INL/CON-23-71408-Revision-0

Development of a Versatile Analysis Method for the Isotopic Characterization of Alpha Spectra

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Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517, DE-AC07-05ID14517

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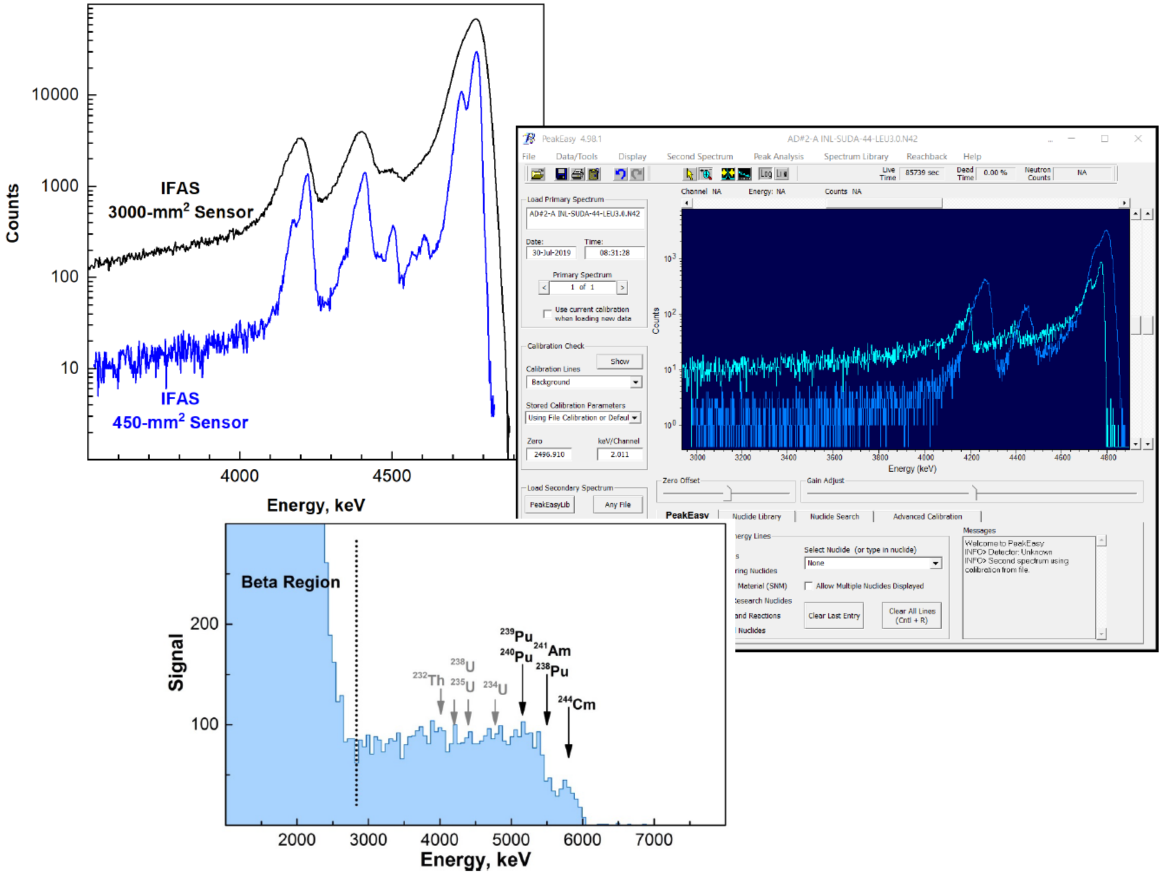
Development of a Versatile Analysis Tool for the Isotopic Characterization of Alpha Spectra

Difficulties with alpha spectra.

- Individual peak shapes have long tails that contribute significant counts across the spectrum, overlapping in energy with counts from other peaks
  - Unable to accurately fit individual peaks locally
- This effect can be magnified by the fact that alpha emissions often occur in multiplets that are close in energy
- Detectors often require stabilization techniques that leave artifacts in recorded spectra
- Difficult to accurately calibrate the spectra



Existing Analysis Tools

- AASIFIT & ADAM
  - Developed at STUK in Finland
  - Monte Carlo components work well (AASI), fitting algorithms are lower TRL and are not freely distributed
- ALPS
  - Developed by Dr. WestmeierGmbH
  - Very powerful, but not currently supported
- ALFITeX
  - Developed at University of Extremadura in Spain
  - Written as an MS Excel macro
- Hand fitting and eyeballing using ROIs
  - Works surprisingly well

What We Set Out to Develop

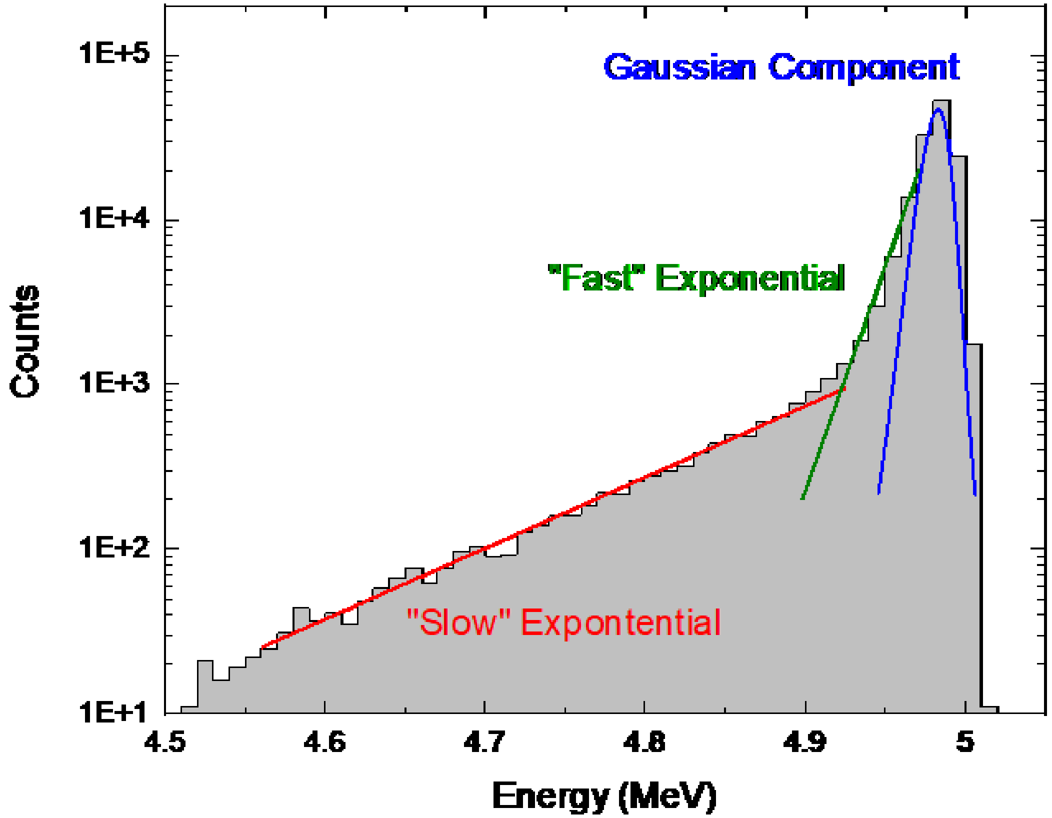
- Consistent and includes error analysis
- Fast
- Uncluttered with features irrelevant to our mission space
- Easy to use –particularly in the field
- Able to digest spectra directly from our detection systems
- Relies on evaluated nuclear data
- Painless to install on standard laboratory equipment

Fitting Method 1: Brute Force Non-Linear Regression

- Using a modified version of Levenberg-Marquardt algorithm that has been optimized for better global convergence
  - Based on the work of Hebden (1973) and More (1977)
- Initially formalized into a set of Fortran subroutines (MINPACK-1) by More, Garbow, and Hillstrom (1980)
- Partial derivative for Jacobians are calculated numerically
- Several tolerance factors can be tweaked for better convergence
- Requires initial guesses for each fit parameter – this includes identifying all emissions prior to fit
- Susceptible to convergence on local minima, particularly with a large set of fitting parameters

The Peak Model

- Single Tail Model - L’Hoir (1984), Bortels and Collaers (1987)
- BEST Model – Pomme and Marroyo (2014)

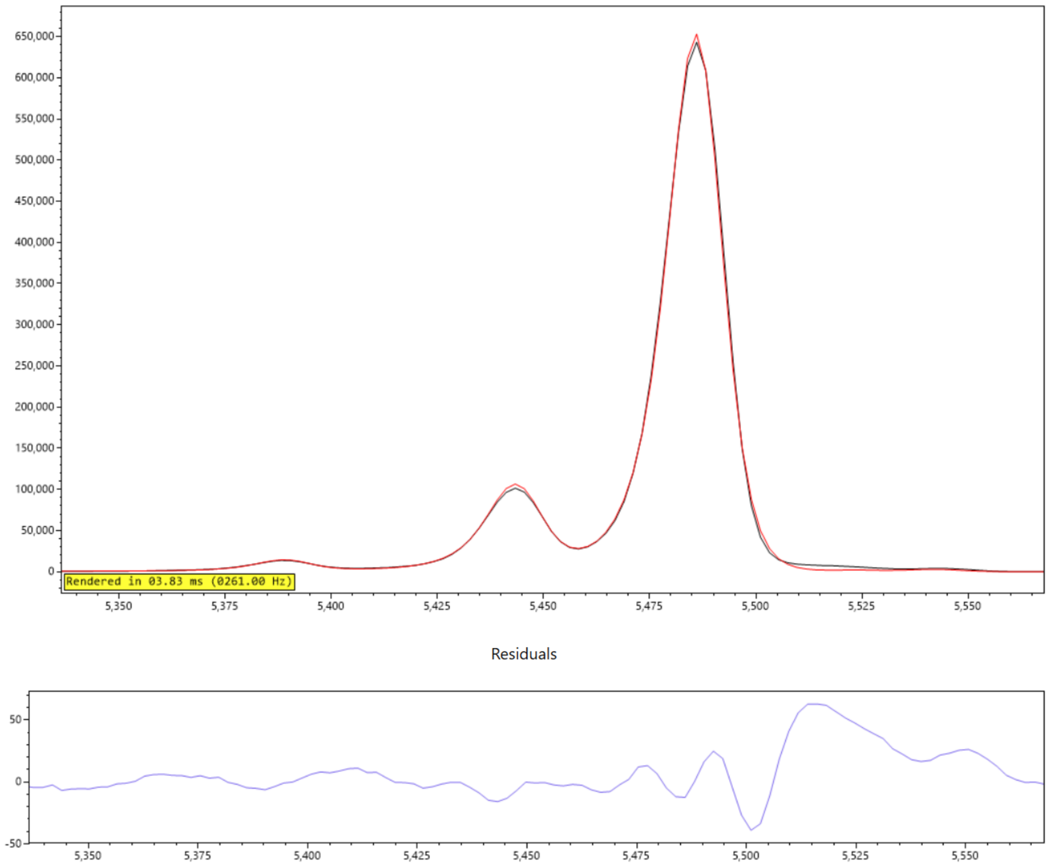


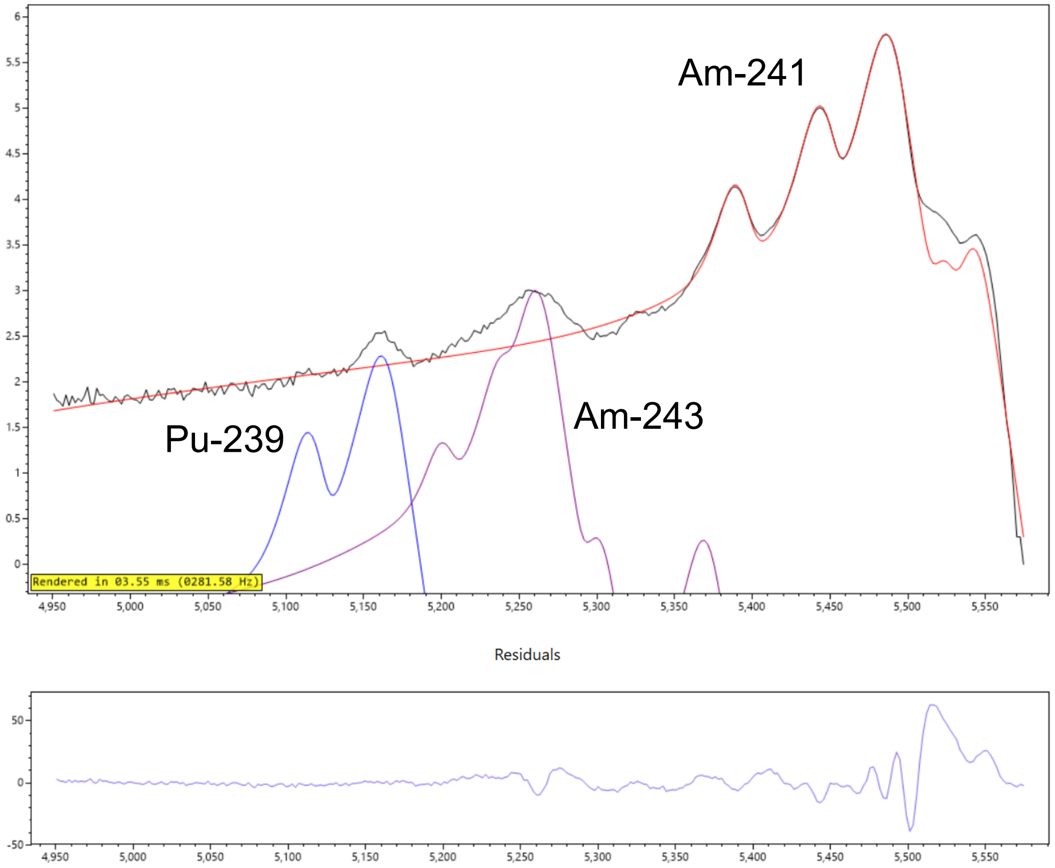
Simulated Single Peak Using AASI

We Fit Isotopes - Not Individual Peaks

- We store nuclear data that includes information like branching ratios, half-lives, specific activities, and individual emissions intensities in a SQLite database
- Individual peak intensities for a given isotope are held fixed, while a single isotope intensity is fit
- If all isotopes are fully represented in the spectra, and assuming efficiencies are flat across the spectrum, we can extract mass and atomic fraction information based on total count ratios:

Examples – Thin Am-241 Calibration Sample

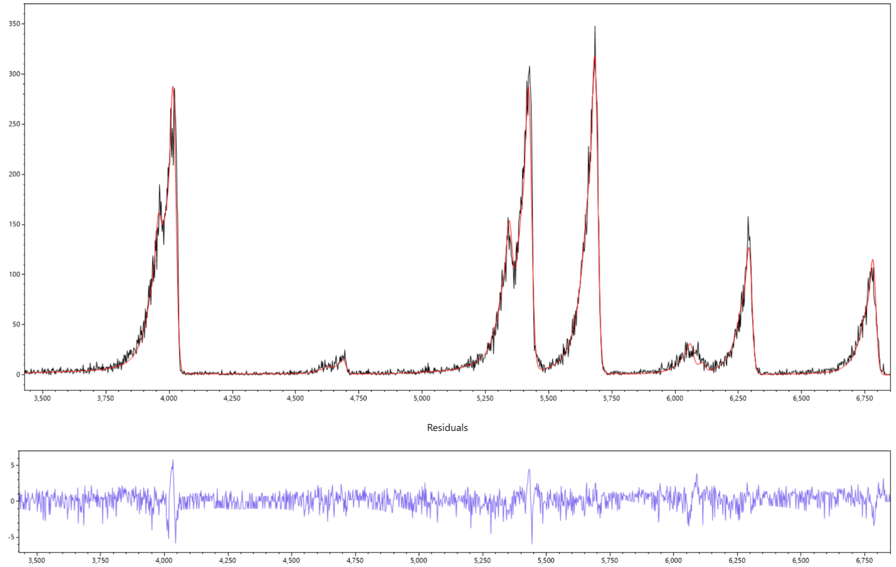


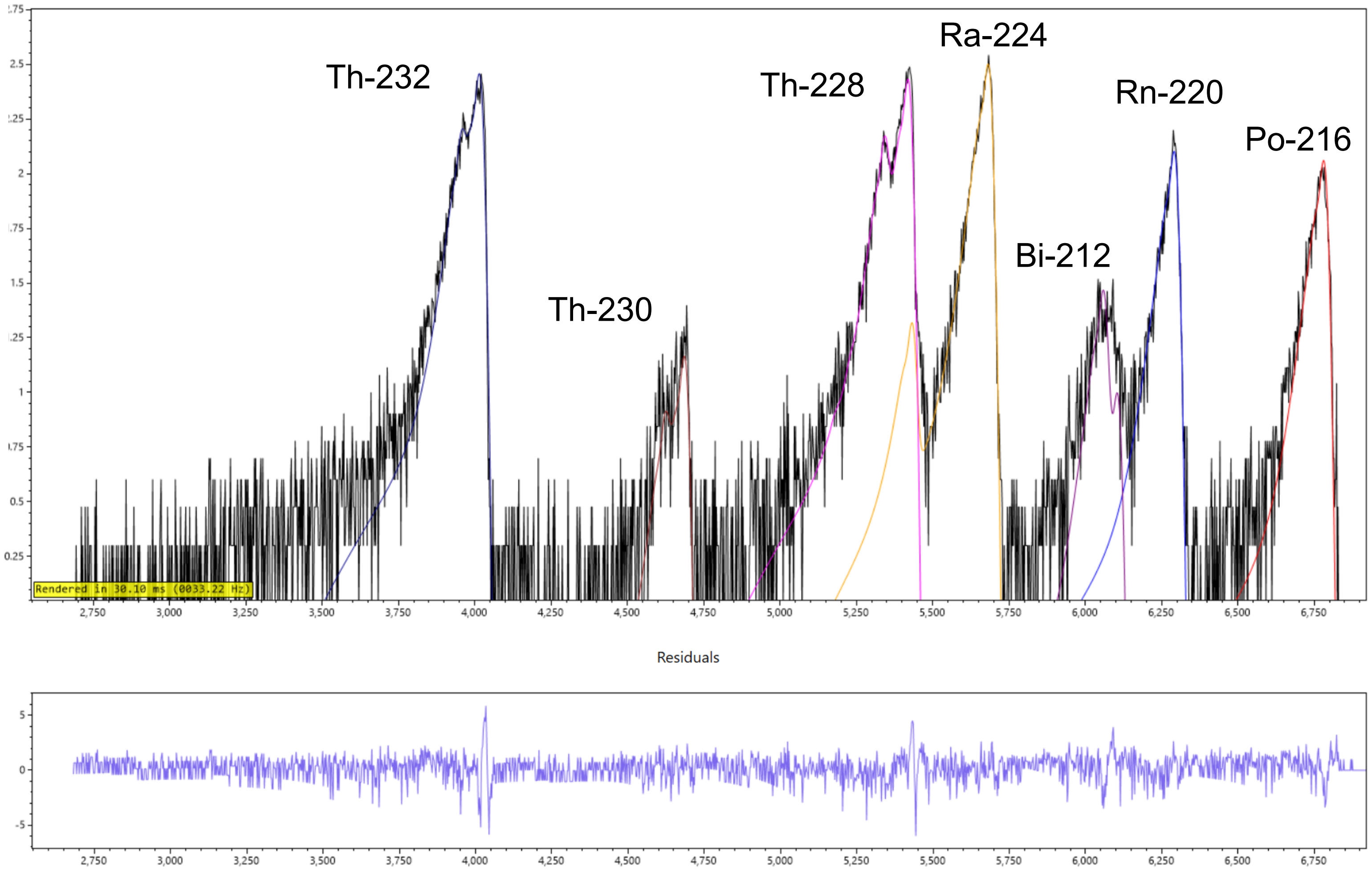


Fit Results: Am-241 95.87%, Am-243 2.53%, Pu-239 1.60%

Used 5 LE tails and 1 HE

Examples – Thin Th-232 Calibration Sample



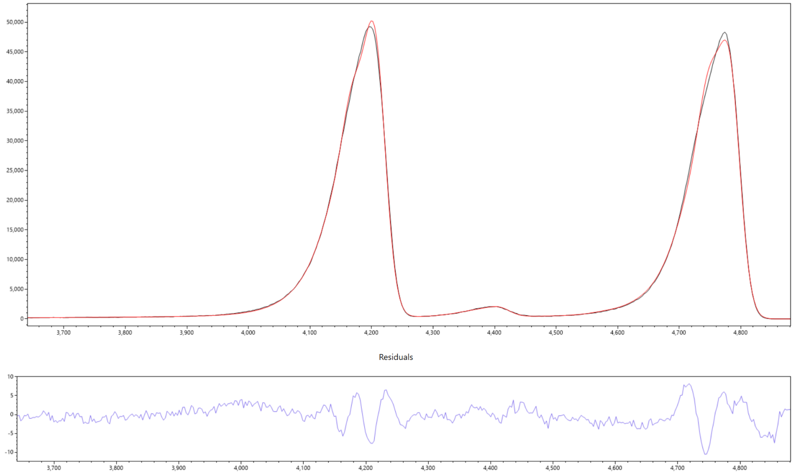


100% Th-232 to 3 significant digits

Used 5 LE tails and 1 HE

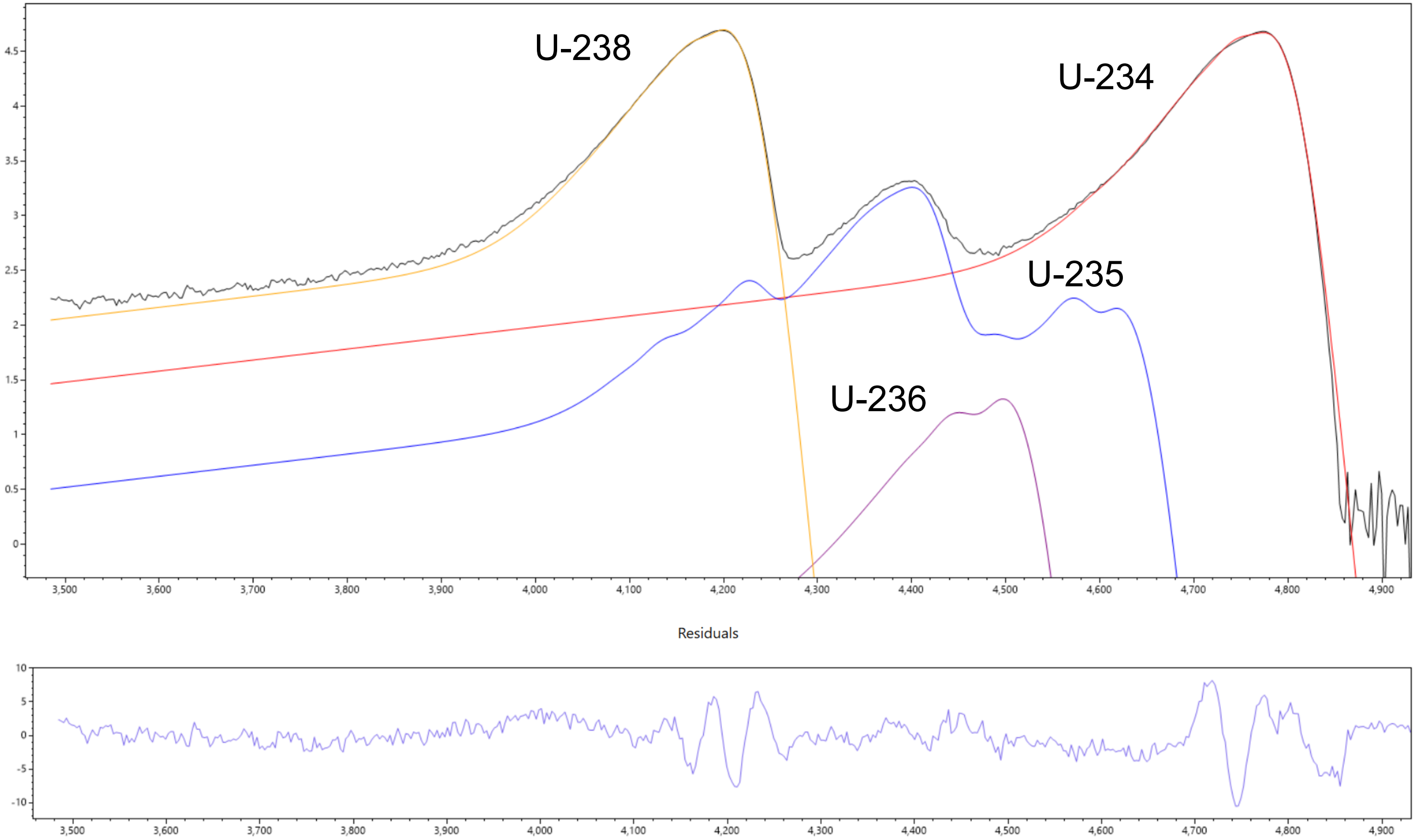
Thin Single-Use Destructive Analysis (SUDA) Sample

U-235: 0.717917 % ± 0.003618



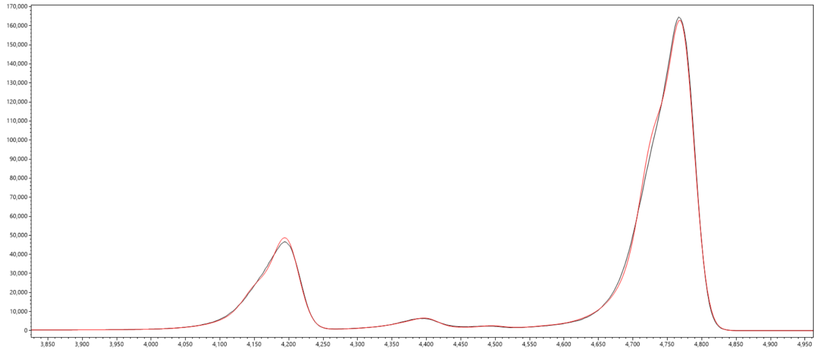
Fit U-235 at 0.707%

Using 4 LE Tails and 1 HE

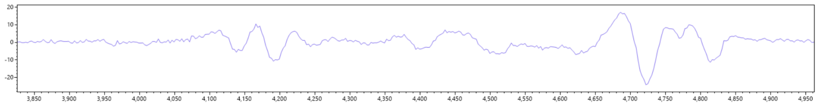


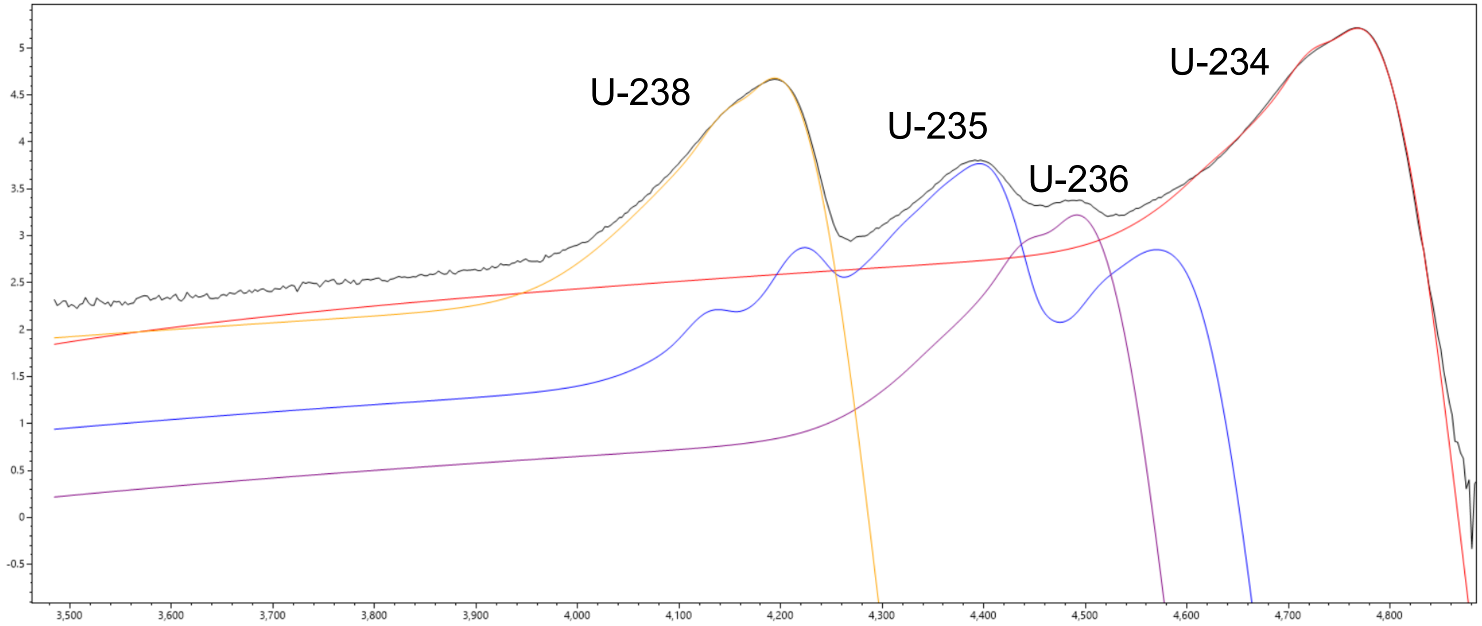
Thin Single-Use Destructive Analysis (SUDA) Sample

U-235: 2.332892 % ± 0.000724

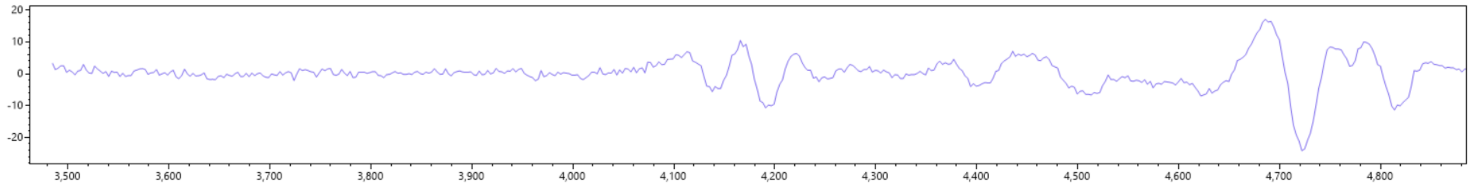


Residuals





Residuals

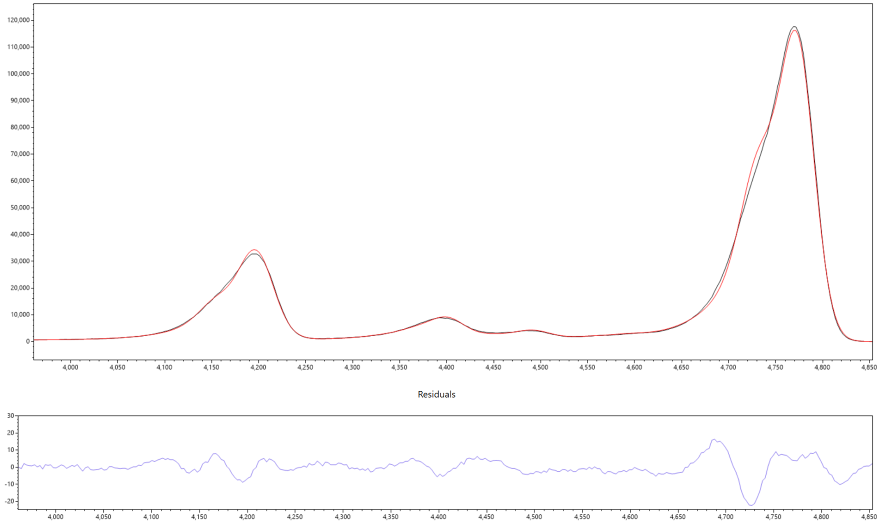


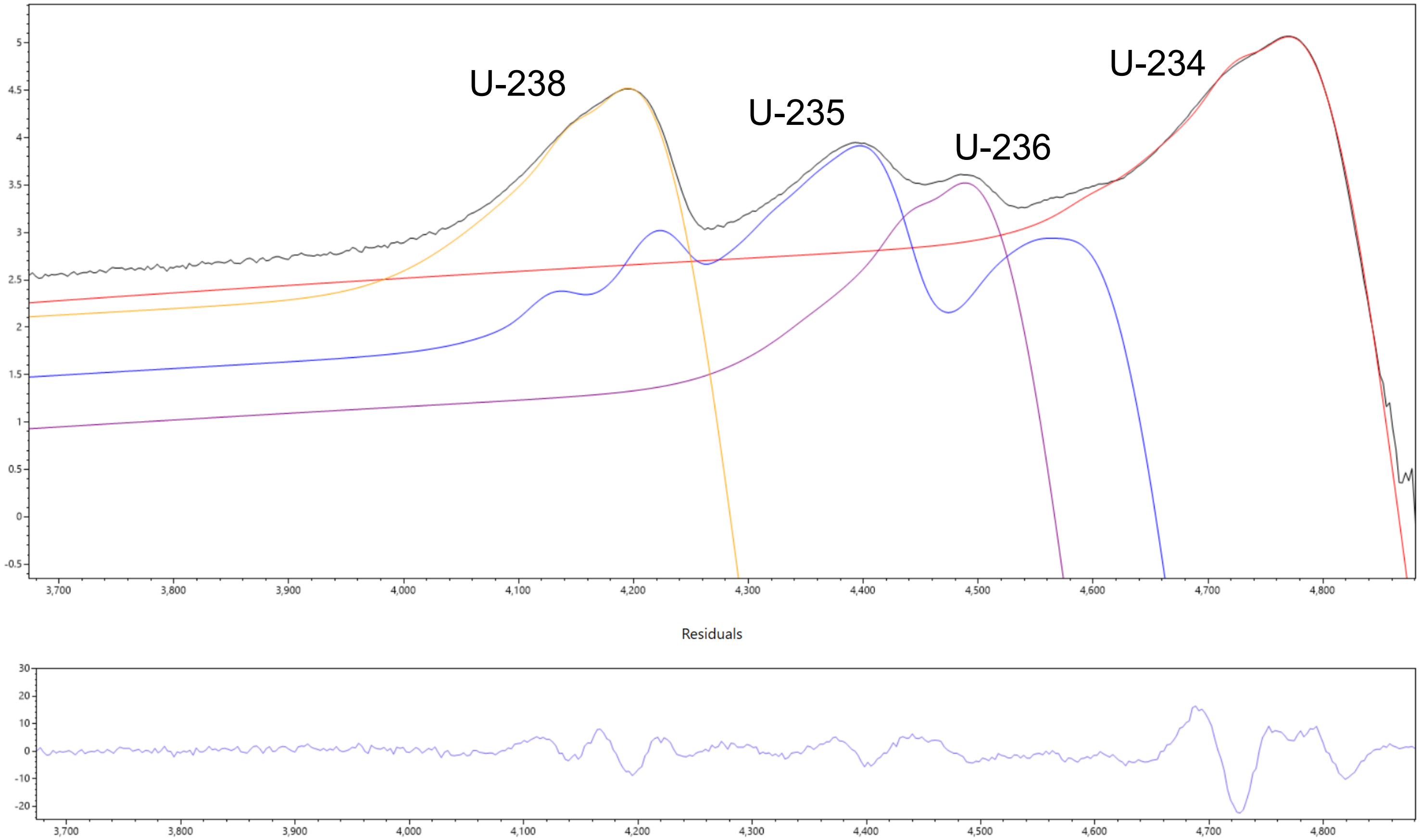
Fit U-235 at 2.428%

Using 5 LE Tails and 1 HE

Thin Single-Use Destructive Analysis (SUDA) Sample

U-235: 4.615002 % ± 0.001213



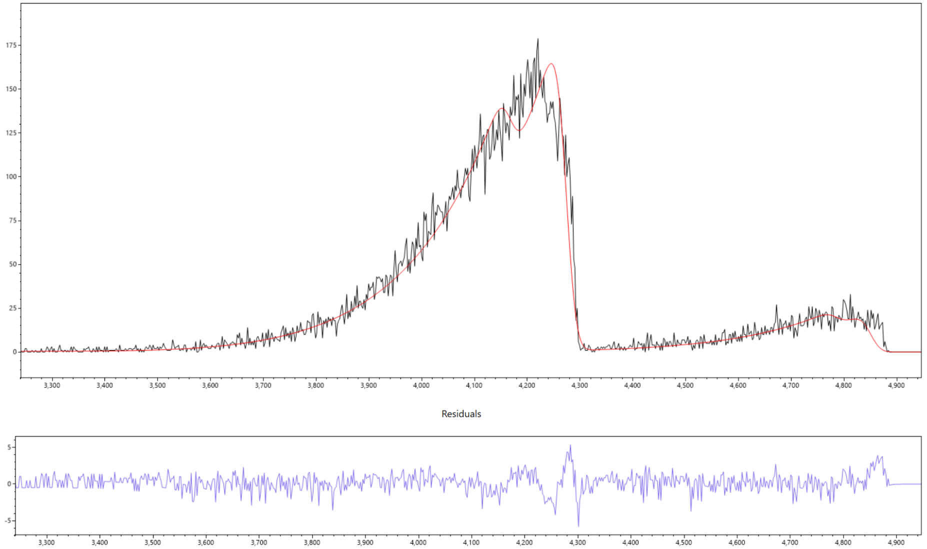


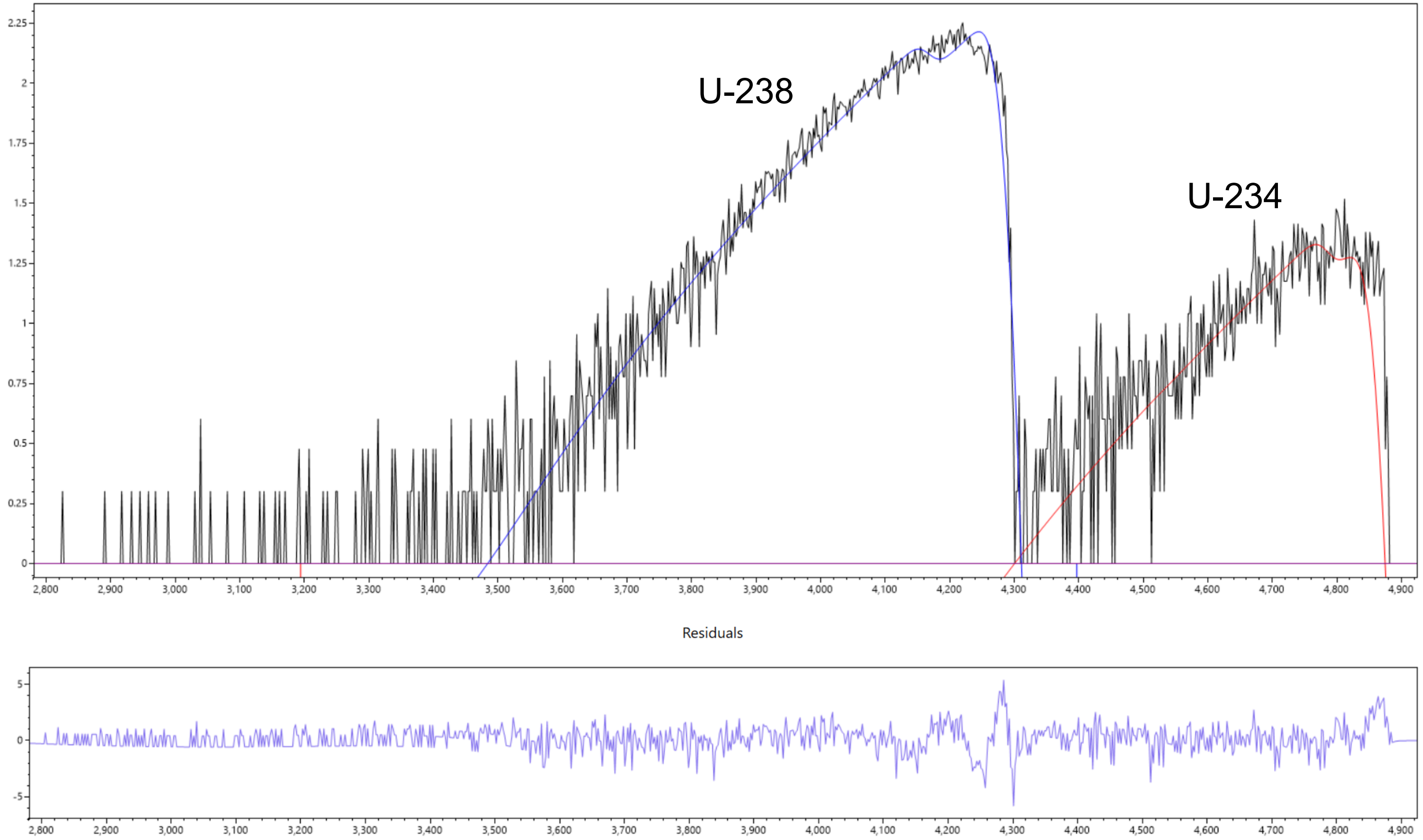
Fit U-235 at 4.788%

Using 5 LE Tails and 1 HE

Thick Single-Use Destructive Analysis (SUDA) Sample

Depleted Uranium

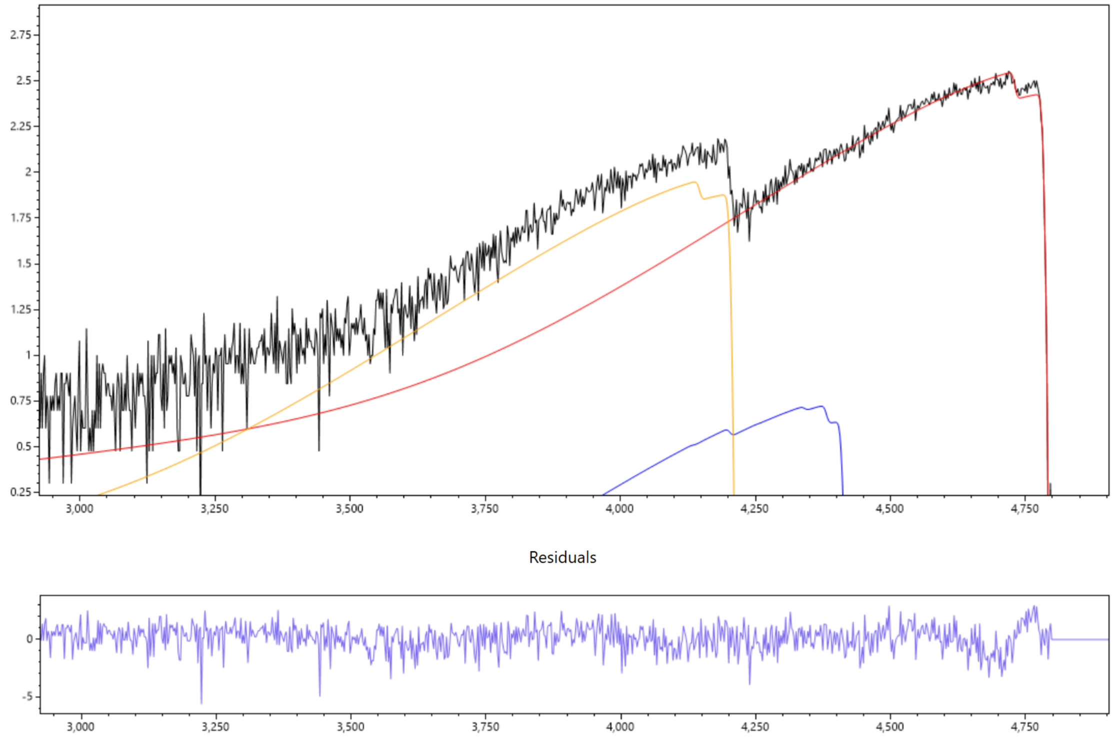




Fit U-238 at 99.999%

Using 5 LE Tails and 0 HE

Thick Single-Use Destructive Analysis (SUDA) Sample



4.621% U-235, Fit = 1.06%

Method 2: Genetic Algorithms (Just Getting Started)

- Most difficult step is using terminology from evolutionary biology
  - Fit Parameters -> Genes
  - Parameter Sets -> Chromosomes
  - A Group of Parameter Sets -> Generation
  - Chi-Squared -> Fitness Function
- Instead of chasing parameters in the direction of negative partial derivatives (and getting stuck in local minima) these algorithms mimic evolutionary processes to find an optimal solution
  - Natural Selection -> Which parameter sets make it to next generation (Elite, Stochastic, Tournament)
  - Crossover -> Using what is learned in a previous generation to create the next group of parameter sets
  - Mutation -> Random whoopsies that insure we do not get stuck in local minima

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