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A portable aerosol monitor with a novel linear deconvolution algorithm for fast and accurate detection of alpha emitters

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***Abstract*—Radiochemistry laboratories and nuclear power plants are required to install aerosol monitors for real-time detection of alpha-particle emitters in air. However, common radioactive aerosol monitors are unsuited for rapid emergency response, since they are neither portable nor easily deployable. Moreover, their spectrometric capabilities are usually limited, showing particularly poor performance of radioisotope identification, which makes it difficult to correctly assess the alpha-emitter concentration in air. We developed a rugged and compact radioactive aerosol monitor, fully battery powered, which can be quickly deployed in field, as well as used as a fixed station for routine monitoring. The monitor is equipped with a silicon detector, and the acquired alpha-particle spectra are analyzed employing a patented linear deconvolution algorithm. A prototype of this device was tested to evaluate its performance of radioisotope identification by using artificial alpha-emitters. Initial encouraging tests provide evidence for alpha-emitters identification with multi-line mixed-isotope alpha sources, and in energy-degraded low-counting-statistics conditions.**

***Keywords* —Alpha-particle spectroscopy; Radioactive aerosol monitor; Linear deconvolution algorithm**

I. INTRODUCTION

ALPHA-particle emitters are commonly found at radiochemistry laboratories and nuclear power plants, during either normal operation or decommissioning. These nuclides pose a serious risk to the workers, and general public, in the case of accidents [1]. In fact, even though their low penetration capability makes somehow negligible the risk due to external exposure, in the case of ingestion or inhalation the committed effective dose might be significantly relevant even in the case of low activity concentrations in air [2]. The installation of aerosol monitors is therefore often mandatory in those workplaces where alpha-emitters are manipulated, and inside and around nuclear sites [3,4].

Several aerosol monitors are available on the market, generally designed as fixed stations devoted to routine monitoring. Even though largely appropriate during normal operation, their usability during an accident is limited. First, they are generally mains powered, and show low Inner Protection (IP [5]) grade; thus, in the case of emergency, they are neither portable nor easily deployable. Second, their usually poor energy resolution and peak deconvolution algorithms, generally with limited capabilities of isotopic recognition, make it difficult to correctly assess the alpha-emitter concentration in air [6]. An accurate isotopic recognition algorithm is also required during routine monitoring, to subtract the contribution of background Naturally Occurring Radioactive Material (NORM), mostly coming from the decay of ^{222}Rn, ^{220}Rn, and related daughter nuclei.

To tackle the aforementioned limitations, a new aerosol monitor is under development. The device is designed to be both a routine monitoring system as well as a readily usable emergency device. It is characterised by a rugged and compact design, fully battery powered, and it is easily deployable in-field. It is equipped with a silicon detector sensitive to alpha- and beta-particles. The software devoted to the analysis is based on a patented linear deconvolution algorithm.

The initial tests performed with a first prototype of the device are discussed herein, focusing on its spectrometric capabilities. The instrument was tested with some multi-line alpha-emitters to simulate accidental contamination from artificial radionuclides.

II. MATERIALS AND METHODS

A. Hardware

The sensitive component of the prototype aerosol monitor is a 450 mm^2 active area silicon detector. The signal from the probe is directly fed to a Charge Sensitive Amplifier (CSA) followed by an analogue shaping stage (1 µs semi-Gaussian shaping). The signal is then digitized by a custom-made Multichannel Analyser (MCA) based on a 125 Ms/s 14-bit Field Programmable Gate Array (FPGA). The amplitude of the digitized pulse is eventually sampled and saved in a histogram. The system allows sampling on different types of filters depending on the specific application. It uses Mixed Cellulose Ester (MCE) Membrane or polytetrafluoroethylene polymer (PTFE) circular filters (standard diameter 47 mm) as standards.

An industrial Single Board Computer (SBC) manages the data acquisition, the real time elaboration, the visualization on a local display and the remote communication of the instrument (Wi-Fi, Bluetooth Low Energy – BLE, 4G). The system uses a low-voltage-powered diaphragm gas pump with a sampling flow rate on the filter of about 3 m^3/h. It is powered by a lithium ion battery pack which guarantees an operating autonomy of approximately 8 hours without recharging. The dimensions of the monitor are 60 cm × 40 cm × 30 cm, with an overall weight < 25 kg. Fig. 1 shows a 3D rendering of the prototype aerosol monitor at the current stage of development.

Fig. 1: 3D rendering of the aerosol monitor.

B. *Deconvolution algorithm*

Alpha-decaying isotopes emit alpha-particles with discrete energies. When a spectroscopy measurement is carried out using a silicon detector, the observed shape of the energy spectrum is determined by the stochastic elastic and inelastic collisions of the alpha-particles in air and in the detector lattice itself. The determination of the radionuclide activity in the measured sample therefore requires radioisotope identification by deconvolving either a multi-peak spectrum or, in general, an energy-degraded low-resolution left-tailed single peak. Therefore, often multiple radioisotopes are identified as candidates to be associated to a specific alpha peak (see e.g. ref. [7]). The patented ENEA approach [8] is adopted to overcome the common problem of ambiguity in the identification of radioisotopes. The algorithm is based on the following calculation steps:

(1) The response function corresponding to the specific silicon detector and the measurement geometry is determined from a reference alpha-particle spectrum;

(2) A library of synthetic alpha-particle spectra is calculated using appropriate modified Gaussian functions [9,10] based on the parameters retrieved in step (1);

(3) Steps (1) and (2) are due once: further measurements will take advantage of this initial effort;

(4) The measured alpha-particle spectrum is then deconvolved by minimizing its difference with a linear combination of the synthetic alpha-particle spectra calculated at step (2).

The goodness of the fitting procedure is evaluated based on the trend of the residuals R, i.e.

$$R = \frac{C_{exp}-C_{fit}}{\sqrt{C_{exp}}} \quad (1)$$

where C_{exp} are the experimental counts and C_{fit} are the ones deconvolved by the fitting algorithm.

The algorithm has proven efficient in laboratory conditions in providing unequivocal identification of radioisotopes in samples, showing a more accurate and sensitive evaluation of the sample activity with respect to traditional approaches [8].

C. *Experimental setup*

Tests were performed at the Casaccia Research Centre laboratories of the Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA). The laboratory hosts several certified multi-line alpha-sources. Among them, the following have been selected to carry out the monitor tests:

- ^{237}Np-^{241}Am-^{244}Cm (150 Bq, 98 Bq, 71 Bq, respectively).
- ^{239}Pu-^{241}Am-^{244}Cm (5546 Bq, 5320 Bq, 2020 Bq, respectively).
- ^{241}Am (213 Bq).

All tests were performed by placing the radioactive source at 2 mm from the silicon detector in air. In the case of the ^{237}Np-^{241}Am-^{244}Cm source, tests were repeated at source-to-detector distances equal to 2 mm, 5 mm and 10 mm, in order to investigate the effects of the distance, i.e. different spectral resolution and counting statistics, on the deconvolution algorithm.

The acquisition time was set to obtain at least about 1000 counts in the most populated spectral bin, with the exception of the test performed at 10 mm from the ^{237}Np-^{241}Am-^{244}Cm source. In this case, the measurement was stopped when the most populated bin reached about 200 counts, in order to investigate the algorithm performance in the case of poor spectral resolution due to low counting statistics.

III. RESULTS AND DISCUSSION

Prior to the measurement with the radioactive sources, a background measurement was launched for 10 minutes, obtaining an average count rate lower than one count per minute over the integral MCA spectrum. Therefore, the monitor is almost blind to background radiation and it is not affected by electronic disturbances.

Fig. 2 shows the counts obtained with the pure ^{241}Am source (red dots) and the reconstructed peak shape calculated by the algorithm (solid line). The peak is correctly recognised by the software. The fit is satisfactory both quantitatively and qualitatively, by visual inspection of the data distribution vs fit function, and residuals distribution vs zero line (discrepancies within ± 10%), without any significant under-/overestimation trend.

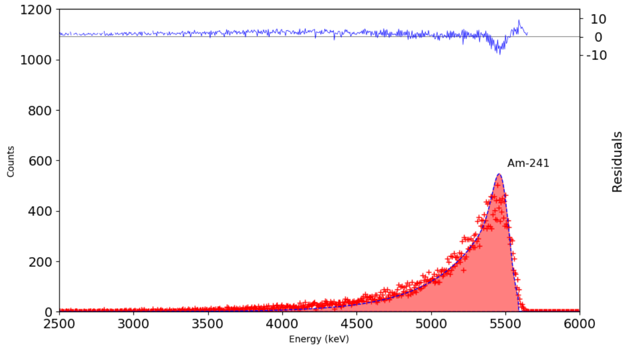

Fig. 2: Experimental counts (dots) and reconstructed peak (dashed line) in the case of the pure ^{241}Am source. The isotope identified by the software is labelled just above of the reconstructed peak. Residuals are compared with the zero line on the top of the graph.

Encouraging results were obtained while measuring a multi-peak source where spectral band overlapping is significant, such as ^{239}Pu-^{241}Am-^{244}Cm (Fig. 3). In this case, the three nuclides of the mixture are correctly deconvolved, represented as solid lines in the graph. Above each peak, the software correctly labels the corresponding isotope: 5.157 MeV peak for ^{239}Pu, 5.486 MeV peak for ^{241}Am, and about 5.8 MeV for ^{244}Cm (differently from the other isotopes of the source, the ^{244}Cm peak is given by the superposition of two emission lines which are almost overlapped, i.e. 5.805 MeV (76.9%) and 5.763 MeV (23.1%)). The spectrum reconstruction (dashed line) is satisfactory both quantitatively and qualitatively, by visual inspection of the data distribution vs fit function, and residuals distribution vs zero line (discrepancies within ± 10%), without any significant under-/overestimation trend. It is worth noting that the algorithm is capable of recognising (and reconstructing) the contribution of the left tail of the ^{241}Am peak, even though about 50% of its area is overlapped with the ^{239}Pu peak.

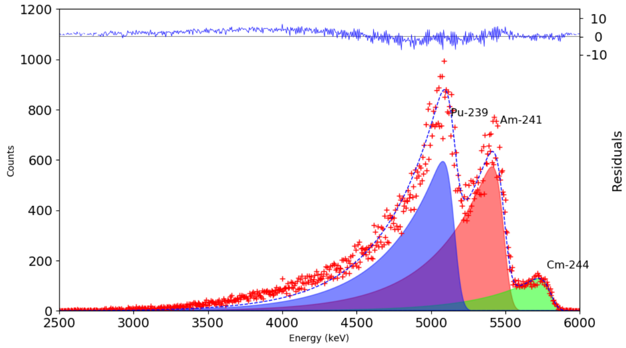

Fig. 3: Experimental counts (dots) and reconstructed peaks (solid lines) in the case of the mixed ^{239}Pu-^{241}Am-^{244}Cm source. The isotope identified by the software is labelled just above of the reconstructed peak. The final convolved spectrum reconstructed by the algorithm is shown as a dashed line. Residuals are compared with the zero line on the top of the graph.

Fig. 4 shows the counts obtained with the mixed ^{237}Np-^{241}Am-^{244}Cm source (red dots), the reconstructed peaks (solid lines and labels), and the reconstructed convolved spectrum (dashed lines), for increasing source-to-detector distances, namely 2 mm (a), 5 mm (b), and 10 mm (c). The spectrometric capabilities of the device are satisfactory even at the largest source-to-detector distances. The full shape of the spectrum is always correctly deconvolved, and the radionuclides are correctly identified for each source-to-detector distance, and for different measurement times, the latter corresponding to distinct counting statistics. The spectrum reconstruction (dashed line) is satisfactory both quantitatively and qualitatively, by visual inspection of the data distribution vs fit function, and residuals distribution vs zero line, without any significant under-/overestimation trend, with the possible exception of the 2 mm distance spectrum, where the largest deviations of the residuals are observed, even though the variations vs zero line still fall between ± 15%. Those deviations are due to chance coincidence between alpha-particles and beta-rays, interactions of x-rays, scattering effects, etc. [11,12,13], which are not directly included in the fit function. However, such effects do not prevent the deconvolution algorithm to correctly identify the alpha-emitters occurring in the spectrum. For the two other cases (5 mm and 10 mm distance), such effects are not appreciable. In fact, the increased source-to-detector distance slightly improves the spectral resolution, since only particles emitted along narrower angles can be detected, i.e. limiting the peak broadening due to alpha-particle collisions in air. Moreover, the lowered counting efficiency due to the increased source-to-detector distance causes a significant drop in the probability of chance coincidence between alpha-particles and beta-/x-rays.

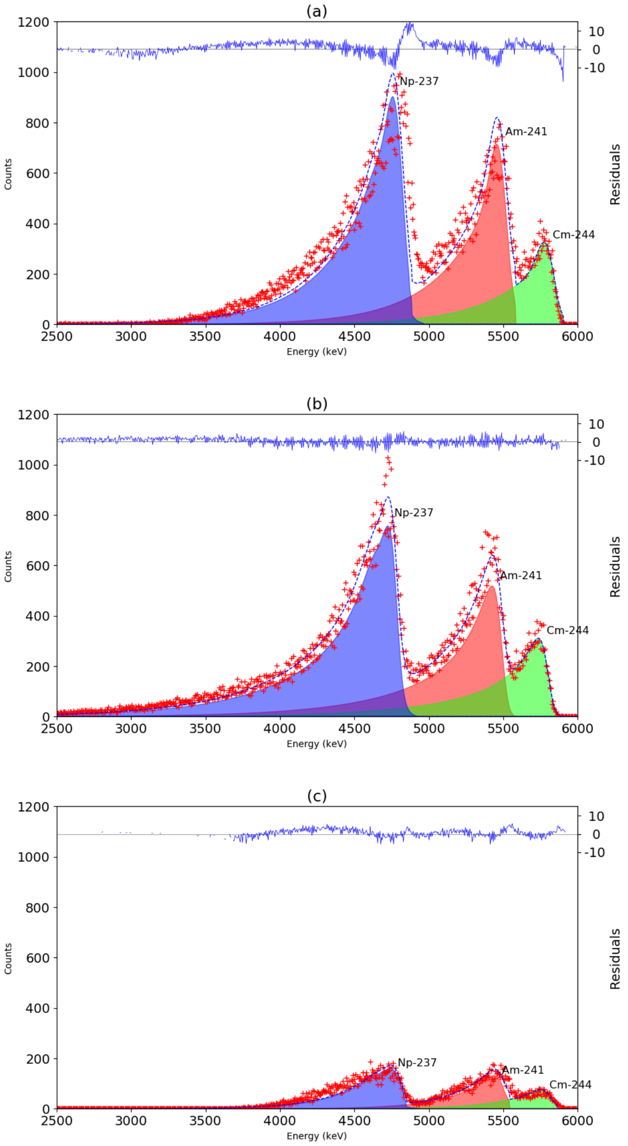

Fig. 4: Experimental counts (dots) and reconstructed peaks (solid lines) in the case of the mixed ^{237}Np-^{241}Am-^{244}Cm source at different source-to-detector distances, namely, 2 mm (a), 5 mm (b), and 10 mm (c). The isotope identified by the software is labelled just above of the reconstructed peak. The final convolved spectrum reconstructed by the algorithm is shown as a dashed line. Residuals are compared with the zero line on the top of the graphs. In the case of 2 mm distance (graph (a)), the relatively large deviations of the residuals (still included between ± 15%) do not prevent the radioisotope identification.

IV. CONCLUSIONS AND OUTLOOKS

A novel alpha-beta aerosol monitor is under development. The device was developed to be either a fixed station, as well as an easily deployable device for emergency use. The implemented algorithm for isotopic recognition was tested with multi-peak alpha sources, showing appropriate and reliable spectrometric performances, both in terms of isotopic identification and spectrum reconstruction (residuals always within ± 10% without showing any significant under-/overestimation trend), also in condition of extremely poor peak resolution and low counting statistics. The device then proved to be equivalent (or superior) to conventional fixed aerosol monitoring stations for isotopic recognition, with the advantage of being designed also for emergency use.

Further steps of the research will involve the investigation of the spectrometric properties of the device for NORM recognition, i.e. background subtraction, and employing the final measurement geometry, i.e. measuring contaminated and uncontaminated filters. This will allow investigating the capability of the system of quantitatively measuring the concentration of artificial radionuclides (or NORM) in the ambient air.

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