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Commissioning of the UK NAtional Nuclear Array

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ABSTRACT

The NAtional Nuclear Array (NANA) is a $LaBr_3$(Ce)-based coincidence gamma-ray spectrometer which can be used to identify, and enhance with respect to the background, signature gamma-ray emissions associated with particular radionuclide decays from a complex multi-component spectrum. Gamma-ray energy coincidence measurements using the NANA have been made using a digital data acquisition system based on CAEN V1751C 1 GHz digitizers. The improved time resolution offered by $LaBr_3$(Ce) crystals compared to similar-sized solid state detectors can provide narrow time-correlated, gamma-ray energy coincidence matrices that can be interrogated to select discrete gamma-ray emissions associated with particular radionuclide decays. This paper provides an overview of the operational characteristics of the NANA spectrometer, including energy resolution and full-energy peak efficiency parameters, and provides an example of double and triple gamma-ray coincidence gating on decays associated with the nuclear fuel waste product ^{134}Cs. The full-energy peak efficiency response of the spectrometer is compared to Monte Carlo GEANT4 simulations.

1. Introduction

The NAtional Nuclear Array, NANA, is a 12 element $LaBr_3$(Ce) scintillation detector array, based at the UK's National Physical Laboratory (NPL). It is designed for use in nuclear spectroscopy measurements by the detection of discrete energy, characteristic gamma rays emitted from excited nuclear states (Bucurescu et al., 2016; Lorusso et al., 2016; Regan et al., 2015a, 2015b). $LaBr_3$(Ce) scintillator crystals are currently of interest to the radiation measurement community as a gamma-ray detector material which combines fast timing response and sufficient energy resolution (Browne et al., 2015; Bucurescu et al., 2016; Lalkovski et al., 2015; Regan et al., 2012, 2013, 2015a, 2015b; Régis et al., 2014a, 2014b; Roberts et al., 2014; Rudigier et al., 2015; Werner et al., 2016). Spectroscopic quality $LaBr_3$(Ce) scintillator detectors are currently utilised in a number of nuclear structure and decay physics experiments worldwide; for example as part of the DESPEC-FATIMA collaboration (Lalkovski et al., 2015; Regan et al., 2012; Roberts et al., 2014) and the RoSPHERE gamma-ray spectrometer at IFIN-HH Bucharest (Bucurescu et al., 2016). In many of these configurations, the $LaBr_3$(Ce) detectors are complemented with high-resolution hyper-pure germanium (HPGe) detectors in hybrid set-ups that provide superior timing triggers for gamma-ray cascades, allowing for the measurement of nuclear excited state lifetimes down to ~10 ps (Régis et al., 2014a, 2014b).

This paper presents the results of commissioning measurements for the NAtional Nuclear Array. Experimentally determined values for the full-energy peak detection efficiency and linearity of energy response are measured using well-characterised, mixed-isotope radioactive sources. These full-energy peak efficiency (FEP) measurements are compared to simulations of the array's response which were made using the GEANT4 Monte Carlo code (Agostinelli et al., 2013). The application of the NANA for the selection of two and three gamma-ray decay cascades from a mixed radioactive source of ^{134}Cs and ^{137}Cs is also investigated, highlighting the potential use of this $LaBr_3$(Ce) array for future radionuclide (waste) assay.

2. Dimensions, modelling and measurement of the response of the NAtional Nuclear Array

In order to maximise the functionality of the array for different applications, a GEANT4 simulation of the spectrometer was constructed within the NPTooL framework (Matta et al., 2016). The GEANT4 simulation includes the front window, the can, the detector material and a borosilicate glass plate representing the photomultiplier tube. A simple representation of the frame has also been constructed within the model. Fig. 1 shows the constructed model defined within NPTool for the GEANT4 simulation of the NANA array.

The complete NANA design comprised twelve individual $LaBr_3$(Ce)

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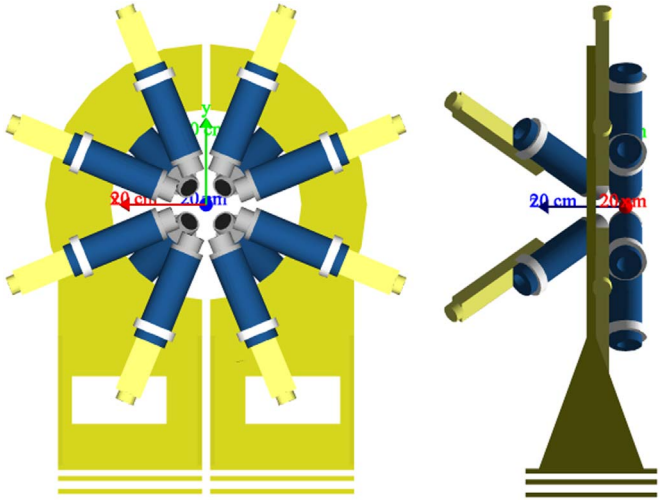

Fig. 1. NPTool model used for the Monte Carlo simulation of the 12 element NANA gamma-ray spectrometer.

Table 1
Detector angles for the twelve positions in the NAtional Nuclear Array. Note that the angles are with respect to the axes given in Fig. 1.

Detector number	Polar Angle/degrees (θ)	Azimuthal angle/degrees (ϕ)
0	90	112.5
1	90	157.5
2	90	202.5
3	90	247.5
4	90	292.5
5	90	337.5
6	90	22.5
7	90	67.5
8	45	45
9	45	135
10	45	225
11	45	315

detectors, with lead shielding to reduce Compton cross talk between detectors. Eight of the detectors are in a central ring with a 45° mutual angular spacing. This central ring is complemented by four additional detectors at backward angles relative to the central annulus. The details of the detector angles in the final NANA geometry are given in Table 1. The typical distance from the central (source) position to the front of the $LaBr_3$(Ce)detector face was 78 mm.

The energy resolution, timing and full-energy peak response of the 8 detectors in the central annulus were measured using a thin film vinyl source mounted on a bespoke 3D printed source holder. The source holder was designed to not shadow any detectors and to minimize any inhomogeneous efficiencies. Using this mixed source, full-energy-peak calibration and efficiency response curves were produced. The latter was used to validate the Monte Carlo simulations for the full-energy peak detection efficiency of the array.

2.1. Linearity and energy calibration of NANA

The $LaBr_3$(Ce) crystal is neither isotopically nor chemically pure, giving rise to an internal radiation background from the detector material (Lorusso et al., 2016). This internal radiation arises from the primordial isotope ^{138}La ($T_{1/2}$=1.02×10^9 years (Bé et al., 2016)), present with an isotopic abundance of 0.090 (1)%. Lanthanum-138 decays by β^- emission (I_{beta}=35%) to the yrast spin/parity $I^\pi=2^+$ state in ^{138}Ce which is followed by the emission of a 788.7 keV gamma ray. The ^{138}La isotope can also decay by electron capture (I_{ec}=65% branch) to the $I^\pi=2^+$ state in ^{138}Ba, followed by a 1435.8 keV gamma ray emission

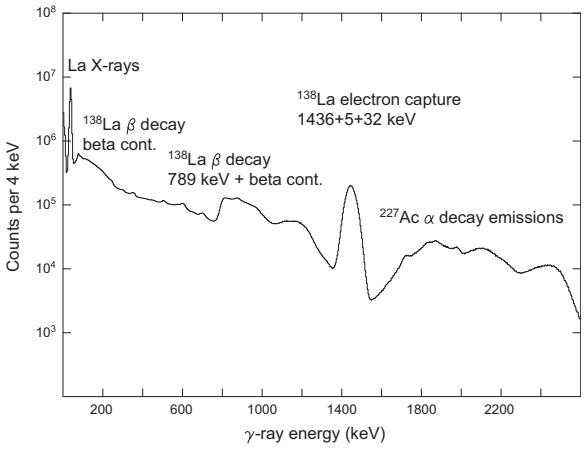

Fig. 2. Measured sum background energy spectrum across the 8 detectors in the central ring of the NANA spectrometer, counted for 86,400 s. The labelled peaks indicate the presence of the primordial ^{138}La isotope in the detector materials and members of the ^{227}Ac decay series. The characteristic Lead X ray caused by the presence of the shield can also be seen.

(Bé et al., 2016) and is accompanied by coincident La K-shell X-rays from the electron capture process. Additional internal radiation is present from the decay of the chemically similar ^{227}Ac ($T_{1/2}$=21.7 years (Browne, 2001)). Actinium-227 decays by β emission to ^{227}Th (and alpha emission to ^{223}Fr) which subsequently decays by alpha emission through the actinium (4 n+3) natural decay series. The alpha decays of the subsequent (main) members of this decay chain (Browne, 2001) ($T_{1/2}(^{227}Th)$=18.697 days (Browne, 2001); $T_{1/2}(^{223}Ra)$=11.4 days (Browne, 2001); $T_{1/2}(^{219}Rn)$=3.96 s (Browne, 2001); $T_{1/2}(^{215}Po)$ =1.78 ms (Browne, 2001)) have characteristic alpha particle decay energies in the range 6–7.5 MeV, but these are quenched by the response of the detector and PMT, and are observed in the detector response spectrum and gamma-ray equivalent energies of between 1.7 MeV and 2.6 MeV.

Fig. 2 shows the energy calibrated response of the sum of the 8 $LaBr_3$(Ce) detectors from the central ring of the NANA spectrometer, with no additional source present.

The variability in the linearity of the energy response across the different $LaBr_3$(Ce) detectors in NANA means that the energy calibration procedure of the $LaBr_3$(Ce) detectors can be more complex than for standard HPGe detectors (for which often a simple linear or quadratic calibration response function will provide good gain matching over an energy range 50 keV–2 MeV). The linearity of energy response for the eight detectors in the central ring of NANA was investigated using a thin VYNS mixed source with 13 prominent characteristic gamma emissions ranging from 60 keV from ^{241}Am to the 1332 keV line of ^{60}Co. The acquired singles spectrum for one of the detectors can be seen in Fig. 3. Peaks were fitted to a Gaussian function with an n^{th}-order polynomial background. The 1836 keV peak was not fitted due to the interference from the quenched internal alpha emissions in this region of the spectrum. The calibrated response of the detectors can be seen in Figs. 4 and 5. The calibration was achieved using a 4th order polynomial. Differences in the response for the high energy ^{60}Co emission (1332 keV) show less than 1% fluctuation across the eight detectors.

During the energy response calibration procedure, the degree of linearity for the detectors was observed to vary across the suite of detectors. The typical range in response from detector to detector can be seen from Fig. 4, where Detector 0 exhibits a greater linearity than Detector 3, whilst operating at the same voltage (1300 V) from the CAEN HV supply unit supplying independent HV to each detector.

The combined energy resolution response of the 8 detectors in the NANA central ring is shown in Fig. 6. In general, the measured full-width half maximum (FWHM) energy resolution for the detectors