

## Chapter 2

# Radioisotopes

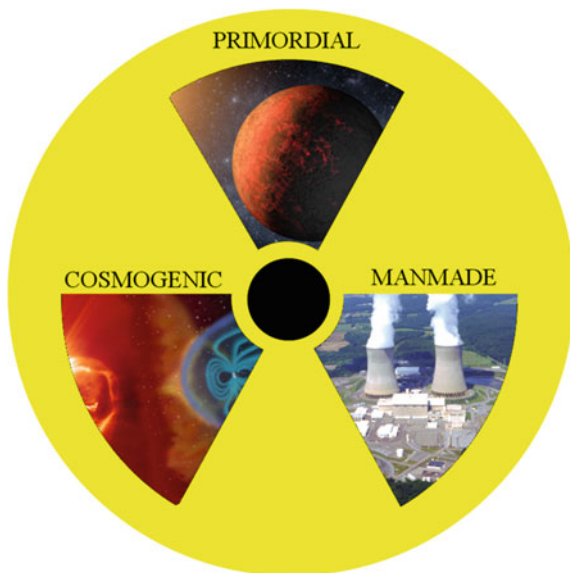
**Abstract** Radioisotopes are of critical importance in nuclear batteries due to their role as the battery's power source. Characteristics of radioisotopes such as type of radiation emitted, decay energy of its radiation, and half-life all influence their usefulness. The type of radiation will determine whether or not there is a good match between the range of radiation and the scale length of the transducer, the decay energy of the radiation determines the effective power density of the source, and the half-life determines the effective lifetime of the nuclear battery. Because of this, special attention must be given during the radionuclide selection process so that it fits the desired design criteria. While simple in concept, the limitations enforced by the physical properties of the substance being used represent an extremely significant barrier to the production of nuclear batteries. Another significant barrier to the production of nuclear batteries arises from limitations in the isotope supply and cost of production. These limitations can be surprising, as a common misconception is that radioisotope supplies are at least abundant, even if they cannot be cheaply produced. For nuclear batteries, supply is an issue because of the large amount of isotopes that would be needed to produce significant power and overcome transducer inefficiencies. For example, a good transducer can only get an average of 1 mW of power per Ci of radionuclide. Production is also an issue because of the cost of producing radionuclide amounts large enough to produce significant power in the nuclear battery. In this chapter, the reader receives a more detailed introduction to radioisotopes, which includes an analysis of origin, existing supplies, production methods, associated production costs, and some basic isotope selection complications.

**Keywords** Radioisotopes supplies • Radioisotope production • Radioisotope costs

### 2.1 Existing Radioisotope Supplies

Radioactive elements existed in the Earth since its formation and can be found in air, water, and soil. Their origin is both terrestrial and extraterrestrial. They can be classified into three groups (Fig. 2.1): primordial, cosmogenic, and manmade.

**Fig. 2.1** Types of radioisotope sources



Primordial radionuclides are the ones created in Earth's formation and their radioactive decay products, cosmogenic radionuclides are the ones produced by cosmic-ray interactions, and manmade radionuclides are those produced by humans after the discoveries of radioactivity and nuclear fission [1]. As both primordial and cosmogenic radioisotopes have a natural origin, they are called Naturally Occurring Radioactive Materials (NORM).

### ***2.1.1 Primordial Radioisotopes***

Some of the primordial radionuclides that were created during the Earth's formation have survived until now due to their long half-lives [2]. One example is U-238, whose half-life is  $4.468 \times 10^9$  years. This time is very close to the age of the Earth, calculated to be around  $4.5 \times 10^9$  years [3]. Some of those long-lived radioisotopes undergo a series of successive radioactive decays, generating radioisotopes with shorter half-lives. Those series of successive radioactive decays are known as decay chains, these chains stop with the production of a stable isotope.

The three existing natural decay chains are the uranium, thorium, and actinium series. Each name originates from the parent radionuclide, which has a much longer half-life than the other radioisotopes in the series. This condition generates secular equilibrium, which is when the activity of the daughter isotope equals the activity of the parent isotope after approximately eight half-lives of the daughter. Taking advantage of this condition, the amount of daughter radioisotopes can be estimated if the amount of the parent radioisotope is known. Uranium and thorium are the two

parent radioisotopes in the three decay chains. Assuming world uranium reserves of about 7,900,000 t and world thorium reserves of 5,385,000 t [4, 5], an estimated inventory of the radioisotopes pertaining to the three decay chains was calculated (Table 2.1). The table was calculated assuming secular equilibrium and an undisturbed sample, which means no loss of gaseous radioisotopes like Radon. The Nuclear data for Table 2.1 was obtained from the National Nuclear Data Center site of Brookhaven National Laboratory [6], and the activities of the daughter were calculated considering the branching probabilities of the decay chains.

One additional decay chain, called the neptunium series, has a parent radioisotope of Np-237. It is no longer found in nature since the half-life of Np-237 is 3 three orders of magnitude less than the age of the Earth. However, this series can be produced artificially through Pu-241, which decays to Np-237.

There are also primordial radionuclides, present in nature, that do not form part of decay chains. They have very long half-lives and their decay product (daughter) is a stable isotope. To have an idea of the amount of the non-series primordial radionuclides, an estimation was done and is shown along with half-lives, relative abundances, and types of radiation in Table 2.2. The supplies were calculated using information about the abundance of elements in the Earth's crust (EC) and in the sea from the Handbook of Chemistry and Physics [7]. Also, it was assumed that the mass of the Earth's crust is  $2.36 \times 10^{22}$  kg, the mass of the hydrosphere is  $1.664 \times 10^{21}$  kg, and seawater is about 97 % of the hydrosphere [7]. Sm-149 and Dy-156 were not included in the table because some references consider them radioisotopes [1], but others [6] label them as stable isotopes. This might be due to fact that it is very difficult to determine if an isotope is radioactive when it has a very long half-life. As improved experiments (greater sensitivity and lower background) become available, the number of confirmed non-series radionuclides may increase [8].

K-40 is the most abundant radioisotope in Table 2.2, which is not surprising because it is the eighth most abundant element in both the Earth's crust and the sea. If we could make 1 kW nuclear batteries using the total amount of K-40 available, that would result in about 423 million batteries. However, it is impossible and infeasible to extract or separate such high quantities of a radioisotope. Further, none of the non-series primordial radioisotopes meet the proper characteristics to be used in nuclear batteries. It may be economical to use primordial radionuclides only if large quantities are not needed due to extraction difficulties.

### 2.1.2 *Cosmogenic Radioisotopes*

Cosmogenic radioisotopes are created by the interaction of cosmic-ray particles with Earth's soil or atmosphere. The flux of cosmic-ray particles is made up of energetic particles (mostly protons with energies of 1–100 MeV) striking Earth's atmosphere. They originate from both the sun and other galaxies in the universe [1]. Most of the atmospheric radionuclide generation takes place in the stratosphere [9] with a higher production around the magnetic poles and lower in the regions near

**Table 2.1** Estimated world supply of natural decay chains' radioisotopes

Series	Isotope	Half-life (year)	World supply (kg)	Total activity (Ci)	Type of radiation
Uranium	U-238	4.47E+09	7.843E+09	2.636E+06	$\alpha, \gamma$
	Th-234	6.60E-02	1.139E-01	2.636E+06	$\beta, \gamma$
	Pa-234 m	2.20E-06	3.796E-06	2.631E+06	$\beta, \gamma$
	U-234	2.46E+05	4.237E+05	2.636E+06	$\alpha, \gamma$
	Th-230	7.54E+04	1.279E+05	2.636E+06	$\alpha, \gamma$
	Ra-226	1.60E+03	2.667E+03	2.636E+06	$\alpha, \gamma$
	Rn-222	1.05E-02	1.714E-02	2.636E+06	$\alpha, \gamma$
	Po-218	5.89E-06	9.469E-06	2.636E+06	$\alpha$
	Pb-214	5.10E-05	8.039E-05	2.636E+06	$\beta, \gamma$
	Bi-214	3.78E-05	5.971E-05	2.636E+06	$\beta, \alpha$
	Po-214	5.21E-12	8.214E-12	2.636E+06	$\alpha, \gamma$
	Pb-210	2.22E+01	3.437E+01	2.636E+06	$\beta, \gamma$
	Bi-210	1.37E-02	2.124E-02	2.636E+06	$\beta, \alpha$
	Po-210	3.79E-01	5.865E-01	2.636E+06	$\alpha, \gamma$
	At-218	4.75E-08	1.528E-11	5.270E+02	$\alpha$
	Pa-234	7.64E-04	4.349E-06	8.692E+03	
Actinium	U-235	7.04E+08	5.691E+07	1.214E+05	$\alpha, \gamma$
	Th-231	2.91E-03	2.284E-04	1.214E+05	$\beta, \gamma$
	Pa-231	3.28E+04	2.570E+03	1.214E+05	$\alpha, \gamma$
	Ac-227	2.18E+01	1.679E+00	1.214E+05	$\beta, \alpha, \gamma$
	Th-227	5.11E-02	3.889E-03	1.197E+05	$\alpha, \gamma$
	Ra-223	3.13E-02	2.370E-03	1.214E+05	$\alpha, \gamma$
	Rn-219	1.25E-07	9.333E-09	1.214E+05	$\alpha, \gamma$
	Po-215	5.64E-11	4.121E-12	1.214E+05	$\alpha, \beta$
	Pb-211	6.86E-05	4.918E-06	1.214E+05	$\beta, \gamma$
	Bi-211	4.07E-06	2.915E-07	1.214E+05	$\alpha, \beta, \gamma$
	Tl-207	9.07E-06	6.356E-07	1.211E+05	$\beta, \gamma$
	Po-211	1.64E-08	3.278E-12	3.397E+02	$\alpha, \gamma$
	Fr-223	4.18E-05	4.372E-08	1.675E+03	$\beta, \alpha, \gamma$
Thorium	Th-232	1.400E+10	5.385E+09	5.927E+05	$\alpha, \gamma$
	Ra-228	5.750E+00	2.174E+00	5.927E+05	$\beta$
	Ac-228	7.016E-04	2.652E-04	5.927E+05	$\beta, \gamma$
	Th-228	1.912E+00	7.227E-01	5.927E+05	$\alpha, \gamma$
	Ra-224	9.944E-03	3.693E-03	5.927E+05	$\alpha, \gamma$
	Rn-220	1.762E-06	6.427E-07	5.927E+05	$\alpha, \gamma$
	Po-216	4.595E-09	1.646E-09	5.927E+05	$\alpha$
	Pb-212	1.214E-03	4.266E-04	5.927E+05	$\beta, \gamma$
	Bi-212	1.151E-04	4.046E-05	5.927E+05	$\alpha, \beta, \gamma$
	Po-212	9.475E-15	2.134E-15	3.797E+05	$\alpha$
	Tl-208	5.805E-06	7.191E-07	2.129E+05	$\beta, \gamma$

**Table 2.2** Estimated world supply of non-series primordial radioisotopes

Radio-isotope	Relative abundance	Half-life (years)	Supply Earth's crust (kg)	Supply sea (kg)	Total activity EC (Ci)	Total activity sea (Ci)	Type of radiation
K-40	0.0117	1.248E+09	5.771E+16	7.327E+13	4.228E+14	5.368E+11	$\beta$ , $\epsilon$
V-50	0.25	2.10E+17	7.080E+15	1.009E+10	2.366E+05	3.371E-01	$\epsilon$ , $\beta$
Rb-87	27.85	4.81E+10	5.915E+17	5.394E+13	5.144E+13	4.691E+09	$\beta$
In-115	96.67	4.41E+14	5.704E+15	3.121E+13	4.027E+07	2.203E+05	$\beta$
Te-123	0.87	9.20E+16	2.053E+11	n/a	6.253E+00	n/a	$\epsilon$
La-138	0.089	1.02E+11	8.192E+14	4.884E+06	2.067E+10	1.232E+02	$\epsilon$ , $\beta$
Ce-142	11.7	5.00E+16	1.836E+17	2.266E+08	9.370E+06	1.156E-02	$\beta$
Nd-144	23.8	2.29E+15	2.331E+17	1.076E+09	2.523E+08	1.164E+00	$\alpha$
Sm-147	15.1	1.06E+11	2.512E+16	1.097E+08	5.635E+11	2.460E+03	$\alpha$
Sm-148	11.35	7.00E+15	1.888E+16	8.244E+07	6.414E+06	2.800E-02	$\alpha$
Gd-152	0.205	1.08E+14	3.000E+14	2.316E+06	6.314E+06	4.876E-02	$\alpha$
Hf-174	0.163	2.00E+15	1.154E+14	1.842E+07	1.156E+05	1.844E-02	$\alpha$
Lu-176	2.588	3.76E+10	4.886E+14	6.266E+06	2.655E+10	3.405E+02	$\beta$
Re-187	62.93	4.33E+10	1.040E+13	4.063E+09	4.609E+08	1.801E+05	$\beta$
Pt-190	0.0127	6.50E+11	1.499E+10	n/a	4.225E+04	n/a	$\alpha$
Pb-204	1.4	1.40E+17	4.626E+15	6.779E+08	5.701E+04	8.355E-03	$\alpha$

the equator. The half-lives of these radionuclides are relatively short, which means that they are present in Earth today due to a continuous process of production, decay, and transport. The distribution of these radionuclides is done by air currents. As such, most isotopes coagulate to aerosol particles that could become condensation nuclei for the formation of clouds, and getting deposited in Earth's surface by precipitation. Cosmogenic radionuclides are also produced in rocks but with a smaller rate, depending on the location, size, and exposure history of the rocks [1].

Two examples of cosmogenic radioisotopes are tritium (H-3) and radiocarbon (C-14). The average inventory of cosmogenic isotopes in the environment is shown in Table 2.3, contrasting the values of two references [1, 9]. Among these radioisotopes only tritium is a possible candidate to be used in nuclear batteries. Its worldwide steady state inventory is estimated to be  $35 \times 10^6$  Ci, and if we look at the power that one can extract from tritium in Table 1.1, the worldwide inventory would produce 1312.5 W. Note that in a nuclear battery this amount of power would be reduced further depending on the efficiency of the battery.

**Table 2.3** Cosmogenic isotope inventory

Radioisotope	Half-life	Inventory (MCi)		Type of radiation
		O'Brien	Masarik	
Be-10	1.39E+06 years	3.5E+00	2.5E+00	$\beta$
Al-26	7.17E+05 years	5.6E-03	1.6E-03	$\epsilon$
Cl-36	3.01E+05 years	1.2E-01	2.7E-01	$\beta, \epsilon$
Kr-81	229,000 years	–	4.9E-05	$\epsilon$
C-14	5700 years	2.4E+02	3.0E+02	$\beta$
H-3	12.32 years	3.5E+01	3.5E+01	$\beta$
Na-22	2.603 years	5.0E-03	–	$\epsilon$
S-35	87.37 days	8.0E-02	–	$\beta$
Be-7	53.24 days	7.7E+00	4.9E+00	$\epsilon$
P-33	25.35 days	4.5E-02	–	$\beta$
P-32	14.26 days	4.7E-02	–	$\beta$
Mg-28	20.92 h	2.3E-03	–	$\beta$
Na-24	15 h	9.7E-03	–	$\beta$
S-38	170.3 min	2.0E-03	–	$\beta$
Si-31	157.3 min	2.2E-02	–	$\epsilon$
F-18	109.77 min	4.2E-03	–	$\epsilon$
Cl-39	56.2 min	1.3E-01	–	$\beta$
Cl-38	37.24 min	8.0E-02	–	$\beta$
Cl-34m	32.00 min	5.4E-03	–	$\epsilon, IT$
Al-29	6.56 min	1.4E-03	–	B
S-37	5.05 min	5.5E-03	–	$\beta$
Ne-24	3.38 min	7.2E-04	–	$\beta$
P-30	2.45 min	5.8E-03	–	$\epsilon$
Al-28	2.24 min	2.0E-02	–	$\beta$

### 2.1.3 Manmade Radioisotopes

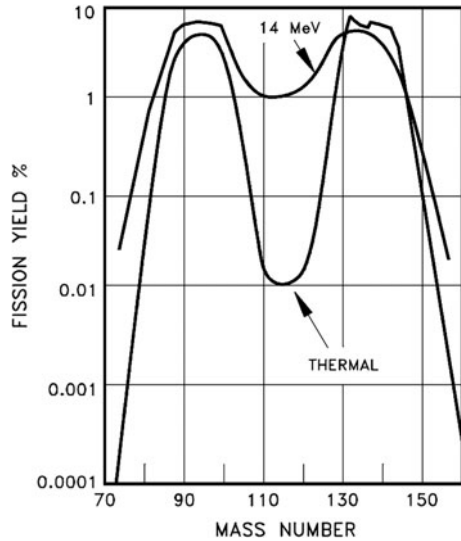
Manmade radioisotopes are generated by the production of nuclear fuels (separation and enrichment of uranium or thorium from natural decay chains), the reprocessing of nuclear fuels (separation of radioisotopes from spent fuel), nuclear power reactors (production of radioisotopes by neutron capture), particle accelerators, and nuclear weapon explosions. The first four production methods are discussed with more detail in Sect. 2.2.

Nuclear reactors use as fuel mostly uranium enriched with the radioisotope U-235. It is obtained from mining, milling, and refining uranium ores. In the process of fabricating nuclear fuel, the gaseous radioisotope Rn-222 could be released. U-235 is utilized because it has a high thermal neutron cross section for fission, which means that when U-235 absorbs a thermal neutron there is a high probability that it splits into two fission fragments and liberates some neutrons and energy. Nuclear reactors take advantage of this to create a controlled chain reaction in which the neutrons released generate more fissions and the heat produced is transformed into electricity. The fuel usually consists of nuclear fuel pellets of uranium dioxide ( $\text{UO}_2$ ), Fig. 2.2, surrounded by a cladding made of a Zirconium alloy [10]. Under normal operation, most of the fission fragments created remain trapped in the nuclear fuel cladding. These fragments, of about 800 different isotopic species, have atomic masses that range from 72 to 160 amu. They are radioactive on the neutron rich side of stability (decay by  $\beta^-$ ), the most probable fission products have masses of 94 and 140 [6, 11, 12]. Figure 2.3 show the fission fragment yields per atomic mass of radioisotope for thermal and fast neutrons. Note that these are the direct yields and that many of these radioisotopes undergo further



**Fig. 2.2** Nuclear fuel pellets [13]

**Fig. 2.3** U-235 fission yields for high- and low-energy (thermal) incident neutrons [11]



beta decays until they reach a stable isotope. Among the isotopes created in the operation of a nuclear reactor, some of them are stable and others have very short half-lives. Around fifty of them live longer than 25 min, and these are the ones that could be dangerous if released during a nuclear accident [1]. In previous nuclear accidents, significant amounts of I-131 and Cs-137 have been released to the environment. However, in normal operation, a very small portion of these radioisotopes leak through the cladding and are released to the moderator (usually light water). However, not all the radioisotopes are generated from fission fragments in a nuclear reactor. The interaction of the neutron flux with the structure materials also produces many radioisotopes such as Cr-51, Mn-54, Fe-55, Fe-59, and Co-60 [1]. Moreover, part of the fuel undergoes neutron capture instead of fission, producing heavier radioisotopes.

Once the fuel is used, a fraction of U-235 and most of the U-238 remains unchanged. Some Pu-239 is created by neutron absorption in U-238. The two radioisotopes of uranium, as well as the plutonium, can be reused to fabricate new fuel. This is called reprocessing. When the nuclear fuel is reprocessed, the radioisotopes created as fission fragments can be separated using different processes discussed in Sect. 2.2. Although it seems a very good option to extend the supply of nuclear fuel and separate useful radioisotopes, it is costly and controversial due to proliferation issues with fissile radioisotopes that could be used to produce weapons like Pu-239 [14].

Once separated from spent fuel, these radioisotopes can be used in different applications like radiopharmaceuticals, medical imaging, tracers, radiation sterilization of food, elimination of insect pests, measuring thickness of materials, remote power sources (RTGs, and nuclear batteries), among others [12].

Worldwide inventories for these radioisotopes are difficult to obtain because the detailed information needed about the nuclear reactors is usually kept confidential and so is not readily available. However, efforts have been made to estimate the world inventory of particular elements and radioisotopes. Some of these, such as plutonium, Am-241, and Kr-85, are useful for nuclear batteries. For example, Pu-238 has been an important radioisotope for space programs since it has been used as a source of power for RTGs. The total world estimated supply of the element (Pu) by the end of 2014 was 2,627 tonnes. Of this, 2,388 tonnes corresponded to civil nuclear reactors (2,113 being irradiated in working nuclear fuel, and 275 in spent fuel), while 240 tonnes corresponded to military stocks [15]. Thus, the most readily available Pu for potential use in nuclear batteries are the 275 tonnes in spent fuel. Two potential Pu sources for nuclear batteries are Pu-238 and Pu-241, which have an estimated concentration of total Pu in typical PWR burned fuel of 1.3 and 4.7 %, respectively [16]. Therefore, there are about 3.57 tonnes of Pu-238 and 12.92 tonnes of Pu-241 worldwide in spent fuel. The amount of Americium that has been produced by the end of 2004 was estimated to be 87 tonnes, with about 68 tonnes being Am-241, a decay product of Pu-241 [17]. In the case of Kr-85, Ahlswede et al. [18] have estimated the global atmospheric content of this radioisotope to be approximately 5400 PBq. This activity corresponds to the amount of Kr-85 that has been released to the atmosphere mainly by nuclear reprocessing facilities, which are the major contributor, and other small sources like nuclear power reactors, naval reactors and isotope production plants. It does not consider the amount of Kr-85 in fuel that has not been reprocessed and thus not released to the atmosphere. Table 2.4 shows the estimated world inventories of Kr-85, Pu-238, and Am-241. The cost of separating these isotopes from spent fuel was considered by the National Research Council [19]. The conclusion of this review was that the feasible approach is to use isotopes that have already been separated in the weapons program to produce Pu-238. The available separated inventory of Pu-238 within the National Laboratories in the United States is about 39 kg and the estimated Np-237 inventory is 300 kg [20]. Pu-238 is created from the  $^{237}\text{Np}(n, \gamma)^{238}\text{Np} \rightarrow ^{238}\text{Pu} + \beta$  reaction. It is feasible to produce Pu-238 from the separated inventory of Np-237 using high neutron flux reactors such as the

**Table 2.4** Estimated world supplies of Kr-85, Pu-238, Pu-241, and Am-241

Radioisotope	Half-life (year)	Year of estimation	Total activity (Ci)	World supply (kg)
Kr-85 <sup>‡</sup>	10.752	2009	1.46E+08	3.73E+02
Pu-238	87.7	End of 2014	6.11E+07	3.57E+03
Pu-241	14.325	End of 2014	1.34E+09	1.29E+04
Am-241	432.6	End of 2004	2.33E+08	6.80E+04

<sup>‡</sup>Global atmospheric content

**Table 2.5** Average information of PWR and BWR reactors in the U.S

Reactor type	First month of operation	Original thermal power (MWt)	Number of assemblies	Initial mass of U (kg)
PWR	Sep-80	2908.9	184	424
BWR	Jan-79	2799.9	755	183

Advanced Test Reactor at Idaho National Laboratory or the High Flux Isotope Reactor at Oak Ridge National Laboratory [19, 21] to produce about 5 kg of Pu-238 per year. The cost of building the production capability is estimated to be 77 million dollars US [19, 20]. The cost of Pu-238 per kg is about 8 million dollars US. Even though the inventory of unseparated Np-237 and Pu-238 worldwide from commercial reactor spent fuel is significant (see Table 2.4) [17], the cost of spent fuel processing is too high to be considered feasible.

Although the estimation of the global inventory has been done only for a few radioisotopes, it is possible to make estimations about the radioisotopes contained in U.S. used nuclear fuel because more information is available. A report by Oak Ridge National Laboratory estimates that as of 2011, there are 67,600 metric tons of heavy metal (MTHM) of commercial used nuclear fuel [22], with 43,900 MTHM correspond to PWR and 23,700 MTHM to BWR. Assuming 70 % (PWR) and 57 % (BWR) as the weight percentages of Uranium found initially in fuel assemblies, the weights corresponding only to spent fuel are: 30,790 tonnes (PWR) and 13,580 tonnes (BWR). Using these masses and the average characteristics of PWRs and BWRs, an estimation of the radioisotope inventories can be made using the methodology proposed by Yancey and Tsvetkov to analyze the spent nuclear fuel [23]. Information representing average reactors is introduced in ORIGEN-ARP (Scale software package) to develop a plant model and estimate the masses of the radioisotopes contained in a used fuel assembly. Once the contents of an “average” PWR and BWR assemblies are obtained, the result is multiplied by the number of assemblies calculated dividing the total mass of spent fuel by the initial mass of U per assembly. The average information about PWR and BWR reactors used in ORIGEN-ARP is shown in Table 2.5 [22]. The estimation of medium half-life radioisotope inventory in spent fuel is shown in Table 2.6. By the end of 2009, there were about 240,000 MTHM of spent fuel worldwide, with 64,500 MTHM corresponding to the U.S. [24], therefore the U.S. spent fuel represents about 27 % of the global amount. Likewise, data in Table 2.6 could represent about 27 % of the global radioisotope supply. This is a gross estimation, because information about U.S. reactors is not necessarily representative of the foreign reactors, which in some cases have different types of fuel, power, etc. However, these numbers provide an idea on how much radioisotope activity might be available by extracting radioisotopes from spent fuel.

**Table 2.6** Estimation of radioisotope supply from spent fuel in the U.S. including potential useful isotopes for nuclear batteries

Radioisotope	Half-life (year)	Total activity (Ci)	Supply (g)
Cs-137	30.08	4.62E+09	5.31E+07
Sr-90	28.9	2.98E+09	2.11E+07
Cm-244	18.1	3.03E+08	3.74E+06
Kr-85	10.752	1.69E+08	4.29E+05
Am-241	432.6	1.58E+08	4.61E+07
Eu-154	8.6	1.01E+08	3.73E+05
Pm-147	2.6234	4.05E+07	4.37E+04
Sm-151	90	1.88E+07	7.16E+05
Cs-134	2.065	1.53E+07	1.18E+04
Eu-155	4.753	1.10E+07	2.23E+04
Sb-125	2.76	3.23E+06	3.08E+03
Cm-243	29.1	1.32E+06	2.56E+04
Sn-121 m	43.9	7.31E+05	1.36E+04
Eu-152	13.528	1.34E+05	7.59E+02
Ru-106	1.02	3.92E+04	1.18E+01
Cd-113 m	14.1	1.15E+04	5.14E+01
Th-228	1.912	1.53E+03	1.86E+00
U-232	68.9	1.51E+03	6.82E+01
Pm-146	5.53	1.33E+03	2.99E+00
Pu-236	2.858	2.84E+02	5.43E-01
Cf-250	13.08	2.39E+01	2.19E-01
Tm-171	1.92	4.80E+00	4.40E-03
Cf-252	2.645	1.12E+00	2.08E-03
Ac-227	21.772	9.10E-02	1.26E-03
Po-210	0.379	8.71E-04	1.94E-07
Pb-210	22.2	8.71E-04	1.14E-05
Ra-228	5.75	1.13E-05	4.14E-08

## 2.2 Radioisotope Production

Due to the limited quantities of radioisotopes available from Naturally Occurring Radioactive Materials and human stockpiles, acquiring significant amounts of many isotopes from existing supplies is unattainable. In order to obtain these substances in quantities sufficient for use, some manner of radioisotope production must be pursued. Unfortunately however, production of these isotopes remains a scientifically complex process. This complexity places a significant barrier on researchers attempting to develop new radioisotope separation methods. Currently, there are only four general methods available for the production of these substances: separation from nuclear reactor spent fuel, separation from natural decay chains, production of radioisotopes through neutron capture in a nuclear reactor, and radioisotope production via the use of a particle accelerator.

### **2.2.1 Separation from Spent Fuel**

The first method of radioisotope production is the separation of isotopes from the spent fuel of a nuclear reactor. In this method, the spent fuel is transported to a reprocessing facility, where it undergoes a series of chemical and/or physical processes. Through these processes, different substances are isolated and removed, leaving the desired chemical elements as products. These chemical elements can then be further processed and refined to yield the proper concentration of the needed radioisotope. Once the process is complete, these recycled isotopes can be used in many applications, including nuclear batteries.

Unfortunately, one of the biggest scientific concerns with this method (in regards to nuclear batteries) is the composition of the isotopes present in spent fuel. Most nuclear reactors in the United States are Light Water Reactors (LWR's). These reactors produce spent fuel that consists of approximately 96 % uranium and less than 1 % plutonium [25]. The remaining 3 % consists of a group of hundreds of radioisotopes known as “fission products,” which were produced during the reactor's operation. It can therefore be concluded that, if a chemical element other than uranium or plutonium is desired for a nuclear battery's operation, an extremely high amount of separation and refinement would be required. This would be a limiting factor in the rate of production. In addition, this problem would also affect the cost of radioisotopes produced using the method, which is an issue discussed later in the chapter.

As was mentioned previously, these concerns apply primarily to scientists interested in the research and development of nuclear batteries. Since this was not the original intention of spent fuel reprocessing, many methods and variations of spent fuel reprocessing have been developed. Before termination of civil spent fuel reprocessing in the United States, the nation employed three different processes to recycle spent fuel: the bismuth phosphate process, the reduction and oxidation (REDOX) process, and the plutonium uranium redox extraction (PUREX) process.

#### **2.2.1.1 Bismuth Phosphate Process**

The first method of radioisotope separation from spent fuel, known as the bismuth phosphate process, originated in the early 1940s from the Metallurgical Laboratory at the University of Chicago. At the time, the University of Chicago was evaluating several chemical separation processes for use in the Manhattan Project [26], a secret military program to design and construct the first nuclear bomb. After evaluation of the potential processes was completed, it was concluded that the bismuth phosphate process was the best choice for separation of plutonium radioisotopes. Three large bismuth phosphate plants were subsequently built by The Manhattan District in Hanford, Washington. These plants were called T, B, and U, but were often nicknamed “canyons” or “Queen Mary's” because of their massive size (see Fig. 2.4). The fundamental concept behind these bismuth phosphate plants was to



**Fig. 2.4** T Plant (*left*) and U Plant (*right*) in Hanford, Washington [27]. “Canyons” such as these could be larger than 800 feet long, 85 feet wide, and 100 feet high

take advantage of different plutonium (Pu) valence states to recycle the spent fuel. In the  $\text{Pu}^{4+}$  valence state, plutonium has the ability to co-precipitate with bismuth phosphate ( $\text{BiPO}_4$ ). However, in the  $\text{Pu}^{6+}$  valence state, plutonium loses this capacity. By manipulating plutonium through this difference in valence states, the bismuth phosphate process produced an end product that consisted of a plutonium nitrate solution.

The first step in the bismuth phosphate process was to remove the aluminum reactor fuel cladding. This was done by submerging the spent fuel in a boiling sodium hydroxide and sodium nitrite solution. The bare fuel, containing mostly uranium and small amounts of plutonium and other fission products, was then dissolved in a concentrated aqueous nitric acid solution. This solution was stabilized with sulfuric acid to stop the uranium from precipitating [28]. To acquire the proper valence state, nitrous acid was added to reduce the plutonium. Bismuth phosphate was added next, in order to co-precipitate with the plutonium. The plutonium and bismuth phosphate were isolated through the use of centrifuges, and then washed [29]. This complete cycle would then be repeated multiple times in order to purify the product. After the cycles had completed, the plutonium solution would be further decontaminated and concentrated via a lanthanum fluoride carrier, shrinking the volume of the solution from about three hundred and thirty gallons down to 8 gal [26]. This volume reduction transformed the plutonium solution into a paste. Further processing of the paste turned it into plutonium metal. For this process, the waste stream contained the uranium and most of the fission products (see Fig. 2.5).

Unfortunately, there were several significant problems with the bismuth phosphate process. First, the spent fuel had to be processed in batches [30]. A second major issue was the loss of several useful products in the waste stream, such as the uranium. One more problem was the low reprocessing rate. On average, a bismuth phosphate plant could only reprocess about one metric ton of fuel per day [26]. Another disadvantage was product quantity: One metric ton of bare fuel produced only 2.5 kg of plutonium product. Additionally, that same ton of bare fuel created about ten thousand gallons of liquid waste. This resulted in approximately 1.5

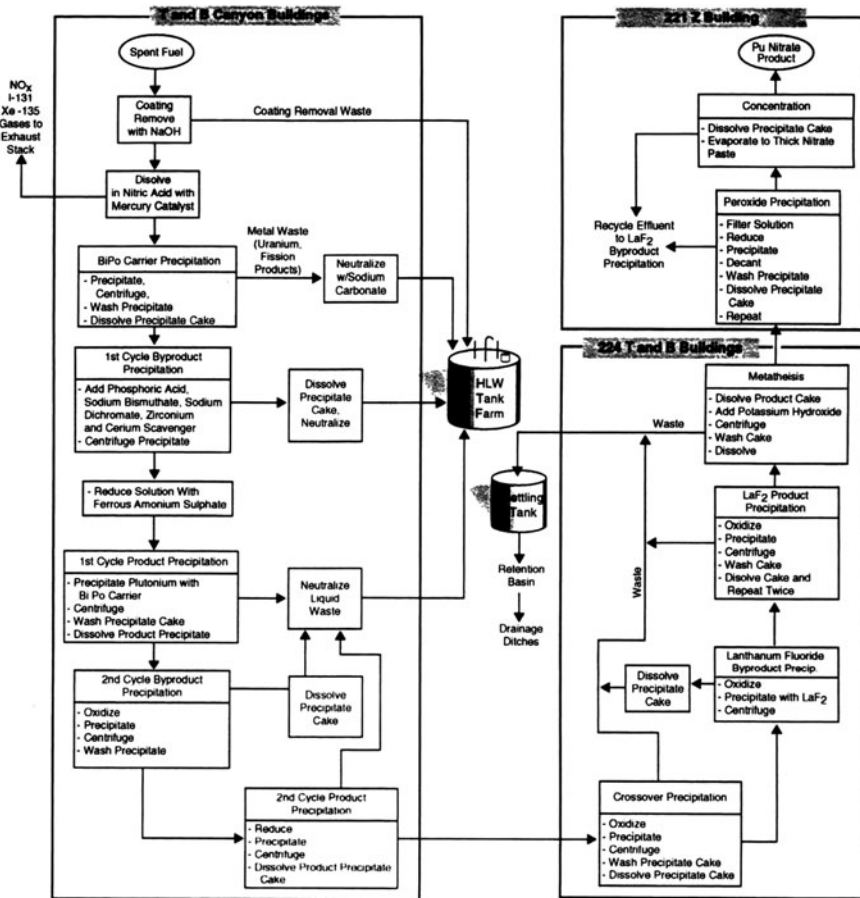


Fig. 2.5 The bismuth phosphate process [26]

million gallons of contaminated wastewater being discharged into the ground each day [31]. Of course, in addition to all these problems, there were issues with the reprocessing plants themselves.

In order to accommodate the necessary radiation shielding for the spent fuel, the T, B, and U reprocessing plants were extremely large. The main buildings (canyons) were over 800 feet long, 85 feet wide, and 100 feet high. Each facility utilized six-foot thick concrete walls to shield workers from the radioactivity. Every plant was divided into at least 20 sections, known as “process cells,” with removable shield covers to separate them. Direct exposure to the fuel or the processing equipment was extremely hazardous due to the radiation dose received, and could reach fatal exposure levels in under a minute [31]. To minimize such exposure, the facilities used overhead cranes and manipulators operated by remote access. In order to successfully operate this equipment, each facility had shielded operating

galleries that ran the length of the building. These contained electrical equipment, control equipment, and pipes, as well as the plant workers themselves. Due to the dangers, a closed-circuit television system was installed to allow equipment operators to see inside the canyons while remaining safe from radiation. Each facility used a ventilation system that passed outside air into the personnel areas, and then drew it into the processing area before it was filtered and sent out via a tall stack [26].

### 2.2.1.2 REDOX Process

As discussed previously, the bismuth phosphate reprocessing method had some significant disadvantages. During the 1940s, difficulties in obtaining uranium made the loss of the uranium in the waste stream the main concern. In order to stop this loss and recover the uranium, the US developed the Reduction and Oxidation (REDOX) fuel separation process [31]. A pilot plant was tested at Oak Ridge National Laboratory in 1948–1949, followed swiftly by a full scale reprocessing facility built at Hanford in 1951 [30]. This facility, known as the “S Plant,” was significantly less massive than its bismuth phosphate predecessors, measuring a mere 470 feet long by 160 feet wide (Fig. 2.6). During its peak operating efficiency, the S Plant could process up to twelve metric tons of spent fuel each day. Over the course of its operational lifetime, the S Plant at Hanford processed roughly 24,000 metric tons of spent fuel. The facility was closed in 1967 [27].

The REDOX method was a critical step forward for the nuclear community in the development of spent fuel reprocessing. Unlike with the bismuth phosphate process, the REDOX technique was a countercurrent, continuous flow solvent extraction process. This eliminated the need to process the spent fuel in batches. Further, the REDOX process produced uranium and plutonium as products. This was accomplished by first removing the reactor fuel cladding in a nitric acid, similar to the bismuth phosphate process. The aqueous solution from this step was then mixed with an organic solvent called Hexone (methyl isobutyl ketone), which is immiscible with water. During the mixing, the uranium and plutonium nitrates



**Fig. 2.6** REDOX Plant in Hanford, Washington [27]

would get extracted from the aqueous solution and transferred into the Hexone. Due to its immiscibility, the Hexone could be easily separated from the aqueous solution. The uranium and plutonium were then chemically reduced and separated. These products were then concentrated and sent to other facilities to be refined and used [26].

In comparison to the bismuth phosphate process, the REDOX technique definitely had both advantages and disadvantages. One obvious advantage of the REDOX method over the bismuth phosphate process was the ability to extract uranium. Another advantage was the consolidation in the physical plant size required while simultaneously increasing the number of plutonium processing operations that could be done inside the facility. Unfortunately, the REDOX process had some serious disadvantages as well. Waste from the plant was far less than ideal: While it was smaller in volume than the waste produced by previous methods, it contained more chemicals and was substantially hotter when it was discharged. Also of major concern was the use of Hexone, as it is volatile and explosive [27]. At only 69 °F, this organic solvent reaches its flash point—the lowest temperature in which the liquid can evaporate enough fluid into the surrounding air to create an ignitable gas. Such a low flash point necessitated the entire REDOX reprocessing operation be conducted in an inert atmosphere [31].

### 2.2.1.3 PUREX Process

Another technique to separate specific radioisotopes from spent fuel is called the Plutonium Uranium Recovery by Extraction (PUREX) process. The PUREX process was developed by Knolls Atomic Power Laboratory in Schenectady, New York during the early 1950s. Part of the driving force for PUREX development was that the reprocessing technique was significantly safer than the REDOX process: As it didn't incorporate Hexone, there was no need for an inert atmosphere or for concern about a Hexone based explosion. After development, the method was tested at Oak Ridge National Laboratory in Tennessee from 1950–1952 [30]. It was then adopted by the Savannah River Site in South Carolina. During November of 1954, the first PUREX reprocessing facility was opened at Savannah River in order to recover the plutonium that had been residing in the spent fuel from five reactors on site [26]. By January 1956, Hanford had constructed their own PUREX reprocessing facility and starting operations (see Fig. 2.7). At over 1000 feet long, 60 feet wide, and 100 feet high, the Hanford PUREX reprocessing facility was the largest chemical processing plant on the site [32].

While similar to the REDOX method in many respects, the PUREX process allowed the countercurrent, continuous flow solvent extraction process to be conducted under a considerably larger amount of safety. It also allowed additional radioisotopes to be extracted. Just like with the REDOX technique, the first step in the PUREX process involved removing the reactor fuel cladding through the use of a nitric acid. The aqueous solution was then mixed with the organic solvent Tributyl phosphate (TBP) [33]. Uranium, plutonium, and other desired radioisotopes (such



**Fig. 2.7** PUREX Plant in Hanford, Washington [27]. The PUREX “canyon” was over 1000 feet long, 60 feet wide, and 100 feet high [32]

as neptunium) got extracted from the aqueous solution and transferred into the TBP through valence state manipulation. These products would then be concentrated in another organic solvent such as paraffin, where they would be subjected to a chemical scrubbing via dilute nitric acid. Both the solvent extraction and scrubbing steps would be repeated multiple times in order to separate and purify the products. Each final product would be in the form of a nitrate [31]. A diagram of the PUREX process is shown in Fig. 2.8.

For the nuclear community, the PUREX process definitely proved itself to be superior to the REDOX technique in almost all respects. As was previously discussed, the PUREX process didn’t incorporate Hexone, which required an inert atmosphere inside the facility and created concern about a Hexone based explosion. The organic solvent used instead, known as TBP, has a much higher flash point (almost 300 °F). This made it possible to use a natural atmosphere with no explosion concerns. Another factor in favor of PUREX was its capability to recover multiple isotopes. Lastly, as might be expected from the previously mentioned benefits, the PUREX process had an higher efficiency than earlier methods [26].

Even though the PUREX process was a significant improvement over the REDOX method, the plutonium product produced with the new technique created a serious concern. Due to its central role in the making of nuclear weapons, the plutonium product quickly became a nuclear weapons proliferation issue. In 1976, President Gerald Ford indefinitely suspended the commercial reprocessing and recycling of plutonium [34]. This directly affected the PUREX method. By the end of April of 1977, President Jimmy Carter banned all civilian reactor nuclear fuel reprocessing in the United States [35].

#### 2.2.1.4 Other Processes

Fortunately, while civil spent fuel reprocessing in the United States was terminated after the creation of the PUREX process, development in this area has continued.

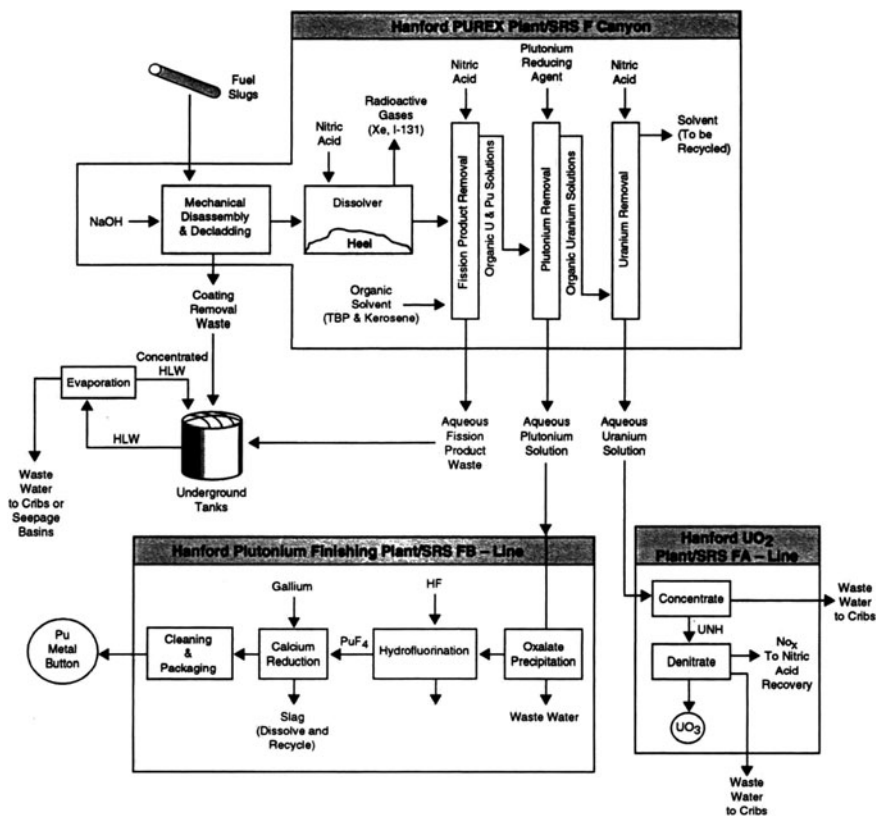


Fig. 2.8 The PUREX Process [26]

This has resulted in numerous different chemical reprocessing methods and their variations. For example, one of the variations to the PUREX process is the TRUEX reprocessing method, which attempts to isolate transuranic elements. Another PUREX reprocessing variation is the DIAMEX-SANEX process, which removes long-lived radioisotopes (like americium and curium) and then separates them into different products. The COEX process extracts a mix containing both uranium and plutonium, as well as a pure uranium stream. Lastly, the GANEX technique extracts a mix containing both uranium and plutonium, similar to COEX, except it also has the ability to separate some of the actinides. Many more examples can be given, however the main concept and issues associated with chemical spent fuel reprocessing remain.

In an attempt to avoid the complications associated with chemical spent fuel reprocessing, some organizations are investigating reprocessing alternatives. One such method worth mentioning is pyroprocessing, currently being investigated by Argonne National Laboratories in Chicago. Unlike other methods, this technique utilizes a process called electrorefining to separate radioisotopes. Similar to

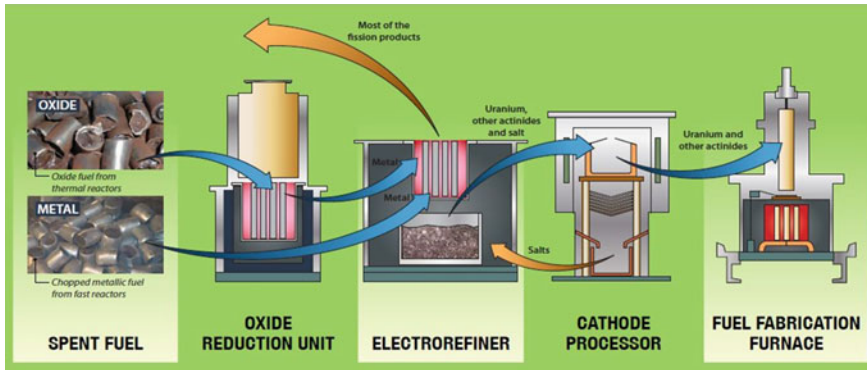


Fig. 2.9 Pyroprocessing Schematic [36]

electroplating, electrorefining involves attaching the spent fuel, already processed into a metallic form, to an anode suspended in a molten salt bath. An electric current is applied across the bath, and the fuel elements dissolve. Recyclable radioisotopes, such as uranium and other actinides, collect on the cathode. These elements are then sent to the cathode processor for further refinement, where any remaining salt is returned to the bath. A schematic of the entire process is shown in Fig. 2.9.

### 2.2.2 Separation from Natural Decay Chains

Another method of radioisotope production involves taking advantage of the three radioisotope decay chains found naturally in the environment. These three decay chains are called the uranium, actinium, and thorium series, and have initial isotopes consisting of uranium-238, uranium-235, and thorium-232, respectively. Usually, these decay chains are approximated as being in “secular equilibrium,” a state in which the activity of each radionuclide in the series is approximately equal. To satisfy the criteria for this definition, each decay chain must have an initial radioisotope with a much longer half-life than any of its following decay products. Additionally, a long time period must have elapsed to allow for the growth of the subsequent radioisotopes. A good approximation for this span is about ten half-lives of the longest living decay product [37].

During each decay process the original radioisotope, known as the “parent,” decays through either alpha or beta particle emission. With the decay particle ejected, the remaining radioisotope is of the form of the new nuclide, known as the “daughter.” This daughter product is more stable than the parent, although it can still decay further. A few of these daughter isotopes are strong gamma emitters. In the context of decay chains, however, gamma emission is not in the same category as an alpha or beta emission. This is because it is the release of excess energy, and by itself does not transform the radionuclide it was emitted from [37].

As was mentioned previously, the first decay chain, known as the uranium series, has an initial parent consisting of U-238. This radioisotope has a half-life of 4.5 billion years. Its decay daughter is Th-234, which is achieved through the emission of an alpha particle. The Th-234 has a half-life of 24 days, during which it emits a beta particle and transforms into Pa-243 m. Pa-243 m is a metastable state, with a half-life less than 1.25 min (75 s). As a parent, Pa-243 m emits a beta, returning the nuclide to an isotope of uranium (U-234). The nuclide then undergoes five consecutive alpha decays. These decays consist of U-234 (with a half-life of 240,000 years) to Th-230, Th-230 (with a half-life of 77,000 years) to Ra-226, Ra-226 (with a half-life of 1,600 years) to Rn-222, Rn-222 (with a half-life of 3.8 days) to Po-218, and Po-218 (with a half-life of 3.1 min) to Pb-214. This radioisotope has a half-life of 27 min and has the decay daughter Bi-214, achieved by beta emission. The bismuth nuclide, having a half-life of 20 min, quickly decays via another beta particle, transforming itself into Po-214. Due to an exceedingly tiny half-life of only 160  $\mu$ s, the Po-214 decays almost instantaneously to Pb-210 through the emission of an alpha particle. Over its 22 year half-life, Pb-210 emits a beta particle, converting the nuclide into Bi-210. Bi-210, with a 5 day half-life, decays via beta emission to Po-210. Lastly, Po-210 emits an alpha particle during its 140 day half-life, transforming into Pb-206. Pb-206 is stable and does not decay further. This entire sequence can be seen in Fig. 2.10.

The actinium series, which is the second decay chain, is shown in Fig. 2.11. Its initial parent is U-235. U-235 is a long-lived isotope, with a half-life of 700 million years. Its decay daughter is Th-231, which is reached through the emission of an alpha particle. The Th-231 has a half-life of 26 h, during which it emits a beta particle and transforms into Pa-231. Pa-231, with a 33,000 year half-life, decays via alpha emission to Ac-227. Interestingly, Ac-227 emits both alpha and beta particles during its 22 year half-life, creating two separate decay paths. Approximately 99 % of the time, Ac-227 decays via a beta particle, yielding Th-227. The Th-227 has a half-life of 19 days, during which time an alpha particle is emitted, resulting in Ra-223. In the other 1 % of the time, Ac-227 decays via an alpha particle, yielding Fr-223. Fr-223, with a 22 min half-life, emits a beta particle, also resulting in Ra-223. The nuclide then undergoes three consecutive alpha decays. These decays consist of Ra-223 (with a half-life of 11 days) to Rn-219, Rn-219 (with a half-life of 4 s) to Po-215, and Po-215 (with a half-life of 1.8 ms) to Pb-211. This radioisotope has a half-life of 36 min and has the decay daughter Bi-211, achieved by beta emission. The bismuth nuclide, having a half-life of 2.1 min, quickly decays via an alpha particle, transforming itself into Tl-207. Lastly, Tl-207 emits a beta particle during its 4.8 min half-life, transforming into Pb-207. Pb-207 is stable and does not decay further.

Last is the thorium series, which has an initial parent consisting of Th-232. This radioisotope has a half-life of 14 billion years. Its decay daughter is Ra-228, which is achieved through the emission of an alpha particle. The Ra-228 has a half-life of 5.8 years, during which it emits a beta and turns into Ac-228. Ac-228, with a half-life of 6.1 h, releases a beta particle, returning the nuclide to an isotope of thorium (Th-228). The nuclide then undergoes four consecutive alpha decays.

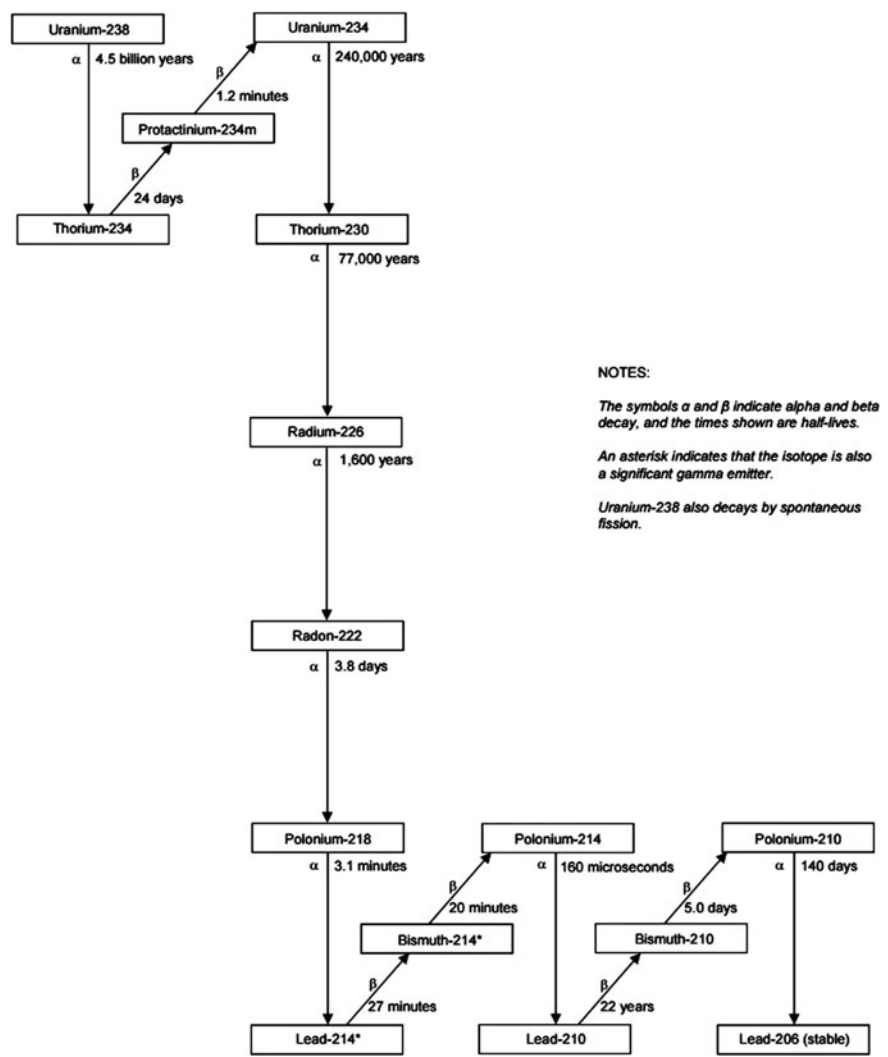


Fig. 2.10 The uranium decay series [37]

These decays consist of Th-228 (with a half-life of 1.9 years) to Ra-224, Ra-224 (with a half-life of 3.7 days) to Rn-320, Rn-320 (with a half-life of 56 s) to Po-216, and Po-216 (with a half-life of 0.15 s) to Pb-212. This radioisotope has a half-life of 11 h and has the decay daughter Bi-212, achieved by beta emission. Similar to the actinium in the actinium series, Bi-212 emits both alpha and beta particles during its 61 min half-life, creating two separate decay paths. Roughly 64 % of the time, Bi-212 yields Po-212 through beta particle decay. The Po-212 has an almost instantaneous half-life of only 310 ns, during which time an alpha particle is emitted, resulting in Pb-208. In the other 36 % of the time, Bi-212 emits an alpha

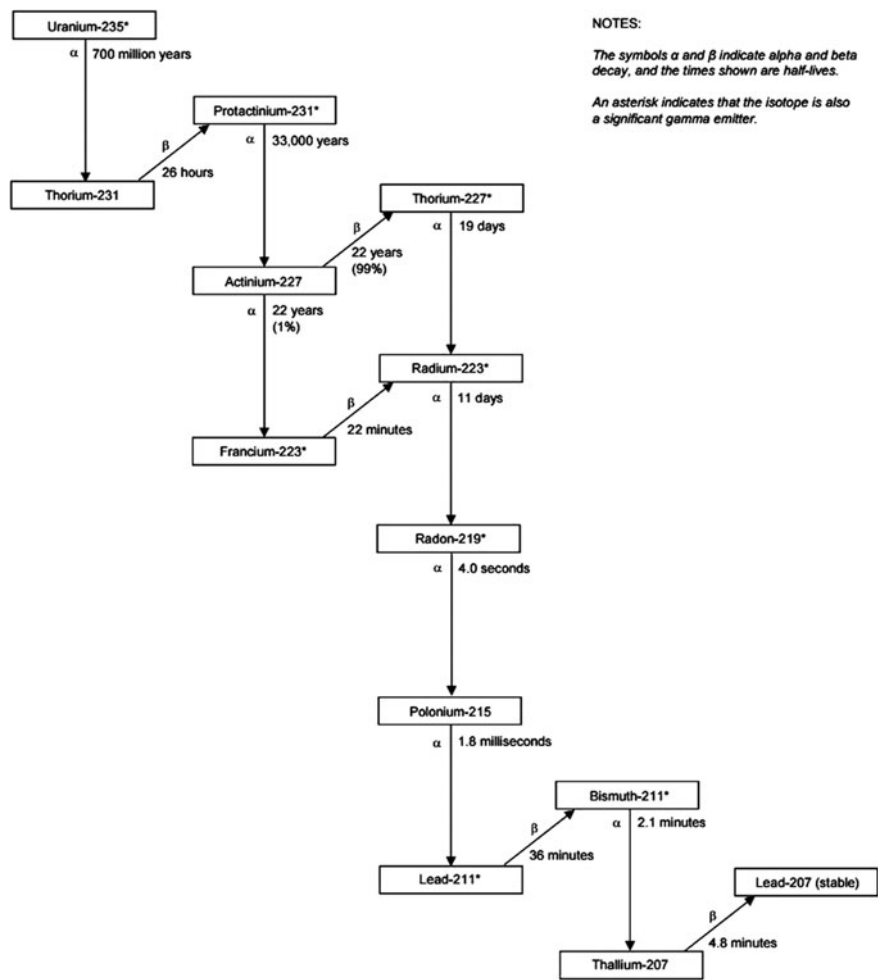


Fig. 2.11 The actinium decay series [37]

particle, giving Tl-208. Tl-208, with a half-life of 3.1 min, emits a beta particle, also resulting in Pb-208. Pb-208 is stable and does not decay further. A diagram of the thorium decay series is shown in Fig. 2.12.

One factor that can significantly disrupt the secular equilibrium of uranium, actinium, and thorium decay chains is whether or not specific radionuclides have been extracted from them. If extraction of a particular isotope has occurred to a considerable extent, its absence will inhibit the formation of its daughter isotopes, effectively severing the decay chain. The remaining daughter isotope sub-series can then be considered a separate (and smaller) series, with one of the long-lived radioisotopes from the original decay chain as its initial parent. For example, if U-238 was extracted from the uranium series in Fig. 2.10, the immediate daughter

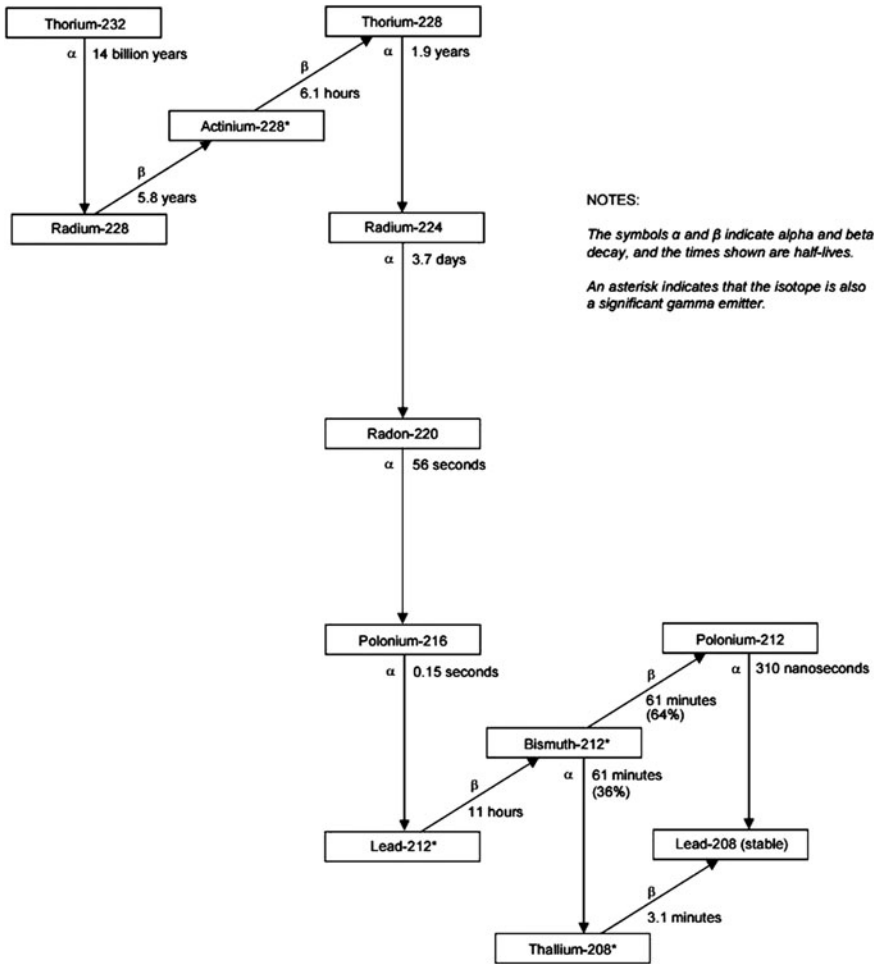


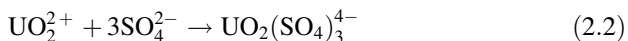
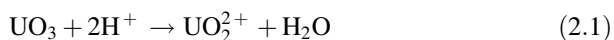
Fig. 2.12 The thorium decay series [37]

product (Th-234) would not continue to form. With relatively short half-lives, both the remaining Th-234 and its daughter Pa-243m would decay out. This would leave the long-lived radioisotope U-234 as the initial parent for a smaller chain, consisting of the subsequent daughters of U-234 originally included in the uranium series. Similarly, if U-234 was extracted from the uranium series, the Th-230 daughter would cease forming. However, since Th-230 itself is a long-lived radioisotope, it would become the initial parent for the remaining isotopes in the decay chain. Other nuclides in the uranium series that can be considered initial parents for smaller chains are Ra-226 and Pb-210 [37].

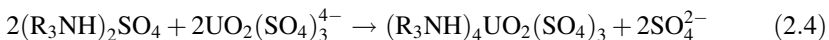
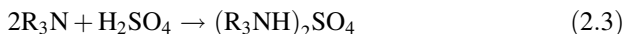
In determining the initial parent for a smaller series, nuclides with half-lives of less than a year are usually not considered. This is because these radioisotopes are

heavily dependent on the isotope supply generated by their parent nuclide, and so reestablish equilibrium with their parent (decay out) within several years [37]. Applying this consideration to the actinium series, it can be determined that it contains the initial parents U-235, Pa-231, and Ac-227. Similarly, applying this consideration to the thorium series reveals the initial parents Th-232, Ra-228, and Th-228.

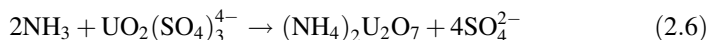
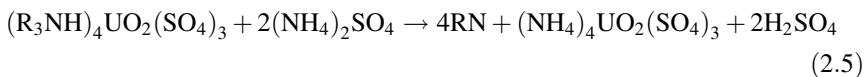
Once an isotope from one of the natural decay chains has been selected for extraction, a chemical separation process is applied to remove the nuclide from the other radioisotopes in the corresponding decay series and manipulate it into a useful form. The exact details of this process depend on the properties of the radioisotope being separated. In the case of uranium, crushed uranium ore ( $\text{UO}_3$ ) is leached with sulfuric acid ( $\text{H}_2\text{SO}_4$ ). The resulting oxidation reaction is [38]:



The uranium solution is then recovered using a resin/polymer ion exchange or liquid ion exchange solvent extraction system. To continue processing, the uranium is stripped from the ion exchange resin/polymer with either a strong acid or chloride solution, or with a nitrate solution in a semi-continuous cycle. In the chemical equation below, an acid is used. The “R” is an alkyl (hydrocarbon) grouping with a single covalent bond [38]:



After the previous reaction, any cation impurities are removed from the solution by the use of sulfuric acid, and anion impurities are removed from the solution by utilizing gaseous ammonia. An ammonium sulfate solution is then introduced, removing the alkyl (hydrocarbon) groupings from the uranium. Once this is completed, gaseous ammonia ( $2\text{NH}_3$ ) is added to the solution. This is done to neutralize the solution and create ammonium diuranate [38]:

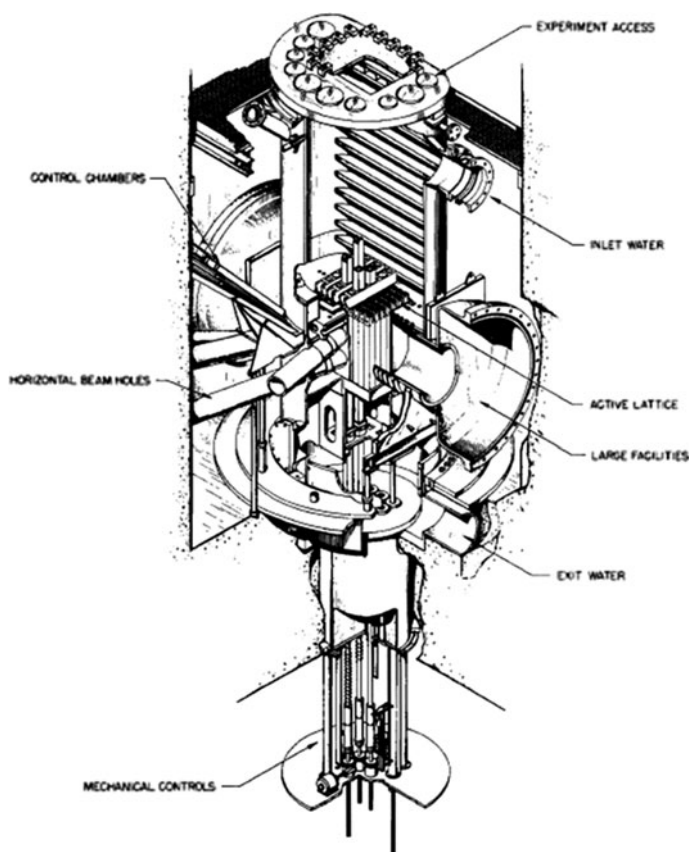


Lastly, the ammonium diuranate is processed. Any remaining water is removed from the solution, and then the ammonium diuranate is then dried at a high temperature [38]. This converts the product to uranium oxide ( $\text{U}_3\text{O}_8$ ), which contains approximately 85 % uranium by mass.  $\text{U}_3\text{O}_8$ , more commonly known as yellow-cake uranium, is then sold to customers.

### 2.2.3 Production by Neutron Capture in a Reactor

A third method, which is commonly used today, is the production of radioisotopes through neutron capture in a nuclear reactor. There are still a few national labs in the United States with this capability. The two current typical methods of neutron exposure to a sample are beam port exposure and pneumatic tube. It is also possible to have a loading dock which remains a stationary area during operation above the reactor, such as demonstrated in the Oak Ridge National Laboratory diagram (Fig. 2.13).

Each research reactor has very different properties associated with the key factors in irradiation (Table 2.7). These key factors include neutron flux, operation time, and volume available for sample exposure. Also important to note is that extra “dead time” occurs due to sample handling procedures and sample loading. This



**Fig. 2.13** Oakridge National Lab diagram of Low-Intensity Testing Reactor. The horizontal beam holes connect to beam ports and the large facilities near the *active lattice* show closer exposure areas

**Table 2.7** Properties of reactor facilities by laboratory

Reactor name	Fast flux (n cm <sup>-2</sup> s <sup>-1</sup> )	Thermal flux (n cm <sup>-2</sup> s <sup>-1</sup> )	Operational time (d year <sup>-1</sup> )	Max volume (cm <sup>-3</sup> )
MIT—**RR	1.2 × 10 <sup>14</sup>	6.0 × 10 <sup>13</sup>	168	460
MURR	6.0 × 10 <sup>14</sup>	6.0 × 10 <sup>14</sup>	180	350
INL **ATR	5.0 × 10 <sup>14</sup>	1.0 × 10 <sup>15</sup>	180–195	15,400*
ONRL Reactors	5.0 × 10 <sup>14</sup>	1.5 × 10 <sup>14</sup>	270–290	14.5 (f) 3.2 (th)

\*Note that INL has an operation port that has a tremendous available volume relative to pneumatic tubes present in other operations. \*\*RR is the abbreviation of Research Reactor, as *MURR* is University of Missouri Research Reactor and *ATR* is Advanced Test Reactor [39–41]

dead time can extend up to 3 or 4 times the exposure time for pneumatic tubes or be as low as 20 % for beam ports/staging areas.

The operational time above is gross of down time, the actual transitioning of material between runs. Thus, effective exposure could be as low as 5 % of the reactor operating time or as high as 70 %. Future calculations will involve an assumption of 50 %.

**2.2.4 Production by Accelerator**

The last method of radioisotope production utilizes a particle accelerator. In these accelerators, beams of charged particles (such as protons) are accelerated by an external magnetic field (see Fig. 2.14). When the appropriate energy level has been reached, these particles are then deliberately collided with target isotope atoms. During the collision, the atom absorbs the particle, transforming itself into a new isotope while releasing subatomic particles. After sufficient irradiation, the target isotope can be removed from the accelerator, and the desired radionuclide can be processed out for use.

One example of particle accelerator based radioisotope production is the Brookhaven Linac (linear accelerator) Isotope Producer, otherwise known as BLIP. This accelerator is located at Brookhaven National Laboratory (BNL) in Upton, New York, and operates by accelerating protons. Since opening in 1972, BLIP has received two upgrades: One in 1986 and the second in 1996 [43]. As shown in Fig. 2.15, BLIP works by diverting some of the particle beam produced by a previous linac. The length of BLIP from the diverging point from the previous linac to the target at the end is 98.4 feet. Target isotopes are formed into round “discs” and are placed inside a vertical 16 inch diameter containment shaft that is filled with water. They are then lowered down approximately 30 feet until they are in line with the incoming particle beam from the linac. Depending on particle beam strength and disc thickness, up to eight target isotopes can be inserted at a time [43]. Currently, BLIP can utilize proton beams with energies of up to 200 MeV, as well as up to 110 μA intensity [44].



Fig. 2.14 A linear accelerator (LINAC) at Brookhaven National Laboratory [42]

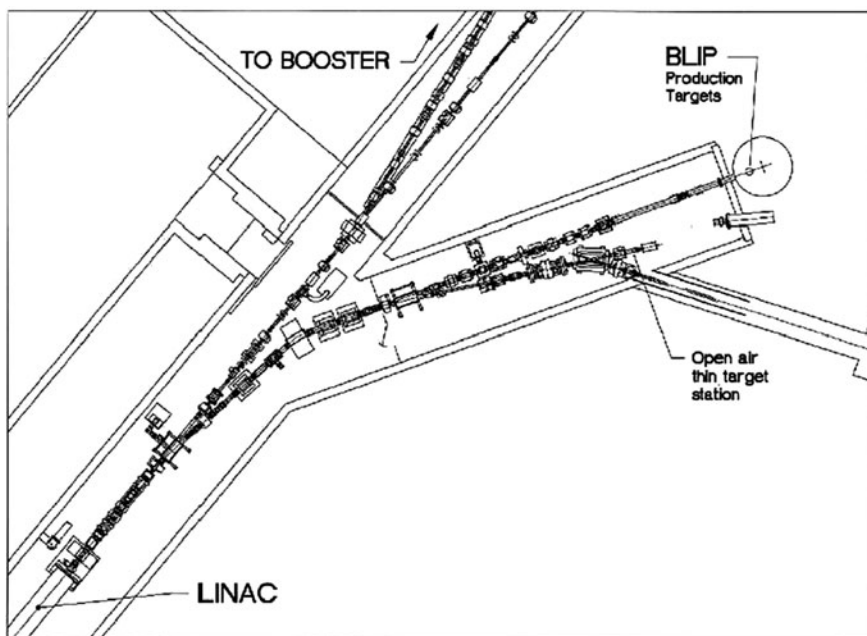


Fig. 2.15 BLIP Beam Line Schematic [42]

Using BLIP, Brookhaven National Laboratory has the capability to produce several radioisotopes. This is illustrated in Table 2.8, which shows the radioisotope production capability for the year 2012. As can be seen from the table, obtaining certain isotopes (such as Be-7 or Fe-52) required placing a special order with BNL. Other radionuclides, such as Ni-63, were readily in stock. Some isotopes were produced monthly (such as Cu-67 and Y-86), while others (such as As-73 and Tc-95 m) were not scheduled to be produced that year. One interesting note is that, for the 2012 radioisotope production year, BLIP was only run from January through July [44]. This means that the longer lived nuclides were available all year, while the supply of shorter lived nuclides quickly died out after BLIP was shut down for the year.

After the desired radioisotopes are produced, they still must be separated from any coproduced isotopes, as well as other target materials. Fortunately, BNL has nine hot cells, eight radiochemistry development laboratories, an instrumentation laboratory for radionuclide and chemical assays, and radioactive waste handling and storage facilities that all enable it to do the required isotope post-processing [43]. In these facilities, BNL employs several radiochemical processing techniques. These methods include ion exchange chromatography, solvent extraction, distillation, and precipitation [43].

A second example of radioisotope production via a particle accelerator is Niowave, a super conducting linear accelerator company located in Lansing, Michigan. Unlike with BLIP (which uses protons), the Niowave super conducting linac accelerates electrons. It operates at only 4 K ( $-452^{\circ}\text{F}$ ) and can produce

**Table 2.8** BLIP radioisotope production capability in 2012 [44]

Isotope	Half-life	Primary usage	Production frequency
Be-7	53.3 days	Research	Special order
Mg-28	20.9 h	Research	Special order
Fe-52	8.3 h	Research	Special order
Fe-55	2.73 years	Commercial	In stock
Ni-63	100.1 years	Commercial	In stock
Zn-65	243.8 days	Commercial	In stock
Cu-67	61.9 h	Research	Monthly
Ge-68	270.8 days	Commercial	Monthly
As-73	80.3 days	Research	Not scheduled
Sr-82	25.4 days	Commercial	Monthly
Rb-83	86.2 days	Research	In stock
Y-86	14.7 days	Research	Monthly
Y-88	106.6 days	Commercial	Not scheduled
Tc-95 m	61 days	Research	Not scheduled
Tc-96	4.3 days	Research	Not scheduled

electrons from 20 to 40 MeV, with the particle beam power ranging from 50 to 100 kW in intensity [45]. In 2014, construction began for a linac facility specifically designed for radioisotope production. When completed in 2015, Niowave intends to pursue production of medical radionuclides Mo-99, Cu-67, and Ac-225. It also plans on investigating production of industrial isotopes Mn-54 and Zn-65 [45].

## 2.3 Cost of Radioisotopes

There are many factors that impact the cost of radioisotope production discussed in this section. The Nuclear Energy Agency OECD 2010 report reviewed production costs of Mo-99 and Tc-99 m. Encompassing all factors can be dissolved down into a per unit cost by the equation:

$$P_{\alpha} = \frac{\sum_t \left( \frac{I_t + O\&M + D_t}{(1+r)^t} \right)}{\sum_t \left( \frac{\alpha_t}{(1+r)^t} \right)} \quad (2.7)$$

In this notation,  $\alpha$ , is the particular radioisotope to be produced; ' $t$ ' is a set particular time, and is summed over all effective times; ' $I$ ' is the investment costs, typically taken at  $t = 0$  but will include interest payments on loans; ' $D$ ' is the decommissioning cost of any facility; ' $O\&M$ ' the operating and maintenance costs at a given time; and ' $r$ ' is the interest rate (to be considered constant, but it also can be dynamic in time). The following are key factors influencing these costs from a per unit basis. Production rates and particular stoichiometry for chemical/nuclear processes are proprietary to each industry, and thus cannot be presented in a reliable fashion. Instead, this section will conclude with previous production costs as a reference point for future work.

### 2.3.1 Cost of Separation

Separation steps exist in every method of source material creation for nuclear batteries. In neutron capture processes, source material is purified typically using an acid and a liquid-liquid extraction step [46]. Accelerator targets are processed in a similar manner. Of particular interest though would be the anticipated cost of a liquid-liquid and liquid-solid extraction processing plants. These are typical designs similar to expected equipment used to harvest material from spent nuclear fuel and convert it to source material.

Separation costs are the composite of capital equipment necessary and processing materials needed. The primary processes involved are: medium based

liquid-liquid extraction, centrifuge separation, dissolving and mixing, distillation, and drying. While the discussion present is a cost guide to extraction of radioactive material from spent nuclear fuel, pieces of the overall process can be extracted to specific methods such as PUREX.

### 2.3.1.1 Cost Analysis

The first consideration in cost analysis is consultation of reactants and recyclable chemicals. While exact stoichiometry and chemical engineering principles are proprietary to companies such as AREVA and their operations in France, by studying operations at the La Hague facility or the US nuclear weapons reprocessing facility at Hanford, WA, one gathers typical chemical reactants used (Table 2.9).

Typically, there are multiple physical modifications steps needed to achieve desired conditions for chemical reactions, thus the discussion begins with Dissolution, Mixing, and Drying Capital Costs.

### 2.3.1.2 Dissolution, Mixing and Drying Equipment

Dissolution, mixing, and drying are the bracket steps to major processes of distillation, liquid-liquid extraction, and centrifuging. Typically, manufacturing of custom capital equipment takes place. But there are major items associated with these processes; equipment such as mixers, dryers, containers, and extraction columns. A facility may need multiple iterations of similar equipment, which cuts down on manufacturing costs (Table 2.10).

**Table 2.9** Typical process materials cost for PUREX

Reactant name	Quantity	Unit	Costs (\$)
Nitric acid	15	L	382.5
Sodium carbonate	12	kg	492.7
H <sub>3</sub> PO <sub>4</sub> —85 % wt	25	kg	234
Sodium bismuthate	100	g	276.5
Sodium dichromate	5	kg	556
Zirconium	100	g	82.7
Cerium in ammonium cerium nitrate	500	g	267.5
Hexone (methyl isobutyl ketone) *98 %	100	ml	30
Sodium hydroxide *97 %	50	kg	971
Tributyl phosphate—TBP	500	ml	174
Kerosene	3.78541	L	3
Gallium	1	kg	339

Note these prices were sourced to be minimums from multiple online and retail sources. They are listed by reactant name, the quantity and unit for the associated cost [47–50]

**Table 2.10** The following are industrial quotes listed as averages from major producers and users of these industrial equipment (Flour, Sulzer, URS)

Device	Cost estimate (\$k)
Industrial dryer	5.00
Packed extraction columns (ECP)	1,000.00
Mixer settler column	2,500.00
Organic solvent nanofiltration	3,000.00

### 2.3.1.3 Sunk Costs Considerations

Referring to the equation at the start of the section it is important to note that investment costs must be considered at time zero. There are instances where these costs can get out of control, resulting in the death of a project. Additionally, in the early studies of nuclear fuel separation the following situation occurred resulting in the destruction of the facility and need for a full redesign.

Nuclear fuel separation occurs via the metal being dissolved in a significant high concentration of acid solutions in preparation for liquid-liquid extraction. Slight oxidation of the metal can occur in long periods of storage. Direct mixing with the TBP had resulted in disaster when the storage container was stainless steel. Via these disasters it is now known that a polymer lining is required in the storage vessel before the combination step. It prevents the violent redux reaction of any oxidized metal in the acid interacting with “TPB,” and removes the possibility of explosive gas generation during storage [51].

### 2.3.2 Cost of Neutron Capture

Projecting the cost of neutron capture is a multi-step process, but can be simplified by dividing the total annual operating and fuel costs by the potential amount of neutrons captured by a given reference material.

$$\frac{C}{n} = \frac{C_{op} + C_{fuel}}{n} \quad (2.8)$$

Note the above formula requires costs to be taken on the same time basis. This investigation takes into account annual operational and fuel costs. Referencing one of the authors, research paper, typical fuel cost for research reactors is roughly \$20 M per year. Additionally, a typical reactor will need a minimum of 50 personnel for operations and primary safety precautions. The median salary in the industry is roughly \$70,000 while the average is closer to \$100,000; the average will be used for conservatism in the estimate [52, 53]. Thus in a year a typical research reactor would cost roughly \$25 M annually for operations and fuel. Typically neutrons available are given in terms of available flux  $\phi$ , so total neutrons:

**Table 2.11** Note that is table represents cost per flux and based on parameters given in Table 2.10

Neutron costs	Fast ( $10^{-14}/(\text{n cm}^{-2})$ )	Thermal ( $10^{-14}/(\text{n cm}^{-2})$ )
MIT-RR	\$2.87	\$5.74
MURR	\$6.43	\$0.53
INL-ATR	\$0.64	\$0.32
ONRL-Reactors	\$1.44	\$0.65

$$n' = \varphi \cdot \Delta t \quad (2.9)$$

Note that  $\Delta t$  is exposure time of the material during operations. Assuming downtime of 50 % the cost estimates from published fluxes of test reactors around the country is shown in Table 2.11.

The number of neutrons though should be more formally analyzed as neutrons are captured. This requires significant number of assumptions and potentially some complexity. First, the neutrons need to be accounted for in terms of thermal and fast neutrons, which have different capture cross sections based on target materials. Second, one is required to take into account decay constants, exposure time, capture cross section, and other physical parameters. Given the complexity, it is easier to find the cost assuming some average parameters for materials and saturation level is reached, removing the specifics of decay effects. To be conservative the decay effects can be coupled with associated downtime of experiments and exposure.

$$n_{cap} = \frac{1}{2} [\varphi_{f,prod} \sigma_f + \varphi_{th,prod} \sigma_{th}] \cdot \rho \cdot V \cdot \Delta t \quad (2.10)$$

Note that sometimes the chambers available for thermal and fast production have different volumes,  $V$ ; where that factor would be distributed. Also density,  $\rho$ , is taken into account, but this equation has the capture cross section in per gram. Typically cross sections are listed on a per atom basis, so multiplying by Avagadro's number  $N_A$  and dividing by molecular weight should be multiplied into the factors. Doing so results in a cost per neutron captured for the test reactors.

Note significant assumptions about the density and capture cross section of materials are taken into account. The table assumes a material with a density of  $8 \text{ g cm}^{-3}$ , a molecular weight of  $80 \text{ g mol}^{-1}$  and fast cross section of 1 barn ( $10^{-24} \text{ cm}^2 \text{ atom}^{-1}$ ) and thermal cross section of 5 barns.

Unfortunately, most production methods for nuclear batteries would like material on the order of grams and not atoms. This requires to multiply the cost by the amount of neutron captures needed to produce a gram of radioactive material, which is on the order of  $10^{23}$ . Suddenly, the costs above reach the 100s–10s of millions per gram of source material produced, from a fuel cost basis. For mass production, beam port operations will be required and only the lower costs presented would be applicable. INL-ATR thermal and MURR thermal neutron production are based on beam port size calculations (Table 2.12).

**Table 2.12** Table is cost per neutron captured, assuming neutrons captured independently of other operations, and a factor of 2 is divided to conserve this operation

Neutron costs	Fast $10^{-17}/\text{n}$	Thermal $10^{-17}/\text{n}$
MIT-RR	\$103.68	\$41.47
MURR	\$232.25	\$3.87
INL-ATR	\$23.23	\$2.32
ONRL-Reactors	\$51.85	\$4.71

As an example of how to go about calculating the cost of isotope production using neutron capture consider the case of Ni-63. It has a half-life of 101 years and emits a beta particle with a maximum energy of 65.9 keV. The long half-life makes it promising for nuclear power battery applications—where long shelf-life is desired. Nickel-63 is produced via neutron capture of Nickel-62 ( $^{63}\text{Ni} (n,\gamma) ^{63}\text{Ni}$ ). The isotopic composition and neutron cross sections of natural nickel are shown in Table 2.13. The large neutron thermal absorption cross section (14.5 barns) as well as the resonance integral (6.6 barns) of Ni-62 increases the likelihood for neutron absorption to occur. A large fraction of natural nickel is Ni-58. To increase the specific activity of Ni-63, Ni-62 is enriched as high as possible and is irradiated with a high neutron flux.

The activity of the radioisotope produced during or at the end of a neutron capture reaction is calculated from the equation below

$$\frac{dN'}{dt} = nv\sigma_{act}N_T - \lambda N' \tag{2.11}$$

where  $N_T$  is the total number of atoms present in the target,  $nv$  is the neutron flux =  $\phi$ ,  $\sigma_{act}$  is the activation cross-section,  $N'$  is the number of activated atoms,  $A$  atomic weight of the target, and  $\lambda N'$  is the decay rate of the product.

The activity can be obtained by integrating the equation above resulting in the equation below.

$$A = \lambda N = \sigma_{act} \phi N_T (1 - e^{-\lambda t}) \tag{2.12}$$

**Table 2.13** Percentage composition of natural nickel isotope

Isotope	Ni-58	Ni-60	Ni-61	Ni-62	Ni-64
% abundance	68.07	26.22	1.14	3.63	0.93
Thermal neutron cross section (barns)	4.6	2.96	2.5	14.5	1.52
Resonance Integral (barns)	2.2	1.5	1.5	6.6	0.98

The above equation does not take into account the following which will reduce the calculated activity:

- Flux depression due to adjacent samples in the reactor especially when such samples are high neutron absorbers,
- Burn-up of the target material with time,
- Destruction of the product nucleus due to subsequent neutron capture,
- Self-shielding effect of the target,
- Power variation in the reactor, and
- Target encapsulation.

A final time,  $t'$  will elapse after irradiation and just before counting. Hence, Eq. 2.12 reduces to

$$A = \sigma_{act} \phi N_T (1 - e^{-\lambda t}) e^{-\lambda t'} \quad (2.13)$$

The quantity  $(1 - e^{-\lambda t})$  is called the saturation term. The equation shows that the activity grows exponentially and reaches a saturated value limited by the flux in the reactor. After sufficient period of irradiation ( $t \gg t_{1/2}$ ) the activity approaches the value

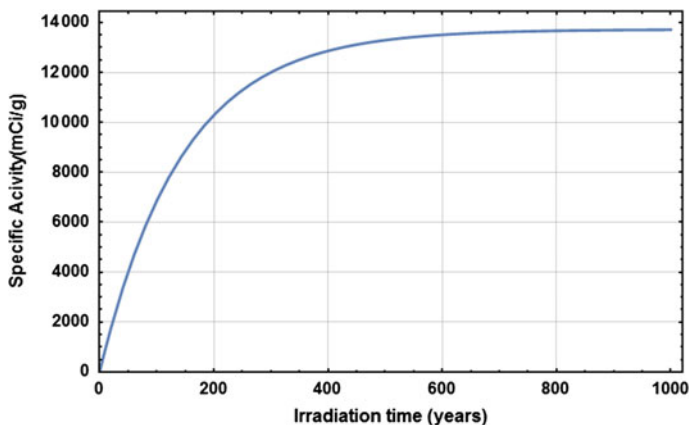
$$A_{sat} = \sigma_{act} \phi N_T \quad (2.14)$$

$A_{sat}$  is called the saturation activity. For relatively short irradiation time comparable to the half-life of the isotope, the saturated term approaches  $\lambda t$  and the activity varies linearly with time.

The neutron flux at the flux trap at the University of Missouri Research Reactor (MURR) is  $6.0 \times 10^{14} \text{ n} \times \text{cm}^{-2} \times \text{s}^{-1}$ . There are many positions within beam port of this reactor that are designated for experimental work. Targets could easily be inserted in and out of this positions during reactor operation. The flux at the H1 position is  $1.0 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$  [54]. Table 2.14 shows the nuclear reactions and calculated specific activity for Ni-63. To minimize the content of Ni-58 in the target, the target is made of mostly Ni-62. The small fraction of Ni-58 in the target would burn in the reactor to produce a small percentage of Ni-59. Figure 2.16 shows the specific activity, defined as the activity per unit mass, for the production of Ni-63 at the H1 position at different irradiation times. The maximum specific

**Table 2.14** Nuclear reactions and specific activity calculation for Nickel-63 at different irradiation times

Reaction	Isotope abundance	Cross section (barns)	Activity at $1.0 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ (mCi/g)		
			1 week	1 month	1 year
$^{62}\text{Ni} (n, \gamma) ^{63}\text{Ni}$	3.63 %	14.5	1.98	7.92	94.7



**Fig. 2.16** Calculated production yields of Nickel-63 by neutron irradiation of Nickel-62 targets as a function of irradiation time at a thermal neutron flux of  $1.0 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$

activity achieved is 13.7 Ci/g, obtained after a few half-lives. Due to the long half-life of Nickel-63, the amount of Ni-63 produced that decays before counting is negligible.

Secondary reactions and products are: (1)  $^{58}\text{Ni} (n, \gamma) ^{59}\text{Ni}$  [ $t_{1/2} = 7.6 \times 10^4$  years, abundance = 68.08%,  $\sigma = 4.6$  barns], (2)  $^{58}\text{Ni} (n, \alpha) ^{55}\text{Fe}$  [ $t_{1/2} = 2.73$  years  $\sigma \leq 1$  mb], (3)  $^{62}\text{Ni} (n, \alpha) ^{59}\text{Fe}$  [ $t_{1/2} = 2.73$  years,  $\sigma = 0.002$  mb], and (4)  $^{64}\text{Ni} (n, \gamma) ^{65}\text{Ni}$  [ $t_{1/2} = 2.51$  h, abundance = 0.9265,  $\sigma = 1.52$  b]

Ni-63 also has an appreciable cross section (24 barns) and is activated in reactors to produce Ni-64. This limits the specific activity of the Ni-63 produced. Generally, 10 g of Nickel-62 powder is encapsulated in an aluminum container with diameter of 4 mm and a height of 2 mm (50.27 mm<sup>2</sup> area) prior to irradiation. Irradiation is done between 8 and 12 weeks [46]. The cost per neutron at MURR has been indicated Table 2.12. Based on this value, the cost of producing 1 Ci/g of Ni-63 is estimated to be US\$4252. The cost of neutrons generally accounts for a small fraction of the total production cost.

### 2.3.3 Cost of Accelerator

Accelerators are used to produce medical isotopes. Medical isotopes are necessary for health related diagnostic imaging and therapy. For these medical related applications, isotopes can be priced at a premium. The diagnostic imaging application typically uses short lived isotopes. The therapeutic isotopes are longer lived. Medical isotopes are created by charged particle reactions (protons, deuterons, helium or neutrons) or a by a photonuclear process with a target material.

**Table 2.15** Cost of manufacturing Mo-99 using various processes

	Cost per Ci	Cost per gram
Reactor	\$1,010.00	\$484,000,000
Photo-nuclear (enriched target)	\$356.67	\$171,000,000
Photo-nuclear (natural target)	\$2,893.33	1,387,000,000
Dedicated cyclotron	\$1,216.67	583,000,000
Multipurpose cyclotron	\$766.67	367,400,000

To illustrate the costs of producing medical isotopes, one of the most studied, Mo-99, is used as an example. Mo-99 is one of the more prominent medical isotopes and has been widely studied [55, 56]. The processes studied included the use of low enriched uranium in a high flux nuclear reactor, the use of Mo-100( $\gamma$ ,n) Mo-99 reaction using an accelerator, or a neutron flux of  $10^{14}$  n cm<sup>-2</sup> s<sup>-1</sup> provided either by a multipurpose 30 MeV cyclotron or a dedicated cyclotron were costed out as shown in Table 2.15.

The high cost of producing a medical radioisotope is acceptable for medical procedures where 10s of mCi of the isotope are needed and the cost premium is worth it for the lifesaving benefits. However, these types of costs are not affordable for nuclear batteries.

New technologies are being developed based on linear accelerators [57]. There are some developments in superconducting linear accelerators. These new approaches and new developments will not have a significant impact on price.

## 2.4 Other Factors Influencing Cost

### 2.4.1 Safety

The costs associated with safety are discussed in further detail in Sect. 5.3.1 of Chap. 5 where regulation based on particular nuclides is discussed. Typically, there will be resident experts whose salaries are in the range of \$90,000–\$300,000 based on qualification and licenses attributed to that individual. Additionally, special precautions such as transportation effects are required.

### 2.4.2 Software

It typically is important to consult the industry standards of modeling tools and regulatory code. A leader in chemical engineering process software is Aspen Technologies, and their license can range from \$5,000 to \$20,000 annually per user depending on detail of specifications needed (quote from call). In the case of nuclear operations, the consent and royalties to LANL, for use and modification of MCNP can be in excess of \$50,000, depending on size of the company.

### **2.4.3 Liquidity of Capital (Cash)**

A big factor recognized in the initial equation is the current interest rate, 'r'. Historically lending of this scale requires that significant interest costs are at a significant premium to central bank rates. Currently, the world is seeing record low interest rates, where the 10 year treasury is going for 1.7 %. The premium for a capital project of this magnitude and risk would typically go for slightly higher rates than that of owning a home, currently 4.3 %. Many companies will issue convertible debt or preferred stock to fund these operations, and yields can be as costly as 10 %. Additionally, should circumstances change or problems come up, it would become necessary to borrow and refinance the loans. In a tight market, noticed in 2008, such costs could be as high as 17 %. Most industrial models will anticipate between 6 and 8 % interest costs, but timing certainly plays an effect in project costs [58].

## **2.5 Isotopes Produced from the Manhattan Project**

The technologies and capabilities available for nuclear battery source material are firmly established, but they are extremely costly, difficult to organize, and filled with regulation. Due to the risks, there are virtually no profitable uses for nuclear energy outside the scale of reactors or premium of medicine. During the beginning of the nuclear era there were a lot of parts of the Manhattan Project and recycling at Hannover, WA was one of those parts. The costs of nuclear fuel separation were a part of the technological development and were assumed by the U.S. government as a means to an end for producing weapons grade plutonium. While this progressed the science, it did not provide good insight into creating a business model for operating profitable sourcing of other radioisotopes [59].

## **2.6 Mixed Oxide Fuel Fabrication Facility (MOX FFF)**

In 1999 the US government approved joint operation of a facility to reprocess spent nuclear fuel at the Savannah River Site in South Carolina. After 16 years and roughly \$8.5B the project is still 'underway' as certain engineering failures, disputes over regulations, and other things have impeded progress. The project has documented over 20 million safe work hours and will be able to process 3.5 metric tons of weapons grade plutonium into a mixed oxide fuel (MOX). MOX fuel contains plutonium and uranium and can be put into many typical reactor designs used today, with the only major change being timing of delayed neutrons. The

facility will have the capability to produce over 150 MOX assemblies a year, and anticipated to finally be online in 2020. All in costs, not including operation will be on the order of \$17B, but was initially projected at \$2-5B [59–61].

## 2.7 Summary

There are limited sources of radioisotopes. Isotopes can come from natural sources and from man-made sources (fission and through other nuclear reactions based on neutron capture, high energy interactions with charged particles or photonuclear). The issue with radioisotopes is the quantity of suitable isotopes that are available from natural sources or fission. These types of radioisotopes have to be separated from other materials which contribute to the cost of the isotopes. If the isotope must be made from nuclear reactions, then the cost of the neutron source, high energy charged particle source or high energy photon source becomes a significant factor.

Availability and cost of isotopes are a problem for the development of nuclear batteries.

## Problems

1. How long would it take for the world's supply of natural Th-234 to reach one gram (Note it is being replenished by a parent isotope)? How about Ac-227? Ra-228?
2. What would be the parent isotopes for sub-series created by extracting the following nuclides: U-238, Th-230, Ra-226, Pa-231, Ac-227, Ra-228, and Th-228?
3. Suppose a sub-series with a parent isotope of U-234 was discovered. What radioisotope would likely have been extracted? What if the parent isotope was Th-228?
4. Which radioisotopes mentioned in this chapter could be used in a nuclear battery?
5. Is Pb-210 a viable isotope for nuclear batteries? Explain your reasoning.
6. Is it feasible to make Pu-238 from the existing inventory of Np-237? Explain your reasoning.
7. Is Am-241 a possible replacement for Pu-238 in a Radioisotope Thermoelectric Generator?
8. How much Kr-85 does a 3 GW thermal commercial reactor with a 90 % capacity factor produce in one year?
9. Estimate how much it costs to produce a gram of Ar-39.
10. Estimate how much it costs to produce a gram of Gd-148.

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