Characterization of a Large Area ZnS(Ag) Detector for Gross Alpha and Beta Activity Measurements in Tap Water Plants

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***Abstract*—In this work we present the characterization of a large area 200 mm $\times$ 200 mm EJ-444 scintillation detector to be used for monitoring gross alpha and beta activity in tap water plants. Specific tests were performed in order to determine the best setup to read-out the light from the detector side. The possibility to stack many detectors and get a compact device with total active area of the order of 1 m^2 has been explored. Alpha/beta discrimination, efficiency and homogeneity tests were carried out with alpha and beta sources. Background from ambient radioactivity was measured as well. Alpha/beta real-time monitoring in drinking water is a goal of the EU project TAWARA_RTM.**

***Index Terms*—Alpha particles, beta rays, radiation monitoring, scintillation counters.**

I. Introduction

THE current national legislations on drinking water security in European [1] and many other countries worldwide foresee periodic controls, including radioactivity tests, performed in qualified laboratories. Liquid scintillation counting (LSC) and proportional counters on dried samples are techniques widely used at present in biomedical and environmental applications [2], [3], [4] to monitor the radioactivity content in the water intended for human consumption. The typical time needed for such controls, from the sample collection to the end, is in the day timescale [5], making it difficult to monitor the contaminant levels on a continuous basis, thus making this method rather inefficient especially in case of an alert for unexpected radioactive/nuclear events. The continuity in the monitoring is a crucial issue in case of unexpected and/or fast water contaminations such as the nuclear disasters caused by natural events (earthquakes, tsunami, …), human errors in the operations of nuclear power plants or voluntary actions (terrorist attacks).

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The main goal of the TAWARA_RTM project [6] is to create a platform for drinking water security against deliberate or accidental Radiological or Nuclear threats.

The TAWARA_RTM platform will provide a real time measurement of the gross alpha and beta activity in the water to verify whether the water is well within the limits set by the European legislation or is reaching the threshold that requires blocking the distribution to the public. In the latter case, a message is sent to the water plant management. At the same time, a second part of the system is activated, to determine the nature of the contamination by gamma-ray spectroscopy. Identifying the nature of the contamination will determine the counter-measures.

The TAWARA_RTM project will make use of state-of-art detectors, read-out electronics and computing tools that will provide an automated management of the system. It will include the development of a complete platform including the fast Real-Time Monitor system (RTM) for the measurement of the gross alpha and beta radioactivity, a Spectroscopic system (SPEC) for the nuclide identification in case of alarm, as well as an Information and Communication System that will be designed to include in the future also chemical and biological sensors. In accordance with the international recommendations in terms of screening levels for alpha and beta radioactivity at the time of the proposal, the TAWARA_RTM platform was conceived to detect a gross alpha/beta activity of the order of 1 Bq/l in a few tens of minutes.

The prototype will be installed at the North Waterworks Plant of Warsaw managed by the Warsaw Waterworks Company for the demonstration campaign. This site has been selected due to the proximity to a Polish National Nuclear Waste storage site. Compared with other EU financed projects on the same topic (see for example SAFEWATER [7] and ISIS [8]), TAWARA_RTM is more focused on improving the screening devices and procedures for the radioactivity in water. In particular, the real-time simultaneous screening of both gross alpha and beta contaminants is a significant step forward with respect to the current security protocols of the European water utilities.

II. Experimental Setup and Read-Out Tests

The basic detection module for the Real Time Monitor (RTM) of the TAWARA_RTM platform consists of a large area 200 mm $\times$ 200 mm EJ-444 scintillation detector [9] manufactured by Eljen Technology.

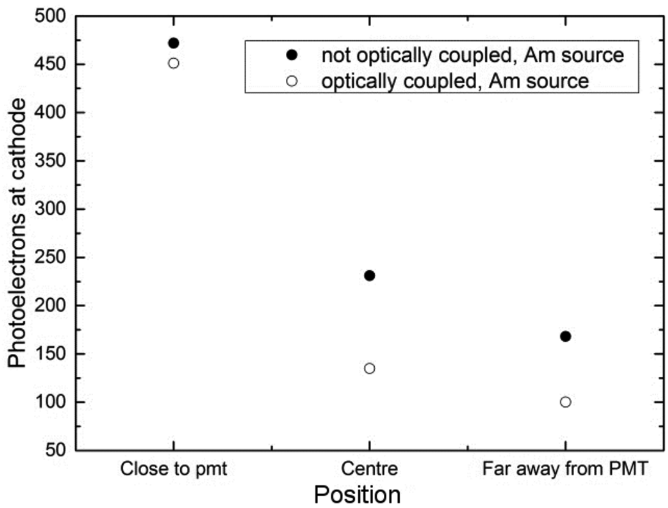

Fig. 1. Light output of alpha particles with different optical couplings between detector and WLS light guide.

Fig. 2. Assembly of the RTM basic module used in the tests.

This detector is made of a thin layer of ZnS(Ag) (about 20 μm, 3.25 g/cm^3 density) deposited on a plastic scintillator layer (EJ-212) with thickness 0.25-0.50 mm. Alpha particles are detected in the ZnS(Ag) layer whereas beta particles are detected inside the plastic layer. The different decay time of the two scintillators (200 ns for the ZnS(Ag) phosphors and 2.4 ns for the plastic scintillator) allows us to separate the two types of particles by pulse shape discrimination (PSD) technique.

Due to the short path length in water of alpha and beta particles of interest, an active area of the order of 1 m^2 is needed to reach the sensitivity required by the EU legislation for the radioactivity level in drinking water. Such a wide area can be obtained by stacking a number of 200 mm $\times$ 200 mm modules, providing a compact geometry suitable for practical applications. This solution implies reading the scintillation light from the sides of the detector module.

The first phase of the R&D of the RTM system was devoted to define the best read-out configuration that may ensure an efficient reading of the light intensity while preserving also the time information needed for the PSD.

Several tests were performed at the SCIONIX labs using standard electronics (analog chain with preamplifier, amplifier and MCA). The light produced by alpha particles (^{241}Am source, $E_\alpha = 5.49$ MeV) and beta particles (^{36}Cl source, $Q_\beta = 709$ keV) were collected from a corner of a light guide supporting two EJ-444 detectors on the opposite sides. It was found that a Wave Length Shifter (WLS) light guide is needed to transport enough light to the corners in order to obtain signals with good signal-to-noise ratio. Using standard light guides only particles interacting in the very proximity of the PMT can be detected. The WLS optical guide consists of a plate of EJ-280. Tests performed by varying thickness and doping showed that a plate of 10 mm thickness with standard doping provides the best results.

The best optical coupling between WLS and detector was studied by measuring the alpha and beta signals at different positions. As an example we show in Fig. 1 the light output of an alpha particle with the source placed at three different positions (close to the PMT, in the center of the detector and in the far corner with respect to the PMT) with/without a standard optical coupling. The latter configuration (no coupling = air coupling) shows the best read-out, especially when the source is far from the PMT. This effect is supposed to be correlated to the large number of total reflections experienced by the light to reach the PMT: an apparently less efficient optical coupling can trap more light inside the WLS. Therefore it appears that the initial deficit in the collection is well compensated by the lower loss during the transport to the PMT.

Further tests were performed at the University of Padova by using digital electronics (fast digitizers with PSD capability). A light-tight test chamber equipped with a 2D positioning system for the radioactive source was built in Padova. The final configuration used in the tests was made of two EJ-444 foils 0.25 mm thick coupled to a 10 mm thick EJ-280 WLS light guide. The light was read out at the two corners with two low-noise Photo Multiplier Tubes Hamamatsu R3550A [10] operated in coincidence to reduce random electronic background (see Fig. 2). Signals were acquired by using a 4 channel Fast Digitizer CAEN DT5730 [11], 14 bit, 500 MHz sampling rate, 250 MHz bandwidth.

Read-out and efficiency tests were performed in air by using bare EJ-444 detectors. The final detectors for the RTM system will be surface protected with a thin layer of plastic material that will increase also the hydrophobicity of the surface and guarantee long-term mechanical stability. The radioactive sources were placed in a source holder that could be moved over the surface to verify the dependence of the light output on the position, as reported in Section IV.

III. Pulse Shape Analysis

Alpha/beta discrimination was obtained using the PSD technique: typical alpha and beta signals recorded by the digitizer are shown in Fig. 3. The different decay times of the two scintillation layers determine a very different time evolution of the signals due to the two kinds of particles. This allows us to apply efficiently PSD algorithms to discriminate the type of detected

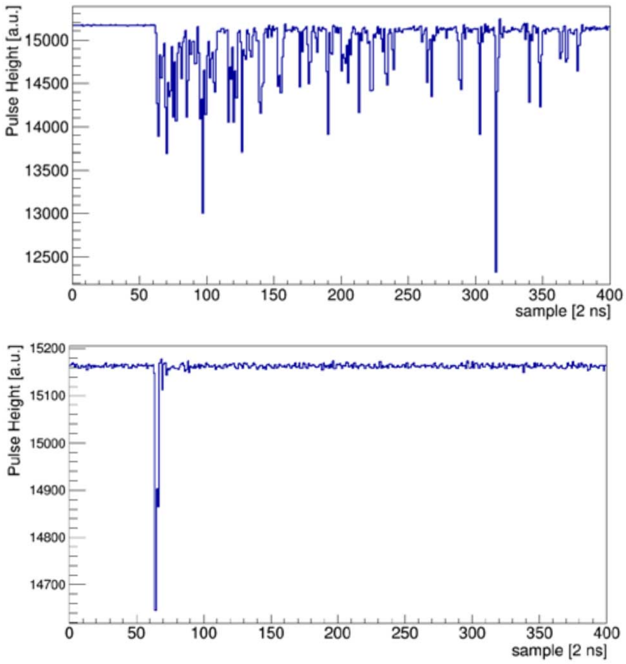

Fig. 3. Typical alpha signal (upper panel) and beta signal (lower panel) recorded with the described experimental setup.

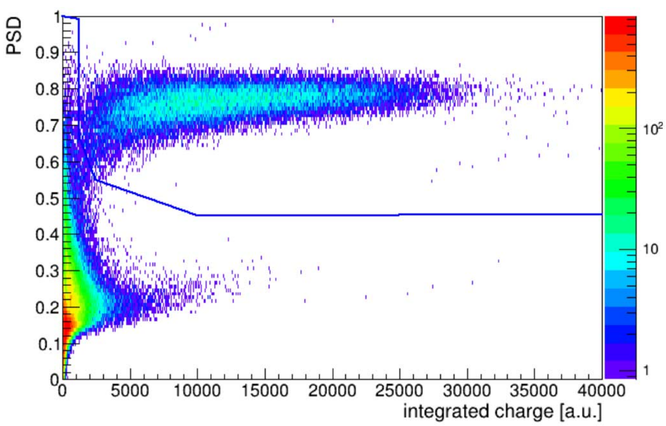

Fig. 4. Example of discrimination plot: alpha particles are separated from the beta particles using a 2D poly-line gate.

particle. It is worth noting that, due to the long decay time of the ZnS(Ag) layer and the low trigger threshold used, a significantly large hold-off time, larger than 50 μs, has to be applied in order to avoid multiple triggers on the same signal.

The main algorithm used for alpha/beta discrimination was the usual PSD parameter defined as the ratio between the integrated charge on the tail and the full signal (see for example [12] for more details on the PSD definition). Fig. 4 shows a typical PSD plot (PSD vs. total integrated charge) for a ^{210}Po alpha source ($E_\alpha = 5.30$ MeV) plus environmental radiation (beta) background.

The separation between the alpha and beta populations was obtained by means of a 2D poly-line gate. It is well known that this discrimination technique works well at mid-high energy, while at low energy the two populations tend to overlap [13]. For this reason the position of the separation line was studied

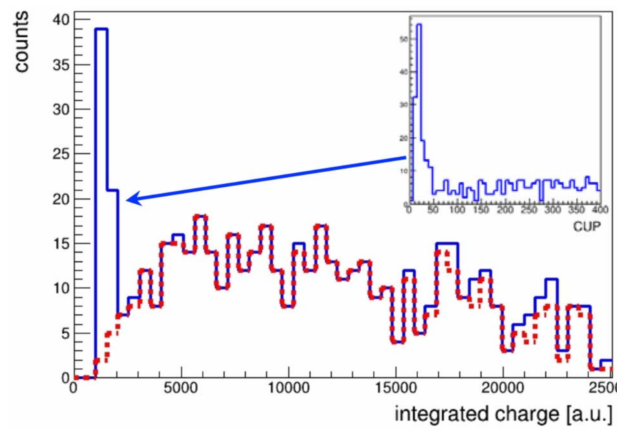

Fig. 5. Integrated charge spectra for events in the alpha region of the PSD plot when the beta source is measured. Solid blue lines refer to selection using only PSD parameter, dotted red line includes also the selection on CUP parameter. See text for details.

by measuring the alpha particles of the Polonium source with an energy degrader as described in the next Section.

After defining the best position of the separation line (blue line in Fig. 4) the contamination of beta signals inside the alpha region was evaluated by using a ^{90}Sr/^{90}Y beta source ($Q_\beta = 546/2280$ keV). The blue spectrum in the main panel of Fig. 5 represents the integrated charge distribution of events identified as alpha particles by the PSD algorithm. On top of the alpha background (see Section V for the background analysis) there is a peak structure at low energy associated to the tail of the beta distribution. This unwanted contamination could be easily removed by using a tighter 2D separation line. Nevertheless this option would increase the energy threshold for the identified alpha particles. For this reason a second algorithm was applied to improve the discrimination at low energies.

This new algorithm calculates a second parameter for each waveform (called CUP in the following) defined as the number of sampled values for which the distance from the baseline is higher than a given threshold value. As shown in the inset of Fig. 5, the low energy peak structure corresponds to events with CUP lower than 50 and can be removed by means of a CUP threshold. A more detailed analysis of the pulse shapes showed that a 2D poly-line in the CUP vs. integrated charge plane can reject also a number of pile-up signals at higher energies.

The red spectrum in Fig. 5 shows the result of the discrimination procedure, where the contamination coming from the beta population has been minimized while keeping the alpha energy threshold as low as possible.

The loss of events due to the alpha discrimination has been evaluated by comparing the net spectra of the alpha source obtained by background subtraction (an example is shown in Fig. 6, solid blue line) with the one provided by the discrimination algorithms (the dashed red line in Fig. 6). It can be seen that the discrimination algorithms affect significantly the alpha detection only at very low integrated charge values. In the case reported in Fig. 6 the alpha source was placed at 15 mm from the detector surface, thus providing a wide distribution of incident energies ranging from zero to 4 MeV and centered at

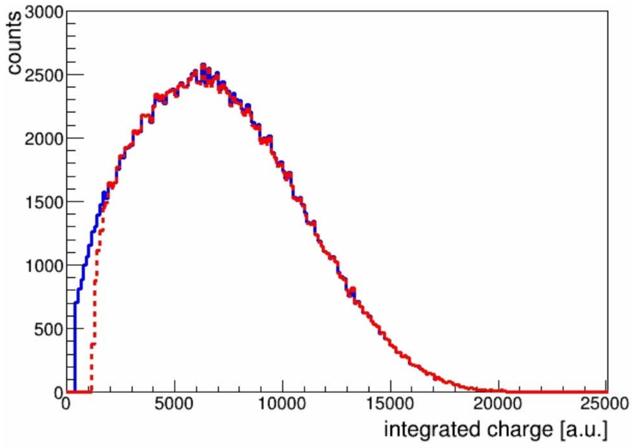

Fig. 6. Example of alpha source background subtracted spectrum (solid blue line) compared with the corresponding spectrum obtained by applying the discrimination algorithms (dashed red line).

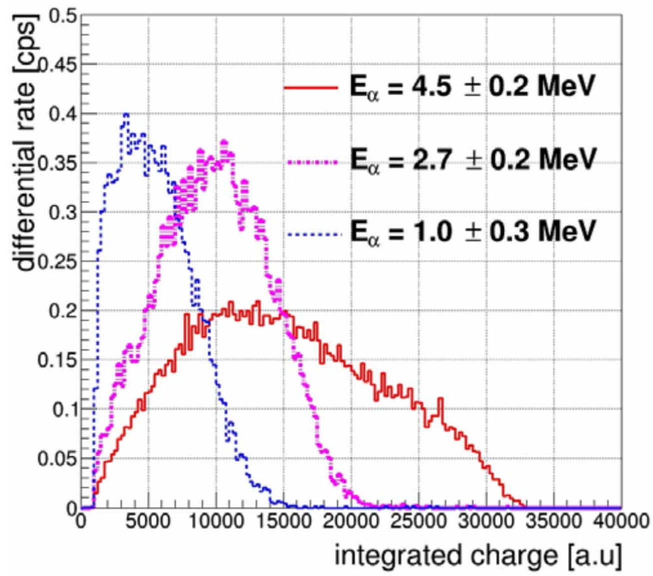

Fig. 7. Examples of alpha particle total integrated charge spectra.

about 2 MeV; the loss of events in this configuration accounts for less than 4% of the data.

The signals acquired by the digitizers were analyzed both off-line and on-line inside the embedded FPGA. It is foreseen that the discrimination parameter will be optimized with an off-line analysis of final prototype performance, then the on-line analysis will be used during the normal operation of the prototype.

IV. Efficiency and Homogeneity Tests

The efficiency and homogeneity of the basic RTM module were tested using a ^{210}Po alpha source and a ^{90}Sr/^{90}Y beta source. The sources were collimated and placed in air at different distances from the detector surface. Thin layers of different materials (aluminum, Mylar) were also used to degrade the energy of the alpha particles to measure the efficiency at lower energy.

A. Alpha Particles

The efficiency for alpha particles was measured by using the ^{210}Po source placed at different distances and positions. Fig. 7 shows an example of three alpha spectra (integrated charge) collected with the source at different distances and using different degraders. The quoted alpha energy was measured by means of a standard planar Si detector in the same geometrical configuration; the uncertainty represents the standard deviation of the alpha distribution measured in the silicon detector. The number of alpha particles measured in the Si detector was used to calculate the efficiency of the EJ-444 detector. An efficiency higher than 90% for alpha particles above 2 MeV was measured over the whole surface of the detector. An efficiency of about 50% was measured for alpha particles with energies down to about 0.5-1.0 MeV. We verified, using a simplified Monte Carlo calculation, that such efficiency values are compatible with the project objectives.

B. Beta Particles

Beta particles leave a small amount of their energy in the plastic active layer. Consequently it is mandatory to minimize the electronic noise in order to keep the threshold as low as possible and be able to collect the smallest beta signals. The electronic noise was almost completely rejected by reading the light with the two PMTs in coincidence. The efficiency was determined by comparing the net beta counting with the reference value measured with a standard planar Si detector, as in the case of the alpha source.

Finally, an efficiency larger than 80% over the whole surface of the detector was measured using the standard thickness (0.25 mm of plastic scintillator).

Further tests using a thicker 0.5 mm plastic layer showed the possibility to increase the efficiency around 90% over the whole active surface, keeping the environmental radiation background at a manageable level.

V. Background

The background due to environmental radiation was measured in air both for alpha and beta signals. The main background is due to beta particles produced by the ambient gamma radiation converting in the active volume of the detector or in the proximity of the surface. A contribution due to cosmic-ray interactions in the detectors is also expected.

A beta background of about 30 cps (counts per second) was measured for the unshielded RTM basic module. This background can be reduced by a factor 4 by means of a passive lead shielding of a few centimeter thickness.

The component of the beta background due to interactions inside the WLS light guide was also measured with the lead-shielded setup by removing the EJ-444 foils from the test module. Fig.8 shows the total background (solid green line) compared to the WLS component (dashed blue line): it can be seen that the lowest part of the total spectrum is almost completely due to gamma interactions inside the WLS light guide.

A further reduction of the background in the beta spectrum can be obtained by reading in coincidence two or more RTM modules in stack, as in the final designed geometry of the RTM system. Coincidence signals, most likely generated by cosmic-

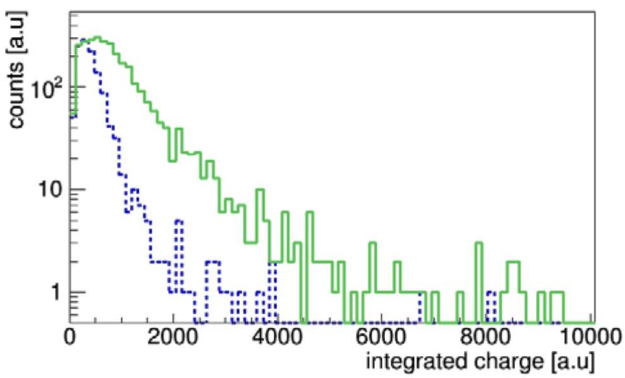

Fig. 8. Environmental radiation background measured with the lead-shielded setup (solid green line). The dashed blue line shows the component due to the WLS light guide.

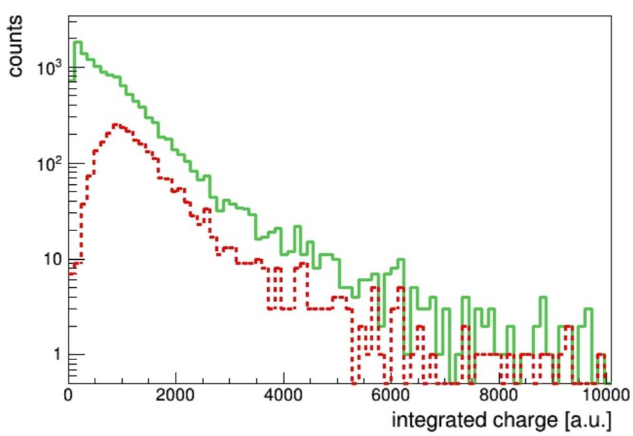

Fig. 9. Environmental radiation background (solid green line) compared with coincidence spectrum (cosmic-rays, dashed red line) obtained by using 2 vertically stacked modules with 4 PMTs in coincidence.

rays passing through the device, can be rejected by anti-coincidence selection in the FPGA board. The measured rate of coincidence events using 2 modules was 4 cps. In Fig. 9 the total environmental radiation background spectrum (solid green line) and the coincidence component (dashed red line) are shown.

The final expected beta background rate for the lead-shielded RTM module is about 5-6 Hz.

A slightly higher background in water can be expected due to the effect of the water as converting material (during the background measurements the two EJ-444 foils were covered with paper). Preliminary tests in water showed indeed an increase of the background of about 10%. An alpha background (i.e. signals tagged as alpha particles by the identification algorithms) of about 0.006 cps per module was also measured. The analysis of this background showed a main component associated to beta interactions in the ZnS(Ag) layer. This component was detected as an increase of the measured rate in the alpha region when the beta source is used. The energy spectrum of these alpha-like signals extends over the whole alpha energy range and the estimated contamination is about 2×10^{-4} counts per incident beta particle. From the point of view of the TAWARA_RTM project, the measured alpha and beta background rates are compatible with the expectations for the final performance of the RTM system.

VI. Conclusions

The basic detection module for the Real Time Monitor (RTM) of the TAWARA_RTM platform based on a large area 200 mm $\times$ 200 mm EJ-444 scintillation detector was realized and tested. Specific tests were carried out in order to determine the best light read-out configuration from the corners of the module. The final setup is made of two EJ-444 scintillator foils coupled to the two opposite faces of a supporting WLS light guide EJ-280 10 mm thick. The light read-out is provided by two low noise Hamamatsu R3550A photomultiplier tubes feeding two Fast Digitizers. Alpha particles are detected in the ZnS(Ag) layer whereas beta particles are detected inside the plastic layer. Experimental tests have been performed to study the alpha and beta particle discrimination, efficiency and homogeneity, the energy thresholds and the background. Results show the possibility of detecting alpha and beta particles with efficiencies of about 90% over the whole detector surface for beta radiation of a ${}^{90}\mathrm{Sr}/{}^{90}\mathrm{Y}$ source and alpha particles with energy larger than 2 MeV. An alpha background rate of 0.006 cps per module and a beta background of 5 cps for a lead-shielded module were also measured. The expected performance in measuring gross alpha and beta activity in water, after the experimental results of the single detector module, is compatible with the TAWARA_RTM objectives.

References

[1] Directive 2013/51/EURATOM, Official J. Eur. Union, 7.11.2013, L296/12.
[2] See for example Packard Application note ABA-005 [Online]. Available: http://www.perkinelmer.com/cmsresources/images/44-73006app_abahtsscreeningalphabetaradion.pdf
[3] ISO 9697, 2008ISO 9697 Water Quality: Measurement of Gross Beta Activity in Non-Saline Water International Organization for Standardization, Geneva, Switzerland, 2008.
[4] ISO 11704, 2010 ISO 11704 Water Quality: measurement of Gross Alpha and Beta Activity Concentration in Non-saline Water—Liquid Scintillation Counting Method International Organization for Standardization, Geneva, Switzerland, 2010.
[5] R. Rusconi *et al.,* in *Proc. Int. Conf. Isotopic and Nuclear Techniques for Health and Environment*, Vienna, 2003 [Online]. Available: http://www.iaea.org/inis/collection/NCLCollectionStore/_Public/34/083/34083020.pdf, Book of Abstract, pg. .21
[6] TAWARA_RTM project–FP7-SEC-2012-1–GA n. 312713 [Online]. Available: www.tawara-rtm.eu
[7] SAFEWATER project–FP7-SEC-2012-1–GA n. 312764 [Online]. Available: https://www.safewater-project.eu
[8] ISIS project–FP7-SEC-201201–GA n. 312330 [Online]. Available: http://www.isis-project.eu
[9] EJ-444 detector [Online]. Available: http://www.eljentechnology.com/index.php/products/zinc-sulfide-shifting-plastics/99-ej-444
[10] R3550 PMT [Online]. Available: http://www.hamamatsu.com/eu/en/product/category/3100/3001/R3550A/index.html
[11] CAEN DT 5730 [Online]. Available: http://www.caen.it/csite/CaenProd.jsp?parent=14&idmod=757
[12] D. Cester *et al.*, "Pulse shape discrimination with fast digitizers," *Nucl. Inst. and Meth. A*, vol. 748, pp. 33–39, 2014.
[13] S. D. Amber *et al.*, "A hybrid pulse shape discrimination technique with enhanced performance at neutron energies below 500 keV," *Nucl. Inst. and Meth. A*, vol. 638, pp. 116–121, 2011, and references therein.