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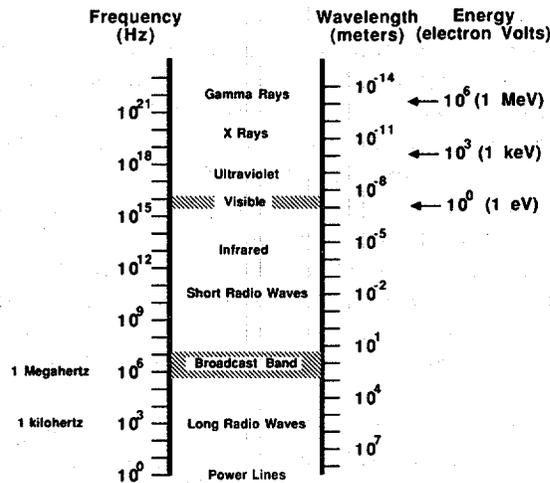
## The Origin of Gamma Rays

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### 1.1 GAMMA RAYS AND THE ELECTROMAGNETIC SPECTRUM

Gamma rays are high-energy electromagnetic radiation emitted in the deexcitation of the atomic nucleus. Electromagnetic radiation includes such diverse phenomena as radio, television, microwaves, infrared radiation, light, ultraviolet radiation, x rays, and gamma rays. These radiations all propagate through vacuum with the speed of light. They can be described as wave phenomena involving electric and magnetic field oscillations analogous to mechanical oscillations such as water waves or sound. They differ from each other only in the frequency of oscillation. Although given different names, electromagnetic radiation actually forms a continuous spectrum, from low-frequency radio waves at a few cycles per second to gamma rays at  $10^{18}$  Hz and above (see Figure 1.1). The parameters used to describe an electromagnetic wave—frequency, wavelength, and energy—are related and may be used interchangeably. It is common practice to use frequency or wavelength for radio waves, color or wavelength for light waves (including infrared and ultraviolet), and energy for x rays and gamma rays.



**Fig. 1.1** The electromagnetic spectrum showing the relationship between gamma rays, x rays, light waves, and radio waves.

Visible light is emitted during changes in the chemical state of elements and compounds. These changes usually involve the outermost and least tightly bound atomic electrons. The colors of the emitted light are characteristic of the radiating elements and compounds and typically have energies of about 1 eV.\* X rays and gamma rays are very high energy light with overlapping energy ranges of 10 keV and above. X rays are emitted during changes in the state of inner or more tightly bound electrons, whereas gamma rays are emitted during changes in the state of nuclei. The energies of the emitted radiations are characteristic of the radiating elements and nuclides.

Knowledge of these high-energy electromagnetic radiations began in Germany in 1895 with the discovery of x rays by Wilhelm Roentgen. After observing that a zinc sulfide screen glowed when it was placed near a cathode-ray discharge tube, Roentgen found that the radiation that caused the glow was dependent on the electrode materials and the tube voltage, that it was not bent by electric or magnetic fields, and that it could readily penetrate dense matter. Natural radioactivity was discovered the following year in France by Henri Becquerel, who observed that uranium salts gave off a natural radiation that could expose or blacken a photographic plate. While studying these phenomena, Marie and Pierre Curie isolated and identified the radioactive elements polonium and radium. They determined that the phenomena were characteristic of the element, not its chemical form.

These "radioactive rays" were intensely studied in many laboratories. In 1899 in England, Ernest Rutherford discovered that 95% of the radiation was effectively stopped by 0.02 mm of aluminum and 50% of the remaining radiation was stopped by 5 mm of aluminum or 1.5 mm of copper. Rutherford named the first component "alpha" and the second, more penetrating radiation, "beta." Both of these radiations were deflected by electric and magnetic fields, though in opposite directions; this fact indicated that the radiations carried electrical charge. In 1900 Paul Villard and Henri Becquerel noted that a photographic plate was affected by radioactive materials even when the plate was shielded by 20 cm of iron or 2 to 3 cm of lead. They also noted that this penetrating radiation showed no magnetic deflection. In 1903 Rutherford named this component "gamma" and stated that "gamma rays are probably like Roentgen rays." Thus the three major radiations were identified and named for the first three letters of the Greek alphabet:  $\alpha$ ,  $\beta$  and  $\gamma$ .

As indicated by the brief description of their discovery, gamma rays often accompany the spontaneous alpha or beta decay of unstable nuclei. X rays are identical to gamma rays except that they are emitted during rearrangement of the atomic electron structure rather than changes in nuclear structure. X-ray energies are unique to each element but the same for different isotopes of one element. They frequently accompany nuclear decay processes, which disrupt the atomic electron shell.

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\* The electron volt (eV) is a unit of energy equal to the kinetic energy gained by an electron accelerated through a potential difference of 1 V; 1 eV equals  $1.602 \times 10^{-19}$  J. This small unit and the multiple units keV ( $10^3$  eV) and MeV ( $10^6$  eV) are useful for describing atomic and molecular phenomena.

Gamma rays from spontaneous nuclear decay are emitted with a rate and energy (color) spectrum that is unique to the nuclear species that is decaying. This uniqueness provides the basis for most gamma-ray assay techniques: by counting the number of gamma rays emitted with a specific energy, it is possible to determine the number of nuclei that emit that characteristic radiation.

## 1.2 CHARACTERISTICS OF NUCLEAR DECAY

### 1.2.1 Nuclear Decay Processes: General

The atomic nucleus is assumed to be a bound configuration of protons and neutrons. Protons and neutrons have nearly the same mass and differ principally in charge: protons have a positive charge of 1 and neutrons are electrically neutral. Different elements have nuclei with different numbers of neutrons and protons. The number of protons in the nucleus is called the atomic number and given the symbol  $Z$ . In the neutral atom, the number of protons is equal to the number of electrons. The number of neutrons in the nucleus is given the symbol  $N$ . The total number of nucleons (protons and neutrons) in the nucleus is called the atomic mass number and given the symbol  $A$  ( $A = Z + N$ ).

For all nuclear decay processes, the number of unstable nuclei of a given species is found to diminish exponentially with time:

$$n = n_0 e^{-\lambda t} \quad (1-1)$$

where  $n$  = number of nuclei of a given species at time  $t$

$n_0$  = number of nuclei at  $t = 0$

$\lambda$  = decay constant, the parameter characterizing the exponential.

Each nuclear species has a characteristic decay constant. Radioactive decay is most commonly discussed in terms of the nuclear half-life,  $T_{1/2}$ , which is related to the decay constant by

$$T_{1/2} = (\ln 2) / \lambda \quad (1-2)$$

The half-life is the time necessary for the number of unstable nuclei of one species to diminish by one-half. (Half-lives are commonly tabulated in nuclear data tables). The decay rate or specific activity can be represented in terms of the half-life as follows:

$$R = \frac{1.32 \times 10^{16}}{A T_{1/2}} \quad (1-3)$$

where  $R$  = rate in decays per second per gram

$A$  = atomic weight

$T_{1/2}$  = half-life in years.

Equation 1-3 is often used to estimate the activity per gram of a sample.

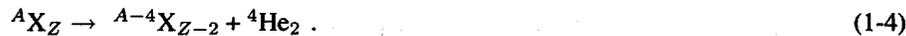
An alpha or beta decay of a given nuclear species is not always accompanied by gamma-ray emission. The fraction of decays that is accompanied by the emission of a specific energy gamma ray is called the branching intensity. For example, the most intense gamma ray emitted by  $^{235}\text{U}$  has an energy of 185.7 keV and a branching intensity of 54%. Uranium-235 decays by alpha-particle emission with a half-life of  $7.038 \times 10^8$  yr. Equation 1-3 thus implies an alpha emission rate of  $7.98 \times 10^4$ /g-s. Only 54% of the alpha particles are accompanied by a 185.7-keV gamma ray; therefore, the specific activity of this gamma ray is  $4.3 \times 10^4$ /g-s.

Of the natural decay radiations only the gamma ray is of interest for nondestructive assay of bulk nuclear materials because the alpha- and beta-particle ranges are very short in condensed matter. Consider the following ranges in copper metal:

$$\begin{aligned} 5\text{-MeV } \alpha: & 0.01 \text{ mm or } 0.008 \text{ g/cm}^2 \\ 1\text{-MeV } \beta: & 0.7 \text{ mm or } 0.6 \text{ g/cm}^2 \\ 0.4\text{-MeV } \gamma: & 12 \text{ mm or } 10.9 \text{ g/cm}^2(\text{mean free path}). \end{aligned}$$

### 1.2.2 Alpha Decay

The alpha particle is a doubly ionized (bare)  $^4\text{He}_2$  nucleus. It is a very stable, tightly bound nuclear configuration. When a nucleus decays by alpha emission, the resulting daughter nucleus has a charge that is two units less than the parent nucleus and an atomic mass that is four units less. This generic reaction can be represented as follows:



The decay can occur only if the mass of the neutral parent atom is greater than the sum of the masses of the daughter and the neutral  $^4\text{He}$  atom. The mass difference between the parent and the decay products is called the Q-value and is equal to the kinetic energy of the decay products:

$$Q = (M_p - M_d - M_{He})c^2 \quad (1-5)$$

where  $M_{p,d,He}$  = neutral atomic mass of the parent, daughter, and  $^4\text{He}$  atom  
 $c$  = velocity of light.

When the parent nucleus decays, most of the energy Q goes to the alpha particle because of its lower mass:

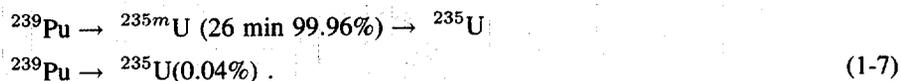
$$E_\alpha = Q(A - 4)/A \quad (1-6)$$

The remainder of the available energy goes into the recoil of the daughter nucleus.

Most of the approximately 160 known alpha emitters are heavy nuclei with atomic numbers greater than 82. The energy of the emitted alpha particle ranges from 4 to 10 MeV and the half-lives vary from  $10^{-6}$  s to  $10^{10}$  yr. The short-lived nuclei emit high-energy alpha particles when they decay.

Immediately after the decay of the parent nucleus, the daughter nucleus can be either in the ground state or in an excited state. In the latter case the nucleus can relax by either of two mechanisms: gamma-ray emission or internal conversion. The radiative relaxation leads to emission of one or more gamma rays (typically,  $10^{-14}$  s after the alpha emission) with discrete energies whose sum equals the original excitation energy. During internal conversion the nucleus transfers the excitation energy directly to one of the most tightly bound atomic electrons, usually a K electron. The electron leaves the atom with an energy equal to the difference of the transition energy and the electron binding energy. The resulting vacancy leads to the emission of x rays or electrons (called Auger electrons) with the characteristic energy spectrum of the daughter element. The probability of internal conversion increases strongly with atomic number ( $Z$ ) and with decreasing excitation energy.

In some cases the alpha decay leads to an excited state that lives much longer than  $10^{-14}$  s. If the lifetime of this state is longer than approximately  $10^{-6}$  s, it is called an isomer of the ground-state nucleus. An example of an isomer is the alpha decay of  $^{239}\text{Pu}$  that leads to  $^{235}\text{U}$ :



The common decay mode of  $^{239}\text{Pu}$  leads first to the isomer  $^{235m}\text{U}$ , which has a half-life of 26 min. The direct decay to  $^{235}\text{U}$  occurs only 0.04% of the time. One of the longest lived isomers is  $^{91m}\text{Nb}_{41}$ , with a half-life of 60 days.

All the alpha particles, gamma rays, and internal conversion electrons emitted during the decay process have discrete, characteristic energies. The observation of these characteristic spectra showed that nuclei have discrete allowed states or energy levels analogous to the allowed states of atomic electrons. The various spectroscopic observations have provided information for developing the nuclear level schemes presented in handbooks such as the *Table of Isotopes* (Ref. 1). An example appears in Figure 1.2 showing the lower energy levels of  $^{235}\text{U}$  populated during the alpha decay of  $^{239}\text{Pu}$ . These levels give rise to the characteristic gamma-ray spectrum of  $^{239}\text{Pu}$ . Note that the characteristic gamma-ray spectrum is commonly associated with the parent or decaying nucleus even though the energies are determined by the levels of the daughter nucleus. Although this practice may seem confusing, it is universally followed for gamma rays. The confusion is further aggravated by the common use of x-ray nomenclature that associates the characteristic x rays with the daughter element. Hence the alpha decay of  $^{239}\text{Pu}$  leads to  $^{235}\text{U}$  and is accompanied by the emission of  $^{239}\text{Pu}$  gamma rays and uranium x rays.

### 1.2.3 Beta Decay

In the beta decay process the atomic number ( $Z$ ) increases or decreases by one unit and the atomic mass number ( $A$ ) stays constant. In effect, neutrons and protons change state. The three types of beta decay are  $\beta^-$ ,  $\beta^+$ , and electron capture.

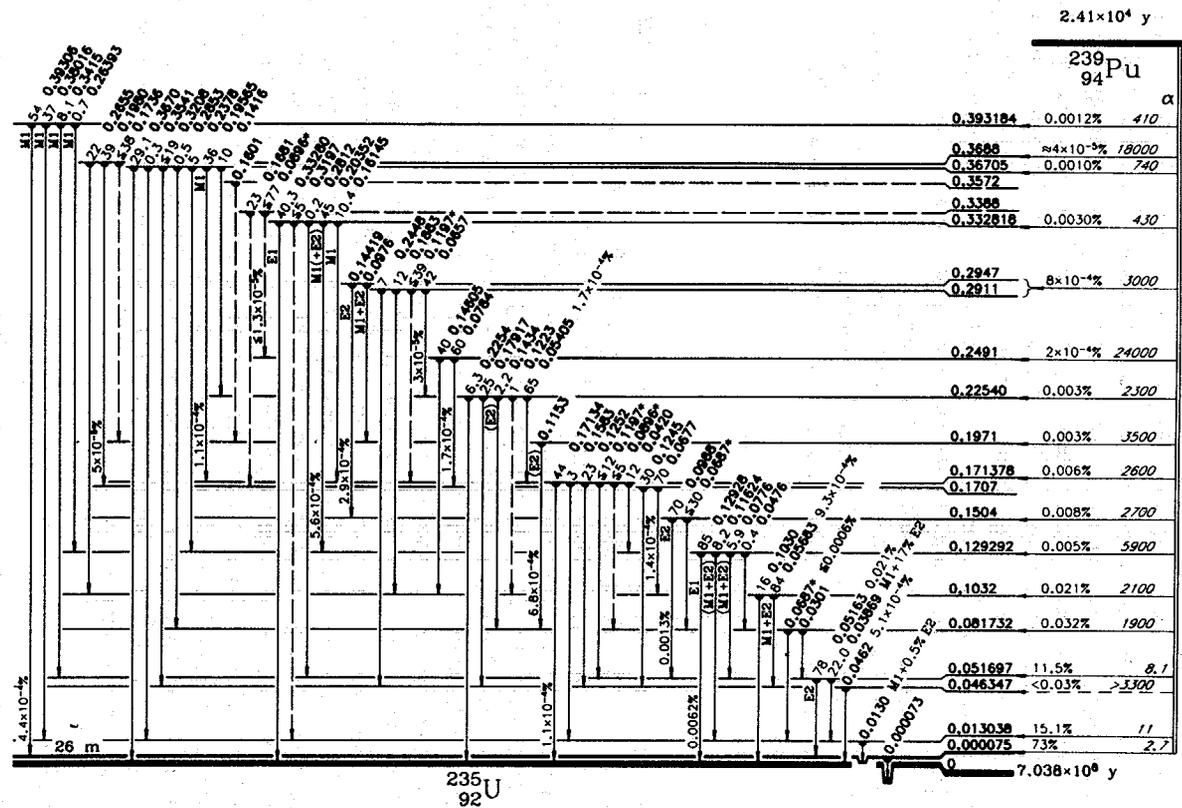


Fig. 1.2 Diagram of some of the nuclear energy levels of  $^{235}\text{U}$ . These levels are populated during the decay of  $^{239}\text{Pu}$  and give rise to the characteristic gamma-ray spectrum of  $^{239}\text{Pu}$ . Figure adapted from Ref. 1.

Beta-minus decay was the first detected process; the  $\beta^-$  particle was found to be a normal electron. During the decay process the nucleus changes state according to the following formula:



The  $\beta^-$  decay process can be thought of as the decay of a neutron into a proton, an electron, and an electron antineutrino. The decay is energetically possible for a free neutron and occurs with a half-life of 12.8 min. This is the common beta-decay process for nuclei with high atomic number and for fission-product nuclei, which usually have significantly more neutrons than protons.

During  $\beta^+$  decay the nucleus changes state according to the following formula:



Electron capture competes with the  $\beta^+$  decay process. The nucleus interacts with an inner atomic electron and, in effect, captures it, changing a proton into a neutron with the emission of a positron and an electron neutrino. The formula for this process is



All unstable nuclei with atomic number less than 82 decay by at least one of the three processes and sometimes by all three (see Figure 1.3). Beta decay occurs whenever it is energetically possible. It is energetically possible if the following conditions are met for the masses of the neutral parent atoms (p) and the potential daughter atom (d):

$$\beta^- \text{ decay: } M_p > M_d$$

$$\beta^+ \text{ decay: } M_p > M_d + 2m_e$$

$$\text{Electron capture: } M_p > M_d . \quad (1-11)$$

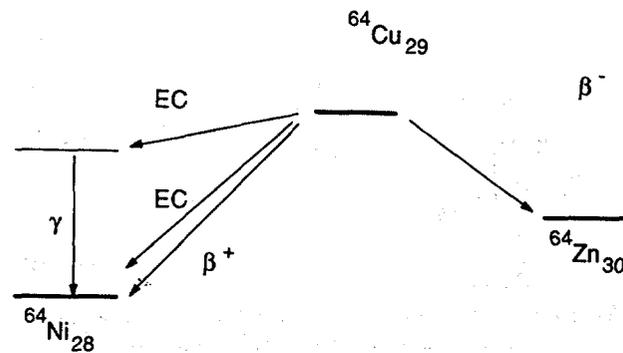


Fig. 1.3 Nuclear decay scheme of  ${}^{64}\text{Cu}$  showing three possible beta decay processes.

Beta decay can be to the ground state or to an excited state in the daughter nucleus. In the latter case the excited state decays by gamma-ray emission or internal conversion.

### 1.3 X-RAY PRODUCTION

#### 1.3.1 The Bohr Model of the Atom

In the simple Bohr model of the atom, the positive nucleus contains protons and neutrons and has an approximate radius of  $1.4 \times 10^{-15} (A^{1/3})\text{m}$  and an approximate density of  $2 \times 10^{14} \text{g/cm}^3$ . The nucleus is surrounded by a cloud of negative electrons in discrete, well-defined energy levels or orbitals. The radii of these orbitals are in the range  $10^{-9}$  to  $10^{-8}$  m. The original Bohr model had well-localized orbits and led to the familiar planetary diagram of the atom. Although not accepted at present, this concrete model is useful for explaining x-ray production.

The different energy levels are designated K,  $L_1$ ,  $L_2$ ,  $L_3$ ,  $M_1$ , ...,  $M_5$ , and so forth. (As an example, consider the electron energy levels of uranium illustrated in Figure 1.4.) The electric force between an electron and the positively charged nucleus varies as the inverse square of the separation; therefore, the electrons closer to the nucleus have a higher binding energy B. The binding energy is the energy required to remove the electron from the atom. The K-shell electrons are always the most tightly bound. Quantum mechanics gives a good description of the energies of each level and how the levels fill up for different elements. The chemical properties of the elements are determined by the electron configuration.

In its normal resting configuration the atom is stable and does not radiate. If an electron moves from a higher to a lower energy level, it radiates an x ray.

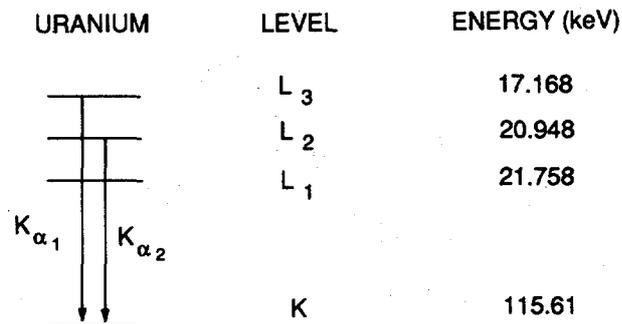


Fig. 1.4 Electron energy levels in uranium. Transitions between the levels shown give rise to the K-series x rays.

### 1.3.2 X-Ray Production Mechanisms

Various interactions ionize or remove an electron from an atom. All energetic, charged particles interact with electrons as they pass through matter. X-ray and gamma-ray photons also interact with atomic electrons. Nuclear interactions such as internal conversion or electron capture can cause the ionization of atomic electrons.

When an electron leaves an atom, the atom is in an excited state with energy  $B_i$  by virtue of the vacancy in the  $i^{\text{th}}$  electron level. This vacancy may be filled by a more loosely bound electron from an outer orbital, the  $j^{\text{th}}$  level. The change in energy level is accompanied by the emission of an x ray with energy  $B_i - B_j$  or by the emission of an Auger electron with energy  $B_i - 2B_j$ . In the latter case the atom transfers its excess energy directly to an electron in an outer orbital. The fraction of vacancies in level  $i$  that result in x-ray emission is defined as the fluorescence yield  $W_i$ . Figure 1.5 shows the variation of the K-shell fluorescence yield with atomic number. X-ray emission is more probable for high-Z elements (for  $Z > 70$ ,  $W_K > 95\%$ ).

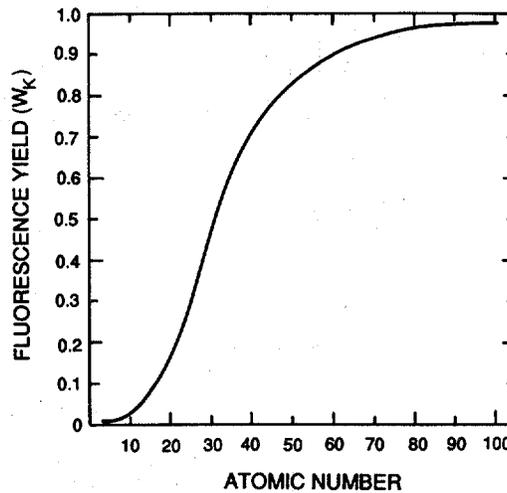


Fig. 1.5 Variation of the K-shell fluorescence yield,  $W_K$ , with atomic number.

High-Z materials have high internal conversion coefficients, which means that their normal decay modes lead to vacancies in an inner electron shell (usually K or L) and the production of characteristic x rays. Because these vacancies occur in the daughter atom, the x-ray energies are characteristic of the daughter element. In condensed materials the charged particles ( $\alpha$ ,  $\beta$ ) and gamma rays from the nuclear decay are stopped in the parent material by a series of interactions with atomic electrons; this leads to the production of x rays that are characteristic of the parent atom. Plutonium metal emits uranium x rays by virtue of the internal conversion process that occurs after alpha decay. It also emits plutonium x rays by virtue of alpha-particle-induced x-ray fluorescence.

### 1.3.3 Characteristic X-Ray Spectra

Each element emits a characteristic x-ray spectrum. All elements have the same x-ray pattern, but the x-ray energies are different. Figure 1.6 shows the characteristic x rays from uranium and lead.

Early investigators developed the system commonly used today for naming x rays. A roman letter indicates the final level to which the electron moves, and a Greek letter plus a number indicates the electron's initial energy level. (The Greek letter was originally related to the x-ray energy and the number to its intensity). Table 1-1 gives the major K x rays of uranium and plutonium. The L and M x rays are of lower energy and are tabulated in the literature.

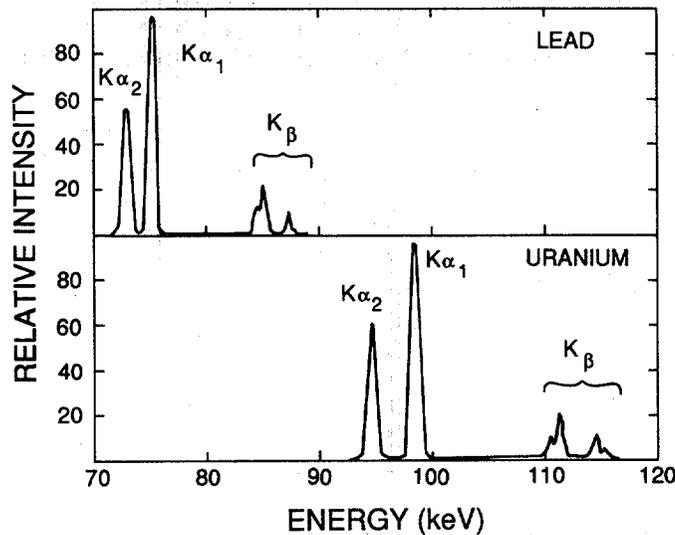


Fig. 1.6 Characteristic x-ray spectra from lead and uranium. Note that the pattern is the same but shifted in energy.

## 1.4 MAJOR GAMMA RAYS FROM NUCLEAR MATERIAL

### 1.4.1 Typical Spectra

Figures 1.7 through 1.12 show typical uranium, plutonium, and thorium gamma-ray spectra. The spectra were measured with high-resolution germanium detector systems. Figure 1.7 shows the spectrum of highly enriched uranium from 0 to 3 MeV, with

Table 1-1. Major K x rays of uranium and plutonium<sup>a</sup>

X Ray	Levels (Final - Initial)	Energy (keV)		Intensity <sup>b</sup>	
		Uranium	Plutonium	Uranium	Plutonium
K <sub>α2</sub>	K - L <sub>2</sub>	94.67	99.55	61.9	62.5
K <sub>α1</sub>	K - L <sub>3</sub>	98.44	103.76	100	100
K <sub>β1</sub>	K - M <sub>3</sub>	111.30	117.26	22.0	22.2
K <sub>β2</sub>	K - N <sub>2-5</sub>	114.5	120.6	12.3	12.5
K <sub>β3</sub>	K - M <sub>2</sub>	110.41	116.27	11.6	11.7

<sup>a</sup>Other x rays in the K series are weaker than those listed here. The energies and intensities are from Ref. 1.

<sup>b</sup>Relative intensity, 100 is maximum.

characteristic gamma rays from <sup>235</sup>U and the <sup>238</sup>U daughter <sup>234m</sup>Pa. The intense gamma rays between 140 and 210 keV are often used for the assay of <sup>235</sup>U (Figure 1.8 shows this region in more detail). For comparison, Figure 1.9 shows a spectrum of depleted uranium; the spectrum shows the <sup>238</sup>U daughter radiations often used for <sup>238</sup>U assay. Figures 1.10 and 1.11 are gamma-ray spectra of plutonium with approximate <sup>240</sup>Pu concentrations of 14% and 6%, respectively. Note the differences in relative peak heights between the two spectra; these differences are used to determine the plutonium isotopic composition (see discussion in Chapter 8). Figure 1.12 shows the characteristic gamma-ray spectrum of <sup>232</sup>Th; all major thorium gamma rays come from daughter nuclides.

#### 1.4.2 Major Gamma-Ray Signatures for Nuclear Material Assay

In principle, any of the gamma rays from nuclear material can be used to determine the mass of the isotope that produces them. In practice, certain gamma rays are used more frequently than others because of their intensity, penetrability, and freedom from interference. The ideal signature would be an intense ( $>10^4$   $\gamma/g\cdot s$ ) gamma ray with an energy of several million electron volts. The mass attenuation coefficients of all materials show a broad minimum between 1 to 5 MeV and there are very few natural gamma rays above 1 MeV that can cause interference. Unfortunately, such gamma rays do not exist for uranium or plutonium.

Table 1-2 lists the gamma rays most commonly used for the nondestructive analysis of the major uranium and plutonium isotopes.

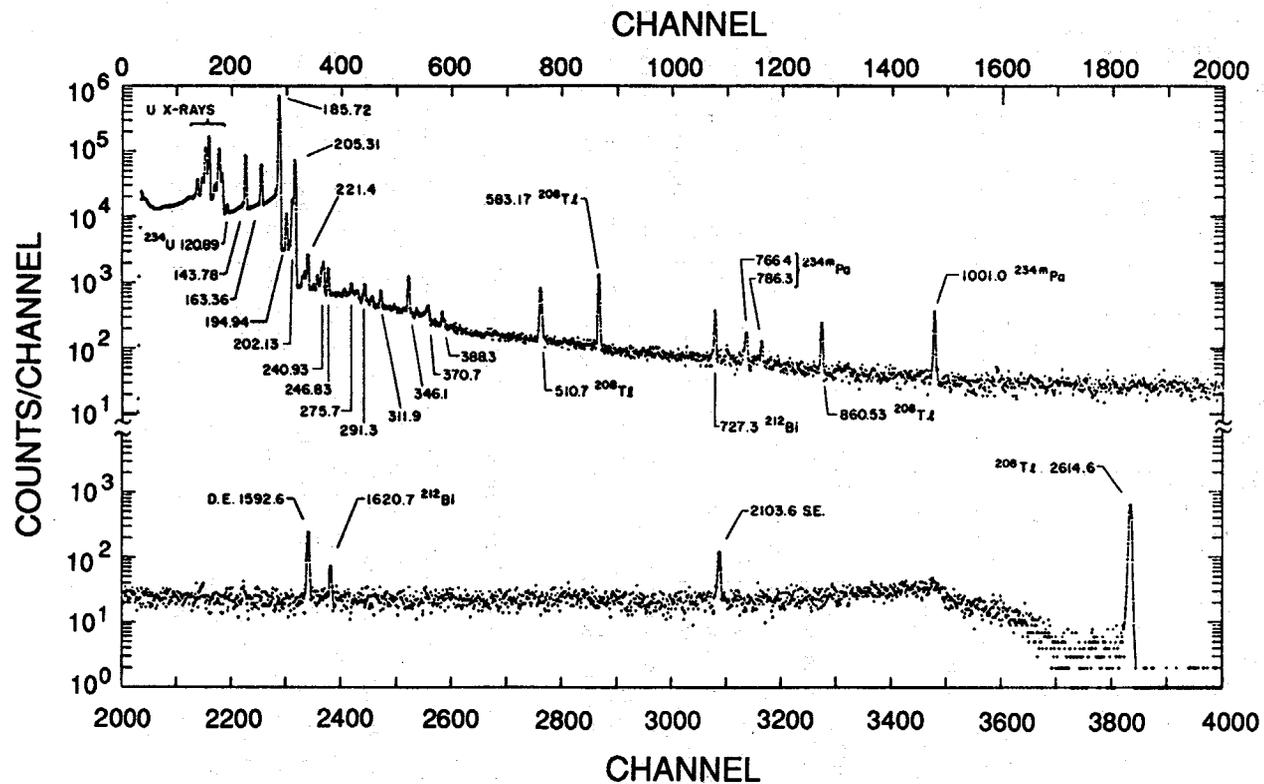


Fig. 1.7 High-resolution gamma-ray spectrum of highly enriched uranium (93%  $^{235}\text{U}$ ). Energies are given in kiloelectron volts. (S.E. and D.E. are the single- and double-escape peaks of the 2614-keV gamma ray.)

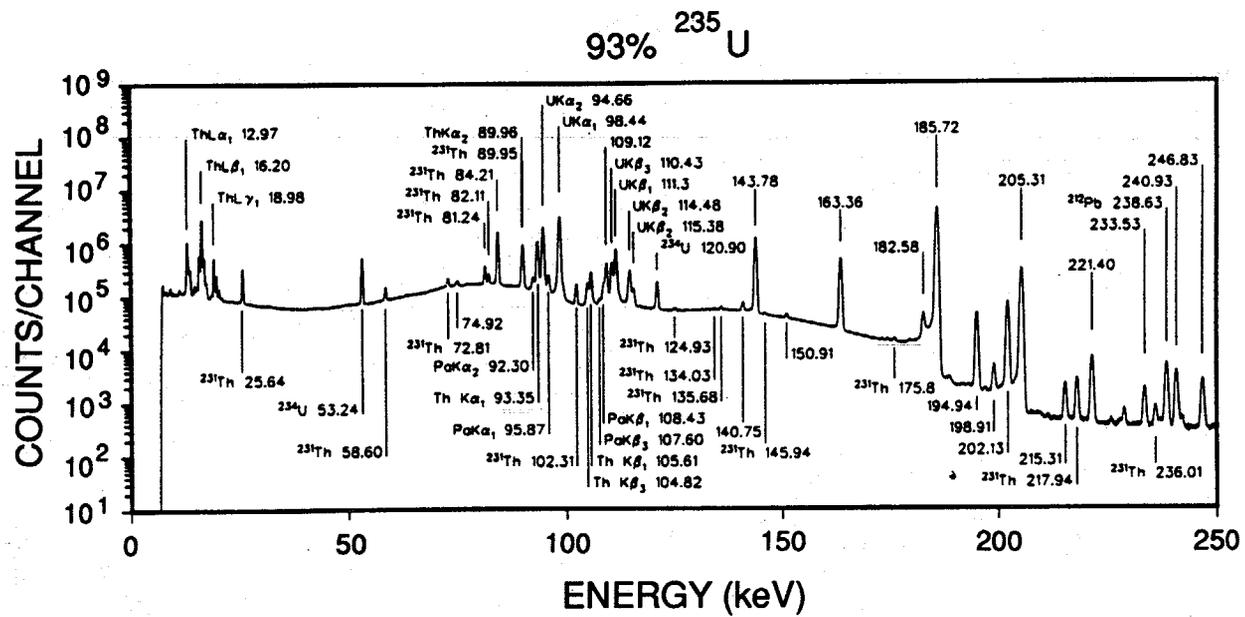


Fig. 1.8 Gamma-ray spectrum of highly enriched uranium showing the intense gamma rays often used for assay of <sup>235</sup>U.

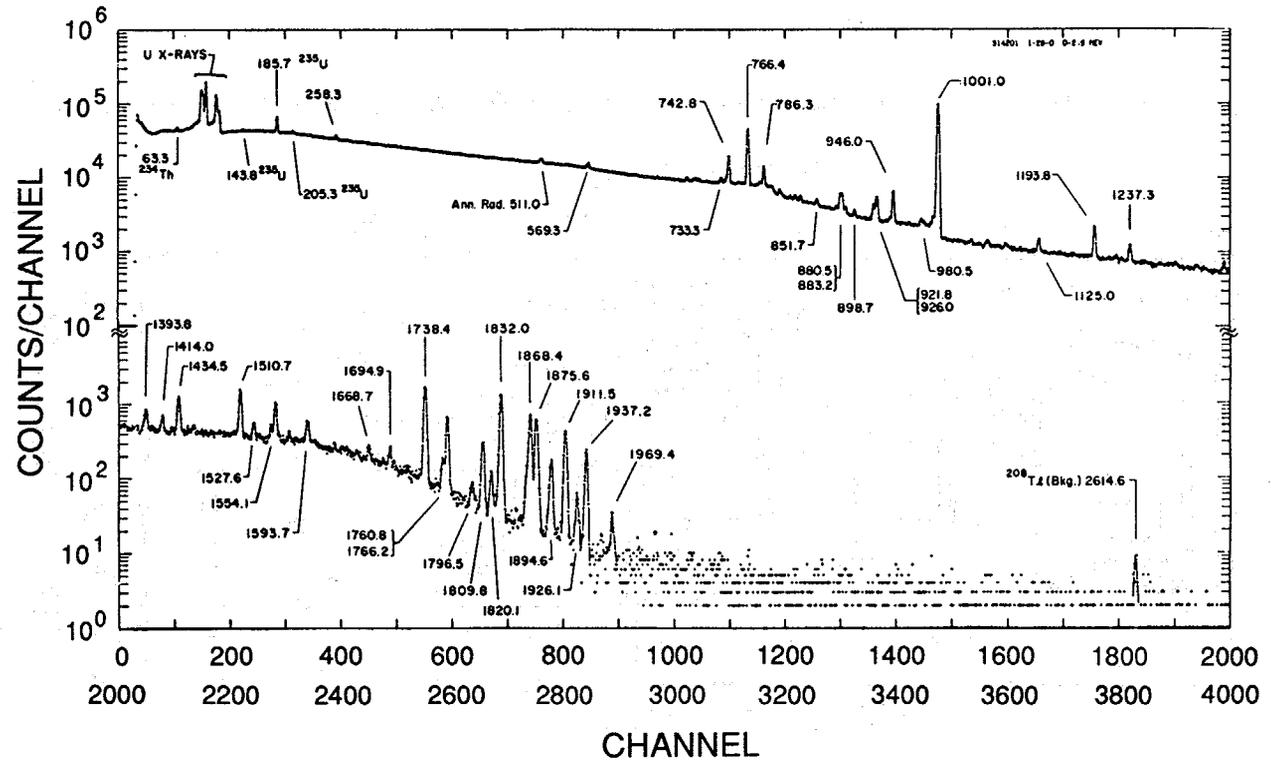


Fig. 1.9 Gamma-ray spectrum of depleted uranium (0.2%  $^{235}\text{U}$ ). The intense gamma rays at 766 and 1001 keV are from the  $^{238}\text{U}$  daughter,  $^{234\text{m}}\text{Pa}$ , and are often used for the assay of  $^{238}\text{U}$ . Most of the weak gamma rays above 1001 keV also come from  $^{234\text{m}}\text{Pa}$ . (Ann. Rad. is annihilation radiation; the small peak at 511 keV is due to positron annihilation.)

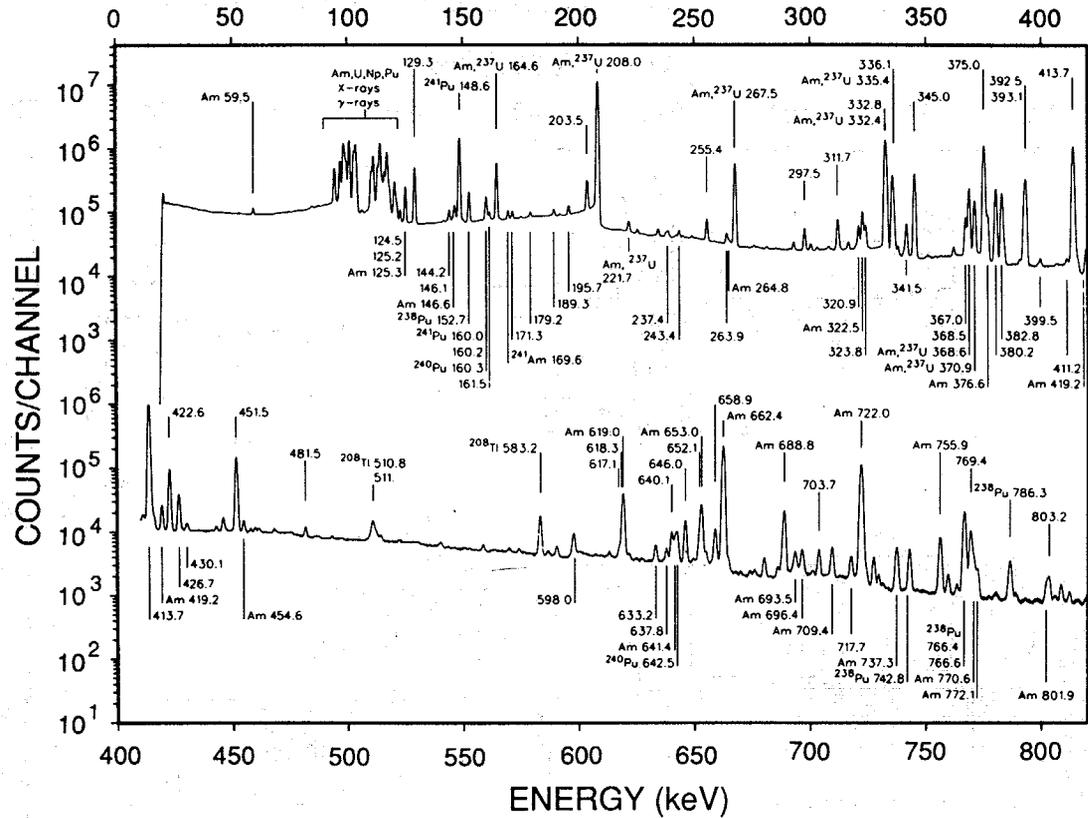


Fig. 1.10 Gamma-ray spectrum of plutonium with 14%  $^{240}\text{Pu}$  and 1.2%  $^{241}\text{Am}$ . Compare this spectrum with that of Figure 1.11, noting the difference in relative intensities. Compare the intensities of the gamma rays at 203.5 keV ( $^{239}\text{Pu}$ ) and 208.0 keV ( $^{237}\text{U}$ , daughter of  $^{241}\text{Pu}$ ).

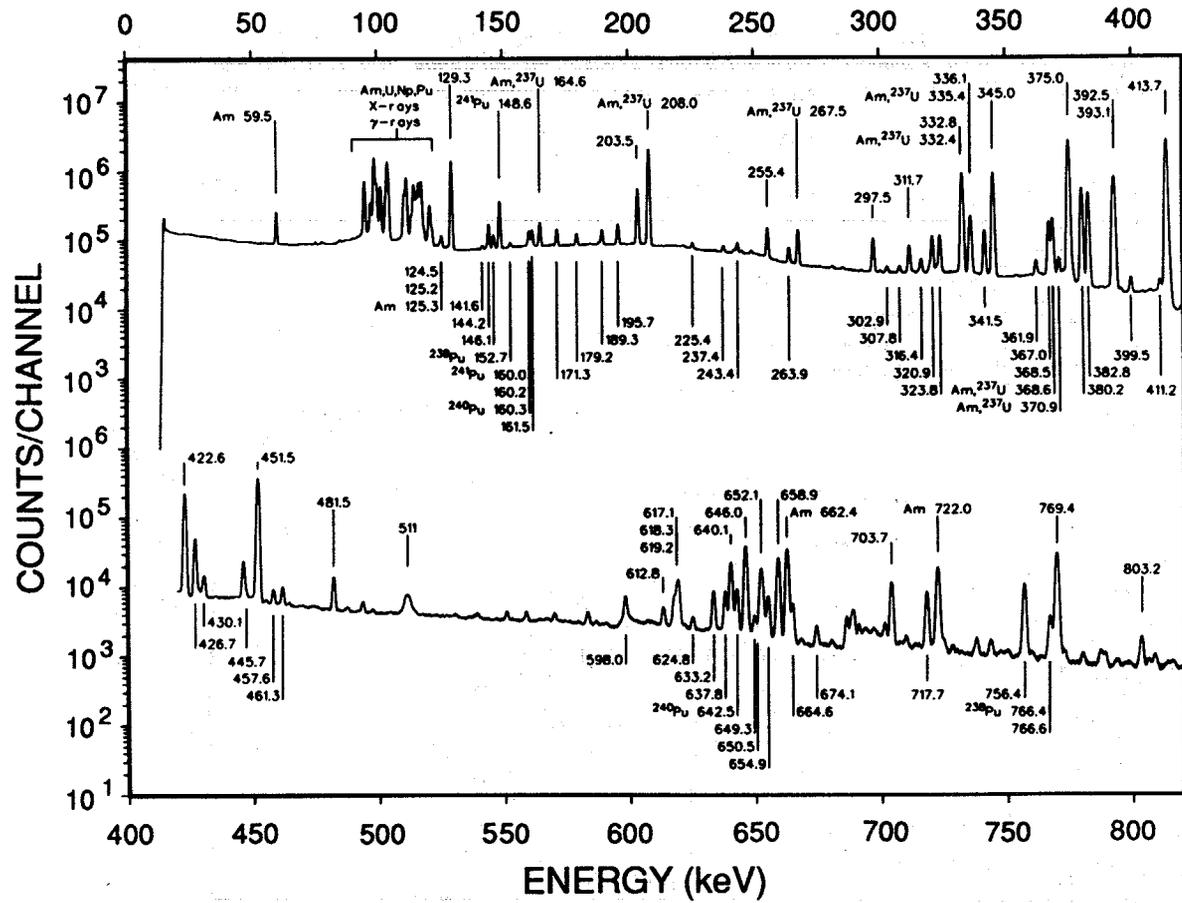


Fig. 1.11 Gamma-ray spectrum of low-burnup plutonium with approximately 6% <sup>240</sup>Pu.

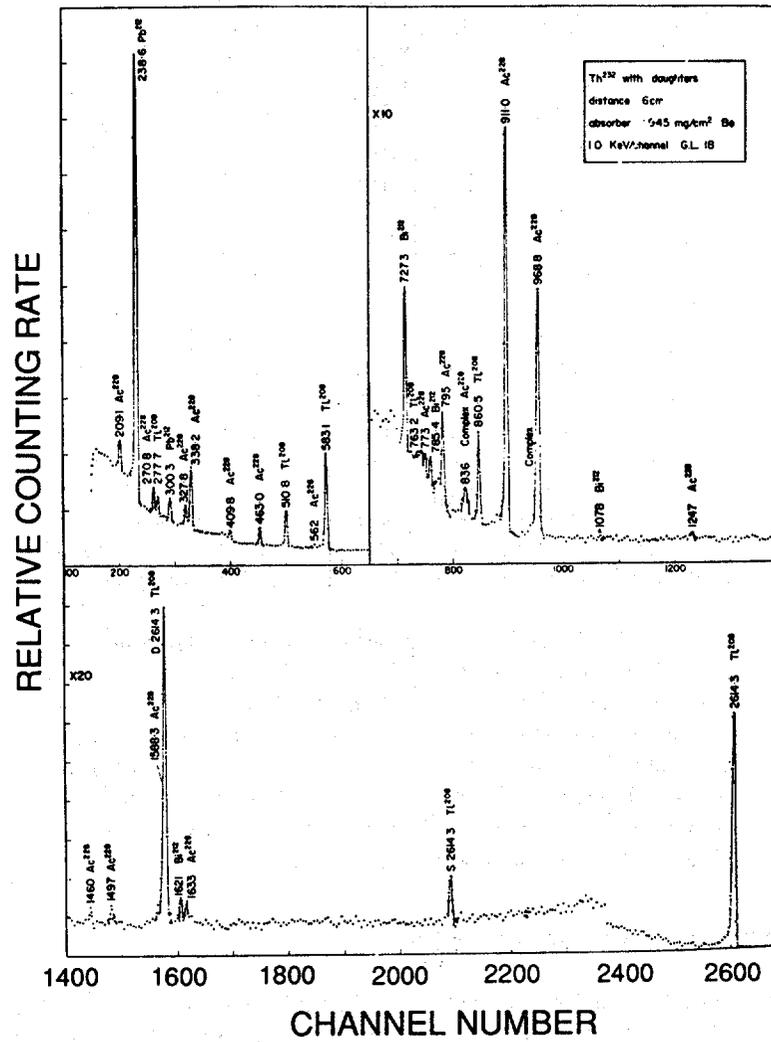


Fig. 1.12 Gamma-ray spectrum of  $^{232}\text{Th}$  and its daughter products. Thorium-232 emits no significant gamma rays of its own. The daughter nuclides grow into equilibrium with the  $^{232}\text{Th}$  parent over a period of approximately 35 yr. [Figure adapted from F. Adams and R. Dams, Applied Gamma-Ray Spectroscopy, 3rd ed. (Pergamon Press, Oxford, 1970).]

Table 1-2. Major NDA gamma-ray signatures

Isotope	Energy <sup>a</sup> (keV)	Activity <sup>a</sup> ( $\gamma/g\text{-s}$ )	Mean Free Path <sup>b</sup> (mm)	
			(High-Z, $\rho$ )	(Low-Z, $\rho$ )
<sup>234</sup> U	120.9	$9.35 \times 10^4$	0.23	69
<sup>235</sup> U	143.8	$8.40 \times 10^3$	0.36	73
	185.7	$4.32 \times 10^4$	0.69	80
<sup>238</sup> U	766.4 <sup>c</sup>	$2.57 \times 10^1$	10.0	139
	1001.0 <sup>c</sup>	$7.34 \times 10^1$	13.3	159
<sup>238</sup> Pu	152.7	$5.90 \times 10^6$	0.40	75
	766.4	$1.387 \times 10^5$	9.5	139
<sup>239</sup> Pu	129.3	$1.436 \times 10^5$	0.27	71
	413.7	$3.416 \times 10^4$	3.7	106
<sup>240</sup> Pu	45.2	$3.80 \times 10^6$	0.07	25
	160.3	$3.37 \times 10^4$	0.45	76
	642.5	$1.044 \times 10^3$	7.4	127
<sup>241</sup> Pu	148.6	$7.15 \times 10^6$	0.37	74
	208.0 <sup>d</sup>	$2.041 \times 10^7$	0.86	83
<sup>241</sup> Am	59.5	$4.54 \times 10^{10}$	0.14	38
	125.3	$5.16 \times 10^6$	0.26	70

<sup>a</sup>Data for uranium isotopes are from Ref. 1; data for plutonium isotopes are from Ref. 2 (energy and branching ratio) and Ref. 3 (half-life).

<sup>b</sup>The mean free path is the absorber thickness that reduces the gamma-ray intensity to 1/e. The mean free path in uranium or plutonium oxide ( $\rho = 10 \text{ g/cm}^3$ ) is given for the high-density, high-atomic-number case (high-Z,  $\rho$ ). The mean free path in aluminum oxide ( $\rho = 1 \text{ g/cm}^3$ ) is given for the low-density, low-atomic-number case (low-Z,  $\rho$ ). Attenuation data are from Ref. 4.

<sup>c</sup>From the <sup>238</sup>U daughter <sup>234m</sup>Pa. Equilibrium assumed.

<sup>d</sup>From the <sup>241</sup>Pu daughter <sup>237</sup>U. Equilibrium assumed.

### 1.4.3 Fission-Product Gamma Rays

Considerable interest has been shown in the measurement of irradiated fuel from nuclear reactors. The irradiated fuel has a high monetary value and a high safeguards value because of the plutonium produced during reactor operation. Gamma rays from the spontaneous decay of uranium and plutonium cannot be used for measurement of irradiated fuel because they are overwhelmed by the very intense gamma rays emitted by fission products that build up in the fuel during irradiation. The total gamma-ray intensity of the fission products from light-water-reactor fuel irradiated to 33 000 MWd/tU (megawatt days per ton of uranium) is approximately  $2 \times 10^{10} \text{ } \gamma/g\text{-s}$  (g = gram of uranium) one year after removal of the fuel from the reactor, whereas

the major uranium and plutonium gamma rays have intensities in the range  $10^3$  to  $10^4$   $\gamma/\text{g}\cdot\text{s}$ . In some instances the intensity of one or more fission products can be measured and related to the mass of the contained nuclear material.

Certain high-Z nuclei can fission or split into two or three medium-Z daughter nuclei. The fission process can occur spontaneously or it can be induced when the parent nucleus absorbs a neutron. Spontaneous fission is more probable in nuclei with even atomic mass numbers (A). Induced fission can occur after absorption of either thermal or fast neutrons in nuclei with odd mass numbers; it only occurs after absorption of fast neutrons in even-numbered nuclei. The fission process was first discovered in 1939 by Otto Hahn and Friedrich Strassman and correctly interpreted in the same year by Lise Meitner and Otto Frisch.

The fission of a nucleus is a cataclysmic event when compared with the alpha- and beta-decay processes described in Sections 1.2.2 and 1.2.3. The energy released in fission is approximately 200 MeV. Most of this energy is carried as kinetic energy by the two (rarely three) daughter nuclei (called fission products or fission fragments). The fissioning nucleus also emits an average of two prompt neutrons and six prompt gamma rays at the instant it splits. A typical fission reaction is illustrated by the formula

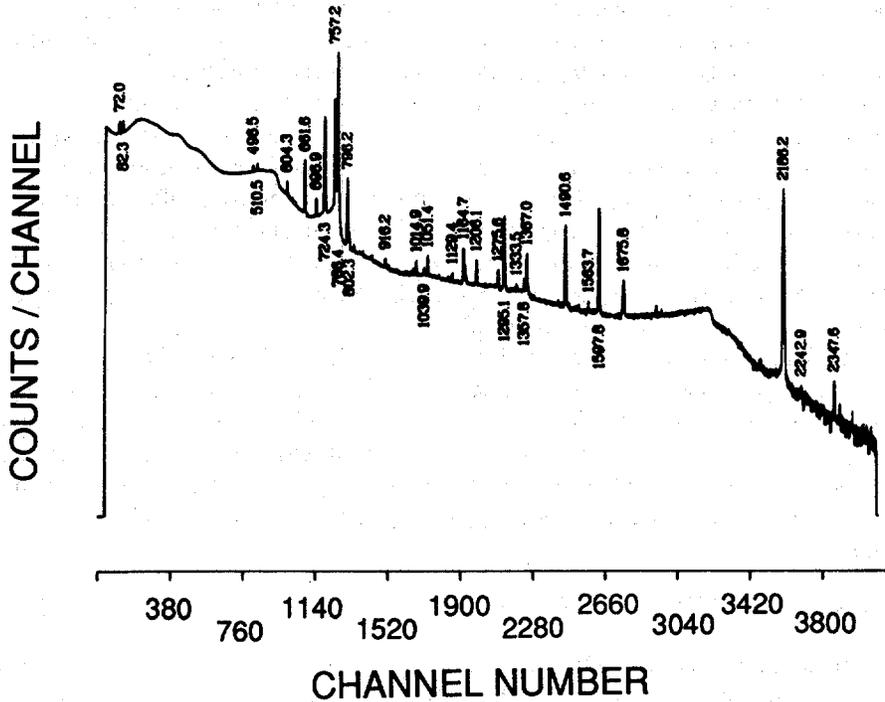


This formula illustrates only one of the many possible fission reactions. The fission-product nuclei themselves are unstable. They have an excess of neutrons and decay by neutron emission or  $\beta^-$  decay (frequently accompanied by gamma-ray emission); the radiations from these reactions are called delayed neutrons and gamma rays. The fission products have half-lives ranging from seconds to years. The gamma rays from fission products can be used to measure irradiated fuel materials.

Typical spectra from irradiated fuel are shown in Figures 1.13 and 1.14. Figure 1.13 shows a spectrum from highly enriched uranium fuel used in a materials test reactor. Figure 1.14 shows an irradiated light-water-reactor fuel spectrum. The most commonly measured fission-product gamma ray is from  ${}^{137}\text{Cs}$  at 661.6 keV. This fission product has a high yield and a sufficiently long half-life (30.2 yr) so that its concentration is proportional to the total number of fissions that have occurred in the fuel. (See Chapter 18 for a more complete discussion of the fission reaction and the measurement of irradiated fuel.)

#### 1.4.4 Background Radiation

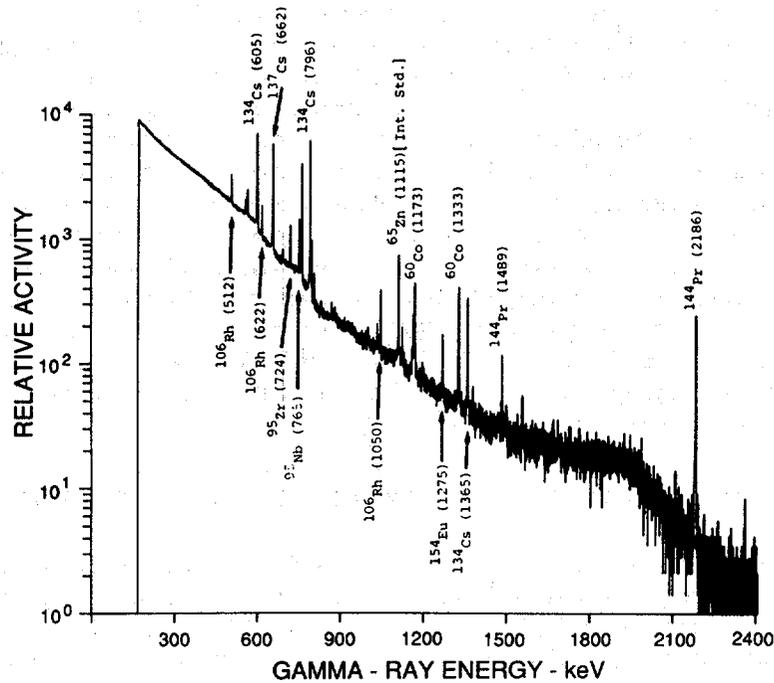
All gamma-ray detectors will give some response even in the absence of a measurement sample. This response is due to the ambient background in the location of the detector. The ambient background consists of radiation from nuclear material in nearby storage areas, cosmic-ray interactions, and natural radioactivity in the local environment.



*Fig. 1.13* Gamma-ray spectrum of highly enriched uranium fuel irradiated in a materials test reactor. The sample had an average burnup of 36.9% and a cooling time of 1.59 yr.

The radiation from the nuclear material stored nearby is of the same nature as the radiation from the samples to be measured. This background spectrum is similar to the spectra shown in Figures 1.7 through 1.12. It often has a high Compton continuum resulting from degradation and scattering in the materials that separate the detector from the storage area. Background radiation from nuclear material can be minimized with a judicious choice of detector location and shielding.

At the earth's surface, cosmic rays consist primarily of high-energy gamma rays and charged particles. Although a neutron component exists, it has little effect on gamma-ray detectors. The charged particles are mostly muons but also include electrons and protons. The muon flux at sea level is approximately  $0.038/\text{cm}^2\text{-s}$ ; at an altitude of 2000 m, the muon flux increases to approximately  $0.055/\text{cm}^2\text{-s}$ . The muon interacts with matter as though it were a heavy electron and its rate of energy loss when passing through typical solid or liquid detector materials is approximately 8.6 MeV/cm. A typical penetrating muon deposits approximately 34 MeV in a 40-mm-thick detector. Because this is much more energy than can be deposited by gamma rays from uranium or plutonium, muon interactions often overload or saturate the detector electronics.



**Fig. 1.14** Gamma-ray spectrum of irradiated pressurized-water-reactor fuel having a burnup of 32 000 MWd/tU (megawatt days per ton of uranium) and a cooling time of 9 months.

For a detector with a front surface area of 20 cm<sup>2</sup>, the typical muon interaction rate at sea level is approximately 0.75/s.

All materials have varying degrees of natural radioactivity. For example: the human body and even gamma-ray detectors have some measurable natural radioactivity; building materials such as concrete can be especially active. The major radioactive species in natural materials are <sup>40</sup>K, <sup>232</sup>Th and its daughters, and <sup>235</sup>U and <sup>238</sup>U and their daughters. Potassium-40 has a natural abundance of 0.0117% and decays by electron capture (10.67%) and  $\beta^-$  decay (89.33%) with a half-life of  $1.277 \times 10^9$  yr. The electron capture is accompanied by the emission of a 1.461-MeV gamma ray that is evident in almost all background gamma-ray spectra. Potassium is present in most organic matter, with <sup>40</sup>K being the major source of radioactivity.

Thorium is a common trace element in many terrestrial rocks. Thorium-232 is the natural parent to the thorium decay series, which goes through 10 generations before reaching the stable nuclide <sup>208</sup>Pb. The half-life of <sup>232</sup>Th is  $1.41 \times 10^{10}$  yr. Its major gamma-ray activity comes from <sup>208</sup>Tl, <sup>212</sup>Bi, and <sup>228</sup>Ac.

Uranium is also found as a trace element in many rocks, although it is less common than thorium. The gamma-ray spectrum of unprocessed uranium ore is much different from that of uranium seen in the nuclear fuel cycle. Because of the long half-life of the daughter  $^{230}\text{Th}$  ( $8 \times 10^4$  yr), later generations take a long time to grow back into equilibrium after any chemical treatment that separates uranium daughters from the natural ore. Figure 1.15 shows a typical spectrum of uranium ore (compare with Figure 1.9). Natural chemical processes in different rocks can often leach out some of the daughter nuclides and cause different ores to have different gamma-ray emissions.

The natural sources discussed above are common and contribute to the background gamma-ray spectrum in most locations. Other sources of background are occasionally encountered, such as materials contaminated by radioactive tracers. Slag from steel furnaces, which can have measurable levels of  $^{60}\text{Co}$ , and uranium tailings are used as a concrete aggregate in some areas. The use of such materials in buildings can contribute to background radiation levels.

## 1.5 ADDITIONAL GAMMA-RAY PRODUCTION REACTIONS

The discussion in Section 1.4 has been limited to gamma rays that come from the natural decay reactions of radioactive nuclides; these gamma rays provide the bulk of the signatures useful for nondestructive assay. This section discusses gamma rays produced in other nuclear reactions. Some of these radiations can interfere with nondestructive analysis.

### 1.5.1 Bremsstrahlung (Braking Radiation)

Charged particles continuously decelerate as they move through condensed materials. As they decelerate, they emit photons with a continuous energy spectrum known as bremsstrahlung; these photons are of interest because their energies are often similar to those used for nondestructive assay.

Beta particles from nuclear decay often emit bremsstrahlung photons while stopping. Although beta particles have a very short range in condensed matter and rarely escape from the host material, the bremsstrahlung photons often escape and are detected along with the gamma rays of interest for nondestructive assay. Internal conversion electrons can also contribute to the production of bremsstrahlung radiation. The discrete gamma rays emitted by a decaying nucleus may be superimposed on a continuous bremsstrahlung background. The electron linac (linear accelerator) uses the bremsstrahlung reaction to produce high-energy photons for nuclear research, nuclear medicine, and active nondestructive assay of nuclear materials (Ref. 5).

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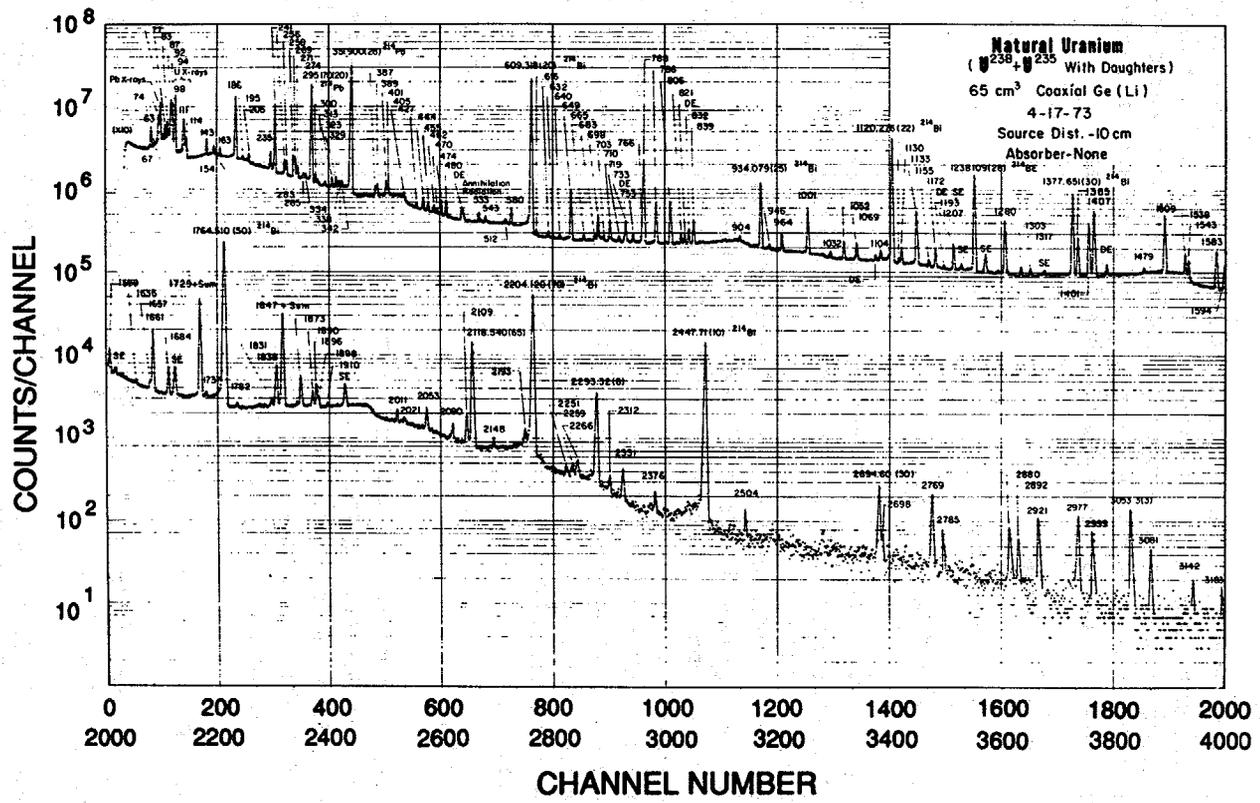
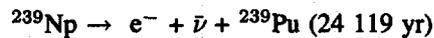
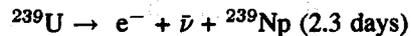
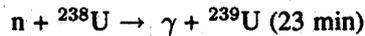


Fig. 1.15 Gamma-ray spectrum of uranium ore. Major radiations are from  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ . Compare the spectrum of processed uranium fuel in Figure 1.9; most daughter products are removed during processing. Figure adapted from R. L. Heath, "Gamma-Ray Spectrum Catalogue," Aerojet Nuclear Corporation report ANCR-1000-2 (March 1974).

### 1.5.2 Particle Reactions

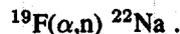
When nuclei interact with other particles, charged or neutral, they often emit gamma rays as products of the interaction. The neutron capture reaction ( $n,\gamma$ ) is a classic example. Usually the new nucleus is radioactive and is created in an excited state from which it can decay by gamma-ray emission. The following formulas illustrate the neutron-capture reaction that breeds plutonium in a fission reactor:



Gamma rays from the capture reaction have discrete energies that are characteristic of the levels of the daughter nucleus. Their energies are typically 8 to 9 MeV for high-atomic-number nuclei.

Inelastic scattering of neutrons ( $n,n'\gamma$ ) is usually accompanied by gamma-ray emission. The gamma rays have discrete energies that are characteristic of the levels in the target nucleus. Gamma rays produced by this reaction are usually not of interest for nondestructive assay.

A major source of neutrons from plutonium compounds and  $\text{UF}_6$  is the interaction of alpha particles from nuclear decay with low-atomic-number nuclei in the compound or surrounding matrix material. The ( $\alpha,n$ ) reaction is frequently accompanied by gamma-ray emission from the excited product nucleus. The fluorine reaction can be written as follows:



The fluorine reaction usually proceeds to the ground state of  ${}^{22}\text{Na}$  and does not result in gamma-ray emission. However, the subsequent  $\beta^+$  decay of  ${}^{22}\text{Na}$  leads to gamma rays with energies of 511 and 1275 keV. These radiations are evident in samples of  $\text{PuF}_4$  and  ${}^{238}\text{PuO}_2$  with trace fluorine impurities. They are not useful as assay signatures, but may be a source of interference.

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3. "Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control," ANSI N15.22-1975 (American National Standards Institute, Inc., New York, 1975) and 1986 revision.
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