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The INAA-INRIM software version 3: presentation and validation of results

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Abstract

Automation in Instrumental Neutron Activation Analysis is present in almost every aspect of the measurement process, from the irradiation, to counting, to the elaboration of results. For the latter task a lot of dedicated software were developed over the years; among them, the k0-INRIM allowed to produce GUM-compliant uncertainty budgets. However, modeling and design limitations hampered its usability in routine analysis.
This paper presents the main features of the newly released INAA-INRIM software, which expands the functionalities of k0-INRIM (introduction of model for direct comparator, multiple counting positions, multiple spectra for same sample, managing of multiple measurements) while keeping the focus on the uncertainty from its predecessor. Notable updates are described and results of an analyzed reference material are reported as evidence supporting software validation.

Keywords: Neutron Activation Analysis, software, uncertainty, validation

1 Introduction

Automation is defined as any process having the goal to transfer tasks to machines working independently from human involvement. The concept ever-growing popularity has been fueled by the parallel improvement of technology at the point to be ubiquitous in present times due to the countless advantages it provides in multiple fields [1]. Within the scientific field, automation is beneficial in many aspects of (among others) metrology applied to analytical measurements, since it allows to minimize the analyst contribution while improving repeatability and reproducibility of the measurement procedure and result throughput using dedicated elaboration software.

Instrumental Neutron Activation Analysis (INAA) is a multi-element analytical technique in bulk based on nuclear reactions produced by neutrons [2, 3]; laboratories applying this technique have been adopting various strategies to automate the main processes of the measurement from designing pneumatic irradiation systems and sample changers for gamma acquisition [4] to the use of artificial intelligence for spectra elaboration [5].

Calculation of measurement results is another aspect of INAA where use of specific software is convenient, especially when the k_0-standardization method [6] is being used since a large amount of elements could be potentially quantified at once. A variety of commercial software is available, although it is not a rare occurrence that each laboratory develops its own tailor-made code [7] in order to satisfy their needs and suitably adapt to their experimental setup.

The need for an automatic support to evaluate measurement uncertainty as described by the Guide to Uncertainty in Measurement (GUM) [8] applied to INAA analysis is what propelled the development of a homemade code at the INRIM radiochemistry and spectroscopy laboratory (now elemental analysis laboratory) culminated with the release of the k0-INRIM software: a tool able to compile uncertainty

budgets for k_0-INAA measurements based on the spreadsheet method [9]. The k0-INRIM software displayed a detailed measurement model inspired from the original k_0 expression but including a custom efficiency ratio function obtained from experimental data and suitable for considering correlations among fitting parameters; in addition, it seamlessly fit into the previously established experimental setup by taking as input the files returned from the spectra elaboration program and yielding as output the GUM-compliant uncertainty budget in Microsoft Excel file format. While the reliability of the implementation was corroborated by the successful participation to a software inter-comparison [10], the routine use in INAA analysis highlighted some technical inconvenience and practical difficulty, namely: all spectra for samples (or standards) needed to be acquired at the same counting position, uncertainty of correlated results (when multiple spectra are acquired for the same sample) were not properly managed, a few corrections (fission and gamma interferences, buoyancy) were missing. In order to overcome these limitations and also expand its applicability to measurements performed with the direct comparator method, a complete revamp of the software, involving measurement models, data structure and user interface, was developed; the name was also changed to INAA-INRIM to better underline the use no longer restricted to the k_0-standardization method following the update.

In this work, the latest release of INAA-INRIM software (version 3.2 at the time of writing) is presented describing the most notable updates. Results of a comparison of software elaborated mass fractions with corresponding consensus values of a certified reference material, meant as software validation [11], are also reported.

2 Method

In the following subsections, the updated measurement model introduced in the software is discussed, other main implemented features concerning interface and usability

are presented, finally, the experimental setup adopted for validation of the code is described.

2.1 Measurement model

The INAA-INRIM was developed with the aim to unify the adopted approach between the two most used standardizations (direct comparator and k_0) in order to get compatible uncertainty budget output allowing seamlessly transition from one another on the same software. This goal was primarily achieved by designing an equation model where the building blocks are macro-parameters (independent units composed by inputs related to common aspects of the measurement). The recently developed comprehensive measurement equation for the relative method [12] was taken as reference, thus, a model comprising of 8 macro-parameters was obtained:

$$w_{\mathrm{smp}} = R_n \; R_d \; R_{k_0} \; R_\phi \; R_\varepsilon \; R_m - K_{\mathrm{blk}} - K_{\mathrm{U}}, \tag{1}$$

where w_{smp} is the mass fraction of the element of interest in the investigated sample (hereafter called measurement sample while the material used to ensure traceability is referred to as standard sample), $R_n = n_{\mathrm{p\ smp}} \left(n_{\mathrm{p\ std}}\right)^{-1}$ is defined as net area ratio and includes input related to counting statistics (magnitude of peak area and direct gamma interferences), $R_d = \left(t_{\mathrm{c\ smp}}\; t_{\mathrm{l\ std}} \left(1 - e^{-\lambda\; t_{\mathrm{c\ std}}}\right)\right) \left(t_{\mathrm{c\ std}}\; t_{\mathrm{l\ smp}} \left(1 - e^{-\lambda\; t_{\mathrm{c\ smp}}}\right)\right)^{-1} e^{\mu\left(\frac{t_{\mathrm{l\ std}}}{t_{\mathrm{c\ std}}} - \frac{t_{\mathrm{l\ smp}}}{t_{\mathrm{c\ smp}}}\right)}\; e^{\lambda\; \Delta t_d}$ is defined as decay ratio and includes input related to the corrections accounting for irradiation, decay and counting times, and detector pileup rejection system, $R_{k_0} = \theta_{i_{E\ \mathrm{std}}}\; M_{\mathrm{smp}} \left(\theta_{i_{E\ \mathrm{smp}}}\; M_{\mathrm{std}}\right)^{-1}$ is defined as k_0 ratio and includes input related to materials' composition, $R_\phi = \frac{1}{1+\beta\; \Delta l} \left(G_{\mathrm{th\ std}} + \frac{G_{\mathrm{e\ std}}}{f} Q_{0\ \mathrm{std}}\left(\alpha\right)\right) \left(G_{\mathrm{th\ smp}} + \frac{G_{\mathrm{e\ smp}}}{f} Q_{0\ \mathrm{smp}}\left(\alpha\right)\right)^{-1}$ is defined as neutron flux ratio and includes input related to the neutron irradiation (flux parameters and neutron self-shielding effects), $R_\varepsilon = \frac{COI_{\mathrm{std}}}{COI_{\mathrm{smp}}}\; k_{\varepsilon\Delta E}\; k_{\varepsilon\Delta d}\; k_{\mathrm{pos}}\; k_{\mathrm{geo}}\; k_{\mathrm{sa}}$ is

defined as efficiency ratio and includes input related to detection efficiency taking into account emissions, samples positioning, samples geometry, self-absorption effects and true-coincidence correction, $R_m = \frac{m_{\mathrm{std}}\ k_{\mathrm{buy}}\ (1-\eta_{\mathrm{std}})\ w_{\mathrm{std}}}{m_{\mathrm{smp}}\ (1-\eta_{\mathrm{smp}})}$ is defined as mass ratio and includes input related to weighting of samples and moisture correction, $K_{\mathrm{blk}} = \frac{m_{\mathrm{blk}}\ w_{\mathrm{blk}}}{m_{\mathrm{smp}}(1-\eta_{\mathrm{smp}})}$ is defined as blank correction and includes input related to the composition of the blank material, $K_{\mathrm{U}} = \Delta w_{\mathrm{U}}$ is defined as U fission correction and includes input related to literature values and U mass fraction in the sample. Inner quantities to the macro-parameters, which symbols are not described here, are discussed in literature [9, 12, 13].

Similar to its original formulation, (1) is suitable for samples co-irradiated in nuclear reactors for which the exposure time is short enough to make burnup effects negligible and only if the elements of interest activate through direct activation-decay schemes [6]. The main convenience brought by the current formulation is that macro-parameters are assembled in order to be uncorrelated to each other thus simplifying the uncertainty propagation; although this claim is not strictly true in every case: for example neutron self-shielding parameters in R_ϕ and γ-self-absorption parameters in R_ε both rely on the composition of the samples, K_{blk} shares a few input parameters (mass of sample and moisture correction) with R_m and K_{U} depends on some neutron flux parameter and literature datum shared with R_ϕ. However, since all these examples described here only affect minor corrections, the extent of the correlation is usually quite limited and the inter-independency of macro-parameters is a safe assumption in most use cases.

Another advantage of the current formulation relies in its flexibility; in fact, (1) allows to be easily extended to be suitable to the application of the k_0 measurement model. This is achieved by simply adjusting the definition of R_{k_0} while keeping all the other macro-parameters of (1) intact:

$$R_{k_0} = \frac{k_{0\ \mathrm{Au}}(\mathrm{m})}{k_{0\ \mathrm{Au}}(\mathrm{a})}, \tag{2}$$

$$R_d = \frac{\lambda_{\mathrm{smp\ (a)}}\ t_{\mathrm{c\ smp}}\ t_{\mathrm{l\ std}} \left(1 - e^{-\lambda_{\mathrm{std\ (m)}}\ t_{\mathrm{irr}}}\right) \left(1 - e^{-\lambda_{\mathrm{std\ (m)}}\ t_{\mathrm{c\ std}}}\right)}{\lambda_{\mathrm{std\ (m)}}\ t_{\mathrm{c\ std}}\ t_{\mathrm{l\ smp}} \left(1 - e^{-\lambda_{\mathrm{smp\ (a)}}\ t_{\mathrm{irr}}}\right) \left(1 - e^{-\lambda_{\mathrm{smp\ (a)}}\ t_{\mathrm{c\ smp}}}\right)} \\ e^{\mu\left(\frac{t_{\mathrm{l\ std}}}{t_{\mathrm{c\ std}}} - \frac{t_{\mathrm{l\ smp}}}{t_{\mathrm{c\ smp}}}\right)}\ e^{\left(\lambda_{\mathrm{smp\ (a)}} - \lambda_{\mathrm{std\ (m)}}\right) t_{\mathrm{d\ std}} + \lambda_{\mathrm{smp\ (a)}}\ \Delta t_d}, \tag{3}$$

where the ratio of composite nuclear constants k_0 replaces the ratio of isotopic abundances and molar masses.

The measurement model adopted for the INAA-INRIM software was based on (1) with the macro-parameter R_{k_0} definition changing depending on the adopted standardization and R_d definition always set to (3) by default with $\lambda_{\mathrm{smp}\ (a)} = \lambda_{\mathrm{std}\ (m)}$ in case of direct comparator standardization. This formulation allowed for a better implementation of the model in the code, improving the flexibility of the analysis and making easier dealing with propagation of uncertainties, especially for multiple measurements of the same measurand, as well as visualization of uncertainty budgets.

2.2 Software features

The following paragraphs represent only a brief summary of the notable features that were added to INAA-INRIM software up to the latest release; a vademecum, including a more in depth description of the functionalities and a step-by-step guide on how to perform basic analysis, can be found in the software official repository [14].

The main feature of INAA-INRIM software is to bring together both direct comparator and k_0-standardization approaches into a single interface and providing full GUM compliant uncertainty budgets for the corresponding measurement results. In order to achieve this, multiple novelties and improvements were necessary from a metrological and a practical standpoint.

From a metrological perspective, the updated measurement model allows to easily combine measurements obtained by different spectra/emissions acquired from the same measurement sample and by different measurement samples from the same material to get an averaged value. The result output is thus organized in three levels: (i) at the base level, a single uncertainty budget related to a measurement sample based on a single spectrum-emission pair, and compiled using (1), is reported, (ii) at the second level, budgets related to the same measurement sample are grouped together and a weighted average is performed to get a single value to be assigned to the sample, (iii) the third and last level considers all calculated averaged values and an arithmetic mean is performed to obtain the overall result to be assigned to the material. For averages performed in levels (ii) and (iii) the uncertainty is propagated by summing the averaged contributions to combined variance of each macro-parameter (with the exception of R_n, whose contribution to variance is reduced depending on statistical uncertainty and number of measurements). The output files, created in Microsoft Excel format, provide information concerning the measurement of an element in a material reporting the measured value and combined uncertainty, as well as expanded uncertainty with coverage factor and effective degrees of freedom, the equation model applied and the percent contribution of each macro-parameter to the combined variance; they also include worksheets for every single budgets, summaries for samples and the total summary, so that all information is traced back to input parameters (figure 1).

From a practical and technical standpoint, features were introduced to enhance usability of the software and enlarge analyst control over the analysis. In detail, the workflow of the analysis (where analysis is still defined as measurement performed from irradiation in a single facility and spectra acquisition on a single detector) has been slightly modified, compared to the previous versions, and the graphical interface was accordingly tweaked to reflect the changes (figure 2).

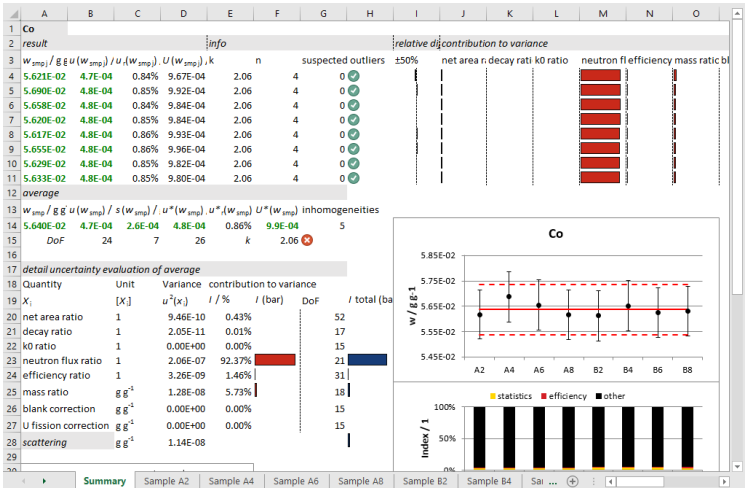


Fig. 1 The uncertainty budget output in Microsoft Excel format

In particular, a more schematic placement of sections (each section is delimited by the black rectangular boxes displaying a title in the upper left corner) is found in the main window which reflects the optimized workflow from left-to-right and from top-to-bottom. Additionally, all information concerning samples and acquired spectra are now stored in their corresponding sections meaning information for measurement samples and standard samples are both found in the *Sample management*, likewise, information for all the spectra, regardless of their origin, are found in the *Measurement spectra* section. Currently, each spectrum requires to be manually linked to its corresponding sample in order to establish a connection determining whether it will be considered a measurement sample spectrum or a standard sample spectrum for the analysis. Thus, with respect to the previous versions, the current workflow allows for an easier management of samples (either measurement samples and standard samples) for which multiple spectra are acquired, moreover, counting positions can now be set individually for each spectrum improving the flexibility of the elaboration. Peaks identification algorithm is also updated allowing the user to check the suspected identity of an emission against the presence of all other emissions from the same emitter found in the investigated spectrum, aiding the analyst by promoting a more robust decisional

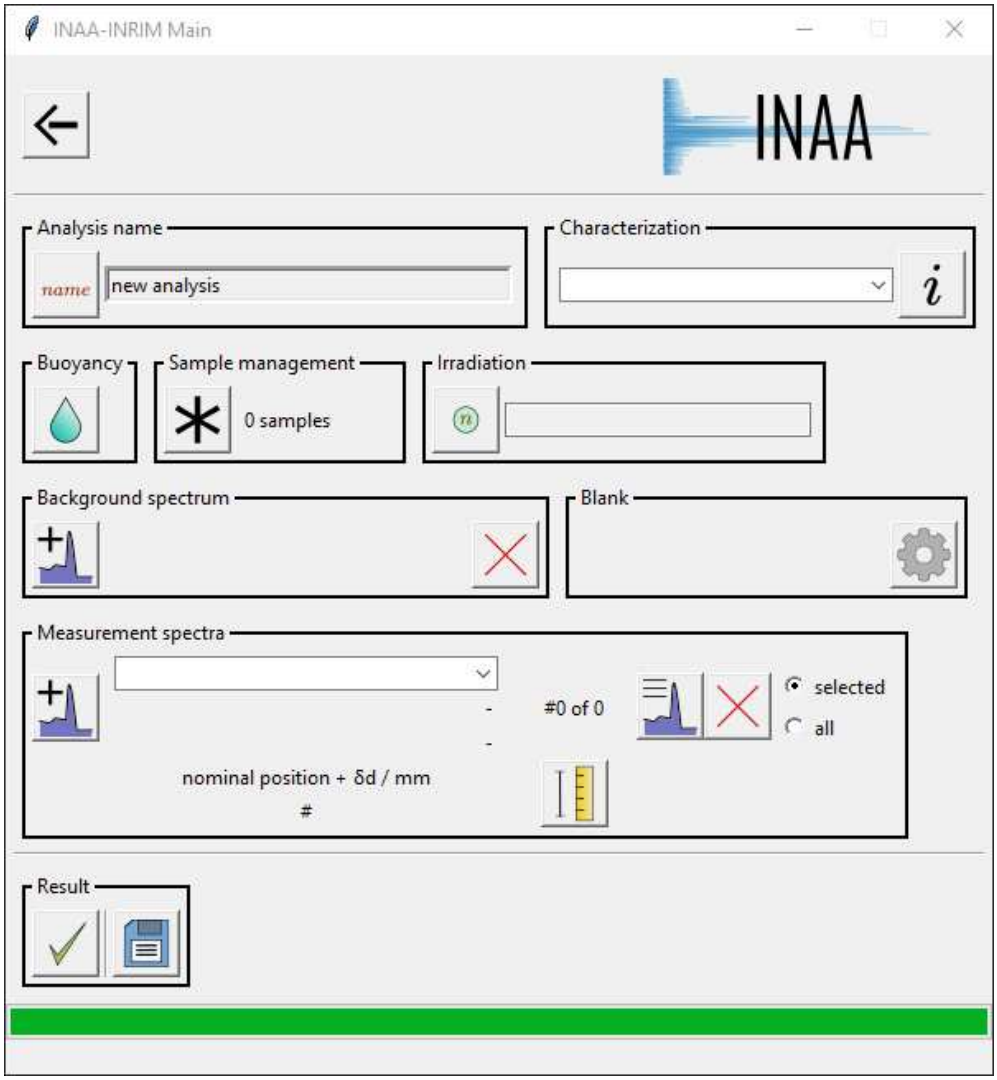


Fig. 2 The main window of the INAA-INRIM software version 3.2

process and giving the opportunity to conveniently assign the detected emitter to all their emissions at once. Additionally, the creation of emissions presets is now possible allowing to instantly search and confirm a predefined set of emissions. Selection and visualization of results is another of the revamped aspects: saved results from different analysis can be recalled to be merged together making possible to quantify measurands

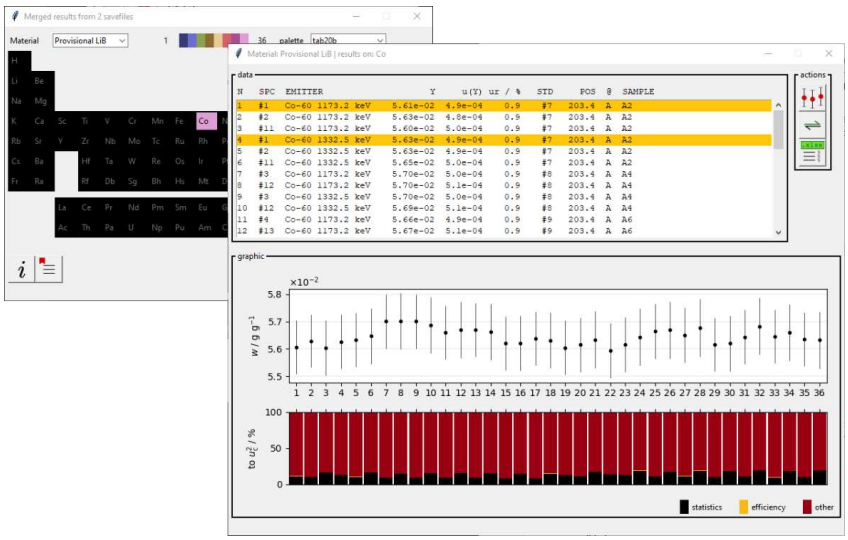


Fig. 3 The results window of the INAA-INRIM software version 3.2

activated from multiple irradiations (as it is common in INAA to perform a short and a long irradiation to detect a wide range of radionuclides) or counted on different detectors. All obtained results are visualized on the classic periodic table style window. However, subwindows can now be accessed to display information concerning each measured element (such as single measured values, absolute and relative uncertainties, percent contributions to combined variance, origin of datum) and a selection of the data to be included in the exported Microsoft Excel output is allowed (figure 3).

Further minor changes involve: a whole section called *Buoyancy* introduced in the main window (figure 1) in order to collect the input parameters required to calculate k_{buy} appearing in the updated model equation for R_m; a switch allowing to select between the standardization methods for data analysis: relative, k_0, or both; and a few other options affecting the general behavior of the software for what concerns peak-identification, default input parameters and output representation.

Despite the soundness achieved by the software in its 3.2 version, INAA-INRIM development is still ongoing, thus, bug correction and addition of even more features could be expected in the future.

2.3 Validation experiment

In order to validate the proper functioning of the software and its implemented measurement model for both experimental methods (direct and k_0-standardization), INAA-INRIM was adopted as the main elaboration tool to provide results for the analysis of certified reference material LNE-LiB01, produced by the Laboratoire National de metrologie et d'Essais (LNE) with the goal to quantify critical elements in Li-ion battery black mass.

The bottle of reference material contained 20 g of crushed Li-ion dry pouch cells with final grain size below 200 µm. After manual shaking of the bottle, 8 aliquots of around 170 mg mass each were weighted on analytical balance and pressed to obtain cylindrical tablets of 10.00(5) mm diameter and 1.05(5) mm height which dimensions were measured with an analogic caliper. Here and hereafter, numbers in parenthesis indicate the standard uncertainty and refer to the last digits of the corresponding value. Additional 4 aliquots of around 1 g mass were weighed and used to evaluate moisture content by means of repeated cycles of 2 h at 105 °C in a ventilated oven until stable mass was reached.

A total of 10 standard samples were prepared by pipetting aliquots of around 60 µL each from a SI traceable solution, $9.988(19) \times 10^{-3}\,\mathrm{g\,g^{-1}}$ Co mass fraction, onto absorbent paper discs, air dried and sealed with adhesive tape to prevent losses. The 8 measurement samples (inserted into polyethylene vials) and 10 sealed standard samples were piled into two irradiation containers in a way that each measurement sample was surrounded by two standard samples in order to keep possible neutron flux gradients under control.

The containers were placed in two irradiation channels situated in the Lazy Susan facility of the TRIGA Mark II reactor in Pavia and irradiated for 30.0(3) min at 250 kW power. After the irradiation, measurement and standard samples were transferred

to counting containers and counted on two HyperPure germanium (HPGe) detectors: ORTEC GEM50P4-83 (50 % relative efficiency, 1.9 keV full-width half maximum at 1332.5 keV energy) and ORTEC GEM20P4-70 (20 % relative efficiency, 1.7 keV full-width half maximum at 1332.5 keV energy) both connected to ORTEC DSPEC multichannel analyzers and controlled by personal computers. Counting distances were set to assure a similar count rate on both detectors despite the difference in efficiency, thus, measurement and standard samples were placed at 203.6(2) mm from the end-cap on the 50 % detector and at 143.6(2) mm from the end-cap on the 20 % detector. Two spectra were collected for each measurement sample having cooling times, relative to end of irradiation, ranging from 46.8 h to 55 h for the first spectrum and from 336 h to 366 h for the second spectrum and acquisition times ranging from 2.5 h to 3.5 h real time for the first one and 6 h real time for the second one, respectively; a single spectrum was collected for each standard sample having cooling times ranging from 168 h to 264 h with real time of 24 h.

Collected spectra were elaborated with HyperLab software [15] adopting the "default settings for moderate count rate, regular peaks" algorithm and relying on energy and resolution curves previously evaluated for both detectors. Spectrum profile and list of information concerning the fitted peaks (hereafter called peaklist) were exported as ascii and csv file formats, respectively, to be digestible as input for the INAA-INRIM software.

Experimental data concerning the analysis (samples, irradiation) were manually introduced to INAA-INRIM through the main window interface (figure 2) and corresponding sub-windows. The elaborated spectra were recalled from their saved ascii (to visualize the profile) and csv (to get vital information about the peaklist) files. Emissions from ^{60}Co (1173.2 keV and 1332.5 keV) and ^{64}Cu (1345.8 keV) were identified and selected for the corresponding lines in the peaklists for measurement and standard sample spectra in the case of Co, and only for measurement sample spectra for the Cu.

Since the current version of the software still doesn't allow to process spectra acquired on multiple detectors within the same analysis, two elaborations were performed separately to overcome this limitation: one concerning spectra acquired with the GEM50P4-83 detector, the other elaboration concerning spectra acquired with the GEM20P4-70 detector; using the INAA-INRIM software the two analysis files were then recalled and merged as one in the results window (figure 3) where all the computed data are summarized and selected for the creation of the budget output. The "relative + k0" method choice, together with the line "Co-60 1332.5 keV" for the k_0 monitor drop down menu were selected to elaborate results for Co and Cu. This combination of options, paired with the setting "elaborate selection only" turned on (found in the settings window) made sure that each emission for Co from the sample spectrum was elaborated with the corresponding emission in the standard spectrum applying the direct comparator method. On the other hand, emissions for Cu were elaborated with the k_0 method adopting the selected ^{60}Co 1332.5 keV emission as k_0 monitor.

In order to correctly assess self-shielding effects due to main matrix components, an estimated composition was assigned to the measurement sample material based on analysis of acquired spectra from a preliminary irradiation performed with k_0 method using Au monitor. Additionally, since Co and Cu were expected to be at percent level, iterative calculation of the results was enabled in the software settings allowing to better estimate self-shielding (especially epithermal) and self-absorption effects mainly due to the abundant presence of these elements.

3 Results

The final Co and Cu mass fraction values resulted from the average of 8 values (once for every measurement sample). In case of Co, each one was calculated as the weighted average of 4 data (1173.2 keV and 1332.5 keV emissions from both acquired spectra)

Table 1 Resulting Co mass fraction, $w_{\mathrm{Co}\ i}$, and uncertainty, $u\,(w_{\mathrm{Co}\ i})$, for any sample of the LNE-LiB01 material, i, and the average; n indicates the number of data from which the value is calculated and k is the coverage factor to get the 95 % interval expanded uncertainty. Relative contributions to combined variance due to R_n, R_ϕ, all other parameters in (1) and scattering of data, s, are reported.

i	$w_{\mathrm{Co}\ i}$ / g g^{-1}	$u\,(w_{\mathrm{Co}\ i})$ / g g^{-1}	n	R_n	R_ϕ	other	s
A2	5.621×10^{-2}	4.7×10^{-4}	4	3 %	90 %	7 %	-
A4	5.690×10^{-2}	4.8×10^{-4}	4	3 %	90 %	7 %	-
A6	5.658×10^{-2}	4.8×10^{-4}	4	3 %	90 %	7 %	-
A8	5.620×10^{-2}	4.8×10^{-4}	4	3 %	91 %	7 %	-
B2	5.617×10^{-2}	4.8×10^{-4}	4	4 %	89 %	7 %	-
B4	5.655×10^{-2}	4.8×10^{-4}	4	4 %	89 %	7 %	-
B6	5.629×10^{-2}	4.8×10^{-4}	4	4 %	89 %	7 %	-
B8	5.633×10^{-2}	4.8×10^{-4}	4	3 %	89 %	7 %	-
	w_{Co} / g g^{-1}	$u\,(w_{\mathrm{Co}})$ / g g^{-1}	k	R_n	R_ϕ	other	s
average	5.640×10^{-2}	4.8×10^{-4}	2.06	0 %	88 %	7 %	5 %

Table 2 Resulting Cu mass fraction, $w_{\mathrm{Cu}\ i}$, and uncertainty, $u\,(w_{\mathrm{Cu}\ i})$, for any sample of the LNE-LiB01 material, i, and the average; n indicates the number of data from which the value is calculated and k is the coverage factor to get the 95 % interval expanded uncertainty. Contributions to combined variance due to R_n, R_ϕ, all other parameters in (1) and scattering of data, s, are reported.

i	$w_{\mathrm{Cu}\ i}$ / g g^{-1}	$u\,(w_{\mathrm{Cu}\ i})$ / g g^{-1}	n	R_n	R_ϕ	other	s
A2	1.041×10^{-1}	2.0×10^{-3}	1	8 %	60 %	32 %	-
A4	1.060×10^{-1}	2.0×10^{-3}	1	8 %	61 %	31 %	-
A6	1.041×10^{-1}	2.0×10^{-3}	1	8 %	61 %	31 %	-
A8	1.031×10^{-1}	2.0×10^{-3}	1	8 %	62 %	30 %	-
B2	1.002×10^{-1}	1.9×10^{-3}	1	10 %	59 %	31 %	-
B4	1.030×10^{-1}	2.0×10^{-3}	1	10 %	59 %	31 %	-
B6	1.028×10^{-1}	2.0×10^{-3}	1	11 %	58 %	31 %	-
B8	1.050×10^{-1}	2.0×10^{-3}	1	11 %	58 %	32 %	-
	w_{Cu} / g g^{-1}	$u\,(w_{\mathrm{Cu}})$ / g g^{-1}	k	R_n	R_ϕ	other	s
average	1.035×10^{-1}	1.9×10^{-3}	2.00	1 %	57 %	30 %	13 %

while for Cu each one corresponded to a single datum (1345.8 keV emission from one spectrum collected in the short acquisition). A summary of the obtained results in the output files is reported in table 1 and table 2.

Obtained results were compared with the certified values for material LNE-LiB01 and reported in figure 4.

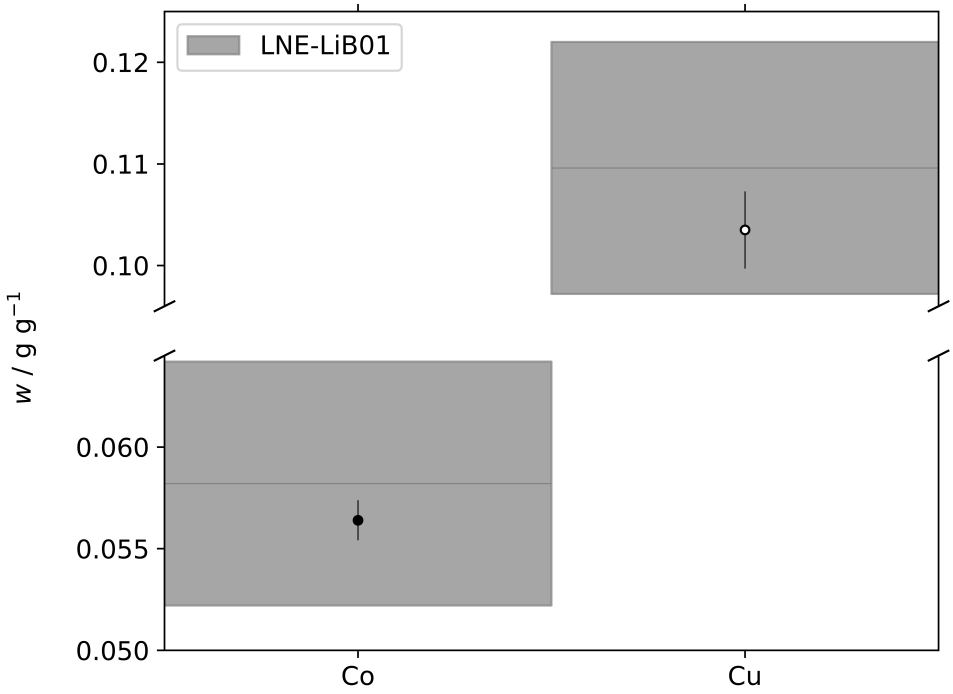


Fig. 4 Results for Co and Cu (black and white dot, respectively) calculated with INAA-INRIM software and compared with the certified values of reference material LNE-LiB01 (horizontal solid line). Vertical error bars and grey areas represent the corresponding expanded uncertainties.

Comparison of results with the reference mass fractions of LNE-LiB01 confirms that INAA-INRIM calculated values are in agreement using both standardizations (direct comparator for Co and k_0 for Cu). According to the information reported in the uncertianty budgets produced by the software itself, and summarized in tables 1 and 2, the main contributor to the combined uncertainty of Co and Cu values was, in both cases, the R_Φ macro-parameter. Specifically, for Co, the epithermal self-shielding correction for the measurement sample, $G_{\mathrm{e\,smp}}$, resulted the main contributor of the combined uncertainty while for Cu, the two main contributions were the Cu resonance integral to thermal cross section ratio, $Q_{0\,\mathrm{Cu}}$, and $G_{\mathrm{e\,smp}}$, as highlighted in tables 3 and 4.

The uncertainties provided by INAA-INRIM are compatible with the perfomed measurement considering that counting statistics was minimized due to high count rate and large number of samples, thus mainly the standardization of choice and experimental setup cointributed to them. As expected, relative uncertianty of Co value (0.9 %) is considerably lower than Cu value (1.8 %) since with direct comparator method many of the activation and detection parameters are neglected and dependency upon composite nuclear constants is minimal.

Moreover, since LNE-LiB01 is considered a challenging material to measure due to very high content of metal elements in the matrix, its use in the validation experiment provided the opportunity to evaluate the proper behavior for most of the corrections implemented in the software model (self-shielding, self-absorption) that would otherwise be negligible in the majority of experimental setup and matrices.

4 Conclusions

INAA-INRIM software expanded the applicability of the previous version of the code adopted at the INRIM elemental analysis laboratory by providing useful features aiming for a better integration with the experimental setup in order to improve flexibility of analysis while reaching the best measurement accuracy. Validation of the overall model (in both direct and k_0-standardization INAA modes) and its implementation is confirmed by the successful results obtained in the analysis of LNE-LiB01 reference material.

Stable 3.2 version of INAA-INRIM software for Windows systems is freely available on the zenodo repository [16], while the most up-to-date (open) source code is available on a github repository: the latter represents the currently development branch thus it provides the most up-to-date features (not discussed here) but is more likely to present bugs and instabilities, also, a python interpreter is needed to run this particular code.

Table 3 Summary of main contributors to the combined uncertainty within the three-stepped uncertainty budget for Co mass fraction. For each step (from left to right: total average, average of results from each measurement sample and single measurement) main contributors with their corresponding relative contributions are reported. The values in parenthesis refer to the sample ID and the spectrum ID in the columns sample average and measurement, respectively. The ×2 symbol after the parenthesis in the parameter column serves to remind that 2 measurements were obtained from each acquired spectrum.

total average		sample average (i)		measurement (j)	
parameter	contribution to u^2 (w_{Co})	parameter	contribution to u^2 ($w_{\mathrm{Co}\ i}$)	parameter	contribution to u^2 ($w_{\mathrm{Co}\ j}$)
R_ϕ	87.9 %	R_ϕ(A2)	89.9 %	$G_{\mathrm{e\ smp}}$(1 & 9)×2	73.9 %
		R_ϕ(A4)	90.4 %	$G_{\mathrm{e\ smp}}$(2 & 10)×2	75.0 %
		R_ϕ(A6)	90.4 %	$G_{\mathrm{e\ smp}}$(3 & 11)×2	75.1 %
		R_ϕ(A8)	90.8 %	$G_{\mathrm{e\ smp}}$(4 & 12)×2	75.5 %
		R_ϕ(B2)	88.9 %	$G_{\mathrm{e\ smp}}$(5 & 13)×2	73.4 %
		R_ϕ(B4)	89.1 %	$G_{\mathrm{e\ smp}}$(6 & 14)×2	74.0 %
		R_ϕ(B6)	89.0 %	$G_{\mathrm{e\ smp}}$(7 & 15)×2	73.8 %
		R_ϕ(B8)	89.2 %	$G_{\mathrm{e\ smp}}$(8 & 16)×2	73.6 %
R_m	5.45 %	not further expanded to inner budgets			
s	4.87 %	not further expanded to inner budgets			
R_ε	1.39 %	not further expanded to inner budgets			
R_n	0.40 %	not further expanded to inner budgets			
R_d	0.01 %	not further expanded to inner budgets			
R_{k_0}	0.00 %	not further expanded to inner budgets			

Interested users are encouraged to try using the software for their analysis and helping the development by providing feedback for future improvements.

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Declarations

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Table 4 Summary of main contributors to the combined uncertainty within the three-stepped uncertainty budget for Cu mass fraction. For each step (from left to right: total average, average of results from each measurement sample and single measurement) main contributors with their corresponding relative contributions are reported. The values in parenthesis refer to the sample ID and the spectrum ID in the columns sample average and measurement, respectively.

total average		sample average (i)		measurement (j)	
parameter	contribution to $u^2\,(w_{\mathrm{Cu}})$	parameter	contribution to $u^2\,(w_{\mathrm{Cu}\ i})$	parameter	contribution to $u^2\,(w_{\mathrm{Cu}\ j})$
R_ϕ	56.6 %	R_ϕ(A2)	60.1 %	$G_{\mathrm{e\ smp}}(1)$ $Q_{0\ \mathrm{Cu}}(1)$	22.8 % 31.5 %
		R_ϕ(A4)	61.4 %	$G_{\mathrm{e\ smp}}(2)$ $Q_{0\ \mathrm{Cu}}(2)$	25.4 % 30.4 %
		R_ϕ(A6)	60.9 %	$G_{\mathrm{e\ smp}}(3)$ $Q_{0\ \mathrm{Cu}}(3)$	25.0 % 30.4 %
		R_ϕ(A8)	61.6 %	$G_{\mathrm{e\ smp}}(4)$ $Q_{0\ \mathrm{Cu}}(4)$	26.2 % 29.8 %
		R_ϕ(B2)	58.8 %	$G_{\mathrm{e\ smp}}(5)$ $Q_{0\ \mathrm{Cu}}(5)$	23.4 % 29.9 %
		R_ϕ(B4)	59.0 %	$G_{\mathrm{e\ smp}}(6)$ $Q_{0\ \mathrm{Cu}}(6)$	23.4 % 30.1 %
		R_ϕ(B6)	57.9 %	$G_{\mathrm{e\ smp}}(7)$ $Q_{0\ \mathrm{Cu}}(7)$	22.0 % 30.4 %
		R_ϕ(B8)	57.8 %	$G_{\mathrm{e\ smp}}(8)$ $Q_{0\ \mathrm{Cu}}(8)$	21.6 % 30.6 %
R_{k_0}	27.7 %	not further expanded to inner budgets			
s	12.7 %	not further expanded to inner budgets			
R_n	1.09 %	not further expanded to inner budgets			
R_m	1.05 %	not further expanded to inner budgets			
R_ε	0.50 %	not further expanded to inner budgets			
R_d	0.25 %	not further expanded to inner budgets			

Competing interests

The authors declare no competing interests.

Author contributions

M Di Luzio: Software, Validation, Writing - original draft; G D'Agostino: Conceptualization, Methodology, Writing - review and editing

Data availability

Original data used for elaboration and result files obtained from the sofware can be found in the zenodo repository: https://doi.org/10.5281/zenodo.19470487.

Code availability

Open source code can be found in the github repository: https://github.com/marcodiluzio/INAA-INRIM.

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