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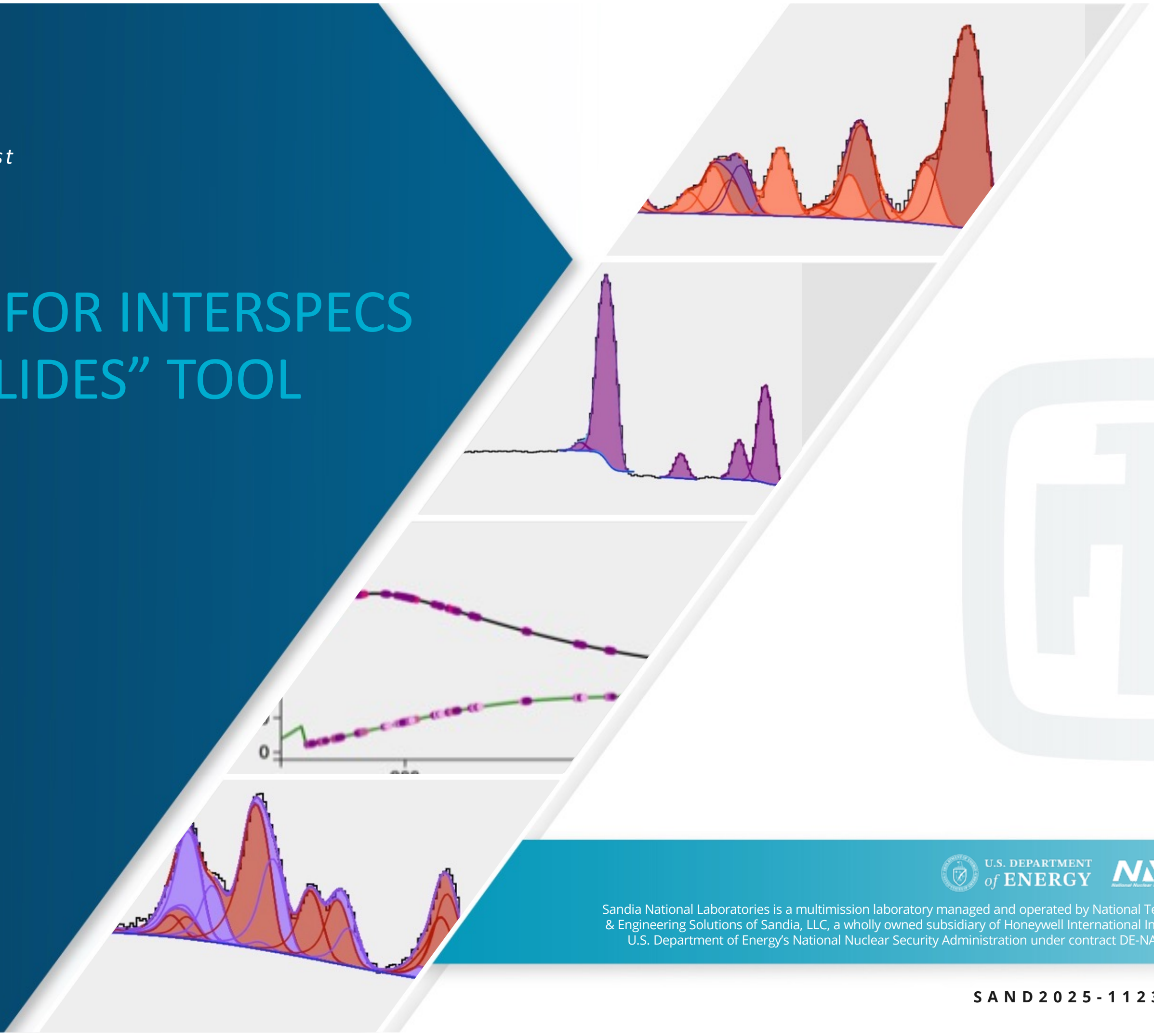
USE INSTRUCTIONS FOR INTERSPECS "ISOTOPICS BY NUCLIDES" TOOL

As of 20250901



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Org 08647

20250901 – support: InterSpec@sandia.gov

SAND2025-11238PE

DISCLAIMER

- These instructions were created using the version of InterSpec available from the “bleeding edge” automated build, as of 20250920, at
  https://github.com/sandialabs/InterSpec/releases/tag/bleeding-edge
  but are expected to be fully applicable to InterSpec v1.0.14
- It is likely there will be some bug fixes and improvements over the next couple months

- Questions, requests, support and feedback at InterSpec@sandia.gov

INTRODUCTION

Presets: Modified HPGe Pu 614 to 668 keV

χ²/dof = 398/0, 84.3% (2σ: 83.34%, 85.27%) Pu239

☑ Fit Energy Cal. ☐ Back. Sub. FWHM Est.: Start from Det/Peaks FWHM Form: sqrt(A0 + A1*E) Peak Skew: GaussExp Add. Uncert: None

Eqn Type: Physical Pu242 corr: By Pu239 Frac. Detector: ORTEC Detective-EX1... ☑ Use Corr. Fcn.

Self Attenuation: Material: Pu (plutonium) Thick.: 2.02901 cm ☑ Fit

External Attenuation: Material: Fe (iron) Thick.: 3.05433 cm ☑ Fit

Nuclide: Pu239 Age: 20.00 y ☐ Fit Age Rel. Act=2.482E9, MassFrac(Pu)=84.3%

Nuclide: Pu240 Age: 20.00 y ☐ Fit Age Rel. Act=1.691E9, MassFrac(Pu)=15.7%

Nuclide: Cs137 Rel. Act=0

☐ Same El. Same Age

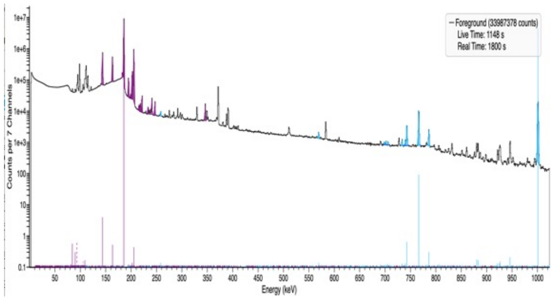
Lower Energy: 614 keV Continuum Type: Cubic

Upper Energy: 671 keV ☑ Force full-range

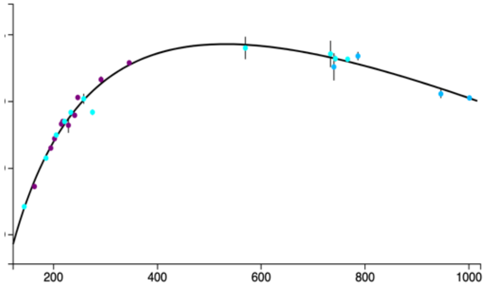
- The “Isotopics by nuclides” tool in InterSpec performs “Relative Efficiency” analysis to tell you amount of each nuclide/source present, relative to each other, as well as possibly determine ages or shielding
  - Relative Efficiency is the technique used by, for example, MGA[†] and FRAM[‡] to determine uranium or plutonium enrichment – but otherwise the techniques use has seemingly been limited
- The tool allows using all gamma or x-ray producing nuclides, fluorescent x-rays, and the nuclear reactions within InterSpec, to quickly and intuitively perform Relative Efficiency analysis – leading to new and powerful analysis capabilities for a number of scenarios
  - U/Pu enrichment, ratio of iodines in an environmental sample, fitting ratio of activities when detector efficiency or sample geometry isn’t known, fitting peaks that overlap or for nuclides that interfere with each other

[†] Buckley, W, Clark, D, Parker, W E, Romine, W, Ruhter, W, & Wang, T F (1999). Plutonium and uranium isotopic analysis: recent developments of the MGA++ code suite.
[‡] Vo, Duc T., & Sampson, Thomas E. (2020). FRAM, Version 6.1 User Manual. https://doi.org/10.2172/1599022

RELATIVE EFFICIENCY ANALYSIS INTRODUCTION



$$C * \frac{Peak\ Area}{Act. \times B.R.}$$



- Relative Efficiency Analysis finds the relative-activities for the nuclides in a spectrum, so that (peak area) / (activity × branching ratio) makes a smooth curve, as a function of energy
- It gives you the nuclide activities/masses, relative to one another – does not give absolute activities/masses
- *Does not* require knowing a detectors efficiency, the distance, shielding, or geometry
- *Does* require that all nuclides and shielding are homogeneous, or the use of multiple relative efficiency curves, and sufficient statics/power in the data to dis-entangle them.

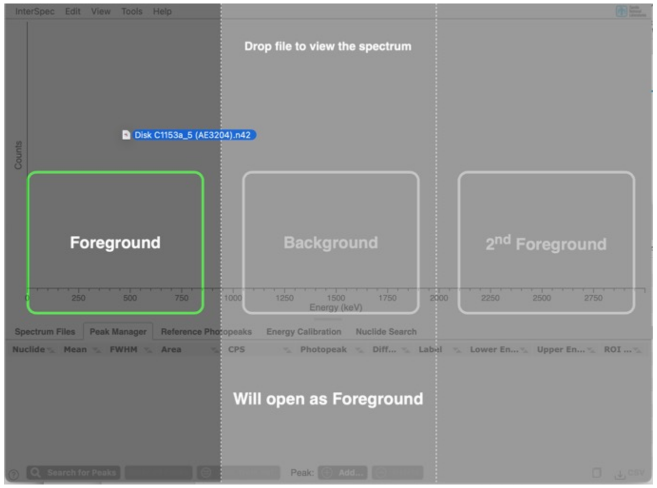
RELATIVE EFFICIENCY REFERENCES

- *Relative Efficiency Curves Demystified*, by Michael Enghauser, SNL, July 2016
  https://www.osti.gov/servlets/purl/1399186
  A great and concise introduction, especially for Uranium enrichment
- *FRMAC Gamma Spectroscopist Knowledge Guide*, by Michael Enghauser, Aug 2019
  https://www.osti.gov/biblio/1763003
  An all-around great gamma spectroscopy manual; see Section 14 for Relative Efficiency
- *Application Guide to Gamma-Ray Isotopic Analysis Using the FRAM Software*, by T. Sampson, T. Kelley, and D. Vo, LANL, Sep 2003
  https://cdn.lanl.gov/files/app-to-isotopic-analysis-using-fram_06e9e.pdf
  FRAM is considered by many to be *the* software for U and Pu enrichment analysis from gamma spectra
- *Peak-Based Relative Efficiency analysis in InterSpec*, W. Johnson, SNL, Aug 2022,
  https://sandialabs.github.io/InterSpec/tutorials/#relative-efficiency-analysis
  Focusses on U-enrichment using manually fit peaks

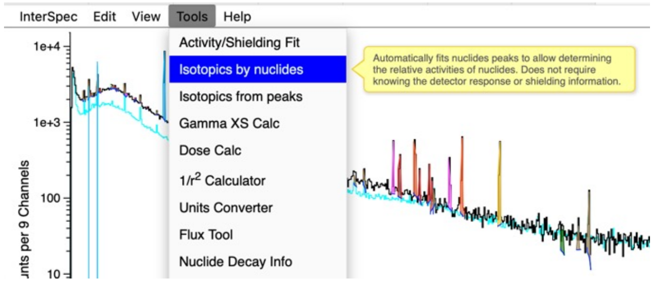
INTRODUCTION TO PERFORMING AN ANALYSIS

Only the more-important or common options will be covered, and will assume some familiarity with InterSpec

INTRODUCTION TO PERFORMING AN ANALYSIS



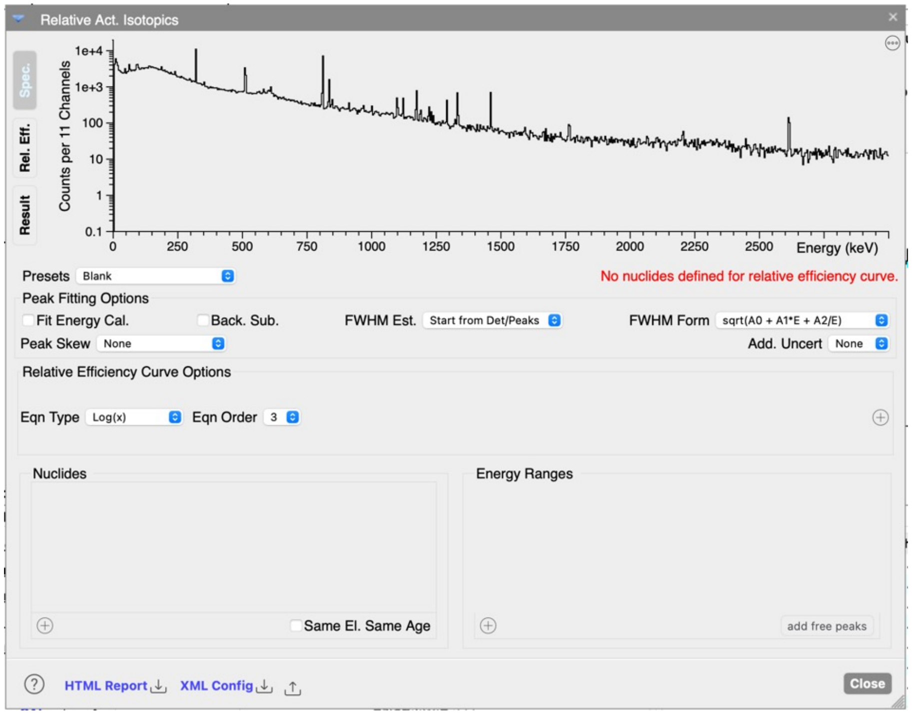
To perform an analysis, first you will load the spectrum of interest as foreground in InterSpec, as well as load a background, if you plan to use it (you may not need to).



Automatically fits nuclides peaks to allow determining the relative activities of nuclides. Does not require knowing the detector response or shielding information.

Then under the “Tools” menu, select “Isotopics by nuclides”

INTRODUCTION TO PERFORMING AN ANALYSIS



Presets: Blank

No nuclides defined for relative efficiency curve.

☐ Fit Energy Cal. ☐ Back. Sub. FWHM Est.: Start from Det/Peaks FWHM Form: sqrt(A0 + A1*E + A2/E)

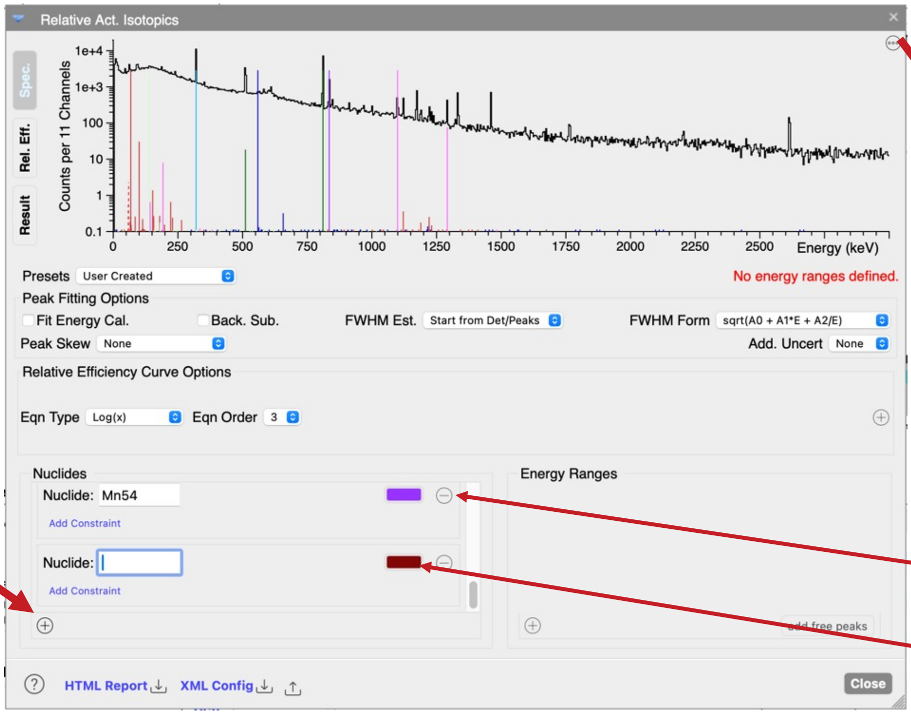
Peak Skew: None Add. Uncert: None

Eqn Type: Log(x) Eqn Order: 3

☐ Same El. Same Age

Once in the tool, you will need to tell it which nuclides/sources you are interested in, what energy ranges to use, as well as other options to apply.

INTRODUCTION TO PERFORMING AN ANALYSIS – ADDING SOURCES



Presets User Created

No energy ranges defined.

Peak Fitting Options

☐ Fit Energy Cal. ☐ Back. Sub. FWHM Est. Start from Det/Peaks FWHM Form sqrt(A0 + A1*E + A2/E)

Peak Skew None Add. Uncert None

Relative Efficiency Curve Options

Eqn Type Log(x) Eqn Order 3

Nuclides

Nuclide: Mn54

Add Constraint

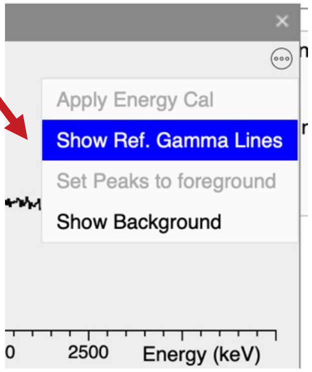
Nuclide:

Add Constraint

Energy Ranges

add free peaks

You can show the source lines on the spectrum from upper-right hand



Click the "+" icon to add nuclides/sources

Click the "-" icon to remove a source

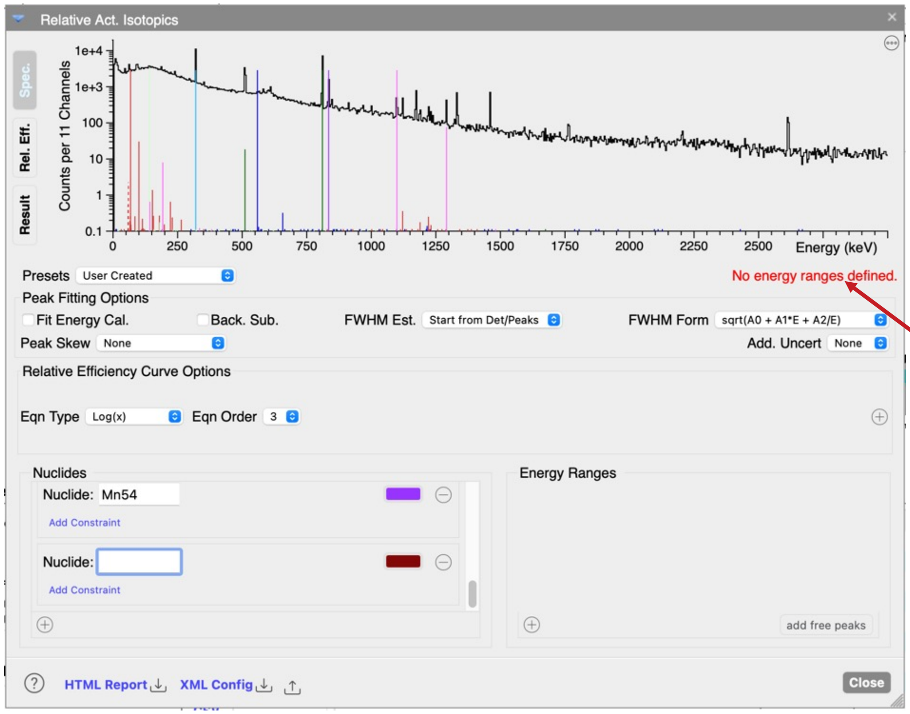
Click the colored square to change the sources color

After adding a source, you can enter any nuclide, (ex. Co60, U235), fluorescent x-ray (ex. W, U), or reaction (ex. Fe(n,g), Be(a,n)).

If aging the source makes sense, you will be given the option to enter an age, or fit for it.

Note: like other places in InterSpec, you only enter parent nuclide (ex. U238), not descendants (ex. Pa234m)

INTRODUCTION TO PERFORMING AN ANALYSIS – ADDING ROIS



Presets: User Created

No energy ranges defined.

Peak Fitting Options

☐ Fit Energy Cal.
☐ Back. Sub.
FWHM Est.: Start from Det/Peaks
FWHM Form: sqrt(A0 + A1*E + A2/E)
Peak Skew: None
Add. Uncert: None

Relative Efficiency Curve Options

Eqn Type: Log(x)
Eqn Order: 3

Nuclides

Nuclide: Mn54
Add Constraint

Nuclide:
Add Constraint

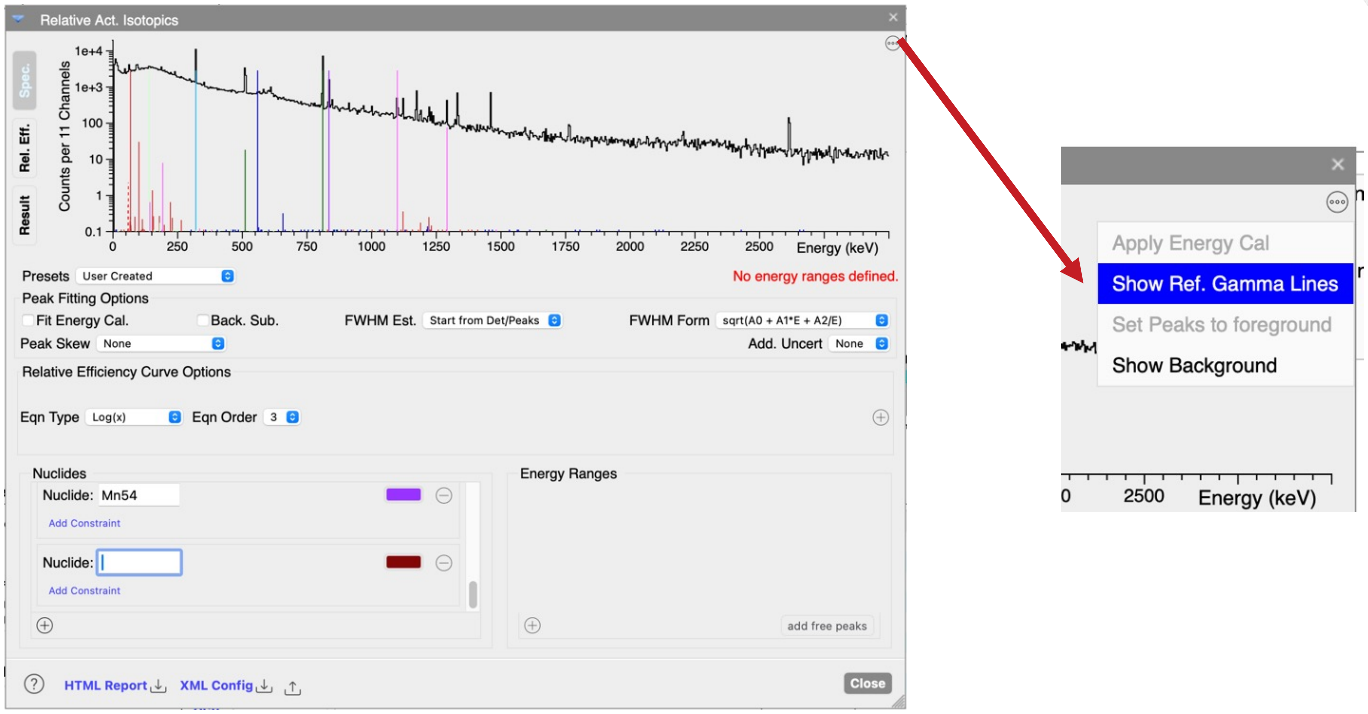
Energy Ranges

add free peaks

Error messages displayed here

You will also need to add “Region of Interest” energy ranges to use in the analysis – this can be done in a number of ways shown on the following slides.
You can add any number of ROIs, but they CAN NOT overlap, or analysis will fail

INTRODUCTION TO PERFORMING AN ANALYSIS – ADDING ROIS (CONT)



Presets User Created

No energy ranges defined.

Peak Fitting Options

☐ Fit Energy Cal. ☐ Back. Sub. FWHM Est. Start from Det/Peaks FWHM Form sqrt(A0 + A1*E + A2/E)

Peak Skew None Add. Uncert None

Relative Efficiency Curve Options

Eqn Type Log(x) Eqn Order 3

Nuclides

Nuclide: Mn54

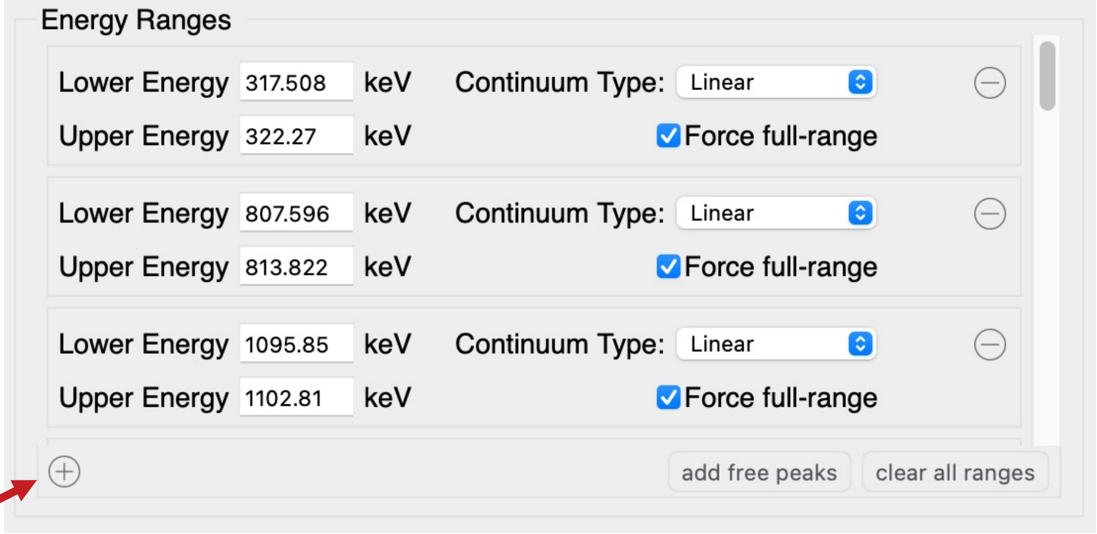
Nuclide:

Energy Ranges

You can show the reference lines for the nuclides/sources you have added, using the upper-right-hand menu, and selecting “Show Ref. Gamma Lines” - this will help to add relevant ROIs.

Putting your mouse over the reference lines on the spectrum will give additional information

INTRODUCTION TO PERFORMING AN ANALYSIS – ADDING ROIS (CONT)



Energy Ranges

Lower Energy 317.508 keV Continuum Type: Linear

Upper Energy 322.27 keV ☑ Force full-range

Lower Energy 807.596 keV Continuum Type: Linear

Upper Energy 813.822 keV ☑ Force full-range

Lower Energy 1095.85 keV Continuum Type: Linear

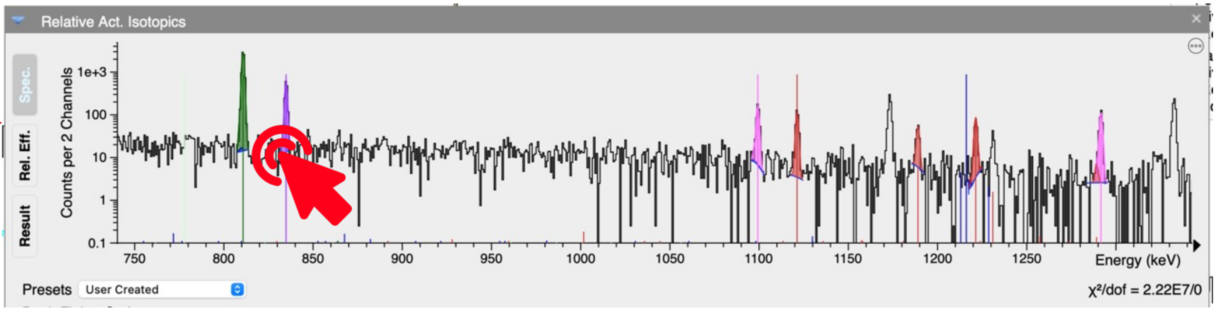
Upper Energy 1102.81 keV ☑ Force full-range

You can click the “+” button in the “Energy Ranges” area to add a new range, and then manually type in the energy extent of a peak, or even the range of a few peaks if they should be fit together (e.g., overlap)

This method is cumbersome and slow – so not recommended!

INTRODUCTION TO PERFORMING AN ANALYSIS – ADDING ROIS (CONT)

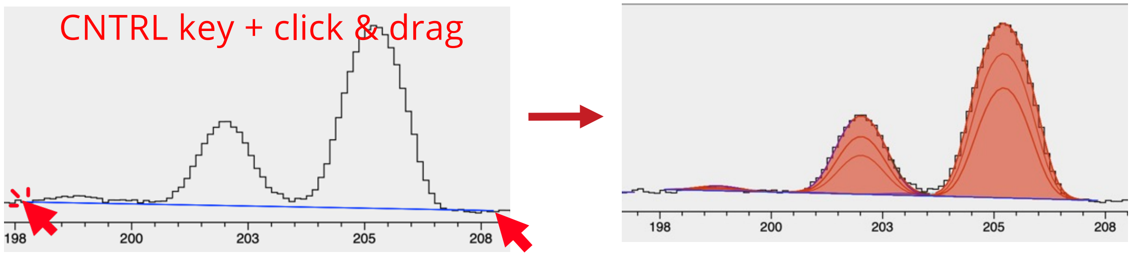
Instead, you can double-click on the spectrum to create a ROI at that energy



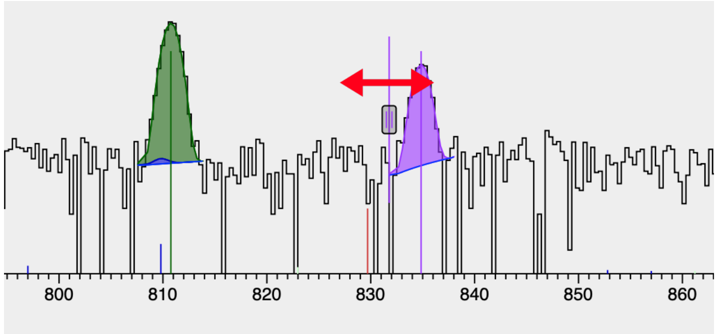
Presets User Created

χ²/dof = 2.22E7/0

Or you can hold down the CNTRL key and click+drag the mouse to define a new ROI

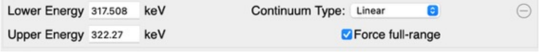


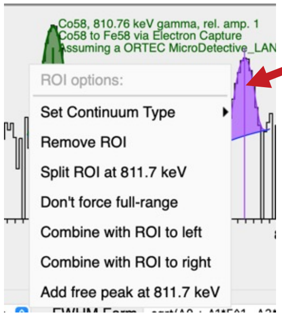
INTRODUCTION TO PERFORMING AN ANALYSIS – ADDING ROIS (CONT)



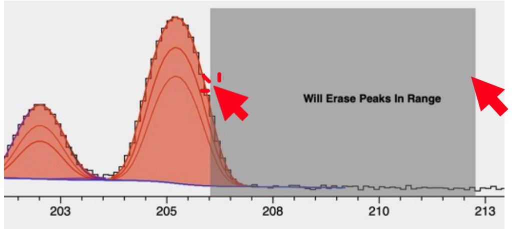
And just like normally in InterSpec, you can move your mouse to the edges of the ROI to adjust its extent

And you can instead adjust properties in the “Energy Ranges” section


Lower Energy 317.508 keV
Upper Energy 322.27 keV
Continuum Type: Linear
☑ Force full-range



You can also right-click on a ROI to adjust its properties

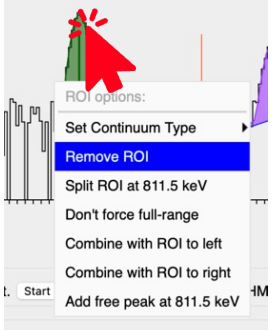


Holding the SHIFT key + click & drag with mouse allows you to delete ROIs, or just remove some of their extent

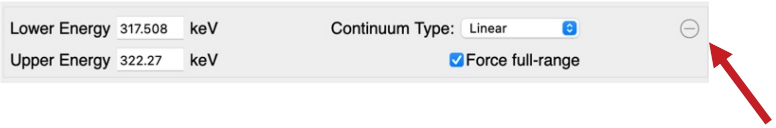
Interactions with the spectrum (zoom-in/out, pan, toggle log/lin, show energy strip chart, etc) in this tool are largely the same as normal within InterSpec

INTRODUCTION TO PERFORMING AN ANALYSIS – ADDING ROIS (CONT)

ROIs can also be removed in a number of ways:



Right click on ROI, and choose “Remove ROI”



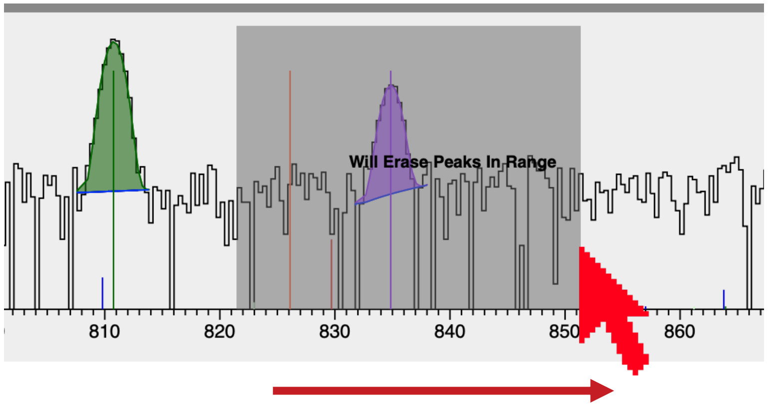
Lower Energy 317.508 keV

Upper Energy 322.27 keV

Continuum Type: Linear

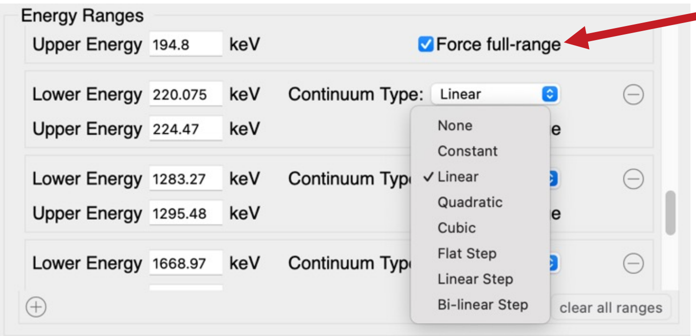
☑ Force full-range

Click the “-” button in “Energy Ranges” area



Shift key + click and drag

INTRODUCTION TO PERFORMING AN ANALYSIS – ADDING ROIS (CONT)



Energy Ranges

Upper Energy 194.8 keV ☑ Force full-range

Lower Energy 220.075 keV Continuum Type: Linear

Upper Energy 224.47 keV

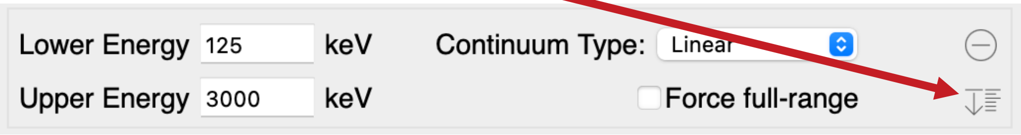
Lower Energy 1283.27 keV Continuum Type

Upper Energy 1295.48 keV

Lower Energy 1668.97 keV Continuum Type

Normally you want to use the entire energy range you specify – that is, a single continuum, plus peaks for however many gammas the sources define, will be fit in the ROI.

However, you can enter a large range (ex. 125 keV to 3000 keV), and un-check this option, to have the tool try and pick out the statistically-significant portions of the spectrum that may have peaks from the entered sources/nuclides – after a solution is found you can expand this ROI to the found regions, and edit from there



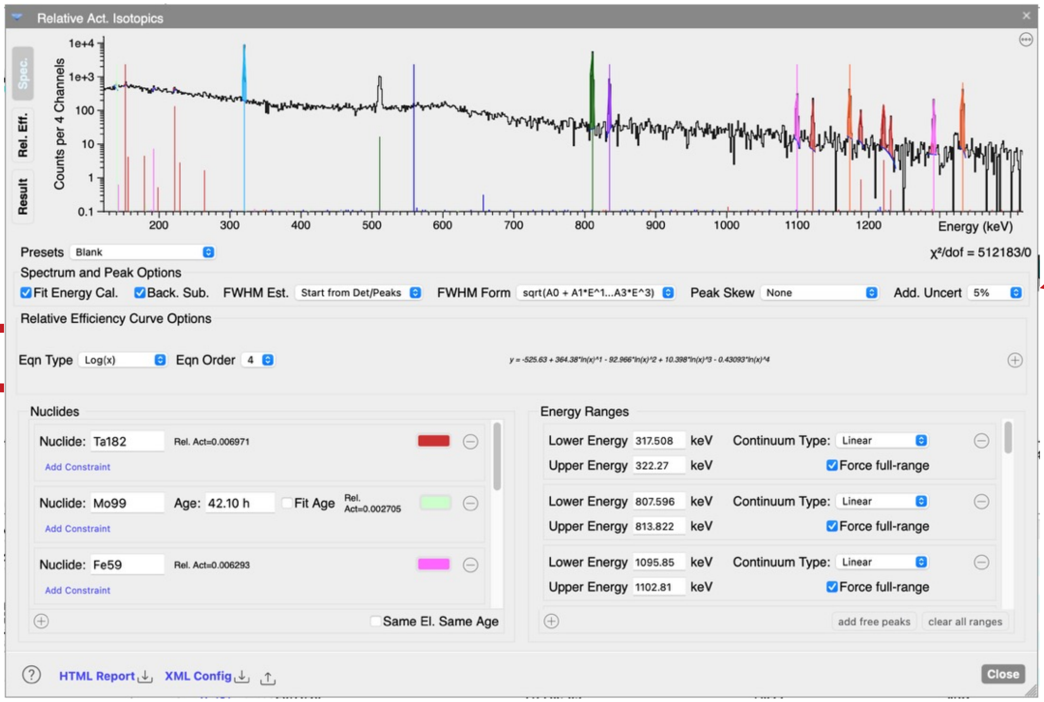
Lower Energy 125 keV Continuum Type: Linear

Upper Energy 3000 keV ☐ Force full-range

Using a large energy range and having the tool pick-out relevant sub-ranges is not recommended!
(sometimes it works okay, but as of 20250630, the implementation could use more work)

INTRODUCTION TO PERFORMING AN ANALYSIS – PICKING OPTIONS

Relative Efficiency curve options



Presets: Blank

χ²/dof = 512183/0

Spectrum and Peak Options

☑ Fit Energy Cal. ☑ Back. Sub. FWHM Est.: Start from Det/Peaks FWHM Form: sqrt(A0 + A1*E^1...A3*E^3) Peak Skew: None Add. Uncert: 5%

Relative Efficiency Curve Options

Eqn Type: Log(x) Eqn Order: 4

y = -525.63 + 364.38*ln(x)^1 - 92.966*ln(x)^2 + 10.398*ln(x)^3 - 0.43093*ln(x)^4

Nuclides

Nuclide: Ta182 Rel. Act=0.006971 🟥
Add Constraint

Nuclide: Mo99 Age: 42.10 h ☐ Fit Age Rel. Act=0.002705 🟩
Add Constraint

Nuclide: Fe59 Rel. Act=0.006293
Add Constraint

☐ Same El. Same Age

Energy Ranges

Lower Energy 317.508 keV Continuum Type: Linear
Upper Energy 322.27 keV ☑ Force full-range

Lower Energy 807.596 keV Continuum Type: Linear
Upper Energy 813.822 keV ☑ Force full-range

Lower Energy 1095.85 keV Continuum Type: Linear
Upper Energy 1102.81 keV ☑ Force full-range

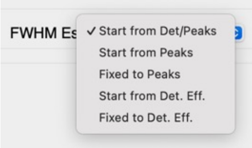
add free peaks clear all ranges

Spectrum and peak options

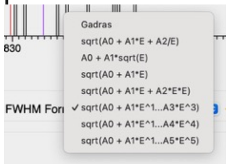
There are two areas that contain options related to the fit.

INTRODUCTION TO PERFORMING AN ANALYSIS – SPEC/PEAK OPTIONS

The FWHM, as a function of energy is usually fit to the data using your chosen ROIs, but you can instead fix it to your det. eff. function, or manually fit peaks



You can choose the functional form of FWHM fit to data



You can add uncertainty to gammas branching ratios – not usually recommended

(its actually an additional uncertainty for each observable peak)

Spectrum and Peak Options

☑ Fit Energy Cal. ☑ Back. Sub. FWHM Est. Start from Det/Peaks FWHM Form sqrt(A0 + A1*E^1...A3*E^3) Peak Skew None Add. Uncert 5%

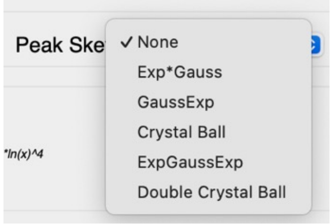
If energy calibration is slightly off, the gain and/or offset can be fit while performing analysis.

It needs to be kinda close to begin with – i.e., peaks from primary gammas should at least overlap with the true energy a little

If the background has peaks or features in any of your ROIs, you can enable background subtraction (if you have a background spectrum), so you don't have to include background nuclides in your analysis

Background subtraction is performed on a channel-by-channel basis, but proper variance is tracked

There are six of different models of peak skew you can choose from



"GaussExp" is usually good for HPGe, while "Double Crystal Ball" is good for CZT.

INTRODUCTION TO PERFORMING AN ANALYSIS – RELATIVE EFF OPTS

The relative efficiency options control the “smooth line” the relative activities are fitted to

Roughly: this line gives the shape of full-energy peak efficiency × attenuation

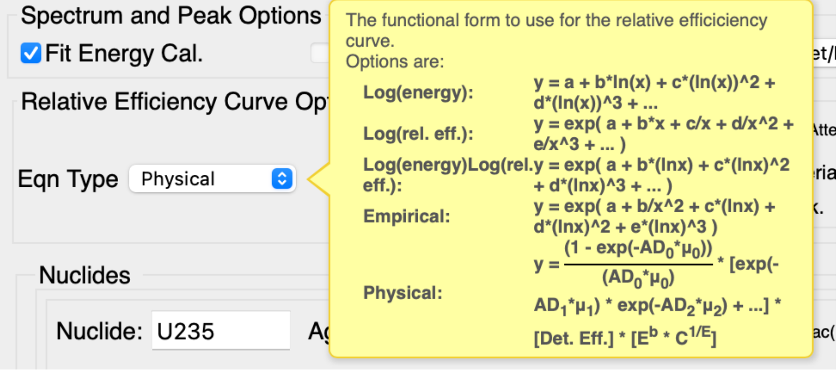
Log(energy)[†], Log(rel eff)[†], Log(energy)Log(rel eff)[†], and Empirical[‡] are polynomial equations, where the only option is order of equation to use – recommendation is to use the lowest order that provides good peak fits

Relative Efficiency Curve Options

Eqn Type: Log(x)
Eqn Order: 3

$y = 110.27 - 68.702*\ln(x)^1 + 13.898*\ln(x)^2 - 0.90482*\ln(x)^3$

(if using for U/Pu enrichment, do not include peaks below the 120 keV x-ray absorption edge, as shape of these curves do not account for this discontinuity)



Spectrum and Peak Options

☑ Fit Energy Cal.

Relative Efficiency Curve Op

Eqn Type: Physical

Nuclides

Nuclide: U235

The functional form to use for the relative efficiciency curve.
Options are:

Log(energy):	y = a + b*ln(x) + c*(ln(x))^2 + d*(ln(x))^3 + ...
Log(rel. eff.):	y = exp(a + b*x + c/x + d/x^2 + e/x^3 + ...)
Log(energy)Log(rel. eff.):	y = exp(a + b*(lnx) + c*(lnx)^2 + d*(lnx)^3 + ...)
Empirical:	y = exp(a + b/x^2 + c*(lnx) + d*(lnx)^2 + e*(lnx)^3)
Physical:	$y = \frac{(1 - \exp(-AD_0 * \mu_0))}{(AD_0 * \mu_0)} * [\exp(-AD_1 * \mu_1) * \exp(-AD_2 * \mu_2) + \ldots] * [\text{Det. Eff.}] * [E^b * C^{1/E}]$

The Physical model[§] allows accounting for k-edge x-ray attenuation (eg. U/Pu), as well as self-attenuation, and external attenuations

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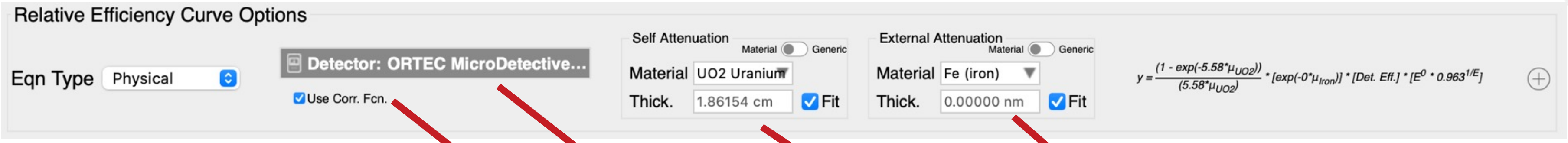
[†] These functional forms are from Mike Enghausers “RelativeEff Uiso” spreadsheet application

[‡] Functional form taken from FRAM

[§] Model used is an adaptation of the models used by MGA and FRAM

INTRODUCTION TO PERFORMING AN ANALYSIS – PHYSICAL MODEL



Relative Efficiency Curve Options

Eqn Type: Physical

Detector: ORTEC MicroDetective...

☑ Use Corr. Fcn.

Self Attenuation (Material / Generic)

Material: UO2 Uranium

Thick.: 1.86154 cm ☑ Fit

External Attenuation (Material / Generic)

Material: Fe (iron)

Thick.: 0.00000 nm ☑ Fit

$$y = \frac{(1 - \exp(-5.58 * \mu_{UO2}))}{(5.58 * \mu_{UO2})} * [\exp(-0 * \mu_{Iron})] * [Det.\ Eff.] * [E^{0} * 0.963^{1/E}]$$

$$RE = [E^{B}C^{1/E}] \times [Det.Eff.] \times \left[\frac{1 - \exp(-\mu_0 x_0)}{\mu_0 x_0}\right] \times \exp(-\mu_1 x_1)$$

Modified Hoerl function[†] accounts for differences between used detector efficiency, and actual efficiency, as well as other small issues. If you know your detectors efficiency, you may not want to use this option.
If used, the B and C coefficients are fit.

Like other places in InterSpec, you can choose your efficiency function. You don't need an exact efficiency function if you use the Corr. Fcn.

This term accounts for self-attenuating sources. Select any material in the InterSpec library – or use generic AN/AD – or leave it blank to not use. Specify a thickness, or fit for it.

External attenuation – same options as self-attenuation

[†] The modified Hoerl function was introduced in FRAM v4, see LA-14018

INTRODUCTION TO PERFORMING AN ANALYSIS – RELATIVE EFF OPTS

- For Uranium or Plutonium problems, the Physical Model is likely better option
    - If the detectors intrinsic efficiency is known, using the Correction Function may not be necessary. If the intrinsic efficiency is not known, just pick the efficiency function that comes with InterSpec that you think is closest, and use the correction function
    - The other functional forms can work well for U/Pu, but you can only use ROIs above 120 keV (i.e., the attenuation k-edge). Pick the equation form whose solution “looks” the best[†], by eye, and use the lowest order of the equation that gives a good answer
- For non U/Pu problems that are not attenuated by a large Z shielding, or only have peaks above the shielding k-edge (if it has one), one of the polynomial forms is usually good-enough – but the Physical Model may still be a good option

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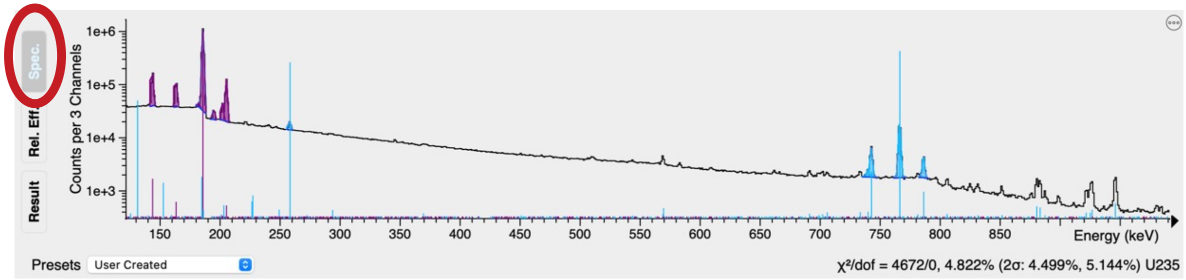
[†] What ”Looks” best will become clearer – but in short it’s the quality of peak fits in each of the chosen ROIs, as well as the rel. eff. function looking like a reasonable/physical curve – this may be a bit unsatisfying for some

INTRODUCTION TO PERFORMING AN ANALYSIS – RESULTS

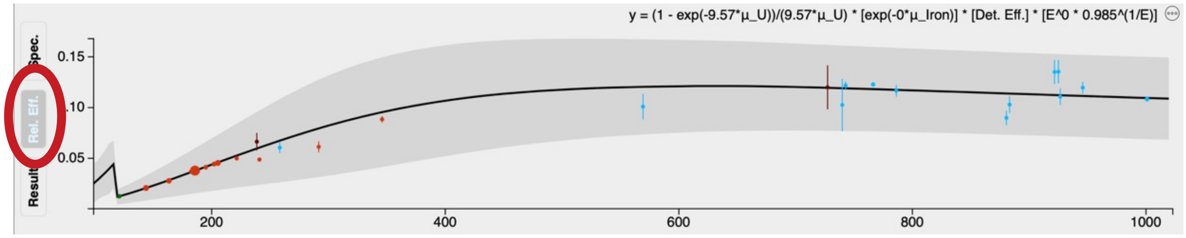
The upper part of the tool lets you easily see results of the fit

Interacting with this spectrum is similar to normally in InterSpec

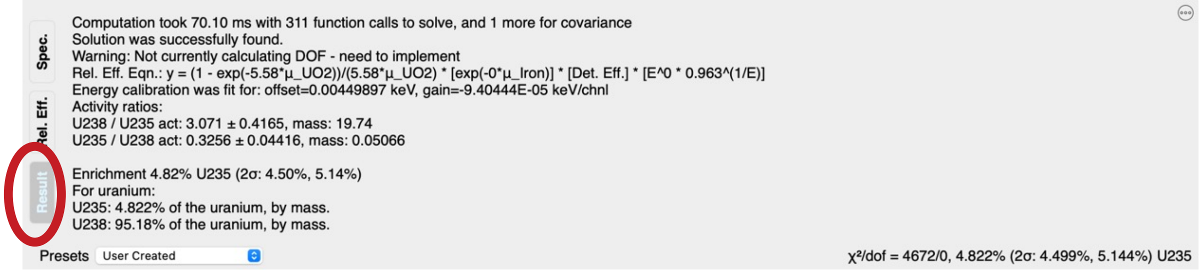


You can zoom-in/out to inspect the quality of peak fit

You can mouse-over data points for a little more info on the Rel. Eff. curve chart



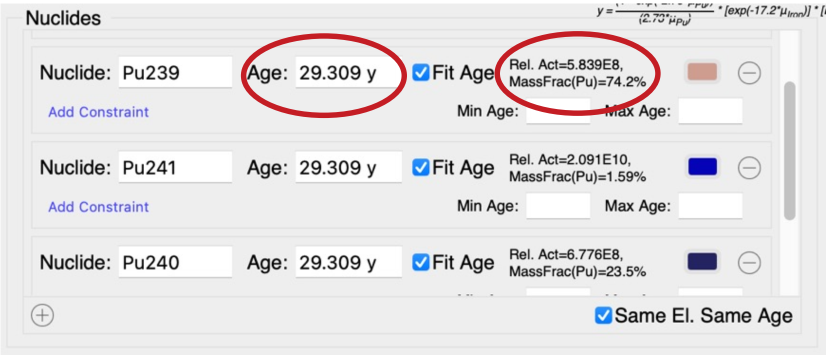
A short summary of the results



```
Computation took 70.10 ms with 311 function calls to solve, and 1 more for covariance
Solution was successfully found.
Warning: Not currently calculating DOF - need to implement
Rel. Eff. Eqn.: y = (1 - exp(-5.58*µ_UO2))/(5.58*µ_UO2) * [exp(-0*µ_Iron)] * [Det. Eff.] * [E^0 * 0.963^(1/E)]
Energy calibration was fit for: offset=0.00449897 keV, gain=-9.40444E-05 keV/chnl
Activity ratios:
U238 / U235 act: 3.071 ± 0.4165, mass: 19.74
U235 / U238 act: 0.3256 ± 0.04416, mass: 0.05066

Enrichment 4.82% U235 (2σ: 4.50%, 5.14%)
For uranium:
U235: 4.822% of the uranium, by mass.
U238: 95.18% of the uranium, by mass.
```

```
χ²/dof = 4672/0, 4.822% (2σ: 4.499%, 5.144%) U235
```

INTRODUCTION TO PERFORMING AN ANALYSIS – RESULTS

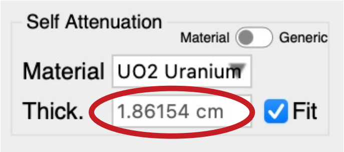

Nuclides

Nuclide: Pu239 Age: 29.309 y ☑ Fit Age Rel. Act=5.839E8, MassFrac(Pu)=74.2%

Nuclide: Pu241 Age: 29.309 y ☑ Fit Age Rel. Act=2.091E10, MassFrac(Pu)=1.59%

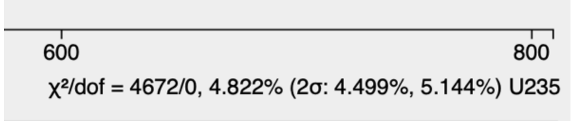
Nuclide: Pu240 Age: 29.309 y ☑ Fit Age Rel. Act=6.776E8, MassFrac(Pu)=23.5%

☑ Same El. Same Age


Self Attenuation

Material: UO2 Uranium

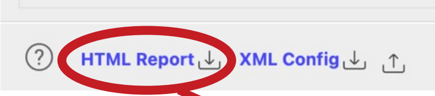
Thick.: 1.86154 cm ☑ Fit

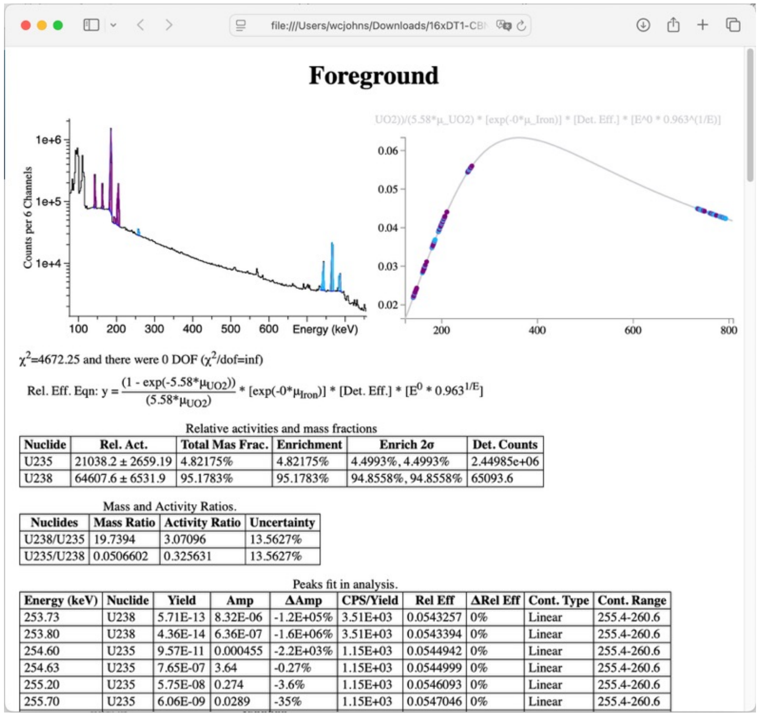


χ^2/dof = 4672/0, 4.822% (2σ: 4.499%, 5.144%) U235

Key fit values are updated in their own areas, but for more info see the “Results” text section of the upper part of the tool.

INTRODUCTION TO PERFORMING AN ANALYSIS – RESULTS





Foreground

χ^2=4672.25 and there were 0 DOF (χ^2/dof=inf)

Rel. Eff. Eqn: $y = \frac{(1 - \exp(-5.58*\mu_{UO2}))}{(5.58*\mu_{UO2})} * [\exp(-0*\mu_{Iron})] * [\text{Det. Eff.}] * [E^0 * 0.963^{1/E}]$

Relative activities and mass fractions

Nuclide	Rel. Act.	Total Mas Frac.	Enrichment	Enrich 2σ	Det. Counts
U235	21038.2 ± 2659.19	4.82175%	4.82175%	4.4993%, 4.4993%	2.44985e+06
U238	64607.6 ± 6531.9	95.1783%	95.1783%	94.8558%, 94.8558%	65093.6

Mass and Activity Ratios.

Nuclides	Mass Ratio	Activity Ratio	Uncertainty
U238/U235	19.7394	3.07096	13.5627%
U235/U238	0.0506602	0.325631	13.5627%

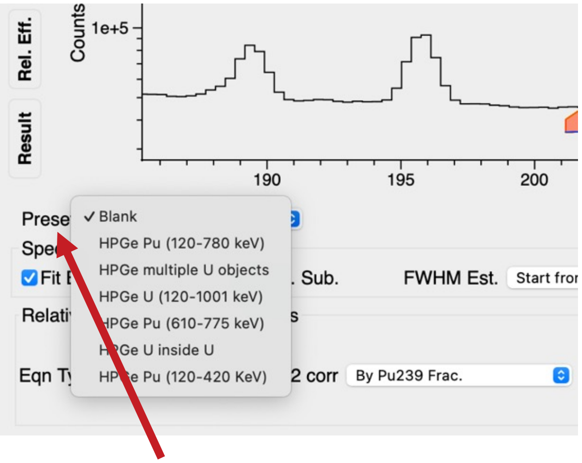
Peaks fit in analysis.

Energy (keV)	Nuclide	Yield	Amp	ΔAmp	CPS/Yield	Rel Eff	ΔRel Eff	Cont. Type	Cont. Range
253.73	U238	5.71E-13	8.32E-06	-1.2E+05%	3.51E+03	0.0543257	0%	Linear	255.4-260.6
253.80	U238	4.36E-14	6.36E-07	-1.6E+06%	3.51E+03	0.0543394	0%	Linear	255.4-260.6
254.60	U235	9.57E-11	0.000455	-2.2E+03%	1.15E+03	0.0544942	0%	Linear	255.4-260.6
254.63	U235	7.65E-07	3.64	-0.27%	1.15E+03	0.0544999	0%	Linear	255.4-260.6
255.20	U235	5.75E-08	0.274	-3.6%	1.15E+03	0.0546093	0%	Linear	255.4-260.6
255.70	U235	6.06E-09	0.0289	-35%	1.15E+03	0.0547046	0%	Linear	255.4-260.6

In the lower left-hand corner of the tool, you can export an HTML report, that is both interactive, and has a ton of information

SAVING CONFIGURATIONS, OR USING PREDEFINED ONES

Defining the setup for each spectrum would take far to long, and be ripe for mistakes



The tool comes with some preset configurations, and it is likely more will be added in the future.

You can export an XML file that contains the full configuration. You can then later re-import it, or just drag-n-drop the XML file onto InterSpec to open the tool with that configuration

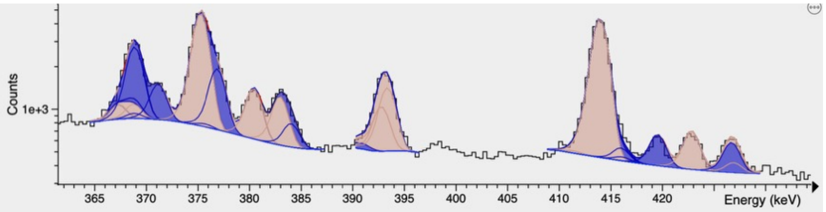
If it's a configuration you use often, you can place the XML file into a directory named "rel_act", in the InterSpec user data directory[†], and then it will appear in the "Presets" drop-down menu

---

[†] The InterSpec user data directory by going to "Help" → "About InterSpec" → "Data", and on windows is usually `C:\Users\<username>\AppData\Roaming\InterSpec`

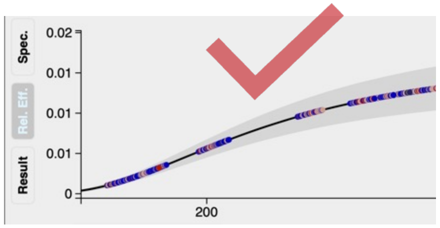
HOW TO JUDGE THE QUALITY OF A SOLUTION

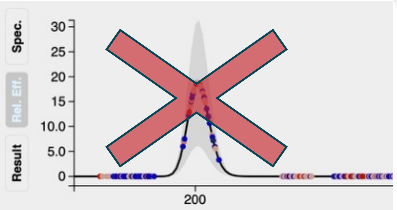
HOW TO TELL IF A SOLUTION IS GOOD



All of the peak fits look good

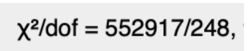
If not, you may need to:
- Change peak skew model
- Change continuum type for one or more ROIs
- Add nuclides, or x-rays, or reactions
- Change the ROI extents
- Fit for an age, or use a pre-determined age
- Background subtract
- Manually correct for non-linearities in energy cal.
- Choose a different Rel. Eff. curve type, or change order fit
- For Physical Model, choose a different shielding type
- For Physical Model, toggle use of correction function
- Add a age, mass-fraction, rel. act. ratio, or rel. act. Constraint, to help get fitter in reasonable starting range
- For Physical Model, choose a detector efficiency closer to your detectors efficiency
- Manually determine FWHM functional value, using the “Fit FWHM…” tool, and then fix this tool to use that value



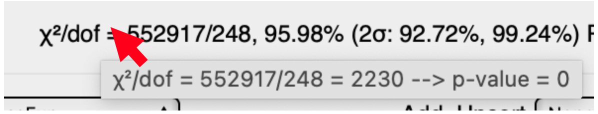


The Rel. Eff. line should look physical

This line is *roughly* detector intrinsic efficiency, convoluted with attenuation effects, so its shape should be something plausible physical.
As of 20250630, the peak markers will always be directly on the Rel. Eff. line, unless an ”Add. Uncert” is used.



χ²/dof = 552917/248,



χ²/dof = 552917/248, 95.98% (2σ: 92.72%, 99.24%)

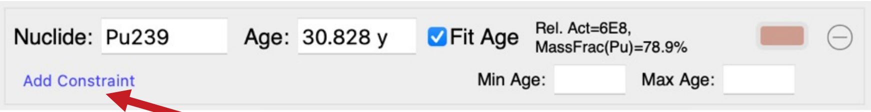
χ²/dof = 552917/248 = 2230 --> p-value = 0

The χ^2/dof can be used to compare different options with each other, with lower values indicating better agreement with data, but you must not change the ROI extents between compared options

(If you place the mouse over this text, a tooltip will show giving the p-value)

CONSTRAINTS

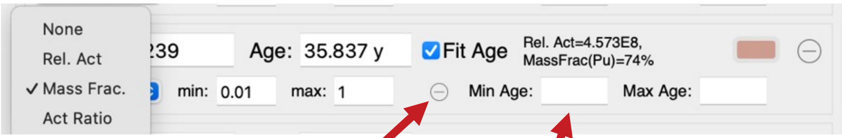
CONSTRAINTS:



Nuclide: Pu239 Age: 30.828 y ☑ Fit Age Rel. Act=6E8, MassFrac(Pu)=78.9%

Min Age: Max Age:

You can add a constraint by clicking “Add Constraint”



239 Age: 35.837 y ☑ Fit Age Rel. Act=4.573E8, MassFrac(Pu)=74%

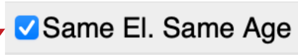
min: 0.01 max: 1 Min Age: Max Age:

Three different types of source constraints are available

Remove constraint

And if “Fit Age” is selected, you can optionally enter an age range to us (if blank, will use 0 to 120 years)

- “Rel. Act.”: Fixed the relative activity value to a range, or fixed value
- “Mass Frac.”: Fixes the fraction of mass of an element that a nuclide comprises, to either a constricted range of values, or a fixed value
- “Act Ratio”: Fixes the rel. activity of one source, to be a ratio of rel. activity of another source



☑ Same El. Same Age

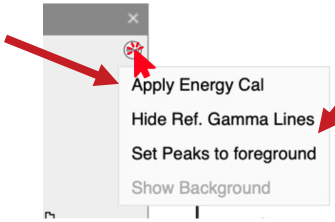
If you know the ages of all nuclides of an element should be the same (eg. U/Pu), and you are fitting age, make sure to check this option

The constraints are mostly useful for low-statistics measurements, or if a given source only has a low-statistics peak, or if you want to use a declared value for one quantity, to help determine the others.

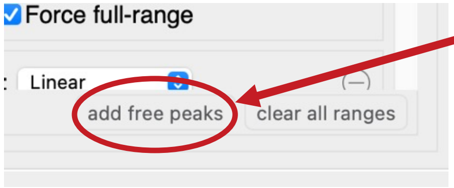
MISC. USEFUL FEATURES

MISC. USEFUL FEATURES

You can apply the fit energy calibration to the foreground spectrum – this can help to bring the reference photopeak lines into the right place on the spectrum, and reduce things being fit for, since you no longer need to fit energy cal



If you want to use these fit peaks to do other calculations with (eg, fit Activity/Shielding, or gammas into 4pi), you can do this here



You can add “free” peaks – not associated with a source, to account for sum peaks in a ROI, escape peaks, or unknown sources. The amplitude, and optionally FWHM of these peaks float freely and don’t effect rel. eff.

HOW THE TOOL WORKS INTERNALLY

DETAILS USEFUL TO UNDERSTAND HOW THE TOOL WORKS

- Given the source, ROIs, and options you have selected, the tool
  - Uses a non-linear optimizer[†] to simultaneously determine:
    {rel. acts, rel. eff. coefs, energy cal., nuclide ages, FWHM coefs, skew coefs}
    This information is combined with source gamma lines (performing any aging necessary) to determine peak amplitude, shape and positions.
  - A linear least squares (using two-sided Jacobi SVD decomposition[‡]) fit is performed to determine the peak-continuum for each ROI. A single continuum is used for each ROI.
  - Then a comparison between the predicted channel counts within ROIs, to the observed channel counts, divided by variance of each channel, is performed to compute the residuals.
  - "automatic differentiation"[§] is used to compute the partial derivative of each residual (ie, data channel comparison to predicted) w.r.t. each parameter being fit, to allow an efficient gradient decent
- There are lots of implementation details concerned with implementing constraints, or estimating initial values, or scaling parameters for better behavior, or making sure to find the optimum solution, but hopefully knowing these wouldn't enlighten anyone

[†] The Google Ceres Solver package, http://ceres-solver.org, is used as the non-linear optimizer in this tool
[‡] As provided by the Eigen C++ template library for linear algebra, https://eigen.tuxfamily.org/
[§] Automatic differentiation is really neat: https://en.wikipedia.org/wiki/Automatic_differentiation

DETAILS USEFUL TO UNDERSTAND HOW THE TOOL WORKS (CONT)

- ROIs are fit using only data from within the ROI. Other tools use a “background” region on either side of the ROI. This tool, and elsewhere in InterSpec, the peak and continuum are fit using just the spectrum-data within the ROI – no normalization to side-channels, or anything like that are performed
  - So feel free to have one ROI go right up to another large peak, with no “flat” channels between – everything will be taken care of for you.
  - You can even have an ROI only partially cover a peak (eg., only go 1 or 2 sigma from mean), and things should be just fine
- ROIs also include effects of peaks whose centroids are not within the ROI; e.g., nearby peaks, or when the peaks have significant tailing, will be accounted for
  - This is useful when the peaks have significant tailing, or a ROI with a small peak is right next to a ROI with a large peak that you maybe don’t want to use

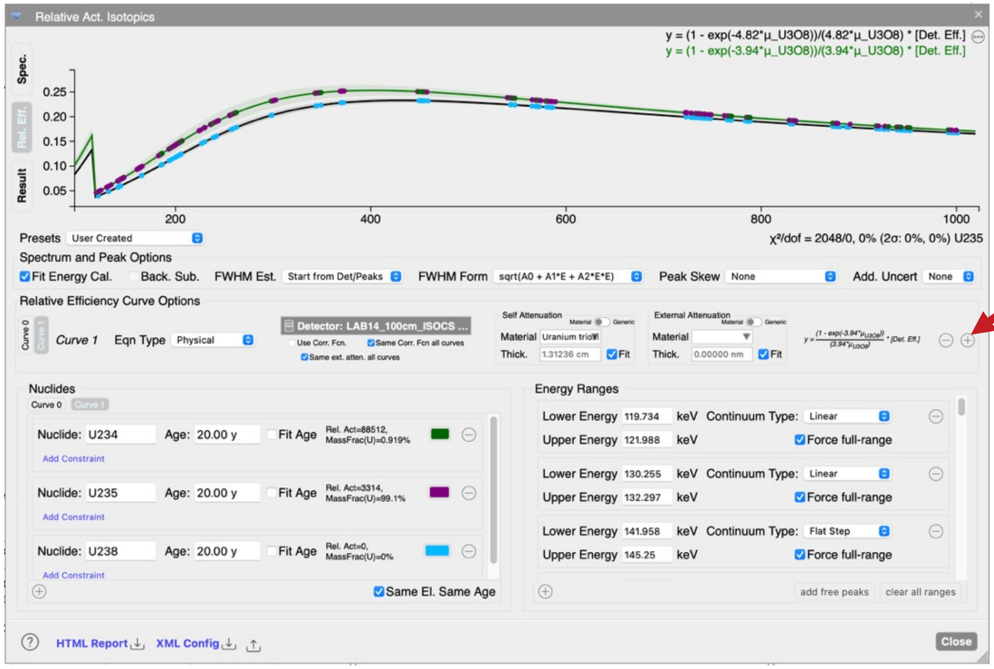
DIFFERENCES TO KEEP IN MIND FROM OTHER TOOLS

- There is essentially no uranium or plutonium specific knowledge embedded into the tool! U/Pu uses the same logic/algorithm/whatever as any other nuclide – no heuristics or priors
  - The only special handling for U/Pu is the Pu242 estimation from correlation, which is applied after the actual fit to data is completed, so doesn't affect fit to data
- When performing Pu enrichment and age calculations, in this tool, do not include Am241 as a source term (unless there was some initially present in the material). Instead, the Pu241 will be aged, and the Am241 will be accounted for in the progeny.
- Other tools (eg., FRAM†) first fit peaks, then from those peaks amplitude/location, determine all the other quantities. This tool fits for everything at the same time, which potentially allows using more information from the spectrum to determine the quantities of interest (but also makes the optimization much more difficult)
- This tool always uses the nuclear data from SandiaDecay – if you want to change any quantities, you will need to edit sandia.decay.xml, which will change values globally in InterSpec - and should not be necessary. Other tools may use different branching ratios for different parameter set.

† See section A.2 (starting on page 130) of https://www.osti.gov/biblio/1599022

MULTIPLE RELATIVE EFFICIENCY CURVES

MULTIPLE RELATIVE EFFICIENCY CURVES



y = (1 - exp(-4.82*µ_U3O8))/(4.82*µ_U3O8) * [Det. Eff.]

y = (1 - exp(-3.94*µ_U3O8))/(3.94*µ_U3O8) * [Det. Eff.]

χ²/dof = 2048/0, 0% (2σ: 0%, 0%) U235

Presets: User Created

Spectrum and Peak Options

☑ Fit Energy Cal. ☐ Back. Sub. FWHM Est. Start from Det/Peaks FWHM Form sqrt(A0 + A1*E + A2*E*E) Peak Skew None Add. Uncert None

Relative Efficiency Curve Options

Curve 1 Eqn Type Physical

Detector: LAB14_100cm_ISOCS ...

☐ Use Corr. Fcn. ☑ Same Corr. Fcn all curves

☑ Same ext. atten. all curves

Self Attenuation (Material / Generic)
Material Uranium trio
Thick. 1.31236 cm ☑ Fit

External Attenuation (Material / Generic)
Material
Thick. 0.00000 nm ☑ Fit

$y = \frac{(1 - \exp(-3.94 \cdot \mu_{U3O8}))}{(3.94 \cdot \mu_{U3O8})} \cdot [Det.\ Eff.]$

Nuclides

Nuclide	Age	Fit Age	
U234	20.00 y	☐	Rel. Act=88512, MassFrac(U)=0.919% 🟩
U235	20.00 y	☐	Rel. Act=3314, MassFrac(U)=99.1% 🟪
U238	20.00 y	☐	Rel. Act=0, MassFrac(U)=0% 🟦

Add Constraint

☑ Same El. Same Age

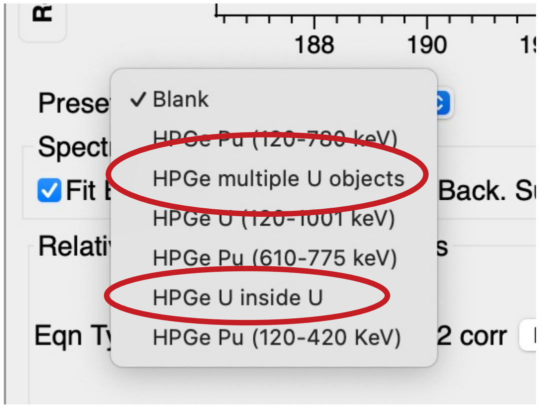
Energy Ranges

Lower Energy	Upper Energy	Continuum Type	Force full-range
119.734 keV	121.988 keV	Linear	☑
130.255 keV	132.297 keV	Linear	☑
141.958 keV	145.25 keV	Flat Step	☑

Add a Rel. Eff. Curve by clicking on the ”+” icon

- If you have multiple sources in view of the detector, that have different attenuations, you can use multiple relative efficiency curves.
  - If the separate sources don’t have any interfering peaks, you could perform analysis for each source separately

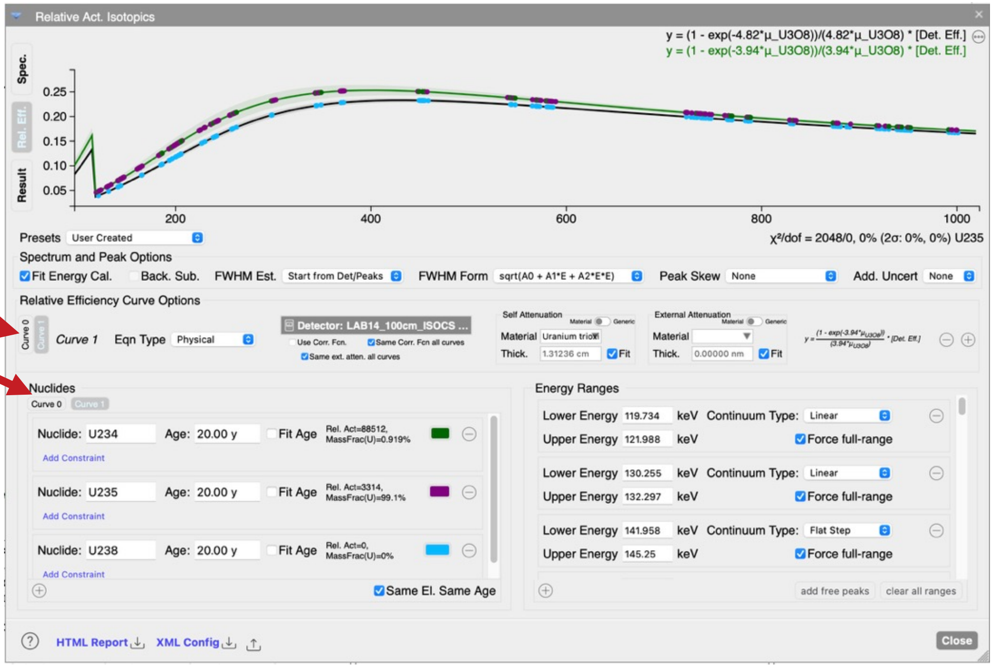
MULTIPLE RELATIVE EFFICIENCY CURVES (CONT)



There are some ”Presets” to help get you started – however, you will likely need to modify ROIs, nuclides, and other options to fit your particular circumstances

MULTIPLE RELATIVE EFFICIENCY CURVES (CONT)

When you have multiple rel. eff. curves, you can select which one the options/sources are displayed for using these menus



y = (1 - exp(-4.82*µ_U3O8))/(4.82*µ_U3O8) * [Det. Eff.]

y = (1 - exp(-3.94*µ_U3O8))/(3.94*µ_U3O8) * [Det. Eff.]

χ²/dof = 2048/0, 0% (2σ: 0%, 0%) U235

Presets: User Created

Spectrum and Peak Options

☑ Fit Energy Cal. ☐ Back. Sub. FWHM Est.: Start from Det/Peaks FWHM Form: sqrt(A0 + A1*E + A2*E*E) Peak Skew: None Add. Uncert: None

Relative Efficiency Curve Options

Curve 0 | Curve 1

Curve 1 Eqn Type: Physical

Detector: LAB14_100cm_ISOCS ...

☐ Use Corr. Fcn. ☑ Same Corr. Fcn all curves

☑ Same ext. atten. all curves

Self Attenuation (Material / Generic)
Material: Uranium triox
Thick.: 1.31236 cm ☑ Fit

External Attenuation (Material / Generic)
Material:
Thick.: 0.00000 nm ☑ Fit

$y = \frac{(1 - \exp(-3.94 \cdot \mu_{U3O8}))}{(3.94 \cdot \mu_{U3O8})} \cdot [Det. Eff.]$

Nuclides

Curve 0 | Curve 1

Nuclide	Age	Fit Age		
U234	20.00 y	☐	Rel. Act=88512, MassFrac(U)=0.919%	🟩
U235	20.00 y	☐	Rel. Act=3314, MassFrac(U)=99.1%	🟪
U238	20.00 y	☐	Rel. Act=0, MassFrac(U)=0%	🟦

Add Constraint

☑ Same El. Same Age

Energy Ranges

Lower Energy	Upper Energy	Continuum Type	Force full-range
119.734 keV	121.988 keV	Linear	☑
130.255 keV	132.297 keV	Linear	☑
141.958 keV	145.25 keV	Flat Step	☑

add free peaks | clear all ranges

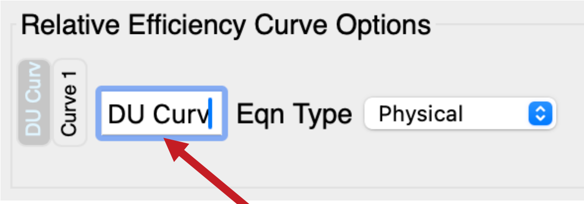
Each rel. eff. curve gets its own set of nuclides/source, and options – but all curves use the same ROIs

MULTIPLE RELATIVE EFFICIENCY CURVES (CONT)

Detector: GADRAS Det-X

☐ Use corr. fcn. ☐ Share corr. fcn.

☑ Share ext. atten. ☐ Shielded by other curves



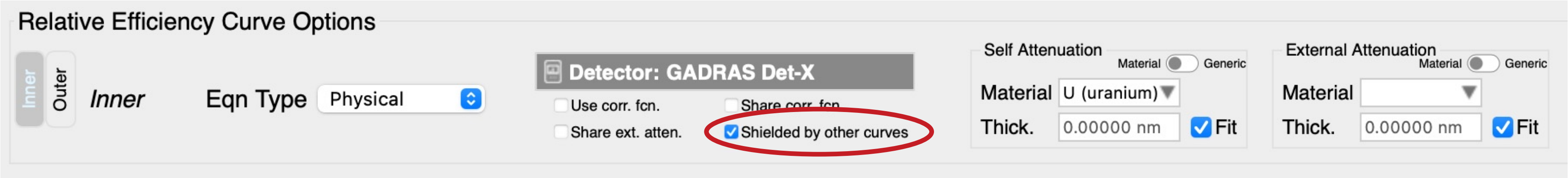
Relative Efficiency Curve Options

DU Curv Eqn Type Physical

You can rename the curves by clicking on the name, and then editing

You can also choose to share the same Modified Hoerl Correction function between curves, and/or the external attenuation (although its best to not use a correction function, particularly if you have a known detector efficiency)

MULTIPLE RELATIVE EFFICIENCY CURVES (CONT)



Relative Efficiency Curve Options

Inner

Eqn Type: Physical

Detector: GADRAS Det-X

☐ Use corr. fcn. ☐ Share corr. fcn.

☐ Share ext. atten. ☑ Shielded by other curves

Self Attenuation

Material: U (uranium)

Thick.: 0.00000 nm ☑ Fit

External Attenuation

Material:

Thick.: 0.00000 nm ☑ Fit

Or, if you know/suspect one source is surrounding another, you can choose for a curve to be attenuated by the other curves.

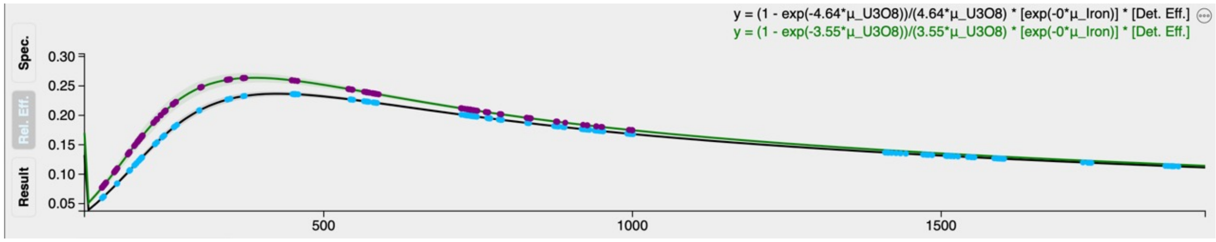
- In this case, the gammas from the “Inner” curve will be self-attenuated by Uranium, then they will go through the “Outer” curves self attenuation and its external attenuation.

This is not a true “spherical” model transport – no gammas from the “Outer” shell are transported through the “Inner” shell (e.g., gammas from the opposite side from the detector, of the outer shell).

SUGGESTIONS FOR MULTIPLE REL. EFF. CURVES

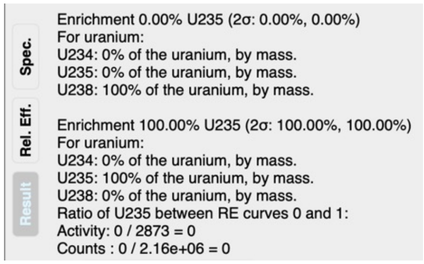
- This work is still fairly-early, so recommendations will likely evolve
- It is best to use the full-energy efficiency function for your detector, or its general model
  - Using the efficiency function from a detector of the same model, but different physical detector, appears to be fine, but there can be notable detector-to-detector differences
- It is best to NOT use the modified Hoerl correction function
  - Use of the correction function, and/or incorrect detector efficiency function, can greatly diminish the ability to discriminate multiple rel. eff. curves from the data
- You will need ROIs over a broad energy range, preferably including below and above ~200 keV
  - The change in attenuation is more significant (both as a function of energy, and of atomic number) below 200 keV or 300 keV, and above this the changes are much shallower
- You will need high-enough statistics data so the multiple efficiency functions can be discriminated
- If you are doing this for uranium – you will likely need the 258 keV peak from U238 to be well-developed - this peak helps to link the low energy U235 ROIs, with the higher energy U238 ROIs – otherwise there the ambiguity of response over such a large energy range with no peaks may make the results untrustworthy
- Sample should be at least 10 or 15 cm from detector, and deadtime low enough so that sum-peaks are not observed
- If you have a HEU object, and the same size LEU object next to each other – they will have the same relative attenuation curves, so this method will not discriminate that there are two different objects present. Similar limitations may apply to other circumstances
- The fit is particularly sensitive to energy variations in spectrum – using non-linear deviation pairs to correct small non-linearities is often-times necessary to get valid discrimination between curves

MULTIPLE RELATIVE EFFICIENCY CURVES – NON-HOMOGENEOUS U



y = (1 - exp(-4.64*µ_U3O8))/(4.64*µ_U3O8) * [exp(-0*µ_Iron)] * [Det. Eff.]
y = (1 - exp(-3.55*µ_U3O8))/(3.55*µ_U3O8) * [exp(-0*µ_Iron)] * [Det. Eff.]

- If you are using multiple relative efficiency curves to determine if a Uranium sample is homogenous or not, to conclude it is not a homogeneous sample:
  - The rel. eff. curves should be of distinct different shapes, and the uncertainties of the curves don’t overlap for at least ~50% of the curves
  - The reported enrichments for each curve should be separate, and the possible ranges should not overlap (eg., ≳3 σ apart)
  - The peak and continuum fits to each ROI must look good
  - The number of counts attributed to both curves must be non-zero, and enrichment defined for each curve



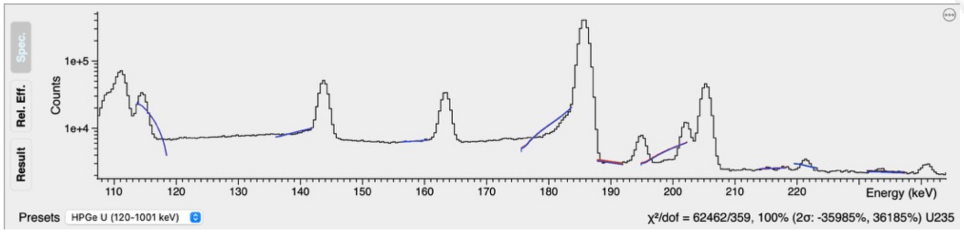
```
Enrichment 0.00% U235 (2σ: 0.00%, 0.00%)
For uranium:
U234: 0% of the uranium, by mass.
U235: 0% of the uranium, by mass.
U238: 100% of the uranium, by mass.

Enrichment 100.00% U235 (2σ: 100.00%, 100.00%)
For uranium:
U234: 0% of the uranium, by mass.
U235: 100% of the uranium, by mass.
U238: 0% of the uranium, by mass.
Ratio of U235 between RE curves 0 and 1:
Activity: 0 / 2873 = 0
Counts : 0 / 2.16e+06 = 0
```

- You may also consider making small changes to the settings, or ROIs used, to make sure the solution is stable, and the fact two curves were fit possible being an aberration

FAILURE MODES AND WORKAROUNDS

A POTENTIAL ISSUE – SOLUTION IS CATASTROPHICALLY BAD

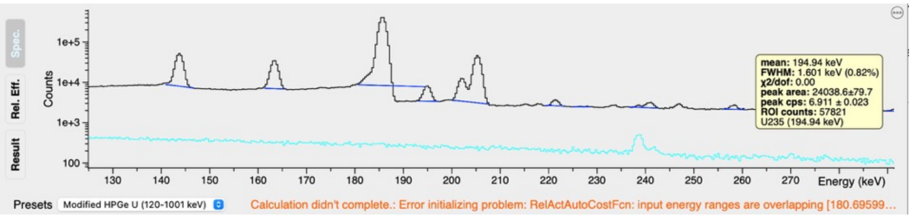
- Try unchecking “Fit Energy Cal.” and/or fix the FWHM to the detector efficiency or spectrum “peaks”
    - The cause for this is believed to be poor stability in the gradients of some fitting parameters, and a fix has been identified
- Make sure you have enough relevant ROIs defined. For example, you cant fit a nuclides age, or many nuclide activities, when only a single ROI is being used, or if you have a nuclide, but it doesn’t have gammas in any of the ROIs
- Check for an error message below the spectrum, and/or in the “Result” tab
- If still not fixed, you can try slightly changing ROIs or other settings
- Improving the starting parameter value estimates in the works



Presets HPGe U (120-1001 keV)

χ^2/dof = 62462/359, 100% (2σ: -35985%, 36185%) U235

Example bad solution – the energy calibration and FWHM fits diverged, and no peaks were fit



mean: 194.94 keV
FWHM: 1.601 keV (0.82%)
χ2/dof: 0.00
peak area: 24038.6±79.7
peak cps: 6.911 ± 0.023
ROI counts: 57821
U235 (194.94 keV)

Presets Modified HPGe U (120-1001 keV)

Calculation didn't complete.: Error initializing problem: RelActAutoCostFcn: input energy ranges are overlapping [180.69599...

Example with overlapping ROIs – see error message in orange below the spectrum

POTENTIAL FUTURE WORK

POSSIBLE (CURRENTLY UNFUNDED) FUTURE WORK:

- This tool was created with about 12 weeks of total effort; given its ambitions, there remains a number of potential upgrades in the future
- Top items on the TODO list:
  - Add mechanism to define ROIs in a peak-resolution independent way, so that a planar HPGe detector could use the same ROIs as a CZT detector
  - Additional improvements to the optimization routines
    - Rarely, the global minimum is not found, and a small perturbation to ROIs or options will result in a drastically better solution found – ensuring the global minimum is reached should be improved
    - The parameters determined by the non-linear fitter (ie, Ceres Solver) could use additional work in creating transformations between the physical quantities, and the quantities the solver works with, to improve the consistency of changes having similar magnitude effects to residuals
  - Improve automatic creation/determination of ROIs, so users don’t have to manually create for many cases
  - Automatically determine the best continuum type
  - More carefully verify, validate, quantify and publish performance of U/Pu enrichment results from a large corpus of known measurements
  - Improvements to the user interface and experience
  - Implement a ray-trace based physical relative efficiency model (reusing Act/Shielding fit code) to allow more detailed and higher fidelity modeling
  - Account for true-coincidence and random summing effects
  - Allow multiple external attenuators (currently calculation code accepts arbitrary number, but GUI only allows one per rel. eff. curve)
  - Optimize, and add more “preset” configurations (essentially no optimization has been done on current presets – they are just starting points)
  - Be able to transfer peak fits, and other results, of this tool to the Activity/Shielding Fit tool to allow quantifying results (or perhaps add some capabilities of the Activity/Shielding Fit tool to this tool)
  - Account for x-ray width; ie, implement Lorentzian/Voigt peak shape
  - Some small domain-specific calculations; ex, heat output of per gram of material, effective neutrons per gram, etc