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

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*Dedicated to
my family*

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

"Alan Turing"

I

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Abstract

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

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

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

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

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

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

Keywords: Tritium in water, Real-time monitor, Nuclear Power Plant, ENvironmental Safety, ...

Nomenclature and Acronyms

Acronyms:

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

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

V_{BR}	— Breakdown Voltage of the SiPM
ΔTV_{op}	— Temperature Coefficient (mV/ $^{\circ}C$)
q_e	— Electron Charge
σ_T	— Total Uncertainty of the Measurement
σ_{st}	— Stadistical Component of the Uncertainty
σ_{si}	— Sistematically Component of the Uncertainty
A_m	— Activity Measured
$10 \mu\text{Sv}/\text{cm}$	— MicroSievert per Centimeter
$HPGe$	— High Purity Germanium Detector
$PHWR$	— Pressurized Heavy Water Reactor
UDL	— Upper Detection Limit
LWR	— Liquid Water Reactor

Atomic and nuclear symbols

${}_1^1\text{H}$	— Hydrogen
${}_1^2\text{H}$	— Deuterium (Non-Radiactive Hydrogen Isotope)
${}_1^2\text{D}$	— Deuterium (Non-Radiactive Hydrogen Isotope)
${}_1^3\text{H}$	— Tritium (Radiactive hydrogen Isotope)
${}_1^3\text{T}$	— Tritium (Radiactive hydrogen Isotope)
${}_6^{14}\text{C}$	— Carbon
${}_{19}^{40}\text{K}$	— Potassium
${}_{86}^{226}\text{Ra}$	— Radon
${}_2^3\text{He}$	— Isotope of the Helium(Non-radiactive, 1 neutrons)
${}_7^{14}\text{N}$	— Nitrogen
${}_6^{12}\text{C}$	— Carbon
${}_3^6\text{Li}$	— Lithium Isotope
${}_3^7\text{Li}$	— Lithium
${}_5^{10}\text{B}$	— Boron
${}_8^{16}\text{O}$	— Oxygen
${}_{86}^{222}\text{Rn}$	— Radon

X

$^{40}_{19}\text{K}$	— Potassium
$^{137}_{55}\text{Cs}$	— Cesium
n	— Free Neutron
H_2O	— Usual Water
D_2O	— Heavy Water
HT	— Air tritium molecule
HTO	— Water tritium molecule
OBT	— Organic tritium molecule
$T_{1/2}$	— Half-life Time of a Radioactive Element
β	— Beta Decay
$\bar{\nu}_e$	— Electron Antineutrino
e^-	— Electron
γ	— Gamma
σ	— Cross Section of a radioactive process
η_{det}	— Intrinsic Detector Efficiency
F_{sci}	— Active surface of the Plastic Scintillator
ε_{det}	— Specific Detector Efficiency
mip	— Minimum Ionizing Particle
Q_β	— Energy released in a radioactive decay
S	— Specific Energy Lost
Z	— Atomic number
$E_\gamma = h\nu$	— Energy of a photon
E_e	— Energy of a electron
m_0	— Rest mass of a electron
c	— Speed of the light in the vacumm
E_b	— Binding Energy
S_{ij}	— Single states of energy levels of electrons in scintillator
T_{ij}	— Triple states of energy levels of electrons in scintillator

Units:

mSv/yr	— Millisievert per Year
mrem	— Millirem
STP	— Standard Temperature ($0^\circ\text{C} = 273\text{K}$) and Pressure (1 atm)
W	— Watt
h	— Hour
$\text{g CO}_2/\text{kWh}$	— Grams of CO_2 per Kilowatt Hour
L	— Liter
Bq	— Becquerel, Nuclear Decay Number per Second
Bq/L	— Becquerel per Liter
Ci	— Curies
Ci/L	— Curies por Liter
yr	— Year
Ci/yr	— Curies per year
GW	— GigaWatt
$\mu\text{S/cm}$	— MicroSivers per Centimeter
kcps	— Kilo Counts per Second
pF	— picoFarads
A	— Ampere (C/s).
C	— Coulomb.
V	— Voltage.
T	— Temperature ($^\circ\text{C}$).
Vol	— Volume (m^3).

Añadir en un futuro:

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

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Chapter 1

Introduction

1.1 Tritium and Nuclear Energy

Radioactivity has been present in the Universe since its inception. It was an important element of the Big Bang¹, which occurred about $14 \cdot 10^9$ years ago. It was also present during the formation of the earth, $4.5 \cdot 10^9$ years ago, which explains why the different layers that make up the earth contain radioactive elements.

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

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

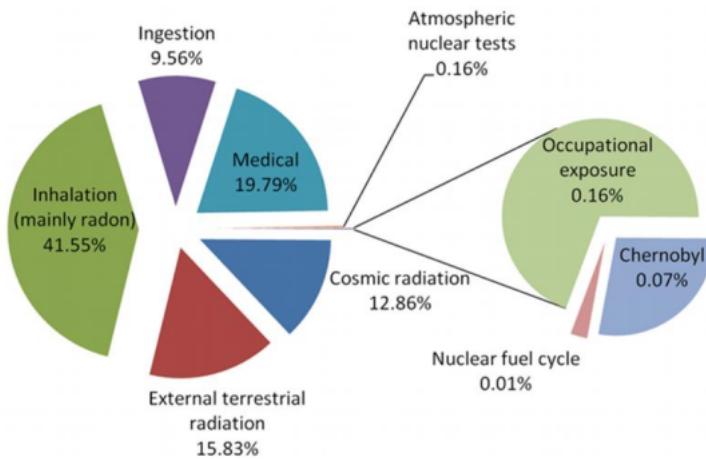


Figura 1.1 – Annual average distribution of the radioactive dose received by the population [1]

As can be seen in Figure 1.1, most of the radioactive dose received by the population is due to both, internal and external natural radiatioactivity, called natural radiation, the effective dose² of which is estimated in 2.42 mSv/yr as can be seen in Table 1.1. It can also be appreciated in Figure 1.1 that the most important part of the artificial radiation received comes from medical treatments.

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

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

Table 1.1: Annual average distribution of the effective dose received by the population due to natural radioactive [2, 3].

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

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

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

1. Firstly, a definition of concepts and units was necessary to quantify the negative effects of radioactivity and, for that, the International Commission of Radiological Units and Measurements, ICRU [4], was created during the first international conference of radiology held in London, in 1925.
2. Secondly, the International Commission on Radiological Protection, ICRP [5], was created in 1928 by the International Society of Radiology, ISR [6]. The ICRP aims to make recommendations and to provide guidance on different aspects of protection against radioactivity. The ICRP does not have the legal capacity to enforce its recommendations, but these are widely accepted the legislation of most countries.
3. Thirdly, the United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR [2], was created in 1955, the objective of which is to estimate and report the levels and effects of ionizing radiation on the population and the environment. These estimates

are taken into account by governments around the world to establish their limits and safety standards.

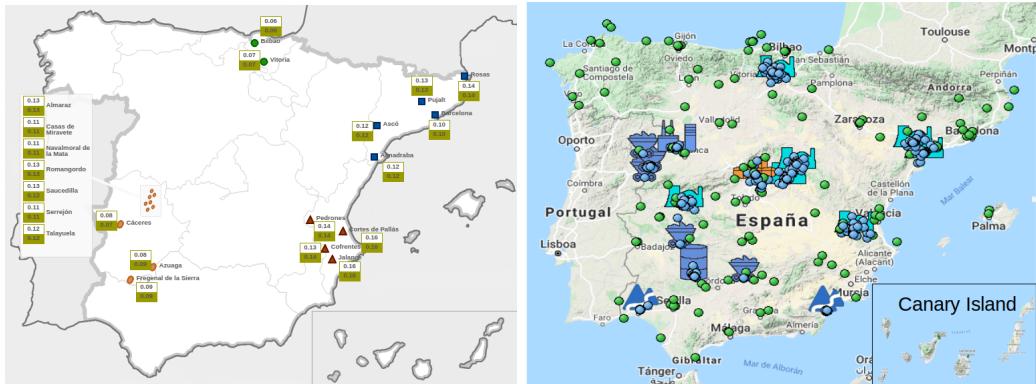
4. Fourthly, the International Atomic Energy Agency, IAEA [1], was created in 1957, which is though to promote the peaceful use of nuclear energy and to avoid its use for any military purpose such us nuclear weapons. Although it is an independent agency, it must to report to the United Nations, UN [7].
5. Fifthly, at the level of the European Union, EU, the European Atomic Energy Community, EURATOM, was created in 1957, which is a international organization stablished by the EURATOM treaty. Its objective is to coordinate research programs for the peaceful use of nuclear energy and the sharing of knowledge, infrastructure and financing of nuclear energy.
6. Finally, at the national level in Spain, the Nuclear Safety Council, CSN for its acronym in Spanish, was created in 1980 [3]. The CNS is the only institution in Spain in matter of nuclear safety and radiological protection and its objective is to reduce to the maximum the radioactivity in the environment due to anthropogenic origins.

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

- (a) On the one hand, the network of automatic stations [8], REA for its acronym in Spanish, shown in Figure 1.2a, which consists of several gamma detectors³ distributed in Spain that measure the radioactive dose in real time. The REA is used for the immediate detection of radiological problems and the application of quick response.

³Detectors that only measure gamma radioactivity

(b) On the other hand, the network of sampling stations [9], REM for its acronym in Spanish, shown in Figure 1.2b, which consists of several interesting points in Spain where samples are taken and transported to a laboratory to be measured. About twenty Spanish laboratories are integrated into this network, the objective of which is to characterize the concentration and evolution of various radioisotopes present in the radioactive background of Spain and to quantify the impact of radioactive facilities on the environment.



(a) Measured points of the REA [8]. The dose and the green box is the monthly average of the gammadots are located around nuclear facilities.

(b) Measured points of the REM [9]. Blue dots are the daily average of the gammadots are located around nuclear facilities. Green dots are uniformly distributed in Spain.

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

There are other networks that measure different parameters such as the concentration of ^{226}Ra in the air and the measurements of all the networks are adapted to the EUROTAM treaty [10].

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 time⁴. This monitor is destined to be finally included in the REA.

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

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

The monitor will be used to ensure that the tritium levels of the Arrocampo dam water are below of the legal limit specified in the EURATOM treaty [10], which is 100 Bq/L. It will be used indirectly to verify the correct operation of the Almaraz NPP, located 4 km above the river since a malfunctioning of it will produce an increase of the tritium activity.

Tritium is one of the most abundantly produced radioisotope in a NPP, as it was verified in the United States Department of Energy complex, (U.S. DOE) [11, 12] and in several research facilities in China [13] and places around them (ground water, surface water and process waste water).

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

Tritium is usually produced in the water used in the nuclear reactor cooling system of some NPPs. It is produced by neutron capture of deuterium, existing in the heavy water (D_2O), semi-heavy water (HDO) or the deuterium created by neutron capture in usual water (H_2O). All these processes have a large probability to happen due to the huge neutron flux in the nuclear reactor, of the order of $10^{14} \text{ n cm}^{-2}\text{s}^{-1}$ [14]. This tritium is finally released partially or totally to the environment with a quantity that depends on the reactor type as it is shown in Table 1.2. The most common way that tritium is released to the environment is HTO [13].

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) [13]

NPPs are operational since more than 60 years and, nowadays, they are essential for providing a large part of the electric power used in the world (more than 20% in Spain [15]). Although the Spanish government is planning to progressively shut down all NPP there are other countries like China [16] or United States, USA [17], that promote their use.

On the one hand, NPPs are an interesting investment since it is one of the cheapest source of energy production. It is stable, as it doesn't depend on meteorological parameters and it doesn't emit greenhouse gases. Although there are other alternative energy sources which are being developed quickly (photovoltaic, wind, tidal energy, etc.), even other concepts of energy production and saving (local production, solar roofs, energy efficiency, smart cities, etc.), today they are not developed enough to fully cover

the population needs. On the other hand, NPPs still have some problems such as the contamination of fresh water from uranium mining, the nuclear waste produced, the nuclear proliferation or the risk of radioactive contamination from accidents as happened in the past: Chernobyl, Fukushima and Three Mile Island [18].

In any case nuclear energy production in the world is not going to stop in the next decade, in fact, it will increase as the United States Energy Information Administration (U.S. EIA) expects [19]. Therefore the development of different types of alarm systems is a good investment. Safety is not a negotiable aspect and there must be mechanisms that warn us of any malfunction of a nuclear power plant.

In addition, it is important to highlight that the developed monitor could be used to verify the correct operation of a nuclear power plant, but it is not our objective. Our much broader objective is to ensure that the levels of tritium in the analyzed water are below the Spanish legal limit. It means that this monitor could be used in many different places with radioactive facilities like the future fusion power plants⁵, nuclear research facilities⁶ or tracking the pathway of tritium discharges to ground water [22].

1.2 Tritium Properties and Radiological Hazards

Tritium is the only radioactive isotope of hydrogen present in the environment. It was for the first time produced in 1934 from neutron capture of deuterium by Ernest Rutherford, Mark Oliphant and Paul Harteck [23] and it was first time isolated in 1939 by Luis Walter Alvarez and Robert Cornog

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

⁶Tritium is one of the main emissions from these sites [20], [21].

[24], 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 ($^{14}\text{N}(\text{n}, ^3\text{H})^{12}\text{C}$) [25] and oxygen ($^{16}\text{O}(\text{n}, ^3\text{H})^{14}\text{N}$) [26]. 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 [13, 25].

Tritium can be produced artificially in the environment from different anthropogenic sources [13, 25]. 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 $8 \cdot 10^9 \text{ Ci}$ ($2.96 \cdot 10^{11} \text{ GBq}$) and a part of which remains to the date. In these nuclear explosions, tritium was produced mainly from the nuclear reactions $^{14}\text{N}(\text{n}, ^3\text{H})^{12}\text{C}$ and $^2\text{H}(\text{n}, \gamma)^3\text{H}$. Tritium can also be produced by commercial producers of radioluminescent and neutron generator devices ($1 \cdot 10^6 \text{ Ci/yr}$), nuclear power and defense industries (around $2 \cdot 10^6 \text{ Ci/yr}$) and several research facilities and nuclear reactor for energy production ($2 \cdot 10^6 \text{ Ci/GWyr}$), through the production cross sections shown in Table 1.3:

Source	Origin	Nuclear reaction	Cross section (b)
^2_1H	Water coolant	$^2_1\text{H}(\text{n}, \gamma)^3_1\text{H}$	$5.2 \cdot 10^{-4}$
^3_2He	Helium coolant	$^3_2\text{He}(\text{n}, \text{p})^3_1\text{H}$	5330
^6_3Li	Moderator	$^6_3\text{Li}(\text{n}, \alpha)^3_1\text{H}$	940
$^{10}_5\text{B}$	Moderator, control rods	$^{10}_5\text{B}(\text{n}, 2\alpha)^3_1\text{H}$	3835

Table 1.3: Most common nuclear reactions of artificial tritium production [13]

Tritium levels in the environment excluding anthropogenic radioactive sources are between 1 and 4 Bq/L, larger than the expected due to the cosmogenic background levels ($0.6 - 1.2 \text{ Bq/L}$, previously mentioned) [27]. It can be explained by the consequences of nuclear weapons tests.

Tritium levels in rivers around a nuclear facility are between 1 and 10 Bq/L and even between 20 and 50 Bq/L at the water discharge site of NPPs [27], where the produced tritium is partially or totally released into the environment, mainly in the HTO water form.

The effect of NPP on tritium levels can be observed from REM date, for example the case of Cofrentes. Cofrentes is the closest nuclear power plant to Valencia, in the measurements of which are involved the LARAM⁷. There, the tritium level is measured in three different places along the Jucar river, marked on the map shown in Figure 1.3. The first place, P1, is located in the river, 6 km upstream from the NPP, the second place, P2, is located 1 km downstream and the third place, P3, is located 5 km downstream. The level of tritium measured in these three locations is shown as a function of the time in Figures 1.4a, 1.4b and 1.4c respectively.

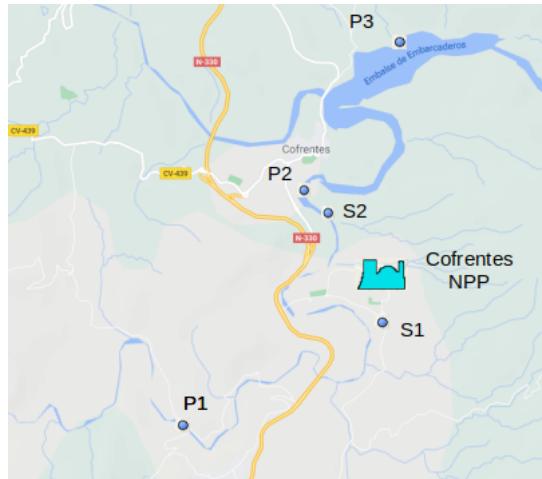
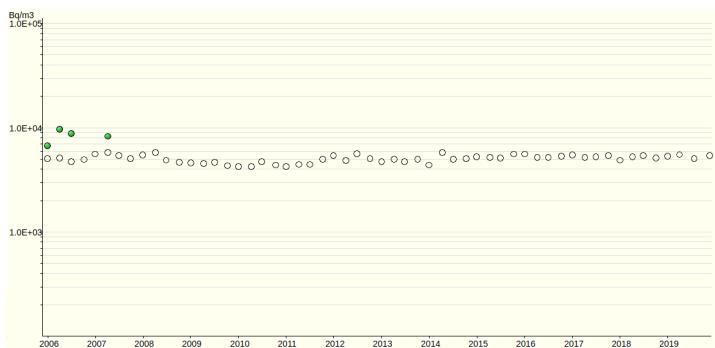


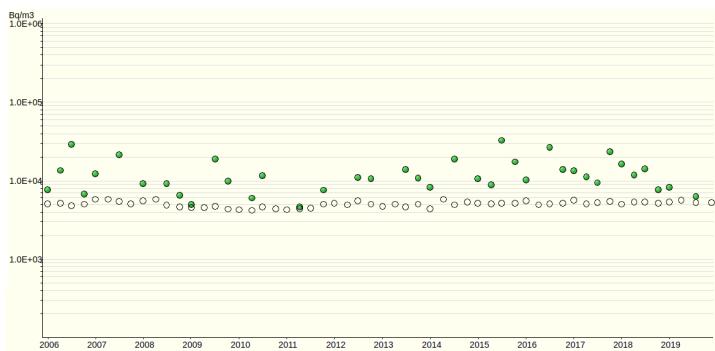
Figura 1.3 – Tritium sampling locations around Cofrentes NPP.

In these figures, the detection limit and the measured activity are shown using white and green dots, respectively. The measured activity is only displayed when it is larger than the corresponding detection limit. The

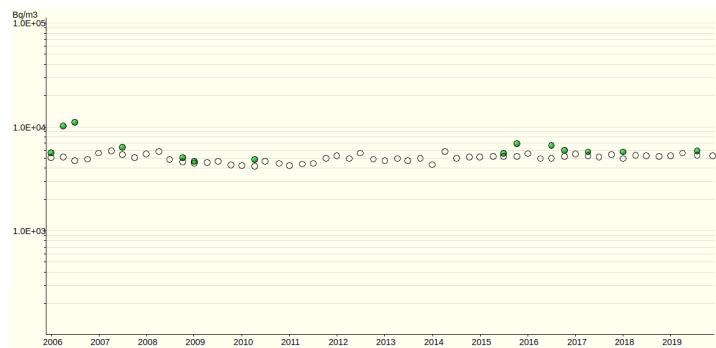
⁷The LARAM is a Valencia laboratory specialized in environmental radioactivity measurements



(a) Tritium activity 6 km upstream.



(b) Tritium activity 1 km downstream.

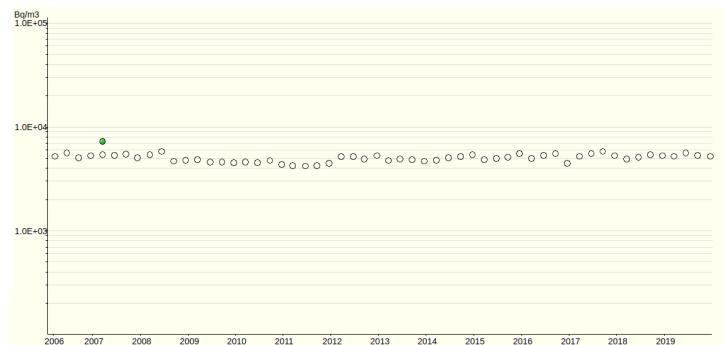


(c) Tritium activity 5 km downstream.

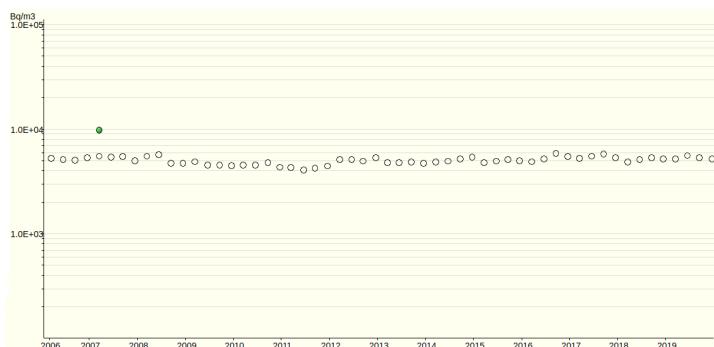
Figura 1.4 – Tritium activity levels in surface water around Cofrentes NPP from January 2006 to November 2019. The white points are used for the detection limit and the green points are used for the measured activity, when it is above the detection limit. [9]

tritium level in the river increases due to the discharge of the NPP and it is diluted again after 4 km downstream, as can be seen from these date.

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



(a) Tritium activity 1 km before NPP.



(b) Tritium activity 1 km after NPP.

Figura 1.5 – Tritium activity levels in groundwater around Cofrentes NPP from January 2006 to November 2019. [9]

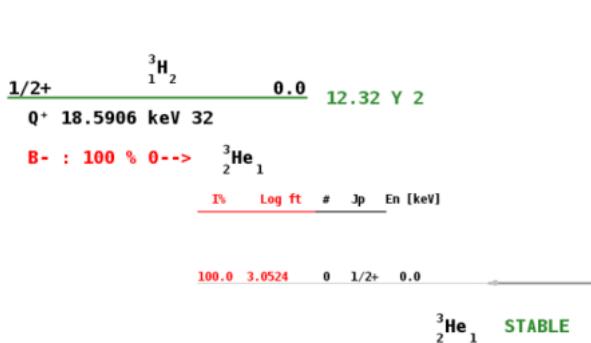
It is important to note that, although environmental tritium level is affected by NPP, these levels are below the maximum allowed limit. The maximum level of tritium measured since of January 2, 2006 is around

32 Bq/L, below to the maximum allowed limit in Europe, 100 Bq/L.

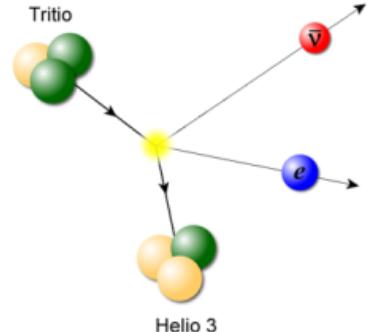
Tritium is a radioactive element with a half-life time of $T_{1/2} = 12.32$ years. It has one proton and two neutrons and decays exclusively through β radiation. It decay 100% directly to the ground state of the ^3_2He isotope of helium, which is a stable nuclei, through the decay scheme of equation 1.1:



In Figure 1.6 the scheme of tritium energy levels is shown. In this decay it is not possible to detect the neutrino because of its extremely weak interaction with matter ($\sigma \propto 10^{-42} \text{ cm}^2$ [28]) and, since ^3He has a much larger mass than electrons and neutrinos, by conservation of energy and momentum, the energy that is taken by this daughter nucleus is very small. Therefore, the detection of tritium is through its decay electron.



(a) Tritium energy levels [29]



(b) Graphic representation of tritium decay [30]

Figura 1.6 – Tritium decay

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.

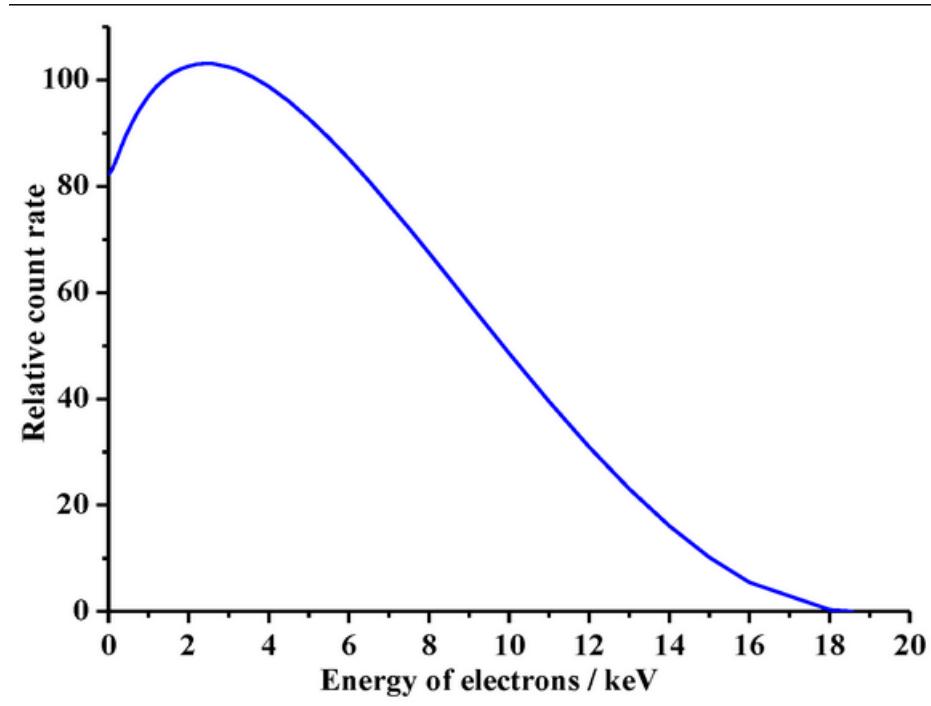


Figura 1.7 – Energy spectrum of tritium electrons [31]

The releasing energy of the tritium decay, is very low. In fact, it is the radioactive isotope with the lowest energy released in its β disintegration [25]. Consequently, the β particle which is emitted in this tritium decay will have a very small mean free path, shown in Table 1.4.

This short mean free path is a major issue in tritium detection, as it makes more difficult the electron detection, which will require a highly sensitive detector. It means that tritium electrons have a low penetration in our body and they are easily stopped with clothes or laboratory gloves, resulting in a low radiological hazard of external tritium.

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

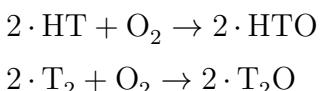
Table 1.4: Penetration depth for decay electron of mean (5, 7 keV) and maximum (18, 6 keV) energies in different media (tritium gas and air at standard conditions of temperature (273 K) and pressure (1 atm), STP, and water) [32]

Nevertheless, the danger of tritium increases when it is ingested or inhaled since it can bind anywhere that hydrogen can and perform the same chemical reactions, sometimes with higher rate if the tritium concentration is high enough to catalyze the reaction.

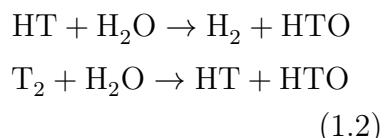
Tritium can be absorbed in our body in three different ways, gaseous tritium (mainly HT), tritiated water (mainly HTO) and organically bound tritium (called OBT).

- The gaseous tritium, which is normally found mixed in the air, is the least important since less than a $3 - 5 \cdot 10^{-3} \%$ is absorbed by the human body, which is insignificant [25]. However, it can be transformed into tritiated water, more harmful from a radiobiological point of interest [25], through the oxidation and exchange reactions by equations 1.2:

Oxidation :



Exchange



- Tritiated water, which is normally found in drinking water and food, has a larger impact since the 99% of it is absorbed [25]. Its biological life time corresponds to the water cycle in the body, around 9.5 days ($\pm 50\%$), time during which tritium will remain in our body [25, 27, 33]. As in water, the biological life time of tritiated water can vary due to various external parameters such as temperature, humidity, drinking habits, etc. or reduced with the use of diuretics [25].
- Organically bound tritium, normally found in food, generally forms a covalent bond with a carbon and it corresponds to 5 – 10 % of tritium absorbed in the body. Although it is less absorbed in the body than tritiated water, it can be more dangerous since it has a longer biological life time. The biological life time of this tritium type depends on the affinity of the organic molecule to the different biological tissues and it can vary from tens to hundreds of days (larger than the ICRP estimate) [27, 33, 34, 35].

There are many studies showing that tritium in living matter can cause the same effects than X-rays or γ rays, which are mutations, tumors, cancer, genetic effects, reproductive effects, etc [36, 37]. In fact, the consequences of tritium radiation can be worse than a similar γ radiations since its biological efficiency⁸ is two or three times larger [36].

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

⁸The 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, which was dealed with in section 1.2, it is important that the current legislation limits the release of tritium to the environment ensuring that the levels are below a safe value for health.

The guidelines setting impose the limit of radioactive elements in drinking water for many countries are based on the radiation protection methodology developed by the ICRP [38] and the recommendations of the world health organization (WHO) [39].

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

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

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

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

Where RDL is the reference dose level, DCF is the dose conversion factor (the normal used value for tritium is $1.8 \cdot 10^{-11}$ Sv/Bq, provided by ICRP [40]) and q is an estimation of the annual volume of drinking water consumed (normally assumed two liters per day, 730 L/yr).

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

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

Most of the countries in the world implement the RDL of 0.1 mSv/yr recommended by the WHO. The legal limit for tritium in drinking water in this case is 76,103 Bq/L but it is often approximated in different ways. Some countries like Switzerland [41] or some organizations like the WHO [39] rounded this value to 10,000 Bq/L. Others like some countries of Canada, such as Ontario and Québec, truncate this value to the first number 7,000 Bq/L [42, 43]. There are other countries like Russia which use the much more accurate approximation value of 7,700 Bq/L [44].

There are other countries like Australia that prefer to implement the RDL of 1 mSv/yr, recommended by the ICRP, the legal limit of which is 76,103 Bq/L [45]. Other countries like Finland are based in the ICRP recommendations and use only the middle of this value, 0.5 mSv/yr, the value of which is rounded to a legal limit of 30,000 Bq/L for tritium in drinking water [46].

There are two different exceptions to these recommendations:

1. On the one hand, most of the USA states like California use a RDL of 4 mrem (0.04 mSv), which corresponds to a legal limit of 20 nCi/L (740 Bq/L)

[47]. This value was proposed by the United States Environmental Protection Agency (US EPA) as a result of an analysis of the available information [48].

2. On the other hand, most of the EU countries, such as France, Germany or Spain, impose an GL of 100 Bq/L, which is one of the most restrictive limit in the world [49, 50, 51]. 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 [52].

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

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

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

1.4 This thesis

Chapter 2

Tritium Detection Systems

2.1 State-of-the-Art

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

Nowadays, the most used technique for measuring tritium in water is liquid scintillator counting (LSC). This technique consists of mixing a liquid sample (some ml for environmental measurements or less for higher activities) with liquid scintillator. This mixture is usually made in a ratio of 50:50 but it depends on the detection system and the samples used [53, 54]. In this technique, the β energy emitted from the sample excites the molecular energy levels of the liquid scintillator which promptly decays emitting several photons with a well-known energy (fluorescence), usually in the visible spectrum. Finally these photons are detected with photosen-

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

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

sors, which convert the optical signal into measurable electrical charge. The liquid scintillator technique has a very good detection sensitivity for low activity levels of tritiated water (< 1 Bq/L) [55] but it has the problems of long measurement time (up to 2 days) and of producing chemical waste, as liquid scintillator contains toluene which is toxic. In addition, this technique requires special staff for sampling, chain-of-custody and lab analysis which require economical and time resources. In order to avoid this problem some unsuccessful efforts have been made in order to build a monitor of tritium with LSC [56].

The ionization chamber (IC) consists of a gas chamber (sample) which contains electrodes connected to different voltages. These electrodes collect the ionization current that is produced due to the β radiation. It is a simple and fast system, but it has the problem of high Low Detection Limit (> 10 kBq) and of requiring the samples to be in a state of gas or steam [57, 58].

The calorimetry method is based on the measurement of the heat generated in the detection medium (normally platinum) [59, 60]. The problem with this technique is that it has a high LDL, of the order of GBq, and requires long measurement time, 2 days or more.

The Beta Induced X-ray Spectrometry (BIXS) is based on the

measurement of the bremsstrahlung radiation produced by the tritium decay electrons, using a NaI(Tl) crystal couplet to a PMT [61, 62] or Silicon Drift Detector (SDD) [63]. The problem with this technique is a high LDL, of the order of MBq.

There are additional methods for tritium detection, although they are less employed or less experimentally developed, each one with its own advantages and limitations. For example, the Avalanche PhotoDiode (APD) cannot be used in contact with water [64], the mass spectrometry which needs to store the sample several months before taking the measurement [65] and the Cavity ring spectroscopy requires a special optical configuration that is not possible outside a laboratory [66].

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

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

1. The study done by M. Muramatsu, A. Koyano and N. Tokunaga in 1967 who used a scintillator plate read out by two PMTs in coincidence [67].
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 [68].

3. The study performed by R. V. Osborne in 1969 that used sixty stacked scintillator plates read out by two PMTs in coincidences [69].
4. The study done by A. N. Singh, M. Ratnakaran and K. G. Vohra in 1985, that used a scintillator sponge read out by PMTs in electronic coincidence [70, 71].
5. The study carried out by K. J. Hofstetter and H. T. Wilson in 1991, that tested different shapes of scintillator plastics like several sizes of beads, fibers, etc. The better result obtained for solid plastic scintillator was a tritium detection efficiency of the order of 10^{-3} [72, 73].

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

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

The results of these experiments are summarized in Table 2.2. As can be seen, in the first column that the intrinsic detector efficiency, η_{det} , is very different in these experiments. As one of the most important factor that affect the efficiency is the active surface of the plastic scintillator, F_{sci} , which varies largely with the detector type, the specific detector efficiency (third column) is used in order to compare these experiments, that is, the intrinsic detector efficiency normalized to this active surface. It can be checked that, effectively, these specific efficiencies are quite similar. The specific efficiency obtained by Moghissi for scintillating fibers is sufficiently high to justify our choice of scintillating fibers as a detection medium. Finally, as can be seen

in the last column, the LDL in all these experiments are of the order of a few tens of kBq/L. Thus, to develop a detector which much lower LDL is essential to comply with the EURATOM directive of 100 Bq/L of tritium in water for human consumption.

2.2 TRITIUM Project

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

To overcome these limitations the *Tritium* project [74], with the title of "Design, construction and commissioning of automatic stations for quasi-real time monitoring of low radioactive levels of tritium in water", was proposed.

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

This project was funded by the Interreg Sudoe program of the European Economical Community, EEC, in the 2016 call with the reference number SOE1/P4/EO214. The purpose of this project is the development of a tritium monitor in quasi-real time. This monitor consists of a ultra-pure water system, which prepare the water sample before introducing it in the detector for tritium measurement, a cosmic veto and a pasive shielding, which reduce the natural background of our tritium detector, and several electronic modules which control all the different parts of the monitor, an-



(a) Arrocampo dam and Almaraz Nuclear Power Plant (b) Tajus river along Spain and Portugal

Figura 2.1 – Arrocampo dam, Almaraz NPP and Tajus river

alyze the tritium measurement and send an alarm if the configured limit (100 Bq/L) is exceeded.

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

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

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

1. The Extremadura group has developed and installed the ultrapure water system to produce water with very low conductivity, $\sigma \approx 10 \mu\text{Sv}/\text{cm}$ (two orders less than sample before the cleaning process, $1000 \mu\text{Sv}/\text{cm}$). This cleaning process is very important for two reasons. On the one hand, for maintaining our detector very clean, which is a critical point. On the other hand, to reduce the natural background since several natural radioactive isotopes presents in this water (except tritium) are removed such as ^{222}Rn , ^{40}K or ^{137}Cs . This system is explained in section 3.3.
2. The french group has develop the pasive shielding for the detector. The shielding is made of ultra radiopure lead with very low intrinsic activity in order to reduce the external natural background of the system. This shielding is presented in section 3.4.1.
3. The Portuguese and Spanish group have collaborated for designing, developing and building four different prototypes of tritium detector and active vetos for removing cosmic events. These prototypes and vetos are explained in chapter 5 and section 3.4.2 respectively.
4. The Portugal and Spanish people have also carried out simulations of this system. The environment chosen to develop these simulations is the package GEANT4 [75], consisting of an extensive C++ library with which the geometry of our detector, the relevant physical processes, etc. can be designed. This simulation are explained in chapter 6.

The important characteristics of the TRITIUM detector must have:

- *Compact*. This is an important point because in the place where this detector will be installed the useful space to be used is very limited.
- *Thin active volume and large active area*. On the one hand, it have to be taken into account that the mean free path of the β particle of

tritium decay is very low so thin detector active volumes are needed. In practice, active thickness beyond the mean free path of the tritium electrons will only contribute to background. On the other hand, as explained in section 2.1, the efficiency of this type of detector scales with the active area, so it is crucial to design the detector with the largest possible active area.

- *High efficiency to tritium.* As the tritium activities to be measured are very low, it is very important to reduce as much as possible the non-detected tritium events.
- *High specificity to tritium.* The detector has to be able to distinguish the tritium signal from the signal due to other radioactive elements present in the sample.
- *Quasi-real time response.* It is important that the system work in quasi-real time in order to detect any problem as fast as possible.
- *Rugged system.* Finally, it has to be taken into account that the final goal is to install an automatical system which works during a number of years without requiring the intervention of specialized people, which requires a rugged monitor.

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

Chapter 3

Design Principles of the Tritium Monitor

3.1 Detector System Overview

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

1. The TRITIUM detector, described in chapter 5, is based on several modules read in parallel. Each module consists of hundreds of scintillating fibers, section 3.2.2, which are in contact with the water sample measured, read by two coincident photosensors, section 3.2.3. The photosensors 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 the deposition of particles onto the fibers because this would prevent the tritium decay electrons from reaching the fibers. Second, particles dissolved in water may contain radioactive isotopes like ^{40}K , which would increase the background. As the water sample has very low tritium counters, to reduce the background is a crucial matter.

3. The background rejection system (section 3.4), that has two different parts. The first one is a passive shield (section 3.4.1), consisting of a lead castle inside of which the TRITIUM detector is located. This castle is employed to eliminate natural radioactive background and cosmic rays with energies of the order of 200 MeV/nucleon . The second part is an active veto (section 3.4.2), consisting of two plastic scintillation blocks located inside of a passive shielding, above and below the TRITIUM detector and read by several photosensors. The goal of this active veto is to remove the remaining high energy events ($> 200 \text{ MeV}$) cosmic rays that can travel through the passive shielding and contribute to background. Contrary to low energy cosmic rays, high energy cosmic rays are difficult to stop so. 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 limit of the tritium level, 100 Bq/s , is exceeded.

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

3.2 TRITIUM Detector

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

- The scintillator, that detects the tritium event, as ionizing radiation hits this material and deposits kinetic energy through ionization and excitation processes, part of the absorbed energy is converted in to photons, mostly in the visible range¹. The produced photons carry information about the particle detected, like its energy, type, etc.
- The photosensor, that detects the photons produced in the scintillator. The most common photosensors in nuclear physics are PMTs and SiPMs. They detect the photons produced in the scintillator with an efficiency and transforms them in electrons with a multiplication factor of around 10^6 . These electrons form a electronic pulse than gives information of the detected photons.
- The electronic system, which is the part of the scintillator detector in charge of processesing and analyzing (first analogically and then digitally) the electrical pulse given by the photosensor. The output of the electronic system is the useful information about the events detected such as number and energy spectrum.

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

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

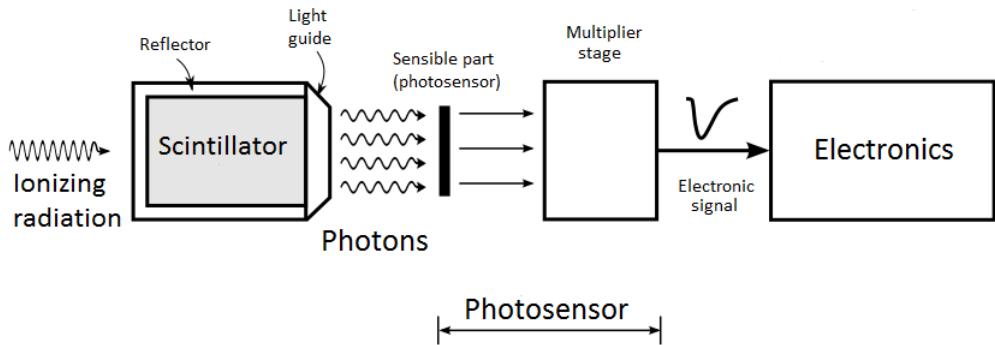


Figura 3.1 – Scheme of the scintillator detector

3.2.1 Interaction of Fast Electrons and Photons with Matter

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

Electrons have charge so their interaction with matter is mainly with the orbital atomic electrons through the Coulomb force. The electron trajectory is much more tortuous than other heavier particles because the mass of both interacting particles is equal. Furthermore, for the same reason, these electrons lose a significant amount of energy in each collision. The specific energy loss is defined as $S = -\frac{dE}{dx}$ which gives the energy loss suffered by the particle per unit of path length. In the case of electrons, this total energy loss has two main contributions, the collisions (elastic and inelastic) and radiative processes (bremsstrahlung) [76, 77]:

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

The radiative part is roughly proportional to the collision part:

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

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

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

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

The photoelectric effect occurs when a photon interacts with an orbital electron in the material, losing all its energy. This energy is absorbed by the electron that is released from the atom (ionization). The energy of the resulting electron, E_e , is [76, 77]:

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

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

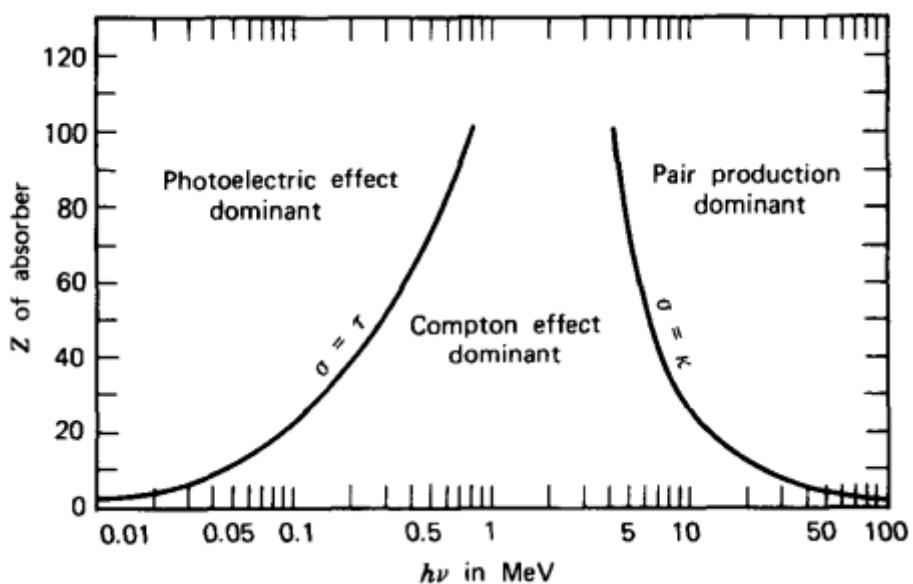


Figura 3.2 – Domain regions of the three most probable types of interactions of gamma rays with matter. The lines show the values of Z and $h\nu$ where the two neighboring effects are equally likely. [76, 77]

the expression [76]:

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

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

The Compton effect occurs when a photon interacts with an orbital electron of the material, transferring part of its energy to the electron, which is released, scattered at an angle θ with respect to the original direction. If the electron binding energy is neglected, the energy, E_e transferred to it is given by [76, 77]:

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

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

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

Finally, for coherent scattering, the atom is neither excited nor ionized and the photon conserves all its energy in the collision. Coherent

scattering is more probable for photons with low energies and materials with high atomic numbers and, as it will be shown in section 3.2.2, it explains why the produced photons are guided along scintillating fibers.

3.2.2 Plastic Scintillators

Scintillators are widely employed for radiation detection in nuclear physics. Scintillator converts kinetic energy of the incoming particles in to light² which can be detected and quantified. Light emission happens through photon de-excitation of excited atoms.

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

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

Organic scintillators are based on a scintillator material dissolved in a base solvent, normally aromatic hydrocarbons as C₁₈H₁₄, C₂₄H₂₂N₂O or C₁₅H₁₁NO with an average atomic numbers of which are between 3,5 and 5.

²The light is made up of photons in the visible energy range.

The scintillator molecules, in which the organic scintillators are based, have a π -electron structure. The energy levels of their electrons are commonly illustrated with a Jablonsky diagram, shown in Figure 3.3, which shows the fundamental singlet state, S_{0i} , where the valence electrons are, the excited singlet states, S_{jk} , and the excited triplet states, T_{lm} . The energy difference between S_1 and S_0 states is around 3 or 4 eV, in the visible range. As it is shown in the figure, each energy states are splitted in close sublevels separated around 0.15 eV. This fine energy structure is due to excitations of molecular vibrational modes tabbed by the second index of the energy states. As the energy levels and sublevels have an energy larger than the thermal energy, 0.025 eV, non-excited electrons are in the ground state S_{00} at STP³.

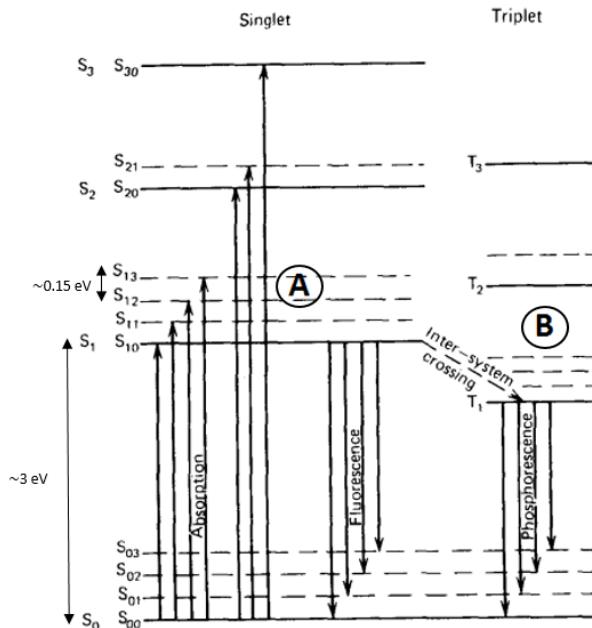


Figura 3.3 – Jablonsky diagram. [76]

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

³Standar temperature and pressure conditions

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

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

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

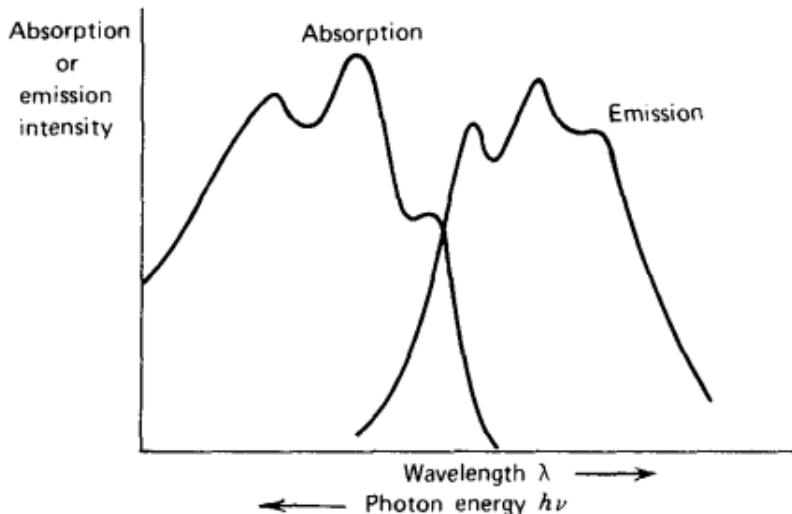


Figura 3.4 – Stokes shift. [76]

The intensity of the fluorescence emission in an organic scintillator

over time is a combination of two exponential functions, one associated with the lifetime of the level, τ (on the order of nanoseconds), and the other associated with the energetic level population, τ_1 (on the order of picoseconds) [76].

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

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



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

As the prompt fluorescence light produces the scintillator signal, detector design should increase it and reduce other possible physical mechanisms. One of the most important parameters is the scintillation yield⁴, defined as the the number of photons emitted by unit of absorbed energy. This yield depends on the type of particle and on other mechanisms that

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

doesn't produce prompt fluorescence, like phosphorescence or delayed fluorescence or even internal conversion. The scintillator yield is normally quoted by the manufacturer for mips⁵.

Plastic scintillators are easy to machine to any desired shape. The chosen shape for TRITIUM detector is the fiber, specifically, commercial fibers BCF-12 from Saint-Gobain Crystals Inc company [78]. This type of fiber was chosen as the result of a comparative study [79] among some of the best-known commercial manufacturers.

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

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

The scintillation light is guided to the sensitive part of the photosensor. A single photon produces a signal with some probability, called the quantum efficiency. Fibers (and scintillators in general) use the optical property of Snell's law [80] to guide their photons to the desired part (ends of the fibers). It is based on the interface created between the core and

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

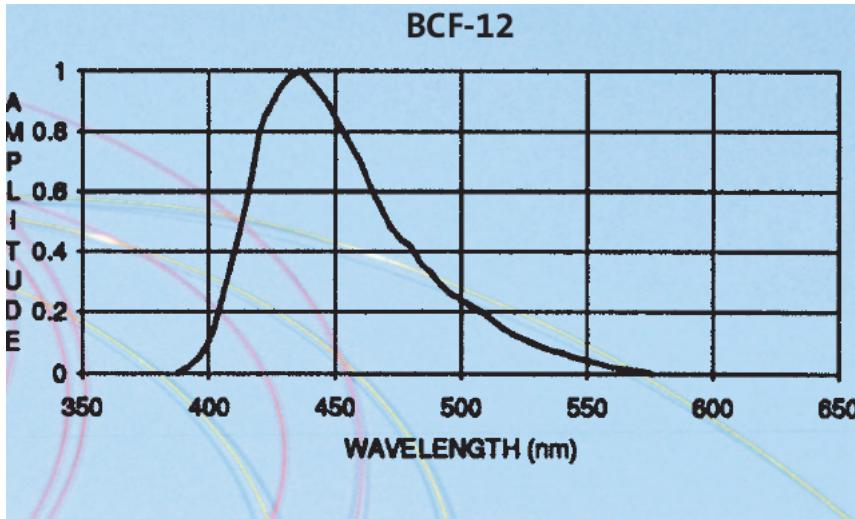


Figura 3.5 – Emission spectrum of BCF-12 fibers of Saint-Gobain. [78]

the surrounding material. When a photon hits this interface, it is refracted (and therefore lost) following the Snell equation, 3.8 [80].

$$n_0 \operatorname{sen}(\theta_0) = n_1 \operatorname{sen}(\theta_1) \longrightarrow \theta_c = \operatorname{asen} \left(\frac{n_1}{n_0} \right) \quad (3.8)$$

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

The trapping efficiency or photon collection efficiency is defined as the efficiency of the scintillator to guide photons. For BCF-12 fibers with optical clad is between 3.44% and 7% per meter of fiber (depending on where the event is detected and is minimum near the fiber axis and maximum near the core-clad interface. For no clad fibers BCF-12 surrounded by water, the trapping efficiency is larger than for cladded fibers. Therefore, from the maximum of 148 photons initially created by a tritium decay electron with

the maximum energy, only 41 photons (for maximum trapping efficiency) are guided in the 25 cm fiber length used in the TRITIUM detector. Thus, the output signal is very weak and is in the range of the spectrum where electronic noise is already significant. As described in the following chapters, a great effort was made to minimize electronic noise by different techniques.

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

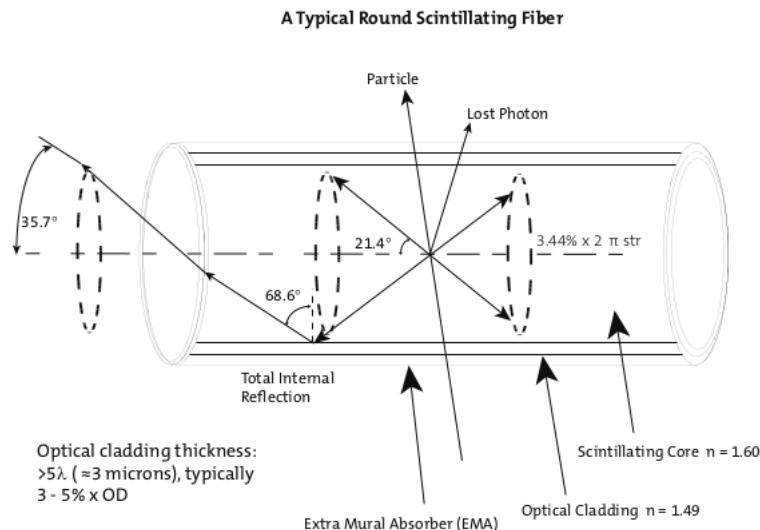


Figura 3.6 – How photons are collected in a fiber with single clad. [78]

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

In the practice, it is difficult to achieve a perfect air-core or water-core interface which affects light collection. As commercial claddings are thicker ($30 \mu\text{m}$) 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

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

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

enough. To overcome this problem a special protocol was developed in the ICMOL laboratory for preparing fibers for tritium detection.

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

3.2.3 Light Detection in Photosensors

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

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

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

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

Table 3.2: Properties of BCF-12 fibers from Saint-Gobain Inc. [78]

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

Photoelectron Multiplier Tubes (PMTs)

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

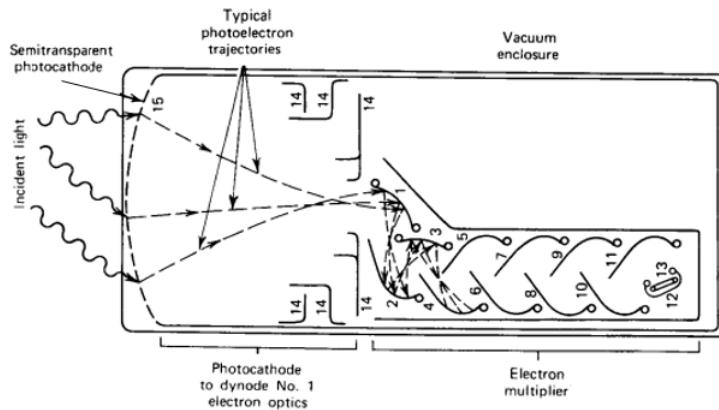


Figura 3.7 – Scheme of a PMT. [76]

The signal production has two phases:

1. In the photoathode photons are converted in photoelectrons through photoelectric effect. The photocathode consists of a thin layer, of the order of nanometers, deposited on the inner surface of the PMT window. The material of the photocathode is chosen to optimize the probability of producing photoelectric effect with the scintillating photons. The PMTs used in TRITIUM experiment are the model R8520-406 from Hammatsu [81] and the material of their photocathode is Bialkali⁶.

⁶The bialkali material is based on the elements ^{121}Sb , ^{85}Rb and ^{132}Cs

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

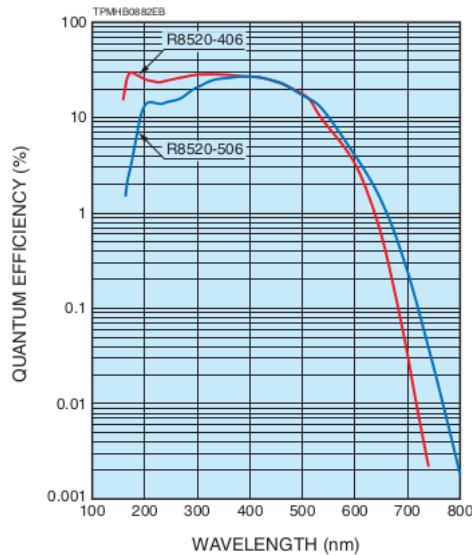


Figura 3.8 – Quantum efficiency spectrum for the PMT used (R8520-406). [81]

The maximum values of the PMT quantum efficiency is usually between 20% and 30% [76] (a little bit less than 30% for the PMTs employed). The emission spectrum of the scintillating fibers used, Figure 3.5, matches the quantum efficiency spectrum of the PMTs used, Figure 3.8 and the position of both peaks is very close, 435 nm for fibers and 420 nm for PMT.

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

Focusing electrodes guide the photoelectrons to the first dynode. They have a collection efficiency (CE) defined as the ratio of the number of photoelectrons reaching the first dynode and the number of photoelectrons leaving the photocathode and its value is around 80%. The dynodes achieve the electron multiplication. A voltage difference between adjacent dynodes accelerates the electrons and produce their multiplication. The multiplication factor of each dynode, δ , is commonly around 5 and is strongly dependent on the HV. If all dynodes have the same gain, the overall gain of a PMT with N dynodes is [76]:

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

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

The multiplication stage adds an uncertainty in the measurement. Working without gain allows to count the number of photons that reach the PMT. This can be done by short-circuiting all the dynodes and the anode and collecting the signal directly from the photocathode. This special setup was used for fiber characterization, described in section 4.1.3.

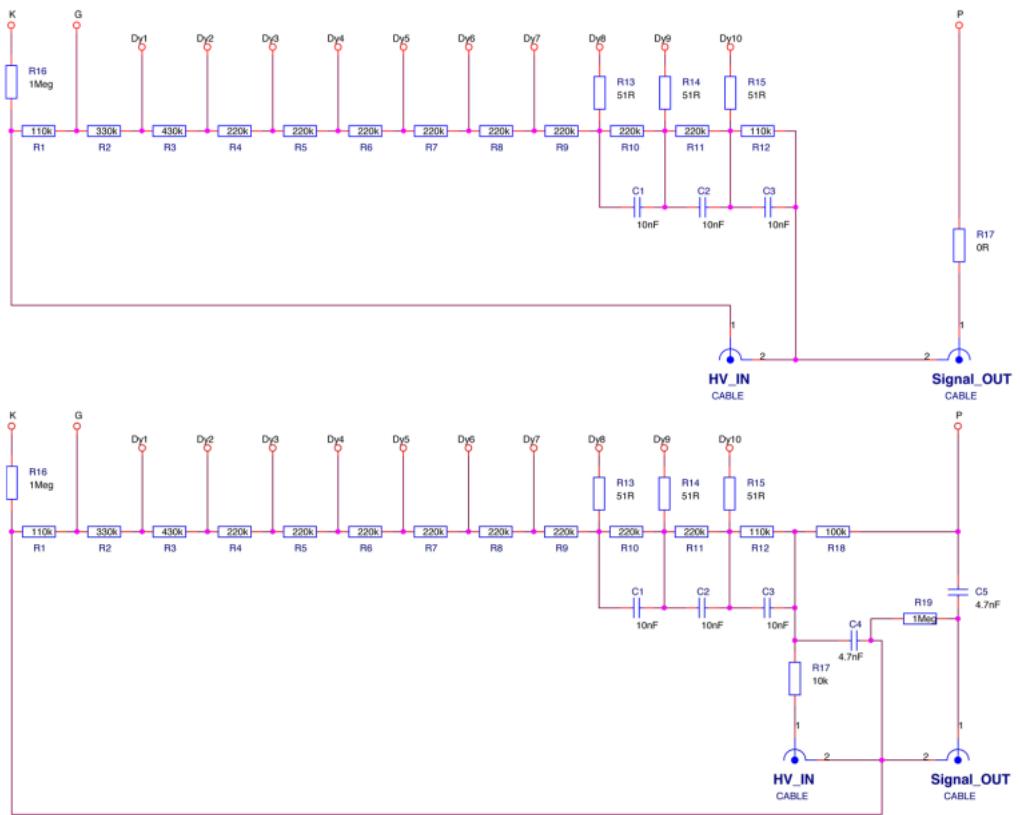


Figura 3.9 – Hamamatsu commercial voltage divider electronic circuit. Upper circuit with negative supply and lower circuit with positive supply. [81]

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

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

The photocathode may emit electrons without any scintillation

light. This signal is named dark current, I_{DC} , and can arise due thermoionic emission and, for the PMTs used, this value is around 2 nA according to their data sheet.

The characterization of the PMTs used for dark current, gain for several HV and quantum efficiency, was done at IFIC in the framework of NEXT experiment [82].

Silicon Photoelectron Multiplier Array (SiPMs array)

The Silicon Photomultiplier (SiPM) are a bind of photosensor, based on semiconductor materials, developed in recent years. They are replacing progressively conventional PMTs in many experiments and applications. They achieve outstanding photon-counting capabilities with high gain and high photodetection efficiency comparing to PMT. They have convenient characteristics as insensitiveness to magnetic fields, low operating voltage and compactness.

SiPM are based on p-n junctions, made with special techniques to achieve a good contact between both surfaces.

The voltage at which the SiPM changes from proportional to geiger mode is called the breakdown voltage, V_{BR} . At a lower voltage it works in proportional mode but at a higher voltage, it switch to Geiger mode. The measurement of the breakdown voltage is one of the most important parameters to characterize the SiPM and its determinations is described in section 4.2.

The SiPM, formed by a matrix of APDs which are photodiodes operating in Geiger mode. A scheme of an APD is shown in Figure 3.10. It has p+ and a n+ layers.

These APDs, called pixels when they are part of a SiPM, are con-

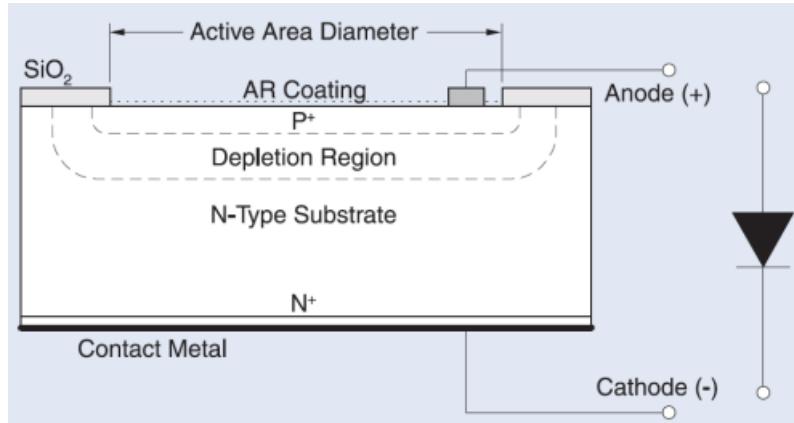


Figura 3.10 – Scheme of a APD and electrical symbol used. [84]

nected in parallel and the sum of all of them is read. The output signal of the pixels are 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. The energy deposited in each APD is not known but, as all SiPM pixels are read at the same time, the charge of the output signal when n photons are simultaneously detected is n times the charge of a single photon, as can be seen in Figure 3.11. Due to this property, after a correct calibration of SiPMs the number of detected photons we have detected is linearly related to the output signal.

As the number of scintillating photons is proportional to the deposited energy, the linearity of its output signal and the deposited energy is obtained.

On top of that, these pixels need to be so small⁷ that, if the photon density to be detected is low enough, we only detect one photon in each pixel. If it doesn't happen, we will detect two or more photons with the same pixel but the output signal will be the same as one detected photon, so we will have a loss of linearity of our output signal. This effect is known as

⁷Pixel sizes for commercial SiPMs are 50 or $75\mu\text{m}$ [85], [86]

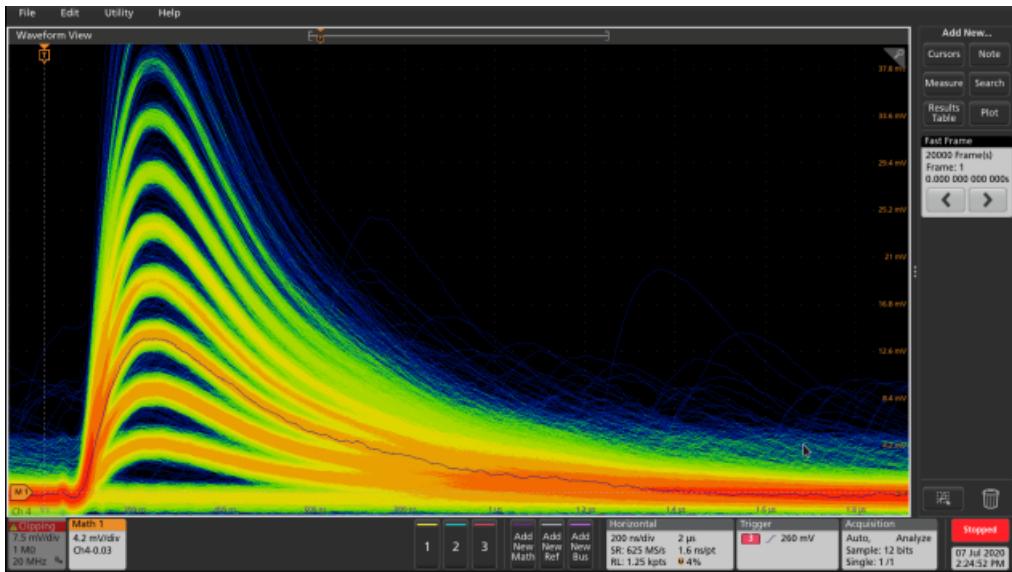


Figura 3.11 – Using persistence on the oscilloscope to show several pulses with different heights. Each height associated with a different number of SiPM pixels lit at the same time.

saturation and it is important to know the photon density at which it happens for our SiPMs. The experimental measurements of this effect, which have been done for our SiPMs, is shown in section 4.2. SI LA MIDO YO PERFECTO, SI NO DECIR QUE PARA NEUSTRO CASO NO ES IMPORTANTE PORQUEU ESTAMOS MIDIENDO MUY POCOS FOTONES POR EVENTO.

Each of these pixels has a quenching resistance⁸ in series that is used to stop the current produced when this pixel has detected a particle. It is used for limit the current drawn by the diode during breakdown and reduce the reverse voltage seen by the diode to one below the breakdown voltage. After that, the voltage seen by the diode is reset to the bias voltage and this pixel is ready to detect a new particle again. In Figure 3.12 (left) a diagram of these chenching resistances and APDs in a SiPM and (right)

⁸The tipical valuer of this quenching resistance for commercial SiPMs is around 500 kΩ

how it works is shown respectively.

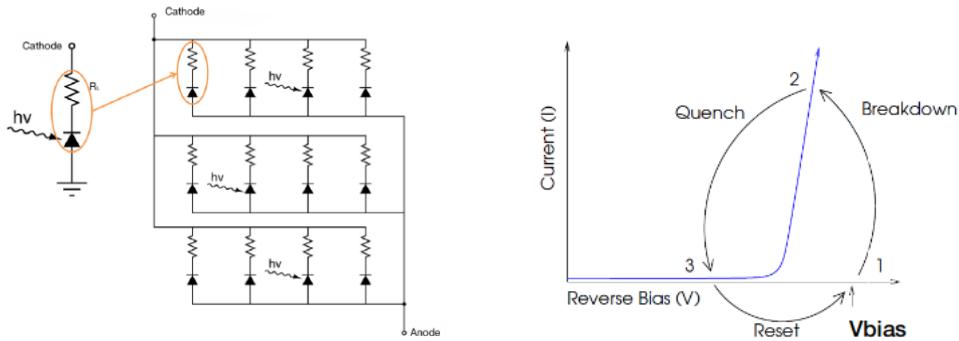


Figura 3.12 – (Left) Electronic scheme of a SiPM and (right) output current of a SiPM as a function of the reverse voltage. It show that the quenching mechanism is essential for working with SiPMs [89]

In this simple electrical scheme we can see that all pixels have a common cathode and anode which means that, as we said before, they are at the same bias voltage and the output is the sum of all of them.

We have a lot of names to refer to these photosensors such as SiPMs, MPPCs, G-APDs, SSPMs, MRS-ADPs or AMPDs. The candidate for TRITIUM project is S13360-6075 from Hamamatsu photonics [86] because its characteristics are the ones that best fit our objectives since this model has super low afterpulses, crosstalk and dark counts than other SiPM models from Hamamatsu. Its characteristics and properties are shown in Table 3.3.

These characteristics and properties will be explained and their experimental measurements will be shown in section 4.2. These numerical values, which appear in Table 3.3, are provided by Hamamatsu photonics but it is only an approximation for this model. These parameters must be determined experimentally for each SiPM used because it can be very different even if it is the same model.

It must be taken into account that we will do this characteriza-

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

Table 3.3: Characteristics of SiPM S13360-6075 from Hamamatsu Photonics [86].

tion at the level of a single SiPM because, at the beginning, it is easier to understand the results but we will work with a matrix of them and we will have to do this characterization for each matrix used.

The matrices under consideration are the model "S13361-6050" from Hamamatsu, which consists of a 4×4 SiPM matrix where the active area of each SiPM is 6×6 mm [87] or the model "S13361-3050" from Hamamatsu, which consists of a 8×8 SiPM where the active area of each is 3×3 mm [88]. They are commercial matrices from Hamamatsu and, as you can see, the total active area that we will cover with these arrangements is the same in both cases, 24×24 mm and it is approximately the same that the active area covered with the PMTs used, which has been shown in the previous section.

These matrices have a common bias voltage and common ground for all SiPMs that are contained and we will have an output signal for each SiPM.

We hope to obtain better results with the 4×4 matrix for theoretical reasons which we will see in section 4.2 like larger PDE, mainly due to a larger active area but it is something that we will have to verify with experimental measurements.

Nuestro SiPM esta dopado? con que?

Comparison of photosensors considered

As we have said before, we are going to use two of the most widely used photosensors in the world, PMT and SiPM. Each has some properties that are better than the other for our experiment and its own problems. We will have to test both and choose the one with which we achieve better results.

The output signal of both photosensors used is proportional to the

number of incident photons and they have a similar internal gain (of the order of 10^6). Both properties are essential for our experiment in order to detect tritium events and obtain a signal large enough to be measured and processed.

They have fast output signals, whose rise time is shorter than nanoseconds, and a wide spectral sensitivity that is similar for both ([200 – 800] ns for PMT and [300 – 900] ns for SiPM).

The supply voltage necessary to work with SiPM, on the order of tens of volts, is much lower than that of PMTs, which require high voltage, that is, on the order of thousands of volts and the PDE at 420 nm, achieved with SiPM is higher, around 50%, than PMT, whose PDE is around 30%. A large PDE is essential because, as we have seen before, the number of photons that we will read in each tritium event will be very low, so we must detect as many photons in each event as we can.

Furthermore PMTs, due to the reason that they consist of a vacuum tube, are more bulky and fragile than SiPMs, which are compactness and robust. It is an advantage of the SiPMs because we want that our detector work during a lot of years so we need this to be durable. Furthermore, because PMTs are manually produced, they are much more expensive, thousands of euros, than SiPMs, tens of euros, which can be mass produced.

On top of that the behavior of the PMTs is affected by magnetic fields, something which doesn't happen with SiPMs with which it has been tested that it can work correctly with magnetic field intensities between 0 and 7 Tesla.

In addition to that, due to their enormous uniformity, SiPMs are capable of distinguishing the exact number of photoelectrons detected and even resolving a spectrum of a single photoelectron, which is not possible with PMTs due to variations in their gain.

On the other hand, the dark current rate for PMTs is much lower (a few counts per second) than for SiPMs, whose dark current rate is between 0.1 and 1 Mcps⁹ (depends on its size) and it happens almost entirely at the level of a single photoelectron. It is a problem of the SiPMs because we need to distinguish the tritium signal from this background. In addition to that, SiPMs have other properties, such as the crosstalk of the afterpulses, that must be measured and extracted since they can affect the correct measurement. We will see how to do it in section 4.2

Also, the SiPMs output signal is affected by a slight change in temperature, something which doesn't happen with PMTs. It is a serious problem of the SiPMs for our experiment because we will work in the field, where we cannot avoid such a low temperature change. As we will see in section 4.2 we will solve this problem with a suitable change in the supply voltage that compensates for this variation.

3.2.4 Electronic Readout

The last part needed to have a complete scintillator detector is the electronic system. This is the part in charge of reading, processing and analyzing the output signal of the used photosensor and providing useful information.

This electronic system will depend on the type of the output signal of the detector, which means that it will be different for the different detector configurations tested, PMT and SiPMs.

Electronical system for PMTs

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

⁹Mega counts per second, 10^6 c/s

reached the PMT photocathode, which can be interesting, for example, to characterize fibers, and, on the other hand, they were used to know the energy of each event detected, which can be interesting, for instance, to obtain an energy spectrum or to discriminate events based on their origin.

In the first case, to know the amount of photons that have reached the photocathode, the PMT should work without the internal gain since it introduces a large uncertainty in the measurement. To do so, the use of the electron multiplication stage (shown in section 3.2.3) must be avoided. It is achieved with a special PCB, shown in Figure 3.13, which was designed, built and tested.

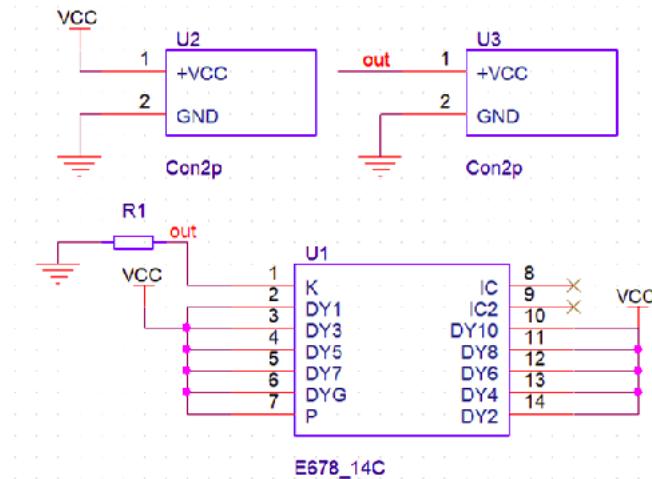


Figura 3.13 – Electronic scheme of the electronic voltage divider circuit used for working with PMTs without its internal gain.).

This PCB relies on a short-circuit all the dindes and read the signal directly from the photocathode. It is designed to be powered with positive supply voltage which is less than the normal situation [0 – 400 V]. This is because it is not needed to create a voltage difference between each pair of dynodes in the chain (we only need to create a voltage difference between the photocathode and the first dinode).

Now, the output signal of our photosensor is very small (currents of the order of tens of nanoamperes¹⁰) since it is not amplified. Therefore, to analyze these types of currents, a special system is needed. The chosen system is Keithley 6487 Picoammeter/Voltage Source [90], which is a commercial system from the Keithley company. It has some interesting options for this study such as automatic baseline correction, the ability to read signals as small as picoamps and the ability to perform some interesting mathematical operation on the signal, such as the average of N measurements with the associated statistical error, where N is programmable by the user ($N = 100$ in all our studies).

This is the configuration used to measure the output current of our photosensors. The amount of photons that has reached the photocathode is calculated using the equation 3.10:

$$N^o \gamma / \text{sec} = \frac{(I_{PMT} - I_{DC})}{q_e \cdot QE \cdot CE} \quad (3.10)$$

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

In the second case, to know the energy of the event that occurred, the internal gain of the PMT is needed since it enlarges the signal (usually a factor of the order of 10^6) which facilitates its processing and analysis. For that the electron multiplication stage shown in section 3.2.3 is used.

In all the studies done, the number of PMTs used are one, two or four, depend on the case. A simplified scheme of the electronic chain

¹⁰1 A = 10^9 nA

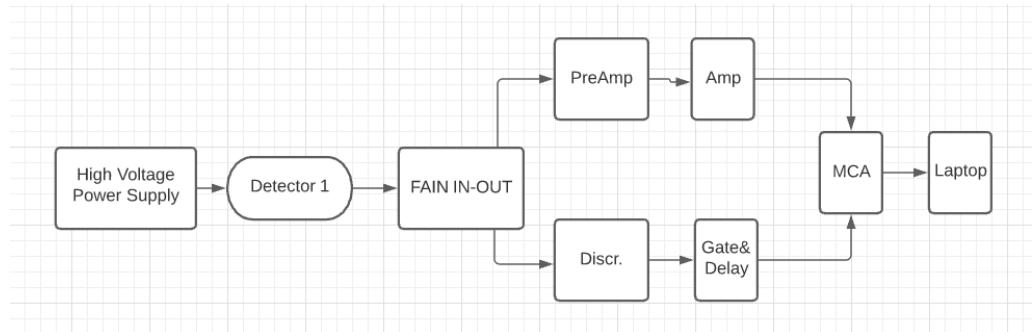
configurations used in each case is shown in Figures 3.14a, 3.14b and 3.14c respectively, which are based on various NIM technology modules¹¹.

The PMTs used in each situation are feeded with the voltage supply "TC 952 High Voltage Supply" from Tennelec [91]. It has four channels for feeding up to four different PMTs. If two or more configurations are needed at the same time, a second voltage supply was used, whose reference is "HV Power Supply N 1130-4" from "Wenzel Elektronik" company [92] with 4 additional channels. The high voltage used in each case will be named in the appropriate section.

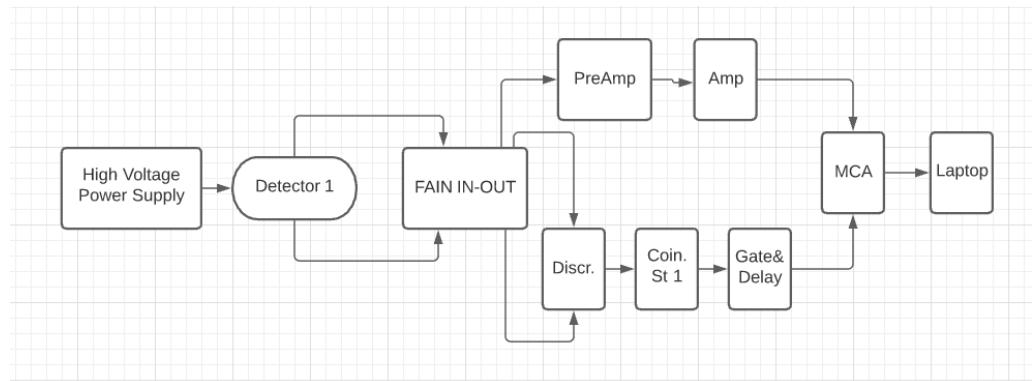
As it can be seen in the three Figures, there are two different lines followed by the output signals of the PMTs, the amplification line and the time coincidence line. Therefore, the first module needed is an analogic FAN IN-OUT module which is used to duplicate the input signal. The module used is the "Quad linear FAN IN-OUT MODEL 740" module from the company "Philips Scintific" [93]. It has four different channels and 4 output signals can be obtained from each one, which are identical to the input signal. One of these output signals is used as the input for the amplification part and another is used as the input for the time coincidence part.

- The amplification line, which is the same for all three configurations, is used to process and amplify the output signal. It contains the energy information and is based on two steps:
 1. One of the output signals of the previous module (FAN IN-OUT) is introduced in a preamplifier, which integrates it, resulting in an output signal the height of which corresponds to the charge of the

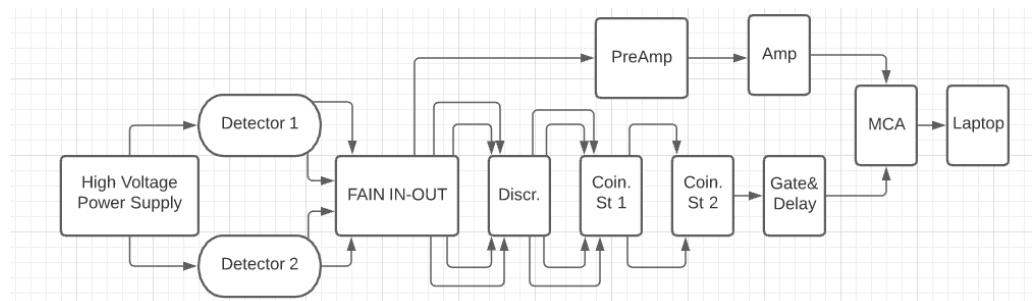
¹¹The Nuclear Instrumentation Module (NIM) is a standard specification convention for electrical and mechanical parameters defined in electronic modules used in experimental nuclear and particle physics.



(a) Electronical configuraiton scheme used to measure with one PMT.



(b) Electronical configuraiton scheme used to measure with two PMTs.



(c) Electronical configuraiton scheme used to measure with four PMTs.

Figura 3.14 – Schemes of the configuraiton of different electronic chains used to measure with PMTs.

input pulse. It has a long tail¹² due to the use of a capacitance. The preamplifier used is "MODEL 9326 FAST PREAMP" from ORTEC [94].

2. The output signal from the preamplifier is introduced into the amplifier which integer the previous singal. The output singal of it has a shape close to the Gaussian function and it is amplificated by a configured factor. The amplifier modules used are "model 575A" and "model 671", both from ORTEC company [95], [96]. An example of the output signal for 575A module is shown in Figure 3.16, green color.
- The time coincidence line, which contain the time information. It is used to obtain the coincidence gate that indicates when we have to save the amplified signal (when all the output signals of the used PMTs are in time coincidence). This line has little difference in all three configurations and it consists of:
 1. One of the output signals of the FAN IN-OUT module of each PMT are introduced into a discriminator module where we obtain an output logic signal with height of -1.2 V and width of 240 ns when a threshold, the one programmed by the user, is exceeded. The discriminators used in our experiments are "Octuple Constant-Fraction Discriminator CF8000" module from ORTEC company [97] and "4 channels discriminator model 84" from CAEN company.
 2. Now the time coincidences are made. It is used to ensure that the detected event comes from the scintillating fibers and to remove other events such as external light or dark current events. As it is shown in section 3.4.2 and chapter 5, each used detectors cointain up two PMTs so time coincidences is done in pairs of

¹²The 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.

photosensors. Due to that, this step is not possible to be applied when only one PMT is used (configuration 3.14a).

Two output logic signals of the discriminator module (which comes from two PMTs that are in the same detector) is connected to a channel of the coincidence module which generate an output signal, with a height of -1.4 V and width of 20 ns, when both are in time coincidence.

The time coincidence modules used are "Coincidence Unit Model 465" from LeCroy [99] and "Coincidence Type N6234" from CERN-NP [100].

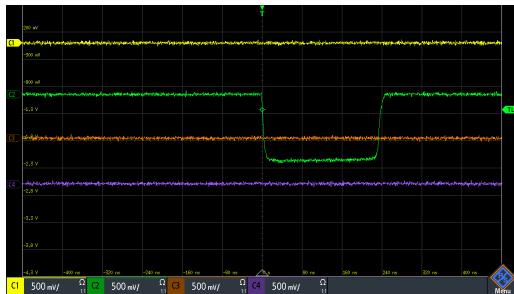
3. An addition step of time coincidence is included when time coincidence between two different detectors (4 PMTs, configuration 3.14c are done. It could be interesting, for example, to detect hard cosmic radiation as we is explained in section 3.4.2.

To do so, a new coincidence step similar to the previous one must be applied. Therefore, two logical output signals of the previous step (coincidence module) are used as input of a second coincidence stage. The output signal of this case has the same parameters, logical signal with a height of -1.4 V and width of 20 ns. The modules used is the same as before.

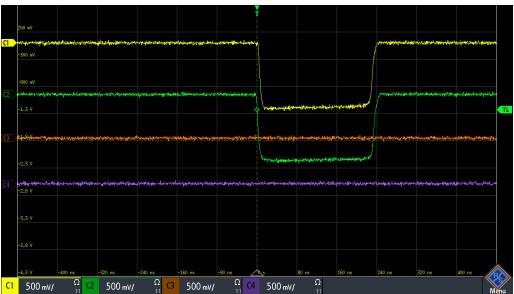
Some interesting cases are shown in Figure 3.15 when time coincidences are made in two detectors, 4 PMTs. There, four logical signals are shown, two of them (channel one and two, yellow and green respectively) come from two PMTs connected to one detector and the other two signals (channels three and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

- (a) In Figure 3.15a only one PMT (channel two) has detected an event. It means that the event detected doesn't come from the scintillator. In this case, the output from coincidence module, stage 1 is not generated and this event is not saved.

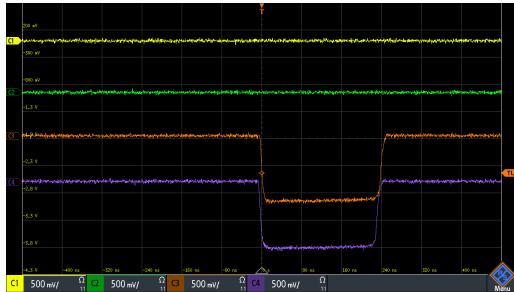
- (b) In Figures 3.15b and 3.15c two PMT signals connected to the same detector have detected an event but the others have not. It means that this event was only detected by one detector. The first coincidence stage will create a output singal but the second not so this event is not saved.
- (c) In Figure 3.15d all signals have detected the event, which means that it is an event that has been detected for both detectors. In this case, a the output signal of both coincidence stage is generated and the event is saved.



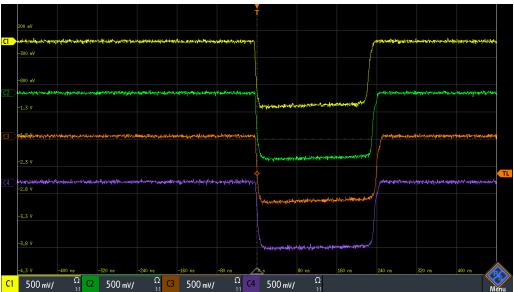
(a) Event detected in only one PMT.



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



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



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

Figura 3.15 – Different situation that can happen when we do time coincidences with PMTs.

4. Finally, the logical output signal of the last coincidence step, is introduced in the "Gate and Delay Generator", model 416A of

the company ORTEC [101]. With this NIM module we obtain a positive logical signal, shown in Figure 3.16, orange color, with a height of 8 V and width of 2 μ s.

At the end, two different output signals are obtained, shown in Figure 3.16, which will be introduced in the MCA 8000D, Pocket MCA from AMPTEK company [102] to be saved. On the one hand the analog signal (output from the amplifier module), which has information about the energy of the event and this is the signal whose information we will save for analyzing. On the other hand the logic signal (output from the Gate and Delay Generator module) that indicates when the amplified signal must be saved.



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

The MCA is a module used to save the signal information as a histogram. The histogrammed variable is the height of the input pulse of the signal (green signal), which, in the case of the electronic configuration used, corresponds to the energy of the event. It is performed

when the value of the signal gate (orange signal) is greater than 4 V, which indicate that it is an interesting event.

Electronical system for SiPMs

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

The electronic system used to process and analyze the output signals of the SiPM arrays is PETSYS [103], which is a commercial system prepared to work with SiPM matrices from Hamamatsu.

Petsys system, Figure 3.17, is a complete acquisition and digitization system that is capable of working with up to 1024 SiPM. It consists of a basic board, which processes the signal, to which 8 different SiPM matrices can be connected with up to 64 SiPM per matrix. This number of channels is needed in the TRITIUM project because, as it is shown in section 5.3, the TRITIUM monitor use dozens of SiPM matrices with 16 channels (SiPMs) per matrix.

Although the capacity provided by PETSYS should be enough for the requirements of the TRITIUM project, TRITIUM is a modular detector with scalable sensitivity. It means that, if an improvement of its achieved limits is needed (improve its sensitivity or further reduce the background) more photosensors are needed. Therefore, the electronic system used should be able to increase its capacity in a scalable way. This requisit is fulfilled by PETSYS since it has an additional module, called Clock and Trigger, with which up to sixteen different PETSYS basic boards can be connected. These sixteen Petsys basic boards are read in parallel, each with the capacity previously mentioned, giving a total system capacity of 256 SiPM matrices



Figura 3.17 – Different parts of PETSYS system. [103]

(16384 SiPMs¹³).

PETSYS is based on C++ and Python scripts that are prepared for the main tasks required for our experiment, such as time coincidence options between SiPM (or even SiPM matrices) or energy discrimination. It is open source, giving the possibility to modify the current scripts or develop others with additional functions.

This system has a time resolution of 250 ps which is one of the best time resolutions of commercial systems available today and its price is around 10€/ channel, which is a cheaper price compared to current similar electronic systems.

As we will see in section 4.2, the temperature of each SiPM matrix is an important parameter to take into account. The PETSYS system has the ability to monitor the temperature of both, the SiPM matrices and ASICS that are used to control them during the measurements. It is an important parameter to ensure the correct functioning of both, the

¹³ $1024 \cdot 16 = 16384$

photosensors and the system, and it offers the possibility of developing new function scripts that allow to implement the stability gain method shown in section 4.2.

Although TRITIUM monitor use SiPM matrices it is important to start the characterizaton at the level of a single channel (only one SiPM) since its reduce the uncertainties in the first results.

In order to do so, an electronic system was designed, developed and built with which we can read up to eight different SiPMs and it has the capacity to monitor the temperature.

This system is based on three different PCBs¹⁴, shown in Figure 3.18 and whose electronical schemes are shown in the appendix B, the output signal of which is connected to an oscilloscope to process and analyze the signal:

1. The first PCB, shown in Figure 3.18a, is used to organize the SiPMs and sensor temperature in this system. 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 "" company, that has a high degree of light tightness. This black box has a small hole, whose diameter is 1 mm, prepared to introduce an optical fiber¹⁵ with which the SiPM can be iluminated with a incoherent light source. The light source used is a LED, model 430L from Thorlabs company [105], whose emission spectrum is shown in Figure 3.18d, which has been experimentaly measured using a spectrometer and fitted to a Gaussian function. We can see that the emission peak of this LED is produced at 436.3 with a FWHM¹⁶ of 19.1 nm. With this LED we intend

¹⁴PCB, Printed Circuit Board

¹⁵The optical fiber used is BCF-98 from Saint-Gobain company [104]

¹⁶The FWHM parameter, Full Width at Half Maximum, of a Gaussian fit can be

to simulate the light emission of the fibers used in the TRITIUM experiment to calibrate the SiPMs at the working wavelength.

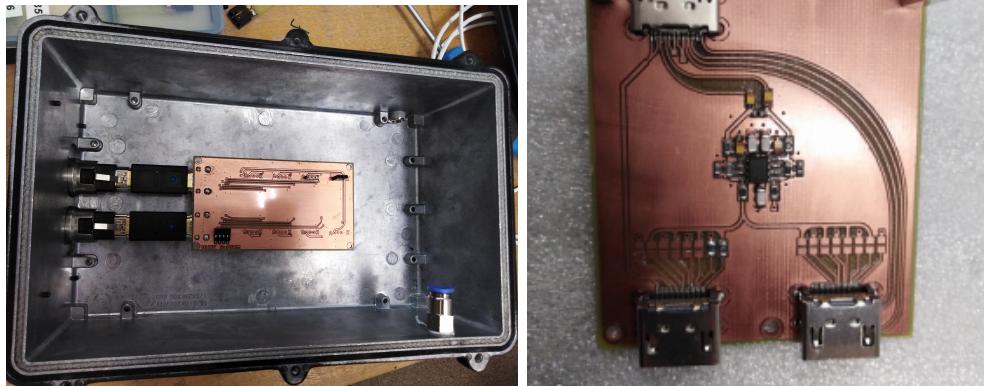
2. The second PCB, shown in Figure 3.18b, is used to sum the different signals of the SiPMs and amplify them by a factor $G = 4187.5$ or $G = 10761.88$ depends on the input resistance oscilloscope, 50Ω or $1 M\Omega$, respectively. This PCB uses a differential amplification with which the electronic noise of the system is reduced and it is connected to the first PCB through two HDMI feedthroughs.
3. The third PCB, shown in Figure 3.18c, is used to arrange all the different input and output signals of this system in an HDMI connection without introducing electrical noise by crosstalk between different signals. This PCB is connected to the second PCB through a HDMI feedthrough.

The input signals of this system are the supply voltage of the SiPMs and the supply voltage of the PCBs (± 6 V) and the output signals are the temperature sensor signal and the summed signal of all SiPMs.

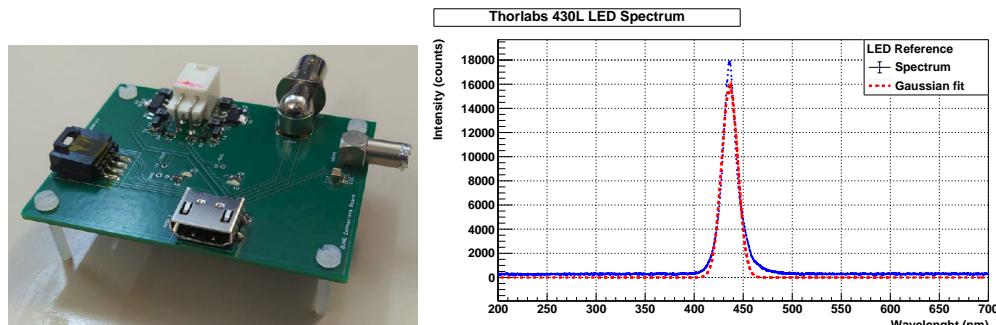
The output signal of the third PCB is connected to an oscilloscope, model ..., where the signal are processed and the data is saved and, subsequently analized by ROOT¹⁷.

calculated from its sigma using the equation: $\text{FWHM} = 2.35 \cdot \sigma$

¹⁷ROOT is a framework for data processing, based on C ++ and object-oriented technology, developed at CERN and widely used in nuclear and particle physics.



(a) PCB 1 used to arrange 8 SiPMs and black box.
(b) PCB 2 used to sum and amplify the output signals of SiPMs



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

(d) Emission spectrum of the LED.

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

3.3 Ultrapure Water System

3.3.1 Introduction to the Ultrapure Water System

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

- On the one hand, it is important because the mean free path of tritium electrons in water, shown in section 1.2, is around $5 \mu\text{m}$ and even less for solid materials like organic material. The electron from the tritium decay must reaches the fiber to be detected, so the detector must be kept very clean. If the analyzed water sample contains particles that can be deposited on the fibers, it can form a layer of matter, which prevents the tritium electrons from reaching the fibers, reducing drastically the tritium detection efficiency until it becomes impossible to measure tritium.
- On the other hand, as it is shown in chapter 5, the tritium monitor does not have any spectrometric capabilities that can be used to distinguish other radioactive elements from tritium. That means that, all the radioactive element included in the analyzed water sample will be computed as a tritium event.

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

In summary, the ultrapure water system is used to keep our detector clean, ensuring the stability of its detection efficiency and to eliminate all radioactive particles other than tritium, maintaining the activity of the tritium in the sample. Both reasons has been tested with experimental measurements, shown in secton 4.3.

3.3.2 Set Up of Ultrapure Water System

The main objectives of this water treatment device are:

- Obtaining a high degree of purification in the processed water sample, reducing its conductivity by approximately two orders of magnitude (from $1000 \mu\text{S}/\text{cm}$ to $10 \mu\text{S}/\text{cm}$)
- Designing of a device with low maintenance (low cost and low man-power)
- Installation of a remote control devices based on probes and valves and development of a software to management it.

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

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

1. First, the raw water from the Tagus River passes through two different filters, the first formed by Silex-Anthracite and the second by granate, with which a gross filtering is made (the largest particles are eliminated). This system has two parallel lines with this first stage which are capable of self-cleaning by injecting ultrapure water in the opposite direction.
2. Next, the outlet water sample of the first stage passes through a $20 \mu\text{m}$ filter (Formed by a synthetic mesh) and activated charcoal filters (one per line in both) that form the fine filtration stage. With the $20 \mu\text{m}$ filter the particles with diameters of up to $20\mu\text{m}$ are filtered and, with the activated charcoal filter, the chlorine and iron particles are removed.

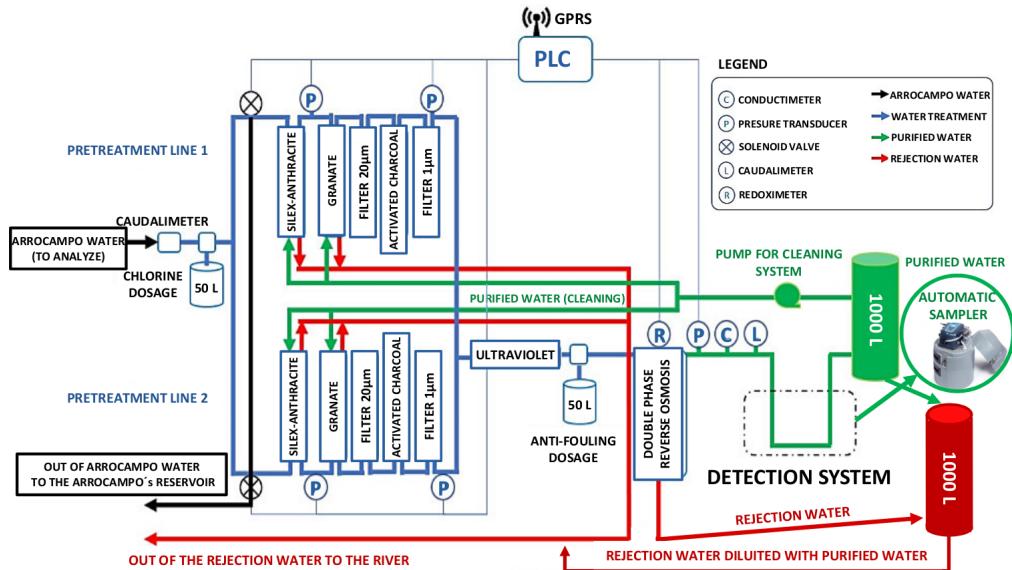


Figura 3.19 – Scheme of water purification system.

3. Then, the outlet water sample of the second stage passes through the super-fine filtering consisting of a $1\ \mu\text{m}$ filter, formed of a dense polypropylene mesh, (one per line), 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, this water sample is introduced in the last stage, double-phase reverse osmosis, thereby reducing the conductivity of the water to values of $5\ \mu\text{S}/\text{cm}$. It was verified that a conductivity of $10\ \mu\text{S}/\text{cm}$ is achieved with only one module reverse osmosis, enough for the needed conditions of tritium detector. Therefore, just one module of reverse osmosis is used for 24 h and the other for another 24 h, reducing the power consumption of the system.

At the end of this system, each water sample is divided into two different outlet samples. The pure water sample, which is the ultrapure water that is introduced into tritium detector, and the rejection water, whose con-

ductivity is even greater than the water sample before treatment because it contains all the particles that have been extracted from the ultrapure water sample.

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

The software used for remote controlling the ultrapure water system is Siemens PLC, with which it can be controlled and the information about its status such as the state of the valves, the pressure probes or water production is obtained in real time.

The appendix C contains several photos about each part of this system, already installed in Arrocampo dam.

3.4 Background Rejection System of TRITIUM Monitor

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

The total uncertainty of the measurement is a quadratic sum of all the different uncertainties present in this measurement, which is mainly the

statistical uncertainty, σ_{st} , (due to the statistical nature of the radioactivity process), the systematic uncertainty, σ_{si} , (due to the manufacture of the detectors), equation 3.11.

The background rejection system of TRITIUM monitor is used to minimize the statistical component. Because of the Poissonian nature of the process, the statistical uncertainty corresponds to the square root of the measured activity, A_m , equation 3.11, which can be reduced by minimizing detected background events.

$$\sigma_T^2 = \sigma_{st}^2 + \sigma_{si}^2; \quad \sigma_{st;bac} = \sqrt{A_{m;bac}} \quad (3.11)$$

The background events that affects the tritium detector is due to natural radioactivity, which is present in all parts of the earth. They can be divided in two different parts, depending on their origin. On the one hand, it can come from radioactive elements that are present in the Earth since its formation, which can be divided in four different natural radioactive series, shown in Table 3.4. On the other hand, it can come from natural radiation received from extraterrestrial sources, called cosmic radiation. It is composed of high-energy particles, mainly protons and α , which, after interacting with the particles in the Earth's atmosphere, generate a shower of muons, photons and neutrons mainly.

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

Table 3.4: Classification of natural radioactive series [106, 107].

Natural radioactivity depends on several parameter like the alti-

tude and latitude of the Earth (the volume of the Earth's atmosphere, with which cosmic rays interact, is different), the height (for the same reason), sea level in our case, and the solar activity cycle (due to the relative position of the earth in the universe). The spatial distribution of cosmic rays, mainly muons, follows a $\cos^2(\theta)$ distribution with the zenith angle.

To remove the effect of this natural radioactivity it is divided into two parts and different techniques are used to prevent these events from affecting the tritium measurement:

- On the one hand, the weak radiation, which is any radiation whose energy emission is below 200 MeV/nucleon. The technique used to avoid that these radiation affect the tritium measurement is to stop them using a lead shield, explained in section 3.4.1,
- On the other hand, the hard radiation, that is any radiation whose energy emission is greater than 200 MeV/nucleon (mainly cosmic radiation). It is much more difficult to stop so, instead of stopping them, the technique used is to build a cosmic veto, explained in section 3.4.2, with which each hard cosmic event is detected and used in anti-coincidence with the TRITIUM detector.

3.4.1 Passive Shield (Lead)

A passive veto is used to stop the weak radiation, which consists of a lead shielding inside which the TRITIUM detector is placed. This lead shielding is used to stop external particles before they reach the tritium detector, affecting the tritium measurement and it is effective for particle energies below 200 MeV/nucleon, which is mainly the earth's natural radioactivity and the weak component of cosmic radiation.

This lead shielding consists of 158 lead bricks with ultra-low in-

strinsec radioactivity, whose thickness are 25 mm. They are shaped like a chevron, shown in Figure 3.20a, specially designed for a perfect fit and easy assembly. As can be seen in Figures 3.20b and 3.20c, these lead bricks are arranged in two layers leaving a total thickness of the lead shielding walls of 50 mm. The space between two lead bricks in the inner layer is overlaid with a lead brick of the outer layer to avoid possible entry of radiation over there.

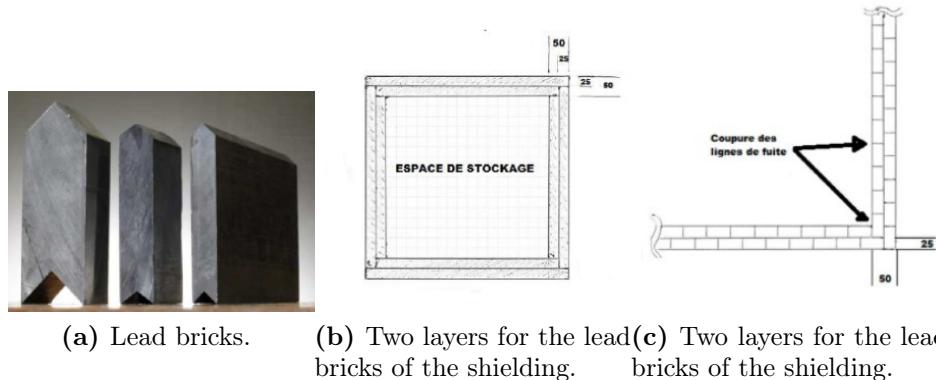
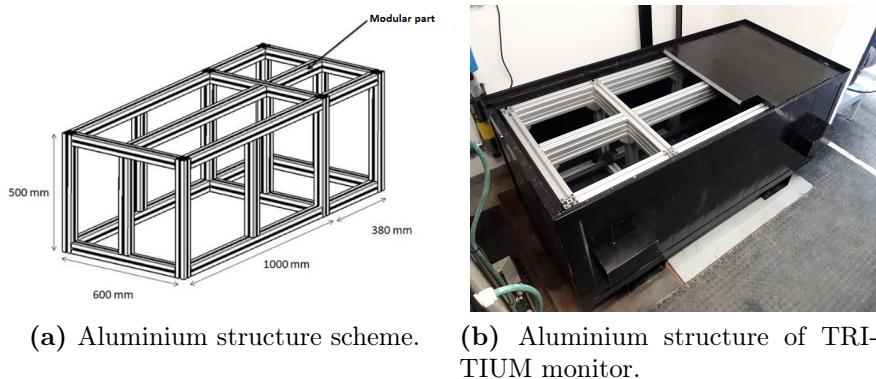


Figura 3.20 – Lead Bricks and their arrangement in the lead shielding.

Mechanical engineering department of CENBG has designed a special aluminum structure, shown in Figure 3.21, to support the total weight of the lead bricks, 2.4 tons.

The internal space of this lead shielding is divided in two parts, shown in Figure 3.20. The one, larger, has an internal dimensions of 90.5 cm long, 41 cm deep, and 51 cm high and it is used to place the TRITIUM detector. The other, smaller, has an internal dimensions of 33 cm long, 41 cm deep, and 51 cm high and it serves to place the electronic system necessary to collect and store data. The external dimensions of this lead shielding are 148 cm long, 60 cm deep and 70 cm high and a weight of 2.5 tons.



(a) Aluminium structure scheme. (b) Aluminium structure of TRITIUM monitor.

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

3.4.2 Active Shield (Cosmic Veto)

As we said before, it is most difficult to stop hard radiation (high energy particles), than weak radiation. It is because the mean free path of particles is proportional to their energy so to stop high energy particles large wall thickness would need. Therefore, instead of stopping it, the so-called cosmic vetos are used.

The cosmic veto consists of several complementary detectors, two complementary detectors for each cosmic veto in the case of TRITIUM monitor, with which the hard cosmic events that affect the tritium measurement will be detected and subtracted from the tritium measurement.

As it is shown in Figure 3.22, the way followed to do that is to place two complementary detectors, called cosmic detectors, one above the TRITIUM cell and the other below it. The distance between both detectors, 34.2 cm for our latest prototype developed, is set by the TRITIUM prototype that has to be placed between both.

The cosmic veto is placed within the lead shielding so that, the weak radiation doesn't affect them with a false hard cosmic events detected.

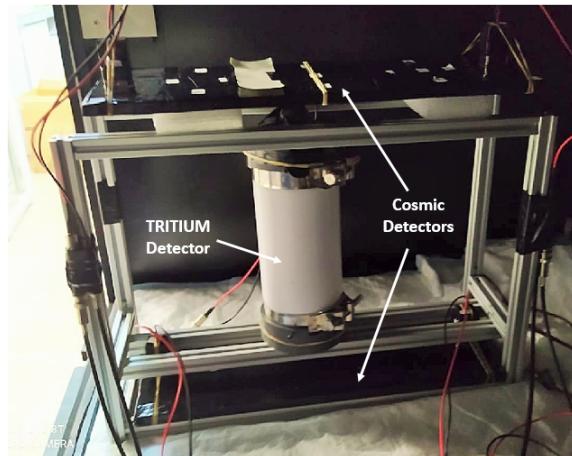


Figura 3.22 – Cosmic veto and Tritium-IFIC 2 prototype in an aluminum mechanical structure developed by IFIC’s mechanical engineering department.

A hard cosmic event that affects the tritium measurement will pass through both cosmic detectors at the same time, shown in figure 3.23a. Each cosmic detector will have two photosensors so there are four photosensors in each cosmic veto. Therefore, to eliminate the hard cosmic events, both cosmic detectors are read out in coincidence with the electron configuration shown in Figure 3.14c. Then, the TRITIUM detector is read out in anti-coincidence with the cosmic veto to save the tritium measurement only when there is not any hard cosmic event.

It is possible that this hard cosmic event detected in the cosmic veto comes from two different hard cosmic events (one detected in each cosmic detector as shown in Figure 3.23b) but it is practically negligible.

The expected hard cosmic rate at sea level for muons, which is the main contributor of cosmic radiation at this height, is $70 \text{ m}^{-2}\text{s}^{-1}\text{sr}^{-1}$ [108, 109], that is, approximately, $10^{-2} \text{ cm}^{-2}\text{s}^{-1}\text{sr}^{-1}$, shown in the cosmic rate plot of Figure 3.23c. Taking into account that time coincidences are doing with signals whose width is of the order of 10 ns, the probability of obtaining two different hard cosmic events in temporal coincidence is less

than 10^{-9} which is insignificant so they are not worth considering.

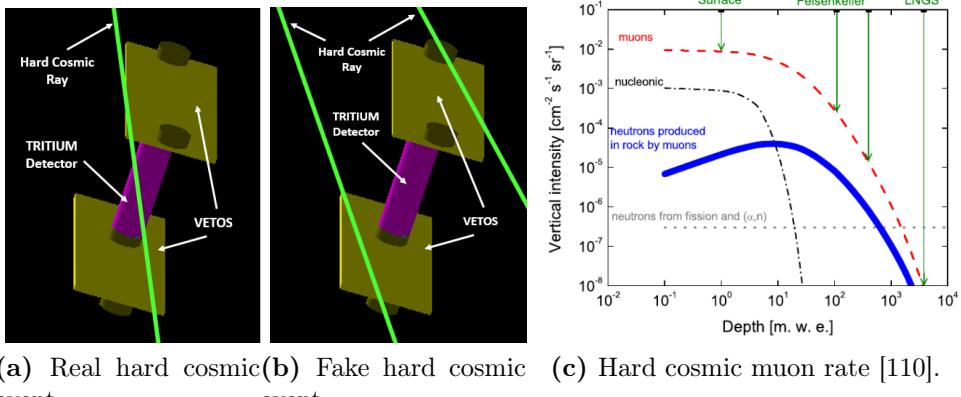


Figura 3.23 – Hard cosmic events detected with the cosmic veto of TRITIUM: a) Affecting to the tritium measurement, b) Does not affecting to the tritium measurement. c) Hard cosmic muon rate [110].

Finally, these individual cosmic detectors on which the cosmic veto is based, consist of a plastic scintillator block from Epic-Crystal [111], whose properties and energy emission spectrum are shown in Table 3.5 and Figure 3.24 respectively.

It has an emission peak very close to that of the scintillating fibers used, so the same photosensors are used to read out them. The dimensions of the scintillator block are 45 cm long, 17 cm deep and 1 cm of thickness and they are covered by three layers, teflon, aluminum and black tape, shown in Figure 3.25. These layers has two objectives. On the one hand, they are used to prevent external photons from reaching the scintillator plastic, giving false hard cosmic events and, on the other hand, they are used to prevent the photons generated by the scintillator plastic from escaping before reaching the photosensor, losing real hard cosmic events.

Two $2.5 \cdot 2.5 \text{ cm}^2$ windows are made done in this coating and used to read the photons produced by the plastic scintillator with photosensors.

Base material	Polystyrene
Growth method	Polymeric
Density (g/cm ³)	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 scintillator blocks from Epic-Crystals. [111]

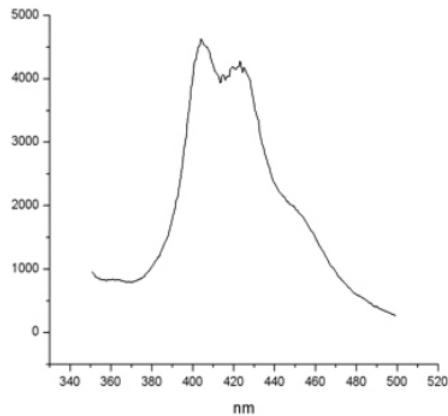


Figura 3.24 – Emission energy spectrum of the plastic scintillation used for the cosmic vetos. [111]



(a) Scintillator with- (b) Teflon coating. (c) Aluminium coat-(d) Black tape coating.
out coating. ing. ing.

Figura 3.25 – Different layers used to cover of the cosmic veto.

The expected hard cosmic rate is calculated for our specifically cosmic vetos. The expected hard cosmic rate at sea level is $10^{-2} \text{ cm}^{-2}\text{s}^{-1}\text{sr}^{-1}$, previously mentioned. Taking into account that, on the one hand, the solid angle of our detectors is $\omega = 0.5434$, which has been calculated by integrating above the area of the TRITIUM cosmic veto, and the area of its cosmic veto is $45 \text{ cm} \cdot 17 \text{ cm} = 765 \text{ cm}^2$, the expected hard cosmic rate on our cosmic vetos should be 2,909 event/s. It is an important calculation used in experimental measurements to determine the efficiency of the cosmic veto.

Chapter 4

Research & Development on Detector Design and Components

This chapter shows the characterization of each individual part of the TRITIUM monitor, including scintillating fibers, SiPMs (at the individual SiPM level and at the matrix level), the ultrapure water system and the background rejection system, consisting of the lead shielding and the active veto.

This characterization is one of the most important things to do because it will help us to understand their behaviour and the results obtained with the full monitor. Furthermore, several developments have been made to improve interesting parameters of the TRITIUM monitor components to enhance the monitor's capabilities of tritium detection.

All these studies have been carried out inside a special light-tight box, called black box, to ensure that the photons we are detecting come from the photon sources used, whether they are emitted by LEDs or by scintillators. In addition, because accurate energy calibration cannot be performed when plastic scintillators are used, most of their energy spectrums

are shown in units of ADC¹ (channels), which are linearly proportional to the units of energy.

4.1 Characterization and R&D on Scintillating Fibers

This section shows experimental measurements of the scintillating fiber parameters that most affect the tritium detection, such as its collection efficiency or their uncertainty in its conditioning process.

Thousands of scintillating fibers are used in TRITIUM detector which need to be prepared and conditioned prior to use in characterization studies or tritium detection. Therefore, various mechanical and electronical devices were developed to automatically prepare a lot of fibers at the same time.

4.1.1 Scintillating Fiber Conditioning Process.

First thing that was done in TRITIUM experiment was to choose the optimal fiber length at which the signal from the tritium events is optimized. To take this decision, it has to be taken into account that, on the one hand, long fibers are interesting because the efficiency of TRITIUM detector is proportional to the active area (proportional to the fiber length), but, on the other hand, in long fibers, scintillating photons need to be reflected in the fiber walls more times to be driven to its ends, where the photosensors are, and, because of that, some photons are lost in each reflection, deteriorating the detector signal.

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

Several simulations were performed using Geant4 [75], a particle and nuclear physics simulation package based on C++, to quantify the importance of this effect. The result of these simulations, shown in section 7.3, Figure ??, was that it is preferable to work with short fiber since a significant loss of photons was observed.

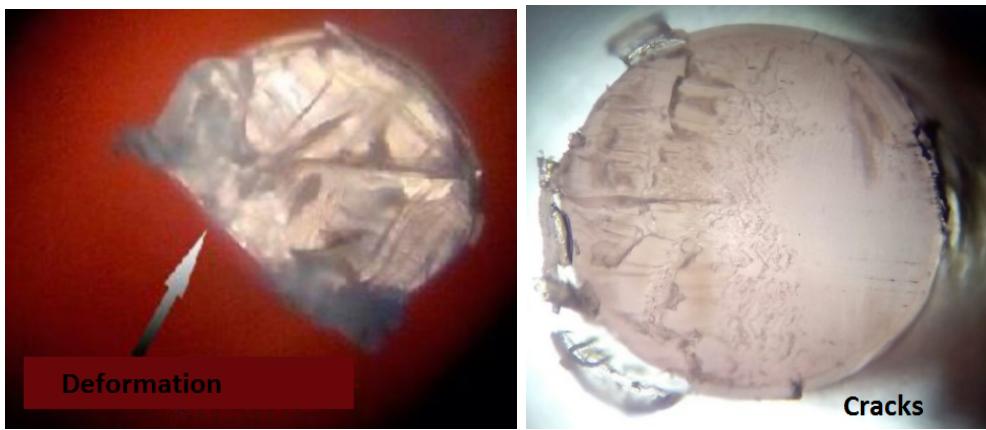
The chosen fiber length for the TRITIUM prototypes developed in Valencia, is 20 cm, which was also the length used for most of the experiments performed in the framework of the TRITIUM experiment.

Due to the length of Saint-Gobain's commercial fibers, which are 1 meter long, an effective scintillator cutting technique was to be developed. It is very important to introduce strict requirements on the cutting quality of the fiber ends since it will greatly affect the transmission of photons and, thus, the efficiency of TRITIUM monitor. This cut must be perpendicular to the fiber and with very low uncertainty in the length of the fiber, both requierments are mandatory to achieve a good coupling with the surface of the photosensor. It is also important that its final state must be as regular as possible, that means, without cracks or deformations that contribute to internal reflections, losing photons and, thus, reducing the tritium signal.

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

Many commercial devices based on blade cleaving, such as the one provided by thorlabs with a diamond tipped blade [112] or others similar to the guillotine designed for industrial fiber optics [113], were tested in an extensive study done with unsuccessful results [79]. As can be seen in Figures 4.1, it presents deformations, cracks or imperfections so the technics considered in this study don't overcome the requirements imposed.

The microscope model PB 4161 from EUROMEX company or the



(a) Fiber end deformation.

(b) Fiber end cracks.

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

Digital Microscope from Jiusion company were used to check the results in the fiber ends.

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

It consists of fourteen rails where the fibers are fixed and a thin blade, fixed on a mobile piece, which is used to cut them. The perpendicular cut, which is one of the requirements imposed, can be ensured since the moving piece, where the blades are fixed, is placed perpendicular to the fibers.

The blade used is the typical commercial razor blade, whose thickness is 0.1 mm, which is the thickness with which we obtain the best results. It was positioned with a slight inclination, 5°, with respect to the horizontal axis since it was seen in several studies that it helps to obtain a less aggressive and cleaner cut [114], [115].

Therefore, as can be seen in Figure 4.3a, with the developed device fiber ends without breaks or deformation was obtained, overcoming other

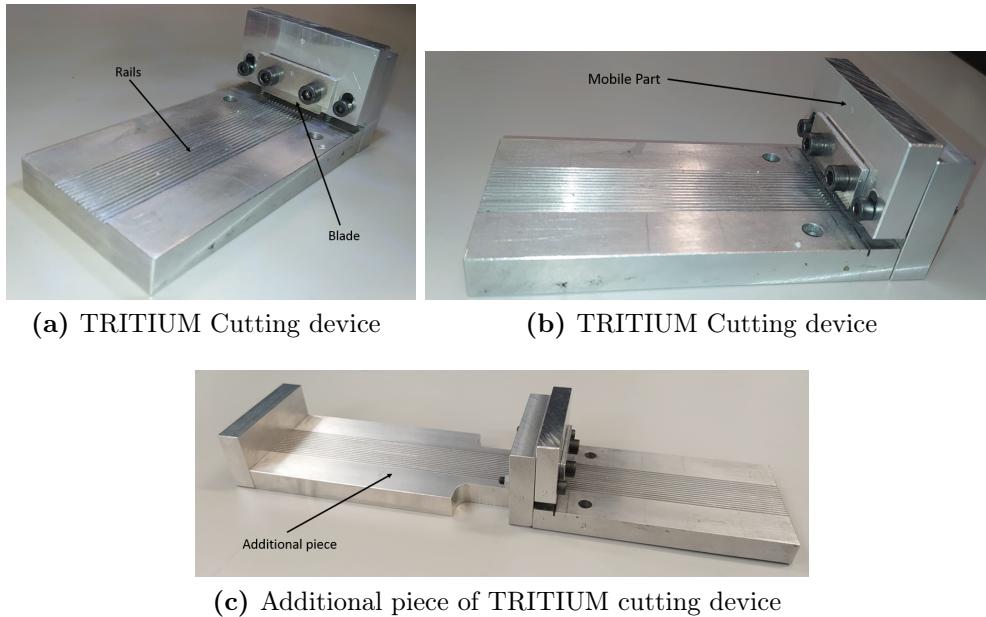


Figura 4.2 – Cutting device developed in the TRITIUM experiment and additional part to make precise measurements of fiber length.

imposed requirement.

Another important parameter that can affect the cutting quality of the fiber ends is the temperature of both, either the fiber or the blade. It was tested in a study in which both were subjected to different temperatures from room temperature (25 degrees) to 110 degrees [79]. No significant conclusions were obtained in the temperature study, so the cutting process is carried out at room temperature to facilitate the cutting technic.

To obtain a low enough length uncertainty, which is the last requirement to overcome, an additional piece was designed and built, shown in Figure 4.2c, which is used to measure the fiber. With this piece we achieve an uncertainty in the measurement of less than 1 millimeter.

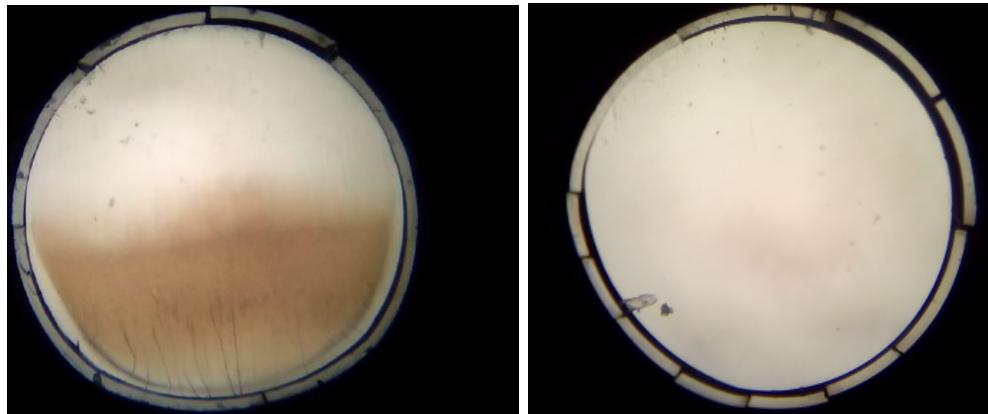
With the designed cutting fiber device we have exceeded all requirements imposed, obtaining a fiber cutting device, whose effect on the

light transmission is minimized.

In Figure 4.3a, which shows the fiber end after cutting process with TRITIUM cutting device, it can be seen a slightly darkened part at the bottom of the fiber, which is an inevitable effect of the cutting process. To reduce the effect of this imperfection, a polishing process developed by thorlabs is included [112].

This polishing process consists of using five different polishing papers, with a decreasing grain size, whose diameters are $30\ \mu\text{m}$, $20\ \mu\text{m}$, $12\ \mu\text{m}$, $5\ \mu\text{m}$ and $0.3\ \mu\text{m}$ respectively, in which we describe movements in the shape of 8 for two minutes (approximately 120 movements).

The result obtained with this polishing process is shown in Figure 4.3b. In Figure 4.3, the quality of both fiber ends, before and after polishing process, can be compared, where it can be appreciated that the darkened part has completely disappeared.



(a) Fiber end after cutting with Tritium de-
vice. (b) Fiber end after cutting and polish-
ing.

Figura 4.3 – Result of the polishing process. a) Fiber end after cutting with TRITIUM devices b) Fiber end after cutting with TRITIUM devices and polishing with Thorlabs technic.

The end of the cut fiber is completely clear after cutting and polishing, without any damage or imperfection, so both tasks, cutting and polishing, make up the conditioning process developed for each fiber before any study or its introduction into the TRITIUM detector.

4.1.2 Automatic Polishing Machine for Scintillating Fibers

At this point it has to be taken into account that, as it is shown in sections 5.2.1 and 5.2.2, hundred of scintillating fibers are used in the latest prototypes developet in TRITIUM experiment, Tritium-Aveiro 0 and Tritium-IFIC 2, which means that tens of thousands of fibers need to be prepared and conditioned for the TRITIUM monitor², section 5.3.

While this amount of fibers is not a problem for the cutting process, as it is very fast, the polishing process would be too time consuming. It takes more than ten minutes to polish each fiber, resulting in an inaccessible amount of time to prepare the needed amount of fibers (years).

Therefore, an automatic polishing machine for scintillating fibers was designed, developed, built and tested, shown in Figure 4.4. 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 increase its capacity.

As can be seen in Figure 4.4, the automatic polishing machine is based on two parts. On the one hand, the polishing table, where the fibers will be polished and, on the other hand, the electronics, based on arduino technology, used to control the movement of the polishing table:

1. The polishing table, shown in Figure 4.5a, can be divided in two parts,

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

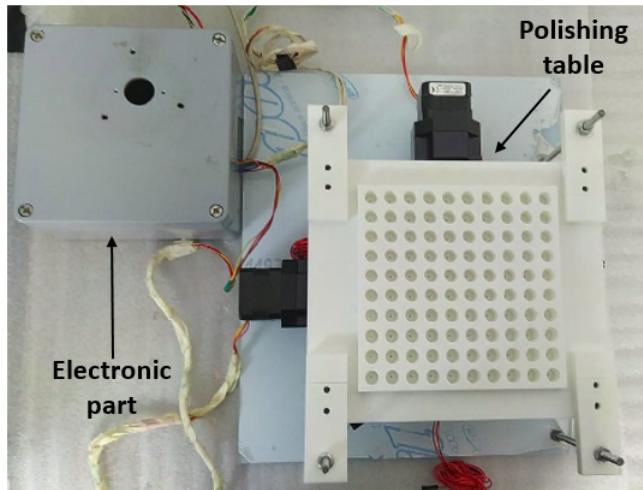


Figura 4.4 – Polishing machine developed in TRITIUM experiment.

the immobile part, where the fibers will be fixed, and the mobile part, where the polishing papers will be fixed. It was decided to move the polishing papers because they are lighter and mechanically easier than fibers.

The immobile part consists of a piece, shown in Figure 4.5a, which was designed and built with a 3D printer, fixed to the system with four vertical screws. There are two nuts on each screw that are used to set the height and the inclination of fibers relative to the polishing papers. This piece contains one hundred holes where the fibers will be placed.

Fibers use their own weight to press the polishing paper. Due to the reason that they are light (0.16 g), we use a plastic belt and a piece of metal with a weight of around 1.5 g, shown in Figure 4.5b, to increase their weight (in the same way as in thorlabs polishing process).

The mobile part consists of a flat PMMA plate, whose dimensions are $18 \cdot 18 \text{ cm}^2$, on which the polishing paper is fixed. This part is fixed to two horizontal screws, perpendicular to each other, shown in Figure 4.5c, that are used to move it around the XY plane (horizontal plane).

This system contains multiple switches, each mounted on a piece designed with a 3D printer, shown in Figures 4.5a, 4.5c and 4.5d, which are used to find the coordinates origin when the system is reinitiated and to stop it if they reach the end of the path.

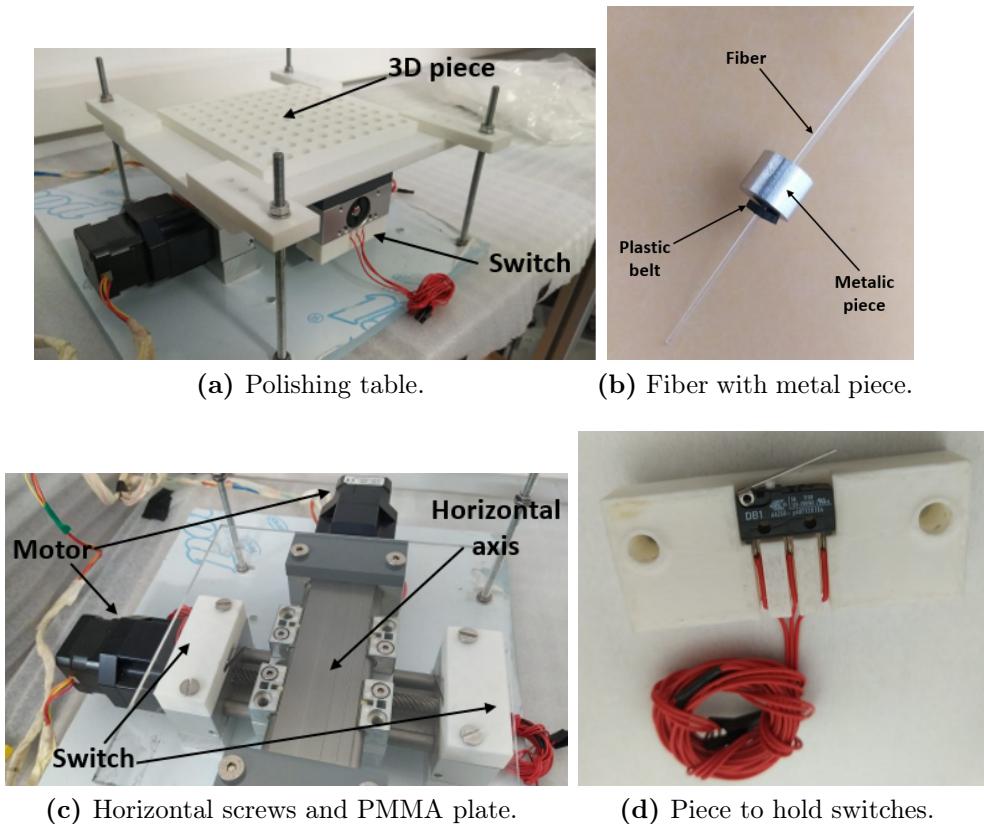


Figura 4.5 – Polishing table of the polishing machine

2. The electronic part, shown in Figure 4.6, is based on arduino technology and it is used to achieve automatic movement of the polishing paper.

It consists of two stepper motors, model NEMA ST4209S1404-A [116], which are used to control the horizontal screws on which the polishing paper is fixed. These motors are controlled by an arduino UNO

[117] that uses a CNC shield [118] 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 it can limit the supply power to the motors, burning the motors in the worst case. Instead of using the Pololu A4988 drivers [119], which is one of the most widely used drivers, the first choice was the DRV8825 driver [120]. 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 was replaced by the TMC2208 [121], much less noisy since it includes the *StealthChop* function with which the noise is practically eliminated. Furthermore, this controller is much more accurate as it has a microstepping mode of 1/256.

The voltage and current used to power the motors are similar to the A4988 (35 V and 2 A) which is sufficient for the system since the used motors are 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 can cause loss of steps, producing movements different from those programmed or even destroying the driver. Therefore, a cooling system is needed to ensure the correct operation of the polishing system.

The cooling system, shown in Figure 4.6, a copper piece³ in contact with both controllers. It has the possibility to use a PELTIER cell to increase the cooling power of this system. A fan is also included to prevent heat accumulation inside the electronics box.

³The copper is one of the best thermal conductor at STP

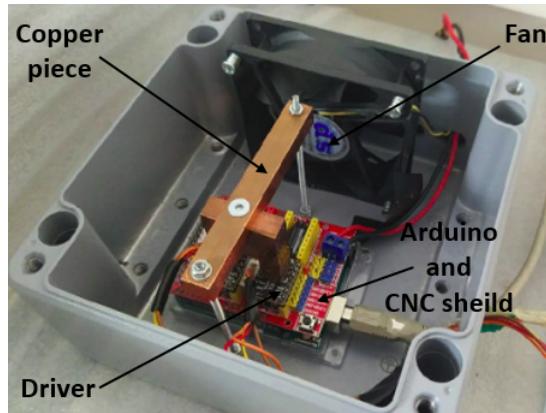


Figura 4.6 – Electronic system of Polishing machine.

This polishing machine is controlled by a computer using the Universal G-code Sender software (a graphical interface based on the GRBL package). It has several useful pre-programmed functions such as "HOME" with which the system, using the previously installed switches, finds its origin coordinate every time the system is turned on.

It also has the ability to load a file containing the g-code to be executed, which is used in TRITIUM experiment to program the 120 movements required for each polishing paper.

To test this machine, twenty fibers with a length of 15 cm were cut and arranged in a bundle. They were fixed in a structure shown in Figure 4.7 with two PMTs located at their ends, which were read in time coincidence using the electronic system described in section 3.2.4, Figure 3.14b.

Two different measurements were taken using two different radioactive sources, a gamma source, ^{60}Co , whose activity was approximately 715 Bq, and a beta source, ^{90}Sr , whose activity was approximately 17.8 kBq. After that, the fiber bundle was polished with the polishing machine developed and the test was repeated.

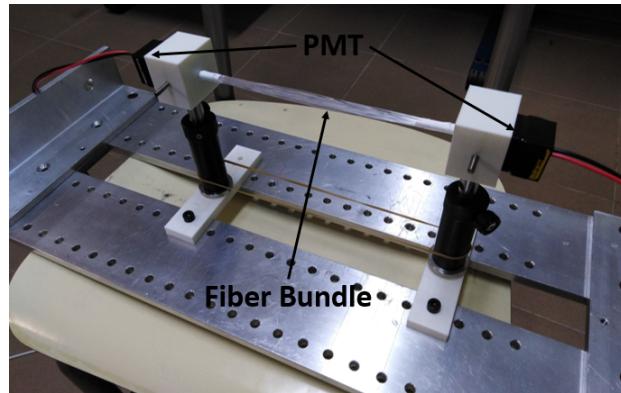


Figura 4.7 – Set up used to test the effect of the polishing machine.

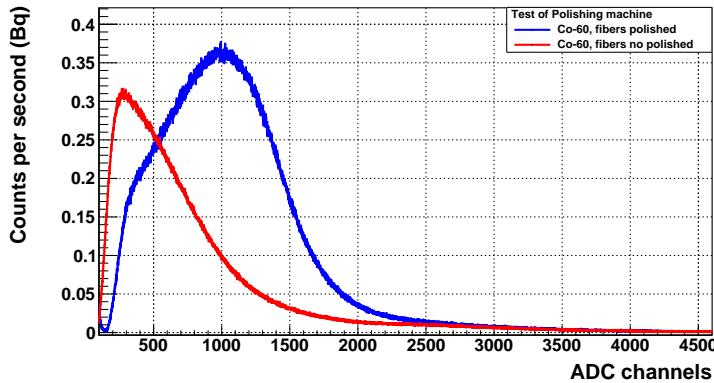
The energy spectrums recorded are shown in Figure 4.8 for each radioactive source, which were placed in the middle of the fiber bunch, that's, 7.5 cm from each PMT.

As can be seen in these figures, both energy spectrums have shifted to the right of the spectrum, which means that the detected events have more energy (more photons per event has reached the PMTs). This increase was quantified in more than 40% (42% for gamma source and 49% for beta source).

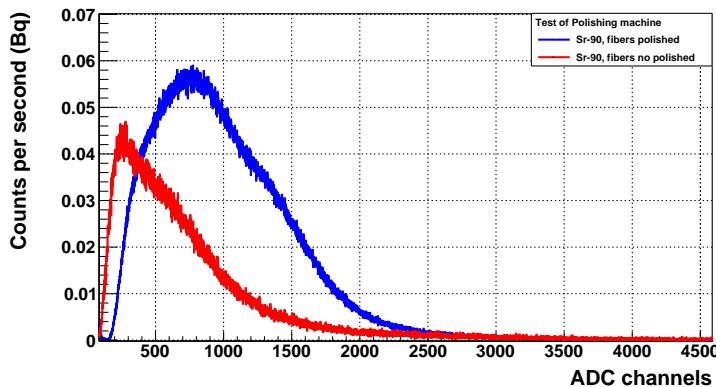
In summary, with the polishing machine, the photon collection efficiency of the fibers after their cut has been improved (mainly for the improvement of the interface between fibers and PMTs). It is very important to achieve a high detector efficiency as the expected number of photons in a tritium event is very low.

4.1.3 Characterization of Scintillating Fibers

This section shows the characterization of no-clad BCF-12 fibers from Saint-Gobain, which are the fibers used in the TRITIUM experiment. These



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



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

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

measurements are compared to other measurements that was made using single clad and multiclad BCF-12 fibers to quantify how the clad affect to several parameters of scintillating fibers.

It is an interesting comparison because, although commercial clads cannot be used for the TRITIUM experiment, they can be developed with a low enough thickness. For example, clads with a thickness of the order of tens of nanometers can be achieved by electrodeposition techniques.

As it was shown in section 3.2.2, the difference between these three types of fibers is that no clad fibers only consist of a polystyrene core with a refractive index of 1.60 whereas, in single clad fibers, the polystyrene core has an acrylic cover (PMMA) with a thickness of 30 μm and a refractive index of 1.49 and, in multiclad fibers, this acrylic cover has another fluor-acrylic cover with a thickness of 10 μm and a refractive index of 1.42.

This characterization was performed at the level of a single scintillating fiber and the parameters measured for every fiber type are the fiber collection efficiency and the uncertainty in the fiber response due to the conditioning process developed in TRITIUM. The reference measure that will be used for that is the number of photons per second that reach the active area of the photosensor.

To measure this parameter, a calibrated PMT, model R8520-06SEL, was used, whose quantum efficiency at the working wavelength, 29.76%, measured by the provider, Hamamatsu. To measure this parameter, the PCB described in section 3.2.4 is needed to work without the internal gain of the PMT and the Keithley 6487 Picoammeter/Voltage Source is used to measure the output current of the PMTs. The number of photons per second can be known from the Keithley measurement using the equation 3.10 with $QE = 0.2976$ and $CE = 1$. A simplified scheme of the set up used for this characterization is shown in Figure 4.9.

It consists of an optical structure in which a LED and a PMT are fixed to the specific distance between them, established by the user. The LED, model LED435-03 from the Roithner LaserTechnik GmbH company [122], is used to reproduce the light emission by the fibers used. The emission spectrum of the LED is shown in Figure 4.10, which was experimentaly measured using a spectrometer and fitted to a Gaussian function. The emission peak of this LED is produced at 433.9 nm with a FWHM of 18.4 nm.

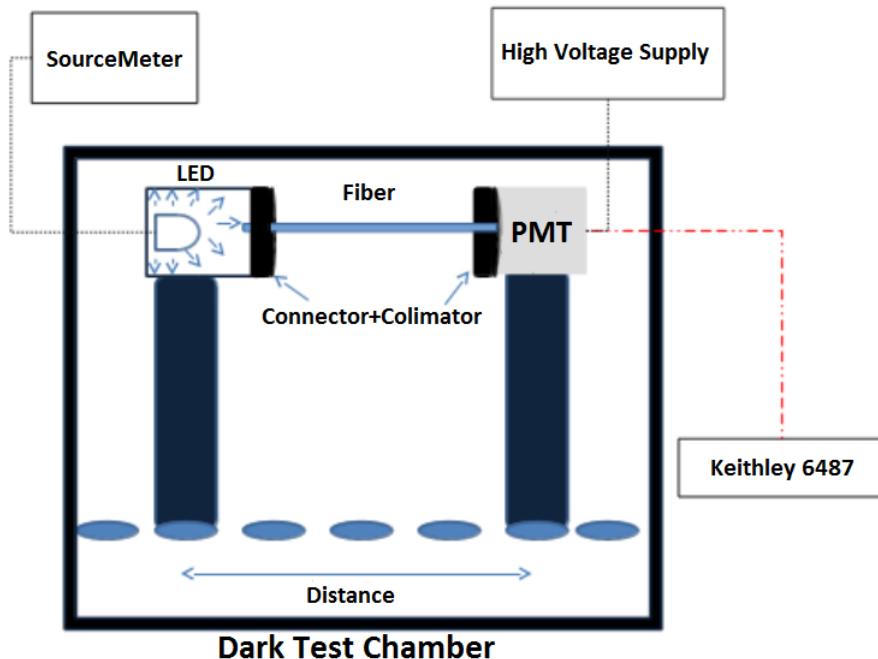


Figura 4.9 – Set up used for fiber characterization.

The fiber will be fixed between the LED and the PMT, whose distance is configured based on the length of the fiber, 20 cm for this study, and optic grease [124] was used for optimal optical coupling between the fiber and the PMT. Two collimators are used to ensure that the photons detected in the PMT are only those that come from the LED and travel through the fiber and two connectors, type FH-ST⁴ from RoHS company [], were placed to the ends of the fiber and used to fix it to the system.

Preparation of the System Used for the Characterization Study.

Before we start with the characterization of the fiber several verification tasks must be performed to verify that the system is working properly. The

⁴FH-ST is a quick assembly connector for 1 mm POF

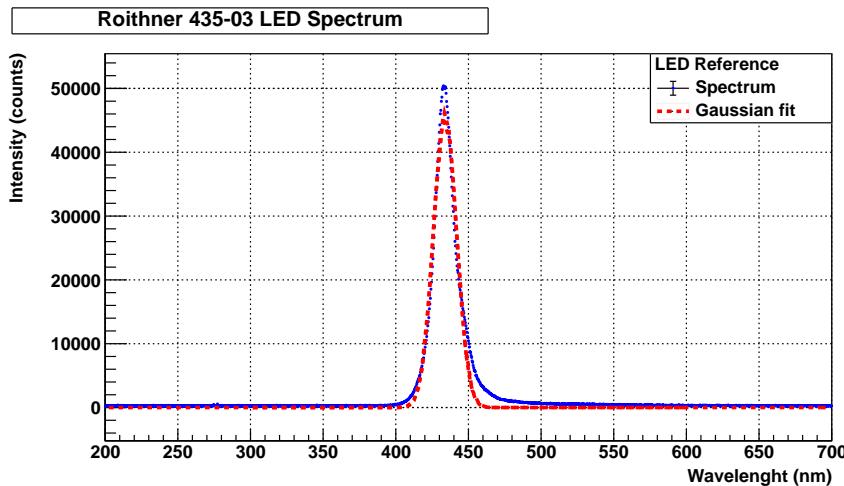


Figura 4.10 – Emission spectrum measured for the LED model 435-03 from Roithner LaserTechnik GmbH Company.

quality of the tightness to the light of the black box used and the correct operation of the PMT for this study, which involves checking the correct operation of the PCB designed and checking the linearity of the PMT output signal in the study range, must be verified.

First, the quality of the light tightness of the black box used was verified. It is important because we are detecting small signals, a few hundred photons per nanosecond, so it must be verified that the background of the system are below that.

This test is performed by measuring a no-clad fiber, whose length is 20 cm, in the previous assembly. This measurement was carried out by feeding the LED with four different intensities (0.05 mA, 0.1 mA, 0.15 mA and 0.2 mA) and this measurement will be repeated covering the set up with a special black blanket from Thorlabs [125], with which it can be ensured that the amount of external photons that reach that system is negligible.

This test was repeated for three different no clad fiber samples and the mean and standard deviation were calculated using the equations 4.1

for each light intensity.

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

The difference between the results obtained in both tests is presented in Figure 4.11.

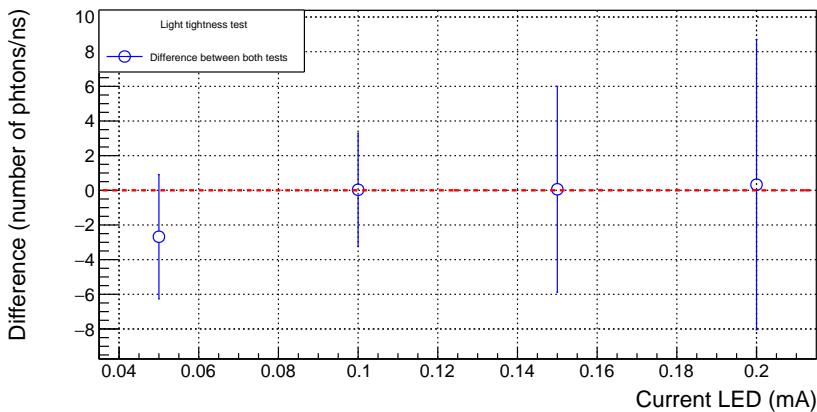


Figura 4.11 – Difference between the results obtained in both tests carried out to check the light-tight quality of the system.

As can be seen in this figure, there are no statistically relevant differences between both situations, which can be verified with the help of the red auxiliary line that marks 0 (no difference). Therefore, it can be ensured that the quality of the light tightness of the black box used is high enough for this study.

Then the operation of the PCB without the internal gain will be verified. It consists of finding the plateau in which the electron collection efficiency in the first dynode is practically 100%.

This test consists of, using the setup previously explained without any fiber, feeding the LED at 1 mA intensity. There, the PMT output current was measured for different supply voltages of the PMT, between 0 and 500 V. The Figure 4.12 show the number of photons detected by the PMT (using a semi logarithmic scale).

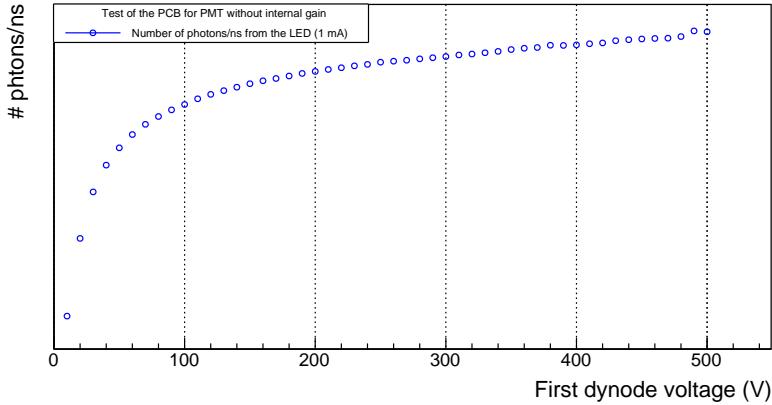


Figura 4.12 – Response of the PMT based on its high voltage using the PCB with which we get no internal gain from the PMT.

As can be seen, this plateau is found at voltages higher than 150 V, where the PMT output response are stable. This is the interesting range for this study so, the chosen voltage at which this study was developed is 250 V.

Finally, the linearity of the PMT is verified. In this study the LED will be powered with several intensities of up to 10 mA (LED linearity range) to ensure that its emission is not saturated.

This linearity is tested in two different ranges, one in the range of the number of photons expected in a tritium event that is only a few tens of photons per tritium event (tens of photons per nanosecond), and second, in the range of this study, whose events will have up to two thousand five hundred photons per nanosecond.

To test the linearity of the PMT at the level of tritium events, the set up explained above is used without any fibers and without the connector that there is in the part of the PMT but keeping the collimators to ensure that the active area of the PMT is the same as the one we use in the study.

To test the linearity of the PMT at the level of more than a thousand photons per nanosecond, we remove the other connector (the one in the LED part) in order to increase the photons emitted by the LED that reach the photosensor and also keeping the collimators.

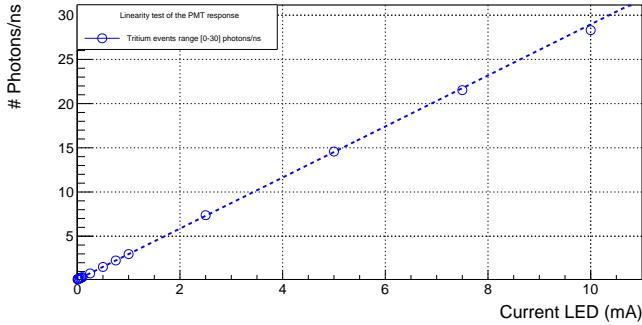
Both results is shown in Figures 4.13, where the uncertainties are included but they are too small to be visible.

As can be seen, the PMT output current is linear in both cases, so the system is ready to start with the characterization of the fibers.

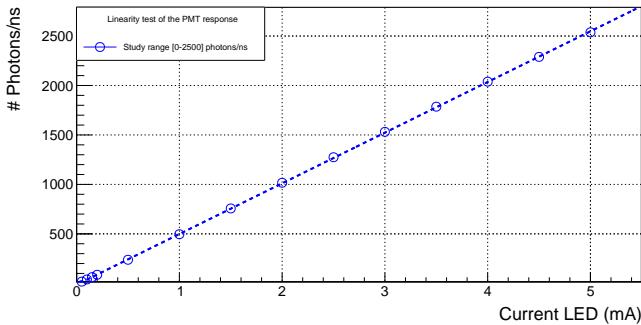
Results of the Characterization of Scintillating Fibers

First, the uncertainty of the conditioning process, σ_{con} , was experimentally measured. This uncertanty appears because, as we saw before, each fiber have to be conditioned, consisting in cutting and polishing it, before using. This is an individual task that can present a small dispersion, affecting the response of each individual fiber. It is an important measurement because this uncertainty will be present in the TRITIUM detector.

To measure it, it has to be taken into account that there is a bit of freedom in this system due to the position of the connectors which are fixed to the fiber (1 mm or less) which means that there is an additional uncertainty, σ_{pos} , in the measurement. Since both uncertainties are not related, the total measurement uncertainty can be calculated as a square sum, equation 4.2.



(a) Verification of linearity in the response of the PMT in the range of tritium events.



(b) Verification of linearity in the response of the PMT in the range of this study ($0 - 2500 \gamma/\text{ns}$).

Figura 4.13 – Linearity tests of the PMT response

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

The uncertainty due to the fiber position are always presented in the measurements of this set up so, the only way to measure the uncertainty due to the conditioning process is to quantify the uncertainty in the fiber position and extract it to the total uncertainty, sum of both, using the equation 4.3. To do so, two different experiments was designed, one where

only the uncertainty in the fiber position are presented ($\sigma_t = \sigma_{pos}$), and other where both uncertainties are involved.

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

The test designed to measure σ_{pos} consisted of prepare one fiber of each type (no clad, single clad and multiclaid) using the conditioning process explained before. Then, fix each fiber in the set up, take a measurement by feeding the LED at an intensity of 0.1 mA and remove this fiber to the set up (and also the connectors). These measurements is repeated ten times with the same fiber, fixing and removing it every time.

Ten different measurements for each fiber type is obtained with this test, the standard deviation of which is only due to the uncertainty in the position. The results is shown in Table 4.1, which was calculated using the equations 4.1 and 4.4.

$$Rel. Std. Des. = \frac{Std. Des.}{\bar{x}} \quad (4.4)$$

Fiber type	Average (γ/ns)	Std. Des. (γ/ns)	Rel. Std. Des. (%)
No Clad	524.088 ± 0.010	17.65	3.37
Single Clad	1071.696 ± 0.01	9.07	0.85
Multiclad	949.930 ± 0.026	9.91	1.04

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

As can be seen, the clad of the fiber reduces the uncertainty due to the position of the fiber, which means that it improves the uniformity of the fiber response. It can be also seen that the use of the clad greatly improves the collection efficiency of the fibers since both types of fibers

with clad have collected more photons than the fiber without clad. It could be because photons are mainly collected in the core of the fiber and the interface created by this core and the surrounding greatly affects the amount of photons collected. This interface can be much more controlled in the case of a single clad or multiclad fibers than in no clad fibers, where it is created between the core and the environment (air or water in the case of TRITIUM), where external conditions, such as the dirt in the room, can affect a lot.

We can also see that the use of a second clad slightly reduce the collection efficiency. One possible reason for this is that, to add a second liner, the polystyrene core must be reduced proportionally.

It is also appreciate in the table that the error of the measurement, provided by the keithley and propagated to the average, is three times smaller than the standard deviation, so it was not taken into account any more.

The second experiment, in which both uncertainties are involved, consists of preparing ten different samples of each fiber type (using the conditioning process) and measuring each fiber under the same conditions as the previous test. This measurement was done for four different LED emission intensities (0.05, 0.1, 0.15 and 0.2 mA) to reduce possibles mistakes.

The case of no clad fibers is shown in Figure 4.14, where it can be seen that, indeed, although each fiber shows a very linear trend with the amount of photons that it collects, a dispersion in the fiber response is clearly seen in each figure. Similar results were obtained for single clad and multiclad fibers, shown in figures 4.15a and 4.15b respectively.

The average of these 10 samples for each type of fibers and its standard deviation are summarized in Tables 4.2, 4.3 and 4.4 and represented in Figure 4.16, where they can be compared.

Led Int. (mA)	Average (γ/ns)	Std. Des. (γ/ns)	Rel. Std. Des. (%)
0.05	243.46	9.82	4.03
0.1	540.62	33.51	6.20
0.15	902.74	36.83	4.08
0.2	1252.62	50.48	4.03

Table 4.2: Average, standard deviation and relative standard deviation of 10 different samples of no clad fibers.

Led Int. (mA)	Average (γ/ns)	Std. Des. (γ/ns)	Rel. Std. Des. (%)
0.05	383.81	33.23	8.66
0.1	922.68	73.97	8.02
0.15	1485.10	119.90	8.07
0.2	2053.78	166.39	8.10

Table 4.3: Average, standard deviation and relative standard deviation of 10 different samples of single clad fibers.

Led Int. (mA)	Average (γ/ns)	Std. Des. (γ/ns)	Rel. Std. Des. (%)
0.05	376.68	14.96	3.97
0.1	870.87	34.58	3.97
0.15	1396.60	55.24	3.95
0.2	1932.57	76.02	3.93

Table 4.4: Average, standard deviation and relative standard deviation of 10 different samples of multi clad fibers.

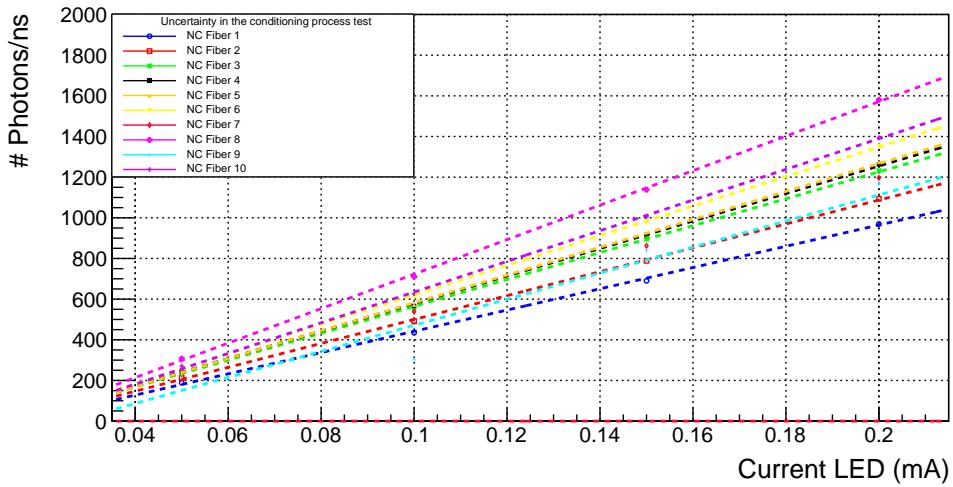


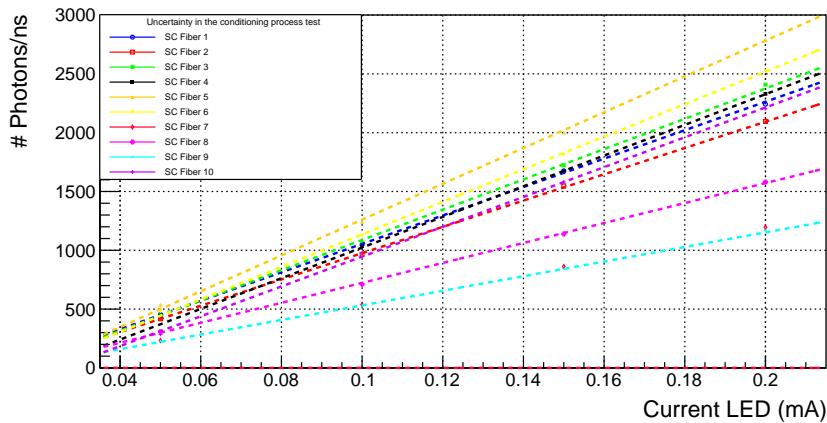
Figura 4.14 – Number of photons/ns reaching the PMT for No Clad fibers.

As it is shown these figures, they have a very linear trend which confirms the correct behavior of the fibers. It can be appreciate that, similar to what happened with the previous test, single clad and multiclad fibers, both, have higher signals than no clad fibers, which means that the clad has an appreciable effect on the fiber collection efficiency and it could be a possible point for futur studies.

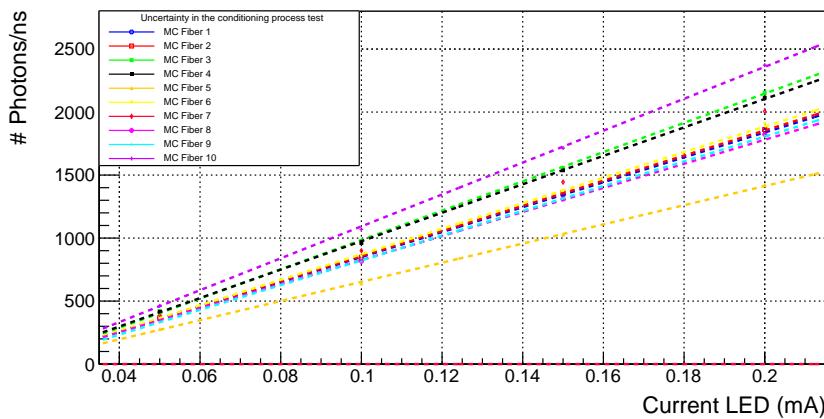
Again, similar to what happened in the previous study, single-clad fibers have higher collection efficiency than multiclad fibers, something that was verified in all the tests performed.

The relative standard deviation are also presented in these tables, where we it can be seen that the dispersion of each fiber type for different LED intensities is practically negligible, which again verifies the correct behavior of the system.

There is only one point (no clad fiber with 0.1 mA) that is higher than we expect. We can see in Table 4.2 that the reason for this is that its



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



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

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

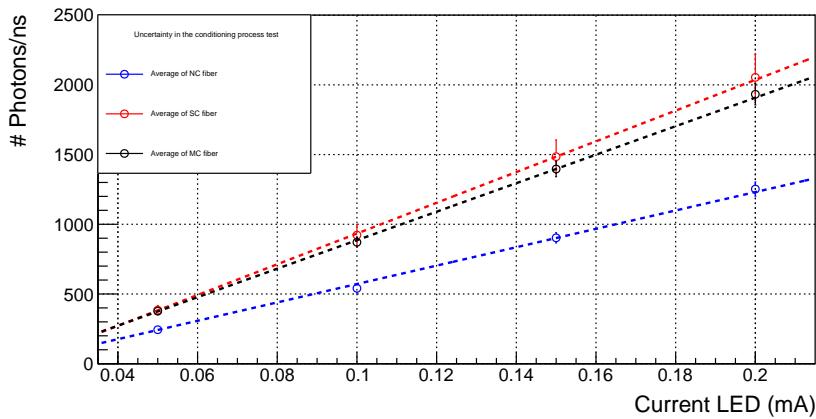


Figura 4.16 – Average of 10 samples for each fiber type (no clad, single clad and multiclads fibers).

standard deviation is too high (as high as the measurement for no clad fibers with 0.15 mA). The reason was found in the sample 9, whose measurement was very different from the average, increasing the standard deviation, probably due to a problem in the measurement process. We discard this sample because this result is not representative.

To uniform the results, an average of these four values is calculated for each fiber type and shown in Table 4.5, where the uncertainty in the fiber position and the uncertainty due to the conditioning process, previously calculated, are also shown.

Fiber type	σ_t (%)	σ_{pos} (%)	σ_{con} (%)
No Clad	4.01	3.37	2.17
Single Clad	8.21	2.17	7.92
Multiclad	3.96	1.04	3.82

Table 4.5: Relative standard deviations (σ_t , σ_{pos} and σ_{con}) measured in this test.

As it can be seen, the least uncertainty in the conditioning process

is found in the no clad fibers, which means that the damage from this process occurs mainly in the fiber clad, which can be checked in Figure 4.3. It was checked under the microscope that this damage only occurs at the end of the fiber.

Also, the largest relative standard deviation in this process is measured for single clad fibers, which means that the second clad increases the resistance of the fiber to this process.

In summary, this study has shown, on the one hand, the use of fiber clad improves the photon collection efficiency, which could be an interesting point for future studies, and, on the other hand, the relative statistical deviation due to the fiber conditioning process developed in the TRITIUM experiment was quantified for each fiber type, where it was checked that the main damage of the conditioning process is produced in the fiber clad so, if a method to create a clad for the fibers is developed, it should be applied after the fiber conditioning process.

Finally, the measurement of the photon collection efficiency of each type of fiber is shown. The collection efficiency is the percentage of photons collected along the fibers. It is usually given by the manufacturer per meter of fiber, EC_{100} .

To measure it, we prepare ten different samples with a length of 10 cm for each fiber type and measure each one using the set up previously explained. Then, the average and standard deviation was calculated using the equations 4.1, whose results are shown in Table 4.6.

The collection efficiency can be calculated by comparing these tests with those performed for a fiber length of 20 cm, whose values has been previously shown in Tables 4.2, 4.3 and 4.4 since both was made under the same conditions. The results is shown in Table 4.7:

As the difference between the fiber length in both studies is only

Led Int. (mA)	No clad (γ/ns)	Single clad (γ/ns)	MultiClad (γ/ns)
0.05	318.35 ± 61.34	549.62 ± 70.79	480.35 ± 83.72
0.1	735.65 ± 143.02	1269.91 ± 164.32	1110.66 ± 193.44
0.15	1183.91 ± 232.07	1983.93 ± 230.97	1777.40 ± 307.19
0.2	1645.18 ± 323.76	2506.97 ± 208.01	2338.43 ± 350.24

Table 4.6: Average and standard deviation of 10 different fibers of 10 cm.

Fiber type	CE_{10} (%)	CE_{100} (%)
No Clad	75.97 ± 7.61	7.597 ± 0.761
Single Clad	77.96 ± 5.66	7.796 ± 0.566
Multiclad	82.60 ± 7.24	8.260 ± 0.724

Table 4.7: Collection efficiency of each fiber type for 10 centimeters, CE_{10} , and 1 meter, CE_{100} .

10 cm, the collection efficiency calculated from these measurements, CE_{10} , is only at that distance. Assuming a linear dependence of this parameter with the distance, the value of CE_{100} can be extrapolated.

The collection efficiency per meter given by the manufacturer Saint-Gobain is between 7% and 3.44% [78]. As collimated photons are used in this study, it can be assumed that it is the best case, 7%.

As can be seen in Table 4.7, our measured values are very close to the one provided by the manufacturer. The difference between this value for the three types of fiber studied is not as large as it was expected. A possible reason is that the difference in fiber length is only 10 cm and it may not be enough to see this effect. It could be interesting to repeat these tests with a larger difference in fiber length.

4.1.4 Cleaning Process for Scintillating Fibers

Finally, an addition step was included to the fiber conditioning process, with the objective of improving the photon collection efficiency of the fibers.

The tritium events detected in the fibers produce few photons, so it is very important to conserve as many photons as possible. As it was shown in the fiber characterization study, the quality of the interface that is created between the core of no clad fibers and the environment (tritiated water in the case of TRITIUM detector) greatly affects the photon collection efficiency.

Therefore, a mechanism, called the fiber cleaning process, was applied to improve the state of this interface. This mechanism aims to remove all external particles deposited on the fibers, such as fat molecules deposited when the fibers are touched, which can affect the quality of the interface, worsening their photon collection efficiency.

As a consequence of this cleaning process, the wetting property of the fibers, shown in Figure 4.17, is improved, preventing air molecules from attaching to the fiber and achieving a uniform water clad around each fiber, resulting in an improvement of their collection efficiency.

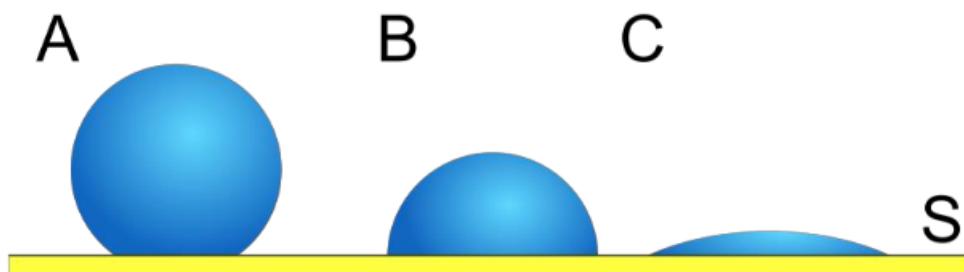


Figura 4.17 – Wetting property produced by the cleaning process. []

This cleaning process was carried out within a clean room at IC-

MOL laboratories⁵ and it was developed by their researchers.

It consists of filling three different beakers, one with alkaline soap, another with millipore water⁶ and the last one with isopropanol. First, the fibers are rubbed for 5 minutes with alkaline soap and then placed in the first beaker for sonication for 3 minutes. Then, the fibers are cleaned with a constant flow of water for 5 minutes. Second, the fibers are placed in the second beaker for sonication for another 3 minutes. Third, the fibers are placed in the third beaker for sonication for another 3 minutes. Finally the fibers are dried with an N_2 air gun and introduced inside of the prototype.

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

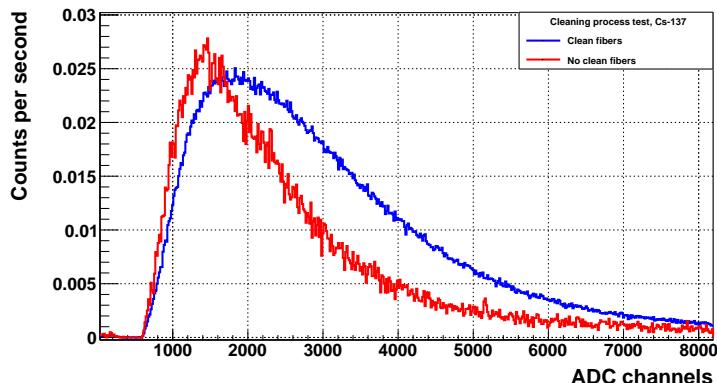
Two radioactive sources were used in this study, a beta source, ^{90}Sr , which is the same as that used in the polishing machine test, and a gamma source, ^{137}Cs , the activity of which is of the order of 500 Bq.

The results are shown in Figures 4.18, where a shift of the spectrum to the right can be seen.

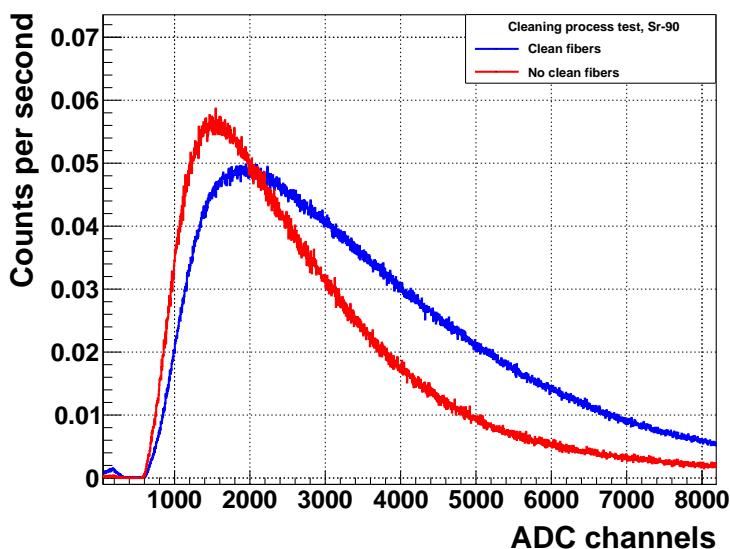
A rude approximation was done to quantify this improvement. It was calculated using the equation 4.5, where A_C is the counts per second measured after the cleaning process and A_{NC} is the counts per second measured before the cleaning process.

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

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



(a) Energy spectrum recorded for the Cs-137 source.



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

Figura 4.18 – Energy spectrums used to test the effect of the Cleaning process

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

The obtained improvement is 20.74% for the gamma source and 20.99% for the beta source so, the improvement of the photon collection efficiency of the fibers was verified using the cleaning process carried out in the clean room of ICMOL laboratories. Nevertheless, it should be taken into account that this test was carried out in air. It could be interesting to repeat it in water to obtain more realistic conclusions since the fibers of the TRITIUM detector will be immersed in water.

4.2 Characterization and R&D on SiPM

4.3 Characterization of the Ultrapure Water System

This section shows the characterization of the ultrapure water system, the objective of which is to ensure that the quality of the water sample used to be measured is good enough to overcome the requirements of the TRITIUM detector. There are three different requirements that this ultrapure water system must meet:

1. A low enough conductivity⁷ of the water needs to be achieved, around $10 \mu\text{Sv}/\text{cm}$, so that external particles dissolved in the water don't be deposited on the fibers, drastically reducing the detector efficiency due to such a low mean free path of the tritium electron.

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

2. It must be noted that this system does not have any spectral capabilities that can be used to distinguish between several radioactive isotopes, so to measure only tritium, this system must remove all external radioactive particles (other than tritium isotope) from the sample.
3. Lastly, the tritium activity should not be affected by this process.

To verify that these requirements have been exceeded, a characterization of the water sample was carried out before and after the ultrapure water system, called raw water and purified water respectively. This characterization consists of measuring the water sample conductivity and the activity of each radioactive element that are present in the sample. The turbidity and the chemical components of the water sample will also be measured.

It must be taken into account that, so that the sample was representative of the raw water sample, it was taken from a place located very close to the input of the ultrapure water system, which is located at 40 meters from the ultrapure water system and two meters deep in the river. It was seen that it is very important since variations of up to 25% in the tritium activity was measured between both points (due to the diffusion of tritium along the river).

First, the chemical composition of the water was measured before the ultra-purification process by a physico-chemical analysis, which was carried out a few years ago. It is shown in Table 4.8.

This table shows that the water sample contains a number of components that must be cleaned to prevent their deposition on the scintillating fibers of the detector, reducing their sensitivity, or affecting the tritium measurement.

Chemical components	Concentration (mg/L)
CO_3H^-	154
Mg	46
Ca	105
NO_3^-	16
Cl^-	196
NO_2^-	0.03
K	11
Na	173
SO_4^-	217
Dry Residue	1029

Table 4.8: Chemical components and turbidity measured in the raw water sample.

Its turbidity⁸ was also measured using the Hanna Hi 9829 portable multiparameter system from Hanna Instruments [126], obtaining a value of 29 NTU, much higher than the WHO recommended limit of 5 NTU.

Second, the conductivity was measured for both, raw and purified water. To do so, the same multiparameter system was used, the Hanna Hi 9829. These measurements, together with the measurement of the conductivity of reject water, explained in section 3.3.2, are presented in Table 4.9.

As can be seen in the first column, the raw water sample has high values of conductivity, caused because it contains many different ions, shown in Table 4.8. It can be appreciate that, in the second column, the conductivity values of pure water was reduced by almost two orders of magnitude, reaching values close to $10 \mu\text{Sv}/\text{cm}$, exceeding a requirement previously mentioned.

⁸The turbidity of water is the loss of its transparency due to dissolved particles, normally measured in NTU, Nephelometric Units of Turbidity, which measure the intensity of the scattered light at 90 degrees.

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 4.9: Measurements of the conductivity for several samples of each water type (raw water, pure water and reject water).

Finally, as can be checked in the third column, the reject water has high values of conductivity. The reason of that is because it contains the ions that was removed to the pure water.

Third, the gamma radioactive elements present in the water sample of both types, raw water and purified water, was identified and their activities was measured using a high purity germanium detector, HPGe. Then, a gamma analysis was carried out to find the natural gamma emitters (those that come from the natural radioactive series, Table 3.4) and the artificial gamma emitters with long enough lifetime to be measured (those that come from the activation of nuclear fission of neutrons).

The radioactive elements found in the raw water sample with activities high enough to be measured by the HPGe detector was 40^{k} and small quantities of 226^{Ra} which were completely disappeared in the purified water sample.

Lastly, the tritium activity was measured to see how it is affected by the ultra-purification process. This measurement was carried out using the Quantulus system, which consists of a liquid scintillator mixed with tritiated water, readout by PMTs.

Before this measurement, each water sample was filtered at 0.45

microns to remove any particles that could cause the extinction of the scintillation signal.

The Table 4.10 show several measurements of the activity for different tritium samples of each water types (raw water, reject water and purified water).

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

Table 4.10: Measurements of the activity for several samples of both water types (raw water and pure water).

As can be see, tritium activity is not affected by this system, exceeding the last requirement of the ultrapure system.

4.4 Characterization of the TRITIUM Cosmic Veto

This last section shows 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.

First, the quality of the veto coverage, shown in Figure 3.25, is verified. This study is done at the level of one detector so the configuration of the electronic chain used is the one shown in Figure 3.14c. To do so, the surface of the veto is divided in 9 points, shown in Figure ??, which was used as a reference to place a gamma source.

FIGURAAAAA

Two different tests were made for this task:

1. The first test was used to quantify the improvement of the veto signal due to its coverage. It consists of, placing a ^{137}Cs source at point 2, measuring with the veto uncovered. Then cover the veto and repeat this measure. The result is shown in Figure 4.19.

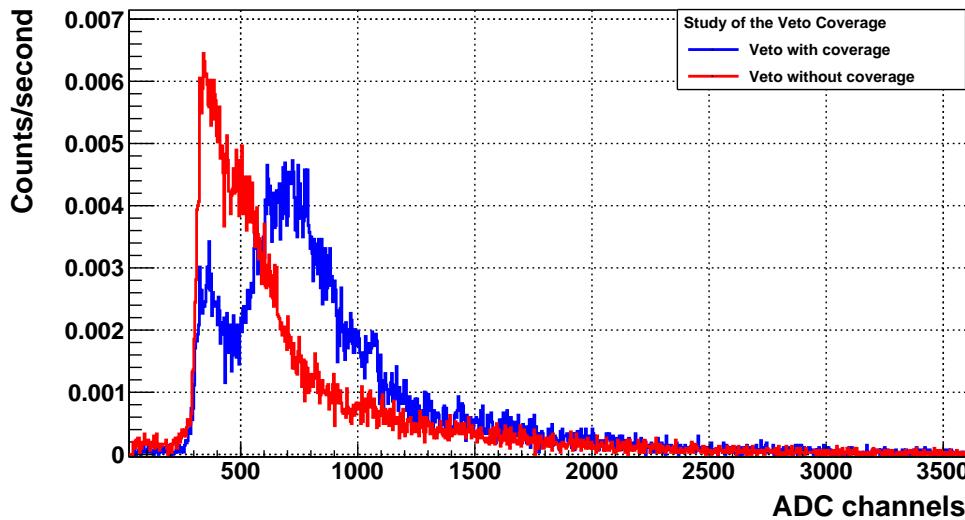


Figura 4.19 – Measurement of a radioactive source ^{137}Cs with the TRITIUM cosmic detector with and without its coverage.

It can be seen that the spectrum has shifted to the right, which means that more photons have been collected per event. No improvement was measured in the number of events detected, only an improvement in their collection efficiency.

2. The second test was used to verify the spatial uniformity of the signal in the covered veto. For this task, a mapping was carried out, which consists of placing a ^{60}Co source at each point and measure the number of events detected in the same time windows (). It was done for two

different veto and the energy spectrum obtained was integrated. The number of count rate obtained in each point is displayed in Table 4.11 for both vetos, the values of which is represented in a bidimensional plot in Figures ?? and ??, respectively.

Point	Veto 1 (counts/s)	Veto 2 (counts/s)
1	18028 ± 3	18293 ± 1.5
2	19133 ± 5	20014 ± 4
3	17858 ± 4	18843 ± 4
4	18969 ± 5	18761 ± 5
5	19893 ± 4	19841 ± 3
6	18573 ± 4	18850 ± 5
7	18200 ± 4	17790 ± 4
8	19725 ± 4	19312 ± 4
9	18030 ± 5	17804 ± 5

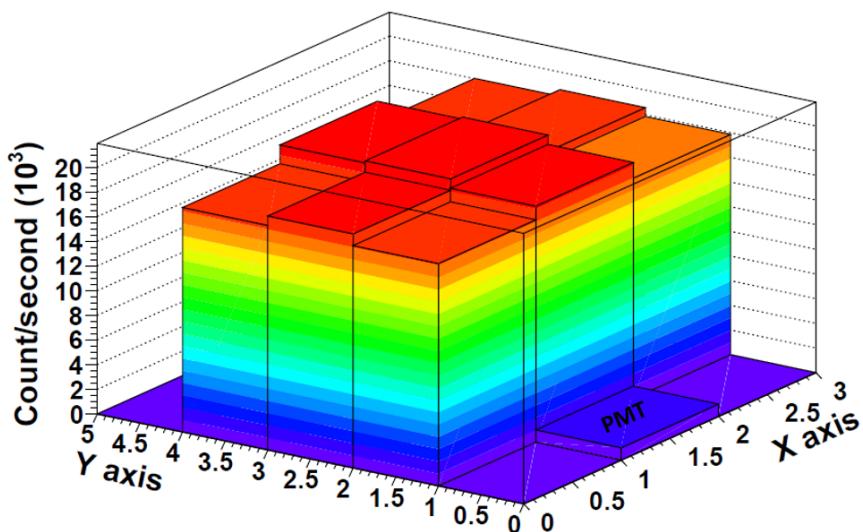
Table 4.11: Count rate measured with two different cosmic detectors using a radioactive source ^{60}Co .

It can be observed that the veto signal has a uniform behavior on its surface, obtaining a fairly similar counting rate in all the points considered in this study.

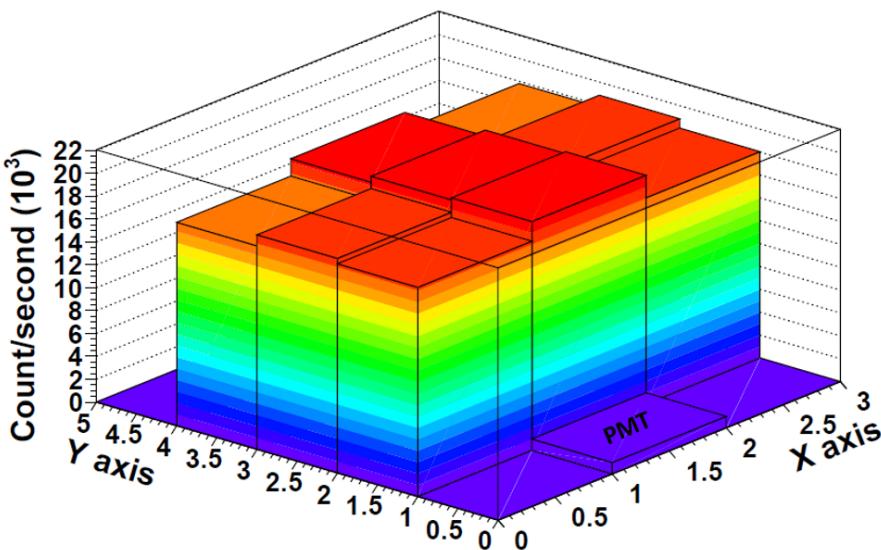
The following studies of this section is done at the level of a cosmic veto (both detectors in coincidence), so the configuration of the electronic chain used is the one shown in figure 3.14c.

With the coverage of the veto correctly tested, the following step is to find the conditions in which the detection of cosmic events is optimized. This optimization consists of, on the one hand, finding the minimum high voltage of PMTs in which their efficiency is stable, and, on the other hand, finding the maximum threshold of the discriminator⁹ at which we start to

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



(a) Mapping of the first TRITIUM cosmic detector.



(b) Mapping of the second TRITIUM cosmic detector.

Figura 4.20 – Bidimensional graph of the count rate (Mapping) measured with two different TRITIUM cosmic detectors using a radioactive source of ^{60}Co .

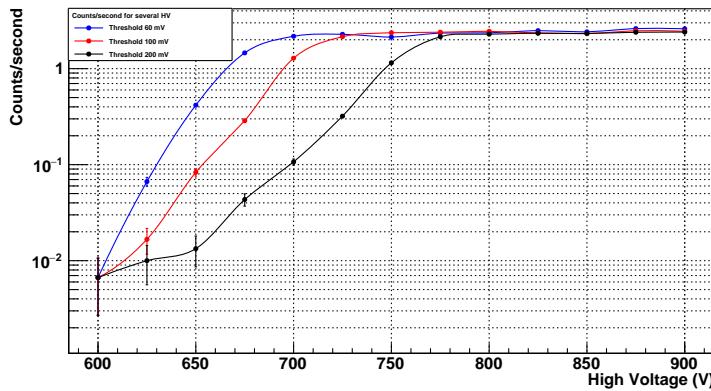
loss cosmic events in their detection. For higher high voltage and smaller thresholds of the found values, a plateau should be found.

To find both parameters, two different studies were carried out, in which the number of coincident events (cosmic events) were measured. On the one hand, it was measured at several high voltages and fixed thresholds and, on the other hand, it was measured at several thresholds and fixed high voltages. Both measurements are shown in Figure 4.21 in which a semi-logarithmic scale is used.

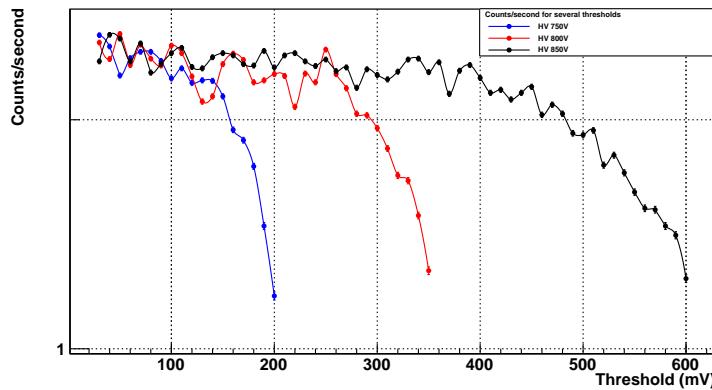
To find the optimized conditions the amplification line of the configuration of the electronic chain 3.14c was eliminated and the output signal of the coincidence module, second stage, was connected to a CAEN Quad Scaler And Preset Counter-Timer module, N. 1145, [127], used to count the number of events in a time window of 300 s.

In Figure 4.21a, the measurements at several high voltage and a fixed thresholds is shown, which was done for three different thresholds, 60 mV, 100 mV and 200 mV. As can be seen, there is a minimum high voltage for each threshold used, 700 V, 730 V and 780 V respectively, at which the plateau start. This minimum voltage is higher when the value of the threshold is increased, as it should happen. The voltage chosen to work is 800 V since it can be assured that it is on the plateau for the three thresholds.

In the same way, in Figure 4.21b, the measurements at several thresholds and a fixed high voltage is shown, which was done for three different high voltages, 750 V, 800 V and 850 V. As can be seen, there is a maximum threshold for each high voltage used, 140 mV, 270 mV and 450 mV respectively, at which the plateau ends. This maximum threshold is increased for higher voltage, as it should happened. The threshold choosen to work is 200 mV since, for the previous election, 800 V, it can be sure of being on the plateau.



(a) Counts per second for several high voltage at three different thresholds.



(b) Counts per second for several thresholds at three different high voltage.

Figura 4.21 – Counts per second at several high voltage and fixed thresholds and several thresholds and fixed high voltage.

Next, the energy spectrum of cosmic events was measured, which is shown in Figure 4.22. For this task the configuration of the electronic chain shown in Figure 3.14c was used with the values previously mentioned, 800 V and 200 mV.

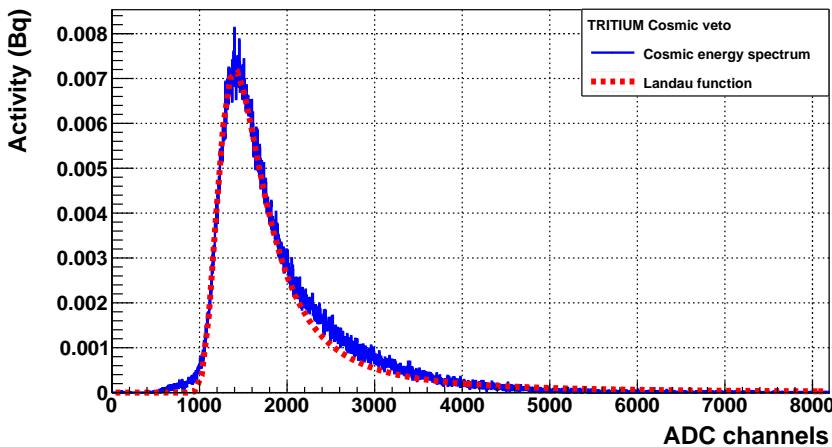


Figura 4.22 – Energy spectrum measured with the cosmic veto.

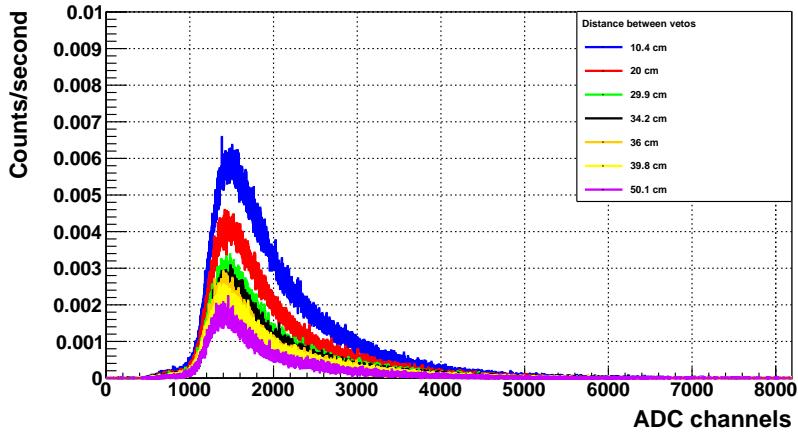
As can be seen, this energy spectrum fits well with a landau function as expected. The number of detected cosmic events can be known by calculating the area integral of this spectrum, whose result is 2,5 event/s. The theoretically expected cosmic rate, calculated in section 3.4.2, is 2,909 event/s, so the efficiency of the active veto developed in TRITIUM experiment for cosmic events detection is 85%, which is a common value of the efficiency of cosmic detectors.

Finally the relationship between the detected cosmic events and the distance between both detectors that form the cosmic veto was obtained. It is interesting because this distance can be changed if other tritium prototypes are used and we need to know the expected cosmic rate for each different situation. To do so, an energy spectrum was measured for five different distances, which are approximately 10 cm, 20 cm, 36 cm, 40 cm and 50 cm, which is shown in Figure 4.23a. The energy spectrum previously

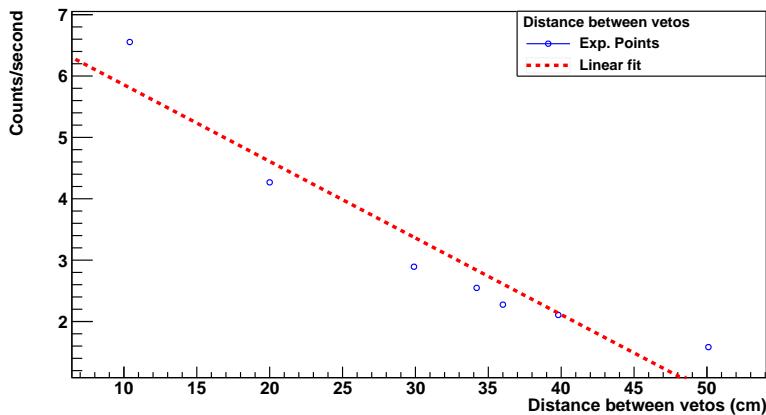
shown in Figure 4.22 was also included.

As we can see, the shape of the spectrum is the same because the energy of the detected events is the same (cosmic events) but the quantity of their events is less for greater distance. The reason for that is that when we increase the distance, the solid angle formed by the active veto is smaller.

The detected cosmic events was calculated by the area integral and they are represented in Figure 4.23b as a function of the distance between both detectors, where a linear fit has been added. With this linear fit, the detected cosmic rate can be easily known if the working distance is changed.



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



(b) Linear fit of counts per second measured with the cosmic veto for several distance.

Figura 4.23 – Measurement of the cosmic veto for several distances between its cosmic detectors.

Chapter 5

TRITIUM Monitor Prototypes

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

On the one hand, the first two prototypes built were preliminary prototypes used to learn about tritium detection and to detect and solve problems in their designs.

On the other hand, the other two prototypes built were prototypes with a well-defined design in which no problems were found. They were built to check more subtle effects.

Each prototype was designed and built in our own workshops (at IFIC, Valencia or Aveiro, Portugal) and it was filled with tritiated water following a protocol specially developed for this task.

In each prototype used, several tightness and filling tests were carried out to guarantee their radiosecurity.

Finally, in the last section, the final monitor of TRITIUM detector will be explained. It is based on modular detection units for easy scalability, where each module will be the chosen prototype (the one with the best results) previously shown throughout this chapter.

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

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

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

5.1.1 TRITIUM-IFIC 0

The Tritium-IFIC 0 prototype was the first prototype developed in TRITIUM experiment and it was used to check the feasibility of the technology proposed by TRITIUM, that's, to verify that we are capable of detecting tritium in water with scintillating fibers.

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

The Tritium-IFIC 0 consists of bundle of 35 fibers, shown in Figure 5.1, whose length is 20 cm, which were cut and polished with the techniques

explained in section ???. This bundle has a metallic piece located in both ends, Figure 5.1a, which are used to fix it to the prototype.

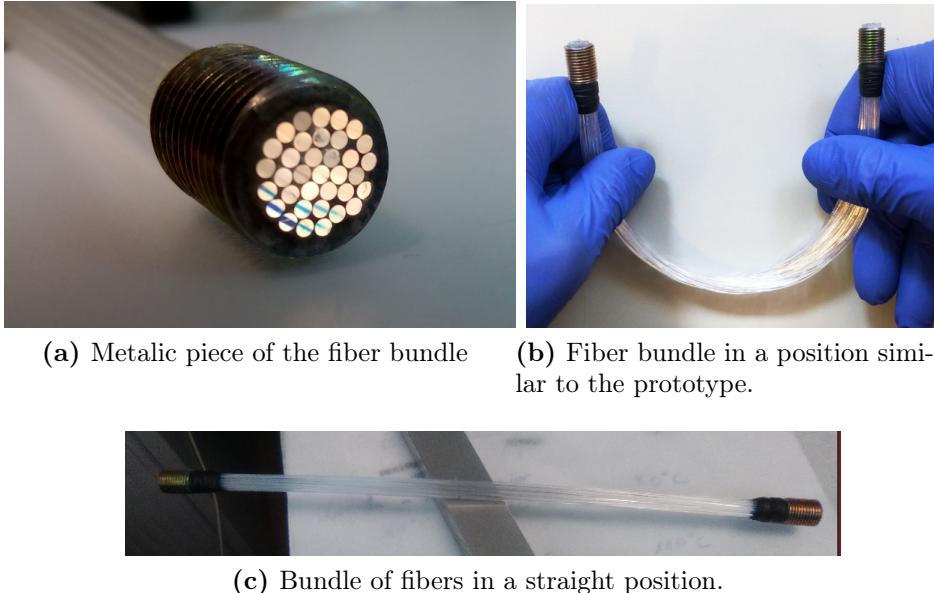


Figura 5.1 – Bundle of 35 fibers, the length of which is 20 cm, used in Tritium-IFIC 0 prototype

This bundle is placed inside of a vessel, whose material is PVC¹ since it is a safe material widely used. This vessel, shown in Figure 5.2, was designed in a U-shape because, in this way, radiological safety was improved, although this shape worsen the efficiency of tritium detection.

As can be seen in Figure 5.2, a piece of methacrylate and steel was designed and built to hold the detector and two calibrated PMTs were optically coupled directly to the fiber bundle ends using optical grease [124].

The used PMTs were the model R8520-460 from Hamamatsu company [81], whose reference number are ZB2771 and ZB2773, and their signals were precessed and analyzed using the electronic configuration shown

¹Polyvinyl Chloride

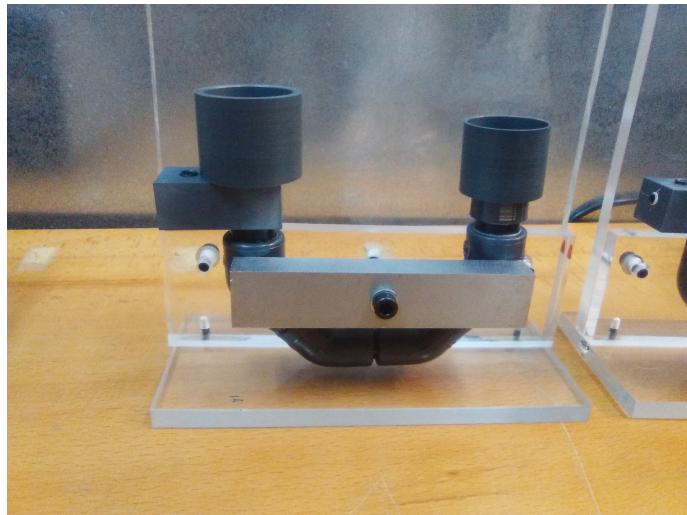


Figura 5.2 – Tritium-IFIC 0 Prototype.

in Figure 3.14b.

A commercial voltage divider electronic circuit, whose electronic scheme is shown in Figure 3.9, was used in each PMT and they were powered to -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.

Two identical prototypes were built and filled following the same protocol but using different liquid solutions.

The first prototype, called Tritium-IFIC 0 Background, was filled only with ultrapure water (39 cm^3 , uncertainty of 0.05%) and it was used to measure the background of our detector whereas the other prototype, called Tritium-IFIC 0 Signal, was filled with a radioactive liquid source of tritium, the preparation of which is explained in the appendix D.

The activity of this liquid source of tritium is 99.696 kBq/L (uncertainty of 2.24%) and the volume used to fill this prototype was the same as the other, 39 cm^3 (uncertainty of 0.05%). Therefore, the total activity

of this tritiated water sample is approximately 3.888 ± 0.087 kBq.

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

A statistically significant amount of time coincident events was not found in both PMTs, which means that the photons produced by a single tritium event cannot reach both PMTs. Therefore the measurement of time coincidence was not possible.

This loss of photons could be caused for several reasons, such as the excessive curvature in the fiber bundle due to the U-shape of Tritium-IFIC 0 prototype, which cause that too many photons escape from the fibers. This loss of photons could also be caused by poor quality of the tritiated water-fiber interface. In fact, the cleaning process explained in section ?? was motivated by this result.

To avoid this problem and obtain some results with this prototype, a measurement was performed with a single PMT. For this task, a small modification of the electronic configuration was done, in which we didn't use any coincidence stage. Therefore, the coincidence gate will be created using the PMT signals that exceed a threshold predefined by the user (the output signal of the discriminator module is connected as input of the Gate & Delay Generator module). The result of this measurement is shown in section 7.1.1, where it will be discussed.

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

The LED shown in section 4.1.3 was used to verify the reduction in photocollection efficiency of the fiber bundle due to this curve.

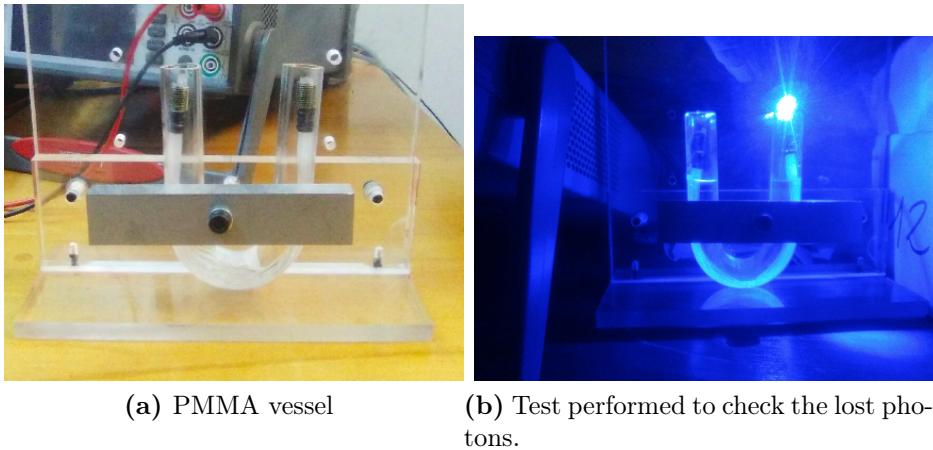


Figura 5.3 – PMMA vessel used to check photon loss due to fiber bundle curve.

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

5.1.2 TRITIUM-IFIC 1

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

1. First, as we said before, the fiber bundle is arranged straight to optimize the photon collection efficiency of the fibers.
2. Second, a special fiber cleaning protocol, previously explained in section ??, was applied on the fibers. It was used to improve the interfaces between fiber and tritiated water, creating a better wetting property

of the fiber, which will result in more tritium events detected and a greater photon collection efficiency.

3. Lastly, as we have seen in our previous fiber characterization study, shown in section 4.1.3, the photon collection efficiency of the fibers used is poor, so a large number of photons will be lost in each tritium event.

It is an innerent characteristic of the fiber which we cannot change but, to reduce its effect, we will use a Teflon vessel for Tritium prototypes.

Teflon is an interesting material for its optical properties, specifically its reflection factor, which is very close to 100% at the working wavelength. It means that practically all the photons that reach the walls of the vessel will be reflected back to the fiber.

Taking into account all this modification, the following prototype was designed, built and tested, whose name is Tritium-IFIC 1. It consists of 64 scintillating fibers, with a length of 20 cm, which are arranged in a straight position using a teflon structure, shown in Figure 5.4, in which these fibers are fixed in an 8×8 square matrix.

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

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

A general view of this prototype is shown in Figure 5.6, which, for radioactivity safety reasons, will be read using only one PMT.

The PMT used was the model R8520-460, from Hamamatsu Photonics company [81] and it was coupled directly to the fiber bundle using

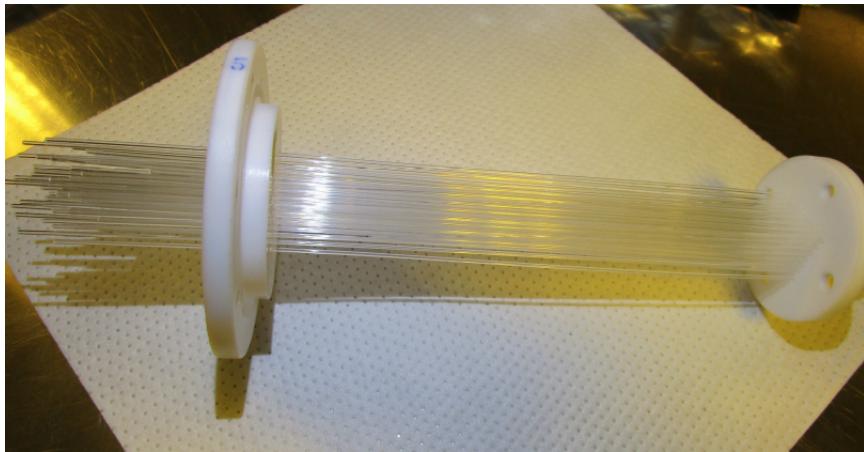


Figura 5.4 – Teflon structure used to arrange the fibers of Tritium-IFIC 1 prototype in a matrix of $8 \cdot 8$.

optical grease [124]. It was powered at -800 V, at which, its quantum efficiency is 28%.

The signal from this PMT was processed and analyzed using the same electronic configuration as that used for the Tritium-IFIC 0 prototype.

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

The result of this measurement is shown in section 7.1.2, where it will be discussed and compared with the result obtained with the previous prototype, Tritium-IFIC 0.

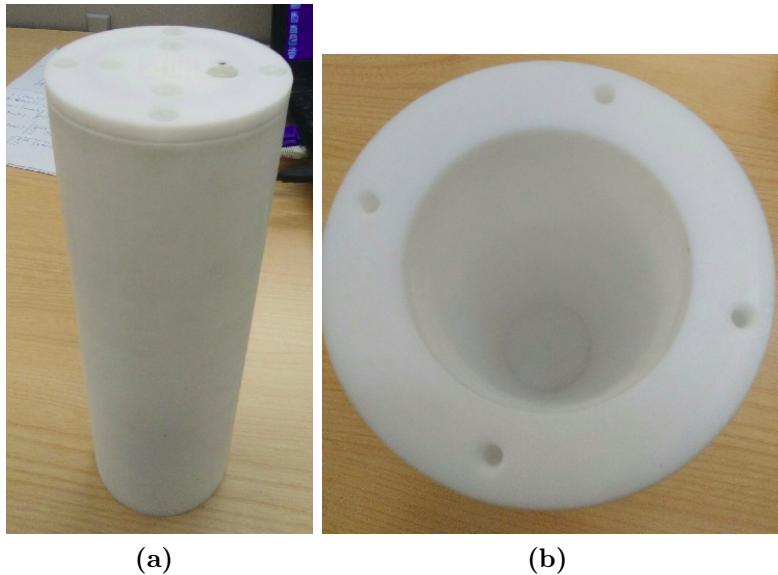


Figura 5.5 – Teflon vessel of Tritium-IFIC 1 prototype

5.2 Latest TRITIUM Prototypes

In this section the last two prototypes are shown, Tritium-Aveiro 0 and Tritium-IFIC 2, in which the problems previously found are solved and a well-defined design is developed for them.

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

In these prototypes we pay particular attention to tritium detection efficiency, which is the reason why they use many more fibers than the preliminary prototypes and a time coincidence reading of these fibers is done using two photosensors.

Furthermore, the activity of the radioactive liquid source of tritium, which is used to fill these prototypes, will be lower since we are inter-

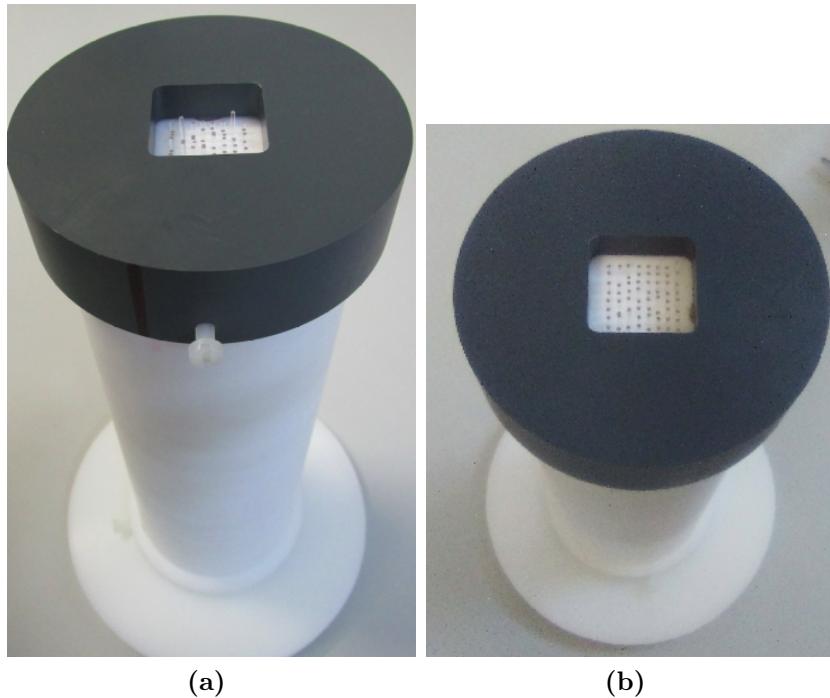


Figura 5.6 – A general view of Tritium-IFIC 1 prototype

ested in measure their sensitivity.

The design of both prototypes is very similar and their objective is to test the subtle effects caused by the little difference, such us the diameter of the fibers used, and to choose the one with the best results which will be the final design of the Tritium detector module.

5.2.1 TRITIUM-Aveiro 0

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

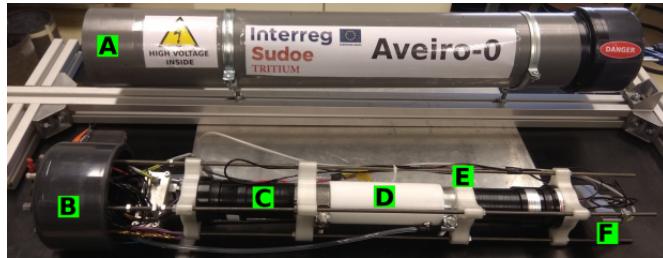


Figura 5.7 – Tritium-Aveiro prototype.

It consists of a teflon vessel (D of Figure 5.7), shown in Figure 5.8, which has an internal cylindrical hole whose diameter and length are 43 mm and 180 mm respectively.



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

This vessel contains 360 no-clad scintillating fibers with a length of 180 mm. The model of the used fibers is BCF-10 from Saint-Gobain company [129], which have practically the same characteristics than the others used up to now (BCF-12 fibers) and its most important difference is the diameter, which is the double, 2 mm.

On the one hand, a larger diameter could be interesting because it facilitates the flow of water around the fibers, reducing the problems related to surface tension and ensuring that the entire active volume of the fibers

is used for tritium detection. In addition, it increase the resistance of the fibers, which is very important since the water is flow around them.

On the other hand, a larger diameter could be detrimental since it worsens the signal-to-background ratio. It happens because, with 2 mm fibers, the active volume that our detector has in the same space is smaller so its signal will be smaller and the part of the fibers where no tritium events reach (they only contribute to the background) is larger so the background will be larger.

In order to quantify the importance of the effect of the fiber diameter, which is needed to choose the best configuration, these measurements will be compared with similar measurements performed with Tritium-IFIC 2 prototype which, as we will see in next section, is based on a similar configuration with 1 mm fibers (BCF-12 model).

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

To ensure the radiosecurity of this prototype, the teflon vessel is totally closed and it has two PMMA windows, whose thick is 10 mm, located at both ends of the fiber bundle which will be used to read it. Two clamps are used to press the Teflon walls against the PMMA windows to ensure the watertightness of the prototype.

PMMA was chosen for its optical properties, especially its transmission coefficient, which was measured for visible light range in the ICMOL

laboratories and it is shown in Figure 5.9.

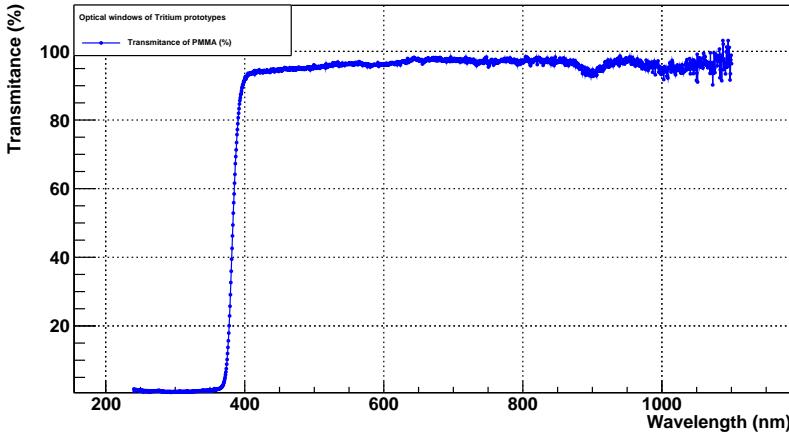


Figura 5.9 – Transmission spectrum of light (in the visible range) in a piece of PMMA of X thickness measured in the ICMOL laboratory.

As can be seen, its transmission coefficient is 95% for the working wavelength (435 nm), which means that the loss of the light in these optical windows is very small. Slightly better transmission coefficients can be achieved with other materials such as quartz or sapphire but they are much more expensive.

Two PMTs (C of Figure 5.7), powered at -1500 V, are used to read this prototype in time coincidence. They are fixed to both fiber bundle ends of the prototype using two pieces (E of Figure 5.7) which has been designed and built with a 3D printer. Both PMTs are optically coupled to the PMMA windows using optical grease [124].

The PMTs used are the model R2154-02 2" from Hamamatsu company [130], whose characteristics, specially its gain and efficiency, are quite similar to the PMTs used in the other prototypes.

All these different parts, together with the electronic system (F of Figure 5.7), is arranged in a structure, seen in Figure 5.7, which is based

on several nuts located on four long stainless-steel screws. These screws are fixed to an external PVC structure, A and B of Figure 5.7, which is used to protect the prototype from physical damage and provide a light-tight operation environment. This PVC structure is equipped with several high voltage power, low voltage power and signals feed-through connectors.

Only one prototype was built, which was designed to be installed in the Arrocampo dam. To do so, on the one hand, a water inlet/outlet were installed in its Teflon vessel to allow a constant water flux through the detector and, on the other hand, several PCBs were specially designed, developed and tested to process and analyze the signals of this system.

On the one hand, a PCB, whose electronic scheme is shown in Figure 5.10, 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 [131], one for each PMT used, which is controlled by a DAC², model MAX5500 from Maxim Integrated company [132]. 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.

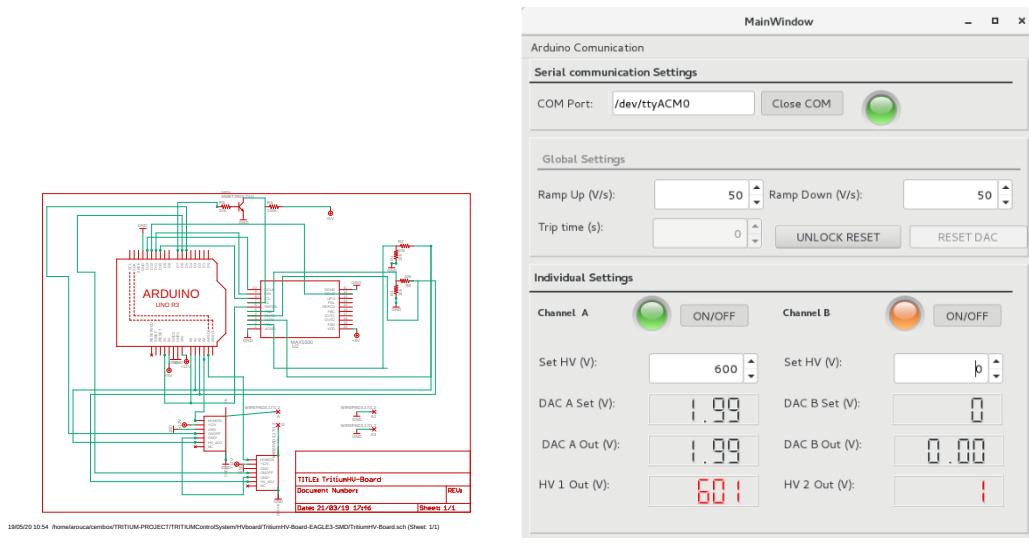
A graphical interface, shown Figure 5.10b, has been developed to manage the different options of this system in a comfortable way.

On the other hand, a electronical chain consisting of several PCBs was used to process and analyze the system signals, whose simplified electronic scheme is shown in Figure 5.11.

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 anticoincidence with a active veto.

To test this electronic chain a plastic scintillation with dimensions of $10 \cdot 10 \cdot 1 \text{ cm}^3$ was used to simulate a veto signal but four different vetos are

²DAC, Digital-to-analog converter



(a) Electronic scheme of the PCB

(b) Graphical user interface

Figura 5.10 – Electronic scheme of the PCB designed to power the PMTs of Aveiro prototype and the graphical user interface developed to control it.

being developed, each one is based on a rectangular plastic scintillations of Saint-Gobain company [133], whose dimensions are $50\text{ cm} \cdot 30 \cdot 2\text{ cm}^3$ with a PMT coupled, model R2154-02 2" from Hamamatsu company [130]. 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 5.11. As a result, each plastic scintillator will be read in anticoincidence with tritium-Aveiro prototype.

Both lines, used to process and analyze the PMT signals of the prototype, are equal and they are used to operate in time coincidence. First, each PMT signal is introduced in a prepamplifier model CR111 from CRE-MAT Inc. company [134], which is used to shape and pre-amplify 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.

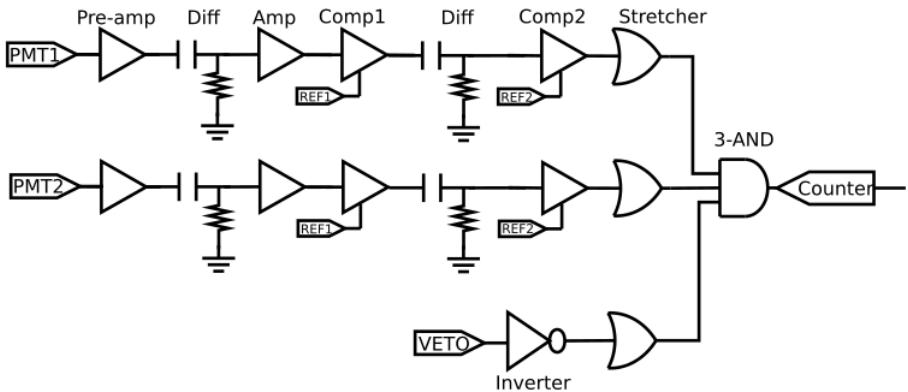


Figura 5.11 – Simplified electronic scheme used to process and analyze the signal of Tritium-Aveiro 0 prototype.

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 [135].

Then, a fast comparator, model LT111 from Linear Technology company [136], 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, $200\ \mu s$, so which too many false coincidence will be registered. To solve this problem a second differentiation stage is included and a second comparator are added to produce a 5V square signal again.

Finally a tunable pulse stretcher based on an OR gate, model SN74AHC1 from Texas Instruments company [137], 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 [138], to perform a logic level comparison. With this last stage we achieve a temporal coincidence of both PMT signals of the prototype and anti-coincidence of them with the veto signal. The output signal of this last stage is simply connected to a pulse counter.

A GPIO pins of a Raspberry Pi is used to communication with the system, control it and configure the different threshold levels and a graphical user interface, whose appearance is shown in Figure 5.12, has been developed to manage in a comfortable way the counter system.

In addition to count, which is the option normally used in our detector, this electronic system include a voltage follower circuit connected to the preamplifier output signal which can be used to obtain a energy spectrum of each PMT of the prototype.

It is important to note that, although this system has a graphical user interface that allows comfortable control of the system, the usual way in which it is controlled is remotely through the computer terminal.

In Figure 5.13 two screenshots are shown to demostrate two different situations of this system. There, we have four different signals. The yellow and cyan signal are input signals of the AND-Gate, which come from the PMT signals of the prototype. The pink signal is the third remaining input signal of the AND-Gate, which come from the PMT signal of the veto. The last signal, green, is the output signal of the AND-Gate.

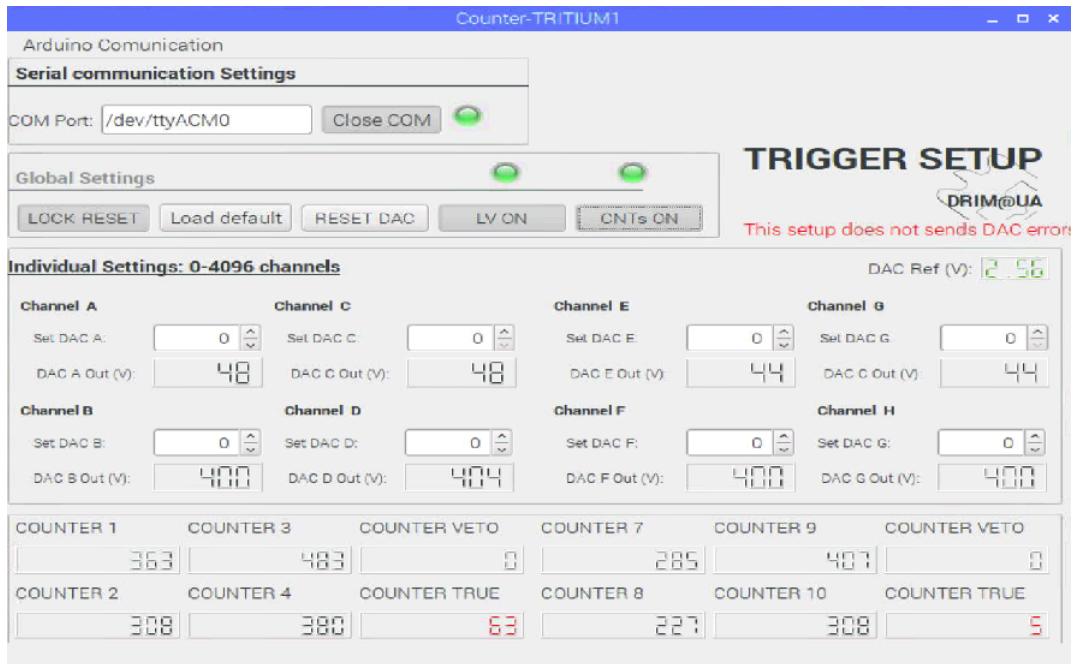
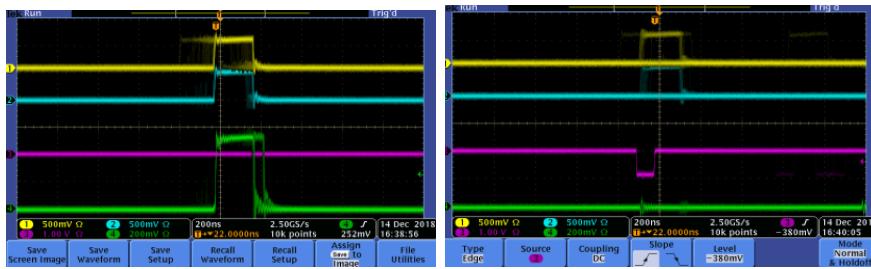


Figura 5.12 – Graphical user interface used to manage the counter system.

As can be seen, in Figure 5.13a 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 5.13b, 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.

First some measurements were taken in the laboratory that were used to characterize the detector. For this task is was filled with ultra-pure water, which was used to measure the background of the detector, and then, it was filled with several radioactive liquid tritium solutions with different activities, 10 kBq/L and 30 kBq/L, which were used the mesure the efficiency and the sensitivity of this module.

Later, it was installed in the arrocampo dam to test its functionality and to begin with the tritium level monitoring.



(a) Event accepted by the electronic system (b) Event rejected by the electronic system

Figura 5.13 – 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

All this measurements will be shown in section 7.1.3, where it will be discussed and compared with the measurements of the previous prototypes.

5.2.2 Advanced IFIC prototype, Tritium-IFIC 2

The last prototype developed in TRITIUM experiment and the second thought to be the final versión of the Tritium detector module was Tritium-IFIC 2, which is shown in Figure 5.14, A.

This prototype was designed and built in the IFIC workshop and it consists of a cylindrical teflon vessel, shown in Figure 5.15, whose shape is similar to the one used in Tritium-Aveiro 0 prototype, whose internal length and diameter are 200 mm and 36 mm respectively.

This prototype contains 800 no-clad scintillating fibers, model BCF-12, with a length of 200 mm. We can check that, as we have said, many more fibers are used than the Tritium-Aveiro 0 prototype which are arranged in less volume.

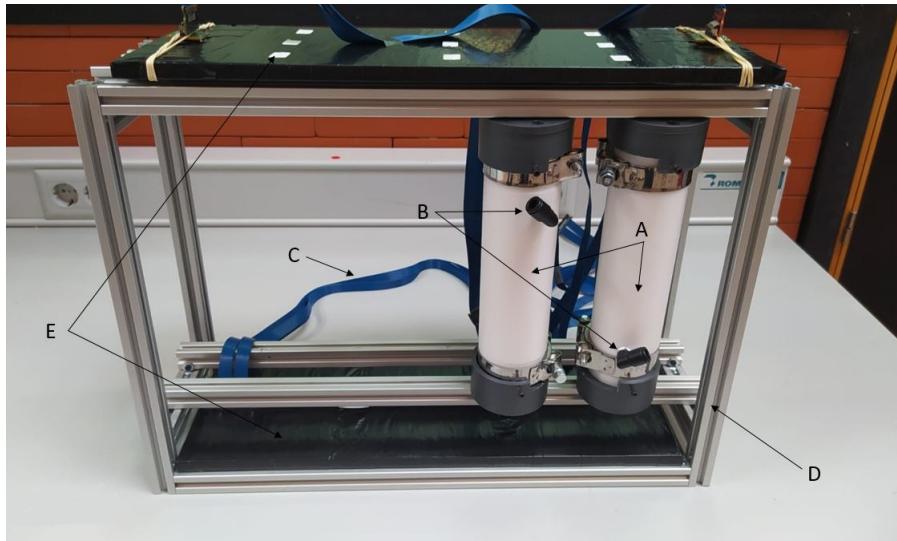


Figura 5.14 – Tritium-IFIC 2 prototype.

The fibers used are cut, polished and cleaned with the conditioning processes previously shown in section ?? since, at that time, the development of the automatic polishing machine had been completed.

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

The width of the PMMA optical windows used is 5 mm, which is sufficient to guarantee radiosecurity since we are working at very low water pressure and two clamps are used to ensure the watertightness of the prototype, similar to the Tritium-Aveiro 0 prototype. We have checked that the transmission coefficient, shown in Figure 5.9, is not affected by the little difference of the PMMA width used in both prototypes.

As can be seen in Figure 5.14, B, and Figure 5.15, a water inlet/outlet was installed in the teflon vessel of this prototype to allow a

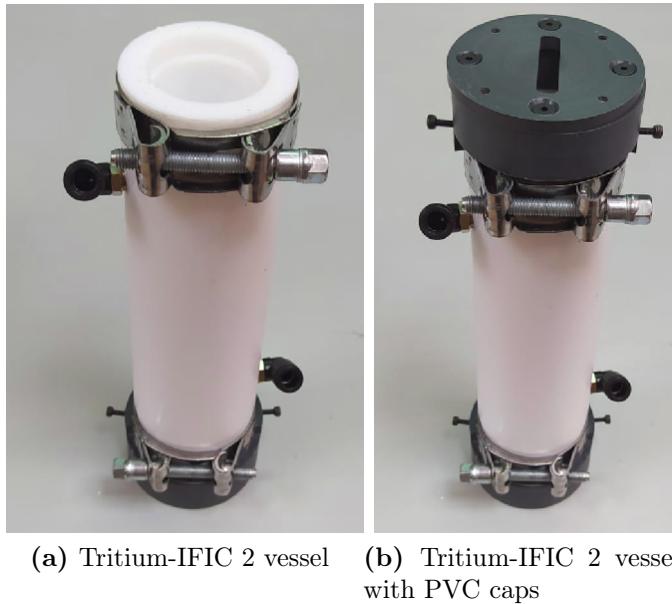


Figura 5.15 – Tritium-IFIC 2 teflon vessel.

constant water flux through the prototype, similar to the Tritium-Aveiro 0 prototype.

For the first laboratory measurements, two PMTs were used, model R8520-460 from the Hamamatsu Photonics company [81], which is useful to understand the results and compare them with the results obtained with the previous prototypes. However, measurements with SiPM arrays have already started whose output signal is connected to PETSYS system through flat wires as can be seen in Figure 5.14, C.

We have to take into account that our final objective will be to install this prototype in Arrocampo dam using SiPM arrays readout by the PETSYS system. This is the reason why we have not developed a electronics chain to process and analyze the PMT signals of this prototype.

Like the Tritium-Aveiro 0 prototype, although PETSYS has a

graphical user interface, shown in Figure 5.16, which allows controlling all the different options such as the voltage with which we feed the SiPM arrays, the thresholds used, etc., normally it will be controlled remotely via computer terminal.

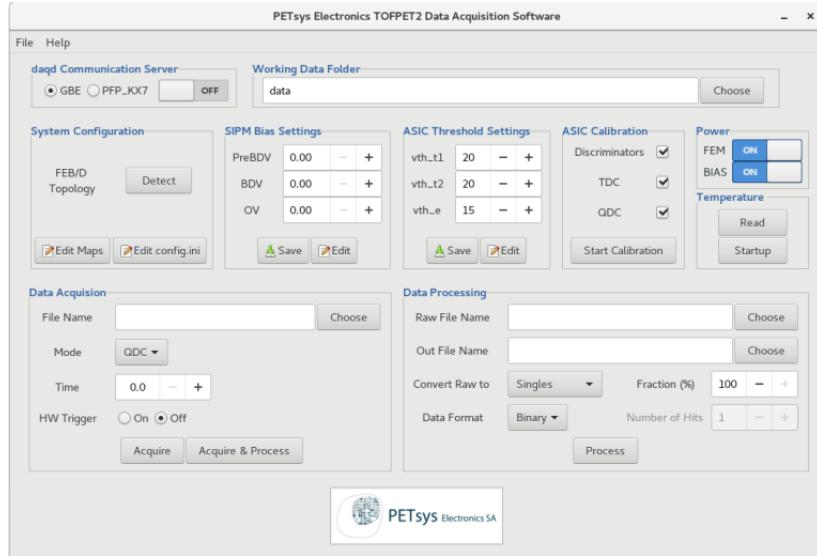


Figura 5.16 – Graphical User Interface (GUI) of PETSYS.

Two PVC caps, located at both ends of the prototype, Figure 5.15b, were used to work with the SiPMs in a light-tight environment and an aluminum structure, shown in Figure 5.14, was designed and built to house up to 10 tritium-IFIC 2 modules and two cosmic vetos, shown in Figure 5.14, E.

At this point we should note that, in the available space of the lead shielding, explained in section 3.4.1, we can accommodate up to 5 structures like the one shown in Figure 5.14. It means that our final Tritium detector is prepared to have up to 50 Tritium-IFIC 2 modules and 10 different cosmic vetos. It means that, due to the reason that our detector scales with the number of tritium modules used, the results obtained with the tritium detector can improve the results obtained with the tritium-ific 2 prototype

by a factor of 50.

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

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

The results of this prototype is shown in section 7.1.4, where they will be compared with the results of the previous prototypes and, specially, with Tritium-Aveiro 0 prototype to choose the design with the best results.

5.3 Modular TRITIUM Detector for In-Situ Tritium Monitoring

Lastly, this section presents the final Tritium detector, a schematic design of which is shown in Figure 5.17.

It consists of several Tritium modules, shown in Figure 5.17, which will be read in parallel. Each module will be the prototypes that achieves better results, Tritium-Aveiro 0 (section 5.2.1) or Tritium-IFIC 2 (section 5.2.2).

These modules will be isolated from environmental radioactivity using three different techniques.

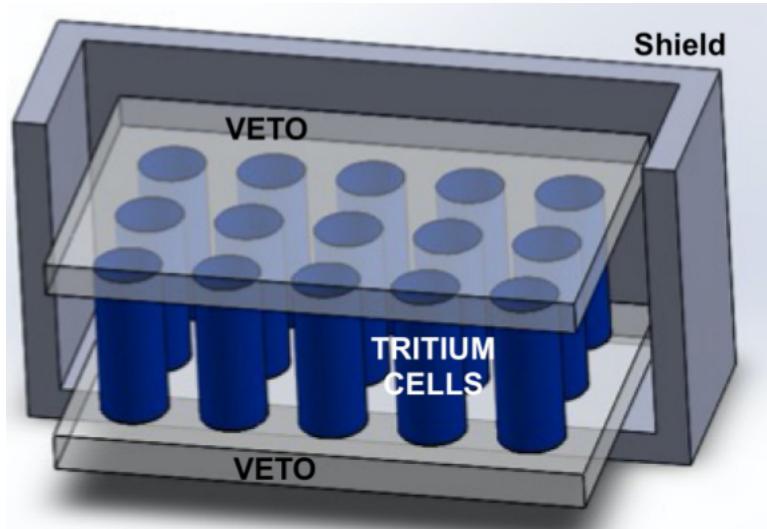


Figura 5.17 – A schematic design of the Tritium detector.

1. First, an external lead shielding, a part of which is shown in Figure 5.17, is used to stop the radioactivity coming from the place where the tritium monitor will be located before it reaches the tritium detector. It has been explained in section 3.4.1.
2. Second, several active vetos, shown in Figure 5.17, placed below and above the tritium modules, will be read in anticoincidence to eliminate the effect on the tritium measurement that has the highest energy events, mainly cosmic events, that will cross through the lead shielding and they reach the tritium modules. They have been explained in section 3.4.2 and 4.4
3. Finally, the radioactive elements present in the water samples, which will be introduced into tritium modules for their measurement, will be eliminated using an ultrapure water system, shown in section 3.3 and appendix C.

The ultrapure water system, lead shielding and a Tritium-Aveiro 0 prototype are installed and currently in operation at the Arrocampo dam.

This entire system has been used to successfully monitor the tritium levels of the water used by the Arrocampo nuclear power plant during X months.

Furthermore, two Tritium-Aveiro 0 prototypes and four active vetos are currently under manufacturing to be installed at the Arrocampo dam, which will be measured in parallel with the current prototype installed.

It should be noted that RaspberryPi, which is used to manage the counter electronic system of Tritium-Aveiro prototype, will have some counting limitations if multiple modules are used. To overcome this problem, it must be replaced with an FPGA-based counter board to ensure reliable counting.

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

One of the most important points of the Tritium detector is its modular design, with which we can achieve scalability to reach the required sensitivity, 100 Bq/L.

If this sensitivity goal is not reached with the current three modules, which will be installed soon, we only need to install some additional modules to improve it.

As we have said before, our only restriction is the available space, which is fixed by the lead shield already built and installed (it is also fixed by the available space in the house where the lead shield is installed). If it is necessary, the lead shielding and the house can be modified to enlarge the available space but is not currently under consideration.

Taking into account the currently available space, five different structures designed for Tritium-IFIC 2, one of which is shown in Figure

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

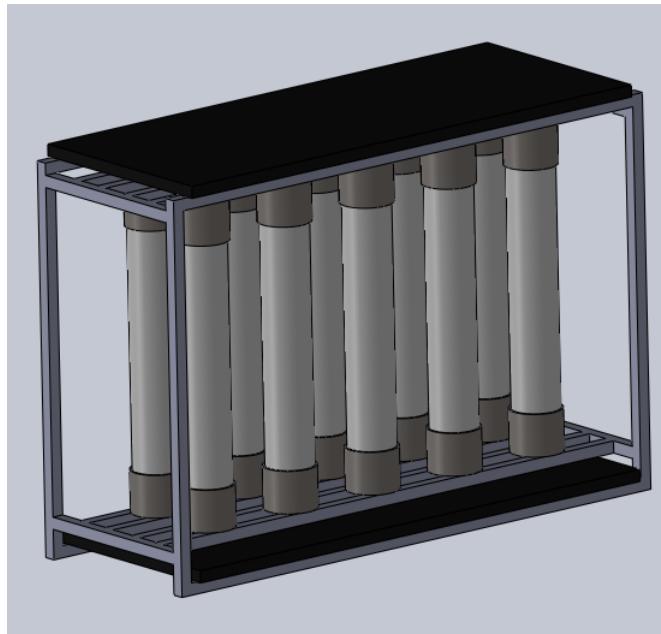


Figura 5.18 – A tritium detector design based on the Tritium-IFIC 2 prototype.

It means that up to 50 Tritium-IFIC 2 modules can be used in parallel, reducing the Tritium detector sensitivity by a factor of fifty. In light of the results, it is not expected to use more than one structure like the one shown in Figure 5.18 (ten modules) to reach the target sensitivity.

Chapter 6

Simulations

This chapter shows the Monte Carlo simulations that have been performed to choose the best design for our detector and understand its behavior. For this task, the used simulation environment is Geant4 [139, 140].

6.1 Geant4 Environment

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

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

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

A physics list used for these simulations was 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 tritium solution), PMMA (to simulate the optical windows of the prototype), polystyrene (to simulate the core of scintillating fibers), teflon (to simulate the prototype vessel), silicone (to simulate the optical grease), silicate glass (to simulate the optical windows of the PMTs) and bialkali (to simulate the photocathode material of the PMT).

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

1. First, the energy spectrums of refractive index and light attenuation was added to the water which was obtained from the reference [143]. Also a electrons emission, uniformly distributed in the water volume, was added to the water whose energy was calculated using the tritium energy spectrum. The used data was obtained from the reference [144].

2. Second, the energy spectrums of refractive index, light attenuation and photon emission was added to the polystyrene, which was obtained from their data sheet [78]. Also the scintillation yield and the decay time was included.
3. Third, the quantum efficiency spectrum was included to the photocatode material of the PMT, whose data was obtained from its data sheet [81] and a refraction index of 1.46 was used for the optical grease.
4. Finally, the optical data for the remaining materials, PMMA windows, teflon and silicate glass, were taken from the reference [145].

It is important to note that this chapter is focused on the Tritium-IFIC 2 prototype since these were the simulations I was primarily working on, but a similar simulation has been performed for the Tritium-Aveiro prototype with which important results has been obtained which will be shown in the section 7.3. In addition, other smaller simulations will be shown, such as a single scintillating fiber with various lengths or various diameters with which the effect of these parameters was quantified.

6.2 Description of the Simulations Performed

Several simulations have been designed and run during the life of the TRI-TIUM project to quantify how different values of interesting parameters affect to the tritium measurement and to understand the behaviour of the different Tritium prototype built which are shown in the following sections.

6.2.1 Tritium Source Shape Optimization

First of all the shape of the simulated tritium source was tested. The objective of this study was to find the tritium source shape that optimaze

the simulation.

Due to the reason that the mean free path of tritium decay in water is only around $5 \mu\text{m}$, there will be many tritium events from the tritium solution that won't reach the scintillating fibers. These will be tritium events that don't provide us with useful information and only contribute to being time consuming and reducing available computing resources.

To optimize the simulation, we found the shape of the tritium source that minimizes the tritium events that do not reach the scintillating fibers avoiding losing the tritium events that reach them.

The simulation that has been designed for this test consists of a scintillating fiber with a length of 20 cm and a diameter of 2 mm and a surrounding tritiated water source with the same length and a thickness 100 times greater than the mean free path of tritium electrons, that's 0.5 mm, to ensure that this study take into account all possible tritium electrons that can reach to the scintillating fiber.

We have to keep in mind that the dimensions of the fiber are not important in this study since we have only simulated energy deposition of tritium events in the fiber. That is, we have not simulated the following steps such as photon generation, propagation of these photons, etc. where the shape of the scintillating fiber becomes important.

The results of this simulation are shown in section 7.3, where it will be discussed.

6.2.2 Fiber Length Optimization

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

For this task, the Tritium-Aveiro prototype was simulated. It consists of 360 scintillating fibers readout by two photosensors, located at both ends, which consist on a windows glass and photocatode. These scintillating fibers are located inside of a teflon tube with two PMMA windows and a optical grease layer with a thickness of 0.5 mm were simulated between the PMMA windows of the prototype and the photosensors. All optical properties mentioned in section 6.1 were included in this study.

The results of this study are shown in section 7.3 where they will be discussed.

6.2.3 Fiber Diameter Effect

A third test was carried out to check the effect of the fiber diameter in the tritium measurement. For this test, the same simulation explained in section 6.2.2 was used, where a fiber length of 20 cm was choseen. Two different diameters were taken into account in this study, 1 mm and 2 mm, which are the commercial options given by Saint-Gobain company.

It doesn't have sense to test it with the tritium source since its efficiency will scale with the active surface¹ of the scintillating fiber. However, an interesting study can be performed to check how the fiber diameter affect to the cosmic detection in the fiber. It is an important result as the background-signal ratio is mainly affected by the cosmic events that hit the Tritium detector.

To do so, the CRY library² [141], [142] was used to generate this cosmic events, which remplaced the tritiated water source used in the previous simulation. The CRY library is able to generate cosmic-ray shower

¹The active surface of the scintillating fiber is the part of the surface of the scintillating fiber that is in contact with the tritiated water.

²CRY library, Cosmic-Ray Shower library

distributions for different particles (muons, neutrons, protons, electrons, photons and pions) at several heights (see level in our case).

The cosmic sources shape used in this simulation is a square of $1 \cdot 1 \text{ m}^2$ located at a height of 35 cm.

The result of this simulation will be shown in section 7.3, where it will be discussed.

6.2.4 Simulation of the Tritium-IFIC 2 Prototype

The Tritium-IFIC 2 prototype simulation was the last simulation developed in the TRITIUM experiment and it was the one I mainly focused on. This simulation is similar to the previous simulation of the Tritium-Aveiro 0 prototype simulation since the design of both detectors are quite similar. There are two main difference between both simulated prototypes:

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

The Tritium-IFIC 2 prototype simulation consists of 800 equispaced fibers distributed in sixteen different circles with increasing radius, which are shown in Figure 6.1. The fibers simulated has a diammeter of 1 mm and the optical properties, mentioned in section 6.1, has been added

to them, whose values has been obtained from the data sheet provided by the manufacturer.

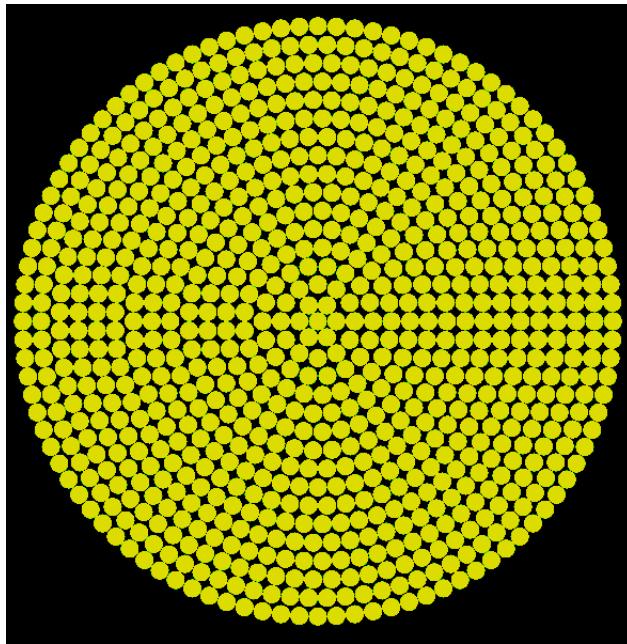


Figura 6.1 – Distribution of the scintillating fibers in the simualtion of Tritium-IFIC 2 prototype.

The used tritiated water source consists of a tritiated water volume with a thickness of $5 \mu\text{m}$ around each scintillating fiber. The Tritium events will be uniformly distributed in all the tritiated water volume.

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

Finally, two PMTs, model R8520-460 from Hamamatsu company [81], were simulated in both ends. The data for the dimensions of them and their quantum efficiency spectrum were obtained from the data sheet

provided by the manufacturer.

The optical properties used for the tritiated water, teflon vessel, PMMA windows and the optical grease are exactly the same as those used for the Tritium-Aveiro 0 prototype simulation.

This simulation is shown in Figure 6.2 in which can be appreciated the PMTs (black), the optical grease (blue), PMMA windows (white), tritiated water (green) and scintillating fibers (yellow). In this image, the Teflon container was not drawn to allow its interior to be seen and several volumes of tritiated water were also not included to allow several scintillation fibers to be seen.

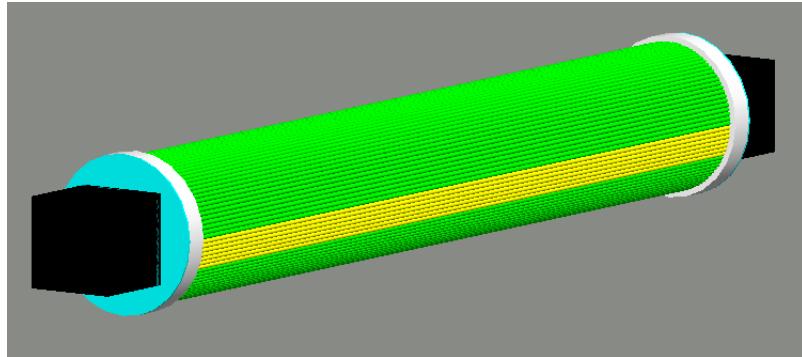


Figura 6.2 – Simualtion of Tritium-IFIC 2 prototype. PMTs (black), the optical grease (blue), PMMA windows (white), tritiated water (green) and scintillating fibers (yellow)

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

6.2.5 Simulation of the Lead Shielding and Cosmic Veto

Finally the lead shielding and active vetos were included in the simulation of the Tritium-IFIC 2 prototype, explained in section 6.2.4. The objective of these simulations was to quantify their effect in reducing cosmic events detected by the prototype and to demonstrate their necessity.

For this task, similar to that done in section 6.2.3, the tritium source was not simulated. Instead of, a cosmic events source was used, which was simulated through the CRY library previously explained.

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

The optical properties given to this plastic scintillators were the same as the one used for the fibers, which are the refractive index and the light attenuation spectrum, whose values are the same that those used for the fibers, and energy emission spectrum, whose values was obtained from their data sheet provided by the manufacturer [111].

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

Finally, a lead shielding was simulated, whose properties were taken from the Geant4 NIST database. Due to the reason that only one Tritium detector module was simulated, it was not necessary to simulate its real dimensions since, in this case, we would need to simulate a larger cosmic veto source and most of this events will not reach neither our active veto nor our Tritium detector module, just contributing to being time consuming and reducing available computing resources.

Instead of that, the dimensions of the lead shielding were smaller

to optimizate the simulation since this change don't affect to the results. The dimensions used were 60 cm long, 60 cm wide, 70 cm high, which is the minimum needed to accomodate the active vetos and Tritium detector module inside. When it is compared to the dimensions mentioned in section 3.4.2, it can be noted that the length of this lead shielding is smaller than the real dimension, 148 cm.

The results of these simulation will be presented in section 7.3, where they will be discussed.

Chapter 7

TRITIUM Monitor Results and Discussion

Comparar las cuentas que espero teóricamente con las obtenidas experimentalmente para tritium IFIC 2. Sale bastante bien.

Ajustar una curva landau a los resultados de cada espectro energético.

Tritium 0 -> Tritium y fuentes radiactivas.

Tritium 1-> Tritium y comparación con Tritium 0

Aveiro -> Calibración inicial, Medida y comparación con la simulación, Reducción de cuentas debida al plomo, figuras 8a y 8b del paper experimental de Carlos, estudio teórico del límite de curié, medida en aire y agua, medida con 10 kBq/L, extensión a mayores tiempos,

Tritium 2 -> Medida y comparación con 1 y 0 (y si se puede con Aveiro). Resultados de la tabla del punto 1.3 de la tesis donde comparo con otros experimentos similares, estabilización, extrapolación teórica.

Finally, the simulated and the experimental spectra are compared, yielding some conclusions regarding the PMT signal and the behavior of the scintillator in tritiated water.

7.1 Results from Laboratory measurements

Mostrar los resultados por unidad de centelleador (unidad de fibra o unidad de superficie) para poder comparar bien los prototipos.

Demostraciones teóricas de las cuentas esperadas en los detectores (en cada apartado)

7.1.1 Experimental Results of TRITIUM-IFIC 0 Prototype

7.1.2 Experimental Results of TRITIUM-IFIC 1 Prototype

7.1.3 Experimental Results of TRITIUM-Aveiro 0 prototype

- Espectro energético.

- Monitorización del fondo durante meses. Desde el mes 3 hasta el mes 8

- Monitorización del fondo y de una disolución de Tritio durante unos días. Estudio de las gaussianas de su distribución

- Effecto del apantallamiento del plomo.
- Volume used.

The first detector module was characterized, commissioned and installed in the discharge channel of Arrocampo dam to the Tagus river. Due to the high sensitivity of the detection module it requires radioactive background mitigation techniques through the use of active and passive shielding. We have obtained a Minimum Detectable Activity lower than 5 kBq/L for a single module being limited by the cosmic background. Far from the primary goal this value is already compatible with a real-time tritium environmental surveillance monitor and a power plant pre-alert system.

Medida con una fuente de ^{55}Fe , ya que tiene gammas muy cercanas a las del tritio, 5.9keV. Esta se situó dentro del tubo de teflón ya que si no se apantallaría y no llegaría a la fibra. Debido a esto no podíamos usar agua en su interior.

7.1.4 Experimental Results of TRITIUM-IFIC 2 prototype

Una sección con la monitorización de la señal y el fondo.

Rellenar con una actividad grande y volver a medir.

Vaciar el prototipo y llenar con actividades más pequeñas. 1000 y 100 Bq/L.

Espectro de tritio + fondo medido y sacar la significancia y la relación señal ruido y todo lo que se pueda.

Comparar fondos en el laboratorio, Cáceres, Almaraz, etc.

COmparar cada propuesta (Aveiro valencia) -> Publicaciones de cada uno y una de las sintesis.

Medir en el prototipo con SIPM.

7.2 Results from Measurements at Arrocampo dam

7.3 Results of the simulations

In this work we present simulation results for a modular tritium in-water real-time monitor. The system allows for scalability in order to achieve the required sensitivity. The modules are composed by 340 unclad scintillating fibers immersed in water and 2 photosensors in coincidence for light readout.

- Simulación del tritio. Vemos que solo se detecta tritio hasta 5 micras. Debido a esto solo simularemos aros de tritio de espesor X.

Vemos la necesidad de tener que utilizar fibras sin clad. También vemos que el espectro de deposición de energía esta centrado en 10 keVs ya que las emisiones de energía pequeña pocas veces llegan a la fibra.

Mostrar aquí la simulación del espectro de tritio. Si eso mostrarla superpuesta a la referencia 10 del paper de carlos (figura 2)

- Simulación del tamaño de las fibras. Vemos que es mejor fibras cortas

Este experimento a consistido en simular

- Simulación fibras de 1 mm y 2 mm. Cósmicos.
- Incluido el coeficiente de Birks ya que este afecta bastante. Mostrar como afecta.

Light yield and Birks' coefficient uncertainties for low energy beta particles is discussed.

A study of the detection efficiency according to the fiber length is presented.

Simulación de cosmisos. Vemos que el resultado es mejor en fibras de 1 mm

- Expandimos esto a tritium ific 2. Empezar por todos los pasos que sigue la luz y demas y ver que todo se simula correctamente (similar a como se hace en el TFM). Luego mostrar los resultados.

Due to the low energetic beta emission from tritium a detection efficiency close to 3.3% was calculated for a single 2 mm round fiber.-> Para Tritium-Aveiro 0 prototype

- Añadimos el blindaje de plomo
- Añadimos los vetos activos
- Suma de ambos efectos.

Si pongo resultados de Tritium Aveiro genial, si no tengo que cambiar el último parrafo de la sección Geant4 Environment.

Chapter 8

Conclusions and Prospects

Que cosas se han conseguido en este experimento? -> DECIR que tanto lo que se ha conseguido con el detector como con las investigaciones de componentes del detector (capítulo 3)

Responder a las grandes preguntas:

- Podemos medir tritio?
- Lo podemos hacer en tiempo quasi real?
- Lo podemos hacer a la actividad que queríamos?
- Que sensibilidad se ha llegado a conseguir?
- Estabilidad temporal?
- Precio?
- Comparación con respecto al resto de experimentos? -> Poner la tabla 1.8 pero incluyéndonos
- Efecto del shield

- Efecto de los vetos
- Efecto de ambas cosas
- Medidas a varias actividades

Hemos llegado a detectar 30 kBq/L con Tritium-Aveiro y se espera llegar a medir hasta menos de 5 kBq/L (superando los actuales límites).

Hemos llegado a detectar 10 kBq/L con Tritium-IFIC 2 (superando los actuales límites) y se espera llegar a medir incluso menos.

Ambos valores, lejos de ser el objetivo del proyecto, sirven para una monitorización en tiempo quasi real. Además, con el monitori final que consiste en varios modiules de estos en apralelo, se pretende llegar al objetivo deseado.

Tenemos datos de tritio en el agua bruta (agua del río) de esa zona desde 1998, pero tengo que solicitar permiso para poder dártelos. En cualquier caso, hay otra manera de conseguirlos, que es a partir de los informes del CSN al Congreso de los diputados (web CSN). Desde 2015 la concentración de tritio en el agua del río Tajo ha disminuido considerablemente porque la CNA instaló unos enfriadores por convección que emiten parte del H3 a la atmósfera. -> Tesis de Antonio Rodríguez y de Elena García.

Appendices

Appendix A

Birks Coefficient Study

Aún faltan cosas por decir.

Appendix B

Electronical Schemes of PCBs Used for SiPM Characterization

Y más cosas aún.

Appendix C

Ultrapure Water System

In this appendix I show several photos of the ultrapure water system in the same order that the water flows through them.

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

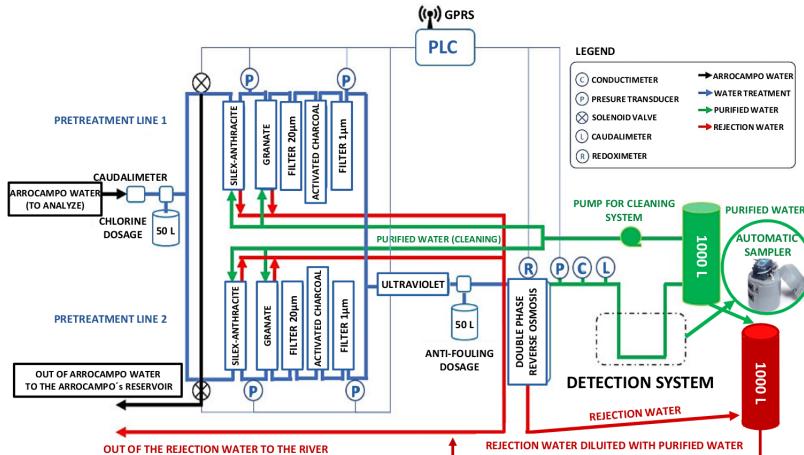


Figura C.1 – Scheme of the ultrapure water system.

Secondly, the Gross filtering stage, made up of Silex-Antracite and

Granate filters, is shown in Figure C.2a:

In third place, the fine filtering stage, consisting of $20\ \mu\text{m}$ filter and active carbon filter, is shown in Figure C.2b:

In fourth place, the superfine filtering, composed of the $1\ \mu\text{m}$ filter and the UV lamps, is shown in Figure C.2c

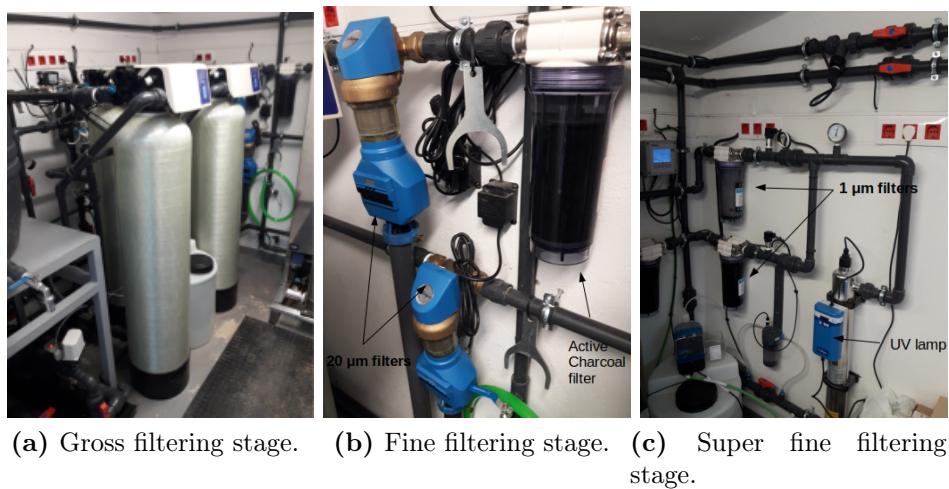


Figura C.2 – Different stages of filtration of the ultrapure water system.

In fifth place, the double phase reverse osmosis is shown in Figure C.3a

In sixth place, the containers in which we store the ultrapure water and the reject water after treatment is shown in Figure C.3b.

In seventh place, the Siemens PLC, software used to control the ultrapure water system, is shown in Figure C.4.

Finally, the complete system of the ultrapure water system is shown in Figure C.5

Just as a curiosity, the three types of water (raw water, rejection



(a) Doble phase reverse osmosis stage and (b) Storage containers of reject and ultrapure water.

Figura C.3 – Doble phase reverse osmosis stage and containers used to store the outlet water of the ultrapure water system.

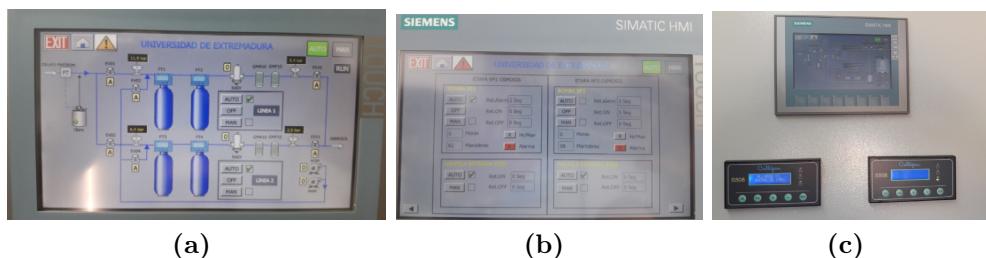


Figura C.4 – Siemens PLC, software for remote control of ultrapure water system.



Figura C.5 – General photo of the complete ultrapure water system.

water and ultrapure water) are shown in Figure C.6, where you can visually check the difference in the turbidity of each type of water.

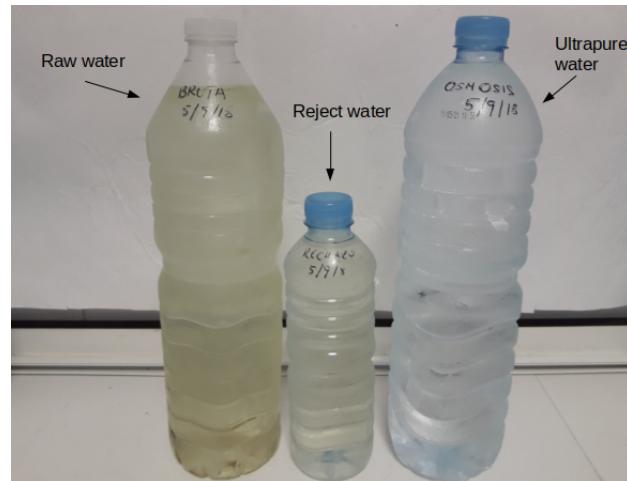


Figura C.6 – Raw water, reject water and ultrapure water obtained with this system.

Appendix D

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 PTB¹, which has a serial number of 2005 – 1442 and reference number of PTB-6.11 – 285/03.2017 [128]

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.

¹Physikalisch-Technische Bundesanstalt, Braunschweig and Berlin, Germany

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Appendix E

Electronic System of TRITIUM-Aveiro 0 prototype

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