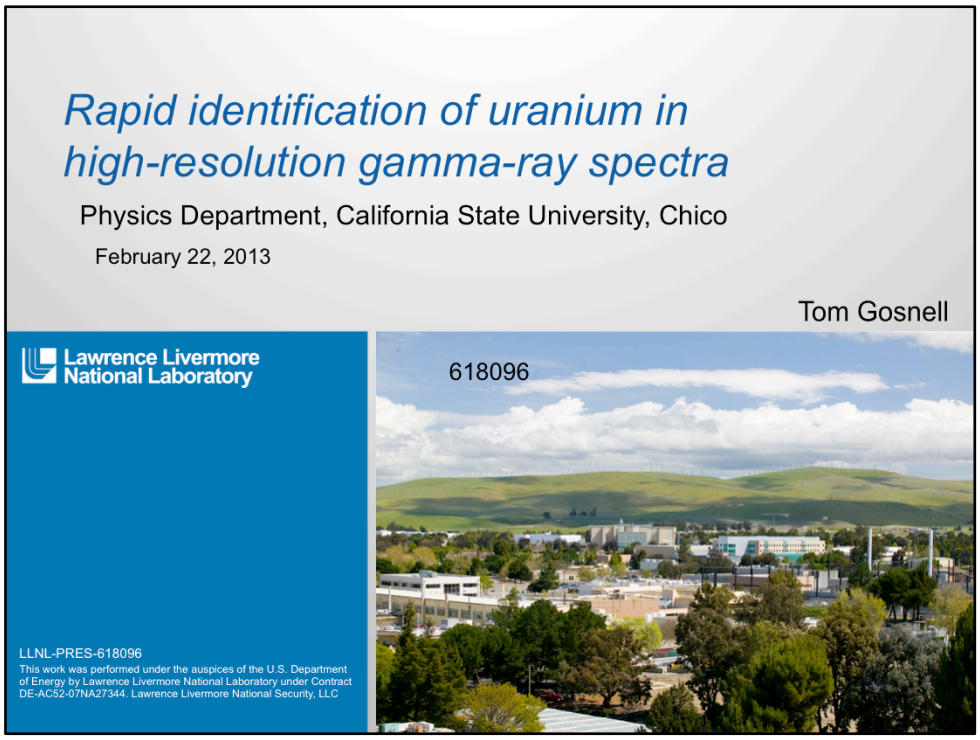


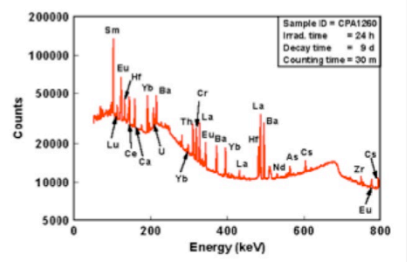
About the speaker

Tom Gosnell was born and raised in Chico. His association with Chico State began at the tender age of six when he was enrolled as a student in the Education Department's laboratory school, Aymer Jay Hamilton. After Aymer Jay, Tom attended Chico Senior High School. He then enrolled as a student at Chico State, with a major in physics. This delightful period ended with the impending jeopardy of his being drafted into the Army. Not fond of the notion of sleeping in rain-filled fox holes or the possibility of being involved in a war where he might be seriously killed, Tom enlisted in the Air Force. After four years in the service, Tom returned to Chico State where he graduated with a physics degree in 1967.

Tom soon married a former Chico State coed, who was then a career high-school biology teacher. They settled in the San Francisco bay area where Tom secured a position as a physicist at a radioanalytical laboratory in Richmond, now the Richmond branch of Eberline Services. There Tom learned some of the fundamentals of radiochemistry, about a great variety of radiation measurement techniques, and the development of computer programs to analyze radiation measurement data. He soon became the supervisor of the radiation measurement laboratory and and computer facility.

Tom then moved on to acquire a Master's Degree in engineering science at the Department of Nuclear Engineering at the University of California at Berkeley. Following that he became a physicist at the Lawrence Livermore National Laboratory, where, for 35 years, he has worked on the application of radiation measurements to problems in areas of national security.

Every gamma-ray spectrum tells a story if you can read its language



What we *can* read from this spectrum

1. Detector is high resolution
2. Probably measured 30+ years ago
3. Detector was small w/low efficiency
4. Detector was probably Ge(Li)
5. Detector was in a counting cave
6. Spectrum likely from Neutron Activation Analysis (NAA)
7. Possibly seeking a trace element
8. Intermediate spectra were likely
9. Scientist possibly a nuclear chemist

What we *can't* read from this spectrum

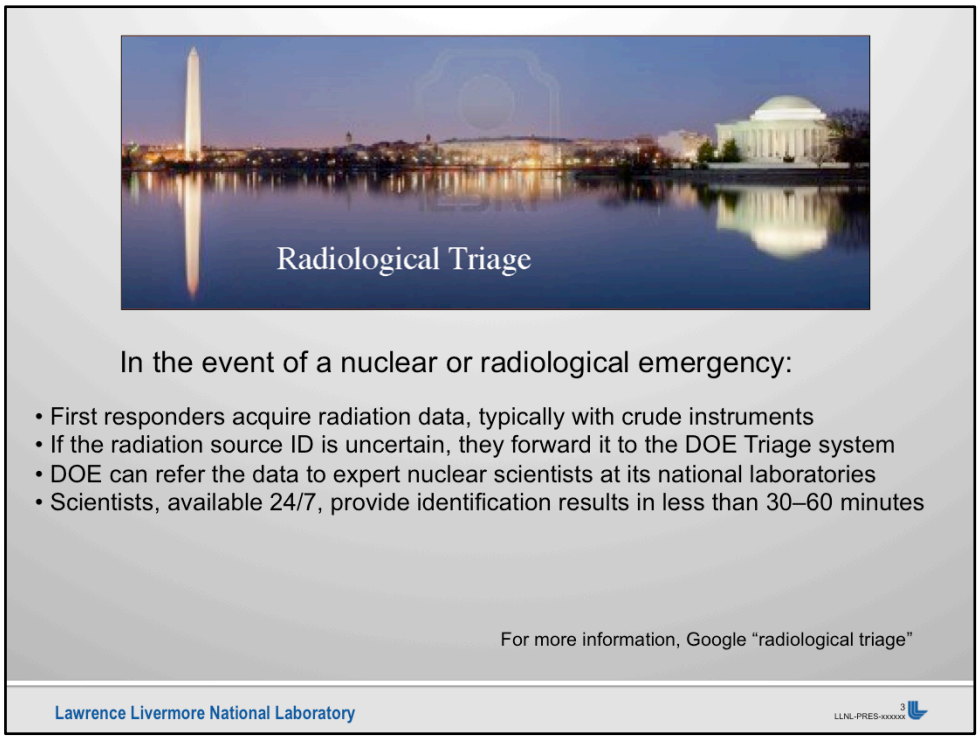
1. Sample provenance (Need more context)
2. What problem was the spectrum to solve? (Need more context)
3. Detailed analysis results (This analyst doesn't speak NAA)

Lawrence Livermore National Laboratory

LLNL-PRES-xxxxxx 2

What we can read from this spectrum

1. Detector is high resolution <= Sharp, narrow peaks
2. Probably measured 30+ years ago <= Time scale in minutes—used before ~1970
3. Detector was small with low detection efficiency <= Detector too old to have high efficiency
4. Detector was probably Ge(Li) <= High-Purity Ge (HPGe) now used since the late 1970s
5. Detector was in a counting cave <= No background peaks present, indicating a massive shield
6. Spectrum likely from neutron activation analysis (NAA) <= Sample was irradiated. There's a dog's breakfast of elements revealed in the spectrum that is not uncommon in NAA
7. Possibly seeking a trace element <= NAA. Sample was allowed to cool for 9 days allowing short-lived nuclides to decay sway so that weak, long-lived peaks could be revealed. Spectra may well have been taken at earlier times for the shorter-lived nuclides.
8. Intermediate spectra were likely <= IF NAA, THEN During the 9-day cooling time spectra may have been taken to analyze for other elements indicated by short-lived nuclides
9. Scientist possibly a nuclear chemist <= Elements are identified not nuclides. Scientist appears familiar with a sophisticated nuclear analysis method



From the Radiological Triage web site:

Google "radiological triage." [Minor additions and corrections are in brackets.]

Mission

The Triage mission is to provide secure on-line nuclear and radiological expertise to first responders within 30-60 minutes of receipt of data This integrated system provides essential time-sensitive information on the nature of the threat, allowing responders to develop and implement appropriate courses of action, and ensure the scope of the response is sufficient to provide for the health and safety of responders and the general public, without placing excessive and unnecessary demands on critical resources.

Steps in the Triage Response

• The NNSA's [National Nuclear Security Administration] 24-hour Watch Office receives the first call that an incident has occurred

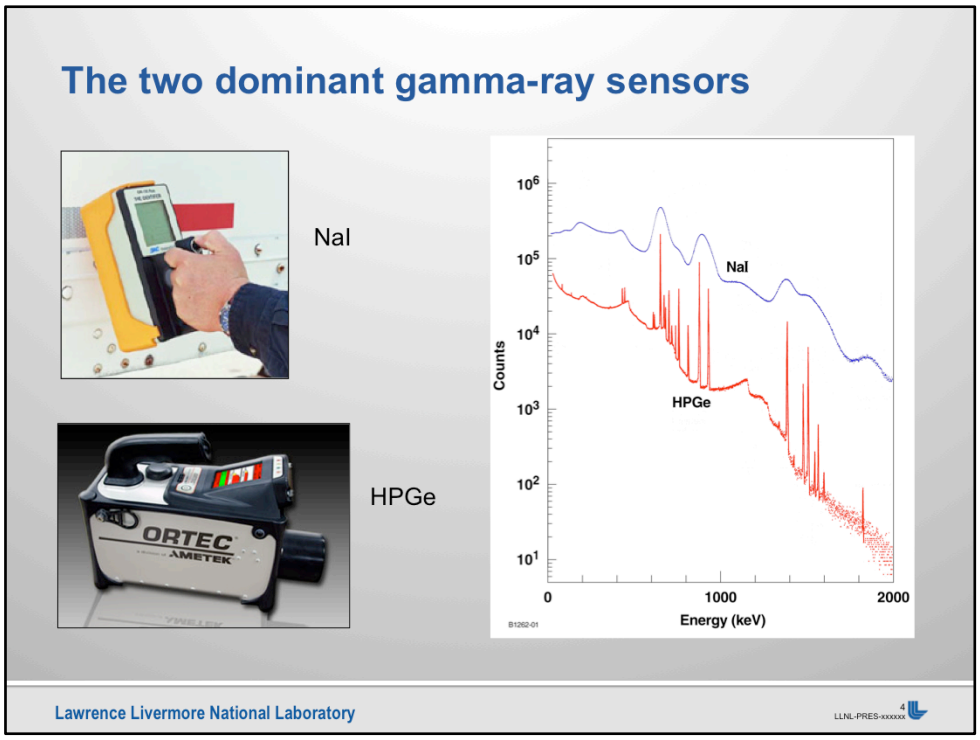
• he Emergency Response Officer (ERO) evaluates the situation, alerts the on-call scientist, and activates Triage. Also, the Radiological Assistance Program teams have the authority to activate the Triage

• Data can be transmitted in an unclassified format to the ERO or directly to Triage via the [Triage] website

• Data [are] collected

• Data analysis begins within 10 minutes of receipt

• Data results provided back to the field within 30-60 minutes



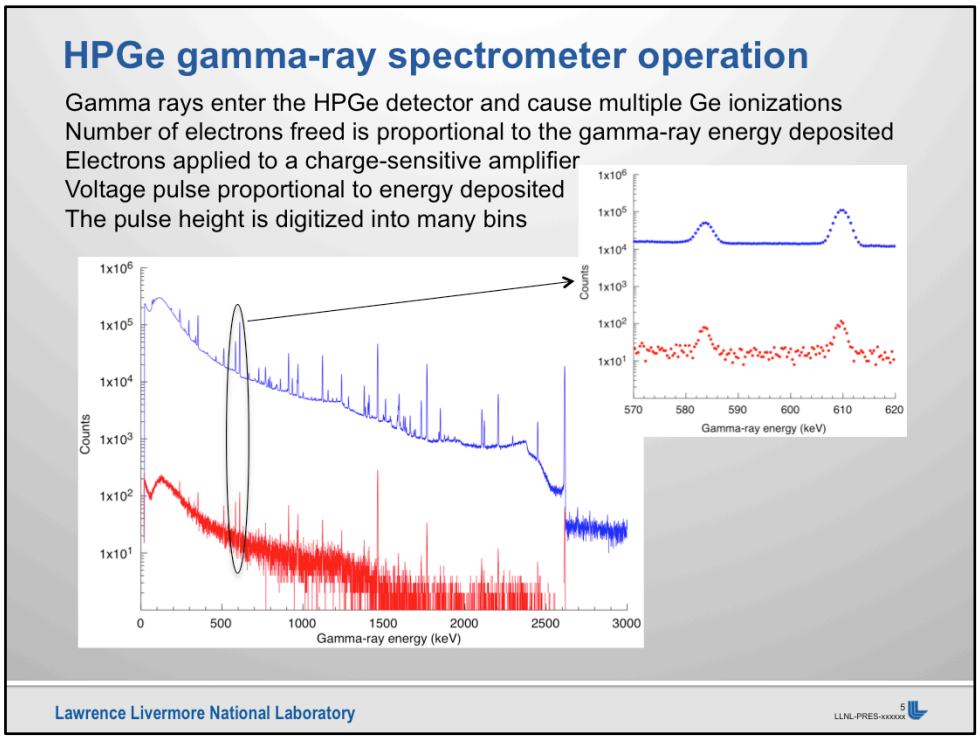
Low- and high-resolution gamma-ray sensors

The two dominant types of portable gamma-ray detection systems for field use employ either low- or high-resolution gamma-ray sensors. The detection system in the upper-left photo employs the most popular sensor used in the field, a sodium-iodide (NaI) scintillation crystal. The detection system in the lower-left photo employs a solid-state diode of High-Purity Germanium (HPGe).

Gamma-ray spectra taken with these two types of sensor are illustrated on the right. The spectra are from a mixture of silver isotopes ^{110m}Ag and ^{118m}Ag. ^{110m}Ag is an industrial calibration and gauging source and ^{108m}Ag is a byproduct of ^{110m}Ag production. There is a considerable mixing of gamma rays from each isotope in the spectra. Because of its superior information content [1], the detailed contributions from each isotope are resolvable in the Ge spectrum but not in the NaI spectrum. For this reason, HPGe excels in the analysis of complex spectra from sources of mixed radionuclides such as uranium.

Gamma-ray spectra collected by nuclear incident first responders are obtained in the field, usually under suboptimal conditions, for short periods of time, and can result in sparse data that cannot reveal the finest details. Nevertheless the superior energy resolution of HPGe allows it to detect uranium, discriminate between natural and processed uranium, and obtain a rough estimate of uranium enrichment.

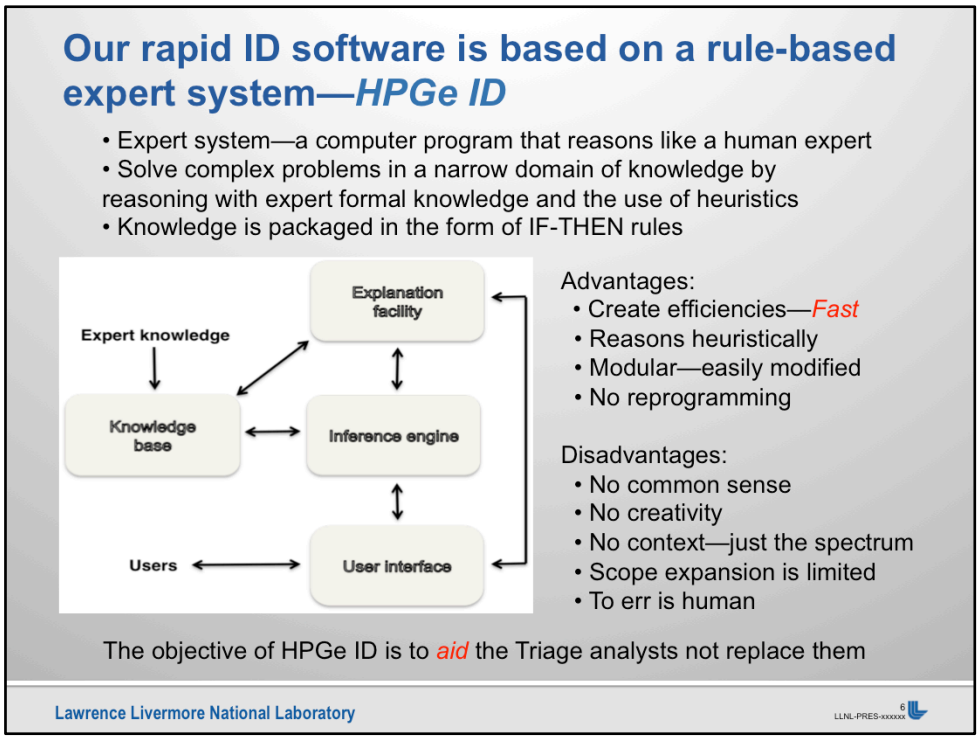
[1] Karl E. Nelson, Thomas B. Gosnell, David A. Knapp, *The effect of energy resolution on the extraction of information content from gamma-ray spectra*, Nucl. Instr. Meth. Phys. Research A **659** (2011) 207.



A gamma-ray spectrometer begins with an energy-resolving detector assembly that ultimately produces charge carriers whose number is proportional (or nearly so) to the amount of gamma-ray energy deposited in the detector. As each gamma ray interacts with the detector the charge carriers are electronically processed and converted into a voltage pulse with a height that is proportional to the number of charge carriers and thus the energy deposition as well. The pulse height is then digitized and one count is added to a computer-type memory register, called a *channel* that corresponds to a narrow energy range that is appropriate for that gamma ray. There are many contiguous memory channels (about 8000 in the figure) that together span the energy range of interest for the measurement.

As gamma ray data acquisition continues, each additional individual gamma ray adds a count to the channel that is appropriate for its amount of energy deposition. After a sufficient number of counts is acquired, the memory can be read out and the results plotted as a series of dots representing the number of counts vs. pulse height. The pulse-height axis is then calibrated to represent gamma-ray energy deposition. This plot is properly called a gamma-ray pulse-height distribution but is more commonly called a gamma-ray spectrum.

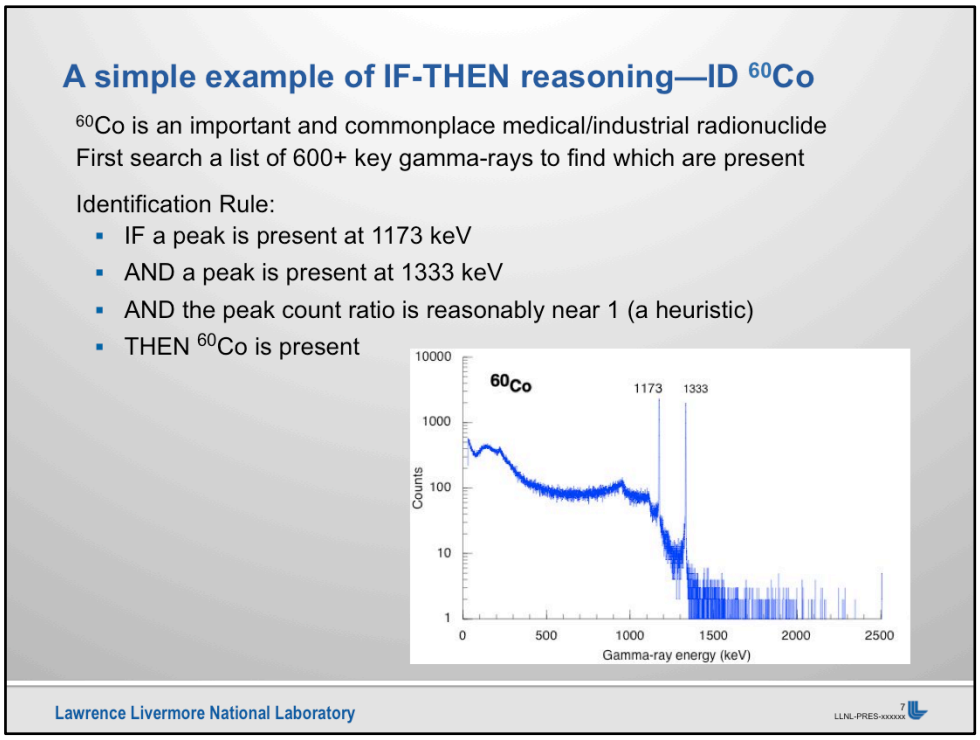
When gamma rays of a particular characteristic energy (often called lines) deposit their full energy in a detector, a peak will appear in the gamma-ray spectrum, *a full-energy peak* or *photopeak* that indicates full energy deposition. Gamma rays do not always deposit their full energy in the detector. They often Compton scatter, causing partial energy deposition, and the scattered gamma ray escapes from the detector. Pulses from partial energy deposition are found in a continuum (the *Compton continuum*) below the full-energy peak.



Rule-based expert systems

Because of the severe time constraints placed on Triage analysis and the nature of field gamma-ray spectra, we implemented *HPGe ID* as a rule-based expert system—a knowledge-based approach rather than an algorithmic approach. Another characteristic of expert systems is that they are heuristic—like humans they reason with expert judgmental knowledge and rules of thumb as well as formal established knowledge. As such *HPGe ID* follows the typical reasoning process used by gamma-ray spectrometrists.

As experience with these field spectra deepens, we need to incorporate newly gained knowledge of their characteristics in a relatively painless manner. An expert system differs from traditional programs by having a unique structure. It is modular, divided into two parts: a knowledge base or rule base that can be easily modified or expanded and an inference engine that reasons with the rules.

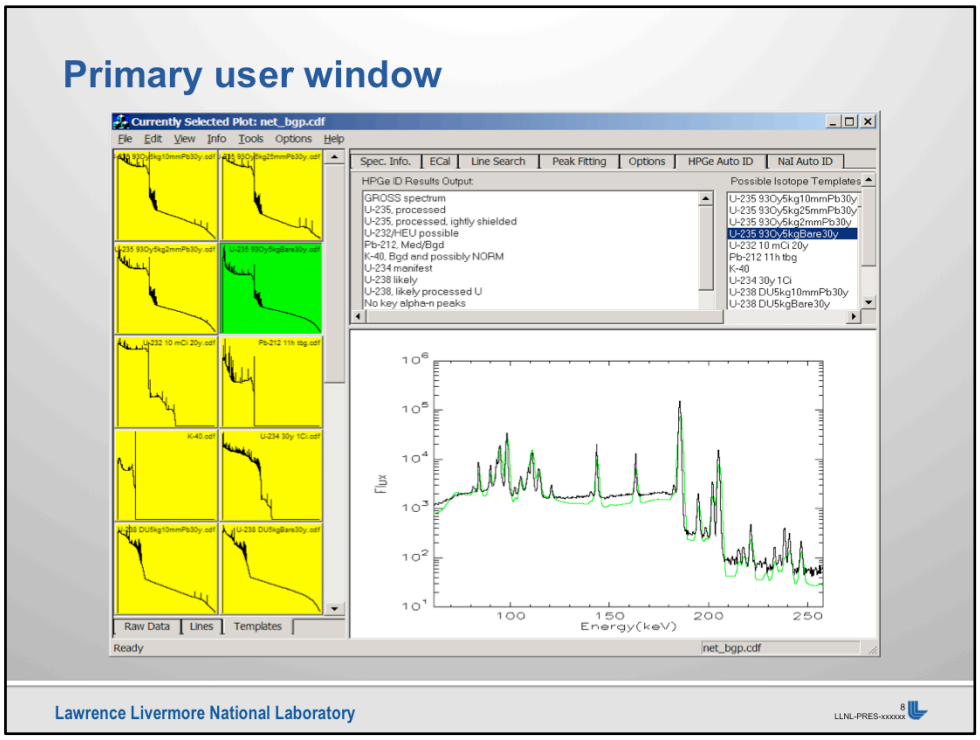


A rule-based expert system is one whose knowledge base contains the domain knowledge coded in the form of IF-THEN rules. *HPGe ID's* domain is radioactive sources that can be encountered in commerce, with a focus on fissile materials.

The analysis of high-resolution gamma-ray spectra is based on photopeaks in the spectrum—their energy and number of counts. *HPGe ID* first applies rules to assemble a working memory to store a database of facts that will later be used as antecedents by further rules. The *HPGe ID* scans the spectrum and stores a Boolean value (TRUE or FALSE) in the database for the presence or absence of 600+ key gamma ray peaks. It also stores the values of the number of counts found in the scan for presence/absence.

To invoke this process the user drags and drops the icon for the gamma-ray spectrum file onto the *HPGe ID* user interface. *HPGe ID* then automatically begins the spectrum scan.

1. *HPGe ID* proceeds using ladder logic. When safely climbing a ladder, ascension to higher rungs depends on successfully ascending each lower rung in turn. The first rung on HPGe ID's ladder is the identification of the presence or absence of key peaks.
2. The second rung is to compute and store store in the database ratio values of the counts in selected gamma-ray photopeak pair. Depending on whether or not these ratios satisfy rule thresholds, a Boolean value, TRUE or FALSE, is assigned to the ratio.
3. The third rung on the ladder is the identification of the likely presence or absence of radionuclides by characteristic photopeak combinations.
4. The fourth and final rung is a report test that includes a Boolean value of TRUE for the nuclide plus other conditions, such as ratios, that must be satisfied to identify the source or determine important characteristics associated with the source, such as uranium enrichment. The TRUE reports are reported in descending order by their their signal-to-noise ratios

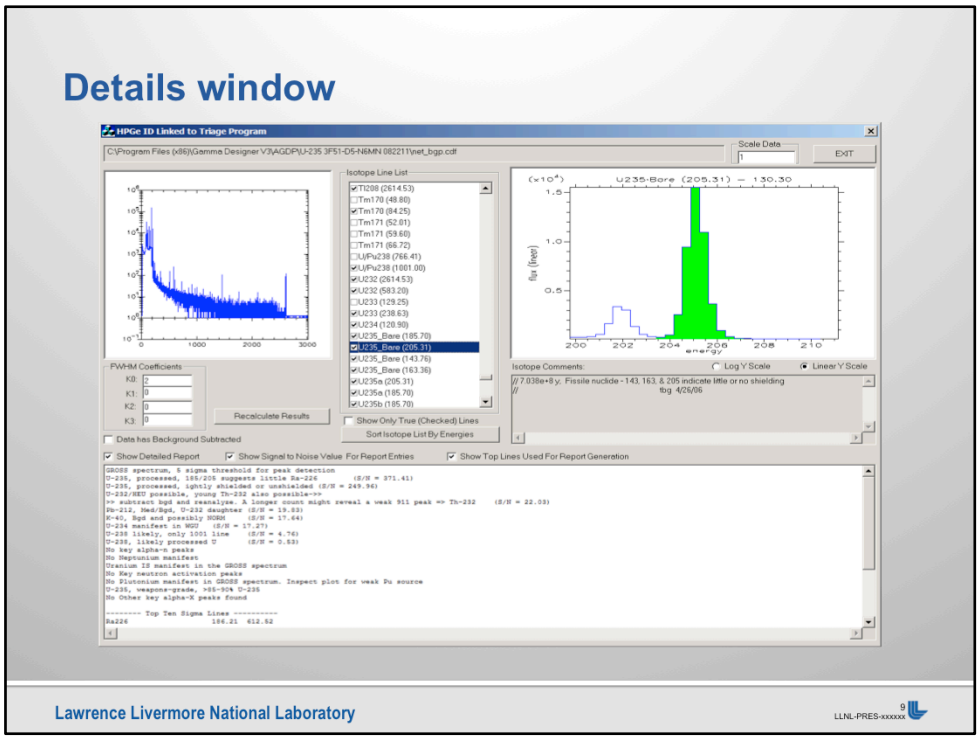


The *HPGe ID* user interface—Primary window

The *HPGe ID* primary user window. In this figure, the user has dragged and dropped a spectral data file in the pulse-height analyzer's proprietary format onto the primary viewer pane. *HPGe ID* translated the proprietary format into a common internal format, displayed the spectrum in the primary viewer pane.

Abbreviated and prioritized result findings appeared in the upper middle pane, indicating that the spectrum is from weapons-grade uranium. A number of candidate spectral templates appeared in the upper right-hand pane for the user to compare with the data. Spectral thumbnails of these candidates are on the left-hand side, shown in yellow, with the exception of the currently displayed template shown as a green thumbnail.

These events unfolded within less than one second. The user has since zoomed the primary view to the first 250-keV of the spectrum and sampled a variety of candidate templates for comparison to the measured data, finally settling on a template of unshielded 93% ^{235}U, shown in green.

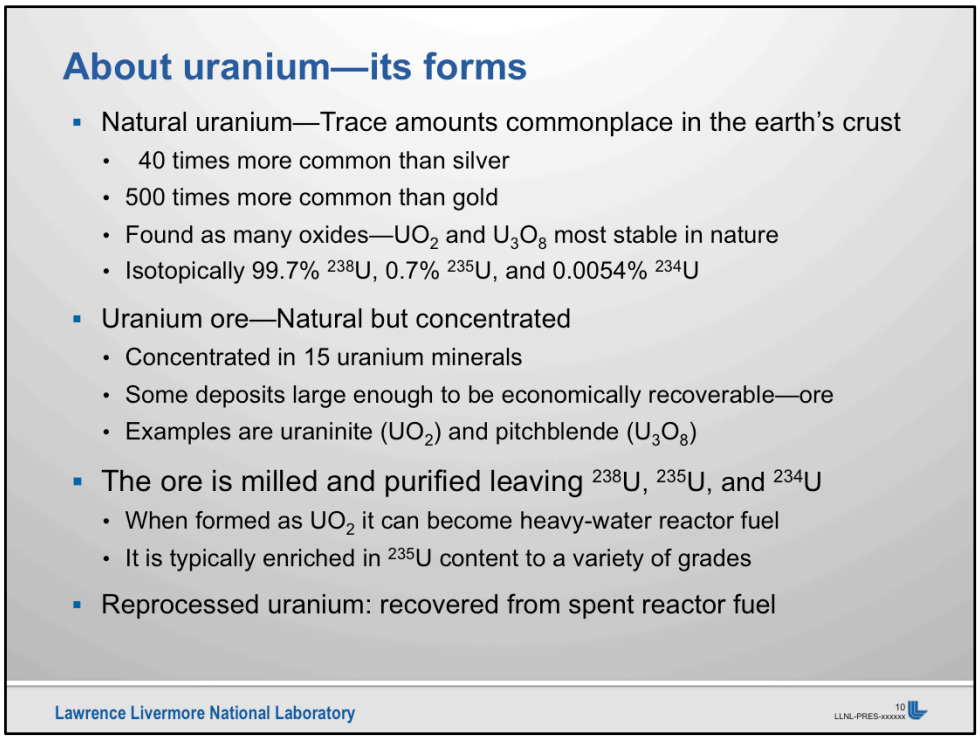


The *HPGe ID* user interface—details window

The *HPGe ID* details user window. In this figure the detailed findings appear in the bottom pane. These include expanded textual results from the primary window. They are prioritized by descending signal-to-noise ratio, S/N, as defined in the Appendix. Finally, they are followed by summary results indicating the presence or absence of uranium, plutonium, neptunium, common neutron activation activity, common alpha-n activity, and other common alpha-X activity.

A thumbnail display of the entire spectrum appears in the upper left pane. The upper center pane reveals the 600+ lines that *HPGe ID* searches for and lists them alphabetically by associated element and mass number. Small boxes to the left of the line descriptions contain checks if the line is an initial candidate for identification. Below the window is a button that will re-sort the lines by gamma-ray energy.

A close-up single-line view is shown in the upper right pane for the 205-keV ^{235}U line, chosen by the user from the isotope line list. The green shading indicates the spectral region used for identification of this peak. Below the line-view pane is a description of the nuclide's provisional identification, its half-life, categorization (e.g. medical, industrial, fissile, research, or impurity) and, usually, its uses/applications.



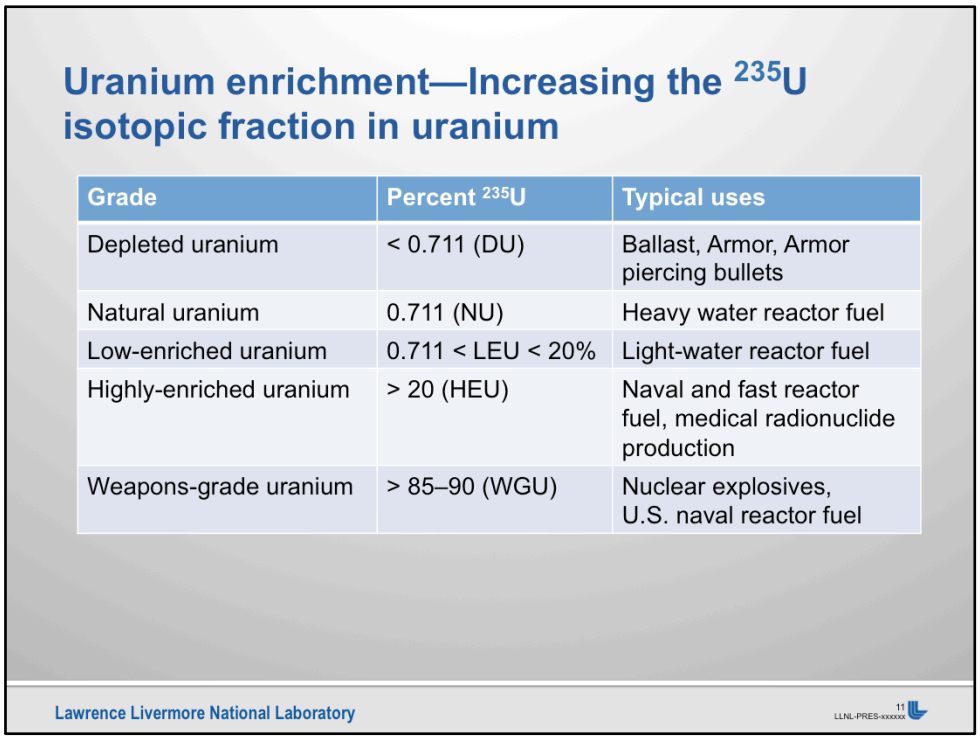
Reprocessed uranium

Reprocessed uranium is the uranium recovered from nuclear reprocessing, as done commercially in France, the UK and Japan and by nuclear weapons states' military plutonium production programs. This uranium actually makes up the bulk of the material separated during reprocessing. Commercial light-water reactor spent fuel contains on average (excluding cladding only four percent plutonium, minor actinides, and fission products by weight. Re-use of reprocessed uranium has not been common because of low prices in the uranium market of recent decades, and because it contains undesirable isotopes of uranium.

During its irradiation in a reactor, uranium is profoundly modified. The composition of reprocessed uranium depends on the initial enrichment and the time the fuel has been in the reactor. A typical vector of uranium isotopes in reprocessed uranium, with attributes, is listed in the Table.

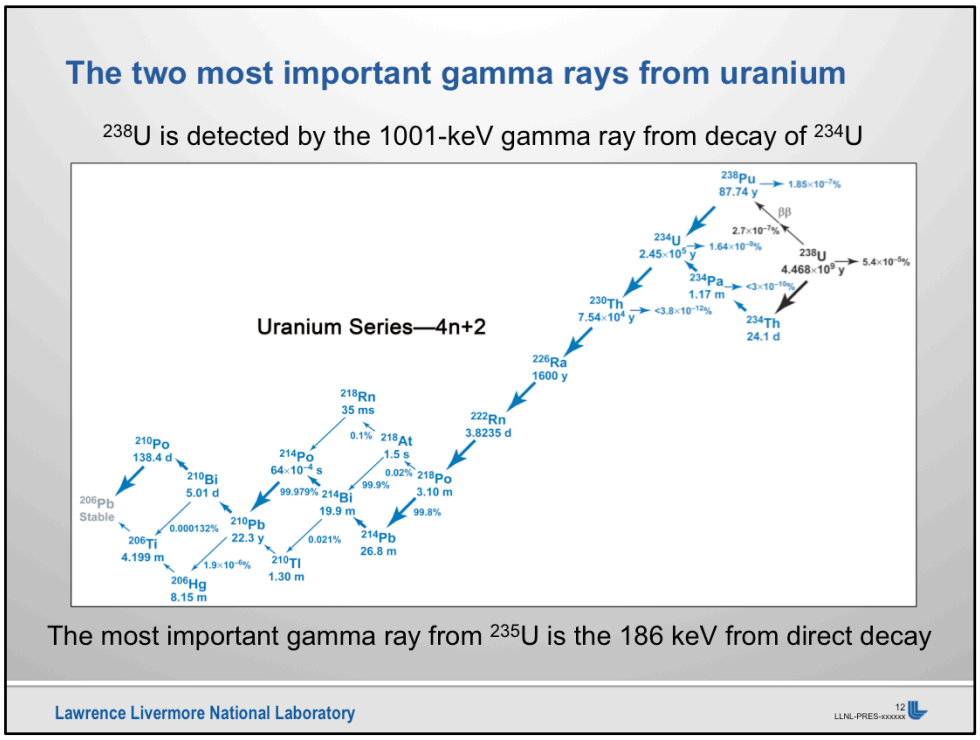
Typical isotopic composition of reprocessed uranium

Isotope	Proportion (%)	Half-life	Characteristics
U-238	99	4.47e9 y	Fertile material
U-237	~0.001	6.75 d	Decays rapidly to Np-237
U-236	0.4–0.6	2.34e7 y	Neutron absorption lowers reactivity, also produces Np-237
U-235	0.4–0.6	7.04e8 y	Fissile material
U-234	>0.02	2.46e5 y	Fertile material, neutron absorption increases reactivity
U-233	trace	1.59e5 y	Fissile material
U-232	trace	68.9 y	Decay product Tl-208 emits strong gamma radiation making handling difficult



Enriched uranium

When processed, uranium is chemically purified, removing the daughters of ^{235}U and ^{238}U, except for its ^{234}U daughter. Processed uranium is typically enriched in the isotopic fraction ^{235}U. If not enriched, the processed uranium can be made into heavy-water reactor fuel. It will have retained its natural isotopic vector and, although processed, is confusingly referred to as natural uranium (NU). The tailings of the enrichment process, depleted uranium (DU), are reduced in their ^{235}U content. Uranium enrichment grades are shown in the Table.



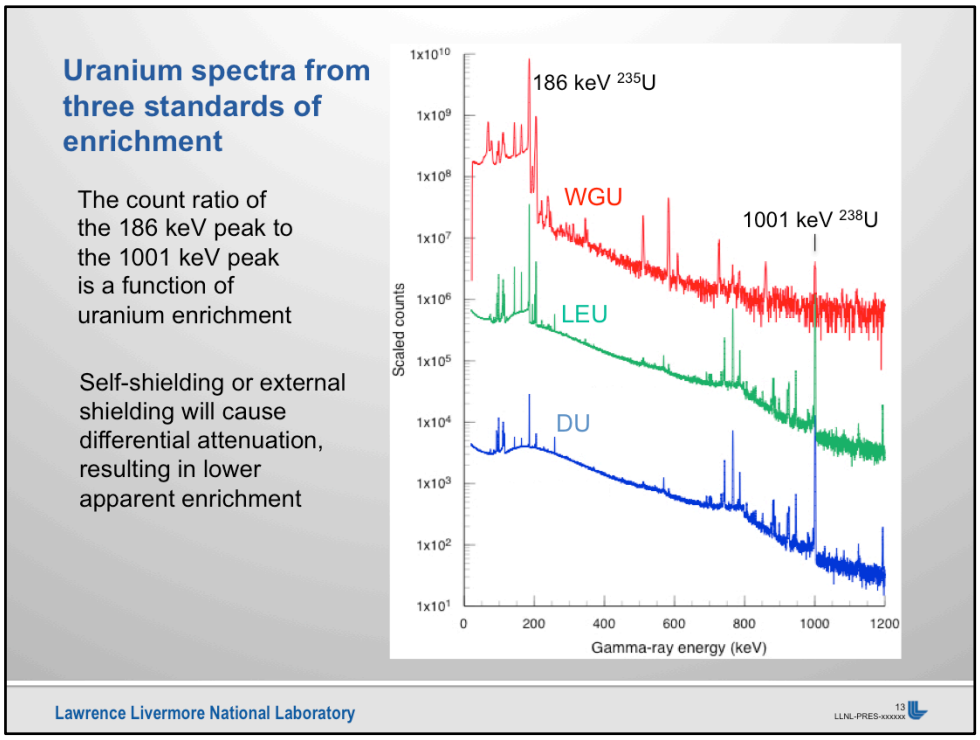
The two most important gamma rays from uranium

Because we are interested in uranium enrichment, the two most important gamma rays from uranium for our purposes are the 1001.03-keV gamma ray from the decay of the ^{234m}Pa granddaughter of ^{238}U and the 185.7-keV gamma ray from direct decay of ^{235}U.

Shown in the figure is the uranium decay series. It is one of the four important heavy element decay series. Three of these, residing in every spade-full of soil, produce much of the bulk of background gamma radiation. Each of the three series has as it's progenitor, a primordial radionuclide, present when the solar system was formed. For the uranium series, ^{238}U, shown in black, is the primordial progenitor.

The ^{234}Th daughter of ^{238}U decays with the relatively weak (2.8 %) emission of a 92.4-keV gamma ray that is easily shielded. This decay populates a long-lived (1.17-m) excited state of ^{234}Pa that beta-decays directly to an excited states of ^{234}U, one of which emits a 1001.03-keV gamma-ray. The emission intensity of this gamma ray is low (0.38 %) but it is very penetrating and is the single most indicative gamma ray for the presence of ^{238}U

We digress here to note that all if the ^{238}U daughters have substantially shorter half-lives than ^{238}U itself. If left undisturbed in the earth's crust for many thousands of years, the daughters will grow into secular equilibrium and decay at the the same rate as ^{238}U. The presence of ^{238}U in background radiation is most notable by the many gamma ray emissions from its distant decay daughters, notably ^{214}Pb and ^{214}Bi. While secular equilibrium is largely present in uranium ore, during geological processes such as erosion, sedimentation, melting, or crystallization, different nuclides in the decay series can become fractionated relative to one another, due to variations in their chemistry or the structural site they occupy. These short half-lives are useful for dating Pleistocene geological events that are too old to be well resolved by radiocarbon dating and too young to be well resolved by methods employing longer half-lives.



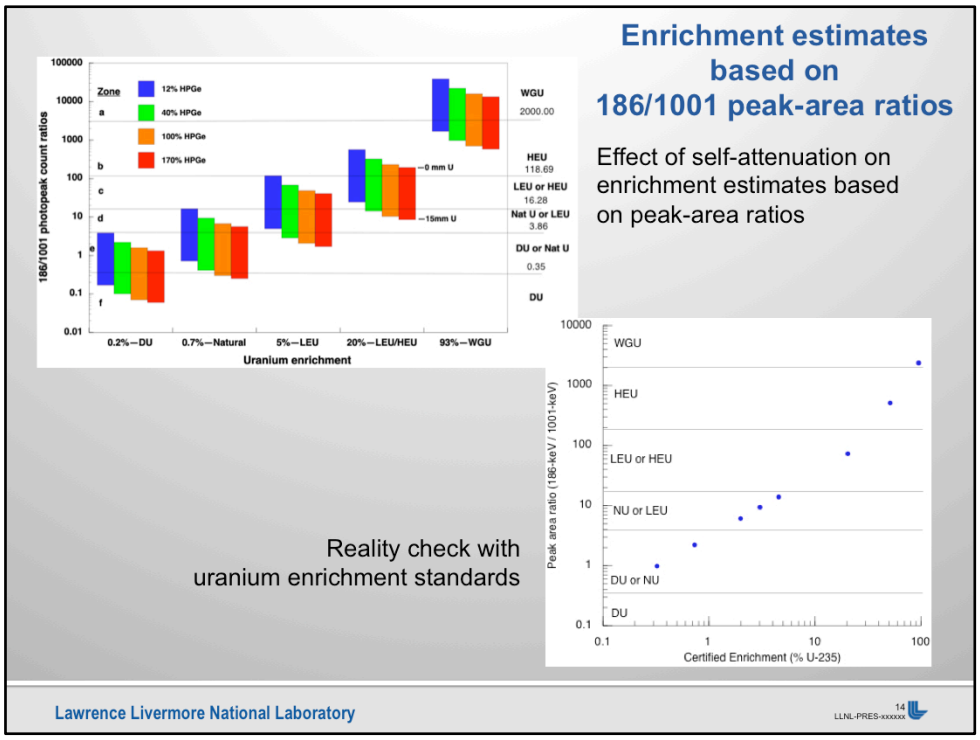
Figure—Uranium spectra from standards of three different enrichments are displayed. The top two spectra have been scaled to separate them for presentation. All three were acquired with HPGe detectors of 20% relative efficiency at a source distance of 100 cm. The lower two spectra were counted for 7000 seconds. The upper spectrum, a weaker source, was counted for only 900 seconds.

Minimum enrichment estimation by peak area ratio

In principle, uranium enrichment can be determined by measuring the ratio of counts in the 186-keV ^{325}U peak and the 1001-keV peak from the ^{234}Pa granddaughter of ^{238}U. In the uranium spectra shown in the figure, the increase in the height of the 186-keV ^{235}U peak relative to the 1001-keV ^{238}U peak as a function of increasing enrichment is readily apparent.

In the early 1970's an effort was made to apply this ratio to obtain high precision, high accuracy results. It was not found to be fruitful for this purpose because certain required information was unavailable for field use. It has also been observed that although the 186- and 1001-keV peaks are easy to measure, it is difficult, because of the large difference in their energies, to determine the relative efficiencies with which they are detected. Furthermore, if there is material intervening between the uranium and the detector, the 186-keV peak will be preferentially attenuated relative to the 1001-keV peak, resulting in an enrichment estimate that is too low, a minimum estimate

Our goal is considerably less ambitious in terms of precision and accuracy. We do, however, wish to make at least a rough estimate of minimum enrichment grade. The task is challenging: to be able to make this enrichment estimate with an automated analysis tool that doesn't require knowledge of the nature of the source, the source-to-detector measurement distance, or the efficiency of the detector.

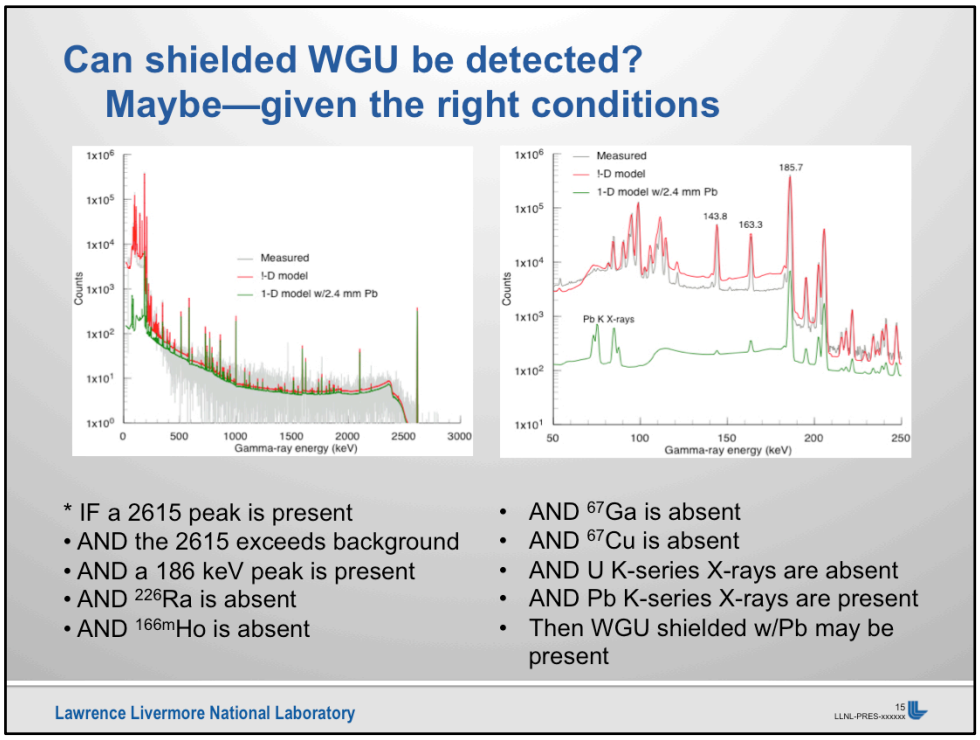


Minimum enrichment estimation by peak-area ratio

The 186/1001-keV peak-area ratio can be used to obtain a rough estimate of uranium enrichment. However, If a significant amount of attenuating material is between the source and the detector, differential attenuation of the two gamma rays will result in erroneously low enrichment estimates. This is because the lower the gamma-ray's energy the more easily it is attenuated. The large energy difference between the 186-keV gamma ray and the 1001-keV gamma-ray can make this differential attenuation significant with a concomitantly erroneously low enrichment estimate.

There are two possible sources of attenuating material. The first is an external absorber with typically unknown properties in Radiological Triage measurements. The second is self-attenuation in uranium itself. Because of the unknown nature of any external attenuators, we can do nothing about estimating their effects. On the other hand, we can calculate a simple self-attenuation model of a line source on the detector axis with an intervening slab attenuator. Gamma rays will be attenuated exponentially at a function of their energy and the linear attenuation coefficient of the absorber, uranium for a particular gamma-ray energy. This function can be integrated to estimate the attenuation for arbitrary thicknesses of uranium for any given energy.

Another source of differential response is the detector itself. Large detectors are more efficient than small detectors, particularly at high energies such as 1001-keV. We calculated the self-attenuation for the 1 86- and 1001-keV gamma rays and took their ratios for five selected enrichments for detectors with a broad range of relative efficiencies and displayed them on a floating bar chart. We did this for no attenuation and attenuation through a maximum uranium thickness of 15 mm .We then separated the ratio values by eye into six generally overlapping zones (left figure). Finally, we obtained enrichment estimates by peak-area ratio for eight unshielded certified enrichment standards to validate the method (right figure). It works.



Detecting shielded HEU

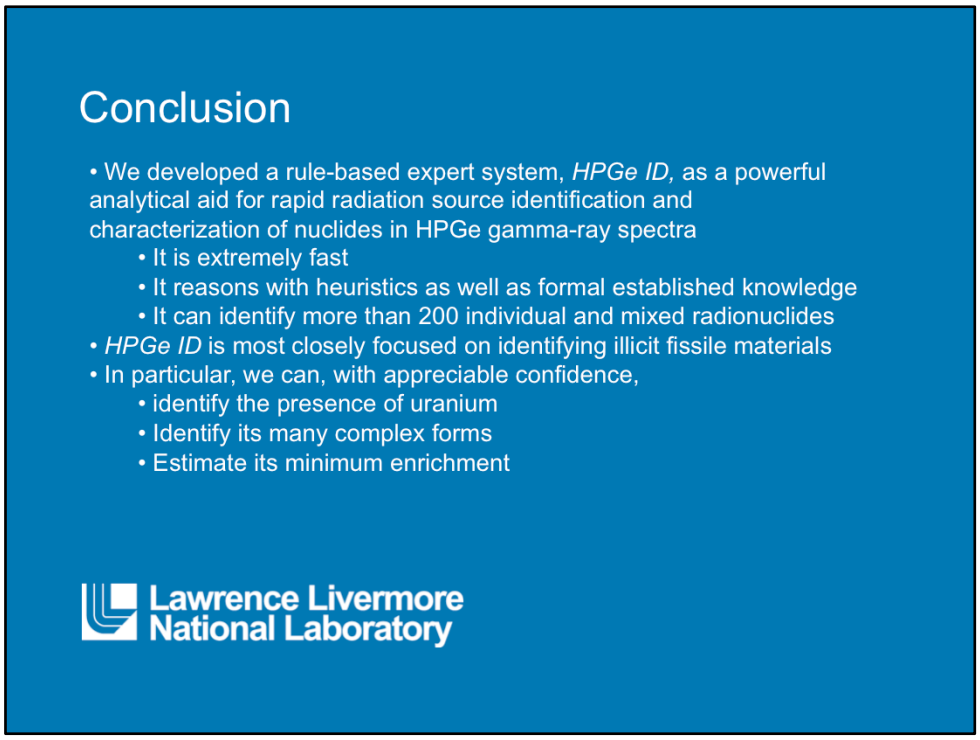
Because it can be used in the manufacture of nuclear explosives, illicit traffic of HEU is a matter of serious concern. Materials intervening between the detector and an HEU source will preferentially reduce the ^{235}U signal, eliminating or greatly reducing the ability to make roughly accurate enrichment estimates by the peak ratio method. An alternative surrogate HEU signature might be that of ^{232}U.

Uranium-232: grasping at straws to detect shielded HEU: With only a 69-year half-life, ^{232}U does not occur in nature but is introduced by reactor irradiation of uranium resulting in a number of complex reaction and decay chains that produce ^{232}U. During enrichment, ^{232}U is swept into the low-mass fraction along with ^{235}U. As a result, the greatest amount of ^{232}U is found in WGU and can be a surrogate for the presence of HEU and, particularly for WGU.

Direct radiation from ^{232}U are too weak to detect. Consequently, we rely on yet another surrogate, ^{208}Tl a distant radioactive daughter of ^{232}U. The decay of ^{208}Tl results in the emission of several gamma rays with strong intensities. Of most interest is the highly penetrating gamma ray at 2614.53-keV with 99% emission intensity. While this gamma ray is readily detectable from WGU, its presence is not unique, so that, without elimination of other possible sources of this radionuclide, it can lead to misidentification of shielded HEU. The most likely interference is the ^{208}Tl signature from background radiation. Counts in the 2615-keV peak in excess of background may be an uncertain indicator of the presence of WGU.

In WGU, the 1001-keV peak from ^{238}U may be too weak to be reliably detectable in WGU but the 186-keV ^{235}U peak that is more than 2000 times as intense may still be measurable when shielded. Therefore if the counts in the 2615-keV peak exceed background, the 186-keV ^{235}U peak can be measured and peaks near 186-kev are absent then the presence of WGU and be inferred. But can such a spectrum possibly exist?

We determined that a 2.4-mm Pb shield could reduce the apparent enrichment by peak-area ratio to 20% ^{235}U. We then computed the HPGe spectra of unshielded WGU and WGU shielded with 2.4-mm of Pb and submitted both to the peak-area test and found that the shielded spectrum gave a peak-area ratio result of LEU or HEU or shielded WGU. The calculations shown in the figures do not include background radiation or spectrum jitter from counting statistics, so rule given in the figure would likely produce doubtful results.



Conclusion

Uranium in its variety of forms, enrichments, and appearance in two reactor fuel cycles presents a challenge to rapidly and unambiguously identify its presence and its qualitative attributes from field measurements. We began this presentation with a sketch of the behavior of our expert system that exploits heuristics to produce these results for uranium and similar, but less complicated. results for more than 200 other radioactive sources. Following that description, we examined some salient radiation signatures of important uranium radioisotopes and their daughters.

HPGe ID is most closely focused on identifying illicit fissile materials. However, our goal was to expand its original limited scope to be able to identify more than 200 other radioactive sources. As a result, at its inception, this expanded version of *HPGe ID* was plagued with frequently occurring misidentifications. For a period extending over nearly a decade we have reduced the frequency of misidentifications to the point that the application is now highly valued by its users. Nevertheless, *HPGE ID* continues to be a work in progress.