11

The Origin of Neutron Radiation

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11.1 INTRODUCTION

The nuclear materials that are accounted for in the nuclear fuel cycle emit neutrons as well as gamma rays. For most isotopes the neutron emission rate is very low compared to the gamma-ray emission rate. For other isotopes the neutron emission rate is high enough to provide an easily measurable signal. If the sample of interest is too dense to permit the escape of gamma rays from its interior, then assay by passive neutron detection may be the preferred technique.

Neutrons are emitted from nuclear materials with a wide spectrum of energies. As they travel through matter, they interact and change their energy in a complex manner (see Chapter 12). However, neutron detectors (see Chapter 13) do not usually preserve information about the energy of the detected neutrons. Consequently, neutron assay consists of counting the number of emitted neutrons without knowing their specific energy. (This is in sharp contrast to gamma-ray assay, where gamma rays of discrete energy are emitted by specific radioactive isotopes.) How then can the assayist obtain a neutron signal that is proportional to the quantity of the isotope to be measured?

This chapter describes the production of neutrons by spontaneous fission, by neutron-induced fission, and by reactions with alpha particles or photons. In many cases these processes yield neutrons with unusually low or high emission rates, distinctive time distributions, or markedly different energy spectra. This information can be used to obtain quantitative assays of a particular isotope *if* the sample's isotopic composition is known and only a few isotopes are present.

The discussion of neutron radiation in this chapter emphasizes features that can be exploited by the assayist. Chapters 14 and 15 focus on total neutron counting techniques that exploit high emission rates or unusual energy spectra. Chapters 16 and 17 describe coincidence counting techniques based on neutron time distributions.

11.2 SPONTANEOUS AND INDUCED NUCLEAR FISSION

The spontaneous fission of uranium, plutonium, or other heavy elements is an important source of neutrons. An understanding of this complex process can be aided by visualizing the nucleus as a liquid drop (Figure 11.1). The strong, short-range nuclear forces act like a surface tension to hold the drop together against the electrostatic repulsion of the protons. In the heaviest elements the repulsive forces are so strong that the liquid drop is barely held together. There is a small but finite probability that the drop will deform into two droplets connected by a narrow neck (saddle point). The two

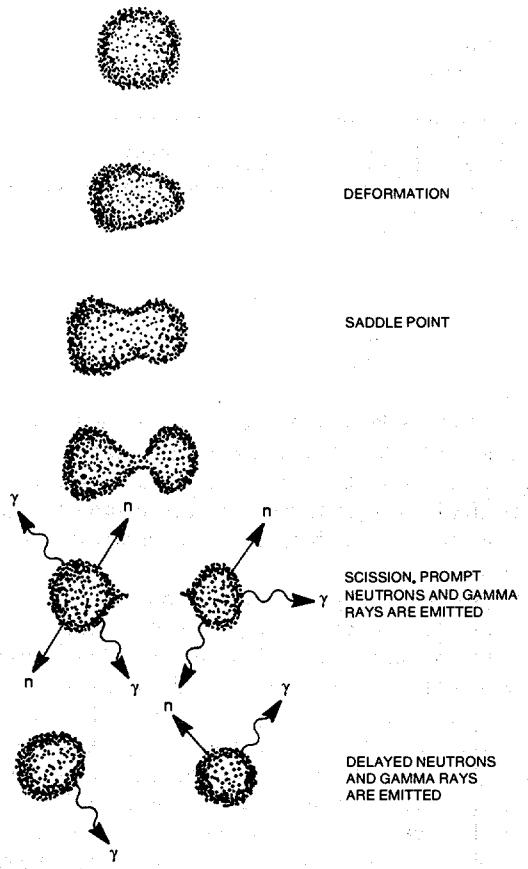

Fig. 11.1 Spontaneous fission of a nucleus represented as the breakup of a liquid drop.

droplets may spontaneously separate (scission) into two fragments. Within 10^{-13} s of scission, each of the two fragments emits a number of prompt neutrons and gamma rays. The fragments are usually unequal in size, with mass distributions centered near atomic numbers 100 and 140 (see Figure 18.2 in Chapter 18). These fission fragments carry away the majority of the energy released in fission (typically 170 MeV) in the form of kinetic energy. Also, within milliseconds or seconds, many of the fragments decay by beta-particle emission into other isotopes that may emit delayed neutrons or gamma rays.

Spontaneous fission is a quantum mechanical process involving penetration of a potential barrier. The height of the barrier, and hence the fission rate, is a very sensitive function of atomic number Z and atomic mass A. The fission yields of some heavy isotopes are summarized in Table 11-1 (Refs. 1 through 6). For thorium, uranium, and plutonium, the fission rate is low compared to the rate of decay by alpha-particle emission, which dominates the total half-life. For californium and even heavier elements, the fission rate can approach the alpha decay rate. The fission yield of ^{240}Pu, 1020 n/s-g (Refs. 4 and 5), is the most important single yield for passive neutron assay because ^{240}Pu is usually the major neutron-emitting plutonium isotope present.

Table 11-1. Spontaneous fission neutron yields

Isotope A	Number of Protons Z	Number of Neutrons N	Total Half-Life[a]	Spontaneous Fission Half-Life[b] (yr)	Spontaneous Fission Yield[b] (n/s-g)	Spontaneous Fission Multiplicity[b,c] ν	Induced Thermal Fission Multiplicity[c] ν
^{232}Th	90	142	1.41×10^{10} yr	$>1 \times 10^{21}$	$>6 \times 10^{-8}$	2.14	1.9
^{232}U	92	140	71.7 yr	8×10^{13}	1.3	1.71	3.13
^{233}U	92	141	1.59×10^{5} yr	1.2×10^{17}	8.6×10^{-4}	1.76	2.4
^{234}U	92	142	2.45×10^{5} yr	2.1×10^{16}	5.02×10^{-3}	1.81	2.4
^{235}U	92	143	7.04×10^{8} yr	3.5×10^{17}	2.99×10^{-4}	1.86	2.41
^{236}U	92	144	2.34×10^{7} yr	1.95×10^{16}	5.49×10^{-3}	1.91	2.2
^{238}U	92	146	4.47×10^{9} yr	8.20×10^{15}	1.36×10^{-2}	2.01	2.3
^{237}Np	93	144	2.14×10^{6} yr	1.0×10^{18}	1.14×10^{-4}	2.05	2.70
^{238}Pu	94	144	87.74 yr	4.77×10^{10}	2.59×10^{3}	2.21	2.9
^{239}Pu	94	145	2.41×10^{4} yr	5.48×10^{15}	2.18×10^{-2}	2.16	2.88
^{240}Pu	94	146	6.56×10^{3} yr	1.16×10^{11}	1.02×10^{3}	2.16	2.8
^{241}Pu	94	147	14.35 yr	(2.5×10^{15})	(5×10^{-2})	2.25	2.8
^{242}Pu	94	148	3.76×10^{5} yr	6.84×10^{10}	1.72×10^{3}	2.15	2.81
^{241}Am	95	146	433.6 yr	1.05×10^{14}	1.18	3.22	3.09
^{242}Cm	96	146	163 days	6.56×10^{6}	2.10×10^{7}	2.54	3.44
^{244}Cm	96	148	18.1 yr	1.35×10^{7}	1.08×10^{7}	2.72	3.46
^{249}Bk	97	152	320 days	1.90×10^{9}	1.0×10^{5}	3.40	3.7
^{252}Cf	98	154	2.646 yr	85.5	2.34×10^{12}	3.757	4.06

[a]Ref. 1.
[b]Ref. 2. Values in parentheses are from Ref. 3 and have estimated accuracies of two orders of magnitude. Pu-240 fission rate is taken from Refs. 4 and 5.
[c]Ref. 6.

The strong dependence of spontaneous fission rates on the number of protons and neutrons is important for assay considerations. The fission rate for odd-even isotopes is typically 10^3 lower than the rate for even-even isotopes, and the fission rate for odd-odd isotopes is typically 10^5 lower. These large differences are due to nuclear spin effects (Ref. 7). As the fissioning nucleus begins to deform, the total ground-state nuclear spin must be conserved. However, the quantized angular momentum orbits of the individual neutrons or protons have different energies with increasing deformation. The lowest energy orbit of the undeformed nucleus may not be the lowest energy orbit in the deformed nucleus. In the case of heavy even-even nuclei, whose total ground-state spin is zero, the outermost pairs of neutrons and protons can simultaneously couple their spins to zero while shifting to the lowest energy orbits. In the case of odd nuclei, a single neutron or proton must occupy the orbit that conserves total nuclear spin even though extra energy is required (Refs. 8 and 9). This effect raises the fission barrier and makes odd-even and odd-odd isotopes more rigid against spontaneous fission than even-even isotopes.

Among the even-even isotopes with high spontaneous fission yields are ^{238}U, ^{238}Pu, ^{240}Pu, ^{242}Pu, ^{242}Cm, ^{244}Cm, and ^{252}Cf. Isotopes with odd neutron numbers or odd proton numbers do not have high spontaneous fission yields, as described above. However, isotopes with odd neutron numbers can easily be induced to fission if bombarded with low-energy neutrons; absorption of an extra neutron yields an unbounded neutron pair whose pairing energy is now available to excite the compound nucleus to an energy near the fission barrier. Among the even-odd isotopes that can be fissioned by neutrons of zero energy but have low spontaneous fission yields are ^{233}U, ^{235}U, and ^{239}Pu. These isotopes are called "fissile." Even-even isotopes, such as ^{238}U and ^{240}Pu, that are not easily fissioned by low-energy neutrons are called "fertile." This term comes from reactor theory and refers to the fact that through neutron capture these isotopes are fertile sources of fissile isotopes. Examples of induced fission cross sections for fertile and fissile isotopes are given in Chapter 12.

11.3 NEUTRONS AND GAMMA RAYS FROM FISSION

Prompt neutrons and gamma rays emitted at the time of scission are the most useful for passive assay because of their intensity and penetrability. Many passive assay instruments, such as coincidence counters, are designed to detect prompt fission neutrons and are often also sensitive to gamma rays. For this reason this section describes both neutron and gamma-ray emissions.

Figure 11.2 shows an energy spectrum of the neutrons emitted during the spontaneous fission of ^{252}Cf (Refs. 10 and 11). The mean energy is 2.14 MeV. The spectrum depends on many variables such as fission fragment excitation energy and average total fission energy release, but can be approximated by a Maxwellian distribution N(E) where N(E) varies as $\sqrt{E}\exp(-E/1.43\ \text{MeV})$. This spectrum is proportional to $\sqrt{E}$ at low energies; it then falls exponentially at high energies. The neutron spectra for spontaneous fission of ^{240}Pu and thermal-neutron induced fission of ^{233}U, ^{235}U, and ^{239}Pu can also be approximated by Maxwellian distributions, with spectrum parameters 1.32, 1.31, 1.29, and 1.33 MeV, respectively (Refs. 12 and 13).

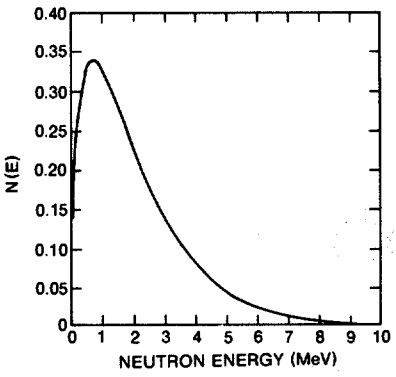

Fig. 11.2 Prompt neutron spectrum from the spontaneous fission of ^{252}Cf, as calculated from a Maxwellian distribution with "temperature" $T = 1.43$ MeV.

The number of neutrons emitted in spontaneous or induced fission is called the neutron multiplicity. Average neutron multiplicities $\bar{\nu}$ are included in the last two columns of Table 11-1. For neutron-induced fission the multiplicity increases slowly and linearly with the energy of the incoming neutron (Ref. 14). The multiplicities given in the last column of Table 11-1 are approximately correct for thermal- or low-energy incident neutrons.

From one fission to another the neutron multiplicity may vary from 0 to 6 or more, depending on the distribution of excitation energy among the fission fragments. Table 11-2 (Refs. 15, 16, and 17) lists the measured prompt neutron multiplicity distributions P(ν) of some important isotopes for spontaneous or thermal-neutron-induced fission. Uncertainties on the individual probabilities vary from 1 to 5% near the maxima to 30 to 50% near the end points. Terrell (Ref. 18) has shown that the multiplicity distributions for both spontaneous and thermal-neutron-induced fission can be approximated by a Gaussian distribution centered at $\bar{\nu}$, the mean multiplicity:

$$P(\nu) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{-(\nu-\bar{\nu})^2/2\sigma^2} \tag{11-1}$$

A distribution width σ of 1.08 can be used as an approximation for all isotopes except ^{252}Cf, where 1.21 should be used.

Information about the neutron multiplicity distribution in fission is used in the analysis of coincidence counting (see Chapter 16). One question that has arisen in this regard is whether the neutron multiplicity and the mean neutron energy are correlated. In other words, if the number of neutrons emitted in a fission is above average, will the mean neutron energy be below average? The available experimental evidence indicates that the mean neutron emission energy is approximately constant and that the number of neutrons emitted increases with the amount of available energy (Ref. 19). Thus the mean energy may be approximately independent of the multiplicity.

After a nucleus undergoes fission, prompt neutrons are evaporated from the fission fragments until the remaining excitation energy is less than the neutron binding energy. At this point, prompt gamma rays carry away the remaining energy and angular momentum. On the average, 7 to 10 prompt gamma rays are emitted, with a total energy of 7 to 9 MeV (Ref. 7). Figure 11.3 shows the prompt gamma-ray spectrum accompanying spontaneous fission of ^{252}Cf, as recorded by a sodium iodide detector (Ref. 20). A

Table 11-2. Measured prompt fission multiplicity distributions

Probability Distribution	^{235}U Induced Fission[a]	^{238}Pu Spontaneous Fission[b,c]	^{239}Pu Induced Fission[a]	^{240}Pu Spontaneous Fission[a]	^{242}Pu Spontaneous Fission[a]	^{252}Cf Spontaneous Fission[a]
P(0)	0.033	0.054	0.011	0.066	0.068	0.002
P(1)	0.174	0.205	0.101	0.232	0.230	0.026
P(2)	0.335	0.380	0.275	0.329	0.334	0.127
P(3)	0.303	0.225	0.324	0.251	0.247	0.273
P(4)	0.123	0.108	0.199	0.102	0.099	0.304
P(5)	0.028	0.028	0.083	0.018	0.018	0.185
P(6)	0.003		0.008	0.002	0.003	0.066
P(7)						0.015
P(8)						0.002
$\overline{\nu}$	2.406	2.21	2.879	2.156	2.145	3.757
$\overline{\nu(\nu-1)}$	4.626	3.957	6.773	3.825	3.794	11.962
$\overline{\nu(\nu-1)(\nu-2)}$	6.862	5.596	12.630	5.336	5.317	31.812

[a]Ref. 15.
[b]Ref. 16.
[c]Ref. 17.

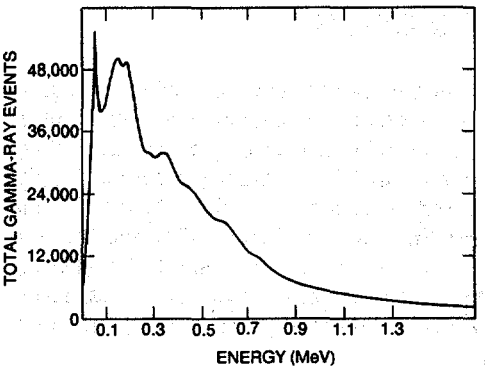

Fig. 11.3 Prompt gamma-ray spectrum accompanying spontaneous fission of ^{252}Cf, as recorded by a sodium iodide detector (Ref. 20).

spectrum obtained with a high-resolution detector might reveal many discrete transitions, although the transitions would be Doppler-broadened by the recoil of the fission fragments. Prompt gamma rays from fission are of much lower intensity than the gamma rays that follow alpha decay (see Chapter 1). Thus they are not useful for passive assay, despite their relatively high energy. However, prompt gamma rays from fission are useful for coincidence counting, where their high multiplicity can lead to a strong signal.

This section includes brief descriptions of the delayed neutrons and gamma rays emitted after fission. In passive assay systems the delayed neutrons and gamma rays are usually masked by the stronger prompt emissions. The time delay, however, is often used by active assay systems to discriminate between the interrogation source and the induced fission signal. For additional details on these delayed signals, see Ref. 21.

Delayed neutrons originate from some of the isotopes produced during beta decay of fission fragments. They are emitted by highly excited isotopes as soon as the isotopes are created by the beta decay of their precursors. Thus, delayed neutrons appear with half-lives characteristic of their precursors. Although there are many such isotopes, delayed neutrons can be categorized into six groups with decay half-lives ranging from 200 ms to 55 s (Ref. 22). The neutron yield of each group is different for each uranium or plutonium isotope. In principle, active assay systems can use this variation as an indication of the isotopic composition of the irradiated sample (Ref. 21), but in practice this use is difficult to implement. Delayed neutron energy spectra are highly structured, as opposed to the smooth Maxwellian distributions of prompt neutrons. Also, the average energy of delayed neutrons is only 300 to 600 keV, as opposed to the 2-MeV average of prompt neutrons. Most important, the number of delayed neutrons is typically only 1% of the number of prompt neutrons. Thus, delayed neutrons contribute to passive neutron measurements, but their effect is not large.

Delayed gamma rays from fission have a higher intensity and slower emission rate than delayed neutrons. Their average multiplicity and energy is comparable to that of prompt gamma rays: 6 to 8 gamma rays, each with an average energy close to 1 MeV. There is no clear-cut distinction between the emission time of prompt and delayed gamma rays as there is for prompt and delayed neutrons. Gozani (Ref. 21) has used a time of 10^{-9} s after fission as a convenient demarcation. The delayed gamma rays so defined are then emitted over times of several seconds or minutes. The intensity of these gamma rays is two orders of magnitude above the intensity of delayed neutrons.

11.4 NEUTRONS FROM (α,n) REACTIONS

Nuclei can decay spontaneously by alpha- or beta-ray emission as well as by fission. Alpha particles are helium nuclei with two protons and two neutrons, and beta particles are energetic free electrons. In principle, all nuclei of atomic mass greater than 150 are unstable towards alpha decay. However, alpha decay is a quantum mechanical barrier penetration process like spontaneous fission, and the Coulomb barrier is high enough to make alpha decay unlikely for all but the heaviest elements. Table 11-3 (Refs. 1, 2, and 23 through 25) lists the alpha decay rates of some heavy elements. The total half-lives of the isotopes listed in the table are almost the same as the alpha decay half-lives, except for ^{241}Pu and ^{249}Bk, where beta decay dominates, and ^{252}Cf, where the spontaneous fission rate is about 3% of the alpha decay rate.

The alpha decay process leads to the emission of gamma rays from unstable daughters (see Chapter 1). Also, the alpha particles can produce neutrons through (α,n) reactions with certain elements. This source of neutrons can be comparable in intensity to spontaneous fission if isotopes with high alpha decay rates such as ^{233}U, ^{234}U, ^{238}Pu, or ^{241}Am are present. This section describes the production of neutrons by (α,n) reactions and provides some guidelines for calculating the expected neutron yield.

Following are two examples of (α,n) reactions that occur in many nuclear fuel cycle materials:

$$\alpha + {}^{18}\mathrm{O} \rightarrow {}^{21}\mathrm{Ne} + \mathrm{n}$$

$$\alpha + {}^{19}\mathrm{F} \rightarrow {}^{22}\mathrm{Na} + \mathrm{n}$$

The alpha particle is emitted from uranium or plutonium with energies in the range of 4 to 6 MeV. Because ^{234}U is the dominant alpha emitter in enriched uranium, the average energy for alpha particles emitted from uranium is 4.7 MeV (see Table 11-3). For plutonium, an average energy of 5.2 MeV is typical. In air, the range of alpha particles from uranium is 3.2 cm and the range of alpha particles from plutonium is 3.7 cm. The range in other materials can be estimated from the Bragg-Kleeman rule (Ref. 26):

$$\text{range} = 0.00032 \frac{\sqrt{A}}{\text{density (g/cm}^3\text{)}} \times \text{range in air} \qquad (11\text{-}2)$$

where A is the atomic weight of the material. The range in uranium and plutonium oxide is roughly 0.006 cm and 0.007 cm, respectively. Thus the alpha particles lose energy very rapidly when traveling through matter. In many cases this short range means that the alpha particle can never reach nearby materials in which (α,n) reactions could take place. If, however, elements such as oxygen or fluorine are intimately mixed with the alpha-emitting nuclear material, an (α,n) reaction may take place because the alpha particle can reach these elements before it loses all its energy.

When the alpha particle arrives at another nucleus, the probability of a reaction depends on the Q-value, the threshold energy, and the height of the Coulomb barrier. The Q-value is the difference in binding energies between the two initial nuclei and the two final reaction products. A positive Q-value means that the reaction will release energy. A negative Q-value means that the alpha particle must have at least that much

Table 11-3. (Alpha,n) reaction neutron yields

Isotope A	Total Half-Life[a]	Alpha Decay Half-Life[a]	Alpha Yield[a] (α/s-g)	Average Alpha Energy[a] (MeV)	(α,n) Yield in Oxide[b] (n/s-g)	(α,n) Yield in UF_6/PuF_4[c] (n/s-g)
^{232}Th	1.41×10^{10} yr	1.41×10^{10} yr	4.1×10^{3}	4.00	2.2×10^{-5}	
^{232}U	71.7 yr	71.7 yr	8.0×10^{11}	5.30	1.49×10^{4}	2.6×10^{6}
^{233}U	1.59×10^{5} yr	1.59×10^{5} yr	3.5×10^{8}	4.82	4.8	7.0×10^{2}
^{234}U	2.45×10^{5} yr	2.45×10^{5} yr	2.3×10^{8}	4.76	3.0	5.8×10^{2}
^{235}U	7.04×10^{8} yr	7.04×10^{8} yr	7.9×10^{4}	4.40	7.1×10^{-4}	0.08
^{236}U	2.34×10^{7} yr	2.34×10^{7} yr	2.3×10^{6}	4.48	2.4×10^{-2}	2.9
^{238}U	4.47×10^{9} yr	4.47×10^{9} yr	1.2×10^{4}	4.19	8.3×10^{-5}	0.028
^{237}Np	2.14×10^{6} yr	2.14×10^{6} yr	2.6×10^{7}	4.77	3.4×10^{-1}	
^{238}Pu	87.74 yr	87.74 yr	6.4×10^{11}	5.49	1.34×10^{4}	2.2×10^{6}
^{239}Pu	2.41×10^{4} yr	2.41×10^{4} yr	2.3×10^{9}	5.15	3.81×10^{1}	5.6×10^{3}
^{240}Pu	6.56×10^{3} yr	6.56×10^{3} yr	8.4×10^{9}	5.15	1.41×10^{2}	2.1×10^{4}
^{241}Pu	14.35 yr	5.90×10^{5} yr	9.4×10^{7}	4.89	1.3	1.7×10^{2}
^{242}Pu	3.76×10^{5} yr	3.76×10^{5} yr	1.4×10^{8}	4.90	2.0	2.7×10^{2}
^{241}Am	433.6 yr	433.6 yr	1.3×10^{11}	5.48	2.69×10^{3}	
^{242}Cm	163 days	163 days	1.2×10^{14}	6.10	3.76×10^{6}	
^{244}Cm	18.1 yr	18.1 yr	3.0×10^{12}	5.80	7.73×10^{4}	
^{249}Bk	320 days	6.1×10^{4} yr	8.8×10^{8}	5.40	1.8×10^{1}	
^{252}Cf	2.646 yr	2.731 yr	1.9×10^{13}	6.11	6.0×10^{5}	

[a]Ref. 1.
[b]Ref. 2.
[c]UF_6, Refs. 23 and 24; PuF_4, Ref. 25.

energy in the center-of-mass reference frame before the reaction can proceed. If this minimum energy requirement is transformed to the laboratory reference frame, it is called the threshold energy:

$$\begin{aligned}&\text{Threshold energy} = -Q(1 + 4/A) \text{ if } Q \text{ is negative}\\&\text{Threshold energy} = 0 \text{ if } Q \text{ is positive.}\end{aligned} \tag{11-3}$$

The Coulomb barrier is the strength of the electrostatic repulsion that the alpha particle must overcome to enter the target nucleus and react.

$$\text{Coulomb barrier (MeV)} = \frac{Z_1 Z_2 e^2}{r_0 (A_1^{1/3} + A_2^{1/3})} \tag{11-4}$$

where $Z_1 = 2$, $A_1 = 4$, $e^2 = 1.44$ MeV-fm, $r_0 = 1.2$ fm, and Z_2 and A_2 refer to the target nucleus (Ref. 27). Thus, an (α,n) reaction is energetically allowed only if the alpha particle has enough energy to (1) overcome or penetrate the Coulomb barrier and (2) exceed the threshold energy. (Note that the two energy requirements are not additive.) Table 11-4 (Refs. 26 and 28) summarizes these properties for a series of low-mass isotopes.

Table 11-4 shows that (α,n) reactions with 5.2-MeV alpha particles are possible in 11 low-Z elements. In all elements with atomic number greater than that of chlorine, the reaction is energetically not allowed. The observed yield of neutrons from (α,n) reactions is given in Table 11-5 (Refs. 29 through 33) for thick targets. A thick target is a material that is much thicker than the range of the alpha particle and one in which the alpha particles lose energy only in the target element. From Equation 11-2, the range of alpha particles in solids is on the order of 0.01 cm.

(Alpha,n) reactions can occur in compounds of uranium or plutonium such as oxides or fluorides and in elements such as magnesium or beryllium that may be present as impurities. The neutron yield per gram of source nuclide in pure oxides and fluorides is given in the last two columns of Table 11-3. In other materials the yield will depend very sensitively on the alpha activity of the nuclear isotopes, the alpha particle energy, the reaction Q-values, the impurity concentrations, and the degree of mixing (because of the short range of the alpha particle).

Equations 11-5 through 11-7 give a prescription for estimating the (α,n) yield in uranium or plutonium oxides with impurities (perfect mixing is assumed). First, the yield in oxide is

$$Y_{\text{oxide}} = \sum_i M_i Y_i \tag{11-5}$$

where M_i is the mass in grams of the ith isotope and Y_i is the neutron yield per gram of each alpha-emitting isotope as given in Table 11-3. The summation over i should include ^{241}Am, which is a strong alpha emitter. The yields for compounds can also be estimated by multiplying the thick target yields in Table 11-5 by the reduction factor K (Refs. 34 and 35):

$$K = \frac{S_t N_t}{S_t N_t + S_\alpha N_\alpha} \tag{11-6}$$

Table 11-4. (Alpha,n) Q-values, threshold energies, and Coulomb barriers

Nucleus	Natural Abundance (%)	Q-Value[a] (MeV)	Threshold Energy[a] (MeV)	Coulomb Barrier (MeV)	Maximum Neutron Energy for 5.2-MeV Alpha[b]
^{4}He	100	−18.99	38.0	1.5	
^{6}Li	7.5	−3.70	6.32	2.1	
^{7}Li	92.5	−2.79	4.38	2.1	1.2
^{9}Be	100	+5.70	0	2.6	10.8
^{10}B	19.8	+1.06	0	3.2	5.9
^{11}B	80.2	+0.16	0	3.2	5.0
^{12}C	98.9	−8.51	11.34	3.7	
^{13}C	1.11	+2.22	0	3.7	7.2
^{14}N	99.6	−4.73	6.09	4.1	
^{15}N	0.4	−6.42	8.13	4.1	
^{16}O	99.8	−12.14	15.2	4.7	
^{17}O	0.04	+0.59	0	4.6	5.5
^{18}O	0.2	−0.70	0.85	4.6	4.2
^{19}F	100	−1.95	2.36	5.1	2.9
^{20}Ne	90.9	−7.22	8.66	5.6	
^{21}Ne	0.3	+2.55	0	5.5	7.6
^{22}Ne	8.8	−0.48	0.57	5.5	4.5
^{23}Na	100	−2.96	3.49	6.0	1.8
^{24}Mg	79.0	−7.19	8.39	6.4	
^{25}Mg	10.0	+2.65	0	6.4	7.7
^{26}Mg	11.0	+0.03	0	6.3	5.0
^{27}Al	100	−2.64	3.03	6.8	2.2
^{29}Si	4.7	−1.53	1.74	7.2	3.4
^{30}Si	3.1	−3.49	3.96	7.2	1.4
^{37}Cl	24.2	−3.87	4.29	8.3	1.0

[a]Ref. 28.
[b]Ref. 26.

where S_t and S_α are the rates at which the alpha particle loses energy in the target material and in the alpha-emitting isotope and N_t and N_α are the atom densities. Some values of the ratio S_α/S_t are given in Ref. 34.

The yield from elements that exist as impurities in the oxide can be estimated by computing the impurity element (α,n) yield relative to the oxide (α,n) yield. The approximation in Equation 11-7 ignores differences in alpha energy between isotopes and differences in target densities that result from the presence of impurities:

$$Y_{impurity} \approx Y_{oxide} \sum_j \frac{P_j A_O I_j S_j}{P_O A_j I_O S_i} \qquad (11\text{-}7)$$

where P_j is the (α,n) neutron yield in the impurity element, from Table 11-5. P_O is the yield in oxygen: 0.059 n/10^6 plutonium alpha particles or 0.040 n/10^6 uranium alpha particles. A_j is the atomic weight of the impurity element, and $A_O = 16$ for oxygen. I_j is

Table 11-5. Thick-target yields from (α,n) reactions (error bars estimated from scatter between references)

Element (Natural Isotopic Composition)	Neutron Yield per 10^6 Alphas of Energy 4.7 MeV (^{234}U)	Neutron Yield per 10^6 Alphas of Energy 5.2 MeV (av. Pu)	References	Av. Neutron Energy (MeV) for 5.2 MeV Alphas (Ref. 29)
Li	0.16 ± 0.04	1.13 ± 0.25	30	0.3
Be	44 ± 4	65 ± 5	31	4.2
B	12.4 ± 0.6	17.5 ± 0.4	29, 30, 33	2.9
C	0.051 ± 0.002	0.078 ± 0.004	29, 30, 31	4.4
O	0.040 ± 0.001	0.059 ± 0.002	29, 30, 31	1.9
F	3.1 ± 0.3	5.9 ± 0.6	29, 30, 33	1.2
Na	0.5 ± 0.5	1.1 ± 0.5	32	
Mg	0.42 ± 0.03	0.89 ± 0.02	29, 30, 31	2.7
Al	0.13 ± 0.01	0.41 ± 0.01	29, 30, 31	1.0
Si	0.028 ± 0.002	0.076 ± 0.003	29, 30, 31	1.2
Cl	0.01 ± 0.01	0.07 ± 0.04	32	

the impurity concentration expressed in parts per million (by weight) of oxide. I_O is the concentration of oxygen expressed in the same way, such as 118 000 ppm for PuO_2 or 154 000 ppm for high-enriched U_3O_8. The total yield from (α,n) reactions is then the sum of Equations 11-5 and 11-7.

The (α,n) yields in Table 11-5 are accurate to 5 to 10% for the best-measured elements. The oxide (α,n) yields in Table 11-3 are known to 10% or better. Thus, the neutron yield calculations are accurate to 10% at best, even with perfect mixing. In moist compounds, liquids, or gases, the estimates given are not valid.

The energy of the neutron emitted in an (α,n) reaction depends on the energy that the alpha particle has at the time of the reaction and on the Q-value of the reaction in the isotope. Average thick-target neutron energies are given in Table 11-5. Maximum neutron energies are given in the last column of Table 11-4. Several spectra are given in Figures 11.4 and 11.5 below.

Another important characteristic of neutrons from (α,n) reactions is that only one neutron is emitted in each reaction. These events constitute a neutron source that is random in time with a multiplicity of 1. This characteristic is exploited by neutron coincidence counters (Chapters 16 and 17), which can distinguish between spontaneous fission neutrons and neutrons from (α,n) reactions.

Note that both (α,n) and (α,p) reactions may leave the nucleus in an excited state, from which the nucleus decays to the ground state by emitting one or more gamma rays. For example:

$$^{19}F(\alpha,n)^{22}Na^* \xrightarrow{\gamma} {}^{22}Na \xrightarrow[2.6\ yr]{\beta^+} {}^{22}Ne^* \xrightarrow{\gamma\,(1275\ keV)} {}^{22}Ne$$

$$^{19}F(\alpha,p)^{22}Ne^* \xrightarrow{\gamma} {}^{22}Ne^* \xrightarrow{\gamma\,(1275\ keV)} {}^{22}Ne\ .$$

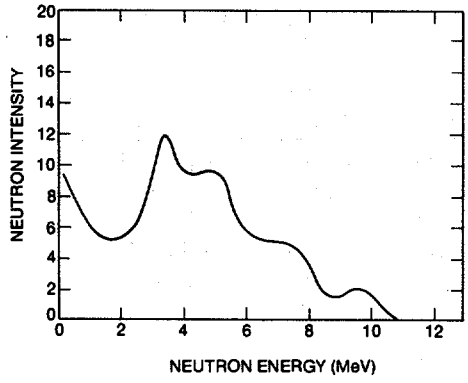

Fig. 11.4 Typical neutron spectrum of an AmBe source.

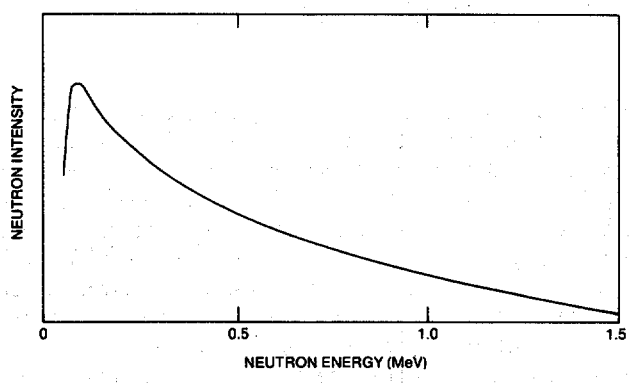

Fig. 11.5 Typical neutron energy spectrum of an AmLi source.

Here the asterisk refers to a nucleus in an excited state. Because many of the gamma rays from these reactions are of high energy and are often emitted nearly simultaneously with the neutron, they can affect the response of total neutron counters or neutron coincidence counters containing detectors that are sensitive to gamma rays.

11.5 NEUTRONS FROM OTHER NUCLEAR REACTIONS

Spontaneous fission, induced fission, and (α,n) reactions are the primary sources of neutrons observed in passive measurements. However, other reactions such as (γ,n), (n,n′), and (n,2n) may take place in the sample or detector assembly and contribute slightly to the observed count rate. This section describes these reactions briefly; examples are given in Table 11-6. These reactions are more important in active nondestructive assay measurements; details can be found in Ref. 21.

Table 11-6. Other nuclear reactions that may affect passive neutron counting

Radiation Source	Threshold Energy[a] (MeV)	Target Material	Reaction	Outgoing Radiation	Outgoing Energy[a]
Gamma	1.665	beryllium	(γ,n)	neutron	8(Eγ−1.665)/9 MeV
Gamma	2.224	deuterium	(γ,n)	neutron	(Eγ−2.224)/2 MeV
Neutron	0	hydrogen	(n,γ)	gamma	2.224 MeV
Proton	1.880	lithium-7	(p,n)	neutron	≥30 keV
Proton	1.019	tritium	(p,n)	neutron	≥0 keV
Neutron	0.1-1.0	lead	(n,n′)	neutron	
Neutron	0.1-1.0	tungsten	(n,n′)	neutron	
Neutron	0.1-1.0	uranium	(n,n′)	neutron	
Neutron	1.851	beryllium	(n,2n)	neutrons	
Neutron	3.338	deuterium	(n,2n)	neutrons	
Neutron	5.340	tungsten	(n,2n)	neutrons	

[a]Ref. 28.

The (γ,n) reaction can produce neutrons in any element if the gamma-ray energy is high enough. The typical minimum threshold energy (~8 MeV) is much higher than the energies of gamma rays emitted from radioactive nuclides. However, the (γ,n) threshold energies for beryllium (1.66 MeV) and deuterium (2.22 MeV) are anomalously low. Thus, it is possible to create a photoneutron source by surrounding relatively intense, long-lived, high-energy gamma-ray sources such as ^{124}Sb or ^{226}Ra with a mantle of beryllium or D_2O. A detailed list of photoneutron sources is given in Table 4.3 of Ref. 21. For passive assay applications it is only necessary to keep in mind that prompt fission gamma rays or gamma rays from some (α,n) reactions can produce extra neutrons if the detector assembly contains beryllium or deuterium. Or, conversely, neutrons can be captured in hydrogen to produce deuterium and 2.22-MeV gamma rays. These possibilities are included in Table 11-6.

Inelastic neutron scattering (n,n′) can occur in heavy nuclei with neutron energies of roughly 0.1 to 1.0 MeV or higher. This reaction is possible if the target nucleus has energy levels low enough to be excited by the neutron. The probability of this reaction is not high, and the number of neutrons present is not altered. However, the average energy of neutrons in the material will decline somewhat faster than would be expected from elastic scattering alone.

The (n,2n) reaction can increase the number of neutrons present, but the threshold energy in most elements is in the range of 10 MeV. For deuterium, beryllium, and tungsten the thresholds are lower, but the number of extra neutrons produced is likely to be small. The possibility of (n,2n) reactions should be considered only when the neutrons are known to have high energy, when deuterium, beryllium, or tungsten are present, and when the coincidence count rates to be measured are very low. In such cases the observed response may be enhanced.

11.6 ISOTOPIC NEUTRON SOURCES

Compact, portable neutron sources are useful for laboratory work, for verifying the proper operation of assay instruments, or for irradiating samples to obtain other induced signals. For accountability or safety purposes it is often important to have sources that contain little or no plutonium or uranium. Such sources can be manufactured by using other isotopes that emit neutrons by spontaneous fission or by taking advantage of (α,n) reactions between strong alpha-particle-emitting isotopes and low-Z materials.

Californium-252 is the most commonly used spontaneous fission neutron source; it can be fabricated in very small sizes and still provide a strong source for a practical period of time. Table 11-7 summarizes some of the properties of ^{252}Cf; Figures 11.2 and 11.3 give the prompt neutron and gamma-ray spectra. For some applications it is important to remember that ^{252}Cf neutrons are emitted with an average multiplicity of 3.757. Thus they are strongly correlated in time and will generate coincidence events.

Sources that emit random, uncorrelated neutrons can be manufactured by mixing alpha emitters such as ^{238}Pu or ^{241}Am with beryllium, lithium, fluorine, or other elements in which (α,n) reactions are possible. Table 11-8 (Refs. 1, 26, and 36) summarizes the characteristics of some common (α,n) sources. One important feature for practical applications is the half-life of the heavy element that emits the alpha particles. The sources listed in Table 11-8 all have long half-lives, with the exception of $^{210}PoBe$. Another important feature is the neutron energy spectrum obtained from the source. In some cases it is important to have a high-energy, highly penetrating neutron source. In other cases it may be important to avoid neutron energies high enough to fission plutonium or uranium isotopes (that is, the source must provide subthreshold interrogation) or high enough to excite (n,2n) reactions.

Table 11-7. Characteristics of ^{252}Cf

Total half-life	2.646 yr
Alpha half-life	2.731 yr
Spontaneous fission half-life	85.5 yr
Neutron yield	2.34×10^{12} n/s-g
Gamma-ray yield	1.3×10^{13} γ/s-g
Alpha-particle yield	1.9×10^{13} α/s-g
Average neutron energy	2.14 MeV
Average gamma-ray energy	1 MeV
Average alpha-particle energy	6.11 MeV
Neutron activity	4.4×10^{9} n/s-Ci
Neutron dose rate	2300 rem/h-g at 1 m
Gamma dose rate	140 rem/h-g at 1 m
Conversion	558 Ci/g
Decay heat	38.5 W/g
Avg. spontaneous fission neutron multiplicity	3.757
Avg. spontaneous fission gamma multiplicity	8

Table 11-8. Characteristics of some isotopic (α,n) sources

Source	Half-Life[a] (yr)	Average Alpha Energy[a] (MeV)	Average Neutron Energy[a] (MeV)	Maximum Neutron Energy[b] (MeV)	Gamma Dose in mrem/h at 1 m/(10^6 n/s)[c]	Curies per Gram[d]	Yield in 10^6 n/s-Ci[c]
^{210}PoBe	0.38	5.3	4.2	10.9	0.01	4490	2-3
^{226}RaBe	1600	4.8	4.3	10.4	60	1	0-17
^{238}PuBe	87.74	5.49	4.5	11.0	0.006	17	2-4
^{238}PuLi	87.74	5.49	0.7	1.5	~1	17	0.07
$^{238}PuF_4$	87.74	5.49	1.3	3.2	~1	17	0.4
$^{238}PuO_2$	87.74	5.49	2.0	5.8	~1	17	0.003
^{239}PuBe	24 120.	5.15	4.5	10.7	6	0.06	1-2
$^{239}PuF_4$	24 120.	5.15	1.4	2.8	~1	0.06	0.2
^{241}AmBe	433.6	5.48	5.0	11.0	6	3.5	2-3
^{241}AmLi	433.6	5.48	0.3	1.5	2.5	3.5	0.06
^{241}AmB	433.6	5.48	2.8	5.0		3.5	
^{241}AmF	433.6	5.48	1.3	2.5		3.5	

[a]Ref. 1.
[b]Ref. 26.
[c]Ref. 36.
[d](Alpha yield/s-g)/(3.7×10^{10} dps/Ci).

Two common (α,n) sources in use today are ^{241}AmBe and ^{241}AmLi. Typical neutron energy spectra for these two sources are given in Figures 11.4 and 11.5. The energy spectra can vary somewhat because of impurity elements or imperfect mixing. [Also, (α,n) spectra can change their shape somewhat in time, depending on the source construction and the particular isotopes involved.] Note that AmLi sources are usually fabricated by mixing $^{241}AmO_2$ with lithium oxide and that (α,n) reactions in the oxide contribute a high-energy tail to the spectrum.

The ^{241}AmBe sources are compact and relatively inexpensive and do not require much gamma-ray shielding. However, the high-energy spectrum permits (n,2n) reactions that will produce coincidence counts. The ^{241}AmLi sources are less compact and more expensive and require tungsten shields. Because of their low-energy neutron spectra, they are the most widely used sources for subthreshold interrogation in active assay and for random-neutron check sources in passive coincidence counting. For the latter application it is important to be aware of the possibility of plutonium contamination in the americium, which can yield spurious coincidence counts from spontaneous fission.

(Alpha,n) sources also emit gamma and beta radiation, and in many cases the dose observed outside the container is dominated by gamma radiation. (For comparison, the dose from 10^6 n/s is about 1 mrem/h at 1 m.) The neutron yield of an (α,n) source relative to its total radiation output in curies may thus be an important selection criterion. This ratio is given in the last column of Table 11-8. Because of their high gamma-ray output, some (α,n) sources should be encapsulated in shielding material. For example, ^{241}AmLi sources are enclosed in 1/4- to 3/8-in.-thick tungsten to shield against the intense 60-keV gamma rays from americium decay.

11.7 CONCLUSIONS

What properties of neutron radiation can be used by the assayist to measure the quantity of specific isotopes? Several important features are summarized below:

1. The odd-even effect in spontaneous fission means that only fertile isotopes like ^{238}U, ^{238}Pu, ^{240}Pu, and ^{242}Pu are strong emitters of high-energy (2-MeV average) neutrons. For metallic samples of plutonium the total neutron emission rate is usually directly related to the masses of the even isotopes that are present. This is also true for metallic uranium, although kilogram quantities are required for practical assays because of the lower neutron emission rate.
2. The prompt neutron multiplicity ($\nu = 2$ to 3) means that coincidence counting techniques can provide a nearly unique signature for the presence of the even isotopes. However, the multiplicity does not vary enough from one isotope to another to permit discrimination between them.
3. The detection of prompt fission gamma rays along with the neutrons can greatly enhance instrument sensitivity. However, the different behavior of neutrons and gamma rays in the sample matrix and in the detector increases the difficulty of relating the measured response to the sample mass. Therefore, this approach is not recommended for most applications. The use of prompt gamma rays alone is an almost untouched field, but relating measured response to sample mass is again likely to be a difficult problem.

4. Delayed neutron yields are too low for passive assay. Delayed gamma rays are usually not detected by neutron detectors, but they contribute to the response of scintillators. Both delayed neutrons and delayed gamma rays are very important for active assay but not for passive assay.
5. Fissile isotopes like ^{235}U and ^{239}Pu are assayed either by active techniques or indirectly by passive assay of adjacent fertile isotopes if the isotopic composition of the sample is known.
6. (Alpha,n) reactions allow good passive assays of compounds such as $^{238}PuO_2$ and $^{234}UF_6$. Again, the quantity of other isotopes can be inferred from the known isotopic composition. (Alpha,n) reactions can also yield unwanted passive emissions that complicate the assay. Neutron coincidence counting is often used to discriminate against (α,n) reactions.

The principles and applications of these techniques are described in Chapters 14 through 17.

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