CHAPTER II

LITERATURE REVIEW

2.1 Alpha Decay

Alpha particles are heavy positively charge particles which are identical to the doubly ionized helium atoms(He^{++}). Each α-particle is composed of two protons and two neutrons. Therefore, in the disintegration of nucleus via alpha decay, the daughter nucleus has an atomic number(Z) two units less and a mass number(A) four units less than the parent nucleus. The nuclear disintegration may be represented as

$$ {}^{A}_{Z}X \longrightarrow {}^{A-4}_{Z-2}Y + {}^{4}_{2}He\ (\alpha) \qquad (2.1.1) $$

The daughter element ${}^{A-4}_{Z-2}Y$ will of course be chemically different from the parent ${}^{A}_{Z}X$

Alpha particles will travel only a short distance in matter before they come to rest. This property is due to the fact that they strongly ionize the meduim through which they pass. Thus their rates of energy loss are very large. The α-particles are monoenergetically emitted from the nucleus. Their range in air is between 3 cm and 7 cm depending on the energy with which they are emitted. The range can be calculated from the empirical relationship:

$$\bar{R} = 0.318 \quad E^{3/2} \qquad (2.1.2)$$

Where E is the energy in MeV and $\bar{R}$ is the mean range in cm. in air at 15°C and 760 mm Hg.

2.2 Neptunium Series (4n+1)

The naturally occurring radioactive elements fall into three series characterized by the integers 4n (thorium series), 4n + 2 (uranium series), and 4n + 3 (actinium series). All of the naturally occurring radioisotopes have A numbers equal to the above three series of numbers. Most of the naturally occurring radioactive isotopes occur among the elements with atomic numbers between Z = 81 and Z = 92. Elements with Z numbers above 92 are called transuranium since they lie beyond uranium in the periodic table. The first of these elements is the element neptunium with a Z = 93. The decay chain of neptunium -237 is shown in Figure 2.1. Because of its comparatively short half-life (2.2×10^6 years) of the neptunium series, the elements appearing in the series are not found in nature, as they have decayed away since the formation of the elements ($\sim 5 \times 10^9$ years ago). The first attempt to create ^{237}Np was made in the 1930's by bombarding uranium with neutrons. During the neutron irradiation, the uranium isotope undergoes fission which results in the production numerous radioactive isotopes.

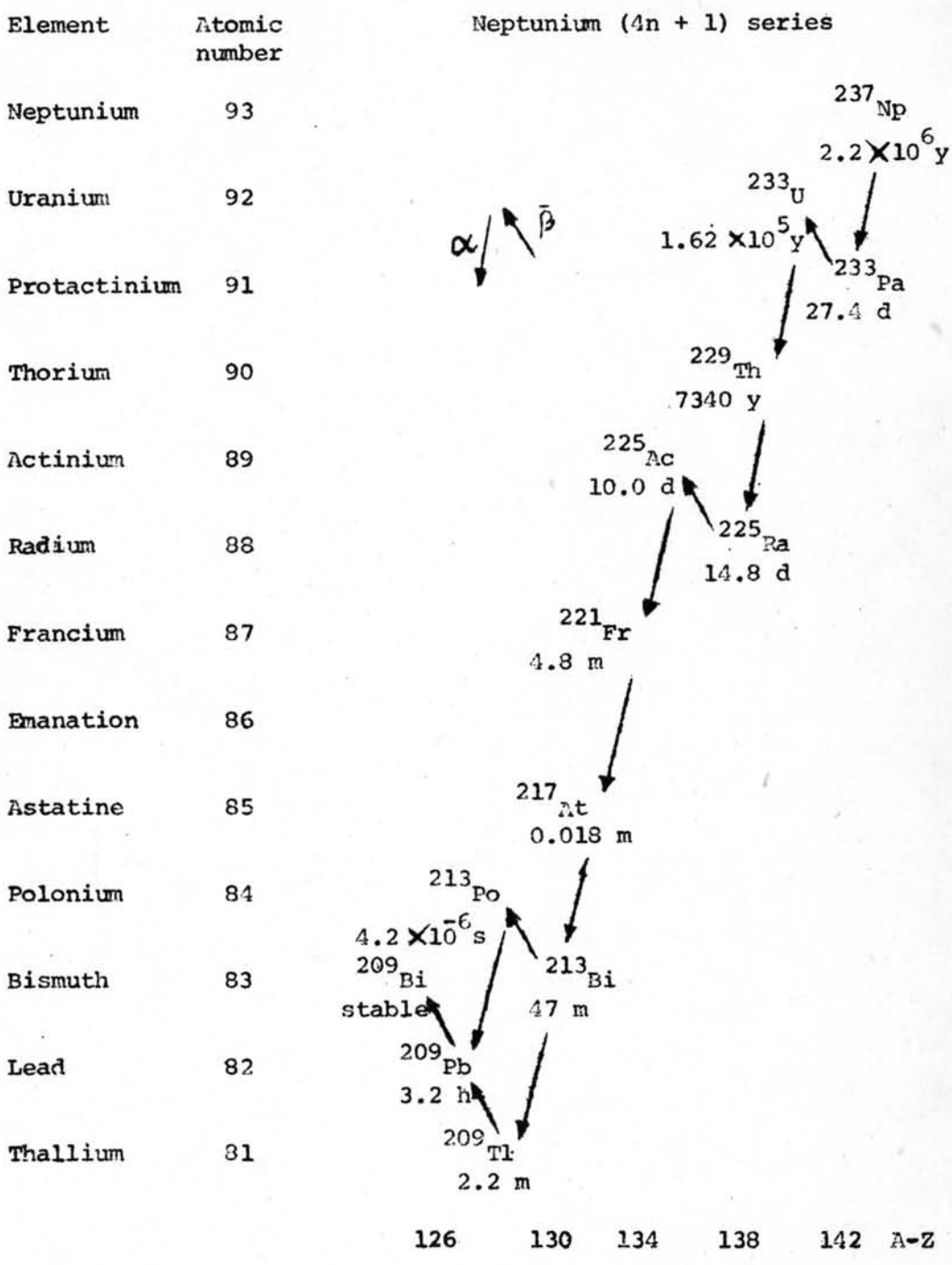

Fig. 2.1 The neptunium (4n + 1) series[4]

---

[4] Earl K. Hyde, Isadore Perlman, Glenn T. Seaborg, The Nuclear Properties of the Heavy Elements. Vol II: Detailed Radioactivity Properties (New Jersy: Prentice-Hall, Inc., 1964). p. 581.

2.2.1 Neptunium -237 ($^{237}_{93}Np$)

This neptunium isotope is the longest lived species of the element neptunium. ^{237}Np is an alpha-emitter with a half-life of about 2.2×10^6 years. It is stable against β -disintegration. It is produced as the decay product of β^- emitting ^{237}U in the reaction:

$$^{238}U\ (n,2n)\ ^{237}U \xrightarrow[7\ \text{days}]{\beta^-} {}^{237}Np \qquad (2.2.1)$$

Figure 2.2 shows the decay scheme of ^{237}Np. The decay scheme shown is of course incomplete in that the spins, parities and excitation characters of the various levels of the ^{233}Pa daughter nucleus.

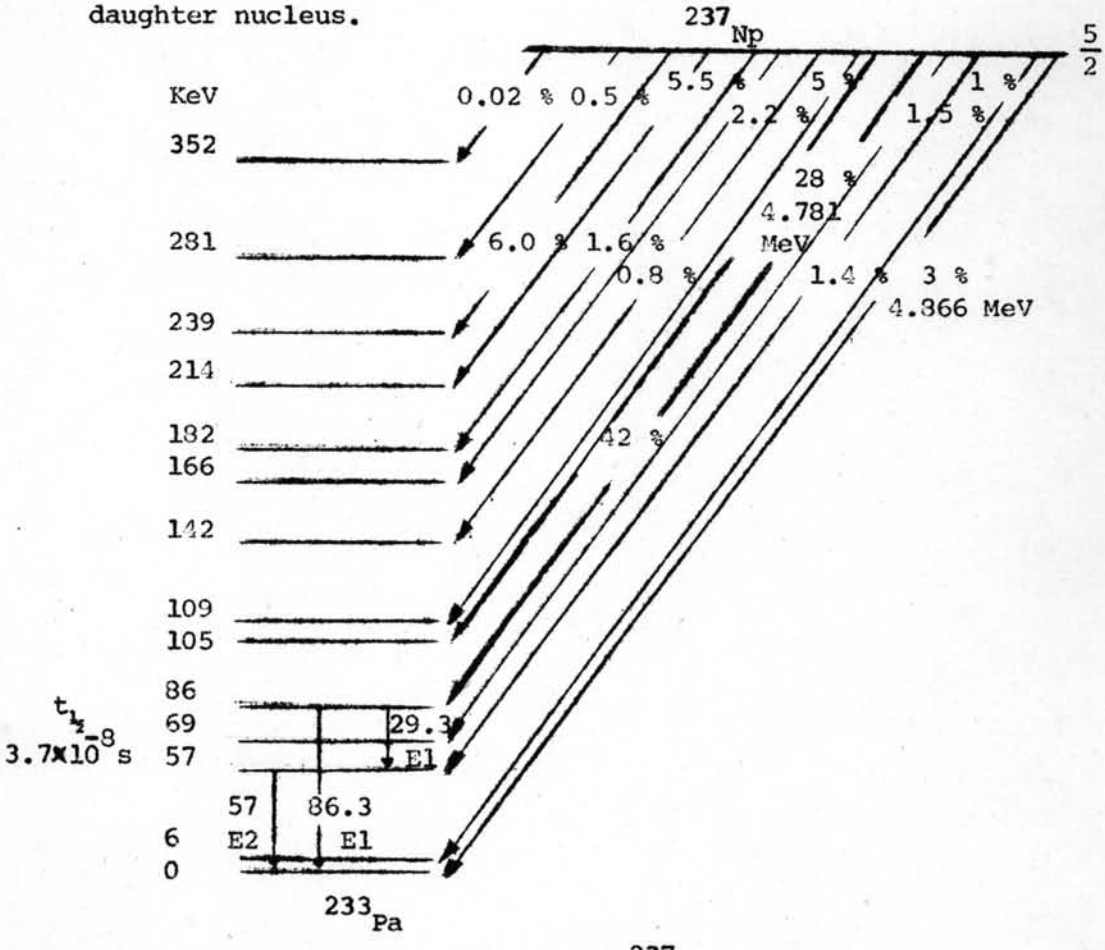

Fig. 2.2 Decay scheme of ^{237}Np

2.3 Americium -241 (Element 95)

Data on americuim -241 is given since the known activity source which will be used to determine the specific activity of the various neptunium -237 sources in a manner to be described in section 2.6, is made with this isotope. The element americium was discovered in 1944 and was produced by the bombardment of ^{239}Pu with neutrons via the reaction:

$$^{239}Pu\ (n,\gamma)\ ^{240}Pu\ (n,\gamma)\ ^{241}Pu \xrightarrow[13\ \text{years}]{\beta^-}\ ^{241}Am \qquad (2.3.1)$$

It was first isolated in the form of a pure compound by Cunningham.[5] By measuring the specific activity, he found the alpha-decay half-life to be 498 years. The alpha decay scheme for ^{241}Am is shown in Figure 2.3 which is based on the data of Asaro, Stephens, and Perlman.[6]

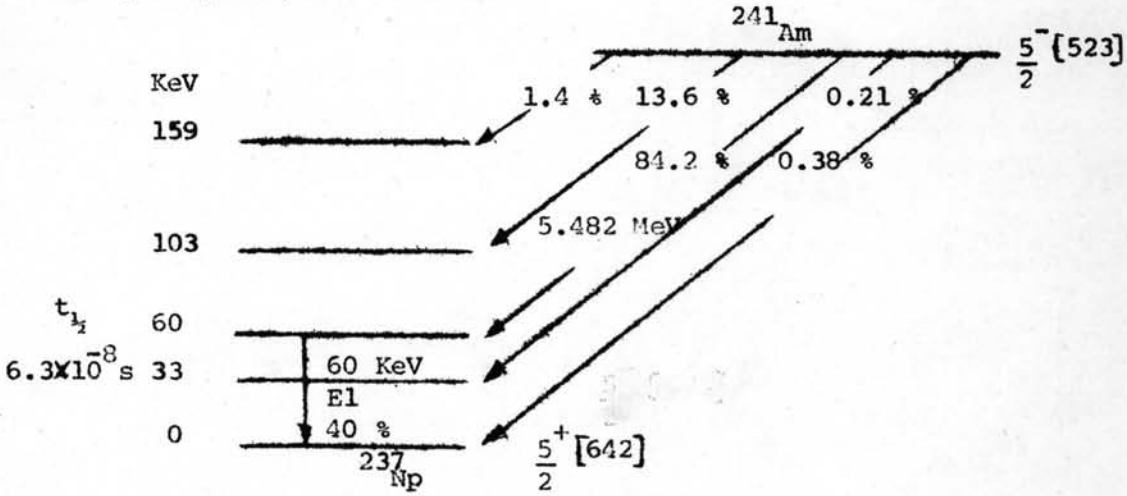

Fig. 2.3 Decay scheme of ^{241}Am

---

[5] B.B. Cunningham, "The Transuranium Elements" Nat.Nucl.Energ.Ser, (Division IV 14 B New York: McGraw-Hill Book Co., Inc., 1949)

[6] F.S. Stephens, Jr., F. Asaro, and I. Perlman, Phys. Rev. 133 (1959) 212.

2.4 The Radioactive Decay Law and the Specific Activity Method

As is well known, radioactive decay is statistical in nature. One can not tell which nuclei will decay in the next time interval. One can only give the probability per unit time that any one nuclei will decay. This statistical nature of the disintegration process leads to the exponential decay law as follows: If λ is the probability per unit time that any one of the nuclei will disintegrate, then λdt will be the probability that the nucleus will disintegrate in the time interval dt. If N is the number of nuclei present at the time t, then the number of nuclei which will disintegrate in an unit time will be

$$dN = -N \lambda \, dt \qquad (2.4.1)$$

The minus sign indicates that N will decrease with time. Integrating Eq.(2.4.1) and assuming that at t = 0, the number of radioactive nuclei present is N_o, we get

$$N(t) = N_0 e^{-\lambda t} \qquad (2.4.2)$$

where N(t) is the number of radioactive nuclei remaining at time t, λ, which is the probability per unit time that any one nuclei will disintegrate, is usually called the disintegration or decay constant.

The activity of a radioactive sample is defined as the number of disintegrations which occur in an unit time, i.e.,

$$\text{Activity} = \left|\frac{dn}{dt}\right| = \lambda N_0 e^{-\lambda t} = \lambda N \qquad (2.4.3)$$

It is the above quantity which is determined when measures the radioactivity of the sample. The decay constant λ can be determined by knowledge of the activity at two time intervals, t_o and $t_{\frac{1}{2}}$ (the half-life of the radioactive sample, defined as the time required for the activity to decrease to one half of its original activity). Since N = No/2 at $t = t_{\frac{1}{2}}$, we have from Eq. (2.4.3)

$$N_o/2 = N_o e^{-\lambda t_{\frac{1}{2}}}$$

$$t_{\frac{1}{2}} = \frac{\ln 2}{\lambda} = \frac{0.693}{\lambda} \qquad (2.4.4)$$

If the radioactive samples have a half-life greater than ten years, the half-life is determined by the specific activity method instead of being determined by measuring the activity at the two different times, t_o and $t_{\frac{1}{2}}$. Since the specific activity S is defined as

$$S = \frac{\text{activity}}{\text{weight unit}}$$

$$S = \frac{\text{activity}}{\text{atomic weight}. \frac{N}{Ao}} \qquad (2.4.5)$$

where Ao is Avogadro's number, and by Eq.(2.4.3)

$$N = \frac{\text{activity}}{\lambda} \qquad (2.4.6)$$

we get

$$S = \frac{\lambda}{\left(\frac{\text{atomic weight}}{Ao}\right)}$$

$$\lambda = S.\left(\frac{\text{atomic weight}}{Ao}\right) \qquad (2.4.7)$$

Therefore, if we know the specific actvity, the half-life can be determined from

$$t_{\frac{1}{2}} = \frac{0.693}{S}\left(\frac{Ao}{\text{atomic weight}}\right) \qquad (2.4.8)$$

2.5 The Detection of Alpha Particles By a Silicon Surface Barrier Detector

Silicon surface barrier detectors have been widely used for charged particle detection. These detectors offer many advantages over the gas-filled detectors and the scintillation detectors. Some of the advantages are:

1. smaller, more compact and convenient size,
2. fast rise time of output pulse,
3. linear response over wide energy range,
4. excellent energy resolution,
5. choice of sensitive depth, area and geometry.

The conventional silicon surface barrier detector is a large area p-n junction diode consisting of an extremely thin p-type layer on the sensitive face of a high purity n-type silicon wafer. Electrical contacts are provided by a thin gold film of approximately 40 $\mu g.cm^{-2}$ thickness on top of the P-type surface layer and a non-rectifying metal contacts on the bottom surface of the n-type silicon wafer.

A cross-section of a typical detector is shown as Figure 2.4. The semi-conductor component of the detector is illustrated in Figure 2.5. In the presence of a reverse bias voltage, free electrons and holes are swept out of a layer between the n-type and

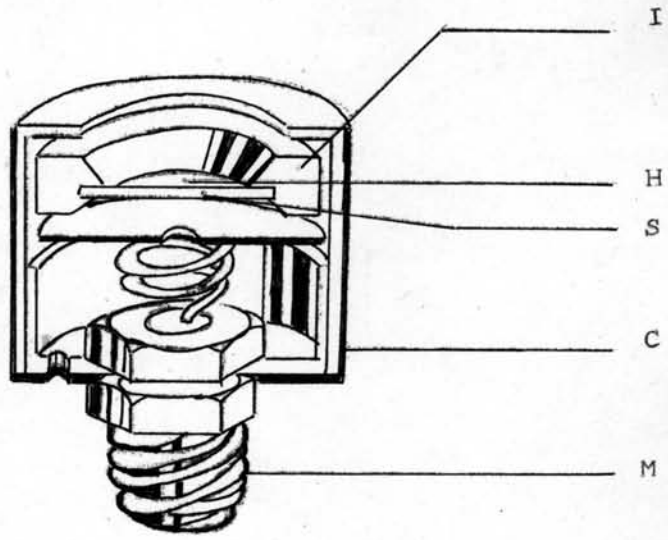

Fig. 2.4 Cross-Section of a Typical A Series Detector

H: the sensitive surface is coated with a thin layer of gold

I: a ceramic ring

S: the circular silicon wafer

C: the metal case

M: connector

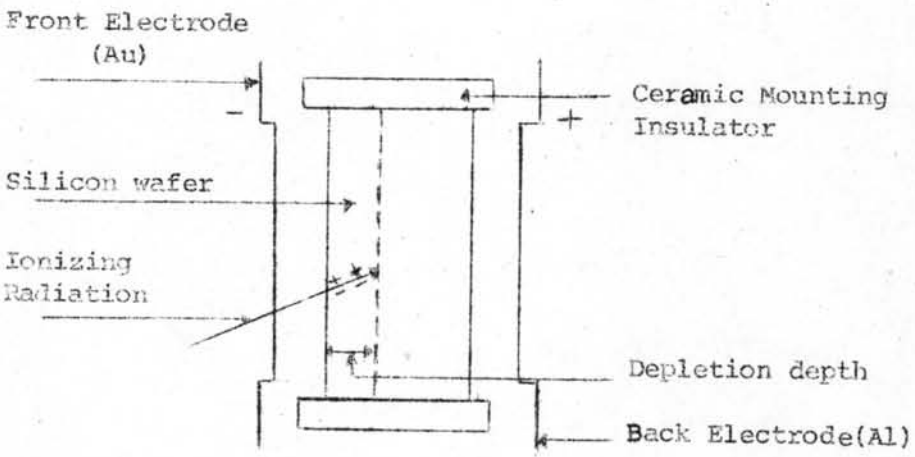

Fig. 2.5 Schemetic of Typical Surface Barrier Detector

P-type regions. This region is called the depletion (sensitive) region and corresponded to the part of the silicon wafer which contains the electric field caused by the externally applied reverse bias voltage. The thickness of the depletion region is given by

$$W \simeq 0.5 \, (\rho_n \, V_b)^{\frac{1}{2}} \qquad (2.5.1)$$

where ρ_n is the room temperature n-type resistivity in ohm-cm, V_b is the applied bias voltage in volts, and W is the depletion depth in microns.

The number of electron-hole pairs created in the depletion region by a passing charged particle is proportional to the energy loss by the charged particle as it passes through the layer. Since it requires 3.55 ± 0.1 eV to produce an electron-hole pair in silicon, an incident particle of energy E will produce E/Eo (Eo = 3.55 ± 0.1 eV) electron-hole pairs if the charged particle comes to rest inside the depletion region: The electron-hole pairs produced in the depletion region are separated by the electric field present. The resulting net current will therefore provide information about the number of electron-hole pairs created by the incident radiation.

Since the detectors are usually made from very high purity substances, there should be no background due to contamination of the silicon. If the walls of the container and supports are carefully treated, background noise of less than 0.5 counts per hour can be obtained. With the present-state of these detectors, the

size and nature of the sensitive area of the surface barrier detectors seriously restrict the solid angle which is subtended by the detector and the amount of ionizing particles which can be counted. In addition, the response of the semi-conductor detector to mono-energetic alpha particles being approximate Gaussian peaks, makes it necessary to collimate the incident flux of alpha-particles if the sensitivity of the sensitive area is not uniform. Otherwise, multiple peaks may be seen.

The high energy resolution property of the surface barrier detector can only be used to advantage if the source is sufficiently thin and uniform. Self absorption or partial absorption of the alpha particles by the source itself will occur if the source is too thick or is not uniform. Care must also be taken to insure that there is no back-scattering of alpha-particles from the backing of the source. Otherwise the information provided by the detector will not be about the radioactive nuclei. The back-scathering is usually minimised by using thin films of low atomic number materials as the backing. It should also be remembered that the film should be of a conductive material in order to avoid charging the source.

2.6 Determination of the Activity of a Radioactive Sample

The technique used to determine the activity of various samples of ^{237}Np was to compare the activity of the ^{237}Np samples with the activity of a known activity source having the same geometry (area, thickness, diameter etc.) and set up in identical detection configuration (solid angles, efficiency, and other counter parameters). In this way, the disintegration rate of the ^{237}Np samples could be determined without having to know the numerical values of all the counter parameters.

The basic formula of this method is

$$A = (A_n/N_n)\ N \qquad (2.6.1)$$

where A is the activity of the ^{237}Np sample, N is the observed counting rate for the ^{237}Np sample and where A_n and N_n are the activity and counting rate for the known activity source. The above formula is obtained by assuming that

1. The solid angles subtended by the detector at the ^{237}Np samples and the known activity source are the same.

2. the efficiency ϵ of the detector for detecting an alpha particle from the ^{237}Np is the same as that for detecting an alpha particle from the known activity source (this assumption is valid if the energies of the alpha particles emitted from the two sources are about the same).

Formula (2.6.1) follows then from the relationship between the counting rate, the activity, the efficiency and the solid angle

$$N = \frac{A \epsilon \Omega}{4\pi} \qquad (2.6.2)$$

2.7 Single Gaussian Fit

As mentioned in section 2.5, the pulse produced by the ionizing charged particle in the depletion region is shaped like a Gaussian distribution, i.e., the spectrum pulse has a form given by

$$Y = Y_o \, e^{-\alpha \left(\frac{X - X_o}{\Gamma} \right)^2} \qquad (2.7.1)$$

where Y_o is the maximum height, x_o is the center of the peak, $\alpha = 4 \ln 2$ if Γ is taken to denote full width at half maximum (FWHM). However, incomplete charge collection causes the Gaussian distribution to have a low energy tail which in turn leads to a distorted energy spectrum. To overcome this drawback, one usually attempts to fit the experimental spectrum with a modified Gaussian distribution whose parameters are determined by a nonlinear least-squares fit technique. To facilitate this fit, it is helpful to rewrite Eq.(2.7.1)as

$$\ln Y(x) = a + bx + cx^2 \qquad (2.7.2)$$

where

$$a = \ln y_o - \frac{\alpha x_o^2}{\Gamma}$$

$$b = 2 \alpha x_o / \Gamma$$

$$c = - \alpha / \Gamma^2$$