Radiation Physics and Chemistry 196 (2022) 110112

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

journal homepage: www.elsevier.com/locate/radphyschem

α-SpecTrim: A new application designed for reproducing alpha-particle spectra

A. Fernández Timón [a,*], M. Jurado Vargas [b], C. Peña Morante [c], J. Sánchez-Oro Calvo [a]

[a] Universidad Rey Juan Carlos, 28933, Móstoles, Madrid, Spain
[b] Departamento de Física, Universidad de Extremadura, 06071, Badajoz, Spain
[c] GMV, 28760, Tres Cantos, Madrid, Spain

ARTICLE INFO

Keywords:
Monte Carlo simulation
Alpha-particle spectrometry
α-SpecTrim application

ABSTRACT

In this work, we introduce a new software application developed to reproduce α-particle spectra measured with silicon detectors. It takes into consideration the main physical processes involved in the interaction of alpha particles with the source and the detector. This application is considerably more user-friendly than multipurpose codes, being also the runtimes to obtain a spectrum much lower. It makes use of the TRIM simulation code for tracking alpha particles in the source and the detector entrance window. Concerning to the response function of a silicon detector, the contribution of electronic excitation and ionization produced by the alpha particles is also obtained by simulation, while the electron-hole pair statistics and the broadening produced by the amplifier system (electronic chain) are included as convolutions of theoretical functions. The code α-SpecTrim is written in Python 3 language using Flask framework, so it can be run in every PC or laptop. It is free and can be provided if requested to the authors.

1. Introduction

As known, alpha-particle spectrometry requires thin sources, usually prepared by deposition, electrodeposition or sublimation in vacuum. The measurements are often made using silicon semiconductor (PIPS) detectors, due to their high energy resolution. In spite of this, the collected spectra involve the asymmetric broadening of alpha lines and consequently an overlapping of the peaks, making often the analysis of alpha-particle spectra difficult.

Steinbauer et al. (1994) provided a survey of the physical processes that contribute to the shape of an alpha-particle spectrum. In summary, they proposed that the final pulse-height distribution could be obtained as a convolution of the energy distribution of the alpha particles emitted from the source, and the other distribution due to the detector response function. This last response is complex and is also divided in a set of individual distributions. In addition, the distribution due to the alpha particles leaving the source can be obtained by theoretical approximations only in the case of very thin sources measured at large distances from the detector. For this reason, the analysis and deconvolution of an alpha-particle spectrum usually requires auxiliary tools, which involve the theoretical fitting to complex mathematical functions, or the use of Monte Carlo simulation.

Monte Carlo methods have become useful tools to be applied in a wide range of issues in nuclear physics, so that many software codes have been written for general or particular purposes. In particular, programs as AlfaMC (Peralta and Louro, 2014), SRIM (Ziegler et al., 2010) or GEANT4 (Agostinelli et al., 2003) have been applied to different tasks related to the detection of alpha particles, and even some works have been carried out to simulate alpha-particle spectra under particular conditions (Díaz-Francés et al., 2017; Krupa and Kurzak, 1991; Siiskonen and Pöllänen, 2004). In spite of this, there is a lack of available tools specifically designed to reproduce alpha-particle spectra under general conditions.

The main goal of this work is the development of a computer application specifically designed to reproduce alpha-particle spectra obtained with silicon semiconductor detectors. The code α-SpecTrim is an user-friendly tool which can be used in many tasks related to alpha-particle spectrometry: study of line shapes for alpha emissions, efficiency calibrations, deconvolution of complex alpha-particle spectra, etc. An intuitive interface helps the user, who only has to introduce the required input data before the program works. Its implementation combines simulation processes with the convolution of theoretical distributions, taking into consideration the physical processes associated to the interaction of alpha particles with matter. All the simulations are

* Corresponding author.
E-mail address: alfonso.fernandez@urjc.es (A. Fernández Timón).

https://doi.org/10.1016/j.radphyschem.2022.110112
Received 13 January 2022; Received in revised form 23 March 2022; Accepted 29 March 2022
Available online 1 April 2022

performed with the TRIM code, an application which is included in SRIM package (Ziegler et al., 2010).

2. Method

The model proposed by Steinbauer et al. (1994) to describe the response function of silicon detectors to alpha particles shows that the final spectrum can be obtained as a convolution of the energy distribution of the alpha particles emitted from the source with the distribution due to the detector response function. Following this model, the procedure to reproduce the shape of an alpha-particle emission could be divided into two main stages: the first phase related to the simulation of alpha particles in the source and through the detector entrance window, and a second stage, involving the convolution of three theoretical distributions that represent the remaining detector response effects. These distributions are related to the ionization–excitation processes into the detector, the generation of hole–electron pairs, and the electronic noise produced in the electronic chain associated to the spectrometer. This was essentially the procedure followed by us in an earlier work (Fernández Timón et al., 2010).

The distributions corresponding to the production of hole-electron pairs and to the contribution of electronic noise can be expressed by means of simple mathematical functions. However, the distribution due to the ionization/excitation produced by alpha particles in the detector is very complex, and it cannot be derived by means of an analytical expression deduced from theoretical models. This contribution has a marked dependence on the energy of alpha particles, as evidenced in a recent study (Jurado Vargas et al., 2020), and it plays an essential role in the alpha line shape. Therefore, it is required to include the ionization/excitation processes occurred in the detector into the proper MC simulation, instead of considering this distribution as a unique function to be convoluted, as carried out as an approximation in a previous work (Fernández Timón et al., 2010).

The software used by us to perform the simulations is the TRIM code (Ziegler et al., 2010). This Monte Carlo code uses text files for the input and output data and will be responsible for performing all the calculations related to the simulation. However, because its usage is complex, the authors of the TRIM code developed some years later the SRIM interface, which supplies a simple way to input data, execute and visualize the simulation results. The code TRIM (via SRIM) has already been used by us in several applications involving alpha-particle interactions with matter (Jurado Vargas and Fernández Timón, 2004, 2005). In this work, the last version of code SRIM-2013 was used.

Our methodology for reproducing alpha-particle spectra starts with the simulation of the tracks of alpha particles in the source material. Alpha particles are initially located in the source substrate, having their initial positions and output directions randomly distributed. For radionuclides with multiple emission energies, the alpha particles are split into several sets proportionally to the corresponding emission probabilities. Emerging particles are filtered, selecting only those ones that would reach the detector according to their positions and directions when they come out of the source. We assume that they follow straight paths from the source to the detector, so no energy losses in the vacuum chamber are considered.

These remaining particles constitute the input for the second simulation, which follows the track of the alpha particles across the entrance window of the detector, which is constituted by a dead layer with a given thickness. Following the previously mentioned procedure, the positions and output directions of alpha particles after crossing the detector window are used as input data for the third (and last) simulation. In this case, a Si layer is chosen with a proper thickness for the residual alpha particles to lose all their remaining energy in the sensitive volume of the detector. An output file of the simulation provides the individual quantity of energy spent by each alpha particle in ionization/excitation processes. These particles are then binned by energy to form a histogram, which must be convolved in the following steps with the theoretical distributions representing the remaining detector response effects.

The energy E spent by each particle in the ionization/excitation processes is available for the production of electron-hole pairs and lattice vibrations, This distribution is well described by a Gaussian with a standard deviation depending on energy E, which is given by $\sigma = \sqrt{wEF}$, where w is the average energy required to produce an electron-hole pair, and F is the Fano factor (Knoll, 1999). Therefore, the histogram obtained after simulating the ionization/excitation processes is convoluted with the distribution due to the production of electron-hole pairs.

Finally, a last convolution must be performed, which takes into account the noise introduced by the electronic equipment. This last distribution is described by a Gaussian, with a standard deviation that can be easily determined experimentally using a pulse generator connected to the preamplifier of the detection system.

Fig. 1 shows, as an introductory example, the application of this methodology to the monoenergetic emission of 5.304 MeV for ^{210}Po. The main detection conditions and input data considered to reproduce this alpha line are shown in Table 1. We can observe the sequence of the stages followed in the methodology aforementioned described, and the effect produced in the alpha-line shape by each step. In particular, it can be noticed the role of the ionization/excitation distribution on the asymmetry of the alpha line and the corresponding displacement towards the low-energy zone of the spectrum. A smoothing of the alpha line is also observed, which is a result of the convolution with the distribution due to the production of electron-hole pairs.

3. Structure of α-SpecTrim

Each stage of the aforementioned process requires some input data. A few of them must be provided by the user, while the remainder information is obtained from the output file of the last step. The data contained in these output files must be processed in order to be included in the input file corresponding to the next stage. These tasks could be manually performed by the user, although they are quite tedious and complex, which may eventually result in errors during simulation. For these reasons, we have developed α-SpecTrim, a simple and intuitive interface to introduce the input data needed for the whole process. One of the main advantages is the fact that it automatizes the running process that corresponds to the simulations and convolutions, including all these tasks in just one execution without requiring additional user attention.

The basic role of α-SpecTrim in the process depends on each particular stage (simulations or convolutions). Fig. 2 shows a flow chart with the different stages of the work performed by the code. As mentioned in the last section, the whole procedure includes three simulations and two convolutions. The application α-SpecTrim is responsible for filling the input files of each simulation or convolution. Moreover, it runs the TRIM execution file for the simulation or the adequate subroutine for the convolution, analyses the output files and prepares them to be part of the input data for the next stage.

The data for the whole process are submitted in different graphical forms, each one associated to a particular task in the procedure: source and backing, detector's dead layer, energy histogram and convolution's features. Fig. 3 shows several screenshots corresponding to different stages of the α-SpecTrim interface. The application offers a database to the user corresponding to a wide set of radionuclides. In the same way, a batch with usual compounds with single and multi-atomic compositions for the backing, the source and the detector window is available. Both databases can be easily extended by the user into the code α-SpecTrim including new radionuclides or compounds.

The progress of the simulations and the convolutions are continuously monitored, showing a progress bar to the user while executing, which favours the user experience. Notice that these processes may eventually last, so the inclusion of these progress bars helps the user to know that it is still executing and not blocked. Even more, the user will

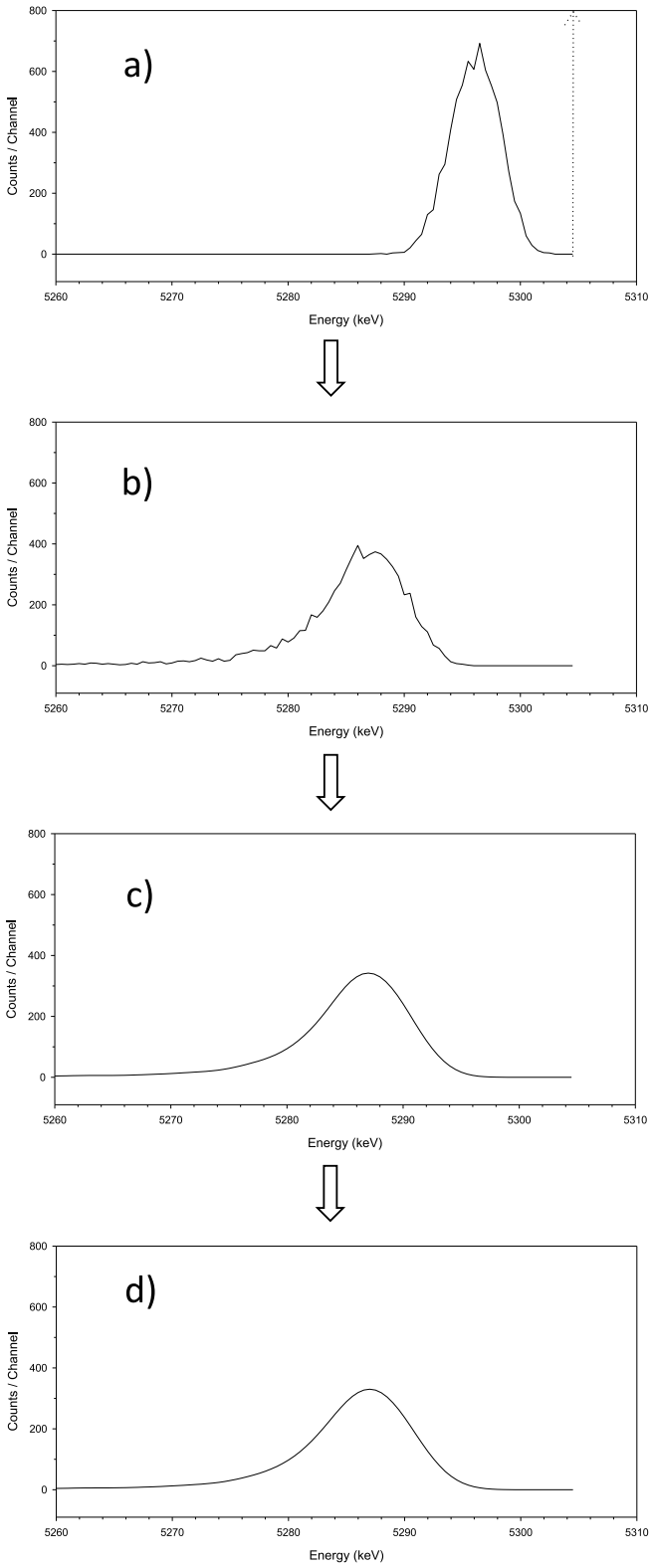

Fig. 1. Evolution of the line shape for the monoenergetic radionuclide ^{210}Po, following the method used by α-SpecTrim. Figure a) shows the output spectrum for the simulation of the source and the detector window. Dotted line marks the emission energy for ^{210}Po. Figure b) shows the output spectrum after the simulation of the I/E processes. Figure c) shows the spectrum as result of the convolution with the hole-electron pair generation distribution. At the end, Figure d) depictures final spectrum after the convolution with the electronic noise distribution.

Table 1
Input data considered to obtain the alpha line of ^{210}Po shown in Fig. 1. The energy considered for the nuclide ^{210}Po was taken from Nucléide-Lara database (Nucléide-Lara).

Source composition	Po (9.25 g/cm^3)
Source thickness	100 Å
Source radius	0.75 cm
Radionuclide	^{210}Po 5304.3 keV (100%)
Backing composition	Stainless Steel (8 g/cm^3)
Detector's window composition	Si (2.32 g/cm^3)
Detector's window thickness	500 Å
Detector's window radius	1.25 cm
Source to Detector Distance	2 cm
Histogram gain	0.5 keV/Channel
Fano's Factor	0.13 eV
Electron-Hole creation energy	3.67 eV
Electronic Noise Sigma	1 keV

know which percentage of the experiment has already finished. The execution time required to generate an alpha-particle spectrum is highly variable, depending on several input data, such as the number of alpha particles, the thickness considered for the backing and for the source substrate, or the energy interval chosen in the convolution processes. For example, the execution time corresponding to the spectrum shown in Fig. 1, using a conventional laptop (Intel Core i5 processor with 8 GB of RAM), was approximately 1 h. This running time was spent mainly in the steps corresponding to the first and third simulation (each one around half an hour), being of the order of 1 min for the other processes.

When the process is ended, an output file is generated, which includes a summary for the input data for the simulations and the convolutions, the calculated detection efficiency and the final histogram giving the number of alpha particles detected versus energy.

The code α-SpecTrim has been implemented in Python 3 programming language, using the framework Flask (Flask framework,). This framework is designed for developing multiplatform web applications under the MVC (Model, View and Controller) pattern. Furthermore, Flask has many available extensions, which ease the development of more complex systems, such as Flask-WTF for forms or Flask login. It also includes a development web server that allows the user to test the project locally before deploying it and supports unitary tests. The only prerequisites to execute α-SpecTrim is to install a Python interpreter (version 3.8.10) and the MongoDB 4.4.5 database (Bradshaw et al., 2019). In order to facilitate the installation to users which are not familiar with these technologies, we provide an installer which checks and installs these prerequisites if necessary. The code α-SpecTrim must be run in a local server, which has to be initialized at the beginning of each session. Once the server is launched, it is only required to navigate to the local direction of the application to initiate α-SpecTrim. The code will be distributed under Creative Commons license upon request to the authors.

4. Application

In order to show the applicability of the code α-SpecTrim, we have measured a source of ^{233}U with a PIPS detector, which involves several alpha emissions overlapped in the spectrum. The source is made by electrodeposition onto a stainless steel backing, with an active radius of 0,75 cm, and was located into a vacuum chamber (Canberra, 7400A) in front of the detector at a distance of 11 mm. The detector is a CANBERRA passivated implanted planar silicon (PIPS) detector with 50 mm^2 active area and 11 keV nominal resolution, connected to a stabilized power supply (CMTE, 3122-D). The signals are passed through a low-noise preamplifier (Canberra, 2004DM), a conventional amplifier (Canberra, 2015A), an analogue-to-digital converter (Canberra, 8713), and finally stored in a multichannel analyzer (Canberra, Accuspec-B). In order to obtain the effect of the electronic equipment on the signal, a

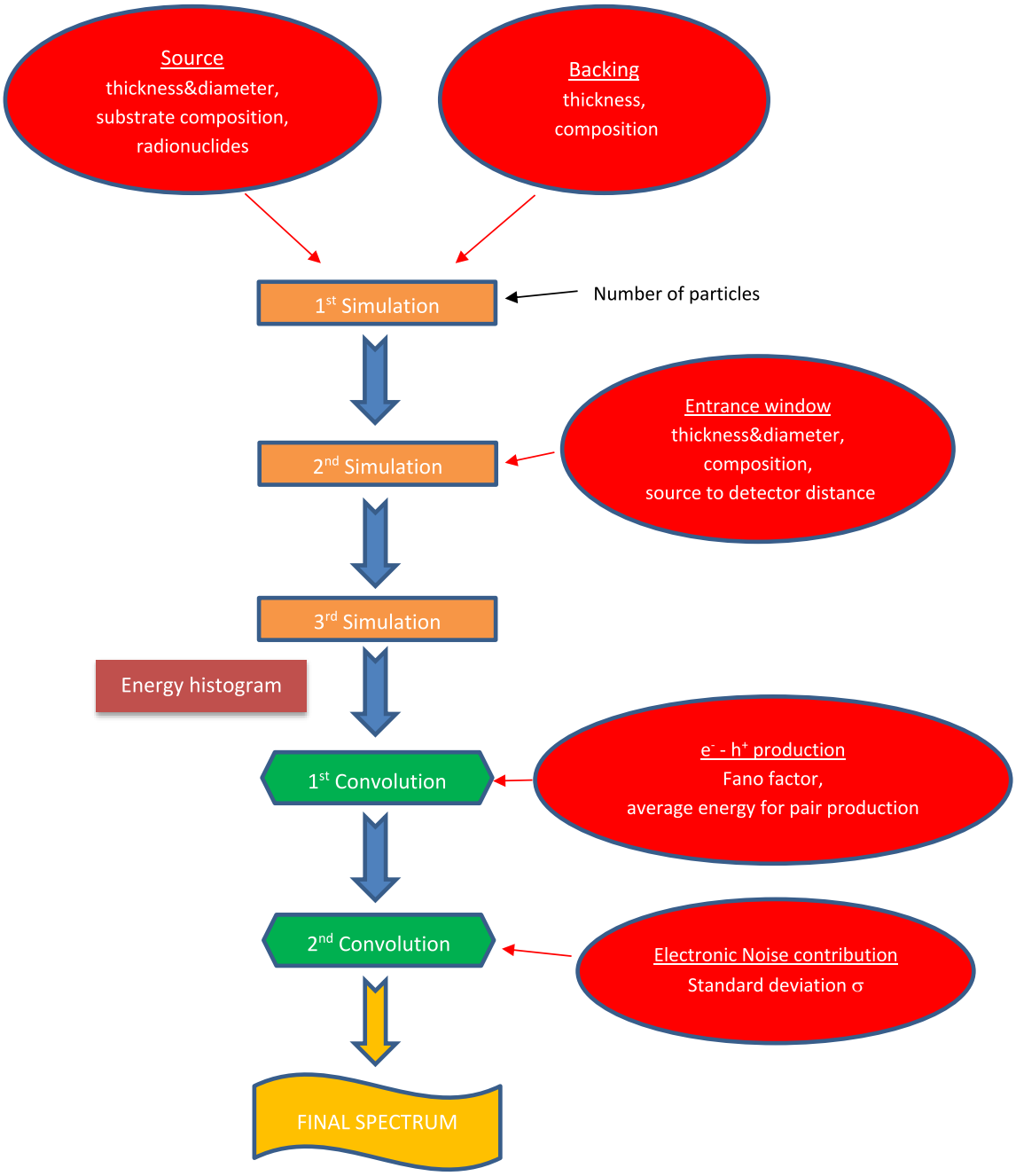

Fig. 2. Flow-chart of α-SpecTrim.

pulse generator (EG&G Ortec, 448) was connected to the input of the preamplifier. The peak of the "pulser'' in the spectrum was fitted with a Gaussian, giving a standard deviation of 3.53 keV.

To reproduce this alpha-particle spectrum with the code α-SpecTrim, several input data were included in the application, taking into consideration all the physical features corresponding to the source and detector. Table 2 shows a summary of these input data, including also the energies and emission probabilities considered for the nuclide ^{233}U. We generated 200.000 alpha particles, which were randomly positioned in the source substrate and with output directions also randomly distributed. The detector window was included as a Si layer of 500 Å of thickness, as indicated by the detector's manufacturer. The values of the Fano factor for silicon and the average energy needed for the production of one electron-hole pair were taken from the work of Steinbauer et al. (1994). The source was considered to mostly consist of UO_2. Because the source thickness is unknown, we generated several spectra assuming different values for this thickness. A value of 500 Å was finally adopted because gave the best agreement between the theoretical and experimental spectrum. The detection efficiency calculated by α-SpecTrim was of 2.33%.

Fig. 4 shows the experimental spectrum and the theoretical one obtained by α-SpecTrim. In general, there is a good agreement over the entire energy interval of alpha-particle emissions. In order to compare both spectra, the differences between the experimental and theoretical values (residuals) must be given in terms of the Poisson standard deviation σ for each channel. The mean value obtained for these weighted residuals was 2.72σ. It represents a good agreement between the theoretical and the experimental spectrum, taking into consideration that all the data included in α-SpecTrim were fixed, so that no fitting or optimization process was performed. The only exception was the source

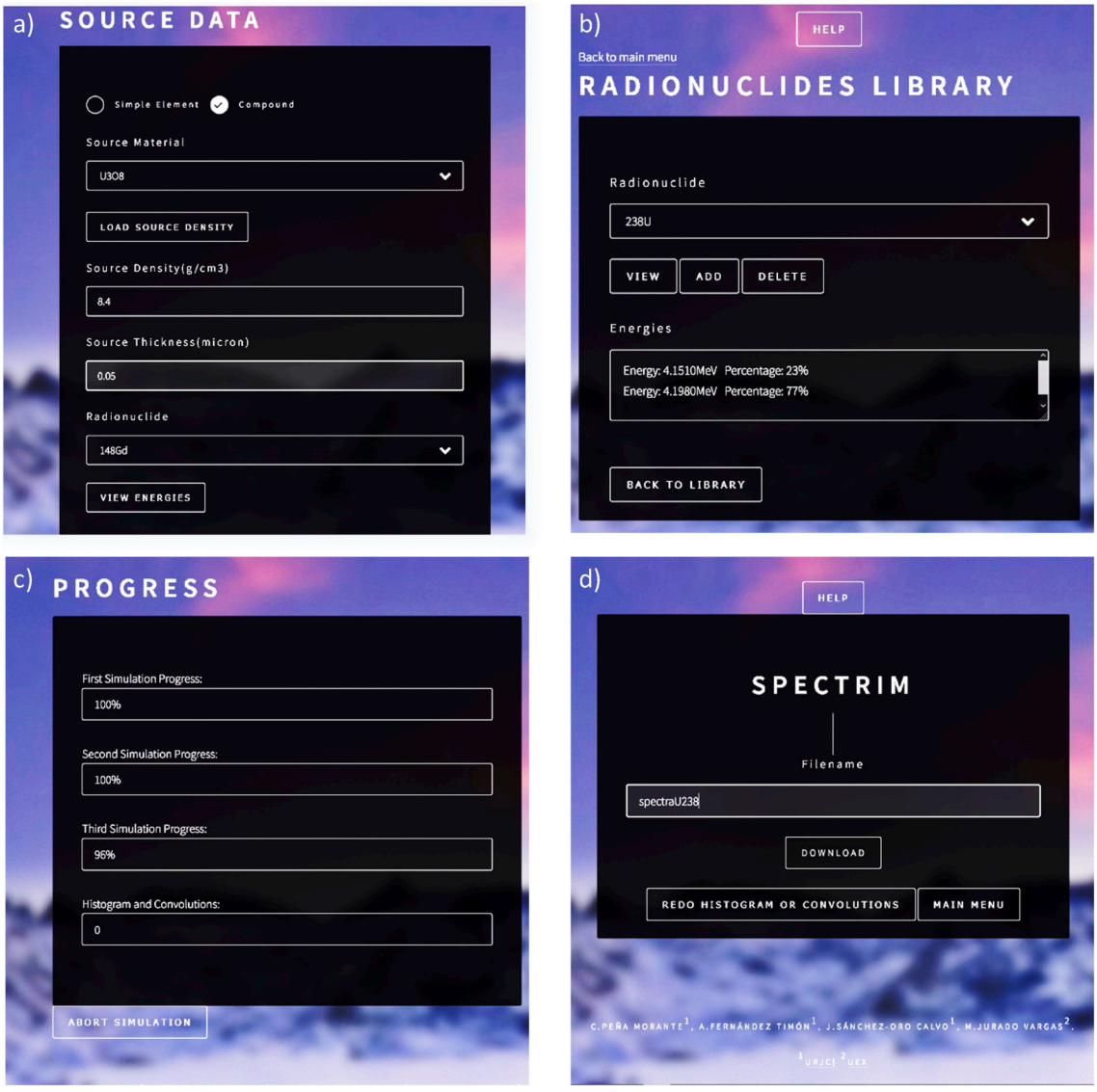

Fig. 3. Screenshots of several steps in α-SpecTrim: source data submission (a), radionuclides library (b), progress bars (c) and final page (d).

Table 2
Input data to reproduce the experimental spectrum of ^{233}U. They include all the physical features corresponding to the source and detector used. The energies and emission probabilities considered for the nuclide ^{233}U were taken from Nucléide-Lara database (Nucléide-Lara).

Source composition	UO_2 (10.97 g/cm^3)
Source thickness	500 Å
Source radius	0.75 cm
Radionuclide	^{233}U
	4729.0 keV (1.62%)
	4754.0 keV (0.19%)
	4783.5 keV (13.26%)
	4796.0 keV (0.28%)
	4824.2 keV (84.65%)
Backing composition	Stainless Steel (8 g/cm^3)
Detector's window composition	Si (2.32 g/cm^3)
Detector's window thickness	500 Å
Detector's window radius	0.4 cm
Source to Detector Distance	1.1 cm
Histogram gain	2.1848 keV/Channel
Fano's Factor	0.13 eV
Electron-Hole creation energy	3.67 eV
Electronic Noise Sigma	3.53 keV

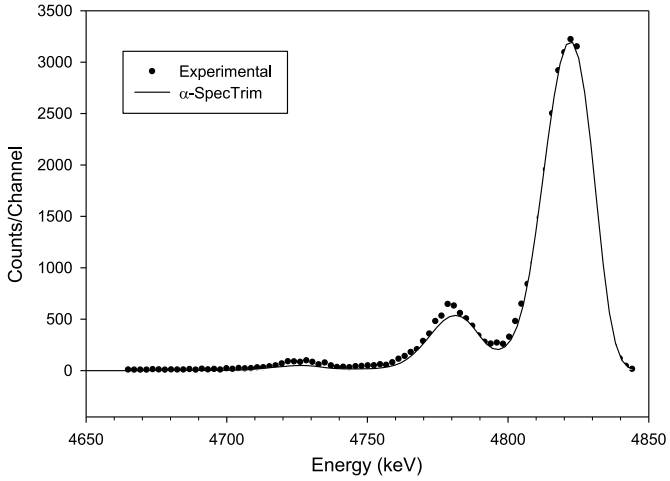

Fig. 4. Experimental spectrum of ^{233}U and the theoretical one obtained by α-SpecTrim.

thickness, which had to be optimized because this value was unknown.

5. Conclusions

We have developed the application α-SpecTrim, implemented in Python language to specifically reproduce alpha-particle spectra measured with silicon detectors. The software includes a part involving the Monte Carlo simulation with the TRIM code, and subsequently a set of theoretical convolutions that represent the remaining detector response. This application takes in account the main processes influencing the shape of an alpha-particle spectrum. This methodology does not use any optimization procedure to fit the theoretical spectrum to the experimental one, i.e., all the parameters are fixed a priori, so they are not free. The application α-SpecTrim can play an important role as an auxiliary tool in alpha-particle spectrometry.

Author statement

Fernández Timón: Conceptualization, Methodology, Software, Formal analysis, Writing - Original Draft, Writing - Review & Editing, Visualization.

Jurado Vargas: Conceptualization, Methodology, Formal analysis, Writing - Original Draft, Writing - Review & Editing, Supervision, Funding acquisition.

Peña Morante: Software, Validation, Writing - Review & Editing.

J. Sánchez-Oro Calvo: Software, Writing - Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Thanks are due to the Junta de Extremadura and Fondo Europeo de Desarrollo Regional FEDER, Spain (project GR18093) and Ministerio de Ciencia, Innovación y Universidades, Spain (project PGC2018-095322-B-C22) for financial support.

References

Agostinelli, S., et al., 2003. GEANT4-a simulation toolkit. Nucl. Instrum. Methods A 506, 250–303. https://doi.org/10.1016/S0168-9002(03)01368-8.

Bradshaw, S., Brazil, E., Chodorow, K., 2019. MongoDB: the Definitive Guide: Powerful and Scalable Data Storage, third ed. O'Reilly Media, Sebastopol.

Díaz-Francés, J.A., Cortés-Giraldo, M.A., Jiménez-Ramos, M.J., Hurtado-Bermúdez, S., 2017. Simulation of the response of a PIPS detector using GEANT4 code. Nucl. Instrum. Methods A 875, 21–26. https://doi.org/10.1016/j.nima.2017.08.033.

Fernandez Timón, A., Jurado Vargas, M., Martin Sánchez, A., 2010. A method to reproduce alpha-particle spectra measured with semiconductor detectors. Appl. Radiat. Isot. 68, 941–945. https://doi.org/10.1016/j.apradiso.2009.10.046.

Flask framework. https://flask.palletsprojects.com/en/2.0.x/. (Accessed 30 July 2021).

Jurado Vargas, M., Fernández Timón, A., 2004. Scattering and self-absorption corrections in the measurement of α-particle emitters in 2π geometry. Nucl. Instrum. Methods B 217, 564–571. https://doi.org/10.1016/j.nimb.2003.12.038.

Jurado Vargas, M., Fernández Timón, A., 2005. Dependence of self-absorption on thickness for thin and thick alpha-particle sources of UO_2. Nucl. Instrum. Methods A 548 (2005), 432. https://doi.org/10.1016/j.nima.2005.04.056.

Jurado Vargas, M., Fernández Timón, A., Ziegler, J.F., 2020. Determination and analysis of ionization/excitation distributions for alpha particles in silicon detectors. Nucl. Instrum. Methods A 971, 164134. https://doi.org/10.1016/j.nima.2020.164134.

Knoll, G.F., 1999. Radiation Detection and Measurements, first ed. John Wiley & Sons, New York.

Krupa, R.J., Kurzak, K., 1991. Simulation and evaluation of α-spectra obtained with semiconductor detectors. Nucl. Instrum. Methods A 307, 469–483. https://doi.org/10.1016/0168-9002(91)90220-K.

Nucléide-Lara. http://www.lnhb.fr/nuclear-data/module-lara/. (Accessed 4 October 2021).

Peralta, L., Louro, A., 2014. AlfaMC: a fast alpha particle transport Monte Carlo code. Nucl. Instrum. Methods A 737, 163–169. https://doi.org/10.1016/j.nima.2013.11.026.

Siiskonen, T., Pöllänen, R., 2004. Simulation of alpha particle spectra from aerosol samples. Appl. Radiat. Isot. 60, 947–953. https://doi.org/10.1016/j.apradiso.2004.02.021.

Steinbauer, E., Bortels, G., Bauer, P., Biersack, J.P., Burger, P., Ahmad, I., 1994. A survey of the physical processes which determine the response function of silicon detectors to alpha particles. Nucl. Instrum. Methods A 339, 102–108. https://doi.org/10.1016/0168-9002(94)91787-6.

Ziegler, J.F., Ziegler, M.D., Biersack, J.P., 2010. SRIM – the stopping and range of ions in matter. Nucl. Instrum. Methods B 268, 1818–1823. https://doi.org/10.1016/j.nimb.2010.02.091.