

Radon test measurements with Ce-doped yttrium aluminum oxide perovskite scintillator

W. Plastino^{1,2}, P. De Felice³ and F. de Notaristefani^{1,2}

¹ Department of Physics, University of Roma Tre, Roma, Italy

² National Institute for Nuclear Physics, Section of Roma Tre, Roma, Italy

³ ENEA, Department of Environment, National Institute for Metrology of Ionizing Radiations, Roma, Italy

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RESUMEN

Se analizaron las propiedades de centelleo para espectrometría gamma de radón y sus descendientes, de un monocristal de perovskita de óxido de Al-Y copado con Ce, acoplado ópticamente a un fotomultiplicador Hamamatsu H5784 con un fotocátodo bialcalino.

Las pruebas se realizaron en agua hasta 100°C y en soluciones ácidas de HCl (37%), H₂SO₄ (48%) y HNO₃ (65%) para simular ambientes de interés geofísico como áreas volcánicas y geotérmicas.

Medidas comparativas con fuentes estándar de radón del Instituto Nacional de Metrología de Radiaciones Ionizantes (ENEA) confirman las propiedades no-higroscópicas del centelleador y poca dependencia con el rendimiento en temperatura y HNO₃.

El monocristal muestra alta estabilidad para espectrometría gamma del radón en ambientes con grandes gradientes de temperatura y altas concentraciones de acidez.

PALABRAS CLAVE: Radón, descendientes del radón, espectrometría gamma, geofísica.

ABSTRACT

Scintillation properties of a Ce-doped yttrium aluminium oxide perovskite monocrystal optically coupled to a Hamamatsu H5784 photomultiplier are analyzed with a standard bialkali photocathode for radon and radon daughters gamma-ray spectrometry. Tests in water up to 100°C and in acidic solutions of HCl (37%), H₂SO₄ (48%) and HNO₃ (65%) were performed to simulate environments of geophysical interest, such as geothermal and volcanic areas. Comparative measurements with standard radon sources provided by the National Institute for Metrology of Ionizing Radiations (ENEA) confirm the non-hygroscopic properties of the scintillator and small dependence of the light yield on temperature and HNO₃.

The Ce-doped yttrium aluminium oxide perovskite monocrystal shows high response stability for radon gamma-ray spectrometry in environments with large temperature gradients and high acid concentrations.

KEY WORDS: Radon, radon daughters, gamma-ray spectrometry, geophysics.

INTRODUCTION

Radon content variations in groundwater and soil have been shown to correlate with the crustal stress-strain changes associated with the seismic and volcanic activity (Hauksson, 1981; King, 1986; Segovia and Mena, 1999; Varley and Armienta, 1999; Plastino and Bella, 2001). To better define these space-temporal radon variations it is necessary to perform the measurements with detectors characterized by a high response stability. We have planned tests with a new scintillator to analyze its performance under extreme physical and chemical conditions.

The Ce-doped yttrium aluminium oxide perovskite monocrystal is characterized by a high light output of 40-

50% relative to NaI(Tl), a short decay time constant of 27 ns, a density of 5.37 g/cm³, an emission peak about 370 nm, a light yield almost independent of the energy deposited by gamma rays in the crystal, a negligible afterglow and a high hardness (Baryshevsky *et al.*, 1991; Baccaro *et al.*, 1995; Tsuchida *et al.*, 1997; Moszynski *et al.*, 1998). The characteristics of this scintillator and particularly its emission peak are well coupled with the sensitivity curve of typical photomultipliers and make this monocrystal a new useful tool for potential applications in gamma-ray spectrometry in geophysical research.

Particularly, the first step of this work was to study the detection properties of this scintillator for radon and radon-daughters gamma-ray spectrometry under extreme physi-

cal-chemical conditions to simulate environments of geophysical interest, particularly those found in geothermal, volcanic and seismic areas. Special attention was paid to measure the light output dependence on temperature and different acidic solutions to estimate the response stability of the scintillator.

METHOD

The experimental array consisted of the radon gamma-ray spectrometer connected serially to the radon Reference Measurement System (RMS) developed at the National Institute for Metrology of Ionizing Radiations (INMRI-ENEA) (De Felice and Myteberi, 1996). The internal volume of the whole circuit was about 10 l and the schematic view of the array used is shown in Figure 1.

The radon gamma-ray spectrometer consisted of a cylindrical monocrystal of Ce-doped yttrium aluminium oxide perovskite (8 mm diameter and 30 mm height) optically coupled to a Hamamatsu H5784 photomultiplier with a standard bialkali photocathode. This monocrystal was positioned

in a stainless steel light- and gas-tight vessel with a volume of 6l. The output signals were integrated with a charge preamplifier and shaped with a spectroscopy amplifier (ORTEC-450). The collected signals were processed by a multichannel analyzer card (ORTEC-Trump) and managed by MCA emulation software (ORTEC-Maestro32).

The RMS of INMRI-ENEA is a cylindrical electrostatic cell with a Si detector. It is used for alpha spectrometry of the electrostatically collected polonium ions produced by the decay of radon (De Felice and Myteberi, 1996). The RMS is used for calibration and testing of radon measuring instruments (De Felice and Myteberi, 1996; De Felice et al., 1996) and it provided the reference radon-in-air activity concentration needed for efficiency calibration of the monocrystal.

The radioactive source section was a radon source in gas sample of about 15 kBq provided by INMRI-ENEA (De Felice and Myteberi, 1996).

The experimental array was scheduled by the following sequence of operations:

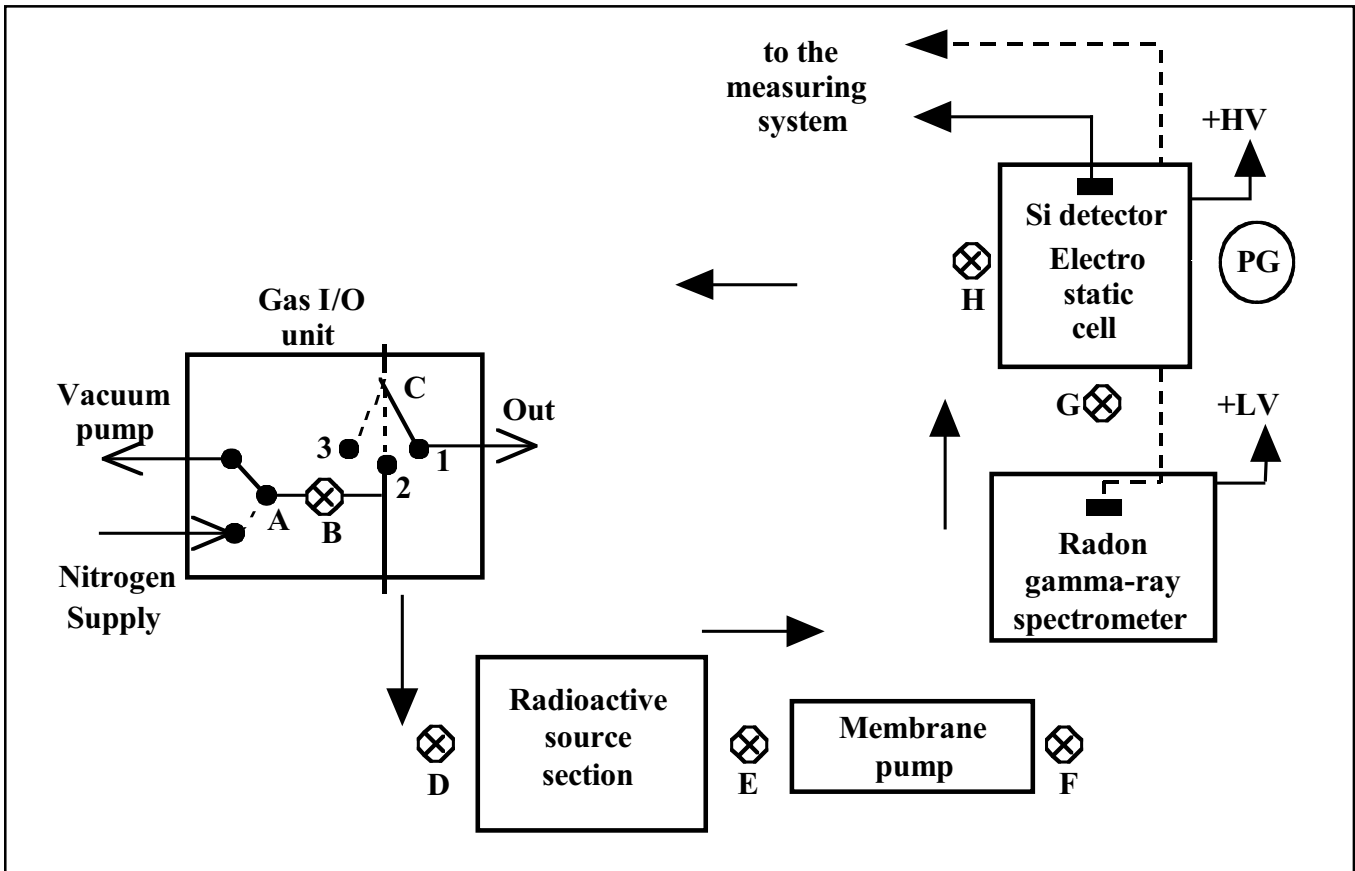


Fig. 1. Schematic view of the experimental array with the radon gamma-ray spectrometer and radon Reference Measurement System of INMRI-ENEA.

a) the radioactive source section was mounted in the RMS circuit as shown in Figure 1 with the valves F, G and H opened and the valves D and E closed; b) the rest of 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 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 and valve H was gradually opened and then the membrane pump was activated (flow rate of 300 cm³/min); f) repeated counting measurements with Si detector and radon gamma-ray spectrometer were performed; g) the valves of the source section were closed and the radon gas was discharged through the gas input/output unit.

The monocrystal was irradiated by means of several radioactive point sources with well spaced gamma-ray energies up to about 700 keV (¹³³Ba, ²⁴¹Am, ⁵⁷Co, ²²Na, ¹³⁷Cs) at 5 cm source-detector distance to obtain the energy calibration curves.

The gamma-ray spectrum determined by this monocrystal in a radon-in-air atmosphere shows the two peaks at about 76 and 609 keV due to the x- and gamma-rays, respectively, emitted by ²¹⁴Bi as shown in Figure 2. The ²¹⁴Bi count rates in the energy windows from 45 to 108 keV and from 532 to 688 keV are assumed as measure of the monocrystal response. These count rates were divided by the radon-in-air activity concentration provided

by the RMS to obtain the counting efficiency values of the monocrystal.

The energy and efficiency calibrations were performed in different test measurements [a: normal; b: water at 100° C; c: HCl (37%); d: H₂SO₄ (48%); e: HNO₃ (65%)]

RESULTS

The energy calibration curves of the radon gamma-ray spectrometer are shown in Figure 3 with the related energy response linearity. Energetic response stability lies within 1%.

Peak centroid and energy resolution for the two observed gamma-ray peaks are independent of the gas pressure inside the radon gamma-ray spectrometer in the range 100-1030 hPa.

The counting efficiencies in normal condition resulted to be 0.029 s⁻¹Bq⁻¹ and 0.019 s⁻¹Bq⁻¹, respectively, for the two selected windows.

To estimate the response stability of the monocrystal related to the different test measurements we calculated the normalized counting efficiency with respect to normal conditions as shown in Figure 4. Both curves show a dependence of the counting efficiency on hot water (10%) and

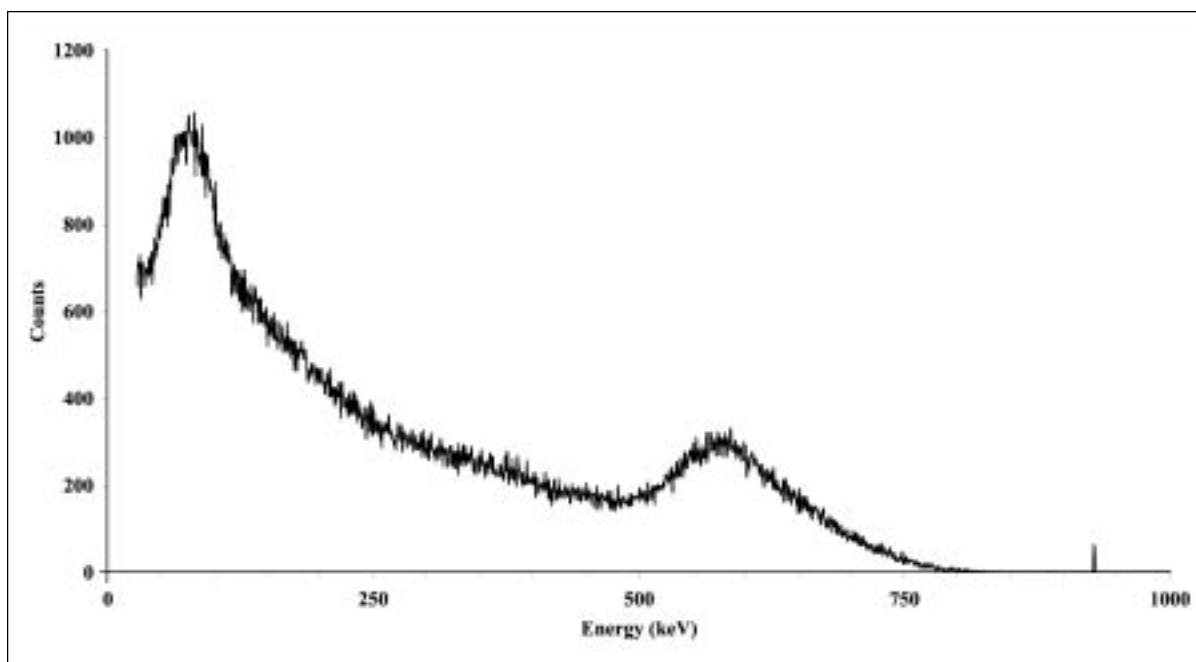


Fig. 2. Energy spectra at normal condition for the Ce-doped yttrium aluminium oxide perovskite monocrystal for radon source in gas sample provided by INMRI-ENEA.

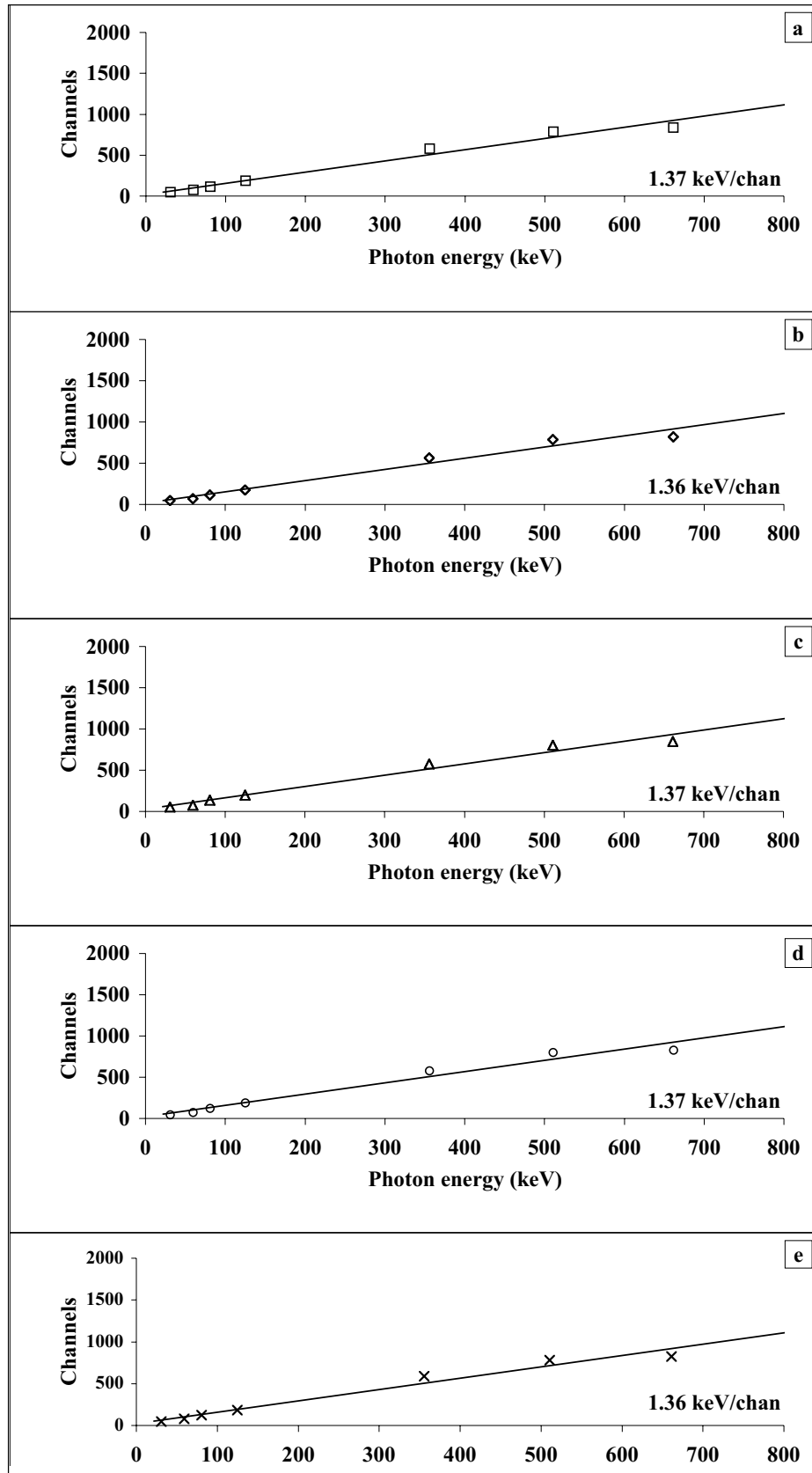


Fig. 3. Energy calibration curves for the Ce-doped yttrium aluminium oxide perovskite monocrystal irradiated by means of several radioactive point sources (^{133}Ba , ^{241}Am , ^{57}Co , ^{22}Na , ^{137}Cs) at the different test measurements [a: normal; b: water at 100°C; c: HCl (37%); d: H_2SO_4 (48%); e: HNO_3 (65%)].

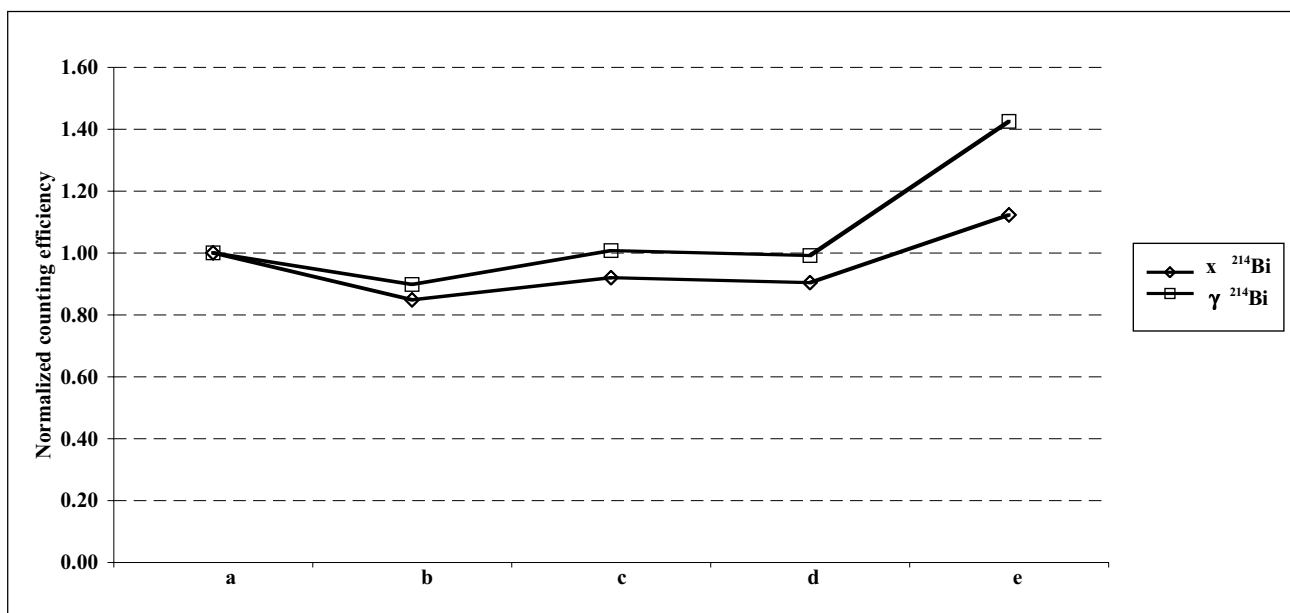


Fig. 4. Normalized counting efficiency with respect to normal condition of the Ce-doped yttrium aluminium oxide perovskite monocrystal at the different test measurements [*a*: normal; *b*: water at 100°C; *c*: HCl (37%); *d*: H₂SO₄ (48%); *e*: HNO₃ (65%)].

HNO₃ (42%) test measurements. These efficiency variations were probably related to temporal changes in the optical properties of the silicon grease spread between the monocrystal and the photomultiplier window. Also, these data have confirmed the non-hygroscopic properties of the scintillator.

CONCLUSIONS

The tests performed with a Ce-doped yttrium aluminium oxide perovskite monocrystal for radon and radon-daughters gamma-ray spectrometry under extreme physical and chemical conditions have pointed out its good response stability. A better performance for this crystal may be obtained after an improvement of the optical coupling with the photomultiplier. Nevertheless, this scintillator seems to be a new useful tool for radon investigations in geophysical environments such as geothermal, volcanic and seismic areas.

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W. Plastino^{1,2*}, P. De Felice³ and F. de Notaristefani^{1,2}

¹ Department of Physics, University of Roma Tre, Via della Vasca Navale, 84, I-00146 Roma, Italy

² National Institute for Nuclear Physics, Section of Roma Tre, Via della Vasca Navale, 84, I-00146 Roma, Italy

³ ENEA, Department of Environment, National Institute for Metrology of Ionizing Radiations, C. R. Casaccia, P.O. Box 2400, I-00100 Roma, Italy

*Corresponding author: tel: +39 06 55177277, fax: +39 06 5579303, e-mail: plastino@fis.uniroma3.it