Introduction to Radiochemistry

Lecture 9

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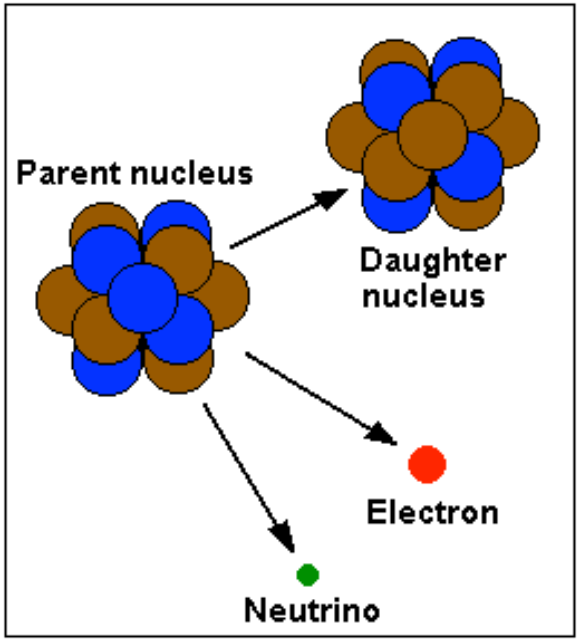
Kinetics of the Radioactive Decays

Introduction

- Single Radioactive Decay

- Decay Chains

- General Problem



Radioactive Decay of a Nuclide

-The radioactive decay is a **random process**

- The probability of decay in a certain time interval dt is history-independent.

- When observing a sample, it is not possible to predict which nucleus will decay but it is possible to predict global properties of the sample.

- The rate is the number of disintegrations in a given time and is commonly called **activity A:**

$$A = -\frac{dN}{dt}$$

- The "-" sign takes into account the decrease of radioactive nuclei.

- The unit of measure of A is the Becquerel (Bq). 1Bq = 1 disintegration/second.

Radioactive Decay of a Nuclide

- The activity is directly proportional to the number of nuclei we have:

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

Solving the above first order differential equation considering:

- A positive proportionality constant λ (the **decay constant**)
- The boundary condition $N(t = 0) = N_0$

we obtain:

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

The constant must have the dimension of [1/time] and therefore we can set $\lambda = 1/\tau$ with τ the **average lifetime**.

Half-Life

- It is useful to know after what time the number N_0 of radioactive nuclides will have been reduced by half.
  We can use the last equation and calculate:

$$\frac{N_0}{2} = N_0 e^{-\lambda t_{1/2}}$$

obtaining:

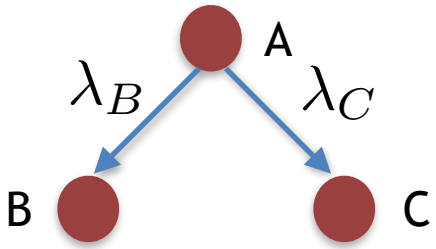
$$\ln \frac{1}{2} = -\lambda t_{1/2} \Rightarrow \boxed{t_{1/2} = \frac{\ln 2}{\lambda}}$$

or, introducing the average lifetime: $t_{1/2} = \tau \ln 2$

Branching Decays

- A nucleus can decay with two (or more) different modes with different probabilities (branching ratios).

- Let's consider the case of two possible decay modes of a nucleus A which can decay either in B or C:



- Branching decays are frequently found in odd-odd nuclei or heavy nuclei. It is common to have alpha-beta branchings.

Branching Decays

- The two branches are completely independent.

  In this case, the decay constants are called **partial decay constants**.

- From the independence, the total activity A_{tot} is given by the sum of the partial activities:

$$A_{tot} = \frac{dN_A}{dt} = \lambda_B N_A + \lambda_C N_A = (\lambda_B + \lambda_C) N_A = N_A \lambda_{tot}$$

- The total decay constant is the sum of the partial ones.

Decay Chains

- A parent radioactive nuclide can decay in another unstable daughter

- What can arise a are decay chains which stop only when a stable nuclide is reached.

- The situation is as follows:

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \longrightarrow \ldots$$

- In a very general situation we should take into account also branching decays.

Two-Step Radioactive Chain

- Let's start with the simplest case: a parent nuclide A with a single unstable daughter B and no branching decays:

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C$$

with C a stable nucleus. We would like to find an expression for the activity of C.

- The parent A will decay with the known formula:

$$\frac{dN_A}{dt} = -\lambda_A N_A \quad (1)$$

- The daughter B will be FORMED with a rate proportional to the amount of A:

$$\frac{dN_B}{dt} = \lambda_A N_A \quad (2)$$

(note the "+" sign now).

Two-Step Radioactive Chain

- While B is formed from the A decay, it is still radioactive and therefore decays with:

$$\frac{dN_B}{dt} = -\lambda_B N_B \quad (3)$$

- The total change in the activity of B is the sum of its formation and decay:

$$\frac{dN_B}{dt} = \lambda_A N_A - \lambda_B N_B \quad (4)$$

- From Eq. (1) we can derive: $N_A = N_A^0 e^{-\lambda_A t}$

- Substituting in Eq.(4) and rearranging the terms:

$$\frac{dN_B}{dt} + \lambda_B N_B - \lambda_A N_A^0 e^{-\lambda_A t} = 0$$

Two-Step Radioactive Chain

- The solution of the last equation is:

$$N_B(t) = \frac{\lambda_A}{\lambda_B - \lambda_A} N_A^0 (e^{-\lambda_A t} - e^{-\lambda_B t}) + N_B^0 e^{-\lambda_B t}$$

where we used also the boundary condition $N_B(0) = N_B^0$.

- The last equation has two relevant limiting cases:

1) **Transient Equilibrium**

2) **Secular Equilibrium**

Transient Equilibrium

- This situation arises when the lifetime of the parent is quite longer than the lifetime of the daughter but still not long enough so that we can see its decays during the experiment.

The corresponding exponentials $e^{-\lambda_B t}$ in the previous equations can be neglected after t~some B lifetimes with respect to $e^{-\lambda_A t}$.
The equation reduces to:

$$\frac{N_B}{N_A} = \frac{\lambda_A}{\lambda_B - \lambda_A}$$

- Given the proportionality between N and activity A, the same equation holds for the activities.

Secular Equilibrium

- If the lifetime of the parent is so long that we basically do not detect its activity during an experiment, we are in the situation of secular equilibrium.

- Again, we can neglect the $e^{-\lambda_B t}$ terms.

- Moreover, $\lambda_B - \lambda_A \approx \lambda_B$ and therefore the transient equilibrium equation derived before becomes:

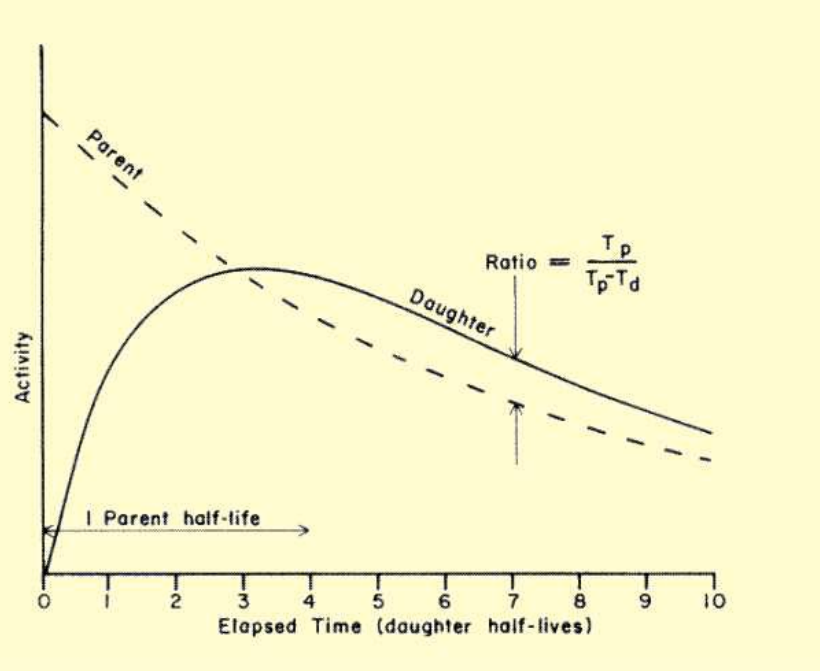
$$N_B \lambda_B = N_A \lambda_A$$

- Since the last equation is an equality between activities, we have also:

$$A_A = A_B$$

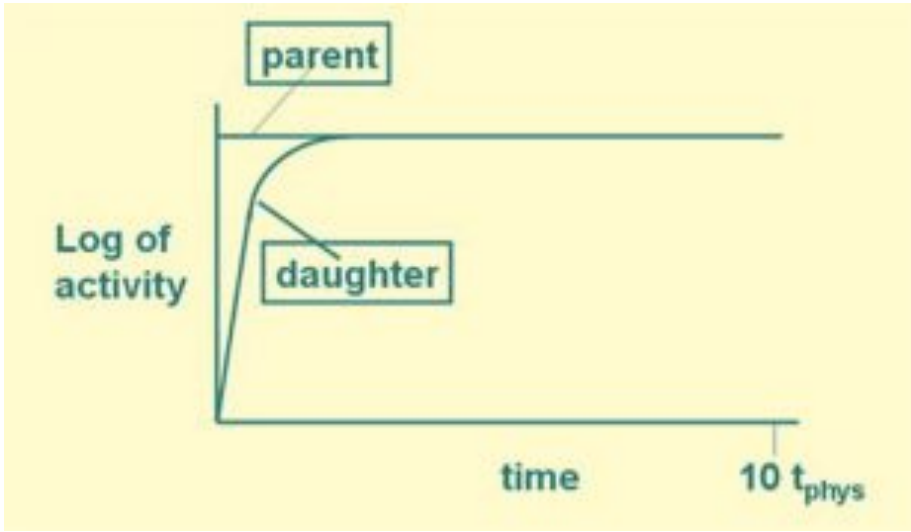
- Conclusion: after some time, the activities of the parent and the daughter become equal (secular equilibrium).

Transient/Secular Equilibria



Transient equilibrium: for long times, the activities decay exponentially with ratio of activities = A1/(A2-A1)

Secular equilibrium: for long times the activity of the daughter is equal to the activity of the parent.



Multiple Radioactive Daughters

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \longrightarrow \ldots$$

The case of multiple decays is more complicated but conceptually similar to the 2-decay case.
For example, the 3-decay case A—>B—>C gives for the activity of C:

$$\frac{dN_C}{dt} = \lambda_B N_B - \lambda_C N_C$$

and having already calculated the activity for B, we can substitute it obtaining:

$$\frac{dN_C}{dt} = -\lambda_C N_C + \lambda_B \left[\frac{\lambda_A}{\lambda_B - \lambda_A} N_A^0 (e^{-\lambda_A t} - e^{-\lambda_B t}) + N_B^0 e^{-\lambda_B t} \right]$$

The solution of the general case is given by the **Bateman equation** (see Notes).

Measurement of Decay Times

- The range of decay times is very large: from ~10-18 to ~1022 years.

- This means that different techniques have to be adopted depending on the decay time.

- We will omit specific details of the detectors since we will treat them in a specific lecture later.

- We divide (roughly) the decay times in 4 categories:

    1) Long decay times (several years)
    2) Medium (from seconds to few years)
    3) Short (seconds to milliseconds)
    4) Very short (below the millisecond range)

Long Decay Times

- In this case the activity stays about the same during the measurement period and therefore measuring the exponential decrease of A is not possible.

- Solution: measure the **specific activity** (SA) instead:

SA = disintegrations /sec /unit mass

The number of radioactive atoms in the sample is:

$$N = \frac{m(g)}{\text{atomic mass}} \cdot N_A$$

if we suppose an isotopically pure sample (otherwise we have to multiply also for the IS: the isotopic abundance). Recalling the definition of activity:

$$A = -\frac{dN}{dt} = \frac{\ln 2}{t_{1/2}} N \Rightarrow \boxed{t_{1/2} = \frac{\ln 2 \cdot N_A \cdot IS}{SA \cdot \text{atomic mass}}}$$

Need to measure SA and sample's mass.

Long Decay Times

Measurement of the **absolute activity** (true number of disintegrations per second):

1) Use of a **calibrated standard** for measuring detector's efficiency.
2) Measure the sample and divide the activity by the efficiency.

Warning: with this method, the thickness of sample and calibrated standard must be comparable for avoiding **self-absorption effects**.

A way to avoid the use of calibrated standards is to use ~100% efficient detectors with 4π coverage.

Long Decay Times

In some cases a **coincidence technique** might be used. For many nuclides, the beta decay is followed almost immediately by a gamma decay (—>why?). There is no angular correlation between the electron/positron and the gamma.

If we place the sample between a gamma and a beta detectors we will measure the following counting rates R:

$$R_\beta = \epsilon_\beta R_0 \qquad\qquad R_\gamma = \epsilon_\gamma R_0$$

where $\epsilon_{\beta/\gamma}$ are the efficiencies of the detectors and R_0 the true rate. Using an appropriate electronics, we can measure the rate at which the gamma and beta decays happen in coincidence. This rate is given by:

$$R_{\beta\gamma} = \epsilon_\beta \epsilon_\gamma R_0$$

The true rate R_0 is therefore given by:

$$R_0 = \frac{R_\beta R_\gamma}{R_{\beta\gamma}}$$

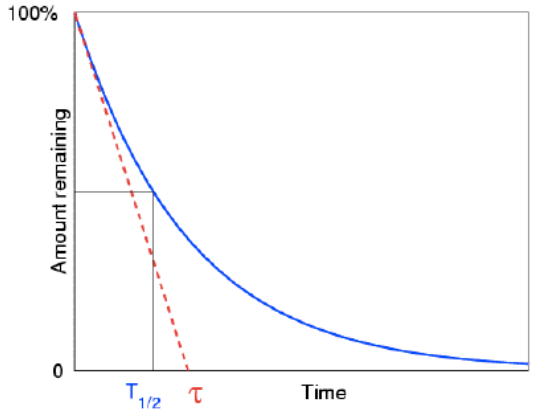
Medium Decay Times

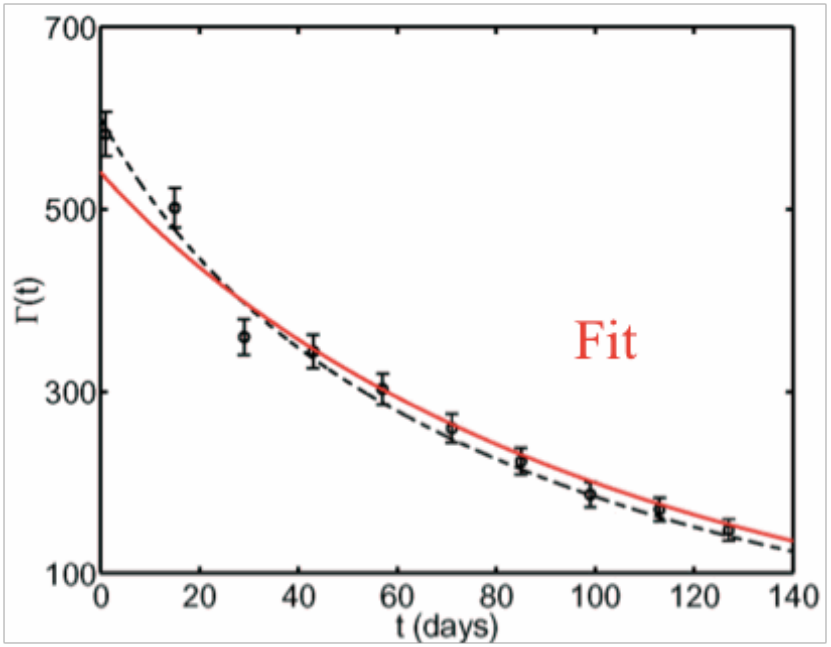
This is the case where we can record sufficient events and where the activity changes significantly during the experiment.

A direct fit to the activity as a function of time can be done. Actually it is easier to do a linear fit to the log(A) vs time data with the formula (just take the logarithm of the decay function):

$$\ln A = \ln A_0 - \lambda t$$

The method can be also sensitive to two decaying species in the sample if their decay times differ sufficiently.





Short Decay Times

Also in this case the direct fit method might work but the experimental complications are bigger.

If we induce the reaction in a target, if the decay time is too short the nuclide might decay within it.

There are methods devised for transporting the produced species outside the target and then count the decays.

One method is to rely on the recoil the produced nuclei have: the recoil can expel some of them from the target and eg. a stream of inert gas will transport them to a counting station.

Very Short Decay Times

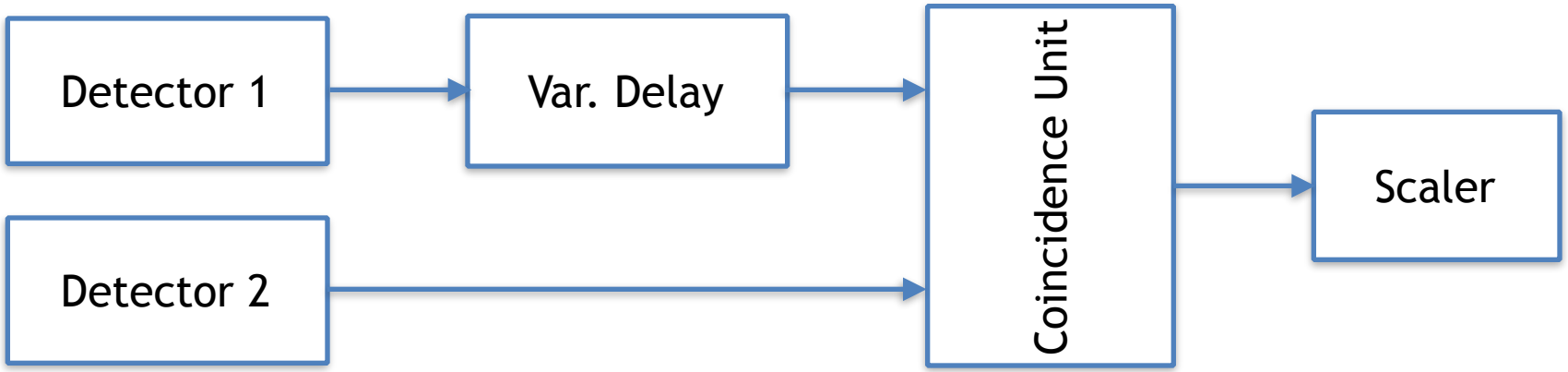
There are various methods and variants. Here we discuss:

1) Delayed coincidences Method

2) Doppler Shift Method

3) Energy Width Method

Delayed Coincidence Method

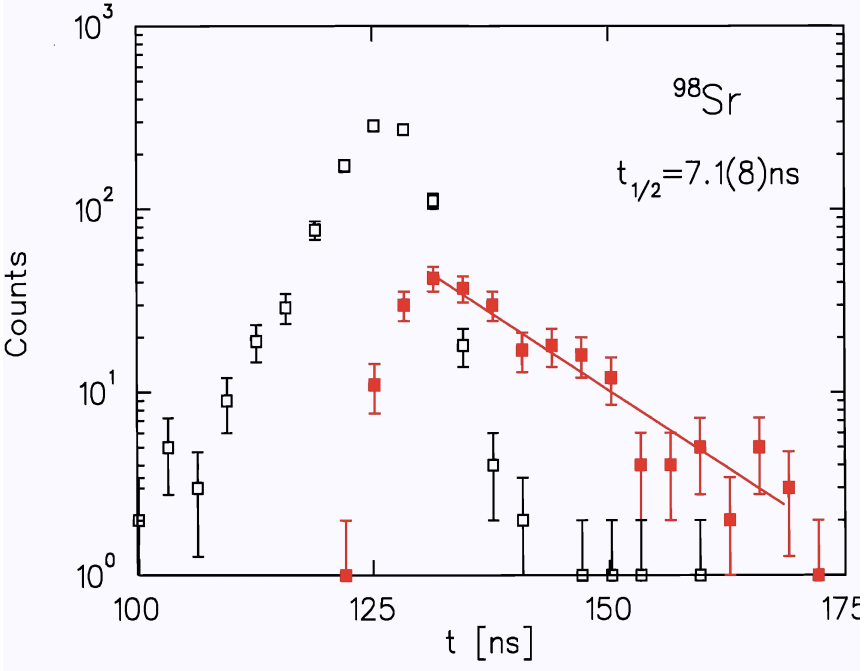


The idea is to detect the formation event (ex. a beta decay) and the decay event (ex. a gamma decay). Starting with delay time =0, the delay is increased until a maximum is reached. After the maximum, the coincidences will decrease. From the slope of the decrease, the lifetime can be derived.

Example:

$$^{24}\mathrm{Ne} \rightarrow ^{24m}\mathrm{Na} + \beta^-$$

$$^{24m}\mathrm{Na} \rightarrow ^{24}\mathrm{Na} + \gamma$$



Doppler Shift Method

The Doppler Shift effect (change in frequency due to the velocity of the emitter) can be used to measure very short lifetimes.

If the unstable nucleus is produced with a certain velocity at a known time.

One can detect gammas produced from the moving nuclei and from nuclei stopped in a material, therefore obtaining the Doppler shift in energy.

From the Doppler shift we can infer the velocity. The relationship between velocity and time in a medium is known experimentally therefore the time between formation and decay can be extracted.

The latter time is the average decay time from which the half-life can be calculated.

Energy Width Method

Recalling the Heisenberg uncertainty principle: $\Delta E \Delta t \geq \hbar$

we see that energy and time cannot be measured with arbitrary accuracy at the same time. The uncertainty on the energy E is called **decay width** Γ.

The decay width is related to the average lifetime of the state by:

$$\Gamma = \frac{\hbar}{\tau}$$

Therefore measuring the energy spread of a state we can calculate the average lifetime of it. The broader the energy peak, the shorter the lifetime.

Summary

1. Basic Decay Law

2. Branching Decays

3. Chain Decays:
   - Two-step decay
     - Transient Equilibrium
     - Secular Equilibrium
   - N-step decay

4. Experimental Measurement of lifetimes
   - Long Lifetimes
   - Medium Lifetimes
   - Short Lifetimes
   - Very Short Lifetimes