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TRITIUM: Design, construction and commissioning of a tritium water detector based on scintillating fibers read out by silicon photomultiplier

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Dedicated to
my family

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Y PENSAR GENTE QUE ME DEJO POR EL CAMINO.

Abstract

Tritium is one of the most frequently 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 legislations 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 at 100 Bq/L, or the U. S. Environmental Protection Agency, 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 a off-line method whose measurement process 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 pasive shielding for reduce the natural background of our system and an ultra pure 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 consisting of several detectors whose objective is to reduce the impact of Nuclear Power Plants to the environment.

Nomenclature and acronyms

Acronyms:

$OECD$	—	Organisation for Economic Co-operation and Development
Btu	—	British termal unit
UN	—	United Nations
$UNFCCC$	—	The United Nations in the framework convention about Climate change
$ITER$	—	International Thermonuclear Experimental Reactor
NPP	—	Nuclear Power Plants
DOE	—	Department of Energy
$U.S.A.$	—	United States of America
$U.S.$	—	United States
EPA	—	Environmental Protection Agency
LDL	—	Lower Detection Limit
PWR	—	Pressurized Water Reactor
BWR	—	Boiled Water Reactor
HWR	—	Heavy Water Reactor
GCR	—	Gas-Cooled Reactor
$PHWR$	—	Pressurized Heavy Water Reactor
$quasi-real$	—	Less than 10 minuts
LSC	—	Liquid Scintillation Counting
LWR	—	Liquid Water Reactor
STP	—	Standard temperature (0°C) and pressure (1 atm)

IC	—	Ionization chamber
$BIXS$	—	Beta Induced X-ray Spectrometry
PMT	—	PhotoMultiplier Tube
SDD	—	Silicon Drift Detector
EEC	—	European Economical Community
PMT	—	PhotoMultiplier Tubs
$SiPM$	—	Silicon PhotoMultiplier
$CNRS$	—	Le Centre National de la Recherche Scientifique, France
$ALARA$	—	As Low As Reasonably Achievable

Atomic and nuclear symbols

CO_2	—	Carbon Dioxide
CH_4	—	Methane
N_2O	—	Nitrous oxide
HFC	—	Hydrofluorocarbons
PFC	—	Perfluorocarbons
SF_6	—	Sulfur Hexafluoride
${}^{1}_{1}H$	—	Hydrogen
${}^{2}_{1}H$	—	Deuterium (Non-radiactive hydrogen isotope, 1 neutron)
${}^{3}_{1}H$	—	Tritium (radiactive hydrogen isotope, 2 neutrons)
${}^{4}_{2}He$	—	Helium
${}^{3}_{2}He$	—	Isotope of the Helium(Non-radiactive, 1 neutrons)
${}^{14}_{7}N$	—	Nitrogen
${}^{12}_{6}C$	—	Carbon
${}^{6}_{3}Li$	—	Lithium isotope
${}^{7}_{3}Li$	—	Lithium
${}^{10}_{5}B$	—	Boron
${}^{16}_{8}O$	—	Oxygen
${}^{222}_{86}Rn$	—	Radon
${}^{40}_{19}K$	—	Potassium

$^{137}_{55}$Cs	—	Cesium
n	—	free neutron
W	—	watt
h	—	hour
g CO_2/kWh	—	grams of CO_2 per kilowatt hour
L	—	Liter
Bq	—	Becquerel, Nuclear decay number per second
Bq/L	—	Becquerel per liter
Ci	—	Curios
Ci/L	—	Curios por litro
H_2O	—	Usual water
D_2O	—	Heavy water
HDO	—	Semi heavy water
yr	—	year
$T_{1/2}$	—	Half-life time of a radioactive element
β	—	Beta decay
$\overline{\nu}_e$	—	Electron antineutrino
e^-	—	Electron
γ	—	Gamma
p	—	Proton
α	—	Alpha (nucleus with two protons)
σ_i	—	Cross section of the "i" process
η_{det}	—	Intrinsic detector efficiency
F_{sci}	—	Active surface of the plastic scintillator
ε_{det}	—	Specific detector efficiency
μS/cm	—	MicroSivers per centimeter

Añadir en un futuro:

$D\&D$	—	Decontamination and Decommissioning.
DWS	—	Drinking water standars

UDL	—	Upper Detection Limit
NA	—	Numerical Apertures
$PMMA$	—	Polymethyl Methacrylates
UV	—	Ultraviolet
WLS	—	Wavelength shifter
T	—	Temperature (°C).
V	—	Volume (m^3).

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Chapter 1

Introduction

1.1 Global energy context

The energy necessities around the world has been increased a 60% in the last 25 years and they are growing each day due to, mainly, the strong population growth (mundial population has been increased in a 40% between 1990 and 2015) and the fast development of emerging countries like China, India and Brazil. [1].

On top of that the outlook is that these energy necessities will keep increasing as you can see in the figure 1.1a, specially for countries which don't belong to the OECD like countries which I have said before. In this figure you can see that the energy quantity which will be used on 2040 is expected to be the double of the one which we used in 2000.

Nowadays, as you can see in the figure 1.1b, the most used elements for getting the energy that we use are liquid fuels, coal and natural gas, namely, natural elements. This fact has two problems. On the one hand, natural elements are limitated and this is a problem due to the huge

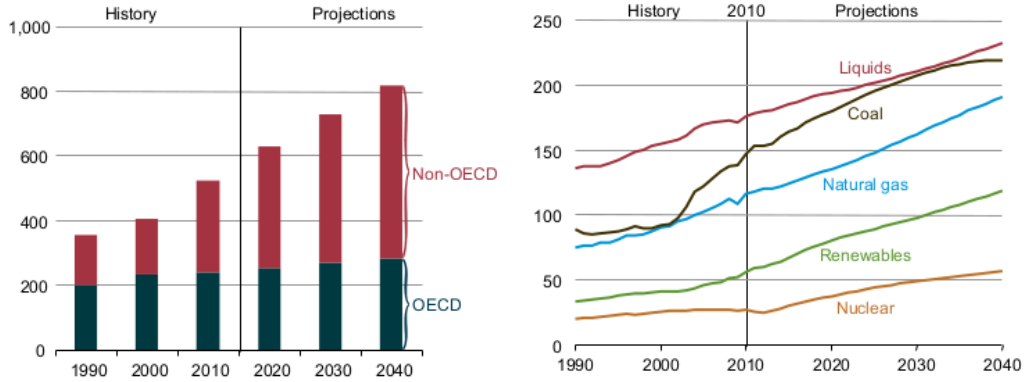


Figura 1.1 – World energy consumption from 1990 up to now and outlooks for the future until 2040 (units in 10^{15} Btu) [2]

increasing of the energy consumed in the world and, on the other hand, obtaining energy from these natural elements produces large amounts of greenhouses gases (mainlty CO_2) which contribute to the environmental contamination, global warming, deforestation, etc. and this is a very big problem, specially right now, since United Nations (UN) did a communication on November 25, 2019 [3], where they claimed that the lifestyle of the world population has not changed in the last years and, as a reason of that, we achieved the highest level of the CO_2 in the history.

In order to control these emissions, the United Nations in the Framework Convention about Climate Change (UNFCCC), has developed a protocol, whose name is Kyoto [4]. The objective of this protocol is to control and reduce the global negative envirionmental impact. It is focused on 6 different gases, carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O) and other three types of fluorinate industrial gases (HFCs, PFCs and SF_6), which are related with the greenhouse effect and whose consecuence is the global warming. This convention only encourage to belonging countries to the United Nations to reduce their greenhouses gases emissions but this protocol commits the countries who has signed it to do so.

Therefore, we currently have a problem because, on the one hand, we want to maintain, even increase, this economic growth and for that we need to produce as much energy as we can but, on the other hand, we need to reduce the negative environmental impact. Hence, what we need is a energy source with which we can get a big quantity of energy with a very low greenhouse gases emisions. One possibility which we have is the nuclear fusion plants. They don't emit greenhouse gases and with its energy, which is practically limitless, we could satisfy the humanity's energy necesities .

The largest representative in this sector is the ITER [5] (International Thermonuclear Experimental Reactor) that is going to be the largest fusion reactor in the world. The ITER is currently under construction in Cadarache, France, and its objective is to provide the concept of sustained fusion. The reaction which ITER try to reproduce in the earth is the fusion reactor using deuterium (${}^{2}_{1}\mathrm{H}$) and/or tritium (${}^{3}_{1}\mathrm{H}$) because it is the only one whose requirements of temperature (several hundreds of millions degrees) and pressure we can fulfill in the earth. Among the possible combinations between both (table 1.1), the choice of ITER is the nuclear reaction between deuterium and tritium in a 50:50 mixture because, as you can see in this table, it is the nuclear reaction with which we get the highest energy release.

Reaction	Products	Energy gain (MeV)
${}^{2}\mathrm{H} + {}^{3}\mathrm{H}$	${}^{4}\mathrm{He} + \mathrm{n}$	17.6
${}^{3}\mathrm{H} + {}^{3}\mathrm{H}$	${}^{4}\mathrm{He} + 2\mathrm{n}$	11.3
${}^{2}\mathrm{H} + {}^{2}\mathrm{H}$	${}^{3}\mathrm{H} + {}^{1}\mathrm{H}$	3.98
${}^{2}\mathrm{H} + {}^{2}\mathrm{H}$	${}^{3}\mathrm{He} + \mathrm{n}$	3.25

Table 1.1: Fusion reactions between deuterium and tritium[6]

Nevertheles, the fusion power plants is not already prepared for working, they are still in experimental phase and its researchers need to solve some important problems like inestabilitiy vortex, materials which withstand such high temperatures, etc. [7]. Hence, although we know that

the nuclear fusion plants will be the energy of the future, nowadays, we have to wait until all their problems has been resolved.

Other possible energy source is the existing Nuclear Power Plants (NPPs). With the NPPs we can practically avoid the problem of the greenhouse gases emission. We have to take into account that, although the nuclear fission reaction doesn't emit greenhouse gasses, the total procces to obtain the energy, which involves the uranium mining and milling, transport, uranium enrichment, etc., has a small contribution to the annual release of greenhouses gases. These emissions are difficult to estimate because, on the one hand, they depend on the NPP that we consider (for instance, there are studies which show that asian NPPs has higher emissions [8]) and, on the other hand, there are some tasks whose greenhouse gases emission are difficult to quantify[8].

There's exist a study [8] which analyzes 19 different studies of different NPPs. His estimation for the total greenhouses gases emision of a NPP is 66 g CO_2/kWh which was obtained as a average of these 19 studies considered. In the table 1.2 this estimation is compared with the estimation for other energy kinds. There you can check that, the emissions due to NPPs is much more smaller, one order or more, than the emissions from burning natural elements.

NPPs are already working and, nowadays, they are essential for providing a big part of the electic power that is used in the current world (more than a 20% in Spain as you can see in the table 1.3).

NPP is one of the cheapest source. It is a stable source which doesn't depend on meteorological parameters and, 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 develop enough to fully supply ourselves with them.

Technology	Estimate (g CO_2/kWh)
Wind	$9 - 10$
Hydroelectric	$10 - 13$
Biogas	11
Solar thermal	13
Biomass	$14 - 41$
Solar PV	32
Geothermal	38
Nuclear	66
Natural gas	443
Fuel cell	664
Diesel	778
Heavy oil	778
Coal	$960 - 1050$

Table 1.2: Estimations of CO_2 emissions for several kinds of energy sources[8]

Type of energy source	Contr.	Type of energy source	Contr.
Nuclear	22.0%	Wind	20.9%
Coal	4.2%	Hydraulics	9.7%
Combined Cycle	20.1%	Solar Photovoltaic	3.5%
Cogeneration	11.8%	Solar thermal	2.0%
No-renewable waste	0.8%	Other renewables	1.4%
Pumping turbine	0.6%	renewable waste	0.3%
		Impoted balance of international exchanges	2.7%

Table 1.3: Contribution of each energy source to the total energy consumed in Spain in 2019 [9]

The detractors of nuclear energy argue that NPPs facilitate nuclear proliferation or there are a risk of radiactive contamination and accidents like it happened in the past: Chernobyl, Fukushima and other accidents with lesser impact such as Three Mile Island, near to Pensilvania, USA [10].

Although we know that the nuclear energy is not the energy of the future since it produces nuclear waste which, by the moment, we don't know how we can eliminate, it is difficult that we leave to use nuclear energy because, now, we don't have a better solution for obtaining the energy which we need.

In Spain the government are doing an effort in order to remove all nuclear power plants since they are not going to build new nuclear reactors and they are only waiting until the NPPs have reached the end of their useful life (approximately 40 years). They expect to close all NPPs between 2020 and 2030 [11].

Other countries like France, where the 77% of their energy consumed is obtained from nuclear sources, prefer to maintain their nuclear facilities and there's even exist other countries that believe that nuclear energy is a safe investment like China who announced in 2016 that they were going to build 60 new nuclear reactors in the next dedade [12] or USA, who made an investment of 35 million of euros in 2019 for development and improvement of nuclear power plants [13].

In any case it is not important if we agree or not with nuclear energy source. The only important thing is that the nuclear energy production in the world is not going to stop in the next decade, in fact, it will increase as you have seen in the outlooks of the figure 1.1b. Therefore the development of different types of alarm systems is a good investment of both, time and money. Safety is not a negotiable aspect and there must be mechanisms that warn us of any malfunction of a nuclear power plant.

Hence, our work has based on the development of a monitor that we can use as early alarm in case of any problem happen in a NPP.

Generally, a nuclear reactor, which is working in normal mode, is characterized by extreme stability and, therefore, by a constant emission of radioactive isotopes so the first alarm signs of any malfunctioning of a NPP is a variation of this radiactive emission rate.

Between all the radioactive elements which is produced in a nuclear power plant, the most frequently produced is tritium as DOE complex [14] [15] and other research facilities in China [16] have seen in their installations and in ground water, surface water, and process waste water around their facilities. However, as we will see in the section 1.3, the current methods which is used for monitoring this radioactive element has some limitations.

For these reasons, the radiactive element which we have chosen for monitoring with our early alarm system is tritium. We have focused our alarm system for working with NPPs but it could be also interesting for other tasks where tritium is involved like monitoring the behaviour of future fusion nuclear plants (ITER will need up to several tens of kilograms of tritium for working, which correspond to several TBq of tritium) or any nuclear research facility (tritium is a commun emission of these places [17],[18]), tracking the movement of tritium contaminated plumes in ground water [19] or demonstrate the compliance with the government agencies which fix the limit of the emitted radionuclides to the environmental.

We have to take into account that the limit of the emission of each radioactive element depends on the government agency who manages it, and the regulation directives that is implemented in that place so, as a consequence, it is different in each country. For example, in Europe, the agency is Council Directive and the limit which they have stablished for tritium in drinking water is $A = 100$ Bq/L [20]. In USA, the organization is United States Envirnomental Protection Agency (U. S. EPA) and this limit

is $A = 20$ nCi/L $= 740$ Bq/L [21].

Tritium is normally produced in the water that there are in the cooling system or the moderator of some NPPs. It usually appear by neutron capture of the deuterium, which exist in the heavy water (D_2O), semi-heavy water (HDO) or the deuterium which has created by neutron capture in usual water (H_2O). All these processes have a big probability to happen due to the huge quantity of neutrons which we has in the nuclear reactor, 10^{14} n cm^{-2}s^{-1} [22].

The quantity of tritium will be different for each reactor type because, as we will see in the next seccion, the cross section of tritium production will depend on the materials which there are in any type of NPP. In the table 1.4 we can see the emissions of different types of nuclear reactors:

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 1.4: Emission of tritium in different types of nuclear reactors[16]

The tritium which is created in the water of the cooling system is finally released partially or totally to the environment. Between these types, the most common way is HTO [16].

Our alarm system will monitor the activity of tritium in the water of the cooling system of the NPP which is emitted to the environmental. It can also work for the water of the moderator in these types of NPP but it's not our objective because it's a close circuit so it's not a emission (unless the moderator is leaking, in which case our alarm system would detect it indirectly due to a variation of tritium activity in the water of the cooling system released to the environment).

The measurement of the tritium activity is one of the systematic environmental control which is performed during energy production by NPPs. It is normally done by LSC technic which has a very good detection capability and precision but it has some problems, for example, it needs too long time for taking a measurement (2 days or more). I will speak more about LSC technic in the section 1.3.

The detection of this tritium in quasi-real time ($<$ 10 min) is important because of the following reasons:

1. It can warn us about the production of an excesive number of neutrons in the nuclear reactor due to the overheating of itself or a leakage of the water from the primary circuit of the cooling system in a nuclear power plant due to some break (perhaps because of an excesive preassure, other alarm sign of a malfunctioning of a nuclear reactor). Both causes could become in a very dangerous problems so the tritium detection in quasi-real time could be important in order to quickly detect and to solve it.

2. The water, which there are in the secondary circuit of the cooling system, will be released to the eviromental, usually rivers or seas, after using it for cooling purposes. Generally this water will be used later for human consuming, irrigation of all kind of plantations or it will arrive to a places where we fish.

   Due to such a low legal limit in comparation with the activities of tritium inside of a nuclear reactor, it is possible that, if the nuclear power plant don't work correctly, the activity of tritium water released will overcome this limit and it become this water in no drinkable and these crops into inedibles. On top of that, the life time of tritium is more than 12 years so these places will remain contaminated during a lot of time.

3. There's exist a lot of rivers or seas, which is used for cooling systems of

the nuclear power plants, that are crossborders, that's, they are shared by several countries like our case as we will see in the seccion 1.4. The emisión of an excessive tritium activity of one country could be affect severely to the other country creating new international conflicts between them.

Because of all these reasons it is very important that we have an alarm system which is capable of measuring such low tritium activities in quasi-real time. Nevertheless, as we will see in the section 1.3, currently there are not any technic with which we can fulfill these requeriments.

All these reasons has motivated the project *Tritium*, whose objective is the development of a system for quasi-real time monitoring of low radioactive levels of tritium in water for security applications in nuclear power plants.

1.2 Tritium properties

Tritium is the only radioactive isotope of hydrogen. It was 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 [24], who checked that tritium is a radiactive element.

Tritium can be found in the environment since it is normally produced through the interaction of cosmics rays and gaseous elements of the upper atmosphere like nitrogen (^{14}N(n, ^{3}H)^{12}C) [25] and oxigen (^{16}O(n, ^{3}H)^{14}N) [26]. Then tritium becomes water (HTO) and reaches the earth's surface as rain with an estimated produccion rate of $4 \cdot 10^6$ Ci/yr
($1.48 \cdot 10^8$ GBq/yr) [16] [25] .

Tritium can be produced artificially in the environment from many different anthropogenic origins. There are a big amount of tritium which was produced on militar nuclear test explosions between 1945 and 1975, whose estimed produccion rate is $8 \cdot 10^9$ Ci ($2.96 \cdot 10^1$1 GBq) and a part of that still remain. It was mainly produced from the nuclear reactions ^{14}N(n, ^{3}H)^{12}C and ^{2}H(n, γ)^{3}H. Tritium can be also produced by commercial producers of radioluminicent and neutron generator devices ($1 \cdot 10^6$ Ci/yr), nuclear power and defense industries (less than $2 \cdot 10^6$ Ci/yr), several research facilities and nuclear reactor operation ($2 \cdot 10^6$Ci/GWyr), whose main production channels are [16] [25]:

$$^{2}_{1}\mathrm{H}(\mathrm{n},\gamma)^{3}_{1}\mathrm{H} \qquad \sigma_{th} = 5.2 \cdot 10^{-4} \text{ b} \quad [16] \tag{1.1}$$

$$^{3}_{2}\mathrm{He}(\mathrm{n},\mathrm{p})^{3}_{1}\mathrm{H} \qquad \sigma_{th} = 5330 \text{ b} \quad [16] \tag{1.2}$$

$$^{6}_{3}\mathrm{Li}(\mathrm{n},\alpha)^{3}_{1}\mathrm{H} \qquad \sigma_{th} = 940 \text{ b} \quad [16] \tag{1.3}$$

$$^{10}_{5}\mathrm{B}(\mathrm{n},2\alpha)^{3}_{1}\mathrm{H} \qquad \sigma_{th} = 3835 \text{ b} \quad [16] \tag{1.4}$$

Tritium is a radioactive element whose half-life time is $T_{1/2} = 12.32$ years. It has one proton and two neutrons and decays exclusively through β radiation, that's, it doesn't have other type of radioactive decay. In this decay, one neutron of tritium is transformed in a proton plus electron and electron antineutrino according to the following equation:

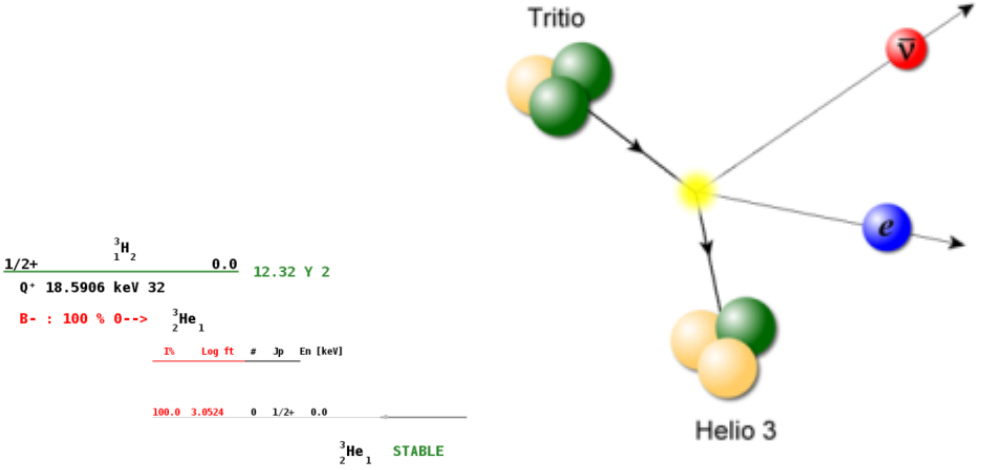
$$\mathrm{n} \longrightarrow \mathrm{p}^{+} + \mathrm{e}^{-} + \overline{\nu}_{\mathrm{e}} \tag{1.5}$$

Then the nucleus of the tritium son has two protons and one neutron so it is a helium isotope, $^{3}_{2}\mathrm{He}$ which is stable. Therefore, the nuclear reaction which descript the β^{-} decay of the tritium is:

$$^{3}_{1}\mathrm{H} \longrightarrow {}^{3}_{2}\mathrm{He} + \mathrm{e}^{-} + \overline{\nu}_{\mathrm{e}} \tag{1.6}$$

In the Figure 1.2 we can see the scheme of tritium energy levels. In this decay we practically don't have the posssibility of detecting the neutrinos because it interacts very weakly with matter and, therefore, with our detector ($\sigma \propto 10^{-42}$ cm^2 [27]) and, since ^{3}He has a much larger mass than electrons and neutrinos, by conservation of energy and momentum, the energy that is taken by its atom is very small. Therefore, we will focus on the electron detection.

The energy that is released in this nuclear reaccion is constant, 18.6 keV, but it is divided between the products of this reaccion. Therefore



(a) Tritium energy levels [28] **(b)** Graphic representation of tritium decay [29]

Figura 1.2 – Tritium decay

not all beta particle (electrons) will have the maximum energy. This is what we can see in the figure 1.3, which is the energetic spectrum of the electrons which are emitted in the tritium decay. The maximum energy of this electrons is 18.6 keV (when beta particles have all the energy), the average energy is 5.7 keV and the most likely value is slightly below of the average energy, around 4.5 keV.

Keep in mind that, although the helium isotope is stable, it will be exited immediately after this decay. As a consequence, after the tritium β^- decay, we will have a subsequent dexcitation of the ^{3}He which will produce fotons, γ, with several well-defined energies that correspond to their energy levels, X-rays. COMPROBAR... It doesn't affect directly to our detector due to the efficiency of our detector at those wavelenghts, as we will se in the chapter **??**, section **??**, but it could be affect indirectly. BUSCAR ESTAS ENERGÍAS Y VER SI SON TAN DIFERENTES COMO PARA NO AFECTAR DIRECTAMENTE O SE PARENCE A LAS DEL TRITIO Y SI AFECTAN DIRECTAMENTE.

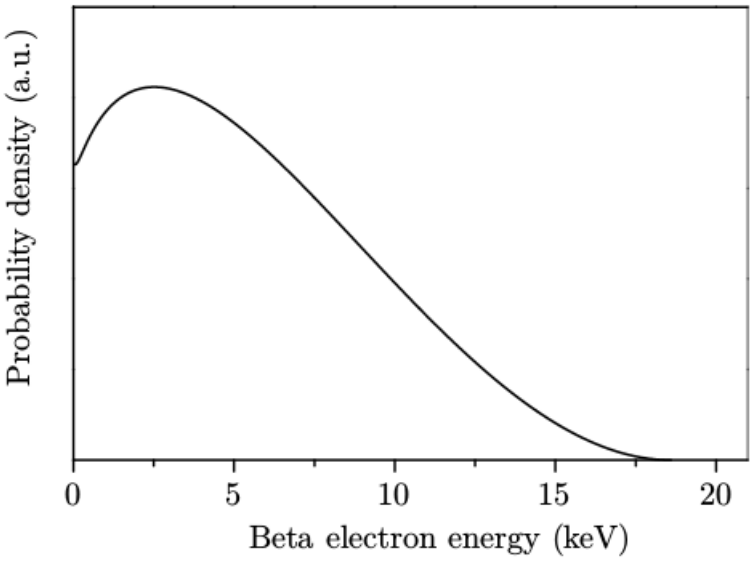


Figura 1.3 – Energy spectrum of tritium electrons [30]

The releasing energy, which is produced in the tritium decay, is very little. In fact, it is the radioactive isotope with the lowest energy released in its β disintegration [25]. As a consequence the β particles which is emitted in this tritium decay will have a very little mean free path as you can see in the table 1.5.

On the one hand, it means that the tritium electrons is easily stopped even for simply walls like our clothes, the laboratories gloves or even the our skin it-self, that's, the radioactive hazard is low. Nevertheless, the danger of tritium is increased when tritium is ingested or inhaled because if it has enough radiactivity it can affect to our internal organs because it has a high biologic life time, 9.5 days [25], time during which tritium remains in our body and we will be receiving dose due to tritium radiation. Therefore, their health hazard is high.

On the other hand, this short mean free path will be a problem when we try to detect tritium and due to that, there are some limitations

Material	Energy (β)(keV)	Penetration Depth
${}^{3}_{1}H_2$, STP	5.7	0.26 cm
${}^{3}_{1}H_2$, STP	18.6	3.2 cm
Air, STP	5.7	0.036 cm
Air, STP	18.6	0.45 cm
Water, soft tissue (solid matter whose density is 1 $g \cdot cm^{-3}$)	5.7	0.42 μm
Water, soft tissue (solid matter whose density is 1 $g \cdot cm^{-3}$)	18.6	5.2 μm

Table 1.5: Mean Free Path of tritium isotope for several energies [25]

which we will have to take into account when we design our detector.

Tritium has different physical properties than other natural isotopes of the hydrogen like different boilling points as you can see in the table 1.6 or the property of auto-radiolysis which exists because the energy released in tritium decay is larger than the energy bond of oxigen and hydrogen in water molecules (5.2 eV) or the ionization energy of water molecules (12.6 eV) so it can break these molecules [31]. Auto-radiolysis only happen when radioactive elements are presented.

Molecule	Boiling point (for gases) (K)	oxidation form
H_2	20.39	H_2O
HD	22.14	HDO
HT	22.92	HTO
D_2	23.66	D_2O
DT	24.38	DTO
T_2	25.04	T_2O

Table 1.6: Gas molecules of hydrogen isotopes and their boiling points

Although tritium has different physical properties it has almost

the same chemical behaviour than other hydrogen isotopes. Tritium, like hydrogen, is a gas at standard conditions of temperature (273 K) and preassure (1 atm) forming a two-atom molecules which can be HT, DT and T_2. It can become in tritium water through oxidation and exchange reactions as you can see in the following chemical ecuations[25]:

$$\begin{aligned} &Oxidation: \\ &2 \cdot HT + O_2 \rightarrow 2 \cdot HTO \\ &2 \cdot T_2 + O_2 \rightarrow 2 \cdot T_2O \end{aligned} \tag{1.7}$$

$$\begin{aligned} &Exchange: \\ &HT + H_2O \rightarrow H_2 + HTO \\ &T_2 + H_2O \rightarrow HT + HTO \end{aligned} \tag{1.8}$$

Due to this chemical similarity tritium water can performe the same chemical process than non-radiactive water, some times with higher rate if the tritium concentration is high enough to catalyze the reaction. Its biological hazard comes from this chemical similarity since tritium water is able to substitute normal water in human body. On top of that, tritium water has a higher absorption in human body, around 99%, than tritium gas, whose absorption in the human body is less than $5 \cdot 10^{-3}\%$ by inalation or practically negligible by skin absorption [25] so it is more dangerous.

1.3 State-of-the-art in tritium detection

Measurement of tritium activity is one of the systematic environmental controls that have been carried out for dozens of years around nuclear power plants during their energy production and around nuclear research facilities.

As a consequence, this measurement has been attempted with many different technologies so far in order to improve the state of the art of each time. The most researched techniques are summarized in the table 1.7.

	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 1.7: State-of-the-art in the tritium detection for different technics [30]

Nowadays, the most used technic for mesuring tritium in water is the LSC. It consists of mixing a liquid sample (some ml for environmental measurements or less for higher activities) with liquid scintillator. In our laboratories, LARAM, at the University of Valencia, this mixture is made in a ratio of 50:50 [32] but it will depend on each system and each sample [33] [34]. In this technic, the β energy that is emitted from the sample excites the molecular energy levels of the liquid scintillator and it is quickly desexcited emitting several photons with a well-know energy (fluorescence), normally in the visible range. Finally these photons are detected with photosensors, processed and analysed.

This technic has a very good detection capability and precision (LDL for tritium better than 1 Bq/L [35]) but it has some problems. On the one hand it need too time for taking a mesurment (more than 2 days)

and, on the other hand, although this sample could be non-radiactive, it contain tolueno which is a toxical chemical waste so we need to follow a special protocol for removing this samplex. On top of that all these technics need special staff for sampling, chain-of-custody and lab analysis which consum economical and time resources. In order to avoid the last problem a monitor of tritium with LSC has been studied [36] but the other problems still remain.

The ionization chamber (IC) is based on a gas chamber (sample) which contains electrodes connected to different voltage. This electrodes recover the ionization current that is produced due to the β radiation. It is a simple and fast system, but the problem is that on the one hand it has too high LDL, more than 10 kBq, and, on the other hand, it needs the state of the sample to be gas or steam [37] [38].

The calorimetry is based on the measurement of the heat generated due to the tritium radiation [39] [40]. The problem with this technic is that it has a too high LDL, of the order of GBq, and it needs too long time, more than 2 days, for taking a measurement.

The Beta Induced X-ray Spectrometry (BIXS) is based on the measurement of the bremsstrahlung with PMTs of NaI [41] [42] or with Silicon Drift Detector (SDD) [43] produced due to the tritium radiation. The problem with this technic is that it has too high LDL, of the order of MBq.

There are many more different methods for tritium detection, although they are less used or less experimentally developed, each one with their own problems for our objective. For example, APD [44], which we cannot use in our case because they cannot function in contact with water, the mass spectrometry [45], which needs to store the sample several months before taking the measurement or Cavity ring spectroscopy [46], which requires a special optical configuration that is not possible outside

the laboratory.

We have to keep in mind that all these techniques are offline methods that take too long to finish the process of taking measurements which include sample taking, sending the sample to the lab, analyzing of the sample so we cannot use them for tritium monitoring. LSC is the only technic which has a LDL enough low to verify the compliance with the established limit, 100 Bq/L. Therefore we will explore this area but, in order to avoid the problems related with this technic (off-line results, no-reusable liquid scintillator and the chemical toxic wastes) we will delve in solid scintillators. There are several studies that have been done so far which intend to do the same as we want with this project, to create a quasi-real time monitor of low tritium activities in water based on solid scintillation:

- First study was done by M. Muramatsu, A. Koyano and N. Tokunaga in 1967 who used a scintillator plate read out by two PMTs in coincidence [47].
- The second study was 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 [48].
- Third study was performed by R. V. Osborne in 1969 who used sixty scintillator plates stacked read out by two PMTs in coincidences [49].
- Fourth study was done by the A. N. Singh, M. Ratnakaran and K. G. Vohra in 1985, who used a scintillator sponge read out by electronic coincidence [50][51].
- Fifth study was carried out by K. J. Hofstetter and H. T. Wilson in 1991, who did different experiments for testing different shapes of scintillator plastic like several sizes of beads, fibers, etc. The better result which Hofstetter got for solid plastic scintillator was a efficiency of the order of 10^{-3} [52][53].

	Efficiency, η_{det} $(cps/(\text{kBq/L}))$	Surface F_{sci} (cm^2)	Specific efficiency $\varepsilon_{det} = \eta_{det}/F_{sci}$	LDL (kBq/L)
Muramatsu	$3.85 \cdot 10^{-4}$	123	$3.13 \cdot 10^{-6}$	370
Moghissi	$4.5 \cdot 10^{-3}$	> 424.1	$< 1.06 \cdot 10^{-5}$	37
Osborne	0.012	3000	$4 \cdot 10^{-6}$	37
Singh	0.041	3000	$1.37 \cdot 10^{-5}$	< 37
Hofstetter	$2.22 \cdot 10^{-3}$	$\sim$ 100	$< 2.22 \cdot 10^{-5}$	25

Table 1.8: Results of different scintillator detector for tritium detection [30]

The results of these experiments are sumarized in the table 1.8. We can see in the first column that the intrinsic detector efficiency, η_{det}, is very different in these experiences. As we know that, in this type of detectors, one of the most important factor, which affect to the efficiency, is the active surface of the plastic scintillator, F_{sci}, and we can see in the second column that it is very different en each detector, we use the specific detector efficiency (third column), in order to compare these experiments, that's, the efficiency normalized to this active surface. Now we can check that, effectively, these specific efficiencies are quite similar. On top of that we can check that the better specific efficiency was obtained for Moghissi who used scintillating fibers. This is a good point which justify our choice about using of fibers like a scintillator. Finally we can see in the last column that the LDL in all these experience are more or less similar and, they are too high for our aim.

To sum up with solid scintillator detectors we can practically avoid all the different problems which other techniques have. The only problem which still remain is that they have a too high LDL. Developing a detector which overcome these LDL is an essential study right now in order to monitoring the tritium levels.

1.4 Tritium project and Tritium monitor

As we have seen in the section 1.3, the current technics which exist nowadays have either higher LDL than the limit established by Council Directive, 100 Bq/L, or they are a off-line method (too slow) so those methods cannot be used for tritium monitoring in quasi-real time.

As a result of these limitations appear the *Tritium* project [54], whose title is "Design, construction and commissioning of automatic stations for quasi-real time monitoring of low radioactive levels of tritium in water".

This project has been funded by Interreg Sudoe program of the 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 sample before we introduce it in our detector, the tritium detector where the tritium measure will be done, the active veto and the pasive shielding which reduce the natural background of our tritium detector and several types of electronic which control all these parts of the monitor, analyze the tritium measurement and will send an alarm if the limit of 100 Bq/L is overcome.

The tritium detector is based on measurements of low energy beta radiation from the radioactive decay of tritium. For doing this task this detector consists of scintillator fibers, that we put directly in contact with water which can contain tritium. We need to put both, scintillator fibers and tritium water, in contact due to such a low mean free path of tritium electrons (table 1.5 of the seccion 1.2). Then, the photons produced on this fibers will be read out by several photosensors. The photosensors which we have tested in this experiment are photomultiplier tubes (PMT) and silicon photomultiplier (SiPM) arrays.

The difficulty when we try to measure tritium is to distinguish these signals from the background. This is because tritium signals are small since tritium events has low energy ($\sim$ keV) and this is the energy range in the spectrum where there are more background counts (the lower energy, the more background events). We will use coincidence techniques in order to reduce the counts from the background.

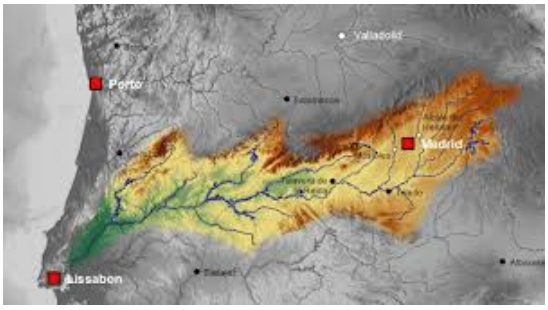
It is important to check the water tightness of each prototype because if the water reaches the photosensor it will be irreparably damaged. On top of that if we use high concentrations of tritium in water for laboratory tests we can contaminate this laboratory, which could be dangerous for the healthy of the workers and it could spoil measurements of future experiments.

Finally this monitor will be installed in the Arrocampo dam, Almaraz, Spain, where the Almaraz nuclear power plant release the water which is used in their cooling system, Figure 1.4. This NPP has two nuclear reactors whose type is PWR. This dam is located near the Tajus river, which is the largest river in Spain, 1007 km. This river cross from Aragon (Spain) to Lisbon (Portugal) and flows into the atlantic ocean. This river is used for an important quantity of animals, plants and even humans because the water of this river is used as drinking water by the spanish and portuguese people. Therefore the international cooperation in order to maintain the quality of this water is very important.

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

FOTOO TRITIUM

(a) Arrocampo dam and Almaraz Nuclear Power Plant



(b) Tajus river along Spain and Portugal

Figura 1.4 – Arrocampo dam, Almaraz NPP and tajus river

Each institution has focused in the development of a different part of all this project:

- First, the Extremadura group has developed and installed the ultra pure water system with wich we get water with very low conductivity, $\sigma \approx 10\ \mu S/\text{cm}$. The conductivity of the water before the cleaning proces is around 1000 $\mu S/\text{cm}$. This clean process is very important for two reasons. On the one hand, it is important for maintaining our detector very clean, which is a critical point. On the other hand, it's important because with this process we reduce the natural background since we remove several natural radiactive isotopes that there are in this water (except tritium) such as ^{222}Rn, ^{40}K or ^{137}Cs. This system will be explained in the chapter 5.

- Second, french group has develop the pasive shielding where our detector will work inside. It is based in ultra radiopure lead with very low intrinsic activity. The objective of this pasive shielding is to reduce the external natural background that affect to our system, for this reason we use lead. Obviously, this shielding doesn't have to affect to the measurement of our system, for this reason we use radiopure elements with very low intrinsic activity. This shielding will be explained

in the chapter 4, section **??**.

- Third, The Portugal and Spanish people has developed the simulations about this system. The program which we have used in this project is GEANT 4, which is a simulation package. It consiste in a extensive C++ library with which we can design the geometry of our detector, the physical processes which happen there, etc. This simulation will be explained in the chapter 7.

- Lastly, The Portugal and Spain people has collaborated for designing, developing and building four different prototypes of tritium detector and active vetos for removing cosmic events. These prototypes and vetos will be explained in the chapter 3.

The tritium level which we want to mesure follow the ALARA principle (As Low As Possible Achievable) and to get it there are important characteristics which our tritium detector must have:

- *Compact.* This is important because in the place where this detector will be installed the useful space that we can use is finite.

- *Thin active volume and large active area.* On the one hand, we have to take into account that, as we have seen in the table 1.5 of the seccion 1.2, the mean free path of the β particle of tritium decay is very low so we need to work with thin active volumes. In the practice, Active thickness beyond the mean free path of the tritium will only contribute to the background. On the other hand, as we have checked in the seccion 1.3 the efficiency of this type of detector scales with the active area so we need to design our detector with the largest possible active area.

- *High sensitivity to tritium.* We are going to work with low tritium activities so we need to reduce as much as possible the non-detected tritium events.

- *High specificity to tritium.* We need that our detector is able to distinguish the tritium signal of the signal of other radiactive elements which can be present in the initial sample.
- *Quasi-real time response.* As we have seen it is important that our sistem can work in quasi-real time in order to detect any problem as fast as possible.
- *Rugged system.* Finally, we have to take into account that our objective will be installing an automatical system which will work during a lot of years without specialized people so we need that our monitor are rugged.

In order to get the measurement in quasi-real time we need to work *in situ*, that's, we need that our detector is able to work in the same place that we take the sample. Whit the work *in site* we achieve:

- a faster monitor because we eliminates the process of taking the sample, the chain-of-custody until this sample arrive to this laboratory and the complexity which involve these tasks.
- a better monitor since if we can work *in site*, our measurements can be more frequent hence we will can identify cahnges in the activity earlier.
- a cheaper monitor because we have not only the material costs attached to the sample collection, chain-of-custody of this sample, shipping of this sample to the laboratory, etc. but we have also eliminated the costs attached to the specialized staff who are involving in these tasks. Our detector will only need frequent calibrations each time in order to ensure its correct operation.
- a safer monitor since the personal exposure dose is reduced and the changes in activity are detected fastly. On top of that we remove the possibles mistakes which can be done by specialized staff.

1.5 Work scheme

Chapter 2

Research & Development on detector design and components

2.1 Design principles of a tritium monitor

2.2 Scintillating fibers

2.2.1 Introduction

2.2.2 R&D on scintillating fibers

2.3 Photosensors

2.3.1 Photomultiplier Tubs (PMTs)

Introduction

2.3.2 Silicon Photomultiplier array (SiPMs array)

Introduction

R&D on the SiPM arrays

Chapter 3

Tritium Monitor prototypes

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

3.1.1 Tritium-IFIC 0

3.1.2 Tritium-IFIC 1

3.1.3 Tritium-Aveiro

3.2 Advanced prototype, Tritium-IFIC 2

3.3 Modular TRITIUM prototype for in-situ tritium monitoring

Chapter 4

Tritium Monitor Background Shields

4.1 Tritium Monitor Background

4.2 Passive shield (Lead)

4.3 Active shield (cosmic veto)

Chapter 5

Ultrapure water system

5.1 Introduction

5.2 Set up of the Ultrapure water System

5.3 Results of the Ultrapure water System

Chapter 6

TRITIUM Monitor results and Discussion

6.1 Results from Laboratory measurements

6.2 Results from measurements at Arrocampo Dam

Chapter 7

Simulations

Chapter 8

Conclusions and prospects

Appendices

Appendix A

Más cosas

Aún faltan cosas por decir.

Appendix B

Y más cosas aún

Y más cosas aún.

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