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Gammastic: towards a Pseudo-gamma Spectrometry in Plastic Scintillators

Matthieu Hamel,* Chrystèle Dehé-Pittance, Romain Coulon, Frédérick Carrel, Philippe Pillot, Éric Barat,* Thomas Dautremer, Thierry Montagu, Stéphane Normand

Abstract—War against CBRN-E threats needs to continuously develop sensors with improved detection efficiency. More particularly, this topic concerns the NR controls for homeland security. A first analysis requires indeed a fast gamma spectrometry so as to detect potential special nuclear materials (SNM). To this aim, plastic scintillators could represent the best alternative for the production of large-scale, low-cost radiation portal monitors to be deployed on boarders, tolls, etc. Although they are known to be highly sensitive to gamma rays, due to their poor resolution, information relative to the nature of the SNM is tricky. Thus, only the Compton edge is obtained after interaction, and no information of the photoelectric peak is observed.

This project concerns new developments on a possible pseudo-gamma spectrometry performed with plastic scintillators.

This project is articulated on a combination of two developments:

- The design of new materials most suitable for recovering the photoelectric peak after gamma interaction with the scintillator. This work concerns mainly plastic scintillators loading with heavy elements, such as lead or bismuth.
- The analysis of the resulting signal with smart algorithms.

This work is thus a pluridisciplinary work performed at CEA LIST and embeds 4 main disciplines: MCNPX simulations (simulated spectra), chemistry of materials (preparation of various plastic scintillators with different properties), instrumentation (lab experiments) and smart algorithms. Really impressive results were obtained with the unfolding of simulated spectra at various energies (from ^{241}Am to ^{60}Co) and an innovative approach was proposed to counter-balance the quenching effect of luminescence by heavy elements in plastic scintillators.

Index Terms— Inference algorithms, Scintillation counters, Nuclear measurements.

I. INTRODUCTION

PROTECTION OF civilians and facilities against CBRN-E (Chemical, Biological, Radiological, Nuclear, and Explosives) threats represents a true challenge due to the constant increase of world's population movements. According to Dr. El Baradei (IAEA director), terrorists who

are unconcerned about exposing themselves to radiation could easily conceal a source in a truck or a suitcase. “*The danger of handling powerful radioactive sources can no longer be seen as an effective deterrent, which dramatically changes previous assumptions. [...] Security of nuclear and other radioactive material has taken on dramatically heightened [in IAEA’s work] significance in recent years.*”

In this context, the Gammastic project aims at developing new solutions to perform a pseudo-gamma spectrometry with plastic scintillators. Due to their low density and effective atomic number, plastic scintillators suffer from the absence of photoelectric peak, and are thus not suitable for a fast detection of Special Nuclear Materials. We describe herein an optimized detector based on the development of new materials (lead- and bismuth-loaded plastic scintillators) and smart algorithms.

II. CHEMISTRY OF MATERIALS

A. Literature Precedents

A rough approximation of the photoelectric effect probability is given by $P_{pe} \approx \text{constant} \times Z_{\text{eff}}^n / (E_\gamma^{3.5})$, where the exponent n varies from 4 to 5. To increase both density and effective atomic number (Z_{eff}), chemists have turned their attention in doping plastic scintillators with heavy elements, without reducing the scintillation properties of the detector.

As early as 1953, Pichat *et al.* reported for the first time the preparation of bismuth- and lead-loaded plastic scintillators, at concentrations up to 9 wt% on small samples (\varnothing 18 mm, h 20 mm) [1]. In the following years, various metals from the IIB, IVA & VA groups (Hg, At, Sb, Bi, Sn, Pb) were incorporated as organometallics into plastic scintillator, with moderate success for the three formers [2-4]. The first ^{137}Cs photoelectric peak was observed by Dannin with highly tin-loaded plastic scintillators [5].

Recently, loading with heavy elements have regained attention. Good energy resolution on the detection of the photoelectric peak for ^{137}Cs were also obtained by using a combination of polyvinylcarbazole, an iridium complex as the fluorescent probe and triphenylbismuth [6, 7] or gadolinium nanocomposites in PVT [8], but on small samples only ($\leq 1 \text{ cm}^3$). In particular, some of us patented the use of cross-linked organolead compounds and their application to the detection of low energy x-rays (10-40 keV) for the Laser Méga Joule facility [9, 10].

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B. Results and Discussion

These last scintillators were designed to present a maximum of scintillation wavelength close to 600 nm. Based on this work, we prepared several plastic scintillators presenting these general properties:

- Bismuth or Lead loading, with a concentration range from 1 to 12 wt%,
- Scintillation wavelength between 400 and 500 nm,
- Morphology \varnothing 5 cm \times h 3-4 cm,
- Organometallics either dispersed into the polymer or cross-linked to the matrix.

Cross-linking of Bismuth organometallics will be discussed in a future paper. Only results of plastic scintillators incorporating triphenylbismuth will be presented herein.

As in [10], we decided to use lead dimethacrylate as the lead source. With the two methacrylate moieties, this chemical is able to be polymerized along with the other monomers so as to be incorporated directly into the polymer matrix, as it is represented in the Fig. 1. Some parallel experiments with other lead organometallics which do not contain a polymerizable moiety did not afford sufficient optical transparency with such sample morphology. To afford the best scintillation yield, several primary (300 \rightarrow 370 nm wavelength shifting) and secondary (370 \rightarrow 420 nm wavelength shifting) were tested. Thus, biphenyl, 2,5-diphenyloxazole, *p*-terphenyl, naphthalene and butyl-PBD were tested as primary fluorophore, and POPOP and bis-MSB were used as wavelength shifters. To access to 450 or even 500 nm emitting scintillators, perylene and 4-butylamino-*N*-(2',5'-di-*t*-butylphenyl)-1,4-naphthalimide, respectively, could be added to the mixture.

All these scintillators were classified from their relative scintillation yield when exposed to gamma sources: ^{241}Am , ^{137}Cs and ^{60}Co (respective activities, 204, 155, and 250 kBq). An Eljen Technologies EJ-200 plastic scintillator with the same dimensions displaying 10,000 ph/MeV was used as a standard for the energy calibration of the spectra. Typically, in a dark box, the sample was interfaced with optical grease to a Hamamatsu H1949-51 photomultiplier, performing between 1800 and 2000 V. The physical signal was pre-amplified, amplified with a Canberra Amplifier timing filter. Data were collected with a PCI EG&G multi channels analyzer and then analyzed with Maestro software. Data acquisition time was generally 900 s.

The concentration of each dye had dramatic influence on the scintillation yield. Some dyes were not compatibles with the reaction conditions for the preparation of the lead-loaded plastic scintillators and generally lead to yellowish, thus non-exploitable materials.

Fig. 2 & 3 show the results of the pulse height spectra of lead and bismuth loaded plastic scintillators, respectively, when exposed to a ^{137}Cs source. From EJ-200 data sheet, this scintillator is assumed to deliver ca. 10,000 ph/MeV. Thus, by comparing the Compton edges, we can determine the relative scintillation yield for our scintillators. The results are given in Table I. Lead-loaded plastic scintillators displayed moderate light yields, close to 4,000 ph/MeV for 1 to 2 wt% of metal. The light yields are even lower than commercial EJ-256-5,

loaded with 5 wt% of lead, which displays 5,200 ph/MeV.

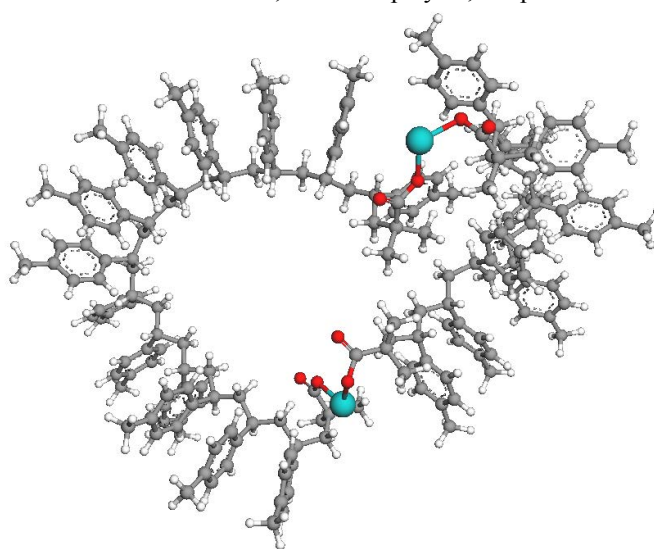


Fig. 1. Computing view of lead(II) methacrylate copolymerized with the other monomers inside the polymer matrix. The lead atoms are in blue, oxygens in red, carbons in grey and hydrogens in white.

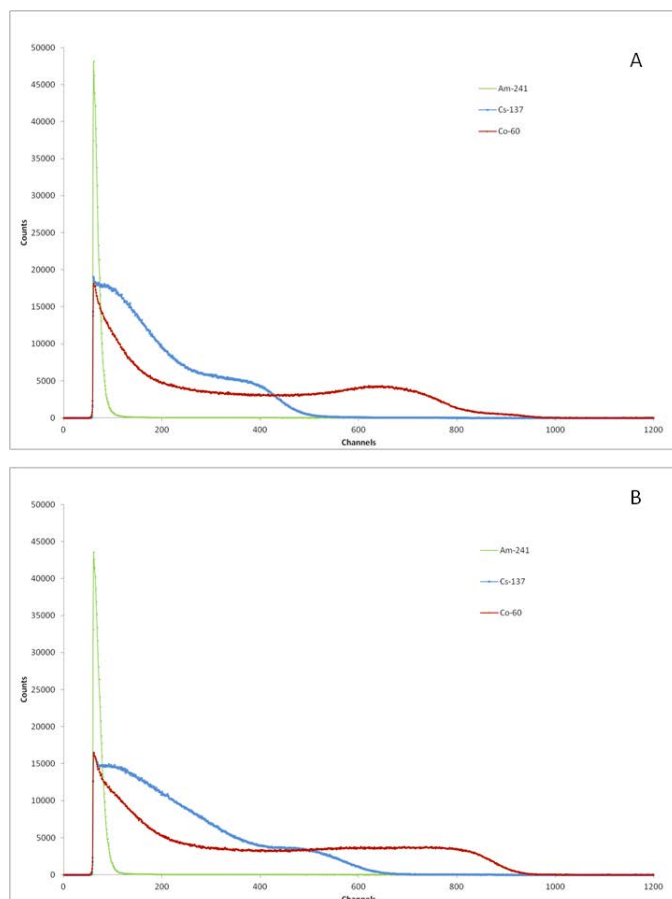


Fig. 2. ^{241}Am , ^{137}Cs and ^{60}Co pulse height spectra of two lead-loaded plastic scintillators (common parameters: 4.9 wt% Pb, diam 4.5 cm, height 3.5 cm) where the first fluorophore concentration has been increased from A (top) to B (bottom) two times.

III. ALGORITHMS

A. Literature Precedents

The “quality” of a spectrum is typically attributed to the resolution of a given peak. For instance, HPGe (High Purity Germanium) is for sure the industrial reference with a resolution $\rho(E) = \text{FWHM}(E)/E \approx 0.15\%$ at 1332 keV. Under such resolution conditions, one can focus only on the photoelectric peak and therefore discard any further information given in other parts of the spectrum. To obtain the information about the quantity of radionuclides, unfolding of the spectrum is thus made possible by using algorithms working with or without [11] a database.

Plastic scintillators cannot display such good resolution, even for low-energy gamma rays, since their response do not display any photoelectric peak. At a given energy, the energetic signature of the detector has therefore to be considered globally, as a response function. With that kind of materials, physics associated to gamma/matter interaction is so complex that any attempt to describe it with an analytical model seems illusory. Moreover, the energy response does not usually display a signature particularly localized in energy. In other words, we can say that from two distinct spectra given in entry, the measured spectra could be very similar. This also means that from a measured spectrum, the inversion process can lead to infinity of solutions for the spectrum in entry. It is therefore necessary to add supplementary information, so-called regularization, so as to reach a satisfactory result.

To be concise, the softer and little-informative the detector response, the trickier the unfolding method. For this reason, one often resorts to spectra classification instead of deconvolution in this situation. As an example, for Radiation Portal Monitors (RPMs), several classification techniques, based on SVM or PCA have been recently proposed [12]. While allowing the detection of particular radionuclides (from a database built with few acquired spectra) or anomalous signature, these approaches fail to consider a full spectrometric analysis (e.g mixture of radionuclides). Algorithms based on full deconvolution of the measured spectrum can be considered as an alternative solution. Three main different deconvolution techniques can be used for gamma-ray spectrometry [13].

The Linear Regularization method (also called Phillips-Townmeyer method [14]) is based on minimizing a criterion combining both data adequacy term (under the form of quadratic errors) and penalty term from the sum of the squares of the gradient of the incident spectrum to be found. The latter term is aimed at regularizing the solutions by imposing a global softness on the entry spectra. This method is rather simple to introduce but limits the production of the peaks in the estimated incident spectra. The penalty term thus tends to erase all these types of solutions.

The Maximum Entropy Method (MEM) is a popular and rather general deconvoluting method [15], which uses entropy as a regularization criterion able to penalize the data bias quadratic term. The main advantage of MEM is the assurance to obtain positive solutions.

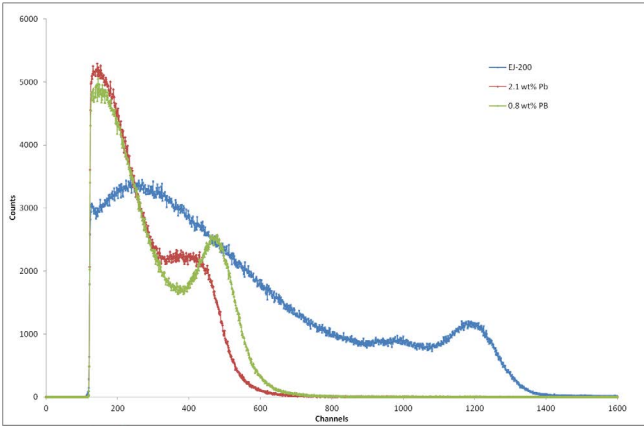


Fig. 3. ^{137}Cs pulse height spectra of two lead-loaded plastic scintillators at the concentrations 2.1 and 0.8 wt%, relative to one sample of EJ-200 of the same size.

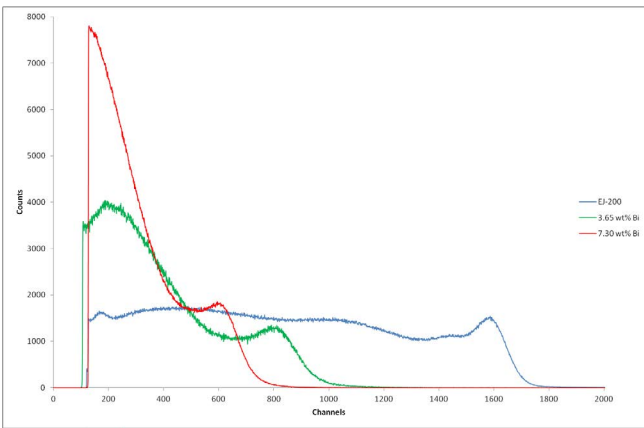


Fig. 4. ^{137}Cs pulse height spectra of two bismuth-loaded plastic scintillators at the concentrations 7.30 and 3.65 wt%, relative to one sample of EJ-200 of the same size.

Results with bismuth loading are much more interesting. Even at high concentrations (up to 7.3 wt%), the light yield remains higher than 4,000 ph/MeV. Future work will focus on the development and improvements of bismuth-loaded plastic scintillators.

According to Table I, despite the fact that both density and Z_{eff} were largely increased, we were not able to observe any photoelectric peak for ^{60}Co and ^{137}Cs . Current experiments are in progress so as to confirm it.

TABLE I
RELATIVE LIGHT YIELDS, DENSITIES AND EFFECTIVE ATOMIC NUMBERS OF
LOADED PLASTIC SCINTILLATORS RELATIVE TO EJ-200

Scintillator	Light yield (ph/MeV)	Density ($\text{g}\cdot\text{cm}^{-3}$)	Z_{eff}
EJ-200	10,000	1.023	≈ 5.7
0.8 wt% Pb	4,300	1.099	19.3
2.1 wt% Pb	3,900	1.099	25.1
3.65 wt% Bi	5,400	1.104	29.7
7.30 wt% Bi	4,100	1.186	36.3
EJ-256-5 (5 wt% Pb)	5,200	1.081	n.d.

Z_{eff} was simulated with X μ DAT software. As the composition of EJ-200 is proprietary, a rough estimation of Z_{eff} is given herein.

Introduced by Shepp and Vardi [16], the Maximum-Likelihood fitting by Expectation Maximization method (ML-EM) is widely spread in tomography. It is based on the expectation maximization (probability density of the observed spectrum for a given entry spectrum) which takes into account, contrary to LR and MEM, the Poisson distribution of spectral data. The EM aspect defines the iterative statistic skill allowing the maximization process. As for tomography, the ML-EM approach can be extended to a bayesian MAP-EM method (*Maximum a posteriori*) by introducing an *a priori* on the awaited spectrum. This *a priori* law governs thus the regularization term, the latter being originally absent from the ML-EM method.

Not to be exhaustive, it is noteworthy to precise that the work performed by Burt [17] which seems to be also the solution used in Symetrica portal monitors, relies on ML-EM approach.

In the next section, we present full deconvolution results obtained on ideal simulated spectra and based on two algorithms: a standard ML-EM algorithm and a new algorithm especially developed in the frame of the GAMMASTIC project.

B. Results and Discussion

Simulated spectra have been obtained using the MCNPX code [18, 19]. Three standard radioactive sources have been considered in these simulations, enabling to cover a large energy range: ^{241}Am (59.5 keV), ^{137}Cs (661.5 keV) and ^{60}Co (1173.2 keV and 1332.5 keV). The MCNPX Gaussian Energy Broadening (GEB) function has been used in order to take the resolution of the plastic scintillator into account. Detector dimensions are the following: 50 mm diameter, 28 mm thickness. Fig. 5 presents the simulated spectrum (top) and the result of a full deconvolution based on a standard ML-EM algorithm (bottom). One can notice that the main gamma-ray emissions can be clearly identified after deconvolution.

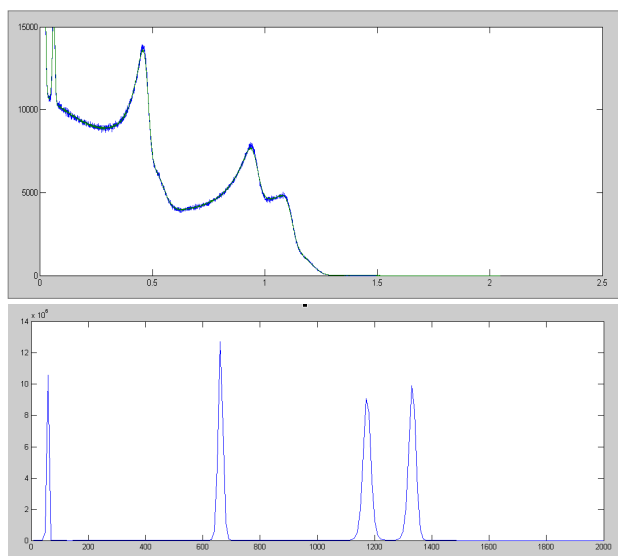


Fig. 5. ML-EM deconvolution: (top) observed spectrum, (bottom) deconvolution results.

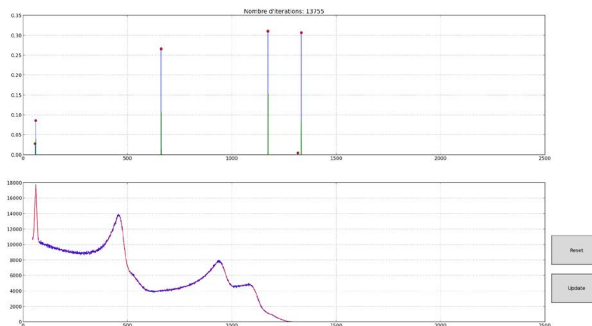


Fig. 6. Deconvolution results obtained with the new algorithm developed in the frame of the GAMMASTIC project: (bottom) observed spectrum, (top) deconvolution results.

Fig. 6 presents the same simulated spectrum but processed using a new algorithm developed in the frame of the GAMMASTIC project. One can notice a significant improvement of the resolution in the spectrum obtained after deconvolution.

IV. CONCLUSION

This work presents an overall solution to increase a plastic scintillator based detector to perform a pseudo-gamma spectrometry for the detection of SNM. Based on both optimization of the material and improvements of the deconvolution algorithms, preliminary results are very promising. Bismuth-loaded plastic scintillators display better relative scintillation yields compared with lead-loaded plastic scintillators. Future work will focus on increasing the metal concentration so as to be able to observe a photoelectric peak for ^{137}Cs . New algorithms were developed and successfully adapted to the deconvolution of simulated spectra. Work on experimental spectra is currently in progress. Ultimately, these two work packages will be combined.

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