22.05 Reactor Physics – Part Two

Neutron Sources and Reactions

1. Neutron Sources:

The title of this course is "Reactor Physics" which implies that the neutron source of interest is nuclear fission. However, reactor physics methods, such as diffusion theory and Monte Carlo, can be used to describe the transport of neutrons in devices other than reactors. Neutrons are extensively used in medicine (neutron capture therapy, treatment of cervical cancer), in imaging (radiography), and in industrial applications (materials). For information on the many uses of radiation, see "Radiation and Modern Life: Fulfilling Madam Curie's Dream," by Alan Walter. Many of these applications use sources other than fission reactors. Accordingly, the major sources are summarized here.

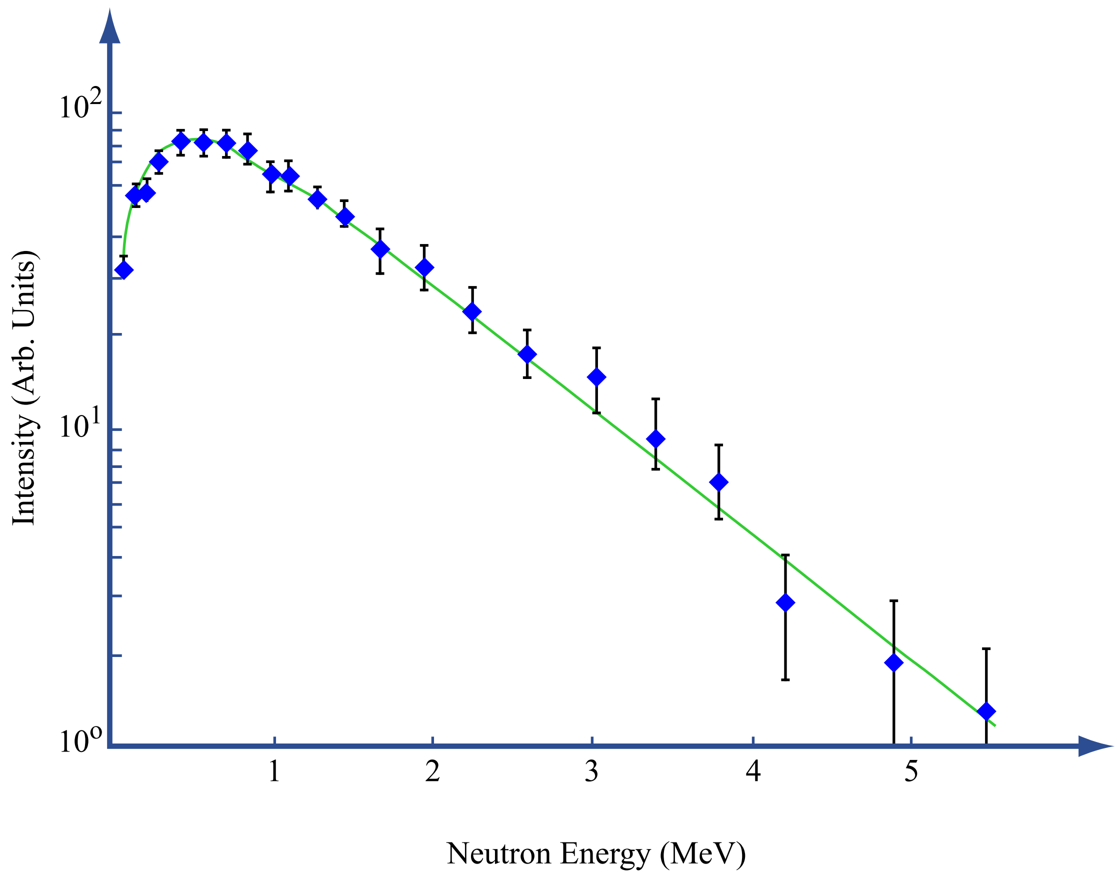
For each source, the student should be able to describe the means of neutron production and characterize the spectrum as either discrete or continuous and why. For discrete spectra, calculate the neutron energies. For continuous ones, draw the spectrum.

a) **Spontaneous Fission**: Many transuranic heavy nuclides undergo spontaneous fission at a rate sufficient to provide a useful neutron source. Each such fission produces several neutrons as well as beta and gamma rays. The most commonly used such source is Californium-252. It undergoes an alpha decay with a 2.65 year half-life. For every 32 alpha emissions, there is a spontaneous fission. The neutron yield is 2.3×10^6 n/s per microgram. Hence, small encapsulated sources provide significant strength. The attached figure shows the spectrum. Intensity peaks between 0.5 and 1 MeV with the most energetic neutrons being 8-10 MeV.

b) **Alpha Neutron Sources**: The most widely used of these sources is:

$$ {}^{4}_{2}He + {}^{9}_{4}Be \rightarrow {}^{1}_{0}n + {}^{12}_{6}C + Q $$

The source of the alpha particle is often Pu-239 which is chosen because it is long-lived and provides an energetic alpha. Other nuclides (Po, Am) can be used. The source is constructed by mixing Pu and Be powder within a double layer of stainless steel. The mixed powder (as opposed to a Pu rod in a Be enclosure) is needed because of the short range of alpha particles. Unless, the Pu and Be are in close contact, the alphas will



Measured neutron energy spectrum from the spontaneous fission of ^{252}Cf. (From Batenkov et al.)

Figure by MIT OCW. From Knoll.

attenuate before striking the Be and no neutrons will be produced. The double layer of stainless steel is needed to contain the toxicity of the Pu. Heat transfer is poor because of the double layer of steel. Hence, Pu-Be sources must not be left in significant neutron fluxes or they will melt. Alpha particles are helium nuclei and form helium atoms once they become electrically neutral. Gas buildup can cause cracking of the steel case. Leak testing, usually every six months, is done for safety by surveying the exterior of the steel case for alpha activity (i.e., wipe the steel with wet filter paper and count same for alpha). The Q-value is 5.74 MeV.

The energy of the alpha particle produced from Pu decay is discrete and the reaction of the alpha on Be is a two body collision. Hence, one expects a discrete neutron spectrum. In reality, the spectrum is continuous because the alpha loses some energy before striking the Be. A figure that gives the spectrum follows. Source strength is typically 10^7 n/s and this requires about 16 mg of material.

c) **<u>Photo Neutron</u>**: A photon that is absorbed by the nucleus creates an excited state from which a neutron is emitted. There are two such sources:

$${}^0_0\gamma + {}^9_4\mathrm{Be} \rightarrow {}^8_4\mathrm{Be} + {}^1_0\mathrm{n};\ Q = -1.67\,\mathrm{MeV}$$

$${}^0_0\gamma + {}^2_1\mathrm{H} \rightarrow {}^1_1\mathrm{H} + {}^1_0\mathrm{n};\ Q = -2.23\ \mathrm{MeV}$$

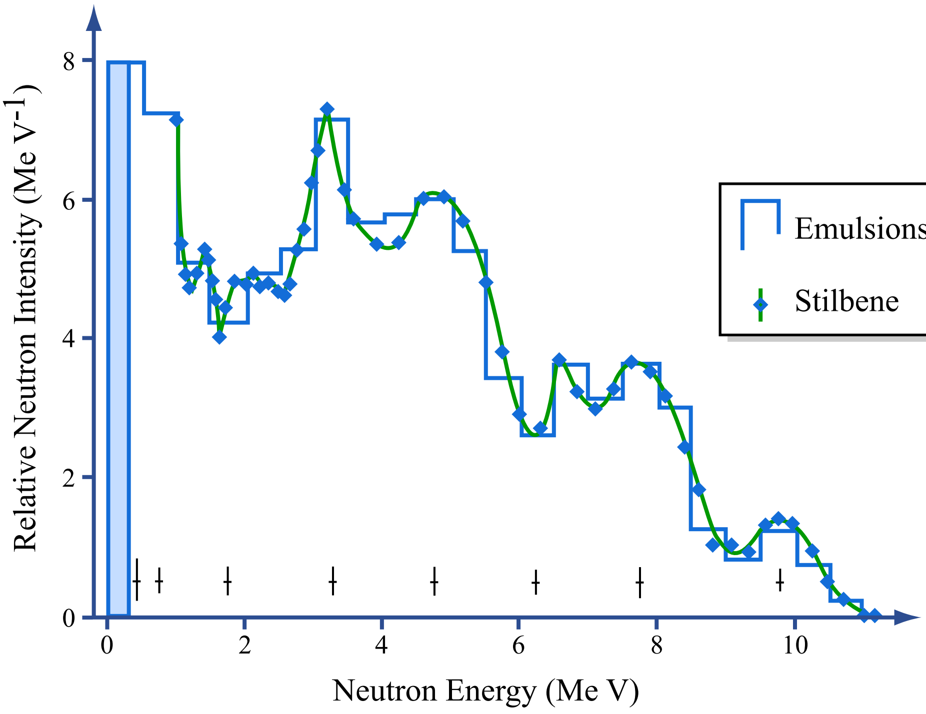
The resulting neutron energies are discrete if the photons are mono-energetic. Roughly, one gamma ray in 10^6 interacts. So, the gamma ray source needs to be very large (as in a fission reactor) for these sources to be appreciable. The most common use is the deuterium reaction as a source of neutrons for the startup of light-water reactors. The source of the photons would be fission products. (<u>Note</u>: Sufficient D_2O exists in light water for this source to be effective in LWRs.)

d) **<u>Accelerated Charged Particles</u>**: Common reactions are deuterons on light nuclei. The Coulomb barrier for such nuclei is not too great and reactions can proceed if the deuterons are accelerated to a few hundred keV. Typical reactions are:

$${}^2_1H + {}^2_1H \rightarrow {}^3_2He + {}^1_0n;\ Q = 3.26\,MeV$$

$${}^2_1H + {}^3_1H \rightarrow {}^4_2He + {}^1_0n;\ Q = 17.6\,MeV$$

The energy of the incident particle is small (keV) compared to the Q-value (MeV). Hence, the monoenergetic neutrons are produced at about 3 and



Measured energy spectra for neutrons from a ^{239}Pu/Be source containing 80g of the isotope. (From Anderson and Neff.)

Figure by MIT OCW. From Knoll.

14 MeV for the above reactions respectively. Yields are high with a 1 mA beam of deuterons providing 10^9 n/s for the deuteron reaction and 10^{11} n/s for the tritium reaction.

e) **Fission/Fusion**: In order to describe these, it is first appropriate to review binding energy.

2. Binding Energy:

The difference between the mass of a nuclide and its constituent nucleons is called the mass defect:

$$\Delta = Zm_p + Nm_n - M_A$$

Where Δ is the mass defect, Z is the atomic number (number of protons), N is the number of neutrons, and m_p, m_n, and M_A are the masses of the proton, neutron, and nuclide respectively. Tabulations of atomic masses are readily available. So, it is useful to convert the above equation to atomic masses by adding Z electrons. Thus:

$$\Delta = Z(m_p + m_e) - Nm_n - (M_A + Zm_e)$$

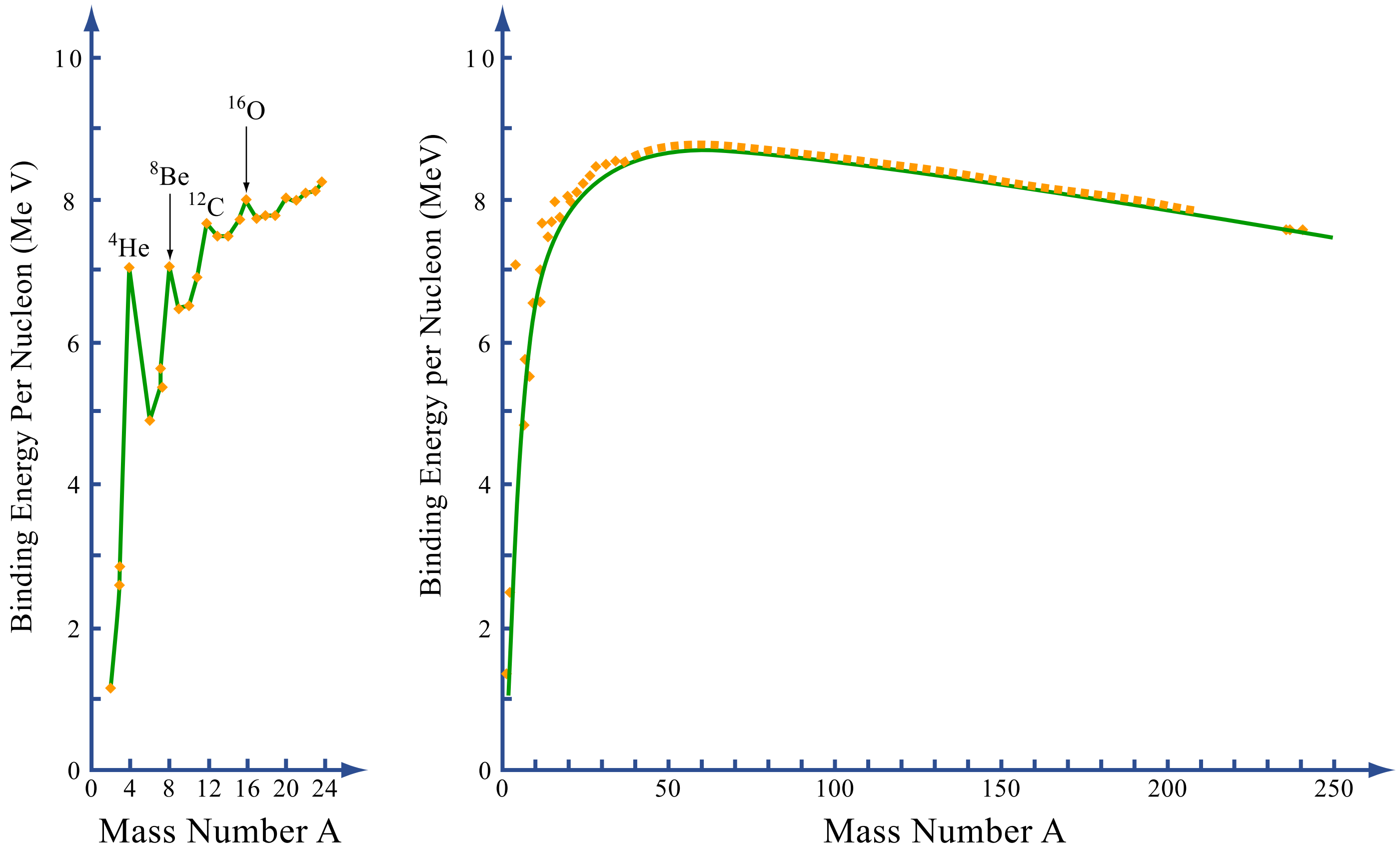
$$= Z(\text{Mass Neutral Hydrogen}) + N(\text{Mass Neutrons}) - \text{Atomic Mass of Neutral Atom}$$

If the mass defect is multiplied by c^2, it is expressed in energy units, and is called the "binding energy" or BE. The attached figure shows the BE per nucleon as a function of mass number. Light nuclei are not tightly bound (small binding energy). The BE rises rapidly with mass number and reaches a peak of about 8.7 MeV for iron. Hence, iron is the most stable nuclide. For nuclides heavier than iron, the BE slowly decreases. The implication of the curve is:

- Fusion occurs for a nuclide below iron. For these nuclides, the BE increases (energy is released) if the light nuclei combine (or fuse) to form a heavier one.

- Fission occurs for nuclides above iron. For these nuclides, the BE increases (energy released) if a heavy nuclide splits (or fissions) to form two light ones.

The BE curve can be used to explain stellar formation and the origin of the elements. Stars form when clouds of hydrogen contract under gravity. The contraction causes the gas to heat up thereby creating a plasma of free electrons and protons (hydrogen nuclei stripped of electrons) and then igniting the reaction:



The average binding energy BE/A of the nucleons in a nucleus versus the atomic mass number A for the naturally occurring nuclides (and ^{8}Be). The left-hand figure shows the detailed variation of the average binding energy for the lightest nuclei, while the right-hand figure shows the overall variation.

Figure by MIT OCW. From Shultis.

$$ {}_{1}^{1}H + {}_{1}^{1}H \rightarrow {}_{1}^{2}D + {}_{+1}^{0}e + \nu; \qquad Q = 0.42\ \text{MeV} $$

When this occurs, the heat generated by the fusion reaction expands the hydrogen cloud and halts the contraction. The star is stable until all the hydrogen is used. Contraction then resumes until another reaction starts. The sequence (contraction, re-ignition, expansion, stable fusion) repeats until the end result is iron. Contraction now resumes. Only this time, no fusion reaction is possible because Fe + Fe would require the input of energy.

Two outcomes are feasible. For low mass stars, the star's remnants become a brown dwarf. For high mass stars, an explosion (supernova) occurs. During these few seconds, neutrons are stripped from some nuclides and bombard other nuclides. Elements heavier than iron are formed as the supernova occurs. This is why iron and the lighter elements are common in the earth's crust while heavier ones (Au, Ag, Pt) are rare. For additional information see Shultis and Faw, pp. 152-157.

3. **Fission**:

The characteristics of the strong (nuclear) force were noted earlier. They are very strong, short range, saturated, and charge independent. The range is typically 10^{-13} cm. If a neutron gets within this distance of a nucleus, it is swept into the nucleus. The nucleus may be thought of as a potential well with the depth of the well being the energy associated with the mass defect. Assume that the neutron in question has no kinetic energy. The process of its joining the nucleus increases the mass defect – the binding energy associated with the last neutron. Or, one could think of the process as equivalent to the neutron "falling" into the well. The potential energy that it had when it was beyond 10^{-13} cm is converted to kinetic energy. This energy is referred to as the "excitation" energy.

An isotope is referred to as "fissile" if the excitation energy provided by the incoming neutron is sufficient to cause fission. Thus, the incident neutron can have zero kinetic energy and still cause fission. The major fissile isotopes are:

U-233, U-235, Pu-239, and Pu-241

Of these, only U-235 is naturally available (0.71% uranium). We note the nuclear reaction needed to produce the others.

a) **U-233**

$$ {}_{0}^{1}n + {}_{90}^{232}Th \rightarrow {}_{90}^{233}Th \rightarrow {}_{-1}^{0}e + {}_{91}^{233}Pa \rightarrow {}_{-1}^{0}e + {}_{92}^{233}U $$

b) **Pu-239**

$$ {}_0^1n + {}_{92}^{238}U \rightarrow {}_{92}^{239}U \rightarrow {}_{-1}^{0}e + {}_{93}^{239}Np \rightarrow {}_{-1}^{0}e + {}_{94}^{239}Pu $$

c) **Pu-241**

$$ {}_0^1\mathrm{n} + {}_{94}^{239}\mathrm{Pu} \rightarrow {}_{94}^{240}\mathrm{Pu} $$

(Occurs 35% of time, rest of time the Pu-239 fissions.)

$$ {}_0^1n + {}_{94}^{240}Pu \rightarrow {}_{94}^{241}Pu $$

Isotopes such as Th-232, U-238, and Pu-240 are designated as "fertile" because they yield fissile material upon capture of a neutron.

Isotopes may also be classed as fissionable if they undergo fission when struck by a neutron that has high kinetic energy.

4. **Fission Process:**

The following table shows the results of the fission process. ~200 MeV of energy is released and distributed.

Product	Energy (MeV)
Fission Products	168
Fission Product Decay	
— Beta	8
— Gamma	7
— Neutrinos	12
Prompt Gamma	7
Fission Neutron	5
	207

The term "prompt" as used above means "emitted at the time of fission." The energy associated with the neutrinos is lost. This loss is balanced by 3-12 MeV of energy associated with capture gamma rays – gammas released upon capture of the fission neutrons.

(i) Fission Products

The fission fragments are usually of unequal mass as shown in the following figure which gives fission yield vs. mass number. There is a decided dip in the curve for asymmetric fission. Note that the vertical axis is log scale. On a linear plot, the dip would be even more pronounced. The nuclides at the lower peak are Mo-99/Tc-99 and at the higher peak I-135/Xe -135.

"The fission fragments, as they appear initially, are unstable with respect to the number of neutrons in their nuclei. Most commonly they become stable again through a series of β decays (emissions of electrons) which, in effect, transform the excess neutrons in the nucleus into protons. Thus, in the course of their radioactive decay to a stable end product, the fission fragments assume a sequence of chemical identities, each successive one having a nucleus containing an additional positive charge. Some of the members of these decay chains are nuclei that have a high probability for capturing neutrons. Such nuclei represent a "poison" in the system in that they compete for neutrons with the fissile material. It is unfortunate that Xe-135, which belongs to the mass 135 decay chain, is the isotope that has the highest probability for capturing low-energy neutrons." (Quoted from Henry, pp 10-11).

(ii) Prompt Neutrons

"There are typically 2 or 3 neutrons emitted per fission. This quantity (2.5 on average) is symbolized as ν. The fraction of the ν neutrons emitted in fission that are remitted between the energies E and E + dE is symbolized by $\chi^j(E)dE$, the quantity $\chi^j(E)$ being called the fission spectrum for isotope j. An empirical expression for the fission spectrum $\chi^j(E)$ that can be used in most applications for all isotopes regardless of the energy of the neutron causing the fission is

$$\chi(E)=0.453e^{-1.036E}\sinh(2.29E)^{1/2},$$

Where E is in MeV. Since $\chi(E)$ is a probability density,

$$\int_0^\infty \chi(E)dE=1$$

A plot of χ(E) is shown in an attached figure.

Note that the most probable energy of an emitted neutron is slightly below 1 MeV and that very few neutrons are emitted with an energy greater than 10 MeV.

The neutrons emitted in fission may be assumed to emerge with equal probability in all directions. In other words, fission neutrons are produced isotropically. They preserve no "memory" of the direction of travel of the neutron that caused the fission." (Quoted from Henry, p. 11)

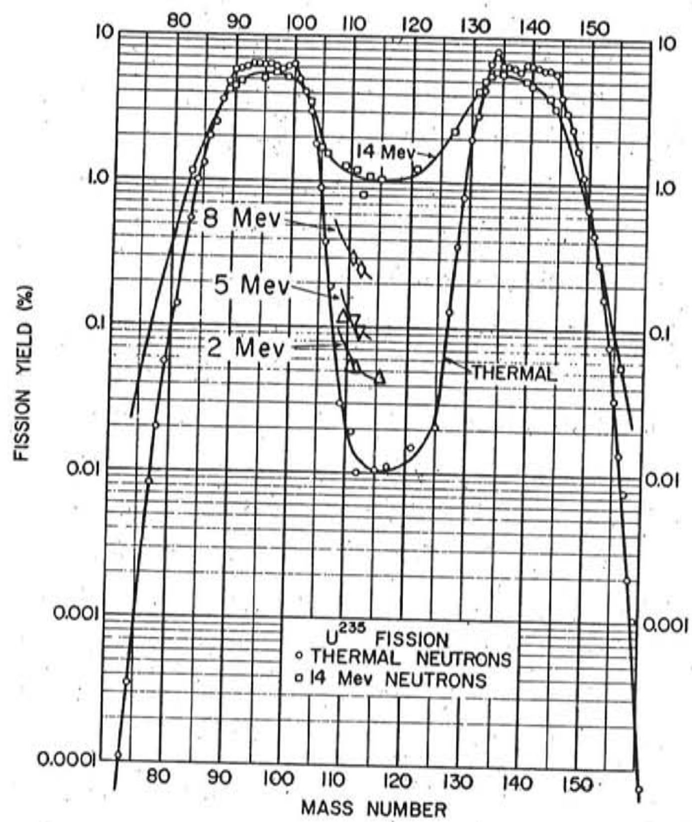


Figure 1.1 Mass-yield curve for fission of U^{235} by thermal, ~2-MeV (fission-spectrum), 5-MeV, 8-MeV, and 14-MeV neutrons. (Thermal and 14-MeV data are from the compilation of Katcoff; intermediate-energy data in the mass valley are from Ford and Gilmore.) From Keepin (1965).

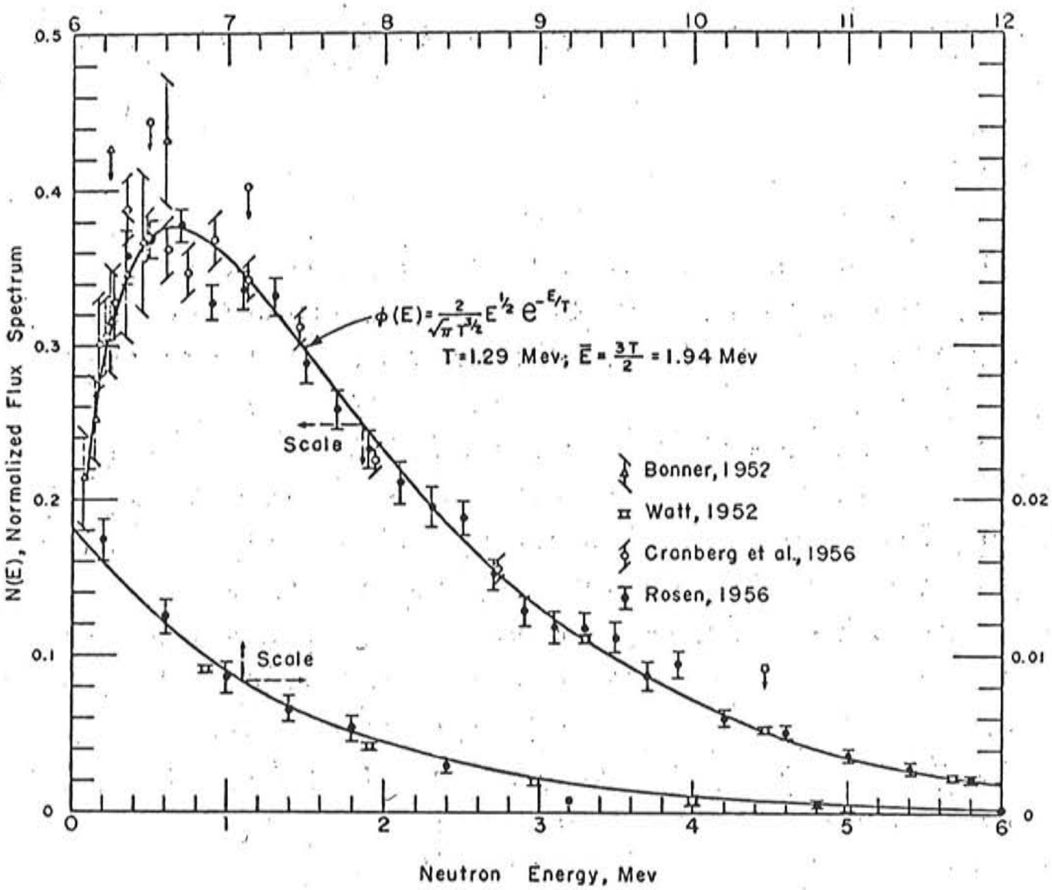


Figure 1.2 Experimental neutron energy spectrum from thermal-neutron-induced fission of U^{235}. Methods of measurement were: Bonner, cloud chamber; Watt, proton recoil; Cranberg, time-of-flight; Rosen, nuclear emulsion. The solid line shows the best-fitted Maxwellian spectral function. Arrows indicate the normalization points for each of the sets of data. From Keepin (1965).

(iii) Delayed Neutrons

Neutrons that appear at the time of fission are termed "prompt." Not all neutrons are prompt. A small fraction (0.65%) are emitted from fission fragments after the fragment has first undergone a beta decay. Recall that the fission fragments have a neutron to proton ratio that places them far from the stability line as given by the Chart of the Nuclides. The beta decay converts a neutron to a proton and if it leaves the daughter nuclide in an excited state, the daughter may then emit a neutron. Such neutrons are called "delayed" with the delay being the time required for beta decay. Fission products that lead to delayed neutrons are called "precursors." To summarize:

A precursor is a fission product that undergoes a beta decay to a daughter nuclide that then emits a neutron.

Delayed neutrons are born at lower energies then prompt neutrons. However, they are still in the range considered as fast.

4. Nuclear Reactions: A nuclear reaction occurs when two nuclei or a nucleus and a nucleon (a proton or a neutron) interact to produce two or more nuclear particles. These reactions are governed by conservation of: number of nucleons, charge, momentum, and energy. The latter includes the rest mass energy.

Consider a reaction: $a + b \rightarrow c + d$

Where M_A and E_A are the mass and kinetic energy of particle a. Same notation for b, c, d.

From conservation of energy.

$$E_a + E_b + M_a c^2 + M_b c^2 = E_c + E_d + M_c c^2 + M_d c^2$$

or

$$(E_c + E_d) - (E_a + E_b) = [(M_a + M_b) - (M_c + M_d)]c^2$$

Thus the change in the kinetic energies of the particles before and after the reaction equals the difference in the rest mass energies of the particles before and after. The latter is called the Q-value for the reaction. Thus:

$$Q = [(M_a + M_b) - (M_c + M_d)]c^2$$

The masses in the above equation are those of the nuclei, which aren't tabulated. We need atomic masses and hence add electron masses to each term. This is acceptable because charge is conserved. Thus,

$$Q = \left[\left[(M_a + Z_a m_e) + (M_b + Z_b m_e)\right] - \left[(M_c + Z_c m_e) + (M_d + Z_d m_e)\right]\right]c^2$$

Each quantity in parenthesis (e.g., $(M_a + Z_a m_e)$) is the rest mass of the neutral atom. Hence, the Q value is the difference in the atomic masses of the products and the reactants.

A reaction is exothermic if Q is positive – mass converted to energy. A reaction is endothermic if Q is negative – energy converted to mass.

There are many important nuclear reactions. Some that are of direct concern in reactor operation are listed here:

- $^{1}_{0}n + ^{16}_{8}O \rightarrow ^{16}_{7}N + ^{1}_{1}p$; $\quad ^{16}O(n,p)^{16}N$

  This reaction occurs in LWRs when the oxygen in the light water coolant absorbs a neutron. The product, N-16, has a 7.6 s half-life and emits a very high energy (6-7 MeV) photon. The resulting radiation level makes PWR/BWR primary piping prohibitively radioactive during reactor operation. Also, carryover of the N-16 in the steam from BWRs renders the turbine/condenser radioactive during normal operation. Fortunately, N-16 activity decays away within minutes following a shutdown. (Note: This reaction is termed a "charged-particle" reaction because a proton is emitted. Most such reactions are endothermic because it takes energy to expel the charged particle through the coulomb barrier. This N-16 reaction requires a neutron of 7 MeV or greater. Very few fission neutrons are born with such energies. Nevertheless, N-16 production is a major operational problem in LWRs.

- $^{1}_{0}n + ^{27}_{13}Al \rightarrow ^{4}_{2}He + ^{24}_{11}Na$; $\quad ^{27}Al(n,\alpha)^{24}Na$

  This reaction occurs in research and test reactors which use uranium-aluminide fuel as well as core structural components that are made of aluminum. The sodium activity dissolves in the coolant. Na-24 has a half-life of 11.99 hours.

- $^{1}_{0}n + ^{40}_{18}Ar \rightarrow ^{41}_{18}Ar + ^{0}_{0}\gamma$; $\quad ^{40}Ar(n,\gamma)^{41}Ar$

Air is 1% argon, So, exposure of air to a neutron flux renders it radioactive. This is why research reactors are often surrounded by cover gases (He, CO_2) or why larger reactors have high ventilation rates in the reactor compartments. Ar-41 has a 1.8 hour half-life.

- $^{1}_{0}n + ^{59}_{27}Co \rightarrow ^{60m}_{27}Co + ^{0}_{0}\gamma; \quad ^{59}Co(n,\gamma)^{60m}Co$

  Cobalt is a constituent of many steel alloys. Upon irradiation, Co-60m is created. This decays with a 15.2 minutes half life to Co-60 which has a 5.2 year half life. Cobalt activity is a major source source of dose for maintenance markers in LWRs. For this reason, enormous effort has been expended to develop cobalt-free steel alloys.

Section 9.7 of Turner lists a number of other important reactions. Students should read this material. We note here the major importance of certain of these reactions:

- Dose to Humans from Neutrons: The principal ones are an (n,γ) on hydrogen and an (n,p) on nitrogen-14.

- Neutron Detection: There are three reactions that are used for neutron detection. These are:

  $^{1}_{0}n + ^{3}_{2}He \rightarrow ^{1}_{1}p + ^{3}_{1}H; \quad Q = 765\,keV; \ \sigma = 5330\,barns$

  $^{1}_{0}n + ^{6}_{3}Li \rightarrow ^{3}_{1}H + ^{4}_{2}He; \quad Q = 4.78\,keV; \ \sigma = 940\,barns$

  $^{1}_{0}n + ^{10}_{5}B \rightarrow ^{4}_{2}He + ^{7}_{3}Li; \quad Q = 2.79\,MeV; \ \sigma = 3840\,barns$

  To be useful for detection, a reaction should have a high Q-value (strong signal) and a high probability of interaction (high efficiency for detection). The He-3 reaction offers the latter while the Li-6 reaction offers the former. The compromise is the B-10 reaction which explains its widespread use.

- Boron Reaction: The $^{10}B(n,\alpha)^{7}Li$ reaction is used for neutron detection; neutron absorption in control devices; nuclear medicine (neutron capture therapy); and shielding.

- Neutron Shielding: Neutron shielding in reactors is achieved in two steps. First, the neutrons are elastically scattered off hydrogen-rich substances (e.g., water, concrete, polyethylene) to reduce their energy. Once at low energy, the neutrons are absorbed in boron, cadmium, or lithium-6. The latter has the advantage of not producing a photon. (Note: The maximum neutron energy from fission is about 10 MeV. From accelerators, one can get much higher energies. Shielding of such high energy neutrons is first done by inelastic scatter off iron and then elastic scatter off hydrogen, and finally absorption at thermal energies.)