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ANNUAL PROGRESS REPORT
GAMMA-RAY SPECTROMETRY OF NEUTRON-DEFICIENT ISOTOPES

R. L. Heath and J. E. Cline

PHILLIPS
PETROLEUM
COMPANY

ATOMIC ENERGY DIVISION

NATIONAL REACTOR TESTING STATION
US ATOMIC ENERGY COMMISSION

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GAMMA-RAY SPECTROMETRY OF NEUTRON-DEFICIENT ISOTOPES**

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Atomic Energy Division

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U. S. ATOMIC ENERGY COMMISSION

ABSTRACT

The objective of this program is to produce a set of pulse-height spectra representing the response of a standard gamma-ray scintillation spectrometer to radiations emitted in the decay of neutron-deficient nuclides. These spectra will be processed and presented in the same manner as those presently included in the Scintillation Spectrometry Gamma-Ray Spectrum Catalogue which originally contained only spectra of neutron-induced activities. The purpose of this collection of data is to provide reference material for experimenters applying the techniques of scintillation spectrometry to the measurement of accelerator produced isotopes. Work during fiscal year 1964 has included the production of a large number of radioactive nuclides by gamma-ray and proton induced reactions. The methods employed for source production, the experimental equipment and measurement techniques are described in some detail. Spectra of over 20 neutron-deficient isotopes are presented in final form. These spectra will be included in a new edition of the Spectrum Catalogue presently being prepared.

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I. INTRODUCTION AND OUTLINE OF PROGRAM

The purpose of this program is to provide basic quantitative information on the radiations emitted in the decay of neutron-deficient isotopes. A major objective is the compilation of gamma-ray spectra of these nuclides. These spectra, obtained under well-defined measurement conditions, provide reference data for experimenters applying scintillation spectrometry to routine problems in the measurement of radiation. These data will be included in supplements to the "Scintillation Spectrometry Gamma-Ray Spectrum Catalogue" which is an information service of the Radioactivity and Decay Schemes Group at the National Reactor Testing Station.

In most applications of radioisotopes it is necessary to utilize some technique for quantitative determination of individual radioactive isotopes. This usually implies the use of an instrumental technique in combination with radiochemical procedures. In the past few years there has been an increasing application of gamma-ray scintillation spectrometry for this purpose. Analogous to the techniques used in emission spectroscopy, the nuclear spectroscopist relies quite heavily on the availability of information on the response of the spectrometer to a given radioactive isotope.

To promote the use of gamma-ray scintillation spectrometry for routine isotopic analysis a program was initiated in this laboratory several years ago to develop standard measurement techniques and to produce an atlas of "standard spectra" for each isotope. Because of the nature of the experimental program and the availability of nuclear reactors, the original emphasis was placed on those isotopes which are produced by neutron capture or fission. In 1958 the original Spectrum Catalogue [1] was issued. This catalogue contained spectra of radioactive isotopes in the form of pulse-height distributions obtained with a standardization scintillation spectrometer. Since that time the cataloguing of spectra has continued.

[1] R. L. Heath, Scintillation Spectrometry Gamma-Ray Spectrum Catalogue, IDO-16408 (1958).

As a result of recent developments, interest in neutron-deficient nuclides has increased. For example, there has been an increase in the application of activation analysis techniques using small neutron generators. These machines provide a convenient method for producing radioisotopes by $(n,2n)$, (n,p) , and (n,α) reactions. The development of high-flux reactors with fission neutron fluxes in excess of 10^{15} n/cm²/sec also will result in reactor production of sizable quantities of radionuclides by the same reactions. Since the methods of gamma-ray spectrometry can be applied to the assay for these nuclides, there is an immediate need for information on the gamma-ray spectra and decay characteristics of these nuclides. For this reason it was deemed desirable to extend the spectrum cataloguing program to include information on these nuclides. With the support of the Isotope Technology Branch, Division of Isotopes Development, U. S. Atomic Energy Commission, this program was extended to include the gamma-ray spectra of neutron-deficient isotopes.

To produce these nuclides it is convenient to use particle accelerators. Most isotopes of interest can be produced with photonuclear, proton or alpha particle reactions. The choice of machines to be used for source production is discussed below.

Electron Linear Accelerators

These machines can be used for the production of radioisotopes by photonuclear reactions. The (γ,n) reaction can be used to form nuclides normally produced in reactors and neutron generators by the $(n,2n)$ reaction. The photonuclear reaction has the advantage of reduced interference from the products of neutron capture. Although these same isotopes can be produced in many cases by particle reactions in a cyclotron, the target problems are much more severe. In many cases the irradiations are simple to make and result in relatively clean samples requiring no chemical purification.

A study of the availability of machines of this type led to the choice of the 45-MeV accelerator operated by the General Atomic Division of General Dynamics Corporation in San Diego, California. This machine is operated on a production schedule and facilities were available for all phases of the experimental program.

Cyclotron

Although reactions involving protons, deuterons, and alpha particles can be induced using a cyclotron, the most useful reactions are (p,n) and $(p,2n)$. The Production Cyclotron at the Oak Ridge National Laboratory is a 22.5-MeV

fixed-energy machine with extremely high beam current capabilities. It is planned to produce many of the desired isotopes in this machine. In cases where it might be advantageous to produce nuclides by alpha-particle reactions, the possibility of using the cyclotrons at the Argonne National Laboratory and the University of Colorado has been investigated. This alternative was necessary since the ORNL machine is used exclusively for proton productions.

Van de Graaff Accelerators

The major advantage of the Van de Graaff generator is an easily accessible external beam of protons. Very effective use of such a machine can be made for the study of very short-lived proton-induced activities. For this purpose, the possible use of the Van de Graaff facilities of The Rice Institute at Houston, Texas, has been investigated. Machines at this laboratory will be used for studies which cannot conveniently be conducted on the other accelerators.

During this fiscal year arrangements were completed with the General Dynamics Corporation and with the Oak Ridge National Laboratory for the use of the linear accelerator and the production cyclotron for source production. Laboratory facilities have been made available at the accelerator sites and complete gamma-ray scintillation spectrometers installed for use at these sites.

A tentative schedule was established for accelerator irradiations to facilitate the design of experiments. The work was divided into four phases:

- (1) Source material procurement and target fabrication,
- (2) production of long-lived isotopes (half-life greater than one day),
- (3) production of medium half-life isotopes (greater than 30 minutes), and
- (4) production of short-lived isotopes (less than 30 minutes).

This division was made to make the best use of available instrumentation and manpower. The long-lived isotopes were produced in the accelerators and shipped back to this laboratory for study. This required almost no labor at the accelerator site and no transportation of equipment to the accelerator. For the shorter-lived isotopes it is necessary to make all measurements and chemical purifications at the accelerator site. The study of short-lived isotopes requires the development and installation of pneumatic rabbit facilities and special source preparation techniques. Most of the long-lived isotopes of interest which could be produced efficiently in the electron linear accelerator have been studied and work is in progress on the production of short-lived activities in this machine.

II. EXPERIMENTAL MEASUREMENTS

In order to provide a set of standard pulse-height spectra for the Gamma-Ray Spectrum Catalogue for comparison with experimental results in other laboratories, the spectrometer and methods used for the experimental measurement of spectra have been standardized. The scintillation spectrometer, including detector, detector shield, source-detector geometry, source preparation, and operating conditions for the electronic equipment, is completely specified for all measurements. To provide the experimenter with an understanding of the reasons for the choice of these standard conditions, the scintillation spectrometer used for all measurements will be described in some detail. The procedure followed to determine isotopic purity will be discussed briefly. Finally, the methods employed for data reduction and preparation of the final spectra will be described.

1. STANDARD LABORATORY SPECTROMETER

1.1 Detector

The 3- x 3-inch NaI(Tl) cylinder was originally chosen as the standard reference detector for all spectra in the spectrum catalogue. The choice of this size detector was a compromise based upon consideration of many factors. This size detector is obtainable from commercial manufacturers with acceptable energy resolution (<8 percent for the Cs-137 gamma ray). The volume of a 3- x 3-inch cylinder of NaI is sufficient to give a reasonable photopeak response in the energy region associated with radioactivity. To obtain appreciable improvement in the photopeak fraction would require increasing the detector size to a 5- x 5-inch cylinder or larger. The larger detectors not only are considerably more expensive but exhibit much poorer energy resolution.

The detector assembly used for all catalogue spectra is shown in Figure 1.

Fig. 1 3 x 3 in. laboratory scintillation detector.

The NaI crystal is contained in a thin-walled aluminum can (0.005-inch wall thickness) to reduce absorption of low-energy photons and Compton scattering from the can. The optical reflector is a (0.005-inch thick) sprayed coating of α -alumina. The crystal is mounted directly on the face of a 3-inch diameter phototube and the can is then evacuated. A mu-metal shield around the dynode structure of the phototube reduces the effects of stray magnetic fields. A more complete description of the detector is given in the original edition of the spectrum catalogue ^[1]. Energy resolution of detectors used in these measurements ranged from 7.5 to 8.0 percent for the Cs-137 gamma ray.

1.2 Detector Shield

Usual laboratory conditions require a radiation shield to reduce the background radiation level to a point where corrections to the data will be small for normal strength sources (1000 to 5000 c/sec). Unfortunately, the use of such a shield produces a serious problem due to scattering of gamma rays from the source off the walls of the detector shield. This scattered radiation produces an undesired distortion in the spectrum. The standard shield configuration which has been adopted, as shown in Figure 2, has inside dimensions 32 x 32 x 32 inches. This shield is constructed of 2- x 4- x 8-inch lead bricks to provide 4-inch thick walls and a 2-inch thickness of lead on top and bottom. A detailed discussion of the various factors influencing detector shield design is given in Reference 1.

1.3 Source-Detector Geometry

For comparison, a well defined source-detector geometry must be chosen since the detailed shape of a pulse-height spectrum for a given isotope is dependent upon the solid-angle intercepted by the detector. Major geometry-dependent effects are discussed in the following paragraphs.

1.31 Photopeak Efficiency. The shape of the pulse distribution representing detector response to a monoenergetic source of radiation will be dependent upon geometry. For a large NaI detector the photopeak results from both photoevents and multiple processes which result in total energy loss in the detector. The probability for the occurrence of multiple events depends upon the path length traversed by a photon in passing through the detector. Since the average path length will depend on source-detector geometry, the shape of the spectrum will change with source-detector distance.

1.32 Coincidence Effects. In most cases the decay of a single nucleus gives rise to two or more coincident gamma rays. There is a finite probability that

Fig. 2 Detailed view of laboratory detector shield.

more than one gamma ray from the decay of this nucleus will enter the detector. The detector output in such a case will be the sum of the energies lost in the detector by each of the photons. This results in a distribution of pulses extending in energy from zero to the sum of the energies of the two coincident gamma rays. This distribution is called the coincidence sum spectrum. Figure 3 shows the spectrum of 20,000-yr Nb-94 which decays with the simultaneous emission of two gamma rays (0.703 and 0.870 MeV). Above the two primary photopeaks the coincidence sum spectrum extends up to the sum of the energies of the two gamma rays.

The probability for detecting two coincident photons in one detector will vary as the square of the solid angle while the probability for detecting a single event will be proportional to the solid angle. Under conditions of large solid angle, analysis of complex spectra could be complicated by the coincidence sum spectrum. The standard configuration adopted was a source-detector distance of 10.0 cm. This solid angle provides reasonable detection efficiency with negligible contributions from coincidence summing in most cases.

1.33 Source Holder. The detection of scattered radiation from material in the immediate vicinity of the source and the detector will result in undesired distortion of the gamma-ray spectrum. For this reason, material used in the source holder and the mass of the source itself should be kept to a minimum. The standard detector, base, and source-holder arrangement are shown in Figure 4. Source mounting cards are positioned 10 cm from the detector by a lattice of four 0.010-inch diameter aluminum wires suspended from the aluminum ring structure.

1.34 Beta Absorber. To prevent the detection of beta particles emitted in the decay of the source, a beryllium absorber (3-inch diameter) is placed directly on the detector. The thickness of absorber used in a given case is dictated by the energy of beta radiation emitted by a given nuclide. The usual values used are 0.164, 0.598, 1.118, or 1.716 g/cm² of Be. The position of the absorber and the choice of Be was made to reduce the production of external bremsstrahlung which results from absorption of the beta radiation.

1.35 Source Preparation. Sources are prepared to approximate a weightless point source as nearly as possible. The source material is deposited between layers of cellulose tape (≈ 1 mg/cm²) and mounted on paper cards 2-1/2-x 3-1/4-inch with a 1-1/2-inch diameter hole punched in the center. In the case of positron emitters, annihilation radiation, emitted largely when the positron comes to rest, complicates the source preparation problem. If the same source-detector geometry is desired for the annihilation quanta as for all other photons

Fig. 3 Spectrum of Nb-94 showing effect of coincidence summing.

Fig. 4 Diagram of detector base and source holder arrangement.

emitted in the decay of the source, then the positrons must be annihilated at the source. Although it results in some deterioration in the quality of the spectrum due to Compton scattering, the convention adopted is to surround the source with copper foil of sufficient thickness to annihilate a majority of the positrons. The amount of absorber used for this purpose is varied with positron energy.

In the production of source material, methods are employed to produce high specific activity; carrier-free chemistry is used when applicable to reduce the mass of inert material in the source.

1.4 Electronic Equipment

The standard spectrometer used for the collection of all pulse-height spectra is shown as a block diagram in Figure 5. The output of the detector is fed to a double-delay line linear amplifier of the type developed by G. G. Kelley of the Oak Ridge National Laboratory [2]. This amplifier provides a bi-polar output pulse which virtually eliminates bias shift problems with changing count rate. It provides better than 0.1 percent integral linearity and excellent gain stability.

The multichannel pulse-height analyzer used is a Nuclear Data Inc. machine (model 130 AT) with arithmetic capabilities and perforated paper tape read-in and read-out. This analyzer provides excellent linearity and stability of zero and ramp slope in the analogue-to-digital convertor. The perforated tape system employs an 8-level IBM tape code which is universally applicable for computer applications.

A detailed outline of test and calibration procedures used at this laboratory to check multichannel analyzers and spectrometer systems is given in reference [3].

Fig. 5 Block diagram of laboratory scintillation spectrometer.

[2] E. Fairstein, Pulse Amplifier Manual, ORNL-3348 (1962)

[3] D. F. Crouch and R. L. Heath, Routine Testing and Calibration Procedures for Multichannel Pulse Analyzers and Gamma-Ray Spectrometers, IDO-16923 (1963).

2. METHODS OF DATA REDUCTION

2.1 Source Production

The production of a pulse-height distribution representing the response of the standard spectrometer to the gamma-ray spectrum of a given nuclide is a process involving many separate steps. First, the possible methods for producing the isotope in question are studied. Factors to be considered include: interference from other isotopes of the same element, half-life, possible elemental contamination of the material to be irradiated, and production cross-sections for various nuclear reactions. The first decision made is the choice of the nuclear reaction to be used. Since it is not usually possible to produce only one nuclide as a result of irradiation of a target in an accelerator, operating parameters for the accelerator are chosen which result in enhanced relative production of the desired nuclide. Variables include bombarding particle energy, irradiation time, and target material. The chemical form of the target material will depend upon chemical procedures to be used in the purification of the sample following irradiation and interference from other activities.

Prior to the production of isotopes in the linear accelerator and cyclotron it was necessary to investigate methods of target preparation and to become familiar with the problems characteristic of bremsstrahlung and proton irradiations. Considerable attention was given to possible interference from neutrons produced in some abundance by both types of accelerators. Certain target materials, designed to yield information on these problems, were irradiated and the resulting reaction products studied in some detail.

Where irradiation of the natural element rather than a mass-separated isotope seemed feasible, a preliminary irradiation was made and the activities produced were examined on the scintillation spectrometer. In many cases obvious elemental contaminants were removed by chemical separation and pure samples of the desired element were prepared. Spectra of these samples were obtained and the spectra were analyzed to determine the energy and intensity of all gamma rays observed. Repeated measurements provided half-life information. Comparison of the observed spectra with available nuclear data usually permitted identification of all isotopes present in a given sample. If no interference from other isotopes were detected, sources were prepared in the standard manner and the spectrum measured under the experimental conditions previously described. In all measurements source strengths were adjusted to provide a

total input counting rate ranging from 1000 to 3000 counts/sec. This restriction was imposed to reduce distortion of the spectrum by random summation of uncorrelated events occurring within the resolving time of the linear amplifier and analyzer. At this input rate the magnitude of the "random sum spectrum" will be a factor of 1000 less than the spectrum due to single events.

In many cases other radioisotopes of the same element were present. If they were present in small quantities relative to the desired isotope and had half-lives considerably longer, the source was allowed to decay and the spectrum of the contaminant was obtained. The contribution from the longer-lived isotope was then subtracted from the original spectrum after normalization of the gain scale and correction for decay. In some cases interference from the production of the undesired isotopes made it necessary to use mass-separated material to obtain a pure spectrum. For isotopes with half-lives shorter than 24 hours it was frequently necessary to make several irradiations to solve all problems of contamination and to obtain sufficient data to ensure sample purity. In many cases an isotope may decay to a radioactive daughter. In these cases the spectrum was measured immediately following chemical separation of the daughter. If the half-life of the daughter is less than 30 minutes the convention adopted was to use the spectrum of the equilibrium mixture.

2.2 Energy Calibration

All spectra in the catalogue are measured using standard energy scales which are related to the energy of the gamma ray emitted in the decay of Cs-137 (0.66162 MeV). The scales are nominally 5, 10, and 20 keV per channel on the multichannel pulse-height analyzer. The calibration procedure takes into account the nonlinear energy response of NaI. The method used to establish the energy scale is described in Reference 3. Precise energy calibration for each spectrum is obtained by measuring a source simultaneously with a Cs-137 source or by using the 0.511-MeV annihilation radiation for sources which decay by positron emission. Although the spectrometers are frequently calibrated with Cs-137 sources, it is usually not practical to set this scale to an accuracy better than 1 percent. The spectrum obtained together with Cs-137 is analyzed with a least squares computer program [4] to determine the position of the Cs-137 photopeak relative to a prominent, well-resolved photoline attributed to the isotope in

[4] R. L. Heath, Recent Developments in Scintillation Spectrometry, IRE Trans. on Nuclear Science, Vol. NS-9, No. 3 (June 1962).

question. The true position of this photopeak on the standard gain scale (eg, 10 keV/channel) may then be determined. A spectrum of the pure isotope is then shifted to place this photopeak at the correct position on the pulse-height vs energy scale using a computer program described in Reference 4. The spectrum is then analyzed to obtain the energies of all major gamma rays for comparison with energy values previously reported in the literature. If more precise energy values have been measured on conversion electron spectrometers or from inelastic scattering, these are used for the final spectrum. If not, the values obtained from this laboratory's analysis are reported.

2.3 Intensity Normalization

In most cases the spectrum is in final form following the gain scale determination. In some cases, however, 4π β - γ coincidence measurements are made on samples of a pure isotope to obtain an absolute decay rate. The same sample is then measured on the gamma-ray spectrometer and a quantitative determination made of the emission rate of a major gamma ray. In cases where the decay scheme for this nuclide is not well established, this information permits the normalization of the intensity of the spectrum to a known decay rate. It is proposed that the branching ratios for major gamma rays be re-measured where possible to provide more accurate information for quantitative assay.

2.4 Data Presentation

The final data are prepared in three forms: (a) a digital listing of the number of counts in each channel of the spectrometer, (b) a point plot (semi-log) of number of counts/channel vs channel number, and (c) perforated paper tapes in 8-level IBM code. The graphs are prepared as templates to overlay the standard 8-1/2 x 11 or 11 x 14 inch K and E 3-cycle semi-log graph paper. Experimental data obtained on any spectrometer may be plotted in this form for comparison. The digital listing of data is provided to permit the experimenters to prepare catalogues of spectra on perforated tape for entry to the core memories of modern pulse-height analyzers. Section III of this report contains spectra which have been compiled during this fiscal year.

3. IRRADIATIONS IN THE ELECTRON LINEAR ACCELERATOR

3.1 Linear Accelerator Facility

During this fiscal year most of the neutron-deficient nuclides studied were produced by photonuclear reactions. These irradiations were made using the

bremsstrahlung beam of the electron linear accelerator operated by the General Atomic Division of General Dynamics Corporation.

A cut-away drawing of the linear acceleration facility is shown in Figure 6. Electrons are ejected into this machine at point 1 as shown in the figure and are accelerated to a maximum of 45 MeV by three Klystron cavities shown at positions 2, 3, and 4. The high energy electron beam is then passed through an analyzer magnet shown at position 5. This magnet is used for energy analysis and to direct the beam into various beam tubes which serve different experimental positions. The energy measurement is accomplished through a determination of the magnetic field required to deflect the beam through a known angle into the tuning port, located at position 6. The tuning-port position has been made available for bremsstrahlung production for this program. The other beam tube facilities shown in the figure are used by other experimenters.

Fig. 6 Cutaway drawing of linear accelerator facility.

For use in this program, the tuning-port facility was terminated about two feet from the analyzer magnet. The beam was directed through a 0.020-inch thick aluminum window into a 1/8-inch thick tungsten alloy water-cooled bremsstrahlung convertor. The particular alloy used was chosen to dissipate the extreme power densities produced by the machine. Normally, an average power of 12 kW is

injected into the convertor by a 2-mm diameter electron beam. Since the cross section for the production of bremsstrahlung is proportional to Z^2 , tungsten serves as a good material for the radiator.

The energy distribution of the bremsstrahlung from monoenergetic electrons of energy E_0 may be approximated by

$$\phi(E)dE = \phi_0 [1 - a(E_0)E] dE.$$

With the high energy photons available from this machine several different photonuclear reactions are possible. Figure 7 shows energy thresholds and cross sections for a typical photonuclear reaction. Curve A shows the (γ, n) reaction cross section. Curve B shows the combined $(\gamma, 2n) + (\gamma, 3n)$ cross sections. The cross section for the (γ, n) reaction exhibits a resonance 3 to 8 MeV in width located from 4 to 15 MeV above the threshold for the particular reaction. Integrated cross sections for the (γ, n) ,

Fig. 7 Typical plot of photonuclear cross sections. Curve A shows the cross section for the γ, n reaction. Curve B shows the cross section for $\gamma, 2n$ plus $3/2 \gamma, 3n$. Also shown are the thresholds for γ, n , γ, np , $\gamma, 2n$, and $\gamma, 3n$ reactions. These data are taken from R. L. Bramblett et al, Phys. Rev. **129**, 2723 (1963).

$(\gamma, 2n)$, and $(\gamma, 3n)$ reactions increase with nuclear mass while the cross section for the (γ, p) reaction remains more or less constant. For this reason the (γ, p) process is almost negligible in the study of the heavier nuclides.

For the production of a particular radioactive nuclide the operating energy of the accelerator was selected to enhance the relative production of the desired activities. In most cases these activities were produced by the (γ, n) reaction and the machine was operated at reduced energy. In those cases where the nuclides to be produced were to be made by the $(\gamma, 2n)$ and $(\gamma, 3n)$ reactions the machine energy was increased to enhance production by these reactions. The remaining operating parameters of the accelerator were set to maximize bremsstrahlung intensity. With the machine operating at 27 MeV, the average intensity of the bremsstrahlung beam was 10^{13} photons/sec. The angular divergence of the bremsstrahlung beam may be approximated by

$$\Omega = \frac{m_0 c^2}{E_0},$$

where E_0 is the energy of the electron beam. At a distance of 2 inches from the bremsstrahlung converter, the average photon flux density is about 2×10^{12} photons/cm²/sec.

3.2 Sample Holder

The sample holder used for most irradiations was designed to provide for the simultaneous irradiation of seven samples. Each irradiation position in the holder received approximately the same bremsstrahlung intensity when the holder was located about 1-1/2 inch from the converter. If fewer samples were to be activated in a given machine run, the holder could be moved closer to the converter to provide higher photon flux. The sample holder is shown in Figure 8. It was constructed of aluminum and cooled by passing water throughout the entire volume. Capsules containing the source material in powdered form were inserted into the holder and cooled by conduction. The capsules also were made of pure aluminum and fabricated as shown in Figure 9. Two types were used. One of these, as shown in the figure, was double-ended, allowing for the irradiation of two different samples in the same capsule. This type was used primarily for the production of short-lived activities and for the irradiation of mass-separated isotopes. Each end of the capsule could

Fig. 8 Cutaway view of sample holder used in linear accelerator irradiations.

Fig. 9 Drawing of capsule used in linear accelerator irradiations.

hold up to 40 mg of material. When larger quantities of material were required, a single-ended capsule was used. The type of construction used for these capsules permitted quick access to the irradiated material. This feature was important for short half-life isotopes. Aluminum was chosen for the fabrication of both capsules and sample holder since activation of aluminum produces primarily a 7-second activity. The quick decay of this activity reduced the radiation hazard to personnel involved in handling the samples after irradiation.

3.3 Experimental Problems

Major difficulties encountered in these irradiations resulted from the production of a high flux of fast and epithermal neutrons in the environment of the samples. These neutrons are produced by photonuclear reactions in the bremsstrahlung converter, the sample holder, and in the samples themselves. Some moderation of these neutrons occurred in the cooling water of the converter and sample holder. Some samples contained contaminant activities produced by neutron capture. This phenomenon was most troublesome in those elements which had high resonance or fast neutron cross sections. Particular examples were indium, molybdenum, and europium. In most cases interference from neutron-induced activities could be reduced considerably by the use of mass-separated isotopes. Mass-separated isotopes also were of considerable value in reducing the presence of undesired activities resulting from photonuclear reactions on isotopes other than those of interest.

Table I shows a list of samples irradiated during the past year at the linear accelerator facility. Included in the table are the sample material, expected activities, half-lives, and the mode of production. Five series of irradiations were made from November 1962 through July 1963. Following these irradiations, the capsules were opened and the material transferred to clean polyethylene containers. At this point, the activity level and purity of each sample was determined on the gamma-ray spectrometer. Chemical purification was performed on samples which showed evidence of contaminant activities. Final samples were then prepared for counting.

TABLE I

SAMPLES IRRADIATED AT THE LINEAR ACCELERATOR FACILITY

Sample Material	Quantity	Isotope Produced	$T_{1/2}$	Reaction	Nov. 1962	Feb. 1963	April 1963	May 1963	June 1963
Graphite	5 mg	C ¹¹	20.4 m	(γ, n)			x	x	x
NH ₄ Cl	1 mg	Cl ^{34m}	32.4 m	(γ, n)			x		x
K ₂ SO ₄	20 mg	K ³⁸	7.8 m	(γ, n)	x		x	x	x
Sc ₂ O ₃	20 mg	Sc ⁴⁴	4 h	(γ, n)			x		
		Sc ^{44m}	2.4 d	(γ, n)			x		
TiO ₂	30 mg	Ti ⁴⁵	3 h	(γ, n)			x		
Cr ⁵⁰	25 mg	Cr ⁴⁹	42 m	(γ, n)			x		
Fe	200 mg	Fe ⁵²	8.2 h	($\gamma, 2n$)			x	x	x
		Fe ⁵³	8.9 m	(γ, n)			x		x
Ni	100 mg	Ni ⁵⁶	6.4 d	($\gamma, 2n$)		x			
		Ni ⁵⁷	37 h	(γ, n)		x			
CuO	10 mg	Cu ⁶¹	3.3 h	($\gamma, 2n$)			x		
		Cu ⁶²	9.8 m	(γ, n)			x		
ZnO	14 mg	Cu ⁶⁷	59 h	(γ, p)			x		
		Zn ⁶²	9.3 h	($\gamma, 2n$)			x		
		Zn ⁶³	38.3 m	(γ, n)			x		
Ga ₂ O ₃	140 mg	Ga ⁶⁷	78 h	($\gamma, 2n$)			x		x
		Ga ⁶⁸	68 m	(γ, n)			x		
GeO ₂	40 mg	Ge ⁶⁷	18.6 m	($\gamma, 3n$)		x			
		Ge ⁶⁸	275 d	($\gamma, 2n$)		x			
		Ge ⁶⁹	39.6 h	(γ, n)		x			
SeO ₂	110 mg	Se ⁷¹	4.5 m	($\gamma, 3n$)			x		
		Se ⁷²	8.5 d	($\gamma, 2n$)			x		
		Se ⁷³	7.1 h	(γ, n)			x		
NH ₄ Br	40 mg	Br ⁷⁷	58 h	($\gamma, 2n$)			x		
		Br ^{78m}	6.4 m	(γ, n)			x		
RbCl	115 mg	Rb ⁸³	83 d	($\gamma, 2n$)		x			
		Rb ^{82m}	23 m	(γ, n)		x			
		Rh ⁸²	33 d	(γ, n)		x			
Sr ⁸⁴	4 mg	Sr ⁸³	36 h	(γ, n)			x		
Y ₂ O ₃	100 mg	Y ^{87m}	14.6 h	($\gamma, 2n$)			x		
		Y ⁸⁷	80 h	($\gamma, 2n$)			x		
ZrO ₂	10 mg	Zr ⁸⁷	94 m	($\gamma, 3n$)			x		
		Zr ⁸⁸	85 d	($\gamma, 2n$)			x		
		Zr ^{89m}	4.4 m	(γ, n)			x		
		Zr ⁸⁹	79 h	(γ, n)			x		
Nb	50 mg	Nb ^{82m}	13 h	(γ, n)			x		
		Nb ⁹²	10.1 d	(γ, n)			x		
(NH ₄) ₆ Mo ₇ O ₂₄	40 mg	Mo ⁹¹	15.5 m	(γ, n)			x	x	
		Mo ^{93m}	6.8 h	(γ, n)			x	x	
Ru	30 mg	Ru ⁹⁴	57 m	($\gamma, 2n$)				x	x
		Ru ⁹⁵	98 m	(γ, n)				x	x
Rh	48 mg	Rh ¹⁰⁰	20.8 h	($\gamma, 3n$)			x		
		Rh ^{101m}	4.7 d	($\gamma, 2n$)			x		
		Rh ¹⁰²	210 d	(γ, n)			x		

TABLE I (Continued)

SAMPLES IRRADIATED AT THE LINEAR ACCELERATOR FACILITY

Sample Material	Quantity	Isotope Produced	T _{1/2}	Reaction	Nov. 1962	Feb. 1963	April 1963	May 1963	June 1963
Ag ₂ O	75 mg	Ag ¹⁰⁵	40 d	(γ , 2n)		x	x	x	x
		Ag ^{106m}	8.3 d	(γ , n)		x	x	x	x
		Ag ¹⁰⁸	24 m	(γ , n)		x	x	x	x
In ₂ O ₃	20 mg	In ^{112m}	20.7 m	(γ , n)			x		
		In ¹¹²	14.5 m	(γ , n)			x		
Sb ¹²¹	10 mg	Sb ^{120m}	5.8 d	(γ , n)			x		
		Sb ¹²⁰	16.4 m	(γ , n)			x		
Cs ₂ O ₃	60 mg	Cs ¹³⁰	30 m	(γ , 3n)		x			
		Cs ¹³¹	10 d	(γ , 2n)		x			
		Cs ¹³²	6.5 d	(γ , n)		x			
La ₂ O ₃	80 mg	La ¹³⁸	9.5 m	(γ , 3n)				x	
Pr ₂ O ₃	120 mg	Pr ¹³⁹	4.3 h	(γ , 2n)				x	x
		Pr ¹⁴⁰	3.4 m	(γ , n)				x	x
Nd ₂ O ₃	70 mg	Nd ¹⁴⁰	3.3 d	(γ , 2n)				x	x
		Nd ¹⁴¹	2.4 h	(γ , n)				x	x
Sm ₂ O ₃	40 mg	Sm ¹⁴²	72 m	(γ , 2n)				x	
		Sm ¹⁴³	8.3 m	(γ , n)				x	
Eu ₂ O ₃	120 mg	Eu ¹⁴⁸	120 d	(γ , 2n)				x	
		Eu ^{150m}	5 y	(γ , n)				x	
		Eu ¹⁵⁰	13.7 h	(γ , n)				x	
		Eu ^{152m}	9.2 h	(γ , n)				x	
		Eu ¹⁵²	13 y	(γ , n)				x	
Tb ₂ O ₃	300 mg	Tb ^{158m}	5.5 h	(γ , 3n)				x	
		Tb ¹⁵⁸	5.2 d	(γ , 3n)				x	
Ho ₂ O ₃	40 mg	Ho ¹⁶⁴	37 m	(γ , n)				x	x
Er ₂ O ₃	80 mg	Er ¹⁶⁰	30 h	(γ , 2n)					x
		Er ¹⁶¹	3.5 h	(γ , n)					x
Tm ₂ O ₃	150 mg	Tm ¹⁶⁸	7.7 h	(γ , 3n)				x	
		Tm ¹⁶⁷	9.6 d	(γ , 2n)				x	
Lu ₂ O ₃	600 mg	Lu ¹⁷²	6.7 d	(γ , 3n)	x				
		Lu ¹⁷³	200 d	(γ , 2n)	x				
		Lu ^{174m}	165 d	(γ , n)	x				
HfO ₂	80 mg	Hf ¹⁷³	23.6 h	(γ , n)				x	x
WO ₂	80 mg	W ¹⁷⁸	22 d	(γ , 2n)				x	x
		W ^{178m}	6 m	(γ , n)				x	x
		W ¹⁷⁹	40 m	(γ , n)				x	x
Au	60 mg	Au ¹⁹⁵	185 d	(γ , 2n)		x			
		Au ^{196m}	10 h	(γ , n)		x			
		Au ¹⁹⁸	5.6 d	(γ , n)		x			

III. INDEX OF GAMMA-RAY SPECTRA

<u>Half-Life</u>	<u>Isotope</u>	<u>Energy Scale</u>	<u>Production Method</u>	<u>Plate No.</u>
32.4 min	Cl-34m	2 keV/PHU	γ, n	17-34m(17-34)1
7.7 min	K-38	2 keV/PHU	γ, n	19-38-2
42 min	Cr-49	1 keV/PHU	γ, n	24-49-1
9 min	Fe-53	1 keV/PHU	γ, n	26-53-1
36 hr	Ni-57	1 keV/PHU	γ, n	28-57-1
61 hr	Cu-67	0.5 keV/PHU	γ, p	29-67-1
78 hr	Ga-67	1 keV/PHU	$\gamma, 2n$	31-67-1
40 hr	Ge-69	1 keV/PHU	γ, n	32-69-1
18 day	As-74	1 keV/PHU	γ, n	33-74-1
7.1 hr	Se-73	1 keV/PHU	γ, n	34-73-1
58 hr	Br-77	1 keV/PHU	$\gamma, 2n$	35-77-1
20 hr	Tc-95	1 keV/PHU	γ, n, β	43-95-1
60 day	Tc-95m	1 keV/PHU	γ, n, β	43-95m(43-95)1
1.7 hr	Ru-95	1 keV/PHU	γ, n	44-95-1
4.7 day	Rh-101m	1 keV/PHU	$\gamma, 2n$	45-101m-1
40 day	Ag-105	1 keV/PHU	$\gamma, 2n$	47-105-1
24 min	Ag-106	1 keV/PHU	γ, n	47-106-1
16 min	Sb-120	1 keV/PHU	γ, n	51-120-1
5.8 day	Sb-120m	1 keV/PHU	γ, n	51-120m-1
6.5 day	Cs-132	1 keV/PHU	γ, n	55-132-1
4.5 hr	Pr-139	1 keV/PHU	$\gamma, 2n$	59-139-1
37 min	Ho-164	0.5 keV/PHU	γ, n	67-164-1
6.1 day	Au-196	1 keV/PHU	γ, n	79-196-1

9 min Fe^{53}
3" x 3" - 3 Na I

6-20-63

ABSORBER 1.1 g Be + 0.3 g Cu
Sandwich

SOURCE DIST. 10 cm (c)

ENERGY SCALE 1 KeV/PHU (Cs)

7.1 hr. Se^{73}

3" x 3" - 21 Na I

4-2-63

ABSORBER 1.18 g/cm² Be + 0.1

Cu Sandwich

SOURCE DIST. 10 cm (c)

ENERGY SCALE 1 KeV/PHU (Cs)

58 hr Br ⁷⁷

3" x 3" - 2 Na

4-5-63

ABSORBER 1.18 g/cm² Be

SOURCE DIST. 10 cm (c)

ENERGY SCALE - 1 KeV/PHU (Cs)

40 day Ag^{105}

3" x 3" - 2 Na I

6-10-63

ABSORBER 1.18 g/cm² Be

SOURCE DIST. 10 cm (c)

ENERGY SCALE - 1 Kev/PHU (Cs)

5.8 day $\text{Sb}^{120\text{m}}$ $3'' \times 3'' - 2 \text{ Na I}$

4-8-63

ABSORBER $1.18 \text{ g/cm}^2 \text{ Be}$

SOURCE DIST. 10 cm (c)

ENERGY SCALE 1 KeV/PHU(Cs)

4.5 hr Pr^{139}

3" x 3" - 3 Na I

6-20-63

ABSORBER 1g/cm^2 Be + 200 mg. Cu
Sandwich

SOURCE DIST 10 cm (c)

ENERGY SCALE - 1 Kev/PHU (Cs)

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