

EPJ Web of Conferences **65**, 02001 (2014)
DOI: 10.1051/epjconf/20146502001
© Owned by the authors, published by EDP Sciences, 2014

Response of parylene-coated NaI(Tl) scintillators at low temperature

Noël Coron¹, Clara Cuesta^{2,3,a}, Eduardo García^{2,3}, Carlos Ginestra^{2,3}, Thomas A. Girard⁴, Pierre de Marcillac¹, María Martínez^{2,3,5,b}, Ysrael Ortigoza^{2,3}, Alfonso Ortiz de Solórzano^{2,3}, Carlos Pobes^{2,3}, Jorge Puimedón^{2,3}, Thierry Redon¹, María Luisa Sarsa^{2,3}, Lidia Torres^{1,c}, Pavol Valko⁶, and José Ángel Villar^{2,3}

¹ Institut d'Astrophysique Spatiale, Bâtiment 121, Université Paris-Sud 11 and CNRS (UMR 8617), 91405 Orsay Cedex, France

² Grupo de Física Nuclear y Astropartículas, Universidad de Zaragoza, C/Pedro Cerbuna 12, 50009 Zaragoza, Spain

³ Laboratorio Subterráneo de Canfranc, Paseo de los Ayerbe s.n., 22880 Canfranc Estación, Huesca, Spain

⁴ Department of Physics & Center for Nuclear Physics, University of Lisbon, Av. Prof. Gama Pinto 2, 1649-003 Lisbon, Portugal

⁵ Fundación ARAID, C/ María de Luna 11, Edificio CEEI Aragón, 50018 Zaragoza, Spain

⁶ Department of Physics, Slovak Technical University, Ilkovicova 3, 812 19 Bratislava, Slovak Republic

Abstract. Despite that it is widely used as a scintillator at room temperature, the hygroscopicity of NaI complicates its handling and limits its application for many purposes, for example as a cryogenic detector. To overcome this problem we study coating materials that can act as humidity barriers, in particular parylene, a polymer that can be deposited in very radiopure, thin and conformal layers. In this work, several NaI(Tl) samples coated with 2-5 μm parylene-C were tested at low temperature. Luminescence spectra under X-ray excitation are presented at several temperatures as well as the light output vs temperature at 1.5-300 K. Several thermoluminescence peaks were observed at around 60, 95 and 150 K during warm up to room temperature. The mechanical resistance of the coating under thermal cycles was also investigated, and we observed a degradation of the optical appearance and the light output after cooling down to about 100 mK, which compromises the reusability of the samples.

1 Introduction

NaI (pure or doped with Tl) are well known scintillators at room and low temperature. They are widely used as detectors for ionizing radiation and in particular they constitute an excellent target for dark matter searches by direct detection, having been used in a number of past and present experiments [1–5]. Nevertheless the high hygroscopicity of this material complicates its handling, especially for low temperature applications, and is one of the reasons why it has not been tested yet as a bolometer, despite its appealing features (see [6] for a discussion about the interest of this target for a bolometric dark matter search). A plausible solution could be to coat the NaI crystals with an appropriate material acting as humidity barrier, being parylene a good candidate.

2 Parylene coating

Parylene stands for a polymer family widely used in the electronic and space industry as moisture and dielectric barrier. It can be deposited in very thin films by vapor-phase condensation polymerization, forming a very homogeneous layer completely conformal to the exposed surfaces. Very thin layers are required to preserve a good op-

tical transmission (see Fig. 1) and to minimize heat capacity, requirements for the building of a good-performance scintillating bolometer. However, the barrier properties of the layer depend on its thickness, and at least several microns are required to allow day scale handling.

The parylene-C coating of the cylindrical samples used in this work was carried out by Comelec¹. Two depositions were needed to completely cover the crystals surface, resulting in $\sim 2 \mu\text{m}$ thickness in the top and bottom faces and $\sim 5 \mu\text{m}$ thickness in the lateral surface. As concerns the radiopurity, starting dimer samples were provided by the company for a HPGe measurement at the Canfranc Underground Laboratory. No contaminations were found for the most common isotopes and only upper limits were determined except for ²³²Th, for which an activity level of $26 \pm 8 \text{ mBq/kg}$ was calculated. Nevertheless, cleaner parylene films are available (see for example [8]).

3 Low temperature X-ray scintillation measurements

The X-ray scintillation measurements were performed at the Institut d'Astrophysique Spatiale (IAS, Orsay). For this study we used one NaI(Tl) cylindrical crystal (25 mm height and 25 mm diameter) with optically polished sur-

^aPresent address: CENPA, University of Washington, US

^be-mail: mariam@unizar.es

^cPresent address: Service d'Astrophysique, CEA Saclay, France.

¹Comelec S.A. (Switzerland) www.comelec.ch

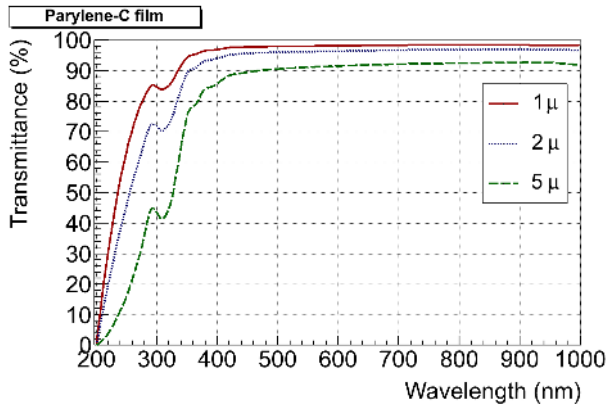


Figure 1. Transmittance (in %) of parylene C films of several thicknesses. Values deduced from [7].

faces provided by DetectEurope² and coated with parylene as described in section 2. For the excitation we used a X-ray BULLET[®] 40 kV source. The sample was mounted in an silver-coated reflecting cavity installed inside a ⁴He cryostat, and an Avantes optical fiber transmitted the light to the optical detector. More experimental details can be found in [6].

3.1 Light yield vs temperature

As light detector we used a Si photodiode HAMAMATSU S1336-18BQ, whose detection range spans from 190 to 1100 nm. The cooling stage was quite fast (around 20 minutes from 300 K to 77 K and 6 minutes from 77 K to 4 K), while the light output dependence with temperature was measured during two warming cycles (from 4 K to 200 K and from 1.5 K to 77 K). Figure 2 shows the results (solid curve), while the open circles represent some points taken during the cooling stage, in good agreement with the warming stage data. The maximum emission was found at around 125 K, followed by a noticeable light decrease below 70 K and a further increment of light below 30 K. The general behaviour is similar to that found by Sailer et al. in [9] for a non-coated sample under α excitation, although the relative intensities and temperatures of the maxima and minima of light emission differ.

3.2 Spectral response

At three stable temperatures (77, 4 and 1.5 K) the spectral emission intensity was measured with an Avantes fiber spectrometer (detector Sony 2048-DUV, diffraction grating VA, 300 lines/mm, detection range: 250-1100 nm). It was necessary to correct experimental data both by the detection efficiency and the cavity transmission dependence with wavelength. The first one was calculated experimentally as the ratio of the known spectra of two commercial sources to the measured ones. The result compares well with the manufacturer specifications³ (see Fig. 3), the biggest differences being below 450 nm, where fiber effects are important. The second one was estimated by

²Detect-Europe, allée de Kerpayen, 56000 Vannes, France

³www.avantes.com

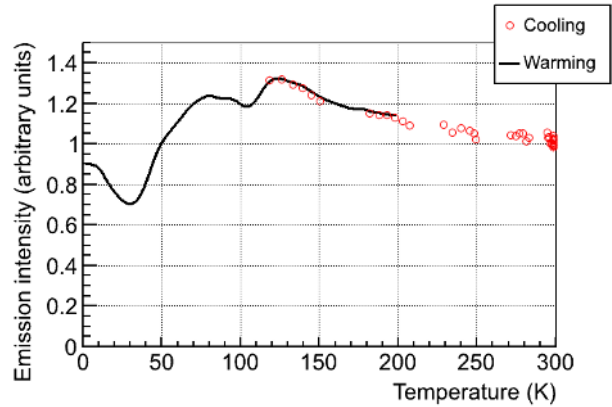


Figure 2. Emission intensity vs temperature of a parylene-coated NaI(Tl) sample under X-ray excitation normalized to the light output measured at 300 K. The solid line represents the integral measurement performed with a Si photodiode during warming up to room temperature, while single points were acquired during the cooling stage (open circles).

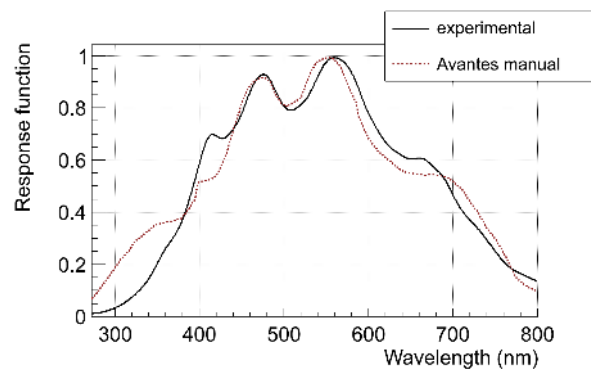


Figure 3. Solid line: detector response function measured for a Sony 2048-DUV (diffraction grating VA, 300 lines/mm) and 3 m Avantes FC-UV 1000 optical fiber. Dotted line: expected sensitivity from Avantes manual. Both curves have been normalized to 1 at the wavelength corresponding to the maximum.

Monte Carlo simulation using Geant4 [10]. The wavelength dependence of parylene transmission and silver reflectivity were taken from [7] and [11] respectively. The transmission efficiency was estimated by following a large number of optical photons and counting those arriving to the optical fiber with the correct angle for transmission (see Fig. 4). A comparison with a simple analytical calculation is also shown in the figure. The calculation follows the approach sketched in [12], but including an absorption factor in the crystal (η), resulting in the following expression for the cavity transmission efficiency:

$$\epsilon_c(\lambda) = \eta x + \frac{R\eta\varphi}{1 - R\eta(1 - \varphi)}\eta(1 - x) \quad (1)$$

where x is the fraction of direct light into the fiber, R is the Ag reflectivity and φ is the probability for a photon of reaching the fiber with the correct angle for transmission. Figures 5 and 6 show the measured spectra at the three stable temperatures before and after applying detec-

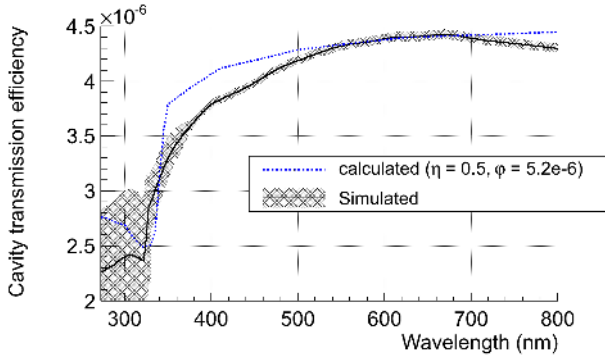


Figure 4. Solid line: cavity transmission efficiency estimated from Geant4 MC simulation. The shadowed area represents the systematic error introduced by a variation of $\pm 1 \mu\text{m}$ in the parylene thickness. Dashed line: analytical calculation of the transmission efficiency (see text for details).

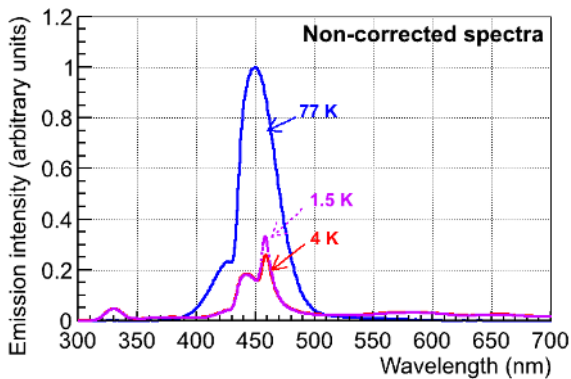


Figure 5. Luminescence spectra of a parylene-coated NaI(Tl) sample at 77, 4 and 1.5 K before applying cavity and detector corrections.

tor and cavity corrections. The effect of systematic errors in the detector and cavity correction, represented by the shadowed area in Fig. 6, is important only below 350 nm. As expected, at 77 K the main emission band is around 440 nm, but a weak emission at 325 nm was also observed at this temperature whose intensity increased steadily while lowering the temperature till becoming the dominant component at 4 K, although the relative intensity between 325 and 440 nm components is strongly affected by systematics previously explained. The spectra of Fig. 6 clearly points out a correlation between the decrease of the light yield below 70 K and the increase below 30 K with the respective decrease and increase of the 440 and 325 nm components. It is worth noting that in the measured spectrum we cannot disentangle the contribution of the parylene itself, whose luminescence could be excited by the UV NaI emission [13].

4 Thermoluminescence

The same experimental setup was used for a thermoluminescence measurement by programming the X-ray source

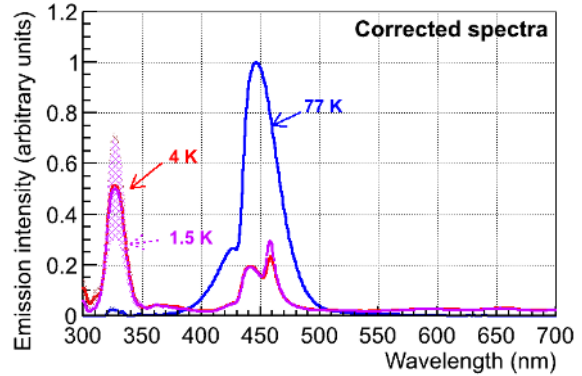


Figure 6. Luminescence spectra of a parylene-coated NaI(Tl) sample at 77, 4 and 1.5 K after applying cavity and detector corrections. Shadowed area represents systematic errors in the corrections (only noticeable below 350 nm).

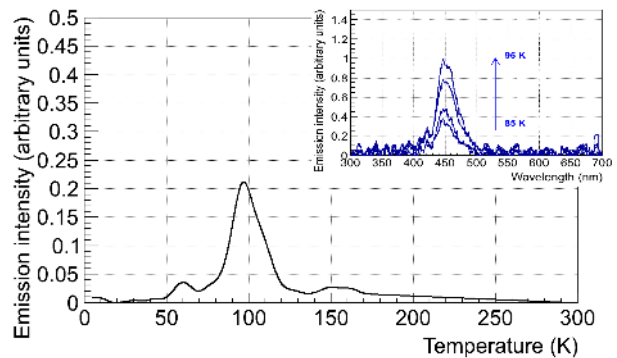


Figure 7. Light output vs temperature of a parylene-coated NaI(Tl) sample recorded during the OFF position of the X-ray recorder, at warming. Three thermoluminescence peaks (60, 95 and 150 K) are observed. In the inset, luminescence spectra corresponding to the main thermoluminescence peak measured at regular time intervals at warming after an initial irradiation at 77 K.

to produce 2 minutes bursts. The light output registered with the Si diode when the source was OFF is displayed in Fig. 7, showing a clear thermoluminescence peak at 95 K and two smaller ones at 60 and 150 K. The emission spectrum of the main peak was also acquired (Fig. 7 inset). Despite the very weak emission, that forced us to use very long integration times and resulted in rather noisy spectra, a clear line centered at 450 nm is observed.

5 Response after thermal cycles

A study of the mechanical resistance and the optical quality after thermal cycles was performed at the University of Zaragoza with two parylene-coated NaI samples (a pure and a thallium-doped one) of the same shape, dimensions and parylene coating thicknesses than the one described in section 3. The light output of the crystals was measured before and after the thermal cycle using a photomultiplier (HAMAMATSU R6233, spectral range: 200-650 nm). The samples were wrapped with Teflon[®] and in-

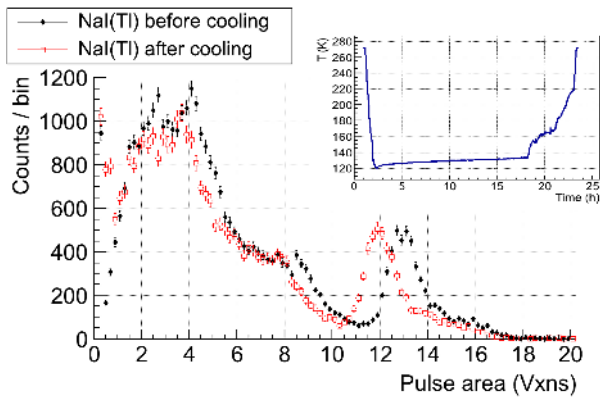


Figure 8. Pulse area spectra for a parylene-coated NaI(Tl) sample before and after cooling down to 100 K under ^{137}Cs excitation. The temperature cycle is shown in the inset.

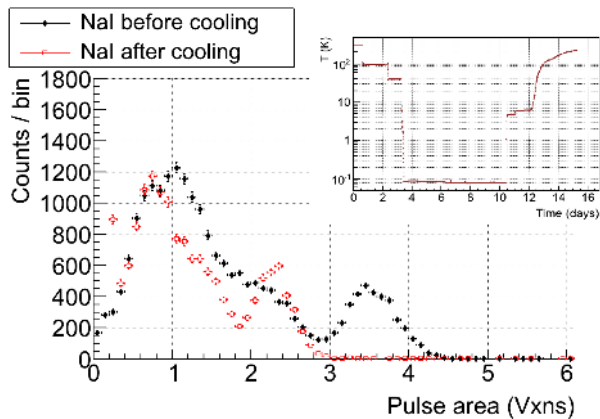


Figure 9. Pulse area spectra for a parylene-coated NaI sample before and after cooling down to ~ 80 mK under ^{137}Cs excitation. The temperature cycle is shown in the inset.

serted in a polyethylene holder that was mounted inside an aluminium cavity facing the PMT. No optical grease was used, losing part of the signal but gaining in reproducibility. The PMT pulses were digitized with a Tektronix scope (1.25 GS/s) and the energy of the pulses was estimated by software integration. The scintillation was excited with 662 keV γ from a ^{137}Cs source.

The doped sample was cooled down to 100 K by connecting the copper crystal holder to a cold finger immersed in a liquid nitrogen bath. A slight decrease in light output of 7% was measured (see Fig. 8). No deterioration was observed in the surface by visual inspection, but the parylene adherence seemed to be lost in some areas. The pure crystal was cooled to mK temperatures by installing it in the mixing chamber flange of a dilution refrigerator. The mounting stage was longer in this case and the crystal was under 30-50% RH conditions during one week. A base temperature of ~ 80 mK was maintained for one week, and then we let it slowly warm up to room temperature. The spectrum taken after this thermal cycle revealed an important decrease of light (around 35%, see Fig. 9), with the surface showing some white areas.

6 Conclusions

Coating NaI (pure or Tl-doped) crystals with an appropriate material facilitate its handling in normal RH conditions and can simplify considerably the design for low temperature applications. We have measured the light output of a parylene-coated NaI(Tl) crystal under X-ray excitation, observing the maximum emission at ~ 125 K followed by a strong decrease, but a further increment in light has been measured below 30 K, that seems to be due to a new emission line at 325 nm. Thermoluminescence has been observed at 60, 95 and 150 K. We have studied the mechanical resistance of the coating under thermal cycles, observing a degradation of the optical appearance and a reduction of the light output after cooling down to about 100 mK, which compromises the reusability of the samples.

Acknowledgements

This work was supported by the Spanish Ministerio de Economía y Competitividad, the European Regional Development Fund (MINECO-FEDER, FPA2011-23749), the French CNRS/INSU (MANOLIA/BOLERO), the grant PDTC/FIS/83424/2006 of the Portuguese Foundation for Science and Technology (FCT), the Nuclear Physics Center of the University of Lisbon, and the Consolider-Ingenio 2010 Programme (MULTIDARK CSD2009-00064). C. Ginestra and L. Torres are supported by MICINN FPI and postdoctoral grants respectively.

References

- [1] R. Bernabei et al., *Eur. Phys. J. C* **67**, 39 (2010)
- [2] S. Cebrián et al., *Astrop. Phys.* **37**, 60 (2012)
- [3] G.J. Alner et al., *Phys. Lett. B* **616**, 17 (2005)
- [4] K. Fushimi et al., *Astrop. Phys.* **12**, 185 (1999)
- [5] M.L. Sarsa et al., *Phys. Rev. D* **56**, 1586 (1997)
- [6] N. Coron et al., *Astrop. Phys.* **47**, 31 (2013)
- [7] Y.S. Jeong et al., *Synthetic Metals* **127**, 189 (2002)
- [8] J. Loach, Talk at Workshop Low Radioactivity Techniques (LRT2010), Aug 28-29 2010, Sudbury (Canada)
- [9] C. Sailer et al., *Eur. Phys. J. C* **72**, 2061 (2012)
- [10] S. Agostinelli, et al., *Nucl. Instrum. Meth. A* **506**, 250 (2003)
- [11] M. Bass, *Handbook of Optics*, 2nd ed. (1995)
- [12] Y. Ortigoza et al., *Astrop. Phys.* **34**, 603 (2011)
- [13] B. Lu et al., *Proc. 15th Int. Conf. on Solid-State Sensors, Actuators and Microsystems (Transducers09)*, 176 (2009)