Investigation of the PGNAA using the LaBr₃ scintillation detector

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ABSTRACT

The Joint Research Centre of the European Commission develops instrumentation for detection of hazardous materials. In relation to this a new experimental facility was constructed for research into methods applying the detection of characteristic gamma rays subsequent to neutron irradiation. This includes the detection of prompt gamma rays from neutron inelastic scattering and neutron capture. For this purpose the device employs LaBr₃ scintillation detectors. The paper investigates the applicability of the LaBr₃ scintillation detector to PGNAA.

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1. Introduction

The Joint Research Centre of the European Commission develops instrumentation and analysis methods for the non-destructive assay of nuclear materials and for detection of contraband materials. In relation to this, a new experimental device was designed and constructed in the laboratory of the Institute for the Protection and the Security of the Citizen (IPSC). The device, called the Pulsed Neutron Interrogation Test Assembly (PUNITA), incorporates a pulsed (D–T) neutron generator. As such the instrument finds applications in many different areas although the main research activities concern NDA methods for nuclear safeguards, and for detection of illicit trafficking of nuclear and non-nuclear materials (Favalli and Pedersen, 2007; Favalli et al., 2007).

A wide range of methods have been proposed in the past for automated, non-destructive detection of shielded hazardous/contraband materials located inside consignments of luggage or goods. In the PUNITA facility we study the methods which apply the detection of characteristic gamma rays subsequent to neutron irradiation (Gozani et al., 2003). Many types of neutron reactions will cause the emission of characteristic gamma rays from a target material. Among these reactions, known to the method of prompt gamma neutron activation analysis (PGNAA), are in-elastic scattering (n,n'γ) by fast neutrons and the neutron capture reaction (n,γ) by thermal neutrons. The gamma energy from such reactions is characteristic to the elements present in the sample. By gamma spectroscopy elements of the sample material can be identified, and to some extent the element composition through determination of gamma line ratios. The elements of our interest include oxygen, hydrogen, nitrogen, carbon, flour and chlorine which emit characteristic gamma energies in the range of 1–11 MeV. An interesting possibility currently being studied in PUNITA consists of combining both inelastic scattering and capture in the same measurement. In relation to this research activity we have taken into operation a newly developed scintillation detector based on the LaBr₃:Ce crystal. This detector offers substantial advantages over the standard NaI(Tl) detector: a better energy resolution < 3% at 662 keV, (Milbrath et al., 2005), and a higher efficiency for high-energy gamma rays (Favalli et al., 2008a). The detector crystal has other advantages such as a high scintillation light output with a fast decay time (Milbrath et al., 2005). Recently we acquired a new and larger lanthanum bromide detector (2×3”) including a PMT which is better adapted to the detection of MeV gamma rays in a complex gamma and neutron background. We have tested the capabilities of the new lanthanum bromide detector with respect to the requirements to perform PGNAA in the PUNITA facility. These requirements include detector shielding configurations, efficiency calibration, energy linearity and resolution. The paper presents the procedures used and the results of this work.

2. Experimental method

A 50.8 mm (diameter) × 76.2 mm (length) (2×3”) LaBr₃:Ce scintillation detector, coupled with a XP5500B/02 photomultiplier, and connected to a standard ORTEC electronic chain (Multichannel Analyzer MCA-927 and Spectroscopy Amplifier-673) was investigated. The gamma spectra were acquired with the MAESTRO MCA software.

2.1. Efficiency evaluation

For the evaluation of a calibration with respect to the full-energy peak efficiency, the distance between the front face of the
scintillation detector and the source was set to 100 and 250 mm, respectively.

At gamma energies below 3 MeV standard radioactive sources such as $^{22}$Na, $^{60}$Co, $^{133}$Ba, $^{137}$Cs, $^{155}$Eu and $^{228}$Th were used. The uncertainty of the source activities were 3% or 5% ($^{228}$Th) at the 95% confidence level. Above ~3 MeV calibrated standard sources are not available. In order to extend the full-energy peak efficiency calibration above 3 MeV, a $^{66}$Ga source was produced.

$^{66}$Ga is an important radionuclide for efficiency calibration because it produces strong gamma lines in the energy region from 500 to 5000 keV (Favalli et al., 2008a; Baglin et al., 2002). The nuclide can be produced by the $^{66}$Zn(p,n)$^{66}$Ga reaction ($T_{1/2}=9.5$ h). For this purpose a natural Zn foil with a thickness of 80 μm was irradiated for 30 min in a 17-MeV proton beam. This service was provided by the MC40 cyclotron of the Joint Research Centre. The activity of the activated $^{66}$Ga was estimated by means of the SRIM (Ziegler, 2002) code to be $3\times10^4$ kBq at the end of the irradiation. After a cooling time of 1.2 day the gamma spectrum was accumulated.

From the numerous strong gamma lines of $^{66}$Ga, we used nine lines in the energy region between 500 keV and 5 MeV for the efficiency calibration. It is important to note that in the energy range below 3 MeV, where radioactive sources were used for calculating the efficiency of the scintillation detector, there are also gamma lines of the $^{66}$Ga. These lines should yield the same quantitative efficiency behavior as those obtained from the radioactive sources. Fig. 1 presents the experimental results obtained. In the plot the errors associated with measurements are also reported. The experimental points exhibit a linear behavior in the range up to 5 MeV in log–log plot, and may be fitted by the following equation:

$$
\varepsilon = a \left( \frac{E}{E_0} \right)^b
$$

where $\varepsilon$ is the full-energy peak efficiency, $E_0$ is an arbitrary energy to make the argument dimensionless [here, $E_0=1$ keV], $a$ and $b$ are the two coefficients of the fitting process of the experimental data. The results of the least squares fit are reported in Table 1.

### 3. Experimental PGNAA measurements

#### 3.1. PGNAA setup description

In order to investigate the applicability of the LaBr$_3$ scintillation detector for the PGNAA method, the following setup was built. An Am–Be neutron source was surrounded by a cylindrical shield in tungsten (W 95%, Fe 3.5%, Ni 1.5%) to attenuate the gamma rays from the neutron source. The neutron source was clad with Cd in order to avoid the interactions of thermal neutrons with the source. A second cylinder of polyethylene was arranged around the tungsten shield in order to thermalize the neutrons produced by the Am–Be source. The test sample was placed outside the cylinder and close to the detector. Additional polyethylene blocks were placed around the sample to achieve a higher thermal neutron flux. A sketch of the whole arrangement is given in Fig. 2.

#### 3.2. Example of PGNAA spectra

For the preliminary experiments we chose ammonium chloride (NH$_4$Cl) as test sample. This salt is interesting as it contains elements typical for contraband materials. The spectrum collected is reported in Fig. 3. Fig. 3 also gives the identification of the characteristic gamma lines. The spectrum shows for example both the characteristic gamma ray at 4.438 MeV emitted by $^{12}$C due to the reaction $^9$Be($^3$n,$^3$p) from the neutron source, and the gamma line at 8.998 MeV due to the presence of Ni in the tungsten shield.

Due to the high efficiency and the high energy resolution (from 2.7% at 662 keV to ~0.7% at 9 MeV) of the scintillation detector, the numerous gamma lines from Cl are well distinguished. The gamma lines of N at 10.829 MeV is also visible. The gamma lines

![Schematic of the set-up employed for the PGNAA measurements.](image)
from N and Cl can be used for the PGNAA evaluation. Also the gamma line from H is clearly visible, but in this case the peak has a large contribution from the H of the polyethylene moderator. For comparison a spectrum of the same sample collected with an HPGe planar detector (P11 portable hand held planar detector by BSI—Baltic Scientific Instruments) is shown in Fig. 4. In this case a cadmium cup was placed on the detector in order to reduce the neutron interactions in the detector. The better energy resolution of the HPGe detector allows the gamma line of Cd at 9.043 MeV to be distinguish from the gamma line at 8.998 MeV of the Ni.

3.3. Linearity

The linearity of the output signal is an important characteristic of a gamma-ray detector when used for a broad range of MeV gamma lines. For an ideal scintillation detector, the scintillation efficiency or the amount of light generated per unit energy loss (dL/dE) would be constant independent of gamma-ray energy. The total light yield is then directly proportional to the incident gamma-ray energy. In order to investigate the linearity response of the LaBr₃ scintillation detector, we have analyzed the spectrum.
measured with the NH₄Cl reported in Fig. 3. Fig. 5 shows the experimental results obtained: experimental points exhibit a linear behavior. Due to this feature the interpretation of the PGNAA spectra is made easy in the gamma energy range of interest. For comparison we recently published (Favalli et al., 2008a, b) experimental results showing how a 1.5” × 1.5” LaBr₃ scintillation detector, coupled with a XP2060B photomultiplier, exhibits a non-linear behavior for gamma-ray energies > ~5 MeV, causing difficulties in the identification of gamma lines at high energies (> ~7 MeV). The superior performance of the present detector may in this respect be attributed to the photomultiplier tube.

4. Conclusions and remarks

The paper describes features of a 2” × 3” LaBr₃ scintillation detector important for PGNAA applications intended for detection of contraband/hazardous materials. The PGNAA method is a promising tool for the detection of contraband/hazardous materials. The paper investigated the applicability of the LaBr₃ scintillation detector to PGNAA in an energy range up to 11 MeV. The results presented show that the detector promises to be a suitable detector for the PGNAA application due to excellent properties such as high energetic resolution, high efficiency and elimination of cooling requirements as for HPGe detectors.

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References


