

Facultad de Física  
Departamento de Física Atómica, Molecular y Nuclear



VNIVERSITAT  
DE VALÈNCIA

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

Marcos Martínez Roig

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Under the supervision of:  
José Díaz Medina  
Nadia Yahlali Haddou



*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 by nuclear facilities and, specifically, by nuclear power plants. Large amounts of tritium are normally produced in the water of their cooling system, which are finally emitted to the environment. Due to the fact that large releases 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 for drinking water in Europe to 100 Bq/L, or the U. S. Environmental Protection Agency, in United States, that limits tritium in drinking water to 20 nCi/L.

Due to the low energy of electrons emitted in the tritium decay, very sensitive detectors are needed for measuring them like LSC. The issue with LSC is that it is an off-line method and the measurement process can take 2 days or more, a time too long to detect a problem in the NPP.

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

The results of the TRITIUM project are presented in this thesis. In the framework of this project a quasi-real time monitor for low tritium

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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 passive shielding for reducing the natural radioactive background 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 that exceeds the legal limits established in Europe. The final idea will be to include this monitor in the early alarm system of Extremadura consisting of several detectors the objective of which is to control the impact of Nuclear Power Plants to the environment.

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

# Nomenclature and Acronyms

Acronyms:

<i>ALARA</i>	— As Low As Reasonably Achievable
<i>APD</i>	— Avalanche Photodiode
<i>BIXS</i>	— Beta Induced X-ray Spectrometry
<i>BWR</i>	— Boiled Water Reactor
<i>CCD</i>	— Charge-Coupled Device
<i>CDF</i>	— Dose Conversion Factor
<i>CE</i>	— Collection Efficiency
<i>CL</i>	— Collection Efficiency
<i>CNRS</i>	— Le Centre National de la Recherche Scientifique, France
<i>CSN</i>	— Nuclear Safety Council
<i>C<sub>t</sub></i>	— Terminal Capacitance of the SiPM
<i>DAQ</i>	— Data Acquisition System
<i>DRIM</i>	— Detecção da Radiação e Laboratório Imagem Médica laboratoire (Laboratory for Radiation Detection and Medical Imaging)
<i>EEC</i>	— European Economic Community
<i>EPA</i>	— Environmental Protection Agency
<i>EU</i>	— European Union
<i>EURATOM</i>	— European Atomic Energy Community
<i>FF</i>	— Fill Factor of a SiPM
<i>G – APD</i>	— Geiger Avalanche Photodiode

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$GCR$	— Gas-Cooled Reactor
$GL$	— Guideline Level
$G_{PMT}$	— Gain of the PMT
$G_{SiPM}$	— Gain of the SiPM
$HPGe$	— High Purity Germanium Detector
$HV$	— High Voltage
$HWR$	— Heavy Water Reactor
$IAEA$	— International Atomic Energy Agency
$IC$	— Ionization Chamber
$ICRP$	— International Commission on Radiological Protection
$ICRU$	— International Commission of Radioactivity Units and Measurements
$I_{DC}$	— Intensity of the PMT due to the dark current
$I_{PMT}$	— Intensity of the PMT when photons are detected
$ISR$	— International Society of Radiology
$LARUEX$	— Laboratorio de Radiactividad Ambiental of the University of Extremadura (Environmental Radioactivity Laboratory of the University of Extremadura)
$LDL$	— Lower Detection Limit
$LED$	— Light-Emitting diode
$LSC$	— Liquid Scintillation Counting
$LWR$	— Liquid Water Reactor
$MAPD$	— Micro-Pixel Avalanche Photodiode
$MDA$	— Minimum Detectable Activity
$MPPC$	— Multi-Pixel Photon Counter
$MRS - ADP$	— Metal-Resistor-Semiconductor Avalanche Photodiode
$NA$	— Numerical Apertures
$NPP$	— Nuclear Power Plants
$P_{av}$	— Avalanche probability in a SiPM
$PCB$	— Printed Circuit Board
$PDE$	— Photodetection Efficiency of the SiPM
$PHWR$	— Pressurized Heavy Water Reactor

<i>PMMA</i>	— Polymethyl Methacrylates
<i>PMT</i>	— PhotoMultiplier Tube
<i>POF</i>	— Region of interest
<i>PVC</i>	— Polyvinylchloride
<i>PWR</i>	— Pressurized Water Reactor
<i>q</i>	— Annual Volume of drinking water consumed
<i>QE</i>	— Quantum Efficiency
<i>quasi-real</i>	— Less than 10 minuts
<i>RDL</i>	— Reference Dose Level
<i>REA</i>	— Red de Estaciones Automáticas
<i>REM</i>	— Red de Estaciones de Muestreo
<i>ROI</i>	— Region of interest
<i>R<sub>q</sub></i>	— Quenching resistance of the SiPM
<i>S</i>	— Energy loss by the particle per unit of path length
<i>SDD</i>	— Silicon Drift Detector
<i>SiPM</i>	— Silicon PhotoMultiplier
<i>SSPM</i>	— Solid State PhotoMultiplier
<i>STP</i>	— Standard Temperature and Pressure conditions
<i>UDL</i>	— Upper Detection Limit
<i>UN</i>	— United Nations
<i>UNSCEAR</i>	— United Nations Scientific Committee on the Effects of Atomic Radiation
<i>U.S.</i>	— United States
<i>USA</i>	— United States of America
<i>U.S.DOE</i>	— United States Department of Energy
<i>U.S.EIA</i>	— United States Energy Information Administration
<i>U.S.EPA</i>	— United States Environmental Protection Agency
<i>V<sub>BD</sub></i>	— Breakdown Voltage of the SiPM
<i>V<sub>bias</sub></i>	— Supply Voltage of the SiPM
<i>V<sub>O</sub></i>	— Potential difference between the n and p layers of the SiPM
<i>V<sub>OV</sub></i>	— Over voltage of the SiPM

$WHO$	— World Health Organization
$\Delta TV_{op}$	— Temperature Coefficient (mV/ $^{\circ}C$ )
$\delta$	— Multiplication factor of a PMT dynode

## Atomic and nuclear symbols

$A_m$	— Activity Measured
$^2_1D$	— Deuterium (Non-Radiative Hydrogen Isotope)
$D_2O$	— Heavy Water
$E_b$	— Binding energy of the electron in a specific material
$E_e$	— Energy of the electron
$E_{\gamma} = h\nu$	— Energy of the photon
$F_{sci}$	— Active surface of the Plastic Scintillator
$mip$	— Minimum Ionizing Particle
$m_0$	— Rest mass of a electron
$NaI(Tl)$	— Thallium doped Sodium Iodide
$OBT$	— Organic tritium molecule
$q_e$	— Electron Charge
$Q_{\beta}$	— Energy released in a radioactive decay
$S$	— Specific Energy Lost
$S_{ij}$	— Single states of energy levels of electrons in scintillator
$T_{1/2}$	— Half-life Time of a Radioactive Element
$T_{ij}$	— Triple states of energy levels of electrons in scintillator
$\varepsilon_{det}$	— Specific Detector Efficiency
$\eta_{det}$	— Intrinsic Detector Efficiency
$\lambda$	— Wavelength
$\lambda_p$	— Maximum wavelength of the associated spectrum
$\sigma$	— Cross Section of a radioactive process
$\sigma^{rel}$	— Relative uncertainty
$\sigma_{si}$	— Sistematically Component of the Uncertainty

- $\sigma_{st}$  Stadistical Component of the Uncertainty
- $\sigma_T$  Total Uncertainty of the Measurement

Units:

- kcps Kilo Counts per Second
- mrem Millirem
- *STP* Standard Temperature ( $0^{\circ}\text{C} = 273\text{K}$ ) and Pressure (1 atm)
- Sv/cm Sievert per Centimeter



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# Chapter 1

## Introduction

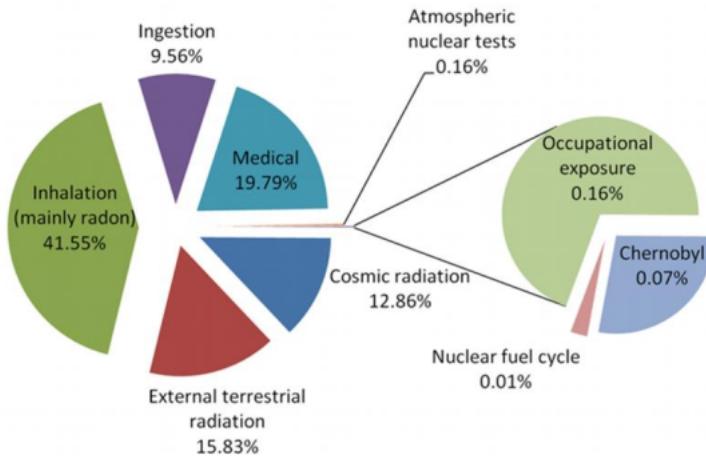
### 1.1 Tritium and Nuclear Energy

Radioactivity is the process in which an unstable atomic nucleus loses energy through the emission of particles such as photons, electrons, etc. This process has been present in the Universe since its inception as it was an important process of the Big Bang<sup>1</sup>. This was also present during the formation of the earth which explains why the different layers that make up the earth contain radioactive elements.

Humanity has always been exposed to radioactivity, whether present in the Earth's crust or in extraterrestrial sources (external natural irradiation). The human being himself is radioactive as radioactive elements are contained in the human body such as  $^{3}\text{H}$ ,  $^{14}\text{C}$  or  $^{40}\text{K}$ , introduced into the body through food or water ingestion or air inhalation (internal natural irradiation). The annual average radioactive dose received by the world 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.



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

As it can be seen in Figure 1.1, most of the radioactive dose received by the population is due to both internal and external natural radioactivity, the effective dose<sup>2</sup> of which is estimated to be 2.42 mSv/yr as shown in Table 1.1.

Since the discovery of radioactivity by Heri Becquerel in 1896, lots

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<sup>2</sup>The effective dose is the radioactive dose absorbed by the population, taking into account the different radiosensitivity of 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 radioactivity [UNS21, CSN21a].

of nuclear-based technology has been developed and applied to several fields such as energy production, research, medicine, industry, etc. Due to this nuclear technological development, various anthropogenic radioactive sources have appeared in society, resulting in a greater amount of radioactive elements released to the environment. It can be noticed in Figure 1.1 that the most important part of the dose received by the population from artificial sources comes from medical practices. The growing knowledge and development of measurement techniques of radioactivity, enable better assessment and characterization of the harmful effects of radioactivity in living organisms. 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 are kept below a safe limit. To accomplish this task, several organizations were created to propose recommendations in radiological protection to the different state organisms and governments at the international level:

1. 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. 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 included in the legislation of most countries.
3. The United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR [UNS21], was created in 1955, with the goal of estimating and reporting the levels and effects of ionizing radiation on the population and the environment. These estimates are taken into account by governments worldwide to establish their safety standards.

4. The International Atomic Energy Agency, IAEA [IAE21], was created in 1957 to promote the peaceful use of nuclear energy and to avoid its use for military purpose such as nuclear weapons. Although IAEA is an independent agency, it must to periodically report to the United Nations, (UN) [UN21].
5. At the level of the European Union (EU), the European Atomic Energy Community (EURATOM) was created in 1957, which is an international organization created through and ruled 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 funding of nuclear energy.
6. In Spain, the Nuclear Safety Council (CSN) was created in 1980 [CSN21a]. The CSN is the only authority in Spain on nuclear safety and radiation protection and its objective is to protect employees, the general population and the environment from the harmful effects of ionising radiation from anthropogenic origins. For this task, the CSN ensure that nuclear and radioactive facilities are operated safely and establish the preventive and corrective measures to apply in all radiological emergencies. The CSN has created various networks consisting of several detectors of radioactivity that are in charge of controlling the levels of radioactivity in the environment and assessing the impact of radioactivity facilities. Two of the most important networks are the network of automatic stations (REA, "Red de Estaciones Automáticas") and the network of sampling stations (REM, "Red de Estaciones de Monitoreo"):
  - (a) The network of automatic stations, REA [CSN21b], shown in Figure 1.2a, consists of several gamma detectors<sup>3</sup> distributed in Spain that measure the radioactive dose in real time. The REA

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

is employed for real-time detection of radiological issues, which enable taking prompt safety measures.

- (b) The network of sampling stations, REM [CSN21c], shown in Figure 1.2b, consists of several strategic points in Spain where samples are taken and transported to a laboratory to be measured. About twenty Spanish laboratories integrate 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  $^{222}\text{Ra}$  in the air. The measurements of all the networks complies with to the EUROTAM treaty [Dir21].

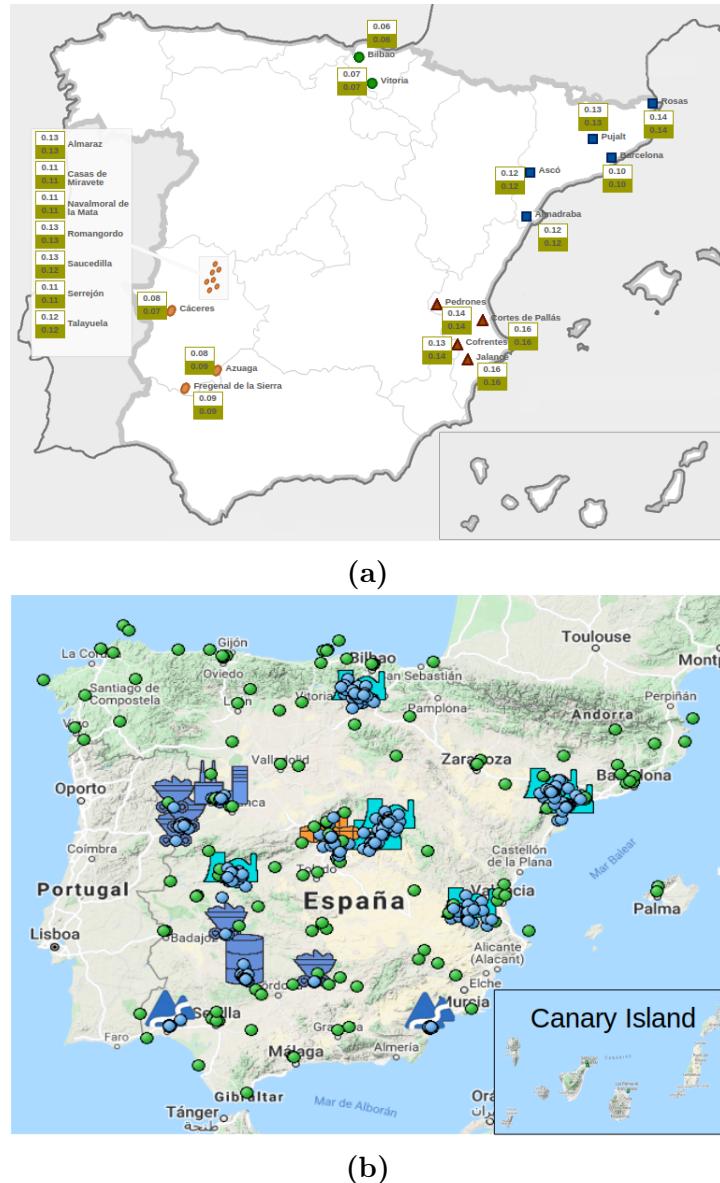
The goal of this thesis and the *TRITIUM* project is to develop a monitor capable of automatically measuring low levels of tritium in water in quasi-real time<sup>4</sup>. This monitor is intended to be finally included in the REA.

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 using the liquid scintillation counter technique (LSC). Due to the limitations of the current tritium detection techniques, which will be described in section 2.1, the *TRITIUM* project has been recently created, the objective of which is to build a tritium detector based on scintillating fibers in contact with the water sample. The photons produced in these scintillating fibers are read out using photosensors, either photomultiplier tubes (PMTs) or silicon photomultipliers (SiPMs).

The *TRITIUM* collaboration is a international group consisting

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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.



**Figure 1.2** – Networks of automatic and sampling stations managed by the Spanish CSN. (Above) Measurement locations 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. (Below) Measurement locations of the REM [CSN21c]. Blue dots are locations near nuclear facilities, and green dots are locations uniformly distributed throughout the country.

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 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 the legal limit of 100 Bq/L specified in the EURATOM Directive 2013/59/Euratom [Dir21]. In addition, this will confirm the correct operation of the Almaraz NPP, since its malfunctioning may produce an increase of tritium activity released.

Tritium is one of the most abundantly produced radioisotope in a NPP, as it was verified in the United States Department of Energy complex, (U.S. DOE) [Berb, Bera] and in several research facilities in China [Hou18], and also places close to them (ground water, surface water and process waste water).

Tritium is produced in the water used for nuclear reactor cooling system of some NPPs by neutron capture of deuterium, existing in the heavy water ( $D_2O$ ), semi-heavy water (HDO) or 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 of the order of  $10^{14} \text{ n cm}^{-2}\text{s}^{-1}$  in the nuclear reactor [?]. This tritium is finally released partially or totally to the environment in a quantity that depends on the reactor type as it is shown in Table 1.2. The most common form in which tritium is released to the environment is HTO [Hou18].

NPPs are operational since more than 60 years and, nowadays, they are essential for providing a large part of the electric power used all over the world (more than 20% in Spain [dE19] and more than a 10% in the world [AIdlE14]). Although the Spanish government is planning to progres-

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].

sively shut down all NPP, there are other countries like China [Pre16] or United States (USA) [New19] that promote their use. NPPs are a profitable investment since they are one of the cheapest source of energy production. Their energy production rate is stable as this doesn't depend on meteorological parameters. Moreover, NPPs do not emit greenhouse gases. Although there are alternative energy sources which are being developed quickly (photovoltaic, wind, tidal energy, etc.), as well as other concepts of energy production and saving (local production, solar roofs, energy efficiency, smart cities, etc.), they are currently not developed enough to fully cover the population needs. On the other hand, NPPs still have some important issues 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, world nuclear energy production is most likely not going to be stopped in the next decade. In fact, the United States Energy Information Administration (U.S. EIA) expects a future increase of nuclear energy production [Cap20]. Therefore the development of different types of alarm systems is an important investment. Safety is not a negotiable aspect and there must be safeguards that warn us of any malfunction of a nuclear power plant. Our 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].

## 1.2 Tritium Properties and Radiological Hazards

Tritium is the only radioactive isotope of hydrogen present in the environment. It was produced for the first time in 1934 in neutron capture of deuterium by Ernest Rutherford, Mark Oliphant and Paul Harteck [MLO34] and it was 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  $4 \cdot 10^6 \text{ Ci/yr}$  ( $1.48 \cdot 10^8 \text{ GBq/yr}$ ), producing a tritium concentration of  $0.6 - 1.2 \text{ Bq/L}$  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  $8 \cdot 10^9 \text{ Ci}$  ( $2.96 \cdot 10^{11} \text{ GBq}$ ), 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 is produced by commercial producers of radioluminescent and neu-

---

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

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

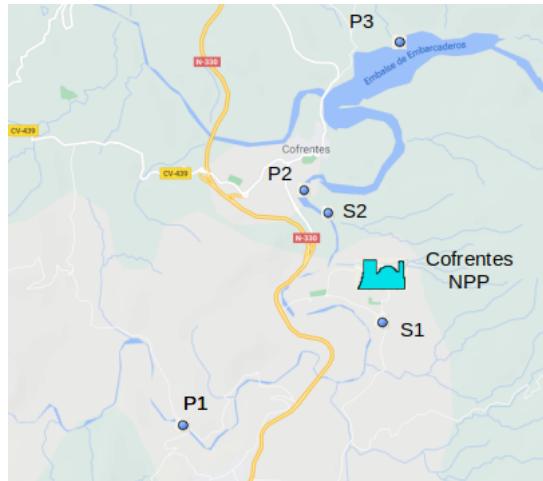
tron generator devices ( $1 \cdot 10^6$  Ci/yr), nuclear power and defense industries (around  $2 \cdot 10^6$  Ci/yr) and several research facilities and nuclear reactors for energy production ( $2 \cdot 10^6$  Ci/GWyr). The production cross sections of the relevant processes are shown in Table 1.3.

Source	Origin	Nuclear reaction	Cross section (b)
${}_1^2\text{H}$	Water coolant	${}_1^2\text{H}(\text{n}, \gamma) {}_1^3\text{H}$	$5.2 \cdot 10^{-4}$
${}_2^3\text{He}$	Helium coolant	${}_2^3\text{He}(\text{n}, \text{p}) {}_1^3\text{H}$	5330
${}_3^6\text{Li}$	Moderator	${}_3^6\text{Li}(\text{n}, \alpha) {}_1^3\text{H}$	940
${}_{10}^5\text{B}$	Moderator, control rods	${}_{10}^5\text{B}(\text{n}, 2\alpha) {}_1^3\text{H}$	3835

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

Tritium levels in the water of the environment, excluding the current 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) [Cal10]. This is attributed to nuclear weapons tests. Tritium levels in rivers around a NPP 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 in the REM data, for example for the case of Cofrentes. Cofrentes is the closest nuclear power plant to Valencia. 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.

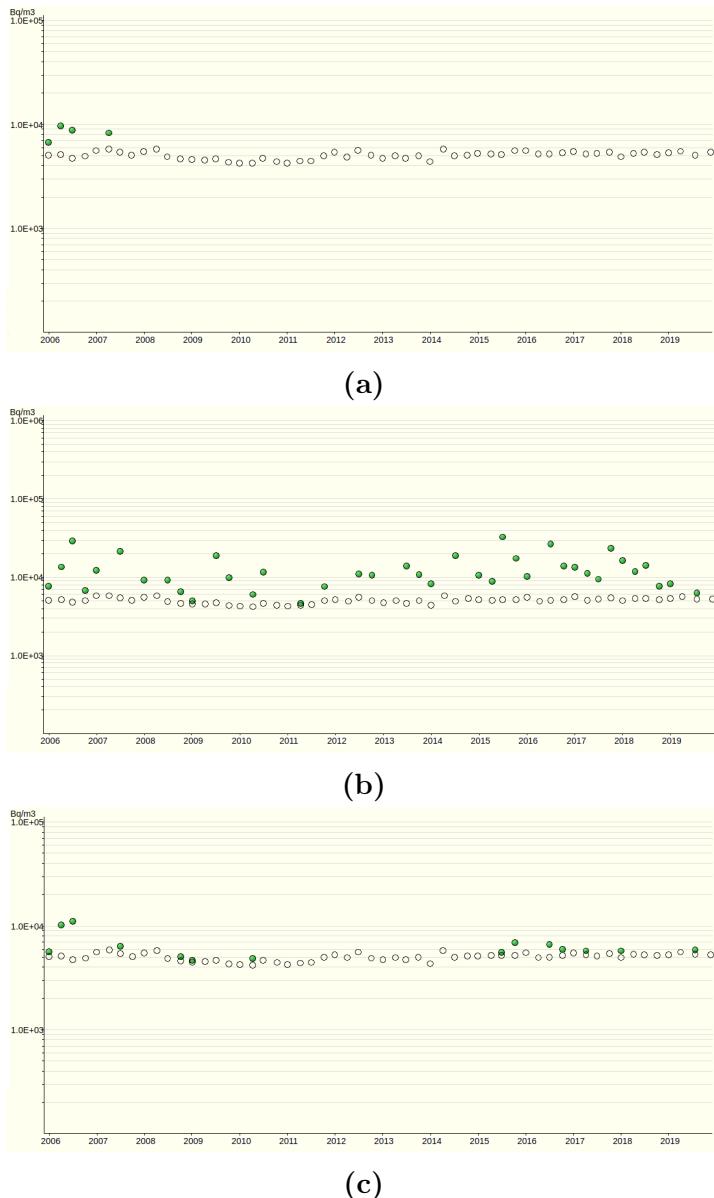


**Figure 1.3 – Tritium sampling locations around Cofrentes NPP.**

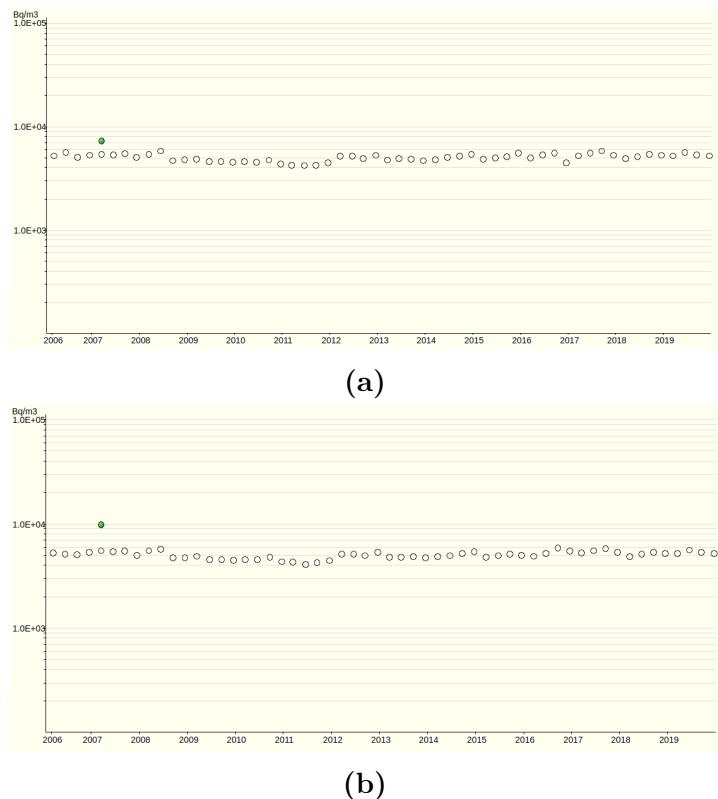
In these figures, the detection limit and the measured activity are plotted with white and green dots, respectively. The measured activity is only displayed when this is larger than the corresponding detection limit. The tritium level in the river increases due to the discharge of the NPP and it is diluted again after 4 km downstream, as can be seen from these data. Two additional measurements of the tritium level in groundwater are included, points S1 and S2 on the map in Figure 1.3, located 1 km before and 1 km after the NPP. Both tritium levels are shown in Figures 1.5a and 1.5b, respectively, where it can be observed that they are below the detection limit.

It is important to note that, although environmental tritium level is affected by the NPP in the case of Cofrentes, these levels are below the maximum allowed limit. The maximum level of tritium measured since January 2, 2006 is around 32 Bq/L, below the limit of 100 Bq/L recommended by the Euratom 2013 Directive.

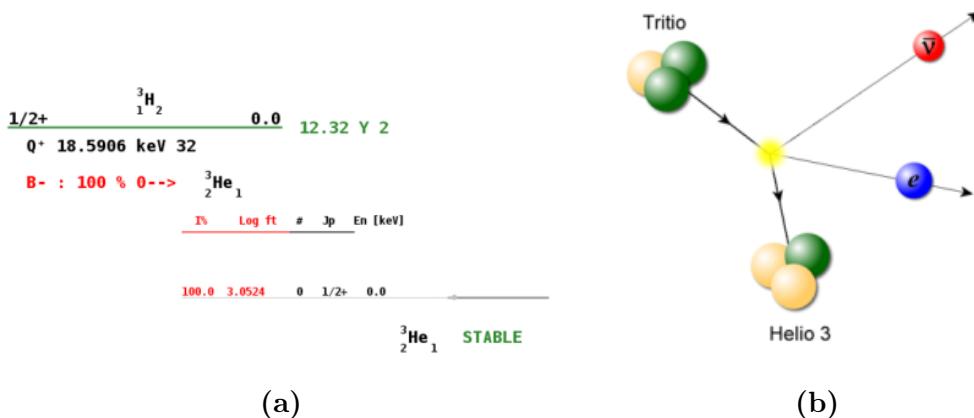
Tritium is a radioactive element with a half-life time of  $T_{1/2} = 12.32$  years. It has one proton and two neutrons and decays exclusively



**Figure 1.4** – Tritium activity levels in surface water around Cofrentes NPP from January 2006 to November 2019. (Above) 6 km upstream. (Middle) 1 km downstream. (Below) 5 km downstream. The white points are the detection limit and the green points are the measured activity, when this is above the detection limit [CSN21c]. The maximum level of tritium measured since of January 2, 2006 is around 32 Bq/L



**Figure 1.5** – Tritium activity levels in groundwater around Cofrentes NPP from January 2006 to November 2019 [CSN21c]. (Above) 1 km before NPP. (Below) 1 km after NPP.



**Figure 1.6** – (Above) Tritium energy levels [IAEA]. (Below) Graphic representation of tritium decay [Pre19].

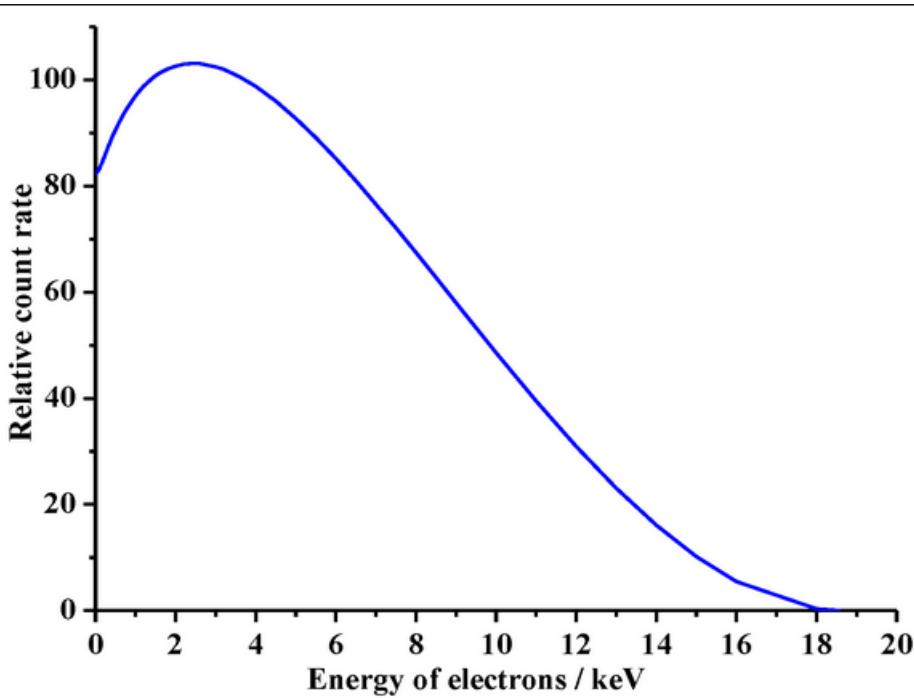
through  $\beta$  radiation. Tritium decays 100% directly to the ground state of the  ${}^3\text{He}$  isotope of helium, which is a stable nuclei, through the process,



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 ( $\sigma \propto 10^{-44} \text{ cm}^2$  [Her07]) and, since  ${}^3\text{He}$  has a much larger mass than electrons and neutrinos, by conservation of energy and momentum, the energy taken by the 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 released energy in tritium decay, is very low. In fact, it is the



**Figure 1.7** – Energy spectrum of tritium electrons [Lin20].

radioactive isotope with the lowest energy released in  $\beta$  decay [USDoEW16]. Consequently, the  $\beta$  particle emitted has a very short mean free path, given in Table 1.4.

This short mean free path is a major issue in tritium detection, as it makes more difficult the electron detection, which requires a highly sensitive detector. It also means that tritium electrons have a low penetration in human body and they are easily stopped by clothes or laboratory gloves, resulting in a low radiological hazard of external tritium. Nevertheless, the danger of tritium increases when this is ingested or inhaled since it binds and perform the same chemical reactions as hydrogen, sometimes with higher rate if the tritium concentration is high enough to catalyze the reaction.

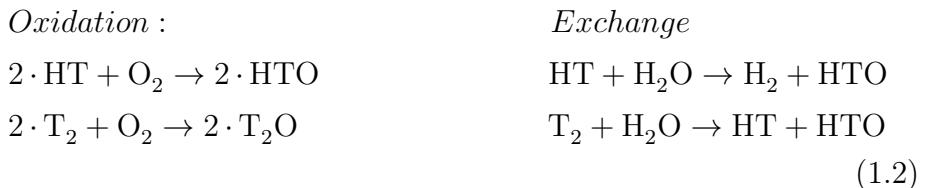
Tritium can be absorbed in our body in three different forms,

Material	P. Depth (5.7 keV)	P. Depth (18.6 keV)
${}^3\text{H}_2$	0.26 cm	3.2 cm
Air	0.036 cm	0.45 cm
Water, soft tissue (solid matter with a 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 STP, standard conditions of temperature (273 K) and pressure (1 atm), and water) [Bla].

gaseous tritium (mainly HT), tritiated water (mainly HTO) and organically bound tritium (OBT).

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



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 ( $\pm 50\%$ ), during which tritium remains in our body

[USDoEW16, Cal10, Mas]. As in the case of 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].

3. Organically bound tritium, normally found in food, generally forms a covalent bond with a carbon. 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 compound 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 may be worse than a similar  $\gamma$  radiations since its biological efficiency<sup>7</sup> is two or three times larger [Str93].

In summary, tritium is a naturally occurring radioactive element. It affects health of living organisms if these are excessively or chronically exposed to tritium. Because of that, several countries have developed a legislation, described in section 1.3, to manage the release of tritium to the environment and ensure that its levels are safe for living organisms.

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<sup>7</sup>The biological efficiency is used to quantify the damage produced in the living cells due to an external radiation.

## 1.3 Current Legislation

Due to the radiological risk of tritium, described above, 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 public health.

The guidelines to limit the radioactive elements in drinking water for many countries. They 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:

1. The justification: The benefit from radiological exposure must outweigh the detriment to health that it causes.
2. The ALARA principle ("As Low As Reasonably Achievable"): The radiological exposure must be kept as low as possible considering social and economic factors.
3. The 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 corresponds to less than 5% of the annual dose due to background radiation, 2.42 mSv/year.

The guideline reference level of each radionuclide in drinking water,

GL, is usually calculated from these recommendations using the equation,

$$GL(\text{Bq/L}) = \frac{RDL}{DCF \cdot q} \quad (1.3)$$

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

The GL calculated for tritium in drinking water according to the ICRP and WHO recommendations is  $76\ 103 \text{ Bq/L}$  and  $7\ 610 \text{ Bq/L}$  respectively. It means that tritiated water with activities below these values is considered not harmful for health.

Based on these recommendations, each country has created organizations in charge of developing its own legislation limits of exposure to radionuclides. In Spain, the responsible organization of this task is the CSN. Most of the countries in the world implement the RDL of  $0.1 \text{ mSv/yr}$  recommended by the WHO. The legal limit for tritium in drinking water in this case is  $7\ 610 \text{ Bq/L}$  but it is often approximated in different ways. Some countries like Switzerland [Dfdl06] or some organizations like the WHO [WHO04] take this value as  $10\ 000 \text{ Bq/L}$ . Others like some territories of Canada, such as Ontario and Québec, truncate this value to the first number  $7\ 000 \text{ Bq/L}$  [otE94, Que16]. There are other countries like Russia which use the much more accurate approximation value of  $7\ 700 \text{ Bq/L}$  [ICfS07]. There are other countries like Australia that prefer to implement the RDL of  $1 \text{ mSv/yr}$ , recommended by the ICRP, the legal limit of which is  $76\ 103 \text{ Bq/L}$  [AG21]. Other countries like Finland are based in the ICRP recommendations and use only half of this value,  $0.5 \text{ mSv/yr}$ , rounded to a legal limit of  $30\ 000 \text{ Bq/L}$  for tritium in drinking water [NEA93].

There are two different exceptions to these recommendations:

1. 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 carried out on the available data. [USEPA76].
2. Most of the EU countries, such as France, Germany or Spain, consider a 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 in water (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 several countries.

## 1.4 This Thesis

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

1. **Chapter 1** provides a brief introduction to tritium detection, reports some important properties of tritium, and discusses the current legislation that limits tritium levels for human consumption in many countries around the world.
2. **Chapter 2** describes the State-of-the-Art of tritium detection and shortly introduces the TRITIUM project.
3. **Chapter 3** outlines the different parts of the TRITIUM monitor, which are the ultrapure water system, the background rejection system (consisting of the lead shielding and the active veto) and the tritium detector.
4. **Chapter 4** reports the calibrations of the different parts of the TRITIUM monitor and describes the developments aimed at improving the efficiency of tritium detection.
5. **Chapter 5** details the geometrical configuration of the different prototypes built in the TRITIUM project.
6. **Chapter 6** details the Monte Carlo simulations performed in the TRITIUM project.
7. **Chapter 7** and **Chapter 8** report the results achieved for the built prototypes and the simulations carried out, respectively.
8. **Chapter 9** summarizes the most important results achieved in this work.



# Chapter 2

## Tritium Detection Systems

### 2.1 Tritium Detection 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 is carried out with different available technologies 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 milliliters 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 on the activity of the samples [AH99, Hof92a]. In this technique, the  $\beta$  particles emitted from the sample excite 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

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

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

with photosensors, which convert the optical signal into a 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 disadvantage of long measurement time (up to 2 days) and producing chemical waste, since liquid scintillator contains toluene which is toxic. In addition, this technique requires special staff for sampling, chain of custody and laboratory analysis which require economical and time resources. In order to overcome these difficulties some efforts have been made in order to build a tritium monitor with LSC but without achieving a low enough LDL [Sig94].

The ionization chamber technique (IC) consists of a gas chamber, filled with gas (sample), which contains electrodes that collect the ionization current produced by the energy deposition of the  $\beta$  radiation in the gas. It is a simple and fast system, but it has a high LDL ( $> 10 \text{ kBq}$ ) and requires the samples to be in a state of gas or steam [Khe02, ZC13]. The ionization chamber technique also requires sample conditioning, chain of custody and laboratory analysis.

The calorimetry method is based on the measurement of the heat generated in the detection medium (normally platinum) [CGA17, BD13]. The disadvantage of this technique is its high LDL, of the order of a 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 coupled to a PMT [Mat07, Mat08] or Silicon Drift Detector (SDD) [Nie15]. The problem with this technique is its high LDL, of the order of MBq.

There are additional methods for tritium detection, although they are less employed or less 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 for sample conditioning [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 activity measurement. Therefore, they cannot be used for in-situ monitoring of tritium in water. The liquid scintillation technique is the only one with sufficiently small LDL to fulfill the requirement of 100 Bq/L of tritium in 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 100 Bq/L in quasi-real time. There are several studies 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 coincidence [Osb70].
4. The study done by A. N. Singh, M. Ratnakaran and K. G. Vohra in 1985, that used a scintillator with several holes 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,  $\varepsilon_{det}$ , of the order of  $10^{-3}(\frac{cps}{kBq/L})$  [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: Efficiencies of different scintillator detectors 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 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 factors 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 detectors, which is the intrinsic

detector efficiency normalized to its active surface. It can be checked that these specific efficiencies are quite similar. 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. The development of a detector with a much lower LDL is thus essential to comply with the EURATOM directive of 100 Bq/L of tritium in water for human consumption.

## 2.2 The 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 high LDL or they work in off-line method.

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 an international consortium of six different European institutions from three European 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, The *Junta de Extremadura* and The University of Valencia, in Spain.

This project was funded by the Interreg Sudoe program of the European Economical Community, EEC, in year 2016 call, with the reference number SOE1/P4/EO214. The purpose of this project is the development of an automatic station for in-water tritium monitoring, in situ and in quasi-real time. The tritium detector consists of a bunch of scintillating fibers in contact with the tritium water sample which are in charge of detecting the

tritium decay electrons. These fibers are read out with several photosensors (photomultiplier tubs, PMTs, or silicon photomultiplier arrays, SiPM) in time coincidence. The specific efficiency obtained by Moghissi for scintillating fibers is sufficiently high to justify our choice of scintillating fibers as a detection medium. Additional elements are used to improve the tritium detection sensitivity such as a ultrapure water system, which prepares the water sample before introducing it in the detector for tritium measurement and a cosmic veto and a passive shielding, which reduces the natural radioactive background of the tritium detector. Several electronic modules which control 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 background events are significant . To reduce the background counts of TRITIUM monitor, coincidence techniques are employed.

The TRITIUM monitor will be installed in the Arrocampo dam, Almaraz (Spain), displayed in Figure 2.1, where the Almaraz NPP releases the water from its secondary cooling circuit. 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 length of 1007 km. This river, shown in Figure 2.1a, rises in Aragon (Spain) and flows into the Atlantic Ocean, through Lisbon (Portugal). The water of this river is used for agriculture and drinking water by both Spanish and Portuguese people. For this reason, an international cooperation is necessary in order to control and maintain the quality of the tagus river water.

Each institution of TRITIUM collaboration has focused its efforts in the development of a different part of this project:

1. The Extremadura group has developed and installed the ultrapure wa-



**Figure 2.1** – (Above) Arrocampo dam and Almaraz Nuclear Power Plant. (Below) Tagus river along Spain and Portugal.

ter system to produce water with very low conductivity,  $\sigma \approx 10 \mu\text{Sv}/\text{cm}$  (two orders of magnitude less than 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 the TRITIUM detector very clean, which is critical for its long-term functionality. On the other hand, to reduce the natural background since several natural radioactive isotopes present 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 developed the passive shielding for the detector. The shielding is made of 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 groups have collaborated for designing, developing and building four different prototypes of the TRITIUM detector and active vetos for reducing cosmic events. These prototypes and vetos are explained in chapter 5 and section 3.4.2 respectively. They have also carried out simulations of this system. These simulations are explained in chapter 6.

The important characteristics of the TRITIUM detector must have are:

1. *Compactness.* Compactness is an important requirement because in the place where the detector is planned to be installed, Arrocampo, there is little space. Compactness also allows portability and cost reduction.
2. *Modularity.* The modularity of the TRITIUM detector is important for its flexibility in the geometrical configuration and to improve its tritium detection sensitivity, which increase in a scalability way. Modularity also makes it easy its construction and maintenance..
3. *Thin active volume and large active area.* The mean free path of the  $\beta$  particle of tritium decay is very short so thin detector active volumes are needed. In practice, active thickness beyond the mean free path of the tritium electrons only contributes to background. In addition, as reported 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.
4. *High efficiency detection for tritium.* As the tritium activities to be measured are very low, we cannot afford the loss of tritium events as it strongly affects the reliability of the tritium measurement..
5. *High specificity to tritium.* The monitor has to be able to distinguish the tritium signal from the signal due to other radioactive elements present in the sample.
6. *Quasi-real time response.* It is important that the system operates in quasi-real time (1 h or less) in order to detect any anomalous tritium release as fast as possible.
7. *Ruggedness.* The final goal of the project is to install an automatic system working during a number of years requiring only scarce intervention of specialized operators. 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 where the water sample is taken. Working *in situ* has some advantages such as: 1) Faster and cheaper maintenance, since the sampling process, chain of custody, etc. are eliminated, 2) Continuous measurements are carried out and 3) Safer monitoring since personal exposure dose is reduced, 4) Changes in activity levels can be detected quickly.



# 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 it is essential to avoid deposition of particles onto the fibers since 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 few 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  $200 \text{ MeV/nucleon}$ . The second part is an active veto (section 3.4.2), consisting of two plastic scintillation plates located inside of a passive shielding, above and below the TRITIUM detector and read by 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 stop. 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,  $100 \text{ Bq/s}$ , is exceeded.

The different parts of the 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

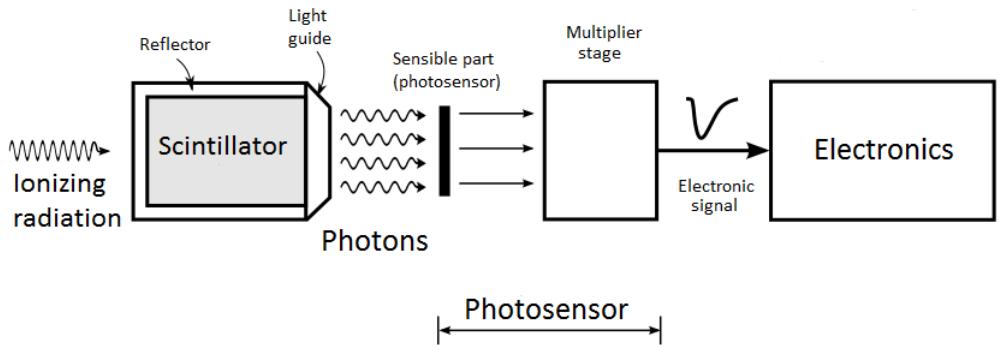
As discussed in section 2.1, the TRITIUM consists in a chain of three main elements:

1. The scintillator, that detects the tritium event. Ionizing radiation hits this material and deposits kinetic energy through ionization and excitation processes. Part of the absorbed energy is converted into photons, mostly in the visible range<sup>1</sup>. The produced photons carry information about the particle detected, like its energy, type, etc.
2. 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 and transform them in electrons with a multiplication factor of around  $10^6$ . These electrons form a electronic pulse than gives information of the detected photons.
3. The electronic system, which is the part of the scintillator detector in charge of processesing and analyzing (first analogically and later 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.

---

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



**Figure 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 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), which are roughly proportional [Kno99, Leo94]:

$$\frac{dE}{dx} \approx \left( \frac{dE}{dx} \right)_c + \left( \frac{dE}{dx} \right)_{br}; \quad \frac{\left( \frac{dE}{dx} \right)_{br}}{\left( \frac{dE}{dx} \right)_c} \approx \frac{EZ}{700} \quad (3.1)$$

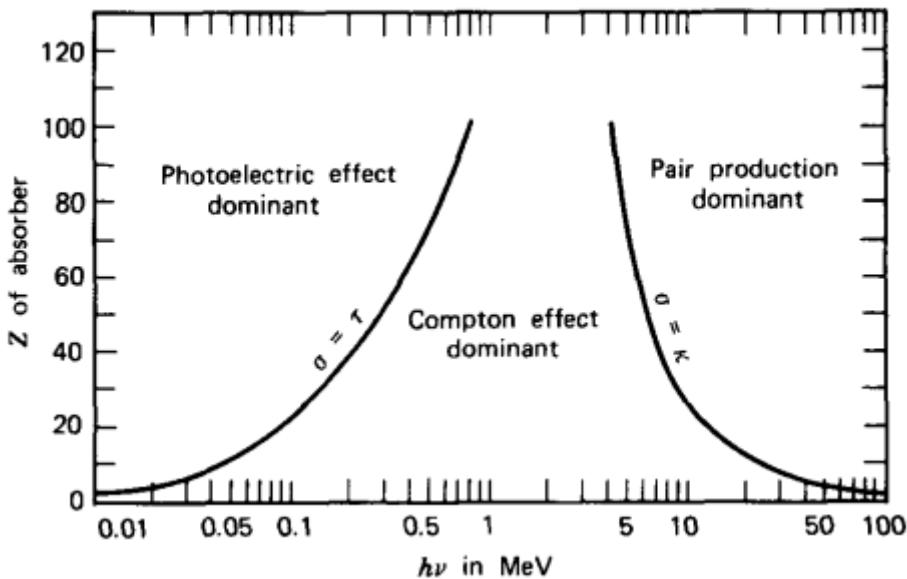
where  $E$  is the energy of the electron in MeV and  $Z$  is the atomic number of the absorbing material. Due to this energy loss, the electrons can only penetrate a material as far as they go before losing their total kinetic energy. This distance is known as range and, in the case of tritium electrons,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

As photons do not have charge, their possible interactions with matter are photoelectric effect, Compton effect, coherent scattering and pair production and the probability of each process, displayed in Figure 3.2, depends on the energy of the photon,  $E_\gamma = h\nu$ , and on the atomic number of the material,  $Z$ . 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 production process does not play any role since this requires a photon energy equal or more than 1.022 MeV.

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]:

$$E_e = E_\gamma - E_b \quad (3.2)$$

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 matter through the variable  $Z$ , and the energy of the electron according to the expression [Kno99]:



**Figure 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].

$$(Pr)_{Ph-eff} \approx \frac{Z^n}{E_\gamma^{3.5}} \quad (3.3)$$

Thus, the photoelectric effect is most probable for elements with high atomic number. This is the reason why this type of elements 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]:

$$E_e = \frac{\frac{E_\gamma^2}{m_o c^2} (1 - \cos\theta)}{1 + \frac{E_\gamma^2}{m_o c^2} (1 - \cos\theta)} \quad (3.4)$$

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 it 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 conserve 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. Scintillators convert kinetic energy of the incoming particles into light<sup>2</sup> which can be detected and quantified. Light emission is produced due to the photon de-excitation of scintillating 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

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

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 identical events emit a different number of photons that follows a Poisson statistics.

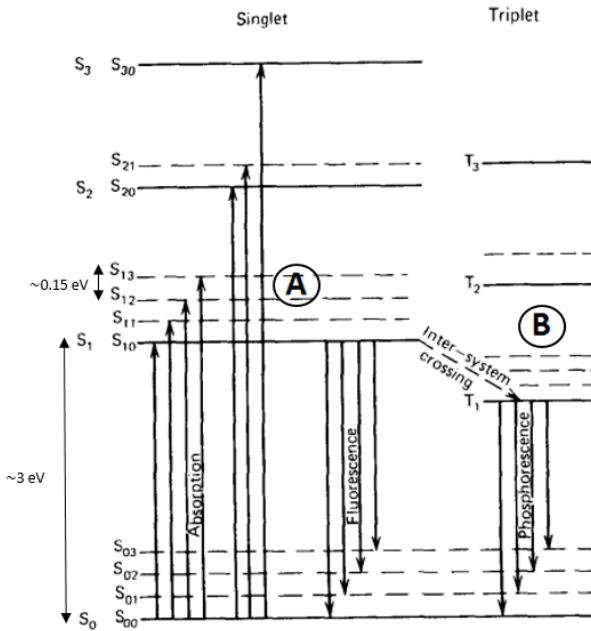
Scintillators can be organic and inorganic. Inorganic scintillators normally have a higher atomic number and density, so their light output is higher. For these reasons they are better for gamma-ray spectroscopy. Organic scintillators are generally faster and they are commonly used for beta spectroscopy 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  $C_{18}H_{14}$ ,  $C_{24}H_{22}N_2O$  or  $C_{15}H_{11}NO$  with an average atomic number between 3.5 and 5. The scintillator molecules of organic scintillators have a  $\pi$ -electron structure. The energy levels of their electrons are commonly illustrated with a Jablonsky diagram, shown in Figure 3.3. This diagram 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 (the visible range). As it is shown in the figure, each energy state is splitted in close sublevels separated around 0.15 eV. This fine energy structure is due to excitations of molecular vibrational modes tagged 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>.

When a particle deposits its kinetic energy in a scintillator, the valence electrons are exited to higher singlet energetic states very fast (times 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-excite to the fundamental single state,  $S_{00}$ ,

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



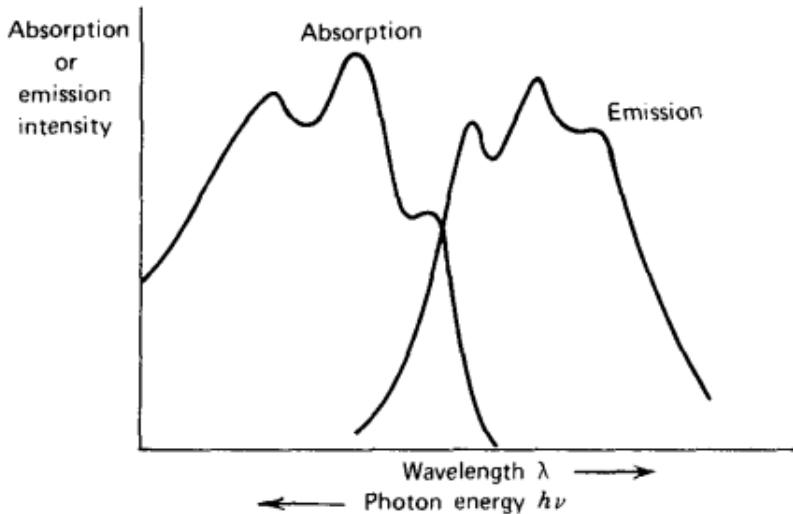
**Figure 3.3 – Jablonsky diagram [Kno99].**

through three different physical mechanisms:

1. 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 energy. This effect is called Stokes shift and it is represented in Figure 3.4.

The intensity of the fluorescence emission in an organic scintillator



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

versus time is the 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].

$$I = I_0 (e^{t/\tau} - e^{t/\tau_1}) \quad (3.5)$$

2. 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.
3. 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, the electron interacts with another electron in a similar state, falling to the first singlet state and quickly de-exciting to the ground state.



This emission has the same emission spectrum as immediate fluorescence, but with a longer lifetime.

As the prompt fluorescence light produces the scintillator signal, the detector design should optimize its detection and reduce other possible physical mechanisms. One of the most important parameters is the scintillation yield<sup>4</sup>, defined as the number of photons emitted per unit of absorbed energy. This yield depends on the type of particle and on other mechanisms that do not produce prompt fluorescence, like phosphorescence, delayed fluorescence, 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 [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 scintillating 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.

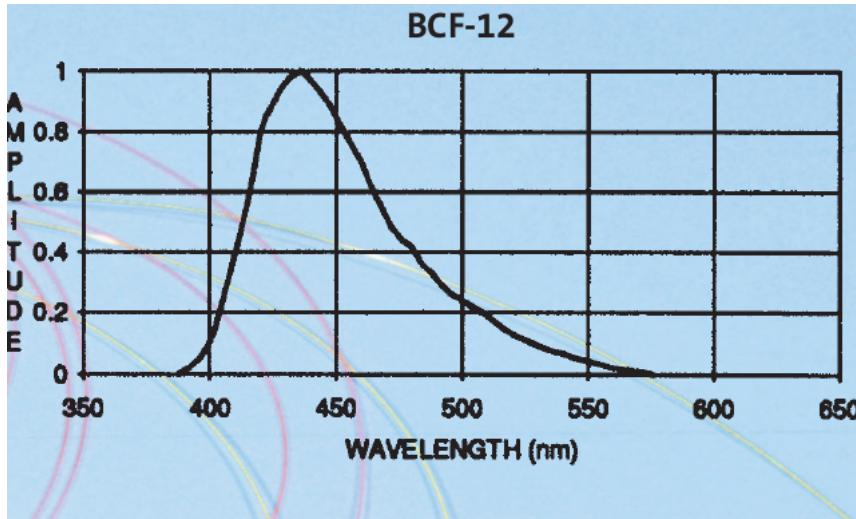
When a particle deposits all or part of its kinetic energy, some photons are produced in the fiber core as a result of the scintillating process. The number of photons produced depend on the scintillation efficiency and its value is around 2.4% for the fibers used (BCF-12), which means that a scintillation yield of about 8000 photons will be produced per MeV for

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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.

<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

a mip. For instance, for tritium electrons, these fibers release a maximum of around 148 photons (when a 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.



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

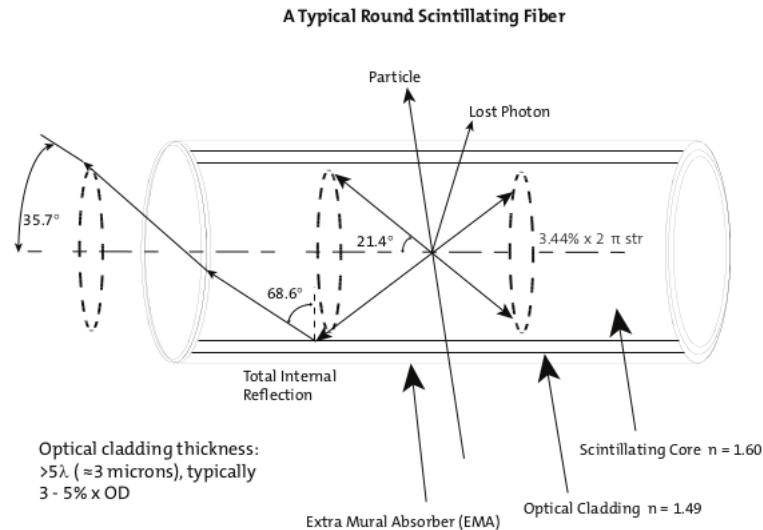
The scintillation light is guided to the sensitive part of the photosensor. A single photon produces a signal with some probability, called the quantum efficiency. Fibers (and scintillators in general) use the optical property of Snell's law [Bor19] to guide their photons to the desired part (ends of the fibers). The guiding mechanism is based on the interface created between the core and the surrounding material. When a photon hits this interface, it is refracted (and therefore lost) following the Snell equation, 3.7 [Bor19].

$$n_0 \sin(\theta_0) = n_1 \sin(\theta_1) \longrightarrow \theta_c = \arcsin\left(\frac{n_1}{n_0}\right) \quad (3.7)$$

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 this efficiency 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 unclad 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 along the 25 cm fiber length of 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.



**Figure 3.6 – Photon collection in a single clad fiber [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 with its corresponding loss of light. Three different cases are shown in Table 3.1, where the cladding effect is illustrated.

Material	Refractive index	critical angle (°)
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.

In the practice, it is difficult to achieve a perfect air-core or water-core interface, and this affects light collection. As commercial claddings are thicker (30  $\mu\text{m}$ ) than the mean free path of tritium decay electrons in water (around 5  $\mu\text{m}$ ) cladded fibers is not an option for the TRITIUM detector. Hence, special attention is needed for achieving a water-core interface good enough. To achieve this goal a special protocol was developed in the ICMOL laboratory for preparing fibers for tritium detection, detailed and tested in section 4.1.4. 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 their number, detection time, etc. There are many available photosensors that rely on various physical processes, such as photoelectron multiplier tubes (PMTs), silicon photoelectron multipliers

Property	Value
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 lenght (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].

(SiPM) or charge-coupled devices (CCD).

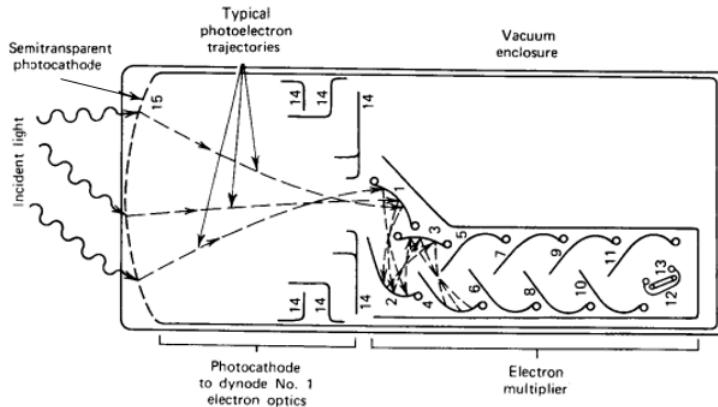
The optimization of the efficiency of a scintillation detector is essential. To do so, the emission spectrum of the scintillator (Figure 3.5 for the fibers used) must overlap as much as possible with the detection efficiency spectrum of the photosensor chosen. The detection efficiency spectrum gives the probability of detecting photons as a function of wavelength. The efficiency of a detector is proportional to the product of both, the emission and the 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%, a high gain (multiplication factor of  $10^6$ ) and need 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 tubes are employed as photosensors in nuclear physics since decades. They detect the scintillating photons that reach its sensitive part, the photocathode, and produce an electronic signal, large enough to be easily measured. In Figure 3.7 a schematic drawing of a PMT is given. This consists of a vacuum tube that has a glass window through which photons can penetrate. The electrons created in the photocathode travel in vacuum.

The signal production has two phases:



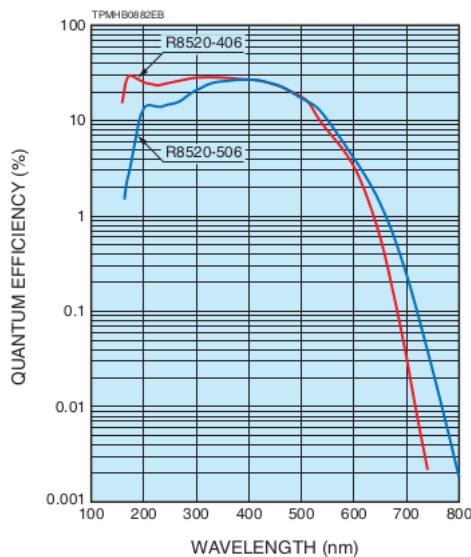
**Figure 3.7 – Scheme of a PMT [Kno99].**

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 the TRITIUM experiment are the model R8520-406 from Hammatsu [K.K19] and the material of their photocathode is Bialkali<sup>6</sup>.

The response of the PMT for long wavelengths is inhibited, 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 inhibited 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. The quantum efficiency (QE) spectrum, shown in Figure 3.8 for the PMT used in TRITIUM, is defined as the ratio of the number of photoelectrons produced at the cathode of the PMT and the number of photons reaching it.

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

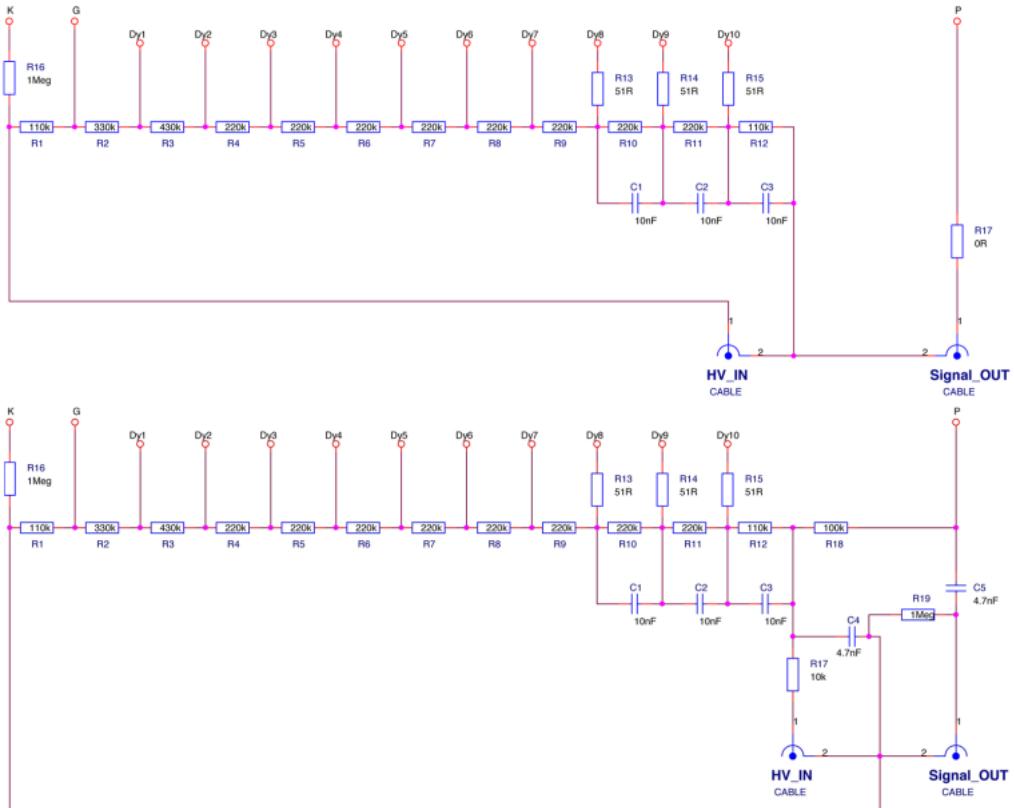


**Figure 3.8** – Quantum efficiency spectrum for the PMT used (R8520-406) [K.K19].

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 by us). 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 positions of both peaks are very close, 435 nm and 420 nm for fibers and PMT respectively.

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 an electronic circuit. A positive HV, grounded in the ph-

tocathode, is convenient 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.



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

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. This 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 usually 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]:

$$G_{PMT} = CE \cdot \delta^N \quad (3.8)$$

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

The multiplication stage adds an uncertainty to the measurement. Working without gain allows us 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 from the photocathode. This special setup was used for fiber characterization, described in 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,  $G_{PMT}$  new electrons are created with a variance of  $\sqrt{G_{PMT}}$ .

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 characteristics.

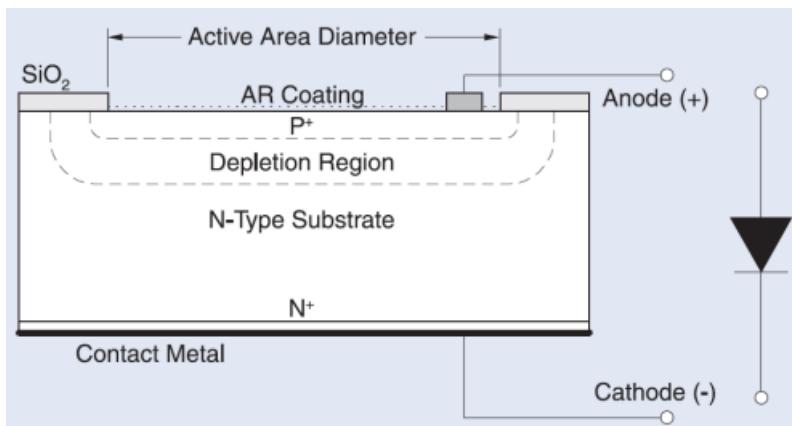
The photocathode may emit electrons in absence of any scintillation light. This signal, called dark current,  $I_{DC}$ , can arise due to thermoionic emission. For the PMTs employed in this work, this value is around 2 nA according to their data sheet.

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

SiPMs are 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.



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

The voltage at which the SiPM starts operating in Geiger mode is called the breakdown voltage,  $V_{BD}$ . At lower voltages, SiPMs work in

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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 sublevels in the forbidden energy gap.

proportional mode in which the signal of the pixel is proportional to the energy deposited but their 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 SiPMs depend on the overvoltage,  $V_{ov}$ . The overvoltage is the voltage applied to the SiPM above their own breakdown voltage and this is expressed as:

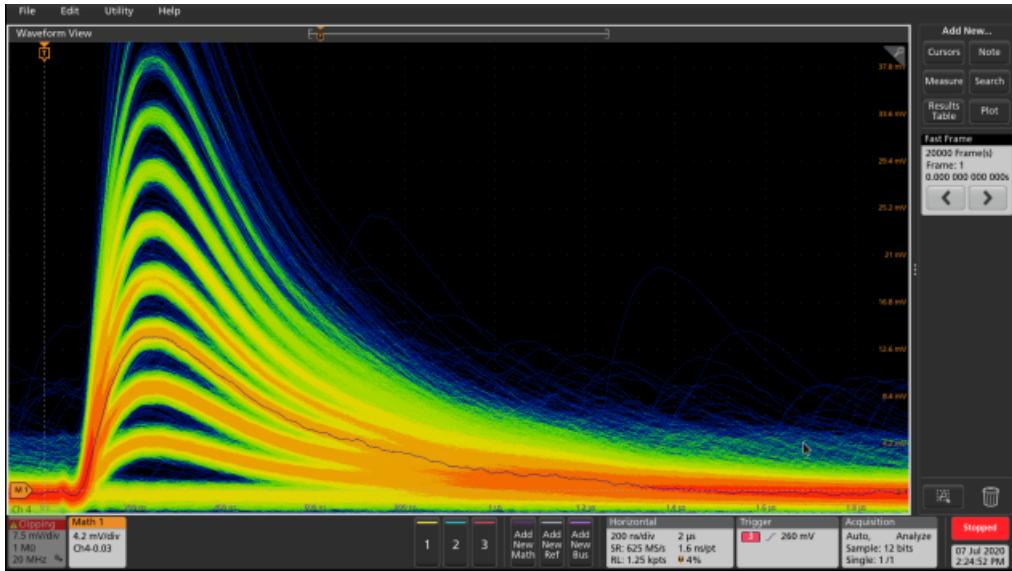
$$V_{bias} = V_{BD} + V_{OV} \quad (3.9)$$

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 detects one photon, the number of detected photons is linearly proportional to the value of the output signal. Hence, after a correct calibration of SiPMs, described in section 4.2, the linearity of the SiPM output signal versus the deposited energy of tritium events is recovered.

The size of a SiPM pixel should be very small<sup>8</sup> to make sure that, for low enough photon fluxes, only one photon is detected in each pixel. If the photon flux is high (typically several thousand of photons per event) more than one photon will impinge on the same pixel, but the output signal would be that of 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 far

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



**Figure 3.11** – SiPM output pulses displayed on oscilloscope, model MSO44X from Tektronix [Tek21]. To easily observe several heights of these pulses, associated with a different number of SiPM pixels fired at the same time, the persistence function of the oscilloscope is used.

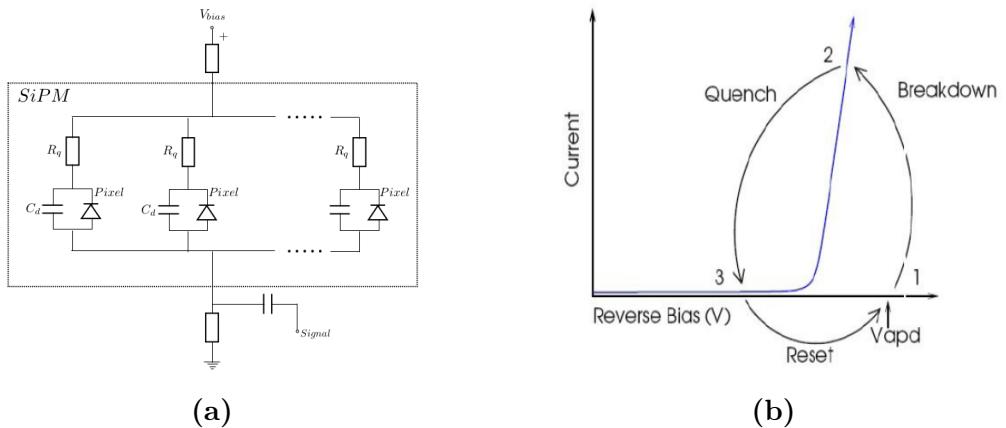
from producing so many photons.

The SiPM can be modeled as an electric circuit, shown in figure 3.12a, in which, due to the charge distribution in the depletion zone, a capacitance is induced by the SiPM. This looks like a reverse diode in parallel with a capacitor of capacitance  $C_d$ . When the pixel detects a photon, the capacitor is discharged, creating an output current (electronic pulse).

In addition, each pixel of a SiPM has a quenching resistance<sup>9</sup> in series,  $R_q$ , that stops the current produced when this pixel is fired. When the discharge is produced, a current flows through the resistance, reducing the reverse voltage seen by the diode below the breakdown voltage. Then, the current that flows through the diode is stopped and the voltage seen by

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<sup>9</sup>The typical value of this quenching resistance for commercial SiPMs is around 500 kΩ



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

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 photon detection is characteristic of a RC circuit, described by the equation:

$$V_{bias}(t) = V(t_0) \left(1 - e^{-t/\tau}\right) \quad (3.10)$$

where  $\tau$  is the recovery time constant of the system, given by  $\tau = C_d \times R_q$ . 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 (typically of the order of  $10^6$ ) 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 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°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 in section 4.2.

An important parameter for the 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 a incident photon generate an electron-hole pair, and the probability that the generated electron or hole produces an avalanche,  $P_{av}$ .

$$PDE = FF \times QE \times P_{av} \quad (3.11)$$

Like PMTs, sometimes the SiPM produces pulses that are unrelated with any incident photon. The current due to these pulses is called the dark current rate. It depends on temperature. At temperatures around 25°C, these pulses are mainly produced by the thermal generation, i.e., when a free electron or hole start an avalanche. The dark current signal is identical to the signal produced by a single photon, so they cannot be discriminated. Therefore, it is very important to determine the importance of the dark current 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

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<sup>10</sup>Around 20 secondary optical photons are emitted in each SiPM output pulse with gains of the order of  $10^6$  [Spi97]

new avalanches. This effect, called optical crosstalk, produces signals that are larger than those corresponding to single photons, producing erroneous information about the number of photons detected. The probability of producing an optical crosstalk event depends on the number of electrons produced in the avalanche (gain) and, therefore, on the temperature and the overvoltage. This probability at the recommended overvoltage and temperatures around 25°C is typically less than 10%.

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 a new avalanche which will be seen as a new event. This events, called afterpulses, are often emitted around 1  $\mu$ s after SiPM output pulses. The afterpulse probability was not measured since it is not interesting for the TRITIUM project. The reason is that the TRITIUM detector uses the SiPM as a counter (without integrating the signal). In addition, time coincidences are made using time windows of 10 ns. At this level, the afterpulse probability is negligible since it normally happen 1  $\mu$ s after the SiPM output pulse.

The initial SiPM cadidate for TRITIUM project and the one which was characterized is the model S13360-1375 from Hamamatsu Photonics [Div16b] because this model has interesting characteristics and properties, shown in Table 3.3. This model was mainly choosen due to its large pixel size, 75  $\mu$ m, which implies a high PDE and a high gain, both important parameters for the TRITIUM project due to the low activity to be detected and to the small signals produced by tritium events. High PDE and high gain are achieved at the cost of reducing the dinamic range, which is not an

issue due to the small signals generated by tritium events.

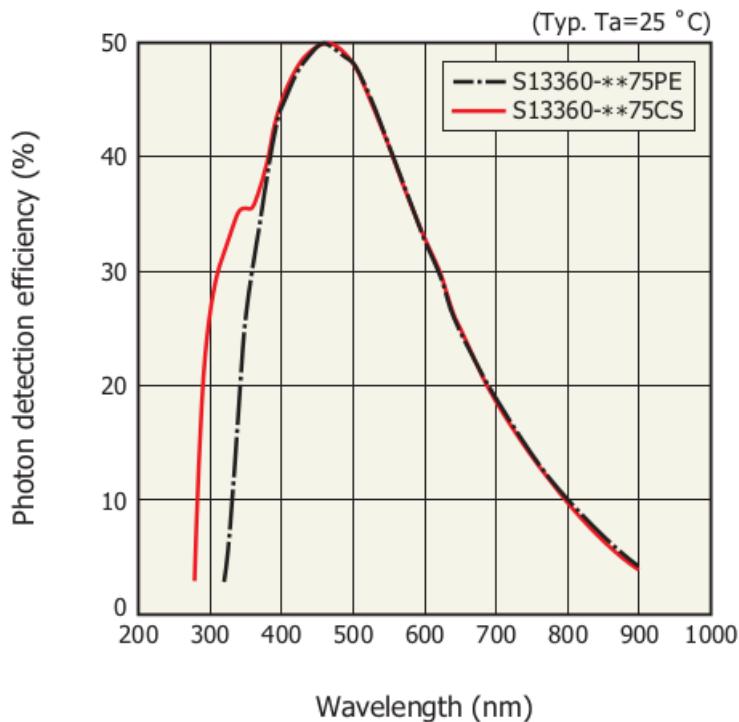
Parameter	S13360-1375	S13360-6075
Series	S13360	S13360
Model	1375	16075
Pixel Pitch ( $\mu\text{m}$ )	75	75
Effective photosensitive area ( $\text{mm}^2$ )	$1.3 \times 1.3$	$6.0 \times 6.0$
Number of pixels	285	6400
Fill factor	82%	82%
Refractive index of windows material	1.55	1.55
Operating temperature range ( $^\circ\text{C}$ )	$[-20, 60]$	$[-20, 60]$
Spectral response range, $\lambda$ (nm)	[320, 900]	[320, 900]
Peak sensitivity wavelength, $\lambda_p$ (nm)	450	450
PhotoDetection Efficiency, PDE, $\lambda = \lambda_p$ (%)	50	50
Dark counts, Typical/Maximum (kcps)	90/270	2000/6000
Terminal capacitance, $C_t$ (pF)	60	1280
Gain, M,	$4 \cdot 10^6$	$4 \cdot 10^6$
Breakdown Voltage, $V_{BD}$ (V)	50.97	53
Cross talk probability(%)	7	7
Temperature coefficient $\Delta TV_{op}$ (mV/ $^\circ\text{C}$ )	54	54

Table 3.3: Characteristics of SiPM S13360-1375 and S13360-6075 from Hamamatsu Photonics [Div16b].

These parameters quoted in Table 3.3, provided by Hamamatsu photonics, are only an approximation for a given element. They can vary significantly even for SiPMs of the same model, so they must be measured for each SiPM used. The measurement of some important characteristics for the TRITIUM project is reported in section 4.2.

This SiPM was also chosen because, as it can be observed in Figure 3.13, its maximum PDE is reached at  $\lambda_{p,SiPM} = 450$  nm, which is very close to the peak of the emission spectrum of the scintillating fibers used,  $\lambda_{p,fiber} = 435$  nm.

This SiPM was later replaced by the more performant model S13360-



**Figure 3.13** – Photon detection efficiency (PDE) spectrum for SiPM S13360-\*\*75 models [Div16b].

6075 from Hamamatsu Photonics [Div16d], whose properties are also listed in Table 3.3. The only difference between both models is their active area ( $6 \times 6 \text{ mm}^2$ ) that allow to read more scintillating fibers with the second model. This improvement is achieved at the price of a higher dark count rate (typically 2 Mcps). In addition, commercial matrices are available, with a total area of  $24 \times 24 \text{ mm}^2$ .

Although TRITIUM detector uses whole SiPM matrices, the characterization 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 A, is already prepared to 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 its advantages and disadvantages, so both were tested to decide the most suitable. The output signal of both photosensors is proportional to the number of incident photons in our range of luminosity and they have a similar gain (of the order of  $10^6$ ). Both properties are essential to detect tritium events and to obtain a large enough signal to be measured and recorded. Both photosensors have fast output signals, with a rise time of the order of 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, of 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 electron detection efficiency at 420 nm, achieved with SiPM is higher, around 50%, than with PMT, about 30%. A large efficiency is essential because the number of photons produced in a tritium event is rather 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 the TRITIUM detector should work during years. Fur-

thermore, PMTs are rather more expensive, than SiPMs. In addition, PMTs are affected by magnetic fields, contrary to SiPMs that work correctly in magnetic field up to 7 Tesla. Moreover, due to their high uniformity, SiPMs are capable of measuring the exact number of photoelectrons detected and even of resolving a single photoelectron, which is not possible with PMTs due to their gain uncertainty.

However, 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 their size, and this happens with SiPMs almost entirely at the level of a single photoelectron. This prevents to separate tritium decay signals from background in the singel photon-detection zone. Another inconvenient of SiPMs is their large crosstalk and afterpulses that need to be corrected. An additional drawback of SiPMs is that their response depends strongly on temperature. As the TRITIUM detector will be installed in an environment with significant temperature variations, this problem is solved by developing a stabilization method of the SiPM gain.

### 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 tritium detection. This electronic system depends on the type of output information that is desired and on the detector configuration used.

#### Electronics for PMTs

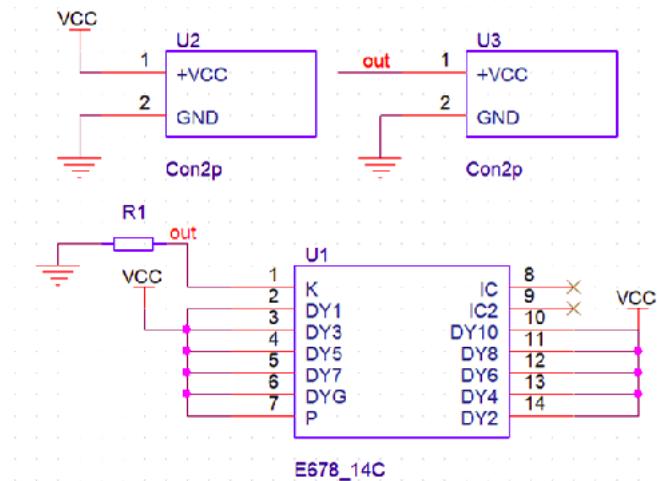
PMTs were used in TRITIUM experiment for two main objectives. On the one hand, to know the amount of incident photons that reach the PMT

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

photocathode, which is important to characterize fibers, and, on the other hand, to know the energy of events, which allows us to discriminate events according to their origin, obtaining an energy spectrum of triggered events.

To know the amount of photons that reach the photocathode, the PMT should work without internal gain since it introduces a large uncertainty in the measurement. For that end, the electron multiplication stage (shown in section 3.2.3) should not be employed. This was achieved with the help of a PCB, shown in Figure 3.14, designed, built and tested for this purpose.



**Figure 3.14** – Electronic scheme of the voltage divider circuit used for working with PMTs without 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 the usual running voltage because this voltage is only needed to create a voltage difference between the photocathode and the first dinode. As the electrons are 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 Pi-

coammeter/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). Finally, the number of photons reaching the photocathode is calculated from,

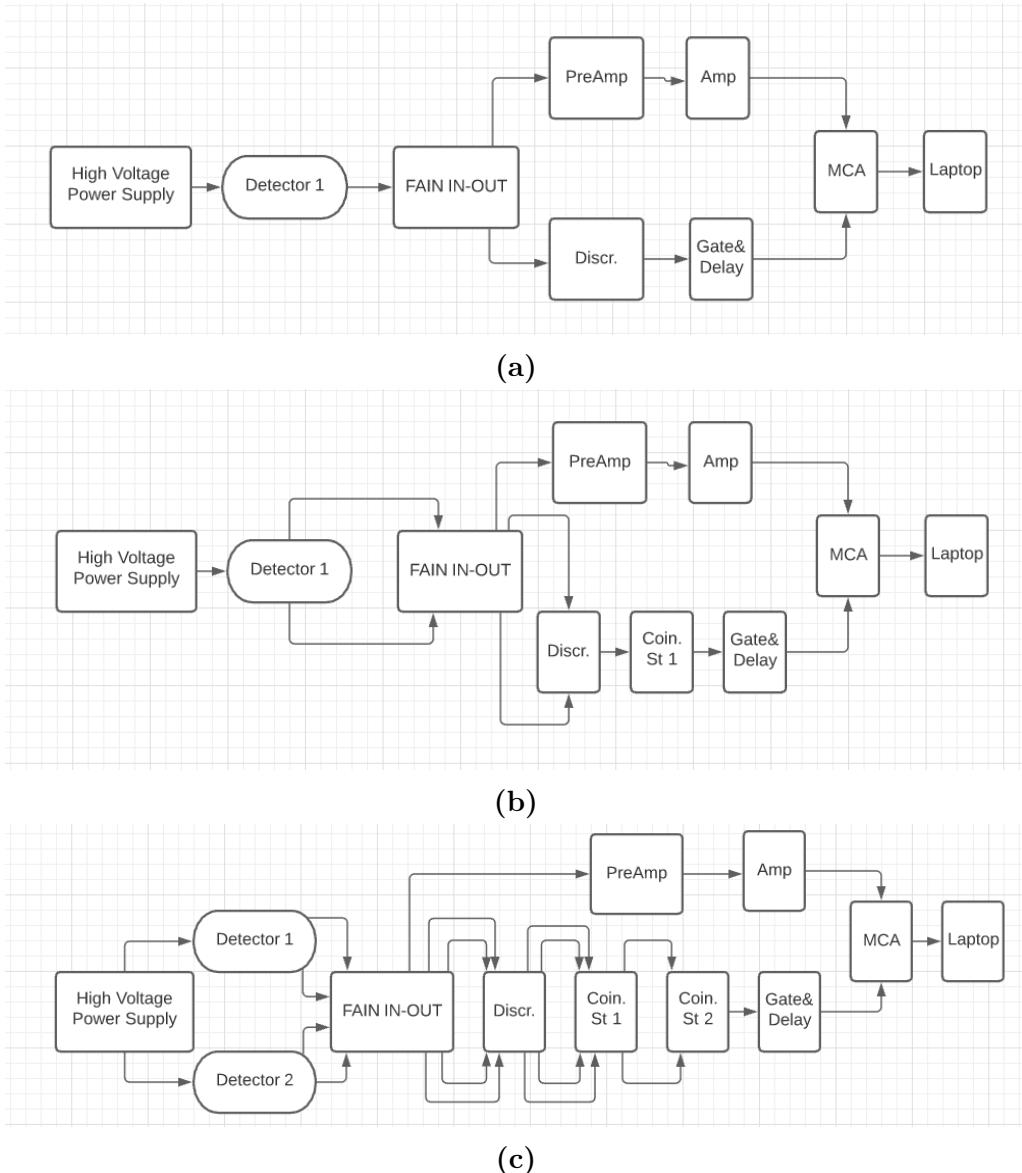
$$N^o \gamma / \text{sec} = \frac{(I_{PMT} - I_{DC})}{q_e \cdot QE \cdot CE} \quad (3.12)$$

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 since the signal is read directly from the photocathode. 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 gain of the PMT has to be restored by removing the short-circuit of the electron multiplication stage. The number of PMTs used simultaneously was one, two or four, depending on the measurement. A simplified scheme of the electronic chain employed in each case is shown in Figures 3.15a, 3.15b and 3.15c, based on various NIM modules<sup>12</sup>.

The PMTs were 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, used to create an energy spectrum, and the time coincidence line, used to make time coincidences. Therefore, an analogic FAN IN-OUT module was used to

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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.



**Figure 3.15** – Schemes of the different electronic for measuring with PMTs.  
 a) Employed when only one PMT is used. b) Employed when two PMTs are used in time coincidence. c) Employed when four PMTs are used in time coincidence.

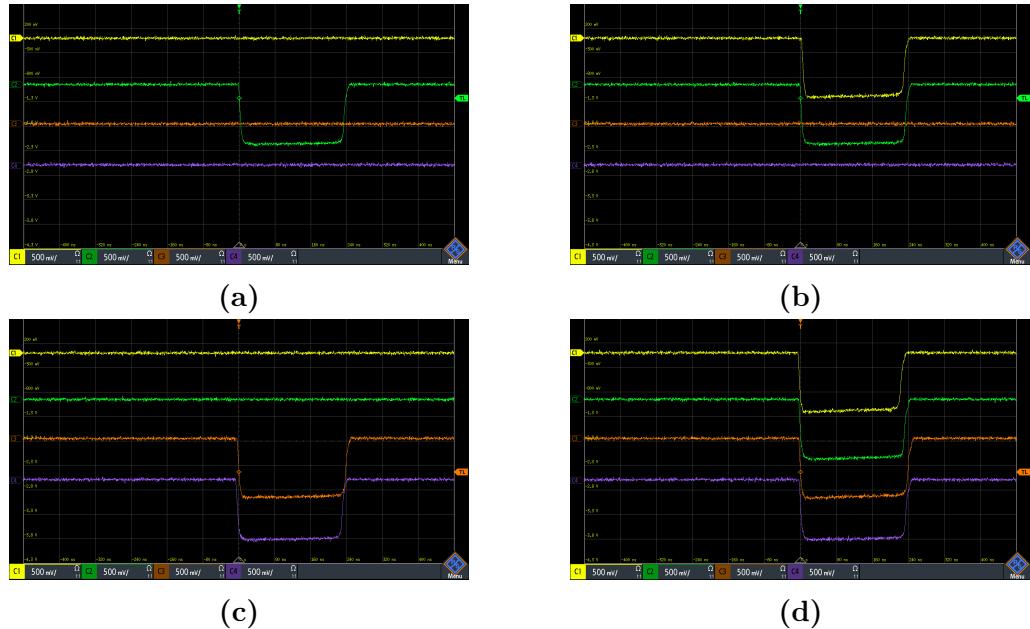
duplicate the input signal. The module employed was the Quad linear FAN IN-OUT MODEL 740 from Philips Scintific [Sci], which has four channels. One output signal was used as the input for the amplification part and the second output was used as input for the time coincidence electronics.

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 signal is integrated by a preamplifier, which gives an output signal with a height proportional to the charge of the input pulse. This signal has a long tail<sup>13</sup> produced by the preamplifier capacitance. The preamplifier used was "MODEL 9326 FAST PREAMP" from ORTEC [ORTd].
  - (b) The output signal from the preamplifier is lead to the amplifier which gives a gaussian shaped output signal. The amplifier modules were 575A and 671 from ORTEC [ORTb, ORTc]. An example of the output signal for 575A module is shown in Figure 3.17, 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 the following branches,
  - (a) The output signal 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 employed are Octuple Constant-Fraction Discriminator CF8000 module from ORTEC [ORTe] and 4 channels discriminator model 84 from CAEN [CAEa].

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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.

- (b) Time coincidences are required to ensure that detected events come from the scintillating fibers and to remove external light and dark current. The two logic signals given by the discriminator from the two PMTs that read a detector are introduced in a coincidence module which generates an output signal of  $-1.4\text{ V}$  height and of  $20\text{ ns}$  width, when both inputs are in time coincidence. The modules used were 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 3.15c) was also studied, which is useful to remove background due to hard cosmic radiation. To do so, a coincidence step similar to the previous one must be applied. The two single detector coincidence signal are checked for coincidence.
- Some examples are shown in Figure 3.16 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 reading the first detector and the other two signals (channels three and four, color orange and violet respectively) come from PMTs reading the second detector.
- i. In Figure 3.16a only one PMT (channel two) detected an event. It means that the event is likely not produced in the scintillator. In this case, no output signal is generated.
  - ii. In Figures 3.16b and 3.16c two PMT signals of one of the detectors are generated but the other detector gives no signal. This event is discarded.
  - iii. In Figure 3.16d the four signals are generated and, consequently, the output signal is generated and the event is recorded.
- (d) The logical output signal, is introduced in the Gate and Delay Generator, model 416A of the company ORTEC [ORTa], which



**Figure 3.16** – Different possibilities when time coincidences with PMTs are done. 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.

gives a positive logical signal, called time windows, shown in Figure 3.17, orange color, with a height of 8 V and width of  $2 \mu\text{s}$ . This module is used to delay the time windows until it overlaps with the energy signal as it is shown in Figure 3.17, orange signal.

As a final output of the electronics, a logical and analogical signals are obtained, shown in Figure 3.17, 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 which information is saved for later analysis. The logic signal (output from the Gate and Delay Generator module) indicates when the amplified signal must be saved.



**Figure 3.17 – Signal amplified and logical gate (input signals of MCA).**

### Electronical system for SiPMs

The SiPMs in the TRITIUM experiment are arranged in matrices of  $4 \times 4$ . The electronic system chosen to process and analyze the output signals of

the SiPM arrays is PETsys [PET], displayed in Figure 3.18, which is a commercial system prepared to work with SiPM matrices from Hamamatsu. PETsys provides time and energy digitalization, including the charge integrations QDCs<sup>14</sup> and TDCs<sup>15</sup>, resulting in a complete acquisition and digitization system capable of working with up to 1024 SiPM. This system consists of a basic board 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 shown in section 5.3, the TRITIUM monitor consists of a large number of SiPM matrices with 16 channels per matrix.



**Figure 3.18** – 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. This means that, if an improvement of TRITIUM

<sup>14</sup>charge-to-digital converter

<sup>15</sup>time-to-digital converter

limits is needed to improve its sensitivity or to further reduce the background, more photosensors would be needed. Therefore, the electronics 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>16</sup>).

PETSYS software is based on C++ and Python scripts to drive the main tasks required, such as time coincidence options between SiPM (or even SiPM matrices) or energy discrimination. This software is open source, giving the possibility to modify the current scripts or to develop others with additional functions. PETsys has a time resolution better than 30 ps which is one of the best time resolutions of commercial systems available and its price is around 10€/ channel, which is cheaper than similar electronic systems.

As reported 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 stabilization method of the SiPM gain reported in section 4.2.

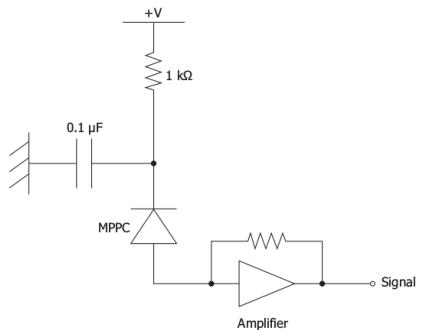
Some characterization measurements were carried out using the PETSYS system to ensure that the system works properly but the SiPM characterization was carried out at the level of a single channel (individual SiPM). The reason is that the output information of PETSYS is already integrated and digitized, so it does not allow the SiPM to be calibrated. Therefore, to characterize a SiPM, a different electronic system was used

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



(a)



(b)

**Figure 3.19 – a)** Electronic board used to provide the SiPM bias voltage and to read the SiPM output signal. b) Electronical scheme in which this PCB is based.

to read up to eight different SiPMs. This system consists of a PCB<sup>17</sup> that provides the SiPM bias voltage and reads the SiPM output signal. An example of the electronic scheme (provided by Hamamatsu) in which this PCB is based is shown in Figure 3.19.

The PCB was feed at  $\pm 6$  V using the voltage source ISOTECH, model IPS-4303 [IT] and the SiPM was feed using the electrometer KETHLEY, model 6517B [Keh], that achieves a resolution of 1 mV, low enough to ensure that this voltage variations does not affect the SiPM gain. The output signal of this PCB is connected to an oscilloscope, model WwaveRunner 625Zi from TELEDYNE LECROY [LEC17] that records the data which were subsequently analized by ROOT<sup>18</sup> scripts.

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<sup>17</sup>PCB, Printed Circuit Board

<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.

## 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. Tritium decay electrons have 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 could be formed, preventing tritium decay electrons from reaching the fibers and reducing drastically the tritium detection efficiency.
2. The tritium monitor does not have any spectrometric capabilities that could 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 tritium is the only radioactive particle that passes through it.

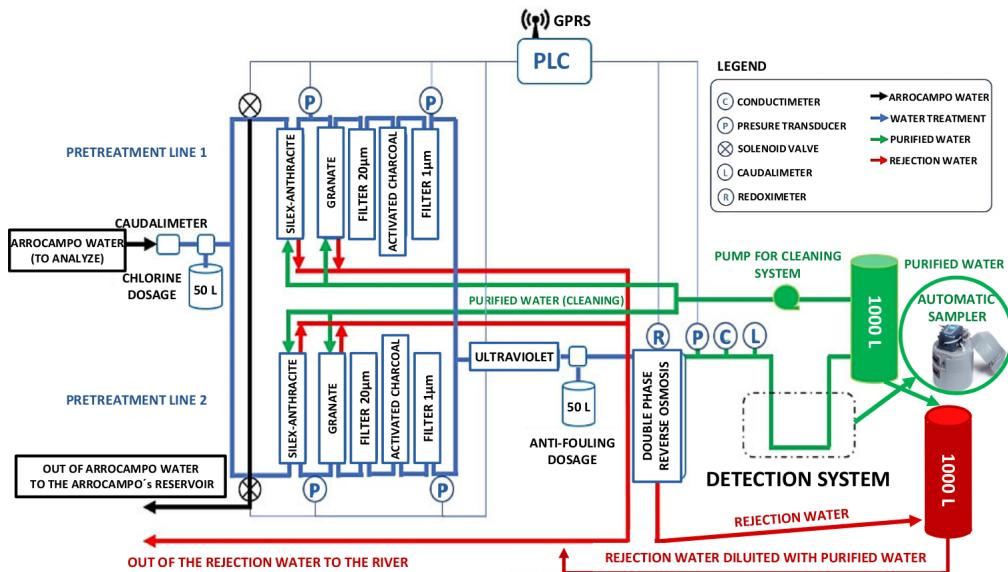
In summary, the ultrapure water system is used to keep our detector clean, ensuring the stability of its detection efficiency and to eliminate all radioactive particles other than tritium.

### 3.3.2 Design of the Ultrapure Water System

The requirements of the ultrapure water system are:

1. 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  $10 \mu\text{S}/\text{cm}$ )
2. Low maintenance (low cost and low manpower)
3. To manage probes and valves 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 3.20.



**Figure 3.20** – Scheme of water purification system.

This system is installed in the Arrocampo dam and consists of four different 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 remove 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. Therefore, only one module of reverse osmosis is used, reducing the power consumption of the system.

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

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 outperforming the requirements of the tritium detector.

The software used for remote controlling the ultrapure water system is Siemens PLC, that gives information such as the state of the valves,

the reading of pressure probes and the amount of 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 the TRITIUM Monitor

The aim 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 the 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 is given by the square root of the measured activity,  $A_m$ , which can be reduced by minimizing detected background events.

$$\sigma_T^2 = \sigma_{st}^2 + \sigma_{si}^2; \quad \sigma_{st;bak} = \sqrt{A_{m;bak}} \quad (3.13)$$

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 the 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  $\alpha$  particles, but, after interacting with the Earth's atmosphere, they generate a shower mainly composed by muons, electrons, photons and neutrons.

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].

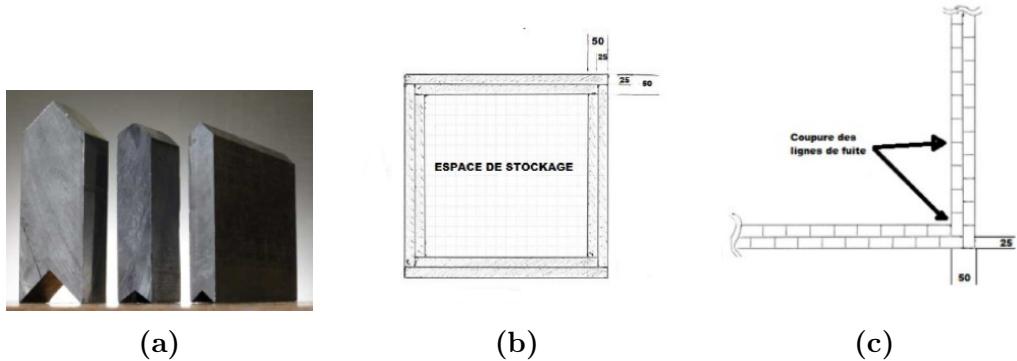
Cosmic radiation depends on several parameter like the longitude, latitude, latitude and the solar activity cycle. The spatial distribution of cosmic rays, mainly muons, follows a  $\cos^2(\theta)$  distribution with the zenith angle.

To remove the background two different techniques are employed for the weak and the hard radiation,

1. 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,
2. The hard radiation, that is any radiation of energy greater than 200 MeV/nucleon, is much more difficult to stop and the technique employed is a cosmic veto in anti-coincidence with the TRITIUM detector, reported in section 3.4.2.

### 3.4.1 Passive Shield (Lead)

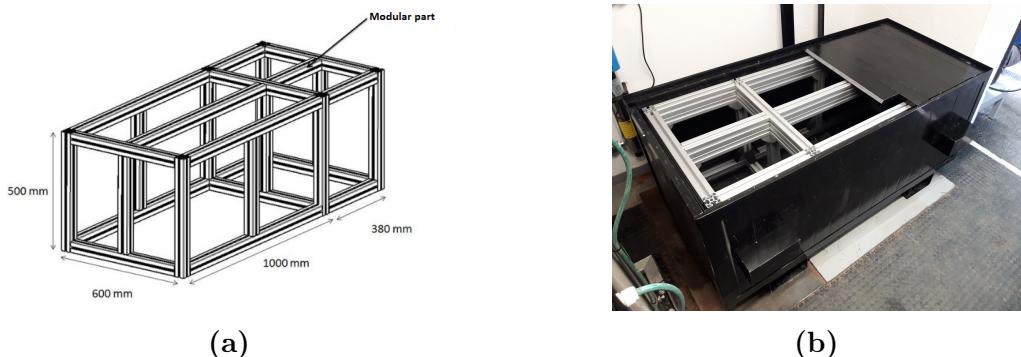
Weak radiation is suppressed by a lead shielding inside which the TRITIUM detector is placed. This lead shielding is effective for particle energies below 200 MeV/nucleon, originating from the Earth's natural radioactivity and the weak component of cosmic radiation. This lead shielding consists of 158 lead bricks of ultra-low intrinsic radioactivity, 25 mm thick. They are chevron shaped, as shown in Figure 3.21a, specially designed for a perfect fit and easy assembly. As can be seen in Figures 3.21b and 3.21c, 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.



**Figure 3.21** – a) Lead Bricks b) and c) Two layers for the lead bricks of the shielding.

A special aluminum structure, shown in Figure 3.22, was designed by mechanical engineering department of CENBG 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 3.21. The larger one has internal dimensions of  $90.5 \times 41 \times 51$  cm $^3$  and is used to place the TRITIUM detector. The smaller one, of dimensions of  $33 \times 41 \times 51$  cm $^3$ , contains the DAQ system. The



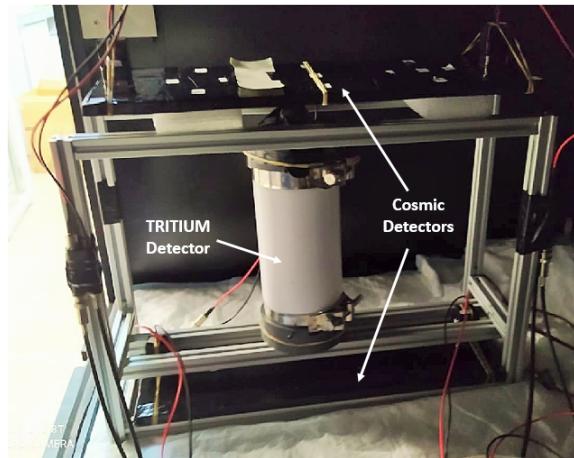
**Figure 3.22** – Aluminium structure of the passive shielding

external dimensions of the lead shielding are  $148 \times 60 \times 70$  cm<sup>3</sup> 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, which consist of at least two complementary detectors in coincidence that reject events simultaneously detected in both of them. As shown in Figure 3.23, the two complementary detectors are 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 inside.

A hard cosmic event crossing simultaneously both cosmic detectors is schematically sketched in figure 3.24a. Each cosmic detector has two photosensors, so the electronic configuration given in Figure 3.15c is used to make time coincidences. The TRITIUM detector is read out in anti-coincidence with the cosmic veto to reject the hard cosmic events from the tritium measurement. Random coincidences from two different hard cosmic events, one in each detector, as drawn in Figure 3.24b, are negligible. The expected hard cosmic rate at sea level for muons is  $7 \times 10^{-3}$  cm<sup>-2</sup>s<sup>-1</sup>sr<sup>-1</sup>

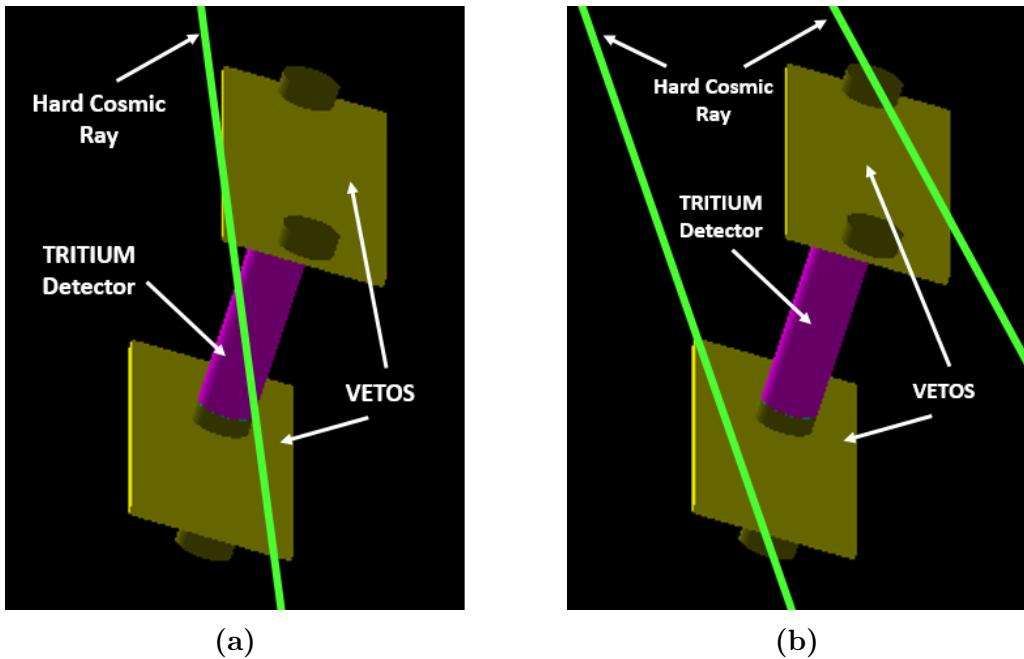


**Figure 3.23** – Cosmic veto and Tritium-IFIC 2 prototype in an aluminum mechanical structure developed by IFIC’s mechanical engineering department.

[ea20, SAG01], as shown in the plot of Figure 3.25. 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 negligible.

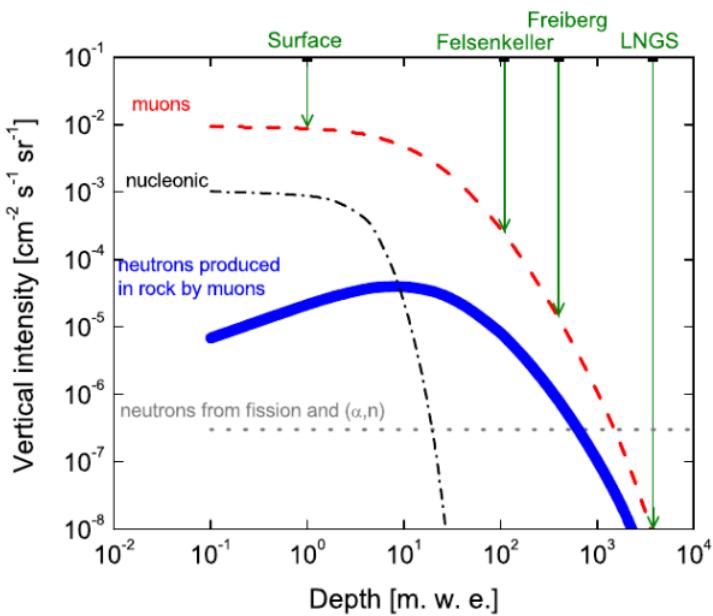
The vetos are made of a plastic scintillator block from Epic-Crystal [Cry20]. Its properties are given in Table 3.5 and its energy emission spectrum is displayed in Figure 3.26.

The energy spectrum has a peak very close to that of the scintillating fibers used, so the same photosensors are used to read them out. The dimensions of the scintillator block are  $45 \times 171 \text{ cm}^2$ . They are wrapped by three different layers, Teflon, aluminum and black tape, exhibited in Figure 3.27. 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.



**Figure 3.24** – Hard cosmic events detected with the cosmic veto of TRIUM: a) Real coincidence, b) Fake coincidence.

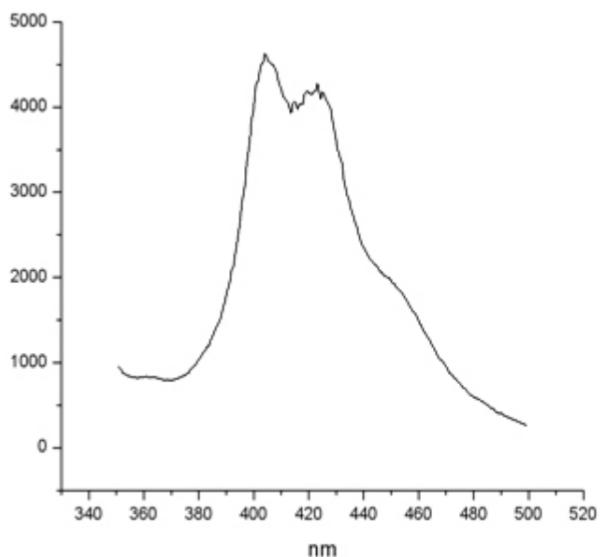
As mentioned above, the expected hard cosmic rate at sea level is  $7 \times 10^{-3} \text{ cm}^{-2}\text{s}^{-1}\text{sr}^{-1}$ . Taking into account that the solid angle of our detectors is  $\omega = 0.5434$ , calculated by integrating the solid angle of one scintillator on the other, and that the area of the veto is  $765 \text{ cm}^2$ , the expected hard cosmic rate on our cosmic vetos should be 2,909 event/s. This is an important result which is used in section 4.4 to determine the efficiency of the cosmic veto.



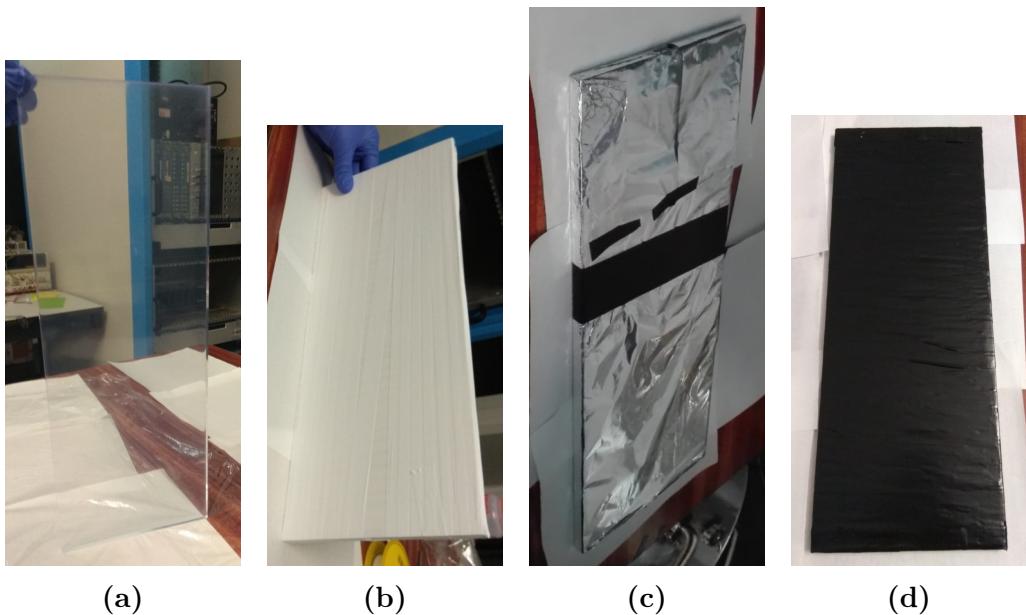
**Figure 3.25** – Hard cosmic muon rate [Szu15].

Property	Value
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].



**Figure 3.26** – Emission energy spectrum of the plastic scintillation used for the cosmic vetos [Cry20].



**Figure 3.27** – Different layers used to wrap the cosmic veto detectors. a) Scintillator without coating. b) Teflon coating. c) Aluminium coating. d) Black tape coating.

# 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, the ultrapure water system and the background rejection system. This characterization is crucial to understand the behaviour of the different parts and the results of the monitor. Furthermore, several developments were made to improve fundamental parameters of the TRITIUM monitor components to enhance their tritium sensitivity. All these studies were carried out inside a special light-tight box, called black box, to ensure that the detected photons 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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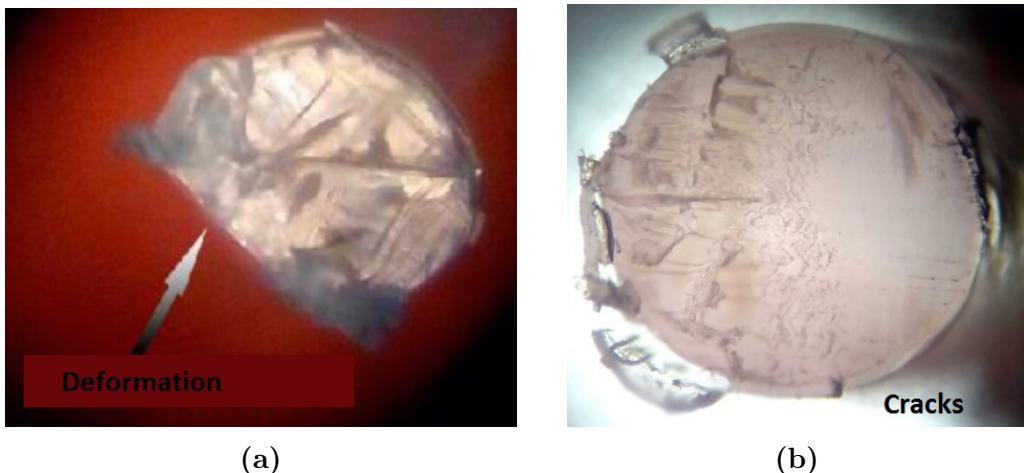
<sup>1</sup>ADC units are the internal units, called channels, in which an analog signal is digitized after an Analog-to-Digital Converter. The ADC units are proportional to the energy and the number of available channels depends on the bits used in its digitization.

## 4.1 R&D for the Plastic Scintillating Fibers

This section reports experimental measurements of the scintillating fiber parameters relevant for tritium detection, such as collection efficiency and uncertainty caused by the conditioning protocol. Thousands of scintillating fibers are used in the TRITIUM detector which were prepared and conditioned prior to characterization studies or tritium detection. Therefore, various mechanical and electronic devices were developed to automatically prepare large number of fibers simultaneously.

### 4.1.1 Scintillating Fiber Conditioning Protocol.

The first step in TRITIUM design was to choose the fiber length at which the signal of tritium events is optimized. On the one hand, long fibers are suitable 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 boundaries many times before reaching the photosensors, that produce a deterioration in the tritium signal. To determine the optimal fiber length, several simulations, described in section 6.2.3, were carried out using Geant4 [Col21a], a particle and nuclear physics simulation package based on C++. It was concluded that it is preferable to employ relatively short fibers. The fiber length for the TRITIUM prototypes developed at IFIC, is 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 cleaving technique had to be developed with strict requirements on the cleaving quality of the fiber ends since this greatly affects the transmission of photons and, consequently, the efficiency of TRITIUM monitor. This cleave must be done perpendicular to the fiber axis and with small uncertainty in the cleaving position to achieve a good end-surface quality that enables optimal coupling to the photosensor. It is also important that the fiber



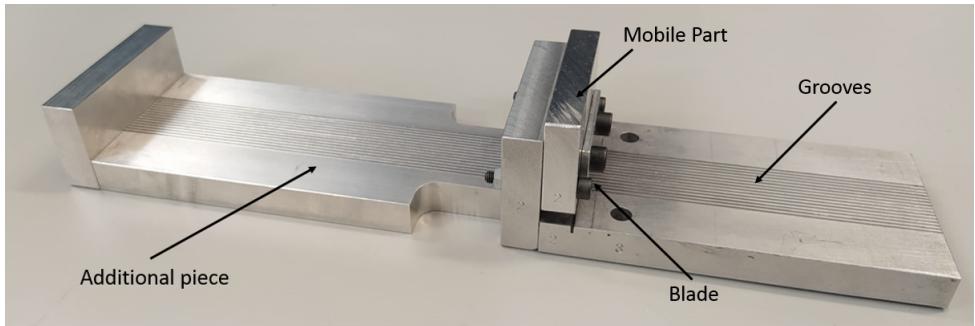
**Figure 4.1** – Unsuccessful results of using commercial techniques to cleave fibers a) Fiber end deformation b) Fiber end cracks.

integrity be preserved, without cracks or deformations that contribute to the loss of photons. Cleaving the end faces of plastic polymer fibers is a current challenge. There are many different techniques such as milling, laser cleaving, focused-ion-beam, blade cleaving, etc. The blade cleaving technique was chosen for TRITIUM experiment because of its mechanical simplicity and because this preserve the integrity of plastic fibers.

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 quality standard requirements.

Because commercial devices are not suitable for our polymer fibers for our scintillating fibers, a cleaving device, shown in Figure 4.2, was designed, built and tested at IFIC laboratory.

This device consists of fourteen grooves to which fibers are held

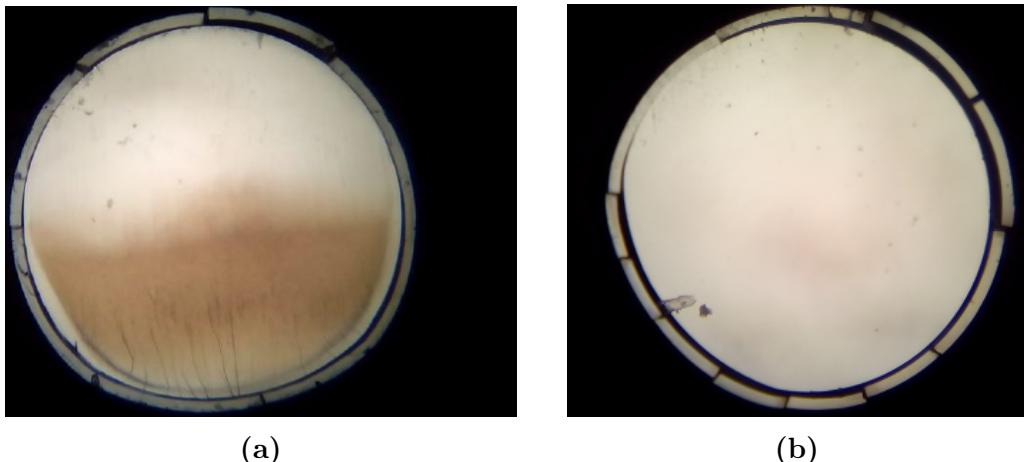


**Figure 4.2** – Cleaving device developed in TRITIUM experiment.

and a thin blade, attached to a mobile piece, which is used to cleave them. The perpendicular cleave, which is one of the requirements, can be ensured since the moving piece, to which the blade is attached, 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 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 cleave [SR15, Law06]. As it can be seen in Figure 4.3a, with this cleaving device fiber ends without breaks or deformation were obtained. An additional parameter that could in principle affect the cleaving quality of the fiber ends is the temperature of both, fiber and blade. A study was carried out in which both were subject to different temperatures from room temperature to 110 degrees. No significant conclusions were obtained [Cam17]. Thus, the cleaving process was carried out at room temperature to make the cleaving process easier.

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

The microscope model PB 4161 from EUROMEX and the Digital

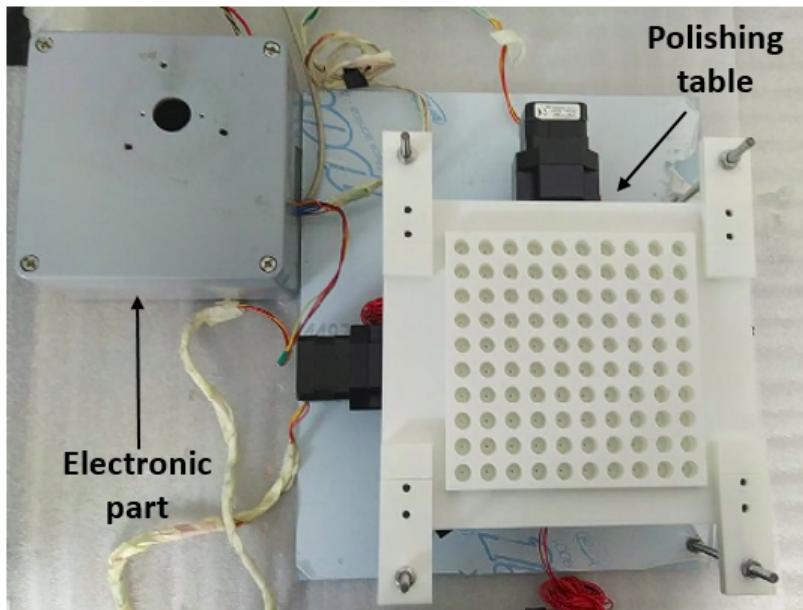


**Figure 4.3** – Result of the polishing process. a) Fiber end after cleaving  
b) Fiber end after cleaving and polishing with Thorlabs technique.

Microscope from Jiusion were used for the quality control of the fiber ends after cleaving. An example of the fiber end after cleaving is shown in Figure 4.3a. The slightly darkened zone at the bottom of the fiber is an unavoidable effect of the cleaving process and, to reduce this effect, a polishing process developed by Thorlabs was applied [Inc06]. It consists on rubbing the fibers during two minutes with 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  $0.3\ \mu\text{m}$ , describing on the paper a shape of an 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, cleaving and polishing, are necessary.

#### 4.1.2 Automatic Polishing Machine for Scintillating Fibers

As mentioned above, tens of thousands of fibers had to be prepared and conditioned for the TRITIUM monitor<sup>2</sup>, section 5.3. Although this number of fibers was not a problem for cleaving, which is very fast process, 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 quantity 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 number of fibers.



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

This automatic polishing machine, displayed in Figure 4.4, consists of two parts: 1) A polishing table, where the fibers are polished 2) The

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

electronics, based on Arduino technology, that operates the movement of the polishing table:

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 locked to the system by four vertical screws. There are two nuts on each screw used 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.

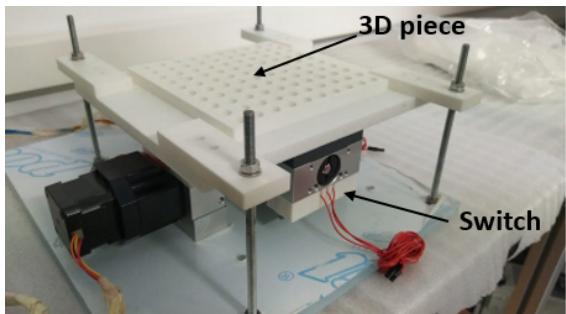
As the fibers are too light (0.16 g) to make the necessary pressure on the polishing paper, a plastic belt and a piece of metal with a weight of around 1.5 g, shown in Figure 4.5b, were employed to increase their contact pressure (similar to the connectors used in the Thorlabs polishing process).

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.

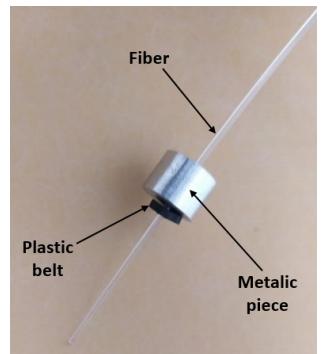
The polishing system contains several switches, mounted on a piece made with the help of a 3D printer, shown in Figures 4.5a, 4.5c and 4.5d, which are used to find the origin of coordinates when the system is reinitiated and to stop the movable part when the end of the path is reached.

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

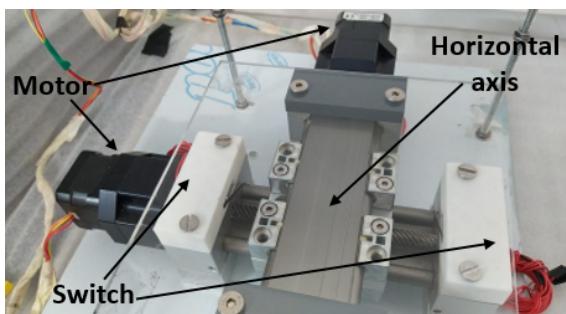
This electronics consists of two stepper motors, model NEMA ST4209S1404-A [Nan], which move the horizontal screws on which the polishing



(a)



(b)



(c)



(d)

**Figure 4.5 – Polishing machine.** a) Polishing table. b) Fiber with metal piece. c) Horizontal screws and PMMA plate. d) Piece to hold switches.

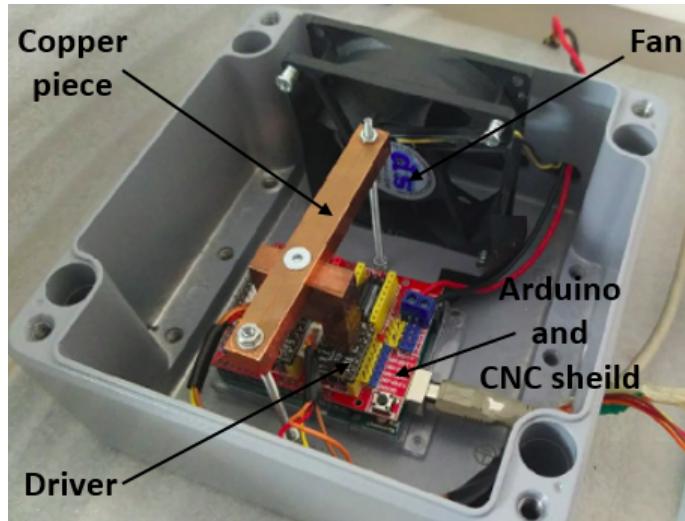
paper is attached. These motors are controlled by an Arduino UNO [ARD] that uses a CNC shield [OSO17] in which two different drivers are connected to control the stepper motors, one driver for each stepper motor.

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 owing to its a microstepping mode of 1/256. The voltage and current used to power the motors are 35 V and 2 A which are 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 wrong movements 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 computer board using the Universal G-code Sender software (a graphical interface based

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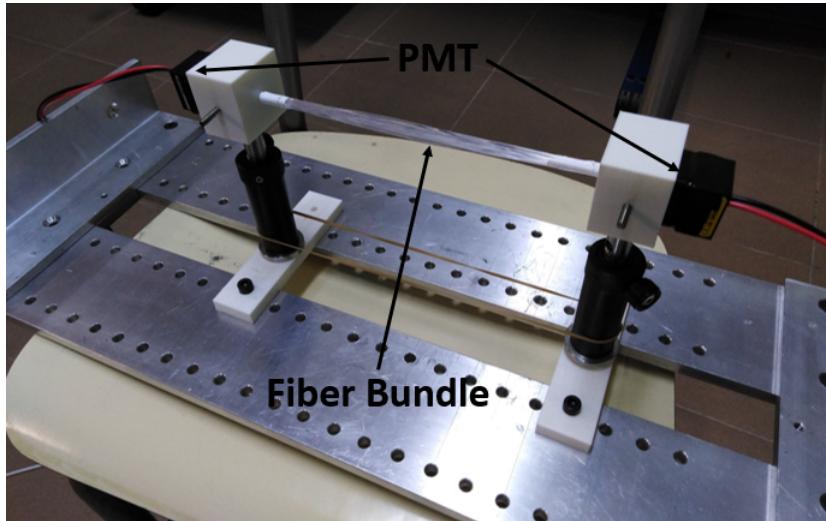
<sup>3</sup>The copper is one of the best thermal conductor at STP



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

on the GRBL package). There are several useful pre-programmed functions such as "HOME" with which the system, using the switches, finds its origin of 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 project, 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 3.15b, monitored the light transmission of the fibers.

The light transmission was measured before and after polishing. These measurements were carried out 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. The energy spectra recorded for both radioactive sources are exhibited in Figure 4.8. The sources were placed in the middle of the fiber bundle, at 7.5 cm from each PMT.

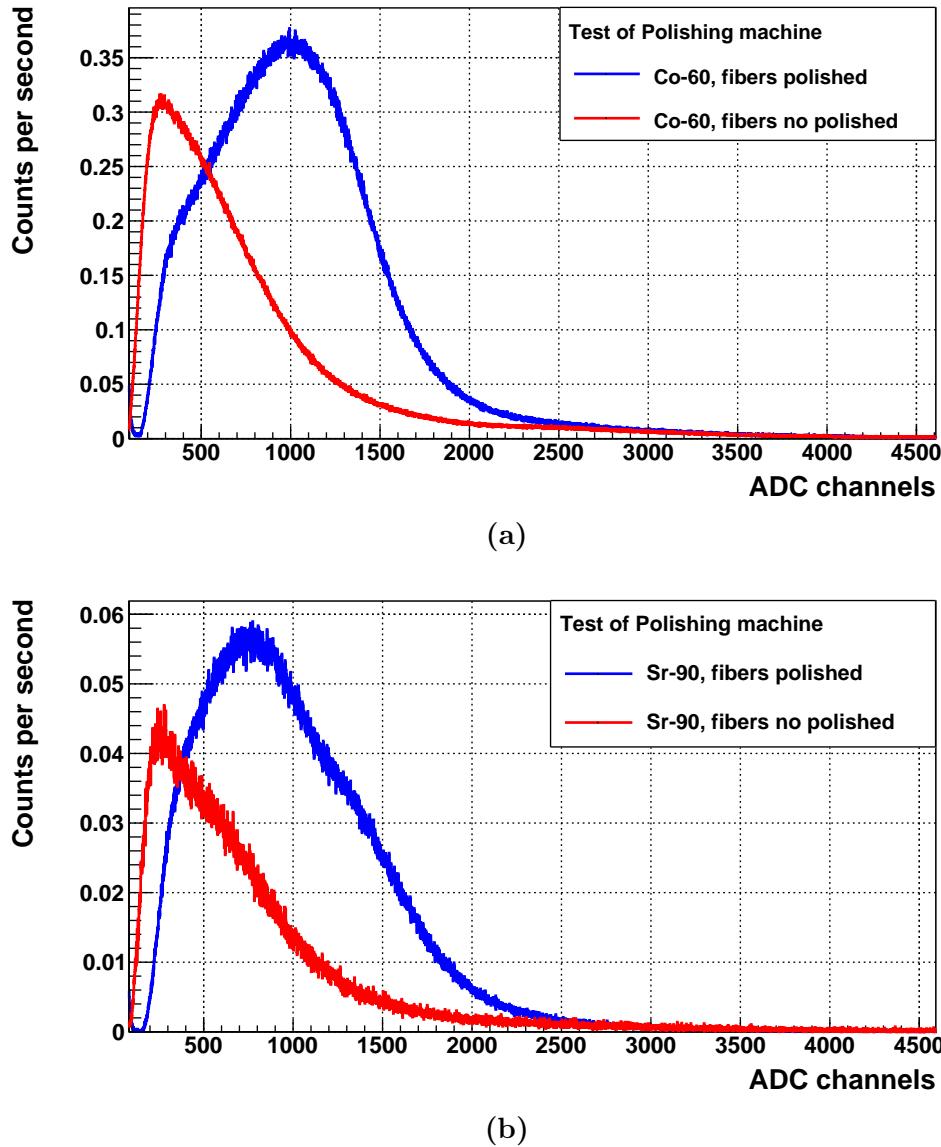


**Figure 4.7** – Setup used to test the effect of the polishing machine.

As it can be seen in Figure 4.8, both energy spectra are shifted to the right after polishing, which means that the detected events have 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 fibers. 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 uncladded BCF-12 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.

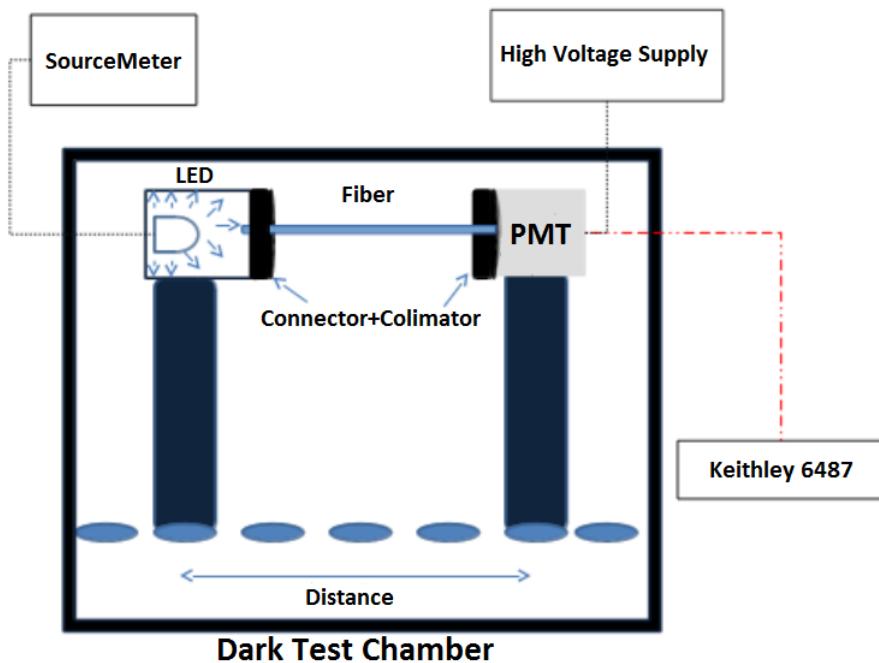


**Figure 4.8 –** Energy spectra recorded with polished and unpolished fibers.  
a) for the  $^{60}\text{Co}$  source b) for the  $^{90}\text{Sr}$  source

Although commercial clads are too thick for the TRITIUM experiment, a low thickness clad could be developed. For example, clads with a thickness of the order of tens of nanometers could be achieved by electrodeposition techniques. 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 for 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 magnitude employed for the characterization was the rate of photons reaching the active area of the photosensor. To measure this magnitude, a calibrated R8520-06SEL PMT of 29.76% quantum efficiency at the working wavelength measured by Hamamatsu was employed. The PCB described in section 3.2.4 was used to switch the PMT internal gain off and the PMT output current was measured by a Keithley 6487 picoammeter/voltage source. The photon rate was obtained from the current measurement using the equation 3.12 with  $QE = 0.2976$  and  $CE = 1$ . A simplified scheme of the setup is shown in Figure 4.9.

This setup consists of an optical structure in which a LED and a PMT are separated a user set distance from each other. A LED435-03 from Roithner LaserTechnik GmbH [Gmb10], simulated the fiber scintillating light. 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 placed 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



**Figure 4.9** – Setup used for fiber characterization.

FH-ST<sup>4</sup> connectors from RoHS company [], were used to fasten the fiber to the system.

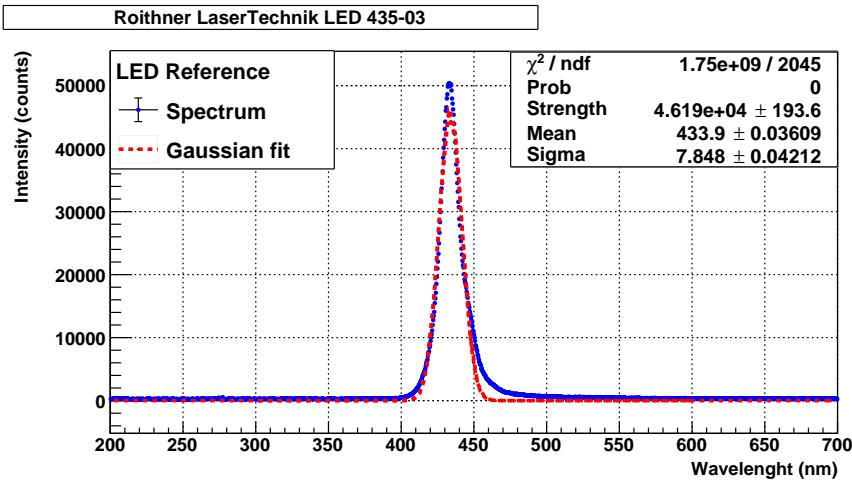
### Preparation of the Characterization System.

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

A light leak in the black box would produce a background larger than the signal. To check the light-tightness of the black box a uncladded fiber of 20 cm length was arranged in the setup. 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

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

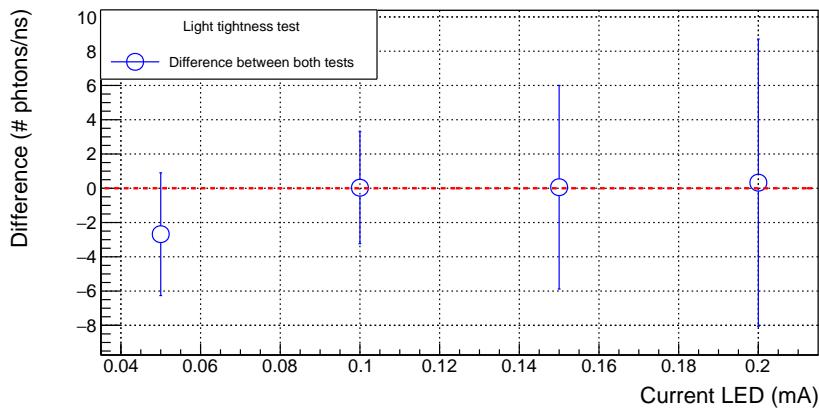


**Figure 4.10** – Emission spectrum measured for the LED model 435-03 from Roithner LaserTechnik GmbH Company.

from Thorlabs [Thoa], that prevents external photons to reach the system. This test was repeated for three different fibers and the mean and standard deviation of the light output were calculated.

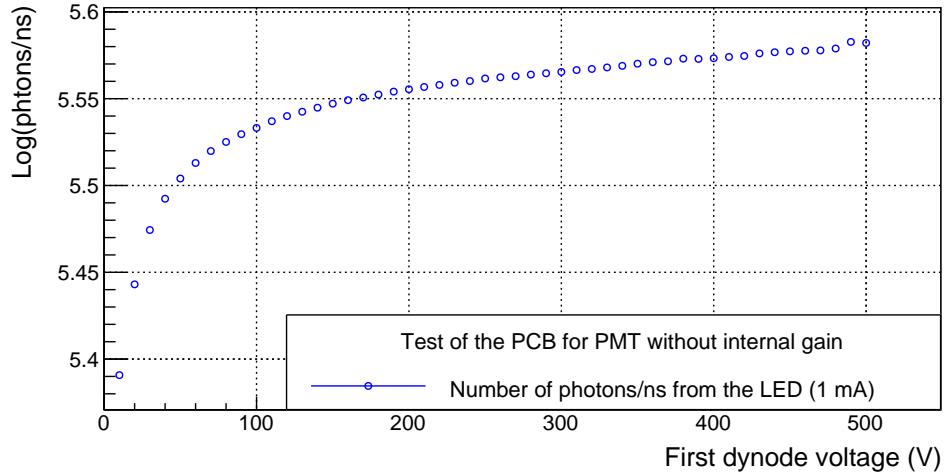
The difference of the PMT responses in both cases is plotted in Figure 4.11 as a function of the LED intensity. As it can be seen in this figure, there are no statistically relevant differences between covered and uncovered fibers. Therefore, the light tightness of the black box is sufficient for this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

The optimal voltage of the PCB without the PMT internal gain was obtained by finding the voltage plateau at which the electron collection efficiency in the first dynode was practically 100%. With no fibers in the setup, the LED was fed at 1 mA intensity 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 starts at voltages higher than 150 V. The chosen voltage for the characterization was 250 V.



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

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 does not saturate. The linearity was tested in the of the number of photons range expected for a tritium event (a few tens of photons per tritium event, which gives tens of photons per nanosecond) and in the range around two thousand five hundred photons per nanosecond. To test the linearity of the PMT in the range of tritium events, the setup described above was used without any fiber but with one of the connectors and the collimators kept to make sure that the active area of the PMT is the same as in the characterization study. To test the linearity of the PMT in the range of more than a thousand photons per nanosecond, the remaining connector was removed in order to increase the photons that reach the photosensor but the collimator was also kept. 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.



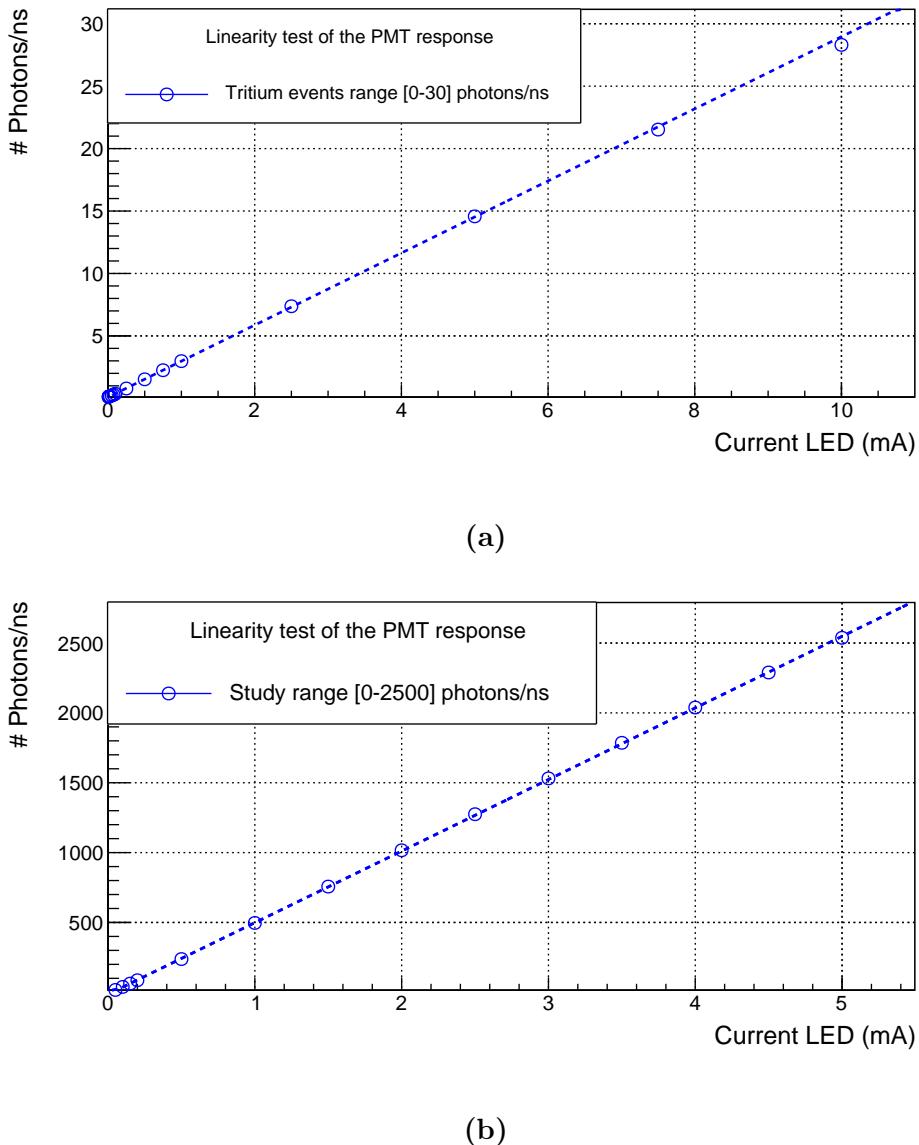
**Figure 4.12** – Response of the PMT as a function of its high voltage using the designed PCB with which no internal gain of the PMT is obtained. Error bars are included but they are too small to be visible.

## Results of the Characterization of Scintillating Fibers

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

$$\sigma_t = \sqrt{\sigma_{pos}^2 + \sigma_{con}^2} \quad (4.1)$$

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 giving only the uncertainty in the fiber



**Figure 4.13 – Linearity tests of the PMT response.** (Above) Response of the PMT in the intensity range of tritium events. (Below) Response of the PMT in the range 0 – 2500 photons/ns. Error bars are included but they are too small to be visible.

position ( $\sigma_t = \sigma_{pos}$ ), and the second to obtain the total uncertainty. The conditioning uncertainty is given by,

$$\sigma_{con} = \sqrt{\sigma_{tot}^2 - \sigma_{pos}^2} \quad (4.2)$$

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 setup, and measurements by feeding the LED with an intensity of 0.1 mA are made. These measurements were repeated ten times with a given fiber. The mean,  $\bar{x}$ , and the standard deviation of the measurements for each fiber type are shown in Table 4.1 where the relative standard deviation,  $\sigma_{pos}^{rel}$ , defined by equation 4.3, is also included.

$$\sigma_{pos}^{rel} = \frac{\sigma_{pos}}{\bar{x}} \quad (4.3)$$

Fiber type	Mean (photons/ns)	$\sigma_{pos}$ (photons/ns)	$\sigma_{pos}^{rel}$ (%)
Uncladded	$524.09 \pm 0.01$	17.65	3.37
Single Clad	$1071.70 \pm 0.01$	9.07	0.85
Multiclad	$949.93 \pm 0.03$	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 noticed, the clad reduces the position uncertainty, which means that it improves the uniformity of the fiber response. It was also found that the clad significantly improves the light collection efficiency of the fibers. The reason could be that photons are mainly collected in the core of the fiber and the interface of core is better defined for single-clad and multi-clad fibers than for uncladded fibers. In the latter case, the interface is provided by the environment (air or water in the case of TRITIUM). 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 radius of 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 of the measurement and was neglected.

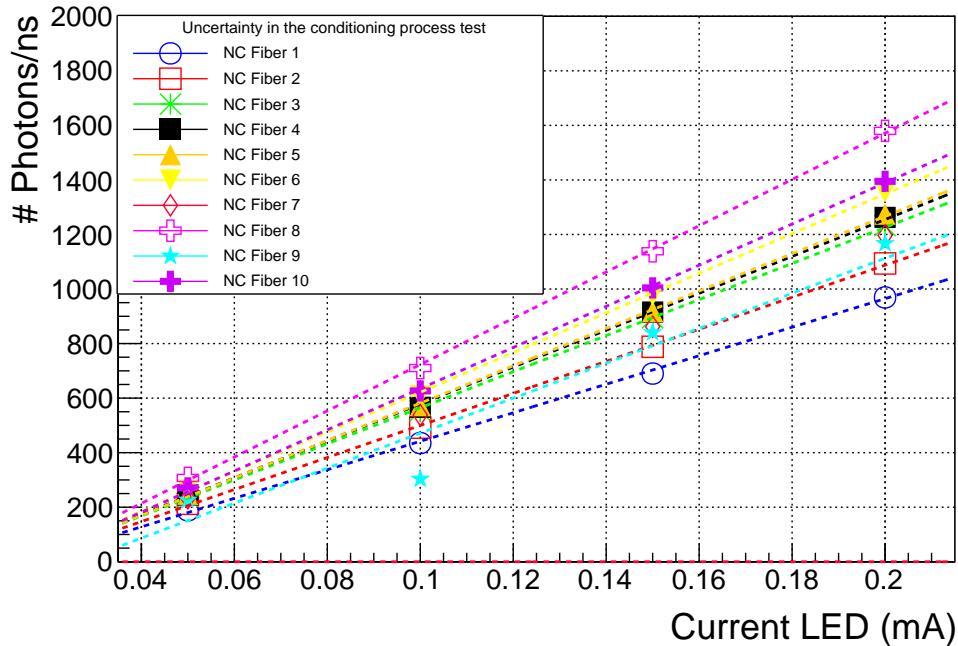
To determinate the whole uncertainty, ten different samples of each fiber type were prepared and each fiber was measured under the same conditions as above. This measurement was done for four different LED emission intensities (0.05, 0.1, 0.15 and 0.2 mA). The results for uncladded fibers are plotted in Figure 4.14, where it can be seen that, although each fiber shows a very linear trend with the number 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 are given in Tables 4.2 and 4.3 respectively and are plotted in Figure 4.16, where they can be compared.

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 collected photons versus LED intensity for the different type of fibers.

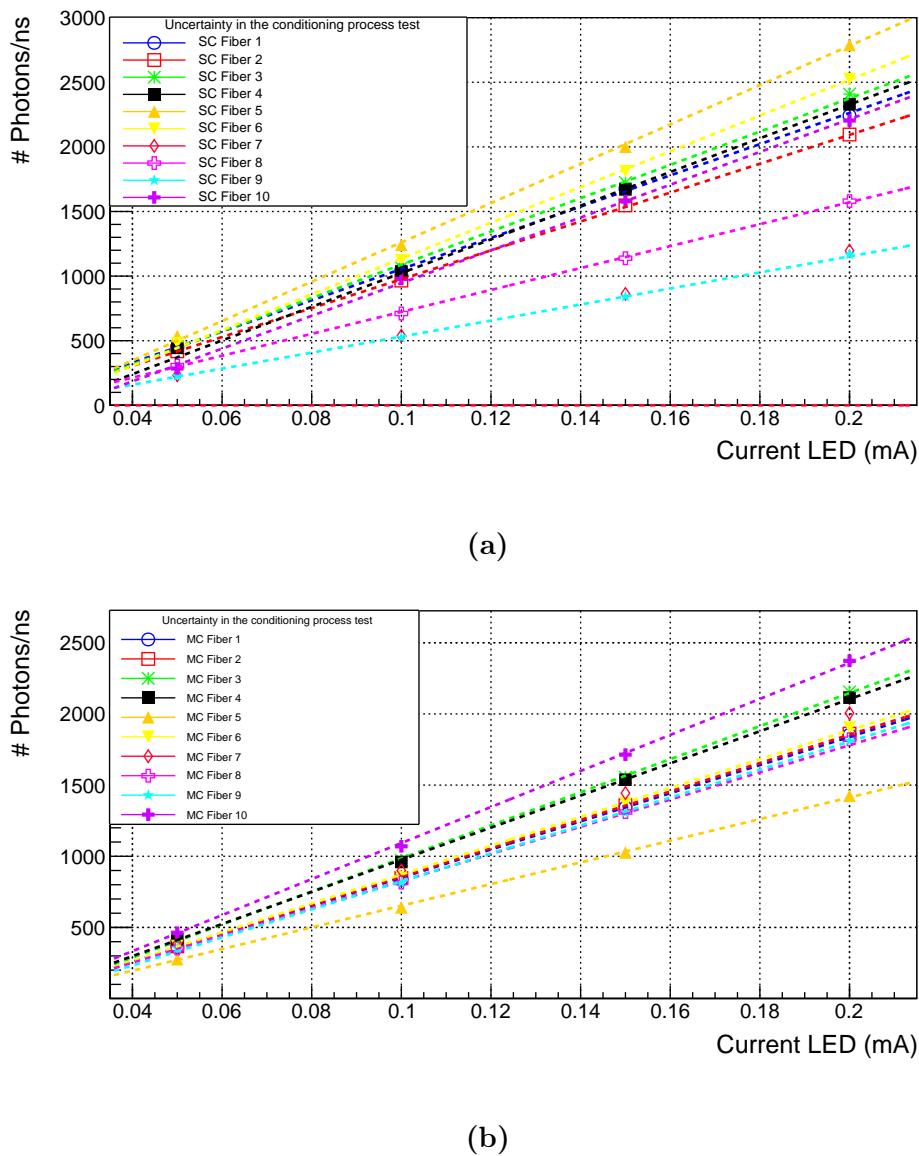
As it can be noticed in Figures 4.14 and 4.15 the fiber response is quite linear and single clad and multiclad fibers have stronger signals than uncladded fibers, which means that the clad has a significant effect on the fiber collection efficiency. It can also be remarked in Table 4.3 that the relative standard deviation,  $\sigma_{pos}^{rel}$ , does not vary with the LED intensity.



**Figure 4.14** – Number of photons/ns reaching the PMT for Uncladded fibers. Error bars are included but they are too small to be visible.

An average of the relative standard deviation caused by the fiber positioning and conditioning is given in Table 4.4. As it can be noticed, the smallest relative standard deviation due to the conditioning process was found for uncladded fibers, which means that the damage from this process occurs mainly in the fiber clad, as illustrated in Figure 4.3. It was checked under 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 increases the resistance of the fiber to the conditioning process.

In summary, this study shows that the use of a fiber clad improves the photon collection efficiency. The relative statistical deviation due to the



**Figure 4.15** – Number of photons/ns reaching the PMT for ten samples of each fiber type. (Above) Single clad fibers (Below) Multi-clad fibers. Error bars are included but they are too small to be visible.

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,  $\sigma_t(\%)$ , versus LED intensity for the different fiber types.

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

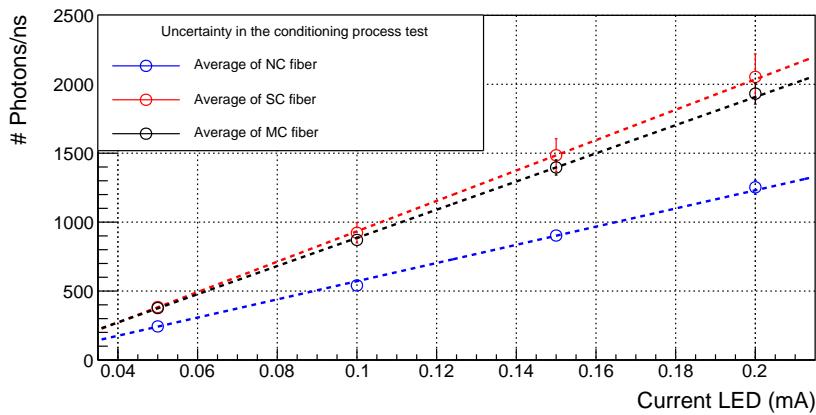
Table 4.4: Relative standard deviations ( $\sigma_t$ ,  $\sigma_{pos}$  and  $\sigma_{con}$ ) measured in this test.

fiber conditioning process was quantified for the different fiber types. It was found that the damage by 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 for each type of fiber is shown. To measure the collection efficiency,  $CE_{100}$ , ten different samples 10 cm length were prepared for each fiber type. Similar measurements, summarized in Table 4.5, were carried out.

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

The collection efficiency,  $CE_{100}$ , given by the manufacturer Saint-



**Figure 4.16** – Average number of photons versus LED current for 10 samples of each fiber type (uncladded, single clad and multi-clad fibers). Error bars are included but they are too small to be visible.

Gobain is in the range 3.44% – 7% [Cer05]. Our measurements, given in Table 4.6, are close but slightly higher than the manufacturer values which could be attributed to our use of collimated photons.

Led Int. (mA)	Uncladded ( $\gamma/\text{ns}$ )	Single-clad ( $\gamma/\text{ns}$ )	Multi-clad ( $\gamma/\text{ns}$ )
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 intensity 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  $CE_{100}$ .

#### 4.1.4 Cleaning Protocol of Scintillating Fibers

The tritium events only produce a few photons in the fibers, so it is very important to detect as many photons as possible. As it was demonstrated 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) affects conspicuously 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.

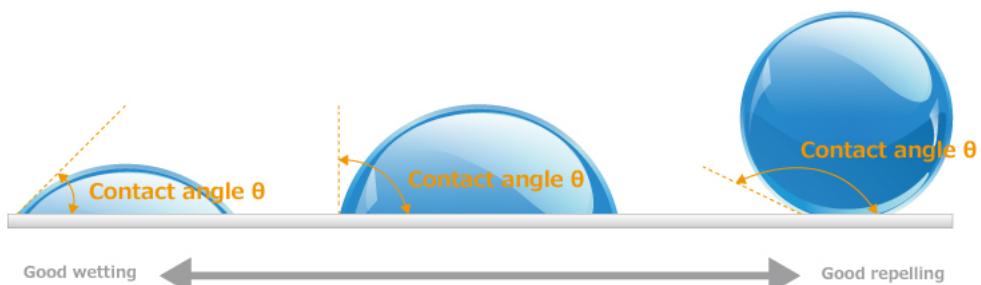


Figure 4.17 – Wetting property produced by the cleaning process. [comb]

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 and they 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 spectra were measured again. Two radioactive sources were used in this study, a <sup>90</sup>Sr beta source, already used in the polishing machine test, and a <sup>137</sup>Cs gamma source, of 500 Bq activity. The results are plotted in Figure 4.18. A shift of the spectrum to higher energies can be noticed for the cleaning fibers. This improvement was quantified by a parameter  $F$  definded as,

$$F = \frac{A_C - A_{NC}}{A_C} \quad (4.4)$$

where  $A_C$  and  $A_{NC}$  ae the integrals of the energy spectra measured after and before the cleaning process, respectively.

The value of  $F$  obtained is about 21% for both radioactive sources.

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<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  $\mu\text{Sv}/\text{cm}^2$

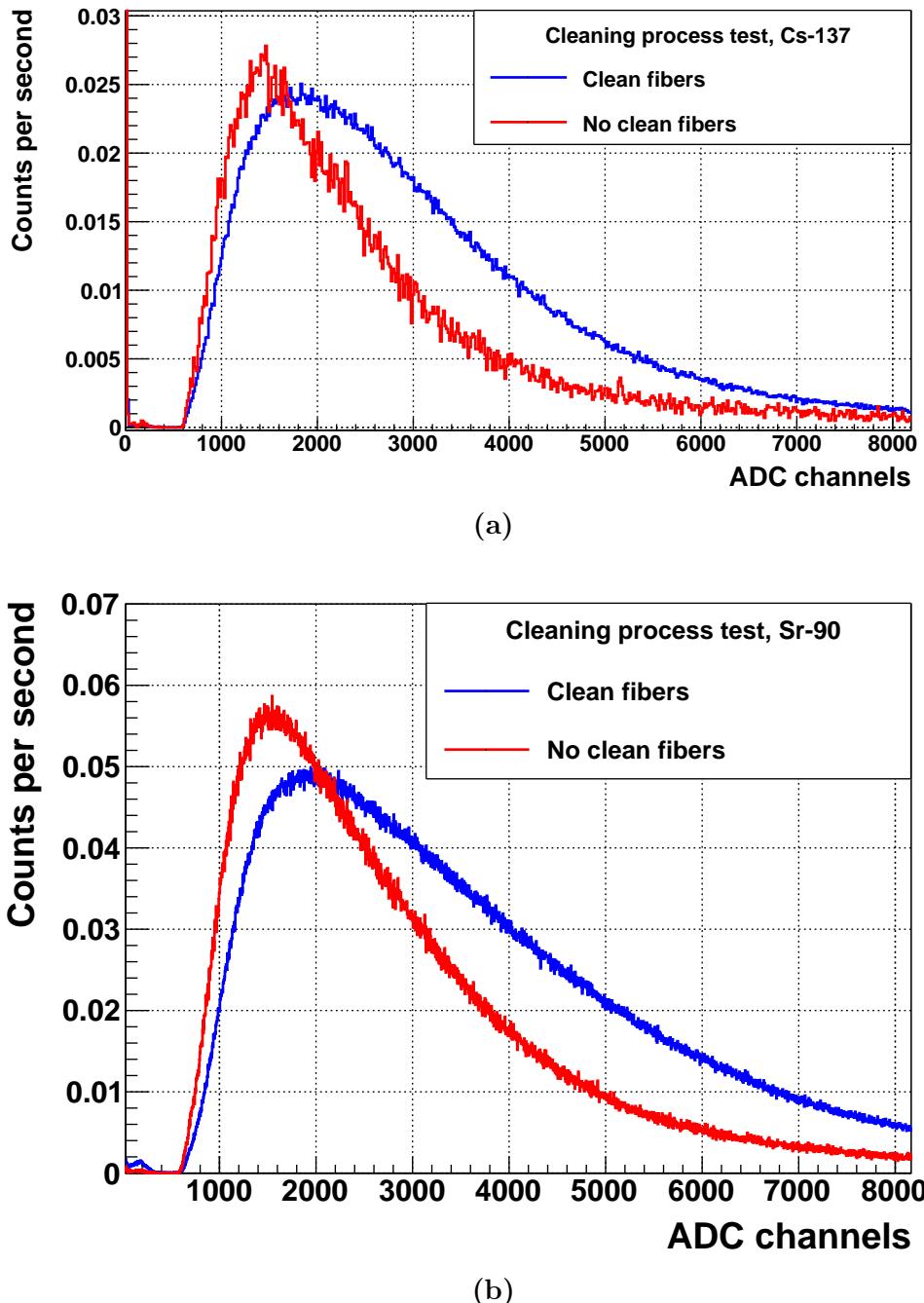
Nevertheless, it should be taken into account that  $F$  was measured in air and the result could differ in water.

## 4.2 Characterization of SiPM

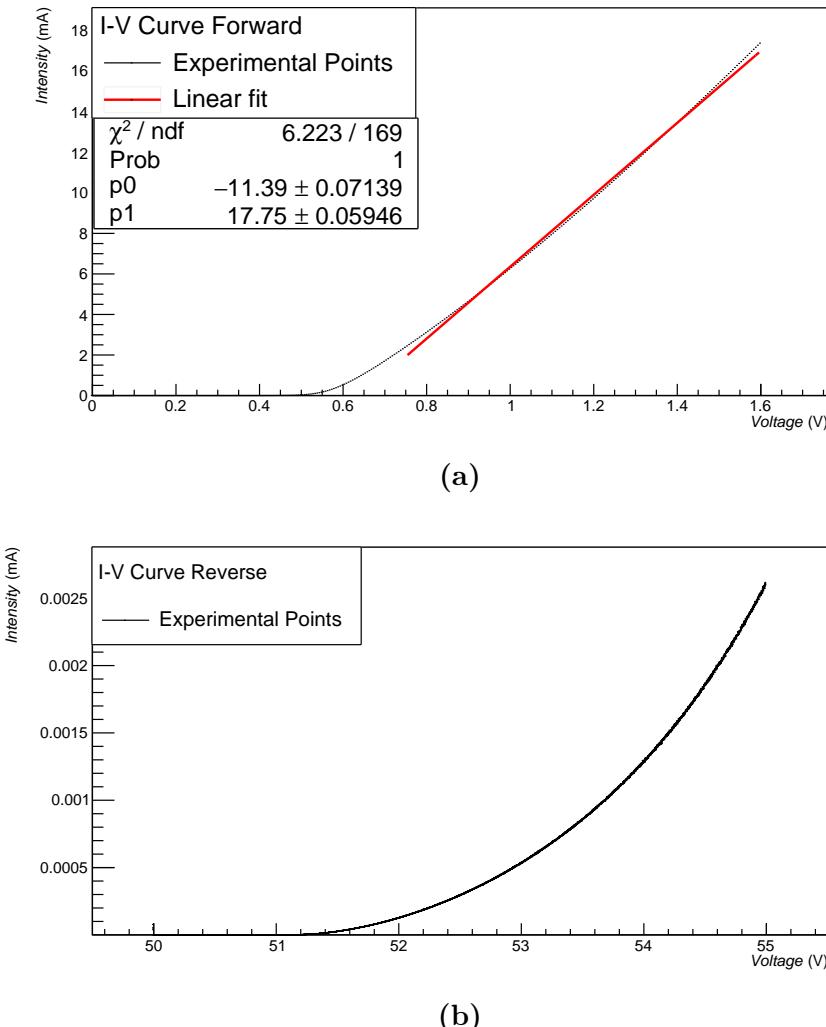
This section details the characterization of the SiPM S13360-1375 model, which was the first chosen as TRITIUM monitor photosensor. This characterization is incomplete since some important SiPM parameters for the TRITIUM monitor, which are PDE, dark count rate and crosstalk probability, were not measured. A complete characterization is already underway for the S13360-6075 model, the latest proposal for the TRITIUM detector, where the relevant parameters, given in section 3.2.3, will be experimentaly determined using the experimental setup, described in appendix A. The SiPM characterization is carried out inside of a climatic chamber, model CCM 81 from DYCOMETAL [DYC]. This climatic chamber allows to control the temperature and humidity with a precision of  $0.1^{\circ}\text{C}$  and 0.1% respectively. In addition, this chamber is a Faraday cage. A special black blanket [Thoa] was used to prevent external photons from reaching the SiPM.

First, the quenching resistance and the breakdown voltage of the SiPM were obtained from the measurement of the current-voltage curves of the SiPM with the bias voltage applied in forward and reverse direction, respectively. This measurement was done without the amplification of the electronic board to achieve a better precision in the dark. The output current of the SiPM was directly measured using the Keithley 6487 Picoammeter/Voltage Source [KEI]. The LabView software was used to take the data. The measured parameters are plotted as a function of the bias voltage in Figure 4.19.

As can be seen, when the bias voltage is applied in forward direction, Figure 4.19a, the output current of the SiPM does not flow until the



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



**Figure 4.19** – I-V curves measured for the SiPM S13360-1375 model with the bias voltage applied in a) forward direction b) reverse direction. Measurements taken at  $T = 25^\circ\text{C}$  and humidity  $H = 45\%$ .

potential difference between the n and p layers is reached, which is approximately  $V_0 = 0.7$  V for silicon photosensors, close to the value experimentally obtained,  $V_0 = 0.5$  V. When the current start to flow, the intensity is linear with the applied voltage. The equivalent resistance,  $R_{eq}$ , was determined from,

$$I = \frac{1}{R_{eq}}V; \quad \frac{1}{R_{eq}} = \sum_{i=1}^N \frac{1}{R_{qi}} = \frac{N}{R_q} \quad (4.5)$$

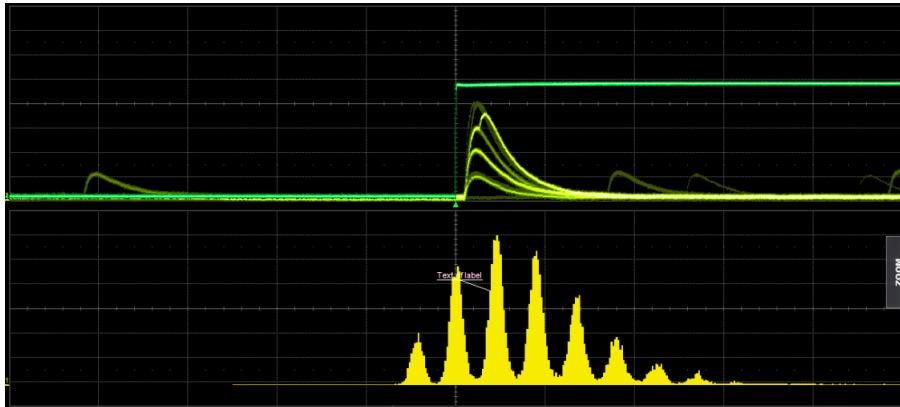
and  $R_{iq}$  are the quenching resistance of each pixel of the SiPM in parallel which have the same value,  $R_q$ . A value of  $R_q = 360.56 \pm 0.07$  k $\Omega$  was obtained, which is in agreement with the typical values given by Hamamatsu.

The breakdown voltage,  $V_{BD}$ , was obtained from the reverse bias voltage plot. This is the points at which the output current of SiPM start to flow, which can be calculated from the maximum of the function

$$f = \frac{1}{I} \frac{dI}{dV} \quad (4.6)$$

The value obtained,  $V_{BD} = 51.02$  V, is in agreement with the value provided by Hamamatsu, Table 3.3.

To measure the SiPM gain,  $G_{SiPM}$ , the electronic board described in section 3.2.4 with an amplification factor of  $F_{amp} = 170$  was used. An incoherent light source, LED435-03 from Toithner LaserTechnik GmbH [Gmb10], described in section 4.1.3, was used to illuminate the SiPM with a low enough flux of  $\lambda = 435$  nm photons. The SiPM output signal shows various well-defined pulse heights, shown in Figure 4.20, corresponding to the number of pixels simultaneously fired. The single photon spectrum, SPS is plotted in Figure 4.20. This spectrum was obtained by integrating and hitogramming the SiPM output pulses with time windows wide enough to contain the full charge of the pulse. The time windows used in these measurements was  $t_w = 500$  ns. The light source gave a trigger signal for the measurement, green line in Fig 4.20.



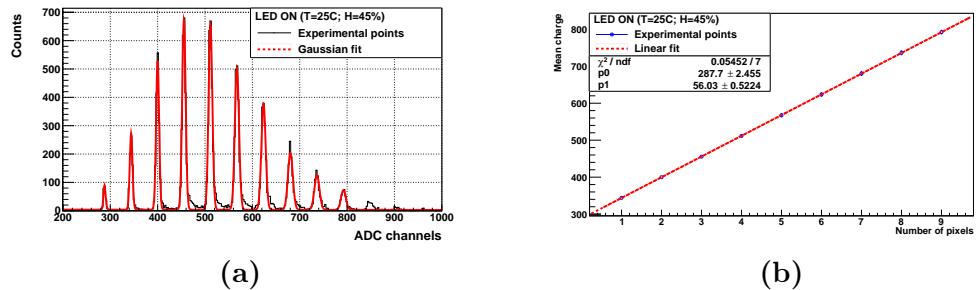
**Figure 4.20** – Above) Trigger signal (green) and SiPM output pulses (yellow). Below) SPS spectrum obtained by integrating and histogramming the SiPM output pulses. This measurement was done at 25°C,  $V_{bias} = 53.98$  and humidity of  $H = 60\%$ .

The well-separated peaks in the SPS spectrum correspond to the charge produced by a different number of fired pixels. The first peak in the spectrum is the pedestal, which is the charge measured when no pixel is fired. This peak is caused by the electronic noise of the system. The second peak corresponds to one fired pixel and so on. The SiPM gain,  $G_{SiPM}$ , can be obtained from the SPS spectrum from the equation,

$$G = \frac{\overline{\Delta Q}(V \cdot s)}{F_{amp}(V/A) \times e^-(C)} \quad (4.7)$$

where  $e^-$  is the electron charge and  $\overline{\Delta Q}$  is the average peak distance in the SPS spectrum, corresponding to the charge released by a fired pixel.

To obtain the value of  $\overline{\Delta Q}$  a macro was written in ROOT [Col21b]. This macro finds and extract the background, which is crucial in some cases like high temperatures or high bias voltages. After that, this macro find all peaks in the SPS spectrum and fits each one to a Gaussian function, shown in Figure 4.21a. The value and error of the charge produced by multiple fired pixels are obtained from the centroid and the sigma of the different



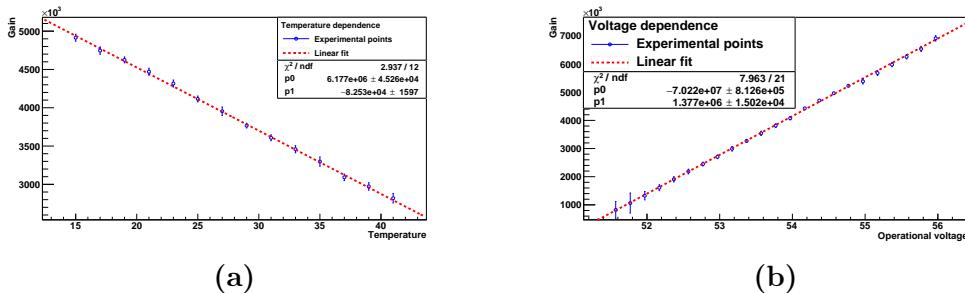
**Figure 4.21** – ROOT analysis performed to obtain the SiPM gain. a) Fit of the SPS spectrum to various Gaussian functions. b) Charge of successive number of pixels as a function of the number of pixels fired. Error bars are within point size. This experience was carried out at  $T = 25^\circ\text{C}$ ,  $V_{bias} = 53.98$  V and humidity of  $H = 45\%$ .

fitted Gaussian functions. The obtained charges are fitted to the number of fired pixels, Figure 4.21b.

Up to 10 simultaneously fired pixels were obtained with a relative uncertainty of the charge measurement of less than 2%. The slope of the straight line in Figure 4.21b corresponds to  $\overline{\Delta Q}$ .

The value obtained for the SiPM gain is  $G_{SiPM} = 4.11 \cdot 10^6$ , very close to the value provided by Hamamatsu, Table 3.3.

A stabilization method for the SiPM gain was implemented. This is necessary for the TRITIUM project since the temperature in the final location of the tritium detector cannot be controlled with enough sensitivity to avoid variations of the SiPM gain. This method consists of compensating for variations in the SiPM gain caused by variations of temperature by controlled variations of the bias voltage. For this task, first, the dependence of the SiPM gain with the temperature and bias voltage was measured. The SiPM gain was measured at several temperatures from  $15^\circ\text{C}$  to  $41^\circ\text{C}$  in steps of  $2^\circ\text{C}$ , which is expected to be the temperature range in the final location. The bias voltage was  $V_{bias} = V_{BD} + 3$ . The SiPM gain was measured at



**Figure 4.22** – Dependence of the SiPM gain with the a) Temperature b) Bias voltage.

several overvoltages from 1 V to 5 V in steps of 0.2 V. The temperature was  $T = 25^\circ\text{C}$ . Both measurements are shown in Figure 4.22.

As can be seen, an excellent linear trend is obtained for both cases. The parameters of the linear fit obtained are,

$$\begin{aligned} G_{SiPM} &= a \cdot T + b; & G_{SiPM} &= c \cdot V_{bias} + d \\ a &= (-82.53 \pm 1.59) \cdot 10^3; & c &= (137.72 \pm 1.50) \cdot 10^4 \\ b &= (617.65 \pm 4.53) \cdot 10^4; & d &= (-762.16 \pm 8.13) \cdot 10^5 \end{aligned}$$

In addition, the breakdown voltage,  $V_{BD}$ , and the terminal capacitance,  $C_t$ , can be obtained from the linear fit of the SiPM gain as a function of the bias voltage,  $V_{bias}$ . Both parameters can be obtained from the definition of the SiPM gain and taking into account that the charge produced in a pixel is proportional to the capacitance of the pixel and the difference voltage in the SiPM,  $V_{OV}$ ,

$$G_{SiPM} = \frac{Q_{pixel}}{e^-} = C_d \frac{V_{bias} - V_{BD}}{e^-} = c \cdot V_{bias} + d \quad (4.8)$$

where  $C_d$  is the pixel capacitance.

From the linear fit obtained in Figure 4.22b, a value of  $V_{BD} = 50.98 \pm 0.59$  V and  $C_d = 220.63 \pm 2.41$  fF are obtained. The terminal capacitance of the SiPM can be calculated assuming all pixels in parallel,  $C_t = N_p \times C_d = 62.88 \pm 0.69$  pF. Both parameters, the breakdown voltage and the terminal capacitance, agree with the values provided by Hamamatsu, Table 3.3.

Finally, the value of the bias voltage to be applied to compensate for the variation in the SiPM gain due to a variation of the temperature can be obtained by applying variations to linear relations:

$$\begin{aligned} G_{SiPM} &= a \cdot T + b \longrightarrow \partial G_{SiPM} = a \partial T \\ G_{SiPM} &= c \cdot V_{bias} + d \longrightarrow \partial G_{SiPM} = c \partial V_{bias} \end{aligned}$$

Therefore, the total variation of the SiPM gain, which is produced by the variation of both parameters, must be cancel:

$$\begin{aligned} \partial G_{SiPM,tot} &= \partial G_{SiPM}(T) + \partial G_{SiPM}(V_{bias}) = 0 \\ \partial G_{SiPM}(V_{bias}) &= -\partial G_{SiPM}(T) \longrightarrow c \partial V_{bias} = -a \partial T \\ \partial V_{bias} &= -\frac{a}{c} \partial T = e \partial T \end{aligned}$$

where the parameter  $e = 59.93 \pm 1.33$  mV/°C is the ratio of  $a$  and  $c$  and agrees with the value of the temperature coefficient provided by Hamamatsu, Table 3.3. Finally, integrating this expression, we obtain:

$$\int_{V_i}^{V_f} \partial V_{bias} = e \int_{T_i}^{T_f} \partial T \longrightarrow \Delta V_{bias} = e \Delta T \quad (4.9)$$

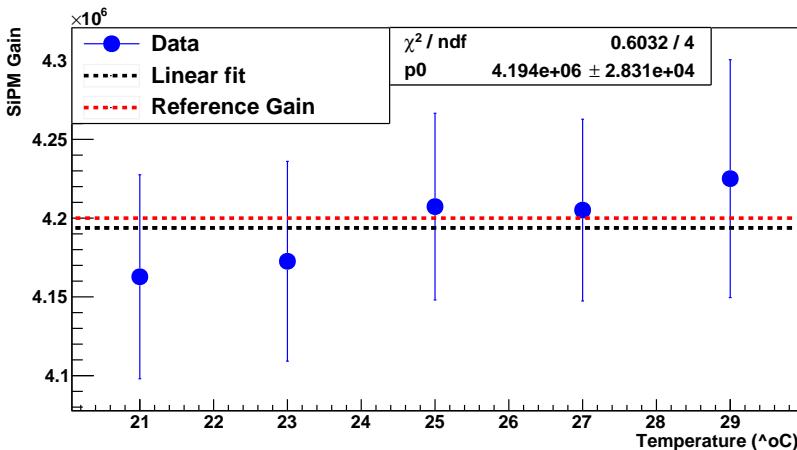
This equation gives the variation of the voltage,  $\Delta V_{bias}$ , that keeps the SiPM gain when there is a variation,  $\Delta T$ , of the temperature. More relevant is to know the bias voltage,  $V_{bias}$  to be applied as a function of the temperature,  $T$ . For this, it is necessary a reference situation. In our case, the reference situation considered is  $V_i = V_{ref} = V_{BD} + 3$  V = 53.98 V and  $T_i = T_{ref} =$

$24^{\circ}\text{C}$ , at which the gain is  $4.2 \cdot 10^6$  (experimentally measured). Thus, we get:

$$(V_{bias} - V_{ref}) = e(T - T_{ref})$$

$$V_{bias}(\text{V}) = 59.9 \cdot 10^{-3} \cdot T(^{\circ}\text{C}) + 52.54 \quad (4.10)$$

Finally, the stabilization method of the SiPM gain developed was tested. The temperature was varied from  $21^{\circ}\text{C}$  to  $29^{\circ}\text{C}$  and the bias voltage was modified according to the equation 4.10. The value of the SiPM gain obtained is shown in Figure 4.23 as a function of the temperature.



**Figure 4.23** – SiPM gain as a function of the temperature with the stabilization method.

A red dotted line is included, indicating the value of the SiPM gain to be kept. The SiPM gain is maintained fairly well by this method.

## 4.3 Characterization of the Ultrapure Water System

This section describes the characterization of the ultrapure water system, that guarantees that the quality of the water sample 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 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. The tritium activity should not be affected by the water purification process.

To verify that these requirements are fulfilled, 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

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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)

the water was measured by a physico-chemical analysis, shown in Table 4.7, before the ultra-purification process. The water sample contains a number of components, that must be removed to prevent their deposition on the scintillating fibers of the detector.

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.

The water turbidity<sup>8</sup> was measured using the Hanna Hi 9829 portable multiparameter system from Hanna Instruments [Ins], obtaining a value of 29 NTU, much higher than the WHO recommended limit of 5 NTU for drinking water. The water conductivity was also measured for both, raw and purified water, using the same system. The results of the conductivity measurements, together with the measurement of the rejected water, described in section 3.3.2, are presented in Table 4.8. 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 re-

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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.

quirement. 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 the purified water.

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.

The gamma radioactive elements present in both, raw 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 absent in the purified water.

The tritium activity was measured by liquid scintillation counting (LSC) to check if the ultra-purification process had modified it. 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 seen in the table, 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 purified water.

## 4.4 Characterization of the TRITIUM Cosmic Veto

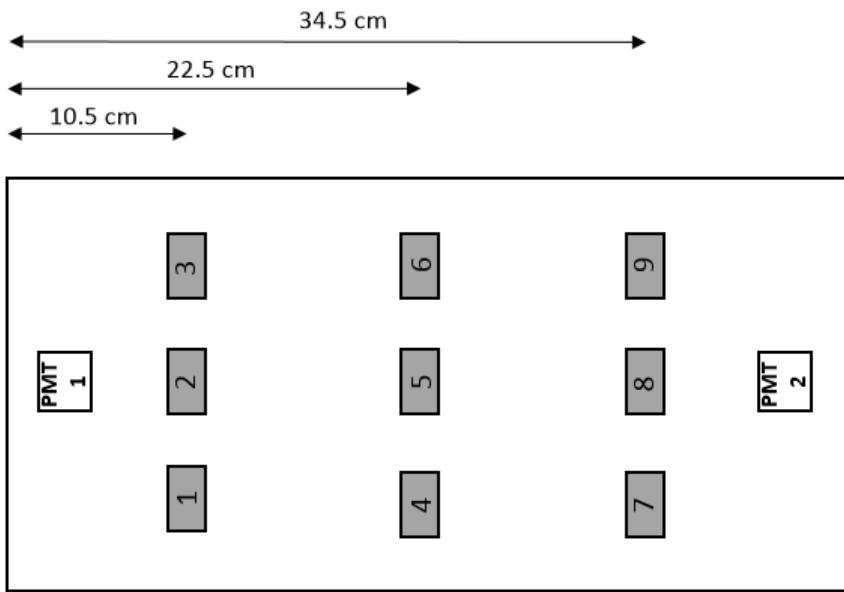
This last section reports on the characterization of the active shield (cosmic veto), which was carried out using PMTs as photosensors. Measurements of the cosmic veto using SiPM arrays has already started and their replacement will be as soon as possible.

The quality of the veto wrapping, shown in Figure 3.27, was checked. This study was done at the level of one detector so the configuration of the electronics is the one given in Figure 3.15b. The surface of the veto was divided in 9 parts, shown in Figure 4.24, in which a gamma source was placed.

Two different tests were made for this task:

1. The improvement of the veto signal due to wrapping. A  $^{137}\text{Cs}$  source was placed at point 2 and a energy spectrum was measured with the veto before wrapping. Next, the measurement was repeated after wrapping. The spectra obtained are shown in Figure 4.25.

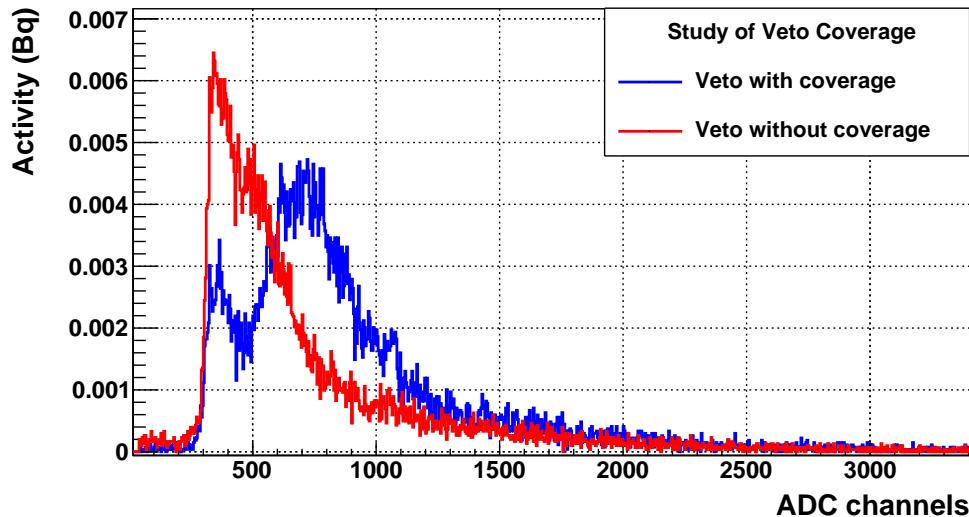
The spectrum of the wrapping veto is shifted to the right, which means that more photons are collected per event. No improvement was obtained in the number of events detected, only in the collection efficiency.



**Figure 4.24** – Reference points used for veto mapping.

2. The spatial uniformity of the signal in the wrapped veto was evaluated. 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 Figure 4.26. It can be observed that the veto signal has a uniform response on its whole surface, giving a fairly similar counting rate in all the points measured.

Next, both vetos in time coincidence was studied, so the configuration of the used electronics was that of Figure 3.15c. The goal was 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,



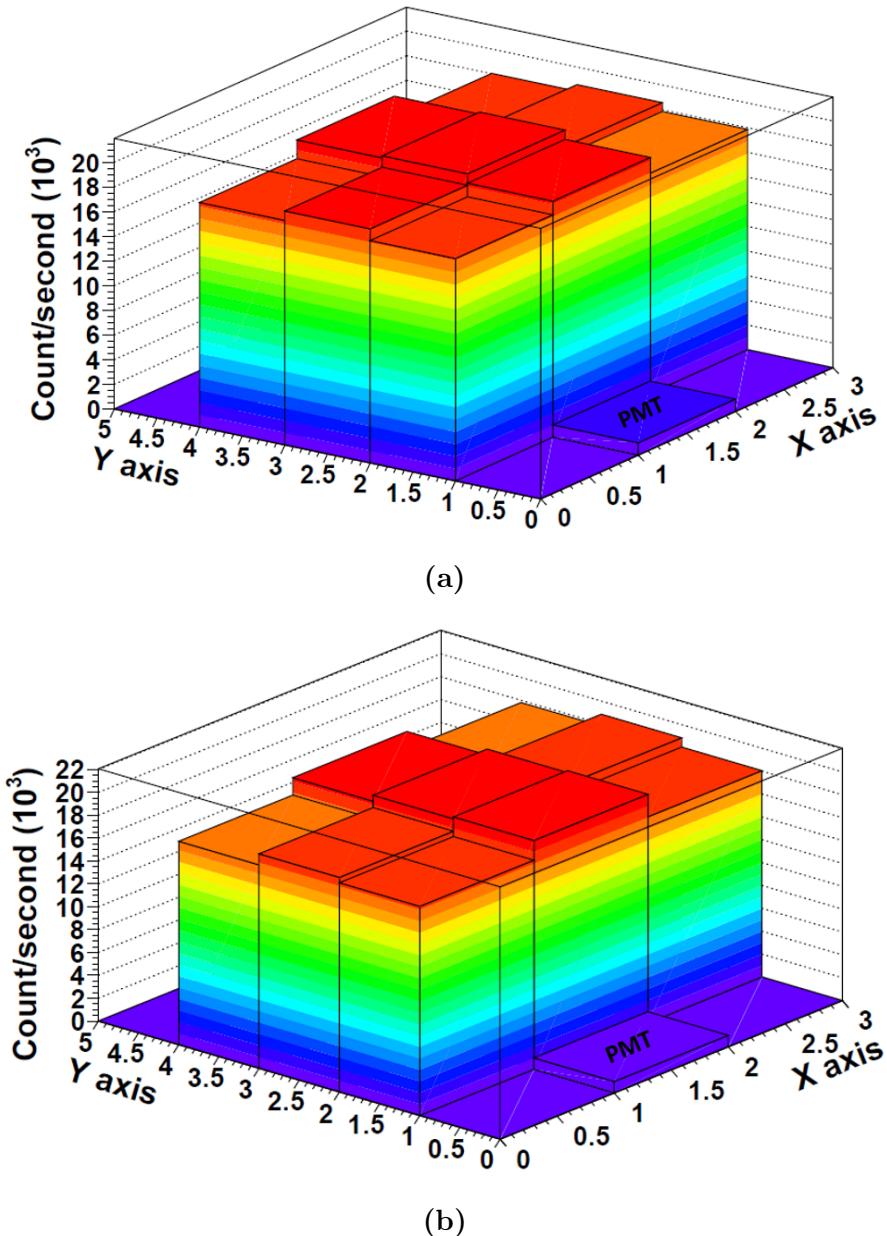
**Figure 4.25** – Measurement of a radioactive source  $^{137}\text{Cs}$  with the TRI-TIUM cosmic detector with and without wrapping.

finding the maximum threshold of the discriminator<sup>9</sup> before starting to loss cosmic events. For higher voltages and a smaller thresholds, a plateau of the counting rate should be obtained.

The counting rate was measured for several high voltages at fixed threshold and for several thresholds at fixed high voltage, plotted in Figure 4.27. To find the optimal conditions, the amplification line of the electronics 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 4.27a, the counting rate at several high voltages for three different thresholds, 60 mV, 100 mV and 200 mV is plotted. 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

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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



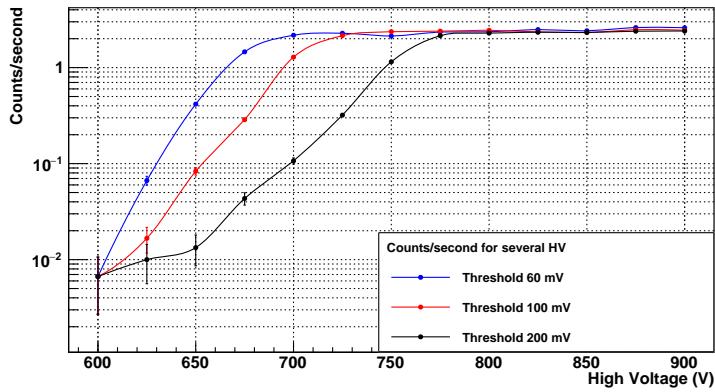
**Figure 4.26** – Bidimensional graph of the count rate (Mapping) measured with two different TRITIUM cosmic detectors using a radioactive source of  $^{60}\text{Co}$ .

when the value of the threshold increases, as it should. 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, plotted in Figure 4.27b. There is a maximum threshold for every high voltage used, 140 mV, 270 mV and 450 mV respectively, at which the plateau ends. This maximum threshold increases with high voltage, as it should. The voltage chosen was 800 V since it is on the plateau for the three thresholds and the threshold chosen was 200 mV which is on the plateau for the selected high voltage.

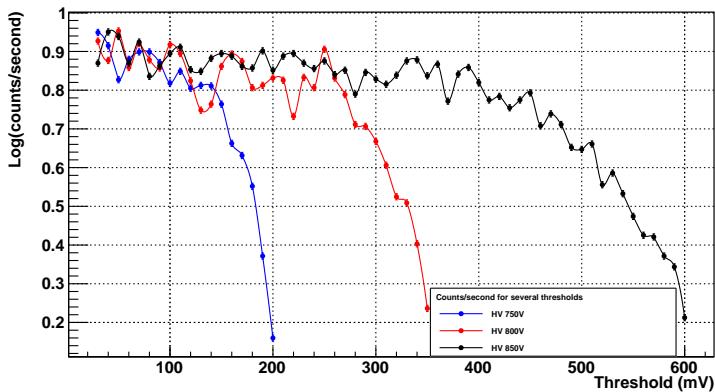
With this setting, the energy spectrum of cosmic events was measured, shown in Figure 4.28.

As expected, this energy spectrum fits well to a Landau function. The cosmic ray rate determined from the area of this spectrum 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 event detection is 85%, which is a usual value for the efficiency of plastic detectors to mips.

Finally the detected cosmic ray rate versus the distance between the two 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 4.29a. The energy spectrum in Figure 4.28 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 4.29b, was fitted to a second degree polynomial which allows to estimate the cosmic rate for a given veto distance.

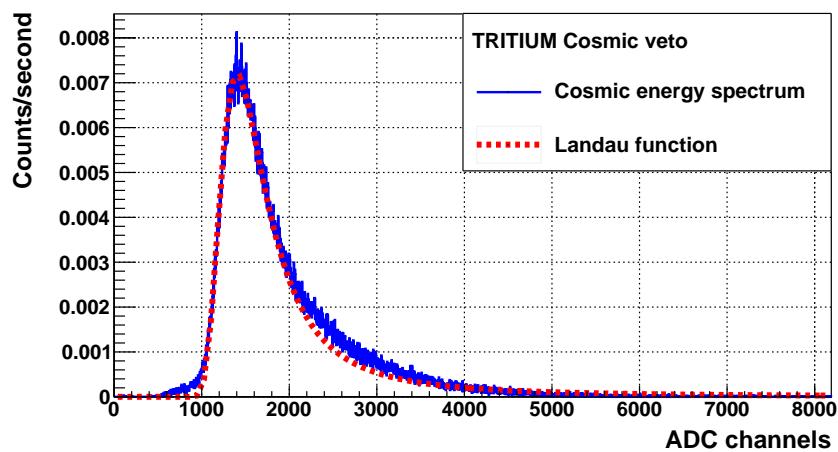


(a)

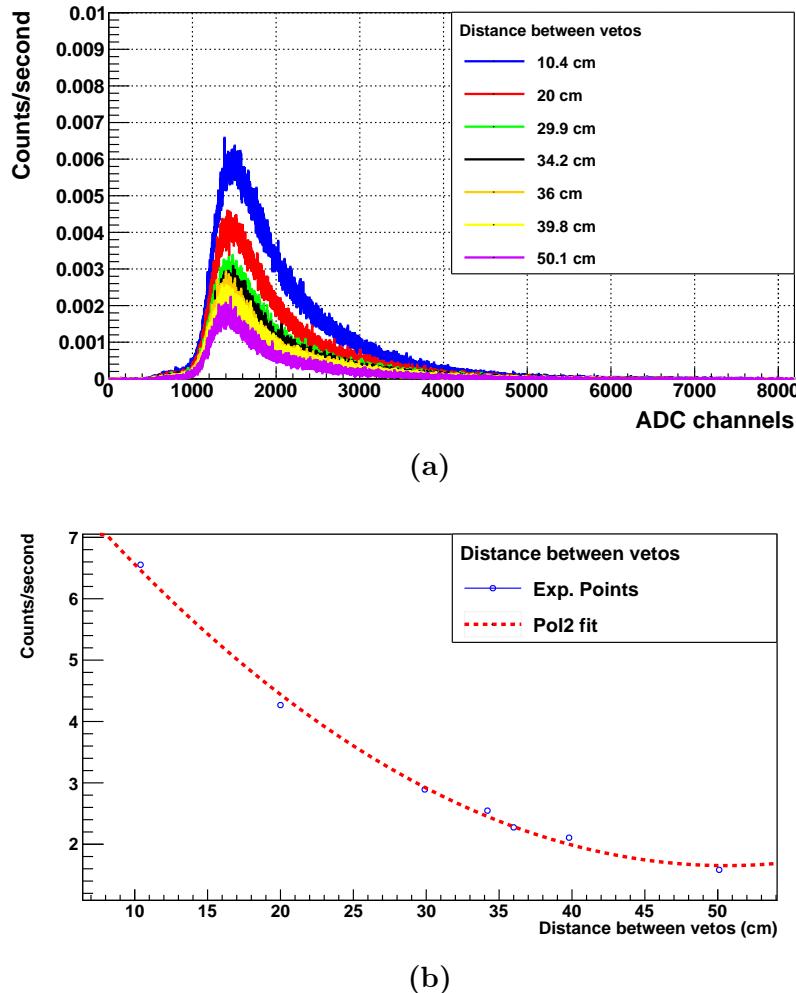


(b)

**Figure 4.27** – Counting rate a) as a function of high voltage for fixed thresholds and b) as function of thresholds for fixed high voltage. in semilogarithmic scale



**Figure 4.28** – Energy spectrum measured with the cosmic veto.



**Figure 4.29** – Measurement of the cosmic veto for several distances between the two scintillators. Above) Energy spectrum of the cosmic veto for several distance. Below) Fit of the cosmic veto rate versus distance to a second degree polynomial.

# Chapter 5

## TRITIUM Monitor Prototypes

This chapter describes the different prototypes developed in the framework of the TRITIUM experiment, which are TRITIUM-IFIC 0, TRITIUM-IFIC 1, TRITIUM Aveiro and TRITIUM-IFIC 2, listed in chronological order of their construction. The first two prototypes built, TRITIUM-IFIC 0 and TRITIUM-IFIC 1, are preliminary prototypes used to learn about tritium detection and to detect and solve problems in their designs. The other two prototypes built, TRITIUM-Aveiro and TRITIUM-IFIC 2, are prototypes with a design in which no significant problems were found. They were built to check more subtle effects.

Each prototype was designed and built in the laboratories of IFIC or Aveiro and it was filled with tritiated water following a protocol specially developed for this task. Several water tightness and filling tests were carried out for each prototype to guarantee its radiosecurity. At the end of the chapter, the final monitor of TRITIUM detector will be described. Its design is a modular structure for easy scalability, composed of as many units of the final prototype as needed to reach the required sensitivity.

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

Two preliminary prototypes, TRITIUM-IFIC 0 and TRITIUM-IFIC 1, are reported in this section, which were designed, developed and built at the IFIC workshop. These prototypes were thought to be a small scale proof of concept of the final TRITIUM detector module and they helped to learn about tritium detection, to detect design problems and to implement improvements for the final prototype.

### 5.1.1 TRITIUM-IFIC 0

The TRITIUM-IFIC 0 prototype was the first prototype developed in TRITIUM experiment to check the feasibility of the technology proposed by TRITIUM, that is, to verify that the possibility of detecting tritium in water with good sensitivity using scintillating fibers.

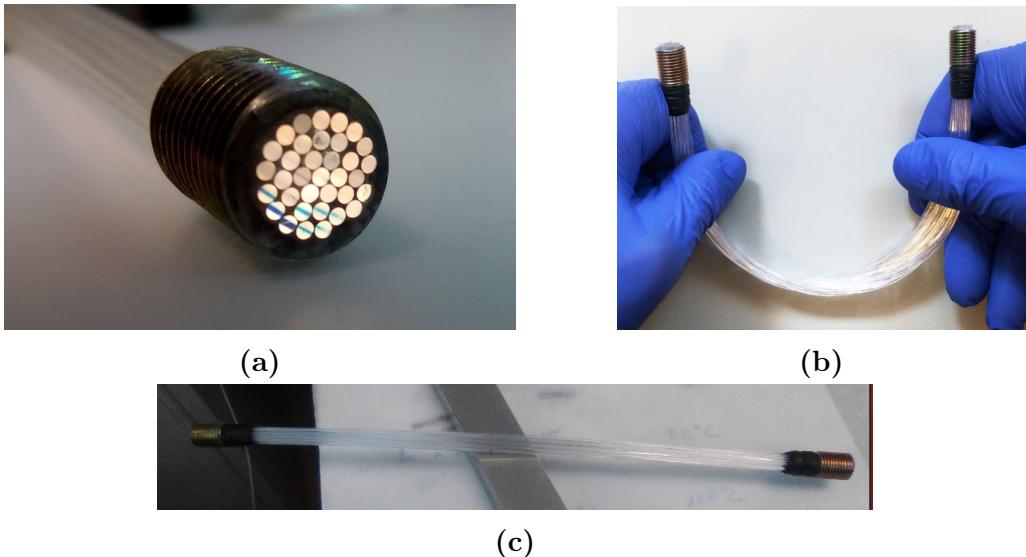
As liquid radioactive sources were involved, the design of this first prototype paid special attention to radiation safety.

TRITIUM-IFIC 0 consists of bundle of 35 fibers, shown in Figure 5.1, with a length of 20 cm, which were cleave and polished with the techniques reported in section 4.1.1. This bundle has metallic pieces located in both ends, shown in Figure 5.1a to attaching it to the prototype vessel.

The fiber bundle was placed inside of a vessel, made of 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 was not efficient for tritium detection as we learned afterwards. As can be seen in Figure 5.2, a frame of methacrylate and steel was designed and built

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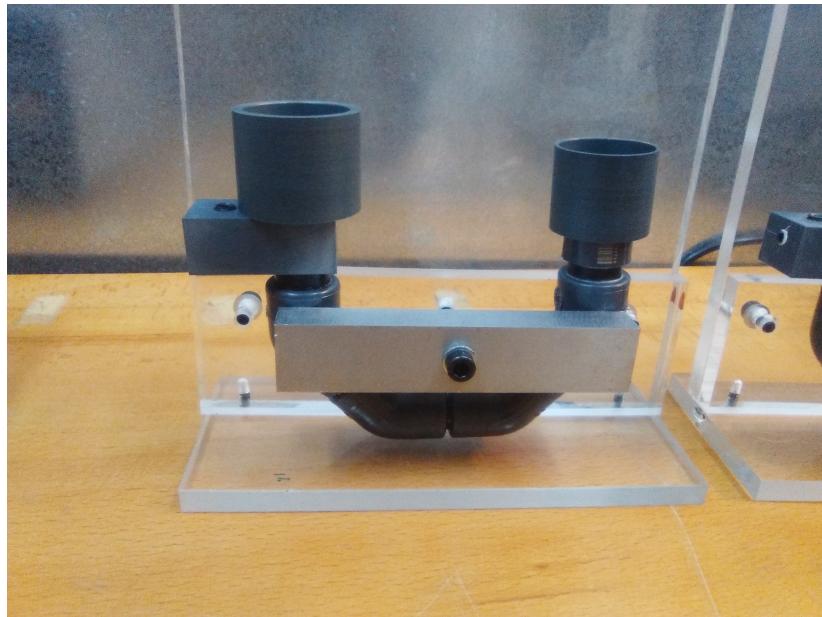
<sup>1</sup>Polyvinyl Chloride, PVC



**Figure 5.1** – a) Metallic piece of the fiber bundle. b) and c) Bundle of 35 fibers, the length of which is 20 cm, used in TRITIUM-IFIC 0 prototype.

to hold the prototype. Two calibrated PMTs were optically coupled to the fiber bundle ends using optical grease [Cera]. The employed PMTs were the model R8520-460 from Hamamatsu company [K.K19] and the high voltage between the dynodes was distributed by the electronic circuit show in fig 3.9. The 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 processed and analyzed by the electronics shown in Figure 3.15b.

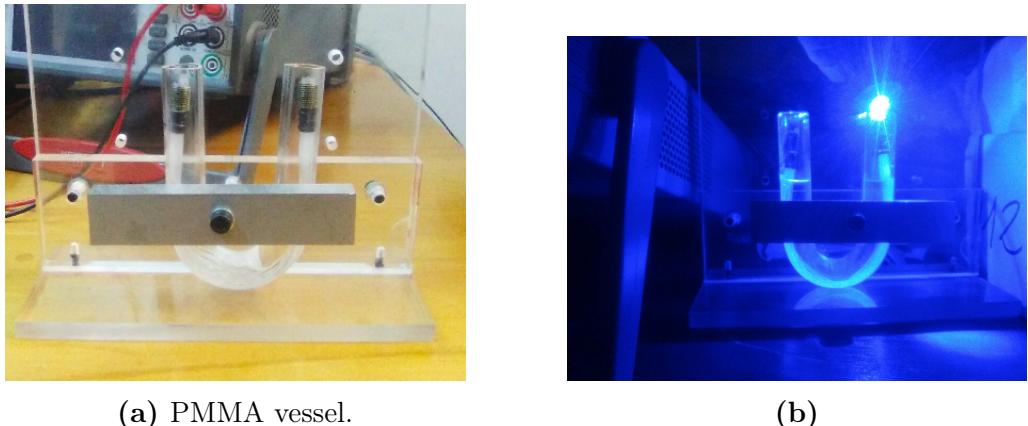
Two identical prototypes were built and filled following the same protocol. The first prototype, called TRITIUM-IFIC 0 Background, was filled with ultrapure water (39 mL, uncertainty of 0.05%) and it was used to measure the radioactive background of the detector whereas the second prototype, called TRITIUM-IFIC 0 Signal, was filled with a radioactive liquid source of tritium, the preparation of which is reported in the appendix C. The specific activity of the liquid source employed was 99.696 kBq/L



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

(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 was approximately  $3.888 \pm 0.087$  kBq. This second prototype was used to measure the signal of the detector (tritium + background), The measured tritium activity was determined by subtracting the background from the signal.

A statistically significant number of time coincident events was not found, so the measurement of time coincidences was not possible. The loss of photons could be caused for several reasons, such as an excessive curvature of the fiber bundle due to the U-shape of the PVC vessel, causing most of the photons escape from the fibers, or the poor quality of the tritiated water-fiber interface (the cleaning process described in section 4.1.4 was motivated by this result). To avoid this problem and to obtain some data with this prototype, a measurement was performed with a single PMT. For this task, the electronic configuration shown in Figure 3.15a was used. The



(a) PMMA vessel.

(b)

**Figure 5.3 – a)** Glass vessel c) Test carried out to check photon loss due to fiber bundle curve.

results of these measurements are reported in section 7.1.1.

An additional test was carried out to find an explanation of the absence of time coincident events in the data. A transparent glass vessel was built similar to the TRITIUM-IFIC 0 prototype vessel, shown in Figure 5.3a, to study the effect of the fiber bundle curvature. The LED described in section 4.1.3 was used to verify the reduction in photocollection efficiency of the fiber bundle due to this curvature. As can be seen in Figure 5.3b, a large percentage of the photons are lost due to the curvature. This problem lead us to use a straight fiber arrangement in next prototypes.

### 5.1.2 TRITIUM-IFIC 1

The next prototype, TRITIUM-IFIC 1, was designed to overcome the problems and limitations found in TRITIUM-IFIC 0. The main improvements were:

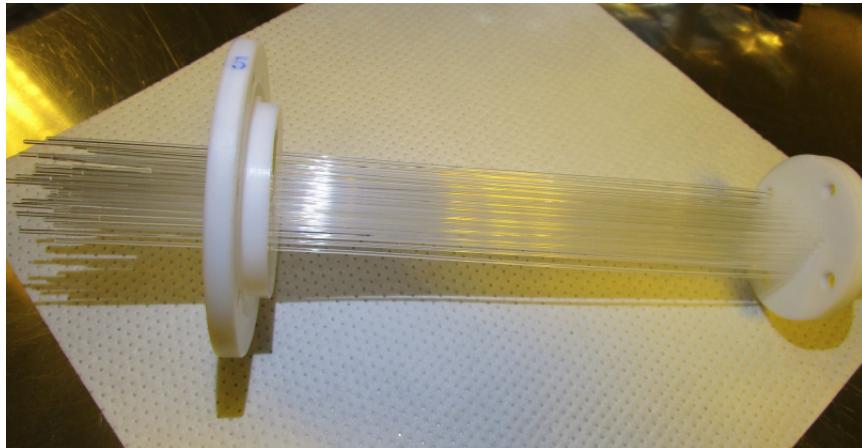
1. The fiber bundle was arranged straight to optimize the photon collec-

tion efficiency of the fibers.

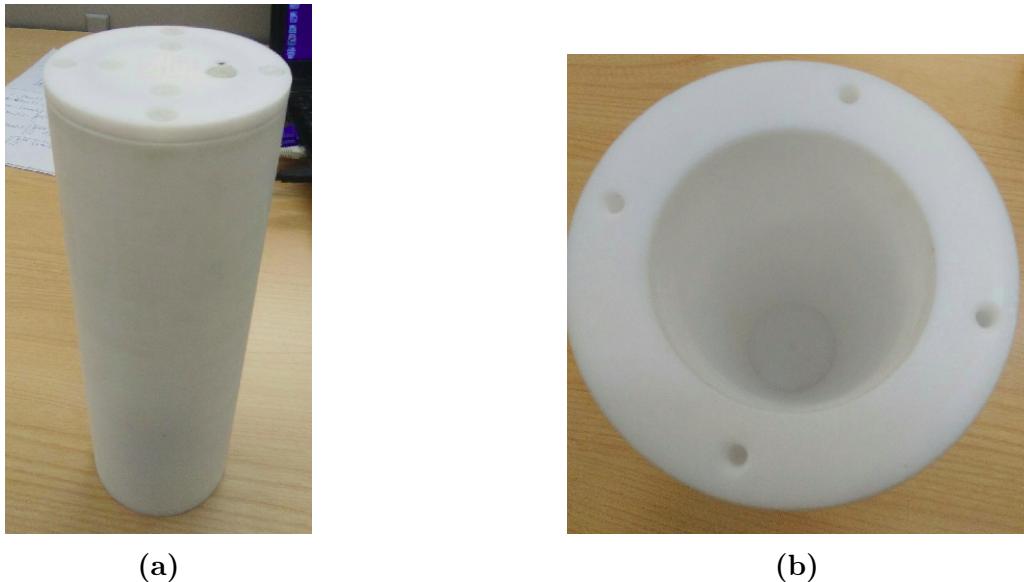
2. A special fiber cleaning protocol, described in section 4.1.4, was applied to the fibers to improve the interfaces between fiber and tritiated water. This protocol produces a better wetting property of the fiber, which improves the photon collection efficiency of the scintillating fibers.
3. A Teflon vessel was used in the Tritium prototypes to improve the photon collection efficiency of the fibers, which is an intrinsic characteristic of the fiber that cannot be changed.

Teflon is a convenient material for its optical properties, specifically its reflection factor, which is very close to 100% at the working wavelength. This means that the photons that escape from the fiber will hit the Teflon walls and go back to the scintillating fiber.

The TRITIUM-IFIC 1 prototype consists of 64 straight scintillating fibers, of 20 cm length, arranged within a Teflon structure in an  $8 \times 8$  square matrix, shown in Figure 5.4.



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



**Figure 5.5** – Teflon vessel of TRITIUM-IFIC 1 prototype.

This structure is placed within a cylindrical Teflon vessel of 48 mm diameter and 200 mm length, shown in Figure 5.5.

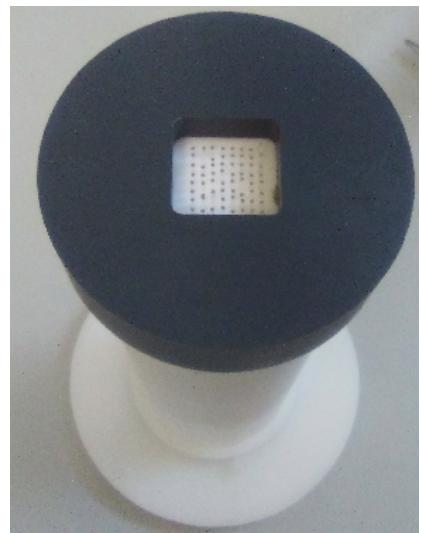
The cleaning process, described in section 4.1.4, was applied to the fibers to achieve a better tritiated water-fiber interface. A PVC piece was used to attach the photosensor to the prototype and prevent external light from being read by photosensors. A general view of this prototype is shown in Figure 5.6.

The prototype was read by a PMT R8520-460, from Hamamatsu Photonics company [K.K19] coupled directly to the fiber bundle using optical grease [Cera]. The electronic circuit, shown in Figure 3.9, was used to distribute the high voltage between the dynodes. The high voltage was  $-800$  V, which corresponds to a quantum efficiency of 28.66%.

The signal from this PMT was acquired by the same electronic configuration used for the TRITIUM-IFIC 0 prototype, shown in Figure 3.15a.



(a)



(b)

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

Unlike the first prototype, only one TRITIUM-IFIC 1 was built. In a first measurement, this prototype 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 with 118 mL (uncertainty of 0.05%) of a tritiated water source of the same activity as the one used for TRITIUM-IFIC 0 prototype, 99.696 kBq/L. The results of these measurements are presented in section 7.1.2, where they are compared to TRITIUM-IFIC 0 prototype results.

## 5.2 Latest TRITIUM Prototypes

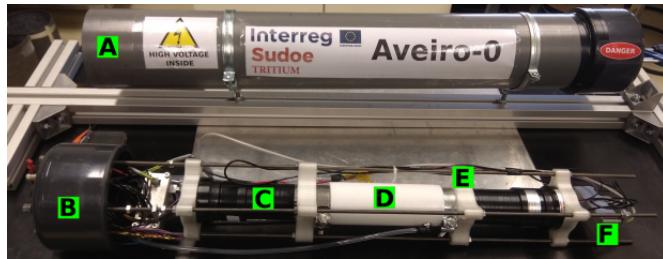
This section reports on prototypes TRITIUM-Aveiro and TRITIUM-IFIC 2. A different design was employed in both prototypes so that they can safely allow the reading of a large number of fibers arranged in a straight position with two photosensors in time coincidence.

Particular attention to tritium detection efficiency was paid in these prototypes, obtained by the use of more fibers than the preliminary prototypes and time coincidences of the photosensors. Furthermore, the activity of the radioactive liquid source of tritium used to fill these prototypes is much lower than that of the first prototypes to be able to measure their low detection level.

The design of both prototypes is quite similar and their objective is to test the importance of to decide the final design of the TRITIUM monitor.

### 5.2.1 TRITIUM-Aveiro

The third prototype built as a proposal of the final TRITIUM detector module was TRITIUM-Aveiro, shown in Figure 5.7, designed and built in the workshop of the University of Aveiro.



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

This prototype consists of a Teflon vessel (marked as D in Figure 5.7), shown in Figure 5.8, with an internal cylindrical hole of 43 mm diameter and 18 cm length.



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

This vessel contains 360 unclad scintillating fibers of 180 mm length. The fibers are BCF-10 from Saint-Gobain company [Cer01], which have similar characteristics than the BCF-12 fibers, except the diameter,

which is the double, 2 mm. A larger diameter is convenient because it facilitates the flow of water around the fibers, reducing problems related to surface tension and ensuring that the entire active volume of the fibers is used for tritium detection. In addition, a large radius increases the rigidity of the fiber. However, this large radius worsens the signal-to-background ratio. The detector active volume for 2 mm fibers is smaller for the same volume, producing a smaller tritium signal and internal volume of the fibers unreachable by tritium decay electrons is larger, producing a larger background. As a result, a lower signal-to-background ratio is obtained. In order to quantify the importance of the fiber diameter, the measurements were compared with similar measurements performed with TRITIUM-IFIC 2 prototype, based on a similar configuration but with 1 mm fibers.

The amount of fibers used in TRITIUM-Aveiro prototype is the maximum which allows the water to flow around the fibers, which are free inside the Teflon vessel.

These fibers were cleaved with the device developed by TRITIUM but they were neither polished nor cleaned because the automatic polishing machine was not yet developed and it was not feasible to polish 360 fibers by hand.

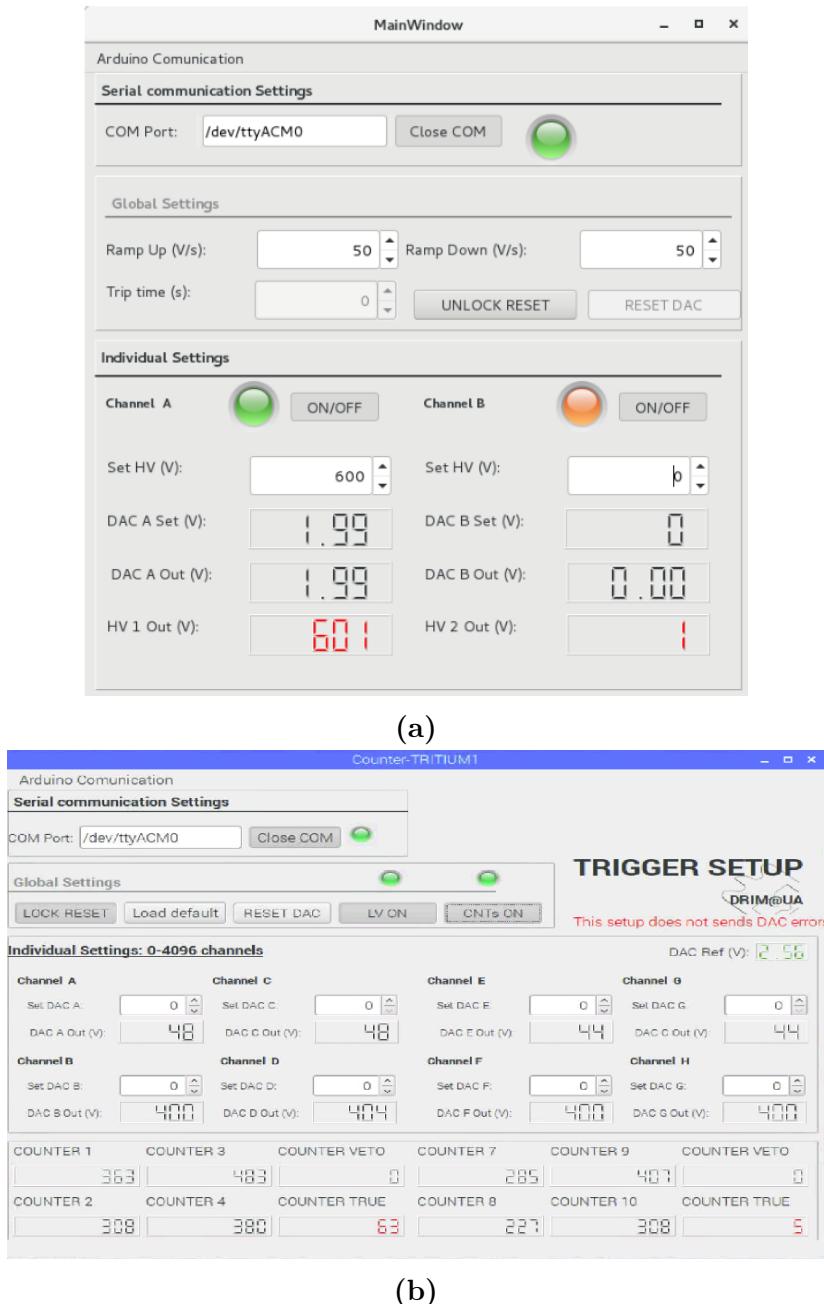
The Teflon vessel is totally closed and a water inlet/outlet were installed in it to allow a constant water flux through it. Two PMMA 10 mm thick windows, located at both ends of the fiber bundle, was used to read the fibers. Two clamps were used to make a tight junction of the Teflon walls and the PMMA. PMMA was chosen for its optical properties, especially its transmission coefficient, which is larger than 95% at the working wavelength. Two PMTs (marked as C in Figure 5.7) were used to read this prototype in time coincidence. HV was  $-1500$  V, at which the quantum efficiency is 26%. PMTs were attached to both fiber bundle ends by two pieces (marked as E in Figure 5.7) built with a 3D printer and they were optically coupled to the PMMA windows through optical grease [Cera]. The PMTs used were

R2154-02 2" from Hamamatsu [K.K10], that have gain and efficiency quite similar to the PMTs used in the other prototypes.

This prototype and its electronics (marked as F in Figure 5.7), were arranged in a structure, shown in Figure 5.7, composed of several clamps and four long stainless-steel screws, locked to an external PVC structure, marked as A and B in Figure 5.7, which protects the prototype from physical damage and provide a light-tight operation environment. This PVC structure is equipped with the necessary feed-through connectors.

Only one prototype was built, which was designed to be installed in the Arrocampo dam and its electronics, based on several PCBs, was specially designed, developed, built and tested to process and analyze the signals of this system, detailed in appendix D. Two interfaces were developed, one to control the PMT power supply, shown in Figure 5.9a, and the other to control the different options of the electronics, such as thresholds and number of counts, shown in Figure 5.9b.

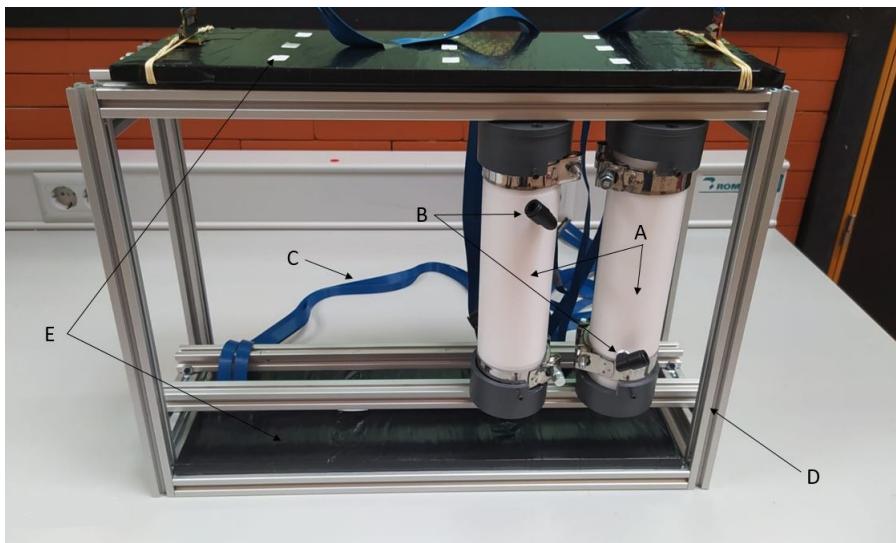
Measurements taken in the laboratory were used to characterize the detector. For this task, the prototype was firstly filled with ultra-pure water, which was used to measure the background of the detector, and next, with a radioactive liquid tritium solution with an activity of 30 kBq/L, which was used to measure the efficiency and the low detection level of the prototype. The volume of ultrapure water and tritium solution used in TRITIUM-Aveiro prototype was 58 mL. This prototype was installed in the Arrocampo dam to test its functionality and to begin with the tritium level monitoring. The laboratory and Arrocampo measurements are reported in chapter 7.



**Figure 5.9 –** Graphical User Interface developed to control a) PMT HV employed. b) the counter system of the TRITIUM-Aveiro prototype

### 5.2.2 Tritium-IFIC 2

The last prototype developed for TRITIUM was TRITIUM-IFIC 2, marked as A in Figure 5.10. This prototype, built in the IFIC workshop, consists of a cylindrical Teflon vessel, shown in Figure 5.11, with a similar shape to that of the TRITIUM-Aveiro prototype. The internal length and diameter of the Teflon vessel were 210 mm and 36 mm respectively.



**Figure 5.10** – TRITIUM-IFIC 2 prototype and active veto within the metallic structure.

This prototype contains 800 unclad BCF-12 scintillating fiber of 200 mm length. This number is larger than that of the TRITIUM-Aveiro prototype and is contained in a smaller volume. The fibers used were cleaved, polished and cleaned with the conditioning process described in section 4.1.

This number of fibers, which were freely arranged, just allowed water to flow through them. Two PMMA windows, located at the ends of the fiber bundle, allowed to read the scintillation light as in the TRITIUM-Aveiro prototype.

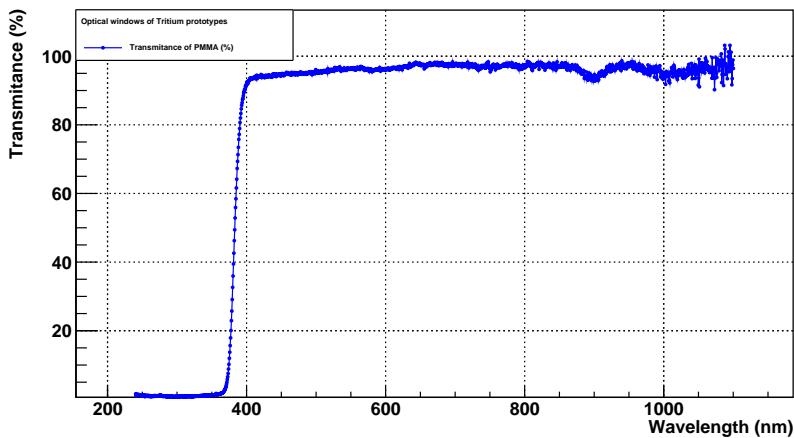


**Figure 5.11** – a) TRITIUM-IFIC 2 Teflon vessel. b) TRITIUM-IFIC 2 Teflon vessel with PVC caps

5 mm width PMMA optical windows is sufficient to guarantee tightness since the detector works at very low water pressure. Two clamps keep the tightness of the prototype, similar to the TRITIUM-Aveiro prototype. PMMA was chosen for its optical properties, especially its transmission coefficient, which was measured for visible spectrum in the ICMOL laboratories, shown in Figure 5.12. This transmission coefficient is approximately 95% for the working wavelength (435 nm). Slightly better transmission coefficients can be achieved with other materials such as quartz or sapphire but they are much more expensive.

A water inlet/outlet was implemented in the Teflon vessel, shown in Figure Figure 5.11, to allow a constant water flux, similar to the TRITIUM-Aveiro prototype.

For the first laboratory measurements, two R8520-460 PMTs from the Hamamatsu Photonics [K.K19], were used to compare the results to



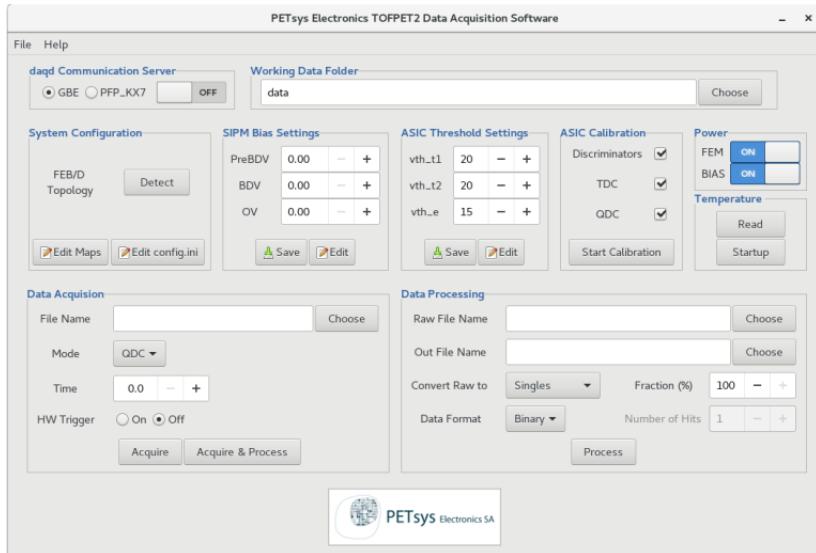
**Figure 5.12** – Transmission spectrum of light by a 5 mm thick of PMMA plate. (measured in the ICMOL laboratory).

those of the previous prototypes. However, measurements of the TRITIUM-IFIC 2 prototype with SiPM arrays and PETSYS electronics have already started since the final version will use them.

PETSYS has a graphical user interface, shown in Figure 5.13. However, they also allow remote control of all the different options such as supply voltage for the SiPM arrays, thresholds, etc., via computer terminal.

Two PVC caps, located at both ends of the prototype were used to provide to the SiPMs a light-tight environment. An aluminum structure was designed and built to house up to 10 TRITIUM-IFIC 2 modules and two cosmic vetos, marked as D in Figure 5.10.

The available space within the lead shielding can accommodate up to 5 structures, which means that the final TRITIUM monitor may accommodate up to 50 TRITIUM-IFIC 2 modules and 5 different cosmic vetos. As the sensitivity of the TRITIUM monitor scales with the number of TRITIUM modules used, the results obtained with the TRITIUM monitor



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

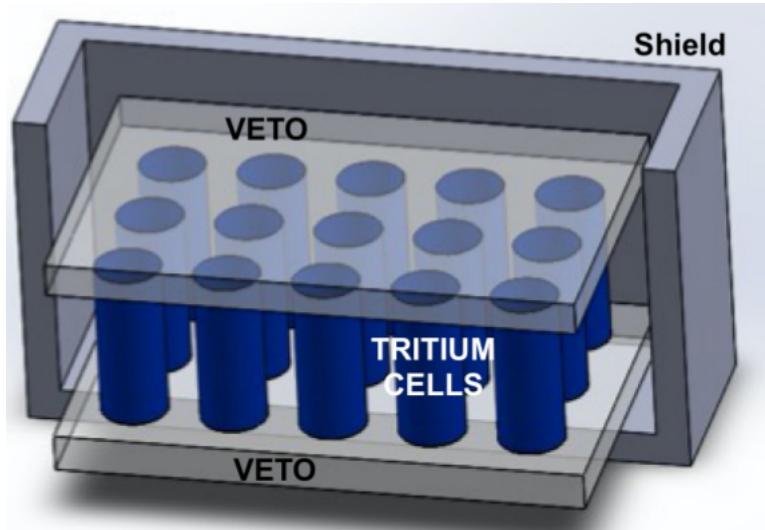
should improve those results by a factor of  $\sqrt{N}$ , where N is the number of modules used.

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 water volume in both cases was 82 mL (uncertainty of 0.05%). The activity of the tritium source used for this prototype was 10 kBq/L (uncertainty of 2.24%), which was prepared by diluting a sample of tritiated water in ultrapure water.

The results provided by this prototype are shown in chapter 7.

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

This section presents the final TRITIUM monitor. A schematic design is shown in Figure 5.14.



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

It consists of a number of TRITIUM modules read in parallel. Each module is the prototypes that achieves better results, TRITIUM-Aveiro or TRITIUM-IFIC 2. These modules are shielded from environmental radioactivity by three different techniques:

1. An external lead shielding, which is used to stop the environmental radioactivity and soft cosmic rays (particles with energies below 200 MeV).
2. Several active vetos, which is placed below and above the TRITIUM modules. These active vetos are read in anticoincidence to suppress

high energy event background, mainly cosmic ray particles with energies above 200 MeV.

3. The radioactive elements present in the water samples, measured by the TRITIUM monitor, are eliminated by the ultrapure water system.

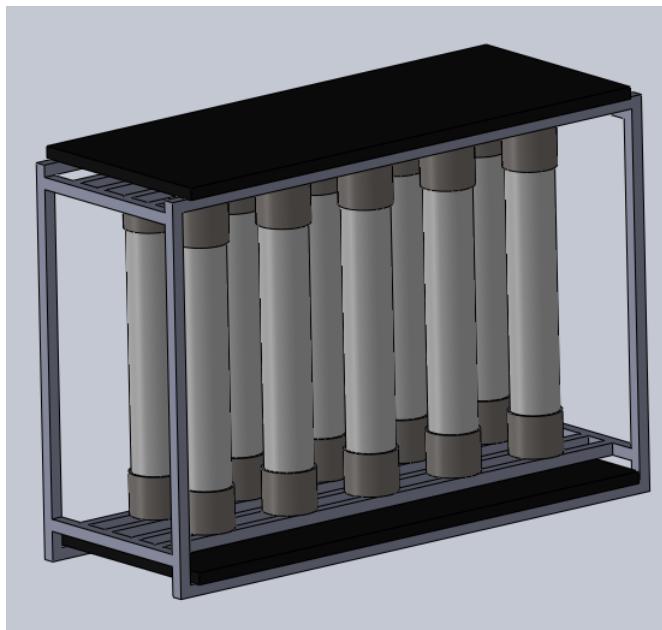
The ultrapure water system, the lead shielding and a TRITIUM-Aveiro prototype are installed and currently in operation at the Arrocampo dam. This entire system is employed to successfully monitor the tritium levels in Arrocampo dam during several months. Furthermore, two additional TRITIUM-Aveiro prototypes and four active vetos are currently under construction and will be measured in parallel with this prototype.

The electronics of the TRITIUM-Aveiro prototype, based on a RaspberryPi, cannot be used for multiple modules due to counting limitations and this must be replaced by an FPGA-based counter board.

Simultaneously, three TRITIUM-IFIC 2 prototypes and an active veto are already built and they will be installed as soon as possible.

One of the most important aspects of the TRITIUM monitor is its modular design, which allows scalability to reach the required sensitivity, 100 Bq/L. It means that if this target sensitivity is not reached with the three modules to be installed, may be obtained by installing additional modules.

The only scalability restriction is the available space, which is set by the lead shield already built and the cabin in which the setup is installed. The currently available space fit five different structures as the one shown in Figure 5.15, each one containing 10 modules and one active veto. If all the 50 TRITIUM modules are installed the sensitivity of the TRITIUM monitor could be improved a factor of around 7 ( $\sqrt{50}$ ) with the respecto to the sensitivity of a single module.



**Figure 5.15** – A TRITIUM monitor design based on the TRITIUM-IFIC 2 prototype.

# Chapter 6

## Simulations

This chapter details the Monte Carlo simulations performed in the TRITIUM experiment to optimize the design of the TRITIUM detector, understand its behavior and investigate its limitations. These simulations are 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. Furthermore, 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. The simulation environment employed was Geant4 [Col21a, Ago03].

### 6.1 Geant4 Environment

Geant4 is a software toolkit for the simulation of the passage of particles through matter developed at CERN, based on object-oriented technology

implemented in the C ++ programming language. Geant4 includes the definition of all the different aspects of the simulation process such as detector geometry, materials used, particles of interest, physical 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. This means that the tritium events are initialized one by one, generating energy, moment, position, etc. The propagation of each tritium decay electron and its interaction with the scintillator is simulated, and 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.

The 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 and polystyrene were taken from the Geant4 NIST database and the other materials were built by specifying their atomic composition. The following optical properties were added to these materials:

1. The refraction index and light attenuation coefficient were added to water, obtained from [Bui94]. A spectrum of tritium decay electrons,

uniformly distributed in the volume, was added to water to simulate tritiated water. The tritium decay spectrum data were taken from [Mer15].

2. The spectra of refractive index, light attenuation and photon emission, obtained from the data sheet of scintillating fibers, [Cer05], were added to the polystyrene. The scintillation yield and the decay time coefficient, also obtained from their data sheet, was included.
3. The quantum efficiency spectrum was added to the photocathode material of the PMTs, taken from their data sheet, [K.K19]. A refraction index of 1.46 was used for 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 [Arg11].

The simulations shown in this thesis are focused on the Tritium-IFIC 2 prototype since these were the simulations I was primarily working on, but similar simulations were performed for the Tritium-Aveiro prototype, which are also summarized.

## 6.2 Simulations to Optimize the TRITIUM Monitor Design

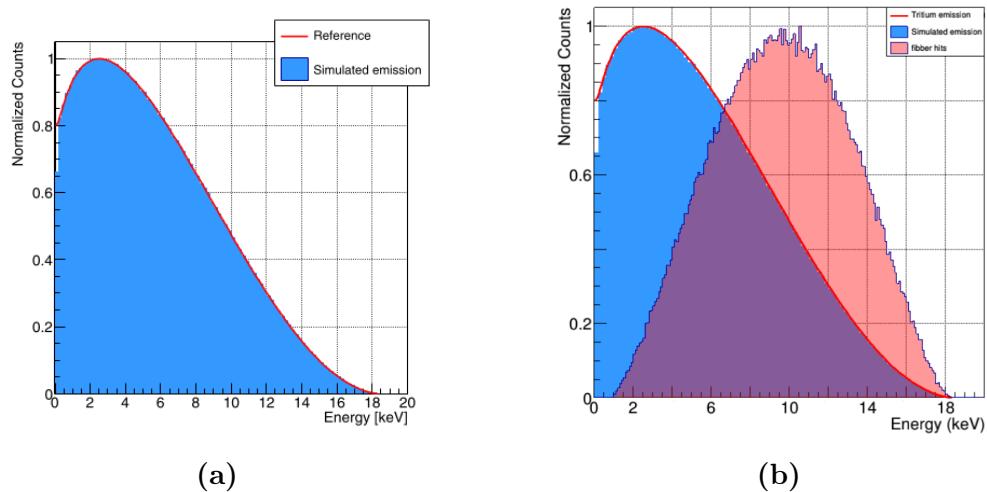
Several simulations were performed during the design of the TRITIUM detector, which were used to quantify the influence of some modifications and to choose those that optimize the tritium detection. The characteristics studied are the diameter and length of the scintillating fibers. As the tritium electrons have a very low mean free path, the shape of the simulated tritium source was also studied to reduce the consumed computing resources.

### 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 is only around  $5 \mu\text{m}$ , so most electrons do not reach the scintillating fibers. These electrons do not provide useful information and only consume computing resources. To optimize the simulation, the dimensions of the simulated tritium source were tested to minimizes the number of tritium events that do not reach the scintillating fibers.

Before that, the initial energy distribution of the simulated tritium events was checked, shown in figure 6.1a, and compared with the input taken from [Mer15], obtaining a good agreement between both. In addition, the distribution of the initial energy of tritium electrons capable of penetrating a fiber and depositing energy is compared to the initial energy distribution of all simulated tritium events, Figure 6.1b. A shift to high energies is observed, creating a peak centred at 10 keV. This shift occurs because the lower energy tritium electrons do not have enough energy to reach and penetrate the fiber and are not detected.

Regarding the optimization of the tritium source shape, 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, were simulated to assess the tritium source. The dimensions of the fiber are not important in this study since only the energy deposition of tritium electrons in the fiber were simulated, excluding optical processes. The objective of this simulation is to find the radial thick of the simulated tritium source at which non significant amount of tritium decay electrons are generated. In Figure 6.2a, a transversal cut of the 2 mm scintillating fiber, the simulated tritium source 0.5 mm thick around the fiber, and the position where the tritium decays happen, the electrons of which has deposited their energy in the scintillating fiber are shown. Furthermore, the distribution of the radial distance between the

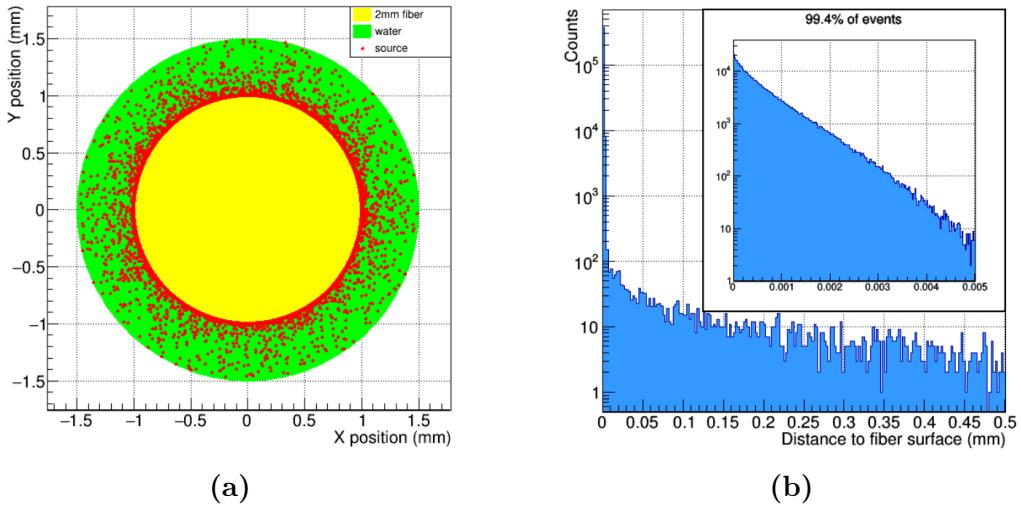


**Figure 6.1** – Energy distribution of a) simulated tritium decays b) Initial energy of tritium decays that reach the scintillating fibers (red histogram) compared the all simulated tritium events (blue histogram) [Aze20].

position where tritium decays take place and the surface of the scintillating fiber is shown in figure 6.2b. As can be seen in the Figure 6.2, 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 6.2b 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 most at this distance.

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

The scintillation yield provided by the manufacturer, 8000 phot./MeV, is only valid for minimum ionizing particles (MIP). As tritium electron energies do not correspond to MIP particles, the output light generated by the scintillating fibers was studied. For this task, the energy deposition



**Figure 6.2** – 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].

of tritium electrons in scintillating fibers and their subsequent emission of scintillation photons was included in to the simulation.

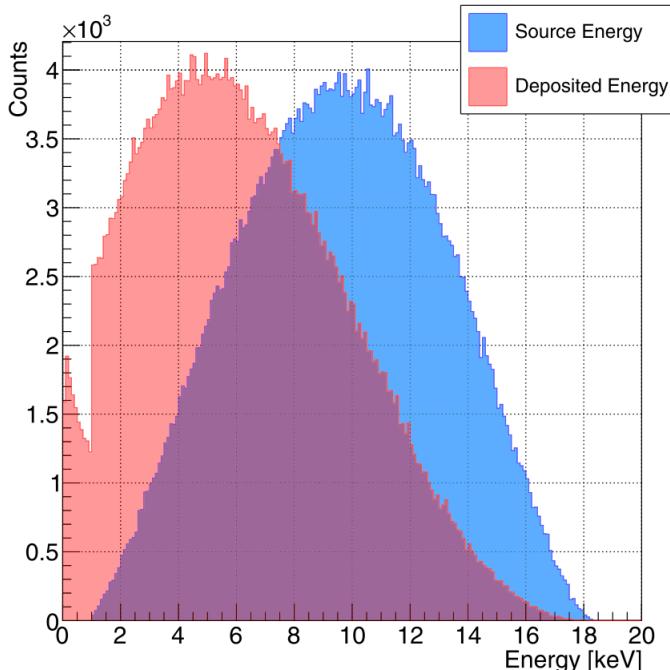
When particles that are not MIP are detected in plastic scintillators, a light quenching effect affects the proportionality between the output light per unit of path length,  $\frac{dL}{dx}$ , and the energy deposited per unit of path length,  $\frac{dE}{dx}$ , through the so-called Birk's coefficient[Bir51].

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

where  $S$  is the scintillation yield, provided by the manufacturer. The value of  $k_B = 0.126 \text{ mm/MeV}$ , typically used for scintillators based on polystyrene [Lev11], was taken for the Birk's coefficient. The objective of this section is to quantify the significance of this effect and how it affects to the tritium

detection.

A verification test of the energy deposition of tritium electron on scintillating fibers was carried out. In Figure 6.3 the initial energy of simulated tritium electrons that reach the scintillating fibers is compared to the energy deposited in scintillating fibers, red histogram.

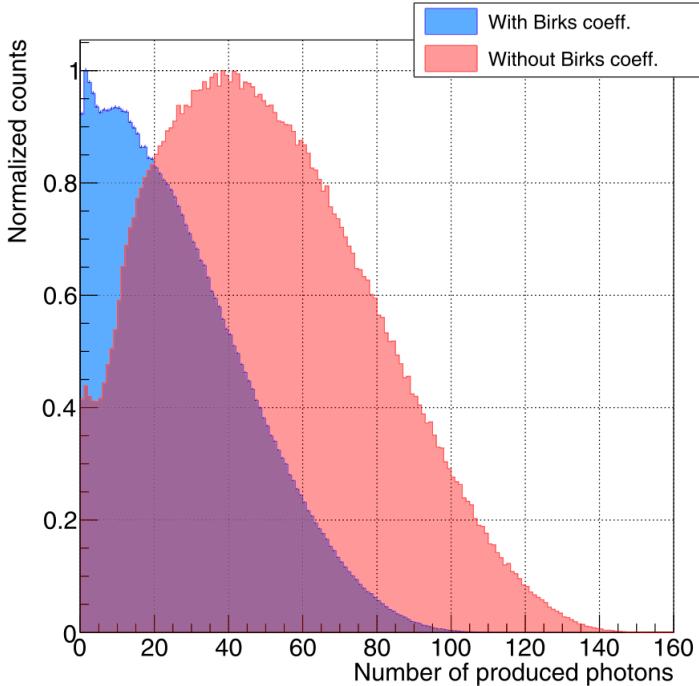


**Figure 6.3** – Distribution of the initial energy of tritium events that reach the scintillating fibers, blue histogram, and the energy deposited, red histogram [Aze20].

A shift to lower energies is observed, which is caused by the loss of energy of tritium electrons in water. A cut of around 1 keV is observed in both energy distributions, produced by the default energy threshold of 990 eV in the G4EmLivermorePhysics physics list.

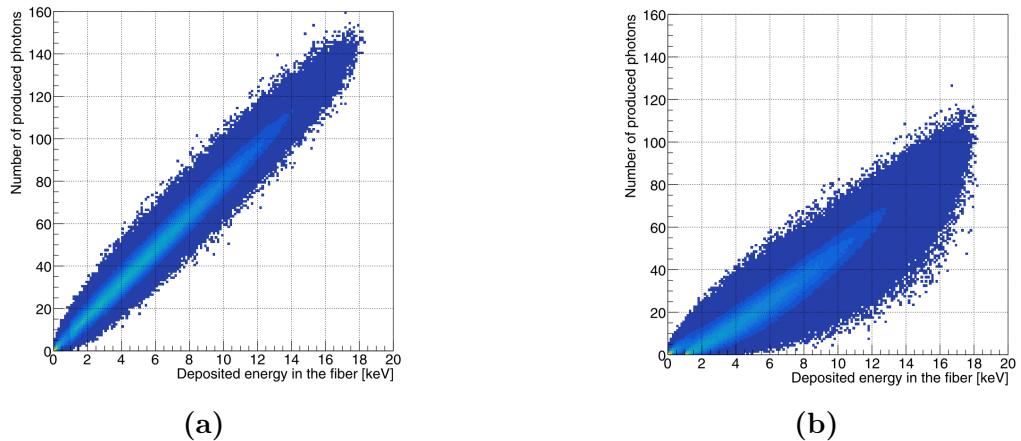
Figure 6.4 shows two distributions of number of photons produced in scintillating fibers by tritium events, one in which the quenching effect

is not considered ( $k_B = 0$ ) and the other in which the Birks coefficient is applied ( $k_B = 0.126 \text{ mm/MeV}$ ).



**Figure 6.4** – Energy distribution of photons produced in the scintillating fiber, Birks coefficient without (red histogram) with (blue histogram)[Aze20].

A distribution with a peak of around 40 photons per tritium event and a maximum of around 150 photons is obtained when the quenching effect is not considered. A significant reduction of the output light is observed when the Birks coefficient is taken into account, producing a distribution peaked at around 10 photons and a maximum of 110 photons. The quenching effect is also observable in Figure 6.5, 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 the number of photons produced per unit of energy deposited, a broader distribution is obtained when the Birks coefficient is considered, indicating an increase of



**Figure 6.5** – 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].

the fluctuations of energy deposition.

### 6.2.3 Fiber Length Optimization

A test was performed to find the fiber length that optimizes the tritium detection efficiency. Two different lengths of scintillating fibers were considered in this study, 1 m and 20 cm and two different tritium source activity were used, 0.5 kBq/L and 2.5 kBq/L. As detected tritium decays are proportional to the active area, 5 detectors were simulated for the case of a 0.20 m fiber length to have the same active area. As the active area of the detector is related to its tritium detection efficiency, the advantage to use long fibers is their large active areas with a small number of cells, reducing the number of photosensors and, consequently, the price of the TRITIUM monitor. However, a smaller length of scintillating fibers reduce de photon absorption produced in the fibers, increasing the tritium detection efficiency per active area.

To find the scintillating fiber length that optimize the tritium detection efficiency, the Tritium-Aveiro prototype, consisting of a similar design as the TRITIUM-IFIC 2 prototype but with 360 scintillating fibers of 2 mm diameter, was simulated. All optical properties were included in this study in which the photon propagation was included.

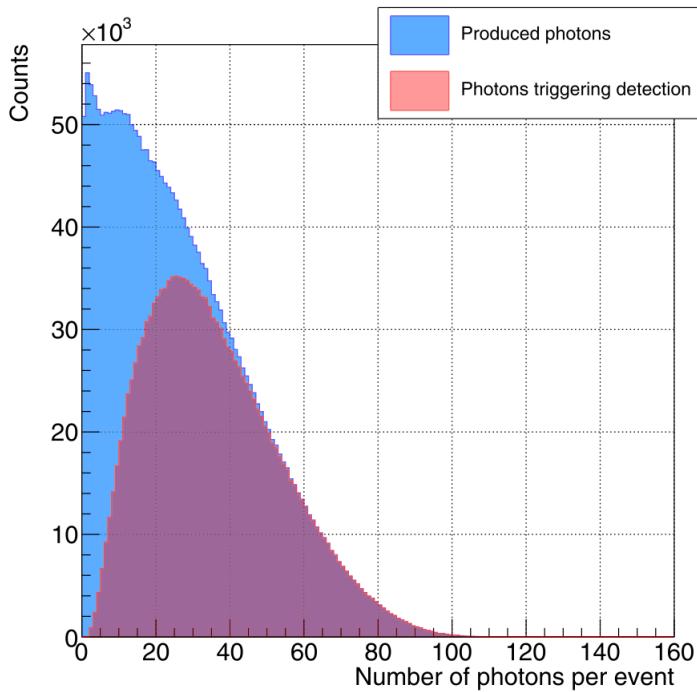
The propagation of photons in scintillating fibers was studied. The number of photons produced in a scintillating fiber per tritium electron was compared between all electrons that reach the scintillating fiber and only those the photons of which are detected in time coincidence by the photo-sensors, shown in Figure 6.6. Tritium events that produce a high number of photons are mostly detected but the events that produce few photons are seldom detected, resulting in a peak centred at around 25 photons.

The counts, integrated over 60 min and taken over a week, are shown in Figure 6.7 as a function of the time for both tritium activities and fiber lengths mentioned above. A 25% larger signal is seen for the shorter fiber length in both cases, due principally to the lower absorption of photons in shorter scintillating fibers and the leakage of some photons due to partial photon collection in the fiber. In addition, non simulation effects like the dirty or mechanical imperfections of scintillating fibers increase accentuate this effect.

#### 6.2.4 Fiber Diameter Effect

A test was carried out to study the influence of the fiber diameter in the tritium measurement. To do so, a simulation, for a single scintillating fiber 20 cm length and two different diameters, 1 mm and 2 mm (the commercial options given by Saint-Gobain), were compared.

An important point is how the fiber diameter affect to cosmic ray detection, which is an important component of the background of the



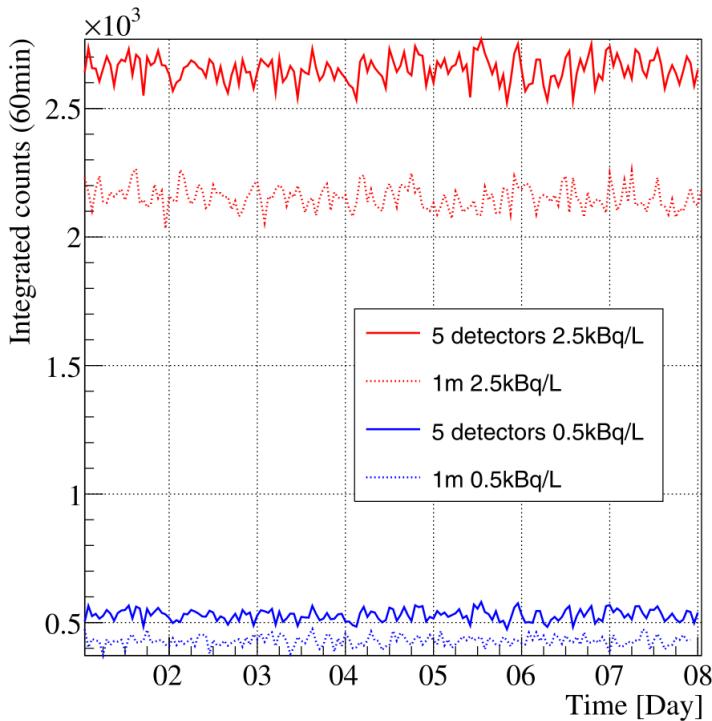
**Figure 6.6** – Number of photons produced in the fiber per tritium event for all tritium events that reach the fiber (blue histogram) and only for tritium events the photons of which are detected by photosensors (red histogram) [Aze20].

TRITIUM monitor. The energy deposited in the scintillating fiber by a cosmic ray is proportional to the active volume crossed, which is larger for 2 mm fibers. Therefore, the cosmic ray signal would be larger for 2 mm diameter fibers. The objective of this study is to find the design with a lower background in the energy region of tritium detection, ROI (up to 18 keV).

For this test, the tritiated water source was replaced by a cosmic ray source, generated by the CRY library<sup>1</sup> [(NA], [Hag07]. The CRY library is a package based on object-oriented technology and implemented in the

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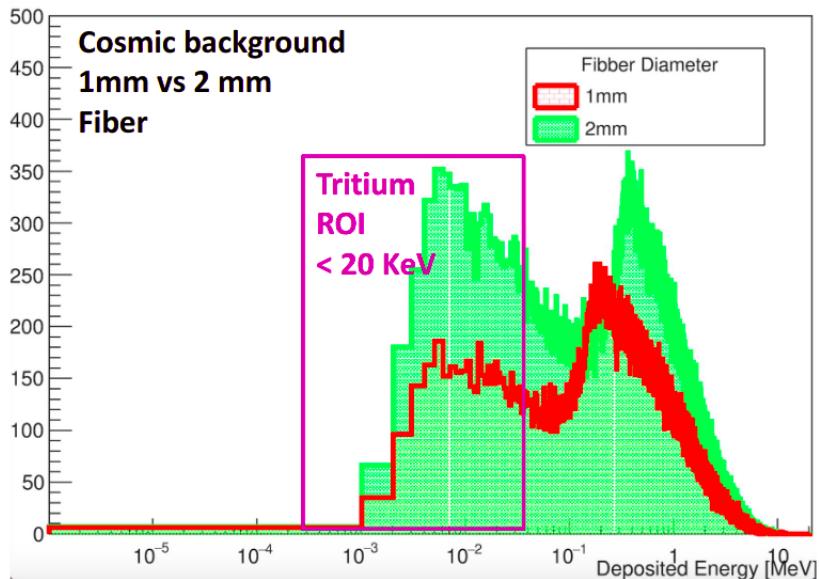
<sup>1</sup>CRY library, Cosmic-Ray Shower library



**Figure 6.7** – Simulations of 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].

C++ programming language. This library is used to generate cosmic-ray shower distributions for different particles (muons, neutrons, protons, electrons, photons and pions). The cosmic source shape used in this simulation is a horizontal square of  $1 \times 1 \text{ m}^2$  located at a height of 35 cm (above the detector) with the typical distribution of cosmic particles at sea level. The distribution of energy deposited in scintillating fibers by cosmic rays is shown in Figure 6.8 for 1 mm and 2 mm fibers.

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



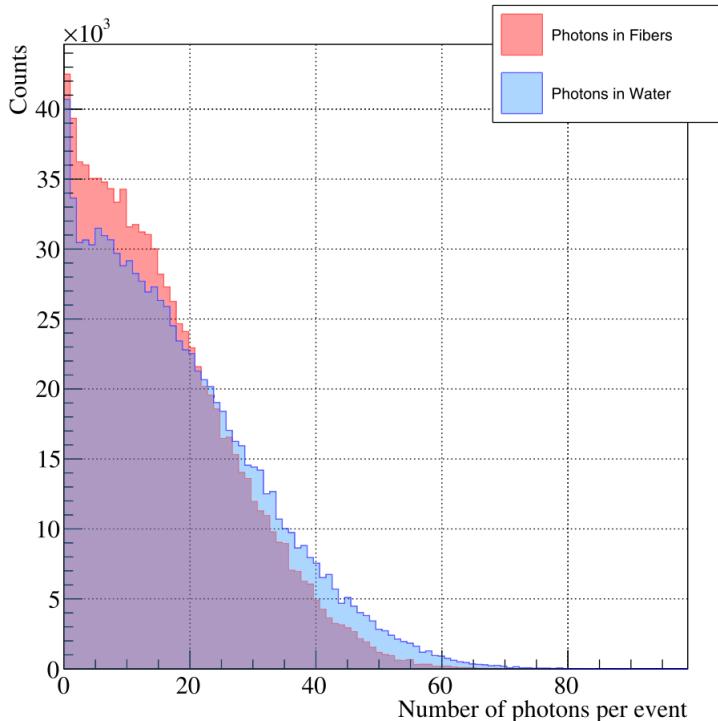
**Figure 6.8** – Comparison of the energy deposition by cosmic rays in scintillating fibers of 1 mm and 2 mm diameter.

us their greater rigidity and better flow of water through them. Thus, a experimental test is needed to choose the best design.

### 6.2.5 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 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. To quantify the importance of the latter contribution, the TRITIUM-Aveiro prototype was simulated. The distribution of the number of photons that reach the PMMA per tritium event is shown in Figure 6.9. Fiber-guided photons are shown in a red distribution, while those traveling in the water

medium are plotted 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. Therefore, PMMA windows improve the tritium detection efficiency by a factor 2.



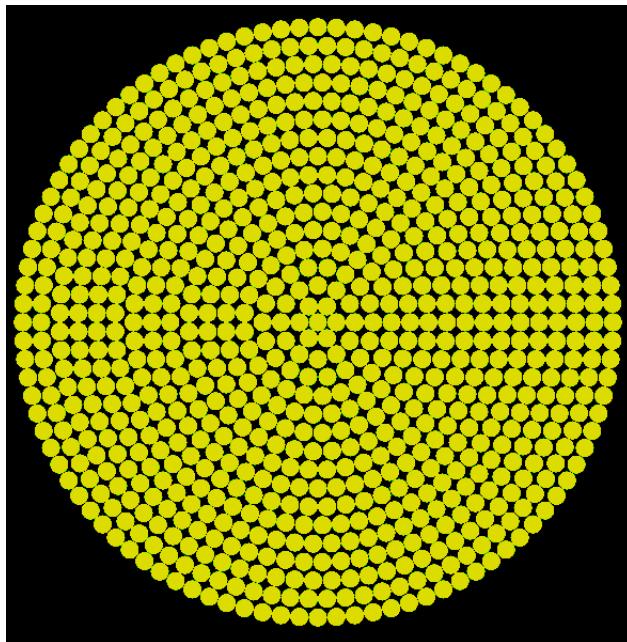
**Figure 6.9** – Distribution of photons reaching PMMA windows. The red histogram corresponds to the photons guided by fibers and the blue histogram to photons traveling in the water [Aze20].

### 6.3 Simulations of the TRITIUM Monitor

This section shows the simulations of the TRITIUM monitor carried out, consisting of various TRITIUM-IFIC 2 prototypes in parallel and a the background rejection system.

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

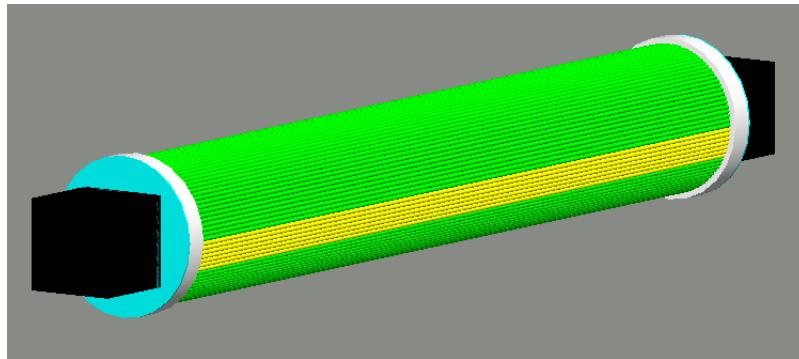
The Tritium-IFIC 2 prototype simulation was the last simulation carried out in TRITIUM experiment. It consists of 800 fibers of 1 mm diameter uniformly distributed in sixteen different circles of increasing radius, as illustrated in Figure 6.10. The optical properties were included.



**Figure 6.10** – Distribution of the scintillating fibers in the simulation of Tritium-IFIC 2 prototype.

The tritiated water source consists of a tritiated water volume with a thickness of  $5 \mu\text{m}$  around each scintillating fiber. Scintillating fibers are located inside of a Teflon vessel, which was simulated with dimensions given above. Two PMMA windows of 5 mm thickness located in both fiber ends and a optical grease layer with a thickness of 0.5 mm located in each PMMA windows were included. Two PMTs, model R8520-460 from Hamamatsu [K.K19], were also simulated as photosensors.

The geometry simulated for TRITIUM-IFIC 2 is shown in Figure 6.11 in which is shown the PMTs (black), the optical grease (blue), PMMA windows (white), tritiated water (green) and scintillating fibers (yellow). In this image, the Teflon container is not drawn to allow its interior to be seen. Several volumes of tritiated water were also excluded to allow several scintillation fibers to be seen.

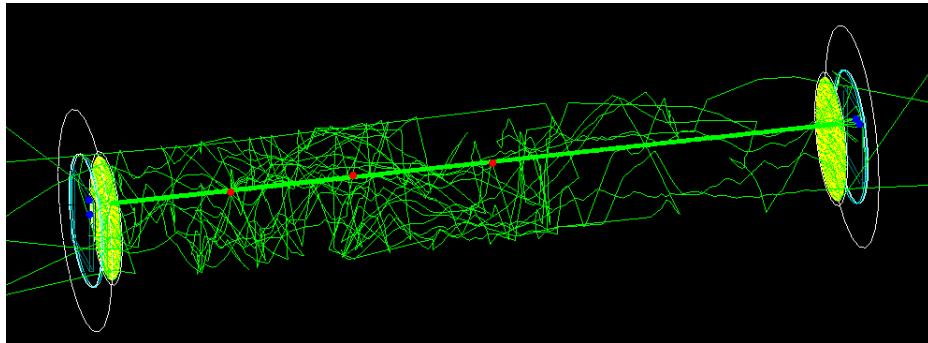


**Figure 6.11** – Simualtion of Tritium-IFIC 2 prototype. PMTs (black), the optical grease (blue), PMMA windows (white), tritiated water (green) and scintillating fibers (yellow).

The used PMTs do not cover the entire active area formed by the scintillating fiber bundle. This is not a problem for the TRITIUM detector since its final version will use SiPM arrays.

The objective of these simulations are to find the Low Detection Limit, LDL, for tritiated water, which is an important parameter of the prototype, and study the activity resolution of the prototype and how both parameters, activity resolution and LDL, can be improved through various parameters such as the increase of the integration time windows of the measurement and the number of prototypes read in parallel. The detection of a tritium event in the TRITIUM-IFIC 2 prototype is shown in Figure 6.12. The path of the photons created in scintillating fibers are represented by green lines which end in red dots when they are absorbed in the fiber or the water and blue dots when they are absorbed in the PMTs (detected).

The fiber that has detected the tritium electron is clearly identified. Some photons go out of the fiber and are not collected. Blue dots in both PMTs indicate that photons are detected on time coincidence.

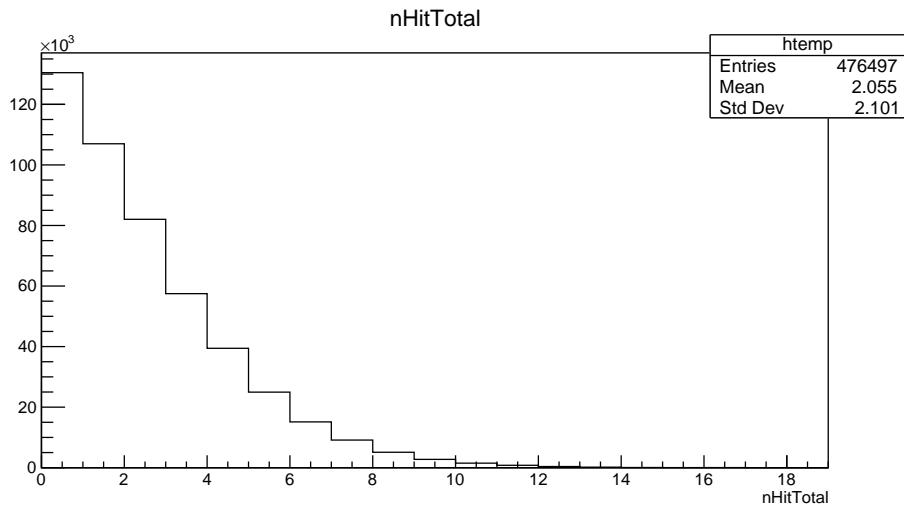


**Figure 6.12** – Tritium electron detected in the simulated TRITIUM-IFIC 2 prototype. The path of the optical photons is represented by green lines and the position in which they are absorbed is represented by red and blue dots (absorbed in water or PMT, respectively).

Several variables were used as tests to verify the different steps of the simulation such as the production of tritium electrons, 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 photosensors per tritium event for the simulated TRITIUM-IFIC 2 prototype is shown in Figure 6.13.

A maximum of 17 photons is obtained for the TRITIUM-IFIC 2 prototype simulations, which is in agreement with the maximum of 15 photons experimentaly measured, shown in Figure 7.9. This confirms that the value used in the simulations for the Birks coefficient,  $k_B = 0.136 \text{ mm/MeV}$ , is quite accurate. The experimental distribution are lower than the simulations between 3 and 8 photons, probably due to imperfections of the prototype which are not included in the simulation.

Activities from 100 Bq/L to 5 kBq/L for three months of simulated data taking and an integration counting time of 10 min were simulated. The

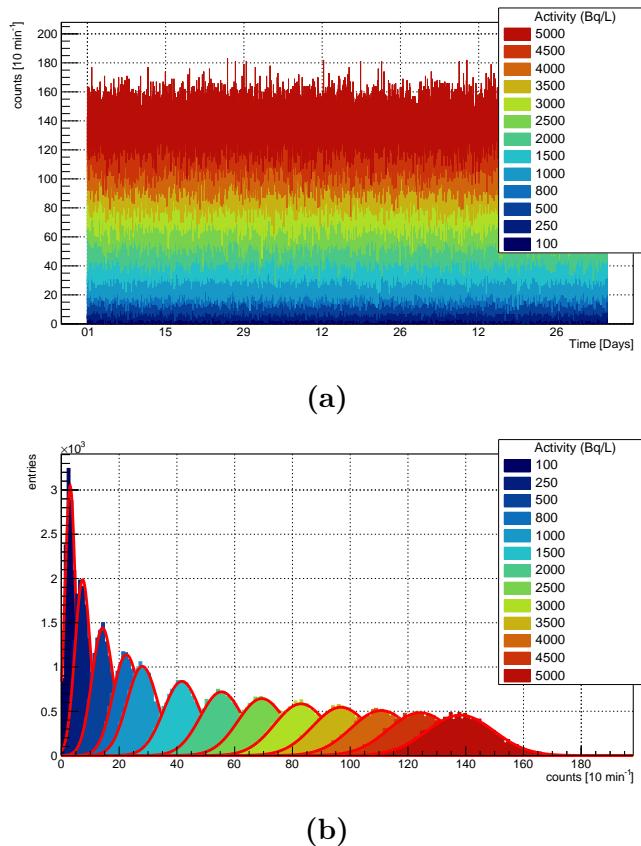


**Figure 6.13** – Photons detected by both PMTs per tritium event in the simulated TRITIUM-IFIC 2 prototype.

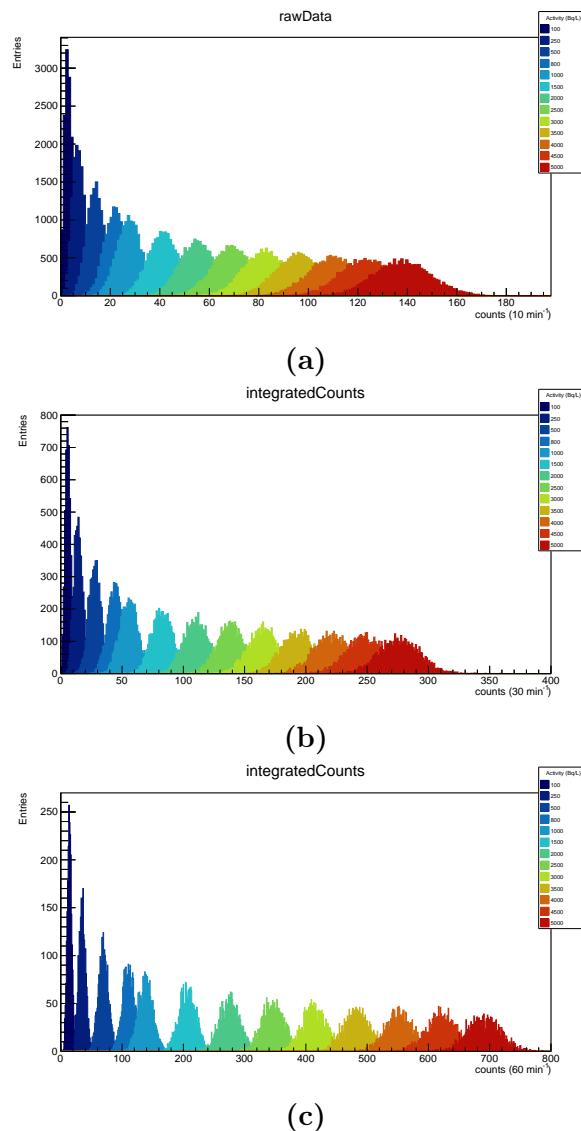
simulation results are presented in Figure 6.14. A difference of 250 Bq/L is not distinguished due to the overlapping of distributions. To reduce the width of the distribution obtained for each activity, the statistics must be increased, which can be done in two different ways, either by increasing the integration counting time or by increasing the number of prototypes read in parallel.

To check the effect of increasing the integration counting time, distributions for increasing integration counting times of 10 min, 30 min and 60 min were generated. They are shown in Figure 6.15.

The effect of increasing the integration counting time is clearly visible in this figure, reducing the 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 counting time of 60 min, which could still be considered as a quasi-real time measurement. Similarly, these distributions are shown in Figure 6.16 for 10 min of integration counting time, for 1, 5 and 10 number of prototypes in paral-



**Figure 6.14** – 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.



**Figure 6.15 –** Distribution of the tritium counts simulated for TRITIUM-IFIC 2 prototype for three different integration time: a) 10 min, b) 30 min and c) 60 min.

lel. 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 resolution, defined as

$$\text{Resolution}(\%) = \frac{\text{FWHM}}{\text{centroid}} \cdot 100 \quad (6.2)$$

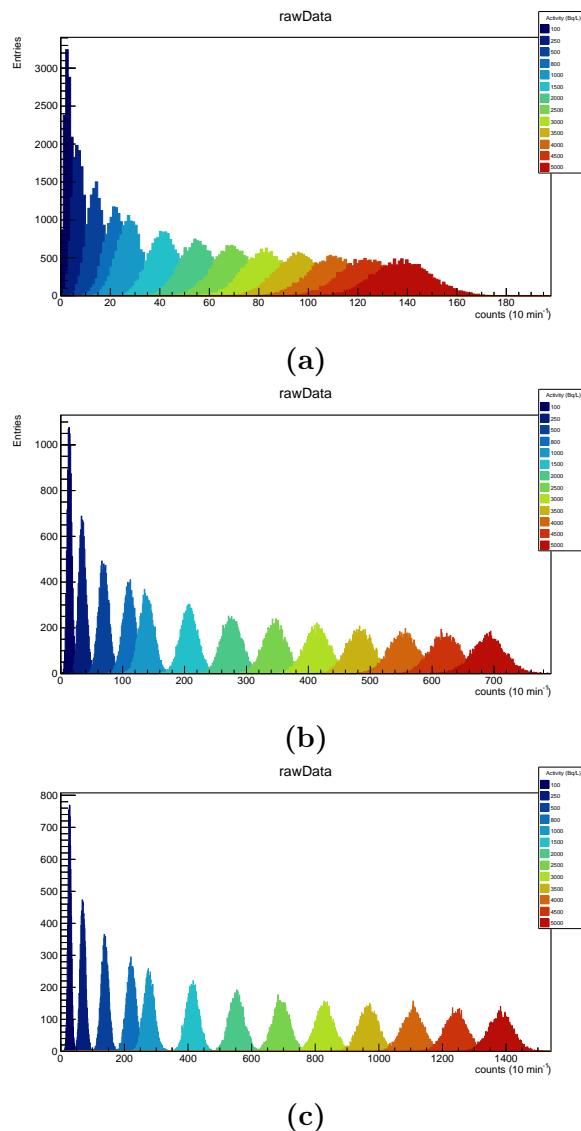
is plotted in Figure 6.17.

It can be observed that the resolution improves with integration time and number of prototypes. Therefore, both parameters must be balanced based on the requirements and funding of the experiment. The activity difference, the distribution peaks are clearly separated for the different integration counting times and number of detectors. The studied cases are summarized in Table 6.1.

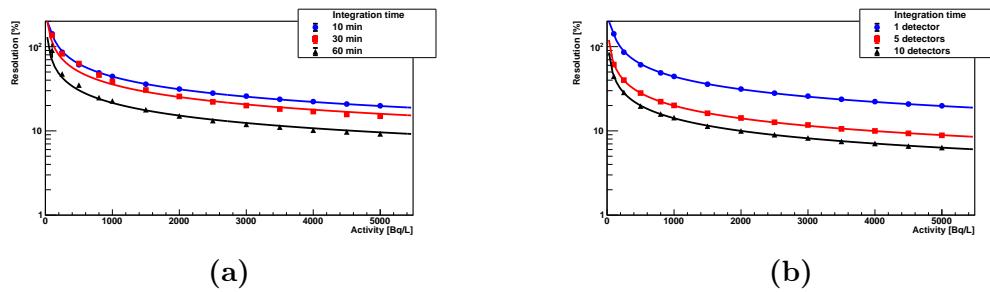
# of Detectors	10 min	30 min	60 min
1	< 1000 Bq/L	500 Bq/L	200 Bq/L
5	200 Bq/L	150 Bq/L	100 Bq/L
10	150 Bq/L	100 Bq/L	$\approx 50$ Bq/L

Table 6.1: Difference in activity that can be resolved for the TRITIUM-IFIC 2 prototype, for different integration times and different number of prototypes.

The decision made in the TRITIUM collaboration is to install 3 different TRITIUM-IFIC 2 prototypes, with which differences of 250 Bq/L are expected to be resolved with an integration time of 30 min. These prototypes are expected to be installed in Arrocampo dam as soon as possible. Two other TRITIUM-Aveiro prototypes are being built and will be installed soon, along the one currently installed.



**Figure 6.16** – Distribution of the tritium counts simulated for different number of TRITIUM-IFIC 2 prototypes: a) 1, b) 5 and c) 10, for an integration time of 10 min.



**Figure 6.17** – Resolution of the TRITIUM-IFIC 2 prototype as a function of the a) integration counting time b) number of prototypes.

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

The lead shielding and active vetos, described above, were included in the simulation of the Tritium-IFIC 2 prototype. The objective of these simulations was to quantify the reduction of cosmic background detected by the prototype. For this task, the tritium source was replaced by the cosmic events source, which was simulated through the CRY library.

The optical properties included to the plastic scintillators of the active veto 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]. Two PMTs, model R8520-460 from Hamamatsu, were simulated to read each plastic scintillator, similar to that presented in section 3.4.2.

The lead shielding was simulated with properties taken from the Geant4 NIST database. The dimensions of the simulated lead shielding were  $60 \times 60 \times 70 \text{ cm}^3$ , which is the minimum needed to accomodate 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.

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 quantify the tritium detection improvement due to 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 rays 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 by the stop of the soft cosmic radiation (energy lower than 200 MeV). It has to be taken into account that the natural backgrounds of the installation site is 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. Around 10% of the cosmic events that penetrate the lead shield and reach the TRITIUM IFIC 2 prototype, which are the hard cosmic rays, are detected by the cosmic veto and, therefore, removed from the tritium background.

Therefore, the usefulness of both parts of the background rejection system, lead shield and active veto, has been demonstrated by the simulations.

# 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 in the laboratory, where external atmospheric conditions, such as temperature, can be better controlled. The second section shows the measurements obtained at the Arrocampo dam, the TRITIUM monitor installation site, where the control of external atmospheric conditions is less accurate.

### 7.1 Results from Laboratory Measurements

This section reports on the experimental results obtained with the different prototypes developed in the TRITIUM experiment in the laboratory. The

facilities used for this task were the IFIC Laboratory, 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 IFIC laboratory.

As stated above, a statistically significant number of events was not obtained when the prototype was read by two PMTs in coincidence. To overcome this problem, a single PMT measurement was taken using the electronic chain configuration shown in figure 3.15a. The energy spectra were measured for both, the signal and background prototypes, which are shown in Figure 7.1a. As reported above, the signal prototype was filled with a tritiated water solution of 99.696 kBq/L activity and the background prototype was filled with ultrapure water. A difference between signal and background, Figure 7.1b, corresponds to the energy spectrum of tritium. The number of counts per second obtained for the three spectra is given in Table 7.1, where the tritium counts are obtained from the difference of signal and backgorund.

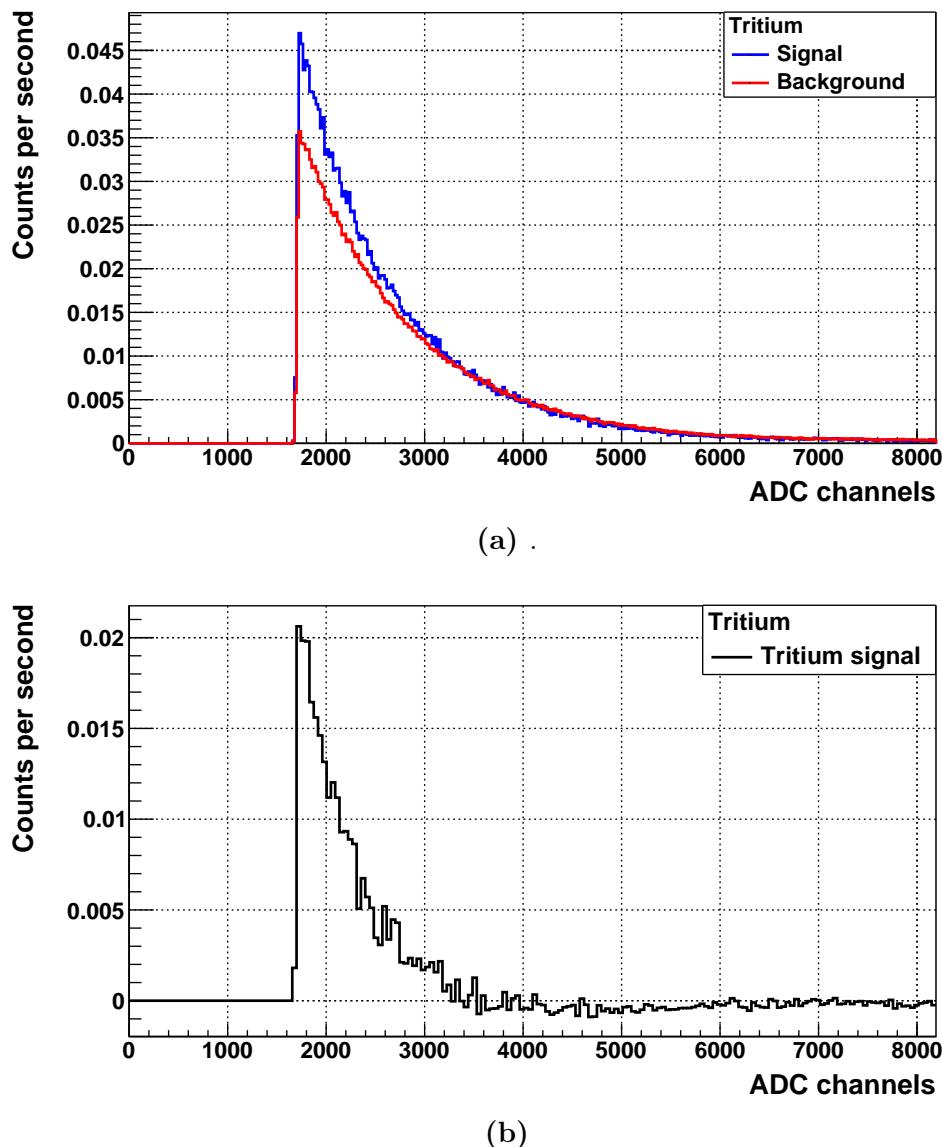
Spectrum	Counts/second
Signal prototype	$2.27 \pm 0.06$
Background prototype	$2.06 \pm 0.06$
Tritium counts	$0.21 \pm 0.085$

Table 7.1: Counting rate obtained with the TRITIUM-IFIC 0 prototype.

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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 (Environmental Radioactivity Laboratory of the University of Extremadura)



**Figure 7.1** – Energy spectra measured with TRITIUM-IFIC 0 prototype. Above) Signal and background energy spectra. Below) Tritium energy spectrum.

The tritium detection efficiency obtained for this prototype is  $(2.11 \pm 0.85) \cdot 10^{-3} \text{ Ls}^{-1}\text{kBq}^{-1}$ , calculated from the ratio of both, the tritium counting rate measured and the specific activity of the tritium liquid source.

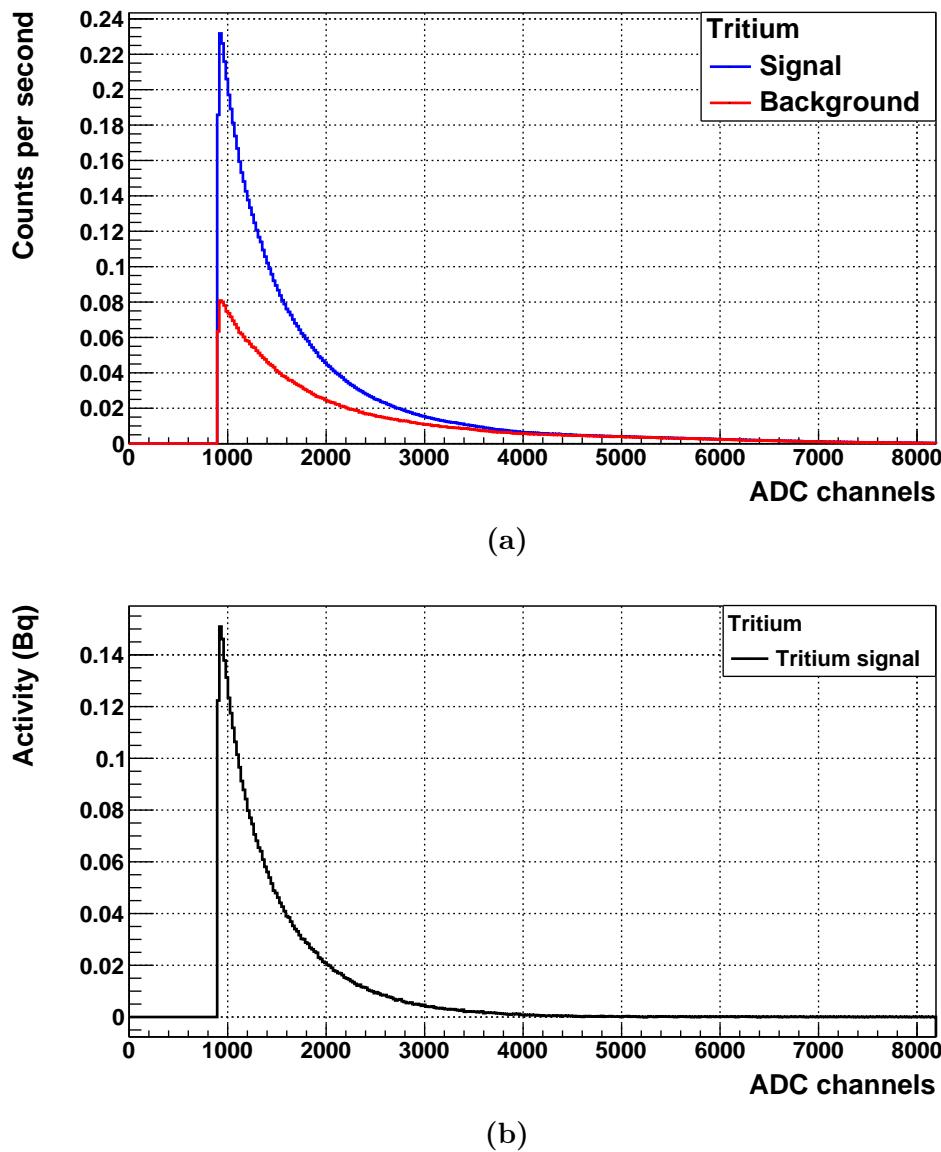
As we reported in section 2.1, the efficiency of scintillating detectors scales with the active area of the scintillator used. Therefore, to compare the efficiency with other detectors and with other prototypes developed in TRITIUM experiment, the specific efficiency of this prototype is calculated by normalizing to the scintillator area, which is  $(9.59 \pm 3.88) \cdot 10^{-6} \text{ Ls}^{-1}\text{kBq}^{-1}\text{cm}^{-2}$ . As can be seen in Table 2.2, the specific efficiency is a somewhat larger than that obtained for Muramatsu and Moghissi [Mog69], which is a low efficiency. This fact can be explained by the loss of photons in the curve of the fiber bunch, as demonstrated in section above.

### 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 IFIC laboratory. Several improvements, such as a Teflon vessel and new straight arrangement of scintillating fibers, was included in the design of this prototype, which were found to be problematic for the previous prototype, reducing its efficiency.

The signal and background energy spectra are shown in Figure 7.2a. The difference between both energy spectra corresponds to the tritium energy spectrum, Figure 7.2b. Furthermore, the tritium detection efficiency of this prototype was improved compared to TRITIUM-IFIC 0 prototype. This improvement can be quantified as in the previous section. The rates measured are given in Table 7.2, where the tritium counts are obtained from the difference of signal and background spectra.

The tritium detection efficiency obtained for TRITIUM-IFIC 1



**Figure 7.2** – Energy spectra measured with TRITIUM-IFIC 1 prototype.  
a) Signal and background energy spectra. b) Tritium energy spectrum.

Spectrum	Counts/second
Signal prototype	$7.82 \pm 0.11$
Background prototype	$3.99 \pm 0.08$
Tritium counts	$3.83 \pm 0.13$

Table 7.2: Counting rate obtained with the TRITIUM-IFIC 1 prototype.

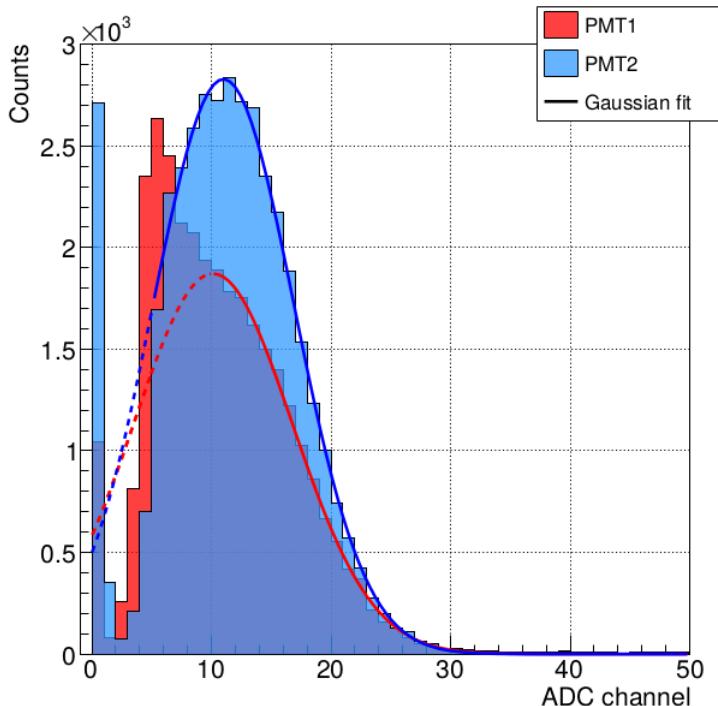
is  $(3.84 \pm 0.16) \cdot 10^{-2} \text{ Ls}^{-1}\text{kBq}^{-1}$ . The efficiency obtained for this prototype is larger than that of TRITIUM-IFIC 0, as expected since this prototype has a larger active area. The specific efficiency obtained is  $(9.56 \pm 0.40) \cdot 10^{-5} \text{ Ls}^{-1}\text{kBq}^{-1}\text{cm}^{-2}$ , which is a factor of ten better than that of TRITIUM-IFIC 0. Furthermore, compared to scintillating detectors developed in other experiments, table 2.2, the efficiency of this prototype is very close to the best result, obtained by Singh, and the specific efficiency, which is the most relevant parameter to compare, is almost 5 times larger than that obtained by Hofstetter [Hof92b, Hof].

### 7.1.3 Experimental Results of TRITIUM-Aveiro prototype

This section reports the results obtained with the TRITIUM-Aveiro prototype during its installation in DRIM and LARUEX laboratories. This prototype was first installed in the University of Aveiro, where the first measurements were taken.

First, the energy distribution of a single photon of the PMT dark current was measured. 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 signal of the PMTs were digitalized, shaped and pulse-height measured by a CAEN V1724 digitalizer [CAEb]. The single-photon

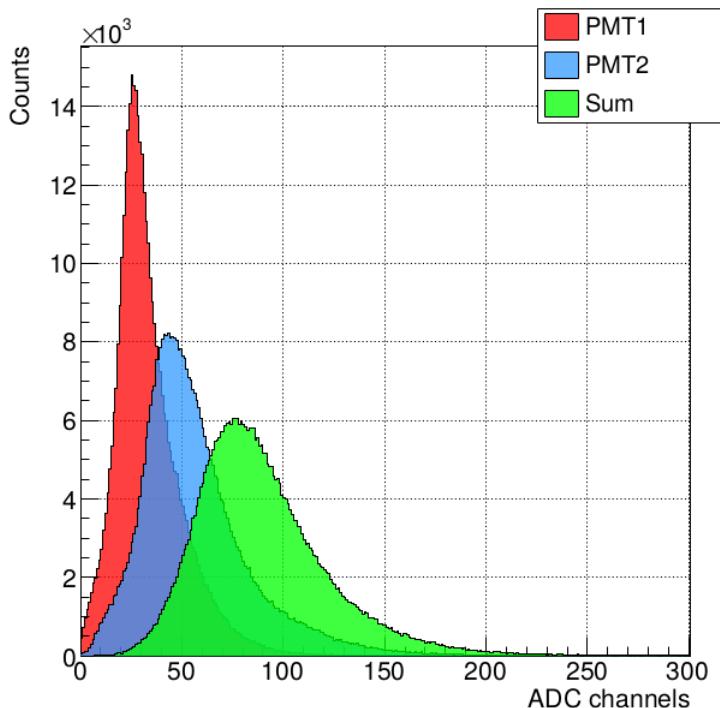
energy distribution of both PMTs is shown in Figure 7.3 in which a fit to a Gaussian function was fitted. Due to the electrical noise of the PMT, an extrapolation (dashed line) was needed to be applied.



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

The distribution obtained with PMT1 deviates from the Gaussian function due to the higher noise in the low energy channels. As DRIM laboratory is not allowed 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. The TRITIUM-Aveiro prototype was coupled to both PMTs using optical grease and, due to its short mean free path in solid materials, the radioactive source was placed inside the Teflon vessel. The prototype was not filled with water for this measurement. The spectra obtained are shown in Figure 7.4. A shift to higher energies is observed

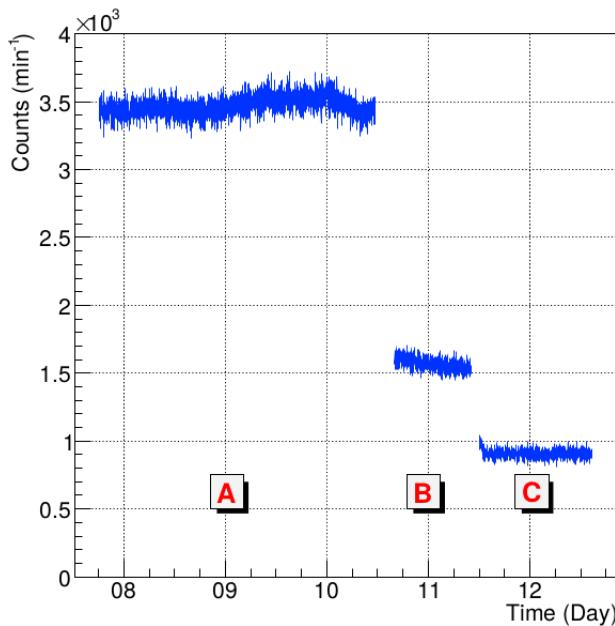
in the PMT2 data due to its higher gain and to the closer distance to the radioactive source.



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

A measurement with a passive shield was also performed in the DRIM laboratory to quantify the attenuation of the background by lead. The  $^{55}\text{Fe}$  radioactive source was removed and a measurement in counting mode was carried out. This measurements, shown in Figure 7.5, were carried out in three different situations. The first, region A, was performed without shielding, the second, region B, with a lead shield of 2.5 mm thickness and the third, region C, with two lead foil layers. As can be seen, in the region A, the average rate of 2.5 days is  $3.5 \cdot 10^3$  counts/min. In the region B, a background suppression by a factor of 2 was observed, measuring an average rate of  $1.6 \cdot 10^3$  counts/min. In the region C, an average rate of  $0.9 \cdot 10^3$

counts/min was obtained, supressing the background by about a factor of 4.

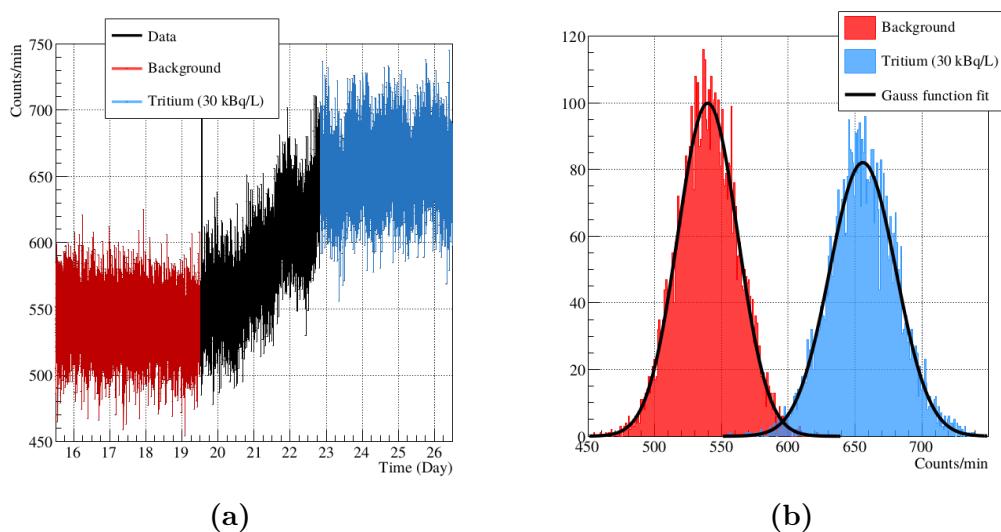


**Figure 7.5** – Measurement of the background with TRITIUM-Aveiro prototype shielded with different layers of lead, A) without shielding, B) with a lead shield of 2.5 mm thickness and C) with two lead shields of 2.5 mm thickness each one [CA].

To work with a tritiated water, the prototype was installed in the LARUEX laboratory, at the University of Extremadura.

The background of the prototype was measured during 4 days with the prototype filled with ultrapure water and covered with lead bricks of 5 cm thickness. The time of each measurement is 1 minut. The data, fitted to a Gaussian function, are shown in Figure 7.6a.

An average rate of 540 counts/min with standard deviation of 22.61 counts/min was obtained. To calculate the Minimum Detectable Activity (MDA), the detection limit concepts developed by Lloyd A. Currie [Cur68]



**Figure 7.6** – Measurements of the background and tritium liquid source (with an activity of 29.8 kBq/L) performed with the TRITIUM-Aveiro prototype and integrated during a minute [CA]. a) Counts per minut measured as a function of time. b) Distribution of the acquired data.

were applied. The minimum net counts with a probability of a false-negative less than a 5%,  $N_D$ , and minimum net current with the probability of a false-positive less than a 5%,  $L_C$ , called critical level, were calculated by the equations,

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

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

where  $\sigma_{Nb}$  is the standard deviation of the average rate of the background measurements.  $L_C$  and  $N_D$  refer to the net rate after background subtraction. Therefore,  $L'_C$  and  $N'_D$ , referred to the detector signal (before background subtraction), are 593 and 648 counts/min respectively.

To find the MDA, tritiated water was slowly added so that the tritium water activity increased continuously up to reach the  $N'_D$  value. An

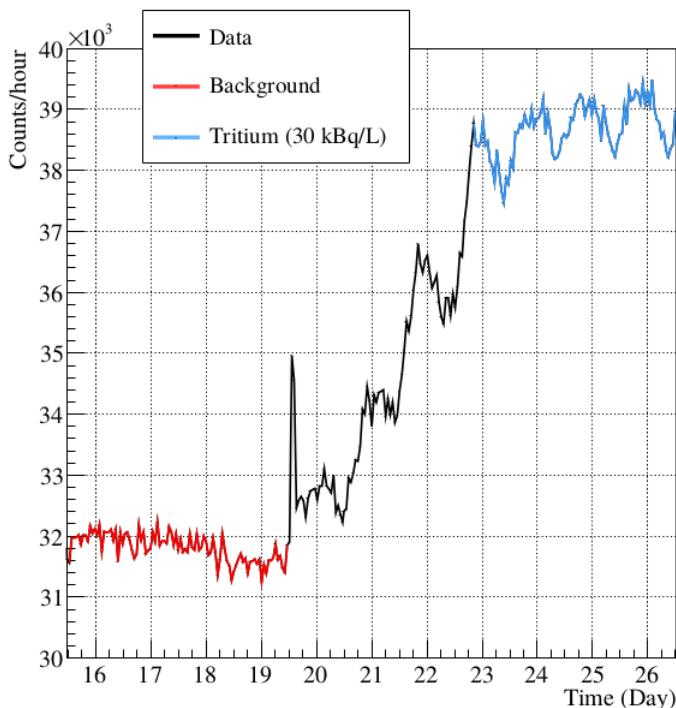
average of  $656 \pm 0.43$  counts/min was obtained, the activity of which was MDA=29.8 kBq/L, obtained with a Quantulus system.

The tritium detection efficiency was calculated from the ratio of the net tritium rate measured,  $1.93 \pm 0.58$  counts/sec, and the activity of the tritium source used. The efficiency obtained is  $(6.49 \pm 1.94) \cdot 10^{-2}$  Ls<sup>-1</sup>kBq<sup>-1</sup>. The specific efficiency is  $(1.59 \pm 0.48) \cdot 10^{-5}$  Ls<sup>-1</sup>kBq<sup>-1</sup>cm<sup>-2</sup>. Comparing to the specific efficiency obtained with scintillating detectors, Table 2.2, the specific efficiency of the TRITIUM-Aveiro prototype is close the largest value, obtained by Hofstetter [Hof92b, Hof]. However this prototype has a lower specific efficiency than 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.

The efficiency uncertainties obtained for this prototype are larger than those obtained in the previous TRITIUM prototypes due to a shorter measurement time, 1 minute, while that used for the previous prototypes is 10 minutes. Because of that, longer measurements are studied to quantify the reduction of the MDA of this prototype. The data for an integrated time of 60 minutes is shown in Figure 7.7.

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

A daily oscillation is clearly observed in the Figure 7.7, indicating that the measurements are affected by external light. This oscillation begins on the 19<sup>th</sup> day, where the water closed circuit pump was installed, so it is likely that a light leak was introduced in the system.



**Figure 7.7** – Measurements of the background and tritium liquid source (with an activity of 29.8 kBq/L) performed with the TRITIUM-Aveiro prototype and integrated during one 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

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 these three spectra is shown in Table 7.3, where the Tritium counts are obtained from the difference of the signal and background counts.

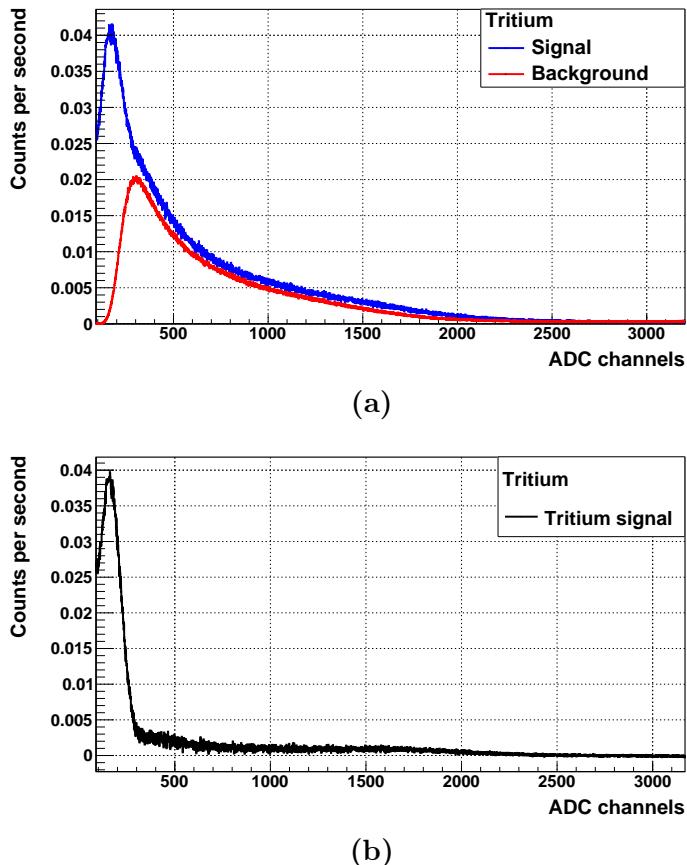
Spectrum	Counts/second
Signal prototype	$19.05 \pm 0.18$
Background prototype	$11.54 \pm 0.14$
Tritium counts	$7.11 \pm 0.23$

Table 7.3: Counts per second obtained with TRITIUM-IFIC 1 prototype.

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.



**Figure 7.8** – Energy spectra experimentally measured with TRITIUM-IFIC 2 prototype. Above) Signal and background energy spectra. Below) Tritium energy spectrum.

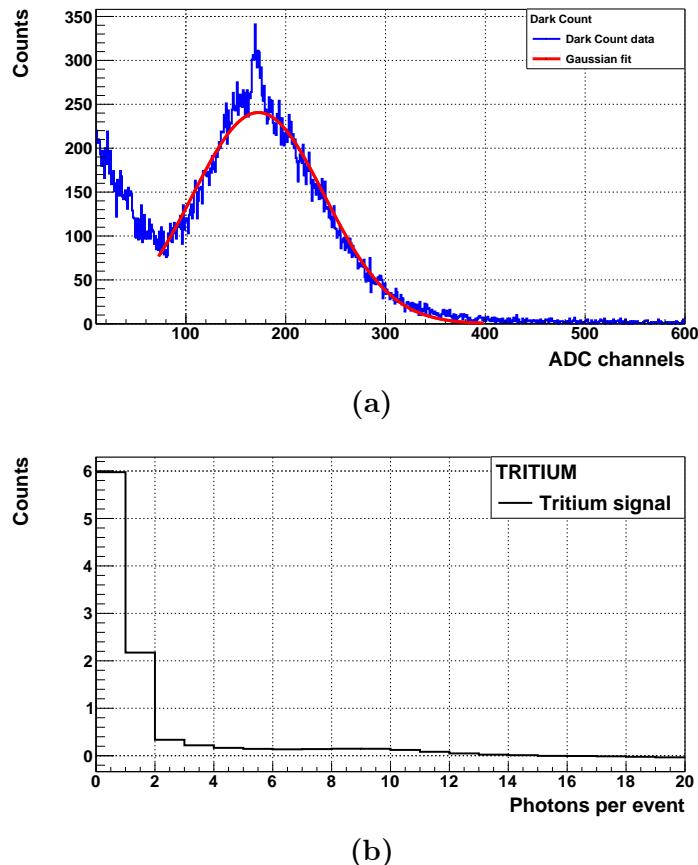
As discussed in chapter 4, the energy spectrum is shown in ADC units, proportional to energy, since it is difficult, even impossible, to do an energy calibration for a plastic scintillator due to its large uncertainty in the number of photons produced per event. Nevertheless, a detector calibration can 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 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 3.15a. 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 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.

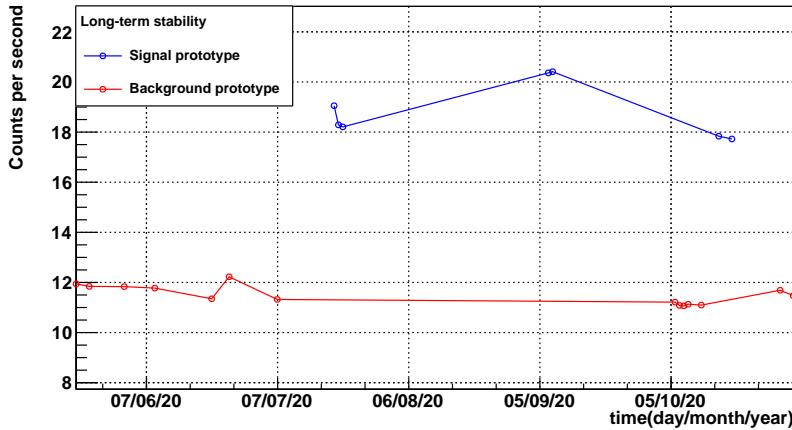
As can be seen, a maximum of 15 photons are generated per tritium event, which is in accordance with the expected result taking in to account the efficiencies involved.

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.

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. Furthermore, it can be verified that the tritium activity used, 10 kBq/L, is not the LDL of the TRITIUM-IFIC



**Figure 7.9** – Above) Single photon distribution of the TRITIUM-IFIC 2 prototype. Below) Tritium energy spectrum measured with TRITIUM-IFIC 2 prototype and expressed in photons detected per event.



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

2 prototype since both measurements, signal and background, are clearly separated along the time.

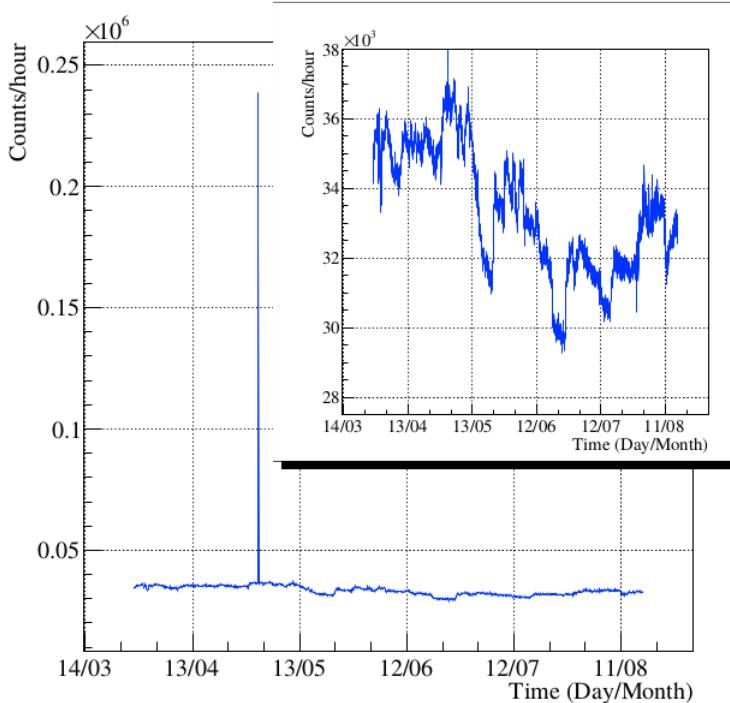
ESTUDIAR EL LDL DEL DETECTOR CON LAS MEDIDAS TOMADAS!!!

INCLUIR QUE NO SE HA CONSEGUIDO MEDIR 1kBq/L!!!

## 7.2 Results from Measurements at Arrocampo Dam

This section shows the measurements obtained with the TRITIUM-Aveiro 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 acquired during this time are shown in the figure 7.11, the measurement time of which is 60 minutes.



**Figure 7.11** – Background measured with the TRITIUM-Aveiro 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

## 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, which was characterized indendently, 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.

First, the components of the tritium monitor, which are scintillating fibers and SiPM, were characterized and some improvements were studied.

- On the one hand, a characterization of the photon collection efficiency of the BCF-12 unclad 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 cleaving, 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.

Due to the strong dependence of the SiPM internal gain on temperature, a stabilization method for the SiPM gain 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 fullfilled. 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 essencial to achieve the activity goal of  $100 \text{ Bq/L}$ .

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 lastest 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 8.1, in which the results obtained with other experiments are also included.

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	$64.87 \pm 19.41$	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 8.1: 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.

As can be seen in the table, the specific efficiency of the latest prototype, TRITIUM-IFIC 2, is almost an order of magnitud 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, 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 and apply the same mathematical method used 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 monitor 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 tri-

tiated water in quasi-real time, improving the specific efficiency and the low detection level of the activity 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 planned 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 installation of which has been delayed due to the coronavirus pandemic.

Finally several Monte Carlo simulation has been developed using Geant4. These simulations were used for three different tasks:

- First, several simulations were carried out to study the different steps of the simulation, such as 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 these 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 as the spectrum of the number of photons obtained per tritium event, ensuring that this prototype

works correctly. 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

## Electronic Readout for characterization the SiPM 13360-6075 Model

This appendix shows the electronic system designed to perform a complete characterization of the SiPM S13360-6075 model. This consists on three different PCBs<sup>1</sup>, shown in Figure A.1:

1. The first PCB, shown in Figure A.1a, 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 black box has a small hole of 1 mm diameter, prepared to introduce an optical fiber<sup>2</sup> to illuminate SiPMs with an incoherent light source. The light source utilized is a LED, model 430L from Thorlabs com-

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<sup>1</sup>PCB, Printed Circuit Board

<sup>2</sup>The optical fiber used is BCF-98 from Saint-Gobain company [Cer21]

pany [Tho18], which gives an spectrum shown in Figure A.1d. 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 palced at 436.3 with a FWHM<sup>3</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 A.1b, 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 A.1c, 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 trought a HDMI feedthrough.

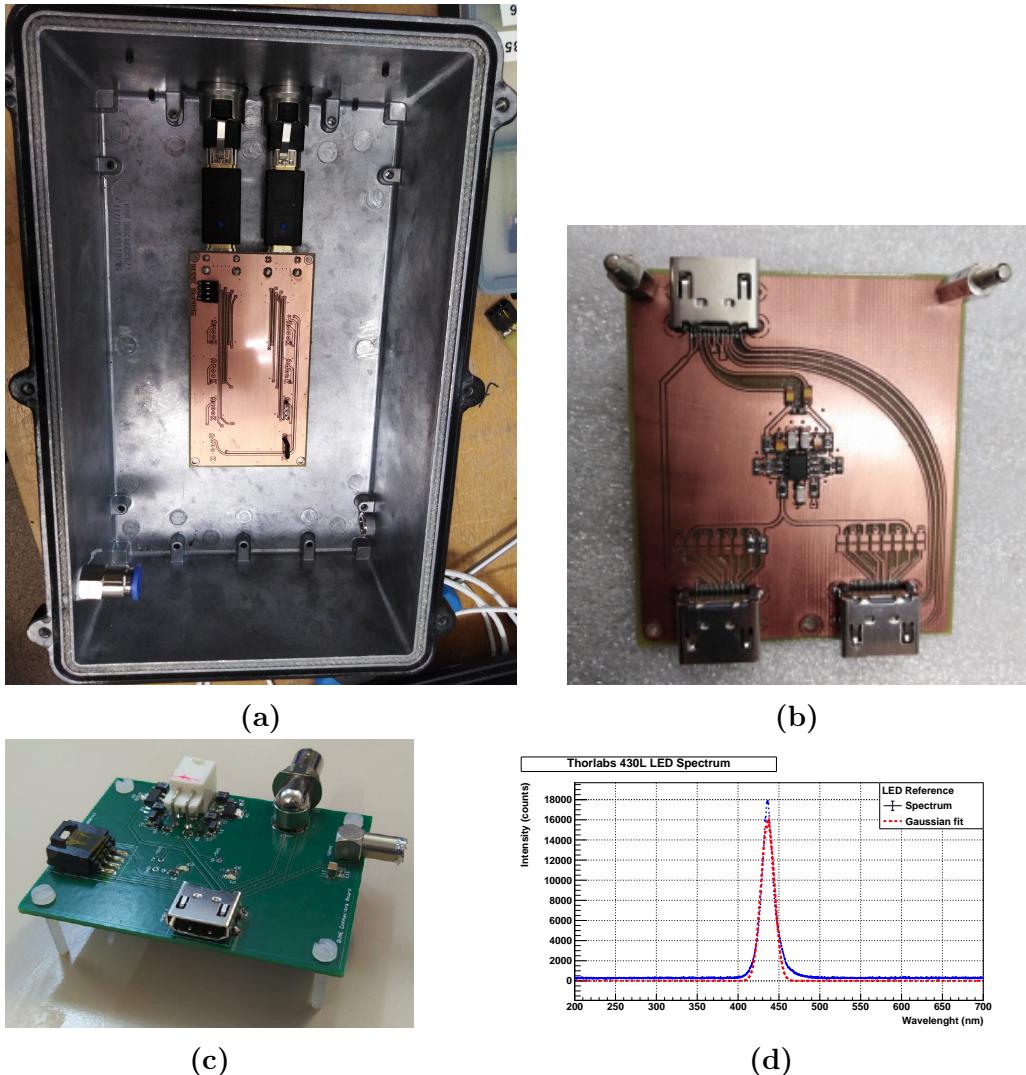
The input signals are the supply voltage of the SiPMs and the supply voltage of the PCBs ( $\pm 6$  V) and the output signals are the temperature sensor signal and the summed signal of all SiPMs.

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>4</sup>.

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<sup>3</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>4</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.



**Figure A.1** – Three PCBs used for the SiPM characterization a) The PCB 1 used to arrange 8 SiPMs and black box. b) The PCB 2 used to sum and amplify the output signals of SiPMs. c) The PCB 3 used to rearrange the different signals of the system. d) The LED emission spectrum.

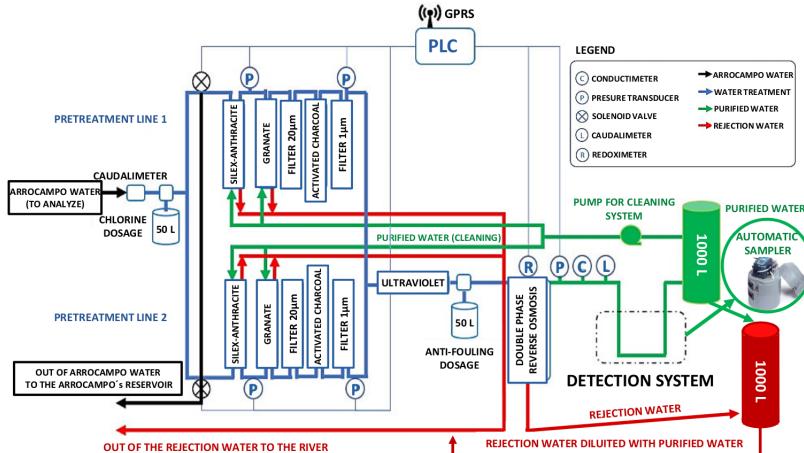


## Appendix B

# Ultrapure Water System

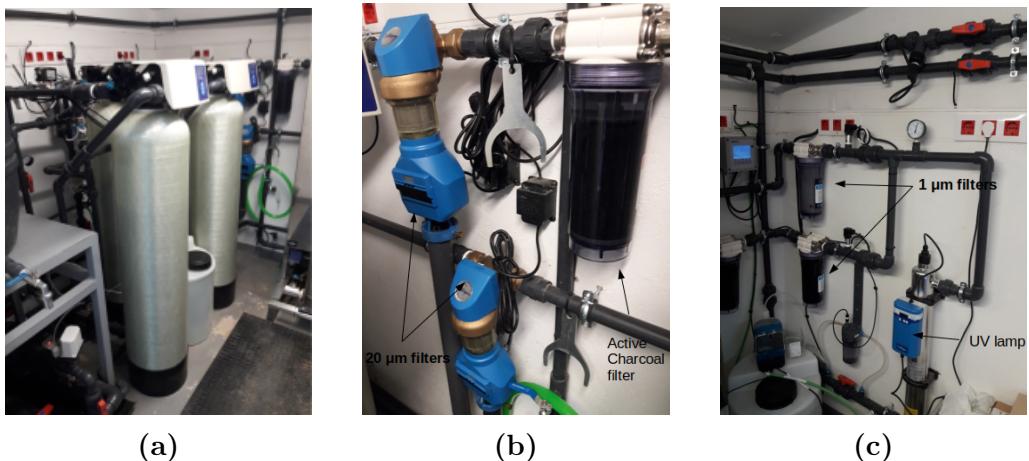
This appendix shows several photos of the ultrapure water system in the same order that the water flows through them.

First of all, the complete scheme of the ultrapure water system is shown in Figure B.1:



**Figure B.1** – Scheme of the ultrapure water system.

The Gross filtering stage, made up of Silex-Antracite and Granate



**Figure B.2** – Different stages of filtration of the ultrapure water system. a) The gross filtering stage. b) The Fine filtering stage. c) The Super fine filtering stage.

filters, the fine filtering stage, consisting of  $20\text{ }\mu\text{m}$  filter and active carbon filter and the superfine filtering, composed of the  $1\text{ }\mu\text{m}$  filter and the UV lamps, are shown in Figure B.2.

The double phase reverse osmosis is exhibited in Figure B.3a and the containers in which we store the ultrapure water and the reject water after treatment is displayed in Figure B.3b.

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 displayed in Figure B.5

Just as a curiosity, the three types of water (raw water, rejection water and ultrapure water) are exhibited in Figure B.6, where it can be visually checked the difference in the turbidity of each type of water.



(a)



(b)

**Figure B.3 – a) Doble phase reverse osmosis stage b) Containers used to store the outlet water of the ultrapure water system.**



(a)



(b)

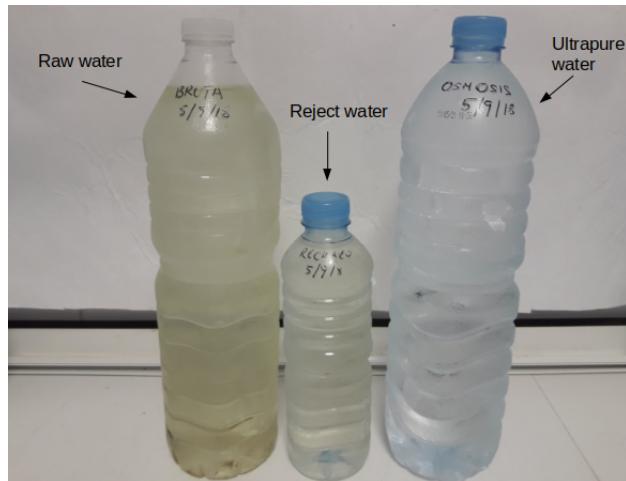


(c)

**Figure B.4 – Siemens PLC, software for remote control of ultrapure water system.**



**Figure B.5** – General photo of the complete ultrapure water system.



**Figure 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

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# Appendix D

## Electronic System of TRITIUM-Aveiro prototype

The electronic system used in TRITIUM-Aveiro prototype consists of several PCB and can be divided into two parts:

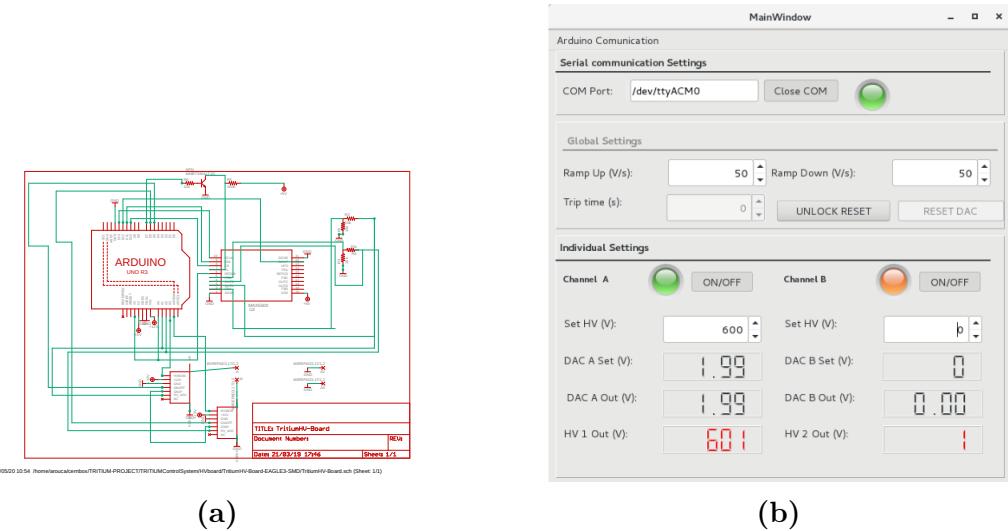
1. A PCB, whose electronic scheme is shown in Figure D.1, was designed to power the PMTs with a negative high voltage. It consists of several high voltage power supply, model C11152-01 from Hamamatsu company [K.K15], one for each PMT used, which is controlled by a DAC<sup>1</sup>, model MAX5500 from 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



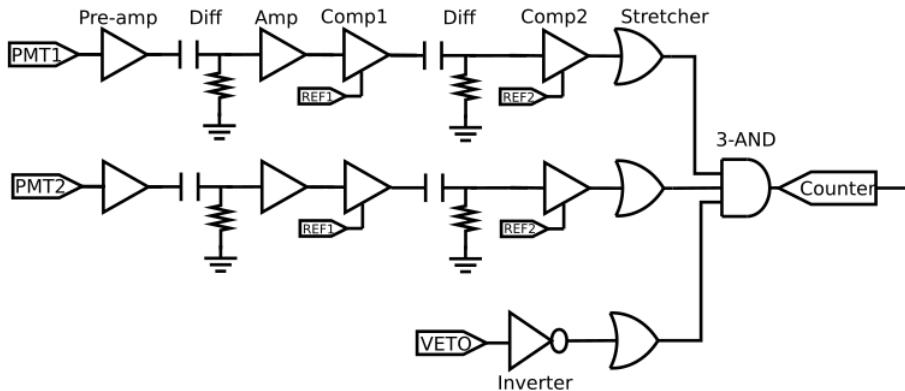
**Figure D.1** – a) The Electronic scheme of the PCB designed to power the PMTs of Aveiro prototype b) The graphical user interface developed to control it.

and analyze the system signals, whose simplified electronic scheme is shown in Figure D.2.

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 an 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 vetos are being developed, based on a rectangular plastic scintillations of Saint-Gobain company [SG], whose dimensions 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 pro-



**Figure D.2** – Simplified electronic scheme used to process and analyze the signal of TRITIUM-Aveiro prototype.

totype, 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 [Inc], 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,  $200 \mu\text{s}$ , with 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.

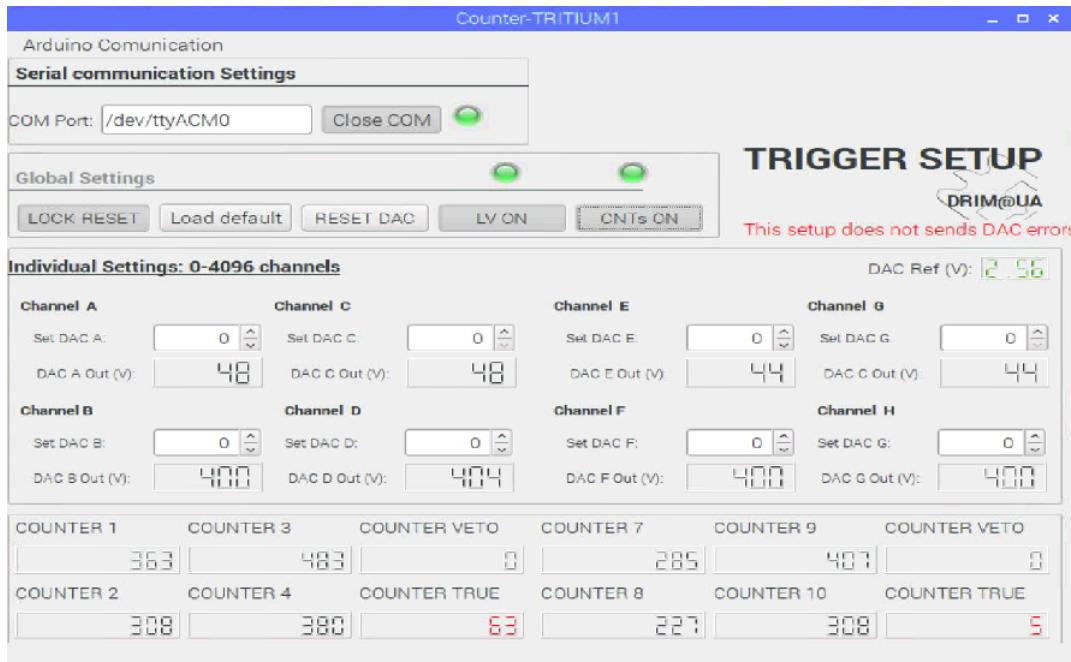
Lastly, these three signals are introduced into a 3-input AND gate, model SN74LVC1G11 from Texas Instruments company [Ins16], to perform a logic level comparison. With this last stage we achieve a temporal coincidence of both PMT signals of the prototype and anti-coincidence of them with the veto signal. 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. A graphical user interface, which is shown in Figure D.3, was developed to manage the counter system in a comfortable way.

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.

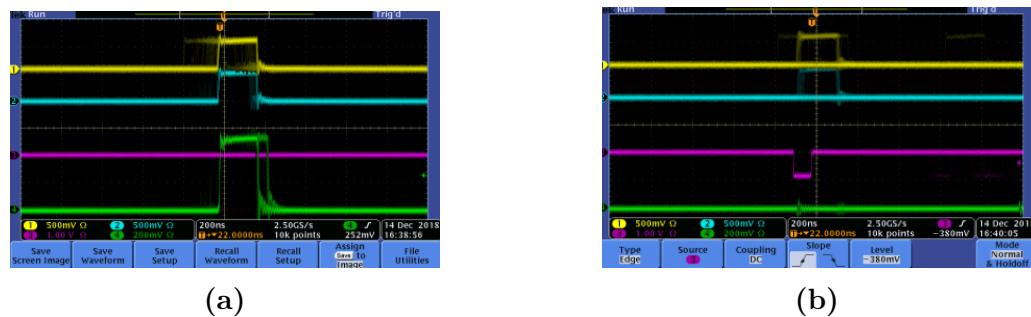
In Figure D.4 two screenshots are shown to demostrate two different situations of this system. There, we have four different signals. The yellow and cyan signal are input signals of the AND-Gate, which come from the PMT signals of the prototype. The pink signal is the third



**Figure D.3** – Graphical user interface used to manage the counter system.

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 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.



**Figure 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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