

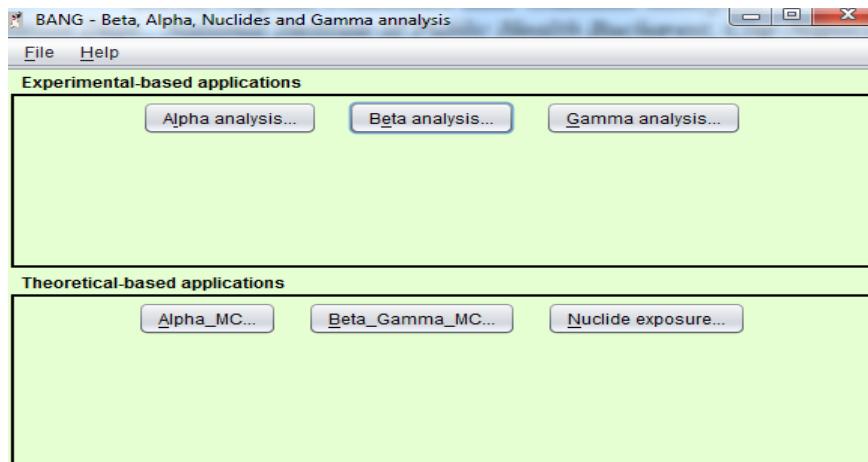
# BANG

Beta, Alpha, Nuclides and Gamma analysis

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The BANG software is intended to help physicists working in alpha and beta radiation detection field, internal and external dosimetry field as well as gamma spectroscopy. It is written entirely in Java and therefore is platform independent.

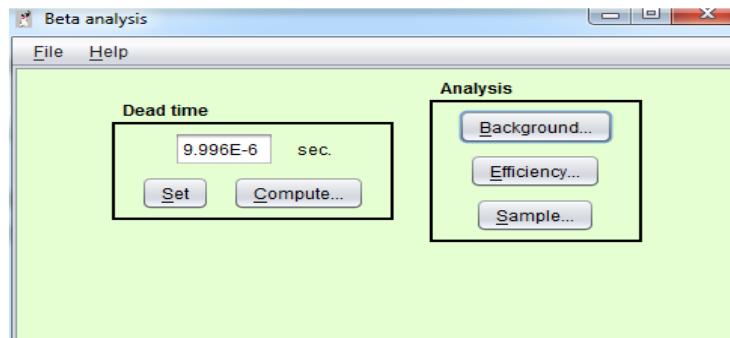
Its main features can be organized in two categories: experimental-based applications and theoretical-based applications. In some cases, a good detector calibration can not be achieved using experimental methods (such as measurement of a standard radiation source) and therefore a theoretical approach must be used to assess the calibration factor (i.e. detector efficiency). This is done using the Monte-Carlo simulation technique for radiation transport in the detector active volume.



## 1. Experimental based applications

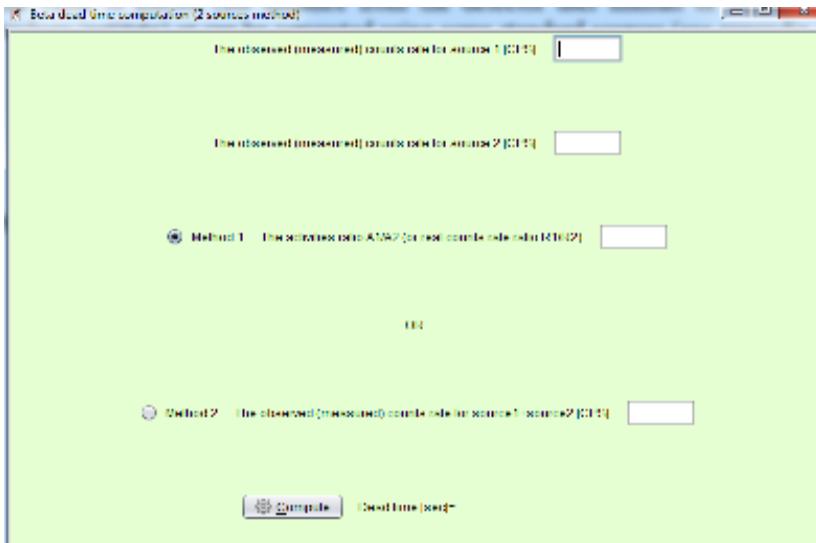
### 1.1 The alpha/beta analysis

The same routine is used both for alpha and beta analysis, the sole difference being the usage of different databases.



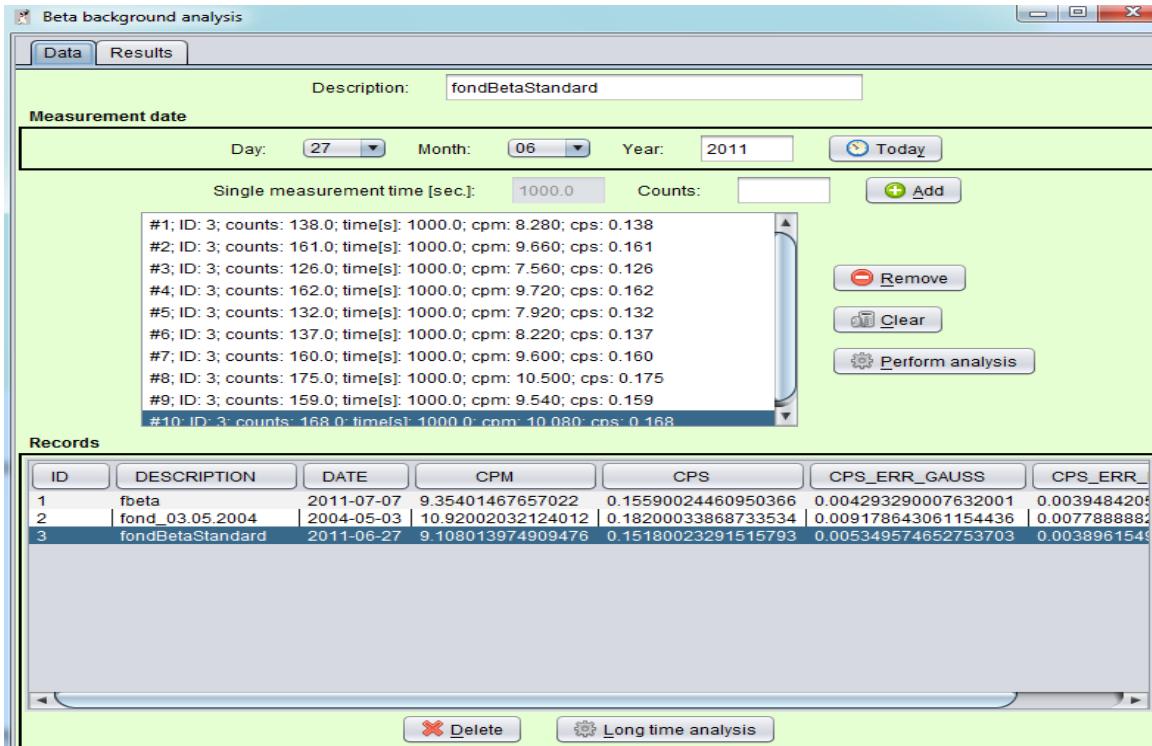
First, one must enter the detector dead-time, which is the short amount of time the detector does not count any pulses. The detector dead time can be manually set if the

value is known (for instance from the detector user manual or supplied by a metrology institute) or can be computed using some standard sources (see Appendix 1).



In order to compute the actual sample activity, the ambient background (counts per seconds for a blank measurement) as well as detector efficiency must be known.

### 1.1.1 The background



When the background window is opened, the interrogation fields are automatically populated based on the record files already present in the database. To perform a new background computation, the user must first press the “Clear” button. Then, the user should provide a suggestive description for the background, set the date and insert the counts as well as the measurement time.

If a single measurement is available, such as a long alpha measurement of about 70000 seconds, then the software assumes a Poisson distribution for handling uncertainties. Otherwise, the Gauss (normal) distribution is used (see Appendix 2 for details). By pressing “Perform analysis” button the uncertainties are computed, compared using the student t-test and Fisher f-test and data can be saved in database. Student t-test and Fisher f-test are performed by using numerical methods rather than some pre-computed coefficients from tables. There are various numerical methods available for handling statistics, solving equations, modeling data by fitting functions such as Numerical Recipes in C [Press et. al].

Having multiple records for background, the user can also perform the long time analysis test to see if the background really changes in time and make the appropriate investigations (see Appendix 3 for details).

### 1.1.2 The efficiency

The screenshot shows the 'Beta efficiency evaluation' application window with the 'Auxiliary data' tab selected. The window has three tabs at the top: 'Auxiliary data' (selected), 'Data', and 'Results'. The main area is divided into sections: 'Nuclide selection', 'Activity date', and a large table for 'Background selection'.

**Nuclide selection:** A dropdown menu shows 'Nuclide: Ac-223' and a 'Description:' input field.

**Activity date:** A date picker shows 'Day: 14', 'Month: 10', 'Year: 2013', and a 'Today' button.

**Background selection:** This section contains two tables.

ID	NUCLIDE	DESCRIPTION	DATE	ACTIV_BQ	ACTIV_BQ_EXTUNC	HALF
1	Sr-90	(Sr+Y)90 SEB 8-31	2000-05-01	823.8	37.071	9.0696
2	K-40	40mg_40K	2004-05-03	0.6453	0.03226499999999995	4.02990
3	K-40	66mg_40K	2004-05-03	1.051	0.05255	4.02990
4	K-40	82mg_40K	2004-05-03	1.3085	0.06542500000000001	4.02990
5	Sr-90	32mg	2007-08-01	378.0	15.12	9.0696
6	Sr-90	20mg	2007-08-01	65.17	2.606800000000002	9.0696
7	Sr-90	40mg	2007-08-01	130.35	5.213999999999995	9.0696
8	Sr-90	60mg	2007-08-01	195.52	7.8208	9.0696
9	Sr-90	80mg	2007-08-01	260.69	10.4276	9.0696
10	Sr-90	100mg	2007-08-01	325.86	13.0244	9.0696

**Delete** button is located below the table.

ID	DESCRIPTION	DATE	CPM	CPS	CPS_ERR_GAUSS
1	fbeta	2011-07-07	9.35401467657022	0.15590024460950366	0.004293290007632001 0.
2	fond_03.05.2004	2004-05-03	10.92002032124012	0.18200033868733534	0.009178643061154436 0.
3	fondBetaStandard	2011-06-27	9.108013974909476	0.15180023291515793	0.005349574652753703 0.

The auxiliary data tab contains the nuclide selection and the background selection. For instance, the measurement of Sr90 standard source implies the selection of Sr90 nuclide. User must provide the source activity, its uncertainty and the date when the

activity is known. These data are usually obtained from the standard source certificate provided by the vendor. The software automatically computes the source activity at measurement date using the well-known radioactive decay law.

The data tab is very similar to the background data tab but the user can also manually set the detector efficiency (e.g. if an external metrology institute performed the detector calibration).

By pressing “Perform analysis” button, the output is shown in the “Result” tab. For details about the detection limit see Appendix 4 and for details about alpha/beta efficiency see Appendix 5.

### 1.1.3 The sample

The screenshot shows the 'Beta sample analysis' software window with the 'Auxiliary data' tab selected. The interface includes tabs for 'Auxiliary data', 'Data', and 'Results'. The main area displays a table for 'Efficiency selection' and another for 'Background selection'.

ID	DESCRIPTION	DATE	EFF	EFF_EXTUNC_GAUSS
1	eff_Sr_Y_90_manual	2004-05-03	0.36854	0.03282
2	eff_40K_40mg	2004-05-03	0.3342134166444593	0.04265939103535494
3	eff_40K_66mg	2004-05-03	0.32699249512844625	0.04459054993473112
4	eff_40K_82mg	2004-05-03	0.31053608912446656	0.03089080210286158
5	eff_90Sr_20mg	2012-02-23	0.1581595386255605	0.006407190507343445
6	eff_90Sr_40mg	2012-02-23	0.16220933128545806	0.0065442733533754216
7	eff_90Sr_60mg	2012-03-01	0.1532183528075324	0.006186181135095067
8	eff_90Sr_80mg	2012-03-01	0.15642371066904617	0.006275393143607988
9	eff_90Sr_100mg	2012-03-01	0.15417524657353043	0.006191124148257726
10	fondBetaStandard	2011-02-24	0.15417524657353043	0.006191124148257726

ID	DESCRIPTION	DATE	CPM	CPS	CPS_ERR_GAUSS
1	fbeta	2011-07-07	9.35401467657022	0.15590024460950366	0.004293290007632001
2	fond_03.05.2004	2004-05-03	10.92002032124012	0.18200033868733534	0.009178643061154436
3	fondBetaStandard	2011-06-27	9.108013974909476	0.15180023291515793	0.005349574652753703

The auxiliary data tab contains the following:

- Final sample activity which is derived from the measured activity by multiplying with a factor. For instance, if one wants to assess the sample activity in Bq/kg, the measured activity is finally divided by the sample mass. In this case, the user chooses the denominator and sets the mass value and its uncertainty (in same units as the mean value) as well as the unit (Bq/kg).

- Efficiency selection from the database which was previously computed.

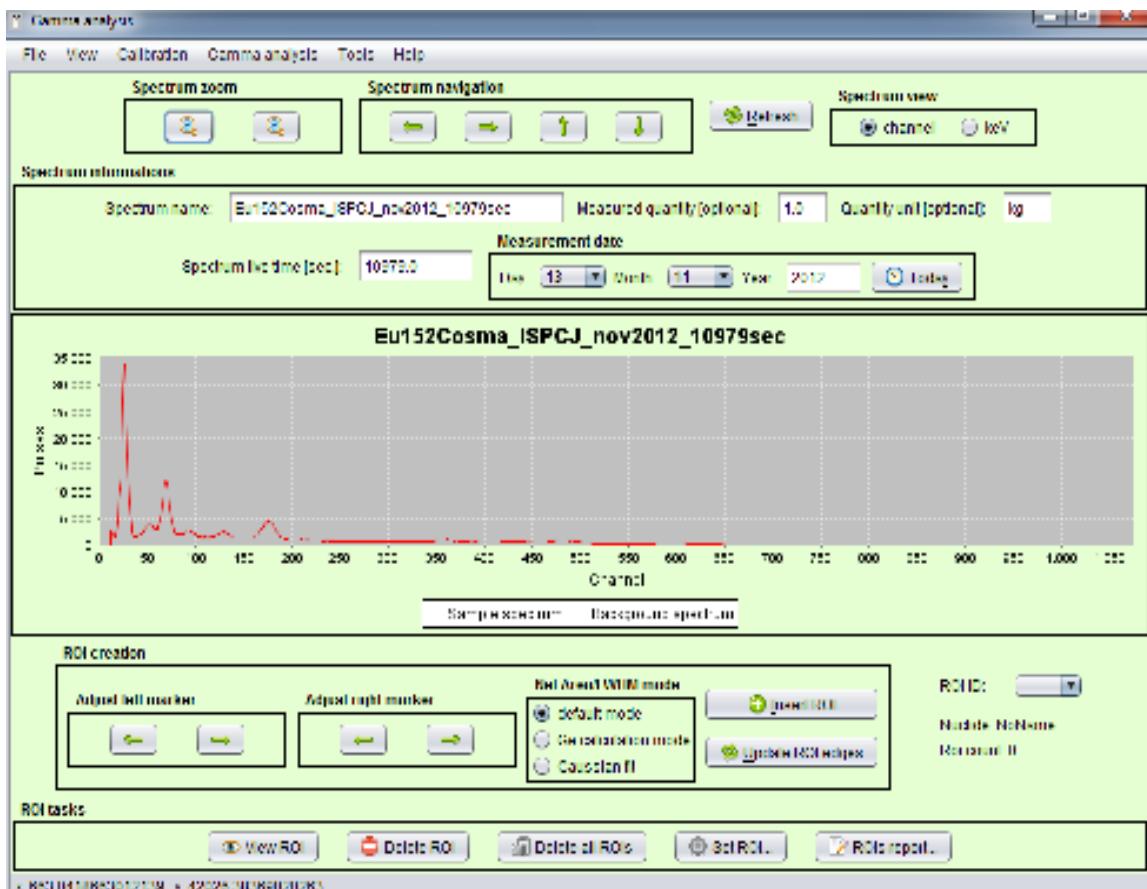
- Background selection from the database.

The data tab is similar to the data tab for efficiency calculation.

By pressing the “Perform analysis” button, the output is shown in the “Result” tab and user has the option to save data or to print the output in pdf format. For details about the detection limit see Appendix 4 and for details about sample activity see Appendix 6.

## 1.2 The gamma analysis

### 1.2.1 Main features



Commercial acquisition software such as Assayer, Gamma2000, Maestro, etc. can save the gamma spectrum in various formats. The ASCII (text) format can be used as input file for this module with some modifications which have to be done by the user. The input file must contain no other text than either one column of data representing the counts from the first channel to the last one or two columns of data representing the channel number and the counts recorded in that channel. In order to load the input spectrum file, the user must choose the “File” menu and then select “Open spectrum file”.

Once the spectrum is loaded, user can navigate it, zoom it or choose between channels or keV if the energy calibration is already performed. The “Refresh” button simply restores the spectrum at the original view. Alternatively, the spectrum navigation can be done by holding both CTRL key and the left (or right) mouse button and moving the mouse. The spectrum zoom can also be performed by using the mouse wheel. The image of gamma spectrum can be copied, saved or printed using the contextual menu (right click on chart).

The user must provide a spectrum name, the spectrum live time (most of acquisition software automatically perform the dead time correction by providing live time –shorter, set by user- and real time –longer, actually this is the real time for the

measurement) and the measurement date. The optional measured quantity and its unit is further used for determination of concentration, e.g. Bq/kg.

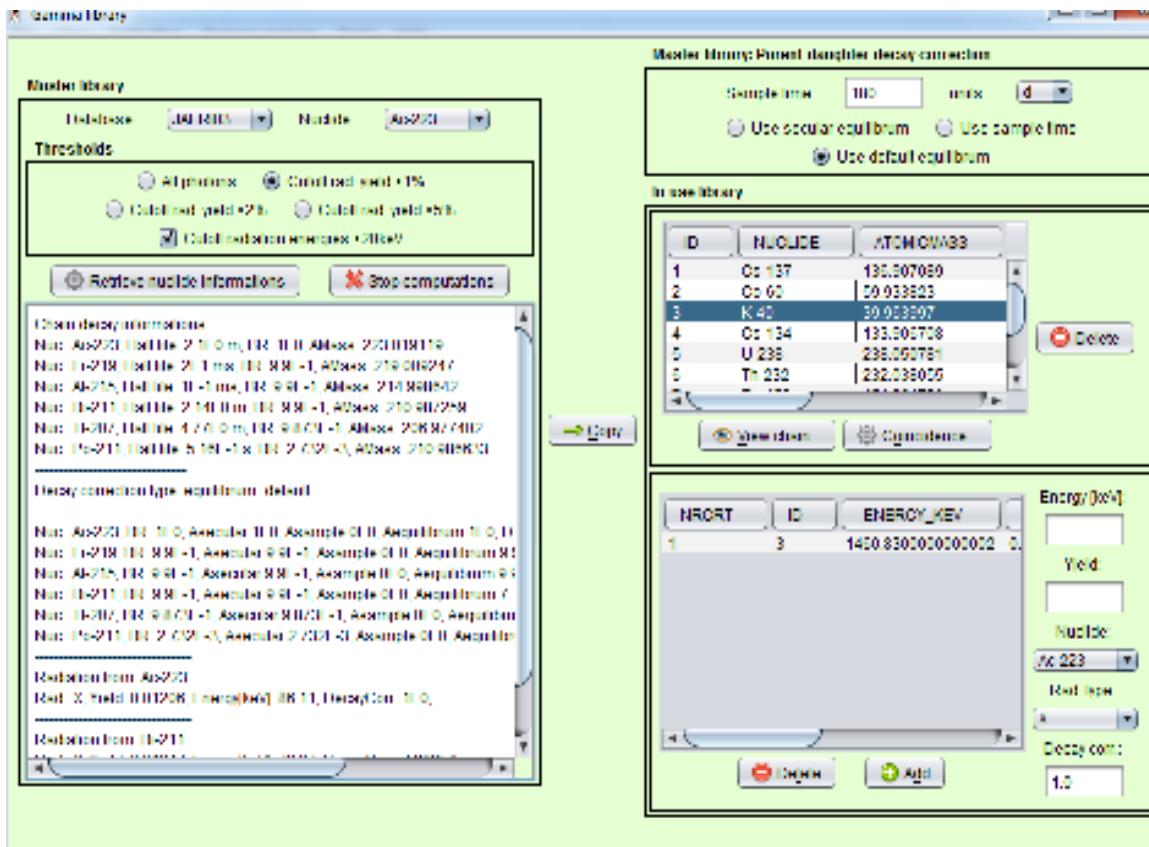
By hovering the mouse over the spectrum, the status bar displays the  $x$  coordinate (channel or keV depending the view) and the  $y$  coordinate (counts or its natural logarithm or its square root depending the selection from the “View” menu).

Creating a Region of Interest (ROI) is done simply by left click twice on spectrum in order to set two markers, usually at peak edges, and by pressing the “Insert ROI” button. The markers can be fine adjusted by using the appropriate controls. After creating the ROI, the user can also chooses to modify only its edges in order to set a more appropriate continuum background if the spectrum is complicated, e.g. overlapped peaks in a spectrum acquired by NaI gamma detectors. This may be done by setting new markers and pressing the “Update ROI edges” button. Using this option, the user can set the edges of the continuum background at large distance from the desired peak in such way that the continuum background is much more appropriate and then perform peak unfolding by using the Gaussian fit operation (see Appendix 7). The net area is computed using a channel by channel subtraction of the continuum Compton background and ambient background for the “default mode” or the “Ge calculation mode”. The “Gaussian fit mode” involves the computation of the net area by approximating the shape of the peak with a Gaussian function (see Appendix 7).

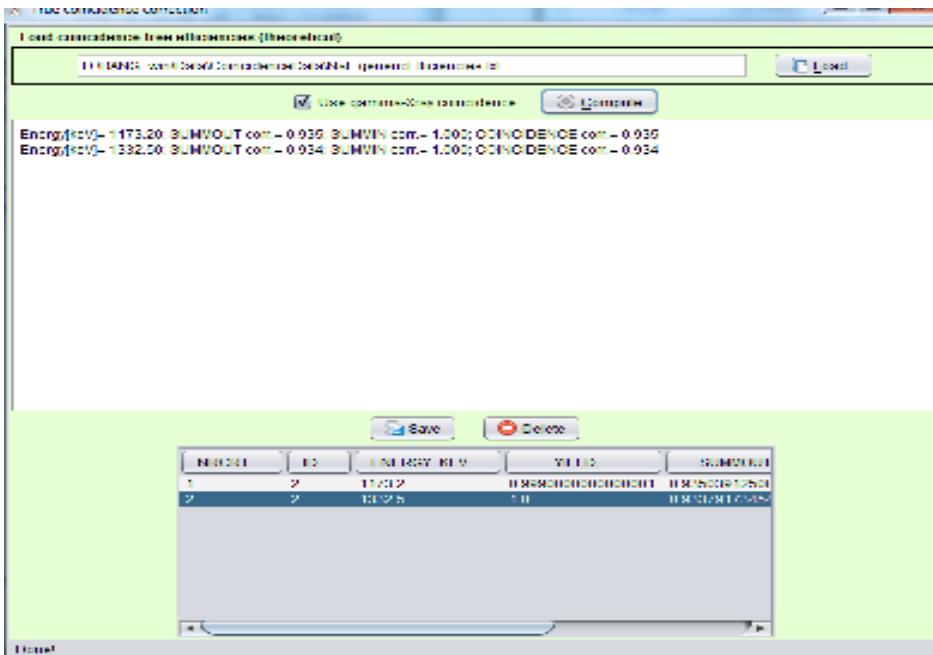
Finally, the user can visualize the ROI (useful when Gaussian fit is performed), delete current ROI or all ROIs. The computation of the activity associated to a particular ROI is done after setting the ROI and then pressing the “ROIs report” button. Setting the ROI means associating the ROI to a particular nuclide from library using “ROI set” button. Details about the corrections done when creating an in-use library, are given in Appendix 8. The spectrum can be saved as being related to background, standard source or regular sample by using the “File” menu and the “Save spectrum” option.

The sample activity is performed by using the information of all ROIs and choosing the “Gamma analysis” menu and then pressing “Sample report”. In order to perform an accurate analysis a good energy and FWHM calibration as well as efficiency calibration is required (see Appendix 9).

Choosing an in-use library of nuclides is a major challenge for first-time users.



First, choose a master library, e.g. JAERI03 [ICRP 1983, Endo 1999, Endo 2001], the nuclide, the thresholds (cutoffs) and the desired parent-daughter decay correction mode (see Appendix 8). Then press the “Retrieve nuclide information” and copy data into the library in use. After copying the nuclide, the user can alter database with its own data (in the lower right side of the window) or use the data computed by the application. The radioactive chain can be visualized and the coincidence correction can be performed (see Appendix 8 for details).



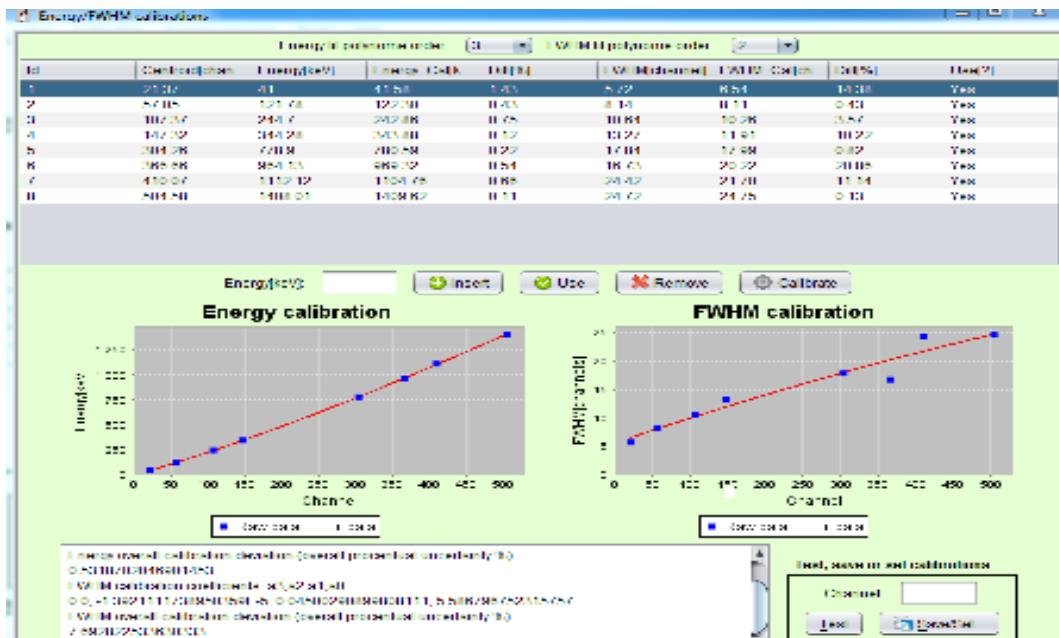
For simplicity, the following approximation is used: the X-ray are emitted only during an EC (electron capture) decay and all X-rays quanta contribute to the main gamma-gamma true coincidence. The user can also choose to perform X-rays - gamma coincidence correction or not. Knowledge of theoretical peak efficiency and total efficiency (coincidence free efficiencies) for several gamma energies are required. These efficiencies are specific to the user's detector and must be tabulated in a 3 columns text file located in the "CoincidenceData" folder. The columns are: energy in keV, peak efficiency (per 100 photons) and total efficiency (per 100 photons). Alternatively, the user may use the generic efficiency files for Ge or NaI crystals but it is strongly recommended to use the theoretical application "Beta\_Gamma\_MC" to perform an accurate Monte-Carlo simulation of radiation transport inside the detector in order to compute the theoretical detector efficiencies.

The nuclide interference correction is not a major concern since these interferences affect mainly the Compton (continuum) background which is subtracted in order to compute the ROI net area. There are Compton counts associated to any real gamma energy in whole spectrum at left side of the peak. The true nuclide interferences refer to multiple lines in a ROI causing the peak to be asymmetrical. Again, this is not a matter of concern since the user has the possibility to choose the peak (few channels near the max value) and its edge separately (wide) and then performing a gaussian fit (unfolding) for that particular peak (ROI).

The sample measurement time (spectrum live time) correction is performed after all ROIs are set (see Appendix 8). Also, in certain conditions (there is only one nuclide in spectrum and global efficiency for that nuclide exists) the sample report contain the gamma global activities.

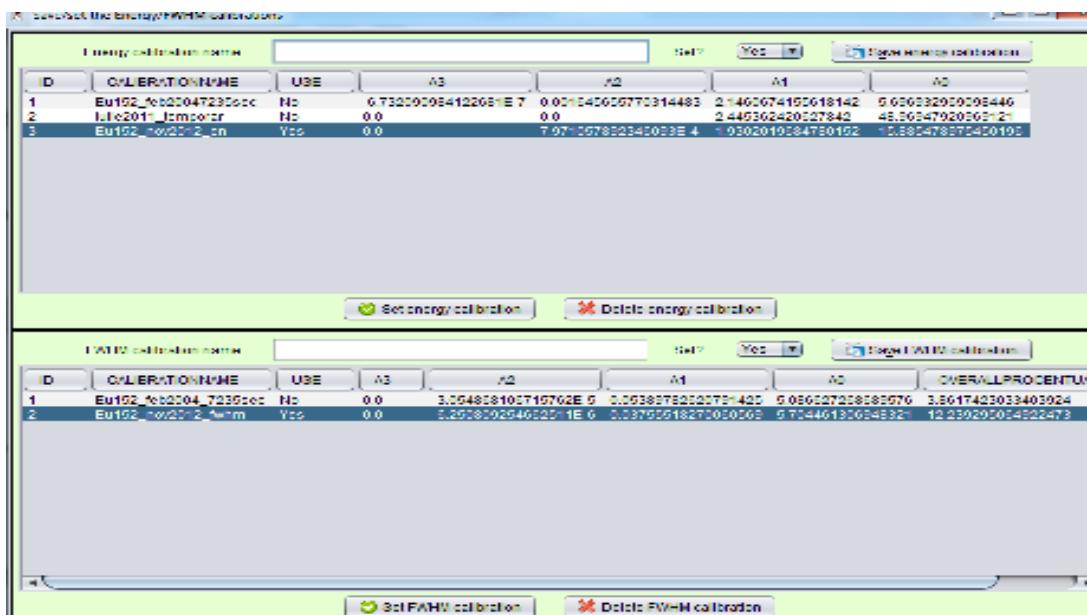
In order to subtract the ambient background, the user must load it from the database. This is done by selecting "File" menu and choosing "Load ambient background spectrum...".

After all ROIs in spectrum are set, the gamma calibrations (energy/FWHM and efficiency calibrations) can be performed by selecting the “Calibration” menu.

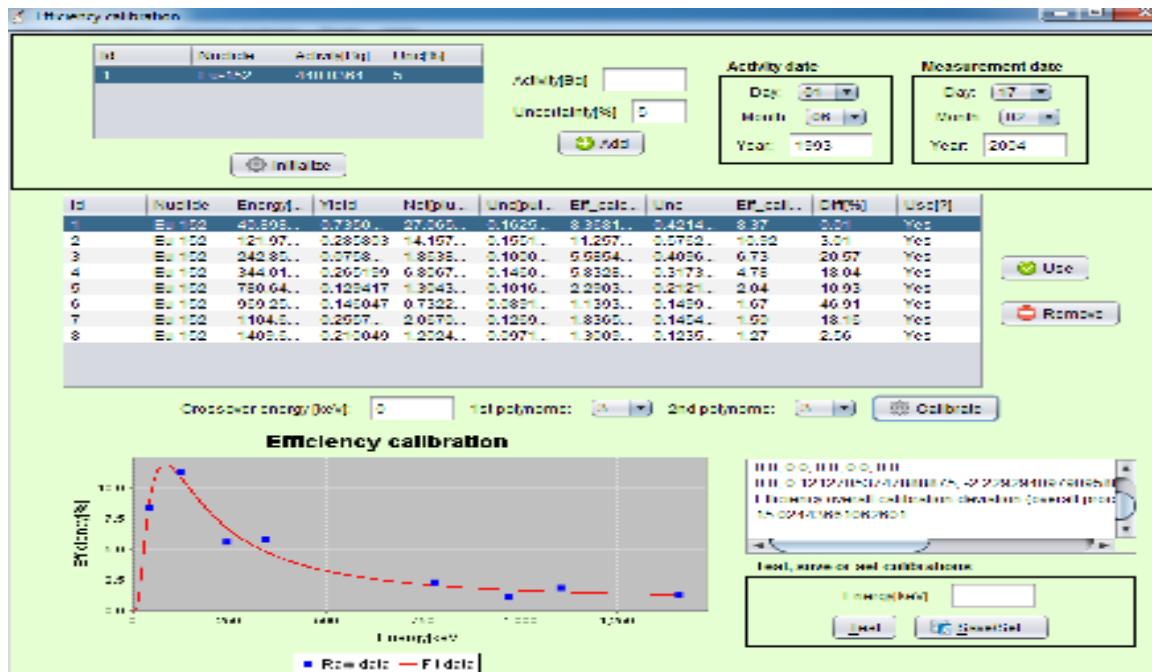


The user must select the desired polynomial order, insert the known energy (e.g. from library) for each ROI centroids, set the ROIs to be used (or removed) for calibration and press the “Calibrate” button.

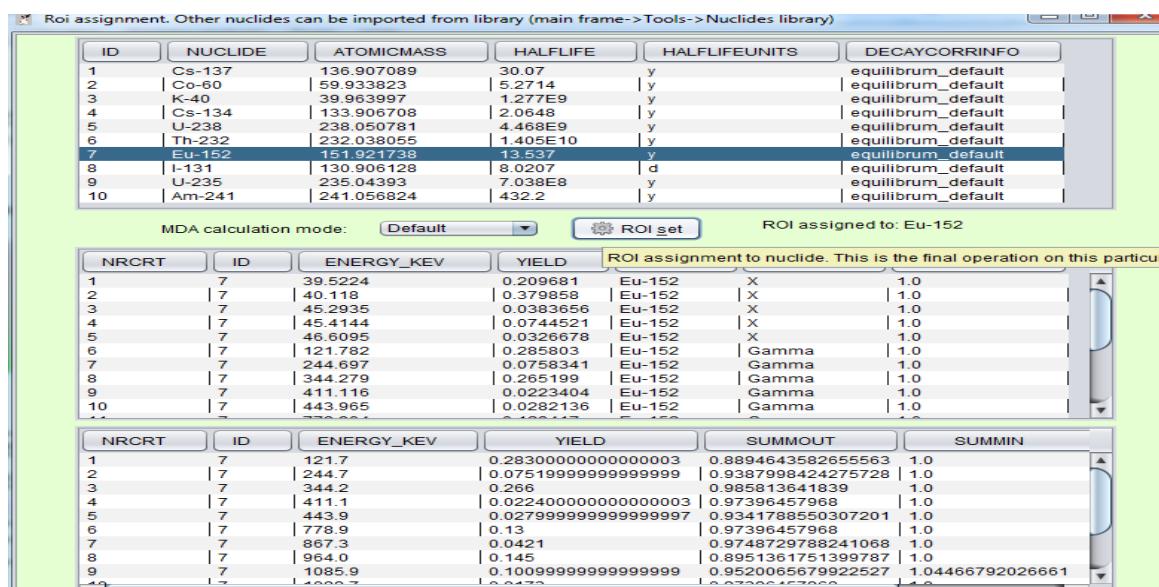
Finally, the calibration can be tested and saved. The user can also choose between multiple calibrations by pressing the “Save/Set” button.



Efficiency calibration requires the knowledge of the activity of the standard source used in calibration. The standard source can contain multiple nuclides, therefore the activity must be known for each of them. After setting the nuclide activity, its uncertainty at activity date and measurement date, the user must press the “Add” button.



The efficiency is computed for each ROI previously associated to a specific nuclide using “ROI set” button.

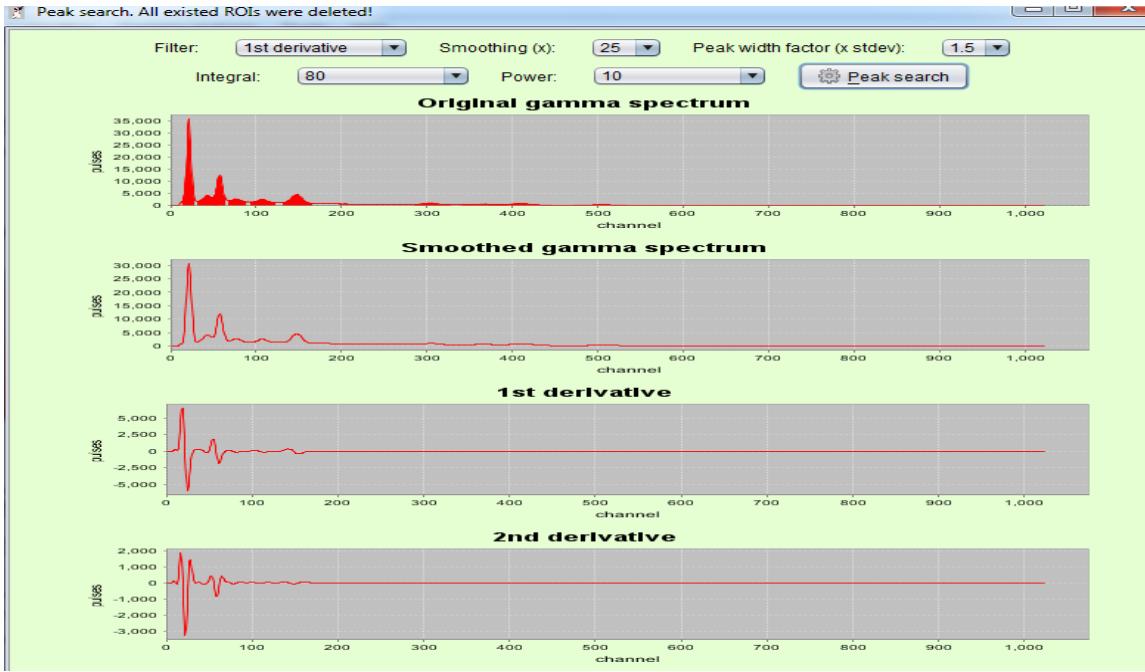


Finally, the user can choose the crossover energy, the polynomial order, can set the ROIs to be used (or removed) for calibration and press the “Calibrate” button. The

calibration can be tested and saved. The user can also choose between multiple calibrations by pressing the “Save/Set” button.

### 1.2.2 Optional features: Automatic peak search and peak identify

These features can be accessed using the “Tools” menu.

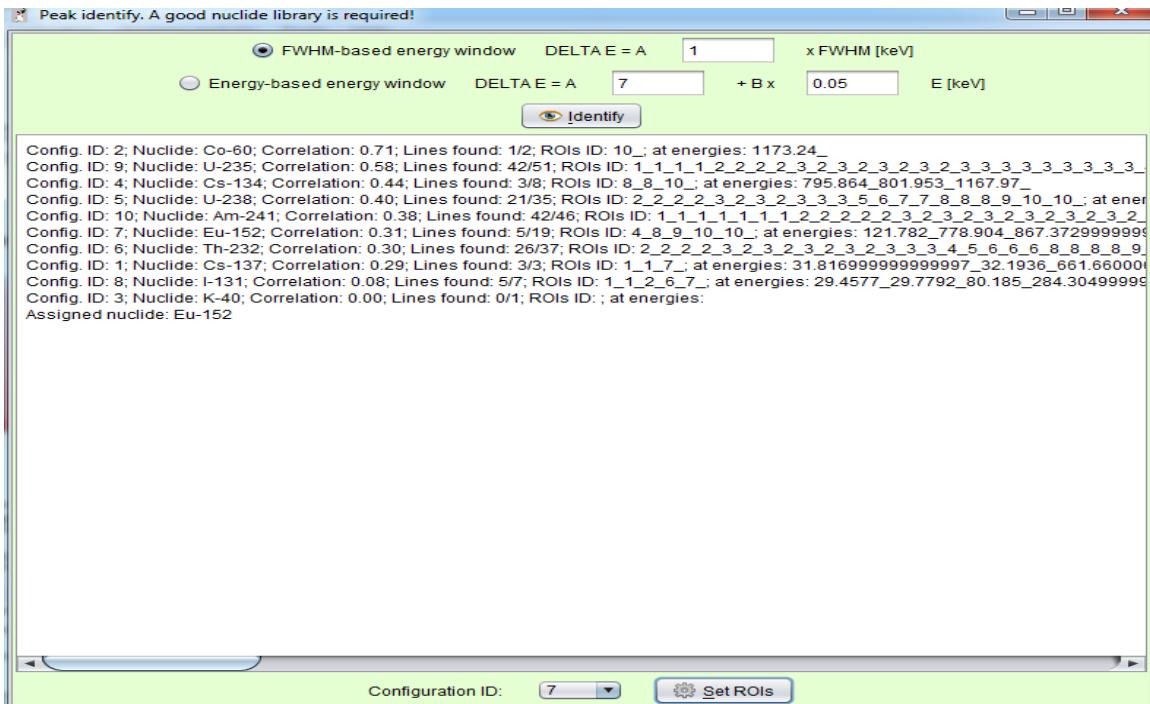


The original spectrum is smoothed using the smoothing moving average relationship counts in each channel (Savitzky & Golay 2<sup>nd</sup> order spline):

$$y_{i-1} = \frac{1}{35}(-3y_{i-3} + 12y_{i-2} + 17y_{i-1} + 12y_i - 3y_{i+1})$$

Then, the peak search is done based on a filter (1<sup>st</sup> derivative or 2<sup>nd</sup> derivative of the smoothed spectrum) and some sensitivity parameters (Integral, Power). Minimum integral and minimum power parameter is set in order to set a valid ROI of width given by the Peak width parameter (times standard deviation) around the peak. The integral parameter refers to the peak area while the power parameter refers to integral parameter divided by peak width [A.N.S.].

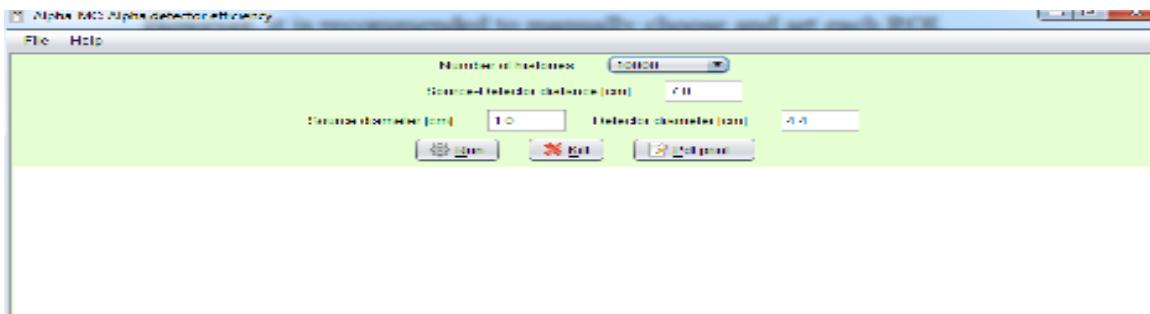
Peak identification is performed by setting a proper energy window using two semi-empirical methods: FWHM based method or Energy based method [A.N.S.]. The program automatically searches for energy lines in the library which match (within the energy window interval) with the ROI centroid energy. Then several possible configurations (solutions) are presented and the user is left to decide which solution is appropriate.



However, it is recommended to manually choose and set each ROI.

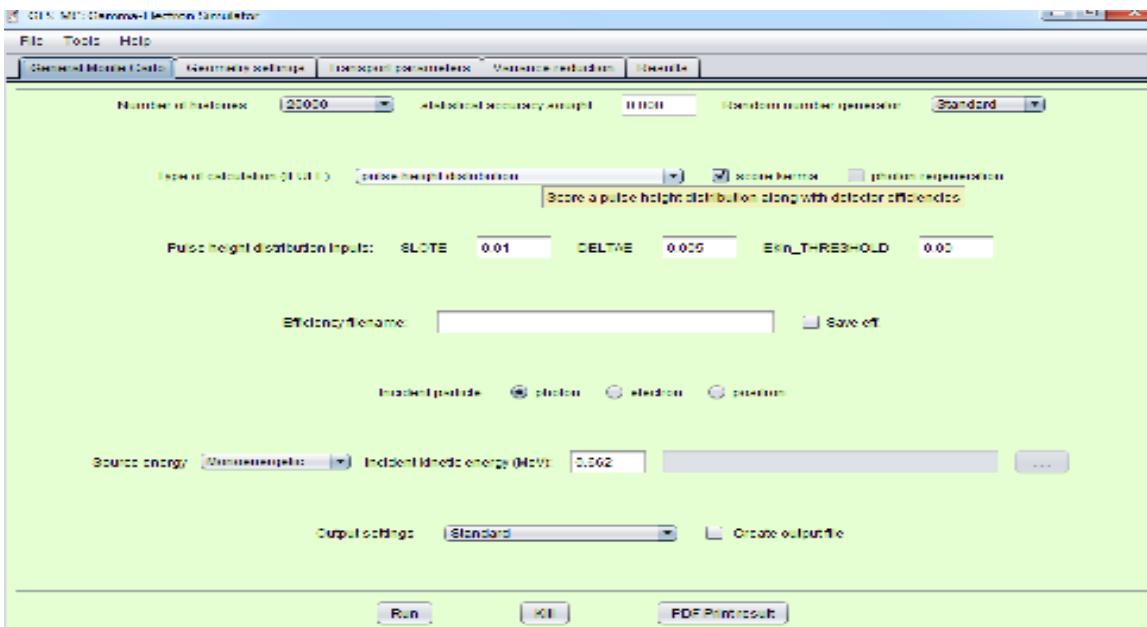
## 2. Theoretical based applications

### 2.1 Alpha\_MC



A typical standard source for alpha analysis has a disk shape (very thin thickness to avoid self-absorption). The efficiency is calculated based on distance from the source to the detector, source diameter and detector diameter (see Appendix 10 for details about this very simple algorithm). Due to the specifics of alpha interactions with matter, if the particle reaches the sensitive detector volume then it is considered to be recorded. Also the possible interaction with air is neglected and therefore, the results are accurate only for alpha systems equipped with a vacuum pump.

## 2.2 Beta\_Gamma\_MC



Monte Carlo sampling for an electron/photon shower is much more complicated than the simulation for alpha particles. This application module is mainly designed for computation of the detector peak efficiency and the detector total efficiency (whole spectrum) which are further used in gamma spectroscopy, for sample activity calculations. The module can also compute the detector efficiency for beta radiation (electrons and positrons), the absorbed dose and kerma in any cylindrical (RZ) geometry, the attenuation and the scatter fraction of radiation in detector walls and it can be used as a radiological application for evaluation of scattered X radiation (secondary radiation) at a user defined distance from the patient. These computations are performed using the Monte Carlo simulation technique and the simulation time can vary from several seconds to several minutes. This module is based on the well-known radiation transport theory and algorithms. The routines are taken from the EGSnrc simulation toolkit source code, developed by SLAC (Stanford Linear Accelerator Center, USA) and NRC (National Research Council, Canada) [Kawrakow, Nelson]. The original EGSnrc toolkit is a general purpose package for the Monte Carlo simulation of the coupled transport of electrons and photons in an arbitrary geometry for particles with energies above a few keV up to several hundreds of GeV.

Summary of EGSnrc capabilities (and this module) concerning the physics behind are:

- the radiation transport of electrons (+ or -) or photons can be simulated for any element, compound, or mixture.
- the transport of both photons and charged particles is performed in steps of random length rather than in discrete steps.
- bremsstrahlung production is calculated using either Bethe-Heitler cross sections or the NIST (National Institute of Standards and Technology, USA) cross sections.

- positron annihilation in flight and at rest (the annihilation quanta are followed to completion).

- multiple scattering of charged particles by Coulomb scattering from nuclei is handled using a new multiple scattering theory which overcomes the shortcomings of Moliere multiple scattering theory. It allows steps of any size and moves seamlessly from a single scattering model for short steps to an accurate multiple scattering model at large steps. The user has the option of scattering based on Rutherford scattering or scattering accounting for relativistic and spin effects.

- Moller ( $e^-e^-$ ) and Bhabha ( $e^+e^-$ ) scattering. The exact rather than asymptotic formulae are used.

- continuous energy loss applied to charged particle tracks between discrete interactions. Total restricted stopping power of charged particle consists of soft bremsstrahlung and collision loss terms. Collision loss is determined by the restricted Bethe-Bloch stopping power with Sternheimer treatment of the density effect in the general case but with provision of using an arbitrary density effect correction and data supplied to use the density effect recommended by the ICRU in Report 37.

- pair and triplet production.

- Compton scattering, either Klein-Nishina or bound Compton and radiative Compton effect.

- coherent (Rayleigh) scattering can be included by means as an option.

- photoelectric effect.

- relaxation of excited atoms after vacancies are created (e.g. after photoelectric or Compton scattering events) to create fluorescent photons (K, L, M shells) and Auger and Coster-Kronig electrons may be produced and tracked if requested.

- the electron radiation transport is handled by using the condensed history theory of type II (the “catastrophic” interactions are separately treated and using the multiple scattering theory for the rest).

- sampling the angular distribution of the photo-electron is available as an option.

- bremsstrahlung angular sampling and pair angular sampling are available for several methods.

- several algorithms for electron step and boundary crossing sampling are available.

- electron impact ionisation can be sampled in several ways.

- a variety of variance reduction techniques have been “built-in” to improve the simulation efficiency.

Summary of this module main features, related improvements and changes in core algorithms are:

- it has an user-friendly graphical interface (GUI) for all the above mentioned applications.

- several minor bugs fixed in original EGSnrc system codes (e.g. reading data from files, some inconsistencies in variable control, etc.). Minor changes in default settings.

- an improved algorithm for solid angle evaluation and incident fluence of radiation. Hence, an important change in starting the shower simulation algorithm has been made.

- additional useful application are: a test for Klein-Nishina sampling using several algorithms (the EGSnrc based algorithm provides the best results. Other well known methods are: Khan and Wielopolsky); material creation for later use in simulation, providing useful information such as computation of cross sections for electrons, positrons and photon interactions as well as suggestive charts; calculation of gamma activities based on computed efficiencies and the comparison with experimental efficiencies; the correction of net counts due to nuclide interferences (e.g. 238U, 232Th and 40K); the correction of net counts due to true coincidence (photons emitted in cascade); calculation for kerma parameter  $g$  as well as mass energy transfer and mass energy absorption coefficients, an application for computing the ideal surface detector efficiency for an electron source with box or point source geometry; a quick gamma global efficiency computation using a hybrid analytical/MonteCarlo algorithm (crude estimate).

General settings are:

- number of histories;
- statistical accuracy sought (if not 0.0, the simulation ends when uncertainties in dose calculation become equal with this value);
  - random number generator picker (standard -default- means the ranlux generator);
    - type of calculation – *dose and stoppers* for dose computation only in the main detector region, *Aatt and Ascat* for computation of the attenuation and the scatter fraction of radiation in the main detector region, *pulse height distribution* used mainly for calculation of detector efficiencies and detector response, *scatter fraction* for dose and scatter fraction in all geometrical regions; various radiation *fluence* calculations, *stopping power ratio* evaluation and *bremssstrahlung* calculation mainly used in simulation of an X-ray generator.
    - score kerma option;
    - photon regeneration option. Caution: this option should not be used for efficiencies calculations. Documentation about this option can be found in some EGSnrc papers [Kawrakow, EGSnrc website].
    - Pulse height distribution inputs: SLOTE and DELTAE- their meaning are shown in tooltip windows and detail documentation about these parameters can be found in EGSnrc papers [Kawrakow]; Ekin\_THRESHOLD is the minimum value of incident electrons kinetic energy required by the simulation to be performed (below this threshold, it is considered to not have a valid signal in detector).
    - if “save eff” checkbox is selected then a suggestive filename must be inserted in the corresponding text field. Efficiencies are automatically saved in application/Data/eff subdirectory and can be viewed with any text editor.
    - source energy can be monoenergetic or spectrum. In the last case the preset spectrum file should be load. The spectrum files can be found in application/Data/spectra subdirectory. Caution: The spectrum inputs can be used for dose evaluation not for efficiency evaluation! If the source is monoenergetic, the user must type the radiation energy for further computation (e.g. efficiency evaluation). The output text file can be generated in several ways (results only, standard and full detail).

Geometry settings contains two parts: Source settings and Detector settings.

### Source settings

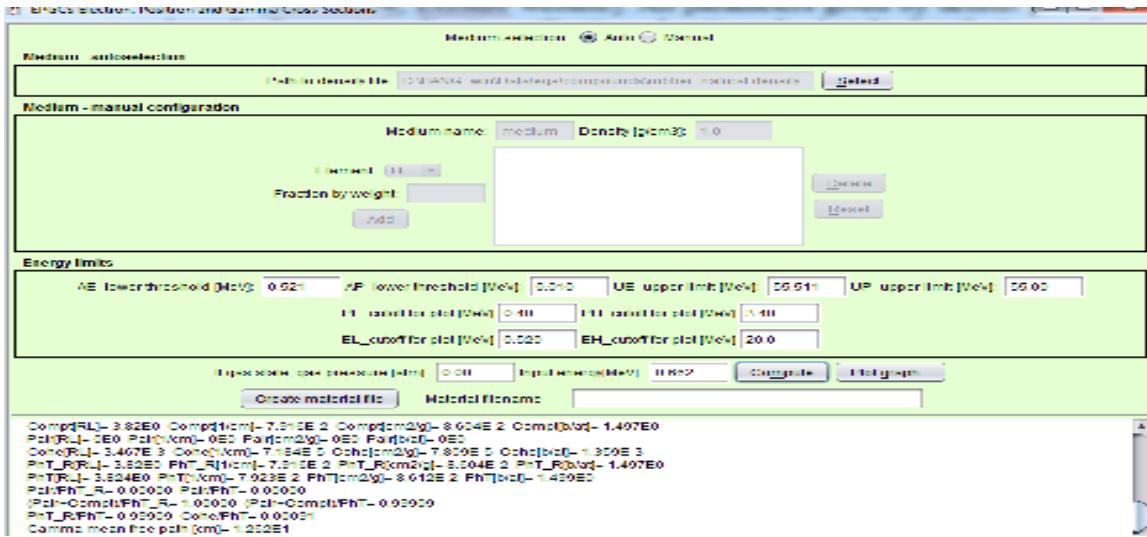
- incident particle type: photon, electron or positron;
- geometry type: Frontal beam, Point source, Sarpagan (means cylindrical) and Marinelli (Marinelli baker)
- source equivalent composition: e.g. H<sub>2</sub>O, NaCl, soil, concrete
- input dimensions for all geometry types.
- save and load option with default filename. The source files are stored in application/Data subdirectory having the “.src” extension.

### Detector settings:

- dimension inputs
  - detector materials for detector active region, surrounding inactive region, mounture (wall) and window. All materials can be created in “create material” section from the “File” menu. The input files (used for material creation) can be found in application/Data/compounds and application/Data/elements subdirectories. The resulted material files (final) are stored in application/Data/interactive subdirectory.
  - other detectors can be set for different custom applications. Radiology application can be used for calculation of scattered X-ray dose (secondary radiation dose) at any point around the patient (phantom).
  - save and load option with default filename. The detector files are stored in application/Data subdirectory having the “.det” extension.

Transport parameters and variance reduction settings are well defined in original SLAC and NRC [Kawrakow, EGSnrc website]. All parameters given by default provide very good results for most applications. Caution: despite *pulse height distribution* mode, for *Aatt* and *Ascat* calculations, the variance reduction techniques must be used!

## Material creation



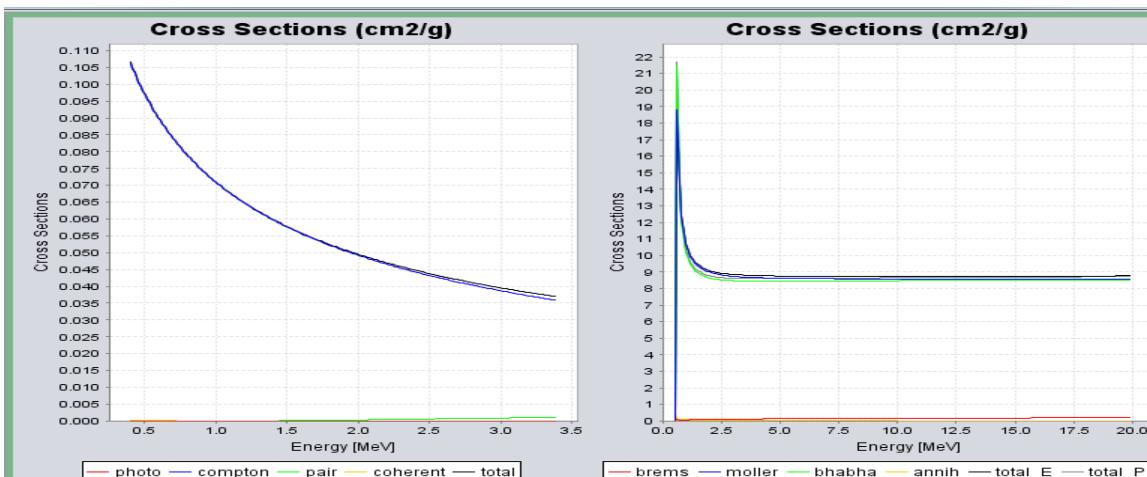
There are two possibilities for generating medium input data-file (containing cross sections parameters):

automatic – the path to the density file is required.

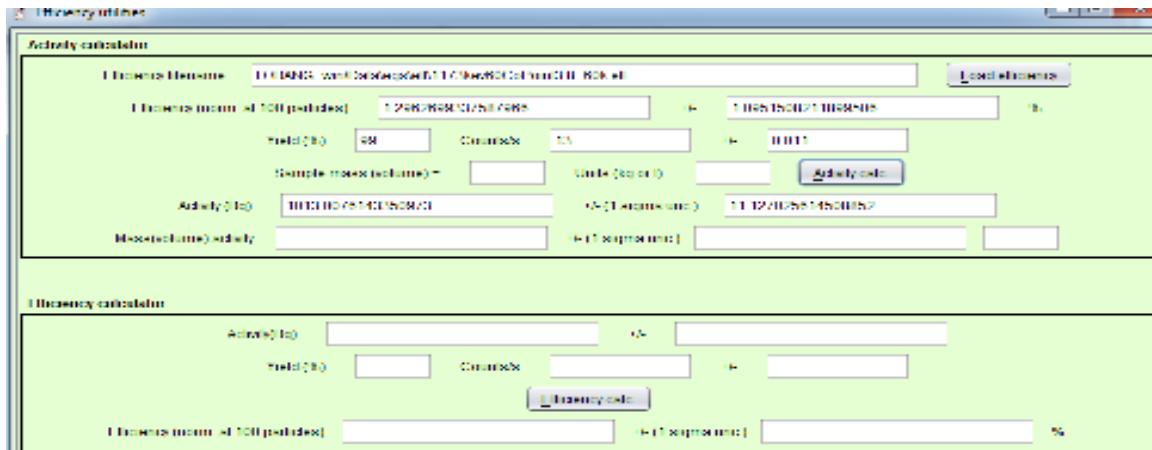
manual – a particular medium can be created having a user specified density (if is gas state, the gas pressure must be known). The user must provide all elements of the medium composition and its fraction by weight.

The energy limits parameters are well defined in SLAC reports [Nelson].

Computation of cross sections and graphical representation are available.

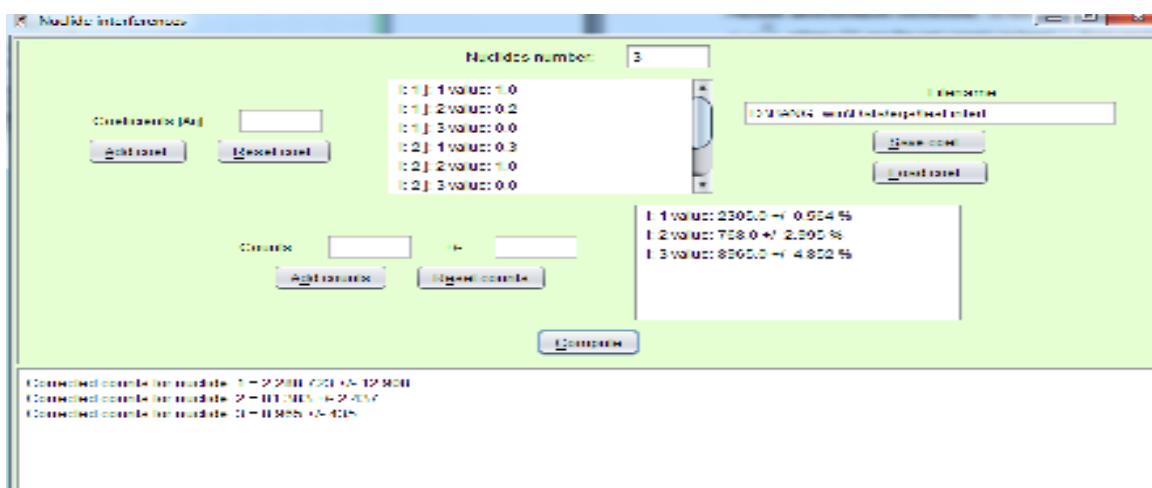


## Efficiency utilities



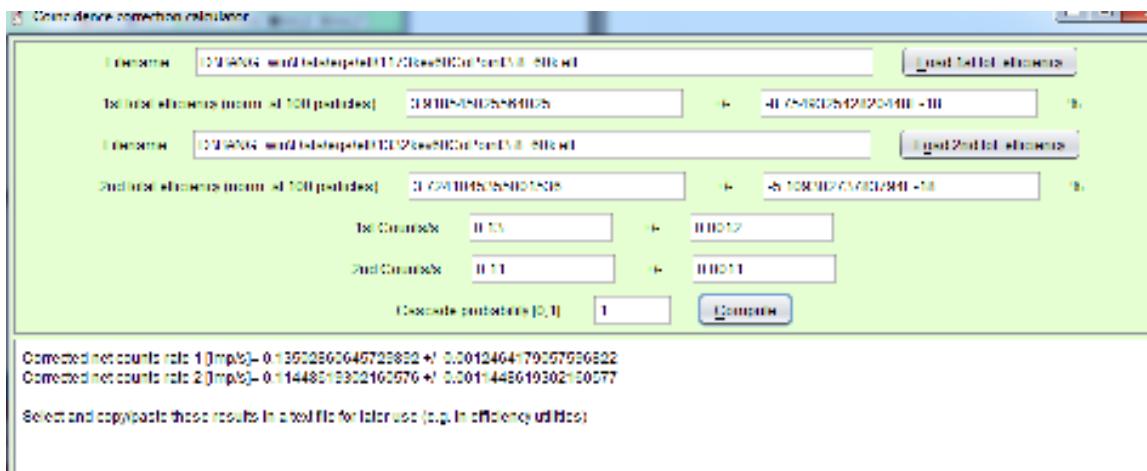
The activity can be easily computed based on the pre-saved efficiency, radiation yield and the net counts (this can be obtained directly from a gamma spectrum acquisition software). Also, the theoretical efficiency computed by Monte Carlo simulation can be compared with the experimental efficiency if the source activity is known.

## Nuclide interferences correction



Details about interference calculations are presented in Appendix 11.  
The corrected net counts can be used in activity calculation (Efficiency Utilities).

### Coincidence correction

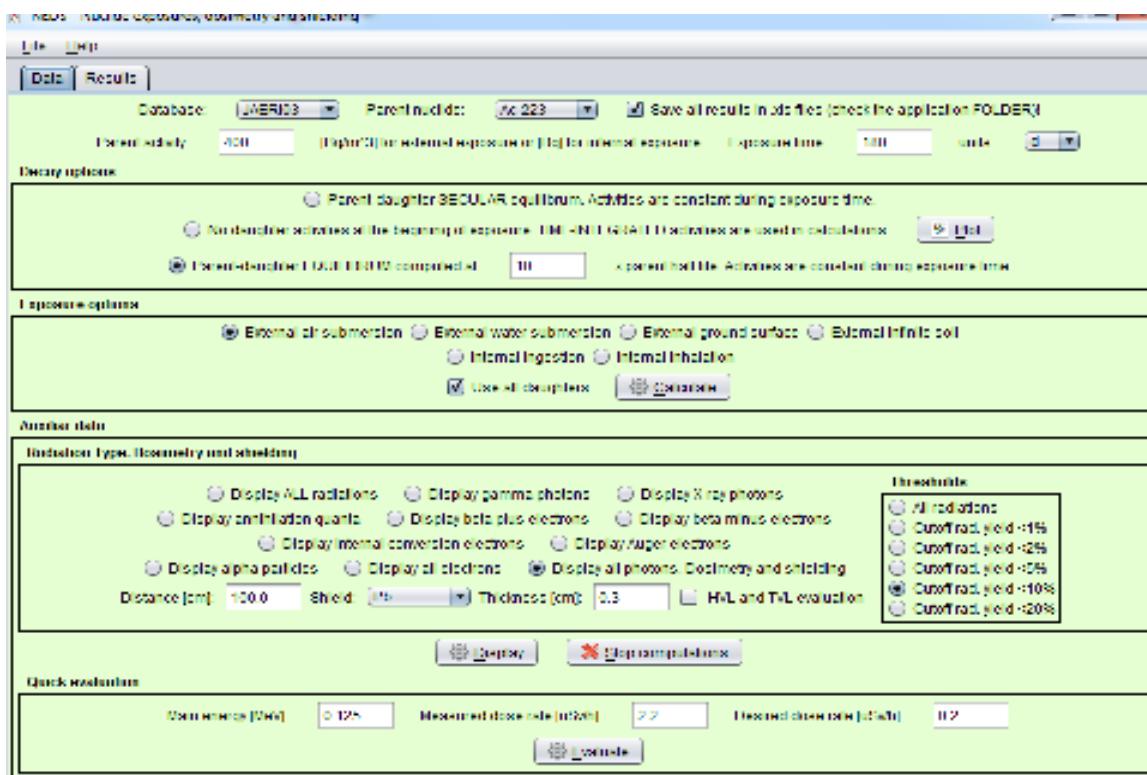


The true coincidence for two photons emitted in cascade is taken into account (e.g. 60Co case). The user must provide the observed net counts for both peaks and load the corresponding total efficiencies (also see Appendix 8).

Then, the corrected net counts can be used in activity calculation (Efficiency Utilities).

### **2.3 Nuclide exposure**

This module is useful for calculation of external and internal exposure of individuals to a specific nuclide of a given activity or concentration. It is also used for computation of chain activities of series of nuclides, retrieving useful information regarding radiation energies and branching ratios and for dosimetry and shielding.



The user must choose the database of nuclides, e.g. JAERI, Parent nuclide, Parent activity and the exposure time [ICRP 1983, Endo 1999, Endo 2001].

Decay options are self-explanatory as well as Exposure options. Then press the “Calculate” button to retrieve the results.

The parent and daughters activities are computed for every possible chain using the Bateman decay law [Bateman, Magill]. Activity of nuclides in each chain is computed at exposure time considering the parent activity is 1 Bq at time 0 and no daughters are present at time 0. Activity at equilibrium is computed at 10 times parent half-life considering parent activity being 1 Bq at time 0 and no daughters at time 0. Integral, i.e. time-integrated activity, is also computed during exposure time. Again, the parent activity is considered to be 1 Bq at time 0 and also no daughters are present at time 0.

Final activities (overall activities) are computed using data for each possible chain using a simple search and summation routine. The activity derived from secular equilibrium is displayed as well as the activity after exposure time as computed by using Bateman, the time-integrated activity and the activity at 10 times parent half-life.

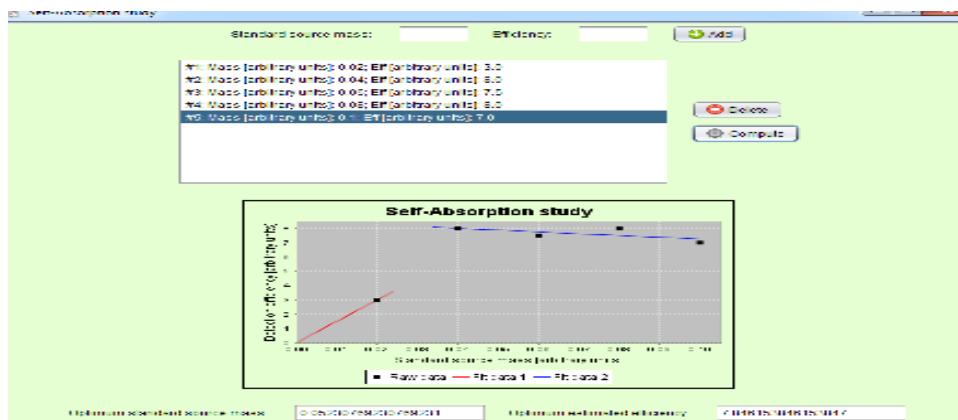
Final activities are used further for computation of organ doses (absorbed dose) of individuals which are subject to an external or internal exposure. Because all activities are previously normalized to 1 Bq, the results are multiplied by the actual parent activity. Therefore, the organ doses as well as the effective dose and the cancer risk parameter correspond exactly to the real case scenario chosen by the user.

External exposure conversion coefficients from activity to dose are taken from FGR12 database [EPA]. Internal exposure conversion coefficients from activity to dose are taken from ICRP2001 database [ICRP 2001]. The cancer risk coefficients are computed using BEIR VII recommendations [BEIR VII].

The radiation type and dosimetry and shielding section provide information about radiation energies and yields for each nuclides according to user needs. If dosimetry is selected then it computes the total tissue dose rate as well as total tissue dose rate after shielding. If HVL and TVL option is checked then it provides information of required absorbent thickness in order to reduce the total tissue dose rate at 1/2 (HVL) or 1/10 (TVL). Quick evaluation section is self-explanatory.

### 3. Other features

#### 3.1 Self-absorption study



It is useful for quick estimation of minimum sample mass to be measured in order to obtain the maximum efficiency as well as reducing self-absorption effect (for alpha and beta measurements).

The evaluation is quickly performed using the cross value of two linear functions computed by chi-fit numerical method.

#### 3.2 Long time stability for alpha beta or gamma detection systems



Long time stability consists in measuring a test source at various time intervals (e.g. each month). The experimental records (counts per seconds and its uncertainty) are compared to the theoretical values computed from the well-known decay law.

The following data are displayed:

-measurement date,

-DELTADAYS which is time interval in days from time 0 when first measurement of test source was made

-test source nuclide

-nuclide half life

- $TV$  which is the theoretical value set at time 0

- $STV3$  which is the 3 times standard uncertainty of  $TV$

- $EV$  which is the experimental value measured after a time interval DELTADAYS

- $SEV3$  which is the 3 times standard uncertainty of  $EV$

-UG is positive value for fixed maximum allowed uncertainty, e.g.  $UG=9\%$ .

It is computed  $TV \pm UG$  as well as  $EV \pm SUMS$  where  $SUMS=STV3+SEV3$ .

All data should lies between red lines otherwise appropriate measures are required.

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## **Appendix 1.**

### **Computation of detector dead time using two standard sources**

Steps:

1. Measure the first standard source and record the counts rate (counts per second, CPS)  $R_1$ .

2. Measure the second standard source and record the counts rate  $R_2$ .

3. At this step two methods are available:

Method 1: the user must provide the ratio of known activities (or their real counts rate) of the standard sources:

$$R = \frac{A_1}{A_2} = \frac{CPS_1}{CPS_2} \quad (\text{A1.1})$$

Method 2: the user must measure both sources at the same time and provide the counts rate for source 1 plus source 2 (S). This can be an experimental difficult task.

Using method 1, the detector dead time is computed by:

$$\tau = \frac{RR_2 - R_1}{R_1 R_2 (R - 1)} \quad (\text{A1.2})$$

Using method 2, the detector dead time is computed by:

$$\tau = \frac{R_1 + R_2 - S}{2R_1 R_2} \quad (\text{A1.3})$$

Proof:

According to the definition of dead time, the corrected counts (subscript c) are given by:

$$R_{c1} = \frac{R_1}{1 - \tau R_1}; R_{c2} = \frac{R_2}{1 - \tau R_2} \quad (\text{A1.4})$$

$$R = \frac{A_1}{A_2} = \frac{R_{c1}}{R_{c2}} = \frac{R_1}{R_2} \frac{1 - \tau R_2}{1 - \tau R_1}$$

Using elementary algebra one can easily find A1.2.

For method 2, we also have:

$$S_c = \frac{S}{1 - \tau S} \quad (\text{A1.5})$$

and

$$S = R_{c1} + R_{c2} \quad (\text{A1.6})$$

by experimental setup.

Substituting A1.4 and A1.5 in A1.6 and neglecting the terms in  $\tau^2$ , one can find A1.3.

## Appendix 2

### Basic statistics

Performing  $n$  independent measurements of a quantity, the results distribution follow the normal (Gauss) distribution where the best estimate of that quantity is the arithmetic mean (the average) given by [EA-4/02]:

$$\bar{q} = \frac{1}{n} \sum_{i=1}^n q_i \quad (\text{A2.1})$$

Its experimental variance is :

$$s^2(q_i) = \frac{1}{n-1} \sum_{i=1}^n (\bar{q} - q_i)^2 \quad (\text{A2.2})$$

and the experimental variance of the mean is associated with the standard uncertainty:

$$u(\bar{q}) = s(\bar{q}) = \frac{s(q)}{\sqrt{n}} \quad (\text{A2.3})$$

In this case, the degrees of freedom is given by:

$$f = n - 1 \quad (\text{A2.4})$$

Alternatively, the individual experimental variance can be computed by:

$$S(q_i)^2 = \frac{1}{n-1} \left[ \sum_{i=1}^n q_i^2 - \frac{1}{n} \left( \sum_{i=1}^n q_i \right)^2 \right] \quad (\text{A2.2'})$$

Radioactivity (by its nature) is subject to Poisson distribution because the number of radioactive nuclei is very big (in most cases the number of atoms/nuclei from a regular sample is very big) and the probability of decay/detection is very small (in most cases the nuclide half-life is very big yielding a decay probability very small). Therefore, if only one sample measurement is performed ( $q$  counts over a given time  $t$ ) based on the fact that it can be safely neglected all other uncertainties involved in measurement (except for inherent Poisson uncertainty of course) then the uncertainty of counts is given by Poisson statistics:

$$u(q) = \sqrt{q} \quad (\text{A2.5})$$

For  $n$  repeatedly measurements, it is useful to compute both Gauss and Poisson uncertainties and compare them to see if there is involved other source of uncertainties beside the inherent Poisson radioactivity uncertainty. The variance of variance is computed by using an approximate formula:

$$s_s^2 = \frac{s^2}{2f} \quad (\text{A2.6})$$

where  $s^2$  is the experimental variance and  $f$  is its degrees of freedom. The comparison of means or the comparison of variances use the same algorithm; the input data are: means and its variances or variances and its variances of variances respectively. It is performed one-tailed t-test for check if there are any significant differences between means or variances respectively [Press et. al]. For variance comparison, it is also used the Fisher f-test.

Having more than one sets of measurements each having its own mean, standard uncertainty (or experimental variance) and degree of freedom and supposing that these sets describe the same statistical population then one can define the overall variance as being:

$$s_g^2 = \frac{\sum_j f_j s^2(q_j)}{\sum_j f_j} \quad (\text{A2.7})$$

If the quantity of interest ( $y$ ) can be expressed as a linear combination of other measured quantity ( $q_i$ ) such as the net counts being the difference of gross counts and the background counts then its degree of freedom can be computed using Welch-Satterthwaite formula:

$$f = \frac{u^4(y)}{\sum_i \frac{u^4(q_i)}{f_i}} \quad (\text{A2.8})$$

For type B uncertainties, when we know the best estimate of a quantity  $q$  and its uncertainty  $\Delta q$ , the number of degrees of freedom can be approximated by:

$$f = 0.5 \left( \frac{q}{\Delta q} \right)^2 \quad (\text{A2.9})$$

The knowledge of degree of freedom is important for student t-test or Fisher f-test.

#### Propagation of uncertainty [Fornasini]

If the quantity of interest can be expressed as a function  $f$  which is a linear combination of some variables  $x_i$  and let  $c_i$  be the linear combination coefficient then we have:

$$\begin{aligned} f &= \sum_{i=1}^N c_i x_i \\ c_i &= \frac{\partial f}{\partial x_i} \end{aligned} \quad (\text{A2.10})$$

Let  $s^2(x_i)$  be the experimental variance of variable  $x_i$ . The variance for function  $f$ ,  $s^2(f)$ , when no correlation is present is computed by:

$$s^2(f) = \sum_{i=1}^N c_i^2 s^2(x_i) \quad (\text{A2.11})$$

In general, if we have correlation between variables we have:

$$s^2(f) = \sum_{i=1}^n c_i^2 s^2(x_i) + 2 \sum_{i=1}^{n-1} \sum_{j=i+1}^n c_i c_j \text{cov}(x_i, x_j) \quad (\text{A2.12})$$

In literature, it is also defined the correlation factor as being :

$$\text{cor}(x, y) = S(x)S(y)\text{cov}(x, y)$$

and it is used instead of the covariance factor.

Example 1: If  $f = ax + by$  and the correlation is present then the general case for this case yields:

$$s^2(f) = a^2 s^2(a) + b^2 s^2(b) + 2ab \text{cov}(x, y) \quad (\text{A2.13})$$

Where the covariance factor is:

$$\text{cov}(x, y) = \frac{1}{(N_1 - 1)(N_2 - 1)} \sum_{i=1}^{N_1} (\bar{x} - x_i) \sum_{j=1}^{N_2} (\bar{y} - y_j) \quad (\text{A2.14})$$

Example 2:

$$f(x, y) = xy \quad (\text{A2.15})$$

Here, there is no linear combination but we can write:

$\ln(f) = \ln(x) + \ln(y)$  and applying A2.12 we finally have:

$$\frac{s^2(f)}{\bar{f}^2} = \frac{1}{\bar{x}^2} s^2(x) + \frac{1}{\bar{y}^2} s^2(y) + 2 \frac{\text{cov}(x, y)}{\bar{x}\bar{y}}; \bar{f} = \bar{xy} \quad (\text{A2.16})$$

Example 3:

$$f(x, y) = \frac{x}{y} \quad (\text{A2.17})$$

Using same techniques as in example 2 one can write:

$$\frac{s^2(f)}{\bar{f}^2} = \frac{1}{\bar{x}^2} s^2(x) + \frac{1}{\bar{y}^2} s^2(y) - 2 \frac{\text{cov}(x, y)}{\bar{x}\bar{y}}; \bar{f} = \frac{\bar{x}}{\bar{y}} \quad (\text{A2.18})$$

Expanded uncertainty  $U$  of an estimate  $X$  is obtained by multiplying the standard uncertainty  $u$  with a coverage factor  $k$ , usually set in such way that any prior measurement yields a result that fall between  $X \pm U$  in 95% cases.

$$U = ku \quad (\text{A2.19})$$

The coverage factor  $k$  is computed using two-tailed student t-test for any level of precision or is taken from literature [Press et. all].

### **Appendix 3**

#### **Background long time analysis**

It is performed the Fisher test for two variances:  $S_a$  which is associated with long time stability and  $S_b$  which is associated with short time stability.

Let  $J$  the number of background records. For each record it is associated an average number of repeated measurements  $K_m$ :

$$K_m = \frac{1}{J} \sum_{j=1}^J k_j \quad \text{where } k_j \text{ is the number of repeated measurements in } j \text{ cycle.}$$

The mean value of counts rate of all background measurements is simply the sum of all counts rate divided by their number:

$$R = \frac{\sum_{i,j} R_{ij}}{\sum_{j=1}^J k_j} \quad \text{where } R_{ij} \text{ is the count rate of index } i \text{ in cycle } j.$$

Let  $R_j$  the mean count rate in a particular cycle  $j$ :

$$R_j = \frac{\sum_{i=1}^{k_j} R_{j,i}}{k_j} \quad \text{where } R_{j,i} \text{ is the } i \text{ counts rate in the particular } j \text{ cycle.}$$

The variance associated with  $k_j$  measurements in  $j$  cycle is:

$$S^2(R_{j,i}) = \frac{1}{k_j - 1} \sum_{i=1}^{k_j} (R_{j,i} - R_j)^2$$

Its degree of freedom is:

$$f_j = k_j - 1.$$

The overall variance for all cycles is given by (A2.7) and describes the short time stability:

$$S^2_b = \frac{\sum_{j=1}^J S^2(R_{j,i})f_j}{\sum_{j=1}^J f_j} \quad (\text{A3.1})$$

having an average degrees of freedom:

$$f_b = J(K_m - 1) \quad (\text{A3.2})$$

Note that the number of degree of freedom can also be estimated using Welch-Satterthwaite formula (A2.8) but the results do not change significantly.

The standard variance associated with J mean cycles is given by:

$$S^2(R_j) = \frac{1}{J-1} \sum_{j=1}^N (R_j - R)^2 \text{ having } J-1 \text{ degrees of freedom.}$$

This is actually the standard variance of mean values  $R_j$  therefore the standard variance associated with long time stability can be estimated as being:

$$S^2_a = K_m S^2(R_j) \quad (\text{A3.3})$$

with

$$f_a = J - 1 \quad (\text{A3.4})$$

degrees of freedom.

#### **Appendix 4**

#### **Detection limit**

Suppose we measure the background and a very low-activity sample. Let  $K_f$  be the mean value for background counts measured over a time  $t_f$  and  $K_q$  be the mean value for sample counts measured over a time  $t_q$ . Background time correction is given by:

$$K_f' = K_f \frac{t_q}{t_f} \quad (\text{A4.1})$$

Let us define the critical level  $N_c$  be the highest number of counts above mean value of background ( $K_f'$ ) and the lowest number of counts below mean value of sample ( $K_q'$ ) when we are uncertain if it is a real activity present in sample or not. By choosing a

coverage factor  $k$  (usually equivalent for 95% confidence) we have (see appendix 2 for details):

$$N_c = K_f' + k\sigma_f' = K_q - k\sigma_q; k = 1.645 \quad (\text{A4.2})$$

Here, the standard uncertainties are designated by the symbol  $\sigma$  and are related to background corrected counts and the sample counts. The coverage factor for 95% confidence is equal with 1.645. It is computed numerically and it can also be found in literature [Press et. all].

Using A4.2, the minimum net counts  $N_D$  (sample counts corrected by background) required for being certain that there is an activity is given by:

$$N_D = K_q - K_f' = 1.645(\sigma_f' + \sigma_q) \quad (\text{A4.3})$$

Referring strictly to inherent radioactivity statistics, the above uncertainties are computed using Poisson distribution and one can write:

$$N_D = 1.645 \left[ \sqrt{\frac{t_q}{t_f} K_f} + \sqrt{K_q} \right] = 1.645 \left[ \sqrt{\frac{t_q}{t_f} K_f} + \sqrt{N_D + \frac{t_q}{t_f} K_f} \right]$$

Solving for  $N_D$  one can finally write:

$$N_D = 2.706025 + 3.29 \sqrt{\frac{t_q}{t_f} K_f}$$

Therefore the detection limit in counts per second will be:

$$L_D[\text{CPS}] = \frac{2.706025}{t_q} + 3.29 \sqrt{\frac{R_f}{t_q}} \quad (\text{A4.4})$$

$$R_f = \frac{K_f}{t_f}$$

## Appendix 5. Alpha/beta efficiency

Assume we have a standard source with a known activity  $A_e$  and an uncertainty  $S_a$ . The program automatically convert the initial activity  $A_0$  at initial date (usually given by source certificate or a metrology institute) using the well-known decay law.

$$A_e = A_0 \exp(-\lambda t) \quad (\text{A5.1})$$

where  $\lambda$  is the decay constant taken from database of nuclides (e.g. JAERI) and  $t$  is the elapsed time from initial date.

Degrees of freedom associated with uncertainty  $S_a$  can be estimated by (see appendix 2):

$$f_a = \frac{1}{2(S_a/A_e)^2} \quad (\text{A5.2})$$

Let  $Q$  and  $F$  be the counts rate for sample and background,  $S_Q$  and  $S_F$  their uncertainties (Gauss and/or Poisson) and  $f_Q$  and  $f_F$  the corresponding degrees of freedom then the net count rate  $R$ , its uncertainty and degrees of freedom are given by (see appendix 2):

$$\begin{aligned} R &= Q - F \\ S_R^2 &= S_Q^2 + S_F^2 - 2\text{cor}(Q, F)S_Q S_F \\ f_R &= \frac{\frac{S_R^4}{S_Q^4 + S_F^4}}{\frac{f_Q}{S_Q^2} + \frac{f_F}{S_F^2}} \end{aligned} \quad (\text{A5.3})$$

The efficiency, its uncertainty and degrees of freedom are then computed by:

$$\begin{aligned} E &= R / A_e \\ \left(\frac{S_E}{E}\right)^2 &= \left(\frac{S_R}{R}\right)^2 + \left(\frac{S_a}{A_e}\right)^2 \\ f_E &= \frac{\left(\frac{S_E}{E}\right)^4}{\left(\frac{S_R}{R}\right)^4 + \left(\frac{S_a}{A_e}\right)^4} \end{aligned} \quad (\text{A5.4})$$

## Appendix 6. Alpha/beta sample analysis

Let  $Q$  and  $F$  be the counts rate for sample and background,  $S_Q$  and  $S_F$  their uncertainties (Gauss and/or Poisson) and  $f_Q$  and  $f_F$  the corresponding degrees of freedom then the net count rate  $R$ , its uncertainty and degrees of freedom are given by (see appendix 2):

$$\begin{aligned}
R &= Q - F \\
S_R^2 &= S_Q^2 + S_F^2 - 2\text{cor}(Q, F)S_Q S_F \\
f_R &= \frac{\frac{S_R^4}{S_Q^4 + S_F^4}}{\frac{f_Q}{S_Q^4} + \frac{f_F}{S_F^4}}
\end{aligned} \tag{A6.1}$$

If the efficiency is taken from a calibration certificate, then the computation of sample activity is given by an algorithm similar to the one used for efficiency calculation (see appendix 5). Instead of efficiency  $E$ , it is computed the activity  $A = R/E$  where efficiency  $E$  has an uncertainty  $S_E$  and the corresponding degrees of freedom are computed by an equation similar to A5.2.

If the efficiency was previously computed, then we can write:

$$A = A_e R / R_e \tag{A6.2}$$

where  $A$  is the sample activity,  $R$  is the background corrected count rate of sample and it is given by A6.1,  $R_e$  is the background corrected count rate of the standard source used in calibration and  $A_e$  is the activity of standard source when the rate  $R_e$  was computed.

According to basic statistics (briefly presented in appendix 2) and let  $S_a$  and  $f_a$  being the uncertainty and degrees of freedom for standard source (see A5.2), one can write:

$$\left(\frac{S_A}{A}\right)^2 = \left(\frac{S_a}{A_e}\right)^2 + \left(\frac{S_R}{R}\right)^2 + \left(\frac{S_{R_e}}{R_e}\right)^2 - 2 \frac{S_R S_{R_e} \text{cor}(R, R_e)}{R R_e} \tag{A6.3}$$

The degrees of freedom for sample activity associated with its uncertainty  $S_A$  are given by Welch- Satterthwaite formula:

$$\begin{aligned}
f_A &= \frac{\left(\frac{S_A}{A}\right)^4}{\frac{\left(\frac{S_a}{A_e}\right)^4}{f_a} + \frac{\left(\frac{S_R}{R}\right)^4}{f_R} + \frac{\left(\frac{S_{R_e}}{R_e}\right)^4}{f_{Re}}} \\
&\quad f_a \quad f_R \quad f_{Re}
\end{aligned} \tag{A6.4}$$

The specific activity ( $\text{Bq/kg}$  or  $\text{Bq/l}$ ) is simply computed by multiplying the above activity in  $\text{Bq}$  with appropriate factors. For instance, if the sample we measured has a mass  $m$  from a total mass  $M$  which is obtained by boiling a volume  $V$  of actual sample then the concentration  $\text{Bq/l}$  is given by:

$$C = A \frac{M}{mV} \quad (\text{A6.5})$$

It is also computed the detection limit (see appendix 4) and its corresponding minimum detectable activity (MDA) in same way the activity is computed from the sample background corrected counts rate  $R$ . The detection limit and  $R$  are also compared using one-tail t-test in order to see if the sample activity is significant above background or not.

## Appendix 7 Gamma analysis. Basics

In order to perform an accurate gamma analysis, the following must be known:  
Channel to energy conversion, i.e. the energy calibration.

Channel (or energy) to FWHM (full width at half maximum) of a ROI conversion, i.e. the FWHM calibration. This is useful to give a hint if there are multiple energy lines inside a particular ROI or not.

Channel (or energy) to efficiency conversion, i.e. the efficiency calibration. This is vital for converting the net area of a ROI to activity in Bq associated to that ROI.

The above calibrations are presented in appendix 9.

Any ROI is subject to two kind of background subtraction: The subtraction of ambient background (counts in all ambient background channels are scaled based on actual sample time) and the subtraction of Compton (continuum) background (each energy line associated to a valid peak in spectrum, generates a continuum background at peak left side).

ROI analysis involves the computation of the following:

1. Start channel, StartCh and its corresponding Start energy.
2. Start edge channel and its corresponding Start edge energy.
3. End channel, EndCh and its corresponding End energy.
4. End edge channel and its corresponding End edge energy.
5. Center channel and its corresponding Center energy computed by:

$$\text{CenterCh} = \frac{\text{StartCh} + \text{EndCh}}{2} \quad (\text{A7.1})$$

6. Centroid channel and its corresponding Centroid energy computed by:

$$\text{CentroidCh} = \frac{\sum_{i=\text{StartCh}}^{\text{EndCh}} G(i) * i}{\sum_{i=\text{StartCh}}^{\text{EndCh}} G(i)} \quad (\text{A7.2})$$

where  $G(i)$  is the total (background included) gross counts in channel  $i$ .

Continuum (Compton) background is computed based on the selection of Start edge channel and End edge channel. Ambient background counts in these channels are given by:

$$B_{StartEdge,t} = B_{StartEdge} \frac{t}{t_B}; B_{EndEdge,t} = B_{EndEdge} \frac{t}{t_B} \quad (\text{A7.3})$$

where  $t$  is the sample live time and  $t_B$  is the ambient background live-time.

Sample counts in these channels corrected by ambient background counts are:

$$\begin{aligned} L_{edge} &= G(StartEdgeCh) - B_{StartEdge,t} \\ H_{edge} &= G(EndEdgeCh) - B_{EndEdge,t} \end{aligned} \quad (\text{A7.4})$$

The Compton counts associated to real Start and End channels in ROI are:

$$\begin{aligned} L &= \frac{L_{edge} * EndEdgeCh - H_{edge} * StartEdgeCh}{EndEdgeCh - StartEdgeCh} + \\ &+ StartCh * \frac{H_{edge} - L_{edge}}{EndEdgeCh - StartEdgeCh} \\ H &= \frac{L_{edge} * EndEdgeCh - H_{edge} * StartEdgeCh}{EndEdgeCh - StartEdgeCh} + \\ &+ EndCh * \frac{H_{edge} - L_{edge}}{EndEdgeCh - StartEdgeCh} \end{aligned} \quad (\text{A7.5})$$

7. Continuum Compton counts  $K$  associated to the selected ROI is simply the area of a trapezoid and is given by:

$$K = (EndCh - StartCh + 1) \frac{H + L}{2} \quad (\text{A7.6})$$

Its standard uncertainty is computed from Poisson distribution,  $u(K) = \sqrt{K}$ .

8. Compton counts rate associated to the selected ROI is:

$$K_{rate} = K / t \quad (\text{A7.7})$$

9. Gross counts associated to the selected ROI is:

$$G = \sum_{i=StartCh}^{EndCh} G(i) \quad (\text{A7.8})$$

having a standard uncertainty computed from Poisson distribution.

10. Gross count rate is Gross counts divided by spectrum live-time.

11. Ambient background counts associated to the selected ROI is:

$$B = \sum_{i=StartCh}^{EndCh} B(i) \frac{t}{t_B} \quad (\text{A7.9})$$

where  $B(i)$  are the ambient counts in channel  $i$  in background spectrum. Its standard uncertainty is computed from Poisson distribution.

12. Ambient background counts rate is given by time corrected ambient background counts  $B$  divided by spectrum live-time  $t$ .

The Compton background counts in channel  $i$ ,  $K(i)$ , of the selected ROI is given by an equation similar to any equations from A7.5, replacing for instance  $H$  with  $K(i)$  and  $EndCh$  with  $i$ . The net counts in channel  $i$ ,  $N(i)$  is therefore given by:

$$N(i) = G(i) - K(i) - B(i) \frac{t}{t_B} \quad (\text{A7.10})$$

Its standard uncertainty is given by propagation of uncertainties using Poisson distribution.

13. Peak pulses, Peak channel and its corresponding Peak energy is related to the maximum value of  $N(i)$  and its corresponding channel and energy.

14. Net area computed from channel by channel subtraction is given by:

$$N = G - B - K \quad (\text{A7.11})$$

having its standard uncertainty computed by propagation of uncertainty (see appendix 2).

Net area can also be estimated using Gaussian fit method. The peak is approximated with a Gaussian function, therefore the Net counts in channel  $i$  are given by:

$$N_i = N_{\max} \exp\left(-\frac{(E_i - \mu)^2}{2\sigma^2}\right) \quad (\text{A7.12})$$

where  $N_{\max}$  are the maximum number of counts in gaussian distribution,  $\mu$  is its energy (or channel) mean corresponding in which  $N_{\max}$  counts are recorded,  $\sigma$  is function standard deviation. ROI data, i.e. the net counts  $N(i)$  from (A7.10), are modeled using the Marquardt-Levenberg algorithm and these 3 parameters are computed [Press et. all].

A general Gauss function is given by:

$$P(k) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\frac{(k - \mu)^2}{2\sigma^2}\right) \quad (\text{A7.13})$$

$$\int_{-\infty}^{\infty} P(x) dx = 1$$

By comparing A7.12 with A7.13 and integrating from  $-\infty$  to  $\infty$ , it can be shown that the ROI net area is:

$$N = N_{\max} \sigma \sqrt{2\pi} \quad (\text{A7.14})$$

Its uncertainty is given by Poisson distribution:  $\sqrt{N_{\max}} \sigma \sqrt{2\pi}$ .

Also, from (A7.13) let  $k = \mu + \lambda\sigma$  for which the probability is reduced at half, then:

$$P(k) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\frac{(\mu - \mu - \lambda\sigma)^2}{2\sigma^2}\right) = P_{\max} \exp\left(-\frac{\lambda^2}{2}\right) = \frac{1}{2} P_{\max}; P_{\max} = \frac{1}{\sqrt{2\pi\sigma^2}}$$

It follows that  $\lambda^2 = -2 \ln \frac{1}{2}$  and therefore:

$$FWHM = \mu + \lambda\sigma - (\mu - \lambda\sigma) = 2\lambda\sigma \text{ or:}$$

$$FWHM = 2\sqrt{-2 \ln\left(\frac{1}{2}\right)}\sigma \approx 2.355\sigma \quad (\text{A7.15})$$

15. FWHM in channels or keV is the full width at half maximum.

In Gaussian fit mode, FWHM is computed from equation (A7.15). Its uncertainty is evaluated to 2 channels. This evaluation is based on initial guess of parameters required by Marquardt-Levenberg algorithm.

In default mode, FWHM is computed by performing a channel by channel scan in ROI in order to find the channels  $C_1$  and  $C_2$  in which the corresponding counts are about half of peak counts. Then:

$$FWHM = C_2 - C_1 \quad (\text{A7.16})$$

Its uncertainty is evaluated to 2 channels (for estimation of each  $C_1$  and  $C_2$ ).

In Ge computation mode, FWHM is computed as follows:

It is assumed that the shape of the peak is a Gaussian function. Let 3 points on that function. One can use A7.12 for each points. By division we have:

$$\begin{aligned} \frac{N_1}{N_3} &= \exp\left[\frac{(E_3 - E_0)^2}{2\sigma^2} - \frac{(E_1 - E_0)^2}{2\sigma^2}\right] = \exp\left[\frac{E_3^2 - E_1^2 - 2E_3E_0 + 2E_1E_0}{2\sigma^2}\right] \\ 2\sigma^2 \ln \frac{N_1}{N_3} &= E_3^2 - E_1^2 - 2E_0(E_3 - E_1); \Rightarrow E_0 = \frac{E_3^2 - E_1^2 - 2\sigma^2 \ln \frac{N_1}{N_3}}{2(E_3 - E_1)} \end{aligned}$$

But  $N_{\max} = N_1 \exp\left[\frac{(E_1 - E_0)^2}{2\sigma^2}\right]$  and we have:

$$N_2 = N_1 \exp\left[\frac{(E_1 - E_0)^2}{2\sigma^2}\right] \exp\left[-\frac{(E_2 - E_0)^2}{2\sigma^2}\right] = N_1 \exp\left[\frac{E_1^2 - E_2^2 - 2E_1E_0 + 2E_2E_0}{2\sigma^2}\right]$$

$$2\sigma^2 \ln \frac{N_2}{N_1} = E_1^2 - E_2^2 + 2E_0(E_2 - E_1)$$

$$2\sigma^2 \ln \frac{N_2}{N_1} = E_1^2 - E_2^2 + 2(E_2 - E_1) \frac{E_3^2 - E_1^2 - 2\sigma^2 \ln \frac{N_1}{N_3}}{2(E_3 - E_1)}$$

Further manipulation gives:

$$\sigma = \sqrt{\frac{(E_2 - E_1)(E_3 - E_2)}{2\left[\ln \frac{N_2}{N_1} + \frac{E_2 - E_1}{E_3 - E_1} \ln \frac{N_1}{N_3}\right]}}$$

or:

$$\sigma = \sqrt{\frac{(E_2 - E_1)(E_3 - E_2)}{2\left[\ln \frac{N_2}{N_1}\right]}}; N_1 = N_3 \quad (\text{A7.17})$$

Using the symmetry property of Gauss function,  $|E_2 - E_1| = |E_3 - E_2|$ .  $N_2$  is chosen to be the maximum net counts in peak channel and the search is done for the closest channel (closest to peak channel) in which there are  $N_1$  counts. Then FWHM is estimated using A7.15.

Its uncertainty is evaluated to 1 channels (for estimation of  $C_1$  associated to  $E_1$ ).

16. FWHM taken from calibration (see appendix 9)

17. Resolution associated to FWHM from both ROI and calibration and their comparison result.

$$\text{Resolution} = 100 \frac{\text{FWHM(keV)}}{\text{Centroid(keV)}} \quad (\text{A7.18})$$

The comparison is made by using one-tailed t-test (see appendix 2).

18. Nuclide information based on ROI set such as: its name, atomic mass, energy yield (probability of emission)

19. Efficiency taken from calibration (see appendix 9)

Let  $Y$  the energy yield and  $\epsilon$  the efficiency associated to selected ROI.

In case of multiple energy lines of the same nuclide in ROI, it is computed the weight:

$$w_i = \frac{Y_i}{\sum_i Y_i}; Y = \sum_i Y_i \quad (\text{A7.19})$$

Let  $\varepsilon_i$  and  $s_i$  the efficiency associated to energy line  $i$  (efficiency is taken from calibration curve, energy line and yield is taken from in-use library) and its corresponding standard uncertainty. The efficiency associated to the selected ROI and its uncertainty is given by:

$$\varepsilon = \frac{\sum_i w_i \varepsilon_i}{\sum_i w_i}; s^2 = \frac{\sum_i w_i^2 s_i^2}{[\sum_i w_i]^2} \quad (\text{A7.20})$$

#### 20. Activity associated to the selected ROI

If efficiency is normalized to 100 photons, i.e. is given in %, then we have the activity associated to selected ROI given by:

$$A = 100 \frac{N}{Y\varepsilon t} \quad (\text{A7.21})$$

where  $t$  is the sample live-time and  $N$  is the net area computed above.

#### 21. Detection limit and its corresponding MDA (see appendix 4)

$$MDA = 100(2.706 + 3.29\sqrt{K})/(tY\varepsilon) \quad (\text{A7.22})$$

where  $K$  is the continuum (Compton) background counts.

22. Comparison between activity and MDA to see if there is an activity in sample or not using one-tail t-test (see appendix 2).

If we have multiple ROIs and from each ROI it is computed the sample activity, then the final nuclide activity is evaluated using the weighted mean formalism.

$$A_i = 100 \frac{N_i}{tY_i\varepsilon_i} \quad (\text{A7.23})$$

where  $N_i$  is the net area of ROI  $i$ ,  $\gamma_i$  and  $\varepsilon_i$  is the peak emission probability and peak efficiency respectively.

The final activity and its uncertainty is given by:

$$A = \frac{\sum_i w_i A_i}{\sum_i w_i}; w_i = A_i / S(A_i)$$

$$S^2(A) = \frac{\sum_i w_i^2 A_i^2}{[\sum_i w_i]^2}$$
(A7.24)

## Appendix 8 Nuclide corrections

### Decay correction

In general case, nuclides are part of radioactive chain, e.g. 238U series, 232Th series. Gamma spectroscopy allows retrieving the parent activity based on net area computation of a ROI around a peak.

$$A_{parent} = \frac{NetArea}{tEY} \quad (A8.1)$$

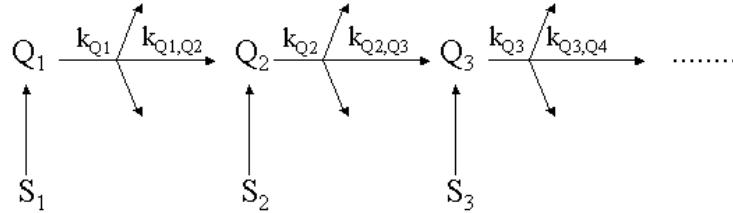
where  $t$  is the spectrum live-time,  $E$  is the peak efficiency taken from calibration curve or computed by Monte-Carlo technique,  $Y$  is the radiation emission probability. Radiation yield is computed by:

$$Y = B_R G \quad (A8.2)$$

where  $B_R$  is the branching ration for daughter nuclide and  $G$  is the gamma emission probability of a particular gamma line in daughter spectrum.

Therefore it is computed the parent activity as being  $A_{parent} = A_{daughter} / B_R$  which is true only if secular equilibrium between parent and daughter (daughters) is reached.

Nuclides decay is a general linear first order process:



where  $Q_i$  are the amounts of species  $i$  present at time  $t$ ,  $k_{Qi}$  is the total removal constant for the species  $Q_i$ ,  $k_{Qi,Qi+1}$  is the partial removal constant leading to the production of species  $Q_{i+1}$ , etc, and  $k_{Qi,Qi+1} = BR_{Qi,Qi+1} \cdot k_{Qi,Qi+1}$  where  $BR_{Qi,Qi+1}$  is the branching ratio. The source terms  $S_i$  are the constant independent rates of production of the  $i_{th}$  species and in most cases are zero.

The differential equations governing the main “chain” shown above are given by:

$$\begin{aligned} dQ_1/dt &= S_1 - k_{Q1} \cdot Q_1 \\ dQ_2/dt &= S_2 + k_{Q1,Q2} \cdot Q_1 - k_{Q2} \cdot Q_2 \end{aligned}$$

$$dQ_i/dt = S_i + k_{Qi-1,Qi} \cdot Q_{i-1} - k_{Qi} \cdot Q_i$$

$$dQ_n/dt = S_n + k_{Qn-1,Qn} \cdot Q_{n-1} - k_{Qn} \cdot Q_n$$

The solution to these equations can be expressed in the form [Bateman, Magill, Skrable]:

$$Q_n(t) = \sum_{i=1}^{i=n} \left[ \left( \prod_{j=1}^{j=n-1} k_{j,j+1} \right) \times \sum_{j=i}^{j=n} \left( \frac{Q_i(0)e^{-k_j t}}{\prod_{\substack{p=i \\ p \neq j}}^n (k_p - k_j)} + \frac{S_i(1 - e^{-k_j t})}{k_j \prod_{\substack{p=i \\ p \neq j}}^n (k_p - k_j)} \right) \right] \quad (\text{A8.3})$$

where  $Q_i(0)$  is amount of the  $i^{\text{th}}$  species at time  $t=0$ . The  $k_n$  are the total removal constant for species  $n$  ( $k = \ln(2)/t_{1/2} = 0.69315/t_{1/2}$ ),  $k_{n,n+1}$  is the partial decay constant (partial removal constant) and is related to the branching ratio  $BR_{n,n+1}$  through the relation  $k_{n,n+1} = BR_{n,n+1} k_n$ . Clearly, from equation (A8.5) the total removal rate constants must be distinctly different, otherwise the terms in the denominator become indeterminate. For radioactive transformations, this is generally the case. The equation A8.3 is known as the Bateman law.

In general case, the secular equilibrium can never be reached so a decay correction is needed. The options are:

1. Secular equilibrium is admitted => no correction is made
2. Correction based on sample time which is the time elapsed from source creation when only parent nuclide was present => computations are based on true Bateman decay law.
3. Sample time is not known but we are sure there is an equilibrium and uncertain if it is secular equilibrium or not. In this case, we assume that the sample-time is  $10 \times$  parent half-life, time sufficiently long for any equilibrium to be reached. Computations are made based on Bateman decay law.

Let parent activity being 1. Then using Bateman law we have daughter activity being  $f$  instead of  $B_R$ . Therefore,

$$A_{\text{parent}} = A_{\text{daughter}} / f = A_{\text{daughter}} / B_R / (f / B_R) \quad (\text{A8.4})$$

So, the correction is made by dividing the “regular” activity (or net area) by a correction factor:

$$\text{Corr} = f / B_R \quad (\text{A8.5})$$

#### Coincidence correction

The gamma-gamma and/or gamma - X-ray true coincidence correction is of main concern since alpha efficiency and beta efficiency are basically zero for gamma detectors. The annihilation quanta (from beta plus decay) has negligible effect in most cases. Based on a quite accurate database (.idt files), the true coincidence is computed for any energy from any daughter nuclide in the radioactive chain [Sudar].

Below, it is presented a simplified version of the true coincidence correction theory:

Suppose we have a real gamma cascade, e.g. 1173 keV and 1332 keV gamma lines from  $^{60}\text{Co}$  nuclide. Let cascade probability being 1, then the true coincidence probability is:

$$p = \gamma_1 \varepsilon_1 \gamma_2 \varepsilon_2 \quad (\text{A8.6})$$

where  $\gamma_1, \gamma_2$  are the emission probabilities for the gamma energy lines (branching ratio included),  $\varepsilon_1, \varepsilon_2$  are the corresponding peak efficiencies.

The net area counts rate for both peaks are:

$$n_1 = A\gamma_1 \varepsilon_1; n_2 = A\gamma_2 \varepsilon_2 \quad (\text{A8.7})$$

where A is the nuclide activity.

The sum-peak is formed and its net area rate is given by:

$$n_{sum} = Ap = A\gamma_1 \varepsilon_1 \gamma_2 \varepsilon_2 \quad (\text{A8.8})$$

In general, if the sum peak presents an emission probability by itself  $\gamma_3$  then the observed counts will be:

$$\begin{aligned} n &= A\gamma_3 \varepsilon_3 \\ n_o &= n + n_{sum} = n(1 + \gamma_1 \varepsilon_1 \gamma_2 \varepsilon_2 / (\gamma_3 \varepsilon_3)) = n(1 + P_{in}) \end{aligned} \quad (\text{A8.9})$$

Due to true coincidence, a certain number of counts are loss in both peaks. The loss depends on number of photons of energy  $E_1$  emitted and totally absorbed  $A\gamma_1 \varepsilon_1$ , the cascade probability (emission of photon 2 right after photon 1, usually being 1 or 100%) and the probability of absorption of photon 2 either totally or partially  $\varepsilon_2^T$ . We have:

$$\dot{n}_1 = A\gamma_1 \varepsilon_1 \varepsilon_2^T = n_1 \varepsilon_2^T \quad (\text{A8.9})$$

The observed counts in a particular peak are:

$$n_{10} = n_1 - \dot{n}_1 = n_1(1 - \varepsilon_2^T) = n_1(1 - P_{out}) \quad (\text{A8.10})$$

Therefore, the correction is made by dividing the “observed” activity (or net area) by a correction factor:

$$Coin\_corr = (1 + P_{in})(1 - P_{out}) \quad (\text{A8.11})$$

### Measurement time correction

During the sample measurement, a particular nuclide from that sample decays, therefore let  $\tau$  be the measurement time (e.g. real time for gamma measurement) then we have:

$$A = A_0 \exp(-\lambda\tau); \lambda = \ln(2)/T_{1/2} \quad (\text{A8.12})$$

where  $A_0$  is the activity at the begining of measurements and  $T_{1/2}$  is the nuclide half-life.

Computing activity  $A$  means the calculation of an average activity during time  $\tau$ . The average (mean) activity is given by:

$$\bar{A} = \frac{\int_0^\tau A_0 \exp(-\lambda t) dt}{\int_0^\tau dt} = \frac{A_0}{\tau} \left| -\frac{1}{\lambda} \exp(-\lambda t) \right|_0^\tau = \frac{A_0}{\lambda\tau} [1 - \exp(-\lambda\tau)]$$

And we have:

$$C = \frac{\bar{A}}{A} = \frac{\lambda\tau}{1 - \exp(-\lambda\tau)} \quad (\text{A8.13})$$

The sample activity at the begining of measurement is finaly computed by multiplying the calculated activity with the correction factor from equation A8.13.

## **Appendix 9.**

### **Gamma calibrations**

#### Energy/FWHM calibration

The energy calibration involves the computation of coefficients for a channel to energy function approximated by a n-order polynome (usually of order 1, 2 or 3).

$$f(x) = \sum_{i=0}^n c_i x^i; n = 1, 2, 3 \quad (\text{A9.13})$$

where  $f$  is the energy depending of channel  $x$  and  $c_i$  are the coefficients.

The coefficients are evaluated using the general least squares numerical method [Press et. all].

Same method and function is used for FWHM calibration.

#### Efficiency calibration

It simply means the computation of an energy to efficiency function. One of the best ways to accomplish that is to consider natural logarithm of energy and efficiency, to choose a crossover energy  $E_c$  and let two polynomes at left and right side of the crossover point [Silena].

$$\begin{aligned} \ln(\text{eff}(x)) &= \sum_{i=0}^n a_i x^i; x = \ln(E); n = 1, 2, 3, 4; E < E_c \\ \ln(\text{eff}(x)) &= \sum_{i=0}^m b_i x^i; x = \ln(E); m = 1, 2, 3, 4; E \geq E_c \end{aligned} \quad (\text{A9.14})$$

The coefficients are evaluated using the general least squares numerical method [Press et. all] and the actual efficiency is evaluated using:

$$\begin{aligned} \varepsilon &= \exp[a_0 + a_1 \ln(E) + \dots + a_n \ln(E)]; E < E_c \\ \varepsilon &= \exp[b_0 + b_1 \ln(E) + \dots + b_m \ln(E)]; E \geq E_c \end{aligned} \quad (\text{A9.15})$$

## Appendix 10

### Monte Carlo algorithm for alpha detector efficiency estimation

Let a coordinate system having its origin at detector window center (or source center). Let  $a$  the source radius,  $b$  the detector radius and  $d$  the source to detector distance.

The evaluation of (x,y) point for radiation emission is done by the following steps[Sima]:

- Let  $r$  a random generated number in  $[0,1]$ . The distance from origin to the point of emission for a disk shape is given by:

$$\rho = a\sqrt{r} \quad (\text{A10.1})$$

- Generate another random number  $r$  and compute the angle:

$$\psi = 2\pi r \quad (\text{A10.2})$$

- Compute x,y coordinate for the emission point as being:

$$\begin{aligned} x &= \rho \cos \psi \\ y &= \rho \sin \psi \end{aligned} \quad (\text{A10.3})$$

Evaluation of weight parameter associated to each particle is necessary for variance reduction, i.e. the particles are generated inside a solid angle chosen in such way that encompass the detector rather than  $4\pi$  (any possible direction). First, let  $D$  be a

distance parameter equal either to source diameter if source diameter is larger than detector diameter or to detector diameter otherwise.

$$D = \begin{cases} 2a, & a > b \\ 2b, & b \geq a \end{cases} \quad (\text{A10.4})$$

The maximum value for the cosine value of the solid angle inside each radiation is emitted is:

$$\cos theta_{\max} = \frac{d}{\sqrt{d^2 + D^2}} \quad (\text{A10.5})$$

The maximum solid angle and the weight of each particles are:

$$\begin{aligned} \Omega_{\max} &= 2\pi(1 - \cos theta_{\max}) \\ w &= \frac{\Omega_{\max}}{4\pi} \end{aligned} \quad (\text{A10.6})$$

Polar angle evaluation is done by the following:

- Let S be normalized maximum solid angle :

$$S = \frac{1 - \cos theta_{\max}}{2} = w \quad (\text{A10.7})$$

- Generate a random number r and compute the cosine and sine of polar angle:

$$\begin{aligned} \cos theta &= 1 - 2rS \\ \sin theta &= \sqrt{1 - \cos theta^2} \end{aligned} \quad (\text{A10.8})$$

The particle traversed distance L is computed by:

$$L = \frac{d}{\cos theta} \quad (\text{A10.9})$$

Azimuth angle evaluation is done by:

- Generate a random number r and compute the angle:

$$\varphi = 2\pi r \quad (\text{A10.10})$$

Initial directional cosine are given by:

$$\begin{aligned} u &= \sin theta * \cos(\varphi) \\ v &= \sin theta * \sin(\varphi) \end{aligned} \quad (\text{A10.11})$$

Next, the particle transport is performed, i.e. its coordinates are updated by:

$$\begin{aligned} X &= x + Lu \\ Y &= y + Lv \end{aligned} \tag{A10.12}$$

And it is tested if hits the detector (scored) or not (missed).

$$X^2 + Y^2 \leq b^2 \quad (\text{A10.13})$$

If the above condition is satisfied then the scoring variable n and its square value  $n_2$  is updated:

$$\begin{aligned} n &= n + w \\ n_2 &= n_2 + w^2 \end{aligned} \quad (\text{A10.14})$$

Note that the scoring variable represents the sum of weights of all particles that reach the detector window.

Let  $N$  be the total number of particles emitted (histories). Then the efficiency and its standard uncertainty (standard deviation of mean) are evaluated to be (see A2.2' and A2.3):

$$\varepsilon = n/N$$

$$u^2 = \frac{n_2 - \frac{1}{N}n^2}{N(N-1)} \quad (\text{A10.15})$$

## **Appendix 11 Nuclide interference correction**

If multiple nuclides are presented in sample then it is possible to have more counts in peak region (ROI) due to nuclide interferences. As an example, the sample having  $^{238}\text{U}$ , $^{232}\text{Th}$  and  $^{40}\text{K}$  nuclides presents interference at 1460keV peak region associated to  $^{40}\text{K}$  nuclide.

Let  $N_i$ ,  $i = 1, 2, \dots, n$  the observed counts associated to ROI around the peak  $i$ . Let  $N_{ij}$  the number of counts in band (ROI)  $i$  due to nuclide  $j$ .

We can write :

$$\begin{aligned} N_{11} + N_{12} + \dots + N_{1n} &= N_1 \\ N_{21} + N_{22} + \dots + N_{2n} &= N_2 \\ \dots \\ N_{n1} + N_{n2} + \dots + N_{nn} &= N_n \end{aligned} \quad (\text{A11.1})$$

From standard source spectrum, one can easily compute the ratios:

$$a_{ij} = \frac{N_{ij}}{N_{jj}} \quad (\text{A11.2})$$

Inserting A11.2 into A11.1 we obtain an equation system of variables  $N_{ii}$  (nuclide of interest in its own band as function of observed counts and the pre-computed ratios  $a_{ij}$ ). This can be solved using various numerical methods such as Gauss, Gauss-Jordan, LU decomposition [Press et. all].