Active Nondestructive Assay of Nuclear Materials

Principles and Applications

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PREFACE

The development of methods for the nondestructive assay (NDA) of nuclear materials virtually started at the dawn of the nuclear era. The fact that nuclear materials, in most cases, emit characteristic radiations made the detection and the quantitative determination of their presence via these radiations a very natural step. However, the status and use of NDA remained limited until the mid-sixties. At that time, the rapid growth of the nuclear power industry and the commercial ownership of nuclear material motivated two comparatively large research and development efforts concerning NDA, especially active NDA (ANDA), to supplement and complement the chemical-analytical and other physical methods used to quantitatively measure nuclear materials. These two programs were instituted by the Office of Safeguards and Material Management of the U.S. Atomic Energy Commission under the late General Delmar L. Crowson. The first program was started in 1966 at the Los Alamos Scientific Laboratory and was based initially on the utilization of 14-MeV neutrons as the main interrogation source for ANDA. The second program, started in late 1967 at the Linac Department of Gulf General Atomic, was based on the use of high-energy gamma rays generated by a linear accelerator as an interrogation source for ANDA. Intensive developmental work in various passive and active NDA methods paralleled the two main efforts, including the following examples: calorimeters for plutonium assay in Mound Laboratory of Monsanto Research Corporation; the Helix counter for plutonium assay at Rocky Flats; the neutron "well" counter at Brookhaven National Laboratory—which then became a leader in evaluating various ANDA systems; and the slowing-down lead spectrometer for reactor fuel pin assay by Inter-atom in Germany.

The main motivation for the rapid development of active NDA was the need to reduce the uncertainties in measurements of fissile material present in mixtures and configuration like waste and scrap, which are either difficult or impossible to measure with conventional chemical-analytical techniques. The reduction of measurement uncertainties

improves the performance of nuclear accountability systems and, hence, the entire safeguards systems.

Though the prime motivation for the development of NDA was nuclear material safeguards, its development brought forth another important benefit: it provided the nuclear industry with an important tool with which to accurately measure the fissile content of finished products, like fuel rods, or the fissile content of various earlier process states, e.g., prior to scrap recovery or waste disposal. Thus, the development of NDA (passive and active) gave the industry new devices which improved both process and quality control. An outstanding example of this is the high-speed fuel rod scanners which are now being used by virtually all manufacturers of low enrichment fuel for light water reactors.

After more than a decade of development, active NDA techniques have reached a certain maturity. The principles, the pitfalls, and the applications of ANDA are well understood. A variety of techniques and commercial instruments are available for measuring the various fissile materials in different compositions and forms. The purpose of this book is to present, coherently and comprehensively, the wealth of available but scattered information on the principles and applications of ANDA. In some instances, gaps existing in the available information have been filled by new, unpublished work by the author and others or with information or data obtained through private communications. In order to promote wide and proper utilization of ANDA, the book is directed to a broad readership: to supervisors of measurements, to instrument operators, and to the nuclear material managers. The book is also intended for the licensing, inspection, and enforcement personnel of both the Nuclear Regulatory Commission (NRC) and the Department of Energy (DOE). System designers and the instrument engineers, including nondestructive testing specialists, as well as graduates of nuclear engineering, will find a wealth of material and references to aid them in developing new ideas or modifying existing ones.

In an attempt to be responsive to such a wide audience, the book is divided into ten chapters. The first chapter provides a background and overview of the entire subject of ANDA, and it could be sufficient for generalists who do not feel the need to go into further detail. The next nine chapters are more detailed and cover ANDA principles (Chapters 2 through 7), applications and electronics (Chapters 8 and 9), and finally, measurement control (Chapter 10). Each chapter has an introductory section describing the relationship of the topic of that chapter to ANDA. Each chapter ends with a section that summarizes the main results and

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conclusions of the chapter, and a reference list. This system of organization will allow the reader to select, within some limits, those parts of the text which are of specific interest to him without his having to read the entire book.

The first buds of this book appeared several years ago; however, the support given in the last few years by the NRC Office of Standard Development through Mound Laboratory of Monsanto Research Corporation was essential to its relatively early conclusion. I would therefore like to express my thanks to Messrs. P. Ting, E. Selleck, and R. J. Jones of NRC. The patience and indulgence of Dr. W. Strohm, who monitored the contract for Mound Laboratory are deeply appreciated as well as his critique and comments. I would also like to acknowledge the help given to me in preparing this manuscript through discussion, written material, and photographs: the safeguards group (Q1) at Los Alamos Scientific Laboratories was very helpful, especially Drs. R. Walton and H. Menlove. Drs. J. Steward and F. Scheonig (GE Nuclear Fuel Division in Wilmington, N.C.); Dr. M. Zucker (Brookhaven National Laboratory); Mr. H. Miller (National Nuclear Corp.); Mr. H. Weber and Dr. D. Bramblett (IRT Corp.); Dr. G. Birkhoff (Euratom, Ispra, Italy); Dr. H. Bucker (Jülich, W. Germany) and many others provided information or written material and deserve my sincere thanks.

This book would probably not have reached completion without the benefit of contributions from two renowned experts in the safeguards area: Professor R. Sher of Stanford University and Dr. J. Glancy of Science Applications, Inc. The results of Professor Sher's comments, writing, critique, and revisions can be found throughout the entire book. Dr. Glancy's general comments and his major contributions to the first and last chapters are gratefully acknowledged.

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Finally, this book would never have been written without the continual encouragement, over many years, from my parents, Rivcca and Arieah Gozani, many miles away, and my wife, Adit. As a small token of my appreciation, I would like to dedicate this book to them.

If this book contributes to a wider and better use throughout the entire nuclear fuel cycle of active NDA methods, facilitating the rapid implementation and safe usage of nuclear energy, an essential energy source, then our toil has been worthwhile.

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CHAPTER 1

BACKGROUND AND OVERVIEW

1.1 INTRODUCTION

Active nondestructive assay (ANDA) is a subset of the relatively new measurement science of nondestructive assay (NDA). In this chapter we introduce the principles underlying different types of NDA and discuss the applications of NDA to the nuclear industry. This discussion highlights differences between ANDA and the passive NDA (PNDA) techniques with which the reader may be more familiar (Ref. 1) and describes the special role of ANDA in safeguards and process or quality control. The advantages and disadvantages of ANDA in various applications are contrasted with those of PNDA and destructive assay techniques based on sampling and chemical analysis. A brief description of the fundamentals of ANDA, which introduces the reader to the concepts and terminology used in this book, concludes this chapter.

1.2 PRINCIPLES OF ANDA

Fissile and fertile nuclei* spontaneously emit characteristic nuclear radiations, such as neutrons and γ -rays, that are sufficiently energetic to penetrate the material and its container or cladding. In addition, when

*Fissile isotopes can be fissioned by low energy (thermal) neutrons. Fertile isotopes require energetic neutrons, above about 0.5 MeV, to induce substantial amount of fission. Fertile isotopes can be transmuted to fissile isotopes. Fissionable isotopes include both kinds. Fissile isotopes are ^{233}U , ^{235}U , ^{239}Pu , and ^{241}Pu . Fertile isotopes are ^{232}Th , ^{234}U , ^{236}U , ^{238}U , ^{236}Pu , ^{238}Pu , ^{240}Pu , ^{242}Pu , and ^{244}Pu . In addition, fissionable isotopes may undergo spontaneous fission. All heavy isotopes undergo fission when bombarded with high-energy γ -rays (>5 MeV). This phenomena is called photofission and is discussed in Chapters 3 and 7.

irradiated by neutrons or γ -rays, certain nuclear materials fission,* emitting highly energetic radiations. Nondestructive assay (NDA) is the observation of spontaneous or stimulated nuclear radiations to determine the amount of one or more nuclear materials in a sample without affecting the sample's physical or chemical form.** NDA can be contrasted with the more traditional, destructive assay of nuclear material performed by sampling and chemical analysis, in which the physical and chemical properties of the sample are altered.

Uranium and plutonium are the nuclear materials commonly assayed nondestructively. Isotopes of each of these materials decay radioactively, either by emitting α -particles (helium nuclei) or β -particles (electrons) and their associated γ -rays, or by fissioning (emitting fission products and their associated neutrons and γ -rays). Table 1-1 lists radiations from uranium and plutonium nuclides commonly observed by NDA. The second column indicates whether α - or β -particle decay is predominant and whether the nucleus will spontaneously fission without stimulation by neutrons and γ -rays. The third and fourth columns give the spontaneous radiations detected by NDA and the intensity or number of radiations emitted per second per gram of the nuclide. The following radiations are listed: γ -rays emitted following α - or β -particle decay; neutrons emitted from spontaneous fission, and neutrons emitted following the reactions of α -particles and other nuclei—called (α, n) reactions. These (α, n) neutrons are emitted from all nuclear materials disintegrating by α -decay, if the nuclear material is mixed with an element having a low atomic number, such as oxygen or fluorine. The fifth column gives the amount of power emitted by spontaneous radioactive decay per gram for plutonium isotopes. This power can be detected by a calorimeter to determine nondestructively the plutonium content.

The last column of Table 1-1 gives the fission cross section*** of the nuclides for fission by low-energy neutrons in thermal equilibrium with the surrounding material. Such neutrons are called thermal neutrons. Cross sections are measured in units of area called barns ($1 \text{ barn} = 10^{-24} \text{ cm}^2$). The larger the cross section, the larger fission probability. Thus, the data in the last column of Table 1-1 show that the probability of thermal neutrons causing fission in nuclides with an odd atomic number— ^{235}U , ^{239}Pu , and ^{241}Pu —is high, but it is low for nuclides with even

*Fission is the splitting or breaking up of an atomic nucleus into two or more smaller nuclei with release of a large amount of energy. A small fraction of this energy is radiated as γ -rays and neutrons.

**Only negligible radioactivity is induced in the material.

***The fission cross section indicates the probability that the nuclide will fission when irradiated with neutrons or γ -rays.

Table 1-1. Natural radiations from U and Pu isotopes observed by NDA.

Spontaneous Radiation Observed in NDA ^a					Thermal ^c Neutron, Fission Cross Section (b)	
Nuclide	Decay Mode	Radiation, Type	Intensity (s ⁻¹ . g ⁻¹)	Calorimetry, Specific Power ^b (W/g)		
²³⁵ U	α-particle	185.7 keV γ-ray	4.3 × 10 ⁴		577 (fissile)	
²³⁸ U	α-particle	1001.1 keV γ-ray ^d	1.0 × 10 ²		<0.5	
		766.4 keV γ-ray ^d	3.9 × 10 ¹			
	fission	fission neutrons	1.7 × 10 ²			
²³⁸ Pu	α-particle	766.4 keV γ-ray	1.5 × 10 ⁵	5.6716 × 10 ⁻¹	18.4	
		152.7 keV γ-ray	6.5 × 10 ⁶			
	(α,n) neutrons, PuO ₂	1.4 × 10 ⁴				
	fission	fission neutrons	2.6 × 10 ³			
²³⁹ Pu	α-particle	413.7 keV γ-ray	3.4 × 10 ⁴	1.9293 × 10 ⁻³	741 (fissile)	
		129.3 keV γ-ray	1.4 × 10 ⁵			
	(α,n) neutrons, PuO ₂	45				
²⁴⁰ Pu	α-particle	no useful γ-rays	—	7.098 × 10 ⁻³	0.03	
		(α,n) neutrons, PuO ₂	170			
	fission	fission neutrons	1.0 × 10 ³			
²⁴¹ Pu	β-particle	208.0 keV γ-ray ^d	2.0 × 10 ⁷	3.39 × 10 ⁻³ ^e	950 (fissile)	
		164.6 keV γ-ray ^d	1.8 × 10 ⁶			
		148.6 keV γ-ray	7.5 × 10 ⁶			
²⁴² Pu	α-particle	no useful γ-rays	—	1.146 × 10 ⁻⁴	—	
		(α,n)neutrons, PuO ₂	10			
	fission	fission neutrons	1.7 × 10 ³			

^a γ-ray energies and insensities and (α,n) neutron intensities from PuO₂ (Ref. 1) Fission neutron intensities (Ref. 2).

^b Specific powers (Ref. 3)

^c Thermal neutron fission cross sections given in barns for neutrons with velocity of 2200 m/s (Ref. 4)

^d γ rays from daughter products

^e This value includes 1.1423 × 10⁻¹ W/g from ²⁴¹Am daughter product

atomic numbers. Odd numbered nuclides are called fissile because they fission when irradiated by thermal neutrons. Even numbered nuclides are called fertile. Neutrons with energy exceeding a threshold energy level are necessary before any significant fission will take place in fertile nuclides.

A wide variety of detectors and detection schemes can be used to observe these radiations. All detectors work essentially on one of three schemes: (1) gross counting of all radiation emitted, (2) net counting of radiation within a narrow energy band, or (3) coincidence or correlation counting of radiation bursts containing two or more radiations correlated in time. For orientation purposes, all NDA instruments and methods can be classified into the four types shown in Figures 1.1 and 1.2. A brief description of the principles of each technique is given below.

1.3 PASSIVE γ -RAY NDA

γ -Rays emitted spontaneously as a result of radioactive α - or β -particle decay have energies characteristic of the particular nuclide from which they come. As shown in Figure 1.1A, some of these γ -rays have sufficient energy to penetrate fissionable material, the nonfissionable matrix in which the material is dispersed, and the container or cladding. Detection of these γ -rays results in electronic pulses proportional to their energy that can be discriminated from pulses belonging to background γ -rays. The net number of counts is interpreted through a predetermined calibration to estimate the kind and amount of nuclide present.

A transmission measurement is a commonly used supplementary measurement when accurate γ -ray assay is required. In this measurement the sample is irradiated by an external source of γ -rays of an energy similar to the ray of interest. Such a transmission measurement allows the correction of the assay for attenuation within dense materials; it has therefore extended the applicability of γ -ray NDA to materials so dense that only a fraction of the spontaneous γ -rays emitted can penetrate the material. If the container is close to the detector, it is usually rotated while it is scanned by the detector, or it is placed at a sufficient distance from the detector to reduce geometrical errors and improve uniformity of the sample response.

An important distinction among various passive γ -ray NDA techniques lies in the ability of each detector to resolve the energy difference between different γ -rays. This energy resolution ranges from medium values characteristic of NaI(Tl) detectors to high values of Ge(Li) or

Figure 1.1. Simplified schematic representation of PNDA.

Figure 1.2. Simplified schematic representation of ANDA.

hyper-pure Ge detectors. Other detectors like Cd(Te) (cadmium-telluride) and BGO (bismuth germanate) detectors have also medium energy resolution. Although passive γ -ray analysis provides information about both the identity and quantity of individual nuclides, there are many materials with densities too high for useful application of passive γ -ray NDA.

1.4 PASSIVE NEUTRON NDA

Neutrons are emitted spontaneously as a result of radioactive decay by fission or as a result of reactions of α -particles with nuclides of low atomic number in the matrix material.* These neutrons have high energies and can penetrate the fissionable and matrix material as well as the container or cladding. As shown in Figure 1.1B, these neutrons are detected by multiple detectors that surround the container, and the gross number of electronic pulses generated by the detectors is counted. The neutron energies are not unique to a specific nuclide, therefore, narrow energy selection is usually not performed. The gross neutron counts are interpreted in terms of the total content of neutron-emitting nuclides. The amount of one specific nuclide can be determined, if the relative abundance and neutron intensity of each neutron-emitting nuclide are known. This technique is limited because spontaneous fission takes place predominantly in ^{238}U and some plutonium isotopes. The gross neutron counting technique is further limited because it requires knowledge, not only of the abundances of the radionuclides but also of the abundances of elements with low atomic number that are targets for the (α, n) reaction.

*The matrix material is all nonfissionable material in the sample.

Spontaneous fission neutrons are emitted in bursts of approximately two or three neutrons simultaneously in time. These neutrons can be distinguished from neutrons resulting from (α, n) reactions (which are emitted singly at randomly spaced intervals) by use of a coincidence gate* that is open only for a short time following detection of the first neutron. The probability of detecting an additional fission neutron in the short time following detection of the first neutron is relatively high because the fission radiations are coincident, that is, they are all emitted simultaneously. On the other hand, the (α, n) neutrons are emitted singly, that is, the probability of detecting an additional (α, n) or any accidental neutron in this interval is no greater than it would be in any other random time interval of equal duration.

An important difference among NDA techniques based on passive neutron is the detectors, which may be gas-filled tubes that detect thermal neutrons or plastic scintillators sensitive to both high-energy neutrons and γ -rays. When plastic scintillators are used, one can count fission neutrons and the fission γ -rays that are coincident with them, or shield the detectors from the γ -rays and count only neutrons, or vice versa. Plastic (and most organic) scintillators have several advantages over other neutron detectors: time response is much faster, pulses are higher, and they can measure coincident radiation in the presence of high background random neutron radiation. The disadvantages are poorer stability, sensitivity to γ -rays, and generally less ruggedness.

1.5 PASSIVE CALORIMETRIC NDA

The energy associated with radioactive decay is contained mainly in the α - and β -particles that do not escape from the radioactive material and its container or cladding. This energy is, therefore, constantly absorbed by the nuclear material, making the material a continuously emitting heat source. When placed in a calorimeter, the radioactive nuclear material increases in temperature because the calorimeter is isolated. Heat cannot be transferred as readily as when the container is cooled by air. As the temperature of the nuclear material increases, more heat is transferred to the calorimeter until the temperature is so high that the heat flow across the resistance thermometer or thermocouple, shown in Figure 1.1C, approaches the nuclear heat generation rate. The measured temperature is interpreted by a calibration in terms of the thermal power of the nuclear material. This thermal power can then be

*Pulse correlation circuit, performing correlations on the pulse train from the detector over a time period of the order of the lifetime of neutrons in the detector, can also be used.

used to determine the total nuclear material content if the relative abundance of all radionuclides in the material is known. Calorimetry is limited to the measurement of plutonium because of the high heat generation rate of plutonium. Its accuracy depends primarily on the precise knowledge of radionuclidic abundances, including the plutonium decay product americium-241 (^{241}Am), that contribute significantly to heat generation.

1.6 ACTIVE NDA (ANDA)

Neutrons or γ -rays which irradiate nuclear material induce fissions that produce, on the average, two to three highly energetic prompt fission neutrons and approximately eight prompt γ -rays. These radiations are called prompt fission radiations because they are emitted at the time of fission. For up to several minutes after fission, the fission product nuclides continue to emit another six to seven γ -rays and approximately 0.01 to 0.02 n/fission. These radiations are called delayed fission radiations. Both prompt and delayed fission radiations can be detected in a manner similar to that previously described for passive neutron counting.

Some ANDA techniques utilize the detection of prompt radiation while the material is being irradiated and others depend on the detection of delayed radiation after irradiation has been terminated. If prompt radiation is detected, the fission neutrons and γ -rays must be distinguished from the irradiation neutrons and γ -rays. As shown in Figure 1.2, radiation from the neutron or γ -ray source is incident upon the nuclear material, but it may also be incident upon the detectors, or it may scatter into the detectors after interacting with the nuclear material or the counting chamber. The discrimination of prompt fission neutrons from the radiation source can be accomplished by using detectors biased to count only neutrons having energy greater than the energy of the source neutrons. An alternative method is to use a random radiation source and coincident counting of the correlated fission radiation. In coincidence counting, it is also necessary either to shield the detectors from the source or to bias the detectors above the energy of the source radiations so that the accidental coincidences from the random source neutrons and γ -rays do not create a large background. Accidental coincidences can be significant, if the number of random radiations detected during the coincidence gate time is large. In the detection of delayed radiation, the irradiation source is quickly removed prior to counting, therefore, gross counting can be used with or without energy discrimination. The source is removed by turning it off, employing a movable shutter, or moving the nuclear material past the source to a shielded counting chamber.

Another important difference among ANDA techniques lies in the type of radiation emitted by the source. Such a source is commonly called an interrogation source because it sends out a signal (interrogating radiation) that triggers an appropriate response from the nuclear material. Interrogation sources are of two types: (1) radioactive isotopic sources and (2) electron, deuteron, or proton accelerator sources. The radioactive sources all emit neutrons, and some also emit γ -rays. Deuteron and proton accelerators serve to produce neutrons and electron accelerators yield γ -rays which can produce neutrons via (γ, n) reaction. Common radioactive sources are (1) californium-252 (^{252}Cf) spontaneous fission sources, (2) alpha-neutron (α, n) sources, such as Am-Li or Pu-Li which emit relatively low-energy neutrons and Am-Be or Pu-Be which emit higher energy neutrons, and (3) photoneutron (γ, n) sources, such as Sb-Be which emit low-energy neutrons. The (α, n) and (γ, n) sources are identified by referring first to the radioactive element emitting the α -particle or γ -ray and then the target element. For example, Am-Li refers to an ^{241}Am α -particle source with a Li target that undergoes reaction with the α -particle to produce a neutron [and a boron (B) nucleus].

The energy of neutrons produced by proton accelerators can be adjusted by using different target elements that will produce neutrons by proton-neutron (p, n) reactions. The maximum energy of the γ -rays produced by electron accelerators determines the maximum neutron energy created by the (γ, n) reaction. The γ -energy can be adjusted by changing the electron energy.

As shown in Figure 1.2, a neutron-moderating assembly is commonly added to an ANDA system between the interrogation source and the nuclear material so that the energy of the interrogating radiation is modified or "tailored" to suit a particular application. Selection of the interrogation source and the tailoring assembly is extremely important because the energy of the interrogating radiation determines the intensity of the response and the measurement accuracy. Tailoring is necessary because both the fission cross section and the penetration of radiation through nuclear material are energy dependent. As a simple illustration of the importance of this feature of ANDA, consider the use of a high-energy neutron source with a tailoring assembly to filter out any low-energy neutrons. The interrogating radiation is highly penetrating, giving a uniform response throughout the nuclear material, but it also gives a low intensity response because the fission cross section is low at high-neutron energy. If the tailoring assembly is modified to moderate the source neutron energy so that the interrogating radiation is primarily low-energy neutrons, then the intensity of the response will greatly increase because of the increase in the fission cross section. However, the

response will no longer be uniform throughout the nuclear material because the low-energy neutrons are not highly penetrating.

It should now be obvious that there are a variety of ANDA techniques that can be used. Table 1-2 presents a list of 23 ANDA techniques.

Table 1-2. Active nondestructive assay techniques.

Radiation				
No.	Source Type	Incident	Observed	Applications
<u>Isotopic Sources</u>				
1	^{252}Cf or (α, n)	Moderated/unmoderated	Prompt neutrons (n_p) and γ rays (γ) using fission multiplicity detectors	Useful for scrap and waste. Sample size up to 55 gal. moderate sensitivity
2	^{252}Cf	Moderated Cf fission neutrons	Gross delayed gamma (γ_d) (possibly n_p and delayed neutrons, n_d)	Samples with low enriched SNM. Most useful for rapid scanning of low enriched fuel pins
3 ^a	Low E (α, n) [e.g., ^{241}Am $(\alpha, n)\text{Li}$]	Unmoderated/partially moderated neutrons	n_p and γ (using FMD)	Same as #1. Source intensity limited
4 ^a	Low E (α, n) [e.g., ^{241}Am $(\alpha, n)\text{Li}$]	Highly/partially moderated	Fast n_p	Low speed LWR fuel rod scanner
5	(γ, n) [e.g., Sb-Be, Ra-Be, etc.]	Sub-MeV neutrons	n_p, n_d	All degrees of SNM ^b enrichments. Various size samples including fuel pins
<u>Accelerators</u>				
6	Electron accelerator	5 to 10 MeV γ -ray (Bremsstrahlung)	Thermalized n_p	
7	Electron accelerator	5- to 10 MeV γ ray (Bremsstrahlung)	n_d and n_p/n_d	All containers, all compositions. Instrument system is complex. Can distinguish between fissile isotopes
8	Electron accelerator	5 to 10 MeV γ -ray (Bremsstrahlung)	γ_d (using low or high resolution detectors)	
9	14 MeV neutron	Unmoderated/partially moderated neutrons	n_d	All containers, all compositions. Low H. Instrument system is complex
10 ^c	14 MeV neutron	Unmoderated/partially moderated neutrons	γ_d	
11 ^c	3-MeV Van de Graaff	Subthreshold neutrons	n_p, n_d	Small samples (no H) with high enriched SNM. Fixed and complex installation. Distinguishes between two fissile isotopes
12	Electron accelerator	MeV/sub-MeV neutrons	n_d	Small or large samples with low to intermediate density of SNM. Complex system
13	14-MeV neutron generator	Low energy, lead moderated neutrons	n_p	Fuel pins with low enrichment. Fixed and complex installation
14	14 MeV neutron generator	High-energy, lead moderated neutrons	n_p (and possibly n_d, γ_d)	Fuel pins with medium size samples (no H) complex system as #13
15	Medium E Linac	Neutrons (via time of flight)	Capture γ	Limited applicability. Can distinguish between several fissile and fertile isotopes. Fixed installation
<u>Reactors as sources</u>				
16	Reactor	Thermal neutrons	n_d	Very sensitive and accurate method for small samples. Good for standards. Fixed installation
17	Reactor	Thermal neutrons	High resolution fission product γ -rays	As in #16, but limited applicability. Can distinguish between ^{239}Pu and ^{235}U . Fixed installation
18	Reactor	Low E (eV) neutrons	Prompt neutrons and neutron transmission	Limited applicability

continued

Table 1-2. Active nondestructive assay techniques (continued).

No	Source Type	Radiation		Applications
		Incident	Observed	
19	Reactor	Low E thermal and low eV neutrons	Capture γ	Limited applicability Can distinguish between several fissile and fertile isotopes Fixed installation
20	Zero power reactor	Thermal neutrons	Reactivity change	Very sensitive Fixed installation Low enrichment Limited applicability
21	Zero power fast reactor	Fast neutrons	Reactivity change	Very sensitive Useful for high enrichment Fixed installation in national labs Limited applicability
<u>x ray and γ-sources</u>				
22	x-ray machines or radioactive sources like ^{241}Am	Filtered and unfiltered x rays	Fluorescent x rays	Complementary rapid method to analytical chemical methods Requires careful sample preparation
23	Radioactive γ sources (^{241}Am , ^{57}Co , ^{137}Cs , ^{60}Co , etc.)	γ rays	Transmitted γ rays	Density determination Low Z/high Z composition determination (e.g., SNM concentration in nitrate solution)

*"Low E" means here neutrons with energy below approximately 0.5 MeV. The very qualitative definitions of "high" and "low" energy, "hard" and "soft" spectrum etc., vary somewhat throughout the book and should be understood within the specific context these adjectives are used.

^bSNM is an acronym for special nuclear material e.g. ^{235}U , ^{239}Pu

^c"Low H" or "no H" mean respectively, low contents of hydrogenous material and absence of such material in the matrix

ANDA also offers a flexibility not found in passive NDA techniques, as the following list, contrasting active and passive techniques, demonstrates:

1. The energy and intensity of the ANDA interrogating radiation can be increased or decreased to provide appropriate response intensity. Passive techniques are limited to the response intensity provided by nature.
2. The energy of the interrogating radiation can be adjusted to generate different responses from different nuclides. This adjustment is primarily useful in measuring first fissile nuclides using low-energy interrogating radiation, and then measuring fertile nuclides using interrogating radiation with energies above the fission threshold. Passive neutron counting measures primarily fertile nuclides, as does calorimetry. Passive γ -ray NDA measures the content of an individual nuclide only, if nature has provided a penetrating γ -ray for that nuclide.
3. ANDA is applicable to both uranium and plutonium, as is passive γ -ray NDA. Passive neutron counting is applicable to plutonium and to low-enrichment uranium, but it is only commonly used for plutonium. Calorimetry applies only to plutonium. In contrast to γ -ray NDA, which can be applied only to uranium compounds and matrix materials with low-bulk density, ANDA can be applied to both low- and high-density materials.

4. Primarily because the interrogating radiation can be adjusted and turned on and off, ANDA measures nuclear materials in the presence of large radiation backgrounds, such as those produced by radioactive fission products.

In spite of the flexibility of ANDA, there are many cases where passive techniques prove as accurate as ANDA and are selected because they are inherently simpler. There are also cases where PNDA is more accurate than ANDA because the sources of error that affect only the spontaneous radiation from PNDA affect both the response and the interrogating radiation from ANDA, thus compounding the error. Presentation of the fundamentals of ANDA in the next chapters will explain the obstacles to accurate assay and show when and how they can be overcome.

The descriptions of the four basic types of NDA techniques (Sections 1.3-1.6) cover briefly the principles involved. A more detailed description of passive γ -ray and passive neutron NDA is presented in "Fundamentals of Passive NDA" (Ref. 1). The fundamentals of passive calorimetric NDA are found in (Ref. 3 and 5).

1.7 APPLICATIONS OF NDA

In the nuclear industry there are four common applications of NDA: (1) nuclear materials accounting, (2) personnel and package search for nuclear material as a safeguard against theft, (3) verification of prior assay, and (4) nuclear materials process and quality control. ANDA can be used in all of these applications. With the exception of process or quality control applications, these uses are part of an integrated safeguards system protecting against theft of nuclear material. Each of these applications is described briefly with emphasis on special advantages or disadvantages ANDA may have.

1.7.1 Nuclear Material Accounting

Nuclear material accounting is one of the most important safeguards against diversion of special nuclear materials—uranium enriched with ^{235}U , ^{233}U , and plutonium—by deterring and detecting thefts of these materials. For safeguards, accounting accuracies of $\pm 0.5\%$ of the material added to the fuel processing operation are required by government regulation to achieve sensitivity to out-of-control situations that may be caused by diversion. Such highly accurate accounting is also required for responsible financial control of operations processing these valuable nuclear materials. These high-accuracy accounting requirements

demand not only accurate measurement of the main material flow and inventory in a nuclear plant, but also measurement of material in scrap, waste, and residue remaining in process equipment after it is cleaned out (holdup). These latter forms of material cannot usually be measured by sampling and chemical analysis because the materials are not homogeneous which makes sampling difficult, and because they are often difficult to dissolve for chemical analysis.

Many nondestructive assay techniques were developed especially to fill the need for measurements of scrap, waste, and residue as seen by studying the example of an accounting material balance presented in Table 1-3. The material balance is divided into receipts (R), beginning plant inventory (BI), shipments (S), and ending plant inventory (EI). The material balance is computed by calculating the material-unaccounted-for (MUF) using the equation $MUF = (R + BI) - (S + EI)$. Each material in the balance is identified in the first column of Table 1-3 and mass is given in the second column. The typical relative measurement error is given in the third column and the absolute error, obtained by multiplying columns two and three, is given in the last column. Although, the masses of scrap, waste, and holdup residue are small relative to the other materials, it is obvious that they must be measured for proper accounting because they make up 13 to 17.5 percent of the receipts added to the process in this particular example. Of course, in

Table 1-3. An example for ^{235}U material balance.

Material		Measurement Error	
Form	Mass (kg ^{235}U)	(Relative Standard Deviation, %)	Absolute (kg ^{235}U)
Receipts- UF_6	1000	0.1	1.0
Beginning inventory			
UO_2 powder	410	0.1	0.41
UO_2 pellets	420	0.2	0.84
Scrap	100	5.0	5.0
Waste	20	25.0	5.0
Holdup	10	50.5	5.0
Shipment- UO_2 Fuel	960	0.2	1.9
Ending inventory			
UO_2 powder	400	0.1	0.40
UO_2 pellets	425	0.2	0.85
Scrap	150	5.0	7.5
Waste	10	25.0	2.5
Holdup	15	50.0	7.5

real plants the proportion may be greater or smaller. As shown in column four, the absolute measurement errors for scrap, waste, and holdup are the largest errors in the material balance. This may not be the case for all real plants, but these errors are still significant in any well-balanced measurement program (Ref. 6). A list of some common scrap and waste materials measured typically by NDA is presented in Table 1-4.

If possible, PNDA is used to measure scrap, waste, and residue because passive assays are inherently less complex than ANDA. However, ANDA should be used when one of the following conditions prevails:

1. No passive radiations exist, or the existing spontaneous emissions are not sufficiently penetrating for accurate NDA, or the spontaneous radiation cannot be distinguished from background radiation.
2. Spontaneous emissions are not intense enough for precise assay within a reasonable amount of time. The ANDA source can be strengthened to overcome most intensity problems where spontaneous emissions are limiting.
3. The attenuation of the spontaneous radiation is excessive. PNDA provides a choice of interrogation and stimulated radiation that reduces self-attenuation.

When the material is very uniform it can be measured by sampling and destructive analysis, but NDA, especially ANDA, may offer a more convenient or economical measurement. Table 1-5 lists materials that are commonly measured or could beneficially be measured by ANDA rather than PNDA or destructive analysis. Table 1-6 lists materials which could be measured by ANDA, but that are usually measured by PNDA. These tables are presented to summarize experience with ANDA and to represent one set of possible measurement strategies. The fundamentals of ANDA presented in the remaining chapters should guide the user in deciding whether ANDA is appropriate for his particular application and should make it possible for him to estimate approximate precision and accuracy (see, for example, Chapter 9).

1.7.2 Real-Time Nuclear Material Accounting

The selection of a nuclear materials measurement technique for safeguards or financial accounting is usually based on accuracy and economic considerations. Recently, however, an additional consideration for safeguards is receiving attention—the timeliness of measurement results. While present accounting can deter theft or detect it after the fact, an accounting system with timely measurements may function as part of preventive safeguards by detecting theft when it occurs, so that a

Table 1-4A. Common scrap and waste material measured by NDA
(Containers: 1 to 4 l bottles).

Description	Material	
	Nuclear Concentration (Weight, %)	Matrix
Ash	High variable, typically 1-5% but may be >10%	Product of combustion, low atomic number, bulk density ~ 2-3 g/cc
Insoluble residues	Typically less than a few percent	Product of repeated acid dissolution, low atomic number, bulk density depends on treatment. Initially, similar to sludge, is sometimes processed to powder after calcination and grinding. Generally aggregated particles of SNM.
Grinder sludge or dust	Variable, 20-30% (high concentration—if dry grinding process of pellets is used)	Metal oxide with lubricant. In dry grinding, oxide is mixed with grinder dust.
Process scrap	High, similar to process material	Contaminated process material such as chipped pellets, etc.
Clean-out sweepings	Variable, 20-80%	Contaminated process material from sweeping gloveboxes, scraping walls of process equipment, etc.

**Table 1-4B. Common scrap and waste material measured by NDA
(Variable containers).**

Material Description	Typical Container	Nuclear Material Concentration (Weight, %)	Matrix Material
Combustible waste	55 gal. drums	Very low, less than 100 g but typically 10 g/drum.	Low density paper, rags, some plastic, etc.
Non-combustible waste	55 gal. drums	Same as above.	Higher density than combustible wastes. Mostly plastics, rubber gloves, some metal, graphite furnace lines, glass parts, etc.
Filters and filter media	Various shapes and sizes. Most fit within 55 gal. drums.	Typically 10-50 g/ container	Filter media, such as fiberglass, asbestos, etc.
Waste boxes	4 ft (120 cm) x 4 ft (120 cm) x 4 ft (120 cm) boxes	Typically 0.5%/box	Assorted waste, including paper, rubber, metal, wood, plastic, and glass.

Table 1-5. Some materials commonly measured by ANDA.

Material	ANDA Technique
1. Lowly enriched uranium fuel rods (^{235}U)	^{252}Cf source with delayed γ -ray detection and/or delayed neutron detection and/or prompt neutron detection using energy discrimination.
2. Mixed-oxide fuel rods (Pu)	^{252}Cf source with delayed γ -ray detection.
3. HTGR fuel particles (^{235}U)	Am-Li source with coincident neutron detection and/or γ -ray detection. ^{252}Cf source with multiple coincident neutron detection and/or γ -ray detection.
4. HTGR fuel rods (^{235}U)	Same as 3, with a neutron moderation to tailor the irradiation neutron energy. ^{124}Sb -Be source with prompt neutron detection after energy discrimination. ^{252}Cf source with delayed neutron detection and γ -ray detection.
5. Small laboratory samples (Pu or ^{235}U)	^{252}Cf source with delayed γ -ray detection.
6. 55 gal. drums of waste (^{235}U and Pu), especially with fission product contamination	14-MeV neutron generator or ^{252}Cf source with neutron detection. Electron accelerator γ -ray source with prompt and delayed neutron detection.
7. High density uranium scrap (lowly enriched and highly enriched ^{235}U)	Am-Li or ^{252}Cf sources with coincident neutron detection and/or γ -ray detection.

Table 1-6. Materials commonly measured by passive NDA.

Material	Passive NDA Technique
Dense plutonium scrap	Calorimetry Passive neutron coincidence
55-gal. drums of low density waste (^{235}U and Pu), without fission product contamination	Passive γ -ray spectrometry and/or passive neutron or neutron- γ coincidence
Low-density plutonium scrap	Passive neutron time correlation and/or Passive γ -ray
Low-density uranium scrap	Passive γ -ray
Residue (holdup)	Passive γ -ray (sometimes passive neutron for plutonium)

response can be initiated to contain the stolen nuclear material and possibly the thief. Such an accounting system is called a real-time or dynamic material control system. Nondestructive assay measurements promise to play an important role in such real-time accounting systems because they can yield measurement results within minutes whereas destructive techniques require hours for completion. Proposed designs of real-time accounting systems are composed of the usual NDA for scrap and waste, new on-line NDA for holdup, and NDA for the main material flow and inventory normally measured by destructive methods. The two latter NDA measurements may be performed on bulk quantities of feed and product or on samples rapidly transmitted to a NDA sample assay station designed to process a large volume of samples in short periods of time. Because ANDA has the capability of reducing measurement time without diminishing precision by increasing the source strength, it is well suited for this application. Another feature of ANDA which makes it useful for real-time and delayed accounting of nuclear material is the inherent invulnerability of ANDA to tampering (Ref. 6) because of the basic complexity of the induced signatures.

1.7.3 Personnel and Package Search

Personnel who are not authorized to carry nuclear material and packages which are not supposed to contain such material routinely leave the areas where nuclear material is processed. Effective safeguards require a search of these personnel and packages to block potential diversion paths. Passive NDA is used for both personnel and package search; ANDA can be used only for package search because of the radiation danger to personnel. While personnel search instrumentation can presently detect lightly shielded 1g of plutonium and 3g of ^{235}U , package search is more difficult because of the often large amount of dense metal that can be present and the large size of some packages. ANDA, applied either as an assay technique or as a transmission check, is ideally suited for package search because the interrogation energy can be high enough to insure effective penetration.

One of the common applications of NDA that satisfies both accounting and package search is the NDA of large barrels or boxes of waste which usually contain small amounts of special nuclear materials (SNM).^{*} NDA systems are used both to assay for the small amount of SNM present and to assure that there are no nuclear materials concealed in the waste.

^{*}Special nuclear material (SNM) means Pu, ^{233}U , ^{235}U , uranium enriched with these isotopes, or any material artificially enriched with any of the foregoing.

1.7.4 Verification of Prior Assays

There are a number of situations when NDA verification of a prior assay can save time and money that would be required to redo the assay: (1) quick check of receipt quantities to assure no large shipper-to-receiver differences, (2) verification of the prior assay of inventory items if the tamper-indicating seal is broken, and (3) measurements by independent inspectors during inventory verification. In most of these applications the assay is semi-quantitative: the amount of detected radiation is usually compared with a previous radiation measurement in such a way that substitution of another element for the one expected in the sample is very difficult. In other cases, a quantitative NDA that may be less accurate than the original assay is used to verify that approximately all the material is present; passive γ -ray or neutron measurements normally suffice for these applications. Calorimetry can be applied in a unique way to check for shipper-receiver differences by determining the "watts" rather than the grams of nuclear material transferred. This method provides a very accurate and secure shipper-receiver comparison. Although ANDA could be used to verify prior assays, it is used less frequently for this purpose than other techniques. A disadvantage for independent inspectors' inventory verification is that ANDA instrumentation is not easily portable. This disadvantage is offset sometimes by measuring many different kinds of materials with ANDA.

1.7.5 Process and Quality Control

The products of nuclear processing operations undergo extensive measurement to assure that rigorous specifications are met. NDA is a useful tool in these processes and quality control. The most common application is the scan of fuel rods and plates to check homogeneity. Both passive and active NDA scans are used, but ANDA offers two advantages: higher throughput and better penetration. ANDA systems for rod scanning are relatively expensive; however, the higher cost is justified because of the importance to reactor performance of knowing rapidly and precisely the amount of nuclear material present and its distribution in the rods. The use of ANDA for this application is increasing.

1.8 OVERVIEW OF FUNDAMENTALS OF ANDA

The successful application of ANDA requires an analyst who understands the fundamentals of ANDA, including the physical principles underlying the technique, and the equipment and electronics used. The remaining chapters provide a description of these fundamentals in detail as required for a full understanding. An overview of these

fundamentals is included below for people other than ANDA analysts or designers and should be helpful to the manager or technician who works with the analyst and desires sufficient knowledge to communicate effectively. It should also be helpful to those who intend to read the whole book by introducing terminology and presenting a brief description of the entire subject before getting into details. Other brief reviews of the principles of ANDA may be found (Ref. 2, 7, and 8).

1.8.1 The Fission Process

Fission of nuclear material is the fundamental process of ANDA. When a fission takes place, a nucleus with atomic number Z and mass A normally separates into two smaller nuclei with atomic numbers Z_1 and Z_2 and masses A_1 and A_2 . The fission of a given isotope (for example, ^{235}U) can proceed in different ways, resulting in a distribution of fission product nuclei with differing masses and atomic numbers. Although the fission products carry approximately 85 percent of the fission energy, they are absorbed within the nuclear material and are thus not useful for ANDA. As shown in Table 1-7, the remaining 15 percent of the energy is radiated as prompt neutrons, prompt γ -rays, delayed γ -rays, delayed neutrons, and β -rays (and neutrinos) resulting from the radioactive decay of the fission products. Because the fission neutrons and γ -rays are highly energetic (in the MeV range) and electrically neutral, they are very penetrating and, thus, provide the useful ANDA signals.

As mentioned earlier, spontaneous fission of certain nuclides provides one of the passive signals observed in NDA. The probability of fission increases dramatically when a neutron or γ -ray interacts with a nucleus and gives it extra energy. Even when a thermal neutron, which has a very small amount of motion energy, is absorbed within a nucleus, a large

Table 1-7. Energy released in the fission of ^{235}U .

Form	Emitted energy (MeV)
Fission fragments	168
Fission product decay	
β -rays	8
γ -rays	7
neutrinos	12
Prompt γ -rays	7
Fission neutrons (kinetic energy)	5
Total	207

amount of energy is imparted to the nucleus. This energy represents the binding energy of the neutron to the nucleus. The mass of the system after the neutron is absorbed by the nucleus is smaller than the sum of the original masses of the neutron and nucleus. The mass loss is equivalent to the neutron binding energy. This energy excites the nucleus, and may be sufficient to cause it to fission.

As the neutron energy increases above thermal energies, the cross section for fission changes. Figure 1.3 shows the fission cross section of ^{235}U from thermal neutron energies up to energies of 15 MeV (a thermal neutron has an energy of 0.025 eV at 20°C). Obviously, the intensity of the ANDA response is proportional to the cross section. Thus, any matrix material or container, which can moderate the energy of the interrogating neutrons, will change the ANDA response and possibly lead to inaccurate assays, unless its effects are accounted for. One way to avoid these errors is to use thermal neutrons that cannot be further moderated. Another approach is to use higher energy interrogating neutrons and to place a thermal neutron detector close to the material to monitor the amount of neutron moderation, therefore, allowing the operator to make corrections.

Figure 1.3. Fission cross section of ^{235}U .

Table 1-1 shows that the thermal neutron fission cross sections for the different nuclides are not the same. The behavior of the cross section with respect to the energy of the interrogating high energy neutrons is not the same either. Figure 1.4 gives plots of the overall energy dependence of the neutron fission cross sections of uranium and plutonium isotopes. A general feature is the decrease in the cross section of the fissile nuclides— ^{235}U , ^{239}Pu , and ^{241}Pu —as the neutron energy increases. It is also clear that the fertile nuclides— ^{238}U , ^{238}Pu , and ^{240}Pu —have effective fission thresholds at energies greater than 0.5 MeV. Thus, if the energy of the interrogating radiation is less than 0.5 MeV, the ANDA response will be primarily a measure of the fissile

Figure 1.4. High energy neutron fission cross sections for uranium and plutonium nuclides measured by ANDA.

nuclides. One measure of an ANDA system is the fissile/fertile response ratio that reflects the assay sensitivity to fertile nuclides. If, the energy of the interrogating neutrons exceeds 1 MeV, the ANDA response measures both fissile and fertile nuclides.

1.8.2 Interactions of Neutrons and γ -Rays with Matter

The interactions of neutrons and γ -rays with matter are discussed in detail in Chapters 2 and 3. In this section we give a brief introduction to the subject.

A beam of neutrons and γ -rays passing through matter is attenuated by scattering and absorption processes. The intensity of the beam decreases according to a simple exponential law that relates the incident beam intensity, I_0 , to the beam intensity, I , which is transmitted through material of thickness, x :

$$I = I_0 e^{-\Sigma x} \quad (\text{neutrons}) \quad (1-1)$$

$$I = I_0 e^{-\mu x} \quad (\gamma\text{-rays}) \quad (1-2)$$

Both Σ , the neutron macroscopic cross section, and μ , the γ -ray attenuation coefficient, are measured in reciprocal length units. The reciprocals of Σ and μ are the mean free paths, or the average distances a neutron or γ -ray travels before interacting.

The quantity Σ relates to bulk matter. Earlier, we discussed cross sections measured in barns that referred to individual nuclei. Those cross sections, called microscopic cross sections (σ), are related to Σ by the quantity N , the number of nuclei/unit volume:

$$\Sigma = N\sigma. \quad (1-3)$$

γ -ray interactions, unlike neutron interactions, are not as dependent on the nucleus as on mass density. It is useful to consider the γ -ray attenuation coefficient, μ , as made of a mass density, (ρ), and a quantity called the mass absorption coefficient, (μ/ρ):

$$\mu = (\mu/\rho) \rho. \quad (1-4)$$

The total cross section or attenuation coefficient is determined by two types of attenuation processes: scattering and absorption. The absorption process removes the neutron or γ -ray; and scattering changes both the direction and energy of the radiation. Both of these processes are important in ANDA. Figure 1.5 shows the total mass absorption coefficient for γ -rays interacting with different materials. γ -Ray energy regions important in ANDA interrogation are also indicated. The penetrability of the interrogating radiation can be determined from these curves. For example, the mass absorption coefficient for interrogating γ -rays in uranium is approximately $0.05 \text{ cm}^2/\text{g}$, and the density of loosely packed uranium oxide is approximately 2 g/cm^3 . Therefore, the linear attenuation coefficient, which equals the average number of interactions/unit length, is $0.1/\text{cm}$. The mean free path, or the average distance between interactions, is 10 cm . By using the exponential attenuation law, we see that about 22 percent of a beam of these γ -rays penetrates to the center of a 15-cm radius cylindrical ("5 gal.") container of uranium oxide. In low-density material ($\ll 1 \text{ g/cm}^3$) with a mass absorption coefficient of $0.02 \text{ cm}^2/\text{g}$, the mean free path is 50 cm . About 40 percent of the beam penetrates to the center of a 55-gal. barrel of 45-cm radius.

γ -Rays emitted by fission have lower energies than interrogating γ -rays. The average energy/fission γ -ray is approximately 1 MeV , giving a mass absorption coefficient in uranium of $0.08 \text{ cm}^2/\text{g}$. Thus, although the energies are lower than those of interrogating γ -rays, the penetration in dense, heavy material is about half. In light materials, the penetration of fission γ -rays is considerably less than that of interrogating γ -rays, but penetration is still very good.

Thermal neutron scattering (σ_s) and absorption (σ_a) cross sections for some materials are shown in Table 1-8. In sharp contrast with the γ -ray absorption coefficients, which behave rather smoothly as the atomic number changes, the neutron cross sections can be drastically different

Figure 1.5. γ -Ray mass absorption coefficient for low and high atomic number (Z) materials.

for elements with similar atomic numbers. For example, the absorption cross section of boron ($Z=5$) is 755 b while that of beryllium ($Z=4$) is 0.1 b, a difference factor of 7550. The neutron cross sections in Table 1-8 can be used in a manner similar to the γ -ray mass absorption coefficients to calculate penetrability. The density of nuclei, N (nuclei/cm³), can be calculated from the following formula:

$$N [\text{nuclei/cm}^3] = \frac{\rho [\text{g/cm}^3] \times \text{Avogadro number} [\text{nuclei/gram-atom}]}{A [\text{g/gram-atom}]} \quad (1-5)$$

For example, for sintered uranium oxide, $\rho = 10 \text{ g/cm}^3$ and $A = 238 \text{ g/gram-atom}$. Avogadro's number = $6.02 \times 10^{23} \text{ nuclei/gram-atom}$ for all

Table 1-8. Scattering (σ_s) and absorption (σ_a) thermal neutron cross sections for some elements.

Element	Cross section (b)		Element	Cross section (b)	
	σ_s	σ_a		σ_s	σ_a
Aluminum	1.4	0.24	Hydrogen	38-100	0.33
Beryllium	7.0	0.10	Iron	11	2.62
Bismuth	9	0.034	Lead	11	0.17
Boron	4	755	Nitrogen	10	1.88
Cadmium	7	2450	Oxygen	4.2	0.0002
Carbon	4.8	0.0034	Silicon	1.7	0.13
Chlorine	16	32.6	Sodium	4	0.53
Deuterium	7	0.0005	Thorium	12.5	7
Fluorine	3.9	0.001	Tungsten	5	19.2
Helium	0.8	0.007	Uranium	8.3	7.68
			Zirconium	8	0.185

elements. Thus, $N = 2.5 \times 10^{22}$ nuclei/cm³, and, from Table 1-8, the total macroscopic cross section (scattering plus absorption) for uranium oxide* is calculated to be 0.4/cm. Thus, thermal neutrons have a mean free path of 2.5 cm in sintered uranium oxide, or 12.5 cm in loosely packed uranium oxide. Our previous calculations revealed that interrogating γ -rays have about the same penetrability in uranium oxide.

Not only does the neutron cross section depend heavily on the element, but as we pointed out earlier (Figure 1.4), it is highly energy-dependent. For uranium-238 (²³⁸U) the total neutron cross section increases from 16 b at thermal energy to 1000 b for neutrons with energy of 10-100 eV and back down to 12-6 b for neutrons with energies from 0.1 to 0.5 MeV. These latter energies are typical of high-energy neutron interrogating radiation.

This discussion of the attenuation of a beam of neutrons or γ -rays is useful in considering (1) penetration of interrogating and stimulated radiations into and out of nuclear materials, (2) design of shields for detectors and personnel, and (3) selection of detector materials. In all of these cases we are always primarily interested in the decrease in total beam intensity, whether by scattering or absorption. However, we cannot neglect the fact that scattered radiation still exists and may still be detected or cause fissions in the nuclear material, because the cross section for fission in fissile material and for detection in detector material increases significantly as the neutron energy decreases.

The ability of different elements to moderate neutron energy by scattering is measured by a parameter known as the slowing-down-power,

*To simplify, we neglect attenuation by oxygen.

a measure of the average decrease in neutron energy/unit path length in the material. The slowing-down-power is calculated by multiplying the macroscopic scattering cross section (number of scatterings/unit length) by the average logarithmic energy loss/scatter. Table 1-9 gives the slowing-down-power of some common neutron-moderating materials. Normal water has the best slowing-down-power of any of these materials. The moderating ratio shown in Table 1-9 is the slowing-down-power divided by the absorption cross section. It is a better measure of neutron-moderating ability of various elements because it reflects the fact that the neutron may be absorbed rather than scattered. Thus, heavy water is by far the best moderator because its absorption cross section is much lower than its scattering cross section.

Another common measure of a substance's ability to moderate neutrons is the average number of scattering collisions required to decrease the neutron energy from the fission neutron energy of 2 MeV to thermal energy. For example, thermalization of neutrons in uranium requires on the average 2171 collisions, but in water there are only 18.

The detailed analysis of neutron slowing-down and γ -ray attenuation is more complex than the simple fundamentals presented here. (See also Chapters 2, 3, and 6.) However, this discussion should help the reader understand the basis of selection of certain materials for neutron-moderating assemblies, neutron and γ -ray shields and detectors, the choice of an interrogating radiation source, and the types of interferences causing error in ANDA.

Table 1-9. Slowing-down properties of moderators.

Moderator	Slowing-Down-Power	Moderating Ratio
Water	1.28	58
Heavy water	0.18	21,000
Beryllium	0.16	130
Graphite	0.065	200

1.9 BRIEF DESCRIPTION OF ANDA SYSTEMS

The simple principles of ANDA, shown schematically in Figure 1.2 and discussed in Section 1.6, indicate that any ANDA system consists of the following five components:

1. *Interrogation source* with associated shielding, collimation, and electronic control.
2. *Neutron-moderating and filtering assembly* for tailoring neutron energies to suit a particular application.

3. *Sample chamber* containing the material being assayed and equipment for scanning and/or rotation.
4. *Detection assembly* including γ -ray shields, neutron moderators to enhance detection, and shields to prevent reflection of radiation into the sample chamber.
5. *Electronic circuits*: analog—for biasing the detectors and analyzing pulses to distinguish and count stimulated radiation; digital—to process the data and calculate the amount of SNM.

In this section, we will briefly describe a few ANDA systems in terms of the five components listed above and include an explanation of the underlying reasons for each ANDA design.

1.9.1 Delayed Neutron ANDA

Delayed neutrons can be used as the observed fission response, but a very intense interrogation source and a highly efficient detector are necessary because the delayed neutron yields are small—only about 0.006 neutrons/fission for ^{239}Pu and 0.016 neutrons/fission for ^{235}U . Delayed neutrons are emitted from approximately 0.1 s–1 min after fission. A very energetic neutron source, such as 14-MeV neutrons generated by deuterium-tritium reactions, can be used to give high intensity and penetrating radiation and thus, a uniform response. This method is especially suitable for measurements of small quantities of highly enriched fissile isotopes (for example, ^{235}U and ^{239}Pu). If considerable amounts of fertile isotopes are present, as with low-enrichment uranium fuel, substantial moderation of the source neutrons is required. This moderation, on one hand, increases the sensitivity of the measurement, but on the other hand, reduces neutron penetrability.

In delayed neutron ANDA, relatively simple detection equipment is required because no energy discrimination is necessary. Typical detection assemblies are BF_3 or ^3He gas-filled thermal neutron detectors surrounded by a good neutron-moderating material, such as polyethylene. The boron-10 (^{10}B) in the BF_3 gas absorbs thermal neutrons with a very high cross section (3700 b), producing α -particles that generate electrical charge by gas ionization. The thermal neutrons react with ^3He to produce tritium and ionizing protons. The cross section for this reaction is 5400 b. The neutron moderator around the detectors thermalizes the delayed neutrons for efficient detection in the gas-filled detectors.

Accelerator neutron sources are the most common interrogation sources used for delayed neutron ANDA. The 14-MeV neutron generator is useful when assaying large containers with material. With other types of accelerators, notably Van de Graaff proton accelerators and accelerator photoneutron sources (see Chapter 4), high neutron intensities,

which have good penetration and energies well below the fission thresholds of the fertile nuclides, are obtained. The accelerators can produce pulses as high as 10^9 to 10^{12} n/s and can be turned off quickly. They have a beam-on to beam-off ratio of 10^7 to 10^9 . Radioactive sources can be used, if the energy of the interrogating neutrons is reduced considerably. The penetration of low-energy neutrons is not as good, but the response is high because of the high fission cross section of thermal neutron. This technique is useful for small samples and uniform materials for which deep penetration of the interrogating radiation is not essential.

Another way to assay a sample is through photofission created by irradiating nuclear material with high-energy γ -rays and prompt and delayed neutrons are counted. In this method, a linear electron accelerator produces electrons with energies up to 10 MeV that impinge on a metal target, such as tantalum, and produce Bremsstrahlung γ -rays.* The technique can be used on a wide variety of materials, even on waste in 55-gal. drums, because these interrogating γ -rays are highly penetrating.

1.9.2 Delayed γ -Ray ANDA

Because delayed γ -rays from fission are 500 to 700 times as intense as delayed neutrons, delayed γ -ray ANDA is more sensitive than delayed neutron counting. The emission rate of delayed γ -rays declines more slowly than that of delayed neutrons, thus relaxing the time requirements on the on-off switching of the irradiation source. Highly efficient NaI(Tl) scintillation detectors are used to detect γ -rays. This technique is commonly used for scanning nuclear fuel rods that move rapidly through an interrogation chamber to a shielded counting chamber. The motion of the rod past the detector provides a scan that shows the uniformity of the distribution of nuclear material in the rod as well as its total content.

Neutrons from a radioactive source are usually the interrogating radiation. The fuel rod scanners mentioned above use ^{252}Cf fission sources and moderating assemblies to thermalize the neutrons. An alternative approach is to move rapidly the ^{252}Cf from the irradiation chamber to a shielded position. This technique, called source jerk or shuffling, can be used with or without neutron moderation, and can be applied to small, medium, and even to large samples like 55-gal. drums.

1.9.3 Prompt Neutron ANDA

One way to detect prompt neutrons from fission is to bias the detectors so that they respond only to fission neutrons and not to

*This type of radiation is described in Chapters 3 and 5.

interrogating radiation. This method usually requires low-energy neutron sources. Helium-4 (^4He) recoil detectors are most often used. They are sensitive only to neutrons above 0.5 MeV, reaching maximum efficiency for neutrons of approximately 1 MeV, the average energy of fission neutrons. The neutrons react with ^4He in an elastic scattering reaction that produces an electrical pulse in the gas-filled detector.

The interrogation source in these ANDA systems can be a ^{252}Cf source, moderated to produce thermal neutrons, or a photoneutron source, such as Sb-Be, that produces neutrons of energies less than 24 keV. The latter source provides adequate penetrability through dense material, and the energy is well below the detection threshold of ^4He and other fast neutron detectors. Two disadvantages of this source are the short half-life of the antimony radioisotope (60 days) and the intense γ -ray background.

1.9.4 Prompt Fission Detection ANDA

Prompt fission radiation can be detected and distinguished from interrogating radiation by coincidence techniques, which are called fission multiplicity detection (FMD) techniques, because they require detection of multiple fission radiations emitted simultaneously. The detectors are usually plastic scintillators sensitive to both neutrons and γ -rays, of which there are approximately eight per fission. Typically, two to four such detectors are used, and coincidences such as 2-out-of-2, 2-out-of-4, or 3-out-of-4 are measured as indications of a fission event. If γ -rays are well shielded and only neutrons are detected, two detectors are adequate. However, if γ -rays are detected, three or four detectors are desirable. The coincidence gate time is very short, of the order of 20-100 ns ($1 \text{ ns} = 10^{-9} \text{ s}$), giving excellent discrimination between true fissions and random pulses. By performing two measurements, one with and one without the interrogation source, the operator can correct for the contribution of spontaneous fissions.

These ANDA systems use a variety of radioactive neutron sources, ^{252}Cf and Am-Li are the most common. The ^{252}Cf source, shielded from the detectors, produces a beam of neutrons that irradiate the sample, which is rotated and scanned to yield uniform response. Single or multiple Am-Li sources are usually placed inside the counting chamber so as to give uniform irradiation of the material. This proximity is possible because the neutron energies from lithium targets are low. The detectors are sometimes shielded with lead to attenuate low-energy γ -rays and with boron to attenuate low-energy neutrons. The induced radiation is then penetrating, resulting in accurate assays of low sensitivity.

The remaining chapters of this book present the fundamentals of ANDA. Chapters 2 and 3 are discussions of the physics of the interaction of neutrons and γ -rays with matter that is basic to the first four components of an ANDA system. In Chapters 4 through 9, each of the five components of ANDA is discussed. Chapters 4 and 5 describe the principles of neutron and γ -ray sources and their characteristics. Chapter 6 treats the transport of neutron and γ -rays through bulk matter, such as the neutron tailoring assembly, the shielding material, the material being assayed, and the detectors themselves. The fundamentals of the physics of neutron and γ -ray interactions with individual atoms and nuclei, described in Chapters 2 and 3, are applied in Chapter 6 to show the gross reaction of neutrons and γ -rays with bulk media. Chapter 7 describes the special characteristics ("signatures") of fission radiations emitted by the materials being assayed. Chapter 8 covers the detectors of the fission radiations. It also includes discussions of the electronics used to process detector pulses for energy discrimination and high-rate pulse counting along with a detailed analysis of coincidence measurement systems. Chapter 9 shows many examples of existing ANDA techniques, which are categorized firstly according to whether delayed or prompt radiation is detected and secondly according to the type of delayed radiation detected and the technique used to discriminate prompt fission radiation from interrogating radiation. Chapter 10 concludes this book with a discussion of instrument analyses and calibration and measurement control programs that are essential for the effective use of ANDA systems.

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CHAPTER 2

INTERACTIONS OF NEUTRONS WITH MATTER

2.1 INTRODUCTION

An understanding of the interaction of neutrons with matter is basic to the analysis of any ANDA measurements based on neutron-induced reactions and detection methods. These interactions occur in each of the generic areas of the basic ANDA system described in Section 1.9: the neutron source, the source spectrum-tailoring assembly, the sample, and the detector. In each of these areas, various types of neutron interactions determine the intensity and energy spectrum of the neutrons. These interactions comprise neutron scattering, neutron absorption, and neutron multiplicative processes, including fission.

In some sources, interactions of neutrons within the source material may slightly change the energy spectrum of the emitted neutrons. The tailoring assembly, designed to shape the neutron energy spectrum to meet the assay needs, is based on an understanding of the interactions between fast neutrons and nuclei of both light and heavy elements. In general, samples consist of a mixture of light and heavy elements. The heavy elements include, usually, both fissile and fertile fissionable isotopes. Thus, in addition to being scattered and absorbed in the sample, neutrons induce fission, which provides the main signature* sought after in ANDA, and also produce more neutrons as a result. Reactions $(n,2n)$ and similar neutron-multiplying interactions may also take place. The neutrons produced by all of these reactions will interact with the sample and either lose energy in scattering or be further multiplied by fission or $(n,2n)$ reactions. Finally, the neutrons must be detected by a selected detector. The proper choice and design of the

*The "signature" of a material comprises the characteristic types, intensities, and energies of the radiations emitted by that material in an assay.

detector (for example, use of a moderator) also require an understanding of neutron interactions in the various components of the detection system.

The neutron interactions which can occur in matter are described in Section 2.2. The notions of interaction cross section, reaction rate and their properties are introduced in Section 2.3. The remaining sections cover, in more detail, each type of interaction and the behavior of its cross section. The last section is a summary of this chapter.

2.2 NEUTRON INTERACTIONS

The various neutron interactions with matter can be conveniently classified as follows (Ref. 1):

1. Scattering
 - a. elastic scattering, (n,n)
 - b. inelastic scattering, (n,n') .
2. Absorption
 - a. radiative capture, (n,γ)
 - b. charged particle reactions, (n,p) , (n,d) , (n,α) , and others.
3. Neutron multiplicative processes*
 - a. fission, (n,f)
 - b. $(n,2n)$.

These interactions are shown schematically in Figure 2.1.

In *elastic scattering*, all of the initial kinetic energy of the incident neutron plus that of the scattering nucleus reappears as the kinetic energy of the two bodies after collision. None of the neutron energy is transformed into the internal energy of the nucleus; the collision can be conceptualized as a classical billiard ball collision. The neutron energy following collision depends on its energy before the collision, the angle through which it is scattered, and the mass of the scattering nucleus. Elastic scattering of neutrons on light nuclei enhances the efficiency of neutron moderation (slowing-down) because the lower the mass of the nuclei, the greater the average loss of neutron energy per collision (see Section 2.4). This fact is important in ANDA since it increases the probability of inducing fission when fissile material is present.

In *inelastic scattering*, some of the incident neutron kinetic energy is transformed during the collision to internal excitation energy of the scattering nuclei. This process occurs only if the neutron energy is large enough to excite the internal energy levels characteristic of the scattering

*These processes are usually included in the absorptive interactions. Here, they are treated separately because of their special role in ANDA.

Figure 2.1. Schematic representations of neutron interactions.

nucleus. In general, inelastic scattering leads to greater neutron energy loss than elastic scattering. It is usually important in heavy and intermediate weight nuclei. As a result of the large energy loss, the neutron energy may be quickly lowered to a level where elastic scattering with light elements, particularly hydrogen, becomes more likely. Thus, (n,n') reactions with heavy nuclei mixed with hydrogen-containing substances can significantly enhance the moderation process of high-energy neutrons.

Absorption reactions result in the loss of the neutron. In most cases, absorption of slow neutrons occurs by the radiative capture process, during which one or more γ -rays are emitted. In some absorptions, a charged particle is emitted [for example, in the (n,p) or (n,α) reactions]. Occasionally, as a result of later disintegration of the product nucleus, one neutron may be eventually emitted. For example, in the $^{17}\text{O}(n,p)$

^{17}N reaction, ^{17}N subsequently decays by delayed neutron emission. The half-life of this process is about 4 s and results in delayed neutron background when 14-MeV neutrons are used in ANDA.

If neutrons are absorbed, through any of the processes mentioned (by materials in the source itself, by spectrum-tailoring assembly or by the sample), the process is known as *self-absorption*. The intensity and energy spectrum of the emerging neutrons depend strongly on this self-absorption.

The *fission* process induced by neutrons (or photons) is the most important reaction in ANDA. It provides the main, if not the sole, signature or combination of signatures which permit ANDA to attain its principal objective—the detection and measurement of fissile materials. As a result of each fission, several neutrons (usually 2 or 3) are released; fission is thus a multiplicative reaction. Another multiplicative process is the $(n,2n)$ reaction, in which a high energy neutron is absorbed by a nucleus and, as a result, two lower energy neutrons are released. Its principal importance in ANDA is to increase source intensity by multiplication and reduce the high-energy of the source neutrons.

2.3 CROSS SECTION AND REACTION RATES

The concept of microscopic and macroscopic cross sections and reaction rates are discussed in this section.

2.3.1 Microscopic Cross Sections

The probability of the occurrence of a neutron-nuclear reaction is related to the concept of a “nuclear cross section.” The cross section can be defined experimentally as follows. Consider a thin target with an incident beam of monoenergetic, monodirectional neutrons, normal to the target. If the rate at which neutron-nuclear reactions occur in the sample is measured, it is found to be proportional to all of the following:

1. The number of neutrons incident upon the target per unit time and per unit area, $I(\text{cm}^{-2} \text{ s}^{-1})$.
2. The cross sectional area of the target, $S(\text{cm}^2)$.
3. The target thickness, $\Delta x(\text{cm})$.
4. The atomic density of the target, $N(\text{cm}^{-3})$.

If σ is the constant of proportionality, then the rate at which reactions occur (the interaction rate) can be expressed as

$$\text{reaction rate} = \sigma INS \Delta x. \quad (2-1)$$

Since the rate has the dimensions of s^{-1} , σ must have the dimensions of an area. An equivalent definition of σ is

$$\sigma \equiv \frac{\text{number of events of a given kind/nucleus/s}}{\text{number of incident neutrons/cm}^2/\text{s}}. \quad (2-2)$$

σS is then the probability that a neutron in the beam will interact with a target nucleus and σ is known as the *microscopic cross section*.

The commonly used unit for σ is the barn (b) (which equals 10^{-24}cm^2 , the approximate geometric cross section of the nucleus). However, σ can be much larger or smaller than the geometric cross section, ranging from 0 to as much as 10^6 b. Neutron-nuclear reaction cross sections relevant to ANDA are usually between 10^{-27} and 10^{-22}cm^2 (millibarns to hundreds of barns).

For each type of neutron-nuclear reaction and each type of nuclide, a different cross section can be defined, depending on the probability of *that particular* reaction taking place. Common types of neutron cross sections include:

$$\begin{aligned} \sigma_s &= \text{elastic scattering} \\ \sigma_i &= \text{inelastic scattering} \\ \sigma_\gamma \text{ or } \sigma_c &= \text{radiative capture} \\ \sigma_f &= \text{fission} \\ \sigma_t &= \text{total} = \sigma_s + \sigma_i + \sigma_\gamma + \sigma_f + \sigma \text{ of other interactions} \\ \sigma_a &= \text{absorption} = \sigma_\gamma + \sigma_f + \sigma(n,p) + \sigma \text{ of other absorption} \\ &\quad \text{interactions} \\ \sigma_{ne} &= \text{nonelastic} = \sigma_t - \sigma_s \\ &\sigma(n,2n) \text{ and other cross sections.} \end{aligned}$$

The total cross section, σ_t , is the cross section for any type of interaction to occur. Because cross sections are essentially probabilities of independent events, they are additive; that is, the total rate of two reactions, A and B , is determined by the sum ($\sigma_A + \sigma_B$).

2.3.2 Macroscopic Cross Sections

In the preceding section, we assumed a beam of neutrons incident upon a very thin target, a condition that assured exposure of each nucleus to the same beam intensity. If the target were thicker, nuclei within the sample would be shielded from the beam by the nuclei on the surface because interactions remove neutrons from the incident beam. To account for thickness effects, we must consider a neutron beam incident upon the surface of a target of an arbitrary thickness.

To derive an equation for the “uncollided” beam intensity, $I(x)$, at any point x in the target, consider a differential thickness of the target

between x and $(x + dx)$. Since dx is very small, the interaction rate in the differential thickness dx can be calculated from Eq. 2-1 as follows:

$$\text{interaction rate in } dx = \sigma IN S dx. \quad (2-3)$$

Since the interaction rate/unit area is equal to the decrease in beam intensity, $-dI$, we obtain

$$-dI = \sigma IN dx. \quad (2-4)$$

The solution of this equation, given an incident beam intensity of I_0 at $x=0$, is

$$I(x) = I_0 e^{-N\sigma x}. \quad (2-5)$$

Eq. 2-5, therefore, defines the "uncollided" beam intensity at any point x in the target.

It is convenient to define the product of the atomic density, N , and the microscopic cross section, σ , as a separate quantity, the *macroscopic cross section*, Σ , which had the dimensions of inverse length.

Since

$$\Sigma = -\frac{1}{I} \frac{dI}{dx} \quad (2-6)$$

Σ can be interpreted as the probability/unit path length that the neutron will react with a nucleus in a small thickness of sample between x and $x + dx$. Furthermore,

$$e^{-\Sigma x} = \text{probability that a neutron moves a distance } x \text{ without interaction} \quad (2-7)$$

so that

$$\begin{aligned} \Sigma e^{-\Sigma x} dx &= \text{probability that the neutron has its first interaction in } dx \\ &\equiv p(x) dx. \end{aligned} \quad (2-8)$$

Therefore, the mean distance a neutron travels before interacting with a nucleus in the sample is (Ref. 2)

$$\bar{x} \equiv \int_0^{\infty} x p(x) dx = \Sigma \int_0^{\infty} x e^{-\Sigma x} dx = 1/\Sigma. \quad (2-9)$$

This distance, known as the *mean free path*, is generally denoted by λ .

Since Σ^{-1} is the mean distance that the neutron travels between collisions, the frequency of collision is given by $v\Sigma$, where v is the neutron speed, and $1/v\Sigma$ is the mean time between reactions.

The macroscopic cross section for a specific reaction is given by N times the microscopic cross section for that reaction, for example, for reactions f , a , t , etc.,

$$\Sigma_f = N\sigma_f, \Sigma_a = N\sigma_a, \Sigma_t = N\sigma_t, \text{ and others.}$$

The macroscopic cross section, for a reaction of type j , of a homogeneous mixture of materials X , Y , and Z (which may, for example, constitute the matrix in the interrogated sample), with atomic densities, N_X , N_Y , and N_Z , respectively, is given by

$$\Sigma_j = N_X\sigma_j^x + N_Y\sigma_j^y + N_Z\sigma_j^z \quad (2-10)$$

where σ_j^x , σ_j^y , and σ_j^z are the microscopic cross sections for nuclides x , y , and z , respectively.

The cross section for any neutron reaction varies from isotope to isotope and depends on the neutron energy. The magnitude and energy dependence of the cross sections are important factors in designing ANDA measurements because they determine the rate of the desirable signal and the undesirable background interactions. Values for cross sections are tabulated in a series of publications (Ref. 3,4) known as the "barn books." Tables and graphs for some important materials for ANDA are presented below (see Section 1.8.1).

Inspection of cross section data reveals the following general features of cross section behavior:

1. In light nuclei ($Z \leq 30$), elastic scattering is usually the dominant interaction process at all neutron energies up to ~ 20 MeV. At neutron energies above ~ 1 MeV, inelastic scattering can also be important.
2. In heavier nuclei ($Z > 30$, except for a few "magic" nuclei), absorption, especially radiative capture, becomes dominant at lower neutron energies (below 10 keV) and inelastic scattering is important at higher energies (above 100 keV).
3. In the energy region between ~ 1 eV and ~ 10 keV, cross sections frequently exhibit a resonance structure (Figure 2.2). In the resonances, the cross section can become very large over a narrow energy interval. Absorption resonances are particularly strong in the intermediate and heavy nuclei. In the light nuclei, the resonances are mainly scattering resonances.
4. Below a few eV, absorption cross sections frequently show an energy variation inversely proportional to the neutron speed: $\sigma_a \sim 1/v$ or $E^{-1/2}$. Scattering cross sections at these low energies are constant and usually amount to a few barns.

Figure 2.2. Total cross section of ^{238}U (Ref. 3).

5. For hydrogen, the absorption cross section is small at all energies (although the absorption can be important). The scattering cross section is large (amounting to 90 b at very low neutron energies) and decreases to 20 b at 1 eV. It is constant at this value up to about 10 keV and then slowly decreases at higher energies to values of about 4 b at 1 MeV and 0.9 b at 10 MeV. The scattering is always elastic, and there are no resonances.

Most of these features can be explained in terms of the formation of a “compound nucleus” (Ref. 1). The interaction may be conceived in two steps: the neutron is first absorbed, resulting in the compound nucleus of mass ($A + 1$); that nucleus then decays either by re-emitting a neutron (elastic or inelastic scattering), by emitting a photon or charged particle (absorption), or by fission. In elastic scattering of this type, the neutron is re-emitted with its original kinetic energy (in the center-of-mass system).

Whereas compound elastic scattering is only significant in the neighborhood of resonances, “potential scattering” is a type of elastic scattering which occurs with neutrons of any energy. It does not involve formation of a compound nucleus but arises simply because of the presence of the nucleus. Another neutron interaction, not involving a compound nucleus process, is called direct interaction which is important at higher neutron energies, well above the average fission neutron energies (~ 2 MeV).

2.4 ELASTIC SCATTERING

With few exceptions, elastic scattering is the most important interaction between neutrons and light nuclei. In hydrogen, for example, the elastic scattering cross section is equal to the total cross section from 10 keV to 40 MeV.

Elastic scattering in a neutron-nuclear reaction results in a discrete loss of neutron energy, varying from zero to the maximum allowable by the laws of conservation of momentum and energy. The loss depends on the

scattering angle and the angle between the neutron's initial and final directions. The largest possible loss occurs when the neutron is scattered backward relative to its original direction (scattering angle = 180°). Solving the equations for conservation of momentum and kinetic energy for this case (Ref. 1) gives the neutron energy after collision, E_1 , in terms of its energy before collision, E_0 , and the atomic mass number of the target, A :

$$E_1 = E_0 \left(\frac{A-1}{A+1} \right)^2. \quad (2-11)$$

This equation shows that in a collision of a neutron with a carbon nucleus ($A = 12$), for instance, the largest possible energy loss is 28 percent, while in elastic scattering with a ^{238}U nucleus, the maximum loss is about 1.7 percent. A collision with a hydrogen nucleus ($A = 1$), however, results in the loss of all the neutron energy.

In addition to the energy loss, the angular distribution of the scattered neutrons is important. To describe this distribution, it is convenient to refer to a coordinate system whose origin is located on the center-of-mass of the neutron-nucleus system. Generally, the neutron scattering on light nuclei in the center of mass system is isotropic for energies below about 0.5 MeV. Neutrons with higher energy (for example, 14 MeV) show forward peaked angular distribution; that is, the neutrons are most frequently scattered through a small angle. In heavy nuclides, such as uranium and lead, anisotropy of the angular distribution prevails well below 1 MeV; for example, at 0.25 MeV (Ref. 3). Neutron scattering by hydrogen is isotropic in the center of mass system for all neutron energies of interest in ANDA. The angular distribution when viewed in the laboratory system is quite different for scattering on a light nuclide target. The average cosine of the scattering angle *in the laboratory system*, $\bar{\mu}$, is a function of the atomic number, A , of the target nucleus

$$\bar{\mu} = \overline{\cos \theta} = 2/3A. \quad (2-12)$$

Hence, for hydrogen ($A = 1$), $\bar{\mu} = 2/3$, and the scattering is mostly in the forward direction. At the other extreme, for scattering by uranium ($A = 238$), $\bar{\mu} \approx 0$. This result should be obvious because for a heavy scatterer, the center-of-mass and the laboratory systems are almost the same (the center-of-mass being essentially fixed at the scattering nucleus).

The effect of the scattering angular distribution on the motion of a neutron, in a scattering medium, may be taken into account very roughly by defining a "transport cross section," σ_{tr} . In a nonabsorbing medium, this cross section is (Ref. 2)

$$\sigma_{tr} = \sigma_s(1 - \bar{\mu}). \quad (2-13)$$

If absorption is also present

$$\sigma_{tr} = \sigma_t - \sigma_s \bar{\mu} = \sigma_a + \sigma_s (1 - \bar{\mu}). \quad (2-14)$$

The transport mean free path is then given by

$$\lambda_{tr} = \frac{1}{1/\lambda_t - \bar{\mu}/\lambda_s}. \quad (2-15)$$

In the case of spherically symmetric scattering, $\bar{\mu} = 0$, Eqs. 2-13 and 2-14 reduce to σ_s and σ_t , respectively. The transport mean free path can be thought of as the average distance that a neutron travels after a collision before it "forgets" its original direction of motion as a result of subsequent collisions.

Because the energy loss depends on the scattering angle, an average energy loss per collision can be obtained by averaging over the angular distribution. It is usually more convenient to use the logarithm of the average energy loss, ξ . It can be shown (Ref. 5) that for isotopic scattering in the center-of-mass system, the average energy loss equals the following:

$$\xi = \langle \ln \frac{E_0}{E_1} \rangle = 1 + \frac{(A-1)^2}{2A} \ln \frac{A-1}{A+1} \quad (2-16)$$

where E_0 is the neutron energy before a collision and E_1 the energy after collision. For values of $A > 2$, this expression may be approximated by

$$\xi \cong \frac{2}{A+2/3} \quad (2-17)$$

within 3.3 percent of the correct value for $A = 2$. For larger values of A this approximation is excellent.

The quantity ξ may be used to calculate the average number of collisions required to slow down a fast neutron from an energy E_0 to E_1 . The average number of collisions required is $[\ln(E_0/E_1)]/\xi$. For example, suppose the initial neutron energy is 2 MeV, and the neutron is slowed down to 0.025 eV by elastic collisions with hydrogen ($\xi=1$). The average number of collisions for this case is 18. The number of collisions required to thermalize a neutron for several elements is summarized in Table 2-1 for $E_0 = 2$ MeV and $E_1 = 0.025$ eV.

However, heavy elements can slow down fast neutrons by means of inelastic scattering. This subject is discussed in more detail in the following section.

2.5 INELASTIC SCATTERING

When a compound nucleus (see Section 2.3.2) is raised to an energy state equal to, or in excess of, the first excited state above ground level

Table 2-1. Some scattering and slowing down properties of nuclei.

Element	Mass Number	ξ	Number of Collisions to Thermalize
Hydrogen	1	1.000	18
Deuterium	2	0.725	25
Beryllium	9	0.209	86
Oxygen	16	0.120	150
Uranium	238	0.00838	2172

of the target nucleus, it may decay to that ground state (or to other level if energetically possible) by emitting one or more γ -rays. A neutron with less energy than the incident neutron is also emitted. The energy of the first level in heavy nuclides is the threshold energy for inelastic scattering. The relative spacing of the energy levels determines the probability distributions of the number and energy of the emitted γ -rays. Where level spacings are wide, as in light nuclei and the stable "magic" nuclei, the threshold is high and decay is more likely to occur by emission of a small number of high-energy γ -rays. When the spacings get closer together, as in heavier isotopes or at high excitation energies, the γ -ray emission spectrum forms a near continuum.

The energy distribution of the inelastically scattered neutrons is determined by the energy levels of the target nucleus. When the energy of the incident neutron is only high enough to excite a few levels in the nucleus, the inelastically scattered neutrons appear in distinct energy groups. Each group corresponds to the excitation of a single level. If the energy of the incident neutron is high enough to excite many levels, the neutron groups corresponding to the more closely spaced levels form a continuous spectrum that is observed in addition to the discrete spectrum.

The inelastic cross sections for lead, representing heavy elements, and for iron, representing intermediate elements, both of which are often used in spectrum-tailoring assemblies, are shown in Figure 2.3. The ability of these elements to slow down fast neutrons by the inelastic process is indicated by the cross sections for emission of specific γ -rays. The energy of these γ -rays is equal to the energy loss of the interacting neutrons. Figure 2.4A shows several γ -emission cross sections for the ^{206}Pb isotope, which constitutes 23.6 percent of the naturally occurring elemental lead. Figure 2.4B shows similar cross sections for the ^{56}Fe isotope, which constitutes 91.7 percent of the naturally occurring iron. These and other curves indicate, for instance, that a 3-MeV neutron has a reasonably high probability of losing either 0.80, 0.54, 0.66, 1.70, 1.36, 1.82, or 1.45 MeV (in decreasing order of probability) in one inelastic

Figure 2.3. Inelastic cross sections of Fe and Pb.

collision with a Pb nucleus. This shows the effectiveness of inelastic scattering in reducing the energy of fast neutrons

The “group transfer cross section” is required in some neutron transport calculations of interest in ANDA applications. The group transfer cross section, $\sigma(E_i \rightarrow E_j)$, is the sum of the cross sections for all processes that result in a neutron whose initial energy is E_i (lying in energy region or “group” i) and is scattered to an energy E_j (in energy region or “group” j). If group j is the same as or is just below group i , both elastic and inelastic scatterings can result in the neutron energy loss $E_i \rightarrow E_j$, and the group transfer cross section is the sum of the cross sections of the two processes. If group j is sufficiently far below group i , elastic scattering cannot cause this transfer (in one collision), because of Eq. 2-11. Here only inelastic scattering is important. For hydrogen, of course, there is no inelastic scattering because all its energy loss is by elastic scattering.

The importance of inelastic scattering in ANDA becomes clear when we remember that in inelastic scattering, a neutron may lose a larger amount of energy in a collision with a heavy nucleus than would be possible through an elastic collision. For example, a 1-MeV neutron loses a maximum of 17 keV in an elastic scattering with ^{238}U as compared to

Figure 2.4A. Cross sections for γ -ray emission in inelastic scattering in ^{206}Pb (Ref. 3).

Figure 2.4B. Cross sections for γ -ray emission in inelastic scattering in ^{56}Fe (Ref. 3).

44 and 146 keV (with equal probability) in inelastic scattering. The slowing-down effectiveness of inelastic scattering in heavy nuclides increases greatly for higher energy neutrons. For example, a 14-MeV neutron will lose on the average about 12 MeV in an inelastic collision with uranium or lead nuclides. Since inelastic scattering cross sections are the largest for intermediate and heavy elements, materials like Fe, Pb, and W are commonly used as a first stage in the moderation of fast neutrons as well as a shield against γ -rays emitted by the neutron source.

The angular distributions of inelastically scattered neutrons is rather simple: up to incident neutron energies of around 10 MeV, the neutron is emitted isotropically in the center-of-mass system. Above 10 MeV, the distribution tends to become peaked forward. For most practical applications, however, it is usually adequate to assume that the inelastic scattering is isotropic in the center-of-mass system. For heavy nuclei, this means that the angular distribution is also isotropic in the laboratory system.

Inelastic cross sections as functions of the incident neutron energy are shown in Figure 2.5 for aluminum, titanium, zirconium, cadmium, gold, uranium, iron, and lead. The cross sections vanish, in each case, below the threshold energy.

Figure 2.5. Inelastic cross sections (σ_{in}) versus incident neutron energy of several elements.

2.6 ABSORPTION

For high-energy neutrons, a number of different absorption processes exist, all of which proceed by compound nucleus formation. The compound nucleus emits one or more particles or γ -rays immediately

following its formation. For fast neutron absorption, the emitted "particles" are usually either a proton, an α -particle, two neutrons, or one or more γ -rays. These reactions can be written in the following ways (the asterisk indicates a nucleus in an excited state):

The new nuclei resulting from these processes, e.g., ${}_{Z-2}^{A-3}Y$ or ${}_{Z-1}^AT$, will go to the ground state from an excited state, usually by rapid emission of one or more γ -rays. For the (n,γ) reactions, these γ -rays are known as (prompt) capture γ -rays, whose energies are in the range 1-10 MeV. They provide unique signatures for most elements. A notable exception are the fissionable isotopes, whose capture γ -ray signatures are not very strong and are usually lost in the background (Ref. 6).

Although fast neutrons may undergo a number of different absorption processes, the probability that these processes will occur is usually very small because cross sections of scattering processes are much larger at high energies than absorption cross sections and because the thresholds of most absorption reactions are usually in the range from 1 to 10 MeV or higher. Only a fraction of the neutrons will cause absorption reactions because they are much more likely to be slowed down below the threshold energies by scattering processes than to undergo fast neutron capture.

Whereas absorption reactions where charged particles are emitted are threshold reactions, the (n,γ) capture reactions are not threshold reactions. Their cross sections are small at high energies but large at lower neutron energies, for example, at thermal neutron energies. It has been mentioned that at thermal energies, the cross section for (n,γ) capture, σ_γ , varies as $1/v$, where v is the neutron speed. The magnitude of σ_γ , at low energy, depends upon whether or not there is a resonance in this energy region. At energies above the resonance region, σ_γ drops rapidly as $1/E$ or faster, to very small values, as a function of energy. The reason for the rapid decrease in σ_γ is that, with the increasing energy of the incident neutron, the compound nucleus is formed in more highly excited states that can decay more readily by neutron emission (or

fission, if this is energetically possible) than by γ -ray emission. At energies above 1 MeV, inelastic scattering, thus, becomes the dominant compound nucleus decay process, at the expense of both elastic scattering and radiative capture. At still higher energies, the $(n,2n)$ reaction can also become important.

Direct application of capture γ -signatures in ANDA measurement is very limited. However, since the low-energy cross section for this process is relatively high for high- Z (Z = atomic number) materials, its effects on the sensitivity and accuracy of ANDA measurements should be considered. γ -Radiation not only increases the background for the neutron detection system, but it can also be detected simultaneously by more than one detector as a result of Compton scattering events (Chapter 3), therefore, increasing the error due to random coincidences in fast coincidence measurement techniques. (See, for example, Chapter 8.) These detection problems are further complicated by annihilation radiation (see Section 3.4) and the presence of materials which decay by γ -cascade, that is, when two or more γ -rays are emitted successively from different excited levels. Radiative capture by hydrogen during the moderation and detection phases of the ANDA measurement technique is also important because it produces a large amount of 2.2-MeV γ -rays, which may be an undesirable background.

2.7 NEUTRON MULTIPLICATIVE PROCESS—FISSION AND $(n,2n)$ REACTIONS

The two main neutron multiplicative processes are the neutron fission (n,f) and the $(n,2n)$ reactions to be discussed in this section.

2.7.1 Neutron Fission

Nuclear fission by neutron (n,f) is by far the most important multiplicative process in ANDA measurements. It provides the main stimulated signature that distinguishes the fissionable materials from the surrounding matrix material in the interrogated sample. Fission may be induced by high-energy or thermal-energy neutrons.* The fissile nuclei, such as ^{233}U , ^{235}U , ^{239}Pu , and ^{241}Pu , undergo fission following the absorption of a thermal-energy neutron. With most other heavy nuclei, the binding energy of the incident neutron is not itself sufficient to supply the compound nucleus with the required critical energy, and the

*Many of the fissionable isotopes also undergo spontaneous fission that is the basis of some PNDA techniques (Ref. 7). Spontaneous fission may constitute one of the background components in ANDA techniques, where spontaneous fission rates are very low as compared to induced fission rates.

neutron must have kinetic energy above a threshold level (usually between 0.5 and 1.5 MeV) to induce fission. Neutrons with energies below this threshold value are labeled *subthreshold* or *sub-MeV neutrons*. A threshold level is always present when the nucleus contains an even number of nucleons (for example, ^{232}Th and ^{238}U), because the binding energy of the incident neutron to an even- A nucleus (for example, ^{238}U) is always less than to an odd- A nucleus (for example, ^{235}U). Nuclei with threshold energies below 10 MeV are called *fertile*.^{*} Although ^{208}Pb may fission when struck by a neutron of energy greater than 20 MeV, it is not considered fertile. On the other hand, ^{238}U can fission if the neutron energy is greater than 1 MeV, hence it is considered fertile. Other fertile isotopes of interest are ^{232}Th , ^{238}Pu , ^{240}Pu , and ^{242}Pu .

Observation shows that any phenomenon that excites a nucleus above the critical energy level can precipitate fission. Besides excitation by thermal- or high-energy neutrons, high-energy γ -rays excite a nucleus and cause a type of fission called photofission. This type of reaction is the basis for ANDA systems employing electron accelerators such as linacs (see Section 3.6).

Following neutron absorption in the fission process, the excited nucleus splits mostly into two large fragments, and emits almost instantaneously, on the average, two or three prompt neutrons and numerous γ -rays. The sequence of events in a fission process is shown schematically in Figure 2.6. It gives the time scale of a fission event and the four types of radiations used as signatures in ANDA techniques: prompt neutrons, prompt γ -rays, delayed neutrons, and delayed γ -rays.

The fission cross section, σ_f , is a function of the target nucleus and the energy of the incident neutron. For the fissile nuclei, σ_f is quite large at low energy and drops roughly, as $1/v$, with increasing neutron velocity (v) or energy up to the eV region where resonances appear. Above approximately 1 keV, resonances can no longer be resolved by present measuring techniques, and the cross section becomes rather smooth, as shown before in Figure 1.3 where σ_f is shown for ^{235}U . Although most of the ANDA techniques discussed in the subsequent chapters are based on fission induced by thermal, epithermal (0.5 to 10 eV), and sub-MeV neutrons, the resonance region in σ_f can possibly be used in ANDA (Ref. 8).

The fission cross section is somewhat different for fertile nuclei, such as ^{238}U , that require an energetic neutron to induce fission. The σ_f is

^{*}The name comes from the fact that the fertile isotopes can capture a neutron and, by radioactive decay, convert into a fissile isotope. This process is called breeding.

Step

1. Initial Condition.

2. 10^{-15} sec after irradiation. The nucleus splits into the two independent fission fragments.

2 Fission Fragments

3. 10^{-20} sec after Step 2.

2 to 3 Prompt Neutrons Per Fission

4. 10^{-17} sec following Step 2.

~8 Prompt Gammas Per Fission

5. 0.1 to 100 sec following Step 2.

~ 10^{-2} Delayed Neutrons and
~8 Delayed Gamma Rays Per Fission

Figure 2.6. Schematic representation of the fission process.

practically zero up to the threshold energy for fission, where it rises rapidly with increasing energy. Since the fission threshold occurs, usually, at energies above the region of resolvable resonances, the fission cross sections of these nuclei tend to be smooth everywhere. The relatively high threshold energy for fission of ^{238}U nuclei provides an efficient way to analyze the content of fissile materials in the sample. The energy spectrum of the source neutrons is tailored so that fission is induced only in the fissile isotopes (Chapters 6 and 9). Figure 2.7 shows the fission cross sections for some fertile and fissile nuclei.

From Figure 2.6, we see that the fission process results in a number of products: energetic fission fragments, neutrons, γ -rays, β -rays and

Figure 2.7. Fission cross section of ^{239}Pu , ^{235}U , ^{238}U , and ^{232}Th .

neutrinos (the last two are not shown in Figure 2.6), emitted either at the instant of fission or sometime later as the fission fragments undergo radioactive decay. Most ANDA techniques are based on measurement of the number of fission events, which occur in the sample, as a result of bombarding it with neutrons of a known intensity and energy.

Measurement of the fission reaction requires that one or more products of the reaction be determined. Fission fragments and β -rays do not travel large distances, from the point of fission in the sample, because of their charge; therefore, they cannot be detected from outside of the sample. γ -Rays and neutrons penetrate much more easily and can be detected outside the sample. All the usable fission signatures are based on these radiations which are discussed in more detail in Chapter 7.

2.7.2 The Reaction $(n,2n)$

The second multiplicative process of importance in ANDA is the $(n,2n)$ reaction, basically an absorption reaction, in which the compound nucleus decays by emitting two neutrons. It is important only at high neutron energies (≥ 7 MeV), although for some nuclei the threshold energy is much lower (1.8 MeV in ^9Be and 3.3 MeV in ^2H). The threshold energy for the $(n,3n)$ reaction is usually so high that it is not significant in most ANDA measurements.

The $(n,2n)$ process is important mainly in the moderation and, to some extent, in the multiplication of 14-MeV neutrons created by the $(d-t)$ reactions. Different interrogating spectra can be produced by surrounding the target of a 14-MeV neutron generator with various combinations of tungsten, lead, carbon, and polyethylene. Owing to the large $(n,2n)$ contribution from Pb and W near 14 MeV, the total neutron leakage from these materials exceeds generally the primary neutron intensity from the target. Since the energy of the neutrons produced by the $(n,2n)$ reaction is lower than that of the primary neutrons, it can also be used to soften the energy spectrum for specific applications. The energy spectra of neutrons resulting from the $(n,2n)$ reactions in Pb and Be are shown in Figure 2.8. The primary neutron energy range is 2 to 15 MeV (with an effective average $\bar{E} = 6.45$ MeV). Note the high intensity of $(n,2n)$ neutrons in Be as compared with Pb (indicated by the area under the histogram) that is explained by the high $(n,2n)$ cross section in Be (0.31 b) relative to the total cross section in this energy range (2.36 b). In contrast, the $(n,2n)$ cross section of Pb in the energy group 2 to 15 MeV is 0.0067 b, although its total cross section is 6.97 b. The inelastic, $(n,2n)$, and $(n,3n)$ cross sections of ^{238}U are shown in Figure 2.9. The rapid increase of the $(n,2n)$ cross section above the reaction threshold

Figure 2.8. Probability of generating a neutron in the i -th energy group by $(n,2n)$ reaction in group 1 (2-15 MeV) for Be and Pb.

Figure 2.9. Inelastic, $(n,2n)$ and $(n,3n)$ cross sections of ^{238}U (Ref. 4).

and the corresponding decrease of the inelastic scattering cross section should be noted.

2.8 SUMMARY

The interactions of neutrons with light and heavy nuclei important in ANDA are summarized in Table 2-2. The distinction between low- and high-energy neutrons is arbitrary because some processes occur in two or more energy ranges. Fission can be produced in various nuclides under different conditions. Slow neutrons cause fission in fissile isotopes but not in fertile ones. Fast neutrons with energies greater than 1 MeV cause fission in both. Inelastic scattering and emission of charged particles occur usually when the energy of the incident neutron is about 1 MeV or higher. The cross sections for reactions in which charged particles are emitted are usually much smaller than those for inelastic scattering and radiative capture, especially when the value of the atomic number (Z) of the target nucleus is high.

Table 2-3 summarizes the relevance and significance of the various neutron interactions to the various basic components of generic ANDA systems. For each component, neutron interactions, which significantly contribute to performance, are arranged in an approximately descending order of importance. The paramount role played by neutron moderation and thermalization is clearly reflected in the table that also indicates various other important elements used or possibly present. A more detailed discussion of the role of these elements will be found in later chapters, especially in Chapter 6.

Table 2-2. Interactions of neutrons with light and heavy nuclei.

Energy	Nuclei	
	Light and Intermediate ($A < 80$)	Heavy ($A > 80$)
Low (0-1 keV)	$(n,n)^a$	(n,γ)
	(n,γ)	(n,f)
		(n,n)
Intermediate (1-500 keV)	(n,n)	(n,n)
	(n,γ)	(n,γ)
		(n,f)
High (0.5 - 10 MeV)	(n,n)	(n,n)
	(n,n')	(n,n')
	(n,γ)	(n,p)
	(n,p)	(n,γ)
	$(n,2n)$	$(n,2n)$
		(n,α)
		(n,pn)
		(n,f)

^aThe symbol (n,n) represents neutron elastic scattering. The other symbols were defined before.

Table 2-3. Important neutron reactions and their relevance to various general components of ANDA systems.

ANDA System Component	Reaction	Materials
Source spectrum-tailoring	(n,n)	Low and medium A^a , (for example, H, Be, D as moderator). Fe, Ni (as fast neutron reflector).
	(n,n')	High and medium A (for example, Pb, W, Fe).
	$(n,2n)$	Be, high A (for example, Pb, W, U).
Interrogated sample	(n,n)	Low A , especially H.
	(n,γ)	All A .
	(n,f)	$A > 232$ (for example, ^{238}U , ^{235}U , ^{239}Pu).
Detector assembly	(n,n)	H, ^4He .
	Capture (n,p) ; (n,α) , etc.	^{10}B , ^3He , ^6Li .
	(n,γ)	All A .

^a A = atomic mass.

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* Available for purchase from National Technical Information Service, Springfield, Virginia 22161.

CHAPTER 3

INTERACTIONS OF γ -RAYS WITH MATTER

3.1 INTRODUCTION

The various interactions of γ -rays with matter influence, in several ways, the proper design, analysis, and use of ANDA methods. First, high-energy (5 to 10 MeV) γ -rays may be used as an interrogation source to stimulate photofission or to produce neutrons by the (γ, n) process. They also may be used to measure density and composition by transmission techniques. Second, prompt and delayed γ -rays resulting from fissions are a significant part of the signatures of the fission events. These signatures may be affected by γ -ray interactions with the sample and surrounding materials. Third, detection and analysis of γ -rays depend on their interaction with the detector materials. γ -Rays constitute also an important portion of the background measured by the detector.

Attenuation of a beam of γ -rays is similar to attenuation of a beam of fast neutrons (see Section 2.2). If a beam of γ -rays penetrates a very thin slab of thickness Δx the change in its intensity is directly proportional to the thickness of the slab and the intensity of the incident beam. Attenuation of γ -rays conforms to the exponential attenuation law for the intensity, I , of transmitted photons:

$$I/I_0 = e^{-\mu x}. \quad (3-1)$$

The constant $\mu(\text{cm}^{-1})$ —the attenuation coefficient—plays the same role as the macroscopic cross section, Σ , in neutron attenuation.

Eq. 3-1 is valid only in “good geometry,” where the γ -rays are monoenergetic, the beam is collimated, and the absorber is thin. It is often used, however, as a first order approximation in other geometries.

The value of the attenuation coefficient depends on the nature of the absorber and the initial energy of the γ -rays. From values of μ tabulated

by Hubbell (Ref. 1) and Davisson (Ref. 2), two general characteristics of μ can be deduced: (a) for a given element, μ first decreases as the energy of the γ -rays increases, passes through a broad minimum, and then increases; and (b) in general, μ is greater for heavy elements than for light ones.

The attenuation coefficient can also be expressed in units of either cm^2/atom (σ_a) or $\text{cm}^2/\text{electron}$ (σ_e). Sometimes it is more convenient to use the mass attenuation coefficient (μ/ρ), cm^2/g . These quantities are related:

$$\mu/\rho = N \frac{Z}{A} \sigma_e = \frac{N}{A} \sigma_a \quad (3-2)$$

where ρ is absorber density; Z is the atomic number; A is the atomic weight; and N is Avogadro's number (6.02×10^{23}).

The mean free path, λ , for a γ -ray in a medium with attenuation coefficient μ is:

$$\lambda = \frac{1}{\mu} \text{ (cm)}. \quad (3-3)$$

For practical purposes, one often defines a *half-value layer*, the thickness (in cm) required to attenuate a beam by a factor of two. The half-value layer is thus equal to $(\ln 2)\lambda = 0.693\lambda$. In Figures 3.1 and 3.2, the half-value layer is plotted as a function of γ -ray energy for various elements (Ref.2).

From Eq. 3-1 it may be deduced that $\mu x = (\mu/\rho) \rho x$ is constant for a given degree of attenuation. The mass attenuation coefficient, μ/ρ , is nearly constant (for Compton scattering) for all elements (except hydrogen, for which its value is about twice the average). Thus, for a given attenuation in different materials, ρx is constant. The thickness required to attenuate the γ -rays by a given fraction is, therefore, inversely proportional to the density.

Three processes are chiefly responsible for the attenuation of γ -rays in the energy range of usual interest for ANDA measurements (0.1 to 3 MeV): photoelectric absorption, Compton scattering, and electron-positron pair production. In other, higher energy ranges and in certain elements, photonuclear reactions such as (γ, n) and (γ, f) should also be considered. These and other interactions of photons with matter are shown in Figure 3.3. The total attenuation coefficient of a given material is obtained by summing the partial coefficients:

$$\mu(E) = \tau(E) + \sigma(E) + \kappa(E) \quad (3-4)$$

where τ , σ , and κ denote the photoelectric, the Compton, and the pair production coefficients, respectively. In Eq. 3-4, the small contribution of the photonuclear reactions is neglected.

Figure 3.1. Half value layer versus photon energy (Ref.2).

Figure 3.2. Half value layer versus photon energy (Ref. 2).

Since γ -ray energies of interest to ANDA are usually from 0.1 to 3 MeV, normally, the most important process is *Compton scattering*. In this process, the photon loses part of its energy as a result of a scattering interaction with an electron, which can be considered free, since the photon's energy is large as compared to the electron's binding energy.

Figure 3.3. Schematic representations of γ -ray interactions.

After scattering, the photon continues to move in a different direction with a lower energy. It may undergo more scattering, until either it escapes from the medium or its energy is reduced to a level where the photoelectric process becomes more probable, and the photon is absorbed. This multiple scattering affects particularly the analysis of fast coincidence measurements because false coincidences may be caused by a single photon scattered two or more times between the coincidence detectors.

The next important process to ANDA is photoelectric absorption. It is the dominant interaction at low photon energies (up to a few hundred keV) and with materials of high atomic number (Z). In this process, the photon transfers all of its energy to a bound electron that is first ejected from the atom and then reabsorbed near where the interaction occurred. At energies above 1 or 2 MeV, photoelectric absorption is rare.

Pair production is the third kind of γ -ray interaction. It is possible at high energies (above 1.02 MeV). In this interaction, the photon is completely absorbed and is replaced by an electron-positron pair whose total energy equals the initial photon energy. It is more important with heavy elements than with light ones.

In the following sections, these interactions of photons with matter are described in more detail. In addition, photonuclear and photofission processes are discussed. For a more complete discussion, see Davisson and Evans (Ref. 3).

3.2 PHOTOELECTRIC EFFECT

Photoelectric absorption is important in several areas of ANDA. It accounts for absorption of delayed γ -rays in fuel rod scanners and also for absorption of γ -rays by heavy materials in a matrix. It is also partially responsible for the effectiveness of γ -ray shielding. In many types of γ -ray detectors, the photoelectric process is the principal mechanism in the determination of γ -ray energy.

The photoelectric process may be visualized as an interaction of a photon with the atomic electron cloud in which the entire photon energy, $h\nu$, is absorbed and an electron is ejected from the atom with kinetic energy, T :

$$T = h\nu - BE \quad (3-5)$$

where BE is the binding energy of the ejected electron. Usually, the electron kinetic energy, T , is 100 keV or less so that the electron does not travel far before it is reabsorbed.

Because a general formula for the probability that a photon of energy $h\nu$ will undergo the photoelectric process is not easy to obtain, several different formulas must be used for photon energies from 0.1 to 10 MeV. A crude, but useful, approximation for the photoelectric absorption coefficient, τ (cm^2/atom), is as follows (Ref.4):

$$\tau \sim \text{const} \cdot \frac{Z^4}{(h\nu)^2} \quad (3-6)$$

where Z is the atomic number. The exponent of Z changes from 4 to 4.6 for $h\nu$ between 0.1 to 3 MeV.

Eq. 3-6 shows that for a given photon energy, photoelectric absorption is stronger in heavier (high Z) materials, such as Pb or U, than in lighter ones, such as Al and Fe. The effect is also much greater at lower photon energies than at higher ones. Values of τ , as a function of photon energy and atomic number, are given in (Ref. 1 and 2). Figure 3.4 shows the photoelectric cross section, (τ/ρ) , in lead as a function of

Figure 3.4. Photoelectric mass attenuation coefficient as a function of photon energy for lead.

the energy of the incident photon. (The discontinuities at low energies occur when the photon energy becomes less than the *K*- and *L*-shell electron binding energies. These electrons cannot then participate in the process, so the cross section decreases.)

3.3 COMPTON SCATTERING

The Compton scattering process lends itself more easily to exact theoretical treatments than the photoelectric absorption process. The process alters the trajectory of the photon, leaving it as a lower energy. This final energy, following a Compton scattering at an angle θ relative to the original direction of the photon, is (Ref. 4, 5):

$$E_1 = \frac{E_0}{1 + \frac{E_0}{MC^2} (1 - \cos \theta)} \quad (3-7)$$

$$MC^2 = 0.511 \text{ MeV}$$

where the scattered (E_1) and initial (E_0) energies are expressed in MeV. Figure 3.5 is a plot of this equation for various values of E_0 and θ . Note that the Compton process is very effective for reducing high energies, whereas photons with low energies (<0.5 MeV) lose little energy, regardless of the scattering angle.

The relative probability of scattering through a given angle is given by the Klein-Nishina formula for the differential scattering cross section

(Ref. 4, 5). Figure 3.6 shows the differential cross section as a function of energy and angle. As the incident photon energy increases, the average scattering angle decreases; that is, the scattered photon distribution becomes more peaked forward. The total scattering cross section is slowly decreasing function of energy. The decrease is quite gradual at low photon energies, it behaves like $1/h\nu$. For energies above 1 MeV, it is roughly proportional to $(h\nu)^{-1}$. Thus, Compton scattering decreases

Figure 3.5. Dependence of Compton-scattered photon energy (E_1) on initial energy (E_0) and scattering angle.

Figure 3.6. Klein-Nishina differential cross section for the number of photons scattered/unit solid angle in the direction θ .

much more slowly with increasing energy than does photoelectric absorption. With a light element it is the main photon interaction process over the entire energy range of interest to ANDA. Even with heavy elements, it is the dominant process in the energy range of 0.6 to 2.5 MeV.

The importance of multiple Compton scattering is demonstrated by a typical pulse height distribution (measured with NaI detector) of scattered photons shown in Figure 3.7 (Ref. 6). The scatterer is an aluminum sphere, and the source is ^{60}Co ($E = 1.25$ MeV). The spectrum consists of two broad maxima: the high-energy, and the low-energy ones. A "single scatter peak," the high-energy maximum, results from photons that scatter once in the sample and escape from the scatterer without a second interaction. The energy of these photons is determined by the Compton scattering formula (Eq. 3-7).

The broader maximum at low-energy results from multiply scattered photons that are subjected to two or more interacting phenomena. In the first scattering, as indicated in Figure 3.5, the energy lost/collision by a photon undergoing successive collisions decreases, both relatively and absolutely, as the photon's energy diminishes. For example, a 1.33-MeV γ -ray scattered through 180° loses 84 percent of its initial energy, but a 50-keV γ -ray scattered through the same angle loses only 16 percent of its initial energy. Thus, a photon loses the bulk of its energy in the first few collisions with the medium, after which it may scatter several more

Figure 3.7. Pulse-height spectrum of ^{60}Co γ -rays backscattered from an aluminum sphere.

times without an appreciable further loss of energy before it is either scattered away from the medium or is absorbed by the photoelectric process. The second phenomenon is represented by the rapidly increasing absorption to scattering ratio caused by the fast increase in the photoelectric cross section as the photon energy decreases (see Section 3.2). This results in the abrupt truncation of the energy spectrum that can be noted in Figure 3.7. As a consequence of these effects, the shape and energy of the multiple scatter "peak" are virtually independent of the source energy.

Because the photoelectric cross section at low energies is relatively large in heavy elements, high-energy photons that have been scattered only once dominate in samples containing significant amounts of high- Z materials, like UO_2 . On the other hand, the effect of multiple scattering should be considered in low- Z moderators and detectors, such as hydrogen-containing materials and plastic scintillators. Figure 3.8 shows the ratio of single to total scattering as a function of the scattering angle, target thickness, and atomic number (Ref. 7). Note that for heavy materials and thin targets, most of the scattered flux consists of singly scattered photons. In Figure 3.9, the contribution of the singly scattered photons from a ^{137}Cs source at an angle of 120° is shown for various scatterers of infinite thickness. For hydrogen-containing materials (for example, plexiglass, plastic and liquid scintillators, etc.), the multiply scattered photons contribute more than 80 percent of the scattered flux, thus requiring complex transport calculations to correct for deleterious effects on the accuracy in single and coincidence counting.

Figure 3.8. Measured ratio of single to total scattering photon yield for various scatterers and detection angles.

Figure 3.10. Pair-production mass attenuation coefficient as a function of photon energy for uranium and beryllium.

Figure 3.11. Mass absorption coefficients for Pb as a function of photon energy.

Figure 3.12. Regions of dominance for photon interaction mechanisms.

pair. The remaining 1.02 MeV resides in the rest masses of the positron and electron. This energy is given up after the positron has slowed down by ionization and radiative collisions and has annihilated itself by combining with another electron. As a result, two 0.511 MeV photons are emitted isotropically with respect to the original primary photon and at 180° with respect to each other. These photons may further interact with the sample by Compton scattering and may finally be absorbed in a photoelectric process or escape the sample.

3.5 PHOTONUCLEAR ABSORPTION

Photonuclear absorption (Ref. 8, 9) consists of nuclear (rather than atomic) interactions initiated by the absorption of a photon. The most likely result of such interactions is the emission of a single neutron (γ, n) reaction, but the emission of charged particles, γ -rays, or more than one neutron are also possible. Any of these processes can occur upon the absorption of a photon of greater energy than the separation energy of the emitted particle (photon, neutron, or α -particle). Thus, photonuclear absorption is a threshold process. Except for deuterium and beryllium, the threshold level lies between 5 to 13 MeV. The cross section increases rapidly above this threshold energy to form the "giant resonance." This is the most characteristic feature of the cross section for the (γ, n) reaction. It is a broad peak, in the absorption cross section, whose center decreases from 24 MeV for lighter nuclei to 12 MeV for the heaviest stable nuclei. The width of the resonance, that is, the energy difference between the points at which the cross section drops to one-half its maximum value, varies from about 3 to 9 MeV, depending on the detailed properties of the nucleus.

The magnitude of the cross section, even at resonance peak energy, is small in comparison with the sum of the competing Compton scattering and pair-production cross sections (see Sections 3.3 and 3.4). Its size never represents more than 10 percent of the total cross section. However, photonuclear absorption can be of considerable importance in ANDA measurements, whenever interrogating photon energies exceeding specific reaction threshold energies are present. Since neutrons are produced in a large fraction of the reactions (about 40 to 100 percent probability depending on photon energy and mass number of the nucleus), photonuclear absorption by matrix materials should be considered in all ANDA techniques involving high-energy γ -rays.

The low threshold energies of deuterium (2.23 MeV) and beryllium (1.69 MeV) present particular difficulties. Deuterium is present in small amounts in all hydrogen-containing materials (0.015 percent of hydrogen content), and its (γ, n) reaction can increase the neutron background relative to the measured signals, namely, prompt neutrons from photofissions. However, photonuclear reactions with deuterium and beryllium can also produce strong intensities of MeV and sub-MeV neutrons as interrogating sources for ANDA (see Sections 4.3 and 4.5).

At γ -ray energies near 20 MeV, $(\gamma, 2n)$ and (γ, np) reactions become possible. The cross sections of these reactions increase with increasing energy while the (γ, n) and (γ, p) cross sections decrease in this energy region. However, because of the high photon energies required for these reactions, they have little effect in most practical ANDA measurements.

3.6 PHOTOFISSION PROCESS

High-energy γ -rays can induce fission. The amount of excitation energy required to make fission possible can be estimated from the height of the nuclear-potential barriers and the dissociation energy for the particular mode of fission. The dissociation energy can be calculated from the mass difference between the fission fragments and the original nucleus.

The photofission process (Ref. 10) is a barrier-penetration process, similar to neutron-induced fission in isotopes with even atomic weight, such as ^{232}Th and ^{238}U (see Section 2.8). Below the effective photofission "thresholds" (about 6 MeV), the photofission cross sections drop extremely rapidly. Photofission cross sections of ^{239}Pu , ^{238}U , and ^{232}Th in the region of interest to the applications of photon-induced reactions in ANDA (4 to 10 MeV) are shown in Figure 3.13 (Ref. 11). The cross sections well above the photofission barrier are about the same for all isotopes. Near and, especially, below the barrier the cross sections are vastly different, a characteristic of prime importance for ANDA

Figure 3.13. Approximate photofission cross section for ^{239}Pu , ^{238}U , and ^{232}Th between 4 to 10 MeV photon energies (Ref. 11). (The cross sections are based on an approximate unfolding of the yield measurements. The neutron binding energy is indicated on the energy abscissa.)

applications. The differing cross sections make it possible to distinguish between the various fissionable isotopes.

To determine the amounts of individual isotopes in a sample, one has to carry out several measurements at different γ -ray energies. At any given energy one can, in principle, simultaneously produce three independent quantities: prompt neutrons, delayed neutrons, and delayed γ -rays. (Prompt γ -rays from photofission are very difficult to measure because of the extremely high background of γ -rays from the source.) These signatures are direct consequences of induced photofission. Because the purpose of the photo-induced ANDA technique is to determine the presence of isotopes, to distinguish between them, and to determine their amounts, the threshold and the subthreshold regions are obviously the most promising regions (see Figure 3.13 for examples). The various signatures resulting from photofission are discussed in Chapter 7, and their applications are described in Section 9.7.

3.7 SUMMARY

There are three principal modes of γ -ray interactions with matter: photoelectric absorption (dominant at low energies and with heavy materials); Compton scattering (dominant at intermediate energies, from 0.6 to 2.5 MeV, and to much higher energies with light materials); and pair production (dominant at high energies). At photon energies above 5 MeV, photofission and (γ, n) reactions are important, especially the former, as a means of interrogation of fissionable material. These interactions are shown schematically in Figure 3.3. The roles played by the various interactions in ANDA are summarized in Table 3-1. It shows the principal effects of these mechanisms on different parts of ANDA systems. Within each portion the listing is roughly in order of decreasing importance.

Table 3-1. Effects of γ -ray interactions on ANDA systems.

System Component	Reaction	Comments
Source	(γ, n)	Neutrons are produced.
Sample	Compton	Attenuation of emitted γ -rays in prompt coincidence and delayed γ -techniques.
	Photoelectric	
	Pair production	Photofission interrogation.
	(γ, f)	
	(γ, n)	
Detector	Pair production	Attenuation of incoming γ -rays in (γ, f) techniques.
	Compton	
	Compton	Provides detection mechanism and creates background in some cases.
	Photoelectric	
	Pair production	Background in neutron detectors in photofission techniques.
	(γ, n)	

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CHAPTER 4

NEUTRON PRODUCTION AND SOURCES

4.1 INTRODUCTION

The interrogation source is an essential part of any ANDA system. It provides the particles that, after some spectral modification, stimulate fissions in samples. The fissions are detected, and the amount of the fissionable materials present is then inferred. The only practical interrogation particles are neutrons and γ -rays because only these radiations can probe deep into the sample. Most ANDA systems are based on neutron sources because of their variety and ease of use. Intense high-energy γ -rays have also proved effective as interrogation particles, but they can be obtained only from electron accelerators. Although they are stable and reliable, the accelerators' complicated operation and maintenance often makes them at present more difficult in field use.

The variety of sources usable in ANDA systems is shown in Table 1-2 (see Section 1.4) that lists ANDA systems generically according to the type of interrogating source. ANDA systems use 14-MeV neutron generators, (α, n) isotopic sources, (γ, n) isotopic sources, ^{252}Cf spontaneous fission sources, nuclear reactors, and accelerator photo-induced neutron sources. Each source has its special merits that make it particularly suitable for specific applications. Some applications, for example, require high-intensity sources. The least expensive and most intense sources are 14-MeV neutron sources with intensities between 10^9 and 5×10^{11} n/s. High intensity ^{252}Cf and Sb-Be (γ, n) sources are also available. Some applications require moderate intensity sources of sub-MeV neutrons which can stimulate fissions in the fissile isotopes without fissioning the more abundant fertile isotopes. Sources based on the (α, n) reaction of Li and (γ, n) sources are the best examples of such isotopic sources. Other sources emit neutrons at energies, generally, above the fission threshold of the fertile isotopes; a tailoring assembly is then required to reduce the neutron energy.

In this chapter, some general properties of charged particle interactions are first discussed and illustrated with (α, n) sources. Then, isotopic photoneutron sources and spontaneous fission sources are presented. Next follows a discussion of accelerator-based sources: neutron generators and accelerator photoneutron sources. The chapter concludes with a comparison of the main sources, which should allow a potential ANDA user or designer to select a proper interrogation source.

4.2 α -EMITTERS AND CHARGED PARTICLE INTERACTIONS

Since Chadwick discovered the neutron in 1932, nuclear reactions have been utilized to produce neutrons. Probably, the most common type of source has been radium-beryllium, using the (α, n) reaction. With the advent of the atomic energy program, other α - and γ -emitters capable of α -neutron and γ -neutron reactions have become more readily available. In general, α -neutron reactions produce more neutrons than γ -neutron reactions, so α -neutron sources are the more common ones. In addition to intensity, however, the suitability for safeguards of an α -neutron source also depends on its half-life, energy, accompanying radiation, and availability.

α -Particles originate either in the natural α -emission of radioactive heavy elements or are produced and accelerated in charged particle accelerators. In the former case, there are 17 light elements that can serve as target materials and over 40 α -emitting isotopes, which theoretically, can be mixed to create neutrons. Among the light elements, lithium, beryllium, boron, and fluorine give the highest neutron yields. Simple, compact, and useful interrogation sources can be made from combinations of a heavy element α -emitter (for example, plutonium, radium, polonium, or americium) and one of these light elements. α -Neutron sources based on other light elements, such as oxygen, are of little value for practical ANDA purposes, although they may be useful in studies of the properties of the α -neutron reaction in these elements. Such studies may be important in understanding background neutrons originating in the oxygen and aluminum present in plutonium oxide samples.

Ten α -emitters have half-lives from a few days to 5,000 yr and five of these have half-lives from a few months to 1,620 yr. In general, the shorter the half-life, the higher the neutron yield for the same target materials. The relationship between activity in units of curies* (Ci) and half-life is:

$$\text{Activity} = 1.124 \times 10^{13} M/T_{1/2} A \text{ Curies} \quad (4-1)$$

*1 curie (Ci) = 3.7×10^{10} disintegrations/s.

where M is the mass (in grams), $T_{1/2}$ is half-life (in seconds), and A is the atomic mass. Thus, α -emitters with long half-lives require greater mass to produce a given activity and, hence, a given neutron flux. Although long-lived α -emitters have the advantage of nearly constant activity, their α -particle energies are generally lower than for short-lived emitters. The lower energies result in greater self-absorption and lower neutron yields.

In addition to (α, n) reactions induced by heavy element α -sources combined with light element targets, (α, n) and other nuclear reactions using accelerated charged particles are frequently used as a source of neutrons. Some of these reactions are:

(α, n)

(p, n)

(d, n)

The energy value given for each of the nuclear reactions is called the Q value of the reaction, or the nuclear disintegration energy (Ref. 1, 2). The Q value expresses the change in the total rest mass (or rest energy) between the projectile (α) plus the target nucleus (for example, ${}^9\text{Be}$) and the emitted particle (for example, neutron) plus the residual nucleus (for example, ${}^{12}\text{C}$):

$$Q = (m_t + m_p)c^2 - (m_n + m_r)c^2 \quad (4-5)$$

where c is the velocity of light and m_t , m_p , m_n , and m_r are the rest masses of the target, projectile, neutron, and the residual nuclei, respectively.

Positive Q values indicate exothermic reactions and negative values indicate endothermic reactions. In exothermic reactions, the kinetic energy of the products (for example, neutrons and residual nuclei) exceeds that of the input particles (for example, target and projectile nuclei), and the reaction can proceed even if the projectile has zero

energy. An endothermic reaction, on the other hand, requires that the projectile have kinetic energy.

The energy and direction of motion of the emerging particles can be derived from the equations of conservation of energy and momentum (Ref. 1). For exothermic reactions, for example, ${}^9\text{Be}(\alpha, n){}^{12}\text{C}$, the neutron energy for zero α -energy is given by:

$$(E_n)_{\min} = Q \frac{m_r}{m_n + m_r}. \quad (4-6)$$

In general, neutron energy is an increasing function of Q . Therefore, beryllium yields the highest neutron energies among (α, n) sources. When the projectile particle, for example, α , has a non-negligible kinetic energy, which is the usual case, the neutron energy also depends on the angle of emission with respect to the direction of the projectile. For example, the ${}^9\text{Be}(\alpha, n){}^{12}\text{C}$ reaction with 7.68-MeV (${}^{214}\text{Po}$, that is, RaC') α -particles yields 13.1 MeV neutrons in the forward direction and 7.7 MeV neutrons in the backward direction. Energies in other directions lie between these two values.

The neutron spectrum resulting from an (α, n) reaction has a complex structure. (See Figure 4.1 for the case of Pu-Be.) The general features of the spectrum can be explained as follows:

1. Neutrons with varied energies are observed, since for each incident α -particle energy the neutron energy is dependent on the angle of emission.
2. The α -particles are continuously slowed down in beryllium; hence, (α, n) reactions occur at all energies between zero and the maximum α -particle energy.
3. The residual nucleus (${}^{12}\text{C}$) can be left in an excited state. This results in a large number of neutrons with a neutron energy smaller than $(E_n)_{\min}$ (5.3 MeV). This is the kinetic energy of the neutrons produced by zero energy α -particles when the ${}^{12}\text{C}$ nucleus is left in its ground state.
4. Low energy neutrons can be produced by the photoeffect in Be.

Endothermic reactions have a threshold energy, $(E_p)_{\text{thres}}$, below which the projectile will not cause the reaction. This threshold is given by (Ref. 1):

$$(E_p)_{\text{thres}} = Q \frac{m_p + m_t}{m_t} \approx Q \quad (\text{for heavy targets}). \quad (4-7)$$

The neutron energy at the threshold, $(E_n)_{\min}$ is given by

Figure 4.1. Comparison of one calculated and two measured neutron spectra. (a) 1 Ci Pu-Be source (Ref. 3). (b) 10 Ci Pu-Be source. [For details on the various experimental and calculational results see (Ref. 3 and 4.)]

$$(E_n)_{\min} = (E_p)_{\text{thres}} \frac{m_p \cdot m_n}{(m_n + m_r)^2}. \quad (4-8)$$

Thus, endothermic (α, n) reactions and other charged particle interactions can supply low-energy neutrons in the sub-MeV region. ${}^7\text{Li}(\alpha, n){}^{10}\text{B}$ and ${}^7\text{Li}(p, n){}^7\text{Be}$ reactions are most useful for this purpose in ANDA.

4.3 ISOTOPIC α -NEUTRON SOURCES

A strong isotopic source that approximates a point source of nearly constant intensity can be made by using the reaction ${}^9\text{Be}(\alpha, n){}^{12}\text{C}$ and the strong α -activity of natural radium, plutonium-238 or americium-241. Radium-beryllium neutron sources are frequently used in laboratories as standard calibration sources. These sources consist of a mixture

of radium bromide and beryllium powder (usually in a ratio of 1:5) compressed under high pressure and carefully placed into a soldered shell of brass or nickel. This shell is usually enclosed in a second shell to prevent the emission of radon, a gaseous daughter product of radium. Sources with as much as 5 g of radium (approximately 5 Ci) have been manufactured. Great care is required in the manufacture of Ra-Be sources, if the neutron intensity is to be constant over long periods. There are two main disadvantages of the Ra-Be source: (1) the strong γ -radiation that accompanies the neutrons, and (2) its complex, broad neutron energy spectrum that varies continuously up to a maximum energy of 13.1 MeV, corresponding to the largest α -energy (RaC') of 7.68 MeV. The most probable energy is 4 MeV and the average energy is about 5 MeV.

Another useful and easily available source is Po-Be. Polonium-210 has an α -energy of 5.305 MeV and a half-life of 138.5 days. It does not emit β - or γ -rays, but its short lifetime is a drawback. About 2.5×10^6 n/s/Ci of ^{210}Po can be obtained from a Po-Be source. The average neutron energy of this source is about 4 MeV. The maximum energy is somewhat less than that of Ra-Be because of the lower α -particle energy.

Plutonium-239, which became available in relatively large quantities a decade ago, has also been used for the manufacture of (α, n) sources. Plutonium-239 decays with a half-life of 24,360 yr. The α -particle energies are 5.15, 5.13, and 5.10 MeV. A ^{239}Pu -Be neutron source emits 2×10^6 n/s/Ci of plutonium. Figures 4.1a and b show calculated and measured neutron energy spectra for 1 and 10 Ci Pu-Be sources (Ref. 3, 4). It is apparent that the shape of the source and the amount of Be affect the emerging neutron spectrum (Ref. 3).

There are several important advantages to ^{239}Pu -Be sources:

1. They are highly reproducible because the plutonium and beryllium are alloyed.
2. They emit no penetrating γ -radiation except for the 4.43 MeV line resulting from de-excitation of ^{12}C , emitted in about 80 percent of the Be (α, n) reactions.
3. They have a very long lifetime.

However, the small specific neutron emission rate from ^{239}Pu -Be sources requires significant amounts of ^{239}Pu to make strong sources (16 g/Ci), which is a distinct drawback, since ^{239}Pu is a fissile isotope and large quantities pose a safety hazard. This disadvantage can be overcome by employing ^{238}Pu as the α -emitter, which has a half-life of 89 yr, and is not truly fissile (it has much lower fission cross section than ^{239}Pu at thermal energies). Its main α -particle energies are about 5.5 MeV with a

very low γ -emission. Plutonium-238 also decays by spontaneous fission, although at a much slower rate than by α -decay. By spontaneous fission, 1 g of ^{238}Pu (17.2 Ci of α -activity) emits about 2500 n/s (about 1100 fissions/s/g), an important fact only when sub-MeV (α -n) sources are used with fission multiplicity detectors. (See Chapter 9.)

If large specific activities are required, ^{210}Po , ^{227}Ac , ^{244}Cm , and ^{248}Cm can be used. Curium-244 has a half-life of 18 yr and is sometimes used instead of ^{252}Cf to simulate the fission spectrum; however, its currently high price seriously limits its use. This situation may change in the future as the demand for strong, long-lived neutron sources increases.

The most common α -emitter in use today is ^{241}Am . Its availability, convenient long half-life (432 yr), low-energy γ -rays, and low fission-ability make it the basis for most (α -n) sources used in ANDA. The neutron spectrum from a 1-Ci Am-Be source is shown in Figure 4.2.

α -Neutron sources utilizing beryllium are characterized by average neutron energies of a few MeV. If sub-MeV neutrons are used to interrogate fissile samples, spectrum tailoring (see Chapters 2 and 6) must be employed. On the other hand, the combination of a target material with a high negative Q value, such as lithium, with an α -source emitting α -particles, just over the threshold energy, will yield a much softer spectrum (that is, lower neutron energy) than that produced by target materials having positive Q values.

Figure 4.2. The neutron spectrum of 1 Ci Am-Be source determined by various methods (Ref. 4). Error bars define the scatter of experimental points.

[For details on the various experimental and calculated results see (Ref. 3 and 4).]

Figure 4.3 compares the main features of the neutron spectra of four important neutron sources: $^{241}\text{Am-Be}$, ^{252}Cf , $^{241}\text{Am-Li}$, and $^{124}\text{Sb-Be}$. The softer spectrum of the $^{241}\text{Am-Li}$ source stands out clearly in comparison to the beryllium sources. It is even softer than the ^{252}Cf spontaneous fission spectrum. The relatively low neutron energies from the $^{241}\text{Am-Li}$ source make it very useful in ANDA instruments assaying material of low enrichment fuel. No substantial moderation of

Figure 4.3. Neutron energy spectra for: a. $^{241}\text{Am-Be}$ and ^{252}Cf sources. b. $^{241}\text{Am-Li}$ source. c. $^{124}\text{Sb-Be}$ source. (Note the different energy scales.)

source neutrons is required to reduce the fission contribution from the abundant fertile isotope. In addition, the high penetrability of the neutrons, which have energies between 100 to 500 KeV, is maintained. In addition to $^{241}\text{Am-Li}$, commonly used sources* with the properties described above include $^{210}\text{Po-Li}$ and $^{238}\text{Pu-Li}$.

The disadvantage of these sources is that the neutron yield is very low, a result of the small (α, n) cross section just above the threshold energy. For example, the yield (n/s·Ci) from a $^{210}\text{Po-Be}$ (α, n) source is about 60 times greater than that of a $^{210}\text{Po-Li}$ source. Thus, the physical size and cost of strong sources can become prohibitive.

Tables 4-1 and 4-2 contain information on the (α, n) sources discussed here, including the background radiation from the sources. In addition to α -particles, most sources emit γ -rays. The γ -dose rate from the various sources is also indicated in the tables. It is apparent that the source most easily shielded is the ^{210}Po α -source. Other sources, like ^{228}Th , actually consist of a number of relatively unstable daughter nuclei which decay by α - and β -particle emission, accompanied by a relatively large number of γ -rays.

Another form of background radiation produced by isotopes like ^{238}Pu , ^{240}Pu , and ^{242}Pu which undergo spontaneous fission as well as

Table 4-1. Data on $\text{Be}(\alpha, n)$ sources.^a

α -emitter	$T_{1/2}$	$\bar{E}_\alpha^b$ (MeV)	γ mrad/h at 1m/10 ⁶ n/s	$\bar{E}_n^b$ (MeV)	n/s/Ci (x10 ⁶)
^{210}Po	138d	5.3	0.02	4.3	2.2
^{226}Ra	1620y	5.6	60	~4.5	13
^{227}Ac	21.8y	6.5	8	~4.7	15
^{228}Th	1.91y	6.4	30	~4.5	20
^{238}Pu	89y	5.5	0.08	~4.5	2.2
^{239}Pu	24360y	5.1	<1	4.5	1.5
^{241}Am	458y	5.5	0.05	~4.5	2.2
^{242}Cm	163d	6.1	0.06	~4.5	2.5
^{244}Cm	18.1y	5.8	0.04		2.5

^a(Ref. 5).

^bAverage energy of α -particle or neutron.

*Whenever the oxide rather than the metallic form of the α -emitter is used, as is the case for ^{241}Am and ^{238}Pu , (α, n) reactions on oxygen contribute significantly to the high-energy tail of the neutron spectrum.

Table 4-2. Comparison of characteristics of isotopic (α, n) sources.^a

Source	Specific Neutron Yield, 10^6 n/(s Ci)	Maximum Neutron Energy, MeV	Approximate Neutron Mean Energy, MeV	γ mrad/h at $1 \text{ m}/10^6$ n/s
$^{210}\text{Po} + \text{Be}$	2.2	11.3	5.3	10^{-2}
$^{210}\text{Po} + \text{B}$	0.8	5.2	3.0	10^{-2}
$^{210}\text{Po} + \text{F}$	1.4	3.0	1.1	—
$^{210}\text{Po} + \text{Li}$	0.1	1.3	0.3	3×10^{-4}
$^{210}\text{Po} + \text{O}$	2.6×10^{-3}			2×10^{-5}
$^{210}\text{Po} + \text{O}^{18}$	1.1	4.3	2.3	1×10^{-2}
$^{226}\text{Ra} + \text{Be}$	13	13	4.7	60
RaBeF_4	3	13	4.7	60
$\text{Rn} + \text{Be}$	14	13	4.7	60
$^{227}\text{Ac} + \text{Be}$	15	13	4.7	16
$^{238}\text{Pu} + \text{Be}$	2.5	11.3	4.7	$6 \times 10^{-3}{}^c$
$^{239}\text{Pu} + \text{Be}$	2.3	11.0	4.5	6^c
$^{241}\text{Am} + \text{Be}$	2.7	11.5	4.5	6^c
$^{242}\text{Cm} + \text{Be}$	2.4	12	—	$6 \times 10^{-3}{}^c$
$^{241}\text{Am} + \text{Li}$	0.04	1.2	0.3	2.5^c

^a(Ref. 6). Differences between sources made of same materials are real and represent different designs.

^b $^{210}\text{Pb} = \text{RaD}$.

^cSoft γ -radiation.

α -decay consists of prompt fission neutrons and γ -rays. The ratio of spontaneous fission neutrons to (α, n) neutrons can range from a negligible amount (7×10^{-6}) for ^{239}Pu to about 2×10^{-2} for ^{238}Pu . Spontaneous fissions, in general, do not significantly enhance neutron production by (α, n) sources. Their presence, however, can constitute an important part of the background in systems based on coincidence detection schemes such as Fission Multiplicity Detectors (see Chapter 9). Further discussion of (α, n) sources in safeguards applications is given by Beyster and Kull (Ref. 7).

4.4 ISOTOPIC PHOTONEUTRON SOURCES

Isotopic photoneutron sources are attractive for ANDA systems, primarily, because they provide ample intensities of sub-MeV neutrons with energies well above thermal energy. These neutrons penetrate well into the samples but do not fission the abundant fertile isotopes. Therefore, it is not surprising that systems with isotopic photoneutron sources—in particular, Sb-Be—were investigated rather early in ANDA development (Ref. 8, 9).

The minimum γ -ray energy necessary for photoneutron interactions with most nuclei is about 8 MeV; however, it is only 1.67 MeV with beryllium and 2.23 MeV with deuterium. Since the γ -energy of radioactive nuclides rarely exceeds 3 MeV, these are the only materials in which photoneutron reactions can be considered.

Beryllium has often been used as a source of photoneutrons by means of the following reaction:

with a Q value of 1.666 MeV. This reaction can produce fairly monoenergetic neutrons with energy from around 24 keV to 1 MeV. The emerging neutron energy (E_n) from this reaction is

$$E_n = \frac{A-1}{A} \left(E_\gamma - Q - \frac{E_\gamma^2}{1861(A-1)} \right) + \frac{E_\gamma}{A} \sqrt{\frac{2(A-1)(E_\gamma - Q)}{931A}} \cos \theta \quad (4-10)$$

where A = mass of the target nuclei = 9, E_γ = γ -ray energy in MeV, Q = threshold energy in MeV = 1.666, and θ = angle between the flight path of the γ -ray and the emitted neutron (Ref. 1). Hence, for a γ -ray with 2.088 MeV of energy,

$$E_n \text{ [keV]} = 375 + 6.6 \cos \theta. \quad (4-11)$$

The intrinsic neutron energy spread in this reaction is 2×6.6 keV, or 13.2 keV. Of course, this intrinsic energy spread does not include the effects of neutron collisions within the source, which become important factors, if the source is fairly large.

A photoneutron source can be created by surrounding an appropriate γ -ray emitting material with a mantle of beryllium or heavy water. Various γ -emitters, such as antimony (${}^{124}\text{Sb}$), yttrium (${}^{88}\text{Y}$), and radium (${}^{226}\text{Ra}$), have been utilized. These neutron sources can be switched off by removing either the photon source or the beryllium converter (Ref. 10). Another advantage of beryllium photoneutron

sources is their compactness. They could be on the order of a few centimeters.

However, photoneutron sources have two disadvantages: high-intensity γ -radiation and small specific neutron yield. In addition, the half-lives of some important γ -emitters (for example, ^{124}Sb and ^{88}Y), are short- and long-lived emitters (for example, ^{226}Ra), and are expensive.

In Figure 4.4, the strength of various sources with half-lives of at least 10 h or more and neutron intensities greater than 5×10^5 n/s/Ci have been plotted as a function of the half-life (Ref. 7). It is apparent from Figure 4.4 that ^{124}Sb and ^{88}Y are the best photoneutron sources for

Figure 4.4. Strength of intense photoneutron sources versus half-life.

ANDA applications because of their relatively high source strength, sufficiently long half-lives, and relatively high specific activities.

Photoneutron energy and intensity are important parameters in selecting photoneutron sources for safeguards applications. Most of the sources emit neutrons with energies below the ^{238}U threshold for neutron fission. For deuterium and beryllium photoneutron sources, the neutron energy (E_n) is given approximately from Eq. 4-10 by:

$$E_n = \begin{cases} \frac{1}{2}(E_\gamma - 2.226) & \text{for deuterium} \\ \frac{8}{9}(E_\gamma - 1.666) & \text{for beryllium} \end{cases} \quad (4-12)$$

where E_γ is the photon energy in MeV. The one-half factor, in the first case, appears because the proton in the deuterium carries half of the total available kinetic energy. Actual photoneutron energy spectra are complicated somewhat by kinematic effects, moderation of neutrons within the source, the presence of more than one source of γ -ray with an energy above the (γ, n) threshold, and neutrons produced by Bremsstrahlung from high-energy β -particles. The neutron energy spectrum from a relatively "clean" ^{124}Sb -Be source is shown in Figure 4.3C.

The neutron source strength is proportional to the γ -ray source strength multiplied by the (γ, n) cross section at the energy of the γ -ray. The (γ, n) cross sections for deuterium and beryllium are shown in Figure 4.5. For a small ^{124}Sb source surrounded by a thick spherical shell of

Figure 4.5. (γ, n) Cross sections for deuterium and beryllium versus γ -ray energy.

beryllium of thickness R , the neutron intensity S_n (n/s)/Ci of ^{124}Sb is approximately:

$$S(n/s \text{ Ci}) = 5 \cdot 10^6 \times R \text{ (cm)}. \quad (4-13)$$

Thus, a 2-cm thick shell of Be surrounding a small, 1 Ci ^{124}Sb source will yield roughly 10^7 n/s.

A typical ^{124}Sb -Be source is shown in Figure 4.6. The neutron energy from this source is 24 ± 2.2 keV. The source can be revived, after the initial decay (with its 60.9 day half-life), by additional irradiation in a nuclear reactor.

Figure 4.6. A typical Sb-Be source
($\sim 5 \cdot 10^6$ n/s/Ci ^{124}Sb).

The ^{88}Y -Be photoneutron source has a longer half-life (107 days) than the ^{124}Sb source, about the same neutron intensity, and a higher source neutron energy ($E_n = 200$ keV). It, thus, offers a greater penetrability which makes it especially useful in ANDA applications. However, production of ^{88}Y , which requires a high-energy, high-intensity charged particle accelerator, is expensive.

Photoneutron sources have been designed in numerous configurations to fit specific requirements. A wide pulse, or step function, of neutron intensity can be obtained if the ^{124}Sb source is ejected from the Be mantle, as is shown in Figure 4.7 (Ref. 10). The source intensity, as a function of time for this design, is shown in Figure 4.8. Within 60 to 100 ms, the source intensity is virtually zero, an important feature whenever delayed neutrons are measured as a signature in an ANDA system. The source neutron energy can also be modified or tailored for a specific

Figure 4.7. Schematic cross section of a step ^{124}Sb -Be source apparatus.

Figure 4.8. Normalized source intensity versus time for the source shown in Figure 4.7.

application by changing the Be thickness and by adding neutron reflectors and absorbers (see Chapter 6).

Characteristics of photoneutron sources with half-lives longer than 1 hr that are generated by thermal neutron capture (for example, by irradiation in reactors), as fission products, or that are naturally available (for example, Ra) are given in Table 4-3. Calculated neutron spectra of some of these sources are shown in Figure 4.9. The shorter-lived isotopes (half-life less than 30 days) have limited applicability because frequent access to a nearby reactor is required for their regeneration. Facilities with their own reactor (or with easy access to one) might benefit by using such sources in suitable ANDA systems. A full discussion of other photoneutron sources is given by Marion and Fowler (Ref. 11) and Beyster and Kull (Ref. 7).

Table 4-3. Characteristics of photoneutron sources.

Isotope	Half-life, $T_{1/2}$	γ -Ray Energy, MeV	Absolute Yield: No. γ -Rays/Disintegration	Target Material	Photoneutrons Energy E_n , keV	Photoneutrons Specific Yield (10^4 n/s. Ci-g)
^{24}Na	14.9 hr	2.750	1.00	Be	964	14
		2.750	1.00	D	262	29
^{56}Mn	2.58 hr	1.80	0.29	Be	119	
		2.10	0.15	Be	386	2.9
		2.56	0.01	Be	—	—
		2.69	0.007	Be	910	—
		2.93	0.004	Be	1124	—
		3.20	0.002	Be	—	—
		2.56	0.01	D	167	0.3
		2.69	0.007	D	233	—
		2.93	0.004	D	352	—
		3.20	0.002	D	866	—
^{72}Ga	14.2 hr	1.72	0.002	Be	—	5.9
		1.82	0.001	Be	—	—
		1.88	0.08	Be	—	—
		2.15	0.001	Be	—	—
		2.20	0.39	Be	475	—
		2.49	0.11	Be	—	—
		2.51	0.18	Be	750	—
		2.68	0.001	Be	—	—
		2.82	0.006	Be	—	—
		3.34	0.001	Be	—	—
		2.49	0.11	D	132	6.9
		2.51	0.18	D	142	—
		2.68	0.001	D	227	—
		2.82	0.006	D	297	—
		3.31	0.001	D	—	—
^{116}In	54.0 min	2.09	0.25	Be	377	0.82

continued

Table 4-3. Characteristics of photoneutron sources (continued).

Isotope	Half-life $T_{1/2}$	γ -Ray Energy, MeV	Absolute Yield: No. γ -Rays/Disintegration	Target Material	Photoneutrons Energy E_n , keV	Photoneutrons Specific Yield (10^4 n/s. Ci-g)
^{124}Sb	60.1 days	1.692	0.50	Be	23	19
		2.088	0.07	Be	375	—
$^{140}\text{Ba} + ^{140}\text{La}$	12.8 days	2.535	0.055	Be	772	0.23
		3.009	0.002	Be	1194	—
La^{140}	40.3 hr	2.535	0.055	D	154	0.97
		3.009	0.002	D	696	—
$^{144}\text{Ce} + ^{144}\text{Pr}$ (MsTh) (RdTh)	284 days	2.190	0.007	Be	466	—
	6.7 yr	1.810	0.038	Be	128	—
	1.91 yr	2.190	0.017	Be	466	—
		2.614	0.346	Be	843	3.5
		2.614	0.346	D	194	9.5
^{226}Ra (^{222}Rn)	1600 yr (3.28 days)	1.728	0.02	Be	55	—
		1.764	0.16	Be	87	—
		1.784	0.03	Be	105	—
		1.791	0.01	Be	111	—
		1.848	0.01	Be	162	—
		1.862	0.02	Be	174	—
		1.900	0.01	Be	208	—
		2.017	—	—	—	—
		2.085	0.01	Be	372	—
		2.117	0.02	Be	401	—
		2.204	0.06	Be	478	—
		2.290	0.004	Be	555	—
		2.450	0.002	Be	697	—
		2.290	0.004	D	32	—
		2.450	0.002	D	112	—

Figure 4.9. Calculated photoneutron energy spectra (integral of spectrum normalized to one).

4.5 SPONTANEOUS FISSION SOURCES

Because of their small size and high-neutron intensity, spontaneous fission sources utilizing californium-252 (^{252}Cf) have recently become the most common ANDA interrogation sources. Spontaneous fission occurs in most heavy nuclei, but usually at very slow rates. The process takes place without the application of external energy to the nucleus and is usually accompanied by other modes of decay. At present, ^{252}Cf (with a spontaneous fission half-life of 87.5 yr) provides the only usable neutron source for this process.

There are some other isotopes that undergo spontaneous fission— ^{238}Pu , ^{240}Pu , and ^{242}Pu —but they cannot be used as interrogating sources because of their low neutron emission rates. (Their spontaneous fission half-lives are of the order of 10^{10} to 10^{11} yr.) Their spontaneous prompt neutrons and γ -rays may, however, constitute an important background component for ANDA detection systems based on the coincidence method.

The main advantages of ^{252}Cf are its availability and its prolific emission of neutrons from spontaneous fission, 2.3×10^{12} n/s/g. Indeed, the emission rate is so high that only milligram or microgram quantities of ^{252}Cf are normally used in neutron sources. Hence, extremely intense, miniature neutron sources are possible with ^{252}Cf . Commercial californium neutron sources with intensities between 10^5 n/s to more than 10^{10} n/s are readily available.

Although ^{252}Cf is usually considered as a neutron source, it in fact emits other radiations in its decay process, among them: neutrons,

α -particles, and γ -rays. As shown in Figure 4.10, 97 percent of the californium atoms decay by α -emission, forming helium gas and the isotope ^{248}Cm . In a practical neutron source, the helium is contained within the capsule and does not present a problem of pressure buildup even at total decay. The *effective* half-life of ^{252}Cf , 2.64 yr, is governed by the α -decay rate of the isotope. This relatively short half-life is one of its main drawbacks, since it leads to rapid depletion of the source.

About 3 percent of californium nuclei undergo spontaneous fission, that provides neutrons having a spectrum of energies similar to the fission spectrum of ^{235}U (Figure 4.11) but with a higher average energy of about 2.35 MeV. In addition to fission fragments, approximately two photons, with the spectrums shown in Figure 4.12, are produced on the average for each neutron released by spontaneous

Figure 4.10. Decay scheme of ^{252}Cf .

Figure 4.11. Neutron energy spectrum of ^{252}Cf .

Figure 4.12. γ -Ray energy spectrum of ^{252}Cf .

fission. In a sealed source, the fission fragments are contained and only neutrons and γ -rays are emitted from the source. If a pure neutron source is desired, the γ -rays (whose average energy is about 0.9 MeV) can be attenuated by a suitable filter. Because of the relative ease with which these softer γ -rays can be absorbed in most applications, these γ -rays are less important than those produced by the interactions of californium neutrons with nearby materials.

Since its average neutron energy (2.35 MeV) is above the threshold energy for fission of ^{238}U , it is possible to use ^{252}Cf for superthreshold-MeV or for subthreshold irradiations. The amount of moderation required for subthreshold irradiation is smaller than that required for $\text{Be}(\alpha, n)$ sources because of the relatively low neutron energy from ^{252}Cf . Californium-252 cannot be used to stimulate fission inside detection systems like the Fission Multiplicity Detector (FMD), however, since the source neutrons would be indistinguishable from fission neutrons produced in the sample (see Chapter 9). In such cases, the source is used outside the system so that the detection probability for more than one ^{252}Cf prompt neutron belonging to the same fission chain is very low.

Figure 4.13 shows a cross section of a typical industrial ^{252}Cf neutron source. The inner capsule (5.5-mm diameter $\times$ 26-mm long) is contained in a second stainless steel capsule (9-mm diameter $\times$ 37-mm long). The source material is californium oxide with over 82 percent ^{252}Cf .

Figure 4.13. Industrial ^{252}Cf source.

4.6 CHARGED PARTICLE ACCELERATORS FOR NEUTRON PRODUCTION

The isotopic sources discussed in earlier sections offer the ANDA user the highest level of reliability. The reactions that produce neutrons are unaffected by the external world. These sources require no maintenance, and their intensity is either constant or very accurately known as a function of time. However, these sources have several disadvantages. The cost of obtaining very high intensities from them, if at all possible, can

be prohibitive. In addition, strong γ -radiation often accompanies neutrons generated by isotopic sources. Finally, because most isotopic sources cannot be switched off, their rapid removal is required in order to measure time-dependent signatures, like delayed neutrons.

Accelerator-based neutron sources offer the ANDA experimenter the highest neutron intensities available, which is probably the single most important feature of this type of source. In some cases, they also can provide a certain energy selectivity, notably, intense sub-MeV neutrons. However, the disadvantage of using these accelerators, especially in the industrial environment, is their relative complexity and their often uncertain reliability. Also, the cost of some types of accelerators and their associated equipment may be prohibitive. Nonetheless, charged particle accelerators played an important role in the development of ANDA, and in some cases, their incorporation in the system is essential. The generation of neutrons by charged particle accelerators—in particular, the Van de Graaff and the Cockcroft-Walton—are discussed in this section.

Van de Graaff Accelerators: This accelerator uses either the proton (p,n) or deuteron (d,n) reactions to produce neutrons. The choice of the reaction depends on the available charged particle energy and the required neutron spectrum and intensity. Both the Van de Graaff and Cockcroft-Walton accelerators endow charged particles with kinetic energy by accelerating them in a vacuum using an electrostatic potential. In the Van de Graaff machine, the high potential is produced by continuous motion of one or more insulating belts.

A uniform proton beam is obtained by introducing hydrogen into the ion source of the machine and filtering any ionized heavy molecules by a magnetic analyzer. Energy-analyzed proton beams up to 100 μ A in strength with continuously variable energy up to 4.5 MeV are available. The energy spread of the beam at 1.0 MeV at the analyzer output can be reduced to as little as 200 eV. Figure 4.14 shows a schematic diagram of a Van de Graaff generator designed to accelerate positive ions.

The most frequently used (p,n) reaction for the production of monoenergetic neutrons is the following endothermic reaction (see Section 4.2):

Data on the energy-dependent cross section for this and other reactions are given by Marion and Fowler (Ref. 11). At the threshold energy for this reaction, neutrons are emitted in the forward direction with energy of 30 keV. The neutron energy increases as the proton energy increases. At proton energies above 2.0 MeV, the ${}^7\text{Be}$ may remain in an excited state and then decays by emitting a γ -ray. A low-energy neutron group

Figure 4.14. Schematic diagram of a Van de Graaff generator operated to accelerate positive ions.

appears corresponding to this mode of reaction. As the proton energy increases to 5 MeV, the intensity of the low-energy neutron group increases to 10 percent of that of the main group of neutrons that accompanies the formation of ${}^7\text{Be}$ in its ground state.

The ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction has been used to produce relatively intense neutrons of 400 to 600 keV energy, which were used to measure small amounts of fissile material (${}^{235}\text{U}$) in the presence of larger amounts of fertile material (${}^{238}\text{U}$). Since delayed neutrons were used as the fission signature, it was necessary to pulse the interrogating neutron beam. With the Van de Graaff, pulsing can be accomplished by a combination of electrostatic deflection of the charged particle beam and a mechanical beam shutter. The on-off ratio, which is the ratio of neutron intensity during the pulse to the neutron intensity between pulses, is the parameter that determines the neutron background in such a measurement. In this manner on-off ratios greater than 10^9 can be obtained with an accelerator (Ref. 12).

There are other useful (p,n) reactions. Among them is the reaction

which makes possible the production of neutrons over a wide energy range (up to 4 MeV). Occasionally, the reactions ${}^{45}\text{Sc}(p,n){}^{45}\text{Ti}$ and ${}^{51}\text{V}(p,n){}^{51}\text{Cr}$ are used for neutron production because they produce relatively low-energy neutrons; however, their neutron yield is very small. Figure 4.15 shows neutron yields as functions of the accelerated charged particle energy for most of the reactions of interest to ANDA.

Figure 4.15. a. Neutron yield and b. neutron energy as a function of bombarding energy for several deuteron and proton induced reactions (Ref. 13).

Since Van de Graaff accelerators are complex and expensive machines, their usefulness for routine analysis in either field or industrial applications of ANDA seems to be limited to large fixed installations.

Low-Energy Cockcroft-Walton (C-W) Accelerators: Since their accelerating voltage is much lower (150-200 keV) than that of the Van de Graaff, simple voltage doubling rectifiers are used in C-W accelerators. For the same reason, these accelerators are much smaller—a few meters long and about a meter wide. They can be installed in small-fixed or mobile laboratories. A modern drift-tube type C-W accelerator is shown in Figure 4.16.

Figure 4.16. Drift tube type neutron generator accelerator assembly (Courtesy Kaman Nuclear, Division of Kaman Sciences Corp., Colorado).

The C-W's low accelerating voltage makes only two reactions useful for neutron production. The first is ${}^2\text{H}(d,n){}^3\text{He}$, or in short $d-d$, yielding a neutron energy of about 2.4 MeV; the second is ${}^3\text{H}(d,n){}^4\text{He}$, or in short $d-t$, yielding a neutron energy of about 14 MeV. At 150 kV accelerating voltage, the $d-t$ reaction furnishes intensities two orders of magnitude higher than the $d-d$ reaction. A C-W accelerator can deliver more than 10^{11} n/s per mA current of charged deuterons on a fresh tritium target. Like the Van de Graaff, the C-W can be pulsed with a high on-off ratio, which is essential for delayed neutron measurements. It can

produce pulse widths down to about $1\ \mu\text{s}$ at repetition rates up to 50 kHz.

Besides the drift tube C-W mentioned above, neutron sources can also be based on a sealed-tube scheme, which eliminates the major components found in conventional differentially pumped accelerators, such as the pump and power supply. In this scheme, neutrons are generated by the $d-t$ reaction: deuterium ions are accelerated from an ion source by an extractor to a nearby target with a tritiated coating. An accelerating potential of up to 150 kV (but usually 120 kV) is used in these sealed tubes.

During operation of the tube, the tritium in the target becomes depleted, but this shortcoming can be overcome by loading a "replenisher" with a mixture of deuterium and tritium. Tritons as well as deuterons are then accelerated into the target so that the net quantity of tritium in the target remains more or less at the same level and the neutron yield is basically constant throughout its lifetime. Typical lifetimes are a few hundred hours, depending on the level of operation. Neutron yields of up to $10^{12}\ \text{n/s}$ can be achieved; average routine yields are 10^9 to $10^{10}\ \text{n/s}$. A schematic representation of a sealed-tube neutron generator is shown in Figure 4.17.

The energy of the neutrons from these generators is about 14 MeV, which is too high for most ANDA measurements. Spectrum moderation is almost always required before exposing the sample to the interrogating neutrons.

The main problems that beset C-W accelerators stem from the short life of the tritiated target in the drift-tube machine operating at full power and the relatively short life (a few hundred hours) of the target in

Figure 4.17. Sealed tube neutron generator
(Courtesy Elliott Automation Radar
System Ltd., England).

the sealed-tube machine. But some improvements have been reported in recent years. Fourteen-megaelectronvolt neutron generators will probably be used more often in ANDA and in neutron activation analysis systems, in general, whenever very strong intensities and/or the need for pulsing are of prime importance.

4.7 ELECTRON ACCELERATOR PHOTONEUTRON SOURCES

The determination of the amount of fissile material (for example, ^{235}U , ^{239}Pu , ^{233}U) in a sample requires techniques that are almost completely insensitive to the presence of fertile materials (for example, ^{238}U or ^{232}Th). Possibly, the most intense practical source of sub-MeV neutrons is provided by photoneutron production from small, low-energy electron accelerators (Ref. 14) (see Chapter 5). With a proper target, such accelerators can provide relatively high-intensity source of neutrons with energies below the fission thresholds of the fertile materials. These neutrons are, thus, highly penetrating and induce fissions only in the fissile materials. In these machines, the accelerated electrons impinge on a target in which Bremsstrahlung radiation is produced whose maximum energy (end-point energy) is equal to the kinetic energy of the electrons. The Bremsstrahlung radiation then interacts with the target material to produce neutrons by the (γ, n) reaction.

A simple and sufficiently intense source of sub-threshold neutrons can be obtained from the $^2\text{H}(\gamma, n)^1\text{H}$ reaction using a D_2O target, shown schematically in Figure 4.18. The maximum neutron energy is determined by the amount that the end-point energy of the Bremsstrahlung exceeds 2.226 MeV, the threshold for the photodisintegration of the deuteron.

One can obtain intense sub-MeV neutron sources by irradiating a deuterium target with a Bremsstrahlung beam generated by electrons with energies from 4 to 6 MeV (see Chapter 5). Typical average

Figure 4.18. Linac sub-MeV neutron source.

intensities are 5×10^7 (at 4 MeV) to 4×10^8 (at 10 MeV) neutrons per second per microamp electron current per cm of D_2O thickness. The neutron spectra from such irradiations are shown in Figure 4.19. The fission spectrum is also plotted for comparison. The sharp high-energy cutoff and the overall soft spectrum are obvious. Figure 4.20 shows the neutron intensity for various accelerator targets as a function of the electron energy. The neutron intensity obtained from a ^{238}U target with electron energies above 7 MeV is higher than that obtained from a thin deuterium or beryllium target. Figure 4.21 is a plot of neutron yield from a ^{238}U target as a function of electron energy, and shows that the intensity grows very quickly. The neutron yields from ^{238}U are very high, much higher than those from a Van de Graaff or C-W neutron generator. However, in order to obtain these high yields, a 15 MeV or higher energy electron accelerator, usually a Linac (linear accelerator), is required. The neutron spectrum from a ^{238}U target is very similar to the fission spectrum; hence, spectrum moderation is required for its use as a source in ANDA systems. When such a machine is available, time-of-flight techniques (see Table 1-2, Chapter 1) become applicable, greatly enhancing ANDA capabilities for small samples. However, its high cost and lack of portability make the high-intensity, medium-energy Linac an

Figure 4.19. Calculated neutron spectrum from the $D(\gamma,n)$ reaction from low energy electron linear accelerator (Ref. 14). (The energy width of the electron energy is assumed to be 4%, which is easily attainable without analyzing magnets.)

Figure 4.20. Neutron yields as a function of electron energy for different converters:

- A: D_2O 1 cm thick
- B: Be 1 cm thick
- C: D_2O 10 cm thick
- D: ^{238}U 1.27 cm thick
- E: W 0.3 cm thick.

unattractive tool for ANDA unless it can be also used for other purposes. Low-energy electron accelerators of the type used for industrial radiography are common, reliable, and less expensive, however; with a properly designed deuterium target, they can provide ample neutron intensities with soft, mainly sub-MeV, spectra.

4.8 OTHER POSSIBLE NEUTRON SOURCES FOR ANDA

The previous subsections covered all the practical sources for ANDA systems. Since each ANDA system or group of systems may have special requirements which make one source "practical" and another one "impractical," the definition of practicality of sources is, of course, very loose. Nonetheless, one can broadly define conditions of practicality for

Figure 4.21. Neutron yield/electron as a function of electron energy for ^{238}U (Ref. 15).

ANDA sources. Such conditions may include reliability, complexity, size, cost, portability, adaptability to industrial environment, and others. No attempt is made here to grade these criteria of practicality. The discussion in the previous sections makes it obvious that isotopic sources qualify very well as practical sources. In the category of accelerators, the C-W and the low-energy electron accelerators can qualify, usually, with some reservations. Other sources which have been used can hardly be justified solely by ANDA requirements, but if available, they can be beneficial for ANDA. Such sources are the proton Van de Graaff or any other proton, deuteron, or α -particle accelerator, medium energy Linacs, and nuclear reactors. High-flux research reactors (peak thermal flux in the reactor is more than $5 \times 10^{12} \text{ n/cm}^2/\text{s}^2$) can provide high-thermal and epithermal fluxes and fast fission fluxes in highly collimated beams to interrogate samples. Small samples can be irradiated inside the reactor core, and their delayed fission radiations (neutrons and γ -rays) can be measured outside. Very high sensitivity and accuracy have been obtained in such measurements. Finally, research reactors and zero-power reactors can be used to accurately measure the fissile content of small samples by means of reactivity methods. For further discussion of these methods, a reactor analysis textbook should be consulted (Ref. 16).

4.9 SUMMARY AND CONCLUSIONS

The various neutron sources available to the ANDA designer and user are reviewed in this chapter. These sources include, among others, isotopic (α, n), especially $\text{Be}(\alpha, n)$ and $\text{Li}(\alpha, n)$; photoneutron sources, in particular $^{124}\text{Sb}(\gamma, n)$; ^{252}Cf spontaneous fission; charged particle accelerators; and Linac-based photoinduced neutron generators. Discussions of the various sources can be summarized by listing the main advantages and disadvantages of the most important ones.

Isotopic (α, n) sources

Advantages:

long half-life; generation of neutrons in the sub-MeV region by some sources, notably, $\text{Li}(\alpha, n)$; usually have a low γ -ray background; reasonably compact; and reasonable cost.

Disadvantages:

spectrum softening required for high-energy neutrons generated; available intensity for reasonable size sources less than 5×10^7 n/s; expensive for large sources; and $\text{Be}(\alpha, n)$ accompanied by highly penetrating, 4.43-MeV γ -rays.

Isotopic (γ, n) sources

Advantages:

generation of sub-MeV neutrons (can produce monoenergetic neutrons); delivery of high neutron intensities, when more spectrum moderation is allowed, by using thicker converters (or target material); a variety of shapes and forms to suit ANDA requirements for some sources, notably, Sb-Be ; neutron production switched on and off; regeneration, by irradiation in reactors, of Sb-Be and other isotopic sources obtained by thermal neutron capture; and possibility of very strong sources.

Disadvantages:

relatively low specific neutron yield; very strong, high-energy γ -ray background requiring heavy shielding; relatively short half-life for some important γ -ray sources (for example, ^{124}Sb , ^{88}Y); the logistics of source utilization are complicated by the need to regenerate short-lived sources; and high cost for long-lived γ -ray sources (for example, ^{226}Ra).

^{252}Cf sources

Advantages:

very intense sources readily available; compact, reasonably low cost per unit-intensity with strong sources; neutron spectrum softer than that delivered by $\text{Be}(\alpha, n)$ source; background of relatively soft γ -rays; and rapid transfer in and out, allowing measurement of delayed radiation.

Disadvantages:

relatively short half-life, which affects the cost of operation; energies of most source neutrons above the fission threshold of the fertile isotopes; and considerable γ -ray background.

*(d-t) 14-MeV neutron sources**Advantages:*

very strong and easily variable source intensities, easy on-off switching and pulsing, lowest cost per unit-intensity among practical sources for ANDA, and very low γ -ray background.

Disadvantages:

very high neutron energy (14 MeV) requiring substantial slowing-down of neutrons, limited target life when accelerator is used at full power, poorer stability and reliability compared to isotopic sources, more manpower required for operation compared with isotopic source, and more space required than for isotopic sources.

*Electron accelerator photoneutron sources**Advantages:*

very strong and easily variable intensities, energies of most neutrons below the fission threshold of the fertile isotopes, no tailoring of the neutron spectrum required, easy to gate on and off and to pulse, practically unlimited target life, and readily designed to accommodate specific ANDA requirements.

Disadvantages:

very strong (though directional) γ -radiation requiring heavy shielding, relatively large capital investment, more manpower required to operate compared with other systems, more complicated maintenance, poorer stability and reliability when compared with isotopic sources (but better as compared with commercial 14-MeV neutron generator), and more space required than for isotopic sources.

These advantages and drawbacks are also shown in Table 4-4. In this table, the various qualitative features of the sources are listed and graded qualitatively from the ANDA point of view. A plus sign is an obvious advantage; a minus sign is an obvious disadvantage; and (+,-) and (-,+) signs are less obvious positive or negative features, respectively. This table may help in selecting proper sources for ANDA or in realizing the limitations of a source already present in the system.

Table 4-4. Main features of neutron sources relevant to ANDA systems.^a

Type of Source	$T_{1/2}$	Availability of Intense Sources	γ -Ray Background	Spectrum	Physical Configuration	Regeneration Possibilities	Switching Capabilities	Portability	Reliability	Cost
(α, n) on Be, B or F	long (+)	$< 5 \times 10^7$ 10^8 (-)	low, 4.43 MeV in Be (α, n) (+)	hard $E_n \sim 5$ MeV (-)	limited usually cylindrical form (-)	not needed (+)	only by rapid removal of source or sample (-)	yes (+)	very high (+)	Reasonable for low and medium intensity (+) high for high intensities (-)
Li (α, n)	long (+)	$< 10^7$ (-)	very low (+)	soft $\bar{E} \sim 0.3$ MeV (+)	limited, as above (-)	not needed (+)	as above (-)	yes (+)	very high (+)	high (-)
^{124}Sb (γ, n)	short (60 days) (-)	$10^6 - 10^9$ (+)	very high, hard γ (-)	soft, can be mono energetic $E_n 1.24$ keV (+)	can be built in variety of shapes to conform to system requirements (+)	reactor irradiation $(-, +)^a$	by rapid removal of Sb or Be mantle $(-, +)$	can be made transportable $(-, +)$	high (+)	reasonable $(+, -)$
^{252}Cf	relatively short (2.6 yr) $(-, +)$	$< 10^{12}$ (+)	high, soft and medium γ $(-, +)$	fission spectrum $E 2$ MeV $(-, +)$	very compact size (+)	not possible (-)	by rapid shuffling of source, $(-, +)$	can be made transportable $(-, +)$	high (+)	lowest cost/ neutron among isotopic sources for high intensity (10^8) sources (+)
14 MeV neutron (D,T) generator	target limited to several hr when full beam is used (-)	$10^9 - 10^{12}$ (+)	low (+)	very hard $E_n 14$ MeV (-)	limited $(-, +)$	not possible, target has to be replaced (-)	excellent (+)	can be made transportable $(-, +)$	relatively complex system, reliability lower than above (-)	lowest cost/ neutron, relatively high capital cost $(+, -)$
Photo neutron production	unlimited (+)	$10^9 - 10^{13}$ (+)	very high (-)	soft $(+, -)$	very flexible as a volume source $(+, -)$	not needed (+)	excellent (+)	can be made transportable $(-, +)$	complex system best among accelerator, lower reliability than isotopic sources $(-, +)$	low cost/ neutron, high capital cost $(-, +)$

^aThe gradings +, -, + and - indicate respectively important advantage, mild advantage, mild disadvantage, and important disadvantage

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CHAPTER 5

GAMMA RAY PRODUCTION AND SOURCES

Active NDA systems can use, as interrogation sources, either neutrons or γ -rays in the energy range of 5 to 10 MeV. γ -Rays with energies less than 1.5 MeV can be employed for transmission measurements. The different neutron sources were discussed in the previous chapter. In this chapter, we shall briefly discuss the general requirements of γ -rays used for interrogation and describe the most important sources: namely, linear electron accelerators and microtrons. The chapter ends with a section on γ -ray sources, including the dual energy γ -ray gauge (DUEGG), used for density and composition measurements.

5.1 γ -RAY BREMSSTRAHLUNG SPECTRUM

The high-energy γ -rays (in our context “high-energy” means between 5 to 10 MeV) form the basis of one class of ANDA systems. This irradiation can cause fission in all fissile and fertile nuclei. The cross sections for this photofission process (see Chapter 3) are all of the threshold type: namely, below a certain energy (typically 6 MeV) the cross section drops very rapidly with decreasing γ -ray energy and above it (in the range of 6 to 10 MeV) it is relatively flat. Most of the differences between the various isotopes appear in the narrow energy band of the “sub-threshold” region. The photofission cross section in this region is very small. Even above the threshold region they are very small, changing from about 0.5 mb* at around 6 MeV to about 15 mb at 10 MeV (see Chapter 3). The basic features of the photofission cross section require that the γ -ray source be very intense and have a well-defined and very stable but variable energy. Monoenergetic γ -rays in the energy region of 5 to 10 MeV are available from proton interactions

*1mb = 10^{-3} barn = 10^{-27} cm²

with fluorine, namely the $^{19}\text{F}(p,\alpha\gamma)^{16}\text{O}$ reaction which yields three γ -ray energies: 6.13, 6.9, and 7.1 MeV. Other monoenergetic γ -rays are available, over wider range of energies, from thermal neutron capture in specific elements. This requires the availability of a nuclear reactor with a sizable thermal flux ($>10^{12}n/\text{cm}^2/\text{s}$).

The electron accelerators are the only practical source of intense γ -rays in the energy range of interest. Generally, they can provide a strong current, or beam, of electrons. The electron beam can be converted into high-energy photons or x-rays by allowing it to strike a converter target, usually, made of high Z elements, such as tungsten. Photons are produced when electrons are decelerated in the Coulomb (electrical) field of the target nuclei. This is the origin of the term Bremsstrahlung, namely, braking radiation, which applies to these γ -rays. The energy spectrum of Bremsstrahlung is continuous with a sharply decreasing intensity as a function of increasing photon energy, Figure 5.1. The maximum photon energy at the Bremsstrahlung end-point is equal to the electron energy, E_e . Most of the Bremsstrahlung radiation appears in the forward direction, that is, in the direction of the electron motion. The higher the electron energy gets, the narrower the angular divergence of the γ -rays becomes.

Figure 5.1. Relationship Between Bremsstrahlung Spectrum $\phi_B(E_\gamma)$, from 6 MeV Electrons, Photofission Cross Section ($\sigma_{\gamma f}$), and Fission Yield (Y_f) for ^{238}U .

When γ -rays, with a continuous energy spectrum, are used as interrogation irradiation, the measured quantity is the yield, for example, fission yield. The fission yield is equal to the integral over the entire γ -ray energy spectrum (for the specific electron energy used) of

the product of the number of photons/unit energy by the photofission cross section. To get the prompt neutron or delayed neutron yield, the fission yield is multiplied by the number of prompt and delayed neutron/fission, respectively (see Chapters 3 and 7).

The relationship between the Bremsstrahlung spectrum, the photofission cross section, $\sigma_{\gamma f}$, and the fission yield for electron energy of 6 MeV is shown in Figure 5.1. The figure shows that the contribution to the yield, from the product of such a spectrum by the fast rising $\sigma_{\gamma f}$, comes mostly from the region just below 6 MeV. The fission yield, Y_f , seems to almost overlap the cross section, at lower energies, and to deviate from it as the threshold value is approached. The curves in Figure 5.1 show why the electron energy, which defines the high-energy tip of the Bremsstrahlung spectrum, has to be very stable. Even small instabilities will introduce errors into the measured yield. The requirement for stability is less stringent once the threshold energy region is passed.

5.2 LOW-ENERGY ELECTRON LINEAR ACCELERATOR (LINAC)

In this section, we will describe the principle of operation of a low-energy electron Linac (Ref. 1, 2) as a γ -ray's source in the energy range of 5 to 10 MeV. Linacs are the most prolific sources of Bremsstrahlung γ -rays because large electron beam currents are attainable from them. So far, only Linacs have been used for safeguards applications (see Chapter 9). The principle of operation of Linacs is as follows:

When a pulse of radio frequency (rf) power is fed into a properly tuned wave guide, an electromagnetic wave will travel down the guide with a velocity determined by the guide dimensions. Electrons injected into the guide at the proper phase are carried along by the wave, much as the surfer is carried shoreward by an incoming ocean wave.

A Linac consists of a series of sections. High-frequency electromagnetic fields are applied between adjacent sections in phase (in such a way that, each time the electron crosses from one section to the next, it is accelerated by the field). The electrons then drift down the section to the next section, where the field is again in phase and this gives the electron more acceleration. The electromagnetic field is generally supplied by large, vacuum tube, rf oscillators (klystrons and magnetrons). The wave can either be of the traveling wave or standing wave type. The beam is generally pulsed. Linacs used for assay purposes have pulse widths on the order of a few microseconds at a repetition rate of approximately 100 pulses per sec (pps).

The high-energy electrons, thus generated finally impinge on a target material (for example, tantalum or tungsten) and are slowed down. As they slow down, Bremsstrahlung radiation is emitted with a continuous energy distribution whose maximum energy is equal to the electron energy. If generation of neutrons is desirable, a proper converter (for example, beryllium or heavy water, D₂O) is placed around the target (see Chapters 4 and 6).

Almost all the work using ANDA methods, based on photofission and photonuclear reactions, was done during 1967-1971 at Gulf Radiation Technology using the 17-MeV, single section Linac and a specially built 10-MeV Linac. The latter machine with all the associated ANDA equipment was installed in a trailer as a mobile laboratory named General Atomic Mobile Assay System (GAMAS) (see Chapter 9). Each Linac used an analyzing magnet to define the electron energy and to assure its stability. The general specification of the GAMAS Linac are listed in Table 5-1 (Ref. 3).

Table 5-1. GAMAS — Safeguards accelerator characteristics.

PHYSICAL:

Waveguide length: 120 cm.
 Dimensions: 90 cm wide, 180 cm high, 270 cm long.
 Weight (excluding shielding): 900 kg.
 Shielding weight: 3700 kg.

ELECTRICAL:

Electron gun: ARCO Model 5.5, gridded.
 Single cavity prebuncher: Stanford Linear Accelerator Center (SLAC).
 Traveling wave structure: SLAC, 2856 MHz.
 Radio frequency source: EEV M5028 4-MW magnetron.
 Magnetic optical system: 4 solenoids, 1 energy analysis magnet, 1 dump magnet.
 Input power requirement: 35 kW.

RADIATION OUTPUT:

Electron energy: 5-10 MeV.
 Direct beam resolution: 7%.
 Analyzed beam resolution: 2.5%.
 Peak current: 0.3 A.
 Pulse width: 1.5 μ s.
 Repetition rate: 60 or 180 Hz.
 Maximum average beam power: 800 W.

The overall mechanical configuration of the GAMAS safeguard Linac is shown in Figure 5.2. The special features of this Linac, at the time of its construction (1969-1970), were compactness, transportability, the

slanted angle of the nonanalyzed beam, and the integrated analyzing magnet and beam shielding.

Figure 5.2. Mechanical Configuration of Prototype Assay LINAC.

A commercially manufactured Linac, suitable for safeguards applications, is the Varian Linatron-1500. This machine, which is manufactured routinely for industrial radiography, has the necessary attributes for field operation. The energy stability and reproducibility of the electron energy spectrum were studied and found satisfactory (Ref. 4). Other important features are its simplicity, safety of operation, and compactness. Since it is a side-coupled standing wave Linac and uses the entire electron beam, an analyzing magnet is not required. The general characteristics of the Linatron-1500 Linac are listed in Table 5-2.

5.3 MICROTRON-CIRCULAR ELECTRON ACCELERATOR (REF. 5)

The two main drawbacks of electron Linacs as ANDA interrogation sources are the instability of the electron energy and the small duty cycle, that is, pulse width times the repetition rate (which rarely exceeds 0.1 percent). Another type of electron accelerator, the Microtron,

alleviates the first drawback and potentially alleviates the second one. The Microtrons have not received the same amount of attention in commercial* and industrial applications as have the Linacs. However, because of their excellent energy resolution and stability, their good electron current capabilities in the energy ranges of interest, and their compactness, Microtrons should be considered as a viable future interrogation source of ANDA.

Table 5-2. General specifications for the Varian Linatron-1500 standing wave, side-coupled electron linear accelerator.

Energy range:	4-10 MeV
Current availability:	Up to 100 mA (peak)
Pulse width:	3-4 μ s
Pulse repetition rate:	Continuously variable between 80 and 320 pps
Maximum average beam power:	950 W
Cooling water requirements:	15 L/min (machine on)
Electrical power requirements:	15 kW
Components:	
x-ray head:	74 cm (29 in.) height 900 kg (2000 lbs.), weight 71 cm (28 in.) width 168 cm (66 in.) depth
Modulator cabinet:	211 cm (83 in.) height 680 kg (1500 lbs.), weight 122 cm (48 in.) width 76 cm (30 in.) depth
Control console:	25 cm (10 in.) height, 27 kg (60 lbs.), weight 50 cm (20 in.) width 38 cm (15 in.) depth

The principles of the Microtron are as follows (Ref. 6):

In the Microtron, electrons are accelerated, in a homogeneous, constant magnetic field, by the electric field in a microwave resonator. The electron orbits form a family of circles which have a common tangent with the

*Only one company, Scanditronix in Sweden, is manufacturing suitable Microtrons for ANDA applications.

resonator gap (Figure 5.3). The source of electrons is an electron gun close to the resonator gap. When the gun is pulsed, a beam of electrons comes through the entrance hole of the resonator and is accelerated by the resonator gap voltage (on the order of 600 kV). Electrons leaving the resonator exit will perform a circular orbit in the magnetic field and arrive again at the resonator entrance. If the magnetic field and the microwave frequency are correctly related to each other, the electrons will pass the resonator in phase with the microwave voltage so that they gain another 600 keV. This process is repeated several times to obtain the desired final electron energy. For example, in a typical Microtron, 11 orbits are required to obtain a kinetic energy of about 6.5 MeV. The beam in the last orbit is extracted from the machine through an iron pipe, which screens the magnetic field, so that the electrons leave the field tangent to the direction of the orbit. The characteristics of a Microtron are given in Table 5-3.

Figure 5.3. Principle of the Microtron Electron Accelerator.

A short study (Ref. 7) was made with a low-power, experimental Microtron to determine its applicability to safeguard problems. The conclusion of this study was that, with proper specifications similar to those listed in Table 5-3, the Microtron could prove very useful for ANDA of nuclear materials.

Table 5-3. Characteristics of a Microtron.^a

Electron energy:	6.5 MeV (kinetic energy) for the fixed-energy-option machine 5 to 10 MeV for the variable-energy-option machine.
Average beam current:	0.2 A for a fixed-energy machine; 0.05 to 0.1 A for a variable-energy machine.
Pulse length:	Continuously variable, 0.5 to 5 μ s.
Pulse rise and decay time:	Typically 0.2 μ s.
Pulse repetition rate:	Up to 200 pps.
Energy spread:	$\pm$ 50 keV, that is, $\Delta E/E$ is less than 1%.
Maximum average beam power:	1400 W for the fixed-energy machine 500 W for the variable-energy machine.

^aMicrotron model M-7, manufactured by Instrument Scanditronix AB, Stockholm, Sweden.

5.4 OTHER HIGH-ENERGY γ -RAY SOURCES

We have mentioned that some charged particle interactions can give off a few monoenergetic γ -rays. Sources employing charged particle accelerators are very inefficient, expensive, and are impractical for ANDA applications. Van de Graaff accelerators (see Chapter 4) can also produce currents of up to a few milliamperes of electrons. Their energy resolution and stability are excellent. Thus, in principle, they can be used for ANDA applications. However, using a Van de Graaff accelerator to produce electrons with energies between 5 to 10 MeV is impractically expensive for ANDA applications when compared to using either a Linac or a Microtron.

5.5 γ -RAY ISOTOPIC SOURCES FOR TRANSMISSION MEASUREMENTS

γ -Ray sources are routinely used for measuring bulk density and are occasionally used for analysis of multicomponent systems. A large number of radioisotopes are available as sources of γ -radiation for transmission measurement. The selection of a source for a specific application depends mainly on the availability, half-life, and energy of the emitted γ -radiation. A list of the common γ -ray sources with some of their pertinent properties is given in Table 5-4.

Cesium-137 is the most commonly used source for density measurement. Its γ -rays have relatively good penetrability, and it is easy to shield personnel from them. Short-lived sources, such as ^{75}Se and ^{169}Yb , are used typically in systems involving the assay of ^{239}Pu or ^{235}U .

Selenium-75, for example, radiates 401-keV γ -rays, which are close in energy to the 414-keV γ -rays of ^{239}Pu . The transmission of γ -rays from ^{75}Se through a sample that contains ^{239}Pu can, therefore, be used to derive a correction factor to account for the attenuations of the γ -rays from ^{239}Pu in the matrix materials of the sample (Ref. 8). Similarly, transmission of γ -rays from ^{137}Cs , ^{54}Mn , and ^{203}Hg through the sample is used to determine the self-attenuation of the most important γ -radiation from ^{238}Pu , ^{238}U , and ^{237}Np , respectively. In any case, the energy of the γ -rays used in transmission measurements is usually chosen to be slightly lower than the γ -energy emitted by the assayed isotope. Possible variations of transmission correction procedures are described in Section 6.12 and (Ref. 8).

Table 5-4. γ -Ray sources useful for transmission measurements.

Nuclide	$T_{1/2}$	Important Energy (MeV) and Yield (%)
^{241}Am	432 yr	0.06 (36)
^{57}Co	270 days	0.014 (8) 0.122 (85) 0.136 (11)
^{153}Gd	242 days	0.070 (8) 0.097 (100) 0.103 (71)
^{75}Se	120 days	0.136 (96) 0.401 (20)
^{169}Yb	32 days	0.177 (58) 0.198 (100)
^{203}Hg	47 days	0.07 (13) 0.28 (81)
^{133}Ba	10 yr	0.081 (34) 0.356 (69)
^{22}Na	2.6 yr	0.511 (181) 1.28 (100)
^{137}Cs	30 yr	0.030 (7) 0.662 (86)
^{54}Mn	314 days	0.84 (100)
^{60}Co	5.26 yr	1.17 (100) 1.33 (100)
^{88}Y	106.5 days	0.90 (91.5) 1.84 (99.5)

The source intensity is generally selected to provide a usable intensity for the measurement period. If a short-lived isotope is used, position adjustment and judicious lead filtering are used to adjust the count rate to a desired level. The transmission source is typically encased in a collimator-shield to minimize personnel exposure.

Two types of measurements are commonly applied. The first is a continuous scan in which the sample moves with a constant speed past the collimator. The second method is based on segmented scans in which the sample is vertically fixed in each position and then counted. Typically, the scan of a container 25-cm high takes about 5 min. For a constant source strength, the measuring time is determined by the required precision and the sensitivity of the method used.

Density measurements by transmission of γ -ray are also used in rod scanners (see Section 9.6) and for materials accounting in general. The response of a transmission system is given by

$$R = R_o e^{-\mu \rho x} \quad (5-1)$$

where R_o is the count rate obtained with an empty container (or cladding), μ is the attenuation coefficient of the bulk material assayed, ρ is the bulk density, and x is the thickness of the sample (see Chapter 3). The measurement error is determined by the counting-statistics errors ($\sqrt{R}$) and by external effects, such as sample vibrations, nonuniformity of the container, and others. The statistical error in the measured density $\sigma(\rho)$ can be readily determined from Eq. 5-1 as

$$\sigma(\rho) = \frac{1}{\mu x \sqrt{R_o}} e^{\frac{\mu \rho x}{2}}. \quad (5-2)$$

Eq. 5-2 shows the basic relationship between the design parameters x , μ (γ -ray source energy), and R_o and the required accuracy of the system.

Analysis of a two-component mixture is possible if two γ -ray transmissions of different energies from two sources are used. Consider, for example, a sample that contains two elements in which one has a low- Z number (carbon, silicon, etc.) and the other has a high- Z number (fissile or fertile materials). The transmission of a particular radiation energy by this sample is given by the exponential relation

$$R_i = R_{io} \exp(-w_L \rho \mu_{L,i} x - w_H \rho \mu_{H,i} x); i = 1, 2 \quad (5-3)$$

where R_i = counting rate of source with energy E_i
 R_{io} = counting rate of source with no sample
 $\mu_{L,i}, \mu_{H,i}$ = attenuation coefficients (cm^2/g) of the light and heavy components, respectively, at energy E_i

w_L, w_H = weight fractions of the light and heavy components, respectively.

By solving Eq. 5-3 for the two γ -ray sources, it is possible to determine the weight fraction of the two components in the sample as follows:

$$W_L = w_L \rho x = D_1/D \quad ; \quad W_H = w_H \rho x = D_2/D \quad (5-4)$$

where

$$D_1 = \ln_1 \mu_{H,2} - \ln_2 \mu_{H,1} \quad ; \quad D_2 = \ln_2 \mu_{L,1} - \ln_1 \mu_{L,2} \quad (5-5)$$

$$D = \mu_{L,1} \mu_{H,2} - \mu_{L,2} \mu_{H,1} \quad (5-6)$$

and

$$\ln_i = \ln[R_i/R_{i0}]. \quad (5-7)$$

The condition necessary for the existence of such a solution is that $D \neq 0$, namely,

$$\frac{\mu_{H,2}}{\mu_{H,1}} \neq \frac{\mu_{L,2}}{\mu_{L,1}}. \quad (5-8)$$

It basically states that the Z dependence of the attenuation coefficients for the two selected γ -ray energies must behave differently, that is, they should be linearly independent. The sensitivity and accuracy of the measurement depends to a large extent on the choice of the two γ -ray energies and on the composition of the sample.

Error analysis of Eq. 5-4 leads to the following expressions for the total errors in the measured material composition:

$$\begin{aligned} \frac{\delta W_L}{W_L} = \frac{1}{D_1} \left\{ \mu_{H,2}^2 (\delta \ln_1)^2 + \mu_{H,1}^2 (\delta \ln_2)^2 + \mu_{H,2}^2 W_L^2 (\delta \mu_{L,1})^2 + \right. \\ \left. \mu_{H,2}^2 W_H^2 (\delta \mu_{H,1})^2 + \mu_{H,1}^2 W_L^2 (\delta \mu_{L,2})^2 + \mu_{H,1}^2 W_H^2 (\delta \mu_{H,2})^2 \right\}^{1/2} \end{aligned} \quad (5-9)$$

$$\begin{aligned} \frac{\delta W_H}{W_H} = \frac{1}{D_2} \left\{ \mu_{L,2}^2 (\delta \ln_1)^2 + \mu_{L,1}^2 (\delta \ln_2)^2 + \mu_{L,2}^2 W_L^2 (\delta \mu_{L,1})^2 + \right. \\ \left. \mu_{L,2}^2 W_H^2 (\delta \mu_{H,1})^2 + \mu_{L,1}^2 W_L^2 (\delta \mu_{L,2})^2 + \mu_{L,1}^2 W_H^2 (\delta \mu_{H,2})^2 \right\}^{1/2}. \end{aligned} \quad (5-10)$$

Generally the high- Z component is determined with greater ease and accuracy, because it dominates the absorption of the low energy γ -rays.

Because $\mu_{L,1}$ is usually larger but reasonably close to $\mu_{L,2}$, all components in Eq. 5-10 may contribute equally to the total error of the high- Z components. The error in the low- Z component is inherently higher. Since $\mu_{H,1} \gg \mu_{H,2} \gg \mu_{L,2}$ the dominant errors are associated with quantities which are related to the higher γ -energy: $\delta \ln_2$, $\delta \mu_{L,2}$, and $\delta \mu_{H,2}$. This is a gratifying situation since there are practically no limitations on the intensity of "point" γ -sources, such as ^{137}Cs or ^{60}Co , and hence one can perform very accurate calibration measurement, which determines the various μ -coefficients and their uncertainties, if good calibration standards are available.

A prototype, dual-energy, γ -gauge (DUEGG) for high-temperature gas reactor (HTGR) fuel pellets (^{238}U , ^{232}Th , and C) with different ratios of low- Z to high- Z materials is discussed in (Ref. 9). In that reference, a system is described in which the absolute amount of the heavy elements was measured in 2 min to an accuracy of 1.5 percent by using a set of attenuation coefficients which were known to an accuracy of about 2 percent. Typical pairs of sources for DUEGG to probe low- and high- Z materials are ^{241}Am and ^{137}Cs , or ^{57}Co and ^{60}Co . In a recent experiment (Ref. 10) the former source-pair was used to determine the density of B_4C powder encapsulated in stainless-steel tubes to an accuracy better than 2.5 percent.

5.6 SUMMARY AND CONCLUSIONS

The basic features of the Bremsstrahlung γ -ray spectrum are reviewed in this chapter. The high-energy tip of the spectrum is the important part in ANDA applications. Because of the fast rising photofission cross section, the stability requirement is very stringent on the high-energy tip of the spectrum and the electron energy that generates it.

Electron Linacs are widely used in the industry, especially for radiography. Versions of radiographic Linacs are suitable for ANDA applications when special care is taken to maintain the stability of the electron energy spectrum. The electron beam intensity and energy variability of Linacs are usually adequate for ANDA applications. Microtrons are very compact and inherently have a very well-defined, stable electron energy. They could be developed for ANDA applications, once there is sufficient demand.

Often, transmission measurements provide invaluable information for the proper utilization of ANDA systems and the interpretation of assay results. General considerations for selections of isotopic γ -ray sources for transmission measurements are briefly reviewed. A list of the most common (and available) sources for such measurements is also given. Applications of the DUEGG concept for low Z /high Z composition determination are discussed.

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* Available for purchase from National Technical Information Service, Springfield, Virginia 22161.

CHAPTER 6

EFFECTS OF NEUTRON AND γ -RAY TRANSPORT IN BULK MEDIA

6.1 INTRODUCTION

In ANDA, neutrons and γ -rays of various energies propagate through light, intermediate, or heavy materials. As they interact with the materials, there are continual changes in the energies of these radiations and, hence, in their interaction cross sections. These changes profoundly affect the design and performance of the assay system and its ability to detect, in an optimal way, fissionable material in the sample. Although the physical laws that characterize the specific transport process involved are well known (see Chapters 2 and 3), analysis of the overall process of radiation propagation through materials is not simple. Modeling the process requires a very large number of data entries. By processing these data and simulating the physical laws on a computer, it is often possible to reduce the complexity of radiation transport calculations to a manageable level. Often, such a computerized simulation helps in the determination of the necessary parameters to be measured, the required accuracy, and consequently, the types of sources and detectors to be used in ANDA.

Both neutron and γ -ray transport are described by the Boltzmann equation (Ref. 1), which is general and exact. This equation, however, cannot be solved rigorously except in certain special cases; therefore, computer calculations are based on a variety of approximations of this equation. These include, in increasing order of complexity: Fermi age approximations, multigroup diffusion approximations, and discrete ordinates approximations. Monte Carlo transport simulations can also be employed. Because these methods are beyond the scope of this work, they will not be discussed. Instead, various results based on both transport calculations and experiments will be presented.

The macroscopic interactions that are discussed include the following:

1. Those between primary source neutrons and moderating materials of the types that may comprise the source "tailoring" system (see Sections 6.2 through 6.6).
2. Those between source neutrons, including those moderated by the tailoring system, and the materials in the sample (see Sections 6.7 through 6.10).
3. Those between γ -rays and the materials in the sample (see Sections 6.11 and 6.12).
4. Those between neutrons and γ -rays, and the detectors (see Section 6.13).

6.2 INTERNAL AND EXTERNAL MODERATOR CONFIGURATIONS

In ANDA measurements, the primary (source) neutron energy is usually modified before irradiating the sample in order to take full advantage of the isotopic discrimination afforded by subthreshold and superthreshold irradiation. The threshold level is the fission threshold for fertile materials. Three categories of output neutron flux are commonly required:

1. Uniform thermal neutron flux over a small area for activation of small samples. A "good" thermal moderator maximizes the fractions of thermal (~ 0.025 eV) neutrons that irradiate the sample.
2. Uniform thermal and epithermal neutron flux ($1/E$ spectrum) over a large area, in fuel rod scanning devices, for example. A "good" $1/E$ moderator simultaneously maximizes both the fraction of neutrons with energies less than the ^{238}U fission thresholds (~ 1 MeV) and the fraction of neutrons with energies less than 0.1 MeV. Such a spectrum provides optimum discrimination against the fertile isotopes, for example, ^{238}U , while greatly enhancing the response of the fissile isotopes of major interest, notably ^{235}U and ^{239}Pu .
3. Higher energy, "hard," subthreshold neutron flux. A criterion for a "good" fast moderator which provides these "hard" neutrons is the maximization of the number of neutrons with energy $< ^{238}\text{U}$ threshold energy, while minimizing the number with energy < 0.1 MeV. Such a spectrum preserves neutron penetrability, although maintaining discrimination against ^{238}U .

Moderators that provide the different neutron spectra are called spectrum "tailoring" systems.

In the first and second categories listed above, irradiation is usually achieved in "internal" configurations; that is, the sample is embedded inside the moderating materials. The third type of irradiation is usually achieved in "external" configurations; that is, a sample outside the moderating assembly is irradiated by the neutrons leaking out of the tailoring assembly (leakage flux). There are, however, some cases in which relatively hard neutron spectra are obtained in internal irradiation configurations. In a few other cases, low-energy epithermal and thermal leakage fluxes are used for external irradiation. In addition, in most cases, a high-intensity neutron flux is required, therefore, the source must be as close to the sample as reasonable.

It should be realized, however, that since the signature of the nuclear materials originates in the fission events, it is more important for design purposes to calculate the fission rate distribution rather than the flux itself. This is especially true for the design of moderators for internal irradiation, such as the fuel rod scanner (see Chapter 9). In terms of fission rate, the design criteria for the latter system are:

1. Uniform fission rate, $F.R. = \int_E \sigma_f(E) (E, r) dE$.
2. Maximum fissile/fertile fission rate ratio.
3. Maximum fission rate.

6.3 EXTERNAL CONFIGURATIONS: SINGLE MODERATORS

In external configurations, a single moderator may be used, such as hydrogen-containing material (like plastic), beryllium, graphite, or a heavy element. Use of a hydrogen-containing moderator results in a sharp decrease in the intensity of the high-energy flux near the source because of the large energy loss per elastic scattering collision. Simultaneously, the relatively large thermal neutron absorption cross section of hydrogen reduces the intensity of the thermal neutron flux away from the source. Use of a heavy element, such as lead, tungsten, or uranium, results in a more uniform spatial flux with substantial energy reduction, from inelastic scattering, but little or no thermal neutron flux.

Figures 6.1 to 6.4 illustrate in more detail some of the foregoing considerations for a single hydrogen-containing moderator (Ref. 2). Figure 6.1 shows the calculated thermal neutron flux inside polyethylene spheres of different radii surrounding a point source of 14-MeV neutrons. Figure 6.2 shows a similar plot for a ^{252}Cf fission source. The thermal flux peak at the source location and its rapid decline with increasing distance from the source are obvious. The peak value of the thermal flux (which includes all neutrons below 0.4 eV), namely, 3.1×10^{-2} /unit source (Figure 6.2), is the upper limit for the thermal flux

Figure 6.1. Thermal neutron flux inside polyethylene (CH_2) spheres of various sizes resulting from 14-MeV neutron source at the center. A unit source intensity is defined as $1n/s$.

available from a fission source by moderation. The peak thermal flux from a 14-MeV source is lower by almost an order of magnitude but the spatial distribution is flatter.

The spectra of the neutrons leaking out of the moderator (and thus available for interrogation of external samples) are shown in Figures 6.3 and 6.4. The neutron spectra are plotted as $E\phi(E)$, where $\phi(E)$ is the total neutron leakage flux at energy E per source neutron per second. The function $E\phi(E)$ is called the lethargy spectrum. A plot of $E\phi(E)$ clearly shows the $1/E$ region of the energy spectrum since $E\phi(E)$ is constant over that region. This portion of the spectrum, which normally extends from about 1 eV to about 100 keV, is an important one, since the neutron absorption and fission cross sections in this energy range are often low enough to provide adequate penetrability of neutrons into the sample.

Figure 6.2. Thermal neutron flux inside polyethylene (CH_2) spheres of various sizes resulting from ^{252}Cf neutron source at the center.

Because the basic characteristics of a hydrogen-containing moderator include (1) the highest thermal flux near the source, (2) large flux gradients, and (3) relatively low fast neutron leakage, it is clear that a tailoring system made of a pure hydrogen-containing moderator is not optimal.

A single heavy moderator might be considered instead of a single light one. Some results for lead are given in Figure 6.5, where calculated neutron lethargy spectra are shown for a lead sphere (50-cm radius) at two locations: close to the source ($r=3.1$ cm), and far from the source ($r=32$ cm). Also shown are the spectra for two types of sources: a high-energy source ($E_n = 14$ MeV) and a fission source. The combination of inelastic scattering in the high-energy region (> 0.5 MeV) and low absorption cross section leads to the net effects of scattering the neutrons almost uniformly over a large volume, and of a slow and continuous reduction of energy with an increasing moderator thickness.

Figure 6.3. Neutron leakage lethargy spectrum (in units of $n/s/\text{unit source}$) from CH_2 spheres of various sizes for 14-MeV neutron source. To obtain the neutron leakage flux/unit energy, the plotted values should be divided by the "effective" group energy. The "effective" group energy is the ratio of the lethargy to energy group widths. For the 9 group calculation shown in Figures 6.3 and 6.4 the effective group energies, starting from the highest energy group are (in eV): 1.6×10^7 , 8.7×10^6 , 2.57×10^6 , 1.9×10^6 , 2.74×10^5 , 2.93×10^4 , 3.07×10^2 , 5.14 , 3.9×10^{-2} .

Figure 6.4. Neutron leakage spectrum from CH_2 spheres of various sizes for ^{252}Cf source. See also caption to Figure 6.3.

Figure 6.5. Neutron lethargy spectrum (in units of $n/\text{cm}^2 \text{ s/unit source}$) in a lead sphere of 50-cm radius for different radial positions (source at the center).

The “hardness” of the neutron spectrum is clearly a function of the position in the sphere; therefore, samples may be positioned inside it according to the desired energy of the interrogating neutrons. However, significantly thermal or very low-energy neutrons are not produced in such a moderator.

Intermediate nuclei, such as Ni or Fe, are used to reflect rather than moderate fast neutrons and thus, to increase the total intensity without softening the spectrum. This effect is caused by their high macroscopic scattering cross section, low absorption at high energies, and relatively

low energy loss/collision. Table 6-1 summarizes the interaction cross sections for neutrons between 110 and 608 keV. The "out-scattering" cross section, given in Table 6-1, is a measure of the probability of reducing the neutron energy below the lower energy boundary of the group (110 keV) and thus it depends on the neutron energy loss/collision.

In heavy and intermediate elements, multiplicative ($n,2n$) reactions also provide a mechanism for reducing the source neutron energy and increasing intensity. These reactions are especially important for 14-MeV neutrons, but they also have some effect on fission sources.

A tailoring system consisting of a single moderator is rarely optimal. Composite moderators consisting of sections of various materials can often provide better tailoring systems for ANDA.

6.4 COMPOSITE MODERATOR ASSEMBLIES FOR 14-MeV NEUTRONS

There are some assays for which the use of 14-MeV neutrons is attractive. These neutrons can be produced with high intensity from a ($d-t$) reaction (see Chapter 4). The main disadvantage of these neutrons is their high energy, which must be moderated. Although single moderators can sometimes be used, it is frequently desirable to have composite moderators to obtain the required spectrum.

The $1/E$ spectrum and the "fast moderated" spectrum are often desired. These spectra can be produced by surrounding the target of a 14-MeV ($d-t$) neutron generator with various combinations of tungsten, lead, carbon, beryllium, and polyethylene, which act as neutron moderators. Such configurations (measuring some 5 to 20 cm in annular thickness) can reduce a large fraction (up to ~80 percent) of the 14-MeV primary source neutrons to energies below the threshold value for ^{238}U fission (Ref. 3). Calculated spectral characteristics of different moderator

Table 6-1. Reflection characteristics for neutrons
in the energy range 110-608 keV.

Element	Macroscopic Cross Sections (cm^{-1})		
	Absorption	Scattering	Out-Scattering
H_2O	3.65×10^{-6}	0.557	2.71×10^{-1}
Fe	4.74×10^{-4}	0.625	3.67×10^{-3}
Ni	8.79×10^{-4}	0.686	7.45×10^{-3}
U	6.33×10^{-3}	0.403	7.53×10^{-3}

assemblies for a 14-MeV primary neutron source are summarized in Table 6-2. The median neutron energy given in Table 6-2 is the flux-weighted average. For many applications, however, a more appropriate average energy is estimated by taking into account the increase in fission rate at low energies. This is accomplished by weighting the neutron energy with the product of the flux and the fission cross section.

The notation for the configuration, in Table 6-2, can be best explained by an example: Pb/C/CH₂-12/10/2.5 means a 12-cm radius core of lead around the source surrounded by a 10-cm thick spherical shell of carbon surrounded by a 2.5-cm thick shell of polyethylene. The thin shell of polyethylene surrounding all of the $1/E$ moderators is necessary to enhance the $1/E$ tail at low energies.

For several of the moderators listed in this table, the indicated total leakage is greater than 100 percent. This apparent neutron multiplication is the result of the large ($n,2n$) cross section in lead and tungsten for 14-MeV neutrons.

It is interesting to compare the characteristics of two different $1/E$ moderators for the 14-MeV source, numbers 11 and 15 in Table 6-2, for example. Number 15 consists of readily available lead, carbon, and polyethylene. Moderator number 11 consists of tungsten, beryllium, and polyethylene. The tungsten inelastic cross section near 14 MeV is about the same as that for lead, but it does not fall off as rapidly in the region of a few MeV. Furthermore, the nuclear density of tungsten is roughly twice that of lead, and the nuclear density of beryllium is considerably higher than that for carbon. Therefore, the radius of the W/Be/CH₂ moderator is only 13 cm, as compared to a radius of 24.5 cm for the Pb/C/CH₂ combination. This impressive reduction in moderator size and weight is obtained without significantly changing the resulting neutron spectrum. In addition, the spectrum from the W/Be/CH₂ moderator (Figure 6.6) exhibits a very desirable minimum just above the ^{238}U threshold value (due to inelastic scattering in tungsten). The spectrum from the Pb/C/CH₂ composite moderator has a maximum in this energy region. These properties make the W/Be/CH₂ combination an attractive one for use with a 14-MeV source.

The calculated spectra (Ref. 4) for various $1/E$ and fast moderator configurations are given in Figures 6.6 and 6.7. An $E\phi(E)$ type plot was chosen for the $1/E$ moderators in Figure 6.6 since this shows best the relative numbers of neutrons in the low-energy region. Figure 6.7 gives the leakage spectra, $\phi(E)$ (not $E\phi(E)$), from a tungsten fast moderator assembly for both 14-MeV and fission-spectrum primary sources. It strikingly shows the desired spectrum peaking effect just below the ^{238}U threshold value.

When 14-MeV neutrons induce fission in samples containing oxygen, the contribution of delayed neutrons from the $^{17}\text{O}(n,p)^{17}\text{N}$ reaction

Table 6-2. Spectral characteristics of various moderator configurations for 14-MeV sources.

Number	Moderator Configuration Thickness (cm) ^a	Total Leakage (%)	Median Energy (MeV)	Fraction of Neutrons		Fission Ratio ^b	
				With $E_n < 0.1$ MeV (%)	With $E_n < {}^{238}\text{U}$ Threshold (%)	$\frac{{}^{235}\text{U}}{{}^{238}\text{U}}$	$\frac{{}^{237}\text{Np}}{{}^{238}\text{U}}$
1	CH ₂ -10	90	—	10	17	—	—
2	Ch ₂ -10(Cd)	85	—	—	11	3.2	2.4
3	Pb-7.5(Cd)	145	1.76	—	39	3.1	3.7
4	Pb-10(Cd)	154	1.76	—	45	3.5	3.7
5	W-7.5(Cd)	135	0.47	—	78	6.5	3.3
6	W-10	139	—	15.2	87	—	—
7	W-10 ^c	86	—	16.2	88	—	—
8	W/C-7.5(Cd)	122	0.19	—	79	13.3	2.8
9	W/C/CH ₂ -7.5/2.5(Cd)	73	0.056	—	69	50.1	2.6
10	W/C/CH ₂ -7.5/2.5	109	0.002	—	80	788	—
11	W/Be/CH ₂ -8/2.5/2.5	115	—	62	80	—	—
12	W/Be/CH ₂ -5/5/2.5 ^c	78	—	69	81	—	—
13	Pb/C/CH ₂ -7.5/7.5/2.5 (Cd)	110	1.14	—	55	20.0	3.14
14	Pb/C/CH ₂ -7.5/7.5/2.5	140	0.47	—	66	312	—
15	Pb/C/CH ₂ -12/10/2.5	142	—	62	83	—	—

^aThe moderators and detectors were covered with 0.76 mm thick Cd where noted.

^bFission ratios are for equal weights of each fissionable isotope. Np has a fission threshold at about 0.6 MeV, substantially lower than that of ²³⁸U.

^cFission neutron source was used instead of 14-MeV source.

Figure 6.6. Neutron leakage lethargy spectra (in units of $n/s/\text{unit source}$) from $1/E$ moderator assemblies

- 1) CH_2 : 10 cm, 14-MeV source
- 2) Pb/C/CH_2 : 12/10/2.5 cm, 14-MeV source
- 3) W/Be/CH_2 : 8/2.5/2.5 cm, 14-MeV source
- 4) W/Be/CH_2 : 5.0/5.0/2.5 cm, fission source.

Figure 6.7. Neutron leakage flux * spectra (in units of $n/cm^2/s/eV/unit$ source) assembly
 1) W: 10 cm, 14-MeV source
 2) W: 10 cm, fission source.

should be considered.** An important function of the spectrum-tailoring assembly is to reduce the ratio of $^{17}\text{O}/^{235}\text{U}$ delayed neutrons and, however, keep the energy spectrum hard enough to ensure large penetration and uniform response in the sample. Neutron transport calculations have been conducted (Ref. 5) to determine the degree of reduction in background radiation that might be expected from the use of a lead moderator around the neutron source. It appears from these calculations that 20 cm of lead should reduce the ^{17}O delayed neutron background by more than an order of magnitude. Another way to reduce this background is to use a lower-energy source since the threshold value of $^{17}\text{O}(n,p)^{17}\text{N}$ reaction is about 8 MeV.

*Note that leakage (or total leakage) from the Moderator assembly can be calculated by integrating the leakage flux over the surface of the assembly.

**Another weak source of delayed neutrons with a half-life of 0.176 s is ^9Li . This isotope is produced by the (n,p) reaction of high-energy neutrons ($E_n > 15$ MeV) on ^9Be .

6.5 MODERATOR ASSEMBLIES FOR ^{252}Cf NEUTRONS

Both single and composite moderators have been investigated with ^{252}Cf sources, and extensive theoretical and experimental data have been obtained. Neutron transport and Monte Carlo results are shown in Figures 6.8, 6.9, and 6.10 (Ref. 6) for a ^{252}Cf source in a 10-cm sphere of W, Be, and D_2O . Results for CH_2 spheres of different radii were already shown in Figures 6.3 and 6.4. In Figures 6.8 through 6.10, the initial fission source spectrum is also plotted (dashed line) for comparison. Tungsten, an excellent moderator for 14-MeV neutrons (Table 6-2), does not appear to be as useful with a ^{252}Cf source because the fission neutrons are rapidly reduced in energy and captured in the resonance region of the tungsten, resulting in only a small increase in the $^{235}\text{U}/^{238}\text{U}$ fission ratio.

The results for W, Be, and D_2O moderators are summarized in Table 6-3 (Ref. 3). Both beryllium and deuterium have $(n,2n)$ cross sections that multiply neutrons at the high energy. Because beryllium has a high nuclear number density ($0.1234 \times 10^{24}/\text{cm}^3$) and a relatively large fractional energy loss associated with each neutron scattering event (due to its relatively low atomic weight), even a small mass of Be moderates neutrons very effectively, without excess thermalization.

Figure 6.11 shows the thermal neutron flux as a function of radial distance from a ^{252}Cf source (Ref. 7) to demonstrate the capability of tailoring the neutron flux for a particular application in which a thermal

Figure 6.8. Neutron leakage spectrum (in units of n/s/unit source) from a tungsten alloy (containing 89 percent W, 7 percent Ni-Cu) spherical moderator (10-cm radius) surrounding a fission-spectrum neutron source. Dashed line indicates the input fission spectrum (ordinate normalized to one source neutron).

Figure 6.9. Neutron leakage spectrum (n/s/unit source) from a beryllium moderator (10-cm radius) surrounding a fission-spectrum neutron source. Dashed line indicates the input fission spectrum (ordinate normalized to one source neutron).

Figure 6.10. Neutron leakage spectrum (n/s/unit source) from a heavy water moderator (10-cm radius) surrounding a fission-spectrum neutron source. Dashed line indicates the input fission spectrum (ordinate normalized to one source neutron).

Table 6-3. Spectral characteristics of ^{252}Cf moderators.

Moderator	Total Leakage	$^{235}\text{U}/^{238}\text{U}$ Fission Ratio (including thermal)	$^{235}\text{U}/^{238}\text{U}$ Fission Ratio (epithermal)
None	100.0%	~ 4.3	~ 4.3
W ^a	81.2%	30.1	30.1
^9Be	105.6%	313.	87.2
D_2O	101.2%	98.2	57.5

^a Alloy containing 89 percent W, 7 percent Ni, and 4 percent Cu.

spectrum is desired. The source is located at the center of the composite moderator materials (CH_2 , D_2O , Be, and C) which are chosen to give high thermal flux and a maximum $^{235}\text{U}/^{238}\text{U}$ fission ratio near the outer region of the spherical moderators. Figure 6.12 plots the $^{235}\text{U}/^{238}\text{U}$ fission ratio as a function of radius for the same moderator configurations. The large $^{235}\text{U}/^{238}\text{U}$ fission ratios ($\sim 10^5$) indicate the possibility of preferentially interrogating the fissile component (^{235}U) in low-enrichment or natural uranium by using a relatively high-energy neutron source (^{252}Cf). Similarly, this high $^{235}\text{U}/^{238}\text{U}$ fission ratio allows the use of fast neutron detectors biased near the ^{238}U fission threshold value (see Chapter 8). The signal to background ratio is proportional to the $^{235}\text{U}/^{238}\text{U}$ ratio, but the signal itself is proportional to the fission rate induced in the ^{235}U sample. Such a composite assembly may therefore offer a significant improvement over a single moderator.

Table 6-4 shows the result of a series of calculations (Ref. 2) on another set of composite moderators. These configurations are typical of fuel rod scanners, which require sample irradiation close to the source to obtain a high neutron flux, but with low flux gradients to be achieved without undue reduction in that flux. The ^{252}Cf source is indicated as Q in the table. The source is 0.3 cm in radius. For each configuration, the table lists the ^{235}U fission rate per ^{235}U atom per source neutron; the ^{238}U fission rate with the same units; the $^{235}\text{U}/^{238}\text{U}$ fission ratio; and the total neutron leakage per source neutron. The fission rates are calculated at a position 5.6 cm from the center of the source, a practical distance between the source and any long sample, such as a fuel pin. The radial dependence of the ^{235}U fission rate is shown in Figure 6.13 for four of the configurations (Numbers 2, 3, 5, and 9). The effect of CH_2 near the source in quickly reducing the ^{235}U fission rate, is clearly shown in cases 2 and 3, but cases 5 and 9 indicate that either Be or Pb near the source flatten the radial dependence of the fission rate because

Figure 6.11. Calculated thermal neutron fluxes ($n/\text{cm}^2\text{s}/\text{unit source}$) versus radial position produced by a ^{252}Cf source in a composite moderator assembly with various moderators in the middle region.

of the low neutron absorption in both of these materials. Cases 2 and 3 give higher fission rates than cases 5 and 9 near the source; however, the steep gradients may render the former configurations less useful than the latter, especially for rod scanning. It is worthwhile mentioning that the lead shell surrounding the source in cases 6 through 10 reduces very effectively the γ -ray background from the ^{252}Cf source. The lead has thus a triple function: moderating fast neutrons, scattering thermal neutrons without absorption, and absorbing γ -rays. The lead that appears in all configurations following the CH_2 or B-CH_2 (borated polyethylene) shells does not affect neutron behavior. Its function is to provide shielding against the 2.2-MeV γ -rays resulting from neutron capture by hydrogen and 0.48-MeV γ -rays accompanying the (n, α) reaction in ^{10}B .

Figure 6.12. Calculated $^{235}\text{U}/^{238}\text{U}$ fission ratios (equal masses of both isotopes) versus radial position for a ^{252}Cf source in a composite moderator assembly with various moderators in the middle region.

The last column in Table 6-4 describes neutron leakage from the system. Because fast neutrons predominate, the leakage is mainly of concern because of radiation hazards. Increasing the thickness of the neutron shielding (for example, case No. 9) cuts the leakage by almost two orders of magnitude.

A comparison of Figures 6.11 and 6.12 to 6.13 shows remarkable results. Two distinct optimized configurations can be observed. The first one (Figures 6.11 and 6.12), consisting of a central region of D_2O , Be, or C, yields reasonably flat thermal flux over a large moderator volume *far away* from the source with a very high $^{235}\text{U}/^{238}\text{U}$ fission ratio. The second one (Figure 6.13), consisting of configurations 5 or 9 of Table 6-4, yields a reasonably flat and high flux over a useful volume of the moderator *near* the source with acceptable $^{235}\text{U}/^{238}\text{U}$ fission ratio.

Table 6-4. Calculated characteristics of some internal moderator configurations for ^{252}Cf source.

Number	Composite Moderator Configuration	FR(235) ^a $\times 10^{-24}$	FR(238) ^a $\times 10^{-28}$	$\frac{\text{FR}(235)}{\text{FR}(238)}$ $\times 10^3$	Neutron Leakage $\times 10^{-2}$
1	Q/CH ₂ /Pb/B-CH ₂ /Pb-0.3/75/7.5/13.5/2.125	4.0	4.2	9.4	2.3
2	Q/CH ₂ /PB/B-CH ₂ /Pb-0.3/10/7.5/11/2.75	5.3	4.2	13.0	2.2
3	Q/CH ₂ /Be/CH ₂ /Be/B-CH ₂ -0.3/5/10/5/10/7.625	3.5	5.2	6.5	1.3
4	Q/CH ₂ /Be/CH ₂ /B-CH ₂ -0.3/3/12/7/10/7.625	3.3	6.8	4.9	1.5
5	Q/Be/CH ₂ /Pb/B-CH ₂ /Pb-0.3/5/7.5/2.5/13.5/2.125	3.5	6.9	5.1	1.7
6	Q/Pb/CH ₂ /Pb/B-CH ₂ /Pb-0.3/2.5/5/5/14/2.125	2.9	6.2	4.7	2.4
7	Q/Pb/CH ₂ /Pb/B-CH ₂ /Pb-0.3/2.5/7.5/5/13.5/1.75	3.4	6.1	5.2	2.3
8	Q/Pb/CH ₂ /Pb/B-CH ₂ /Pb-0.3/5/7.5/2.5/13.5/2.125	4.1	8.7	4.8	2.2
9	Q/Pb/CH ₂ /Pb/Li-CH ₂ -0.3/5/9/7.5/38	4.5 ^b	8.7	5.2	0.04
10	Q/Pb/ZrH/Pb/B-CH ₂ /Pb-0.3/2.5/7.5/5/13.5/2.125	2.9	6.2	1.3	2.2
11	CH ₂ /Q/CH ₂ /B-CH ₂ /Pb-7.5/0.3/5/7.5/10.625	3.8	1.9	2.0	5.3

^aFR is the fission rate.^bThermal Flux, $E_n < 0.5$ eV, at this point is calculated to be $\sim 10^{-2}$ n/cm²-s/unit source.

Figure 6.13. Spatial distribution of ^{235}U fission rate in composite moderator with ^{252}Cf source (see Table 6-4 for configuration number).

The use of various materials to reflect neutrons in a fast neutron assay system based on ^{252}Cf has been investigated (Ref. 8). In this system, the neutron spectrum is adjusted to optimize neutron penetrability and the fissile/fertile fission ratio at the same time. The basic design consists of a moderating core surrounded by fast neutron reflectors and a biological shield. Some of the results obtained by neutron transport calculations for this configuration are listed in Table 6-5. As expected, the hydrogen-containing materials lower the neutron energies too much for adequate penetration. Table 6-5 shows also beryllium as an intermediate moderating material and nickel as the fast neutron reflector both giving a very useful combination. The calculated neutron spectra for a combination of this type with and without the nickel reflector are shown in

Table 6-5. Fission rate calculations for spherical ^{252}Cf moderator configurations.

Moderator Material and Shell Thickness (cm) ^a	$^{239}\text{Pu}/^{238}\text{U}$ Fission Ratio (Equal Masses)		^{239}Pu Fissions / g / Source Neutron ^b	
	100 eV	1 eV	100 eV	1 eV
W/CH ₂ /Be/Fe-1/1/6/21	87	305	3.75×10^{-5}	1.32×10^{-4}
W/CH ₂ /Be/Fe-1/2/5/21	82	314	2.87×10^{-5}	1.16×10^{-4}
W/B/Fe-1/7/21	89	288	4.07×10^{-5}	1.32×10^{-4}
W/Be/Fe-2/6/21	99	309	4.06×10^{-5}	1.27×10^{-4}
W/Be/Fe-3/5/21	107	313	3.94×10^{-5}	1.16×10^{-4}
W/Be/Fe-1/7.5/20.5	92	311	4.23×10^{-5}	1.43×10^{-4}
W/Be/Ni-1/7.5/20.5	118	410	5.42×10^{-5}	1.88×10^{-4}

^aThe designation W(3), Be(5), Fe(21) corresponds to a 3-cm radius W core surrounded by a 5-cm-thick layer of Be and 21-cm-thick layer of Fe. All assemblies are enclosed in additional biological shielding.

^bThe fissionable material is located at a radius of 10 cm for all cases. The fission rate in Pu is given in units of fissions/g/unit source.

Figure 6.14. The nickel increases the total signal rate by about 70 percent.

Neutron tailoring assemblies for ^{252}Cf are sometimes designed to produce various fast neutron spectra for the interrogation of small samples, as well as fuel pellets and rods. For some measurements, two or more different neutron energies are required to assay the fissile and fertile content of unknown samples. The moderating assembly can also be employed in these cases to change the neutron energy spectrum, as required, by varying the construction materials and configuration. Beryllium is used in spectrum-tailoring assemblies because it can reduce the energy of fission neutrons below 1 MeV without thermalizing the majority of them. Cadmium is often used to absorb thermal neutrons

Figure 6.14. Calculated neutron spectra from a ^{252}Cf source (a) with and (b) without the cylindrical nickel reflector.

around the sample, while iron and nickel are applied to reflect fast neutrons, which reduces leakage losses. An example of a spectrum-tailoring assembly that can be used to obtain more than one energy spectrum is shown in Figure 6.15 (Ref. 9). Owing to the asymmetric design of the system, the energy spectrum depends on the angle. This assembly is then capable of both subthreshold and superthreshold irradiation. The neutron spectrum is changed by rotating, relative to the sample, the californium source with its moderator. The spectra for $\theta = 0^\circ$ (hardest spectrum), $\theta = 90^\circ$, and $\theta = 180^\circ$ (softest spectrum) are shown in Figure 6.16.

Figure 6.15. Schematic diagram of a cross section of the cylindrical moderating and shielding assembly for the multi-energy californium assay system.

The beryllium cylinder containing the source and nickel segment are designed to rotate through $0^\circ < \theta < 180^\circ$.

In this view, $\theta = 180^\circ$.

Figure 6.16. Calculated average neutron spectra in the central 5 cm of the irradiation channel as a function of θ (see Figure 6.15).

6.6 MODERATOR ASSEMBLIES FOR PHOTONEUTRON SOURCES

When sub-MeV neutron sources based on electron linear accelerators (Linacs) or isotopic (γ, n) reactions are utilized (see Chapter 4), the target material can be used efficiently for spectrum tailoring. Since the neutron yield is a function of target thickness, various optimization techniques can be used to design a system which provides both maximum intensity and the desired sub-MeV energy spectrum. For example, by irradiation of a deuterium target with a Bremsstrahlung beam of 4 to 6 MeV maximum energy, one can obtain an intense sub-MeV neutron source. These neutrons are characterized by a relatively narrow energy spectrum with a sharp high-energy cut-off. However, if a relatively thick D_2O target is used to increase the neutron intensity, the moderating effect of the heavy water softens the neutron spectrum even more and substantially reduces the number of neutrons with energies above 1 MeV. This effect is shown in Figure 6.17 (Ref. 10). In a representative case with the Gulf Radiation Technology Safeguards Linac, a 15-cm thick D_2O target yielded, at an electron energy of 10 MeV, about 3×10^{11} n/s. This type of source and target thickness results in a discrimination ratio* between ^{235}U and ^{238}U of about 50 when prompt neutrons are used as signatures, and a ratio of 22 when delayed neutrons are used. In contrast, the discrimination ratio for a thin D_2O target at this electron energy is

*The discrimination ratio is defined as the ratio of the responses from equal masses of isotopes stimulated by the same irradiation.

**Figure 6.17. Neutron lethargy spectrum ($E\phi(E)$) from a $(x\gamma,n)^*$ reaction in thin and thick D_2O targets (maximum γ -ray energy: 10 MeV).
A: Thin D_2O Target, 1 in. (2.5 cm).
B: Thick D_2O Target, 15 in. (37.5 cm).**

only 8 for prompt neutrons and 3.3 for delayed neutrons. At an electron energy of 4 MeV, the $^{235}\text{U}/^{238}\text{U}$ discrimination ratio is 1644 for thick targets, 172 for thin targets of D_2O when prompt neutron signals are used, and 679 and 41, respectively, when delayed neutron signals are used.

The same considerations apply to isotopic $\text{Be}(\gamma,n)$ sources, such as Sb-Be . Because of the low (γ,n) cross section (10^{-3} barn), it is desirable to have a relatively thick mantle of Be around the Sb source. However, too thick a mantle slows down the neutrons considerably and increases the self-shielding in samples loaded with fissile materials. In addition, since most fast neutron detectors are sensitive to various degrees of γ -radiation, it is imperative to shield the source with a high- Z material, such as lead, which has a very low absorption cross section and slows

*The notation $(x\gamma,n)$ is a (γ,n) reaction with Bremsstrahlung γ -ray spectrum.

down neutrons only slightly. Thus, Be and Pb can serve as target-shielding and spectrum-tailoring materials. Calculational results of the γ -ray flux and the relative fast neutron signal (the relative prompt neutron production rate in ^{235}U samples) for a typical Sb-Be configuration are shown in Figure 6.18 (Ref. 11). Within the first 4 cm from the source center, prompt neutron production rate is strongly affected by beryllium. Near the outer boundary (beyond about 20 cm) the production rate declines strongly due to the strong neutron leakage from the lead. In the main volume of the lead, the fission rate is attenuated approximately exponentially as a function of position with an attenuation constant of about 0.13 cm^{-1} (curve A in Figure 6.18). The attenuation constant for the ^{124}Sb γ -rays in lead is much higher, about

Figure 6.18. Relative prompt neutron production (A) and γ -ray flux (B) from shielded ^{124}Sb -Be neutron source (Ref. 11).

A: Relative ^{235}U prompt neutron production rate.

B: Relative total flux of γ -radiation.

0.55 cm^{-1} . Thus, reasonably high neutron fluxes with a low γ -ray background are attainable with such configurations (see Section 4.3).

6.7 NEUTRON INTERACTIONS IN THE SAMPLE—INTRODUCTION TO MATRIX EFFECTS

Whenever ANDA techniques are applied to scrap materials and large samples, it is necessary to determine the influence of the sample matrix materials* and sample inhomogeneities on measurement. This information should make it possible to minimize the sensitivity of the assay technique to the physical form and chemical composition of the sample and to reduce the effect of the spatial distribution of materials in the sample. Generally, the matrix effects are analyzed in two steps; first, their effects on the primary radiation, and second, their effects on the stimulated prompt and delayed radiations moving toward the detector. The basic interaction processes in the sample are the same as those which take place in the nearby moderating assemblies (see Sections 6.2 through 6.5). However, while the materials and configuration of the moderating assemblies are well defined and even subject to design modifications, matrix composition and inhomogeneities may be poorly known and are often uncontrolled. Thus, it is not surprising that the interaction of the matrix materials with the incident and stimulated radiations is probably the largest source of error in ANDA techniques.

In general, neutron ANDA techniques are mostly affected by the hydrogen content of the matrix, while γ -ray measurements are especially sensitive to the presence of heavy (high Z) matrix materials. The main effect of introducing hydrogen-containing materials into the sample is to shift the energy spectrum of the interrogating neutrons downward, which, in turn, increases the rate of induced fissions. At higher hydrogen density and with large samples, the competing effect of thermal neutron absorption by hydrogen also takes place. Matrix effects are more pronounced for low-energy neutrons (for example, delayed neutrons) because of their larger hydrogen scattering cross sections ($\sim 8 \text{ b}$ for delayed neutrons and $\sim 4 \text{ b}$ for prompt neutrons). Computer simulations and sophisticated calibration techniques are usually required to obtain qualitative and semiquantitative results for these effects in practical geometries and compositions.

The problem of neutron multiplication in the sample poses additional analytical and experimental difficulties. The sample containing fissionable materials surrounded by moderating and reflecting materials

*Matrix refers to all nonfissionable materials in the sample. If these are light materials, they influence neutron interactions (chiefly scattering); if they are heavy, they affect γ -ray transmission.

constitutes a miniature multiplying assembly or (subcritical) nuclear reactor; the neutrons from fission may induce more fissions. The more fissionable material in the sample, the greater the probability of induced fissions and, therefore, the greater the enhancement of the counting rate. Various analytical and experimental techniques have been proposed (Ref. 12) to correct for this effect in highly enriched fissile samples.

6.8 MATRIX EFFECTS — RESULTS OF CALCULATIONS AND MEASUREMENTS

The delayed neutron response resulting from active interrogation of small amounts of fissile material interspersed with large amounts of various scrap materials has been simulated by using computers. These parametric computations, summarized in Figure 6.19 and later in Figure

Figure 6.19. Delayed neutron response using 14-MeV interrogating neutrons of a simulated 55-gal. scrap barrel containing 1g of ^{235}U at 15 cm radial position in representative matrix materials. The dashed lines indicate the response with no matrix materials present, ± 20 percent.

6.22, indicate that samples containing almost any common scrap material other than hydrogen-containing compounds are effectively transparent to fast neutrons, whether these are interrogating neutrons or delayed neutrons resulting from fission. If no hydrogen is present, assay of fissile materials is essentially independent of matrix composition in which the fissile material is interspersed.

The case of a hydrogen-containing matrix is, however, much more difficult. Quantitative results for the effects of hydrogen density and other matrix materials on the response of delayed neutrons were obtained experimentally (Ref. 13, 14) and analytically (Ref. 14) for samples in 1-gal. cans. The effect of the various matrix materials on the plutonium fission rate in the can was measured using two types of fission probes. A small plutonium fission chamber was placed in the moderator cavity beside the gallon can at 90° to the accelerator beam axis. In addition, a 0.0025-cm thick ^{235}U foil was wrapped around the exterior of the can to act as an in situ probe. The fission detector response indicated the change in neutron flux caused by the matrix material in the can, but the delayed neutron response from the in situ uranium reflected both the change in the interrogating neutron flux and delayed neutron absorption by the matrix material.

The different materials investigated are listed in the first column of Table 6-6. The second and third columns list the delayed neutron response from the foil probe and the plutonium fission detector response, respectively. Both of these responses have been normalized to unity for the case of any empty can. Thus, the deviation from unity represents the net effect of the added matrix material. Table 6-6 shows that errors in the data, normalized to the foil response, are roughly less by a factor of two than the errors in the data normalized to the fission chamber response. This is due to the moderation and absorption by the matrix material of the delayed neutrons from the foil. Since the larger matrix effects are caused by the hydrogen content in the samples, the first column of Table 6-6 gives also the effective hydrogen density (ρ_H) in grams of hydrogen/unit volume for some of the samples.

To observe the delayed neutron response as a function of the material's position in the can, irradiations were performed in which a thin, metallic plutonium disc (0.01-in. (0.025-cm) $\times$ 2-in. (5-cm) diameter) was moved throughout the volume of the can that was, in turn, slowly rotated about its axis of symmetry. Figure 6.20 shows a plot of the delayed neutron response, as a function of the radial position of the foil, for the different matrix materials. Very little radial variation occurred in the response for cans which did not contain hydrogen. The delayed neutron response increased for increasing amounts of hydrogen, tending to saturate in the full density case ($\rho_H = 0.11 \text{ g/cc}$).

Table 6-6. Matrix material effects on delayed neutron response of fissile material in 1-gal. recovery cans.

Matrix Material	Normalized Response	
	^{235}U Foil (in situ probe)	^{239}Pu Fission Detector
Empty can	1.00	1.00
Pb shot	1.00	1.12
Metal parts	1.04	1.11
Carbon pellets	1.15	1.32
Sand-slag-crucibles	1.23	1.41
Concrete	1.30	1.69
String-flo-filters	1.96	3.13
Poly-chips ($\rho_{\text{H}} = 0.0093 \text{ g/cc}$)	1.89	2.80
Poly-chips ($\rho_{\text{H}} = 0.027 \text{ g/cc}$)	4.08	7.75
Plastic-Benelux-leadglass	5.00	10.0
Water ($\rho_{\text{H}} = 0.11 \text{ g/cc}$)	5.12	13.6

If the matrix materials do not include hydrogen, a calibrated fission chamber placed next to the sample would indicate which response curve in Figure 6.20 should be used to determine the amount of fissionable material in the can. Hence, nonhydrogen-containing matrix errors can be reduced to around ± 10 percent; if good standards are available, assay accuracy, within a few percent, can be attained. More sophisticated calibration techniques should be used for a hydrogen-containing matrix in which response is a function of position. The response shown in Figure 6.20 can be changed slightly, if the primary neutron energy or the spectrum-tailoring assembly is modified (see Section 6.10).

The experimental results of the effect of nonfissionable matrix materials on delayed neutron assay were supplemented by a series of calculations (Ref. 15). Figure 6.21 shows the delayed neutron response/g of plutonium, as a function of the radial position inside the can, for the full range of densities of the polyethylene matrix. All results are normalized to the delayed neutron response of 1 g of plutonium placed in the void 0.5 cm from the edge of a can containing no matrix material. The calculations assumed a "flat-response" detector. The drop in the response curve for full-density ($\rho = 0.92 \text{ g/cm}^3$) polyethylene near the center of the can, Figure 6.21, is partially compensated for in the measurements shown in Figure 6.20, because the response of the neutron

Figure 6.20. Measured delayed neutron response versus radius for 1-gal. cans with Pu distributed in various matrix materials. These cans are representative of those found in some industrial nuclear processing and recovery facilities.

counter is not really flat: the counter is more efficient for the lower energy neutrons coming from the center of the can. The degree of compensation depends on sample size and hydrogen density. The response curves for CH_2 densities greater than $\rho = 0.4 \text{ g/cm}^3$ are clearly compressed. This effect does not seriously affect the assay of dilute systems, but it can cause difficulties in the assay of high plutonium concentration samples. It is important to note that the higher CH_2 densities (for example, between $\rho = 0.2$ and 0.92 g/cm^3) are not usually found in normal assay applications.

Figure 6.21. Calculated relative delayed neutron response/g versus radius in 1-gal. cans for Pu distributed uniformly in polyethylene matrices of various densities.

Substantial improvement in the delayed neutron response of the 55-gal. scrap barrel (see Figure 6.19) can be obtained by the normalization technique described. Such a normalization can also be obtained by a direct determination of the delayed neutron response of a small amount (for example, 1 g in situ probe) of each fissile species, of interest, measured directly in the neutron energy spectral environment of the actual system under interrogation. To achieve this, 1 g of material may be irradiated in the closest possible proximity (internally or peripherally) to the unknown sample that is surrounded, during

irradiation, by a hydrogen-containing reflector. The delayed neutron response is then normalized to the response of the 1 g standard. The application of this normalization to the results shown in Figure 6.19 are shown in Figure 6.22. This procedure further suppresses matrix effects, including those of the hydrogen-containing materials, but it does not by any means eliminate the hydrogen effect.

6.9 SELF-SHIELDING IN THE FISSIONABLE MATERIAL

Analysis of matrix effects should also include study of the self-shielding effect of the fissionable material. The results shown in Figure 6.20 and 6.21 are based on the assumption that the fissile content of the sample is so low that it does not significantly alter the neutron flux in the matrix material when plutonium is not present (that is, infinitely dilute plutonium). Figure 6.23 shows the results of calculations for cans containing 20 and 200 g of plutonium in a polyethylene matrix of density $\rho = 0.2 \text{ g/cm}^3$. Clearly, such masses of plutonium can no longer be considered dilute.

Figure 6.22. Normalized (to 1 g in situ probe) delayed neutron response of the scrap barrel of Figure 6.19 surrounded by a 2.5-cm polyethylene reflector.

Figure 6.23. Calculated relative delayed neutron response/g versus radius in 1-gal. cans for three masses of Pu distributed uniformly with no matrix material and in a polyethylene matrix ($\rho = 0.2$ g/cc).

Neutron absorption by plutonium causes almost no change in the shape of the response curve, but it does produce a significant change in the magnitude of the response relative to the infinitely dilute case. If, however, the delayed neutron response is normalized to the response of a plutonium fission chamber located outside, but close to, the can, the effects of the amount of plutonium on the magnitude of delayed neutron response are significantly reduced. This normalized response/g of plutonium for the center, middle, edge, and volume-average of cans containing no matrix material, and polyethylene with a density of

$\rho = 0.2 \text{ g/cm}^3$ are given in Table 6-7. For comparison, the response normalized to the response of the "empty" can is also shown. If fission chamber normalization is used, the delayed neutron response/g in the active interrogation assembly is reasonably independent of a plutonium loading of up to at least 200 g.

It should be noted that the modeling and calculation discussed above take detailed account of all major neutron interactions (fission, moderation, capture, leakage) and, thus can provide very effective guidance to the development of optimized assay systems for a wide range of sample sizes (from a few cubic centimeters to gallons), of matrix materials, and of physical and chemical forms in which assay samples occur in the nuclear industry.

6.10 MATRIX EFFECTS AND THE INTERROGATING NEUTRON SPECTRUM

All the results presented in Section 6.8 reflect the combined effects of the matrix on both the interrogating neutrons from a 14-MeV source and the emitted delayed neutrons. In this section, some data are presented which show the effects produced by several, specific matrix configurations when different interrogating spectra are used. Figure 6.24 shows the effect on the response of tailoring the source spectrum (Ref. 14) when the sample matrix is CH_2 . The untailored 14-MeV curve is identical to the CH_2 curve of Figure 6.19. The curves for the $1/E$ -tailored and fast-tailored spectra indicate the increased sensitivity obtainable for dilute CH_2 matrices. For dense CH_2 matrices, the response is governed by the matrix rather than the source-tailoring system, and differences between the curves are small.

Table 6-8 shows similar results for a 14-MeV source and two tailored spectra with average neutron energies of 1.8 and 0.1 MeV, respectively, for a variety of sample matrices (Ref. 16). The fission rate in ^{235}U is strongly enhanced by tailoring the source spectrum when the sample matrix is a strong moderator, but it is essentially unchanged when the sample matrix is nonmoderating. For the ^{238}U fission rate, a moderating matrix reduces response.

Figure 6.25 shows the measured effect of surrounding a small sample of ^{235}U (5g) with various thicknesses of polyethylene (Ref. 17). A partially moderated ^{252}Cf interrogation source was used in these measurements. The fission rate was measured, in this case, by the fast coincidence method based on prompt γ -rays (see Chapters 8 and 9). Since the sample was small, it was practically transparent to all neutrons. The degradation of the interrogating neutron spectrum, which increases as a function of the polyethylene thickness (up to a certain limit, which

Table 6-7. Delayed neutron response/g of plutonium (relative to response of dilute distribution).

Matrix	Pu Loading (g)	Relative Delayed Neutron Response			Fission Counter Normalization			
		Center	Middle	Edge	Center	Middle	Edge	Vol Av
CH ₂	20	0.88	0.87	0.86	1.07	1.06	1.05	1.06
($\rho = 0.2$)	200	0.39	0.39	0.40	1.11	1.13	1.16	1.15
None	20	0.98	0.98	0.98	0.99	0.99	1.00	1.00
("empty" can)	200	0.88	0.89	0.91	0.96	0.97	1.00	0.98

Figure 6.24. Comparison of delayed neutron response from 1 g of ^{235}U placed at $r = 15$ cm in a simulated scrap barrel containing CH_2 for three-neutron source spectra (14 MeV, "Fast," and "1/E").

was not reached in these experiments), resulted in a large increase in fission rate. The increase was slightly tempered by reduction in the efficiency of detecting fast neutrons as a result of their slowing-down in the polyethylene surrounding the uranium.

As a final illustration of response increase due to degradation of the neutron spectrum, Figure 6.26 shows the delayed neutron response of U_3O_8 samples as a function of the ^{235}U enrichment for various interrogating spectra (Ref. 18). Curve (a) corresponds to 14-MeV neutrons and curve (b) is the hard spectrum obtained with a Pb-W moderator. The linear behavior of curve (b) indicates that there is little neutron self-absorption, even in the highly enriched samples. For curve (c), the tailoring moderator consists of concentric layers of tungsten and

Table 6-8. Matrix material effects on delayed neutron response.

Interrogation Sample ^a	Normalized Delayed Neutron Response		
	D-T Source ($E_n = 14.9$ MeV)	Pb Moderator ($\bar{E}_n \sim 1.8$ MeV)	W, Fe Moderator ($E_n \sim 0.1$ MeV) ^b
²³⁵ U foil	1.00	1.00	1.00
²³⁵ U foil + (calcined residue)	0.98	0.96	1.03
" + Al ₂ O ₃	0.95	0.97	1.05
" + ZrO ₂	0.96	0.94	1.03
" + C	0.97	0.99	1.06
" + Pb	1.03	0.97	1.00
" + CH ₂ bottle (1/3 L)	1.06	1.11	1.36
" + NH ₄ NO ₃	1.13	1.76	4.00
" + H ₂ O	2.41	5.15	9.86
²³⁸ U foil	1.00	1.00	1.00
²³⁸ U foil + (calcined residue)	0.92	0.92	0.94
" + Al ₂ O ₃	0.92	0.95	0.94
" + ZrO ₂	0.94	0.96	0.97
" + C	0.94	0.93	0.93
" + Pb	0.93	0.97	0.96
" + CH ₂ bottle (1/3 L)	0.98	0.99	0.99
" + NH ₄ NO ₃	0.87	0.89	0.84
" + H ₂ O	0.77	0.76	0.66

^aVolume of added matrix material ~ 300 cm³.^b $\bar{E}_n$ corresponds to the median neutron energy.

Figure 6.25. Enhancement of fission response of a small ^{235}U sample by moderation of high-energy neutrons in polyethylene.

carbon. At high enrichments, this response curve is slightly nonlinear because there is some neutron absorption in the resonance region. Curve (d) was obtained by adding a CH_2 reflector to the W-C configuration of curve (c), which resulted in many low-energy neutrons, strongly self-absorbed at high enrichments. Note that the effect of decreasing the average energy of the interrogating spectrum in this configuration is to increase the response, but at some sacrifice in linearity.

6.11 γ -RAY TRANSPORT IN THE SAMPLE

The sample matrix can also affect γ -ray interrogations, such as those using electron accelerators. At higher energies (>5 MeV) of interrogating radiation, the only important interaction processes are pair production and Compton scattering (see Chapter 3). Pair production simply removes γ -rays and produces two secondary, 0.511 MeV, γ -rays. Compton scattering lowers the energy and changes the direction of γ -rays. Since the interrogating photons* are in a beam and the photofission cross section drops sharply at the threshold value, pair production and Compton scattering effectively *remove* photons from the interrogating

*Photons and gamma rays are the same, and the terms are used interchangeably in this text.

Figure 6.26. Delayed neutron response from cylindrical samples of U_3O_8 for different enrichments of ^{235}U (total mass of U held constant). Curve (a) corresponds to a 14-MeV neutron irradiation, and curves (b), (c), and (d) correspond to irradiations using moderated spectra with successively decreasing average energies. (The broken lines indicate linear response curves.)

beam. The effect on the matrix is simply to reduce the intensity of the interrogating beam without changing its spectrum. This is the main advantage of using γ -rays over neutron-active interrogation, where the spectrum is changed and along with it the system response.

Figure 6.27 (Ref. 17) shows a comparison of assays of samples consisting of 1-gal. jars weighing about 4 kg and filled with highly enriched ^{235}U , wet SiO_2 , and chemical residue. The samples were assayed both by detecting the 185-keV γ -rays from ^{235}U and by active interrogation using moderated ^{252}Cf neutrons and Linac photons. Photon interrogation led to a linear response (Figure 6.27, curve 1) since the sample and its matrix were essentially transparent to photons. Both the passive γ -ray response (185 keV from ^{235}U) and the active neutron response were strongly nonlinear. In the first case, because of self-absorption of the 185-keV γ -rays by uranium and the matrix material (sand), and in the second case, because of the strong moderation of the source neutrons by the water present. The low-energy neutrons were strongly self-absorbed in samples containing larger amounts of uranium.

The (γ, n) reactions in some isotopes with low neutron-binding energies, such as ^9Be , ^{13}C , and especially ^2D , constitute an important perturbation to techniques based on photonuclear interactions. This is particularly true when the matrix contains large amounts of hydrogen-containing materials because of the presence of 0.015% of deuterium.

Figure 6.27. Results of nondestructive assays with three different techniques. (1): Photon-induced reactions. (2): Neutron-induced interactions. (3): Passive detection of 185-keV γ -rays.

However, since the threshold value for this reaction in deuterium (2.23 MeV) is well below the photofission threshold levels, one can identify and isolate this background and subtract it.

6.12 TRANSMISSION OF γ -RAYS IN BULK MEDIA

In the previous section, the effect of the sample matrix on the interrogating γ -radiation was discussed. Correcting for self-attenuation of the signature γ -rays as they leave the sample is another central problem in γ -ray analysis of bulk samples. This attenuation problem is most important in the analysis of fissionable materials; because of their high atomic numbers, they severely attenuate γ -rays (see Chapter 3). In fact, γ -ray assay for high density plutonium and uranium is often impossible because the required corrections cannot be made accurately. Nevertheless, there are many cases and classes of materials for which a careful γ -ray assay is the most precise, most accurate, and cheapest method for determining the amount of uranium and plutonium.

The most effective way to account for self-attenuation in the sample is to calculate or measure the proper correction factor. The correction factor may be defined in several ways. The most useful form is the correction factor with respect to a nonattenuating sample. Symbolically, the definition may be expressed as

$$C(0) = CF \times C(\mu) \quad (6-1)$$

where $C(0)$ is the count that would have been obtained from the sample with no attenuation and $C(\mu)$ is the count obtained in an actual measurement. The correction factor (CF) is computed from the sample's μ and the geometry of the assay. With this definition, the corrected count rate/g of material assayed is constant; that is, the calibration curve is linear in terms of mass and corrected count rate.

Another useful quantity is the correction factor with respect to a nonattenuated point source, (CF_p). Symbolically, this is expressed as

$$C_p(0) = CF_p \times C(\mu) \quad (6-2)$$

where $C_p(0)$ is the count that would have been obtained if the sample had been condensed to a nonattenuated point at the center of the cylinder and $C(\mu)$ is the actual count from the cylindrical sample, precisely as in the previous case. This form of the correction factor is particularly useful in the assay of a large sample (such as a 55-gal. drum) with respect to a small standard sample placed in the center of the drum. If the sample attenuation coefficient and the appropriate dimensions and distances are known, the two forms of the correction factor are about equally difficult to compute. The problem is even more complicated, if

the sample is nonhomogeneous and, if the effect of matrix materials on the primary radiation [for example, by (n, γ) reactions] is taken into account, that is, if the γ -ray sources are not uniformly distributed in the sample. Price (Ref. 18) and Fitzgerald (Ref. 19) summarize results for CF for many geometries of practical interest. Parker and Reilly (Ref. 20) review the methods used to determine correction factors in uniform cylindrical samples. Although their discussion is directed toward passive NDA methods, it can be applied to active methods as well. They also present numerical results for cylindrical samples far from the detector.

Because of the lack of simple and exact analytical forms for the correction factor, much use has been made of approximate forms, perhaps the most useful of which is

$$CF = k \ln(T)/(T^k - 1) \quad (6-3)$$

where T is the sample transmission, namely $e^{-\mu x}$ and k is a constant equaling ~ 0.8 .

In the near field (that is, when the sample is close to the detector), the correction factors become functions not only of sample transmission but also of the sample radius, height, and distance from the detector. A simple two-dimensional model that demonstrates these dependencies is sufficiently accurate for some practical work. The model assumes a circular sample of radius R and zero height whose center is at a distance D from a point detector that is in the plane of the sample. In this model, the CF is a function of T and the ratio D/R only, which means, for example, that given the same value of T , a sample of 1-cm radius 10 cm from the detector has the same CF as a 10-cm radius sample 1 m from the detector. Figure 6.28 gives the CF as a function of D/R for various values of T . These and all other near-field results were obtained by computerized numerical integrations. Figure 6.28 shows that, essentially, the CF decreases as D/R decreases and the more drastic changes occur for small values of T . This behavior is qualitatively consistent with the inverse square law.

When possible, a far-field assay geometry is preferable because of the less stringent requirements for positioning of the sample and the smaller dependency of the CF on the exact height and diameter of the sample. Unfortunately, count rate considerations usually force the use of a near-field situation. In any case, but especially in near-field situations ($D/R < 10$), it is best to use the CF values computed for that combination of D/R and T rather than using Eq. 6-3. However, obtaining these values is often difficult because of the lack of a simple analytic form for $CF(T)$. For dense samples of heavy elements, the transmission factors may be so small that γ -ray assay is not practical.

Figure 6.28. Near-field correction factors for a cylinder as a function of D/R for various transmission values. D is the distance of the detector to the center of the sample. R is the radius of the sample.

6.13 INTERACTION OF RADIATION WITH THE DETECTION SYSTEM

Neutrons and γ -rays can only be detected indirectly: neutrons by using nuclear reactions and γ -rays by atomic reactions that produce charged secondary particles. In the following section, some overall macroscopic considerations, mainly relevant to neutron-based ANDA systems, are presented.

ANDA techniques involve detection of prompt and delayed neutrons that could be moderated to epithermal or thermal energies. Two different kinds of measurements are commonly applied: the first kind involves measurement of neutron intensity* and (roughly) neutron energy by using threshold detectors (for example, fast scintillators, ^4He or H proportional counters, or threshold fission chambers); in the second

*Strictly speaking, neutron detectors generally do not measure neutron intensity (that is, energy and space integrated flux over the detector volume) but a similar integral of the product of the detector response by the neutron flux. This is the meaning of neutron intensity measurements as used in the text.

method, only the intensity is measured. Prompt fast neutron measurement is possible if the background due to the tailored source neutrons and γ -ray field is below the energy bias of the detection system. However, the measurement is sensitive to the rate of those neutron interactions in the sample that might reduce the neutron energy below the detection energy bias. Since neutrons emitted from the sample outer surface are more likely to reach the detector without further interaction with the sample, fast neutron measurements give higher weight to the sample peripheral region than to its core. The effect is more pronounced for large samples having a hydrogen-containing matrix material and for low-energy delayed neutrons.

Neutron intensity measurements can be carried out using BF_3 or ^3He proportional counters, which have very high efficiency for detection of low-energy and thermalized neutrons. However, the efficiency of ordinary BF_3 detectors drops by a factor of 20 when the neutron energy is changed from 0.0253 eV (thermal neutron) to 10 eV. Because the neutrons emitted from the center of hydrogen-containing samples are slowed down on their way to the detector, they have a higher probability of detection. Using an unmoderated BF_3 or ^3He proportional counter, therefore, overestimates the importance of the sample's core as compared to the periphery, unless some moderating materials are interposed between sample and detectors and around the detectors. If the density of the hydrogen-containing material is high and the sample size is large enough, the neutrons generated deep inside the sample may be absorbed by hydrogen. Thus, the sensitivity of the external detector, to fissile material in the core of the sample, is again reduced.

To detect fast neutrons with a BF_3 or ^3He counter, the neutrons must first be moderated, as is done with a "long counter" (see Chapter 8). This detector is a standard arrangement of a BF_3 or ^3He counter in a paraffin moderator. Because this counter detects fast neutrons with an efficiency nearly independent of neutron energy over a wide range of energies, it reduces, to some extent, the spatial dependence of the measurement. However, distinguishing between source and fission neutrons is impossible after moderation of the source neutrons. The method is very powerful for delayed neutron measurements whereby stimulated delayed neutrons are measured after the source is turned off. The delayed neutrons can thereby be distinguished from the interrogating neutrons. The design of the counter moderation assembly is similar to that of the spectrum-tailoring system (see Section 6.2). Design considerations and discussions of various detector systems for ANDA are presented in Chapter 8.

6.14 SUMMARY AND CONCLUSION

Neutron and γ -ray interactions with materials constituting or surrounding the source, the sample, and the detector affect the ability of an ANDA system to determine the fissile and fertile components of the sample. For neutron-based systems, desirable spectral characteristics of the interrogating neutrons can be achieved by surrounding the source with single or composite spectrum-tailoring media. Hydrogen-containing materials moderate rapidly fast neutrons, and also absorb thermal neutrons relatively strongly; therefore, they are most useful when high thermal neutron fluxes are desired close to the source. Nonhydrogen-containing, low absorbing materials, such as D_2O , Be, C, W, and Pb, moderate much more slowly, but result in a flatter spatial dependence. They are often useful for large volume samples where good penetrability is desired. The heavy moderators, for example, lead and tungsten, are especially useful in that they also provide a compact γ -ray shield for the source.

The presence of matrix materials in the sample affects penetrability of the interrogating radiation, the fission response, and the transmission of the signature radiation. Light materials, especially hydrogen, affect fast neutrons by moderation. Heavy materials affect mainly γ -rays through absorption processes. Often, these effects can be corrected for empirically by applying calculated correction factors or by carrying out measurements on similar samples of known composition.

The response of neutron detectors is also affected by nearby materials, especially moderators. For efficient, energy-insensitive fast neutron detection, "long counters," or other configurations of thermal neutron detectors embedded in moderator blocks can be used.

Tables 6-9 and 6-10 summarize qualitatively characteristics of moderating and matrix materials and their respective effects on ANDA. Some idea of the quantitative effects can be gained from Figures 6.19 or 6.24. Delayed neutron response can vary by two orders of magnitude depending on the (perhaps unknown) matrix composition and density. These uncertainties can be considerably reduced in some cases, however, by normalizing to the response of a small sample, as is shown in Figure 6.22.

In any ANDA application, all of these effects should be considered in the design and use of the system.

Table 6-9. Characteristics of basic neutron moderating assemblies.^a

Entity	Single Moderators			Composite Moderators	
	Hydrogen-containing (<i>H</i>)	Low <i>Z</i> (for example, Be,C)	High <i>Z</i> (for example, Pb,W)	High <i>Z/H</i>	High/Low <i>Z/H</i>
Neutron spectra	Very soft	Soft, sub-MeV	Hard, sub-MeV	Soft, sub-MeV	Soft, sub-MeV
Penetrability	Limited	Good	Very good	Good	Fair
Sensitivity	Very high	Fair	Low	High	High
Peak thermal flux	High	Low	Very low	Medium	Medium
Flux gradients	High	Low	Low	Low	Medium
Leakage	Low	Fairly high	High	Low to fair	Low
Fissile/Fertile Fission ratio	Very high	Medium	Low	High	Very high
Special features	Very compact; inexpensive	(<i>n,2n</i>) in Be contributes to source intensity, especially for 14-MeV neutrons	(<i>n,2n</i>) contributes to source intensity, especially for 14- MeV neutrons; provides good shielding to source γ -rays	Low flux gradients in high <i>Z</i> region; provides good shielding to source γ -rays	Low flux gradients in low <i>Z</i> region

^a*Z* represents the atomic number of the element; *H* represents hydrogen-containing moderators.

Table 6-10. Sensitivity of neutron assay results to various matrix materials.^a

Spectrum	Matrix Materials			Self Absorption in Fissile Material
	Hydrogen- containing	Low Atomic Number (<i>Z</i>)	High Atomic Number (<i>Z</i>)	
Hard	Very sensitive	Fairly insensitive	Fairly insensitive	Very Low
Epithermal	Very sensitive	Sensitive	Insensitive	Low
Thermal	Fairly insensitive	Insensitive	Insensitive for nonabsorbing high <i>Z</i> elements (for example, Pb); sensitivity to absorbing elements (for example, W)	High

^aSee Figures 6.19 to 6.26 and Tables 6-6 to 6-8.

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CHAPTER 7

SIGNATURES OF NEUTRON- AND PHOTON-INDUCED FISSIONS

7.1 INTRODUCTION

In ANDA, discrimination between the various fissile and fertile isotopes, which may be present in the sample, is made possible by the fact that each isotope emits a characteristic set of radiations after fissioning. These penetrating radiations, which consist of prompt neutrons, prompt γ -rays, delayed neutrons, and delayed γ -rays, constitute the "signature" of the isotope. Figure 2.6 shows the time scale on which these prompt and delayed radiations are emitted: prompt neutrons within the 10^{-15} s after fission, prompt γ -rays within 10^{-15} s,* delayed neutrons within approximately 1 min, and delayed γ -rays, most of which are also emitted within the first minute or so. In addition, x-rays may be given off (both prompt and delayed), and β -ray emission will result from fission products decay. These latter radiations are not usually used as signatures because they are easily absorbed by the sample.

The different properties of the signature radiations vary not only from fissionable isotope to isotope, but also depend on the energy of the radiation inducing the fissions and on the mechanism causing the fissions (that is, neutron-induced, photon-induced, or spontaneous). In this chapter, we review the signatures encountered in ANDA systems and their use in distinguishing among the various fissionable isotopes.

*In most ANDA systems all γ -rays emitted within about 10^{-8} s can be considered as prompt radiations.

7.2 PROMPT NEUTRONS

Prompt neutrons can play an important role in ANDA when it is relatively easy to distinguish them from source neutrons. This distinction can be made by the energy differential between source and induced neutrons and is applicable in the following cases:

1. When the source neutrons are created at low energies such as with the (γ, n) sources (Sb-Be) or with low-energy (α, n) sources ($^{241}\text{Am-Li}$) (see Chapter 4).
2. When moderation of neutrons from high-energy neutron sources can provide the required energy differential (see Chapter 6).
3. When photon-induced fission is used and there are, of course, no source neutrons [except those from the background (γ, n) reactions].

Distinction between source and prompt fission neutrons can also be made on the basis of the high multiplicity of fission neutrons. Prompt neutrons from spontaneous fission, which have properties somewhat similar to those from induced fission, often constitute an important part of the background in ANDA systems. In this section, the number of prompt neutrons emitted and the energy spectra of these prompt neutrons are discussed for neutron-induced, photon-induced, and spontaneous fission.

7.2.1 Prompt Neutron Yields and Multiplicities

Most fissions are accompanied by emission of prompt neutrons. In any given fission, the number of prompt neutrons emitted is between 0 and about 8 or 9. The *average* number of prompt neutrons per fission, $\bar{\nu}_p$, has a characteristic value which is determined by the fissioning isotope and the type and energy of the fission-inducing radiation.

In general, $\bar{\nu}_p$ lies between 2 and 3 for thermal neutron-induced fission and between 4 and 5 for 14-MeV neutron-induced fission. For a given isotope, $\bar{\nu}_p$ increases almost linearly with the energy of the incident neutron. For fissile isotopes, $\bar{\nu}_p$, as a function of the incident neutron energy (E_n), can be well represented by the formula:

$$\bar{\nu}_p(E_n) = \bar{\nu}_p(\text{thermal}) + aE_n \quad (7-1)$$

where a is a constant whose value is approximately 0.14 MeV^{-1} and E_n is the incident neutron energy in MeV.

Table 7-1A gives values of $\bar{\nu}_p$ (thermal), a , and $\bar{\nu}_p$ (14 MeV) for the fissile isotopes (Ref. 1).

For fertile isotopes, an equation of the same form as Eq. 7-1 can be used to obtain $\bar{\nu}_p(E_n)$ from the threshold energy ($\sim 1 \text{ MeV}$) to 14 MeV. These relations are shown in Table 7-1B.

Table 7-1. Prompt neutrons from various fission reactions.

A. Average number of prompt neutron/fission for fissile isotope.

Isotope	$\bar{\nu}_p$ (thermal)	$a(\text{MeV}^{-1})$	$\bar{\nu}_p$ (14 MeV)
^{233}U	2.480 ± 0.007	0.123	4.208 ± 0.020
^{235}U	2.407 ± 0.007	0.141	4.387 ± 0.008
^{239}U	2.874 ± 0.009	0.148	4.942 ± 0.012
^{241}Pu	2.924 ± 0.012	0.143	4.92 ± 0.15

B. Average number of prompt neutron/fission for fertile isotopes.

^{232}Th :	$\bar{\nu}_p(E_n) = 1.852 + 0.1513 E_n^a$
^{238}U :	$\bar{\nu}_p(E_n) = 2.640 + 0.1279 E_n^a$
^{240}Pu :	$\bar{\nu}_p(E_n) = 2.861 + 0.1467 E_n^a$

^a $E_n \leq 15 \text{ MeV}$.

C. Average number of prompt neutrons/fission and fission rate in spontaneous fission.

Isotope	$\bar{\nu}_{SF}^a$	$SF/\text{g}\cdot\text{s}$
^{238}U	1.95	5.64×10^{-3}
^{238}Pu	2.26	1.1×10^3
^{240}Pu	2.17	4.71×10^2
^{241}Pu	2.16	8.0×10^2
^{252}Cf	3.8	6.16×10^{11}

^a SF stands for spontaneous fission.

To get the average value of $\bar{\nu}_p$ for a spectrum of interrogating neutrons, for example, if the interrogation source is a “fast moderated” ^{252}Cf source, one must calculate:

$$\bar{\nu}_p \text{ (source spectrum)} = \frac{\int \bar{\nu}_p(E_n) \Sigma_f(E_n) \phi(E_n) dE_n}{\int \Sigma_f(E_n) \phi(E_n) dE_n}. \tag{7-2}$$

In other words, one must weigh the fission rate at each incident energy E_n , $\Sigma_f(E_n) \phi(E_n)$, by the value of $\bar{\nu}_p$ at that E_n . This requires that the neutron spectrum and macroscopic fission cross sections be known. Usually, it is sufficient to use $\bar{\nu}_p(\bar{E}_n)$, where $\bar{E}_n$ is a characteristic average interrogating neutron energy.

The average $\bar{\nu}_p$ from spontaneous fission (SF), which may constitute a background in $\bar{A}NDA$ systems, is given in Table 7-1C, along with the spontaneous fission rate. The prompt neutron emission rate is the product of these two, if there are no other neutron emitting processes present, such as (α, n) reactions.

Because of the small differences between the $\bar{\nu}_p$ of different isotopes and the uncertainty introduced by the interrogating neutron spectrum, the $\bar{\nu}_p$ by itself is, usually, *not used* as a signature. The fact that $\bar{\nu}_p$ is larger than 2, however, makes it possible to use the coincidence techniques described in Chapters 8 and 9.

Note that $\bar{\nu}_{SF}$ is lower than that from neutron-induced fission by about 20 to 30 percent for all but one isotope. On the other hand, the $\bar{\nu}_{SF}$ of ^{252}Cf is substantially higher than $\bar{\nu}_p$ from neutron-induced fission, typically by about 60 percent. Hence care should be taken when a ^{252}Cf source is used for calibration of ANDA systems which are based on prompt neutron multiplicities.

The difference between $\bar{\nu}_{SF}$ of ^{240}Pu ($\bar{\nu}_{SF} = 2.17$) and ν_p from induced fission of ^{239}Pu [for example, $\bar{\nu}_p$ (thermal) = 2.87] can enable one to determine the contribution of neutron multiplication to the total signal in the sample of neutrons originating from the spontaneous fission of ^{240}Pu . This is important in passive coincidence measurements of samples with a high concentration of fissile plutonium.

The distribution function $P(\nu_p)$, known as the multiplicity distribution, is shown in Figure 7.1 and is closely approximated by a normal (Gaussian) curve (Ref. 2, 3). The function $P(\nu_p)$ represents the fraction of fissions (in a given isotope under a given interrogation condition) in which *exactly* ν_p prompt neutrons are emitted. From $P(\nu_p)$, we can obtain certain averages, for example,

$$\bar{\nu}_p = \sum_{\nu_p=0}^9 \nu_p P(\nu_p) \quad (7-3)$$

$$\overline{\nu_p(\nu_p-1)} = \sum_{\nu_p=0}^9 \nu_p(\nu_p-1)P(\nu_p) \quad (7-4)$$

and others. These averages, or moments, are useful in the interpretation of ANDA coincidence measurements (see Chapter 8). The curve $P(\nu_p)$ shown in Figure 7.1 is a "universal" curve since it is given in terms of $\nu - \bar{\nu}_p$. If $\bar{\nu}_p$ for any isotope is known, say from Eq. 7-1, then $P(\nu_p)$ can be obtained from Figure 7.1.

7.2.2 Prompt Neutron Energy Spectra

The energy spectra of the prompt neutrons are similar for all fissionable materials and can be expressed by the formula:

$$N(E) = Ae^{-aE} \sinh \sqrt{bE} \quad (7-5)$$

Figure 7.1. Prompt neutron multiplicity distribution. Neutron emission probability P_ν versus $(\nu - \bar{\nu})$ for $^{235}\text{U}(n_{\text{thermal}}, f)$. The curve is a plot of Terrell's universal Gaussian, with width $\sigma = 1.08$.

where E is the neutron energy in MeV, and A , a , and B are fitted constants and $N(E)dE$ is the number of prompt neutrons per fission emitted with energies between E and $E+dE$. The constant a is approximately 1 MeV^{-1} and b is approximately 2 MeV^{-1} . The values of a and b differ from isotope to isotope and depend slightly on the incident neutron energy. The constant A is chosen so that the integral of $N(E)dE$ is equal to $\bar{\nu}_p$. For the thermal neutron fission of ^{235}U , $A = 0.484$. Eq. 7-5 is known as the Watt spectrum. Other representations of the fission spectrum are also used. Figure 7.2 (Ref. 4) shows a comparison of the measured and calculated prompt neutron spectrum for 0.53 MeV neutron-induced fission of ^{235}U . The spectra are plotted as $N(E)/\sqrt{E}$.

From the Watt spectrum, it is possible to obtain the average energy of the prompt neutron. The value is approximately 2 MeV, but the exact value depends on parameters a and b . This high energy makes it possible to distinguish these neutrons from moderated source neutrons by employing a fast neutron detector biased at sufficiently high energy. This

Figure 7.2. Fission neutron spectrum from ^{235}U obtained at 0.53-MeV incident energy. The line and the dashed curve are least squares fits to the experimental points assuming energy distributions according to Maxwell and Watt, respectively. The former is given by $N(E)/\sqrt{E} = \text{Exp}(-E/1.44)$ and the latter by Eq. 7-5.

can be done in ^4He gas counter or proton recoil counter. The small differences between the prompt neutron spectra do not, however, make them useful as signatures to distinguish between different isotopes.

7.2.3 Prompt Neutrons from Photofission

The yields and energy spectra of the prompt fission neutrons resulting from photon-induced fission are not nearly as well known as those from neutron-induced or spontaneous fission (as in the case of ^{252}Cf). For Bremsstrahlung irradiation from Linacs, the prompt neutron yield is a combination of fission neutrons plus photoneutrons from (γ, n) reactions in the fissionable isotopes. It is difficult to separate these, unless the Bremsstrahlung energy is below the (γ, n) threshold value or sophisticated γ -compensated coincidence circuits are employed. Figure 7.3 shows the overall neutron yield (prompt and delayed)/unit γ -ray dose in several isotopes as a function of the end-point energy of the incident γ -radiation (which is equal to the electron energy) (Ref. 5). Figure 7.4 gives the variation of the prompt neutron yield *per fission* as a function of the end-point energy (Ref. 6). In both figures, photoneutrons are included whenever the electron energies exceed the (γ, n) threshold levels (6.34 MeV for ^{232}Th , 6.04 MeV for ^{238}U , and 5.24 MeV for ^{235}U).

The prompt neutron energy spectrum is a combination of the fission neutron (Watt) spectrum plus the spectrum of the (γ, n) neutrons, and therefore, it depends on the end-point energy. Not much is known about the overall prompt neutron spectrum. However, for all practical purposes it can be assumed that the spectrum is very similar to the neutron fission spectrum.

As in neutron-induced fission, the prompt fission neutrons in photofission can be used to detect fission events by employing a Bremsstrahlung energy below the (γ, n) threshold value. Since there are no source neutrons (except those arising from (γ, n) reactions in nearby materials), the prompt fission neutrons can be detected rather easily.

The absolute yields and their variations with the Bremsstrahlung energy below the (γ, n) thresholds (Figure 7.3) can be used as signatures. Figures 7.5 and 7.6 show some discrimination ratios (ratios of prompt neutron yields) as functions of Bremsstrahlung end-point energy (Ref. 5, 6). At low energies, large discrimination ratios are obtained between ^{235}U and ^{232}Th , ^{235}U and ^{238}U , and even between the fissile isotopes ^{235}U and ^{239}U . However, because of low energies, absolute yields are low, and unless high power low-energy electron accelerators are employed, the method may be difficult to use in practical ANDA situations (see Chapter 5).

Figure 7.3. Prompt and delayed neutron yields in fissile and fertile material produced by Bremsstrahlung from electrons of energy E_e .

7.3 DELAYED NEUTRONS

Delayed neutrons from fission are the basis of several ANDA methods. Their chief advantage over prompt neutrons is that their detection can be accomplished at some time after fission has occurred. Hence, time discrimination between source neutrons and delayed neutrons can be used when a pulsed source is employed. In addition, their absolute yields and their yields as functions of time vary significantly from fissionable isotope to isotope, and thus they constitute attractive signatures. The delayed neutron yields are very small, however. We will now examine their properties in more detail.

Figure 7.4. Ratio of prompt neutron to fission yields for photon-induced reactions in fissionable materials (Ref. 6).

7.3.1 Delayed Neutron Yields and Their Time-Dependence

Delayed neutrons are emitted from radioactive decay processes in many fission products. When a fission product is in a certain unstable state, it may preferentially de-excite by emitting a neutron rather than a γ -ray. Neutron emission takes place immediately after the unstable nucleus is created, as a result of β -decay of its precursor, in the fission product chain. Therefore, the “half-life” of delayed neutrons from that particular decay product appears to be the same as the precursor β -decay half-life.

Figure 7.5. Prompt neutron discrimination ratios of ^{238}U and ^{239}Pu relative to ^{235}U as a function of electron energy.

An example of a delayed neutron transition is shown in Figure 7.7. ^{87}Br β -decays with a half-life of 54.4 s. Some of the transitions are to excited states of ^{87}Kr , which then de-excites by neutron emission, rather than γ -ray emission, and forms ^{86}Kr . The delayed neutron half-life from this transition appears to be 54.4 s.

The overall total yield of delayed neutrons is small, ranging from approximately 0.007/fission in ^{239}Pu to about 0.05/fast fission in ^{238}U or ^{232}Th . Although there are more than 50 known precursors, the half-lives of many of these are close to one another. Therefore, it is

Figure 7.6. Prompt neutron discrimination ratios of ^{232}Th relative to ^{235}U and ^{238}U as a function of electron energy.

conventional to lump the delayed neutrons together into six groups. The yields and half-lives of these six groups are shown in Table 7-2A and B for various fissioning isotopes (Ref. 7). The absolute group yields are obtained by multiplying the total yields by the group fractional yields.

As can be seen from the tables, there are substantial variations in group yields from isotope to isotope, and, therefore, both the total yields and the overall time dependence of delayed neutron emission vary considerably. Thus, either of these parameters or a combination of both can be used directly as signatures for individual fissile isotopes. Furthermore, since there are very few nonfission reactions which produce delayed neutrons, their presence is an almost certain indicator that fission has taken place. The only reactions that can seriously interfere, with the use of delayed neutrons as signatures, are the (γ, n)

Figure 7.7 Decay scheme of a delayed neutron emitter.

reactions in beryllium and deuterium that are present in or near the sample, and which can be induced by high-energy delayed γ -rays. Any reaction that results in ^{17}N [for example, $^{17}\text{O}(n,p)^{17}\text{N}$] may cause interference, since ^{17}N also decays by emission of β -particles with a 4-s half-life, followed by instantaneous delayed neutron emission.

Figure 7.8 shows the delayed neutron yield in ^{233}U , ^{235}U , ^{238}U , and ^{239}Pu as functions of the energy of the incident neutron (Ref. 7). At high incident neutron energies, the yield is seen to fall dramatically. The yield at 14-MeV incident energy is about half its value at lower energies. This behavior is true of other fissionable isotopes as well.

Delayed neutron signatures allow discrimination between different isotopes by the delayed neutron yield and the difference among various groups. The latter affects the shape of the decay curve of delayed neutron. Thus, a "shape discrimination ratio," S_{dsh} , between two isotopes, 1 and 2, can be defined (Ref. 8) as:

$$S_{\text{dsh}}(1,2) = \frac{D_1(t, t_o)/D_1(0, t_o)}{D_2(t, t_o)/D_2(0, t_o)} \quad (7-6)$$

where $D_i(t, t_o)$ is the delayed neutron yield from isotope i at time t following an irradiation of length t_o . Table 7-3 shows some values of this parameter for an incident fission neutron energy spectrum for the isotopes ^{235}U , ^{238}U , and ^{239}Pu .

To be useful in ANDA, S_{dsh} must be as different from unity as possible. Table 7-3 indicates that delayed neutron signatures of this type cannot be used efficiently to distinguish between ^{235}U and ^{239}Pu , but they are useful for the pairs ^{238}U - ^{239}Pu and ^{238}U - ^{235}U . These differences supplement the relatively large differences between the delayed neutron yields (β) of the various isotopes (Table 7-2A), for example, $\beta(^{239}\text{Pu})/\beta(^{235}\text{U}) = 0.386$, and $\beta(^{238}\text{U})/\beta(^{235}\text{U}) = 2.656$.

Table 7-2. Delayed neutron parameters.

A. Recommended absolute delayed neutron yields.

Isotope	Combined ^a	Fast	Thermal	Isotope	Combined ^a	Fast	Thermal
²³² Th	0.0545 ±0.0011 ±2.1%	0.0545 ±0.0012 ±2.2%		²³⁸ Pu	0.00456 ±0.00012 ±1.8%	0.00456 ±0.00013 ±2.0%	0.00024 3.9%
²³³ U	0.00698 ±0.00013 ±1.9%	0.00729 ±0.00019 ±2.7%	0.00664 ±0.00018 ±2.8%	²³⁹ Pu	0.00655 ±0.00012 ±1.8%	0.00664 ±0.00013 ±2.0%	0.00624 3.9%
²³⁴ U	0.0106 ±0.0012 ±11.3%	0.0106 ±0.0012 ±11.3%		²⁴⁰ Pu	0.0096 ±0.0011 ±11.5%	0.0096 ±0.0011 ±11.5%	
²³⁵ U	0.01697 ±0.00020 ±1.2%	0.01714 ±0.00022 ±1.3%	0.01654 ±0.00042 ±2.5%	²⁴¹ Pu	0.0160 ±0.0016 ±10.0%	0.0163 ±0.0016 ±10.0%	0.0156 ±0.0016 ±10.0%
²³⁶ U	0.0231 ±0.0026 ±11.3%	0.0231 ±0.0026 ±11.3%		²⁴² Pu	0.0228 ±0.0025 ±11.0%	0.0228 ±0.0025 ±11.0%	
²³⁸ U	0.04508 ±0.00060 ±1.3%	0.04510 ±0.00061 ±1.3%					

^aRecommended value based on combined processing of fast and thermal data (Ref. 7).

Table 7-2. Delayed neutron parameters (continued).

B. Recommended group parameters for delayed neutrons^a

Group <i>i</i>		Fractional Group Yield <i>a</i>	Decay Constant λ (sec ⁻¹)	Group		Fractional Group Yield <i>a</i>	Decay Constant λ (sec ⁻¹)
²³² Th	1	0.034 ± 0.003	0.0124 ± 0.0003	²³⁹ U	1	0.038 ± 0.004	0.0129 ± 0.0003
	2	0.150 ± 0.007	0.0334 ± 0.0016		2	0.280 ± 0.006	0.0311 ± 0.0007
	3	0.155 ± 0.031	0.121 ± 0.007		3	0.216 ± 0.027	0.134 ± 0.004
	4	0.446 ± 0.022	0.321 ± 0.016		4	0.328 ± 0.015	0.331 ± 0.018
	5	0.172 ± 0.019	1.21 ± 0.13		5	0.103 ± 0.013	1.26 ± 0.17
	6	0.043 ± 0.009	3.29 ± 0.44		6	0.035 ± 0.007	3.21 ± 0.38
²³³ U	1	0.086 ± 0.004	0.0126 ± 0.0006	²⁴⁰ Pu	1	0.028 ± 0.004	0.0129 ± 0.0006
	2	0.274 ± 0.007	0.0334 ± 0.0021		2	0.273 ± 0.006	0.0313 ± 0.0007
	3	0.227 ± 0.052	0.131 ± 0.007		3	0.192 ± 0.078	0.135 ± 0.016
	4	0.317 ± 0.016	0.302 ± 0.036		4	0.350 ± 0.030	0.333 ± 0.046
	5	0.073 ± 0.021	1.27 ± 0.39		5	0.128 ± 0.027	1.36 ± 0.30
	6	0.023 ± 0.010	3.13 ± 1.00		6	0.029 ± 0.009	4.04 ± 1.16
²³⁵ U	1	0.038 ± 0.004	0.0127 ± 0.0003	²⁴¹ Pu	1	0.010 ± 0.003	0.0128 ± 0.0002
	2	0.213 ± 0.007	0.0317 ± 0.0012		2	0.229 ± 0.006	0.0299 ± 0.0006
	3	0.188 ± 0.024	0.115 ± 0.004		3	0.173 ± 0.025	0.124 ± 0.013
	4	0.407 ± 0.010	0.311 ± 0.012		4	0.390 ± 0.050	0.352 ± 0.018
	5	0.128 ± 0.012	1.40 ± 0.12		5	0.182 ± 0.019	1.61 ± 0.15
	6	0.026 ± 0.004	3.87 ± 0.55		6	0.016 ± 0.005	3.47 ± 1.7
²³⁸ U	1	0.013 ± 0.001	0.0132 ± 0.0004	²⁴² Pu	1	0.004 ± 0.001	0.0128 ± 0.0003
	2	0.137 ± 0.003	0.0321 ± 0.0009		2	0.195 ± 0.032	0.0314 ± 0.0013
	3	0.162 ± 0.030	0.139 ± 0.007		3	0.161 ± 0.048	0.128 ± 0.009
	4	0.388 ± 0.018	0.358 ± 0.021		4	0.412 ± 0.153	0.325 ± 0.020
	5	0.225 ± 0.019	1.41 ± 0.10		5	0.218 ± 0.087	1.35 ± 0.09
	6	0.075 ± 0.007	4.02 ± 0.32		6	0.010 ± 0.003	3.70 ± 0.44

^a(Ref. 7).

Figure 7.8. Measured values of delayed neutron yield for:

- A: ^{233}U (n,f)
- B: ^{235}U (n,f)
- C: ^{238}U (n,f)
- D: ^{239}Pu (n,f)

7.3.2 Delayed Neutron Time-Dependence and Yields from Photofission

The delayed neutron yield from photofission shows a very steep increase for a slight increase in the electron energy (E_e), similar to the prompt neutron yield (see Figure 7.3). This yield varies greatly among the various fissionable isotopes revealing the same trend as with neutron fission, namely, delayed neutron yields decrease in the order $^{232}\text{Th} > ^{238}\text{U} > ^{235}\text{U} > ^{239}\text{Pu}$. The absolute magnitude of the delayed neutron yields for low E_e is very low, however. Unless powerful, low-energy electron accelerators are employed, the delayed neutrons are, usually, used as signature for $E_e > 7 \text{ MeV}$, especially for ^{235}U and ^{239}Pu .

Some ratios of delayed neutron yields versus E_e are shown in Figure 7.9 (Ref. 5). These curves indicate the degree of discrimination between individual isotopes allowed by the delayed neutron yields.

Table 7-3. Shape discrimination ratios.

$S_{\text{dsh}}(238,235) = \frac{D_{238}(t,t_o)/D_{238}(0,t_o)}{D_{235}(t,t_o)/D_{235}(0,t_o)}$					$S_{\text{dsh}}(239,235) = \frac{D_{239}(t,t_o)/D_{239}(0,t_o)}{D_{235}(t,t_o)/D_{235}(0,t_o)}$			
t sec	$t_o=10^{-3}$ sec	$t_o=10^{-1}$ sec	$t_o=10^1$ sec	$t_o=10^2$ sec	$t_o=10^{-3}$ sec	$t_o=10^{-1}$ sec	$t_o=10^1$ sec	$t_o=10^2$ sec
1.0	0.68	0.70	0.82	0.82	1.04	1.04	1.02	1.03
4.2	0.48	0.49	0.66	0.67	1.08	1.07	1.07	1.09
10.5	0.39	0.41	0.58	0.61	0.16	1.15	1.15	1.16
20.5	0.34	0.35	0.52	0.58	0.26	1.26	1.22	1.21
52.5	0.33	0.33	0.51	0.55	0.40	1.39	1.33	1.25
100.5	0.30	0.30	0.46	0.48	0.39	1.39	1.32	1.20
204.5	0.21	0.22	0.33	0.35	0.21	1.32	1.14	1.04
300.5	0.18	0.18	0.28	0.30	0.09	1.09	1.04	0.96

Figure 7.9. Delayed neutron discrimination ratios of ^{232}Th , ^{235}U , ^{238}U , and ^{239}Pu . (Vertical error bars indicate measurement accuracy.)

A very powerful signature by fissionable isotopes is shown in Figure 7.10 (Ref. 5) which gives ratios of the prompt to delayed neutron yields as functions of E_e . These constitute large, usable discrimination ratios for various isotopes. Experimentally, this signature has some special merits. The same neutron detector measures both the prompt and delayed neutron under very similar conditions. Hence, problems with instrumental instabilities are alleviated. Also, matrix effects are partially canceled, as well as other possible effects which influence both prompt

Figure 7.10. Ratio of prompt to delayed neutron yields as a function of electron energy.

and delayed neutrons (for example, multiplication and photon energy fluctuations).

The time dependencies of delayed neutrons following photofission in ^{235}U and ^{238}U for 10-MeV Bremsstrahlung irradiation are shown in Figure 7.11 (Ref. 9). The time dependence of delayed neutrons resulting from photofission is very similar to that resulting from neutron-induced fission, except for slight differences at early times following short

Figure 7.11. Delayed neutron decay from ^{238}U and ^{235}U following irradiation with Bremsstrahlung from 10-MeV electrons.

irradiations. Table 7-4 shows the relative (fractional) delayed neutron group yields in ^{235}U and ^{238}U at incident photon energies of 8 and 10 MeV (Ref. 9).

Because of the similarity of the decay curves resulting from photofission to those resulting from neutron-induced fission, the shape discrimination ratios will be similar to those listed in Table 7-3.

7.3.3 Delayed Neutron Energy Spectra

In contrast to prompt neutrons, which have an average energy of 2 MeV and a smooth characteristic Watt spectrum, delayed neutrons have a lower average energy (on the order of 0.4 to 0.5 MeV), and highly structured spectra (especially at lower energies). Figures 7.12 and 7.13 show delayed neutron spectra evaluated from the thermal fission of ^{235}U and the fast fission of ^{238}U (Ref. 10). Spectra for other isotopes

Table 7-4. Relative abundance of delayed neutrons from photofission at E_e of 8 and 10 MeV.

Approximate Half-Life (sec)	Relative Abundance %			
	^{238}U		^{235}U	
	8	10 MeV	8	10 MeV
55	2.5 ± 0.1	2.1 ± 0.1	5.2 ± 0.5	5.6 ± 0.2
20	17.3 ± 0.4	16.3 ± 0.1	25.3 ± 0.3	24.5 ± 0.5
5	17.0 ± 1.1	16.7 ± 0.3	21.3 ± 1.3	24.4 ± 0.9
0.6	17.9 ± 1.1	18.1 ± 1.0	4.4 ± 2.2	9.1 ± 1.0
0.2	9.2 ± 1.4	10.5 ± 1.1	8.8 ± 3.2	2.8 ± 1.1

Figure 7.12. Equilibrium delayed neutron spectrum from thermal neutron fission of ^{238}U (Ref. 10).

Figure 7.13. Equilibrium delayed neutron spectrum from fast-neutron fission of ^{235}U (Ref. 10).

have the same general appearance. Table 7-5 gives the mean energies of delayed neutron spectra.

Table 7-5. Mean energies of delayed neutron spectra (keV).^a

Isotope	Eccleston	Keepin	Fieg	Sloan	Saphier	Batchelor	Evans
²³³ U	350	390	—	—	—	—	—
²³⁹ Pu	369	400	425	—	—	—	509
²³⁵ U	385	430	435	339	471	430	457
²³² Th	355	490	445	—	—	—	—
²³⁸ U	347	490	—	—	—	—	542

^aFor references on the various evaluations and experiments, see (Ref. 10).

The energy spectra of delayed neutrons do not serve usefully as signatures. However, they are important in the design of the system, especially with regard to the detector characteristics and possible matrix effects.

7.4 PROMPT γ -RAYS

γ -Rays associated with fission are emitted continually after fission. Roughly speaking, truly "prompt" γ -rays are emitted within 10^{-15} sec as a result of energy redistribution accompanying fission. As the highly excited fission fragments de-excite, γ -rays are emitted up to a few microseconds after fission, as a result of isomeric transitions.* At longer times, the β -decay of the fragments continues to result in associated γ -ray emission. Since these processes merge into each other, there is no clear separation between prompt and delayed γ -rays. We arbitrarily define "prompt" to include γ -rays emitted up to ~ 10 ns after fission since experimental devices, which detect these γ -rays, have time resolutions of this order.

The total prompt γ -ray energy released per fission is on the order of 6.5 to 7.0 MeV. Neglecting γ -rays and x-rays below 0.14 MeV, the average γ -ray energy is about 0.97 and 0.94 MeV for thermal neutron fission of ²³⁵U and ²³⁹Pu, respectively, and 0.88 MeV for spontaneous fission of ²⁵²Cf. Therefore, about 6 to 8 prompt γ -rays are released/fission. The prompt γ -ray energy spectra for these fissioning nuclides, obtained with NaI detectors, are shown in Figure 7.14 (Ref. 11). Table

*Isomeric transitions represent de-excitation of a nucleus from an excited state to its ground state by the emission of a γ -ray. These transitions usually have very short half-lives ($\ll 1$ s), though occasionally the half-lives are considerably longer (meta-stable transitions).

Figure 7.14. Prompt γ -ray energy spectra for the thermal neutron fission of ^{235}U and ^{239}Pu and for the spontaneous fission of ^{252}Cf (Ref. 11).

7-6 gives the number of prompt γ -rays/fission and the prompt γ -ray energy/fission for several γ -ray energies (Ref. 11). The differences between isotopes are probably too small to make these spectra or multiplicities directly useful as signatures; but, with neutron-induced fission, the high multiplicities can be advantageously used in coincidence methods. When photofission is employed, the intense γ -ray burst from the source completely “swamps” the prompt fission γ -rays, thus making the coincidence circuit inoperable.

Little is known about the multiplicity distribution function of prompt γ -rays, $P(\gamma_p)$. The available evidence indicates that it is probably Gaussian, like the corresponding prompt neutron function, $P(\nu_p)$. A slight correlation between the prompt γ -ray multiplicities and their energy spectra has been observed (Ref. 12). This correlation does not affect ANDA measurement techniques based on the fission multiplicity concept, unless absolute calibration based on computation, rather than comparison to known standards, is attempted.

In addition to prompt fission γ -rays, prompt capture γ -rays will also be emitted as a result of neutron capture (n, γ) reactions when a neutron

Table 7-6. Energy emitted/fission in the form of prompt γ -rays and the number of γ -rays/fission for various γ -ray energy intervals.

Photon Energy Range (MeV)	Protons/Fission			MeV/Fission		
	^{235}U	^{239}Pu	^{252}Cf	^{235}U	^{239}Pu	^{252}Cf
0.14-0.3	0.833	1.267	1.270	0.181 $\pm$ 0.005	0.275 $\pm$ 0.007	0.282 $\pm$ 0.007
0.3-0.5	1.318	1.473	1.855	0.532 $\pm$ 0.013	0.596 $\pm$ 0.015	0.759 $\pm$ 0.019
0.5-0.7	1.182	1.219	1.613	0.709 $\pm$ 0.028	0.737 $\pm$ 0.019	0.963 $\pm$ 0.024
0.7-1.0	1.191	1.097	1.067	1.021 $\pm$ 0.026	0.939 $\pm$ 0.023	0.908 $\pm$ 0.023
1.0-1.5	1.072	0.998	0.838	1.337 $\pm$ 0.034	1.241 $\pm$ 0.031	1.044 $\pm$ 0.026
1.5-2.0	0.461	0.450	0.442	0.804 $\pm$ 0.020	0.787 $\pm$ 0.020	0.778 $\pm$ 0.020
2.0-2.5	0.258	0.268	0.292	0.582 $\pm$ 0.015	0.606 $\pm$ 0.015	0.660 $\pm$ 0.017
2.5-3.0	0.158	0.175	0.183	0.434 $\pm$ 0.012	0.482 $\pm$ 0.014	0.503 $\pm$ 0.013
3.0-4.0	0.143	0.173	0.160	0.490 $\pm$ 0.015	0.597 $\pm$ 0.017	0.550 $\pm$ 0.014
4.0-5.0	0.050	0.0714	0.058	0.220 $\pm$ 0.015	0.318 $\pm$ 0.019	0.254 $\pm$ 0.010
5.0-6.0	0.021	0.0265	0.019	0.116 $\pm$ 0.011	0.145 $\pm$ 0.018	0.102 $\pm$ 0.009
6.0-7.0	0.0098	0.0104	0.0057	0.064 $\pm$ 0.015	0.067 $\pm$ 0.012	0.038 $\pm$ 0.005
7.0-10.0	0.0027	0.0015	0.0004	0.019 $\pm$ 0.015	0.019 $\pm$ 0.012	0.0019 $\pm$ 0.0014
0.14-10.0	6.70	7.23	7.80	6.51	6.81	6.84

source is used. These capture γ -rays have a total energy equal to the neutron binding energy (approximately 6 to 8 MeV). The spectra of prompt capture γ -rays vary considerably from one element to another. Sometimes one and sometimes a few high-energy γ -rays are emitted whose total energy is equal to the binding energy. In other cases, a cascade of several low-energy γ -rays is emitted whose total energy is then equal to the binding energy. The capture γ -rays can be both advantageous and deleterious to ANDA. If they result from capture by fissionable isotopes of interest, they enhance the prompt fission γ -rays as part of their signature. On the other hand, if they result from capture by other materials in the sample matrix or nearby materials, they form an undesirable source of background, especially for coincidence methods. Even if a single high-energy capture γ -ray is emitted, it can undergo successive Compton scatterings in the detectors of a coincidence system and, thus cause spurious coincidence counts (see Chapter 8). Hence, one should be aware of highly absorptive elements contained in the sample matrix, such as chlorine, and select other measurement techniques, or take some measures to reduce the background due to the (n,γ) captures.

7.5 DELAYED γ -RAYS

Delayed γ -rays which are emitted within seconds and minutes after fission are very important signatures in ANDA systems. These are used, in rod scanners, for example, to determine total fissile content and pellet to pellet variations in fissile content (see Chapter 9). The delay times between irradiation and emission of delayed γ -rays are on the order of several seconds. The total delayed γ -ray energy/fission is roughly the same as the total prompt γ -ray energy, about 6 to 8 MeV (within 25 percent for all fissioning isotopes of interest). The average delayed γ -ray energy is also about the same as for the prompt γ -rays, about 1 MeV. Thus, approximately six to eight delayed γ -rays are emitted/fission, most within the first minute. This is in contrast to the small number of delayed neutrons (~ 0.007 - 0.05 /fission) emitted mostly within the much shorter time of 10 to 20 s.

As has been pointed out in previous sections, γ -rays are emitted continuously after fission. γ -Rays that are emitted within a short time, up to about 1 μ s, arise mainly from isomeric transitions in the fission products. The spectra of these delayed γ -rays are complicated and different from the other "slow" delayed γ -rays. These γ -rays are of some interest because they can interfere with prompt neutron detection or contribute to prompt γ -ray coincidence techniques.

7.5.1 Delayed γ -Ray Energy Spectra

The spectra of delayed γ -rays are complex since many fission products can contribute to them. Although it is almost impossible to resolve

individual lines with NaI detectors, this can be accomplished with high resolution Ge(Li) detectors. About 200 γ -ray peaks in the energy range up to 1.1 MeV have been found in this manner. Figure 7.15 shows the delayed γ -ray spectrum resulting from a 30-s irradiation by a highly moderated ^{252}Cf neutron source and measured for a 30-s period with a NaI detector (Ref. 13).

Figure 7.16 shows high resolution, delayed γ -ray spectra from ^{235}U and ^{239}Pu (Ref. 14). Samples of fissile materials were irradiated for 2 min in a thermal neutron flux of 10^6 $n/\text{cm}^2/\text{s}$ and the γ -spectra for different waiting times were measured with a Ge(Li) detector. The results shown in Figure 7.16 are for a waiting time of 30 min. In general, the ^{239}Pu and ^{235}U spectra are quite similar, except for a few peaks where substantial differences were observed. The peak at 1248 keV is six times higher in ^{235}U than in ^{239}Pu , and the peak at 767 keV is about twice as high in ^{239}Pu as in ^{235}U . Furthermore, the ^{105}Ru peak at 724 keV appears in the ^{239}Pu spectrum, but not in the ^{235}U spectrum.

Figure 7.17 shows a portion of the delayed γ -ray spectra from the fissile isotopes ^{233}U , ^{235}U , ^{239}Pu , and ^{241}Pu taken with a Ge(Li) detector 90 min after a 5-min thermal neutron irradiation (Ref. 15). The spectra are quite similar, although a few individual peaks such as the 724-keV line from ^{105}Ru and some of the ^{134}I peaks have different relative amplitudes.

Figure 7.15. Delayed γ -ray pulse height distribution measured with NaI detector (30 s irradiation time, 0.5 s delay, and 30 s measuring time).

Figure 7.16. ^{239}Pu and ^{235}U pulse height distribution due to fission product γ -ray measured with Ge(Li) detector (Ref. 14).

Similar results were observed following high intensity Bremsstrahlung irradiation (Ref. 16). In those measurements, discrimination ratios on the order of factor of 3 were observed between ^{235}U , ^{239}Pu , and ^{238}U . The main drawback to the use of these signatures, whether induced by neutron or photofission, is the requirement for high intensity sources of interrogating radiation.

Figure 7.17. Comparison of a portion of the fission product γ -ray spectra from ^{241}Pu , ^{239}Pu , ^{235}U , and ^{233}U . The lines labeled “B” are background from the natural activity present in the samples (Ref. 15).

The average pulse height of a NaI-delayed γ -ray spectrum corresponds to a γ -ray energy of approximately 0.6 to 0.8 MeV.* This energy is less than the ~ 1 MeV average energy of γ -rays because the detector is more efficient for lower energy γ -rays. The 0.6 to 0.8-MeV value is, however, higher than the passive γ -ray energies in ^{235}U (0.185 MeV) and ^{239}Pu

*It depends on detector size, measuring configuration, and length of irradiation and delay times.

(~ 0.400 MeV). Therefore, these γ -rays have a better chance of escaping from the sample than do the passive radiations. The penetrability of the detected γ -rays can be significantly increased by biasing out all energies below 1 MeV, for example. This method is often used, however, it incurs a substantial loss in signal intensity.

7.5.2 Delayed γ -Ray Time-Dependence

Two basic characteristics of delayed γ -rays from fission distinguish them from delayed neutrons: much higher intensities and much slower decay, which is referred to as time dependence.

The time-dependence of delayed γ -rays varies somewhat from fissile to fertile isotopes, but does not vary significantly from one fissile isotope to another, a situation analogous to the delayed neutron time-dependence.

For times up to 2×10^3 s after fission, the delayed γ -ray activities from ^{235}U and ^{238}U have been conveniently* fitted into five groups (Ref. 17) similar to the six delayed neutron groups. The group half-lives and group yields are shown in Table 7-7. The delayed γ -ray intensity/fission for ^{235}U as calculated from these group constants for different irradiation times (t_i) is given in Table 7-8. This table shows the loss of long-lived delayed γ -rays when short irradiation times are used. Calculations indicate that an irradiation time of about 60 s excites about 50 percent of the delayed γ -rays. Delayed γ -ray decay for ^{235}U fission, with an irradiation time of 1 s, is shown in Table 7-9. Since only fast γ -ray groups are excited within such a short irradiation time, the decay is quite rapid. Note that even at 30 s after an irradiation of 1 s, the number of delayed γ -rays/fission exceeds the total number of delayed neutrons/fission for most fissionable isotopes.

The total relative intensities of delayed γ -rays versus time for ^{235}U and ^{239}Pu are shown in Figure 7.18 (Ref. 18). These curves were obtained with a low resolution plastic scintillator. By comparing the intensity at early times with that at later times, a discrimination ratio of about 1.30 was obtained for these isotopes, which is slightly higher than that obtained from delayed neutron kinetics (see Table 7-3). However, the initial and integrated intensity of delayed γ -rays/fission in ^{235}U is higher by about 1.7 than that in ^{239}Pu . This discrimination ratio is fully usable since the absolute intensities are high as compared to those of the delayed neutrons. More data on the delayed γ -ray spectrum and its time dependence can be found (Ref. 19).

*There is very little physical meaning to these groups, since delayed γ -rays are being emitted by many isotopes with many different half-lives. However, the grouping, done by a least-squares fit method, does facilitate calculations which are useful for the design and understanding of ANDA techniques that are based on this signature.

Table 7-7. Delayed γ -ray groups.

Group (<i>i</i>)	$T_{1/2}^i$ (s)	Abundances (yield/fission)	
		$a_i(^{235}\text{U})$	$a_i(^{238}\text{U})$
1	0.29	0.067	0.54
2	1.7	1.05	1.76
3	13	1.92	2.81
4	100	1.73	2.02
5	940	1.55	1.55

Table 7-8. Delayed γ -ray intensity $\gamma_d(t)$ (photons/fission) at the end of irradiation time t_i of ^{235}U .

t_i (s)	$\gamma_d(t)$
2	0.87
3	1.13
5	1.49
8	1.84
10	2.02
20	2.62
100	4.00
1000	5.58
10000	6.32

Table 7-9. Decay of delayed γ -rays following 1 s irradiation.

Time After End of Irradiation	Yield
t (s)	$\gamma_d(t)$
0.00	1.32×10^0
1.00	5.26×10^{-1}
5.00	1.61×10^{-1}
10.00	8.27×10^{-2}
15.00	6.03×10^{-2}
20.00	4.79×10^{-2}
25.00	3.90×10^{-2}
30.00	3.22×10^{-2}

Figure 7.18. Relative intensity of γ -rays ($E_\gamma > 380$ keV) versus time following neutron-induced fission of ^{235}U and ^{239}Pu (Ref. 18).

7.6 SUMMARY AND CONCLUSIONS

Prompt and delayed neutrons and γ -rays are emitted after fission. The yields, multiplicities, and energy spectra of the prompt neutrons depend on the fissioning isotope and the type and energy of the radiation inducing fission. Prompt neutron multiplicities are advantageously used in ANDA coincidence detectors. The high average energy of prompt neutrons also makes it possible to distinguish them from the source neutrons in techniques using low-energy or thermalized neutron sources. Prompt neutrons from photon-induced reactions, in fissionable isotopes, are easily discernible from the interrogating γ -radiation.

Delayed neutron time dependencies and yields vary among fissionable isotopes. They can be used as signatures and to obtain unambiguous evidence that fission has occurred. When applicable (that is, in photon-induced fission), the ratio of prompt to delayed neutron yield provides a very useful signature which is, to a large extent, free of effects that can beset the individual components of the measurement.

Prompt γ -rays provide high multiplicities for use alone or in conjunction with neutrons in coincidence detectors in ANDA systems that employ neutron interrogation. Delayed γ -rays can be used, in devices such as rod scanners, as measures of the total fissile content or

variations in fissile content. Delayed γ -rays are abundant as compared to delayed neutrons. Their penetrability through the sample is much higher than that of the passive γ -rays emitted naturally by the fissile isotopes. Thus, delayed γ -rays constitute one of the most usable signatures in ANDA systems.

Table 7-10 summarizes the main features of these radiations in ANDA systems.

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Table 7-10. Main signatures for ANDA.

Conditions	Neutrons		γ -Rays	
	Prompt	Delayed	Prompt	Delayed
Number/fission	2.5-3 (n_{th}, f) 4-5 (n_f, f)	0.007-0.05	6-8	6-8
Average energy (MeV)	~ 2	0.4-0.5	~ 1	~ 1
Properties used	yield, multiplicities	yield, time dependence	multiplicities	yield, time dependence, gross energy
Ease of detection	fair to high	high	high in (n, f), impossible in (γ, f)	high
Usefulness in coincidence based techniques	high	none	high in (n, f)	none
Usefulness in gross counting techniques	high for (n_{th}, f) and (γ, f) low to fair in other cases	high	high for (n, f), impossible for (γ, f)	high
Possible obtainable SNM sensitivity	high	fair in (n, f) high in (γ, f)	high in coincidence technique, low in others	high
Discrimination ratio	low in (n, f) high in (γ, f)	fair in (n, f) high in (γ, f)	low	fair

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CHAPTER 8

NEUTRON AND PHOTON DETECTION SYSTEMS AND ELECTRONICS

8.1 INTRODUCTION

All ANDA systems rely on the detection of neutrons and/or photons which are emitted by the sample. In some techniques, fast neutrons are detected; in others, neutrons may be slowed down before detection, possibly to thermal energies. Many ANDA methods are based on coincidence counting—the simultaneous detection of two or more neutrons and/or photons emitted by the same nuclear decay (usually fission). Some coincidence techniques require very fast timing in order to respond to prompt fast neutrons and/or photons. Other coincidence techniques have relatively slow timing and require high efficiency rather than fast response. Energy resolution of the detected radiation may also be of some importance, for example, one may want to detect γ -rays of a certain energy or in a specified energy region.

All of these considerations enter into the choice and proper design of the detectors and the associated electronics. In this chapter, the principal types and features of the commonly used slow neutron, fast neutron, and γ -ray detectors are described. Pulse formation and shaping by the associated electronics and some aspects of coincidence methods important in ANDA are also discussed.

8.2 NEUTRON DETECTORS

Detection of neutrons is accomplished by detection secondary particles which result from interactions of neutrons with nuclei (see Chapter 2). Some of these reactions are:

1. The absorption of the neutron by a nucleus with the prompt emission of a charged particle, for example, (n,p) or (n,α) . The

charged particle and recoil product nucleus then cause ionization in the detector.

2. Neutron-induced fission, (n,f) . The detector registers ionization caused by fission fragments or counts the prompt or delayed neutrons and γ -rays.
3. Neutron scattering. The recoil nucleus produces ionization in the detector.
4. The formation of a radioactive nuclide after absorption of a neutron. The nuclide's subsequent activity can then be measured, but this method is not employed in ANDA.

The first reaction is used for slow neutron detection. Because the ${}^6\text{Li}(n,\alpha)$, ${}^{10}\text{B}(n,\alpha)$, and ${}^3\text{He}(n,p)$ reactions have high cross sections for thermal neutrons, they are commonly used. The neutron threshold energies of other reactions of this type are usually above 1 MeV and have small cross sections, hence, they are seldom used in ANDA. The fission reaction (reaction 2) can be used for both slow and fast neutron detection. Neutron scattering, reaction 3, is the most common mechanism used for fast neutron detection, although the absorption reaction, reaction 1, can also be used.

8.2.1 Thermal Neutron Detectors

The boron detector is a simple and common device used for detection of thermal neutrons. It consists of a cylindrical metallic tube filled with boron trifluoride (BF_3) gas, which in most safeguard applications has been highly enriched (up to 96 percent) with ${}^{10}\text{B}$. The tube's casing acts as a cathode, and the anode is a central wire. A high voltage is applied between the anode and cathode. Boron-coated counters*—their walls coated with a compound containing boron and filled with an ordinary counting gas—also exist, but are less commonly used, except in high γ -ray fields, such as those near spent fuel elements (see below).

Neutron capture in ${}^{10}\text{B}$ leads to an exothermic (n,α) reaction, which can proceed in two ways:[†]

*In ANDA, detectors which detect reactions are attached to counters that count the reactions. The whole unit can be considered a counter, since reactions must be detected before they can be counted. The terms "detector" and "counter" are often used interchangeably in this text.

[†] ${}^7\text{Li}^*$ indicates ${}^7\text{Li}$ nucleus in an excited state.

The split between these two reactions (branching ratio) depends on the energy of the incident neutron. For thermal neutrons (0.0253 eV), 6.1 percent of the reactions are branch 1 reactions and 93.9 percent are branch 2 reactions. The cross section for this reaction at 0.0253 eV is 3840 b. The reaction products, the ${}^7\text{Li}$ recoil nucleus and the α -particle, lose their kinetic energy by ionizing the gas. The primary ions in turn produce many more secondary ions (gas multiplication); the gas multiplication factor is typically 10^3 to 10^4 . All the ion-electron pairs are swept to the electrodes because of the electric field: the electrons drift to the anode, inducing a charge on it. Converting the charge to voltage produces an output pulse whose amplitude is proportional to the initial ionization, and hence, to the total kinetic energy of the reaction products (but *not* to the energy of the incident neutron).

Figure 8.1 shows the pulse spectrum of a BF_3 detector irradiated by thermal neutrons. The small peak near channel 150 corresponds to the 2.79 MeV branch of the ${}^{10}\text{B}(n,\alpha){}^7\text{Li}$ reaction; the large peak at channel 100 is the other branch. Some of the α -particles or ${}^7\text{Li}$ nuclei produced near the wall or at the ends of the counter can leave the sensitive counter volume or strike the walls before they give up to the gas all of their

Figure 8.1. Typical pulse height spectrum of BF_3 (1 atm) gas proportional detector.

energy as ionization energy. Because of these end and wall effects, there is a tail below the main peaks corresponding to smaller pulses. There may be a further increase in the spectrum corresponding to very small pulses that are due to γ -rays producing electrons in the chamber's wall. Neutron pulses can be distinguished easily from γ -rays by use of a suitably biased discrimination (see Section 8.4).

The detection efficiency of a boron counter varies with its sensitive length, the pressure of the gas it contains, and the $^{10}\text{B}(n,\alpha)$ cross section (which varies with neutron energy as $1/\sqrt{E}$). At a pressure of 1 atm, the efficiency of a 20-cm long BF_3 counter is about 0.9 for axially incident thermal neutrons, but it drops to about 0.03 for axially incident 100-eV neutrons. The required operating voltage increases with pressure. A pressure of 140 cm of mercury (2 atm) is generally considered a practical maximum for constructing standard BF_3 counters.

BF_3 detectors are widely used when low to medium γ -radiation fields are encountered (less than about 10 rad/hr) because of their excellent discrimination between the large neutron pulses and the much smaller gamma γ -ray pulses. They can be made in a variety of sizes, ranging from approximately 0.6 cm in diameter and 2.5 cm in length to tubes that are 5 cm by 60 cm. Figure 8.2 shows how the count rate varies with voltage. The count-rate plateau typical of these devices is about 300 V wide, and its slope is less than 1 percent/100 V, which explains the great stability of these detectors.

Counters coated with ^{10}B and filled with ordinary counting gas have poorer pulse height resolutions than BF_3 gas-filled counters because the reaction products lose energy in escaping from the coating into the

Figure 8.2. Plateau curve for $^{10}\text{BF}_3$ proportional counter.

gas-filled volume of the tube. Thin coatings minimize the energy loss but result in low efficiency. Figure 8.3 shows a pulse height spectrum for a ^{10}B -coated counter, the poorer resolution is evident. The poorly defined peaks near channels 50 and 90 correspond to detection of the ^7Li and ^4He particles, respectively, because either ^7Li or ^4He will be directed into the active volume of the detector. The count-rate plateau is also much worse than that of the BF_3 counter; its slope is -10 percent/100 V, indicating poorer stability relative to BF_3 counters.

Figure 8.3. Typical pulse height spectrum of ^{10}B -coated proportional detector.

The effect of γ -ray fields on the pulse height distribution of a ^{10}B -coated detector is shown in Figure 8.4 (Ref. 1). This detector is useful even for average γ -ray fields of 10^3 to 10^4 rad/s encountered near spent nuclear fuel assemblies. ^{10}B -coated detectors are also used when low operating voltage (less than 1000 V), high signal (requiring only small amplification), and fast charge collection time (typically less than 50 ns) are desired.

Improved detection efficiency can be obtained by using a proportional detector filled with ^3He instead of BF_3 . ^3He undergoes the following reaction:

Figure 8.4. Pulse height distribution in ^{10}B neutron detectors as a function of the average γ -ray field created by a Linac (Ref. 1).

The cross section of this reaction is about 5300 b at 0.0253 eV and varies as $1/\sqrt{E}$ over a wide energy range. The pulse height distribution from a ^3He counter for thermal neutrons is shown in Figure 8.5. Its single peak results from the complete absorption of reaction products into the counter gas. At low energies, the pulse height distribution due to γ -radiation is similar to that shown in Figure 8.1 for the BF_3 counter. The ^3He gas is usually combined with gas additives chosen to optimize such parameters as operating voltage, pulse rise time, gas multiplication, spectral resolution, neutron sensitivity, and γ -ray sensitivity. A ^3He detector can almost be 100 percent efficient for thermal neutrons at 4 atm pressure. For higher energy (epithermal) neutrons, the efficiency can be improved by increasing the pressure. The count-rate plateau is quite good, its slope is about 1 percent/100 V.

^3He counters have better pulse height resolution and higher efficiency than BF_3 counters. These improved characteristics result from the cross

Fig: re 8.5. Typical pulse height distribution of $^3\text{He}(4\text{ atm})$ gas proportional counter.

section of $^3\text{He}(n,p)$ which is larger than that of $^{10}\text{B}(n,\alpha)$, and from the ability of the ^3He detectors to operate at higher fill pressures (up to 40 atm, as compared to the 2 atm limit for BF_3 detectors). On the other hand, the advantages of BF_3 counters are that they are cheaper and require a lower operating voltage* (usually less than 2500 V).

Fission counters can be used both for thermal neutron detection and for gross fast neutron spectroscopy. Because the total energy of fission fragments is about 160 MeV, it is possible to make fission chambers for neutron detection that are insensitive to γ -rays. Fission chambers are generally filled with argon and operate at low voltage—a few hundred volts (this is the ionization region, with no gas multiplication). To minimize self-absorption, the electrodes are thinly coated ($\leq 1\text{ mg/cm}^2$) with fissionable material. As a result, the efficiency of these detectors is generally low. However, because of their very high discrimination against γ -rays, they may be the best detector when spent fuel elements are being assayed in ANDA. Fission chambers can be used in γ -ray fields as high as 10^7 rad/hr .

Thermal neutrons can also be detected with relatively high efficiency by certain types of scintillation counters. Various substances emit

*However, for the same pressure or the same detection efficiency BF_3 counters require higher operating voltages.

scintillations of light when irradiated by charged particles or photons. For thermal neutron detection, secondary charged particles or photons (resulting from neutron reactions in the detector) produce the scintillation: two such reactions are $^{10}\text{B}(n,\alpha)$ and $^6\text{Li}(n,\alpha)$. The light scintillation causes photoelectric emission of electrons from the cathode of a photomultiplier tube (PMT) which is optically coupled to the scintillator. The emitted electrons are further multiplied at successive tube electrodes (dynodes). The output current pulse at the tube anode is then amplified and measured.

Scintillation counting, thus, involves five consecutive stages:

1. Absorption of the nuclear radiation by the scintillator.
2. Conversion of the energy dissipated in the scintillator to light (photons).
3. Collection of photons on the photosensitive cathode of the photomultiplier tube.
4. Emission of photoelectrons.
5. Multiplication of electrons.

Good scintillators require four characteristics: (1) high efficiency in converting the energy of nuclear particles into the energy of fluorescent radiation, (2) transparency to their own fluorescent radiation, (3) short decay time for the fluorescent radiation, and (4) a spectral distribution of the scintillation that matches the responses of available photocathodes. Other properties, such as density, form, state of matter, and versatility in sizes and shapes, should also be considered in designing a good scintillator. The requirement for good time resolution is particularly important in coincidence experiments. The shorter the resolution time, the smaller will be the ratio of chance coincidences to true coincidences (see Section 8.6.1).

Some operating characteristics of the counters discussed in this section are outlined in Table 8-1. As can be seen from the table, the main disadvantage of scintillation counters for ANDA applications is that they cannot retain both high efficiency and good discrimination against γ -rays. They also have poorer stability than BF_3 or ^3He detectors.

One type of scintillation detector used for thermal neutrons consists of $\text{ZnS}(\text{Ag})$ mixed with the (n,α) "converter" such as boron or lithium compounds and a plastic bonding medium. Neutron pulses are larger than γ -ray pulses, in this type of detector, and also take longer to decay in the scintillator. Both of these properties can be used to distinguish neutron pulses from γ -ray pulses and to reject the latter. Figure 8.30 (Section 8.4.4) illustrates the principle of a "pulse-shape discriminating circuit" that rejects the shorter γ -ray pulses while passing the neutron

Table 8-1. Thermal Neutron Detectors.

Detector, Type	Nuclear Reactions Applied	γ -Ray Sensitivity	Neutron Detection Efficiency	Comments
BF ₃ -filled proportional counter	$^{10}\text{B}(n,\alpha) \text{ } ^7\text{Li}$	Some γ -ray discrimination is possible; functional in γ -ray field of <10 rad/hr	Bare detectors are effective only for thermal neutrons	Good pulse-height spectrum. Very good stability.
^{10}B -coated proportional counter	$^{10}\text{B}(n,\alpha) \text{ } ^7\text{Li}$	Functional in γ -ray field of <10 ⁵ rad/hr		Requires moderately low voltage. Very fast response. Poor pulse-height spectrum.
^3He -filled proportional counter	$^3\text{He}(n,p) \text{ } ^3\text{H}$	γ -ray discrimination is possible; functional in γ -ray fields of <2 rad/hr	Effective for thermal neutrons; used for neutron spectroscopy in the range 0.2-2 MeV	Very good pulse-height spectrum. Very good stability.
Boron+ scintillator (e.g., NaI(Tl))	478 keV γ -ray from $^{10}\text{B}(n,\alpha) \text{ } ^7\text{Li}$	Sensitive to background γ -radiation	10% Efficiency for neutrons up to 10 keV	Suffers from PMT instability
ZnS(Ag) + B ₂ O ₃	$^{10}\text{B}(n,\alpha) \text{ } ^7\text{Li}$	Insensitive to γ -rays	Effective only for thermal neutrons	Small detector
Lithiated scintillator, 95% ^6Li in ZnS(Ag)	$^6\text{Li}(n,\alpha) \text{ } ^3\text{H}$	Insensitive to background γ -rays	High efficiency for slow neutrons	Only small detectors
LiI(Eu), ^6Li glass scintillation	$^6\text{Li}(n,\alpha) \text{ } ^3\text{H}$	Sensitive to γ -radiation	Useful for fast neutron spectroscopy	PMT instabilities
Fission counter	Fission	Insensitive to γ -rays; functional in γ -ray field in excess of 10 ⁶ rad/hr	Used for slow and fast neutrons	Requires low anode voltage. Very stable. Very fast response. Very low efficiency.

pulses. Figure 8.6 shows bias curves (count rate versus pulse amplitude) with and without pulse shape discrimination and indicates the improvement in the neutron to γ -ray ratio resulting from the pulse shape discrimination (Ref. 2).

Finally, it should be mentioned that one can use semiconductor detectors (see Section 8.3.2), which respond to charged particles, to detect thermal neutrons. To do so, one places a converter foil next to the detector. Charged particles from (n,p) , (n,α) , or (n,f) reactions in the converter are then detected. These detectors have not been used in ANDA systems so far, although they have some attractive characteristics such as very fast response. Their main disadvantages are small size and high susceptibility to radiation damage.

Comprehensive discussion of all kinds of neutron detectors is presented (Ref. 2-5).

Figure 8.6. Bias curves for boron-ZnS(Ag) scintillator
(a) with pulse shape discriminator,
(b) without pulse shape discriminator.

8.2.2 Moderated Fast Neutron Detectors

One way to detect fast neutrons is to first slow them down and then use any of the thermal neutron detectors discussed above. This is a common approach employed in ANDA of nuclear materials. Two examples of detectors of this type are the "long counter" and the "slab detector."

The long counter (Ref. 6) consists of one or more BF_3 or ^3He tubes in a relatively long shielded cylindrical moderator, as shown in Figure 8.7. The counter is sensitive to fast neutrons entering the right-hand side. Neutrons entering from other directions are moderated and then captured in the borated paraffin. The arrangement of counters and moderator is such that the efficiency for detection of fast neutrons entering from the front is independent of the neutron energy (Figure 8.8). The intrinsic efficiency* of the arrangement in Figure 8.7 is about 11.5 percent as compared to a standard long counter (one BF_3 tube) efficiency of about 2.3 percent.

The slab detector (Ref. 7) contains a number of ^3He tubes in a (polyethylene) moderator, in parallel slab geometry as shown in Figure 8.9. This arrangement is useful for detection over large areas or large solid angles.

Figure 8.7. Design features of a high-efficiency modified long counter.

This configuration consists of five ^3He (6 atm) counters imbedded in a 9-in.(22.5 cm)-diameter polyethylene cylinder.

*The probability of detection of neutrons impinging on the detector system, in this case from the front.

Figure 8.8. Efficiency of the modified long counter as a function of neutron energy as determined by radioactive sources. The absolute efficiency for detection of neutrons from sources placed 1.0 m from the detector face are given by the left-hand ordinate. On the right-hand ordinate, the intrinsic efficiency for detection of neutrons striking the front face of the detector is presented as a function of neutron energy.

Figure 8.9. Cross-sectional view of a ^3He -slab detector. This configuration is used to obtain coarse neutron-energy spectrum information. In the flat-response mode of operation, all materials between the front and back counter banks are removed.

The detector shown in Figure 8.9 has 13 6-atm ^3He tubes arranged in two banks embedded in polyethylene. The sensitive area is about 50×60 cm, and the intrinsic efficiency is about 10 percent. Coarse indication of the neutron spectrum can be obtained if a slab of polyethylene is placed between the front and rear banks. Figure 8.10 shows the count-rate ratio of the front to back banks as a function of the energy of the incident neutrons. For a flat response, the polyethylene slab is removed. The resulting efficiency curves for each bank separately and for the entire unit are shown in Figure 8.11.

Table 8-2 gives intrinsic efficiencies for various moderated detectors and fast neutron sources (Ref. 8).

8.2.3 Nonmoderated Neutron Detectors

In many ANDA applications, especially those which use fast coincidence techniques, fast neutron detectors that depend on first moderating the neutrons are much too slow to be useful. The slowing-down time for the neutrons is typically 10 to 100 μs , but fast systems must detect the neutrons within 10 to 100 ns. Gas-filled proportional counters and scintillators are used to directly detect the fast neutrons by scattering rather than absorption reactions. For these detectors, neutrons must not be slowed down.

Figure 8.10. Response ratios between front and back counter banks of the ^3He slab detector versus incident neutron energy.

Figure 8.11. Efficiency of ³He slab detector (absolute efficiency under the same condition as in Figure 8.8).

Table 8-2. Intrinsic efficiency of fast neutron detectors.

Reference Source	Average Neutron Energy (MeV)	Intrinsic Efficiency (%)			
		Long Counter (1 tube)	Large Slab (13 tubes)	Small Slab (5 tubes)	5-cm Pilot ^a
²⁴¹ Am-Li	0.5	2.3 ± 0.1	—	15.0 ± 0.7	34 ± 5
²³⁸ Pu-Li	0.65	2.3 ± 0.1	18.5 ± 0.7	14.3 ± 0.6	44 ± 3
²³⁸ Pu-Be	4.2	1.9 ± 0.1	15.3 ± 0.7	9.6 ± 0.4	54 ± 4

^aFast scintillator.

Helium-4 and hydrogen or light hydrogen-containing compounds, like methane, are commonly used in such counters. The scattering cross sections of H and ⁴He are shown in Figure 8.12 along with the fission neutron spectrum. The ⁴He cross section matches the fission spectrum quite well, it is highest in the energy region where the fission spectrum peaks.

Typically, a ⁴He-recoil proportional counter operates at about 10 to 20 atm pressure at an efficiency up to about 1.5 percent (see Figure 8.13). Hydrogen recoil counters operate at a few atmospheres at efficiencies of about 1 percent. The pulse height distribution in recoil counters is quite broad because the energy of the recoil particle (α or proton) depends on the scattering angle. For hydrogen recoils, the pulse

Figure 8.12. Neutron scattering cross section for ^1H and ^4He .

height distribution is essentially flat from the energy of the incident neutron to zero. However, a number of effects can perturb the response curve: wall and end effects, multiple scattering of the neutron, and scattering from nonhydrogen-containing material in the gas (for example, carbon, if methane is used). Figure 8.14 shows a calculated pulse height distribution for a hydrogen counter. A pulse height spectrum for ^4He is shown in Figure 8.15.

To understand the pulse height spectra of Figures 8.14 and 8.15, recall that for a monoenergetic fast neutron of energy E_n , the energy loss per scattering can have any value from zero up to $(1-\alpha)E_n$ with equal probability* (see Chapter 2). For H, α equals 0. For ^4He , α equals $9/25 = 0.36$. The ideal pulse height spectrum in an H-filled detector for a monoenergetic fast neutron is flat from pulse amplitude zero to a maximum corresponding to the neutron energy (E_n). For the He detector, the maximum pulse height corresponds to $0.64 E_n$. The response

*This is strictly correct if the scattering is isotropic in the center of mass system.

Figure 8.13. Efficiency of $^4\text{He}(18 \text{ atm})$ proportional counter.

Figure 8.14. Calculated response function for 500-keV neutrons in a hydrogen-filled proportional counter.

function to monoenergetic neutrons is not flat but peaked.* For a fission spectrum distribution of fast neutrons, such as from ^{252}Cf , the pulse height distribution is the convolution of the distributions from monoenergetic neutrons, weighted by that of the fission spectrum. This

*This is because of the anisotropy of the neutron scattering from the nuclei in the center of mass system.

Figure 8.15. Typical pulse height spectrum of ^4He proportional counter
(a), (b) spectrum of fission neutrons from ^{252}Cf source,
(c) noise spectrum,
(d) spectrum from 50 mrad/hr (2.6 MeV) γ -ray field.

convolution (Figure 8.15) results in the broad peak (a) in the figure. However, because some neutrons are scattered more than once, there is an augmentation of low-amplitude pulses corresponding to the pulse height distributions for lower energy, scattered neutrons. This augmentation accounts for the rise (b) in low-amplitude pulses. At low pulse amplitudes, amplifier noise (c) or small amplitude γ -ray pulses (d) may cause a further sharp increase in the pulse height distribution. Similar considerations apply to hydrogen-recoil counters. Because of the broad pulse height spectra, characteristic of recoil counters requiring unfolding of the spectra to obtain the original neutron spectra, they are not easy to use as fast neutron spectrometers. Nevertheless, proton (H) recoil detectors have been used in certain applications, such as measurement of nuclear reactor spectra and determination of delayed neutron spectra.

The time resolutions of ^4He and CH_4 -filled tubes range from $0.1\ \mu\text{s}$ to over $100\ \mu\text{s}$, depending on the size of the tube and the type, pressure, and multiplication factor of the gas (Ref. 9). Judicious choice of these parameters can render these detectors useful for fairly fast ($1\text{--}10\ \mu\text{s}$) coincidence detection of prompt, fast-fission neutrons, however, this application is practical only in passive NDA.

^3He detectors can also be used to detect fast neutrons as well as thermal neutrons. They can give resolution in the (n,p) peak as good as $50\ \text{keV}$ for 600-keV neutrons; however, the pulse height spectra (Figures 8.16 A and B) show a broad distribution of pulses due to scattering recoils and a "thermal" peak due to moderation of neutrons in nearby materials. Since there is always some moderating material present in ANDA applications, ^3He counters cannot normally be used for fast neutron detection because of this relatively poor pulse height distribution.

8.2.4 Scintillators for Counting Fast Neutrons

Scintillation detectors can be used for detection of fast as well as for thermal neutrons. For fast neutrons, the charged particles needed to produce scintillations come from neutron scattering reactions, usually

Figure 8.16A. Theoretical energy spectrum of charged particles generated by fast neutrons incident on a ^3He detector.

*Thermal neutrons are removed by suitable filter; for example, cadmium.

Figure 8.16B. Actual energy pulse height distribution of 1.27-MeV neutrons measured with high resolution ^3He spectrometer (courtesy Seforad Ltd. Emeck Hayarden, Israel).

the (n,p) reaction on hydrogen present in the scintillator. For this application, various solid (plastic) or liquid scintillators and Hornyak counters are useful. The Hornyak counter is a mixture of a hydrogen-containing material (wax or lucite) with $\text{ZnS}(\text{Ag})$ phosphor, similar to the scintillator described previously for thermal neutron detection. Although these counters have low efficiency, they are virtually insensitive to γ -ray background. Finally, liquid scintillators can also be used, these fast scintillators can be made very large and in a variety of shapes. Since all of these scintillators respond to recoils, their pulse height distributions are broad.

Table 8-3 shows some of the properties of important plastic scintillators, and Table 8-4 gives similar data for liquid scintillators. Table 8-5 summarizes general properties of different fast neutron detectors.

Pulse height distributions and efficiencies for a liquid scintillator detector (NE-213, see Table 8-4) have been obtained for nearly monoenergetic neutrons from 0.2 to 22 MeV (Ref. 10). These results, which also apply to the NE-211 and to plastic scintillators, are shown in Figure 8.17. The general features of these spectra—flat distribution up to a maximum pulse height corresponding to the energy of the incident neutrons—are explained in Section 8.2.3.

For use in ANDA applications, fast neutron scintillants must have fast response. All of the scintillators listed in Tables 8-3 and 8-4 have short

Table 8-3. Properties of representative plastic scintillators of interest as fast neutron detectors.

Manufacturer ^a	Identification	Light Output ^b (%)	Decay Constant (ns)	Wavelength of Max. Emission ^c (nm)	H/C Atomic Ratio	Comments
(1)	NE 102 A	65	2.4	425	1.104	General applications
(1)	NE 104	68	1.8	405	1.099	Fast timing
(1)	NE 111	55	1.7	375	1.093	Ultrafast timing cannot be used in large sizes because of light self-absorption
(1)	Pilot B	68	1.8	408	1.100	General applications
(1)	Pilot U	67	1.36	391	1.100	Ultrafast timing
(2)	SPF	55 - 65	4	430	1.0	General applications
(3)	KL 211	60	2.2	420	0.992	General applications
(3)	KL 236	65	1.87	410	0.992	Ultrafast timing

^aSee Table 8-4.^bLight output relative to that of anthracene.^cnm = nanometer = 10^{-7} cm.

Note: For all the scintillators shown:

Density = 1.03 - 1.06 g/cm³

Refractive index = 1.58 to 1.59

Softening temperature = 75 to 85°C

Table 8-4. Properties of liquid scintillators.

Manufacturer ^a	Identification	Light Output ^b (%)	Decay Constant (ns)	Wavelength of Max. Emission (nm)	H/C Atomic Ratio	Comments
(1)	NE 211	78	2.6	425	1.248	General use
(1)	NE 213	78	3.7	425	1.213	For pulse shape discrimination against γ -rays
(1)	NE 228	45	—	385	2.11	High H/C ratio

^aManufacturer code:

- (1) Nuclear Enterprises, Ltd. (CA)
- (2) Amperex Electronic Corp. (NY)
- (3) Koch-Light Laboratories, Ltd. (GB)

^bLight output relative to that of anthracene.

Table 8-5. Unmoderated^a fast neutron detectors.

Detector Type	Nuclear Reaction	Sensitivity		Comments
		γ -Ray	Neutron	
³ He-filled proportional counter	³ He (n,p) ³ H	Low to medium	Low	Can be used as a spectrometer
⁴ He-filled proportional counter	⁴ He (n,n) ⁴ He	Medium	Low	Good for fission neutrons; can be used in large banks to increase efficiency
H-filled (H or methane) proportional counter	(n,p) scattering	Medium	Low	The only practical direct detector for sub-MeV neutrons down to 50-keV; can be used in large banks
Hornyak	(n,p) scattering	Very low	Low	Good for high γ -ray fields
Plastic scintillator	(n,p) scattering	Very high	Medium	Highest efficiency detector for fast neutrons ($E_n > 0.5$ MeV). Insensitive to sub-MeV neutrons. Very fast response; good for coincidence techniques; relatively low cost; main deficiency—high γ -ray sensitivity
Liquid scintillator	(n,p) scattering	Very high	Medium	As above, low cost for large volume detectors; some scintillants can distinguish between γ -ray and neutrons; this is applicable to low γ -ray fields and relatively small detectors
Fission chambers	(n,f) on ²³⁸ U, ²³² Th, ²³⁷ Np, ²³⁴ U, etc.	Extremely low	Very low to low	Can be used in very high γ -ray field; fast response; relatively expensive

^aOther fast neutron detectors, like the long counter discussed in Section 8.2.1, first moderate neutrons to thermal energies and then detect them as thermal neutrons.

Figure 8.17. Absolute intrinsic differential efficiency of a 4.60-cm by 4.65-cm-diameter NE-213 (and NE-211) scintillator, normalized to 1 neutron incident on cross-sectional area. The neutron beam is perpendicular to the scintillator axis. (Note the change of scale for 8.03 MeV and 14.01 MeV incident neutron energies.)

decay constants and sufficiently high light output. NE-102, for example, is commonly used in fast ANDA coincidence circuits (for example, fission multiplicity detectors—FMD), which are discussed in further detail here and in Chapter 9.

In general, plastic and liquid scintillators are characterized by high efficiency for both neutrons and γ -rays. Therefore, they can be small, which may be important, in ANDA applications where space is limited. The pulse height distributions are also broad for neutrons because of the recoil proton energy distribution, and are broad for γ -rays because of the small amount of full energy absorption. The broad pulse height distributions make energy determination difficult; however, this determination is often not too important in ANDA. The short decay times (see Tables 8-3 and 8-4) allow scintillation detectors to be used for fast counting or fast coincidence methods.

8.2.5 Neutron Spectrometers

Fast neutron spectrometry is seldom used in ANDA applications, mainly because of its low efficiency. Also, very few fission signatures

require spectrometry, one of them is the spectrum of delayed neutrons from different fissile isotopes (see Chapter 7). The principal methods used in fast neutron spectrometry are briefly described in this section.

Fast neutron detectors, which respond to recoils, have broad pulse height distributions for monoenergetic neutrons. The pulse height spectra can be unfolded, with some effort, to obtain the original neutron spectra. Since this type of spectrometer has relatively high efficiency (compared with alternate methods to be described), it is preferred in certain circumstances. As a result, computer programs are available for determining the response function and for unfolding the neutron spectrum.

However, several more direct techniques can sometimes be used for fast neutron spectrometry: ${}^6\text{Li}$ and ${}^3\text{He}$ semiconductor sandwiches, proton recoil "telescopes," and time-of-flight techniques, among others. In the lithium sandwich detector, a thin layer of material containing lithium (LiF) is placed between two semiconductor detectors. For low neutron energies, the reaction products from the ${}^6\text{Li}(n,\alpha){}^3\text{He}$ reaction are detected in the two semiconductors. "True" events are detected by a coincidence between the two detectors. The total energy deposited on the two detectors is equal to the Q -value of the reaction (MeV) plus the incident neutron energy. One drawback of this method is that, as the incident neutron energy becomes larger, the reaction products are not, sometimes, emitted in opposite directions, and both can end up in the same detector. In this case, the event will not be recorded since there is no coincidence. ${}^3\text{He}$ can be used instead of ${}^6\text{Li}$ because it results in somewhat better resolution. The charged particles do not lose as much energy in the ${}^3\text{He}$ gas as they do in the ${}^6\text{Li}$ coating. Compared to ${}^6\text{Li}$, the ${}^3\text{He}$ system has also a higher cross section and a higher efficiency because the range of the charged particles limits the amount of ${}^6\text{Li}$ that can be used.

The proton recoil telescope responds only to recoil protons emitted at a given angle with respect to the incident neutron direction. If the angle is fixed, there is a one-to-one correspondence between the proton energy (which can be easily determined) and the neutron energy. The drawback of the telescope is its very low efficiency due to the small angular region in which events are measured.

Time-of-flight techniques measure the time between the emission of the neutron in a fission event, for instance, and its later pickup by a fast response detector, such as a plastic scintillator, after the neutron has traveled an accurately known distance from source to detector. This measurement gives directly the velocity and, hence, the neutron energy, but, like proton recoil telescopes, it has extremely low efficiency.

8.3 PHOTON DETECTORS

ANDA often involves γ -rays measurement. These may originate either in the interrogation source or in the sample as a result of fission or other reactions. Two types of detectors, scintillators and solid-state germanium detectors, are described in the following sections. Both types have wide application in the assay of fissionable materials. These and other types of γ -rays detectors used for specialized detection problems are discussed extensively (Ref. 3, 11, and 12).

8.3.1 Scintillation Detectors

When γ - and x-rays pass through matter, such as the detector, they transfer part of or all of their energy to electrons by the processes described in Chapter 3: photoelectric absorption, Compton scattering, and pair production. In scintillation detectors, these secondary electrons dissipate their energy in turn by ionization or excitation of molecules, which emit light scintillations, during their return to normal states. The light scintillation is then converted by a photomultiplier tube into a pulse of current or voltage. Under proper conditions, the height of the pulse is proportional to the energy lost by the ionizing particle in the crystals. Thus, this system can be used to measure energy distributions as well as the number of γ -rays. The process is quite similar to that previously described for neutron detection.

A wide variety of scintillators are used for detection of γ -rays. Several of the more important ones along with some of their characteristics are listed in Table 8-6. The first two materials in the table are single organic

Table 8-6. Properties of γ -ray scintillators.

Scintillator	Type ^a	Density (g/cm ³)	Index of Refraction	Light Output (% of Anthracene)	Decay Time (ns)
Anthracene	Org. crystal	1.25	1.62	100	30
Stilbene	Org. crystal	1.16	1.63	50	4.5
NE102	Plastic	1.032	1.58	65	2.4
NE213	Org. liquid	0.874	1.51	78	3.7
NE901	Glass	2.64	1.58	28	75
NaI(Tl)	Inorg. crystal	3.67	1.85	230	230
CsI(Na)	Inorg. crystal	4.51	1.84	219	630
CsI(Tl)	Inorg. crystal	4.51	1.80	104	1000
CsF	Inorg. crystal	4.11	1.48	12	5
CdWO ₄	Inorg. crystal	7.90	2.2	39	500,20 000
Bi ₄ Ge ₃ O ₁₂	Inorg. crystal	7.13	2.15	18	300

^aOrg. = Organic; Inorg. = Inorganic.

material crystals. The next two materials are organic materials dissolved in organic solids or liquids, and the others are inorganic crystals or glass. In addition to these materials, xenon and other inert substances in gaseous, liquid, and solid phases and various glasses can be used as scintillators.

The most commonly used scintillator for γ -rays detection is sodium iodide doped with a small amount of thallium impurity, NaI(Tl). NaI is transparent to its own fluorescent light. The fluorescent spectrum is well matched to photocathodes used in some photomultiplier tubes. This scintillator has high efficiency and fairly good resolution and can be made in all sizes up to 30 cm by 30 cm. The most common sizes are 7.5 cm by 7.5 cm and 12.5 cm by 12.5 cm. Figure 8.18 shows a NaI crystal-photomultiplier combination. To prevent moisture retention and light loss, the crystal is encapsulated in an air- and light-tight reflecting material such as aluminum coated with magnesium oxide. Photoelectrons emitted from the photocathode are multiplied at each of the dynode stages in the photomultiplier tube.

A typical pulse height spectrum for incident γ -rays of a particular energy (^{137}Cs , 662 keV) is shown in Figure 8.19. A peak results, if the γ -ray is totally absorbed by the crystal and its energy converted to fluorescent radiation that is then detected without loss in the photomultiplier. Ideally, this peak would be represented by a line, but losses and statistical fluctuations in the photomultiplication process broaden the

Figure 8.18. Schematic of a scintillation detector showing the scintillation crystal (NaI) and photomultiplier tube (PMT).

Figure 8.19. Pulse height distribution of ^{137}Cs γ -ray source measured by a 3-in. (7.5-cm) $\times$ 3-in. (7.5-cm) NaI(Tl) Detector.

line. Some of the γ -rays undergo Compton scattering rather than photoelectric absorption and give up only part of their energy to the crystal before escaping from it. This results in the broad, relatively flat pulse height distribution shown below the peak, known as the Compton shelf. If the crystal is large enough, many of the photons scattered by the Compton process will eventually be absorbed after several collisions, and their total energy will be converted, placing them in the peak and giving a high peak-to-Compton shelf ratio.

The detector efficiency depends on its size and the γ -ray energy, Figure 8.20 shows this dependence. The figure includes a geometry (solid angle) factor, which makes the maximum efficiency 50 percent rather than 100 percent when the source is placed next to the detector (zero source to crystal distance).

The resolution of a NaI-detector is defined as $\Delta E/E$, where ΔE is the full width at half the maximum (FWHM) of the photopeak of the 662-keV γ -ray from ^{137}Cs , and $E = 662$ keV. A good, medium sized (7.5 cm $\times$ 7.5 cm) detector with a high-quality photomultiplier should have a resolution of better than 7.5 percent. In fact, superior detectors with resolution smaller than 6.5 percent are now available. Larger cylindrical detectors should have a resolution better than 8.0 percent. Detectors whose resolutions are 10 percent or worse have limited use for γ -ray energy spectrum determinations, but they can serve for gross γ -ray detection. Sodium iodide detectors are used in most ANDA instruments,

Figure 8.20. Calculated efficiencies of NaI [3-in. (7.5-cm) $\times$ 3-in. (7.5-cm)] crystal.

based on the detection of delayed γ -rays from fission, because of their high efficiency and their ability to select specific energy regions.

In coincidence systems, plastic scintillators (for example, NE-102A) are often used for both neutron and γ -ray detection. The γ -ray energy resolution for plastic scintillators is quite poor as compared with NaI detectors since there is very little total energy absorption. Typical pulse height spectra for various γ -ray energies from a detector 12.7 cm $\times$ 10 cm are shown in Figure 8.21 (Ref. 13), the Compton edge (see Chapter 3) for each is indicated by an arrow. The solid lines are pulse height spectra, calculated by Monte Carlo methods. Because of the broad pulse height distribution in hydrogen-containing scintillators, the detection efficiency depends strongly on the pulse height bias, or discriminator setting. Using Figure 8.22 (Ref. 14), one can estimate the intrinsic detection efficiency* of a plastic scintillator, for a certain incident photon energy, by selecting the curve that corresponds to the discriminator bias setting employed in the system to read the value of μ . The efficiency is then estimated by calculating $1 - \exp(-\mu d)$, where d is the effective thickness of the scintillator. For example, the efficiency of a 12.5-cm thick scintillator is about 45 percent for 1 MeV γ -rays when the equivalent bias is set at an energy of 0.25 MeV.

*The intrinsic detection efficiency is the probability that a γ -ray incident on the detector face will induce an electronic pulse higher than the discriminator setting.

Figure 8.21. Pulse height distribution in a plastic scintillator for three γ -ray energies. The arrows indicate the location of the Compton edge. Continuous curve indicates Monte Carlo calculations, broken line indicates experimental results (Ref. 12).

Figure 8.23 shows the detection efficiency, as a function of γ -ray energy, including source-detector geometrical effects, of a large plastic scintillator in a typical ANDA application. Here, the sample is a uniform fissile sample in which fissions are induced by neutrons from a source 20 in. (50 cm) away. This arrangement, which is typical of a group of ANDA instruments based on fission multiplicity detection (see Chapter 9), shows a zero bias efficiency of over 30 percent for low-energy γ -rays. The efficiency at 1 MeV is greater than 20 percent.

Figure 8.22. Linear absorption coefficient μ (cm^{-1}) for hydrogen-containing scintillator versus incident γ -ray energy for different detector bias settings.

Figure 8.23. Efficiency of plastic scintillator in an ANDA system configuration of fission multiplicity detector.

8.3.2 Semiconductor Detectors

Semiconductor detectors are characterized by very high energy resolution. They are, however, among the most delicate photon detectors and have low efficiency. Semiconductor detectors are often used in passive NDA applications, but they are not commonly used in active

systems, except in some developmental work relating to delayed γ -rays following neutron-induced fission and photofission (see Chapters 7 and 9) and in neutron capture studies of fissionable isotopes (Ref. 15). Properties of the important semiconductor detectors are briefly reviewed in this section.

Semiconductor detectors used for γ -ray work include lithium-drifted germanium [Ge(Li)] and high purity, "intrinsic" germanium. Lithium-drifted silicon [Si(Li)] is also used for x-ray or low-energy γ -ray work. In the lithium-drifted detectors, Li ions are diffused into the crystal and combine with impurity sites to form a charge-free (depleted) region. A high purity crystal does not require Li-drifting. In either case, secondary electrons, resulting from γ -ray interactions in the detector, cause electron-hole (that is, positive ion) pairs to be formed in the depleted region, and these are collected by electrodes (as in a gas proportional counter) when a suitable bias voltage is applied.

These detectors are generally smaller than NaI detectors, ranging up to depleted volumes in excess of 100 cm^3 . Their low efficiencies are quoted as the ratio to the efficiency of a 7.5-cm by 7.5-cm (3-in. $\times$ 3-in.) NaI detector for the 1.33-MeV γ -rays of ^{60}Co . A 100-cc Ge(Li) detector has an efficiency of about 20 percent. Recently, single detectors with efficiencies in excess of 40 percent have become available. However, the resolution of Ge(Li) or hyperpure Ge detectors is vastly superior to that of NaI detectors. A good large detector has a resolution of less than 2 keV (FWHM) at the ^{60}Co peaks, and a small detector can have even better resolution—several hundred eV for incident energies of less than 100 keV.

One serious drawback to Ge(Li) detectors is the need to cool them, at all times, to liquid nitrogen temperature. At room temperature, the depleted region is removed by diffusion processes and the crystal is then no longer useful as a detector. They are, therefore, mounted on a heat-conducting rod that is inserted into a Dewar vessel containing liquid nitrogen. Hyperpure Ge detectors and Si(Li) detectors need be cooled only when in use.

Figure 8.24 shows a typical Ge(Li) spectrum for ^{137}Cs . A comparison with Figure 8.19 shows the dramatic improvement in resolution.

8.4 DETECTION ELECTRONICS

The signal that emerges from any detector caused by a neutron or photon is a small charge or voltage pulse. For simple detection purposes it may be sufficient to distinguish this pulse from noise pulses and record it. Usually, however, much more processing is required. In detecting γ -rays, the energy spectrum is almost always needed. This requires

Figure 8.24. ^{137}Cs pulse height spectrum as measured by a Ge(Li) detector.

measuring the pulse height distribution from the detector. In general, a detection and pulse height analysis system requires preamplification, pulse shaping, further amplification, pulse height discrimination or pulse height analysis, and pulse recording or storage. In addition, many ANDA systems involve coincidence techniques. All of these functions are accomplished by the detector's electronics system.

8.4.1 Basic Electronic Circuits

A typical, simple system of nucleonic instrumentation is shown in Figure 8.25. The components of the system will be described emphasizing important factors that are under the control of the operator.

There are, as discussed before, three categories of detectors: semiconductors, scintillators, and gas-filled counters (ion chambers or proportional counters). In all these, the electrical output signal is an impulse of charge (current $\times$ time). The area of the impulse (the total charge generated by each ray or particle absorbed by the detector) is directly proportional to the absorbed energy. This energy, as has been pointed out previously, may not be the same as the energy of the incident radiation.

The duration of the output current depends on the type of detector and the nature and energy of the radiation. The currents of shortest duration (0.1 to 10 ns) occur in plastic scintillators and thin-depletion-depth semiconductor detectors; the longest (0.1 to 5 μs) occur in

Figure 8.25. Block diagram of basic instrumentation system.

gas-filled detectors.* The current duration in scintillators, such as sodium iodide and cesium iodide, is 0.25 to 1.5 μ s.

In semiconductor detectors, the energy conversion factors (W , the energy required to form an electron-hole pair) are 2.8 and 3.5 eV for germanium and silicon, respectively. Charge multiplication does not occur in semiconductor detectors. In scintillation counters, the effective W is 3 to 30 keV/electron, multiplied by as much as 10^5 to 10^8 by the photomultiplier. In gas-filled detectors, the conversion factor is 25 to 36 eV/ion pair. No multiplication occurs in ionization chambers, but multiplication factors of as much as 10^4 are common for proportional counter detectors. Thus, depending on the detector and the radiation energy, signal heights may vary from small values comparable to the noise level of the amplifier to high amplitudes at which amplifier noise is negligible.

*Ionization chambers and some special gas proportional counters have short electron charge collection times (10 to 100 ns).

Figure 8.26 shows various input couplings between the detector and the preamplifier. Because there is capacitance in the signal line to the preamplifier, these couplings are called AC. When high count rates are anticipated, direct coupling (DC) is usually preferred (see Section 8.5.1). Semiconductor detectors and some gas proportional counters require low to very low noise preamplifiers. Scintillation detectors do not require

Figure 8.26. Detector to preamplifier circuits. For the scintillation detector, I and II can be either current sensitive preamplifier or fast impulse amplifier.

preamplifiers; their output can be fed directly into amplifiers that accept narrow (impulse) pulses.

Low noise preamplifiers distort the pulse shape, which is then retrieved in the main amplifier by differentiation. Simple differentiation results in undershoot—a distortion of the amplitude of a pulse preceded by another that has not been completely integrated by the preamplifier. Most amplifiers provide a compensating network (pole zero compensation = P.Z.C.) (Ref. 16) which, if correctly adjusted, completely eliminates the undershoot. Because P.Z.C. is necessary at high counting rates, the P.Z.C. network in the main amplifier should be carefully adjusted for the contemplated count rate range. Figure 8.27 shows typical pulse shapes from detectors, preamplifiers, and amplifiers.

Figure 8.27. Detector, preamplifier, and amplifier pulse shapes.

8.4.2 Pulse Shaping and Pile-up

The output obtained from a scintillation detector, or the differentiated output from a preamplifier, is amplified and finally fed to a shaping network. This network assures that the pulse amplitude, at its output, is proportional to the area of the pulse at its input. Hence, the pulse amplitude at the output of the main amplifier will be proportional to the charge delivered by the detector, which in turn is proportional to the energy deposited in the detector.

Ideally, the output pulse of voltage preamplifiers will be a step function whose height is proportional to the charge collected at the detector. It decays with a long time constant (of the order of $50\ \mu\text{s}$), as is indicated in Figure 8.27. The rise time of the step is determined by the charge collection time of the detector. For very fast detectors (with rise times less than $50\ \text{ns}$), the inherent rise time (typically 30 to $100\ \text{ns}$) of the preamplifier, which is also affected by the detector's capacitance, is dominant.

At high count rates, the "piling-up" of the pulses on the long tail may cause the response to be nonlinear. To obtain the undistorted amplitude of the pulse, no other pulse should be present at the precise instant the amplitude is to be measured. Piling-up is a particularly serious problem in high resolution systems, such as Ge(Li) .

To reduce pile-up, the output of the preamplifier is first differentiated in the amplifier. This effectively removes the slow tail and produces a pulse whose amplitude is proportional to the initial rise of the preamplifier's pulse, and hence, to the detector's signal. The pulse can be shaped in various ways: by C-R, C-R-L or delay-line differentiation and R-C, R-L-C or delay-line integrating networks. Figure 8.28 shows some simple pulse-shaping circuits.

In general pile-up may be reduced by five means:

1. By choosing a pulse shape device which ensures that the output pulse returns to zero as fast as possible after having reached its peak. Several commercially available amplifiers use a particular filter which provides practically equal rise and fall time ("semi-Gaussian" shaping) to assure the closest to optimum performance obtainable with a linear time-invariant shaping network.
2. By making the shaped pulses as narrow as possible so that the probability of overlapping pulses is reduced. At very high counting rates, an improvement in resolution may be achieved by reducing the shaping time-constant to a value shorter than the one required for optimum signal-to-noise (S/N) ratio at low counting rates. Hence,

Figure 8.28. Pulse shaping networks and resulting pulse shapes (τ is the appropriate time constant, D_T is the time delay introduced by the delay-line).

there is a tradeoff between an optimal S/N ratio and the ability to count at high rates. As a rough guide, the following time-constants are recommended for semiconductor detectors: at low counting rates, 6.4 or 3.2 μs for high-capacitance detectors, and 1.6 μs for low-capacitance detectors. With cooled FET* preamplifiers the

*Field-effect transistor.

optimum time-constant is shorter than $1.6 \mu\text{s}$ and at medium and high counting rates, these values should be reduced by a factor of 2 to 4.

3. By using a pile-up rejection (PUR) circuit, the effect of residual pile-up may be further reduced. This circuit prevents those pulses whose amplitudes have been distorted by pile-up from reaching the spectrum analyzer. Obviously, if the level of pile-up is high, for example, with a very high count rate at low energies, the PUR will be busy most of the time in rejecting pulses and very little useful information on the desired low-intensity, high-energy lines will be accumulated. This is the basic deficiency of the conventional PUR.
4. By DC coupling the entire system or using baseline restoration. The latter is essentially a nonlinear filter that heavily attenuates low-frequency components of the signal delivered by the main amplifier, thus partially eliminating the slow tail which may be left by incomplete P.Z.C.
5. By using a time-variant filter which responds to both the shape and the width of the pulses instead of the one parameter measured by the Gaussian (time-invariant) filter. The time variant method can probably give the best resolution for high count rate cases (Ref. 17), but it is more complicated. Although of limited usefulness in ANDA, it could be useful in passive γ -ray NDA.

It must be stressed that none of the above pile-up reducing techniques alone will suffice; only a judiciously chosen combination of the available methods will achieve optimum results.

8.4.3 Systems for Counting and Energy Spectrometry

Figure 8.25 shows a block diagram of a simple counting system. All pulses exceeding the energy level chosen by the integral discriminator will be counted. Such a system is useful for counting a single energy line when no other lines are present (for example, in gas counters). The discriminator simply suppresses the counts originating from noise.

A more advanced system is shown in Figure 8.29A. Here the integral discriminator of Figure 8.25 is replaced by a single channel analyzer that counts all pulses whose height falls within a certain range. This system can count the intensity of a single energy line in the presence of others. By selecting the proper channel width and by gradually advancing the value of the baseline, one can obtain a complete spectrum. This procedure is time-consuming, but often suffices, especially in the field, where multichannel analysis is not available.

Figure 8.29. Block diagrams of counting systems.

Figure 8.29B shows a system that employs a multichannel analyzer (MCA). The MCA consists of an analog-to-digital converter (ADC) that converts the peak amplitude of incoming pulses into a digital number that is stored by a digital memory in a “channel.” The data can be retrieved later in a suitable form for display of the recorded spectrum on a cathode ray tube and/or for storage for further analysis on a permanent medium such as punched tape, magnetic tape, disc, and others. MCA spectra have been shown in Figures 8.19, 8.21, and 8.24. Figure 8.29C demonstrates the additional use of a window amplifier, which focuses on only a certain region of the energy spectrum, thereby greatly reducing the waste of memory space on channels that would otherwise record only insignificant information, if any.

8.4.4 Pulse Shape Discrimination

In addition to pulse height discrimination in scintillators used for detecting neutrons, it is often desirable to discriminate against γ -ray

pulses by other means. γ -Ray pulses have often shorter decay times than neutron pulses because their secondary particles are electrons rather than heavy, charged particles. The difference in decay times can be used to reject pulses caused by γ -rays. This method is known as pulse shape discrimination (PSD).

Figure 8.30 shows the principle of one type of PSD circuit (Ref. 5). The pulse is delayed about 50 ns, which is shorter than the decay time of a neutron pulse and longer than the decay time of a γ -ray pulse. The delayed pulse is counted in coincidence with the original pulse. As a result, there will be a coincident pulse, if the event is caused by a neutron. These kinds of circuits can reduce the γ -ray background in neutron scintillation detectors by a factor of 10 or more. PSDs have also been used in ANDA systems that count fast neutrons from fissions stimulated by low-energy (sub-MeV) neutrons, for example, from Sb-Be sources (see Section 9.3.1).

8.4.5 Detector and Electronic Stability

Stability of the detection system is essential. The most important cause of instability, temperature variation of the detector, can be mitigated by keeping the detector at constant temperature. Temperature instability is particularly serious for NaI detectors and fast scintillators. Ge(Li) detectors are kept at cryogenic temperature, and in some cases, their preamplifiers are also cooled.

Although gas proportional counters, especially BF_3 and ^3He , are temperature-insensitive, they often perform poorly in high humidities

Figure 8.30. Principles of a pulse shape discriminator circuit.

because of high-voltage leakage and breakdown across insulators. In addition, gas proportional counters undergo long-term deterioration due to radiation damage and depletion of the neutron-absorbing isotope. For instance, ^3He tubes at 4 atm pressure have been found to deteriorate in a neutron flux of 10^7 to 10^8 n/cm²/s after a few hundred hours of operation (Ref. 18). This phenomenon may result from the buildup of electronegative gaseous products that capture the drifting electrons thus, reducing the pulse amplitude. To prevent the buildup, a layer of activated charcoal that absorbs these gases is placed on the tube cathode. After 1000 hr, the gas multiplication factor of charcoal-coated tubes drops about 10 percent, but there is no deterioration of the shape of the pulse height spectrum. These tubes also have better resolution than uncoated tubes (Ref. 18).

Count rate is still another cause of instability: some photomultipliers show gain characteristics that depend on the count rate. Several methods exist for stabilizing such systems. If the count rates are not too high (for example, less than 100,000 cps) and good resolution detectors are used (NaI), stabilization may be accomplished by circuits that sense the amplitude drift of a prominent γ -ray peak and change either the amplifier's gain or the high voltage of the photomultiplier. These spectrum stabilizers, commercially available, have proven reliable in the field. Sometimes the peak that the stabilizer responds to is an artificial one, provided either by an α -particle emitter "seeded" in the crystal or by a constant intensity light-emitting diode (LED). In either case, the pulse height of the artificial peak should be well above the energy region of the detected γ -rays to prevent undesirable interferences. For applications with count rates at about 100,000 cps, resolution of instability problems is discussed in Section 8.5.

8.4.6 Deadtime Corrections

All detectors and their associated electronics are subject to errors due to "deadtime," the interval following the detection of a pulse during which a succeeding pulse cannot be detected or recorded. Both detectors and electronics have deadtimes. For example, in a gas-proportional counter, a second event may not be detected while the ionization process of a first event is taking place. Here, the detector is said to be fully or partially "paralyzed." Deadtimes in electronics occur because of the width of the pulse or because it takes time for a circuit to recover from a previous pulse. The deadtime of the system (detector plus all electronics) is equal to the longest deadtime of any of its components. Deadtimes range from less than 1 ns in fast scintillator detectors to ~ 100 μs in some gated electronic circuits.

It is always necessary to correct observed count rates for the counts presumed to have been missed during the system deadtime. A simple correction formula can be derived as follows: let the deadtime following each detected event be τ (s) and the average observed count rate (cps) be N_o . The expected total deadtime per second is $N_o\tau$, and during this time $N_o(N_o\tau)$ events are presumably missed. Therefore, the true (corrected) count rate, N_t , is given by

$$N_t = N_o + N_o^2\tau = N_o (1 + N_o\tau). \quad (8-3)$$

A more rigorous derivation yields

$$N_t = N_o / (1 - N_o\tau) \quad (8-4)$$

which agrees with Eq. 8-3 if $N_o\tau \ll 1$ (low count rates). Eq. 8-3 and 8-4 assume total paralysis of the system during deadtime. Different formulas replace Eq. 8-4, if partial paralysis is assumed, but these also reduce to Eq. 8-3 at low count rates. For some gated circuits (see Section 8.6), the correction formulas can be much more complicated and the corrections much larger.

As a numerical illustration: if τ is $0.1 \mu\text{s}$, the deadtime correction $N_o\tau$ will be 1 percent, if N_o is 10^5 cps, and 10 percent, if N_o is 10^6 cps. For τ at $100 \mu\text{s}$, 1 percent correction is required at an N_o of 100 cps and 10 percent at an N_o of 1000 cps.

8.5 HIGH-COUNTING RATE PROBLEMS IN ANDA

In many ANDA applications, it is essential to be able to count at very high rates. High counting rates can stem from large γ -ray backgrounds in measurements where neutrons are used as a signature (for example, in spent fuel rod measurements), or in measurements of photofissions. High counting rates also arise from the high transient source neutron intensity of 14-MeV or photoneutron sources in delayed neutron or γ -ray measurements. Finally, when high accuracy is desired in a very short measuring time, very high counting rates are required to yield statistically significant results. This is the situation in high-speed fuel rod scanners (see Section 9.6).

Fast detectors, such as scintillators, ionization chambers, and coated-cathode (for example, ^{10}B) proportional counters, can provide the capability to accurately measure high count rates. The following discussion will be limited to scintillation detectors. A special high counting rate ANDA application of BF_3 and ^{10}B -coated detectors in which the γ -ray flash from a Linac is canceled by subtracting detector signals is discussed in the literature (Ref. 19).

8.5.1 Pulse Counting with Fast Organic Scintillators

The fastest possible counting can be obtained by plastic scintillators with fast photomultipliers. Count rates in excess of 10^7 pulses per second can be achieved without major difficulties. The basic circuit, shown in Figure 8.26B, is usually followed by a fast, nonshaping amplifier and a fast discriminator. The purpose of the amplifier is to allow the photomultiplier tube (PMT) to operate at lower gain (that is, lower high voltage) and hence, lower average anode current, which improves the stability of the photomultiplier. The stability of the voltages between the dynodes is also important. To assure that these voltages remain constant, regardless of the count rate and the pulse amplitudes, one can replace the voltage distribution resistors of some of the stages by Zener diodes, which give a fixed voltage drop regardless of current. A simpler approach is to connect the last dynodes to ground by relatively large capacitors (typically $0.01 \mu\text{F}$), as is shown in Figure 8.26B. The capacitors limit the variations of the voltage across the resistors when large, individual pulses occur.

Usually, stable operation of photomultipliers requires that the current flowing across the high voltage divider-string is at least a factor of 100 higher than the average anode current. The approximate anode current can be computed from the measure total count rate, and the shape of an average pulse, which can be obtained by observing the signal on a fast oscilloscope. Two such pulse shapes are shown in Figure 8.31. The first was obtained from a ^{22}Na (511- and 1275-keV) γ -ray source and a 15.2-cm x 15.2-cm cylindrical plastic scintillator (NE-102A). The second was obtained from an Am-Li neutron source and a long (15.2-cm x 61-cm) NE-102A scintillator. A fast photomultiplier tube (XP-1040) with a rise time of 2 ns was used in the first case; the second used a "slow" PMT, EMI-9530K, with a rise time of a little less than 40 ns.

Figure 8.31. Anode pulse shapes for two sources, two sizes of a plastic scintillator, and two types of PMT's.

The area of each of the triangles shown in Figure 8.31 multiplied by the average count rate gives a rough approximation to the average anode current. For 10^5 pps, the current from the 15.2 cm x 15.2 cm scintillator was 26 μ A. For the same count rate, the current in the larger scintillator exposed to the neutron source was about 260 μ A. Obviously, the reason for the much higher current in the second case is the larger width of the pulse, probably caused by the multiple scattering the neutron undergoes as it is slowed down in the large hydrogen-containing scintillator. For stable operation, the average PMT current should not significantly exceed 1 μ A. Hence, for the long scintillator, amplifying the signal gain by a factor of 10 to 50 and reducing the high voltage, lowers the average current to the desired level without loss of sensitivity.

The fast integral discriminator (Figure 8.25) that follows the amplifier serves to standardize the pulses, to eliminate noise pulses, and, if desired, to provide some rough discrimination among energies. The energy discrimination is coarse because of the poor pulse height distribution obtained by plastic scintillators (see Figures 8.17 and 8.21).

8.5.2 Fast Pulse Counting with Sodium Iodide (NaI) Detectors

Inorganic scintillators, of which NaI(Tl) is the most common, have relatively long decay time constants, between 250 and 1000 ns. One exception is the CsF detector. Although it has a short decay time (~ 5 ns), it has very low light-emitting efficiency. Hence, it exhibits very poor energy resolution, although it is somewhat better than organic scintillators. The long decay time of the light pulse in inorganic scintillators is a drawback for high counting rate applications. However, their high sensitivity, efficiency, and good energy resolution make these detectors, especially NaI(Tl), very attractive even for applications with high count rates. There are two ways in which NaI(Tl) detectors are used: discrete pulse counting and current measurements. In pulse counting, the long decay time, 250 ns for NaI, causes superposition of pulses on the trailing edges of the previous pulses. In order to reduce this pile-up, one can differentiate the signal, thereby reducing the effective pulse width. Without differentiation, the signal takes about 1 μ s to fall to about 2 percent of its peak value. With proper differentiation, one can reduce this time to between 0.5 and 0.2 μ s. A moderate or substantial loss in energy resolution results because part of the scintillation light is rejected. However, even with strong differentiation, which allows count rates in excess of 500,000 cps, the energy resolution is quite often sufficient to afford good separation between background and signal.

An example of delay line differentiated ("clipped") signals from a 5.1-cm x 5.1-cm NaI(Tl) detector measuring ^{137}Cs (662 keV) and ^{241}Am (60 keV) γ -ray lines is shown in Figure 8.32. The figure shows

Figure 8.32. Delay line (50 ns) differentiated signals from NaI(Tl) detector [2 in. (5 cm) $\times$ 2 in. (5 cm) crystal with RCA 8850 PMT and fast amplifier] counting 300,000 cps.

that in spite of the high count rate (300,000 cps) one can easily separate the low and high energy lines by placing the discriminator level at the gap between the lines, that is, around 100 mV. Here, gain shifts of less than 25 percent will not greatly change the observed count rate because of the gap width. The presence of oscillations on the trailing edge of the pulse in Figure 8.32, due to poor impedance matching, underlines the importance of proper termination and impedance matching in fast counting systems.

8.5.3 Current Measurements

Discrete pulse measurements with NaI(Tl) are limited, in practice, to count rates of less than 500,000 cps. Thus, if higher counting rates are desired, the detection of individual pulses must be foregone and measurement of the average current must be substituted. The main advantage of the current measurement technique is its virtual independence of counting rate. It applies to low counting rates, if the average current is above the background current ("dark-current") as well as to count rates of many million pulses/second, regardless of the decay time characteristics of the scintillation. In the examples mentioned in Section 8.5.1, the average PMT was 26 and 260 μ A for the short and long scintillators, respectively. These currents may be reduced by a factor of 10 to 50 with proper preamplification. Currents of these magnitudes or even 1000 times smaller can easily be measured by DC coupling the PMT anode to a current integrator (for example, Ortec Model 439 Digital Current Integrator), which replaces the current-sensitive preamplifier in Figure 8.26B. Figure 8.33 shows this arrangement.

Figure 8.33. Photomultiplier system with ground (DC-coupled) anode.

- (I) Current sensitive preamplifier or fast impulse amplifier for counting discrete pulses, or current integrator for current measurements.
- (II) Charge sensitive preamplifier or fast impulse amplifier.

In Figure 8.33 the anode operates at ground potential, and the cathode operates at the required negative high voltage. The absence of a capacitor in the anode line also makes this coupling the best one for discrete high pulse rate applications. The great sensitivity of the integrator makes the PMT more stable by reducing the PMT gain (by lowering the high voltage).

One source of long term instability in inorganic scintillators is the phosphorescence created in the glass envelope of the PMT by the strong light generated in the scintillator. The intensity of this light is high enough to cause phosphorescence only when a highly efficient crystal like NaI(Tl) is exposed to strong radiation fields, for example, in high-speed fuel rod scanners (see Section 9.6). Phosphorescence is also a function of the type of glass used. Because it appears as a low constant current, it does not affect discrete pulse measurements, however, it does affect current measurements. By keeping the average radiation field constant, one can account for the effect of phosphorescence.

It should be recalled that the quantity that is measured by current integration is different from the quantity of discrete pulses. In pulse counting, one measures the number of pulses above a certain energy or pulse height level or within an energy band per unit time. In current measurement, the integral of the product of the number of pulses times their pulse height (or intensity of the emitted light) is measured. Figure 8.34 compares these two quantities and shows the pulse height distribution owing to delayed γ -rays (Figure 7.15), $\phi(E)$, and the

Figure 8.34. Delayed γ -ray pulse height and energy deposition distributions in 3-in. (7.5-cm) $\times$ 3-in. (7.5-cm) NaI (Tl) detector (30 s irradiation and measuring time).

spectrum of energy deposited by these particles in the detector, $E\phi(E)$.* The discrete pulse rate, C , at the output of the discriminator with a bias d , is

$$C = \int_d^{E_{\max}} \phi(E) dE. \quad (8-5)$$

The measured current, I , is proportional to

$$I \propto \int_0^{E_{\max}} E\phi(E) dE. \quad (8-6)$$

*Note that we use the symbol of energy, but actually what we have is pulse height distribution, which has been calibrated in terms of the energy of monoenergetic calibration γ -ray sources.

One can define the average energy, $\bar{E}$, for current measurement as follows:

$$\bar{E}(\text{current}) = I/C. \quad (8-7)$$

The average spectrum energy, as measured by current from the delayed γ -rays, is about 740 keV, substantially higher than the mean energy of the discrete pulses (about 500 keV in the present case). Current measurement does not provide any means of controlling background or spectral response, except by γ -ray filters that attenuate the low-energy part of the spectrum.

The statistical uncertainties associated with discrete pulse and current measurements are considerably different. For the former, the variance is given by

$$(\delta C)^2 = \int \phi(E) dE = C \quad (8-8)$$

and for the current measurements the variance is given by

$$(\delta I)^2 \propto \int E^2 \phi(E) dE \neq I. \quad (8-9)$$

Hence, one cannot estimate statistical uncertainty from a current measurement alone by taking the square root of the total count. This quantity can, however, be determined empirically by repeating the measurement several times and computing the standard deviation.

In summary, the direct current measurement of a scintillator detector offers a counting system totally independent of count rate and deadtime. As long as the spectrum of the measured radiation does not significantly change during the measurement, the measured current is proportional to the count rate of the incident radiation.

8.6 COINCIDENCE COUNTING METHODS

Several ANDA methods are based on coincidence counting techniques, that is, the nearly simultaneous detection, in two or more detectors, of radiation emitted from an event. There are, on the average, about 2.5 to 3 prompt fast neutrons and 7 to 8 prompt γ -rays with an average energy of ~ 1 MeV emitted in each fission. This large multiplicity of emitted radiation makes coincidence counting an attractive method for detecting fission events. Coincidence systems designed for this purpose are called fission multiplicity detectors (FMDs). Such coincidence counting schemes commonly employ two to four detectors, which may respond either to neutrons or to γ -rays, or to both. The longest interval, within which two or more such detected events result in an output coincidence,

is called the resolving time of the system. If the resolving time is less than about 100 ns, the system is referred to as fast, if the resolving time is much greater (up to $\sim 100 \mu\text{s}$ in some applications), the system is slow. The resolving time, or coincidence time, should not be confused with the deadtime (see Section 8.4.6).

In order to count prompt fission radiations in ANDA applications, fast coincidence systems are usually used. These give a high signal-to-background ratio. The background consists of accidental, or random, coincidences that result from detection within the resolving time of two or more radiations which do not originate from a common event. The rate of accidental coincidences is proportional to the resolving time, τ , for twofold coincidences (or to τ^{n-1} for n -fold coincidences) so that short resolving times are necessary for low accidental coincidence backgrounds. Fast coincidence systems are described in the following sections.

Coincidence systems that detect prompt neutrons, after they have been slowed down to thermal or near thermal energies, use slow coincidence circuits, since the slowing-down and diffusion times in the detectors are $\sim 100 \mu\text{s}$. This technique (see Section 8.6.2), is much more common in passive NDA methods that detect spontaneous fissions in ^{240}Pu , however, one active system based on the same principle has been designed (see Section 9.5.6).

8.6.1 Fast Coincidence Counting

A typical ANDA fast coincidence system has up to four detectors, and an output pulse will be recorded if any two, three, or all four detectors (at the choice of the operator) sense an incoming neutron or photon within the resolving time of the system. The detectors used are often plastic scintillators (NaI is also possible), which have rapid responses to both fast neutrons and γ -rays (see Tables 8-3 and 8-6). The choice of the coincidence level is dictated by the background level. Generally, the fission signal-to-background ratio improves considerably when higher levels of coincidence are used, but at the same time the signal level declines substantially. Often the best compromise is to use 3-out-of-4 coincidences. Since a typical fission will emit about from 2.5 to 3 neutrons and 7 or 8 photons, the chances of detecting two or more of these are good, if the detectors' efficiencies are high enough.

There are two types of circuits that accept coincidence pulses from detectors and reject those that are not in coincidence: "gates" and "summers." Gates are opened by the first of the coincident pulses to be received and then pass the other coincident pulses. They are open for a preset interval, usually about 50 ns for fast coincidence circuits or 10 to

100 μ s for slow circuits. Summers add the pulses: if the pulses overlap within their respective pulse widths, they generate an output pulse of amplitude equal to the sum of the individual amplitudes and width equal to the overlap time. A suitably biased discriminator will pass the overlap (summed) pulse but reject the smaller nonoverlapping pulses. The principles of both types of systems are shown in Figures 8.35 and 8.36.

Figure 8.37 shows a schematic diagram of the fast coincidence system used in Isotopic Source Assay System (ISAS) (see Section 9.5). The circuit shown contains basically most attributes of fast coincident NDA electronics. After pulse height discrimination and shape standardization, the signals from the four detectors are fed to the logic* mixer (Figure 8.38) whose outputs correspond to the desired multiplicity (2-out-of-4, 3-out-of-4, or 4-out-of-4). The logic mixer plus the second quad discriminator provide for the majority coincidence using the principle demonstrated in Figure 8.36. The photomultiplier's signals are also fed to the linear mixer and delay (Figure 8.39). The output of the logic mixer opens a gate that passes the output from the linear mixer. The signal from the linear mixer is delayed to arrive at the linear gate concurrently with the signal of the majority coincidence. The linear mixer actually sums the pulses from the four scintillators.

Figure 8.35. An AND gate as a coincidence circuit.

*Linear pulses are analog pulses that maintain the original shape of the detector pulses. Logic pulses are pulses with constant standard shape. The linear and logic mixers are electronic circuits that mix linearly the linear and logic pulses, respectively.

Figure 8.36. Principles of "Summer" coincidence circuits.

The linear sum that gets through the linear gate, which is opened only if the desired coincidence (or majority) level is obtained, is related to the total energy deposited by the nuclear signature in the detectors. Since the total prompt photon energy from the fission event is usually higher than the total energy released by other nuclear processes [including (n, γ) neutron capture], the measurement of the total energy deposited in the detectors affords another degree of discrimination against nonfission background. This fact is demonstrated in Figure 8.40 that shows the normalized total pulse height distribution of a four-detector ISAS for 3-out-of-4 coincidences for a small fission source (^{252}Cf) and for the background induced by a neutron source placed in the ISAS source-collimator. The pulse height distribution from ^{22}Na (1.275 and 0.511 MeV) γ -ray source provides for energy calibration. Figure 8.40 shows that improved discrimination against background can be obtained by biasing the linear system (the amplifier in Figure 8.37) at around 1 MeV

Figure 8.37. Basic ISAS FMD electronics.

(about channel 90). Obviously, such a high bias will cause substantial loss in signal. Thus, the use of the linear channel is recommended when the presence of elements with high-neutron capture cross sections and associated high prompt γ -ray emissions (for example, chlorine) is expected.

Figure 8.39. Schematic of a linear passive mixer (ISAS).

Figure 8.40. Pulse height distribution of total energy deposited in a 4-detector FMD for 3-out-of-4 coincidence events.

Several different slow coincidence circuits have been designed and built. One is simply a gate which is open for a duration of the order of the diffusion time in the thermal neutron detector. A detected neutron opens this gate, which then passes any succeeding neutron detected while the gate is open. An identical gate, delayed by several hundred μs from the first, passes signals from neutrons that do not originate in the fission event and can, therefore, be used to determine the accidental coincidence rate. This method requires significant corrections for deadtime (Ref. 20). In another circuit, shift register logic (Ref. 21) is used. Here, each neutron pulse is stored in a shift register and is shifted at a fixed rate (for example, $1/\mu\text{s}$) for a time corresponding to the length of time the gate is open (τ). If a subsequent pulse(s) arrives while the first is in the register, coincidences are recorded. These coincidences are registered on a scaler, at a rate R . In effect, each incoming pulse causes the scaler to monitor the pulses stored in the shift register. All pulses are also recorded in a second ordinary scaler, this count rate is G . It can be shown that the fission rate is proportional to $(\tau G^2 - R)$. If, only accidental (uncorrelated) coincidences are present, $R = \tau G^2$. The smaller corrections, for deadtime necessary for the shift register circuit relative to the simple gate circuit, permit much higher count rates in the shift register.

A simple slow-coincidence circuit consists of two scalers, one of which registers all counts, except for those missed during its short deadtime. The other is constructed with an artificially long deadtime, approximately equal to the neutron diffusion time in the detector. It will, therefore, not detect the subsequent pulses (after a given pulse) due to the correlated neutrons. The differences between these two scalers, corrected for their respective short and long deadtimes, is to first order proportional to the fission rate (Ref. 22). The circuit, known as the variable deadtime circuit (VDC) has been found vulnerable to changes in the rate of accidental coincidences during the measurement, since it measures only the average accidental rate during the run.

Table 8-7 shows a comparison of the circuits discussed above for an assay of a 483-g Pu sample under conditions of high, low, and variable background (accidental) rates (Ref. 23). The poorer performance of the VDC, especially in run No. 3, where the background rate was changed, is evident. The "updating" circuit listed in Table 8-7 is basically the first circuit described above, with two successive gates for "real" and "accidental" counts.

8.7 COINCIDENCE EFFICIENCY

One of the most important attributes of fast coincidence techniques for ANDA is their ability to detect fission events with high efficiency. It

Table 8-7. Intercomparison of thermal neutron coincidence circuits for variable background rates.^a

Run	Average Total Rate 10 ³ cps	VDC Assay (g)	Shift Register Assay (g)	Updating Circuit Real-Time Assay (g)	Updating Circuit Livetime Assay (g)
1	1.6	456 ± 1	483 ± 3	472 ± 2	471 ± 3
2	12.0	446 ± 5	482 ± 10	477 ± 15	480 ± 4
3 ^b	6.6	3098 ± 3	483 ± 4	481 ± 7	479 ± 4

^aThe standard deviation was obtained by repeating each run three times.

^bFor run 3, the random background rate was changed by more than a factor of 10 during the measurement.

is probably this feature that made them the most usable techniques for ANDA. The efficiency of a coincidence system is the product of the efficiencies of the individual detectors. Hence, any change in detector size, the distance between source and detector, or the detector shielding will greatly affect the efficiency of coincidence detection. For example, assume the geometrical efficiency, ϵ_g (that is, the solid angle subtended by the fissile or fertile material at the detector) of each detector in a four-detector FMD is changed by increasing the distance of the four identical detectors from the sample. Then the efficiency of detecting a 4-fold coincidence, which is proportional to ϵ_g^4 , will change accordingly. Thus, a reduction of 10 percent in the efficiency of each individual detector will reduce the efficiency of a typical 4-fold coincidence system by 34 percent. The calculation of the individual detector efficiency and, for that matter the coincidence efficiency, is generally quite difficult. Among other variables, it should take into account the distribution of fissions in the sample, the presence of matrix materials, the density of the materials, the geometric configuration, the size and shape of the detector, and the energy bias. Such calculations are possible for simple situations, for example, a point or line fission source and spherical or cylindrical detectors with zero bias.

Because of the complexity of the configuration, the Monte Carlo technique is the most appropriate for determining detector efficiency. This technique is used to calculate the efficiency of an ISAS like FMD, as shown in Figure 8.23. However, it is much more practical to measure the detection efficiency of individual detectors and the entire coincidence system in actual applications. In the following section, coincidence detection efficiency will be derived on the assumption that the detection efficiencies of individual detectors comprising the coincidence instrument are known and equal. The analytical expressions for

the efficiency of the FMD, allow qualitative determination of the following effects on detection efficiency:

1. Variation of the effective fission multiplicity (for example, by absorption of a portion of the prompt γ -ray spectrum) and the efficiency of individual detectors.
2. Change in the number of detectors.

8.7.1 FMD Efficiency and Multiplicity-Dependence

A general analytical expression for the efficiency of an FMD is given in Appendix A. For an FMD with four identical detectors and a fission event of average multiplicity, ν , the coincidence efficiencies, $\epsilon_{\ell/L}$, are given by the following equations:

$$\epsilon_{1/4} = 1 - (1 - 4\epsilon)^\nu \quad (8-10)$$

$$\epsilon_{2/4} = 1 - 4(1 - 3\epsilon)^\nu + 3(1 - 4\epsilon)^\nu \quad (8-11)$$

$$\epsilon_{3/4} = 1 - 6(1 - 2\epsilon)^\nu + 8(1 - 3\epsilon)^\nu - 3(1 - 4\epsilon)^\nu \quad (8-12)$$

$$\epsilon_{4/4} = 1 - 4(1 - \epsilon)^\nu + 6(1 - 2\epsilon)^\nu - 4(1 - 3\epsilon)^\nu + (1 - 4\epsilon)^\nu. \quad (8-13)$$

If the efficiency of a single detector is small, that is, $\epsilon \leq 1$ percent, the expressions above reduce to a simpler form:

$$\epsilon_{\ell/L} \cong \frac{L!}{\ell!(L-\ell)!} \frac{\nu!}{(\nu-\ell)!} \epsilon^\ell \quad (8-14)$$

where ℓ is the degree (or "fold") of the coincidence and L is the number of detectors. $\epsilon_{\ell/L}$ signifies the probability of detecting any coincidences between ℓ detectors (ℓ -fold coincidence) out of all L detectors of the system.

For the case of a two-detector FMD, the efficiency is equal to

$$\epsilon_{2/2} \cong \nu(\nu-1)\epsilon^2. \quad (8-15)$$

For a three-detector FMD,

$$\epsilon_{2/3} \cong 3\nu(\nu-1)\epsilon^2 \quad (8-16)$$

$$\epsilon_{3/3} \cong \nu(\nu-1)(\nu-2)\epsilon^3. \quad (8-17)$$

For a four-detector FMD,

$$\epsilon_{2/4} \cong 6\nu(\nu-1)\epsilon^2 \quad (8-18)$$

$$\epsilon_{3/4} \cong 4\nu(\nu-1)(\nu-2)\epsilon^3 \quad (8-19)$$

$$\epsilon_{4/4} \cong \nu(\nu-1)(\nu-2)(\nu-3)\epsilon^4. \quad (8-20)$$

The dependence of efficiencies for $\epsilon = 0.2$ as given in Eq. 8-10 through 8-13 are listed in Table 8-8. The dramatic decline in the high-fold coincidence efficiency as a result of multiplicity reduction ($\delta\nu$) is quite obvious. When the efficiency of the single detector is low, that is, $\epsilon \leq 0.01$, Eq. 8-14 through 8-20 can be used. These results are shown in Table 8-9. The relative (normalized) coincidence efficiency becomes independent of ϵ . The relative efficiency increase from the minimum value (which is quite small for $\epsilon = 0.01$ as indicated in the table) as a function of the increasing multiplicity is even more dramatic in this case.

The FMD efficiency based on Eq. 8-10 and 8-11, as a function of single detector efficiency in the realistic range of less than 15 percent (typical of FMDs accommodating samples larger than 1 gal. canisters) for different multiplicities, is shown in Figures 8.41 and 8.42. The relatively higher sensitivity of 2-out-of-4 versus 3-out-of-4 for different multiplicities and single detector efficiencies is shown clearly in Figure 8.43 (Ref. 24). As the single detector efficiency increases, the efficiency advantage of 2-out-of-4 over 3-out-of-4 declines.

Table 8-8. FMD efficiency for single detector with 0.2 efficiency.

Multiplicity ν	Multiplicity Change $-\delta\nu$	Efficiency		
		2/4	3/4	4/4
8	0	0.997	0.904	0.427
6	2	0.993	0.753	0.215
4	4	0.902	0.422	0.038
3	5	0.768	0.192	0
2	6	0.480	0	0

Table 8-9. Relative FMD efficiency for a single detector with a low efficiency ($\epsilon < 1\%$).

Multiplicity ν	Multiplicity Change $-\delta\nu$	Efficiency Normalized to the Lowest Possible Multiplicity		
		2/4	3/4	4/4
8	0	28	56	70
7	1	21	35	35
6	2	15	20	15
5	3	10	10	5
4	4	6	4	1 ^a
3	5	3	1 ^a	0
2	6	1 ^a	0	0
1	7	0	0	0
0	8	0	0	0

^aThe absolute efficiency (for $\epsilon = 0.01$) is $6 \cdot 10^{-4}$, $2.4 \cdot 10^{-5}$, $2.4 \cdot 10^{-7}$ for 2/4, 3/4, and 4/4, respectively.

Figure 8.41. 2-out-of-4 coincidence efficiency vs. single detector efficiency for different multiplicities.

The tables and figures in this section illustrate amply the strong dependence of FMD efficiency on the observed fission multiplicity and the efficiency of the single detector. It is, therefore, very important for the designer and user of an FMD-ANDA system to carefully consider any measures he takes that may reduce them. The effective multiplicity is reduced by placing a γ -ray filter between sample and detectors to suppress the low-energy prompt γ -rays that are more susceptible to self-absorption (see Chapter 9). Because this technique results in a substantial efficiency reduction, it should be used only if *significant* γ -ray self-absorption is anticipated (for example, if a large sample of high density UO_2 is measured). The multiplicity may also be decreased if poorly designed electronics are used, which require high discrimination levels against noise, thus eliminating low-energy pulses from neutrons or

Figure 8.42. 3-out-of-4 coincidence efficiency vs. single detector efficiency for different multiplicities.

photons. The single detector efficiency is reduced in an FMD, if the sample cavity is enlarged to accommodate larger containers. Thus, selection of containers for ANDA samples should be carefully considered if detection sensitivity and measurement time are important.

8.7.2 Effect of the Number of Detectors

Eq. 8-14 describes the effect on efficiency of the number of detectors, L , in an FMD. To demonstrate the gain in efficiency, if the number of detectors in the FMD is increased, let us assume that we split each of the existing detectors into k detectors. Thus, the overall FMD efficiency for single events is unchanged:

Figure 8.43. Efficiency of the fission multiplicity detector (FMD) as a function of the multiplicity of prompt-fission γ -rays.

$$\epsilon_{\ell/kL} \cong \frac{(kL)!}{\ell! (kL - \ell)!} \frac{\nu!}{(\nu - \ell)!} \left(\frac{\epsilon}{k}\right)^\ell. \quad (8-21)$$

It can be shown that for $k \geq 1$, $\epsilon_{\ell/kL} \geq \epsilon_{\ell/L}$, therefore, the FMD efficiency increases by splitting the individual detectors into smaller detectors. The reason for this behavior is apparent: two particles that enter one detector in the original configuration and, therefore, do not register as a coincidence, however, can now enter two detectors and register as a coincidence.

The maximum theoretical improvement in detection efficiency is given when $k \gg 1$;

$$\lim_{k \rightarrow \infty} \left[\epsilon_{\ell/kL} / \epsilon_{\ell/L} \right] \cong \lim_{k \rightarrow \infty} \frac{(kL)!}{(kL - \ell)!} \frac{(L - \ell)!}{L!} \frac{1}{k^\ell} = L \frac{\ell(L - \ell)!}{L!}. \quad (8-22)$$

Table 8-10 shows the gain in FMD efficiency obtained by "splitting" the detectors. The main gains are in measurement of high-fold coincidences, where improvement is generally needed, since the efficiencies are indeed low.

The improvement of detection efficiency by splitting detectors, or better, by adding more of them, rests, of course, on the assumption that the multiplicity is large. If fission multiplicity is substantially reduced, as discussed previously, the gains in efficiency will be much smaller than those shown in Table 8-10. Furthermore, splitting the detectors into many small detectors (each with its own PMT and fast discriminator) obviously creates practical difficulties. Nevertheless, one can get an idea of the degree of improvement to be expected; for example, a three- or four-detector system is generally better than a two-detector system.

Table 8-10. Ratio of FMD efficiency between split and unsplit FMD.

FMD		Detectors, Original No. (<i>L</i>)	Coincidence Level (<i>q</i>)	Split Factor	Efficiency Ratio
Split	Unsplit				
$\epsilon_{2/4}$		2	2	2	1.5
$\epsilon_{2/6}$	$\epsilon_{2/2}$	2	2	3	1.67
$\epsilon_{2/\infty}$		2	2	∞	2
$\epsilon_{2/6}$		3	2	2	1.25
$\epsilon_{2/9}$	$\epsilon_{2/3}$	3	2	3	1.33
$\epsilon_{2/\infty}$		3	2	∞	1.5
$\epsilon_{3/6}$		3	3	2	2.5
$\epsilon_{3/9}$	$\epsilon_{3/3}$	3	3	3	3.11
$\epsilon_{3/\infty}$		3	3	∞	4.5
$\epsilon_{2/8}$		4	2	2	1.17
$\epsilon_{2/12}$	$\epsilon_{2/4}$	4	2	3	1.22
$\epsilon_{2/\infty}$		4	2	∞	1.33
$\epsilon_{3/8}$		4	3	2	1.75
$\epsilon_{3/12}$	$\epsilon_{3/4}$	4	3	3	2.04
$\epsilon_{3/\infty}$		4	3	∞	2.67
$\epsilon_{4/8}$		4	4	2	4.38
$\epsilon_{4/12}$	$\epsilon_{4/4}$	4	4	3	6.11
$\epsilon_{4/\infty}$		4	4	∞	10.67

8.8 SELF-SHIELDING AND FISSION MULTIPLICITY

In the previous section we dealt with detector efficiency that is described by a single parameter, ϵ (see Section 8.7). Actually, the detector efficiency is a very complex function of the sample's geometry, (size, shape, and position relative to the detector) and the detector's geometry. It is also a function of the energy of the emitted particles, the sample's self-shielding, and the detector's material. Accurate calculation of the single detector efficiency or the FMD's efficiency is, in general, very difficult. However, analysis of a general expression for detector efficiency and taking into account properties of its components shed light on some very useful properties of the coincidence efficiency. The most notable is the reduction of self-shielding in the sample (Ref. 25).

General expressions for coincidence efficiencies, incorporating the previous factors, are discussed in Appendix B. It is shown that to a first order, for a point fission source (for example, small fissile sample) the ratio of the product of the count rate of each individual detector (N_1 and N_2 , in a two-detector FMD) to the coincidence count rate (N_{12}) is independent of the efficiency of the single detectors:*

$$N_1 N_2 / N_{12} = \left[(\bar{\nu})^2 / \nu(\nu - 1) \right] N_f. \quad (8-23)$$

The ratio is equal to the fission source strength, N_f , multiplied by a physical constant, hence it is independent of geometrical or self-shielding effects. This result, which is an extension of the well-known nuclear physics method for the absolute determination of source intensity by β - γ rays coincidences, is important for ANDA since it allows, in principle, the determination of the fission rate independently of the effects of self-shielding. This formulation is rigorously true for small samples, but is also approximately valid, under some conditions, for the more general case.

Expressions similar to Eq. 8-23 can be derived for a three-detector FMD:

1. Using single and twofold coincidences,

$$\frac{N_i \cdot N_j}{N_{ij}} = \frac{(\bar{\nu})^2}{\nu(\nu - 1)} N_f \quad i \neq j = 1, 2, 3. \quad (8-24)$$

*Definitions of the various averages of the multiplicity ν are given in Eq. 7-3 and 7-4 and Appendix B.

2. Using two- and threefold coincidences,

$$\frac{N_{12} N_{13} N_{23}}{N_{123}^2} = \frac{(\overline{\nu(\nu-1)})^3}{(\overline{\nu(\nu-1)(\nu-2)})^2} N_f. \quad (8-25)$$

3. Using onefold and threefold coincidences,

$$\frac{N_1 N_2 N_3}{N_{123}} = \frac{(\bar{\nu})^3}{(\overline{\nu(\nu-1)(\nu-2)})} N_f^2. \quad (8-26)$$

The signal from an FMD is usually a majority-coincidence (for example, any 1-out-of-3, that is, $N_{1/3}$ or $N_{2/3}$ or $N_{3/3}$) rather than particular two- or threefold coincidences. One can relate the majority-coincidence signals, which are more numerous and statistically more significant, if the detector efficiencies and the various coincidence rates are equal, that is, if $N_i = N_s$ and $N_{ij} = N_d$ independently of i or j (for example, $N_d = N_{12} = N_{13} = N_{23}$):

$$N_i = N_s = \frac{1}{3}N_{1/3} + \frac{1}{3}N_{2/3} + \frac{2}{3}N_{3/3} \quad (i = 1, 2, 3) \quad (8-27)$$

$$N_{ij} = N_d = \frac{1}{3}N_{2/3} + \frac{1}{3}N_{3/3} \quad (i \neq j = 1, 2, 3) \quad (8-28)$$

$$N_{123} = N_{3/3}. \quad (8-29)$$

The left-hand side of Eq. 8-24, 8-25, and 8-26 can be replaced with N_s^2/N_d ; $N_d^3/N_{3/3}^2$ and $N_s^3/N_{3/3}$, respectively.

Expressions similar to Eq. 8-24 through 8-26 can be derived for a four-detector FMD. The expressions involving onefold, twofold, and threefold coincidences are actually identical to those given for a three-detector FMD with, of course, more possible combinations. Combinations involving fourfold coincidences are as follows:

$$\frac{N_1 N_2 N_3 N_4}{N_{1234}} = \frac{(\bar{\nu})^4}{\overline{\nu(\nu-1)(\nu-2)(\nu-3)}} N_f^3 \quad (8-30)$$

$$\frac{N_{ij} \cdot N_{k\ell}}{N_{1234}} = \frac{\overline{\nu(\nu-1)}^2}{\overline{\nu(\nu-1)(\nu-2)(\nu-3)}} N_f \quad \begin{matrix} i \neq j = 1, 2, 3, 4 \\ k \neq \ell = 1, 2, 3, 4 \end{matrix} \quad (8-31)$$

$$\frac{N_{123} N_{124} N_{134} N_{234}}{(N_{1234})^3} = \frac{(\overline{\nu(\nu-1)(\nu-2)})^4}{(\overline{\nu(\nu-1)(\nu-2)(\nu-3)})^3} N_f. \quad (8-32)$$

If all the detectors are identical and the various coincidence rates are equal—namely $N_i = N_s$, $N_{ij} = N_d$, $N_{ijk} = N_t$ for $i \neq j \neq k = 1, 2, 3, 4$ then one can express Eq. 8-30 through 8-32 and those equations pertaining to lower level coincidences in terms of the majority-coincidence signals. This is done through the following relationships.

$$N_s = \frac{1}{4}N_{1/4} + \frac{1}{4}N_{2/4} + \frac{1}{2}N_{3/4} + \frac{3}{4}N_{4/4} \quad (8-33)$$

$$N_d = \frac{1}{6}N_{2/4} + \frac{1}{6}N_{3/4} + \frac{1}{3}N_{4/4} \quad (8-34)$$

$$N_t = \frac{1}{4}N_{3/4} + \frac{1}{4}N_{4/4}. \quad (8-35)$$

With these expressions, the left-hand side of Eq. 8-30 through 8-32 can be written respectively as follows: $N_s^4/N_{4/4}$, $N_d^2/N_{4/4}$, and $N_t^4/N_{4/4}^3$.

Eq. 8-23 to 8-26 and 8-30 to 8-32 involve ratios of coincidences (including onefold coincidence, which is equivalent to single detector count rate). The ratios tend to reduce the self-attenuation and spatial variation of prompt fission signatures. The degree of reduction depends on the instrument, the interrogated samples, and the selected ratio. The best ratios for a given set of samples should be determined by the user during instrument calibration. In the selection of the proper ratio, the user should recall that the signal-to-background ratio is considerably improved by using higher-fold coincidences over lower ones. Conversely, the absolute signal is substantially lower if higher-fold coincidences are used. The optimum, dependent on all the factors mentioned above, will often be obtained within intermediate level coincidences, namely, two- and threefold coincidences.

A few experiments found in the literature demonstrate that the coincidence ratio suppresses signature self-shielding and spatial variations. In the first case (Ref. 25), a small ^{252}Cf spontaneous fission source was placed at the center of ISAS. The onefold (N_i) and twofold (N_{ij}) coincidence rates were measured as a function of the lead thickness (providing the attenuation) around the source. Whereas, the onefold and twofold count rate decreased by 30 and 50 percent, respectively, the ratios of the kind given by Eq. 8-23 did not change. These results are shown in Table 8-11.

Another confirmation of the merits of coincidence ratio was provided during trials of the Isotopic Source Assay Fissometer (ISAF) (see Section 9.5.2). The normal majority-coincidence signal, $N_{3/3}$, was measured as well as the ratio $R_{s,t} = [N_1 N_2 N_3 / N_{3/3}]^{1/2}$, which is to the first order, equal to the fission rate. Figure 8.44 shows $N_{3/3}$ for a 100-g sample of ^{235}U as a function of its axial position within the counting volume. The

Table 8-11. Effect of shielding on single and twofold coincidence count rate (²⁵²Cf point source).

Pb Thickness (in.)	$\frac{N_1}{N_1^0}$	$\frac{N_2}{N_2^0}$	$\frac{N_3}{N_3^0}$	$\frac{N_4}{N_4^0}$	$\frac{N_{14}}{N_{14}^0}$	$\frac{N_{23}}{N_{23}^0}$	$\frac{(N_1 \cdot N_4)/N_{14}}{(N_1^0 \cdot N_4^0)/N_{14}^0}$	$\frac{(N_2 \cdot N_3)/N_{23}}{(N_2^0 \cdot N_3^0)/N_{23}^0}$
0	0.0	1.0	1.0	1.0	1.0	1.0	1.0	1.00
1/4	0.825	0.872	0.843	0.832	0.673	0.733	1.01	1.00
1.2	0.757	0.785	0.757	0.752	0.551	0.599	1.02	1.00
1.25 ^a	0.828	0.652	0.778	0.654	0.523	0.496	1.025	1.02

^aNonsymmetrical source shielding—less shielding in direction of detectors Nos. 1 and 3.

Figure 8.44. Axial response of ISAF as determined by $N_{3/3}$ signal and by the coincidence ratio $R_{s,t}$ (see text).

zero axial distance corresponds to the location of the point furthest into the counting volume, that is, the table on which the sample is placed. The ratio $R_{s,t}$, drawn on the same figure, shows a much flatter response, indicating successful suppression of spatial effects on the efficiency. Other measurements, using 300 to 2500 g of highly enriched-uranium (93 percent) in UO_2 , indicate that the scatter of the measured points around the curve is significantly lower for $R_{s,t}$ than for the $N_{3/3}$ signal.

The instrument response to low-quality samples made of calcined ash containing highly enriched-uranium is shown in Figure 8.45. The curves given in Figure 8.45 show the saturation effect due to the self-shielding of the interrogating neutrons. Here again the $R_{s,t}$ ratio demonstrates a much smoother locus for the points as compared to the normal $N_{3/3}$ signal. This effect indicates that the proper coincidence ratio can indeed suppress variations, which affect fission signatures, and that are very difficult to avoid or control.

8.9 DISTRIBUTION OF PROMPT FISSION SIGNATURES OVER TIME

One can distinguish between prompt fission signatures (namely, prompt neutrons and γ -rays) and background radiation because the signatures are emitted simultaneously with the fission event or very shortly thereafter (10^{-15} s) (see Chapter 7). Fast detectors like plastic

Figure 8.45. Response of three-detector FMD (ISAF) to calcined ash containing ^{235}U . (Note the smoother locus of the points for $R_{s,t}$ in comparison to the normal $N_{3/3}$ signal.)

scintillators can detect these emissions within several tens of nano-seconds (<100 ns). Most plastic and liquid scintillators have very short response times—less than 5 ns (see Tables 8-3 and 8-4).

The PMTs that amplify scintillation pulses are usually also very fast. Their ability to amplify fast pulses without distortion is measured by the variation in the electron transit time in the PMT or by the magnitude of the anode pulse's rise time. The rise time of fast tubes are less than 5 ns, but the rise times of slow PMTs are between 30 to 40 ns.

Despite the high speed of the scintillators and PMTs, fast coincidences in ANDA systems are detected with time resolutions of 30 to 100 ns. These relatively long times result from neutron flight time within the sample and detectors and the large number of reflections the scintillation light may have to undergo to reach the photocathode of the PMT. This multiple reflection effect is especially strong in large scintillators, that are often required in ANDA to assure high efficiency. In addition, as mentioned above, one has to take into account the time-of-flight of a neutron in the scintillator before it interacts (by proton recoil) to yield a flash of light. For example, a 1 MeV neutron takes 7 ns to travel 10 cm. Thus, the mean time for a fast neutron interaction in the scintillator can easily be of the order of 10 to 20 ns. For a γ -ray, the mean interaction time is less than 1 ns. Thus, the time distribution of coincident pulses, characterized by τ , will be the convolution of the time distribution of the interaction of radiation in the scintillator (characterized by τ_i), the scintillation decay time (characterized by τ_d), and the PMT rise time distribution (characterized by τ_p). Since these are mean times, averaged over statistical distributions, τ is given by

$$\tau = [\tau_i^2 + \tau_d^2 + \tau_p^2]^{1/2}. \quad (8-36)$$

The coincidence-resolving time is actually 2τ (see Section 8.10). For γ -rays in typical fast scintillators and PMTs, $2\tau = 6$ ns. For neutrons it can be as much as 40 to 80 ns since τ_i can be of the order of 20 to 40 ns. The time distribution in a simple coincidence measurement setup is shown in Figure 8.46. The figure shows (1) the time distribution of coincident pulses from a bare ^{252}Cf fission source (curve a), (2) the same distribution with the prompt neutrons suppressed by shielding the source with 3.5 in. (8.8 cm) of polyethylene (curve b), and (3) the distribution with the prompt γ -rays suppressed by shielding the source with 0.25 in. (0.63 cm) of Pb (curve c). All distributions are normalized to the same centroid height. The time characteristics of the distributions

shown in Figure 8.46 are summarized in terms of their FWHM* and FWTM* in Table 8-12. Figure 8.46 indicates that the prompt neutrons, although fewer, are mostly responsible for the "slow" tail of the time distribution. Conversely, any narrowing of the resolving time of a coincidence circuit below 30 to 40 ns will mostly reduce neutron detection. If one desires to reduce the contribution of γ -rays, one has to exclude the time region of ± 5 ns by a delayed coincidence circuit. This is done in one version of the Random Driver (see schematic in Figure 9.9) with a substantial loss in intensity and sensitivity (see also Section 8.7.1).

8.10 CORRECTION FOR ACCIDENTAL COINCIDENCES

The large efficiencies for fission detection offered by FMD were discussed in Section 8.7. Another important advantage of FMDs is that they use scintillators with fast time response, as discussed in Section 8.9. The fast time response means short resolving time, hence, allowing very high counting rates. The short resolution time (a few ns) in fast coincidence systems is required because accidental or random coincidences must be minimized. These are radiations detected in coincidence

Figure 8.46. Time distribution of coincident pulses.

- a: Unshielded ^{252}Cf source.
- b: ^{252}Cf shielded with 3.5 in. (8.8 cm) of polyethylene.
- c: ^{252}Cf shielded with 0.25 in. (0.63 cm) of lead.

*FWHM: Full Width of distribution at Half of its Maximum amplitude. FWTM: Full Width at Tenth of its Maximum amplitude.

Table 8-12. Characteristics of time distribution of coincidences in FMDs.

Source	Shielding	Radiation Suppressed	FWHM (ns)	FWTM (ns)
^{252}Cf	None	None	4.3	21.1
^{252}Cf	Pb, 0.25 in. (0.63 cm)	γ	4.7	23.5
^{252}Cf	polyethylene, 3 in. (7.5 cm)	Neutron	4.5	9.1
^{22}Na	None	None	3.5	6.7

that do not arise from a common nuclear event. General expressions for correcting accidental coincidences are given, followed by a discussion of their application to a four-detector system.

8.10.1 General Expressions

For a two-fold coincidence system, it is easy to show that the accidental coincidence rate (accidental coincidence counts/second), A_{12} , is given by

$$A_{12} = 2\tau N_1 N_2 \quad (8-37)$$

where τ is the mean time within which coincidences will be registered. Therefore, the resolving time of the system is 2τ . For example, as is shown in Figure 8.35, the sum of pulse width, $\tau_1 + \tau_2$, is equal to the resolving time. Hence, τ is actually equal to the average pulse width. N_1 and N_2 are the single count rates in the two detectors. If N_1 and N_2 are each proportional to the fission source strength, N_f , there follows:

$$N_i = \bar{\nu}\epsilon_i N_f \quad (i = 1, 2). \quad (8-38)$$

Then, A is proportional to the product τN_f^2 , as is shown:

$$A_{12} = 2\tau\epsilon_1\epsilon_2(\bar{\nu})^2 N_f^2. \quad (8-39)$$

The true coincidence rate, N_{12} , is independent of τ , was shown in Eq. 8-24 to be:

$$N_{12} = \frac{\nu(\nu-1)}{(\bar{\nu})^2} \left(\frac{N_1 N_2}{N_f} \right) = \frac{\nu(\nu-1)}{(\bar{\nu})^2} \epsilon_1 \epsilon_2 N_f. \quad (8-40)$$

Therefore,

$$A_{12}/N_{12} = \frac{(\bar{\nu})^2}{\nu(\nu-1)} (2\tau) N_f. \quad (8-41)$$

A_{12}/N_{12} is proportional to τN_f , which should be made as small as possible by reducing the resolving time. However, $\tau N_f \ll 1$ provides only an absolute maximum limit to the magnitude of the accidental coincidences because the single detector count rates, N_1 or N_2 , are not usually governed by the fission signatures, but by background radiation (that is, N_i is *not* dependent only on N_f). Background can result from the sample radioactivity, from the interrogating source, and others. Hence, the accidental to true coincidence ratio should be determined in actual situations.

Generalization of Eq. 8-37 to a system with more than two detectors is not trivial. The proper generalization (Ref. 26, 27) of the formula for two-fold accidental coincidence to n -fold accidental coincidence is given as follows:

$$A_n = n N_1 N_2 \dots N_n \tau^{n-1} + \text{terms in } \tau^n. \quad (8-42)$$

Hence, for a three-detector system,

$$A_3 \cong 3 N_1 N_2 N_3 \tau^2 \quad (8-43)$$

and for a four-detector system,

$$A_4 \cong 4 N_1 N_2 N_3 N_4 \tau^3. \quad (8-44)$$

The accidental rate mentioned so far describes the chance overlap of n pulses, each in one detector. There are, however, other types of overlaps. Especially important in ANDA applications is the overlap of a single pulse with a genuine $(n-1)$ -fold coincidence. This rate is given by the usual twofold accidental coincidence.

$$A_{n-1,1} = 2\tau N_i^{(n-1)} N_i \quad (8-45)$$

where $N_i^{(n-1)}$ is the rate of genuine $(n-1)$ -fold coincidences that are not accompanied by a pulse in the i -th detector. Accidental coincidences also occur from the overlap of genuine $(n-2)$ -fold coincidences with two pulses, and so on. These terms are of the order of τ^2 , and so on. They should generally be added only if the higher terms in Eq. 8-42 are taken into account. However, in ANDA applications, the genuine coincidence rate, for example, $N_i^{(n-j)}$ rate, can be so high that its τ^j term can be comparable or even higher than the τ^{j-1} term of the purely accidental overlap of j -pulses.

8.10.2 Applications to Four-Detector FMD

The nominal quantities in the FMD are the majority-coincidences ℓ -out-of- L : 1-out-of-4, 2-out-of-4, 3-out-of-4, and 4-out-of-4. Of course, one can also record the following:

1. The four single-detector count rates.
2. The six combinations of twofold coincidences.
3. The four combinations of threefold coincidences.

There is much to be gained from this additional information, as shown in previous sections, but the systems needed to acquire it become more complex. At the minimum, the FMD user should be able to use readily all the majority-coincidences. These quantities consist of true, $N_{\ell/L}$, and accidental, $A_{\ell/L}$, coincidences. Thus, the gross coincidence rate $R_{\ell/L}$ is

$$R_{\ell/L} = N_{\ell/L} + A_{\ell/L} \quad (8-46)$$

or more specifically,

$$R_{1/4} = 4N_s - R_{2/4} - 2R_{3/4} - 3R_{4/4} \quad (8-47)$$

$$R_{2/4} = N_{2/4} + A_{2/4} - R_{3/4} - 2R_{4/4} \quad (8-48)$$

$$R_{3/4} = N_{3/4} + A_{3/4} - R_{4/4} \quad (8-49)$$

$$R_{4/4} = N_{4/4} + A_{4/4} \quad (8-50)$$

where, assuming all detectors and efficiencies are equal,

$$A_{2/4} = 12N_s^2\tau \quad (8-51)$$

$$A_{3/4} = 4N_{2/4} N_s\tau + 12N_s^3\tau^2 \quad (8-52)$$

$$A_{4/4} = 2N_{3/4} N_s\tau + 3N_{2/4} N_s^2\tau^2 + 4N_s^4\tau^3. \quad (8-53)$$

The terms on the right-hand side of Eq. 8-51, 8-52, and 8-53 are listed according to descending importance in ANDA.

To illustrate the significance of the correction for accidental coincidences and the relationship of the data output from the FMD, we take the example of an ISAS measurement (Ref. 28) of high-temperature gas reactor (HTGR) fuel rods that contain ^{232}Th , with its radioactive daughter products, in addition to ^{235}U . The presence of thorium results in high onefold and twofold coincidence backgrounds.

The measurement results for two samples, A and B, are given in Table 8-13A. In Table 8-13B the calculated parameters, based principally on Eq. 8-51, 8-52, and 8-53 are also given. The calculations are based on $2\tau = 38$ ns. Table 8-13 demonstrates the importance of the correction for accidental coincidences of the case studied. It shows that the onefold count rate has a large background component. The background also contributes significantly to the accidental coincidences. Table 8-13 also shows that the accidental coincidences contribution can reliably be calculated during the calibrations of the system. Indeed, FMD systems

are often provided with a small microprocessor that makes the necessary correction.

The accidental coincidences can be measured directly by randomizing coincident pulses, that is, by delaying them several times the system resolving time. Typically, 200 ns delay times are used. Any coincidence that occurs after the delay must be accidental by definition. Simple signal delay is entirely proper for a two-detector FMD. For other detectors, the accidental coincidences of genuine $(n-1)$ -fold coincidence, with a random pulse have to be measured.

A simple circuit has been devised for that purpose (Ref. 28). The $R_{1/4}$ signal is delayed and routed to "veto" a 2-out-of-4 majority-coincidence unit. The other four inputs are the signals from the four detectors of the FMD. (In a three-detector FMD, a 2-out-of-3 majority-coincidence veto unit is required.) Any coincidence not vetoed in the unit is an accidental coincidence between a genuine $2/4$ coincidence and a third randomized pulse or between 3 random uncorrelated pulses. Using a delayed $R_{1/4}$ signal, rather than an external random pulser, saves the cost of an additional source or electronic pulser and gives a higher statistical accuracy to the measurement.

The relationship between this 2-out-of-4 coincidence, $V_{2/4}$ (veto rate), and the accidental coincidence rate can be shown to be:

$$V_{2/4} = 2N_{2/4}R_{1/4}\tau + 1.5A_{2/4}R_{1/4}\tau. \quad (8-54)$$

Based on Eq. 8-47 through 8-52, one can rewrite the expression for $V_{2/4}$ as

$$V_{2/4} = \frac{2}{1+\alpha} A_{3/4} + \frac{4}{1+\alpha} 12N_s^3\tau^2 \quad (8-55)$$

where

$$\alpha = (R_{2/4} + 2R_{3/4} + 3R_{4/4})/R_{1/4}. \quad (8-56)$$

For a three-detector FMD,

$$V_{2/3} = \frac{3}{1+\beta} A_{3/3} + \frac{6}{1+\beta} 3N_s^3\tau^2 \quad (8-57)$$

where

$$\beta = (R_{2/3} + 2R_{3/3})/R_{1/3}. \quad (8-58)$$

The adequacy of the $V_{2/4}$ measurements is demonstrated in Table 8-13 by the good agreement between the calculated $A_{3/4}$ and the measured

one (by $V_{2/4}$).^{*} Another measuring scheme for accidental coincidences for a three-detector system has been recently suggested (Ref. 29).

8.11 SUMMARY

ANDA systems depend on the detection of neutrons and/or photons. Neutrons are detected either by slow neutron detectors (common detectors are BF_3 or ^3He gas tubes, or scintillators), after having been slowed down or by fast neutron detectors via recoils from scattering reactions. Fast neutron detectors include ^4He or hydrogen-filled tubes and hydrogen-containing scintillators.

Photons are detected by scintillators, especially sodium iodide (NaI) or by semiconductors [$\text{Ge}(\text{Li})$].

To process and record detected events, associated electronics are required, consisting generally of a preamplifier, amplifier plus pulse shaping network, discriminator, and scalers or a multichannel pulse height analyzer. These electronics should have good linearity, fast response, and low noise characteristics.

Fission multiplicity detectors utilize fast or slow coincidence circuits. Generally, four detectors are used, and 2-out-of-4 or 3-out-of-4 coincidences are detected. When prompt fast neutrons and photons are detected, fast coincidence circuits with resolving times between 40 to 100 ns are used. For slow neutron "well" coincidence detectors, high-efficiency thermal neutron detectors are utilized with slow coincidence circuits having coincidence gates of 20 to 100 μs .

Various formulas to correct for deadtime, accidental (chance) coincidence rates, self-shielding reduction by multiplicity ratios, and the effects of multiplicity and detector splitting on efficiency are given. These effects strongly influence the optimal design of a coincidence system for a given ANDA application. Methods to empirically determine accidental corrections concurrently with the basic data are shown to yield reliable results. Methods using the wealth of information furnished by the coincidence systems are described in detail in this chapter. The ability to reduce the self-attenuation of the prompt fission signatures (neutrons and γ -rays) and the effects of variation in fissile material distribution within the sample are presented both theoretically and experimentally.

^{*}A better agreement could be achieved, if the resolving time used in the calculation was 36 ns.

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CHAPTER 9

REPRESENTATIVE ANDA SYSTEMS

9.1 INTRODUCTION

In the foregoing chapters we have reviewed the basic nuclear interactions of radiations and their transport through media. The various available sources of radiations and the special features of radiations resulting from fission events have been discussed. The fundamentals of the design of the four building blocks of ANDA systems have been analyzed, namely:

- radiation sources,
- source spectrum tailoring and shielding devices,
- sample chambers, and
- detectors and electronics.

Now, the material of the previous chapters can be synthesized to describe representative ANDA systems. In Table 1-2 (see Chapter 1), 23 ANDA techniques have been listed. These are characterized by the source type (isotopic neutron sources of high or low energy, charged particle accelerators, reactors, x-ray or γ -ray sources), the incident radiation (determined by the source and its spectrum-tailoring system), and the observed signature radiation (prompt and delayed neutrons and γ -rays, or combinations of these). Within these categories, further classification by detection method (see Chapter 8) is also possible. Signature radiation is the primary classification in this chapter.

Most of the ANDA systems described in this chapter are actually used continuously or periodically in the nuclear industry or in major laboratories around the world. But a few systems are only in the design stage or have been only suggested for some special applications. The list of the ANDA systems discussed in this chapter, long as it is, should not

be construed as being complete. The systems described here are, nevertheless, representative of what has been or could be achieved in applications of ANDA techniques to nuclear materials control and measurement in the nuclear industry.

The descriptions of the various representative systems include statements on "precision" and "accuracy." The term "precision" describes the reproducibility of measurements by a given method or system and it can be determined by assaying the same sample several times. Accuracy describes the systematic errors encountered in a given method or system. These errors are caused, for example, by calibration errors, nonrepresentative standards, or improper operating procedures. Systematic error is normally determined by assay of a known standard, or in absence of the latter, the error can be described by the differences between assay results for the same sample at different plants and laboratories. The precision and accuracy values are given normally at the one relative standard deviation (one sigma level).

9.2 DELAYED NEUTRON SYSTEMS

Delayed neutrons form a convenient and useful signature because they can be easily distinguished, by time discrimination, from source neutrons and prompt fission neutrons (see Chapter 7). Of course, the source must be pulsed or intermittent so that the delayed neutrons can be detected during the absence of source or prompt fission radiation. Pulsed accelerators, movable sources, movable samples, and shutters are used in delayed neutron systems.

9.2.1 Pulsed 14-MeV Sources

Sources of 14 MeV, such as Cockcroft-Walton accelerators (see Chapter 4), are a natural choice when delayed neutrons are detected since these sources can be easily pulsed to give high on-off ratios.* The source intensity is also high—in excess of 10^{11} n/s during the pulse. In addition, the high neutron energy results in good penetration of the sample. Pulsed 14-MeV systems have been used for samples of all sizes: small samples, 1-gal. containers, and 55-gal. drums. They have been proposed for the assay of spent fuel rod assemblies and leached hulls of spent fuel rods.

The principle of the delayed neutron technique is demonstrated in Figures 9.1 and 9.2, which show the delayed neutron response curves of

*The on-off ratio is the ratio of source neutron intensity, during the pulses, to source neutron intensity between pulses. This ratio should be high so that the source neutrons appearing between pulses are not confused with delayed neutrons.

Figure 9.1. Delayed neutron time response of ^{238}U and ^{235}U .

Figure 9.2. Delayed neutron time response of ^{238}U , ^{239}Pu , and composite sample of 54% ^{238}U and 46% Pu.

several samples during and after a one second irradiation (Ref. 1). The curves are normalized to equal counts during the last second of counting to bring out the differences in slope during the first seconds of counting. These curves lead to isotopic discrimination ratios of about 1.25 between ^{238}U and ^{235}U and between ^{238}U and ^{239}Pu .

The Los Alamos MONAL (Mobile Nondestructive Assay Laboratory) contains a typical 14-MeV neutron source and delayed neutron detection

system (Ref. 2). The source strength in MONAL is about 2.5×10^{11} n/s and the on-off ratio is about $\sim 10^8$. Spectrum-tailoring assemblies of Pb and Pb-C (see Chapter 6) are used to produce interrogating neutrons of ~ 3 -MeV and ~ 0.5 -MeV energy, respectively. A large sample cavity permits samples of various sizes (1-gal. cans, 55-gal. drums) to be assayed. Figure 9.3 shows a schematic of the spectrum-tailoring assembly and sample cavity when assaying 1 gal. cans. The tailoring assembly for 55-gal. cans is shown in Figure 9.4. In both cases, delayed neutrons were detected with a slab detector of ^3He counters (see Chapter 8). The fission counter shown in Figure 9.3 is employed for normalization to reduce the effects of neutron moderation in the matrix materials (see Sections 6.8, 6.9, and 6.10).

Table 9-1 shows results of a MONAL assay of 1-gal. cans. Results indicate good agreement and linearity of response for the two standards to within 5 percent. The differences between the assay results for the other samples and their "by difference" weights were attributed to inaccuracy of the "by difference" content (Ref. 2). The ANDA results, shown in Table 9-1, agree also with passive NDA results (not shown in the table) within their respective errors (± 3 percent for the active assay and ± 10 -15 percent for the passive assay).

Figure 9.3. Active interrogation system for 1-gal. cans.

Figure 9.4. Schematic of experimental arrangement for the assay of 55-gal. barrels. (^{238}U counter serves to normalize to source intensity. ^{235}U counter serves to monitor matrix effects.)

Figure 9.5 shows results (Ref. 3) of an assay of a set of 55-gal. standards that were designed to simulate as closely as possible some plutonium-bearing solid process materials and residues encountered in the plant. The best statistical accuracy is demonstrated by the passive γ -ray assay ("Passive 2" in Figure 9.5); but it also shows a bias, probably because of the self-attenuation of the γ -rays. The wide statistical scattering of the results obtained with the 14-MeV interrogation arises from the low yield of delayed neutrons from plutonium and because large neutron background from the (α, n) and spontaneous fission has to be subtracted. Table 9-2 shows the composition of the samples involved

Table 9-1. Active Pu assay of recovery cans containing sand, slag, and crucible matrix materials.

Can No.	"By Difference" Weight (g Pu)	Active Assay ^b	
		E ~ 3 MeV (g Pu)	Pb-C Moderator (g Pu)
G-1 ^a	17.64	Standard	Standard
G-2	176.40	178.0 $\pm$ 5.0	168.0 $\pm$ 5.0
XBL-40-1	13	22.5 $\pm$ 0.7	20.6 $\pm$ 0.6
XBL-40-2	13	16.6 $\pm$ 0.5	15.0 $\pm$ 0.8
XBL-42	11	14.4 $\pm$ 0.5	14.7 $\pm$ 0.6
XBL-43	23	24.0 $\pm$ 0.7	21.0 $\pm$ 0.8
XBL-44	18	15.8 $\pm$ 0.5	15.8 $\pm$ 0.5
XBL-45	24	16.5 $\pm$ 0.5	15.0 $\pm$ 0.5

^aG-1 and G-2 are standard cans with known Pu content, as listed.

^bThe uncertainties quoted are standard (one sigma) statistical errors.

Figure 9.5. Active and passive assay results of rocky flats 55-gal. standards. ("Passive 1" and "Passive 2" measurements use spontaneous fission neutrons and plutonium γ -rays, respectively.)

in the measurements just discussed. Some knowledge of composition is important for the understanding of the nature and magnitude of the matrix's influence on assay results.

Other results obtained with a 14-MeV generator and delayed neutron detectors have also been reported. At Hannover University (Ref. 4), the ^{235}U content of spherical fuel elements of high temperature reactors (HTR)* has been assayed. The irradiation and counting were performed inside a 15- × 15- × 15-cm graphite block surrounded by a lead cube, and the delayed neutrons were detected by a ^3He slab detector on top of the cube. Table 9-3 shows some results obtained for highly enriched MTR** fuel elements and ^{235}U - ^{232}Th HTGR fuel spheres. Similar work

*High Temperature "Pebble Bed" Reactor.

**MTR stands for Materials Testing Reactor.

Table 9-2. Summary of rocky flats 55-gal. barrel standards.

Description	Matrix	Matrix Composition (Wt. %)	Matrix Av. Net Weight (kg)	Matrix Av. Density (g/cm ³)	Plutonium (as PuO ₂) Loadings Assayed (g Pu)
Graphite molds	60-mesh graphite	100	110	0.53	60, 145, 195
Dry combustibles	Carbon	90	24	0.12	10, 165, 175
	Plastics	5			
	Cellulose	5			
Wet combustibles	Cellulose	80	51	0.25	28.5, 166
	Water	15			
	Plastics	5			
Washables	Polyvinyl	42	32	0.15	10, 90, 160
	Lead gloves	28			
	Polyethylene	20			
	Cellulose	7			
	Surgeon gloves	3			
Raschig rings	Pyrex glass w. 12% Boron as B ₂ O ₃	100	82	0.39	40, 95, 185
Resin	Dowex 1 × 4	100	—	—	25, 110
Benelex-Plexiglas	Benelex-Plexiglas	—	—	—	75

Table 9-3. Assay of highly enriched-uranium with a 14-MeV generator with delayed neutron detection.^a

SNM	Sample Description	(P) (A) Precision Accuracy	Assay Time Per Sample
^{235}U	MTR-fuel elements, ^{235}U , 89.9% enrichment, 180 g uranium (measurement without graphite pile).	(P) $\pm 2.7\%$ (1 σ) (A) $\pm 4\%$ (1 σ)	3 min
^{235}U	HTR-spherical fuel elements, 1 g ^{235}U (93%) + 5 g ^{232}Th measured in graphite pile.	(P) $\pm 4.3\%$ (1 σ)	30 s

^a(Ref. 4).

was done at Julich (Ref. 5). Both the total fissile content ($^{235}\text{U} + ^{233}\text{U}$) and the $^{233}\text{U}/^{235}\text{U}$ ratio were determined in HTR fuel elements, with statistical errors between 2 and 8 percent.

Since 14-MeV neutron generators are not forbiddingly large (some are even portable), this ANDA method is well suited for use in the field. MONAL, for example, is housed in a trailer that can be moved to the installation where the assays are to be performed. Other considerations relating to field use of 14-MeV neutrons are discussed in Chapter 4.

9.2.2 Pulsed Van de Graaff Sources

The pulsed, 3.75-MeV Van de Graaff accelerator at the Los Alamos Small Sample Station yields about 10^{12} n/s and its on-off ratio is 10^9 . It is operated with a cycle of 100 ms: 35-ms irradiation, 25-ms delay, and 40-ms counting period. Neutrons with 400- to 600-keV energies are produced by the $^7\text{Li}(p,n)$ reaction. They provide good penetration and interrogate fissile isotopes only. The delayed neutrons are counted with a high-efficiency slab arrangement of ^3He counters. This system is particularly useful for sample sizes from 0.1 to 10 g of fissile material. However recently, medium and large size assay systems based on the Van de Graaff source, using both thermalized and 0.1-0.4 MeV neutrons have been designed (see Ref. 6). The expected range of fissile content with these systems is 1 to 1000 g.

Moderating the source neutrons to thermal energies improves considerably sensitivity,* since as little as 1 mg of natural uranium may be detected. In large ($\sim 8\text{-L}$) samples, sensitivities of the order of 4 parts per million (ppm) of natural uranium or 0.03 ppm of enriched uranium are obtained.

*Sensitivity denotes the smallest amount of material concentration that can be detected unambiguously.

Because Van de Graaff sources are large and complex, they are not particularly well suited for field work. The Van de Graaff at the Los Alamos Scientific Laboratory (LASL) (Ref. 6) facility is the only one in use. Van de Graaff accelerators must be more or less permanently installed at a field facility, and they require highly trained personnel for operation and maintenance.

9.2.3 Irradiation Step Techniques

Because they are smaller and less complex, isotopic accelerators may be combined well with delayed neutron detection. However, since the source is not pulsed, some other means must be used to obtain intermittent irradiations.

1. The source can be brought up to and removed from the sample.
2. The sample can be brought up to and removed from the source.
3. A neutron shutter between source and sample can be used.

All of these techniques are denoted by the term "irradiation step function technique." In these methods, the sample is irradiated for a prescribed time, then irradiation is stopped to allow the induced delayed radiation to be measured. The pulsed, 14-MeV neutron generator and the Van de Graaff accelerator are examples of obtaining an irradiation step function by shutting off the ion beam that generates neutrons. This can be done very rapidly, typically within microseconds. The methods described in this section are slower, but are nevertheless fast enough to be useful. It is only necessary, that the time for shutting off the beam be short as compared to the delayed neutron half-lives and that it be accurately reproducible. We will describe systems based on the first two of the above techniques since shutters are not widely used, except when a nuclear reactor served as a neutron source.

An example of a system in which the source is moved to and from the sample is the Shuffler (Ref. 7). The Shuffler utilizes a ^{252}Cf source that is "shuffled" from a shielded area (the "Dwell tank" in Figure 9.6) to an irradiation position in less than 0.5 s. To obtain both precision and flexibility, the source speed, source position, sample position, and the irradiation and count times are controlled by stepping motors. The source size is about 1 mg ($2.4 \times 10^9 \text{ n/s}$). The irradiation and count times are each a few seconds. Figure 9.6 shows a schematic of the Shuffler.

A soft neutron source spectrum makes the Shuffler quite sensitive: less than 1 mg of ^{235}U can be detected. Its sensitivity is somewhat poorer for plutonium, owing to the smaller delayed neutron yield and the additional emission of spontaneous fission and (α, n) neutrons from

Figure 9.6. Modulated ^{252}Cf assay system, "Shuffler," showing the shielding tanks and fast neutron irradiation chamber. The top drawing is a cross-sectional view of the system shown in the lower drawing (Ref. 7).

the sample. Obviously, the sensitivity of the system to the hard neutron spectrum is much lower. Samples with high γ -ray activity (such as waste barrels contaminated with fission products) can be surrounded with a lead shield, which also slightly increases the source flux (because of scattering in the lead), without increasing the background (Ref. 8). This has been incorporated in another application of the source shuffling technique that has been recently proposed (Ref. 9). The proposed scanner measures small amounts of fissile material present in 55- and 80-gal. drums, of possibly highly radioactive waste, from reprocessing plants. Figure 9.7 shows a schematic of the barrel scanner. The heavy lead (not shown) and iron shielding is one feature of the system that also moderates and reflects the source neutrons. Another feature is the sequential positioning of the source, in the three positions indicated in the figure, to achieve uniform, axial irradiation.

The delayed neutron pellet assay gauge (Ref. 10) is an example of a system in which the sample is transported to and from the source. Figure 9.8 is a schematic of this system. Two ^{252}Cf sources (5 mg each) are

Figure 9.7. ^{252}Cf -based barrel scanner. (Underground concept, suitable to assay highly radioactive 55- and 80-gal. drums.)

Figure 9.8 Sketch of light water breeder reactor (LWBR) pellet assay gauge.

used. The delayed neutron detector consists of six ^3He detectors. The irradiation and counting times are each 100 s, and the sample transport time is a few seconds. During the transport time, the delayed neutron intensity decays by about a factor of four. This loss in signal and the requirement of very high precision are reasons for the high strength of the sources. This machine is employed for the assay of $^{233}\text{UO}_2\text{-ThO}_2$ pellets. The delayed neutron contribution, from fast fission in the thorium (about 2.5 percent of the total), is determined by running through the machine a ThO_2 -pellet of the same dimensions as the mixed $\text{UO}_2\text{-ThO}_2$ pellets. The calibration curve is shown in Figure 9.9. The excellent agreement between the chemically determined ^{233}U content and the ANDA measurements for unirradiated fuel rods made up of this type of pellet is shown in Table 9-4 (Ref. 11). A similar system with heavier γ -ray shielding around the delayed neutron detectors (^{10}B -coated proportional counters) will be used to measure the total residual fissile content in the irradiated fuel pins of the light water breeder reactor (LWBR).

Figure 9.9. Pellet assay gauge calibration curve for the determination of fissile content in $^{233}\text{UO}_2\text{-ThO}_2$ fuel pellet fissile ($5\text{-mg}^{252}\text{Cf}$ source, 100-s irradiation and counting times, 4-s delay time).

Table 9-4. LWBR pellet delayed-neutron assay gauge results for unirradiated fuel rod assay.^a

System Type	"Known" ^{233}U Loading ^b (g ^{233}U)	Assay Result (g ^{233}U)
Thermal interrogation	6.036 ± 0.027	5.950 ± 0.059
Epithermal interrogation	6.036 ± 0.027	6.03 ± 0.15

^aCorrection for self-adsorption was applied to the thermal interrogation. Epithermal interrogation was achieved by absorbing the thermal neutrons with an appropriate absorber (for example, Cd).

^bThis "known" loading is based on the Westinghouse Bettis Laboratory destructive chemical and mass spectroscopic analysis of the pellet lots used in this rod.

9.3 PROMPT NEUTRON SYSTEMS

Prompt neutrons are easier to detect than delayed ones because of their much higher intensity. On the other hand, one must then distinguish the prompt neutrons from the source neutrons, which are many times more abundant. The task of distinguishing the prompt fission neutrons from source neutrons can be accomplished either by detecting their difference in energy or by detecting the coincidence properties of the fission neutrons and γ -rays. Various neutron sources can be used with each of these methods.

If low-energy neutron sources are used (intrinsically low-energy sources, fast-moderated sources, or thermalized sources), the prompt fission neutrons can be distinguished from the source neutrons by energy-biased detectors, (such as ^4He gas proportional counters), proton recoil detectors (either gas or scintillator), or other scintillation detectors. The properties of these detectors are described in Chapter 8.

9.3.1 Low-Energy Isotopic Sources

Low-energy isotopic sources, especially (γ, n) sources such as Sb-Be and Ra-Be, are particularly effective in the determination of fissile content in the presence of large amounts of fertile material, as in low enrichment fuel. These sources are flexible: they may be made in various shapes and configurations as required by the needs of the specific system. The most common low-energy source is Sb-Be, which yields neutrons with energies below 30 keV (see also Chapters 4 and 6).

An early study (Ref. 12) discussed various possible measuring techniques employing Sb-Be sources, including:

- direct, prompt, fast neutron detection with biased detectors,
- age (or slowing-down distance) difference technique, and
- delayed neutron technique.

A system based on the first method has been built (Ref. 12) and is shown schematically in Figure 9.10. Its liquid scintillation detector, highly efficient for fission neutrons, is also sensitive to γ -ray radiation. The γ -ray background was reduced both by the massive lead shield shown in the figure and by a pulse-shape discrimination circuit in the detector's electronics. This circuit distinguishes γ -ray pulses from neutron pulses by their difference in decay time in the liquid scintillator and then rejects the γ -ray pulses (see Chapter 8).

Typical results obtained with this system are shown in Table 9-5. The samples were short HTGR fuel rod consisting of ^{235}U and Th in a graphite matrix, with a Th/ ^{235}U ratio of about 17. Sample No. 2 was used as a reference sample for normalization purposes. Good agreement between the manufacturer's values and those by Sb-Be ANDA (and, also

Figure 9.10. Detailed cross section of the sub-MeV assembly based on Sb-Be neutron source.

**Table 9-5. Nondestructive assay of HTGR fuel elements
(using Sb-Be source and a liquid scintillator).**

From Manufacturer			Low Sub-MeV Results		Passive Scanning [Ge(Li)]	
Sample No.	²³⁵ U (g)	Normalized	Net Signal (Counts/s)	Normalized	Net Signal	Normalized
1	0.94	1.12	5.06	1.10 ± 0.01	1.42×10^5	1.12 ± 0.005
2	0.84	1	4.60	1	1.26×10^5	1
3	0.74	0.88	4.04	0.88 ± 0.02	1.10×10^5	0.88 ± 0.005

in this case the PNDA using a Ge(Li) detector) are shown. The use of liquid scintillators limits the method to samples, which do not contain fission products, like fresh fuel rods or clean scrap.

Two other instruments that combine Sb-Be sources and prompt fast neutron detection are described. The first apparatus, schematically shown in Figure 9.11, is intended for intermediate-sized samples of fissile material (Ref. 13). It demonstrates an efficient use of the keV-neutrons from the Sb-Be for maximum penetrability without undue moderation. The resulting linearity of the system to ^{235}U mass is shown in Figure 9.12, which indicates that neutron self-absorption begins only above 50 g of ^{235}U . The attempt to design the system to obtain also flat spatial responses was reasonably successful, as indicated by Figure 9.13.

A second instrument that also requires the high penetrability of the Sb-Be neutrons is described (Ref. 14). The main purpose of this is to assay FFTF* fuel rods, which contain about 25 percent PuO_2 . The source surrounds the sample fuel rod, as is shown in Figure 9.14. The Ni ring surrounding the Sb-Be source acts as a source neutron reflector, a 2.2-cm thick Ni reflector, nearly doubles the signal. Similarly, the presence of a fast neutron reflector outside of the detector ring enhances the signal: about 7 cm of iron results in a 50 percent increase in count rate. Greater thicknesses do not further increase the signal.

Another example of an elaborate system based on Sb-Be and fast neutron detection is the Euratom photoneutron source assay system

Figure 9.11. Experimental photoneutron assay system (top view).

*Fast Flux Test Facility.

Figure 9.12. Photoneutron assay system response versus ^{235}U mass.

Figure 9.13. Spatial dependence (deviation from the response at the center position) of the photoneutron assay system response.

Figure 9.14. Cross section of a cylindrical assembly used in the assay of fast reactor fuel rods.

(Ref. 15). This system can measure larger samples: the inner dimensions of the irradiation chamber are 40 cm wide, 60 cm high, and 60 cm long. The system allows rotation and vertical scanning of the sample container in order to obtain a more uniform response. The beryllium mantle of the Sb-Be can be removed pneumatically in order to "shut off" the source, so that the sample can be placed in the system. This capability allows, in principle, for measurements of delayed neutrons, but such measurements have not yet been attempted. The fast fission neutrons are detected by banks of ^4He recoil detectors. Response curves for bulk samples of uranium are shown in Figure 9.15.

An experimental arrangement, proposed in 1969 (Ref. 16), has been set up (Ref. 17) to distinguish the fission-induced neutrons from the intense, low-energy Sb-Be source neutrons by the "Fermi-age" or "selective transport properties." A more recent setup (Ref. 18) is shown in Figure 9.16. In this arrangement, the sample is surrounded by a Sb-Be ring source. The detector is separated from the sample (and source) by approximately 12 cm of borated water that slows down and absorbs, preferentially, the low-energy source neutrons (see Chapter 6). Spherical HTR fuel elements containing graphite, thorium, and uranium oxides with different amounts of ^{235}U and various burn-ups were counted at 12 cm, using a 100-Ci ^{124}Sb source. The neutron count rates for different types of fuel elements are given in Table 9-6. This table indicates that the measured neutron count rate/gram of fissile uranium is 92.5 ± 5 counts/s, independent of fuel composition and burn-up. A large portion of this error is believed to reflect the inaccuracies of the

Figure 9.15. Calibration curves for a Euratom Sb-Be assay system.

Figure 9.16. An experimental arrangement for the Fermi age (or selective transport) technique.

Table 9-6. Fermi age technique: neutron count rate
of different HTR spherical fuel elements^a

Element, Type ^b	Original Content of U-235 (g)	Burn-up (fifa) ^c	Final Content of Fissile Uranium (g)	Neutron Count Rate		Neutron Count Rate/g of Fissile Uranium (counts/s/g/U _{fissile})
				Measured	Net	
Blank	—	—	—	1.48	0.0	—
Graphite ball	—	—	—	1.41	-0.07	—
U (natural) ball	0.0134	—	0.0134 U-235	2.88	1.40	104
UCC	1	—	1	89.56	88.16	88
Wall paper	1	—	1	87.68	86.28	86
Pressed el.	1	—	1	89.12	87.72	88
34% fifa ^c	1	34%	0.58 U-235 0.04 U-233	58.55	57.15	92
			0.62 U			
64% fifa ^c	1	64%	0.28 U-235 0.07 U-233	35.40	34.0	97
			0.35 U			
						Average 92.5 ± 5

^aSource strength: ~100 Ci Sb-124.

Working distance: 12 cm.

^bSee Ref. 18 for description of the samples used.

^cUnit of burn-up given by the number of fissions/initial number of fissile atoms.

standards. These tests demonstrate the potential use of the technique for highly radioactive samples.

Systems based on isotopic photoneutron sources other than Sb-Be have been investigated, including Ra-Be (Ref. 19). Recently, a ^{88}Y -Be system has been considered (Ref. 20) for measurements of spent light water reactor (LWR) fuel elements.

Another early system based on detection of fast prompt neutrons was built by National Nuclear Co. It used two proton recoil, ZnS(Ag) (mixed with hydrogen-containing substance) scintillation detectors (Hornyak detectors) to sense fast neutrons emitted by a sample (up to 5 gal.) located on a turn-table surrounded by a hydrogen-containing moderator. A ^{238}Pu -Li source was positioned midway between the two detectors. Although the neutron detection efficiency of this kind of detector is quite low because of its limited thickness (see Chapter 8), it is extremely insensitive to γ -rays. A system based on ZnS(Ag) detectors can, therefore, be used for irradiated material containing significant amounts of fissile materials.

9.3.2 Moderated Neutron Source Systems

The advantage of moderated (tailored) neutron source systems is their higher intensity, either accelerators or relatively intense ^{252}Cf sources can be used. On the other hand, an often massive source tailoring system is required. Furthermore, an accelerator source, in particular, is expensive and complex. The tailoring system may not completely remove high-energy source neutrons that interfere with prompt fission neutrons. If carefully designed, however, the average interrogating neutron energy can be higher than the characteristic 30 keV of Sb-Be sources. This results in better sample penetration and a much lower γ -ray background. The design of spectrum-tailoring systems has been discussed in Chapter 6.

A reactor fuel rod scanner, which measures the total fissile content, is a commonly used system based on such a moderated neutron source and fast neutron detection. An example of such a scanner is the LWR fuel rod assay system with a D_2O -moderated ^{252}Cf source and biased ^4He detectors. It was designed and built at Los Alamos (Ref. 21). Figure 9.17 shows the moderator-detector assembly with a fuel rod in one of the six fuel-rod channels. A programmable calculator handles data acquisition from the eight detectors, data reduction, and printout. The net count rate for a typical boiling water reactor (BWR) fuel rod (2.34 percent enrichment) is about 1200 counts/s for a ^{252}Cf source of 100 μg . For a PuO_2 fuel rod with a loading of 0.15 g of Pu/in., the count rate is about 1400 counts/s.

Figure 9.17. Moderator and shield assembly for a 6-channel ^{252}Cf -based total fissile assay system for LWR fuel rods.

The calibration of these fuel rod scanners is based on fuel rod standards supplied by the rod manufacturer. For low-enrichments, the response is adequately described by the function

$$R = A(1 - e^{-BU}) \quad (9-1)$$

where R is the net count rate, A and B are constants, and U is the ^{235}U content of the rod. The exponential term arises from the flux depression in the rod. The response is linear for small amounts of ^{235}U . A typical calibration curve is shown in Figure 9.18.

Similar systems, measuring total fissile content of fresh fuel rods, are incorporated in some pellet-to-pellet fuel rod scanners based on delayed γ -ray detection (see Section 9.6). The detection of the prompt fast fission neutrons, which introduces a certain complexity into the system, alleviates, however, some of the problems encountered in determining the total fissile content from the delayed γ -ray signature (for example, scan speed variation and its effects on the time-dependence of the delayed γ -ray signal, and photomultiplier phosphorescence effects).

An important example of a moderated neutron source and fast neutron detector is a proposed ^{252}Cf system for the measurement of the residual fissile content in spent fuel assemblies from LWRs (Ref. 22). In this system, moderation of the fast source neutrons (^{252}Cf) is provided by the water in the spent-fuel storage pool. The fast neutrons resulting from induced fissions are detected, either inside or close to the fuel assembly, by a threshold fission detector, for example, a ^{238}U fission chamber, which can stand the extremely high fields of γ -ray rays typical

Figure 9.18. Calibration curve for a ^{252}Cf -based fuel rod assay system (ordinate gives the net counting rate due to the fuel rod).

of spent nuclear fuel. It is estimated that reasonable sensitivities to residual fissile content (^{235}U , ^{239}Pu) and accuracies of better than 5 percent per assembly can be obtained.

9.4 ACCELERATOR PHOTOFISSION AND PHOTONEUTRON TECHNIQUES

Electron linear accelerators (Linacs) can be used as interrogation sources in two ways: either the γ -rays (Bremsstrahlung) can directly cause photofission in the sample, or the γ -rays can produce photoneutrons in a converter like D_2O , which then interrogate the sample (see Chapter 4). In Chapter 7, we plotted curves of the yield of both prompt and delayed neutrons after several isotopes underwent photofission by Bremsstrahlung at various energies. The manner in which *both prompt and delayed neutrons* are measured following repetitive pulses of Bremsstrahlung radiation is indicated in Figure 9.19 (Ref. 23). This figure shows the prompt and delayed neutron time-dependence for a

Figure 9.19. Time-dependence of neutron count rate in a moderated neutron detector following Bremsstrahlung-induced photofission.

^{235}U sample at an electron energy of 7.6 MeV. The apparent decay constant ($71\ \mu\text{s}$) of the prompt neutrons is actually a characteristic of the moderated neutron detector system (BF_3 counters in a polyethylene block).^{*} Prompt neutrons can be detected for a relatively long time (~ 30

^{*}The prompt neutrons are slowed down in polyethylene and then diffuse in it. The "decay" is a result of their capture by polyethylene and the BF_3 counters and their leakage from the system.

to 700 μ s) because of the moderator's "die-away" time, hence, enabling the experimenter to conveniently separate neutrons from the γ -ray flash. Delayed neutrons dominate the count rate after about 100 μ s. Since there are no source neutrons, it is possible to count both prompt and delayed neutrons.

9.4.1 Delayed Neutron Techniques

The assay of some samples of low-enrichment uranium oxide (Ref. 24) can illustrate how the Linac works as an interrogating source for detecting only delayed neutron signatures. The total uranium content of the sample was determined to within ± 2 percent by comparing the delayed neutron yields from photofission at 10 MeV with the yields from standards. A correction of about 7 percent had to be made for self-absorption of the Bremsstrahlung radiation in the samples. Delayed neutron response was more reliable than prompt neutron response because delayed neutrons can only come from the fission products, whereas some prompt neutrons can arise from (γ, n) reactions in various materials near the sample and detector.

For measuring the amount of ^{235}U , sub-MeV neutrons were used to interrogate the sample (see Section 4.7). These were generated by the (γ, n) reaction in a D_2O -target irradiated by 9.5-MeV Bremsstrahlung radiation. The calculated neutron spectrum for this Bremsstrahlung energy (see Figures 4.21 and 6.17) showed that most of the neutrons were below the ^{238}U threshold value. One standard and one unknown sample were irradiated for about 10 min each, and the relative delayed neutron yields were measured. A comparison of these measurements gave the ^{235}U content of the unknown sample within 10 percent. The major part of this error was caused by the low count rate in the simple, nonoptimized system used. Other results demonstrating the potential capabilities of the accelerator sub-MeV technique are given (Ref. 25).

9.4.2 Prompt Neutron Technique

We have already discussed one Linac system using photofission for total uranium determination by the delayed neutron signature. Another example is the barrel scanner constructed by Gulf-General Atomics as part of their mobile laboratory GAMAS (General Atomic Mobile Assay System) (Ref. 26). GAMAS uses electron Linac-generated photons to induce photofission in ^{235}U , ^{238}U , and ^{239}Pu . The advantages of this machine, which is housed in a trailer, stem from the high intensity and penetration of the interrogation photons (5 to 10 MeV) and of the stimulated fission neutrons. In the original GAMAS, both prompt and

delayed neutrons were detected by two banks of moderated BF_3 counters. Samples (up to 55-gal. drums) were offset horizontally, rotated, and scanned vertically. An on-line computer processed the data, controlled the scanning sequence, calculated all corrections, and printed out the fissionable content of each sample. Typical accuracies for uranium and plutonium waste in 55-gal. drums ranged from 2 to 15 percent (with poor standard). For certain fuel elements, 1 percent precision was reached.

Figure 9.20 shows a schematic drawing of GAMAS, and Figure 9.21 gives the original sample-detector arrangement. Figure 9.22 shows a later, improved arrangement which has a much higher efficiency and lower background than the original. It allows the delayed neutrons to be used both as a measure of total fissionable material, and, in conjunction with the prompt neutrons, as a measure of ^{235}U enrichment.

GAMAS can also analyze passive γ -rays emitted from ^{235}U and ^{239}Pu . In some samples, the presence of large amounts of uranium or thorium makes passive assay impossible because of the large self-absorption. Active assays can be done at two different Linac energies to distinguish thorium from uranium, since the relative prompt and delayed neutron yields from thorium and uranium differ as functions of Linac energy (see Figure 7.3).

Figure 9.20. Cutaway drawing of GAMAS showing the improved barrel scanner.

Figure 9.21. Schematic representation of a barrel, neutron detectors, and Bremsstrahlung collimation system.

Figure 9.22. Cross section of the improved barrel scanner showing details of the neutron detector arrangement.

Typical results obtained by using GAMAS in the active mode are given in Tables 9-7 and 9-8. Table 9-7 shows one important source of background (for the prompt neutron signature stimulated by the photofission process): the (γ, n) neutrons created by γ -rays interaction with deuterium traces in natural hydrogen present in water inside the drum or in the polyethylene moderator of the detector. By using a low-energy electron (and hence photon) beam, one can correct for this background. As seen in Table 9-7, this correction is quite reliable, even for relatively "wet" samples (Ref. 27). The fact that the yield per gram is practically constant demonstrates the adequacy of the techniques for these difficult-to-measure samples.

Table 9-8 shows similar results for Pu-bearing 55-gal. drums* with a large variety of matrix materials. The fissile material in these waste drums was measured by observing the prompt neutrons from (γ, n) and (γ, f) reactions and correcting for the (γ, n) background. The overall agreement, within 7.4 percent between the measured and nominal mass of Pu, was quite good for waste barrel measurements. The largest deviation, which was in wet combustibles, resulted probably from the increased detection efficiency for the partially moderated neutrons in the detectors.

Table 9-7. GAMAS: measured prompt neutron yield from
55-gal. U-scrap standards.

Nominal U (g)	Barrel Weight (kg)	Yield at 7.5 MeV (counts/ monitor)	Deuterium Correction (counts/ monitor)	Net Yield (counts/ monitor)	Net Yield/g Uranium (counts/ gmonitor)
5.27	37.4	11.6 ± 0.2	8.0 ± 1.0	3.6 ± 1.0	0.68 ± 0.19
15.8	36.5	15.8 ± 0.2	6.5 ± 0.8	9.3 ± 0.8	0.59 ± 0.05
30.0	24.0	21.8 ± 0.2	2.3 ± 0.8	19.3 ± 1.0	0.64 ± 0.03
50.1	42.2	39.0 ± 0.4	9.5 ± 1.2	29.5 ± 1.3	0.59 ± 0.05
0 (cs) ^a	37.0	15.3 ± 0.4	16.5 ± 1.7	-1.2 ± 2	
0 (cs) ^a	45.4	20.0 ± 0.7	21.0 ± 2.2	-1.0 ± 2.4	
0 (cs) ^a	54.5	25.3 ± 0.4	24.7 ± 2.5	0.6 ± 2.6	
0 (cs) ^a	81.6	31.5 ± 0.3	28.5 ± 2.9	3.0 ± 3.0	

^a(cs) = clean scrap (no uranium).

*Many of the barrel standards were also interrogated by 14-MeV neutrons, as mentioned previously (see, for example, Table 9-2 and Figure 9.5).

Table 9-8. Results obtained by GAMAS on standard barrels containing Pu.

Content	Pu		Measured ÷ Nominal
	Measured (g)	Nominal (g)	
Dry combustibles	180.1	170	1.059
	181.9	180	1.011
	22.9	25	0.916
	166.4	155	1.074
Raschig rings	57.4	61	0.941
	193.6	185	1.047
	80.0	76	1.052
	158.0	161	0.981
	111.0	110	1.009
	48.1	50	0.962
	175.4	194	0.904
Wet combustibles	34.7	28	1.239
	142.2	166	0.857
	113.2	110	1.029
Resin	48.6	50	0.973
	25.2	25	1.008
	84.3	85	0.992
	163.5	175	0.934
Washables	90.6	85	1.066
	(6.5)	10	
	47.9	50	0.952
Graphite	191.8	195	0.984
	164.1	155	1.058
Benelex-Plexiglas	123.9	125	0.991
	50.4	50	1.008

Average ratio of measured to nominal = 1.003 ± 0.078 .

9.5 PROMPT RADIATION TECHNIQUES USING COINCIDENCE METHODS

Earlier chapters have noted that a total of about 10 particles (two to three prompt neutrons plus seven to eight prompt γ -rays) are emitted on the average in a fission event. This high multiplicity makes possible use of fast coincidence techniques to distinguish these radiations from background radiations, which generally appear one at a time. ANDA systems that make use of this method to measure fission events are sometimes called fission multiplicity detection (or detectors) (FMD) systems. Such systems have several detectors, typically four, on different

sides of the sample and coincidences between any two or three detectors are counted. A detailed discussion of the coincidence method and its electronics has been presented in Chapter 8.

9.5.1 Isotopic Source Assay System (ISAS)

One of the earliest active coincidence systems was developed at Gulf General Atomics (GGA). Shortly afterwards, Los Alamos, followed by the National Nuclear Corp., developed similar types of systems. The GGA machine was called the Isotopic Source Assay System (ISAS). One of the most studied ANDA systems (Ref. 28), ISAS originally was designed and used for determination of the fissile content of bulk liquid and solid materials. The accuracy of the instrument depended upon the quality, type and homogeneity of the material, and length of the measurement time. The range of types of material to be assayed could be extended by varying the energy of the interrogating neutrons by using different spectrum-tailoring arrangements.

The ISAS has three major components (see Figures 9.23 and 9.24):

1. Source container or shielding and collimator (SCC), which contains about 25 μg of ^{252}Cf and has appropriate personnel shielding.

Figure 9.23. Basic configuration of FMD in its active mode (ISAS) (horizontal cross section).

Figure 9.24. Different measuring configurations of ISAS. A: ISAS with long sample carrier for samples up to 8 by 51 in. (20 × 127.5 cm). B: Horizontally scanning ISAS detector system.

Collimators may be adjusted in both design and size to fit the particular type of sample being assayed.

2. Fission multiplicity detector (FMD), which houses the rotating sample holder and a set of four plastic scintillators mounted in pairs on opposite sides of the sample. The FMD accommodates containers up to 8 in. (20 cm) in diameter and about 50 in. (125 cm) long.
3. Modular electronics and data-handling equipment.

The fission multiplicity detector in ISAS detects fissions and other nuclear reactions specific to fissionable materials that are induced by neutrons from the source.

The several detectors, usually four plastic scintillators, are electronically connected so that time-coincident events of a selected multiplicity are registered as an indication of fissionable material. The scintillators detect both prompt fast neutrons and prompt γ -rays in coincidence, the resolving time is about 20 to 60 ns depending on the type of photomultiplier used. Usually the 3-out-of-4 coincidence mode is employed (see Chapter 8). Plutonium-240, which fissions spontaneously, can be measured without a neutron source and can be used to indicate total plutonium content, if the isotopic composition is known.

To measure ^{235}U or ^{239}Pu , which does not fission spontaneously, a neutron source is used. To minimize self-shielding effects in samples with high fissile content, ISAS uses a neutron beam filtered by boron. With the filtered beam, a measurable portion of the response results from fast neutron fission of ^{238}U . By taking a second measurement, without the filter, the response is almost entirely due to ^{235}U . The two measurements can be used to determine ^{238}U and ^{235}U in kilogram-sized samples enriched to about 20 percent in ^{235}U . Under optimum conditions, as little as 0.2 g of ^{235}U can be measured in 100 s with a precision of 15 percent. A typical sensitivity curve for ISAS is shown in Figure 9.25.

Figure 9.26 shows a response curve for ISAS for larger amounts of ^{235}U . For these samples, the response is observably nonlinear about 100 g of ^{235}U . The nonlinearity of the response curve is caused by both the attenuation of the incoming neutron beam by the fissile material and the scattering and self-absorption of the stimulated prompt neutron and γ -rays in the uranium itself. The first effect can be reduced by absorbing the low-energy source neutrons in a boron filter, the second effect can be reduced by absorbing the low-energy γ -rays in a high- Z filter (for example, lead) placed between the sample and the detectors. However, each of these remedial actions results in a substantial reduction in sensitivity.

An example of reducing neutron self-shielding by filtering out low-energy neutrons from the interrogating beam is shown in Figures 9.27 and 9.28, which demonstrate the great improvement in linearity of the response curve for ^{239}Pu obtained with a B_4C filter. Indeed, when large quantities of fissile material are present in the sample, the loss in sensitivity accompanying the use of a filtered beam is usually acceptable, since a much better response results. The effect of removing the γ -ray self-shielding is shown in Figure 9.29. It points out a considerable improvement in both the linearity of the response curve and the scatter

Figure 9.25. Lower limits of detection, ISAS (3 out of 4 coincidences).

Figure 9.26. Measurement of ^{235}U quantities in the range of up to 200 g.

Figure 9.27. Measurement of fissile Pu quantities in the range of up to 400 g with soft neutron spectrum.

Figure 9.28. Measurement of fissile Pu quantities in the range of up to 400 g with hard neutron spectrum.

Figure 9.29. Effect of attenuation correction on ISAS response curve (Ref. 29).

of the points in the uncorrected curve caused by variation of the thorium-to-uranium ratio among the samples used (Ref. 29).

ISAS was applied to a large variety of materials in samples of different sizes and matrices encountered mainly in fuel fabrication and scrap recovery. It is difficult to define accuracies for such a general purpose ANDA system. Nonetheless, it was found that standard ISAS units under average conditions routinely gave (relative) accuracies of about 1 to 2 percent (at one standard deviation) for concentrations of 50 g or more fissile material homogeneously distributed throughout a 1-gal. volume of dense matrix material. Accuracies of about 2 to 10 percent were routinely obtained for the same amount of fissile material heterogeneously distributed throughout the matrix material. The sensitivity was typically 0.1 to 0.25 g of fissile material heterogeneously distributed throughout a 1-gal. bulk volume of matrix material.

9.5.2 Isotopic Source Adjustable Fissometer (ISAF)

The Isotopic Source Aadjustable Fissometer (ISAF) (Ref. 30), designed and built by IRT Corporation (formerly Gulf Rad. Tech.—San Diego), also uses an isotopic source and fission multiplicity detection. ISAF can handle sample containers of up to 5 gal. The system contains three 24-in. (60-cm) long plastic scintillators positioned at 90° from one another in three of four quadrants which surround the sample. The fourth quadrant contains an Am-Li source (1.2×10^6 ns/s). The basic geometrical arrangement of ISAF is shown in Figure 9.30. In most applications, moderator wedges, located between the detectors and hydrogen-containing moderating material surrounding the source on three sides, further tailor the source spectrum. For applications, which require more penetrability of the source neutrons, the moderator can be replaced by iron and the wedges replaced by neutron absorbers. A highly thermalized source can make the response insensitive to specific sample matrix effects, for example, those due to moisture. The source can be positioned anywhere along the 24-in. length of the detectors, or it can be removed if passive assay is desired. The FMD signal is 3-out-of-3 coincidences (2-out-of-3 can also be used). The coincidence resolution time is about 80 ns.

Figure 9.31 shows the response as a function of the vertical position of the source and sample. Note that the most uniform response with respect to the sample occurs when the source is about 5 in. (12.5 cm) above the rotating turntable. This is because the portion of the sample

Figure 9.30. Schematic representation of ISAF and associated electronics.

Figure 9.31. Vertical response vs. various source locations above the rotating table.

that receives maximum source intensity has a lower efficiency for detection. The efficiency falls off as the distance between that portion of the sample and the detector photomultiplier tube is increased. Furthermore, the uniformity of response is not improved by scanning the source vertically at a uniform rate: the response functions simply average, as shown by curve No.4 in the figure. To improve uniformity of response, it is necessary to decrease the speed of the source motion in proportion to the decreasing efficiency of the detector as the source irradiates portions of the sample further from the turntable.

The fission efficiency of ISAF, defined as the number of recorded coincidence events/spontaneous fission in a calibrated ^{252}Cf source located at a fixed position (for example, center), is quite high. A typical efficiency curve for 3-out-of-3 coincidences is shown in Figure 9.32. The efficiency varies for 3/3 coincidences from a maximum of 6 to 10 percent for a 3.5-in. (8.8 cm) diameter sample to a minimum of 0.4 to 1

Figure 9.32. Efficiency for detection of ^{252}Cf spontaneous fission as a function of sample size in ISAF.

percent for a 15-in. (37.5 cm) diameter sample. The range of efficiencies results from differences in ISAF systems. The efficiencies for 2-out-of-3 coincidences (2/3) are about a factor of 5 higher. This decrease in efficiency of about an order of magnitude caused by the sample container size underlines the importance of proper selection of containers for ANDA applications. The 3/3 count rate from a fissile plutonium sample, in the form of thin metal foils in a large sample configuration (20-cm diam), is about 2.3 counts/g/s, with a background equivalent to about 0.5 g of Pu. The passive response from ^{240}Pu , in the same configuration for the same metal foils, is 5 counts/g/s, with a background equivalent to 0.3 g of ^{240}Pu . Thus ISAF demonstrates high efficiencies both for passive and active NDA applications.

The measured response of ISAF for different irregular materials from a light water fuel fabrication plant contained in 5-gal. pails is shown in Figure 9.33. These materials have different total weights and different fissile contents (0.7 to 3 percent enrichment). The accuracy with which the fissile content is determined based on these curves ranges from 3.5 to 5 percent. The different slopes and approach to saturation manifested in Figure 9.33 clearly show the dependence of the instrument response on the density of the sample matrix material.

Figure 9.33. Comparison between responses from different types of fuel scrap using ISAF.

9.5.3 The Los Alamos Scientific Laboratory Random Driver

The Los Alamos Scientific Laboratory (LASL) coincidence system, called the Random Driver (Ref. 31), uses an Am-Li source ($\sim 5 \times 10^5$ n/s). The detectors are two plastic slab scintillators (made of Pilot F) that detect mostly prompt fission neutrons, the resolving time is about 100 ns. Because the Am-Li source neutrons have an average energy below 1 MeV (see Chapter 4), they induce fission mostly in the fissile material of the sample. The distance between detectors can be varied to accommodate samples up to 5 gal. in size. Lead and boron can be interposed between the sample and the detectors to reduce neutron and γ -ray self-absorption. The source can be moved vertically to traverse the rotated sample. Two ^3He detectors can monitor the thermal neutron flux to correct for the effects of a hydrogen-containing matrix that increases the fission rate as a result of increased thermalization of source neutrons. In a later version, shown schematically in Figures 9.34 and 9.35, four fixed Am-Li sources, rather than a single moving one, are used in order to obtain a more uniform vertical response.

The Random Driver (RD) was designed to minimize the contribution of prompt fission γ -rays, unlike ISAS and ISAF which were designed to sense both prompt neutrons and γ -rays (but they can, when desired, suppress the response of either radiation). The design of the RD stems from the desire to obtain a linear response curve for high-density samples

Figure 9.34. Cutaway view of Random Driver Mod-III. The two ^3He proportional counters located along diagonal corners of the sample cavity are not shown.

Figure 9.35. Top view of Random Driver Mod-III. The instrument will accommodate samples up to 153-mm-diam by 365-mm-high.

with high fissile content, accepting the penalty of lower instrument sensitivity by a factor of 5 to 10. In the RD, γ -ray coincidences are reduced by lead-shielding and then rejected by a time-of-flight discrimination circuit (Figure 9.36) that senses only those events separated by 5 to 40 ns (Ref. 32), thus excluding coincident γ -rays which have a multiple flight-time of about 2 to 5 ns from sample to scintillator (see Chapter 8). The lead-shielding interposed between sample and detectors reduces the passive γ -ray signals from the sample, from the Am-Li source, and lowers the accidental coincidences. It also increases the neutron detection efficiency by reflecting source neutrons and makes neutron detection less sensitive to the variations in the heavy element matrix in the samples. However, excessive thicknesses of lead or other moderators will reduce the energy of the stimulated neutrons and, therefore, their detection probability.

Figure 9.37 shows calibration curves obtained by using an early version of the RD (Ref. 33) for 2-L bottles containing U_3O_8 . The source strength of this system was 2×10^5 n/s. The "fast neutron interrogation" curve was obtained by using the bare Am-Li source. The "thermal neutron interrogation" curve was obtained by using the Am-Li source enclosed in a 1.5-in. (3.75 cm) by 2-in. (5 cm) by 4.5-in. (11.25 cm) block of polythylene. Both curves were corrected by the ^3He monitor for matrix effects in the samples. The increased sensitivity, due to thermalization of the source neutrons, was accompanied by some loss in linearity, presumably because the increased thermal neutron absorption in the larger samples was not completely corrected for by the monitor. Figure 9.38 shows the assay precision of this system for both thermal and fast neutron interrogation modes.

Figure 9.39 shows a calibration curve for a set of HTGR standards made of carbon-coated $\text{ThC-}^{235}\text{UC}$ fuel particles obtained with the Random Driver Mod-III (Ref. 34). A count time of 1000 s typically gave an assay precision of 1 percent for a sample whose fissile mass was 300 g, that is similar to the performance shown in Figure 9.38. The slightly nonlinear response curve, R_c was fitted to the function

$$R_c = \frac{aM}{1 + bM} \quad (9-2)$$

where M is the fissile mass and a and b are fitted constants. Based on this calibration, the results of the Random Driver assay for this fuel agreed, to within a few percent, with the chemical assay. The precision of these measurements (1000 s each) was about 0.5 percent.

Figure 9.36. Block diagram for the electronics for the Random Driver Mod-III.

The gate generator output is 35 ns and delays A and B are 5 ns, yielding a coincidence gate of 40 ns, and resolving time of 80 ns. Delays C and D are 155 ns, resulting in an accidental time window from 155 to 190 ns.

Figure 9.37. Calibration curves for both fast and thermal neutron interrogation. These curves have been normalized to the ^3He flux monitor.

9.5.4 National Nuclear Corporation Random Driver

The National Nuclear Corporation (NNC) coincidence system, designed for 5-gal. samples, has four plastic detectors and a source segment which surround the sample in a five-sided array. Pu-Li sources of $\sim 10^6$ n/s are used. In some models, two sources (top and bottom) are used; in others, a single central source with a special FMD is claimed, by its manufacturer, to achieve uniform response. Usually the 3-out-of-4 coincidence mode is used to detect neutrons and γ -rays with a resolving time of 60 ns. Other systems are designed for sample sizes that range from $\sim \text{cm}^3$ to ~ 55 gal.

A detailed description and evaluation of the earliest 5-gal. NNC system is given (Ref. 35). This system can be used either passively, responding to spontaneous fissions in ^{238}Pu , ^{240}Pu , or ^{242}Pu , or actively, responding to induced fissions in ^{235}U , ^{239}Pu , or ^{241}Pu . The substantial coincidence “background” from spontaneous fissions must be corrected for, when the system is used in the active mode.

Figure 9.40 shows 3-out-of-4 coincidence rate measurements with a partially thermalized neutron source with and without a low-Z matrix

Figure 9.38. Assay precision for various amounts of uranium using the LASL random source interrogation system with a 2×10^5 n/s Am-Li source and 50 ns coincidence gate. Assay precision is defined as the ratio of the standard deviation of the assay to the assay value.

surrounding the sample, and with and without a removable Pb-B shield in front of the detectors. These shields reduce fissions in the samples caused by back-scattered, thermalized neutrons and reduce detection of low-energy fission γ -rays. All the curves are linear, indicating, in this case, absence of neutron or γ -ray self-absorption. Figure 9.41 shows similar curves obtained in the 2-out-of-4 mode. The 3-out-of-4 mode is less sensitive (lower counts per gram fissile) than the 2-out-of-4, but it exhibits a more linear response to the fissile content in the presence of the low-Z matrix.

9.5.5 Comparison of Fast Coincidence Systems

Because all ANDA systems based on the FMD (ISAS, Random Driver, and ISAF) are fundamentally similar, they can, in principle, be transformed from one to another. ISAS uses an externally tailored,

Figure 9.39. Calibration curve for HTGR fuel particles based on weighted least squares fit of data on 14 standards ($a = 0.2834$ counts/s/g, $b = 2.127 \times 10^{-5}$ /g).

Figure 9.40. Active assay 3-out-of-4 coincidences.

Figure 9.41. Active assay 2-out-of-4 coincidences.

high-energy source (^{252}Cf) to induce fissions and a four-detector FMD to measure both prompt neutrons and γ -rays. ISAF uses an internally moderated or nonmoderated low-energy source (Am-Li) and a three-detector FMD that also measures neutrons and γ -rays. The Random Driver uses internal low-energy sources (Am-Li or $^{238}\text{Pu-Li}$) and a FMD with two detectors (LASL) or four detectors (NNC) that measure prompt neutrons but partially suppress signals from prompt γ -rays despite a substantial loss in detection efficiency. Suppression of prompt γ -rays, which provide most of the coincident signals from fission events (see Chapter 8), may sometimes be desirable when absorption of mainly low-energy γ -rays in large and dense samples (for example, kilogram samples of UO_2 or thorium) can occur. Thus, if this portion of the signature is suppressed, for example, by interposing a lead shield between sample and detectors, the assay results will be less dependent on sample density. However, a lead shield that is too thick (>5 cm) can reduce the fission neutron energy below the threshold level of the scintillator. When dense samples of fissile materials are measured, the low-energy portion of

the interrogating neutron spectrum should be removed by interposing a neutron absorber (for example, a borated, or better, a lithiated hydrogen-containing shield) between the source and the sample and between the sample and the plastic (hydrogen-containing) scintillators. Neutron and γ -ray absorbers, in principle, can be added to or removed from each of the systems discussed. This important flexibility allows for a strong signal and a resulting high sensitivity for those samples which are not subject to either neutron or γ -ray self-shielding. Conversely, for samples in which self-shielding is significant and the calibration curve becomes strongly nonlinear, proper absorbers of neutrons and/or γ -rays will greatly improve response.

Detection of different multiplicities by the FMD leads to another independent method of reducing the attenuation effects of the prompt fission γ -rays in the sample. For example, it is shown in Chapter 8 that the ratio of the product of all possible combinations of coincidences between two detectors in a three-detector FMD to the threefold coincidence rate is relatively independent of sample attenuation. Similar expressions can be derived using different levels of coincidences.

When the γ -ray attenuation is constant or unimportant, the same mentioned ratios give information on fast neutron multiplication within the sample. When a fast neutron from a fission event (stimulated or spontaneous) causes another fission, it happens within the time resolution of the coincidence circuits (30 to 80 ns) and the apparent multiplicity of the event is greatly increased. The higher multiplicity favors high-level coincidences (for example, 3-out-of-4 or 3-out-of-3) over lower level coincidences (for example, 2-out-of-4 or 2-out-of-3). Thus, the ratio of high-level to low-level coincidences can provide an indication of self-multiplication (Ref. 36), which is important for samples with high fissile content.

Various implications of the number of detectors in the FMD, for example, two, three, four, or even more, have been discussed in Chapter 8. Generally, for high multiplicities (for example, prompt fission γ -rays with or without prompt neutrons), using more detectors increases considerably detection efficiency. However, if only neutrons are to be detected (low multiplicity), it does not make sense to use FMD with more than three detectors, since more detectors will only lead to higher accidental coincidence rates with little increase in true coincidence efficiency.

9.5.6 Active-Well Coincidence Counters

The passive neutron-well slow coincidence counter has been described (see Section 8.6.2), where it was pointed out that this device is useful for

plutonium assay since it responds to spontaneous fission neutrons, mainly from ^{240}Pu . Passive coincidence counters of this type cannot be used for uranium bulk-sample assay because of the very low spontaneous fission rates in uranium. An instrument that combines an Am-Li neutron interrogation source with a ^3He thermal neutron-well coincidence detector has recently been designed and tested at Los Alamos (Ref. 36). It is called the active-well coincidence counter, and it is intended for use in the assay of ^{233}U or ^{235}U . The advantages of this instrument over conventional fast coincidence FMDs are its light weight, portability,* increased stability, and smaller sensitivity to matrix effects, and γ -ray backgrounds. Its insensitivity to γ -ray background should make the device particularly suitable for ^{233}U -Th cycle fuel materials, which often have high γ -ray backgrounds from the decay of ^{232}U . The main disadvantages of the instrument are its limited sensitivity due to the limit of the source strength (which is determined by the rate of the chance coincidences) and its more complex electronics.

Normally, an interrogation source used with a thermal neutron-well coincidence counter results in extremely large accidental coincidence count rates because of the long (20 to 100 μs) coincidence gate lengths. In the active-well coincidence counter, the Am-Li source is placed inside a moderating shield, and the operator attempts to take advantage of the greater transmission of the induced prompt fission neutrons (~ 2 MeV average energy) as compared with the source neutrons (< 0.4 MeV) to reduce the accidental rate. The source shield, which partially shields the source neutrons from the ^3He detectors, has an open channel to the sample in the well. In this manner, the ratio of accidental coincidences due to the source neutrons to the true coincidence rate due to induced fissions in the sample is reduced by more than an order of magnitude as compared with the unshielded source.

9.6 DELAYED γ -RAYS SYSTEMS

In addition to prompt neutrons, prompt γ -rays, and delayed neutrons, several delayed γ -rays are emitted following fission as a result of radioactive decay of fission fragments. In the first minutes after fission, approximately six to eight such delayed γ -rays, with average energies of the order of 1 MeV, are emitted. In this section, systems that are based on the detections of delayed γ -rays will be described. They include fuel rod scanners and the small sample assay system (SSAS).

*FMDs were not designed, so far, with light weight and portability as design criteria. Inherently, neutron/ γ -ray FMD is not heavy, since it is based on plastic scintillators, hence it can be made relatively light and portable.

The assay of nonirradiated fuel rods by so-called rod scanners often uses delayed γ -rays. Fuel rod scanners are used in industry for two important functions:

1. To accurately determine total fissile content in the rod.
2. To determine pellet-to-pellet variations in the fissile content.

The first function is required for safeguards, quality, and process control. Prompt neutrons, delayed neutrons, or delayed γ -rays are used to determine total fissile content. The second function is required mainly for plant quality assurance. Because very high counting rates are required for good statistical precision, delayed γ -rays or intense γ -rays (for example, those emitted by radioactive decay of ^{241}Am in mixed oxide rods) are used to determine pellet-to-pellet variation in practically all systems.

To measure total fissile content of a fuel rod, a fuel rod scanner should satisfy a number of criteria, including high reliability and low operating cost. The most important criterion, however, is that the response above background be due entirely to the fissile material in the rod. For instance, if delayed neutrons are detected, it is necessary to insure that there is little or no response to γ -rays (natural or induced), to neutrons from spontaneous fission in the fuel, or to fissions in the fertile isotopes in the fuel. Other effects which might perturb responses include the following: thermal neutron absorption by fuel impurities, effects of variations in cladding thickness on neutron absorption, effects of "crosstalk"* among different fuel rods being assayed simultaneously and end-effects.** Such perturbations can be minimized by careful design (Ref. 37) (see Chapter 10).

In a typical rod scanner, a ^{252}Cf source irradiates a section of the fuel rod. The rod is driven through the system at a uniform speed on the order of several centimeters/second. The source strength is in the range of 10^8 to 10^{10} n/s (50-5000 μg ^{252}Cf). The source neutrons are moderated by a large moderating assembly, usually made of polyethylene or more exotic materials, for example, D_2O -impregnated resin, designed to maximize the fissile-fertile fission ratio (see Chapter 6). The moderator is surrounded by a thick neutron and γ -ray shield. The entire

*Crosstalk is an effect caused by (1) γ -rays from one rod being counted by the detector for an adjacent rod, or (2) the fast neutrons created in one rod causing fissions in another.

**End effects are caused by the fact that each end of the rod obviously is not shielded by fuel material, thus signal and background are different from the rest of the rod.

configuration is designed to provide the highest flux in the moderator consistent with low γ -radiation, from the source, at the counting position. The shield is typically about 2 m in diameter. Figure 9.42 shows a schematic of a representative rod scanner.

Maintaining uniform rod-drive speed requires precision drive such as that afforded by the screw, belt, pincher roller, or sprocket chain. Pusher rod or a combination of pusher rod and crawler belts has been used as well as endless belt carrying fuel rod through irradiator and detectors. The scanner is typically 15 m in length for LWR fuel rods (including the fuel rod input and unloading sections). Usually, several rods (from two to six) can be assayed simultaneously by running them through separate channels placed symmetrically around the source.

The delayed γ -rays are detected from a collimated section (one to several pellets long) of the rod as it travels past a γ -ray detector consisting of either one or more NaI crystals or plastic scintillators. Fast neutron detectors are often also used to monitor prompt fission neutrons as a measure of the total fissile content of the rod.

Figure 9.43 shows typical delayed γ -ray scans from a rod scanner (of the type shown in Fig. 9.42) assaying fuel rods containing anomalously enriched pellets. The data readout is usually designed to sound an alarm

Figure 9.42. Schematic diagram of the pin and pellet assay system (PAPAS). The direction of fuel rod travel is from right to left. (WEP: Water extended polyester resin made by Ashland Oil Co.)

Figure 9.43. A typical delayed γ -ray scan of a fuel rod. The scan is of a 3.3% enriched PWR fuel rod, 66-in. (165-cm) long, with pellets of lower enrichments interspersed as shown above. The lower curve is a smoothed version of the raw data in the upper curve (the error bars represent 2σ uncertainties). Each point represents the total counts accumulated in 0.4 s for a rod-feed rate of 8 ft/min (244 cm/min).

or reject the rod, when an off-spec pellet is detected. Off-spec pellets are those whose fissile content deviates by more than the specification from the nominal value. Typical specifications of LWR pellet enrichment are ± 10 percent. Deviations from specifications can occur if the size, density, or enrichment of the pellet are incorrect. Size and density variations are usually determined separately by γ -ray transmission measurement techniques discussed briefly in Chapter 5 and later in this section.

The ability of a rod scanner to reject an off-spec pellet but not a good pellet is a strong function of γ -ray counting statistics. Typical requirements are that a pellet that is 10 percent off specification be detected and rejected 95 percent of the time and that no more than 5 percent of the fuel rods be falsely rejected for containing off-spec pellets. Since there may be several hundred pellets in a rod, the false rejection rate must be less than one in several thousand pellets. False rejection occurs when a count corresponding to a particular pellet is anomalously high or low because of statistical fluctuation in the count rate, or other reasons unrelated to the fissile content. (Section 9.6.5 contains a more detailed discussion of this topic.)

High throughput is necessary in rod scanners, since they are normally used to monitor the entire production run in fuel fabrication plants. This requirement places severe demands on the design of the rod scanner, especially on its counting system. Very fast detectors and electronics are required to obtain the high count totals necessary for good counting statistics. Highly efficient detectors are also necessary (but some, such as

NaI, are slow), therefore, special designs must be employed for the system to perform satisfactorily. On-line computers are often also included to reduce and analyze the count data and give immediate indication of off-spec elements.

Many rod scanners also measure γ -ray transmission through the rod as it moves past a fixed, highly collimated γ -ray source, usually ^{137}Cs . Transmission through the sample is a function of the sample's mass or density (see Chapter 3). Therefore, this method affords an easy and accurate measure of density variations in the fuel rod. By monitoring transmission as the rod moves past the γ -ray source, pellets of incorrect length and gaps between pellets can also be detected. The total length of the fuel column in the rod can also be determined accurately.

9.6.1 General Electric Rod Scanner (Ref. 38)

The first fully developed, high-speed fuel rod (or pin) scanner was designed and built by the General Electric Vallecitos Nuclear Center and has been in use at the G.E. Wilmington fuel fabrication plant since 1972. This system, "Fat Albert," assays BWR fuel rods using a 4-mg ^{252}Cf source and NE-102 plastic scintillators with fast photomultiplier tubes. There are six fuel rod channels. A 1-Ci ^{137}Cs source serves to measure the density uniformity of the fuel rods. Finally, a pair of NaI detectors monitors ^{238}U daughter activity and background resulting from any residual delayed γ -ray activity, if rods are rescanned. The system uses fast (20 ns) electronics to handle the high count rates (50,000 to 500,000 counts/s). Figure 9.44A shows a view of "Fat Albert" in which the six fuel rod channels and detector photomultipliers can be seen. An overall view of the system is shown in Figure 9.44B.

Calibration is performed by placing a standard rod in the system and entering a standard number in the data analysis computer. The measured values for enrichment, density, and total ^{235}U content are automatically compared to their standard values, and the computer software then adjusts the constants of the calibration model to recalibrate the system. The rod transit time is about 4 min/rod and the throughput is approximately 90 rods/hr. Crosstalk effects are on the order of maximum 5 percent, when a natural uranium rod is next to two rods containing 3 percent ^{235}U enrichment.

9.6.2 Fast Breeder Reactor (FBR) Rod Scanner

A rod scanner for assaying fast breeder reactor type fuel elements has been designed and constructed at LASL and has been used at the Westinghouse-Hanford Engineering Development Laboratory (HEDL)

Figure 9.44A. General Electric fuel rod scanner. The photograph shows several rods emerging from the irradiator after passing in front of the delayed γ -ray detectors, of which four are shown. (Courtesy of the Nuclear Fuel Division, General Electric, Wilmington, S.C.)

Figure 9.44B. General Electric fuel rod scanner: overall view showing the irradiator and the unloading racks. (Courtesy of the Nuclear Fuel Division, General Electric, Wilmington, S.C.)

plutonium fuel facility in Richland, Washington (Ref. 39). This instrument may be employed for both active and passive NDA. For the active method, a 200- to 600- μg ^{252}Cf source (Figure 9.45) is placed in the center of the moderator, and the fuel rod moves past it and into the detectors, two 5-in. (12.5 cm) by 5-in. (12.5 cm) NaI crystals with through-holes. For determination of total fissile (plutonium) content, the high-energy delayed γ -rays ($E_\gamma > 1.2$ MeV) are detected. They have high penetration through the rod and a high signal-to-background ratio.

For pellet-to-pellet analysis, the same detectors are used to examine different portions of the low-energy γ -ray spectrum. A 0.2-in. (0.5-cm) collimator is used. One detector counts 60-keV γ -rays (mainly from ^{241}Am in the fuel), while the other counts 100 to 500-keV γ -rays (primarily from plutonium). The intensity of the ^{241}Am line, which is much higher than the higher energy lines, acts as a batch monitor since the ^{241}Am content (the decay product of ^{241}Pu) of the fuel is time-dependent. If all pellets in the rod are made of the same batch, variations in the ^{241}Am activity will indicate variations in the fissile content.

Figure 9.45. Schematic diagram of the ^{252}Cf fast neutron assay system for FBR-type rods. The delayed γ -rays induced by fast neutron irradiation are subsequently counted by the two NaI detectors, which also measure passive γ -rays, to determine pellet-to-pellet uniformity (Ref. 39).

The fuel rod is first moved through the NaI detectors (toward the source) to obtain a background count. Then the motion direction is reversed, so that the detectors count the delayed high-energy γ -rays after irradiation by the ^{252}Cf source. Figure 9.46 shows the rod scanner response to a mixed-oxide fuel rod containing pellets with different Pu enrichments.

The delayed γ -ray response is nonlinear with respect to total plutonium content because of a neutron flux depression that occurs when the Pu content becomes large. A calibration function of the type $R = A(1 - e^{-BM})$ represents the net count rate R , as a function of total Pu mass, M . A and B are fitted constants obtained by counting standard rods with known fissile contents. Since A depends on the strength of the ^{252}Cf source, frequent recalibration is required.

In general, this machine determines total fissile content in these fuel elements with a total accuracy of better than 1 percent, comprising of

Figure 9.46. FFTF fuel pin with various combinations of plutonium enrichments for pellet-to-pellet scanning. Top curve corresponds to 60-keV energy window and bottom curve corresponds to 100- to 500-keV window.

The small enrichment variation, 1.7 to 3.1% relative, can be easily observed in the top curve but not so readily in the bottom curve (Ref. 39).

less than 0.2 percent systematic and less than 1 percent random errors. Table 9-9 shows a comparison of the rod scanner assay with chemical assays on a number of fuel rods.

Recently National Nuclear Corp. (see Section 9.6.4) has built a scanner for Westinghouse-HEDL for the assay of FFTF fuel. This design is patterned on the LASL unit described above. Total fissile is measured using fast neutron interrogation with fissile content being determined by measuring delayed gammas of over 1.2 MeV. Pellet-pellet variation is determined using passive counting of the ^{239}Pu gammas. In order to minimize the effect of the ^{241}Pu daughter ^{237}U , provisions are made to eliminate or subtract counts of less than 330 keV. This system has a total of 10 counters in parallel in the "passive" detection system. These counters include a pellet densitometer, an ^{241}Am detector and eight ^{239}Pu detectors. While these are known as passive detectors, actually about 50 percent of the counts come from active interrogation (fission products). A 2-mg ^{252}Cf source is used with this system.

9.6.3 Pin and Pellet Assay System (PAPAS)

Figure 9.42, shown before, is a schematic of the Los Alamos pin and pellet assay system (PAPAS) (Ref. 40), designed to assay LWR fuel rods. This machine uses energy-biased ^4He counters to detect prompt fission neutrons for total fissile content and delayed γ -rays for pellet-to-pellet analysis. The delayed γ -rays are detected by a 2-in. (5-cm) by 2-in. (5-cm) NaI detector with a 0.75-in. (1.9-cm) through-hole. The detector is located near the end of each of the four fuel rod channels. The rod scan rate is 8 ft/min (4 cm/s). With a 500- μg ^{252}Cf source, delayed γ -ray count rates of about 88,000 counts/s are typically obtained from 3.3 percent enriched PWR fuel rod. Figure 9.43 shows a pellet-to-pellet scan obtained with PAPAS for a PWR fuel rod containing a number of pellets of anomalously low- and high-enrichments.

PAPAS determines total fissile content to better than 1 percent and detects a nominally 3.3 percent enriched pellet, which is approximately 7 percent off-spec, about 97 percent of the time.

9.6.4 Commercial Rod Scanners

National Nuclear Co. has built two types of rod scanners. One of these (Ref. 41), uses a small FMD assembly made of four plastic scintillators through which the fuel rod moves. Since prompt coincidences due to thermal neutron fissions are the signal detected by the FMD, it must be exposed to a very high neutron and gamma fluxes. These fluxes and the captured γ -rays associated with the thermal neutron flux create a very

Table 9-9. Los Alamos FBR rod scanner: precision nondestructive assay versus destructive chemical assay of fuel pins (g fissile Pu).

Conditions		(A) Vendor Chemical Assay	(B) HEDL Chemical Assay ^a	(C) HEDL Fuel Pin Scanner ^b	(A-B) Shipper- Receiver Difference	(A-C) Shipper- Receiver Difference	(B-C) HEDL Difference
Inner Core (15 Pins)	Totals	448.18	447.68	447.82	+0.50	+0.36	-0.14
	Mean/pin	29.879	29.845	29.855	(+0.11%)	(+0.08%)	(-0.03%)
	Std dev	0.2380	0.2261	0.2319	_c	_c	_c
Outer Core (34 Pins)	Totals	1246.16	1240.83	1244.72	+5.43	+1.44	-3.89
	Mean/pin	36.652	36.495	36.609	(+0.43%)	(+0.12%)	(-0.31%)
	Std dev	0.3254	0.2853	0.2640	_c	_c	_c

^aHEDL chemical assay values are based on averages of three to six pellet samples/pin.

^bHEDL fuel pin scanner values are based on averages of 6 scans/pin.

^cNot a significant difference.

high background of uncorrelated single events, which may limit the applicability range of the device. However, by a judicious choice of source intensity (typically < 0.5 mg of ^{252}Cf), source-detector distance, and degree of coincidence measured (for example, 3-out-of-4), a useful and relatively inexpensive system results that is applicable to cases where scanning speed and throughput requirements are not too demanding. For high throughput cases, a "conventional" approach has been taken, namely a strong ^{252}Cf source (2 mg) and delayed γ -ray detection downstream with NaI detectors for pellet-to-pellet variation. Moderated delayed neutron detection with ^3He counters is used far from the source zone for total fissile content determination. The accuracy claimed for this determination is 0.25 percent at 1σ confidence level. This type of rod scanner is claimed to routinely detect 10 percent off-spec pellets in LWR fuel rods scanned at a speed of about 25 ft/min (750 cm/min) or higher with 95 percent confidence level and 5 percent false rejection.

IRT Corporation has designed and built several fuel rod scanners since 1972. Stimulated delayed γ -rays measured with multiple NaI detectors are used for the determination of pellet-to-pellet variation and of total fissile content. Californium-252 neutron sources of 1 to 2 mg are used in the IRT two-channel system. The claimed accuracy for the total fissile content is less than 1 percent (at 2σ level). Scanner specifications for detection of discrepant pellets with ± 10 percent variation in the fissile content, at scanning speeds of 25 to 35 ft/min, are similar to the value stated above for the NNC scanner.

The commercial systems use a minicomputer to perform on-line all the required analyses and to decide on the acceptance or rejection of fuel rods. Data analysis involves some smoothing and unfolding (Ref. 42) as discussed in the next section.

9.6.5 Statistical Considerations in Rod Scanners

The problem of off-spec pellet rejection has been briefly alluded to in the introduction to this chapter. We now examine it in more detail. Consider first, a simple model in which a succession of pellets moves past a detector. Suppose each pellet is counted, one at a time, and the successive counts are recorded. If the pellets are perfectly uniform, the counts will be statistically normally* distributed about some mean value N , with a standard deviation, σ , equal to $\sqrt{N}$. The rod scanner is set to reject pellets whose enrichments are typically 10 percent high (or low). If

*Counting statistics for radiation emissions usually have a Poisson distribution. The distinction between this and the normal distribution is especially important at the tail regions. We use here the normal distribution for the sake of simplicity.

a 10 percent high pellet is detected, its counts are expected to be drawn from another normal distribution whose mean is $1.1N$ with a standard deviation equal to $\sqrt{1.1N} \approx \sqrt{N}$. These two distributions are sketched in Figure 9.47. It should be noted that these distributions result purely from the counting statistics.

Suppose one desires to reject 10 percent off-spec pellets with 95 percent confidence. The reject point must then be set at N_1 which is determined by requiring that the shaded area under curve II from N_1 to infinity be 95 percent of the total area under curve II. N_1 is thus approximately 1.65σ below the mean of curve II. If the separation between curves I and II is such that the distance of N_1 from the mean (N) of curve I is, for instance, only 1σ , then there is an approximately 33 percent probability that a good pellet would give a count rate high enough to be rejected. For good pellets to be rejected at a rate of, for instance, no more than 1 in 20,000 (corresponding to a 1 percent rod reject rate, if there are 200 pellets/rod), it would be necessary that $N_1 - N$ be about 4σ . Thus $(1.1N - N)$ must be approximately $5.65\sigma = 5.65\sqrt{N}$, which means $N = 5.65\sqrt{N}/0.1$, or $N = 3200$.

This analysis assumes that the system counts one pellet at a time. In fact, the pellets are moving past the detector, and the detector is responding to more than one pellet at any instant. Suppose it "sees" three pellets. Then, instead of responding to a 10 percent difference, the system must respond to a 3.3 percent increase in count rate. (The expected total count of three good pellets is N' , and of two good plus one bad pellet is $1.033N'$). Because we still require a spacing of 5.65σ , N' must now be about 32,000.

To obtain the necessary high throughput of rods, the scanning speed must be several pellets/second, and high count rates are required to meet both of the rejection criteria. In PAPAS, for example, the count rate for

Figure 9.47. Distributions of normal pellets and "high" pellets.

PWR fuel rods is about 90,000 counts/s. The estimated count rate in the IRT scanner is about 300,000–500,000 counts/s. As has been mentioned, this may place severe demands on the detectors and electronics.

The problem may be ameliorated by various methods of “smoothing” the raw count data. The main purpose of these methods is to enhance the signal from off-spec pellets, which is embedded in a large statistical (and nonstatistical) noise. Smoothing and other techniques (Ref. 40, 42) range from simple summing of successive count totals to more sophisticated analyses that use correlation functions that are based on the measured response of the detector to a single pellet.

It should be emphasized again that the simple considerations outlined rest on two assumptions: (1) that counting variations are due only to counting statistics (and not to rod speed variations, vibrations, unstable electronics, and others), and (2) that only pellets with a minimum off-spec enrichment are rejected. It should be clear, for example, in the illustration given, that a large fraction of pellets whose enrichment is between the specified enrichment and the off-spec value (10 percent) will also be rejected. It is always necessary to demonstrate experimentally that the design criteria are in fact satisfied by a particular rod scanner.

Examples of unsmoothed and smoothed data are shown in Figures 9.43 and 9.48A-D (Ref. 42). Figure 9.43 shows a scan of an LWR fuel

Figure 9.48. A, B: Strip chart records of data obtained from a good rod. A correction has been applied for the end pellets in both the smoothed and unsmoothed data. C, D: Strip chart records from a bad rod showing unsmoothed and smoothed data (Ref. 42).

rod with some anomalous pellets interspersed within it. In Figure 9.48 parts A and B are shown raw and smoothed data obtained with an early version of the IRT scanner from a good rod. Parts C and D show data from another rod containing some off-spec pellets. The effectiveness of the smoothing in suppressing the statistical "noise" is evident, although there is also some spreading of the apparent deviation over several pellets.

9.6.6 High-Precision Small-Sample Assay System (SSAS)

Another ANDA system which uses delayed γ -rays very efficiently is the small sample assay system (SSAS) built by IRT (Ref. 43). The system is based on the principle of thermal (or epithermal) neutron activation analysis, where the delayed γ -rays emitted from fission products a few seconds to a few minutes after fission are the observed response.

During system development, four techniques were investigated to determine which would routinely give ± 0.5 percent relative precision measurements, within a short period of time, when low-enrichment UO_2 fuel pellets were measured. The following techniques were investigated:

1. Passive measurement of the 185-keV γ -rays emitted by ^{235}U .
2. Active measurement using neutron interrogation with either coincidence counting techniques for detection of prompt fission events (that is, neutron and γ -rays) or prompt neutron detection using detectors with an energy threshold.
3. Activation analysis using delayed neutron measurement.
4. Activation analysis using delayed γ -ray measurement.

The first two techniques were rejected because of their low counting rates, excessively long counting times, and/or too large backgrounds. The third technique was rejected because it requires a large (3- to 10-mg) ^{252}Cf source, therefore, its costs became prohibitive. The fourth technique proved to be very successful. This approach takes advantage of the high initial yields of delayed γ -rays by starting the counting very shortly after the end of the irradiation (see Chapter 7).

The SSAS operates with a basic timing cycle, which begins with a background measurement, continues by irradiating the sample in the neutron flux of a properly moderated ^{252}Cf source, and concludes with a measurement of the signal plus background. The basic physics of the system, which makes efficient use of the delayed γ -spectrum and time-dependence, yields very high count rates/gram of fissile material, even when moderately small Cf sources (10 to 50 μg) are used. A

simplified sketch showing the main features of an IRT SSAS is shown in Figure 9.49. The instrument allows efficient use of the delayed γ -ray signature while maintaining simplicity and transportability.

The SSAS can be operated with either dead-timeless electronics (current measurements, see Chapter 8) or with fast linear electronics that retain pulse height information. The latter was found to be very helpful, especially for Pu-U mixed oxide pellets.

Extensive tests to determine the precision of the SSAS were performed with sintered LWR pellets and natural to 3.3 percent enriched UO_2 powder (Ref. 43). Results showed that the precision typically obtained for pellets and for oxide powder, with variable bulk density, is better than about 0.5 and 1 percent, respectively, at the $1\text{-}\sigma$ confidence level.

Another small sample assay system based on delayed γ -detection has been designed by LASL (Ref. 44). This instrument, called MECAS (multi-energy californium assay system), has the unique capability to irradiate the sample by three different neutron spectra, each with a different degree of hardness. This allows determination of both the fertile and fissile content of the sample with small self-attenuation effects. Extensive performance results from this system are not yet available, but preliminary runs with BWR fuel pellets showed agreement with chemical assay between 0.5 and 1 percent at 1σ level.

VIEW WITH OUTER COVER REMOVED. AUTOMATIC SAMPLE
LOADING AND UNLOADING TRAYS NOT SHOWN

Figure 9.49. Simplified sketch of the Gulf Rad Tech (now IRT Corp. San Diego, CA) high-precision small-sample assay system.

9.7 OTHER ANDA METHODS

In closing this chapter, we briefly mention several other ANDA techniques which have been investigated or used. These include the lead slowing-down spectrometer, methods based on the use of nuclear reactors as neutron sources, and x-ray fluorescence.

9.7.1 The Lead Slowing-Down Spectrometer

The lead slowing-down spectrometer is a large (1 to 8 m) solid block of pure lead, into which a pulse of fast (for example, 14 MeV or photoneutrons) neutrons is introduced. Shortly after the burst the average neutron energy of fast neutrons decreases rather slowly with time, and the energies of most of the neutrons do not deviate much from the average energy. Thus, at least to a first approximation, the neutrons slow down as a group. If a fissionable sample and a prompt neutron detector are placed in the block, the count rate variation with time, after the neutron pulse is introduced, will depend on the fission cross section variation with neutron energy. This dependence allows for both detection of the fissionable material and isotopic discrimination. Total fissile content can be determined by comparison with standards. Figure 9.50 is a schematic of a lead slowing-down spectrometer used for fuel pin assay (Ref. 45). A pulsed 14-MeV generator is the neutron source. Neutrons are detected with methane proportional recoil counters.

In another application of the lead slowing-down spectrometer (Ref. 46), the prompt fission neutrons from ^{235}U and ^{239}Pu in fuel elements were detected, at two different times, when the mean interrogating neutron energies were about 0.3 eV and 0.025 eV. Because of the strong resonance in the fission cross section of ^{239}Pu , the counts at the time corresponding to an interrogating neutron energy of 0.3 eV come mostly from the ^{239}Pu in the sample, while those at the time corresponding to 0.025-eV neutron energy come from both of the fissile isotopes, if self-shielding can be neglected. If the counting rates at both interrogating neutron energies are measured for a standard rod containing known amounts of both isotopes, one can determine the ^{235}U and ^{239}Pu contents of the unknown rod. The accuracy in this work was better than 5 percent for a measurement which lasted 30 min.

The system (Ref. 46) uses a 2-m by 2-m by 2-m lead pile. The fuel pin is brought to the machine and unloaded into a magazine on the input side. There is a similar magazine on the output side. These magazines have a capacity of 100 pins and act as a buffer to decouple the spectrometer from the production line. The fuel pin to be measured is automatically fed into the spectrometer and pushed through it at constant speed. As it enters, the neutron generator is switched on and as

Figure 9.50. Setup of neutron slowing-down-time spectrometer. The sample location $1/3L$ reduces neutron flux harmonics, which facilitates theoretical calculations (Ref. 45).

the fuel pin leaves, the generator is switched off, and the data analysis system is switched on. For data analysis, an on-line computer compares the count rates with stored count rates from a standard rod. $^{10}\text{BF}_3$ counters are used to monitor the neutron source strength, that is also normalized to the reference rod data. The computer then determines the fissile ^{235}U and ^{239}Pu content of the rod and classifies the rod as "good" or "bad." This method can be used either for mixed oxide fuel elements or to determine the burnup of UO_2 elements.

As has been mentioned, the use of the 0.3 eV and thermal energy regions may pose problems because of the strong self-shielding effects at 0.3 eV and the long period between pulses required to allow for the die away of the thermal neutrons. In an early work (Ref. 47), it was shown by calculations that by using the high-energy sub-MeV portion of the slowing neutron spectrum, which is established in the lead spectrometer within a few microseconds, one can obtain a good discrimination ratio

between ^{239}Pu and ^{235}U (for example, factor of 2) without self-shielding. Also, the neutron flux is much higher at these early times, that helps to compensate for the low fission cross sections. The high-energy sub-MeV approach was successfully tested on the Rensselaer intense spectrometer (RINS) (Ref. 48) using fuel pellets of ^{233}U - ^{233}Th .

A later lead slowing-down spectrometer studied in detail by calculations the region of 50 keV to few eV (Ref. 49). These results suggest that a high discrimination between ^{235}U and ^{239}Pu can be obtained, if the energy regions around 30 and 10 eV are utilized. The results also indicate that for typical LWR fuel elements, 1 percent counting statistics can be obtained in assays of less than 10 min.

9.7.2 Reactor Methods

Neutrons from a nuclear reactor can be used for interrogation in active assays. In one application of this method, at Gulf Radiation Technology, samples were irradiated in the outer fuel ring of a TRIGA reactor (Ref. 50). A rapid pneumatic transfer system then moved the samples to a counting station where delayed neutrons were detected with $^{10}\text{BF}_3$ counters. ^{232}Th and ^{238}U contents were also measured by detecting γ -rays from the neutron activation products, ^{233}Pa and ^{239}Np , with Ge(Li) or NaI detectors. A later application of this method, including the preparation of samples and standards, is given in (Ref. 51). Typically, the sample to be assayed is irradiated in the reactor for 30 s, then it is removed from the reactor for a 30-s decay period, followed by a 50-s counting interval. The delayed neutrons are moderated by a polyethylene or paraffin block and are detected by a bank of ^3He thermal neutron counters. Accuracies of better than 0.5 percent in the determination of ^{235}U and about 1 percent for ^{232}Th have been achieved relative to the standards. Precision was a factor of 2 to 5 or better. To assay thorium (in the presence of ^{235}U), a thick ^{10}B shield is necessary around the sample during the reactor irradiation to minimize thermal neutron activation in the ^{235}U . Very high sensitivities are obtained for ^{235}U with 2×10^{-10} to 10^{-11} g of ^{235}U , depending on whether steady-state or pulsed irradiation was used.

Another reactor-based method determines the reactivity worth of a sample (the change in the reactor multiplication factor) due to the presence of the sample (Ref. 52) that can be related to its fissionable content. This technique (Ref. 53) does not require a high-flux reactor. It has been used with a zero-power critical assembly, the advanced reactivity measuring facility (ARMF) (Ref. 54, 55) and even with a subcritical assembly plus a ^{252}Cf source (Ref. 56).

9.7.3 X-Ray Fluorescence

In x-ray fluorescence (Ref. 57), a γ -ray source, for example, ^{109}Cd , irradiates the sample and induces fluorescent x-rays characteristic of the elements present in the sample. The x-rays are detected with high resolution solid-state detectors such as $\text{Si}(\text{Li})$. For samples diluted with water, sensitivities of about 10 ppm U or Pu can be reached. However, for dense samples, only the surface material can be analyzed because of the extremely large self-absorption of x-rays. Discrimination between isotopes of the same element is not possible, since the x-rays come from transitions between orbital electron energy levels and do not originate in nuclear processes.

9.7.4 Other Techniques

Few other generic techniques that were not discussed in this chapter are cited in the literature: resonance self-indication (Ref. 58), resonance transmission (Ref. 59), and active calorimeter (Ref. 60).

9.8 SUMMARY

Many types of ANDA systems have been investigated. Systems which respond to delayed neutron signatures utilize either 14-MeV source neutrons from pulsed accelerators (either Cockcroft-Walton or Van de Graaff) or use isotopic sources which intermittently irradiate the sample (step irradiation techniques). Accelerator sources are characterized by high intensities, high on-off ratios, and good discrimination ratios. Isotopic sources, such as ^{252}Cf , are smaller, and their instruments are less complex and more reliable. Delayed neutron detection is generally characterized by fair sensitivities. They can be used when γ -ray backgrounds are high, as in irradiated fuel, but this requires very strong sources.

Systems based on prompt neutron detection either utilize energy-biased fast neutron detectors to distinguish fission neutrons from source neutrons, or use fast coincidence techniques to take advantage of the high multiplicities of prompt fission neutrons and γ -rays. Energy-biased neutron detectors are used with either low-energy sources (for example, Am-Li or Sb-Be) or with high-energy sources (for example, ^{252}Cf plus spectrum-tailoring to reduce the interrogation energy). Fission multiplicity detectors (FMD) can respond either to prompt neutrons alone or to a combination of prompt neutrons and prompt γ -rays, they can be made relatively insensitive to matrix effects. The active-well counter can also be designed to assay samples with high γ -ray backgrounds.

Linacs provide Bremsstrahlung radiation that can induce photofission in the sample or can be converted to a neutron source by the (γ, n) reaction in a convertor such as D_2O . With photofission, either the delayed neutrons can be detected or both the prompt and delayed neutrons can be detected (in different segments of the time interval between accelerator pulses). High sensitivities and discrimination ratios can be obtained, but the Linac is, like other accelerators, complex and expensive.

To determine total fissile content of fuel rods and to examine pellet-to-pellet variations in enrichment, size, or density, fuel rod scanners are the instrument of choice. These contain an isotopic (^{252}Cf) source plus a spectrum-tailoring assembly through which the fuel rod passes. Total fissile content is measured by prompt or delayed neutrons or delayed γ -rays. Pellet-to-pellet variations are usually detected by delayed γ -rays. Rod scanners are characterized by high throughputs and the ability to detect reliably and reject rods containing anomalous pellets, and maintaining an adequately low false-rejection rate.

Other less widely used ANDA techniques include lead slowing-down spectrometry, delayed neutron detection following fission induced in a nuclear reactor, measurement of reactivity worth in a reactor, and others. x-Ray fluorescence is widely used as a supplement or alternative to analytical chemical destructive assay.

Table 9-10 summarizes some qualitative features of the various ANDA methods. Examples of nuclear materials common in the nuclear fuel cycle and measured with ANDA are listed in Table 1-5 in Chapter 1 and in Chapter 10.

Table 9-10. Qualitative characteristics of representative ANDA systems.

Signatures	Sources	Detectors	Advantages	Drawbacks	Some Representative Systems
Delayed neutrons	Spectrum tailored 14 MeV, Van de Graaff, $E_n < 1$ MeV Tailored ^{252}Cf , Linac photofission and/or photoneutrons switchable Sb Be	Moderated thermal neutron detectors, e.g., BF_3 or 3He "Slab Detector"	Is specific to fissile materials, no interference from source neutrons, applicable to highly γ radioactive ("hot") samples	Requires very high source intensity, hence shielding is heavy, handling complicated, and costs high Isotopic sources require "on off" switching mechanism Very high "on off" ratio	MONAL (LASL), GAMAS (GA IRT) Cf Shuffler
Prompt neutrons	Low energy (α, n) sources, Sb Be Tailored (highly moderated) high energy sources (e.g., ^{252}Cf , 14 MeV), Linac photofission	Fast neutron detectors gas proportional counter 4He , H, fast scintillators, threshold fission chamber can be used with intense sources (e.g., Cf) Moderated neutron detectors as in (1) can be used with Linac	High sensitivity and flexibility, applicable to "hot" sample	Strong spectrum moderation is required for tailored system, thus susceptibility to neutron self shielding	Sb Be (GA, LASL, EURATOM, Julich, etc.) Total Fissile rod scanner (LASL), GAMAS (GA IRT), Spent LWR Assembly, system (EPRI SAI)

continued

Table 9-10. Qualitative characteristics of representative ANDA systems (continued).

Signatures	Sources	Detectors	Advantages	Drawbacks	Some Representative Systems
Fission multiplicity (FMD)					
Fast	Low energy (α, n) sources (e.g., Am-Li, Pu-Li) Tailored and collimated $^{252}\text{Cf}_{(\alpha, n)}$ sources.	Fast scintillators (2, 3, 4, or more) in coincidence with or without neutron and/or γ -ray filters, fast coincidence electronics	Flexible, good sensitivity, relatively simple, transportable, can tolerate relatively high neutron background, reasonably inexpensive, delivers wealth of useful information, can determine Pu isotopics	Not applicable to "hot" samples, interrogation source intensity is limited.	ISAS (GA-IRT) ISAF (GA-IRT) Random Driver (LASL, NNC, TN)
Slow	Directionally shielded low energy (α, n) sources	Moderated neutron detectors using slow coincidence circuit ("well counter").	Increased stability and transportability, applicable to "hot" samples	Limited sensitivity because of inherent limitations on source intensity. Applicable only to samples with low neutron background.	Active-well counter (LASL).
Delayed γ -rays	Isotopic sources—most common ^{252}Cf (accelerator neutron sources also possible).	NaI(Tl) or plastic scintillators with fast electronics	Very high sensitivity, suitable for on-line applications	Not applicable to "hot" samples, susceptible to γ -ray self-attenuation.	Large systems, Rod scanners (GE, LASL, IRT, NNC) Small systems: SSAS (IRT), MECAS (LASL).

Commercial Vendors

NNC National Nuclear Corp., Redwood City, CA
 IRT IRT Corp., San Diego, CA
 TN Texas Nuclear Corp., Austin, TX

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CHAPTER 10

INSTRUMENT ANALYSIS, CALIBRATION, AND MEASUREMENT CONTROL FOR ANDA

10.1 INTRODUCTION

Ideally, the ANDA analyst would like to specify and procure an instrument especially suited for his particular measurement problem, however, for a number of reasons, this is not always possible. First, only a limited number of ANDA instruments are manufactured, and procuring the ideal instrument may mean requesting a custom design. Second, even a custom-designed ANDA system cannot optimally solve all measurement problems, even in a single setting. In certain situations, the analyst must compromise by getting an assay instrument flexible enough to solve numerous measurement problems but not optimized for any one.

The application of ANDA requires careful and sometimes extensive effort in three areas: analysis of the instrument's sensitivity to a host of parameters, preparation of representative and unbiased calibration standards, and monitoring the performance of ANDA by, for example, comparison of ANDA measurements with those of independent assays to assure measurement control. These three tasks, performed by the ANDA analyst, are as important to accurate ANDA as the design and construction of a high-quality, fundamentally sound, ANDA instrument. Careful analysis, calibration, and measurement control are likely to result in highly accurate ANDA, even when the instrument is applied to diverse measurement problems. On the other hand, the analyst's carelessness and shortcutting can result in inaccurate ANDA, even for an instrument ideally suited for a particular application.

This chapter provides advice on instrument analysis, calibration, and measurement control. This advice relies on the understanding of the physical process discussed in the previous chapters and on some

knowledge of the nature of the materials contained in the tested sample. Further, the advice is based on the reported experience of previous ANDA users, and it is illustrated with examples taken from the literature cited in this and previous chapters.

Instrument analysis, calibration, and measurement control are discussed together because they are closely related. Instrument analysis teaches the analyst about the important parameters that influence the active radiation response. This information is required for preparation of calibration standards because preparing a complete set of standards representing all unknown materials is impossible. Practical calibration standards attempt to simulate unknown samples in those aspects that significantly influence the response. Even then, preparation of calibration standards requires considerable effort and is expensive. Instrument analysis also provides information needed for establishing a measurement control program. The approximate error model for ANDA, determined in the analysis of the instrument, defines the requirements for replicate analysis and the control standards necessary for proper measurement. Finally, part of a quality measurement control program is comparison of the ANDA results with those of assays by independent methods, destructive or nondestructive. Results of these comparisons should be fed back to the calibration program to correct biases and add calibration data points.

It is not possible to provide examples and specific discussions of all ANDA methods and applications. Therefore, the following sections treat generally each of the topics. However, an attempt has been made to refer mainly, throughout this chapter, to specific examples for two types of ANDA: the fission multiplicity detector (FMD) techniques for measurement of small to medium containers of nuclear material, and the delayed γ -ray scanning of fuel rods and pellets.

10.2 ANDA INSTRUMENT ANALYSIS

The instrument analysis required for proper application of any measurement technique is also necessary for the design of quality instrumentation. Instrument analysis is equally mandatory for the analyst who wishes to perform an assay of a specific material in a particular container. The principles of ANDA and the representative ANDA systems presented in previous chapters of this book provide the background for such an analysis.

In this section, we discuss seven parameters that must be analyzed because of their potential influence on the ANDA response. This section ends by describing how results of analyses are combined in an attempt to

estimate total ANDA performance. This discussion is not complete because it identifies only the parameters of major importance:

1. Response Profile—the change in the efficiency of interrogation and detection as a function of location within the counting chamber or container, or along an item being scanned.
2. Fissile/fertile response ratio—the ratio of responses from equal amounts of fissile and fertile nuclides.
3. Cladding and container effects—the attenuation or moderation of interrogating or stimulated γ -rays and neutrons passing through the cladding or container.
4. Matrix effects—the attenuation or moderation of interrogating or stimulated γ -rays and neutrons passing through the matrix materials.*
5. Self-shielding effects—the attenuation of γ -rays and neutrons by the nuclear material that is being measured.
6. Multiplication—the increase in radiation emission per gram of material caused by fission of nuclear materials induced by other stimulated radiation rather than the interrogating radiation.
7. Cross-talk in multiple channel systems—radiation leakage from one counting or interrogating channel to another (such as in fuel rod scanners) during assay.

Other parameters not listed here are (a) the influence of background neutrons and γ -rays, (b) instrument instability, (c) the relative abundances of radionuclides, and (d) end effects in fuel rod scanners. Background and instability, problems common to all measurements, are discussed briefly in Chapters 8 and 9. The abundances of radionuclides are more difficult parameters to measure,** however, their effects are so great that they must be accurately known for each assay.

* Matrix material comprises all the elements in the sample except the actinides to be measured (thorium, uranium, and plutonium) and those elements that are chemically bound to or mixed with the actinides.

** Distinction among the fertile isotopes (^{232}Th , ^{238}U , ^{238}Pu , ^{240}Pu) and their fissile counterparts is feasible and can be made by some of the available ANDA systems. Some of the fertile isotopes that have sufficiently high spontaneous fission rates can be sometimes distinguished from the other fertile isotopes through coincidence counting. It is much more difficult to distinguish among the fissile isotopes, but the delayed neutron yield may provide the desired signature to separate ^{235}U from ^{239}Pu (see Chapter 7).

10.2.1 Response Profile

The interrogation efficiency of ANDA systems can be defined as the probability of detecting a unit mass of fissile material placed in the sample chamber of the ANDA system. This efficiency includes all the factors that influence the response of the system to the presence of fissile material.

For a better understanding of the design of an ANDA system or of its operational characteristics, it is instructive to specifically identify the two components that together form the interrogation efficiency. The first one is the detection efficiency, that is, the probability that the system's detectors measuring a fission event occurring within the sample chamber. This is commonly measured by moving a small spontaneous fission source (for example, ^{252}Cf or ^{240}Pu)* over the entire volume of a "typical" sample known to have the matrix materials and other features expected in the unknown sample. Such a measurement provides the experimenter with a profile of the detection efficiency. The second component of the interrogation efficiency of an ANDA system is the fissioning efficiency, that is, the probability of inducing fissions in a unit mass of fissile (or fertile) material. Because it is proportional to the neutron flux, one can determine the fissioning efficiency by measuring the flux.

The interrogation efficiency itself can be measured directly by moving a unit mass of fissile material over the volume of a "typical" or standard sample, thus furnishing a profile of the instrument's response to the fissile material. The degree of response nonuniformity varies greatly among different systems, depending on the type of system and its configuration. In large systems, such as those for barrel assay, the detection efficiency alone may vary by 10 to 20 percent (Ref. 1). The variation in fissioning efficiency increases the total nonuniformity of the interrogation efficiency to more than 20 percent. Systems that measure smaller containers (~ 4 L) show smaller variations in interrogation efficiencies. Well-designed systems show typical variations with the position of the source of ~ 5 percent (Ref. 2, 3). Even systems designed to measure small samples show significant efficiency variation (Ref. 4). If all assayed material is homogeneous, the variation in efficiency will not influence the assay. Nonuniformity of response, due to the uneven distribution of fissile material within the sample, may introduce measurement errors, therefore, every effort must be made when assaying

*In quantitative efficiency measurements based on the multiplicity of particles emitted in fission events, ^{240}Pu is preferred because its fission multiplicity is much more representative of the fissionable isotopes. The higher fission multiplicity of ^{252}Cf can lead to overestimation of detection efficiency (see Chapter 7).

heterogeneous samples to obtain as uniform a response as possible. Unfortunately, in most applications the material has inhomogeneities that may otherwise lead to significant errors in the assay.

Because measuring changes in interrogation efficiency as a function of sample position are straightforward, this is one of the first tests that should be performed for an ANDA system. Experiments should be carried out to determine the interrogation efficiency at a few points, calculations may then be made to determine the efficiency at other points. The tests consist of two types: differential and integral response measurements.

Figure 10.1 shows a differential interrogation response along the vertical axis of the isotopic source assay system (ISAS) (see Chapter 9). This measurement was done while moving a small vial of highly enriched U_3O_8 along the vertical axis of the system. The signal recorded was the 3-out-of-4 coincidences. The response of a large sample can be obtained by integrating this response curve over the height of the sample. Similar

Figure 10.1. Differential vertical response profile (ISAS).
The rectangles at the top indicate the relative position of the four detectors. Also indicated are the widths at 90%, 2.7 in. (6.75 cm) and 50%, 7.1 in. (17.8 cm) of the maximum response.

measurements across the horizontal plane between the detectors (see Chapter 9) showed little response variation. Figure 10.2 shows an integral vertical response profile for the Random Driver. The relatively flat axial response is the result of spacing apart the two interrogation sources. Figure 9.31 shows a simpler method for obtaining flat response, albeit, over a somewhat smaller axial segment. This integral response profile was measured directly by filling the container with nuclear material and measuring the response per gram as a function of fill height.

The type of measurements described give the user a complete interrogation response profile for the instrument. Upon concluding that a sample with a particular nonuniform distribution of fissile material cannot be adequately measured by the instrument, the user can take steps to reduce the effects of the nonuniformity. One of the most common methods is to rotate the sample during irradiation, thus reducing the effects of radial nonuniformities in fissile material. To reduce or eliminate the effects of axial nonuniformity (for example, containers filled to different levels) the sample may be scanned axially when it is moved over the entire volume of the detection system. Appendix C covers this point.

Figure 10.2. Plot of relative vertical response versus sample fill height. The flatness in response for fill heights between 75 and 200 mm was achieved by vertically separating the sources as shown.

Effective axial scanning requires two conditions. The first is zero response at the initial and final scanning points. Although this condition can always be met, it can add more background to the total signal. However, this deleterious effect can be reduced, if the interrogation response profile is as flat as possible and declines sharply at the initial and final scanning points.

The other condition for effective axial scanning—equal response per unit fissile mass and per unit length or height in the calibration standard and unknown sample—is more difficult to reach, especially when high accuracies are required. A case in point is the scanning of fuel rods, where the interrogation response changes near the ends of the rod. The effect is demonstrated in Figure 10.3, where the solid curve shows the true nonlinear response of a fuel rod scanner. The dotted line shows a hypothetical linear response that represents the change in response with rod length for a fixed enrichment or fissile concentration. The solid curve was obtained by using calibration standard rods of a single length. The rod with mass m_0 has the same length (ℓ_0) as the standards and is accurately measured. Assuming that a rod of the same enrichment as m_0

Figure 10.3. Response of hypothetical fuel rods of length ℓ_0 , ℓ_1 , and ℓ_2 versus ^{235}U content of m_0 , $m_0\ell_1/\ell_0$, and $m_1\ell_2/\ell_0$ of the rods.

but of a shorter length (ℓ_1) is measured, the true mass is $(\ell_1/\ell_0)m_0$; but the assay gives an erroneous mass m_1 . The ANDA device effectively interprets the rods as having a lower enrichment than m_0 rather than a smaller length. An opposite effect occurs for longer (ℓ_2) rods. Rod scanners have reported end effects of approximately ± 0.8 percent for a 1 percent length change (Ref. 5). This error can be reduced by determining the actual active length of the fuel rod, that is, the length of the fissile column of the fuel rod. The actual active length can be deduced either from the profile of the signal as measured by the rod scanner or by separate γ -ray densitometer measurement (see Section 9.6).

10.2.2 Fissile/Fertile Response Ratio

Fissions in fertile nuclides may produce a significant response in ANDA systems, especially, if high energy neutron interrogation is used. The response ratio can be calculated if the interrogating neutron energy spectrum is known. The fission cross sections for the fissile and fertile nuclides are averaged over the neutron energy spectrum, and the ratio is computed as follows:

$$\text{Response ratio} \left(\frac{\text{fissile}}{\text{fertile}} \right) = \frac{\int \Phi(E) \sigma_{\text{fissile}}(E) dE}{\int \Phi(E) \sigma_{\text{fertile}}(E) dE} \quad (10-1)$$

where $\Phi(E)dE$ is the neutron flux between energies E and $(E + dE)$, and $\sigma_{\text{fissile}}(E)$ and $\sigma_{\text{fertile}}(E)$ are, respectively, the fissile and fertile nuclide fission cross sections in the same energy region.

Extensive calculations of the fissile to fertile response ratio are reported in Chapter 6 for different composite moderators and interrogation sources. These calculations can be verified by experimentally measuring the ratio using pure materials or, less directly, using materials of different enrichments. Light water reactor (LWR), low-enrichment uranium fuel rod scanners have calculated and measured fissile/fertile response ratios of 10^4 to 10^5 . Thus, even for natural uranium, the fertile ^{238}U contributes only 1.4 to 0.14 percent to the response (Ref. 5).

Because the fertile nuclides can be stimulated to fission by fission neutrons from the fissile nuclides, the fissile/fertile response ratio should be measured in all ANDA systems, even in those using interrogating neutrons with energies below the effective fission threshold. For example, in the assay of a uranium-thorium mixture using 300- to 600-keV neutrons from a Van de Graaff accelerator, the thorium in the sample will increase the response by 0.12 percent/gram of thorium (Ref. 4). Another example which shows the importance of the fissile/fertile response ratio is in the assay of uranium mixtures using sub-MeV

neutrons induced by Bremsstrahlung 4-MeV electrons (see Chapter 5 and Section 6.6.) Although most of the interrogating neutrons are well below 1 MeV for a thick D_2O target, the presence of ^{238}U increases the response of 1 g of ^{235}U by about 0.06, and 0.15 percent per gram of ^{238}U when prompt and delayed neutron signatures are used, respectively (Ref. 6).

10.2.3 Cladding and Container Effects

The size, shape, composition, and thickness of the containers of nuclear materials influence the performance of ANDA systems. Probably, the most important characteristics of containers are their size and shape because these dictate, to some extent, the size and the shape of the system. They affect, above all, the systems' efficiency. For some ANDA systems, the detection efficiency is inversely proportional to the fourth power of its size! The larger the ANDA system, the more difficult it is to attain uniform response to the special nuclear materials (SNM).*

Other important effects on measurements due to the container are attenuation, scattering, and spectral modifications (for example, from the slowing of neutrons) of the passive signature, the interrogation, and the stimulated radiations. Interaction of the radiations with the container material can also create background radiation, for example, by neutron capture.

Thus, containers affect:

- efficiency of detection,
- uniformity of response,
- attenuation of interrogation and signature radiations,
- scattering of interrogation and signature radiations,
- spectral modification of interrogation and signature radiations, and
- background.

Because of these effects, the containers and cladding enclosing the ANDA calibration standards should always be the same as those used for material routinely assayed. Even then, however, unavoidable variations in the mass of a container and/or the thickness of a container's wall or cladding may affect the assay. Table 10-1 (Ref. 7) shows the change in the response from a subthreshold fission multiplicity instrument (Random Driver), when plastic rather than aluminum containers are used. The 3He detector of the instrument, which measures the neutron moderation caused by the plastic container, provides data used to correct the

*Special Nuclear Materials (for example, ^{233}U , ^{235}U , ^{239}Pu , ^{241}Pu).

Table 10-1. Effect of container composition
on a Random Driver assay.

Plastic Container Volume (gal.)	^{235}U Mass (g)	% Change in Response Relative to Aluminum
1/2	57	+7.8% $\pm$ 1.6%
1/2	69	+4.6% $\pm$ 2.7%
1/2	154	+5.5% $\pm$ 1.2%
		Mean = +6.0% $\pm$ 1.0%
1	111	-4.3% $\pm$ 1.1%
1	157	-8.1% $\pm$ 1.5%
1	167	-3.4% $\pm$ 0.7%
		Mean = -5.3% $\pm$ 1.4%

response. The table shows, however, that even with this correction, the response is not the same with different containers. Nor is there a constant bias between aluminum and plastic, as might be expected: the 0.5-gal. container has a positive bias relative to aluminum, but the 1.0-gal. container has a negative bias. The response, then, is affected by both the mass and by the diameter of the container. In the example of 1-gal. container, analysis will probably show that if measurements more accurate than ± 6 percent are required, a single type of container must be used.

The change in ANDA response due to changes in the wall thickness of 10-L polyethylene bottles filled with highly enriched uranyl nitrate, as determined by ISAS, is shown in Table 10-2. In all cases (using hard epithermal, soft epithermal, and thermal neutron spectra) the extra polyethylene increased the response per unit mass of fissile material.

Table 10-2. Change in response caused by a 10% change in wall thickness of a 10-L polyethylene bottle of uranium bearing solution, measured by ISAS.

Spectrum	Response Change (%)	
	10% Change Inside Diameter	10% Change Outside Diameter
"hard" (boron liner, borated paraffin collimator)	(0.69 $\pm$ 0.10)	(0.19 $\pm$ 0.08)
"epithermal" (no boron liner, borated paraffin collimator)	(0.79 $\pm$ 0.16)	(0.18 $\pm$ 0.09)
"thermal" (no boron liner, unborated paraffin collimator)	(0.80 $\pm$ 0.05)	(0.09 $\pm$ 0.10)

For a change in the outside diameter of the bottle, the increase in response is primarily due to the additional moderation of the neutrons by the increased wall thickness. For change in the inside diameter, the increase in response is mostly caused by the increased sample surface area to volume ratio. The data indicate that, for the boron liner and borated paraffin collimator, a 15 percent variation of the wall thickness (keeping the outside diameter constant) causes an approximately 1 percent change in response. Changes in the outside diameter have a much smaller effect.

For more accurate assay systems, such as fuel rod scanners, the effect of minor variations in cladding thickness can be calculated. The change in response of an LWR fuel rod scanner, due to changes in the thickness of stainless steel cladding, has been calculated to be -12.2 percent/mm (-0.31 percent/mil) primarily because of the large thermal neutron absorption cross section of iron (Ref. 5). For Zircaloy-2 cladding, the calculated effect is smaller: -1.14 percent/mm (0.029 percent/mil). The variation in a typical production rod, ± 0.03 mm with Zircaloy-2, causes assay variations of ± 0.03 percent, much smaller than the overall measurement accuracy. The effect of variations of this size in stainless steel containers is ± 0.37 percent.

10.2.4 Matrix Material Effects

Matrix material affects ANDA through attenuation and moderation of both the interrogating and the fission response radiations. Hydrogen in water and in waste materials, such as plastics, causes severe moderation of high energy neutrons. Impurities, such as chlorine, absorb neutrons and release captured γ -rays which create background in coincidence systems. Burnable poisons, such as Gd_2O_3 , that are added to fuel rods make thermal neutron assay nearly impossible.

The effects of matrix moderation and attenuation can be calculated, in a manner similar to the calculation of the fissile/fertile ratio, provided the neutron energy spectrum is known. For example, the cadmium ratio of a ^{235}U detector can be measured to determine the fraction of the interrogating flux in the resonance (epicadmium) and thermal region. A knowledge of the materials which compose the matrix and their distribution is then sufficient for a rough calculation of the size of the resonance and thermal neutron absorption effects. Unfortunately, this knowledge is often very hard to obtain.

Matrix material effects can also be directly measured. Several examples of such measurements are discussed in Chapter 6, primarily, in Sections 6.8 and 6.10. We provide additional examples in this chapter to underline the importance of matrix effects in ANDA applications.

Figure 10.4 shows data from a test on the effect of hydrogen on a Van de Graaff, small sample assay system using delayed neutrons as the signature (see Section 9.2). Hydrogen was introduced by mixing U_3O_8 with dextrose as the hydrogen-containing material (Ref. 4). About 75 mg of hydrogen in small uranium samples raised the estimate of the fissile content by about 8 percent.

The effect of larger quantities of hydrogen-containing material was measured with the Random Driver. The fissile material was contained in HTGR fuel. A batch of HTGR fuel particles were placed in an aluminum container with plastic added to its outer surface. The response of the instrument and the count rate of the thermal flux monitor (^3He detector) were then measured. The results are shown in Table 10-3. The flux ratio (F), which represents the degree of neutron moderation by the sample, is equal to the ratio of the monitor count rate (when the sample is inside the measuring cavity) to the monitor count rate of the empty cavity. The factor k corrects to the first order (Ref. 8) for the softening of the interrogation neutron spectrum caused by the sample. Table 10-3 shows that, for the particular Random Driver used in this test, the practical limit on the flux ratio is about 1.6. Above 1.6, the corrected results are not reliable. The flux ratio is shown in Figure 10.5 as a function of the amount of plastic (polyethylene bags and chips) added to a fixed amount of depleted uranium. The flux correction ratio may be applied here, only when the mass of the plastic is below 200 g.

The magnitude of the neutron moderation effects caused by low- Z matrix materials is also specific to the type of instrument used and to the type, size, and weight of the samples. For example, if the neutron interrogation spectrum used is soft from the outset, the presence of hydrogen-containing material is much less important than when a hard neutron spectrum is used. In systems, which measure primarily the fast prompt neutrons with threshold detectors, such as ^4He or plastic scintillators, the presence of large amounts of high- or intermediate- Z materials (for example, iron, aluminum, various oxides, or lead) can reduce the energy of the stimulated neutrons and substantially diminish their detection probability. Partial correction for the reduced system response may be obtained by a modification of the flux factors (Ref. 9).

Heavy actinide elements (for example, ^{238}U , ^{232}Th) mixed with the fissile nuclear material often cause significant attenuation of neutrons and γ -rays. Special mixtures of the fissile material with different amounts of the heavy actinides can be measured to obtain the data necessary to estimate the size of the effect or to correct for attenuation (Ref. 10).

Figure 10.4. Relative response as a function of sample hydrogen content for various uranium loadings. Samples used were mixtures of U_3O_8 and dextrose.

Table 10-3. Random Driver flux correction factor.

Plastic Mass (g)	Uncorrected Response (Relative to Zero Plastic)	Flux Ratio (F)	Flux Factor $K = (F + 1)/2$	Corrected Response
0	1.018	1.036	1.018	1.000
5	1.029	1.057	1.029	1.000
12.5	1.034	1.090	1.045	0.990
25	1.065	1.149	1.074	0.991
50	1.106	1.270	1.135	0.973
100	1.226	1.616	1.308	0.937
250	1.684	3.220	2.110	0.798
400	2.144	5.516	3.258	0.658

10.2.5 Self-Shielding Effects

Because fissile material attenuates the interrogating radiation, it reduces the amount of fission stimulated in fissile material in the center of the container. Self-shielding can also occur within particles made of highly absorbing fissile materials or other materials. These effects, called self-shielding, can be calculated by using the exponential law for the attenuation of neutrons and γ -rays, if the energy of the radiation and the cross section or mass absorption coefficient are known (see Chapter 6).

Figure 10.5. Measurement of the increase in thermal neutron flux (flux ratio) caused by plastic material.

Self-shielding can also be measured experimentally, but these experiments are not as straightforward as others discussed in this chapter. Self-shielding, within particles, can be measured by preparing particles of different diameters and by arranging the experiment so that the shadowing of one particle by another is negligible or so that the two effects—within particle self-shielding and particle to particle self-shielding—are considered together. Self-shielding of materials in the center of a container can be measured by preparing samples of different diameters or thicknesses. Results of this less direct measurement are shown in Figure 10.6 (Ref. 11). Here, either delayed γ -rays or fast neutrons, resulting from thermal neutron fissions, were counted.

The response of neutrons, which are more penetrating than the delayed γ -rays (see Chapter 7), is less affected by self-shielding in fuel rods than the response of the γ -rays. Self-shielding occurs in the numerous fuel particles distributed along with thorium oxide particles in a carbon matrix. Since the distribution of particles is not fully controlled, the larger self-shielding effect appears as a larger scatter in the delayed γ -ray response when compared to the fast neutron response.

Generally, comparing results of two or more methods, which differ in all or some aspects, offer the experimenter the possibility of determining systematic errors. For example, by comparing the results of the calorimeter measurement, which is virtually matrix independent, and that of a FMD-based system, one can experimentally determine various corrections over a range of sample sizes, containers, matrices, and material densities.

Figure 10.6 Response variations for fast neutron counting as compared with delayed γ -ray counting for thermal-neutron interrogation of HTGR fuel rods.

10.2.6 Multiplication

The complete effects of multiplication (see also Section 6.7) on response are not easy to determine because the multiplication depends partly on how much neutron moderation occurs. However, they can be experimentally determined simply by measuring the change in the specific response due to an increase in the mass, assuming other effects such as matrix moderation, matrix attenuation, and self-shielding are constant or known (see also Section 8.7 on measurement of multiplication in coincidence systems). The approximate size of the multiplication may be calculated roughly by using neutron transport codes. The effects of multiplication are usually included in the calibration because standards, spanning the range of fissile masses expected in the assayed sample, are used.

10.2.7 Cross-Talk

In multiple channel systems, such as fuel rod scanners (see Section 9.6), the effect on the results of assaying simultaneously rods of different enrichments must be determined by placing rods of the same enrichment in all channels and measuring the response of a single rod. The other rods are then replaced with rods of different enrichment, and the response measurement is repeated. Finally, all rods except the rod

being assayed are removed, and the response with empty channels is measured. These data can then be used to compute corrections for cross-talk or to arrive at a more reliable calibration and assay scheme (Ref. 5).

10.2.8 Combined Effects

We have discussed seven parameters that can affect the response:

$$R_{\text{ANDA}} = R(p, f, c, m, s, M, t) \quad (10-2)$$

where

p = response profile

f = fissile/fertile response ratio

c = container and cladding attenuation and moderation

m = matrix attenuation and moderation

s = self-shielding effects caused by increased density, particle size, or container diameter

M = mass of fissile material including the effect of multiplication

t = cross-talk between channels.

The relative change in response from changes in these parameters can be calculated by using the following formula:

$$\frac{\Delta R}{R} = \sum_i \frac{\Delta R/R}{\Delta \rho_i} \Delta \rho_i \quad (10-3)$$

where $(\Delta R/R)/\Delta \rho_i$ is the coefficient for the relative change in response per change in the parameter ρ_i , and $\Delta \rho_i$ is the total change in the parameter ρ_i .

As an example, assume the following coefficients for a Random Driver system measuring ^{235}U in HTGR fuel:

Coefficient	$(\Delta R/R)/\Delta \rho$	$(\Delta R/R)/\Delta f$	$(\Delta R/R)/\Delta c$	$(\Delta R/R)/\Delta m$	$(\Delta R/R)/\Delta s$
Value	0.1	10^{-5}	0.01	0.1	0.05
Units	cm^{-1}	(unit ratio) $^{-1}$	mm^{-1}	g^{-1}	$(100\mu)^{-1}$

Let us also assume that the parameter changes from the mean or average values are as follows:

Parameter	$\Delta \rho$	Δf	Δc	Δm	Δs
Value	0.1	5	1	0.01	10
Entity	fill height	ratio	wall thickness	g Hydrogen	particle size
Units	cm	dimensionless	m	g	μm (micrometer)

The total maximum effect, assuming all effects are in the same direction, is calculated as follows:

$$\frac{\Delta R}{R} = \underbrace{0.01}_{\text{response profile}} + \underbrace{5 \times 10^{-5}}_{\text{fertile material}} + \underbrace{0.1}_{\text{container wall}} + \underbrace{0.001}_{\text{hydrogen in matrix}} + \underbrace{0.005}_{\text{particle size}} \quad (10-4)$$

$$\frac{\Delta R}{R} = 0.02605. \quad (10-5)$$

Thus, here the response is increased by 2.6 percent. If the calibration relationship between the response and the uranium content is approximately linear, then the uranium assay will be biased upward by 2.6 percent.

If changes in the parameters are not unidirectional, but independent, then the root-mean-square rather than the sum provides an estimate of the variation in response due to variations in the parameters:

$$\frac{\Delta R}{R} = \sqrt{(0.01)^2 + (5 \times 10^{-5})^2 + (0.1)^2 + (0.001)^2 + (0.005)^2} \quad (10-6)$$

$$\frac{\Delta R}{R} \simeq \pm 0.014. \quad (10-7)$$

Thus, for this case the expected variation is ± 1.4 percent.

10.3 ANDA CALIBRATION

Calibration is a method to determine the relationship between the observed stimulated fission radiation and the mass of a nuclide or nuclides. The calibration is performed by using calibration standard materials that (1) are similar to the materials to be assayed in all properties affecting the assay, and (2) have nuclide masses that are known as a result of an assay by independent techniques traceable to national standards.

10.3.1 Calibration Standards

Figure 10.7 (Ref. 12) shows a recommended sequence of events for preparing ANDA (or any NDA) calibration standard. This sequence is outlined below:

1. Instrument analysis, as described in the preceding sections, is performed to determine the effect on the response caused by a change in any of the various parameters affecting that response.

Figure 10.7. Typical sequence of events to prepare NDA calibration standards.

2. The material to be measured is sampled and assayed by both destructive and nondestructive tests. Necessary information, such as container composition and wall thickness, may be available from the supplier. Other information, such as chemical composition and moisture content of the material, may be available from process data.
3. The maximum variation in each parameter is multiplied by the response coefficient to determine the maximum effect. These data are assembled so that a decision can be made as to which parameters

should be duplicated in the calibration standards and which ones can be ignored.

4. The calibration standards are prepared under closely controlled conditions. All steps and results are documented and verified.

Calibration standards can be categorized according to the manner in which they are prepared:

- *Synthetic Standards:* Materials are mixed together and formed into the proper geometric configurations to duplicate the unknowns to be assayed. For example, uranium oxide may be mixed with graphite flour to duplicate an ash standard, or uranium oxide may be formed into pellets in a special press and placed in cladding tubes to prepare fuel rod standards. Table 10-4 presents some common matrix materials useful in preparing synthetic calibration standards. The last two columns give, respectively, the thermal neutron cross section and the absorption coefficients of each material for 1-MeV γ -rays.
- *Production Materials Standards:* Materials used for routine and production processes, which have been analyzed and tested by using independent techniques, can be used as standards. For example, fuel particles may be analyzed for elemental content, particle diameter, density, etc., by sampling a large batch. A portion of this batch may then be used as a standard for ANDA. Similarly, fuel pellets from a large batch that has been sampled may be used to prepare fuel rod standards.

Calibration standards can also be categorized according to geometry and homogeneity:

- *Homogeneous Standards:* The standard is uniform throughout its entire volume. For example, pellets of a single enrichment in a fuel rod or a homogeneous distribution of uranium oxide in graphite flour make homogeneous standards. The nuclear content of these standards is fixed. A number of standards must be prepared to duplicate a range of masses. Figure 10.8 shows a homogeneous fuel rod standard (Ref. 13).
- *Modular Standards:* Various small homogeneous standards are mixed together to duplicate a variety of conditions. Examples of modular standards include fuel pellets of different enrichments in a single fuel rod or packages of uranium oxide-graphite mixtures dispersed in a graphite matrix. By combining a limited set of modular standards, a variety of conditions can be simulated. Figure 10.9 shows the fuel column makeup of both homogeneous and modular fuel rod standards (Ref. 14). A modular waste standard is shown in Figure 10.10.

Table 10-4. Common matrix materials for preparation of calibration standards.

Compound	Chemical Form	Z	Density ^a (g/cm ³)	Thermal Neutron Cross Section (b)	1-MeV γ Attenuation Coefficient (cm ² /g)
Diatomaceous earth (celite)	—	—	0.5	—	—
Graphite flour	C	6	1.5	4.8	0.0637
Aluminum oxide	Al ₂ O ₃	13	3	2.84	0.0625
Iron oxide	Fe ₃ O ₄	26	5	10.93	0.0607
Tin oxalate	SnC ₂ O ₄	50	3	4.50	0.0599
Lead oxalate	PbC ₂ O ₄	82	5	9.17	0.0614
Red lead oxide	Pb ₃ O ₄	82	8	10.55	0.0685
Silicone rubber ^b	—	—	1.5	7.26	0.0646

^aThese are powders, so actual bulk density is variable, usually less than value listed here.

^bApproximate composition: H(4%), C(18%), O(28%), Si(25%), Fe(24%), Al, K, Na (<0.2%).

Material Description:

	Thermal Rods	Fast Rod
Cladding material	Zircaloy — 2	Type — 304 Stainless steel
Rod outside dia	0.523 ± 0.004 in.	0.250 ± 0.010 in.
Cladding wall thickness	0.040 ± 0.003 in.	0.015 ± 0.001 in.

Figure 10.8. Encapsulated fuel rod standard.

- **Heterogeneous Standards:** Materials are mixed together to simulate a nonuniform distribution. Often, the mixing process is not planned to systematically produce a certain distribution but, rather, produces a random mixture of ingredients. In effect, the ingredients are “thrown together.” If this process is repeated in many standards, the assumption is that the net result will be a random set of

Figure 10.9. Mixed-oxide fuel pin standards.

Figure 10.10 55 gal. drum test container for waste assay.

heterogeneous standards. This is an attempt to duplicate the unknowns that may also be mixtures that have been “thrown together.” Figure 10.11 presents radiographs of three heterogeneous waste standards (Ref. 13).

Homogeneous, modular standards provide the most control in the calibration process because the analyst has systematically built in the effects he feels are important. Homogeneous standards provide a good reference set of standards to start a calibration. All parameters, except the nuclide content, are prepared at the mean values of the unknown. For example, if the moisture content of the unknown is expected to

Figure 10.11. Radiographs of typical nondestructive waste assay standards.

range from 10 to 50 mg of water per sample, then the standards contain 30 mg of water. A number of standards are prepared in the same way, by changing only the nuclide content.

Experience in preparing fuel rod standards has shown that the use of production material pellets for the standards is a satisfactory approach for both LWR and mixed-oxide rods (Ref. 5, 14, 15, 16). In one case, conditions were carefully controlled and audited during the fabrication of the pellets. A sampling plan was devised prior to fabrication and was followed closely from powder blending through the completion of pressing. The pellet press and other equipment were cleaned to assure proper enrichment. The cladding was selected from in-process stock but was closely inspected to assure uniform wall thickness. Special calibration and auditing procedures were performed on scales and analytical measurements to assure accurate assays of the content of the standards. The procedures included frequent checks of calibration by measuring standards and the use of a number of analytical techniques and/or laboratories.

A similar approach has been used to prepare production scrap standards for calibrating a low-enrichment uranium fission multiplicity detector (Ref. 3). The scrap was first classified into three categories to reduce variations in density within each category. Five standards were prepared in each category by sampling the population of scrap materials and extracting 10 samples from each material for laboratory analysis. The sampling and analytical errors caused an uncertainty of ± 5 percent in relation to the content of the standards. In this particular example, the fill height of the container was controlled to ± 5 percent. The moisture content was also controlled. Therefore, the standards were all basically similar, except for then different uranium enrichments.

10.3.2 Point and Range Calibration

The standard approach to ANDA calibration is to prepare standards with a range of nuclide concentrations or masses and to determine an analytical relationship between the response and a particular concentration or mass. These relationships are rarely linear, most often they are exponential or rational relationships of the following forms:

$$R = A(1 - e^{-BM}) \quad \text{exponential} \quad (10-8)$$

$$R = \frac{AM}{1 + BM} \quad \text{rational} \quad (10-9)$$

where

R = response

M = nuclide mass

A, B = calibration constraints.

There are two limitations when using a range calibration. First, the assay should be performed only on material with masses that fall within the range because extrapolation is not permitted. Second, the extent of the range should be limited to approximately an order of magnitude increase in nuclide concentration or mass. For example, a rod scanner can be calibrated to assay from 0.5 to 5 percent ^{235}U enrichments, or a fission multiplicity scrap assay system can be calibrated to assay from 1 to 10 g or 10 to 100 g of uranium. Extending an accurate calibration range beyond a factor of 10 requires an impractically large number of standards and a complex calibration equation.

The use of a range calibration is convenient when measuring a wide variety of materials. However, if the nuclide mass of the unknown is concentrated at a few points, a point calibration may be more accurate. In point calibration, a number of standards are prepared for each of the masses, and the average response for each mass is determined. The unknowns are then assayed by comparing the response directly to the response of the standard and assuming linearity of the response for the slight deviations in mass about each point.

Point and range calibrations are shown in Figure 10.12. The range calibration provides the best fit of the calibration data from standards with masses M_1 , M_2 , M_3 , and M_4 , provided the assay is performed uniformly on unknowns with mass from M_1 to M_4 . The insert in Figure 10.12 shows the point calibration for unknowns with masses near M_3 . A similar calibration is performed at each of the other masses, M_1 , M_2 , and M_4 . The point calibration is useful and more accurate than a range calibration but only if unknowns with approximately these four discrete

Figure 10.12. Range and point calibration for ANDA.

masses are assayed, and none with masses in between are assayed. The calibration equation obtained from the standards used for the point calibration shown in the insert of Figure 10.12 is of this form:

$$M_u = R_u \left(\frac{\overline{M_s}}{\overline{R_s}} \right) \quad (10-10)$$

where

$$\begin{aligned} M_u &= \text{assayed mass of the unknown} \\ R_u &= \text{response from unknown} \\ \left(\frac{\overline{M_s}}{\overline{R_s}} \right) &= \text{average mass per response from the standards.} \end{aligned}$$

10.3.3 Calibration Data Analysis

Calibration data consist of a set of data points (M_i, R_i) , where M_i is the nuclide mass and R_i is the response for standard (i) . The mass has an uncertainty associated with the analytical techniques and the preparation methods used to make the standard. The response has an uncertainty associated with the precision of the ANDA technique. Theoretically, the uncertainty in the response can be reduced to an insignificantly small value by repeatedly measuring the standard. The mean response is then used for the calibration.

The calibration data set (M_i, R_i) is used to determine the calibration coefficients in the calibration equation:

$$R = f(M, A, B) \quad (10-11)$$

where the function $f(M, A, B)$ may be the exponential or rational relationship, previously discussed, or some other mathematical form. The calibration constants A and B are estimated by a straightforward, least squares method to fit the calibration relationship to the data (M_i, R_i) . The calibration data can be weighted by standard deviations in the data, to obtain the best fit, by defining a function (Q) of the following form (Ref. 12):

$$Q = \frac{D}{S_D} \quad (10-12)$$

Where D is the difference between the measured data and the assumed response function, namely,

$$D = F - f(M, A, B). \quad (10-13)$$

S_D is the variance of this difference:

$$S_D^2 = S_R^2 + \left(\frac{\partial f}{\partial M} \right)^2 S_M^2. \quad (10-14)$$

For example, for the exponential function,

$$\begin{aligned} R &= A(1 - e^{-BM}) \\ D &= R - A(1 - e^{-BM}) \\ S_D^2 &= S_R^2 + A^2 B^2 e^{-2BM} S_M^2. \end{aligned} \quad (10-15)$$

The function (Q) is then fitted to the calibration data points.

As an example, Table 10-5 presents sample data for calibration of a fission multiplicity detector (the Random Driver, here). Fissile mass, response, and the standard deviations for five standards were used to determine calibration constants A and B , their standard deviations, and the covariance (Cov) between A and B . The total variance (S^2) in the mass of material assayed by ANDA can be computed as follows:

$$S_M^2 = \frac{A^2}{(A \cdot RB)^4} S_R^2 + \frac{R^2}{(A \cdot RB)^4} S_A^2 + \frac{R^4}{(A \cdot RB)^4} S_B^2 - \frac{2R^3}{(A \cdot RB)^4} \text{Cov}(A, B). \quad (10-16)$$

For example, if the measured response from an unknown is 21.02 ± 0.09 counts/s, the assay fissile mass is (see Eq. 10-9):

$$M = \frac{R}{A \cdot RB} = 165.81 \text{ g.} \quad (10-17)$$

Table 10-5. Sample ANDA calibration data analysis.

Standard	Fissile Mass $\pm$ Std Dev (g)	Response $\pm$ Std Dev (counts/s)
1	74.51 $\pm$ 0.42	9.89 $\pm$ 0.03
2	115.63 $\pm$ 0.24	14.94 $\pm$ 0.04
3	152.43 $\pm$ 0.66	19.31 $\pm$ 0.03
4	156.45 $\pm$ 0.24	19.75 $\pm$ 0.05
5	345.88 $\pm$ 0.27	41.63 $\pm$ 0.05

NOTE: Calibration Function: $R = AM/(1+BM)$. Result of Weighted Least Squares Fit: $A = 0.13338 \pm 0.00044$ (0.33%); $B = 0.0003145 \pm 0.0000127$ (4.04%); $\text{Cov}(A,B) = 0.5326 \times 10^{-8}$.

The variance in the mass is

$$S_M^2 = 0.56 + 0.33 + 0.12 - 0.38 = 0.63 \quad (10-18)$$

$$S_M = (S_M^2)^{1/2} = (0.63)^{1/2} = 0.79 \text{ g } (=0.48\%). \quad (10-19)$$

The total relative standard deviation of 0.48 percent is composed of the ANDA counting precision standard deviation of 0.45 percent and a calibration error standard deviation of 0.16 percent.

Although this formula can be used to predict the calibration error for individual assays, it is sometimes necessary or convenient to determine a single measure of the calibration error. One measure is the root-mean-square error, or the standard error in the fit. Table 10-6 shows the calculation of this calibration error for data previously presented in Table 10-5. The calibration error is stated as ± 0.91 g or ± 0.95 percent.

10.3.4 Traceability and Documentation

The ANDA calibration is the basis for quantitative assay. The calibration standards preparation and the calibration procedures should therefore be documented and audited to assure that all steps are correct. Figure 10.13 is a sample of a form used to document standards preparation. All laboratory analysis reports would be attached, as well as a radiograph, a γ -ray scan, an x-ray transmission scan, or some other proof of the distributions of the SNM within the container.

Traceability of ANDA to other nuclear measurements is confirmed in three ways. First, the content of standards are measured using analytical techniques that are calibrated with the National Bureau of Standards' Standard Reference Materials because these materials are used as the reference for nuclear measurements. Second, the method of preparing

Table 10-6. Prediction of calibration error
for calibration in Table 10-5.

Fissile Mass of Standard, g		
Assigned Value	Predicted by ANDA	Differences, g
74.51	75.92	+1.41 (+1.9%)
115.63	116.10	+0.47 (+0.41%)
152.43	151.68	-0.75 (-0.49%)
156.45	155.30	-1.15 (-0.74%)
345.88	346.09	±0.21 (+0.06%)
Root-Mean-Square		= ±0.91 (±0.95%)
		$\left(\frac{\sum S^2}{\sum M^2}\right)^{1/2} = \pm 0.47\%$

URANIUM STANDARD CERTIFICATION

DOCUMENTATION AND HISTORY OF MAKE-UP

Type of Standard

Ash
(Particle, ash, insol, combustible waste, etc)

Log Number of Standard

1007

U-235 Mass of Standard

28.64 gms

1) Standard origin (describe origin of standard material, analytical method, analyst, laboratory characterizing standard material. Attach laboratory reports)

(U,Th)O₂, batch SW-85 1001 from scrap recovery. Analyzed by modified Davies-Grey titration for uranium. Four samples, each analyzed in duplicate, performed 11/7/75 by A B Jones, FQIL. Analyzed for uranium isotopic composition by LFE Environmental. Reports attached.

2) Standard composition (list weight % element composition as well as physical and chemical composition and U-235 enrichment)

a Oxide - 26.023 ± 0.028% U, 92.038% U-235, 59.30% Th, 1.08% insols

b Matrix - GE RTV 60 silicone rubber. 1.47 g/cc, 28.5% O, 25.4% Si, 23.7% Fe, 17.5% C, 4.4% H, 0.2% Al, 2% Na or K

c Xylene thinner added to original mix (evaporated)

3) Standard geometry (describe form of standard, container, how made, etc., include drawing as appropriate)

Standard is contained in a 1/2-gallon aluminum wide mouth bottle. Cylindrical geometry. 4.75" (12.07 cm) diameter. 6" (14.25 cm) fill height. Total bottle height = 9" (22.86 cm). The bottle tare = 204.22 gms. gross weight after oxide added = 323.78 (FQIL scale. Mettler K-7 accuracy < 0.3 gms)

4) Uranium content and error (list uranium content, U-235 content and errors associated with each number)

The U content is determined solely by the oxide net weight and the analytical results above. No loss occurred. The uranium is added to the tared bottle and weighed. Xylene is added to wet the oxide and the RTV is mixed in with a wooden stick. All mixing takes place in the bottle. After curing, the small amount of RTV on the stick is removed and added to the bottle. Oxide net = 119.56 ± 26.023% = 31.11 gms U ± 92.038% = 28.64 ± 0.09 gms U-235. Uniformity profile attached.

5) Seal number (violation of seal invalidates standard certification)

Standard Prepared by

Date

Preparation Certified by

Date

Figure 10.13. Uranium standard certification documentation
and history of makeup.

the standards conforms to accepted proven practice, and the instrument analysis substantiates the adequacy of the standards. Finally, the ANDA results should be compared with results, on the same materials, obtained by other independent methods. This final step is part of measurement control, discussed in the next section.

10.4 ANDA MEASUREMENT CONTROL

Measurement control is the process of monitoring the performance of the ANDA to detect biases, so that corrections can be made and to quantify errors needed for the estimation of the uncertainty in ANDA. Measurement control is performed in three ways:

1. Repeating measurements to assure that the results are approximately the same and to quantify the random measurement error.
2. Measuring standards to verify that they have not changed and that they represent material being assayed, to assure that the calibration is still valid, and to quantify bias and systematic error associated with drifts.
3. Comparing ANDA results with assays by independent techniques to assure that the ANDA method is consistent with other nuclear measurement techniques and to quantify biases and systematic errors between techniques.

10.4.1 Random Error in Repeated Measurements

Random error is estimated by repeating the measurement of unknowns and analyzing the data to compute a standard deviation. Because the random error of ANDA is usually a function of the mass of nuclear material assayed, data should be taken on the entire range of material to be assayed. An example of replicate measurements to determine random errors is provided (Ref. 10), that describes how six calibration samples spanning the range of fissile content of interest were measured in an ISAS device over a period of 6 days, one per day. The results are given in Table 10-7 and show that the lowest degree of precision was obtained in samples having a relatively low U content. By adjusting the counting time, one can improve the precision. These measurements indicate that the overall system reproducibility is about 0.7 percent at the 1σ level. The inherent precision for a single determination of 3-out-of-4 rate is between 0.3 to 0.4 percent at the 1σ level. This value was determined by recycling the equipment with a single sample, without any movement of source and sample, a technique required in ISAS for determining the background.

Table 10-7. ISAS precision runs.

Calibration Set	U (g)	Th (g)	1	2	3	4	5	6	Average	σ	σ (%)
A	20.50	707	0.3643	0.3603	0.3540	0.3601	0.3631	0.3605	0.3603	0.0036	1.00
B	25.55	446	0.4920	0.4889	0.4823	0.4949	0.4838	0.4834	0.4875	0.0051	1.05
C	31.80	475	0.5910	0.5941	0.5823	0.5889	0.5917	0.5924	0.5901	0.0042	0.71
D	41.25	542	0.6803	0.6821	0.6849	0.6818	0.6716	0.6844	0.6809	0.0048	0.70
E	46.25	548	0.8866	0.8872	0.8767	0.8651	0.8846	0.8927	0.8838	0.0067	0.76
F	68.40	548	0.9989	0.9990	1.0121	0.9950	1.0070	0.9911	1.0000	0.0077	0.77

10.4.2 Control Standards

The response of an ANDA instrument may drift so that the calibration is no longer valid. For this reason, standards should be measured frequently, both to prevent large drifts and to measure the size of small drifts. Usually, one or more standards may be measured daily or at the beginning of each shift to normalize or standardize the calibration for that day. Other standards are then measured, interspersed with the measurement of the unknowns. Control limits are usually established for both the normalization standard and the control standard assay.

The data from the assay of the standards are averaged to compute a bias over some time period. The establishment and use of control limits prevent this bias from becoming large. The standard deviation in the bias is an estimate of the systematic error that could exist in the ANDA results. An example of the selection of control standards, frequency of measurements, and other related matters is provided in the next section.

10.4.3 Comparative Measurements

The total error in ANDA can be estimated by combining the following errors:

- Unknown Effects: Instrument analysis defines the effects of various parameters on ANDA response. Significant effects that cannot be corrected for are duplicated in the standards. The maximum size of other effects, deemed to be insignificant by the analyst, can be estimated by calculation and measurement.
- Calibration error.
- Random error.
- Bias and systematic error associated with drift.

The combined error is an estimate of the type of agreement expected between ANDA and other techniques. This agreement should be verified through comparative measurements, such as:

1. ANDA of large containers, like barrels, can be compared with ANDA of smaller containers of the same material by dividing the contents of the large container into a number of small units. This comparison is usually valuable because the ANDA of small containers is considerably more accurate than ANDA of large containers.
2. ANDA at one facility or laboratory can be compared with ANDA at other facilities and laboratories.
3. ANDA can be compared with assays by passive NDA, such as calorimetry, passive neutron counting, and passive γ -ray counting.

4. ANDA can be compared with chemical assay. Extensive sampling and homogenization of the material by grinding and blending is usually required.

The results of comparative assays are valuable in confirming error estimates, but they can also be used to improve ANDA calibration (Ref. 17). If the analyst is confident in the comparative assay, the ANDA response and the comparative assay result form a data point that can be added to the original calibration data set. A new least-squares-fit can then be performed.

The study on a Random Driver (Ref. 7) provides an instructive example for instrument analysis, use of control standards, and comparative measurements. Several parameters that influence assay results were studied, including short term drift due to temperature effects, photomultiplier tube fatigue (which causes a slow efficiency decline), neutron moderation by the container, and container fill height. The result of the tests led to several operational instructions: (1) ambient temperature should be controlled to $\pm 2^\circ\text{F}$, (2) fill height should be measured and used in the calculation, (3) only aluminum containers should be used, and (4) a calibration should be made every four hours. The precision of the Random Driver under these controls is shown in Figure 10.14 along with the precision predicted based on counting statistics alone. The observed difference in errors is the main component of the systematic error.

The Random Driver mentioned above was calibrated using seven standards of HTGR particles. Each of three standards was analyzed by

Figure 10.14. Comparison of measured error to calculated precision.

four different analytical techniques: delayed neutron activation analysis, Davies-Gray titration (Ref. 18), x-ray fluorescence, and Van de Graaff small sample assay (see Chapter 9, Section 9.2). The other four standards were analyzed by the Van de Graaff and x-ray fluorescence methods only. Table 10-8 shows the results of comparing the Random Driver assay results with the assay results from sampling and analysis by both Davies-Gray titration and x-ray fluorescence. The second column gives the number of independent batches compared. Columns 3 and 4 give the total ^{235}U masses by each technique, and column 5 gives the relative bias on the total ^{235}U and a 2σ estimate of the uncertainty in the bias. For particle categories 4 and 5, the ^{235}U masses ranged over 200 g; therefore, proportional and constant biases are also recorded. The last two columns show the measured standard deviation of the differences from batch to batch and the standard deviation predicted from the combined errors in the chemical assay and the Random Driver assay. The chemical assay errors include the sampling error determined on three samples from each batch. The Random Driver (RD) error includes the calibration error predicted from the weighted, least-squares-fitting routine. A significant bias is present only for the inner-coated particles (large kernels).

10.5 CONCLUSIONS

ANDA systems generally do not provide absolute results, therefore, the performance of such systems depends greatly on careful instrument analysis and calibration against well-accepted and independent techniques, such as chemical assay of representative samples. When this is properly done, the results of the ANDA measurements are highly reliable, even for samples which could not be measured directly by conventional techniques. A more thorough discussion of the applications of statistical methods in the area of nuclear material control and accountability can be found in J.L. Jaech's book (Ref. 19).

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Table 10-8. Comparison of Random Driver and chemical assay on HTGR coated particles.

Category	Number Batches	Total ^{235}U		Relative Bias + 2σ	Standard Deviation In Differences	
		Chemistry	RD		Measured	Predicted ^a
1. Inner carbon-coated particles (large kernels)	13	5,298	5,368	$+1.30\% \pm 0.88\%$	$\pm 1.59\%$	$\pm 1.48\%$
2. Silicon carbide-coated particles (small kernels)	15	4,795	4,751	$-0.92\% \pm 0.88\%$	$\pm 1.70\%$	$\pm 1.55\%$
3. Silicon carbide-coated particles (large kernels)	14	5,440	5,408	$-0.59\% \pm 1.10\%$	$\pm 1.66\%$	$\pm 1.40\%$
4. Outer carbon-coated particles (small kernels)	6	692.1	687.8	(0.63%) $-1.88\% \pm 5.50\%^b$ $+1.45 \pm 6.0 \text{ g}^c$	$\pm 3.23 \text{ g}$	$\pm 1.44 \text{ g}$
5. Outer carbon-coated particles (large kernels)	7	1,522	1,502	(-1.31%) $-0.39\% \pm 1.20\%^b$ $-2.0 \pm 3.0 \text{ g}^c$	$\pm 1.87 \text{ g}$	$\pm 2.15 \text{ g}$

^aIncludes sampling error for chemical assay and calibration error for Random Driver.^bProportional bias over range of 200 g.^cConstant bias over range of 200 g.

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APPENDIX A

ANALYTICAL EXPRESSIONS FOR FMD EFFICIENCY

The binomial distribution is the fundamental frequency distribution governing random events. This type of distribution is used to arrive at the efficiency of the FMD for a given multiplicity. If the FMD has four detectors, then let ϵ_1 , ϵ_2 , ϵ_3 , and ϵ_4 be the detector efficiencies and q be the probability that *no* interaction will take place in any of the detectors. For an event of multiplicity ν , all possible occurrences are represented by the following expansion:

$$1 = (\epsilon_1 + \epsilon_2 + \epsilon_3 + \epsilon_4 + q)^\nu = \sum \frac{\nu! \epsilon_1^{n_1} \epsilon_2^{n_2} \epsilon_3^{n_3} \epsilon_4^{n_4} q^{n_q}}{n_1! n_2! n_3! n_4! n_q!} \quad (\text{A-1})$$

where the sum is taken over all possible combination of $(n_1, n_2, n_3, n_4, n_q)$ such that

$$n_1 + n_2 + n_3 + n_4 + n_q = \nu. \quad (\text{A-2})$$

The terms in the sum are defined as: n_1 particles are detected in detector no. 1, n_2 particles are detected in detector no. 2, n_3 particles are detected in detector no. 3, and n_4 particles are detected in detector no. 4, while n_q particles escape detection.

The probability that a 4-out-of-4 coincidence (4/4) is recorded is, then, the sum of all terms in the expansion such that the terms n_1, n_2, n_3, n_4 are equal to or greater than 1. Of course, when ν is large, the number of terms involved become large and the evaluation of the various probabilities become difficult. Therefore, it is sometimes easier to find the probability of an occurrence *not* happening and then subtracting this probability from 1. For example, the probability of *not* recording a 4/4 coincidence is the sum of the terms where at least one of the terms n_1, n_2, n_3, n_4 is zero. A useful expansion for this procedure is the following:

$$1 = (\epsilon_1 + \epsilon_2 + \epsilon_3 + \epsilon_4 + q)^\nu \quad (\text{A-3})$$

$$= \nu! \sum_{\ell=0}^{\nu} \sum_{m=0}^{\nu-1} \sum_{n=0}^{\nu-\ell-m} \sum_{j=0}^{\nu-\ell-m-n} \frac{\epsilon_1^{\ell}}{\ell!} \frac{\epsilon_2^m}{m!} \frac{\epsilon_3^n}{n!} \frac{\epsilon_4^j}{j!} \frac{q^{(\nu-\ell-m-n-j)}}{(\nu-\ell-m-n-j)!}.$$

If all the efficiencies of the detectors are assumed equal, the following efficiencies are obtained.

$$\epsilon_{1/4} = 1 - (1 - 4\epsilon)^{\nu} \quad (\text{A-4})$$

$$\epsilon_{2/4} = 1 - 4(1 - 3\epsilon)^{\nu} + 3(1 - 4\epsilon)^{\nu} \quad (\text{A-5})$$

$$\epsilon_{3/4} = 1 - 6(1 - 2\epsilon)^{\nu} + 8(1 - 3\epsilon)^{\nu} - 3(1 - 4\epsilon)^{\nu} \quad (\text{A-6})$$

$$\epsilon_{4/4} = 1 - 4(1 - \epsilon)^{\nu} + 6(1 - 2\epsilon)^{\nu} - 4(1 - 3\epsilon)^{\nu} + (1 - 4\epsilon)^{\nu} \quad (\text{A-7})$$

$$\text{where } \epsilon = \epsilon_1 = \epsilon_2 = \epsilon_3 = \epsilon_4 \text{ and } q = 1 - 4\epsilon. \quad (\text{A-8})$$

Some obvious general limits of the efficiency can easily be derived:

$$\lim_{\nu \rightarrow 0} \epsilon_{\ell/L} = 0 \quad (\text{A-9})$$

$$\lim_{\epsilon \rightarrow 0} \epsilon_{\ell/L} = 0. \quad (\text{A-10})$$

Note also another obvious property of $\epsilon_{\ell/L}$:

$$\epsilon_{\ell/L} = 0 \quad \ell > \nu. \quad (\text{A-11})$$

If ϵ is a small number, which is quite common, the expression for $\epsilon_{\ell/L}$ is simplified. Hence, for $\epsilon \ll 1$ ($\epsilon \lesssim 10^{-2}$):

$$\begin{aligned} \epsilon_{1/4} &\cong 4\nu\epsilon \\ \epsilon_{2/4} &\cong \frac{4!}{2!(4-2)!} \nu(\nu-1)\epsilon^2 = 6\nu(\nu-1)\epsilon^2 \end{aligned} \quad (\text{A-12})$$

$$\epsilon_{3/4} \cong \frac{4!}{3!(4-3)!} \nu(\nu-1)\epsilon^3 = 4\nu(\nu-1)(\nu-2)\epsilon^3 \quad (\text{A-13})$$

$$\epsilon_{4/4} \cong \frac{4!}{4!(4-4)!} \nu(\nu-1)\epsilon^4 = \nu(\nu-1)(\nu-2)(\nu-3)\epsilon^4 \quad (\text{A-14})$$

or generally,

$$\epsilon_{\ell/L} \cong \frac{L!}{\ell!(L-\ell)!} \frac{\nu!}{(\nu-\ell)!} \epsilon^{\ell}. \quad (\text{A-15})$$

These equations have been used in Section 8.7.

APPENDIX B

GENERALIZED EXPRESSIONS FOR COINCIDENCE EFFICIENCIES

Assume that two detectors face a point fissile source. The count rates in each individual detector (N_1 and N_2) and in coincidence (N_{12}) are as follows, assuming $\epsilon \ll 1$:

$$N_1 \cong \bar{\nu} N_f g_1(r_1) \int f(E) e^{-\mu(E)r_1} \epsilon_1(E) dE \quad (\text{B-1})$$

$$N_2 \cong \bar{\nu} N_f g_2(r_2) \int f(E) e^{-\mu(E)r_2} \epsilon_2(E) dE \quad (\text{B-2})$$

$$N_{12} \cong N_f \overline{\nu(\nu-1)} g_1(r_1) \cdot g_2(r_2) \int f e^{-\mu r_1} \epsilon_1 dE \cdot \int f e^{-\mu r_2} \epsilon_2 dE \quad (\text{B-3})$$

where N_f is the number of fissions per second (source intensity), P_ν (Eqs. 8-4 and B-4) is the multiplicity distribution (see Chapter 7), r_1 or r_2 is a position variable, g_i is the solid angle subtended by the point fissile sample at the i -th detector, $f(E)$ or f is the energy distribution of the prompt fission photons, $\epsilon_i(E)$ or ϵ_i is the intrinsic efficiency of the i -th detector, and $\mu(E)$ or μ is the photon macroscopic removal cross section of the matrix material around the small fissile sample. Two other definitions are best given in equation form:

$$\bar{\nu} = \sum_{\nu=1} P_\nu \cdot \nu \quad \overline{\nu(\nu-1)} = \sum_{\nu=2} P_\nu \nu(\nu-1). \quad (\text{B-4})$$

This leads to:

$$N_1 \cdot N_2 / N_{12} \cong N_f (\bar{\nu})^2 / \overline{\nu(\nu - 1)}. \quad (\text{B-5})$$

In the more general case of a distributed fissile source, one has the following expressions:

$$N_1 \cong \bar{\nu} N_f \int_{\vec{r}} \int_E S(\vec{r}) f(E) W_1(\vec{r}, E) dE d\vec{r} \quad (\text{B-6})$$

$$N_2 \cong \bar{\nu} N_f \int_{\vec{r}} \int_E S(\vec{r}) f(E) W_2(\vec{r}, E) dE d\vec{r} \quad (\text{B-7})$$

$$N_{12} \cong \overline{\nu(\nu - 1)} N_f \int_{\vec{r}} S(\vec{r}) \left[\int_E f(E) W_1(\vec{r}, E) dE \cdot \int_E f(E) W_2(\vec{r}, E) dE \right] d\vec{r} \quad (\text{B-8})$$

$$\begin{aligned} \frac{N_1 N_2}{N_{12}} &= \frac{(\bar{\nu})^2}{\overline{\nu(\nu - 1)}} \times \\ &\times N_f \frac{\int_{\vec{r}} \int_E S(\vec{r}) f(E) W_1(\vec{r}, E) dE d\vec{r} \cdot \int_{\vec{r}} \int_E S(\vec{r}) f(E) W_2(\vec{r}, E) dE d\vec{r}}{\int_{\vec{r}} S(\vec{r}) \left[\int_E f(E) W_1(\vec{r}, E) dE \cdot \int_E f(E) W_2(\vec{r}, E) dE \right] d\vec{r}} \end{aligned} \quad (\text{B-9})$$

where $W_i(\vec{r}, E)$ is the probability that a photon born in position $\vec{r}$ and at energy E will be detected by the i -th detector. This probability is also affected by photon attenuation inside the sample, the solid angle, and the intrinsic detector efficiency. $S(\vec{r})$ is the normalized distribution of fissile material in the sample ($\int_{\vec{r}} S(\vec{r}) d\vec{r} = 1$).

It is clear from Eq. B-9 that, in general, the detection probabilities *will not* cancel out.

Nonetheless, it can be generally stated that the sensitivity of the ratio $N_1 N_2 / N_{12}$ to the mentioned probabilities is lower than that of the individual count rates, N_1 , N_2 , or N_{12} . This is because the expression of $N_1 N_2 / N_{12}$ is a ratio of two quantities of similar nature.

Partial cancellation of various components of the detection probabilities can occur in a limited but important number of instances. Two such cases are when the absorption is small or when the photon removal cross section becomes independent of energy. The photon attenuation factors evidently will not affect the ratio $N_1 N_2 / N_{12}$ when $\mu r \ll 1$. Here, the probabilities associated with space and energy are independent and separable, that is,

$$W_i(\vec{r}, E) = W_i(\vec{r}) \cdot \epsilon_i(E) \quad (\text{B-10})$$

where $\epsilon_i(E)$ is the intrinsic detector efficiency. When the latter is integrated over the fission spectrum, it drops out of the ratio, leaving the following ratio:

$$\frac{N_1 N_2}{N_{12}} = \frac{(\bar{\nu})^2}{\nu(\nu - 1)} \frac{\int_{\vec{r}} S(\vec{r}) W_1(\vec{r}) d\vec{r} \cdot \int_{\vec{r}} S(\vec{r}) W_2(\vec{r}) d\vec{r}}{\int_{\vec{r}} S(\vec{r}) W_1(\vec{r}) W_2(\vec{r}) d\vec{r}}. \quad (\text{B-11})$$

Since the fission prompt γ -ray spectrum spans a relatively wide range of energies, one can accentuate the higher energy region (the "Compton" region) by various means where (1) μ is a weak function of energy, and/or (2) μ is small enough so that $\mu r \ll 1$ for a range of sample sizes and materials. The cancellation of the intrinsic efficiency components in Eq. B-9 requires only condition (1).

Varying the degree of the dependence of the ratio $N_1 N_2 / N_{12}$ or other similar ratios on the attenuation effect offers a means of detecting large material density variations among samples of the same kind. One simple way is to compare ratios involving higher degrees of coincidences to ratios involving lower degrees of coincidences. A typical example is Eq. B-4. Another example is:

$$\frac{N_{12} N_{13} N_{23}}{N_{123}^2} = N_f \frac{[\nu(\nu - 1)]^3}{[\nu(\nu - 1)(\nu - 2)]^2} \frac{\langle G_{12} \rangle \langle G_{13} \rangle \langle G_{23} \rangle}{\langle G_{123} \rangle^2} \quad (\text{B-12})$$

where

$$\langle G_{i,j,\dots} \rangle = \int_{\vec{r}} S(\vec{r}) d\vec{r} \left[\int f(E) W_i(\vec{r}, E) dE \int f(E) W_j(\vec{r}, E) dE \int \dots \right]. \quad (\text{B-13})$$

A second way to control the variation of $\mu(E)$ is to insert successively thicker layers of lead-shielding between the sample and the detectors. A third way to vary and study the effect of prompt fission signature self-attenuation is to vary the detector bias, thus reducing the contribution to the count rates of γ -rays and neutrons of lower energies.

In summary, one can assume the possibility of reducing the dependence of the assay results on self-attenuation, provided the FMD system is inherently insensitive to spatial effects in the sample cavity, that is, if $W_i(r)$ and its products are approximately spatially independent.

Measurement with localized sources have demonstrated that the rotation and vertical scanning of the samples in the FMD and by other means (see Chapter 9) reduces variation in the detectors' response due to large spatial change to less than 10 percent.

APPENDIX C

SAMPLE SCANNING

In ANDA systems where the sample can be scanned along its axis, the response (nonuniform as it may be) in principle does not effect the results. We shall prove here this analytically, stating the various assumptions and their consequences.

Assume an ANDA system with an interrogation response distribution of $h(Z)$ where Z is the axis of motion and $h(Z)$ is the count rate of a unit fissile mass. For example, 1 g of ^{235}U at a position between Z and $Z+dZ$ yields a counting rate of $h(Z)dZ$ counts/s. The mass (in grams) is $g(Y)dY$ at a position Y of the ^{235}U in the sample.

Figure C.1. Schematic ANDA system with its interrogation response profile and a distribution of the fissile content in the sample.

In ANDA one usually deals with a standard sample and an unknown sample. The standard will have fissile content distribution of $g_s(Y)$, and the unknown will have a distribution of $g_u(Y)$. The corresponding total masses are $G_s = \int_{-\infty}^{\infty} g_s(Y)dY$ and G_u gram, respectively. Now, suppose that the sample is moved at a constant speed, v , from a position at the bottom (Z_b) through the ANDA system to a position at the top (Z_t).

(Z_o is the position at center.) At a particular time, t , the count rates will be given by the following:

$$C_s(t) = \int_{Z_b}^{Z_t} g_s(Z - Z_o - vt)h(Z)dZ + b \quad (C-1)$$

$$C_u(t) = \int_{Z_b}^{Z_t} g_u(Z - Z_o - vt)h(Z)dZ + b \quad (C-2)$$

where b is the known background count. Assume now that Z_b (the position at time $t = 0$) and Z_t (the position at time $t = T$, the total time taken to move from Z_b to Z_t) are far enough apart that the system response, $h(Z)$ is zero: namely,

$$C_s(0) = C_u(0) = C_s(T) = C_u(T) = b. \quad (C-3)$$

The total number of counts accumulated during the scan samples (N_s and N_u) are, respectively, in the standard and the unknown sample,

$$N_s = \int_0^T C_s(t)dt \quad N_u = \int_0^T C_u(t)dt \quad (C-4)$$

where

$$\begin{aligned} N_s &= \int_0^T dt \int_{Z_b}^{Z_t} g_s(Z - Z_b - vt)h(Z)dZ + bt \\ &= \int_{Z_b}^{Z_t} h(z)dZ \int_0^T g_s(Z - Z_b - vt)dt + bt. \end{aligned} \quad (C-5)$$

N_u may be expressed similarly.

The net count N is given by

$$n_s = N_s - bT \quad \text{and} \quad n_u = N_u - bT. \quad (C-6)$$

The change in the order of the interrogation in Eq. C-5 allows us to substitute for the second integral with G_s/v . Hence,

$$n_s = (G_s/v) \int_{Z_b}^{Z_t} h(Z)dZ \quad (C-7)$$

and

$$n_u = (G_u/v) \int_{Z_b}^{Z_t} h(Z) dZ. \quad (C-8)$$

Note that N_s and N_u are measured quantities (for example, total numbers of net coincidence signal) and that G_s is known. Hence, the fissile mass of the unknown sample is as follows:

$$G_u = G_s \frac{N_u}{N_s}. \quad (C-9)$$

The fissile content of the unknown sample is given in terms of the known fissile content of the standard sample, and the total net signal count from the unknown and standard samples. The result is entirely independent of either the fissile material distribution or the profile of the system response.

To recapitulate the assumptions that led us to Eq. C-9, we note that:

1. The net detected signal rate per unit fissile mass, $h(Z)dZ$, being a property of the ANDA system and, hence, should be the same for the standard and unknown samples. This requires that the self-shielding and multiplication (if any) be about the same in both samples.
2. The initial and final scanning points have zero response.