The Decay of Radiochemistry

and

The Decay of Don Wiles

A short tour through the

History of Radiochemistry

Canadian Nuclear Society
Ottawa, 19 March, 2009

Radium discovery and Development

Artificial Radionuclides

Nuclear Fission

Applications and new Discoveries

What's Next?

Early People involved

Henri Becquerel
Marie Skladowska
Pierre Curie
André DeBierne

Early Assistants:
Bertha Karlik, Elisabeth Rona, Ellen Gleditsch

Discoveries:

Radiation and its behaviour
New elements and their purification
Medical uses of radiation

Later People involved

Kasimir Fajans, Fritz Paneth, Frederick Soddy, George de Hevesy

Radium discovery and Development

Artificial Radionuclides

Nuclear Fission

Applications and new Discoveries

What's Next?

1932-34 was a time of Major Advance

The Neutron was discovered

The Cyclotron invented

Nuclear transformations were started

Fission was seen but not recognized

People involved

Fermi, Joliot, Hahn

$^{23}Na\ (n,\gamma)\ ^{24}Na \rightarrow\ ^{24}Mg$

$^{238}U\ (n,\gamma)\ ^{239}U \rightarrow\ ^{239}X$

They found many 'isotopes'

In fact, it was nuclear fission!

Discovery of the Missing Elements

43 Perrier, Segre - 1937

61 Marinski, Glendennin, Coryell - 1945

85 Corson ... Segre - 1940

87 Perey - 1939

And the creation of new ones

Neptunium: MacMillan, Starke

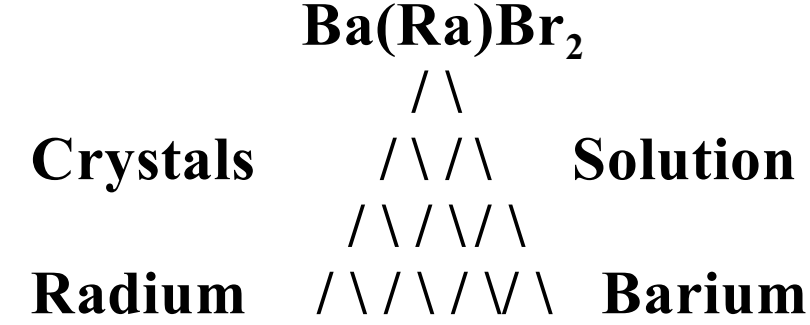
Plutonium:

Berkeley, Dubna, Darmstadt

Many more (110 now?)

Meanwhile back at Port Hope

Fractional Crystallization of Radium
By Marie Curie's procedure:



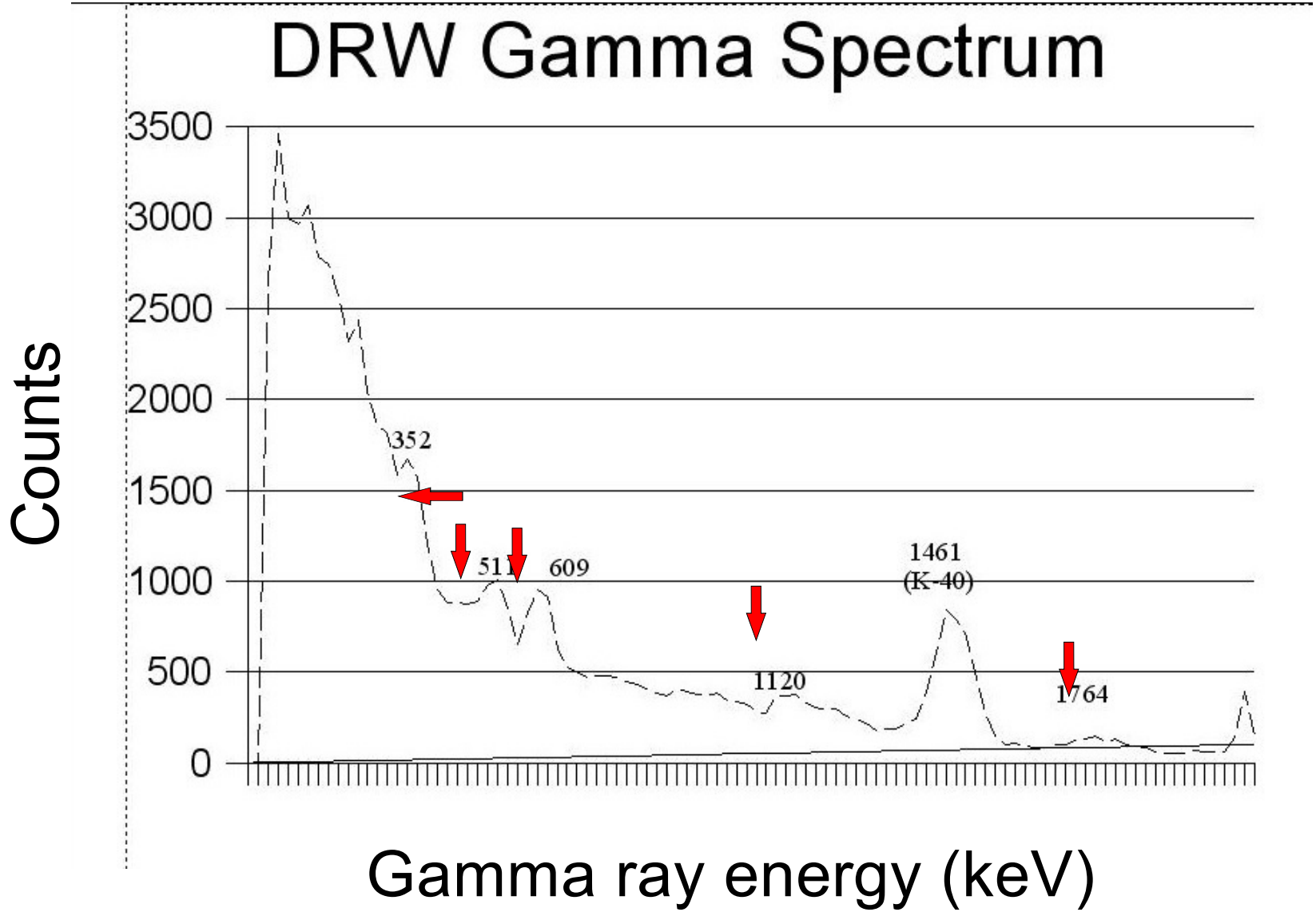
The Radiation was Intense

Monday Mornings the quartz crucibles were brown

On being calcined, the solid glowed violet

The Radium flame test is Red

DRW Fingerprints


DRW Gamma Spectrum
Counts
3500
3000
2500
2000
1500
1000
500
0
352
511
609
1120
1461
(K-40)
1764
Gamma ray energy (keV)

Radium discovery and Development
Artificial Radionuclides
Nuclear Fission
Applications and new Discoveries
What's Next?

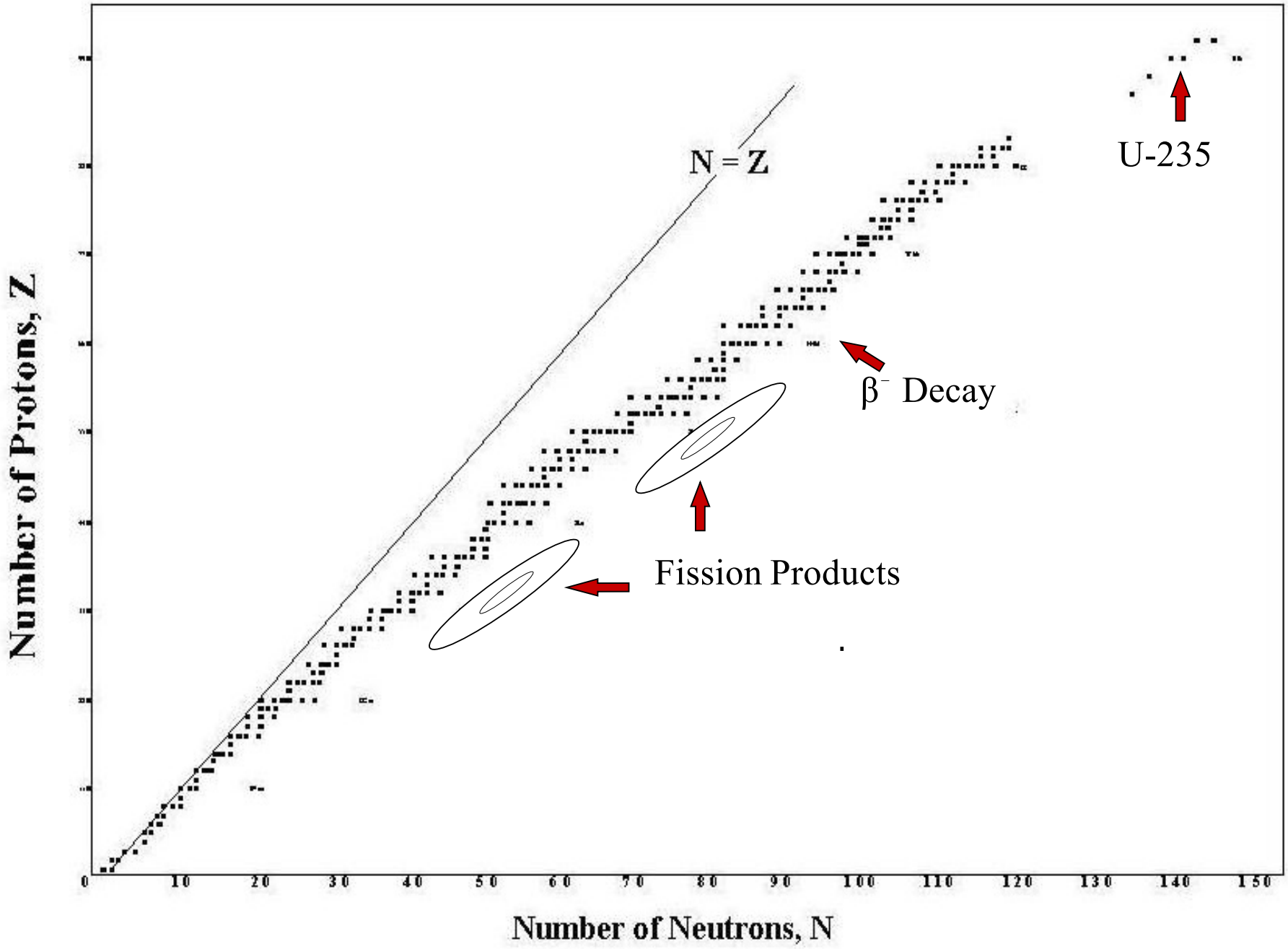
People Involved

Hahn, Meitner, Strassmann

Seaborg, Sugarman, Coryell

Thode

Wilkinson, Harvey,
Grummitt, Yaffe

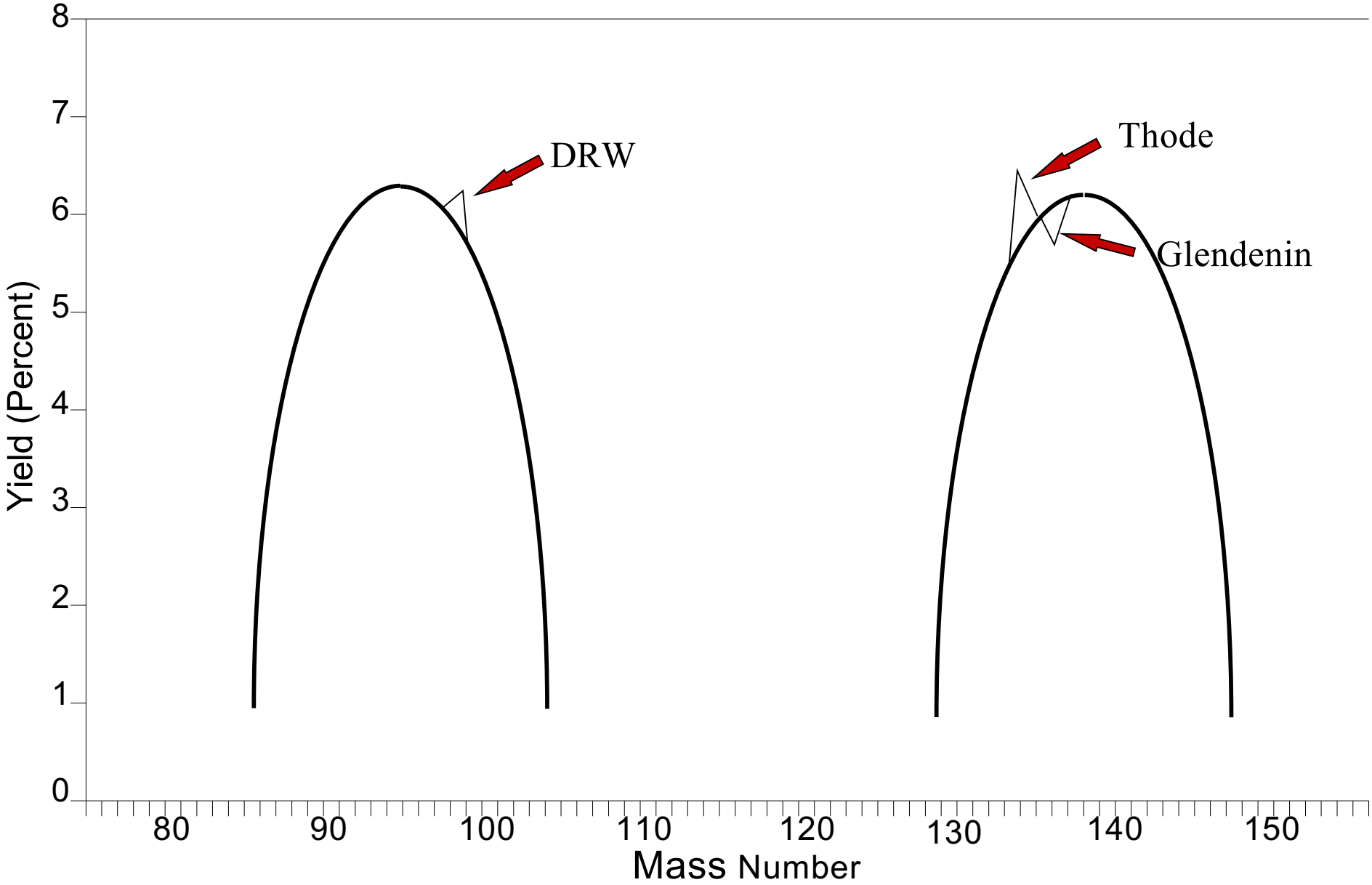

N = Z
U-235
β⁻ Decay
Fission Products
Number of Protons, Z
Number of Neutrons, N
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150

Fission Yields

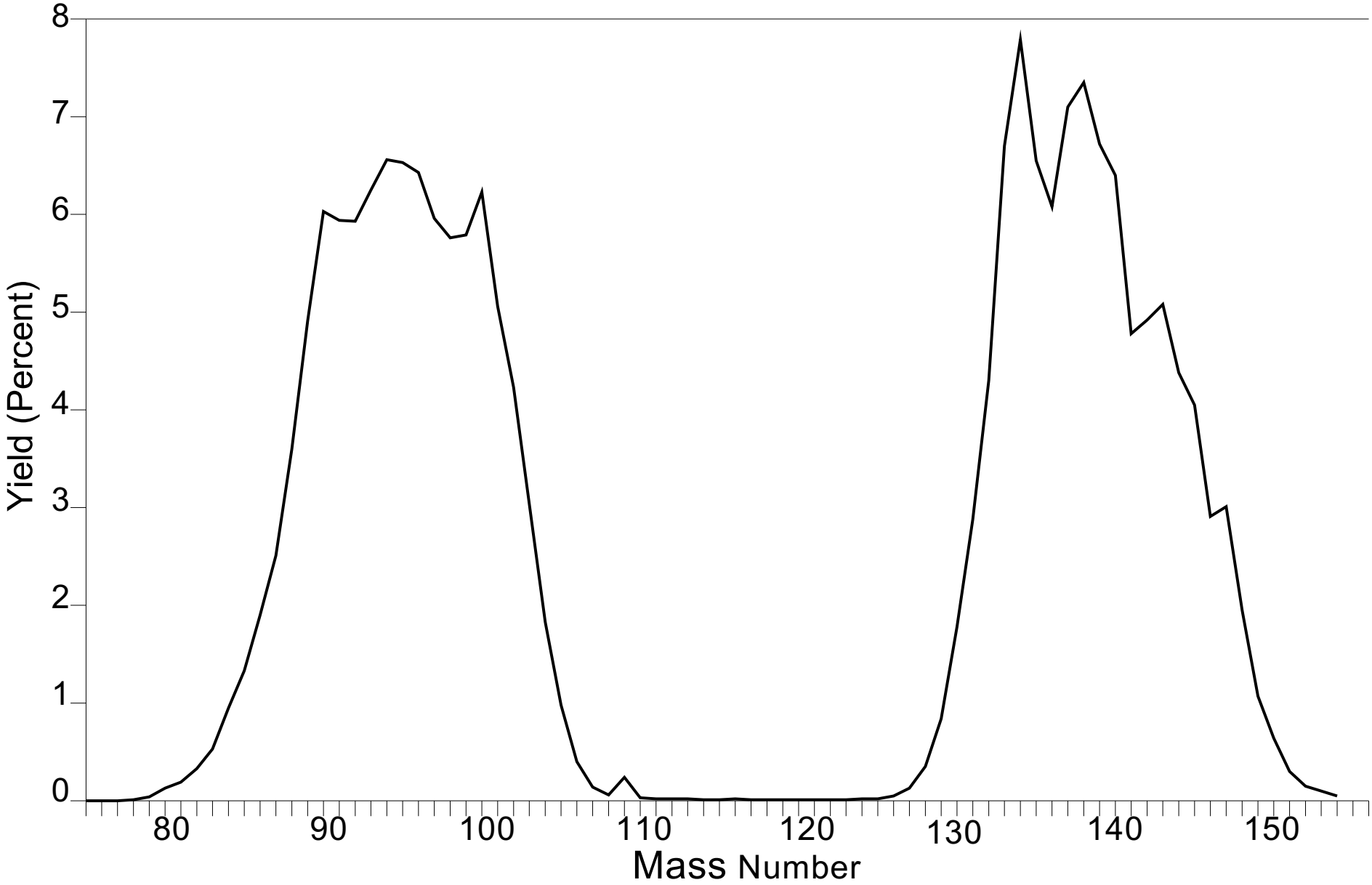
became the order of the day

$$^{140}\mathrm{I} \rightarrow {}^{140}\mathrm{Xe} \rightarrow {}^{140}\mathrm{Cs} \rightarrow {}^{140}\mathrm{Ba} \rightarrow {}^{140}\mathrm{La} \rightarrow {}^{140}\underline{\mathrm{Ce}}$$

Fission Yields in U-235



Fission Yields in U-235



Secrecy!!

Senator Joseph McCarthy

Radiochemistry in Norway

Identified Tin-132

Radiochemistry at Carleton

Fifty years.

Several different areas

Taught many students

Radiation Sources:

One would need:

Reactors
Cyclotrons
Neutron Sources

Radium-Beryllium

Photo-neutron sources
Sb-124 - Be

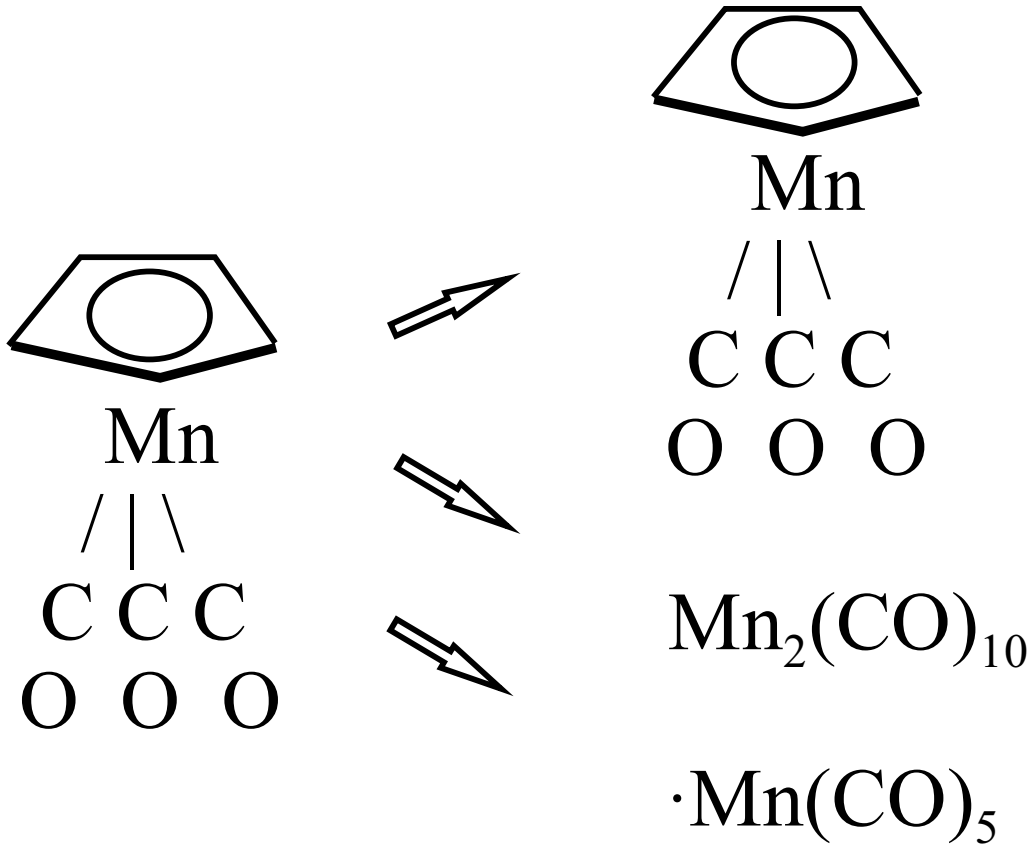
Radium discovery and Development
Artificial Radionuclides
Nuclear Fission
Applications and new Discoveries
What's Next?

Hot atom reactions became important

What happens to the chemistry of a

Newly-radioactive atom?

Too difficult for current experiments and theories

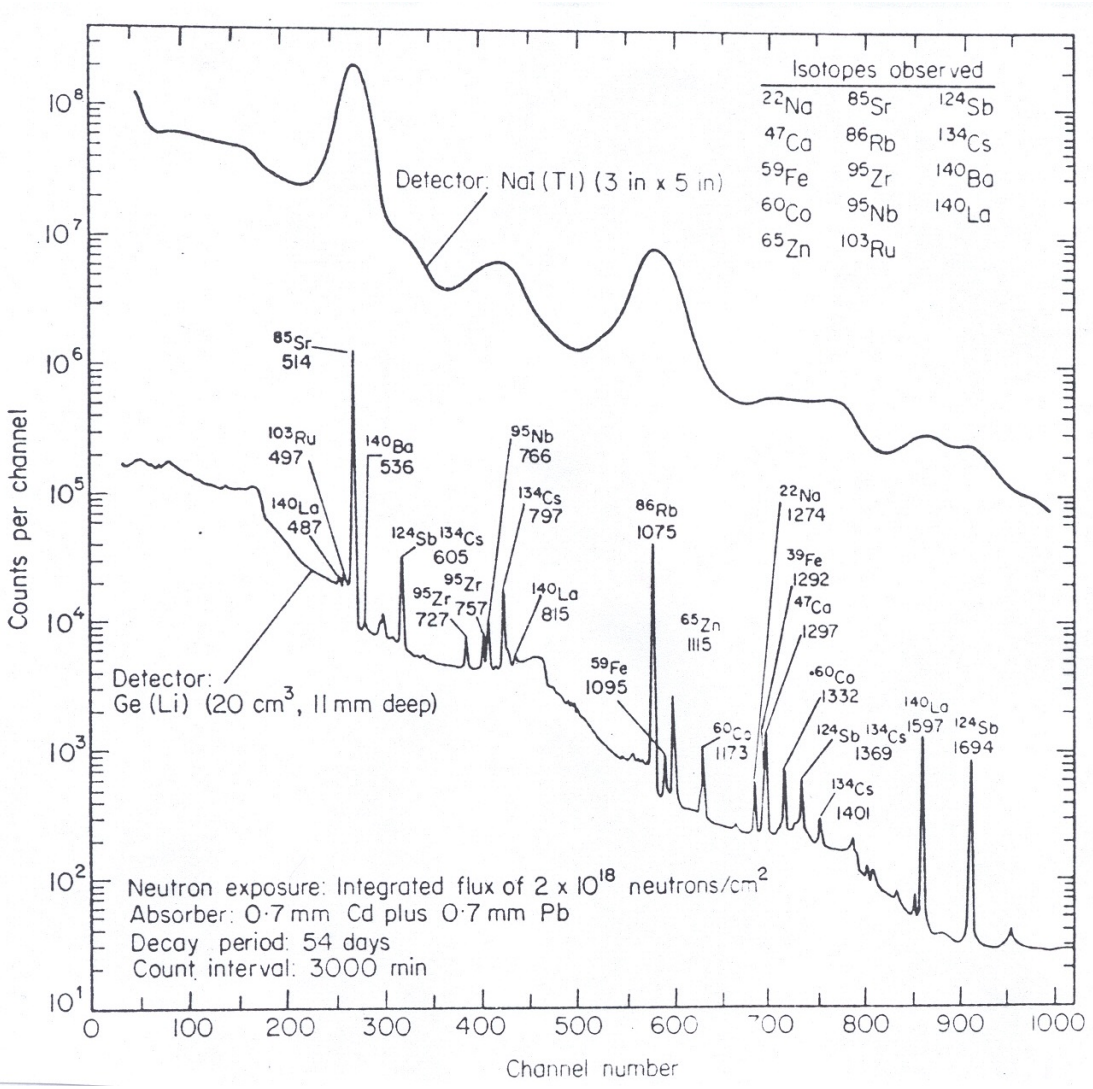


What is happening?

Activation Analysis was a Big Thing

in Archaeology

Especially with high-resolution Detectors


Isotopes observed
22Na 85Sr 124Sb
47Ca 86Rb 134Cs
59Fe 95Zr 140Ba
60Co 95Nb 140La
65Zn 103Ru
Detector: NaI (Tl) (3 in x 5 in)
Detector: Ge (Li) (20 cm3, 11 mm deep)
85Sr 514
103Ru 497
140Ba 536
140La 487
124Sb 134Cs 605
95Nb 766
134Cs 797
95Zr 727
95Zr 757
140La 815
86Rb 1075
65Zn 1115
59Fe 1095
22Na 1274
39Fe 1292
47Ca 1297
60Co 1332
60Co 1173
124Sb 134Cs 1369
140La 1597
124Sb 1694
134Cs 1401
Neutron exposure: Integrated flux of 2 x 10^18 neutrons/cm^2
Absorber: 0·7 mm Cd plus 0·7 mm Pb
Decay period: 54 days
Count interval: 3000 min
Counts per channel
Channel number
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
0
100
200
300
400
500
600
700
800
900
1000

But we had a better idea!

Nitrogen in proteins

$$^{15}N\ (n,\gamma)\ ^{16}N$$

Reactor neutrons have too high energy:

$$^{16}O\ (n,p)\ ^{16}N$$

Use a photoneutron source

$^{9}Be\ (\gamma,n)\ ^{8}Be$

Using ^{124}Sb, (E_γ = 1.76 MeV) the maximum neutron energy would be about 25 KeV.

We had the largest neutron source in the world

The flux was only 10^{8}

Not strong enough

Mössbauer Spectroscopy

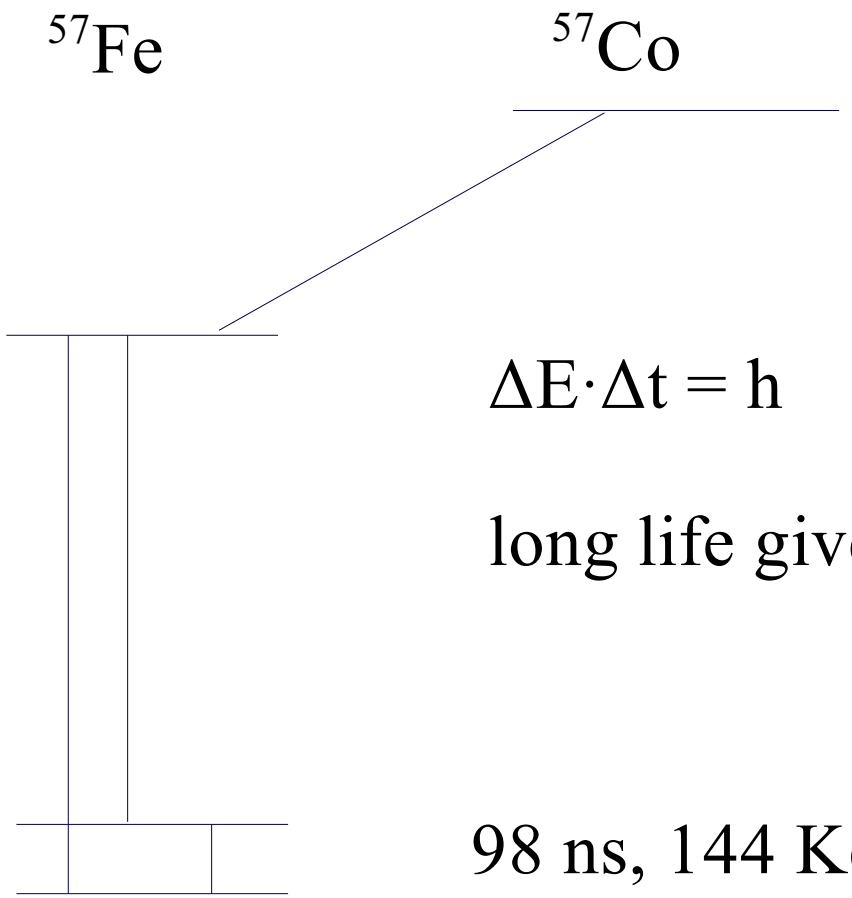
became the thing to do

$$\mathbf{E_\gamma = E - recoil}$$

$$\mathbf{\Delta E = h/\Delta t}$$

$\mathbf{\Delta E} \approx$ Chemical Energies

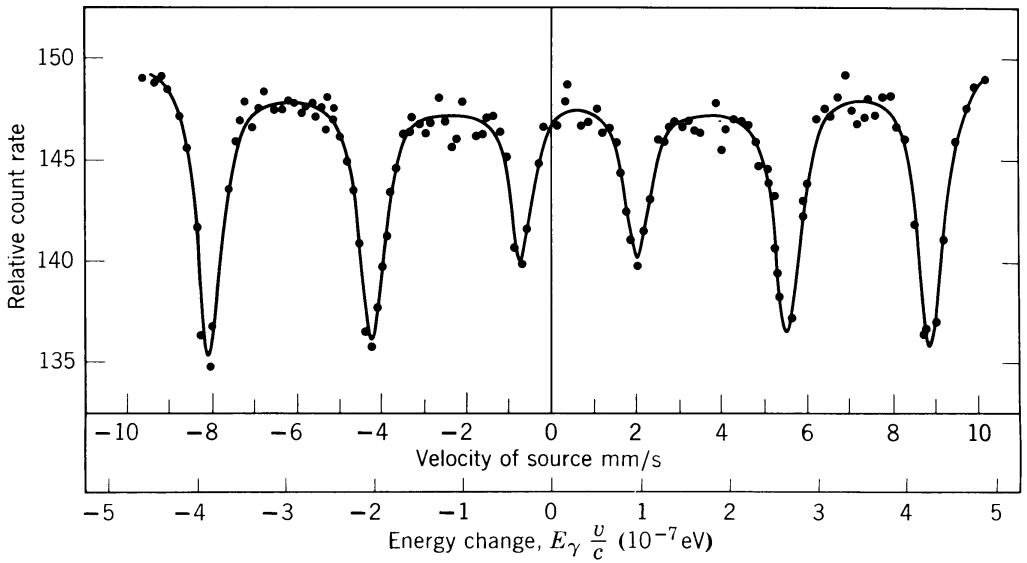
$\mathbf{\Delta E} \approx$ Doppler Energies

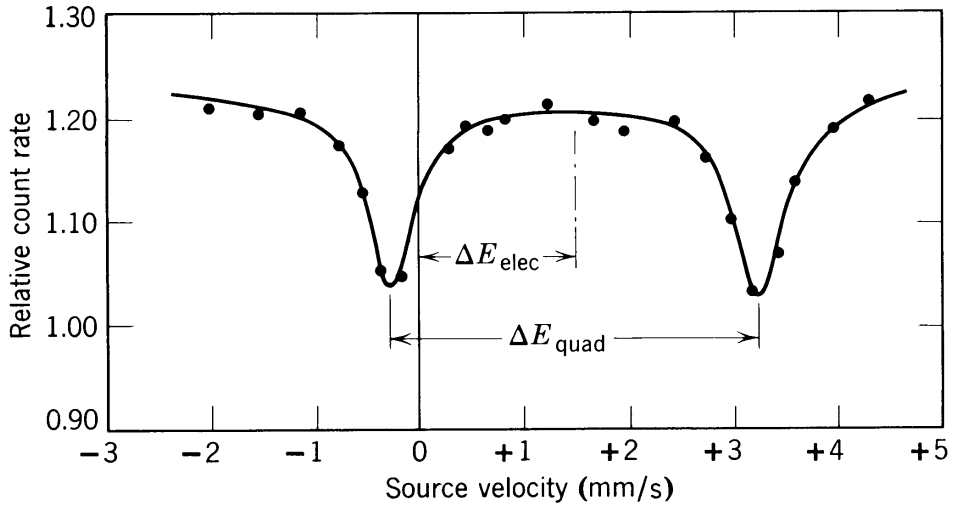


$\Delta E \cdot \Delta t = h$

long life gives precise energy

98 ns, 144 KeV


150
145
140
135
Relative count rate
−10 −8 −6 −4 −2 0 2 4 6 8 10
Velocity of source mm/s
−5 −4 −3 −2 −1 0 1 2 3 4 5
Energy change, $E_\gamma \frac{v}{c}$ (10^{-7} eV)


1.30
1.20
1.10
1.00
0.90
Relative count rate
ΔE_{elec}
ΔE_{quad}
−3 −2 −1 0 +1 +2 +3 +4 +5
Source velocity (mm/s)

Measuring alpha particles

became the thing to do

Led to environmental Radiochemistry

Nuclear Waste Disposal

Radium discovery and Development
Artificial Radionuclides
Nuclear Fission
Applications and new Discoveries
What's Next?

Synthesis of radiopharmaceuticals

Development of new irradiation techniques

(theraspheres)

Radiochemistry has Changed

Radiochemistry has now become a servant to other fields of study:

Pharmaceutical and Medicinal Radiochemistry

Environmental Radiochemistry

Chemistry of fission products in the environment

Iodine-129
Chlorine-36
Technetium-99
Others?

How do we counter public apprehension?

Where would one go now
To Study in Radiochemistry?

La Dernière Classe

Environmental Radiochemistry?