SECTION 15: Nuclear Reactions

Nuclear reactions provide the energy that fuels our universe. From the perspective of basic research, the focus is on understanding nuclear reaction mechanisms and structure, as well as the synthesis of exotic nuclei that do not exist on earth. More broadly, nuclear reactions find application in a spectrum of research interests. For example, nuclear astrophysics attempts to understand the reactions that are involved in stars and the evolution of our universe. As an analytical tool, neutron and proton activation analysis are applied to a wide range of problems, from environmental contaminants to forensic science. Nuclear power generates more than 20% of the electrical energy in the United States. Among the many other applications are the study of space-radiation effects, both biological and on semiconductor materials, relevant to satellites and space travel, and in medical science for producing radioisotopes for diagnostic procedures and for therapeutic purposes such as tumor treatment.

Basic Definitions

A nuclear reaction can be defined as a collision between two nuclei that produces as change in the nuclear composition and/or the energy state of the interacting species. As such, nuclear reactions fall in the category of a second-order kinetic process. Under normal circumstances, a reaction involves a **projectile**, a nucleus that is produced in a particle accelerator (Sec. 4) or nuclear reactor (Sec.18). Current technology can accelerate particles of all elements up to energies in excess of 10^6 MeV and velocities v ~ c. Table 15.1 summarizes several projectile sources in the U.S.

Table 15.1

Projectile type	**Facilities**
Neutrons	Nuclear reactors, see Appendix 15.A
Light ions ($A \leq 4$)	FNL Fermi National Laboratory Synchrotron (IL) AGS Alternating Gradient Synchrotron (NY))
Heavy ions (A = He to U)	NSCL National Superconducting Cyclotron Laboratory (MSU) Texas A&M Cyclotron Institute ATLAS Argonne National Laboratory (IL) RHIC Relativistic Heavy-ion Collider (NY) Lawrence Berkeley Laboratory 88-inch Cyclotron (CA) HHIRF Hollifield Heavy-ion Facility (TN)
Electrons & Photons	CEBAF Jefferson Laboratory (VA) SLAC Stanford Linear Accelerator (CA)
Exotic beams (π, K, antiprotons)	FNL and AGS

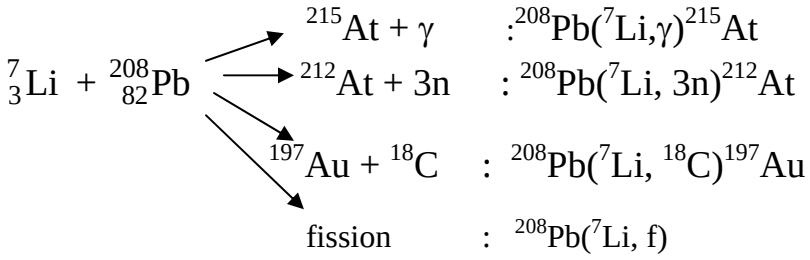
Radioactive beams: NSCL, ATLAS, HHIRF, Texas A&M Cyclotron Institute

The projectile is incident upon a **target** nucleus, which is usually fixed (v = 0), but in some accelerators colliding beams are possible, thereby increasing the center-of-mass energy of the collision. The **products** of a reaction may include any possible nuclei that are permitted by the conservation laws (mass-energy, baryons, charge, etc.). By accelerating the projectile, kinetic energy can be converted to mass in order to overcome negative Q-values.

The notation for a nuclear reactions is

Target(projectile, light products)heavy product(s)

As example, consider the following reaction and several possible products:



Often the heavy product can be omitted if there is no ambiguity.

For each of the four possible reactions above there is a separate Q-value, where

Q = ∑Δ(reactants) – ∑Δ(products) = available energy (Eq.15.1)

As an example, for the $^{208}Pb(^{7}Li, \gamma)$ reaction

$Q = \Delta(^{208}Pb) + \Delta(^{7}Li) - \Delta(^{215}At) - \Delta(\gamma,)$

$Q = -21.759 + 14.908 - (-1.263) - 0$

$Q =$ – 5.589 MeV ENDOTHERMIC

Since this reaction has a negative Q-value, it means that at least 5.589 MeV of energy must be converted into mass in order for the reaction to occur. However, if a ^{7}Li nucleus is accelerated to 5.589 MeV, no reaction is observed. Why?

Reaction Energetics

Two quantities are important energetic considerations in evaluating a nuclear reaction:

(1) The **threshold energy E_{th}** is the minimum projectile energy necessary to satisfy mass-energy and momentum conservation in a nuclear reaction to form products in their ground state, i.e. the energy necessary to compensate for a negative Q-value), and

(2) The **excitation energy E*** is the excess energy above the ground state for the product of a nuclear reaction.

In order to derive these quantities, mass-energy must first be conserved. For the generic reaction

projectile(p) + target(T) → composite nucleus (CN) + E*,

mass-energy conservation states

$$E_p + \Delta_p + \Delta_T = \Delta_{CN} + E_{CN} + E^*$$

$$E_p + \Delta_p + \Delta_T - \Delta_{CN} = E_p + Q = E_{CN} + E^* \qquad (\text{Eq. } 15.2)$$

where E_p denotes the kinetic energy of the projectile and E_{CN} that of the product. It is assumed that the target is at rest. Linear momentum conservation requires that

$$\vec{p}_p + \vec{p}_T = \vec{p}_{CN} \text{ , or non-relativistically}$$

$$\sqrt{2M_pE_p} + 0 = \sqrt{2M_{CN}E_{CN}}\,.$$

Approximating particle mass M by mass number A, this becomes

$$A_pE_p = A_{CN}E_{CN.} \qquad (\text{Eq. } 15.3)$$

Combining Eqs. 15.2 and 15.3 gives

$$E_p\left(\frac{A_{CN} - A_p}{A_{CN}}\right) = E^* - Q \text{ , or since } A_p + A_t = A_{CN},$$

$$E_p = (A_{CN}/A_T)(E^* - Q). \qquad (\text{Eq. } 15.4)$$

If **E* = 0**, the minimum energy or ground state, then $\mathbf{E_p = E_{th}}$ and eq. 15.4 becomes

$$E_{th} = (A_{CN}/A_T)(-Q). \qquad (\text{Eq. } 15.5)$$

For reactions in which the Q-value is negative, conservation of momentum requires that the composite nucleus have some kinetic energy and therefore the projectile energy must be greater than the negative Q-value.

If **E* > 0**, the excess energy above the ground state is given by

$$E^* = (A_T/A_{CN})\, E_p + Q\,. \qquad \text{(Eq. 15.6)}$$

The excitation energy is then dissipated by emission of photons or nuclear particles.

Problem: What is the threshold energy for the reaction of ^{7}Li with ^{208}Pb to form the composite nucleus ^{215}At? What is the kinetic energy of the ^{215}At nucleus?

$$E_{th} = -Q\left(\frac{A_{CN}}{A_T}\right) = -(-5.589\ \text{MeV})\,(215/208) = \underline{\mathbf{5.777\ MeV}}$$

$$E_{CN} = E_{th} - E_p = 5.777 - 5.589 = \underline{\mathbf{0.188\ MeV}}$$

In this reaction, 5.589 MeV of kinetic energy is converted into mass overcome the negative Q-value and 0.188MeV goes into the kinetic energy of the ^{215}At. However, if this reaction is carried out with a 5.777 MeV ^{7}Li, no reactions are observed. Why?

Problem: What is the excitation energy for the above reaction at a ^{7}Li bombarding energy of 40.0 MeV?

$$E^* = (208/215)\ 40.0\ \text{MeV} - 5.589 = \underline{\mathbf{33.1\ MeV}}$$

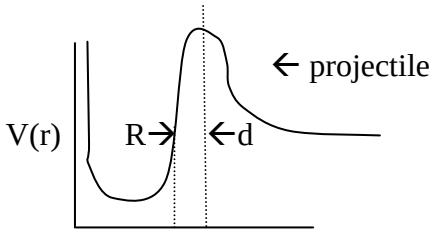
This excitation energy can also be expressed in terms of a nuclear temperature T,

$$T = (E^*/a)^{1/2}\,, \qquad \text{(Eq. 15.7)}$$

where a is the **level density parameter**, a ~ A/10 MeV^{-1} , which is related to the density of nuclear energy levels at a given excitation energy. The conversion from MeV to Kelvin temperature is 1.16 x 10^{10} K/MeV, so that the temperature of the compound nucleus formed in the above reaction at a bombarding energy of 40.0 MeV is 4 x 10^{11} K.

<u>The Coulomb Barrier</u>

The failure to observe nuclear reaction `at a bombarding energy of 5.777 MeV, even though energy and momentum are conserve can be traced to the repulsion between two positively-charged nuclei, the **Coulomb Barrier**, which imposes another restriction on the reaction energetics. Just as alpha particles must penetrate the Coulomb barrier to escape the attractive nuclear potential, the same barrier inhibits charged particles from entering the region of nuclear attraction, as illustrated in Fig. 11.1.



r

Fig. 15.1 Potential energy diagram of a projectile encountering the Coulomb Barrier of a target nucleus. R represents the half-thickness radius (Section 4) and d represents the distance at which the Coulomb barrier is a maximum.

According to Coulomb's Law,

$$V_{Coul} = \frac{(Z_p e)(Z_T e)}{d} = \frac{Z_p Z_T e^2}{R_p + R_T} \quad ,$$

where $\mathbf{d = r_o(A_p^{1/3} + A_T^{1/3})}$ with $r_o \sim 1.60$ fm being a typical value for most reactions. The difference between the radius parameter r_0 for the Coulomb barrier and for the nuclear potential (~1.44fm) is due to the diffuse nature of the nuclear surface. As the nuclei approach one another, the diffuse tails may overlap , permitting reactions to occur at a larger radius. Inserting the radius parameter d and the electric charge constant e into the above equation gives

$$V_{Coul}^{cm} = \frac{1.44\, Z_p Z_T \text{ MeV} - \text{fm}}{r_0(A_p^{1/3} + A_T^{1/3})} = \frac{0.90\, Z_p Z_T}{(A_p^{1/3} + A_T^{1/3})} \text{ MeV} \qquad (\text{Eq. } 15.8)$$

As was the case with the threshold energy, linear momentum conservation requires that the target nucleus carry off some momentum and kinetic energy. The same center-of-mass-to laboratory(cm) conversion factor (eq. 15.5) applies in this case as well, giving

$$V_{Coul}^{lab} = \left(\frac{A_{CN}}{A_T}\right) V_{Coul}^{cm} \, . \qquad (\text{Eq. } 15.9)$$

Problem: Calculate the Coulomb Barrier for the reaction of ^{7}Li with ^{208}Pb. From Eq. 15.8,

$$V_{Coul}^{lab} = \left(\frac{215}{208}\right) \frac{(82)(3)(0.9)}{(208^{1/3} + 7^{1/3})} = \mathbf{29.1\ MeV}$$

At this bombarding energy, which is much larger than the threshold energy, nuclear reactions begin to occur.

In summary, the threshold energy is an exact condition, set by conservation of mass-energy and linear momentum. In contrast, the Coulomb Barrier is an approximate condition due to the diffuse nature of the nuclear surface. The bottom line as to which of these two conditions must be met is that **the largest of either the threshold energy OR the Coulomb Barrier sets the minimum projectile kinetic energy for the reaction to occur.**

There is also one additional barrier, the **centrifugal barrier**, that must be considered if angular momentum is imparted to the target nucleus. The Coulomb trajectory for a projectile is depicted in Fig.15.2.

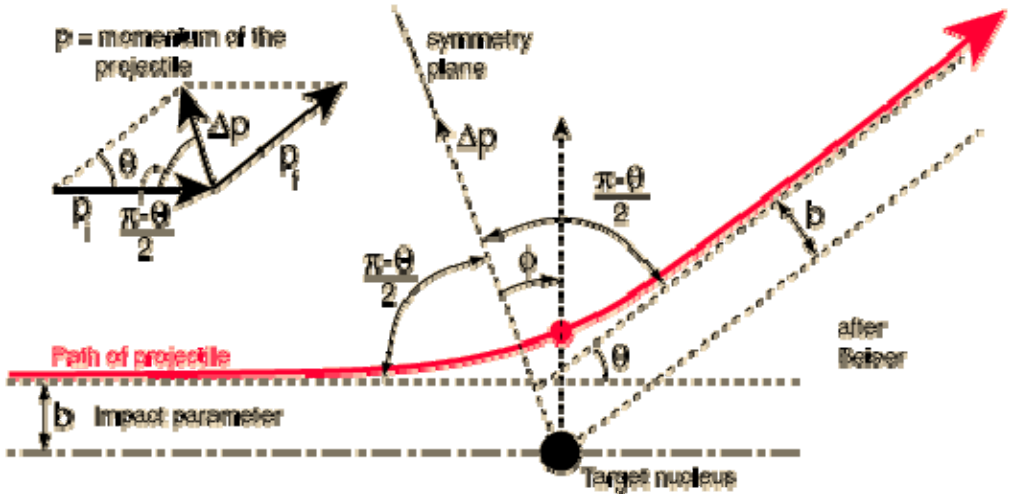


Fig 15.2 The Coulomb trajectory of a projectile incident upon a target nucleus, where θ isthe angle of deflection from the incident path; b is the impact parameter, the distance from the trajectory that coincides with the center of the target nucleus and Δp is the momentum change. It was this sort of analysis that led Rutherford to deduce the size of the nucleus.

The rotational energy of a nucleus is given by

$$E_{rot} = \frac{\ell(\ell+1)\hbar^2}{2I}$$

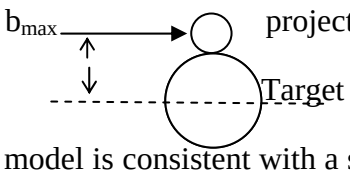
Here the orbital angular momentum ℓ = mvb and I = μr^2, where μ is the reduced mass of the system. Since the orbital angular momentum can always be zero, this is not a minimum condition for reaction.

In general, for most reactions the Coulomb barrier is higher than the threshold energy, the exception being found among light target nuclei. For neutrons, of course, there is no Coulomb Barrier and the Q-values (binding energies also) are positive so that there is no minimum energy for most neutron-induced reactions.

Reaction Probability

The probability for a nuclear reaction is expressed in terms of a **cross-section**, denoted by **σ**, in effect the relative cross-sectional area presented by the target to the incoming projectile. Since a nuclear reaction involves a collision between two nuclei, this corresponds to a second-order rate law, just as in chemical reactions.

As an initial simplification, we will adopt a simple geometric model of touching-spheres.



$b_{max} = R_p + R_T = r_o(A_p^{1/3} + A_T^{1/3})$

Such a model is consistent with a square-well potential (Sec. 9): i.e. the nuclear force is either on or off. Thus if the target and projectile touch ($b \leq R_p + R_T$), there is a reaction. If $b > R_p + R_T$, then there is no reaction. The probability that the projectile will hit the target is given by the cross-sectional area defined by the two touching spheres, called the **total reaction cross section σ_R**, where

$$\sigma_R = \text{area} = \pi(R_P + R_T)^2 = \pi r_0^2 (A_p^{1/3} + A_T^{1/3})^2 . \qquad \text{(Eq. 15.10)}$$

As mentioned above, an empirical value of $r_o \sim 1.60$ fm gives a good approximation to the data. The standard cross-section unit for nuclear reactions is the **barn (b) ,**

$$\textbf{1 barn} = 1 \times 10^{-24}\ \textbf{cm}^2 = \textbf{1b.}$$

A nuclear reaction is a sequential process that evolves through the initial collision stage, followed by the redistribution of energy and particles and finally disintegration of the system into the final products, as illustrated below.

COLLISION STAGE		DECAY STAGE
(entrance channel)		(exit channel)
$\tau \sim 10^{-21} - 10^{-23}$ s	MASS – ENERGY	$\tau \sim 10^{-15} - 10^{-21}$ s
	EQUILIBRATION →	
σ_R		$\sigma(a,b)$
Collision Probability		Probability of forming b

The time scale of the collision ($\sim 10^{-21} - 10^{-23}$ s) depends upon the velocity of the incident projectile and the size of the target. The intermediate that is formed during the mass-energy equilibration process is analogous to an activated complex in chemistry. Reactions in which the projectile and target fuse together are called **complete fusion** or **composite nucleus formation** reactions. Some reactions, especially at higher energies involve interaction of the projectile with only a portion of the target nucleons. These are usually referred to as **direct reactions**. The time scale of the decay stage depends upon the type of particle(s) that is emitted and the excitation energy of the composite nucleus. The final stage of the reaction may result in several products, each characterized by a **partial cross section, $\sigma(a,b)$.** The sum of all partial cross sections constitutes the total production cross section and is equal to the reaction cross section,

$$\sigma_R = \sum \sigma(a,b).$$

As an example, consider the following reaction

$$40\text{-MeV } {}^{4}\text{He} + {}^{232}\text{Th} \rightarrow {}^{236}\text{U}^{*} \rightarrow \text{products.}$$

the measured partial cross sections are shown in Table 15.2, along with their sum.

Table 11.2

Reaction	Cross Section	Product
$\sigma(\alpha, pn)$	= 0.010 b	${}^{234}_{91}\text{Pa}$
$\sigma(\alpha, 2n)$	= 0.080 b	${}^{234}\text{U}$
$\sigma(\alpha, 3n)$	= 0.245 b	${}^{233}\text{U}$
$\sigma(\alpha, 4n)$	= 0.125 b	${}^{232}\text{U}$
$\sigma(\alpha, f)$	= 1.190 b	fission
σ_R	= 1.650 b	= $\sum\sigma(\alpha, x)$

The total reaction cross section for charged-particle-induced reactions is usually 3 b or less.

Nuclear Reaction Rates

The rate law for an elementary bimolecular reaction

$$A + B \rightarrow C + D$$

is given by the second-order expression

$$\text{Rate} = -\frac{d[A]}{dt} = k[A][B].$$

The terms in this expression can be interpreted as follows. The Factor [**A][B]** defines the **collision probability** and depends on collision geometry. For two gases or liquids, concentration accounts for the fact that reactions can occur throughout the total volume. Instead, for a molecular beam incident on a gas, it is the number of beam particles per unit time that must be considered. The rate constant k defines the probability of a reaction -- if a collision occurs -- and is a function of the activation energy, temperature, structural orientation of the molecules, etc.

For nuclear reactions there are two general cases: particle-beam-induced reactions and reactions in a nuclear reactor. For this purpose the general second-order rate equation can be modified by

replacing the collision factor [A][B] by $n_p n_T$, the product of the number of projectiles times the number of target nuclei, each defined by the collision geometry, described below. The rate constant k is the cross section σ. The rate expression then becomes

$$\text{Rate} = dN/dt = \sigma\, n_p n_T,$$

where the rate may indicate the number of reactions per unit time (σ_R) or the number of product nuclei per unit time (σ(a, b)).

For reactions induced by a beam of charges particles, n_p (i) = N(i)/t, the number of projectiles per unit time (a current) incident on a target of thickness x is attenuated to n_p (f) = N(f)/t particles per unit time, as shown in Fig. 15.3.

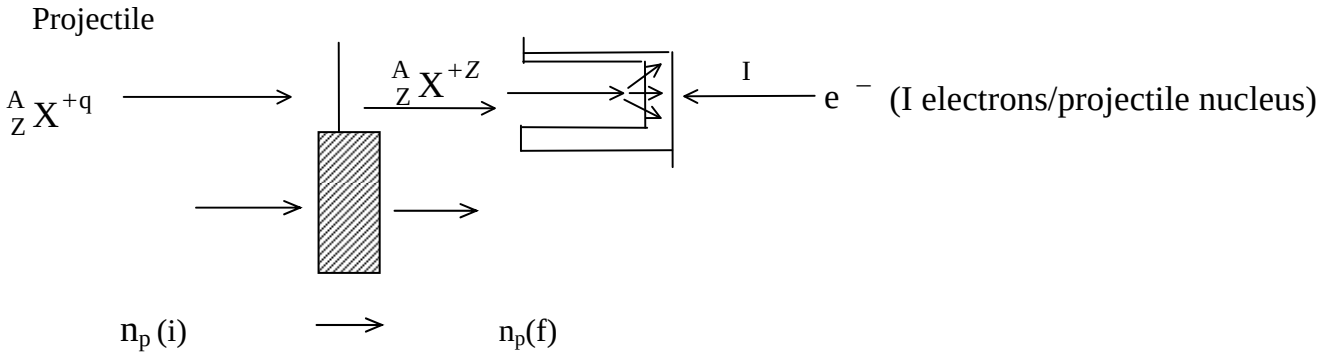


Fig 15.3 Schematic layout of a beam-induced nuclear reaction. Top: A beam of charged particles ^{A}X with atomic ionization state +q and initial intensity n_p(i) is accelerated and made incident on a target. In passing through the target the projectile is stripped of any remaining electrons to a charge state +Ze . A device called a Faraday cup collects the electric charge of the beam, which when integrated, permits counting of the beam. Bottom: Expanded view of the target, showing the decrease in beam intensity from n_p(i) to a final intensity n_p(f) .

The rate of reaction in this geometry is

$$\text{Rate} = n_p(i) - n_p(f) = dn_p/dx = \sigma\, n_t\, n_p\, dx\,,$$

where $\mathbf{n_t}$ is the number density of target nuclei $\mathbf{N/cm^3 = \rho\, N_0/(g\text{–}at.\ wt.)}$. Integrating this expression gives

$$n_p = n_p(i)\ e^{-n\sigma x}\,, \qquad \text{(Eq.15.11)}$$

which is recognized as the Beer-Lambert Law, familiar to chemical analysis. Here we assume that the cross section does not change with target thickness due to the energy degradation of the beam in passing through the target Not a valid assumption for very thick targets). The number of projectiles per unit time is measured by the beam current I in Amperes A, (1 A = 6.28 X 10^{18} e^- /second), where

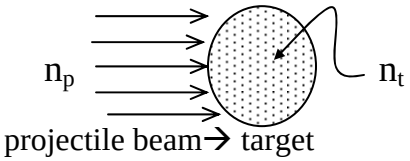
$$n_p = I/Z \,, \qquad \text{(Eq. 15.12)}$$

and Z = +q, the charge on the projectile ion after passing through the target.

.

In the case of a thin target ($\Delta x \sim 0$) , which is frequently the case, there are no shadowed nuclei and the target can be approximated as presenting a cross-sectional area to the beam. Expanding Eq. 15.11 in the limit $\Delta x \sim 0$, the rate (number of reactions per unit time, dN/dt)

$$\text{Rate} = (I/Z)\sigma(n_t x) \,. \qquad \text{(Eq. 15.13)}$$



The factor $n_t x$ in Eqs. 15.11 and 15.13 has the units of atoms per unit volume times thickness, or atoms/cm^2 and can be expressed in two ways, either

$n_t x$ = density x thickness x N = (g/cm^3)x cm x (N_o/g-at.wt.) or

$n_t x$ = aerial density x N = g/cm^2 x N_o/g-at.wt.) .

Problem: After passing through a target, the beam current of ^{12}C ion is 10.0 nA. How many ^{12}C ions per second is this?

$n_p = I/Z = (10.0 \times 10^{-9}\text{A})(6.28 \times 10^{18}\text{e/A-s})/6\text{e}^- =$ **1.05×10^{10} ^{12}C/s**

Problem: What is the production rate of $^{266}_{106}Sg$ if a 100 μg/cm^{2}target of $^{248}_{96}\text{Cm}$ (thin) is bombarded with a 1.0 μA beam of ^{22}Ne ions? What is the fission rate?

σ (^{22}Ne, 4n) = 1.0 nb σ (^{22}Ne, f) = 2.5 b

R = $(I/Z)\sigma(n_t x)$ for thin targets

$\sigma = (1.0 \times 10^{-24}\ \text{cm}^2)(10^{-9}) = 1.0 \times 10^{-33}\ \text{cm}^2$

$$(n_t x) = \frac{(100\times10^{-6})\text{g}\ (6.02 \times 10^{23}\ \text{atoms/mole})}{248\ \text{g/mole}} = 2.43 \times 10^{17}\ \text{atoms/cm}^2$$

$I/Z = 1.0\ \mu\text{A} \times 6.28 \times 10^{12}\ \text{e}^-/\mu\text{A}\ /\ (10\ \text{e}^-/\text{Ne}) = 6.28 \times 10^{11}\ \text{Ne/s}$

$R = 1.5 \times 10^{-4}\ {}^{266}_{106}Sg\ /s = 0.54/hr$

$R = 7.6 \times 10^{5}$ fission fragments/s NOTE: 2 fragments/fission

Decay During Bombardment

When the product of a nuclear reaction has a half-life that is short with respect to the bombardment time, Eqs. 15.11 and 15.13 must be modified to account for radioactive decay in competition with production. Assuming that the production rate is independent of time, this problem is analogous to secular equilibrium. Decay during bombardment is an important consideration in problems such as the production of radioisotopes for nuclear medicine and in understanding ^{14}C production in the atmosphere.

$$\frac{dN}{dt} = \underset{\text{Production}}{R} - \underset{\text{Decay}}{\lambda N}$$

The solution to this differential equation is

$$N = \frac{R}{\lambda}\left(1 - e^{-\lambda T}\right), \qquad \text{(Eq.15.14)}$$

or in terms of activity, $N\lambda = R\left(1 - e^{-\lambda T}\right) = \frac{A}{c}$,

where T is the duration of the bombardment. The factor $\mathbf{(1 - e^{-\lambda T})}$ is called the **saturation factor** and is an important consideration in deciding the length of bombardment time required for obtaining a desired amount of product. As T becomes long with respect to the half-life, the exponential term goes to zero and Eq. 15.14 become constant. Accelerator time may be expensive and therefore the law of diminishing returns sets in; e.g.

$T = t_{1/2}$,	$N = (R/\lambda)(1\text{-}1/2)$	=	$(R/\lambda)(1/2)$; 50% of maximum
$T = 2t_{1/2}$,	$N = (R/\lambda)(1\text{-}1/4)$	=	$(R/\lambda)(3/4)$; 75% of maximum
$T = 3t_{1/2}$,	$N = (R/\lambda)(1\text{-}1/8)$	=	$(R/\lambda)(7/8)$; 87.5% of maximum, etc.

The chronology of an irradiation is outlined below:

time ↓

(1) Accelerator ON $\left\{\begin{array}{l} A + B \rightarrow C \\ C \rightarrow D \end{array}\right\}$ time = 0 $N_c(T) = (R/\lambda)(1 - e^{-\lambda T})$

$N_c(t\text{-}T) = N_c(T)\ e^{-\lambda(t-T)})$

(2) Accelerator OFF _____ T

(3) Measure Sample _____ | t

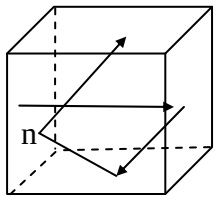
In step (1) (accelerator on) the product is being produced and undergoing decay simultaneously. Step (2) (accelerator off) involves only decay of the sample after the irradiation has ceased. Step (3) involves measurement of the sample as it decays away according to the first order decay law.

Neutron-Induced Reactions

For neutron-induced reactions, there is no Coulomb barrier. The binding energies of all stable nuclei in their ground states are positive, and since $B_n = Q$, **neutron-capture reactions**

$$ {}^{1}_{0}n + {}^{A}_{Z}X \rightarrow {}^{A+1}_{Z}X + Q . $$

are always energetically allowed for stable isotopes. If such reactions are induced by a beam of neutrons, Eqs. 11.10. 11.12 and 11.13 apply, just as with beams of charged particles. The more common situation, however, involves irradiation in a nuclear reactor. Schematically, this situation is analogous to a neutron gas inside the reactor that permeates a solid or liquid target. Thus all target nuclei are accessible to neutrons, as the neutrons fill the volume of the sample and are reflected from the walls of the reactor.



The collision term n_p for the neutrons depends on their concentration, neutrons per unit volume n_n, and their velocity v. This product is called the **neutron flux Φ,** where

$$ \Phi = n_n v. $$

All target atoms are available for reaction so that $n_T = N_T$, the number of target nuclei. With these definitions Eq. 15.13 becomes

$$ \text{Rate} = R = \sigma \Phi N_T \quad \text{and} \qquad \text{(Eq. 15.15a)} $$

$$ N\lambda = R(1 - e^{-\lambda T}). \qquad \text{(Eq. 15.15b)} $$

Unlike charged-particle reactions, neutron-capture cross sections are highly variable, spanning a range from ~ 0.1 to 10^4 b. Two important reactions in nuclear reactor technology are $^{10}B(n,\alpha)$

with a cross section of 3838 b and $^{113}Cd(n,\gamma) = 2.0 \times 10^4$ b. Neutron-capture cross sections tend to be large and span a broad range of values from ~0.1 to to 10^4 b. Very large cross sections can result when the energy of the incident neutron oveerlaps with an energy state in the A + 1 daughter nucleus, creating a **resonance effect** that enhances the reaction probability. The cross sections also become large when the irradiation is carried out with **thermal neutrons** (i.e. neutrons with kinetic energies comparable to room temperature). For very low energy the cross section increases as the velocity decreases – the **1/v law** that is important in the operation of nuclear reactors.

Neutron Activation Analysis (NAA)

One widespread use of neutron-capture reactions is **neutron-activation analysis**, which is capable of identifying about two-thirds of the chemical elements. With a sensitivity of picograms, it is particularly applicable to many heavy elements that are low-level environmental contaminants. However, it does not yield information about chemical composition, only the elements present, and is not sensitive for the biological elements hydrogen, carbon, nitrogen and oxygen. Among the many applications of the technique are:

- Environmental pollutants relevant to atmospheric chemistry, marine samples, water quality; e.g. V,Cu, Cd,PCBs, Hg
- Cosmology: trace elements in meteorites and lunar samples
- Geology: mineral analysis, volcanic versus meteoritic theories of Cretaceous-Tertiary extinction
- Medicine: trace elements in body fluids
- Criminolgy: forensic clues in gunpowder residues
- Art history: detection of art forgeries from trace elements in paint bases
- Agriculture: herbicide and pesticide residue migration
- Electronics, metallurgy and petroleum engineering: identification of trace element contaminants

The procedure is illustrated in Fig. 15.4.

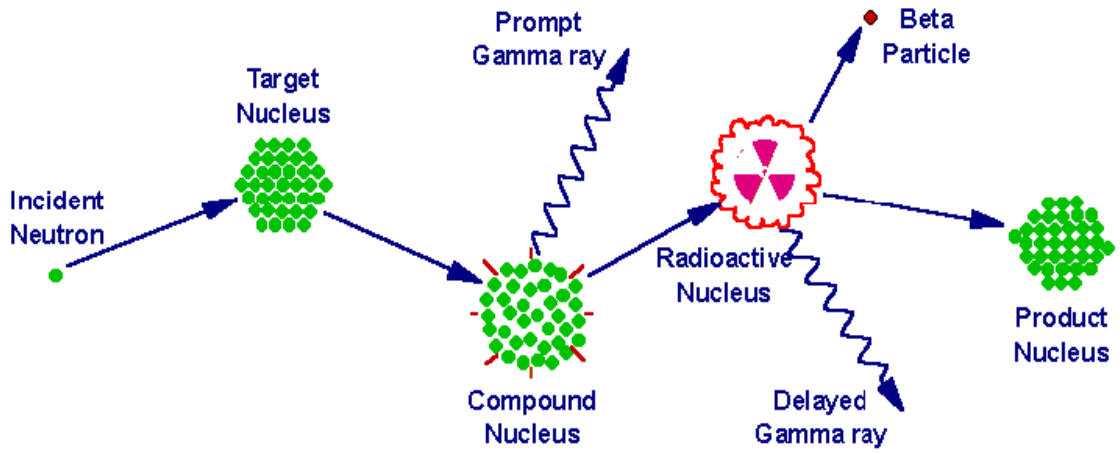


Fig. 15.4 Incident neutrons irradiate target nuclei in an unknown sample to form a compound nucleus, which emits a prompt gamma ray to produce a radioactive product nucleus. The decay of the product emits beta particles and gamma rays that can be used as fingerprints of the product A + 1 nucleus. Because neutrons do not increase the atomic number, identification of the product defines the element and isotope(s) of the constituents of the sample.

In order to achieve maximum sensitivity, a large flux of low-energy neutrons is desired, conditions that are readily available in a nuclear reactor. Fig. 15.5 show the range of neutron energies (in eV) as a function of relative neutron flux in a typical reactor.

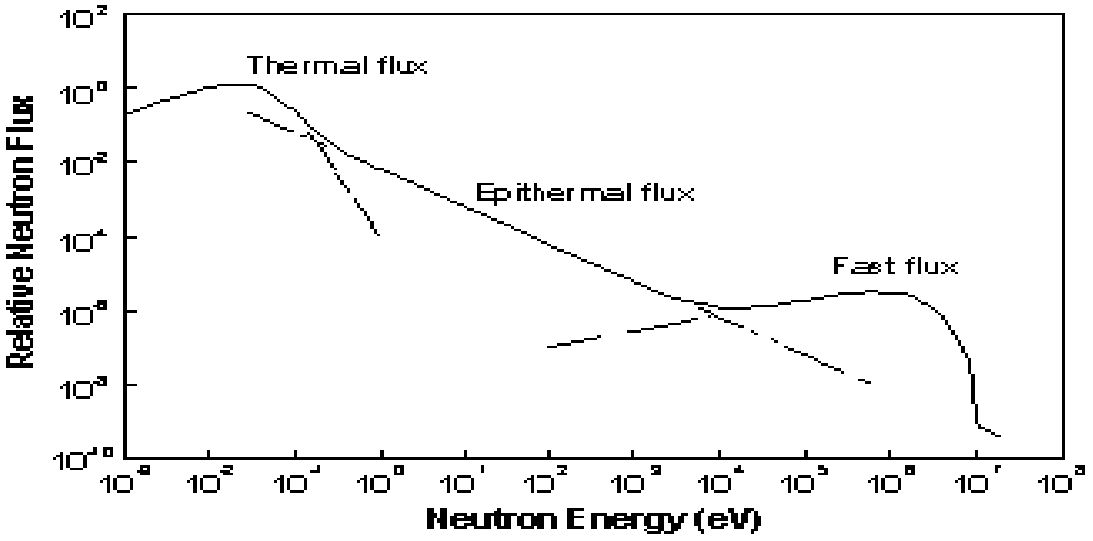


Fig. 15.5 Three major neutron flux distributions in a nuclear reactor: thermal flux, corresponding to room temperature velocities (meV); epithermal flux at intermediate energies (eV – keV), and fast flux due to fission reactions (keV – MeV).

Element identification is achieved by gamma-ray detection. The gammas may be either prompt gammas associated with the capture reaction (PGNAA) or gammas that have their origin in the radioactive decay of the product nucleus (NAA). A typical NAA spectrum is shown in Fig 15.6, showing the identification of 19 different elements.

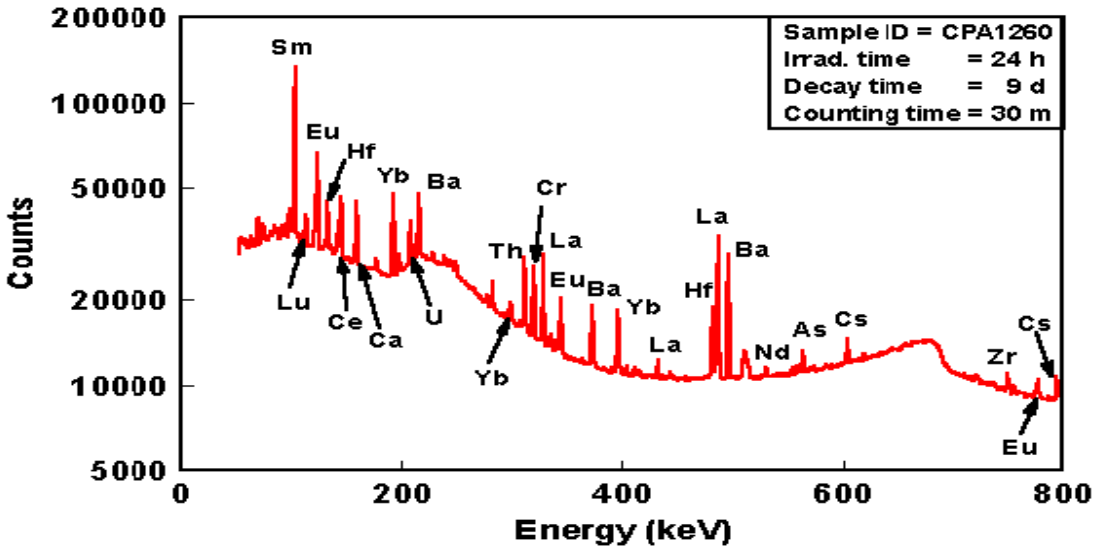


Fig. 15.6 Gamma-ray spectrum of a sample of pottery that has been irradiated for 24 hours and counted for 30 minutes nine days later.

PGNAA is most useful when the cross section is large, since it has the advantage of instant feedback during the irradiation. Conventional NAA requires removal of the sample and off-line counting of the sample. The following problem illustrates the application of the NAA technique.

Problem: What is the sensitivity (in grams) for chromium with NAA?

Step I: Select the **best product** nucleus for detection from the four naturally-occurring isotopes of Cr:

$^{50}Cr(4.35\%)\ ;\ \sigma(n,\gamma) \quad = 15.96\ b \quad \rightarrow {}^{51}Cr(27.7\ d\ EC) + \gamma$

$^{52}Cr(83.79\%)\ ;\ \sigma(n,\gamma) \quad = \rightarrow {}^{53}Cr(stable) \quad$ not useful

$^{53}Cr(9.50\%)\ ;\ \sigma(n,\gamma) \quad = \rightarrow {}^{54}Cr(stable) \quad$ not useful

$^{54}Cr(2.36\%)\ ;\ \sigma(n,\gamma) \quad = 0.386\ b \quad \rightarrow {}^{55}Cr(3.55\ min\ \beta^-)$

Which is better, ^{50}Cr or ^{54}Cr ? ^{50}Cr has a larger abundance and larger cross section but its half-life is a bit long. However, it decays to an excited state in ^{51}Cr that emits an easily detectable gamma ray. ^{54}Cr undergoes beta decay with a relatively short half-life, which is more difficult to quantify. Therefore **select ^{50}Cr** and **count ^{51}Cr**.

Step II: Irradiate the sample, remove it from the reactor and count. As is the case in any analytical procedure, sample preparation is important, especially the need to avoid introducing contaminants and in the case of reactors, guaranteeing the thermal stability of the sample. Ideally, it is desirable to count gamma rays. Gammas provide unique isotopic signatures and penetrate matter readily, removing the need for chemical separations that can affect yield determinations and introduce contaminants.

Step III: Calculate yield

For optimum sensitivity we make the following assumptions:

$\Phi = nv = 1.0 \times 10^{16}$ n/cm^2-s (usually 10-100 times less)

$T_{irradiation} = 10.0$ d

Detection limit = 0.100 dps for ^{51}Cr (corrected for background)

t - T = 5.0 days

- Due to the time between the irradiation and counting of the sample, correction for decay must be made since).100 dps is the **final** desired rate This requires correction for decay :

$$A(^{51}Cr)_{t-T} = A(^{51}Cr)_T\, e^{-\lambda t}$$

$$0.100 \text{ dps} = A(^{51}Cr)_T\, (e^{-0.693(5.0d)/27.7d})$$

$$A(^{51}Cr)_T = \frac{0.10 \text{ dps}}{0.882} = \mathbf{0.113 \text{ dps at end of irradiation.}}$$

- The reaction rate R then becomes:

$$R = \sigma\, N(^{50}Cr)\Phi = (15.96 \times 10^{-24}\ \text{cm}^2)(\mathbf{N[^{50}Cr]})(1.0\times10^{16}\ \text{n/cm}^2\text{–s}).$$

- Accounting for the 10.0 day bombardment, the activity of ^{51}Cr nuclei is then

$$N_T(^{51}Cr)\lambda(^{51}Cr) = R\,(1 - e^{-0.693(10.0\text{ d}/27.27\text{ d})}) = R(1 - 0.779) = 0.221\,R = 0.113 \text{ dps}$$

$$(0.221)(15.96 \times 10^{-24}\ \text{cm}^2)(1.0\times10^{16}\ \text{n/cm}^2\text{–s})\mathbf{N(^{50}Cr)} = 0.113 \text{ dps}$$

$$N(^{50}Cr) = (0.113)/(0.221)(15.96)(10^{-8}) = \mathbf{3.20 \times 10^6 \text{ atoms of } ^{50}Cr}$$

- In order to determine total chromium, the isotopic abundance of ^{50}Cr must be taken into account:

  $N(Cr) = N(^{50}Cr)/abundance = 3.20 \times 10^6/0.0435 =$ **7.36×10^7 atoms**

  wt (Cr) = $(7.36 \text{ X } 10^7)(52.0 g/mole)/(6.02 \text{ X } 10^{23}) =$ **6.2×10^{-13} g**

APPENDIX 15.A

Centers of NAA

See http://www.missouri.edu/~glascock/naa_over.htm for an overview of NAA.

http://www.ncnr.nist.gov/instruments/nactanal.html NIST, Center for Neutron Research

http://ares.jsc.nasa.gov/Labs/neutronAAlab.htm, NASA

http://reactor.engr.wisc.edu/naa.html, University of Wisconsin, Nuclear Reactor Lab.

http://www.chem.tamu.edu/services/naa/, Texas A & M,

http://geology.cr.usgs.gov/facilities/gstr/, US Geological Survey $30/sample

and MANY more