U-006-101.27

2705

THE ORIGIN OF K-65 MATERIAL

01-27-92

DOE/BLEECKER PUBLISHING
DOE-750-92
338
LETTER

R-008-101.47

Department of Energy
Fernald Environmental Management Project
P.O. Box 398705
Cincinnati, Ohio 45239-8705
(513) 738-6357

2705

JAN 2 7 1992

DOE-750-92

Mr. Steve Conliff
Fernald Investigative Group
% Bleecker Publishing
P. O. Box 21785
Columbus, Ohio 43221

Dear Mr. Conliff:

THE ORIGIN OF K-65 MATERIAL

In response to questions asked about the origin of K-65 material, all available information was reviewed to define the source of this material. The K-65 Silos are two (2) large concrete storage structures which were built in 1951 and 1952 at the Feed Materials Production Center (FMPC) at Fernald, Ohio. Their purpose was to provide storage for the residues of pitchblende processing. These residues were referred to by the designated code name "K-65". The name refers specifically to various radium bearing raffinate wastes generated in the processing of pitchblende and other radium bearing ores. A code name was also used for the pitchblende ore before processing, it was called "Q-11".

The pitchblende ore was processed for its uranium value. Since the concentration of uranium was quite high in this ore, the amount of radium, as would be expected, was also significant. The great majority of this ore came from one mine, the Shinkolobwe, in the Belgium Congo. This mine began operation in 1921 for the purpose of obtaining radium. With the increased importance of uranium during the 1940's it was reopened in 1943 and operated for its uranium content. As the radium was still of considerable value at that time, an agreement was reached between the Atomic Energy Commission (AEC) and African Metals based on the ore being processed for removal of uranium, with the provision that the residue from this processing would be returned to its owner, African Metals. The agreement, although more cumbersome to administer than a simple purchase of ore, was necessary due to the lack of adequate alternative sources of uranium ore for the expanding program of the AEC.

The residues which are stored in the K-65 Silos were generated at the Mallinckrodt Chemical Works (MCW) in St. Louis, Missouri, as well as by NLO at FMPC. The process which was used for extraction of uranium at Mallinchrodt Chemical Works was the dual-cycle ether process. This process was different from TBP-kerosene extraction system utilized at the FMPC.

The residues of this processing were originally sent back to African Metals. However, beginning April 1949 the residues were no longer returned to African Metals following processing, but were stored for future shipment. Due to continued production, storage began to be a problem As a result, shipments of the drummed K-65 residues were sent to Lake Ontario Ordnance Works (L.O.O.W.) near Niagara Falls, New York. Also, the two K-65 Silos were planned as a part of the uranium processing plant due to be constructed at Fernald, Ohio. Their completion so early in the construction of the plant may indicate the priority placed on their role in the uranium production process at that time. It is significant to note that the availability of concentrates during subsequent years changed the need for such storage of radium containing residues.

Some of the drums which were sent to L.O.O.W. were emptied into a concrete water tower at that site. A smaller number (about 6,000) were shipped to Fernald by rail. Also, the continuing production at Mallinchrodt resulted in about 25,000 drums being sent form St. Louis to FMPC. These shipments began in 1951. These drums were sent to a drum handling building which had been built specifically to transfer the contents of the drums of offsite origin into the silos. The system used was based on slurrying the material into the silo (the same method as would be utilized later from the Hot Raffinate area, the primary difference being the distance form the silos). The result of the additions of the material from L.O.O.W. and from St. Louis was the completion of filling of Silo 1.

The operation of the FMPC Refinery ran concurrently with the completion of processing of the offsite drums through the drum handling building. It is understood, therefore, that although Silo 1 was filled completely from offsite drums, Silo 2 material is a mixture of offsite K-65 material and FMPC generated K-65 material. A reference to the chronology of silo filling with MCW and NLO generated material is included. The primary information which can be gained form this reference is that prior to April 1956 the material placed in the silo was a mixture of MCW and NLO material. During 1957 and 1958, the residue of processing Australian pitchblende was added to the silo. The total amount was 380,451 lbs (in contrast to 19,004,675 lbs. of Congo origin). The remainder of the material added until the final decanting of the silo was residue generated by NLO from the remaining Congo ores.

After Silo 2 was decanted, some drummed material was added. Most of this material was soil which was contaminated with K-65 material. This occurred due to the drum storage adjacent to the drum handling building.

In summary, the great majority of K-65 material comes from processing ore from a mine in the Belgian Congo. A much smaller amount come from processing pitchblende ore from Australia. This material was processed with either the dual-cycle ether process or the TBP-kerosene extraction system.

In regard to your question regarding the K-65 residue as Manhattan Project material, a letter is enclosed on this subject. Similarly, in regard to the issues raised in your newsletter, the FEMP approach is summarized in "Radioactivity Detection and Evaluation at the FMPC", also enclosed. Finally, in regard to your question regarding the material in Silo 3, the enclosed flow diagram from the 60's addresses this issue.

If any further information is desired, please contact Teressa C. Kwiatkowski at 513-738-6004 or me at 513-738-6158.

Sincerely,

Randi B. Allen

Randi B. Allen
Operable Unit 4 Manager

FO:Allen

Enclosure: As stated

cc w/encl.:

L. C. Bogar
J. P. Hopper
D. A. Nixon
AR File
ERA Project File
Central File

[redacted]

2705-C 7-3-1

April 2, 1957

K-65 SURVEY

C. Schiman

R. B. Wolf

CENTRAL FILES

[redacted]

1. When was drummed K-65 received from MCW removed from the railroad cars by excavating services?
   Not available from our files.

2. What period of time did we dump MCW material into the tanks?
   July 16, 1952 thru January, 1956.

3. Why was operating labor high in the following months?
   <u>MARCH 1954</u> - Operators dumped 810 drums and spent extra time in area due to excessive maintenance work.
   <u>AUGUST 1954, SEPTEMBER 1954</u>, OCTOBER 1954, - Averaged 400 drums per month dumped and 241 drums of K-65 from the Pilot Plant which was dumped, trucked to the Hot Raffinate Are, sampled and pumped to storage.
   <u>MARCH 1955</u> - Equiv. of 23 shifts dumped 1076 drums.
   <u>APRIL 1955</u> - Equiv. of 16 shifts dumped 805 drums.
   <u>SEPTEMBER 1955</u> - 2 shifts dumped 24 drums and a plugged tank and excess water in the pipe trench necessitated extra pumpings from this area.
   <u>OCTOBER 1955</u> - 20 shifts dumped 334 drums, received K-65 slurry and pumped excess water to the scrap pit.
   <u>NOVEMBER 1955</u> - 14 shifts dumped 260 drums. Plagued with frozen lines, returned clear liquor to Hot Raffinate.
   <u>FEBRUARY 1956</u> - Pumped clear liquor and thawed out frozen lines.
   <u>APRIL 1956</u> - Dumped 59 drums of clean out K-65 from MCW, pumped clear liquor.
   <u>AUGUST 1956</u>- Not determined.

4. When did we stop unloading and dumping MCW material?
   Unloading - January, 1956; dumping - April, 1956.

5. During what periods of time did we process Q-11 in the NLO refinery?
   <u>Campaign</u>
   A - 4/[illegible]/54 to 6/7/54
   Pilot Plant - K-65 - 9/24/54 to 10/14/54
   B - 10/25/55 to 2/3/56
   C - 7/31/56 to 10/30/56
   D - 3/7/57 to continuing at present.
   Note: These are the dates that the k-65 produced in the refinery was campaigned to the storage tanks. Introduction of Q-11 to the refinery would precede these dates by one or two weeks.

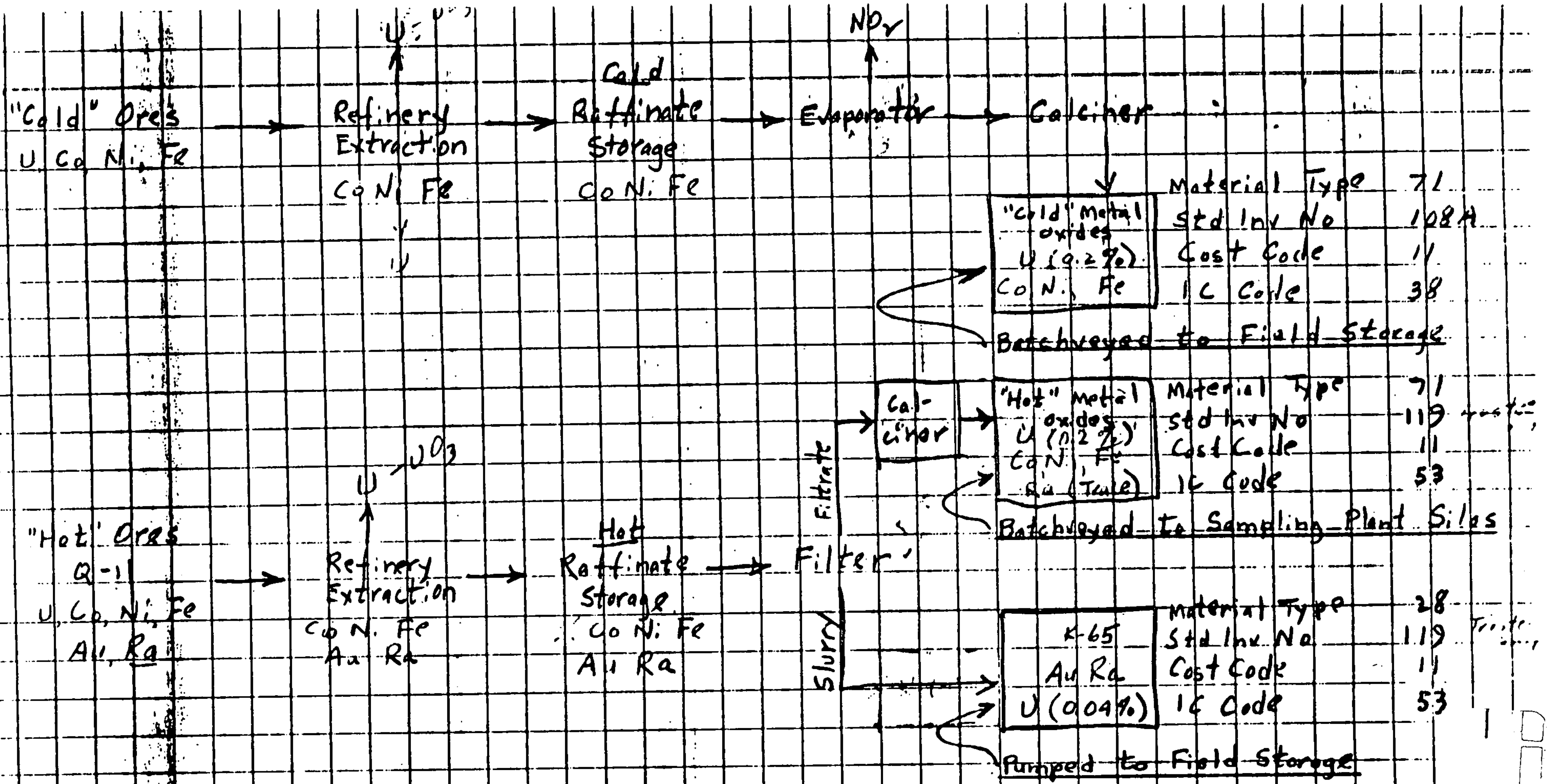
R. B. Wolf

R. B. Wolf

[redacted]

cc:
Central Files

RBW/hg



[redacted]

[redacted]

[redacted]

5

2705

— [redacted] 2705 —

From: R. A. Vogel (6748) WMCO:CP:OU4:91-009

Date: January 29, 1991 [redacted]

Subject: THE K-65 SILOS IN RELATION TO THE MANHATTAN PROJECT

To : D. A. Nixon

The question has been raised as to the correctness of the use of the term Manhattan Project wastes in relation to the material in the K-65 silos. This is not an accurate reference since the residues in the K-65 silos were not generated as a part of the Manhattan Project.

The material in the K-65 silos is residue remaining from pitchblende ore which was treated to remove uranium. Most of the ore used in this process came from one mine in the Belgian Congo. The uranium removal was done at the FMPC by NLO and at Mallinckrodt Chemical Works (MCW) at St. Louis. The extraction at Mallinckrodt Chemical Works was based on a dual-cycle ether process. At the FMPC, extraction was based on a TBP-kerosene process. In either case uranium was removed form the ore, leaving the residues we refer to as K-65 residues.

To understand the separateness of these residues from the Manhattan Project, it is important to note that none of the K-65 residues stored at FMPC were generated prior to April of 1949. Prior to that time, it had been the policy to return the residues to the Belgian owners (African Metals) of the material. Beginning in April of 1949 this policy was no longer followed. (Page 3 Attach. 1) The material generated after this time was placed in drums for eventual storage at Lake Ontario Ordinance Works and at the FMPC.

The Atomic Energy Commission took over the functions of the Manhattan Engineering District in 1946. The Manhattan Project was essentially complete at this time. Any pitchblende residues which may have remained from the project are understood to have been returned to African Metals prior to 1949.

Robert A Vogel

R. A. Vogel
Operable Unit 4

RAV:lem

Attachment

c: D. J. Carr
J. R. Craig, DOE/FMPC
S. W. Coyle
F. F Haywood, Lee Wan
S. Wolinsky, ASI/IT
ERA Project File
AR Files
Central Files

[redacted]

[illegible] Moore, Director, Construction and Engineering Division
R. J. Smith, Jr., Chief, Operations Branch, Production Division

March 1, 1951 2705

RADIUM CAKE STORAGE (K-65)

[redacted]

SYMBOL: PO:RJS:bh

[redacted]

[redacted]

of 3 pages.
Series A

In accordance with recent agreement, there is set forth below pertinent information regarding Radium Cake Storage (K-65).

1. Selected Excerpts from Pitchblend Ore Supply Contract (Administered by RMO):

"(b) Unless otherwise mutually agreed, all K-65 bearing sludges shall be weighed and sampled by the Buyer at the location where the ore and ore concentrates are processed for the extraction of Q-11 in accordance with the methods set forth in 5(d), Article II and in the presence of a representative of the Seller if the Seller shall so desire. These sludges so weighed and sampled shall be packed in containers suitable for shipment to Location S and shall be delivered to the Seller F.A.S. port in S or H.

"(c) If the Seller so elects the K-65 bearing sludges, after weighing and sampling, shall be placed in outdoor bulk storage and the Buyer agrees to construct or acquire and maintain outdoor bulk storage facilities acceptable to the Seller and the Buyer. Delivery of such sludges shall be made to the Seller after weighing and sampling and such sludges shall be at the risk of the Seller after they have been placed in bulk storage. The Buyer agrees that it will store such sludges for the Seller until ten (10) years after the date of the first delivery of such sludges resulting from the processing of the ore and ore concentrates under this contract from the Buyer to the Seller.

"(d) The Seller shall have the right to inspect such storage facilities at any time and the Buyer agrees at the Buyer's expense to withdraw from storage and ship such quantities of K-65 bearing sludges as the Seller may require packed in suitable containers to the Seller F.A.S. port in S or H. The Seller further agrees that it will give reasonable notice to the Buyer of its requirements of such sludges to be shipped by the Buyer and the Seller will not require the Buyer to withdraw from bulk storage and ship quantities of less than one hundred (100) tons of such sludge nor require the Buyer to ship quantities less than fifty (50) tons where such sludges have not been placed in bulk storage.

SPECIAL RE-REVIEW
FINAL DETERMINATION
UNCLASSIFIED
By [illegible]
Date 12-7-87

OFFICE →	PRODUCTION DIV.	0248292		ORO 59392
SURNAME →				
DATE →	3/2			

7

"(e) The cost of such storage facilities and the expense of maintaining such storage, including the guarding of the storage area, shall be [redacted] paid for by the Buyer but with respect to K-65 bearing sludges which have been placed in bulk storage which the Seller subsequently wishes to withdraw from such storage and ship as specified in subparagraph (d), the Seller shall reimburse the Buyer for the cost of such storage facilities by paying the Buyer an amount equal to the cost of containers, packing and shipment of such sludges until such amounts expended by the Buyer for the acquisition and construction of such storage facilities shall have been paid for by the Seller. The Seller agrees to reimburse the Buyer quarterly for one-half (1/2) of the maintenance expenses, including the cost of guarding. The Seller shall have an option to purchase such storage facilities from the Buyer at any time during the ten (10) year period specified in subparagraph (c) at an amount equal to the Buyer's total cost of acquiring and construction of such facilities, reduced by such amounts as the Seller shall have paid to the Buyer as amounts equivalent to the cost of containers, packing, and shipment of such sludges withdrawn from bulk storage.

[illegible]

"(f) In the event that the Seller does not exercise the option to purchase provided for in subparagraph (e) and gives no instructions to the Buyer regarding the shipment of K-65 bearing sludges which remain in bulk storage at the end of the ten (10) year period named in subparagraph (c), above, such sludges shall be treated as abandoned by the Seller to the Buyer unless otherwise mutually agreed.

"(g) Residues and sludges other than K-65 bearing sludges which shall remain after the extraction of Q-11 shall, unless otherwise mutually agreed, be stored by the Buyer in outdoor bulk storage acceptable to the Buyer and the Seller and shall be at the risk of the Seller. At any time during the ten (10) year period named in subparagraph (c) above, the Buyer agrees to pack and ship such residues and sludges to points designated by the Seller but the costs of packing and shipping such residues and sludges shall be borne by the Seller. If the Seller gives no instructions to the Buyer regarding the disposition of such residues and sludges with the ten (10) year period named in subparagraph (c) above, such residues and sludges shall be treated as abandoned by the Seller to the Buyer unless otherwise mutually agreed."

SPECIAL REVIEW
FINAL DETERMINATION
UNCLASSIFIED
By [illegible]
Date 12-7-[illegible]

"(c) The K-65 content of the ore and ore concentrates containing Q-11 shall be based on assays for K-65 content made by the National Bureau of Standards for 'S' and by the National Physical Laboratory for 'H'. The Buyer agrees to return to the Seller in the form of sludges not less than ninety-eight (98) per cent of the K-65 contained in ore and ore concentrates, assaying not less than forty-seven and five tenths (47.5) per cent Q-11, dry basis; not less than ninety-six and five tenths (96.5) per cent of the K-65 content in ore and ore concentrates assaying less than forty-seven and five-tenths (47.5) per cent Q-11, dry basis, and not less than twenty-five (25) per cent, dry basis."

Note: Penalty is \$4.83 per mg. of Ra."

[redacted]

[redacted]

B. Background to Date:

Prior to April 1, 1949, radium cake was returned to the vendor as fast as possible. At the request of the vendor, the Commission on that date commenced storing the radium cake in the United States. The considerations pertaining to storage at that time are contained in report titled "Radium Cake Storage" prepared by E. C. Sargent and H. F. Reichard, dated April 22, 1949. The Sargent-Reichard report recommended storage in specially lined drums. For medical reasons and because of the uncertainty as to length of time in storage, an existing above ground concrete water tower at LOOW was selected for bulk storage. The LOOW tower was inspected for the vendor by the Stearns-Roger Manufacturing Company on December 22, 1949 and December 28, 1949 who first rejected the idea and then gave limited approval to store not more than 3750 tons in the tower. Details of the Stearns-Roger Reports are in your files.

As of February 28, 1951, some 2100 tons of K-65 have been dumped into the LOOW tower and an additional 2000 tons are in drums at LOOW awaiting dumping.

Based on consumption of 120 tons/mo of pitchblend ore at the MCW Refinery, there will be K-65 production at the rate of about 140 tons/mo.

Distribution

Copy 1 - D. C. Moore
Copy 2 - Operations Branch
Copy 3 - Operations Branch
Copy 4 - M & R ✓

SPECIAL REREVIEW
FINAL DETERMINATION
UNCLASSIFIED
By:
Date 12-7-87

[redacted]

[redacted] 2705

[redacted]

WESTINGHOUSE MATERIALS COMPANY OF OHIO

RADIOACTIVITY DETECTION AND EVALUATION AT THE FMPC

NOVEMBER 29, 1990

[redacted]

[redacted]

TABLE OF CONTENTS

[redacted]

1.0 INTRODUCTION

The following report is intended to be utilized as a radiological information source. It is also intended to serve as a link between the radiological sampling and analysis groups with those individual responsible for preparing Removal Site Evaluations (RSEs), Removal Actions, and other documents which must evaluate radiological contamination levels and determine the associated health effects. Also, it is hoped that this document will be an evolving information source to satisfy the needs or problems associated with radioactivity concepts and calculations. In order to attain these objectives, and others which may develop, any comments are greatly appreciated.

Contained in this report is a general discussion of radioactivity all centered around the basic units and definitions commonly used by both the sampling groups and other engineering groups. Additional information concerning the mechanisms of radiation detection and the various instruments used is also included in an effort to describe the processes and to provide insight to the limitations involved. A section on laboratory methods of radionuclide detection is included in order to provide engineers with the details of measurement practices so that the proper perspective is available when developing removal action plans and schedules. The final section deals with the specifications put forth as part of the Quality Assurance Project Plan (QAPP).

The evolution of this document is intended to provide that information which is both specific and commonly needed in those areas discussed above. The intent is to avoid producing a document that is either cumbersome or unfriendly. Instead the approach was taken to provide both a format for delivering pertinent and useful information as well as the initial material expected to be desired by all of the organizations involved. The material contained here can be either expanded or reduced as the needs arise to continue to meet the objectives and the needs of the concerned parties.

[redacted]

2.0 UNITS AND DEFINITIONS

The purpose of this section is to introduce the basic units and their definitions used in performing radiological investigations. This section will cover several areas of radioactivity calculations which are pertinent to the discussions of radiological measurement and analysis. Additional information which may be needed by the users of this document can refer to the reference section for texts providing further explanation.

RADIOACTIVITY

The radiations of primary concern at the Feed Materials Production Center (FMPC) and for the purposes of this document are those which originate in atomic or nuclear processes. More specifically, the radiations which will be the focus of this radiological fact sheet are listed in Table 2.1. They can be separated into three general types: 1) fast electrons, 2) heavy charged particles and 3) electromagnetic radiation. Fast electrons include beta particles (positive or negative) emitted in nuclear decay. For the purposes of this document heavy charged particles denote alpha particles. In reality, heavy charged particles are defined as all energetic ions with mass of one atomic mass unit or greater, which would include alpha particles, protons, fission products, or the products of many nuclear reactions (Knoll, 1979). Electromagnetic radiation are X-rays, which are emitted in the rearrangement of electron shells of atoms, and gamma-rays which originate from transitions within the nucleus (Knoll, 1979).

TABLE 2.1: CLASSES OF IONIZING RADIATIONS PERTINENT TO THE FMPC RADIOLOGICAL INVESTIGATIONS

Type of Radiation	*Category of Radiation*
Charged Particle Radiation	Fast Electrons Heavy Charged Particles
Uncharged Radiation	Electromagnetic Radiation - Gamma-Rays and X-Rays

[redacted]

The **activity** of a radionuclide is defined as its rate of decay, and is given by the fundamental law of radioactive decay, shown in Equation 2.1.

$$\frac{dN}{dt} = -\lambda N \qquad (2.1)$$

where the terms of Equation 2.1 are define as follows:

N = Number of radioactive nuclei.

λ = Decay constant for the radionuclide, units of $(\text{seconds})^{-1}$.

The unit of activity has been described by the curie (Ci), which is defined as exactly **3.7×10^{10} disintegrations / second.** The more conventional unit currently being employed is the SI equivalent, the becquerel (Bq). The becquerel is defined as one disintegration per second or one becquerel is equal to **2.703×10^{-11}** Ci. Activity measures a source disintegration rate. Or the other hand, the emission rate of radiation produced by decay is not synonymous with activity, since often a given radiation will be emitted in only a fraction of all the decays. A knowledge of the decay scheme of the particular isotope is needed in order to infer a radiation emission rate from its activity.

The specific activity of a radioactive source is defined as the activity per unit mass of the radioisotope. The specific activity is defined by Equation 2.2.

$$\text{Specific Activity} \equiv \frac{\text{Activity}}{\text{Mass}} = \frac{\lambda N}{NM/A_v} = \frac{\lambda A_v}{M} \qquad (2.2)$$

where the terms of Equation 2.2 are defined as follows:

M = Molecular Weight of sample.

A_v = Avogadro's Number (6.02×10^{23} nuclei/mole).

λ = Radioisotope decay constant (Ln 2 / half-life).

[redacted]

RADIATION INTERACTIONS

The operation of any radiation detector is dependent upon the mechanism in which the radiation interacts with the material of the detector. Therefore, in order to understand the response of a particular detector to a given radiation it is necessary to understand the fundamental mechanisms by which radiations interact and lose their energy in matter. In order to gain a thorough description of these interactions one can consult any number of texts, several of which are listed in the reference section.

Heavy Charged Particles

Alpha particles interact with matter primarily through their strong positive charge interaction with the electrons of the absorber. The interaction produces positive ions and free electrons. Alpha particles are characterized as nonpenetrating radiation since their travel distances in air are typically less than a few centimeters, while in tissue alpha particles travel in the range of microns (μm or 1×10^{-6} meters).

Fast Electrons

Fast electrons lose energy at a much slower rate in an absorbing medium than heavy charged particles. In addition the path of fast electrons through materials is much less straight than heavy charged particles would exhibit, such as the alpha particle. Tortuous paths are expected for fast electrons since their mass is equal to that of the orbital electrons with which they are interacting as a result they are easily deflected during collisions.

Gamma-Rays

Gamma-ray interactions are qualitatively different from that of either alpha or beta radiation. Heavy charged particles and fast electrons have definite ranges in matter, and as a result can be completely absorbed; therefore, gamma-rays can only be reduced in intensity by using increasingly thicker absorbers. Gamma radiation can not be completely absorbed.

There are primarily three interaction mechanisms by which gamma-rays interact with matter: 1) photoelectric absorption, 2) Compton scattering, and 3) pair production. A more detailed description of these processes can be found elsewhere, it sufficient to state that all of these processes lead to the partial or complete transfer of the gamma-ray photon energy to electron energy. These processes cause the gamma-ray to either disappear entirely or be scattered through a large average angle. Gamma-ray interactions are therefore in sharp contrast to the charged particle radiations discussed earlier which slow down gradually through multiple interactions with numerous absorber atoms.

DECAY SCHEMES

The transformations of natural radioisotopes occur by one of two processes: alpha or beta emission. These processes result with conservation of charge, and with mass changes that are strictly accounted for either by the energy change of the products according to Einstein's law of energy and mass equivalence, or by the emission of a gamma-ray. Alpha decay results in a product isotope four units lower in mass and two units lower in nuclear charge. Beta decay produces a very small mass change, but nuclear charge increases by one unit. In some situations a radioisotope can decay by either beta or alpha emission, for example the case of Bismuth-214 (atomic number 83) as shown below. This process is called "branching" and the relative probability of decay by each process is called the "branching ratio."

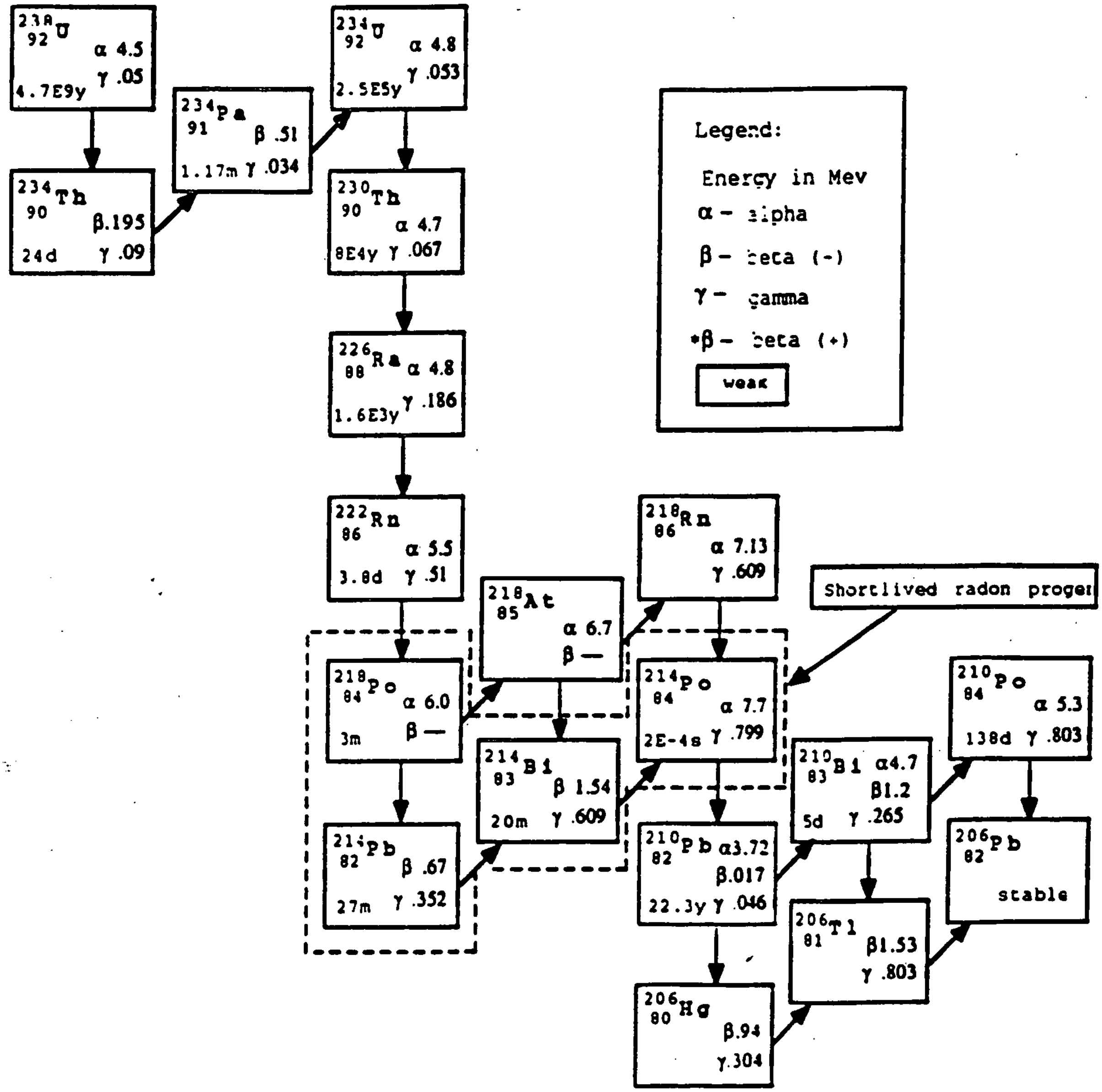
$$\mathrm{Bi}^{214}_{83} \rightarrow \mathrm{Tl}^{210}_{81} + \mathrm{He}^{4}_{2} \qquad 0.04 \text{ Percent}$$

$$\mathrm{Bi}^{214}_{83} \rightarrow \mathrm{Po}^{214}_{84} + \mathrm{e}^{0}_{-1} \qquad 99.96 \text{ Percent}$$

The isotopes formed by a sequence of transformations constitute the members of a radioactive series. A natural radioactive series originates with a long-lived element and ends with a stable isotope. There are four principal radioactive series: 1) uranium-radium series (4n + 2), 2) the thorium series (4n + 0), 3) the actinium series (4n + 3), and 4) the neptunium series (4n + 1), which no longer occurs in nature. These are the only possible radioactive series, since beta-ray emission does not change the atomic weight, usually denoted by a capital "A", and since proton or neutron emissions do not occur. Figures 2.1, 2.2, and 2.3 illustrate the decay chains of the uranium series, with the parent of uranium-238, the thorium series, with the parent of thorium-232, and the actinium series with the parent uranium-235, respectively.

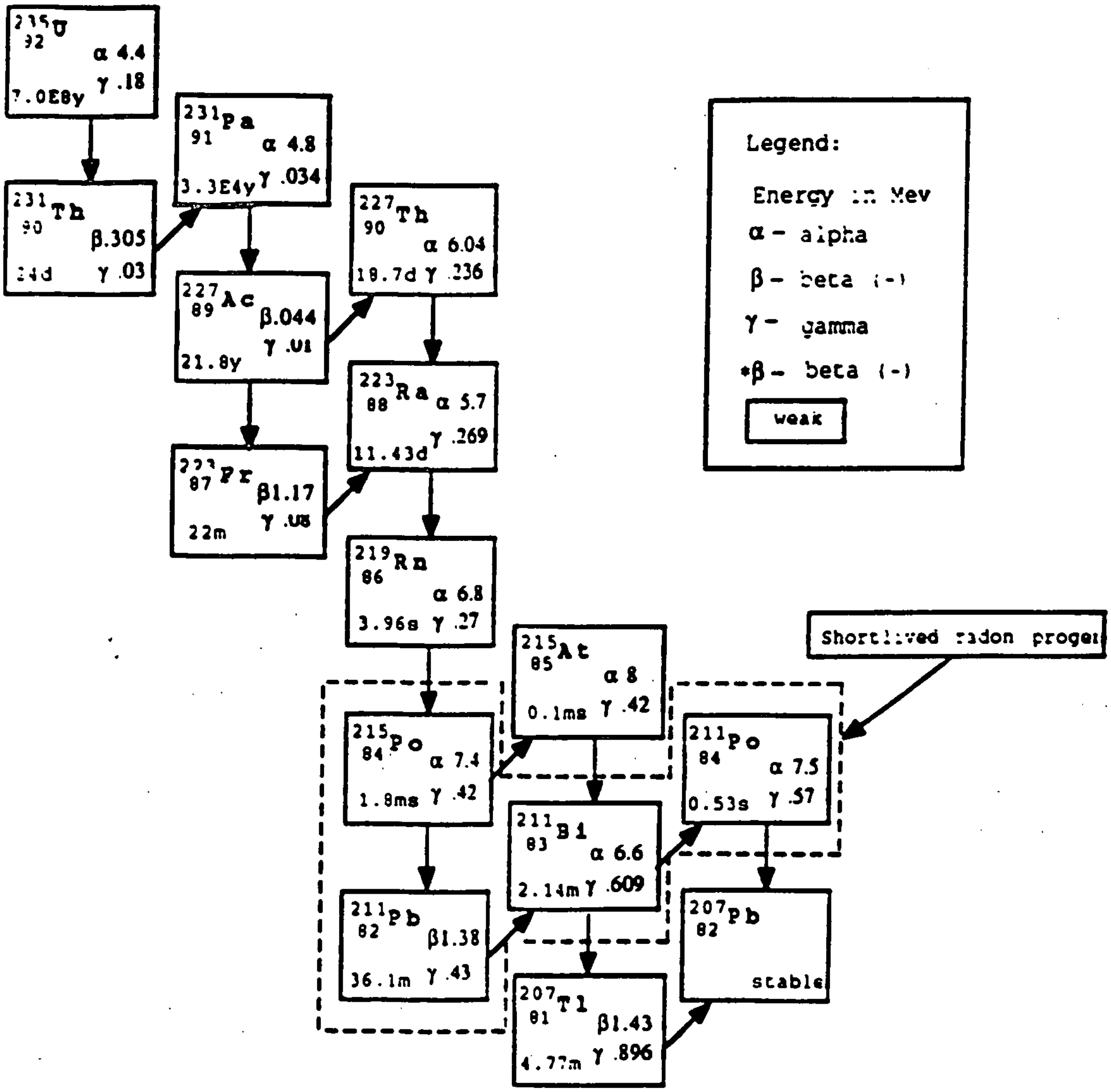
[redacted]

FIGURE 2.1: URANIUM-238 DECAY SERIES



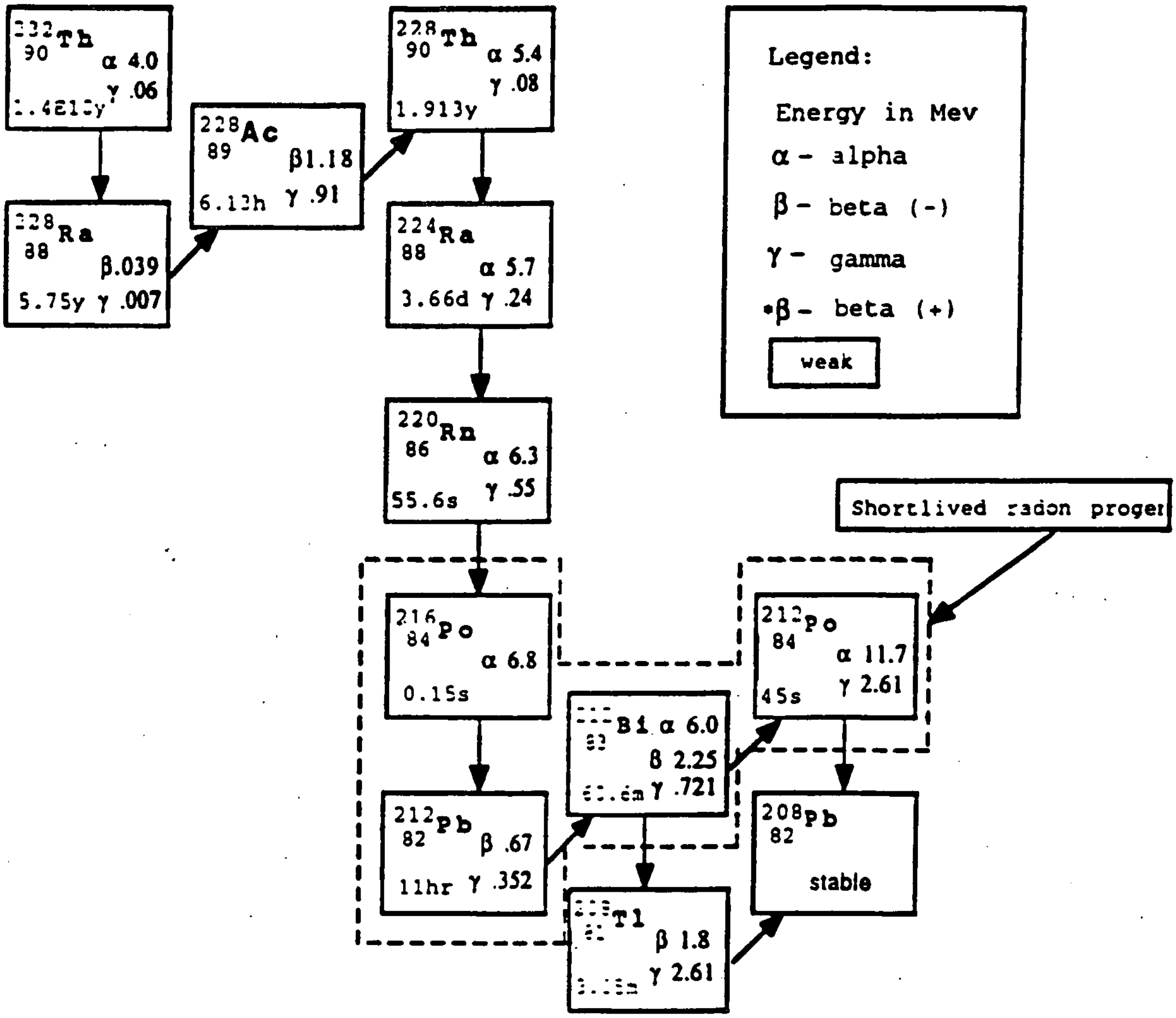
[redacted]

FIGURE 2.2: URANIUM-235 DECAY SERIES



[redacted]

FIGURE 2.3: THORIUM-232 DECAY SERIES



[redacted]

SECULAR EQUILIBRIUM

Secular equilibrium refers to the relatively rapid buildup of a very short-lived daughter from a long-lived parent. For instance, consider the five member series starting with krypton-90 and ending with zirconium-90 (Cember, 1987):

$$Kr_{36}^{90} \xrightarrow{\beta} Rb_{37}^{90} \xrightarrow{\beta} Sr_{38}^{90} \xrightarrow{\beta} Y_{39}^{90} \xrightarrow{\beta} Zr_{40}^{90}$$

33 s 2.74 m 19.9 y 64.2 h

In this series Kr36 decays by beta emission, with a half-life of 33 seconds, to Rb37. Due to the short half-life of Kr36, any amount of Kr36 will in short time period, approximately 10 minutes, be transformed completely into Rb37. As a result the activity of strontium-90 will quickly build up. The Y39, daughter of strontium-90, will accumulate much faster than the decay of strontium-90. After approximately seven half-lives of yttrium-90, a point is reached whereby the instantaneous amount of strontium-90 that transforms is equal to that of yttrium-90. At this point the yittrium-90 is said to be in secular equilibrium with the strontium-90. At equilibrium the activity of the parent equals the activity of the daughter:

$$N_{parent}\lambda_{parent} = N_{daughter}\lambda_{daughter}$$

[redacted]

[redacted]

RADIOACTIVE EQUILIBRIUM

Two other general cases of radioactive equilibrium should be considered. Unlike the case of secular equilibrium where the daughter half-life is very much less than the parent, $(\lambda_A << \lambda_B)$ these cases deal with the situations where: 1) the parent half-life is only slightly greater than the daughter, $(\lambda_A < \lambda_B)$ and 2) the case where the parent half-life is less than that of the daughter,$(\lambda_A > \lambda_B)$.In the first of these two cases, where the parent half-life is slightly greater than the daughter and given that the parent is initially pure and free of any daughter activity, the relationship between the daughter and the parent develops into what is known as transient equilibrium. The daughter activity starts at zero rises to a maximum and then seems to decay with the same half-life as the parent. At this point the daughter is undergoing transformations at the same rate as it is being produced. The following expression explicitly represents the activity of the daughter.

$$\lambda_B N_B = \frac{\lambda_B \lambda_A N_{A_0}}{\lambda_B - \lambda_A}(e^{-\lambda_A t} - e^{-\lambda_B t})$$

where:

λ_A = Decay constant for the parent.

λ_B = Decay constant for the daughter.

N_A = Initial number of parent atoms.

After a sufficiently long time, given that the decay constant for the daughter is greater than that of the parent, $(\lambda_A < \lambda_B)$, then the expression representing transient equilibrium can be written as follows:

$$\lambda_B N_B = \frac{\lambda_B \lambda_A N_A}{\lambda_B - \lambda_A}$$

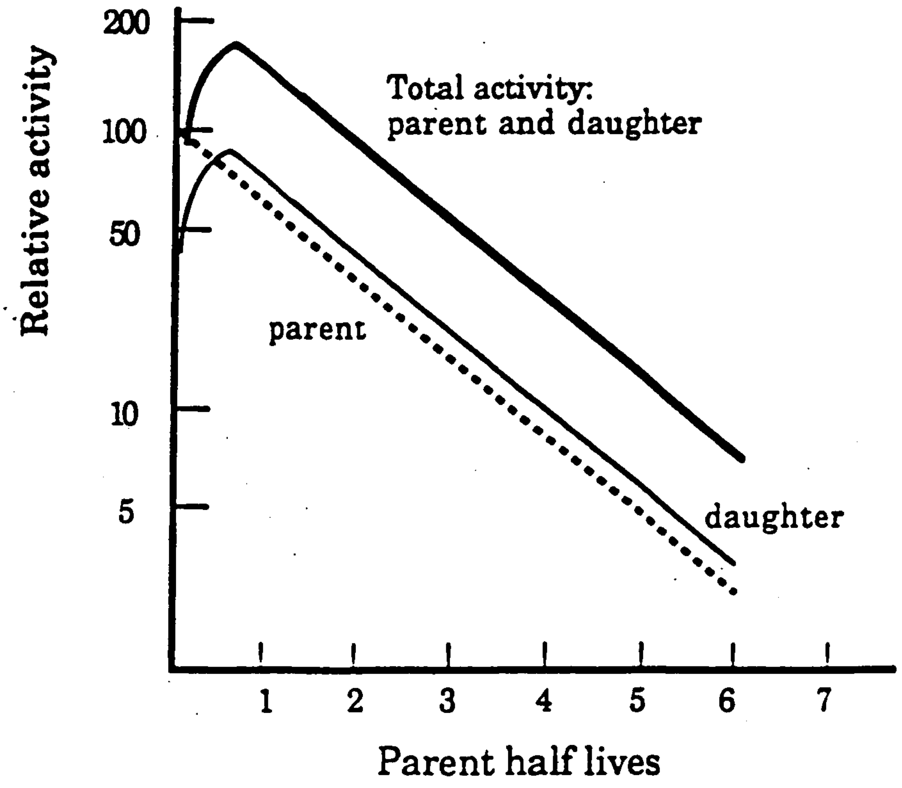
[redacted]

[redacted]

Figure 2.4 represents the buildup and subsequent decay of a daughter and parent in a transient equilibrium situation. The time when the parent and daughter isotopes may be considered to be equilibrated depends on their respective half-lives. The shorter the half-life of the daughter, relative to the parent, the more rapidly will the equilibrium be attained.

In the case where the half-life of the daughter exceeds that of the parent, no equilibrium is possible. The daughter activity reaches a maximum and then reaches a point where it decays at its own characteristic rate. This case is represented in Figure 2.5.

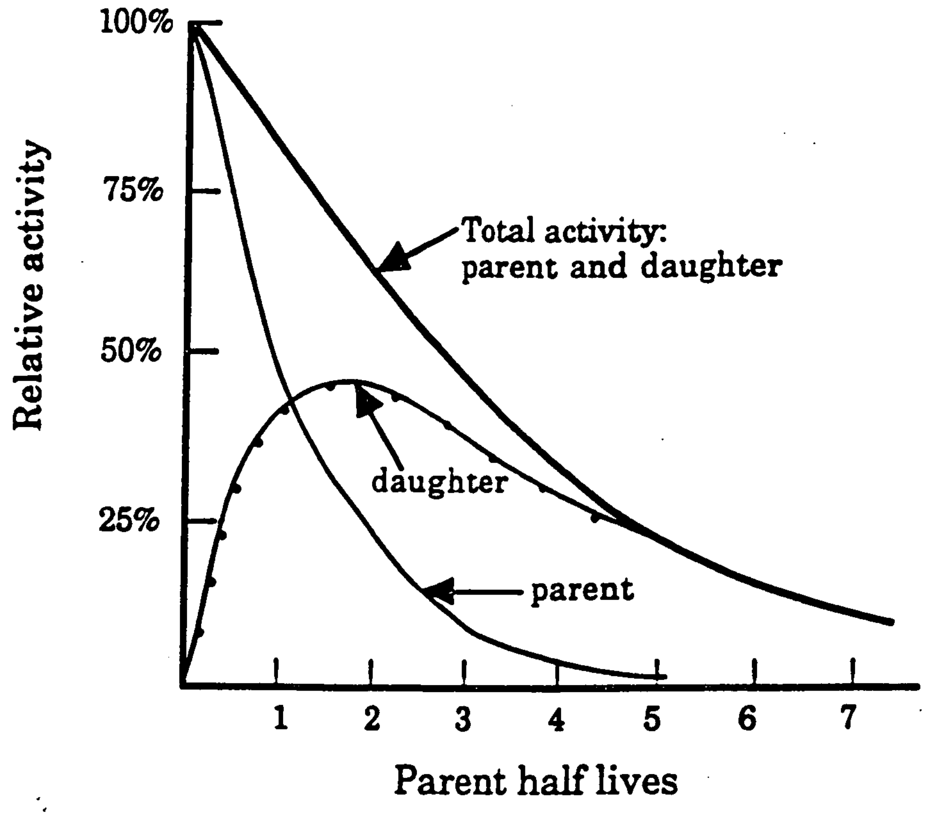
FIGURE 2.4: TRANSIENT EQUILIBRIUM



[redacted]

[redacted]

FIGURE 2.5: NO EQUILIBRIUM CONDITION



RADIOACTIVITY CALCULATIONS

The purpose of this section is to illustrate some of the basic calculations previously discussed. The following calculations are described: 1) derivation of natural uranium activity fractions, 2) calculation of specific activity, and 3) formulae used for determining percent enrichment of uranium-235 in a sample.

Derivation of Natural Uranium Activity Fractions

Activity is defined by Equation 2.3, where "N" has units of atoms per gram and λ is the decay constant with units of inverse seconds.

[redacted]

$$\text{Activity} = N \times \lambda \qquad (2.3)$$

[redacted]

Consider, for instance, the calculation of natural uranium activities in a one gram sample. Equation 2.4 illustrates the formula for determining the activity distributions for natural uranium. Figures 2.1 through 2.3 list the half-lives for the three isotopes of uranium. The terms "N" and λ shown in Equation 2.4 are defined by Equations 2.5 and 2.6. Natural uranium contains 99.283 percent uranium-238, 0.711 percent uranium-235, and 0.0054 percent uranium-234.

$$\underset{\text{Activity}}{\text{Natural Uranium}} = N\lambda(\%\ \text{U-238}) + N\lambda(\%\ \text{U-235}) + N\lambda(\%\ \text{U-234}) \qquad (2.4)$$

$$N = \frac{A_v}{M.W} = \frac{\frac{\text{atoms}}{\text{mole}}}{\frac{\text{grams}}{\text{mole}}} \qquad (2.5)$$

$$\lambda = \frac{\text{Ln } 2}{\text{half-life} \times 365\,\frac{\text{d}}{\text{yr}} \times 24\,\frac{\text{hr}}{\text{d}} \times 3600\,\frac{\text{s}}{\text{hr}}} \qquad (2.6)$$

Using Equations 2.5 and 2.6 to determine both "N" and λ for each isotope and then inserting the results into Equation 2.4 gives the following activities in the one gram sample:

Uranium-238 = 1.23×10^4 disintegrations / s

Uranium-235 = 5.65×10^2 disintegrations / s

Uranium-234 = 1.23×10^4 disintegrations / s

Notice that the activities for uranium-238 and uranium-234 are equal. This is expected for natural uranium since the half-life for uranium-234 is much smaller than the half-life for uranium-238 and therefore the uranium-234 has come into secular equilibrium with the its parent uranium-238.

[redacted]

[redacted]

A curie is defined as 3.7×10^{10} disintegrations / s. Therefore, dividing the activities above by 3.7×10^{10} gives the activity of each radionuclide in units of curies. Using the fact that one picocurie (pCi) is equal to 1×10^{-12} curies, the following picocurie activities can be expressed:

Uranium-238 = 3.3×10^5 pCi; fraction = 0.4887

Uranium-235 = 1.5×10^4 pCi; fraction = 0.02266

Uranium-234 = 3.3×10^5 pCi; fraction = 0.4887

Notice that the fraction of uranium-235, found by dividing the activity of uranium-235 by the total activity, is equal to 0.02266. If a total uranium value is given with units of picocuries per gram and the isotopic activity distribution is known to be natural, or normal, then one can simply multiply the total activity by the fractions above in order to obtain the isotopic activities.

Determination of Percent Uranium-235 Enrichment for an Isotopic Uranium Sample

If a sample is expressed isotopically in units of micrograms per gram (µg/g) for uranium-238, uranium-234, and uranium-235, then the percent enrichment of uranium-235 would be described by Equation 2.7. However, when the concentrations are in activities this procedure can not be used. Instead one must use the specific activity of each isotope in order to convert the activities to a mass unit. Recall the specific activity was defined by Equation 2.2. Table 2.2 lists the specific activity for uranium-238, uranium-235, and uranium-234.

$$\%\,\text{U-235} = \frac{\text{U-235 }(\mu g/g)}{(\text{U-235, }\mu g/g) + (\text{U-238, }\mu g/g) + (\text{U-234, }\mu g/g)} \times 100 \qquad (2.7)$$

[redacted]

[redacted]

TABLE 2.2: SPECIFIC ACTIVITIES FOR URANIUM-238, URANIUM-235, AND URANIUM-234

RADIONUCLIDE	*SPECIFIC ACTIVITY*
Uranium-238	3.34×10^5 pCi/g
Uranium-235	2.46×10^6 pCi/g
Uranium-234	6.19×10^9 pCi/g

[redacted]

[redacted]

3.0 MECHANISMS OF RADIATION DETECTION

The purpose of this section is to discuss the five principal mechanisms used to detect radiation: 1) ionization, 2) proportional, 3) Geiger-Mueller, 4) scintillation, and 5) semiconductor. Ionization chambers, proportional counters, and Geiger tubes all function, in somewhat different ways, by creating an electrical signal when ion pairs are formed within the gas as radiation interacts with the detector. Scintillation detectors basically function by changing the energy of an ionizing particle into a flash of light. Semiconductor detectors are based on counting "electron-hole pairs" which are created by the charged particle (primary or secondary radiation) as it passes through the detector.

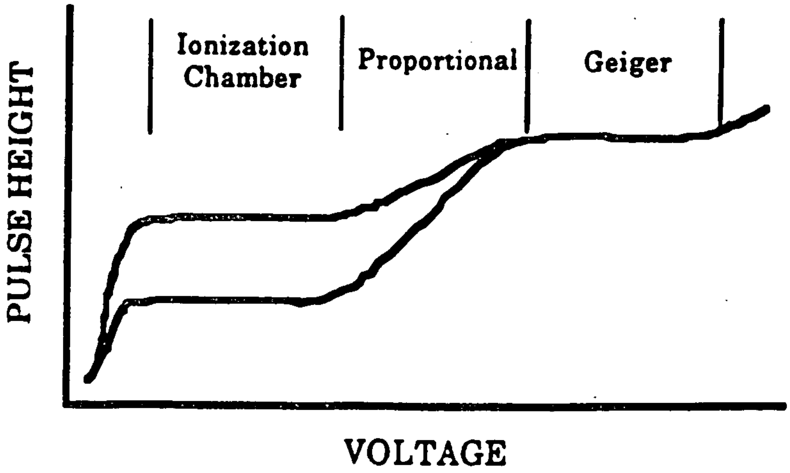
Ionization Chamber Counters

Ionization chambers are considered the simplest of all gas-filled detectors. The term "ionization chamber" has come to be known as any type of detector in which ion pairs are collected from gases. The normal operation of an ionization chamber counter is based on collecting the charges, using an electric field, which result when ionization occurs using relatively low voltages within the gas of the detector. When a constant amount of radiation is allowed to pass through a detector, and the voltage is varied, several distinct regions of significance appear which are important to radiation measurement (Cember, 1987). Figure 3.1 illustrates this concept (Cember, 1987). The low voltage region is referred to as the ionization chamber region. Cember describes "low voltages" as a voltage great enough to collect the ions before a significant fraction of them are able to recombine, yet not great enough to accelerate the ions sufficiently to produce secondary ionization by collision (Cember, 1987). Furthermore, in the ionization chamber region the number of electrons collected by anode (positive terminal) will be equal to the number produced by the primary ionizing particle (Cember, 1987). As a result, the pulse size will be independent of the voltage and will only depend upon the number of ions produced by the primary ionizing particle as it passes through the detector (Cember, 1987). This linear relationship, with constant current, between pulse size and the number of ions produced for ionization chamber counters makes them able to distinguish between radiations of different specific ionization, such as alphas and betas or gammas (Cember, 1987 and Knoll, 1979).

[redacted]

[redacted]

FIGURE 3.1: PULSE HEIGHT VERSUS VOLTAGE DIAGRAM ILLUSTRATING THREE REGIONS OF GAS-FILLED DETECTOR



Proportional Counters

Similar to Geiger-Mueller counters, which will be discussed next, proportional counters are based on the phenomenon of gas multiplication to amplify the charge created by the ion pairs which formed as the ionizing radiation passes through the gas of the detector. A disadvantage of operating a counter in the ionization chamber region is the weak output pulse, which requires either a large amount of amplification or a high degree of input sensitivity in the scaler (Cember, 1987). A method for over coming this problem, and yet still taking advantage of pulse size dependence to distinguish between radiation types, is to operate the counter in the proportional region (see Figure 3.1) (Cember, 1987). The proportionality region occurs as the voltage across the counter is increased beyond the ionization chamber region and at the point where secondary electrons are produced by collision. Proportional counters can distinguish between alpha and beta radiation, and more specifically beta particle activity can be obtained from the alpha and beta count rate.

[redacted]

[redacted]

Geiger Counter

If the voltage is increased in the gas filled detector beyond the proportional region, an avalanche of secondary electrons extends along the entire length of the anode and the Geiger region begins (Cember, 1987). In the Geiger region the size of all pulses, regardless of the specific radiation type, is the same. Therefore, in the Geiger region a counter cannot distinguish between several types of radiation. The advantage of operating in the Geiger region is that the very large output pulses, due to the high gas amplification, eliminates the need for a pulse amplifier or an amplifier that does not have to meet the specific requirements of high pulse amplification (Cember, 1987).

Scintillation Counter

A scintillation counter detects ionizing radiation by producing visible light, which is recorded by a photonmultiplier tube, as the ionizing radiation interacts with the scintillation crystal. A variety of materials can be used as a scintillator. The most widely applied scintillators comprise the group of inorganic alkali halide crystals, such as the sodium iodide crystal (NaI). Scintillation detectors are based on a principle called fluorescence which refers to the prompt emission of visible radiation from a substance following its excitation. Scintillation counters are commonly used to count gamma-rays and low-energy beta rays. Gas filled detectors have low detector efficiencies for gamma-rays since the photons are easily lost before they can be counted. Solid scintillating crystals, however, have high detection efficiencies for gamma-rays.

Semiconductor Detectors

As compared to scintillation detectors, semiconductor detectors offer the important advantage of providing much better energy resolution. In scintillation detectors the process where radiation is converted to light and then used to generate an electrical signal is somewhat lengthy and inefficient (Knoll, 1979). As a result, the energy needed to produce one photoelectron, signal carrier, is at least 1000 electron volts and the number of carriers produced after the radiation interaction is usually no more than a few thousand (Knoll, 1979). "The statistical fluctuations with such a small number place an inherent limitation on the energy resolution that can be achieved under the best of circumstances, and nothing can be done to improve the energy resolution beyond this point (Knoll, 1979)." "Semiconductor detectors reduce the statistical limit on energy resolution by increasing the quantity of information carriers per pulse (Knoll, 1979)."

[redacted]

[redacted]

Similar to the ion pairs created in the scintillation crystal, the basic signal carriers in semiconductor detectors are the electron-hole pairs, which are created along the path of incident radiation.

4.0 SURVEY INSTRUMENTS

Survey meters are described as portable field measurement instruments and include the capability for detecting a variety of radiations, such as alphas, betas, gamma-rays, and even neutrons.

Since alpha radiation is low penetrating, and therefore normally does not constitute an external dose problem, alpha survey instruments are needed only for detection of surface contamination. Typically the sensitivity of alpha survey instruments is approximately 500 alpha/minute per 100 square centimeters based on a total source emission (4π) and a counting efficiency of 15% (DOE, 1980).

Beta and gamma radiation can present both a surface contamination problem as well as an external radiation problem. Typically reliable detection levels for beta radiation are approximately 600 beta/minute per probe area for betas of 500 keV energy, using a thin window detector with a window thickness of 1.4 to 2.0 mg/cm^2 (DOE, 1980).

Ion chamber type instruments are usually used for beta and gamma radiation dose rate measurement (DOE, 1980). Since potential exposure rates in the field can vary considerably, two types of instruments are often used. A large chamber instrument is used for low to moderate exposure rates and a small ion chamber is used for high exposure rates. Both of these types of instruments usually have the capability to permit measurements of nonpenetrating radiation by sliding or removing the outer shield. Table 4.1 provides a summary of these survey instruments (DOE, 1980 and Kathren, 1974).

[redacted]

[redacted]

TABLE 4.1: OVERVIEW OF PORTABLE SURVEY METERS
(Kathren, 1974)

Ionizing Radiation	Type of Instrument	Comments
Beta Radiation		
Low Level	Geiger Counter	Readings should be made with window open and should not be considered as dose rate measurements but merely an indication. Closed window response should be subtracted to provide better estimates. Highly energy dependent.
High Level	Ion Chamber	
Photon Radiation		
Low Level	Geiger Counter & Ion Chamber	In mixed beta-photon fields, readings should be made with window closed. In pure gamma fields the arithmetic average of the open and closed window readings will provide a more accurate measurement. Cannot be used in pulsed fields.
High Level	Ion Chamber	
Alpha Radiation	Air Proportional	Susceptible to altitude (atmospheric pressure) changes: also can be adversely affected by humidity.

[redacted]

[redacted]

TABLE 4.1: OVERVIEW OF PORTABLE SURVEY METERS CONTINUED
(Kathren. 1974)

Ionizing Radiation	Type of Instrument	Comments
Alpha Radiation	Gas Flow & Proportional	Bulky, use time limited by gas bottle contents; requires purging.
Alpha Radiation	Scintillation	High Sensitivity; susceptible to light leaks. Pinhole in window can cause spurious response.

5.0 LABORATORY METHODS OF RADIONUCLIDE DETECTION AT THE FMPC

The purpose of this section is to provide an overview of the scope and principles developed by the analytical laboratory at the FMPC to determine isotopic analyses. Appendix A contains the analysis file print-out for CHOICE samples. This print-out provides a variety of information about the isotopic labboratory, such as the different analyses which are performed, method number, analytical procedure, and default units determined in the analyses. Two of the methods listed in Appendix A, method number 4024 - isotopic thorium analyses and method number 6003 (now 6006) - isotopic uranium-235 analysis are in draft form and are in the process of being obtained.

Determination of Isotopic Uranium (wt %) - Method No: 6005

Method number 6005 describes a thermal ionization mass spectrometry (TMS) procedure for determining isotopic uranium. This method is used to determine uranium isotopic analysis, regardless of the isotopic concentrations, from microgram quantities of uranium. Uranium is initially separated into its matrix material and then purified prior to the analysis.

[redacted]

The TMS analysis for isotopic uranium takes about two hours. Once the purified uranium sample, U_3O_8, is obtained it is dissolved with nitric acid. The solution of the purified uranium is deposited on two filaments and dried. These filaments are attached to the ion sources and then inserted into the spectrometer.

The material on the filaments is thermally excited, ionized, and accelerated through the magnetic field for ion separation. The isotopes are measured individually and sequentially using an electron multiplier by changing the accelerating potential on the ion source. The intensity of each isotopic species is proportional to the abundance of that species in the original sample. The total of all isotope signals measured represents the total uranium signal, therefore, each individual isotope signal is a percentage of the total.

Radiometric Determination of Thorium-234 and Pa-234m in Effluents - Method No: 4022

Method 4022 describes the analysis of thorium-234 and protactinium-234m radionuclides in water effluents by gamma spectrometry using a germanium (Ge) detector. This method is applicable to effluents containing less than 2.5 g/L of soluable impurities. The activity of a one liter sample measured for 24 hours has a lower limit of detection of 1 pCi/L. After the sample is appropriately prepared, it is placed in a 100 mL polystyrene vial on a gamma spectrometer. The efficiency corrected net counts for the 93 keV (Th-234), 765 keV and 1001 keV (Pa-234m) photopeaks are used to calculate the specific activities of the two radionuclides of interest. An NBS mixed radiometric standard reference material is used to provide efficiency corrections for the photopeaks measured. Measured activities are also corrected for ingrowth and decay, back to the sampling time.

Radiometric Determination of Radium-226 and Radium-228 in Various Matrices - Method No: 4002

Method 4002 describes the analysis of sump effluents and production by-products materials that are soluable in mineral acids. This method is used to separate, and analyze, Ra-226 and Ra-228 from solutions containing uranium, thorium, decay products, and nonradioactive materials. Typical analysis time for both Ra-226 and Ra-228 is three days. The radium isotopes can be determined to a lower detection limit of 0.010 d/m-mL.

[redacted]

Radium is separated from other interfering elements using cation exchange and coprecipitation with barium from a concentrated nitric acid solution. The precipitate is further purified by passage through heated cation exchange columns. Ac-228 is allowed to ingrow into a purified radium solution. its activity is proportional to the amount of Ra-228 present in the sample. The Ra-226 alpha activity is measured on a solid state alpha spectrometer. The Ac-228 beta activity is measured, following elution of these radionuclides from the resin, using a low background proportional counter.

Radiometric Determination of Plutonium - Method No: 4019

This method is applicable to the separation of plutonium from uranium fission products, soil, vegetation, and effluents. Analysis time is dependent on sample matrix and activity level. Sample preparation and purification are accomplished in 6-7 hours while the actual analysis time requires an additional 24 hours. The method has an approximate lower limit of detection of 0.35 d/m-mL.

The appropriately prepared sample of plutonium is inserted into the alpha spectrometer and the plutonium activity is measured on the silicon surface barrier detector. A correction is made for variations in plating (preparing the plutonium sample) using a neptunium-237 spike.

Radiometric Determination of Neptunium-237 - Method No: 4003

The method is applicable to the separation of neptunium (Np) from uranium fission products, soil, vegetation, and effluents. Analysis time is dependent on sample matrix and activity level. Sample preparation and purification are accomplished in 6-7 hours and counting time requires an additional 24 hours. Precision values are progressively better for higher concentration levels. The method has a lower limit of detection of 0.27 d/m-mL of sample.

Radiometric Determination of Technetium-99 - Method No: 4007

This method is applicable to various uranium and copper matrices that can be solubilized in weak acids or a basic media. The lower detection limit for technetium-99 in uranium is 2.25 (U Basis) parts per billion (ppb) with a relative precision of +/- 21.1% (95% confidence limit). The lower detection limit for Tc-99 in copper is 20.3 ppb (U Basis) with a relative precision of +/- 24.8% (95% confidence limit). Unsatisfactory results occur when NO_3^- is present, therefore nitric acid is not used for digestion.

[redacted]

Radiometric Determination of Cesium-137 - Method No: 4006

This method describes the radiometric determination of cesium-137 (Cs-137) in uranium matrices. Self-absorption losses may be considered negligible if the sample contains less than four grams per liter of cesium. A lower detection limit of five disintegrations per minute per milliliter, is obtained by ensuring that the counting precision is at least 10% (relative to its own count rate). An individual Cs-137 determination can be performed in 4 to 6 hours.

[redacted]

[redacted]

6.0 RI/FS RADIOACTIVITY MEASUREMENT INSTRUMENTATION SPECIFIED IN QAPP

The purpose of this section is to list the instruments utilized by the International Technologies Laboratory to analyze RI/FS samples. The instruments are listed for radioactivity type as well as specific radionuclides. This information was obtained from the draft document of the Quality Assurance Project Plan (QAPP).

Radionuclide/Radioactivity	Instrument
Gross Alpha	Tennelec Low background alpha/beta counting system Ludlum Scaler Scintillation Counters Models: 1000, 2000, 2200.
Gross Beta	Tennelec low background alpha/beta counting system.
Gamma Spectrometry	Intrinsic Germanium Detector and Nuclear Data Computer based Gamma Spectrometer, ND9900.

Radionuclide	Instrument
Radium-226	Alpha Spectrometer and/or Nuclear Data Computer based Gamma Spectrometer, Model ND9900.
Radium-228	Tennelec Low Background alpha/beta Counting System.
Strontium-90	*Same as Radium-228.*

[redacted]

[redacted]

Radionuclide	Instrument
Technetium-99	Packard Liquid Scintillation Spectrometer.
Isotopic Thorium	Alpha Spectrometer and Nuclear Data Computer based Gamma Spectrometer, Model ND9900.
Uranium Total	Alpha Spectrometer
Uranium Isotopic	Nuclear Data Computer based Gamma Spectrometer, Model ND9900.
Isotopic Plutonium	Alpha Spectrometer and Nuclear Data Computer based Gamma Spectrometer, Model ND9900.
Neptunium-237	Alpha Spectrometer and Nuclear Data Computer based Gamma Spectrometer, Model ND9900.
Cesium-137	Intrinsic Germanium Detectors and Nuclear Data Computer based Gamma Spectrometer, Model ND9900.

[redacted]

[redacted]

7.0 REFERENCES

Cember, H., 1987. *Introduction to Health Physics*. 2nd ed. Pergamon Press, New York.

Knoll, G. F., 1979. *Radiation Detection and Measurement.* Wiley, New York.

U.S. Department of Energy, 1980. *A Guide to Reducing Radiation Exposure to As Low As Reasonably Achievable (ALARA),* Contract No: EY-76-C-06-1830, prepared by R. L. Kathren and J. M. Selby, Pacific Northwest Laboratory.

WMCO, 1989. *Radiometric Procedure for the Determination of Cesium-137,* Method No: 4006.

WMCO, 1986. *Radiometric Determination of Radium-226 and Radium-228 in Various Matrices,* Method No: 4002.

WMCO, 1988. *Radiometric Determination of Th-234 and Pa-234m in Effluents,* Method No: 4022.

WMCO, 1989. *Radiometric Procedure for the Determination of Technetium 99*, Method No: 4007.

WMCO, 1989. *Radiometric Procedure for the Determination of Neptunium-237,* Method No: 4003.

WMCO, 1987. *The Determination of the Uranium Isotopes (wt %) by Thermal Ionization Mass Spectrometry (TMS),* Method No: 6005.

WMCO, 1989. *Radiometric Procedure for the Determination of Plutonium,* Method No: 4019.

Evans, R. D., 1955. *The Atomic Nucleus,* McGraw-Hill, New York. [redacted]