k0-INRIM User's Manual

version 2.1

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1 Introduction

A software package, called k0-INRIM, was developed to assist NAA analysts during data elaboration and uncertainty evaluation following the recommendations of the guide to the expression of uncertainty in measurement GUM [1]. The main aim was to make automatic the compiling of the uncertainty spreadsheet for the k_0-NAA method described in [2].

Specifically, the software accepts as an input spectra information and experimental data to obtain the mass fractions of the investigated elements including their uncertainty budgets and detection limits.

The equation model (eq. 35) implemented in the present version applies to activation and decay path types I, IIB, IVB and VI defined in [3]. A warning message appears in the case of complex activation decay paths that are not yet implemented. Similarly, the user is informed when non-$1/v$ nuclides requiring the application of Westcott formalism are selected.

It is worth to note that the use of the current version allows performing analysis that feature experimental setups concerning sample (containing the investigated elements) and standard (containing the monitor element) gamma acquisitions in different counting positions, albeit at least one of the two should be in the reference position (see paragraph 3.3.1); moreover, extended geometry of samples and standards are taken into account.

The calibration of the detection system represents the focal point of the whole analysis; it is entirely experimental and requires the measurement of gamma reference sources at the counting positions of the samples in order to define them all and also evaluate additional parameters linked to other efficiency corrections.

Information concerning installation and use of the software is given in sections 2 and 3, respectively. It is worth to note that the structure of the section 3 corresponds to the workflow of the analysis.

2 Software installation

The k0-INRIM is potentially multi-platform as it is expected to work flawlessly also on macOS and linux systems although this version has been heavily tested only on 64 bit Windows systems (versions 7 and 10).

The software running requires the previous installation of the Python compiler, version 3.6 or 3.8 [4], freely available at `https://www.python.org/downloads/`. During the Python installation, click the "Add python3.x to PATH" check-box. Moreover, the k0-INRIM uses the additional modules "xlrd", "xlsxwriter", "numpy", "scipy", "pandas" and "matplotlib" [5, 6] available at the

"pip" internet repository.

The k0-INRIM can be found as a compressed folder download-able from the github repository (`https://github.com/marcodiluzio/k0-INRIM/releases/tag/v2.1`); once the zip (or, alternatively, the tar.gz) folder, named Source code, has been downloaded on computer, a simple unpack is required. In the unpack process, it is recommended to avoid system-reserved destinations, such as the C://Programs path, as this will prevent the writing of working files unless the user has administrator privileges; the «user» or «Documents» folders are suggested. Once unpacked, a «k0-INRIM» folder will appear in the destination. The main folder includes two additional «data» and «classes» folders, and the «k0-INRIM.py», «k0-INRIM.pyw», «LICENSE» and «requirements.txt» files.

The «k0-INRIM.py» is the main Python script using Python objects defined in the «naaobj.py», «GUI_things.py» and «recovery.py» supporting script files included in the «classes» folder. Those represent the source codes, accessible for expert users for further development. The «requirements.txt» file includes information of the required additional modules and «LICENSE» a copy of the software GNU general public license. The file «k0-INRIM.pyw» is equivalent to «k0-INRIM.py» and works only under Windows to start the program without the underlying console.

The software main settings are defined in the «k0-set.cfg» file included in «data» along with the i) «characterization», ii) «coincidences», iii) «facility», iv) «irradiation», v) «k0data», vi) «literature-data», vii) «models», viii) «monitor_elements», ix) «samples», x) «saves» and xi) «sources» folders storing information on i) detector characterizations, ii) literature data for true-coincidences correction, iii) irradiation facilities and detectors, iv) performed irradiations, v) nuclear data from the k_0 database [7], vi) data concerning γ-emissions, fast neutron reactions and self-absorption correction, vii) measurement models, viii) elements to be used as monitors for flux ad gradient evaluations, ix) samples involved in analysis, x) savefiles and xi) γ sources certificates, respectively.

3 Use

3.1 Main window

The software main window is opened by double clicking on the «k0-INRIM.py» file (or «k0-INRIM.pyw») on Windows, while if other systems are in use (macOs or linux) the corresponding terminal should be used to start the script. A supporting prompt window (console) is also opened to display user messages and software errors, if the software is started from the «k0-INRIM.py» file; be aware that closing this prompt window stops the program.

At first start-up the k0-INRIM automatically attempts to install the missing additional modules; for this procedure a working internet connection is required. (Figure 1)

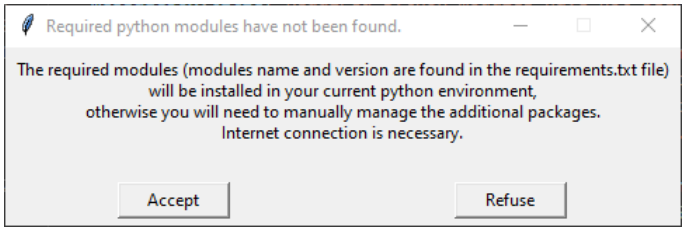


Figure 1: The start-up check for missing modules

In case some error occurs, modules have to be manually installed by opening a Windows prompt (or macOS/linux terminal) and entering the command *pip install module-name==version* by replacing for each *module-name==version* each name (with version number) as present in the «requirements.txt» file. Use instead the command *pip3 install module-name==version* if macOS or linux systems are running.

In the main windows (Figure 2) 9 separated regions can be identified, with titles in the upper left side, suggesting the preferable workflow to follow; they are: 1) *Utility*, 2) *Characterization*, 3) *Analysis name*, 4) *Irradiation*, 5) *Background*, 6) *Standard*, 7) *Sample*, 8) *Limits* and 9) *Results*.

Each region contains buttons, drop-down menus, sliders and spinboxes to perform actions. Finally, the bottom of the window includes a label where hints are printed and a progress bar to monitor advancement of tasks.

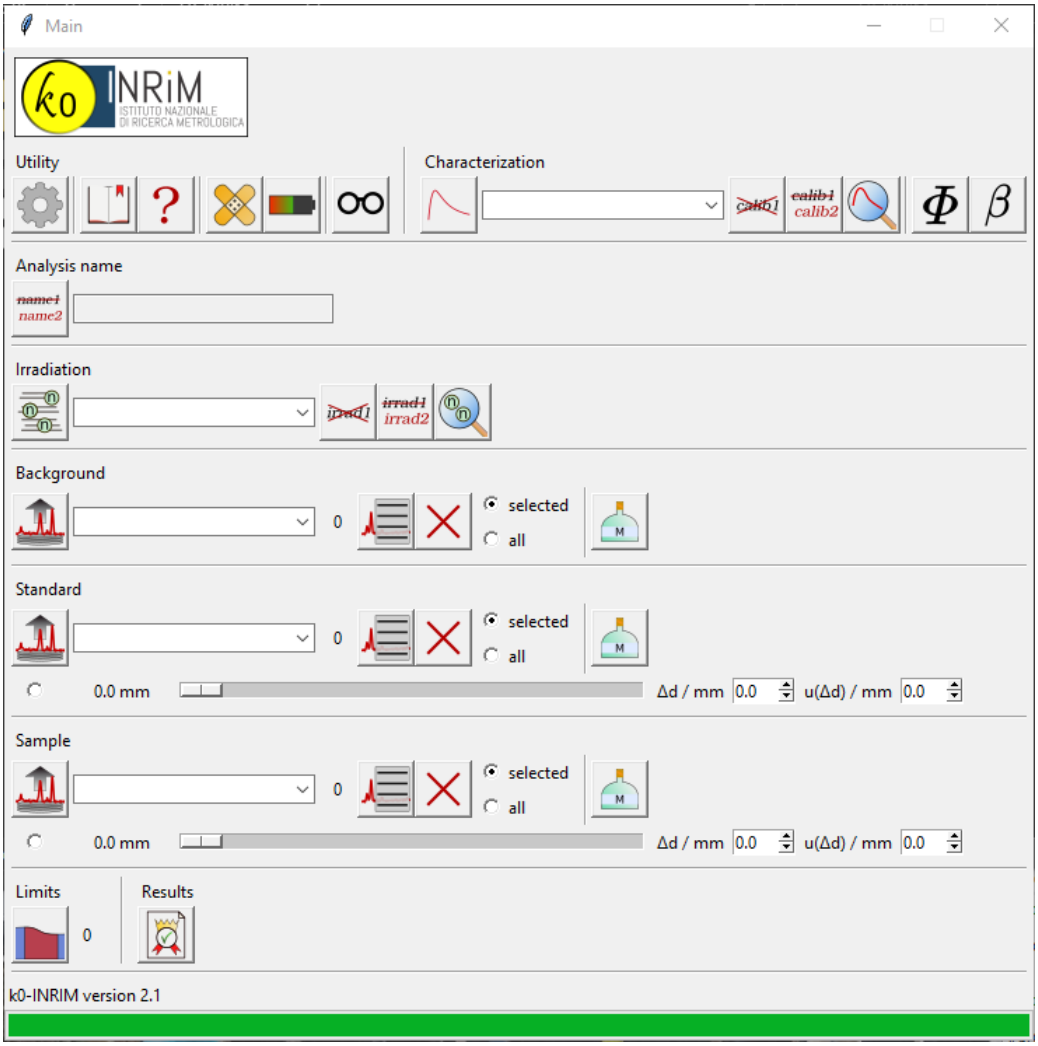


Figure 2: The main window of k0-INRIM software

3.2 Utility

This section provides buttons for *Settings*, *Browse databases*, *Credits*, *Save*, *Load* and *Predict experimental setup*.

3.2.1 Settings

The button allows to set the general behavior of the software (Figure 3). Any change, if confirmed, will automatically make the current settings effective by restarting the program.

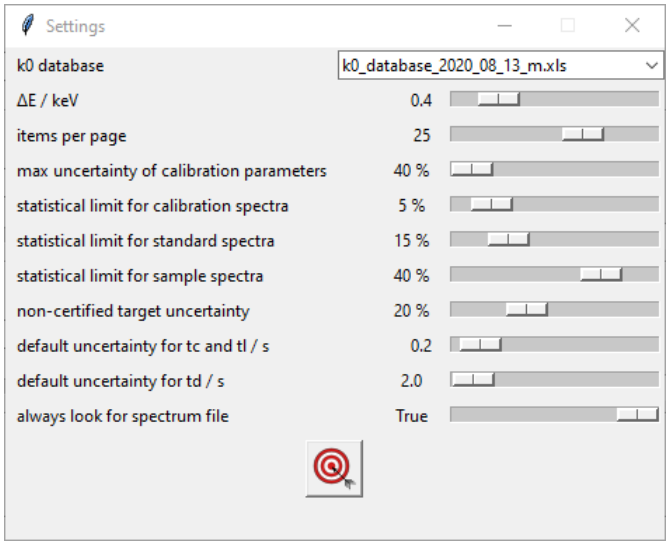


Figure 3: The settings window

The first setting is the k_0 database version used to retrieve nuclear data. In the case of release of an updated version, save the new file in the «k0data» folder and replace the current selection with the new filename that appears in the drop-down menu.

The k_0 database installed by default is the most up to date version at the time of writing [7] with a couple of minor but necessary changes. Details of these changes are given in the «README_k0_database.txt» file in the «k0data» folder.

The second setting is the energy tolerance, ΔE, used to identify the gamma peaks of the spectra by comparison with the emission energies reported in the k_0 database, E_p. The software searches for entries satisfying the relationship: $E_\mathrm{p} - \Delta E < E_\mathrm{meas} < E_\mathrm{p} + \Delta E$, where E_meas is the energy of the measured peak. The ΔE value shouldn't be neither too low or too high. In the former case the identification fails if the energy calibration is affected by minor biases while in the latter case multiple correspondences are returned. The default 0.3 keV value might be a reasonable compromise. If some mis-recognition is spotted it might depend from some energy bias present in the imported elaborated spectra, thus, a check of those imported files is always suggested prior to starting the elaboration, alternatively, an enlargement of the ΔE setting might be a viable option.

The third setting affects the visual aspect of the peak list window (see paragraph 3.6) by adjusting the maximum number of listed peaks to appear in the screen.

The fourth setting defines the maximum allowed relative uncertainty for the parameters, a_i, modeling the detection efficiency (see paragraph 3.3).

The following three settings define the threshold maximum allowed peak statistical relative (percent) uncertainty while importing a spectrum peak-list; these settings are independent for calibration, standard and sample spectra, respectively.

The following one sets the uncertainty of those elements for which certified mass fraction values are not available.

The following two settings define the default values for the standard uncertainties of the input quantities t_c and t_l (this also affects Δt_d) and t_d (here and hereafter, see the equation model 35 for the adopted symbols of the input quantities).

The last one sets whether looking for the spectrum file (in .chn or .asc format) while importing peaklists; the spectrum file includes information on the spectrum profile (to display the spectrum or for detection limits calculations) and real, live and start acquisition times. If the spectrum file is required by the software (as while importing peaklists of .csv format) it will be imported anyway regardless of the selection for this setting.

The *Confirm* button accepts the changes and restarts the program.

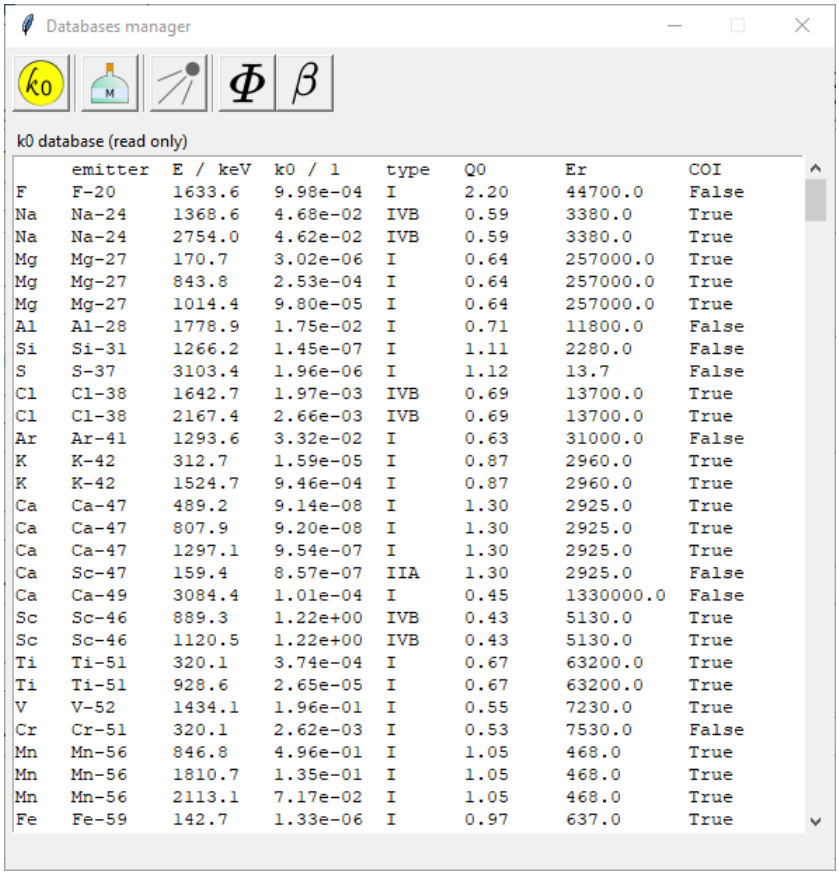
3.2.2 Browse database

The button opens the Databases manager subwindow offering the possibility to display and modify useful data to be used in the analysis. In particular, information about k_0 database, investigated samples, reference γ sources and irradiation facilities can be recalled here from the corresponding buttons: *k0 database*, *material database*, *source database*, *flux evaluation history database* and *beta evaluation history database*. These buttons change the behavior of the Databases manager subwindow by showing different pages with the default being the k0 database.

The button displays a read only window showing entries of the k_0 database currently in use (Figure 4). For each line of the database target element, emitter, emission energy, k_0 value, activation/decay type, Q_0 value, $\bar{E}_r$ value and occurrence of true-coincidences are showed.

The button displays information about materials used in the analysis and stored in the «samples» folder. Material name and mass fractions (and corresponding standard uncertainties) of constituent elements are assigned, recalled or modified together with optional information such as description, type and physical state (Figure 5).

At the bottom of the window, *add a new material*, *update changes to this material*, *delete material* buttons allow to perform the corresponding actions. On the right side the element drop-down menu and two entry fields enable to select a specific element (ordered by Z number) and assign mass fractions and corresponding standard uncertainties, respectively. The *update*



	emitter	E / keV	k0 / 1	type	Q0	Er	COI
F	F-20	1633.6	9.98e-04	I	2.20	44700.0	False
Na	Na-24	1368.6	4.68e-02	IVB	0.59	3380.0	True
Na	Na-24	2754.0	4.62e-02	IVB	0.59	3380.0	True
Mg	Mg-27	170.7	3.02e-06	I	0.64	257000.0	True
Mg	Mg-27	843.8	2.53e-04	I	0.64	257000.0	True
Mg	Mg-27	1014.4	9.80e-05	I	0.64	257000.0	True
Al	Al-28	1778.9	1.75e-02	I	0.71	11800.0	False
Si	Si-31	1266.2	1.45e-07	I	1.11	2280.0	False
S	S-37	3103.4	1.96e-06	I	1.12	13.7	False
Cl	Cl-38	1642.7	1.97e-03	IVB	0.69	13700.0	True
Cl	Cl-38	2167.4	2.66e-03	IVB	0.69	13700.0	True
Ar	Ar-41	1293.6	3.32e-02	I	0.63	31000.0	False
K	K-42	312.7	1.59e-05	I	0.87	2960.0	True
K	K-42	1524.7	9.46e-04	I	0.87	2960.0	True
Ca	Ca-47	489.2	9.14e-08	I	1.30	2925.0	True
Ca	Ca-47	807.9	9.20e-08	I	1.30	2925.0	True
Ca	Ca-47	1297.1	9.54e-07	I	1.30	2925.0	True
Ca	Sc-47	159.4	8.57e-07	IIA	1.30	2925.0	False
Ca	Ca-49	3084.4	1.01e-04	I	0.45	1330000.0	False
Sc	Sc-46	889.3	1.22e+00	IVB	0.43	5130.0	True
Sc	Sc-46	1120.5	1.22e+00	IVB	0.43	5130.0	True
Ti	Ti-51	320.1	3.74e-04	I	0.67	63200.0	True
Ti	Ti-51	928.6	2.65e-05	I	0.67	63200.0	True
V	V-52	1434.1	1.96e-01	I	0.55	7230.0	True
Cr	Cr-51	320.1	2.62e-03	I	0.53	7530.0	False
Mn	Mn-56	846.8	4.96e-01	I	1.05	468.0	True
Mn	Mn-56	1810.7	1.35e-01	I	1.05	468.0	True
Mn	Mn-56	2113.1	7.17e-02	I	1.05	468.0	True
Fe	Fe-59	142.7	1.33e-06	I	0.97	637.0	True

Figure 4: The k0 database window

element's value and *delete element's value* buttons perform the corresponding actions on the selected element. The g g-1 button switches to ppm and % mass fraction values.

A new material is created by pressing the *add a new material* button and information are not saved on disc until the *update changes to this material* button is pressed.

The button displays information about γ reference sources adopted in the characterization tasks and stored in the «sources» folder. Source name, reference date and γ-emitting radionuclide information are assigned, recalled or modified (Figure 6). Moreover, the gamma source database window allows to combine stored sources in a single (virtual) source, with a common reference date, useful for following detector characterization processes.

At the bottom of the window, *add a new source*, *update changes to selected source*, *delete the selected source* buttons allow to perform the corresponding actions. Just below, there are two sections divided by a vertical separator: on the left section emitters can be selected and added from an internal database while on the right section single emission information can be managed.

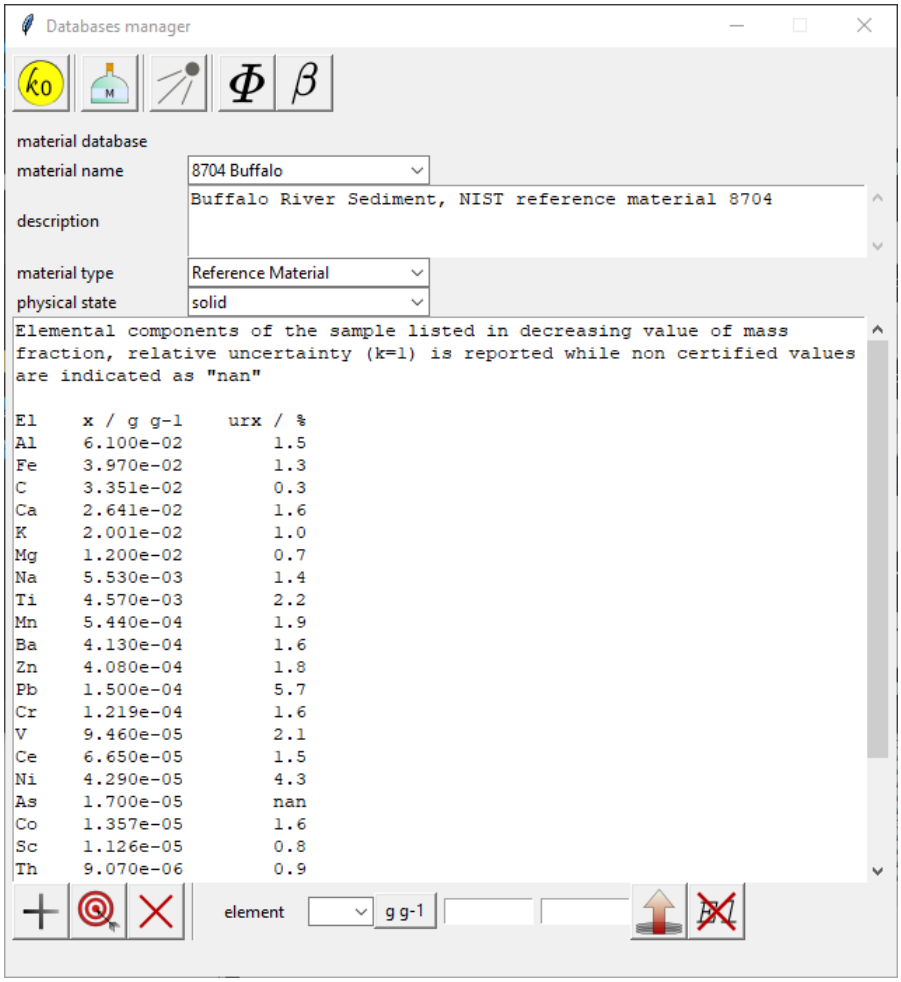


Figure 5: The material database window

A new source is created by pressing the *add a new source* button and information are not saved on disc until the *update changes to selected source* button is pressed.

Once a new source is created, the name is assigned in the corresponding drop-down menu, reference date is set by pressing the button; the reference date is confirmed with the *confirm new date* button in the dedicated sub-window (Figure 7).

From the emitter drop-down menu a radionuclide can be chosen and added to the selected source by pressing the *add or update an emitter* button after setting the activity and checking the half-life in the corresponding entry fields.

A single γ-emission can be selected using the drop-down menu and deleted by pressing the *delete emission* button in the right section. γ-yield and COIfree status (if checked means that the emission is considered as coincidence-free during the detector characterization) of the selected γ-emission can be modified using the corresponding entry field and checkbox, respectively. Changes must be

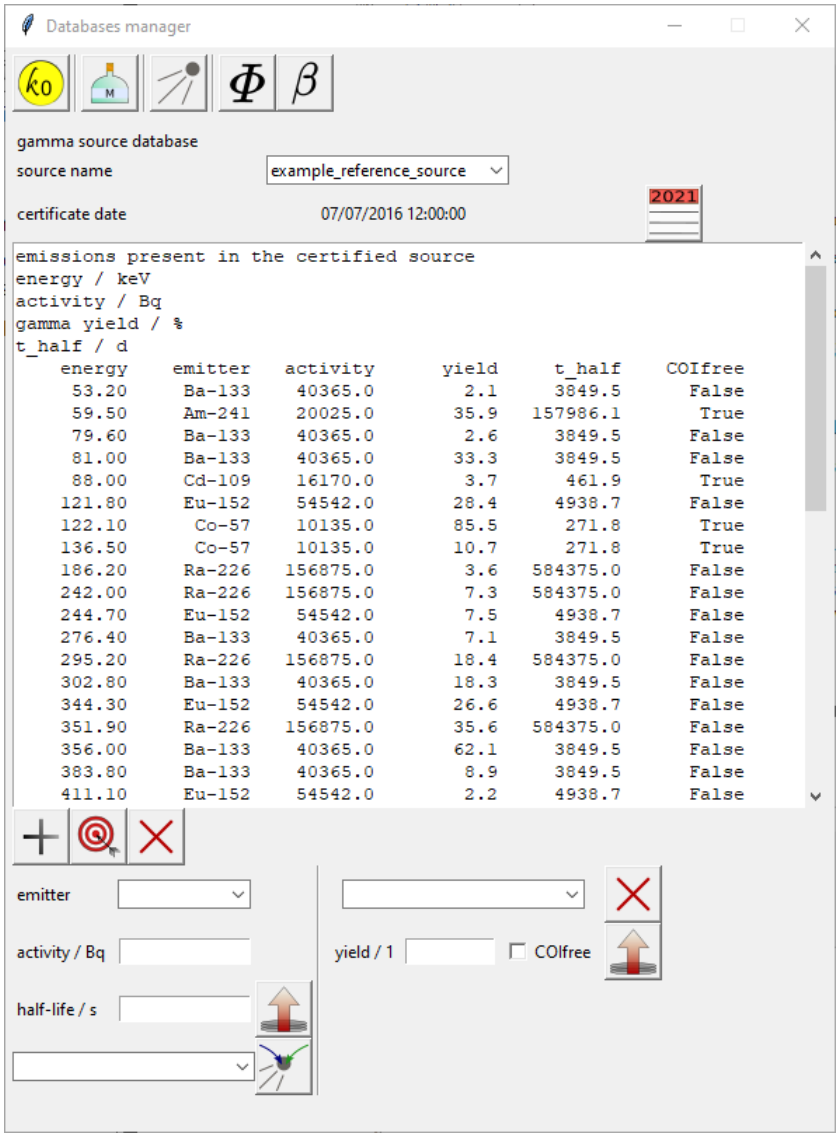


Figure 6: The γ reference source database window

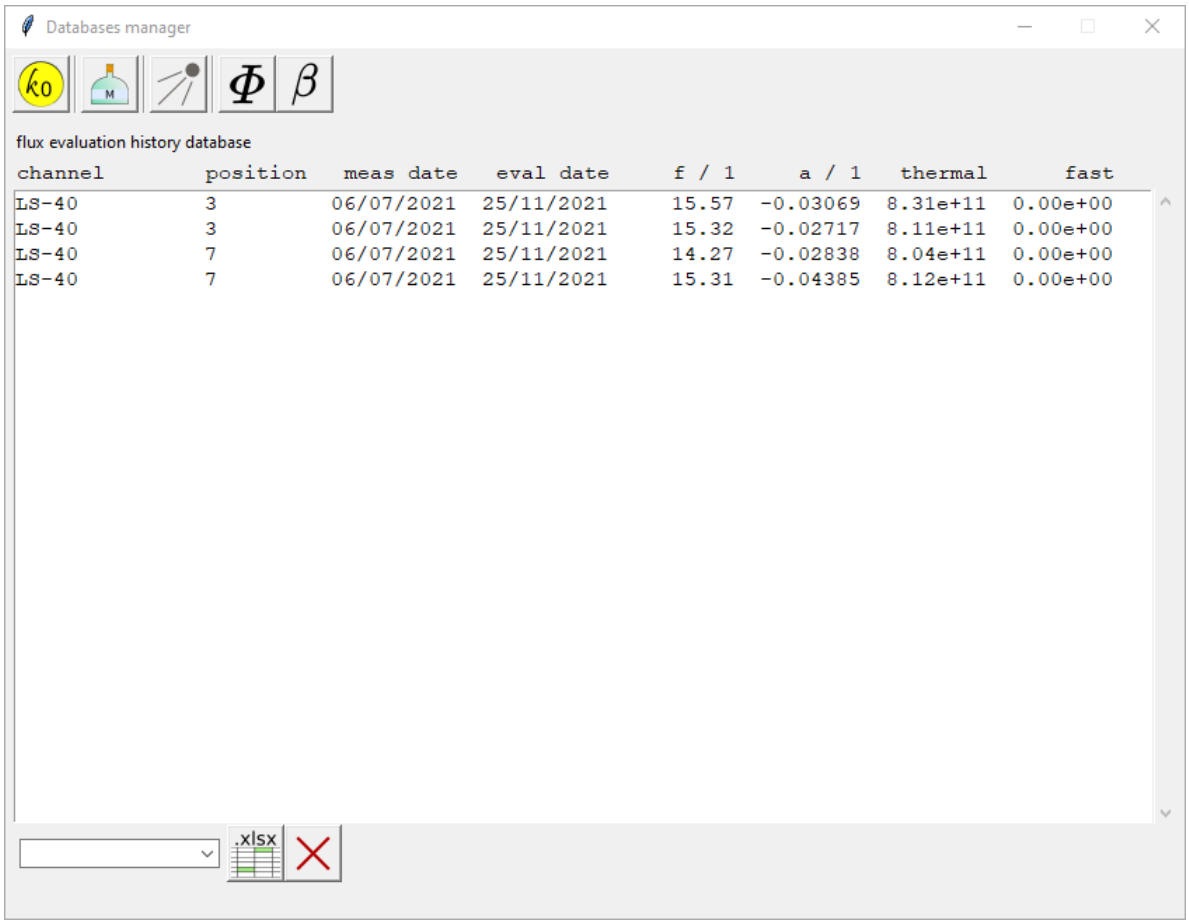
confirmed by pressing the *update data for the current emission* button in the bottom right section.

In case different γ-sources are used to characterize the detector, a virtual γ-source must be added to the database by pressing the *add a new source* button; it is suggested to assign a reference date close to the acquisition of the γ-sources having the shortest half-lives to avoid unreasonable activity values. To combine different γ-sources select an existing source in the database from the *merging source name* drop-down menu and confirm by pressing the *merge with selected source*

Figure 7: The change reference date sub-window

button. The emissions of the merging source are added to the virtual source and the activities of the radionuclides are referred to the reference date.

The Φ button displays information about characterizations performed on the irradiation facilities (see paragraph 3.3.5); those data are stored in the «facility» folder in a file called «channel.csv». For each entry the channel name, position within the channel, measurement date, evaluation date, f value, α value, conventional thermal flux and fast flux values are reported (Figure 8).



channel	position	meas date	eval date	f / 1	a / 1	thermal	fast
LS-40	3	06/07/2021	25/11/2021	15.57	-0.03069	8.31e+11	0.00e+00
LS-40	3	06/07/2021	25/11/2021	15.32	-0.02717	8.11e+11	0.00e+00
LS-40	7	06/07/2021	25/11/2021	14.27	-0.02838	8.04e+11	0.00e+00
LS-40	7	06/07/2021	25/11/2021	15.31	-0.04385	8.12e+11	0.00e+00

Figure 8: The flux evaluation history database window

At the bottom of the window the selection can be filtered by channel name and then saved to an Excel file, or selected entries can be deleted with the corresponding buttons, *export selection as .xlsx file* and *delete selected measurement*, respectively.

The β button displays information about characterizations performed on the irradiation facilities (see paragraph 3.3.6); those data are stored in the «facility» folder in a file called «beta.csv». For each entry the position within the channel, measurement date, evaluation date, β value, $u(\beta)$ value are reported (Figure 9).

At the bottom of the window the selection can be filtered to show exclusively β values referring

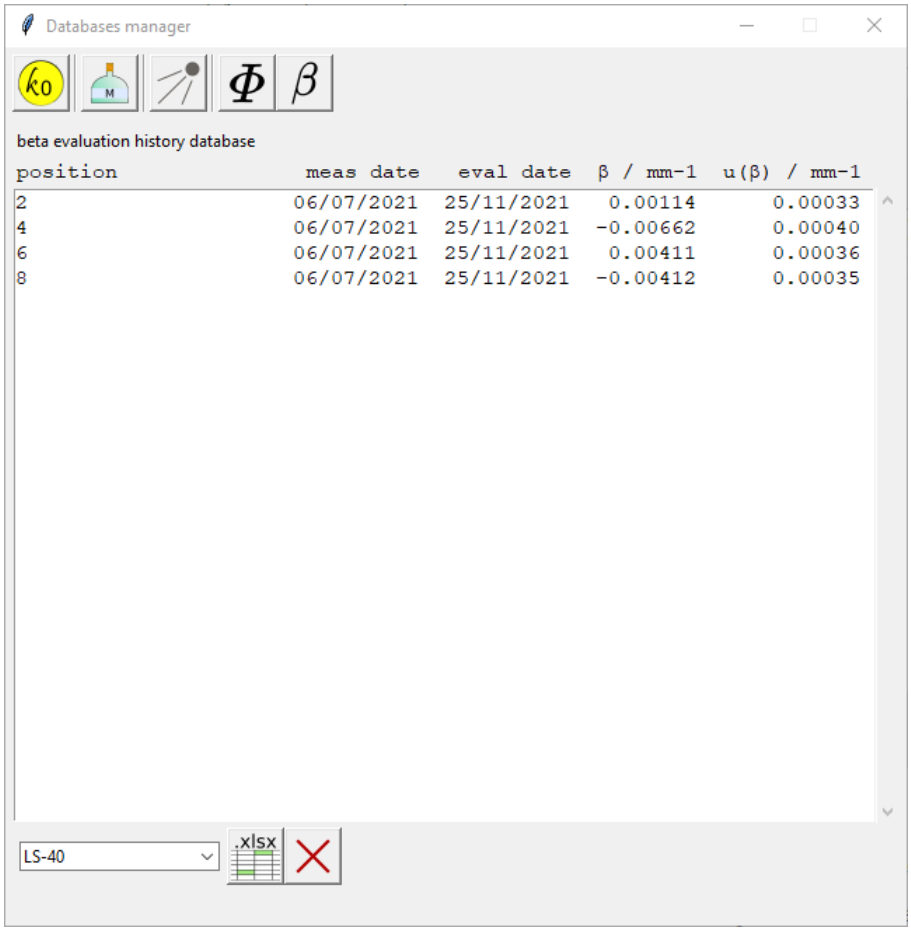


Figure 9: The flux evaluation history database window

to the specified channel name; the whole selection can then be saved to an Excel file, or selected entries can be deleted with the corresponding buttons, *export selection as .xlsx file* and *delete selected β measurement*, respectively.

3.2.3 Credits

This subwindow includes information about the version of the k0-INRIM used, contact emails and reference to scientific literature; moreover, it displays a copy of the GNU GENERAL PUBLIC license shipped with the software.

3.2.4 Save

The button allows to save the current state of the program. It opens a subwindow (Figure 10) allowing to choose the filename, whether include the date and a further button to perform

the save.

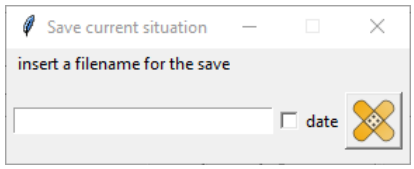


Figure 10: The save data sub-window

If an analysis name is already been introduced (see section 3.4) it will be the suggested filename for the save.

3.2.5 Load

The button allows to restore the k0-INRIM to a previously saved state (settings included). It opens a subwindow (Figure 11) allowing to select one of the previously saved files from the drop-down menu and, once selected recall it with the *recall saved file* button or delete it with the *delete saved file* button.

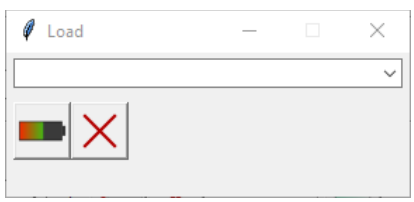


Figure 11: The load data sub-window

By pressing either button a confirmation subwindow will open requiring the user to confirm the choice. It is worth to note that recalling the saved file will restart a new instance of the program while losing all unsaved activity. Recalling a saved file also recalls all the values for settings adopted when the save was performed with exception of the number of listed peaks that will remain unchanged since it is just a visual option.

The drop-down menu in this window looks for .sav files stored in «saves» folder within «data» of the main k0-INRIM folder; it is possible to recall specific savefiles (also form a different pc) just by moving the suitable .sav file to the «saves» folder and select it via the drop-down menu after restarting the Load window.

3.2.6 Predict experimental setup

The button allows to predict the expected emissions' count rate for a defined experimental setup. It opens a subwindow (Figure 12) allowing to identify the suitable conditions for the following analysis based on knowledge deriving from previous facility and detector characterizations.

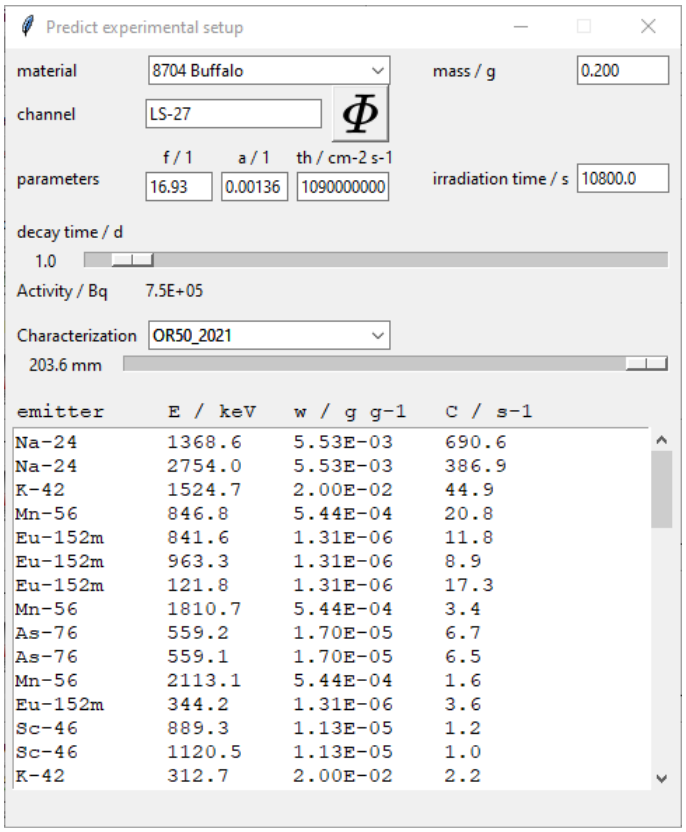


Figure 12: The predict experimental setup sub-window

The drop-down menu next to the label `'material'` allows to choose the material, among those stored in the «samples» folder in «data», for which count rate values will be returned; the corresponding mass has to be inserted in the entry next to the label `'mass / g'`. Irradiation information have to be entered in the entries next to the labels `'channel'`, `'parameters'` and `'irradiation time / s'`; the `'parameters'` section provides entries for typing f, α and the Hogdahl conventional thermal flux values, respectively. Alternatively, with the button it is possible to recall information for one of the facilities saved in the *flux history database* subwindow in the *Browse database* section. The cooling time before γ-counting might be selected by scrolling the slider below the label `'decay time / d'` within the range 0-20 days with 0.1 day steps. The adopted detector characterization and the counting distance can also be selected through the drop-down menu labeled `'characterization'` and slider underneath it; if no detector characterization is selected, efficiency equal to 1 is assumed.

After declaration of all required input, the text box at the bottom of the window displays the

predicted count rate after given cooling time for any emission expected to be produced from the material in the selected experimental conditions; in particular, each line gives information about the emitter, the corresponding emission energy, the mass fraction within the material and the calculated count rate. Additionally, a rough estimate of the expected total activity is reported next to the label `'Activity / Bq'`.

The count rates are obtained through application of the INAA absolute measurement model relying on k_0 database data:

$$C(t_\mathrm{d}) = m\, w\, k_{0\,\mathrm{Au}}(n)\, N_k\, \Phi_\mathrm{th} \left(1 + \frac{1}{f}\left(\frac{Q_0 - 0.429}{\bar{E}_\mathrm{r}^{\alpha}} + \frac{0.429}{(2\alpha+1)\,0.55^{\alpha}}\right)\right)\left(1 - \mathrm{e}^{-\lambda\, t_\mathrm{i}}\right)\mathrm{e}^{-\lambda\, t_\mathrm{d}}\, \varepsilon, \quad (1)$$

where $C(t_\mathrm{d})$ is the count rate evaluated at cooling time t_d, m is the mass of irradiated material, w is the mass fraction of the investigated element, $k_{0\,\mathrm{Au}}(n)$ is the k_0 value for the specific emission, $N_k = \frac{N_\mathrm{A}\,\Gamma_\mathrm{Au}\,\sigma_{0\,\mathrm{Au}}\,\theta_\mathrm{Au}}{M_\mathrm{Au}} = 0.2885$ is a composite constant based on literature values of parameters referred to gold (gamma emission rate, $2200\,\mathrm{m\,s^{-1}}$ capture cross section, isotopic fraction and molar mass) and Avogadro constant, Φ_th is the conventional thermal neutron flux, f is the thermal to epithermal flux ratio, Q_0 is the resonance integral to thermal cross section ratio, $\bar{E}_r$ is the effective resonance energy, t_i is the irradiation time and ε is the counting efficiency.

Count rates obtained from this calculation are approximated since multiple corrections are neglected (self-shieldings, self-absorption, gradients etc) and need to be considered just as a guideline to choose the suitable experimental setup. Moreover, since the k_0-database is used to gather literature information a limited number of reaction are considered: only delayed (n,γ) leading to direct decays, thus, the value reported in the label accounting for the total activity is likely to be underestimated.

3.3 Characterization

This section provides tasks to manage characterization of detection efficiency and irradiation facilities. It provides a *New detector characterization* button, a drop-down menu to recall a previously saved detector characterization, three *Delete detector characterization*, *Rename detector characterization*, *Display detector characterization* buttons to perform corresponding actions on the selected detector characterization, and two *Flux evaluation* and *Gradient evaluation* buttons to perform measurements on the irradiation facility.

The detector characterization drop-down menu looks for a triplet of files (.pos, .pkl and .txt, while only .pos is mandatory it is suggested to have them all to recall full information) stored in

«characterization» folder within «data» of the main k0-INRIM folder; it is possible to recall specific detector characterizations obtained with the same software version (also from a different pc) just by moving the suitable files to the «characterization» folder and select it via the drop-down menu; in order to make the change effective it is required to restart the software.

3.3.1 New detector characterization

The *New detector characterization* task involves the creation of a new instance of detector characterization where data about detection efficiencies and other useful information are processed. The detector characterization consists of acquisitions of reference γ-sources in all counting positions. In order to benefit of the full potential of this procedure at least four different positions need to be used (acquiring all emissions in the farthest positions and only coincidence-free emissions in the others) otherwise some functionality, as corrections for vertical displacement and sample-geometry, will be missing.

The detector characterization window appears by pressing the button in the main window (Figure 13).

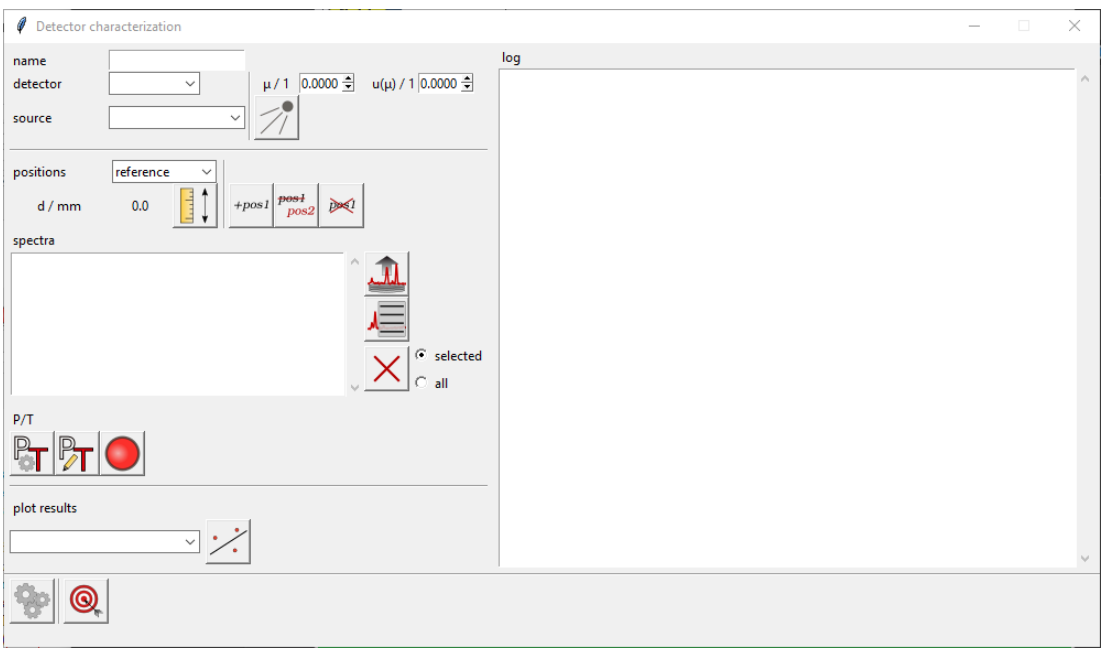


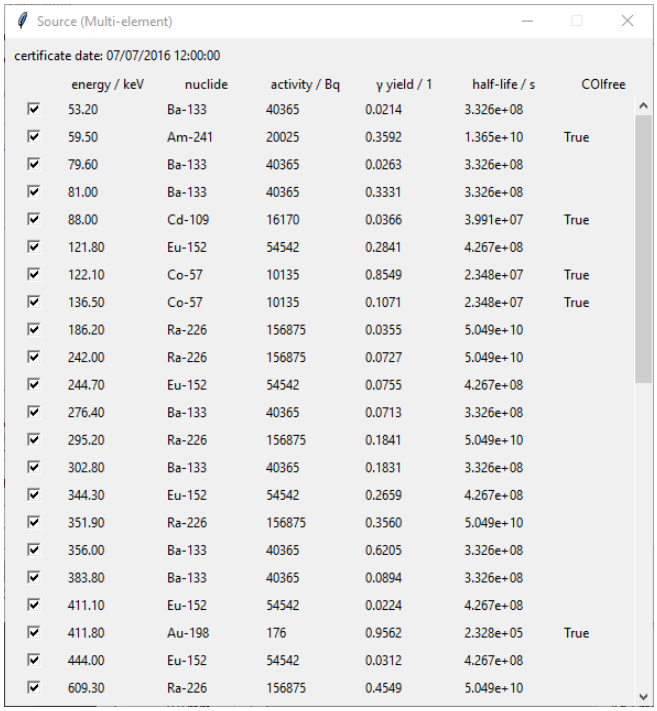
Figure 13: The new detector characterization window

The window is composed by some sections on the left hand side divided by horizontal separators. At the top of the window, information about the chosen name for the detector characterization, detector information and data about the γ source are required.

The detector name can be chosen by the drop-down menu which keeps in memory all previously

inserted detector information, moreover, values and uncertainty of excess counting loss constant of the detection system, denoted as μ, are required; these information are also stored (in the «detlist.csv» file present in the «facility» folder).

The source drop-down menu allows to select the γ source to be used in the characterization, the source should represent all the emissions adopted for the task even if they come from different reference sources, and should be created accordingly in the *gamma source database* subwindow in the *Browse database* section (3.2.2). Gamma emissions from the source can be included or excluded for the incoming characterization by selecting them through the button. The selection emissions subwindow (Figure 14) displays the name and reference date of the source and lists all the emissions included in it; each emission line provides information about energy, nuclide identity, activity, gamma-yield, half-life, whether it is considered a coincidence free emission and has a checkbutton that can be selected or deselected: if it's deselected that emission will be ignored in the following calculations.



Source (Multi-element)

certificate date: 07/07/2016 12:00:00

	energy / keV	nuclide	activity / Bq	γ yield / 1	half-life / s	COIfree
☑	53.20	Ba-133	40365	0.0214	3.326e+08	
☑	59.50	Am-241	20025	0.3592	1.365e+10	True
☑	79.60	Ba-133	40365	0.0263	3.326e+08	
☑	81.00	Ba-133	40365	0.3331	3.326e+08	
☑	88.00	Cd-109	16170	0.0366	3.991e+07	True
☑	121.80	Eu-152	54542	0.2841	4.267e+08	
☑	122.10	Co-57	10135	0.8549	2.348e+07	True
☑	136.50	Co-57	10135	0.1071	2.348e+07	True
☑	186.20	Ra-226	156875	0.0355	5.049e+10	
☑	242.00	Ra-226	156875	0.0727	5.049e+10	
☑	244.70	Eu-152	54542	0.0755	4.267e+08	
☑	276.40	Ba-133	40365	0.0713	3.326e+08	
☑	295.20	Ra-226	156875	0.1841	5.049e+10	
☑	302.80	Ba-133	40365	0.1831	3.326e+08	
☑	344.30	Eu-152	54542	0.2659	4.267e+08	
☑	351.90	Ra-226	156875	0.3560	5.049e+10	
☑	356.00	Ba-133	40365	0.6205	3.326e+08	
☑	383.80	Ba-133	40365	0.0894	3.326e+08	
☑	411.10	Eu-152	54542	0.0224	4.267e+08	
☑	411.80	Au-198	176	0.9562	2.328e+05	True
☑	444.00	Eu-152	54542	0.0312	4.267e+08	
☑	609.30	Ra-226	156875	0.4549	5.049e+10	

Figure 14: The emission source selection window

The emission selection is kept in memory until another source is selected.

Below the left top section, a region requiring information about distances in which the characterization is performed and the acquired spectra from the source, together with tools to perform a Peak-to-Total (PT) evaluation is present.

The drop-down menu next to the label `'position'` allows to select a specific counting position; all actions taken in this section refer to the currently selected counting position. The reference

position, entered by default, represents the farthest counting position and its presence is mandatory.

The value next to the label `'distance'` indicates the distance from the detector end-cap of the current position; it is 0 by default but can be changed by pressing the *Modify distance* button. This button will open a subwindow to type the new value and validate it by pressing enter from the keyboard.

The following

Add a counting position,

Rename a counting position and

Delete a counting position buttons perform the corresponding actions. There is no limit to how many positions can be set, however, if no spectra or no emissions are found in a particular position it will be excluded from processing. Also, reference position is the only one that cannot be renamed or deleted.

The listbox below the label `'spectra'` includes the filename of the spectra that are acquired at the current position (i.e. at the same distance from detector end-cap defined by the label `'d / mm'`).

The *Add spectra to the current counting position* button allows to select spectra to include in the current position; valid input are peak lists (in .csv file format) from HyperLab elaborated spectra or reports (in .rpt file format) obtained from GammaVision. When one of these file types is selected the software proceeds to open it to gather the required information about gamma peaks, in the meanwhile the software also looks for a file in the same folder with same name but different type (.asc or .chn) in order to get information concerning the complete gamma spectrum and acquisition times. In case .asc or .chn spectrum is not found an error is raised (although, for compatibility reasons this behavior can be modified for .rpt files in the settings window through the option `'always look for spectrum file'` (Figure 3)). The filenames of correctly recalled spectra will be visible in the listbox.

The *Show the peaklist of selected spectrum* button allows to browse information about the currently selected spectrum (if no spectrum is selected the first one of the list is automatically open, if any). The peaklist window shows information about the directory where the file is located, and the name of files uploaded and whether they are identified as peak list (.csv or .rpt files) or spectrum (.asc or .chn files), the start acquisition time, real, live and dead time, together with the list of all γ-peaks reported in the peaklist (Figure 15).

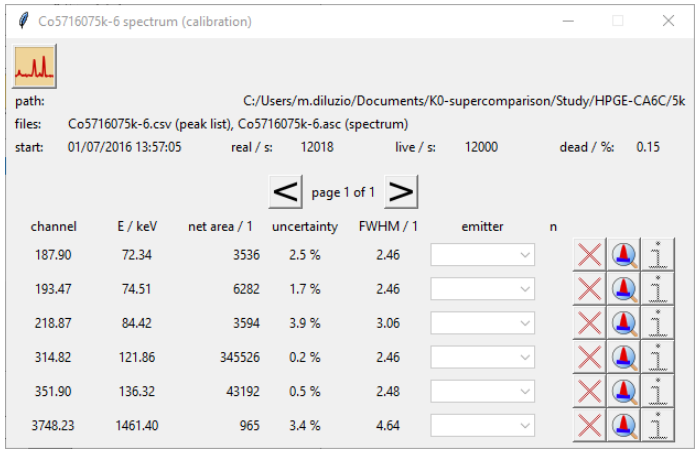


Figure 15: The peaklist window

The *Show spectrum profile* button, placed at the top-left of the window, allows to open a visualization of the spectrum profile (Figure 16), if any spectrum filetype was found when recalling the spectra.

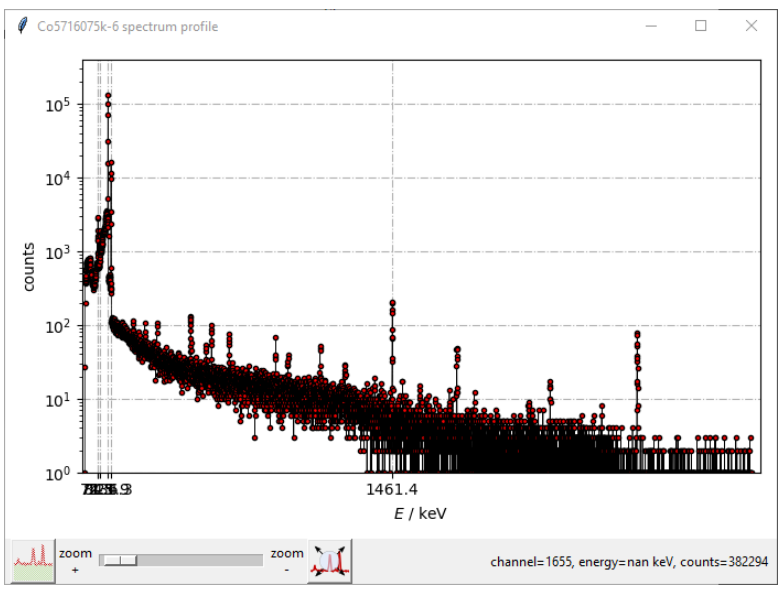


Figure 16: The spectrum window

The spectrum window displays a plot of spectrum profile with energies (in keV) on x-axis and logarithm of counts in the y-axis. If selected from the main window (see paragraph 3.6.1), a superimposed background is visualized (or hidden) by pressing the *Show background profile* button; the background profile, already scaled to the spectrum live time, is drawn as a green line. It is possible to navigate the spectrum profile plot by scrolling with the mouse-wheel; a zoomed-in visualization will be automatically prompted while the zoom level is adjusted by the corresponding slider between the `'zoom +'` and `'zoom -'` labels. The *Zoom y-axis* button adjusts the y-

axis to the current content while navigating through the spectrum profile. At the bottom-right of the window, information such as channel, energy and counts are displays depending on mouse position over the plot; a characterization should be selected in the main window for the energy to be shown.

Back to the peaklist window, the < *Previous page* and > *Following page* buttons browse the peaklist in order to display all the peaks since the visualization is split in pages. Each page includes a number of lines selected in the settings menu (see paragraph 3.2.1). Each line provides information or actions referred to the corresponding peak; in particular, the information are the centroid of the peak, the energy at given centroid, the net area with relative uncertainty, FWHM (in channels) and the number of correspondences found in k_0 database for that energy; those are reported below the labels `'channel'`, `'E / keV'`, `'net area / 1'`, `'uncertainty'`, `'FWHM / 1'` and `'n'` respectively. Actions allow to select a specific emission from those suggested by the software using the drop-down menu below the label `'emitter'`, clear the selection, visualize the peak in the spectrum profile and get more information on the selected emission via buttons. When dealing with calibration spectra the action *Go to info* is disabled and the emitter selection drop-down menus give the possibility to select emissions from the selected source certificate (and not from the k_0-database). The button clears the selection and the button promptly adjusts the x-axis of the spectrum profile plot to center the corresponding peak.

Back to the Detector characterization window, the spectra appearing in the spectra listbox can also be deleted from the corresponding position by the *Delete the selected spectra* button. The selector next to it allows to choose if only the selected spectrum should be deleted or all spectra of the current position; in both cases, a confirmation subwindow will be prompted. This action does not erase from disc the original files of peak lists and spectra.

At the bottom of this section the PT region is found. Here, three buttons are present to perform actions on PT data: *Evaluate PT for the current position*, *Insert PT values for the current position* and *Display PT values for the current position*.

PT is a key component to calculate coincidence corrections for closer counting positions; in the instance of the k0-INRIM software it is optional to perform a PT evaluation and its computation is, to some extent, independent from the rest of detector characterization meaning that a different reference source might be used with completely different emissions and γ-source spectra; due to this particularity, it is always suggested to perform the PT evaluation after all counting positions are defined but before pressing the button to process the regular detector characterization. PT evaluation is achievable by pressing or button. In the first case an automatic evaluation based on acquisition of true-coincidence emissions is performed.

A subwindow will open requiring the introduction of all the needed information such as the background and true-coincidence free emissions spectra acquired at current position (Figure 17).

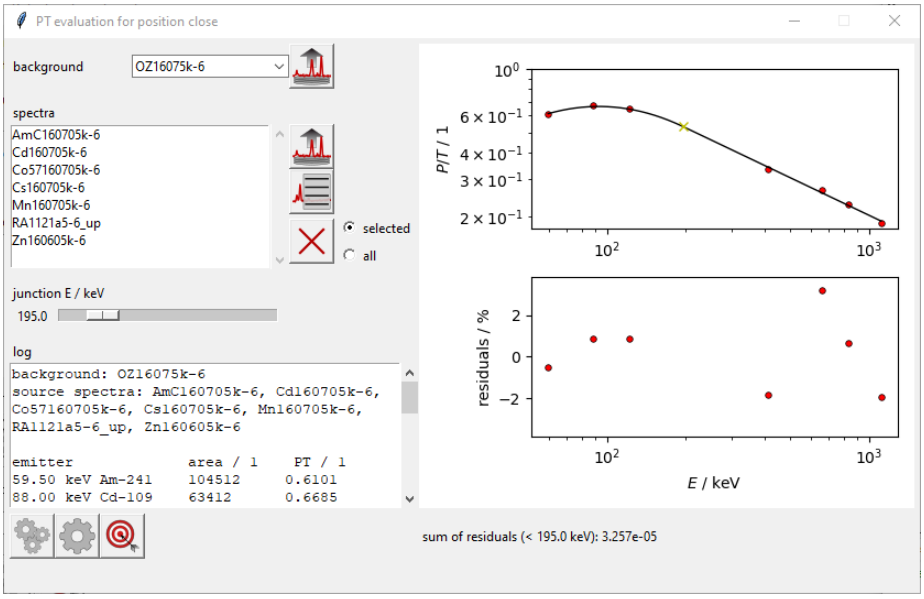


Figure 17: The PT evaluation window

Management of spectra is done with the usual buttons (, , and). It is worth to note that if a background is already selected in the main window, as well as reference emission spectra in the current position, they are automatically recalled in the PT evaluation window. The slider below the label `'junction E / keV'` indicates the energy where the software will set the discontinuity between the two equations used to draw the PT curve; in particular, below said point a polynomial exponential equation is adopted while above that point a linear exponential equation is adopted, respectively [3] (eq. 2),

$$PT = \begin{cases} 10^{p_1(\log_{10} x)^2 + p_2 \log_{10} x + p_3} & \forall x : 0 < x \leq E_{\text{junction}} \\ 10^{s_1 \log_{10} x + s_2} & \forall x : x > E_{\text{junction}} \end{cases} \tag{2}$$

While x represents the energy of emission, parameters p_1, p_2 and p_3 define the polynomial fit and parameters s_1 and s_2 define the straight line fit, respectively.

The log box in the lower part of the window and graphs at the right-hand side will display useful information while the evaluation process is started.

Once all the required information are introduced, the click of button starts the PT evaluation; the process inquires the presence of the background spectrum and checks whether the number of channels corresponds to that of the reference emission spectra, then, reads the reference source file to retrieve information related to the true-coincidence free emissions and looks for the presence of corresponding γ-peaks in the acquired spectra. The emission selection is performed automatically by the software unless a label is specified by the user in the corresponding peaklist. For each found

emission the software gathers the information of net peak area from peaklist and calculates the total spectrum area, T, by subtracting the integral of the live time corrected background to the integral of spectrum profile, eq. 3:

$$T = \sum_{i=\text{channel}_0}^{\text{channel}_n} counts_{\text{src}}(i) - \frac{t_{\text{l src}}}{t_{\text{l bkg}}} counts_{\text{bkg}}(i), \tag{3}$$

with index i spanning over the number of spectra channels for the emitter source (subscript src) and background (subscript bkg), respectively, with the latter adjusted to comply the live time of the former. On top of that a further correction is also introduced at low channel numbers in order to take into account the lower energies being filtered by the discriminator; thus, a constant extrapolation until 0 energy is performed taking as reference the maximum count value among the first 20 channel with non-zero counts. Finally, in order to avoid non-physical behaviors all corrected channels showing a negative count are set to 0.

At this point, the dataset which is composed by energies and PTs (net area of peak divided by total spectrum profile) is split depending of the selected E_{junction}; data coming from energies greater than E_{junction} are fitted with the corresponding linear fitting function (eq. 2). Once parameters s_1 and s_2 are obtained, the remaining part of dataset is fitted by minimizing the sum of residual function, σ_{res}, performed on the logarithm of first function in eq. 2:

$$\sigma_{\text{res}} = \sum \left(y_{\text{exp}} - p_1 \left(\log_{10} x\right)^2 + p_2 \log_{10} x + p_3 \right)^2, \tag{4}$$

where, y_{exp} represents the experimental PT values.

On top of that, a couple of constraints are added to the minimization process in order to assure continuity around the E_{junction} point; to this aim the first derivative of the polynomial fitting equation is forced equal to the slope of the linear fitting equation and equality of y between the two fits is imposed at $x = E_{\text{junction}}$ point (eq. 5).

$$\begin{cases} 2p_1 \log_{10} E_{\text{junction}} + p_2 - s_1 = 0 \\ p_1 \left(\log_{10} E_{\text{junction}}\right)^2 + p_2 \log_{10} E_{\text{junction}} + p_3 - \left(s_1 \log_{10} E_{\text{junction}} + s_2\right) = 0 \end{cases} \tag{5}$$

Whether a successful elaboration is completed a textual log will appear in the log box together with plots of experimental points, fitting curves and residuals in the graphics at the right-hand side. Moreover, a value identified as sum of residuals, including only data with energy below E_{junction}, is reported at the bottom of the graphics and is intended as an aid for the minimization of E_{junction} value itself since this task it is not performed automatically. Anytime the E_{junction} value is changed by the corresponding slider, button has to be clicked again to confirm the new value.

The button allows to manually tweak the values of the dataset, computed on the experimental data, if some mistake is suspected to be happened.

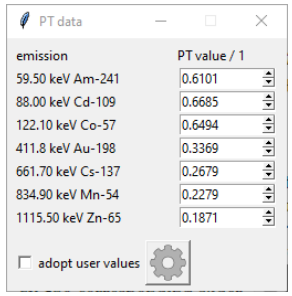


Figure 18: The window to manually tweak PT dataset

The subwindow (Figure 18) lists any energy and PT value, the latter is presented in a spinbox allowing manual modification. To make the dataset changes effective, the box with label `'adopt user values'` needs to be checked before clicking the button.

Finally the *Confirm elaboration* button saves the current PT evaluation for the corresponding counting position.

The second PT evaluation method consists in the manual introduction (or modification) of the fitting parameters. By pressing the button a subwindow appears with room to change the value of any parameter and a plot to see the outcome due to the updated values (Figure 19).

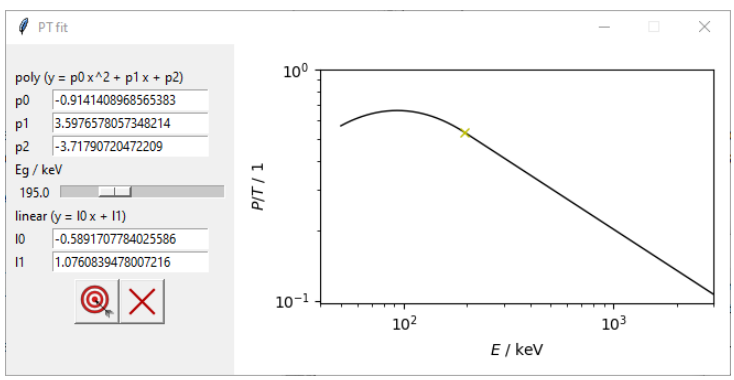


Figure 19: The window to manually tweak PT fitting parameters

The *Save PT evaluation* button confirms the changes and updates the plot while the *Delete PT evaluation* button deletes the current PT evaluation.

Independently from the evaluation method chosen, when a PT elaboration is saved for a certain

counting position the button becomes making clear that the PT is now available for said position; clicking the button shows the actual PT fitting parameters and plot similar to Figure 19 but with all options disabled. It is worth to note that the PT computation is issued without uncertainty evaluation, the uncertainty for the whole true-coincidence correction (see section 4) will be given in the budget.

The penultimate section in the Detector characterization window allows to recall plots at the various counting positions to have a glimpse of the fits, this is a useful resource in combination with the log box lying at the right-hand side of the window where textual information are reported. The selection of the plot is performed by the drop-down menu below the label `'plot results'` and the plot is visualized by clicking on the *See details of fits* button; two types of plot can be recalled, a so-called position plot which shows information (ε or $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ and d_0', see eq. 6 and eq. 35 for reference) about a single counting position and the so-called emission plot which shows information (variation of count rate depending on counting position) about a single reference source emission. Each plot is accompanied by a further graphics bringing relative residuals information.

At the bottom of the window the two *Elaborate detector characterization* and *Save the current detector characterization* buttons allow to perform the corresponding actions.

By pressing the button the elaboration starts. The first task consists of an integrity check to test whether all required information are correctly introduced. The software gathers data about the selected emissions of the reference source and looks for them in the spectra of reference position, with the discriminant being the ΔE setting in section 3.2.1. A unique peak must be assigned to the reference emission; if the routine identifies multiple peaks assignable to the same emission, the one with the lowest statistical uncertainty is automatically selected (when this happens a notification is raised with detail of the selected peak). Otherwise, if for a certain emission no suitable peaks are found in the spectra, the emission is neglected for all the following calculations.

For any emission identified in the spectra at reference counting position information concerning channel centroid, literature energy taken by the source file, FWHM, efficiency and efficiency relative uncertainty are returned. The latter two data, denoted with ε and $u_{\mathrm{r}}(\varepsilon)$, are calculated by means of experimental and literature input parameters:

$$\varepsilon = \frac{n_{\mathrm{p}}\,\lambda\,t_{\mathrm{c}}\,\mathrm{e}^{\mu\left(1-\frac{t_{\mathrm{l}}}{t_{\mathrm{c}}}\right)}}{t_{\mathrm{l}}\,\mathrm{e}^{\lambda t_{\mathrm{d}}}\,\left(1-\mathrm{e}^{\lambda t_{\mathrm{c}}}\right)\,\Gamma\,A}, \tag{6}$$

where, n_{p} is the identified net peak area, λ is the emitter decay constant, t_{c} and t_{l} are the real and live counting time, respectively, t_{d} is the decay time with respect to the certificate date, μ is the excess counting loss constant of the detection system, Γ is the emission gamma yield and A is the activity of the emitter. The relative uncertainty of ε is then assumed equal to the relative statistical uncertainty of the peak area.

Energy, FWHM and efficiency fits are calculated on data obtained in the previous process. For energy, a linear equation is adopted:

$$E = b_1\, ch + b_2, \tag{7}$$

where, b_1 and b_2 are fitting parameters and ch is the channel number. For FWHM, the square root of a linear equation is preferred:

$$\mathrm{FWHM} = \sqrt{c_1\, ch + c_2}, \tag{8}$$

where, c_1 and c_2 are fitting parameters and ch is the channel number. Efficiency data are fitted with an exponential polynomial equation [8]:

$$\varepsilon = \mathrm{e}^{a_1\, E + a_2 + a_3\, E^{-1} + a_4\, E^{-2} + a_5\, E^{-3} + a_6\, E^{-4}}, \tag{9}$$

where, a_1, a_2, a_3, a_4, a_5 and a_6 are fitting parameters and E is the energy expressed in MeV. While energy and FWHM curves only affect detection limits calculations, efficiency fit (at reference) is key for the calculation of $k_\varepsilon \Delta E$ parameter. All these fits are returned with covariance matrices for uncertainty evaluation.

Calculations on all other counting positions are performed by selecting the true-coincidence free emissions from the reference source. The coincidence free emissions are looked for in the spectra of all positions and a 2d array including emissions count rate at every position is obtained:

$$\begin{matrix} C_{0,\mathrm{ref},E_1} & C_{0,\mathrm{ref},E_2} & C_{0,\mathrm{ref},E_3} & C_{0,\mathrm{ref},E_4} & C_{0,\mathrm{ref},E_5} & \cdots & C_{0,\mathrm{ref},E_n} \\ C_{0,\mathrm{pos}\,1,E_1} & C_{0,\mathrm{pos}\,1,E_2} & C_{0,\mathrm{pos}\,1,E_3} & C_{0,\mathrm{pos}\,1,E_4} & C_{0,\mathrm{pos}\,1,E_5} & \cdots & C_{0,\mathrm{pos}\,1,E_n} \\ C_{0,\mathrm{pos}\,2,E_1} & C_{0,\mathrm{pos}\,2,E_2} & C_{0,\mathrm{pos}\,2,E_3} & C_{0,\mathrm{pos}\,2,E_4} & C_{0,\mathrm{pos}\,2,E_5} & \cdots & C_{0,\mathrm{pos}\,2,E_n} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ C_{0,\mathrm{pos}\,n,E_1} & C_{0,\mathrm{pos}\,n,E_2} & C_{0,pos\,n,E_3} & C_{0,\mathrm{pos}\,n,E_4} & C_{0,\mathrm{pos}\,n,E_5} & \cdots & C_{0,\mathrm{pos}\,n,E_n} \end{matrix} \tag{10}$$

where, C represents a value of count rate evaluated at a certain time, for a well defined emission energy and at a counting position, in particular, C is calculated similarly to eq. 6:

$$C_0 = \frac{n_\mathrm{p}\, \lambda\, t_\mathrm{c}\, \mathrm{e}^{\mu\left(1 - \frac{t_\mathrm{l}}{t_\mathrm{c}}\right)}}{t_\mathrm{l}\, \mathrm{e}^{\lambda t_\mathrm{d}}\, \left(1 - \mathrm{e}^{\lambda t_\mathrm{c}}\right)}, \tag{11}$$

where C_0 is the count rate reported at a defined time, also in this case it is the reference certificate date.

The array in 10 is normalized with respect to the corresponding count rates at reference position, inverted and square rooted in order to get a pseudo linear trend of the normalized count rate over the counting distance along the columns [8].

$$\begin{matrix} 1 & 1 & 1 & 1 & 1 & \cdots & 1 \\ \sqrt{\frac{C_{0,\mathrm{ref},E_1}}{C_{0,\mathrm{pos}\ 1,E_1}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_2}}{C_{0,\mathrm{pos}\ 1,E_2}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_3}}{C_{0,\mathrm{pos}\ 1,E_3}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_4}}{C_{0,\mathrm{pos}\ 1,E_4}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_5}}{C_{0,\mathrm{pos}\ 1,E_5}}} & \cdots & \sqrt{\frac{C_{0,\mathrm{ref},E_n}}{C_{0,\mathrm{pos}\ 1,E_n}}} \\ \sqrt{\frac{C_{0,\mathrm{ref},E_1}}{C_{0,\mathrm{pos}\ 2,E_1}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_2}}{C_{0,\mathrm{pos}\ 2,E_2}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_3}}{C_{0,\mathrm{pos}\ 2,E_3}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_4}}{C_{0,\mathrm{pos}\ 2,E_4}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_5}}{C_{0,\mathrm{pos}\ 2,E_5}}} & \cdots & \sqrt{\frac{C_{0,\mathrm{ref},E_n}}{C_{0,\mathrm{pos}\ 2,E_n}}} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ \sqrt{\frac{C_{0,\mathrm{ref},E_1}}{C_{0,\mathrm{pos}\ n,E_1}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_2}}{C_{0,\mathrm{pos}\ n,E_2}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_3}}{C_{0,\mathrm{pos}\ n,E_3}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_4}}{C_{0,\mathrm{pos}\ n,E_4}}} & \sqrt{\frac{C_{0,\mathrm{ref},E_5}}{C_{0,\mathrm{pos}\ n,E_5}}} & \cdots & \sqrt{\frac{C_{0,\mathrm{ref},E_n}}{C_{0,\mathrm{pos}\ n,E_n}}} \end{matrix} \tag{12}$$

In case values from matrix 12 are missing or not recognized by the software, the whole column will be dismissed for the following calculations. If the number of non-missing values along a column is more or equal to 4 (in matrix 12 at least 4 counting positions should be covered), a three-parameters polynomial fit is performed along each column to describe their $\sqrt{\frac{C_{0,\mathrm{ref},E}}{C_{0,\mathrm{pos}\ n,E}}}$ pseudo linear trend over distance for a defined source emitting at energy E:

$$\sqrt{\frac{C_{0,\mathrm{ref}}}{C_{0,d}}} = r_1 d^2 + r_2 d + r_3, \tag{13}$$

where, r_1, r_2 and r_3 are fitting parameters and d is the distance from detector end-cap in mm, deduced from the counting positions. These fitting parameters are used to calculate the d'_0 value (the theoretical distance inside the detector providing the maximum γ-absorption [8]) at any counting position and energy. The d'_0 is evaluated by assuming a linearization of the trend described in eq. 13 along the distance:

$$y = r'_1 x + r'_2 \tag{14}$$

where, y is the linearized $\sqrt{\frac{C_{0,\mathrm{ref}}}{C_{0,\mathrm{pos}}}}$ trend, x is the independent variable (distance from detector end-cap), r'_1 and r'_2 are the linearized parameters that can be defined in terms of r_1, r_2 and r_3:

$$r'_1 = \frac{\partial y}{\partial d} = 2 r_1 d + r_2, \tag{15}$$

$$r'_2 = y - r'_1 d = -r_1 d^2 + r_3. \tag{16}$$

The wanted d'_0 value is obtained by solving eq. 14 for x while $y = 0$, each time replacing d with the distance of a counting position:

$$d'_0 = -\frac{r'_2}{r'_1} = -\frac{-r_1 d^2 + r_3}{2r_1 d + r_2}, \tag{17}$$

where, d represents the distance at a certain counting position. Since d'_0 values are expected to be negative (due to the fact that they represent a distance inside the detector while the origin is on the end-cap), if they turn out as positive they are discarded since cannot be accepted for following processes. Uncertainties of d'_0 are taken into account by propagating uncertainties of parameters in the eq. 17.

A 2d array including d'_0 values for every position and energy is finally obtained:

$$\begin{matrix} d'_0(\mathrm{ref}, E_1) & d'_0(\mathrm{ref}, E_2) & d'_0(\mathrm{ref}, E_3) & d'_0(\mathrm{ref}, E_4) & d'_0(\mathrm{ref}, E_5) & \dots & d'_0(\mathrm{ref}, E_n) \\ d'_0(\mathrm{pos}\,1, E_1) & d'_0(\mathrm{pos}\,1, E_2) & d'_0(\mathrm{pos}\,1, E_3) & d'_0(\mathrm{pos}\,1, E_4) & d'_0(\mathrm{pos}\,1, E_5) & \dots & d'_0(\mathrm{pos}\,1, E_n) \\ d'_0(\mathrm{pos}\,2, E_1) & d'_0(\mathrm{pos}\,2, E_2) & d'_0(\mathrm{pos}\,2, E_3) & d'_0(\mathrm{pos}\,2, E_4) & d'_0(\mathrm{pos}\,2, E_5) & \dots & d'_0(\mathrm{pos}\,2, E_n) \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ d'_0(\mathrm{pos}\,n, E_1) & d'_0(\mathrm{pos}\,n, E_2) & d'_0(\mathrm{pos}\,n, E_3) & d'_0(\mathrm{pos}\,n, E_4) & d'_0(\mathrm{pos}\,n, E_5) & \dots & d'_0(\mathrm{pos}\,n, E_n) \end{matrix} \tag{18}$$

From calculated d'_0 points for each counting position (along the rows of 18), an exponential polynomial weighted fit (eq. 19) is performed after first guess parameters are evaluated by performing a similar unweighted fit on $\ln |d'_0|$ values:

$$d'_0 = -\mathrm{e}^{z_1 E + z_2 + z_3 E^{-1} + z_4 E^{-2} + z_5 E^{-3}}, \tag{19}$$

where, z_1, z_2, z_3, z_4 and z_5, are fitting parameters and E is the emission energy in MeV. These fits will be adopted in case geometrical corrections are needed (small variability on nominal counting positions, extended samples).

In order to account the situation when sample and standard are acquired in different counting positions, a parameter called $k_{\varepsilon \Delta d_{\mathrm{ref}}}$ is introduced defining the efficiency ratio at reference position with respect to any other counting position [9]; its value is experimentally evaluated as in 12 but

without applying the square root:

$$\begin{matrix} 1 & 1 & 1 & 1 & 1 & \dots & 1 \\ \frac{C_{0,\mathrm{ref},E_1}}{C_{0,\mathrm{pos}\ 1,E_1}} & \frac{C_{0,\mathrm{ref},E_2}}{C_{0,\mathrm{pos}\ 1,E_2}} & \frac{C_{0,\mathrm{ref},E_3}}{C_{0,\mathrm{pos}\ 1,E_3}} & \frac{C_{0,\mathrm{ref},E_4}}{C_{0,\mathrm{pos}\ 1,E_4}} & \frac{C_{0,\mathrm{ref},E_5}}{C_{0,\mathrm{pos}\ 1,E_5}} & \dots & \frac{C_{0,\mathrm{ref},E_n}}{C_{0,\mathrm{pos}\ 1,E_n}} \\ \frac{C_{0,\mathrm{ref},E_1}}{C_{0,\mathrm{pos}\ 2,E_1}} & \frac{C_{0,\mathrm{ref},E_2}}{C_{0,\mathrm{pos}\ 2,E_2}} & \frac{C_{0,\mathrm{ref},E_3}}{C_{0,\mathrm{pos}\ 2,E_3}} & \frac{C_{0,\mathrm{ref},E_4}}{C_{0,\mathrm{pos}\ 2,E_4}} & \frac{C_{0,\mathrm{ref},E_5}}{C_{0,\mathrm{pos}\ 2,E_5}} & \dots & \frac{C_{0,\mathrm{ref},E_n}}{C_{0,\mathrm{pos}\ 2,E_n}} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ \frac{C_{0,\mathrm{ref},E_1}}{C_{0,\mathrm{pos}\ n,E_1}} & \frac{C_{0,\mathrm{ref},E_2}}{C_{0,\mathrm{pos}\ n,E_2}} & \frac{C_{0,\mathrm{ref},E_3}}{C_{0,\mathrm{pos}\ n,E_3}} & \frac{C_{0,\mathrm{ref},E_4}}{C_{0,\mathrm{pos}\ n,E_4}} & \frac{C_{0,\mathrm{ref},E_5}}{C_{0,\mathrm{pos}\ n,E_5}} & \dots & \frac{C_{0,\mathrm{ref},E_n}}{C_{0,\mathrm{pos}\ n,E_n}} \end{matrix} \tag{20}$$

Fits for $k_{\varepsilon\Delta d}$ data are performed for each counting position (along the rows of 20) using an exponential polynomial unweighted fit (eq. 21):

$$k_{\varepsilon\Delta d_{\mathrm{ref}}} = \mathrm{e}^{j_1 E + j_2 + j_3 E^{-1} + j_4 E^{-2} + j_5 E^{-3} + j_6 E^{-4}}, \tag{21}$$

where, $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ denotes the efficiency ratio of the reference position to the currently investigated position, j_1, j_2, j_3, j_4, j_5 and j_6 are fitting parameters and E is the energy in MeV. The covariance matrix of the parameters is also returned in order to be able to evaluate the uncertainty of the curve. A check on the parameters' uncertainty is performed after a successful fit elaboration and the parameter which highest relative uncertainty, if is above the limit set in the settings window (see section 3.2.1), is discarded; the fit (and subsequent check) is reiterated until the condition is satisfied or in case only 4 fitting parameters are left.

During the elaboration process the software looks for PT fits already performed and includes them in the current characterization instance; if PT fits are missing in some position, a default value of 0 is adopted and coincidence corrections won't take place for those counting positions. It is thus necessary to manage PT elaborations before starting the detector characterization computation.

When a successful detector characterization is concluded, a confirmation message will appear while the logbox is compiled with useful information and the plot result section becomes active to display plots. If the elaboration stops due to some problem, the logbox should report the occurrence causing the halt. Anyway, the most probable causes of error might be:

- biases in the peaks of the peaklist or a non-suitable ΔE parameter resulting in misidentification of peaks,

- errors in introduction of distances for counting positions,

- erroneous creation or recall of the source certificate or selection of emitting energies too close to each other leading to ambiguities in the peak recognition algorithm.

The button allows to save the successful elaboration with filename chosen in the entry with label `'name'`. The process creates three files with the same name but different extensions that are saved in the «characterization» folder: the actual data recalled by the software (.pos), a copy of logfile (.txt) and a copy of the plots for fitted and experimental values (.pkl). It is worth to note that if the Detector characterization window is closed before the elaboration is saved all progresses are lost, for this reason warnings and confirmation messages will appear anytime the window is going to be closed. It should also be acknowledged that while saving, the software does not check if files with same name are already present in the folder, thus, it will overwrite those without asking for permission.

3.3.2 Delete detector characterization

If a previously saved detector characterization is selected from the drop-down menu it can be deleted from disc by pressing the button; a confirmation subwindow will appear before completing the procedure.

3.3.3 Rename detector characterization

If a previously saved detector characterization is selected from the drop-down menu it can be renamed by pressing the button; a subwindow will appear to type the new name (Figure 20).

Figure 20: The characterization rename subwindow

Pressing the enter key from keyboard allows to check the validity of the new name and confirm it, if successful.

3.3.4 Display detector characterization

If a previously saved detector characterization is selected from the drop-down menu its information can be displayed by pressing the button; this will open a subwindow recalling all information concerning the characterization that are stored on disc (Figure 21):

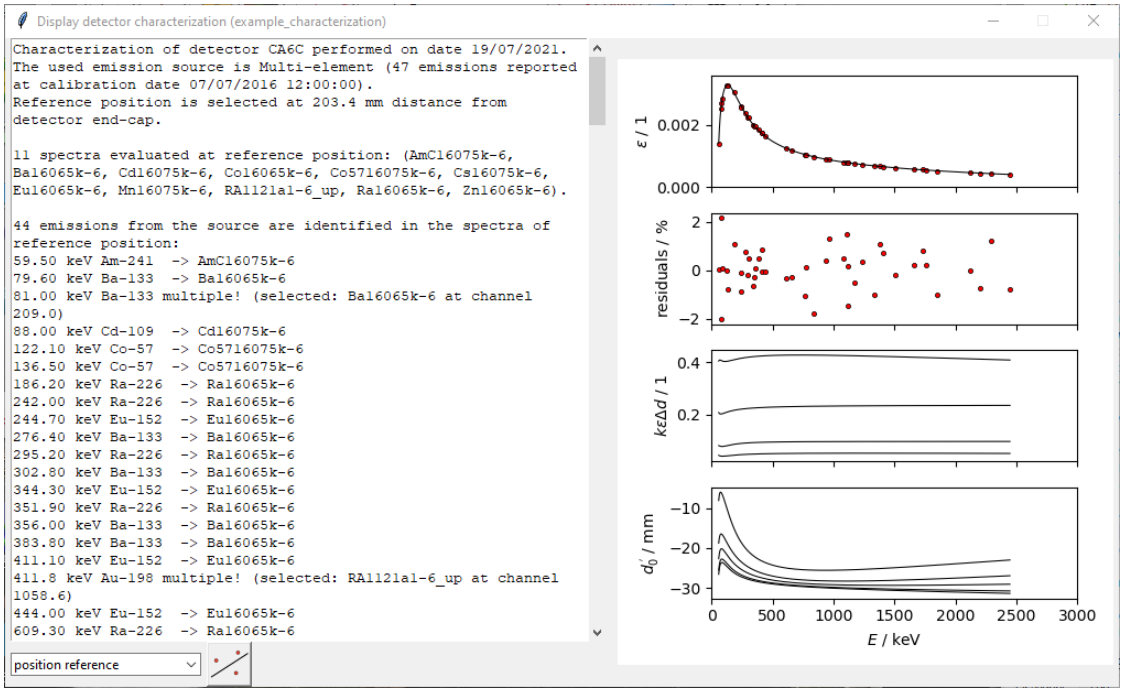


Figure 21: The detector characterization display subwindow

On the left-hand side the log is reported while on the right-hand side a summary of plots for detection efficiency at reference position, relative residuals of detection efficiency, $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ values at counting positions other than reference and d'_0 values at all counting positions are present. At the bottom-left of the window a region similar to the `'plot results'` in new detector characterization window (see section 3.3.1) is present; similarly, this allows to display plots recorder during the characterization process.

3.3.5 Flux evaluation

The flux evaluation process, accessible with button, introduces the capability to elaborate measurement of neutron flux parameters by means of the bare triple monitor method [10].

The bare triple monitor consists in the co-irradiation of a flux monitor set (usually containing Au and Zr) and subsequent acquisition of activated nuclides to measure neutron flux parameters. The instance of the method implemented in this software is able to evaluate the thermal to epithermal conventional flux ratio (f), correction to the E^{-1} epithermal flux trend (α), conventional thermal flux (Φ_{th}), conventional epithermal flux (Φ_{e}), and fast flux (Φ_{f}), all defined within the Hogdahl convention.

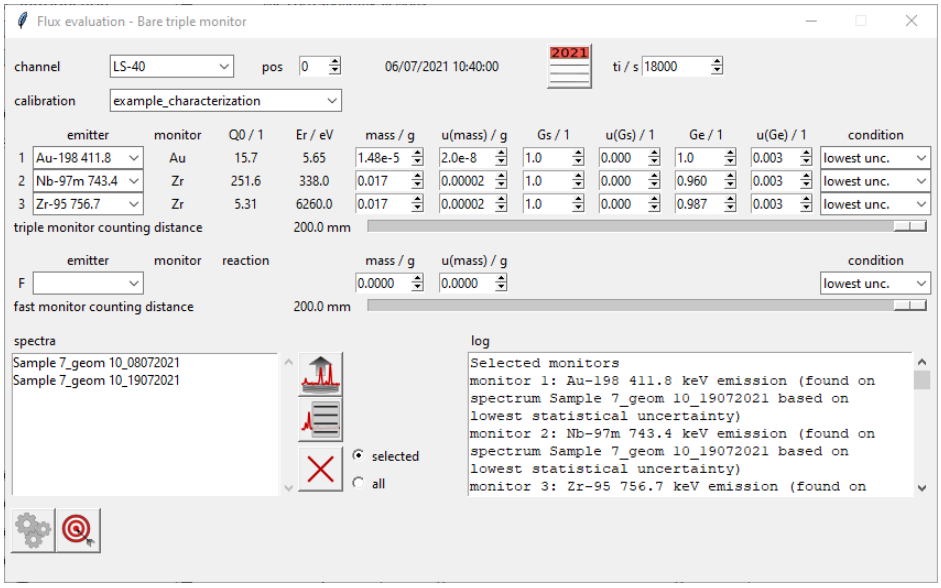


Figure 22: The bare triple monitor subwindow

The flux evaluation subwindow requires multiple information concerning the measurement experiment (Figure 22). In particular, the name of the channel being investigated and an indication of the position within the channel are required in the entries labeled `'channel'` and `'position'`, respectively; commas are not allowed in those entries. The time of end irradiation for the monitor set is modified with the button, the default being the current date; the irradiation time in seconds is entered in the spinbox labeled `'ti / s'`. The detector characterization in use is selected, among the stored ones, by the drop-down menu labeled `'calibration'`; the choice of the characterization activates the independent sliders labeled `'triple monitor counting distance'` and `'fast monitor counting distance'` allowing to select the counting distances used for acquisition of the corresponding monitors. While the monitor for fast flux might be acquired at different distance with respect the triple monitor, the three emitters composing the triple monitor have to be acquired at the same position with respect to each other (and likely as far as possible to detector end-cap) to assure minimum measurement uncertainty.

For each monitor (the triplet labeled as `'1'`, `'2'` and `'3'` and the fast labeled as `'F'`) a line of information has to be filled. For each one the emitter has to be identified by means of the drop-down menu below the label `'emitter'`. Said drop-down menu includes all emitters from Au and Zr by default, however, additional emitters might be added by writing the atomic symbols of their targets, separated by a space, in the first line of «flux.txt» file found in the «monitor_elements» folder. The software will search the k_0 database for any emission obtained from the targets in the «flux.txt» file which undergo activation-decay types I (and similar) or IIA. Mass of the element in the monitor and its uncertainty are required in the corresponding spinboxes labeled `'mass / g'` and `'u(mass) / g'`. Room to insert self-shielding corrections for the triple monitor set (thermal and epithermal) is placed in the corresponding spinboxes labeled `'Gs / 1'`, `'u(Gs) / 1'`, `'Ge / 1'` and `'u(Ge) / 1'`, respectively. The drop-down menu labeled `'condition'` allows to choose various condition to discriminate the peak to adopt for calculation when multiple spectra of the monitor set are recalled; the options are: `'lowest unc.'` selecting the peak with lowest

statistical uncertainty, `'shortest'` selecting the peak found in the spectrum with smaller live time, `'longest'` selecting the peak found in the spectrum with greater live time, `'earliest'` selecting the peak found in the spectrum acquired after shortest decay time, `'latest'` selecting the peak found in the spectrum acquired after longest decay time.

The listbox in the bottom left-hand side, labeled `'spectra'`, lists all the recalled spectra adopted for the evaluation; the three , , and buttons allow to perform actions such as import, display and delete spectra, respectively.

The textbox in the bottom right-hand side, labeled `'log'` provides useful information when a successful elaboration is completed.

At the bottom of the window the two *Compute flux parameters* and *Confirm obtained values* buttons allow to perform the corresponding actions.

The button starts the elaboration process by searching the recalled spectra to find the peaks related to the selected monitors, successively, specific count rate (C_{sp}) is calculated for each emitter (eq. 22 for type I activation-decay, eq. 23 for type IIA activation-decay):

$$C_{\mathrm{sp}} = \frac{n_{\mathrm{p}}\,\lambda\,t_{\mathrm{c}}\,\mathrm{e}^{\mu\left(1-\frac{t_{\mathrm{l}}}{t_{\mathrm{c}}}\right)}}{t_{\mathrm{l}}\,\left(1-\mathrm{e}^{-\lambda t_{\mathrm{i}}}\right)\mathrm{e}^{-\lambda t_{\mathrm{d}}}\left(1-\mathrm{e}^{-\lambda t_{\mathrm{c}}}\right)\,COI\,m}, \tag{22}$$

$$C_{\mathrm{sp}} = \frac{n_{\mathrm{p}}\,\left(\lambda_3-\lambda_2\right)\,t_{\mathrm{c}}\,\mathrm{e}^{\mu\left(1-\frac{t_{\mathrm{l}}}{t_{\mathrm{c}}}\right)}}{t_{\mathrm{l}}\,COI\,m\,\left(\frac{\lambda_3}{\lambda_2}\left(1-\mathrm{e}^{-\lambda_2 t_{\mathrm{i}}}\right)\mathrm{e}^{-\lambda_2 t_{\mathrm{d}}}\left(1-\mathrm{e}^{-\lambda_2 t_{\mathrm{c}}}\right)-\frac{\lambda_2}{\lambda_3}\left(1-\mathrm{e}^{-\lambda_3 t_{\mathrm{i}}}\right)\mathrm{e}^{-\lambda_3 t_{\mathrm{d}}}\left(1-\mathrm{e}^{-\lambda_3 t_{\mathrm{c}}}\right)\right)}, \tag{23}$$

where, n_{p} is the net peak area, λ is the decay constant (λ_2 and λ_3), μ is the excess counting loss constant of the detection system, t_{c} is the real counting time, t_{l} is the live counting time, t_{i} is the irradiation time, t_{d} is the decay time from irradiation end, COI is the true-coincidence correction factor and m is the monitor element mass. The uncertainty is estimated by propagation of the relative uncertainties of n_{p}, COI and m.

The α value is calculated by solving the corresponding implicit function [11]:

$$\left(\frac{1}{\frac{C_{\mathrm{sp},1}\,k_{0,2}\,\varepsilon_2}{C_{\mathrm{sp},2}\,k_{0,1}\,\varepsilon_1}-1}-\frac{1}{\frac{C_{\mathrm{sp},1}\,k_{0,3}\,\varepsilon_3}{C_{\mathrm{sp},3}\,k_{0,1}\,\varepsilon_1}-1}\right)G_{e,1}\left(\frac{Q_{0,1}-0.429}{\bar{E}^{\alpha}_{\mathrm{r},1}}+\frac{0.429}{(2\alpha+1)\,0.55^{\alpha}}\right)+$$
$$-\left(\frac{1}{1-\frac{C_{\mathrm{sp},2}\,k_{0,1}\,\varepsilon_1}{C_{\mathrm{sp},1}\,k_{0,2}\,\varepsilon_2}}\right)G_{e,2}\left(\frac{Q_{0,2}-0.429}{\bar{E}^{\alpha}_{\mathrm{r},2}}+\frac{0.429}{(2\alpha+1)\,0.55^{\alpha}}\right)+ \tag{24}$$
$$+\left(\frac{1}{1-\frac{C_{\mathrm{sp},3}\,k_{0,1}\,\varepsilon_1}{C_{\mathrm{sp},1}\,k_{0,3}\,\varepsilon_3}}\right)G_{e,3}\left(\frac{Q_{0,3}-0.429}{\bar{E}^{\alpha}_{\mathrm{r},3}}+\frac{0.429}{(2\alpha+1)\,0.55^{\alpha}}\right)=0,$$

where, subscripts 1,2 and 3 indicate monitors labeled `'1'`, `'2'` and `'3'`, respectively, C_{sp} is the specific count rate at saturation (eq. 22,23), k_0 is the composite nuclear constant defined by De Corte [3], ε is the detection efficiency that in this instance does not account for position variability, G_e is the epithermal self-shielding factor, Q_0 is the resonance integral to thermal cross section ratio and E_r is the effective resonance energy.

The uncertainty of α is evaluated by reiterating eq. 24 several times while changing one input parameter at a time of a value equal $\pm$ its standard uncertainty. Those sensitivity coefficients are propagated with the covariance matrix of input parameters to get the variance of α and, subsequently, the uncertainty.

The knowledge of α is introduced in the following calculations to evaluate f:

$$f=\frac{\frac{k_{0,2}\,\varepsilon_2}{k_{0,3}\,\varepsilon_3}G_{e,2}\left(\frac{Q_{0,2}-0.429}{\bar{E}^{\alpha}_{\mathrm{r},2}}+\frac{0.429}{(2\alpha+1)0.55^{\alpha}}\right)-\frac{C_{\mathrm{sp},2}}{C_{\mathrm{sp},3}}G_{e,3}\left(\frac{Q_{0,3}-0.429}{\bar{E}^{\alpha}_{\mathrm{r},3}}+\frac{0.429}{(2\alpha+1)0.55^{\alpha}}\right)}{G_{\mathrm{s}}\left(\frac{C_{\mathrm{sp},2}}{C_{\mathrm{sp},3}}-\frac{k_{0,2}\,\varepsilon_2}{k_{0,3}\,\varepsilon_3}\right)}, \tag{25}$$

with its uncertainty evaluated by propagation of sensitivity coefficient and covariance matrix. It is worth to take into account that eq. 25 assumes the thermal self-shielding parameter (G_{s}) being the same for the two monitors; moreover, monitor 2 and 3 are chosen since in the default configuration they represents the two monitors of Zr element, giving the best uncertainty on f evaluation as some of the parameters cancel out.

Thermal and epithermal conventional fluxes are calculated only if Au-198 441.8 keV is selected among the monitor since the software needs some literature value that, for the Au emission, is hard-coded in the source code file itself:

$$\Phi_{\mathrm{th}}=\frac{C_{\mathrm{sp,Au}}\,M_{\mathrm{Au}}}{\theta_{\mathrm{Au}}\,\Gamma_{\mathrm{Au}}\,\varepsilon_{\mathrm{Au}}\,N_{\mathrm{A}}\,\sigma_{0,\mathrm{Au}}\left(G_{\mathrm{s}}+\frac{G_{\mathrm{e,Au}}}{f}\left(\frac{Q_{0,\mathrm{Au}}-0.429}{\bar{E}^{\alpha}_{\mathrm{r,Au}}}+\frac{0.429}{(2\alpha+1)0.55^{\alpha}}\right)\right)}, \tag{26}$$

$$\Phi_{\mathrm{e}} = \frac{C_{\mathrm{sp,Au}}\, M_{\mathrm{Au}}}{\theta_{\mathrm{Au}}\, \Gamma_{\mathrm{Au}}\, \varepsilon_{\mathrm{Au}}\, N_{\mathrm{A}}\, \sigma_{0,\mathrm{Au}} \left(G_{\mathrm{s}}\, f + G_{\mathrm{e,Au}} \left(\frac{Q_{0,\mathrm{Au}} - 0.429}{\bar{E}^{\alpha}_{\mathrm{r,Au}}} + \frac{0.429}{(2\alpha+1)0.55^{\alpha}} \right) \right)}, \tag{27}$$

where, the subscript Au refers to the Au-198 411.8 keV monitor, M is the molar mass, θ is the isotopic abundance, Γ is the γ-yield, N_A is Avogadro constant, σ_0 is the (n,γ) reaction cross section at $2200\,\mathrm{m\,s^{-1}}$. The hard-coded values are: $M_{\mathrm{Au}} = 196.966\,569\,\mathrm{g\,mol^{-1}}$, $\theta_{\mathrm{Au}} = 1$, $\Gamma_{\mathrm{Au}} = 0.9562$ and $\sigma_{0,\mathrm{Au}} = 9.870(1) \times 10^{-23}\,\mathrm{cm^2}$.

The last elaboration performed concerns the fast flux, whether a fast monitor is selected in the corresponding drop-down menu in the flux evaluation window:

$$\Phi_{\mathrm{f}} = \frac{C_{\mathrm{sp,F}}\, M_{\mathrm{F}}}{\theta_{\mathrm{F}}\, \Gamma_{\mathrm{F}}\, \varepsilon_{\mathrm{F}}\, N_{\mathrm{A}}\, \sigma_{\mathrm{f,F}}}, \tag{28}$$

where, the subscript F refers to the selected fast monitor and σ_f is the U-235 averaged fast reaction cross section for which information can be found in the «fast_data.csv» file within the «literaturedata» folder.

Uncertainty of all fluxes is evaluated by propagation of sensitivity coefficients and covariance matrix.

A successful elaboration of flux parameters fills the logbox at the right-hand side of the window with useful information about the process and results; it also gives, for α and f index of the five most important contributors to their estimated uncertainties. In addition, after a couple of checks on the calculated results, it allows to save the obtained data with the button. Saved information are appended to the «channel.csv» file in the «facility» folder and can be recalled by the browse databases command (see section 3.2.2).

3.3.6 Gradient evaluation

The gradient evaluation process, accessible with button, introduces the capability to elaborate measurement of neutron flux gradients within vertical positions of an irradiation channel.

The so-defined vertical count rate gradient due to unit distance of irradiation position (β) is evaluated as the ratio of specific count rate for two known samples over the distance between their irradiation positions.

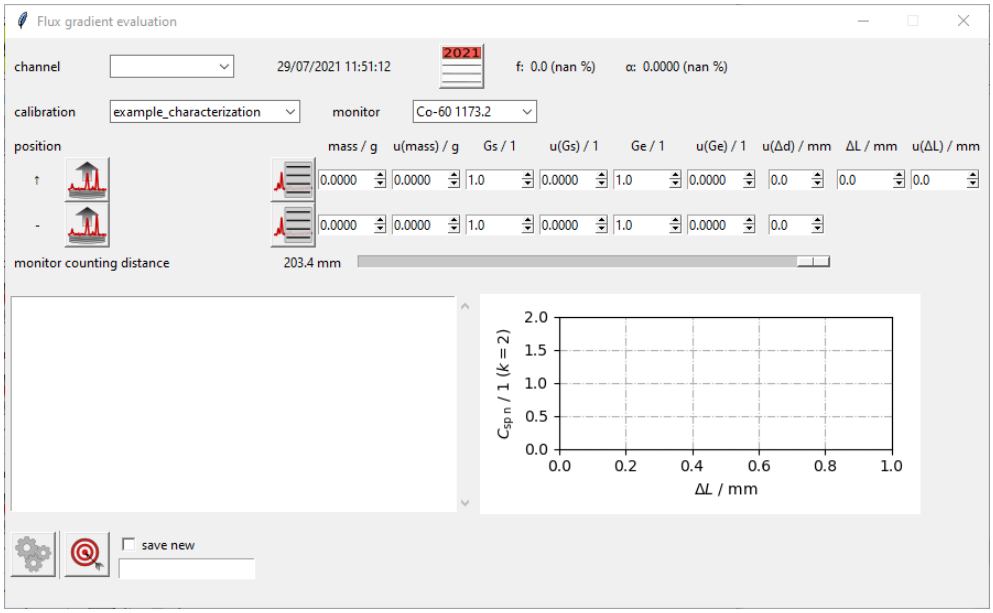


Figure 23: The flux evaluation subwindow

The flux gradient evaluation subwindow requires multiple information concerning the measurement experiment. In particular, the name of the channel being investigated is recalled by the drop-down menu labeled `'channel'`; when a channel is selected the corresponding f and α values appear in the labels `'f:'`, `'$\alpha$:'` at the right-hand side just as an indication and do not interfere with the following β calculations. The time of end irradiation for the monitor set is modified with the button, the default being the current date. The detector characterization in use is selected, among the stored ones, by the drop-down menu labeled `'calibration'`; the choice of the characterization activates the slider labeled `'monitor counting distance'` allowing to select the counting distances used for acquisition of the corresponding monitors. The emission to be used as monitor is selected in the drop-down menu labeled `'monitor'`; all the simple activation-decay emissions present in the k_0-database are eligible as monitor, thus, additional emitters might be added to the default ones (Au and Co) by writing the atomic symbols of their targets, separated by a space, in the first line of «gradient.txt» file found in the «monitor_elements» folder. The two monitors are identified with the labels `'↑'` and `'-'`, referring to the one placed higher and the one placed lower in the irradiation facility, respectively. A spectrum for both monitors needs to be recalled with the button and can be displayed with button. Values and uncertainties for masses (labeled `'mass / g'` and `'u(mass) / g'`), thermal self-shieldings (labeled `'Gs / 1'` and `'u(Gs) / 1'`), epithermal self-shieldings (labeled `'Ge / 1'` and `'u(Ge) / 1'`) are required in the corresponding spinboxes. In addition, uncertainty on counting positioning might be included for both monitors in the spinboxes labeled `'u(Δd) / mm'`. Finally, the distance with uncertainty of the higher monitor with respect to the lower one is required in the corresponding spinbox labeled `'ΔL / mm'` and`'u(ΔL) / mm'`; this distance should always be positive since the reference system used for this software considers distances going upwards as positive.

In the lower part of the window a log box is found in the left-hand side and a plot in the right-hand side; those will provide useful information while starting the process.

At the bottom of the window, *Evaluate* and *Save elaboration* buttons allows to perform the corresponding actions.

Clicking the starts the evaluation of gradient by retrieving literature data of the selected monitor from the k_0-database and inspecting the spectra to find the monitor peaks. For both monitors specific count rate of nuclides at a reference time (and considering the uncertainty on positioning), ς, is calculated:

$$\varsigma = \frac{n_{\mathrm{p}}\, t_{\mathrm{c}}\, \lambda\, \mathrm{e}^{\mu\left(1-\frac{t_{\mathrm{l}}}{t_{\mathrm{c}}}\right)}}{t_{\mathrm{l}}\, \mathrm{e}^{-\lambda t_{\mathrm{d}}}\left(1-\mathrm{e}^{-\lambda t_{\mathrm{c}}}\right) m \left(\frac{\mathrm{d}_{\mathrm{ref}}-d_0'}{\mathrm{d}_{\mathrm{ref}}+\Delta d-d_0'}\right)^2 \left(G_{\mathrm{s}} + \frac{G_{\mathrm{e}}}{f}\left(\frac{Q_0-0.429}{E_r^{\alpha}} + \frac{0.429}{(2\alpha+1)0.55^{\alpha}}\right)\right)}, \tag{29}$$

and the vertical irradiation gradient correction, β is computed accordingly:

$$\beta = \frac{\frac{\varsigma_{\uparrow}}{\varsigma_{-}} - 1}{\Delta L}, \tag{30}$$

where subscripts $\uparrow$ and $-$ indicate the monitors put in the higher irradiation position and lower irradiation position, respectively, β is the vertical count rate gradient per mm vertical distance of irradiation position and ΔL is the distance between the irradiation positions of the two monitors. The uncertainty is evaluated by propagation of variances of input parameters through eq. 29 and 30. It is worth to note that the software considers Δd in equation 29 as a variable with value 0 and uncertainty $u\left(\Delta d\right)$, this doesn't mean the two monitors have to be acquired at one of the reference positions. In fact, until they are counted at the same distance with respect to detector end-cap, $\mathrm{d}_{\mathrm{ref}\uparrow} = \mathrm{d}_{\mathrm{ref}-}$ and $\Delta d_{\uparrow} = \Delta d_{-}$, no biases are introduced in this calculations; however, in case of significant difference between the two Δd, the proposed implementation could not be suitable to evaluate β value.

A successful elaboration displays all crucial information in the log box and allows to save the obtained results by pressing after giving a valid (non-empty and without commas) position name. It allows to save the β elaboration by appending resulting data to the «beta.csv» file in the «facility» folder; those information can be recalled by the browse databases command (see section 3.2.2).

3.4 Analysis name

This section allows to assign a name to the k_0-NAA analysis defined from this point onwards. The term analysis here refers to the experimental setup and elaboration performed to obtain mass

fraction results of analytes target elements from samples all referred to their corresponding single standard. This name should unambiguously identify any analysis since this will be the default suggested filename while outputs are saved.

This section includes a *Modify analysis name* button and a readonly label showing the current analysis name.

3.4.1 Modify analysis name

The name of current analysis can be changed by pressing the button; a subwindow will appear to type the new name (Figure 24).

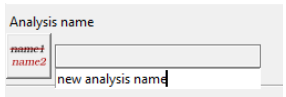


Figure 24: The modify analysis name subwindow

Pressing the enter key from keyboard allows to check the validity of the new name and confirms it, if successful.

3.5 Irradiation

This section provides tasks to manage information concerning the irradiations performed on the samples to analyze. It provides a *New irradiation* button, a drop-down menu to recall a previously saved neutron irradiations, three *Delete irradiation*, *Rename irradiation*, *Display irradiation* buttons to perform corresponding actions on the selected irradiation data.

3.5.1 New irradiation

The *New irradiation* task involves the creation of a new instance of neutron irradiation where data concerning activation of samples are processed. This task will open a subwindow (Figure 25) allowing to introduce information about date and time of neutron exposure and details about the facility where it was performed.

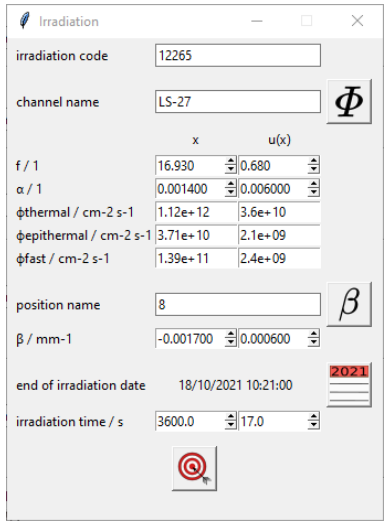


Figure 25: The new irradiation subwindow

The entry at the top of the window labeled `'irradiation code'` allows to introduce the unique code of irradiation assigned to the neutron exposure, it is not a mandatory field, however; it might be left blank but this will save the information of the current only temporary under a fictitious code «_». The entry labeled `'channel name'` allows to introduce the reference for the adopted irradiation channel. Data for this section can be inserted by hand or a selection among the already characterized facilities might be performed through the *select flux parameters from database* button. This button pops up a visualization of flux parameters database from which the wanted set of parameters can be selected (Figure 26).

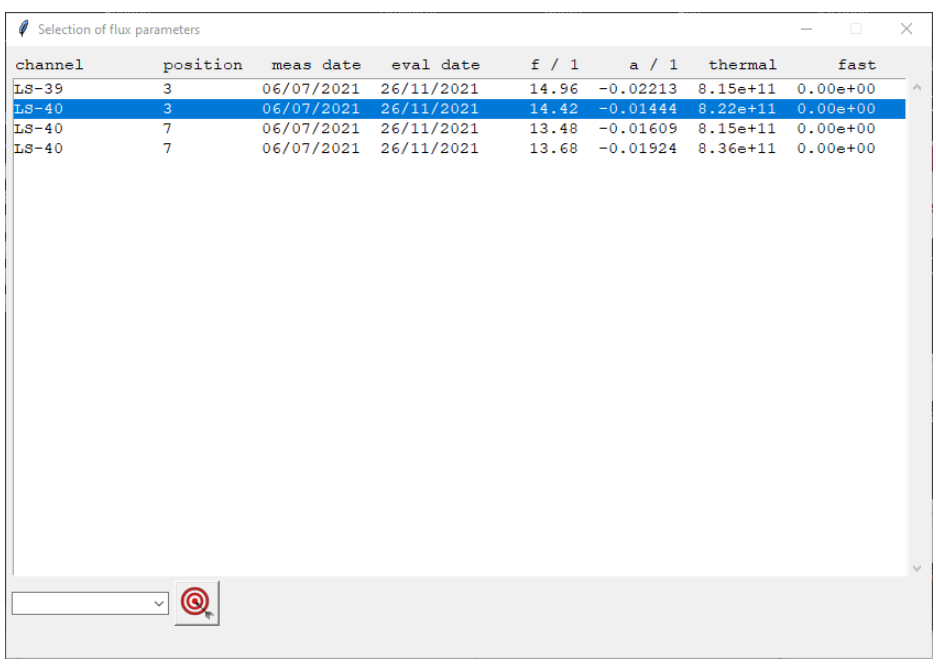


Figure 26: The selection of flux parameters subwindow

The list of data displays useful information, for each entry, in order to correctly identify the suitable set of flux parameters related to the current irradiation; in particular, channel name, indication of position within the facility where the flux was measured, date of measurement and date of evaluation through the software, f value, α value, conventional thermal and fast fluxes (in cm^{-2} s^{-1}) are reported in the columns `'channel'`, `'position'`, `'meas date'`, `'eval date'`, `'f / 1'`, `'a / 1'`, `'thermal'` and `'fast'`, respectively. The selection can be filtered by channel name through the drop-down menu at the bottom-left of the window. When a line is highlighted by clicking on it with the mouse and confirmed through the *select the current flux parameters* button, all related information as channel name, f, α, thermal, epithermal and fast fluxes are automatically filled in the corresponding spinboxes of the irradiation window.

Once the main flux parameters are selected, it is possible to introduce β and $u(\beta)$ values in the corresponding spinboxes by hand or by picking the suitable ones from the beta database through the *select beta parameters from database* button. This button pops up a visualization of beta parameters database from which the wanted one can be selected (Figure 27).

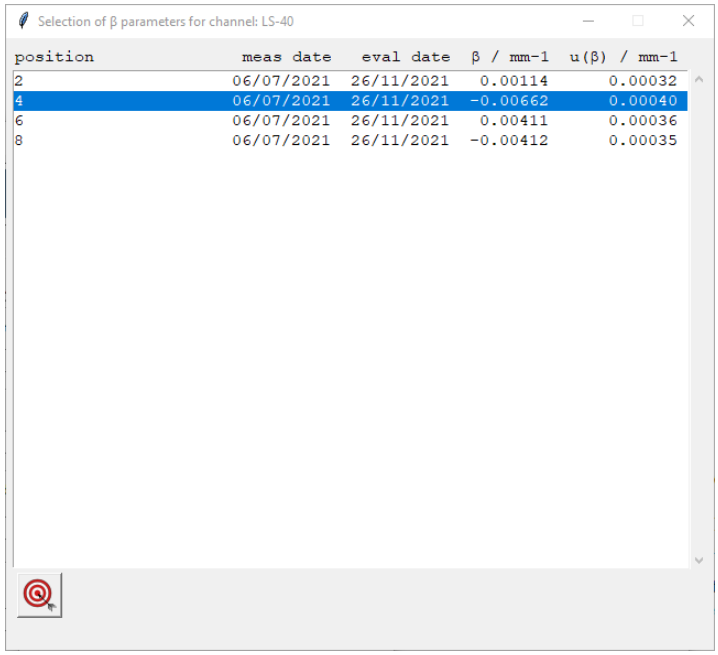


Figure 27: The selection of flux parameters subwindow

The list of data displays useful information, for each entry, in order to correctly identify the suitable beta parameter related to the current irradiation position; in particular, indication of position within the facility where the flux was measured, date of measurement and date of evaluation through the software, β value and $u(\beta)$ value are reported in the columns `'position'`, `'meas date'`, `'eval date'`, `'`β` / mm-1'` and `'u(`β`) / mm-1'`, respectively. The selection is automatically filtered according to the value reported on the channel name entry in the irradiation window.

When a line is highlighted by clicking on it with the mouse and confirmed through the *select the current β parameter* button, all related information as position name and β are automatically filled in the corresponding spinboxes of the irradiation window.

The end of irradiation date and time is showed in the text under the label `'end of irradiation date'`, it can be modified by pressing the button (similarly to paragraph 3.3.5). The length of the neutron exposure in seconds is required in the spinbox labeled `'irradiation time / s'`.

The button checks the consistency of provided data and confirms them, if successful; the just introduced irradiation is automatically selected as the current one in the irradiation drop-down menu of the main window.

3.5.2 Delete irradiation

If a previously saved irradiation is selected from the drop-down menu it can be deleted from disc by pressing the button; a confirmation subwindow will appear before completing the procedure.

3.5.3 Rename irradiation

If a previously saved irradiation is selected from the drop-down menu it can be renamed by pressing the button; a subwindow will appear to type the new name (Figure 28).

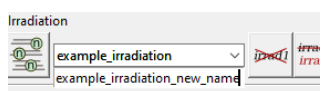
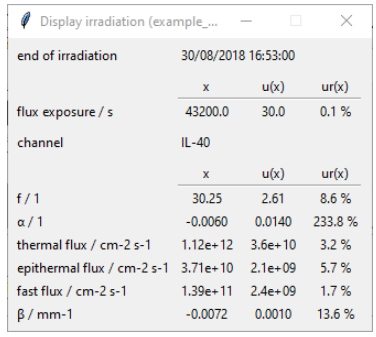


Figure 28: The rename irradiation subwindow

Pressing the enter key from keyboard allows to check the validity of the new name and confirms it, if successful.

3.5.4 Display irradiation

If a previously saved irradiation is selected from the drop-down menu its information can be displayed by pressing the button; this will open a subwindow recalling all information concerning the irradiation that are stored on disc (Figure 29):



Display irradiation (example_...

end of irradiation	30/08/2018 16:53:00		
	x	u(x)	ur(x)
flux exposure / s	43200.0	30.0	0.1 %
channel	IL-40		
	x	u(x)	ur(x)
f / 1	30.25	2.61	8.6 %
α / 1	-0.0060	0.0140	233.8 %
thermal flux / cm-2 s-1	1.12e+12	3.6e+10	3.2 %
epithermal flux / cm-2 s-1	3.71e+10	2.1e+09	5.7 %
fast flux / cm-2 s-1	1.39e+11	2.4e+09	1.7 %
β / mm-1	-0.0072	0.0010	13.6 %

Figure 29: The display irradiation subwindow

3.6 Background

The background section of the main window allows to introduce information about the γ background profile and blank in order to correct their influence from the elaboration. It comprises a *Open background* button, a drop-down menu with a spectrum counter to identify the name of spectrum currently selected and the number of recalled spectra, a *Background peaklist* button to display data of the selected spectrum and the peaklist (the list of all the elaborated peaks of the selected spectrum), a *Delete background* button with a switcher to delete spectra and a *Define material* button to provide information related to the physical sample.

3.6.1 Open background

The *Open background* task allows to recall spectrum files from disc; a subwindow asking to select a file pops up (Figure 30).

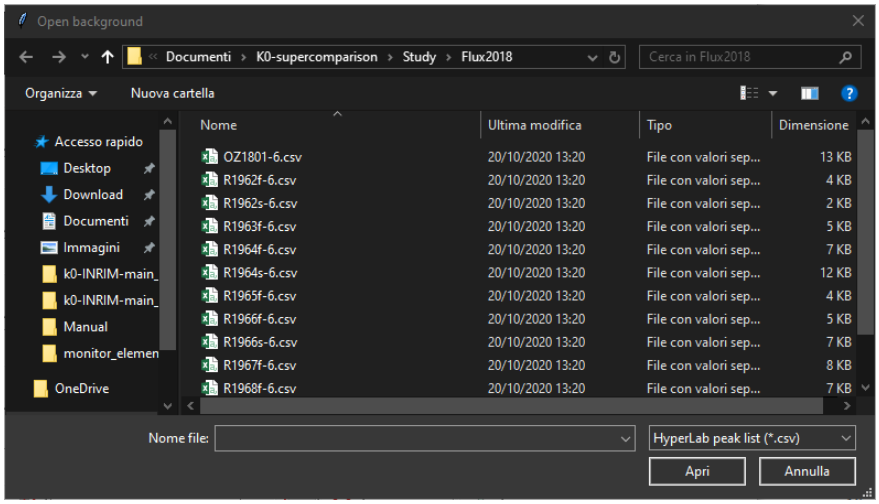


Figure 30: The window to recall spectra

The filetype to import can be changed with the bottom-right drop-down menu; the available filetypes are .csv (elaboration output from HyperLab software) or .rpt (elaboration output from GammaVision software). Once the *Open* button is pressed the software tries to open the selected file and gathers data from it, if the *always look for spectrum file* option in the settings window (see subsection 3.2.1) is set to True, also information for the spectrum profile are searched while, if not necessary, they are skipped in case the setting is False. To look for the spectrum profile the same folder of the selected peaklist is inspected for a file with same name but extension .asc (ascii formatted spectrum obtainable from HyperLab software) or .chn (default integer spectrum file from GammaVision software), in this order. For macOS and linux filesystems the spectrum file search might lead to some issue since these systems are case sensitive meaning that *some_filename.asc* $\neq$ *some_filename.ASC*. If a spectrum profile is not correctly identified on macOS or linux systems this can be the cause of the problem.

When spectrum data are correctly uploaded, the name of the selected file is appended to the drop-down menu next to the button and the spectrum counter is updated. Since the background section only accepts data relative for one spectrum, if a spectrum is already present in the drop-down menu when a new one is uploaded, the newly recalled will replace the old one.

3.6.2 Background peaklist

The *Background peaklist* button allows to browse information about the currently selected spectrum. The peaklist window shows information about the directory where the file is located, the name of uploaded files and whether they are identified as peak list (.csv or .rpt files) or spectrum (.asc or .chn files), the start acquisition time, real, live and dead time, together with the list of all γ-peaks reported in the peaklist (Figure 31).

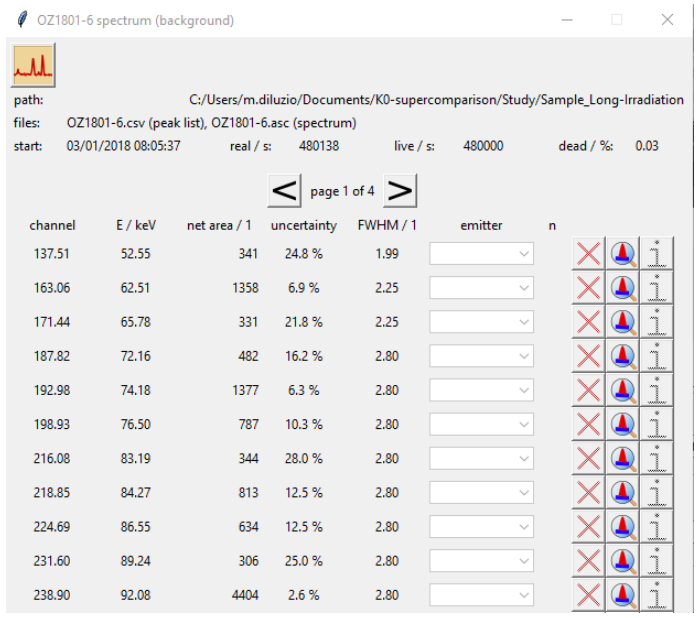


Figure 31: The peaklist window

The *Show spectrum profile* button, placed at the top-left of the window, allows to open a visualization of the spectrum profile (Figure 32), if any spectrum filetype was found when recalling the spectra.

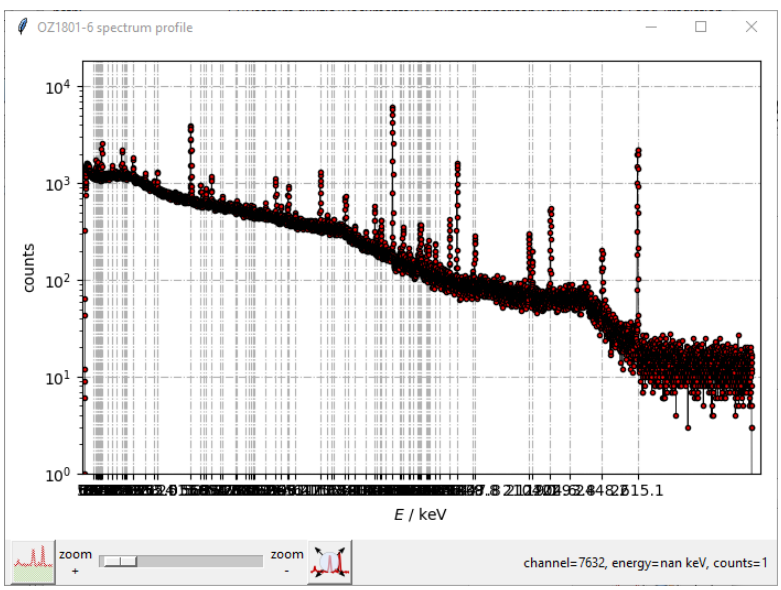


Figure 32: The spectrum window

The spectrum window displays a plot of spectrum profile with energies (in keV) on x-axis and logarithm of counts in the y-axis. It is possible to navigate the spectrum profile plot by scrolling with the mouse-wheel; a zoomed-in visualization will be automatically prompted while the zoom level is adjusted by the corresponding slider between the `'zoom +'` and `'zoom -'` labels. The

Zoom y-axis button adjusts the y-axis to the current content while navigating through the spectrum profile. At the bottom-right of the window information such as channel, energy and counts are displayed depending on mouse position over the plot; a characterization should be selected in the main window for the energy to be showed.

Back to the peaklist window, the < *Previous page* and > *Following page* buttons browse the peaklist in order to reach all the peaks since the visualization is split in pages. Each page includes a number of lines selected in the settings menu (see paragraph 3.2.1). each line provides information or actions referred to the corresponding peak; in particular, the information are the centroid of the peak, the energy at given centroid, the net area with relative uncertainty, FWHM (in channels) and the number of correspondences found in k_0 database for that energy; those are reported below the labels `'channel'`, `'E / keV'`, `'net area / 1'`, `'uncertainty'`, `'FWHM / 1'` and `'n'` respectively. Actions allow to select a specific emission from those suggested by the software using the drop-down menu below the label `'emitter'`, clear the selection, visualize the peak in the spectrum profile and get more information on the selected emission via buttons. When dealing with background spectra all these actions but *Go to peak* are disabled. The button promptly adjust the x-axis of the spectrum profile plot to center the corresponding peak.

3.6.3 Delete background

The selected (or every) background spectrum can be deleted by the button. The selector next to it allows to choose if only the selected spectrum should be deleted or all spectra; in both cases the outcome will be the same since for background section only a single spectrum is allowed; however, a confirmation subwindow will be prompted. This action does not erase from disc the original files of peak lists and spectra, it only clears them from the software memory.

3.6.4 Define material

The *Define material* task initiates a window (Figure 33) allowing to introduce information about the analysis blank, since the model equation (see eq. 35) provides this kind of correction.

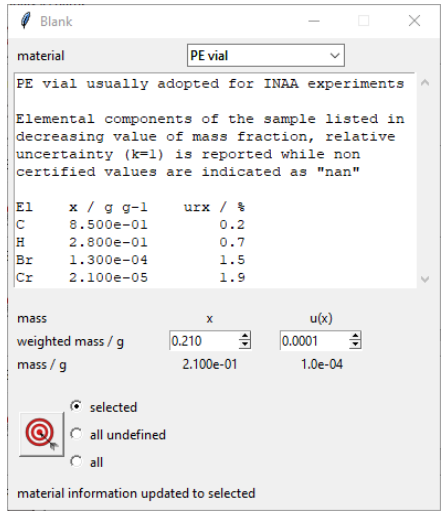


Figure 33: The window to introduce information for blank

The window allows to assign a material for the blank from the drop-down menu labeled `'material'`; the elemental composition of the chosen material will be applied to the blank for correction purposes.

Spinboxes to introduce the mass (in grams) and uncertainty of the blank are present.

The button confirms the submitted information regardless of what option is chosen in the adjacent list of radiobuttons.

3.7 Standard

The standard section of the main window allows to introduce information about the γ-spectrum containing the target element adopted as the comparator in the elaboration. It comprises a *Open standard* button, a drop-down menu with a spectrum counter to identify the name of spectrum currently selected and the total number of recalled spectra, a *Standard peaklist* button to display data of the selected spectrum and the peaklist (the list of all the elaborated peaks of the selected spectrum), a *Delete standard* button with a switcher to delete spectra and a *Define material* button to introduce information related to the physical sample. In addition, it provides a slider with a radiobutton and spinboxes to identify the actual counting position of the sample.

3.7.1 Open standard

The *Open standard* task is similar to that for the background spectrum (see paragraph 3.6.1).

3.7.2 Standard peaklist

The *Standard peaklist* button allows to browse information about the currently selected spectrum. The peaklist window shows information similarly to the background peaklist window (see paragraph 3.6.2) with the difference that, in this case, the emission selection drop-down menu and all three buttons are active (Figure 34).

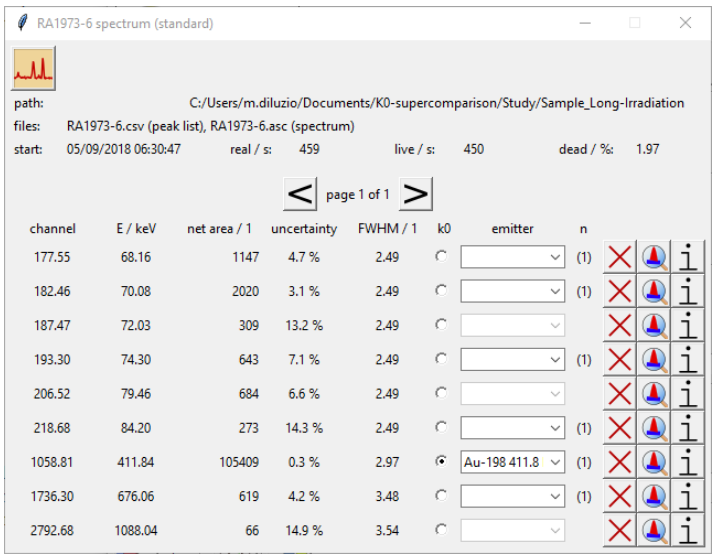
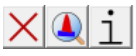


Figure 34: The standard peaklist window

The emission selection drop-down menu (under the column labeled `'emitter'`) allows to assign an emission label among the possible ones, to the corresponding peak. The emission label comprises the emitting radionuclide and the γ-energy in keV; the list of possible emissions is produced by matching the energy of the corresponding peak with the k_0-database in use within the energy tolerance set in the option $\Delta E \ / \ keV$ (see the paragraph 3.2.1); the number of entries satisfying the matching condition is indicated in the column labeled `'n'`.

With the three function  buttons it is possible to interact with the current peak; if an emission label is assigned to it, with the left button the current selection is cleared, or with the right button literature information about the emission are displayed (Figure 35), the central button opens the spectrum profile centered on the selected peak.

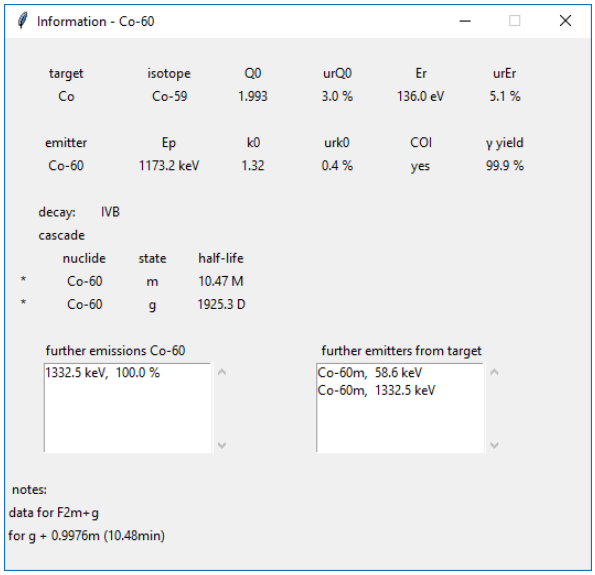


Figure 35: The emission information window

The standard peaklist window also provides the functionality to select one, and only one, peak as the designated target monitor for the analysis through the radiobutton in the column labeled `'k0'`; clicking on one of the radiobuttons will activate it and the corresponding peak is defined as the monitor. The selection is changed by clicking on any other radiobutton of by pressing the *X* button from .

All selections (emitters from drop-down menus and k0 radiobutton) are persistent even after the peaklist window is closed.

3.7.3 Delete standard

The *Delete standard* task is similar to that for the background spectrum (see paragraph 3.6.3).

3.7.4 Define material

The *Define material* task relative to standard initiate a window (Figure 36) allowing to introduce information, more than the blank window (see paragraph 3.6.4), referred to standard physical sample.

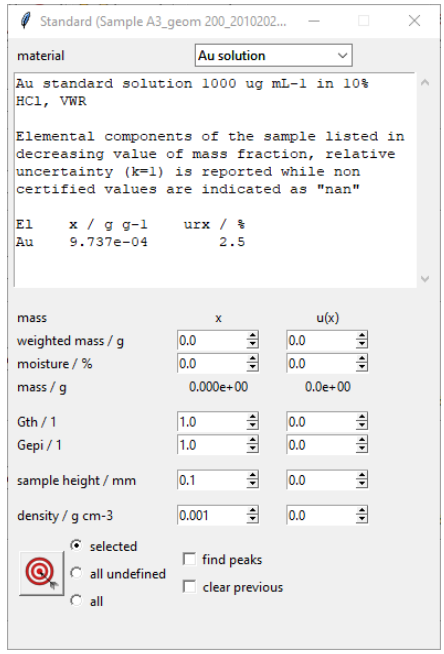


Figure 36: The window to introduce standard information

Like the blank window (see paragraph 3.6.4) there is the possibility to assign a sample composition for the standard from the drop-down menu labeled `'material'`; in this instance the choice is mandatory since the elemental composition is needed as a source of information for the target comparator mass fraction which will be used in the analysis.

Spinboxes to introduce the mass (in grams) and uncertainty of the standard are present as well as the additional spinboxes to take into account the moisture correction (as a percentage). The required information is the percent moisture of the weighted sample since the adopted correction applies to the weighted mass value as a factor:

$$m_{\mathrm{corr}} = m_{\mathrm{w}} \left(1 - \eta\right), \tag{31}$$

where m_{corr} is the moisture corrected mass, m_{w} is the uncorrected weighted mass and η is the relative moisture which is calculated by dividing for 100 the value introduced in the spinbox of the current window, $\eta = \frac{\eta_{\%}}{100}$.

In addition, specific spinboxes are present to introduce information about self-shielding corrections (thermal self-shielding G_{th} and epithermal self-shielding G_{e} labeled `'Gth / 1'` and `'Gepi / 1'`, respectively) to be applied on the comparator.

Other information are required, in order to perform further corrections such as sample extended geometry and γ self-absorption (see the measurement model eq. 35). In particular, the sample

height (in mm) and sample density (in g cm^{-3}) values are requested in the corresponding spinboxes labeled `'sample height / mm'` and `'density / g cm-3'`, respectively.

It is worth to notice that any input parameter has also place to introduce the corresponding standard uncertainty.

The button confirms the submitted information while checking the consistency of the inserted data. The options labeled `'selected'`, `'all undefined'` and `'all'` indicate to which spectrum those data are applied, albeit it is ineffective in this instance since only a single spectrum can be recalled as standard. The options in the checkbuttons labeled `'find peaks'` and `'clear previous'` define whether the software should perform an automatic search to identify peaks in the corresponding spectrum; when the first option is checked the software performs an automatic assignment of each peak of the peaklist to an emission label, only in case the match is unambiguous; if a sample is chosen, only emissions deriving from the targets in the selected sample composition are taken into account otherwise all entries from the k_0 database are tested. This functionality does not overwrite a selection that was previously assigned, thus, checking the second option overrides this behavior allowing to clean the selection before starting the automatic assignment.

3.7.5 Select counting position

The standard counting position region at the bottom of the standard section comprises a radiobutton, a slider with indication of nominal counting positions, and spinboxes to introduce value and uncertainty for the parameter taking into account vertical variability with respect to the nominal position.

When a detector characterization is selected from the corresponding drop-down menu in the main window the counting distance identified as the reference (the counting distance labeled as reference in the *New detector characterization* task, paragraph 3.3.1) is automatically set for both sliders referred to standard and sample. The k0-INRIM software allows to manage two different counting positions for sample and standard [9], however, one of the two has to be at the reference position. Thus, when the radiobutton is activated, the corresponding slider can be moved to any other counting position, with closer sample-detector distances going towards the left-hand side, otherwise it is set on the reference counting position and unable to move.

The value reported on the slider indicates the nominal counting position, that is the position where the calibrated source was while the detector characterization has been performed. Counting positions of sample and standard are defined as actual counting positions. To get from the nominal counting position to the actual counting position the value of the spinbox labeled `'Δd / mm'` should be adjusted according to:

$$d_{\text{actual}} = d_{\text{nominal}} + \Delta d, \tag{32}$$

with, d_{actual} the actual counting position, d_{nominal} the nominal counting position and Δd the difference between actual and nominal counting positions, respectively.

In the convention adopted in this software all positions are taken into account at the bottom of the corresponding sample (either calibration sources, standards, samples). The axis of coordinates has its 0 at the detector endcap while, as a general rule, distances with positive signs are going upwards and negative signs are going downwards.

3.8 Sample

The sample section of the main window allows to introduce information about the γ-spectra containing the target elements to be investigated. It comprises a *Open sample* button, a drop-down menu with a spectrum counter to identify the name of spectrum currently selected and the total number of recalled spectra, a *Sample peaklist* button to display data of the selected spectrum and the peaklist (the list of all the elaborated peaks of the selected spectrum), a *Delete sample* button with a switcher to delete spectra and a *Define material* button to introduce information related to the physical sample. In addition, it provides a slider with a radiobutton and spinboxes to identify the actual counting position of the sample.

3.8.1 Open sample

The *Open sample* task is similar to that for the background spectrum (see paragraph 3.6.1) with the only difference that multiple selection is here allowed.

3.8.2 Sample peaklist

The *Sample peaklist* button allows to browse information about the currently selected spectrum. The peaklist window shows information similarly to the standard peaklist window (see paragraph 3.7.2) (Figure 34).

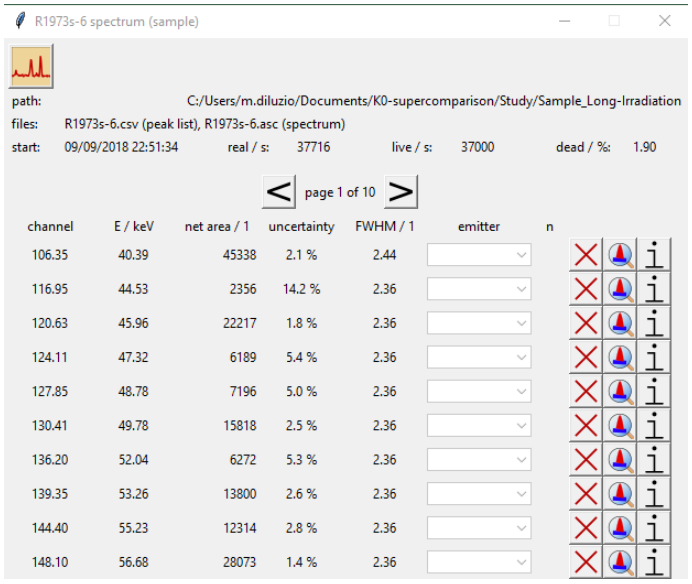


Figure 37: The sample peaklist window

The difference with the standard peaklist windows is that, the column labeled `'k0'` is not present and all the emissions identified in the column labeled `'emitter'` are investigated in the analysis.

3.8.3 Delete sample

The *Delete sample* task is similar to that for the background spectrum (see paragraph 3.6.3), with the difference that since in the sample section multiple spectra are allowed, there is an actual difference in behavior whether selecting the `'selected'` or `'all'` label from the switcher.

3.8.4 Define material

The *Define material* task relative to sample initiate a window (Figure 38) allowing to introduce information concerning the investigated material, similarly to the standard window (see paragraph 3.7.4).

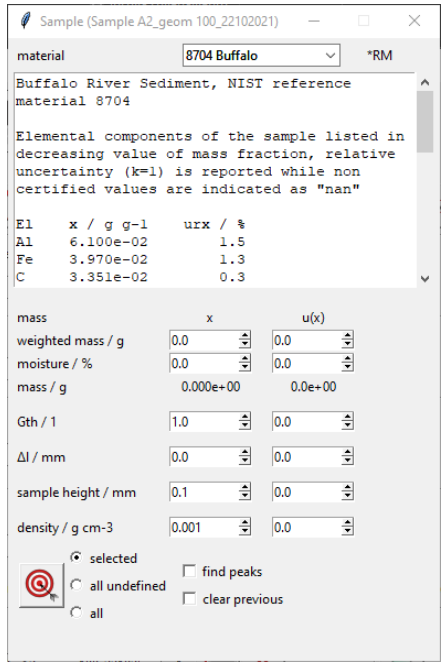


Figure 38: The window to recall spectra

The only difference with it is that the spinbox to insert G_e information is replaced by the Δl one (see eq. 35) labeled `'Δl / mm'`. This input refers to the difference in the vertical placement within the irradiation channel of sample with respect to the standard. Also in this case the coordinate convention is maintained, thus, this distance has negative sign if the sample is lower with respect to the standard or positive otherwise with the only difference being the reference points; this distance should be calculated between the centers of the sample and standard involved since the correction based on it adopts a linear approximation.

Also, in case multiple samples are recalled the options `'selected'`, `'all undefined'` and `'all'` have effect: `'selected'` applies the provided data only to the currently selected spectrum, `'all undefined'` applies the provided data to all spectra for which the defines spectrum task is not performed yet and `'all'` applies the provided data to all spectra, thus also, those that have been already defined are overwritten.

Lastly, if the type of selected material was defined as "Reference material" during its creation (see paragraph 3.2.2), a label `'*RM'` appears next to the material name to notify the user that the current selection is suitable for performing validation of results task (see paragraph 3.10.1).

3.8.5 Select counting position

The sample counting position region at the bottom of the sample section shares similar features and actions with respect to the standard counting position region (see paragraph 3.7.5).

However, it is worth noticing that the counting position information introduced in this region are applied to all spectra in the sample list.

3.9 Limits

The limits section comprises a *Detection limits* button and a counter label. The first one allows to select the elements for which detection limits have to be investigated, in case their corresponding peaks are not found in the sample spectra, the second one informs the user about the number of target elements currently selected.

A periodic table-like subwindow pops us by pressing the button (Figure 39).

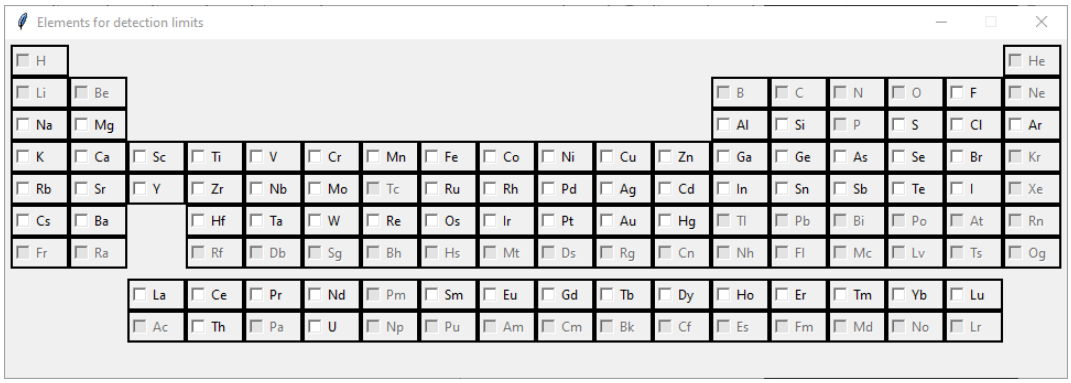


Figure 39: The element selection window

Among all elements, only those present in the adopted k_0-database are activated and selectable. Once a target element is checked, by clicking upon its cell, it will be taken into account for detection limits calculations, and the counter label is promptly updated. To deselect an element just click on an already selected cell.

3.10 Results

The Results section comprises a *Elaborate* button. This one performs the analysis with all the input data provided and reports the results in different ways.

3.10.1 Elaborate

The elaboration starts by clicking the button. A number of checks are performed on the input data to verify if all conditions are met. In case some condition is not satisfied a subwindow appears giving information about what input data caused the issue.

If all checks are passed, the elaboration subwindow appears giving a general overview of the analysis and all actions that can be performed on it (Figure 40).

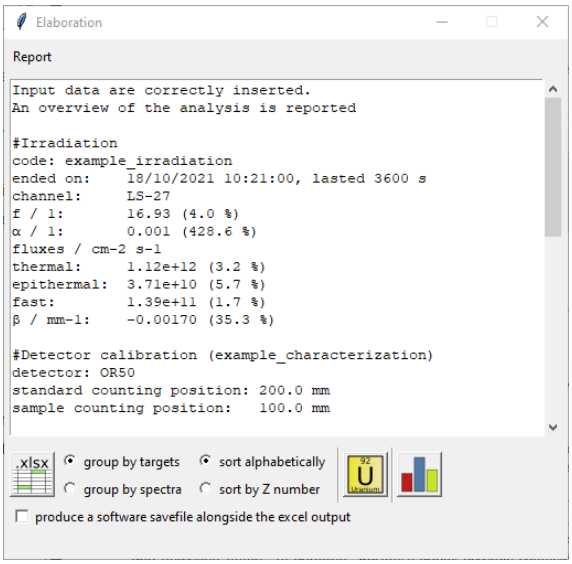


Figure 40: The elaboration report window

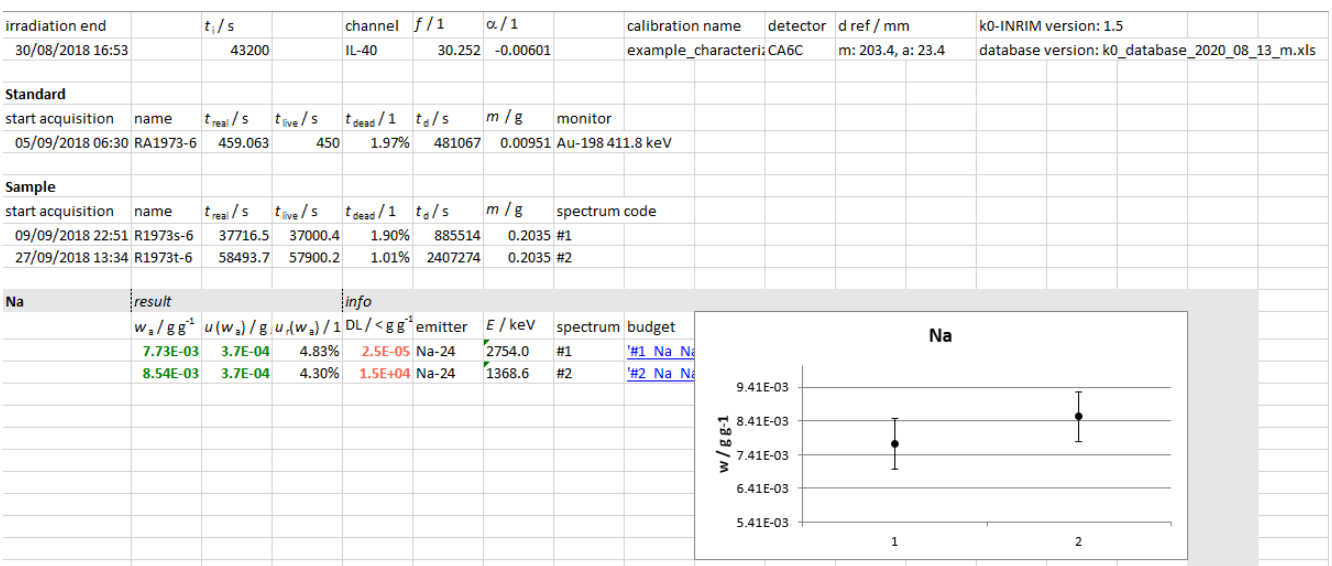
At the top of the window the log labeled `'Report'` contains information concerning irradiation (neutron exposure data and adopted irradiation channel), detector characterization (adopted detector and selected counting positions), background (whether is selected a background for background correction), standard (spectra data and monitor information) and samples (spectra data and number of analyte peaks selected). At the end of the report a summary is found indicating how many budgets will be issued if the elaboration is exported as it is and discriminates between quantified and detection limits. In addition, warnings about possible conflicts among uncertainty budgets (whether the same emitter is identified in more peaks) are reported here. This kind of conflicts are automatically managed by assigning a unique peak to a single emitter (the first occurrence) or it might be resolved manually from the peak selection in the sample peaklist window (see paragraph 3.8.2).

Uncertainty budgets are statements of a measurement uncertainty, of their components and of their calculation and combination [12]. Each budget is translated in an instance of a python class that includes all the required information to compute the mass fraction of an analyte, or its detection limit if a peak is not identified. All calculations, and corrections, are internally computed within that class; the resulting values are calculated though the measurement model adopted in the software (see section 4, eq. 35) together with the complete uncertainty budget output. Within the

scope of the current analysis each uncertainty budget is represented by a unique code $\#n_S_T_S_E$-$AA\ EEE.E$ where n is the index of the spectra (an integer depending on the list of sample spectra based on the uploading order), S_T represents the symbol of target element that is quantified, S_E represents the symbol of the emitter element adopted to evaluate the target S_T with AA the mass number identifying the isotope, $EEE.E$ is the γ-emission energy adopted for the evaluation (e.g. #1_Sc_Sc-46 889.3 refers to the uncertainty budget obtained from elaboration performed on sample spectrum with index 1 aimed to quantify Sc from the emission 889.3 keV of Sc-46).

At the bottom of the window three options applicable to the uncertainty budgets can be activated with the *Process Excel output*, *Overview* and *Perform a validation inspection* buttons.

The button exports the uncertainty budgets in a Microsoft Excel spreadsheet which also includes a summary (found as first sheet of the document) and a section reporting measurement model data and explanations (found as last sheet of the document). It provides two options to group and sort budgets that will slightly change the appearance of the summary page. The group option allows to choose between `'group by targets'` and `'group by spectra'`; the first one groups together all the emissions deriving from the same target across all spectra (Figure 41) while the second one groups all emissions from different targets found in the same spectrum (Figure 42). The sort option allows to choose between `'sort alphabetically'` and `'sort by Z number'`; the first lists the target elements within a same group alphabetically while the second lists them by increasing atomic number.



irradiation end		t_i / s		channel	f / 1	α / 1		calibration name	detector	d ref / mm	k0-INRIM version: 1.5
30/08/2018 16:53		43200		IL-40	30.252	-0.00601		example_characteri	CA6C	m: 203.4, a: 23.4	database version: k0_database_2020_08_13_m.xls

Standard

start acquisition	name	t_{real} / s	t_{live} / s	t_{dead} / 1	t_d / s	m / g	monitor
05/09/2018 06:30	RA1973-6	459.063	450	1.97%	481067	0.00951	Au-198 411.8 keV

Sample

start acquisition	name	t_{real} / s	t_{live} / s	t_{dead} / 1	t_d / s	m / g	spectrum code
09/09/2018 22:51	R1973s-6	37716.5	37000.4	1.90%	885514	0.2035	#1
27/09/2018 13:34	R1973t-6	58493.7	57900.2	1.01%	2407274	0.2035	#2

Na	*result*			*info*				
	w_a / g g^{-1}	$u(w_a)$ / g	$u_r(w_a)$ / 1	DL / < g g^{-1}	emitter	E / keV	spectrum	budget
	7.73E-03	3.7E-04	4.83%	2.5E-05	Na-24	2754.0	#1	'#1_Na_Na
	8.54E-03	3.7E-04	4.30%	1.5E+04	Na-24	1368.6	#2	'#2_Na_Na

Figure 41: The summary sheet with `'group by targets'` option activated

irradiation end		t_i / s		channel	f / 1	α / 1		calibration name	detector	d ref / mm		k0-INRIM version: 1.5			
30/08/2018 16:53		43200		IL-40	30.252	-0.00601		example_characteri:	CA6C	m: 203.4, a: 23.4		database version: k0_database_2020_08_13_m.xls			
Standard															
name	RA1973-6														
start acquisition		t_{real} / s	t_{live} / s	t_{dead} / 1	t_d / s	m / g	monitor								
05/09/2018 06:30		459.063	450	1.97%	481067	0.00951	Au-198 411.8 keV								
Sample															
name	**R1973s-6**														
spectrum info															
start acquisition		t_{real} / s	t_{live} / s	t_{dead} / 1	t_d / s	m / g									
09/09/2018 22:51		37716.5	37000.4	1.90%	885514	0.2035									
element	*result*			*info*											
target	w_a / g g^{-1}	$u(w_a)$ / g	$u_r(w_a)$ / 1	DL / < g g^{-1}	emitter	E / keV	budget								
Na	7.73E-03	3.7E-04	4.83%	2.5E-05	Na-24	2754.0	'#1_Na_Na-24 2754.0'!A1								
K	2.10E-02	6.1E-03	29.12%	3.1E-02	K-42	1524.7	'#1_K_K-42 1524.7'!A1								
Ca	This comp	0.0E+00	-	0.0E+00	Sc-47	159.4	'#1_Ca_Sc-47 159.4'!A1								
Ca	3.47E-02	2.4E-03	6.85%	2.3E-02	Ca-47	489.2	'#1_Ca_Ca-47 489.2'!A1								
Ca	2.61E-02	2.8E-03	10.86%	1.6E-02	Ca-47	807.9	'#1_Ca_Ca-47 807.9'!A1								
Ca	3.13E-02	1.4E-03	4.42%	1.4E-03	Ca-47	1297.1	'#1_Ca_Ca-47 1297.1'!A1								
Sc	4.64E-06	2.1E-07	4.42%	6.2E-09	Sc-46	889.3	'#1_Sc_Sc-46 889.3'!A1								

Figure 42: The summary sheet with `'group by spectra'` option activated

In both cases, the summary page gives valuable general information about the software itself (versions of code and k_0 database) and experimental setup (irradiation and γ-acquisition data with statement of adopted irradiation channel and flux parameters, counting positions, detector). Also, results are reported and links to the corresponding uncertainty budget sheets are provided.

Each uncertainty budget sheet provides a header with information concerning target, emitter and emission energy. Underneath that, a table with all input parameters is found. For each input, data as unit, value, standard uncertainty, relative uncertainty, sensitivity coefficient and contribution to variance are reported, plus two intermediate data ($y\left(x_i^+\right)$ and $y\left(x_i^-\right)$ corresponding to the resulting y obtained by increasing and decreasing x_i by 1 standard uncertainty) that are hidden by default (Figure 43). The sensitivity coefficients are numerically evaluated from the measurement model:

$$\frac{\partial y}{\partial x_i} \approx \frac{y\left(x_i + u\left(x_i\right)\right) - y\left(x_i - u\left(x_i\right)\right)}{2u\left(x_i\right) + \delta x}, \tag{33}$$

with $\frac{\partial y}{\partial x_i}$ the sensitivity coefficient referred to the x_i input parameter with $u(x_i)$ its uncertainty, $y\left(x_i \pm u\left(x_i\right)\right)$ is the evaluation of y depending on the variation of the x_i value, and δx is a negligible offset in the order of 2×10^{-9} introduced to make sure the program doesn't raise an error if $u(x_i)$ is set to 0.

Target	Sc	Emitter	Sc-46	E_p / keV	889.3			
Quantity		Unit	Value	Std unc	Rel unc	Sensitivity coef.	Contribution to varia	
X_i		$[X_i]$	x_i	$u(x_i)$	$u_r(x_i)$	c_i	I / %	
t_i		s	43200	30	0.07%	-6.53595E-12	0.00%	
n_{pa}		1	848568.8	957.4	0.11%	5.46768E-12	0.07%	
λ_a		s^{-1}	9.57E-08	2.28E-11	0.02%	-44.19064154	0.00%	
Δt_d		s	404447	0.2	0.00%	4.4402E-13	0.00%	
t_{ca}		s	37716.5	0.2	0.00%	2.21876E-13	0.00%	
t_{la}		s	37000.4	0.2	0.00%	-1.25396E-10	0.00%	
COI_a		1	0.920414	0.015917	1.73%	-5.04239E-06	15.32%	
m_{sm}		g	0.2035	0.00008	0.04%	-2.27995E-05	0.01%	
η_{sm}		1	0	0	-	4.6397E-06	0.00%	
$k_{0Au}(a)$		1	1.22	0.00488	0.40%	-3.80309E-06	0.82%	
G_{tha}		1	1	0	0.00%	-4.57412E-06	0.00%	
G_{ea}		1	1	0	0.00%	-6.55777E-08	0.00%	
Q_{0a}		1	0.43	0.086	20.00%	-1.59165E-07	0.45%	
E_{ra}		eV	5130	872.1	17.00%	-1.88216E-16	0.00%	

Figure 43: An uncertainty budget sheet, input data

At the right of the input parameter table lie the correlation and covariance matrices. The correlation matrix is produced by the software which takes care of the inputs that are strictly correlated to each other (as it is the case in relative analysis). The covariance matrix is then calculated upon it by including also the variance of single parameters (Figure 44).

Corr. Matrix	t_i	n_{pa}	λ_a	Δt_d	t_{ca}	t_{la}	COI_a	m_{sm}	η_{sm}	$k_{0Au}(a)$	G_{tha}
t_i	1	0	0	0	0	0	0	0	0	0	0
n_{pa}	0	1	0	0	0	0	0	0	0	0	0
λ_a	0	0	1	0	0	0	0	0	0	0	0
Δt_d	0	0	0	1	0	0	0	0	0	0	0
t_{ca}	0	0	0	0	1	0	0	0	0	0	0
t_{la}	0	0	0	0	0	1	0	0	0	0	0
COI_a	0	0	0	0	0	0	1	0	0	0	0
m_{sm}	0	0	0	0	0	0	0	1	0	0	0
η_{sm}	0	0	0	0	0	0	0	0	1	0	0
$k_{0Au}(a)$	0	0	0	0	0	0	0	0	0	1	0
G_{tha}	0	0	0	0	0	0	0	0	0	0	1
G_{ea}	0	0	0	0	0	0	0	0	0	0	0
Q_{0a}	0	0	0	0	0	0	0	0	0	0	0

Figure 44: An uncertainty budget sheet, matrices

At the bottom of the uncertainty budget sheet the result is calculated (see section 4) including value, standard uncertainty and relative uncertainty if the peak is identified, otherwise only the detection limit value is issued. If in the *define material* window a composition is selected (see paragraph 3.8.4) the γ self-absorption correction is automatically evaluated; moreover, if a mass fraction value for the target element is found in the selected composition, the zeta score, z_{score}, is also reported:

$$z_{\text{score}} = \frac{w_{\text{meas}} - w_{\text{true}}}{\sqrt{u^2\left(w_{\text{meas}}\right) + u^2\left(w_{\text{true}}\right)}}, \tag{34}$$

with, w_{meas} the measured mass fraction, w_{true} is the mass fraction for the same element assumed

as the true value with $u^2\left(w_{\mathrm{meas}}\right)$ and $u^2\left(w_{\mathrm{true}}\right)$ their variances, respectively.

Additional information (Figure 45) are reported such as $\frac{\varepsilon_{\mathrm{geo\,m}}}{\varepsilon_{\mathrm{geo\,a}}}$, $k_{\varepsilon\Delta\mathrm{d\,a}}$, ω_{a}, $F_{\mathrm{abs\,a}}$, $k_{\varepsilon\Delta\mathrm{d\,m}}$, ω_{m}, $F_{\mathrm{abs\,m}}$, $Q_{0\,\mathrm{a}}\left(\alpha\right)$, $Q_{0\,\mathrm{m}}\left(\alpha\right)$, $\left(1+\beta\Delta l\right)$, m_{a}, $m_{\mathrm{a\,blank}}$, $n_{\mathrm{p\,a'}}$, $n_{\mathrm{p\,b}}$, $n_{\mathrm{p\,i}}$, $n_{\mathrm{p\,f}}$ (see section 4). The cells referring to $n_{\mathrm{p\,i}}$ and $n_{\mathrm{p\,f}}$ values, that are blank by default, can be filled with γ-interference correction and fast activation correction as their effect on the analyte peak evaluated in number of counts (see section 4), respectively. As a last cell, a link to the measurement model sheet is present.

Quantity	Unit	Value	Std unc	Rel unc	Contribution to variance
Y	$[y]$	y	$u(y)$	$u_r(y)$	l / %
w_a	$g\,g^{-1}$	4.64E-06	2.1E-07	4.42%	100.00%
DL	$<g\,g^{-1}$	6.2E-09			
zeta score	1	-			

Additional information

Quantity	Unit	Value	Std unc	Rel unc	Contribution to variance
X_i	$[X_i]$	x_i	$u(x_i)$	$u_r(x_i)$	l / %
$\varepsilon_{geo\,m}$ / $\varepsilon_{geo\,a}$	1	0.100401	-	-	2.14%
$k\varepsilon\Delta d\,a$	1	1.034611	-	-	
ω_a	1	1.076259	-	-	
$F_{abs\,a}$	1	1	-	-	
$k\varepsilon\Delta d\,m$	1	1.012469	-	-	
ω_m	1	1.000444	-	-	
$F_{abs\,m}$	1	0.999999	-	-	
$Q_{0,a}(\alpha)$	1	0.433713	-	-	
$Q_{0,m}(\alpha)$	1	15.86337	-	-	
$1+\beta\,\Delta l$	1	0.982075	-	-	
m_a	g	9.44E-07	-	-	
$m_{a\,blank}$	g	0	-	-	
$n_{p\,a'}$	1	848568.8	957.4	0.11%	
$n_{p\,b}$	1	0	0	-	
$n_{p\,i}$	1			-	
$n_{p\,f}$	1			-	

Measurement model

'Models'!A1

Figure 45: An uncertainty budget sheet, results and additional information sections

It is important to notice that, aside for values and uncertainties of input parameters and correlation matrix, which are filled with numbers written by the k0-INRIM software, all other cells compute their result in real time since Excel functions are implemented in them; it means that modifying values in input cells also modifies all cells linked to them immediately displaying the effect to the result.

The last sheet of the workbook includes images of the measurement models implemented in the software with explanation of the adopted symbols (Figure 46).

Type I activation-decay

$$w_{\mathrm{a}} = \left(\frac{\lambda \frac{(n_{\mathrm{p}}/COI)(t_{\mathrm{c}}/t_{\mathrm{l}})e^{\mu(1-t_{\mathrm{l}}/t_{\mathrm{c}})}}{(1-e^{-\lambda t_{\mathrm{i}}})(1-e^{-\lambda t_{\mathrm{c}}})}\Big|_{\mathrm{a}} e^{(\lambda_{\mathrm{a}}-\lambda_{\mathrm{m}})t_{\mathrm{d\,m}}+\lambda_{\mathrm{a}}\Delta t_{\mathrm{d}}} \frac{1}{1+\beta\Delta l_{\mathrm{a}}} \frac{k_{0\,\mathrm{Au}}(\mathrm{m})}{k_{0\,\mathrm{Au}}(\mathrm{a})} \frac{G_{\mathrm{th\,m}} + \frac{G_{\mathrm{e\,m}}}{f}\left(\frac{Q_{0\,\mathrm{m}}-0.429}{E_{\mathrm{r\,m}}^{\alpha}} + \frac{0.429}{0.55^{\alpha}(1+2\alpha)}\right)}{G_{\mathrm{th\,a}} + \frac{G_{\mathrm{e\,a}}}{f}\left(\frac{Q_{0\,\mathrm{a}}-0.429}{E_{\mathrm{r\,a}}^{\alpha}} + \frac{0.429}{0.55^{\alpha}(1+2\alpha)}\right)} k_{\varepsilon\Delta E}\, k_{\varepsilon\Delta \mathrm{d_{ref}}} \frac{\left(\frac{\mathrm{d_{ref\,m}}-d'_{0\,\mathrm{m}}}{\mathrm{d_{ref\,m}}+\Delta d_{\mathrm{m}}-d'_{0\,\mathrm{m}}}\right)^2}{\left(\frac{\mathrm{d_{ref\,a}}-d'_{0\,\mathrm{a}}}{\mathrm{d_{ref\,a}}+\Delta d_{\mathrm{a}}-d'_{0\,\mathrm{a}}}\right)^2} \frac{1-}{1-} \right.$$

Parameters

w_{a} : mass fraction of analyte in the sample / g g^{-1}

λ_{a} : decay constant of analyte emitter / s^{-1}

$n_{\mathrm{p\,a}}$: net peak area (corrected for counts due to background, interferent peaks and fast activations) of analyte / 1

COI_{a} : true coincidence correction factor for analyte emission / 1

$t_{\mathrm{c\,a}}$: real counting time of sample spectrum / s

$t_{\mathrm{l\,a}}$: live counting time of sample spectrum / s

μ : excess counting loss constant of the detection system / 1

t_{i} : irradiation time of sample and standard / s

λ_{m} : decay constant of monitor emitter / s^{-1}

$n_{\mathrm{p\,m}}$: net peak area (corrected for counts due to background, interferent peaks and fast activations) of monitor / 1

COI_{m} : true coincidence correction factor for monitor emission / 1

t : real counting time of standard spectrum / s

Figure 46: The measurement model sheet

The filename and directory of the Excel output is selected by the user after pressing the

button. If the checkbutton labeled `'produce a software savefile alongside excel output'` is checked a typical .sav file is created alongside the Excel output and saved on the same destination; this .sav file provides a way to recall the whole elaboration process, if the file is moved into the «saves» folder the whole elaboration can be recalled by the software.

The

button displays the results evaluated from the current uncertainty budgets in a periodic table window without providing any output file (Figure 47). It might be useful for a fast consistency check of the analytical results before exporting the Excel file.

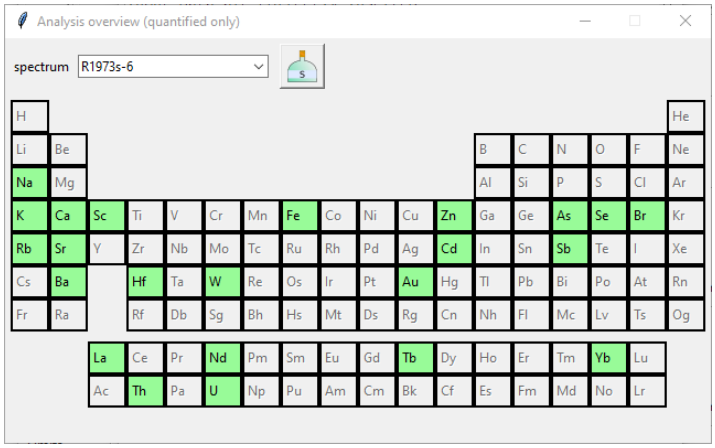


Figure 47: The quantified elements overview window

The drop-down menu labeled `'spectrum'` allows choosing among the investigated sample spectra. In the periodic table-like section quantified elements within the selected spectrum are highlighted in green. It is possible to click on highlighted element buttons to open subwindows containing useful information concerning the list of emitters adopted to quantify the target element such as

emitter identity, mass fraction, relative uncertainty, detection limit, and z_{score}, labeled `'emitter'`, `'w / g g-1'`, `'urw / %'`, `'DL / g g-1'` and `'z / 1'`, respectively (Figure 48).

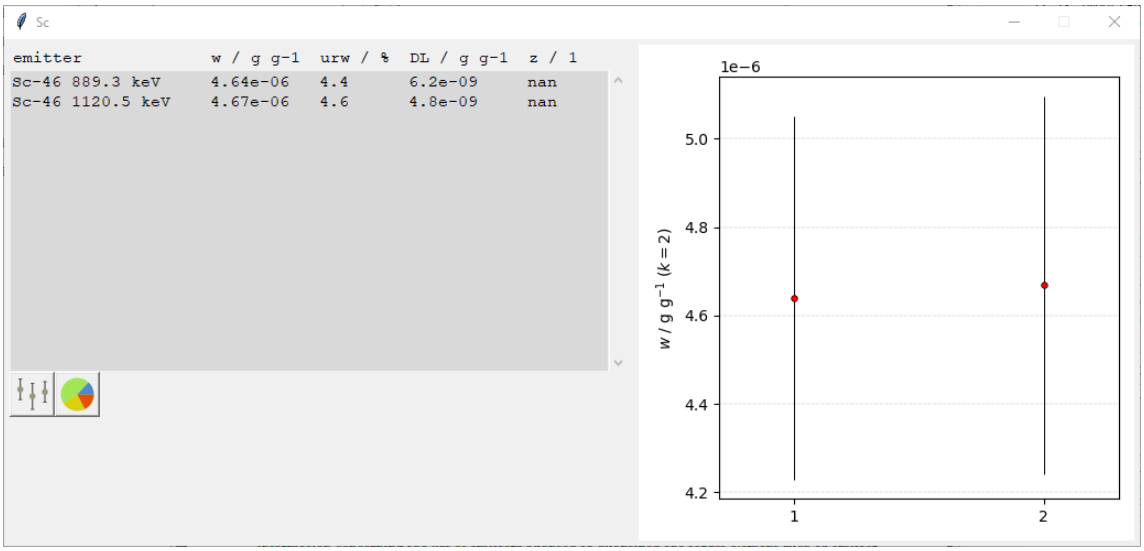


Figure 48: The quantified element subwindow, overview

In addition, the graph at the right-hand side displays a plot of mass fractions for all entries in the list. Conversely, while one of the items of the list is selected, pressing the *Display contributors to variance* button will show a pie chart displaying the five most important contributors to the combined uncertainty of the selected emission (Figure 49).

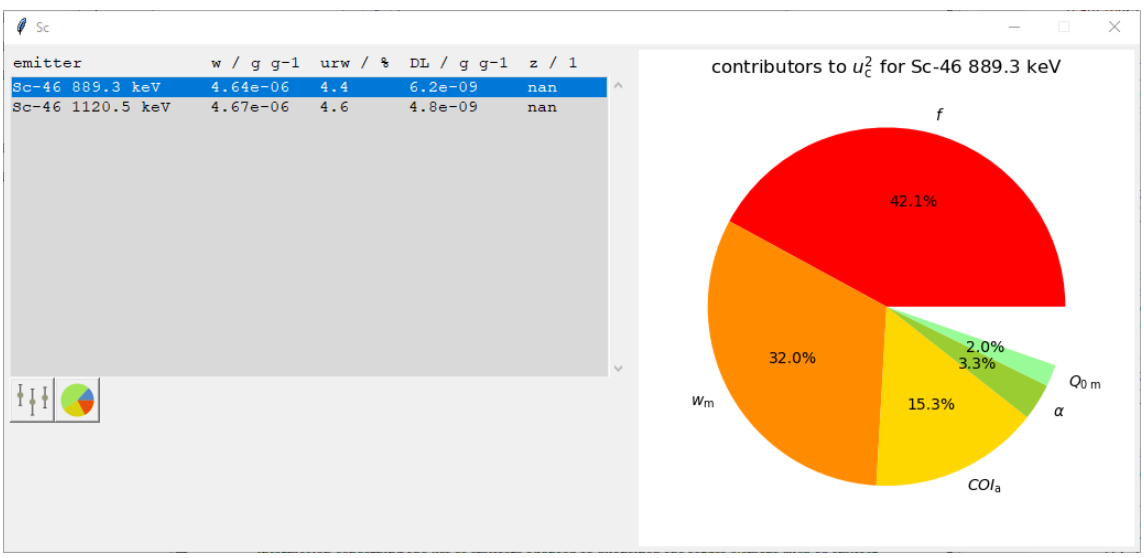


Figure 49: The quantified element subwindow, contribution to variance

The *Display overview plot* button, instead, reverts to the overview plot (Figure 48).

The *Save as material* button allows to convert the data displayed in the periodic table-like window into material data type usable by the software (Figure 50). The subwindow that pops up provides an overview of the elements spotted in the analysis of the selected sample and a radiobutton to choose how to manage multiple emitter: whether to adopt an aritmethic or weighted (using variances as weights) average and whether adopting Inter Quartile Range (IQR) or manual selection for outliers management.

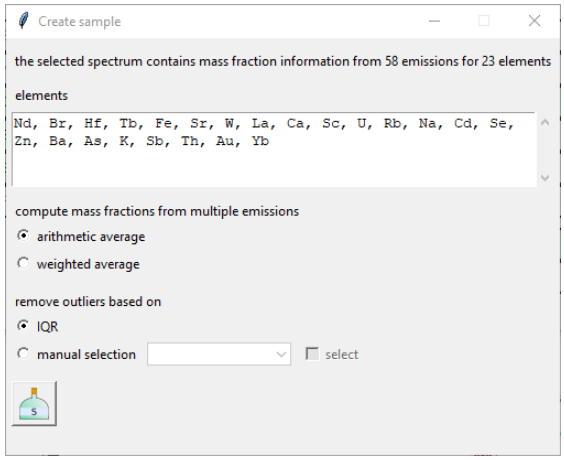
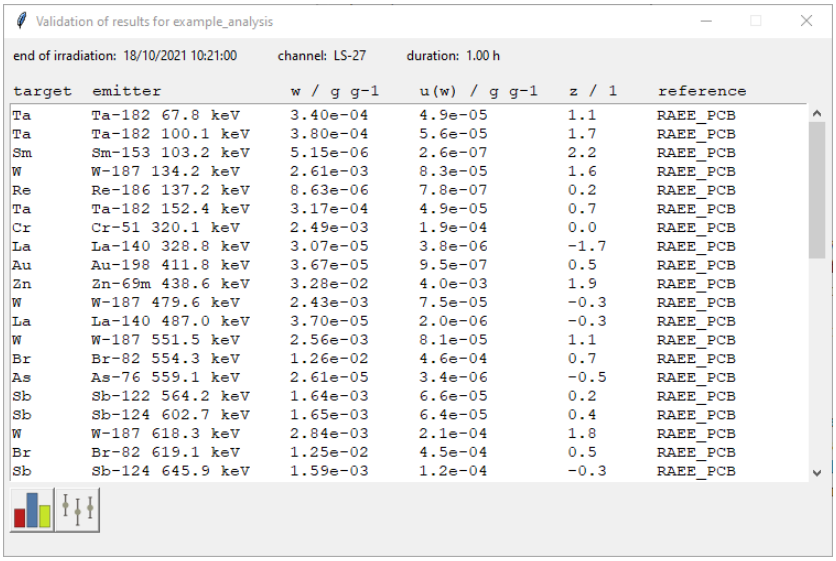


Figure 50: The save as sample quantified element subwindow

The button calculates the concentration of the target elements and opens a material window similar to that in the browse database subsection (see paragraph 3.2.2) to further modify and save the information. At this point any modification of the material is still possible (name, description, composition) and is saved on disc through button while is discarded by closing the subwindow.

The button selects and displays data from the current analysis for validation purposes; the required condition to perform this task is that one or more samples should be labeled with the tag *RM (see paragraph 3.8.4) meaning that is regarded by the software as a reference material. The measured elements' mass fractions for the sample are compared with the stored composition data (Figure 51).

The validation of results window displays information about the reference material under investigation; in particular, experimental setup data are reported as the end of irradiation, channel name and duration of irradiation while for each quantified emitter data as target, γ-emission, measured mass fraction, evaluated uncertainty, z_{score} and material name are listed, in the columns `'target'`, `'emitter'`, `'w / g g-1'`, `'u(w) / g g-1'`, `'z / 1'` and `'reference'`, respectively.



Validation of results for example_analysis

end of irradiation: 18/10/2021 10:21:00 channel: LS-27 duration: 1.00 h

target	emitter	w / g g-1	u(w) / g g-1	z / 1	reference
Ta	Ta-182 67.8 keV	3.40e-04	4.9e-05	1.1	RAEE_PCB
Ta	Ta-182 100.1 keV	3.80e-04	5.6e-05	1.7	RAEE_PCB
Sm	Sm-153 103.2 keV	5.15e-06	2.6e-07	2.2	RAEE_PCB
W	W-187 134.2 keV	2.61e-03	8.3e-05	1.6	RAEE_PCB
Re	Re-186 137.2 keV	8.63e-06	7.8e-07	0.2	RAEE_PCB
Ta	Ta-182 152.4 keV	3.17e-04	4.9e-05	0.7	RAEE_PCB
Cr	Cr-51 320.1 keV	2.49e-03	1.9e-04	0.0	RAEE_PCB
La	La-140 328.8 keV	3.07e-05	3.8e-06	-1.7	RAEE_PCB
Au	Au-198 411.8 keV	3.67e-05	9.5e-07	0.5	RAEE_PCB
Zn	Zn-69m 438.6 keV	3.28e-02	4.0e-03	1.9	RAEE_PCB
W	W-187 479.6 keV	2.43e-03	7.5e-05	-0.3	RAEE_PCB
La	La-140 487.0 keV	3.70e-05	2.0e-06	-0.3	RAEE_PCB
W	W-187 551.5 keV	2.56e-03	8.1e-05	1.1	RAEE_PCB
Br	Br-82 554.3 keV	1.26e-02	4.6e-04	0.7	RAEE_PCB
As	As-76 559.1 keV	2.61e-05	3.4e-06	-0.5	RAEE_PCB
Sb	Sb-122 564.2 keV	1.64e-03	6.6e-05	0.2	RAEE_PCB
Sb	Sb-124 602.7 keV	1.65e-03	6.4e-05	0.4	RAEE_PCB
W	W-187 618.3 keV	2.84e-03	2.1e-04	1.8	RAEE_PCB
Br	Br-82 619.1 keV	1.25e-02	4.5e-04	0.5	RAEE_PCB
Sb	Sb-124 645.9 keV	1.59e-03	1.2e-04	-0.3	RAEE_PCB

Figure 51: The window for analysis validation

The *plot z values for all results* and *plot element agreement with reference* buttons provide graphical visualization of data present in the validation of results window. The task accessed from button allows to display an overview of the z_{score} values for the whole dataset (Figure 52). All values are represented in a vertical bar chart with any emission (in increasing γ-energy) on the abscissa and z_{score} on the ordinate. Regions including z_{score} values satisfying $2 \leq |z_{\text{score}}| \leq 3$ and $3 < |z_{\text{score}}| < \infty$ are highlighted with light green and light red colors, respectively, in order to clearly identify which data fall in those critical areas (green for questionable values, red for outliers); this outcome is also resumed in the title of the graph together with the total number of emissions under investigation.

At the bottom of the window, a slider and a few buttons are present to perform actions on the visual aspect of the graphic. The *adjust left y axis* button works in combination with the adjacent slider to symmetrically change the limits of the ordinate axis; the *reset axis when resized* button spreads the graphic over the available window area when the latter is manually resized; the *show/hide the legend* button alternatively shows or hides the figure legend, by clicking it multiple times also moves the position of the legend counterclockwise; the *show/hide ticklabels* button shows or hides the labels on the abscissa used to identify the investigated emissions; the *save figure* button saves the current figure into a .png file with name and destination chosen by the user.

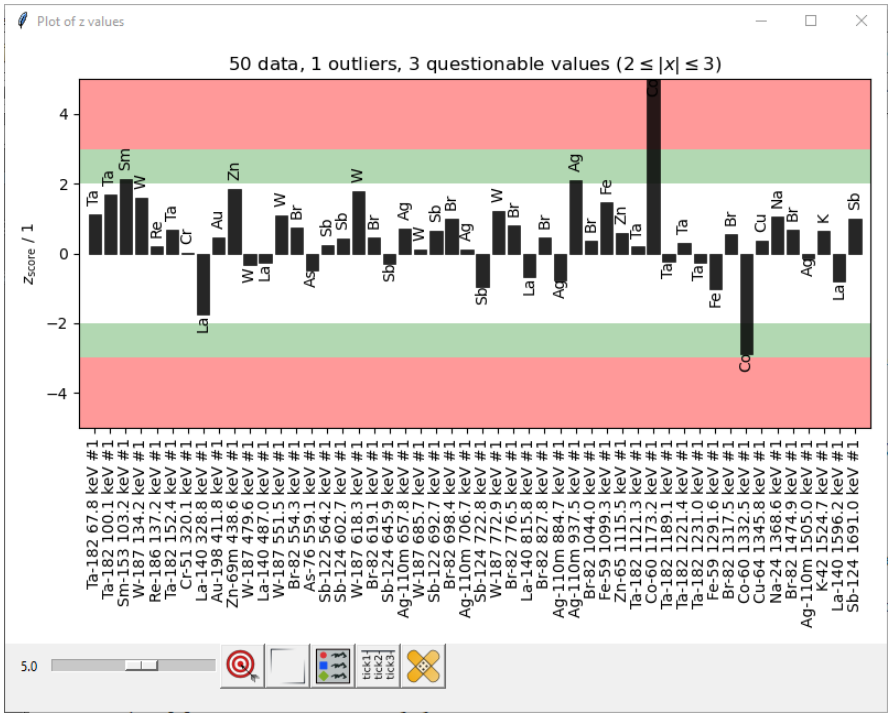


Figure 52: The window for analysis validation, z_{score} graph

The button produces an element by element series of graphics depicting the agreement between the measured and reference mass fraction values (Figure 53).

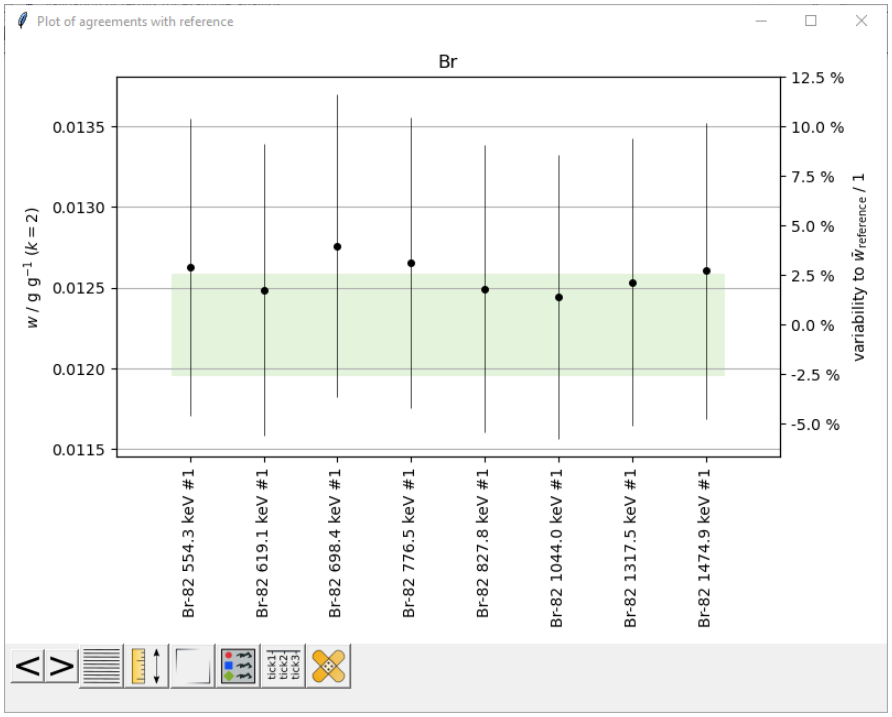


Figure 53: The window for analysis validation, targets agreement graph

In the plot of agreements with reference window, data for a single element are displayed at a time; the measured mass fractions (with extended uncertainties, $k = 2$) deriving from the corresponding investigated emissions are depicted together with a light green area representing the confidence interval ($k = 2$, approximately 95%) of the reference value. The graphic includes two ordinate axes, the left one expresses mass fraction with unit g g^{-1} while the right one expresses variability to the reference mass fraction as $\frac{w_i - w_{\mathrm{ref}}}{w_{\mathrm{ref}}}$; in case multiple reference materials are selected for the samples a weighted average is calculated to return w_{ref}.

The buttons at the bottom of the window are < *go to previous element*, > *go to following element*, *switch axis of y grid*, *adjust right axis limits*, *reset axes when resized*, *show/hide the legend*, *show/hide ticklabels* and *save figure*. The < and > buttons allow to change the currently visualized element over a list sorted in alphabetical order; the title of the graphic indicates the current element while hovering on the < and > buttons suggests what are the previous and next elements. The button allows to alternate the horizontal grid between the two axes. The button opens a small frame 83.70 -61.77 with two entries allowing enter the new limits (higher limit on top, lower limit at the bottom) of the right-hand axis as percent values and a button to confirm the changes. The remaining buttons perform the same actions of the similar buttons in the plot of z values window (Figure 52).

4 Measurement model

The measurement model adopted in the k0-INRIM software derives from the original equation of k_0 method [3] but is modified to comply with the experimental setup, adopted by the k0-INRIM, and application of uncertainty propagation formula [1].

For simple activation-decay types (I, IIB, IVB, VI) the measurement model is as follows:

$$
\begin{aligned}
w_{\mathrm{a}} = \frac{1}{m_{\mathrm{sm}}\left(1-\eta_{\mathrm{sm}}\right)} \Bigg(& \frac{\lambda \frac{(n_{\mathrm{p}}/COI)(t_{\mathrm{c}}/t_{\mathrm{l}})e^{\mu(1-t_{\mathrm{l}}/t_{\mathrm{c}})}}{(1-e^{-\lambda t_{\mathrm{i}}})(1-e^{-\lambda t_{\mathrm{c}}})}\Big|_{\mathrm{a}}}{\lambda \frac{(n_{\mathrm{p}}/COI)(t_{\mathrm{c}}/t_{\mathrm{l}})e^{\mu(1-t_{\mathrm{l}}/t_{\mathrm{c}})}}{(1-e^{-\lambda t_{\mathrm{i}}})(1-e^{-\lambda t_{\mathrm{c}}})}\Big|_{\mathrm{m}}} e^{(\lambda_{\mathrm{a}}-\lambda_{\mathrm{m}})t_{\mathrm{d\,m}}+\lambda_{\mathrm{a}}\Delta t_{\mathrm{d}}} \frac{1}{1+\beta\Delta l_{\mathrm{a}}} \frac{k_{0\,\mathrm{Au}}(\mathrm{m})}{k_{0\,\mathrm{Au}}(\mathrm{a})} \\
& \times \frac{G_{\mathrm{th\,m}} + \frac{G_{\mathrm{e\,m}}}{f}\left(\frac{Q_{0\,\mathrm{m}}-0.429}{\bar{E}^{\alpha}_{\mathrm{r\,m}}} + \frac{0.429}{0.55^{\alpha}(1+2\alpha)}\right)}{G_{\mathrm{th\,a}} + \frac{G_{\mathrm{e\,a}}}{f}\left(\frac{Q_{0\,\mathrm{a}}-0.429}{\bar{E}^{\alpha}_{\mathrm{r\,a}}} + \frac{0.429}{0.55^{\alpha}(1+2\alpha)}\right)} k_{\varepsilon\Delta E}\, k_{\varepsilon\Delta \mathrm{d_{ref}}} \\
& \times \frac{\left(\frac{\mathrm{d_{ref\,m}}-d'_{0\,\mathrm{m}}}{\mathrm{d_{ref\,m}}+\Delta d_{\mathrm{m}}-d'_{0\,\mathrm{m}}}\right)^2 \frac{1-e^{-\nu_{\mathrm{m}}h_{\mathrm{m}}\rho_{\mathrm{m}}}}{\nu_{\mathrm{m}}h_{\mathrm{m}}\rho_{\mathrm{m}}}}{\left(\frac{\mathrm{d_{ref\,a}}-d'_{0\,\mathrm{a}}}{\mathrm{d_{ref\,a}}+\Delta d_{\mathrm{a}}-d'_{0\,\mathrm{a}}}\right)^2 \frac{1-e^{-\nu_{\mathrm{a}}h_{\mathrm{a}}\rho_{\mathrm{a}}}}{\nu_{\mathrm{a}}h_{\mathrm{a}}\rho_{\mathrm{a}}}} \frac{\left(1+\frac{h_{\mathrm{a}}}{\mathrm{d_{ref\,a}}+\Delta d_{\mathrm{a}}-d'_{0\,\mathrm{a}}}\right)}{\left(1+\frac{h_{\mathrm{m}}}{\mathrm{d_{ref\,m}}+\Delta d_{\mathrm{m}}-d'_{0\,\mathrm{m}}}\right)} m_{\mathrm{std}}\left(1-\eta_{\mathrm{std}}\right) w_{\mathrm{m}} - m_{\mathrm{blank}} w_{\mathrm{blank}} \Bigg),
\end{aligned}
\tag{35}
$$

where subscripts a and m refer to the analyte and monitor, respectively, and the input quantities are briefly described as follow:

- $\lambda = \ln(2)/t_{1/2}$ is the decay constant of a radionuclide having an half-life time $t_{1/2}$,

- n_{p} is the number of counts in the full-energy γ-peak corrected for interferences and background (see eq. 36),

- COI is the true-coincidence correction factor,

- t_{c} and t_{l} are the counting and live times of the detection system,

- $t_{\mathrm{d\,m}}$ is the decay time of the monitor,

- $\Delta t_d = t_{\mathrm{d\,a}} - t_{\mathrm{d\,m}}$ is the difference between the analyte and monitor decay times at their acquisition start times,

- t_{i} is the neutron irradiation time,

- μ is the excess counting loss constant of the detection system,

- β is the linear flux variation within the irradiation channel depending on the vertical distance (see paragraph 3.3.6),

- Δl_{a} is the vertical distance of sample irradiation position with respect to the standard irradiation position,

- $k_{0\,\mathrm{Au}}$ is the k_0 factor,

- G_{th} and G_{e} are the thermal and epithermal neutron self-shielding correction factors,

- f is the (conventional) sub- to epi-cadmium neutron flux ratio (see paragraph 3.3.5),

- α is the epi-cadmium neutron shape factor (see paragraph 3.3.5),

- Q_0 is the ratio of the resonance integral (for a $1/E$ neutron spectrum in the epi-cadmium region) to the thermal cross section,

- $\bar{E}_{\mathrm{r}}$ is the effective resonance energy,

- $k_{\varepsilon\Delta E}$ is the monitor to analyte efficiency ratio calculated at reference position,

- $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ is the efficiency ratio of the analyte (or monitor) between actual and reference counting positions,

- d_{ref} is the nominal counting position (the one selected through the corresponding sliders in the main window, see paragraphs 3.7.5 and 3.8.5,

- $d_0^{'}$ is the distance travelled by the γ-emission within the detector,

- Δd is the vertical position variability with respect to the nominal position (d_{ref}),

- ν is the mass attenuation coefficient,

- h is the height of sample,

- ρ is the density of sample,

- w_{a}, w_{m} and w_{blank} are the mass fractions for analyte, monitor and blank, respectively,

- m_{std}, m_{sm} and m_{blank} are the standard, sample and blank masses, respectively, and

- η_{std} and η_{sm} are the relative moisture correction to apply to standard and sample masses, respectively.

In detail, for sample:

$$n_{\mathrm{p}} = n_{\mathrm{p}}^{'} - n_{\mathrm{b}} - n_{\mathrm{i}} - n_{\mathrm{f}}, \tag{36}$$

with, $n_{\mathrm{p}}^{'}$ the net area counts returned by the elaboration software (either GammaVision or HyperLab) and reported in the peaklist, n_{b} the number of counts due to background, n_{i} the number of counts due to γ interferences and n_{f} the number of counts due to fast activation interferences.

The net area background correction is automatically performed by the software if the background spectrum is selected. The net area for the interfering peak found in the background spectrum is reported to the live time of the investigated spectrum:

$$n_{\mathrm{b}} = n_{\mathrm{b}}^{'} \frac{t_{\mathrm{l\,smp}}}{t_{\mathrm{l\,bkg}}}, \tag{37}$$

where, $n_{\mathrm{b}}^{'}$ is the net area of interfering peak found in the background spectrum and subscripts smp

and bkg refer to sample and background spectrum, respectively.

The COI correction is performed based on P/T values calculated from the fit obtained in the characterization task and literature values gathered from De Corte tables [3]:

$$COI = \left(1 - \sum_i loss(i)\right)\left(1 + \sum_i sum(i)\right) \tag{38}$$

with $loss(i)$ and $sum(i)$ depending on the decay schemes of the investigated emitter, with combined uncertainty on COI calculated after the assumption $u_{\mathrm{r}}\left(loss(i)\right) = u_{\mathrm{r}}\left(sum(i)\right) = 20\,\%$.

The $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ parameter differs from 1 if sample and standard are acquired in different counting positions with respect to each other. Since one of the two counting positions needs to be the reference position, assigned during the characterization process (see paragraph 3.3.1), two scenarios might take place. In case the standard is counted at reference position:

$$\varepsilon = \frac{\varepsilon_{\mathrm{m\,ref}}}{\varepsilon_{\mathrm{a}}}, \tag{39}$$

with ε the efficiency ratio, $\varepsilon_{\mathrm{m\,ref}}$ the efficiency for monitor emission at reference position and ε_{a} the efficiency for analyte emission at counting position,

$$\varepsilon = \frac{\varepsilon_{\mathrm{m\,ref}}}{\varepsilon_{\mathrm{a}}}\frac{\varepsilon_{\mathrm{a\,ref}}}{\varepsilon_{\mathrm{a\,ref}}}, \tag{40}$$

since $\frac{\varepsilon_{\mathrm{m\,ref}}}{\varepsilon_{\mathrm{a\,ref}}} = k_{\varepsilon\Delta E}$:

$$\varepsilon = \frac{\varepsilon_{\mathrm{m\,ref}}}{\varepsilon_{\mathrm{a\,ref}}}\frac{\varepsilon_{\mathrm{a\,ref}}}{\varepsilon_{\mathrm{a}}} = k_{\varepsilon\Delta E}\, k_{\varepsilon\Delta d_{\mathrm{ref}}}, \tag{41}$$

thus, $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ is equivalent to the ratio between the analyte emission at reference and at the current counting position; since the reference is by definition the farthest counting position ($\varepsilon_{\mathrm{a\,ref}} < \varepsilon_{\mathrm{a}}$), $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ results in a value lower than 1.

In case the sample is counted at reference position:

$$\varepsilon = \frac{\varepsilon_{\mathrm{m}}}{\varepsilon_{\mathrm{a\,ref}}}, \tag{42}$$

$$\varepsilon = \frac{\varepsilon_{\mathrm{m}}}{\varepsilon_{\mathrm{a\,ref}}} \frac{\varepsilon_{\mathrm{m\,ref}}}{\varepsilon_{\mathrm{m\,ref}}}, \tag{43}$$

again, since $\frac{\varepsilon_{\mathrm{m\,ref}}}{\varepsilon_{\mathrm{a\,ref}}} = k_{\varepsilon\Delta E}$:

$$\varepsilon = \frac{\varepsilon_{\mathrm{m\,ref}}}{\varepsilon_{\mathrm{a\,ref}}} \frac{\varepsilon_{\mathrm{m}}}{\varepsilon_{\mathrm{m\,ref}}} = k_{\varepsilon\Delta E}\, k_{\varepsilon\Delta d_{\mathrm{ref}}}, \tag{44}$$

thus, $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ is equivalent to the ratio between the monitor emission at the current counting position and at reference; since the reference is by definition the farthest counting position ($\varepsilon_{\mathrm{a\,ref}} < \varepsilon_{\mathrm{a}}$), $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ results in a value greater than 1.

The $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ parameter automatically adjusts to the correct scenario based on the user selection in the main window (which radiobutton is checked in section 3.8.5 or 3.7.5) and is evaluated at any energy through the $k_{\varepsilon\Delta d_{\mathrm{ref}}}$ fits calculated during the characterization process (see eq. 21 in paragraph 3.3.1).

The ν value represents the mass attenuation coefficient which is a key parameter to evaluate the γ-self-absorption correction; it is calculated from literature data and composition of the corresponding sample.

$$\nu = \sum_i w_i (\mu/\rho)_i(E), \tag{45}$$

where, w_i is the mass fraction of the ith element of sample and $(\mu/\rho)_i(E)$ is the mass attenuation coefficient gathered from a NIST database [13] for that specific element at the investigated energy, E. Since the database is composed of data evaluated at defined energies, a linear approximation is performed to interpolate values. If the sample composition is unknown, ν value is set to 0 and the correction is not performed.

For simple activation-decay types the software also returns a value for the detection limit following the Currie's method [14]. The model is similar to eq. 35 in which $n_{\mathrm{p\,a}}$ is substituted with

$2.71 + 4.65\sqrt{B}$ where B is evaluated differently in case the peak is identified or not: for identified peaks background average intensity is evaluated based on a 3 channel range at both ends (1.5FWHM away) of the peak, otherwise the integral of spectrum over a 3FWHM range, centered on the centroid, is performed. Moreover, for the detection limit calculation blank correction is not applied.

5 Sources of input quantities

Source		X_i
Input spectra: GV[1)] / HL[2)]	x_i	$n_{\mathrm{p\,m}}$, $n_{\mathrm{p\,a}}$, $t_{\mathrm{c\,m}}$, $t_{\mathrm{c\,a}}$, $t_{\mathrm{l\,m}}$, $t_{\mathrm{l\,a}}$
	$u(x_i)$	$n_{\mathrm{p\,m}}$, $n_{\mathrm{p\,a}}$
Interface input fields	x_i	t_{i}, μ, $G_{\mathrm{th\,m}}$, $G_{\mathrm{th\,a}}$, $G_{\mathrm{e\,m}}$, m_{m}, m_{a}, w_{m}, Δl_{a}, Δd_{m}, Δd_{a}, h_{m}, h_{a}, ρ_{m}, ρ_{a}, η_{std}, η_{smp}
	$u(x_i)$	t_{i}, μ, $G_{\mathrm{th\,m}}$, $G_{\mathrm{th\,a}}$, $G_{\mathrm{e\,m}}$, m_{m}, m_{a}, w_{m}, Δl_{a}, Δd_{m}, Δd_{a}, h_{m}, h_{a}, ρ_{m}, ρ_{a}, η_{std}, η_{smp}
Software defaults[3)]	x_i	$G_{\mathrm{e\,a}}$
	$u(x_i)$	$G_{\mathrm{e\,a}}$, $t_{\mathrm{c\,m}}$, $t_{\mathrm{c\,a}}$, $t_{\mathrm{l\,m}}$, $t_{\mathrm{l\,a}}$, Δt_{d}, $t_{\mathrm{d\,m}}$
k_0-database	x_i	$k_{0\,\mathrm{Au}}(\mathrm{m})$, $k_{0\,\mathrm{Au}}(\mathrm{a})$, $\bar{E}_{\mathrm{r\,m}}$, $\bar{E}_{\mathrm{r\,a}}$, $Q_{0\,\mathrm{m}}$, $Q_{0\,\mathrm{a}}$, λ_{m}, λ_{a}
	$u(x_i)$	$k_{0\,\mathrm{Au}}(\mathrm{m})$, $k_{0\,\mathrm{Au}}(\mathrm{a})$, $\bar{E}_{\mathrm{r\,m}}$, $\bar{E}_{\mathrm{r\,a}}$, $Q_{0\,\mathrm{m}}$, $Q_{0\,\mathrm{a}}$, λ_{m}, λ_{a}
Computation results	x_i	COI_{m}, COI_{a}, β, f, α, $k_{\varepsilon\Delta E}$, $k_{\varepsilon\Delta d_{\mathrm{ref}}}$, $d'_{0\,\mathrm{m}}$, $d'_{0\,\mathrm{a}}$, ν_{m}, ν_{a}
	$u(x_i)$	COI_{m}, COI_{a}, β, f, α, $k_{\varepsilon\Delta E}$, $k_{\varepsilon\Delta d_{\mathrm{ref}}}$, $d'_{0\,\mathrm{m}}$, $d'_{0\,\mathrm{a}}$, ν_{m}, ν_{a}

Table 1: Input quantities, X_i, of the measurement model grouped depending on the input source of their values, x_i, and uncertainties, $u(x_i)$. [1)] ORTEC GammaVision, [2)] HyperLab, [3)] some of those default uncertainties can be modified in the settings window (see section 3.2.1)

6 Bibliography

References

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7 Versions

- Changes of version 2.1

  - implementation of tool for experimental setup prediction

  - correction of bug in the browse database leading to corruption of data involving flux and beta values

- Changes of version 2.0

  - implementation of the possibility to assign different counting positions to standard and sample spectra despite one has to be at reference, the overall detector characterization task is completely redesigned including a new method to deal with counting position variability correction

  - extended sample geometry effect and γ-self-absorption corrections automatically implemented based on literature data (NIST compilation) and sample information

  - COI correction automatically implemented based on the literature data from De Corte thesis

  - latest k0 database (k0_database_2020_8_24) adopted by default

  - implementation of only simple (I, IIB, IVB, VI) activation decay paths due to the rewriting of measurement model

  - inclusion of analysis validation section to compare results with reference certificate values

- Changes of version 1.2

  - introduction of an additional module in the Irradiation window to automatically evaluate f and α parameters with triple bare method (Au+Zr)

  - introduction of thermal flux correction factor (β) and vertical position variability in the irradiation channel in the measurement model in order to evaluate the effect of positioning in phase of irradiation

  - implementation of some complex activation-decay paths (IIA and more)

- Changes of version 1.1

  - installation procedure is simplified, download and installation of Anaconda package is no longer necessary nor recommended; presence of additional packages is checked during the first start-up of k0-INRIM software, however, a working internet connection is still required

  - calibration window is completely redesigned and calibration savefiles now contain more information; moreover, when a new calibration is saved the calibration drop-down menu is instantly updated, thus, the Refresh button is removed

- symbols and units not appearing correctly in the output file have been corrected

- options to reduce the number of peaks displayed in the peaklist by including a check on the maximum statistical uncertainty are added depending on the type of spectrum

- latest k0 database (k0_database_2019_04_04) adopted by default

o Changes of version 1.01

- equation 4 is corrected (an exponent was misplaced in the previous version due to a typing error); please always refer to equation 4 reported in the current user's manual version

- figure 25 is modified according to the corrected model