

Scintillating fibre based beta spectrometer: Proof of concept by Monte-Carlo simulation and first experimental assessment

Nicolas Dufour, Adrien Sari, Guillaume H. V. Bertrand, Frédérick Carrel

► To cite this version:

Nicolas Dufour, Adrien Sari, Guillaume H. V. Bertrand, Frédérick Carrel. Scintillating fibre based beta spectrometer: Proof of concept by Monte-Carlo simulation and first experimental assessment. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2021, 1010, 165548 (7 p.). 10.1016/j.nima.2021.165548 . cea-04550635

HAL Id: cea-04550635 https://cea.hal.science/cea-04550635

Submitted on 17 Apr 2024 $\,$

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Scintillating fibre based beta spectrometer: proof of concept by Monte-Carlo simulation and first experimental assessment

N. Dufour*¹, A. Sari¹, G. H. V. Bertrand¹, F. Carrel¹

¹Université Paris-Saclay, CEA, List, F-91120 Palaiseau, France <u>*nicolas.dufour3@cea.fr</u>

Abstract

9 10

1

2 3

4

We developed a new beta measurement method based on scintillating fibres, able to discriminate incident beta 11 particles based on their energy, using various cladding thicknesses, which could find useful applications in the field of 12 nuclear decommissioning and dismantling (D&D). Indeed, thanks to Monte Carlo simulations, we found that high-13 14 energy beta particles were less affected by thicker cladding than medium-energy beta particles. Therefore, combining 15 different cladding thicknesses and a deconvolution method, we were able to identify and quantify convoluted beta 16 spectra in a simulated scenario. In this paper, we describe the Monte Carlo simulations combined with the 17 deconvolution algorithm based on the Maximum Likelihood – Expectation Maximization method (ML-EM), and the 18 associated experimental setup. We demonstrated the feasibility of such a method, using both simulated and experimental data, by deconvoluting ³⁶Cl and ⁹⁰Sr-⁹⁰Y spectra, considering ³⁶Cl as a medium-energy beta emitter and 19 20 ⁹⁰Sr-⁹⁰Y as a high-energy beta emitter. Using fibres with only one thickness of cladding, we found that we could not 21 estimate both contributions correctly, with errors reaching up to 71% of the actual activities. However, using two 22 different acquisitions of fibres each one with a different cladding thickness, we were able to estimate both 23 contributions with an error of 26% for ³⁶Cl and 10% for ⁹⁰Sr-⁹⁰Y. Study of the improvement of these performances is 24 presented and could help to fine-tune activities estimated. The next step of this work would consist in designing a beta 25 spectrometer, based on scintillating fibres, and assess its performances. Taking advantage of both deformability 26 capacity of scintillating fibres and their potential lengths up to a few meters, we expect that such a detection system 27 will be well adapted to the radiological characterization of large surfaces, and suitable for deployment on uneven 28 surfaces.

29 30 I. Introc

31

21

I. Introduction

32 Once a nuclear facility has ceased its activity, nuclear decommissioning and dismantling (D&D) ensures that all contamination is removed and treated accordingly. Depending on the past operations conducted in the facility, 33 expected contamination hotspots can vary in terms of radionuclides present and associated activities. Different 34 particles can be emitted by a single radioactive spot, thus hampering the characterization of a particular signal. For 35 example, ⁹⁰Sr is a fission product, beta emitter only, found along with ¹³⁷Cs, beta and gamma emitter. On-site 36 characterization of ⁹⁰Sr contamination can be estimated using scaling factors between ⁹⁰Sr and ¹³⁷Cs [1], but this 37 38 methodology can only be applied where such scaling factors exist, such as in nuclear power plants [2] only under the 39 hypothesis that these scaling factors are applicable and correct. Moreover, this method cannot be applied in cases in 40 which the beta contamination is not linked with a gamma emitter. Different detectors and methodology have been 41 developed specifically to resolve this problem [3]–[6]. Since the emission spectrum of a beta particle is continuous, there are no characteristic figures to rely on in order to identify beta emitters. Thus, deterministic and statistical 42 analysis were developed in order to deconvolute beta spectra [7]-[9]. A new technical solution based on the usage of 43 plastic scintillating fibres (PSF) with various cladding thicknesses, and a deconvolution method using Maximum 44 45 Likelihood-Expectation Maximisation (ML-EM) [10], is presented in this paper. This new technical solution is 46 adapted to the radiological characterization of large surfaces, and able to fit uneven surfaces from the intrinsic 47 deformability of scintillating fibres.

PSF used in this study are composed of a core made in polystyrene, and a cladding primarily made in polymethymethacrylate (PMMA). Both materials are composed of elements characterized by low atomic numbers, carbon and hydrogen. Thus, an organic scintillating fibre is intrinsically less sensitive to gamma-rays than beta particles, reducing any parasite signal coming from gamma emitters. PSF can be manufactured with lengths up to a few meters, first from a polymerisation step and then a fiber-drawing step. Such methodology is described in more details in the literature [11]. Applications of PSF are various and range from particle tracking [12], the detection of radionuclides in drinking water [13], to high-energy calorimetry [14]. Using their deformability capabilities, PSF would enable the detector to adapt to the measured surface, reducing losses due to the air layer when measuring beta particles, and enabling a better radiological characterization of uneven surfaces.

57 In this paper, we study the feasibility of a beta spectrometer based on scintillating fibres. First, we will present the 58 principle of the separation of two convoluted beta signals based on Monte Carlo simulation, combined with the ML-59 EM algorithm. Then, we will present the experimental setup and the results, obtained using ³⁶Cl and ⁹⁰Sr-⁹⁰Y beta 60 sources, used to evaluate the methodology of using scintillating fibres with various cladding thicknesses, combined 61 with the ML-EM algorithm. This methodology showed an improvement of the deconvolution process, especially in the 62 determination of the high-energy beta emitter.

- 64 II. Monte Carlo simulation and radionuclide identification
- 66 A. Principle and simulation study

67 68 In the research and development of nuclear detectors, a simulation phase is classic as it allows an estimation of the 69 detector response, amongst other calculations, before any experimental measurements. As such, using the MCNP6.2 70 simulation code [15], we designed the geometry of a single polystyrene scintillating fibre and studied its response 71 when exposed to beta sources. The simulated fibre consists of a polystyrene core with a diameter of 250 µm, and two 72 cladding thicknesses were simulated: 7.5 µm and 500 µm, both in PMMA. Sources used were pure-beta emitters ⁹⁰Sr 73 and ⁹⁰Y, generated using the BetaShape software [16]. The mentioned radionuclides were chosen for their disparities in their beta emission spectrum: ⁹⁰Sr has a beta energy endpoint at 546 keV and an average beta energy of 196 keV, 74 while these values are 2280 keV and 926 keV respectively for ⁹⁰Y. We calculated the deposited energy spectra from 75 incident beta particles normalised per emitted particle, called the F8 tally in MCNP6.2. Without further considerations, 76 77 we can estimate that the F8 tally result is comparable to a detection efficiency; therefore, we will designate the result as a simulated detection efficiency. Results of the simulations are presented in figure 1, and figure 2 presents the 78 79 electronic stopping power of polystyrene.

80

63

- 81
- 82



Fig. 1. Simulated detection efficiency as a function of beta particle energy, simulated with MCNP6.2 for different beta radionuclides. One standard deviation uncertainties are shown.



Fig. 2. Electronic stopping power of polystyrene ($\rho = 1.05$). Data collected from ESTAR [17].

These results illustrate the impact of the cladding. More specifically, ⁹⁰Y data show the same peak centered around 88 89 40 keV, for both cladding thicknesses. A partial energy deposit, characterized by a constant energy deposition rate of 90 the high-energy beta particles can explain this peak. Indeed, the stopping power of polystyrene for electrons between 91 500 keV and 2 MeV is equal to 2 MeV/cm, as shown in figure 2. Calculation of the deposited energy in 250 µm of polystyrene crossed gives 50 keV, which is coherent with our simulation and shows that these particles do not deposit 92 93 all of their energy. Higher energy depositions are explained by electron paths greater than 250 µm, expected for any non-perpendicular incident vector or any diffusion inside the fibre. The limited impact of the cladding to the peak is 94 95 explained by a low energy deposition in the PMMA. Indeed, energy deposition in 7.5 µm and 500 µm of PMMA is 96 1 keV and 75 keV respectively, when the electron energy is comprised between 500 keV and 2 MeV. Thus, any 97 electron with an incident energy greater than 575 keV will induce the same stopping power in polystyrene whether 98 there is a 500 µm cladding or not. Therefore, high-energy beta emitters will yield the same peak whether the cladding 99 is 7.5 μm or 500 μm.

However, the decrease for the ⁹⁰Sr signal is significant, and is due to the energy deposition in the cladding being 100 101 more important. From figure 2, we can see that the polystyrene stopping power increases exponentially as the electron 102 energy decreases below 500 keV, this phenomenon being also found for PMMA. Since ⁹⁰Sr beta emission spectrum 103 ranges from 0 to 546 keV, we expect all electrons emitted to be affected by the thicker cladding. For example, the average ⁹⁰Sr beta particle of 196 keV will deposit 2 keV for 7.5 µm of PMMA, but will not go through 500 µm of 104 105 PMMA. The energy deposition for a cladding of 500 µm will change the electronic stopping power from 2.7 MeV/cm to 3.6 MeV/cm. Therefore, any expected spectrum at 7.5 µm cladding will be different from a 500 µm cladding 106 spectrum, combined with a shielding effect blocking low-energy beta particles. 107

We conclude that a contrast was achieved, with a low impact on the high-energy beta emitter, and a significant 108 109 decrease of the medium-energy beta emitter. Therefore, by using scintillating fibres with various cladding thicknesses, 110 we expect to be able to create a contrast between different beta distributions. By using more scintillating fibres with 111 different cladding thicknesses, we expect contrasts to be sufficient to identify and quantify beta signatures. However, 112 such a detection system would require the use of a deconvolution algorithm to interpret the measurements. In the 113 frame of this study, we chose the Maximum Likelihood – Expectation Maximisation (ML-EM). 114

115

116

B. Identification with ML-EM

117 The Maximum Likelihood – Expectation Maximisation (ML-EM) algorithm is a method used in matrix inversion problems. It is based on an iterative calculation of the maximum likelihood achievable between an input dataset and a 118 predetermined database, where the input dataset follows a Poisson distribution. The iterative nature of the ML-EM 119 approach ensures the convergence of the calculated estimation, but imposes the definition of a starting value and a 120 next sequence step equation. Mathematical development leading to the final equation for the ML-EM resolution has 121 122 already been detailed in the literature [10], [18], [19], and more specifically to gamma-ray spectra deconvolution [20]– 123 [22]. A general formula for the number of measured counts from a radioactive source can be written as: 124

$$Y = A_{source} \ I \ \varepsilon \ t \tag{1}$$

126 With Y the measured spectrum, A_{source} the source activity, I the emission intensity of the measured particle, ε the 127 detection efficiency and t the measurement time. The equation can be simplified using λ as the total number of 128 emitted particles by the source, over the measurement time t, factored by the emission intensity I:

129

$$\lambda = A_{source} I t \tag{2}$$

130

131 Considering each bin of the measured spectrum, Y can be seen as a vector of B values (for *Binning*), each value 132 describing one energy bin. In such case, there is a specific detection efficiency for each bin corresponding to the 133 particle energy, thus ε has at least one dimension. Moreover, if there are multiple sources tested, λ can be seen as a 134 matrix of S values (for *Sources*). Therefore, ε must possess another dimension corresponding to the detection 135 efficiency for each radionuclide. For example, if we have a sampling of 1024 values for each spectrum, the dimension 136 of Y will be [1024,1]. Moreover, if we evaluate the contribution of three possible beta emitters, dimension of ε will be 137 [1024, 3], representing the three detection efficiency spectra for each beta source. Then, the dimension of λ will be 138 [1,3].

139 We can rewrite equation 1 using matrix notations

140

$$[Y]_B = [\lambda]_S [\varepsilon]_{B,S} \tag{3}$$

141

142 The measured spectrum matrix $[Y]_B$ is obtained through measurements, $[\varepsilon]_{B,S}$ is the associated database of detection 143 efficiencies and $[\lambda]_S$ is the emission spectrum, which is the desired information from the deconvolution process. The 144 detection efficiency database $[\varepsilon]_{B,S}$ is an array composed of each detection efficiency corresponding to each 145 radioactive source evaluated. This database can be generated through Monte Carlo simulations or experimental data.

146 Let $\hat{\lambda}$ be the estimator of λ . The ML-EM expression for the methodology defines the following next sequence step 147 equation:

148

$$\hat{\lambda}_{j}^{(n+1)} = \frac{\hat{\lambda}_{j}^{(n)}}{\sum_{i=1}^{B} \varepsilon_{i,j}} \sum_{i=1}^{B} \frac{\varepsilon_{i,j} Y_{i}}{\sum_{k=1}^{S} \varepsilon_{i,k} \hat{\lambda}_{k}^{(n)}}$$
(4)

149 Where $\hat{\lambda}_{j}^{(n+1)}$ is the estimator for the emission spectrum. Since the ML-EM algorithm is iterative, $\hat{\lambda}_{j}^{(n+1)}$ is the 150 estimator at the n + 1 step, $\hat{\lambda}_{j}^{(n)}$ is the estimator at the *n* step, *i* represents the iterator linked to the binning dimension 151 and both *j* and *k* are iterators linked to the source dimension. The first value, at the n = 1 step, must be input. 152 Following [23], the first value can be the average value written as:

154

$$\hat{\lambda}_{j}^{(1)} = \frac{\sum_{i=1}^{B} Y_{i}}{\sum_{i=1}^{B} \sum_{j=1}^{S} \varepsilon_{i,j}}$$
(5)

We simulated the response of the scintillating fibre with other beta emission spectra: ¹⁴C, ⁹⁰Sr, ⁹⁰Y, ²¹⁰Bi, ²¹²Bi and 155 212 Pb, for the two cladding thicknesses previously mentioned: 7.5 μ m and 500 μ m, in the same fashion as the results 156 presented in figure 1. To do so, we used MCNP6.2 to calculate the simulated detection efficiencies for each source, 157 158 using the F8 tally. This dataset was then used to construct a database of responses, corresponding to the $\varepsilon_{i,i}$ matrix, by 159 merging all F8 tally results. The input spectrum Y_i consisted of the simulation of a measured spectrum of ⁹⁰Sr and ⁹⁰Y, 160 in secular equilibrium, with a total activity of 2000 Bq. To construct this simulated measured spectrum, we added both 90 Sr and 90 Y simulated detection efficiencies, and multiplied the result with the simulated activity of 2000 Bq. 161 162 Furthermore, to simulate the uncertainty associated with the counting statistics, each bin of the simulated measured 163 spectrum was varied using a Poisson law. We expect a perfect identification and quantification for an ML-EM 164 reconstructed activity of ⁹⁰Sr and ⁹⁰Y equal to 1000 Bq, and other radionuclides to 0 Bq. Table 1 presents the ML-EM 165 results, for 200 iterations. The number of iterations was chosen based on the convergence of the ML-EM output 166 values. 167

100						
	^{14}C	⁹⁰ Sr	⁹⁰ Y	²¹⁰ Bi	²¹² Bi	²¹² Pb
Input activity (Bq)	0	1000	1000	0	0	0
Reconstructed	<1	963	990	5	<1	19

activity (Bq)						
169	Table 1. Input and reconstructed activities using the ML-EM algorithm.					

Table 1. Input and reconstructed activities using the ML-LW argo

From these results, we validated the methodology of using scintillating fibres with various cladding thicknesses to create a discrimination between beta emission spectra. This enables the use of deconvolution methods allowing to identify and quantify beta emitters present in an unknown mixture. We note that the ML-EM algorithm has a threshold below which identification may not be certain, as it can be seen on the reconstructed activity of ²¹²Pb. From the validation of the concept using simulations, we developed a setup to assess the experimental feasibility of the methodology.

177

178 III. Experimental setup and acquisitions

179

181

180 A. Methods and materials

182 Following simulation results, we developed an experimental setup to evaluate the feasibility of the methodology 183 presented above. A PSF bundle was manufactured and is composed of one hundred BCF-10 scintillating fibres [24] mounted onto two photomultipliers tubes 9205SB [25] using homemade adaptors. We chose one hundred scintillating 184 fibres instead of a single one, similar to the simulation results presented above, because a single fibre was not enough 185 186 to obtain a measurable signal. Optical grease has been applied to the interface. A single fibre has a core diameter of 187 250 µm and a surrounding optical cladding of 7.5 µm-thick. Fibre core is made of polystyrene and cladding is made of 188 polymethyl methacrylate (PMMA). Voltage applied to the photomultiplier tubes (PMTs) was 850 V. Anode outputs 189 are linked to a CAEN DT5743 digitizer [26], and measurements were done in coincidence to reduce background 190 signal. The coincidence windows was taken with a value of 15 ns, corresponding to the temporal length of a pulse. 191 Experimental setup has been placed in a dark environment, inside a metallic box, preventing any visible light to 192 interfere. A photography of the experimental setup inside the box is shown in figure 3. Cladding thickness was 193 modified using layers of PTFE. Each layer of PTFE is 200 µm-thick. Up to four were used one on the top of each 194 other to obtain a total thickness of 800 µm as is shown in figure 4. One acquisition set was done without any layer.





196

197

198 199

200

Fig. 3. Photography of the experimental setup with scintillating fibres. The bundle of fibres is shown in the middle. The aluminium adapters to the PMts are also visible, the mechanical pressure between the fibres and PMts being exerted by the rubber bands. In the middle is the source holder. Both black cylinders, on the left and right handsides, are the PMts.



201 202 203

Fig. 4. Photography of the experimental setup with scintillating fibres and four PTFE layers stacked (total thickness of 800 μ m).

Table 2 sums the characteristics of the two beta sources we used, ³⁶Cl and ⁹⁰Sr-⁹⁰Y. They were chosen for the difference in energy distribution and endpoint. Data listed in table 2 was taken from the Nucléide LARA database [27].

 Chlorine-36
 Strontium-90 + Yttrium-90

 Activity
 6470 ± 320 Bq
 5170 ± 260 Bq

	Particles emitted	Beta ¹	Beta	
	Average beta energy	246 5 keV	195.9 keV (⁹⁰ Sr)	
	(keV)	240.3 KeV	933 keV (⁹⁰ Y)	
	Maximum beta energy	700.2 koV	546.2 keV (90 Sr)	
	(keV)	709.3 KeV	2281.5 keV (⁹⁰ Y)	
Table 2. List	of radioactive beta sources used	during our measurement campa	aign. Uncertainties given at one st	andard deviation.

208 209

¹We will consider ³⁶Cl as a pure beta emitter since the other decay line possible is of low intensity (β +, 1.9%). 210

211 B. Acquisition results – Single source measurement

212

The first set of measurements was done with a single source during each acquisition. Figure 5 presents experimental data for the naked fibre bundle and 800 μ m of PTFE, and figure 6 presents the summed detection efficiency of each spectrum, as a function of the PTFE thickness. For visualisation purposes, the detection efficiency scale is constant for both graphs in figure 5. Horizontal axis in figure 5 is shown in pC units, and corresponds to the charge of the digitized pulse. This is a standard output of the CAEN DT5743 digitizer and allows the construction of an acquisition histogram.



Fig. 5. Detection efficiency of the fibre bundle without any PTFE layer on the left, and with 800 μm of PTFE on the right, for the ³⁶Cl and ⁹⁰Sr-⁹⁰Y sources.



PTFE thickness (µm)
 Fig. 6. Summed detection efficiency as a function of the PTFE thickness, for the ³⁶Cl and ⁹⁰Sr-⁹⁰Y sources. One standard deviation uncertainties are shown.

220 221

222 223

Detection efficiency for the ³⁶Cl source tends towards zero as more and more layers of PTFE are added, with a 228 slower decrease from 0 µm to 200 µm compared with the thicker layers, visible in figure 6. ⁹⁰Sr-⁹⁰Y detection 229 efficiency does not follow the same trend. Indeed, detection efficiency increases until 600 µm, before decreasing. In 230 parallel to this phenomenon, we also note that the maximum pulse charge has also slightly increased. In figure 5 for 231 the naked bundle, ⁹⁰Sr-⁹⁰Y detection efficiency curve reaches zero at 7 pC, while it reaches 0 at 9 pC for 800 µm of 232 233 PTFE. We explain both of these effects from the increase of the trapping efficiency. Indeed, a naked BCF-10 fibre has a trapping efficiency of 3.44% [13], meaning that 96.56% of all generated scintillation photons are lost. Adding a 234 235 reflecting layer of PTFE increases the percentage of captured photons. A better trapping efficiency allows more photons to be detected, increasing both the detection efficiency and the maximum resulting charge. However, energy 236 loss due to thicker layers of PTFE will overlap the benefits of a better trapping efficiency, as can be seen for the 237 detection efficiency for ${}^{90}\text{Sr}$ - ${}^{90}\text{Y}$, decreasing after 600 µm of PTFE. Overall, for the ${}^{90}\text{Sr}$ - ${}^{90}\text{Y}$ source, we can hypothesize that only the most energetic part of the emission spectrum is 238

Overall, for the 90 Sr- 90 Y source, we can hypothesize that only the most energetic part of the emission spectrum is detected, thus the impact from the strontium part of the beta emission spectrum is negligible. Even though the trapping efficiency is maximised when adding PTFE, this increase does not compensate for the energy loss of 36 Cl beta particles crossing the cladding, for all thicknesses. Therefore, from these first experimental results, we demonstrate the feasibility of the multi-cladding PSF detector principle, to develop a beta counter able to discriminate incident beta particles in gross energy ranges.

245

In this section, we will evaluate the detector's response when two beta source are placed together, to check if we find any discrepancies with single source acquisitions. Moreover, this data will be used as the input data for testing ML-EM algorithm performances. In this set of measurements, the two beta sources used, ³⁶Cl and ⁹⁰Sr-⁹⁰Y, were measured simultaneously for the different thicknesses of PTFE. Figure 7 present the measurements results for both the naked bundle and with 800 μ m of PTFE, expressed in counting rate (s⁻¹). For visualisation purposes, the counting rate scale is constant for figure 7. Figure 8 shows the summed counting rate for each thickness of PTFE.



Fig. 7. Fibre bundle counting rate when both sources are present, with no PTFE layer on the left and with 800 µm of PTFE on the right.





255

The distribution endpoint can help identifying the most energetic beta emitter radionuclide, from the maximum charge read, but not identify precisely any other beta emitter. However, even in a lack of prior information point-ofview, we would be able to say that the measured sample contains at least one high-energy beta emitter, since an 800 µm-thick PTFE layer was not able to stop incident radiations, under the hypothesis that the signal is in fact due to beta particles.

When both sources are present, behaviours described for the single source measurements are not visible here. Indeed, figure 6 shows a constant increase of the summed detection efficiency up to $600 \mu m$ and a slight decrease at mono-sources measurements and dual sources measurements. Causes for this discrepancy between summed differences in geometrical efficiency, and an inhomogeneity of the PTFE layers, discussed in more detail in the spectrum identification section of this article.

From the highest contrast achieved with the thickest layer of PTFE, we expect the ML-EM code to correctly evaluate high-energy beta emitters, and then deduce medium-energy beta emitters present on the other acquisition spectra. As we have seen, single source acquisitions show some limitations concerning the representativity of single source measurements. To tackle this issue, we are studying possible improvements of the experimental setup, especially concerning the homogeneity of PTFE layers, and ensuring that they do not overlap or loosen. In order to obtain better results, new protocols with three or more sources are being studied and should provide information about limitations on the identification and quantification processes, in terms of number of sources present.

282

283 IV. Spectrum identification

284

285 286

A. Deconvolution results – Dual sources measurements with various cladding thicknesses

Using single source acquisitions as the ML-EM database, we tested the algorithm ability to separate the ³⁶Cl and ⁹⁰Sr-⁹⁰Y beta spectra, from the double source acquisitions. We chose two thicknesses for the database, 200 μ m and 800 μ m of PTFE, because of the high contrast between ³⁶Cl and ⁹⁰Sr-⁹⁰Y detection efficiency spectra (*cf.* figs. 4 and 5). Moreover, we empirically found better ML-EM reconstructions using these two spectra, and a comparatively worse reconstruction using any other combinations, including all acquisition spectra together. This behaviour will be discussed in the next section. One hundred iterations were enough for the convergence of ML-EM output data, and are presented in figure 9.

- 294
- 295



296

Fig. 9. Measured spectra and ML-EM reconstruction of the counting rate, for 200 μm of PTFE on the left and 800 μm on the
 right, when both ³⁶Cl and ⁹⁰Sr-⁹⁰Y sources are present.

300 No visual discrepancies are visible between experimental data and the ML-EM reconstruction. Actual sources 301 activities and the ML-EM reconstructed counterpart are presented in table 3. Percentage biases were calculated in 302 order to evaluate the difference between experimental and ML-EM reconstructed activities.

		Source activity	Reconstructed activity	Percentage biases
--	--	-----------------	------------------------	-------------------

Chlorine-36	6470 ± 324 Bq	4780 Bq	-26%
Strontium-Yttrium-90	5168 ± 258 Bq	5776 Bq	+12%

Table 3. Sources and ML-EM reconstructed activities for 200 µm and 800 µm spectra inputs.

Comparing data from table 3, we can see that the ³⁶Cl reconstructed activity does not match the experimental 306 value, with an underestimation over five standard deviations. However, ⁹⁰Sr-⁹⁰Y activity is correct under three 307 standard deviations. Discrepancies will be discussed by comparing obtained results with expected ones. 308

309

310 B. Deconvolution results – Dual sources measurements with a single cladding thickness

311

312 In order to validate the methodology, we tested the ML-EM reconstruction using only one acquisition spectrum. 313 Table 4 presents the ML-EM reconstructed activities using single spectrum databases, and their associated percentage

biases. Each ML-EM column corresponds to a database and measured spectrum couple, corresponding to 314 315 measurements done using the specified PTFE layer. For example, ML-EM $- 0 \mu m$ corresponds to the construction of a 316 database using single source measurements with no layers of PTFE, and the measured spectrum corresponds to the measurement of both sources together without PTFE. 317

318

	³⁶ C1	³⁶ Cl	90 c , 90 v	⁹⁰ Sr- ⁹⁰ Y
	CI	percentage bias	51- 1	percentage bias
Source activity	6470 ± 324 Bq	N/A	5168 ± 258 Bq	N/A
$ML-EM - 0 \mu m$	4136 Bq	-36%	2552 Bq	-51%
ML-EM – 200 µm	1852 Bq	-71%	7844 Bq	+52%
ML-EM – 400 µm	9313 Bq	+44%	2678 Bq	-48%
ML-EM – 600 µm	6962 Bq	+8%	2502 Bq	-52%
ML-EM – 800 µm	9348 Bq	+44%	5103 Bq	-1%

319

320

321 Comparing table 4 with table 3 percentage biases, we note that some single PTFE thickness configurations obtain 322 better results on a single beta emitter than with two PTFE thicknesses configurations. For example, 800 µm PTFE single spectrum obtains a percentage bias of 1% with the expected ⁹⁰Sr-⁹⁰Y activity, but has a percentage bias of 44% 323 with the expected ³⁶Cl activity. On the other hand, 600 µm PTFE single spectrum obtains a percentage bias of 8% with 324 the expected ³⁶Cl activity, but has a percentage bias of 52% with the expected ⁹⁰Sr-⁹⁰Y activity. From the percentage 325 bias values in table 3, 26% and 12% for the ³⁶Cl and ⁹⁰Sr-⁹⁰Y respectively, we conclude that the methodology 326 presented here constitutes the best compromise for the deconvolution of the two beta spectra. 327

328

329 C. Deconvolution results – Expected results

330

331 Considering the database, we calculated the expected spectra if the reconstructed activities were correct, *i.e.* equal to the experimental activities presented in table 3, for the setup with 200 and 800 µm of PTFE. Figure 10 presents the 332 333 expected reconstructed spectrum.



Table 4. Sources and ML-EM reconstructed activities, with associated percentage biases, for single spectrum databases.

Fig. 10. Measured spectra and expected ML-EM reconstruction of the counting rate for exact activities, for 200 μm of PTFE on
 the left and 800 μm on the right, when both ³⁶Cl and ⁹⁰Sr-⁹⁰Y sources are present.

338

339 Both figs. 11 and 12 are similar in shape and intensity without any major visual discrepancy. We do note that the 340 expected ML-EM reconstruction of figure 10 does not overlap completely the measured counting rate on the second 341 peak, while it is the case for figure 9. Average maximum value for both measured and reconstructed spectra for the second peak in figure 9 is 0.13 s⁻¹, while for figure 10, average maximum value is 0.11 s⁻¹ for the expected ML-EM 342 343 reconstruction: even when using the experimental activities, we obtain a slight difference between the experimental 344 spectra and reconstructed ones. We explain this difference from a lack of representativeness of the database, possibly 345 due to statistical variation at the maximum peak values. Another factor may be the solid angle differences being too 346 important between one source and two sources measurements. However, we do not expect significant differences, because sources were placed together along the fibres bundle, only shifting their position from the single source 347 348 location by a few centimetres. In addition, we note that PTFE layers were changed or readjusted between measurements for the database and dual sources acquisitions. PTFE thicknesses may not have been consistent and thus 349 350 created discrepancies. From this study, we conclude that the small variations may impede the correct reconstruction.

351

352 Conclusion and outlook

353

354 We recall that the objective of this paper was to develop a new methodology of beta identification and 355 quantification system using scintillating fibres, to tackle the issue of radiological characterization of large and possible 356 uneven surfaces. In this paper, we presented the principle of a discrimination method based on scintillating fibres with 357 various cladding thicknesses, for medium and high-energy beta emitters, enabling beta spectrometry using 358 deconvolution algorithms. We studied using Monte Carlo simulation the methodology principle by identifying and 359 quantifying beta signatures using the ML-EM algorithm. A database of multiple simulated beta signatures was 360 constructed and used effectively with satisfying deconvolution results. Then, an experimental setup was designed, and showed that the methodology can be used on a detection system. First results showed an improvement of the 361 362 identification and quantification of convoluted beta emitters when compared to single cladding scintillating fibers 363 presented in IV. B.

Further, we plan to acquire spectra with longer measurement time to construct a database with less statistical variation and check the prevalence of this issue on the representativity. Moreover, we are planning to manufacture specific bundles with preset PTFE layers. These improvements will help in developing a first prototype of beta spectrometer using Monte Carlo simulation and then assess its performances in laboratory. Two application layouts are studied: spectrometry using deconvolution algorithms like presented in this paper, and a counting mode, where identification is limited to the energy range of the beta emitter, for example, between medium and high-energy beta emitter. In this case, there would be no need for a deconvolution methodology. Studies with more than two beta sources present are planned, and will cover larger energy ranges. Studies of the actual detector sensitivity to gamma rays is currently underway. Finally, study of the counting rate evolution as a function of the detector deformability is also planned.

374

376 References

- W. Irrek, K. Müller, D. Fouquet, and A. P. Froggatt, "Comparison among different decommissioning funds methodologies for nuclear installations," 2007.
- Radiological Characterization of Shut Down Nuclear Reactors for Decommissioning Purposes, no. 389.
 Vienna: International Atomic Energy Agency, 1998.
- A. R. Lang *et al.*, "Cesium and Strontium Contamination of Nuclear Plant Stainless Steel: Implications for
 Decommissioning and Waste Minimization," *ACS Omega*, vol. 4, no. 11, pp. 14420–14429, Sep. 2019.
- G. Turkington, K. A. A. Gamage, and J. Graham, "Beta detection of strontium-90 and the potential for direct in situ beta detection for nuclear decommissioning applications," *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 911, pp. 55–65, Dec. 2018.
- A. Leskinen, P. Fichet, M. Siitari-Kauppi, and F. Goutelard, "Digital autoradiography (DA) in quantification of trace level beta emitters on concrete," *J. Radioanal. Nucl. Chem.*, vol. 298, no. 1, pp. 153–161, Oct. 2013.
- P. Fichet, A. Leskinen, S. Guegan, and F. Goutelard, "Characterization of Beta Emitters for Decommissioning," in Volume 2: Facility Decontamination and Decommissioning; Environmental Remediation; Environmental Management/Public Involvement/Crosscutting Issues/Global Partnering, 2013.
- A. Grau Malonda, L. Rodriguez Barquero, and A. Grau Carles, "Radioactivity determination of 90Y, 90Sr and 89Sr mixtures by spectral deconvolution," *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 339, no. 1–2, pp. 31–37, Jan. 1994.
- T. M. Semkow, "Statistical deconvolution of beta spectra: Implications for neutrino mass detection," *Appl. Radiat. Isot.*, vol. 46, no. 5, pp. 341–349, May 1995.
- R. Remetti and A. Sessa, "Beta spectra deconvolution for liquid scintillation counting," *J. Radioanal. Nucl. Chem.*, vol. 287, no. 1, pp. 107–111, Jan. 2011.
- [10] L. A. Shepp and Y. Vardi, "Maximum Likelihood Reconstruction for Emission Tomography," *IEEE Trans. Med. Imaging*, vol. 1, no. 2, pp. 113–122, Oct. 1982.
- P. Rebourgeard *et al.*, "Fabrication and measurements of plastic scintillating fibers," *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 427, no. 3, pp. 543–567, May 1999.
- 406 [12] R. E. Ansorge *et al.*, "The UA2 scintillating fibre detector," *Nucl. Inst. Methods Phys. Res. A*, vol. 273, no.
 407 2–3, pp. 826–832, 1988.
- T. Bernard *et al.*, "SAFEWATER Innovative Tools for the Detection and Mitigation of CBRN Related
 Contamination Events of Drinking Water," *Proceedia Eng.*, vol. 119, pp. 352–359, 2015.
- 410 [14] H. Burmeister, "Electromagnetic calorimetry using scintillating plastic fibres," pp. 530–533, 1984.
- 411 [15] C. J. Werner et al., "MCNP Version 6.2 Release Notes," Los Alamos, NM (United States), Feb. 2018.
- 412 [16] X. Mougeot, "Reliability of usual assumptions in the calculation of β and v spectra," *Phys. Rev. C*, vol. 91, no. 5, p. 055504, May 2015.
- 414 [17] M. J. Berger, J. . Coursey, M. . Zucker, and J. Chang, "ESTAR, PSTAR, and ASTAR: Computer Programs 415 for Calculating Stopping-Power and Range Tables for Electrons, Protons, and Helium Ions (version 1.2.3)," 416 *National Institute of Standards and Technology, Gaithersburg, MD*. [Online]. Available: 417 http://physics.nist.gov/Star. [Accessed: 04-Jul-2021].
- 418 [18] M. R. Gupta, "Theory and Use of the EM Algorithm," *Found. Trends*® *Signal Process.*, vol. 4, no. 3, pp. 223–296, 2011.
- M. Frandes, B. Timar, and D. Lungeanu, "Image Reconstruction Techniques for Compton Scattering Based Imaging: An Overview [Compton Based Image Reconstruction Approaches]," *Curr. Med. Imaging Rev.*, vol. 12, no. 2, pp. 95–105, 2016.
- L. K and C. R, "EM reconstruction algorithms for emission and transmission tomography.," J. Comput.
 Assist. Tomogr., vol. 8, no. 2, pp. 306–316, 1984.
- 425 [21] L. Bouchet, "A comparative study of deconvolution methods for gamma-ray spectra," *Astron. Astrophys.*426 *Suppl. Ser.*, vol. 113, pp. 167–183, 1995.
- L. J. Meng and D. Ramsden, "An inter-comparison of three spectral-deconvolution algorithms for gamma-ray spectroscopy," in *IEEE Nuclear Science Symposium. Conference Record. 1999 Nuclear Science Symposium and Medical Imaging Conference (Cat. No.99CH37019)*, 1999, vol. 2, pp. 691–695.
- 430 [23] H. Lemaire, "Développement d'une caméra gamma de troisième génération," p. 206, 2015.
- 431[24]Saint-Gobain, "Scintillating Optical Fibres," Product Datasheet.[Online].Available:432https://www.crystals.saint-gobain.com/sites/imdf.crystals.com/files/documents/fibre-product-sheet.pdf .
- 433 [25] ET Enterprises, "9205B series data sheet," *Product Datasheet*. [Online]. Available: http://et-434 enterprises.com/images/data_sheets/9205B.pdf.
- 435 [26] CAEN, "DT5743," Product Datasheet. [Online]. Available: https://www.caen.it/products/dt5743/.

436 437 M.-M. Bé and V. Chisté, "Table de radionucléides," LNE-LNHB/CEA, 2007. [27]