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Title:	The Nuclear Fuel Cycle and Associated Gamma-Ray Signatures
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The Nuclear Fuel Cycle and Associated Gamma-Ray Signatures

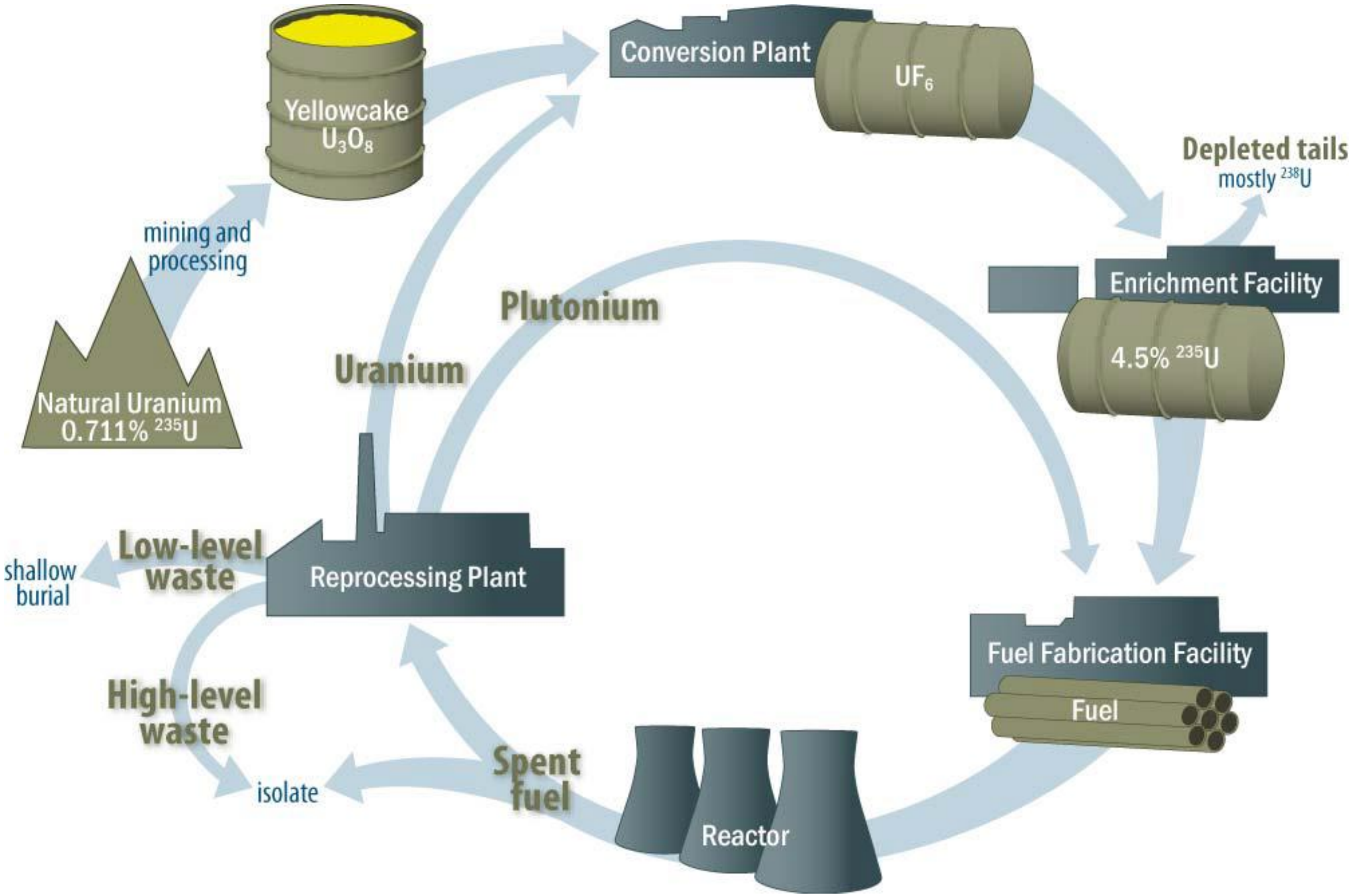
Los Alamos National Laboratory

LA-UR-xx-xxxx

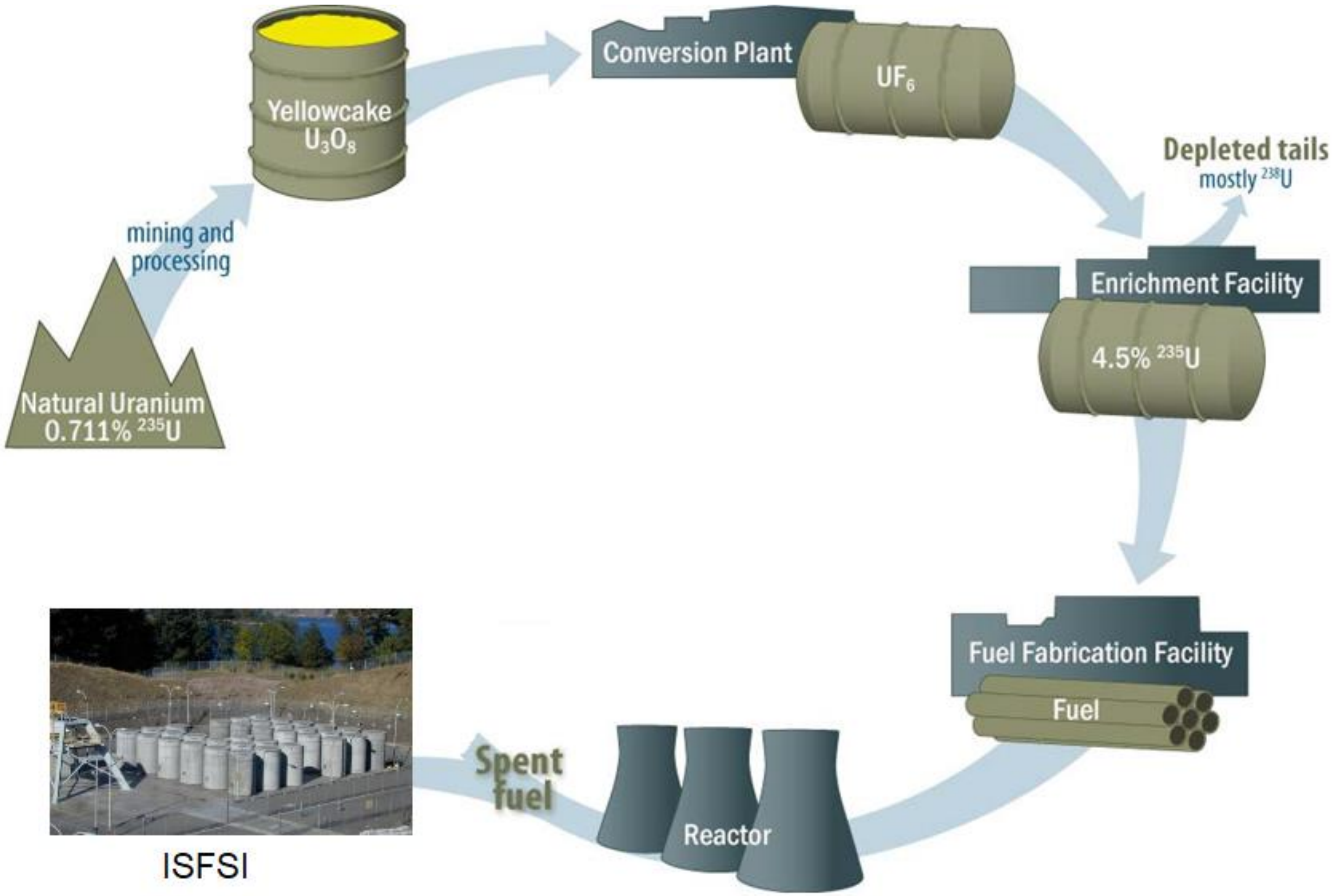
Introduction

- The nuclear fuel cycle concerns the life of nuclear material:
  - extraction from the earth in raw form
  - processing and enrichment
  - use in reactors or weapons
  - reprocessing or disposal
- Throughout the process, signature of the material changes
  - radiation
  - appearance

The Uranium Cycle



The Uranium ~~Cycle~~ Line



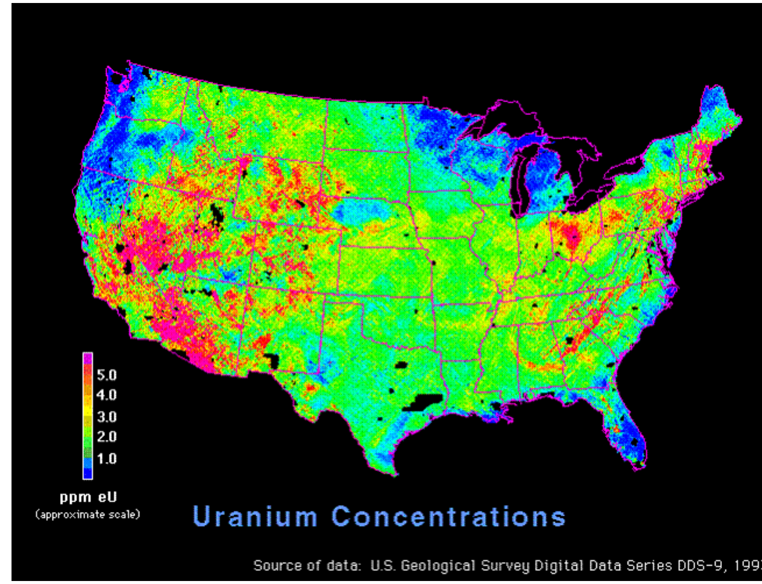
Definition of Uranium Enrichment

- Only three isotopes of uranium are found in nature
  - ^{238}U (99.27%)
  - ^{235}U (0.72%)
  - ^{234}U (0.006%)
- Categories of Enrichment (E = % of ^{235}U)

– Depleted Uranium (DU)	$E < 0.72\ \%$
– Natural Uranium (NU)	$E = 0.72\ \%$
– Enriched Uranium	$E > 0.72\%$
• Low Enriched Uranium (LEU)	$0.72\% < E < 20.0\ \%$
• High-Assay Low Enriched Uranium (HALEU)	$5\% < E < 20\%$
• High Enriched Uranium (HEU)	$E \geq 20.0\ \%$

Uranium Mining

Uranium is naturally occurring at about 1.8 - 2.7 ppm in the earth's crust. U content in ores can range from ~0.02 to ~20 %.



Uranium ore can appear in many different forms, from the primary mineral uraninite, to the colorful secondary minerals shown below.

Uraninite (UO_2 / U_3O_8) "pitchblende"

Torbernite $Cu(UO_2)_2(PO_4)_2 \cdot 12\ H_2O$

Carnotite $K_2(UO_2)_2(VO_4)\ 3H_2O$

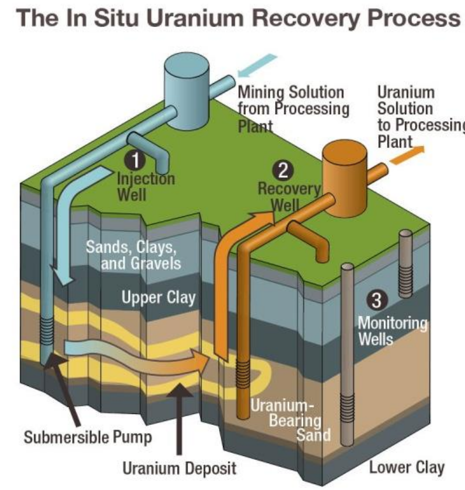
… 'ore', it could look just like a rock

http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Mining-of-Uranium/Uranium-Mining-Overview/

Uranium Mining

Mining Methods:

- Open Pit
- Underground
- In Situ Leach (ISL)

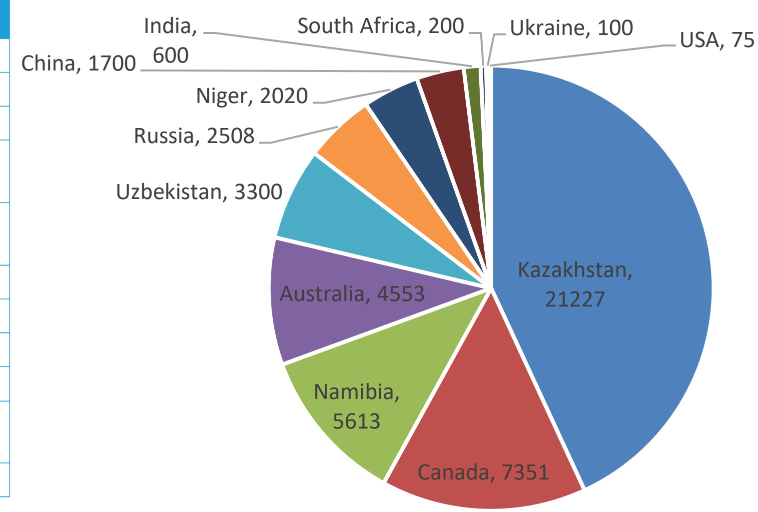


Top Ten Uranium Mines 2022

Mine	Country	Main owner	Type	Production (tonnes U)	% of world
Cigar Lake	Canada	Cameco/Orano	underground	6928	14
Husab	Namibia	Swakop Uranium (CGN)	open pit	3358	7
Inkai, sites 1-3	Kazakhstan	Kazatomprom/Cameco	ISL	3201	7
Olympic Dam	Australia	BHP Billiton	by-product/underground	2813	6
Karatau (Budenovskoye 2)	Kazakhstan	Uranium One/Kazatomprom	ISL	2560	5
Rössing	Namibia	CNNC	open pit	2255	5
SOMAIR	Niger	Orano	open pit	2020	4
Four Mile	Australia	Quasar	ISL	1740	3
Central Mynkuduk	Kazakhstan	Ortalyk	ISL	1650	3
South Inkai 4	Kazakhstan	Uranium One/Kazatomprom	ISL	1600	3
Top 10 total				**28,125**	**57%**

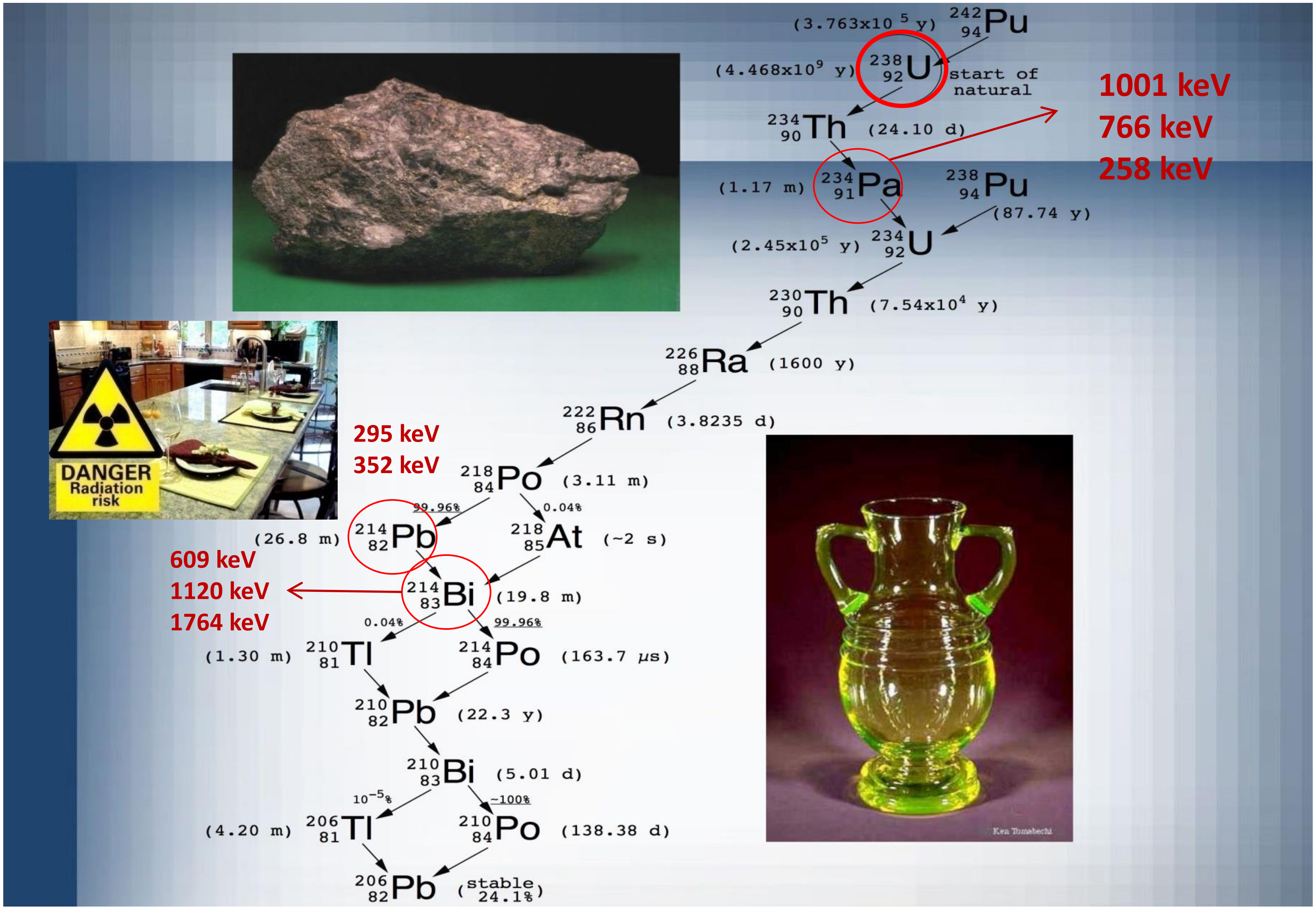
http://www.world-nuclear.org/uploadedFiles/org/WNA/Publications/Nuclear_Information/Pocket%20Guide%20Uranium.pdf/

Top 10 Uranium Mining Nations 2018

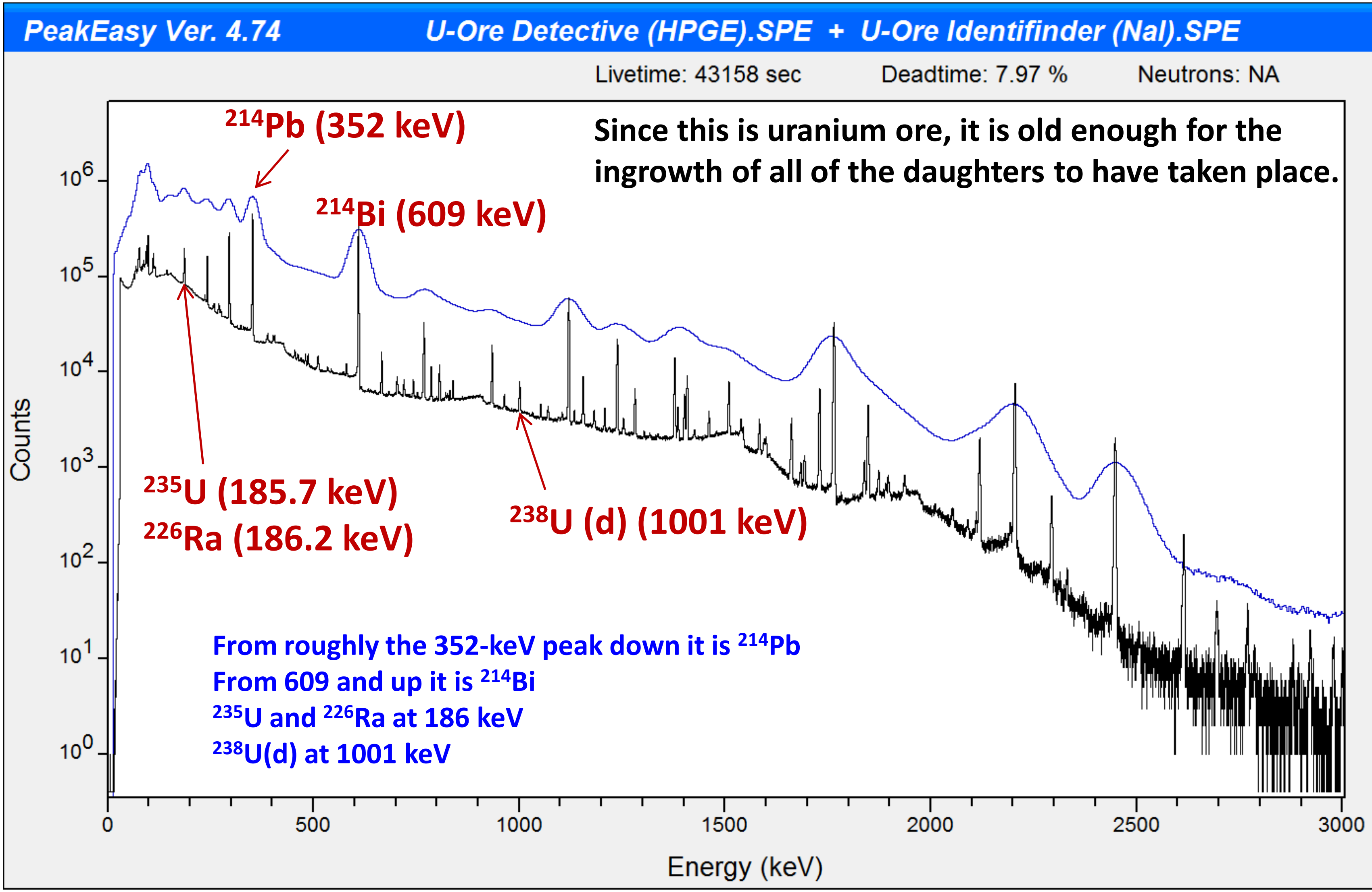


Values in tonnes U

U-238 Decay Series

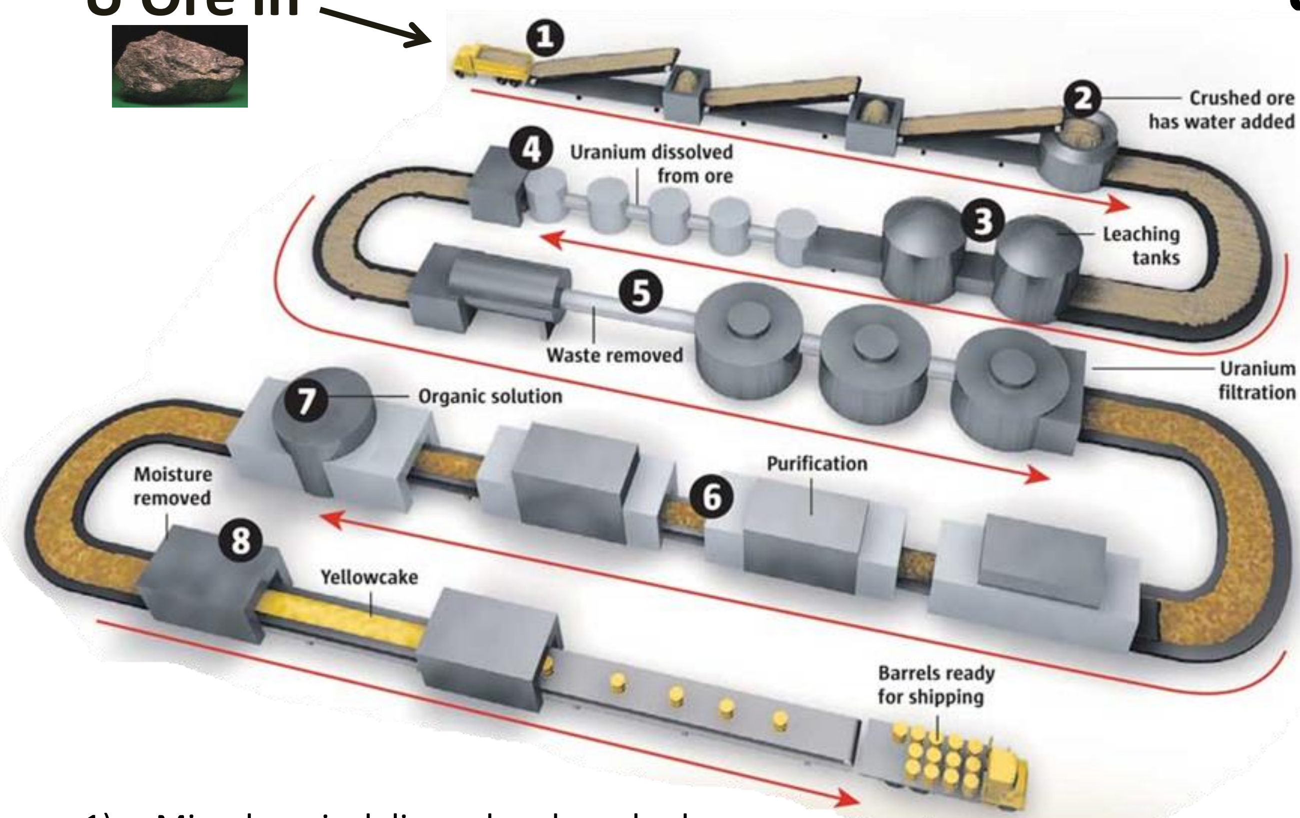


Uranium Ore Spectrum



Uranium Milling

U Ore in

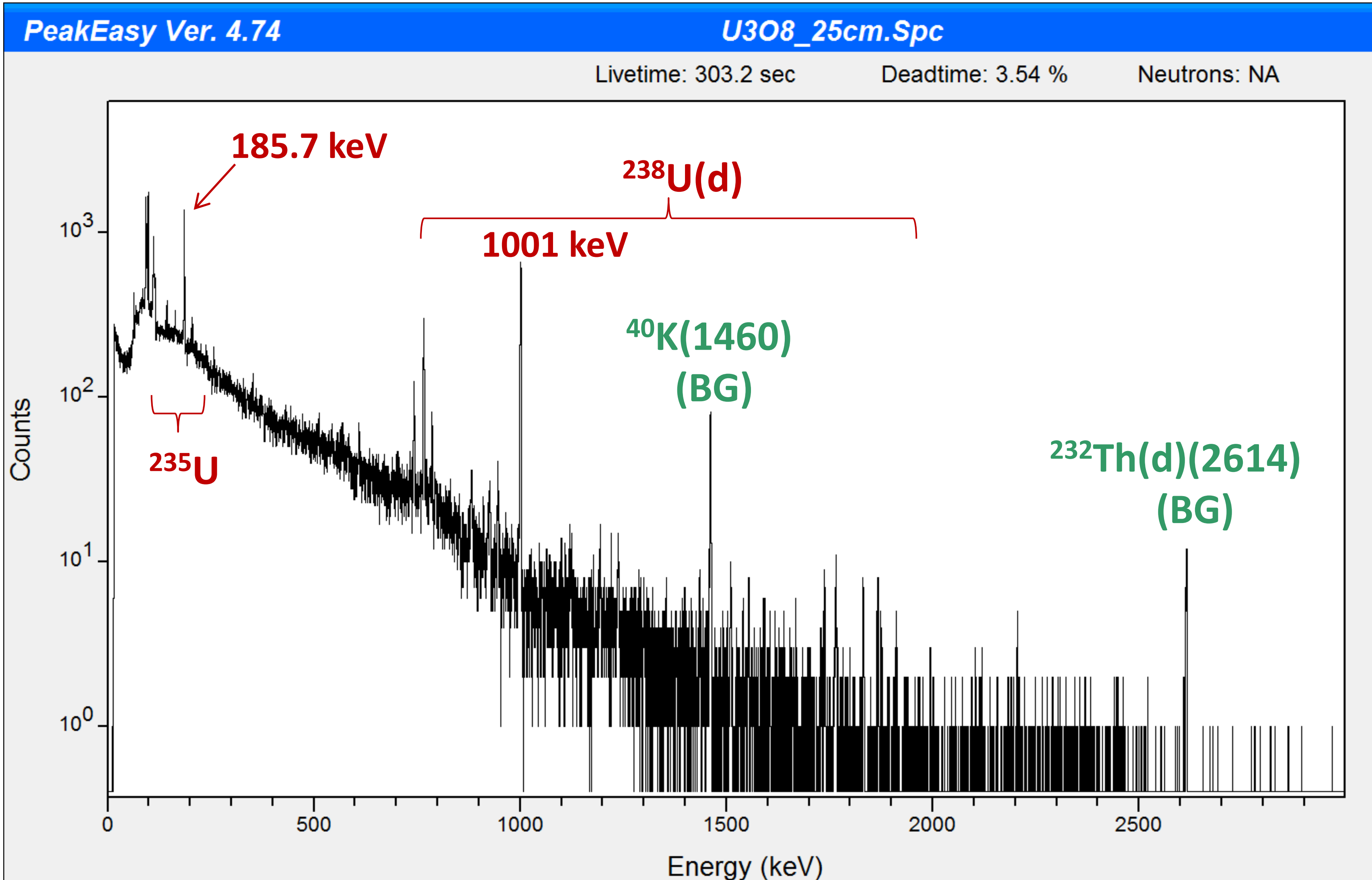


1) Mined ore is delivered and crushed
2) Water added, ore ground into a fine sand (slurry)
3) Slurry pumped into leach tanks
4) H_2SO_4 and H_2O_2 added to dissolve U from ore
5) Waste is separated and stored in tanks
6) U purified and extracted using organic solution
7) U extracted from organic solution using ammonium sulfate
8) Excess moisture removed yielding U_3O_8 "yellowcake"

U_3O_8 out

(or UO_3)

Yellowcake Spectrum

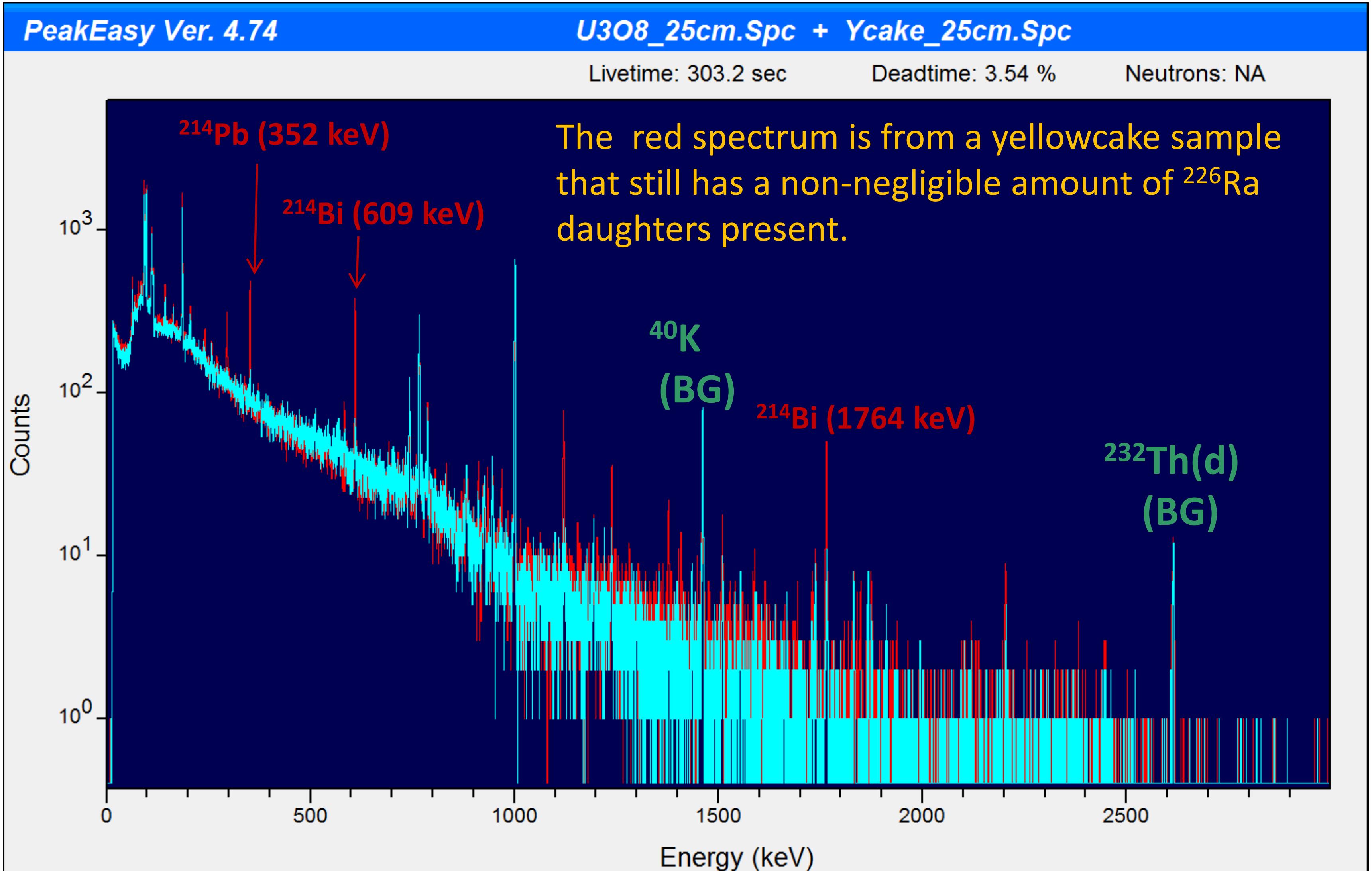


“Yellowcake” is not always yellow

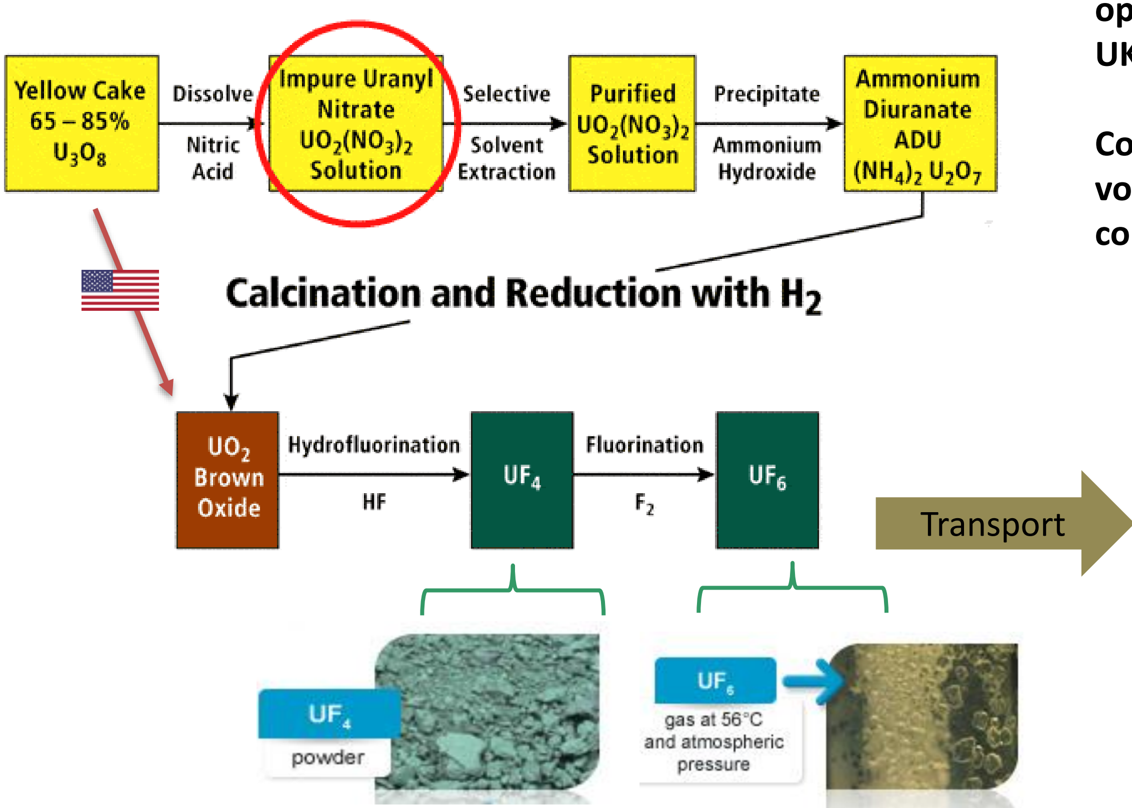
“Yellow cake is the final product of the milling of uranium ore. It contains about 80 % of uranium oxide and is usually represented by the formula U_3O_8. The yellow cake produced by most modern mills is actually brown or black, not yellow; the name comes from the color and texture of the concentrates produced by early mining operations due to impurities from ammonium diuranate $(NH_4)_2U_2O_7$.”

https://www.euronuclear.org/info/encyclopedia/y/yellow-cake.htm

Yellowcake is not always 'pure'



Uranium Conversion



Plants to convert uranium oxide to UF_6 are operating commercially in USA, Canada, France, UK, Russia, China.

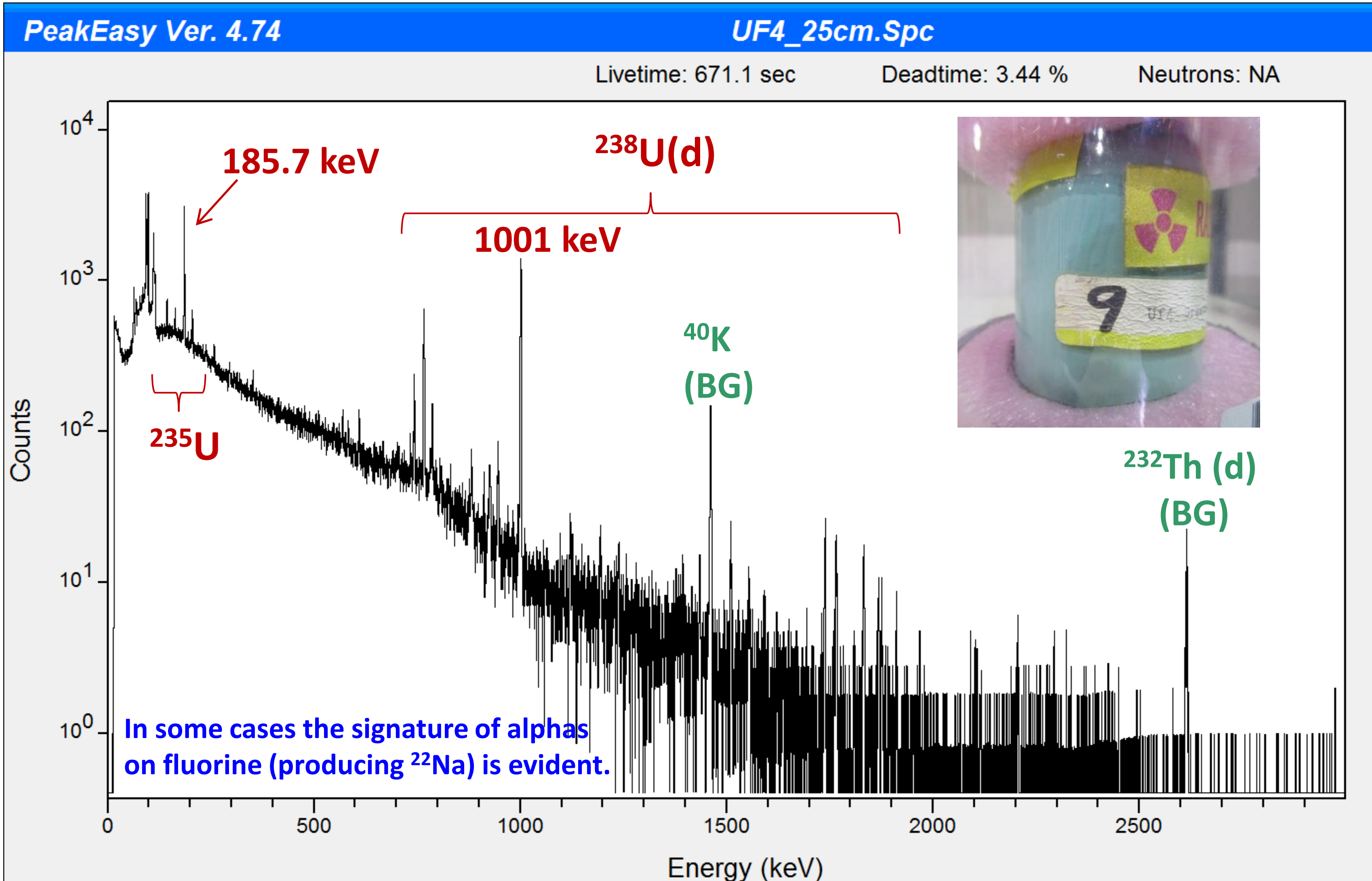
Conversion to UF_6 is achieved by a dry fluoride volatility process in the USA, while all other converters use a wet process.

- Used for natural and depleted uranium
- Holds 12,500 kgs of UF_6 (**8,450 kgs U**)
- A 48Y cylinder filled with natural uranium contains **60.1 kgs of ^{235}U**.
- Nominal wall thickness 16 mm

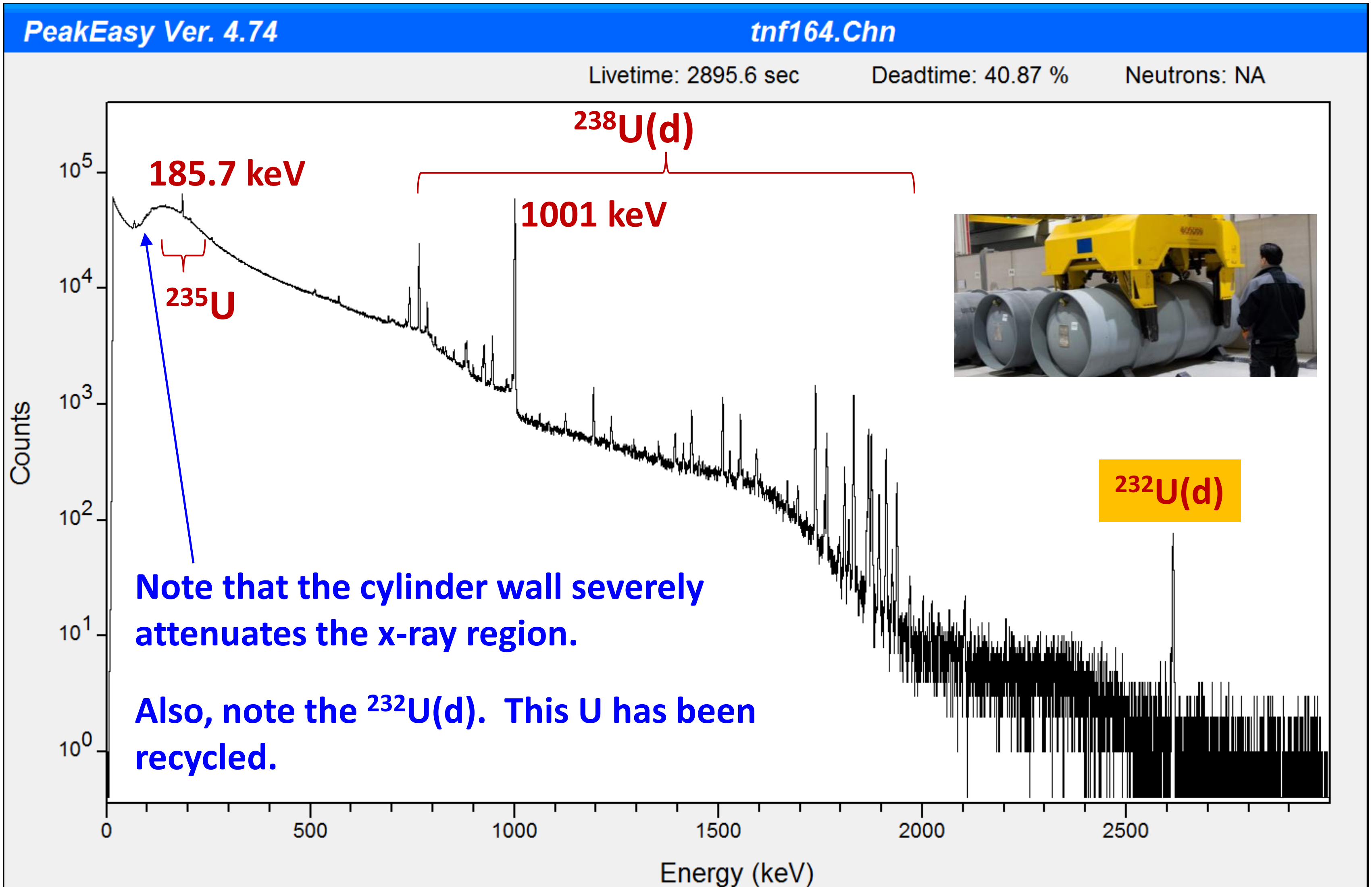
Note: Type 30B Cylinders can hold 2270 kg UF6 (1540 kg U), can transport UF6 up to 4.95% ^{235}U.

http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Conversion-Enrichment-and-Fabrication/Conversion-and-Deconversion/
http://web.ead.anl.gov/uranium/guide/prodhand/sld018.cfm
http://www.areva.com/EN/operations-757/conversion-the-fluorination-of-uranium-in-2-stages.html

UF_4 Spectrum



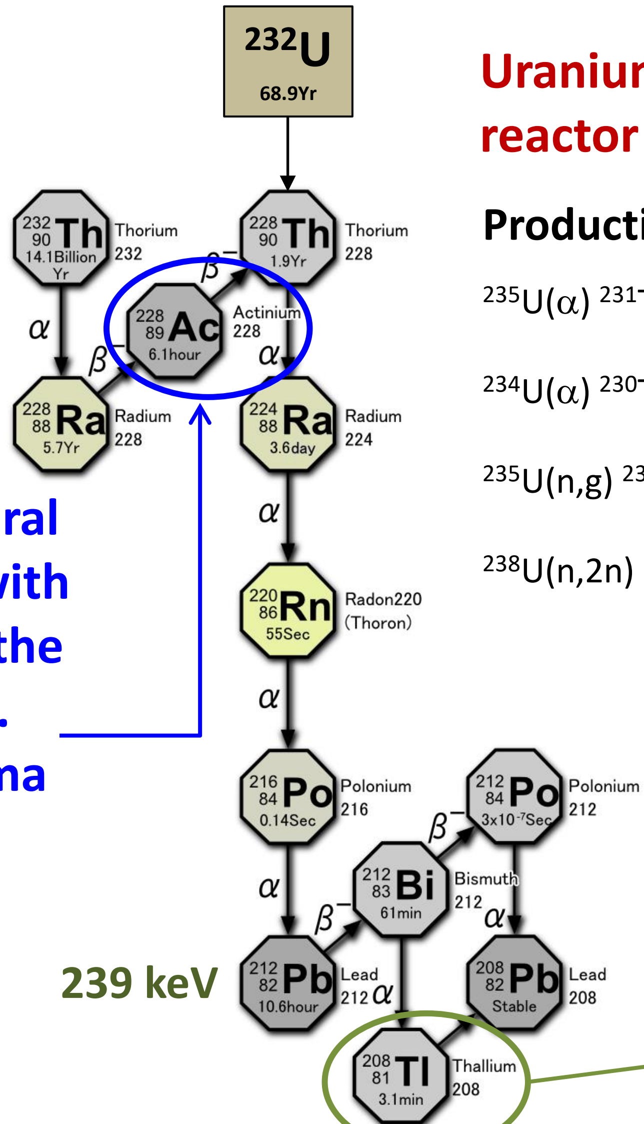
UF_6 Spectrum (~'Natural' U)



Question Time!

- Why does a spectrum of separated uranium look different from uranium ore after 20 y?
  - A) after 20 years it has become stable
  - B) because of the ingrowth of Am-241
  - C) equilibrium of daughters takes ~ 1E6 years
  - D) the enrichment has changed with time

An Aside: ^{232}U in Recycled U



Uranium that has been through a reactor (recycled) contains ^{232}U .

Production of ^{232}U in a Reactor:

$^{235}U(\alpha)\ ^{231}Th(\beta-)\ ^{231}Pa(n,\gamma)\ ^{232}Pa(\beta-)\ ^{232}U$

$^{234}U(\alpha)\ ^{230}Th(n,\gamma)\ ^{231}Th(\beta-)\ ^{231}Pa(n,\gamma)\ ^{232}Pa(\beta-)\ ^{232}U$

$^{235}U(n,g)\ ^{236}U(n,\gamma)\ ^{237}U(\beta-)\ ^{237}Np(n,2n)\ ^{236m}Np(\beta-)\ ^{236}Pu(\alpha)\ ^{232}U$

$^{238}U(n,2n)\ ^{237}U(\beta-)\ ^{237}Np(n,2n)\ ^{236m}Np(\beta-)\ ^{236}Pu(\alpha)\ ^{232}U$

Thorium in the natural background starts with ^{232}Th, and contains the decay product ^{228}Ac. This emits the gamma rays:

911 keV
969 keV
338 keV

239 keV

2614 keV
861 keV
583 keV

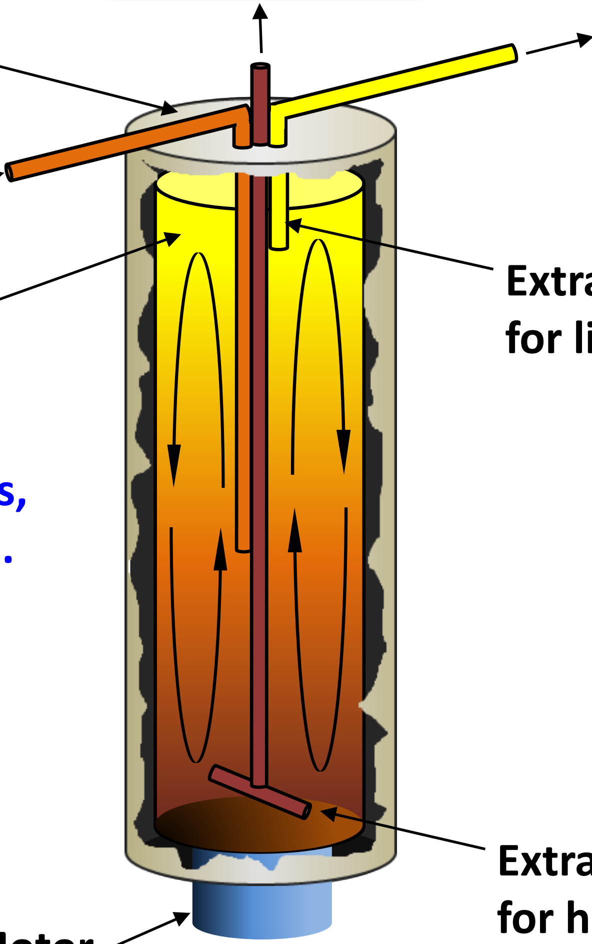
*Peurrung, A.J., "Predicting ^{232}U Content in Uranium", PNNL Document 12075

Enrichment: Gas Centrifuge

A gas centrifuge has a spinning rotor within a case. UF_6 gas is fed to the rotor and the more massive ^{238}U atoms drift to the outside leaving the lighter ^{235}U atoms in the center.

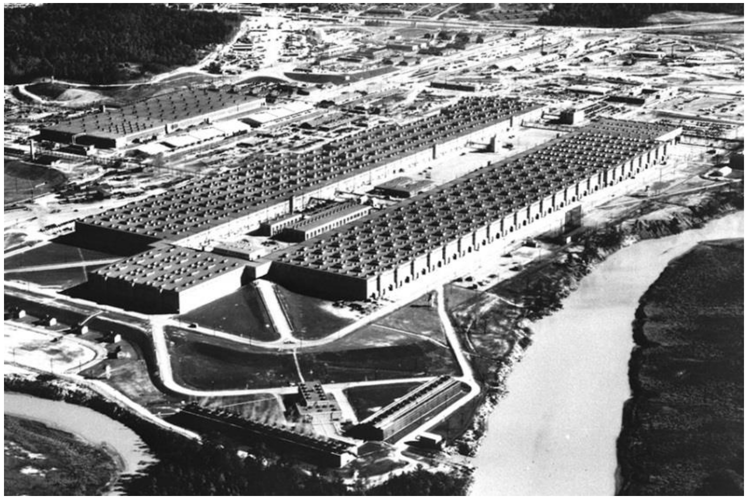
Thermal gradients induce convection currents, which further aid in the separation of ^{238}U and ^{235}U.

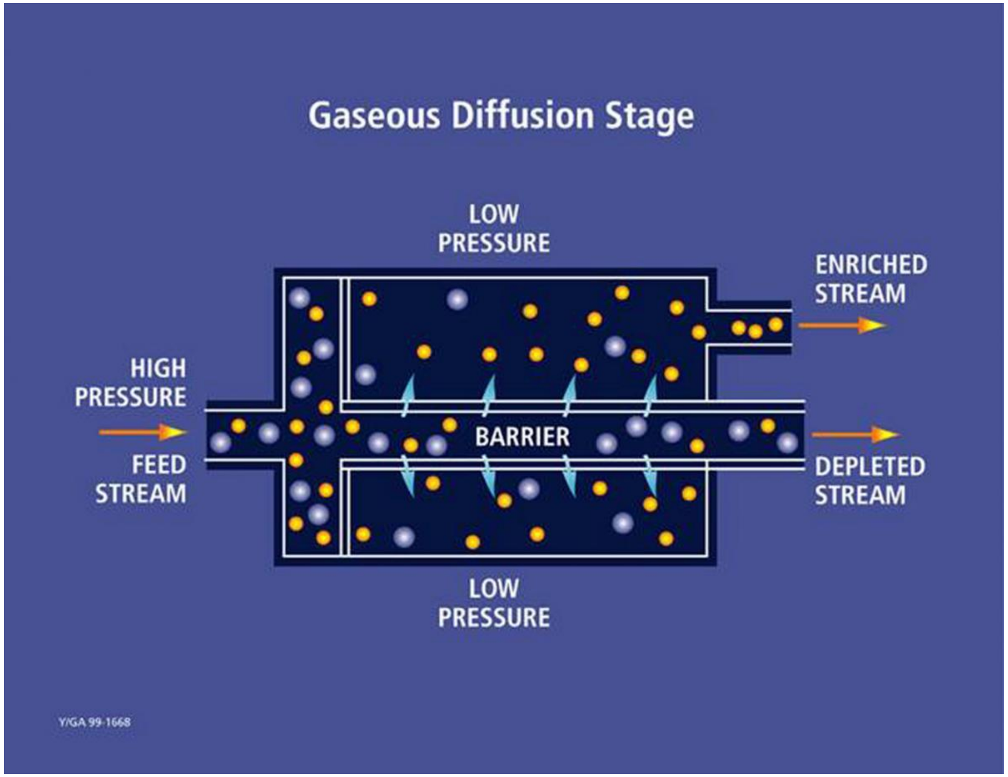
A cascade of centrifuges is used in this process, where the enrichment increases in each stage.



Enrichment: Gaseous Diffusion

In the gaseous-diffusion process UF6 gas is filtered by a semi-porous membrane. The less massive ^{235}U atoms drift through the membrane more easily than ^{238}U atoms.

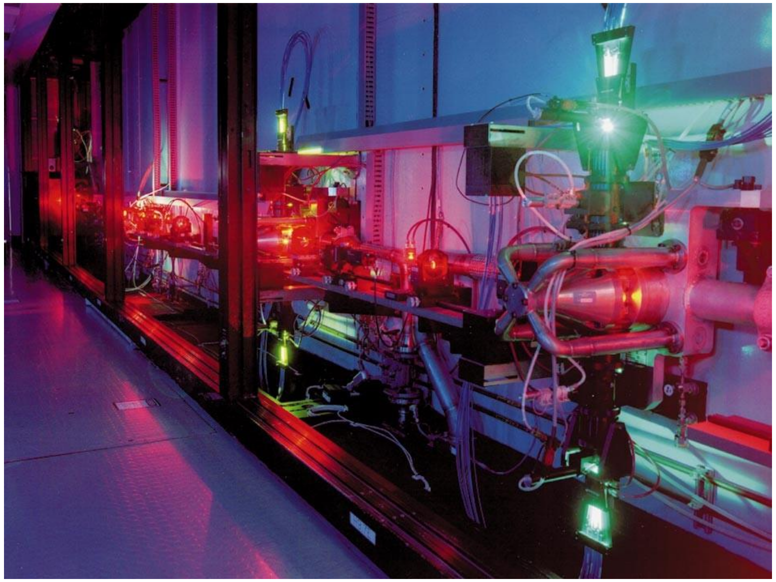


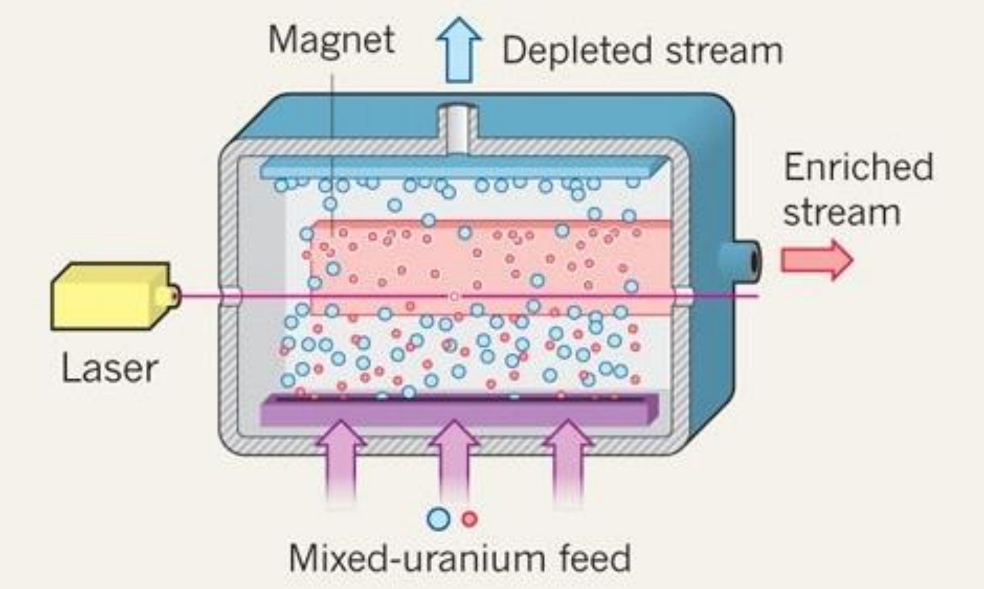


A cascade of stages is used in this process, where the enrichment increases in each stage.

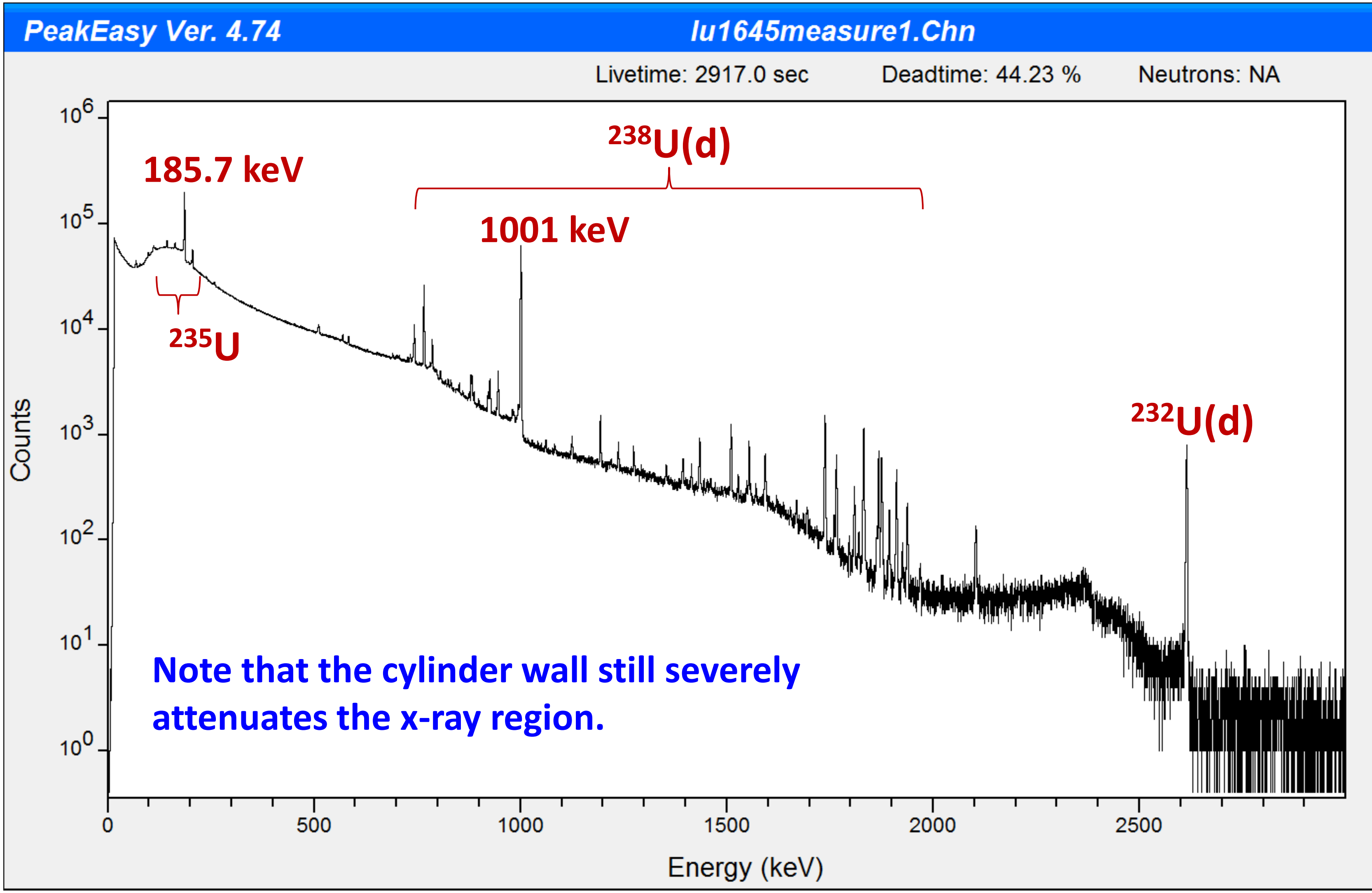
Enrichment: Laser Isotope Separation

In laser-isotope separation, a tunable laser excites and ionizes ^{235}U atoms. These charged atoms are then collected electrostatically or electromagnetically and separated from the neutral ^{238}U atoms.

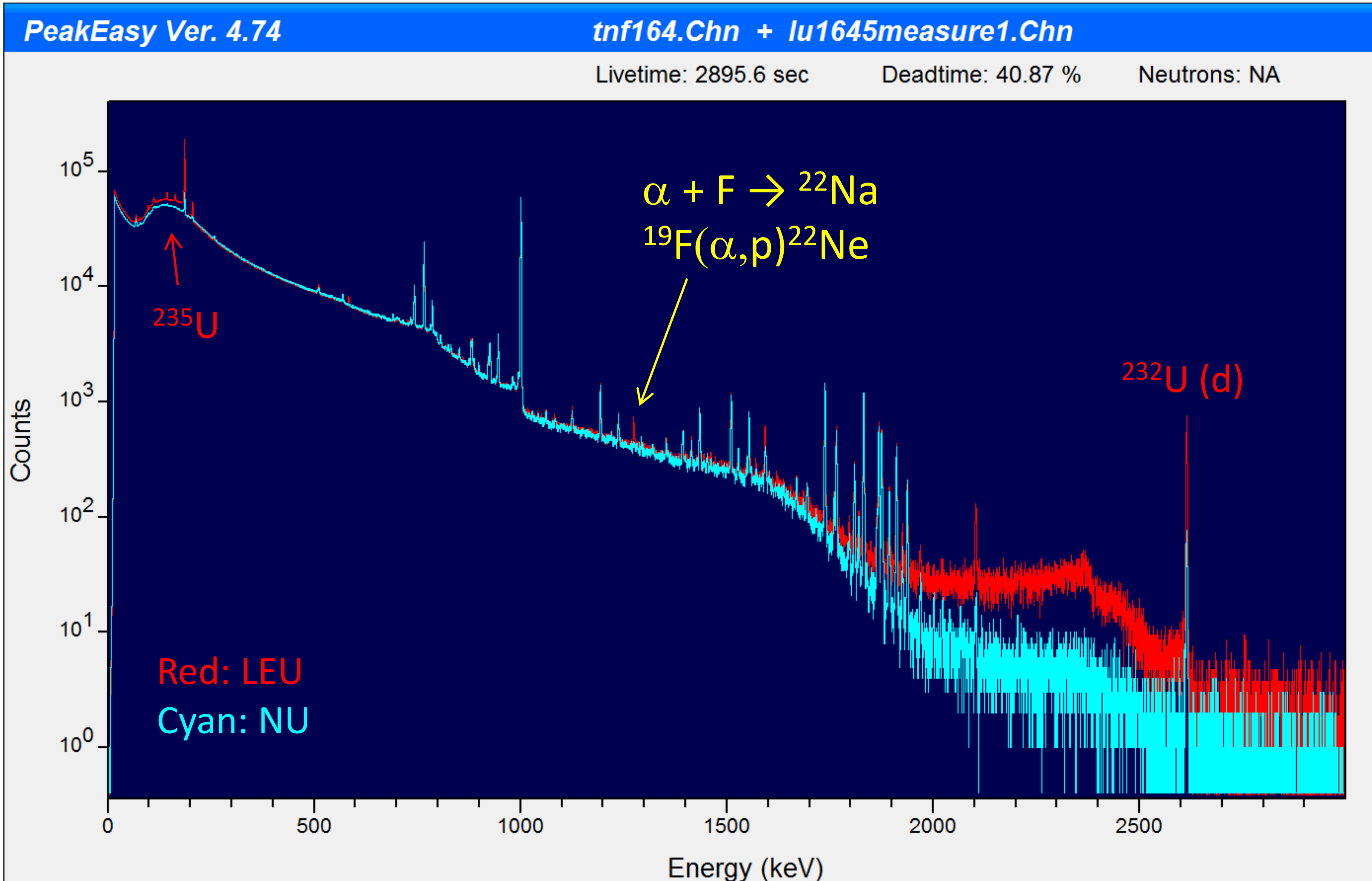




UF_6 Spectrum (LEU)



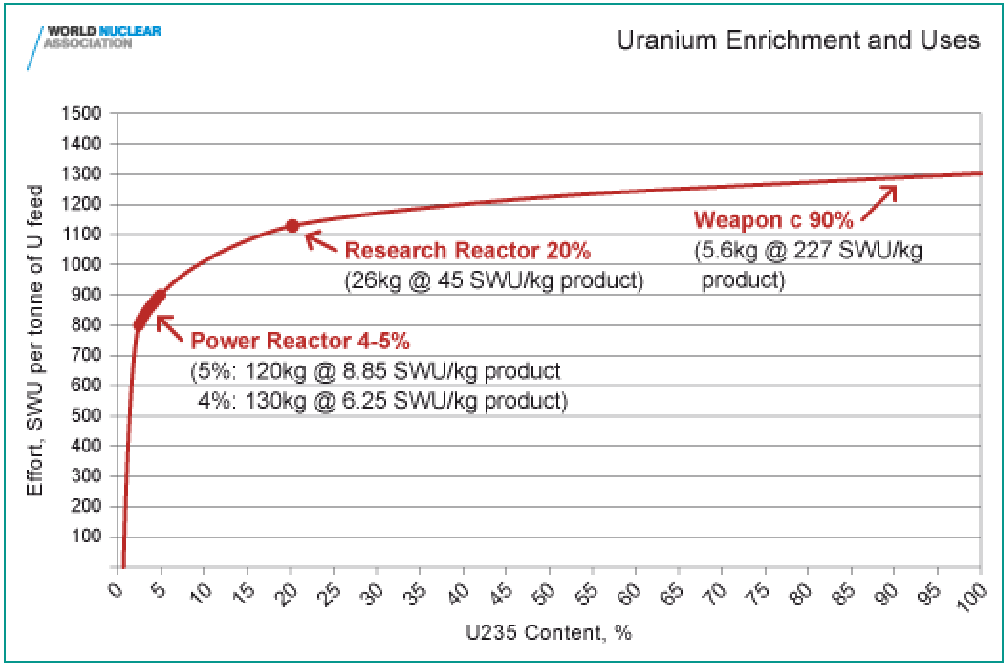
UF_6 Spectra (NU v. LEU)



Relative Enrichment Effort

Separative Work Unit (SWU): This is a complex unit that represents the effort that is required to enrich uranium.

The bulk of the effort is in taking the enrichment from NU to 20 %!



http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Conversion-Enrichment-and-Fabrication/Uranium-Enrichment/

Congressional Research Service
Informing the legislative debate since 1914

Natanz Pilot Enrichment Plant

Iran is using IR-1 centrifuges in this facility to produce LEU containing approximately 20% uranium-235. Iran is also testing several types of centrifuges in the facility. Iran's production of LEU enriched to this level has caused concern because such production requires approximately 90% of the effort necessary to produce weapons-grade HEU, which, as noted, contains approximately 90% uranium-235.[14]

http://fpc.state.gov/documents/organization/234999.pdf

Who Enriches Uranium?

World enrichment capacity – operational and planned (thousand SWU/yr)

Country	Company and plant	2013	2015	2020
France	Areva, Georges Besse I & II	5500	7000	7500
Germany-Netherlands-UK	Urenco: Gronau, Germany; Almelo, Netherlands; Capenhurst, UK.	14,200	14,400	14,900
Japan	JNFL, Rokkaasho	75	75	75
USA	USEC, Piketon	0*	0	0
USA	Urenco, New Mexico	3500	4700	4700
USA	Areva, Idaho Falls	0	0	0
USA	Global Laser Enrichment, Paducah	0	0	0
Russia	Tenex: Angarsk, Novouralsk, Zelenogorsk, Seversk	26,000	26,578	28,663
China	CNNC, Hanzhun & Lanzhou	2200	5760	10,700+
Other	Various: Argentina, Brazil, India, Pakistan, Iran	75	100	170
	Total SWU/yr approx	**51,550**	**58,600**	**66,700**
	Requirements (*WNA reference scenario*)	49,154	47,285	57,456

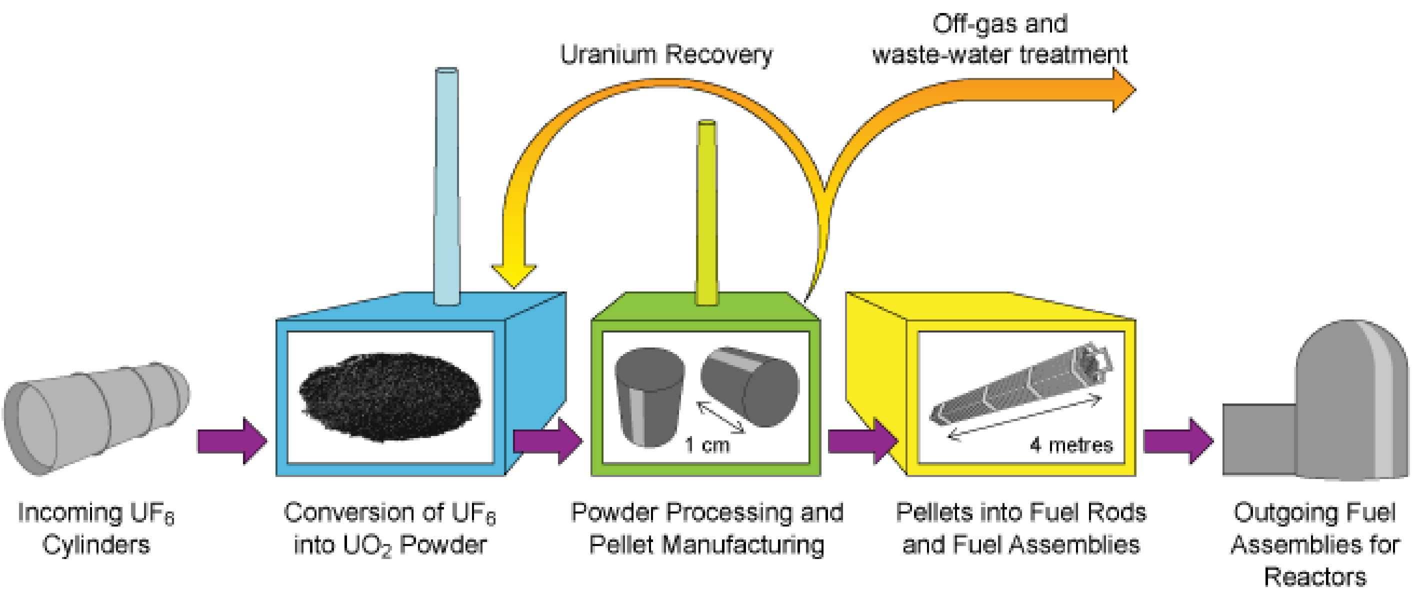
https://www.world-nuclear.org/information-library/nuclear-fuel-cycle/conversion-enrichment-and-fabrication/uranium-enrichment.aspx/

Question Time!

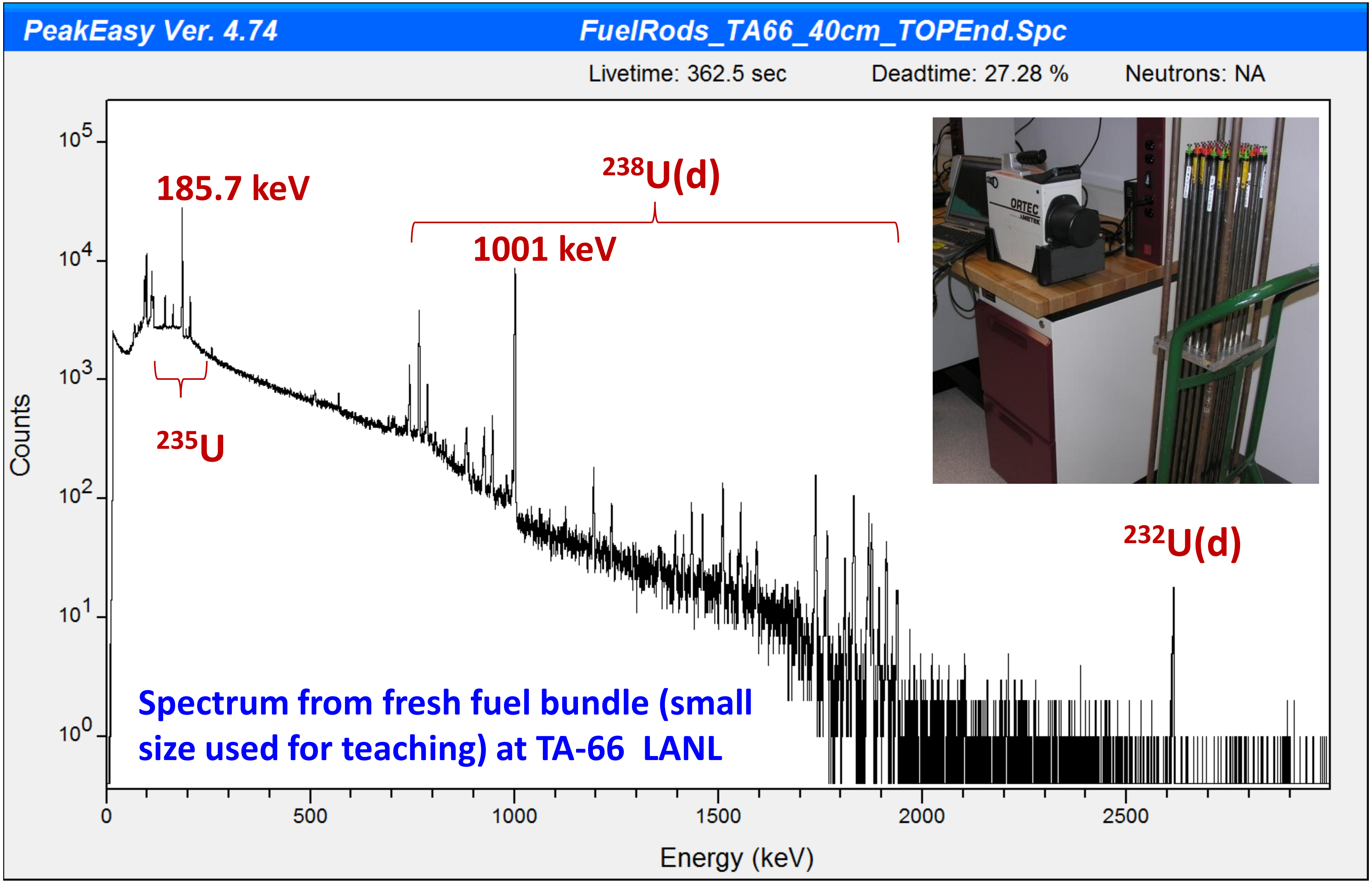
- The presence of U-232 in uranium items:
  - A) enhances the intensity of the 2614.5 –keV peak
  - B) mimics the natural background from uranium
  - C) indicates that the material is 100% natural
  - D) significantly alters the enrichment

Uranium Fuel Fabrication

- Produce pure uranium dioxide (UO_2) from incoming UF_6 or UO_3.
- Produce high-density, accurately shaped ceramic UO_2 pellets.
- Fabricate the rigid metal framework for the fuel assembly

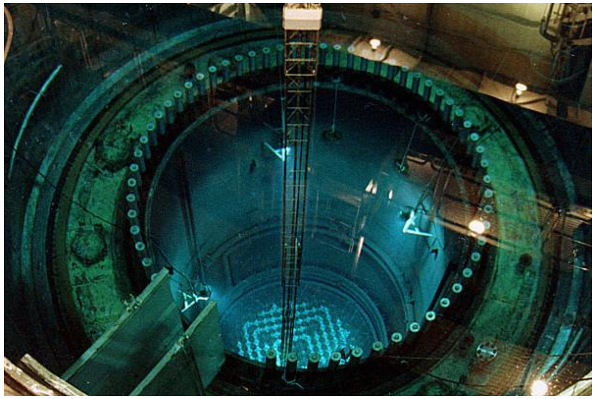


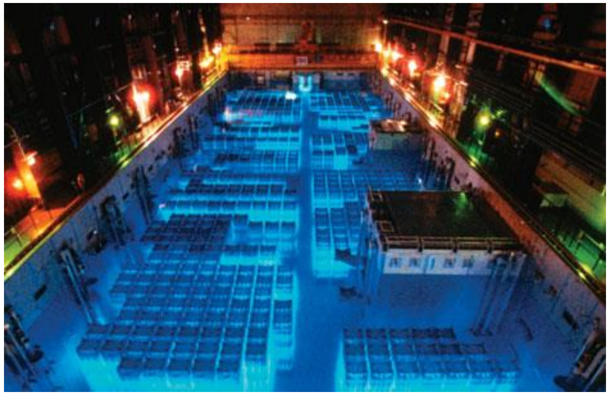
UO_2 (Fuel Bundle) Spectrum



Spent Fuel

Spent fuel is removed from the reactor directly to a spent-fuel pond, where it may stay for several years as it is highly radioactive and thermally hot due to fission fragments and decay products.



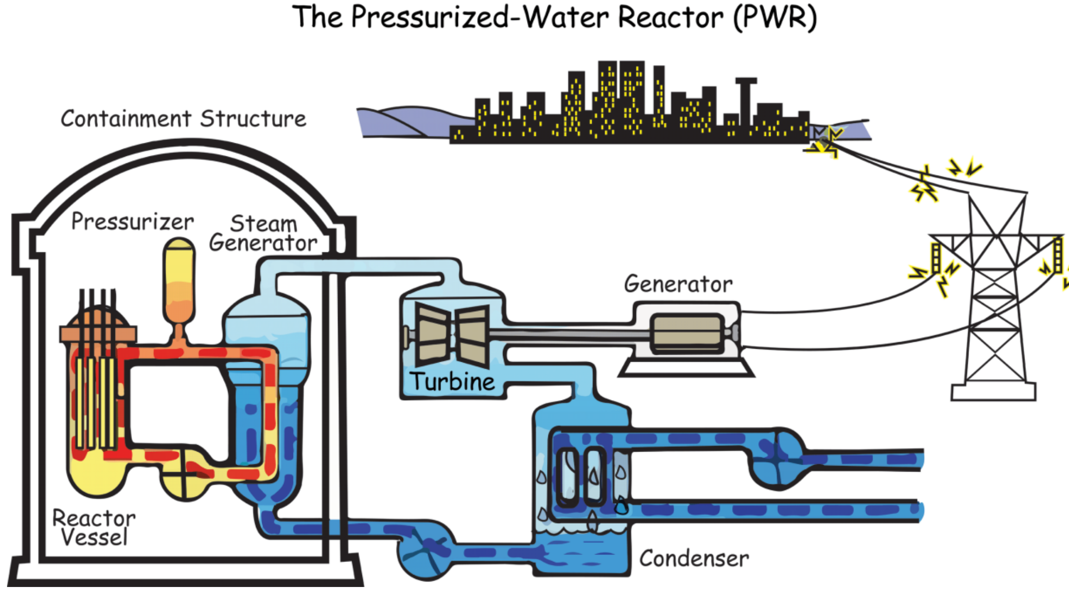


Following this it is moved to dry cask storage.

Used fuel will typically have 95% ^{238}U, about <1% ^{235}U and ~1% plutonium.

The high output of radiation from the fission products make gamma-ray measurements of spent fuel very difficult.

In the Reactor



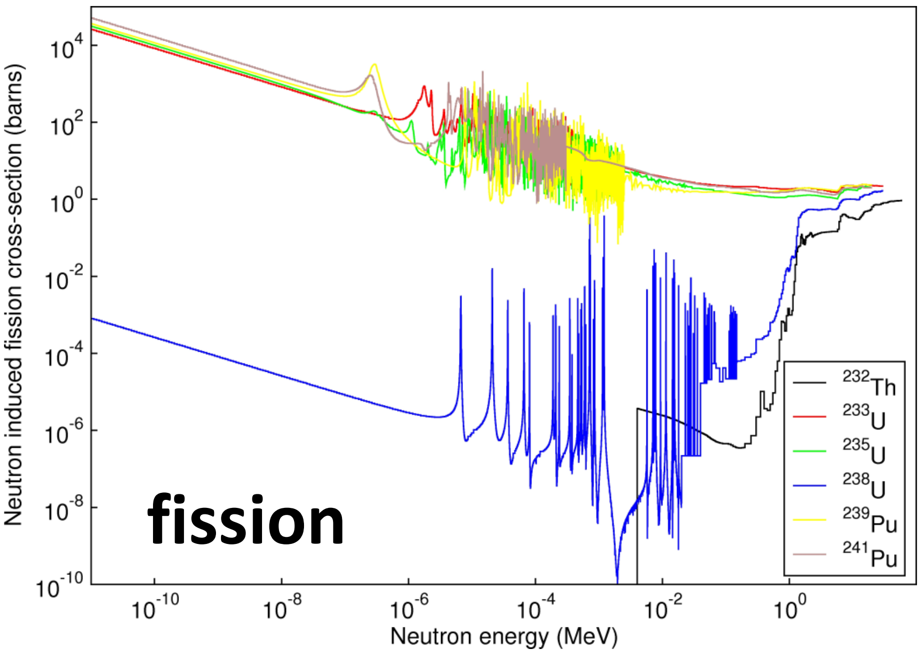
- For a reactor with an output of 1000 megawatts (MWe), the core would contain about 75 tonnes of low-enriched uranium.

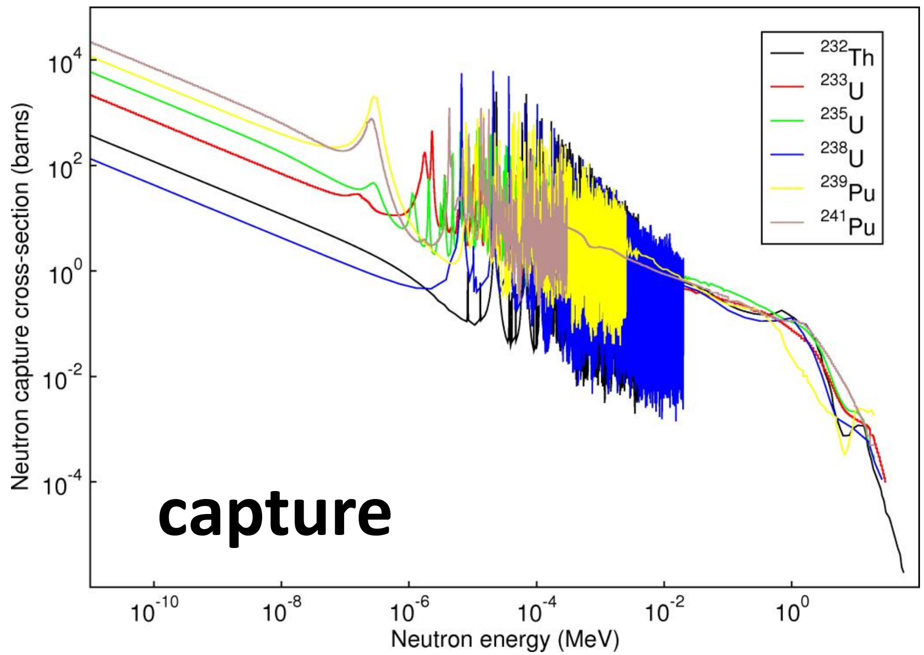
- Some of the ^{238}U in the reactor core is turned into plutonium (which in turn provides ~1/3 of the total energy).

$$^{238}_{92}U + n \rightarrow {}^{239}_{92}U \xrightarrow[23.5m]{\beta-} {}^{239}_{93}Np \xrightarrow[2.35d]{\beta-} {}^{239}_{94}Pu$$

Fission vs. Capture

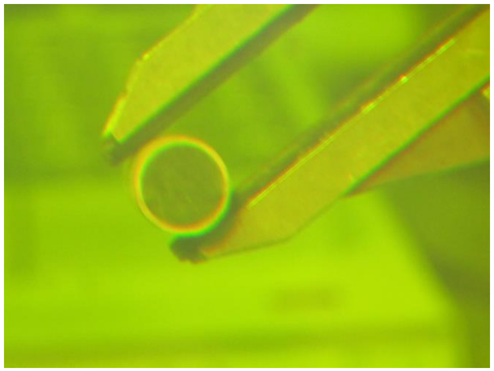
^{235}U and ^{239}Pu behave similarly with respect to neutron capture and fission.





^{238}U is far more likely to capture a neutron below 1 MeV (and make ^{239}Pu !)

Spent Fuel Fission Product Data



58 g sample of spent BWR fuel (cool down time ~21 years)

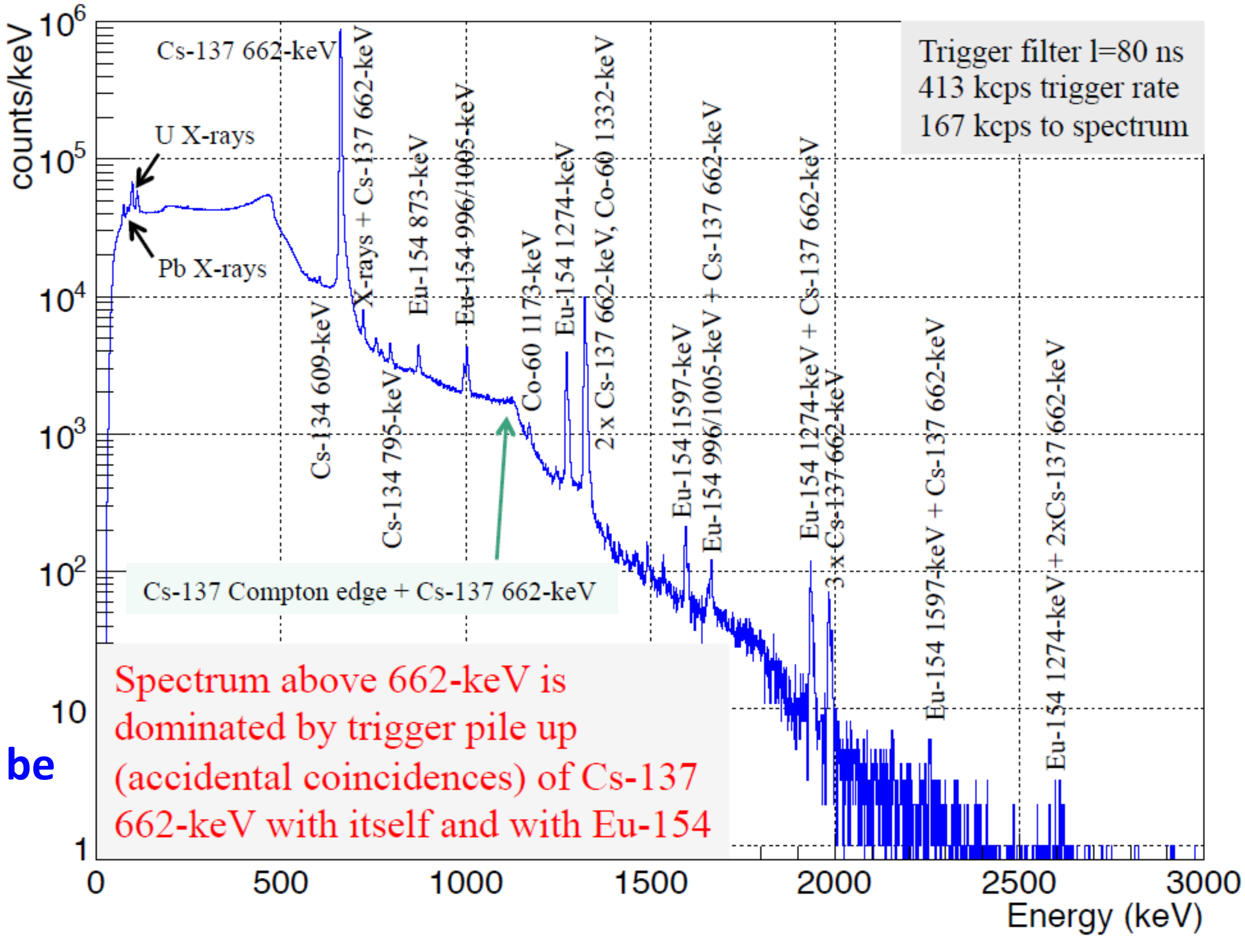


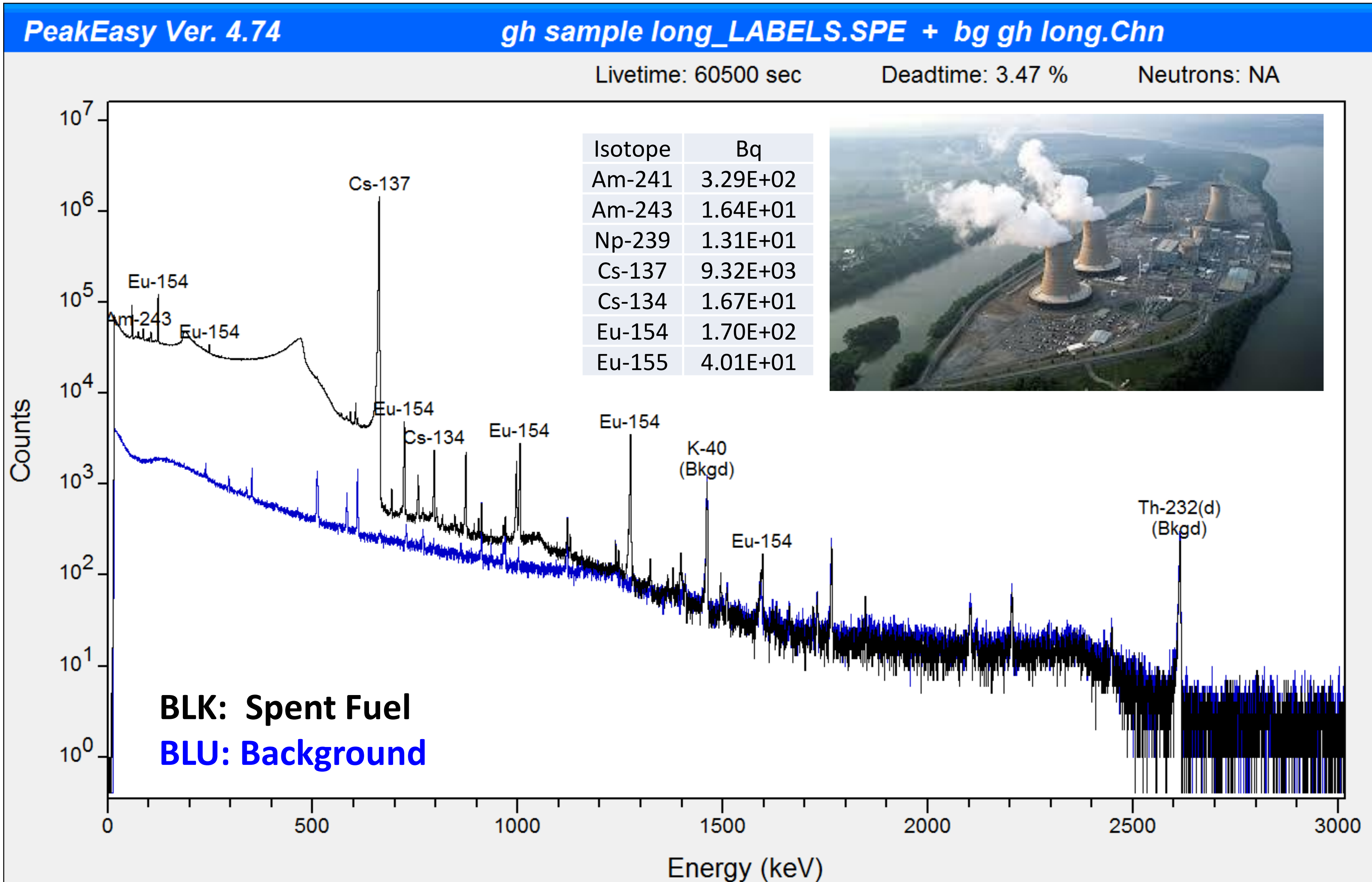
Figure 5: Energy spectrum obtained at 803 cm with no shielding.

Activity ratios of nuclides can be used to determine exposure (burnup) of fuel.

- **$^{134}Cs/^{137}Cs$**
- **$^{154}Eu/^{137}Cs$**

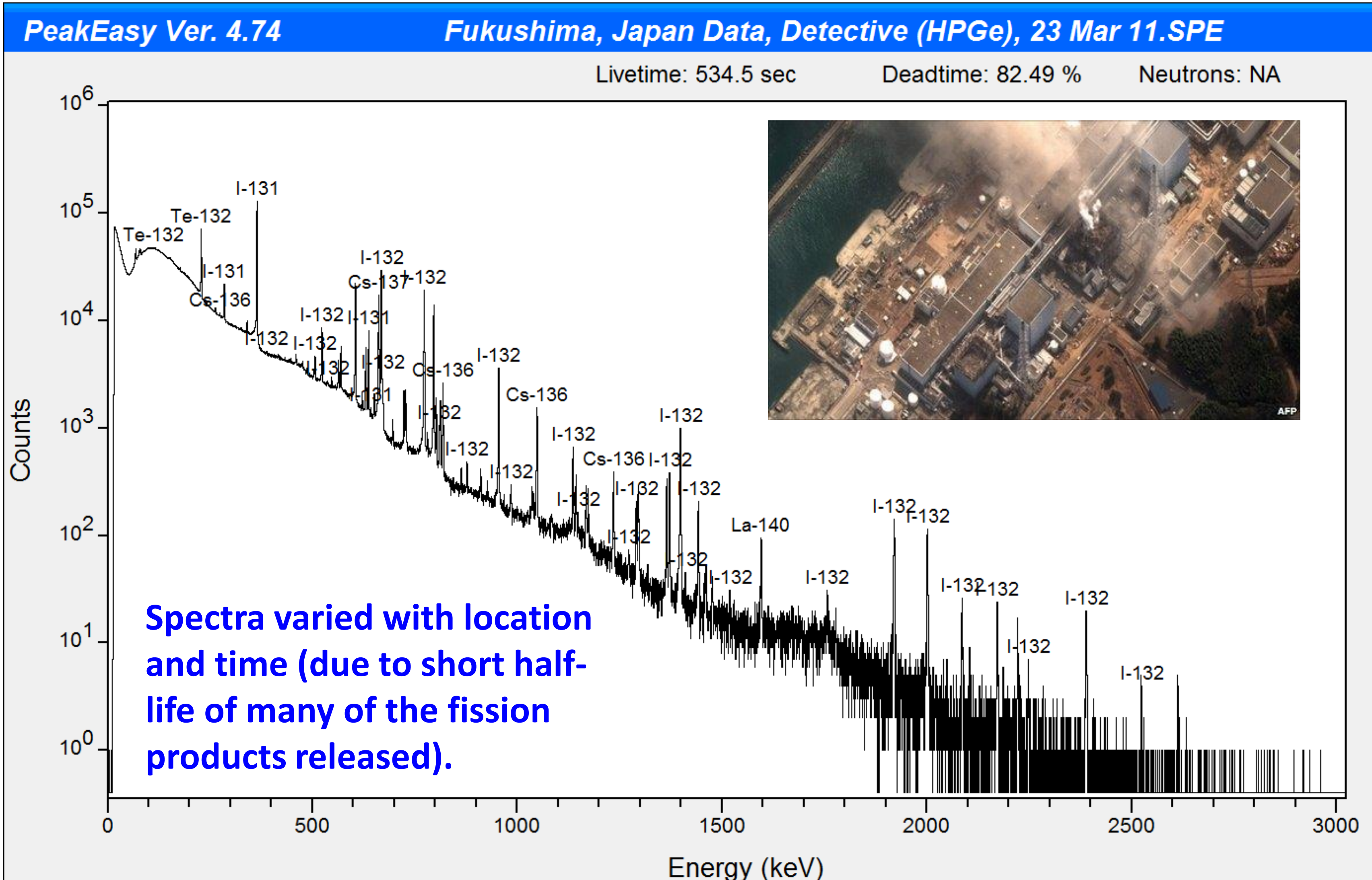
Fast, J.E. et al., "Spent Fuel Measurements", PNNL Document 23561

Spent Fuel from TMI



Isotope	Bq
Am-241	3.29E+02
Am-243	1.64E+01
Np-239	1.31E+01
Cs-137	9.32E+03
Cs-134	1.67E+01
Eu-154	1.70E+02
Eu-155	4.01E+01

Fukushima



PeakEasy Ver. 4.74
Fukushima, Japan Data, Detective (HPGe), 23 Mar 11.SPE
Livetime: 534.5 sec
Deadtime: 82.49 %
Neutrons: NA
Counts
Energy (keV)
Te-132
I-131
Cs-136
Cs-137
I-132
La-140
AFP
Spectra varied with location and time (due to short half-life of many of the fission products released).

Fukushima and Chernobyl

	Half-Life	Released into the environment (Ci) Chernobyl	Fukushima	Fukushima/ Chernobyl
Xe-133	5 d	1.76E+08	2.97E+08	1.69
I-131	8 d	4.76E+07	4.32E+06	0.09
Cs-134	2 y	1.27E+06	4.86E+05	0.38
Cs-137	30 y	2.30E+06	4.05E+05	0.18
Sr-90	29 y	2.70E+05	3.78E+03	0.01
Pu-238	88 y	4.05E+02	5.14E-01	0.00127
Pu-239	24,100 y	3.51E+02	8.65E-02	0.00025
Pu-240	6,540 y	4.86E+02	8.65E-02	0.00018
Total	-	2.27E+08	3.03E+08	**1.3**

Ratio of radionuclides accumulated in the reactor core at the time of the accidents that were released into the environment

Nuclides	Chernobyl NPS [f]	TEPCO's Fukushima Daiichi NPS [g]
Xenon (Xe)-133	Nearly 100%	Approx. 60%
Iodine (I)-131	Approx. 50%	Approx. 2-8%
Cesium (Cs)-137	Approx. 30%	Approx. 1-3%

Note: The reported release from Fukushima varies significantly between sources!

https://www.env.go.jp/en/chemi/rhm/basic-info/1st/02-02-05.html

Fukushima over time

	Release	6 Months	1 Year	10 Years	100 Years
Xe-133	2.97E+08	3.1E-03	3.1E-14	5.3E-212	0.0E+00
I-131	4.32E+06	2.2E+00	1.1E-06	3.8E-120	0.0E+00
Cs-134	4.86E+05	4.1E+05	3.4E+05	1.5E+04	4.3E-10
Cs-137	4.05E+05	4.0E+05	4.0E+05	3.2E+05	4.0E+04
Sr-90	3.78E+03	3.7E+03	3.7E+03	3.0E+03	3.5E+02
Pu-238	5.14E-01	5.1E-01	5.1E-01	4.7E-01	2.3E-01
Pu-239	8.65E-02	8.6E-02	8.6E-02	8.6E-02	8.6E-02
Pu-240	8.65E-02	8.6E-02	8.6E-02	8.6E-02	8.6E-02

https://www.env.go.jp/en/chemi/rhm/basic-info/1st/02-02-05.html

Chernobyl over time

	Release	6 Months	1 Year	10 Years	100 Years
Xe-133	1.76E+08	1.8E-03	1.9E-14	3.1E-212	0.0E+00
I-131	4.76E+07	2.4E+01	1.2E-05	4.2E-119	0.0E+00
Cs-134	1.27E+06	1.1E+06	9.0E+05	4.0E+04	1.1E-09
Cs-137	2.30E+06	2.3E+06	2.2E+06	1.8E+06	2.3E+05
Sr-90	2.70E+05	2.7E+05	2.6E+05	2.1E+05	2.5E+04
Pu-238	4.05E+02	4.0E+02	4.0E+02	3.7E+02	1.8E+02
Pu-239	3.51E+02	3.5E+02	3.5E+02	3.5E+02	3.5E+02
Pu-240	4.86E+02	4.9E+02	4.9E+02	4.9E+02	4.8E+02

https://www.env.go.jp/en/chemi/rhm/basic-info/1st/02-02-05.html

Reprocessing

Used fuel still contains about 96% of its original uranium (<1% U-235)
About 3% of the used fuel comprises waste products
and 1% is plutonium.

Over the last 50 years the principal reason for reprocessing used fuel has been to recover unused plutonium, along with less immediately useful unused uranium.

(tonnes per year)

LWR fuel	France, La Hague	1700
	Russia, Ozersk (Mayak)	400
	Japan (Rokkasho)	800*
	Total LWR (approx)	2100
Other nuclear fuels	UK, Sellafield (Magnox)	1500
	India (PHWR, 4 plants)	260
	Total other (approx)	1760
Total civil capacity		3860

** now expected to start operation in 2022*

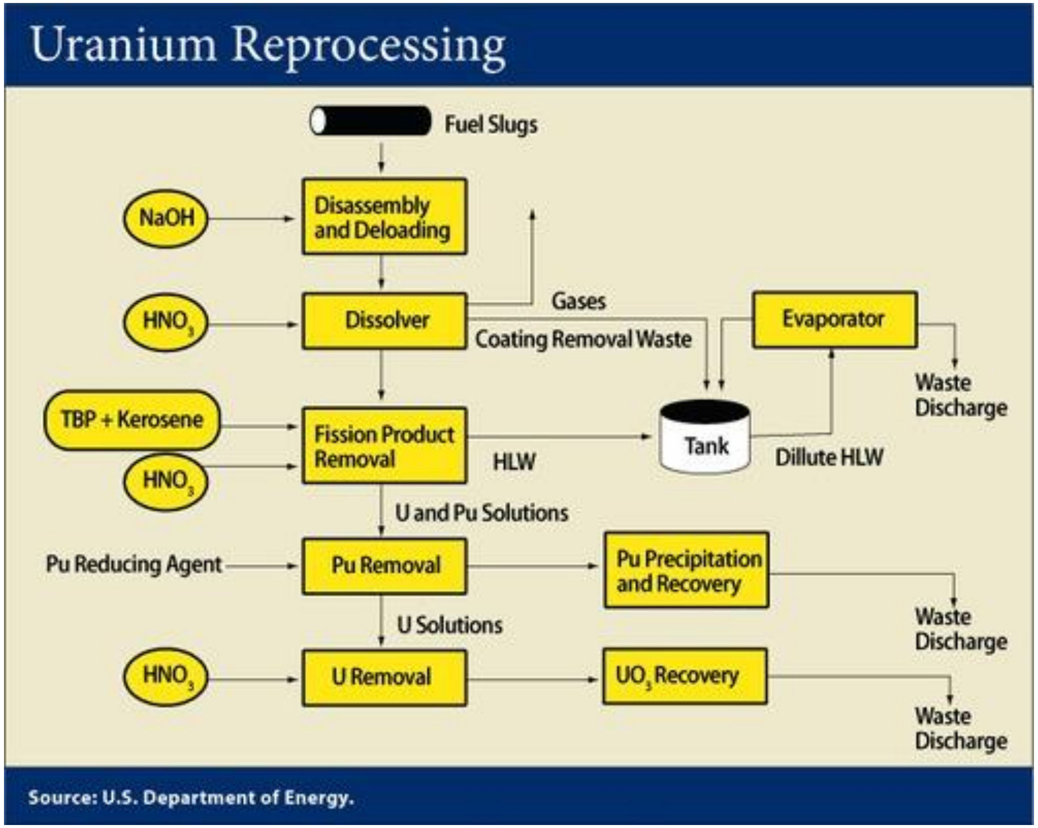
Sellafield, UK

https://www.world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/processing-of-used-nuclear-fuel.aspx

PUREX

All commercial reprocessing plants use the well-proven hydrometallurgical PUREX (plutonium uranium redox extraction) process

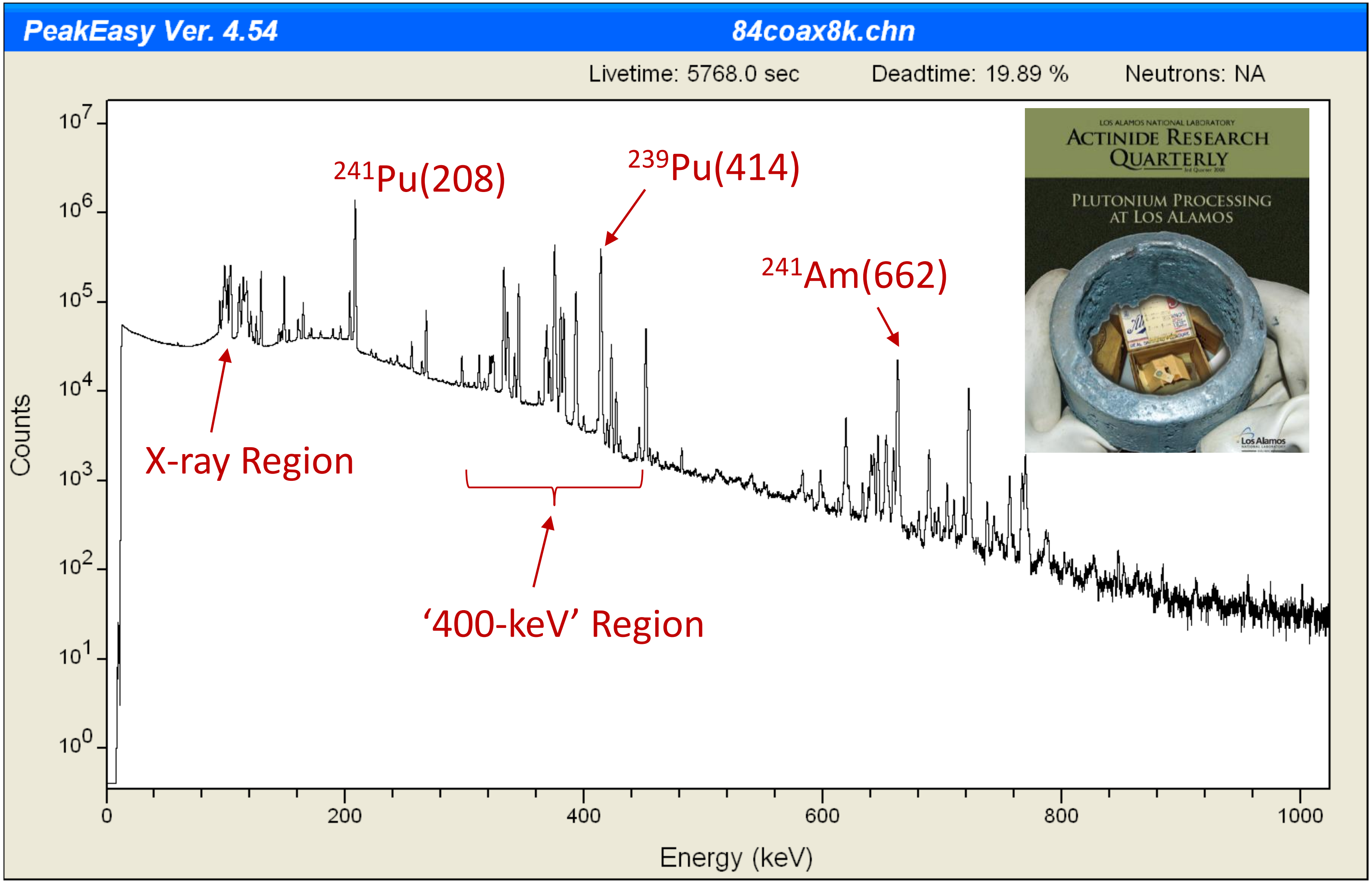
- fuel elements dissolved in concentrated nitric acid
- U and Pu (and perhaps others) are separated



The Pu and U can be returned to the input side of the fuel cycle: the uranium to the conversion plant prior to re-enrichment* and the plutonium straight to MOX fuel fabrication.

* If the ^{236}U content is high the uranium is better suited for use in MOX fuel.

Plutonium Spectrum



Mixed-Oxide (MOX) Fuel

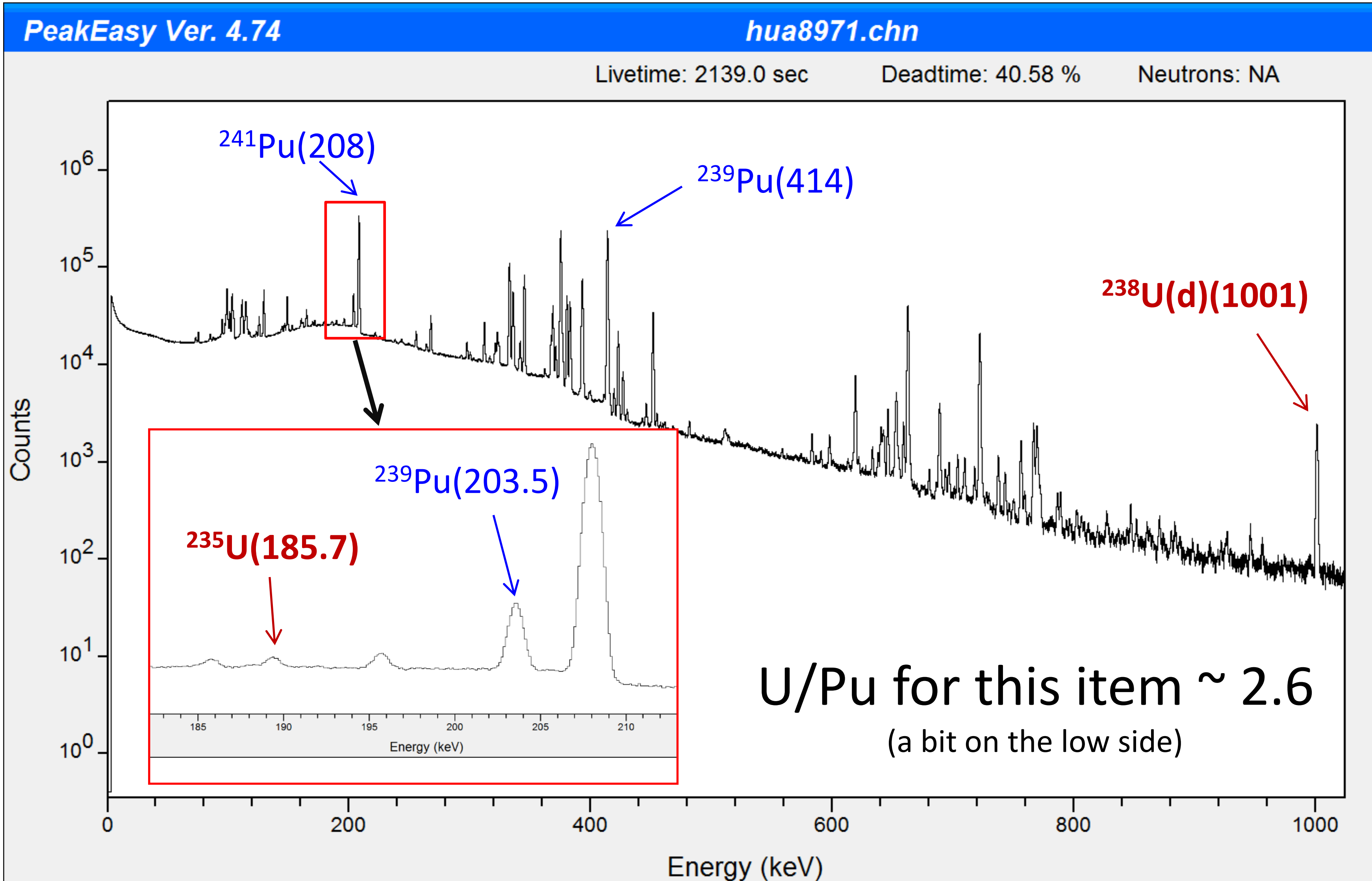
- Most of the separated plutonium is used almost immediately in mixed oxide (MOX) fuel.
  - MOX ≈ $PuO_2 + UO_2$ (NU *or* DU *or* Recycled U)
  - PuO_2 content typically ~ 1.5 – 30 wt. %
  - MOX essentially can be used in lieu of LEU

World mixed oxide fuel fabrication capacities (t/yr)

	2017	2020
France, Melox	195	195
Japan, Tokai	10	10
Japan, J-MOX Rokkasho	0	140
Russia, MCC Zheleznogorsk	60	60
Total	**265**	**405**

https://www.world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/mixed-oxide-fuel-mox.aspx

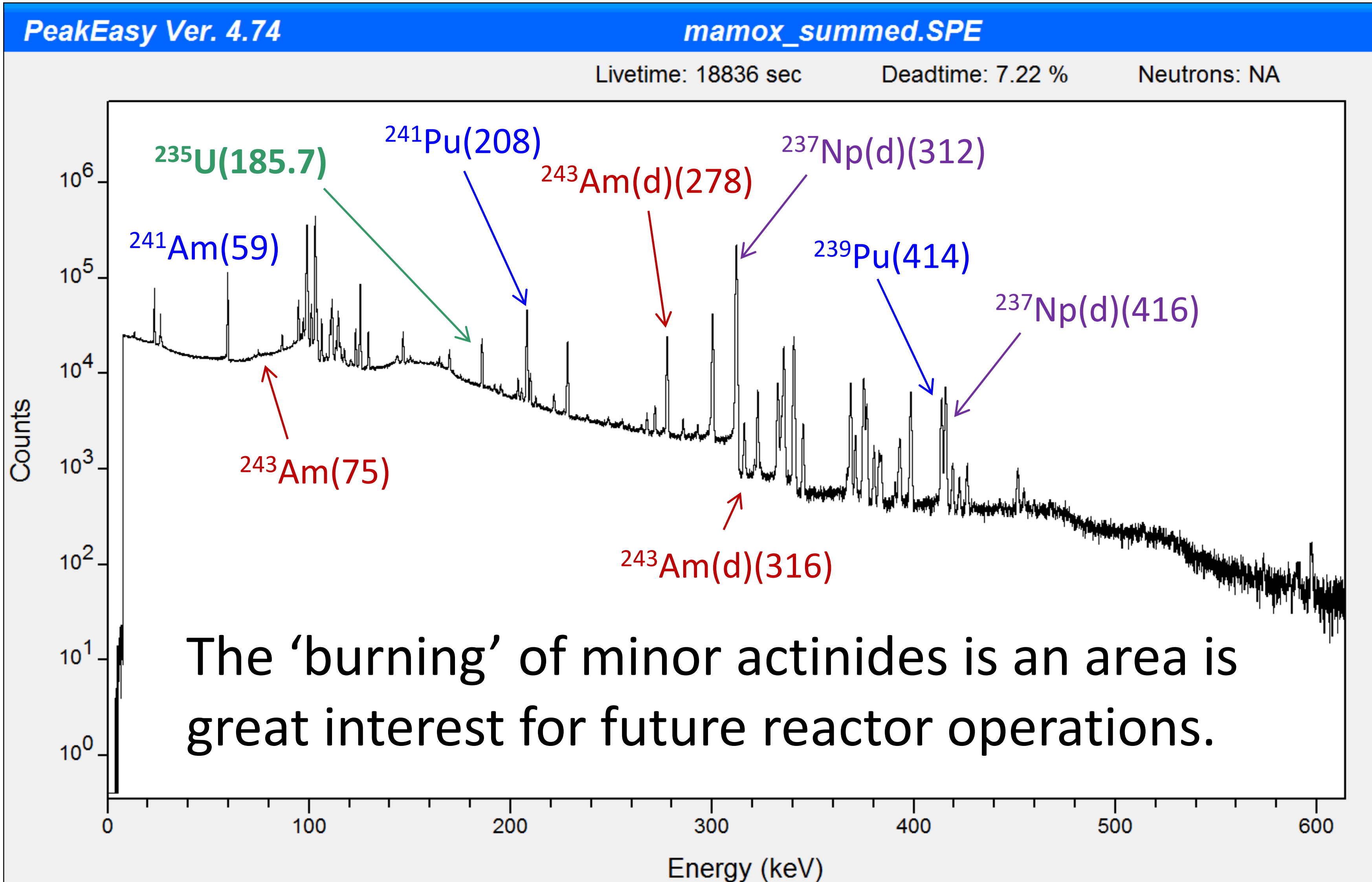
Example MOX Spectrum



U/Pu for this item ~ 2.6

(a bit on the low side)

Minor-Actinide MOX Spectrum



The 'burning' of minor actinides is an area is great interest for future reactor operations.

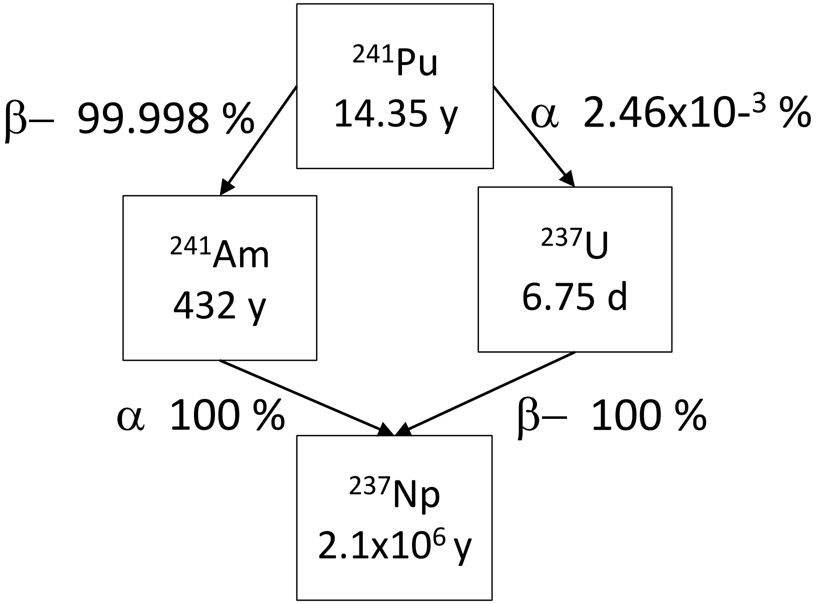
Neptunium-237

^{237}Np is a minor actinide separated from spent fuel.

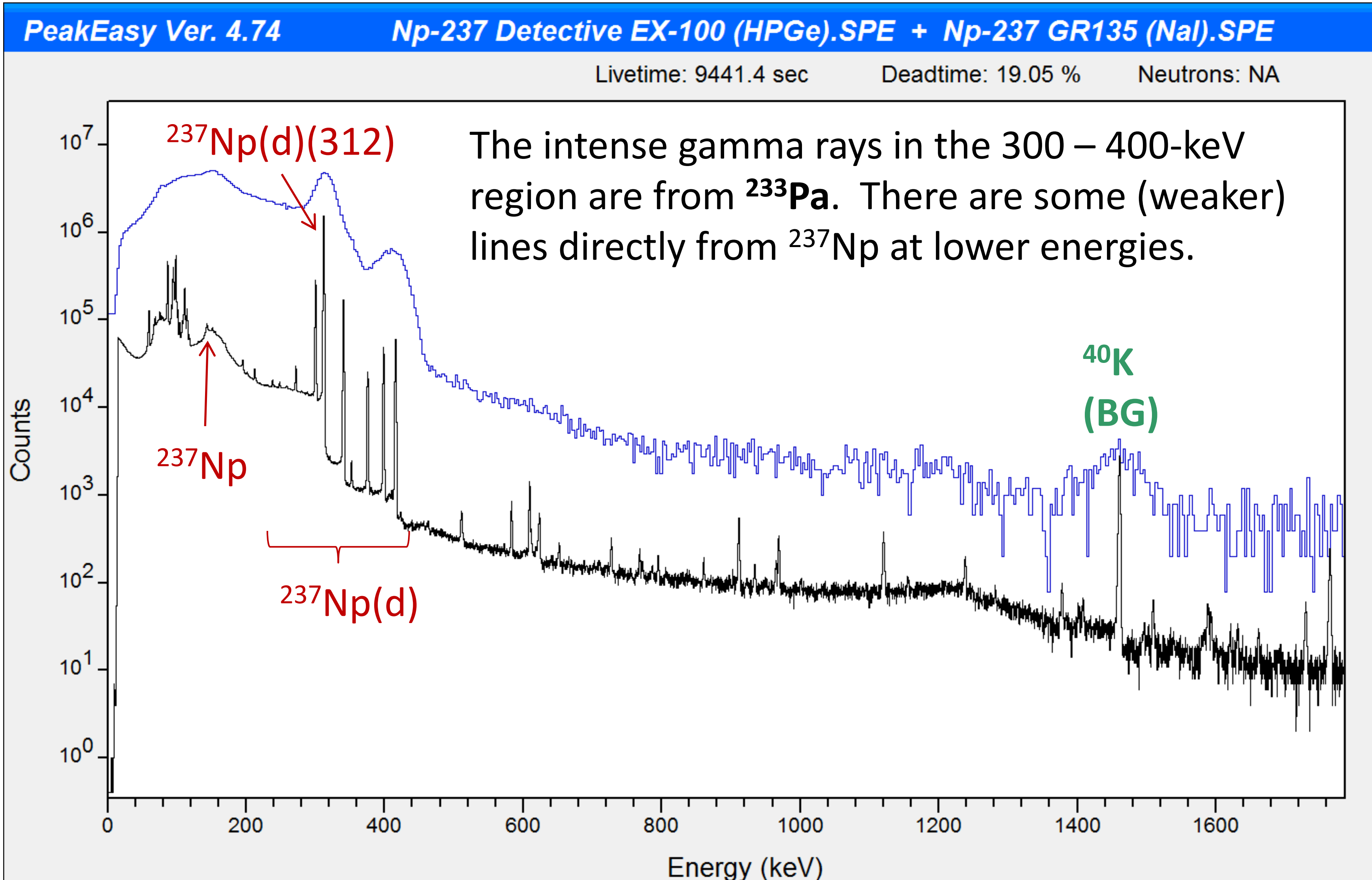
- Half Life: 2.14 x 10^6 years
- Critical Mass: ~60 kG (fast neutrons only)
- ^{237}Np is used in ^{238}Pu production
- **Major gamma rays (e.g. 312 keV) are from daughter ^{233}Pa**
- produced through bombardment of ^{235}U with neutrons or the decay of ^{241}Pu

$$^{235}_{92}U + n \rightarrow ^{236}_{92}U$$

$$^{236}_{92}U + n \rightarrow ^{237}_{92}U \overset{\beta-}{\underset{6.75d}{\rightarrow}} ^{237}_{93}Np$$



^{237}Np Spectrum

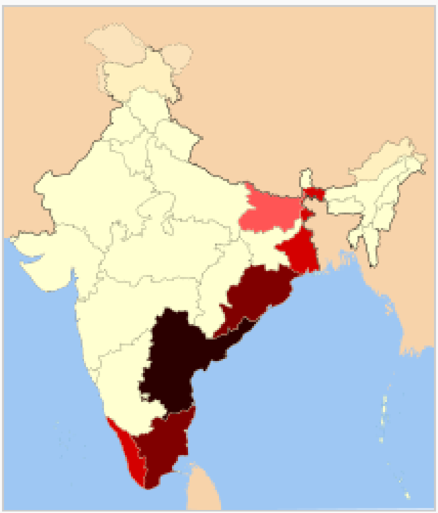


The intense gamma rays in the 300 – 400-keV region are from **^{233}Pa**. There are some (weaker) lines directly from ^{237}Np at lower energies.

Energy from Thorium

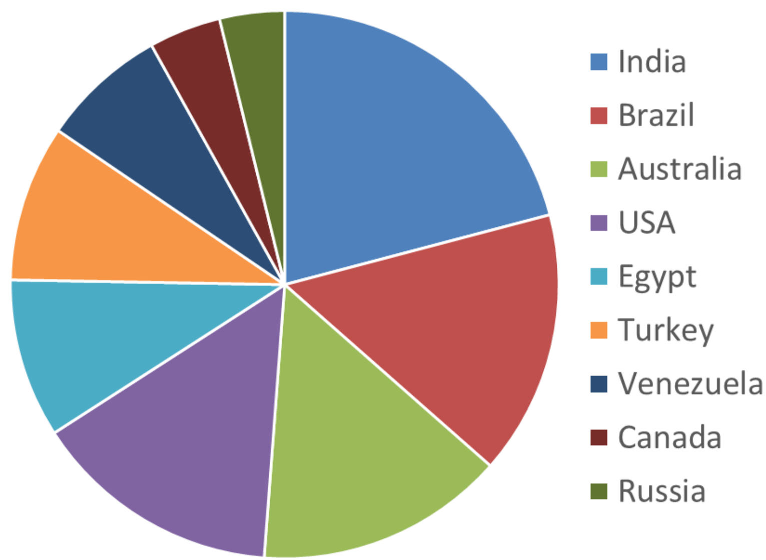
- ^{232}Th is not fissile but it is fertile and ~3x more abundant than U
- Obtain ThO_2 from monazite (6 – 12 % Th typically)
- Put it around a reactor to make ^{233}U, which is fissile

$$^{232}_{90}Th + n \rightarrow {}^{233}_{90}Th \xrightarrow[22m]{\beta-} {}^{233}_{91}Pa \xrightarrow[27d]{\beta-} {}^{233}_{92}U$$



India's thorium is mostly found in a contiguous belt formed by its eastern coastal states.
2012 reserve estimates:[9]
- 35% (Andhra Pradesh, excluding Telangana)
- 15–20% (Tamil Nadu, Odisha)
- 10–15% (Kerala, West Bengal)
- 0–5% (Bihar)

Thorium Reserves: Top 10 Nations 2014

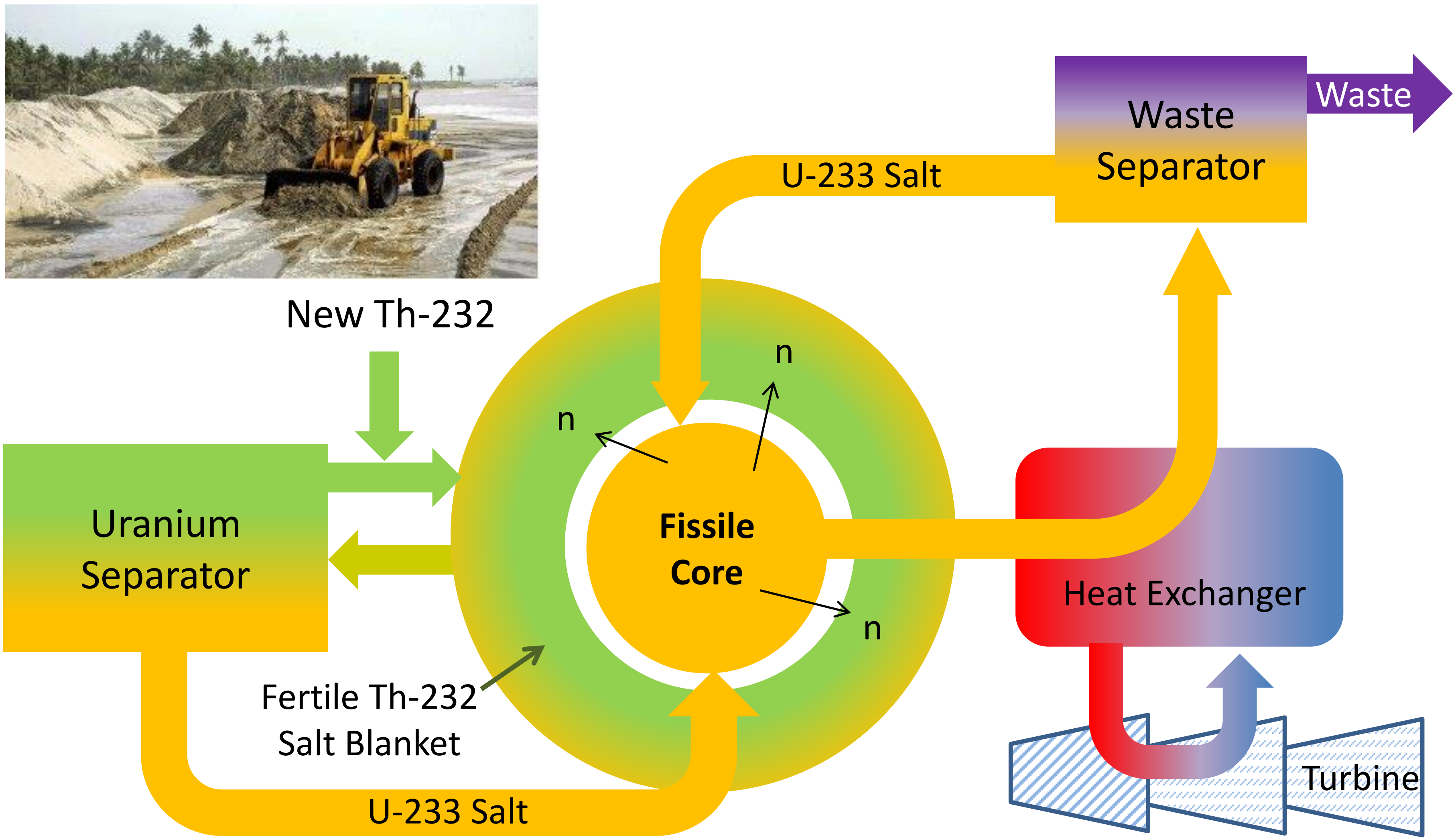


(These numbers vary depending on the source)

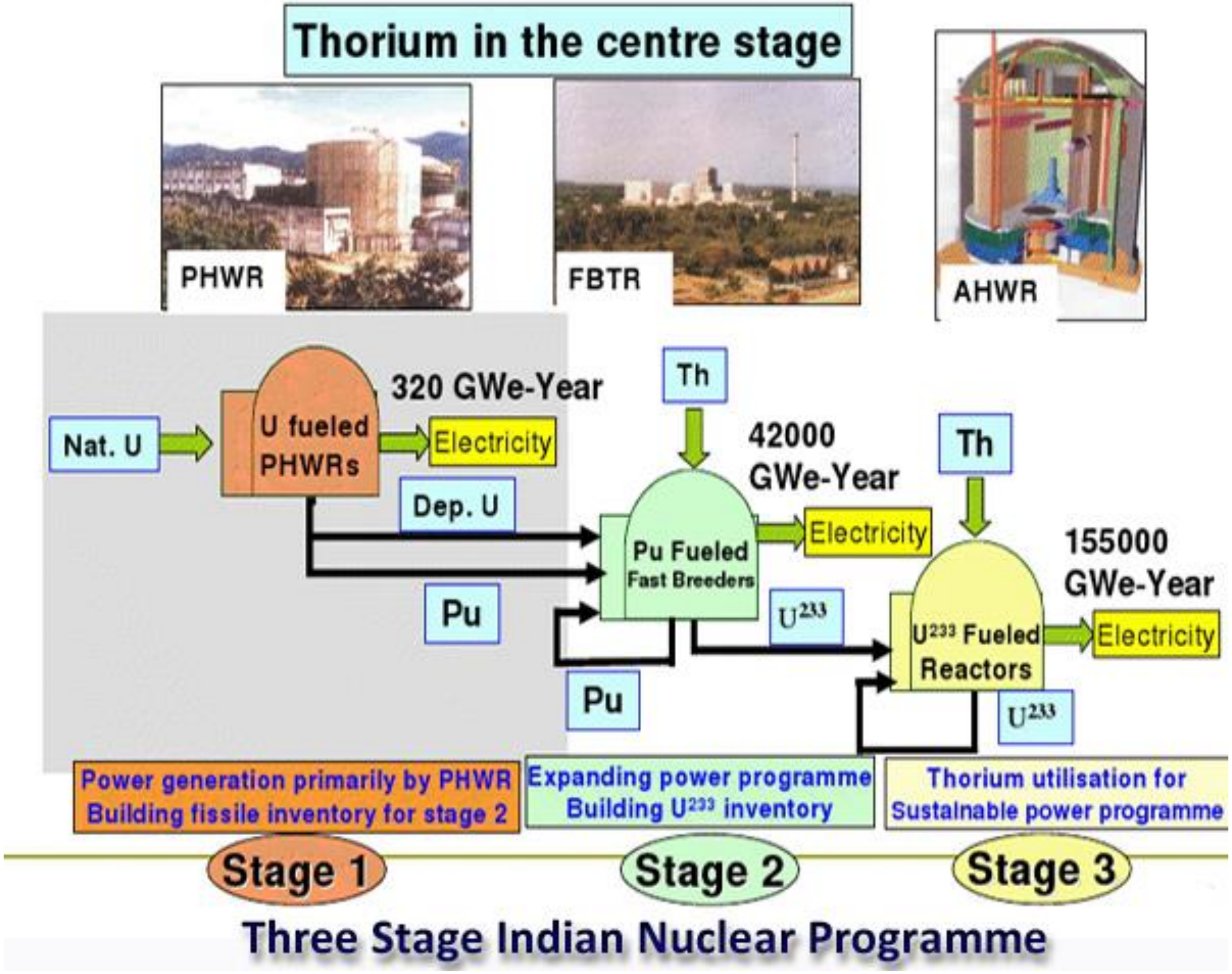
Question Time!

- A typical fresh MOX spectrum:
  - A) looks like 50/50 for gammas from U/Pu
  - B) is complicated by fission product peaks
  - C) can’t be measured due to high dead time
  - D) looks mostly like plutonium

Liquid Fluoride Thorium Reactor



India's Approach to Using Thorium



http://www.barc.gov.in/rcaindia/rca_indian_expertise_energy_1.html#India%27s

But you also get ^{232}U!

$$^{232}_{90}Th + n \rightarrow ^{233}_{90}Th \xrightarrow[22m]{\beta-} ^{233}_{91}Pa \xrightarrow[27d]{\beta-} ^{233}_{92}U + n \rightarrow ^{232}_{92}U + 2n$$

$$^{232}_{90}Th + n \rightarrow ^{233}_{90}Th \xrightarrow[22m]{\beta-} ^{233}_{91}Pa + n \rightarrow ^{232}_{91}Pa + 2n \xrightarrow[1.3d]{\beta-} ^{232}_{92}U$$

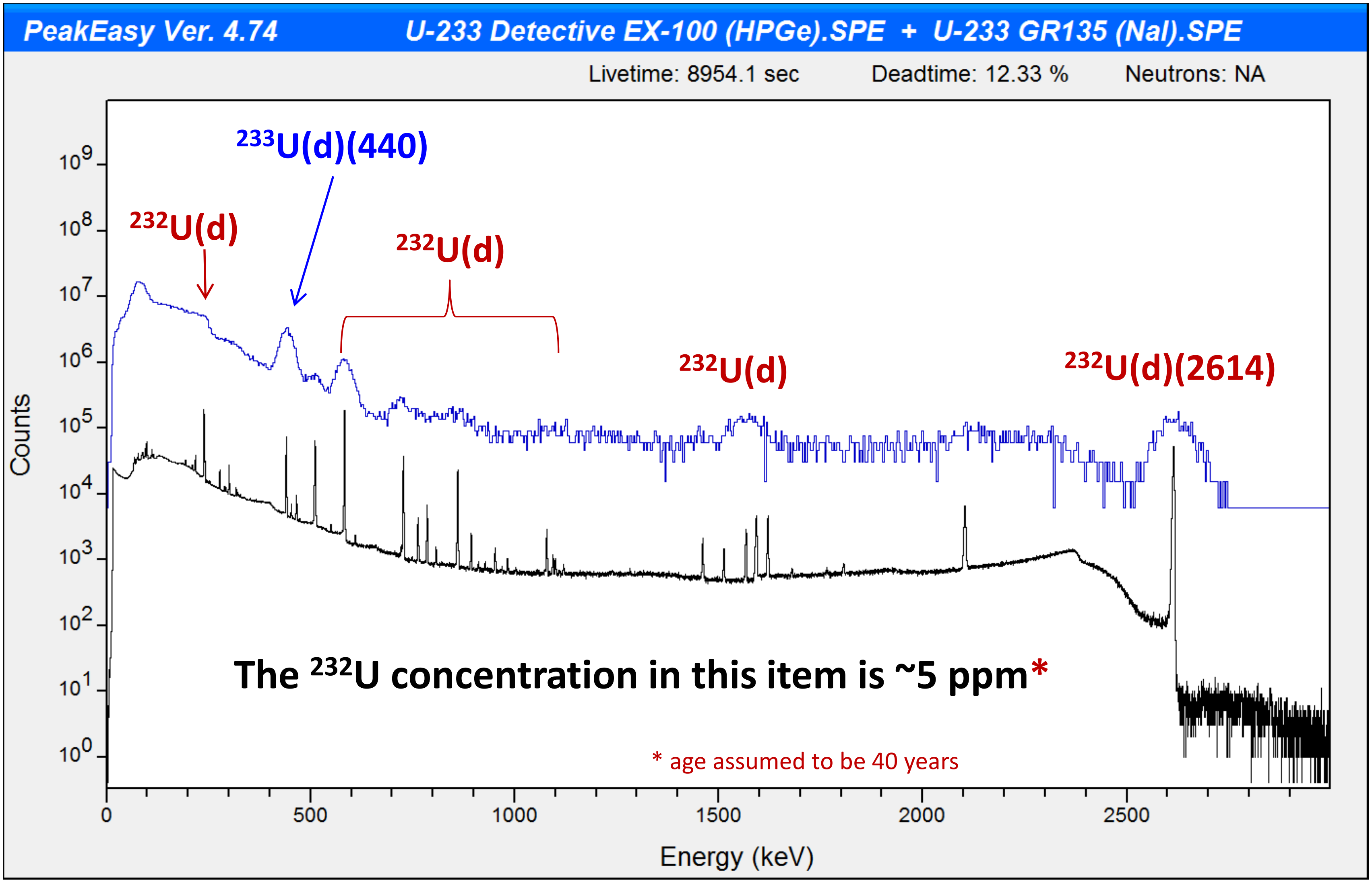
$$^{232}_{90}Th + n \rightarrow ^{231}_{90}Th + 2n \xrightarrow[1.0d]{\beta-} ^{231}_{91}Pa + n \rightarrow ^{232}_{91}Pa \xrightarrow[1.3d]{\beta-} ^{232}_{92}U$$

Approximate Rules of Thumb for U233/U232 Dose Rate

Age [y]	Dose Rate [R/h/kg] @ 1 ft
1	3
10	10

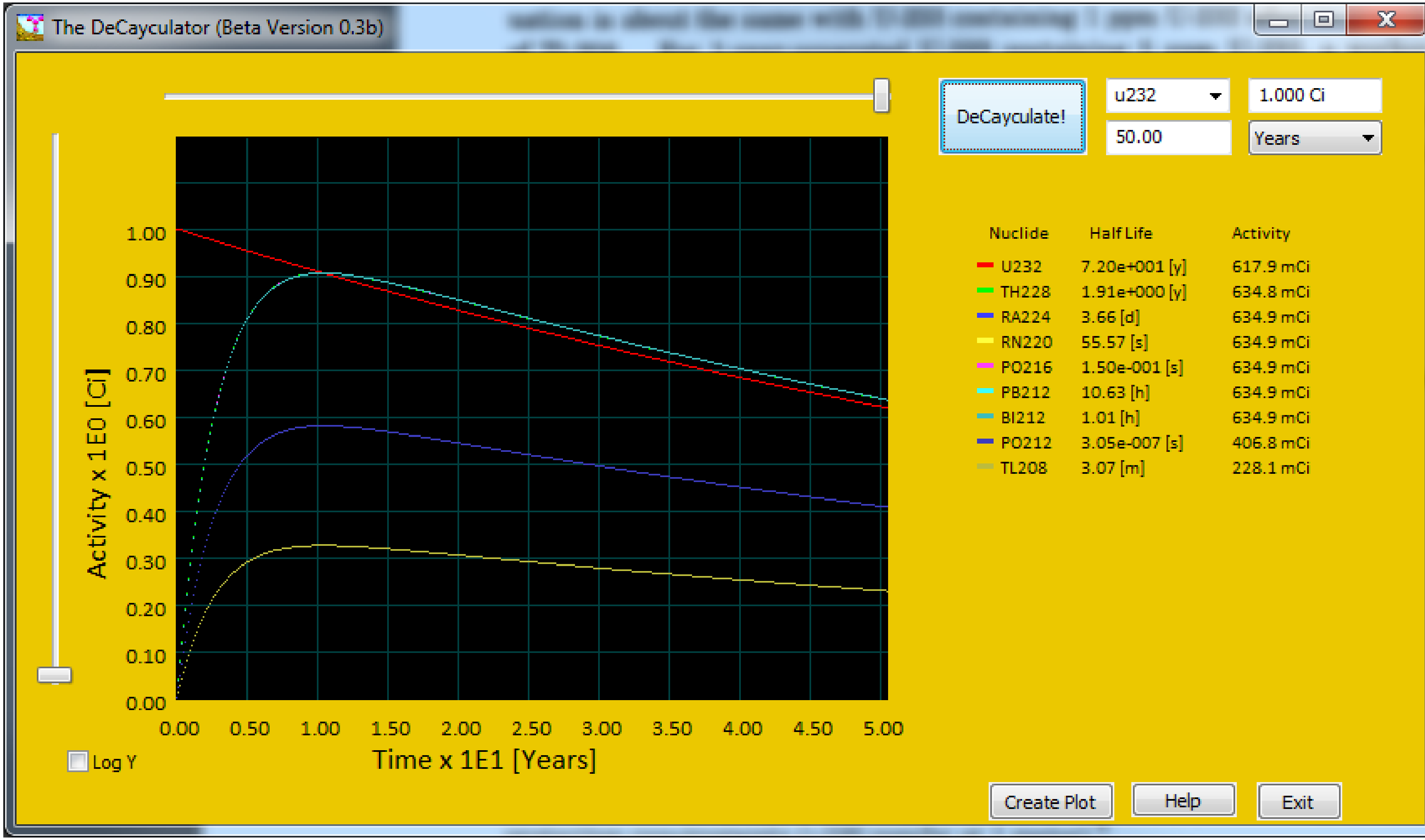
Owen, F.E., "Beta and Gamma Dose From U-232 in U-233", 1964

^{233}U Spectrum

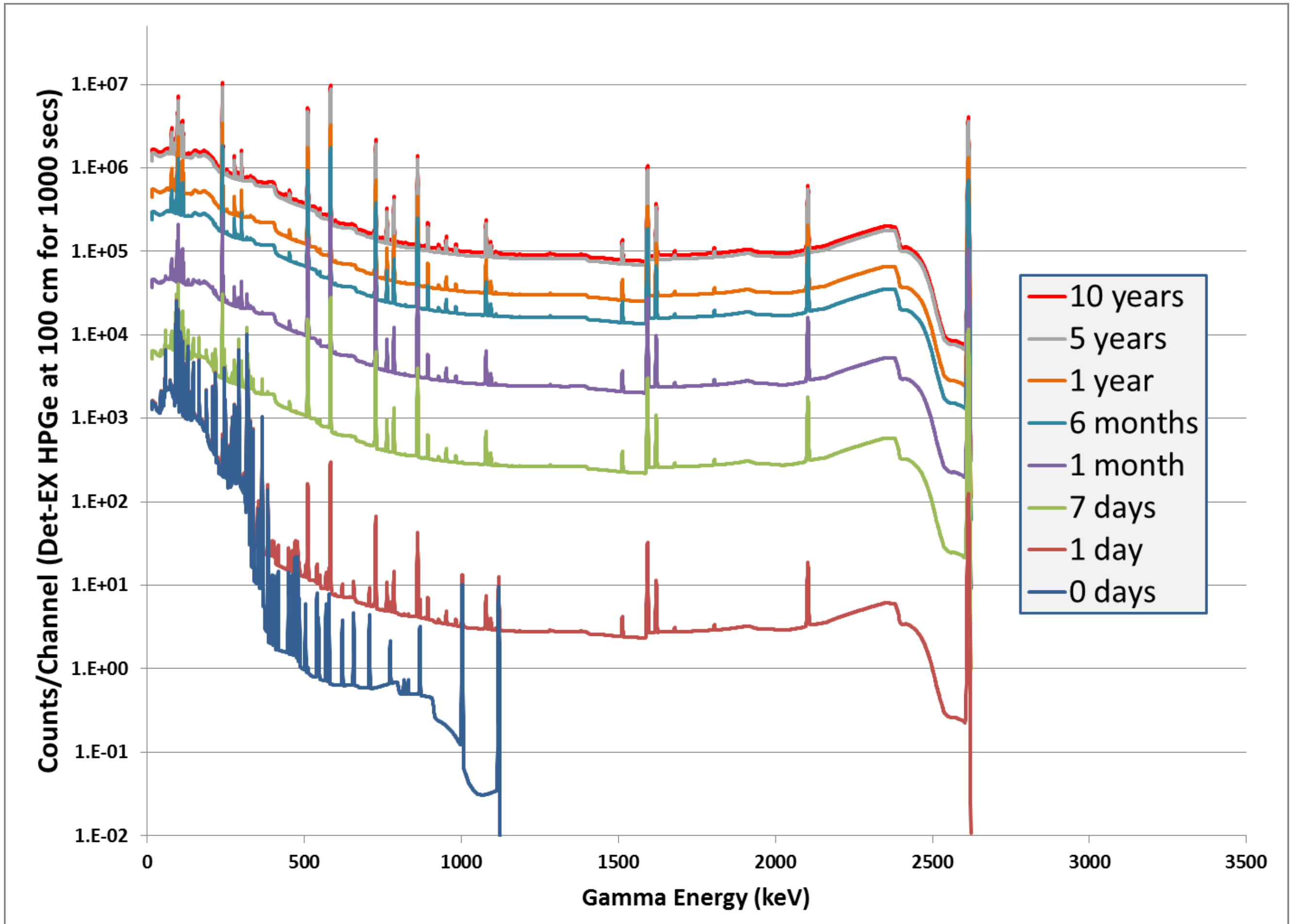


^{232}U Progeny

Max dose from ^{232}U is after about 10 years.



$^{233}U/^{232}U$ Spectra over Time



Modeled high-resolution gamma spectra for a 1-kg metal sphere of U-233 with U-232 at 100 ppm, for an Ortec Detective-EX detector at 100 cm for 1000 seconds.*

* Wimer, N. Et al., “Field Detection and Identification of U-233: Technical Challenges and Opportunities”, Lawrence Livermore National Laboratory Document LLNL-TR-661514