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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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
- The challenge of timeliness
A Glossary

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

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)


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 (α)**

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

$$^{14}C \rightarrow ^{14}N + \beta^- + \bar{\nu}_e$$
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,$^\nu$
- 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
  \[ ^{22}_{\text{Na}} \rightarrow ^{22}_{\text{Ne}} + \beta^+ + \nu_e \]
- 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
  \[ e^- + ^{209}\text{Bi} \rightarrow ^{209}\text{Pb} + \nu_e \]
- 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, γ-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 γ-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.)

• γ-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, i.e., the number of decays/s is proportional to the number of nuclei present.
- In equation form, \(-\frac{dN}{dt} = \lambda N\) where the constant \(\lambda\) is the decay constant.
- \((A = \lambda N)\)
- Rearranging:

\[
\frac{-dN}{dt} = \lambda N
\]

\[
\frac{dN}{N} = -\lambda dt
\]

\[
N = N_0 e^{-\lambda t}
\]

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

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

Graphically
Radioactive Decay Equilibria

Consider 1 → 2 → 3 →
rate of change of 2 = rate of production of 2 by decay of 1 - rate of decay of 2

\[
\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2
\]
\[
dN_2 + \lambda_2 N_2 dt = \lambda_1 N_1 dt
\]
\[
N_1 = N_1^0 e^{-\lambda_1 t}
\]
\[
dN_2 + \lambda_2 N_2 dt = \lambda_1 N_1^0 e^{-\lambda_1 t} dt
\]
\[
e^{\lambda_2 t} dN_2 + \lambda_2 N_2 e^{\lambda_2 t} dt = \lambda_1 N_1^0 e^{(\lambda_2 - \lambda_1) t} dt
\]
\[
d(N_2 e^{\lambda_2 t}) = \lambda_1 N_1^0 e^{(\lambda_2 - \lambda_1) t} dt
\]
\[
N_2 e^{\lambda_2 t} \bigg|_0^t = \frac{\lambda_1 N_1^0 e^{(\lambda_2 - \lambda_1) t}}{\lambda_2 - \lambda_1} \bigg|_0^t
\]
\[
N_2 e^{\lambda_2 t} - N_2^0 = \frac{\lambda_1 N_1^0 (e^{(\lambda_2 - \lambda_1) t} - 1)}{\lambda_2 - \lambda_1}
\]
\[
N_2(t) = \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t}
\]
\[
A_2(t) = \frac{\lambda_1 \lambda_2 N_1^0}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + A_2^0 e^{-\lambda_2 t}
\]
\[ N_2(t) = \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t} \]

\[ A_2(t) = \frac{\lambda_1 \lambda_2 N_1^0}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + A_2^0 e^{-\lambda_2 t} \]
Special Cases

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

\[
\frac{dN_2}{dt} = \lambda_1 N_1 \\
\int dN_2 = \lambda_1 N_1 dt = \lambda_1 N_1^0 e^{-\lambda_1 t} dt \\
N_2 = \frac{\lambda_1 N_1^0}{-\lambda_1} (e^{-\lambda_1 t}) \bigg|_0^t = N_1^0 (1 - e^{-\lambda_1 t})
\]
Special Cases

• Transient Equilibrium \((\lambda_2 \sim 10x \lambda_1)\)

\[
\lambda_2 > \lambda_1 \\
e^{-\lambda_2 t} \ll e^{-\lambda_1 t} \\
N_2 e^{-\lambda_2 t} \to 0 \\
N_2(t) = \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t} \\
N_2(t) \approx \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} e^{-\lambda_1 t} \\
N_1 = N_1^0 e^{-\lambda_1 t} \\
\frac{N_1}{N_2} = \frac{\lambda_2 - \lambda_1}{\lambda_1}
\]
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 ($\lambda_2 >> \lambda_1$)

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

\[
\frac{N_1}{N_2} = \frac{\lambda_2}{\lambda_1}
\]

\[
\lambda_1 N_1 = \lambda_2 N_2
\]

\[
A_1 = A_2
\]
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

\[ M(Z, A) = Z \cdot M(^1H)c^2 + (A - Z)M(n)c^2 - B_{\text{tot}}(Z, A) \]

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

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

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

This is the equation of a parabola, \( a + bZ + cZ^2 \)
Where is the Minimum of the Parabolas?

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

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

\[
\frac{Z_A}{A} \approx \frac{1}{2} \frac{81}{80 + 0.6A^{2/3}}
\]
Valley of Beta Stability
The $\beta$-Stable Valley

\[ Z_A \approx \frac{A}{2 + A^{2/3}a_c/(2a_{sym})} \]
Reaction Types and Mechanisms
### Reaction Types and Mechanisms

<table>
<thead>
<tr>
<th>Incident particle interacts with</th>
<th>Type of force</th>
<th>Effect on energy of particle</th>
<th>Effect on nucleus</th>
<th>Name of process</th>
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<td>Coulomb</td>
<td>Slight reduction</td>
<td>Unchanged</td>
<td>Atomic ionization and excitation</td>
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<td>Coulomb</td>
<td>Reduced</td>
<td>Increased</td>
<td>Coulomb or Rutherford scattering</td>
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<td>Nuclear</td>
<td></td>
<td>Varies</td>
<td>Nuclear reaction</td>
</tr>
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</table>
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° in CN frame.
If emitted particles are neutrons

\[ N(\varepsilon) d\varepsilon = \frac{\varepsilon}{T^2} \exp\left(-\frac{\varepsilon}{T}\right) d\varepsilon \]
If Emitted Particles Are Charged Particles

\[ N(\varepsilon)d\varepsilon = \frac{\varepsilon - \varepsilon_s}{T^2} \exp\left(-\frac{\varepsilon - \varepsilon_s}{T}\right) d\varepsilon \]
Overview of fission
Fission Barriers
Mass distribution
Normal fission
Subbarrier fission
Isomer fission
Spontaneous fission
Negative energy in eV

Fission cross section in fm²
Fission probability

- Fission probability \( \frac{N_f}{N_f + N_n + N_{\text{gamma}} + N_{\text{ch.p.}}} \)
Multiple chance fission
Energetics of Fission

- Q value ~ 200 MeV
- TKE ~172 MeV  \( TXE = Q - TKE \)
- Neutrons ~18 MeV
- Gammas ~ 7.5 MeV
- \( \beta, \text{etc} \) ~2.5 MeV ("Decay heat")
Fission Product Distributions

TKE Distribution

\[ TKE = \frac{Z_1 Z_2 e^2}{1.8 \left( A_1^{1/3} + A_2^{1/3} \right)} \ MeV \]
Fission Product Charge Distributions

\[ P(Z) = \frac{1}{\sqrt{c}} \exp\left[ \frac{(Z - Z_p)^2}{c} \right] \]
Prompt Neutrons
Prompt Neutrons
Prompt Neutron Spectra

- Average neutron energy ~ 2 MeV
- Spectrum:
  frame of moving fragment; Maxwellian
  \[ P(E) = E_n \exp\left(-\frac{E_n}{T}\right) \]
  lab frame; Watt spectrum

\[
P(E_n) = e^{-\frac{E_n}{T}} \sinh\left(\frac{4E_n E_{f}}{T^2}\right)^{1/2}
\]
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

\[ ^{222}\text{Rn} \]
\[ ^{40}\text{K} \]
\[ ^{3}\text{H} \]
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

Upcoming Webinars

• Nuclear Fission/Nuclear Devices
• Uranium Resources
• Chronometry

NAMP website: www.wipp.energy.gov/namp