LITERATURE REVIEW AND COMMERCIAL SOURCE
EVALUATION OF AMERICIUM-241

FINAL REPORT

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ABSTRACT

An exhaustive literature review survey of the fabrication and applications of Am-241 is presented. Commercial Am-241 photon and Am/Be neutron sources were obtained and a limited evaluation program was carried out in conjunction with the study. A survey of Am-241 fabricators and facilities employing the isotope in instrumentation, was performed and is reported.

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I. AMERICIUM-241 THE MULTI-PURPOSE RADIOISOTOPE

A. Historical View*

Americium-241 or element 95 was discovered at the Metallurgical Laboratory of the University of Chicago by the team of G. Seaborg, A. Chiorso, R.A. James and L.O. Morgan during a period of several months (Between 1944-1945). After several attempts at making elements of atomic number greater than 94, plutonium-239 was bombarded with 32-MeV helium ions in the Berkely 60 - in Cyclotron. In the experiment it was speculated that the element $^{241}_{95}\text{Am}$ was produced.

Later it was shown that Americium-241 could be produced by the following neutron reactions:

The isotope was also produced by the reaction:

The element produced by beta decay of $^{241}_{94}\text{Pu}$ was characterized by alphas with a range in mica as depicted in Fig. 1. By the latter part of 1945, a sufficient quantity of Americium-241 had been accumulated such that B.B. Cunningham of the Metallurgical Laboratory was able to prepare the first observ-

able quantity of a pure compound of the isotope (the hydroxide).

The neutron-plutonium-239 reaction that was initially used to produce ^{241}Am is employed today to produce it in quantity. The AEC's Rocky Flats Plant near Denver, Colorado has, to date, produced more than 10 kg⁽¹⁾ of Americium-241 by "milking" the available plutonium stock. Figure 2 shows the past and the predicted future production of the actinide element $^{241}_{95}\text{Am}$.

The number of applications of $^{241}_{95}\text{Am}$ are far more numerous than any of the other actinide elements. Most of the applications of the source as a photon and alpha emitter are given in Table I. All of the applications and the characteristics of the source for the different kinds of particle emissions are discussed in the subsequent review.

Up until very recently, the half-life of $^{241}_{95}\text{Am}$ was taken to be 458 years as measured by low-geometry absolute alpha counting. However, Oetting and Gunn⁽²⁾ (1967), using six- and fifteen-gram samples of ^{241}Am metal, determined the half-life to be 432.7 ± 0.7 years by the calorimetric method. Nevertheless, throughout this report the previous value of 458 years will be used, since the difference in half-life is of little consequence for most applications.

* The information in this historical view is taken mostly from Reference 1.

ORIGINAL MICA ABSORPTION DATA
SHOWING PRESENCE OF 95^{241} ALPHA
PARTICLES IN DEUTERON - IRRADIATED PLUTONIUM

OCTOBER 17-18, 1944

G.T. SEABORG, L.O. MORGAN, R.A. JAMES, AND A. GHIORSO

49 DD #31 - RANGE CURVE FOR 95^{241}

49 DD #32 - COMPARISON RANGE CURVE
FOR Pu^{239} FRACTION

Figure 1. Original laboratory plot of October 1944 data characterizing new alpha activity. (Photographic reproduction, Reference 1.)

Table I. Applications of Americium-241

²⁴¹ Am	Alpha Source	Fire detector via ionization of surrounding atmosphere
		Charged particle activation-detection of air borne beryllium
		Alpha induced fluorescence, for detection of light elements
		Static charge elimination, paper and other industries
		Density measurements, extra terrestrial atmospheres
		Altitude measurements, via density determination
		Gas pressure measurement via alpha range determination
		Heat generation, current generation
		Ionization of air, in lightning rod application
		Changing charge imbalance after air conditioning via ionization
		Determination of thickness in thin metal foils, polymers
		Hole indicator device
		Humidity measurement
		Determination of gas components in mixtures
		Ion source in gas chromatography

²⁴¹Am * Neutron Source
 *Am/Be is employed
 unless otherwise noted.

Am/Be, soil moisture determination

Am/Be in conjunction with a gamma source, e.g.
 Cs for simultaneous determination of soil
 moisture and density

Road logging

Well logging
 Neutron-neutron logging (thermal and epithermal)
 Neutron-γ logging
 Activation logging, γ spectroscopy

²⁴¹Am Neutron
 Activation Techniques

C in Fe ores

Chromium in chrome concentrated sample

For aluminum in a variety of processes

Phosphorous in human bones

²⁴¹Am Neutron
 Source process Control
 Measurements

Moisture content in sintered material mixtures
 in blast furnace operations

Moisture content in glass manufacture, and con-
 crete mixes

Testing of tar impregnation of magnesite bricks

Radiography

Radiography

²⁴¹Am Photon Source
 (Attenuation techniques)

Water content measurement during transient flow
 in impact sand

Water content measurement during infiltration
 and redistribution in soils

Determination of water content and bulk density
 of soil in conjunction with Cs-137

	Bone density measurement
	Bone mineral content determination
	"In Vitro" measurement of fat/lean ratios
	Thickness gauging of glass and primary metals
	Thickness measurement of aluminum materials
Photon Source Backscatter techniques	Analysis of concrete
	Analysis of coal ash
	Analysis of road surfaces
Photon Source Fluorescence Methods	Fabrication of gamma excited x-ray sources
Spectroscopy	Determination of the concentrations of zinc in zinc ores. Determination of the concentration of Ag in Ag ores using an ^{241}Am photon source with Sb and I target assemblies
	Tin in Tin ores determination with ^{241}Am , Cs Target Assemblies
	Heavy element ore analysis for determination of La, Ce, Pr and Nd.
	On stream determination of zinc in zinc slurries using Am-241-Cs γ -X source
	On stream determination of tin in tin ore processing Am-241-Ba γ -X and Am-241 - Cs γ -X
	Determination of Ba in Baryte, Am-241/Sm and /Ba

	Alloy analysis--using Am-241 with various targets. (e.g. tin, copper, and molybdenum)
	Assay of Pure Gold
	Coating thickness measurements <ul style="list-style-type: none"> a) tin on steel b) zinc on steel c) nickel on steel d) aluminium on steel e) feasibility of corrosion measurements f) wear measurements of rubber tile g) paper weight density determination
	Clinical In ViVo imaging of the thyroid gland and the liver
	Monitoring of cerebral blood flow
	Quantity determination of Iodine in various organs of the body
Trace element analysis	Barium in ocean clams. Calcium and strontium in egg shells. Arsenic content in soil. Trace mineral content salmon
	Trace determination of lead in air sampling filters. Cd in water samples. Barium in Barium solution. Yttrium concentrations.
	Concentrations of Uranium in Uranium solutions using Am-241/I targets
	Iodine determination in pharmaceuticals

Figure 2. Potential Isotopes from U.S. Nuclear Plants. (Reference 3).

B. Extraction Processes for Americium - 241

The estimated growth nuclear power and spent fuel processing in the U.S.A. for the period of 1968 thru 1984 is given in figure 3.⁽⁴⁾ americium can be separated from lanthanide elements which do not involve high concentration of any salt or acid by the Talspeak method of Weaver and Kappelmann.⁽⁵⁾ The Talspeak method promotes highly preferential extraction of the lanthanides with hydrocarbon solutions of monacidic organophosphates of phosphonates by preferential complexing of the trivalent actinides in an aqueous solution with a suitable aminopolyacetic acid. Fig. 4 shows a flow sheet for this method of separation for americium and curium from the lanthanides and yttrium.

Moore⁽⁶⁾ reported a method for isolation of trivalent Actinide - Lanthanide elements from nitrate solutions. This method is based on the liquid-legend extraction method for extracting the HEDTA complex of ^{241}Am and $^{152-4}\text{Eu}$ Tracers with 20% Aliquat. A 336-5-Xylene solution method has also been described by Moore⁽⁷⁾.

Guseva and Tikhomirona⁽⁸⁾ demonstrated the possibility of separating the transplutonium elements from the rare earth elements of the yttrium group, using mixtures containing 1-2 M NH_4SCN + 70% CH_3OH or 1-2 M NH_4SCN + 50 - 70% $\text{C}_2\text{H}_5\text{OH}$; $\text{C}_3\text{H}_7\text{OH}$ in place of concentrated ammonium thiocyanate solutions, which is very important in work with large concentrations of α - emitters. Baybarz⁽⁹⁾ describes a phosphonate extraction process for separation of transplutonium elements from americium and curium which consisted of a preferential solvent extraction of the transcurium elements with 2 - ethylhexylphenylphosphoric acid from dilute HCL solutions. He found that extractions varied with the inverse third power of the acid composition in the

Fig. 3. Estimated growth of nuclear power and spent fuel processing in the United States for the 15 years.

(After Ref. - 4)

Fig. 4. Flowsheet for Purification of Americium and Curium.

(Ref. - 5)

range 1-4 N and directly with the third power of the extractant concentration. Most of the methods described above (like several others (10) (11)) have not been applied on a large scale.

At the Rocky Flats Processing Plant the americium is recovered from the filtrate of a plutonium precipitation operation. Initially the volume and pH is reduced by evaporation, the acidity of the evaporated "crude" feed is adjusted by dilution to $\sim 0.4N$, and finally the feed is loaded onto a cation-exchange resin column. 5 - M NH_4CNS is used to elute the americium and rare earths, and the eluate is subsequently loaded onto an anion-exchange resin column. The rare earths are eluted by washing with additional 5-M NH_4CNS and the Americium is stripped with 0.1- M HCL . The americium is loaded onto a cation-exchange column and stripped a second time with .6 - M HCL . This solution with the addition of small quantity HNO_3 for stabilization of residual $Pu(IV)$, is loaded in an anion-exchange resin column to remove plutonium. The americium is recovered by adjusting the H^+ concentration of the feed to $2N$ and then precipitating the Americium as the oxalate. Several other methods are discussed by Baybarz,⁽¹²⁾

Richardson⁽¹³⁾, discussed the pilot plant runs of a counter current solvent extraction⁽¹⁴⁾ technique. The process employs a solvent containing 30 vol. % dibutylbutyl phosph $\odot$ ate (DBBP) in CCL_4 diluent. $Pu(IV)$ is nearly quantitative in this system over a wide range of conditions. The extraction of Americium is strongly dependent on the total nitrate salting strength and nitric acid concentration of the aqueous feed. A solvent extraction flowsheet for separation of Americium from - rare earths (e.g. La and Ce) is given in Figure 5 (15).

Fig. 5 Solvent Extraction Flowsheet for Separation of Americium from Rare Earths (La and Ce). (From Reference 15)

REFERENCES

1. G.T. Seaborg, "The Synthetic Actinides--From Discovery to Manufacture," Nucl. Appl. & Tech. 9, PP 830-50, (1970).
2. F.L. Oetting and S.R. Gunn, J. Inorg, Nucl. Chem., 29, P. 2659, (1967).
3. Nuclear Industry, "Optimum is Balanced by Caution in Isotope Market Appraisals", Nov.-Dec., 15(11 and 12), 1968.
4. C.A. Rohrmann, "Values in Spent Fuel From Power Reactors," Isotopes and Radiation Technology, 6(1), (1968).
5. B. Weaver and F.A. Kappelmann, "Talspeak: A New Method of Separating Americium and Curium from the Lanthanides by Extration from an aqueous solution of an Amenopolyacetic Acid Complex with a Monoacidic Organo-phosphate or Phosphate," ORNL-3559, (1964).
6. F.L. Moore, "Improved Extraction Method for Isolation of trivalent Actinide-Lanthanide Elements from Nitrate Solutions," Analytical Chemistry, P. 510, (1965).
7. F.L. Moore, "Liquid-Liquid Extraction of Anionic Americium and European Complexes of Hydroxyethylethylenediamine-Triacetic Acid and DTPA," Anal. Chem., 38(7), P 905, (1966).
8. L.I. Gusena and G.S. Tikhomirava, Radiokhimiya, 19(2), PP 246-49, (March-April, 1968).
9. R.D. Baybarz, "Separation of Transplutonium Elements by Phosphate Extraction," ORNL-3273 (1962).
10. A.K. Williams, USAEC. Rep. RFP-349, (1964).
11. P.G. Hagan and G.W. Miller, USAEC. RFP-442, (1965).
12. R.D. Baybarz, Atomic Energy Review, 8(2), IAEA, Vienna, (1970).
13. G.L. Richardson, "Americium Recovery by DBBP Solvent Extraction Pilot Plant Demonstration" BNWL-CC-1503. (1968).
14. I.N. Taylor, Jr., "Flowsheet for Continous Solvent Extraction of Plutonium and Americium in Waste Treatment Facility," ARH-210, (1967).
15. E. H. Smith and W. Bowes. "Isotopic Power Sources .. A Compendium," MND -P -2581-1, (1961).

II. AMERICIUM-241 AS AN ALPHA SOURCE

Americium-241 is an artificially produced radioelement with a half-life of 458 ± 10 years that decays to ^{237}Np by alpha emission followed by lower energy gamma emissions (the gamma spectrum of an encapsulated ^{241}Am photon source is covered in section IV of this review). Americium-241 is present as a by-product in the waste streams of plutonium reprocessing plants and is also recovered from power reactor fuel reprocessing with mixtures of ^{241}Am and ^{243}Am .

The nuclear and chemical properties of Americium-241 are covered extensively in an early review (1962) by Strain and Leddicotte⁽¹⁾ and in a later review (1972) which was with alpha sources in general, by Greene Pressly and Case⁽²⁾. For additional information on the nuclear properties, one might consult The Atomic Nucleus, by R. D. Evans. Alpha emitting radionuclides can be used to produce intense sources of alpha particles. These alphas can be applied in a variety of tasks ranging from the production of x-rays (by bombarding low Z elements with alpha particles) to determining chemical composition. There are many other unique applications of ^{241}Am as an alpha source and these will be covered later. However, the most common use of ^{241}Am as an alpha emitter is in neutron sources through the (α, n) reaction on beryllium. Most of the americium produced is used in the manufacture of neutron sources.

The half-life of ^{241}Am is long enough to preclude frequent decay corrections. The specific activity is ~ 3.43 curies per gram or 3.96×10^9 ⁽²⁾ alpha disintegrations per milligram of ^{241}Am .

(3)
The principal alpha emission energies are 5.44 and 5.48 MeV at percentages
of 13% and 85% respectively. The decay scheme of ⁽⁴⁾241Am is shown in Fig.1.
A recent measurement of the spectra of ²⁴¹Am along with that of ²⁴⁴Cm and
²³⁴Pu obtained using a GaAs ⁽⁵⁾ surface barrier is shown in Fig.2 . The
most important parameters that influence one's choice of an alpha-emitter
are half-life and activity and in both cases ²⁴¹Am has acceptable
parameters for most applications. Table I , gives some of the properties
of several of the commonly used alpha sources along with ²⁴¹Am.

Figure I . Decay Scheme of Am^{241} (Ref. 4.)

Figure 2 . A typical α -spectra of ^{239}Pu , ^{241}Am and ^{244}Cm taken with GaAs detector (EX-62, #3) at room temperature. (Ref-5)

NUCLIDE	HALF-LIFE	PRINCIPAL ALPHA EMISSION ENERGIES (MeV)	CURIES PER GRAM OF NUCLIDE	ALPHAS** (Cpm/mg)
$^{210}\text{Po}^*$	138 d	5.30 (100%)	4500	5.19×10^{12}
^{228}Th	191 y			
$^{238}\text{U}^*$	4.47×10^9 y	4.20 (77%) 4.15 (23%)	3.36×10^{-7}	388
$^{238}\text{Pu}^*$	87.5 y	5.50 (72%) 5.46 (28%)	17.2	1.99×10^{10}
$^{239}\text{Pu}^*$	2.44×10^4 y	5.15 (88%) 5.11 (11%)	6.13×10^{-2}	7.08×10^7
$^{241}\text{Am}^*$	458 y	5.48 (85%) 5.44 (13%)	3.43	3.96×10^9
$^{244}\text{Cm}^*$	18 y	5.81 (76%) 5.77 (24%)	81	9.38×10^{10}
^{210}Pb	22 y	5.3		
^{226}Ra	1620 y	4.5 7.7		

* Values taken from Reference 2 .

** 52% geometry.

TABLE I. Properties of several commonly used alpha sources.

A. Preparation of ^{241}Am Alpha Sources

In the preparation of alpha sources the two most important qualities are adherence and uniformity of thickness. The first quality is important because good adherence prevents the loss of active material. Uniformity of thickness is important because it prevents changes in the alpha spectrum caused by variation in absorption.

There are at least six techniques for preparing alpha sources. These range from the more common electro deposition method to the less common enamel power melting technique.⁽⁶⁾ The various techniques along with descriptions of the methods are given in Table II. Detailed descriptions of the techniques of source preparation presented in Table II can be found in the review of alpha sources by Green et al.⁽²⁾ Reference 2 also presents several other methods of source preparation not mentioned in Table II.

The encapsulation^s of alpha sources must be done in such a manner as not to severely affect the spectrum of the emitted alphas but also to insure that the active material will be, at all times, contained. Organic film cover materials, such as Mylar, Zapon and Polyethylene, while being relatively transparent for α 's suffer radiation damage in short period of time. In a test conducted by using⁽¹⁾ .20 grams of AmO_2 covered with 0.1 mil polyethylene film, large holes developed in the polyethylene after less than eight hours. The results of a test ran by Strain and Leddicotte⁽¹⁾ in which a 0.1 mil thick nickel foil was used as a cover is shown in Figure 5. The transmitted integral alpha count through the nickel foil was 72% of that of the uncovered ^{241}Am source with a noticeable shift in energy of alphas emitted. Protective coverings in some cases reduce energy of the alphas by as much as 50%. This is a problem

Table II. Methods of Preparing Alpha Sources

METHOD	DESCRIPTION OF METHOD	COMMENTS
Electrodeposition	The alpha emitter is electrolytically deposited from an electrolyte, ammonium oxalate solution onto a metallic substrate (the cathode)	Good adherence and quantitative recovery for $< 100 \mu\text{g}/\text{cm}^2$ of ^{241}Am . (See Fig. 3).
Electrospraying	High voltage placed on a wire inserted in a capillary, from which the radioactive material in solution is forced in a tiny droplets onto a grounded backing plate.	Used in preparing thin layer sources ($\text{few } \mu\text{g}/\text{cm}^2$) on areas up to 100 cm^2 . Uniform deposit and high yields. (See Fig. 4).
Sputtering	In this technique an argon plasma is impinged on a target of fused oxide or metal causing a spallation of the target material onto the substrate.	Adoptable to a wide range of materials. Good uniformity and good control over source layer thickness. possibility of significantly stronger α -sources up to $2 \text{ mCi}/\text{cm}^2$.
Vacuum Evaporation	Electron bombardment or RF induction heating is used to evaporate the source material. This creates vapor beam which is directed toward the substrate for deposition.	Good adherence and uniformity of the condensed layer. Thickness of $< 1 \mu\text{g}/\text{cm}^2$ to $> 50,000 \mu\text{g}/\text{cm}^2$ efficiency $>$ that of the sputtering technique. 20% yields.

Continuation of Table II . Methods of Preparing Alpha Sources

METHOD	DESCRIPTION OF METHOD	COMMENTS
Solution Evaporation	Evaporation of an aqueous or organic solution of the source material onto a substrate.	Fair adherence and 100% yields are obtained using this method.
Painting	A dilute solution of cellulose nitrate and active material is literally painted onto the backing material and after drying the residue is fired.	Thick films can be obtained by this method. But the uniformity is a function of the operation.

* Private Communication-- J.H. Jarrett (1973).

only in those cases where the source is being used as a reference source or in alpha spectrometry. Commercial alpha sources employ gold, stainless steel and other metals as covering materials.

Figure 3 . Electroplating cell. Preparation of thin uniform Targets of actinide elements: uranium and americium. (Reference 7).

Figure 4 . Basic diagram of electrospraying machine. (Reference 8).

Figure 5 . Firsh Grid Chamber Measurement of Emitted Alpha Particle Energy from a Covered and Uncovered Mount. (Reference 1).

B. Methods of Evaluation of ^{241}Am -Alpha Sources

A test of adherence of alpha emitters is of prime importance since loss of adherence can cause contamination of the environment of the source or result in increased self-absorption in the source. ⁽⁹⁾ The integrity of an alpha source, as far as release of α 's is concerned, can be ascertained by a simple smear test. A smear test on a cover or uncovered source should not show a release of activity greater than 0.005 μCi (see Reference 2) of the established limit.

The emission rates of an alpha source can be accurately determined using surface barrier detectors, magnetic spectrometers and ionization chambers. The uncovered spectra of ^{241}Am , ^{243}Am and ^{244}Cm obtained using a surface barrier detector are shown in Figure 6. In addition to surface barrier detectors, magnetic spectrometers and ionization chambers, secondary reactions can be used to measure the intensity of alpha sources. One such reaction is the (α, n) reaction in suitable low Z materials. ⁽¹⁰⁾

These neutrons can be used to produce activation and the resulting activity of the radioisotope related to alpha emission.

For sources with large amounts of radioactivity, the output of the alpha source can be determined by measuring the current produced in a collector plate by alpha particles. The source and collector are housed in a vacuum called an Evacuated Alpha Chamber. ⁽¹¹⁾ Similarly, when the source is not placed in an evacuated chamber, the current produced by the ionization of the air surrounding the source causes the current to flow between the source and collector. ⁽¹²⁾ This current, when measured at various source-to-collector distances, can be used to estimate by extrapolating the alpha flux of the source surface.

Figure 6. Illustration of high resolution obtained in alpha-spectrometry application. Taken with surface-barrier detector of 25 mm² area, operated at room temperature. (Reference 16).

The energy distribution of alpha particles emitted by an alpha source is affected by the uniformity of the thickness of the active material.

The effect of thickness has been studied by several experimentalist.⁽¹⁴⁾ ⁽¹⁵⁾ ⁽¹⁶⁾ Utilizing the information obtained in these studies if the energy spectrums from other complex alpha sources can be predicted. Knowledge of the composition of the spectrum of an alpha source can be used to estimate its thickness.

The uniformity of the deposition of active material can be determined by autoradiography,⁽¹⁷⁾ densitometric⁽¹⁸⁾ techniques and counting at different small locations of the source through masking metal or other material templates.

Table III lists methods of source evaluations that have been used by various investigators in the study of ^{241}Am or alpha sources in general. A more comprehensive listing of methods used in source evaluation and the parameters measured can be found in Reference 2 .

TABLE II ^{241}Am and Alpha Sources Evaluations

RADIOISOTOPE	PARAMETER MEASURED	METHOD USED	REFERENCE
^{241}Am	Alpha intensity	Alpha-gamma coincidence	19-31
Alpha emitters	Alpha intensity	(α, n) and (n, γ) reactions	20
Alpha emitters	Alpha backscatter coefficient	Intensity measurement	21
^{241}Am , ^{238}U ,	Uniformity	Autoradiography	22
^{241}Am	Absorption of alpha in stainless steel	Energy spectrum	23
Alpha emitters	Dose depth	Calculations and TLD	24
Alpha emitters	Self-absorption	Energy spectrum	25
Alpha sources	Dose rate	Theoretical and semi-empirical relations	26
Alpha emitters	Alpha dose rate uniformity	Actinometric photographic plates	27
^{234}U , ^{241}Am	Effect of absorbers on alpha energy	Surface barrier detector	28
Alpha emitters	Energy spectra	Comparison to standard	29
Alpha emitters	Effect of thickness Effect of thickness	Energy spectra Angular intensity	30

RADIOISOTOPE	PARAMETER MEASURED	METHOD USED	REFERENCE
^{241}Am	Alpha angular intensity Alpha energy	Surface barrier detector	31

C.Applications of ^{241}Am as Source of Charged Particles

The application of ^{241}Am as an alpha source in radioisotopic neutron sources has been covered in the section III of this report

Herein, we shall examine only those uses where the source is designed primarily as an alpha source.

The maximum range and high specific ionization of alpha particles limits their use to two broad areas. These are: gaging applications, ionization of gases and radioisotope generators.

1. Ionization of gases

Due to its large mass the principal process that causes energy loss is ionization by electron collisions. The small amount of energy lost due to angular deviation, nuclear-scattering and reactions is not significant.

The high specific ionization of alpha particles has made their use almost a must in the detection of fire, ionic conditioning of air, elimination of static electricity and in lightening rods. The interaction of alpha particles with matter is discussed in several texts.⁽³²⁾ ⁽³³⁾

a. Fire detection

The principle of ionization via alpha particles has been used in fire detection apparatuses. Dance⁽³⁴⁾ describes a smoke detector which employs alpha sources of strengths of the order of 10 - 100 mCi (usually ^{226}Ra and ^{241}Am). Two ionization chambers are employed in the fire detection device. One of the chambers is closed and the other one is open to the environment. The circuit of the smoke detector is shown in Figure 7, along with the characteristic ionization

Figure 7-A. Electrical circuit of a smoke detector. (Ref.34).

Figure 7-B. Characteristic curves for the ion chamber (Ref.34).

current versus the voltage across each chamber. Under no smoke conditions, the voltage V_t is less than enough to trigger the trigger tube but when smoke in sufficient quantities enter the open chamber the cathode voltage increases and activates the alarm. The ionization current of the open chamber can be increased by as much as 10% when the ~~comb~~ustion products in the air reaches 1 part in 10,000 by weight.

Several ^{(35) (36) (37)(38)} fire warning systems based on the ionization principle have been reported in the literature and in most of these systems ^{241}Am can be used as the alpha source.

2. Charge Particle Activation

Use is made of the α , n reaction in neutron sources, but seldom is it used as a means of analysis. Strain and Leddicottee⁽¹⁾ used 12 mg of ^{241}Am in solution with varying amounts of beryllium to demonstrate that beryllium in small concentration (μg) could be detected by monitoring the prompt gamma radioactivity produced in the following reaction

They suggest that ^{241}Am be used instead of ^{210}Po as the alpha source in monitors designed (See figure 8) to determine the presence of air borne beryllium. The same mechanism of activation can be applied for quantitative control in products where Boron, Fluorine and Aluminium are present in analytical chemistry.⁽³⁹⁾

AIR FLOW IS AROUND PHOTO TUBE AND ALPHA SOURCE. THIS ARRANGEMENT MINIMIZES α CONTAMINATION.

Figure 8 . Source Mounting Detail for Air Borne Be Monitor. (Ref. 1)

Alpha-x-ray fluorescence and luminescence are also forms of activation analysis. Alpha - x - ray fluorescence using α 's from ^{241}Am will be dealt with in detail in a later section of this report.

3. Static Charge Elimination

In most industries where mechanical materials with poor electrical conductivity are processed, electrostatic charges are built up. This problem is particularly prevalent in the printing and textiles industries. But it is also present in the plastics, rubber and various machinery in mechanical plants,

Static charges have many adverse effects on the productivity of workers and on the quality of production. For example in the printing industry, the speed that paper fed into printing presses is limited by the presence of static charges.

Radiation (nuclear) has gained widespread use as ionizers employed in the elimination of static charges. Principally, alpha^{(40) (41) (42) (43) (44) (45)} and beta particles are used as the ionizing particles. The beta particles have a much larger radius of action but they are less effective.

Legrand⁽⁴⁶⁾ developed an ionatron device which used ^{241}Am as its source of alphas. The range of ionization of this device is a maximum of 75 mm.

This small radius of action made its practical use somewhat limited.

Malcoci and Gheorghin⁽⁴⁷⁾ examined the various static charge elimination devices and found that the most commonly used radioactive sources are:

^{241}Am (alphas) and ^{90}Sr (betas).

and to a lesser degree ^{210}Po (alpha) and ^{226}Ra (alpha). A block ionization unit is shown in Figure 9. This unit consists of identical elements, each with disc electrodes on which the radioactive source is mounted and a grounded grid. The electrode is connected to a high voltage supply.

4. Gas Pressure and Density Gauges

Gas density gages can be designed to operate on anyone of the following principles:

- (a) Energy - which uses as an indicator the residual energy of the alphas after they have passed through the gas to measure its density.
- (b) Range - which uses as indicator the number of alphas passing through a thickness of sample to imply the gas density.

In addition to density, the energy and range principles can be applied to thickness, temperature and pressure measurements. Hansen and Sellers⁽⁴⁸⁾ have applied the above principles in a feasibility study of alpha-particles for measuring planetary atmospheric density. The system uses either a "thin" or "thick" source design. The "thin" source system uses a monoenergetic alpha source (e.g. ^{241}Am), separated from the detector and measures the average energy loss. The thick source uses an alpha source with a broad energy spectrum and measures the count rate above a certain threshold. Either system can be employed in combination with CO_2 , N_2 and Ar atmospheres and the measured density, ρ_m , given by

$$\rho_m = \rho_{\text{CO}_2} + \rho_{\text{N}_2} + 0.67 \rho_{\text{Ar}} \quad (2)$$

Figure 9 . Block ionization unit with radioactive sources:
 1 - active electrode; 2 - collecting electrode;
 3 - source of supply; 4 - metallic housing earthed;
 5 - partition insulators. (Ref. 47)

to within $\pm 2\%$. The thin source system is more accurate at high densities, while the thick source gives better results at low densities. The system used with an x-ray attenuation system allows the quantities $\rho_{\text{CO}_2} + \rho_{\text{N}_2}$ and ρ_{Ar} to be determined separately. Even without composition information, the particle system alone will measure the density of the Martian atmosphere to within $\pm 8\%$.

Sellers and Morel⁽⁴⁹⁾ described an altimeter setting indicator system capable of operating up to ten digital displays with an indicated error of $\pm .005$ inches of mercury. ^{241}Am was selected as the isotopic source. The sensor is placed in a temperature controlled environment (i.e. enclosure) and the measured density inside the chamber is used to infer the pressure (See Fig. 10).

An alpha-particle densitometer with variable response has been described by Brandt et al⁽⁵⁰⁾. The instrument makes use of ^{241}Am as the source of alphas for the gas density measurements. They derived relations to establish the connection between a specified densitometer response and the corresponding source distribution. Shaped sources were constructed by mounting contoured baffles on the alpha sources. Figure 11, shows responses for a parabolic source.

In general, gas density measuring devices can use ^{241}Am as a source of alphas. Some of the devices and theory of alpha emitters for measuring gas densities are discussed in references (51) (52) (53)(54) (55)

In the measurement of pressure, the amount of current produced in a gas by an ionizing particle moving through it is directly related to the pressure in the medium. The radiation source works in principle like the filament of a conventional ion gage. This type of gage has the advantages

Figure 10. Simplified diagram of temperature chamber. (Ref. - 49)

Figure 11. Experimental instrument response for an $E^{1/2}$ - source versus air pressure on a linear scale. Source 10 mCi ^{241}Am , source-counter distance $D=100$ cm, counting time per point ~ 3 min. (Ref. 50)

of (a) no burn out of the filament; (b) no possibility of chemical reaction between the gas and filament; (c) consistency in the alpha source and lack of instabilities resulting from power supply fluctuations. Its main disadvantage is that only mean-level or analog output is possible which makes it sensitive to fluctuations inherent in analog signals.

Andrieux and Iapteff⁽⁵⁶⁾ used the Alphovac-500 ionization gages to demonstrate the pressure range of ionization gages. The Alphovac-500 is capable of pressure measurements between $.5 \times 10^{-4}$ and 1000 torr with a precision of $\pm 3\%$ and it uses a ^{241}Am alpha source.

Gilly and Jourdan⁽⁵⁷⁾ used alpha sources to study the determination and stabilization of altitude. A 90% enriched ^{235}U source was used with a semiconductor detector for determining altitude and a ^{241}Am source was used with a semiconductor detector for stabilizing the altitude. The results were identical with those given by a barometer in a stratospheric balloon test.

Gilly, Jourdan and Denis⁽⁵⁸⁾ have also developed a machmeter using semiconductor detectors and ^{241}Am alpha sources. With two sources, one is able to measure static and impact pressure when each source is paired with a detector and the necessary electronic equipment.

5. RADIOISOTOPIC HEAT GENERATORS

The heat generated in a radioisotope results from exothermic decay reactions. In the exothermic decay reaction, the difference between the mass of the decay products and the initial mass of the isotope is called the mass defect and represents the mass that appears in the form of energy. The heat is usually converted into electrical energy in compact power devices by thermoelectric or thermionic means.

Generally two types of radioisotopes are used in radioisotopic fuels. These are either alpha or beta emitters. Beta emitters in most cases, have longer lifetimes, lower power densities (i.e. Watts) and lower cost than the alpha emitters. The heat generators that utilize alpha emitters require less shielding since they present less of a health hazard than X- and gamma rays.

The specific power of ^{241}Am (i.e. $0.1145 \pm .0002$ watts/gms (59) (60)) is low; that is, when compared with other alpha emitters of much shorter half lives. Some of the commonly used alpha emitters and their specific powers are given in Table IV.

TABLE IV
ALPHA EMITTING ISOTOPES USED IN POWER SOURCES (61)

ISOTOPE	DECAY	HALF LIFE (years)	THERMAL POWER (watt/gm.)
^{238}Pu	α	90	0.48
^{244}Cm	α	18	2.3
^{242}Cm	α, n	0.44	120
^{210}Po	α	0.38	140

Very few radioisotopic heat generators have been designed primarily with Am as the energy source. In most instances where the source is mentioned, it is simply referred to as one of many possibilities (62) (63) (64) (65) ^{241}Am does have some limited applications as in situations where a constant current source is required. (66) (67)

6. Radioactive Lightning Rods

The use of ionized air around lightning rods as a means of providing better protection against lightning has recently gained widespread acceptance. (68) The E. F. ("electricite froide") lightning rod (69) (70) consist of a stainless steel cylindrical rod having a sharp point at the top with several ridges running longitudinally down the rod, and an annular electrode having sharp

edges directed toward the rod. The rod and electrode on which several

Am sources are mounted are separated by an insulator. The radioactive lightning rod is based on the Szilard's Theory that ionized air would provide better protection than the Franklin Rod. The device is designed such that when the potential gradient between the rod and the electrode reaches high enough values there will be electrical discharges between the sharp edges of the electrode and the ridges of the rod. The discharge is aided by the ionization of the air by the alpha particles emitted by the ²⁴¹Am sources. The electrical discharge causes additional ionization of the gases, the net effect being to create an ionized cloud around the lightning rod that provides easy path for the lightning to the grounded rod.

The present design of the E. F. Lightning Rod uses eight specially developed ²⁴¹Am sources. A schematic diagram of the E. F. Lightning Rod is shown in Figure 12. In a test comparing the E.F. Lightning Rod with a Franklin Rod on top of a skyscraper in France, the E.F. Rod gave an ionization rate about 10^6 times higher than the Franklin rod. (71) Delhove (72) (73) reported recently that there are between five and ten thousand such rods installed in Europe. In spite of the popularity of the E.F. Lightning Rods in Europe, these devices have not gained widespread use in the U.S. A. (74)

7. Air Conditioning by Ions

The question of air ionization has become supremely acute in large cities where, because of pollution, buildings have to be aired by ventilation systems which remove all ionization from the air we breathe. Filtering of air for air conditioning removes negative ions from the air giving rise to an over abundance of positive ions. Positive ions,

Figure 12 Schematic Diagram of E.F. Lightning Rod. (Reference 69)

unlike negative ions, tend to have a detrimental effect on one's feeling of well-being.

By utilizing a radioactive source⁽⁷⁵⁾ it is possible to change the electric sign of the air giving it a preponderance of negative ions suitable for human beings. Invernizzi⁽⁷⁶⁾ has described the "Energie Froide" ion generator which used alpha sources. The system uses an ^{241}Am source capable of producing 5×10^9 ions/sec. The alpha particles generate both positive and negative particles in ionizing the air. The positive ions are removed, electrostatically collected and eliminated while the negative ions are emitted to the ambient air. If the air has been conditioned, the E.F. generators reestablishes the acceptable ionic condition. Under polluted conditions the E.F. generators cause the precipitation of the pollutants and tend to restore the proper negative ion balance in the ambient air.

8. Thickness Gauges

Recently, several reviews have been published that are devoted either entirely or in part to methods of thickness measurements. Howe⁽⁷⁷⁾ has surveyed the methods of film thickness determination that are applicable to nuclear targets in which alpha particles were covered. Moret⁽⁷⁸⁾ in a review of methods of thickness measurements also treated the alpha absorption method. More recently, Greene, Pressly and Chase⁽⁷⁹⁾ reviewed exclusively the use of alpha particles in thickness measurements.

This technique is best applied to the problem of determining the thickness of a material with a high degree of accuracy. Such a case is the determination of target foil thickness. The energy loss method is used in lieu of such methods as balance weighting, ellipsometry, change in frequency of a quartz crystal, resistivity measurements, and others since it alone is readily adaptable to thickness or uniformity measurement. The method involves measuring the change in energy of particles as they pass through the target foil. H. L. Adair⁽⁷⁹⁾ has presented a thorough discussion of the use of the method in the measurements of carbon, gold, and nickel foil thickness. (See Fig.'s 13 and 14). The source used was an 0.8 g ^{241}Am . Figs 13 and 14 show the alpha energy loss as a function of the thickness of gold and nickel targets, respectively. A patent on a transmission alpha thickness apparatus has been awarded to Anderson⁽⁸⁰⁾ for a device consisting of an alpha source (which may or may not be ^{241}Am) and a semiconductor junction diode detector which relate the thickness of the material to the shift in the energy spectrum of the alpha particles.

Teranishi⁽⁸¹⁾ then later Nonaka and Sugai⁽⁸²⁾ using an improved gage have described the measurement of the thickness of thin polymer films with a semiconductor alpha particle detector and a ^{241}Am alpha source. In the latter the accuracy of the method was improved by using a larger ^{241}Am source.

Burke and Tippet⁽⁸³⁾ have applied Rutherford scattering to target thickness measurements. By counting the rate of scattered particles, the thickness of foils were determined. The method is particularly suited for thickness measurements of high Z materials that have been deposited on low Z backings.

Figure 13. Americium-241 Energy Loss Measurements for 100-700 $\mu\text{g}/\text{cm}^2$ Gold Foils. (Ref. 79)

Figure 14. Americium-241 Energy Loss Measurements for 0.2-5.0 mg/cm^2 Nickel Foils. (Ref. 79)

9. Other Applications

The introduction of electron capture detection in gas chromatography has opened up new areas of investigation. These electron capture detectors makes use of a radioactive source that ionizes the gas and functions as one of the ionization chambers electrodes. Shoemaker et al (84) compared ^{241}Am , ^{99}Te , ^3He and ^{226}Ra for their utility in gas chromatographic ionization detectors. Their results showed that ^3He , ^{241}Am and ^{226}Ra work equally as well as radiation sources in electron capture ionization detectors. For temperatures above 225°C only ^{226}Ra and ^{241}Am were recommended with ^{241}Am being favored because of its relative low energy gammas.

Alpha particles are also used in the measurement of the relative humidity of air. (85) The relative humidity of the air is determined by measuring the transmission of alpha particles through a thin film containing an equilibrium proportion of moisture. The method reduces the interferences normally encountered with electrical hygrometers caused by pollutants in the air.

Gardner et al, (86) have suggested a new application of alpha particles. Observing that various gases exhibit stopping powers that differ from others in magnitude and in relative magnitude as a function of energy, they concluded that if the components of a gas mixture and their stopping power are known, a measure of the concentration each component can be obtained. Tests were conducted on different gas mixtures of N_2 and CO_2 using a ^{241}Am source and a surface barrier detector. The results of these preliminary tests are shown in Table IV.

Table V. Results for Alpha - Particle Stopping Power Gauge Method of
Measuring the Composition of N_2 - CO_2 Mixtures. (ref. 66)

Approximate CO_2 Concentration by Orifice (Mol Fraction)	Measured CO_2 Concentration by Alpha Particle Stopping Power Gauge (Mol Fraction)
0.0	0.0
0.2	.228
0.25	.292
0.30	.364
0.50	.569
0.05	.093
0.05	.091
0.60	.682
0.50	.593

A recent paper by Patterson et al, ⁽⁸⁷⁾ discussed the use of ^{241}Am in the preparation of high-level alpha-particle sources for the NASA surveyor alpha scattering experiment. These high intensity alpha-particle sources can be prepared by neutron irradiation of ^{241}Am in a reactor to produce ^{242}Cm . The alpha activity ratio ($^{242}\text{Cm}/^{241}\text{Am}$) was $10^7 : 1$.

REFERENCES

1. J.E. Strain and G.W. Leddicottee, "The Preparation; Properties, and Uses of Americium-241, Alpha-Gamma- and Neutron Sources", ORNL-3335 (1962).
2. R.E. Greene, R.S. Pressly, and F.N. Case, "A Review of Alpha Radiation Source Preparation Methods and Applications," ORNL-4819 (1972).
3. D. Stromenger, J.M. Hollander, and G.T. Seaborg, "Table of Isotopes," Rev. Modern Phys., 30, No. 2, Part II, P. 826 (1958).
4. Phys. Rev. Vol. 30, P. 286, (1958).
5. T. Kobayoshi et al. "Performance of GaAs Surface-Barrier Detectors Made From High-Purity Gallium Arsenide", IEEE Transactions on Nuc. Sci, NS-19, 3, (June 1972).
6. C. Cimetiere, H. Desroches, and C. Routier, "Process for the Fabrication of Alpha Sources, Patent 2,041,947 (French) Jan. (1971).
7. J.C. Laul, M.V. Ramonial and C.L. Rao, "Preparation of Thin Uniform Targets of Actenide Elements: Uranium and Americium," International J. of App. Rad. and Isotopes, 17, PP 306-308, (1966).
8. P.S. Robinson, "The Production of Radiolactive Sources by the Electro spraying Method," 40, PP 136-40, (1966).
9. Industrial Nucleonics Corp., Columbus, OH., "Radiation Source Capsule," Br. Pat. 1, 187,210 (Apr. 1970).
10. E.G. Segre, "Method and apparatus for Measuring Strong Alpha Emitters", American Patent 2,573,069, Oct. 30, 1951 assigned to USAEC.
11. ORNL, Evacuated Alpha Chamber, CAPE-1831.
12. R.F. Itzhaki, "An α -ray Source and Its Dosimetry," Physics in Med. Biol., 5, PP 61-72 (1960).
13. J.L. Blankenship and C.J. Borkoweki, IRE Trans. on Nuclear Sci., NS. 8 (No. 1): 19 (1961).
14. V.P. Grigoror, "Approximate Method of Calculating Alpha Radiation Spectra from Thick Emitters," Gosudorstvenngi Komitet Po Ispol.' Zovaniyu. Atomnoi Energii USSR (1963).
15. N.K. Abrosimov and G.E. Kocharov, "The Effects of the Thickness of The Source on the Shape of the Energy Distribution and Angular Distribution Curves of α -Particles," Izvest. Akad. Nawk. SSR, Ser. Fig. 26, PP 237-44 (1962).

16. I. Chudacek, "Energy Spectrum of Particles Emitted from Sources of Different Thickness," Czechoslov. J. Phys., 8, PP 396-403. (1958).
17. A.T. Kazakevich and V.M. Surin, "Application of Autoradiography for Control of Irregularity of Actinide Element Layers," At. Energ. (USSR) 21, 132-4 (1966) (In Russian).
18. G.A. Dupetit, "Preparation of Neptunium Targets by Electrodeposition", J. Inorg. Nucl. Chem., 24, PP 1297-8, (1962).
19. Annual Progress Report for the Period of July 1961-June 1962, NBL-188, New Brunswick Laboratory (December 1962).
20. E.G. Segre, "Method and Apparatus for Measuring Strong Alpha Emitters," American Patent 2,573,069, Oct. 30, 1951; assigned to U.S. Atomic Energy Commission.
21. M. Movchet, M. Boulloud, and M. Gasc, "Study of the Variation of the Efficiency of α Emitting Sources as a Function of the Nature of the Support and the Applied Threshold, CEA-2128, Commissariat a l'Energie Atomique Centre d'Etudes Nucleaires, Saclay (1962).
22. A.T. Kazakevich and V.M. Surin, "Application of Autoradiography for Control of Irregularity of Actinide Element Layers," At. Energ. (USSR) 21, 132-4 (1966) (In Russian).
23. R.A. Fluegge, "A Study of Alpha Particle Reactions in Atmospheric Gases, NYO-5708-2, Cornell Aeronautical Lab, Inc. (November 1966).
24. P.J. Walsh and D.I. McRee, "Depth-Dose Curves for α Particles from Area Sources and Point Sources," Health Phys. 20, 352-3 (1971).
25. V.P. Grigorov, "Matrix Method of Calculations of Alpha-Ray Spectrum of Thick Sources," At. Energ. (USSR) 21, 517-18 (1966) (In Russian).
26. Dosimetry, MIT-952-5 (Part I), PP 137-83, Massachusetts Institute of Technology, Cambridge.
27. R.F. Itzhaki, "An α -ray Source and Its Dosimetry," Physics in Med. Biol. 5, 61-72 (1960).
28. L.H. Peshori, P.P.V.J. Nambiar, and D.V. Gopinath, "Effect of Absorbers on Alpha Spectral Measurements," Health Phys. 18, 685-91 (1970).
29. V.P. Grigorov, Approximate Method of Calculating Alpha-Radiation Spectra from Thick Emitters," Gosudarstvennyi Komitet po Ispol'zovaniyu Atomnoi Energii SSSR, 1963.
30. N.K. Abrosimov and G.E. Kocharov, "The Effect of the Thickness of the Source on the Shape of the Energy Distribution and Angular Distribution Curves of α -Particles," Izvest. Akad. Nauk S.S.R., Ser. Fiz. 26, 237-44 (1962) (In Russian).

31. D. Yeates, S. Harris, and C. Doust, "Study of the Angular Distribution and Energies of Alpha Particles Emitted by ^{241}Am Sources," Nucl. Instrum. Methods 80, 320-4 (1970).
32. W.J. Price, Nuclear Radiation Detection, McGraw-Hill Book Co., New York, 2nd Ed. (1964).
33. H.A. Bethe and J. Ashkin, Experimental Nuclear Physics, Vol. 1, John Wiley & Sons, New York, (1953).
34. J.B. Dance, "Radioisotopes Speed Fire Detection," Instrument Practice, PP 975-79, (Dec. 1966).
35. E. Meili and T. Lamport, "Ionization Fire Alarm System", U.S. Patent #3,353,170 (Nov. 14, 1967) .
36. H. Carbonnel, "Low Current Ionization Chambers for Fire Detection", (In French), Presented at the Conf. on Rad. and Isotope Tech. In Civil Engineering, Brussels, (Oct. 1970).
37. G. Berio, "The Use of Ionization of Air for Protection from Lightening," No. 46, Bureau of Eurisotop, Brussels, Belgium (1970) (In French). Presented at the Conference on Radiation and Isotope Techniques in Civil Engineering, Brussels, Oct. 28-30, 1970.
38. C.M. Porsar, "Ionization Fire Detection Device", U.S. Patent #3,582,646, (June 1, 1971).
39. G.I. Mikhailov and L.P. Starchik, "Isotopic Analysis of Lithium Based on the Yield of the reaction $^7\text{Li}(\alpha, \alpha')^7\text{Li}$. At. Energ. (USSR), 24, 6, P 575 (1968).
40. R.N. Walz, "Nuclear Static Eliminators Design and Development," from 6th Annual Packaging Industry Technical Conference, Hartford, Conn., May 1966, ORAU. Gmelin (Conf. 660549) (AED Conf. 66-141-1).
41. F.J. Miller and P.S. Baker, "Radioisotopes and Radiation in the Textile Industry, ORNL-IIC-29, Oak Ridge National Laboratory (Feb. 1972).
42. J.L. Putman, "Static Elimination," AERE Radioisotope Review Sheet A6. October 1963.
43. I.A. Polonik, "Nuclear Radiation as a Means of Air Ionization for Neutralizing Electrostatic Charges in the Processing of Synthetic Fibers," in The Uses of Radioactive Isotopes and Nuclear Radiation in the USSR, Vol. 2, Proc. All Union Conf., Riga, April 12-16, 1960, PP. 50-4, P.I. Antyshev et al (Eds.), Moscow (1961), AEC-tr-6466/2, 1966.

44. I.V. Romeikov and A.V. Vladimirov, "Radioactive Isotope Method of Removing Static Electricity Charges," Poligrafiya 1, 29-30 (1965) (In Russian).
45. Z. Satorie and V. Borcevsky, "Radioactive Electrostatic Charge Eliminators," Jad. Energ, 15 (8), PP 278-81 (1969).
46. J.C. Legrand, "Elimination of Electrostatic Charges in the Textile, Paper, Rubber, and Plastics Industries with the use of the Ionotron Source," Supplement to Bulletin d' Information A.T.E.N. No. 68, (1967).
47. V. Mulclci and E. Gkeorghiu, "The Elimination of Electrostatic Charges in the Paper Industry," Inst. of Paper Chemistry, Appleton, W.I., (1969).
48. F.A. Hansen and B. Sellers, "Feasibility Study of Alpha Particle Densitometer for Measuring Planetary Atmospheric Density"; Final report, July 1968-May 1969, N-69-38649, Panametrics, Inc. (June 1969). Also summarized in Isotop. Padiat. Technol. 8, PP 185-6 (1970).
49. B. Sellers and P. Monee, "Design and Development of an Isotopic Altimeter Setting Indicator," FAA-RD-69-38 (Vol 1 and 2) Panametrics Inc. (June 1969).
50. W. Brandt, M.D. D'Agostino and A.J. Favale, "Alpha-Particle Denistrometer with Variable Response," Nucl. Technol, 11, (1) PP 99-104, (1971).
51. A.M. Nechushkin, P.M. Belash, V.F. Belov and A.L. Sarkisov, "Semiconductor Pressure Transducers for Gas Mixtures," translated by J. Bradley from Izv. Vyssh. Ucheb. Zaved, Neft. Gaz. 7(11), 89-91 (1964), SLAC-trans-43, Stanford Linear Accelerator Center.
52. R. Dagnac, "Vacuum Gauges Using Radioactive Sources Emitting Alpha Rays or Beta Rays," Vide 19, 135-40 (1964), bibliographic review in French.
53. Ya. Yu. Rebo and D.N. Ziv, "The Use of Alpha Emitters for Measuring Gas Density," translated from Trudy Vsesoyuz, Nauch., Tekh. Konf. Primenen. Radioaktiv. i Stabil. Izotopov i Izluchenii v Narod. Khoz. i Nauke, Moscow, 1957 (1958). Mashinostr. i Priborostr., PP. 280-5, AEC-tr-4492, PP. 322-8.
54. R.N. Brown, D. Balber, and G.V. Rylsky, "Device for Measuring Gas Pressure by Means of Alpha Particles," U.S. Patent No. 3,141,970, July 21, 1964 (filed May 15, 1961); assigned to General Precision, Inc.

55. R.P. Gardner and R.L. Ely, Jr., Radioisotope Measurements Application In Engineering, PP 287-302, Reinhold, N.Y. (1967).
56. P. Andrieux and B. Iapteff, "Improvement of Ionization Gauges Employing a Radioactive Source: L'Alphavac-500," Proc. of the Fourth International Vacuum Congress, Manchester England, (April 1968).
57. L. Gilly and P. Jourdan, "Testing in a Stratospheric Ballon a Semiconductor Detector ALtimeter," CEA-R-3516 (1968), Also see: CEA-R-3281 (1967).
58. L. Gilly, P. Jourdan and P. Denis, "Mach Meter Using Semiconductor Detectors for Nuclear Radiation," CEA-R-4107 (1971) - L. Gilly, "Nuclear Machmeter," Aeronaut. Astronaut (Paris), 27, PP 23-34 (1971).
59. F.L. Oetting, "Recent Calorimetric Determinations of the Specific Powers and Half-Lives for Certain Long-Lived Alpha-Emitting Radio-nuclides," The Dow Chemical Company, Golden Colo. US.A. (19).
60. F.L. Oetting, and S.R. Gunn, " A Calorimetric Determination of the Specific Power and Half-Life of Americium-241" J. Inorg. Nucl. Chem., 29, PP 2659-2664, (1967).
61. H.L. Davis, "Isotope Cost and Availability", In Hand Book of Nuclear Research and Technology, Nucleonics, McGraw-Hill.(1965)
62. C.A. Rohrmann, "Radioisotopic Heat Sources", HW-78180, HW-76323, (1963).
63. J. Freund, "Radioisotope and Thermoelectric, Conversion Generators", Energ. Nucl.(Paris) 10(7), PP 430-44, (1968).
64. V. Keshisian, "Radioisotope Generator with Limited Protective Shielding," Pub. Oct. 30, 1967.
65. "Apparatus to Supply Fluid for Heating Garments, e.g. Space and Diving Suits," Published Aug. 6, 1969.
66. R.T. Santoro, F.E. Gillespie and P.M. Aebensold, "Constant Current Source for Instrument Calibration," Rev. Sci. Instrum., 37(7), PP 919-22, (1966).
67. S.V. Damie and G. Joseph, "Radioactive Pulse Light Source for In-Flight Calibration of Scintillator Photo Multiplier Assembly," Nucl Instrum Methods, 65, PP 123-4, (Oct 1968).
68. G. Berio, "The Use of Ionization of Air for Protection from Lightning" (In French) Bureau Eurisotop, Brussels, Belgium (1970).
69. K. Ritter, "Lightening Rod with Ionizing Field," U.S. Patent No. 3,328,508, Published June 27, 1967 assigned to General Protection Establishment, Liechtenstein.

70. K. Ritter, "Lightning Rod with Great Ionizing Power," U.S. Patent No. 3,350,496, Oct. 31, 1967, assigned to General Protection Establishment, Liechtenstein.
71. T. Rommel, "Lightning Conductor System Using Radioisotopes", Conf. on Rad. and Isotope Tech in Civil Eng. (Sept. 1970).
72. J. Delhove, "A Study of the Radiological Aspects of the Manufacture and Use of Radioactive Lightning Conductors," EUR-4292.F. Euratom Brussels, Belgium (1970).
73. J. Delhove, "Study of Radiologic Aspects Connected with the Manufacture and Use of Radioactive Lightning Rods," J. Belge Radiol, 52, II-VII, (1969).
74. "Lightning Rods Go Radioactive," Elec. World, 173,(3) PP 36-7 (1970).
75. C.G. Invernizzi, "Air Conditioning by Ions," Conf on Rad. and Isotop Tech in Civil Eng., Eurisotop 51, (Sept. 1970).
76. C.G. Invernizzi, "Ionic Conditioning of Air," No. 53, Bureau Eurisotop Brussels, Belgium (1970) (In French), English: Translated by H. Warren, ORNL-tr-2427.
77. F.A. Howe, "A Critical Survey of Methods for Determination of Film Thickness Applicable to **Nuclear** Targets," Proc. of the Seminar on The Preparation and Standardization of Isotopic Targets and Foils, Harwell, Oct. 20-21, 1965. PP 123-7, M.L. Smith(Ed) AERE-R-5097 (Dec. 1965) (Conf. 651055).
78. H. Moret, "Thickness Measurement and Mass Determination of Evaporated Layers," Proc. of the Seminar on the Preparation and Standardization of Isotopic Targets with Foils, Harwell, Oct. 20-21, 1965, PP 128-34, M.E.Smith (ed). AERE-R-5097, (1965) (Conf. 651055).
79. H.L. Adair, "Target Thickness and Uniformity Measurement Using Charged Particles," Proc. of the Third International Symposium on Research Materials for Nuclear Measurements, Galtinburg, Tenn., 1971, PP 272-82, (Conf. 711002).
80. H.L. Anderson, "Thickness Measurements Using Alpha Particles," U.S. Patent 3,193,680, July 1965 (filled in 1960).
81. K. Teranishi, "Measurements of the thickness of Thin Polymer Films With a Semiconductor α -particle Detector," Nucl Eng. (Tokyo), 11, PP 46-56 (1965).
82. I. Nonaka and I. Sugai, "Alpha-ray Thickness Gauge," INS-T1-112, (July 8, 1971) (In Japanese).

83. D.G. Burke and J.C. Tippet, "An Application of Rutherford Scattering to Target Thickness Measurements," Nucl. Instrum. Methods, 63, PP 353-6, (1968) .
84. G.R. Shoemake, D.C. Fenimore and A. Zlatkin, "Radiation Sources for Ionization Detectors in Gas Chromatography," J. of Gas Chromatography, PP 285-86, Aug. (1965).
85. H.K. Hallows and A.E. Hodgson, "An Alpha-Particle Hygrometer", In Radioisotope Instruments in Industry and Geophysics, Proc. of the Sym. on Radioisotopes Instr. in Industry and Geophysics., IAEA, Vienna 1966, (Conf. 651023-8).
86. R. P. Gardner, K. Verghese, and J. I. Cehn, "A Mathematical Model for the measurement of Gas Properties with Alpha - Particle Gauges", Nucl. Tech. 16, (Nov. 1972)
87. J. H. Patterson et al., "Preparation of High Level Alpha Particle Sources for the Surveyor Alpha Scattering Experiment", Nucl Tech, 28, pp. 277 - 285, (1973)

D. Alpha Induced Fluorescence

Introduction

A very brief general discussion of fluorescence can be found in a later section on photon induced x-ray emission. For a formal treatment of charged particle excitation the reader is referred to the work by Metzbacher.⁽¹⁾

The cross-section for the excitation soft x-rays by incident alpha particles varies as Z^{-12} within the energy region of naturally emitted alpha particles.

The production of soft K x-rays from low Z elements ($Z \leq 15$), and it can similarly be shown for L and M x-rays from heavier elements, is therefore a most sensitive and efficient process. In addition the massiveness of the alpha itself, the impending collision process is not accompanied by any significant Bremsstrahlung radiation as would be the case if electrons were employed.

The most comprehensive work, particularly in the area of radioisotope induced fluorescence has been carried out by Sellers, Ziegler and their collaborators. Any investigator contemplating the use of α -emitting isotopes should definitely refer to their original papers^{(3) (4) (5)}.

Sellers and Ziegler have developed a yield equation and employ an approximate relationship (See Eq. 1) to predict α -excitation x-ray yields from proton data already available in the literature.

$$Y_{\alpha}(E) = 4Y_p(E/4). \quad (1)$$

In their various investigations they employed a ^{210}Po (α -5,3 MeV) source, and experimentally determined the x-ray yield per incident α particle. If one employed ^{241}Am (5.48 MeV alphas) similar yields would be attained. A composite table of Sellers' results over a period of years is contained in Table 1.(a,b) As it indicates the fluxes of x-rays obtainable per millicurie of alphas is quite acceptable especially in the case of K-x-rays for elements below $Z=13$.

The work by Sellers and his collaborators includes an investigation into secondary and accompanying interfering radiation produced in the collision process along with the fluorescent x-rays.⁽⁴⁾ Employing ^{210}Po , they found the intensity of α -photons and other emitted particles to be several orders of magnitudes less than the flux of excited x-rays. Alpha excited x-rays from a pure element target can thus result in the fabrication of a highly monoenergetic soft x-ray source. ^{241}Am which, is by large a prolific photon emitter, (Np x-rays and 60 KeV α photon) would not of course yield a monoenergetic source of soft alpha produced x-rays.

Applications

Several researchers have designed spectrometers utilizing α -emitting radioisotopes. Sellers and Ziegler performed feasibility studies considering ^{210}Po α 's for the study of extraterrestrial⁽⁴⁾ material surfaces.

Adler and Trombler, reported the design of and development of a lunar probe for just this purpose, under NASA funding.

Their first report⁽⁶⁾ summarizes the results of studies employing 20 mc ^{210}Po - α sources to study meteorite samples. In a later paper⁽⁷⁾ they indicate the parameters for a lunar probe capable of being transported by astronauts. Further discussion of the results of employing a lunar probe in the 1967 Surveyor V mission are contained in review of Radio-isotope Instruments by Cameron and Clayton⁽⁸⁾.

In papers delivered at the 2nd Low Energy X- and Gamma Source Symposium, workers from Japan⁽⁹⁾ and France⁽¹⁰⁾ report the use of α excited fluorescent x-rays to detect the presence of light elements with high S/n ratios. It is reported here and in other proceedings that Ushida⁽¹¹⁾ and co-workers have developed on-line-instrument, for the mixing control of cement raw materials, where such light elements as Mg, Al, Si are to be determined in a matrix containing various heavy elements. They employed two sources of ^{210}Po of 10 mCi activity.

At the same conference Martinelli and Robert et al. reported the use of ^{210}Po sources in investigations with samples of magnesium aluminum and agglomerated silica,⁽¹⁰⁾ and the initial phase of a study to determine the sodium content of glass. An application of ^{241}Am in α excited fluorescence is attributed to these individuals. Isotopes and Radiation Technology⁽¹²⁾ reported that Martinelli and his co-workers have developed a portable x-ray fluorescent analyzer utilizing both ^{210}Po and ^{241}Am alphas. Fig. 2, depicts a schematic diagram of the instrument.

Am-241--Possibilities

As the French have apparently demonstrated ^{241}Am does offer the possibility of being employed in the production of "soft" x-rays thru alpha excitation. The energy of emitted alphas are comparable to ^{210}Po

and it has the advantage of being a long lived radioisotope (458 years as compared to 138 days for ^{210}Po , 168 for ^{242}Cm).

The disadvantage of ^{241}Am is, of course, the accompanying emission of x and gamma photons. This may however be overcome by designing suitable detector systems and may in fact lead to an advantage which has not yet been fully realized. The simultaneous excitation of light and heavy elements, with ^{241}Am alphas excited by the characteristic K x-rays from the light elements, and the accompanying γ - and x-ray photons exciting the heavier elements. This detection technique has been employed by NASA with the ^{242}Cm sources in the Lunar Probe apparatus.

Table 1a
K Shell Alpha Excited X-Ray Yields
 $E_\alpha = 2.75 \text{ MeV}$ (Ref 6.)

Energy keV	Target	Principal Element	Z	(X-rays / α -sr)
0.28	C	Carbon	6	1.2×10^{-3}
0.39	BN	Nitrogen	7	2.9×10^{-4}
0.53	H ₃ BC ₃	Oxygen	8	2.9×10^{-4}
0.68	Teflon	Fluorine	9	5.8×10^{-4}
1.04	NaBH ₄	Sodium	11	1.7×10^{-4}
1.25	Mg	Magnesium	12	5.3×10^{-4}
1.49	Al	Aluminum	13	$2.7\text{--}2.4 \times 10^{-4}$
4.9	V	Vanadium	23	3.0×10^{-6}
8.9	Cu	Copper	29	5.8×10^{-6}

TABLE 1b Thick Target Characteristic X-Ray Yields (Ref 5.)

Energy kev	Element	Z	Shell	Yield (X-Rays/Alpha-Steradian)		
				2 mil Be Counter	5 mil Be Counter	Est. from Eq.
1.49	Aluminum	13	K		2.4×10^{-4}	4.4×10^{-4}
1.70	Tantalum	73	M		7.7×10^{-5}	
1.77	Tungsten	74	M		8.2×10^{-5}	
2.30	Molybdenum	42	L	2.7×10^{-5}	5.8×10^{-5}	2.3×10^{-5}
2.35	Lead	82	M	1.3×10^{-5}	4.6×10^{-5}	2.7×10^{-5}
2.83	Paladium	46	L	1.0×10^{-5}		
2.98	Silver	47	L	1.1×10^{-5}		1.6×10^{-5}
3.17	Uranium	92	M	7.3×10^{-6}		1.4×10^{-5}
3.28	Indium	49	L	8.6×10^{-6}		
3.44	Tin	50	L	8.2×10^{-6}	1.3×10^{-5}	
4.9	Vanadium	23	K	4.6×10^{-6}	8.0×10^{-6}	
5.9	Cobalt	27	K	3.3×10^{-6}		
7.5	Nickel	28	K	3.5×10^{-6}		
8.0	Copper	29	K	4.2×10^{-6}	5.8×10^{-6}	5.4×10^{-7}
8.1	Tantalum	73	L	1.9×10^{-6}	2.8×10^{-6}	6.3×10^{-7}
8.3	Tungsten	74	L	2.0×10^{-6}	2.3×10^{-6}	5.2×10^{-7}
8.6	Zinc	30	K	4.0×10^{-6}		
9.4	Platinum	78	L	1.1×10^{-6}		2.7×10^{-7}
9.7	Gold	79	L	1.2×10^{-6}		2.3×10^{-7}

Fig. 2. Schematic diagram of Ar iel Analyzer.

REFERENCES

1. Mertzbacher, E. Lewis, H.W., X-ray Production By Heavy Charged Particles, In Handbuch der Physik, Vol. 34, PP 166-192, Springer Verlag Berlin, 1958.
2. Mertzbacher, E., Inner Shell Ionization by Simple Charged Particles Invited paper given at the International Conference on Inner Shell Ionization Phenomena, Atlanta Georgia, April 1972.
3. Sellers, B., Zeigler, C.A., Generation and Practical Use of Monoenergetic X-rays from Alpha emitting Isotopes", ORNL-11C-5, Proceedings of the Symposium on Low Energy X and Gamma Sources, PP 353-373, Chicago, Oct 20-21, 1964.
4. Sellers, B., Generation and Practical Use of Monoenergetic X-rays from Alpha emitting Isotopes", Annual Reports, NYO-3491-1, -2, 3.
5. Sellers, B., Wilson, H.H., Papadopoulos J., Heavy Particle Excitation of Low Energy Characteristic X-radiation-Analytical and Experimental Correlations," Proceedings of 2nd Symposium on Low-energy X and Gamma sources and application P 576 Austin, Texas, 1967.
6. Adler, I., Trombka, J. "Rock Analysis by alpha excitation of x-rays-A possible Lunar probe," Proceedings of 2nd Symposium on Low energy x and gamma sources and applications P 929, Austin, Texas, March 27-29, 1967.
7. Trombka, J.I., Adler, I., et al, "Combined X-ray Neutron Experiment For the Exploration of Lunar and Planetary Surfaces" Applications of Low Energy X and Gamma Rays. Charles Ringler editor, Gordon and Breach, New York, 1971.
8. Cameron, J.F. and Clayton, C.G., Radioisotope Instruments Part I, PP 115-117.
9. Kato, Masao, "Present Status of research and Applications of Low Energy X and Gamma-ray sources in Japan.
10. Uchida, K. Tominaga, H., Imamura, H., Light elements Simultaneous Analyzer by the X-ray emission method using Alpha and X-ray Sources for cement raw mix control" Symposium Radioisotope instruments in Industry and Geophysics, Warsaw 1965, Vol I, 113, IAEA Vienna.
11. Martinelli, P., Robert A., and Cauailles J., Progress in the study and application of low energy-photon radioactive sources, Proceedings of 2nd Symposium on Low Energy X and Gamma sources and applications. P. 696.
12. French x-ray analyzer, Isotopes and Radiation Technology, Vol. 8-1 P. 122.

III. Americium-241 in Composite Neutron Sources

A. Encapsulation Methods and Am-Be Source Designs

The basic design of (α ,n) neutron sources has remained virtually unchanged. The sources normally consist of a mixture of an alpha emitter and beryllium (for example, americium - 241 and beryllium), the active material being in the form of a powder, pressed pellet or alloy. The main developments in neutron source fabrication ~~have~~ come in the form of encapsulation materials, sealing techniques and testing. A pressed pellet double encapsulation design is shown in Fig. 1. A detailed discussion of the fabrication of an Am-Be neutron source can be found in the review of Strain and Leddicotte⁽¹⁾ which dealt only with americium and its uses.

Recent review papers by Tolmie⁽²⁾ and Ansell and Hall⁽³⁾ have discussed the most modern developments in radioisotope neutron sources. In the review paper by Ansell and Hall⁽³⁾, several special purpose neutron source designs are discussed. Annular ^{241}Am -Be sources of neutron yields of $\sim 10^5 - 10^6$ n/sec which are used in soil moisture studies are reviewed (See Fig. 2). A similarly designed source containing 100 mCi of ^{241}Am has passed all the requirements for safe transport of radioactive materials. The annular source has also been used in fluorine analysis.

A rotory swaging technique developed at the Radiochemical Centre, Amersham is used to produce line sources. The technique is similar to that investigated in Japan for the production of fuel rods. The neutron sources are fabricated as follows: (a) The $^{241}\text{AmO}_2$ and beryllium metal powder are thoroughly mixed; (b) The powder mixture is then loaded into an aluminium tube and both ends of the tube sealed; (c) The sealed compact is extended in a rotory swaging machine to a wire of the required diameter; (d) The swaged wire is cut into the desired lengths and the wire is then doubly encapsulated

Fig. 1. Schematic drawing of Pressed Pellet Neutron Source.

Fig. 2. ANNULAR SOURCE FOR MOISTURE METER

(Ref. - 3)

Fig. 3. AMERICIUM-241-BERYLLIUM LINE NEUTRON SOURCE

in stainless steel tubing and sealed via argon arc welding. A design of a source produced by this method is given in figure 3. In spite of the length of this source, the variation of the neutron output along its active length is small ($\sim \pm 10\%$ see Ref. 2). Typical design data for both the pressed pellet and the line source are given in tables I and II.

All of the sources using stainless steel as a containment capsule are not recommended for use in situations where high temperatures are encountered, because beryllium becomes corrosive to stainless steel at temperatures around 600°C . The use of high temperature sintered compacts with beryllium oxide as a target represents one possible alternative to the standard neutron source design. The yield (n/sec/ci) for the mixed Be/AmO_2 and BeO/AmO_2 and the high temperature sintered compacts of Be/AmO_2 and BeO/AmO_2 are shown in Fig's. 4 and 5.

Steady state neutron sources consisting of intimate mixtures of an alpha source and beryllium have wide spread applications but there are uses where a radioisotopic source capable of being turned on and off has some advantages. One such design is described in reference 3. This source was developed to serve as a calibration standard in a neutron dosimeter. It consists of two half shells of α - emitting foil and beryllium attached to a small button magnet. The magnet equipped with spindles is mounted and sealed in a stainless steel capsule. A second magnet is mounted external to the sealed capsule and equipped with an operating spindle, rotation of which brings the alpha emitter and the beryllium target in to close proximity (See fig. 6). With 5 m ci of ^{241}Am , an output of 10^4 n/sec was obtained in the on position and 1.5×10^2 n/sec in the off position.

Fig. 4. EFFECT OF TARGET/NUCLIDE RATIOS ON NEUTRON YIELD
(Ref. 3)

Fig. 5. COMPARISON OF NEUTRON YIELD PER UNIT VOLUME
(ref. - 3)

Table I. Typical Design Data for Pressed Pellet Am - Be Neutron Source

Beryllium	0.50 gms
*Americium -241	91 m ci (.027 gms)
Chemical form	Am O ₂
Emission Rate	~ 2.61 X 10 ⁵ n/sec
Dimension	0.5" OD X 0.5 " OL

TABLE II

Americium-241-Beryllium Line Neutron Sources(Ref. 3).

EXTERNAL DIA. OF SWAGED ROD IN.	ACTIVITY/FOOT AT 10:1 Be/AmO ₂ ratio	NEUTRON OUTPUT/FOOT
0.050	30	7 x 10 ⁴
0.075	100	2.2 x 10 ⁵
0.100	150	3.3 x 10 ⁵
0.150	340	7.5 x 10 ⁵

Fig. 6. VARIABLE OUTPUT NEUTRON SOURCE

(Ref. - 3)

References

1. J.E. Strain and G.W. Leddicotte, "The Preparation, Properties and Uses of Americium-241, Alpha-Gamma-and Neutron Sources," ORNL-3335 (1962).
2. R. W. Tolnie, "Recent Developments in Radioisotope Neutron Sources", Proc. of ANS Topical Meeting on Neutron sources and Applications, VI, Augusta, GA, (1971).
3. K. H. Ansell, and E. G. Hall, "Recent Developments in (α ,n) Sources", Proc. of ANS Topical Meeting on Neutron Sources and Applications, Augusta, GA, VII, April (1971)
4. IAEA, "Regulations for the Safe Transport of Radioactive Materials". 1967 Edition., Vienna; 1967, IAEA Safety Series, No. 6.
5. K. Naruki, "Vib-Swaying Process for Fuel Rod", J. Nucl. Sci. and Tech., 7, p. 327 (1970)
6. J. D. Baird, G. A. Geach, A. G. Knapton and K. B. C. West, "Compatibility of Beryllium with other Metals used in Reactors", : Proc of the 2nd UN Int. Conf. on Peaceful Uses of Atomic Energy, 5, p. 328 (1958).

B. NEUTRON SPECTRUM -- ^{241}Am -Be ^{241}Am - ^{242}Cm -Be SOURCES

1. Review of Methods of Measuring Absolute Neutron emission rates and energy spectra of neutron sources.

The $^9\text{Be}(\alpha, n)^{12}\text{C}$ reaction ($Q=5.7$ MeV) is the most commonly used reaction in radioactive neutron sources. (1) (2) Only after the early sixties did Americium-241 become available in sufficient quantities to allow its use in the preparation of neutron sources. Prior to this time the α -emitters most often used were ^{210}Po , ^{239}Pu and ^{226}Ra . All of these alpha emitters are widely used to date and, depending on the applications, each has advantages and disadvantages. ^{210}Po has a short half-life (138 d), ^{226}Ra is an intense γ -emitter and ^{239}Pu neutron sources tend to be bulky. We have deliberately excluded ^{252}Cf neutron sources because it is not a (α, n) neutron source. ^{241}Am seems to have the desirable properties of an isotope for use in neutron sources. It has a low energy gamma emission, low bulk and a simple decay correction owing to its very long-lived daughter (2.2×10^6 y) and it can be obtained isotopically pure.

Neutron sources are used in a variety of applications. In those applications where the neutron source is used as a standard, it is important that one know something about emission rate and energy distribution of the neutrons from the source.

Coswell⁽³⁾ has reviewed the methods of neutron emission rates and spectra measurements with emphasis on the manganous sulfate bath method. In this method, neutrons emitted from the source located at the center of the bath, enter the bath and are slowed down by a series of collisions, and eventually captured by the ^{55}Mn nuclei present in the bath. The activity of the ^{56}Mn

nuclei is then measured to ascertain the emission rate of the source. The measured ^{56}Mn activity with corrections for leakage of neutrons and capture by elements other than ^{55}Mn is used to estimate the source emission rate. The principle of the method is shown in Fig. 1.

Geiger and Hargrove⁽⁴⁾ measured the energy spectrum of an alloyed ^{241}Am -Be source using a photon recoil spectrometer and a time-of-flight method. The photon recoil telescope measures the energy of recoil photons produced by the neutrons in a thin hydrogenous radiators. Only those photons scattered through small angles in the forward direction are accepted by the telescope.⁽⁵⁾ Two radiators of glycerol tristearate, evaporated on tantalum (1.17 and 3.28 mg/cm² thick) and 17.5 mg/cm² thick polyethylene on a tantalum backing were used to measure a partial spectra (~1.20 Mev to 8 MeV . The spectrums measured by both methods are shown in Fig.2.

The ^{241}Am -Be neutron spectrum was measured and calculated taking into account the anisotropy in the angular distribution of neutrons from the ^9Be (α ,n) reaction by Notarrigo.⁽⁶⁾ The ^{241}Am -Be source supplied by Radiochemical Center Amersham had an activity of one Ci (2.5×10^6 neut/sec). The neutron spectrometer consisted of a polyethylene foil shaped to allow only photons ejected with a fixed angle ($\theta = 45^\circ$) to reach the detector. (A silicon detector of 0.5 cm² sensitive area). The counter had cylindrical symmetry.

A comparison of peak energy positions (MeV) obtained by Notarrigo et al with values obtained by others^{(4) (7)} for ^{241}Am -Be Sources is given in table I,

Figure 1 . Manganous Sulfate bath source calibration (Schematic).

Figure 2 . Bottom: Neutron spectrum of $\text{Am}^{241}\text{-Be}(\alpha, n)$ source. The dotted line is an estimate for the low energy group. Centre: The "classical" Po-Be spectrum of Whitmore and Baker). Top: Partial spectrum for transitions into the 4.43 MeV state of C^{12} . The dotted spectrum is calculated). (Reference 4).

Am-Be ⁽⁷⁾	Am-Be ⁽⁴⁾	Am-Be ⁽⁶⁾
3.3	3.25	3.6
		4.1
5.25	5.0	4.8
5.8		5.5
7.1		6.7
8	8	7
	10	9

Table I. Comparison of peak energy positions (MeV) of measured spectra of ²⁴¹Am-Be.

The peak energy values of Thompson and Taylor⁽⁷⁾ were obtained by using a photon recoil spectrometer. This consisted of a 2" diameter by 1" thick stilbene crystal mounted on a photomultiplier tube. The 100 mCi Am-Be was placed 6" in front of the detector which was shielded on all sides by 2" of lead and 6" of paraffin. The neutrons were recorded as recoil photons resulting from the scattering of neutrons by hydrogen nuclei in the scintillator. The measurement covered the range of energies from ~ 2.5 MeV up to above 11.5 MeV. (See figure 3).

A recent measurement of the ^{241}Am -Be spectrum including the low energy region has been reported by Werle⁽⁸⁾ and is shown in Figure 4. This spectrum was measured down to energies below 200 keV. A unique method of determining the spectrum of the ^{241}Am (α, n)Be source has been applied by Geiger and Van Der Zwan⁽⁹⁾. This method involves bombarding a thick ^9Be target with a beam of alphas of a particular energy to simulate the neutron spectrum.

Below we shall report on the results of the spectra measurements for ^{241}Am -Be completed in connection with this work.

2. Results of Spectra Measurements of Two Commercial ^{241}Am -Be sources

Muckenthaler, Freestone and Henry⁽¹⁰⁾ of the Tower Shielding Facility at ORNL made a recent attempt to measure the spectra of two low intensity ($\sim 10^5$ n/sec) commercial sources at the request of the authors of this report. The detectors were spherical proportional counters, 4.64 cm in diameter, containing 1, 3 and 10 atmospheres of hydrogen. The pulse height spectra were unfolded to produce neutron energy spectra using the IBM 360 Program, SPEC 4, of Benjamin and Kemshall⁽¹¹⁾.

The sources used in the measurements were supplied by Monsanto and

Figure 3 Spectrum of neutrons from Am- α -Be source. (Reference 7).

Amersham/Searle and were of low intensity, 2.61×10^5 n/sec and 2.4×10^5 n/sec respectively.

The Amersham source was placed at 1-30/32 in. from the detector (center to center), while for the Monsanto source the separation distance was 1-13/16 in. The results of these measurements are given in tables II and III and Figures 5 and 6. The general shapes of the distributions are very similar as was expected. These spectra agree with those of Werle⁽⁸⁾ for an Am-Be neutron source. The sharp increase in intensity at 100 keV and below were attributed to the effect of gamma field associated with the source or the detector response. The gamma spectrum of an Am-Be neutron source as measured by the Radiochemical Centre of Amersham is presented in Fig. 7.

TABLE II

TUSKEGEE AM-BE SOURCE FROM AMERSHAM AT 1 15/16 in. FROM DETECTOR

ENERGY BOUNDARY (MEV)		FLUX (N/SQ-CM/MEV/MIN)	ERROR (PERCENT)
0.0612	0.0740	0.7523E 06	0.67
0.0740	0.0904	0.1115E 06	2.78
0.0904	0.1105	0.3859E 05	6.77
0.1105	0.1342	0.2167E 05	11.18
0.1342	0.1635	0.1806E 05	11.85
0.1436	0.1756	0.2723E 05	3.62
0.1635	0.2000	0.1732E 05	10.64
0.1756	0.2121	0.1520E 05	6.57
0.2121	0.2577	0.1283E 05	7.04
0.2577	0.3169	0.1250E 05	6.21
0.3169	0.3853	0.8448E 04	9.18
0.3853	0.4674	0.8278E 04	8.80
0.4591	0.5591	0.8646E 04	4.13
0.4674	0.5723	0.7832E 04	8.00
0.5591	0.6864	0.7212E 04	4.52
0.5723	0.7000	0.6666E 04	8.50
0.6864	0.8409	0.5696E 04	5.51
0.8409	1.0227	0.4405E 04	7.04
1.0227	1.2500	0.4078E 04	6.78

TABLE III

TUSKEGEE AM-BE SOURCE FROM MONSANTO AT 1 13/16 in. FROM DETECTOR

ENERGY BOUNDARY (MEV)		FLUX (N/SQ-CM/MEV/MIN)	ERROR (PERCENT)
0.0637	0.0744	0.2021E 06	1.62
0.0744	0.0888	0.4419E 05	5.34
0.0888	0.1031	0.3596E 05	7.21
0.1031	0.1229	0.2281E 05	8.54
0.1229	0.1444	0.1121E 05	17.71
0.1444	0.1695	0.1084E 05	17.26
0.1695	0.2000	0.1056E 05	15.76
0.1894	0.2259	0.1184E 05	7.82
0.2259	0.2669	0.1240E 05	7.58
0.2669	0.3125	0.1303E 05	7.35
0.3125	0.3673	0.9157E 04	9.64
0.3673	0.4312	0.7496E 04	11.23
0.4312	0.5087	0.7112E 04	10.72
0.4682	0.5500	0.7960E 04	9.71
0.5087	0.6000	0.5686E 04	12.57
0.5500	0.6500	0.6816E 04	10.58
0.6500	0.7682	0.5984E 04	11.70
0.7682	0.8955	0.4114E 04	18.36
0.8955	1.0591	0.4217E 04	15.25
1.0591	1.2500	0.2954E 04	20.97

Figure 4. ^{241}Am -Be Neutron Spectrum Measured with Proton-Recoil counter by Werle (8). The spectrum is compared to that of Geiger and Hargrove (4).

FIG. 5 AMERSHAM AM-BE (AMN 100-4148) AT 1 15/16 IN.

FIG. 6 TUSKEGEE AM-BE SOURCE AT 1 13/16 IN FROM DETECTOR.

Fig. 7. GAMMA SPECTRUM FROM
Am241/Be Neutron Source,
Measured on 3" x 3" NaI crystal

3. Americium-Beryllium-Curium Source

To fabricate the ^{241}Am -Be- ^{242}Cm neutron source, pressed pellets composed of ^{241}Am oxide on powdered beryllium metal are irradiated in a reactor to produce ^{242}Cm by the reaction $^{241}\text{Am}(n, \alpha) ^{242}\text{Am} \xrightarrow{\beta^-} ^{242}\text{Cm}$ (See Fig. 8). From Fig. 8 one observes that a fraction of ^{241}Am undergoes fission⁽¹¹⁾. Also, the product is divided between two isomeric states of ^{242}Am (an excited and ground state). Only the ground state decays to ^{242}Cm . The ground state (isomer also has an electron capture mode of decay to ^{242}Cm). The maximum ^{242}Cm production regardless of quality, is obtained at a neutron flux of $8 \times 10^{14} \text{ n/cm}^2 \text{ sec}$.

Gillette⁽¹²⁾, in a review of the radioisotopes Program at ORNL, presented neutron spectrums for an ^{241}Am -Be- ^{242}Cm and an ^{241}Am -Be in the energy range of 3 to 11 MeV (See Fig. 9).

Figure 8. Production of ^{242}Cm by neutron irradiation of ^{241}Am . (Reference 11).

Figure 9 . Neutron Spectrum from $^{241}\text{Am Be-}^{242}\text{Cm}$ and $^{241}\text{Am Be}$ Sources.
(Reference 12),

Summary

The neutron energy spectrum of the $^{241}\text{Am-Be}$ and $^{241}\text{Am-Be-}^{242}\text{Cm}$ neutron sources have been measured by several investigators. Most of the accurate spectra measurements have been in the range of one MeV or greater. The measurements of Werle⁽⁸⁾ and Muckenthaler et al⁽⁹⁾ (The work of Ref. 9 was done in connection with this study) are the only two measurements that have been done below 500 keV (as far as could be determined by the authors). The measurement of Werle extended down to 10 keV while that of Ref. 9 extended down to ~ 60 keV.

REFERENCES

1. W.N. Hess, "Neutrons from (α ,n) Sources," Annals of Phys., 6, P. 115 (1959).
2. A. Rubbino and D. Zubke, "Neutron from (α , n) Sources", IL Nuovo Cimento, XLIV B, N.I., (1966).
3. R.S. Coswell, "Review of Measurements of Absolute Neutron Emission Rates and Spectra from Neutron Sources", In Neutron Sources and Applications, Proc. of the Am. Nucl. Soc., Augusta, Ga., Conf-710402, (April 19-21, 1971).
4. K.W. Geiger and C.K. Hargrove, "Neutron Spectrum of an ^{241}Am -Be (α ,n) Source," Nucl. Phys., 53, PP 204-08, (1964).
5. K.W. Geiger and C.J.D. Jarvis, Can. J. Phys. 38, PP. 569, (1960).
6. S. Notarrigo, F. Porto, A. Rubbino and S. Sambataro, "Experimental and Calculated Energy Spectra of Am-Be and Pu-Be Neutron Sources," Nucl Phys A 125, PP 28-32, (1969).
7. M.N. Thompson and J.M. Taylor, "Neutron Spectra From Am- α -Be and Ra- α -Be Sources", Nucl Instr, 37, P. 305 (1965).
8. H. Werle, "Spektrumsmessung Radioaktiver Neutronen-quellen im Energiebereich von 10 keV bis 10 MeV mit. Protonenruckstoss-Proportionalzahlrohren", Kernforschungszentrum Karlsruhe, Report INR-4 120 - 25 (1970).
9. K.W. Geiger and L. Von der Zwan, "The Neutron Spectrum of a ^{241}Am -Be (α ,n) Source as Simulated by Acceleration produced α -Particles," Int. J. of Appl. Rad. and Isotopes, 21(4), (1970).
10. F.J. Muckenthaler, R.M. Freestone and K.M. Henry, "Am-Be Neutron Spectra in the Kilovolt Region," ORNL-CF-73.5.57 (1973).
11. P.W. Benjamin and C.D. Kemshall, "The Analysis of Recoil Proton Spectra", AWRE Report No. 0-7167 (1967).
12. R.D. Baybarz, "Recovery and Application of the Transuranium Elements: ^{237}Np , ^{238}Pu , ^{241}Am , ^{242}Cm , ^{244}Cm and ^{252}Cf ," AT.Energy Rev., 8, No. 2, IAEA, Vienna, (1970).
13. J.H. Gillette, "Review of Radioisotopes Program, 1964," ORNL-3802,

C. ²⁴¹Am-Be As a Source of Fast Neutrons in Neutron Moisture Gauges

1. Introduction

The quest for a clear understanding of hydrological phenomena in order that mankind may be successful in water resource planning has lead to the development of a broad range of nuclear techniques for use in hydraulics. Single and dual photon attenuation techniques have been used by many investigators in soil moisture studies to determine both bulk density and moisture content (See section IV page 194). The tendency is to employ both gamma and fast neutron sources for measurements of soil bulk density and moisture content. Nucleonic techniques are used in soil moisture studies because they are:

- a) Non-destructive
- b) Repetitive
- c) Rapid (requiring only 2-5 min/measurement)
- d) Convenient

and

- e) In Situ measurements are possible

There are some disadvantages associated with the nuclear techniques and these are:

- a) High cost of apparatuses
- b) All commercially available units are heavy

and

- c) Two different probes are required to measure the moisture content of the whole soil profile.

Various neutron sources are used in soil moisture studies. In the past the most widely used neutron source was ²²⁶Ra-Be, but it has the disadvantage of having a high γ/η ratio (See table I).

TABLE I

SOURCE	HALF LIFE	NUMBER OF ALPHAS	E _{AV} ALPHAS	n/sec/Ci*(1)	MR/hr/ Meter per 10 ⁶ Neutrons
Ac-Be	22 Y			1.5×10^7	8
²⁴¹ Am-Be	458 Y	1	5.4	2.2×10^6	1
²⁵² Cf	2.65 Y	NONE		1.0×10^6	--
²³⁹ Pu-Be	24400 Y	1	5.1	1.5×10^6	<1
²¹⁰ Po-Be	138 d	--	--	2.5×10^6	0.04
²³⁸ Pu-Be	86.4 Y	1	5.5	2.2×10^6	<1
²²⁶ Ra-Be	1620 Y	4	5.6	1.3×10^7	60

* The neutron yield of ²⁵²Cf is in terms of n/sec/μg.

TABLE I. Neutron sources used in Soil Moisture Studies.

In terms of neutron yield per mCi, ^{226}Ra -Be is about 6 times as efficient as ^{241}Am -Be but the lower specific gamma activity makes Am-Be sources preferable in most cases.

Methods of determining moisture in solids can be classified as either direct or indirect. In the direct category are those methods that determine the weight loss after the moisture has been removed, those where the moisture is condensed and its volume measured and lastly, those that involve quantitative chemical analysis. The indirect methods utilize the variation of some measureable property as an indication of moisture content. The nucleonic method constitutes one of many different indirect methods of measuring moisture content. (A review of methods of moisture measurements and comparison is given by Szulmager.(2)).

The neutron moisture gauge was originally utilized in ground water surveys and concrete moisture measurement, following the publication of results obtained in soil moisture measurements studies in the early 1950's. Since that time the use of this technique has grown steadily. The rate of acquisition and accumulation of neutron moisture gauges in the USA is given in figure 1. The accuracy of neutron moisture gauges is a function of the precision of the gauge, errors in calibration, and the compatibility between lab and field conditions. However, these gauges typically are capable of accuracy to within $\pm 0.01 \text{ g H}_2\text{O/cm}^3$ (4) for very precise devices and they can be used to measure soil moisture from full saturation up to dry conditions.(5)

Fig. 1. Cumulative number of neutron moisture meters years purchased up to 1967.

*Data taken from a survey by Gardner⁽³⁾.

2... Principles of the use of fast neutrons in moisture detection

In general neutrons emitted from neutron sources are not mono-energetic. The spectrum of energies of emitted neutrons ranges from 0 to ≈ 11 MeV.⁽⁶⁾ Since the bulk of these neutrons have energies above 100 kev, the sources are commonly referred to as fast neutron sources.

The fast-neutrons emitted from the source move into the medium, the moisture content of which is to be determined, and undergo collisions with the atomic nuclei of the medium. The decrease in energy of the neutron as a result of a collision is strongly dependent on the mass of the target nucleus. The scattering of the neutron may be elastic or inelastic, depending on whether the energy loss suffered by the colliding neutron is partly spent on exciting the target nucleus or is merely transferred to the target without changing form. Since the inelastic scattering threshold of light nuclei is relatively high, the elastic scattering process is the most dominant mode of interaction for neutron energies greater than 1 ev with light nuclei. This results from the fact that the energy of the first excited state is generally greater than 4 MeV (e.g. 4.43 MeV for ^{12}C and 6.42 MeV for ^{16}O) and the incoming neutron must have a laboratory energy of

$$E_{\text{lab}} = \left(\frac{A + 1}{A} \right) E \quad (1)$$

where

A = mass number

E = energy of first excited state

Utilizing the fact that both kinetic energy and momentum are conserved in an elastic scattering event, it is easy to show that the final neutron energy after a head on collision is

$$E_I = E_F + \frac{4A}{(A+1)^2} E_I \quad (2)$$

where

E_I = Initial energy of the neutron

E_F = Final energy of the neutron

A = The target nucleus Mass Number

Equation (2) reveals the importance of the mass of target nucleus. For example, for hydrogen, with a mass number of one, a neutron can lose all of its energy in a single collision. But since soils are composed of several different elements all of which present to the neutron a certain probability of interaction, the total microscopic cross-section for a multicomponent media is

$$\Sigma = 6.02 \times 10^{-3} \rho \sum_{i=1}^m \frac{w_i \sigma_i}{A_i} \quad (3)$$

where

w_i = weight percentable of i^{th} element.
 σ_i = the microscopic X-section of i^{th} element.
 A_i = its mass number
 ρ = the density of the material

Of the various types of nuclear interactions, the elastic collisions are of prime importance in understanding the moderation of the fast neutrons.

For neutron energies in the range of 1 ev or greater scattering generally predominates over absorption and the moderation of neutrons in the medium is governed almost exclusively by the water present. As the neutron reaches the thermal energy range (0-1 ev), absorption becomes an important mechanism, and the amount of absorption is directly related to the composition of the medium through Σ_a . The problem of the absorption process removing some of the thermalized neutrons can be eliminated by measuring the number of neutrons in an energy range where scattering is still predominant and absorption processes are small (i.e. $E \approx 1 \rightarrow 10$ eV). Neutrons in this range are essentially only scattered neutrons, their number is directly related to the water content, and the measurement is not dependent on the composition of the medium. The microscopic absorption cross-section of the absorbers is substantially reduced due to their $1/v$ dependence in the 1 - 10 ev range, while the value of Σ_s of water has dropped only from about 100 to 40 barns.⁽⁷⁾ Measurement of the epithermal flux has one other advantage, i.e., the epithermal energy is greater than the binding energy of the molecules; hence, the incoming neutron sees individual atoms of the H₂O molecule. However, the epithermal flux has the disadvantage of being 1/10 that of the thermal flux. This problem can be ameliorated by counting for longer times. In spite of the fact that an epithermal neutron meter would be relatively insensitive to the composition of the medium, thermal neutron moisture meters have enjoyed wide use. These probes are sensitive to the composition and dry bulk density of the soil under investigation and thus one can not obtain a general calibration curve that holds for all soils. Application of these probes in the field on different soils, then necessitates the development of practical theoretical models

that would allow for the variation in the dry bulk density and the chemical composition of soils.

3. Theoretical Models

There are several methods of arriving at analytical models to compare with experimental measurements of soil moisture content. These models make use of Fermi-Age Theory, multigroup theory, neutron transport theory and the Monte Carlo methods. Table II gives some of the analytical expressions subsurface probes (see figure 2) derived using some of these methods. Invariably many simplifying assumptions are made in most of the theories used. For example, the Fermi-Age theory method assumes that there is a negligible amount of absorption of the neutrons while slowing down. However, with the use of this method and the more complex methods, The agreement between theory and measurements are good enough to predict the response of neutron gauges to various factors. Several authors⁽¹¹⁾ ⁽¹²⁾ have investigated the effects of detector shape and geometry on the average neutron flux at the detector. The inclusion of the source-detector geometrical effects and epithermal absorption of neutrons in the detector tends to make the determination of analytical expressions for $\phi_T(r)$ more difficult. Other techniques of considerable complexity such as the transport theory and the Monte Carlo methods⁽¹³⁾ have been used to determine the thermal neutron flux. But the large amount of computational time required to obtain sufficient accuracy on digital computers have made their methods "last resort" techniques.

4. Surface Moisture and Transmission Gauges

Surface moisture gauge. (see figure 3) has been treated by Gardner and Roberts⁽¹⁴⁾ using a two group diffusion model for a point source located in the center of a spherical cavity in an infinite homogeneous medium. Recently,

TABLE II. Some analytical expressions describing the thermal flux in infinite mediums.

METHOD	NEUTRON FLUX AS A FUNCTION OF POSITION
Fermi-Age	$\phi_T(r) = \frac{S e^{\tau_T/L_T^2}}{8\pi\bar{D}r} \left(e^{r/L_T} \left[\operatorname{erf}\left(\frac{r}{\sqrt{\tau_T}}\right) + \frac{\sqrt{\tau_T}}{L_T} \right] + \exp\left(-\frac{2r}{L_T}\right) \operatorname{erf}\left(\frac{r}{2\sqrt{\tau_T}} - \sqrt{\tau_T}/L_T\right) - \right.$ $\left. 2e^{-r/L_T} \sinh r/L_T \right] \quad (8) \quad (9)$
One-speed	$\phi_T(r) = \frac{S}{4\pi\bar{D}r} e^{-r/L}$
Multigroup Theory (Two-groups)	$\phi_T(r) = \frac{S L_T^2 [(\exp(-r/L_T) - \exp(-r/\sqrt{\tau_T}))]}{D_T 4\pi r (L_T^2 - \tau_T)} \quad (10)$
Three-groups	$\phi_T(r) = \frac{S}{4\pi\bar{D}r} \left(\frac{L_T^2}{L_{f1}^2 - L_{f2}^2} \left(\frac{L_{f1}^2}{L_{f1}^2 - L_T^2} \right) \exp(-r/L_{f1}) + \frac{L_{f2}^2 L_T^2 \exp(-r/L_2)}{(L_{f2}^2 - L_T^2)(L_{f2}^2 - L_T^2)} + \right.$ $\left. \frac{\exp(-r/L_T) L_T^4}{(L_T^2 - L_{f1}^2)(L_T^2 - L_{f2}^2)} \right) \quad (11)$
	<p>where: L_T, L_{f1}, and L_{f2} are the slowing down length for thermal, the highest fast and lower fast groups, respectively. τ_T is neutron age to thermal energies. $\bar{D}$ is the mean diffusion coefficient. S is the source strength (neutrons/seconds)</p>

Figure 2. Typical subsurface: 1) Fast neutron source, 2) detector, 3) preamplifier, 4) container, 5) cable.

Figure 3. Surface Moisture Gauge: 1). neutron detectors; 2). neutron source; 3). soil.

Gemmell, McGregor and Mass⁽¹⁵⁾ simulated the surface probe technique using a digital computer solution to the Boltzmann Transport Equation in multi-energy group form. A surface probe experiment on concrete was analyzed and the agreement between theory and experiment was better than 15 per cent.

The simplest moisture gauge theory is that of the attenuation on transmission type. The bulk density and the moisture content are given by⁽¹⁶⁾

$$\rho_s = \frac{\mu_w \ln(I/I_o)_n - \mu_{wn} \ln(I/I_o)_\gamma}{\mu_{wn} \mu_{s\gamma}^X - \mu_{sn} \mu_{w\gamma}^X} \quad (4)$$

and

$$\theta = \frac{\mu_{s\gamma} \ln(I/I_o)_n - \mu_{sn} \ln(I/I_o)_\gamma}{\mu_w \mu_{sn}^X - \mu_{s\gamma} \mu_{wn}^X} \quad (5)$$

respectively, where I_o , I , μ_{sn} , μ_{wn} , $\mu_{s\gamma}$, and $\mu_{w\gamma}$ are the radiation intensities before and after passage through the soil sample and container and the respective neutron and gamma attenuation coefficients for oven dried soil and water (cm^2/g). The subscripts n and γ refers to neutrons and gamma rays, respectively.

These models can be used to calibrate moisture gauges if all of the parameters are known. Normally, a model is constructed that describes the response of the gauge over the range of moisture contents, dry compositions and dry densities encountered in the field. Then a number of laboratory calibration samples can be used to determine the constants of the chosen model for a particular gauge. Some of the factors that influence moisture gauges response are given in Table III.

TYPE OF GAUGE	FACTORS INFLUENCING MOISTURE GAUGE RESPONSE
Both Surface and Subsurface gauges	<ul style="list-style-type: none"> a) Source-detector geometry b) Type of detector c) Construction material of access tube d) Soil parameters - <ul style="list-style-type: none"> 1. Dry bulk density 2. The amount of bound water content 3. Concentration of elements with large absorption cross section.
Subsurface Probes	<ul style="list-style-type: none"> a) Compaction of the soil when the access tube is inserted and cavities b) Variation in the access tube diameter and it's wall thickness c) Leakage of water and condensation in the access tube d) The effect of near surface measurements
Surface Probes	<ul style="list-style-type: none"> a) Reproducible geometry b) Smoothness of the surface of the soil c) Compression of the soil by the instrument.

TABLE III. Some factors that influences the response of a neutron moisture gauge.

4. Radiological Hazards

Neutron moisture gauges unlike conventional moisture measuring apparatuses are radiological hazards. The gamma exposure rate in mR/hr/m per 10^6 neutrons for several neutron sources that are used in soil moisture studies are given in table I. One sees that there is a negligible amount of exposure from photons for sources of this strength at one meter for all of the sources except $^{226}\text{Ra-Be}$, but they all pose neutron hazards to potential users. ^{252}Cf is included in table I for comparison since it has recently been employed as a source of fast neutron in a moisture study by Corey et al. (16)

The most commonly used neutron sources (i.e. in moisture gauges) are $^{241}\text{Am-Be}$ and $^{226}\text{Ra-Be}$. For unshielded $^{241}\text{Am-Be}$ and $^{226}\text{Ra-Be}$ point isotropic sources of 30 mCi and 10 mCi, respectively, the variation of neutron dose rates as a function of positions are given in figures 4 and 5. However, these dose rates are significantly lower for a commercially available gauge. The unshielded dose rate for a 3 μg ^{252}Cf source ($\approx 3 \times 10^6$ neutrons/sec) is ≈ 6.6 m rem/hr at one meter. (18) The cost of ^{252}Cf sources of adequate strengths for accurate neutron moisture measurements is prohibitive, thus it is, at the present time, not truly competitive with $^{241}\text{Am-Be}$ or $^{226}\text{Ra-Be}$. The negligible photon hazard of small $^{241}\text{Am-Be}$ neutron sources obviously enhances their employment as the source of fast neutrons in gauges. The source strength can be materially increased and the weight of the probe and accessories decreased, thereby increasing the precision and convenience of the neutron gauge used.

Figure 4. Variation with distance of neutron dose rate from a 30 mCi ^{241}Am -Be neutron source. (17)

Figure 5. Variation with distance of neutron and photon dose rates from a 10 mCi ^{226}Ra -Be neutron source. (17)

5. Results of Soil Moisture Studies

Soil moisture determination in which the neutron scattering principle was used had their beginning in the early 1950's. Early measurements used ^{226}Ra -Be as the primary source of fast neutrons for the neutron meters. Today, even though ^{241}Am -Be is becoming more common as the source of fast neutrons, in those cases where the experimentalist or technician are interested in the simultaneous determination of bulk density and moisture content of soils a single ^{226}Ra -Be⁽¹⁹⁾ neutron source can be used. Whereas in a similar situation ^{241}Am -Be and a gamma emitter (e.g. ^{137}Cs) would be needed for the same measurements. If it is desired to determine the water content by weight percentage one needs to know the density of the material involved; that is, an accurate density measurements cannot be performed on a system with varying moisture content without a parallel determination of the moisture content and vice versa.

Calibration of neutron moisture gauges is covered in detail in, Neutron Moisture Gauges⁽¹⁸⁾. This guide book covers the theory of neutron interaction with matter, gauge design, accuracy, health and safety from a radiological point of view and the factors influencing the selection of a particular gauge. A large number of the reports on neutron moisture gauges have been devoted to the calibration of these instruments. Efforts have ranged from the experimental methods using soil samples to theoretical models described in previous section. McHenry and Gill⁽²⁰⁾, examined the influence of bulk density, slow neutron

absorbers and time on the calibration of neutron probes. Measurements with several different probes using ²²⁶Ra-Be and ²⁴¹Am-Be were carried out over a range of moisture content (oven dry to saturation) on Grenada silt loam soil. The results for several dry densities are shown in figure 6 and the effect of a slow neutron absorber in figure 7. Jensen and Somer⁽²¹⁾ using a 30 mCi ²⁴¹Am-Be source measured calibration curves for the same soil type of different dry-soil bulk densities (see figure 8). Unlike the measurements of reference⁽²⁰⁾ in which a 100 mCi ²⁴¹Am-Be source was employed these results reveal significant changes in count rates as a function of dry density changes. In both cases, however, it was shown that the linear calibration curve was of the form

$$Y = m(CR) - b \quad (6)$$

where

Y = moisture content in vol %

m = slope of the calibration curve $\left(\frac{dw}{dCR}\right)$

CR = count rate

b = dry-soil water equivalent (mx count rate at 0% moisture).

In equation (6) m and b for a moisture-reading count rate meter corresponds to range and zero suppression. Most of the experimental results, in which ²⁴¹Am-Be has been employed in moisture meters to date, have monitored thermal neutrons. This type of moisture gauge is greatly affected by the types and quantities of solids in the soil or media. A device, that makes use of epithermal neutrons, in a moisture meter capable of field measurements was designed and built by Ringle, Bell and Hurley.⁽²²⁾ The moisture meter can be used in measurements of moisture over a range of 20 to 350% with an average error in the moisture content of 16% (maximum 32%) if no information on the sediment is available; and 9% (maximum 23%) if information is available.

Figure 6. Calibration of a neutron probe using an Am-Be source in a Grenada silt-loam soil at differing dry bulk densities. (20)

Figure 7. The effect of boron concentrations on the slow neutron flux measured by a USDA Sedimentation Laboratory neutron probe at differing moisture percentages. (20)

FIG. 4. Calibration curves (vol. % H_2O).
Rising soil at different dry densities.

Figure 8. Influence of density on calibration curve.
Variations in dry-soil bulk density affect the calibration curve of a neutron-moisture gauge. Higher densities give higher count-rates for the same moisture content. (21)

It was mentioned at the beginning of this section that one of the disadvantages of neutron gauges was that a subsurface probe can not be used near the soil surface. This is caused by the lack of resolution of all scattering neutron probes, which is related to the sphere of influence. The radius of the sphere of influence is given by

$$R_C = r_{H_2O} \left(\frac{100}{\text{vol } \% H_2O} \right)^{1/3} \quad (7)$$

where

$$r_{H_2O} = \text{radius of the sphere of influence in water}$$

De Boever and De Boodt⁽²³⁾ used cast nylon as an auxiliary moderator and cadmium to shield the detector in two separate experiments in an effort to reduce the sphere of influence for moisture contents by volume varying from 11.0% to 53% and 11.95% to 100%, respectively. In measurements where cast nylon was used as an auxiliary moderator a reduction of 10% in R_C was observed when the cadmium was used to shield the scintillation from thermal neutrons, but again as in the cast nylon case there was a corresponding loss of sensitivity.

A calibration measurement by Van Bavel and Stirck⁽²⁴⁾ at a 10 cm depth with a ^{241}Am -Be source depth probe, when compared with results using ^{226}Ra -Be, suggested that the lower average source energy of neutrons from an Am-Be versus a ^{226}Ra -Be source resulted in better resolution in a neutron measurement. This is an obvious fact because the radius of the sphere of influence is a function of the migration length of the fast neutrons. That is, R_C is a function of M , where M is defined as

$$M = \sqrt{L_T^2 + \tau_T^2} \quad (8)$$

where

L_T^2 = Diffusion length for thermal neutrons

τ_T^2 = Net vector distance traveled by a fast neutron to it's attainment of thermal energy.

Thus sources such as $^{241}\text{Am-Be}$ or ^{252}Cf should provide increased resolution because of their lower average neutron energies. In the former the disadvantage is its lower neutron output and the prohibitive cost of the latter requires that detail investigation of the actual gains be made before these sources can be recommended. Table IV gives some of the applications of neutrons in soil moisture studies.

In a comparative study entitled, Commercial Portable Gauges for Radiometric Determination of the Density and Moisture Content of Building Materials; ⁽²⁵⁾ sponsored by the IAEA, a series of tests on a number of commercial portable neutron moisture gauges were conducted to determine: The relationships between certain gauge parameters and characteristics; suitability of gauges for certain uses and techniques and procedures of calibration. Gauges were requested from 22 companies representing several countries. Eleven manufacturers participated in the study. The types of neutron gauges are given in table V. These tests were conducted during the year 1968 and from the number probes utilizing $^{241}\text{Am-Be}$ sources the increased use of $^{241}\text{Am-Be}$ fast neutron sources is evident.

Most of the moisture meters used in the U.S. are supplied by Nuclear Chicago* or Troxler Electronic Laboratories**. A report of a survey conducted by the U.S. Work Group on Nuclear Techniques in Unsaturated and Saturated Zones. The types and strength of fast neutron sources (cont. on p.116)

*Nuclear-Chicago Corp., a subsidiary of G.D. Searle & Co. 333 E. Howard Ave., Des Plaines, Ill. 60018.

**Troxler Electronic Lab. Inc, P.O. Box 5997, Raleigh, N.C. 27607.

SOURCES		TYPE AND PURPOSE OF THE MEASUREMENT	PROBE AND TYPE DETECTOR	REFERENCE AND YEAR
²⁴¹ Am-Be (100 mCi)		Measurements made on silt loam soil at different dry bulk densities, the effect of slow neutron absorbers in the soil and long term stability in the electronics.	End Placement (USDA Sed. Lab.)	McHenry and Gill (21) (1967)
²⁴¹ Am-Be		Determination of water movement in soils with a view toward estimating meters of soils likely to be contaminated by the spilling radioactive liquids		Rancon (26) (1967)
30 mCi ³⁴¹ Am-Be 3 mCi ¹³⁷ Cs		Subsurface simultaneous moisture density measurement. The effect of soil parameters changes on the linearity of the calibration curve	Subsurfaces-Probe ⁶ Li glass scintillation	Jensen and Somer (22) (1967)
²⁴¹ Am-Be and ¹³⁷ Cs		Discussion of an instrument the densitratest compaction-which consist of a double sealed source Transmission method.	Detectors selectively sensitive to gamma rays and high energy neutrons.	Wack et al (30) (1970)
²⁴¹ Am-Be 30 mCi		Moisture measurement on laboratory samples of sand. Sensitivity of the surface gauge and subsurface gauge were	Subsurface - Probe Detection BF ₃ Surface - Probe	Aditya and Batra (31) (1969)

SOURCES	TYPE AND PURPOSE OF THE MEASUREMENT	PROBE AND TYPE DETECTOR	REFERENCE AND YEAR
$^{241}\text{Am}-^9\text{Be}$ (200 mCi)		End Placement probe with a BF_3 - detector in an aluminum case, 3.4 cm and 17 cm long.	Long and French ⁽²⁹⁾ (1967)
$^{241}\text{Am}-^9\text{Be}$	Fractional water content by volume measured on two days on an adelanto clay loam profile both have and ryegrass cover	End placement Troxler N 104 Probe fitted with an $^{241}\text{Am}-\text{Be}$ source	Van Bauel and Stirk ⁽²⁴⁾ (1967)
$^{241}\text{Am}-\text{Be}$ (50 mCi) Ra-Be	Comparison of theoretical and experimental data; a method of representation by which it is possible to assess the ability of a three group model in a true situation	End placement-Probe with Lithium-Glass scintillation	Cauchat ⁽²⁷⁾ (1967)
$^{241}\text{Am}-\text{Be}$ (300 mCi)	Examine the possibility of three flux intensity measurement obtained by gamma diffusion. The flux intensity meas- urements were of a) epithermal near source, and b) two thermal at the source and approx. 10 cm away from the source.	Thermal neutron det- ector BF_3 and a ^3He detector covered with Cadmium and 2mm of d'endium, pour to detection of the ep- thermal neutrons	Corompt and Calmels ⁽²⁸⁾ (1967)

SOURCES		TYPE AND PURPOSE OF THE MEASUREMENT	PROBE AND TYPE DETECTOR	REFERENCE YEAR
$^{241}\text{Am-Be}$		Road Logger-Instrument capable of producing a continuous record of density and moisture of an area 15" wide, 6' long and 8" deep.	surface - Probe Detection - BF_3	Unknown In Pennsylvania Highway Department Report
$^{241}\text{Am-Be}$ $2 \times 10^5 -$ 2×10^6 n/sec		Design and Calibration Ocean nuclear probe for sediment water content determination. Theoretical model (7.10 groups) used to check calibration	Depth - Probe Cd-covered LiI detector	Ringle Bell and Hurley (22) (1970)
Uses fast neutron sources such as $^{241}\text{Am-Be}$		Patent: Description and uses of portable apparatus-Troxler electronic labs		Lowery and Clements (1970) (Patent United States of America)

SOURCES	TYPE AND PURPOSE OF THE MEASUREMENT	PROBE AND TYPE DETECTOR	REFERENCE AND YEAR
$^{241}\text{Am}-\text{Be}$ (100 mCi)	Test conducted to determine the influence of a cast nylon housing surrounding the source (in one experiment) and that of a thin cadmium foil covering the counter on the resolution of a neutron moisture meter	Depth(end placement) probe ^6Li -glass-scintillation	De Boever and De Boodt ⁽²³⁾ (1965)

Table IV Applications of $^{241}\text{Am}-\text{Be}$ in soil moisture studies.

SOURCE	TYPE OF PROBE	TOTAL NUMBER OF SOURCES	NUMBER AND STRENGTH OF SOURCES
²⁴¹ Am-Be	Surface	3	3-30 mCi
	Subsurface	8	4-30 mCi 3-100 mCi 1-10 mCi
²²⁶ Ra-Be	Surface	4	2-2 mCi 1-4 mCi 1-3 mCi
	Subsurface	3	2-10 mCi 1-5 mCi
²³⁹ Pu-Be	Surface	1	10 ⁵ neutrons/sec
	Subsurface	1	10 ⁵ neutrons/sec

Table V. Types of neutron gauges and sources used in the IAEA Commercial neutron moisture gauges (1968).

used in these probes are given in table VI.

TYPE OF NEUTRON SOURCE	TOTAL NUMBER OF PROBES USING SOURCE	RANGES OF THE STRENGTHS OF SOURCES USED
$^{241}\text{Am-Be}$	56	39-100 mCi 8-80 mCi 4-50 mCi The remaining 5 sources ranged in strengths from as low as 5 mCi up to 200 mCi.
$^{226}\text{Ra-Be}$	146	10-2 mCi The remaining sources, with the exception of a few in the 8-12 mCi class were in the 3-5 mCi class.
$^{239}\text{Pu-Be}$	3	5, 1 and .125 curies

Table VI. Some results of a survey conducted on the use of neutron meters in soil-moisture measurements prior to august 1, 1967. ⁽⁴⁾

Summary

The ^{241}Am -Be neutron source has become the most widely used sealed neutron source in soil moisture studies since the mid nineteen sixties. ^{241}Am -Be has the obvious advantage of a low gamma/neutron yield ratio.

The lower average source energy of ^{241}Am -Be versus those emitted by ^{226}Ra -Be results in better resolution near the soil surface. This source however must always be used in conjunction with a gamma source in those situations where the bulk density and moisture content of the soil must be determined simultaneously.

REFERENCES

1. K. Ansell and E.G. Hall, "High Intensity (α , n) Sources," In Neutrons Sources and Applications, Conf. 710402, P. I-100, (1971).
2. W. Szulmayer, "Humidity and Moisture Measurement," CSIRO Fd. Presero, Q., V 29, 2, (1969).
3. W.R. Gardner, "Neutron Meter Use in Soil-Moisture Measurements," (A Survey) Isotopes and Radiation Technology, V. 7, 3 (1970).
4. L.F. Ballard and R.P. Gardner, "Density and Moisture Content Measurements by Nuclear Methods," Res. Prog. Rep. Res. Triangle Inst., Durham, N.C. 14, (1965).
5. C.G. Clayton, "Nucleonic Soil Density and Moisture Gauges," Nuclear Techniques and Minearal Resources, IAEA, Vienna (1969).
6. Reactor Physics Constants, ANL-5800 (1967).
7. R. Lamarsh, Nuclear Reactor Theory,
8. J.W. Holmes, "Calibration and Field use of the Neutron Scattering Method of Measuring Soil Water Content," Aus. J. Appl. Sci., 7 (1956).
9. W. Gardner, D. Kirkham, "Determination of Soil Moisture by Neutron Scattering," Soil Sci., 73, PP 391-401, (1952).
10. R.A. Semmler, "Neutron Moderation Moisture Meters. Analysis of Application to Coal and Soil," Final Rep. C00-712-73 (1963).
11. P.L. ØLgaard, V. Kaakr, "Comparatice Experimental and Theoretical Investigation of the DM Neutron Moisture Probe," Nucl. Engr. Design, 5, PP 311-24 (1967).
12. P. Couchat, "La Methode Neutronique de Mesure de L'humidite' des Sols," Rep. CEA-R 3298 (1967).
13. S. Kasi and H. Koskinen, "Analysis, Calculations and Measurements concerning the Moisture Measuring by the Neutron Method," Nucl Engr Design, 3, PP 74-82, (1966).
14. R.P. Gardner and K.F. Roberts, "Density and Moisture Content Measurements by Nuclear Methods," Research Triangle Institute, North Carolina (1966).
15. W. Gemmell, B. McGregor and G.F. Moss, "Estimation of Moisture Content by Neutron Scattering: Theory, Calculation and Experiment," Int. J. Appl. Radiat. Isotopes, 17, PP 615-20, (1966).

16. J.C. Corey, A.R. Boulogne and J.H. Horton, "Determination of Soil Density and Water Content by Fast Neutrons and Gamma Rays," Water Resources Res., V 6, 1, PP 223-29, (1970).
17. Neutron Moisture Gauges, Tech Rep. Series No. 112, IAEA, Vienna (1970).
18. C.N. Wright, "Protection for Safe Handling of ^{252}Cf Sources," Health Phys., 15(5), PP 466-69, (1968).
19. A'. Z. Nagy and T. Ra'zga, "Radioisotopic Combined Moisture-density Meter," J. Sci. Instrum., V 43, (1966).
20. J.R. Mchenry and A. Gill, "The Influence of Bulk Density, Slow Neutron Absorbers, and Time on the Calibration of Neutrons-Moisture Probes," In Isotope and Radiation Techniques In Soil Physics and Irrigation Studies, Proc. of a Symp., Istanbul, June (1967).
21. P.A. Jensen and E. Somer, "Scintillation Techniques In Soil-Moisture and Density Measurements" In Isotope and Radiation Techniques in Soil Physics and Irrigation Studies, Proc. of a Symp. Istanbul, June (1967).
22. J.C. Ringle, J.R. Bell and J.L. Hurley, "Design and Calibration of Deep Ocean Nuclear Probe for Sediment Water Content." Report No. CR 70;016 (1970).
23. J. DeBoever and M. De Boodt, "The Application of Cadmium and Cast Nylon in the Measurement of Soil Moisture with a Neutron Moisture Meter," In Second International Symposium, Application of Isotope Techniques in Agronomy Research (1965).
24. C.H.M. Van Bavel and G.B. Stirk, "Soil Water Measurement with an ^{241}Am -Be Neutron Source and an Application to Evaporimetry," J. of Hydrology, 5, PP 40-46 (1967).
25. A. Honig et al (Eds.), Commercial Portable Gauges for Radiometric Determination of the Density and Moisture Content of Building Materials, Technical Rep. Series No. 130; IAWA, Vienna (1971).
26. D. Rancon, "Evolution de l' humidite' d'un Sol Soumis a' un apport. continu d'eau Sur une pet'te Surface" Symposium on the Use of Isotopes and Radiation Techniques In Soil Physics and Irrigation Studies, Istanbul, June (1967).
27. P. Couchat, "De'termination de la courbe de'etalonnage de l'humidimetre a neutrons a partir de l'analyse chimique des sols," Symposium on the Use of Isotopes and Radiation Techniques In Soil Physics and Irrigation Studies, Istanbul, June (1967).

28. E.P. Corompt and P. Calmels, "Exploritation des informations continues dans un flux de neutrons thermalises et possibilities d'applications a la mesure de l'humidite' et de la densite' d'un Sol," Symposium on the Use of Isotopes and Radiation Techniques In Soil Physics and Irrigation Studies, Istanbul, June (1967).
29. I.F. Long and B.K. French, "Measurement of Soil Moisture in the field by Neutron Moderation," J. of Soil Sci. V 18, 1, P. 49 (1967).
30. B. Wack et al, "Non-destructive Measurements of density and Moisture by Direct Transmission of fast Neutrons and Gamma Rays,-Application to Soil Testing," Abstract In Eurisotop No 51, Part 1, August (1970).
31. P.K. Aditya and A.K. Batra, "Moisture Measurement by Neutron Moderation," Indian Journal of Pure and Applied Physics, V 7, 5, P 323 (1969).

2. Am-241-Be in Neutron Logging Techniques

1. General discussion of the principles of neutron well logging.

For almost 32 years neutron logging has been used in ore, coal, oil and in general mining geophysics. Recently the neutron logging tool has been suggested as a means of studying Ground-Water Hydrology. Several detailed review papers on Nuclear techniques in the oil industry^{(1), (2), (3), (4)} and geophysics⁽⁵⁾ discusses along with other methods (such as gamma logging) neutron logging techniques. The theory of neutron-logging methods is based on the penetration of neutrons in rock media. Rock media can be categorized as wet (with strong (ore geophysics) and weak (oil geophysics)) or dry (with strong and weak). The various neutron methods of logging can be delineated as given in figure 1. In figure 1 those methods of logging that are not applicable to sealed neutron sources are not presented.

The physics of neutron logging methods are similar to those encountered in nuclear reactor theory or in soil moisture studies. The three group methods as well as the Fermi age approximation are used to calculate calibration curves. Also as in soil studies the slowing down length, L_f , the thermal diffusion length, L_T , and the diffusion coefficient D , for thermal neutrons are required.

Of the nuclei present, hydrogen has the greatest influence on the behavior of neutrons. Most hydrogen is present in either water, gas or oil, in the void spaces of the rock medium. Since neutrons are sensitive to the amount of hydrogen in a medium, neutron logs measure the amount of water (hydrogen) present in the unsaturated zone and the amount of porosity of the medium in the saturated zone. Of the methods of neutron logs given in Figure 1, the epithermal neutron log technique has the advantage of being less sensitive to the penetrating effects of thermal neutron absorption in the rock matrix and

Figure 1. Neutron Borehole logging techniques that can be conducted using sealed isotopic neutron sources (e.g. ^{239}Pu -Be, ^{226}Ra -Be, ^{241}Am -Be and ^{252}Cf and others).

and salts present in the water in a borehole. Use is made of epithermal neutrons in sidewall neutron porosity measurements. The epithermal neutron detector (e.g. High pressure - helium - 3 counter) and a neutron source, such as $^{239}\text{Pu-Be}$ or $^{241}\text{Am-Be}$, are mounted in a slide and is pressed firmly against the wall of an uncased hole. The detector is shielded such that it has directional sensitivity and it responds primarily to neutrons coming from the formation rather than the fluid in the borehole.

The distance between source and detector affects the quantitative results of the log. Experiments ⁽⁶⁾ indicated that by increasing both source strength and source-detector spacing the radius of investigation is increased. The radius of investigation is particularly useful for hydrologic investigations because it eliminates some of the present limitations to the method caused by irregularities of borehole diameters, cement and gravel packs. A typical neutron logging system is shown in Figure 2.

For cased holes, large source detector separations are used, which requires sources of strengths of approximately 10^8 neutrons/sec. The large separation between the detectors and source is chosen because, on a relative basis, the liquid in the borehole is a stronger attenuator of neutron (thermal and epithermal) than the formation. Hence the sensitivity of the tool to the formation is heightened giving a natural directional sensitivity. Theoretical and experimented studies ^{(7) (8)} have shown that the ratio of the thermal fluxes at two large distances along the borehole axis is essentially identical to the epithermal thermal ratio and is a measure of the slowing down length of the formation. The slowing down of neutrons is strongly dependent on the hydrogen content of the formation and thus is an indication of the porosity of the medium. In cases where the lithology is not known a second porosity tool is

LEGEND FOR FIGURE 2

1. Surface Electronics
2. Pre amp
3. Photomultiplier Tube
4. Scintillator
5. Neutron Shield (hydrogenous)
6. Neutron Source (e.g. $^{241}\text{Am}-\text{Be}$)
7. Steel Casing
8. Borehole (uncased)
9. Earth Formations

Figure 2 -- Typical neutron logging system.

used and a cross-plot of the data from the two logs is used to determine it. This porosity information is used to properly scale the neutron log. To recognize gas deposits two logs, (e.g., a neutron and formation density and a acoustic velocity) are used. The effect of the presence of gas on these tools is to cause the apparent porosity to be low in the neutron case and too high in the formation density and acoustic velocity cases. The separation between the neutron log and either of the other two can be used to qualitatively identify the presence of gas.

Neutron - well logging is routinely used in the petroleum industry. According to Caldwell⁽⁹⁾ in 1970 more than 30,000 new wells were drilled by the oil industry and in every case either a gamma or neutron log or both were run in these wells. This does not include the nuclear logs conducted on old wells.

The requirements of good statistical accuracy and fast logging speeds and the advent of ^{252}Cf high intensity neutron sources ($\approx 10^9$ n/sec) have stimulated the development of neutron sources such as $^{227}\text{Ac-Be}$, $^{238}\text{Pu-Be}$ and $^{241}\text{Am-Be}$ of $\approx 10^8$ n/sec all of which have much longer half lives than ^{252}Cf and have very competitive prices.

Steady-state neutron sources can not compete with neutron generators where neutron-gamma inelastic logs are required, e.g., in detection of carbon or other elements. This results from the fact that energy of neutrons needed to excite the energy levels in the nucleus exceeds the average neutron energies of these sources. Nor can they be used in neutron life-time logging applications. However, a new type of mechanically activated pulsed neutron source comprising an alpha source (e.g. ^{241}Am) and a beryllium target separated

rotating shutter was recently patented by Cladwell and Gwens⁽⁹⁾. The rotating shutter allows bursts of alphas to pass and when they impinge on the beryllium target they create a pulse of neutrons.

2. Experimental results of measurements.

Unfortunately there is a scarcity of technical papers concerning the use of ^{241}Am -Be in actual field logging and laboratory situations. The source is, however, one of the most frequently used steady state sources in systems designed to determine porosity and salinity content by measuring thermal neutrons on capture gamma rays. The various service companies have published much literature on the design, calibration and performance of their respective tools. In most cases they suffer from being sensitive to changes in borehole parameters and from being unable to separate the responses to porosity and salinity.

The sidewall neutron porosity system reduces most of the environmental effects that have complicated neutron logging techniques. Figure 3 shows⁽¹⁰⁾ a SNP log made in carbonate rock, together with a gamma ray, caliper and density log. The neutron porosity log shows the results of two separate runs with and without caliper compensation for variations in the borehole diameter, as the solid and dotted curves, respectively. A comparison between neutron logs obtained using a 3 curie ^{241}Am -Be (8.6×10^6) and a 50 millicuries ^{252}Cf (2.1×10^8) source conducted by Keys and Boulogne⁽¹¹⁾ showed that the greater neutron field of the ^{252}Cf -source made the log with this source: more sensitive to the lithology, less sensitive to borehole diameter variations, and less noisy. A set of neutron logs made in a cased well in which the

Figure 3 -- SNP Log in evaporite section. Ref. 10.

Figure 4. A comparison of three neutron logs made with different sources in a cased section of a well. Reference 11.

results obtained with the two sources given above and the log made by a commercial company in the same well in which a 5 Ci ^{239}Pu -Be (8×10^6 n/sec) source was used is shown in Figure 4. Figure 4 shows the effect of the source intensity on the log, the ^{252}Cf -log is qualitatively superior to that of the ^{241}Am -Be source. Because of its relatively short half life, ^{252}Cf , may not possess clear superiority (with all things considered) to ^{241}Am -Be as a source for use in conventional porosity and moisture logging. There are however, other advantages of ^{252}Cf as a neutron source in neutron logging. But these will not be discussed here. Table I gives a comparison of neutron sources used in well logging. As a source that does not need periodic decay correction, ^{241}Am is definitely an attractive source of neutrons.

	<u>HALF LIFE</u>	<u>MEAN ENERGY (MeV)</u>	<u>YIELD NPS</u>	<u>APPROX. COST (\$) 10⁶ NPS</u>
²³⁹ Pu-Be	24,360 y	4.5	1.7 x 10 ⁷	91
²⁴¹ Am-Be	458 y	~4	10 ⁷	250
			10 ⁸	200
²²⁷ Ac-Be	22 y	~4	10 ⁸	40
²¹⁰ Po-Be	138 d	4.3	2.5 x 10 ⁸	21
²⁵² Cf	2.65 y	2.3	10 ⁸	4.3
			10 ⁹	4.3

TABLE I. Comparison of Neutron Sources for Well Logging⁽¹²⁾.

REFERENCES

1. G.L. Meyer, Application of Neutron Sources to Ground Water Hydrology, In Neutron Sources and Applications, Conf. - 710402, Vol I, (1971).
2. F.E. Senftle, Nuclear Techniques for Mineral Exploration, In, Neutron Sources and Applications, Conf. - 710402, Vol. I, (1971).
3. R.L. Caldwell, Use of Neutrons for Petroleum and Gas Exploration, In, Neutron Sources and Applications, Conf. - 710402, Vol. I, (1971).
4. G. Clayton, "A Survey of the Application of Radiation Techniques in Oil and Mineral Bore holes," AERE-R 5368 (1967).
5. Threadgold, P., "Application of Well Logging Techniques to Mining Exploration Boreholes," Inst. of Mining and Metallurgy, London; 17, PP 16, (1969).
6. Tittman, J., Sherman, H., Nugel, W.A., Alger, R.P., "The Sidewall Epithermal Neutron Porosity Log, J. Petrol Technol 18, P. 1351, (1966).
7. L.S. Allen, C.W. Tittle, W.R. Mills, R.L. Caldwell, "Dual-Spaced Neutron Logging for Porosity," Geophysics, 32 (1) PP 60-68, (1967).
8. H.S. Parp, H.D. Scott, "The Use of Cf-252 as a Neutron Source for Well Logging," Sym. on App. of Radioisotopes Am. Inst. Chem Engrs. 63rd Annual Mtg., Chicago (1970).
9. R.L. Caldwell and W.W. Gwens, US Patent No. 3,389,257 Pulsed Neutron Source Comprising a Plurality of Alpha sources and associated targets with a rotatable shutter there-between (June 1968).
10. R.L. Caldwell, "Nuclear Logging Methods," Radioisotopes, (Tokyo), 17(4): PP 171-85, (1968).
11. W.S. Keys and A.R. Boulogne, Transactions of the SPWLA 10th Annual Logging Symposium, Houston, (1969).
12. H.J. Parp, and H.D. Scott, "The Use of ²⁵²Cf as a Neutron for Well Logging," Conf. - 710402, Vol. III, April 19-21 (1971).

B. Neutron Activation Using the Radioisotopic Neutron Source ^{241}Am .

Although sealed radioisotopic - neutron sources has been used and will continue to be applied in activation analysis in a number of laboratories through out the world, their relatively low intensities have placed severe limitations on their applicability to many problems in the field of neutron activation analysis. Up until very recently before the advent of $^{241}\text{Am} - ^{242}\text{Cm}-\text{Be}$ (ABC), ^{252}Cf , $^{243}\text{Cm}-\text{Be}$, $^{238}\text{Pu}-\text{Be}$, and $^{124}\text{Sb}-\text{Be}$ neutron sources, the typical radioisotope neutron source had a yield of $\sim 10^6$ neutrons/sec. The ABC, $^{210}\text{Po}-\text{Be}$, $^{228}\text{Th}-\text{Be}$ and $^{244}\text{Cm}-\text{Be}$ (1) (2) sources can be obtained with yields of 10^8 neutrons/seconds. More recently, californium-252, a man made radioisotope, with a spontaneous neutron emission of $\sim 10^6$ neutrons/second/mg has become available (at $\sim \$10.00/\mu\text{g}^{(3)}$).

All of the sources mentioned above are fast neutron emitters with the most probable energy of neutrons being 4 MeV for $^{241}\text{Am}-^{242}\text{Cm}-\text{Be}$ and $^{244}\text{Cm}-\text{Be}$ and approximately 2 MeV for ^{252}Cf .

1. Principles of Radioisotopic Activation

The Americium-Beryllium neutron source (as is most neutron sources) emits neutrons that are predominately fast with a significant thermal neutron yield. Fast neutrons interact with materials through: inelastic scattering reaction ($n, n'\gamma$) radiative capture (n, γ), radioactive capture, and particle reactions ($n, 2n$), (n, α) and (n, p). The inelastic scattering reaction is a dominate mode of interaction for neutron with energies above 2 MeV with elements with atomic weights less than ≈ 70 amu's. Particle reactions occur at high neutron energies and are essentially threshold reaction.

Thermal neutrons interact at a significant rate with materials through the radiative capture process. Many trace and base metallic elements (e.g. Ni, Fe, Cu, Ti, Mn, Cr and Pb) have distinct transition energies and can be identified via this process. In this process gamma-rays emitted in the decay of the compound nuclei formed in (n, γ) reactions have energies largely below 3 MeV. The radioactive capture reaction is also significant for thermal neutrons. This process allows one to measure half lives and the characteristic radiations of relatively short half-lived (< 10 min) isotopes which provide a powerful confirmatory identification test.

The procedure for determination of an element by activation analysis can be described in five basic steps, as illustrated in Fig. 1.

Fig. 1. Block diagram of Activation Analysis Procedure.

The advantage of activation analysis lies mainly in its extreme sensitivity. It is in some cases more sensitive than emission spectrography. Also, it is the only analytical method for determination of trace elements which is not affected by errors due to blank values. Lastly, the possibilities of non - destructive determination are much greater than with any other method. The major disadvantage of this method is that some elements cannot be determined practically by the neutron - activation technique.

The detection limit of activation analysis depends on the fact that a certain minimum activity is needed for sureness of detection. The activity of a sample after irradiation can be expressed as:

$$A = \phi \sigma N t \left(\frac{.693}{t_{1/2}} \right) \quad (1)$$

for

$$t_{1/2} > t$$

$$A = \phi \sigma N (1 - e^{-.693(t/t_{1/2})}) e^{-.693(t'/t_{1/2})} \quad (2)$$

In Eq. (1) and Eq. (2), ϕ , σ , N , t , $t_{1/2}$ and t' are the neutron flux, microscopic - cross section, number of atoms of the element in the sample, irradiation time, half-life of the radioactive product and cooling time, respectively. For saturation activity, the detection limit, as expressed in grams, is

$$N_{min} = 5.10^{-24} M/\phi \sigma \quad (3)$$

where

$$M = \text{the atomic weight of the element}$$

If we consider the case of ^{64}Cu ($\sigma \sim 2$ barns) and the typical thermal neutron flux from an ^{241}Am -Be source of $\sim 10^5$ N/cm²/sec, the detection limit in grams for ^{64}Cu is found to be $\sim 1.6 \times 10^{-3}$ grams.

2. Applications of the ^{241}Am -Be Neutron Source

Stewart⁽⁴⁾ used a 3-Curie ^{241}Am -Be source to illustrate the technical feasibility of measuring carbon in fly ash by the inelastic scattering of the neutrons from the source while passing through moving material on a conveyor belt. The results of one of their test on 20 and 198 pound samples of iron ore sinter is shown in Fig. 2. They estimate that based on a tonnage flow of ash of 10 to 100 tph through a 40 inch diameter vessel and using a 30-curie neutron source in conjunction with a 5 x 5 inch Crystal detector, a $\pm 0.5\%$ change in ^{12}C should be detectable in 70 seconds and a $\pm 0.2\%$ change in 3 minutes.

The prompt (n, γ) reaction offers one the opportunity to make rapid and non-destructive measurements. This method has been applied in a wide range of fields and processes. Tiwari⁽⁵⁾ utilized the (n, γ) technique as a means of determining the protein in grain samples. The "prompt" γ -rays produced by neutron capture have been employed in non destructive analysis of boron in glass.⁽⁶⁾ A 300 mCi ^{241}Am -Be source was used in this study. In a recent experiment by Tiwari⁽⁷⁾ et al, he observed the prompt gammas from the capture reaction of neutrons from an ^{241}Am -Be source to determine the nitrogen content of organic materials.

In some applications the delayed γ -rays from a radioactive product contains the information desired. One such application is in the use of neutron activation to determine certain elements in a slurry. Starnes⁽⁸⁾, used 3 Ci of ^{241}Am -Be along with a NaI(Tl) crystal to determine the amount

Figure 2. Nuclear Measurement of Carbon in Iron Ore Sinter Mix.
(Reference 4).

of fluorine and barium in a small recirculating loop of barytes and fluoirt. The fluorine and barium were activated by the $^{19}\text{F}(\text{n}, \alpha) ^{16}\text{N}$ and $^{137}\text{Ba}(\text{n}, \text{n}') ^{137\text{m}}\text{Ba}$, respectively.

Small ^{241}Am -Be neutron sources in the 10^6 n/sec class can only be employed to activate high cross-sectional elements when the pollutant is present in a few parts per million. An early review of the applications of ^{241}Am -Be neutrons by Strain and Leddicotte⁽⁹⁾ reported certain elements could be detected in parts per million after a few minutes of exposure to a 10^4 neutron flux. Small ^{241}Am -Be sources have been evaluated along ^{239}Pu -Be and ^{252}Cf as possible neutron sources for use in measuring sulfur in coal and oil⁽¹⁰⁾ through the n, γ reaction.

The utilization of neutron activation with ^{241}Am -Be and ^{252}Cf sources for process control analysis in the primary metal, stone, clay and glass has been examined by Kuusi⁽¹¹⁾ with plant scale comparisons. Neutron activation analysis using a radioisotopic sources has considerable potential for industrial application to process and quality control. Kuusi⁽¹²⁾ (See Fig. 3) described a silicon, chromium and aluminum concentration analyzer⁽¹³⁾ which employed a 5 Ci ^{241}Am -Be source with an output of 1.25×10^7 n/sec. The silicon concentration is determined by counting the number of counts accumulated under the 1.78 MeV photopeak of the spectrum of ^{28}Al (^{28}Al (2.3 min) is produced via the $^{28}\text{Si}(\text{n}, \text{p}) ^{28}\text{Al}$ reaction), Likewise chromium and aluminum concentrations are determined by counting the 1.43 MeV gammas from ^{52}V ($^{52}\text{Cr}(\text{n}, \text{p}) ^{52}\text{V}$) and the .85 and 1.01 MeV gammas from ^{27}Mg ($^{28}\text{Al}(\text{n}, \text{p}) ^{27}\text{Mg}$), respectively. Calibration curves obtained by Kuusi for determining the amount of ^{52}Cr and ^{28}Al for a time of 10 min are given in figure 4 . For a sample weight of 980 g. the statistical error of counting, including background statistics corresponds to $\pm .09$ wt % SiO_2 when the SiO_2 content

Figure 3. Schematic diagram of the construction of the analyzer and the scheme of the analysis. (Reference 12).

Figure 4. Calibration curves for the determination of (a) chromium and (b) silicon in chrome-concentrate samples. Activation time 10 min, transfer time 0.5 min, counting time 10 min. Sample weight varied from 900 to 1100 g. (Reference 12).

is 5 wt.%. Similarly, the statistical error of counting for Cr_2O_3 and Al_2O_3 corresponds to $\pm 0.5\%$ Cr_2O_3 when the Cr_2O_3 content is 40% and $\pm 0.13\%$ Al_2O_3 when the Al_2O_3 content is 14%. As much larger neutron sources are employed the irradiation time and counting time can be reduced. Wood⁽¹⁴⁾ reported on the use of a 33 Ci ^{241}Am -Be source in a iron foundry to determine silicon concentration in freshly cast iron. Irradiation and counting times were 3-and-2-minutes, respectively.

Boddy and Gloros⁽¹⁵⁾ recently reported on the measurement of phosphorus in human bone using radioisotopic neutron sources. The neutron source used was a one Ci ^{241}Am -Be source of 2.2×10^6 n/sec yield. Measurements were made sequentially on the lower leg of a human skeleton. The source was positioned with its geometrical centre at 2 cm from the ankle (lateral malleolus) for an irradiation time of 3 min. The induced radioactivity was measured by a NaI (Tl) crystal.

These measurements were done only to test feasibility of measuring phosphorus via the $^{31}\text{P}(n, \alpha)^{28}\text{Al}$, in vivo. The ^{241}Am -Be source employed was sub-optimal but it did enable the validity of the technique to be examined with respect to reproducibility, geometries and counting statistics. The coefficient of variation due to counting statistics ranged from 7.8 - 10.5 % for skeleton ankles and was 8.7% for skeleton hand. The monitoring and irradiation geometries for this study are shown in figure 5 , along with recommended irradiation and monitoring geometries in Fig. 6 .

Two Instruments for Activation Analysis of Solutions by a dialution method has been described by Louwrier, et al⁽¹⁶⁾ for determination of the concentration of a main component in an aqueous solution. One method activates eight solutions at the same time. A diagram of the apparatus is shown in figure 7 . The eight vessels containing the solutions to be analyzed are placed on a small rotating table. The positions of the vessels are identical relative to the axis of the turntable. The source, a ^{239}Pu -Be or ^{241}Am -Be neutron source is placed in a lucite plug filled with paraffin at the center of the table.

Figure 5 . Irradiation and monitoring geometries proposed for in vivo activation analysis in man using radioactive neutron sources. (Reference 15).

Figure 6 . Irradiation and monitoring geometries used to demonstrate the feasibility of measuring phosphorus, in parts of a human skeleton, by a method suitable for in vivo activation analysis using $^{241}\text{Am-Be}$ neutron sources. (Reference 15).

The second method utilizes a large cylinder or sphere containing the liquid to be activated in which the neutron source can be placed in the centre where the counters are placed after the neutron source is removed. In this case instead of activating unknown and known sample at the same time the known and unknown samples can be irradiated separately. For an ^{241}Am -Be source vessel of 20 cm in radius is sufficient. The equations governing the resolution of the experimental data obtained by these two methods is sufficiently different from the normal activation equations. They will be summarized here. The activity induced in the pure standard and impure solutions are:⁽¹⁶⁾

$$A_1 = K \cdot N_{C_1} (1 - \alpha_C N_{C_1}) \quad (1)$$

$$A_2 = K \cdot N_{C_2} (1 - \alpha_C N_{C_2} - \alpha_i N_{i_2})$$

and the corresponding counts are given by

$$R_1 = A_1 e_o (1 - \epsilon N_{C_1}) \quad (2)$$

$$R_2 = A_2 e_o (1 - \epsilon N_{C_2} - \epsilon_i N_{i_2})$$

where

$K =$ a constant

$N_{C_1} =$ concentration of known element in pure standard

$N_{C_2} =$ concentration of unknown element in impure sample

$\alpha_C =$ flux depression proportionality constant for element to be activated

$\alpha_i =$ flux depression proportionality constant for element to be activated for impurities.

ϵN_{C_2} and ϵN_{C_1} = relative reduction in efficiency by dissolved material to be determined

$\epsilon_i N_{i_2}$ = reduction in efficiency by impurities

e_o = counting efficiency for activity in the presence of pure water

To determine N_{C_2} for the above equations one must know more than the values of R_1 and R_2 . By diluting the original inactive solutions of both the standard and the unknown with pure water or sulphuric acid to a concentration per volume which is a fraction of the original one, a series of count rates R' for various f values can be obtained. The R'/F values are plotted as a function of F both for the impure solution and the standard and the extrapolate to $F = 0$. The ratio of these extrapolated values of R'/F at $f = 0$ is equal to the ratio of N_{C_1} and N_{C_2} .

In the second method, the activity A' of a solution containing N_C atoms/cm³ obtained after diluting with pure water to a fraction f of the original concentration is

$$A' = k \frac{f N_C \sigma_C}{(f N_C (\sigma_a + \sigma_s) + N_H \sigma_H + r(1-f) N_C \sigma_C + f \sum N_i \sigma_i + (1-f) \sum \rho N_i \sigma_i (1-q'))}, (3)$$

where

k $\equiv$ proportionality constant of activation

N_C $\equiv$ atom/cm³ of the element to be determined

σ_C and σ_a $\equiv$ activation and absorption cross-section of unknown

N_H $\equiv$ atom/cm³ of hydrogen present

σ_H $\equiv$ absorption cross-section of hydrogen.

- σ_i $\equiv$ absorption cross-section of impurities
 σ_s $\equiv$ absorption cross-section of the elements stoichiometrically associated with the element to be determined.
 ΣN_i $\equiv$ sum of atoms/cm³ of impurities
 r $\equiv$ number of hydrogen atoms entering the solution to replace one atom of the material to activated when the latter is removed. ρ is the corresponding number for impurities.
 q' $\equiv$ fraction of neutrons escaping the vessel.

The count rate for different dilutions of each solution are given by the equation

$$R' = \epsilon' A' \quad (4)$$

Substituting Eq. (4) into Eq. (3) and extrapolating to $f=0$ one obtains the ratio f_0/R_0 as

$$f_0/R_0 = \frac{1}{k\epsilon'} \frac{N_H\sigma_H + rN_C\sigma_c + \Sigma\rho N_i\sigma_i}{N_C\sigma_C} \frac{1}{(1-q'_0)} \quad (5)$$

As far as the parted specific volume of the main solute and of the impurities are constant, the value of $N_H\sigma_H + N_C\sigma_c + \Sigma\rho N_i\sigma_i$ is the same for different solutions. Thus if the influence of concentration on the terms $1/\epsilon'k$ and $1/(1 - q')$ can be neglected the extrapolation of f/A' to $f=0$ for both standard solution and the unknown solution gives two numbers the ration of which is equal to N_{C1}/N_{C2} .

The first instrument has the advantage of using less material and of being less susceptible to disturbing influences due to impurities or differences in concentration. The second instrument is, however, easier obtained as it requires no special equipment except a neutron source.

Figure 7. Apparatus for the simultaneous activation of eight aqueous samples in an identical neutron flux. An Am-Be source can be used in this apparatus. (Reference 16).

3. Applications of ^{241}Am - ^{242}Cm -Be Neutron Sources

Acree in 1964 developed a technique of producing high-output ^{242}Cm neutron sources. The ^{241}Am - ^{242}Cm -Be neutron source was produced by blending milligram quantities of ^{241}Am O_2 with milligram quantities of beryllium metal powder to a weight ratio of 1:10. The blends were pressed into 6-mm diam pellets, encapsulated in aluminum and irradiated at a flux of 2×10^{14} for 30 days. Approximately 21% of the ^{241}Am was converted to ^{242}Cm . ^{242}Cm has a half-life of 163 days and can be used to produce neutron sources in the 10^8 - 10^9 neutrons/second class.

The ABC (^{241}Am - ^{242}Cm -Be) source has been the subject of much interest because of its high neutron yield. The source has been considered for neutron radiography, process control and fast neutron activation. Wing and Wahlgren⁽¹⁷⁾ used a ABC neutron source with a flux of approximately 9×10^7 neutrons/cm²-sec (fast) to determine the relative detection sensitivities of 73 chemical elements (all the radioactively stable elements except beryllium, lutecium, osmium and the noble gases). The results of one run to establish detection sensitivities with this source which has a neutron spectrum that covers the energy range from thermal to ten⁽¹⁷⁾ MeV is shown in Fig. 8.

Figure 8 . Detection sensitivities of chemical elements vs. the gamma energies detected, for one-hour irradiation, one-hour cooling, 40-minute count. (Reference 15)

Stone⁽¹⁸⁾ has presented figures to show the relative use of neutron sources in industry for the period starting 1955 and projected up to 1975. These are reproduced below in figure 9. Figure 9 reflects the apparent acceptability of radioisotopic neutron sources. When the three sources of neutrons are compared it is clear that all of them present problems to a potential user. A reactor (of $\approx 10^8$ n/cm²/sec) is expensive, and technically and administratively complex to operate. (α ,n) radioisotope sources while capable of this level of neutron flux would be prohibitively expensive. Accelerators using the D-T reaction are capable of neutron fluxes of $\approx 10^8$ n/cm²/sec, but they require frequent target changes and enormous amounts of deuterium to operate at a high beam current for any significant length of time. Thus the radioisotope neutron sources of (α , n), (γ ,n) and spontaneous fission type are finding wide and increasing acceptances in industry. These sources are normally in the 10^6 - 10^8 neutrons/sec class.

The neutron fast yields of these sources are several orders of magnitude lower than some neutron generators, hence making it impossible to analyze trace quantities of elements using these sources. They can however be used to determine macro-quantities of elements with high enough effective activation cross-sections particularly in samples in which the matrix is only

slightly activated.

These sources are relatively low in price, easy to install and reliable. In installing these sources it is sufficient to surround the source with moderation (e.g. water and paraffin). The moderator (or moderator containment vessel) is in some cases surrounded by a cadmium sheet to absorb thermal neutrons reaching the surface of the shielding. Unlike many other neutron sources with high intensity gamma radiation, $^{241}\text{Am-Be}$ does not require the use of gamma shielding.

In many applications of neutron sources in instrumentation, where high neutron yields are not necessary and constancy of outputs is more important, sources like ^{244}Cm , ^{238}Pu and $^{241}\text{Am-Be}$ are more likely to be used.

Figure 9. Relative industrial use of neutron sources for process and quality control. 1955 to 1975 (Reference 18).

REFERENCES

1. M.A. Wohlgren and D.C. Stewart, "Evaluation of a ^{244}Cm -Be Neutron Source For Activation Analysis", in Conf-710402, Vol II, (1971), U.S.A.
2. K.H. Ansell and E.G. Hall, "High Intensity (α , n) Sources", in Conf-710402, Vol. II, (1971), U.S.A.
3. Californium-252 Progress, 4, August (1970).
4. R.F. Stewart and F.L. William Jr., "Nuclear Measurement of Carbon in Fly Ash", Proc: Edison Electric Institute - National Coal Association-Bureau of Mines Symposium, Pitts., Pa. 3/14-16/67.
5. P.N. Tiwari, "Rapid and Non-Destructive Determination of Protein In Grain Samples by Prompt (n , γ) Technique", Radiochem Radioanal. Lett., 6(6): PP 363-76 (1971).
6. Y. Kusnka, H. Tsuji and M. Egawa, "Rapid Determination of Boron in Glass By Prompt γ -ray Measurement" Presented at the 9th Radiochemistry Symposium of October (1965).
7. P.N. Tiwari et al, "Determination of Nitrogen in Organic Materials by Prompt n, γ Techniques", Int. J. App. Rad. Isot. (1971).
8. Starnes (Hilger and Watts, Ltd., London), In "Applications of Neutron Activation to On-Stream Analysis," by J.R. Rhodes, Isot and Rad. Tech., 6, (1969).
9. P.G. Geffrey and J.M. Bakes, "The Determination of Fluorine and Fluorite Ores and Concentrates by Isotope Source, Fast Neutron Activation Analysis", The Analyst, Vol. 92, P. 151, (1967).
10. J.E. Srain and G.W. Leddicotte, "The Preparation, Properties and Uses of Am-241 Sources", ORNL-3335 (1962).
11. U.S. Bureau of Mines; Californium-252 Progress Rep. #4, (1970).
12. J. Kuusi, "Radioisotope Neutron Activation Analyzer for Process Control Analysis", Nucl. Appl. Technol 8(5): PP 465-72 (1970).
13. J. Kuusi, "Use of Radioisotopic Neutron Sources for Process-Control Activation Analysis", Isotopes and Radiation Technol, 9(1), PP 48-52 (1971).

14. D.E. Wood, "Activation Analysis in the Metals Industry," Nucl News, 9(9), PP 12-17 (1966).
15. K. Boddy and D. Gloros, "The Measurement of Phosphorous in Human Bone Using Radioactive Neutron Sources--A Technique for Partial Body In Vivo Activation Analysis, Int. J. of App. Rad. and Instop., 24, PP 147-51, (1973).
16. P.W.F. Louwnier, A.H.W. Aten, Jr., and J.V. Hooivonk, "Two Instruments for Activation Analysis of Solutions by a Dilution Method," Int. J. of App. Rad. and Isotop., 24, PP 147-51, (1973).
17. J. Wing and M.A. Wohlgren, "Detection Sensitivities in Nuclear Activation with an Isotopic Fast-neutron Source," Analytical Chemistry, Vol. 39, 1, PP 85-89, (Jan.1967).
18. L.R. Stone, "Industrial Uses of Neutrons for Process and Quality Control," In Conf-710402 Vol I, (1971).

F. The Use of ^{241}Am -Be in Process Control

There are several methods of measuring moisture content on line in the steel industry. Most of the techniques are indirect or inferential. These techniques include the neutron scattering method. Most existing neutron probes utilized ^{241}Am -Be fast neutron sources for several different reasons not the least of which is the reduced radiological health hazard.

1. Discussion of Results

Neutron moisture probes are divided into three broad types. These are: the insertion type, which is inserted into the specimen; the surface type, which is pressed against the surface of the specimen; and lastly the transmission type where the source and detector are placed on opposite sides of the specimen (See figure 1). Papez and Cameron⁽¹⁾ reviewed the merits of these systems and gave experimental results on measurement sensitivity and other factors. These results and the comparisons are given in Table I. The results of a comparison test in which the insertion and surface types were used in the measurement of moisture content of the same sinter raw mixture are given in Figure 2. In 1958 Migagawa⁽²⁾ et al. started a series of basic experiments and on-line tests in which a neutron moisture gauge was employed in the measurement of sinter raw materials and blast furnace coke. A method of automatically correcting and weighing of the moisture content of the lump coke was developed. They compared the relationship between raw material (Sinter) density to moisture content by counting the output of a moisture meter (See Figure 3). The effect of density changes, as in soil moisture studies, is seen. Moisture measurement of the blast furnace coke was found to have accuracy of $\pm 0.95\%(1\sigma)$, with possible coke control per change of ~ 50 Kg and an accuracy of $\pm .28\%(1\sigma)$. The various errors of moisture measurements in the steel making industry are also examined and

Figure 1. Typical examples of the basic geometries of moisture meters.
(Reference 1).

GAUGE	4 π	2 π	TRANSMISSION
<u>Gauge Details</u> Energy emitted-detected Geometry Source: Activity Time constant Detector Wear Protection Reflector <u>Installation</u> Typical installation Modification to plant Disturbance to material flow	Fast-slow Backscatter $^{241}\text{Am}/\text{Be}$: 100mCi 10 sec 1 ^3He prop. counter: 150 mm x 25 mm ϕ 4 atm ^3He Steel access tube 12 mm thick NONE In hopper Mod. reqd. Flow disturbed	Fast-slow Backscatter $^{241}\text{Am}/\text{Be}$: 300 mCi 10 sec 2 ^3He prop. counters Radiation "window" 3 mm steel + 30 mm ceramic Steel On hopper Mod. reqd. NONE	Fast-slow Transmission $^{241}\text{Am}/\text{Be}$: 600 mCi 20 sec 1 LiI (Eu) Scint. counter: 4 mm x 38 mm ϕ NONE Steel On conveyor belt Very little mod. reqd. NONE
MATERIAL	COKE SINTER MIX	COKE SINTER MIX	COKE SINTER MIX
<u>Sensitivity</u> $(\frac{\delta I}{I} / \frac{\text{Sm}}{\text{m}})$ at 5% moisture by wt. <u>Precision (1σ)%</u> Moisture by wt. at 5% moisture by wt. <u>Factors affecting accuracy</u> Source decay Instrument drift per week Wear of protective device(Typical wear in 6 months) Material build-up Radius of sphere of influence(1) at 5% moisture by wt.	0.54 0.39 0.21 0.19 Negligible error <0.02% H ₂ O equiv. 0.05% H ₂ O equiv. Eliminated in 80 cm 40 cm	0.21 0.30 0.15 0.10 Negligible error <0.04% H ₂ O equiv. 0.1% H ₂ O equiv. good installation 30 cm 15 cm	- 0.54 - 0.22 (at .80 kg/m) Negligible error <0.02% H ₂ O equiv. Zero error 50% of load measured

TABLE I. Comparison of different neutron moisture gauges.(1)

Figure 2. Comparison of insertion type and surface type of detecting head on the sinter raw mixture. ° Insertion type; · Surface type. (Reference 1).

Figure 3 . The effect of density of material output of moisture meter. (Reference 2).

maintenance steps are mentioned.

In construction, controlling the fluctuation of the surface moisture content of aggregates, which has a significant effect on the workability and strength of concrete mixed in the concrete batching plant, is a difficult problem. Yamamoto⁽³⁾ used a type PB-1 moisture probe, which contained a 30 mCi $^{241}\text{Am-Be}$ source, mounted in a sand bin to measure the moisture content of each batch. This information was then used to adjust the weights of water and sand automatically to maintain the proper mixture. This method has the advantages of providing: (a) short measuring time, (b) a signal that can be connected to the continuous logging and automatic control system to ensure accurate fast-acting control of the supply of water and sand.

The continuous variation of the moisture content of the coke used in the making of pig iron affects the working of the blast furnace and quality of the pig iron. A neutron probe was installed on the coke bin of the Ougree' experimental blast furnace and operated continuously through 1965 and 1966. The source, a 100 mCi $^{241}\text{Am-Be}$, was inserted into the bin and the accuracy of the system was found to be $\pm 0.54\% \text{ H}_2\text{O}$.

Neutron probes are being used widely in the steel making, glasswork, casting, and other industries as a means of process control. In Japan alone prior to 1970, Sakaki⁽⁴⁾ reported that more than 100 neutron moisture meters were in use. Table II contains an incomplete list of reported industrial utilization of $^{241}\text{Am-Be}$ in process control applications.

Table 2. ^{241}Am -Be In Process Control Applications

SOURCE	INDUSTRY	PROBE TYPE	RANGE IN MOISTURE CONTENT	REFERENCE
^{241}Am -Be (30 mCi)	Concrete-Moisture	Surface, PB-1 Moisture Probe Detector, BF_3	0-10 weight % H_2O reproductivity ± 0.5 wt % H_2O	Yanomato ⁽³⁾ 1970
^{241}Am -Be	Glass-Production Moisture Measurement	Surface - Detector - Scintillation	Range of Moisture content unknown. Statistical re- liability of humidity determination is $\pm 0.1\%$ abs.	Caimann ⁽⁶⁾
^{241}Am -Be (100 mCi)	Testing of tar im- pregnation of mag- nesite bricks -	Unknown	N.A.	Frevert and Rotzer ⁽⁷⁾
^{241}Am -Be (100 mCi)	Steel-measurement of moisture content of the cake in a cake bin	Surface-Probe Detector- Scintillation	Accuracy of Probe data 0.20 % H_2O	Lucker ⁽⁵⁾ 1967
^{226}Ra -Be (10-20 mCi)	Steel-demonstration of the use of neutron gauges in Process Control in an on-line mode.	Surface and encenter Probes Detectors - BF_3		Myagawa ⁽²⁾ et al 1969

REFERENCES

1. K. Papez, J.F. Cameron and B. Machaj, "On-Line Neutron Moisture Gauges In the Steel Industry," IAEA, Sym. on the Use of Nucl. Tech. In the Basic Metal Industries, Helminski (1972).
2. K. Myagawa, I. Mishima, T. Takeda and Y. Tanabiki, "Application of Neutron Moisture-Gauge In the Steel Industry," Trans. ISIJ, V 9, PP 285-296 (1969).
3. M. Yamamoto, "Quality Control of Concrete by Neutron Moisture Gauge," EURISOTOP #51 (1970).
4. M. Sakaki, "Moisture Measurement by Neutrons", Toshiba Review, Jan.- Feb. (1969).
5. J. Luckers, "Continuous Measurement of the Moisture Content of Coke by Means of a Neutron Probe," G.N.R.M. No. 11 (1967).
6. V. Caimann, "Experience with Sand Humidity Measurement Using Neutron Probes," EURISOTOP #51 (1970).
7. , Frevert and , Rotzer, "Radiometric Measurements on Magnesite Bricks Testing the Tar Impregnation of Magnesite Bricks," EURISOTOP #51 (1970).

G. Neutron Radiography

1. Introduction

The development of neutron radiography during the period 1937-1960 parallels that of X-ray radiography for the period of 1900-1930. The adoption of the X-ray technique was slow until reliable and inexpensive X-ray sources became available. The idea of using a neutron beam as a nondestructive testing medium was introduced by Kallman⁽¹⁾ whose paper (1948) and earlier joint patents with Kuhn⁽²⁾ (3) outlined the basic principles of neutron radiography. Kallman⁽¹⁾ investigations of the use of neutrons in radiographical applications were conducted with a small accelerator with the strength of 2-3 gram Radium-beryllium. Peter⁽⁴⁾ (9146) using an accelerator of strength approximately equal to 10 kg of $^{226}\text{Ra-Be}$ also employs neutrons to make radiographs.

There were no further development in neutron radiography until Thewlis⁽⁵⁾ (1956) and Thewlis and Derlyshire⁽⁶⁾ initiated the application of the thermal nuclear reactor to neutron radiography. Once the nuclear reactor was applied to this problem as a neutron source, the attention of investigators turned developing detection methods and collimation techniques. In an extensive review on neutron radiography prior to 1965, Berger⁽⁷⁾ suggested that the extremely long lag time between discovery of the neutron and its application in radiography was caused by the paucity of suitable neutron sources.

The number of laboratories with work going on in this field have grown from approximately three⁽⁸⁾ in 1964 to well over 76 as of 1972. Facilities using nonreactor sources have been set up at several locations, e.g. Argonne National Laboratory's Facility at Idaho Falls, the Marshall Space Flight Center at Huntsville, Alabama, and the LSU Demonstration Center at Baton Rouge, La..

The various types of neutron sources used in neutron radiography are presented in table I.

The most commonly used radioisotope neutron sources are given in table II.

A major draw back of these sealed sources is their low neutron intensities. Watts,⁽⁹⁾ estimated a loss in intensity by a factor of approximately 10^6 in going from the MeV energy range to a collimated thermal neutron flux. Hence, for a thermal flux of $\approx 10^5$ thermal neutrons/cm²-sec, an uncollimated fast-neutron flux of $\approx 10^{11}$ neutron/sec is needed. ²⁴¹Am-Be neutron sources are available with neutron intensities of 10^8 neutron/sec.⁽¹⁰⁾

Several extensive reviews (other than Ref. 1) on neutron radiography have been conducted by Spowart,⁽¹¹⁾ Barton⁽¹²⁾, and Gerrard⁽¹³⁾. The types of neutron sources are discussed in detail and in most cases comparisons made between these sources (relative to: use, cost of source and portability). For thermal neutron radiography with small neutron source, an important quantity that has to be considered is the thermalization factor. The thermalization factor is defined as the ratio of the isotropic yield of a neutron source (neutrons/sec) to the Peak thermal flux (neutron/cm²-sec). Barton⁽¹²⁾ estimates a thermalization factor of 500 for ²⁴¹Am-Be/²⁴¹Am-²⁴²Cm-Be neutron sources.

SOURCE	TYPICAL NEUTRON FLUX (n/cm ² -sec) AT THE FILM	BEAM DIAMETER (cm)	RESOLUTION
Reactor Core Moderator	$10^7 - 10^9$	20 - 80	Excellent/high gamma content in beam
Reactor-Thermal Column	$10^5 - 10^6$	30	Excellent/lower gamma content
Accelerator Sealed Tube-Moderator	$10^3 - 10^5$	10 - 20	Fair/longer exposure times
Radioisotope Neutron Sources	$10 - 10^4$	10	Excellent-poor/ gamma content in beam dependent on choice of source
Van de Graaff	10^5	- -	Excellent/low gamma content- short exposure time
Cyclotrons (Table Top Type)	10^7	- -	Excellent

Table 1. Common Sources of Neutrons, typical neutron flux at the film, beam diameter and resolution.

TABLE II
Isotopic neutron sources tested for neutron radiography (Reference 12).

SOURCE	HALF-LIFE (APPROX)	MAIN REACTION	GAMMA- RADIATION EFFECT ON DIRECT EXPOSURE NR	THERMA- LIZATION FACTOR IN WATER	AVAIL- ABLE SOURCE INTENSITY SUIT- ABLE FOR NR (n/sec)	PEAK THERMAL NEUTRON FLUX IN MODERATOR (n/cm ² -sec)	PRESENT COST FOR EACH SOURCE OF STATED INTENSITY (\$)
²¹⁰ Po-Be	4 1/2 mon	α - Be	Just Acceptable	About 500	10^7	2.10^4	800
²³⁹ Pu-Be	24,000 yr	α - Be	Just Acceptable	About 500	10^7	2.10^4	1,200
²⁴¹ Am-Be	458 yr	α - Be	Just Acceptable	About 500	10^7	2.10^4	3,000
²⁴¹ Am- ²⁴² Cm- Be	5 1/2 mon	α - Be	Just Acceptable	About 500	10^9	2.10^6	2,000
²⁴⁴ Cm-Be	18 yr	α - Be	Just Acceptable	About 500	10^9	2.10^6	17,000
²⁵² Cf	2 1/2 yr	Pission	Just Acceptable	About 100	10^9	10^7	7,000
¹²⁴ Sb-Be	2 mon	γ - Be	Too High	About 10	1000 Ci	10^8	1,500

2. Principles of Neutron Radiography

For practical reasons neutron radiography is limited to samples of thickness less than a few mean free paths (< 6 mfp). For a thickness greater than a few mean paths the reduction of the uncollided neutron flux is such that interpretation of the radiograph is difficult. Thus, the prime requirement of radiation in a radiographic system is its ability to penetrate the material under test. The ideal situation for neutron radiography is that of a composited system with a light material surrounded by a dense metal (e.g. fluids in pipes, and ceramic cores in metal containers).

If a beam of neutrons of flux, $\phi(E)$, is incident on an object - Film-Foil System as shown in figure 1, the Total reaction rate is given by⁽¹⁵⁾

$$R_T = \int_0^{\infty} R(E) dE \quad (1)$$

where

$$R(E) = \phi(E) e^{-\mu_{ob}(E)} (1 - e^{-\mu_f})$$

$$\phi(E) = \text{Neutron flux at the object}$$

$$\mu_{ob}(E) = \ell_1 \Sigma_1 + \ell_2 \Sigma_2$$

$$\mu_f(E) = \Sigma_f \ell_f$$

The film density, D , for direct exposure is assumed directly proportional to the reaction rate per unit area in the foil times the exposure time, i.e.

$$D = kRt \quad (2)$$

The film density change (contrast) produced by a thickness ℓ_n of a material described by $\Sigma_n(E)$ is given by

$$\frac{dR}{d\ell_n} = \int_0^{\infty} -\Sigma_n(E) R(E) dE \quad (3)$$

and the change in contrast as a function object thickness is found to be

$$\frac{dD}{d\ell} = -\bar{\Sigma}_{\eta} D \quad (4)$$

where

$$\bar{\Sigma}_{\eta} = \frac{\int_0^{\infty} \Sigma_{\eta}(E) R(E) dE}{R}$$

The flux in equation (1) must be specified by the Maxwellian distribution at a certain neutron temperature.

There are several other factors that influence the quality of neutron radiography. These are:

- (a) Collimator Ratios
- (b) Collimator Optimum Aperture
- (c) Geometric Unsharpness
- (d) Detector Efficiencies
- (e) Exposure times

All of these factors have been studied by many investigators and the reader may consult anyone of the several reviews mentioned in the introduction for more detail.

Figure 1. Schematic representation of detection system for direct-exposure neutron radiography.

3. Experimental Results and Discussion

The interest in radioisotope neutron sources has increased steadily from the first application of ¹³⁴Sb-Be to neutron radiography by Warman 1965.⁽¹⁶⁾ The earliest published reports of the use of ²⁴¹Am-Be as a neutron source were those of Barton and Boutaine (1967)⁽¹⁷⁾ and Cutforth (1968).⁽¹⁸⁾ The radiography work by Barton and Boutaine using ²⁴¹Am-Be was to test the usefulness of the Americium-241-Beryllium in neutron radiography. Hawkesworth et al⁽¹⁹⁾ made use of an 5 Ci ²⁴¹Am-Be source of $\approx 10^5$ n/cm² couple with a granular scintillator, to compensate for the low neutron flux, to make radiographs of fibre health physics dosimeters. The ²⁴¹Am-²⁴²Cm-Be source has been used by Albertson and Rathke⁽²⁰⁾ and Barton and Klozan.⁽²¹⁾ There has been very little use made of either of the sources i.e. ²⁴¹Am-Be/²⁴¹Am-²⁴²Cm-Be in neutron radiography. The lack of applications of ²⁴¹Am-Be sources probably results from the event of ²⁵²Cf. ²⁵²Cf is considered, on the basis of technical performance, to be the best isotope source for neutron radiography. However, it does present certain problems related to availability and cost. The choice between ²⁴¹Am-²⁴²Cm-Be, ²⁵²Cf and ²⁴⁴Cm-Be, the sources capable of yields of $10^9 - 10^{10}$ neut/sec, depends to a large extent on the expected life time of a particular facility.

REFERENCES

1. H. Kallmann, "Neutron Radiography", Research, 1, PP 254-260 (1948).
2. H. Kallmann and E. Kuhan, Patents, German - 694, 634, 753, 326 (1938); U.S. 1, 230, 618; 2, 185, 757 (1937); 2, 245, 787 (1939); 2, 246, 443; French 886, 513; 185, 516 (1938).
3. H. Kallmann, "Neutron Radiograph", Research, PP 234-60 (1948).
4. O. Peter, Naturf, 1, P-557 (1946).
5. J. Thewlis, "Neutron Radiography", Br. J. Appl. Phys., 7 PP 345-350, (1956).
6. J. Thewlis and R.T.P. Derbyshire, AERE M/TN 37, (1956).
7. H. Berger, Neutron Radiography, New York, Elsevier (1965).
8. H. Berger, J.P. Barton and K. Ogawa, (Eds), Neutron Radiography Newsletter, American Society for Nondestructive Testing, Evanston, Ill. (Nos. 1-10, 1966-1969).
9. H.V. Watts, "Research on Neutron Interactions in Matter as Related to Image Formation", USAEC Reports ARF-1164-3-27. Armour Res. Foundation, Chicago (1960-62).
10. J.P. Barton, "Foreseeable Application of ^{252}Cf to Neutron Radiography", Isotopes and Radiation Technology, PP 285-291, (1970).
11. A.R. Spowart, "Neutron Radiography", J. or Phy. E, Sci Instr., V 5, #6, PP 497-511 (1972).
12. J.P. Barton, "Sources for Industrial Neutron Radiography", In Neutron Sources, Conf. (1971).
13. M. Gerrard, "Uses of Neutron Radiography", Isotope Technology and Development, PP 1-17 (1969).
14. J.P. Barton, "Radioisotope Sources and Neutron Radiography", Chem. Engr. Progress Symp. Series 66, PP 91-100, (1970).
15. M.J. Flynn, G.F. Knoll and A.K. Pozanski, "Neutrography in Medical Research and Pathology", In Neutron Sources, Conf. (1972).
16. E.A. Warman, "Neutron Radiography in Field Use," Matr. Eval., PP 543-550, (1965).
17. J.P. Barton and J.L. Boutaine, "Early Developments of Neutron Radiography In France", ORNL-tr-1780, (1967).

18. D.C. Cutforth, "Mater, Evaluation", 26, No. 449, (1968).
19. M.R. Hawkesworth et al, J. Mater. Sci., 4, P 817, (1969).
20. D.C. Albertson and D.P. Rathke, "Applications of Neutron Radiography Using a Radioactive Source", GE NEODRO 68 69-NED-42, (1969); Trans. Am. Nucl. Soc., 12, 2, 462 (1969).
21. J.P. Barton and M.F. Klozar, "Experiments with ^{241}Am - ^{242}Cm -Be for Neutron Radiography", presented at Spring Natl. Conf. Am. Soc. Nondestructive Testing (1971).

12 Am-241 --- A Photon Source

A. Am-241 Photon Source Encapsulation and Design

Earlier source fabrication techniques are discussed in Strain and Leddicottes' review⁽¹⁾. ^{241}Am is reported as being typically fabricated into source configurations in form of powdered AmO_2 . Strain et. al., describe the preparation of the oxide material by first precipitating out of a "stock" solution, the hydroxide, $\text{Am}(\text{OH}_3)$ by addition of NH_4OH to the initial mixture, separating the precipitate out by centrifugation, rinsing it with water and finally igniting it in a platinum crucible. The AmO_2 obtained is then ground to a fine powder, and encapsulated in a suitable container.

Though encapsulation methods have advanced since this review was issued (1962) stainless steel is still predominately employed as the capsule material for most commercial sources with usual source windows of about .010 inch. The attenuation of the stainless steel windows on photon emission is depicted on Fig. 1.

Small source configurations of ^{241}Am suffer from a high degree of absorption and as Strain and Leddicotte report surface area must be increased to attain high activity. Fig. 2 (a) illustrates an earlier design that would yield maximum photon emission with .25 gram of Am-241. Any additional source material did not increase the yield due to self absorption effects. An improvement, is the design depicted in Fig. 2(b) it could contain up to 3 grams of AmO_2 again encapsulated in stainless steel. Ansell and Stevenson⁽²⁾ (1967) report the use of a ceramic enamel matrix with ^{241}Am to attain source concentrations of over 300 mCi/cm^2 . Their encapsulation design is depicted in Fig. 3; again stainless steel is employed, with the radioactive material inset into a depression in a gold foil to reduce backface transmission.

Fig. 1. Absorption of Gamma Radiation from ^{241}Am in Stainless Steel.

(a) Source holder for AmO_2 up to 0.25 gram.

(b) Source holder for AmO_2 up to 3 grams.

Fig. 2. Source Holders Used in the Preparation of $\text{Am}^{241}\text{O}_2$ Gamma Sources.

A special glass "bead" source incorporating ^{241}Am and attaining activities up to 14 mCi has been developed by Ansell and collaborators for use in source/target assemblies. The ceramic point source is mounted in the apex of a truncated gold foil cone (See Gamma-Excited X-ray Sources), the entire cone encapsulated in an aluminum envelope.

As noted by earlier investigators, intense Am-241 sources can be attained by dispersing the radioactive material over an extended area configuration Giaugue⁽³⁾ (1968) and Pressly⁽⁴⁾ describe the use of annular source configurations to obtain intense gamma-excited x-ray sources. Al_2O_3 powder is used to absorb a given solution of AmCl_3 , then the resulting mixture is loaded into an annealed aluminum tube, (1/16 O.D.; .0005 in. wall thk.). The tube is then bent into an annular shape and sealed into a lead holder with expoy resin. The Al_2O_3 matrix facilitates even distribution of the activity and acts as an absorber for the alpha radiation.

Much of the recent source development work with ^{241}Am , evolves around the use of ceramic or ceramic matrix enamels for primary radioactive containment. Incorporation of the radioisotope into a ceramic matrix decreases the possibility of released particulates, substantially reducing safety hazards. Ceramic enamels loaded with a radioisotope have the added capability of being spread evenly over relatively large areas. Precautions must be taken in the choice of ceramic material employed, so as to avoid (α, n) and (γ, n) reactions with the matrix material and resulting neutron emission. Ansell and Hall⁽⁵⁾ report the development of a "new" enamel that has a low neutron emission rate, good adhesion to stainless steel and excellent spreading capability. Table I lists the characteristics of the new enamel.

Another configuration that attains the safety aspects of the ceramic and enamel sources is the rolled metal foil fabrication technique, Fig. 4. This design has adequate emission activities for the Np-L x-rays as well as the 60 keV γ -photon.

With the increased use of ^{241}Am in the process control industry in gauging applications, not only are high activity sources needed but special geometries must be employed. Radioisotope gauges to control thickness of strip steel must incorporate long line sources or utilize a point source in a scan operation. Ansell and Hall report the development of long line sources, utilizing ^{241}Am . The type of loadings achieved is summarized in Table II. The fabrication technique involves the incorporation of the radioisotope in insoluble form in a sintered metal matrix, cladding it with an inactive layer and working it to the desired length and diameter. Aluminum is reported to be employed as the metal matrix. Using this technique, a 50 mCi, ^{241}Am source several feet in length has been fabricated.

Most major source fabrication facilities utilize the above mentioned fabrication techniques. Fig. 5 through Fig. 9, depict some of the configurations with noted source preparation technique.

Figure 3. Source Capsule for γ 50 keV. Ref. 2.

TABLE 1 Properties of americium-241 enamel

Melting range	1000-1200 C
Max. act./unit area (photon output 60 KeV)	400 mCi/cm ²
Wipe test bare enamel (loading 400 mCi/cm ²)	0.015 μ Ci
8 hour 50 C immersion test (loading 400 mCi/cm ²)	0.01 μ Ci
Neutron emission	10-12 n/sec/mCi
Other neutron emissions - for comparison purposes	
AmO ₂ only	3-4 n/sec/mCi
AmO ₂ in Al rolled foil	6-7 n/sec/mCi

TABLE I

FIGURE 4 Rolled foil mounted in holder

TABLE II

O.D. of Wire inches	mCi/cm
0.100	125
0.088	100
0.070	62
0.060	46
0.040	22
0.027	10

Fig. 5. A New England Nuclear annular source /Target assembly preliminary design; up to 100 mCi, Am-241.

Fig. 6. Am-241, extended area disc source, activity 1Ci. Am-241 is incorporated into a ceramic enamel (Amersham/Searle).

Fig. 7. A CEA/IRE Am-241 source, stainless steel encapsulation, can accommodate from 30 mCi to 2 Ci activity.

Fig. 8. Design of Am-241 100 mCi line source, glass beads encapsulated in a stainless steel capsule, this type source emits neutrons due to (γ, n) reactions with elements (e.g. Si, Al, O) in the matrix, (Dimensions in mm.) (Courtesy of Amersham/Searle).

ANT SERIES CAPSULE

STAINLESS STEEL: Am 241
ALUMINUM: Cd-109 & Co-57

Fig. 9. Annular capsule design; (Courtesy Isotope Products Laboratories).

B. Photon Spectrum and Commercial Source Comparison

An energy diagram showing the several gamma-photons emitted following ^{241}Am alpha decay is illustrated in an earlier section (See ^{241}Am -- The Alpha Source). The decay process leaves the atomic structure in an excited state and subsequently Np-X-rays are emitted along with the gamma photons. Table III lists the prominent γ 's and x-rays.

As part of this program ^{241}Am source fabricators were contacted in order to obtain commercially available sources for evaluation purposes. Four facilities were initially contacted Amersham/Searle, Monsanto (Dayton), Isotopes Products Laboratories, and General Nuclear. All responded favorably but due to our requirements and the sale of General Nuclear's source fabrication facilities, photon sources were only procured from Amersham and Monsanto (Dayton). The sources obtained were both of disc geometry, stainless steel encapsulated with approximately equal windows.

The original objectives of the evaluation program were to compare commercially available sources as to absolute emission rate, and emitted photon spectrum. All testing was to be performed at ORNL's Isotope Development Center under our direction. Due to time, and equipment problems, the set of tests originally proposed was not completed. However, a low energy spectrum of both the Amersham and Monsanto sources was obtained, utilizing the Isotopes Development Centers Si(Li) detector system. Table IV lists the Si(Li) system parameters. The detector was designed especially for low energy work and the overall efficiency for the higher 60 keV photon is very low.

(7)

The data obtained is illustrated in Fig. 10 and 11. No information was available for the detector efficiency at the source to detector distance employed, and therefore only raw counts are plotted.

A comparison between the resulting low energy pulse height spectrum reveals no anomalous photon peaks in either source that might have occurred due to differing impurity materials, in the source. There is, however, a significant difference in the raw counts in the Np 26 keV x-ray peak (over a factor of 2) between the two sources. Generally the data indicates that the low energy photon emission rate in the Monsanto source is lower than the Amersham/Searle source. This would indicate a greater attenuation in the Monsanto source for lower energy photons. (See graphs).

The information supplied by the manufacturers, lists the source window thickness's as .010", .254 mm, (Monsanto) , .3mm (Amersham) and both employ stainless steel encapsulating material. The difference in low energy photon emission rates cannot be attributed, therefore, to vastly different window materials or thicknesses. Along with their source Amersham supplied a pulse height diagram indicating the relative intensities of the various emitted photons. (See Fig. 12).

RADIOISOTOPE	HALF LIFE	USEFUL RADIATIONS	INTENSITY PER DISINTEGRATION (6)
Americium-241	458 years	13.9 keV NpL α X-rays	0.14
		17.8 keV NpL β X-rays	0.18
		20.8 keV NpL γ X-rays	0.05
		26.4 keV γ -ray	0.03
		59.6 keV γ -ray	0.36

TABLE III

TABLE IV

Source to Detector Window Distance	--	18 in.
Counting Time	--	200 sec.
Detector	--	Si(Li)
Preamplifier	--	Ortec 117A
Lin-Amplifier	--	Ortec 951
Analyzer -- Nuclear Data Analyzer 510	--	4096 channels
Dead Time	--	35%

Figure 10. Source Data from Supplier

Amersham/Searle, AMC-17 Disc Type Photon Source
Nominal Output: 302 mCi
Am-241 Incorporated in a Ceramic Enamel
Source window: .3mm
Active Dia.: 12mm

RAW COUNTS (200 sec)

LOW ENERGY PULSE HEIGHT SPECTRUM (7)
FOR AM-241 COMMERCIAL SOURCE

CHANNEL NO.

Figure 11. Source Data from Supplier

Monsanto to Research Corporation, MRC-740 Disc Type Photon Source
Nominal Output: 301 mCi
Am-241 as AmO₂
Source Window: .254 mm (.010")
Outer Diameter of source container as .425", (approx. 10.8mm)

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Figure 12.

Low Energy γ /x-ray Spectrum from Am-241 Source AMC.3797
Measured on Si(Li) semiconductor detector.

REFERENCES

1. Strain, J.E. and Leddicottee, "The Preparation Properties, and Uses of Am-241, Alpha, Gamma and Neutron Sources," ORNL-3335
2. Ansell, K.H., and Stevenson, J., "The Development of Low Energy Photon Sources in the United Kingdom," ORNL-11C-10, Pg. 764.
3. Giaugue, R.D., "A Radioisotope Source-Target Assembly for X-ray Spectrometry," Analytical Chemistry, Vol. 40, No. 13, Nov. 1968, PP 2075-2077.
4. Pressly, R.S., "Design and Use of Low Energy Gamma and X-ray Sources," Applications of Low-Energy Gamma and X-ray Sources, PP 380, Zeigler, editor Gordon and Breach, N.Y. 1970.
5. Ansell, K.H., and Hall, E.G., "Recent Developments in Low Energy Photon Sources," Applications of Low-Energy Gamma and X-ray Sources PP 380, Zeigler, editor, Gordon and Breach, N.Y. 1970.
6. Lederer, C.M., Hollander, J.M., and Perlman, "Table of Isotopes," John Wiley and Sons, Inc., N.Y. (1968).
7. Pulse Height Spectrum Measurements made by R.S. Pressly, ORNL Isotopes Development Center.

C. Photon Attenuation and Backscatter Techniques

1. SOIL DENSITOMETRY -- TRANSMISSION METHODS

a.) Introduction

The use of the gamma transmission technique in soil densitometry was first suggested by Vomcil ⁽¹⁾ (1954) and Bernhard and Chsek ⁽²⁾ (1955) as a practical means of measuring the density of soil without destruction or disturbance of the sample. Sources which emitted high energy photons were initially applied to this task; mainly ^{137}Cs and ^{60}Co ⁽³⁾ ⁽⁴⁾. Since the initial work ⁽²⁾ in 1955, this method has been successfully used for measuring soil-water content in laboratories and in the field ⁽⁵⁾ (i.e. in situ). This technique of measuring soil-water content makes use of the single (SGEM) and the dual (TGEM) gamma energy approaches.

The principal justification for using gamma attenuation for measuring the water content of soil IN SITU resides in: (a) the resolving power of the method as it relates to the size of soil body seen by the detector, (b) the accuracy of the method, (c) the absence of a time lag in the availability of the information on the moisture content.

b.) Theory

Since the transmission method makes use of the well known exponential attenuation law, only the resulting expressions that are used to extract the desired information will be presented in this discussion.

The soil bulk density in a simple gamma transmission experiment can be expressed as:

$$\rho_s = \frac{\ln(I_0/I)}{X(\mu_s + \mu_w \theta_w)} \quad (1)$$

where I is the counts collected with a soil sample in the beam path. I_0 is the count rate with no sample; X is the sample thickness (cm); μ_w is the mass attenuation coefficient for water (cm^2/gm); μ_s is mass attenuation coefficient for dry soil; and θ_w is the gravimetric water content (gm/gm). Assuming that the mass attenuation coefficients μ_s and μ_w are known, Equation 1 still contains, besides the quantities X , I and I_0 , which are directly measurable, the unknown quantities ρ_s and θ_w . This makes it necessary for one to determine the dry bulk density of the soil or the gravimetric water content by other methods before the gamma scanning technique can be used to determine the remaining unknown. For example; gravimetric samples can be taken in equal thickness of soil to obtain a profile of θ_w ; the uncollided photon intensity measured at vertical increment; then using the values of θ_w and the measured values of I and the known values of X , μ_s , μ_w and I_0 the bulk density profile of the soil can be calculated utilizing equation (1). The same holds true for the case in which values of ρ_s are known and θ_w is to be measured using the gamma scanning technique.

This method assumes that the soil does not shrink or swell when the water content changes. Soane⁽⁶⁾, and Gardner and Calissendorff⁽⁷⁾, have demonstrated that this problem can be negated by applying a dual photon source method. Theoretically, the bulk soil density (gm/cm^3) and the water content (gm/cm^3) can be obtained from the simple exponential attenuation law as:

$$\rho_s = \frac{\mu_w \delta_1 \ln(I/I_0)_{\delta_2} - \mu_w \delta_2 \ln(I/I_0)_{\delta_1}}{\mu_w \delta_2 \mu_s \delta_1 X - \mu_s \delta_2 \mu_w \delta_1 X} \quad (2)$$

and

$$\theta_w = \frac{\mu_s \delta_1 \ln(I/I_0)_{\delta_2} - \mu_s \delta_2 \ln(I/I_0)_{\delta_1}}{\mu_w \delta_1 \mu_s \delta_2 X - \mu_s \delta_1 \mu_w \delta_2 X}$$

respectively. Therefore, ρ_s and θ_w can be determined if $(I/I_0)_{\delta_2}$, $(I/I_0)_{\delta_1}$ and corresponding mass attenuation coefficients are known. The two Gamma Energy methods provide a method whereby the bulk density and water content are both measurable by the gamma scanning method. The use of a two source combination made up of high and low energy photon emitter exploits the tremendous difference in the attenuation coefficient of soil for photons of energies >100 kev and those with energies <100 kev.

For example, it is desirable to have one source which is attenuated by water. There are many sources which satisfy these conditions. But there are other factors that influence the selection of sources. (See section ii page 209 of the summary bone densitometry for a discussion of these factors). However, with all factors considered, the $^{241}\text{Am} - ^{137}\text{Cs}$ combination is the best available for moisture and bulk density measurements (10).

In both the SGEM and TGEM the electronic instrumentation consists of the conventional detection and pulse height analysis equipment. In the laboratory, the detector, source and collimation geometry are not unlike other transmission set-ups. Field photon moisture source--detector probe arrangement for gamma transmission study is shown in Figure 1.

Figure 1. Source and detector probe arrangement for gamma transmission apparatus. (Reference 9).

C. MEASUREMENTS AND RESULTS

Even though the single energy method has been utilized in bulk density and moisture content measurements by numerous investigators, (5) ^{241}Am has been used sparingly (See table 1).

In soil density studies, using the single energy method, it is desirable to use a source that emits a photon with an energy high enough such that the attenuation cross section of the sample for that energy of gamma is independent of atomic number. (See Figure 2). For moisture content measurements, in which the dry bulk density of the soil is known, the energy of the photon is less crucial because hydrogen is a very slowly varying function of photon energy. However, the sensitivity of attenuation coefficients of the constituents of soil to photons of energies below 300 kev makes ^{241}Am ideal as the low energy photon in the two gamma energy method.

Soane (6) demonstrates the effectiveness of the Dual Gamma source method for measuring bulk density and water content of soils. Using 350 mCi of ^{241}Am and 30mCi of ^{137}Cs , he measured the mass thickness of water and soil of three samples of known soil-water mixtures and illustrated that the TGSM results agree very well with the known mass thickness of water and soil.

Figure 2. Total gamma attenuation coefficient for 5 common soil elements (Reference 10).

Isotopes	Location And Soil	Application	Method	Ref. & Year
²⁴¹ Am	Laboratory Sand	Water content measurement during transient, <i>non-</i> hysterectic flow of water in packed sand.	SGEM	Smiles et al ⁽¹⁴⁾ 1971
²⁴¹ Am	Laboratory Sand	Water content measurement during infiltration and redistribution in a column of soil of varying water content.	SGEM	Vachaud & Thony ⁽¹⁵⁾ 1971
²⁴¹ Am & ¹³⁷ Cs	Laboratory Sand	Demonstration of the utility of the TGSM for both water content and bulk density measurements.	TGEM	Soane ⁽⁶⁾ 1967
²⁴¹ Am & ¹³⁷ Cs	Laboratory Cecil and Houston Black	Measurement of water content and density changes in swelling and nonswelling soil.	TGEM	Corey et al ⁽¹¹⁾ 1970
²⁴¹ Am & ¹³⁷ Cs	Laboratory Simulated Soil (glass plates and water filled glass cells).		TGEM	Gardner and Calissendorff ⁽⁷⁾ 1967
²⁴¹ Am	Laboratory	Water content measurement of 1mm thick and 5cm diameter soil samples.	SGEM	Swart and Groenevelt ⁽⁸⁾ 1972.

Recently, Corey et al., (11) used a single collimator with an ^{241}Am source mounted in front (i.e. on the sample side) of the ^{137}Cs source. This arrangement makes it possible to construct a simple dual source system for IN SITU measurements of water content and dry soil bulk densities. He demonstrated the need for two gamma sources in moisture measurement of swelling soil. (See Figure 3).

The accuracy and precision of the dual gamma method of soil bulk density and moisture content measurements have been thoroughly examined by Gardner et al, (12). They estimated that the standard deviation in both water content and bulk density measurements to be $0.007 \text{ g/cm}^3/10^6$ counts and $0.005/\text{g/cm}^3/2.510^6$ counts when measured in air and a precision limit of about 0.0035 g/cm^3 at midrange values of water content and bulk density.

Both the SGEM and the TGEM have been used in soil-water content measurements in laboratories. The real strength of this method resides in field application where up to the present it has seen limited use. In addition to the need for site calibration for application of this technique, tests in the field have shown that the instrumentation is subject to electronic drift caused by electronic and temperature effects. IN SITU measurements by Reginato and Jackson (13) have been made which compensate for temperature effects. They found that with temperature compensation the absolute gravimetric water content can be measured with an average difference between gamma and gravimetric values of .009.

Figure 3. Water content and soil densities determined by the two gamma energy method prior to and 24 hours following the addition of water to a 5.1 x 5.1-cm clear plastic column partially filled with Houston Black soil. The dashed line indicates the water content that would have been calculated if the measurement had been made by cesium attenuation alone.

Summary

The photoelectric effect is important in the attenuation process for gammas in the energy range of ^{241}Am (i.e. 59.6 keV). From Figure 2, it is evident that the total attenuation coefficient is strongly dependent on the composition of the soil. Variations in the elements composition of soil from site to site can make the use of ^{241}Am for dry bulk density soil studies highly undesirable. This could result in large errors in the measured dry bulk density. The source (i.e. ^{241}Am) is quite acceptable for moisture content measurements using the SGEM because the total gamma attenuation coefficient for both hydrogen and oxygen are slowly varying functions of the incident gamma energy (See Figure 2.)

In the TGEM, ^{241}Am has been proven to be suited for the low energy gamma role (11). Several laboratory experiments have been conducted to demonstrate this technique but no IN SITU experiments have been reported in which the TGEM was employed. Ultimately the utility of any soil density measuring technique must be verified in the field.

- (1) Vomocil, J.A., "In situ Measurement of Soil Bulk Density", Agr. Eng. 39, pp631-54 (1954).
- (2) Bernhard, R.K. and Chasek, M., "Soil Density Determination by direct Transmission of Gamma Rays", Reprint 86 , ASTM, Phil., Pa. (1955).
- (3) Ferguson, H. and Gardner, W.H., "Water Content Measurement in Soil Columns by Gamma Ray Absorption", Soil Sci. Soc. Amer. Proc., 26, pp 11-14, (1962).
- (4) Van Bael, C.H.M., "Soil densitometry by Gamma transmission", Soil Sci., 87, pp 50-58, (1959).
- (5) McKinney, F.E., "Measuring Moisture with Gamma Radiation", Isotope and Radiation Technology, 7, (1), pp 29-36, (1969).
- (6) Soane, B.D., "Dual Energy Gamma-ray Transmission for coincident measurement of Water Content and Bulk Density of Soil", Nature, V 21, (1967).
- (7) Gardner, W.H. and Calissendorff, C., "Gamma ray and Neutron Attenuation in Measurement of Soil Bulk Density and Water Content", In Isotope and Radiation Techniques in Soil Physics and Irrigation Studies, IAEA, (1967).
- (8) Swart, J.C., DE, and Groenevelt, P.H., "Column Scanning with ke V gamma radiation", Soil Science, 112, pp 419-24, (1971).
- (9) Reginato, R.J. and Van Bael, C.H.M., "Soil Water Measurement with Gamma Attenuation", Soil Science Society of American Proceedings, pp 721-24, (1964).
- (10) Van Bael, C.H.M., " Soil Density Measurement with Gamma Radiation", Trans., 7th Int. Cong. Of Soil Sci., 1, pp 284-89,
- (11) Corey, J.C., Peterson, S.F., and Wakat, M.A., "Measurement of Attenuation of ^{137}Cs and ^{241}Am Gamma Rays for Soil Density and Water Determinators', Soil Sci. Soc. Amer. Proc., 35, pp 215-19, (1971).
- (12) Gardner et al, "Systematic and Random Errors in Dual Gamma Energy Soil Bulk Density and Water Content Measurements," Soil Sci. Soc. Amer. Proc., 36, pp 393-398, (1972).
- (13) Reginato, R.J. and Jackson, R.D., "Field Measurement of Soil-Water Content by Gamma-Ray Transmission Compensated for Temperature Fluctuations". Soil Sci. Soc. Amer. Proc., 35, pp 529-33, (1971).
- (14) Smiles, D.E., G. Vachaud and M. Vauclin, "A Test of the Uniqueness of the Soil Moisture Characteristic During Transient, Nonhysteretic Flow of Water in a Rigid Soil," Soil Sci. Soc. Amer. Proc., Vol 35, (1971).
- (15) Vachaud, G., and J.L. Thony, 1971, "Hysteresis Effects During Infiltration and Redistribution in a Soil Column at Different Initial Water Contents," Water Resources Res. 7(1), PP 111-127.

2. Bone Density and Mineral Content Measurement

a. Introduction

The direct photon absorptiometric method of bone-mineral measurement utilizes a narrow monoenergetic photon beam from a low-energy radionuclide source passed through the heterogeneous system (i.e. bone and tissue). The changes of transmittance, which are directly related to the mineral mass in the scan path, can be monitored with a suitable sensor and pulse height analyzer.

Bone density and mineralization measurements using low-energy photons were first reported by Cameron, *et. al.* (1) in 1962. In this initial attempt ^{125}I was employed as the photon source. Later in 1963, Cameron and Sorenson (2) reported the use of ^{241}Am in conjunction with ^{125}I as sources in the measurement of bone mineral. Mazess (3) *et. al.*, in 1964 presented a report on the accuracy of the method which showed that the correlation between the bone mass, as determined by the scanning technique, and the actual bone weight was 0.96. They also compared the scan results for ^{125}I and ^{241}Am using excised bones and found that both sources yield the same correlation with the actual bone mineral content.

b. Principle

(i) Single Photon Method*

The expressions for attenuation of monoenergetic radiation after passage through a thickness of tissue and bone material plus tissue, are given by, (4), (5), (6)

$$(1-a) \quad I_T = I_0 \exp (- \mu_T \rho_T \ell)$$

and

$$(1-b) \quad I = I_0 \exp (- \mu_B \rho_B \ell_B - \mu_T \rho_T \ell_T)$$

The subscripts B and T in equation (1) refer to bone and tissue respectively. The quantities μ_T , ρ_T , ℓ_T , μ_B , ρ_B , and ℓ_B and ℓ are the mass absorption coefficient for tissue in units of cm^2/g , the density of tissue, the thickness of tissue, coefficient for bone, the density of bone, the thickness of bone and ℓ is the total thickness of the sample.

After rearranging equations 1, one finds that equivalent thickness of bone mineral of density ρ_B is given by,

$$(2) \quad \ell_B = [\log_e (I^T/I) / (\mu_B \rho_B - \mu_T \rho_T)]$$

and the mass of mineral per unit area (gm/cm^2) by

$$(3) \quad m_B = \rho_B \ell_B$$

Cameron and Sorenson were first to present equation (2) and make measurements to test its validity. When "I" is measured at closely spaced intervals, Δx , across the bone one obtains a series of equivalent thickness, ℓ_B which when summed gives the equivalent cross-sectional area of the bone. The mass of bone mineral per unit length of bone is obtained as

$$(4) \quad M_B = m_B \Delta x$$

*This method of bone mineral measurement will be referred to as the single gamma energy method throughout the remainder of this text.

Equation (2) and (3) assumes that one has precise knowledge of the constant μ_B , μ_T and the corresponding densities. A schematic view of a typical system for measuring the bone mineral content by making measurements at equidistant point is given in Figure 1.

Figure 1 - A schematic view of a typical system for measuring the bone mineral content by making measurements at equidistant points.

Spring (7) has proposed a variation of the point measuring method in which, instead of measurements in equal spaced intervals, the source-detector system moves continuously at a constant speed. The method was checked by using a ^{241}Am source. Later, Mazess et al. (8) employed this continuous sweep method using a two energy method with ^{125}I and ^{241}Am as sources.

Bone mineral analyzers with the flexibility of utilizing ^{241}Am or ^{241}Am plus one or more additional monoenergetic photon sources have been manufactured by the Packard Instrument Company (9), (10) and the Sloan-Kettering Institute(11). With ^{125}I and ^{241}Am as sources these systems have the capability of making accurate measurements on both large and small bones. The 27 kev photon of ^{125}I provides good discrimination between tissue and bone, while the 60 kev photon of ^{241}Am adds the ability to analyze large bones. (See Figure 2 and 3)

(2) TWO PHOTON METHOD

This method requires that the absorbing medium be made up of two components which differ substantially in mass absorption coefficients for a low energy photon but are equal for a second photon of higher energy. While this is the optimum situation, it is certainly not a requirement. The mass absorption coefficients of two components of the medium for a high energy photon may differ, provided this difference is less than that at the low energy photon. (See figure 4, for example at 100 kev)

Sorenson and Cameron (14) proposed this technique for determining the fractional composition of a two-component system by using the differential absorption of two "monochromatic" photon beams.

Figure 2 - Scans of a child's femur with different radionuclides (12).

Figure 3-Scans of an adult radius with different radionuclides (12).

Figure 4 -- a plot of the mass absorption coefficient versus photo energy for fat and fat free tissue from 20 to 100 keV₀. (Schmonsees and Preuss⁽¹³⁾)

Taking the ratio of the logarithms of the expressions governing the attenuations of the beams from two sources while passing through the samples they obtain the relation

$$(4) \quad R = (\mu_{g1A}f_A + \mu_{g1B}f_B)/(\mu_{g2A}f_A + \mu_{g2B}f_B)$$

where μ_{g1A} , μ_{g1B} , μ_{g2A} , μ_{g2B} , f_A and f_B are mass attenuation coefficients for components A and B, for photons g_1 and g_2 and the fractions of components A and B.

Using the fact that

$$(5) \quad f_A + f_B = 1$$

the fraction of component A is given by

$$(6) \quad f_A = (\mu_{g1A} - R\mu_{g2A})/(R\mu_{g2B} + \mu_{g1B} - \mu_{g1A})$$

where f_A is a function of the known mass absorption coefficients and the measureable quantity R . The lack of dependence of Equation 6 on the thickness and density of the IN VIVO sample makes it a powerful and popular tool for determining fat and lean composition of IN VIVO samples. Cameron and Sorenson ⁽¹⁴⁾ using ^{125}I and ^{241}Am to measure the composition of samples of animal muscle and fat obtained results within 3% of that obtained by other methods of measurements. In a measurement of bone mineral content Roos et al ⁽¹⁵⁾ also applied the technique in a measurement of human vertebrae. The sources used in their measurements were ^{137}Cs and ^{241}Am .

d. Discussion of ^{241}Am as a Source of Photons in Bone Densitometry

The Single Gamma Energy Method

When the single source method of bone densitometry is applied there is no one source whose photon energy is optimum for all bones in IN VITRO studies. The suitability of certain sources for different skeletal locations is evident from the scan of a child's femur and that of an adult radius shown in figures (2) and (3). The dependency of the mass absorption coefficient on the photon energy is given in table I for the most commonly used radionuclides. In addition to the size of the bones, Judy (16) found that the optimum energy of photons for bone mineral content measurements increases with the increasing thickness of soft tissue cover. By far the most often used sources are ^{125}I and ^{241}Am . Both sources are essentially monoenergetic or can be made so with the use of filters. As a single gamma source ^{241}Am is more suitable for heavier bones (17) (18) (19), such as the humerus and femur. Its 458 year half-life practically means that it does not have to be replaced. However, its specific activity and the self absorption of its 59.6 keV photon make the use of relative large diameter sources almost mandatory in order to have sources that produce high enough counts, after passing through the sample to obtain the accuracy needed in a short exposure time. For example, a 100mCi ^{241}Am source has a diameter of 5mm compared to a 150mCi of ^{125}I with a source diameter of 3mm. The larger width of the photon beam (20) causes errors in the estimation of bone mineral. Even considering the sensitivity of ^{241}Am to bone edge criterion caused by the width of the beam and the irregularity of bone shapes, americium - 241 has proved to be useful for density measurements of the trabecula bone in the peripheral skeleton in population studies and in cases where the total thickness is of the order of 5 to 30mm.

NUCLIDE	HALF-LIFE (YRS.)	MODE OF DECAY (principal)	PHOTON ENERGY KEV	% OF DECAY	SPECIFIC ⁽²¹⁾ ACTIVITY THEORETICAL mCi/mg	SPEC. ACTIVITY COMMER. mCi/mg	MASS ABSORPTION COEFFICIENT ⁽²²⁾		COST %100 mCi
							Bone (cm ² /gm)	Muscle (cm ² /gm)	
^{99m} Tc	.2	Electron Cap- ture	140	90	5.0 X 10 ⁶		.0310	.0276	
¹²⁵ I	468	Electron Cap- ture	35 27 TeK	7 138	1.7 X 10 ⁴		.463 for the 35 Kev photons and 2.22 for the 27 Kev Photons	.110 for the 35 Kev Photons .459 for the 22 Kev Photons	\$75 ^a
¹⁰⁹ Cd	1.27	Electron Cap- ture	88 22 X-Rays	4 107	2.6 X 10 ³	1-3 ^a	.047. for the 88 Kev Photons 1.914 for the 22 Kev Photons	.0264 for the 88Kev Photons .459 for the 22 Kev Photons	\$75 ^a

TABLE I COMMONLY USED LOW ENERGY PHOTON SOURCES

NUCLIDE	HALF-LIFE YEARS	MODE OF DECAY (principal)	PHOTON ENERGY (KEV)	% OF DECAY	SPECIFIC ACTIVITY THEORETICAL mCi/mg	SPEC. ACTIVITY COMMER. mCi/mg	MASS ABSORPTION COEFFICIENT (cm ² /gm)		COST/100 mCi
							Bone	Muscle	
²⁴¹ Am	468	Alpha	59.6 trace to 0.72 Np L X-Ray	36	3.3	3 ^a	.0941	.0330	\$65 ^a
²¹⁰ Pb	22	Beta	47 11-13 Bi L X-Rays (Plus Bremsstra Lungs up to 1.17 Mev.	4 24	0.8 X 10 ²	10 ^a	~.179 for the 47 Kev Photons	.0432 for the 47 Kev Photons	\$65 ^a
¹³⁷ Cs 216	30	Beta	662	85 From 137mBa	~1.0 X 10 ²		~.0309	~.321	
¹⁵³ Gd	.65	Electron Capture	103 70 97 41 EuK X-Ray	20 2.6 30 110	3.3 X 10 ³	40-80 ^a	~.391 for the 100 Kev Pho-- tons	~.0263 for the 100 Kev Photons	

TABLE I- COMMONLY USED LOW ENERGY PHOTON SOURCES

For extended uses, radiation dose rate of any source must be considered. In terms of permissible dose rates, Cameron and Sorenson ⁽²⁾ estimated the exposure of .15 rem with 5mc of ^{125}I (27.3 kev) and .05 rem with 5mc of ^{241}Am (59.6 kev). This is well within the maximum permissible dose for the forearms of children of 7.5 rem per year and is much lower than the dose rate of an X-ray tube source ⁽²⁾ ⁽²³⁾ ⁽²⁴⁾ a comparable number of photons reaching the sensor.

Obviously, there are particular skeletal locations where one low energy photon source is more desirable than another one. One would not, for example, choose a ^{241}Am source for use in a study of the radius. (See figure 2) But in those cases where large bones or bones with thick tissue covers to be studied using the single source method, ^{241}Am is the most acceptable photon source to use. This can be seen from table I by comparing cost, half-lives, monochromaticity and photon energies.

Table II contains all the clinical applications and investigative studies in which ^{241}Am has been used up to the year 1972. In those cases where it might prove helpful the skeletal location studied is also given.

ii The Two Gamma Method of Bone Densitometry and Fat-Lean Ratios IN VIVO

The use of ^{125}I and ^{241}Am and other dual sources have proven to be efficacious in the determination of fat-lean ratios and bone mineral content. (See Table II) The 27.3 and 59.6 kev photons of ^{125}I and ^{241}Am are very well suited for these measurements. If the only errors involved are statistical in nature, an accuracy of to within less than 2% can be expected in bone mineral and fat-lean ratios measured by this method. For example, 1% for a 200 mCi and a 40 mCi ^{241}Am sources.

^{241}Am and ^{137}Cs has also been used in the two source method ⁽¹⁵⁾. This combination offers some advantages over the ^{241}Am - ^{125}I combinations in large bone studies with thick tissue covers. Schomensees and Preuss ⁽¹³⁾ ⁽⁵²⁾ compared the ^{125}I

PHOTON SOURCES USED	SKELETON LOCATION	MOTIVATION	REF. NO. & YEAR	METHOD
^{241}Am	Femur	Skeletal loss of calcium by patients being dialyzed	(46) Atkinson <u>et al</u> 1970	Single Gamma Energy Method
^{241}Am	Orstal end of the femur shaft	Serial studies of skeletal loss of Calcium on lacating mothers	(42) Atkinson <u>et al</u> 1970	Single Gamma Energy Method
^{241}Am and Strontium Isotopes	Pelvis and Spine	Transmission scanning of various skeleton locations	(48) Briggs 1968	Single Gamma Energy Method
^{241}Am	Anterior iliac Crest bone	Analysis of bone mineral content after renal transplant (metabolism)	(43) Bright 1969	Single Gamma Energy Method
^{218}Ra D & E and ^{241}Am	None: Dipotassium hydrogen phosphate as bone mineral substitute	Measurement of bone mineral content using the two gamma source measurement; estimation of the accuracy of the technique	(51) Burch and Bloch 1970	Two Gamma Energy Method
^{241}Am , ^{125}I , ^{210}Pb , ^{109}Cd , and ^{153}Gd	Non-Skeletal Components (fat and non-fat components) and long bones	To obtain quantitiative IN VIVO information for assessment of various clinical problems	(28) Cameron 1972	Both Single and Two Energy Method
^{241}Am and ^{125}I	Calf of the leg	Determination of the relative amount of fat in the calf of the leg	(29) Cameron and Sorenson 1966	Two Gamma Energy Method
^{241}Am , ^{125}I , and X-Ray Tube	Excised Bone	Determination of mineral content and a comparison of methods	(30) Cameron and Mazess, Sorenson 1968	Two Gamma Energy Method

PHOTON SOURCES USED	SKELETON LOCATION	MOTIVATION	REF. NO. & YEAR	METHOD
^{241}Am , ^{125}I	None	Test of the two source method for two-phase material Paraffin and CaCO_3	(2) Cameron and Sorenson 1963	Two Gamma Energy Method
^{241}Am and ^{125}I	Long Bones	Determination of bone mineral IN VIVO by absorption of monoenergetic photons	(32) Cameron 1970	Single Gamma Energy Method
^{241}Am , ^{125}I and ^{210}Pb	Adult radius Child Femur	Study of optimum design parameters (eg. choice of radiation source with consideration of cost, half life physical dimensions, and activity.)	(12) Cameron and Sorenson 1968	Single Energy Method
^{241}Am , ^{125}I	Os Calis (Physical Element Model)	Numerical Evaluation of two Gamma source method. Error and Dose Estimate	(23) Cohen and Gilson 1965	Two Gamma Energy Method
$^{219}\text{X-Ray}$, ^{241}Am and ^{125}I		Comparison of methods of bone mineral measurements etc. for clinical use	(49) Evens <u>et al</u> 1971	
X-Ray, ^{241}Am and ^{125}I	extreme skeletal locations	Comparison of four methods of bone mineral estimation in normal and osteoporotic women	(50) Goldsmith <u>et al</u> 1971	Single Gamma Energy Method
^{241}Am ^{125}I	Small Bone Sites	IN VIVO determination of bone mineral content.	(32) Johnston <u>et al</u> 1968	Single Gamma Energy Method
^{241}Am , X-Ray and ^{125}I	Radius	IN VIVO Measurement of bone mass and comparison of methods	(39) Johnston <u>et al</u> 1968	Single Gamma Energy Method
^{241}Am	Humerus Femur and Os Calcis	Use in bone mineral scanners	(26) July, P. <u>et al</u> 1967	Single Gamma Energy Method

PHOTON SOURCES USED	SKELETON LOCATION	MOTIVATION	REF. NO. & YEAR	METHOD
^{241}Am , ^{125}I	Calcaneus and Phalanx	Bone Mineral Measurement	(33) Lanzl <u>et al</u> 1970	Single Gamma Energy Method
^{125}I and ^{241}Am		Studies of absorptiometry in two component systems	(47) Leahy <u>et al</u>	Two Gamma Energy Method
^{241}Am	Small bone sites (non human)	Determination of the bone density and mineral content of Beagles	(45) Lloyd <u>et al</u> 1970	Single Gamma Energy Method
^{241}Am and ^{125}I 220	a) Forearm (IN VIVO) b) Excised bone	Experimental investigation of the use of monochro- matic radiation (princip- ally ^{125}I) for direct measurement of bone mineral. Compared Am-241 with I-125 in the study of Excised Bones.	(3) Mazess <u>et al</u> 1964	Two Gamma Energy Method
^{241}Am	Adipose Tissue	Excised bone and measure- ment of lean-fat composi- tion of soft tissue	(8) Mazess <u>et al</u>	Two Gamma Energy Method
^{241}Am	Femur	Measurement of Bone densities in athletes. Body height and weight comparison of bone density of athletes with non-athletes	(25) Nilsson and Westlen 1971	Single Gamma Energy Method
^{241}Am	Distal Femur	Measurement of: (1) bone mineral content in cases of fracture of tribal shaft (2) bone mineral content in the humerus in cases of osteopenia in the frozen shoulder (3) Influence on bone density of fluoride content in drinking water	(35) Nilsson 1968	Single Energy Method

PHOTON SOURCES USED	SKELETON LOCATION	MOTIVATION	REF. NO. & YEAR	METHOD
^{125}I and ^{241}Am	Two phase system (i.e. fat and lean) study of lamb carcass	A method for rapid estimation of fat content of fresh meat is examined	(34) Ringkob 1970	Two Gamma Energy Method
^{241}Am and ^{137}Cs	Lambar Vertebrae	Determination of bone mineral content in the study of Myeloma and demineralisation	(15) Roos <u>et al</u> 1970	Two Gamma Energy Method
^{57}Co , ^{125}I	Several Sites	Quantitative Determination of mineral content of bones Mineral, and Osteoporosis	(38) Rossow and Stureter 1969	Two Gamma Energy Method
^{125}I , ^{109}Cd , ^{241}Am	Lipid	^{109}Cd as a two photon source and ^{241}Am and ^{125}I were evaluated for dual beam absorptometry	(13) Schmonsees and Preuss 1972	Single Gamma Energy Method
^{241}Am , ^{137}Cs , (two source method) and ^{170}Th & ^{241}Am (single so.)	Femur, Radius, Metacarpes, and Ulna	Density measurements of bone. Mineral analysis	(44) Shimmins <u>et al</u> 1968	Single and two Gamma Energy Method
^{241}Am	Excised Humerus	Measurement of bone mineral to study: (a) Accuracy of the method (b) Reproducibility of the method (c) Clinical applicability of the method	(37) Sorenson <u>et al</u> 1968	Single Gamma Energy Method
^{125}I and ^{241}Am	Cortical Bone	Determination of bone mineral content as a function of its diameter	(41) Sorenson <u>et al</u> 1967	Two Gamma Energy Method

PHOTON SOURCES USED	SKELETON LOCATION	MOTIVATION	REF. NO. & YEAR	METHOD
^{241}Am , ^{125}I , and ^{99}mTc		Evaluation of different nuclide for use in rectilinear scanners and scintillation and cameras	(27) Sorenson and Cameron 1969	Single Gamma Energy Method
^{241}Am and ^{125}I		Measurement of composition of two component systems	(14) Sorenson and Cameron 1964	Two Gamma Energy Method
^{241}Am 222	Radius of the forearm	Development of a simplified method for bone mineral measurements IN VIVO using a continuously scan method	(7) Spring 1967	Single Gamma Energy Method
^{241}Am and ^{125}I	Os Calis	To develop a method of bone densitometry for clinical use so as to permit serial studies	(36) Strandjord <u>et al</u> 1968	Single Gamma Energy Method
^{241}Am , ^{75}Co , and ^{125}I		Measurement of mineral content of bone. Comparison of sources	(40) Strueter and Rossow 1969	Single Gamma Energy Method
^{241}Am	Femur	Discussion of parameters to describe bone density, accuracy and reproducibility. Particular reference to the femur.	(17) West and Reed 1970	Single Gamma Energy Method

PHOTON SOURCES USED	SKELETON LOCATION	MOTIVATION	REF. NO. & YEAR	METHOD
^{241}Am	Femur	Changes in bone mineral content in post and menopausal women.	() Nilsson 1970	Single Gamma Energy method
^{125}I and ^{241}Am		Osteodensitometer designed for single and two source scanning methods	(11) Zeitz 1970	Single and Two Gamma Energy Methods

and ^{241}Am combination with ^{109}Cd as sources of two distinct gammas to be used in dual beam absorptometry. ^{109}Cd , which emits a 22 kev and 88 kev photon, exhibits more desirable features for fat-nonfat density studies. One of these features is the higher intensity of its low energy beam relative to that of ^{125}I . Figure (2) also shows graphically that a ^{109}Cd source better satisfies the fundamental assumption of this method (52) as it relates to fat- nonfat ratio measurements.

REFERENCES

1. Cameron, J. R., Grant, R. M. and MacGregor, R., "An Improved Technique For The Determination of Bone Mineral Content in IN VIVO", Radiology 78 III, Jan. (1962)
2. Cameron, J. R. and Sorenson, J., "Measurement of Bone Mineral IN VIVO: An Improved Method" Science 142: 230-232, (1963)
3. Mazess, R. B. et al, "Accuracy of Bone Mineral Measurement", Science, Vol. 145 (1964)
4. Evans, R. D., The Atomic Nucleus, pp 711-736, McGraw-Hill Book Co. (1955)
5. Garrett, C. W., "On The Physics of Radiographic Bone Densitometry", In Progress in Methods of Bone Mineral Measurements, Conf. 680-211--2
6. Sorenson, J. A., "Evaluation of Radionuclide Sources for Transmission Scanning", Invest. Radiol. 5, pp. 92-102 (Mar.-Apr., 1970)
7. Spring, E. A., "Simplified Method For Bone Mineral Measurements IN VIVO", International Journal of App. Rad. and Isotopes, Vol., 18, pp. 700-11 (1967)
8. Mazess, R., Cameron, J. R., and Sorensom, J. A., "Determining Body Composition by Radiation Absorption Spectrometry", Nature, Vol. 228, Nov. 21, (1970)
9. Case, F. A., "Precision of the Norland Instruments Bone Mineral Analyzer", Proceedings of Bone Mineral Measurement Conference, Chicago, Ill., Cameron, J. R. (ed.), conf. 700515, (1970)
10. Schrodtt et al, "Features and Performance Characteristic of Packard Osteodensitometer", Proceedings of Bone Measurement Conference, Chicago, Ill. Cameron, J. R., (ed.), 700515 (1970)
11. Zeitz, L. and Freed, B., "Design and Calibration of Sloan-Kettering Institute Osteodensitometer", In Progress of Bone Measurement Conference, Chicago, Ill., Cameron, J. R. (ed.), Conference- 700515 (1970)
12. Cameron, J. R., and Sorensom, J. A., "Measurement of Bone Mineral by The Direct Photon Absorption Method: Principles and Instrumentation". In Progress in Methods of Bone Mineral Measurement, (1968)
13. Schmonsess, W. G., and Preuss, L., "Two Photon Absorptiometric Analysis, ^{109}Cd ." In Applications of Low Energy X and Gamma Rays, Ziegler, A. A. (ed.) (1972)

14. Sorenson, J. A. and Cameron, J. R., "Body Composition Determination by Differential Absorption of Monochromatic X-Rays, ORNL-11C-5, (1964)
15. Roos, B., Rosengren, B. and Skoldborn, "Determination of Bone Mineral Content in Lumbar. Vertebrae by a Double Gamma Ray Technique" "In Conference 700515", (1970)
16. Judy, P. F., "Theoretical Accuracy and Precision in the Photon Attenuation Measurement Of Bone Mineral", Proceedings of Bone Measurement Conf., May (1970), Cameron, J. R., (ed.) Conf. 700515
17. West, R. R. and Reed, G. W., "The Measurement of Bone Mineral IN VIVO by Photon Beam Scanning", Br. J. Radiol. 43. pp. 886-93. (1970)
18. Nilsson B., "Post-Traumatic Osteopenia, A Quantitive Study of the Bone Mineral Mass in the Femur Following Fracture of the Tibia in Man Using Americium -241 as a Photon Source", ACTA ORTHOP SCAND, 37, Suppl, 91 pp. 1-55 (1966).
19. Cameron, J. R., "Applications of the Direct Photon Absorption Technique For Measuring Bone Mineral Content IN VIVO". Determination of Body Composition IN VIVO. Progress Report (1969)
20. Sorenson, J. A., "Measurement Of Bone Mineral by the Direct Photon Absorption Method: Experimental Results", In Progress in Methods of Bone Mineral Measurements, conference 680 211-2 z (1968)
21. Wang Yen, (ed.), Handbook of Radioactive Nuclides, the Chemical Rubber Co., Pub. (1969)
22. Veigle, W. J. and Henry, E. M., "X-Ray Absorption in Bone and Muscle", In Applications of Low Energy X and Gamma Rays, Ziegler, A. A., (ed.) (1972)
23. Cohen, M.J. and Gilson, "Precision Methods using Soft Penetrating Radiation for Bone Densitometry", Progress in Development of Methods in Bone Densitometry, NASA Sp-6A, (1965)
24. Mazess, R. B., Cameron, J. R., and Sorenson, J. A., "A Comparison of Radiological Methods for Determing Bone Mineral Content", In Conf. of Progress In Methods of Bone Mineral Measurements, Bethesda, MD. Feb. (1968)
25. Nilsson, B. E. and Westlin, N. E., "Bone Density in Athletes", Clin. Orthop, 77 pp. 179-82, (1971)
26. July, P. et al], "New Bone Mineral Scanners", Conf.1422-10 (1967)
27. Sorenson, J. A. and Cameron, J. R., "Evaluation of Radionuclide Sources for Transmission Scanning", Conf. 1422-58, (1969)
28. Cameron, J. R., "Body Composition by Transmitted Gamma Ray Measurements", Tran. Am N c. Soc., Vol. 15 #1 (1972)
29. Cameron, J. R. and Sorenson, J. A. "Measurement of Bone Mineral and Body Composition 10 IN VIVO", pp. 37-42 of IVth Nordic Meeting on Clinical Physics, Proceedings. Rytila, A. Spring, E. (eds.) ((1966)

30. Cameron, J. R., Mazess, R. B. and Sorenson, J. A., "Precision and Accuracy Of Bone Mineral Determination by Direct Photon Absorptiometry", Invest. Radiol. 3 141 (1968)
31. Cameron, J. R., "IN VIVO Diagnostic Applications of External Radioactive Sources", Trans. Am. Nuc. Vo. 13 #2 (1970)
32. Johnston, D. C. Jr., Smith, D. M., Pao-lu Yu, and Deiss, W. P., Jr., Metabolism, Clinical and Experimental., pp. 1140-53, Dec. (1968)
33. Lanzl, L. H., Cox, Z., and Dobben, G., "An Additional System for Bone Denistrometry Studies", Conf. 700515 (1970)
34. Ringkob, T. P., "I. Estimation of Meat Composition by Differential Absorption of Gamma Rays. II. Effects of Parental Size on Carcass Composition and Performance of Lambs", PhD Thesis, Cornell University (1970)
35. Nilsson, B., "Bone Denistometry Using a Single Beam from a Radionuclide Source", In Progress in Methods of Bone Mineral Measurement, p. 319 Fe. (1968)
36. Strandjord, N. M. et al, "Clinical Applications", In Progress in Methods of Bone Mineral Measurement, p 308 Feb. (1968)
37. Sorenson, J. A. et al, "Measurement of Bone Mineral by Direct Photon Absorption Method: Experimental Results", In Progress in Methods of Bone Mineral Measurement, p. 269, Feb. (1968)
38. Rossow, J., and Strueter, H. D., "Radioisotope Method For Quantitative Determination of Mineral Content of Bones" Part II. Double Isotope Method". Fortschn Ge. Roentgenstr. Nuklearmed., 111 (2) pp. 155-63 (1969)
39. Johnston, D. C. et al, " IN VIVO measurement of Bone Mass in the Radius" Metabolism, Clinical and Experimental. Henry M. Stratton, Inc. (1968)
40. Strueter, H. D. and Rossow, J., "A Procedure for Quantitative Determination of Mineral content of Bones with Radioisotopes", Fortschn, Geb., Roentgenstr.
41. Sorenson, J. A. et al., "Bone Mineral Content as a Function of Bone Diameter in Cortical Bone" Coo-1422-17, (1967)
42. Atkinson, P. J. and West, R. R., "Loss of Skeletal Calcium in Lactating Women" J. Obstet. Gynae. Brit. Comm. 77, pp. 555-60.
43. Bright, R. W., Kalenak, A. and Elmore S. M., "Bone Metabolism Studies In Renal Transplant Recipients", Surg Forum, 20, pp. 461-3, (1969)
44. Shimmins, J. et al., "The Measurement of Bone Mineral IN VIVO by Photn Absorption", Calcif Tissue Res. Suppl: 40 (1968)
45. Lloyd, R. D. et al, "Americium -241 Studies in Beagles", Health Phys., 18, pp. 149-56 (1970)
46. Atkinson, P. J. et al., "Loss of Skeletal Calcium by Patients on Maintenance Dialysis" Biot. Med. Journal :29. pp. 490-2 (1970)

47. Leaky, D. M., Preuss, L. E. and Bugenis, C. K., "Some Studies of Absorptiometry in Two Component Systems Using Isotopic X-Ray Sources", Journal of Nuclear Medicine: 8. p. 338 (1967)
48. Briggs, R. C., Wilson, E. B., Sorenson, J. A., "Combined Emission-Transmission Scanning of the Skeleton", Radiology:90, pp. 348-50 (1968)
49. Evens, R. G. , Pak C. Y. C., Bantter, F. C. and Ashburn, W., "Clinical Use of an IN VIVO measurement of Bone Mineral Content", In Cameron Photon Absorption Technique of Bone Mineral Analysis, Norland Instruments, Ft. Atkinson, WI. pp. 19-31, (1971)
50. Goldsmith, N. F., Johnston, J. O., Ury, H., Vose, G. Colbert, C. "Bone Mineral Estimation in Mormal and Osteoponatic Women", In Cameron Photon Absorption Techniqe of Bone Mineral Analysis, Norland Instruments, Ft. Atkinson, WI., pp. 47-65. (1971)
51. Burch, W. and Black, P. "Two Wavelength Techniques for the Measurements of Bone Mineral Content IN VIVO", In Proceeding of Bone Measurement Conf, May (1970), Cameron, J. R. (ed.), Conf. 700515.
52. Schmonsees, W. G. and Preuss, L., "Two absorptiometric Analysis- ¹⁰⁹Cd", In Proceedings of Bone Measurement Conference, Chicago, ILL. May (1970), Cameron, J. R. (ed.) Conference 700515

3. THICKNESS MEASUREMENTS

a. Introduction

If a material is placed between a radioactive source and a detector, then depending (See Fig. 1) on the type and energy of the radiation, the atomic number of the material, the density and the thickness of the material, the radiation is blocked completely or attenuated. The amount of reduction in the initial intensity of the radiation is used to infer some property of the material. A second geometrical configuration is used in the backscatter gage. In this case the thickness and density are measured with the source and the detector on the same side of the material.

Several thorough reviews on the use of radioisotopes in thickness measurements⁽¹⁾ using both the attenuation process and the backscatter techniques are available. Cameron and Clayton⁽²⁾ (1971) in a comprehensive study dealt with the application and design of radioisotope instruments. This study includes a survey of radioisotope instruments used in thickness, mass per unit area and coating thickness measurements and the corresponding industries where the properties are measure. Table I contains a partial list (after Cameron and Clayton--Reference 2) of applications in several industries where ^{241}Am - might be used in thickness measurements.

Figure 1. Transmission gauge geometry.

Figure 2. Backscatter gage geometry.

Table I. Industries where ^{241}Am might be used in thickness measurements.
(i.e. thickness, coating thickness and mass per unit area).

INDUSTRY	APPLICATIONS
Agriculture, Forestry and Fishing	Mass per unit area of leaves in studies of water economy and balance in plants.
Mining and Quarrying	Thickness of residual layers of coal on floor and roof during automatic coal cutting.
Manufacturing Industries Food and Beverages	Mass per unit area of foods produced in sheet form, such as dough for biscuits and cakes, chocolate, cheese, chewing gum and meat products.
Textiles	Control of coating and impregnation processes in production of cellulose fabrics, tufted carpets, leathercloth, tyre fabric, artificial leather, linoleum, adhesive and abrasive cloth; control of warpknit fabrics in heat setting; thread mass per unit length; pick-up of moisture and extent of drying; wear of fabrics and garments.
Paper and Paper Products	Mass per unit area of paper and paperboard of all qualities and thickness; "bone-dry" weight of paper (measured in combination with a dielectric moisture gauge): studies of paper formation; control of coating and impregnation processes, e.g. polyethylene coatings on paper, gum on paper, clay content of paper and in production of laminated plastics.
Printing, Publishing and Allied Industries	Coating thickness of bi-metallic casts for offset printing, stereotypes and printing plates; thickness of ink and coatings on paper.
Rubber Products	Mass per unit area of rubber sheet, foam rubber and rubber coated or impregnated material such as tyre cords and floor coverings; studies of tyre wear.

INDUSTRY	APPLICATIONS
Chemicals and Chemical Products	Thickness and mass per unit area of plastic film issuing from extruders, plastic-coated paper and fabrics, laminated plastics; wall thickness of plastic tubes and bottles and of pipes and tanks in chemical plant.
Products of Petroleum and Coal	Asphalt impregnated products (roofing paper and shingles).
Non-metallic Mineral Products	Mass per unit area of abrasive papers and cloths, glass sheet, glass and asbestos fibres, asbestos-cement sheet, slate; selection of refractory bricks.
Basic Metals	Detection of "pipes" in ingots; thickness of hot-and coal-rolled sheet metals tubes and rods; steel sheet sorting; wall thickness of pipes and tanks; wear of furnace walls.
Metal Products	Thickness and/or coating thickness of tin and zinc plate and painted aluminium-coated and plastic-coated metals and a variety of coated and laminated metal products such as nuts and bolts, fixing brackets, miscellaneous domestic appliances.
Machinery	Thickness and mass per unit area of sheet materials used in machinery such as strip metal, coated and impregnated papers and textiles, solid and porous rubber and plastic sheets, coatings or platings on components, e.g. condenser paper, transformer windings and porous rubber sheets and plates for batteries.
Transport Equipment	Thickness or mass per unit area of sheet materials used in transport equipment such as steel for car bodies and ships hulls; leathercloth and other plastics for upholstery; thickness of plastic-coated radomes; width of cooling passages in turbine blades; engine wear research; wear of missile nose cones.

INDUSTRY	APPLICATIONS
Miscellaneous	Thickness of photographic emulsions and coating thickness on photographic base papers; coating of precious metals in jewellery manufacture.
Electricity, Gas, Water and Sanitary Services	Wall thickness of pipes, boiler tubes and hoppers; measurement of soot, water and snow depositon.

b. Theory

(1) Gamma Ray Attenuation Methods

The mechanism of attenuation is expressed by the equation

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

where I is the measured intensity, I_0 is the intensity of the source, μ is the mass absorption coefficient of the material, ρ is the material density, and x is the material thickness. This equation may also be expressed in terms of the area density

$$F = \rho x \quad (2)$$

It is clear then, that the attenuation method will permit one to measure either area density, density of a specimen of known thickness, or thickness of a material of known density. (3)

The ratio I/I_0 is known as the transmittance. The transmittance curve of Fig. 3 shows that a particular source-detector geometry may be used for a wide range of thicknesses. However, when used over such a wide range of thicknesses, the detector will experience large variation in intensity over the full range of thicknesses which is undesirable in practice due to the limits of sensitivity and stability of the detector. It is necessary in the general case to limit the measuring range. This is accomplished by adoption of the Principle of Constant Absorption. Utilizing this method the transmittance curve is reduced to one, two, or three partial ranges with measurements made only within these ranges. Macedo and Sercdynski⁽⁴⁾ gave an excellent discussion of the application, limitations. and sources of error of this principle in hot metal thickness gauging.

Figure 3. Transmittance curves for two source-detector geometries.(Reference 4).

The relationship between area density, F , of an absorptive material and the measured intensity, I , after attenuation can be described by the absorption curve. The absorption curve for ^{241}Am is shown in Figure 4. The ratio of I/I_0 can be represented by the equation

$$I/I_0 = \text{Exp}\left(-\frac{F_0}{F}\right) \quad (3)$$

where F_0 is a constant of the source, called the $1/e$ area density and depending on the atomic number Z of the absorber material. As seen in Figure 4, this dependence is considerable for ^{241}Am .

Gamma attenuation is by far the predominant technique in determining thickness with an ^{241}Am source. The sheet steel industry has stimulated a good deal of research on this technique and others. Table II gives comparison data for various techniques and source geometries. The THAM geometry type is capable of noise level or $1/4$ that of the THA at the same speed of response. Conversely THAM could be operated at a speed of response 16 times as fast as THA with the same noise level.

(2) Gamma Backscatter Techniques

The problem of measuring thickness in cases where there is present an interface of two media can be solved by utilizing the technique of gamma ray backscatter. A typical such application is in determining metal-on-metal or coating-on-base thickness determination.

The energy flux of back-scattered gamma radiation per unit solid angle is given by

$$dI_{\text{scat}} = QK_f E_0 \Omega_E a(E_0, \theta_0, Z, \theta, \phi) \quad (4)$$

where E_0 is the energy of gamma quanta reaching a reflecting surface; Q is the source strength, MeV/sec; K_T is the yield of gamma quanta of energy E_0 per decay event; Ω is the solid angle in which the primary radiation is emitted; $a_E(E_0, \theta_0, Z, \theta, \phi)$ is the differential energy albedo of the gamma quanta. (5)

It can be seen from ξ_0-4 that the intensity of back-scattered radiation is a function of the atomic number Z . This functional relationship is exploited by applying the method to two materials of different Z . Typical scattering materials are carbon, aluminum, plexiglas, iron, (Steel), cadmium, and lead. (3)

Given a sheet of back-scattering material of thickness greater than saturation thickness it is observed that the intensity of back-scattered radiation increases with increasing thickness of the overlay if the atomic number of the overlay is higher than that of the base and decreases with increasing thickness of the overlay if its atomic number is lower than that of the scattering material. (3)

The literature does not divulge a large number of gamma back-scattering devices utilizing ^{241}Am . In principle the isotope could be used in a large number of industrial applications. Among these are:

1. Measuring the single wall thickness of pipe
2. Measuring the amount of shale in coal
3. Determining the thickness of a coal vein
4. Gauging metal coating thickness

Table II. Standard Geometries for Steel Application

Geometry Type	Source	Half Life, Years	Type Radiation	Strength milli-curies	Energy, mev	Range for steel, thousandths of an inch.		Air gap in.	Frequency response d-c to cps	Source statistics as a % of thickness at 1 and 1/2 % resolution			
						1% res.	1/2% res.			1% low	1/2% low	1/2% high	1% low
TLK	Kr-85	10	Beta	1000	0.67max.	0.1-5	0.4-3	1/2	0.3 0.6 1.6	1.0 1.5 2.2	0.4 0.6 0.9	0.2 0.2 0.3	0.2 0.3 0.6
TM	Sr-Y-90	28	Beta	25	2.28max	1-20	2-18	1-1/2	0.3 0.6	1.5 2.2	0.8 1.1	0.3 0.6	0.4 0.6
TM	Sr-Y-90	28	Beta	100	2.28max	1-20	2-16	1-1/2	0.3 0.6 1.6	0.8 1.1 1.7	0.4 0.6 0.9	0.2 0.2 0.4	0.2 0.3 0.5
THAM	Am-241	458	Gamma	1000	0.06mono	5-150	12-120	6	0.3 0.6 1.6 3.5	0.4 0.5 1.3 1.0	0.2 0.3 0.6 0.9	0.1 0.1 0.2 0.3	0.1 0.1 0.3 0.4
THAM	Am-241	458	Gamma	1000	0.06mono	5-150	12-120	12	0.3 0.6 1.6 3.5	0.6 0.8 1.3 1.9	0.3 0.4 0.8 0.9	0.1 0.2 0.2 0.3	0.1 0.3 0.3 0.4
THA	Sr-Y-90	28	Brem.	1500	2.28max	5-300	10-50	4	0.3 0.6	1.3 1.9	0.6 0.7	0.4 0.5	0.4 0.5
THA	Sr-Y-90	28	Brem.	1500	2.28max	5-300	10-50	6	0.3 0.6	1.6 2.3	0.7 1.0	0.5 0.7	0.5 0.7
TH	Sr-Y-90	28	Brem.	300	2.28max	10-600	40-250	6	0.3	2.3	1.0	0.6	0.6
TH	Sr-Y-90	28	Brem.	300	2.28max	10-600	40-250	12	0.3	3.4	1.4	0.9	1.2
TH	Sr-Y-90	28	Brem.	700	2.28max	10-600	40-250	6	0.3 0.6	1.5 2.2	0.7 0.9	0.4 0.5	0.5 0.7
TH	Sr-Y-90	28	Brem.	700	2.28max	10-600	40-250	12	0.3 0.6	2.3 3.2	1.0 1.4	0.6 0.6	0.8 1.1

Figure 4. Absorption curve for ^{241}Am (0.06 MeV) for Aluminum ($Z=13$), Steel ($Z=26$) and Copper ($Z=29$). (Reference 3).

Figure 5. Source-scatter -detector geometry for backscatter measurements. (Reference 6).

It is also possible to combine radiation techniques in a single-duty instrument. A patent has been granted for a dual mode fluorescence and back-coating thickness measuring gauge which measures the thickness of zinc coating on a ferrous base in a typical galvanizing process.⁽⁷⁾ The instrument requires a 0.60 MeV gamma radiation source.

The gauge source of radiation is used for exciting the coating and base material, causing both to fluoresce, and for producing back-scattered radiation. The gauge is equipped with a pair of detectors which have response characteristics including both fluorescent emission and the back scattered radiation.

Trost⁽⁸⁾, used the backscattering of the 60 keV photons from ²⁴¹Am to measure the thickness of low atomic number substances (e.g. glass, rubber and plastics). ²⁴¹Am was found to be better suited for thicknesses greater than the practical range of Beta particles (e.g. 0.5 and 5 g/cm²). Using a prototype for the thickness gauging of 3-mm glass a precision of ± 0.003 mm was obtained with count rate circuit with a 2 sec time constant. Table III shows some of the applications of ²⁴¹Am in thickness measurements.

Table III. List of applications of ^{241}Am in thickness measurements

ISOTOPES	APPLICATION	REFERENCE & YEAR
^{241}Am	The determination of the ash content of coal, thickness gauging of glass, plastics and metals with ^{241}Am -rays using the backscatter technique.	Trost ⁽⁸⁾ (1967)
^{90}Sr , ^{147}Pm ^{241}Am , ^{226}Ra and ^{85}Kr	Review: Primary metal industry; measurement and control system; Thickness	Griffith ⁽¹³⁾ (1967)
^{137}Cs and ^{241}Am	Metal thickness measurement used in primary metal industry also examines statistical errors.	Macelo and Seredynski ⁽⁴⁾ (1970)
^{147}Pm , ^{210}Po ^{241}Am , ^{90}Y , ^{241}Am , ^{137}Cs and Co	Thickness and density measurements in the paper, plastic, and fabrication industries	Langel ⁽³⁾ (1970)
^{147}Pm , ^{210}Po , ^{241}Am , ^{90}Sr , ^{35}S , ^{204}Ti , ^{14}C , ^3H , ^{137}Cs , and ^{60}Co	Measurement of Metallic Corrosion using backscattering and radiation-induced x-rays.	Berk ⁽⁹⁾ (1965)

Table III continued--

ISOTOPES	APPLICATION	REFERENCE & YEAR
^{241}Am , ^{90}Sr	Thickness Measurement on plate glass	Langel ⁽¹⁰⁾ (1965)
^{85}Kr and ^{241}Am	Thickness gauging with special respect to aluminum materials	(11) (1967)
^{241}Am , ^{147}Pm , ^{85}Kr , ^{204}Ti , ^{90}Sr , ^{106}Ru , ^{137}Cs and ^{60}Co	Review: Along density measurements, discusses thickness measurements as it relates to measurement equipment for technical use.	Kosmowski ⁽¹²⁾ (1966)

REFERENCES

1. Isotope and Radiation Technology, "Isotope Technology Development," 1(2), Winter (1963-1964).
2. J.F. Cameron and G. Clayton, Radioisotope Instruments, Part I, Pergamon Press Ltd., OX., England (1971).
3. H. Langel, "Thickness and Density Measurements with the Aid of Nuclear Radiation," Kerntechnik 12, January, 1970.
4. F.X. Macedo, and , "Isotope Gauges: An anlaysis of Some Errors in Hot metal Thickness Measurement,"
5. B.P. Bulaton, and I. Ya. Serebrennikov, "Effect of Density of the Medium on Height and Level Measurements Made with Aid of Backscattered Gamma Radiation," in Radiation Technology, No. 3, Proceedings of the Institute of Radiation Technology, ed. A.S. Sutan' (Jerusalem, 1971).
6. J.H. Tolan and W.T. McIntosh, :Investigation of Applications of Compton Backscatter," USAEC Report NYO-2779, (1960).
7. You Min Chen, "Dual Mode Flourescence and Back-Scatter Coating Thickness Measuring Gauge," U.S. Pat. No. 3,497,691, issued Feb. 24, 1970.
8. A. Trost, "Determination of the Ash Content of Coal, Thickness Gauging of Glass, Plastics and Metals with Americium-241 Gamma-Rays," In Radioisotope Instruments in Industry and Geophysics, I, Vol 1, PP 435-45, IAEA, (1966).
9. S. Berk, Measuring Metallic Corrosion by Radiation Backscattering and Radiation-Induced X-rays," Mater. Protect., 4(11), PP 39-41. (1965).
10. H. Langel, "Thickness Measurements on Plate Glass by Means of Nuclear Radiation," (in German), Ind-Anz, 87(82), PP 1974-7, (1965).
11. "Thickness Gaging by Means of Kr-85 Bremsstrahlung and Am-241. Sources with special respect to Aluminium Materials," (in German), Isotopenpraxis, 3(7), PP 263-5, (1967).
12. A Kosmowski, "Measurement Equipment for Technical Use of Radionuclides," Z. Instrum, 74(9), PP 291-4 (1966).
13. J.L. Griffith, "Progress Report-Radioisotope Thickness Measurement Systems and Techniques," Iron and Steel Engineer, February, 1967.

4. Densitometry -- Backscatter Techniques

a) Introduction

Almost all of the isotopic density measurement systems in use are based on the principles of beta backscatter or gamma transmission. There are many applications (e.g. Highway construction) where the gamma backscatter technique is well suited. The initial studies of Hine and McCall⁽¹⁾(1954) and Hayward and Hubbell⁽²⁾(1954) established much of the basis for this method. In principle, the radiation emanating from a source propagates through the medium to be measured and interacts in a manner such that some of the radiation is scattered back to a detector. The amount of interaction or scattering is primarily a function of the density of the scattering medium. Thus the amount of radiation detected is a good measure of the density of the scattering medium.

b) THEORY

The interaction of the incident radiation and the electrons of a sample can be described mathematically by the classical Thomson scattering cross-section. According to the Thomson scattering cross-section, the fraction of low energy radiation scattered by an electron is given by

$$\frac{d\sigma}{d\Omega} = \frac{e^4 (1 + \cos^2 \theta)}{32\pi^2 \epsilon_0^2 (M_0 c^2)^2}$$

where $\frac{d\sigma}{d\Omega}$ = fraction of incident radiation scattered per unit solid angle at and angle θ WRT the incident angle.

e = electronic charge

ϵ_0 = permittivity of the vacuum

θ = angle of scatter

Using a single-scattering approximation (Fig 1), the gamma flux scattered at an angle θ_2 (WRT normal to the surface) is given by

$$I_s = \frac{6.02 \times 10^{23} \rho Z S}{A H^2 (\mu_0 + \mu_1 \cos \theta_1 / \cos \theta_2)} \frac{d\sigma}{d\Omega} I_0 \quad (2)$$

where ρ is the density of the scattering medium, 6.02×10^{23} is Avogadro's number; H is the distance from the scattering medium, cm; S is the surface area of the scatter; Z and A are the atomic number and mass number respectively; μ_0 and μ_1 are the linear absorption coefficients of the incident and scatter photon, respectively. Equation 2 indicates that I_s is a maximum when the incident beam is tangential to the surface ($\theta_1 = 90^\circ$) and minimum when it is normal to the surface.

Fig. 1. Schematic diagram of an elementary scattering event of a gamma quantum: $E_0 = h\nu$ - energy of incident quanta; $E_1 = h\nu$, - energy of scattered quanta; θ - scattering angle. $\theta = 180^\circ - (\theta_1 + \theta_2)$

The intensity of the scattered radiation of the detector is obtained by integrating eq. 2. over the solid angle subtended by the detector and the thickness of the scattering medium.

Thus:

$$I_D/I_0 = K \frac{Z}{A \rho} \quad (3)$$

where

$$K = \int_0^x \int_{\Omega_D} \left(\frac{6.02 \times 10^{23} S d\sigma/d\Omega}{A H^2 (\mu_0 + \mu_1 \cos\theta_1 / \cos\theta_2)} \right) dx d\Omega_D .$$

In eq. 3. I_D is the intensity of scattered radiation reaching the detector.

c. Measurements and Results

Quantitative analysis of cement in concrete is made possible by virtue of cement's relatively unique and substantially constant chemical composition. The backscatter technique, where low energy gamma rays are used, is one of the several nuclear techniques used in determining cement in concrete. Some of the geometrical arrangements in which low energy photons can be employed are shown in figure 2. Berry⁽³⁾ made a detail study of the depth backscatter probe. The probe consisted of one - in. diameter by one - in. thick NaI(Tl) crystal scintillation detector and a 2 - mm diameter spherical 14 mCi ²⁴¹Am source mounted on a conical radiation shield and axially separated from the crystal by a distance of about 2 in. The source-and-detector assembly is encased in a 0.020 in-wall stainless-steel tube (See Fig. 3). The theoretical performance of the gage in terms of the backscattered ²⁴¹Am intensity as a function of cement content, water content and bulk density was compared with experimental results by Berry⁽³⁾ and the results are shown in Fig. 4.

Fig. 2. Geometrical arrangements employing low-energy radiation scatter and transmission for cement analysis. (Ref. - 3)

Fig. 3. Probe Layout. (Ref. - 3)

Fig. 4. Comparison of theory and experiment. (Ref. - 3)

Tanemura and Suita⁽⁴⁾, conducted a systematic study on γ - and X-ray sources of energy below 60 keV employing the backscatter technique, as a means of coal ash analysis. ^{241}Am was used as a source of photons to excite the characteristic X - rays in a target material.

Figure 5. shows the reciprocal intensity of scattered X - rays and γ as a function of ash content for three sources, ^{241}Am - Ba, ^{241}Am - Ag and ^{241}Am . The gradient of the calibration lines gives the quantity:

$$\alpha = \frac{\mu_{1a}' + \mu_{2a}'}{\mu_{1c} + \mu_{2c}} - 1, \quad (4)$$

The quantities μ_{1a}' , μ_{2a}' , μ_{1c} and μ_{2c} are the linear attenuation coefficients for the ash and carbon, respectively. Then the content of ash is given by

$$\omega_a = (I_c/I_s - 1)/\alpha \quad (5)$$

where

I_c = intensity of scattered γ - or X-rays from coal without ash.

I_s = intensity of scattered γ - or X-rays with ash content.

Eight samples with a constant ash content of 20%, but with varying amounts of Fe_2O_3 from 0.5 to 4.0% content were also tested. The ^{241}Am - γ source was less sensitive to changes in Fe_2O_3 content as in the case (Fig.6) where the ash content was varied.

Crosland et al⁽⁵⁾, describes the types of ash monitors available and suitable for use in coal preparation plants. Included in the description of different instruments is a discussion on the Simcar (an

instrument produced by Simon Carbes, Ltd) which uses ^{241}Am .

The Simcar recorder is sensitive to ion variation and can be used for larger coal.

The backscatter technique affords the experimentalist the opportunity to use a very simple experimental arrangement. This results from the fact that the source and detector can be contained in a single container.

Fig. 5. Reciprocal intensity of scattered γ - or X-rays vs. ash content.

REFERENCES

1. G. J. Hine and R. C. McCall, "Gamma-Ray Backscattering," Nucleonics, 12(4), PP. 27-30, (April 1954).
2. E. Haywood and J. H. Hubbell, "The Backscattering of the ^{60}Co Gamma Rays from Infinite Media," J. Appl Phys., 25, PP. 506-509, (1954)
3. P. F. Berry, "Nuclear Applications in Highway Research," Isotopes, and Radiation Technology Volume 8, No 2. Winter (1970-1971).
4. T. Tanemura and H. Suita, "Studies on Radioactive X- and Gamma- Ray Sources applied to the Determination of Ash Content of Coal", Oyo Buturi, 36(6), PP 444-51, (1967).
5. R. Crosland, D. A. Hall and M. K. Laverick, "Continuous Ash Monitoring and Coal Blending Processes," In "Mineral and Extractive Metallurgy," M. J. Jones (ed), Inst of Mining and Metallurgy, London, PP 179-200, (1971).

D. Low Energy Photon Induced Fluorescence

1. Introduction

The 60 kev γ -ray and the lower energy Np x-rays emitted in the decay of ^{241}Am to ground state Np are of sufficient energy and occur with adequate intensity, to be utilized as a source of incident radiation to produce fluorescent x-rays in suitable target elements.⁽¹⁾ Over the past 15 years the use of low energy photon excitors has grown tremendously in a variety of fields. A report published in 1970 by a panel of IAEA⁽²⁾ presents a thorough review of radioisotope induced fluorescence and its many applications. Our purpose here is to specifically identify the applications, where ^{241}Am is employed.

Generally, fluorescence is employed as an analytical tool to determine the presence of particular elements in a target material. In this regard radioisotope induced fluorescence is complementary to conventional x-ray tube techniques, atomic absorption methods or neutron activation analysis. Though the resulting intensity from radioisotope induced fluorescence is 10^7 times smaller than conventional x-ray tube excitation and can never achieve the sensitivity of neutron activation analysis, it has the advantage of being incorporated into a portable analysis system capable of being employed in the field and in "on stream" analysis systems. Radioisotope fluorescence systems are less complicated and less expensive than other analytical techniques and with the advent of high resolution solid state detectors, radioisotope analysis does approach the sensitivity of x-ray tube methods, notably in the

excitation of x-ray spectra between 20 to 100 keV. Long lived photon emitters like Am-241 provide stable excitation sources and unlike x-ray tube sources, radioisotope emitters can be chosen to be essentially monochromatic.

2. Physics of Fluorescence

In order to understand the limits and advantages of Am-241, in radioisotope induced fluorescence, it would be worth while to give a brief summary of the physics involved in the primary interaction. A more thorough treatment of the subject can be found in standard texts and various papers in the field.^{(3) (4) (5)} Generally speaking, x-ray spectroscopy deals with the excitation characteristics of elements in a target material, by some form of incident radiation (heavy charged particles, electrons, photons). The interest here is in photon excitation.

An incident photon, via a photo-electric interaction, with an orbital electron in a target element can provide sufficient energy to eject the inner orbit electron from the lower energy level thus leaving a vacancy.

This vacancy is then filled by a transition of an electron from an outer (higher) energy orbit to the lower inner orbit. The inner orbit thus excited by the transition, loses this energy by either emitting x-rays, characteristic of the energy separation between the two respective energy levels, or by transferring this energy to an electron in the lower energy shell, thereby ejecting it. The emitted electrons are known as Auger electrons and this process competes with the emission of the characteristic x-rays.

An indication of the ability of a particular target element to emit x-rays under the influence of incident photon radiation, rather than Auger electrons is termed the fluorescent yield of that element. (Eq. 1) An element of high fluorescent yield is a good candidate to undergo fluorescent excitation.

$$\text{Fluorescent yield} = \frac{\text{No. of x-rays emitted}}{\text{No. of vacancies produced}} \quad (1)$$

As indicated in Fig. 1 the fluorescent yield increases as Z of the target element increases. The fluorescent process, since it occurs due to a photo electric interaction with a bound electron is highly energy dependent. In Figure 2 is plotted the cross-section for creating a K or L vacancy versus the energy of the incident photon. As the graph indicates when the energy of the incident radiation is vastly different from the energy difference between orbital levels, the cross-section decreases rapidly. Where photo-electric absorption does occur, production of the K-shell vacancy predominates, L-vacancies occurring with significant intensity in the heavier elements.

In the practical application of radioisotope induced fluorescence the production of radiation is not the only consideration. Secondary, interactions of the emitted photon with the host material must be considered. In heavy elements the intensity of the emitted radiation will be appreciably attenuated and in any case scattering of the emitted x-ray photons will occur, affecting the sensitivity of the method. These problems will be treated separately as they occur in a particular application.

Figure 1. Fluorescence yield vs. atomic number. $\square$, theoretical, from E.J. Callen, Preprint 1963, unpublished; also Bulletin of the American Physical Society, 7: 416(1962). Δ , theoretical, from M.A. Listengartebm Izvestiya Akademii Nauk Armyanskoi SSR, Seriya Fizika, 24: 1041 (1960). $\blacksquare$, theoretical, R.A. Rubenstein and J.N. Spyder, The Physical Review, 97: 1635 (1955). $\circ$, experimental values. $\bullet$, precise experimental values. (Reprinted from Isotopes & Radiation Technology, Vol 7 #4).

Figure 2. Cross section for K- (or L-) shell vacancy formation in several elements as a function of energy. (5)

5

E. Photon Induced Fluorescence With ^{241}Am

As previously indicated the decay of ^{241}Am results in the production of a 59 keV photon and others below 26 keV. Of primary importance is the emission of the 59.6 keV photon, occurring for 36% of all alpha disintegrations, though Np x-rays are also emitted with sufficient intensity to be of use in applications of fluorescence spectroscopy.

Rhodes (3) in an experimental arrangement with a conical 25 mc Am-241 measured the fluorescent emission of various pure target elements. The results indicated in Figure 3, show the fluorescent emission of photons from targets $Z = 25$ to $Z = 68$, K absorption edge 49.7 keV. The discontinuity in the emission between 10-20 keV, is due to the combined effect from the incident 60 keV γ and accompanying Np X-rays.

The data indicates that ^{241}Am can be a suitable K-x-ray excitation source for elements from $Z = 25$ to $Z = 68$. ^{241}Am of course, is most effective as the exciting source for elements with the K absorption slightly below that of the 60 keV photon. In addition, L-x-rays of the heavier elements can be sufficiently excited. (See Fig. 2). In a particular application however, it sometimes is profitable to employ sources of slightly higher energies than the K or L absorption edge in order to diminish the contribution of Compton-scattered x-rays. Figure 4 indicates the range of ^{241}Am along with various other radioisotopes. In Fig. 5 spectral purity and excitation efficiency of K x-rays by ^{241}Am and ^{109}Cd are compared.

Figure 3. Curves of spectral purity and emission as functions of target atomic number and K_{α} x-ray energy with a 2.5 mC Americium-241 primary source. (3)^a

Figure 4 . Most suitable energy regions for various monoenergetic γ - and X-ray sources. In the hatched area coherent and Compton scatterings take place. The shadowed area represents the Compton continuum. Reference 2 .

Fig. 5

Fluorescence excitation efficiency and spectral purity for K X-rays
excited by Am^{241} and Cd^{109}

○ x Experimental points

— Calculated curves

Ref. 6

REFERENCES

1. C.M. Lederer, J.M. Hollander and I Perlman, Table of Isotopes, 6th ed., John Wiley and Sons, New York (1968).
2. C.G. Clayton, "Radioisotope X-Ray Fluorescence Spectrometry," Report Of IAEA Panel, Vienna, May 13-17, 1968,
3. J.R. Rhodes, "Radioisotope X-Ray Spectrometry," Analyst, 91, PP 683-99 Nov. 1966.
4. Leibhafsky, H.A., Pfeiffer, H.G., Winslow, E.G., and Zernany, P.D., "X-Ray Absorption and Emission in Analytical Chemistry," John Wiley and Sons, Inc, New York and London, 1960, PP 9-20, 26-37.
5. F.S. Goulding, and J.M. Jaklevic, "Trace Element Analysis By X-Ray Fluorescence," UCRL-20625, NTIS, 1971.
6. Rhodes, J.R., "Ore and Coal Analysis Using Radioisotope Techniques", ORNL-11C-5.

1. Am-241 GAMMA EXCITED X-RAY SOURCES

a.) Introduction

The range of applicability of a monoenergetic radioisotope photon emitter in fluorescence work is enhanced by utilizing the isotope to excite characteristic x-rays in various pure element targets. The resulting excited x-rays from the target material are then utilized to excite x-rays in lower atomic number elements. The energy range of such sources extends from about 10 keV up to 100 keV.

Since the resulting radiation is to be employed in some further application, the primary radioisotope must be capable of producing within the target a sufficiently high intensity of x-rays. This limits the targets to materials above $Z = 30$, whose K absorption edge is near the emitted primary photon energy. To be of analytic use the resulting secondary radiation must be highly monoenergetic with a negligible background contributions. Am-241 can therefore be employed as the primary source in γ/X sources to efficiently excite characteristic x-rays in targets from $Z = 35$ to about 65.

Table I - Energy Ranges Over Which K X-Rays
Comprise Greater Than 90 Per Cent
of Output Of γ X Sources Using
Appropriate Targets (1)

Radioisotope	Energy Range (keV)
Am-241	20 - 51
Cd-109	8.6 - 19
Gd-153	19 - 77
Eu-155	31 - 77
Sm-145	14 - 32

As noted earlier the addition to the 60 keV photon, the emitted Np x-rays, are also quite effective in exciting suitable target materials. Table I indicates a general range of applicability of γ -x sources for various primary isotopes where the produced K-x-ray comprises at least 90% of the radiation output of the target material. ^{241}Am and ^{153}Gd can be employed to generate x-rays in approximately the same energy region, however, ^{241}Am with a $t_{1/2}$ - 470 years, has an advantage over the shorter lived ^{153}Gd ($t_{1/2}$ = 236 days).

b) Am-241-Source/Target Assemblies

Various source target assemblies and geometries have been presented in the literature (See references 1,2,3,4,5,6,7). The systems all incorporate back scatter geometry to afford x-ray emission spectral purity of over 90%.

Figure 1 (a,b,c,d) illustrate very common configurations utilizing a central conical source irradiating a target cone of a given element. The curves presented in the previous section were obtained with a 7.5 mc Am-241 source in a geometric configuration similar to Fig. 1c with suitable target materials. Table II presents a comparison between the various geometries.

Watts ⁽³⁾ has developed a theoretical expression to calculate expected efficiencies for x-ray production versus γ -incident energy for a point source conical target geometry. Figure 3 illustrates the predicted efficiencies from this model which are within experimentally obtained data. Best results with this particular geometry with ^{241}Am would be for target materials, from $Z = 58 - 68$, and from about 35-40. It is important to consider with this configuration, the source to target distance, detector position and type. Because of the relative high self absorption of ^{241}Am , the actual geometric construction

(a)

(b)

(c)

(d)

Fig. 1 Geometrical arrangements of source, sample and detector. Ref(1)

TABLE II Measurements with Various Arrangements of Source, Sample, and Detector Ref(1)

Geometry ⁺	Crystal diam. (cm)	Sample to source-detector distance d * (cm)	Relative efficiency [†]	% counting rate change within 3 mm about d	Sample area (cm ²) ★
Fig. 2a	4.5	3.5	1	0.4	125
Fig. 2b	4.5	2.4	0.95	0.4	87
Fig. 2c	{ Internal 4.7 External 7.6	1.03	9.2	1.1	28
Fig. 2d	{ Internal 4.7 External 7.6	0.64	8.8	3.8	33

- + Am-241 sources used were supplied by Radiochemical Centre, U.K.
 (i) Flat disc, RCC code AMC.6.
 (ii) "Conical" source¹⁷, with conical shield and Am-241 in 2 mm dia. ceramic sphere. Additional shielding added to approximate geometry of Fig. 2a.
- * Sample at position of maximum counting rate.
- † Counting rate per effective radioisotope disintegration rate, for the same sample with each system.
- ★ Counting rate due to sample of this area is 90 per cent of that from an extended sample.

of the source is very critical. Ansell et al. have reported obtaining 14mc of activity in the 2 mm diameter bead sources used in the S/T assembly (See Fig.4) As noted in Fig. 2, the source and target assemblies are designed around scintillation detectors employed with photomultipliers or proportional counters. To employ the high resolution characteristics of solid state detectors which do not have the sensitive area of the scintillation systems, a new configuration employing an annular source target geometry has been developed. (See Figure 5a,b). These ^{241}Am annular sources are available from several firms. The target element is interchangeable to make it a useful source over a wide energy range. Preparation of such sources was discussed earlier. Annular source target assemblies can be fabricated to obtain much more intense sources than point source/cone geometries.

A comparison of the photon emission characteristics of a ^{147}Pm and ^{241}Am sources in this type configuration is illustrated in Fig. 6. The output spectrum of a 50 mCi Am/Te source target assembly (Fig. 5b) measured by a Si(Li) detector is depicted in Fig. 7. The spectral purity of the excited radiation is vividly demonstrated.

FIG.2. Geometrical arrangement used in calculation of K X-ray output from conical shell of target material.

FIG.3. Efficiency of the γ -ray excited X-ray source vs. atomic number of target material for various energy γ -rays.

Fig. 4. Conical Source. Ref(4)

Figure 5a Source-target assembly(5)

Fig. 5b Source-target assembly
(dimensions in mm.) (6)

Fig. 6. K Absorption Energy (kev) of Target Material
(Ref (6))

Figure 7. X-ray spectrum of 50 μ Ci Am-241/Te assembly
measured on Si(Li) detector Ref. (6)

REFERENCES

1. Watt, J.S., "Some Recent Developments in Low Energy X and Gamma Ray Sources and Applications in Australia," ORNL-11C-10, PP 665.
2. Watt, J.S., "The Use of Gamma Ray Excited X-ray Sources in X-ray Fluorescence Analysis," International Journal of Applied Radiation and Isotopes, Vol. 18, PP 383-391, 1967.
3. Watt, J.S., "Gamma Ray Excited X-ray Sources," Int. J. Appl. Radiat. Isotop., 15(10); PP 617-25, 1964.
4. Ansell, K.H. and Stevenson, "The Development of Low Energy Photon Sources In the United Kingdom," ORNL-11C-10, PP 764-783.
5. Giaque, R.D., "A Radioisotope Source-Target Assembly For X-ray Spectrometry," Analytical Chemistry, Vol. 40, No. 13, Nov. 1968.
6. Ansell, K.H. and Hall, E.G., "Recent Developments of Low Energy Photon Sources," Applications of Low Energy X and Gamma Rays, P 360, Zeigler, editor, Gordon and Breach, New York, 1970.
7. Pressly, R.S., "Design and Use of Low Energy Gamma and X-ray Sources," P 380, Ziegler, editor, Gordon and Breach, New York, 1970.

2. X-ray Fluorescence Spectrometry:

a. Introduction

Several review papers consider the various facets of fluorescence spectroscopy systems over a wide field of applications (1), (2), (3), (4). A concise, comprehensive treatment of most is contained in the cited review by the IAEA panel. A general system is depicted in the block diagram in Figure 1. The choice of source, energy and activity, detector, geometrical configuration and readout system depends on the specific application.

Fig. 1. Schematic of X-ray spectroscopy systems

b.) General Discussion of Analysis Systems

The general objective of fluorescent spectroscopy is to produce characteristic x-rays within a sample material to obtain some knowledge of the elements therein by the detection of the excited characteristic radiation.

As outlined in the initial section the maximum yield of fluorescent x-rays is obtained by choosing the energy of the exciting photon just above the K-absorption edge of the target element. The simple yield equation

however, only applies when the target is composed of a homogenous material. The effects of a non-fluorescent matrix, the physical state of the matrix (powder, solution, crystal) other fluorescent elements present in the sample, especially those that are of neighboring atomic number and the physical aspects of the sample material, such as particle size and composition must be considered in most applications in order to select the energy of the incident radiation to obtain the maximum sensitivity for a particular element determination.

An appreciation of the difficulty an investigator ordinarily encounters in correlating the K-x-ray yield from a particular element within a sample to its concentration is represented in Fig. 2, where the variation in yield is depicted as a function of differing matrices.⁽³⁾ If the sample is a powder, particle size and composition add to the complexity. Various investigators have done much work in this area and models have been developed to account for the variation of fluorescent yield of particular target samples (Berry^(5,6) Carr-Brion⁽⁷⁾). The proper choice of exciting energy can compensate or overcome some of the troublesome effects. Carr-Brion⁽⁷⁾, for example, suggests that though the lower limit of detection is achieved by

Fig. 2 Intensity - concentration relationship for tin(IV) oxide in different matrices: A, silica; B, iron(III) oxide; C, tungsten trioxide; and D, lead(IV) oxide

employing an exciting energy that just can be separated from the characteristic x-rays of the wanted element by employing the energy resolving power of the detector, heterogeneity effects may be reduced by the use of the highest energy characteristic x-rays and inefficient excitation. For some applications complex computer systems have been developed to overcome matrix effects utilizing photon absorption methods along with XRF. To date no general theory exists for predicting the x-ray yield from a heterogeneous sample, and the choice of exciting energy to obtain the maximum sensitivity can only be determined for particular applications.

In addition to the above mentioned studies, the reader can obtain an overview of the effects of particle size and composition, matrix effects on x-ray yield from the various reviews on x-ray fluorescent spectroscopy (Rhodes (2) (4), Carr-Brion and Payne⁽³⁾, and the IAEA panel report. Most of these effects arise in the type of samples encountered in ore assay and mineralogical applications. In-depth studies have been done on these particular applications. (See later sections).

Once selection of the proper exciting energy is made, the investigator must then determine a suitable detector system. Again many factors other than the energy of the characteristic radiation must be considered. Ruggedness, portability, simplicity of operation and stability are as important to a particular application as the ability of a system to efficiently detect the produced characteristic radiation.

Primarily, though, the detector system must be sufficiently sensitive to the produced characteristic radiation and capable of differentiating between these x-rays and scattered primary and secondary radiation.

The problem is aptly illustrated in Fig. 3, typifying a spectrum from a sample excited by a low energy photon, such as from ^{241}Am .

The objective is to identify the characteristic x-rays superimposed on the background noise. Detector systems (including detector and accompanying analyzing electronics) must properly discriminate against the background radiation and provide the investigator an output dependent upon the excited fluorescent radiation. Generally the type of detectors employed in radioisotope x-ray spectrometry fall into four categories; various types of proportional counters, scintillation detectors (NaI), ion chambers and solid state detectors. Use of crystal diffraction spectrometers is not feasible due to the low-photon output of the radioisotope sources. The final choice, again, is warranted by the application.

Most analysis work involves samples containing various elements, often times of neighboring atomic number. The detector system must then be capable of not only discriminating against scattered radiation but be able to resolve, the different characteristic x-ray peaks (See Fig. 3).

In many systems this is accomplished by employing x-ray filters in conjunction with scintillation detectors. The filters are materials interposed between the detector and target sample, to ~~absorb selectively~~ emitted radiation below a certain energy utilizing the K absorption edge behavior. Two such materials are employed of neighboring atomic number and yield a energy window for incident radiation. (Fig. 4). Metals, plastics and epoxy materials have been employed in this manner (IAEA Panel Report ^{(2) (3) (4) (9) (10) (11)}).

The rapid advances in the past 5 years in the application of radioisotope x-ray fluorescent spectroscopy has been sparked by the use of solid

Figure 3. . Idealized spectrum observed by an X-ray fluorescence spectrometer.

Fig. 4. Principle of balanced filters - (II)

state detector (Si(Li) and Ge(Li)) together with pulse height analyzer systems⁽¹²⁾. These detector systems, though having low efficiency compared to scintillation counters and thus requiring higher exciting source strength do have excellent resolution capabilities (See Fig.5 and Fig.6) and make possible simultaneous detection of elements of neighboring atomic number.

A typical arrangement is depicted in Fig.7 taken from a patent application assigned to Bowman, Thomson, Hyde, Jared⁽¹³⁾. The detectors either Ge(Li) or Si(Li) require cryogenic cooling and do restrict system portability as compared to those employing scintillators or proportional counters. In recent years many workers have studied the use of such systems and have opened the field of radioisotope x-ray fluorescent spectrometry to encompass a broad range of applications.^(12 - 21)

Such systems are becoming laboratory analytical instruments in trace element analysis studies and with the advent of large area detectors⁽¹⁶⁾ with increased efficiency, will greatly enhance the application of radioisotope induced fluorescence.

A very informative comparison of the various type detectors was completed by Burkhalter and Campbell⁽¹⁸⁾ in conjunction with the development of a system to analyze low grade silver and gold ores. A summary of their results as of 1967 is depicted in Table I, The reader must be aware that solid state detector technology has made great strides since that time, the response time, for instance is on the order of nanoseconds.

The selection of detector system and primary exciting energy cannot of course be independently selected. A choice in a particular energy

Figure 5. Comparison resolutions of various x-ray detectors (IAEA Panel Report).

FIGURE 6
RESOLUTION AS A FUNCTION OF COUNT RATE FOR TWO DIFFERENT STATE-OF-THE-ART Si X-RAY SPECTROMETER SYSTEMS: Notice that the count rate scale is logarithmic which tends to compress the data at high rates Ref.(12)

H. R. BOWMAN ET AL.
SEMICONDUCTOR X-RAY EMISSION SPECTROMETER.

Filed Sept. 16, 1966

Fig. 1

INVENTORS
HARRY R. BOWMAN
STANLEY G. THOMPSON
EARL K. HYDE
RICHARD C. JARED
ATTORNEY

Table I Comparison of X-ray detectors.

Gas Proportional Detector		Scintillation Detector		Semiconductor Detector	
Advantages					
Good detection efficiency	>70%, 2-15 kev range, insensitive to high energy radiation	High detection efficiency	>90% for X-ray range >5 kev	High detection efficiency	Near 100% if X-ray completely stopped in depletion depth
Moderate pulse resolution	Dispersion power Z^2j at 2σ level	Short decay time	0.25 μ second NaI(Tl)	Excellent energy resolution	Dispersion power better than Z^2j at 2σ with small detectors
Excellent energy linearity	Entire X-ray range	Size	Photomultiplier tubes with diameters $\frac{1}{2}$ inch available for drill hole probes	Linear energy response	Excellent energy linearity
Short rise time	Electron collection time $\sim 0.2 \mu$ second	Rugged	Ruggedized photomultiplier tubes	Negligible background	Excellent signal-to-noise
Low background	1-2 counts/minute	Moderate cost	\$500-\$1000	Fast rise times	Several hundred nano-seconds
Good stability	Few spurious pulses				
Size	Wide variety of shapes and sizes				
Rugged	Rugged detectors available				
Low cost	\$300-\$500				
Disadvantages					
Space charge	Large pulse shifts with X-ray intensity has restricted use of proportional detector for energy dispersion analysis	Poor pulse resolution	5 times poorer than gas and 6-7 times poorer than Si semiconductor detectors	Temperature sensitivity	Cryogenic conditions restrict versatility
Escape peaks	Magnitude dependent on gas used and detector design	Energy linearity	Poor below ~ 40 kev	Size	High detection efficiency limited to <40 kev, loss of pulse resolution with larger size detectors
		Temperature sensitivity	Ordinary photomultiplier tube limited to 85°C	Cost	Large detectors are expensive, \$1000-\$3000
		Handling	NaI extremely hygroscopic, photomultiplier tube can be damaged by overvoltage	Low output signal	Low noise amplifiers necessary
		High background	1-10 counts/second, poor signal-to-noise at low energy	Stability	Not stable for long storage times at ambient temperatures

region of soft characteristic x-rays may deem it necessary to employ proportional counters rather than solid state detectors. The geometric configuration of source-target detector is also of the utmost importance in order to achieve maximum intensity of excited characteristic radiation, minimize the detection of the primary radiation and thus achieve a low limit of detection. Spectrometers with proportional or scintillator counters employ the central source configuration to achieve maximum benefits. Fig. 8, a, b, c, depicts variations on this geometry as per particular requirement.^{(22) (23)}

To offset the decreased efficiency of solid state detectors, extended area sources of high intensity have been developed using annular and line construction and the geometries as depicted in Fig. 9 (a,b). As pointed out by Jones and Carpenter⁽²⁵⁾ such annular configurations have the added advantages of providing uniform flux distribution over the surface of the sample and are basically insensitive to small geometric variations.

Kneip and Laurer⁽²⁶⁾ have also shown that the concentric source geometry, Fig. 9b, can be employed to attain a factor of 25 or greater sensitivity than the annular configuration. Source intensities for these types of source configurations are on the order of tens of curies. (See section on photon sources).

For the excitation of low Z elements source target assemblies have been developed to be employed with solid state detector systems (See section Am-241- γ /X sources) utilizing annular sources in a backscatter geometry, (Fig. 10).

As the reader should now appreciate the particular choice of radioisotope spectrometer system for an application must be a well thought out decision weighing all facets of the type of investigation to be performed. Solid state systems may not be appropriate where an inexpensive

Fig. 8 Source, sample, and counter arrangements for X-ray fluorescence analysis: (a) compact; (b) double backscatter; (c) noncompact.

Figure 9. Typical geometries employed with solid state detector systems.

Figure 10. S/T assembly for solid state detector systems (Reference 27).

REFERENCES

1. Cameron, J.F. and Florkowski, T., "Radioisotope Sources of Low Energy Electromagnetic Radiation and their Use in Analysis and Measurement of Coating Thickness," ORNL-11C-5.
2. Rhodes J.R., Futura, Hall J.D., "X-ray Analysis Using Radioisotope Sources" Contra AT-(40-1)-3224, July 14, 1967.
3. Carr-Brion K.G., Payne, K.W., "X-ray Fluorescence Analysis," The Analyst, Dec. 1970, Vol. 95, No. 1137 PP 997-991.
4. Rhodes, J.R., "Radioisotope X-ray Spectrometry", The Analyst, Nov. 1966, Vol 91, No. 1088 PP 683-699.
5. Berry, P.F., "A Study of Particulate Heterogeneity Effects In X-ray Spectrometry", OR003847-2.
6. Berry, P.F., "Particle Size Heterogeneity Phenomena in X-ray Analysis", Applications of Low Energy X and Gamma Rays C.A. Ziegler (ed) Gordon & Beach, N.Y. 1971.
7. Carr-Brion, K.G., "The Selection of Exciting Energy in Radioisotope X-ray Fluorescence Analysis", The Analyst, March 1969, Vol 94, PP 177-181.
8. Reproduced from "Trace Element Analysis by XRF" F.S. Goulding and J.M. Jaklevic, May 1971, UCRL-20625.
9. Dunne, J.A. and Nickle, L.L., "Balanced Filters for the Analysis of A, Si, K, Ca, Fe, and Ni," Proceedings of 2nd Symposium on Low Energy X and gamma Sources and applications, Austin Texas, March 27-29, 1967, ORNL-11C-10.
10. Rhodes, J.R., Morgan, I.L., Futura, T. "A New Method for X-ray Energy Selection Using a Scintillation Counter and Balanced Filters", Applications of low Energy X and Gamma Rays, Edit. C.A. Ziegler, Gordon and Breach (N.Y. 1971).
11. Broquet, C., Robin, G., Vacher, M., "Realization and Use of X-ray Filters for Energy Selection," ORNL-11C-10, P 356.
12. Walter, F.J., "The Impact of Semicountors on X-ray Spectroscopy," IEEE Transactions on Nuclear Science NS-17, No. 3 (June 1970).
13. Semi Conductor X-ray emission Spectrometer Patented March 18, 1969, Pat. No. 3,433,954, Bowman et al.

14. Sandburg, A.O., "Energy Deispersion X-ray Analysis with electron and Isotope excitation" ASTM Special Publication 4850-48500-39. Presented at 73rd Annual Meeting ASTM, Toronto, Canada, June 21-26, 1970.
15. Horth, R.L., Frankel, R.S., Gehrke, R.J., Barstow, J. "Energy Dispersive X-ray Spectrometry with Dedicated Computer Data Reduction,"
16. Frankel, R.S., Atkin, D.W., "Large Area Silicon X-ray Detectors," Applications of Low Energy X and Gamma Rays, C.A. Ziegler, editor, Gordon and Breach, N.Y., 1971.
17. Heath, R.I. "Review of Solid State Detector Systems for X-ray Spectrometry) ORNL 11C-10, P 465.
18. Jones, B.J., Carpenter, R.A., "X-ray Fluorescent Analysis of Multi-Component Samples by a Non-Dispersive System," ORNL-11C-10.
19. Bowman, H.R., Hyde, E.K., Thompson, S.G., Jared, R.C., "Application of High Resolution Semiconductor detectors in X-ray emission Spectrography," Science, Vol. 51, P 562, Feb. 4, 1966.
20. Hyde, E.K., Bowman, H.R., Sisson, D.H., "Analytical Capabilities of Semi-Conductor X-ray Emission Spectrometer," UCRL-16845, May 1966.
21. Burkhalter, P. and Cambell, W.J., "Comparison of Detectors For Isotopic X-ray Analyzers", ORNL-11C-10.
22. Cameron, J.F. and Florkowski, T., "Radioisotope Sources of Low Energy Electromagnetic Radiation and Their Use in the Analysis and Measurement of Coating Thickness," Source Configurations Described, ORNL-11C-5.
23. Figures reproduced from Isotopes Radiation Technology, Vol.3, No. 3.
24. Bablan, R.J., Brill, A.B., Datton, J.A., "Applications of Von Dispersive X-ray Fluorescent Techniques for In Vitro Studies," IEE Transactions on Nuclear Science, NS-20, No. 1 , P 385, Feb. 1973.
25. Jones, W.B., Carpenter, R.A., "Nondispersive X-ray Fluorescent Spectrometer," Advances in X-ray Analysis, Vol. II, P. 215, (N.Y. 1968).
26. Kneip, T.J., Laurer, G.R., "Isotope Excited X-ray Fluorescences," Anal. chem, 44 No. 14, Dec. 1972.
27. Goulding, F.S., Jaklevic, "Trace ELEMENT Analysis By X-ray Fluorescence," UCRL-70625, May 1971.
28. Kobayuski, Sugita, Takayanagi, Iio and Sasaki, "GaAs Biomedical Probes and Their Applications in Nuclear Medicine," P 310, NS-20, No. , Feb. 1973.

3. Am-241 X-Ray Fluorescent Spectroscopy Applications

a. Introduction

The purpose here will be to report on the particular applications of ^{241}Am in radioisotope induced fluorescence reported in the literature.

For a thorough review involving the general applications of radioisotopes the reader is referred to the IAEA Panel Report and numerous other review articles. Situations concerning other sources will only be made in so far as they had been compared with possible ^{241}Am excitation.

b. Ore Analysis-- In Field Assay Techniques

Zinc in Zinc Ores

Utilizing a portable analyzer with central source geometry, Rhodes⁽¹⁾ reports that concentrations of copper and zinc in ores greater than 1% can be determined by employing primary radiation from $^3\text{H/Zr}$ ^{109}Cd , ^{241}Am or $^{197}\text{Pm/Al}$

bremsstrahlung and balanced filters for energy selection. For sensitive analysis, ^{109}Cd ^{(2) (3)} is reported to be the most efficient exciting source, utilizing nickel and cobalt balanced filters for energy selection, to discriminate against Cobalt (up to 2%) concentration and Iron x-rays (.5 to 7%) ordinarily present the ore matrix. With ^{109}Cd the scattered radiation is confined to a range between 20-22 keV and therefore is well resolved by a proportional counter from the fluorescent x-rays (approximately 8 keV).

Silver in Silver Ores

Introduction

Extensive work has been done in applying radioisotope x-ray fluorescent methods to the determination of silver in silver ores. Rhodes^{(4) (5)} and Burkhalter^{(6) (7) (8)} have performed investigations in this area considering a variety of exciting sources and the utilization of portable analyzers with

scintillation detectors in addition to solid state detector systems.

Silver Ore Mineralogy and System Requirements

Silver in silver ores ranges from 1000 ppm to an average of 35 ppm in a low grade ore mined in the United States. Spectroscopic systems must then have a limit of detection below this value to be useful. The ore matrix is frequently quartz or composed of materials similar in atomic number to quartz. The elements Fe, Cu, Zn, As, Sb, Bu, and Pb are commonly associated with the presence of silver in significant concentrations with, ppm concentration of elements like Sr, Zr, Mo, and Cd.⁽⁶⁾ An accurate determination of silver in silver ores therefore, is complicated by absorption of the excited radiation within the matrix interference from the excitation of Ba-K α X-rays (32 keV) and varying concentration of the accompanying elements which would vastly change matrix absorption.

Systems Employed and Results

The feasibility of a portable analyzer system utilizing a Na(I) detector system with single channel analyzer was examined by Burkhalter and Rhodes. Burkhalter employed the configurations depicted in Fig(1) with the following designated sources:⁽⁶⁾

- (a) 14 mCi source of ^{241}Am
- (b) A central source target assembly with Sb.
- (c) The emitted x-rays of Tellurium from ^{125}I , together with the 35.5 α - photon.
- (d) $^{147}\text{Pm}/\text{Al}$ Bremsstrahlung source.

Rhodes⁽⁴⁾ compared various ^{241}Am S/T assemblies (I, Te, Sb) in the geometry depicted in Fig. 1(a) and found $^{241}\text{Am}/\text{Sb}$ to be the most sensitive exciting source.

FIG. 1. Geometrical arrangements of source, sample and detector.

Table I⁽⁶⁾ is a summary of the detection limits normalized to source excitation activity for silver in a 1000 ppm sample of silver in a prepared silver matrix. (Note that θ is the scatter angle between the source of radiation and the axis of the detector in the geometry depicted in Fig. 1b. Burkhalter measured the intensities from his prepared silver loaded silica sample, then obtained an intensity measurement from pure silica to obtain the ratio of I_{Ag}/I_b , eliminating any matrix effects. Table II gives the results from prepared samples with 1000 ppm silver doped with various elements present in natural silver ores. The effect on the intensity of the emitted characteristic AgK α radiation is indeed pronounced.

Rhodes⁽⁴⁾ developed a method for overcoming matrix effects by the use of another S/T assembly in the head of the portable analyzer in conjunction with $^{241}\text{Am}/\text{Sb}$. The method, initially proposed by Watt⁽⁷⁾, measures the absorption coefficient within the matrix of Compton scattered radiation from a source-target assembly whose target characteristic radiation is in a suitable energy region but cannot excite the element of interest. Rhodes employing a $^{241}\text{Am}/\text{Ag}$ assembly obtained the results depicted in Fig. 2 and 3. The dual head method precludes use as a "portable" device but can definitely be employed in "on stream" or process analysis of slurries. Rhodes concluded that for a portable instrument employing a scintillation counter and single channel analyzer, $\text{Pm}^{147}/\text{Al}$ was the best available source and matrix effects can be compensated for by measuring the intensity of the scattered intensity raised to the n th power, n being arbitrarily chosen to eliminate the matrix effect between two different values of matrix composition at zero percent silver. (See Fig. 4).

TABLE I. Peak-to-background ratios and normalized detection limits for 1000 ppm silver in a silica matrix

Source	Radiation	ϕ deg	I_{Ag}/I_b	Detection limit (normalized), ppm
^{241}Am	Sb K	90–120	0.095	21
^{241}Am	Sb K	$130 \pm 15\%$	0.13	18
^{125}I	Tc K	90–140	0.175	16
X-Ray generator	Ba K	$165 \pm 5\%$	0.18	15.5
X-Ray generator	Ba K	$130 \pm 10\%$	0.265	13
$^{147}\text{Pm}/\text{Cs}$	brems	110–150	0.085	22
$^{147}\text{Pm}/\text{Al}$	brems	110–150	0.095	21

TABLE II. Intensity measurements, silver intensities, peak-to-background ratios, and detection limits for 5-percent heavy-metal additions in silica matrices

Sample matrix plus 1000 ppm Ag	N_{Ag} c/s	I_b c/s	I_{Ag} c/s	I_{Ag}/I_b	Detection limit, ppm	I'_{Ag} c/s
SiO_2	18,350	16,150	2200	0.14	17	2200
$\text{SiO}_2 + 5\% \text{ Fe}$	12,350	11,110	1240	0.11	25	–3800
$\text{SiO}_2 + 5\% \text{ Zr}$	15,770	14,400	1370	0.10	25	–380
$\text{SiO}_2 + 5\% \text{ Sb}$	25,980	24,990	990	0.04	45	+9830
$\text{SiO}_2 + 5\% \text{ Ba}$	12,740	10,830	1910	0.18	16	–3410
$\text{SiO}_2 + 5\% \text{ Pb}$	6760	5880	880	0.15	25	–9390

FIGURE 2
CALIBRATION CURVE USING ARTIFICIAL SILVER ORE
SAMPLES AND ^{241}Am - Sb SOURCE TARGET
ASSEMBLY

Ref.(4)

FIGURE 3

PLOT OF "FLUORESCENT / SCATTER" RATIO USING
 ^{241}Am - Sb AND ^{241}Am - Ag SOURCE-TARGET ASSEMBLIES

Figure 4. $KI_f/(I_s)^{2.16}$ against silver content, illustrating partial elimination of the matrix absorption effect.

With this method, employing $^{147}\text{Pm}/\text{Al}$, Rhodes attained detection limits of 32 ppm, With the dual source target assembly, in a 100 second count time 26 ppm sensitivity was attained, at 30 sec, 47 ppm.

Burkhalter⁽⁸⁾ continued his study of prepared 1000 ppm silver samples employing Si(Li) and Ge(Li) detectors with exciting sources of $^{241}\text{Am}/\text{Sb}$, I^{125} , $^{241}\text{Am}/\text{I}$, and $^{147}\text{Pm}/\text{Cs}$, utilizing the procedure in his earlier work. Fig. 5 and Fig. 6 illustrate the output from the two systems providing the best resolution. A summary of results is presented in Table III Rhodes^{(4) (5)} employing a 30 mm² 2 mm thick Si(Li) detector and compensating for its low efficiency with makeshift geometry, Fig. 7 , together with a $^{241}\text{Am}/\text{I}$ source target assembly obtained detection limits of 4 ppm accumulating 10^6 counts in 2 hours; twenty-two parts per million sensitivity was attained in 4 minutes. With a 150 mc Source, a 4 ppm sensitivity is predicted in 15 minutes.

Rhodes reported on the analysis of a variety of natural silver ores with the $^{241}\text{Am}/\text{I}$, Si(Li) system^{(4) (5)}. The ore sample obtained from the Bureau of Mines were first ball-milled to minus-32 mesh to eliminate particle size effects and examined as dry powder samples. Table IV represents the results; fig. 8 is a typical spectra.

As the pulse height spectra indicates the BaK x-rays are sufficiently suppressed overcoming any appreciable enhancement effects. The presence of lead however greatly attenuates the AgK_α radiation (Note: the Bunker Hill sample). In an effort to compensate for matrix effects Rhodes takes a ratio of the fluorescent to the Compton scattered radiation. Fig.9 illustrates these results.

FIG. 5 Silver and background spectra obtained with I excitation and Si (Li-drifted) semiconductor detector.

FIG. 6 Silver and background spectra obtained with Cs excitation and Si (Li-drifted) semiconductor detector. Ref (8)

FIG.7
ARRANGEMENT OF SOURCE-TARGET ASSEMBLY, COOLED Si(Li)
DETECTOR, AND SILVER ORE SAMPLE (REF.5)

TABLE III

**X-RAY INTENSITY MEASUREMENTS FOR 1000 PPM SILVER IN SILICA AND
CALCULATED DETECTION LIMITS BASED ON 100-SECOND COUNTING INTERVALS (Ref.8)**

Detector	Source	Radiation	I_t c/s	N_{Ag} c/s	I_b c/s	I_{Ag} c/s	I_{Ag}/I_b	Detection Limit, (ppm)	Detection Limit (Normalized) (ppm)
Silicon	$^{241}\text{Am}/\text{Sb}$	Sb K	200	7.2	3.0	4.2	1.4	150	5.7
Silicon	^{125}I	Te K	4400	230	64	166	2.6	21	4.8
Silicon	$^{241}\text{Am}/\text{I}$	I K	200	7.05	1.75	5.3	3.0	110	4.7
Silicon	$^{137}\text{Ba}/\text{Cs}$	Cs K	300	9.7	3.1	6.6	2.1	110	5.1
Germanium	^{125}I	Te K	350	18.8	4.5	14.3	3.2	70	4.6

TABLE IV

Table
(5)

Natural Silver Ores, Properties and Results

No.	Description		Silver Oz/Ton	Pb %	Zn %	Cu %	Fe %	SiO ₂ %	I _F (C/S)	I _F /I _S	Remarks on Matrix
1	Ward	D. Nev. 35.1	2.6	6.2	1.0	0.06	21.1	36.5	2.76	0.116	Heavy; high iron lead ore
2	Badger	Mont. 70.2	3.0	0.4	4.6	0.26	3.9	61.0	4.19	0.111	Light; quartz
3	Flathead	Mont. 68.1	3.8	0.8	0.05	0.07	2.8	78.3	5.38	0.114	Light; quartz
4	Orphan Girl	Mont. 70.1	4.4	4.8	2.0	0.05	2.3	75.3	3.85	0.120	Fairly heavy; lead ore
5	Robinson	D. Nev. 34.1	6.3	1.3	0.05	0.0	4.2	83.4	5.35	0.120	Light; quartz
6	Mammoth No. 6	Dut. 56.2	12.6	3.6	1.4	2.1	21.2	27.9	3.66	0.143	Heavy; high iron
7	Taylor No. 2	D. Nev. 36.2	13.9	0.4	1.2	0.06	0.3	1.2	7.02	0.167	Medium; carbonate ore
8	Taylor No. 1	D. Nev. 36.1	14.1	1.3	0.7	0.4	1.2	52.1	7.02	0.143	Light; quartz
9	Mammoth Raise	Dut. 56.4	15.8	2.8	0.1	0.08	5.0	67.8	5.28	0.154	Fairly heavy
10	Mammoth No. 5	Dut. 56.1	15.9	0.45	1.5	2.4	32.8	16.1	3.72	0.152	Heavy; high iron
11	Crescent	Id 44.2	17.2	0.05	0.05	0.34	15.9	53.4	5.67	0.155	Fairly heavy; high iron
12	Baker Property	Colo 119.1	17.4	0.4	0.4	0.1	3.6	35.2	4.53	0.185	Heavy; barium ore containing 26.6% Ba
13	Mammoth No. 14	Dut. 56.3	19.9	1.4	0.2	0.09	4.4	68.6	6.96	0.170	Light; quartz
14	Bunker Hill	Id 44.1	25.1	27.9	3.65	0.05	7.0	32.4	3.23	0.173	Very heavy; high grade lead ore
15	Escalante	Dut. 55.1	26.7	0.5	3.9	0.05	3.9	78.6	8.27	0.197	Light; quartz

FIG 8 SPECTRUM FROM BAKER PROPERTY SILVER
(5) ORE, EXCITED BY $^{241}\text{Am}/\text{I}$ AND
DETECTED BY $\text{Si}(\text{Li})$

FIG. 9 Ag K SCATTERED RATIO AS A FUNCTION OF SILVER CONTENT OF NATURAL ORES. SOURCES:— $^{241}\text{Am}/\text{I}$.

Comments

With the development of more efficient Si(Li) detectors and greater source loading of ^{241}Am in extended area sources such as annular rings, a sensitive analysis of silver in silver ores can be obtained using a $^{241}\text{Am}/\text{I}$ target assembly. Matrix effects, especially dealing with the inherent heavy element attenuation, do need further study. Scintillator counter portable systems employing Pm/Al for in field determination provide the best answer for that purpose. The dual target $^{241}\text{Am}/\text{Sb}$ - $^{241}\text{Am}/\text{Ag}$ method is well suited for process control or on stream analysis of silver ore slurries.

Tin in Tin Ores

As reported by Rhodes⁽¹⁾⁽¹⁰⁾, ^{241}Am and $^{241}\text{Am}/\text{Cs}$ were compared with ^{153}Gd , and $^{147}\text{Pm}/\text{Al}$ bremsstrahlung radiation as excitation sources with Ag and Pb balanced filters for the analysis of tin in tin ores, utilizing a portable analyzer system with a scintillation counter and rate-meter (Hilger and Watts). The results obtained indicated that for this system $^{147}\text{Pm}/\text{Al}$ was the most sensitive excitation source, (detection limits - .03% tin in dry powdered samples, .05% in field assay determinations). With the $^{241}\text{Am}/\text{Cs}$ assembly (4 mCi, ^{241}Am) a detectable limit of 90 ppm tin in dry powder was achieved and .03% tin in solids (10% solids) from artificial sample slurries. Increased source activity to 20 mc, can increase the sensitivity to 40 ppm and .014% respectively. With the target assembly a compensation method was studied to overcome inter-element effects and compared to the balanced filter technique used with the single excitation sources. The compensation

method involves the monitoring of the intensity of characteristic radiation from an interfering element present in the matrix. (For further discussion of Tin Ore Analysis, see "On Stream" applications).

Heavy Element Ore Bearing Samples

Introduction

Ordinarily in the case of heavy element analysis, with x-ray fluorescence equipment, it is L x-ray excitation that is employed, rather than K x-ray fluorescence. The use of the K x-ray excitation does however eliminate some of the problems encountered with the excitation of the softer L x-rays. Kunzendorf and Wollenberg⁽¹¹⁾, and recently Kuusi et al.⁽¹²⁾ have reported the use of ^{241}Am , together with a solid state detector system to determine the presence of rare earth elements to the 10 ppm level with sufficient accuracy (approximately $\pm 6\%$).

Methods and Results

Kunzendorf et al., were interested in the determination of the presence of the rare earth elements La, Ce, Pr, and Nd in unprepared flat rock samples. The 60 keV photon of ^{241}Am was determined as a suitable source of excitation in this region (See Table V). The target rock sample and source were set up as indicated in Fig. 9. The Ge(Li) detector had an active area of $.9 \text{ cm}^2$ with a resolution of 1 keV; the source - a 30 mCi disc configuration. The auxiliary electronics included a 1024 P. H. A. A sample spectrum of an irradiated rock sample is shown in Fig. 10 (Background subtracted).

To determine the content of the element under investigation they applied a least squares method to an expression describing the x-ray spectrum of the four elements.

$$Y_t(E)dE = \sum_{j=1}^4 f_j \left(\sum_k a_{jk} \exp[-(E - E_{jk})^2/2\sigma^2] \right) dE \quad (1)$$

where

$$k = k_{\alpha_2}, k_{\alpha}, k_{\beta_1}, k_{\beta_2},$$

and to the measured spectrum $Y_m(E)dE$, (background subtracted), with the aid of a computer calculated the fractions, f_j , of the element present. (See Eq. 1). A comparison between a calculated spectrum and a portion of actual spectrum is shown in Fig. 11. Background measurements were accomplished utilizing a like mineral compound, (arfvedsonite) containing no rare earths, in place of the sample, and subtracting this spectrum from the sample spectrum. X-ray determinations of the four elements in samples were compared to mass spectroscopic data and agreement within a 95% confidence level was attained. The results of a scan of a longitudinally cut drill core are illustrated in Fig. 12.

Kuusi⁽¹²⁾ et al. employed Co-57 in most of their investigative work to determine the presence of lead, cerium, tungsten and molybdenum in several types of samples using K x-ray excitation techniques. They found however, that ^{241}Am would be superior to Co-57 in cerium determination. A Si(Li) solid state detector system was employed in conjunction with a 30 mCi, ^{241}Am disc source. Fig. 12 and Fig. 13 are calibration curves for ascertaining the concentration of cerium in drill core material and process water samples, respectively.

TABLE V

K X-ray energies, normalized intensities and fluorescence yield ω_K for La, Ce, Pr and Nd.

Z	Element	$EK\alpha_2$ (keV)	$EK\alpha_1$ (keV)	$EK\beta_1$ (keV)	$EK\beta_2$ (keV)	ω_K
57	La	33.03 (27.96%)	33.44 (53.75%)	37.8 (15.05%)	38.7 (3.23%)	0.89
58	Ce	34.28 (27.81%)	34.72 (53.48%)	39.2 (15.51%)	40.2 (3.21%)	0.89
59	Pr	35.55 (27.81%)	36.03 (53.48%)	40.7 (15.51%)	41.8 (3.21%)	0.90
60	Nd	36.85 (27.81%)	37.36 (53.48%)	42.2 (15.51%)	43.3 (3.21%)	0.90

Fig. 9. Experimental set-up.

Fig. 10. Rare-earth X-ray spectrum of a sample excited by ^{241}Am , showing positions of characteristic $K\alpha$ and $K\beta$ X-ray energies. The background has been subtracted.

Fig. 11. Part of a measured X-ray spectrum which has been used for the calculation of La, Ce, Pr and Nd in the sample. For comparison the spectrum computed from the calculated fractions (f_i) is superimposed on the measured spectrum.

Fig. 12. The distribution of f_{Ce} and La/Ce over the flat surface of a piece of drill core cut longitudinally.

Fig. 13. Calibration curve established for determining the cerium content of drill core material samples.

Fig. 14 Calibration curve established for determining the cerium content of process water samples.

REFERENCES

1. Rhodes, J.R., "Ore and Coal Analysis Using Radioisotope Techniques," Proc. of Symposium on Low Energy X and Gamma Sources and Applications, P 206, Chicago, Oct. 20-21, ORNL-11C-5.
2. Rhodes, J.R., "Some Examples of Ore and Alloy Analysis Using Multi Purpose PORTable Analyzer", Proc. of 2nd symposium on Low Energy X and GAMMA Sources and Applications", P. 843, Austin, Texas, March 27-29, 1967, ORNL-11C-10.
3. Rhodes, J.R., Ahler, T.G., and Poole, D.O., "Analysis of Zinc and Copper Ores by Radioisotopes," Paper presented at Symposium of Analytical Instrumentation Wind-Seale, Dec. 4-5, 1963.
4. Rhodes, J.R., Furuta, T., Hall, J.D., "X-Ray Analysis Using Radioisotope Sources," AEC Report ORO-3224-12, July 1967.
5. Rhodes, J.R., "Optimization of excitation and Detection Techniques of Silver Ores By Radioisotope X-ray Spectrometry," ORNL-11C-10, P 442, 2nd Symposium on Low Energy X and Gamma Source and Applications.
6. Burkhalter, P.G., "Detection Limit of Silver by Energy Dispersion X-ray Analysis Uisng Radioisotopes," International Journal Applied Radiation and Isotopes, Vol 20, PP 353-362.
7. Watt, J.S., "Some Recent Developments in Low Energy X and Gamma Sources And Applications , In Australia", P 663, 2nd Sympsoium on Low Energy X and Gamma Sources and Applications.
8. Burkhalter, P.G., "Radioisotope X-ray Analytical Techniques for Gold and Silver Ores", Nuclear Techniques and Mineral Resources, IAEA, Vienna, 1969.
9. Burkhalter, P.G., "X-ray Intensity Measurements From Ores Using Semiconductor Detectors and Radioisotope Excitation," Applications of Low Energy X- and Gamma Rays, C. Ziegler (ed), Gordon and Breach, N.Y., (1971), P 147.
10. Rhodes, J.R., Aher, T.G., and Boyce, T.S., "Determination of Tin in Tin Ores By Radioisotope X-ray Flourescence," Radiochemical Methods of Analysis, Vol. II, IAEA Pub. STI/PUB/88, Vienna, 1965.

11. Kunzendorf and Wollenberg, "Determination of Rare-Earth Elements in Rocks by Isotope-Excited X-ray Fluorescence Spectrometry," Nuclear Instruments and Methods, 87, PP 197-203, (1970).
12. Kuusi, J., Virtanen, W., and Jauho, "Heavy Element Analysis By Isotope-Excited X-ray Fluorescence," Nuclear Technology, Vol. 13, Feb. 1972.

c. Mineral Analysis--"On Stream" Methods

Introduction

The preceding section was primarily concerned with the possibility of ascertaining mineral presence with "field" samples or in field analysis. The restriction on the system was portability or semi-portability. For "on stream" applications within the ore processing industry portability is not a factor and multiple measurement techniques to overcome matrix effects can be employed. A general review of such applications can be found in Cameron and Florkowski, 1964⁽¹⁾ and more recently in (Ellis et al, 1969⁽²⁾, Clayton, 1969⁽³⁾ and Carr-Brion and Payne⁽³⁾ (1970)), the IAEA Panel Report; Rhodes⁽⁵⁾ (1971); Papez and Cameron⁽⁶⁾ (1972).

The difficulty in employing X-ray fluorescence techniques to determine element concentration in ore samples or slurries with any degree of sensitivity is the lack of homogeneity in the material under investigation. In most mineral analysis, matrix effects make it impossible to correlate characteristic intensity from the sample with the concentration of an element under investigation. As discussed earlier theories have been developed to predict x-ray yield given variable matrix parameters and some simple techniques like choosing the highest possible exciting energy have been successful.⁽⁷⁾

Rhodes (See previous section) employed a ratio technique and dual
241

Am S/T heads to overcome the effect of Fe_2O_3 in silver determination but, again as he noted this is not a simple field method. Ordinarily it is necessary to employ more complicated measurement techniques to obtain any degree of sensitivity. Ellis and Fookes⁽²⁾ and Carr-Brion⁽⁸⁾ have proposed methods involving multiple measurements (absorption and x-ray fluorescence)

to overcome matrix effects in ore samples. Source/target assemblies such as Am/Cs, Am/Sn, Am/Ba, Am/Sm were found to be efficient exciting sources.

Matrix Correction Methods

Ellis et al. based their method on a simple equation; the emitted characteristic x-ray intensity (I_f) is given by:

$$C = I_f a_1 (\mu_E + \mu_F) \quad (1)a$$

where C is the concentration of the wanted element and (a) is a constant to be determined. Carr-Brion employs an expression with an multiplicative exponential factor. (Notation has been changed from the literature to show similarity).

$$W = \frac{I_f (\mu_E + \mu_F)}{KI_E W (1 - e^{-(\mu_E + \mu_F) \rho X})} \quad (1)b$$

where

- K = Constant depending on element being determined, geometry of the analyzer, etc.
- I_E = intensity of the exciting X-rays
- W = weight fraction of the element in the slurry
- ρ = slurry density
- X = slurry depth

In both equations,

- μ_E = X-ray attenuation coefficient of the slurry for the exciting x-rays.
- μ_F = X-ray attenuation coefficient of the slurry for the characteristic x-rays.

If $(\mu_E + \mu_F) \rho X$ is large (> 5) the exponential term is negligible and the expressions are practically identical. However, W , represents the concentration of the element in the whole slurry not only the solid component (i.e. the ore), thus obtaining W, W_s must still be computed. The C is concentration in percent weight of the contained solids. (See accompanying graphs). The utilization of both expressions requires a measurement of I_f and a value of $(\mu_E + \mu_F)$. The method employed depends on the particular application.

Applications--Results

Zinc

A simple technique investigated by Ellis and his co-workers was to make a single measurement (N_s) with the incident x-ray energy just greater than the K-shell absorption edge of zinc, utilizing a ^{238}Pu Ge γ -X source with a Zn filter. The results illustrated in Fig. 1a are close to that obtained using balanced filters, indicating that most of the error is due to changes in absorption caused by variation in matrix components. Transmission and scatter measurements were then made, the scatter measurements employing an $^{241}\text{Am/Cs}$ γ X source (5 mCi, Am-241) and for the transmission measurements the straight 5 mCi source.

The corrections employed utilizing these measurements are summarized in Table I. Fig. 1b depicts the function of the combined signal employing equation 2e. The samples were prepared by cold pressing 100 g of dry solids mixed with 15 g of paraffin wax into discs. A study of flotation-fed zinc ores was made. The results are reported in Table II. (Note the sensitivity to particle size.) The data indicates that the matrix effects are less for large particle size, and in addition (N_s) measurements may be omitted.

FIG 1a. Single measurement (N_1).

FIG 1. (b) N_1 compensated by both lead concentration and scattered X-ray measurement (N_s).

TABLE I Precision of determination of zinc in ground lead-zinc ore samples using various methods of matrix correction. Mean zinc concentration is 12.7 per cent by weight

Equation	Method of correction	RMS error (% Zn by weight)
2a	$a_1 \cdot N_1 + a_2$	2.38
2b	Balanced filters*	2.30
2c	$\frac{a_1 \cdot N_1}{N_s} + a_2$	1.33
2d	$a_1 \cdot N_1 \cdot \log(N_0/N_t) + a_2$	3.50
2e	$a_1 \cdot N_1 \cdot \left(\frac{1}{N_s} + a_2 \cdot C_{Pb} \right) + a_3$	0.57
2f	$a_1 \cdot N_1 \cdot [\log(N_0/N_t) + a_2 \cdot C_{Pb} + a_4] + a_5$	0.76
2g	$a_1 \cdot N_1 \cdot (N_1 + a_2 \cdot C_{Pb} + a_3) + a_4$	1.54
2h	$a_1 \cdot (N_1 + a_2 \cdot C_{Pb} + a_3) \cdot (N_1 + a_4 \cdot C_{Pb} + a_5)$	1.23

N_1 : Pu-238/Gc γ X source 25 mg/cm² Zn filter.

N_s : Am-241/Cs γ X source, 220 mg/cm² Al filter.

N_t : Am-241 60 keV transmission.

C_{Pb} : Lead concentration.

* Balanced filters: Pu-238/Gc γ X source, Cu and Ni filters.

FIG. 1. Determination of zinc in lead-zinc ore, showing how compensation reduces errors caused by sample matrix variations.

Precision of determination of zinc in lead-zinc mineral products†

Mineral product	Sample preparation	Zinc (% by weight)		RMS error (% by weight)		
		Range	Mean	N_1	$(N_1, C_{Pb})^*$	$(N_1, C_{Pb}, N_s)^*$
Lead-zinc ore	ground	0-34	12.7	2.38	1.23	0.57
Flotation Feed series 1	ground	9-24	16.0	1.64	0.90	0.65
	finely	9-24	16.0	2.06	1.21	0.36
	ground					
Flotation Feed series 2	unground	5-23	13.3	1.58	0.88	0.80
	ground	5-23	13.3	3.46	1.82	0.61
Zinc concentrate	unground	51.5-54.5	53.1	0.52	0.53	0.40
	ground	51.5-54.5	53.1	0.45	0.41	0.41
Residue	ground	0.1-1.5	0.64	0.13	0.053	0.018

* Terms used in the equations for determining zinc.

† Samples from Broken Hill, N.S.W.

 N_1 : Pu-238/Ge γ X source, 25 mg/cm² Zn filter. N_s : Am-241/Cs γ X source, 220 mg/cm² Al filter. C_{Pb} : lead concentration.

TABLE II

Tin

As opposed to Ellis et al, Carr-Brion's (8) (9) investigation of tin concentration was undertaken in a tailing stream system utilizing the set-up depicted in Fig. 2, and (further described in the noted reference) where several liters of slurry were discharged intermittently into a recirculating loop along side the process stream. He employed a 14 mc, Am-241 source with a bonded BaSO₄ target in a S/T assembly with a Pd foil absorbing filter and silver transmitting foil. A compensation for changes in ($\mu_E + \mu_F$) was accomplished by measuring the backscattered x-ray radiation through the absorbing filter and taking the ratio of Sb-K γ intensity to backscattered intensity. The results obtained are indicated in Table III.

Ellis and Fookes et al, applied the same analytical technique--taking the ratio of N_1/N_S --to determine tin concentration in dry tailing samples. Measurements were taken employing the "Dual Target" method rather than balanced filters. They excited the SnK α -x-rays with an ²⁴¹Am/Cs assembly and exchanged the Cs target for Sn to obtain a measure of backscatter intensity. These results are indicated in Table IV, Case 1 and 5. Further tests were performed on samples of widely varying matrix composition--slime sulphides--containing up to 15% lead and 44% iron. By examining the function of μ vs $1/N_S$, (Fig. 3) utilizing a 60 keV transmission measurement, they arrived at an additional correction term increasing sensitivity (Case 3, Table IV). In addition, samples of widely varying tin content 0-23% by weight were studied. A correction term was put forth to compensate for tin absorption, coherent scattering caused by the tin, and the additional SnK γ x-rays excited by the 60 keV gamma photon from Am-241. The correction resulted in the expression:

$$C = a_1(N_1 + a_2[N_s - a_6N_1] + a_3) \left(\frac{1}{(N_s - a_6N_1) + a_4N_1 + a_5} \right) \quad (3)$$

Unground ore samples of slime takings were investigated using the same expression. (Case 4).

Barium

On stream, Barium analysis was conducted by Carr-Brion⁽⁸⁾, without employing balanced filters. He employed the dual target method, implementing an "on stream" system in a barytes flotation pilot plant. His choice of a source was 7.5 mc source of ^{241}Am with alternate targets of Sm_2O_3 and BaSO_4 . Results of a calibration run are plotted in Fig. 4.

Conclusion

Ellis' results must be qualified in that they are not slurry determinations. In aqueous solutions, there will be absorption of the characteristic radiation by the water reducing sensitivity scattered intensities will also be increased. As pointed out to determine the element concentration in a solid within a slurry, the solids slurry content may need to be monitored. Carr-Brion demonstrated that measured slurry density can be monitored if need be with a Cs-137 gamma gauge.

Fig. 4 Calibration graph for modified analyser obtained on-line using samples subsequently analysed

Table III. Comparison of On-line and laboratory tin analysis (Ref. 8)

SAMPLE ROUGHER TAILING NO.	TIN FROM ANALYZER %	TIN IN SAMPLE %
21	.26	.29
22	.22	.19
23	.21	.17
24	.18	.19
25	.14	.11

Fig. 2. Source-target-detector assemblies with filters and twin slurry presenter (diagrammatic) Ref 9

TABLE IV

Precision of determination of tin in mineral products *Ref. 2.*

Mineral product	Sample preparation	Tin (% by weight)		RMS error (% by wt. tin)		
		Range	Mean	N_1	N_1/N_2	Minimum (see text)
1. Mixed ores, residues and concentrates (Renison)	ground	0.2-23	3.4	2.33	0.85	0.21
2. Slime rougher tailings*	ground	0.08-0.2	0.14	0.016	0.007	0.006
3. Slime sulphides*	unground	0.5-3	1.0	0.22	0.068	0.034
4. Slime tailings*	unground	2-10	4.0	1.61	0.90	0.13
5. Slime tailings*	ground	0.5-2	1.3	0.27	0.03	0.028

 N_1 : Am-241/Cs γ X source, 53 mg/cm² Ag filter. N_2 : Am-241/Sn γ X source, 58 mg/cm² Pd filter.

* Samples from Ardath, N.S.W.

FIG. 3 Slime sulphides: relationship between the scattered X-ray measurement (N_1) and the sample mass absorption coefficient for 25 keV X-rays (μ).*Ref. 2.*

REFERENCES

1. Cameron, J.F. and Florkowski, "Radioisotope Sources of Low Energy Electromagnetic Radiation and Their Use in Analysis and Measurement of Coating Thickness," ORNL-11C-5, 1964.
2. Ellis, W.K., Fookes, R.A., Gravitis, V.L., Watt, J.S., "Radioisotope X-ray Techniques for On-Stream Analysis of Slurries. Feasibility studies Using Solid Samples of Mineral Products," Int. J. Appl. Radiat. Isotop., 20(10): 691-701 (1969).
3. Clayton, C.G., "Applications of Radioisotope X-ray fluorescence Analysis in Geological Assay Mining and Mineral Processing", Nuclear Techniques and Mineral Resources, IAEA Vienna, 1969, STI/PUB/198.
4. Carr-Brion, K.G., and Payne, K.W., "X-ray Fluorescence Analysis -- A review," The Analyst, Vol. 95, No. 1137, P 977.
5. Rhodes, J.R., "Progress in Industrial Application of Low Energy Radioisotope X-ray Techniques," Applications of Low Energy X and Gamma Rays, Ziegler (ed), Gordon and Breach, N.Y. 1971.
6. Papez, K, Cameron, J.F., "Applications of Non-dispersive X-ray Fluorescence Analyzers in the Basic Metals Industry," Symposium on the use of Nuclear Techniques in the Basic Metal Industries, Vienna, IAEA/Sm-159/16.
7. Carr-Brion, K.G., "The Selection of Exciting Energy in Radioisotope X-ray Fluorescence Analysis," The Analyst, Vol. 94, March 1969, PP 177-181.
8. Carr-Brion, K.G., "Performance of An On-Stream Radioisotope X-ray Fluorescence Analyser," Trans. Inst. Mining Met., 76, C94-100, June 1967.
9. Carr-Brion, K.G., Rhodes, J.R., "On-Stream X-ray Fluorescence Analysis of Ore Slurries with Radioisotope X-ray Source," Inst. Pract., Vol. 19, Nov. 1965.

d.) Finished Metal Analysis

Introduction

Radioisotope x-ray excitation has been explored as a possible means of alloy identification, alloy sorting and gold impurity analysis. The Hollstein and Devoe⁽¹⁾ study again points out the increasing impact of solid state detector techniques on the use of radioisotope induced fluorescence.

Alloy Analysis

Feasibility studies employing Am-241 excitation and a solid state detector (Si(Li)) for the determination of the Mo and Sn in alloys have been performed by Hollstein and Devoe⁽¹⁾ Three methods of sample preparation were evaluated; dissolution, followed by evaporation onto paper discs; dissolution alone; and chip preparation of the solid material. As noted in the graphs, chip preparation yielded the most sensitive results. The exciting source (1/8" dia capsule of 10 mCi of ^{241}Am) and detector were mounted on a flat aluminum plate in side by side geometry inside the cryostat.

Two types of material structures were involved in the study--tin in copper base alloys and molybdenum in steel. A typical pulse height spectra is illustrated in Fig. 1. The graphs, Fig. 2, and 3, depict the results of the investigation and an evaluation of the sensitivity of the methods of sample preparation. Matrix correction factors were employed to account for the increased absorption of the $\text{MoK}\alpha$ radiation in the Fe based alloys.

Heath⁽²⁾ has explored the possibilities of Si(Li) detector systems in determining alloy composition, using a 14 mCi, ^{241}Am S/T assembly with a Tellurium target ($\text{K}_\alpha - \text{K}_\beta$ x-rays from 27.2 to 31.7 keV) in a configuration similar to Rhodes (See Silver in Silver Ores).

He excited various stainless steel samples and found that their pulse height "signatures" could be employed for identification purposes. Fig. 4 .

A portable analyzer system consisting of an Ar-CO₃ proportional counter, balanced filters and radioactive sources of ²⁴¹Am (10 mCi) and ⁵⁵Fe (15 mCi) is suggested by Sellers et al.,⁽³⁾ in the sorting of various alloy materials. Their technique is based on the premise that in a family of alloys, rapid identification may be possible by determining the presence of some basic material then, when ambiguities arise, proceed to identify a second element. The method of analysis consists of first choosing a particular identifying element (e.g. in high-temperature alloys this could be nickel), then making an initial measurement with a "pass" filter in place, storing the resulting counts in the "up" mode of an up/down scaler and finally making another measurement with a proper absorber filter in place, the scaler now counting in the "down" mode. The resulting measurements thus produce two signals an "up" count reading proportional to the concentration of the characteristic element and other lower elements, and a difference reading which is primarily dependent on the concentration of the selected characteristic element. A plot of "up" versus difference counts for nickel in high temperature alloys is shown in Fig. 5 . Some identification can be ascertained but at least in cases like (I), (E), (J), another measurement must be made. In this case a molybdenum investigation may have the desired effect, a second measurement in the same alloys for Mo is illustrated in Fig. 6 . As noticed from the plots either "up" or diff. or both counts can be employed in identification.

Fig. 1 Spectrum of Copper-Base Alloy Sample No. 124d

Ref(1)

Fig. 2. Determination of Sn in Copper-Base Alloys

Fig. 3. Determination of Mo in Steel Ref(1)

Fig. 4 · X-ray spectra of stainless steel samples. Ref(2)

Fig. 5. Plot for nickel in hi-temperature alloys

Fig. 6. Plot for molybdenum in hi-temperature alloys

Re4(3)

The method is also examined for aluminum, titanium and copper alloys, Fe-55 being used as the exciting source for titanium alloys.

Gold Assay

Though Cd-109 is the predominant source employed in the investigation of gold ores, a method of assaying high purity gold in the gold refining process has been developed using ^{241}Am . (4)

The application is to ascertain the purity of gold within the 99.40-99.80 percent range. Since the impurity at this stage is mainly due to the presence of silver, the idea is to excite the AgK α -x-rays, their measured intensity being indicative of the purity of the gold. ^{241}Am was chosen because it offers an efficient means of exciting AgK α -x-rays, is a long-lived radioisotope and in practice did not produce a very high background of direct or scattered radiation. The analyzer itself is of typical central source backscatter geometry, employing an Argon filled proportional counter. A dual channel analyzer system was employed in the measuring process, one set with a window to respond only to the excited gold L-x-rays, and the other to the AgK x-rays). Count time was controlled by monitoring the Au-L x-ray intensity and using a preset scaler to automatically cut off the "silver" single channel analyzer at 3×10^6 counts in the gold channel. The instrument was calibrated to directly measure the purity of gold.

REFERENCES

1. Hollstein, M.G., DeVoe, J.R., "Analysis for Medium-Weight-Elements by Gamma-Excited X-ray Fluorescence," In Proceedings of 2nd Symposium on Low-Energy X and Gamma Sources and Applications, Austin, Texas, March 27-29, 1967, Vol. 1, P.S. Baker and M. Gerrard, Eds. PP 483-502.
2. Heath, R.L., "Review of Solid-State Detector Systems for X-ray Spectrometry," In Proceedings of 2nd Symposium on Low-Energy X-and Gamma Sources and Applications, Austin, Texas, March 27-29, 1967, Vol. 1. PP 424-41.
3. Sellers, B., Brinkerhuff, J.M., Hanser, , "Rapid Alloy Solting with a Radioisotope Analyzer," Application of Low Energy X and Gamma Rays, C. Zielger editor, P. 19, Gordon and Breach, 1971, N.Y.
4. Snyman, G.C., "Rapid Assay of High Purity Gold by Means of Radioisotope X-ray Fluorimetry," Int. J. Appl. Radiat. Isotop., 18(4), PP 243-6, 1967.

e. Coating Thickness Measurement

(1) Introduction

In the summary compiled by Cameron⁽¹⁾ on the use of radioisotope x-ray fluorescent techniques for coating thickness measurement, Am-241 is employed in over 30% of the reported applications. Its use in the measurement of tin on steel and galvanized coatings may probably account for a substantial amount of its commercial sales.

(2) General Discussion and Theory (2)

There are two basic methods for the measurement of coating thickness with an x-ray fluorescent gauge:

(A) The excitation of characteristic x-rays within the coating and measurement of the emitted intensity, given by

$$I = I_{SAT} [1 - \exp \{-(\mu_e + \mu_f)\} m] \quad (1)$$

(B) The excitation of characteristic x-rays in the backing material where the measured intensity is given by

$$I = I_{SAT} \exp \{-(\mu_e + \mu_f)\} m \quad (2)$$

where

m = mass per unit area of the coating

μ_e = mass attenuation coefficient of the exciting energy

μ_f = mass attenuation coefficient of the fluorescent radiation within the material in which it is excited

TABLE I Some on-line and laboratory measurements

Ref 1

Application	Source	Activity	Radiation detected	Detector	Filters and/or p.h.a.	Thickness range	Precision ($\pm 2\sigma$)	Measuring or response (5RC) time
<i>On line</i>								
Electroplated tin on steel	$^3\text{H}/\text{Zr}$	30 Ci	FeK	Ion Cham.	None	1 to 20 g/m ²	± 0.05 g/m ²	10 sec.
	^{241}Am	30 mCi	SnK	Prop.	Plastic	1 to 20 g/m ²	$\pm 2\%$ at 10 g/m ²	50 sec.
	^{241}Am + Cs target	170 mCi	SnK	Scint.	Ag	1 to 20 g/m ²	$\pm 1.5\%$ at 10 g/m ²	
Hot-dipped zinc on steel	^{241}Am	1 Ci	ZnK	Ion Cham.	Ni/Cu	30 to 500 g/m ²	± 3 g/m ²	15 sec.
	^{241}Am	30 mCi	ZnK	Prop.	Al	50 to 200 g/m ²	± 3.7 g/m ²	50 sec.
<i>Laboratory</i>								
Electroplated tin on steel	$^3\text{H}/\text{Zr}$	20 Ci	FeK	Ion Cham.	None	0.02 to 1 μm	$\pm 1\%$	25 sec.
	$^3\text{H}/\text{Zr}$	3 Ci	FeK	Scint.	None	0.02 to 1 μm	$\pm 2\%$	50 sec.
	^{238}Pu	10 mCi	FeK	Scint.	Mn	0.0 to 1.35 lb/b.b.	± 0.014 lb/b.b. at 0.77 lb/b.b.	10 sec.
Electroplated zinc on steel	$^3\text{H}/\text{Zr}$	3 Ci	FeK	Scint.	Ni, 0.013 mm	0.5 to 13 μm	$\pm 1\%$	30 sec.
	^{238}Pu	10 mCi	ZnK	Scint.	Ni/Cu Bal.	0.0 to 2.4 oz/ft ²	± 0.004 oz/ft ² at 0.312 oz/ft ²	10 sec. each filter
Hot-dipped zinc on steel	^{241}Am	10 mCi	ZnK	Scint.	Cr	30-500 g/m ²	$\pm 2\%$	40 sec.
Aluminium on steel	^{241}Am	1Ci	FeK and Back-scattered	Ion Cham.	None	0 to 100 μm	± 0.75 μm at 25 μm	15 sec.
Ferrochrome compressed powder on steel	^{238}Pu	30 mCi	CrK	Scint.	Ti/V Bal.	12.0 to 33.7 g/ft ²	± 0.20 g/ft ² at 15.2 g/ft ²	40 sec. each filter
Nickel plated on steel	^{238}Pu	30 mCi	NiK	Scint.	Co/Fe Bal.	0 to 10 μm	± 0.1 μm at 5 μm	10 sec. each filter
Gold coating on steel	^{57}Co	1 mCi	AuK	Scint.	None	0 to 32.5 μm	± 0.25 μm at 32.5 μm	10 sec.
Silver plated on non-magnetic metal	$^{147}\text{Pm}/\text{Al}$	500 mCi	AgK	Scint.	None	0 to 51 μm	± 0.1 μm at 11 μm	10 sec.
Pt electroplated on Ti	$^3\text{H}/\text{Zr}$	3 Ci	TiK	Scint.	None	0.05 to 1.3 μm	1%	30 sec.
	^{238}Pu	10 mCi	PtL	Scint.	Al, 0.025 mm	0 to 30 mg/cm ²	± 0.2 mg/cm ² at 20 mg/cm ²	10 sec.
TiO ₂ -Based paint on Polyester backing	$^3\text{H}/\text{Zr}$	2.5 Ci	TiK		None	20 to 100 μm		
	^{238}Pu	10 mCi	TiK		None	0 to 100 g/m ²	± 0.5 g/m ² at 20 g/m ²	100 sec.
Latex on steel formers	^{241}Am	100 μCi	Back-Scattered Np L X-rays			0.15 to 0.25 μm		
Plastic coating on Aluminium	^{55}Fe	5 mCi	Back scattered Mn K X-rays	Scint.	None	0 to 25 μm	± 0.75 μm	
Copper in "copper" electroplating solution	^{238}Pu	30 mCi	CuK	Scint.	Co/Ni Bal.	0 to 60 g Cu/l	± 2.6 g Cu/l at 60 g Cu/l	10 sec. each filter

where

$$I_{\text{SAT}} = KI_o/\mu_e + m_{\text{SAT}} \quad (3)$$

and

$$m_{\text{SAT}} = 3/(\mu_e + \mu_f) \quad (4)$$

For coatings which are compounds or mixtures of several elements values of μ_e and μ_f can be calculated from the relation

$$\mu = \sum_i \mu_i r_i \quad (5)$$

where μ_i and r_i are the mass attenuation coefficient and weight fraction of the i th element.

Theoretically when employing backing x-ray excitation no upper limit exists for measurable coating thickness. However, because of scattering from the coating and absorption of the incident radiation, the upper measurable limit of coating thickness for backing excitation is about the same as for coating excitation. Fig. 1 is a plot of m_{SAT} for $\mu_e = 0$ and can be used to determine the upper limit of measurable thickness for pure element coating by either method.

(3) Metal Coating Weight Gauges

Tin on Steel

Watt⁽³⁾ (4) employing an $^{241}\text{Am}/\text{Cs}$ target assembly (37 mCi of ^{241}Am)

demonstrated the practicality of employing x-ray fluorescent techniques to measure tin coatings on steel plate.

FIG. 1. Maximum coating thickness measurable by K X-ray excitation as a function of atomic number.

Employing the side by side geometry , Watt determined the tin coating weight on steel from emitted fluorescent radiation, with the results indicated in Fig. 2 .

Zinc on Steel

Margolinas⁽⁵⁾ reports on on-line zinc coating gauge developed in France, utilizing Am-241 as the exciting radiation and a proportional counter detector with an aluminum filter. (See Fig.3).

The proportional counter is employed due to its relativity insensitivity to Compton scattering of the intial radiation; the aluminum filter to improve the discrimination between the iron and zinc x-rays. Though eliminating 95% of the excited Fe radiation, the filter passes 50% of the Zn K x-rays. Measurements are reported to be made with better than 5% accuracy.

Chen and Cahill⁽⁶⁾ have developed a zinc coating gauge utilizing dual ion chamber detectors producing signals of opposite polarity. Employing backscatter geometry (Fig. 4) and two filters (one of Cu another of Ni), they obtained a spectral response function as indicated in Fig. 5 . The output signal of the apparatus is directly proportional only to the the 8.64 keV radiation and is thus a direct indication of the weight of the zinc coating. A gauge accuracy is reported of better than $\pm .01 \text{ oz./ft}^2/\text{side}$ over a wide range of substrate hardness (from full hard to dead soft), and maintains accuracies of better than $\pm .02 \text{ oz./ft}^2/\text{side}$ with varying sample composition. The advantage of this gauge in actual process control is indicated in Fig. 6 and Fig. 7 , for a double coating processes.

Fig. 2 Determination of the coating weight of tin on steel. Ref 4.

Fig 3 Schematic cross-section of the gage head.

Fig. 4 Typical zinc coating measurement Ref. 6.

FIGURE 5 Differential spectral response for Zn on steel (6)

FIGURE 6 Individual coating weight distribution before-after gage

FIGURE 7 Total coating weight distribution before-after gage

FIG. 8 Measurement of nickel coatings on steel. Ref(7.)

Miscellaneous Applications--Metal Coatings

Radioisotope x-ray fluorescent systems with Am-241 exciting sources have been employed to measure aluminum coatings on steel utilizing Fe-K excited x-rays.⁽¹⁾ Duftschmid⁽⁷⁾ reports the feasibility of measuring nickel coatings on steel with a specially developed portable x-ray fluorescent analyzer, incorporating a permanent 30 mCi, ^{241}Am source. Results of measurements employing this instrument are indicated in Fig. 8. The measurement of metal corrosion has also been suggested as a possible application of x-ray fluorescent methods.⁽⁸⁾ The migration of oxygen atoms into the surface of the material decreases the number of metal atoms exposed to exciting radiation and would thus decrease the fluorescent intensity of the characteristic radiation to an uncorroded surface.

(4.) Wear Measurements and Paper Weight Density

Introduction

An ingenious application of x-ray fluorescent thickness gauging to measure wear of a rubber flooring material has been reported by Kemper.^{(9) (10)} Employing the principles of backing excitation and using an Am-241 source in conjunction with a silver foil imbedded beneath the flooring material

Kemper has correlated the yield of the excited AgK α x-rays through the overburden tile with tile wear. (See Fig. 9). Kuusi,^{(11) (12)} reports a similar method to measure the amount of water removal, in a particular stage of paper processing.

Instrumentation and Results

After an evaluation of available low energy γ emitting isotopes (Cd-109, (88 keV- γ , and 22 keV - x-rays) ^{153}Gd (47 keV and 103 keV- γ ; 42 keV x-rays) and ^{241}Am), Kemper⁽⁹⁾ selected an ^{241}Am silver foil combination as the most

- a: characteristic target X rays
 b: radiation scattered in target
 c: radiation scattered in object
 d: direct radiation through shielding and scattered by materials of probe
 e: radiation scattered in backing

Fig 9 SCHEMATIC INDICATION OF THE METHOD WITH DIFFERENT CONTRIBUTIONS TO DETECTOR SIGNAL REF(9)

Fig10 Signal observed, plotted against wear produced in accelerated wear experiment. Counting time: 200 sec.

Fig11 Control of wear assessed by γ -X-fluorescence technique. Horizontally: wear measured with a micrometer. Vertically: wear read from calibration curve Ref (10)

practical and efficient method of producing the fluorescent x-rays. Rather than a central source geometry, an annular ring configuration (Fig. 9), was selected to provide increased detector surface and provide more space for the insertion of a collimator. Silver was judged to be the best target to insure separation of the characteristic x-ray peak from the backscatter peak and provide a reasonable production of fluorescent x-rays. Filters of Cg, Cu and Al were employed in front of the NaI detector to screen off any direct radiation, the output routed to an P.H.A., and ratemeter.

A detailed study was made of the contributions to the measured x-rays from backscatter radiation and calibration procedures were ascertained. The results of an accelerated wear experiment are depicted in Fig. 10, while Fig. 11, compares micrometer measurements versus that from the x-ray fluorescence method for a control wear experiment.

Kuusi⁽¹¹⁾ has demonstrated that this same technique can be used to determine the weight density of paper web as an indication of water removal in the processing of paper products. The apparatus (Fig. 12) employs an ^{241}Am 45 mCi point source, an Argon-methane proportional counter with central source geometry, and measures the excited characteristic radiation of CuK-x-rays from a bronze wire, attenuated by the overburden paper web. The paper web at this point in the process consists predominately of water and, therefore a measure of the attenuation of the characteristic radiation can be correlated to water removal.

A set of calibration curves for measured intensity versus weight density of water is presented in Fig. 13, Kuusi reports an accuracy, on the order of 1% with a fixed mounted instrument and a 20 sec measuring period over a wide range of paper weight density (300-3000 g/m²).

Fig. 12 Block diagram of the instrument. Ref(II)

Fig. 13. A set of calibration curves. Ref(III)

REFERENCES

1. Cameron, J.F, and Florkowski, "Radioisotope Sources of Low Energy Electromagnetic Radiation and Their use in Analysis and Measurement of Coating Thickness." ORNL-11C-5, 1964.
2. Clayton, C.G., "Applications of Radioisotope X-ray Fluorescence Analysis in Geological Assay Mining and Mineral Processing", Nuclear Techniques and Mineral Resources, IAEA, Vienna, 1969, STI/Pub/198.
3. Rhodes, J.R., "Progress in Industrial Application of Low Energy Radioisotope X-ray Techniques", Applications of Low Energy X and Gamma Rays, Ziegler (ed), Gordon and Breach, N.Y., 1971.
4. Carr-Brion, K.G. and Payne, K.W. "X-Ray Fluorescence Analysis -- A Review", The Analyst, Vol 95, No. 1127, P 977.
5. Papez, K, Cameron, J.F., "Applications of Non-Dispersive X-ray Fluorescence Analyzers in the Basic Metals Industry." Symposium on the use of Nuclear Techniques in the Basic metal industries, Vienna, IAEA/Sm-159/16.
6. Chen, Y.M., and Cahill, B.B., "Industrial Applications of Low Energy Gamma Emitting Isotopes," Applications of Low Energy X and Gamma Rays C. Ziegler, editor, Gordon and Breach, N.Y., 1971.
7. Duftschmid, K.E., "A Versatile Field Instrument For X-ray Fluorescence Analysis", Nuclear Techniques and Mineral Resources, IAEA Vienna, 1969, P 325.
8. Berk, S., "Measuring Metallic Corrosion By Radiation Backscattering and Radiation Induced X-rays,"
9. Kemper, A., "Wear Measurements by Means of γ -X Fluorescence", Euratom Report, 1967.
10. Kemper, A., "Wear Measurements by Means of γ -X Fluorescence," Wear, 12, 1968, PP 55-68.
11. Kuusi, J., Hietala, M., Puolakka, H., Lehtinen, A.I., "A Radioisotope X-Ray Method for On-Line Measurement of Fourdrinier Machine Water Removal on the Wire Section," Tappi, 52(12) PP 2378-81, (1969).
12. Kuusi, J., Puolakka, "Use of Radioisotope X-ray Techniques in the Paper Making Process and Quality Control Measurements," Peaceful Uses of Atomic Energy, Vol. 14, IAEA 1972.

f. "IN VIVO" FLUORESCENT IMAGING TECHNIQUES

(1.) Introduction

An interesting development in use of x-ray fluorescence in the biomedical field in the past decade is its application in the area of organ imaging. Its success has been directly coupled with the advance of solid state detector technology. Generally, "In Vivo" imaging techniques require the ingestion of radioactive tracers (I-125, Tc-99, etc.), this is not required with fluorescent techniques. X-rays fluorescence imaging depends on the ability of an outside radiant source to excite a "stable" tracer or residual element to some excited state and the detection of the emitted fluorescent radiation. With the radioactive source outside the body, the patient is administered a non-toxic stable compound capable, of course, of being excited to a fluorescent state. If an "organ" is known to accumulate a sufficient amount of a naturally occurring element, a fluorescent image of that organ may be obtained by exposing the organ to incident radiation capable of exciting K, or L x-rays of that residual element. This technique employed by Hoffer et al. (1) (2) (3) was successfully carried out at the University of Chicago in 1968. Their group successfully "imaged" the thyroid gland by monitoring fluorescent radiation from stable iodine (28.5 keV), excited by an Americium source (60 keV). The clinical arrangement (figure 1) included the use of a solid state detector (a lithium drifted silicon crystal) capable of resolving the emitted fluorescent peak from the scattered radiation produced in the surrounding tissue. As noted by Hoffer (14) with

Figure 1. Schematic representation of fluorescent scanning head currently in use. The radionuclide source is well collimated and produces a narrow beam of radiation which excites only one portion of the gland at a time. The detector is virtually totally uncollimated, allowing maximum solid angle for detection of characteristic X rays produced. The unit actually utilizes three 5-Ci ^{241}Am gamma-ray sealed sources, each focused at the same point. The entire scanning head is mounted on a rectilinear scanner. The detector is a solid state lithiumdrifted silicon crystal of cross section 30 mm^2 , 3mm depletion depth. (Reference 1).

Figure 2 not available at time of publication.

ordinary scintillation detectors the fluorescent method could not be employed (Figure 2).

This initial effort has since been followed by other developments, particularly in the area of monitoring cerebral blood flow. (5)(6)

Fluorescent scanning offers some advantages over tracer scanning. As far as the patient is concerned, it means a reduction in radiation exposure. In addition, Hoffer (3) has shown that as far as thyroid scanning is concerned, fluorescent techniques offer improved diagnostic capabilities, along with more precise imaging. Utilizing the particular geometric configuration as illustrated in Fig 1, one can see that this method is inherently laminographic and is being this successfully employed in the advance of organ aminography. (5)(6)

(2) Analytical Background

A thorough analytical treatment of "In Vivo" x-ray fluorescent imaging is presented by J.F. Tinney. (7) He has developed an expression for the fluorescent yield of a tracer element present in a given organ as a function of tracer element, organ depth, excitation energy and several other parameters. (See Eq. 1)

$$\frac{C_K^Z}{D} = \frac{6.25 \times 10^7 (\pi R_1^2) A_2^Z \epsilon_K^Z \omega_K^Z}{E_1 (\mu_{m-abs})_E^T (4\pi R^2) \sin^2 \phi} \cdot \frac{r_K^Z - 1}{r_K^Z} \cdot \frac{e^{-\alpha^T X_1} \tau_1^{Z,0}}{\alpha^0} \left\{ 1 - e^{-\alpha^0 (X_2 - X_1)} \right\} \quad (1)$$

where

C_K^Z	=	tracer element K-series fluorescent x-ray count rate (counts/sec)
D	=	tissue surface dose rate (mR/sec)
πR_1^2	=	effective collimator resolution area (cm ²)
ω_K^Z	=	tracer element fluorescent yield
E_1	=	excitation beam energy (keV)
$(\mu_{m-abs})_{E_1}^T$	=	mass energy absorption coefficient for primary photons in tissue (cm ² /g).
R	=	organ-detector separation distance (cm)
r_K^Z	=	tracer element K-edge absorption jump ratio
α^T	=	$\frac{\mu_1^T}{\sin \phi} + \mu_K^T$
μ_1^T	=	linear attenuation coefficient of primary photons in tissue (cm ⁻¹)
μ_K^T	=	linear attenuation coefficient of tracer element K x-rays in tissue (cm ⁻¹)
X_1	=	organ depth (cm)
$\tau_1^{Z,0}$	=	tracer element linear absorption coefficient for primary radiation in the organ (cm ⁻¹)
α^0	=	$\frac{\mu_1^0}{\sin \phi} + \mu_K^0$
μ_1^0	=	linear attenuation coefficient of primary x-rays in the organ (cm ⁻¹)
μ_K^0	=	linear attenuation coefficient of tracer element K x-rays in the organ (cm ⁻¹)
$X_2 - X_1$	=	organ thickness (cm)
A_2	=	detector area (cm ²)
ϵ_K^Z	=	detector efficiency.

Figure 3 Tinney (7)

Figure 4 . Iodine is the tracer element. Incident angle ϕ of the source collimator, is adjustable. (Reference 7).

Figure 5. Iodine tracer element. (7)

Figure 6 . Results obtained by normalization $C_K^Z/D = 1$ for $Z = 63$. Excitation energy chosen in this case to just exceed the K absorption edge energy of each tracer element. (7)

The expression is arrived at by an integration over the attenuation depth $X_2 - X_1$ for the emitted fluorescent radiation at the organ. The geometric variables in the above expression are referred to in the experimental arrangement in Figure 3.

Figures 4, 5 and 6 illustrate various dependencies of the resulting expression C_Z^K/D . A glance at figure 5, which shows the dependence of C_Z^K/D as a function of excitation energy, shows that with iodine as the fluorescing target having a K absorption edge of 33.2 keV, an excitation energy of about 45 keV would be optimum. This however neglects the attenuation of the incoming radiation by overburdened tissue. Employing Eq. 1, Tinney has shown that for an organ depth of 8 cm, an optimum excitation energy for iodine would be about 50 keV, making ^{241}Am a suitable source to supply the incident radiation, for thyroid imaging.

(3) Imaging Applications

X-ray fluorescent imaging of the thyroid does have clinical possibilities. There are several advantages to the method relative to tracer technique. A most important aspect is that due to the relative high content of iodine (.04% by weight) in the thyroid a "cold" fluorescent image can be obtained, whereas with the old tracer technique the resulting image was wholly dependent on the metabolic activity of the thyroid and the resulting uptake of the I-125.⁽¹⁾⁽³⁾ Indeed, Hoffer and his group have found that it may be very useful to compare "tracer" images with a fluorescent image. It has been demonstrated that certain areas

of metabolic inactivity, containing significant amounts of iodine related compounds can be located employing fluorescent imaging.

Fluorescent imaging significantly reduces the overall body dose administered (only 15×10^{-3} rad to the thyroid per scan) ⁽¹⁾ to the patient, since only a stable tracer rather than a radioactive tracer is employed. With the similar experimental arrangements as illustrated in Figure 1, a "laminographic" picture of the thyroid can be obtained, since the radiation detected must emanate from a specific irradiated volume, defined by the intersection of the detector and source line of sight. A further discussion of x-ray fluorescent radioisotope laminography follows.

The 60 keV photon emitted in the Am-241 decay process is a suitable source for thyroid imaging. The fluorescent yield of the ^{241}Am , 60 keV photon-Iodine interaction is 87%, 80% of the emitted radiation being comprised of 28.5 keV K x-rays. (3,8) As was mentioned earlier the 60 keV photon is within the optimum range of exciting the K x-ray of the stable iodine present within the thyroid, considering organ depth within the body.

For all practical purposes ^{241}Am is a monoenergetic photon source with the emitted α -particle being absorbed within the source encapsulation material. For the patient this means he is not exposed to any appreciable extraneous dose of radiation. In addition the incident radiation is well above any possible emitted iodine radiation so that by employing pulse height analysis and a relatively good solid state detector system, it is easily separated out from the subsequent fluorescent radiation.

Fig.7a

Fluorescent scanning device consisting of an annular collimated 17.2 Ci source of Am-241 in a doubly encapsulated steel ring, and a collimated Si(Li) detector. (Reference 4).

Figure 7b . Ge(Li) detector (left) and Si(Li) detector with ^{241}Am fluorescence source mounted on Ohio-Nuclear rectilinear scanner. (Reference 4).

Figures 1 and 7 (a,b) depict two fluorescent scanning arrangements.^{(1) (4)}

Imaging -- Other Organs & Blood Flow Studies

Applying x-ray fluorescent imaging to the thyroid was a natural consequence of the iodine presence in the organ. In other organ scanning situations one must find a suitable non-toxic, inert element of suitable atomic number capable of being excited by low energy incoming photons. As reported by Moody⁽⁵⁾ and Kaufman⁽¹¹⁾, fluorescent scanning has been utilized in the investigation of cerebral blood flow. They ascertained that stable iodine could be employed as a tracer through an inter-arterial injection of (Renografin 1-6) meglumine diatrizoate, 37% Iodine). Their initial studies performed with monkeys indicated that a very small amount of Renogiafin (29% Iodine) would give suitable results and could be tolerated by humans. Moody and his group also investigated the feasibility of employing stable Xenon as a tracer material in the study of blood flow. This would eliminate any interarterial injection to the patient. The advantage of these x-ray fluorescent scans, rather than the classic radioactive tracer techniques is again that whole body dose is reduced and most important diagnostically, the detected emitted radiation is confined only to the region of interest. Kaufman⁽¹¹⁾ recently reports employing this technique in studies involving six patients using two collimated 2.5 Ci, ^{241}Am line sources.

Figure 9 Diagram demonstrating the tomographic effect on detection within the region of interest. Detection of gamma rays is possible only within the area of intersection of the exciting beam and collimated field. Regions outside the beam from the source remain nonradioactive. The source is a 3.3 curie annular ring. Ref(5)

FIGURE 10 Block diagram of the system used to correct for variations of the intensity of the exciting X-radiation Ref 6.

Figure 9 shows the experimental arrangement of Moody et al. M.M. Ter-Pogossian⁽⁶⁾ has devised a scheme similar to the above, to study "In Vivo" blood flow, and has taken into account any error that he involved with radiation emanating from any volume not defined by the solid angle intersections of the source and detector. This case would occur if any high atomic number "Z" element was interposed between the source and the "irradiated volume" or between the irradiated volume and the detector. (See figure 10). Though Ter-Pogossian in his studies employed an x-ray unit, it seems that a suitable ^{241}Am source could be adopted to this application.

Laminography

The techniques of x-ray fluorescent imaging enable the investigator to view that portion of the organ that is intersected by the incident beam of radiation and the line of sight of the detector. (See figure 1 & 9). As mentioned earlier one can utilize this method of triangulation to obtain a 3-dimensional picture of the organ under investigation. By administering various tracer agents one could obtain laminographic pictures of the distribution of the tracers throughout the human body. This method has been demonstrated by Kaufman⁽⁹⁾ and others. With their laminographic camera, they administered a stable tracer and irradiated a "plane" within the patients neck and detected the fluorescent radiation with a multiwire proportional chamber (see figure 11 and Table I).

Figure 11 . Schematics of the Laminographic Excitation Camera. Ref 9.

REVELANT PARAMETERS OF THE LAMINOGRAPHIC EXCITATION CAMERA

Sensitive Area: $10 \times 12 \text{ cm}^2$
Intrinsic Resolution of the Wire Chamber: 0.14 cm FWHM
Collimator Resolution at the surface: 0.3 cm
Collimator Resolution at 2.5 cm: 0.5 cm
Detection Efficiency: 2.9×10^{-5} (8.7×10^{-5})*
Energy Resolution: 10% FWHM (approximately 5% with a tin filter)
Source Strength: 7.0 Ci of ^{241}Am (Effective Strength 5.0 Ci of ^{241}Am)
Laminographic Cut: 0.1 to 2 cm
Typical Counting Rate: 650 counts/cm³/min (1950 counts/cm³/min)*
Maximum Counting Rate: 3×10^7 /min
Typical Sensitivity: 500 counts/cm³/rad (1500 counts/cm³/rad*)

TABLE I .

Recent Advances

In very recent publications Hoffer⁽¹⁰⁾, and Kaufman, Nelson, et al⁽¹¹⁾ suggest the possibility of obtaining actual quantitative estimates for the amount of tracer present in a specific irradiated volume. It is pointed out that it would be extremely valuable in thyroid⁽¹¹⁾, liver and kidney⁽¹¹⁾ diagnostics. Ordinarily, in an organ picture, whether taken by x-ray fluorescent, imaging, or tracer techniques, the result is a picture of the relative distribution of tracer material within the organ of interest. Due to interposing unknown thicknesses of overburden tissue no quantitative information can be obtained. In tracer scanning, Hoffer noted, that R.D. Rollo⁽¹²⁾ by determining the relative depth of the gland under investigation, obtained quantitative information of tracer distribution. This was done by obtaining measurements of the emitted radiation from two different source to organ-detector to organ distances. (Fig.12).

Kaufman and his co-workers, in efforts to make an absolute determination of iodine concentration in the liver, "in vivo", have developed an alternate method employing a "calibration" procedure with tissue equivalent materials to ascertain the attenuation of the excited characteristic I-K x-rays emanating from the liver. The results of a study are indicated in Table II. The separation in concentration between the normal patients and the 6 "clinically" diagnosed as suffering from thyroiditis support the proposition that ^{241}Am radioisotope fluorescence may be employed as a diagnostic tool.

Studies are presently underway to determine iodine concentration in the liver, "in vivo". Kaufman reports that "in vivo" studies to are being performed to determine iodine concentration in blood, bile and urine. In this work a 300 mCi, ^{241}Am source of area 1 cm^2 is employed as the excitation source. Concentrations of iodine down to a level of .002 mg/cc (counting time: 500 sec) and 40 mg/cc (counting time: 20 sec) have been measured, in such biological fluids.

$$\text{RATIO } \frac{\text{COUNT AT } D_1}{\text{COUNT AT } D_2} = \frac{(D_2 \text{ SOURCE} + d)^2}{(D_1 \text{ SOURCE} + d)^2} \times \frac{(D_2 \text{ DETECTOR} + d)^2}{(D_1 \text{ DETECTOR} + d)^2}$$

Figure 12. The mean depth of the iodine content of the gland can be estimated by use of the inverse square principle. The detector and source are placed at a measured distance from the neck surface. The counts are recorded as "count at D_1 ." Both the detector and source are then moved closer to the neck surface, and the counts are recorded as "count D_2 ." Since the distance between the source and the neck at D_1 and D_2 and the actual counts per unit time are known, the distance d , which represents the mean depth of the iodine content it is not necessary actually to know the value of d . It is sufficient to use constant values for D_1 detector, D_2 detector, D_1 source, and D_2 source, and to know the ratio of counts at D_1 and D_2 . (Ref 10)

THYROIDAL IODINE CONCENTRATION (mg/cc)

CLASS	NUMBER	RANGE
Normals	11	0.085 - 0.65
Diffuse Toxic Goiter	1	0.14
Non-Thyrototoxic Autonomous Function	3	0.075 - .20
Thyroiditis (Clinical Diagnosis)	6	0 - 0.080
Papillary Carcinoma in Lymph Node	1	0

Table II (Reference 11).

Outlook

X-ray fluorescent imaging techniques are proving to be quite valuable in improving the diagnostic capabilities of medical personnel and reducing the radiation hazard to the patient. ^{241}Am has proven useful in thyroid imaging and has been demonstrated to be of use in cerebral blood flow analysis. The fact that ^{241}Am is highly monoenergetic means that the patient is no longer exposed to "useless" radiation. Its long life and relative low cost make it a perfect candidate to be incorporated into imaging equipment. Encapsulation is not a problem, since the only interest is in the 60 keV photon. The low energy of the source will be a disadvantage in some cases due to heavy tissue attenuation. With the recent discoveries of heavy trace mineral elements present in body fluids and tissues (66 to date) it may be possible to develop a greater family of non-toxic, inert, ingestible tracer compound that would further advance the application of x-ray fluorescent imaging to other body organs, and the use of ^{241}Am

REFERENCES

1. Hoffer, P.B., Bernstein, Joel and Gottschalk, Alexander, "Flourescent Techniques in Thyroid Imaging," Seminars in Nuclear Medicine, Vol. 1, No. 3, July 1971, PP 379-382.
2. Hoffer, Paul, B., "Flourescent Scanning: A medical Application of Flourescent Spectroscopy." Isotopes Radiation Technology, Vol. 6, No. 3, Spring 1969.
3. Hoffer, P.B., "Fluorescent Thyroid Scanning," Amer. J. Roentegencl, Radium Ther. Nucl. Med., 105(4) PP 721-7, 1969.
4. Brill, A.B., Patton, J.A., Baglan, R.J., "An experimental Comparison of Scintillation and Semiconductor Detectors for Isotope Imaging and Counting, " IEEE Transactions on Nuclear Science, June 1972, Vol. NS-19, No. 3, PP 179-189.
5. Moody, R.A., Hoffer, P.B., Polcyn, R.E., Lowe, H.J., Gottschalk, A., Dobben, G.D., "K-shell Fluorescence for the Study of Regional Cerebral Blood Flow," J. Neurosurg, 35, 181-4, Aug. 1971.
6. Ter-Pogossian, M. M., "In Vivo Flow Studies By Means of Excited X-ray Characteristic Radiation," Application of Low Energy X and Gamma Rays editor, Zeigler, Gordon and Breach Science Publishers, PP 241-248.
7. Tinney, J.F., "In-Vivo X-ray Fluorescence Analysis: Concepts and Equipment," UCRL-72962, Conf.-710204-1, 1971.
8. Clark, G.L., The Encyclopedia of X-rays and Gamma Rays, Reinhold, New York, 1963, PP 1124-1129.
9. Kaufman, L., Perez-Mendez, V., Powell, M., "Laminographic Excitation Camera for Thyroid Imaging," UCRL-20229, 1970.
10. Hoffer, P.B., Beck, R., Stark, V., and Peck, T., "Applications of Flour-
escent Scanning in Nuclear Medicine," IEEE Transactions on Nuclear Science, Feb. 1973, Vol. NS-20, No. 1, PP 375-378.
11. Kaufman, L., Nelson, J., Price, D., Shames, D., Wilson, C.J., "Some ap-
plications of Si(Li) detectors to Clinical Problems," IEEE Transactions
on Nuclear Science, Feb. 1973, Vol NS-20, No.1, PP 402-409.
12. Rollo, F.D., "¹²⁵I for Measuring The Radioiodine Uptake of the Thyroid,"
Journal Nuclear Medicine, 12:8, 1971.

g Trace Element Analysis

(1) Introduction

The advent of high resolution solid state detectors and the interest in the presence of trace amounts of metal elements in various matrices has led to the use of radioisotope x-ray fluorescent techniques, in several fields, and various feasibility studies to determine future applicability. (1) (2) (3) (4) (5) (see other references in previous section). As indicated, radioisotope excitation in conjunction with solid state detectors, though generally inferior to x-ray tube excitation and neutron activation analysis, does have the capability of providing a semi-portable analysis system, with PPM sensitivity. The work of Kaufman et al. and Hoffer and his co-workers (cited in the previous section) demonstrates the type of analysis that can be accomplished. As noted by both investigators, systems have been developed to *determine quantitatively* the concentration of iodine in the thyroid gland and the liver. Analysis of stainless steels (Heath) and the determination of silver in silver ores (Rhodes) with solid state detectors further attest to the wide range of ^{241}Am x-ray fluorescent spectrometers employing either Si(Li) or Ge(Li) detector systems.

(2) System Design and Sensitivity

The range of elements where Am-241 would provide an adequate excitation source has been cited in previous sections. A study completed by Baglan⁽³⁾ et al. to determine the sensitivity of solid state systems and the use of ^{241}Am suggests that Si(Li) detectors are generally the best choice in

non-dispersive analysis Employing a 20 Ci, Am-241 annular source, they determined a measure of the relative sensitivity attainable with an ²⁴¹Am Si(Li) system. The results are indicated in Table I . Cranston and Anspaugh⁽⁶⁾ in a feasibility study to determine the design of a radioisotope x-ray fluorescent system as applied to biological analysis, reported an expression to calculate the number of counts/sec that will be accumulated in a given photo peak (N_p) for a particular excited x-ray, taking into account several system parameters.

$$N_p = .282 \times 10^{33} \left(\frac{A_2 \epsilon \cdot f m \sigma S \eta}{AW d_1^2 d_2^2} \right) \quad (1)$$

The calculation is based on the assumption that the source and sample are point parameters.

- A₂ = Area of Detector
- ε = Photopeak efficiency of detector
- f = fluorescence yield in sample
- m = mass of unknown element in sample
- σ = photon-capture cross-section per electron in sample
- S = source strength in Ci
- η = intensity of desired gamma ray
- AW = atomic weight of element under study
- d₁ = source-to-sample distance
- d₂ = sample-to-detector distance

Table I Relative sensitivity ($N/\sqrt{BKG}$) versus atomic number using ^{241}Am exciting source. (See Appendix)

Z	K α (keV)	Cross- Section (Barns)	Net Counts(N)	BKG	$N/\sqrt{BKG}$
51	26.2	1330	11100	800	400
53	28.5	1550	12450	900	420
55	30.3	1770	13650	900	455
57	33.7	2000	14900	1800	346
59	35.8	2300	16600	2500	332
61	38.4	2580	18000	3100	321
63	41.2	2920	19700	5000	282
65	44.1	3220	21000	6000	272
67	47.1	3680	23400	22000	158
69	50.2	4000	24700	38000	127

(3) Biological and Environmental Studies

Goulding and Jaklevic⁽⁴⁾, Cranston and Anspaugh⁽⁶⁾ discuss the general applicability of radioisotope induced fluorescent systems to the study of trace mineral content in biological samples. It is worth noting that in most biological samples backscattered radiation is due mainly to interaction with light elements in the sample matrix. A continuum of incident low energy photons would result in a continuum of backscattered radiation, restricting the sensitivity of analysis. The use of monoenergetic excitation, available with radioisotope sources, confines the backscatter peak to the high energy portion of the spectrum. The reported biological investigations employ extended area source configurations.

Cranston et al., studied, in detail, various aspects of Am-241-Ge(Li) systems in the detection of barium. Preliminary investigations were performed with a barium-doped cellulose material, — Avicel, and a spectrum from an irradiated ocean clam specimen (*Saxidomus*) is reported (Fig. 1, and Fig. 2). The ²⁴¹Am source employed was an extended area configuration disc type placed in combined source/sample holder illustrated in Fig. 3. One disc source was used although the holder had been designed to accommodate four such sources.

Fig. 1 The Ba K α line from a 176 ppm Ba in Avicel sample.

Fig. 2 Portion of the spectrum from an ocean clam (*Saxidomus*).

Ref 6

Fig. 3. Sectional sketch of the source-sample holder. Ref 8.

Fig. 4 . Americium-241-Tin X-ray Excitation of Calcium and Strontium X-rays of Egg Shells

Fig. 5. Arsenic Content of Soil vs. Count Rate of Arsenic K_{α} X-ray

A news item in a commercial application newsletter has reported use of a Si(Li)-Americium (20 mCi) system to obtain a pulse height "signature" of trace material content in nine varieties of salmon to determine their geographic origin. The study is reported to be underway at the Fisheries Research Board of Canada, under the direction of Dr. John Calaprice.

Cate⁽⁷⁾ reports the use an ^{241}Am , 30 mCi annular source assembly to investigate the possibility of employing radioisotope x-ray fluorescence methods to detect lead in air sampling filters. ^{241}Am was selected over ^{109}Cd to excite the characteristic L-x-rays of Pb because of the high signal to background ratio it provided (See Table II). Artificial samples were made up by filtering lead acetates, of varying concentration. No interfering elements were in the solution and therefore were not deposited on the filter samples as would be the case in air sampling. This is an important qualifying factor since Ba occurs with lead in the air and would definitely effect Pb determination. The results obtained are indicated in Table III. Cate⁽⁶⁾ notes the minimum detectable quantity as $.072 \mu\text{g Pb/cm}^2$.

Pressly⁽⁸⁾ has utilized an ^{241}Am , 300 mCi annular source in source/target/sampler holder assembly, to excite characteristic x-rays from various sample materials. Fig. 4 is a portion of pulse height spectrum produced by Snk-x-rays from an $^{241}\text{Am}/\text{Sn}$ source target assembly, incident on egg shells, indicating the presence of calcium and strontium. An $^{241}\text{Am}/\text{Mo}$ assembly was also employed by Pressly to excite AsK-x-rays in soil samples to ascertain a possible correlation between soil arsenic content and measured AsK x-ray yield. The results are indicated in Fig. 5.

TABLE II
RESULTS OF EXCITER OPTIMIZATION EXPERIMENTS

SOURCE	X-RAY LINE S^2/B		
	$L\alpha$	$L\beta$	$L\alpha + L\beta$
Cd^{109}	1.3×10^4	9.91×10^3	2.27×10^4
Am^{241}	1.36×10^5	3.27×10^4	1.32×10^5

TABLE III
Results of samples prepared with the solution lead acetate

CONC. $\mu g/cm^2$	<u>NET COUNT RATE</u> COUNTS/MIN $\pm 2\sigma$
100	12.403 ± 20
50	6.328 ± 178
10	1357 ± 39
5	769 ± 24
2	323 ± 23
1	167 ± 14
1/2	78 ± 12
BKg 0 $\mu g/cm^2$	121 ± 8.6

In contrast to the growing trend to employ solid state detector systems in x-ray fluorescent analysis, Florkowski et al, reported the use of a proportional counter in the determination of trace metal content in water. ^{109}Cd (2.5 mCi), ^{239}Pu (10 mCi) and ^{241}Am (5 mCi) sources were used to provide the exciting incident radiation in determining the presence of elements indicated in Table IV. Calibration curves to ascertain trace element content down to the P.P.M. level were obtained, and subsequent investigations were undertaken. Americium excitation was particularly employed to determine the presence of Cd. A sample calibration curve is depicted in Fig. 6. The results obtained are tabulated in Table IV. The indication from the data is that ^{241}Am can be employed to ascertain the presence of Cd to an accuracy of ± 4 PPM.

(4) General Chemical Analysis

Though other laboratory analysis techniques are inherently more sensitive than radioisotope induced x-ray fluorescence, ^{241}Am - solid state and ^{241}Am - scintillator systems, have been studied as complementary chemical analysis tools, particularly where "on-line" techniques are needed, as in a chemical processing operation.

Feasibility studies with an ^{241}Am Si(Li) system have been reported by Hyde et al.⁽¹⁰⁾ to determine barium concentration in barium solutions at the one to ten percent level and yttrium concentrations at the milligram or trace level. In both cases the K-x-ray yield is correlated to the element concentration.

Though most trace element studies with ^{241}Am depend on the excitation of characteristic K-x-rays within the sample material, Shenberg and Amiel⁽¹¹⁾ have demonstrated that L-x-ray excitation of uranium in uranium solutions can be successfully correlated to uranium concentration. Using an $^{241}\text{Am}/$

Table IV. SUMMARY OF RESULTS

Source	Content (ppm)	I/I _{H₂O} for elements:					
		Zn Zn K	Cu Cu K	Fe Fe K	Pb Pb L	Hg Hg L	Cd Cd K
¹⁰⁹ Cd	0	1.000	1.000	1.000	1.000	1.000	
	5	1.010	0.004	-	1.013	1.015	
	10	1.026	1.022	1.015	1.037	1.027	
	25	1.056	1.055	*1.05	1.072	1.035	
	50	1.123	1.105	1.13	1.124	1.094	
	100	1.232	1.22	1.27	1.223	1.222	
²³⁸ Pu	0	1.000	1.000	1.000	1.000	1.000	1.000
	5	1.019	1.009	1.003	1.002	1.020	1.028
²⁴¹ Am	10	1.038	1.017	-	1.066	1.040	1.043
for Cd	25	1.097	1.046	1.028	1.029	1.080	1.063
	50	1.181	1.097	1.053	1.053	1.123	1.125
	100	1.371	1.227	1.132	1.100	1.249	1.240
¹⁰⁹ Cd		Mixture of equal amounts					
	0			1.000			
	10			1.020			
	20			1.031			
	40			1.055			
	60			1.070			
	20			1.100			
	100			1.120			
¹⁰⁹ Cd		After chemical extraction					
	2	1.03					
	5	1.24					
	10	1.53					
	20	2.24

Fig. 6 Example of calibration curves (²⁴¹Am source).

Iodine source/target assembly, (1 mCi annular ^{241}Am source) and Si(Li) detector, they were able to obtain a linear correlation between the ratio U-L α , emission/I-K α scattered incoherent radiation, and uranium concentration. (See Fig. 7).

In an effort to develop a system to determine the concentration of Br, Ag and I in pharmaceutical products Svitel and Schiller,⁽¹²⁾ employed ^{153}Gd , ^{109}Cd and ^{241}Am as excitation sources with a scintillator detector system. ^{241}Am was found to be the best choice for iodine determination. In the pharmaceutical, Yastyl, (iodoantipyrine) iodine concentration of .55% was able to be ascertained with an absolute analysis accuracy of .0067%. Bowman *et al.*⁽²⁾ have demonstrated that pulse height "signatures" identifying solder and phosphor bronze can be clearly obtained using an ^{241}Am exciting source and solid state Si(Li) detector system. In solder, the incident radiation results in the excitation of Pb L-x-rays and Sn K-x-rays while in phosphor bronze the K-rays of Cu and Sn are excited.

Fig. 7 Comparison between UL_a and $UL_a/I K_a \text{ incoh}$ as a function of U concentration Ref 11

REFERENCES

1. Walter, F.J., "The Impact of Semi-Conductor Detectors on X-ray Spectroscopy," IEEE Trans on Nuclear Science, NS-17, No. 3, PP 196, June, 1970.
2. Bowman, H.R., Hyde, K, Thompson, S.G., Jared, R.C., "Application of High-Resolution Semiconductor Detectors in X-ray Emission Spectrography," Science, 151, PP 562-8, 1966.
3. Baglan, R.J., Brill, A.B., Patton, J.A., "Applications of Non-Dispersive X-ray Fluorescence Techniques for In Vitro Studies," IEEE Trans. on Nuclear Science, NS-20, No. 1, Feb. 1973.
4. Goulding, F.S., and Jaklevic, "Trace Element Analysis by X-ray Fluorescence," UCRL-20625, May 1971.
5. Heath, R.I., "Review of Solid State Detector Systems for X-ray Spectrometry," ORNL-11C-10, PP 465.
6. Cranston, F.D., Anspaugh, L.R., "Preliminary Studies in Non-Dispersive X-ray Fluorescent Analysis of Biological Materials," UCRL-50569, Jan. 1969.
7. Cate, J.C., Jr., "Determination of Lead in Air Sample Filters by X-ray Fluorescence Analysis," UCRL 51038, 23 Apr. 1971.
8. Pressly, R.L. "Design and Use of Low Energy Gamma and X-ray Sources," Applications of X and Gamma rays, Ziegler editor, Gordon and Breach, N.Y., 1971.
9. Florkowski, T., Holynska, B., Niewodniczanski, J., "Feasibility of Radioisotope X-ray Fluorescence Analysis for Measuring Water Pollution by Metals," Nuclear Techniques in Environmental Pollution, Austria, Oct. 26-30, 1970, C.N. Welsh, ed. IAEA Vienna, Austria, PP 335-45.
10. Hyde, E.K., Bownam, H.R., and Sissor, P.H., "Analytical Capabilities of a Semiconductor X-ray Emission Spectrometer," UCRL-16845, May 1966.
11. Shenberg, C. and Amiel Saadia, "Analytical Significance of Peaks and Peak Ratios in X-ray Fluorescence Analysis Using a High Resolution semiconductor Detector," Analytical Chemistry, Vol. 43, No. , July 1970, PP 1025.
12. Svitel, J., Schiller, P., "The Determination of Br, Ag, and I in Pharmaceuticals Using X-Ray Fluorescence Excited By Gamma Sources," J. Radioanal. Chem, 5(2), PP 343-51, 1970.

V. Miscellaneous Applications

Photons from ^{241}Am and ^{137}Cs are used in differential gamma ray absorption in spent fuel⁽¹⁾ to detect the presence of elements of $A > 230$. This method depends on the fact that low energy gammas is absorbed in a manner proportional to the fifth power of the atomic weight of the absorber, whereas the absorption of high energy gammas goes as the first power of the atomic weight.

A similar approach has been discussed by Charbucinski⁽²⁾ for determination of the percentages contents of heavy element content in ore. Charbucinski⁽³⁾ also developed nomographs for ores with 0-15% zinc content and 0-4% lead content. They can be used to interpret measured results obtained using the selective non-spectrometric - method of determining the Zn and Pb content in polymetallic ores. Gamma ray absorption spectroscopy has been shown to be a fast, accurate, and non-destructive method for determining the Si and Ge content in SiGe alloys.⁽⁴⁾

^{241}Am , as an alpha source, has been used by Eggelsmann and Rasp⁽⁵⁾ to make area measurements. The area to be measured is placed between a large-area radioactive source and a suitable radiation detector. The loss in counting rate at the detector caused by the absorbing material gives a direct measurement for its area. An unique application of ^{241}Am is in an indwelling intracranial pressure sensor. The pressure sensor⁽⁶⁾ consists of a two fluid system (See Fig. 1). One fluid is placed in a tambour located within a cerebral centricule. Variation in the intracranial pressure causes compression or extension of the tambour. When the pressure increases the neutral fluid in the tambour is forced into the collection chamber of Fig. 1., where the radioactive fluid

Figure 1. Indwelling intracranial pressure sensor. (Ref. 6).

METHOD OF ANALYSIS	RADIATION SOURCE	$\frac{\sigma W}{W}$, % ESTIMATED FROM SPREAD OF THE EXPERIMENTAL POINTS	$\frac{\sigma W}{W}$, % (reproducibility)
1. X-ray fluorescence	$^3\text{H/Zr}$	10 - 20	3.5 - 14.0
2. X-ray fluorescence	^{109}Cd	8 - 17	2.5 - 8.5
3. Scattering	^{109}Cd	5 - 10	2.5 - 5.0
4. Scattering	^{241}Am	—	2.0 - 7.0
5. Scattering-transmission	^{241}Am	—	1.0 - 4.0
6. Transmission	^{241}Am	3 - 5	1.5 - 3.5

Table I. The estimate of $\frac{\sigma W}{W}$ for x-ray fluorescence, scattering and transmission methods. (Ref. 7).

(^{241}Am in solution is housed). The radioactive fluid is separated by a diaphragm and as the intracranial pressure increases the radioactive fluid is displaced. The displaced fluid is forced into a pressure readout chamber. The gammas from ^{241}Am are counted by a 1" scintillator crystal. For a fluid containing 10 n Ci of ^{241}Am , the dose rate to the neutral tissue was calculated to be .01 Mrem/day at 1 cm

Dzfunikowski⁽⁷⁾ examined the methods of x-ray fluorescence and scatter-with-transmissions for use in the measurements of the Fe concentration of Fe-rich sands. It was found that for accuracy and sensitivity the transmission method using ^{241}Am was better than that obtained with a portable x-ray fluorescence source. The relative standard deviation of the results was determined to be less than 5% with little sensitivity to variation in the distribution of Fe in the sample. A comparison of the different methods is given in Table I.

A gamma scattering method has been used to examine the feasibility of measuring atmospheric density. (See Fig. 1). But it was found that at high altitudes the instrument was limited by high background levels. Gardner and Whitaker⁽⁸⁾, in regards to these high background levels, has employed a method where the source (^{241}Am) and the detector are suitably spaced with shielding interposed to prevent the direct unscattered gammas from reaching the detector. The number of gammas reaching the detector are then assumed to be scattered. The calibration arrangement used in the experiment is shown in Fig. 2.

Figure 2. Gamma-ray density sensor and boost vehicle used on flight tests, (Ref. 8),

The experimental gross response of several sources to density at two different source-to-detector positions is given in Fig. 3.

Figure 3. Illustration of calibration arrangement showing tower, source, shield and detector. (Ref. 8).

Figure 4. Gross experimental response to density for all sources.
(Ref. 8),

REFERENCES

1. J.E. Strain, W.J. Ross, G.A. West and J.W. Landry, "The Design and Evaluation of a Delayed-Neutron Teached-Hull Monitor", ORNL-Feb. (1968).
2. J. Charbucinski, "A Gamma-Gamma Scattering Method for Simultaneous Determination of Two Heaby Elements Content In Ores", Radiochem Radioanal. Letters , 4(3), PP 157-64 (1970).
3. J. Charbucinski, "A Selective Non-Spectrometric Gamma-Gamma Method of Determing the Zn and Pb Content in Polymetallic Ores," Nukleonika 15, PP 563-72, (1970).
4. , "The Determination of the Si and Ge Content in SiGe Alloy by Gamma Ray Absorption Spectroscopy",
5. H. Eggelsmann and W. Rasp, " Area Measurement Method with A Radioactive Preparation," PTB Mitt, 80, PP 436-9, (1970).
6. G. Bloom, K. Murphy, and T. Cox, "An Isotope-Powered Indwelling Intracranial Pressure Sensor", Trans Am. Nucl. Soc., 13(2), Nov. (1970).
7. B. Dziunikowski "Methods of Measuring the Concentration of Iron in Quartz by Means of Gamma-and X-rays", Inst. Mining Met., Trans., Sect. B, 76, (1967).
8. R.P. Gardner and D.R. Whitaker, "Experimental and Theoretical Studies on the Gamma-Ray Scattering Technique for Measuring Atmospheric Density", Nucl Eng. and Des., PP. 13-28, (1968).

VI SURVEY

A direct mail survey of Am-241 source fabricators and distributors, and instrument manufacturers possibly employing Am-241 has been performed in relation to this study.

The mailing list was compiled mainly from the Science Buyer Guide, (1972-1973) and complimented by information from the yearly publication of Nuclear Industry, a list of radioisotope suppliers from Nuclear Technology and from information obtained during the literature review study. A cover letter and respective questionnaire were sent to the firms involved (See examples A, B,).

The information contained in the following Tables I, II, III was taken from the questionnaire responses and any additional information submitted by the responding firms. The firms involved were most cooperative in offering information.

Am-241 - Survey

(EXAMPLE A)

FIRM: _____

Does your firm fabricate Am-241 sources? Yes__ No__

Does your firm market Am-241 sources? Yes__ No__

If yes, designate type and range of activity.

(a) Photon source

(b) Alpha source

(c) Composite Neutron source

Type _____

If possible, please indicate below or on an additional sheet, your principal customers.

Does your firm manufacture instrumentation utilizing Am-241?

Yes__ No__

If so, what type? _____

Additional Comments: (Unusual applications, specialty, items.)

*(If not in violation of propriety customers' agreements.)

(EXAMPLE B)

DATA SHEET

(Survey of Companies Utilizing Am-241 in Instrument Fabrication.)

1. Company: _____ 2. Person(s) completing questionnaire:
(name and position) _____

3. Type of instrument(s) manufactured:

4. Principle industries where your equipment is employed.

5. Principle applications:

6. Does your equipment utilize Am-241? (Circle one) Yes No
(If so, please complete the following if possible)

7. Am 241 is utilized (circle where appropriate)
(a) As a component in a composite neutron source
(Name configuration (Am-Be, Am-Li, etc.) _____)
(b) As an α - Source
(c) As a monoenergetic source of low energy photons
(d) In a source/target assembly to produce monoenergetic X-rays
(e) Other _____

8. With respect to the above, complete the following.

Source

Activity/Yield

(Note: In composite source configurations, please indicate activity of Am-241 and yield of secondary radiation.)

9. Name(s) of your principle supplier(s).

PLEASE DO NOT FEEL RESTRICTED TO JUST COMPLETING THIS FORM. FORWARD ANY AND ALL INFORMATION YOU DEEM APPROPRIATE, (SPECIFICATIONS, DATA SHEET, ETC.).

TABLE I
SURVEY OF RADIOISOTOPE SUPPLIERS

FIRM	AM-241 Source Fabricator				DISTRUBUTOR		PHOTON SOURCE		S/T ASSY.		ALPHA SOURCE		NEUTRON SOURCE	
	YES	NO	NO	REPLY	YES	NO	POS. REPLY	TYPE/ ACTIVITY	POS. REPLY		POS. REPLY		POS. REPLY	
AB Atomenergi Isotop- service														
AB Isotopeteknik	SEE ISOTOPES				TECHNIQUES LAB									
Acec-Ateliers De Con- structions Electri- ques De Charleroi			X											
Aeg-Telefunken	Referred to				Alkem Division --		No reply							
Aktiebolaget Atomenergi Isotopservice, Sweden	X				X		X	To 10 mCi (Most for inhouse, manufactured instrumentation)						
American Atomics Corp.		X				X								
American Nuclear Corp.						X								
American Radiochemical Corp.						X								
Amersham/Buchler	X				X		X	1-3000 mCi	X		X	.01-10μCi	Am/Be, Am/B, Am/Li Am/F 1-25mCi	
Amersham/Searle	X				X		X	3 mCi to 1 Ci	X	upon customer request	X	3mCi-1Ci	X	Am/Be 1mCi-5Ci Am/B 100mCi- 3 Ci Am/F 100 mCi- 3 Ci Am/Li 100 mCi- 1 Ci
Argonne National Lab.		X				X								

TABLE I (con't)
SURVEY OF RADIOISOTOPE SUPPLIERS

FIRM	AM-241 Source Fabricator				DISTRUBUTOR		PHOTON SOURCE		S/T ASSY.		ALPHA SOURCE		NEUTRON SOURCE	
	YES	NO	NO	NO	YES	NO	POS.	TYPE/ ACTIVITY	POS.	REPLY	POS.	REPLY	POS.	REPLY
	REPLY						REPLY		REPLY		REPLY		REPLY	
Atomchem Corp.			X											
Atomic Corp. of Amer.		X				X								
Atomic Energy Research Est. (England)	X					X								
Atome Industriel														
Atomic Energy of Can- ada, Ltd.	X				X		X		X	High energy 3.85 MeV-Am/B SOURCE				
Atomic Energy Research Establishment (Den- mark)	X					X								
Atomics International		X				X								
Austrialian Atomic Energy Commission			X											
Automation Industries		X				X								
Battelle Northwest Laboratories	X					X					X	Up to 2 mCi/cm ²		
Baird Atomic Inc.														
Bio-Nuclear Division of American Bio- medical Corp.														
Bio-Rad Laboratories		X				X								
California, University of Los Alamos Sc- inetific Lab.	X					X					X			
Canrad Precision Ind.														
CEA/IRE, (France/Bel- gim)	X				X		X	100 μ Ci - 2Ci	X		X	various types .1 μ C to 2 mCi		Am/Be 10 mCi to 7Ci

TABLE I (con't)
SURVEY OF RADIOISOTOPE SUPPLIERS

FIRM	AM-241 Source Fabricator				DISTRUBUTOR		PHOTON SOURCE		S/T ASSY.		ALPHA SOURCE		NEUTRON SOURCE	
	YES	NO	NO REPLY		YES	NO	POS. REPLY	TYPE/ ACTIVITY	POS. REPLY		POS. REPLY		POS. REPLY	
C.E.N.-S.C.K.														
Cambridge Nuclear Corp														
Capintec Nuclear		X				X		(Isotope calibration standards for nuclear medicine)						
Chemapol			X											
Cumberland Research Corp.		X				X								
Douglas Lab, Donald W.		X				X								
Douglas United Nuclear Inc.		X				X								
Dow Chemical Co.		X				X								
E.I. Dupont de Nemours and Co.		X				X								
Eberline Instrument Corp.	X				X						X	activity range up to 5μCi		
Energie Froide, Switzerland		X				X								
Eidg. Institut fur Reaktorforschung (Switzerland)		X				X								
Gamma Industries Inc.	X				X		X	COMMERCIAL SOURCES, mCi - 1000mCi level					X	Am/Be 1 mCi-20Ci, Am-Be-Cs 1mCi-500 mCi
General Electric, Vallecitos, Nuclear Center	X				X									Am-Be-Cm 3000Ci Am-Be 10 Ci
General Nuclear Inc.					(Source fabrication facilities acquired by Gamma Industries)									
Georgia Institute of Technology		X				X								
Gesellschaft fur Kernforschung M.B.H.														
Gulf General Atomic		X				X								

*npts -- Nuclear transformations per second.

TABLE I (con't)
SURVEY OF RADIOISOTOPE SUPPLIERS

[illegible]

TABLE I (con't)

FIRM	AM-241 Source Fabricator				DISTRUBUTOR	PHOTON SOURCE		S/T ASSY.	ALPHA SOURCE	NEUTRON SOURCE
	YES	NO	NO		YES NO	POS.	TYPE/ ACTIVITY	POS.	POS.	POS.
	REPLY					REPLY		REPLY	REPLY	REPLY
Nuclear Materials and Equipment Corp.	X				X					X Am/Be 1-10Ci/sec
Nuclear Radiation Developments, Inc.	X				X				X up to 1500µC/in ²	
Nuclear Research Corp.										
Nuclear Supplies		X			X					
Nuclear Technology			X							
Nucleonic Corp. of America										
Nukem-Nuklear Chemie Und Metallurgie GMBH			X							
Ortec, INc.						X .1µCi-25mCi			X .1µCi	
Parkwell Laboratories	X				X (NO DIRECT REPLY, INFORMATION FROM A USER)					
Phillips-Duphar, N.V.										
Picker, Corp.										
Radiation Materials Co., Inc.										
Technical Operations Inc.										
Teledyne-Isotopes		X			X					
United Kingdom Treasury and Supply Delegation										
United States Radium Corporation		X			X					
Universal Radio-isotopes, Inc.			X							

MANUFACTURERS EMPLOYING RADIOISOTOPES IN GAUGING INSTRUMENTATION
(ABSORPTION)

[illegible]

TABLE II (con't)

FIRM	DISTRIBUTOR OR FABRICATOR EMPLOYING RADIOISOTOPES			AM-241		TYPE OF GAUGE OR INSTRUMENT	SOURCE AND ACTIVITY	DETECTOR	OUTPUT/ READOUT	APPLICATIONS
	YES	NO	REPLY	YES	NO					
Cryogenic Research			X							
Delta Engineering			X							
Digital Displays			X							
EG&G	X			X			alpha-10 μ Ci			Neutron tissue dose det. LET prop. counter cal.
E. Vernon Hill			X							
Forestry Supplies			X							
Foxboro			X							
General Electric Space Technology Products	X				X					
Glenco Scientific										
Gould										
Honeywell	X			X		Not specifically stated	alpha			Trace mineral detection in the atmosphere prob- ably XRF. Also invest- igating source as static eliminator
Kahl Instrument		X			X					
Kahn			X							
Kistler Instrument			X							

TABLE II (con't)

MANUFACTURERS EMPLOYING RADIOISOTOPES IN GAUGING INSTRUMENTATION
(ABSORPTION)

FIRM	DISTRIBUTOR OR FABRICATOR EMPLOYING RADIOISOTOPES		AM-241		TYPE OF GAUGE OR INSTRUMENT	SOURCE AND ACTIVITY	DETECTOR	OUTPUT/ READOUT	APPLICATIONS
	YES	NO	YES	NO					
Kent Instrument		X			X				
Kratos Instrument				X					
ICC Cryogenics				X					
Instruments for Res. & Ind.		X			X				
James Electronics				X					
Lab-Items Associates		X			X				
Lab Line Instrums		X			X				
Landis & Gyr.	X			X	Moisture gauge	Am/Be 100- 300 mCi			Moisture measurement in concrete manufacture
					Thickness gauges Transmission and Backscatter	60 keV photon 10-2000 mCi	Depends on application	depends on customer needs	Thickness gauging and control for variety of continuous manufacturing processes.
					(e.g. gauge on cold rolling mill for steel plate C-frame)	300 mCi	Thermostati- cally con- trolled ion chamber	Analog sig- nal for line or point re- corders	Carbon steel thickness in continuous production line.
					Level gauge				Control of the heights of changes in blast furnaces filling heights in cupola furnaces.
					Promixity gauges				
LFE, Trapelo Division	X			X	C-frame thick- ness gauge for on- line process con- trol.	60 keV photon -- 1 C	Not infor- mation	Direct dig- ital read- out record- er outputs, others	Thickness meas. and con- trol in steel and copper brass manufacturing

TABLE II (con't)

MANUFACTURERS EMPLOYING RADIOISOTOPES IN GAUGING INSTRUMENTATION
(ABSORPTION)

FIRM	DISTRIBUTOR OR FABRICATOR EMPLOYING RADIOISOTOPES				TYPE OF GAUGE OR INSTRUMENT	SOURCE AND ACTIVITY	DETECTOR	OUTPUT/ READOUT	APPLICATIONS
	YES	NO	AM-241	YES NO					
Labtest Equip. Co.	X			X					
Livingston Atmometer			X						
Luft Instruments			X						
Manufactureres Eng. & Equipment		X		X					
Matheson Gas Products									
Microwave Instruments		X		X					
New Port of North Am- erica		X		X					
Nuclear Chicago		X		X					
Nuclear Enterprises Limited (England)	X			X	Portable gauge, moisture meas. and a combined moisture density probe	AM/Be 30 mCi 1 mCi, Cs-137 employed for density meas. in combined probe	³ He prop. counter and Li glass scint. EMI PM tube in the com- bined probe	Scalar spect- rometer 12" x12"x6.5" Digital read out.	Moisture measurements of soils building materials etc. Geological sur- veying
					On line fluid den- sity gauging sys- tems	60 keV photon		Meter and recorder output	Oil interface detection; control and meas. water content in cement slurry; control of mineral content in mineral floatation; meas. of specific gravity of various materials; fluid density
					On line thickness gauge and weight control meas.	60 keV photon (1-4 Ci)		A variety of readouts available	Production of steel strip and other materials con- tinuously produced.

* See also X-ray flourescent Equipment Survey.

TABLE II (con't)

MANUFACTURERS EMPLOYING RADIOISOTOPES IN GAUGING INSTRUMENTATION
(ABSORPTION)

FIRM	DISTRIBUTOR OR FABRICATOR EMPLOYING RADIOISOTOPES YES NO	AM-241 NO REPLY	TYPE OF GAUGE OR INSTRUMENT YES NO	SOURCE AND ACTIVITY	DETECTOR	OUTPUT/ READOUT	APPLICATIONS
Nuclear Enterprises Limited (England) Cont'd.				online moisture gauges	Am/Be 100-300 mC	A variety of inter-faces	Moisture content of coke, in blast furnace operations, in granular materials and the manufacture of paper and plastics.
Nuclear Radiation Development	X		X	Hole detector	Alpha source 2 mCi/in. 1"x2"xwidth of web investigated	Ion chamber Pen recorder meter readout	Meas. of holes in transparent or opaque materials
Nuclear Equipment		X					
Nuclear Equipment Chem		X					
Nuclear Sources & Services							
Oxford Instrument	X		X				
*Industrial Nucleonics (Accu-Ray)	X		X	On line meas. bulk density thickness measurement, level gauges	60 keV photon no details given. In-house design		Paper industry; Thickness measurement of steel aluminum and other metals; rubber and plastic industry controls; Meas. control and rejection system in cigarette manufacture; level control instru. variety of industries

TABLE II (con't)
MANUFACTURERS EMPLOYING RADIOISOTOPES IN GAUGING INSTRUMENTATION
 (ABSORPTION)

FIRM	DISTRIBUTOR OR FABRICATOR EMPLOYING RADIOISOTOPES		AM-241		TYPE OF GAUGE OR INSTRUMENT	SOURCE AND ACTIVITY	DETECTOR	OUTPUT/ READOUT	APPLICATIONS
	YES	NO	YES	NO					
				REPLY					
Industrial Dynamics	X		X		On-line Fill level gauges	NO INFORMATION SUPPLIED			On line measurement and control equipment for liquid level monitoring used by beverage manufact- urers, and others.
Ohmart, Corp.	X		X		Level control density and thickness gauges	(60keV photons) 100 to 1000 mCi. Am/Be 5x10 ⁶ n/sec			Industrial process control equipment
Pennsylvania Electronic Technology			X						
Photobell			X						
Photometric Data Systems			X						
Photovolt		X		X					
PMF Electronics			X						
Frank Petree			X						
Rank Precision Industries			X						
Scientific Instruments	X			X					
SGA Scientific									
Soil Moisture Equip.				X					
Soltec Corporation		X		X					
Spatial Data		X		X					

TABLE II (con't)
MANUFACTURERS EMPLOYING RADIOISOTOPES IN GAUGING INSTRUMENTATION
 (ABSORPTION)

FIRM	DISTRIBUTOR OR FABRICATOR EMPLOYING RADIOISOTOPES			AM-241		TYPE OF GAUGE OR INSTRUMENT	SOURCE AND ACTIVITY	DETECTOR	OUTPUT/ READOUT	APPLICATIONS
	YES	NO	NO REPLY	YES	NO					
Standard Controls			X							
Statham Instruments		X			X					
Testing Machines			X							
Texas Nuclear	X				X					
Trans Sonics			X							
Troxler Electronic Labs., Inc.	X			X		Moisture and den- sity measurement (portable)	8mCi, Cs-137 50 mCi, AmBe 125 n/s, Cs/ AmBe; 1000 mCi Am Be 2.5 n/s; 300 mc Am Be 7.5 n/s			Measurement of soil and asphalt density and soil moisture and asphalt content for highway con- struction industry and agriculture research
Studsuk Medical In- struments AB Atom- energi (Sweden)	X			X		Bone densito- meter	60 keV photon 10 mC and 100 mC	NaI(Tl) cry- stal 2 x 7 mm, type XP- 1110 PM tube	Digital and printout of bone width, equiv. min- eral content use of on- line comput- er	Measurement of bone den- sity and low mineral content.

TABLE III

SURVEY OF X-RAY FLUORESCENCE EQUIPMENT MANUFACTURERS

FIRM	EMPLOY RADIO- ISOTO- PES	AM- 241	TYPE OF INSTRU- MENT	SOURCE AND ACTIVITY	DETECTOR	SOURCE/ TARGET DETECTOR GEOMETRY	OUTPUT/ READOUT	APPLICATIONS/COMMENTS
Angstrom		No						Marketer for x-ray spect- rograph (Pitchford Ind.)
Applied Research Labs	Yes	No						
Atomic Energy of Canada *	Yes		Portable Analy- zer					
Baird -Atomic								
Bausch & Lomb								
Biomedical Systems								
Blake Industries	No							
Boyle Industrial Gauging Systems*	Yes		(Information not Obtained)					Zinc coating thick- ness gauges, process con- trol*
Canberra Industries	Yes	No						
Cartner Group Ltd. Mintec Division *	Yes		(Information Not Obtained)					ange of instru- mentation for continous analysis of minerals, ores and allied materials.*
CEA/Sac lay (France)	Yes	Yes	"Areil" Analyzer Portable	Alpha	windowless circulating gas pro- portional counter	Backscatter geometry cen- tral source inside gas	Pulse height analyzer, paper type punch	Portable Analyzer for determination of elements down to magnesium, α ex- citation employed for B, C, N, O, Ne.
CGEI Lepaute*	Yes		(Information Not Obtained)					XRF gauge for the measurement of Ca content of raw materials*

*Information obtained from IAEA Panel Report, not from direct mail.

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TABLE III (con't)
SURVEY OF X-RAY FLUORESCENCE EQUIPMENT MANUFACTURERS

FIRM	EMPLOY RADIO- AM- ISOTO- 241 PES		TYPE OF INSTRU- MENT	SOURCE AND ACTIVITY	DETECTOR	SOURCE/ TARGET DETECTOR GEOMETRY	OUTPUT/ READOUT	APPLICATIONS/COMMENTS
Landis-Gyr	Yes	Yes	On-line thick- ness gauge for Zn & Sn coatings on steel	(DETERMINED	BY APPLICA	TION)		Thickness coating on steel, on line process control.
Nuclear Associates								
Nuclear Chicago		No						
Nuclear Diodes								
Nuclear Enterprises	Yes	Yes*	Portable Ana- lyzer (Am-241 also noted as employed for backscatter source	Photon 60 keV 10 mCi (Tin determination	NaI	Central sou- rce backscat- ter geometry, balanced fil- ters	P.H.A. out- put; rate- meter	Alloy analysis; slag analysis; prospecting; analysis of cem- ent aggregate; ore analysis coating thickness measurement.
Nuclear Equipment Corporation	Yes	Yes	Si(Li) x-ray spectrometers	60 keV and Np x-rays up to 300 mCi also S/T assemb- lies	Si(Li)	Usually sour- ce annular configuration		Basic and industrial research; process control; analysis Mining metals and chemical in dust
Nuclear Semicon- ductor	Yes	Yes	Laboratory sol- id state ana- lyzer Si(Li)	Si(Li) top hat detector		Pulse height analyzer		Trace element analysis noted application in advertisement employs 20 mCi, Am-241 for trace analysis of elements "in-culto" and sockeye salmon
Nucleonic Data Systems, Inc.	Yes	Yes						U.S. Manufacturer of EIC coating thickness gauge (See EIC.)
Nucleometre*	Yes	Yes	Zinc coating gauge on-line	Photon (60KeV)	Proportion- al counter employs Al foil to filter out			Monitoring zinc coatings on steel sheets.

*Information obtained from IAEA Panel Report, not from direct mail.

**Isotopes and Radiation Technology 1970, Vol 8-1, P. 122-123.

*Information obtained from IAEA Panel Report, not from direct mail.

**Isotopes and Radiation Technology 1970, Vol 8-1, P. 122-123.

TABLE III (con't)
SURVEY OF X-RAY FLUORESCENCE EQUIPMENT MANUFACTURERS

FIRM	EMPLOY RADIO- ISOTO- PES	AM- 241	TYPE OF INSTRU- MENT	SOURCE AND ACTIVITY	DETECTOR	SOURCE/ TARGET DETECTOR GEOMETRY	OUTPUT/ READOUT	APPLICATIONS/COMMENTS
CGR Medical Columbia Scientific Industries	Yes	Yes	XRF analysis of air pollu- tants rock soil and plant sam- ples	Am-241/Ag S/T assemblies 14mCi * Am-241				Am-241 employed only as reference source.
EKCO Electronics LTD.								(See Nuclear Enterprises Ltd.)
EIC Elektronik tronische Instru- mente und Kontrol- leinrichtungen	Yes	Yes	XRF Analyzer Process control	60 Key photon up to 1 C.	Proportion- al counter *		SCA ana- lyzer Dig- ital scal- ar *	Coating thickness in meas- urements for steel, computer film industry.
Hilger and Watts, Ltd.*	Yes	Yes	Portable anal- yzer	Interchange- able sources	NaI Two filters	Central Sour- ce	Ratemeter with low level dis- criminator recorder socket	Field and laboratory applic- ations.
			Laboratory Analyzer	Interchange- able sources	NaI(Tl) interchan- geable filter slides	Central source	Ratemeter socket same as above	Field and laboratory applic- ations.
Honeywell, Inc.	Yes	Yes		Alpha Source				Unknown (related to trace mineral detection)
Humphrey Electronics								
KeveX Corp.**	Yes		Si(Li) solid state spectro- meters	Cd-109 photon source	Si(Li)	Side by Side backscatter geometry	Pulse height analyzer and atten. equip.	Applications, trace mineral analysis; ar authentication used Cd-109.

TABLE III (con't)
SURVEY OF X-RAY FLUORESCENCE EQUIPMENT MANUFACTURERS

FIRM	EMPLOY RADIO- ISOTO- PES	AM- 241	TYPE OF INSTRU- MENT	SOURCE AND ACTIVITY	DETECTOR	SOURCE/ TARGET DETECTOR GEOMETRY	OUTPUT/ READOUT	APPLICATIONS/COMMENTS
Ortec	Yes	Yes	Laboratory analyzer	60 keV photon 25 mCi alone & in S/T ass.	Si(Li) or Ge(Li)	Single channel analyzer log/linear ratemeter P.H.A. Computer interface capability		Analysis of materials; pollution analysis; artifact authentication.
Packard Instrument Co.	Yes	Yes	Tri-Carb-liq. scintillator spectrometers	60 keV photon 130 µC				External standardization of instrumentation
Panamet *	Yes		Portable analyzer	Interchangeable sources		Scintillation or proportional counter with interchangeable filters.	SCA with Digital Display	Field, laboratory, plant use
Philips Electronic		No						
Philips Medical Systems								
Pickar Medical Products								
Pickar Scientific Apparatus, Div.	No							
Princeton Gamma Tech.	Yes	Yes	XRF instruments for mineral mining processes; petroleum and chemical analysis	Gamma source 10-50 mCi S/T assembly 100 to 500 MCi of Am-241				Inspection and control of concentration of various elements in mineral, chemical, mining petroleum industry.

TABLE III(con't)

SURVEY OF X-RAY FLUORESCENCE EQUIPMENT MANUFACTURERS

FIRM	EMPLOY		TYPE OF INSTRUMENT	SOURCE AND ACTIVITY	DETECTOR	SOURCE/TARGET DETECTOR GEOMETRY	OUTPUT/READOUT	APPLICATIONS/COMMENTS
	RADIO-ISOTO-PES	AM-241						
Rigaku Denki Co., Ltd.	Yes	Yes	Portable analyzer	60 keV photon 30 mCi	Xe and Ar filled proportional counters	Backscatter geometry with filters	P.H.A. scalar, 5 digit ratemeter	Alloy Analysis, Sn coating, Zn coating.
Siemens								
Syntex Analytical Instruments								
Telsec Instrument Ltd.	Yes	No	Radioisotope non-dispersive analyzers	Interchangeable*	Proportional counters Be window*	Source sealed in sensing unit.	Integrating ratemeter	Portable analyzer analysis transition elements mining industry.
Texas Nuclear Div.	Yes	Yes	Series 9200 Portable analyzer	Alpha-photon source interchangeable*	Scintillation counter, two filters*		SCA with digital display attachment for MPHA*	Sn in ore samples.
Vacuum Technology Assoc.								Do not manufacture or design radioisotope XRF systems
Varian Associates								

VII Partial list of patents in which ^{241}Am is cited as a possible source of radiation*.

PATENT NUMBER AND DATE ISSUED	SUBJECT	TYPE OF PARTICLE USED
3,508,055 4/21/70	Security System utilizing radiation	Photons
3,263,082 7/26/66	Geological prospecting comprising directional irradiation and det- ection	Photons
3,562,531 2/9/71	Nucleonic Measuring ap- paratus with automatic fire safety rad. source, shutter closing and lock- ing means comprising a spring retained by a fusible Barrier which melts at high tempera- tures.	Photons/Betas
1,065,918 1967(British)	Improvements in or re- lating to apparatus for x-ray analysis	^{241}Am as a Bremsstrah- lung source.
3,448,264 6/3/69	x-ray source and measur- ing means for back sc- atter analysis of samples	Photons
3,402,709	Suit heater	Alphas
3,399,303 8/27/68	Radioactive metal cor- rosion evaluation and methods therefore	Alphas
3,328,508 6/27/67	Lighting rod with ionizing field	Alpha

PATENT NUMBER AND DATE ISSUED	SUBJECT	TYPE OF PARTICLE USED
1,017,595 1966 (British)	Improvements in or re- lating to radiometric analysis techniques	^{241}Am as a Bremsstrahlung source
3,353,170 11/14/67	Ionization fire alarm system	Alpha and photon
1,187,210 1970 (British)	Radiation source cap- sule	Alpha
Unknown	Method and apparatus for producing x- rays of particular wave lengths and applications therefore	Alphas
3,505,520 4/7/70	Measuring the incombustible content of Mine Dust using backscatter of low energy gamma rays	Photons
3,133,196 5/12/64	Radioactive heat source and container with He- lium permeable window in the container	Alpha
3,515,875 6/2/70	Alpha-Particle emit- ting radioisotope generator	Alpha
3,578,406 Date unknown (1971)	SO_2 Measurement System	Photon

PATENT NUMBER AND DATE ISSUED	SUBJECT	TYPE OF PARTICLE USED
864,656 (Can. Pat.) 2/23/71	Irradiation Device	Photons
3,497,691 2/24/70	Dual mode fluorescence and backscatter coating thickness measuring gauge	Photons
2,887,358 5/19/59	Large scale methods for the production and purification of Curium	Not applicable
3,463,619	Solvent extraction process for pur- ifying Americium and Curium	Not applicable
3,329,817 7/4/67	Radiation Source	
3,360,477 12/26/67	Neutron Source	
3,554,867 1/12/70	Method of obtaining Americium	
3,492,479 1/27/70	Apparatus for meas- uring hydorgeneous material utilizing radioactive source	Neutrons ($^{241}\text{Am}-\text{Be}$)
1,193,003 (British) 5/28/70	Improvement in or re- lating to devices for measuring the moisture content of a volume of material	<u>Neutron ($^{241}\text{Am}-\text{Be}$)</u> Photons
3,581,090 5/25/71	Utilizing penetrating radiation	Photons

PATENT NUMBER AND DATE ISSUED	SUBJECT	TYPE OF PARTICLE USED
3,389,257 6/68	Pulsed neutron source comprising a plurality of alpha sources and associated targets with a rotatable shu- tter there between	Alphas
3,460,917 8/12/69	Recovery of ^{241}Am from plutonium metals using molten salts	
3,445,201 5/20/69	Ion exchange process for recovering am- ericium and curium	
3,520,819 (Canadian)	High Energy gamma ray source	Alphas
824,227 (Canadian)	Fast Neutron Well Logg- ing	^{241}Am -Be neutron
3,546,454	Multiple Detector neu- tron Logging Apparatus	Neutron
1,034,539	Manufacture of Radio- Isotopes	
3,497,691	Dual made fluorescence and backscatter coating thickness measuring gauge	Photon
3,433,954 3/18/69	Semiconductor X-ray Em- ission Spectrometer	Photon

*Unless otherwise specified the patents were issued in the U.S.

PATENT NUMBER AND DATE ISSUED	SUBJECT	TYPE OF PARTICLE USED
1,065,919 (Canadian)	Improvements in or re- lating to Methods of X-ray Analysis	Photon