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Advances on the development of the detection system of C-BORD's rapidly relocatable tagged neutron inspection

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The European H2020 project entitled "effective container inspection at border control point" (C-BORD) focuses on the development and in-situ tests of a comprehensive, cost-effective solution for the generalized non-intrusive inspection (NII) of containers and large-volume freight at the European Union border. The opening procedures of suspect containers are time consuming and expensive for economical and safety reasons; therefore, to reduce such operations, the C-BORD project aims to develop a set of technologies that can improve the quality of NII. Among these techniques, a tagged neutron inspection system is being developed in the C-BORD project. It will be a second-line defense system, to be used on sealed containers to detect explosives, illicit drugs, and chemical agents in suspect voxels (elementary volume units). This method employs a beam of tagged neutrons and a set of NaI(TI) and LaBr₃(Ce) scintillators, which will be used to detect prompt gamma rays produced by the neutron interactions. Here we report the advances on the development of the C-BORD's rapidly relocatable tagged neutron inspection system, in particular the comprehensive characterization of the NaI(TI) and LaBr₃(Ce) gamma detectors (time and

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energy resolutions, high-count-rate behavior), the digital analysis for time-coincidence measurements and the data acquisition system (DAQ).

Keywords: Tagged neutron inspection system; non-intrusive inspection; C-BORD.

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1. Introduction

The mission of C-BORD¹ is to develop and test a comprehensive cost-effective solution for the generalized inspection of container and large-volume freight. To protect European Union (EU) borders, the project copes with a large range of container non-intrusive inspection (NII) targets, including explosives, chemical warfare agents, illicit drugs, tobacco, and special nuclear materials. In comparison with the technologies currently used (mainly imaging system based on X or gamma rays), C-BORD is expected to increase interdiction of illicit material in containerized freight and deliver new capabilities against critical operational requirements and constraints by: increasing throughput of containers per time unit; reducing need for costly, time-consuming, and dangerous manual container inspections; and lowering false negative and false positive alarm ratios. The main technical objective of C-BORD is to develop a toolbox of TRL-7 first-line and second-line devices employing different non-destructive passive and active techniques: advanced radiation management, next-generation cargo X-ray, tagged neutron inspection, photo-fission and evaporation-based detection. The idea is to develop a unique graphical user interface (GUI) for all systems: a data-fusion display, decisionsupport software, and a common data format. At the end of the project, recommendations and tools for standardized tests for the different C-BORD NII technologies will be elaborated. In this work we present the progress on the development of the C-BORD's rapidly re-locatable tagged neutron inspection system (RRTNIS). In particular, we give the results concerning the comprehensive characterization of the gamma scintillator detectors. We studied the response of three different photomultipliers, to be coupled to large-sized NaI:Tl crystals, to find the one that satisfies all the requirements of our system. Moreover, we assembled two new LaBr₃:Ce detectors that were completely characterized.

2. C-BORD's Tagged Neutron Inspection System

The concept of the C-BORD's tagged neutron inspection technique can be explained simply as follows. Fast neutrons ($E_n = 14$ MeV), produced using the D-T fusion reaction (D + T \rightarrow n + α), are tagged in time and direction by detecting the associated alpha particles using a position-sensitive detector (YAP:Ce scintillator coupled to a multi-anode photomultiplier). Characteristic de-excitation gamma rays are emitted in all directions when these neutrons interact, mostly by inelastic scattering, with some nucleus belonging to a large target (for example a cargo container). Using a gamma detector array, one can record the neutron-gamma time-of-flight (ToF) spectrum and the gamma energy spectrum of the emitted photons. By selecting only a window of the ToF spectrum and a particular tagged neutron incident direction, one can inspect a small voxel of the entire target. The analysis of the selected gamma energy spectra is performed by comparing those with a database of elementary gamma signatures. A simple scheme of the C-BORD's Tagged Neutron Inspection System (TNIS) is shown in Fig. 1.



Fig. 1. General scheme of the C-BORD's tagged neutron inspection system (TNIS).

The detection module of C-BORD's TNIS consists of a gamma detector array composed of twenty large-sized 5 in \times 5 in \times 10 in NaI:Tl and four 3 in \times 3 in LaBr₃:Ce scintillators (see Fig. 2). The electronics chain for signal processing is based on fast digitizers, and the custom-made data acquisition software denominated ABCD² complete the system. The nuclear electronics chain is composed of two CAEN V1730 VME Digitizers, 16 ch, 14 bit, 500 MS/s (Samples/second); five CAEN V6533 VME HV supplies, 6 ch (NEG), 4 kV/3 mA; and one CAEN V2718 VME-PCI Optical Link Bridge.



Fig. 2. View of NaI:Tl (left) and LaBr₃:Ce (right) scintillators.

The NaI:Tl detectors that will be used to assemble the C-BORD's TNIS were also used in a previous project, called EURITRACK.^{3–5} These detectors must satisfy the C-BORD project requirements. The main requirements are good energy resolution (\sim 7–8%

at 662 keV), good time resolution (< 3 ns, thresh. ~500 keV), linearity, and gain stability for counting rates up to ~200 kcps (counts-per-second). In the case of LaBr₃:Ce detectors, the requirements for the linearity and gain stability were for counting rates up to ~20 kcps, as well as for energy and time resolutions approximately $2\times$ the NaI:Tl detectors.

3. Experimental Set Up

After a preliminary assessment of the energy resolution, time resolution, and gain stability of the NaI:Tl, it was determined that not all the detectors satisfy the requirements of the C-BORD's TNIS.² Therefore, it was necessary to refurbish at least 10 of the 20 NaI:Tl detectors. In particular, the photomultipliers (PMTs) and voltage dividers had to be replaced. The physical integrity of all crystals was checked as well. The crystal/PMT optical couplings were tested with several optical interfaces (optical greases with different viscosities and silicon pads). The best results were obtained using optical grease with high viscosity.

In this work we present a comparative study of three different PMTs to select the best match for the project's requirements. Table 1 shows the main technical characteristics of the PMTs under study. Active voltage dividers (VD), delivered by Hamamatsu and ETEL, were used respectively.

Table 1. Comparison of the main technical characteristics of the three 1 wirs under study.					
	ET9390	Hamamatsu R877-100	Hamamatsu R11833-100HA	XP4512*	
Photocathode	bialkali				
Spectral range (nm)	300-630	300–650		290–630	
Active diameter (mm)	115	111		110	
Luminous sensitivity (muA/Im)	75	90	105	70	
QE (%)	28		35	24	
Nominal anode sensitivity (A/Im)	50	40	50	70	
Gain at nominal	7×10^5	4.4×10^5	2×10^5	2×10^7	
Dark current typ (nA)	1	10	8	90	
Rise time (ns)	13	20	4	2.5	
Transit time (ns)	60	115	49		

Table 1. Comparison of the main technical characteristics of the three PMTs under study.

* Legacy Photonis PMT from the former project. No longer commercially available.

We give the results of only two of the four LaBr₃:Ce detectors. These two crystals were coupled, using an EJ-560 silicon pad, to two Hamamatsu R10233-100-01 photomultipliers, which is a model especially designed for this kind of crystal, with very high light outputs.

Two experimental set ups were used to perform a comprehensive characterization of the gamma detectors. With the first set up, the energy resolution, the linearity, and the gain stability of the detectors were tested. With the second set up, the time resolution of the detectors was measured. The electronic chain and the DAQ used to perform these measurements were the same that are going to be used in the final integration of the TNIS (described in Sec. 2).

3.1. Energy resolution, linearity and gain stability

The energy resolution, the linearity, and gain stability of the detectors were measured as a function of the counting rate. To perform such tests, a set of calibration gamma sources (¹³⁷Cs, ²²Na, ⁶⁰Co, ⁵⁴Mn and ⁸⁸Y) was used; each source had activities ~370 kBq. All the sources were used at the same time to reach a counting rate between 250 kcps and 300 kcps when the sources were placed very close to the NaI:Tl detectors. Two detectors were evaluated in one run (we performed several runs). A run consisted of moving all the sources from a position far away from the detectors (~2.5 m, having a counting rate ~2.5 kcps in the case of NaI:Tl detectors) to a very close position. When moving the sources, the energy spectra of both detectors were continuously acquired. For each event, the digitizer saved the timestamp and the long integration of the corresponding analog pulses. Therefore, this information let us reconstruct the energy spectra at some instant $t \pm \Delta t$ (corresponding to some counting rate value, $C_R \pm \Delta C_R$).

3.2. Time resolution

Coincidence measurements using ²²Na and ⁶⁰Co sources were performed to get the time resolution of the detectors at different energy thresholds. A 2 in \times 2 in EJ-228 fast plastic scintillator was used as the start detector in the case of the NaI:Tl scintillator study. Regarding the LaBr₃:Ce scintillators, being fast and identical detectors, the coincidence measurements were done using just the two detectors under study. For all time measurements, a new firmware, developed for the V1730 digitizer, containing a digital constant-fraction discriminator was employed. In this way, the reconstruction of the coincidences and the time spectrum were done on-line without any off-line post-processing.

4. Results

4.1. NaI(Tl) detectors: energy resolution and linearity

Figure 3 shows the energy calibration and resolution curves of the NaI:Tl detectors coupled to the three PMTs under evaluation. All detectors exhibit a very linear behavior, at least up to 2.734 MeV, which is the most energetic gamma photon of our calibration set (⁸⁸Y). The energy resolution curves are very similar; the small differences could be explained because we used three different crystals to perform the measurements. The energy resolution obtained at 662 keV was ~8%.



Fig. 3. (left) Energy calibration and (right) resolution curves for the three PMTs under study.

4.2. NaI(Tl) detectors: high counting rates

Figure 4 presents the energy resolution and the relative peak position (at 662 keV) as a function of the counting rate for the three NaI:Tl detectors under study. The energy resolution increases as the counting rate gets higher; however, at 200 kcps the Hamamatsu photomultipliers show an increase less than 5% of the energy resolution value at low counting rates.



Fig. 4. (left) Energy resolution and (right) relative peak position at 662 keV as a function of the counting rate for the three PMTs under study.

Concerning the gain stability of the detectors, we explored the relative peak positions (at 662 keV) as a function of the counting rate, and we concluded that the ET9390 and the Hamamatsu R11833-100HA are the best PMTs. Those detectors exhibited a very stable behavior up to 200 kcps. In fact, the R11833-100HA is able to work properly even at more than 260 kcps. On the other hand, the R877-100 PMT showed a very large gain shift (around 50% at 200 kcps) and was consequently discarded to refurbish our NaI:TI detectors.

4.3. LaBr₃(Ce) detectors: energy resolution and linearity

Figure 5 shows the energy calibration and resolution curves (up to 2.7 MeV) of the two LaBr₃:Ce detectors. Both PMTs working at -1200 V show very good linearity.

Concerning the energy resolution, we obtained similar results for both detectors, having an average resolution value of \sim 3.5% at 662 keV being acceptable according to the literature.^{6,7}



Fig. 5. (left) Energy calibrations and (right) energy resolution curves for the two 3 in \times 3 in LaBr₃:Ce scintillators.

4.4. LaBr₃(Ce) detectors: high counting rates

Figure 6 presents the energy-resolution values and the relative peak position (at 662 keV) as a function of the counting rate for both detectors under study. The energy resolution is very stable even up to 120 kcps, the variations are estimated to be lower than 5% with respect to the values at low counting rates for both detectors. Concerning the gain stability, we observed that LaBr – A is more stable than LaBr – B; however, the gain shift exhibited by the latter is very small (~1% at 120 kcps).



Fig. 6. (left) Energy resolution and (right) relative peak position at 662 keV as a function of the counting rate of the two 3 in \times 3 in LaBr₃:Ce scintillators.

4.5. Time resolution

The on-line digital constant fraction discrimination (DCFD), embedded in the firmware of the digitizer V1730, has to be properly configured to achieve an optimal performance.

The parameters delay (D) and fraction (F), which are used to determine the fine timestamp of the event, were studied for both types of detectors (NaI:Tl and LaBr₃:Ce). As an example, Fig. 7 presents the time-resolution values of the LaBr₃:Ce detector's system for different possible values of D and F, coming from the ²²Na coincidence measurement (using the two 511 keV annihilation photons). In this case, using D = 40 ns and F = 25%, we obtained the best timing performance of the LaBr₃:Ce detectors. The time resolution is represented by the full width at half maximum value (FWHM) of the time-coincidence spectrum. Using the ²²Na coincidence measurements, we obtained 0.50 ± 0.01 ns of time resolution (thresh. ~500 keV) for each LaBr₃:Ce detector. With the ⁶⁰Co, 1173 keV and 1332 keV correlated photons, (thresh. ~1 MeV) the result was 0.40 ± 0.01 ns.



Fig. 7. Time resolution (FWHM) of the LaBr₃:Ce system as a function of the DCFD delay for different values of fraction.

Concerning the NaI:Tl detectors, Table 2 shows the time-resolution values for the three different PMTs under study. From our tests, we found out that Hamamatsu R11833-100HA PMT shows the best performance, with a time resolution lower than 3 ns using an energy threshold ~500 keV. The others PMTs do not comply with the requirements of the C-BORD's TNIS.

Table 2. Time resolution results for the NaI:Tl detectors.				
РМТ	Time Resolution (²² Na), ns	Time Resolution (⁶⁰ Co), ns		
ET9390	3.9 ± 0.2	3.0 ± 0.1		
R877-100	3.5 ± 0.1	2.5 ± 0.1		
R11833-100HA	2.5 ± 0.1	1.8 ± 0.1		

5. Conclusions

In this work we presented the advances on the development of the detection module of the C-BORD's TNIS compared to our previous work in Ref. 2. In particular we performed a complete characterization of a part of the gamma detectors set. We studied the energy resolution, linearity, time resolution, and gain stability at high counting rates of different crystal/PMT combinations. We evaluated three different PMTs to be coupled to the large-sized NaI:Tl crystals, and we assembled and characterized two new $3 \text{ in } \times 3$ in LaBr₃:Ce detectors.

The LaBr₃:Ce scintillators (coupled to a Hamamatsu R10233-100-01 PMT) exhibited an energy resolution around 3.5% at 662 keV and a time resolution (FWHM) around 0.5 ns (with a threshold of 500 keV). These values are in good agreement with the literature. Concerning the refurbishment of the NaI:Tl detectors, we found that the Hamamatsu R11833-100 PMT exhibits the best performance. The detector assembled with this PMT shows an energy resolution ~8% at 662 keV, a time resolution ~2.5 ns (²²Na coincidence measurements), a very good linearity, and finally, a very good stability at high counting rates (no gain shift up to 260 kcps).

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