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Radon gamma-ray spectrometry with YAP:Ce scintillator

Wolfango Plastino[a,b,*], Pierino De Felice[c], Francesco de Notaristefani[a,b]

[a] *Department of Physics, University of Roma Tre, via della Vasca Navale 84, I-00146 Roma, Italy*
[b] *National Institute for Nuclear Physics, Section of Roma Tre, via della Vasca Navale, 84, I-00146 Roma, Italy*
[c] *ENEA, Department of Environment, National Institute for Metrology of Ionizing Radiations, C.R. Casaccia, P.O. Box 2400, I-00100 Roma, Italy*

Abstract

The detection properties of a YAP:Ce scintillator ($YAlO_3$:Ce crystal) optically coupled to a Hamamatsu H5784 photomultiplier with standard bialkali photocathode have been analyzed. In particular, the application to radon and radon-daughters gamma-ray spectrometry was investigated. The crystal response has been studied under severe extreme conditions to simulate environments of geophysical interest, particularly those found in geothermal and volcanic areas. Tests in water up to a temperature of 100°C and in acids solutions such as HCl (37%), H_2SO_4 (48%) and HNO_3 (65%) have been performed. The measurements with standard radon sources provided by the National Institute for Metrology of Ionizing Radiations (ENEA) have emphasized the non-hygroscopic properties of the scintillator and a small dependence of the light yield on temperature and HNO_3. The data collected in this first step of our research have pointed out that the YAP:Ce scintillator can allow high response stability for radon gamma-ray spectrometry in environments with large temperature gradients and high acid concentrations. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

The YAP:Ce monocrystal has been emphasized as a good detector for gamma-ray spectrometry [1,2]. The properties of YAP:Ce can be summarized as follows [3–6]:

- high light output of 40–50% relative to NaI(Tl);
- density of 5.37 g/cm^3;
- short decay time constant of 27 ns;
- average Z of 39;
- broad spectrum emission peaking at 370 nm;
- very good mechanical and chemical properties; and
- light yield almost independent of the energy deposited by gamma rays in the crystal and negligible afterglow.

The properties of YAP:Ce monocrystal and particularly its emission peak are well coupled with the sensitivity curve of typical photomultipliers and make this detector a useful tool for potential applications in gamma-ray spectrometry in geophysical research.

*Corresponding author. Department of Physics, University of Roma Tre, via della Vasca Navale 84, I-00146 Roma, Italy. Tel.: +39-06-55177-277; fax: +39-06-5579-303.

E-mail address: plastino@fis.uniroma3.it (W. Plastino).

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Tests to estimate the detection properties of this scintillator for radon and radon-daughters gamma-ray spectrometry for environments of geophysical interest such as geothermal, volcanic and seismic areas have been performed. Special attention was paid to measure the light output dependence on temperature and the effect of treatments with acid solutions to estimate the response stability of the scintillator under extreme physical and chemical conditions.

2. Experimental array and method

The radon gamma-ray spectrometer consisted of a bare cylindrical crystal of YAP:Ce (supplied by the Crytur, Ltd.—Czech Republic) optically coupled to a Hamamatsu H5784 photomultiplier with standard bialkali photocathode. The crystal size was 8 mm diameter and 30 mm height. The crystal was positioned at the geometrical center of a 6 l stainless-steel light- and gas-tight vessel equipped with gas input/output and a pass-through electrical connectors.

Output signals were integrated with a charge preamplifier, shaped with a spectroscopy amplifier (ORTEC-450) and processed by a multichannel analyzer card (ORTEC-Trump). The collected pulse-height spectrum was managed by a suitable MCA emulation software (ORTEC-Maestro32).

The radon gamma-ray spectrometer was serially connected to the radon Reference Measurement System (RMS) developed at the National Institute for Metrology of Ionizing Radiations (INMRI-ENEA) [7]. The schematic view of the experimental array is shown in Fig. 1. The RMS is routinely used for calibration and testing of radon measuring instruments [7,8] and it provided the reference radon-in-air activity concentration needed for efficiency calibration of the YAP:Ce radon spectrometer. The RMS is based on a cylindrical electrostatic cell with a Si detector. It is used for the alpha spectrometry of the electrostatically collected polonium ions produced in the decay of radon [7]. The radioactive source section consisted in a 35 cm^3 glass bulb filled with about 15 kBq radon in air. The internal volume of the whole circuit was about 10 l.

The experimental array was used with the following sequence of operations: (a) the radioactive source section was connected to the circuit with the valves F, G and H opened and the valves D and E closed; (b) the rest of the circuit was evacuated, the valve H was closed and the valve C was set in position 3; (c) the valves of the source section were gradually opened in a sequence

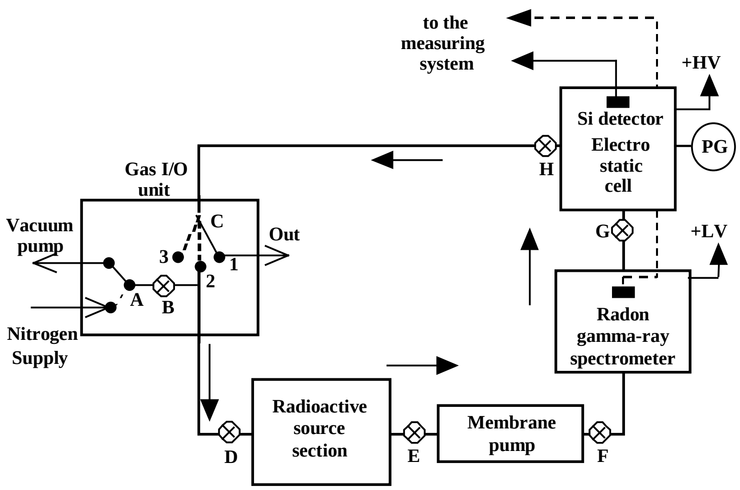

Fig. 1. Schematic view of the experimental array with the radon gamma-ray spectrometer and radon Reference Measurement System of INMRI-ENEA.

opposite to the gas stream direction; (d) the HV and LV power supplies were switched on; (e) the valve C was put in position 2, the valve H was gradually opened and the membrane pump was activated (flow rate of 300 cm^3/min); (f) repeated counting measurements with RMS and with the radon gamma-ray spectrometer were performed; and (g) the valves of the source section were closed and the radon gas was discharged through the gas input/output unit.

The energy calibration curves of the radon spectrometer were obtained irradiating the crystal by means of a set of radioactive point sources of different radionuclides (^{133}Ba, ^{241}Am, ^{57}Co, ^{22}Na, ^{137}Cs) positioned at 5 cm source–detector distance. These provided a number of photons with well-spaced gamma-ray energies up to about 700 keV.

As shown in Fig. 2, the gamma-ray spectrum given by the radon spectrometer when the crystal is introduced in a radon-in-air atmosphere shows two main peaks at about 76 and 609 keV. These are due, respectively, to the X- and gamma-rays emitted by ^{214}Bi. The count rates in the energy windows from 45 to 108 keV and from 532 to 688 keV were taken as crystal response. Corresponding counting efficiency values were obtained dividing these count rates by the radon-in-air activity concentration given by the RMS.

Both energy and efficiency calibrations were performed in normal condition (a) and repeated after different test treatment consisting in crystal immersion, for about 30 min, in each of the following solutions: (b) water at 100°C; (c) HCl (37%); (d) H_2SO_4 (48%); and (e) HNO_3 (65%).

3. Results

A typical energy calibration curve of the radon spectrometer is reported in Fig. 3, in which the energy response linearity is shown. The slopes of the different energy calibration curves normalized to that of normal condition are shown in Fig. 4. These emphasized a good energetic response stability, within 1%.

The peak centroids and the energy resolution for the two observed gamma-ray peaks were independent on the gas pressure inside the radon gamma-ray spectrometer, in the range 100–1030 hPa. For lower pressure values, changes in photomultiplier performance were observed.

The counting efficiencies in normal condition resulted to be 0.029 s^{-1} Bq^{-1} l and 0.019 s^{-1} Bq^{-1} l, respectively, for the two selected counting windows. The counting efficiencies measured after each test treatment were normalized to those obtained in normal condition. The results are shown in Fig. 5.

Both curves show a slight effect of hot water (10%) and HNO_3 (42%) treatments on the counting efficiency. These efficiency variations were probably related to temporal changes in the

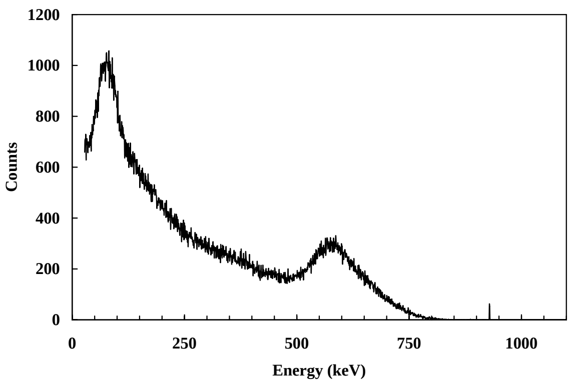

Fig. 2. Energy spectra at normal condition for the YAP:Ce crystal for radon-in-air atmosphere.

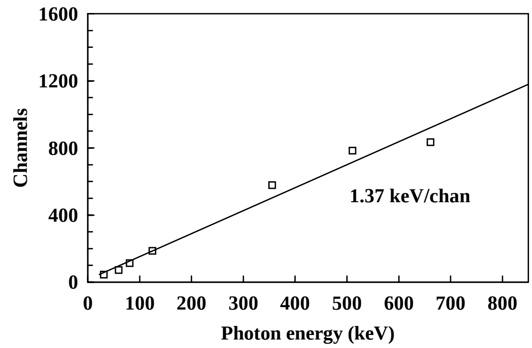

Fig. 3. Energy calibration curve for the YAP:Ce crystal irradiated by means of a set of radioactive point sources of different radionuclides (^{133}Ba, ^{241}Am, ^{57}Co, ^{22}Na, ^{137}Cs) after the treatment (c).

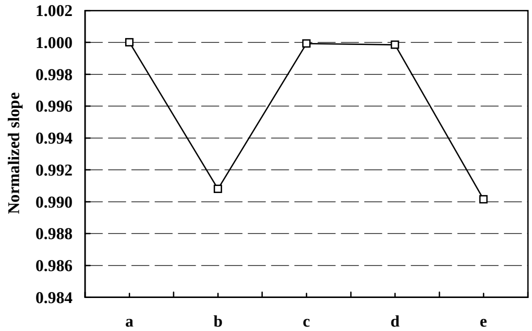

Fig. 4. The normalized slope of the different energy calibration curves respect to normal condition for the YAP:Ce crystal irradiated by means of a set of radioactive point sources of different radionuclides (^{133}Ba, ^{241}Am, ^{57}Co, ^{22}Na, ^{137}Cs) after the different test treatment [a: normal; b: water at 100°C; c: HCl (37%); d: H_2SO_4 (48%); and e: HNO_3 (65%)].

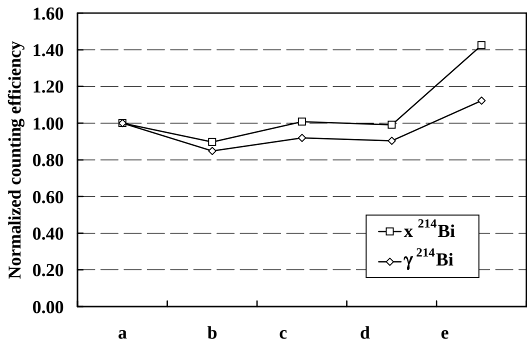

Fig. 5. Normalized counting efficiency respect to normal condition of the YAP:Ce crystal at the different test treatment [a: normal; b: water at 100°C; c: HCl (37%); d: H_2SO_4 (48%); and e: HNO_3 (65%)].

optical properties of the silicon grease spread between the crystal and the photomultiplier window. Also, these data have emphasized the non-hygroscopic and good mechanical properties of the YAP:Ce monocrystal.

4. Conclusions

Tests performed with a YAP:Ce crystal for radon and radon-daughters gamma-ray spectrometry in simulated environments of geophysical interest such as geothermal, volcanic and seismic areas have pointed out the good crystal response stability.

A better performance for the YAP:Ce monocrystal may be obtained after improvement of the crystal to photomultiplier optical coupling. Nevertheless, this scintillator seems to be a new useful tool for radon investigations under extreme physical and chemical conditions.

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