



# Radiochemistry Webinars

## Introduction to Nuclear Forensics



*In Cooperation with our University Partners*



## Meet the Presenter...

*Dr. Walter Loveland*



Dr. Walter Loveland is a Professor of Chemistry at Oregon State University. He holds a B.S. degree in chemistry from The Massachusetts Institute of Technology and a Ph.D. in chemistry from the University of Washington. His current research is aimed at understanding the synthesis of the heaviest nuclei and the synthesis of new neutron-rich heavy nuclei using radioactive beams and multi-nucleon transfer reactions. He is also involved in the study of the fission process especially the total kinetic energy release and neutron emission. Dr. Loveland has published over 215 papers in the refereed literature and is the author of several monographs and textbooks in nuclear and radiochemistry. Dr. Loveland is a distinguished recipient of the Sigma Xi Award for Research, Beaver Champion Award, and the F.A. Gilfillan Memorial Award for Distinguished Scholarship in Science at Oregon State University, the Outstanding Referee Award of the American Physical Society and the American Chemical Society 2014 Glenn T. Seaborg Award in Nuclear Chemistry.



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# Nuclear Forensics An Introduction

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Oregon State University



**National Analytical Management Program  
(NAMP)**

TRAINING AND EDUCATION SUBCOMMITTEE

# What is Nuclear Forensics?

- “The investigation of nuclear materials to find evidence of the source, the trafficking, and the enrichment of the material” Wikipedia
- “A methodology that aims at re-establishing the history of nuclear material of unknown origin” , Mayer et al, 2011
- K.J. Moody, P.M. Grant and I.D. Hutcheon, Nuclear Forensic Analysis, 2nd Edition (CRC Press, Boca Raton, 2015) --The “bible” of the field
- The challenge of timeliness

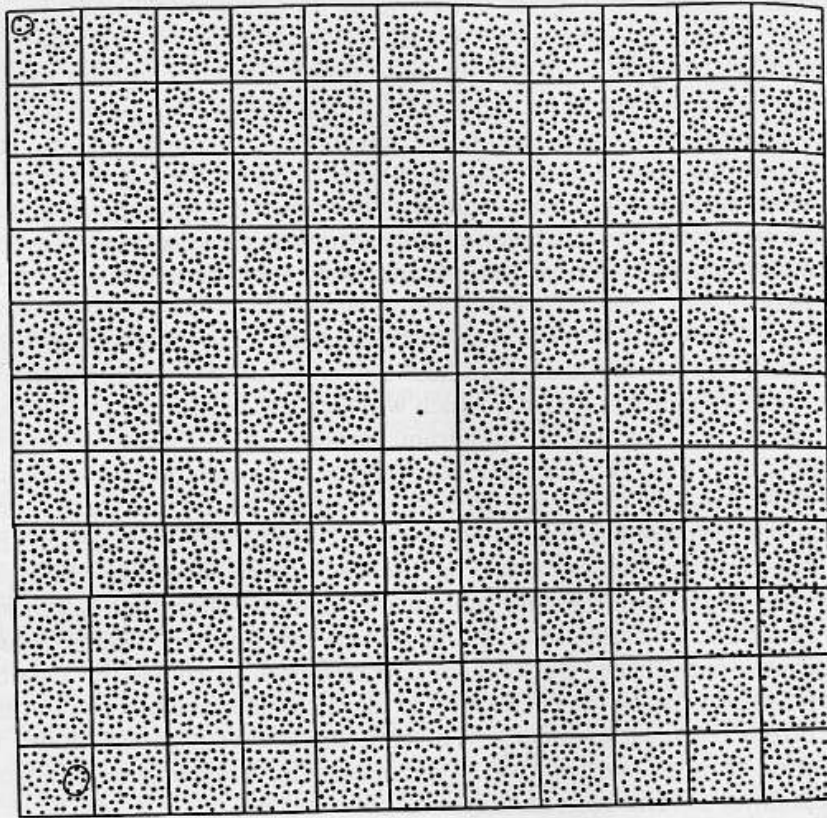
# A Glossary

- Nuclear Material = Radioactive material with attention to Special Nuclear Materials ( $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ )
- IND Improvised Nuclear Device
- RDD Radiologic Dispersal Device = Dirty Bomb = Weapon of Mass Disruption
- HEU  $> 90\%$   $^{235}\text{U}$
- LEU  $< 20\%$   $^{235}\text{U}$

# The “Realities”

- Nuclear Power and Pu
  - 1 GWe reactor generates 200 kg Pu/yr (~50 bombs)
  - World production = 70 t Pu/yr
- “The Arms Race”
  - US 100 t Pu, ~1000 t HEU SU was similar
  - In 2011, the US had 5000 warheads and Russia had 8000-10000
  - The “nuclear club” also includes France, China, Britain, Israel, India, Pakistan, N. Korea
- Atmospheric Testing
  - At peak (1962) 72 Mt of fission products were put into the atmosphere/yr.
- Current Situation
  - 2200 t of Pu (90-100 t military)
  - 3800 t fissile material, ~2000 t in weapons usable form





**Figure 10.1** A realistic view of the world armaments situation. The chart shows the world's current firepower in terms of the firepower of World War II. The dot in the center square represents all the firepower of World War II (including the atomic bombs dropped on Hiroshima and Nagasaki): three megatons. The other dots represent the world's present nuclear weaponry. This comes to 18,000 megatons, which equals 6000 World War IIs. The US (and allies) and the SU (and allies) share this firepower approximately equally. The top lefthand circle enclosing nine megatons represents the weapons in just one Poseidon submarine. This is equal to the firepower of three World War IIs and is enough to destroy over 200 of the Soviet Union's largest cities. We have 31 such submarines and ten similar Polaris submarines. The bottom lefthand circle enclosing 24 megatons represents one new Trident submarine with the power of eight World War IIs: enough to destroy every major city in the Northern hemisphere. The Soviets have similar levels of destructive power. Just two squares on this chart (300 megatons) represent enough firepower to destroy all the large and medium size cities in the entire world (from Hof 85).

Source: D. R. Hofstadter, *Metamagical Themas*, (Basic, New York, 1985)

# A Brief Incomplete History

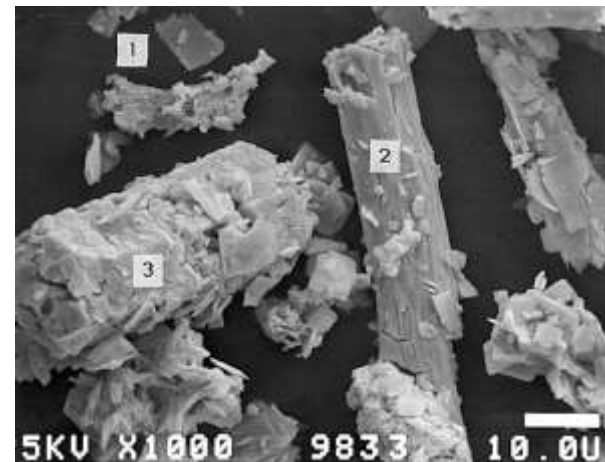
- 1944—Detection of  $^{133}\text{Xe}$  in the atmosphere as a signature of German nuclear program
- Monitoring of Russian nuclear program in post WWII

“Joe-1 test in August 1949”



U.S. Weather Bureau Report on Alert Number 112 of the Atomic Detection System, 29 September 1949PSF: Subject File 1940-1953, National Security Council – Atomic Files, Box 173, Folder: “Atomic Bomb: Reports,” Harry S. Truman Presidential Library

1994 Munich Plutonium  
Intercepted at Munich Airport in August 1994 on a Lufthansa flight from Moscow  
363 grams of plutonium (87% Pu-239) and  
122 grams of uranium (560 grams of  
plutonium and uranium oxide)



K.-R. Lü tzenkirchen, CSI: Karlsruhe, Actinide Research Quarterly, 2007



# Basic Ideas of Nuclear Science

- Types of radioactive decay
- Nuclear decay kinetics
- Masses and binding energies
- Actinide decay schemes
- Nuclear reactions/fission
- Natural radioactivity

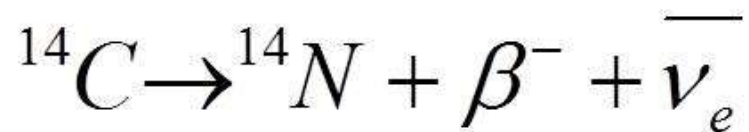
# Types of Radioactive Decay

## Alpha decay ( $\alpha$ )

- Decay by the emission of doubly charged helium nuclei  ${}^4\text{He}^{2+}$ .  $\Delta Z = -2$ ,  $\Delta N = -2$ ,  $\Delta A = -4$
- ${}^{238}\text{U} \rightarrow {}^{234}\text{Th} + {}^4\text{He}$
- All nuclei with  $Z \geq 83$  decay by  $\alpha$ -decay as do some rare earth nuclei
- Alpha emitters are internal radiation hazards having a short range in matter.
- The emitted  $\alpha$ -particles are monoenergetic

# Beta -decay

- Beta decay is a term used to describe three types of decay in which a nuclear neutron (proton) changes into a nuclear proton (neutron). The decay modes are  $\beta^-$ ,  $\beta^+$  and electron capture (EC).
- $\beta^-$  decay involves the change of a nuclear neutron into a proton and is found in nuclei with a larger than stable number of neutrons relative to protons, such as fission fragments.
- An example of  $\beta^-$  decay is



# Beta decay (cont)

- In  $\beta^-$  decay,  $\Delta Z = +1$ ,  $\Delta N = -1$ ,  $\Delta A = 0$
- Most of the energy emitted in the decay appears in the rest and kinetic energy of the emitted electron ( $\beta^-$ ) and the emitted anti-electron neutrino,  $\bar{\nu}_e$
- The decay energy is shared between the emitted electron and neutrino.
- $\beta^-$  decay is seen in all neutron-rich nuclei
- The emitted  $\beta^-$  are easily stopped by a thin sheet of Al

# Beta decay (cont)

- The second type of beta decay is  $\beta^+$  (positron) decay.
- In this decay,  $\Delta Z = -1$ ,  $\Delta N = +1$ ,  $\Delta A = 0$ , i.e., a nuclear proton changes into a nuclear neutron with the emission of a positron,  $\beta^+$ , and an electron neutrino,  $\nu_e$
- An example of this decay is

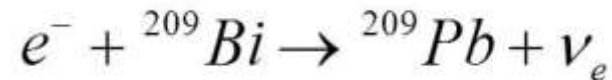


- Like  $\beta^-$  decay, in  $\beta^+$  decay, the decay energy is shared between the residual nucleus, the emitted positron and the electron neutrino.
- $\beta^+$  decay occurs in nuclei with larger than normal p/n ratios. It is restricted to the lighter elements
- $\beta^+$  particles annihilate when they contact ordinary matter with the emission of two 0.511 MeV photons.



# Beta decay (cont)

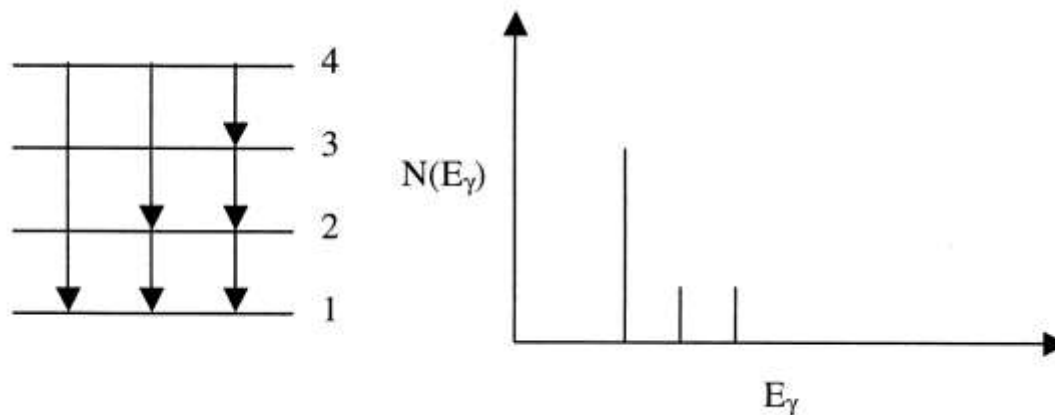
- The third type of  $\beta$  –decay is electron capture decay (EC). In EC decay an orbital electron is captured by a nuclear proton changing it into a neutron with the emission of an electron neutrino.
- An example of this type of decay is



- The occurrence of this decay is detected by the emitted X-ray (from the vacancy in the electron shell).
- It is the preferred decay mode for proton-rich heavy nuclei.

# Electromagnetic decay

- There are two types of electromagnetic decay,  $\gamma$ -ray emission and internal conversion (IC). In both of these decays  $\Delta N = \Delta Z = \Delta A = 0$ , with just a lowering of the excitation energy of the nucleus.
- In  $\gamma$ -ray emission, most of the emitted energy appears in the form of a photon.
- These emitted photons are mono-energetic and have an energy corresponding to almost all of the energy difference between the final and initial state of the system. This is typically depicted as



# Electromagnetic decay (cont.)

- $\gamma$ -rays are the most penetrating nuclear radiation and to attenuate them requires massive shielding. They represent an external radiation hazard.
- The second type of electromagnetic decay is internal conversion. In IC decay, the emitted energy is transferred (radiationlessly) to an orbital electron, ejecting that electron which carries away most of the decay energy.

# Basic Decay Equations

- Radioactive decay is a first order process, ie, the number of decays/s is proportional to the number of nuclei present
- In equation form,  $-dN/dt = \lambda N$  where the constant  $\lambda$  is the decay constant
- ( $A = \lambda N$ )
- Rearranging

$$\frac{-dN}{dt} = \lambda N$$

$$\frac{dN}{N} = -\lambda t$$

$$N = N_0 e^{-\lambda t}$$


where  $N_0$  is the number of nuclei present at  $t=0$ .

If we remember the basic equation relating activity to number of nuclei in a sample,  $A = \lambda N$ , then we can write

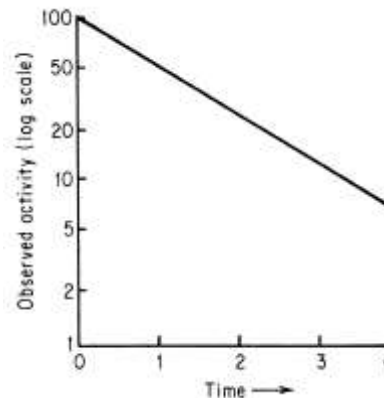
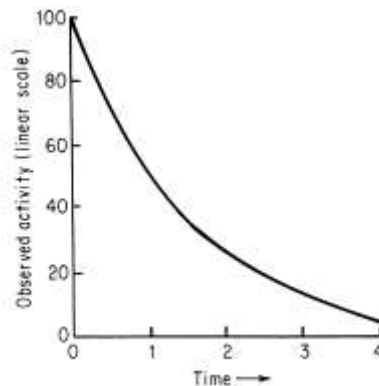
$$A = A_0 e^{-\lambda t}$$

Thus we have two equations that look the same, but have very different meanings

$$N = N_0 e^{-\lambda t}$$

$$A = A_0 e^{-\lambda t}$$


Graphically





# Radioactive Decay Equilibria

Consider  $1 \rightarrow 2 \rightarrow 3 \rightarrow$

rate of change of 2 = rate of production of 2 by decay of 1 - rate of decay of 2

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$

$$dN_2 + \lambda_2 N_2 dt = \lambda_1 N_1 dt$$

$$N_1 = N_1^0 e^{-\lambda_1 t}$$

$$dN_2 + \lambda_2 N_2 dt = \lambda_1 N_1^0 e^{-\lambda_1 t} dt$$

$$e^{\lambda_2 t} dN_2 + \lambda_2 N_2 e^{\lambda_2 t} dt = \lambda_1 N_1^0 e^{(\lambda_2 - \lambda_1)t} dt$$

$$d(N_2 e^{\lambda_2 t}) = \lambda_1 N_1^0 e^{(\lambda_2 - \lambda_1)t} dt$$

$$N_2 e^{\lambda_2 t} \Big|_0^t = \frac{\lambda_1 N_1^0 e^{(\lambda_2 - \lambda_1)t}}{\lambda_2 - \lambda_1} \Big|_0^t$$

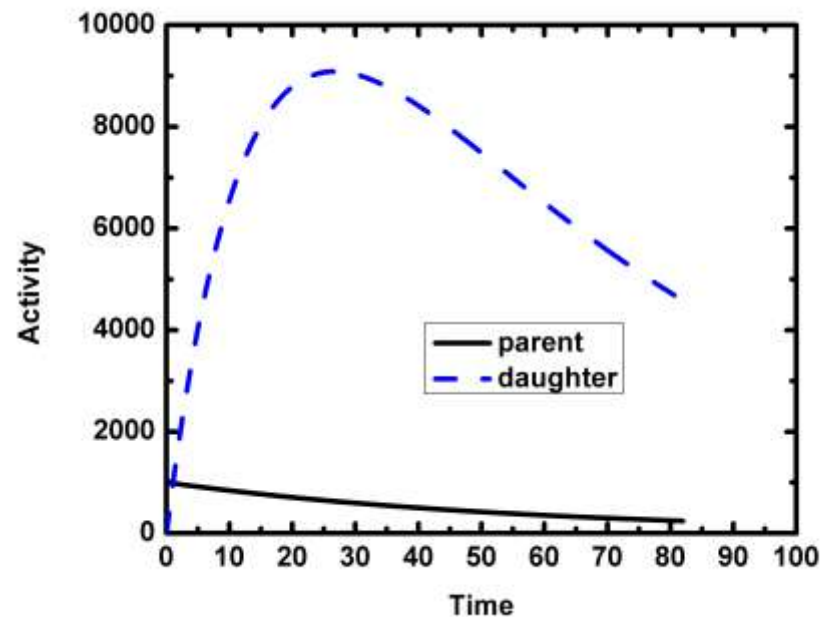
$$N_2 e^{\lambda_2 t} - N_2^0 = \frac{\lambda_1 N_1^0 (e^{(\lambda_2 - \lambda_1)t} - 1)}{\lambda_2 - \lambda_1}$$

$$N_2(t) = \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t}$$

$$A_2(t) = \frac{\lambda_1 \lambda_2 N_1^0}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + A_2^0 e^{-\lambda_2 t}$$

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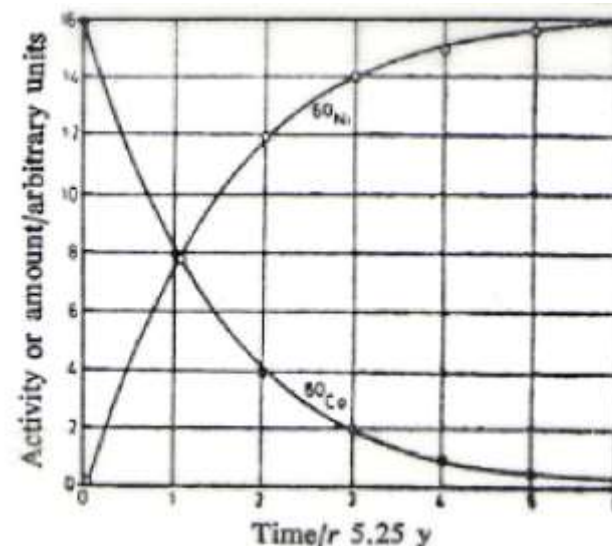
# Special Cases

- No equilibrium, product is stable ( $\lambda_2=0$ )

$$\frac{dN_2}{dt} = \lambda_1 N_1$$

$$dN_2 = \lambda_1 N_1 dt = \lambda_1 N_1^0 e^{-\lambda_1 t} dt$$

$$N_2 = \frac{\lambda_1 N_1^0}{-\lambda_1} (e^{-\lambda_1 t}) \Big|_0^t = N_1^0 (1 - e^{-\lambda_1 t})$$



# Special Cases

- Transient Equilibrium ( $\lambda_2 \sim 10 \times \lambda_1$ )

$$\lambda_2 > \lambda_1$$

$$e^{-\lambda_2 t} \ll e^{-\lambda_1 t}$$

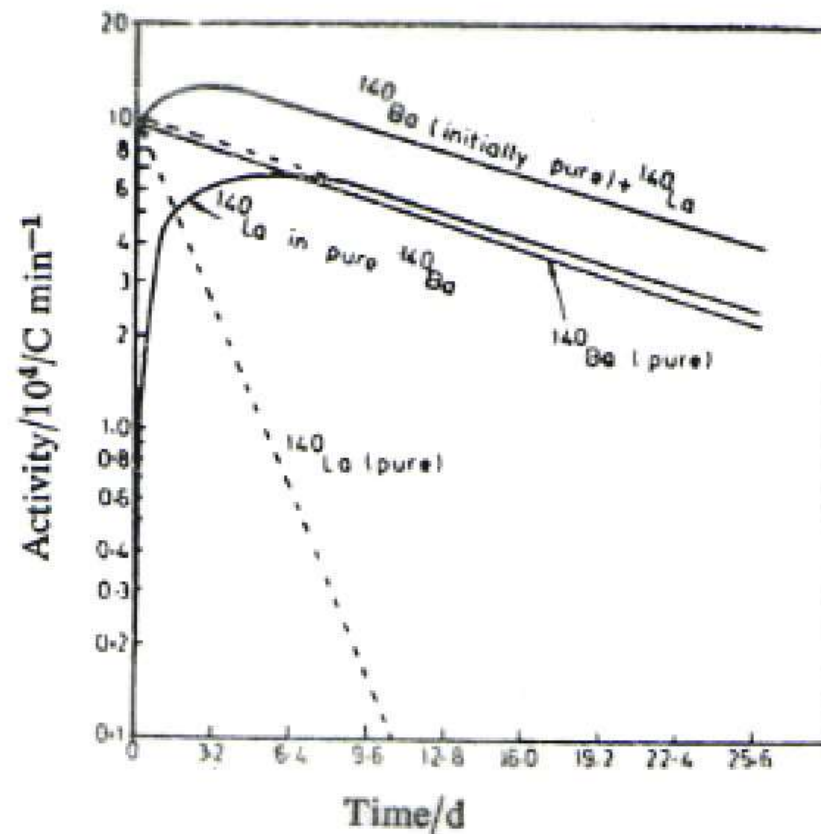
$$N_2 e^{-\lambda_2 t} \rightarrow 0$$

$$N_2(t) = \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t}$$

$$N_2(t) \approx \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} e^{-\lambda_1 t}$$

$$N_1 = N_1^0 e^{-\lambda_1 t}$$

$$\frac{N_1}{N_2} = \frac{\lambda_2 - \lambda_1}{\lambda_1}$$



Note that the daughter activity is maximum at  $t_{\text{max}}$

$$t_{\text{max}} = \frac{1}{\lambda_2 - \lambda_1} \ln \frac{\lambda_2}{\lambda_1}$$



# Special Cases

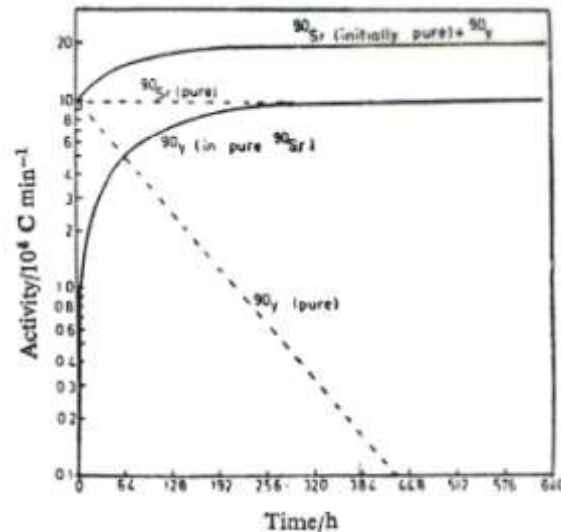
- Secular Equilibrium ( $\lambda_2 \gg \lambda_1$ )

$$\frac{N_1}{N_2} = \frac{(\lambda_2 - \lambda_1)}{\lambda_1}$$

$$\frac{N_1}{N_2} = \frac{(\lambda_2)}{\lambda_1}$$

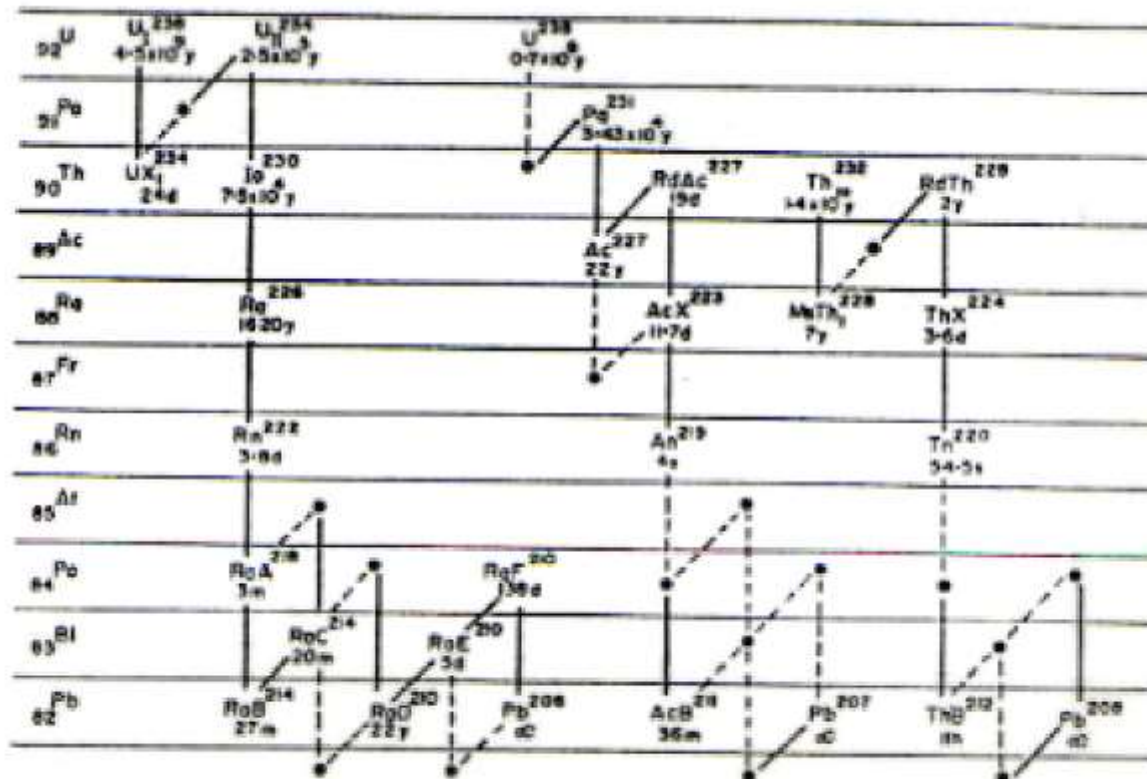
$$\lambda_1 N_1 = \lambda_2 N_2$$

$$A_1 = A_2$$



# Importance of Secular Equilibrium

- Naturally occurring decay series



# Importance of Secular Equilibrium

- Production of radionuclides in a nuclear reaction
- Nuclear reaction  $\rightarrow 2 \rightarrow$

$$N_2^0 = 0$$

$$Rate = R - \lambda_1 N_1$$

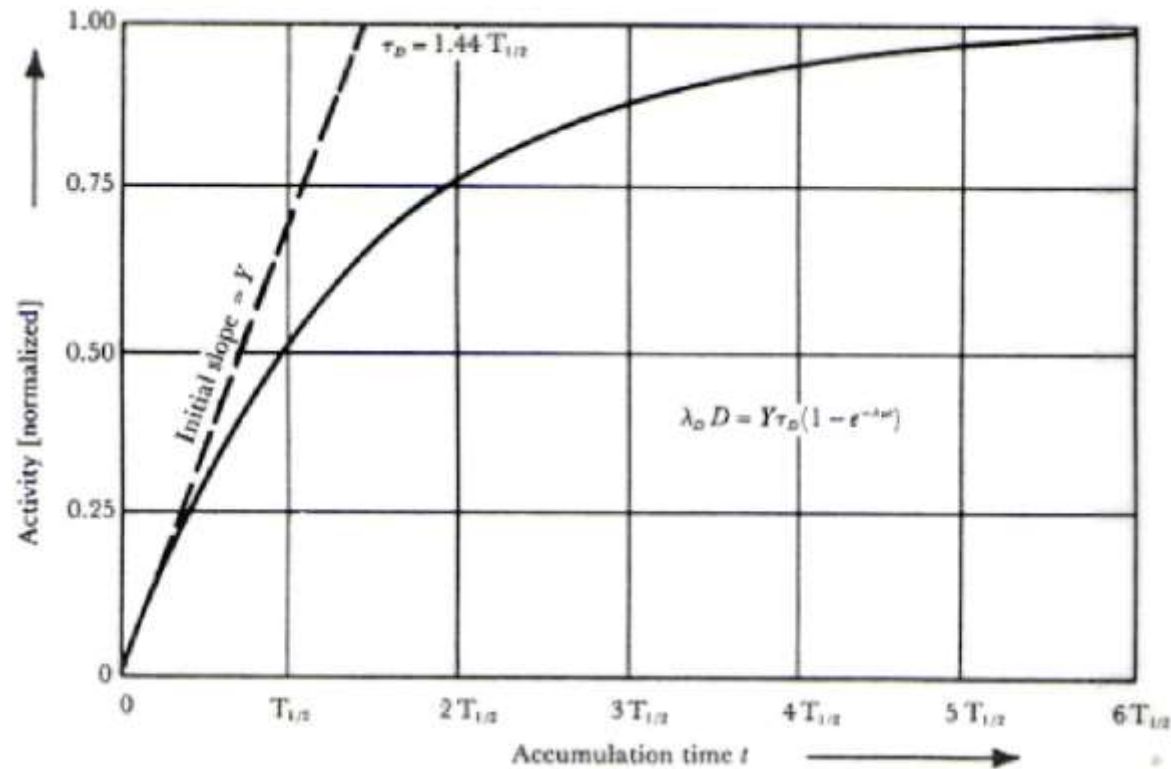
$$N_2(t) = \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t})$$

$$\lambda_1 \ll \lambda_2$$

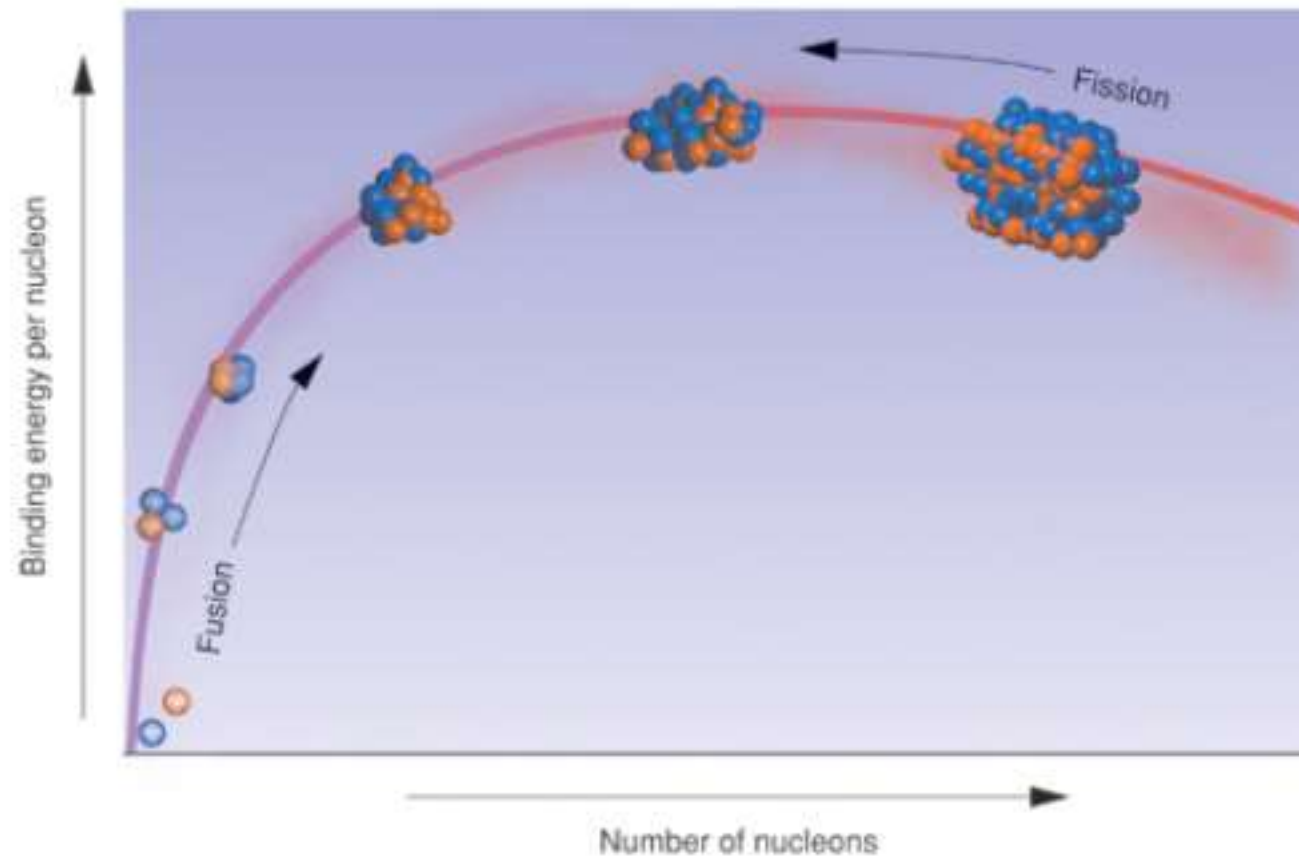
$$N_2(t) = \frac{\lambda_1 N_1^0}{\lambda_2} (1 - e^{-\lambda_2 t})$$

$$A_2(t) = \lambda_2 N_2 = R(1 - e^{-\lambda_2 t})$$

# The “Economics” of Irradiating Samples



At  $t=\infty$ ,  $A=A_{\text{saturation}}$



# Mass Parabolas and Valley of Beta Stability

$$M(Z, A) = Z \bullet M(^1H)c^2 + (A - Z)M(n)c^2 - B_{tot}(Z, A)$$

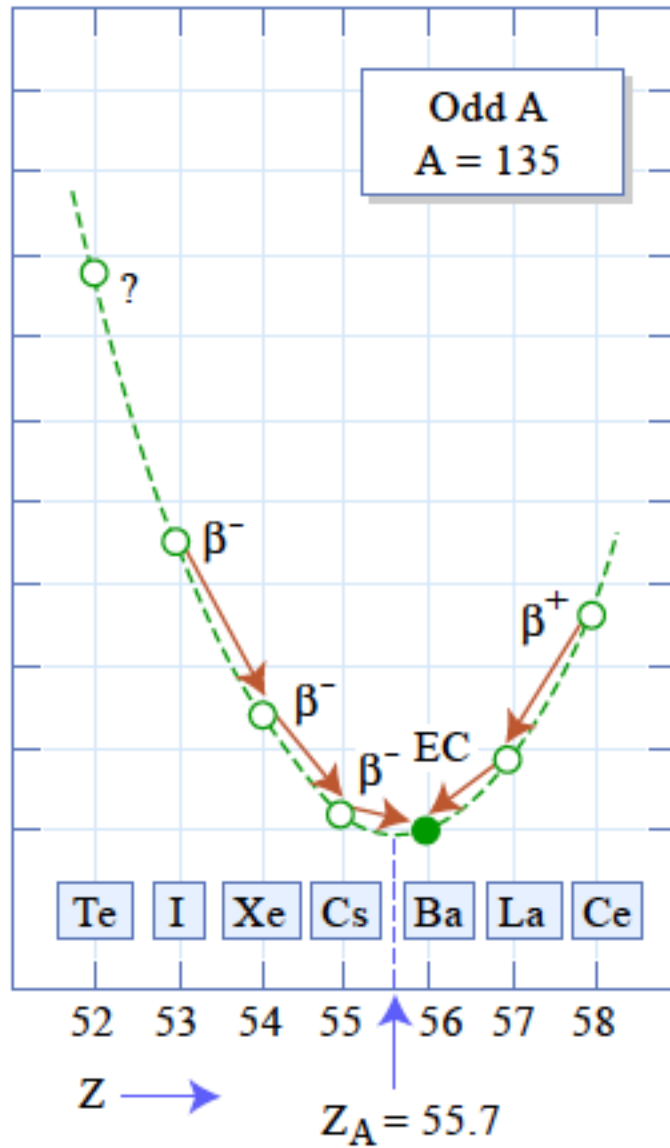
$$B_{tot}(Z, A) = a_v A - a_s A^{2/3} - a_c \frac{Z^2}{A^{1/3}} - a_a \frac{(A - 2Z)^2}{A}$$

$$a_a \frac{(A - 2Z)^2}{A} = a_a \frac{A^2 - 4AZ + 4Z^2}{A} = a_a \left( A - 4Z + \frac{4Z^2}{A} \right)$$

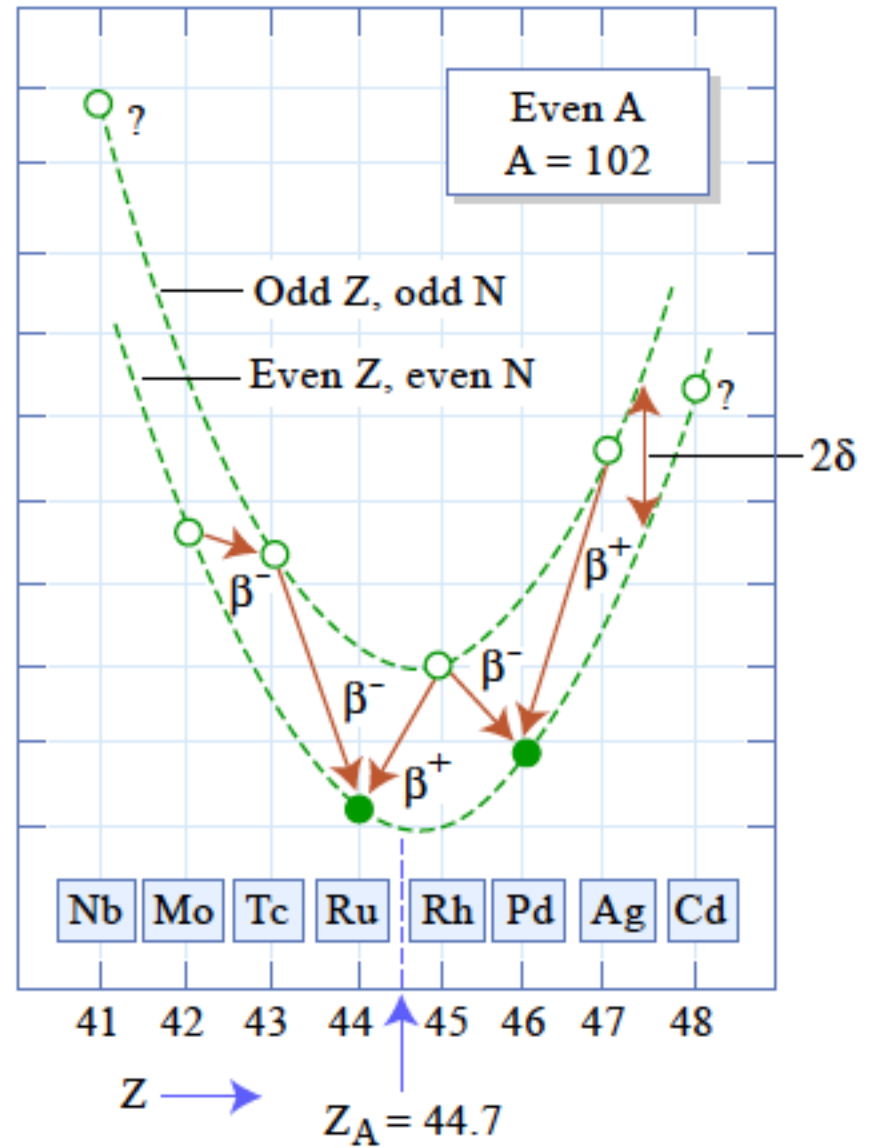
$$M = A \left[ M(n)c^2 - a_v + \frac{a_s}{A^{1/3}} + a_a \right] + Z \left[ M(^1H)c^2 - M(n)c^2 - 4a_a \right] + Z^2 \left( \frac{a_c}{A^{1/3}} + \frac{4a_a}{A} \right)$$

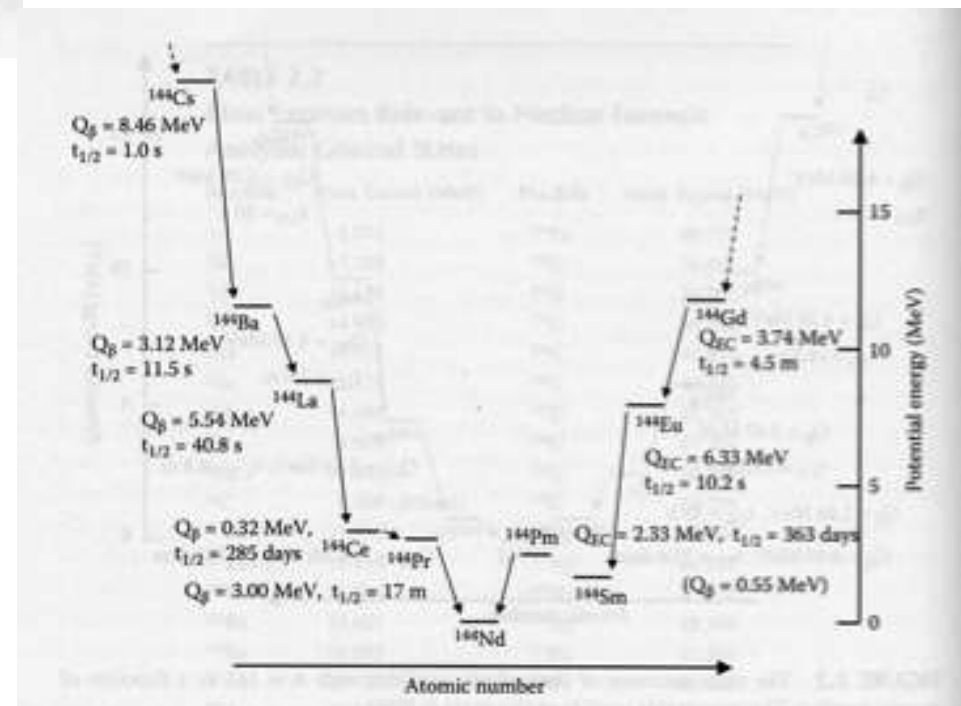
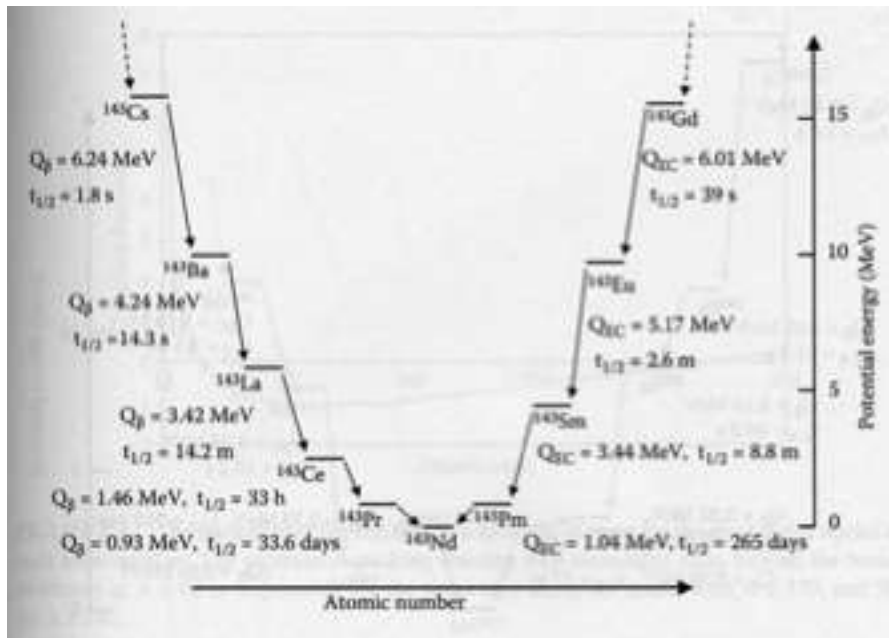
This is the equation of a parabola,  $a+bZ+cZ^2$

Atomic Mass (M)



Atomic Mass (M)







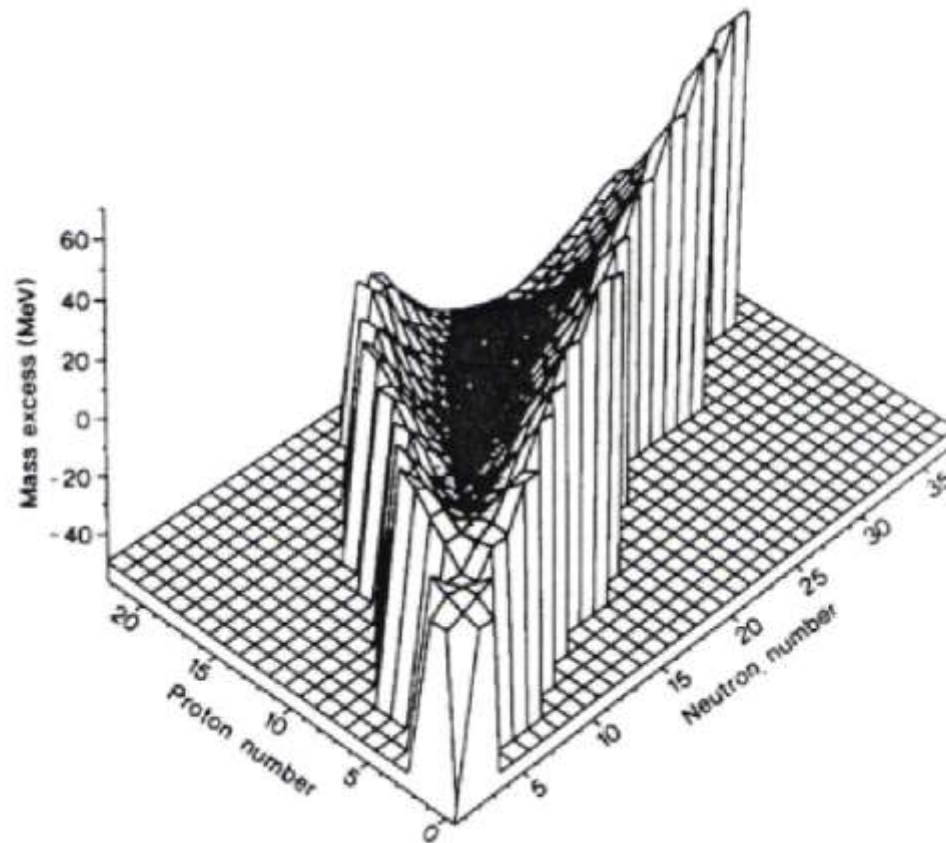
# Where is the Minimum of the Parabolas?

$$\left(\frac{\partial \mathcal{M}}{\partial Z}\right)_A = 0 = b + 2cZ_A$$

$$Z_A = \frac{-b}{2c} = \frac{M(^1H) - M(n) - 4a_a}{2\left(\frac{a_c}{A^{1/3}} + \frac{4a_a}{A}\right)}$$

$$\frac{Z_A}{A} \approx \frac{1}{2} \frac{81}{80 + 0.6A^{2/3}}$$

# Valley of Beta Stability



# The $\beta$ -Stable Valley

*Experimental Chart of Nuclides 2000*

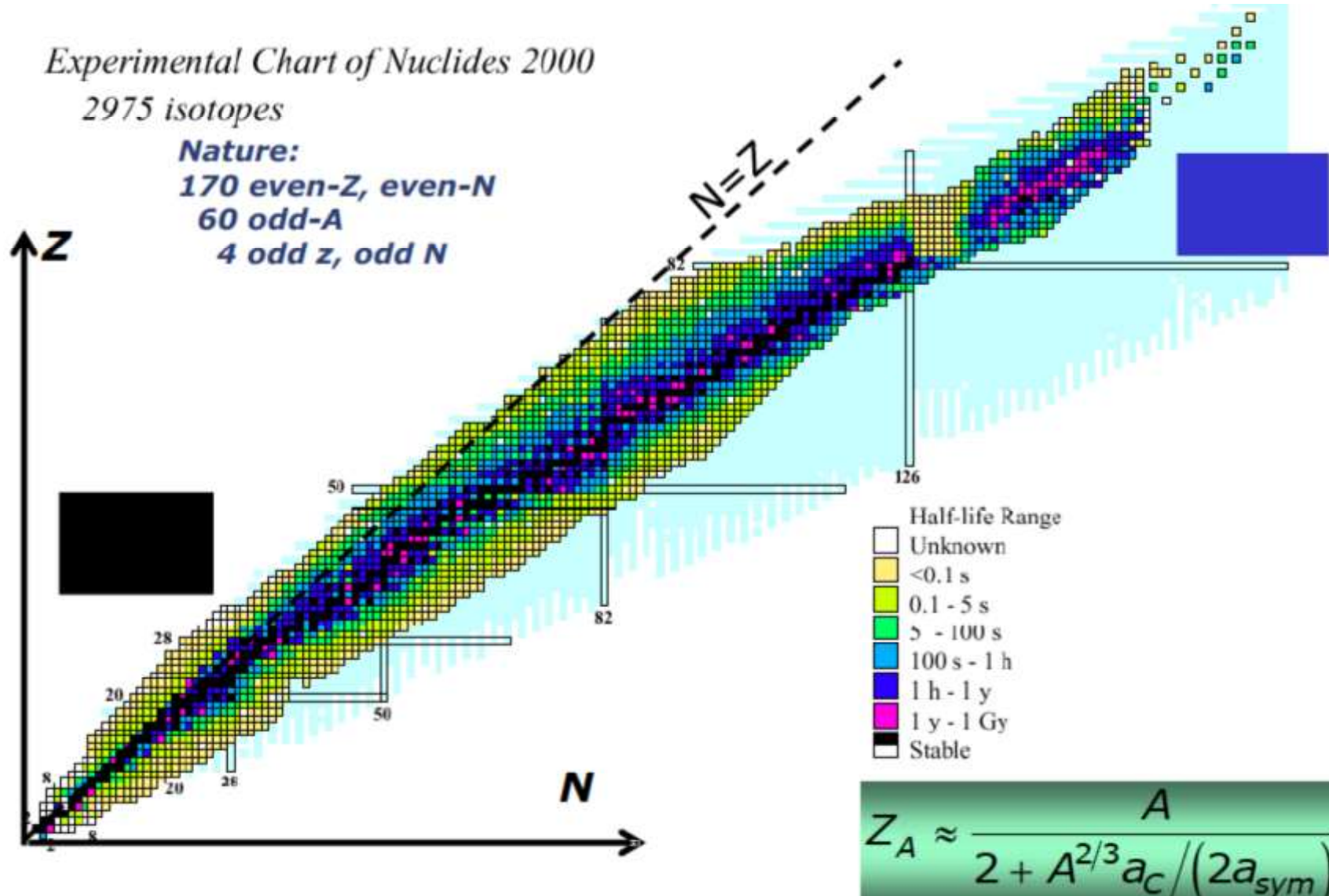
2975 isotopes

**Nature:**

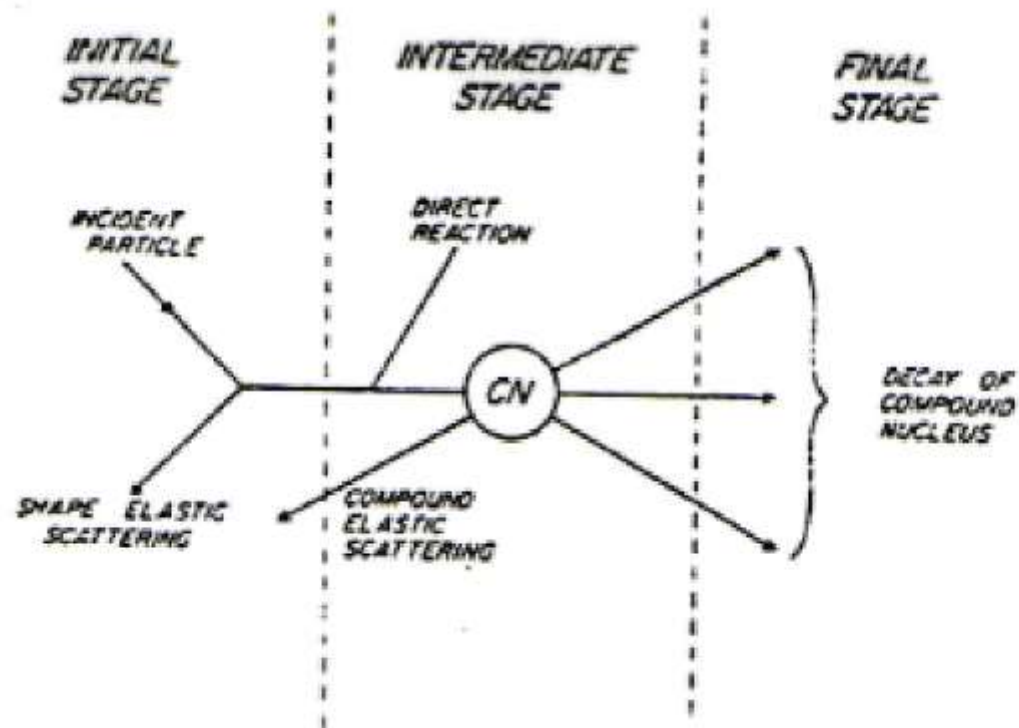
170 even-Z, even-N

60 odd-A

4 odd z, odd N



# Reaction Types and Mechanisms



# Reaction Types and Mechanisms

Incident particle interacts with	Type of force	Effect on energy of particle	Effect on nucleus			Name of process
			Kinetic energy	Internal energy	Identity	
Atomic electron	Coulomb	Slight reduction	Unchanged	Unchanged	Unchanged	Atomic ionization and excitation
Nucleus	Coulomb	Reduced	Increased	Unchanged	Unchanged	Coulomb or Rutherford scattering
Nucleus	Coulomb	Reduced	Increased	Increased	Unchanged	Coulomb excitation
Nucleus	Nuclear	Reduced	Increased	Unchanged	Unchanged	Nuclear elastic scattering
Nucleus	Nuclear	Reduced	Increased	Increased	Unchanged	Nuclear inelastic scattering
Nucleus	Nuclear	Particle transmuted or absorbed	Increased	Varies	Transmuted	Nuclear reaction

# Compound Nuclear Reactions

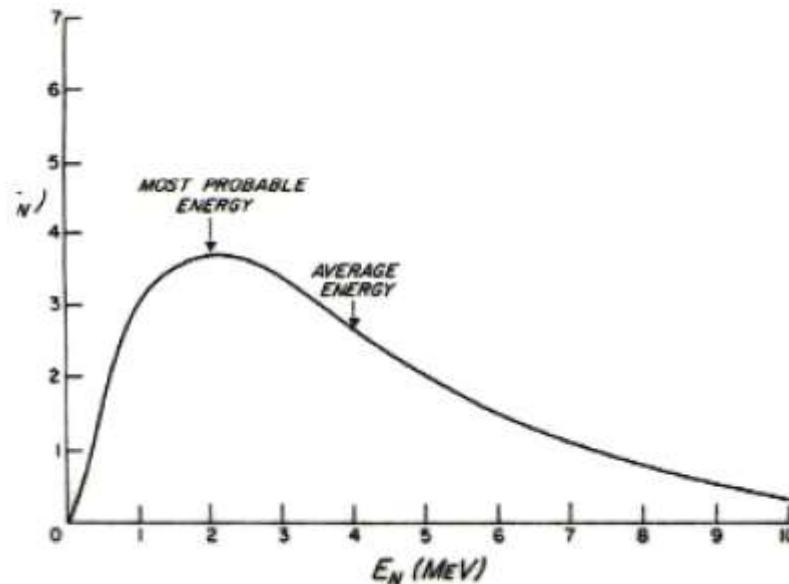
- The compound nucleus is a long-lived reaction intermediate that is formed by a complex set of interactions between the projectile and the target.
- The projectile and target nuclei fuse and the energy of the projectile is shared among all the nucleons of the composite system.
- The lifetime of the CN is  $\sim 10^{-18}$ - $10^{-16}$  s. This lifetime is directly measurable by crystal-blocking techniques.

# Independence Hypothesis or “Amnesia Assumption”

- The mode of decay of the CN is independent of its mode of formation.
- Caveat: Conservation laws apply.
- Experimental evidence: the Ghoshal expt.
- Angular distributions symmetric about  $90^\circ$  in CN frame.

# If emitted particles are neutrons

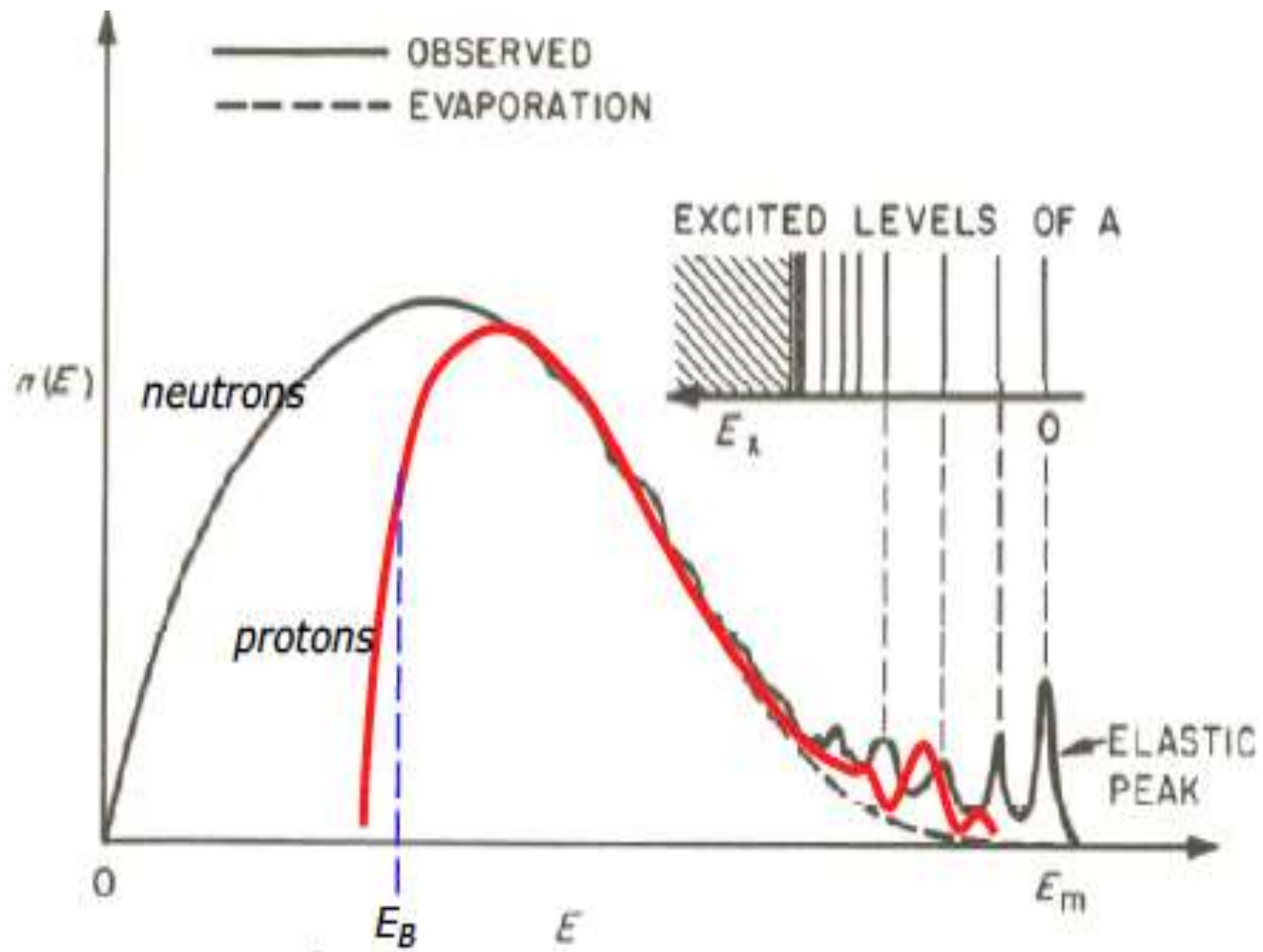
$$N(\varepsilon)d\varepsilon = \frac{\varepsilon}{T^2} \exp(-\varepsilon/T) d\varepsilon$$



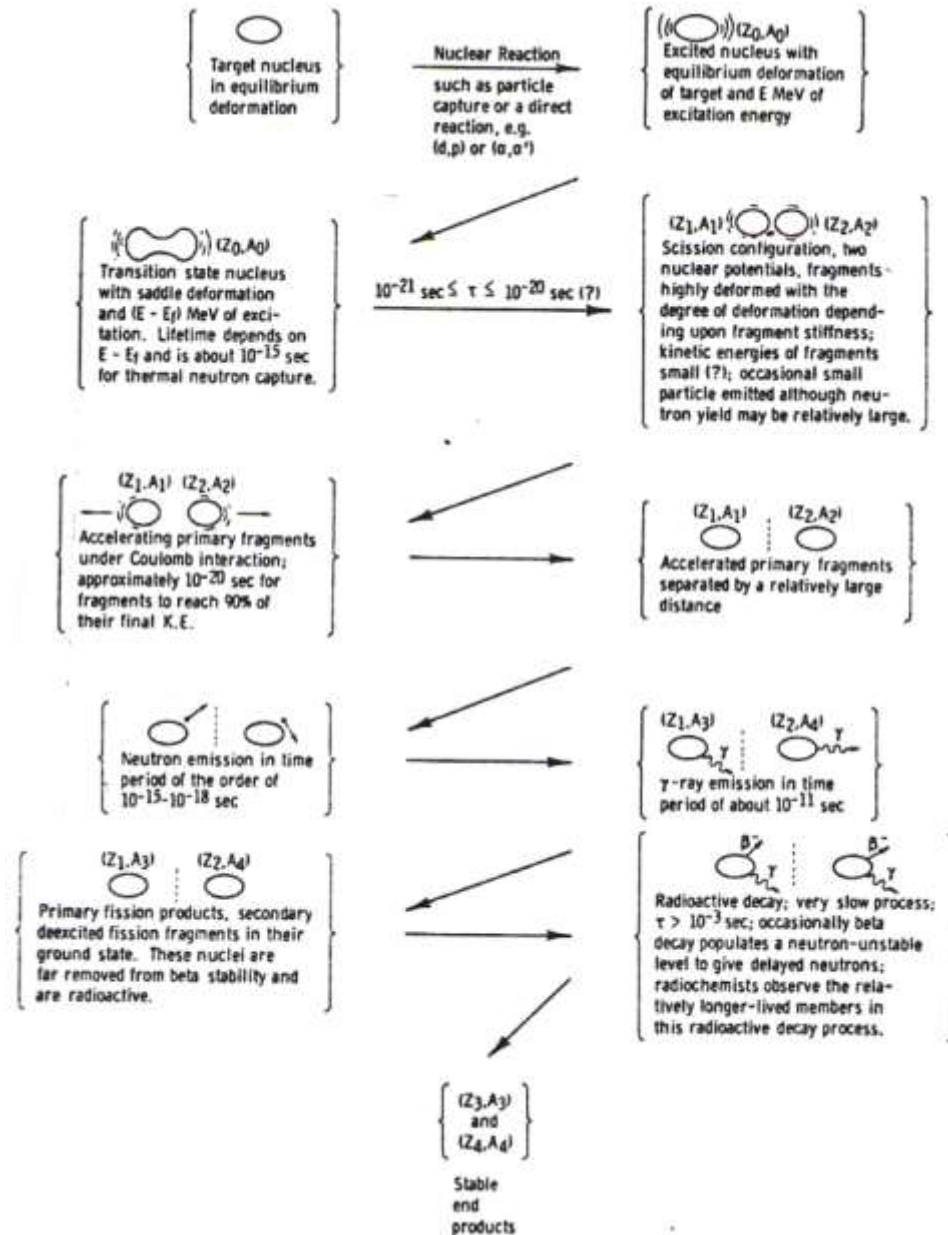


# If Emitted Particles Are Charged Particles

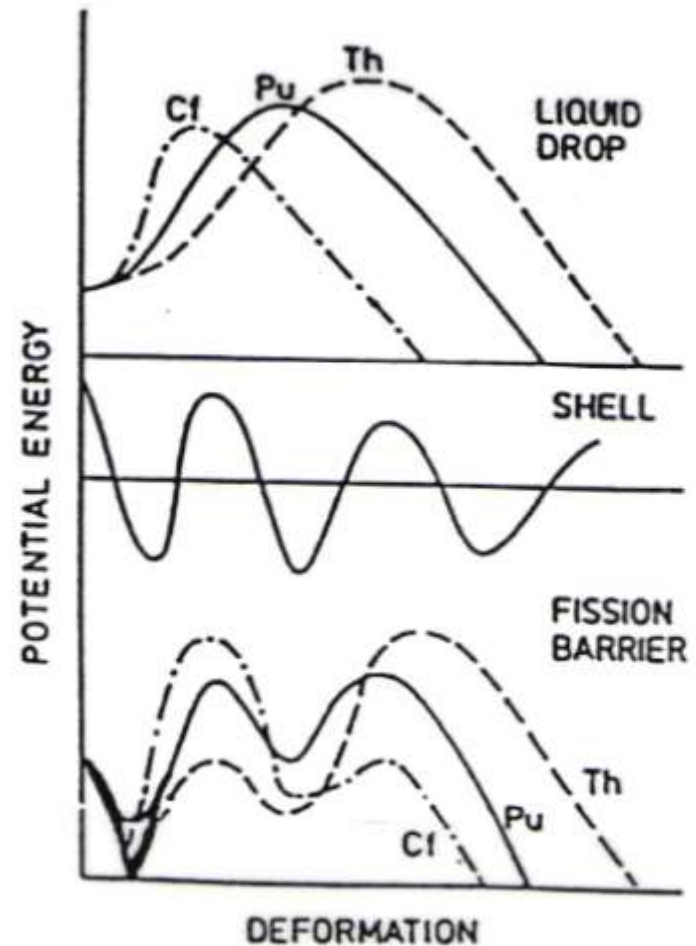
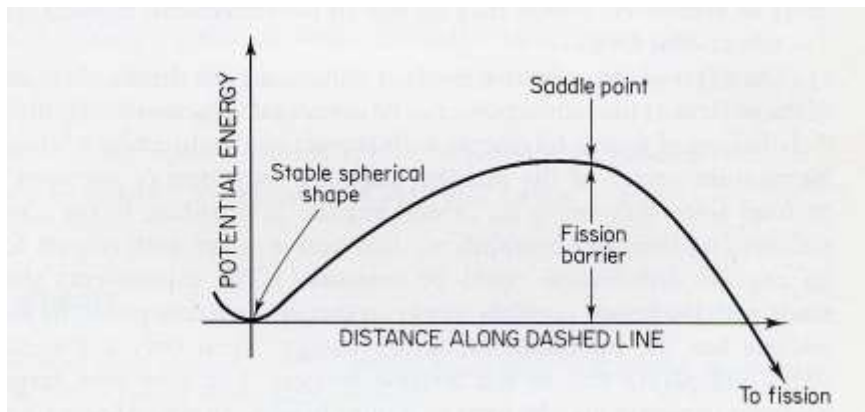
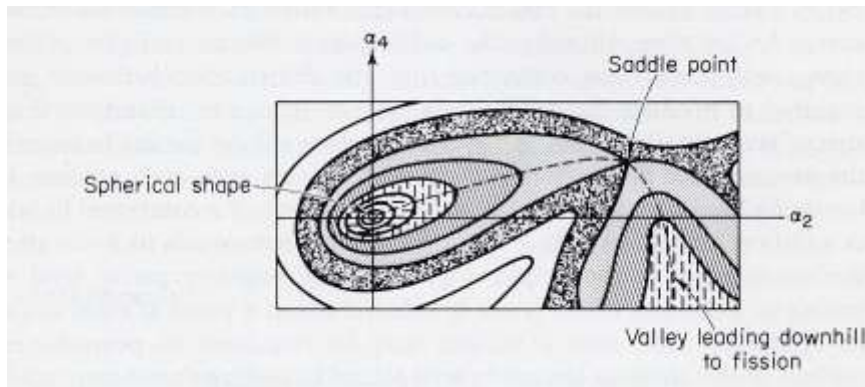
$$N(\varepsilon)d\varepsilon = \frac{\varepsilon - \varepsilon_s}{T^2} \exp(-(\varepsilon - \varepsilon_s)/T) d\varepsilon$$

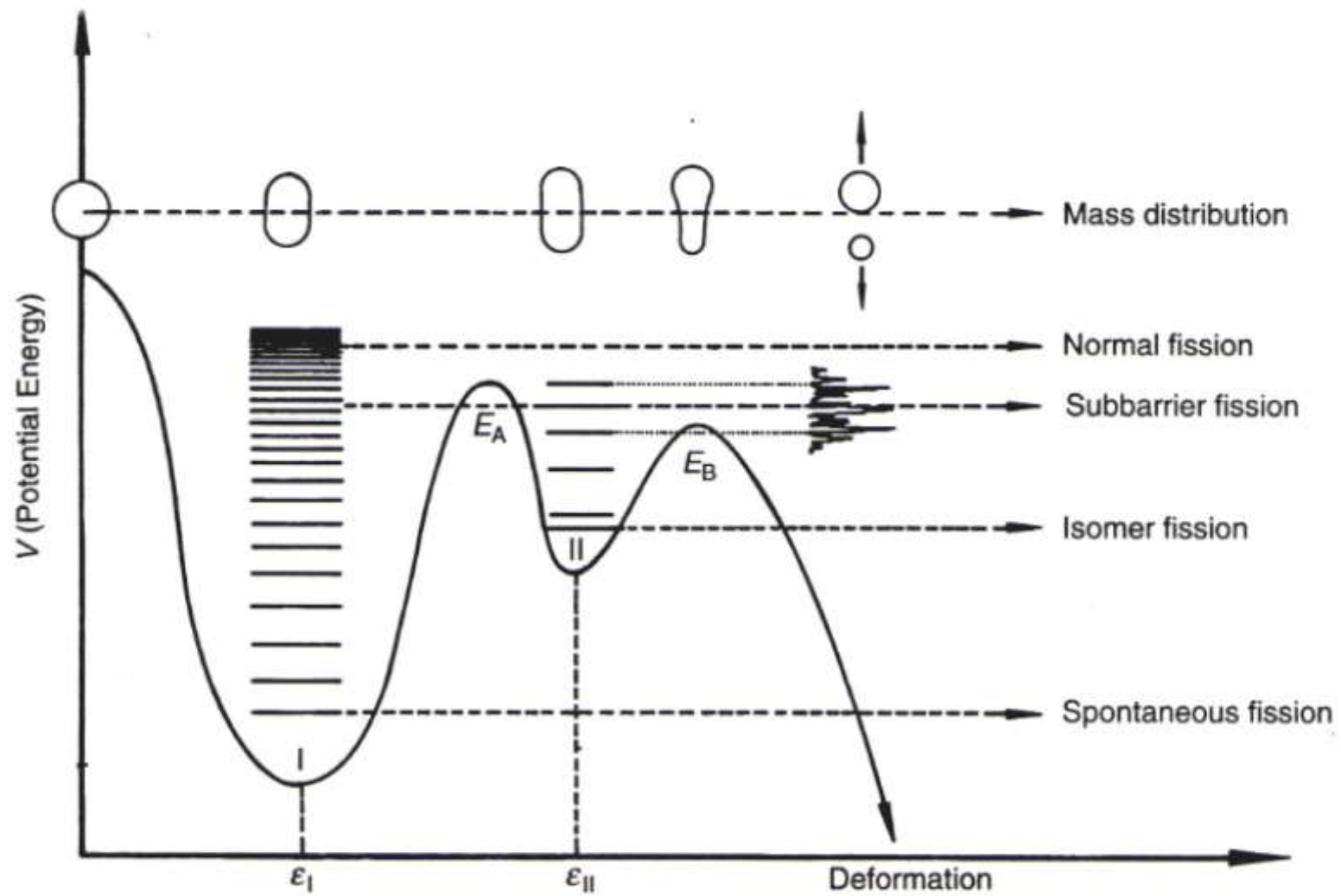


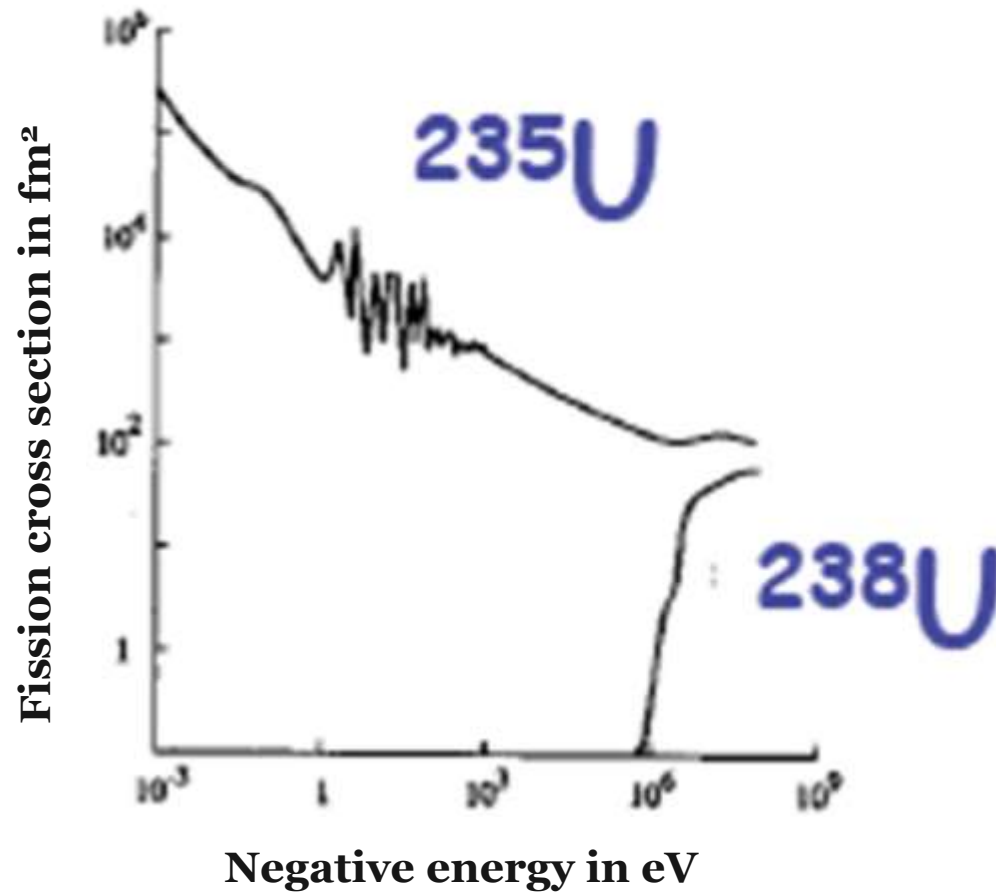
# Overview of fission



# Fission Barriers

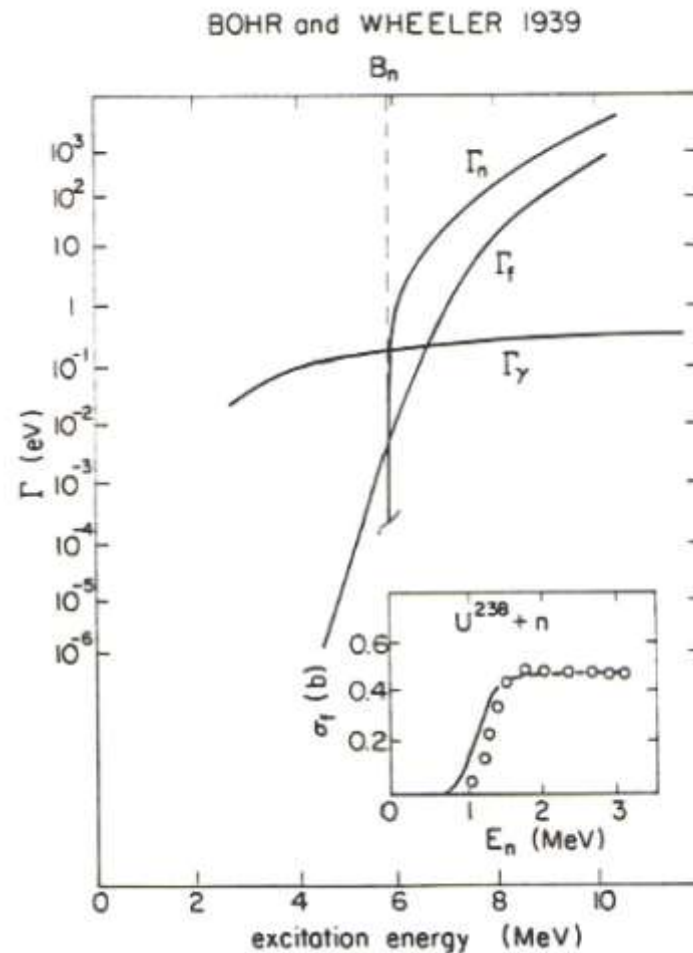




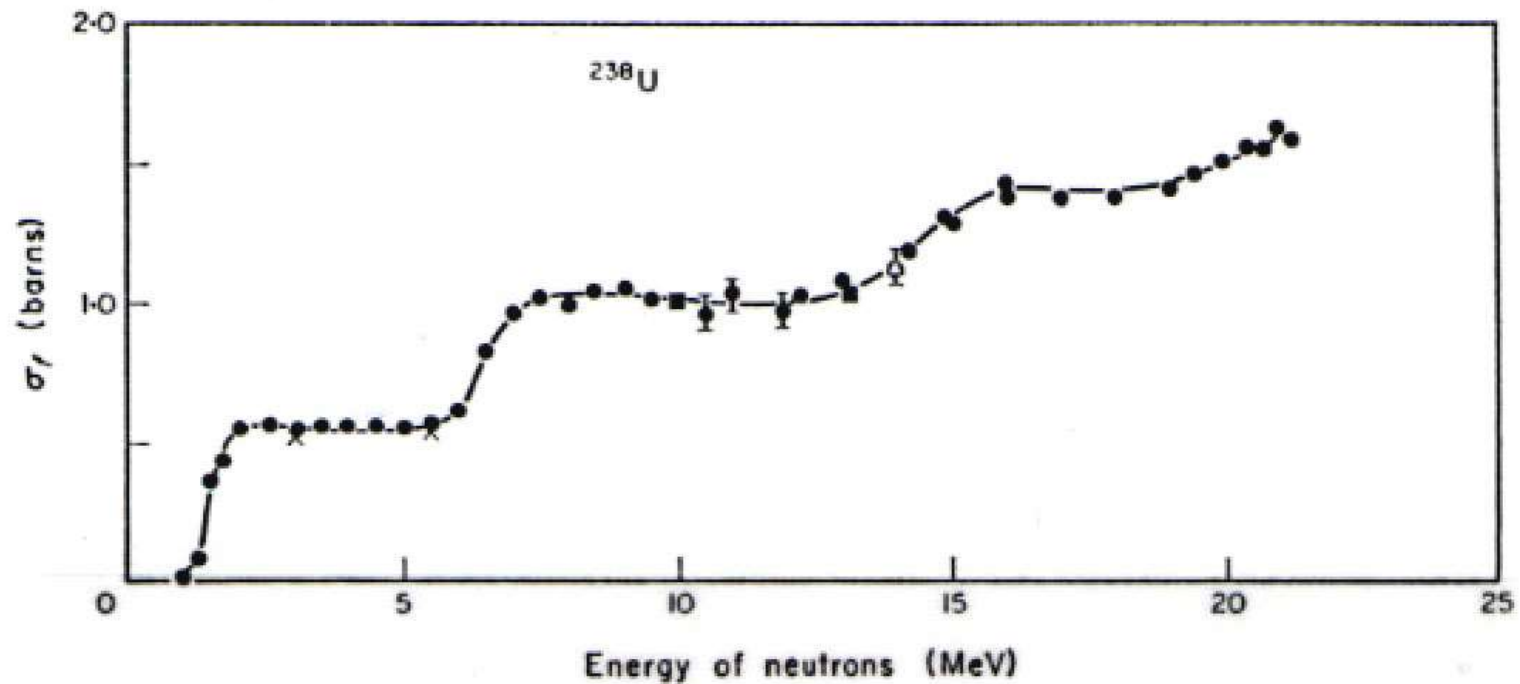


# Fission probability

- Fission probability ( $N_f/(N_f+N_n+N_{\text{gamma}}+N_{\text{ch.p.}})$ )



# Multiple chance fission





# Energetics of Fission

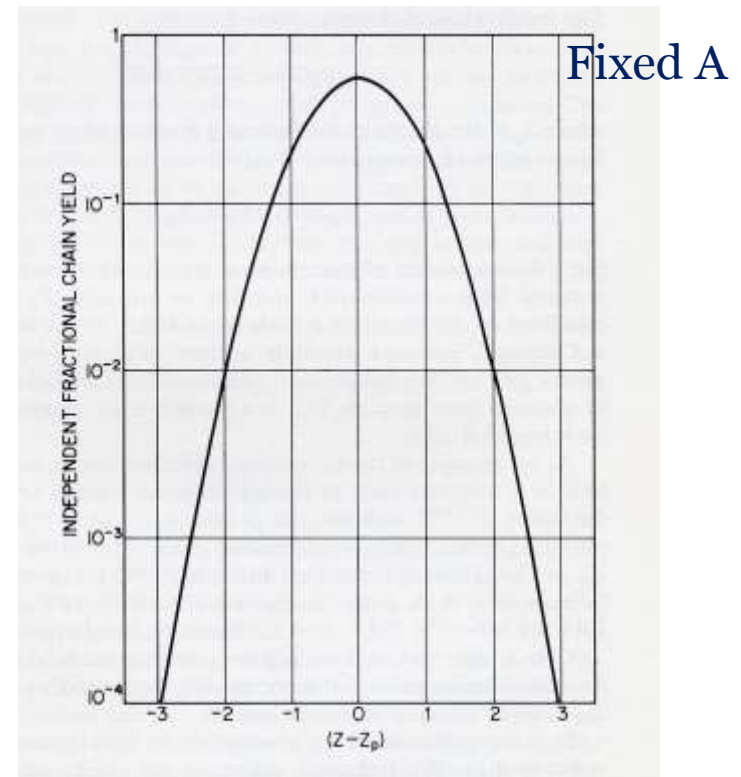
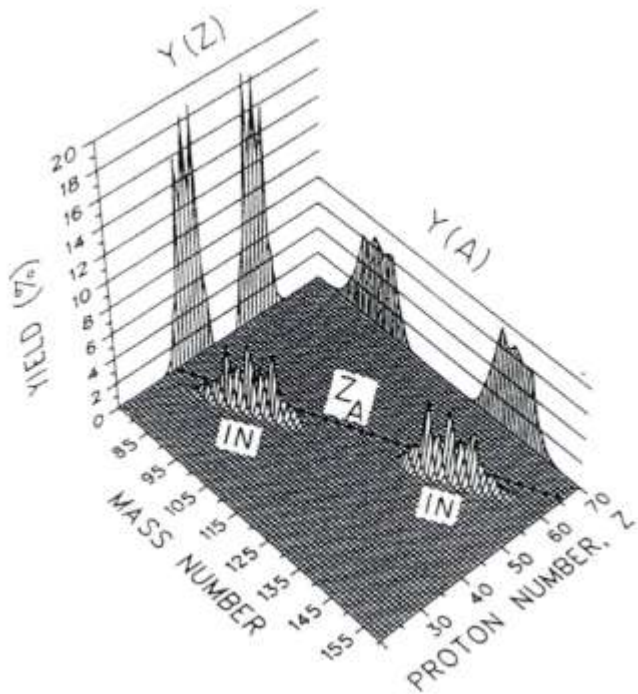
- Q value  $\sim 200$  MeV
- TKE  $\sim 172$  MeV     TXE = Q - TKE
- Neutrons  $\sim 18$  MeV
- Gammas  $\sim 7.5$  MeV
- $\beta$ , etc  $\sim 2.5$  MeV (“Decay heat”)

# Fission Product Distributions

## TKE Distribution

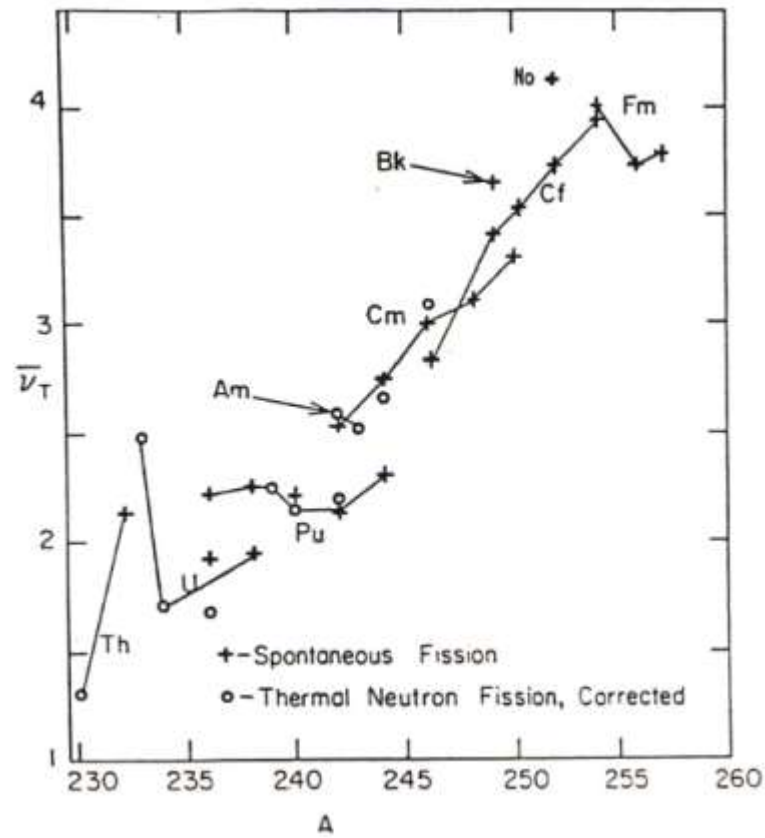
$$TKE = \frac{Z_1 Z_2 e^2}{1.8(A_1^{1/3} + A_2^{1/3})} \text{MeV}$$

# Fission Product Charge Distributions

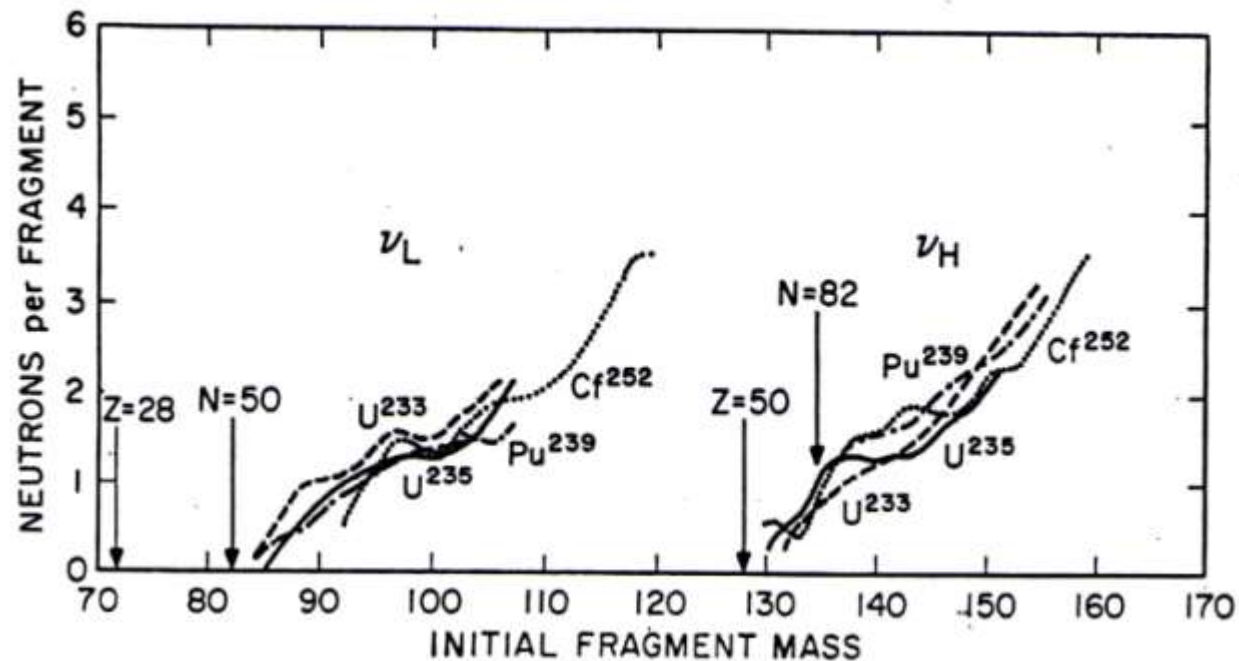


$$P(Z) = \frac{1}{\sqrt{c\rho}} \exp[-(Z - Z_p)^2 / c]$$

# Prompt Neutrons



# Prompt Neutrons



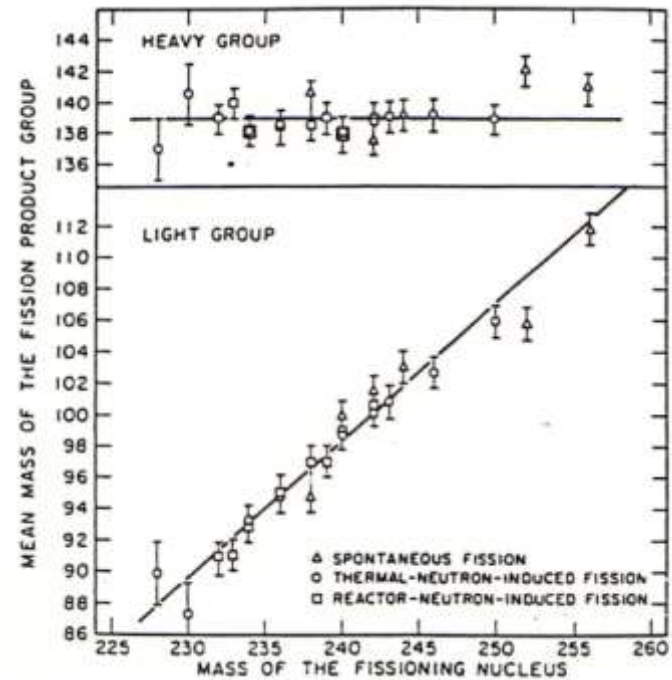
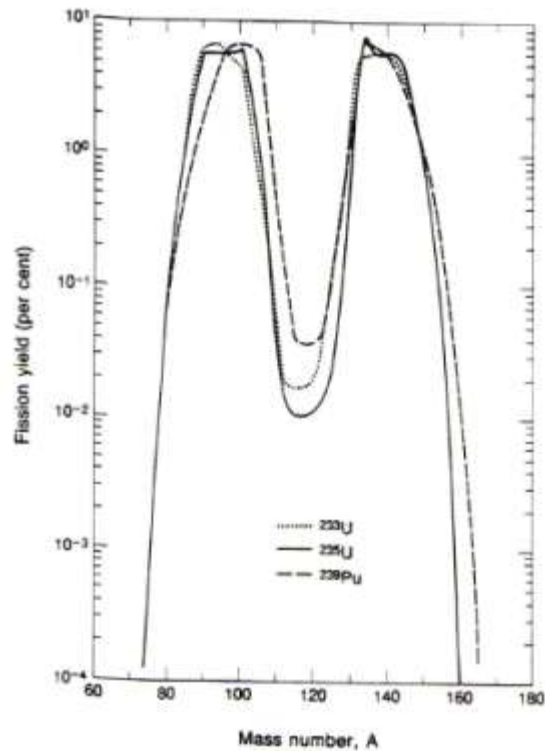
# Prompt Neutron Spectra

- Average neutron energy  $\sim 2$  MeV
- Spectrum:  
frame of moving fragment; Maxwellian  

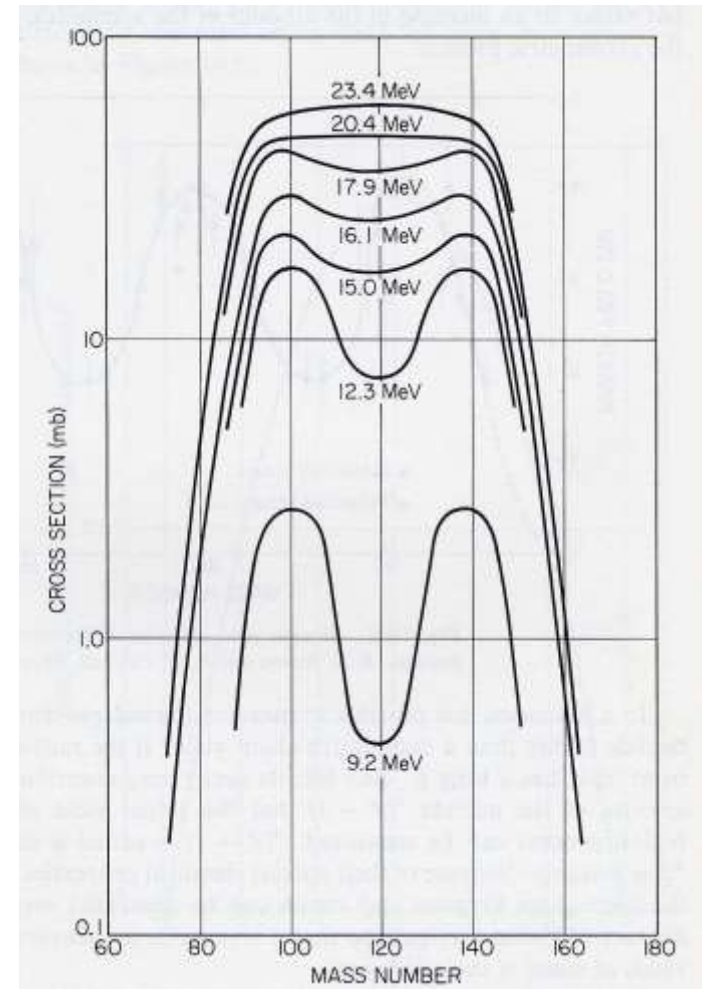
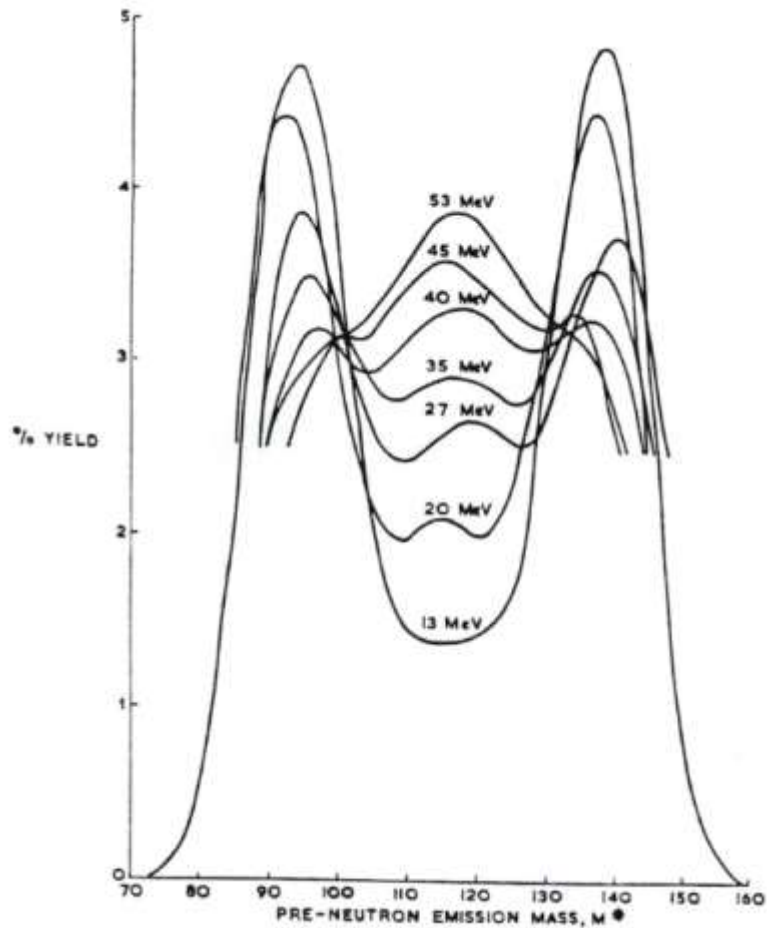
$$P(E) = E_n \exp(-E_n/T)$$
lab frame; Watt spectrum

$$P(E_n) = e^{-E_n/T} \sinh(4E_n E_f / T^2)^{1/2}$$

# Fission Mass Distributions

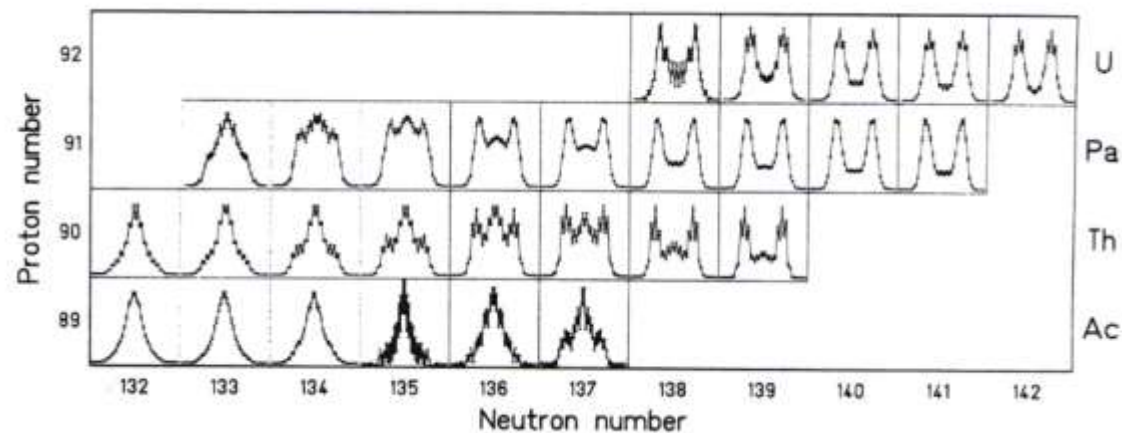
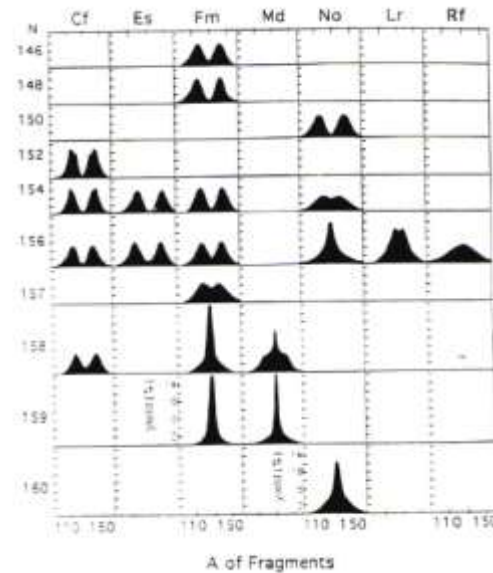


# Fission Mass Distributions

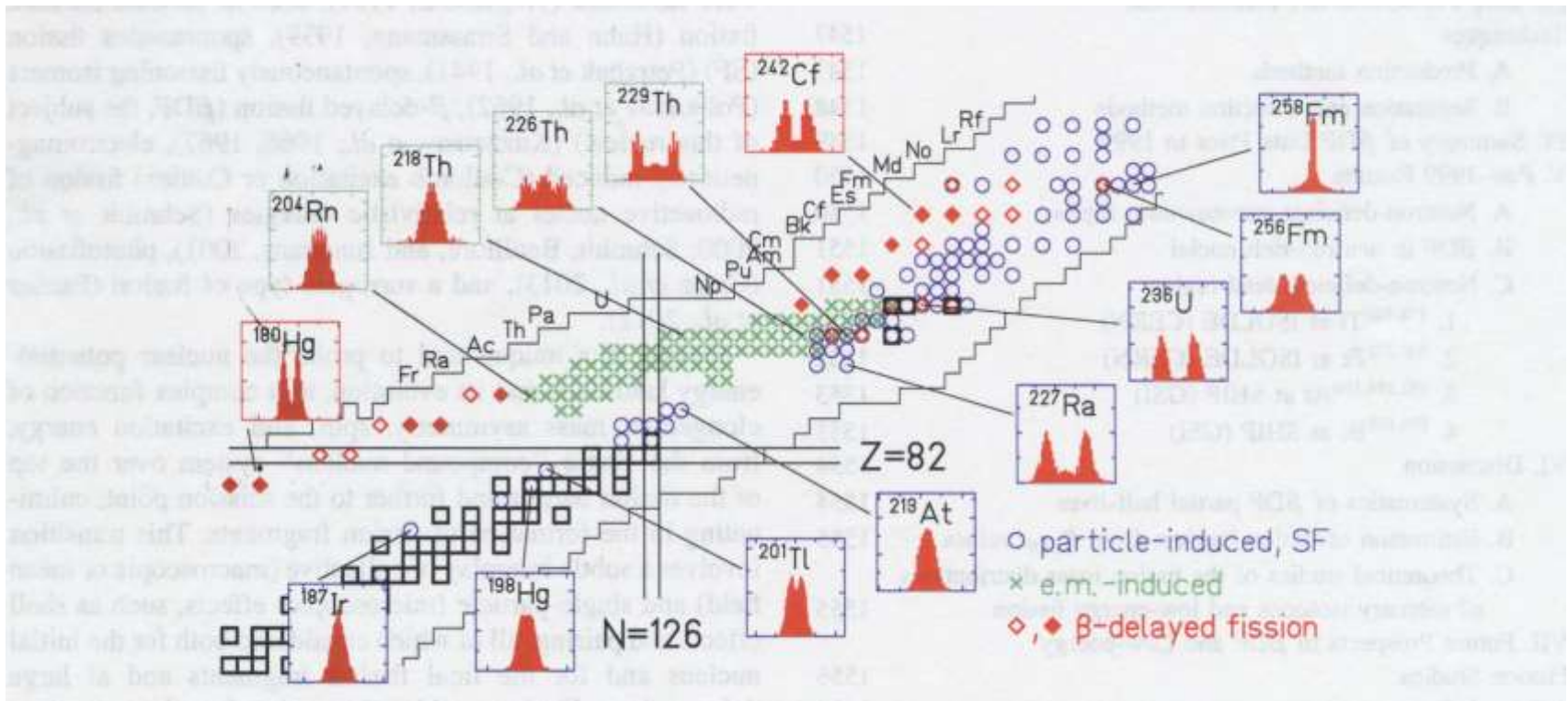




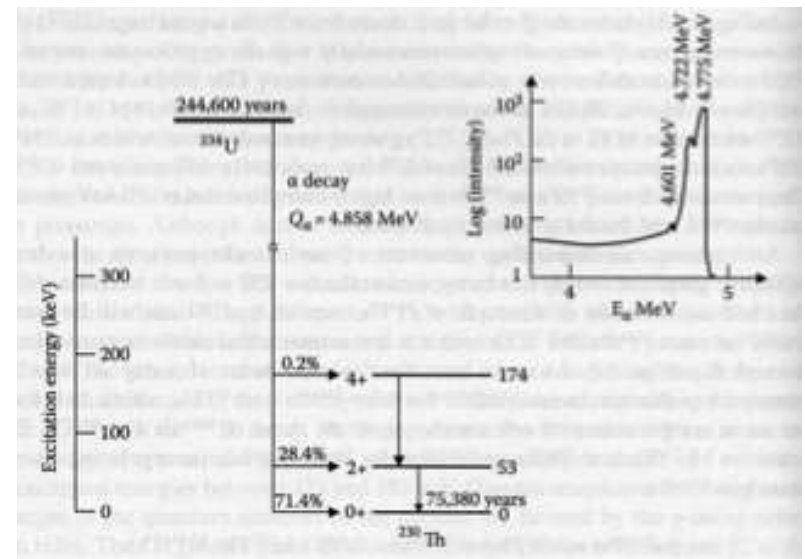
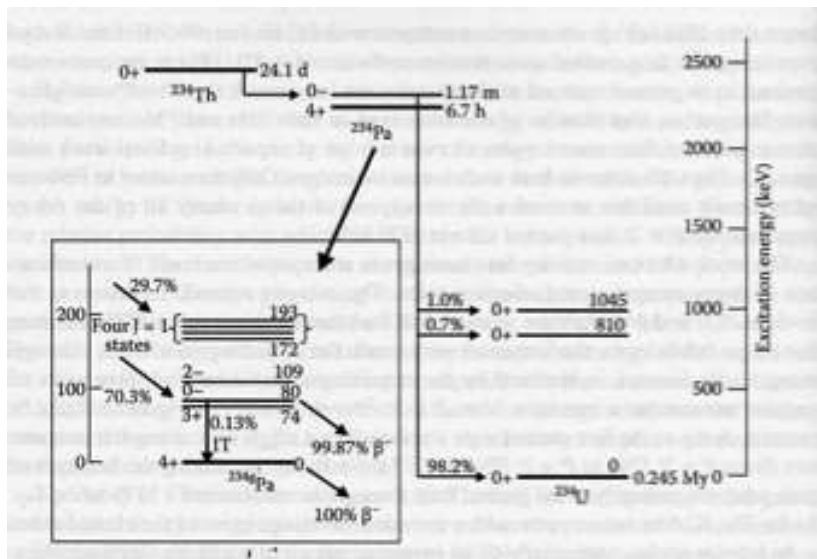
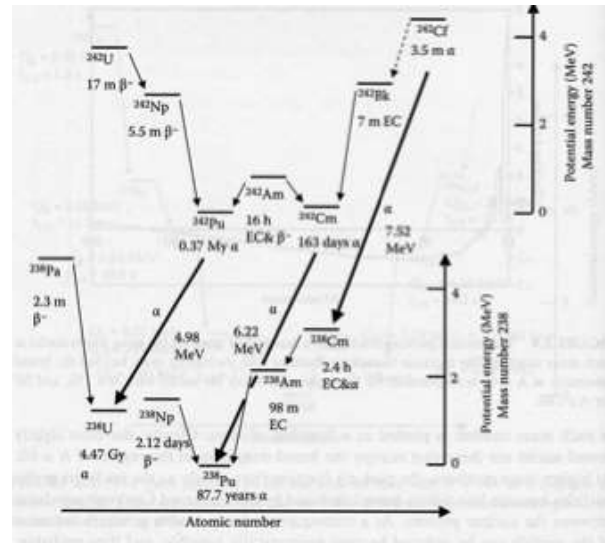
# Fission Mass Distributions



# Mass Distributions in Low Energy Fission



# Actinide decay schemes



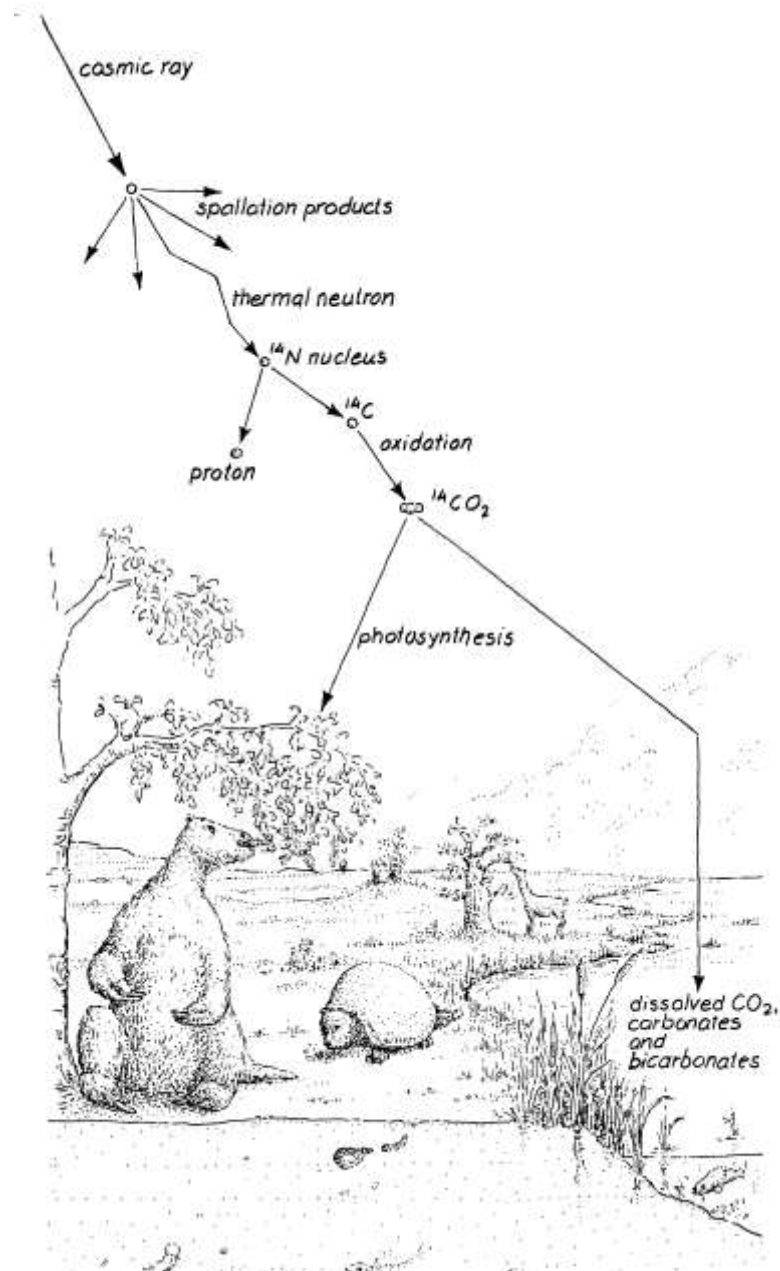
# Natural Radioactivity

- Primordial
- Cosmogenic
- Anthropogenic

Environmentally interesting radionuclides



# $^{14}\text{C}$



# Simple Radionuclide dating

$$A(t) = A_0 e^{-\lambda t}$$

$$t = \frac{\ln(A_0 / A)}{\lambda} = \frac{\ln(N_0 / N)}{\lambda}$$

## Tricks

- AMS
- Variations in  $A_0$  or  $N_0$

# Parent->Daughter Dating

$$D(t) + P(t) = P(t_0) = P_0$$

$$P(t) = P_0 e^{-\lambda t}$$

$$t = \frac{1}{\lambda} \ln \left( 1 + \frac{D(t)}{P(t)} \right)$$

# References

- K.J. Moody, P.M. Grant and I.D. Hutcheon, Nuclear Forensic Analysis, 2<sup>nd</sup> Edition (CRC Press, Boca Raton, 2015)
- K. Mayer et al., J. Phys. Conf. Series 312, 062003 (2011).



# Upcoming Webinars

- Nuclear Fission/Nuclear Devices
- Uranium Resources
- Chronometry

NAMP website: [www.wipp.energy.gov/namp](http://www.wipp.energy.gov/namp)