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# TRITIUM: Design, Construction and Commissioning of an In-Water Tritium Detector

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*Dedicated to  
my family*

Sometimes it is the people no one imagines anything  
of who do the things that no one can imagine.

"Alan Turing"

I



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# Abstract

Tritium is one of the most abundantly emitted radioisotopes in a nuclear power plant. Large quantities of tritium are normally produced in the water of their cooling system, which are finally emitted to the environment. Due to the fact that high quantities of tritium could be dangerous for human health and for the environment, there exist several regulations around the world which try to control this radioactive emissions in each country, like the Directive Europeen 2013/51/Euratom, which establishes the tritium limit in drinking water in Europe to 100 Bq/L, or the U. S. Environmental Protection Agency, in United States, whose tritium limit in drinking water is established at 740 Bq/L.

Nowadays, due to such a low energy emitted in the tritium decay, high sensitive detectors are needed for measuring it like LSC. The problem with LSC is that it is an off-line method the measurement process of which can take up to 3 or 4 days, too much time if there are any problem with the NPP.

Detectors based on solid scintillators is a promissing idea for building a tritium detector that works in quasi-real time. This type of detectors has been developed so far succesfully but without achieving enough sensibility for measuring the legal limits.

In this study the results of TRITIUM project is presented. In the

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framework of this project a quasi-real time monitor for low tritium activities in water have been developed . This monitor is based on a tritium detector that contains several detection cells which are read in parallel, several active vetos and a pasive shielding for reducing the natural background of our system and an ultrapure water system to prepare the sample before being measured. Each detection cell is made up of hundreds of scintillating fibers read out by PMTs or SiPM arrays.

The final objective of this monitor will be the radiological protection around the nuclear power plant. This monitor will provide an alarm in case of an unexpected tritium release. It will be included in the early alarm system of Extremadura consisting of several detectors the objective of which is to reduce the impact of Nuclear Power Plants to the environment.

**Keywords:** Tritiated water, Real-time monitor, Nuclear Power Plant, Environmental Safety, ...

# Nomenclature and Acronyms

Acronyms:

|                   |  |
|-------------------|--|
| <i>ICRU</i>       | — International Commission of Radioactivity Units and Measurements       |
| <i>ICRP</i>       | — International Commission on Radiological Protection                    |
| <i>ISR</i>        | — International Society of Radiology                                     |
| <i>UNSCEAR</i>    | — United Nations Scientific Committee on the Effects of Atomic Radiation |
| <i>IAEA</i>       | — International Atomic Energy Agency                                     |
| <i>UN</i>         | — United Nations   |
| <i>EU</i>         | — European Union   |
| <i>EURATOM</i>    | — European Atomic Energy Community                                       |
| <i>CSN</i>        | — Nuclear Safety Council   |
| <i>REA</i>        | — Network of automatic stations  |
| <i>REM</i>        | — Network of sampling stations   |
| <i>quasi-real</i> | — Less than 10 minutes   |
| <i>LSC</i>        | — Liquid Scintillation Counting  |
| <i>PMT</i>        | — PhotoMultiplier Tub  |
| <i>SiPM</i>       | — Silicon PhotoMultiplier  |
| <i>NPP</i>        | — Nuclear Power Plants   |
| <i>U.S.DOE</i>    | — United States Department of Energy                                     |
| <i>U.S.</i>       | — United States  |
| <i>PWR</i>        | — Pressurized Water Reactor  |

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|                  |   |
|------------------|---|
| <i>BWR</i>       | — Boiled Water Reactor                                    |
| <i>HWR</i>       | — Heavy Water Reactor                                     |
| <i>GCR</i>       | — Gas-Cooled Reactor                                      |
| <i>USA</i>       | — United States of America                                |
| <i>LARAM</i>     | — Laboratorio de Radiactividad Ambiental                  |
| <i>WHO</i>       | — World Health Organization                               |
| <i>ALARA</i>     | — As Low As Reasonably Achievable                         |
| <i>GL</i>        | — Guideline   |
| <i>EPA</i>       | — Environmental Protection Agency                         |
| <i>LDL</i>       | — Lower Detection Limit                                   |
| <i>IC</i>        | — Ionization Chamber                                      |
| <i>BIXS</i>      | — Beta Induced X-ray Spectrometry                         |
| <i>SDD</i>       | — Silicon Drift Detector                                  |
| <i>APD</i>       | — Avalanche Photodiode                                    |
| <i>EEC</i>       | — European Economic Community                             |
| <i>CNRS</i>      | — Le Centre National de la Recherche Scientifique, France |
| <i>PMMA</i>      | — Polymethyl Methacrylates                                |
| <i>CCD</i>       | — Charge-Coupled Device                                   |
| <i>HV</i>        | — High Voltage  |
| <i>QE</i>        | — Quantum Efficiency                                      |
| <i>CE</i>        | — Collection Efficiency                                   |
| <i>MPPC</i>      | — Multi-Pixel Photon Counter                              |
| <i>G – APD</i>   | — Geiger Avalanche Photodiode                             |
| <i>SSPM</i>      | — Solid State PhotoMultiplier                             |
| <i>MRS – ADP</i> | — Metal-Resistor-Semiconductor Avalanche Photodiode       |
| <i>MAPD</i>      | — Micro-Pixel Avalanche Photodiode                        |
| $\lambda$        | — Wavelength  |
| $\lambda_p$      | — Maximum wavelength of the associated spectrum           |
| <i>PDE</i>       | — Photodetection Efficiency of the SiPM                   |
| $C_t$            | — Terminal Capacitance of the SiPM                        |
| $G_{SiPM}$       | — Gain of the SiPM  |

|                             |   |
|-----------------------------|---|
| $V_{BR}$                    | — Breakdown Voltage of the SiPM               |
| $\Delta TV_{op}$            | — Temperature Coefficient (mV/ $^{\circ}C$ )  |
| $q_e$                       | — Electron Charge                             |
| $\sigma_T$                  | — Total Uncertainty of the Measurement        |
| $\sigma_{st}$               | — Stadistical Component of the Uncertainty    |
| $\sigma_{si}$               | — Sistematically Component of the Uncertainty |
| $A_m$                       | — Activity Measured                           |
| $10 \mu\text{Sv}/\text{cm}$ | — MicroSievert per Centimeter                 |
| $HPGe$                      | — High Purity Germanium Detector              |
| $ROI$                       | — Region of interest                          |
| No-clad fibers              | — Uncladded fibers                            |
| $POF$                       | — Region of interest                          |
| $PHWR$                      | — Pressurized Heavy Water Reactor             |
| $UDL$                       | — Upper Detection Limit                       |
| $LWR$                       | — Liquid Water Reactor                        |

### Atomic and nuclear symbols

|                        |   |
|------------------------|---|
| $^1_1\text{H}$         | — Hydrogen  |
| $^2_1\text{H}$         | — Deuterium (Non-Radiactive Hydrogen Isotope)       |
| $^2_1\text{D}$         | — Deuterium (Non-Radiactive Hydrogen Isotope)       |
| $^3_1\text{H}$         | — Tritium (Radiactive hydrogen Isotope)             |
| $^3_1\text{T}$         | — Tritium (Radiactive hydrogen Isotope)             |
| $^{14}_6\text{C}$      | — Carbon  |
| $^{40}_{19}\text{K}$   | — Potassium   |
| $^{226}_{86}\text{Ra}$ | — Radon   |
| $^3_2\text{He}$        | — Isotope of the Helium(Non-radiactive, 1 neutrons) |
| $^{14}_7\text{N}$      | — Nitrogen  |
| $^{12}_6\text{C}$      | — Carbon  |
| $^6_3\text{Li}$        | — Lithium Isotope                                   |
| $^7_3\text{Li}$        | — Lithium   |

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|                        |   |
|------------------------|---|
| $^{10}_5\text{B}$      | — Boron   |
| $^{16}_8\text{O}$      | — Oxygen  |
| $^{222}_{86}\text{Rn}$ | — Radon   |
| $^{40}_{19}\text{K}$   | — Potassium   |
| $^{137}_{55}\text{Cs}$ | — Cesium  |
| n                      | — Free Neutron  |
| $\text{H}_2\text{O}$   | — Usual Water   |
| $\text{D}_2\text{O}$   | — Heavy Water   |
| HT                     | — Air tritium molecule  |
| HTO                    | — Water tritium molecule                                      |
| OBT                    | — Organic tritium molecule                                    |
| $T_{1/2}$              | — Half-life Time of a Radioactive Element                     |
| $\beta$                | — Beta Decay  |
| $\bar{\nu}_e$          | — Electron Antineutrino                                       |
| $e^-$                  | — Electron  |
| $\gamma$               | — Gamma   |
| $\sigma$               | — Cross Section of a radioactive process                      |
| $\eta_{det}$           | — Intrinsic Detector Efficiency                               |
| $F_{sci}$              | — Active surface of the Plastic Scintillator                  |
| $\varepsilon_{det}$    | — Specific Detector Efficiency                                |
| mip                    | — Minimum Ionizing Particle                                   |
| $Q_\beta$              | — Energy released in a radioactive decay                      |
| S                      | — Specific Energy Lost  |
| Z                      | — Atomic number   |
| $E_\gamma = h\nu$      | — Energy of a photon  |
| $E_e$                  | — Energy of a electron  |
| $m_0$                  | — Rest mass of a electron                                     |
| c                      | — Speed of the light in the vacumm                            |
| $E_b$                  | — Binding Energy  |
| $S_{ij}$               | — Single states of energy levels of electrons in scintillator |
| $T_{ij}$               | — Triple states of energy levels of electrons in scintillator |

Units:

|                            |   |
|----------------------------|---|
| $\text{mSv/yr}$            | — Millisievert per Year   |
| $\text{mrem}$              | — Millirem  |
| $\text{STP}$               | — Standard Temperature ( $0^\circ\text{C} = 273\text{K}$ ) and Pressure (1 atm) |
| $\text{W}$                 | — Watt  |
| $\text{h}$                 | — Hour  |
| $\text{g CO}_2/\text{kWh}$ | — Grams of $\text{CO}_2$ per Kilowatt Hour                                      |
| $\text{L}$                 | — Liter   |
| $\text{Bq}$                | — Becquerel, Nuclear Decay Number per Second                                    |
| $\text{Bq/L}$              | — Becquerel per Liter   |
| $\text{Ci}$                | — Curies  |
| $\text{Ci/L}$              | — Curies por Liter  |
| $\text{yr}$                | — Year  |
| $\text{Ci/yr}$             | — Curies per year   |
| $\text{GW}$                | — GigaWatt  |
| $\mu\text{S/cm}$           | — MicroSivers per Centimeter  |
| $\text{kcps}$              | — Kilo Counts per Second  |
| $\text{pF}$                | — picoFarads  |
| $\text{A}$                 | — Ampere (C/s).   |
| $\text{C}$                 | — Coulomb.  |
| $V$                        | — Voltage.  |
| $T$                        | — Temperature ( $^\circ\text{C}$ ).   |
| $\text{Vol}$               | — Volume ( $\text{m}^3$ ).  |

Añadir en un futuro:

|        |  |
|--------|--|
| $D\&D$ | — Decontamination and Decommissioning. |
| $DWS$  | — Drinking Water Standars              |
| $NA$   | — Numerical Apertures                  |



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# Chapter 1

## Introduction

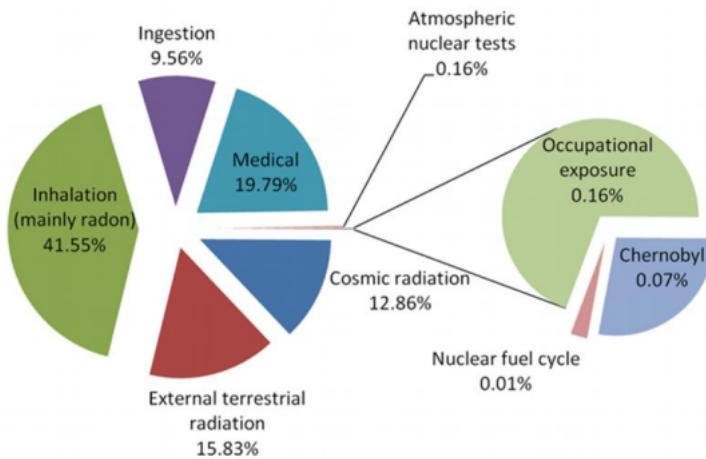
### 1.1 Tritium and Nuclear Energy

Radioactivity has been present in the Universe since its inception. It was an important element of the Big Bang<sup>1</sup>, which occurred about  $14 \cdot 10^9$  years ago. It was also present during the formation of the earth,  $4.5 \cdot 10^9$  years ago, which explains why the different layers that make up the earth contain radioactive elements.

Therefore, humanity has been exposed to radioactivity since its origin, whether present in the Earth's crust or in the universe (external natural irradiation). Even the human being himself is radioactive as radioactive elements are contained in the human body such us  ${}^3\text{H}$ ,  ${}^{14}\text{C}$  or  ${}^{40}\text{K}$ , introduced into the body through food or water ingestion or air inhalation (internal natural irradiation). The annual average of the radioactive dose received by the population is presented in Figure 1.1 and Table 1.1.

---

<sup>1</sup>The Big Bang is the most acceptable hypothesis that explains the formation of the universe and its development over time so far.



**Figura 1.1** – Annual average distribution of the radioactive dose received by the population [IAE21]

As can be seen in Figure 1.1, most of the radioactive dose received by the population is due to both, internal and external natural radiatioactivity, called natural radiation, the effective dose<sup>2</sup> of which is estimated in 2.42 mSv/yr as can be seen in Table 1.1. It can also be appreciated in Figure 1.1 that the most important part of the artificial radiation received comes from medical treatments.

Since the discovery of radioactivity, made by Hènri Becquerel in 1896, a lot of technology based on nuclear concepts has been developed and applied to several fields such as energy production, research, medicine, industry, etc.

Due to the introduction of radioactivity in the society, various anthropogenic radioactive sources have appeared in the environment, resulting in increased levels of its radioactive elements, called radioactive background.

As our knowledge about radioactivity and our measurement tech-

---

<sup>2</sup>The effective dose is the radioactive dose absorbed by the population, taking into account the different radiosensitivity in each organ or tissue.

| Radiation source       | Eff. dose (mSv/yr) | Typical range (mSv/yr) |
|------------------------|--------------------|------------------------|
| Cosmic (external)      | 0.39               | 0.3 – 1.0              |
| terrestrial (external) | 0.48               | 0.3 – 0.6              |
| Inhalation (internal)  | 1.26               | 0.2 – 10               |
| Ingestion(internal)    | 0.29               | 0.2 – 0.8              |
| Total                  | 2.42               | 1 – 12.4               |

Table 1.1: Annual average distribution of the effective dose received by the population due to natural radioactive [UNS21, CSN21a].

niques advanced, the negative effects of radioactivity was observed and characterized. Because of that, it is important to control the level of radioactive background to which the population is exposed and to ensure that these levels is kept below of a safe limit. For this task, several organizations were created to forward recommendations in radiological protection to the different organizations and governments of the world.

1. Firstly, a definition of concepts and units was necessary to quantify the negative effects of radioactivity and, for that, the International Commission of Radiological Units and Measurements, ICRU [ICR21b], was created during the first international conference of radiology held in London, in 1925.
2. Secondly, the International Commission on Radiological Protection, ICRP [ICR21a], was created in 1928 by the International Society of Radiology, ISR [ISR21]. The ICRP aims to make recommendations and to provide guidance on different aspects of protection against radioactivity. The ICRP does not have the legal capacity to enforce its recommendations, but these are widely accepted the legislation of most countries.
3. Thirdly, the United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR [UNS21], was created in 1955, the ob-

jective of which is to estimate and report the levels and effects of ionizing radiation on the population and the environment. These estimates are taken into account by governments around the world to establish their limits and safety standards.

4. Fourthly, the International Atomic Energy Agency, IAEA [IAE21], was created in 1957, which is though to promote the peaceful use of nuclear energy and to avoid its use for any military purpose such us nuclear weapons. Although it is an independent agency, it must to report to the United Nations, UN [UN21].
5. Fifthly, at the level of the European Union, EU, the European Atomic Energy Community, EURATOM, was created in 1957, which is a international organization stablished by the EURATOM treaty. Its objective is to coordinate research programs for the peaceful use of nuclear energy and the sharing of knowledge, infrastructure and financing of nuclear energy.
6. Finally, at the national level in Spain, the Nuclear Safety Council, CSN for its acronym in Spanish, was created in 1980 [CSN21a]. The CSN is the only institution in Spain in matter of nuclear safety and radiological protection and its objective is to reduce as much as possible the radioactivity in the environment due to anthropogenic origins.

For this task, the CSN has created a number of networks consisting of several detectors of radioactivity that are in charge of controlling the levels of radioactivity in the environment and checking the impact of radioactivity facilities to it. Two of the most important networks are the network of automatic stations and network of sampling stations:

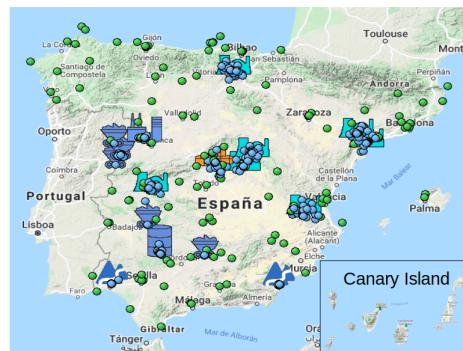
- (a) On the one hand, the network of automatic stations [CSN21b], REA for its acronym in Spanish, shown in Figure 1.2a, which consists of several gamma detectors<sup>3</sup> distributed in Spain that

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<sup>3</sup>Detectors that only measure gamma radioactivity



**(a)** Measured points of the REA [CSN21b]. The white box is the daily average of the gamma dose and the green box is the monthly average of the gamma dose.



**(b)** Measured points of the REM [CSN21c]. Blue dots are located around nuclear facilities. Green dots are uniformly distributed in Spain.

**Figura 1.2** – Networks of automatic and sampling stations managed by the spanish CSN.

measure the radioactive dose in real time. The REA is used for the immediate detection of radiological problems and the application of quick response.

- (b) On the other hand, the network of sampling stations [CSN21c], REM for its acronym in Spanish, shown in Figure 1.2b, which consists of several interesting points in Spain where samples are taken and transported to a laboratory to be measured. About twenty Spanish laboratories are integrated into this network, the objective of which is to characterize the concentration and evolution of various radioisotopes present in the radioactive background of Spain and to quantify the impact of radioactive facilities on the environment.

There are other networks that measure different parameters such as the concentration of  $^{226}\text{Ra}$  in the air and the measurements of all the networks are adapted to the EUROTAM treaty [Dir21].

The goal of this thesis and the *TRITIUM* project is to develop a monitor capable of automatically measuring low levels of tritium in water in quasi-real time<sup>4</sup>. This monitor is destined to be finally included in the REA.

Tritium is one of the radioactive isotopes routinely measured in REM tests and it is detected through the low-energy electrons produced in tritium beta decay, mainly through the liquid scintillation counter technique, LSC. Due to the limitations of the current methods, which will be shown in section 2.1, the objective of the *TRITIUM* project is to build a tritium detector based on scintillating fibers that will be put directly in contact with the sample (water). The photons produced in these scintillating fibers will be read out using photosensors, either photomultiplier tubes (PMTs) or silicon photomultipliers (SiPMs).

The *TRITIUM* collaboration is an international group consisting of a consortium of 6 different European institutions of 3 different countries: Portugal, France and Spain. The final emplacement of the *TRITIUM* monitor is the Arrocampo dam, Extremadura, Spain, the water of which is used for the cooling system of the Almaraz fission nuclear power plant, NPP. This detector will be installed 4 km downstream from the Almaraz Nuclear Power Plant.

The monitor will be used to ensure that the tritium levels of the Arrocampo dam water are below of the legal limit specified in the EURATOM treaty [Dir21], which is 100 Bq/L. It will be used indirectly to verify the correct operation of the Almaraz NPP, located 4 km above the river since a malfunctioning of it will produce an increase of the tritium activity.

Tritium is one of the most abundantly produced radioisotope in a NPP, as it was verified in the United States Department of Energy complex,

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<sup>4</sup>Quasi-real time is an approximation of real-time measurements. It means a relatively small time, like ten minutes.

(U.S. DOE) [Berb, Bera] and in several research facilities in China [Hou18] and places around them (ground water, surface water and process waste water).

Tritium is usually produced in the water used in the nuclear reactor cooling system of some NPPs. It is produced by neutron capture of deuterium, existing in the heavy water ( $D_2O$ ), semi-heavy water ( $HDO$ ) or the deuterium created by neutron capture in usual water ( $H_2O$ ). All these processes have a large probability to happen due to the huge neutron flux in the nuclear reactor, of the order of  $10^{14} \text{ n cm}^{-2}\text{s}^{-1}$  [?]. This tritium is finally released partially or totally to the environment with a quantity that depends on the reactor type as it is shown in Table 1.2. The most common way that tritium is released to the environment is HTO [Hou18].

| Reactor type | Gaseous discharge (GBq/y) | Liquid discharge (GBq/y) |
|--------------|---------------------------|--------------------------|
| PWR          | $3.70 \cdot 10^3$         | $2.59 \cdot 10^4$        |
| BWR          | $1.85 \cdot 10^3$         | $3.70 \cdot 10^3$        |
| HWR          | $7.40 \cdot 10^5$         | $1.85 \cdot 10^5$        |
| GCR          | $7.40 \cdot 10^3$         | $1.11 \cdot 10^4$        |

Table 1.2: Emission of tritium per year from different types of nuclear reactors. Pressurized Water Reactor (PWR), Boiled Water Reactor (BWR), Heavy Water Reactor (HWR) and Gas-Cooled Reactor (GCR) [Hou18]

NPPs are operational since more than 60 years and, nowadays, they are essential for providing a large part of the electric power used in the world (more than 20% in Spain [dE19] and more than a 10% in the world [AIIdlE14]). Although the Spanish government is planning to progressively shut down all NPP there are other countries like China [Pre16] or United States, USA [New19], that promote their use.

On the one hand, NPPs are an interesting investment since it is one of the cheapest source of energy production. It is stable, as it doesn't

depend on meteorological parameters and it doesn't emit greenhouse gases. Although there are other alternative energy sources which are being developed quickly (photovoltaic, wind, tidal energy, etc.), even other concepts of energy production and saving (local production, solar roofs, energy efficiency, smart cities, etc.), today they are not developed enough to fully cover the population needs. On the other hand, NPPs still have some problems such as the contamination of fresh water from uranium mining, the nuclear waste produced, the nuclear proliferation or the risk of radioactive contamination from accidents as happened in the past: Chernobyl, Fukushima and Three Mile Island [Ass20].

In any case nuclear energy production in the world is not going to stop in the next decade, in fact, it will increase as the United States Energy Information Administration (U.S. EIA) expects [Cap20]. Therefore the development of different types of alarm systems is a good investment. Safety is not a negotiable aspect and there must be mechanisms that warn us of any malfunction of a nuclear power plant.

In addition, it is important to highlight that the developed monitor could be used to verify the correct operation of a nuclear power plant, but it is not our objective. Our much broader objective is to ensure that the levels of tritium in the analyzed water are below the Spanish legal limit. It means that this monitor could be used in many different places with radioactive facilities like the future fusion power plants<sup>5</sup>, nuclear research facilities<sup>6</sup> or tracking the pathway of tritium discharges to ground water [AS00].

---

<sup>5</sup>The International Thermonuclear Experimental Reactor, ITER, will need up to several tens of kilograms of tritium to function, which correspond to various TBq of tritium.

<sup>6</sup>Tritium is one of the main emissions from these sites [Fer19], [(BN)].

## 1.2 Tritium Properties and Radiological Hazards

Tritium is the only radioactive isotope of hydrogen present in the environment. It was for the first time produced in 1934 from neutron capture of deuterium by Ernest Rutherford, Mark Oliphant and Paul Harteck [MLO34] and it was first time isolated in 1939 by Luis Walter Alvarez and Robert Cornog [Alv39], who checked that tritium is a radioactive element.

Tritium is naturally produced in the environment through the interaction of cosmic rays and gaseous elements of the upper atmosphere like nitrogen ( $^{14}\text{N}(\text{n}, ^3\text{H})^{12}\text{C}$ ) [USDoEW16] and oxygen ( $^{16}\text{O}(\text{n}, ^3\text{H})^{14}\text{N}$ ) [Hai14]. Around 99% of cosmogenic tritium forms water (HTO) and reaches the Earth's surface as rain with an estimated production rate of  $4 \cdot 10^6$  Ci/yr ( $1.48 \cdot 10^8$  GBq/yr), producing a tritium concentration of 0.6 – 1.2 Bq/L in precipitation [Hou18, USDoEW16].

Tritium can be produced artificially in the environment from different anthropogenic sources [Hou18, USDoEW16]. There is a large amount of tritium which was produced in military nuclear test explosions between 1945 and 1975, with an estimated total production of  $8 \cdot 10^9$  Ci ( $2.96 \cdot 10^{11}$  GBq) and a part of which remains to the date. In these nuclear explosions, tritium was produced mainly from the nuclear reactions  $^{14}\text{N}(\text{n}, ^3\text{H})^{12}\text{C}$  and  $^2\text{H}(\text{n}, \gamma)^3\text{H}$ . Tritium can also be produced by commercial producers of radioluminescent and neutron generator devices ( $1 \cdot 10^6$  Ci/yr), nuclear power and defense industries (around  $2 \cdot 10^6$  Ci/yr) and several research facilities and nuclear reactor for energy production ( $2 \cdot 10^6$  Ci/GWyr), through the production cross sections shown in Table 1.3:

Tritium levels in the environment excluding anthropogenic radioactive sources are between 1 and 4 Bq/L, larger than the expected due to the cosmogenic background levels (0.6 – 1.2 Bq/L, previously mentioned)

| Source            | Origin                     | Nuclear reaction                                 | Cross section (b)   |
|-------------------|----------------------------|--|---------------------|
| $^2_1\text{H}$    | Water coolant              | $^2_1\text{H}(\text{n}, \gamma)^3_1\text{H}$     | $5.2 \cdot 10^{-4}$ |
| $^3_2\text{He}$   | Helium coolant             | $^3_2\text{He}(\text{n}, \text{p})^3_1\text{H}$  | 5330                |
| $^6_3\text{Li}$   | Moderator                  | $^6_3\text{Li}(\text{n}, \alpha)^3_1\text{H}$    | 940                 |
| $^{10}_5\text{B}$ | Moderator,<br>control rods | $^{10}_5\text{B}(\text{n}, 2\alpha)^3_1\text{H}$ | 3835                |

Table 1.3: Most common nuclear reactions of artificial tritium production [Hou18]

[Cal10]. It can be explained by the consequences of nuclear weapons tests.

Tritium levels in rivers around a nuclear facility are between 1 and 10 Bq/L and even between 20 and 50 Bq/L at the water discharge site of NPPs [Cal10], where the produced tritium is partially or totally released into the environment, mainly in the HTO water form.

The effect of NPP on tritium levels can be observed from REM date, for example the case of Cofrentes. Cofrentes is the closest nuclear power plant to Valencia, in the measurements of which are involved the LARAM<sup>7</sup>. There, the tritium level is measured in three different places along the Jucar river, marked on the map shown in Figure 1.3. The first place, P1, is located in the river, 6 km upstream from the NPP, the second place, P2, is located 1 km downstream and the third place, P3, is located 5 km downstream. The level of tritium measured in these three locations is shown as a function of the time in Figures 1.4a, 1.4b and 1.4c respectively.

In these figures, the detection limit and the measured activity are shown using white and green dots, respectively. The measured activity is only displayed when it is larger than the corresponding detection limit. The tritium level in the river increases due to the discharge of the NPP and it is diluted again after 4 km downstream, as can be seen from these date.

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<sup>7</sup>The LARAM is a Valencia laboratory specialized in environmental radioactivity measurements



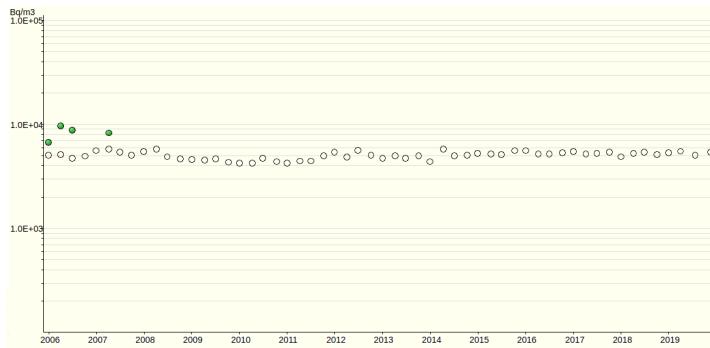
**Figura 1.3** – Tritium sampling locations around Cofrentes NPP.

Two additional measurements of the tritium level in groundwater have been included, points S1 and S2 on the map in Figure 1.3, which are located 1 km before and 1 km after the NPP. Both tritium levels are shown in the figure 1.5a and 1.5b respectively, where it can be observed that they are not affected by the nuclear power plant.

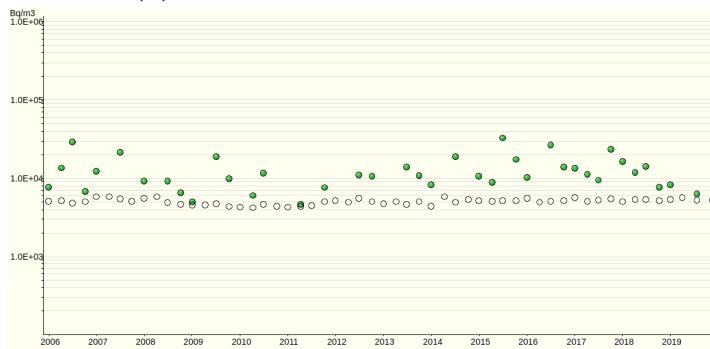
It is important to note that, although environmental tritium level is affected by NPP, these levels are below the maximum allowed limit. The maximum level of tritium measured since January 2, 2006 is around 32 Bq/L, below to the maximum allowed limit in Europe, 100 Bq/L.

Tritium is a radioactive element with a half-life time of  $T_{1/2} = 12.32$  years. It has one proton and two neutrons and decays exclusively through  $\beta$  radiation. It decay 100% directly to the ground state of the  $^3_2\text{He}$  isotope of helium, which is a stable nuclei, through the decay scheme of equation 1.1:

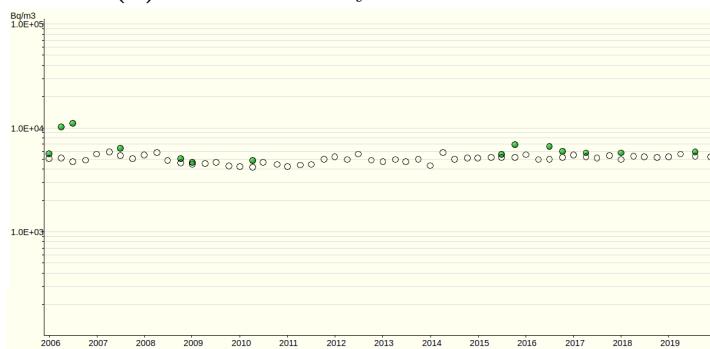




(a) Tritium activity 6 km upstream.

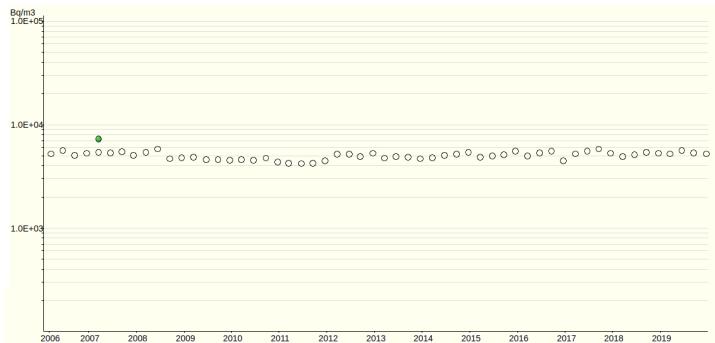


(b) Tritium activity 1 km downstream.



(c) Tritium activity 5 km downstream.

**Figura 1.4** – Tritium activity levels in surface water around Cofrentes NPP from January 2006 to November 2019. The white points are used for the detection limit and the green points are used for the measured activity, when it is above the detection limit. [CSN21c]

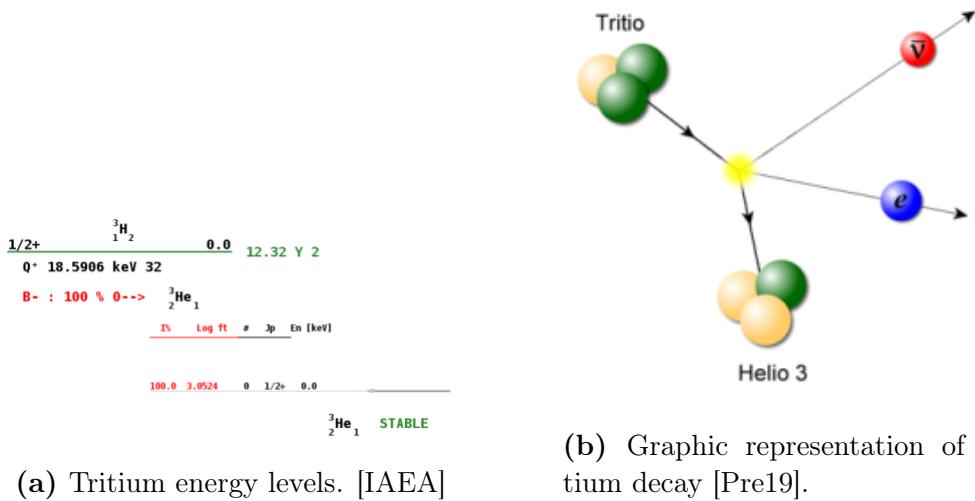


(a) Tritium activity 1 km before NPP.



(b) Tritium activity 1 km after NPP.

**Figura 1.5** – Tritium activity levels in groundwater around Cofrentes NPP from January 2006 to November 2019. [CSN21c]



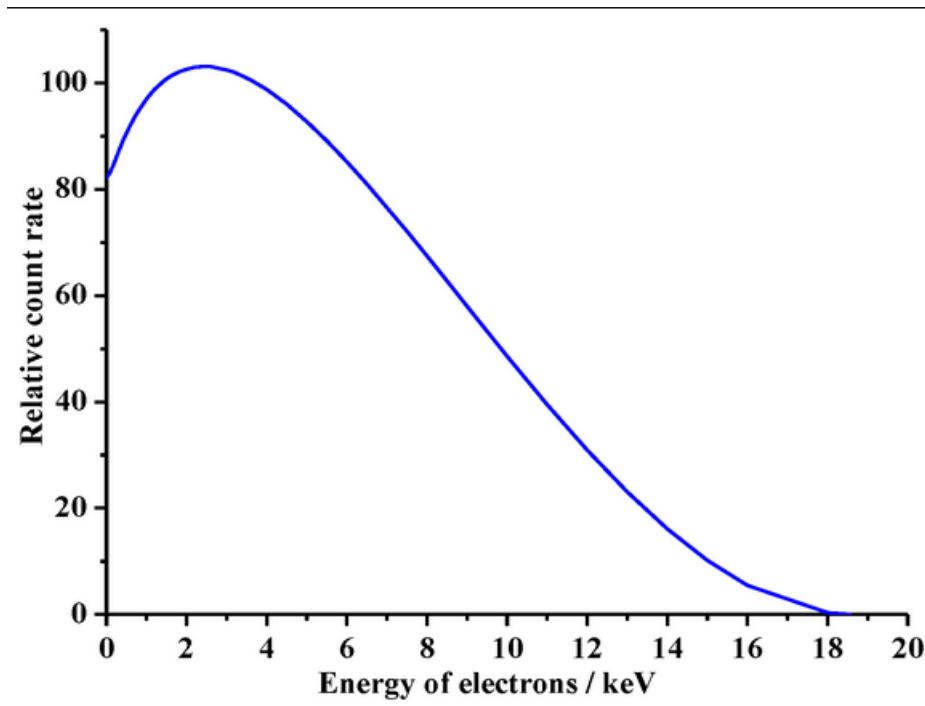
**Figura 1.6 – Tritium decay**

In Figure 1.6 the scheme of tritium energy levels is shown. In this decay it is not possible to detect the neutrino because of its extremely weak interaction with matter ( $\sigma \propto 10^{-42} \text{ cm}^2$  [?]) and, since  $^3\text{He}$  has a much larger mass than electrons and neutrinos, by conservation of energy and momentum, the energy that is taken by this daughter nucleus is very small. Therefore, the detection of tritium is through its decay electron.

The energy released in the tritium decay is  $Q_\beta = 18.6 \text{ keV}$ , shared between the decay products. Therefore, the energy spectrum of the decay electrons is a continuum with a maximum value of 18.6 keV, as shown in Figure 1.7. This energy spectrum has an average energy of 5.7 keV and the most likely energy is slightly below, around 4.5 keV.

The releasing energy of the tritium decay, is very low. In fact, it is the radioactive isotope with the lowest energy released in its  $\beta$  disintegration [USDoEW16]. Consequently, the  $\beta$  particle which is emitted in this tritium decay will have a very small mean free path, shown in Table 1.4.

This short mean free path is a major issue in tritium detection, as



**Figura 1.7** – Energy spectrum of tritium electrons [Lin20]

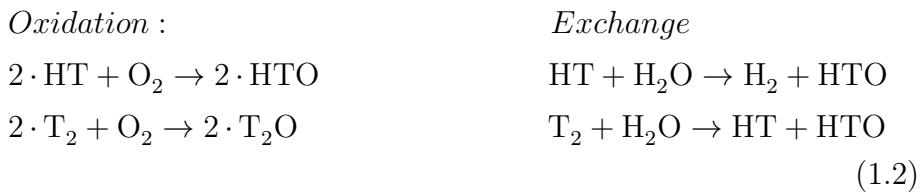
| Material  | P. Depth (5.7 keV) | P. Depth (18.6 keV) |
|---|--------------------|---------------------|
| ${}^3_1\text{H}_2$  | 0.26 cm            | 3.2 cm              |
| Air   | 0.036 cm           | 0.45 cm             |
| Water, soft tissue<br>(solid matter with a<br>density of $1 \text{ g} \cdot \text{cm}^{-3}$ ) | $0.42 \mu\text{m}$ | $5.2 \mu\text{m}$   |

Table 1.4: Penetration depth for decay electron of mean (5, 7 keV) and maximum (18, 6 keV) energies in different media (tritium gas and air at standard conditions of temperature (273 K) and pressure (1 atm), STP, and water) [Bla]

it makes more difficult the electron detection, which will require a highly sensitive detector. It also means that tritium electrons have a low penetration in our body and they are easily stopped with clothes or laboratory gloves, resulting in a low radiological hazard of external tritium. Nevertheless, the danger of tritium increases when it is ingested or inhaled since it can bind anywhere that hydrogen can and perform the same chemical reactions, sometimes with higher rate if the tritium concentration is high enough to catalyze the reaction.

Tritium can be absorbed in our body in three different ways, gaseous tritium (mainly HT), tritiated water (mainly HTO) and organically bound tritium (called OBT).

- The gaseous tritium, which is normally found mixed in the air, is the least important since less than a  $3 - 5 \cdot 10^{-3} \%$  is absorbed by the human body, which is insignificant [USDoEW16]. However, it can be transformed into tritiated water, more harmful from a radiobiological point of interest [USDoEW16], through the oxidation and exchange reactions by equations 1.2:



- Tritiated water, which is normally found in drinking water and food, has a larger impact since the 99% of it is absorbed [USDoEW16]. Its biological life time corresponds to the water cycle in the body, around 9.5 days ( $\pm 50\%$ ), time during which tritium will remain in our body [USDoEW16, Cal10, Mas]. As in water, the biological life time of tritiated water can vary due to various external parameters such as

temperature, humidity, drinking habits, etc. or reduced with the use of diuretics [USDoEW16].

- Organically bound tritium, normally found in food, generally forms a covalent bond with a carbon and it corresponds to 5 – 10 % of tritium absorbed in the body. Although it is less absorbed in the body than tritiated water, it can be more dangerous since it has a longer biological life time. The biological life time of this tritium type depends on the affinity of the organic molecule to the different biological tissues and it can vary from tens to hundreds of days (larger than the ICRP estimate) [Cal10, Mas, PF81, Mar72].

There are many studies showing that tritium in living matter can cause the same effects than X-rays or  $\gamma$  rays, which are mutations, tumors, cancer, genetic effects, reproductive effects, etc [Str93, Ryt]. In fact, the consequences of tritium radiation can be worse than a similar  $\gamma$  radiations since its biological efficiency<sup>8</sup> is two or three times larger [Str93].

In summary, tritium is a naturally occurring radioactive element that can affect health if it is released excessively. Because of that, each country has developed a legislation, shown in section 1.3, to manage the release of tritium and ensure that these background levels are safe for health.

## 1.3 Current Legislation

Due to the radiological risk of tritium, which was deal with in section 1.2, it is important that the current legislation limits the release of tritium to the environment ensuring that the levels are below a safe value for health.

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<sup>8</sup>The biological efficiency is used to quantify the damage produced in the living cells due to an external radiation.

The guidelines setting impose the limit of radioactive elements in drinking water for many countries are based on the radiation protection methodology developed by the ICRP [ICoRP91] and the recommendations of the world health organization (WHO) [WHO04].

The objective of the international radiation methodology is to protect people and the environment from the negative effects of ionizing radiations but allowing beneficial activities that involve a reasonable radiation exposure. It is based on three main points, which are the justification (the benefit from radiological exposure must outweigh the detriment to health that it causes), the ALARA principle, "As Low As Reasonably Achievable" (the radiological exposure must be kept as low as possible considering social and economic factors) and dose limitation (limit that must never be exceeded).

While the ICRP recommends a maximum dose of 1 mSv/yr, excluding the natural background and medical interventions, the WHO is more conservative, recommending a maximum dose of 0.1 mSv/yr, which correspond to less than 5% of the annual dose due to background radiation, 2.42 mSv/year, Table 1.1.

The guideline reference level of each radionuclide in drinking water, GL, is usually calculated from these recommendations using the equation

$$GL = \frac{RDL}{DCF \cdot q} \quad (1.3)$$

where RDL is the reference dose level, DCF is the dose conversion factor (the normal used value for tritium is  $1.8 \cdot 10^{-11}$  SvBq, provided by ICRP [ICoRP96]) and q is an estimation of the annual volume of drinking water consumed (normally assumed two liters per day, 730 L/yr).

The GLD calculated for tritium in drinking water according to the ICRP and WHO recommendations is 76,103 Bq/L and 7,610 Bq/L

respectively. It means that tritiated water with activities below these values is considered not harmful to health.

Based on these recommendations, each country has created organizations in charge of developing its own legislation on radionuclide limits. In Spain, the responsible organization of this task is the CSN.

Most of the countries in the world implement the RDL of 0.1 mSv/yr recommended by the WHO. The legal limit for tritium in drinking water in this case is 7,610 Bq/L but it is often approximated in different ways. Some countries like Switzerland [Dfdl06] or some organizations like the WHO [WHO04] rounded this value to 10,000 Bq/L. Others like some countries of Canada, such as Ontario and Québec, truncate this value to the first number 7,000 Bq/L [otE94, Que16]. There are other countries like Russia which use the much more accurate approximation value of 7,700 Bq/L [ICfS07].

There are other countries like Australia that prefer to implement the RDL of 1 mSv/yr, recommended by the ICRP, the legal limit of which is 76,103 Bq/L [AG21]. Other countries like Finland are based in the ICRP recommendations and use only the middle of this value, 0.5 mSv/yr, the value of which is rounded to a legal limit of 30,000 Bq/L for tritium in drinking water [NEA93].

There are two different exceptions to these recommendations:

1. On the one hand, most of the USA states like California use a RDL of 4 mrem (0.04 mSv), which corresponds to a legal limit of 20 nCi/L (740 Bq/L) [OoEHHA07]. This value was proposed by the United States Environmental Protection Agency (US EPA) as a result of an analysis of the available information [USEPA76].
2. On the other hand, most of the EU countries, such as France, Germany or Spain, impose an GL of 100 Bq/L, which is one of the most restrictive limit in the world [Idredsn17, BfS05, CdSN13]. This value

arise from the consideration that it is an indicator of the presence of other radionuclides more dangerous than tritium. These limits are fixed by the EURATOM Council Directive [EAEC13].

All limits mentioned in this section are summarized in table 1.5.

| Country/Agency | Legal limit of tritium (Bq/L) |
|----------------|-------------------------------|
| ICRP           | 76, 103                       |
| WHO            | 10,000                        |
| Switzerland    | 10,000                        |
| Canada         | 7,000                         |
| Russia         | 7,700                         |
| Australia      | 76, 103                       |
| Finland        | 30,000                        |
| United States  | 740                           |
| European Union | 100                           |

Table 1.5: Legal limit of tritium in drinking water established in each country.

## 1.4 This Thesis

This thesis is divided into nine different chapters that structure the information as follows:

**Chapter 1** provides a brief introduction of the importance of tritium detection, shows some important properties of tritium, and discusses the current legislation that limits tritium levels in many countries around the world. **Chapter 2** exhibit the State-of-the-Art in tritium detection and shortly introduces the TRITIUM project. **Chapter 3** details the different parts on which the TRITIUM monitor are based, which are the ultrapure water system, the background rejection system (consisting of the lead shielding and the active veto) and the tritium detector. **Chapter 4** presents the

calibrations of the different parts of the TRITIUM monitor and shows the developments aimed at improving the efficiency of tritium detection. **Chapter 5** details the geometric configuration of the different prototypes built in the TRITIUM project, the measurements of which are presented **Chapter 7**. **Chapter 6** explains the Monte Carlo simulations performed, the result of which are shown in **Chapter 8**. Finally, **Chapter 9** summarized the most important results achieved in this work.



# Chapter 2

## Tritium Detection Systems

### 2.1 State-of-the-Art

Measurement of tritium activity is one of the routine environmental controls that are carried out in the vicinity of nuclear research facilities and nuclear power plants during their energy production lifetime. Consequently, this measurement has been carried out with different available technologies under development to improve the state of the art of tritium detection. The most employed techniques are summarized in Table 2.1.

Nowadays, the most used technique for measuring tritium in water is liquid scintillator counting (LSC). This technique consists of mixing a liquid sample (some ml for environmental measurements or less for higher activities) with liquid scintillator. This mixture is usually made in a ratio of 50:50 but it depends on the detection system and the samples used [AH99, Hof92a]. In this technique, the  $\beta$  energy emitted from the sample excites the molecular energy levels of the liquid scintillator which promptly decays emitting several photons with a well-known energy (fluorescence), usually in the visible spectrum. Finally these photons are detected with

|                   | LSC                   | IC                 | Calorimetry | BIXS       |
|-------------------|-----------------------|--------------------|-------------|------------|
| Measured quantity | Scintillation photons | Ionization current | heat        | X-rays     |
| LDL               | $\sim$ Bq             | 10 – 100 kBq       | $\sim$ GBq  | $\sim$ MBq |
| Sample form       | Liquid                | Gas, vapor         | All         | All        |

Table 2.1: State-of-the-art in the tritium detection for different techniques. This table show the measured quantity, low detection level (LDL) and the sample form for four different techniques, liquid scintillator counting (LSC), ionization chamber (IC), calorimetry and beta induced X-rays spectrometry (BIXS)

photosensors, which convert the optical signal into measurable electrical charge. The liquid scintillator technique has a very good detection sensitivity for low activity levels of tritiated water ( $< 1 \text{ Bq/L}$ ) [Pal07] but it has the problems of long measurement time (up to 2 days) and of producing chemical waste, since liquid scintillator contains toluene which is toxic. In addition, this technique requires special staff for sampling, chain-of-custody and lab analysis which require economical and time resources. In order to avoid this problem some unsuccessful efforts have been made in order to build a monitor of tritium with LSC [Sig94].

The ionization chamber (IC) consists of a gas chamber (sample) which contains electrodes connected to different voltages. These electrodes collect the ionization current that is produced due to the  $\beta$  radiation. It is a simple and fast system, but it has the problem of high Low Detection Limit ( $> 10 \text{ kBq}$ ) and of requiring the samples to be in a state of gas or steam [Khe02, ZC13].

The calorimetry method is based on the measurement of the heat generated in the detection medium (normally platinum) [CGA17, BD13]. The problem with this technique is that it has a high LDL, of the order of GBq, and requires long measurement time, 2 days or more.

The Beta Induced X-ray Spectrometry (BIXS) is based on the measurement of the bremsstrahlung radiation produced by the tritium decay electrons, using a NaI(Tl) crystal couplet to a PMT [Mat07, Mat08] or Silicon Drift Detector (SDD) [Nie15]. The problem with this technique is a high LDL, of the order of MBq.

There are additional methods for tritium detection, although they are less employed or less experimentally developed, each one with its own advantages and limitations. For example, the Avalanche PhotoDiode (APD) cannot be used in contact with water [Sha97], the mass spectrometry which needs to store the sample several months before taking the measurement [JB10] and the Cavity ring spectroscopy requires a special optical configuration that is not possible outside a laboratory [Bra15].

All the above techniques are offline methods that need long time for sample collection, shipment to the laboratory and activities measurements. Therefore, they cannot be used for in-situ monitoring of tritium in water. The liquid scintillation technique is the only one with sufficiently small Low-Detection-Limit to fulfill the compliance of 100 Bq/L in tritium of the water samples, established by the EURATOM directive.

The purpose of the TRITIUM project is to develop an alternative method, based on solid scintillators, that allows to accomplish the requirements of in-situ monitoring of levels as low as the legal limit in Europe, 100 Bq/L, in quasi-real time. There are several studies that was developed with solid scintillators so far:

1. The study done by M. Muramatsu, A. Koyano and N. Tokunaga in 1967 who used a scintillator plate read out by two PMTs in coincidence [Mur67].
2. The study carried out by the A. A. Moghissi, H. L. Kelley, C. R. Phillips and J. E. Regnier in 1969 that used one hundred plastic fibers

coated with anthracene powder and read out by two PMTs in coincidence [Mog69].

3. The study performed by R. V. Osborne in 1969 that used sixty stacked scintillator plates read out by two PMTs in coincidences [Osb70].
4. The study done by A. N. Singh, M. Ratnakaran and K. G. Vohra in 1985, that used a scintillator sponge read out by PMTs in electronic coincidence [Sin85, MR00].
5. The study carried out by K. J. Hofstetter and H. T. Wilson in 1991, that tested different shapes of scintillator plastics like several sizes of beads, fibers, etc. The better result obtained for solid plastic scintillator was a tritium detection efficiency of the order of  $10^{-3}$  [Hof92b, Hof].

| Study      | $\varepsilon_{det}(\frac{cps \cdot 10^{-3}}{kBq/L})$ | $F_{sci}$ (cm <sup>2</sup> ) | $\eta_{det}(\frac{cps \cdot 10^{-6}}{kBq/L \cdot cm^2})$ | LDL (kBq/L) |
|------------|--|------------------------------|--|-------------|
| Muramatsu  | 0.39   | 123                          | 3.13   | 370         |
| Moghissi   | 4.50   | > 424.1                      | < 10.6   | 37          |
| Osborne    | 12   | 3000                         | 4  | 37          |
| Singh      | 41   | 3000                         | 13.7   | < 37        |
| Hofstetter | 2.22   | ~ 100                        | < 22.2   | 25          |

Table 2.2: Results of different scintillator detector for tritiated water detection. This table shows the efficiency of the detector ( $\varepsilon_{det}$ ), its active surface ( $F_{sci}$ ), its specific efficiency ( $\eta_{det} = \varepsilon_{det}/F_{sci}$ ), defined as its efficiency normalized to its active surface, and its low detection-level (LDL) for each study listed above.

The results of these experiments are summarized in Table 2.2. As can be seen in the first column, the intrinsic detector efficiency,  $\varepsilon_{det}$ , is very different in these experiments. As one of the most important factor that affect the efficiency is the active surface of the plastic scintillator,  $F_{sci}$ , which varies largely with the detector type, the specific detector efficiency (third column) is used in order to compare these experiments, that is, the intrinsic

detector efficiency normalized to this active surface. It can be checked that, effectively, these specific efficiencies are quite similar. The specific efficiency obtained by Moghissi for scintillating fibers is sufficiently high to justify our choice of scintillating fibers as a detection medium. Finally, as can be seen in the last column, the LDL in all these experiments are of the order of a few tens of kBq/L. Thus, to develop a detector with much lower LDL is essential to comply with the EURATOM directive of 100 Bq/L of tritium in water for human consumption.

## 2.2 TRITIUM Project

As a conclusion of section 2.1, the current techniques cannot be used for tritium monitoring in quasi-real time since they have either a higher LDL or they work in off-line method (too slow).

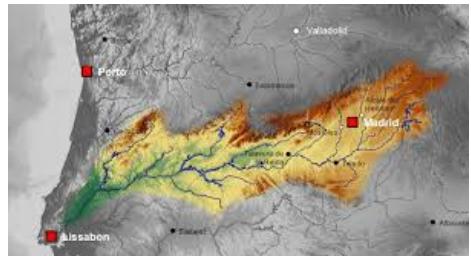
To overcome these limitations the *Tritium* project [Pro21], with the title of "Design, construction and commissioning of automatic stations for quasi-real time monitoring of low radioactive levels of tritium in water", was proposed.

The *Tritium* collaboration is a international group consisting of a consortium of 6 different european institution of 3 different countries: The University of Aveiro, in Portugal, The University of Bordeaux and the National Center for Scientific Research, CNRS (Section Aquitaine-Limousin), in France and the University of Extremadura, *Junta de Extremadura* and University of Valencia, in Spain.

This project was funded by the Interreg Sudoe program of the European Economical Community, EEC, in the 2016 call with the reference number SOE1/P4/EO214. The purpose of this project is the development of a tritium monitor in quasi-real time. This monitor consists of a ultra-



**(a)** Arrocampo dam and Almaraz Nuclear Power Plant.



**(b)** Tajus river along Spain and Portugal.

**Figura 2.1** – Arrocampo dam, Almaraz NPP and Tajus river

pure water system, which prepare the water sample before introducing it in the detector for tritium measurement, a cosmic veto and a passive shielding, which reduce the natural background of our tritium detector, and several electronic modules which control all the different parts of the monitor, analyze the tritium measurement and send an alarm if the configured limit (100 Bq/L) is exceeded.

A crucial problem is to distinguish tritium signals from the background because tritium events has low energy ( $\sim$  keV) and fall in an energy range of the spectrum where there is significant background. To reduce the background counts of TRITIUM monitor, coincidence techniques are used.

The TRITIUM monitor will be installed in the Arrocampo dam, Almaraz, Spain, where the Almaraz NPP release the water of the secondary cooling circuit, displayed in Figure 2.1. This NPP has two nuclear reactors of PWR type. Arrocampo dam is located near the Tagus river, shown in Figure 2.1b, which is the longest river in Spain, with a long of the 1007 km. This river, shown in Figure 2.1a, rises in Aragon (Spain) and flows into the Atlantic Ocean, Lisbon (Portugal). The water of this river is used for agriculture and drinking water by both, Spanish and Portuguese people. Therefore the international cooperation in order to maintain the quality of the tajus river water is very important.

Each institution has concentrated its efforts in the development of a different part of this project:

1. The Extremadura group has developed and installed the ultrapure water system to produce water with very low conductivity,  $\sigma \approx 10 \mu\text{Sv}/\text{cm}$  (two orders less than sample before the cleaning process,  $1000 \mu\text{Sv}/\text{cm}$ ). This cleaning process is very important for two reasons. On the one hand, for maintaining our detector very clean, which is a critical point. On the other hand, to reduce the natural background since several natural radioactive isotopes presents in this water (except tritium) are removed such as  $^{222}\text{Rn}$ ,  $^{40}\text{K}$  or  $^{137}\text{Cs}$ . This system is explained in section 3.3.
2. The french group has develop the pasive shielding for the detector. The shielding is made of ultra radiopure lead with very low intrinsic activity in order to reduce the external natural background of the system. This shielding is presented in section 3.4.1.
3. The Portuguese and Spanish group have collaborated for designing, developing and building four different prototypes of tritium detector and active vetos for removing cosmic events. These prototypes and vetos are explained in chapter 5 and section 3.4.2 respectively.
4. The Portugal and Spanish people have also carried out simulations of this system. The environment chosen to develop these simulations is the package GEANT4 [Col21], consisting of an extensive C++ library with which the geometry of our detector, the relevant physical processes, etc. can be specified. This simulation are explained in chapter 6.

The important characteristics of the TRITIUM detector must have:

- *Compact.* This is an important point because in the place where this detector will be installed the useful space to be used is very limited.

- *Thin active volume and large active area.* On the one hand, it have to be taken into account that the mean free path of the  $\beta$  particle of tritium decay is very low so thin detector active volumes are needed. In practice, active thickness beyond the mean free path of the tritium electrons will only contribute to background. On the other hand, as explained in section 2.1, the efficiency of this type of detector scales with the active area, so it is crucial to design the detector with the largest possible active area.
- *High efficiency to tritium.* As the tritium activities to be measured are very low, it is very important to reduce as much as possible the non-detected tritium events.
- *High specificity to tritium.* The detector has to be able to distinguish the tritium signal from the signal due to other radioactive elements present in the sample.
- *Quasi-real time response.* It is important that the sistem work in quasi-real time in order to detect any problem as fast as possible.
- *Rugged system.* Finally, it has to be take into account that the final goal is to install an automatical system which work during a number of years without requiring the intervention of specialized people. Therefore, a rugged monitor is required.

In order to get the measurement in quasi-real time it is needed to work *in situ*, that is, in the same place that the water sample is taken. Working *in situ* has some benefits for the detector such as faster and cheaper maintenance, since the sampling process, chain of custody, etc. are eliminated, more frequent measurements are carried out and safer monitoring since personal exposure dose is reduced, changes in activity levels can be detected quickly and possible errors due to specialized staff are eliminated.

# Chapter 3

## Design Principles of the Tritium Monitor

### 3.1 Detector System Overview

The objective of the TRITIUM project is the design, development, construction and commissioning of an automatic station for real-time monitoring of low levels of tritium in water. To achieve this aim, the TRITIUM group has developed a monitor consisting of several parts, listed below:

1. The TRITIUM detector, described in chapter 5, is based on several modules read in parallel. Each module consists of hundreds of scintillating fibers, section 3.2.2, which are in contact with the water sample measured, read by two coincident photosensors, section 3.2.3. The photosensors considered are photomultiplier tubes (PMT) (section 3.2.3) and silicon photomultipliers (SiPM) (section 3.2.3).
2. The ultrapure water system (section 3.3) that prepares the water sample before measurement. This system removes all the organic parti-

cles dissolved and all the particles with a diameter greater than  $1 \mu\text{m}$  without affecting the tritium content of the sample. This system is important for two reasons: First, because the mean free path of tritium in water is very short, 5 or  $6 \mu\text{m}$ , so this is essential to avoid the deposition of particles onto the fibers because this would prevent the tritium decay electrons from reaching the fibers. Second, particles dissolved in water may contain radioactive isotopes like  $^{40}\text{K}$ , which would increase the background. As the water sample has very low tritium counters, to reduce the background is a crucial matter.

3. The background rejection system (section 3.4), that has two different parts. The first one is a passive shield (section 3.4.1), consisting of a lead castle inside of which the TRITIUM detector is located. This castle is employed to eliminate natural radioactive background and cosmic rays with energies up to 200 MeV/nucleon. The second part is an active veto (section 3.4.2), consisting of two plastic scintillation blocks located inside of a passive shielding, above and below the TRITIUM detector and read by several photosensors. The goal of this active veto is to remove the remaining high energy events ( $> 200 \text{ MeV}$ ), cosmic rays that can travel through the passive shielding and contribute to background. Contrary to low energy cosmics rays, high energy cosmic rays are difficult to be stopped. The technique employed to eliminate their contribution consists of reading the TRITIUM detector in anti-coincidence with the active veto.
4. A monitoring electronic system sends an alarm if the signal limit of the tritium level, 100 Bq/s, is exceeded.

The different parts of TRITIUM monitor were subjected to tests to verify their correct operation before installing them in the Arrocampo dam. The final goal is to include TRITIUM in the network of automatic stations, REA (section 1.1).

## 3.2 TRITIUM Detector

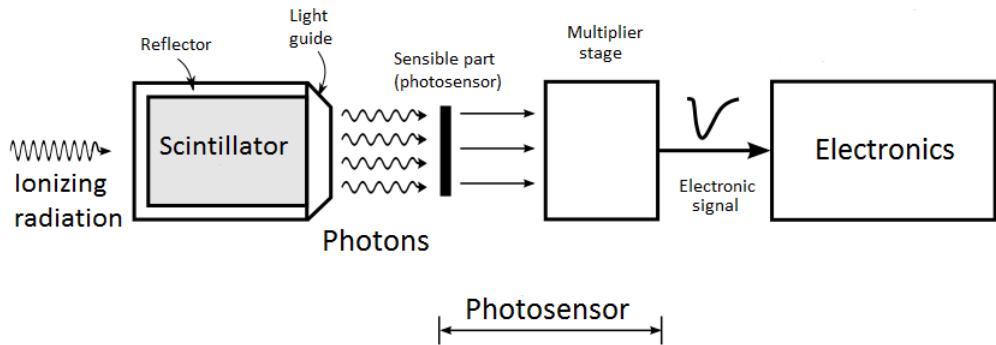
As discussed in section 2.1, the TRITIUM consists in a chain of three main elements:

- The scintillator, that detects the tritium event, as ionizing radiation hits this material and deposits kinetic energy through ionization and excitation processes, part of the absorbed energy is converted in to photons, mostly in the visible range<sup>1</sup>. The produced photons carry information about the particle detected, like its energy, type, etc.
- The photosensor, that detects the photons produced in the scintillator. The most common photosensors in nuclear physics are PMTs and SiPMs. They detect the photons produced in the scintillator with an efficiency and transforms them in electrons with a multiplication factor of around  $10^6$ . These electrons form a electronic pulse than gives information of the detected photons.
- The electronic system, which is the part of the scintillator detector in charge of processesing and analyzing (first analogically and then digitally) the electrical pulse given by the photosensor. The output of the electronic system is the useful information about the events detected such as number and energy spectrum.

In Figure 3.1 a scheme of a scintillation detector is shown. There, the scintillator detects ionizing radiation and produces photons that are guided by the reflector and the light guide to the photosensor. Some of the photons that reach the sensitive part of the photosensors are converted and multiplied, forming an electronic pulse. The output signal of the photosensor (electronic pulse) is processed and analyzed by the corresponding electronics.

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<sup>1</sup>The visible range is made up by photons with a wavelength between 380 nm and 750 nm



**Figura 3.1** – Scheme of the scintillator detector

### 3.2.1 Interaction of Fast Electrons and Photons with Matter

This section describes the interaction of particles with matter, focusing on the particles and energy range relevant for this thesis, electrons (0–18 keV) and photons in the visible range (approx. 380 – 750 nm).

Electrons have charge so their interaction with matter is mainly with the orbital atomic electrons through the Coulomb force. The electron trajectory is much more tortuous than other heavier particles because the mass of both interacting particles is equal. Furthermore, for the same reason, these electrons lose a significant amount of energy in each collision. The specific energy loss is defined as  $S = -\frac{dE}{dx}$  which gives the energy loss suffered by the particle per unit of path length. In the case of electrons, this total energy loss has two main contributions, the collisions (elastic and inelastic) and radiative processes (bremsstrahlung) [Kno99, Leo94]:

$$\frac{dE}{dx} \approx \left( \frac{dE}{dx} \right)_c + \left( \frac{dE}{dx} \right)_{br} \quad (3.1)$$

The radiative part is roughly proportional to the collision part:

$$\frac{\left(\frac{dE}{dx}\right)_{br}}{\left(\frac{dE}{dx}\right)_c} \approx \frac{EZ}{700} \quad (3.2)$$

where  $E$  is the energy of the electron in MeV and  $Z$  is the atomic number of the absorbing material. Due to this energy loss, the electrons can only penetrate a material as far as they go before losing their total kinetic energy. This distance is known as range and, in the case of tritium electrons, its value is quoted in Table 1.4.

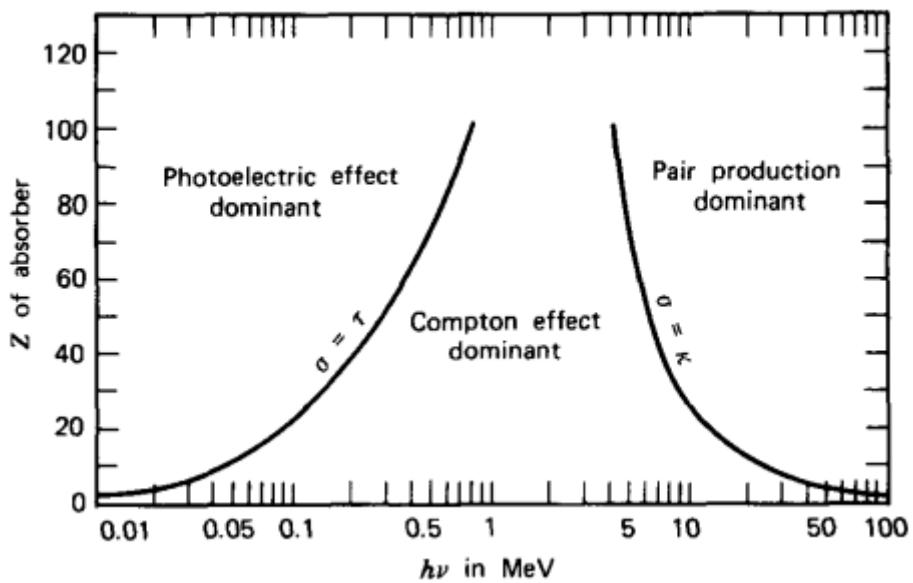
As photons don't have charge, their possible interactions with the matter are photoelectric effect, Compton effect, coherent scattering and pair production and the probability of each process depends on the energy of the photon,  $E_\gamma = h\nu$ , and on the atomic number of the material,  $Z$ , displayed in Figure 3.2.

The only relevant photons for this thesis are in the visible range, between 400 and 700 nm, that corresponds to energies of the order of the eV. Therefore, pair productions, which requires a photon energy equal or more than 1.022 MeV, does not play any role.

The photoelectric effect occurs when a photon interacts with an orbital electron in the material, losing all its energy. This energy is absorbed by the electron that is released from the atom (ionization). The energy of the resulting electron,  $E_e$ , is [Kno99, Leo94]:

$$E_e = E_\gamma - E_b \quad (3.3)$$

where  $E_b$  is the binding energy of the electron in this material. The probability of this effect depends on the number of available electrons in the matter through the variable  $Z$ , and the energy of the electron according to



**Figura 3.2** – Domain regions of the three most probable types of interactions of gamma rays with matter. The lines show the values of  $Z$  and  $\hbar\nu$  where the two neighboring effects are equally likely. [Kno99, Leo94]

the expression [Kno99]:

$$(Pr)_{Ph-eff} \approx \frac{Z^n}{E_\gamma^{3.5}} \quad (3.4)$$

Thus, the photoelectric effect is most probable for elements with high atomic number. This is the reason why elements with high atomic number are the best insulators against gamma radiation and why the passive shielding of TRITIUM monitor consists of lead bricks ( $Z = 82$ ) (section 3.4.1).

The Compton effect occurs when a photon interacts with an orbital electron of the material, transferring part of its energy to the electron, which is released, scattered at an angle  $\theta$  with respect to the original direction. If the electron binding energy is neglected, the energy transferred to it,  $E_e$ , is given by [Kno99, Leo94]:

$$E_e = \frac{\frac{E_\gamma^2}{m_0 c^2} (1 - \cos\theta)}{1 + \frac{E_\gamma^2}{m_0 c^2} (1 - \cos\theta)} \quad (3.5)$$

where  $m_0$  is the rest mass of the electron and  $c$  is the speed of the light in the vacuum. The probability of the Compton effect is proportional to the atomic number (available electrons in the matter),  $Z$ , and decreases with the energy of the photon.

As can be seen in Figure 3.2, for photon energies in the visible spectrum (of the order of eV), the Compton effect is only likely for very light materials, ( $Z < 4$ ). For heavier materials the photoelectric effect is the dominant effect.

Finally, for coherent scattering, the atom is neither excited nor ionized and the photon conserves all its energy in the collision. Coherent scattering is more probable for photons with low energies and materials with

high atomic numbers and, as it will be shown in section 3.2.2, it explains why the produced photons are guided along scintillating fibers.

### 3.2.2 Plastic Scintillators

Scintillators are widely employed for radiation detection in nuclear physics. Scintillator converts kinetic energy of the incoming particles in to light<sup>2</sup> which can be detected and quantified. Light emission happens through photon de-excitation of excited atoms.

Light production is linear in a wide energy range of incoming particles. Scintillators should have good optical properties, such as being transparent to the wavelength of their own emission and having a refractive index as close as possible to that of glass for optimizing optical coupling with photosensors. Photon emission in scintillators is a statistical process, which means that two identical events will emit a different number of photons that follows a poisson statistics.

Scintillators can be organic and inorganic. Inorganic scintillators normally have a higher atomic number and density so their light output are higher. Due to these reasons they are better for gamma-ray spectroscopy. Organic scintillators are generally faster and they are commonly used for beta spectroscopy and neutron detection. This section is focussed on organic scintillators since they are the ones used in the TRITIUM project.

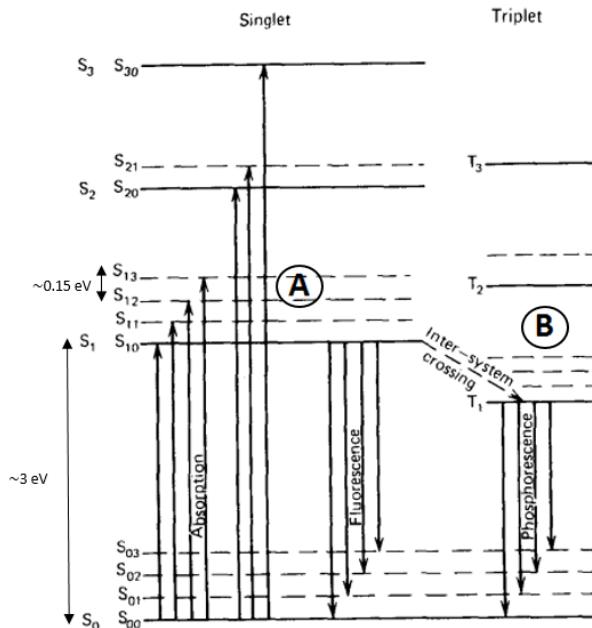
Organic scintillators are based on a scintillator material dissolved in a base solvent, normally aromatic hydrocarbons as C<sub>18</sub>H<sub>14</sub>, C<sub>24</sub>H<sub>22</sub>N<sub>2</sub>O or C<sub>15</sub>H<sub>11</sub>NO with an average atomic numbers of which are between 3,5 and 5.

The scintillator molecules, in which the organic scintillators are

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<sup>2</sup>The light is made up of photons in the visible energy range.

based, have a  $\pi$ -electron structure. The energy levels of their electrons are commonly illustrated with a Jablonsky diagram, shown in Figure 3.3, which shows the fundamental singlet state,  $S_{0i}$ , where the valence electrons are, the excited singlet states,  $S_{jk}$ , and the excited triplet states,  $T_{lm}$ . The energy difference between  $S_1$  and  $S_0$  states is around 3 or 4 eV, in the visible range. As it is shown in the figure, each energy states are splitted in close sublevels separated around 0.15 eV. This fine energy structure is due to excitations of molecular vibrational modes tabbed by the second index of the energy states. As the energy levels and sublevels have an energy larger than the thermal energy, 0.025 eV, non-excited electrons are in the ground state  $S_{00}$  at STP<sup>3</sup>.



**Figura 3.3 –** Jablonsky diagram. [Kno99]

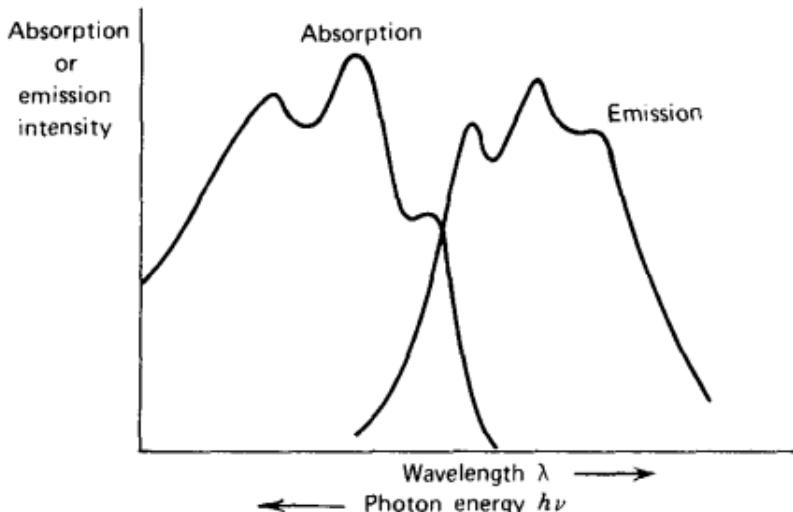
When a particle deposits their kinetic energy in a scintillator, their valence electrons are excited to higher singlet energetic states very fast (times

<sup>3</sup>Standard temperature and pressure conditions

of the order of picoseconds) and are quickly de-excited to the first singlet excited state,  $S_{10}$ , through non-radiative processes known as internal conversion. These electrons can de-excited to the fundamental single state,  $S_{00}$ , through three different physical mechanisms:

- Prompt fluorescence(process A in Figure 3.3), where the electron in the  $S_{10}$  energy level is de-excited to some sublevel of the ground state  $S_{0i}$ , emitting a photon. This process happens immediately after the excitation of the scintillator molecules (around tens of nanoseconds after excitation). Each scintillator has a characteristic emission spectrum that defines its response due to the fluorescence mechanism.

Organic scintillators are practically transparent to their own fluorescence emission because there exist a quenching effect in each de-excitation process by which all emitted photons by the scintillator have less energy than the excitation. This effect is called Stokes shift and it is represented in Figure 3.4.



**Figura 3.4 – Stokes shift. [Kno99]**

The intensity of the fluorescence emission in an organic scintillator

over time is a combination of two exponential functions, one associated with the lifetime of the level,  $\tau$  (on the order of nanoseconds), and the other associated with the energetic level population,  $\tau_1$  (on the order of picoseconds) [Kno99].

$$I = I_0 (e^{t/\tau} - e^{t/\tau_1}) \quad (3.6)$$

- Phosphorescence, where the electron that is in the first single excited state cross to a triple excited state (process B in Figure 3.3) with a process called "intersystem crossing". This is a metastable state with a longer lifetime than phosphorescence. This process happens around  $10^{-3}$  seconds after scintillator excitation.
- Delayed fluorescence, which occurs when an electron is in a triple excited state but its transition to the ground state is forbidden. In this case, this electron interacts with another electron in a similar state, falling and return to the first singlet state and quickly de-exciting to the ground state.



This emission has the same emission spectrum as immediate fluorescence, but occurs later.

As the prompt fluorescence light produces the scintillator signal, detector design should increase it and reduce other possible physical mechanisms. One of the most important parameters is the scintillation yield<sup>4</sup>, defined as the the number of photons emitted by unit of absorbed energy. This yield depends on the type of particle and on other mechanisms that doesn't produce prompt fluorescence, like phosphorescence or delayed fluorescence

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<sup>4</sup>The scintillation yield is a way of expressing the efficiency of the scintillator in converting the energy deposited by the particle into photons.

or even internal conversion. The scintillator yield is normally quoted by the manufacturer for mips<sup>5</sup>.

Plastic scintillators are easy to machine to any desired shape. The chosen shape for TRITIUM detector is the fiber, specifically, commercial fibers BCF-12 from Saint-Gobain Crystals Inc company [Cer05]. This type of fiber was chosen as the result of a comparative study [Cam17] among some of the best-known commercial manufacturers.

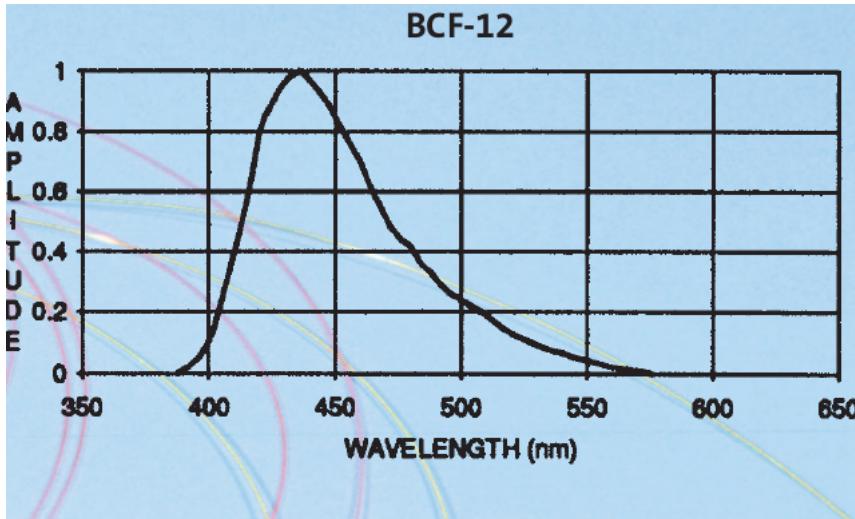
The BCF-12 fibers consist of a scintillating of polystyrene with the possibility of surrounding it by a cladding of polymethylmethacrylate (PMMA) (smaller refractive index than core in order to achieve a critical angle) or a multicladding (second cladding) with even smaller refractive index.

When a particle deposits all or part of its kinetic energy, some photons are produced in the fiber core as a result of the scintillating process. The number of photons produced depend on the scintillation efficiency and its value is around 2.4% for the fibers used (BCF-12), which means that a scintillation yield of 8000 photons will be produced per MeV for a mip. For instance, for tritium electron, these fibers release a maximum of around 148 photons (when tritium electron has the maximum energy, 18.6 keV), or less as electrons of these energies are not mips. The emission spectrum of the fibers employed in this work, is shown in Figure 3.5.

The scintillation light is guided to the sensitive part of the photosensor. A single photon produces a signal with some probability, called the quantum efficiency. Fibers (and scintillators in general) use the optical property of Snell's law [?] to guide their photons to the desired part (ends of the fibers). It is based on the interface created between the core and the surrounding material. When a photon hits this interface, it is refracted

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<sup>5</sup>The MIP, Minimum Ionized Particles, is a particle that has the speed that generate minimum ionization, that's, for example, electrons with 500 keV or more



**Figura 3.5** – Emission spectrum of BCF-12 fibers of Saint-Gobain. [Cer05]

(and therefore lost) following the Snell equation, 3.8 [?].

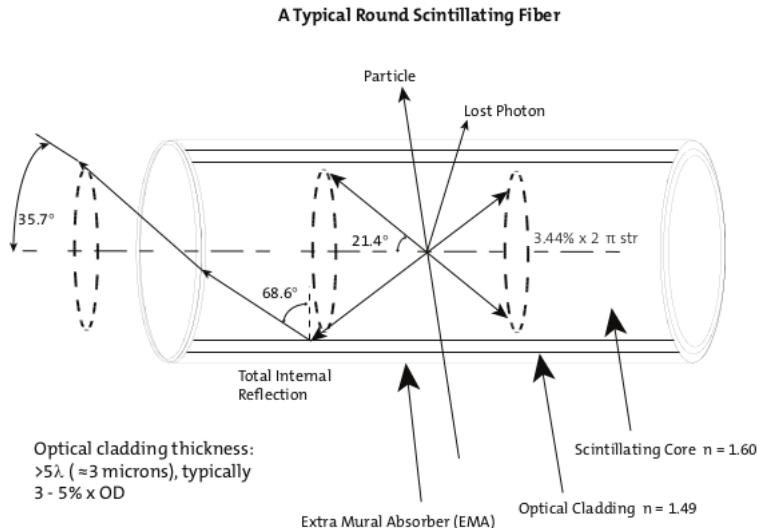
$$n_0 \sin(\theta_0) = n_1 \sin(\theta_1) \longrightarrow \theta_c = \arcsin\left(\frac{n_1}{n_0}\right) \quad (3.8)$$

If the surrounding material has a lower refractive index than the core of the fiber, there exist a critical angle,  $\theta_c$ , beyond which photons will be totally reflected and therefore kept within the fiber as illustrated in Figure 3.6.

The trapping efficiency or photon collection efficiency is defined as the efficiency of the scintillator to guide photons. For BCF-12 fibers with optical clad is between 3.44% and 7% per meter of fiber (depending on where the event is detected and is minimum near the fiber axis and maximum near the core-clad interface). For uncladded fibers BCF-12 surrounded by water, the trapping efficiency is larger than for cladded fibers. Therefore, from the maximum of 148 photons initially created by a tritium decay electron with the maximum energy, only 41 photons (for maximum trapping efficiency)

are guided in the 25 cm fiber length used in the TRITIUM detector. Thus, the output signal is very weak and is in the range of the spectrum where electronic noise is already significant. As described in the following chapters, a great effort was made to minimize electronic noise by different techniques.

In Figure 3.6 the light collection in a fiber is illustrated.



**Figura 3.6** – How photons are collected in a fiber with single clad. [Cer05]

The cladding material is useful for protecting the core surface from dirt or aggressive external agents that may reduce the light collection but at the cost of increasing the critical angle which its corresponding loss of light. Three different cases are shown in Table 3.1, where the cladding effect is illustrated.

In the practice, it is difficult to achieve a perfect air-core or water-core interface which affects light collection. As commercial claddings are thicker (30  $\mu m$ ) than the mean free path of tritium decay electrons in water (around 5  $\mu m$ ) cladded fibers is not an option for the TRITIUM detector. Hence, special attention is needed for achieving a water-core interface good enough. To overcome this problem a special protocol was developed in the

| Material         | Refractive index | critical angle ( $^{\circ}$ ) |
|------------------|------------------|-------------------------------|
| Air              | 1                | 42.98                         |
| Water            | 1.33             | 62.47                         |
| Cladding of PMMA | 1.49             | 76.26                         |

Table 3.1: Critical angles associated to different interfaces created with polystyrene,  $n_0 = 1.6$ , and other materials

ICMOL laboratory for preparing fibers for tritium detection.

The most important parameters of scintillating fibers of TRITIUM are given in Table 3.2.

### 3.2.3 Light Detection in Photosensors

The scintillating photons created in the core of the fiber and guided to its end are detected by photosensors. Photosensors have a sensitive part that is optimized to detect photons in a range of energy (usually in the visible range) with a certain probability, called quantum efficiency. The photosensors produce an electronic signal that carries information about the detected photons such as number, detection time, etc.

There are many available photosensors that rely on various physical processes, such as photoelectron multiplier tubes (PMTs), silicon photo-electron multiplier (SiPM) or charge-coupled device (CCD).

To optimize the efficiency of a scintillation detector is essential that the emission spectrum of the scintillator (Figure 3.5 for the fibers used) overlaps as much as possible with the detection efficiency spectrum of the photosensor chosen. The detection efficiency spectrum shows the probability of detecting photons at a function of wavelength. The efficiency of a detector is proportional to the product of both, the emission and the

|                                      |   |
|--------------------------------------|---|
| Core material                        | Polystyrene                               |
| Core refractive index                | 1.60                                      |
| Density (g/cm <sup>3</sup> )         | 1.05                                      |
| Cladding material                    | Acrylic (PMMA)                            |
| Cladding refractive index            | 1.49                                      |
| Cladding thickness ( $\mu\text{m}$ ) | 30  |
| Numerical aperture                   | 0.58                                      |
| Trapping efficiency                  | 3.44% minimum                             |
| No. of H atoms per cc (core)         | $4.82 \cdot 10^{22}$                      |
| No. of C atoms per cc (core)         | $4.85 \cdot 10^{22}$                      |
| No. of electrons per cc (core)       | $3.4 \cdot 10^{23}$                       |
| Radiation length (cm)                | 42  |
| Emission peak (nm)                   | 435 (Blue)                                |
| Decay Time, (ns)                     | 3.2                                       |
| 1/e Length (m)                       | 2.7                                       |
| Scintillator yield (# $\gamma$ /MeV) | $\sim 8000$                               |
| Operating Temperature                | $-20^\circ\text{C}$ to $50^\circ\text{C}$ |

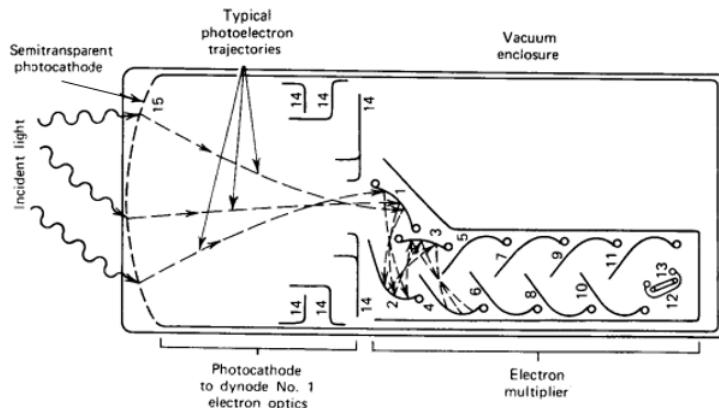
Table 3.2: Properties of BCF-12 fibers from Saint-Gobain Inc. [Cer05]

detection efficiency spectra, and this is largest when both spectra match.

The proposal of TRITIUM is to use SiPM arrays because they are very fast (of the order of ns) and have a high photodetection efficiency of about 50%, high gains (multiplication factor of  $10^6$ ) and a low voltage supply. The most important reason of this choice is that SiPM arrays are able to detect a single photon with high efficiency, which is a fundamental aspect due to the low amount of photons generated by tritium decay. The PMTs, which are the conventional choice, were also tested because they have lower dark count rate than SiPM and similar properties like gain and timing.

## Photoelectron Multiplier Tubes (PMTs)

Photoelectron multiplier tube, PMT, employed as photosensors in nuclear physics during decades, detect the scintillating photons that reach its sensitive part, photocathode, and produce an electronic signal, large enough to be easily measured. In Figure 3.7 a schematic drawing of a PMT is given. The PMT consists of a vacuum tube that has a glass window through which photons can penetrate. The electrons created in the photocathode travel in vacuum.



**Figura 3.7** – Scheme of a PMT. [Kno99]

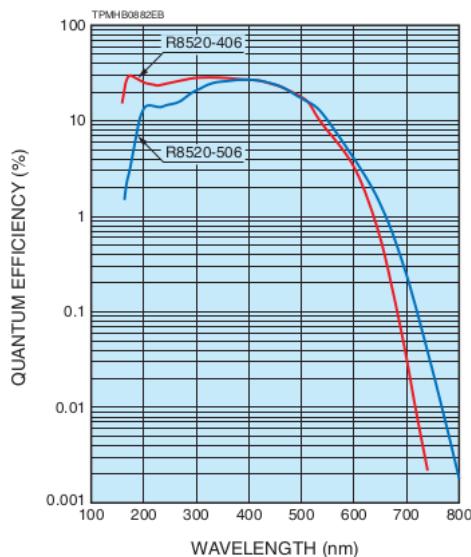
The signal production has two phases:

1. In the photocathode, photons are converted in photoelectrons through photoelectric effect. The photocathode consists of a thin layer, of the order of nanometers, deposited on the inner surface of the PMT window. The material of the photocathode is chosen to optimize the probability of producing photoelectric effect with the scintillating photons. The PMTs used in TRITIUM experiment are the model R8520-406 from Hammatsu [K.K19] and the material of their photocathode is Bialkali<sup>6</sup>.

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<sup>6</sup>The bialkali material is based on the elements  $^{121}\text{Sb}$ ,  $^{85}\text{Rb}$  and  $^{132}\text{Cs}$

The response of the PMT at long wavelengths is limited mainly because photon energy is not enough to produce a photoelectric effect or the emitted photoelectron does not have enough energy to overcome the material-work function. The response of the PMT at short wavelengths is limited due to absorption in the window material, quartz in our case. Thus, the response of the PMT has a strong dependence on the energy of the photon and the quantum efficiency (QE) spectrum defined is given by the ratio of the number of photoelectrons produced at the cathode of the PMT and the number of photons reaching it. For PMTs used in the TRITIUM experiment, the QE is showed in Figure 3.8.



**Figura 3.8** – Quantum efficiency spectrum for the PMT used (R8520-406). [K.K19]

The maximum values of the PMT quantum efficiency is usually between 20% and 30% [Kno99] (a little bit less than 30% for the PMTs employed). The emission spectrum of the scintillating fibers used, Figure 3.5, matches the quantum efficiency spectrum of the PMTs used, Figure 3.8 and the position of both peaks is very close, 435 nm for

fibers and 420 nm for PMT.

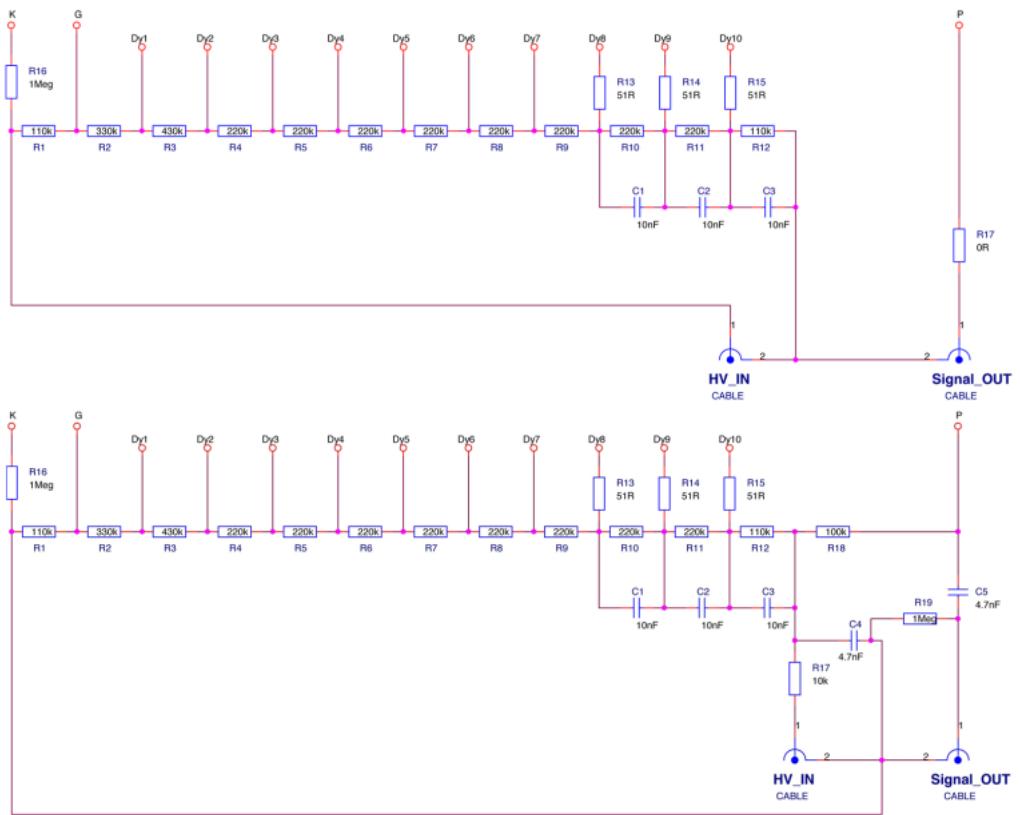
2. As the number of photoelectrons produced in the photocathode is very small, an electron multiplication stage is employed to obtain an electronic signal of sufficient size to be processed by the electronic system. The amplification stage is based on three elements, focusing electrodes, dynodes and anode, which are metallic plates with a shape and position designed to optimize the collection and multiplication of electrons. A high voltage (HV) is applied to the PMT which is distributed between all this elements, including the photocathode, with the help of electronic circuit. A positive HV, grounded in the photocathode, is interesting for measuring PMT currents, and a negative HV, grounded in the anode, gives a faster response. The commercial electronic circuits of Hamamatsu are shown in Figure 3.9.

Focusing electrodes guide the photoelectrons to the first dynode. They have a collection efficiency (CE) defined as the ratio of the number of photoelectrons reaching the first dynode and the number of photoelectrons leaving the photocathode and its value is around 80%. The dynodes achieve the electron multiplication. A voltage difference between adjacent dynodes accelerates the electrons and produce their multiplication. The multiplication factor of each dynode,  $\delta$ , is commonly around 5 and is strongly dependent on the HV. If all dynodes have the same gain, the overall gain of a PMT with  $N$  dynodes is [Kno99]:

$$G = CE \cdot \delta^N \quad (3.9)$$

that give an overall gain of a PMT of the order of  $10^6$ , strongly dependent on the applied HV.

The multiplication stage adds an uncertainty in the measurement. Working without gain allows to count the number of photons that reach the PMT. This can be done by short-circuiting all the dynodes and the anode and collecting the signal directly of the photocathode. This special setup was used for fiber characterization, described in



**Figura 3.9** – Hamamatsu commercial voltage divider electronic circuit. Upper circuit with negative supply and lower circuit with positive supply. [K.K19]

section 4.1.3.

The output pulse of a PMT has a width of the order of tens of nanoseconds. The multiplication process can be described as a Poisson statistical process. For each electron in the first dynode,  $G$  new electrons are created with a variance of  $\sqrt{G}$ .

The output signal of a PMT is linear with the number of photons that reach its sensitive part up to a saturation limit, at which the linearity

is lost. This limit depends on the PMT model.

The photocathode may emit electrons without any scintillation light. This signal is named dark current,  $I_{DC}$ , and can arise due thermoionic emission. For the PMTs used, this value is around 2 nA according to their data sheet.

The characterization of the PMTs used for dark current, gain for several HV and quantum efficiency, was done at IFIC in the framework of NEXT experiment [Pé10].

### Silicon Photoelectron Multiplier Array (SiPMs array)

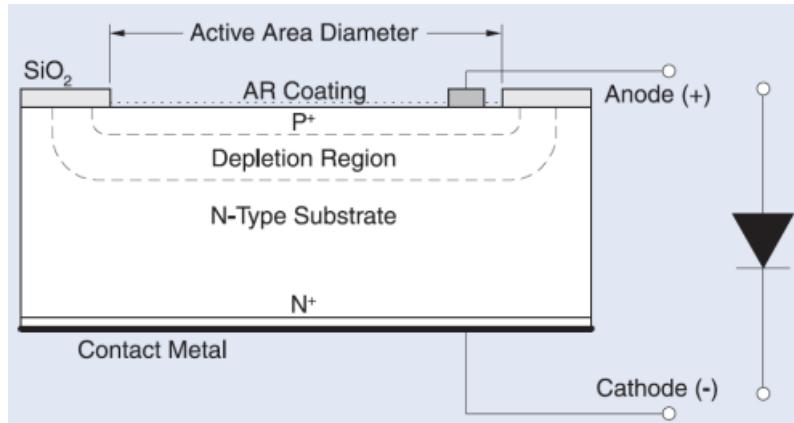
The Silicon Photomultiplier (SiPM) are a kind of photosensor, based on semiconductor materials, developed in recent years. They are replacing progressively conventional PMTs in many experiments and applications. They achieve outstanding photon-counting capabilities with high photodetection efficiency comparing to PMT and similar gain. They have convenient characteristics as insensitiveness to magnetic fields, low operating voltage and compactness. The main problem with the SiPMs are their high dark count rate (between 100 kHz and 1 MHz).

The SiPM is formed by a matrix of APDs connected in parallel which are photodiodes operating in Geiger mode. APDs, the scheme of which is shown in Figure 3.10, are based on p-n junctions<sup>7</sup> made with special techniques to achieve a good contact between both surfaces.

The voltage at which the SiPM starts operating in geiger mode is called the breakdown voltage,  $V_{BR}$ . At lower voltages SiPM works in proportional mode in which the signal of the pixel is proportional to the

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<sup>7</sup>A p-n junction is a junction of a p+ and n+ layer, which are a tetravalent material doped with a trivalent or pentavalent material respectively, creating interesting sublevels in the forbidden energy gap.



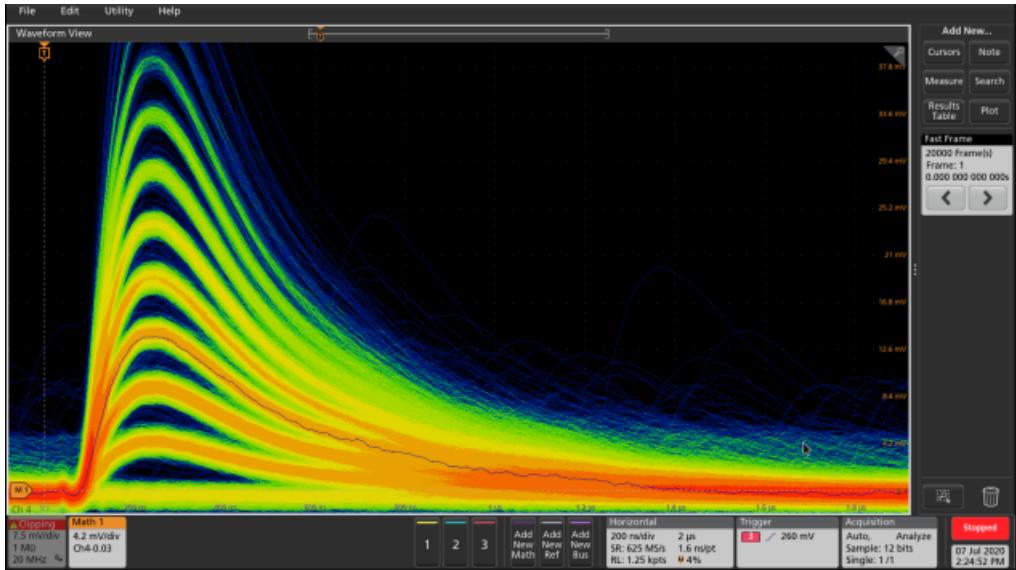
**Figura 3.10** – Scheme of a APD and electrical symbol used. [Opt]

energy deposited but its gain is lower than in Geiger mode. The experimental measurement of the breakdown voltage, described in section 4.2, is an important measurement to characterize a SiPM since many properties of the SiPM depends on the overvoltage,  $V_{ov}$ . The overvoltage is the voltage applied to the SiPM above their own breakdown voltage and this is expressed as:

$$V_{bias} = V_{BR} + V_{OV} \quad (3.10)$$

These APDs, called pixels when they are part of a SiPM, are connected in parallel and the sum of all of them is read. The output signal of an individual pixel is quite similar regardless of the energy deposited, with some difference because of the uncertainty due to the SiPM manufacturing process and the statistical nature of the detection process. Therefore, the energy deposited in each APD is not known but the charge of the output signal when  $n$  pixels are simultaneously fired is  $n$  times the charge of a single pixel, as can be checked in Figure 3.11. Due to this property, considering that each pixel only detect one photon, the number of detected photons is linearly proportional to the area of the output signal. Hence, after a correct

calibration of SiPMs, shown in section 4.2, the linearity of the SiPM output signal and the deposited energy by the tritium event is recovered.



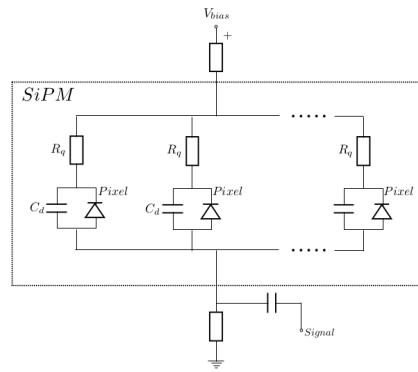
**Figura 3.11** – Using persistence on the oscilloscope to show several pulses with different heights. Each height associated with a different number of SiPM pixels lit at the same time.

The size of a SiPM pixel has to be very small<sup>8</sup> to make sure that, for low enough photon densities to be detected, only one photon is detected in each pixel. If the photon density is high (typically several thousand of photons per event) more than one photon will be detected by the same pixel, generating an output signal equal to one detected photon. This effect, known as saturation, produces a loss of linearity of the output signal. However, this effect is not important for the TRITIUM detector since its scintillating signals are further away from producing it.

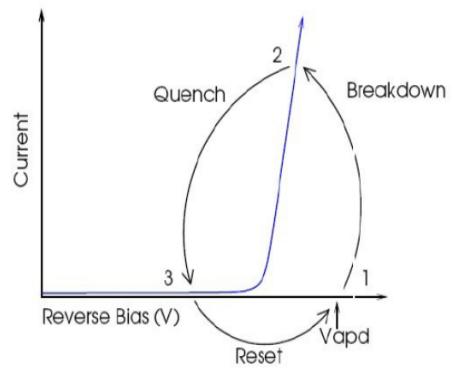
The SiPM can be modeled as the electric circuit, shown in figure 3.12a, in which, due to the charge distribution in the depletion zone, a capacitance are induced by the SiPM. This looks like a reverse diode in

---

<sup>8</sup>Pixel sizes for commercial SiPMs are 25, 50 or 75 $\mu\text{m}$  [Div16a], [Div16c], [Div16d]



(a) Photon Detection Efficiency, PDE.



(b) Output current of a SiPM.

**Figura 3.12** – (Left) Electronic scheme of a SiPM and (right) output current of a SiPM as a function of the reverse voltage. It show that the quenching mechanism is essential for working with SiPMs [sl17].

parallel with a capacitor with a capacitance of  $C_d$ . When the pixel detects a photon, the capacitor are discharged, creating an output current (electronic pulse).

In addition, each pixel of a SiPM has a quenching resistance<sup>9</sup>,  $R_q$ , in series that is used to stop the current produced when this pixel is fired. When the discharge is produced, a current flow through the resistance, reducing the reverse voltage seen by the diode to one below the breakdown voltage. Then, the current that flow through the diode is stopped and the voltage seen by the diode is reset to the bias voltage. This pixel is ready to detect a new photon again. This behaviour is schematically shown in Figure 3.12b.

The recovery of the bias voltage seen by the SiPM after a photon detection is that characteristic in a RC circuit, described by the equation:

---

<sup>9</sup>The tipical value of this quenching resistance for commercial SiPMs is around  $500\text{ k}\Omega$

$$V_{bias}(t) = V(t_0) (1 - e^{-t/\tau}) \quad (3.11)$$

where  $\tau$  is the recovery time constant of the system, the value of which is  $\tau = C_d \times R_q$  for RC circuits.

In section 4.2 the capacitance  $C_d$  and the quenching resistance  $R_q$  are experimentally measured and the recovery time constant extrapolated from both.

SiPM gain is defined as the charge produced when a single pixel is fired. This can be measured experimentally from its Single Photon Spectrum, SPS, which is the spectrum obtained when the SiPM output signal is integrated (charge) and histogrammed. The SiPM gain is typically of the order of  $10^6$ . The experimental measurement of the SPS and the calculation of the gain is presented in section 4.2.

It has to be taken into account that the SiPM gain is highly dependent on temperature, which cannot be controlled with sufficient sensitivity (less than  $1^\circ\text{C}$ ) in the final emplacement of the TRITIUM monitor. Therefore, a gain stabilization method was developed to compensate for the temperature effect. This method is detailed and tested in section 4.2.

Other important parameter for a SiPM used in the TRITIUM project is the photon detection efficiency, PDE. This is defined as the probability of recording the electrical pulse produced by a photon that hit the SiPM. The PDE of a SiPM consists of a product of three different parameters, the fill factor ( $FF$ ), which is the ratio between the active area of the SiPM and the total area, the quantum efficiency for photoelectron conversion ( $QE$ ), which is the probability that an incident photon generates an electron-hole pair, and the probability that the generated electron or hole produces an avalanche,  $P_{av}$ .

$$PDE = FF \times QE \times P_{av} \quad (3.12)$$

Like PMTs, sometimes the SiPM produces pulses that are unrelated with any incident photon, called dark current rate. The dark current rate depends on the temperature and, at temperatures around 25 °, these pulses are mainly produced by the thermal generation, which is produced when a free electron or hole start an avalanche. This signal is identical to the signal produced due to a detection of a single photon so they cannot be differentiated. Therefore, it is very important to determinate their importance on the tritium signal.

Electrons contained in an avalanche of a SiPM pixel emit secondary optical photons<sup>10</sup>. These optical photons can reach other pixels, producing new avalanches. This effect, called optical crosstalk, produces signals that are larger than those corresponding to the detected event, producing erroneous information about the number of photons read. The probability of producing an optical crosstalk event depends on the number of electrons produced in the avalanche (gain) and, therefore, depends on the temperature and the overvoltage. This is typically less than 10% at the recommended overvoltage and temperatures around 25°C.

The PED, dark count rate and Crosstalk are not experimentaly measured yet since a different setup, shown in reference [Yah13], is needed. These parameters will be measured for the SiPM model used in the final version of TRITIUM monitor.

Due to imperfections existing in the cristal lattice of a SiPM, called traps, an electron of an avalanche can be captured and released after a characteristic time,  $\tau_t$ . If this characteristic time is longer than the time used to recover the charge of the pixel, typically  $3\tau$ , this electron can trigger

---

<sup>10</sup>Around 20 secondary optical photons are emitted for SiPMs with gains of the order of  $10^6$  [Spi97]

a new avalanche which will be seen as a new event. Afterpulses events are often emitted around  $1 \mu\text{s}$  after some SiPM output pulses so they don't affect to the tritium measurements since time coincidence are always done, using time windows of the order of 100 ns.

The initial candidate for TRITIUM project and the one which was characterized is the model S13360-1375 from Hamamatsu Photonics [Div16b] because this has interesting characteristics and properties, shown in Table 3.3. A large pixel size,  $75 \mu\text{m}$ , was chosen because this implies high PDE and high gain, both parameters are important for the TRITIUM project due to the low activity to be detected and the small signals produced by tritium events, respectively. This is achieved at the cost of reducing the dynamic range, which is not a problem due to the small signal generated by tritium events.

These values, provided by Hamamatsu photonics, are only approximate for a given element. Therefore, these parameters must be experimentally measured for each SiPM used because they can vary significantly even for SiPMs of the same model. Some most important characteristics for the TRITIUM project are experimentaly measured and given in section 4.2.

This model was later replaced by the model S13360-6075 from Hamamatsu Photonics [Div16d]. Its main difference is its higher active area ( $6 \times 6 \text{ mm}^2$ ) with which larger active area of scintillating fibers can be read. This is achieved at the price of a higher dark count rate (typically 2 Mcps) and crosstalk probability (7%). In addition, commercial matrices are available covering a total size of  $24 \times 24 \text{ mm}^2$ .

Although TRITIUM detector uses whole SiPM matrices, the characterizations has been carried out at the level of a single SiPM to learn about the values of the SiPM parameters and to test the gain control method. A new experimental setup, detailed in appendix ??, is already prepared to

| Parameter  | Numerical value  |
|--|------------------|
| Serie  | S13360           |
| Model  | 1375             |
| Pixel Pitch ( $\mu\text{m}$ )                                    | 75               |
| Effective photosensitive area ( $\text{mm}^2$ )                  | $1.3 \times 1.3$ |
| Number of pixels   | 285              |
| Fill factor  | 82%              |
| Refractive index of windows material                             | 1.55             |
| Operating temperature range ( $^\circ\text{C}$ )                 | [-20, 60]        |
| Spectral response range, $\lambda$ (nm)                          | [320, 900]       |
| Peak sensitivity wavelength, $\lambda_p$ (nm)                    | 450              |
| PhotoDetection Efficiency, PDE, $\lambda = \lambda_p$ (%)        | 50               |
| Dark counts, Typical/Maximum (kcps)                              | 90/270           |
| Terminal capacitance, $C_t$ (pF)                                 | 60               |
| Gain, M,   | $4 \cdot 10^6$   |
| Breakdown Voltage, $V_{BR}$ (V)                                  | 53               |
| Cross talk probability(%)  | 7                |
| Temperature coefficient $\Delta TV_{op}$ (mV/ $^\circ\text{C}$ ) | 54               |

Table 3.3: Characteristics of SiPM S13360-1375 from Hamamatsu Photonics [Div16b].

perform a complete characterization of the SiPM model S13360-6075.

### Comparison of photosensors considered

The photosensors employed in TRITIUM are both, PMT and SiPM. Each kind of photosensor has his advantages and disadvantages, so both were tested to make a final choice. The output signal of both photosensors is proportional to the number of incident photons and they have a similar gain (of the order of  $10^6$ ). Both properties are essential to detect tritium events and to obtain a signal large enough to be measured and processed. Both photosensors have fast output signals, with a rise time shorter than nanoseconds, and a wide spectral sensitivity (200 – 800 ns for PMT and 300 – 900 ns for SiPM). The supply voltage necessary to work with SiPM, on the order of tens of volts, is much lower than that of PMTs, which require a high voltage, of the order of a thousand volts. The PDE at 420 nm, achieved with SiPM is higher, around 50%, than with PMT, which have a PDE about 30%. A large PDE is essential because the number of photons produced in a tritium event is very low. Furthermore PMTs, as they consist of a vacuum tube, are more bulky and fragile than SiPMs, which are compact and robust. This is an advantage for the SiPMs because TRITIUM detector should work during years. Furthermore, PMTs are rather more expensive, than SiPMs. In addition, PMTs are affected by magnetic fields, contrary to SiPMs that works correctly in intensities of magnetic field up to 7 Tesla. Moreover, due to their high uniformity, SiPMs are capable of distinguishing the exact number of photoelectrons detected and even of resolving a single photoelectron, which is not possible with PMTs due to variations in their gain.

On the other hand, the dark current of PMTs is much lower (a few counts per second) than that of SiPMs, that have a dark current between

0.1 and 1 Mcps<sup>11</sup>, depending on they size, and this happens almost entirely at the level of a single photoelectron. This prevents to separate tritium decay signals from background in the signal photon-detection zone. Another inconvenient of SiPMs is large crosstalk and afterpulses that need to be corrected.

An additional drawback of SiPMs is that their output signal depends strongly with the temperature. As TRITIUM detector will be installed in an environment with significant temperature variations, this problem is solved by a suitably changing the supply voltage to compensate temperature variations.

### 3.2.4 Electronic Readout

The electronic system is in charge of reading, processing and analyzing the output signal of photosensors and providing output information about the tritium detection. This electronic system depends on the type of detector configurations employed

#### Electronical system for PMTs

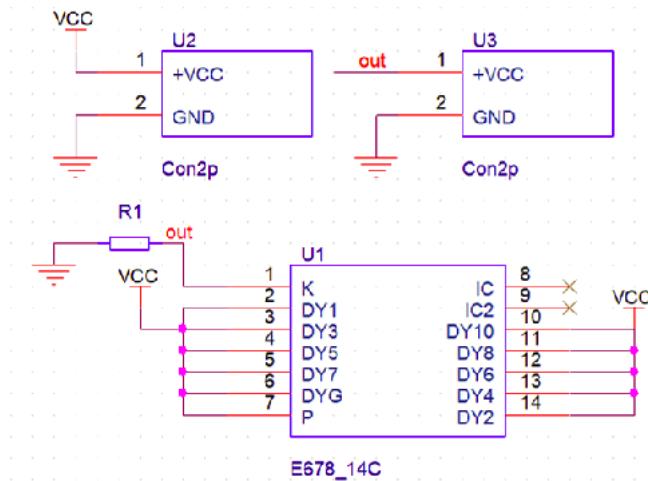
PMTs were used in TRITIUM experiment for two main objectives. On the one hand, to know the amount of incident photons that reached the PMT photocathode, which is important to characterize fibers, and, on the other hand, to know the energy of events, which allow us to obtain an energy spectrum and to discriminate events according to their origin.

To know the amount of photons that have reached the photocathode, the PMT should work without internal gain since it introduces a large uncertainty in the measurement. To this end, the electron multiplication

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<sup>11</sup>Mega counts per second,  $10^6$ c/s

stage (shown in section 3.2.3) must be employed. This is achieved with the help of a PCB, shown in Figure 3.13, designed, built and tested for this purpose.



**Figura 3.13** – Electronic scheme of the electronic voltage divider circuit used for working with PMTs without its internal gain.).

This PCB short-circuits the dynodes and reads the signal directly from the photocathode. This PCB was designed to be supplied with a positive voltage smaller than usual running voltage because it is only needed to create a voltage difference between the photocathode and the first dynode. As the signal is not multiplied, the output pulse of the photosensor is very small (currents of the order of tens of nanoamperes and a special readout system is needed. The chosen system is Keithley 6487 Picoammeter/Voltage Source [KEI], a commercial system from Keithley. This system has some useful options such as automatic baseline correction, the ability to read currents of the order of picoamperes and the possibility of carrying out mathematical operation on the signal, such as the average of N measurements with the associated statistical error, where N is programmable by the user ( $N = 100$  in all our studies). This is the configuration set to measure the output current of our photosensors. The number of photons

that has reach the photocathode is calculated from:

$$N^o \gamma / \text{sec} = \frac{(I_{PMT} - I_{DC})}{q_e \cdot QE \cdot CE} \quad (3.13)$$

where  $I_{PMT}$  is the output current of the PMT when it detects photons and  $I_{DC}$  is the dark current. This equation takes into account the quantum efficiency of the PMT, which is close to 30%, and the capture efficiency in the dynodes, equal to 1. In addition, it is assumed that each detected photon only generates one electron, the charge of which is  $q_e$ .

To determine the energy of the events, the internal gain of the PMT has to be restablished.

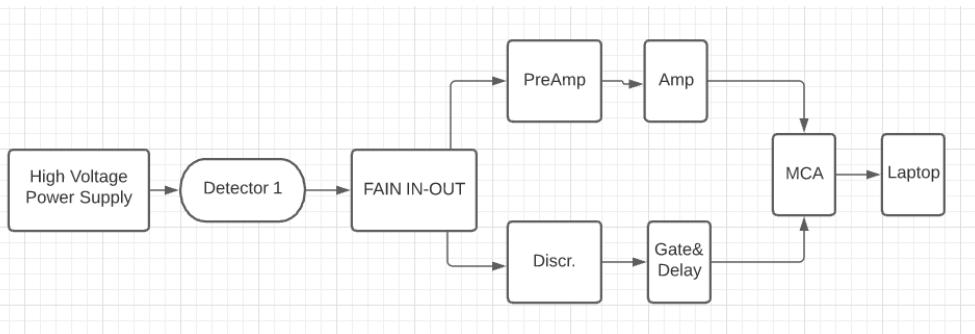
The number of PMTs used are one, two or four, depending on the measurement. A simplified scheme of the electronic chain employed in each case is shown in Figures 3.14a, 3.14b and 3.14c, based on various NIM technology modules<sup>12</sup>.

The PMTs are supplied in all the cases by TC 952 High Voltage Supply from Tennelec [Cam], which has four channels. If two or more configurations are needed, a second voltage supply HV Power Supply N 1130-4 from Wenzel Elektronik company [Ele] with 4 additional channels, was employed.

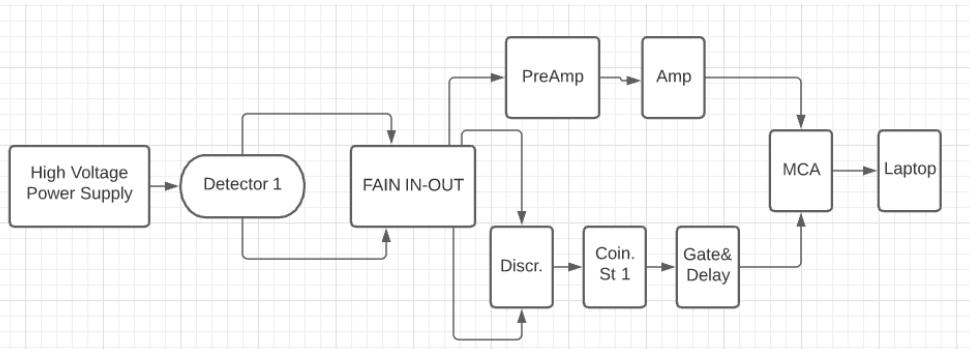
As it can be seen in the figures, there are two different lines followed by the PMT output signals, the amplification line and the time coincidence line. Therefore, the first module needed is an analogic FAN IN-OUT module which is used to duplicate the input signal. The module employed is the Quad linear FAN IN-OUT MODEL 740 from Philips Scintific [Sci], which has four channels. One output signals is used as the input for the amplification part and a second is used as the input for the time coincidence

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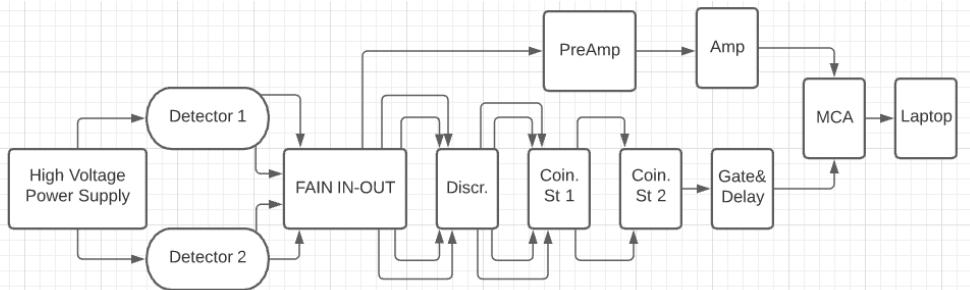
<sup>12</sup>The Nuclear Instrumentation Module (NIM) is a standard specification convention for electrical and mechanical parameters defined in electronic modules used in experimental nuclear and particle physics.



(a) Electronic scheme employed when only one PMTs are used in time coincidence.



(b) Electronic scheme employed when two PMTs are used in time coincidence.



(c) Electronic scheme employed when four PMTs are used in time coincidence.

**Figura 3.14** – Schemes of the different electronic for measuring with PMTs.

part.

1. The amplification line, which is the same for the three configurations, provides the energy information and is based on two steps:
  - (a) The output signals is integrated by a preamplifier, which gives an output signal with a height corresponding to the charge of the input pulse. This signal has a long tail<sup>13</sup> produced by the preamplifier capacitance. The preamplifier used is "MODEL 9326 FAST PREAMP" from ORTEC [ORTd].
  - (b) The output signal from the preamplifier is lead to the amplifier which gives an amplified output signal with a shape close to a Gaussian function. The used amplifier modules are 575A and 671 from ORTEC [ORTb, ORTc]. An example of the output signal for 575A module is shown in Figure 3.16, green color.
2. The time coincidence line contains the time information and gives the gate that triggers coincident signals of both PMTs. This line consists of:
  - (a) The output signals of the FAN IN-OUT module of each PMT is introduced into a discriminator module that gives a logic signal of -1.2 V height and of 240 ns width when a given threshold is exceeded. The discriminators utilized are Octuple Constant-Fraction Discriminator CF8000 module from ORTEC [ORTe] and 4 channels discriminator model 84 from CAEN [CAEa].
  - (b) Time coincidences are required to ensure that detected event comes from the scintillating fibers and to remove background like external light and dark current.

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<sup>13</sup>The length of the tail is,  $\tau = RC$ , where R is the input resistance and C is the capacitance used. It is the typical output signal in RC circuits.

The two logic signals given by the discriminator module that come from the two PMTs in the same detector are introduced in the coincidence module which generates an output signal of  $-1.4$  V height and of  $20$  ns width, when both are in coincidence.

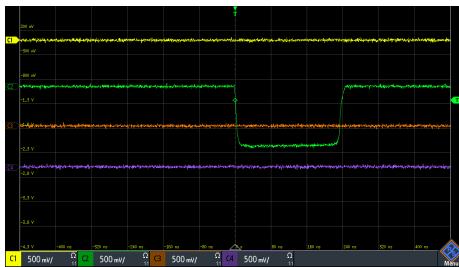
The modules used are Coincidence Unit Model 465 from LeCroy [LeC] and Coincidence Type N6234 from CERN-NP [CERb].

- (c) Time coincidence of two different detectors (4 PMTs, configuration 3.14c) was also studied, which is useful to remove background due to hard cosmic radiation.

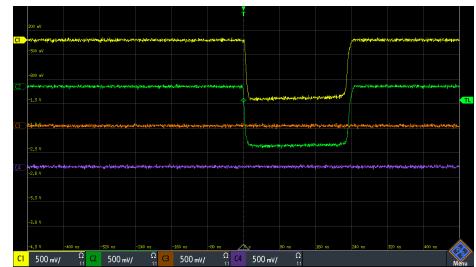
To do so, a new coincidence step similar to the previous one must be applied. The two logical output signals of the single detector coincidence are checked for coincidence.

Some examples are shown in Figure 3.15 for time coincidences of two detectors (4 PMTs). There, four logical signals are shown, two of them (channel one and two, yellow and green respectively) come from two PMTs connected to the first detector and the other two signals (channels three and four, color orange and violet respectively) come from PMTs connected to the second detector.

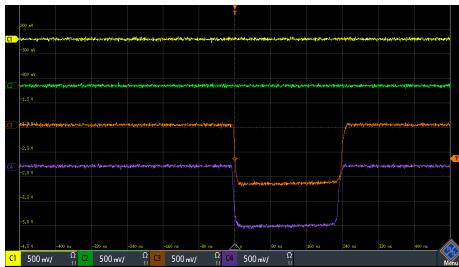
- i. In Figure 3.15a only one PMT (channel two) has detected an event. It means that the event is likely not produced in the scintillator. In this case, no output is generated.
- ii. In Figures 3.15b and 3.15c two PMT signals of the same detector are generated but the other detector gives no signal. This event is discarded.
- iii. In Figure 3.15d the four signals are detected, which means that the output signal is generated and the event is recorded.
- iv. The logical output signal, is introduced in the Gate and Delay Generator, model 416A of the company ORTEC [ORTa], which gives a positive logical signal, shown in Figure 3.16, orange color, with a height of  $8$  V and width of  $2 \mu\text{s}$ .



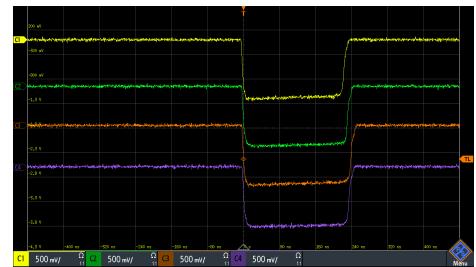
(a) Event detected in only one PMT, one detector.



(b) Event detected in two PMTs, one detector.



(c) Event detected in two PMTs, other detector.



(d) Event detected in all PMTs, both detector.

**Figura 3.15** – Different situation that can happen when time coincidences with PMTs are done.

At the end, a logical and analogical signals are obtained, shown in Figure 3.16, which are recorded by the MCA 8000D, Pocket MCA from AMPTEK [Amp]. The analogical signal has information about the energy of the event and this is the signal whose information we will save for analyzing. On the other hand the logic signal (output from the Gate and Delay Generator module) that indicates when the amplified signal must be saved.



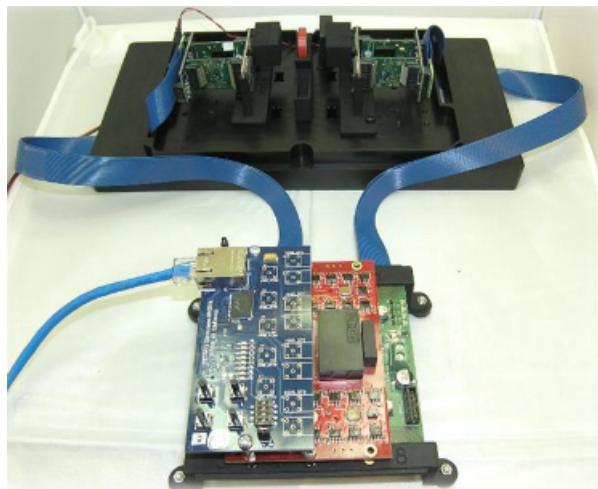
**Figura 3.16** – Signal amplified and logical gate (input signals of MCA).

### Electronical system for SiPMs

The SiPMs are used in the TRITIUM experiment in two different ways, at the level of a single SiPM and at the level of several SiPMs arranged in a matrix. Both studies were carried out to characterize the SiPM arrays used in the TRITIUM monitor.

The electronic system chosen to process and analyze the output signals of the SiPM arrays is PETsys [PET], which is a commercial system prepared to work with SiPM matrices from Hamamatsu.

PETsys is a complete acquisition and digitization system that is capable of working with up to 1024 SiPM. This system consists of a basic board, which processes the signal, to which 16 different SiPM matrices can be connected with up to 64 SiPM per matrix. This number of channels is needed in the TRITIUM project because, as it is shown in section 5.3, the TRITIUM monitor consists of a large number of SiPM matrices with 16 channels (SiPMs) per matrix. The PETsys system used in TRITIUM is displayed in Figure 3.17.



**Figura 3.17** – Different parts of PETSYS system. [PET]

Although the capacity provided by PETSYS should be enough for the requirements of the TRITIUM project, TRITIUM is a modular detector with scalable sensitivity. It means that, if an improvement of its limits is needed to improve its sensitivity or to further reduce the background, more photosensors would be needed. Therefore, the electronic system should be able to increase its capacity in a scalable way. This requirement is fulfilled by PETSYS since it has an additional module, called Clock and Trigger, to which up to sixteen different PETSYS basic boards can be connected. These sixteen PETsys basic boards are read in parallel, giving a total system

capacity of reading 256 SiPM matrices (16384 SiPMs<sup>14</sup>).

PETSYS is based on C++ and Python scripts that are prepared for the main tasks required, such as time coincidence options between SiPM (or even SiPM matrices) or energy discrimination. It is open source, giving the possibility to modify the current scripts or develop others with additional functions. PETsys has a time resolution of 250 ps which is one of the best time resolutions of commercial systems available and its price is around 10€/ channel, which is cheaper compared to similar electronic systems.

As described in section 4.2, the SiPM matrix temperature is an important parameter. The PETsys system has the ability to monitor the temperature of the SiPM matrices and ASICS employed to control them. Temperature monitoring is important to ensure the correct functioning of both, photosensors and system. PETsys has the possibility of developing new scripts to implement the stability gain method reported in section 4.2.

Although the TRITIUM monitor use SiPM matrices it is important to start the characterizaton at the level of a single channel (only one SiPM) to reduce the uncertainties in the first results. In order to do so, an electronic system was designed, developed and built to read up to eight different SiPMs and to monitor their temperature.

This system is based on three different PCBs<sup>15</sup>, shown in Figure 3.18 (electronical schemes shown in the appendix A):

1. The first PCB, shown in Figure 3.18a, is used to organize the SiPMs and sensor temperature. This PCB place up to 8 different SiPMs and a temperature sensor and arrange their output signals on two HDMI connections. This PCB is placed inside a special black box, from Thorlabs company [Thob], that has a high degree of light tightness. This

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<sup>14</sup> $1024 \cdot 16 = 16384$

<sup>15</sup>PCB, Printed Circuit Board

black box has a small hole of 1 mm diameter, prepared to introduce an optical fiber<sup>16</sup> to illuminate SiPMs with an incoherent light source. The light source utilized is a LED, model 430L from Thorlabs company [Tho18], which gives an spectrum shown in Figure 3.18d. The spectrum was experimentaly measured with a spectrometer and fitted to a Gaussian function. It can be seen that the emission peak of this LED is placed at 436.3 with a FWHM<sup>17</sup> of 19.1 nm. With the help of this LED the light emission of the fibers used in TRITIUM experiment is simulated to calibrate the SiPMs at the working wavelength.

2. The second PCB, shown in Figure 3.18b, sums the different signals of the SiPMs and amplify them by a factor  $G = 4187.5$  or  $G = 10761.88$ , depending on the input resistance of the oscilloscope,  $50 \Omega$  or  $1 M\Omega$ , respectively. This PCB uses a differential amplification that reduce the electronic noise of the system and is connected to the first PCB through two HDMI feedthroughs.
3. The third PCB, shown in Figure 3.18c, rearranges all the different input and output signals in an HDMI connection to avoid crosstalk between different signals. This PCB is connected to the second PCB through a HDMI feedthrough.

The input signals are the supply voltage of the SiPMs and the supply voltage of the PCBs ( $\pm 6$  V) and the output signals are the temperature sensor signal and the summed signal of all SiPMs.

The output signal of the third PCB is connected to an oscilloscope, model MSO44X from Tektronix [Tek21] that records the data which are subsequently analized by ROOT<sup>18</sup>.

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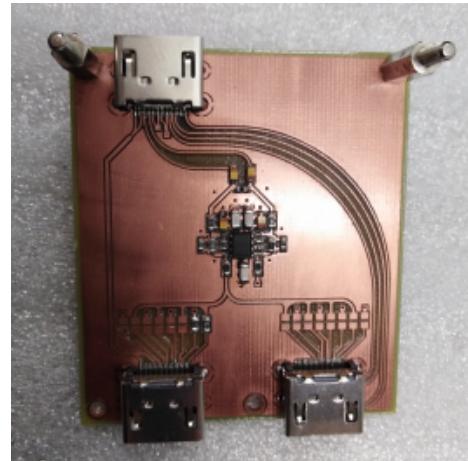
<sup>16</sup>The optical fiber used is BCF-98 from Saint-Gobain company [Cer21]

<sup>17</sup>The FWHM parameter, Full Width at Half Maximum, of a Gaussian fit can be calculated from its sigma using the equation:  $\text{FWHM} = 2.35 \cdot \sigma$

<sup>18</sup>ROOT is a framework for data processing, based on C ++ and object-oriented technology, developed at CERN and widely used in nuclear and particle physics.



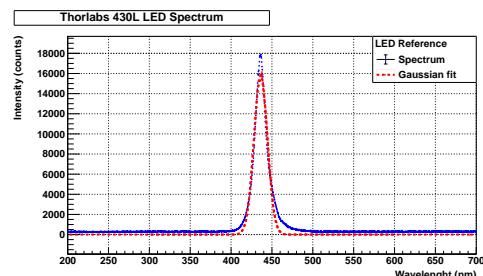
(a) PCB 1 used to arrange 8 SiPMs and black box.



(b) PCB 2 used to sum and amplify the output signals of SiPMs.



(c) PCB 3 used to rearrange the different signals of the system.



(d) Emission spectrum of the LED.

**Figura 3.18** – Three PCBs used for the SiPM characterization and LED emission spectrum.

## 3.3 Ultrapure Water System

### 3.3.1 Introduction to the Ultrapure Water System

The objective of the ultrapure water system is to purify the water sample before the measurement. This system is important for two reasons:

1. The mean free path of tritium electrons in water is around  $5 \mu\text{m}$  and even less in solid materials like organic material. The electron from the tritium decay has to reach the fiber to be detected and, consequently, the detector must be kept very clean. If the analyzed water sample contains particles that may be deposited on the fibers, a layer of matter can be formed, preventing the tritium electrons from reaching the fibers and reducing drastically the tritium detection efficiency.
2. The tritium monitor does not have any spectrometric capabilities that can be used to distinguish other radioactive elements from tritium. That means that, any radioactive event in the analyzed water sample would be counted as a tritium event.

The ultrapure water system was designed to remove all particles up to a diameter of  $1 \mu\text{m}$  and organic matter, which means that the only radioactive particle that passes through it is tritium.

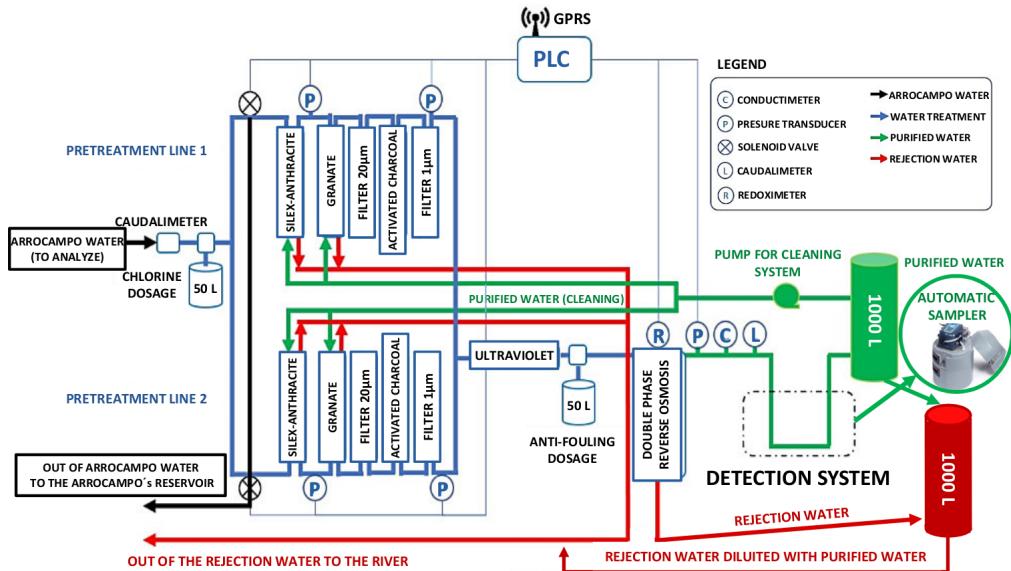
In summary, the ultrapure water system is used to keep our detector clean, ensuring the stability of its detection efficiency and to eliminate all radioactive particles other than tritium.

### 3.3.2 Set Up of Ultrapure Water System

The requirements of this water treatment device are:

- to obtain a high degree of purification of the processed water sample, reducing its conductivity by approximately two orders of magnitude (from  $1000 \mu\text{S}/\text{cm}$  to  $10 \mu\text{S}/\text{cm}$ )
- to require of low maintenance (low cost and low manpower)
- to install a remote control device with probes and valves controlling by software.

The LARUEX laboratory in Extremadura, one of the six collaborators of the TRITIUM experiment, has designed, developed and built the ultrapure water system, a scheme of which is shown in Figure 3.19.



**Figura 3.19** – Scheme of water purification system.

This system is installed in the Arrocampo dam and consists of four different consecutive stages:

1. The raw water from the Tagus River passes through two different filters, the first made of silex-anthracite and the second of garnet, with

which a rough filtering is made (the largest particles are eliminated). This system has two parallel lines and implements self-cleaning by injecting ultrapure water in the opposite direction.

2. The outlet water sample of the first stage, called fine filtration stage, passes through a  $20\ \mu\text{m}$  filter (formed by a synthetic mesh) and activated charcoal filters (one per line) that removes chlorine and iron particles.
3. The outlet water of the second stage passes through a super-fine filtering consisting of a  $1\ \mu\text{m}$  filter, formed of a dense polypropylene mesh and UV lamps. The first filter removes all the particles up to diameters of  $1\ \mu\text{m}$  and the UV lamps remove the organic matter present in the sample.
4. Finally, the water is introduced in the last stage, double-phase reverse osmosis, that reduces the conductivity of the water to about  $5\ \mu\text{S}/\text{cm}$ . It was verified that a conductivity of  $10\ \mu\text{S}/\text{cm}$  is achieved with only one module of reverse osmosis, enough for the needed conditions of tritium detector. Therefore, only one module of reverse osmosis is used for 24 h, reducing the power consumption of the system.

As a result of the purification process, besides the ultrapure water that is introduced into TRITIUM detector, a rejection water, with conductivities greater than the original water containing the particles extracted from the ultrapure water is produced.

The ultrapure water system is able to process up to  $0.850\ \text{m}^3/\text{h}$  with a single line operating or  $1.480\ \text{m}^3/\text{h}$  with both, greatly overestimating the requirements of the tritium detector.

The software used for remote controlling of the ultrapure water system is Siemens PLC, that gives the information such as the state of the valves, the pressure probes or water production in real time.

The appendix B contains several pictures of different parts of this system, installed in Arrocampo dam.

## 3.4 Background Rejection System of TRITIUM Monitor

The objective of the background rejection system is to reduce the TRITIUM radioactive background. The TRITIUM project follows the ALARA principle for the tritium activity measurement, that is, to measure tritium activity "as low as reasonably achievable". The detection limit of tritium activity is set by the uncertainty in the activity of the radioactive background since tritium activities below this uncertainty cannot be distinguished from the background. Therefore, the background uncertainty must be reduced as much as possible.

The total uncertainty is a quadratic sum of all the different uncertainties related to the measurement, i. e., the statistical uncertainty<sup>19</sup>,  $\sigma_{st}$ , the systematic uncertainty<sup>20</sup>,  $\sigma_{si}$ , etc.

The background rejection system of TRITIUM monitor minimizes the statistical component. Because of the Poissonian nature of the process, the statistical uncertainty corresponds to the square root of the measured activity,  $A_m$ , which can be reduced by minimizing detected background events.

$$\sigma_T^2 = \sigma_{st}^2 + \sigma_{si}^2; \quad \sigma_{st;bak} = \sqrt{A_{m;bak}} \quad (3.14)$$

The background of TRITIUM is due to natural radioactivity and

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<sup>19</sup>Uncertainty due to the statistical nature of the radioactivity process

<sup>20</sup>uncertainty due to the manufacture of the detectors

has two different sources. On the one hand, radioactive elements that are present in the crust of Earth, mainly  $^{40}\text{K}$  and elements from the four different natural radioactive series, shown in Table 3.4. On the other hand, the cosmic ray radiation. The primary cosmic radiation is composed of high-energy particles, mainly protons and  $\alpha$ , but, after interacting with the Earth's atmosphere, they generate a shower mainly composed by muons, electrons, photons and neutrons mainly.

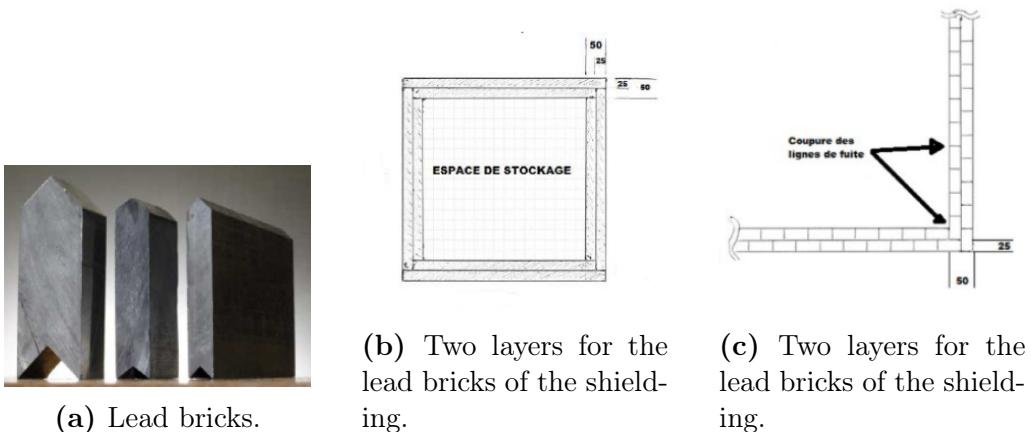
| Mass Num. | Series           | Prim. el.         | Half life (y)        | Final isotope     |
|-----------|------------------|-------------------|----------------------|-------------------|
| 4n        | Thorium          | $^{232}\text{Th}$ | $1.41 \cdot 10^{10}$ | $^{208}\text{Pb}$ |
| 4n+1      | Neptunium        | $^{237}\text{Np}$ | $2.14 \cdot 10^6$    | $^{209}\text{Pb}$ |
| 4n+2      | Uranium-Radium   | $^{238}\text{U}$  | $4.51 \cdot 10^9$    | $^{206}\text{Pb}$ |
| 4n+3      | Uranium-Actinium | $^{235}\text{U}$  | $7.18 \cdot 10^8$    | $^{204}\text{Pb}$ |

Table 3.4: Classification of natural radioactive series [The96, Eva96].

Cosmic radiation depends on several parameter like the altitude and latitude of the Earth, the altitude, sea level in our case, and the solar activity cycle. The spatial distribution of cosmic rays, mainly muons, follows a  $\cos^2(\theta)$  distribution with the zenith angle.

To remove the effect of background two different techniques are employed:

- On the one hand, the weak radiation, which is any radiation with energy below 200 MeV/nucleon is stopped by a lead castle, described in section 3.4.1,
- On the other hand, the hard radiation, that is any radiation of energy greater than 200 MeV/nucleon, is much more difficult to stop and the technique used is a cosmic veto, reported in section 3.4.2 in anti-coincidence with the TRITIUM detector.



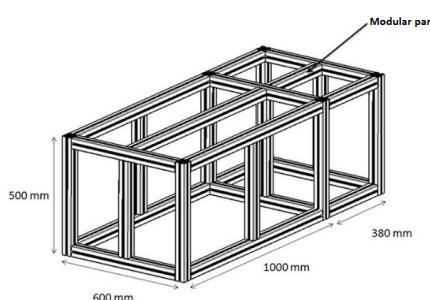
**Figura 3.20 – Lead Bricks and their arrangement in the lead shielding.**

### 3.4.1 Passive Shield (Lead)

Weak radiation is suppressed by a lead shielding inside which the TRITIUM detector is placed. This lead shielding is effective for particle energies below 200 MeV/nucleon, which is due to the Earth's natural radioactivity and to the weak component of cosmic radiation. This lead shielding consists of 158 lead bricks with ultra-low intrinsic radioactivity, 25 mm thick. They are chevron shaped, shown in Figure 3.20a, specially designed for a perfect fit and easy assembly. As can be seen in Figures 3.20b and 3.20c, these lead bricks are arranged in two layers with a total thickness of 50 mm. The junction of the inner layer lead bricks is shielded by a lead brick of the outer layer to avoid any leak of radiation.

Special aluminum structure was designed by mechanical engineering department of CENBG, shown in Figure 3.21, to support the total weight of the lead bricks, 2.4 tons.

The internal room of the lead shielding is divided in two parts, as exhibited in Figure 3.20. The larger one has internal dimensions of  $90.5 \times 41 \times 51$  cm<sup>3</sup> and is used to place the TRITIUM detector. The smaller



(a) Aluminium structure scheme.



(b) Aluminium structure of TRITIUM monitor.

**Figura 3.21** – Lead Bricks and their arrangement in the lead shielding.

one, of dimensions of  $33 \times 41 \times 51 \text{ cm}^3$ , contains the DAQ system. The external dimensions of the lead shielding are  $148 \times 60 \times 70 \text{ cm}^3$  and it weighs 2.5 tons.

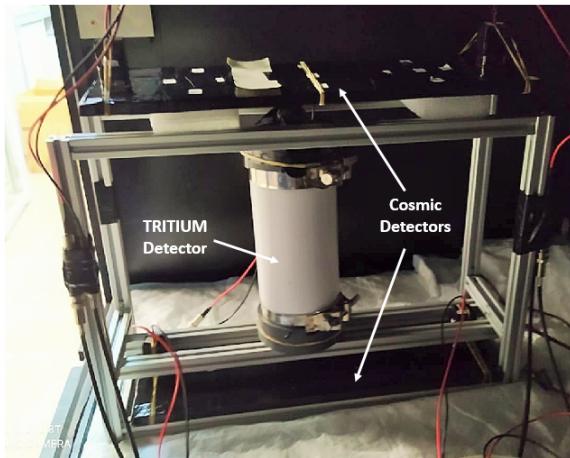
### 3.4.2 Active Shield (Cosmic Veto)

As hard radiation cannot be stopped by a moderate lead thickness cosmic vetos are employed.

A cosmic veto consists of at least two complementary detectors in coincidence that reject simultaneous events in both.

As shown in Figure 3.22, the two complementary detectors were placed one above and the other below the TRITIUM detector. The distance between both detectors, 34.2 cm for our latest prototype developed, is set by the TRITIUM prototype to be placed between both.

A hard cosmic events simultaneous through both cosmic detectors is displayed in figure 3.23a. Each cosmic detector has two photosensors. Hard cosmic events are rejected when both detectors are in coincidence



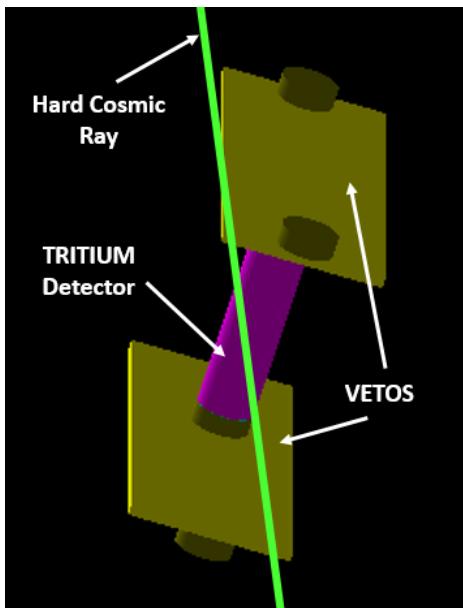
**Figura 3.22** – Cosmic veto and Tritium-IFIC 2 prototype in an aluminum mechanical structure developed by IFIC’s mechanical engineering department.

with the electronic configuration given in Figure 3.14c.

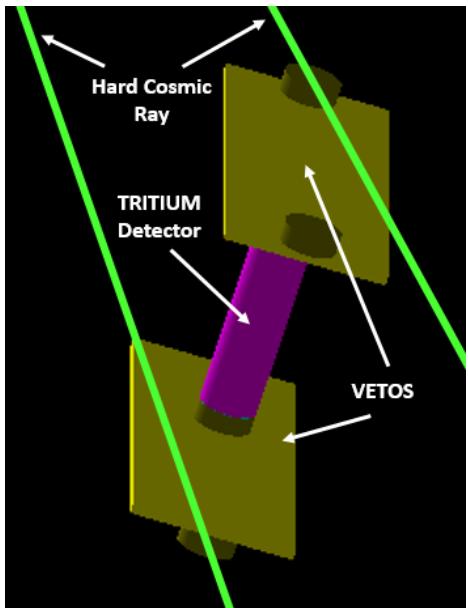
The TRITIUM detector is read out in anti-coincidence with the cosmic veto. Random coincidence from two different hard cosmic events, one in each detector, shown in Figure 3.23b are negligible. The expected hard cosmic rate at sea level for muons, main contributor, is  $70 \text{ m}^{-2}\text{s}^{-1}\text{sr}^{-1}$  [ea20, SAG01], that is  $7 \times 10^{-3} \text{ cm}^{-2}\text{s}^{-1}\text{sr}^{-1}$ , shown in the cosmic rate plot of Figure 3.24. As time coincidences are triggered by logical gates of about 10 ns, the probability of recording two different hard cosmic events in temporal coincidence is less than  $10^{-9}$  which is not worth considering.

The vetos are made of a plastic scintillator block from Epic-Crystal [Cry20]. Its properties are given in Table 3.5 and its energy emission spectrum is displayed in Figure 3.25.

The energy spectrum has a peak very close to that of the scintillating fibers used, so the same photosensors are used to read out them. The dimensions of the scintillator block are  $45 \cdot 171 \text{ cm}^3$  and they are wrapped by



(a) Real hard cosmic event.

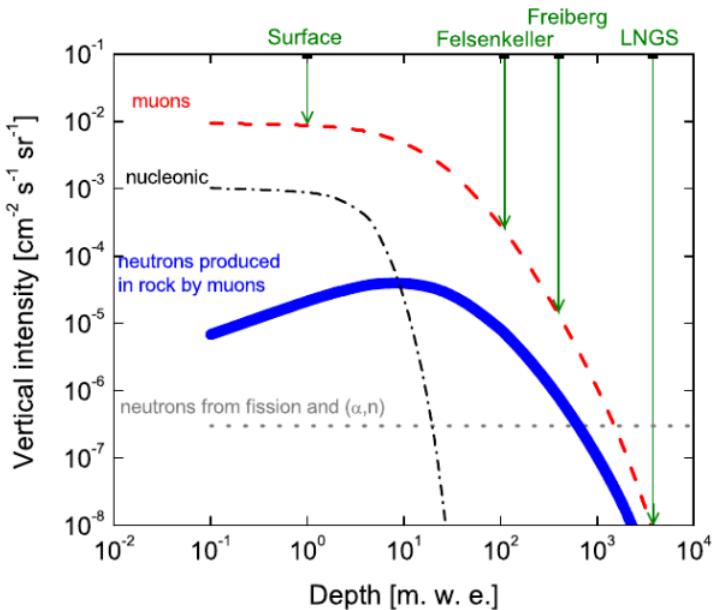


(b) Fake hard cosmic event.

**Figura 3.23** – Hard cosmic events detected with the cosmic veto of TRI-TIUM: a) Affecting to the tritium measurement, b) Does not affecting to the tritium measurement.

|                              |             |
|------------------------------|-------------|
| Base material                | Polystyrene |
| Growth method                | Polymeric   |
| Density (g/cm <sup>3</sup> ) | 1.05        |
| Refractive index             | 1.58        |
| Soften temperature (°)       | 75-80       |
| Light output (Anthracene)    | 50-60%      |
| H/C ratio                    | 1.1         |
| Emission peak (nm)           | 415 (Blue)  |
| Decay Time, (ns)             | 2.4         |
| Hygroscopic                  | No          |

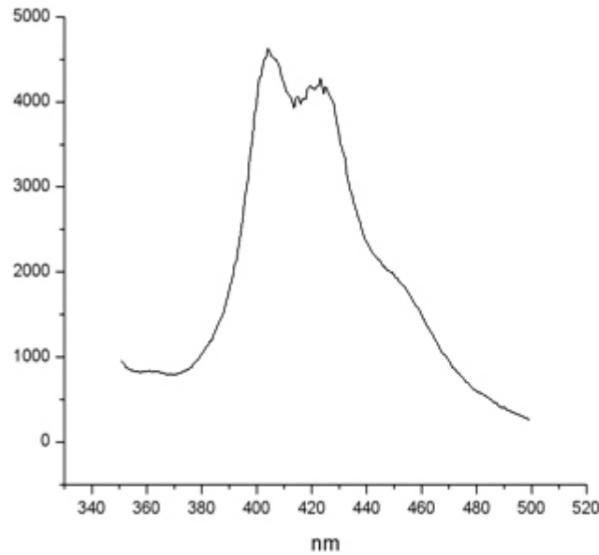
Table 3.5: Properties of plastic scintillators from Epic-Crystals. [Cry20]



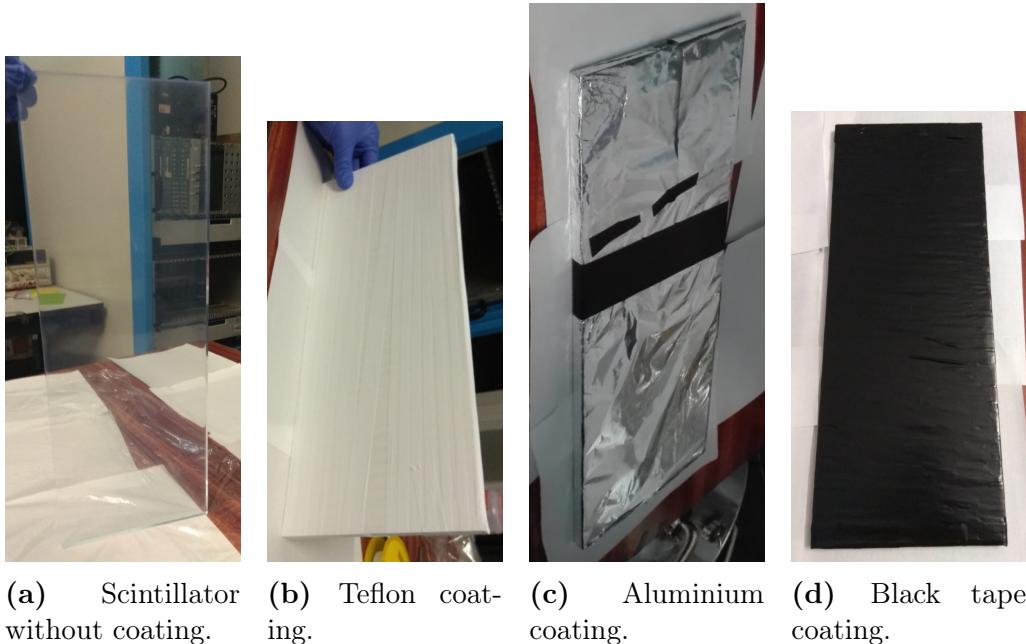
**Figura 3.24** – Hard cosmic muon rate [Szu15].

three different layers, teflon, aluminum and black tape, exhibited in Figure 3.26. These layers prevent external photons from reaching the scintillator plastic and avoid photons generated by the scintillator plastic from escaping before reaching the photosensor. Two  $2.5 \times 2.5 \text{ cm}^2$  windows are made on the wrapping to allow reading by the photosensors.

As previously mentioned, the expected hard cosmic rate at sea level is  $7 \times 10^{-3} \text{ cm}^{-2}\text{s}^{-1}\text{sr}^{-1}$ . As the solid angle of our detectors is  $\omega = 0.5434$ , calculated by integrating the solid angle of one scintillator on the other, and the area of the veto is  $765 \text{ cm}^2$ , the expected hard cosmic rate on our cosmic vetos should be 2,909 event/s. Thus is important to determine the efficiency of the cosmic veto.



**Figura 3.25** – Emission energy spectrum of the plastic scintillation used for the cosmic vetos. [Cry20]



(a) Scintillator without coating. (b) Teflon coating. (c) Aluminium coating. (d) Black tape coating.

**Figura 3.26** – Different layers used to cover of the cosmic veto.

# Chapter 4

## Research & Development on Detector Design and Components

This chapter describes the characterization of the different parts of the TRITIUM monitor, including scintillating fibers, SiPMs (at the individual SiPM level and at the matrix level), the ultrapure water system and the background rejection system. This characterization is crucial to understand their behaviour and the results of the monitor. Furthermore, several developments were made to improve fundamental parameters of the TRITIUM monitor components to enhance it's sensitivity. All these studies were carried out inside a special light-tight box, called black box, to ensure that the photons we are detecting come from the sources used, either LEDs or scintillators. In addition, as the energy response of plastic scintillators has a rather large uncertainty, most of the energy spectra are shown in ADC<sup>1</sup> channels, which are linearly proportional to energy.

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<sup>1</sup>ADC units are the internal units, called channels, in which an analog signal is digitized after an Analog-to-Digital Converter. The number of available channels depends on the bits used in its digitization.

## 4.1 Characterization and R&D on Scintillating Fibers

This section reports experimental measurements of the scintillating fiber parameters that most affect the tritium detection, such as collection efficiency and conditioning uncertainty. Errors are included in all graphs displayed in this section but they are too small to be visible. Thousands of scintillating fibers are used in TRITIUM detector which were prepared and conditioned prior to characterization studies or tritium detection. Therefore, various mechanical and electronical devices were developed to automatically prepare many fibers at the same time.

### 4.1.1 Scintillating Fiber Conditioning Process.

The first step in TRITIUM design was to choose the optimal fiber length at which the signal of tritium events is optimal. On the one hand, long fibers are interesting because the efficiency of TRITIUM detector is proportional to the fiber length, but, on the other hand, in long fibers, scintillating photons are reflected on the fiber walls more times before reaching the photosensors, and, as some photons are lost in each reflection, the detector signal is deteriorated.

Several simulations were carried out using Geant4 [Col21], a particle and nuclear physics simulation package based on C++, to determine the optimal length. The result of these simulations, described in section 6.2.3, and displayed in section 8.1.3, Figure 8.7, was that it is preferable to work with short fibers.

The fiber length for the TRITIUM prototypes developed in Valencia, was 20 cm, which was also the length used for most of the experiments carried out in the framework of the TRITIUM experiment. As Saint-Gobain

commercial fibers are 1 meter long, an effective scintillator cutting technique had to be developed with strict requirements on the cutting quality of the fiber ends since this greatly affect the transmission of photons and, consequently, the efficiency of TRITIUM monitor. This cut must be perpendicular to the fiber and with very low uncertainty in the cutting position to achieve a good coupling of the fiber with the surface of the photosensor. It is also important that the fiber integrity be preserved, without cracks or deformations that contribute to the loss of photons.

Cutting the end faces of polymer fibers is one of current challenges. There are many different techniques such as milling, laser cutting, focused-ion-beam, blade cutting, etc. The blade cleaving technique was chosen for TRITIUM experiment because of its mechanical simplicity.

Many commercial devices based on blade cleaving, such as the one provided by thorlabs with a diamond tipped blade [Inc06] or others similar to guillotine designed for industrial fiber optics [fo], were tested in an extensive study with unsuccessful results [Cam17]. As it can be seen in Figures 4.1, Commercial techniques produce deformations, cracks and imperfections so they do not fulfill the requirements previously imposed.

The microscope model PB 4161 from EUROMEX and the Digital Microscope from Jiusion company were used to check the results in the fiber ends.

Because commercial devices do not work for our scintillating fibers, a cutting device, shown in Figure 4.2, was designed, built and tested.

This device consists of fourteen rails to which the fibers are hold and a thin blade, fixed on a mobile piece, which is used to cut them. The perpendicular cut, which is one of the requirements, can be ensured since the moving piece, to which the blades are fixed, is set perpendicular to the fibers. The blade used is a typical commercial razor blade, of 0.1 mm thickness, which is the thickness that gave the best results. The blade was

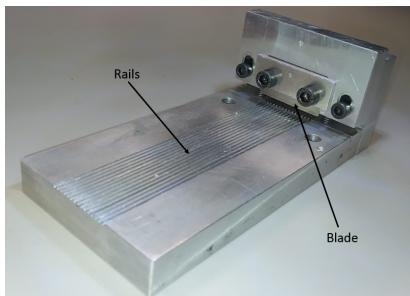


(a) Fiber end deformation.

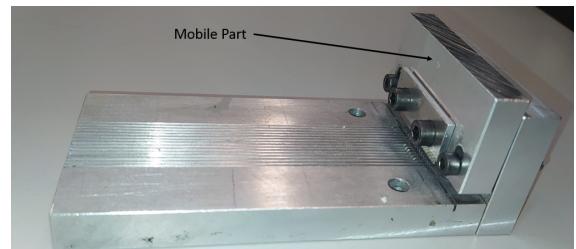


(b) Fiber end cracks.

**Figura 4.1** – Unsuccessful results of using commercial techniques to cut fibers.



(a) TRITIUM Cutting device.



(b) TRITIUM Cutting device.



(c) Additional piece of TRITIUM cutting device.

**Figura 4.2** – Cutting device developed in the TRITIUM experiment and additional part to make precise measurements of fiber length.

adjusted with 5° tilt, with respect to the horizontal axis since it was found in several studies that this helps to obtain a less aggressive and cleaner cut [SR15, Law06]. As it can be seen in Figure 4.3a, with this cutting device it was obtained fiber ends without breaks or deformation.

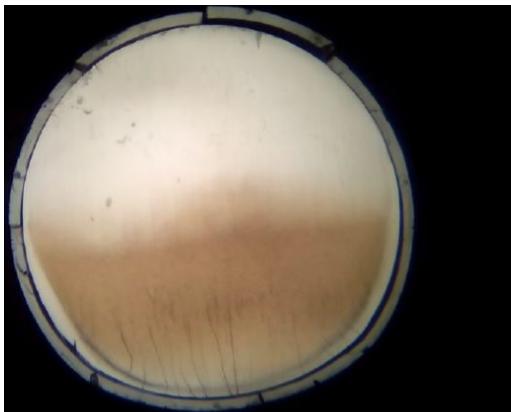
An additional parameter that could affect the cutting quality of the fiber ends is the temperature of both, the fiber and the blade. A study was carried out in which both were subjected to different temperatures from room temperature to 110 degrees. No significant conclusions were obtained [Cam17]. Thus, the cutting process was carried out at room temperature to make the cutting easier.

To set the fiber length, which is the last requirement, an additional piece was designed and built, shown in Figure 4.2c. With the help of this piece an uncertainty in the fiber length of less than 1 millimeter was achieved. With the designed cutting fiber device all requirements imposed have been fulfilled, obtaining fibers with optimal light transmission.

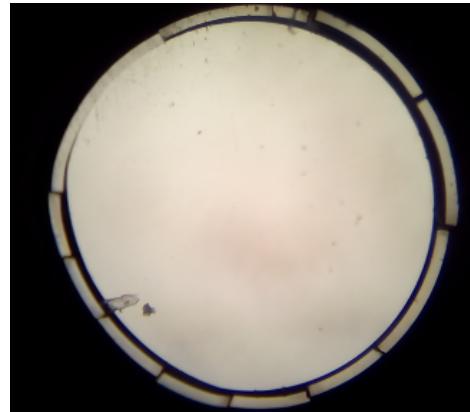
The fiber end after cutting is shown in Figure 4.3a. The slightly darkened zone at the bottom of the fiber is an inavoidable effect of the cutting process and, to reduce this flaw, a polishing process developed by Thorlabs was applied [Inc06].

Five different polishing papers, with a decreasing grain size, 30  $\mu\text{m}$ , 20  $\mu\text{m}$ , 12  $\mu\text{m}$ , 5  $\mu\text{m}$  and 0.3  $\mu\text{m}$  respectively, were rubbed during two minutes with the fiber describing on the the paper a shape of 8 (approximately 120 movements).

The result obtained after polishing is shown in Figure 4.3b, where it can be noted that the darkened zone has completely disappeared and the fiber end is completely clear without any damage or imperfection. Therefore, both tasks, cutting and polishing, are necessary.



(a) Fiber end after cutting with Tritium device.



(b) Fiber end after cutting and polishing.

**Figura 4.3** – Result of the polishing process. a) Fiber end after cutting b) Fiber end after cutting and polishing with Thorlabs technique.

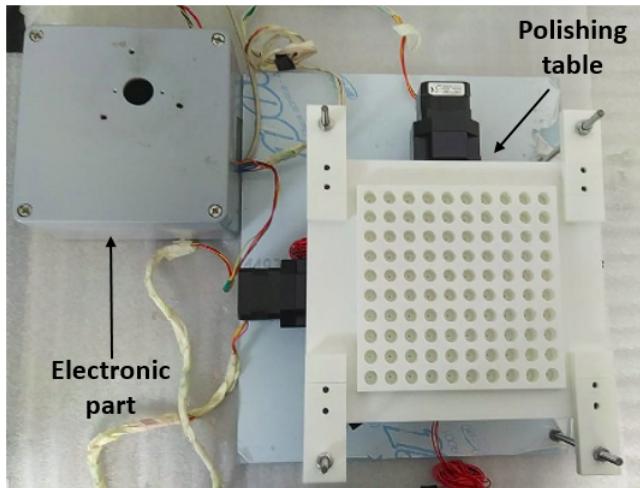
#### 4.1.2 Automatic Polishing Machine for Scintillating Fibers

As mentioned above, tens of thousands of fibers need to be prepared and conditioned for the TRITIUM monitor<sup>2</sup>, section 5.3. Although this amount of fibers was not a problem for cutting, which is very fast, the polishing process is quite time consuming. It takes more than ten minutes to polish each fiber, that would result in an unaffordable amount of time to prepare the needed amount of fibers. Therefore, an automatic polishing machine for scintillating fibers was designed, built and tested. This polishing machine is able to polish up to one hundred scintillating fibers at the same time and automatically. Furthermore, it is easily scalable to larger capacity.

This automatic polishing machine, displayed in Figure 4.4, consists of two parts: 1) A polishing table, where the fibers are polished 2) The electronics, based on Arduino technology, that operates the movement of the polishing table:

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<sup>2</sup>Tritium prototype will be a module of TRITIUM monitor, based on dozens of modules.



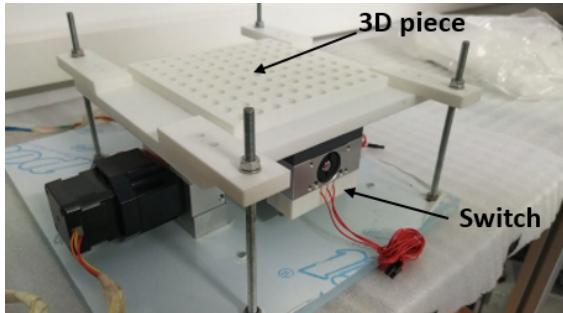
**Figura 4.4** – Polishing machine developed in TRITIUM experiment.

1. The polishing table, shown in Figure 4.5a, is divided in two parts: the static part, where the fibers are fixed, and the movable part, where the polishing papers are fixed. It was decided to set the polishing papers on the movable part because they are lighter and less fragile than fibers.

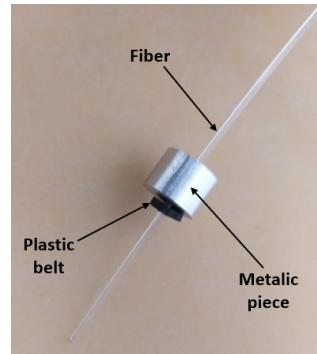
The static part consists of a piece, shown in Figure 4.5a, built with a 3D printer and fixed to the system by four vertical screws. On each screw there are two nuts to set the relative height and the inclination of fibers to the polishing papers. This piece contains one hundred holes in which the fibers are placed.

The own weight of fibers press the polishing paper. As they are too light (0.16 g), a plastic belt and a piece of metal with a weight of around 1.5 g, shown in Figure 4.5b, is employed to increase their weight (in the same way as in Thorlabs polishing process).

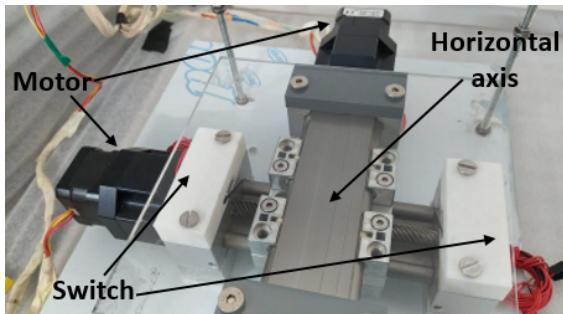
The movable part consists of a flat PMMA plate of  $18 \times 18 \text{ cm}^2$  to which the polishing paper is attached. This part is locked to two horizontal screws, perpendicular to each other that are used to set its position in the XY plane (horizontal plane), as shown in Figure 4.5c.



(a) Polishing table.



(b) Fiber with metal piece.



(c) Horizontal screws and PMMA plate.



(d) Piece to hold switches.

**Figura 4.5 – Polishing table of the polishing machine**

The polishing system contains several switches, mounted on a piece made by a 3D printer, shown in Figures 4.5a, 4.5c and 4.5d, which are used to find the origin of coordinates when the system is reinitiated and to stop the movable part when the end of the path is reached.

2. The electronics, shown in Figure 4.6, is based on Arduino technology which controls the automatic movement of the polishing paper.

The electronics consists of two stepper motors, model NEMA ST4209S1404-A [Nan], which move the horizontal screws on which the polishing paper is attached. These motors are controlled by an Arduino UNO [ARD] that uses a CNC shield [OSO17] in which two different drivers

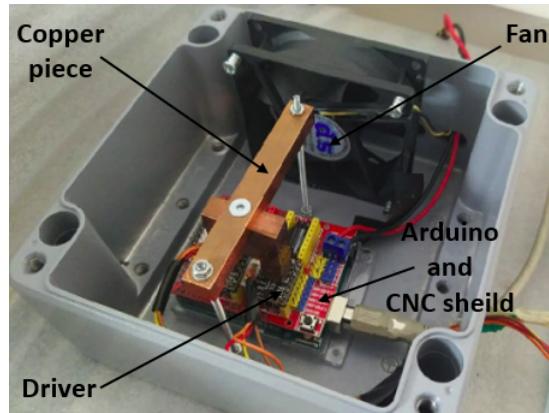
are connected to control the stepper motors, one driver for each stepper motor.

Drivers are controllers that allow to manage stepper motors in a simple way. It is very important to choose the correct controller for the system because the controller limits the supply power to the motors, avoiding burning of the motors in the worst case. Instead of using the Pololu A4988 drivers [All12], which is one of the most widely used drivers, the first choice was the DRV8825 driver [Ins14]. DRV8825 allows to power the motor with higher voltage and intensities (45 V and 2.5 A) than A4988 (35 V and 2 A). Also, the DRV8825 controller includes a new microstepping mode (1/32) compared to the A4988 (1/16) with which we get more accurate and smooth movements. Finally the drivers were replaced by the TMC2208 [Cir19], much less noisy since it includes the *StealthChop* function with which the noise is practically eliminated. Furthermore, this controller is much more accurate as it has a microstepping mode of 1/256. The voltage and current used to power the motors are 35 V and 2 A which is sufficient for the whole system since the current of the motors is limited to 1.33 A. The excess current will be transformed into heat that has to be dissipated from the system. Overheating of the drivers may cause loss of steps, producing different movements from those programmed or even destroying the driver. Therefore, a cooling system is needed to ensure the correct operation of the polishing system. The cooling system, shown in Figure 4.6, consists of a copper piece<sup>3</sup> in contact with both controllers and a fan, used to prevent heat accumulation inside the electronics box. The cooling power can be improved by using a PELTIER cell.

This polishing machine is controlled by a Raspberry Pi using the Universal G-code Sender software (a graphical interface based on the GRBL package). There are several useful pre-programmed functions such as "HOME"

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<sup>3</sup>The copper is one of the best thermal conductor at STP



**Figura 4.6** – Electronic system of Polishing machine.

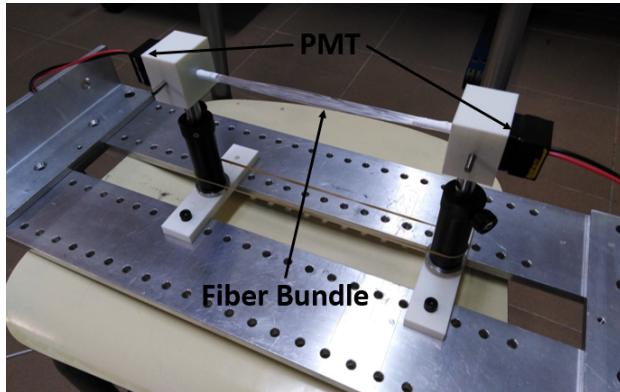
with which the system, using the switches, finds its origin coordinate every time the system is turned on. The software also has the possibility of loading a file containing the g-code to be executed, In the TRITIUM, the 120 movements required for each polishing paper are loaded in this way.

This machine was tested with twenty fibers of 15 cm length arranged in a bundle. The fibers were fixed to the structure shown in Figure 4.7 and two PMTs located at the bundle ends, read in coincidence as described in section 3.2.4, Figure 3.14b, monitored the light as transmission of the fibers.

Two different measurements were taken using two radioactive sources, a  $^{60}\text{Co}$  gamma source of 715 Bq activity, and a  $^{90}\text{Sr}$  beta source of 17.8 kBq activity. After that, the fiber bundle was polished and the test was repeated.

The energy spectra recorded for both radioactive source are exhibited in Figure 4.8. The sources were placed in the middle of the fiber bundle, at 7.5 cm from each PMT.

As it can be seen in Figure 4.8, both energy spectra are shifted to the right after polishing fibers, which means that the detected events have



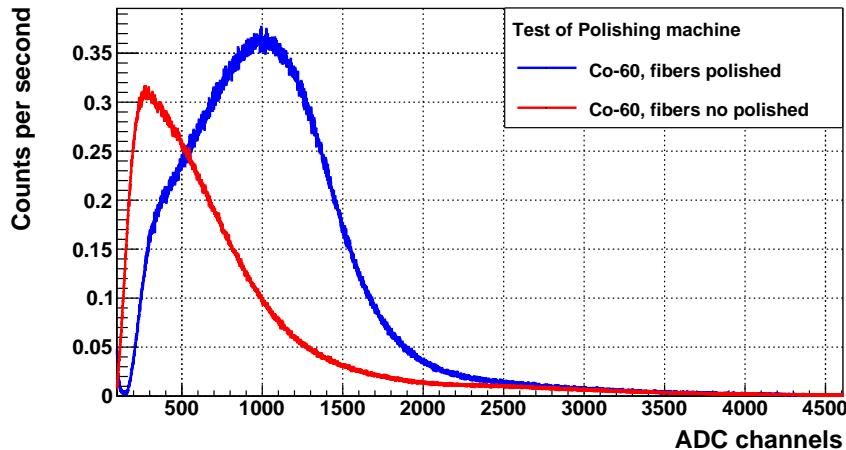
**Figura 4.7** – Set up used to test the effect of the polishing machine.

more energy (more photons per event reach the PMTs). This energy increase was more than 40% ( $(42 \pm 4.6)\%$  for gamma source and  $(49 \pm 8.4)\%$  for beta source) with respect the unpolished fiber. In summary, with the polishing machine, the photon collection efficiency of the fibers was improved (mainly due to the improvement of the interface between fibers and PMTs). It is very important to achieve a high detection efficiency as the expected number of photons per tritium event is quite low.

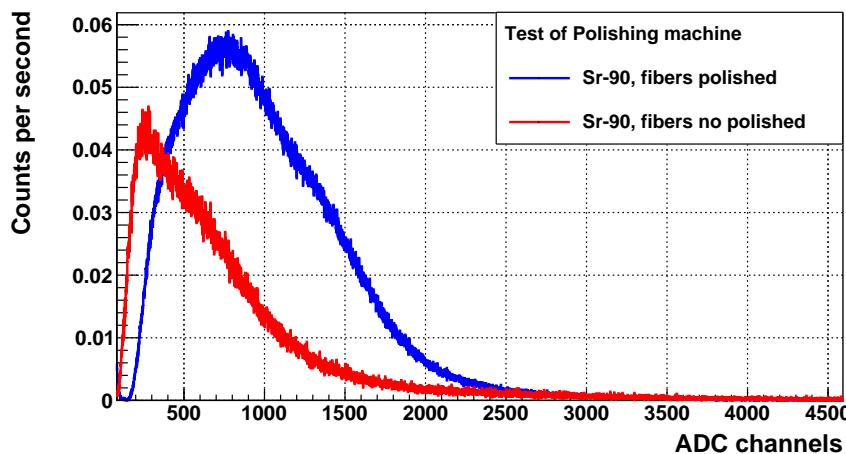
### 4.1.3 Characterization of Scintillating Fibers

This section describes the characterization of unclad BCF-12 fibers, called no-clad fibers, from Saint-Gobain, which are the fibers selected for the TRITIUM experiment. These fibers are compared to single clad and multiclad BCF-12 fibers to quantify the influence of the clad in the relevant parameters of the scintillating fibers.

Although commercial clads are too thick for the TRITIUM experiment, the necessary low thickness clads could be developed. For example, clads with a thickness of the order of tens of nanometers can be achieved by electrodeposition techniques.



(a) Energy spectrum recorded for the Co-60 source.



(b) Energy spectrum recorded for the Sr-90 source.

**Figura 4.8** – Energy spectrums used to test the effect of the Polishing machine

The difference between these three types of fibers is that uncladded fibers only consist of a polystyrene core with a refractive index of 1.60 whereas, single clad fibers, have an acrylic clad (PMMA) of 30  $\mu\text{m}$  thickness and a refractive index of 1.49 and, multiclad fibers, have a second fluor-acrylic clad of 10  $\mu\text{m}$  thickness and a refractive index of 1.42.

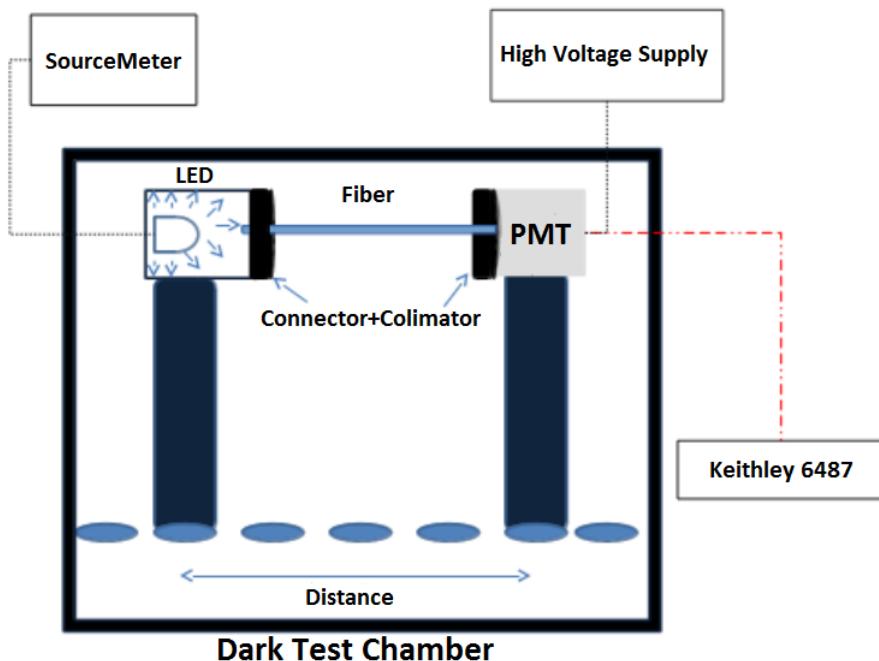
This characterization was carried out at the level of a single scintillating fiber. The parameters measured for each fiber type were the fiber collection efficiency and the uncertainty of the fiber response due to the conditioning process. The reference magnitude employed for the characterization is the rate of photons that reach the active area of the photo-sensor. To measure this magnitude, a calibrated R8520-06SEL PMT was used, whose quantum efficiency at the working wavelength, 29.76%, was measured by Hamamatsu. The PCB described in section 3.2.4 was used for working without the PMT internal gain and the PMT output current was measured by a Keithley 6487 Picoammeter/Voltage Source. The photons rate was obtained from the current measurement using the equation 3.13 with  $QE = 0.2976$  and  $CE = 1$ . A simplified scheme of the used set up is shown in Figure 4.9.

This setup consists of an optical structure in which a LED and a PMT are fixed to a user set distance between them. A LED435-03 from Roithner LaserTechnik GmbH [Gmb10], simulated the light emission by the fibers. The emission spectrum of the LED, given in Figure 4.10, was experimentaly measured by a spectrometer and fitted to a Gaussian function. The LED emission peak is at 433.9 nm with a FWHM of 18.4 nm.

The fiber was fixed between the LED and the PMT. The length of the fiber was 20 cm. Optical grease [Cera] was used for optimal coupling between the fiber and the PMT. Two collimators were used to ensure that only photons detected from the LED were detected in the PMT. Two FH-ST<sup>4</sup> connectors from RoHS company [], were used to fix the fiber to the

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<sup>4</sup>FH-ST is a quick assembly connector for 1 mm POF, Plastic Optical Fiber



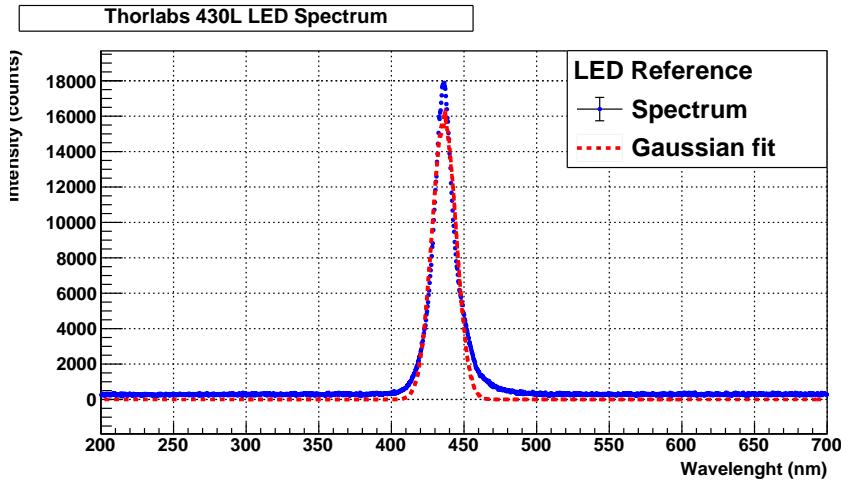
**Figura 4.9** – Set up used for fiber characterization.

system.

### Preparation of the Characterization System.

Before characterizing a fiber several tasks had to be performed to check that the black box was light-tight enough and the PMT response was lineal.

A light leak in the black box would produce a background larger than the signal. To check the light-tightness a no-clad fiber of 20 cm length was fixed in the previous assembly. The LED was fed with four different intensities (0.05 mA, 0.1 mA, 0.15 mA and 0.2 mA) and the PMT response was measured with and without a special black blanket from Thorlabs [Thoa], that prevents external photons to reach the system.



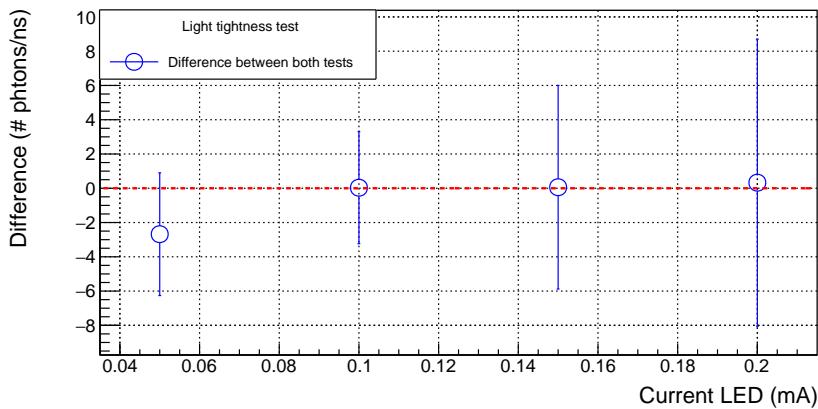
**Figura 4.10** – Emission spectrum measured for the LED model 435-03 from Roithner LaserTechnik GmbH Company.

This test was repeated for three different fibers and the mean and standard deviation of the light output were calculated by:

$$\bar{x} = \frac{\sum_{i=0}^N x_i}{N}; \quad \sigma = \sqrt{\frac{\sum_{i=0}^N (x_i - \bar{x})^2}{N-1}}; \quad (4.1)$$

The difference of the PMT responses in both cases is presented in Figure 4.11. As it can be seen in this figure, there are no statistically relevant differences between covered and uncovered fiber. Therefore, the light tightness of the black box is sufficient for this study.

The optimal voltage of the PCB without the PMT internal gain was obtained by finding the voltage plateau in which the electron collection efficiency in the first dynode was practically 100%. The LED was fed at 1 mA intensity without any fiber and the PMT output current was measured for different PMT supply voltages, between 0 and 500 V. The number of photons detected by the PMT is plotted in Figure 4.12. As it can be seen, the plateau is located at voltages higher than 150 V, where the PMT output



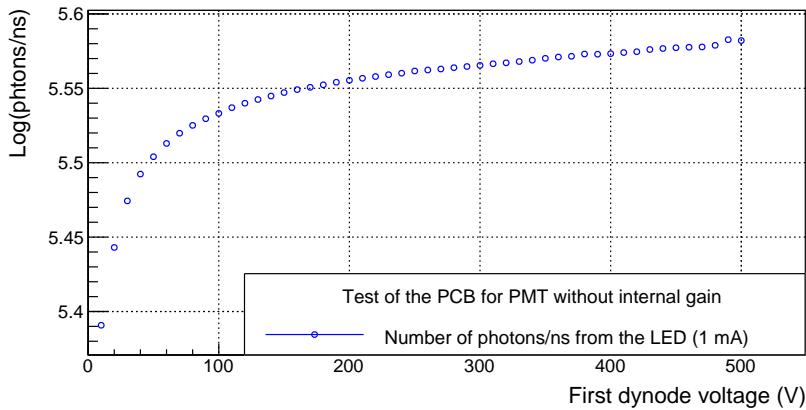
**Figura 4.11** – Difference between the results obtained in both tests carried out to check the light-tight quality of the system.

response is stable. The chosen voltage at which the characterization was carried out was 250 V.

Finally, the linearity of the PMT was verified. The LED was powered with intensities ranging from 0 to 10 mA (LED linearity range) to check that the LED emission light do not saturate. The linearity was tested in the range of the number of photons expected in a tritium event (a few tens of photons per tritium event, tens of photons per nanosecond) and in the range of up to two thousand five hundred photons per nanosecond.

To test the linearity of the PMT in the range of tritium events, the set up described above was used without any fiber and one of the connectors but the collimators was kept to make sure that the active area of the PMT is the same as the use in the characterization study.

To test the linearity of the PMT at the level of more than a thousand photons per nanosecond, the remaining connector was removed in order to increase the photons that reach the photosensor and the collimators was also kept.



**Figura 4.12** – Response of the PMT as a function of its high voltage using the designed PCB with which no internal gain of the PMT is obtained.

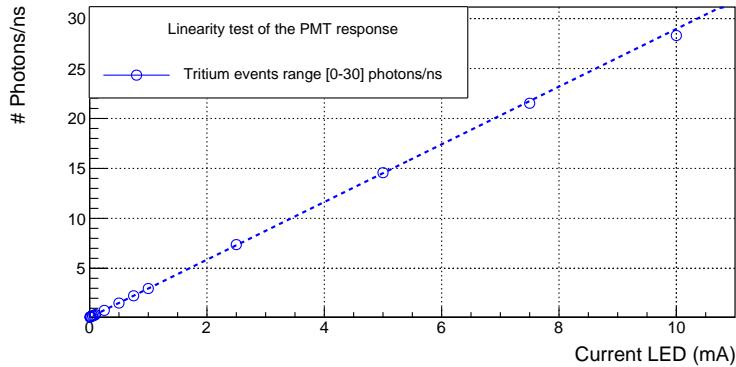
The results for both intensity ranges are shown in Figures 4.13. As it can be seen, the PMT output current is linear in both intensity ranges.

## Results of the Characterization of Scintillating Fibers

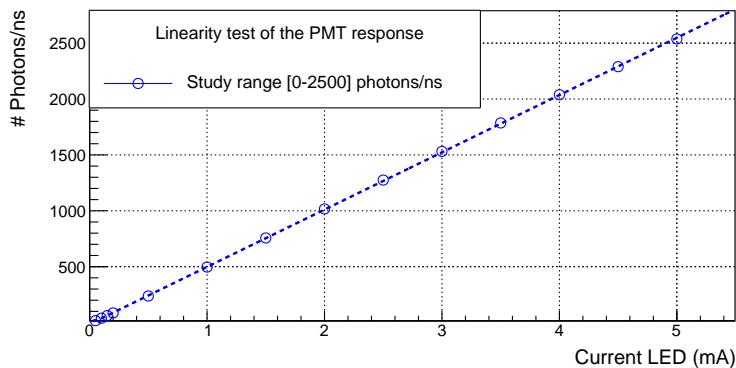
The conditioning process, consisting of cutting and polishing the fibers, is an individual task that generates a small dispersion in the response of each individual fiber,  $\sigma_{con}$ . This is an uncertainty that will be present in the tritium measurement of the monitor.

To measure this dispersion, it has to be taken into account that the position of the connectors that lock the fiber in the experimental setup produce an additional uncertainty,  $\sigma_{pos}$ , in the measurement. Since both uncertainties are independent, the total uncertainty is given by:

$$\sigma_t = \sqrt{\sigma_{pos}^2 + \sigma_{con}^2} \quad (4.2)$$



- (a) Response of the PMT in the intensity range of tritium events. Error bars are smaller than the point size.



- (b) Response of the PMT in the range  $[0 - 2500]$   $\gamma/\text{ns}$ . Error bars are smaller than the point size.

**Figura 4.13** – Linearity tests of the PMT response

The uncertainty due to the fiber position has to be quantified to extract the conditioning uncertainty from the total uncertainty. Two different experiments were designed, the first with only the uncertainty in the fiber position ( $\sigma_t = \sigma_{pos}$ ), and the second with both uncertainties. The conditioning uncertainty is given by:

$$\sigma_{con} = \sqrt{\sigma_{tot}^2 - \sigma_{pos}^2} \quad (4.3)$$

The test designed to measure  $\sigma_{pos}$  consisted of preparing one fiber of each type (uncladded, single clad and multiclaid) by the conditioning process reported above. Each fiber was locked in the set up, and a measurement by feeding the LED with an intensity of 0.1 mA was done. These measurements were repeated ten times with the same fiber. Ten different measurements for each fiber type was obtained. The standard deviation of the measurements is only due to the position uncertainty. The results are shown in Table 4.1.

$$\sigma_{pos}^{rel} = \frac{\sigma_{pos}}{\bar{x}} \quad (4.4)$$

| Fiber type  | Mean ( $\gamma/\text{ns}$ ) | $\sigma_{pos}$ ( $\gamma/\text{ns}$ ) | $\sigma_{pos}^{rel}$ (%) |
|-------------|-----------------------------|---------------------------------------|--------------------------|
| Uncladded   | $524.088 \pm 0.010$         | 17.65                                 | 3.37                     |
| Single Clad | $1071.696 \pm 0.01$         | 9.07                                  | 0.85                     |
| Multiclad   | $949.930 \pm 0.026$         | 9.91                                  | 1.04                     |

Table 4.1: Mean and standard deviation (due to fiber position in the setup) of photons per nanosecond that reach the PMT for 0.1 mA LED intensity.

As it can be seen, the clad reduces the position uncertainty, which means that it improves the uniformity of the fiber response. It was also found that the clad markedly improves the light collection efficiency of the fibers. The reason could be because photons are mainly collected in the

core of the fiber and the interface of core is better defined in the case of a single clad or multiclad fibers than for uncladded fibers. In the latter case, the interface is provided by the environment (air or water in the case of TRITIUM), and external conditions, as dirt, may produce noticeable interface fluctuations.

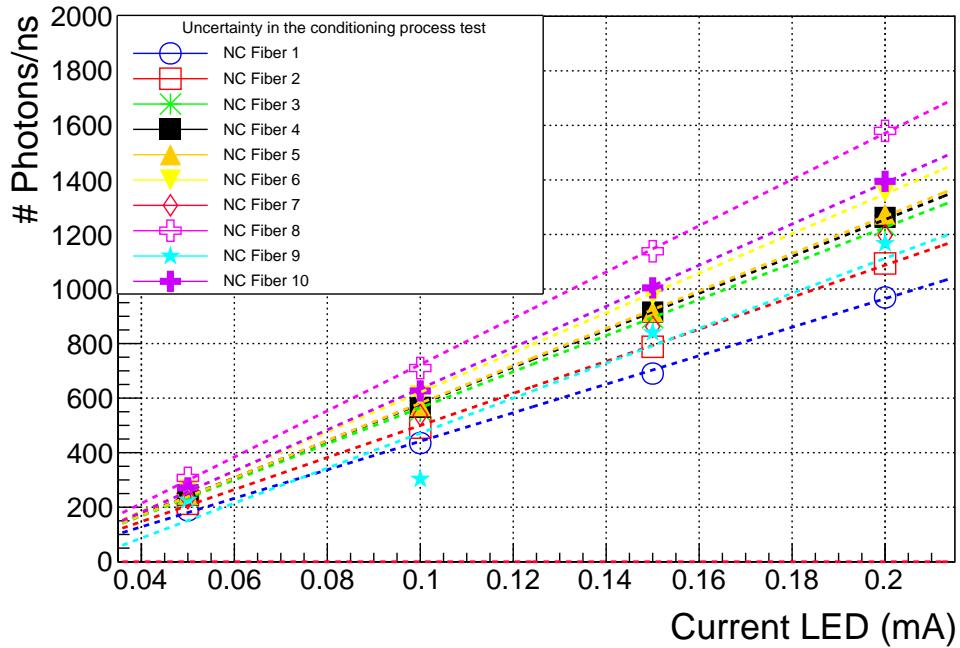
It is also seen in the table that a second clad slightly reduce the collection efficiency. The reason could be that a second clad layer reduces the fiber core proportionally.

Concerning the error of the measurement, the error of the Keithley device was three orders of magnitude smaller than the standard deviation, and it was not taken into account.

To determinate the whole uncertainty, ten different samples of each fiber type were prepared and each fiber was measured under the same conditions as the previous test. This measurement was done for four different LED emission intensities (0.05, 0.1, 0.15 and 0.2 mA) to reduce possible mistakes. The results of uncladded fibers are plotted in Figure 4.14, where it can be seen that, although each fiber shows a very linear trend with the amount of collected photons, a dispersion in the fiber response is clearly seen. Similar results were obtained for single clad and multiclad fibers, displayed in figures 4.15a and 4.15b respectively.

The average number of collected photons versus LED intensity and the relative standard deviation for each type of fiber is given in Tables 4.2 and 4.3 respectively and represented in Figure 4.16, where they can be compared.

As it can be noticed in Figures 4.14 and 4.15 the fiber response is quite linear and that single clad and multiclad fibers have stronger signals than uncladded fibers, which means that the clad has a significant effect on the fiber collection efficiency. Also, it was found that single-clad fibers have higher collection efficiency than multiclad fibers. It can also be remarked in

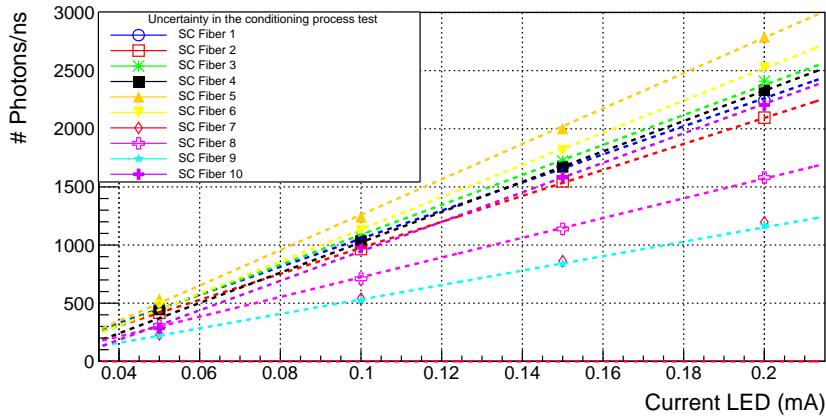


**Figura 4.14** – Number of photons/ns reaching the PMT for Uncladded fibers.

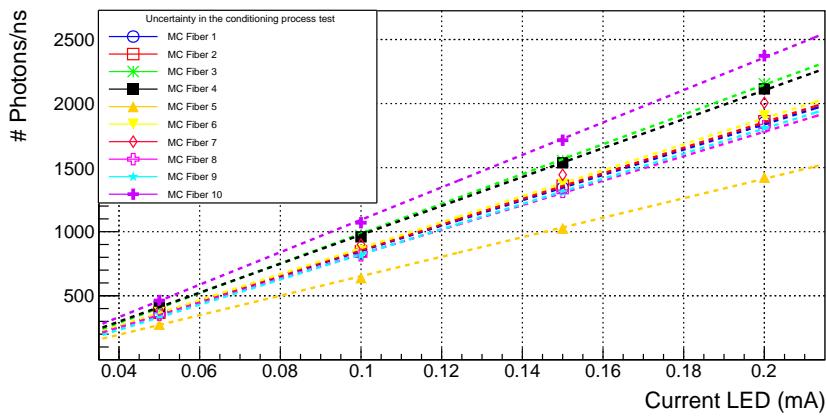
Table 4.3 that the relative standard deviation,  $\sigma_{pos}^{rel}$ , does not vary with the LED intensity.

An average of the uncertainty due to the fiber position and the conditioning process are given in Table 4.4.

As it can be seen, the smallest uncertainty in the conditioning process was found in the uncladded fibers, which means that the damage from this process occurs mainly in the fiber clad, as can be checked in Figure 4.3. It was checked under the microscope that this damage only occurs at the end of the fiber. Also, the largest relative standard deviation in this process is measured for single clad fibers, which means that the second clad



(a) Number of photons/ns reaching the PMT for Single Clad fibers.



(b) Number of photons/ns reaching the PMT for MultiClad fibers.

**Figura 4.15** – Number of photons/ns reaching the PMT for ten samples of each fibers type.

| Led Int. (mA) | Uncladded     | Single Clad    | MultiClad     |
|---------------|---------------|----------------|---------------|
| 0.05          | $245 \pm 11$  | $384 \pm 33$   | $377 \pm 15$  |
| 0.1           | $572 \pm 26$  | $923 \pm 74$   | $871 \pm 35$  |
| 0.15          | $915 \pm 39$  | $1485 \pm 120$ | $1397 \pm 55$ |
| 0.2           | $1267 \pm 55$ | $2054 \pm 166$ | $1933 \pm 76$ |

Table 4.2: Number of the collected photons versus LED intensity for the different type of fibers.

| Led Int. (mA) | Uncladded | Single Clad | MultiClad |
|---------------|-----------|-------------|-----------|
| 0.05          | 4.38      | 8.66        | 3.97      |
| 0.1           | 4.59      | 8.02        | 3.97      |
| 0.15          | 4.34      | 8.07        | 3.95      |
| 0.2           | 4.36      | 8.10        | 3.93      |
| Mean          | 4.42      | 8.21        | 3.96      |

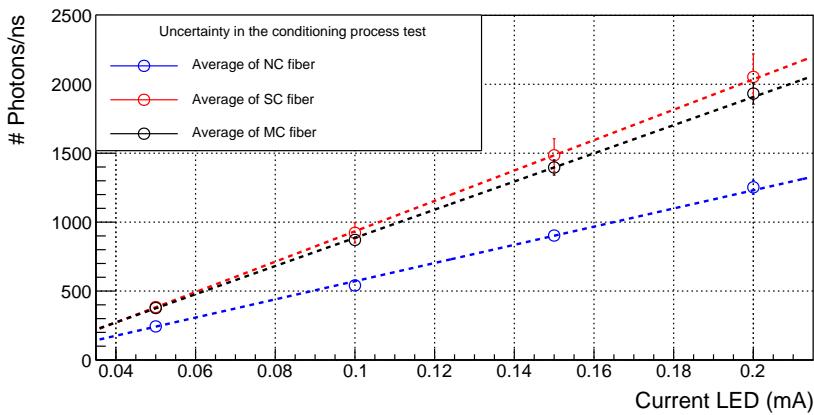
Table 4.3: Relative standard deviation versus LED intensity for the different fiber types.

increases the resistance of the fiber to the conditioning process.

In summary, this study shows that the use of fiber clad improves the photon collection efficiency. The relative statistical deviation due to the fiber conditioning process was quantified for the different fiber types. It was found that the damage of the conditioning process is produced mainly in the fiber clad. Thus, if a method to build a clad for fibers is developed, it should be applied after the fiber conditioning process.

Finally, the measurement of the photon collection efficiency of each type of fiber is shown. The collection efficiency is the percentage of photons collected along the fibers. It is usually given by 1 m fiber length,  $CE_{100}$ .

To measure the collection efficiency,  $CE_{100}$ , ten different samples 10 cm length was prepared for each fiber type. The results are given in



**Figura 4.16** – Average of 10 samples for each fiber type (uncladded, single clad and multiclad fibers).

| Fiber type  | $\sigma_t$ (%) | $\sigma_{pos}$ (%) | $\sigma_{con}$ (%) |
|-------------|----------------|--------------------|--------------------|
| Uncladded   | 4.42           | 3.37               | 2.86               |
| Single Clad | 8.21           | 2.17               | 7.92               |
| Multiclad   | 3.96           | 1.04               | 3.82               |

Table 4.4: Relative standard deviations ( $\sigma_t$ ,  $\sigma_{pos}$  and  $\sigma_{con}$ ) measured in this test.

Table 4.5.

The collection efficiency in 10 cm fiber length,  $CE_{10}$ , was calculated by comparing these tests with those performed for a fiber length of 20 cm and the collection efficiency,  $CE_{100}$ , was calculated from  $CE_{10}$  by assuming a linear dependence on length.

The collection efficiency,  $CE_{100}$ , given by the manufacturer Saint-Gobain is between 7% and 3.44% [Cer05]. As collimated photons were used in this study, the fact that our results are in the best side is justified. As it can be seen in Table 4.6, our measured values are very close to those

| Led Int. (mA) | Uncladded ( $\gamma/\text{ns}$ ) | Single clad ( $\gamma/\text{ns}$ ) | MultiClad ( $\gamma/\text{ns}$ ) |
|---------------|----------------------------------|------------------------------------|----------------------------------|
| 0.05          | $318 \pm 61$                     | $550 \pm 71$                       | $480 \pm 84$                     |
| 0.1           | $736 \pm 143$                    | $1270 \pm 164$                     | $1111 \pm 193$                   |
| 0.15          | $1184 \pm 232$                   | $1984 \pm 231$                     | $1777 \pm 307$                   |
| 0.2           | $1645 \pm 324$                   | $2507 \pm 208$                     | $2338 \pm 350$                   |

Table 4.5: Number of the collected photons versus LED intesntiy for 10 different fibers of 10 cm length.

| Fiber type  | $CE_{10}$ (%) | $CE_{100}$ (%) |
|-------------|---------------|----------------|
| UnCladded   | $76 \pm 8$    | $7.6 \pm 0.8$  |
| Single Clad | $78 \pm 6$    | $7.8 \pm 0.6$  |
| Multiclad   | $83 \pm 7$    | $8.3 \pm 0.7$  |

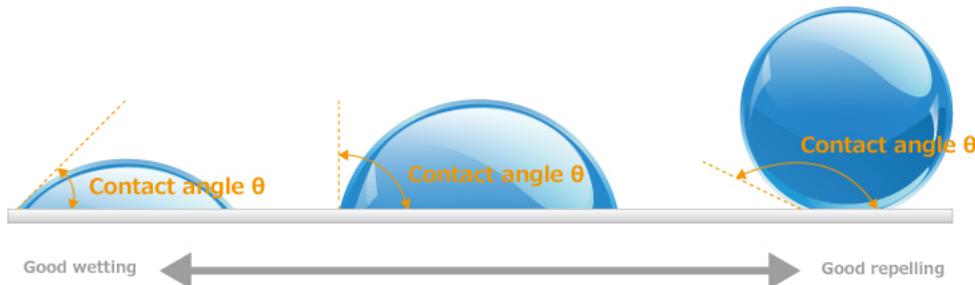
Table 4.6: Collection efficiencies  $CE_{10}$  and  $CE_{100}$ .

provided by the manufacturer.

#### 4.1.4 Cleaning Process for Scintillating Fibers

The tritium events detected in the fibers produce a few photons, so it is very important to conserve as many photons as possible. As it was shown in the fiber characterization study, the quality of the interface between the core of uncladded fibers and the environment (tritiated water in the case of TRITIUM detector) conspicuously affects the photon collection efficiency. To improve the quality of the interface a fiber cleaning process was included, aiming to remove external particles deposited on the fibers, such as dust and fat that worsen the photon collection efficiency. Through this cleaning process, the wetting property of the fibers, illustrated in Figure 4.17, is improved, preventing air molecules from attaching to the fiber and achieving a uniform water clad around the fibers, which results in an improvement of

their collection efficiency.



**Figura 4.17** – Wetting property produced by the cleaning process. [comb]

This cleaning process was developed and carried out in the clean room of ICMOL laboratory<sup>5</sup>. It consists of filling three different glass beakers, one with alkaline soap, another with millipore water<sup>6</sup> and the last one with isopropanol. First, the fibers are rubbed for 5 minutes with alkaline soap and then placed in the first beaker for sonication for 3 minutes. Then, the fibers are cleaned with a constant flow of water for 5 minutes. Second, the fibers are placed in the second beaker for sonication for another 3 minutes. Third, the fibers are placed in the third beaker for sonication for another 3 minutes. Finally the fibers are dried with an N<sub>2</sub> air gun and introduced inside of the prototype.

The improvement in fiber response was verified using a bundle of twenty fibers of 15 cm length that was prepared with the conditioning process described. This bundle of fibers was arranged in the setup described in section 4.1.2, Figure 4.7, and several energy spectra were taken using different radioactive sources. Then, these fibers were cleaned with the fiber cleaning process and these measurement was repeated in the same conditions.

<sup>5</sup>ICMOL, Institute of Molecular Science, is a research institute located in the Science Park of the University of Valencia.

<sup>6</sup>The millipore water is water in which all the ions were removed, producing a very low conductivity of it-self, on the order of 10  $\mu\text{Sv}/\text{cm}^2$

Two radioactive sources were used in this study, the  $^{90}\text{Sr}$  beta source, used in the polishing machine test, and the  $^{137}\text{Cs}$  gamma source, of 500 Bq activity. The results are shown in Figures 4.18, where a shift of the spectrum to higher energies can be noticed.

This improvement was estimated by:

$$F = \frac{A_C - A_{NC}}{A_C} \quad (4.5)$$

where  $A_C$  is the integral of the energy spectrum measured after the cleaning process and  $A_{NC}$  is the integral of the energy spectrum measured before the cleaning process.

The F obtained is about 21% for both radioactive sources. Nevertheless, it should be taken into account that this test was carried out in air which may give somewhat different results than in water.

## 4.2 Characterization and R&D on SiPM

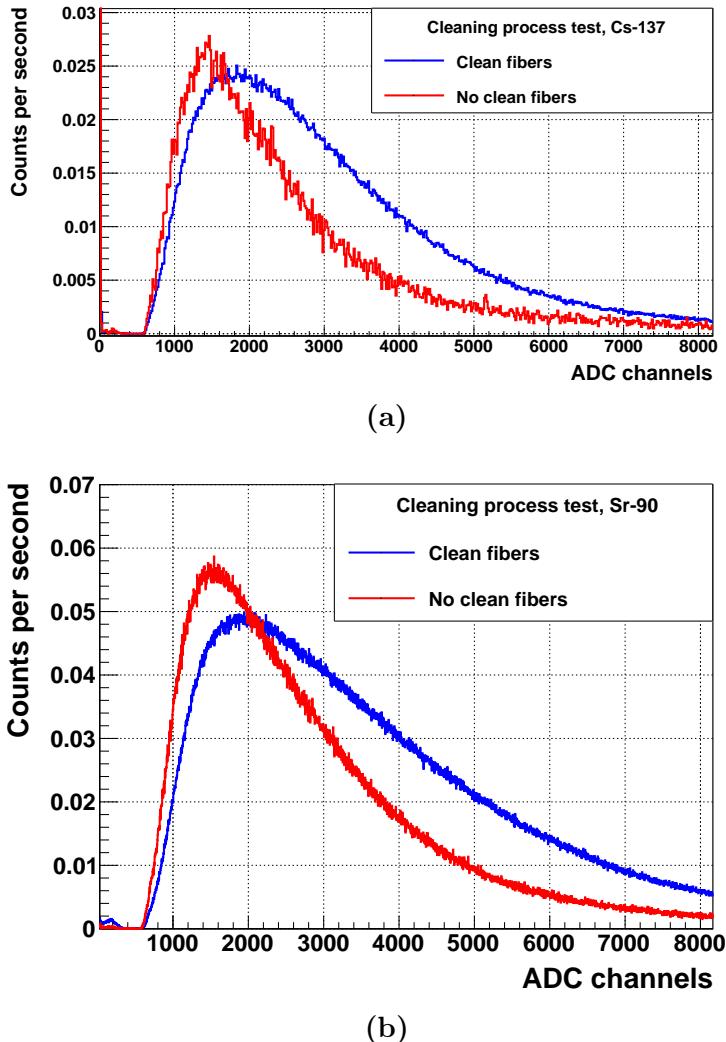
## 4.3 Characterization of the Ultrapure Water System

This section describes the characterization of the ultrapure water system, employed to ensure that the quality of the water sample used to be measured fulfills the requirements of the TRITIUM detector. There are three different requirements that this ultrapure water system must satisfy:

1. A quite low conductivity<sup>7</sup> of the water, around 10  $\mu\text{Sv}/\text{cm}$ , to avoid

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<sup>7</sup>Conductivity is the ability of a material to conduct electrical current. In liquids, conductivity is related to the presence of salts (presence of positive and negative ions)



**Figura 4.18** – Energy spectra obtained before and after the cleaning process using a radioactive source of a)  $^{137}\text{Cs}$  and b)  $^{90}\text{Sr}$

that external particles dissolved in the water be deposited on the fibers, drastically reducing the detector efficiency.

2. The radioactive particles (other than tritium isotope) from the water sample should be removed because tritium cannot be separated from other radioactive isotopes.
3. Lastly, the tritium activity should not be affected by the water purification process.

To verify that these requirements are complied with, a characterization of the water sample for both, raw water and purified water, was done. This characterization consisted of measuring the water sample conductivity and the activity of the different radioactive element present in the sample. The turbidity and the chemical components of the water sample were also measured. The sample of the raw water was taken at 40 meters from the ultrapure water system and two meters deep in the river since it is the place where the samples used in TRITIUM monitor will be taken. Variations of up to 25% in the tritium activity was measured between both points (due to the diffusion of tritium along the river).

The chemical composition of the water was measured before the ultra-purification process by a physico-chemical analysis, shown in Table 4.7.

The water sample contains a number of components given in the Table, that must be removed to prevent their deposition on the scintillating fibers of the detector, that would reduce their sensitivity to tritium activity.

The water turbidity<sup>8</sup> was measured using the Hanna Hi 9829 portable multiparameter system from Hanna Instruments [Ins], obtaining a value of

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<sup>8</sup>The turbidity of water is the loss of transparency due to dissolved particles, normally measured in Nephelometric Units of Turbidity, NTU, as the intensity of scattered light at 90 degrees.

| Chemical components     | Concentration (mg/L) |
|-------------------------|----------------------|
| $\text{CO}_3\text{H}^-$ | 154                  |
| Mg                      | 46                   |
| Ca                      | 105                  |
| $\text{NO}_3^-$         | 16                   |
| $\text{Cl}^-$           | 196                  |
| $\text{NO}_2^-$         | 0.03                 |
| K                       | 11                   |
| Na                      | 173                  |
| $\text{SO}_4^-$         | 217                  |
| Dry Residue             | 1029                 |

Table 4.7: Chemical components and turbidity measured in the raw water sample.

29 NTU, much higher than the WHO recommended limit of 5 NTU for drinking water.

The water conductivity was measured for both, raw and purified water, using the same system, the Hanna Hi 9829. The results of these measurements, together with the measurement of the conductivity of rejected water, described in section 3.3.2, are presented in Table 4.8.

| Date    | Raw ( $\mu\text{Sv}/\text{cm}$ ) | Pure ( $\mu\text{Sv}/\text{cm}$ ) | Reject ( $\mu\text{Sv}/\text{cm}$ ) |
|---------|----------------------------------|-----------------------------------|-------------------------------------|
| 1/8/18  | 970                              | 11.85                             | 1442                                |
| 7/8/18  | 958                              | 11.8                              | 1632                                |
| 14/8/18 | 966                              | 12.04                             | 1725                                |
| 22/8/18 | 980                              | 12.54                             | 1702                                |
| 28/8/18 | 987                              | 9.9                               | 1692                                |
| 5/9/18  | 1009                             | 12.02                             | 1645                                |

Table 4.8: Measurements of the conductivity for several samples of water.

As it can be seen in the first column, the raw water sample has

high values of conductivity, due to its content of ions, shown in Table 4.7. It can be noticed in the second column of the table that the conductivity of pure water was reduced by almost two orders of magnitude, to values close to  $10 \mu\text{Sv}/\text{cm}$ , fulfilling the requirement. In the third column, it can be remarked that the rejected water conductivity is higher than that of raw water, because this water contains the removed ions from pure water.

The gamma radioactive elements present in both, raw water and purified water, were identified and their activities measured by a HPGe, high purity germanium detector. A gamma analysis was carried out to determine the emitters with long enough lifetime to be measured. The radioactive isotopes found in the raw water sample with measurable activities were  $^{40}\text{K}$  and  $^{226}\text{Ra}$  which were completely removed in the purified water.

The tritium activity was measured by liquid scintillation counting (LSC) to find its modification by the ultra-purification process. The raw water was filtered at 0.45 microns to remove any particles that could cause the extinction of the scintillation signal. Table 4.9 show several measurements of the tritium activity for different water samples before and after purification. As it is seen, tritium activity is not affected by the purification process.

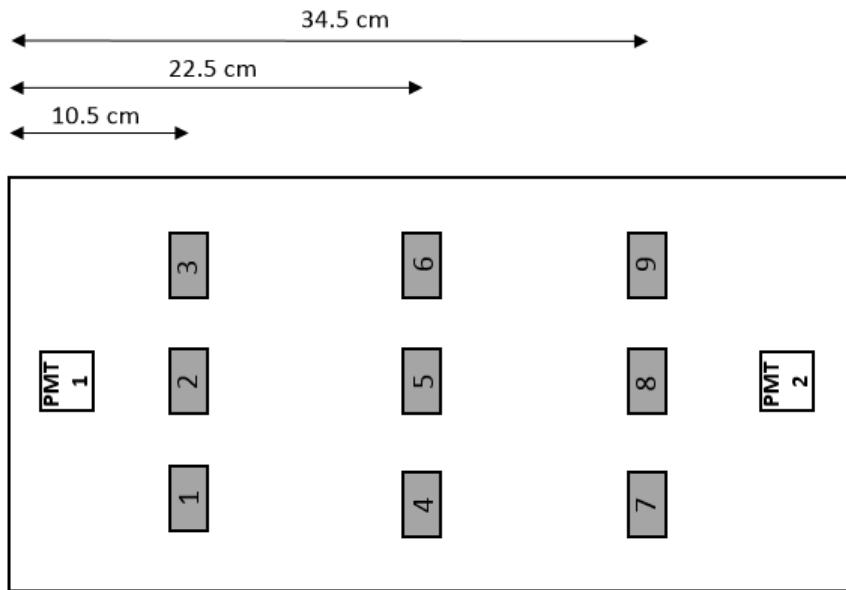
| Date     | Raw (Bq/L)     | Pure (Bq/L)     |
|----------|----------------|-----------------|
| 7/8/18   | $24 \pm 3$     | $26 \pm 4$      |
| 11/12/19 | $13.2 \pm 2.1$ | $13.85 \pm 2.2$ |
| 15/01/20 | $30.6 \pm 4.2$ | $30 \pm 4$      |

Table 4.9: Measurements of the tritium activity for several samples of both, raw and pure water.

## 4.4 Characterization of the TRITIUM Cosmic Veto

This last section reports on the characterization of the active shield (cosmic veto), which was carried out using PMTs as photosensors. Measurements of the cosmic veto using SiPM arrays has already started and their replacement will be as soon as possible.

The veto coverage, shown in Figure 3.26, was verified. This study was done at the level of one detector so the configuration of the electronics is the one shown in Figure 3.14b. The surface of the veto was divided in 9 parts, shown in Figure 4.19, in which a gamma source was placed.

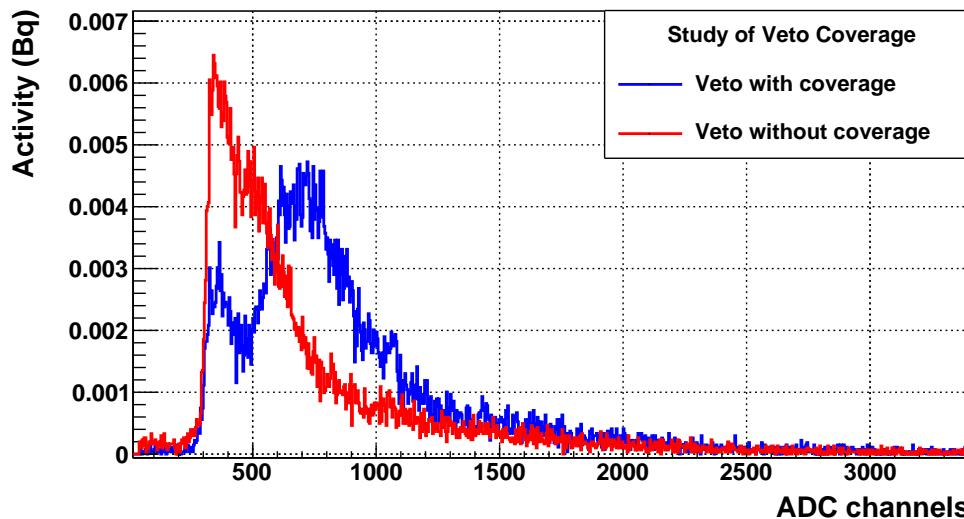


**Figura 4.19** – Reference points used for veto mapping.

Two different tests were made for this task:

1. The first test was used to quantify the improvement of the veto signal

due to its coverage. A  $^{137}\text{Cs}$  source was placed at point 2 and a energy spectrum was measured with the veto uncovered. Next, the measurement was repeated with the veto covered. The result is shown in Figure 4.20.

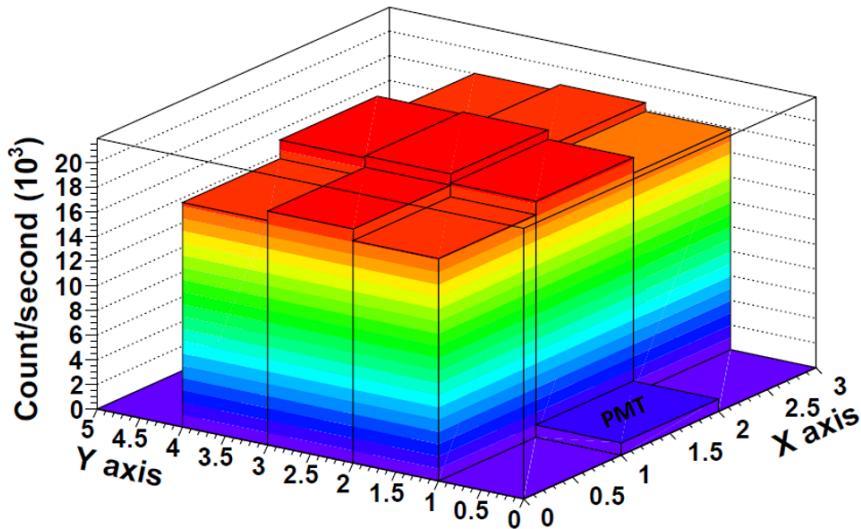


**Figura 4.20** – Measurement of a radioactive source  $^{137}\text{Cs}$  with the TRI-TIUM cosmic detector with and without its coverage.

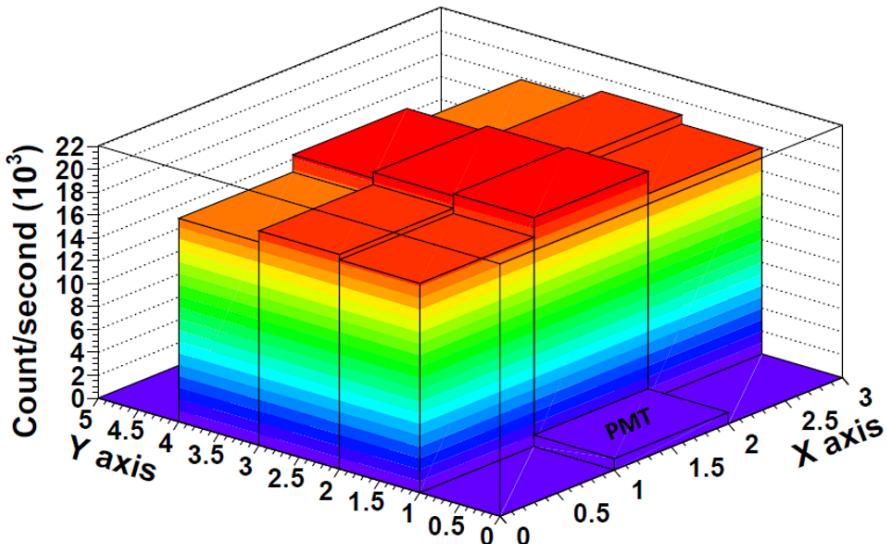
The spectrum is shifted to the right, which means that more photons have been collected per event. No improvement was obtained in the number of events detected, only in the collection efficiency.

2. The second test was done to verify the spatial uniformity of the signal in the covered veto. For this task, a mapping was carried out, which consisted of placing a  $^{60}\text{Co}$  source at each point and measuring the number of events detected in the same time window. This test was done for two different veto modules and the energy spectrum obtained was integrated. The count rates obtained are plotted in Figures 4.21a and 4.21b, respectively.

It can be observed that the veto signal has a uniform response on its



(a) Mapping of the first TRITIUM cosmic detector.



(b) Mapping of the second TRITIUM cosmic detector.

**Figura 4.21** – Bidimensional graph of the count rate (Mapping) measured with two different TRITIUM cosmic detectors using a radioactive source of  $^{60}\text{Co}$ .

surface, giving a fairly similar counting rate in all the points considered.

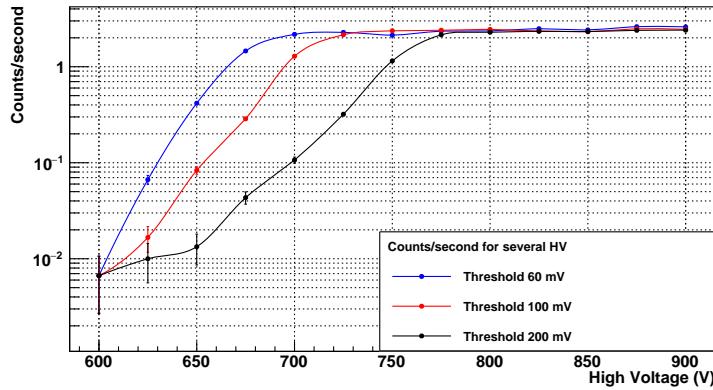
The following studies of this section were done at the level of a cosmic veto (both detectors in coincidence), so the configuration of the electronics was that of Figure 3.14c. The following step is to find the conditions in which the detection of cosmic events is optimized. This optimization consists of, on the one hand, finding the minimum high voltage of PMTs for which their efficiency is stable, and, on the other hand, finding the maximum threshold of the discriminator<sup>9</sup> before starting to loss cosmic events. For a higher high voltage and a smaller threshold, a plateau of the counting rate should be found.

The counting rate was measured for several high voltages at fixed thresholds and for several thresholds at fixed high voltages. Both measurements are shown in Figure 4.22. To find the optimal conditions the amplification line of the configuration of the electronics 3.14c was eliminated and the output signal of the coincidence module was connected to a CAEN Quad Scaler And Preset Counter-Timer module, N. 1145, [coma]. The counting rate was measured in a time window of 300 s.

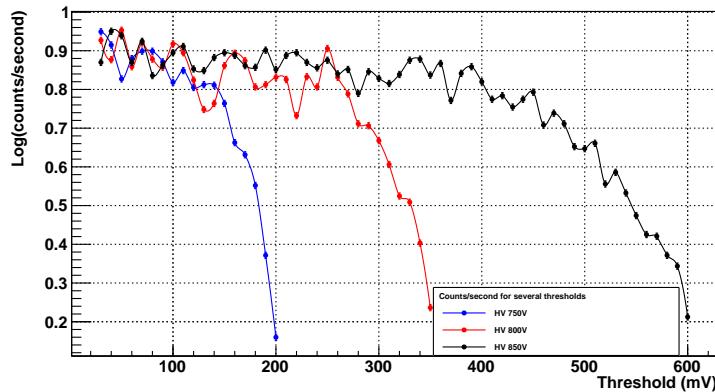
In Figure 4.22a, the measurements at several high voltage is exhibited, which was done for three different thresholds, 60 mV, 100 mV and 200 mV. As it can be observed, there is a minimum high voltage for each threshold, 700 V, 730 V and 780 V respectively, at which the plateau start. This minimum voltage is higher when the value of the threshold is increased, as it should happen. The voltage chosen was 800 V since it is on the plateau for the three thresholds. Analogously, the counting rate for several thresholds and fixed high voltage was measured for three different high voltages, 750 V, 800 V and 850 V, which is plotted in Figure 4.22b. There is a maximum threshold for every high voltage used, 140 mV, 270 mV and 450 mV

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<sup>9</sup>The threshold is the voltage value that the PMT output signals must exceed to contribute to the cosmic detection



(a) Counting rate as a function of high voltage for three different thresholds in semilogarithmic scale.

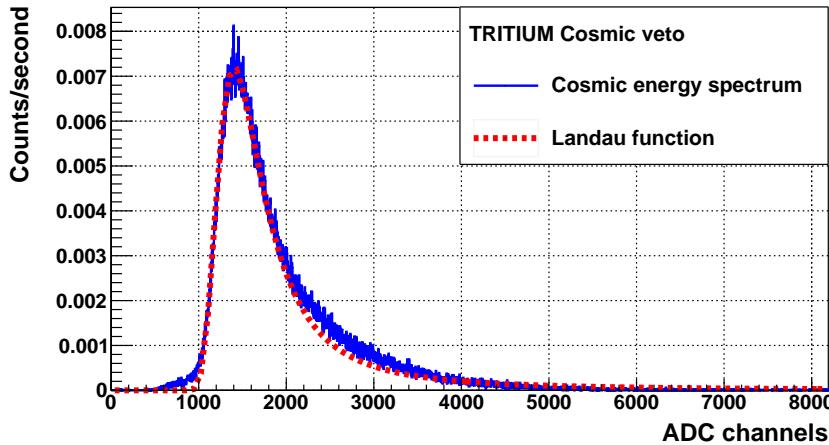


(b) Counting rate as a function of threshold for three different high voltage in semilogarithmic scale.

**Figura 4.22** – Counting rate as a function of high voltage for fixed thresholds and as a function of thresholds for fixed high voltage.

respectively, at which the plateau ends. This maximum threshold increases with high voltage, as it should happen. The threshold chosen was 200 mV which is on the plateau for the selected high voltage, 800 V.

Next, the energy spectrum of cosmic events was measured, which is shown in Figure 4.23. For this task, the configuration of the electronics is given in Figure 3.14c for 800 V and 200 mV of HV and threshold, respectively.

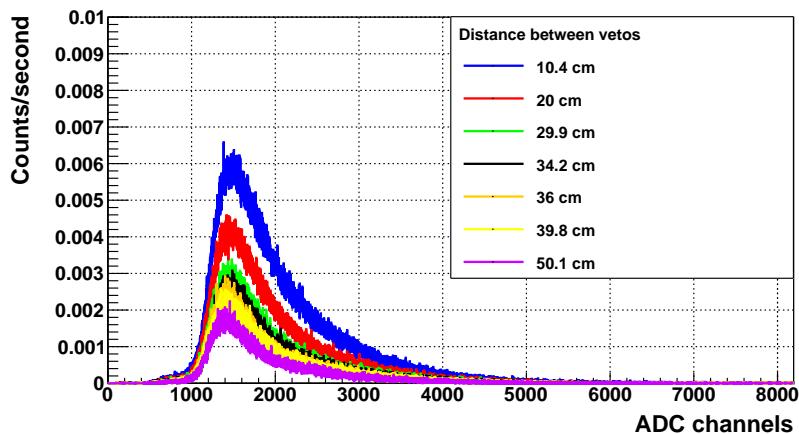


**Figura 4.23** – Energy spectrum measured with the cosmic veto.

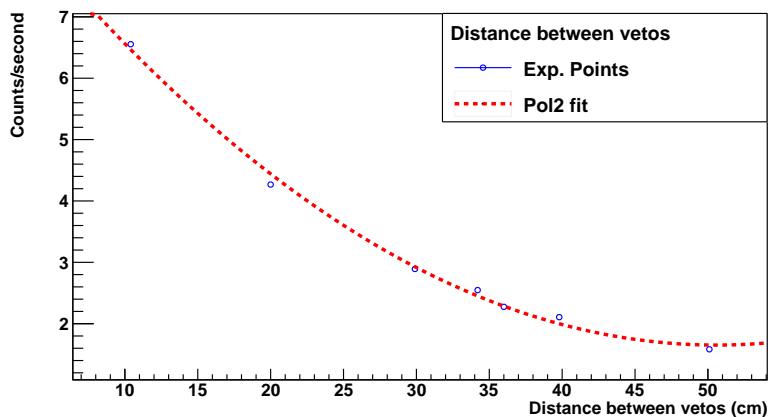
As it is expected, this energy spectrum fits well to a landau function. The cosmic ray rate can be determined from the area of this spectrum, which is 2,5 event/s. The expected cosmic rate, calculated in section 3.4.2, is 2,9 event/s, so the efficiency of the active veto developed in TRITIUM experiment for cosmic events detection is 85%, which is a usual value for the efficiency of plastic detectors.

Finally the relationship between the detected cosmic ray rate and the distance between the two plastics of the cosmic veto was obtained. The energy spectrum was measured for five different distances, namely 10 cm, 20 cm, 36 cm, 40 cm and 50 cm. The spectra are plotted in Figure 4.24a.

The energy spectrum of Figure 4.23 was also included. As it is expected, the counting rate decreases with the distance but the spectrum shape remains the same. The integrated spectra as a function of distance, plotted in Figure 4.24b, was fitted to a second degree polynomial which allows to estimate the cosmic rate if the veto distance is changed.



(a) Energy spectrum of the cosmic veto for several distance.



(b) Fit of counts per second measured with the cosmic veto for several distance to a second degree polynomial.

**Figura 4.24** – Measurement of the cosmic veto for several distances between its cosmic detectors.



# Chapter 5

## TRITIUM Monitor Prototypes

This chapter describes the different prototypes that was developed in the framework of the TRITIUM experiment, which are TRITIUM-IFIC 0, TRITIUM-IFIC 1, TRITIUM Aveiro 0 and TRITIUM-IFIC 2, listed in chronological order of their construction.

The first two prototypes built are preliminary prototypes used to learn about tritium detection and to detect and solve problems in their designs.

The other two prototypes built are prototypes with a well-defined design in which no problems were found. They were built to check more subtle effects.

Each prototype was designed and built in the laboratories of the university (IFIC, Valencia or Aveiro, Portugal) and it was filled with tritiated water following a protocol specially developed for this task, described in appendix C. Several water tightness and filling tests were carried out in each prototype to guarantee its radiosecurity.

Finally, the final monitor of TRITIUM detector will be explained.

It is based on modular detection units for easy scalability, where each module is the chosen prototype (the one with the best results).

## 5.1 Preliminary IFIC prototypes, TRITIUM-IFIC 0 and TRITIUM-IFIC 1

Two preliminary prototypes, TRITIUM-IFIC 0 and TRITIUM-IFIC 1, are shown in this section, which were designed, developed and built at the IFIC workshop.

These prototypes were thought to be a small scale of the final TRITIUM detector module and they were used to learn about tritium detection, to detect possible problems in their designs and to verify the improvements applied to them.

### 5.1.1 TRITIUM-IFIC 0

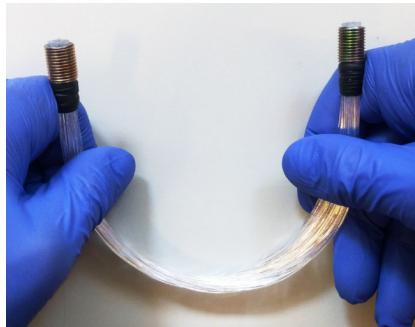
The TRITIUM-IFIC 0 prototype was the first prototype developed in TRITIUM experiment and it was used to check the feasibility of the technology proposed by TRITIUM, that's, to verify that it is possible to detect tritium in water using scintillating fibers.

Due to the problems that arise when liquid radioactive sources are used, the design of this first prototype paid special attention to radiation safety, rather than in detecting tritium efficiency.

TRITIUM-IFIC 0 consists of bundle of 35 fibers, shown in Figure 5.1, with a length of 20 cm, which were cut and polished with the techniques explained in section 4.1.1. This bundle has a metallic pieze located in both ends, shown in Figure 5.1a, which are used to fix it to the prototype.



(a) Metallic piece of the fiber bundle.



(b) Fiber bundle in a position similar to the prototype.



(c) Bundle of fibers in a straight position.

**Figura 5.1** – Bundle of 35 fibers, the length of which is 20 cm, used in TRITIUM-IFIC 0 prototype

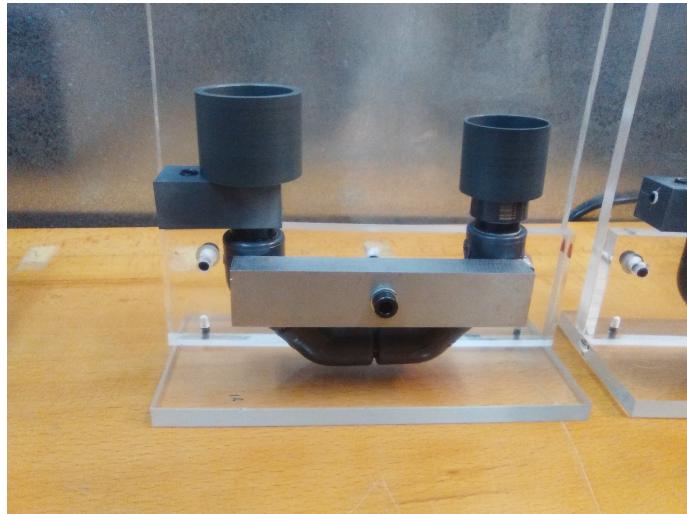
This bundle is placed inside of a vessel, whose material is PVC<sup>1</sup> since it is a safe material widely used. This vessel, shown in Figure 5.2, was designed in a U-shape to improve the radiological safety, although this shape worsen the efficiency of tritium detection.

As can be seen in Figure 5.2, a piece of methacrylate and steel was designed and built to hold the detector. Two calibrated PMTs were optically coupled directly to the fiber bundle ends using optical grease [Cera].

The employed PMTs were the model R8520-460 from Hamamatsu company [K.K19], whose reference number are ZB2771 and ZB2773, and the electronic circuit, shown in figure 3.9, was used to distributed the high voltage between the dynodes. The employed high voltage was  $-800$  V, at which their gain are  $1.26 \cdot 10^6$  and  $1.01 \cdot 10^6$  and their quantum efficiency are 29.76% and 28.66% respectively. Their signals were precessed and analyzed

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<sup>1</sup>Polyvinyl Chloride, PVC



**Figura 5.2 – TRITIUM-IFIC 0 Prototype.**

using the electronic configuration shown in Figure 3.14b.

Two identical prototypes were built and filled following the same protocol but using different liquid solutions. The first prototype, called TRITIUM-IFIC 0 Background, was filled only with ultrapure water (39 mL, uncertainty of 0.05%) and it was used to measure the radioactive background of the detector whereas the other prototype, called TRITIUM-IFIC 0 Signal, was filled with a radioactive liquid source of tritium, the preparation of which is explained in the appendix C. The specific activity of the liquid source employed was 99.696 kBq/L (uncertainty of 2.24%) and the volume used to fill this prototype was the same, 39 mL (uncertainty of 0.05%). Therefore, the total activity of this tritiated water sample is approximately  $3.888 \pm 0.087$  kBq.

This second prototype was used to measure the signal of the detector (tritium + background) and the measured tritium activity can be known by extracting the background (measurement of TRITIUM-IFIC 0 Background) to the signal (measurement of TRITIUM-IFIC 0 Signal).

A statistically significant amount of time coincident events was not found in both PMTs, so the measurement of time coincidence was not possible.

The loss of photons could be caused for several reasons, such as an excessive curvature in the fiber bundle due to the U-shape of TRITIUM-IFIC 0 prototype, causing that too many photons escape from the fibers, or the poor quality of the tritiated water-fiber interface (the cleaning process explained in section 4.1.4 was motivated by this result).

To avoid this problem and obtain some results with this prototype, a measurement was performed with a single PMT. For this task, the electronic configuration shown in Figure 3.14a was used. The results of these measurements are shown in section 7.1.1, where they are discussed.

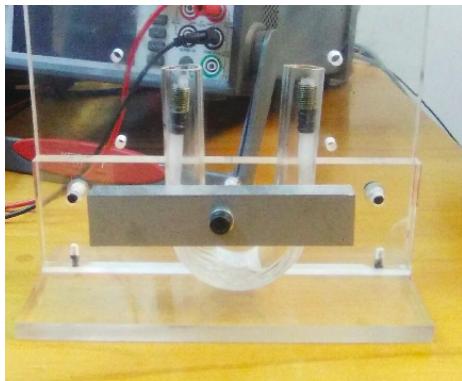
In addition, a test was carried out to explain why it was not possible to measure both PMTs in time coincidence. For this task a transparent PMMA vessel, shown in Figure 5.3a, was built in a similar shape to that of the TRITIUM-IFIC 0 prototype vessel to check the effect of the fiber bundle curve.

The LED described- in section 4.1.3 was used to verify the reduction in photocollectio efficiency of the fiber bundle due to this curve.

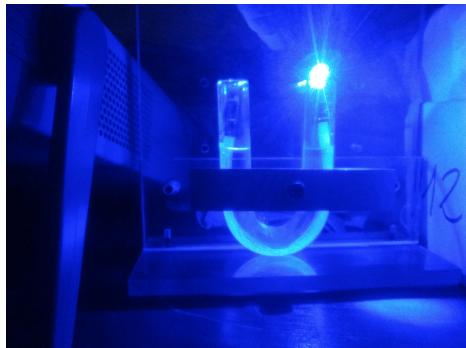
As can be seen visually in Figure 5.3b, a large percentage of the photons are lost due to the curve, which can be easily solved by using a straight fiber arrangement in next prototypes.

### 5.1.2 TRITIUM-IFIC 1

The next prototype, TRITIUM-IFIC 1, was intended to overcome the problems and limitations found in the previous one, TRITIUM-IFIC 0, section 5.1.1. To do so, some improvements were applied on it:



(a) PMMA vessel.



(b) Test performed to check the lost photons.

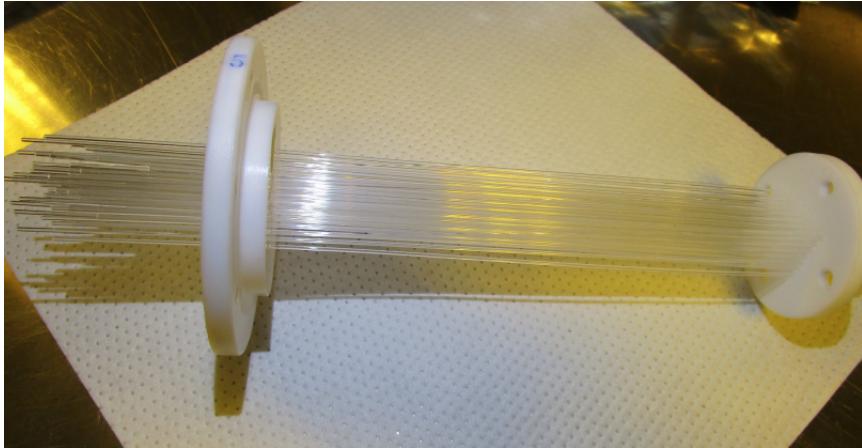
**Figura 5.3** – PMMA vessel used to check photon loss due to fiber bundle curve.

1. The fiber bundle is arranged straight to optimize the photon collection efficiency of the fibers.
2. A special fiber cleaning protocol, explained in section 4.1.4, was applied on the fibers. It was used to improve the interfaces between fiber and tritiated water, creating a better wetting property of the fiber, which will result in more tritium events detected and a larger photon collection efficiency.
3. Teflon vessel was using in the Tritium prototypes to reduce the effect of the small photon collection efficiency of the fibers, measured in section 4.1.3, which is an innerent characteristic of the fiber which cannot be changed.

Teflon is an interesting material for its optical properties, specifically its reflection factor, which is very close to 100% at the working wavelength.

The TRITIUM-IFIC 1 prototype consists of 64 scintillating fibers, with a length of 20 cm, that are arranged in a straight position using a

teflon structure, shown in Figure 5.4. The fibers are fixed in an  $8 \cdot 8$  square matrix.



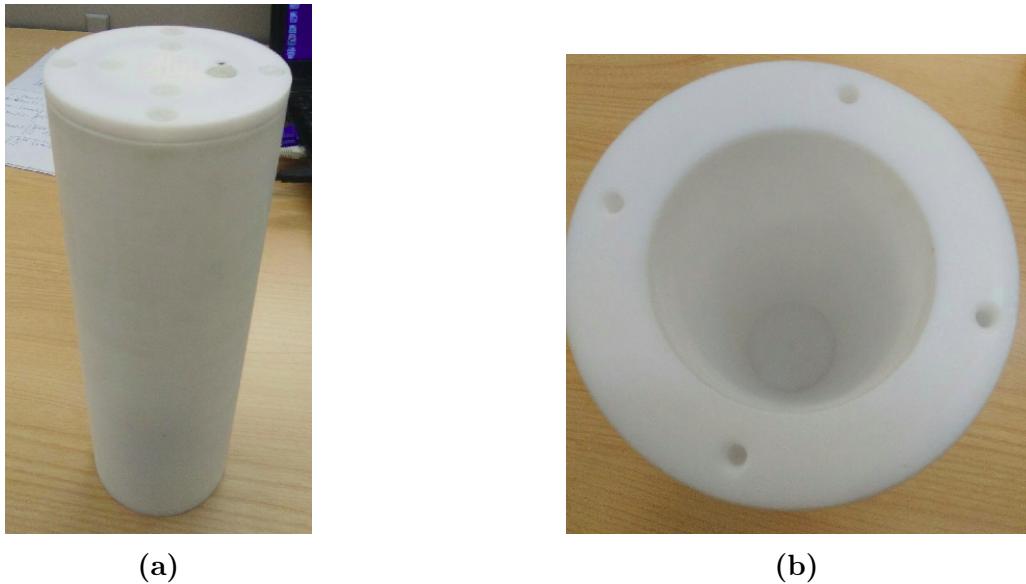
**Figura 5.4** – Teflon structure used to arrange the fibers of TRITIUM-IFIC 1 prototype in a matrix of  $8 \cdot 8$ .

A new teflon vessel was designed and built, shown in Figure 5.5. It has a cylindrical hole whose internal diameter and length are 48 mm and 200 mm respectively, where the fiber structure will be placed.

In addition to cutting and polishing the scintillating fibers, a cleaning process, described in section 4.1.4, was applied to them to achieve a better tritiated water-fiber interface.

A piece of PVC was used to fix the photosensor on the prototype and prevent the photosensor from being affected by external light. A general view of this prototype is shown in Figure 5.6, which, for radioactivity safety reasons, will be read using only one PMT.

The PMT used was the model R8520-460, from Hamamatsu Photonics company [K.K19] and it was coupled directly to the fiber bundle using optical grease [Cera]. The electronic circuit, shown in Figure 3.9, was used to distributed the high voltage between the dynodes. The employed



**Figura 5.5** – Teflon vessel of TRITIUM-IFIC 1 prototype

high voltage was  $-800$  V, at which, its quantum efficiency is 28.66%.

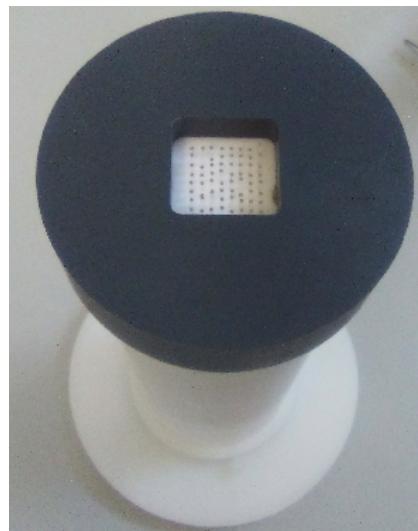
The signal from this PMT was processed and analyzed using the same electronic configuration as that used for the TRITIUM-IFIC 0 prototype, shown in Figure 3.14a.

Unlike the previous prototype, only one TRITIUM-IFIC 1 was built. First, it was filled with ultrapure water (118 mL, uncertainty of 0.05%) and several background measurements were taken over a week. Then, it was emptied and refilled using 118 mL (uncertainty of 0.05%) of the radioactive liquid source of tritium explained in appendix C.

The results of these measurements are shown in section 7.1.2, where they are discussed and compared with the result obtained with the previous prototype, TRITIUM-IFIC 0.



(a)



(b)

**Figura 5.6** – A general view of TRITIUM-IFIC 1 prototype

## 5.2 Latest TRITIUM Prototypes

This section shows the last prototypes, TRITIUM-Aveiro 0 and TRITIUM-IFIC 2, in which the problems previously found are solved and a well-defined design is developed for them.

A different design was employed in both prototypes so that they can allow the reading of a large number of fibers arranged in a straight position with two photosensors in time coincidence, in a safe way.

Particular attention to tritium detection efficiency was paid in these prototypes, which is the reason why they use many more fibers than the preliminary prototypes and a time coincidence reading of these fibers is done using two photosensors.

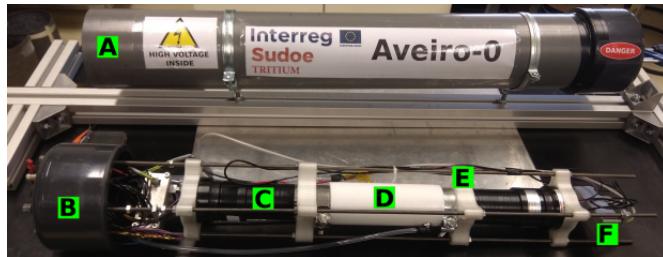
Furthermore, the activity of the radioactive liquid source of tritium, which is used to fill these prototypes, is much lower than previous prototypes since the interesting parameter to be measured is their low detection level, LDL.

The design of both prototypes is very similar and their objective is to test the subtle effects caused by the little difference, such as the diameter of the fibers used, and to choose the one with the best results which will be included in the final design of the TRITIUM monitor.

### 5.2.1 TRITIUM-Aveiro 0

The third prototype built and the first thought to be the final version of the TRITIUM detector module was TRITIUM-Aveiro 0, shown in Figure 5.7, which was designed and built in the workshop of the University of Aveiro.

It consists of a teflon vessel (D of Figure 5.7), shown in Figure 5.8,



**Figura 5.7** – TRITIUM-Aveiro prototype.

which has an internal cylindrical hole the diameter and length of which are 43 mm and 18 cm respectively.



**Figura 5.8** – Teflon structure and fiber bundle used in TRITIUM-Aveiro 0 prototype.

This vessel contains 360 no-clad scintillating fibers with a length of 180 mm. The model of the used fibers is BCF-10 from Saint-Gobain company [Cer01], which have practically the same characteristics than the others used up to now (BCF-12 fibers) and their most important difference is the diameter, which is the double, 2 mm.

A larger diameter could be interesting because it facilitates the flow of water around the fibers, reducing the problems related to surface tension and ensuring that the entire active volume of the fibers is used for tritium detection. In addition, it increase the resistance of the fibers, which

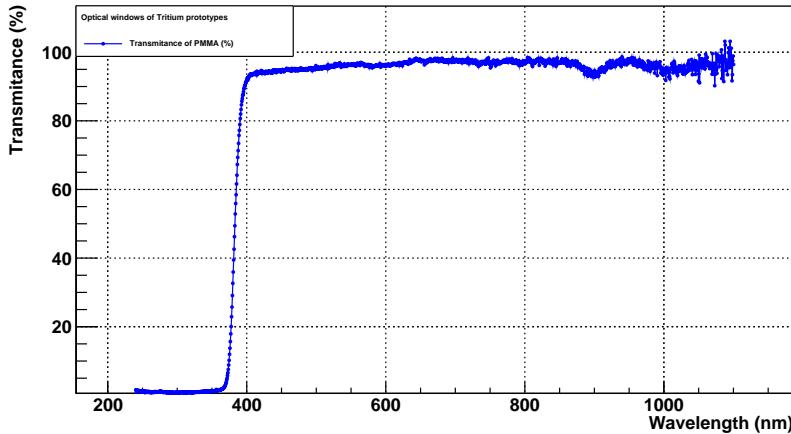
is very important since the water is flow around them. However, it could be detrimental since this worsens the signal-to-background ratio. The detector active volume for 2 mm fibers in the same space is smaller, producing a smaller tritium signal and the part of the fibers where no tritium events reach (they only contribute to the background) is larger, producing a larger background.

In order to quantify the importance of the fiber diameter effect, the measurements were compared with similar measurements performed with TRITIUM-IFIC 2 prototype, shown in section 5.2.2, based on a similar configuration with 1 mm fibers (BCF-12 model).

The amount of fibers used in TRITIUM-Aveiro 0 prototype is the maximum which allows the water to flow around the fibers and a structure was not feasible to use to fix them due to the large amount of them used. These fibers were cut with the fiber cutting device developed by TRITIUM but they were neither polished nor cleaned. The reason for this is that the automatic polishing machine was not yet developed and it was not feasible to polish 360 fibers by hand. In fact, the automatic polishing machine was motivated by the amount of fibers used in this prototypes.

To ensure the radiosecurity of this prototype, the teflon vessel is totally closed and a water inlet/outlet were installed in it to allow a constant water flux through it. Two PMMA windows was used to read the fibers, whose thick is 10 mm, which was located at both ends of the fiber bundle. Two clamps are used to press the Teflon walls against the PMMA windows to ensure the water tightness of the prototype. PMMA was chosen for its optical properties, especially its transmission coefficient, which was measured for visible light range in the ICMOL laboratories, shown in Figure 5.9.

As can be seen, its transmission coefficient is approximately 95% for the working wavelength (435 nm). Slightly better transmission coeffi-



**Figura 5.9** – Transmission spectrum of light (in the visible range) in a piece of PMMA of X thickness measured in the ICMOL laboratory.

lients can be achieved with other materials such as quartz or sapphire but they are much more expensive.

Two PMTs (C of Figure 5.7) are used to read this prototype in time coincidence, which are powered at  $-1500$  V, at which the quantum efficiency is 26%. They are fixed to both fiber bundle ends of the prototype using two pieces (E of Figure 5.7) which was designed and built with a 3D printer. Both PMTs are optically coupled to the PMMA windows using optical grease [Cera].

The PMTs used are the model R2154-02 2" from Hamamatsu company [K.K10], whose characteristics, specially its gain and efficiency, are quite similar to the PMTs used in the other prototypes.

All these different parts, together with the electronic system (F of Figure 5.7), is arranged in a structure, shown in Figure 5.7, which is based on several nuts located on four long stainless-steel screws. This screws are fixed to an external PVC structure, A and B of Figure 5.7, which is used to protect the prototype from physical damage and provide a light-tight

operation environment. This PVC structure is equipped with several high voltage power, low voltage power and signals feed-through connectors.

Only one prototype was built, which was designed to be installed in the Arrocampo dam and an electronic chain, based on several PCBs, was specially designed, developed, built and tested to process and analyze the signals of this system, shown in appendix D.

Two interfaces were developed, one to control the power supply to the PMTs, shown in Figure 5.10a, and the other to control the different options of the electronic reading chain, such as thresholds, shown in Figure 5.10b.

First some measurements were taken in the laboratory, which were used to characterize the detector. For this task it was firstly filled with ultra-pure water, which was used to measure the background of the detector, and then, with a radioactive liquid tritium solution with an activity of 30 kBq/L, which were used to measure the efficiency and the low detection level, LDL, of the prototype. The volume of ultrapure water and tritium solution used in TRITIUM-Aveiro 0 prototype is 58 mL. Later, it was installed in the arrocampo dam to test its functionality and to begin with the tritium level monitoring. The laboratory and Arrocampo measurements, both, are shown in section 7.1.3, where they are discussed and compared with the measurements of the previous prototypes.

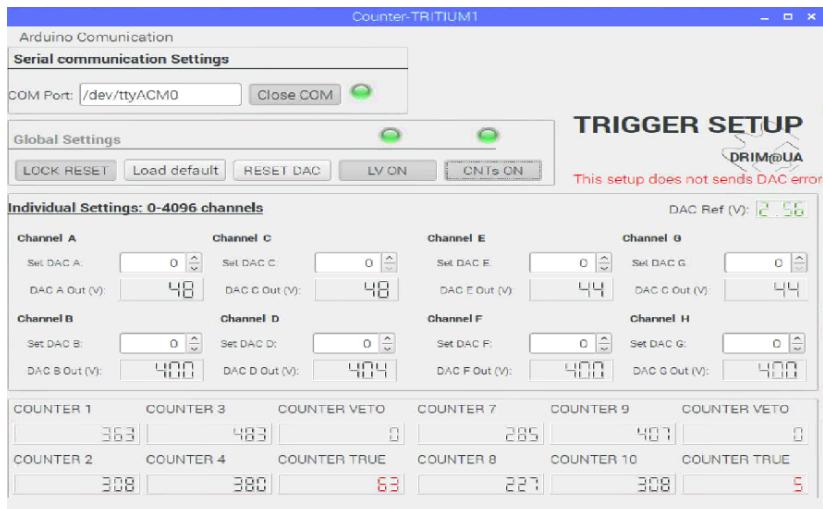
### 5.2.2 Advanced IFIC prototype, Tritium-IFIC 2

The last prototype developed in TRITIUM experiment and the second thought to be the final version of the TRITIUM detector module was TRITIUM-IFIC 2, which is shown in Figure 5.11, A.

This prototype was designed and built in the IFIC workshop and

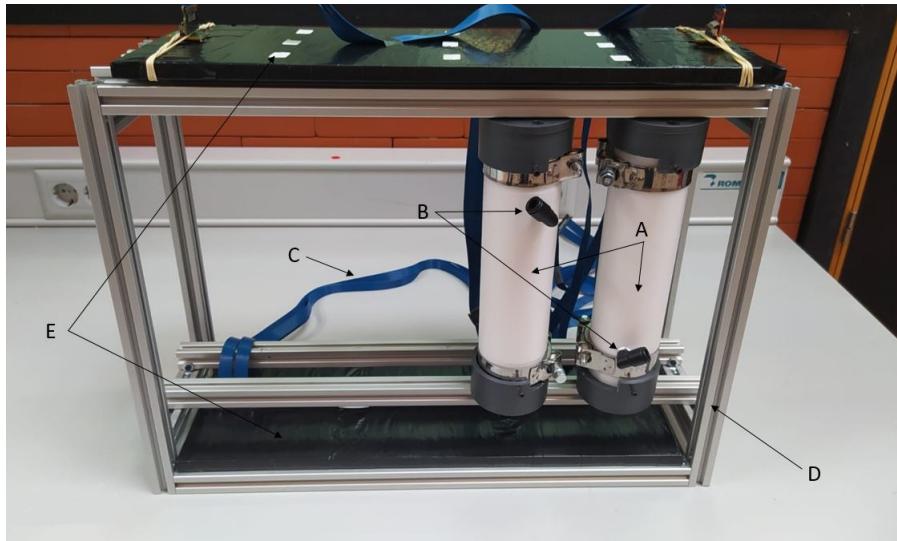


(a) Graphical user interface to manage the power supply voltage of PMTs.



(b) Graphical user interface used to manage the counter system.

**Figura 5.10** – Graphical User Interface developed to control the TRITIUM-Aveiro prototype.



**Figura 5.11 – TRITIUM-IFIC 2 prototype.**

it consists of a cylindrical teflon vessel, shown in Figure 5.12, the shape of which is similar to the one used in TRITIUM-Aveiro 0 prototype. The internal length and diameter of the teflon vessel are 210 mm and 36 mm respectively.

This prototype contains 800 no-clad scintillating fibers, model BCF-12, with a length of 200 mm, a larger number of fibers than the TRITIUM-Aveiro 0 prototype which are arranged in less volume.

The fibers used are cut, polished and cleaned with the conditioning processes previously shown in section 4.1 since, during the construction of the prototype, the development of the automatic polishing machine was completed.

These fibers are freely arranged, with a density that allows water to flow through the fibers and two PMMA windows, located at the ends of the fiber bundle, were used to read this system, similar to the TRITIUM-Aveiro 0 prototype.



(a) TRITIUM-IFIC 2 vessel.



(b) TRITIUM-IFIC 2 vessel with PVC caps.

**Figura 5.12** – TRITIUM-IFIC 2 teflon vessel.

The width of the PMMA optical windows used is 5 mm, which is sufficient to guarantee radiosecurity since the detector works at very low water pressure and two clamps are used to ensure the watertightness of the prototype, similar to the TRITIUM-Aveiro 0 prototype. The transmission coefficient, shown in Figure 5.9, is practically unaffected by the little difference of the PMMA width used in both prototypes.

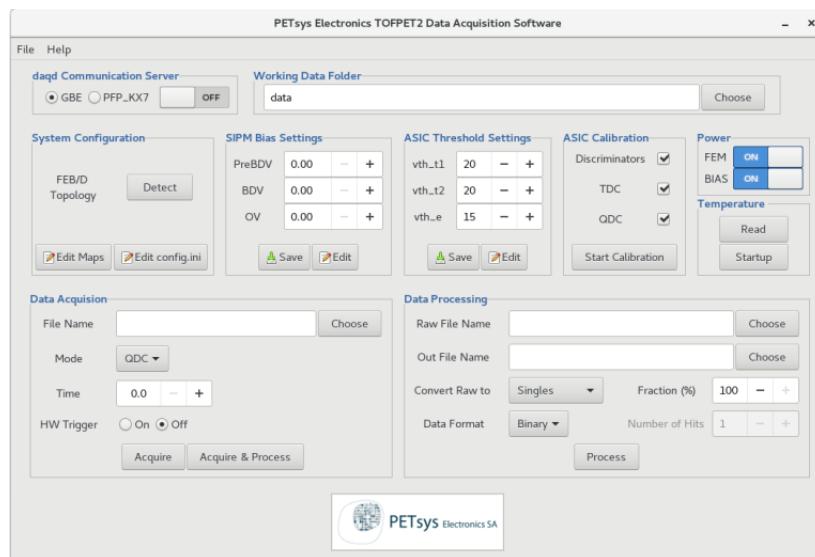
As can be seen in Figure 5.11, B, and Figure 5.12, a water inlet/outlet was installed in the teflon vessel to allow a constant water flux through it, similar to the TRITIUM-Aveiro 0 prototype.

For the first laboratory measurements, two PMTs were used, model R8520-460 from the Hamamatsu Photonics company [K.K19], which is useful to understand the results and compare them with the results obtained with the previous prototypes. However, measurements with SiPM arrays have already started the output signal of which is connected to PETSYS

system through flat wires as can be seen in Figure 5.11, C.

As SiPM arrays readout by the PETSYS system will be employed in the TRITIUM-IFIC 2 prototype that will be installed in Arrocampo dam, it is not necessary to develop a electronics chain to process and analyze the PMT signals of this prototype.

Like the TRITIUM-Aveiro 0 prototype, although PETSYS has a graphical user interface, shown in Figure 5.13, which allows controlling all the different options such as the voltage with which the SiPM arrays is fed, the thresholds used, etc., normally it will be controlled remotely via computer terminal.



**Figura 5.13 – Graphical User Interface (GUI) of PETSYS.**

Two PVC caps, located at both ends of the prototype, Figure 5.12b, were used to work with the SiPMs in a light-tight environment and an aluminum structure, shown in Figure 5.11, was designed and built to house up to 10 TRITIUM-IFIC 2 modules and two cosmic vetos, shown in Figure 5.11, E.

The available space of the lead shielding, explained in section 3.4.1 is enough to accommodate up to 5 structures like the one shown in Figure 5.11. It means that the final TRITIUM module has the capacity to contains 50 TRITIUM-IFIC 2 modules and 5 different cosmic vetos. As the sensibility of the TRITIUM monitor scales with the number of TRITIUM modules used, the results obtained with the TRITIUM monitor should improve the results obtained with the TRITIUM-IFIC 2 prototype by a factor of N, where N is the number of cells used.

Two identical TRITIUM-IFIC 2 prototypes were built, similar to the TRITIUM-IFIC 0 prototype, one of them was filled with ultrapure water and used to measure the background and the other was filled with a radioactive liquid source of tritium and used to measure the signal. The volume used in both cases was 82 mL (uncertainty of 0.05%).

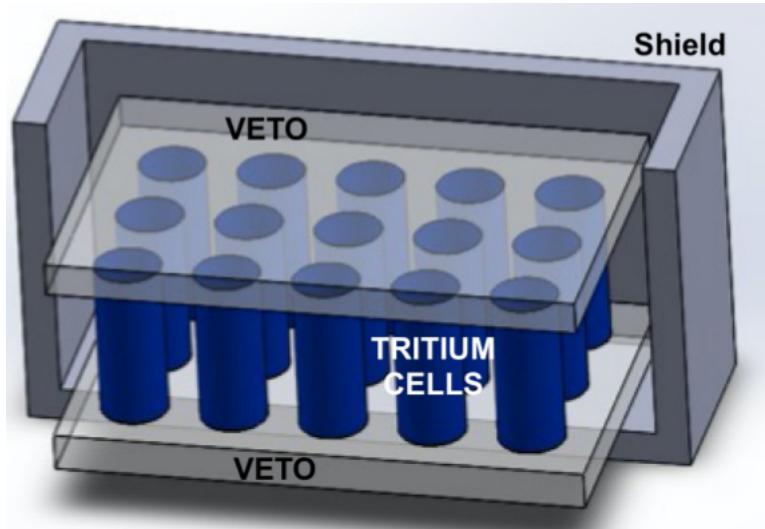
The activity of the tritium source used for this prototype is 10 kBq/L (uncertainty of 2.24%), which was prepared by diluting a sample of tritiated water explained in appendix C with ultrapure water until the desired activity was achieved.

The results of this prototype is shown in section 7.1.4, where they are compared with the results of the previous prototypes and, specially, with TRITIUM-Aveiro 0 prototype.

## 5.3 Modular TRITIUM Detector for In-Situ Tritium Monitoring

Lastly, this section presents the final TRITIUM monitor, a schematic design of which is shown in Figure 5.14.

It consists of several TRITIUM modules, shown in Figure 5.14,



**Figura 5.14** – A schematic design of the TRITIUM detector.

which are read in parallel. Each module will be the prototypes that achieves better results, TRITIUM-Aveiro 0 (section 5.2.1) or TRITIUM-IFIC 2 (section 5.2.2).

These modules are isolated from environmental radioactivity using three different techniques.

1. First, an external lead shielding, explained in section 3.4.1, a part of which is shown in Figure 5.14. This is used to stop the environmental radioactivity which increase the radioactive background measurement of the TRITIUM monitor
2. Second, several active vetos, explained in section 3.4.2 and characterized in section 4.4, which is shown in Figure 5.14, placed below and above the TRITIUM modules. These active vetos are read in anti-coincidence to eliminate the effect of the high energy events of the background (mainly cosmic events) on the TRITIUM measurement.
3. Finally, the radioactive elements present in the water samples, intro-

duced into TRITIUM modules to be measured, are eliminated using an ultrapure water system, shown in section 3.3 and appendix B and characterized in section 4.3.

The ultrapure water system, lead shielding and a TRITIUM-Aveiro 0 prototype are installed and currently in operation at the Arrocampo dam. This entire system was used to successfully monitor the tritium levels in Arrocampo dam during three months. Furthermore, two additional TRITIUM-Aveiro 0 prototypes and four active vetos are currently under manufacturing to be installed, which are measured in parallel with the current prototype installed.

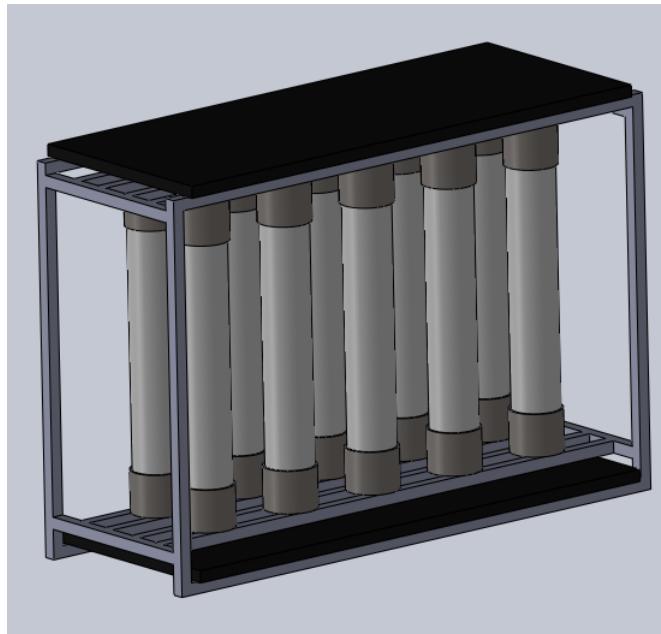
The RaspberryPi, in which the counter electronic system of TRITIUM-Aveiro prototype is based, has some counting limitations if multiple modules are used. To overcome this problem, it must be replaced with an FPGA-based counter board to ensure reliable counting.

At the same time, three TRITIUM-IFIC 2 prototypes and an active veto have already been built, the installation of which at the Arrocampo dam was delayed due to the coronavirus pandemic restrictions imposed in Spain. They will be installed as soon as possible.

One of the most important points of the TRITIUM detector is its modular design, with which an scalability can be achieved to reach the required sensitivity, 100 Bq/L. If this sensitivity goal is not reached with the three modules that will be installed, only some additional modules need to be installed to improve it.

The only scalability restriction is the available space, which is fixed by the lead shield already built and installed (it is also fixed by the available space in the house where the lead shield is installed). Taking into account the currently available space, five different structures designed for TRITIUM-IFIC 2, one of which is shown in Figure 5.15, can be used, where

ten different modules (grey) and an active veto (black) can be accommodated in any of them.



**Figura 5.15** – A TRITIUM detector design based on the TRITIUM-IFIC 2 prototype.

It means that up to 50 TRITIUM-IFIC 2 modules can be used in parallel, reducing the TRITIUM detector sensitivity by a factor of fifty. In light of the results obtained with these prototypes, it is not expected to use more than one structure like the one shown in Figure 5.15 (ten modules) to reach the target sensitivity.

# Chapter 6

## Simulations

This chapter details the Monte Carlo simulations that were performed in the TRITIUM experiment. Their goal were to choose the best design for the TRITIUM detector, understand its behavior and find its limitaitons. The simulation environment used for this task is Geant4 [Col21, Ago03].

### 6.1 Geant4 Environment

Geant4 is a software toolkit for the simulation of the passage of particles through matter. It is a package developed at CERN that is based on object-oriented technology implemented in the C ++ programming language.

This includes the definition of all the different aspects of the simulation process such as detector geometry, materials used, particles of interest, pyhisics processes that handle particle and matter interactions, response of sensitive detectors, generation, storage and analysis of event data and visualization.

Geant4 simulates particle-by-particle physics. It means that the tritium events are initialized one by one, whose energy, moment, position, etc. are determined. Then, the propagation of each tritium event and its interaction with the scintillator is simulated, in which optical photons are created. The propagation of these optical photons are also simulated one by one and the simulation ends when all tritium events have been simulated and all created optical photons have been absorbed by either the sensitive detector or other materials present in the simulation.

A physics list used for these simulations is Livermore, G4EmLivermorePhysics, which is specially designed to work with low energy particles. This list includes the most important electromagnetic process at low energies such as Bremsstrahlung, Coulomb scattering, atomic de-excitation (fluorescence) and other related effects.

The materials used in these simulations were water (to simulate the tritiated water source), PMMA (to simulate the optical windows of the prototype), polystyrene (to simulate the core of scintillating fibers), teflon (to simulate the prototype vessel), silicone (to simulate the optical grease), silicate glass (to simulate the optical windows of the PMTs) and bialkali (to simulate the photocathode material of the PMT).

The properties of water, teflon, polystyrene were taken from the Geant4 NIST database and the other materials were built by specifying their atoms. Optical properties was added to these materials:

1. First, the spectrum of the refractive index and the light attenuation were added to the water which was obtained from the reference [Bui94]. Furthermore, an electron emission, uniformly distributed in the volume, was added to the water, the energy of which was calculated using the tritium energy spectrum. This emission of electrons simulates the disintegration of the dissolved tritium particles in the water sample. The used data was obtained from the reference [Mer15].

2. Second, the spectrums of the refractive index, the light attenuation and the photon emission were added to the polystyrene, which was obtained from the data sheet of scintillating fibers, [Cer05]. Also the scintillation yield and the decay time was included.
3. Third, the quantum efficiency spectrum was included to the photocathode material of the PMTs, the data of which was obtained from their data sheet, [K.K19] and a refraction index of 1.46 was used for the optical grease, also obtained from its data sheet, [Cera].
4. Finally, the optical data for the remaining materials, PMMA windows, teflon and silicate glass, were taken from the reference [Arg11].

It is important to note that the simulations shown in this theses are focused on the Tritium-IFIC 2 prototype since these were the simulations I was primarily working on, but a similar simulation was performed for the Tritium-Aveiro prototype, some of their most important results are also presented. In addition, other smaller simulations are shown used to choose the best design of the TRITIUM detector.

## 6.2 Description of the Simulations Performed

Several simulations was designed and run during the life of the TRITIUM project to quantify how different values of interesting parameters affect to the tritium measurement and to understand the behaviour of the different TRITIUM prototype built which are shown in the following sections.

### 6.2.1 Tritiated Water Source

First of all the shape of the simulated tritiated water source was optimized.

The mean free path of tritium electrons in water are only around  $5 \mu\text{m}$ , so there are many electrons that don't reach the scintillating fibers. These are electrons that don't provide useful information and only contribute to being time consuming and reducing available computing resources.

To optimize the simulation, the dimensions of the simulated tritium source was studied. The goal of this study was to minimizes the tritium events that do not reach the scintillating fibers avoiding missing the tritium events.

A simulation test was performed to ensure that the tritium source is simulated correctly. This test consists of a scintillating fiber 20 cm long and 2 mm in diameter and a surrounding tritiated water source of the same length and 0.5 mm thick (100 times greater than the mean free path of tritium electrons) to ensure that this study take into account all possible tritium electrons that can reach to the scintillating fiber.

The dimensions of the fiber are not important in this study since only the energy deposition of tritium electrons in the fiber were simulated. That is, this simulation doesn't include the following steps such as photon generation, propagation of these photons, etc. in which the shape of the scintillating fiber becomes important.

The results of this simulation are shown in section 8.1.1, where they are discussed.

### 6.2.2 Energy Deposition and Light Output of Scintillating Fibers

The scintillation yield provided by the manufacturer, 8000 phot./MeV, only works for Minimum Ionizing Particles (MIP). As tritium electron energies are far from being MIP particles, the output light generated by the scin-

tillating fibers was studied. For this task, the energy deposition of tritium electrons in scintillating fibers and their subsequent emission of scintillation photons was added to the simulation.

When no MIP particles are detected in plastic scintillators a light quenching effect affect to the proportionality between the output light per unit of path length,  $\frac{dL}{dx}$ , and the energy deposited per unit of path length,  $\frac{dE}{dx}$ , through the so-called Birks coefficient, following equation [Bir51].

$$\frac{dL}{dx} = S \frac{\frac{dE}{dx}}{1 + k_B \frac{dE}{dx}} \quad (6.1)$$

where  $S$  is the scintillation yield, provided by the manufacturer. A value of  $k_B = 0.126 \text{ mm/MeV}$  was considered for the Birk's coefficient which is the one used for scintillators based on polystyrene [Lev11]. The effect of this correction is shown and discussed in section 8.1.2.

### 6.2.3 Fiber Length Optimization

Another test was performed to find the fiber length that optimizes the tritium detection efficiency. Two different lengths of the scintillating fiber were considered in this study, 1 m and 25 cm.

As the active area of the detector is related with its tritium detection efficiency, the advantage to use a longer fibers is that larger active areas can be achieved with a less number of cells, considerably reducing the number of used photosensors and, as a consequence, the price of the TRITIUM monitor. However, smaller length of scintillating fibers reduce de photon absorption produced in the fibers, increasing the tritium detection efficiency for the same active area.

For this task, the Tritium-Aveiro prototype was simulated, consisting of a similar design of TRITIUM-IFIC 2 prototype but using 360 scintillating fibers of 2 mm diameter and readout by two different photo-sensors, model R2154-02 2" from Hamamatsu company [K.K10]. All optical properties mentioned in section 6.1 were included in this study.

The results of this study are shown in section 8.1.3, where they are discussed. It must be taken into account that the result of this study is also applicable to the design of the TRITIUM-IFIC 2 prototype.

#### 6.2.4 Fiber Diameter Effect

A third test was carried out to check the effect of the fiber diameter in the tritium measurement. For this test, the same simulation explained in section 6.2.3 was used, where a fiber length of 20 cm was choseen. Two different diameters were taken into account in this study, 1 mm and 2 mm, which are the commercial options given by Saint-Gobain company.

It doesn't have sense to test it with the tritium source since its efficiency will scale with the active surface of the scintillating fiber. However, an interesting study can be performed to check how the fiber diameter affect to the cosmic detection in the fiber. It is an important result as the background-signal ratio is mainly affected by the cosmic ray events.

For this task, the tritiated water source was replaced by a cosmic ray source, which was generated using the CRY library<sup>1</sup> [(NA], [Hag07]. The CRY library is a package based on objected-oriented technology and implemented in the C++ programming language. This library is used to generate cosmic-ray shower distributions for different particles (muons, neutrons, protons, electrons, photons and pions) with several options that can be varied like several altitude.

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<sup>1</sup>CRY library, Cosmic-Ray Shower library

The cosmic sources shape used in this simulation is a horizontal square of  $1 \cdot 1 \text{ m}^2$  located at a height of 35 cm (above the detector) with the typical distribution of cosmic particles at sea level.

The result of this simulation are shown in section 8.1.4, where they are discussed.

### 6.2.5 Simulation of the Tritium-IFIC 2 Prototype

The Tritium-IFIC 2 prototype simulation was the last simulation developed in the TRITIUM experiment and this was the one I mainly focused on. It consists of 800 equispaced fibers distributed in sixteen different circles with increasing radius, which are shown in Figure 6.1. The fibers simulated has a diameter of 1 mm and the optical properties, mentioned in section 6.1, was included.

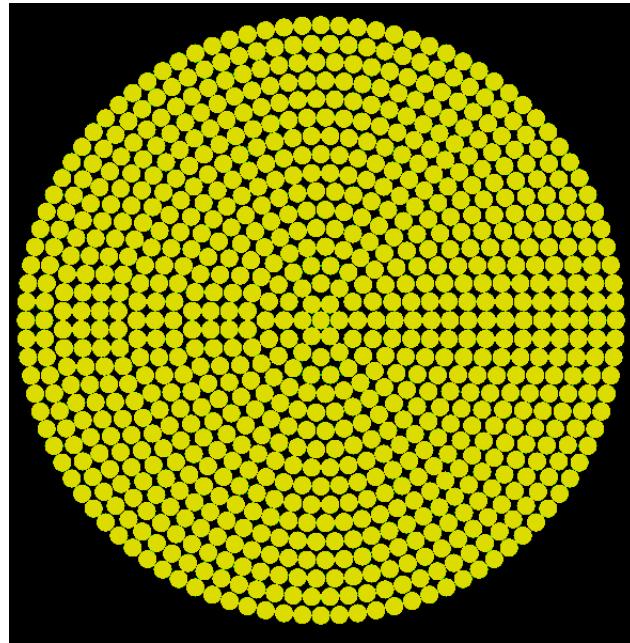
The tritiated water source used consists of a tritiated water volume with a thickness of  $5 \mu\text{m}$  around each scintillating fiber.

Scintillator fibers are located inside of a teflon vessel, which was simulated with the dimensions mentioned in section 5.2.2. Two PMMA windows with a thickness of 5 mm were simulated and located in both fiber ends and a optical grease layer with a thickness of 0.5 mm was included in each PMMA windows.

Finally, two PMTs, model R8520-460 from Hamamatsu company [K.K19], were simulated in both ends.

The optical properties used for the tritiated water, teflon vessel, PMMA windows and the optical grease, mentioned in section 6.1, are included in this simulation.

The simulation of TRITIUM-IFIC 2 is shown in Figure 6.2 in which

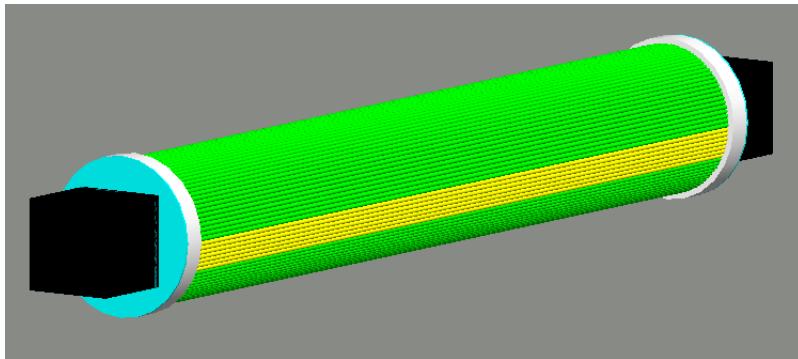


**Figura 6.1** – Distribution of the scintillating fibers in the simualtion of Tritium-IFIC 2 prototype.

can be appreciated the PMTs (black), the optical grease (blue), PMMA windows (white), tritiated water (green) and scintillating fibers (yellow). In this image, the Teflon container was not drawn to allow its interior to be seen and several volumes of tritiated water were also not included to allow several scintillation fibers to be seen.

As can be seen in this figure, the used PMTs don't cover the entire active area formed by the scintillating fiber bundle. It's not a problem for the Tritium detector since its final version will not include this photosensors. The final version of Tritium-IFIC 2 prototype will use SiPM arrays and the PMTs model used in the Tritium-Aveiro 0 prototype are circular PMTs with which the full active area is covered.

The simulation of the Tritium-Aveiro 0 prototype is similar to this since the design of both detectors are quite similar. There are two main



**Figura 6.2** – Simualtion of Tritium-IFIC 2 prototype. PMTs (black), the optical grease (blue), PMMA windows (white), tritiated water (green) and scintillating fibers (yellow)

difference between both simulated prototypes:

1. The diameter of the fibers used, which is 1 mm for Tritium-IFIC 2 prototype and 2 mm for Tritium-Aveiro 0 prototype. As the internal volume of the teflon vessel is filled, this difference imply a difference number of the scintillating fibers used, causing a difference in the signal-background ratio.
2. The photosensors used since, although both are PMTs, the model of the used PMTs is different and it cause a different active area readout, affecting to the tritium detection efficiency.

The results obtained with the simulation of the TRITIUM-IFIC 2 prototype are shown in section 8.2.1, where they are discussed.

### 6.2.6 Simulation of the Lead Shielding and Cosmic Veto

Finally the lead shielding and active vetos, detailed in section 6.2.5, were included in the simulation of the Tritium-IFIC 2 prototype. The objective

of these simulations was to quantify its effect in reducing the cosmic events detected by the prototype, demonstrating its need.

For this task, similar to that done in section 6.2.4, the tritium source was replaced by the cosmic events source, which was simulated through the CRY library.

As can be seen in Figures 3.23a and 3.23b, two plastic scintillators were simulated with the dimensions mentioned in section 3.4.2 and located above and below of the Tritium-IFIC 2 prototype simulated.

The optical properties included to this plastic scintillators are the refractive index, the light attenuation spectrum and energy emission spectrum, the values of which were obtained from their data sheet provided by the manufacturer [Cry20].

As shown in this figure, two PMTs, model R8520-460 from Hamamatsu company, were simulated to read each plastic scintillator, similar to that presented in section 3.4.2.

Finally, a lead shielding was simulated, whose properties were taken from the Geant4 NIST database. The dimensions of the simulated lead shielding were  $60 \cdot 60 \cdot 70 \text{ cm}^3$ , which is the minimum needed to accommodate an active veto and Tritium detector module inside. The length of the simulated lead castle, 60 cm, is smaller than real dimension, 148 cm. The reason for this is that only one tritium detector module was simulated, so the dimension of the lead shielding can be reduced to optimize simulation time and computing resources.

The results of these simulation are shown in section 8.2.2, where they are discussed.

# Chapter 7

## TRITIUM Monitor Results and Discussion

This chapter shows and discusses the experimental results obtained with the different prototypes developed in the TRITIUM experiment. This is divided into two sections, according to the place where they were carried out. The first section shows the measurements obtained at the University Laboratories, where external atmospheric conditions, such as temperature, can be controlled. The second section shows the measurements obtained at the Arrocampo dam, the final TRITIUM monitor emplacement, where the control of external atmospheric conditions is less precise.

### 7.1 Results from Laboratory Measurements

This section shows, compares and discusses the experimental results obtained with the different prototypes developed in the TRITIUM experiment during their installation in the laboratory. The facilities used for this task

were the Nuclear Radiation Laboratory, at the IFIC, Valencia, the DRIM<sup>1</sup>, at the University of Aveiro, and the LARUEX<sup>2</sup> laboratory in Extremadura.

### 7.1.1 Experimental Results of TRITIUM-IFIC 0 Prototype

This section shows the measurements obtained with the TRITIUM-IFIC 0 prototype during its installation in the Nuclear Radiation Laboratory at IFIC, the design of which was explained in section 5.1.1.

As stated in section 5.1.1, a statistically significant number of events was not obtained when the prototype was read with two PMTs in time coincidence. To overcome this problem, a single PMT measurement was taken using the electronic chain configuration shown in figure 3.14a. The energy spectra were measured for both, the signal and background prototypes, which are shown in Figure 7.1a. As it was mentioned in section 5.1.1, the signal prototype was filled with a tritiated water solution with an activity of 99.696 kBq/L and the background prototype was filled with ultrapure water.

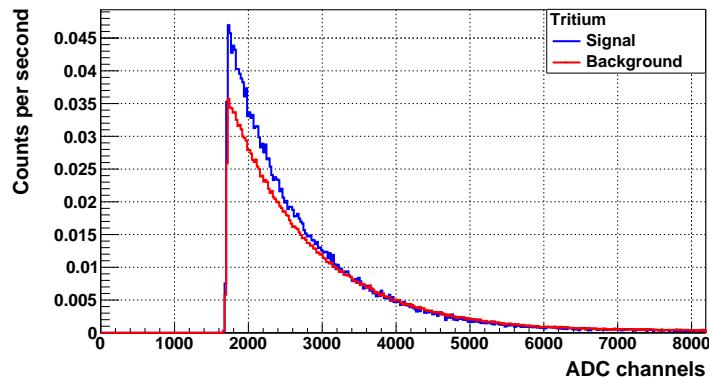
In this figure, a difference between both energy spectra is clearly visible, which correspond to the energy spectrum of tritium, Figure 7.1b. A number of counts per second of  $2.27 \pm 0.06$  and  $2.06 \pm 0.06$  was obtained for the both measurements, signal and background, respectively. Therefore,  $0.21 \pm 0.085$  counts per second was obtained for the tritiated water source used.

The tritium detection efficiency obtained for this prototype is  $(2.11 \pm$

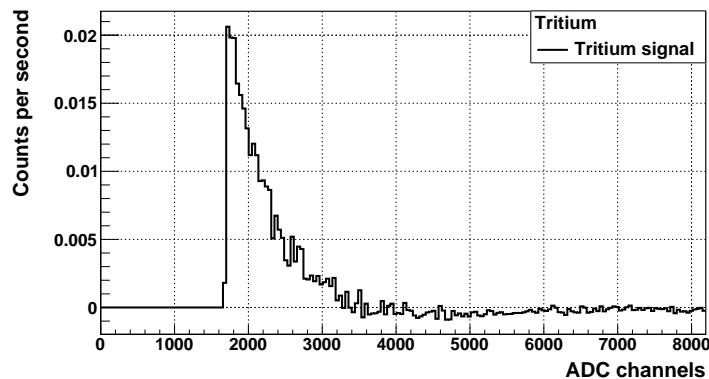
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<sup>1</sup>DRIM, Deteção da Radiação e Laboratorio Imagem Médica laboratoire (Laboratory for Radiation Detection and Medical Imaging)

<sup>2</sup>LARUEX, Laboratorio de Radiactividad Ambiental de la Universidad de Extremadura



(a) Signal and background energy spectra.



(b) Tritium energy spectrum.

**Figura 7.1** – Energy spectra experimentally measured with TRITIUM-IFIC 0 prototype.

$0.85) \cdot 10^{-3} \frac{\text{c/s}}{\text{kBq/L}}$ , which is calculated from the quotient of both, the counts per second measured and the specific activity of the tritium liquid source used.

Comparing with the detectors developed so far by other experiments, Table 2.2 the efficiency obtained is of the order of the detectors the worst results, obtained by Moghissi and Muramatsu.

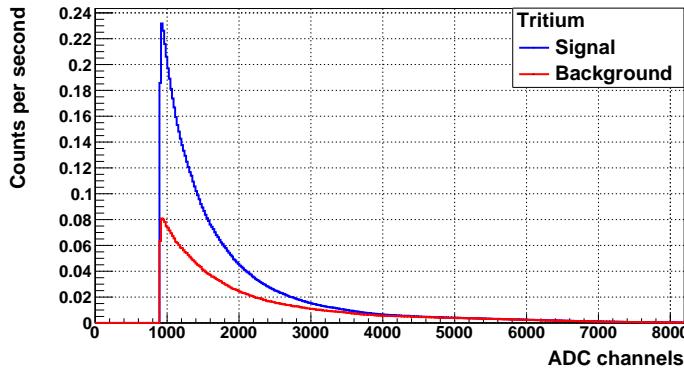
As we explained in section 2.1, the efficiency of scintillating detectors scales with the active area of the scintillator used. Therefore, to compare the efficiency with other detectors and with other prototypes developed in TRITIUM experiment, the specific efficiency of this prototype is calculated, the value of which is  $(9.59 \pm 3.88) \cdot 10^{-6} \frac{\text{c/s}}{\text{kBq/L cm}^2}$ .

As can be seen in the Table 2.2 the specific efficiency is a little bit larger than the detectors with the worsen specific efficiency developed up to now, Muramatsu and Moghissi. This fact can be explained with the loss of photons produced in the curve of the fiber bunch, discussed and experimentally demonstrated in section 5.1.1.

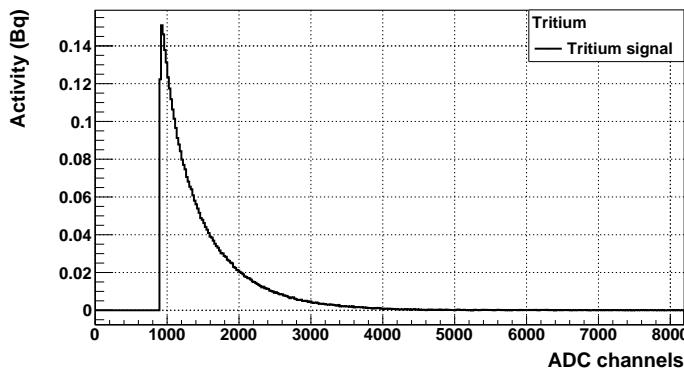
### 7.1.2 Experimental Results of TRITIUM-IFIC 1 Prototype

This section shows the results obtained with the TRITIUM-IFIC 1 prototype during its installation in the Nuclear Radiation Laboratory at IFIC. Its design is explained in section 5.1.2, which includes several improvements, such as a teflon vesel and new straight arrangement of scintillating fibers, which were found to be a problems in the previous prototype, reducing its efficiency.

The energy spectra of both, signal and background, were measured with TRITIUM-IFIC 1 prototype, which is shown in Figure 7.2a. As it was



(a) Signal and background energy spectra.



(b) Tritium energy spectrum.

**Figura 7.2** – Energy spectra experimentally measured with TRITIUM-IFIC 1 prototype.

mentioned in section 5.1.2, to measure the signal, the prototype was filled with a tritiated water solution with an activity of 99.696 kBq/L and, to measure the background, the prototype was filled with ultrapure water.

As can be seen, the difference between both energy spectra is clearly visible, which corresponds to the tritium energy spectrum, Figura 7.2b. Furthermore, compared to the energy spectra obtained with the previous prototype, Figure 7.1a, the difference between both is larger, which implies that the tritium detection efficiency of this prototype was improved.

This efficiency improvement can be quantified using similar calculations than those used in the previous section. The number of counts per second measured for signal and background, both, are  $7.82 \pm 0.11$  and  $3.99 \pm 0.08$  respectively. Therefore,  $3.83 \pm 0.13$  counts per second were obtained for the tritiated water source used.

The tritium detection efficiency of TRITIUM-IFIC 1 is  $(3.84 \pm 0.16) \cdot 10^{-2} \frac{\text{c/s}}{\text{kBq/L}}$ , which is calculated using the same quotient explained in the previous section. As can be seen, the efficiency obtained for this prototype is larger than that obtained for the previous prototype, TRITIUM-IFIC 0. It is an expected result since this prototype uses a larger active area. The specific efficiency is calculated to eliminate the effect of different active areas between both prototypes and quantify the improvement in the efficiency due to modifications in the design. The specific efficiency obtained is  $(9.56 \pm 0.40) \cdot 10^{-5} \frac{\text{c/s}}{\text{kBq/L cm}^2}$ .

Therefore it was verified that the specific efficiency of this prototype was improved by a factor of ten due to the modifications applied to its design which confirms and quantifies the usefulness of these modifications. Furthermore, compared with scintillating detectors developed in other experiments, table 2.2, on the one hand, the efficiency of this prototype is very close to the best result, obtained for Singh, and, on the other hand, the specific efficiency, which is a most relevant value to compare, is almost 5 times larger than the best results, obtained for Hofstetter.

It must be taken into account that in the first two prototypes the Low Detection Level, LDL, was not studied, since their objective was to improve its design and find the problems that reduced their efficiency. In fact, it can be seen in Figure 7.2 that the activity used is further to be the LDL of the TRITIUM-IFIC 1 prototype. The LDL was only studied in the final prototypes.

### 7.1.3 Experimental Results of TRITIUM-Aveiro 0 prototype

This section shows the results obtained with the TRITIUM-Aveiro 0 prototype during its installation in Aveiro and Extremadura laboratories. The design of this prototype is shown in section 5.2.1.

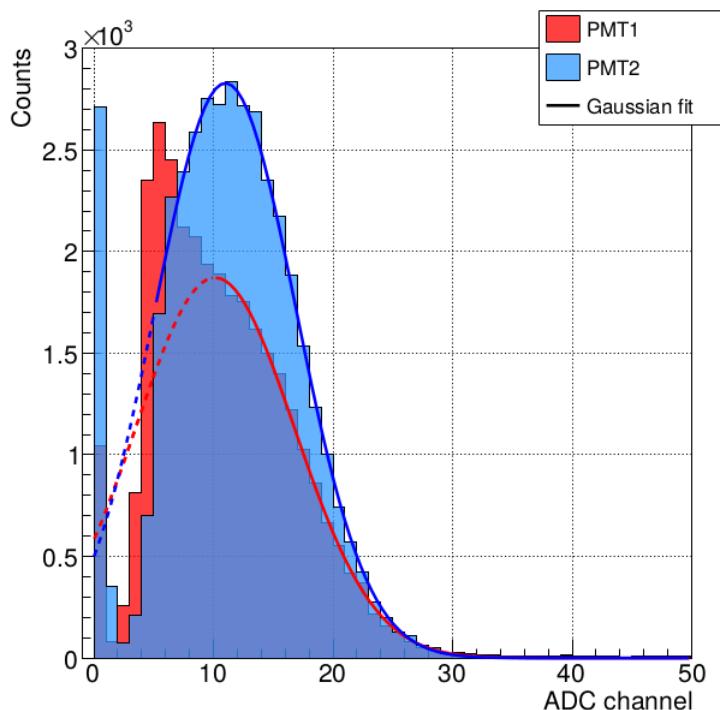
This prototype was first installed in the DRIM laboratory, at the University of Aveiro, where the first measurements were taken. These measurements were used to find and solve the problems, learn about low energy detection and develop a functional scintillation prototype for TRITIUM.

First, the energy distribution of a single photon was measured from the self-emission of PMTs (dark current). To avoid the environmental light detection, the TRITIUM-Aveiro prototype was removed and the measurement was carried out only with the PMTs, the windows of which were covered with black caps. The output signals of the PMTs were digitalized, shaped and pulse-height measured by a CAEN V1724 digitalizer [CAEb].

The single-photon energy distribution of both PMTs is shown in Figure 7.3 in which a gaussian function was fitted. Due to the electrical noise of the PMT, an extrapolation (dashed line) was needed to be applied.

As can be seen, the distribution obtained with PMT1 deviates from the Gaussian function due to the higher noise in the low energy channels. This fact indicates that it could be interesting to use PMT with very low background for future prototypes.

As DRIM laboratory was not equipped to work with liquid radioactive source such as tritiated water, the first measurements were taken with a  $^{55}\text{Fe}$  radioactive source since the energy of its  $\gamma$  emission, 5.9 keV, is very close to the energy of tritium electrons. To do so, the TRITIUM-Aveiro 0 prototype was coupled to both PMTs using optical grease and, due to its

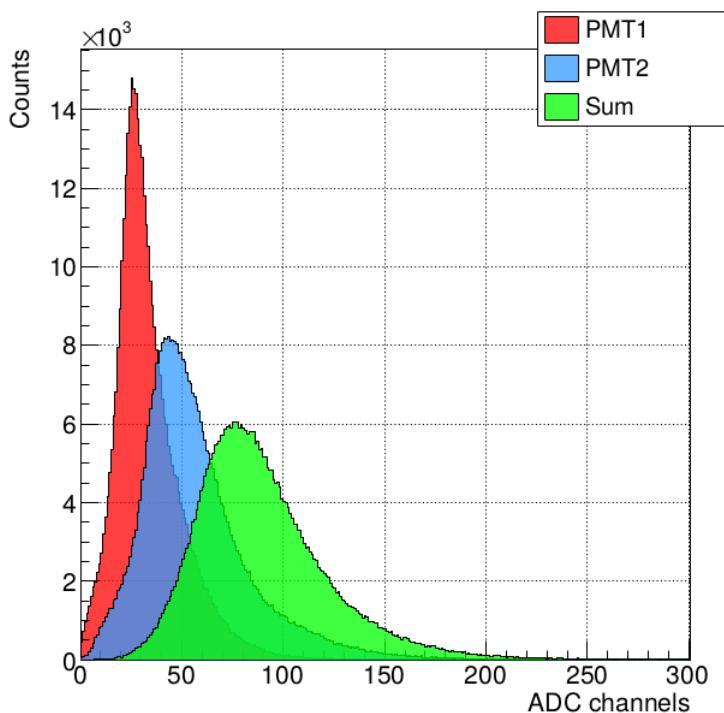


**Figura 7.3** – The single-photon energy distribution of both PMTs used in the TRITIUM-Aveiro 0 prototype and their sum [CA].

low mean free path in solid materials, the radioactive source was placed inside the teflon vessel. This prototype was not filled with water because of the presence of the radioactive source. This measurement is shown in Figure 7.4.

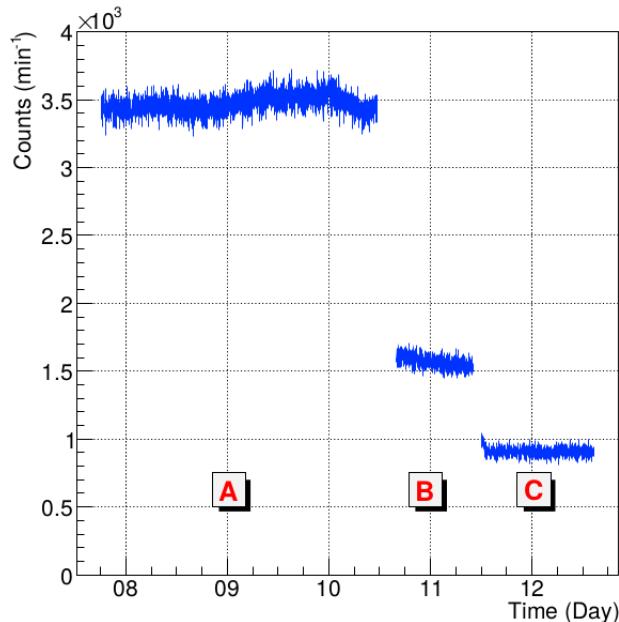
A shift to the right side is observed for the PMT2 data, which is produced because this PMT has a higher gain and the radioactive source was placed closer to it, reducing the attenuation of the photons.

Lastly, a passive shield test was performed in the DRIM laboratory to quantify the attenuation of the background produced by lead. To do so, the  $^{55}\text{Fe}$  radioactive source was removed and the electronical chain explained in appendix D was used in counting mode for this test.



**Figura 7.4** – Measurement of a  $^{55}\text{Fe}$  radioactive source with the TRITIUM-Aveiro 0 prototype [CA].

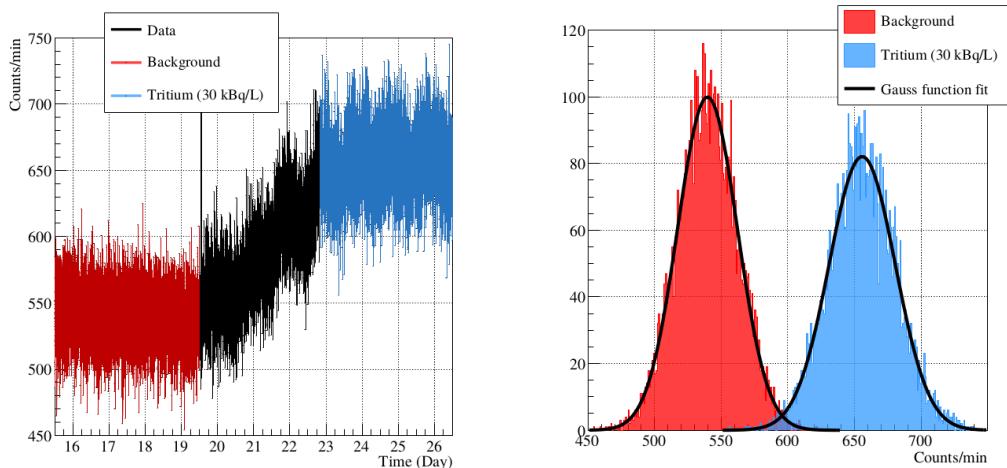
The measurements, shown in Figure 7.5, were carried out in three different situations. The first, region A, in which the measurement was performed without using any lead foil, the second, region B, in which lead foil with a thickness of 2.5 mm was used and the third, region C, in which another lead foil layer with the same thickness (total thickness of 5 mm) was used.



**Figura 7.5** – Measurement of the background with TRITIUM-Aveiro 0 prototype covered with different thicknesses of lead [CA].

As can be seen, in the region A, the average of the data adquired during 2.5 days is  $3.5 \cdot 10^3$  counts/min (58 counts/sec). In the region B a reduction of more than two times was observed, measuring an average of  $1.6 \cdot 10^3$  counts/min (26 counts/sec). In the region C a reduction of about 4 times relatively to the region A is observed, measuring an average of  $0.9 \cdot 10^3$  counts/min (15 counts/sec).

It has to be taken into account that the case C is the real situation that is present in Arrocampo since, as it has been shown in section 3.4.2,



(a) Counts per minut measured as a function of time.

(b) Distribution of the acquired data.

**Figura 7.6** – Measurements of the background and tritium liquid source (with an activity of 29.8 kBq/L) performed with the TRITIUM-Aveiro 0 prototype and integred during a minute [CA].

the thickness of the lead shielding ise 5 mm.

As it was said, the DRIM laboratory was not equipped to work with a liquid radioactive source such as tritiated water, so this prototype was installed in the LARUEX laboratory, at the University of Extremadura to finalize with the characterization measurements.

First, the background of the prototype was measured during 4 days. For this task, the prototype was filled with ultrapure water and covered with lead bricks with a thickness of XXX. The time of each measurement is 1 minut and the data is shown in Figure 7.6a and 7.6b in which a gaussian fit was done.

As a result of the gaussian fit, an average ( $N_B$ ) of 540 counts/min and standard deviation ( $\sigma_{Nb}$ ) of 22.61 counts/min was obtained. To calculate the Minimum Detectable Activity (MDA), the detection limit concepts

developed by Lloyd A. Currie [Cur68] was applied. With this concepts, the minimum net counts with the probability of a false-negative less than a 5%,  $N_D$ , and minimum net currents with the probability of a false-positive less than a 5%,  $L_C$ , called critical level, are calculated using the equations:

$$L_C = 2\kappa\sigma_{Nb} = 53 \text{ counts/min} \quad (7.1)$$

$$L_D = \kappa^2 + 2L_C = 108 \text{ counts/min} \quad (7.2)$$

Both values refer to the net counts per minute after background subtraction, so,  $L'_C$  and  $N'_D$  referred to the detector signal (before background subtraction) are 593 and 648 counts/min respectively.

To find the MDA associated to this  $N'_D$ , tritiated water was slowly added so that the tritium water activity increased continuously up to an average of  $656 \pm 0.43$  counts/min. Then, the activity of this solution was measured with a Quantulus system, obtaining a MDA of 29.8 kBq/L.

The tritium detection efficiency can be calculated from the quotient of the net tritium counts per second measured,  $1.93 \pm 0.58$  counts/sec, and the activity of the tritium source used. The efficiency obtained is  $(6.49 \pm 1.94) \cdot 10^{-2} \frac{\text{c/s}}{\text{kBq/L}}$ . The value obtained with TRITIUM-Aveiro 0 prototype is larger than the efficiency reported by other similar experiments, Table 2.2. This is also larger than the efficiency obtained with previous TRITIUM prototypes, an expected result since the active area of this prototype is on order of magnitude larger.

The specific efficiency is calculated to compare with other scintillating detectors, the value of which is  $(1.59 \pm 0.48) \cdot 10^{-5} \frac{\text{c/s}}{\text{kBq/L cm}^2}$ . Comparing with the specific efficiency obtained with scintillating detectors developed in other experiments, Table 2.2, the value obtained for TRITIUM-Aveiro 0 prototype is close to the largest specific efficiency, obtained by Hofstetter. However this prototype has a worsen specific efficiency than other

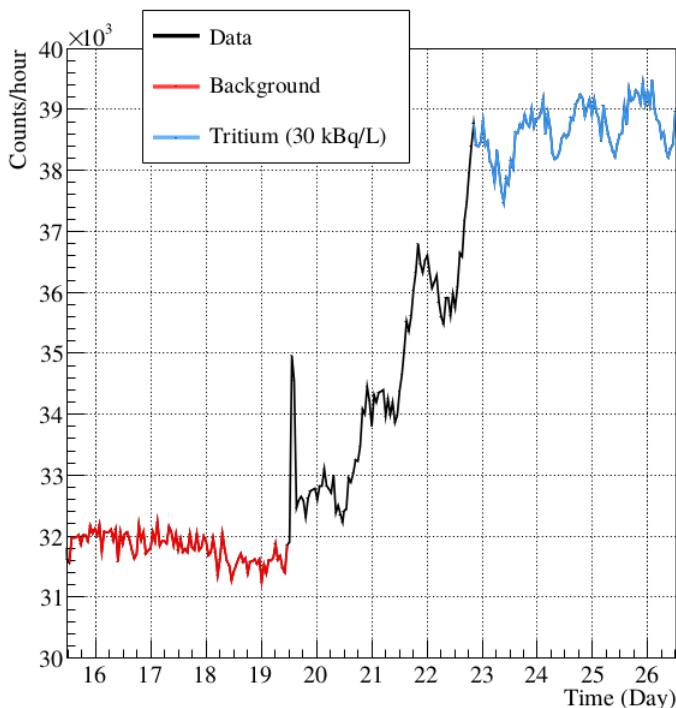
prototypes developed in TRITIUM experiment such us TRITIUM-IFIC 1. A possible reason is that the fibers used in this prototype are not polished or cleaned. Therefore, the importance of the fiber polishing and cleaning process is again exhibited.

It can also be noted that the efficiency uncertainties obtained for this prototype are greater than those obtained in the previous TRITIUM prototypes. The reason of that is a difference in the measurement time. The measurement time used for the TRITIUM-Aveiro prototype is 1 minute, while that used for the previous prototypes is 10 minutes. Smaller uncertainties are achieved in the measured counts and, therefore, in the efficiency when longer measurements are taken.

Finally, as lower uncertainties in the measured counts allow a lower MDA to be achieved, longer measurements are studied to quantify the reduction of the MDA of this prototype. For this task, groups of 60 successive measurements are integrated, resulting in several measurements of 60 minutes. The acquired data is shown in Figure 7.7, where it can be checked a smaller relative uncertainty than the values for 1 minute.

In this case, the average and uncertainty of the measured background data are  $3.186 \cdot 10^4$  and 228 counts per hour respectively. Using the equations 7.1 and 7.2, the values of  $L_C = 530$  and  $N_D = 1043$  counts per hour are obtained respectively. Assuming linearity between the measured counts for the background and the tritiated water,  $3.872 \cdot 10^4$  counts per hour, this  $N_D$  corresponds of a MDA of 4.53 kBq/L.

A diarly oscilation is clearly observed in the Figure 7.7, showing that the measurements are affected by external light. This oscilation begins on the 19<sup>th</sup> day, where the water closed circuit pump was installed, so it is likely that the light leak is produced through this system.

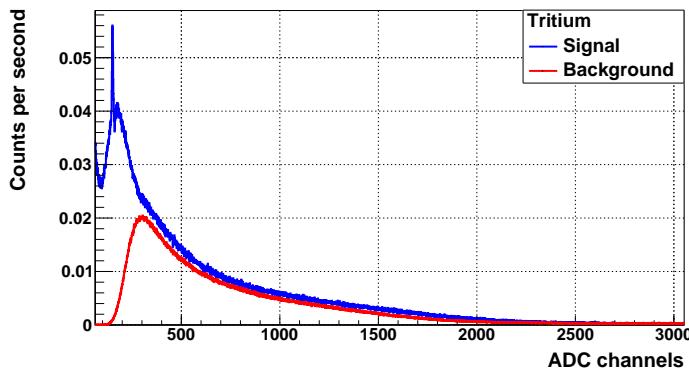


**Figura 7.7** – Measurements of the background and tritium liquid source (with an activity of 29.8 kBq/L) performed with the TRITIUM-Aveiro 0 prototype and integrated during an hour [CA].

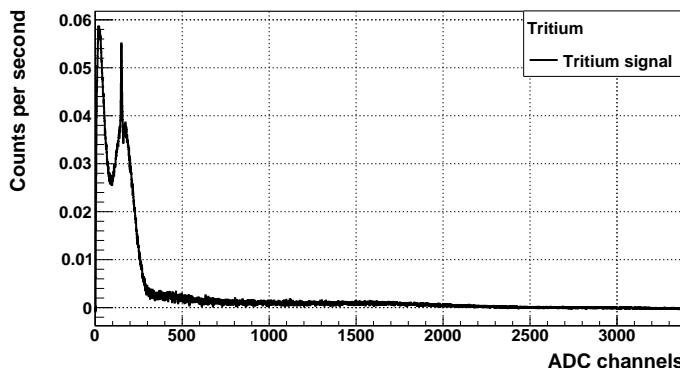
#### 7.1.4 Experimental Results of TRITIUM-IFIC 2 prototype

This section shows the results of the lastest prototype developed in the TRITIUM experiment, TRITIUM-IFIC 2, during its installation in the Nuclear Radiation Laboratory at IFIC. The design of this prototype is shown in section 5.2.2.

The energy spectra of the signal and background prototypes were measured, which are shown in Figure 7.8a. As it was mentioned in section 5.2.2, the signal prototype was filled with a tritiated water solution with an



(a) Signal and background energy spectra.



(b) Tritium energy spectrum.

**Figura 7.8** – Energy spectra experimentally measured with TRITIUM-IFIC 2 prototype.

activity of 10 kBq/L and the background prototype was filled with ultrapure water.

A difference between both signals is clearly visible, which corresponds to the energy spectrum of tritium, Figure 7.8b. The number of counts per second obtained for both, the signal and background, are  $19.05 \pm 0.18$  and  $11.54 \pm 0.14$  respectively. Therefore,  $7.11 \pm 0.23$  counts per second was obtained for the tritiated water source used.

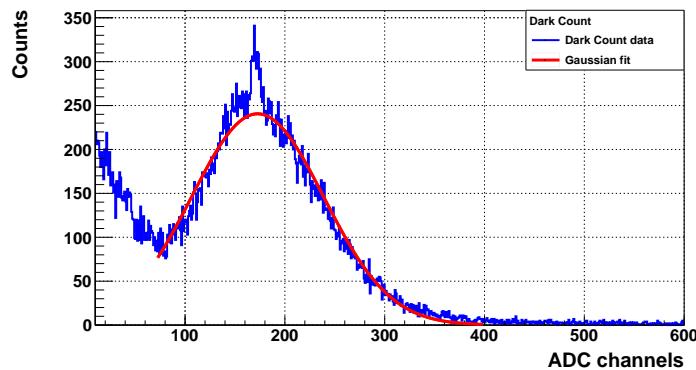
The tritium detection efficiency obtained for this prototype is  $(7.11 \pm 0.28) \cdot 10^{-1} \frac{\text{c/s}}{\text{kBq/L}}$ , calculated from the quotient of both, the counts per second measured and the specific activity of the tritium liquid source used. This efficiency is larger than all scintillating detectors developed so far, including the prototypes developed in TRITIUM experiment, Table 2.2 and sections 7.1.1, 7.1.2 and 7.1.3. It is an expected result since the active area used in this prototype is larger than those used in others.

To remove the effect of different active area, the specific efficiency is measured, obtaining a value of  $(1.59 \pm 0.48) \cdot 10^{-5} \frac{\text{c/s}}{\text{kBq/L cm}^2}$  for this prototype. Again, it can be observed that this prototype has the largest specific efficiency obtained so far with a scintillating detector used for tritium detection.

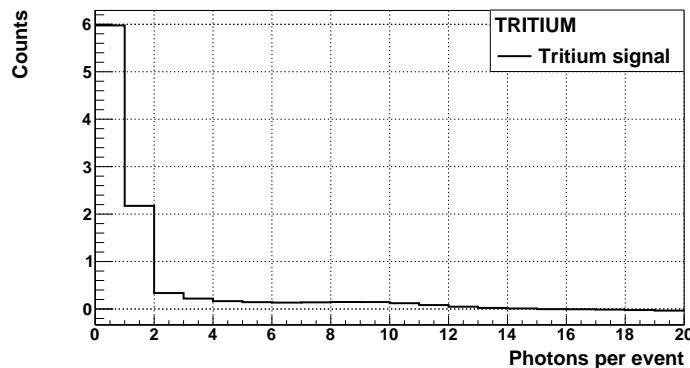
Therefore, as it has demonstrated, the intrinsic and specific efficiency obtained so far for scintillating detectors used for tritium detection has been exceeded with the last TRITIUM prototype, TRITIUM-IFIC 2.

As discussed in chapter 4, the energy spectrum is shown in ADC units, proportional to energy, since it is difficult, even impossible, to do an energy calibration for a plastic scintillator due to its large relative uncertainty in the number of photons produced per event. Nevertheless, a detector calibration can be performed to express the results in units of photons detected per event. It was carried out using the single-photon distribution of the used PMTs, which was obtained from their self-emissions. Similar to the TRITIUM-Aveiro 0 prototype, the PMT used to read this prototype was decoupled to the prototype and covered with a special black blanket to ensure that external photons did not reach the PMT. The output signal of the PMT was analyzed using the electronical chain shown in Figure 3.14a. The distribution measured is shown in Figure 7.9a, in which a gaussian function was fitted.

As can be seen, the mean and uncertainty of the signal produced



(a) Single photon distribution.



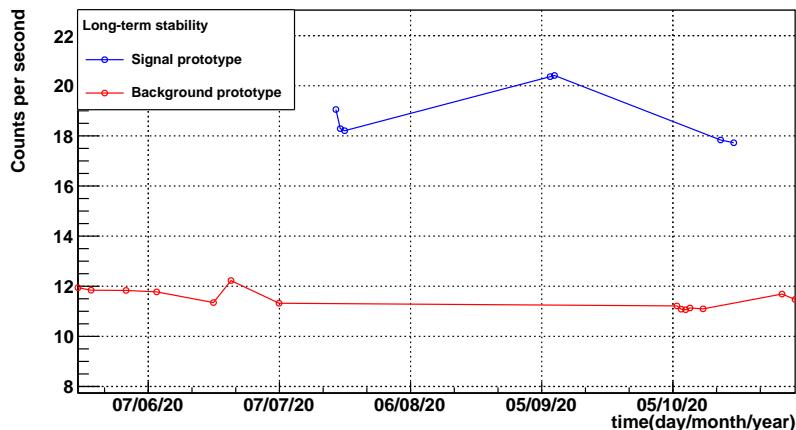
(b) Tritium energy spectrum.

**Figura 7.9** – Tritium measurement with TRITIUM-IFIC 2 prototype and expressed in photons detected per event.

for a single photon detected with the PMT used are 172.71 and 66.19 (in ADC units) respectively. Therefore, the tritium signal, Figure 7.8b, can be expressed in number of photons detected per event, Figure 7.9b, by simply dividing this spectrum by the mean measured for the single-photon distribution.

As can be seen, a maximum of 15 photons are generated per tritium event, which is in accordance with the expected result taking in to account the efficiencies involved.

Finally, a monitoring of both prototypes, signal and background, were carried out for several months, the measurements of which are shown in Figure 7.10.

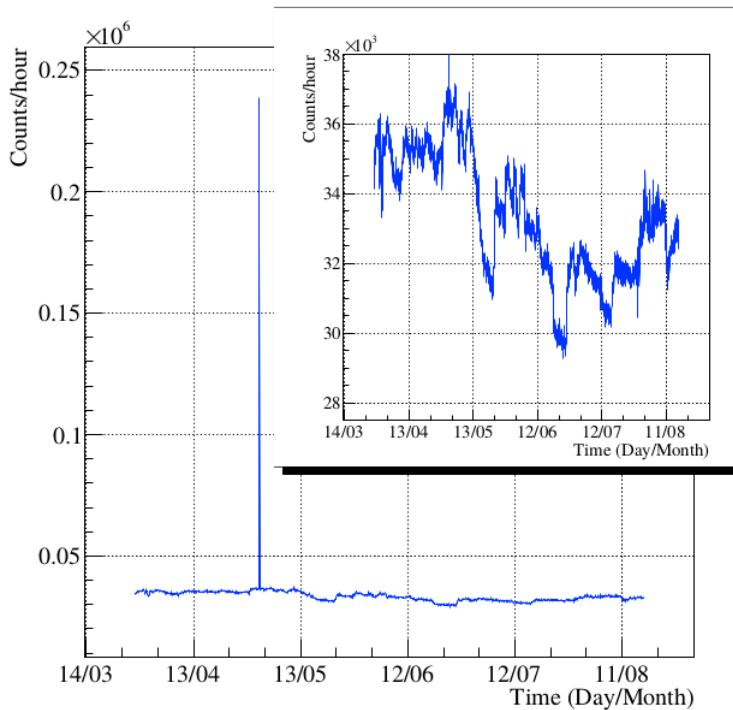


**Figura 7.10** – Monitoring of the signal and background prototypes for several months.

As can be seen, the signal of both was not reduced, ensuring the maintainance of the detector efficiency during 3 months for the signal and 6 months for the background.

## 7.2 Results from Measurements at Arrocampo Dam

This section shows the measurements obtained with the TRITIUM-Aveiro 0 prototypes during their installation in the Arrocampo dam, which has been the only prototype installed in Arrocampo dam so far. This prototype was installed and working there for more than four months, from March 27, 2019 to August 18, 2019, during which time this prototype was taking background measurements. The data adquired during this time are shown in the figure 7.11, the measurement time of which is 60 minutes.



**Figura 7.11** – Background measured with the TRITIUM-Aveiro 0 prototype during its installation in Arrocampo dam [CA].

This data shows good stability in the working months, measuring

a mean value of around 9.31 counts per second. A large peak is observed on May 2, 2019, caused by an opening of the roof of the lead shield to access the prototype. In the small box of the figure, the data is zoomed for a better visualization. The MDA measured in Arrocampo dam for 60 minute integration counting data is 6 times larger than that calculated in section 7.1.3. This variations can be caused by electric noise induced by the electric pumps of the water purification system and instabilities observed in the electronic boards.

A cosmic veto currently under development is planned to be installed and used in anti-coincidence along with two additional prototypes.

Furthermore, three TRITIUM-IFIC 2 prototypes and a cosmic veto, explained in section 5.2.2 and 3.4.2 respectively, are also planned to be installed in Arrocampo dam as soon as possible.

# Chapter 8

## Results of the TRITIUM Simulations

This chapter shows the results obtained in the simulations described in chapter 6, which is divided in two different sections. The first section contains the results of several simulations used to improve the design of the TRITIUM detector while the second exhibits the results obtained for the simulation of a full TRITIUM monitor based on an active veto read in anticoincidence with several TRITIUM-IFIC 2 prototypes. As shown throughout the different sections of this chapter, several tests were carried out to verify the correct simulation of the different steps such as the simulated tritium source, the energy deposition in the fibers and the production of photons in them.

## 8.1 Optimization of the TRITIUM Monitor Design

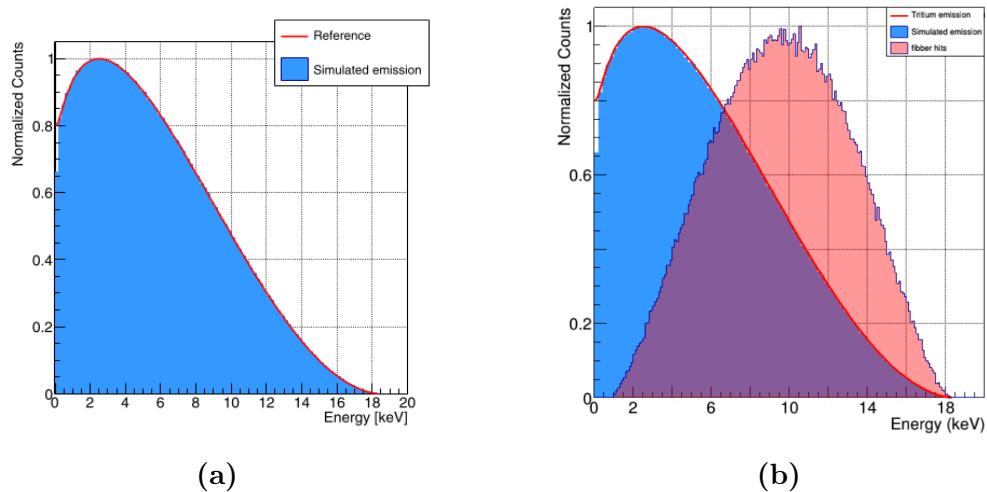
Several simulations were performed during the design of the TRITIUM detector, which were used to quantify the effect of some modifications and to choose those that optimize the tritium detection. The characteristics studied are the diameter and length of the scintillating fibers. As the tritium electrons has a very low mean free path, the shape of the simulated tritium source was also studied to reduce the consuming time and computing resources.

### 8.1.1 Optimization of the Tritiated Water Source

This section show and discuss the results obtained with the simulation described in section 6.2.1, the objective of which is to find the radial thick of the simulated tritium source that reduce the consuming time and computing resources used in the simulations.

First, the initial energy of the simulated tritium events are verified. For this task, the energy distribution of the simulated tritium electrons is obtained, shown in figure 8.1a, and compared with that obtained in the reference [Mer15]. As can be checked, there is a good agreement between both.

In addition, a spectrum of the initial energy of tritium electrons that are capable of reaching the fiber and depositing energy are shown in Figure 8.1b, red histogram, which is compared to the energy distribution of all simulated tritium events, blue histogram. A shift to the right side in the red histogram is observed, creating a peak centred at 10 keV. This shift occurs mainly because the lower energy tritium electrons don't have enough energy to reach the fiber and overcome the water-fiber interface, producing

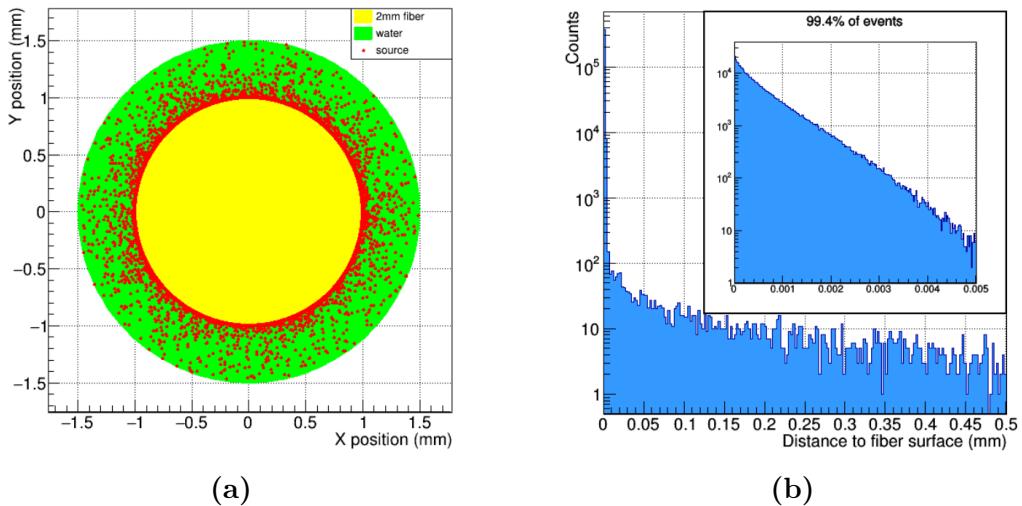


**Figura 8.1** – Energy distribution of a) simulated tritium decays b) Initial energy of tritium decays that reach the scintillating fibers (red histogram) compared the all simulated tritium events (blue histogram) [Aze20].

a non-detected tritium event.

Regarding the radial thickness of the tritiated water source, Figure 8.2a shows a transversal cut of the 2 mm scintillating fiber, yellow, the simulated tritium source 0.5 mm thick around the fiber, green, and the tritium decays the electrons of which has deposited their energy in the scintillating fiber, red dots. Furthermore, the distribution of the radial distance between the position where tritium decays take place and the surface of the scintillating fiber are shown in figure 8.2b.

As can be seen in both figures, most of the tritium decays that are detected occur in close proximity to the scintillating fiber. A zoom is applied in the inset box of the Figure 8.2b for better viewing. The chosen thickness of the simulated tritium source is  $5 \mu\text{m}$  since the 99.4% of the events that are able to deposit energy in fibers are produced at least of this distance.



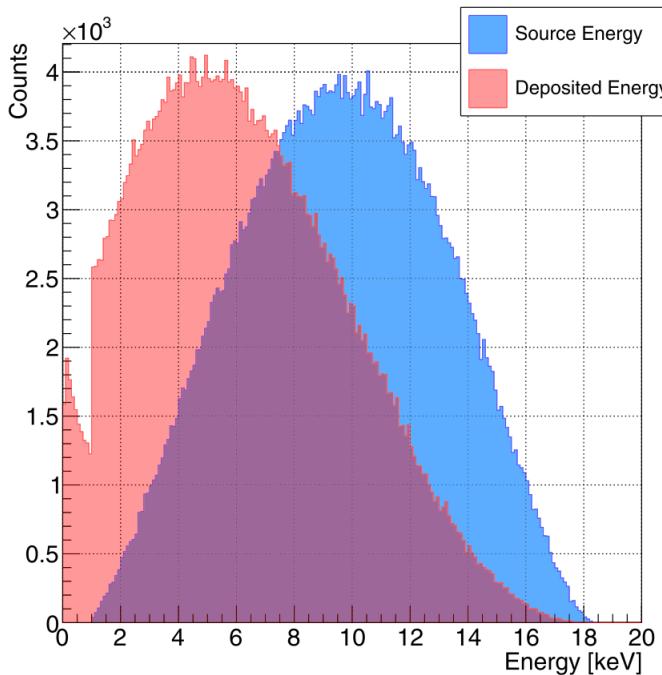
**Figura 8.2** – a) Transversal cut of simulated scintillating fiber (yellow) and tritium source (green) with various tritium decays (red dots) b) Distribution of the radial distance between the position where the tritium decay takes place and the surface of the scintillating fiber [Aze20].

### 8.1.2 Simulation of the Output Light of Scintillating Fibers

As tritium electrons are far from being a MIP a quenching effect is produced in the output light generated by scintillating fibers, following the Birks law (equation 6.1). This quenching effect produces a reduction in the photons emitted by the scintillating fibers. The objective of this section is to quantify the significance of this effect and how it affects to the tritium detection.

First, a verification test of the energy deposition of tritium electron on scintillating fibers is carried out. In Figure 8.3 the initial energy of simulated tritium electrons that has reach the scintillating fibers, blue histogram, is compared with their energy deposited in scintillating fibers, red histogram.

A shift to the left side of the spectrum (smaller energies) is ob-

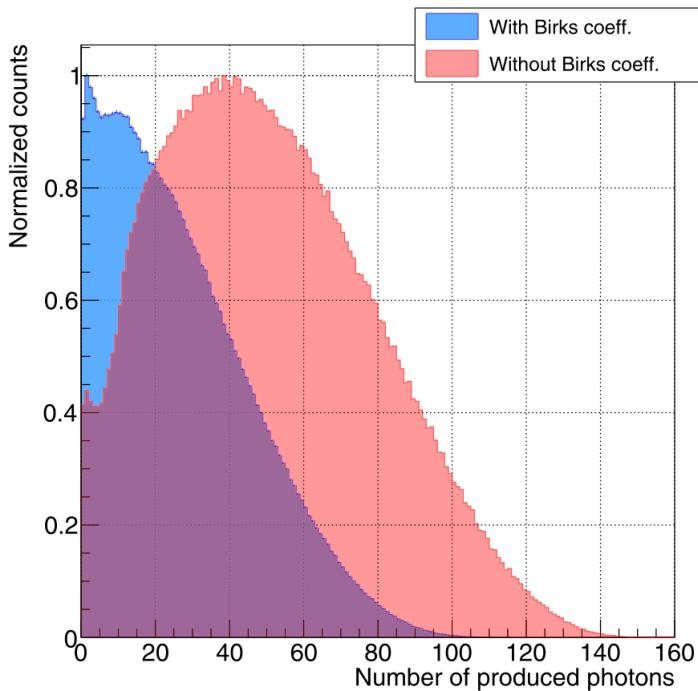


**Figura 8.3** – Distribution of the initial energy of tritium events that has reach the scintillating fibers, blue histogram, and the energy deposited, red histogram [Aze20].

served between the blue histogram, with a peak around 10 keV, and the red histogram, with a peak around 5 keV. This displacement is mainly caused by the loss of energy of tritium electrons in the water. A cut of around 1 keV is observed in both energy distributions, produced by the default energy threshold of 990 eV that exist in the G4EmLivermorePhysics physics list.

Figure 8.4 shows two distributions of number of photons produced in scintillating fibers by tritium events, one in which the quenching effect has not been considered ( $k_B = 0$ ), red histogram, and other in which the Birks coefficient has been applied ( $k_B = 0.126 \text{ mm/MeV}$ ), blue histogram.

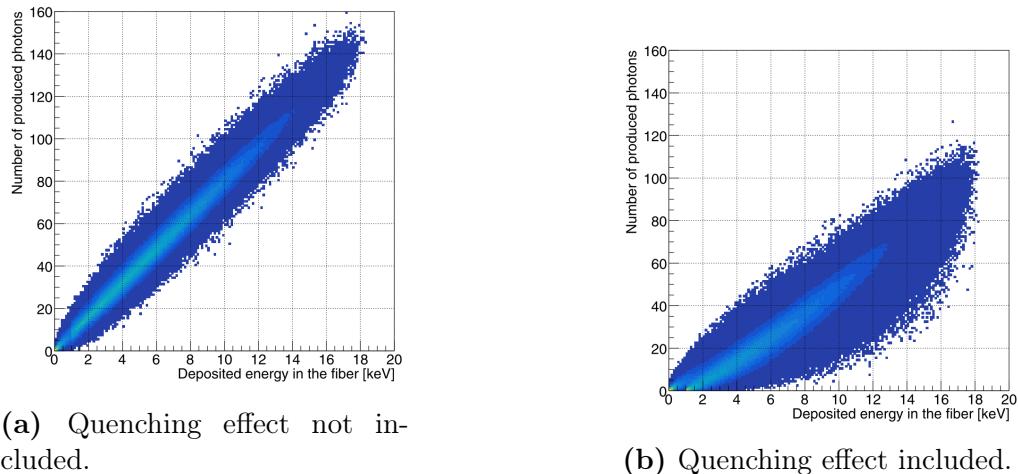
As expected, a distribution with a peak of around 40 photons per



**Figura 8.4** – Energy distribution of photons produced by the scintillating fiber when the birks coefficient is not considered, red histogram, and when this is considered, blue histogram [Aze20].

tritium event and a maximum of around 150 photons is obtained when the quenching effect was not considered. A significantly reduction of the output light is observed when the Birks coefficient is taken into account, producing a distribution with a peak centred around 10 photons and a maximum of 110 photons. The quenching effect is also observable in Figure 8.5, where the number of produced photons as a function of the energy deposited in the fiber is displayed in a bidimensional plot.

In this figure, in addition to a reduction of number of photons produced per energy deposited, a broader distribution is obtained when the Birks coefficient is considered, showing an increasement of the fluctuations of energy deposition.



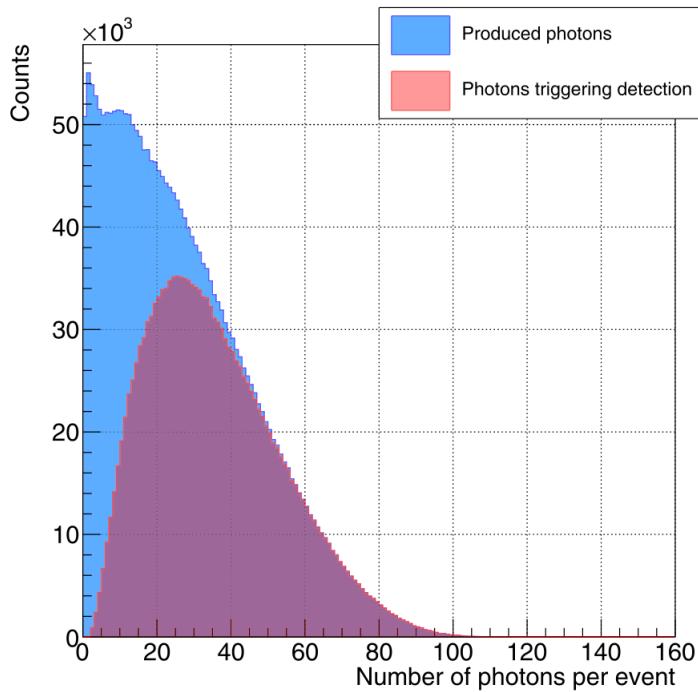
**Figura 8.5** – Number of photons produced in front of the energy deposited in the scintillating fibers when a)the birks coefficient is not considered ( $k_B = 0$ ) b) the Birks coefficient is considered ( $k_B = 0.126 \text{ mm/MeV}$ ) [Aze20].

### 8.1.3 Optimization of the Scintillating Fiber Length

A study was carried out to choose the length of scintillating fibers that optimize the tritium detection efficiency. For this task, the complete TRITIUM-Aveiro 0 prototype was simulated, in which the photon propagation was included.

First, the propagation of photons in scintillating fibers was studied. The blue distribution of Figure 8.6 shows the number of photons produced in the fiber per tritium event that reaches them and the red distribution shows the same information but only for those events detected for both photosensors in time coincidence.

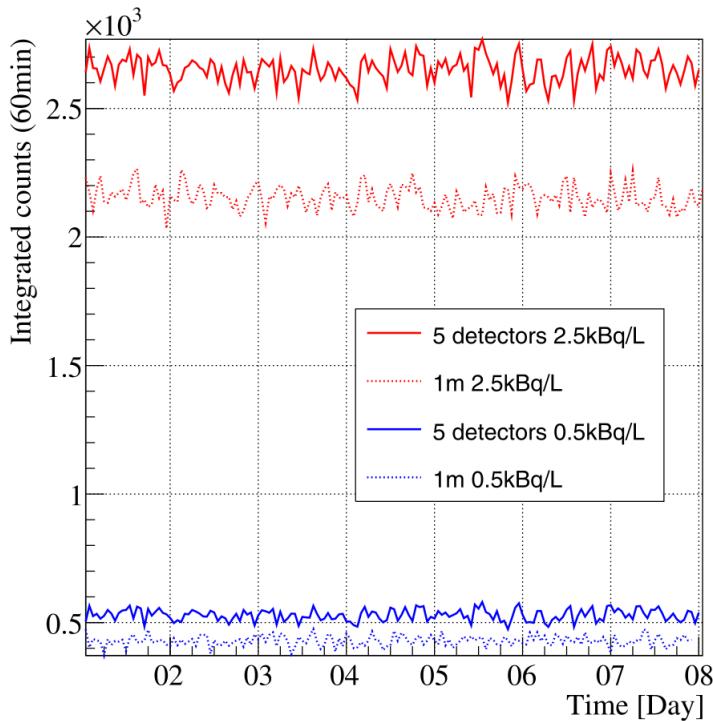
It can be seen that tritium events that produce a high number of photons are practically always detected but most of the events with fewer photons produced in the fibers are not detected, producing a peak centred of around 25 photons.



**Figura 8.6** – Number of photons produced in the fiber per tritium event for all tritium events that reach the fiber (blue histogram) and only for tritium events the photons of which are detected by photosensors (red histogram) [Aze20].

Regarding the fiber length study, two different lengths were compared, 1 m and 0.20 m, and two different tritium source activity were used, 0.5 kBq/L and 2.5 kBq/L. As detected tritium counts is proportional to the active area, 5 detectors were simulated for the case of 0.20 m fiber length to normalize the study to the same active area. The counts, which were integrated over 60 min and taken over a week, are shown in Figure 8.7.

A larger signal is seen for shorter fiber lengths in both cases, producing a increasement in tritium detection efficiency of approximately 25%, principally caused by a lower absorption of photons in shorter scintillating fibers and the leakage of some photons due to a non-perfect photon collec-



**Figura 8.7** – Counts integrated over 60 min, normalized to the same active area and taken over a week for a fiber length of 1 m, dashed lines, and 20 cm, solid lines and two different activities, 0.5 kBq/L, blue lines, and 2.5 kBq/L, red lines [Aze20].

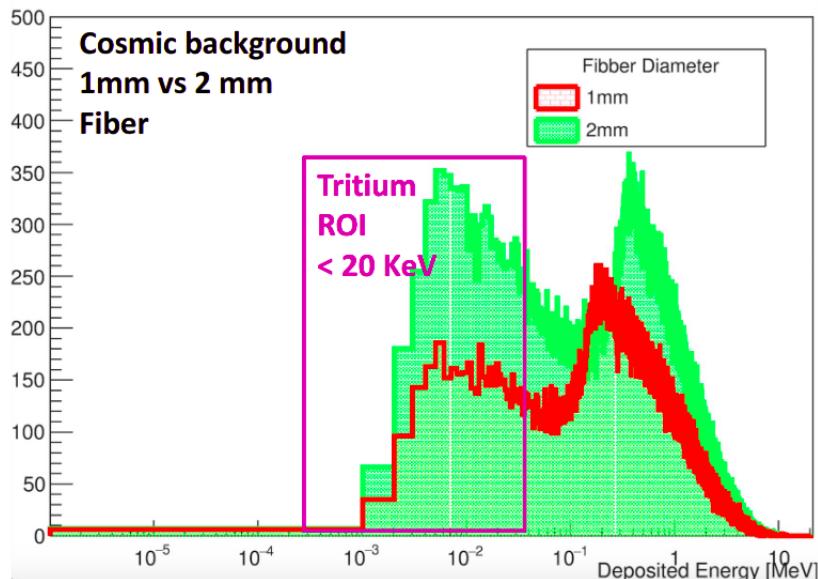
tion in the fiber.

#### 8.1.4 Optimization Scintillating Fiber Diameter

This study quantifies the effect of the scintillating fiber diameter on the detection of cosmic ray events. The energy deposited in the scintillating fiber by a cosmic ray event is proportional to the active volume crossed, which is larger for 2 mm fibers. Therefore, the cosmic ray signal obtained for a measured cosmic event will be larger for a detector based on 2 mm

diameter. The objective of this study is to find the design with which a lower background is obtained in the region of interest of tritium detection, ROI (up to 18 keV).

The distribution of energy deposited in scintillating fibers by cosmic ray events are shown in figure 8.8 for both cases, 1 mm and 2 mm fibers.



**Figura 8.8** – Comparison of the energy deposition of cosmic ray events in scintillating fibers of 1 mm and 2 mm in diameter.

As can be seen in the figure, a smaller background is measured for fiber diameters of 1 mm, which reduces the low detection level LDL of the detector. There are other reasons that favor the use of 2 mm fibers, such as their greater resistance and an improvement to the passage of water through them, so an experimental test is needed to choose the best design.

### 8.1.5 Effect of the PMMA windows

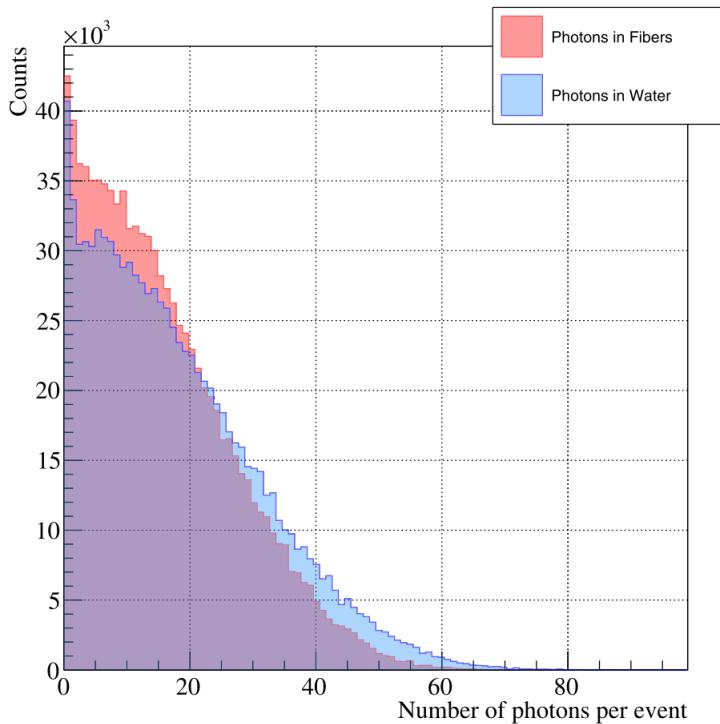
In the first prototypes, TRITIUM-IFIC 0 and TRITIUM-IFIC-1, the fibers were directly coupled to the photosensor, so the detected photons were only those guided by fibers. However, in the last prototypes, TRITIUM-Aveiro 0 and TRITIUM-IFIC 2, two PMMA windows are used, which allows the detection of photons guided by fiber and photons that come from the water medium.

To quantify the importance of this effect, the TRITIUM-Aveiro 0 prototype was simulated and the distribution of the number of photons that reach the PMMA per tritium event are shown in Figure 8.9. Fiber-guided photons are shown in a red distribution, while those traveling in the water medium are represented in the blue histogram.

It can be seen that the tritium signal obtained from the water is as important as that obtained from the fibers, contributing half of a signal. Therefore, an improvement in tritium detection efficiency is achieved using PMMA windows.

## 8.2 Simulation Results of TRITIUM Monitor

This section shows the results obtained in the simulation of the TRITIUM monitor, consisting of a study of the tritium detection with various TRITIUM-IFIC 2 prototypes in parallel, section 8.2.1, and a study of the effect of the background rejection system, section 8.2.2.

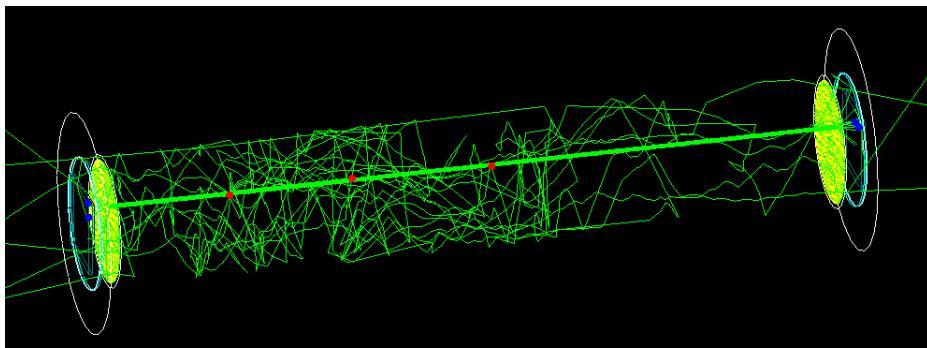


**Figura 8.9** – Distribution of photons reaching PMMA windows. The red histogram includes those guided by fibers and the blue histogram includes those traveling in the water medium [Aze20].

### 8.2.1 Simulation Results of TRITIUM-IFIC 2

This section shows the results obtained with the simulation of the TRITIUM-IFIC 2 prototype, which was used for two different objectives. On the one hand, these simulations were carried out to find the Low Detection Limit, LDL, of this prototype for tritiated water activity, which is an important parameter to know the limitation of the prototype. On the other hand, these simulations serve to study the activity resolution of the prototype and how it can be improved through various parameters such as the increasement of the integration counting count time of the measurement or the number of prototypes read in parallel.

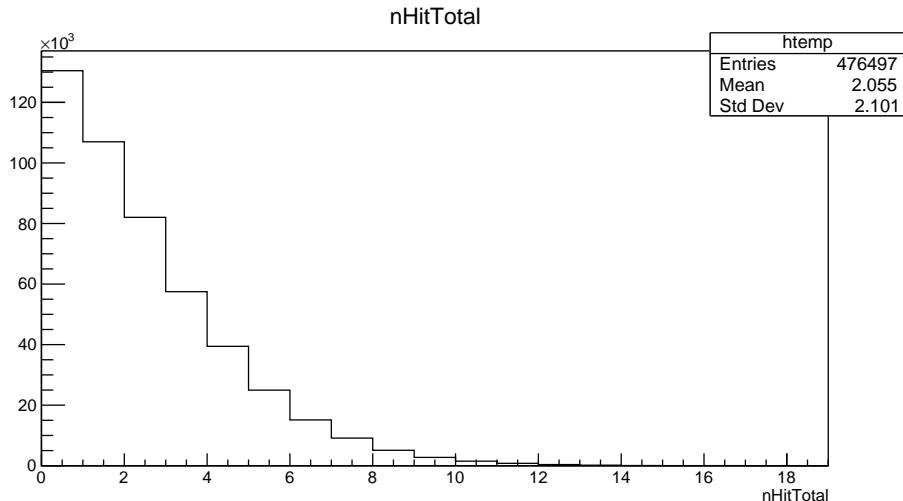
The detection of a tritium event by the TRITIUM-IFIC 2 prototype is shown in Figure 8.10, in which, the path followed by the photons created in scintillating fibers are represented by green lines which end in red dots when it is absorbed in the fiber or the water and blue dots when it is absorbed in the PMTs (detected). The fiber that has detected the tritium electron is clearly identified and the photons out of this are those that has not been collected due to the critical angle. Blue dots are obtained in both PMTs for this event, indicating that this is detected on time coincidence.



**Figura 8.10** – Tritium electron detected in the simulated TRITIUM-IFIC 2 prototype. The path of the optical photons is represented by green lines and the position in which it is absorbed is represented by red and blue dots (absorbed in water or PMT, respectively).

Several variables were used as tests in each simulation to verify the different steps of the simulation such as the production of tritium events, the energy deposition in scintillating fibers and their subsequent photon emission, spatial distribution of generated events, detected events, etc.

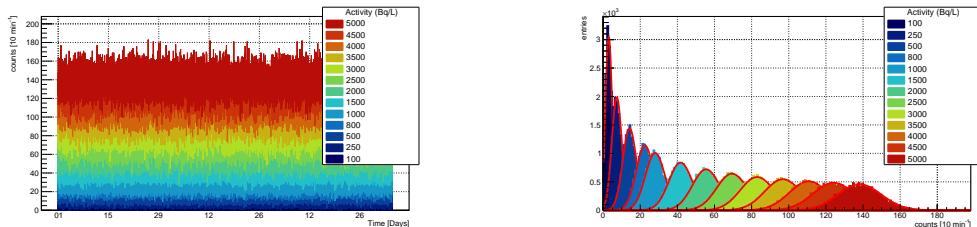
The distribution of the number of photons detected by both PMTs per tritium event was obtained for the simulated TRITIUM-IFIC 2 prototype, shown in Figure 8.11.



**Figura 8.11** – Photons detected by both PMTs per tritium event in the simulated TRITIUM-IFIC 2 prototype.

A maximum of 17 photons is obtained for the TRITIUM-IFIC 2 prototype simulations, which are in agreement with the maximum of 15 photons experimentaly measured by the experimental experience, Figure 7.9b. This confirms that the value used in the simulations for the Birks coefficient,  $k_B = 0.136 \text{ mm/MeV}$ , is adequate. The experimentally obtained distribution are a bit small between 3 and 8 photons, probably due to imperfections of the prototype which are impossible to simulate.

Various activities were simulated from 100 Bq/L to 50 kBq/L for three months of simulated data taking and an integration counting time of



(a) Counts as a function of the time.

(b) Distribution.

**Figura 8.12** – Tritium counts detected with a simulated TRITIUM-IFIC 2 prototype using a integration counting time of 10 min a) as a function of the time b) distribution of them.

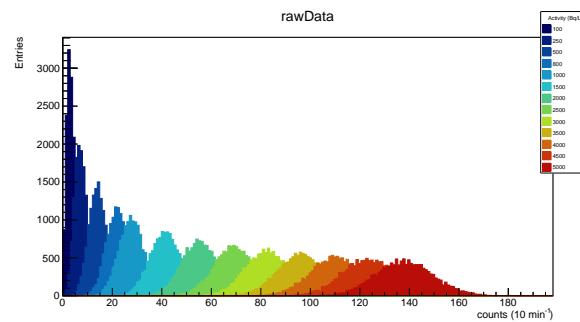
10 min was used.

The measurements obtained are presented in Figure 8.12a as a function of time, which are histogrammed in Figure 8.12b. In this figure a zoom is applied from 100 Bq/L to 5 kBq/L (interesting range) for better visualization.

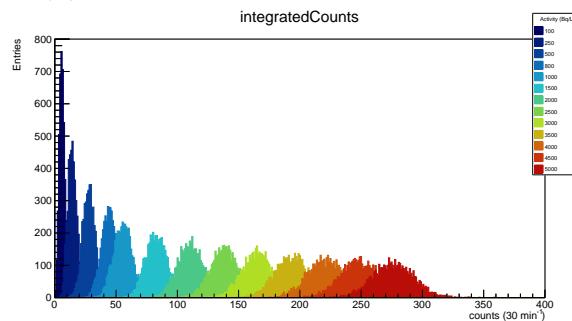
Difference of 250 Bq/L is not distinguished due to the overlapping of several distributions. To reduce the width of the distribution obtained for each activity, the statistics must be increased, which can be done in two different ways, increasing the integration counting time or increasing the number of prototypes read in parallel.

To check the effect due to an increase of the integration counting time similar distributions are obtained for three increasing integration counting times (10 min, 30 min and 60 min), which are shown in Figure 8.13.

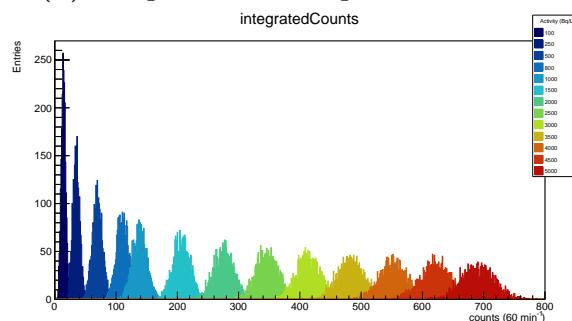
The effect of increasing the integration counting time is clearly visible in this figure, reducing the relative distribution width and improving the activity resolution of the TRITIUM monitor. Differences as low as 250 Bq/L are clearly distinguished using only one detector and an integration



(a) Integration counting time of 10 min.



(b) Integration counting time of 30 min.



(c) Integration counting time of 60 min.

**Figura 8.13** – The distribution of the tritium counts detected with a simulated TRITIUM-IFIC 2 prototype for three different integration counting time, a) 10 min b) 30 min and c) 60 min.

counting time of 60 min, which we can still consider a quasi-real time measurement. Similarly, this distributions are shown in Figure 8.14 for 10 min of integration counting time, in which three increasing number of prototypes were read in parallel (1, 5 and 10).

Again the reduction of the distribution width is clearly visible in these figures, improving the activity resolution of the detector. In this case, differences of 250 Bq/L are clearly distinguished using a integration counting time of 10 min and measuring with 5 TRITIUM-IFIC 2 prototypes.

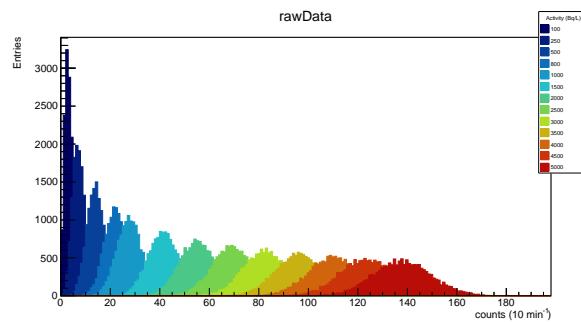
The effect on the resolution, defined as the equation 8.1, is also studied as a function of both TRITIUM monitor characteristics, integration counting time and number of used prototypes, and shown in Figure 8.15.

$$\text{Resolution}(\%) = \frac{\text{FWHM}}{\text{centroid}} \cdot 100 \quad (8.1)$$

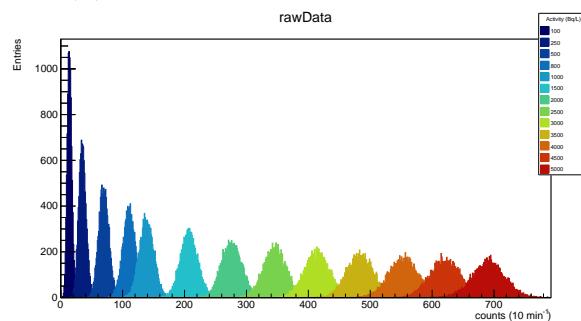
A growing improvement can be observed in both cases and, as can be verified with the lines in the figure, the resolution of both cases fit to the expected behavior of the inverse of the root of accounts.

Therefore, both characteristics must be balanced based on the requirements and financial budget of the experiment. The activity difference, the distribution peaks of which are clearly distinguished for each different case of integration counting time and number of detectors used, is summarized in Table 8.1.

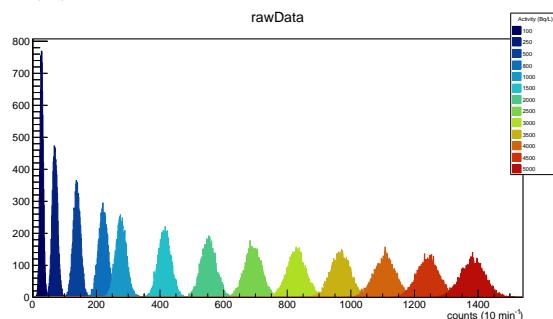
The decision made in the TRITIUM experiment is to install 3 different TRITIUM-IFIC 2 prototypes, with which differences of 250 Bq/L are expected to be distinguished using an integration counting time of 30 min. These prototypes are expected to be installed in Arrocampo dam as soon as possible. Two other TRITIUM-Aveiro 0 prototypes are being built and will be installed soon, along the one currently installed.



(a) One TRITIUM-IFIC 2 prototype.

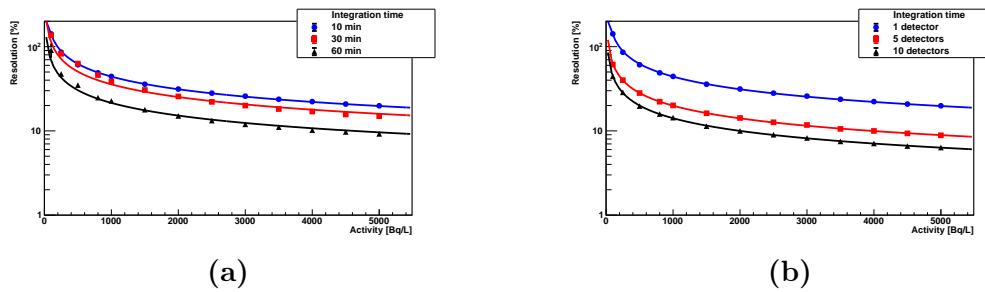


(b) Five TRITIUM-IFIC 2 prototypes.



(c) Ten TRITIUM-IFIC 2 prototypes.

**Figura 8.14** – The distribution of the tritium counts detected with several simulated TRITIUM-IFIC 2 prototypes a) 1, b) 5 and c) 10 for an integration counting time of 10 min.



**Figura 8.15** – Resolution of the TRITIUM-IFIC 2 prototype as a function of the a) integration counting time b) number of prototypes.

| # of Detectors | 10 min      | 30 min   | 60 min            |
|----------------|-------------|----------|-------------------|
| 1              | < 1000 Bq/L | 500 Bq/L | 200 Bq/L          |
| 5              | 200 Bq/L    | 150 Bq/L | 100 Bq/L          |
| 10             | 150 Bq/L    | 100 Bq/L | $\approx 50$ Bq/L |

Table 8.1: Difference in activity that can be clearly distinguished for various cases of the TRITIUM-IFIC 2 prototype based on different integration counting times and different number of prototypes.

### 8.2.2 Simulation Results of Background Rejection System

Finally, this section shows the results obtained with the simulation of a TRITIUM-IFIC 2 prototype when the background rejection system, detailed in section 6.2.6, is included. These simulations quantify the effect on the tritium measurement of both parts of the background rejection system, lead shield and cosmic veto.

Similar to the simulations used for the study of the TRITIUM-IFIC 2 prototype, analogous variables were used as tests, which were systematically verified, to ensure that all the steps of the simulations were carried out correctly.

Three different simulations were carried out to independently quantify how the tritium detection is affected due to both, the lead shield and the cosmic veto. The first simulation consists of a TRITIUM-IFIC 2 prototype and the cosmic ray source, in the second simulation a lead shield was added and for the third simulation, the cosmic veto was also included.

The cosmic events detected by the TRITIUM-IFIC 2 prototype are reduced around 5.5 times when a lead shielding with walls of 5 cm is included. This reduction is mainly caused due to the stop of the weak cosmic radiation (energy lower than 200 MeV). It has to be taken into account that the natural backgrounds of the place are not included in this simulation. This radioactive background will be also stopped by the lead shielding, so the expected reduction of the radioactive background due to the passive veto is even better.

Regarding to the cosmic events that pass through the lead shield and reach the TRITIUM IFIC 2 prototype, which are the hard cosmic radiation, a percentage of around 10% is detected with the cosmic veto and, therefore, removed to the tritium measurement.

# Chapter 9

## Conclusions and Prospects

This chapter contains a brief summary of the most important achievements reached in this work and highlights the main conclusions obtained.

The design of a tritium detector capable of measuring low tritium activities in quasi-real time is mandatory since this is one of the first sign of a malfunctioning of a nuclear facility, such us nuclear power plants, future nuclear fusion reactors and laboratories of high energy physics.

The goal of the TRITIUM project is to design, build, install and commission a tritium monitor that will measure tritium activities as low as 100 Bq/L (legal limit imposed for European Council Directive 2013/51/EU-RATOM for drinking water consumption) in quasi-real time (1 hour or less).

The TRITIUM monitor developed in the TRITIUM project has different parts, which are detailed in this thesis. The different parts are an ultrapure water system, which prepares the water sample before the measurement, a tritium detector, consisting of scintillating fibers readout by photosensors (PMTs or SiPM arrays), and a passive and active shielding, which is used to reduce de radioactive background measured by the tritium

detector.

Several studies were carried out to characterize the different parts of the TRITIUM monitor independently:

First, the components of the tritium monitor, which are scintillating fibers and SiPM, were studied.

- On the one hand, a characterization of the photon collection efficiency of the BCF-12 no clad scintillating fibers was performed, which was compared with single clad and multiclads fibers to check the importance of the clad.

In addition, a conditioning process of scintillating fiber was developed, tested and implemented, consisting of cutting, polishing and cleaning them, the objective of which is to increase the tritium detection efficiency. Due to the large number of scintillating fibers used, it was necessary to develop an automatic polishing machine, based on arduino technology, which is capable of polishing up to one hundred scintillating fibers at the same time. The improvement in photon collection efficiency due to the polishing and cleaning process was quantified in more than 40% and 21%, respectively.

- On the other hand, a characterization of the SiPM used (Hamamatsu model S13360-6075) was carried out at the level of a single SiPM. In this characterization several interesting parameters such as quenching resistance, terminal capacitance, internal gain of SiPM, breakdown voltage, temperature coefficient, etc. were experimentally measured. An additional calibration was carried out at the level of a SiPM matrix in which the probability of crosstalk between different SiPMs was measured, obtaining an insignificant probability of happening. The linearity of the SiPM signal as a function of the the number of scintillating fibers was also verified.

Due to the strong dependence of the SiPM internal gain on temperature, a temperature compensation mechanism was developed and experimentally tested. The objective of this mechanism is to compensate for temperature variations with variations in the operating voltage of the SiPM, maintaining the internal gain of the SiPM during its operation.

Second, a characterization of the ultrapure water system was carried out, in which it was checked that the imposed requirements were fullfit. The requirements are to prepare water samples with very low conductivity (of the order of about  $10 \mu\text{Sv}/\text{cm}^2$ ) in which the organic matter and all particles up to 1 mm diameters is removed without affecting the tritium levels.

Third, a characterization of the active veto was carried out in which several interesting parameters were experimentally measured.

- First, the quality of the coverage of the plastic scintillator, consisting of a layer of teflon, aluminium and black tape, was tested and quantified, obtaining an improvement in the uniformity and quality of the photon collection efficiency.
- Second, the high voltage and threshold that optimize the detection of hard cosmic rays ( $> 200 \text{ MeV}$ ) was experimentally found.
- Third, a hard cosmic events was experimentaly measured with the active veto developed, obtaining an energy spectrum with a shape similar to a Landau functions, as it is expected, and an efficiency of hard cosmic rays detection of 85%. In addition, the relationship between the number of the hard cosmic rays measured and the distance between both plastic scintillators of the active veto was obtained, allowing this distance to be changed without the need to perform a new calibration of the active veto.

This background rejection system, consisting of a lead shielding and an active veto, is essential to achieve the activity goal of 100 Bq/L.

Fourth, four different prototypes of the tritium detector have been developed. The first two prototypes, TRITIUM-IFIC 0 and TRITIUM-IFIC 1, was used to detect potential problems that affect to the tritium measurement as well as to test several improvement in the detector design. The last two prototypes, TRITIUM-Aveiro and TRITIUM-IFIC 2, are two different designs to be used in the final tritium cell of the TRITIUM monitor. Each design has its own advantages and disadvantages and the one with the best results will be used as a final design of the TRITIUM cell.

With the different prototypes, an increasing sensitivity has been achieved, showing the effect of the applied improvements. The best tritium detection efficiency was obtained with the latest prototype developed, TRITIUM-IFIC 2, with which the State-Of-The-Art of tritium detection has been overcomed. The most important results of each prototype developed in TRITIUM project are presented in Table 9.1, in which the results obtained with other experiments are also included.

As can be seen in the table, the specific efficiency of the latest prototype, TRITIUM-IFIC 2, is almost an order of magnitude better than the best result obtained in other experiments (Hofstetter). Special attention need to be paid for the specific efficiency obtained for the TRITIUM-Aveiro prototype, which is smaller than the expected. A possible reason is because the used fibers was not polished nor cleaned, reducing the tritium events detected. It could be interesting to develop a new TRITIUM-Aveiro prototype, the fibers of which are prepared with the conditioning process detailed in sections 4.1.1 and 4.1.4 to decide which tritium cell design optimizes the tritium detection.

A low detection level, LDL, of 29.8 kBq/L has been measured for the TRITIUM-Aveiro prototype using 1 minutes of integration time,

| Study      | $\varepsilon_{det}(\frac{cps \cdot 10^{-3}}{kBq/L})$ | $F_{sci}$ (cm <sup>2</sup> ) | $\eta_{det}(\frac{cps \cdot 10^{-6}}{kBq/L \cdot cm^2})$ | LDL (kBq/L) |
|------------|--|------------------------------|--|-------------|
| Muramatsu  | 0.39   | 123                          | 3.13   | 370         |
| Moghissi   | 4.50   | > 424.1                      | < 10.6   | 37          |
| Osborne    | 12   | 3000                         | 4  | 37          |
| Singh      | 41   | 3000                         | 13.7   | < 37        |
| Hofstetter | 2.22   | ~ 100                        | < 22.2   | 25          |
| T-IFIC 0   | $2.11 \pm 0.85$                                      | 219.91                       | $9.59 \pm 3.87$  | 100*        |
| T-IFIC 1   | $38.42 \pm 1.61$                                     | 402.12                       | $95.55 \pm 4.01$   | 100*        |
| T-Aveiro 0 | $64.87 \pm 19.41$                                    | 4071.50                      | $15.93 \pm 4.77$   | 29.8        |
| T-IFIC 2   | $711.03 \pm 27.77$                                   | 5026.55                      | $141.45 \pm 5.52$  | 10*         |

Table 9.1: Results of scintillator detector developed for several experiments (including the TRITIUM project) for tritiated water detection. This table shows the efficiency of the detector ( $\varepsilon_{det}$ ), its active surface ( $F_{sci}$ ), its specific efficiency ( $\eta_{det} = \varepsilon_{det}/F_{sci}$ ), defined as its efficiency normalized to its active surface, and its low detection-level (LDL) for each study listed above. The "\*" symbol indicates that this is the specified activity that the detector can distinguish from the background, but it is not its LDL.

slightly improving the State-Of-The-Art. It is expected to be improved up to 5 kBq/L by increasing the integration time up to 1 hour, which is still considered quasi-real time.

A better result was obtained for the TRITIUM-IFIC 2 prototype, being able to clearly measure an activity of 10 kBq/L, improving the best results obtained in other experiments. However this is not the LDL of the prototype. To measure this, it is necessary to take many more measurements, similar to how it was done for the TRITIUM-Aveiro prototype.

Nevertheless the low detection level achieved with this prototypes is further from being the goal of the TRITIUM project, 100 Bq/L. This is not a problem since the TRITIUM monitor will consists of several TRITIUM cells readout in parallel, becoming the TRITIUM detector in a scalable detector. It means that more tritium cells can be used, readed in parallel, to improve the results obtained. The activity goal is expected to be achieved using three different cells of TRITIUM-IFIC 2 read in anti-coincidence with an active veto.

In summary, two different prototypes has been developed in the TRITIUM project with which it is possible to measure low activities of tritiated water in quasi-real time, improving the specific activity and the low detection level currently achieved with other experiments. In addition, the stability of the tritium detection efficiency of both prototypes has been verified during several months.

Currently, the lead shielding, the ultrapure water system and a TRITIUM-Aveiro prototype are installed in Arrocampo dam, near to Almaraz Nuclear Power Plant. Two additional TRITIUM-Aveiro prototypes and an active veto are planed to be installed as soon as possible. In addition, three prototypes of TRITIUM-IFIC 2 and an active veto are ready to be installed too, the instalation of which has been delayed due to the coronavirus pandemic.

Finally several Monte Carlo simulation has been developed using Geant4. These simulations was used for three different tasks:

- First, several simulations were carried out to study the different steps of the simulation, such us the energy deposition of tritium electrons on scintillating fibers (spectrum peaked of around 5 keV), the number of photons produced by the fibers (spectrum peaked of around 10 photons) or which of this are detected by the photosensors in time coincidence (spectrum peaked of around 25 photons). They were also used to quantify the importance of the reduction of the scintillating fiber signals because tritium electrons are not MIP particles (Birks effect).
- Second, these simulations were used to test different tritium detector designs, such as different fiber lengths or fiber diameters, and choosing the one with the best results, that is, the one that optimizes the tritium detection efficiency.
- Third, these simulations were used to verify the results obtained with the last two TRITIUM prototype, such us the spectrum of the number of photons obtained per tritium event. They were also used to find the sensibility of each different prototype and how the integration time and the number of cells used can improve to the tritium detection of the TRITIUM monitor.



# Appendices



## Appendix A

# Electronical Schemes of PCBs Used for SiPM Characterization

Y más cosas aún.

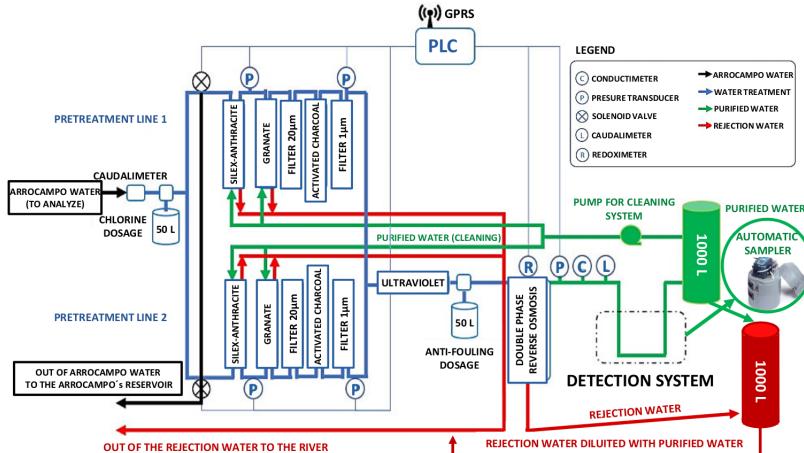


## Appendix B

# Ultrapure Water System

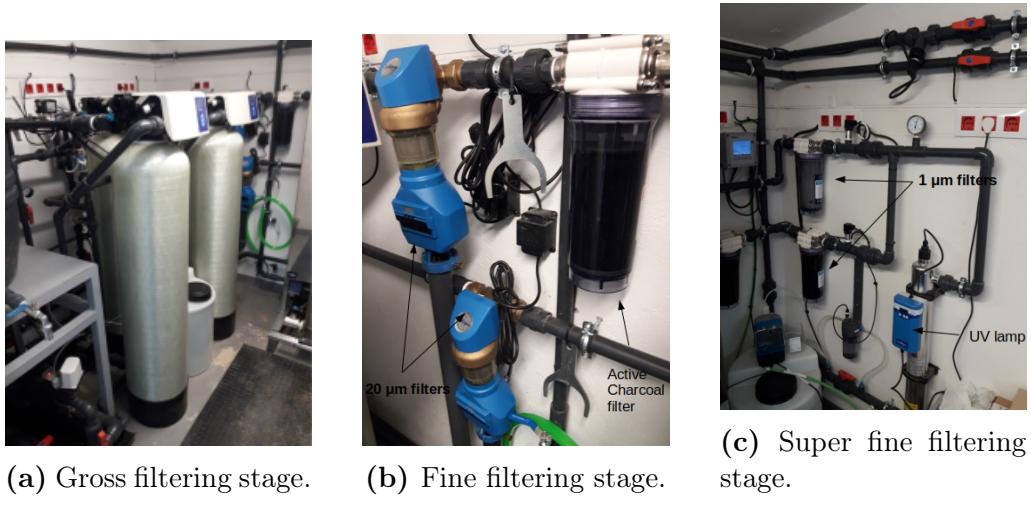
In this appendix I show several photos of the ultrapure water system in the same order that the water flows through them.

First of all, the complete scheme of the ultrapure water system is shown in Figure B.1:



**Figura B.1** – Scheme of the ultrapure water system.

Secondly, the Gross filtering stage, made up of Silex-Antracite and



**Figura B.2** – Different stages of filtration of the ultrapure water system.

Granate filters, is shown in Figure B.2a:

In third place, the fine filtering stage, consisting of  $20\text{ }\mu\text{m}$  filter and active carbon filter, is shown in Figure B.2b:

In fourth place, the superfine filtering, composed of the  $1\text{ }\mu\text{m}$  filter and the UV lamps, is shown in Figure B.2c

In fifth place, the double phase reverse osmosis is shown in Figure B.3a

In sixth place, the containers in which we store the ultrapure water and the reject water after treatment is shown in Figure B.3b.

In seventh place, the Siemens PLC, software used to control the ultrapure water system, is shown in Figure B.4.

Finally, the complete system of the ultrapure water system is shown in Figure B.5



(a) Doble phase reverse osmosis stage.



(b) Storage containers of reject and ultrapure water.

**Figura B.3** – Doble phase reverse osmosis stage and containers used to store the outlet water of the ultrapure water system.



(a)



(b)



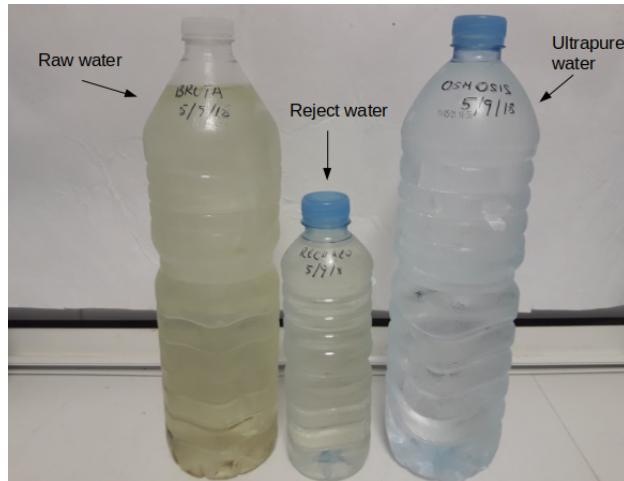
(c)

**Figura B.4** – Siemens PLC, software for remote control of ultrapure water system.



**Figura B.5** – General photo of the complete ultrapure water system.

Just as a curiosity, the three types of water (raw water, rejection water and ultrapure water) are shown in Figure B.6, where you can visually check the difference in the turbidity of each type of water.



**Figura B.6** – Raw water, reject water and ultrapure water obtained with this system.

# Appendix C

## Preparation of Liquid Radioactive Source of Tritium

To prepare this radioactive liquid source, 1.86 g (uncertainty of 0.05%) of tritium was purchased from the Germany company PTB<sup>1</sup>, which has a serial number of 2005 – 1442 and reference number of PTB-6.11 – 285/03.2017 [PTB]

The activity of this tritium source is 26,8 MBq/g (uncertainty of 2.24%), reference data of 1 of January of 2017, and it was dissolved in 500 mL (uncertainty of 0.05%) of ultrapure water, giving 500 ml of tritium water, to which we will call standard solution, with an activity of 100.096 kBq/g (uncertainty of 2.24%), that's, 99.696 kBq/L (uncertainty of 2.24%), which was measured with the TRI-CARB 2810 system, based on liquid scintillation readout by PMT.

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<sup>1</sup>Physikalisch-Technische Bundesanstalt, Braunschweig and Berlin, Germany

212APPENDIX C. PREPARATION OF LIQUID RADIOACTIVE SOURCE OF T

# Appendix D

## Electronic System of TRITIUM-Aveiro 0 prototype

The electronic system used in TRITIUM-Aveiro 0 prototype consists of several PCB and can be divided into two parts:

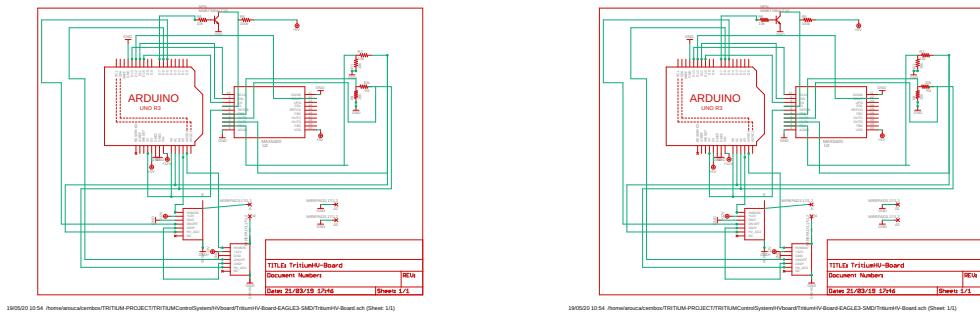
1. A PCB, whose electronic scheme is shown in Figure D.1, was designed to power the PMTs with a negative high voltage. It consists of several high voltage power supply, model C11152-01 from Hamamatsu company [K.K15], one for each PMT used, which is controled by a DAC<sup>1</sup>, model MAX5500 from Maxim Integrated company [Int09]. An Arduino Mega is used for the DAC communication and cross-checking the output values and it is connected to a Raspberry Pi to control the system.

A graphical interface, shown Figure D.1b, has been developed to manage the different options of this system in a comfortable way.

2. A electronical chain consisting of several PCBs was used to process

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<sup>1</sup>DAC, Digital-to-analog converter

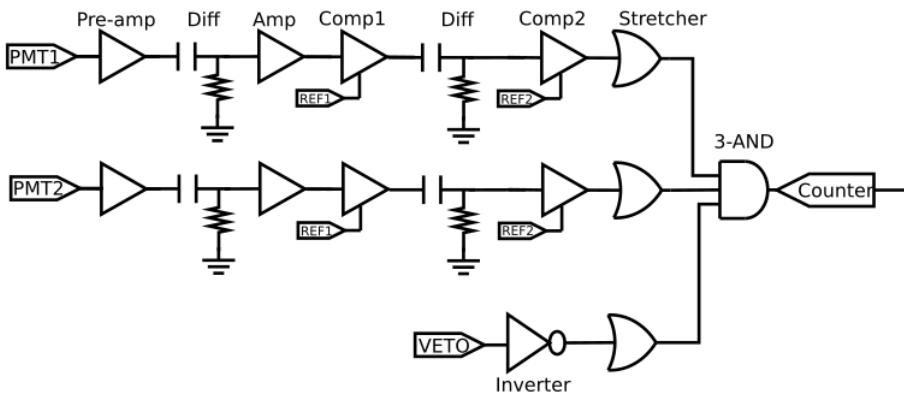


(a) Electronic scheme of the PCB.

(b) Graphical user interface.

**Figura D.1** – Electronic scheme of the PCB designed to power the PMTs of Aveiro prototype and the graphical user interface developed to control it.

and analyze the system signals, whose simplified electronic scheme is shown in Figure D.2.



**Figura D.2** – Simplified electronic scheme used to process and analyze the signal of TRITIUM-Aveiro 0 prototype.

It consists of three different lines, two of them are used for the PMT signals of the prototype and the remaining line is used for doing anti-coincidence with a active veto.

To test this electronic chain a plastic scintillation with dimensions of  $10 \cdot 10 \cdot 1 \text{ cm}^3$  was used to simulate a veto signal but four different

veto are being developed, each one is based on a rectangular plastic scintillations of Saint-Gobain company [SG], whose dimensiones are  $50\text{ cm} \cdot 30 \cdot 2\text{ cm}^3$  with a PMT coupled, model R2154-02 2" from Hamamatsu company [K.K10]. The output signal of these PMTs will be input in a OR stage, whose response will be introduced in the veto line shown previously in Figure D.2. As a result, each plastic scintillator will be read in anticoincidence with tritium-Aveiro prototype.

Both lines, used to process and analyze the PMT signals of the prototype, are equal and they are used to operate in time coincidence. First, each PMT signal is introduced in a preamplifier model CR111 from CREMAT Inc. company [Incl], which is used to shape and preamplify the signal. To reduce electronic noise and signal loss, both preamplifiers are connected as close as possible to the PMTs and they are located inside of aluminum boxes which act like a Faraday cage.

Each preamplifier is followed by a differentiation stage, which is used to reduce the time width of the signal, and amplification stages, used to amplify the signal. The amplification used is the model OPA656 from Texas Instruments [Ins15].

Then, a fast comparator, model LT111 from Linear Technology company [Tec], is used to set a threshold which will be used to remove the PMT signals whose amplitude are below this value (dark counts of the PMT). A MAX5500 DAC is used to configure the thresholds.

The time width of the preamplifier output signal is too large,  $200\text{ }\mu\text{s}$ , so which too many false coincidence will be registered. To solve this problem a second differentiation stage is included and a second comparator are added to produce a 5V square signal again.

Finally a tunable pulse stretcher based on an OR gate, model SN74AHC1 from Texas Instruments company [Ins17], is used to set the time width of each signal at 100 ns, with which the time coincidence windows of our adquisition system is 200 ns, narrow enough to have a negligible false coincidence rate.

In the remaining line, used for the veto signal, an inverter is used in the first stage. With it, the signal will always be in the high level, 5 V, except when a cosmic particle is detected, in which case the signal will be in the low level, 0 V. Then, another stretcher is used to create a signal with the same time width than the others, 100 ns.

Lastly, these three signals are introduced into a 3-input AND gate, model SN74LVC1G11 from Texas Instruments company [Ins16], to perform a logic level comparison. With this last stage we achieve a temporal coincidence of both PMT signals of the prototype and anti-coincidence of them with the veto signal. The output signal of this last stage is simply connected to a pulse counter.

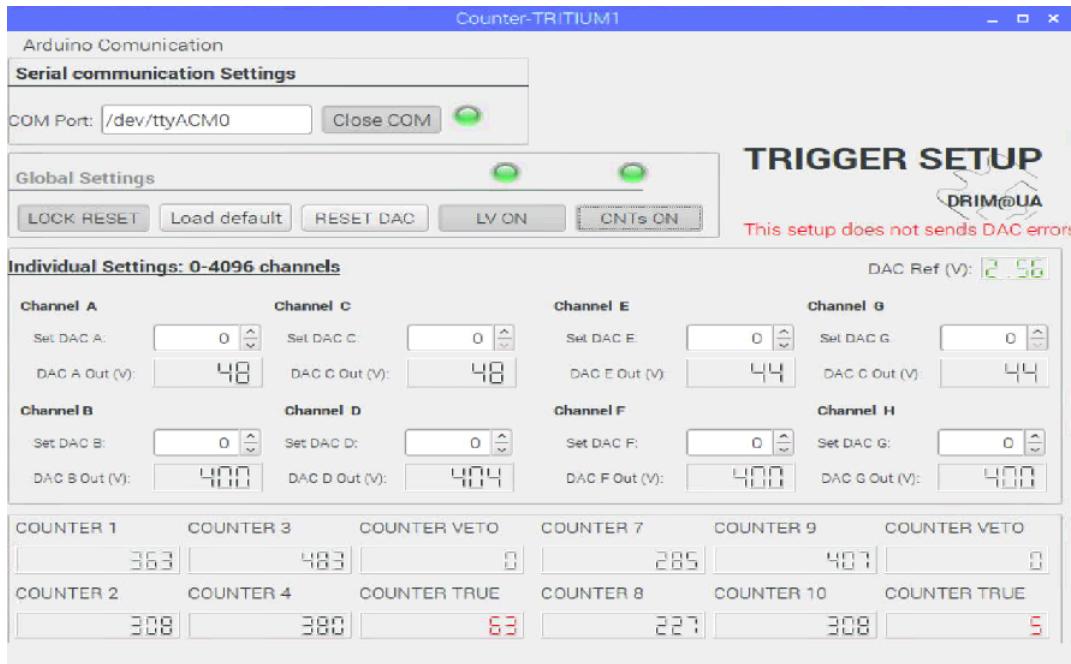
A GPIO pins of a Raspberry Pi is used to communication with the system, control it and configure the different threshold levels and a graphical user interface, whose appearance is shown in Figure D.3, has been developed to manage in a comfortable way the counter system.

In addition to count, which is the option normally used in our detector, this electronic system include a voltage follower circuit connected to the preamplifier output signal which can be used to obtain a energy spectrum of each PMT of the prototype.

It is important to note that, although this system has a graphical user interface that allows comfortable control of the system, the usual way in which it is controlled is remotely through the computer terminal.

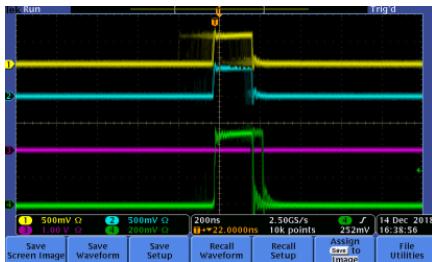
In Figure D.4 two screenshots are shown to demostrate two different situations of this system. There, we have four different signals. The yellow and cyan signal are input signals of the AND-Gate, which come from the PMT signals of the prototype. The pink signal is the third remaining input signal of the AND-Gate, which come from the PMT signal of the veto. The last signal, green, is the output signal of the AND-Gate.

As can be seen, in Figure D.4a both PMTs of the prototype have detect a time coincident event, which has not been detected for the

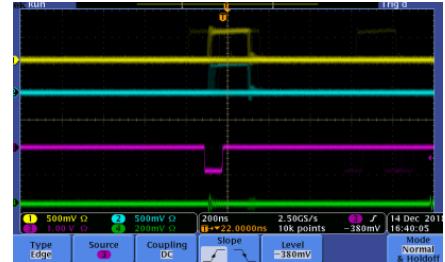


**Figura D.3** – Graphical user interface used to manage the counter system.

veto, so this event is counted. In Figure D.4b, a time coincidence event has been observed in the three PMTs, which means that it is a cosmic event, so this event is not counted.



(a) Event accepted by the electronic system.



(b) Event rejected by the electronic system.

**Figura D.4** – Two different situations of the electronic chain response. A.- Event accepted since veto has not detected it. B.- Event rejected since veto has detected it

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