Chapter 2

THEORY

2.1 Natural radioactive series

Among the natural elements, those with atomic number greater than 83 are radioactive. Almost half of these radionuclides belong to natural radioactive series. There are three main natural radioactive series, called the uranium series, thorium series and actinium series.

2.1.1 Uranium Series

This radioactive series begins with uranium-238 (U-238) which undergoes chain disintegration into more stable product atoms. U-238 has a long half-life (4.5×10^{9} years). There are 14 steps of the disintegration process which ends when stable lead-206 (Pb-206) forms.

2.1.2 Thorium Series

In this series, thorium-232 (Th-232) with the half-life of 1.42×10^{10} years transforms spontaneously into 10 steps and stops when stable Pb-228 forms.

2.1.3 Actinium Series

Uranium-235 is the parent radionuclide in this series. Its half-life is 7.14×10^{8} years. There are 14 steps of disintegration and the end product of the series is stable Pb-207.

It is concluded that the intermediate decay products of

these series, since they are short lived compared to their parents, exist naturally in equilibrium.[10,17]

The disintegration of these series are shown in Table 2.1-2.3.

Apart from the above radionuclides, K-40 is also one of the main primordial radionuclides. It is only 0.0118 percent of natural potassium but, because potassium is widespread in environment, K-40 is always found in nature. It has the half-life of 1.28×10^{9} years.

2.2 Gamma Radiation

After the decay of a nucleus by either alpha (α) or beta(β) decay, the resultant nucleus is often left in an exited state. Then it decays to the ground state by the emission of gamma-ray (γ).

Gamma-ray is an electromagnetic photon originating in the nucleus. It is emitted corresponing to the difference in the energy levels. In many cases, a series of γ rays are emitted and these are characteristic of the particular nuclide.

When gamma- rays interact with matter, there are three main processes involved: photoelectric effect, Compton scattering and pair production.

In the photoelectric effect, the photon ejects an electron from an inner shell and all the gamma ray energy is given to the electron. This effect is dominant for low gamma-ray energy.

For Compton scattering, only a portion of the gamma ray energy is given to the electron. Then a new photon of lower energy is created and scattered in a different direction from the original one.

Pair production process may occur if the gamma- ray energy is

Table 2.1: Decay chain of U-238

Element	Half-life	Principal gamma activity (energy in MeV and % bundance)
U-238	4.5×10^9 y	0.048(0.075%)
↓		
Th-234	24.1 d	0.063(5.7%), 0.093(6.8%)
↓		
Pa-234m	1.17 m	1.001(0.59%)
↓		
U-234	2.48×10^5 y	0.053(0.68%), 0.121(0.23%)
↓		
Th-230	7.7×10^4 y	0.068(0.4%)
↓		
Ra-226	1.6×10^3 y	0.186(4%)
↓		
Rn-222	3.82 d	0.51(0.07%)
↓		
Po-218	3.1 m	Nil
↓		
Pb-214	26.8 m	0.352(36%), 0.295(18.9%) 0.075(15%),0.242(7.6%)
↓		
Bi-214	19.7 m	0.609(46%)*, 1.764(15.8%) 1.120(13.6%), 0.285(5.2%) 1.378(4.7%)
↓		
Po-214	164μs	0.799(0.014%)
↓		
Pb-210	20.4 y	0.047(4%)
↓		
Bi-210	5 d	Nil
↓		
Po-210	138.4 d	0.803(0.0011 %)
↓		
Pb-206	Stable	

Data from Erdmann and Soyka (Julich, FG)

(* from CANBERRA MCA Series 35 Plus User Mannual)

Table 2.2: Decay chain of Th-232

Element	Half-life	Principal gamma activity (energy in MeV and % bundance)
Th-232	1.41×10^{10} y	Nil
↓		
Ra-228	5.75 y	Nil
↓		
Ac-228	6.13 h	0.911(25%)*, 0.969(13.3%), 0.964(3.7%), 0.339(9.3%), 0.795(3.9%)
↓		
Th-228	1.913 y	0.084(1.6%), 0.216(0.29%), 0.132(0.19%)
↓		
Ra-224	3.64 d	0.241(4.2%), 0.084(0.16%), 0.082(0.1%)
↓		
Rn-220	54.5 s	0.542(0.07%)
↓		
Po-216	0.15 s	Nil
↓		
Pb-212	10.6 h	0.239(43.1%), 0.300(3.27%), 0.077(17.5%), 0.075(9.6%)
↓		
Bi-212	60.55 m	0.727(6.5%), 0.040(1.1%), 0.785(1.1%), 1.620(1.51%)
↓		
Po-212	0.3 μs	Nil
↓		
Tl-208	3.1 m	0.583(30%*), 0.511(22.5%), 0.860(12%), 0.277(6.5%)
↓		
Pb-208	Stable	

Data from Erdmann and Soyka (Julich, FG)

(* from CANBERRA MCA Series 35 Plus User Mannual)

Table 2.3: Decay chain of U-235

Element	Half-life	Principal gamma activity (energy in MeV and % bundance)
U-235	7.1×10^{8} y	0.186(54%), 0.144(9.72%), 0.205(5%),0.163(4.6%)
↓		
Th-231	25.52 h	0.026(12%), 0.084(5.1%), 0.090(1.1%)
↓		
Pa-231	3.25×10^{4} y	0.303(2%), 0.300(1.9%), 0.284(1.3%), 0.330(1.09%)
↓		
Ac-227	21.8 d	0.995(0.028%)
↓		
Th-227	18.5 d	0.050(8.8%), 0.236(10.4%), 0.256(6%), 0.330(2.37%)
↓		
Ra-223	11.4 d	0.270(13.6%), 0.324%(4.0%), 0.154(5.6%),0.144(3.24%)
↓		
Rn-219	3.92 s	0.270
↓		
Po-215	1.8 ms	Nil
↓		
Pb-211	36.1 m	0.405(3.7%), 0.832(3.0%), 0.427(1.9%)
↓		
Bi-211	2.16 m	0.351(14%)0.405(3.0%) 0.832(3.3%)
↓		
Tl-227	4.77 m	0.898(0.27%)
↓		
Pb-207	Stable	

Data from Erdmann and Soyka (Julich, FG)

(* from CANBERRA MCA Series 35 Plus User Mannual)

greater than 1.02 MeV and the probability of this interaction increases exponentially with increasing of the gamma-ray energy. The incident photon is absorbed and a pair of electrons, one positive (so called "positron", e^+), and the other negative (so called "negatron", e^-), are produced. The positron, often lossing its kinetic energy, will combind with an electron in the surrounding resulting in the emission of two 0.511 MeV gamma-rays.

2.3 Radioactive Decay Law

The radioactive decay is the process which an unstable nuclide undergoes nuclear transformation to gain more stability. It is generally accompanied by the emission of charge particles and gamma ray. This process occurs in a random manner and is not influenced by any environmental, physical or chemical parameters. However, a probability of disintegration in a time interval can be stated. This probability of disintagration per unit time is called the decay constant (λ) and is characteristic of the particular mode of decay of that particular nuclide.

The disintegration rate of a radionuclide is proportional to the number of active nuclide present. Therefore, by using the notation of the differential calculus, we can write:

$$-dN/dt \quad \alpha \quad N$$

or

$$-dN/dt = \lambda N$$

then we get;

$$N = N_o e^{-\lambda t}$$

where N is the number of active nuclei still present at any time t, N_o is the original number present at time t= zero and λ is the decay constant.

The radioactivity (A) of a substance is the disintegration rate of a substance:

$$A = \lambda N$$

2.4 Chain Disintegration

In this process, a parent radionuclide decays to a series of unstable daughters until a stable product forms. In this case, the expression for the number of atoms of each species can be written as follows:

$$N_1 = N_o \exp(-\lambda_1 t)$$

Where N_o represents the original number of parent atoms

N_1 is the number of parent atoms having a decay constant ($\lambda 1$)

t is decay time

2.5 Equilibrium in Disintegration Process

2.5.1 Transient Equilibrium :

$$\lambda_2 > \lambda_1$$

where λ_1 = decay constant of the parent nucleus

λ_2 = decay constant of the daughter

In transient equilibrium, the half-life of the parent is longer than that of the daughter, but still not very long. In a

freshly purified parent fraction, the daughter activity builds up then decays with the same half-life as the parent. This relationship is illustrated graphically in Fig. 2.1.

At equilibrium :

$$A_2 = [\lambda_2 / (\lambda_2 - \lambda_1)] A_1$$

where A_1 = activity of the parent nuclei

A_2 = activity of the daughter

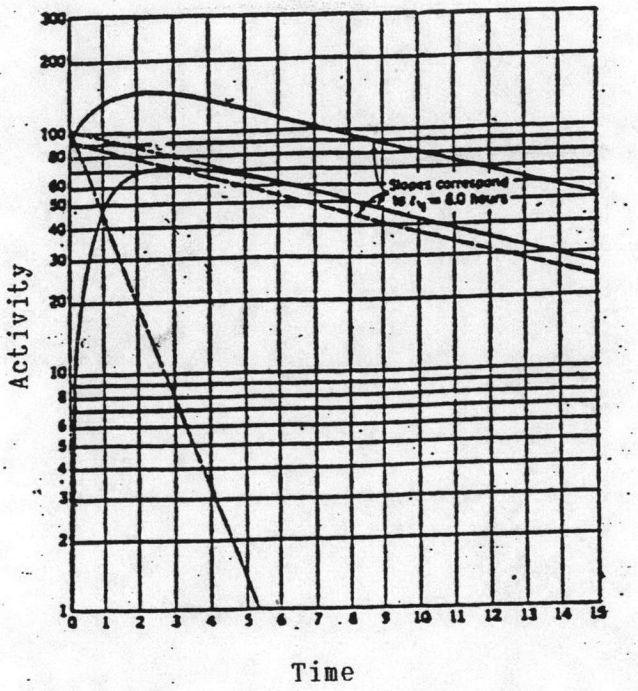

Figure 2.1 Transient Equilibrium of the Disintegration Process

2.5.2 Secular Equilibrium :

$$\lambda_2 \gg \lambda_1$$

In this case the half-life of the parent is much longer than that of the daughter. The buildup of activity in a freshly isolated parent fraction occurs as shown in Fig. 2.2.

At secular equilibrium :

$$A_2 = A_1$$

Thus, the activity of the daughter is equal to the activity of the parent.

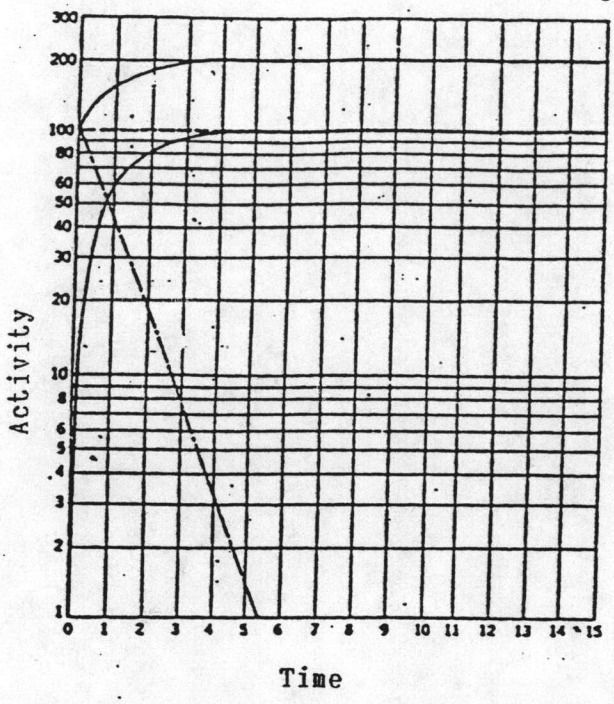

Figure 2.2 Secular Equilibrium of the Disintegration Process

2.6 Series Relationships for Naturally Occurring Radionuclides

2.6.1 U-238 and Ra-226 relationship (secular equilibrium)

At equilibrium : $A_1 = A_2$

Where A_1 = activity of U-238

A_2 = activity of Ra-226

or $\lambda_1 N_1 = \lambda_2 N_2$

or

$$(N_1 / N_2) = (T_1 / T_2) = (4.51 \times 10^9 \text{ yr}) / (1620 \text{ yr}) = 2.8 \times 10^6$$

Where N_1 and T_1 = number of atoms and half-life of U-238 respectively

N_2 and T_2 = number of atoms and half-life of Ra-226 respectively

In natural uranium, therefore, 1 part of radium is present to about every 3 million parts of uranium.

2.6.2 Ra-226 and Rn-222 Relationship

At equilibrium : $A_1 = A_2$

Starting with pure radium, it would take about 7 half-lifes of Rn (7 x3.8 = 27 days) for 99 % equilibrium to be established.

2.6.3 Rn-222 and Daughter Products Relationship

The daughter products of Rn-222 (i.e., Po-210, Pb-214 Bi-214 and Po-214) are short-lived compared to the parent. Starting with freshly purified radon, equilibrium will be established in four hours, after which approximately equal amounts of radon and daughters would be present.

2.7 High Resolution Gamma Spectrometry

Gamma spectrometry is a mean for identification and quantification of radionuclides present in a sample using the emitted gamma radiation.

A gamma spectrometric procedure in general comprises three steps : measurement, spectrum analysis and interpretation.

The measurement of a gamma radiation emitting sample results in a pulse height distribution, refered to as gamma-ray spectrum. In this distribution, the pulse height is proportional to the gamma-ray

The shape of the distribution is determined by the response function of the spectrometer. Major parts of this function are: full energy peak or photopeak, Compton continuum, escape peak and backscatter peak.

In high resolution gamma spectrometry with semiconductor or solid state detector, the positions of photopeaks and their areas are the bases for identification and quantification. These pieces of information are extracted from the gamma ray spectrum in the spectrum analysis step of the procedure. In the interpretation step the peaks are assigned to the relevant radionuclides and the activities of these nuclides in the sample are calculated.

A typical gamma spectrometer composes of a gamma-ray detector, electronics for analog processing the signals produced by the detector and a multichannel analyser (MCA) for digitizing the signals and spectrum accumulation. The general outline of a gamma spectrometer can be seen in Figure 2.3.

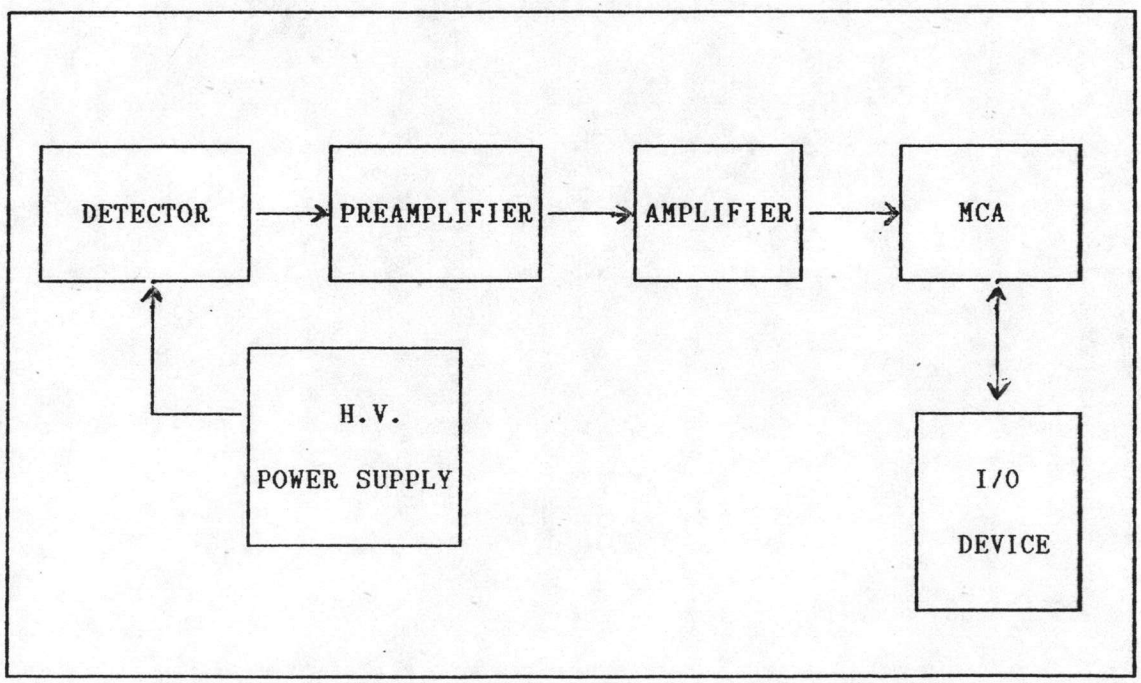

Figure 2.3 General Outline of a Gamma Spectrometry System

The energy calibration of a gamma spectrometer links channel numbers to gamma ray energies. This calibration has to be regularly repeated.

The efficiency calibration yields the efficiency curve which represent the relation between absolute photopeak efficiency and gamma ray energy. Figure 2.4 illustrates the typical absolute efficiency curve of a coaxial hyperpure germanium detector.

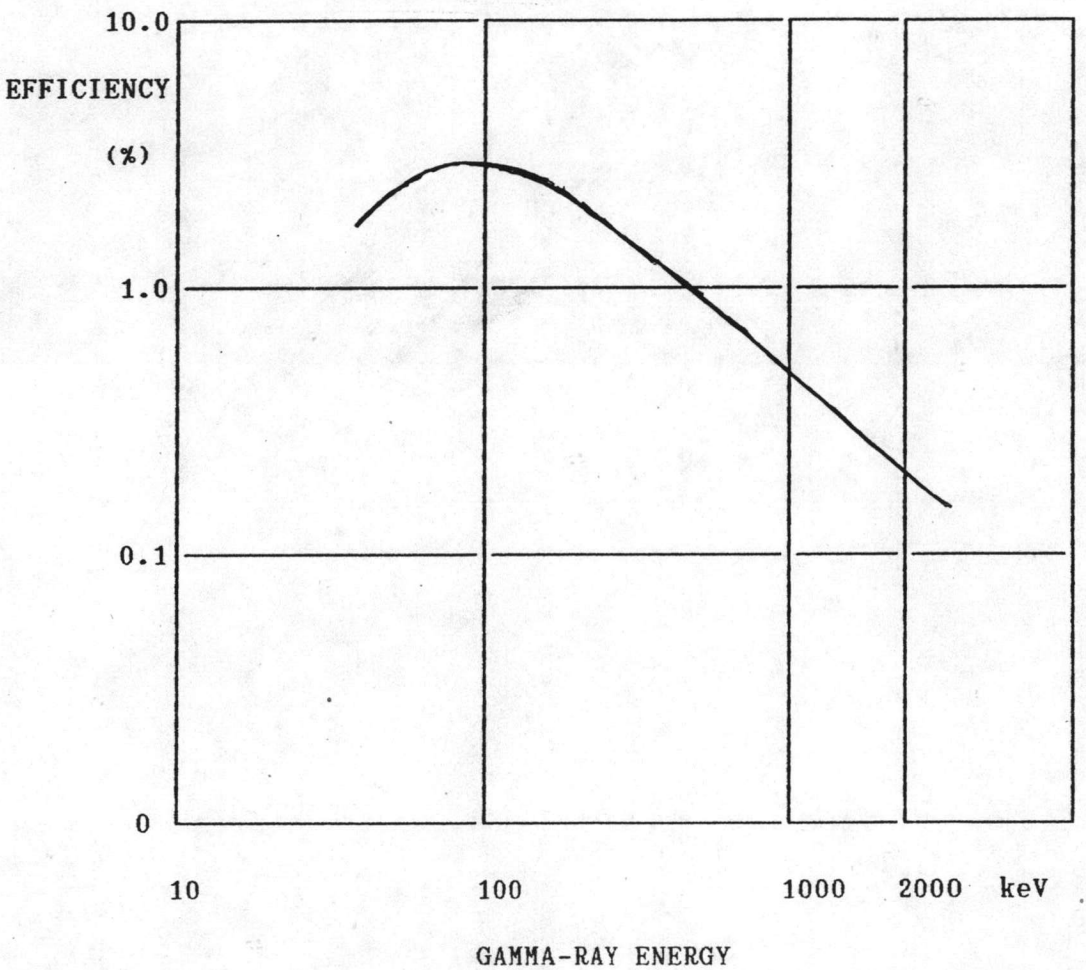

Figure 2.4 Typical Absolute Efficiency Curve for Coaxial Germanium Detector

2.8 Lower Limit of Detection (LLD)

The detection limit of any gamma spectrometer with germanium detector for gamma ray depends on the background of the same period of time to be expended on the measurement, and also on the stability of the electronics during the counting process. Background refers to both the natural background and Compton scattered events contributed by all detected radiations whose energies exceed the energy of interest. A useful rule of thum for just being able to detect a peak due to a gamma-ray of energy E is

$$N_1(E) - N_u(E) > 3[(N_u(E)]^{1/2}$$

Where $N_1(E)$ is the number of counts in the peak of interest, $N^u(E)$ is that of background while $(N_u(E))^{1/2}$ is the standard deviation in the background count. Thus, a detector limit R_a at T can be expressed as follows:

$$R_a = 3[(N_u)^{1/2}]/T$$

To indicate that a peak will be detected, an emission rate must be greater than $\sqrt{2}R_a$.