Phoswich scintillator assemblies: Application to the Simultaneous detection of Particle and Gamma radiation

ENSAR 2
Work Package 9 – JRA1 PASPAG
Report period: 1 March 2019 – 31 August 2021

Proposal Acronym: ***PASPAG***

Activity number: ***WP9*** *-* ***JRA1***

Descriptive title of the activity: ***Phoswich scintillator assemblies: Application to the Simultaneous detection of Particle and Gamma radiation***

Webpage: *http://www.iem.cfmac.csic.es/departamentos/nuclear/WEB-PASPAG/webstyle1/*

OBJECTIVES

The PASPAG JRA has worked on R&D in order to optimize gamma-ray spectrometry combining a maximum of solid angle with good rate capability and energy resolution. Combinations of novel scintillator materials and photon sensor technologies in combination with high granularity have been the key tools to tackle the challenges associated with extreme Doppler shift at relativistic energies, high fluences and time and energy resolutions. The obtained knowhow has in several cases been applied to novel societal applications

DESCRIPTION OF WORK

PASPAG has exploited novel scintillator materials and light-sensors to obtain new techniques and concepts for gamma-ray detection. We have focused on R&D in order to simultaneously detect gamma rays, neutrons and charged particles at high fluencies.
We have also taken this technology out of basic science to exploit societal applications within, the areas of nuclear medicine and homeland security. Depending on the applications, features like energy resolution, position sensitivity, high-rate capability, and insensitivity to magnetic fields or radiation hardness have been tackled. During this 3rd period we have mainly focused on prototyping and testing of devices, meaning that not all Tasks and groups have been active during the prolongation period.

PROBLEMS ENCOUNTERED DURING THIS PERIOD

In principle all Milestones and Deliverables were delivered during the previous report-period, however, the final test results were still to be delivered. Delays due to negotiations with the suppliers of photo-sensors and delays in the delivery of new scintillators were already indicated in the 2nd period report. Due to the first prolongation of the ENSAR2 project, the preparations for final prototyping could continue over 2019 preparing for the final test-beams.
However, the Covid-19 pandemic closed down all R&D and experimental activity for almost a year (2020), especially all possibilities for travel and perform any experiments were impossible. Luckily, we have been able to profit from the 2nd prolongation of the ENSAR2 to perform these final experiments, and to hand in the final Deliverable D9.6 according with updated time-schedule.
The activity of the WP9 during this 3rd period of ENSAR2, between March the 1st 2019 and February the 31st 2021, very much coincide with deliverable D9.6.

Phoswich scintillator assemblies: Application to the Simultaneous detection of Particle and Gamma radiation

TASK1 Novel Scintillator Materials: (INFN – CNRS)

During this period at INFN-Milano in collaboration with CNRS the activity was concentrated on the imaging properties of large volume (3”x3”) scintillation detectors and on the readout of these scintillators using SIPM. In 2019 a 1-year fellowship provided by ENSAR2 was assigned at INFN to perform the experimental work.

A LaBr3(Ce;Sr) 3x3” crystal coupled to a matrix of 144 SIPM has been studied and compared to the same crystal coupled with a standard 3” PhotoTube. The SiPM-array is composed form of 6 x 6 mm^2 individual SiPM that together covers the full crystal. (Fig. 1)

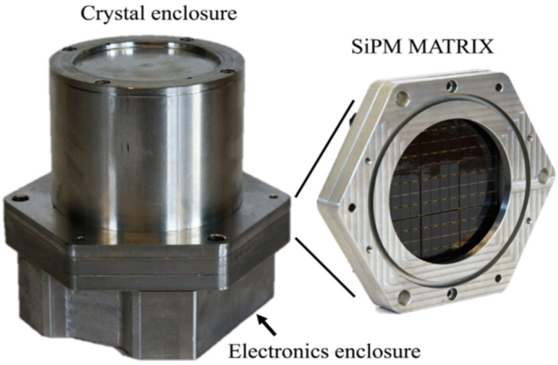

Figure 1: Photo of the $LaBr_3$(Ce;Sr) 3x3” crystal encapsulated and coupled to an array of 144 6x6mm^2 SiPMs.

An interesting detail was noticed, if one study the internal ^{227}Ac alpha-contamination, see Fig. 2, there is a shift in the alpha peak positions obtained when using the SiPM compared to the standard PM, a shift in the order 50 keV is observed. The crystal is the same so the reason must come from the sensors and have to be investigated further.

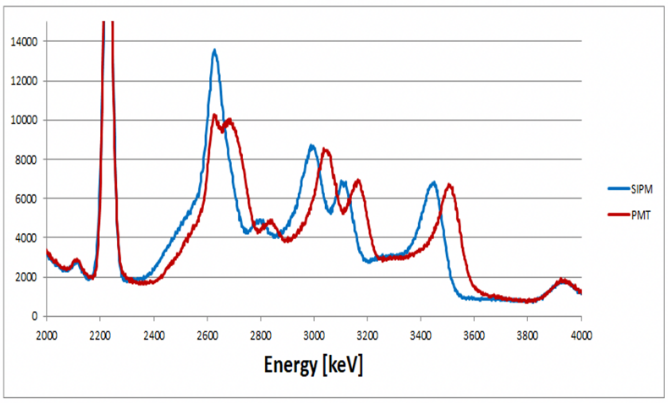

Figure 2: Spectrum obtain from the internal ^{227}Ac alpha contamination that is always present in $LaBr_3$ and $LaCl_3$ crystals. A shift in the order of 50 keV is observed, note that the gamma-ray peak though coincide perfectly.

Two types of studies were performed in order to see if it is possible to determining the position of the gamma-ray impact when using the SiPM array.

- Using a collimated ^{137}Cs source, placed so that the gamma ray-beam enter on the crystal in a definite x-y or z-y -position, see Fig. 3.
- Using a non-collimated ^{137}Cs source i.e. the full crystal is irradiated on its axis from a distance of approximately 20 cm. We call this measurement 'FLOOD'.

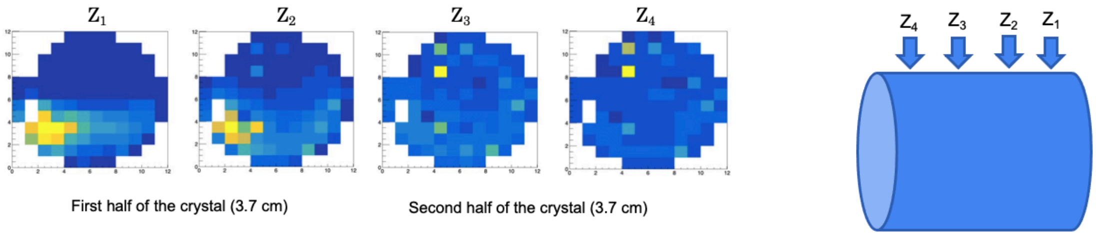

Figure 3: The figure shows the hit-patterns obtained when irradiating the crystal with gamma-rays from a collimated ^{137}Cs source at different positions along the Z-axis, the SiPM is mounted at Z_0.

In the spectra associated to the collimated source we notice that the spectrum measured by a single SIPM present a gaussian curve if it is far away from the collimation axis, while a tail appears to the right-side if it is near the collimation axis. See deliverable D9.6 for more details.

TASK2 Phoswich detectors: (CSIC – USC - CTH)

During this period a lot of efforts has been placed at CSIC and USC and at the associate CTH to mitigate the problem observed in the big LaBr/LaCl phoswich CEPA-clusters for R3B delivered by Saint Gobain. In several of the milestones and deliverables it has been reported on the high dynamic range to be covered but also on the un-linearity found both in efficiency and energy resolution in these crystals. These crystal materials are so hygroscopic that the standard method of lapping is not possible. Two ways are being followed to find a solution; the first one is to mitigate the problem by adding a lightguide to the crystals, Saint Gobain is preparing this solution and we can expect to get refurbished crystals back late 2021. Monte Carlo simulations indicates that this should be a possible solution with a minor loss in efficiency. The second solution is to go for another type of phoswich CsI-GYGAGG suggested and investigated by the USC group. Simulations has been performed and initial laboratory test with sample crystals have been performed with good results. This solution was presented and accepted by the R3B collaboration in order to have a detector for Phase1 of FAIR.

TASK3 Hybrid arrays and their applications: (CSIC – INFN – IFJ PAN – IFIN HH)

At IFIN-HH one has been working on a method to determine the enrichment level of nuclear fuel. The same type of crystal (LaBr3(Ce;Sr) coupled with similar array of 56 SiPM as mentioned in TASK 1 was used as a prototype to be able to estimate the enrichment of ^{235}U in a bulk ^{238}U sample. This is of course a very important application in order to be able to control and oversee the strategic use of nuclear reactors in different countries. The results obtained, shown in Fig. 4, illustrate that the prototype is sensible to the ^{235}U-enrichment of the sample especially that the 63.3 keV of ^{238}U and the 185 keV of ^{235}U lines are clearly identified and can be used to estimate the enrichment level. In field operation it is of course much easier to handle a scintillator than a bulky LN_2 cooled HPGe detector.

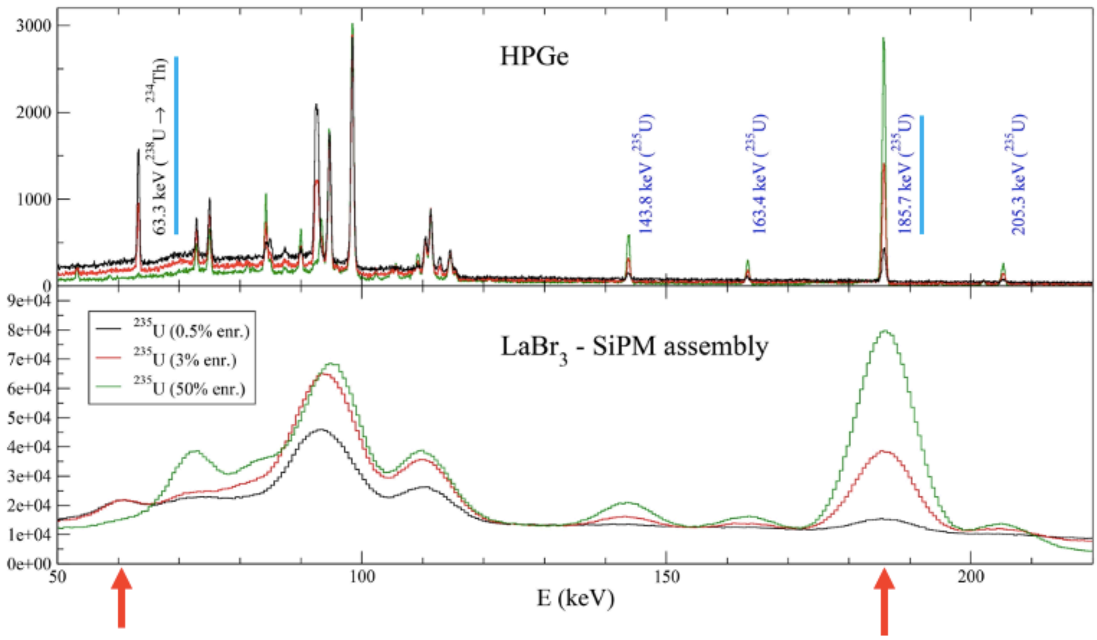

Figure 4: Spectra showing that the scintillator is sensible to the ^{235}U-enrichment of the sample and that the 63.3 keV of ^{238}U and the 185 keV of ^{235}U lines are clearly identified and can be used to estimate the enrichment level.

At IEM – CSIC one has been working on a p-CT imaging prototype, treatment plans made via proton-CT images will offer more accurate estimations and better control of the treatment than with the present X-ray imaging. To this aim, R&D to perform medical imaging with proton beams via p-CT using particle detectors extensively used in Experimental Nuclear Physics, the Double-Sided-Silicon-Strip-Detectors as tracking devices in combination with a big LaBr scintillator capable to fully stop the high energy protons and determining the energy. As proof-of-concept test experiments were performed in 2019 at the local Madrid 5MV tandem accelerator. Of course, at this proton energy (10 MeV) only tracking through very thin phantom could be made, however, the reconstruction algorithm could be evaluated. Three Master students performed and defended their final thesis on this subject in the summer of 2019.

Phoswich scintillator assemblies: Application to the Simultaneous detection of Particle and Gamma radiation

In 2020 due to the pandemic no presential work could be done, we however, could refine the simulations and designed a pCT prototype for bigger phantoms (6 cm diameter) optimized for the proton energies (110 MeV) available at Centrum Cyklotronowe Bronowice (Kraków, Poland) the home lab of our collaborator IFJ PAN. Also, here we have had help o 3 master students (to be defended September 2021). The experiment could finally be performed in June 2021. The equipment was shipped from Madrid to Krakow and 3 researchers followed to mount and perform the experiment with participation and help from our collaborators of IFJ PAN and financial support from ENSAR2 access. Figure 5 show a sketch of the experimental set-up as it was represented in GEANT4.

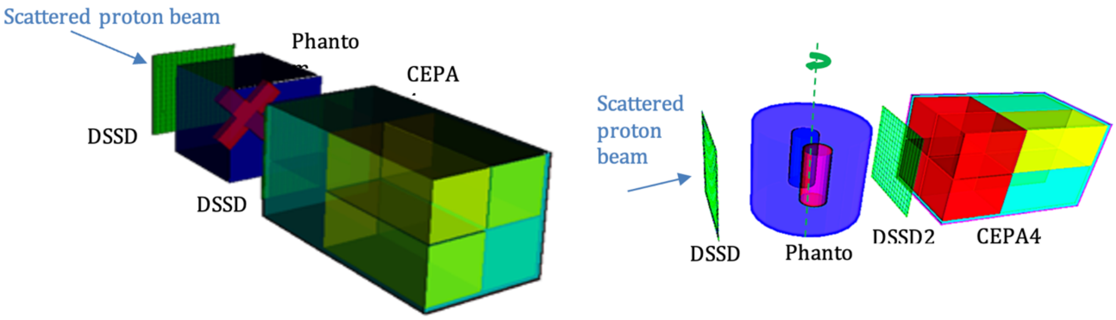

Figure 5: Geometry of the experimental setup as included in Geant4 simulations. Two different phantoms are shown. The cross shaped (red) aluminum inserts on a PMMA cubic matrix shown on the left used to take a simple projection to evaluate the image. The PMMA phantom with inserts of Alcohol and Water shown in the right sketch was evaluated at several rotation angles in order for a 3D- reconstruction.

The analysis shows very promising results (Fig 6) and we are preparing for new experiments with even thicker phantoms and realistic treatment energies up to 230 MeV.

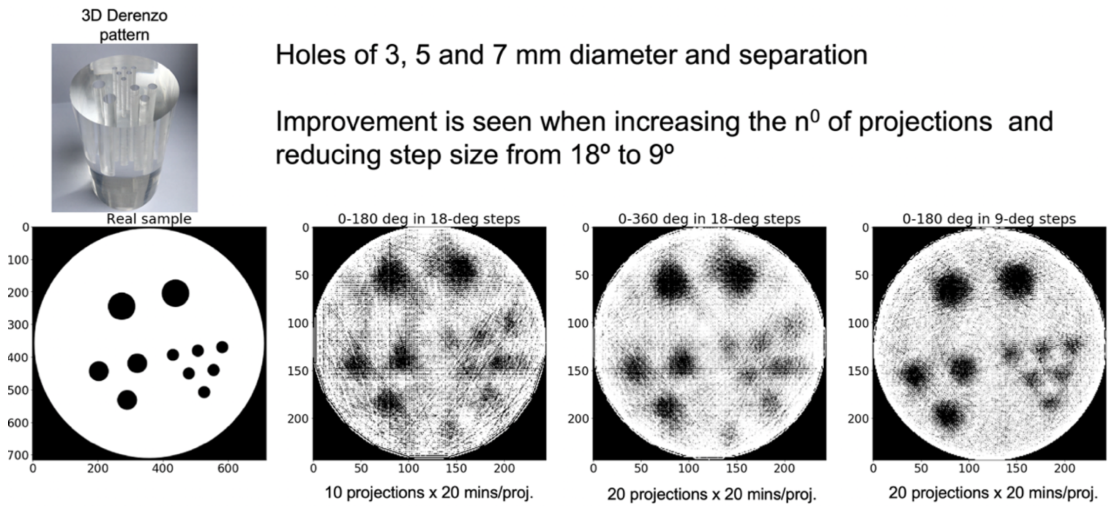

Figure 6: On top is shown the 3D-derenzo phantom (PMMA) used in the experiment. To the left is shown the real sample, followed by the reconstruction result from the different measurements made.

Subtask 3.3 Secondary Electron Emission: (GSI – U Cologne – U Rzeszow)

The SEE-collaboration has made improvements on their device to be able to measure the fluence in the experimental beamlines of our facilities. The secondary electrons stimulated near the material surface have a better chance to escape the material, therefore nanomaterials may improve the SEE yield through the manipulation of material geometry. Further modification of the surface coating can be used to decrease the energy necessary for the electrons to leave the materials surface, enhancing the SEE yield (SEY).

The purpose of this R&D was to find a nanomaterial that has a higher SEE than gold. A measurement of the SEE properties from 1D (one-dimensional) nanostructures of ZnO and ZnO/GaN (ZnO with GaN coating) composed of a mostly regular pattern of nanotubes grown on a thin Si3N4 substrate has been performed. A 4.77 MeV/u Au beam. From the GSI UNILAC was directed to the microbeam experiment setup.

It was observed an average increase of 2.5 secondary electrons emitted from the 1D ZnO nanotubes compared to gold. The data also shows that GaN coating does not lead to a higher SEY yield. One possible direction in future investigations is: (1) search for coating leading to larger SEY (2) investigating materials with smaller nano-tube diameter and larger density. When considering thin detectors, we suggest the use of a nanomaterial made by growing high aspect ridges on a thin substrate, with ridges regularly spaced and perpendicular to the substrate. If this material is placed at an angle relative to the beam the ions will pass through multiple ridges. Utilizing this approach, the SEY is increased due to passing through multiple surfaces. It is worth noting that presenting further surfaces due to the to the higher energy δ-electrons bumped by a neighbouring nanostructure, the SEY will be further increased via secondary interactions with the material.

It has been performed a comparison of SEE properties between novel nano-materials with one and three-dimensional nano-structures composed of a mostly regular pattern of rods. The nano-structured materials investigated show enhanced SEE properties when compared with gold (standard material used in SEE). Results from this work will enable the development of new radiation detectors for science and industry. During 2020-21 analyses of data and preparation of publication have been performed. Full details of these investigations are published in OPEN ACCESS in the NATURE Scientific Reports.

General considerations during 3rd period:

Risks:
As we are at the end of the project eventual risks during the time of project have all been mitigated; the objectives are fulfilled and all milestones and deliverables have been handed in on time.

Publications:

- O. Tengblad, M. Garcia Castaño, I. Marroquin
  Phoswich scintillator assemblies: Application to the Simultaneous detection of Particle and Gamma radiation – PASPAG
  Nucl. Instr. Mehtods B 463, 15 January 2020, Pages 415-417
  https://reader.elsevier.com/reader/sd/pii/S0168583X19302228?token=B5B7445E9318535980A19258DC919419E14A0B52E90E6D5EBBD542CB9717C43DE4F9EC51BA3CDB12466441E90F1BA0F6&originRegion=eu-west-1&originCreation=20210826144909

- Boutachkov, P., Voss, K.O., Lee, K. et al. An investigation of secondary electron emission from ZnO based nanomaterials for future applications in radiation detectors. Sci Rep 11, 737 (2021). https://doi.org/10.1038/s41598-020-80788-y
  https://www.nature.com/articles/s41598-020-80788-y

- Cholewa, M., Cappellazzo, M., Ley, M. et al. In search of nano-materials with enhanced secondary electron emission for radiation detectors. Sci Rep 11, 10517 (2021). https://doi.org/10.1038/s41598-021-89990-y https://www.nature.com/articles/s41598-021-89990-y .

Master theses defended 2019:

- Alejandro Ortiz Cortés: CEPA Energy range adjustment with muons. University of Complutense Madrid, Spain 10/09/2019 https://www.iem.cfmac.csic.es/departamentos/nuclear/fnexp/reports/TFM/TFM-A_Ortiz.pdf
- María Inmaculada Posadillo de Bringas: Desarrollo de un prototipo con DSSDs para realizar imagen médica con protones. University of Sevilla, Spain 10/09/2019 https://www.iem.cfmac.csic.es/departamentos/nuclear/fnexp/reports/TFM/TFM-Posadillo_de_Bringas.pdf

- Simulaciones Monte Carlo para el desarrollo de un prototipo de tomo´grafo de protones con aplicación en protonterapia. University of Sevilla, Spain 10/09/2019 https://www.iem.cfmac.csic.es/departamentos/nuclear/fnexp/reports/TFM/TFM-V_Tavora.pdf

Master theses in preparation for September 2021.

- Pedro Martínez Moreno: Reconstrucción de imágenes tomográficas del prototipo de escáner pCT del IEM. University of Complutense Madrid, Spain
- Carlos Ballesteros Bejarano: Estudio de capturas radiográficas de fantomas gruesos empleando el prototipo de escáner de protones del IEM-CSIC. University of Complutense Madrid, Spain
- Amanda Nerio: Experimental tests of a scanner prototype for medical imaging with protons at IEM-CSIC. University of Complutense Madrid, Spain

Deviations:
No major deviations in the project have occurred. However, some scintillator materials could not be thoroughly tested due to reduced availability of crystals, and certain combinations of materials could just not be obtained due to lack of producers.

Gender equality:
The WP9 have aimed for equal gender opportunities.

Milestones:
The final deliverable D9.6 – Summary of test results was handed in July 2021.

Funds:
The funds of the WP9 have been fully exhausted. INFN have had one person employed for 1 year (2019) for the study of large-volume position imaging. In the case of IEM-CSIC funds saved during the covid-19 period was used (after previous approval from EC project officer) for the procurement of a novel digitizer MDPPq-16ch digitizer and a MVMLC VME controller to improve the readout

electronics for the p-CT prototype. Also, thanks to the prolongation the remaining funds for an engineer could be used and a technician Vicente Garcia Tavora was employed (1.10.2020 – 30.08.2021) in order to do the successful final preparations, simulations and experiment with the p-CT prototype.

Final Conclusion

Within the PASPAG collaboration several studies of new promising scintillator materials have been performed. These new materials have been characterized with gamma rays and charged particles. This investigations and comparisons of material and sensors and different couplings have led to prototyping actual performing detectors.

During this 3rd period very promising prototype detectors have been tested.

- We have shown that with specialized algorithms we can, combining the right scintillator with the best readout sensor determining the interaction point of gamma-ray radiation and thus obtain a detector with directional sensitivity that is of outmost interest for directional detecting movement of radioactivity in a crowd of people e.g., at a transfer hall of an airport.
- We have shown another application where we can determine the ^{235}U enrichment in bulk ^{238}U samples that is of highest importance to observe and control of international agreements on the composition of fuel for nuclear reactors are being followed.
- We have also shown very promising results of a prototype for p-CT imaging.

Our R&D will not stop here even though the ENSAR2 is coming to an end. In the case of p-CT imaging we already have beamtime approved to further improve of the prototype and to move towards even bigger phantoms and higher proton energies to fully match with the actual clinical situation.