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Final Report

STUDIES ON X-RAYS AND BREMSSTRAHLEN FROM SOURCE-TARGET MIXTURES

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L. E. Brownell
E. W. Coleman
C. J. Fox
J. V. Nehemias

RESEARCH AND DEVELOPMENT DIVISION
OFFICE OF THE SURGEON GENERAL, DEPARTMENT OF THE ARMY
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ABSTRACT

Homogeneous mixtures of beta-emitting radioisotopes with stable target materials have been investigated as possible radiation sources for medical radiography. Mixtures containing calcium-45, thallium-204, or promethium-147 as chemical compounds have been prepared and their spectra determined. Radiographic testing of these sources indicates that promethium-147, or other low-energy beta emitters, may find use as medical x-ray sources. However, the present sources do not have sufficient output intensity to produce radiographs in reasonable exposure times. Anticipated improvement factors can reduce exposure times to the order of minutes, but more efficient conversion of radiation to images by films and screens is necessary before practical application is expected.

A. INTRODUCTION

Portable x-ray units utilizing radioactive sources of radiation have been reported by the Army Medical Research Laboratory.[1-5] While such units present certain advantages over conventional field radiographic equipment, the quality of radiographs produced is not considered acceptable.

Radiographs were best described as "flat," with insufficient differentiation of bone and tissue. This difficulty was attributed to a higher-energy bremsstrahlung spectrum inherent in extended sources containing high-energy beta-emitting radioisotopes. This difficulty also occurs in heterogeneous source and target systems.[3-5] In addition, the efficiency of photon production, in terms of photons produced per beta particle incident upon the target, is usually less than 1%. An upper limit is imposed upon the number of incident beta particles obtainable, by specific activity and self-absorption considerations.

This project was established to investigate the feasibility of using mixtures of radioisotopes and stable target materials as radiation sources for diagnostic radiography. The spectral distribution and total production could be expected to be improved because: 1) the total activity that can be utilized is not limited by beta-particle range in the source; 2) the probability of first collision interaction with target atoms is increased; 3) backscatter losses from the target are omitted; and 4) the spectral distribution function of thickness observed in thulium can be utilized.[6,7] This report, together with the Preliminary Report and Literature Survey, completes presentation of the studies conducted under the contract. The initial report included a detailed discussion of the problems peculiar to diagnostic radiography, and the possible methods of using radioisotopes to solve them. A literature survey on the production of low-energy photons was presented, with an empirical evaluation of bremsstrahlen production in radiography sources.

B. SCOPE OF INVESTIGATION

The original proposal advocated investigation of systems of thulium-170 with aluminum and bismuth, strontium-90 with tungsten, and promethium-147 with barium, lead, or tungsten.

After conferring with the contract representative in December, 1956, the emphasis of the investigation was shifted somewhat. The major conclusions were: 1) in view of logistic problems expected in storing, transporting, and replacing sources of short half-life, such as thulium-170 (127 days), only those isotopes with a half-life of one year or longer should be investigated (a "use-life" of three to four half-lives is believed reasonable, based on operating experience with thulium-170); and 2) due to the uncertainties of evaluating an x-ray source on the basis of spectrum alone, the spectral studies should be supplemented by radiographic testing. The experimental work with thulium-170 was therefore discontinued, and the nature of the program redesigned to permit radiographic testing.

The practical application of isotopes of very long half-life is questionable, since long half-life and low specific activity are implicitly related, and low specific activity and small focal spot size are incompatible with reasonably short exposures. Maximum specific activity and half-life are related by the equation:

$$a = \frac{1.13 \times 10^{13}}{TA} ,$$

where

a = theoretical specific activity of pure isotope,
T = half-life, in seconds, of isotope, and
A = atomic mass of the isotope.

The calculated maximum specific activities of selected isotopes in curies/gm are tabulated below to illustrate this relationship. Even more important to radiographic application is the volume specific activity in curies/cc. Using theoretical metal densities, the maximum volume specific activity has been calculated.

TABLE I

CALCULATED MAXIMUM SPECIFIC ACTIVITIES FOR SELECTED ISOTOPES

Isotope	Half-Life[8]	Metal Density	Maximum Specific Activity	
			Curies/gm	Curies/cc
W-185	73.2 days	19.3 gm/cc	9,680	187,000
Tm-170	127 days	9.3	6,060	56,500
Ca-45	163 days	1.55	17,820	27,650
Pm-147	2.6 yr	6.9	938	6,480
Tl-204	4.0 yr	11.85	439	5,200
Sr-90	28 yr	2.6	142	370
Am-241	~460 yr	~20	~3.2	~64

It should be understood that the above values are specific activities of sources containing 100% of the radioactive isotope in question. In practice, the percent of the radioactive specie may be only a small fraction of the total weight or volume of the source.

Because of the low specific activity of strontium-90, the investigation of strontium source-target mixtures was withdrawn.

On the basis of these considerations, calcium-45, promethium-147, and thallium-204 were selected for study.

C. EXPERIMENTAL RESULTS

Source-target compounds have been investigated and a number of spectra have been determined. Radiographic testing has been undertaken with those sources producing sufficient radiation intensity.

The results are encouraging with regard to improved spectra, but the question of ultimate output efficiency remains unanswered. The studies included three isotopes, in quantities from ten microcuries to one curie.

The properties of the radioisotopes studied are presented in Table II.

TABLE II

PROPERTIES OF RADIOISOTOPES STUDIED

Isotope	Tl-204	Ca-45	Pm-147
Half-life	4.1 yr	163 days	2.6 yr
Beta energy (Mev)	0.765 (98%)	0.254	0.223
Gamma energy (Mev)	None	None	None
Other	EC (2%)	None	None

The source-target combinations were selected to include a number of elements having characteristic x-rays in the diagnostic region. The energies of these x-rays are tabulated in Table III.

TABLE III

CHARACTERISTIC X-RAY ENERGIES OF SELECTED ELEMENTS

Element	Atomic No.	Characteristic X-Ray Energies (Kev)			
		α_2	α_1	β_1	β_2
Iodine	53	28.37	28.66	32.35	33.08
Barium	56	31.87	32.25	36.43	37.91
Neodymium	60	36.89	37.42	42.34	43.38
Promethium	61	38.2*	38.8*	44.0*	45.0*
Tungsten	74	58.07	59.42	67.28	69.26
Thallium	81	72.58	73.00	82.57	85.23
Lead	82	72.90	75.05	84.86	87.75

*Estimated

1. SPECTRAL STUDIES

a) Thallium-204.—Sources containing 10 and 100 microcuries of Tl-204 as TlI showed a strong peak at about 85 kev. This energy would be expected from the electron capture decay of thallium to mercury. A minor peak was observed at about 33 kev. This peak was attributed to iodine-characteristic x-rays. The shape of the spectra indicated a hard bremsstrahlung component, but these low-level samples did not permit evaluation of the spectra above 120 kev. A subsequent source of 4 millicuries of thallium-204 iodide indicated that the bremsstrahlung spectrum extended well beyond 300 kev. This indicates that most of the problems encountered in thulium-170 sources are also inherent in thallium-204 sources. The only improvement would be an increased half-life.

b) Calcium-45.—Since calcium and promethium have similar chemical properties, much of the exploratory work for promethium-147 sources was carried out with calcium-45. This greatly reduced waste-disposal problems (shorter decay storage) and provided a source material with stable carrier. In general, the spectra of Ca-45 sources were very similar to comparable Pm-147 sources. The major differences were the absence of Pm-x-rays and a slightly higher-energy bremsstrahlung spectrum due to the higher beta energy of calcium-45.

c) Promethium-147.—The most extensive studies were with promethium-147. There are several problems involved in the preparation of Pm-147 source-target mixtures which have an effect on the source spectrum. First, the promethium solutions currently available have a variable concentration of stable and radioactive impurities. Since a millicurie of promethium is only about 1 microgram (about 5×10^{15} atoms), any effective tracer-level x-ray source must consist of promethium dispersed in a carrier. The major impurity present in tracer-level promethium solutions is neodymium. This is fission-decay neodymium

which is incompletely separated from the promethium during processing. The solutions generally contain less than 1 mg/mc of total solids, and less than 10 ppm of heavy metals, which may include plutonium, curium, and americium. The decay of these heavy-metal alpha emitters is accompanied by low-energy gamma rays, which may be apparent in multicurie sources.

Since no stable isotopes of promethium are known, and the quantity of Pm-147 in millicurie sources is too small for practical handling, another element must be used as carrier in low-level promethium studies. Neodymium was chosen as carrier since its chemical behavior is identical with promethium and it was already present in the promethium solution as an impurity. This practically eliminates the possibility of observing promethium x-rays, since photoelectric absorption of promethium characteristic x-rays in neodymium is very high.

The first Pm-147 sources were prepared by evaporation of promethium solutions on dry precipitates of $BaWO_4$ and $PbWO_4$. These sources each contained about 15 microcuries of Pm-147. Very little difference was observed in their spectra. The level of activity was too low to permit detailed spectral analysis.

A 4.5-mc Pm-147 (Nd carrier) source was prepared as precipitated tungstate. To compare the effect of target atomic number on the spectrum, a similar source was prepared as the molybdate. The experimental spectra of these sources are shown in Fig. 1.

The major difference in these spectra is in the characteristic x-ray region. The major peak of the tungstate source is due to tungsten x-rays, while the molybdate source has a peak corresponding to neodymium-promethium x-rays.

Beyond the characteristic x-ray region the spectra are practically identical. This is in agreement with theory, since the distribution of bremsstrahlung is independent of atomic number for atoms other than the parent, and internal bremsstrahlung has equal probability in the two sources. However, the magnitude of bremsstrahlung production is dependent upon atomic number (Z^2 law), and the curves have been normalized to compare the spectra rather than the relative efficiency.

d) Comparison of Spectra.—Figure 2 compares the spectra of three possible radiographic sources: thulium-170, thallium-204 iodide, and promethium-147 tungstate. Although all three sources show peaks in the characteristic x-ray region, the broad, flat, higher-energy bremsstrahlung spectra of the thulium-170 and thallium-204 sources indicate that shielding difficulties and poor radiographic contrast may be expected from these sources. The spectrum of the promethium sources is a definite improvement over thulium-170 with regard to higher-energy components. The major peak is at a higher energy than the thulium, but is still in the diagnostic region. (A lower-energy peak could be obtained by use of molybdate instead of tungstate, as shown in Fig. 1.)

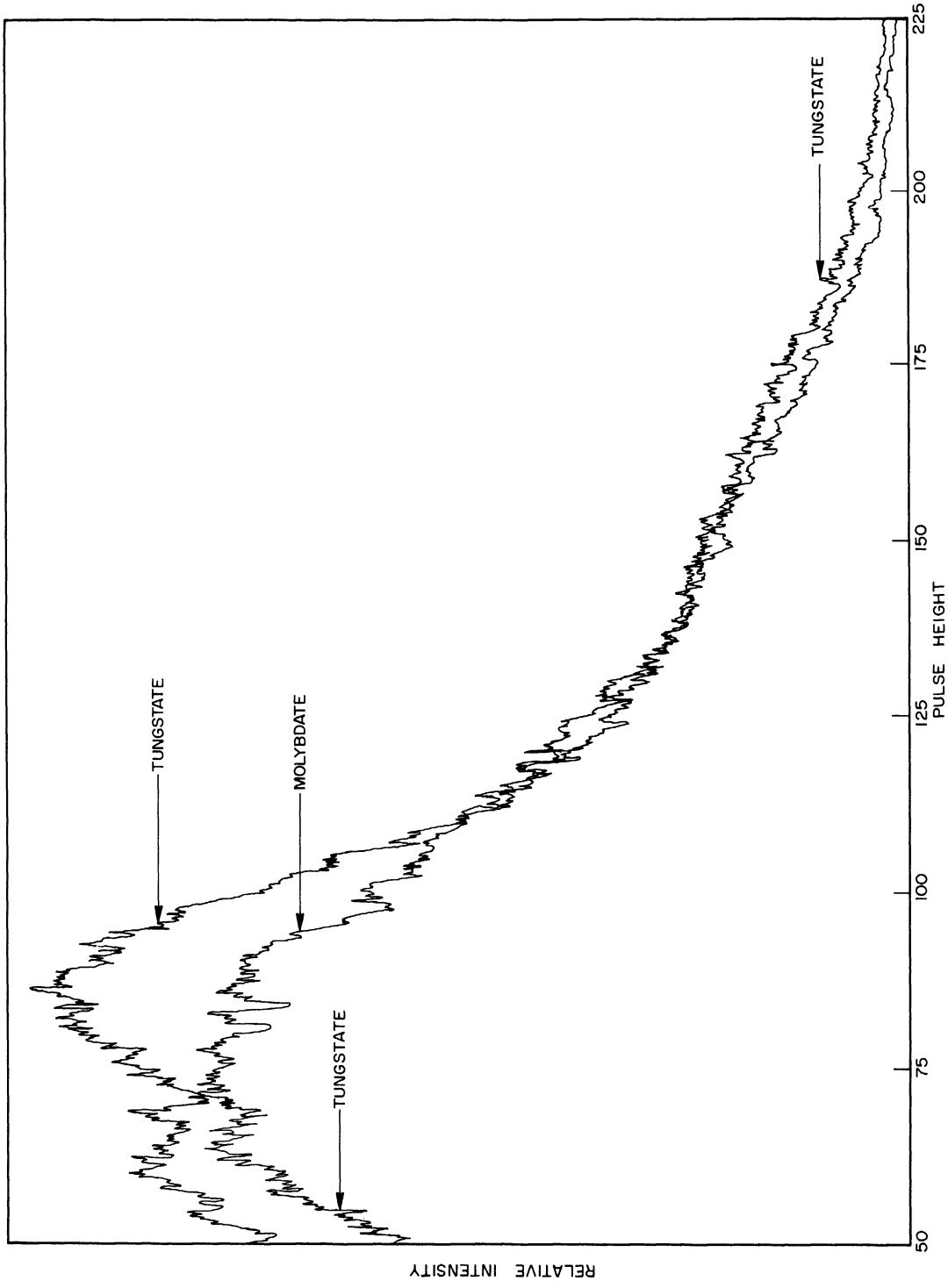


Fig. 1. Comparison of the spectra of promethium-147 (neodymium) tungstate and molybdate source-target mixtures.

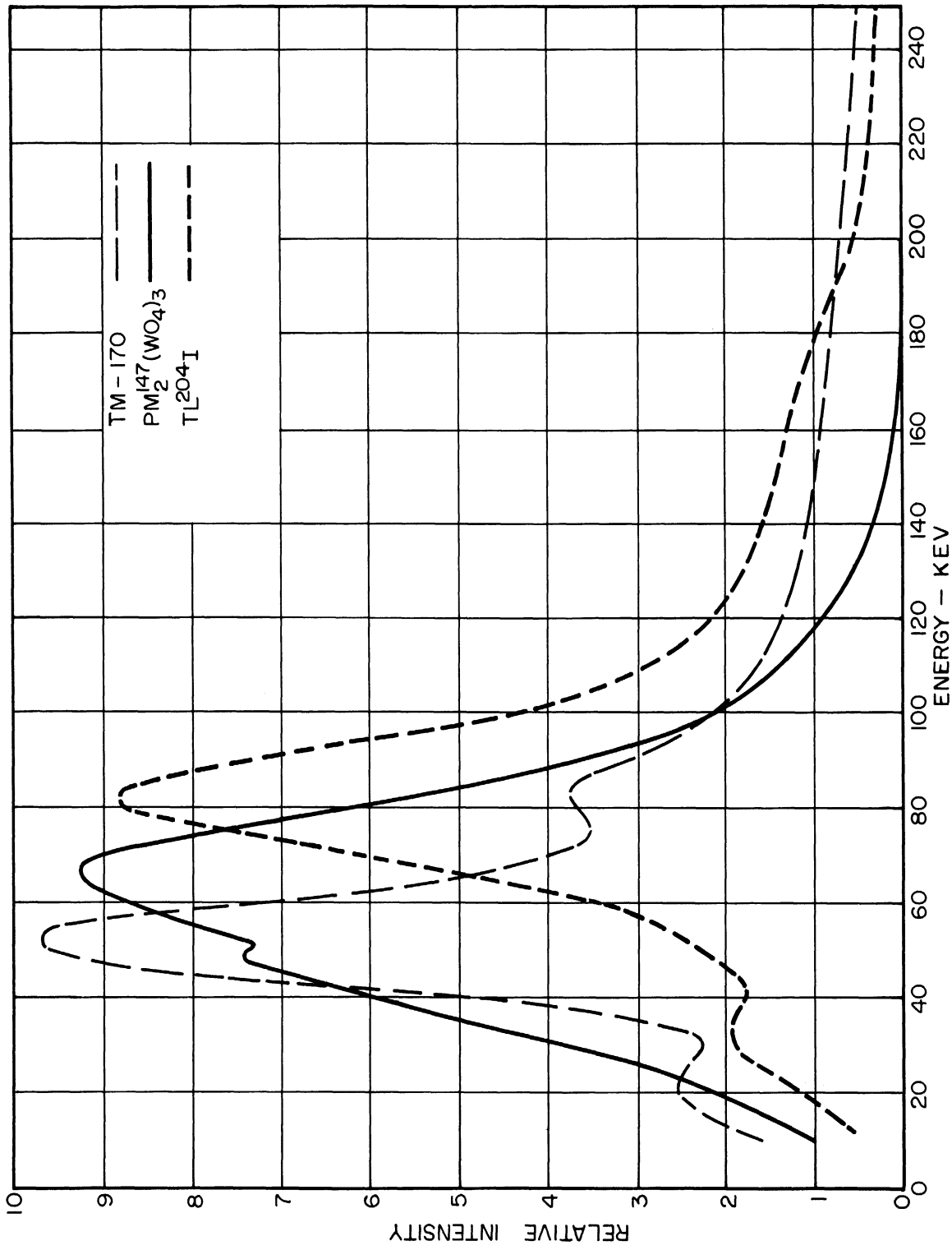


Fig. 2. Comparison of the spectra of three radiographic sources.

e. One Curie Pm-147 Tungstate Source.—A one-curie promethium-147 tungstate source with a $Nd_2(WO_4)_3$: $Pm_2(WO_4)_3$ ratio of 16.5 was prepared by the Oak Ridge National Laboratory. The source was encapsulated in a metal capsule with a 0.005-in. stainless-steel window. It is felt that future sources of this type should have a beryllium window to reduce absorption loss. The spectrum of the one-curie source is presented in Fig. 3. This figure also shows the calculated spectrum for the same source with a 0.020-in. beryllium window. A thinner beryllium window may be feasible, with a correspondingly higher output.

2. RADIOGRAPHIC STUDIES

The radiographs taken with the experimental sources are reproduced as positive images. These images are contact prints made from the original radiographs. This method allows sharper detail and better contrast comparison for films varying widely in degree of exposure.

The radiographs are displayed in sets for ease of comparison. Figure 4 shows three radiographs of an eviscerated, preserved mouse sealed in a Lusteroid plastic test tube.

Comparison of the radiographs of Fig. 4 is difficult because of the wide variation in focal spot size and source-film distance used. Even with these considerations in mind, the improvement achieved with Pm-147 over Tl-204 is evident. The higher degree of contrast is even more pronounced in the original radiographs. Although the high degree of resolution obtained in Fig. 4c is reduced in the Pm-147 radiograph, the contrast is comparable. The definition of the rib cage is adequate for gross examination, and the definition of vertabrae is fair. The sutures used in the abdomen wall are also distinguishable.

Due to the long exposure times required, no radiographs of human subjects were feasible. To evaluate performance on human subjects, a phantom was constructed of human bone imbedded in paraffin. A section of sterile lyophilized human pelvis was obtained from The University of Michigan Hospital and embedded in a paraffin block one inch thick. Wood screws of various lengths were embedded in one edge of the block in a step pattern, with the shortest screw nearest the bottom and the longest screw nearest the top of the block. Fine steel wires were embedded in the lower surface of the block, and the phantom was wrapped in a protective film of mylar plastic.

Figure 5 presents a Pm-147 radiograph of the phantom with a machine x-ray for comparison. The results are favorable, and the structural detail of the bone marrow is almost as clear in the Pm-147 radiograph. (The difference in definition is partially attributed to the smaller focal spot size and larger source-film distance of the machine radiograph.) Numerous defects in the phantom, such as void volumes and cracks, are clearly recognizable in either radiograph. The mylar film is also evident at the end where the radiation was received tangentially.

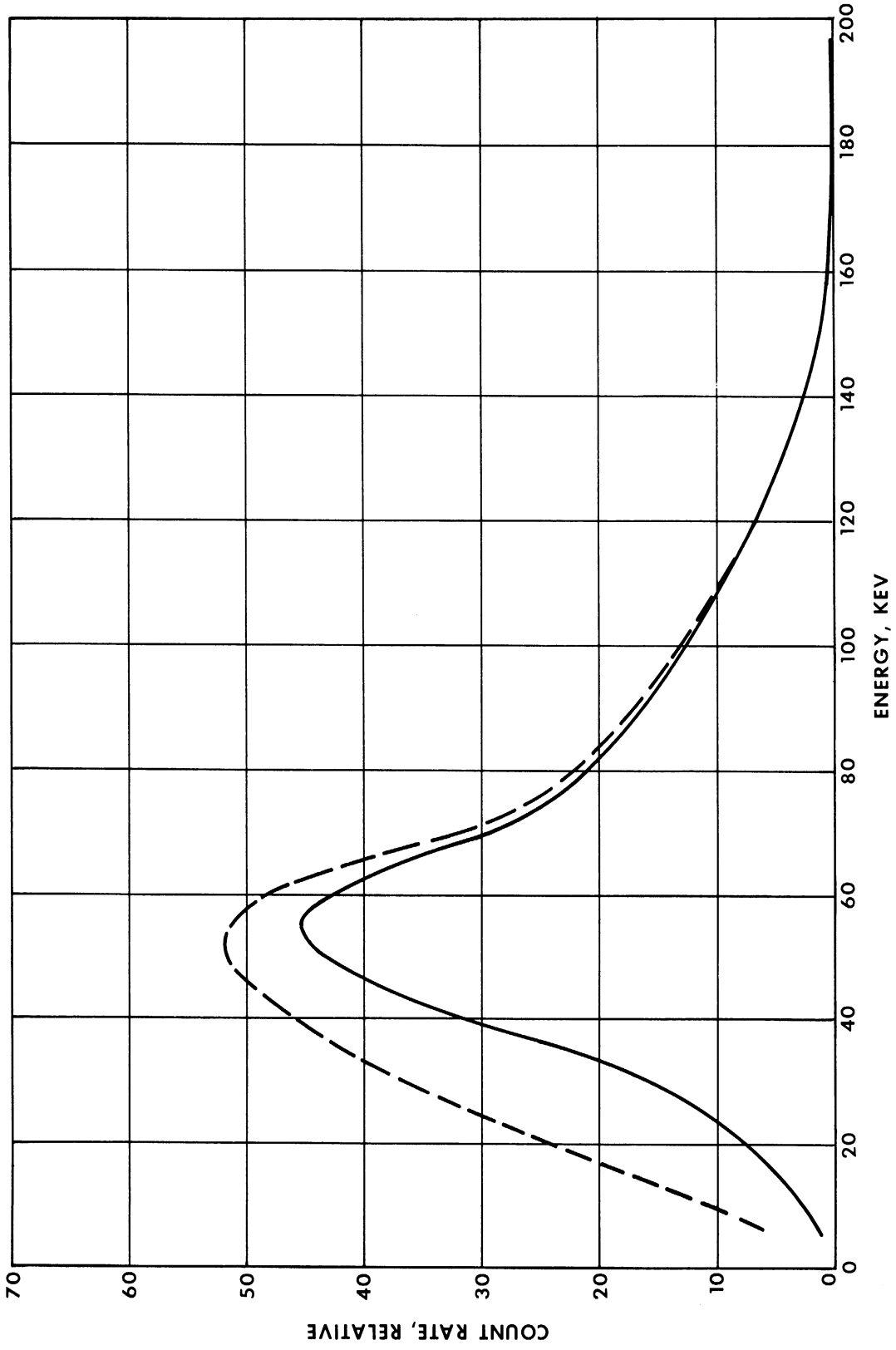
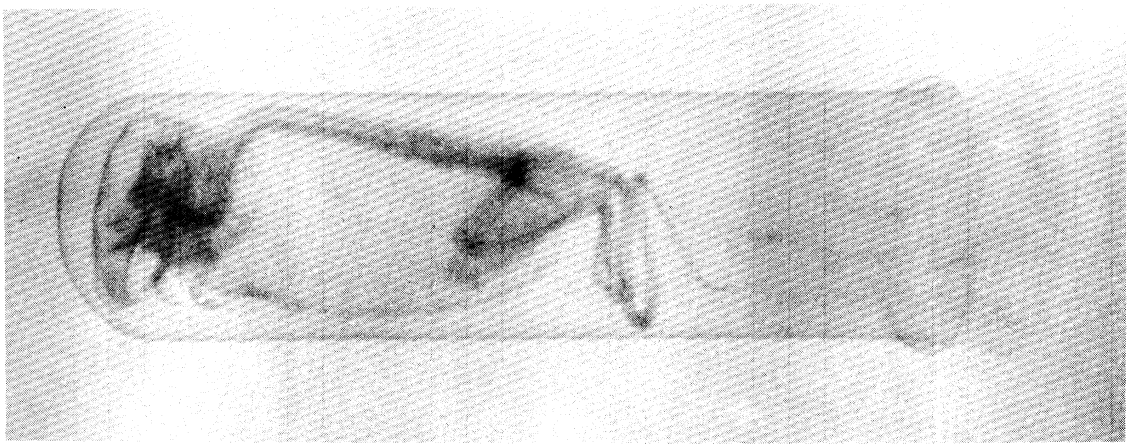
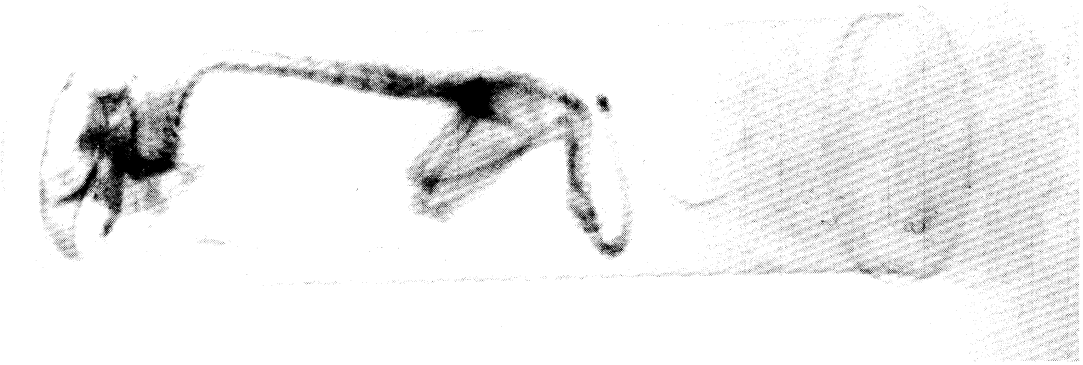


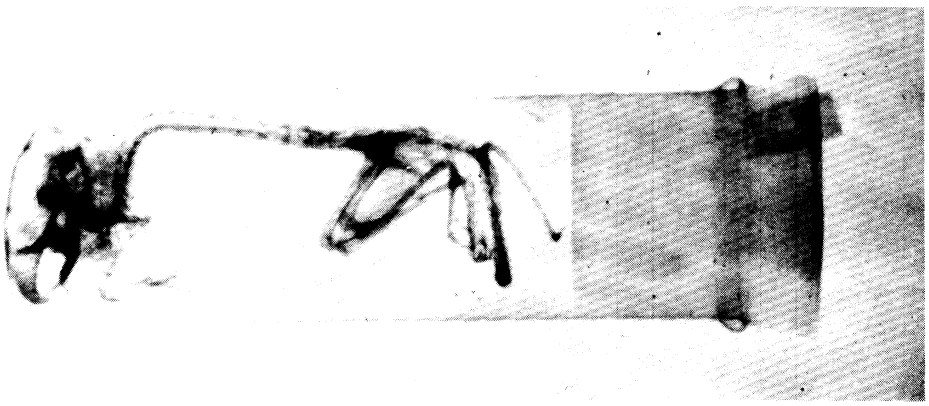
Fig. 3. Spectrum of the 1-curie Pm-147 tungstate source (solid curve) and calculated spectrum of the same source with a 0.020-in. beryllium window.



a. Tl-204 iodide, 4-mc source, 2-mm diam. Exposure: 240 hr at 5 in.

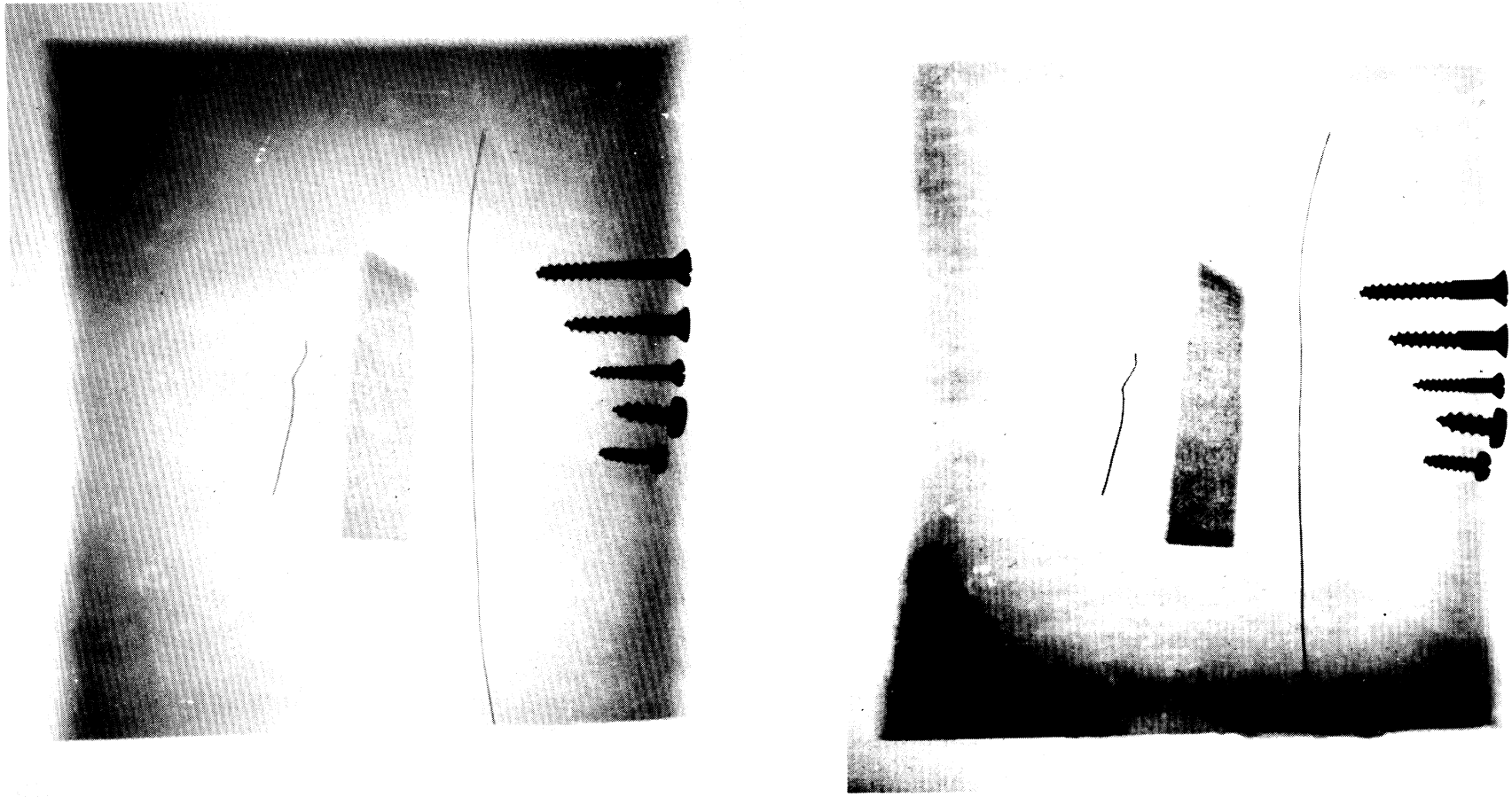


b. Pm-147 tungstate, 1-curie source, 4-mm diam. Exposure: 15 hr at 8 in.



c. Machine x-ray: 67 kvp, 1.25 mas at 30 in.

Fig. 4. Radiographs of preserved mouse.



a. Pm-147 tungstate source: 1-curie source, 4-mm diam. Exposure: 68 hr at 19 in.

b. Machine x-ray: 67 kvp, 1.75 mas at 30 in.

Fig. 5. Radiographs of paraffin phantom.

Figure 6 is a radiograph of a number of objects presenting a broad variety of materials and densities. Unfortunately the reproduction does not show the fine detail of the original radiograph. The detail of the flexible watch strap is excellent. The mica supporting discs in the electron tube are clearly shown, and the inner construction of the resistor is well detailed. The radiograph of the scissors faintly shows the screw hinge, and has a good density variation with metal thickness. Perhaps the most striking result is shown in the radiograph of the stopwatch, since similar radiographs with Tm-170 have been published.[1] The Pm-147 radiograph exhibits only partial details of the internal mechanism, in contrast to the fine detail produced by higher energy Tm-170 sources. This provides radiographic evidence supporting the spectral determination of lower bremsstrahlung energy in Pm-147 sources.

3. ABSORPTION STUDIES

Absorption studies of the promethium tungstate sources supplemented the spectral evaluation. Figure 7a is the aluminum absorption curve obtained from analysis of the 4.5-mc Pm-147 tungstate source. The absorption curve, indicated by the data points, could be resolved into three absorption energy groups, all below 100 kev. The mass absorption coefficients for these energy groups are indicated on the graph.

The 4.5-mc source was too weak to permit accurate lead absorption measurements.

Figure 7b is the stainless-steel absorption curve of the 1-curie Pm-147 tungstate source.

4. SUMMARY OF EXPERIMENTAL RESULTS

Source-target mixtures containing calcium-45, thallium-204, or promethium-147 have been prepared and evaluated. Spectral studies and radiographic testing show that source-target mixtures containing low-energy beta emitters, such as Pm-147, produce radiation applicable to diagnostic radiography without the objectionable higher-energy "fogging" components of thulium-170 or thallium-204 sources. The low-energy bremsstrahlung spectrum of promethium sources, reinforced by characteristic x-radiation, has a distribution very similar to that of a tungsten anode x-ray tube.

Although exposure times required are extremely long at this time, several improvements can be made to increase the source output. However, present technology does not indicate that application of promethium sources for medical radiography is immediately practical. This will be discussed in detail after presentation of the experimental data in the next section.

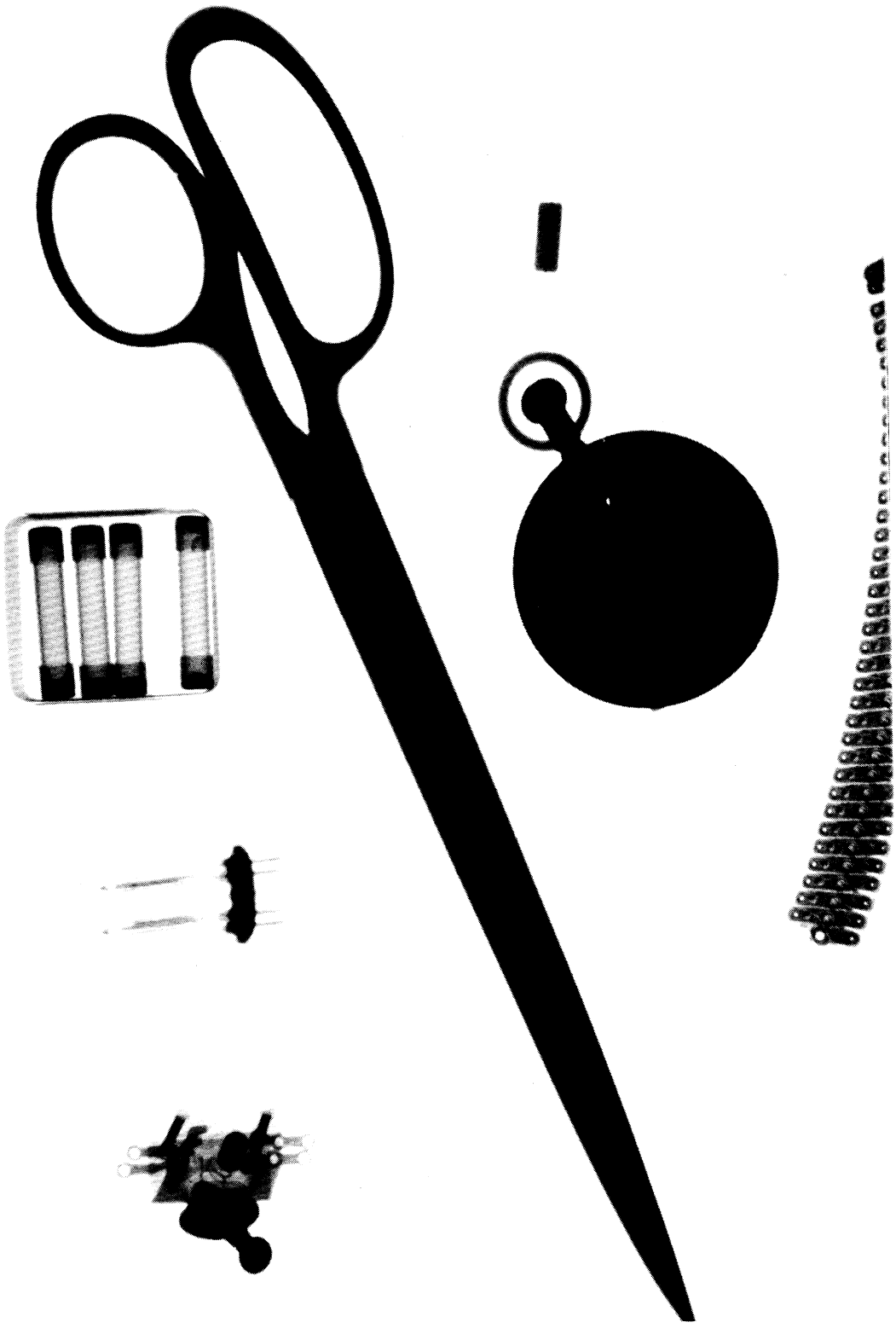


Fig. 6. Technical radiograph with promethium-147 tungstate source; 1-curie Pm, 44 hr at 10 in.

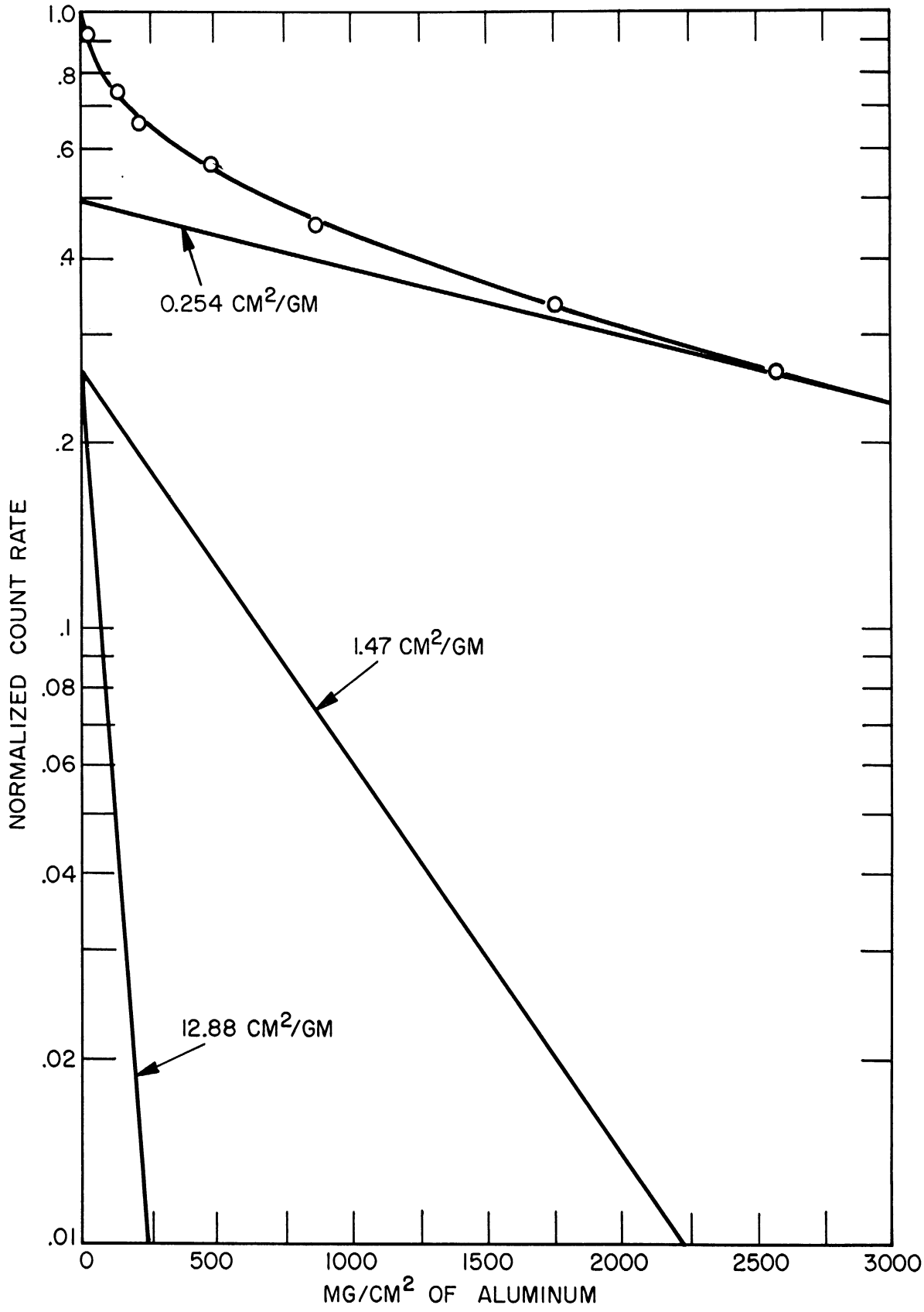


Fig. 7a. Aluminum absorption curve and aluminum absorption analysis for 4.5-mc Pm-147 tungstate source.

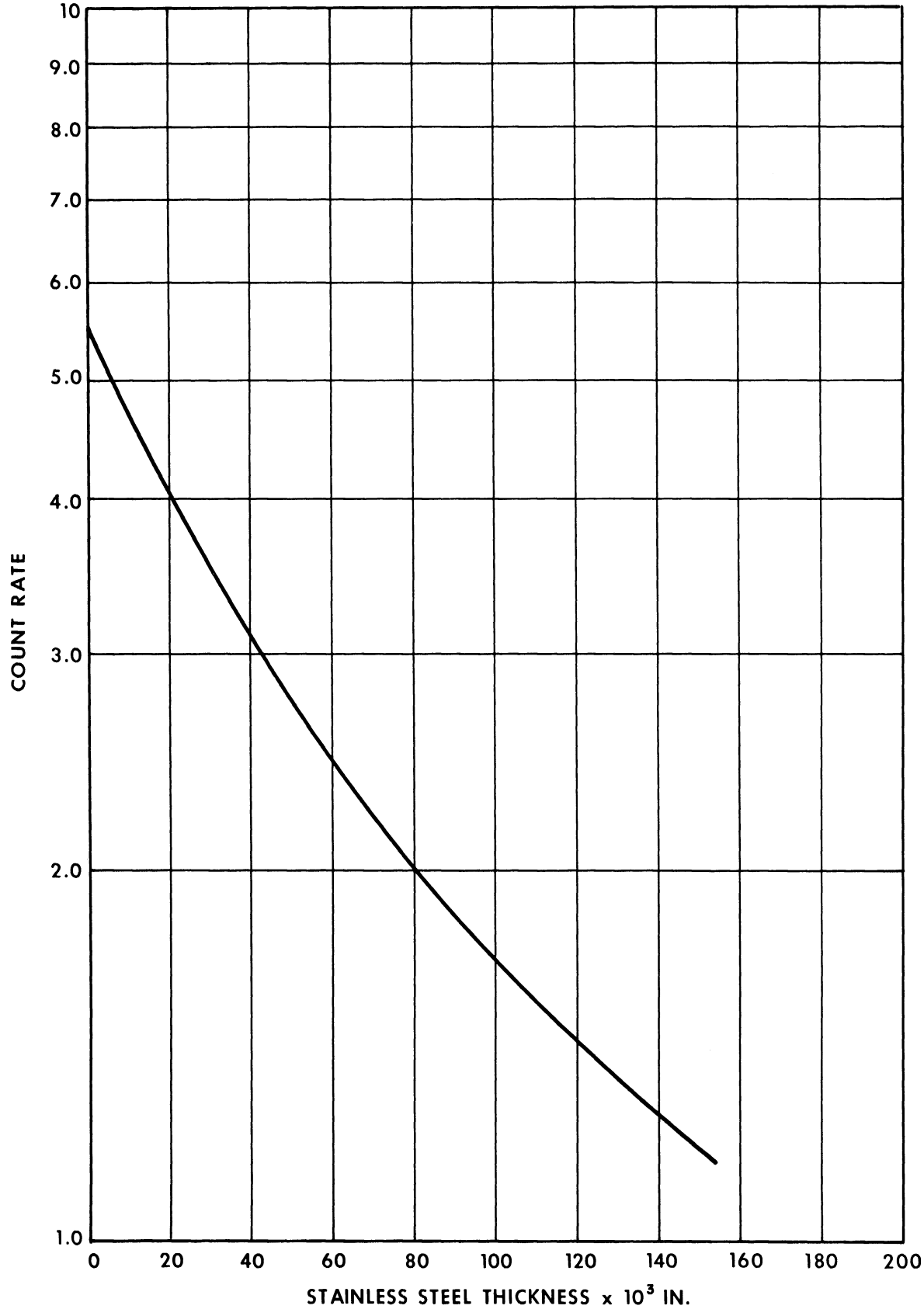


Fig. 7b. Stainless-steel absorption curve for 1-curie Pm-147 tungstate source.

D. EXPERIMENTAL METHODS

The experimental results reported in the preceding section have been obtained through the cooperation of many departments of The University of Michigan. The authors wish to express sincere appreciation to Professor F. J. Hodges, Chairman of the Department of Radiology, School of Medicine, for the use of x-ray cassettes and darkroom facilities. The assistance of Mr. A. H. Emmons of the Phoenix Memorial Laboratory in arranging use of the gamma spectrometer greatly facilitated this study.

1. SOURCE CAPSULES

All the sources used in this study were encapsulated, sealed sources. This provided a convenient means of containing the source in a fixed geometry with minimum danger of contamination. The fabrication and performance of various source capsules is discussed.

a) Lucite Capsules.—Figure 8 shows the Lucite capsule and the components from which it was constructed. The sleeve at the left is 1/2-in. diameter by 1/4 in. high, with a 2-mm axial cavity. The thicker disc at the right serves as the "window" of the assembled capsule, providing 3/16 in. of beta absorber. To charge the capsule, the window section is welded in place using chloroform as solvent. The radioactive source material is then pressed into the cavity. In practice, the quantity of source material is insufficient to fill the cavity. To assure a fixed source geometry within the capsule, a paraffin follower plug is inserted and pressed tight. The paraffin plug also wipes the sides of the cavity clean of radioactive material and serves as a primary seal. The excess paraffin is carefully removed with a scalpel and the thin Lucite disc welded in place with chloroform. These capsules were used for the thallium-iodide sources and some of the preliminary promethium and calcium sources. None of these source capsules has shown surface radio-contamination, indicating no leakage. The major disadvantage of these capsules is their small volume and the difficulties encountered in machining small-diameter holes in Lucite rod without causing striation and cracking.

b) Cast Plastic Capsules.—To overcome the disadvantages experienced with the Lucite capsules, a set of molds was prepared to construct capsules using a cold-casting resin.

Figure 9 shows the capsule components and a sealed capsule. The capsule base has a 3/4-in. diameter, with a 1/4-in.-diameter cavity 3/4 in. deep. The cavity and plug have a slight taper providing a seated fit when closed. The capsule base was machined after casting to provide the proper window thickness for the beta energy of source material to be encapsulated. These capsules were

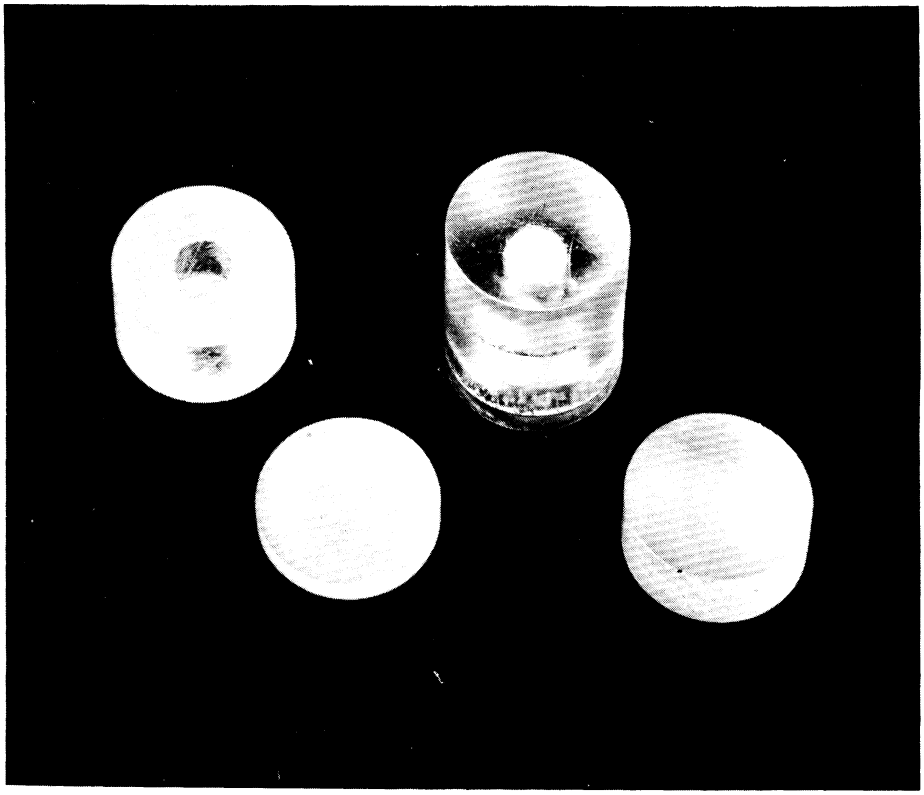


Fig. 8. Lucite capsule and components.

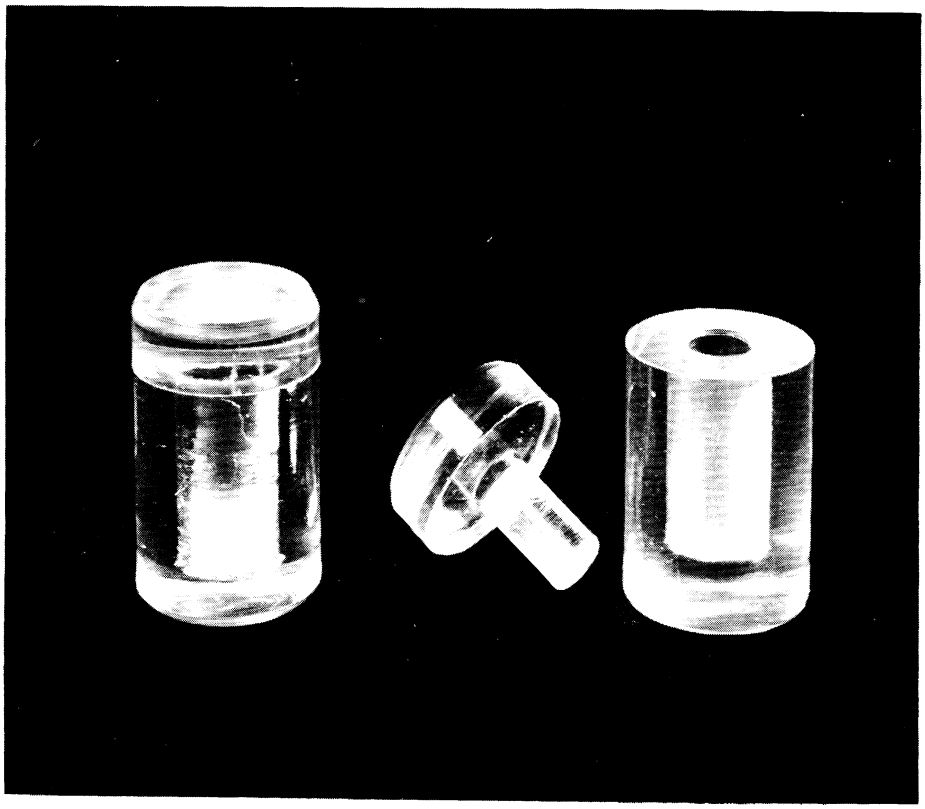


Fig. 9. Cast plastic capsule and components.

loaded in the same manner as the Lucite capsules, except that a sized paraffin follower plug was used. The capsule plug was sealed on using the cold-casting resin as cement. This was found to provide a seal which could not be broken by repeatedly throwing the capsule on a concrete floor. When the assembled capsule was clamped in a vise and subjected to torsion, the capsule body failed, rather than the seal. These capsule were deemed highly satisfactory, and were used in the majority of the calcium and promethium studies.

c) Stainless-Steel Capsule.—Although the plastic capsules were considered satisfactory for low-level sources, they might be expected to deteriorate under the intense beta-particle flux of curie-level sources. Figure 10 is a sketch of the capsule used for the 1-curie promethium tungstate source. This capsule is a modification of a standard Oak Ridge capsule, and was constructed and loaded by personnel of the Oak Ridge National Laboratory. The modification consisted of pressfitting a 4-mm-ID aluminum sleeve into the source cavity and machining the window thickness to 0.005 in. Aluminum was chosen for the sleeve to minimize bremsstrahlung and characteristic x-ray production in the vertical

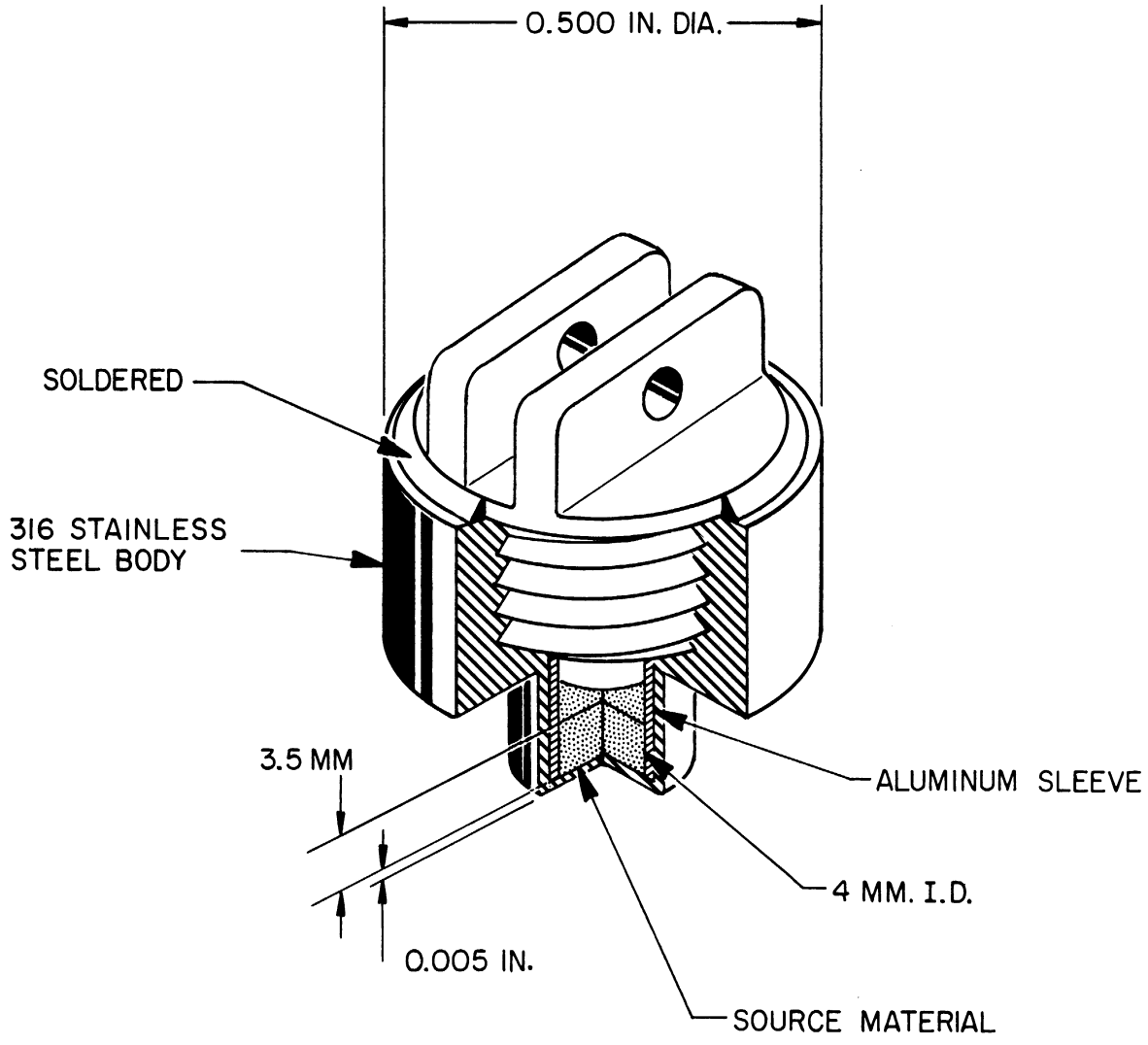


Fig. 10. ORNL stainless-steel capsule modified for 1-curie promethium-147 source.

walls of the source cavity, so that the effective focal spot size would be 4 mm. The capsule was silver-soldered after filling and placed in a larger Lucite capsule with an air-path aperture to the window. The plastic capsule was used only to eliminate the possibility of accidental contact with the thin window during handling. The question of mechanical strength in a 0.005-in. stainless-steel window, and the relatively high absorption factor for low-energy radiation ($e^{-ux} \simeq 0.25$ at 25 kev) suggests that a thicker beryllium metal window would be better. The capsule has shown no surface radio-contamination in periodic smear tests.

2. SOURCE PREPARATION

The source-target mixtures prepared in this study were of two types: 1) mechanical mixtures, and 2) chemical compounds. All sources were prepared in a glove box. "Cold" runs using stable isotopes were used to determine appropriate methods before actual source preparation.

a) Thallium-204.—The thallium-204 was obtained from Oak Ridge Batch No. 5 shipped November 9, 1956. The solution contained 3.58 (± 10%) mc/ml of thallium-204 as thallium nitrate in 2.72 N nitric acid. The specific activity was 123 mc/gm of thallium, with a dilution of 29 mg thallium/ml. The source-target mixtures of thallium-204 were chemical compounds of thallium iodide. A carrier solution of stable thallium nitrate (Fisher Scientific No. T-85, Control No. 529247) was prepared to dilute the activity before preparation of sources. The procedure for preparation of thallium-iodide sources is as follows: In a centrifuge tube place 1 mc of Tl-204 solution and 1 ml (100 mg) of thallium nitrate solution. Dropwise, add 47% hydroidic acid to 3 drops excess. Centrifuge the thallium iodide precipitate and decant the supernatant liquid. The precipitate is washed twice with distilled water by stirring, centrifuging, and decanting. The damp precipitate is transferred to a watch glass and dried at 60°C. This quantity of dry precipitate is divided into two sources containing one-third and two-thirds of the total precipitate.

A 4.5-mc source was prepared for radiographic testing of thallium-204 iodide source-target mixtures using the above method without addition of stable carrier.

b) Calcium-45.—Calcium-45 has a beta-decay energy of 254 kev and a half-life of 163 days. Since the beta energy is about the same as promethium-147 (223 kev), it was convenient to use calcium-45 in part of the experiments. The advantages were: 1) a stable calcium carrier could be used, 2) the shorter half-life simplified waste storage and handling, 3) the absence of measureable calcium x-rays in the spectra allowed better evaluation of various target materials, and 4) the chemistry of calcium is very similar to that of promethium.

Calcium-45 was used in conjunction with various targets (salts) to provide sources for calibration of the NaI(Tl) crystal used in spectral studies.

The activity was obtained from Oak Ridge Batch No. 115 shipped January 3, 1957. The solution contained 4.03 (± 10%) mc/ml.

The preliminary study on target materials included preparation of sources containing Ca-45 evaporated on precipitates of $BaWO_4$ and $PbWO_4$. The procedure was as follows: Into a cast plastic capsule containing 100 mg of dry 120-mesh precipitate, 0.02 ml of Ca-45 solution was pipetted and the mixture allowed to evaporate at room temperature (about 24 hr). The capsule was then sealed in the manner described previously in Section D.

c) Promethium-147.—The first Pm-147 source was prepared by evaporation on $BaWO_4$, as described in the preceding section. The spectrum of this source showed tungstate to be a suitable target, and a promethium-neodymium tungstate source was prepared. The neodymium carrier solution was prepared as follows: 25 gm of purified neodymium chloride (Fisher Scientific Company No. N-33, Lot No. 543281) was dissolved in 0.13N HCl. Since the salt was in the hydrated form, the solution was standardized after preparation. The method of standardization included precipitation of oxalate and ignition to oxide as weighing form. The solution was found to contain 45.5 mg/ml of Nd.

The cold runs with neodymium carrier indicated a slight solubility of the tungstate in water. To lower the solubility, an ethanol-water solution was used. The neodymium tungstate prepared in the cold runs was found to be a pale pink material easily ground to a fine powder. The precipitate fluoresces soft blue in ultra-violet or white fluorescent light. The material was also used to determine the bulk density of < 120-mesh precipitate when compacted in a cast plastic capsule. A bulk density of 0.8 gm/cm^3 was determined, and 100 mg was chosen as the loading for these capsules. The promethium-147 was obtained from Oak Ridge Batch No. 18 shipped November 8, 1956. The solution contained 1.55 (± 20%) mc/ml of Pm-147 in 0.13N HCl. The total solids were less than 1 mg/ml with less than 10 ppm of heavy metals. The procedure used in preparing neodymium tungstate-promethium-147 tungstate sources is given in the following paragraph.

To a 50-ml beaker containing 1.00 ml of neodymium carrier solution, 3.00 ml of promethium solution and 10 ml of distilled water were added. The solution was neutralized with NaOH until a cloudy precipitate began to form. Dropwise addition of 0.13N HCl with stirring was then used to redissolve the precipitate. To the clear solution, 5 ml of absolute ethanol were added. The tungstates were then precipitated by dropwise addition of 1 ml of 0.53N Na_2WO_4 solution. After settling for 15 minutes, the clear supernatant liquid was removed by pipette and the precipitate washed by stirring in 5 ml of 1:1 ethanol-water solution. After settling, the supernate was again removed and a final wash with pure ethanol was performed. As much liquid as possible was removed by decantation, then the precipitate was dried in the beaker on a 50°C hotplate. The precipitate cake was broken up, then dried overnight at 100°C. The dry precipitate was crushed with a glass rod and transferred to a cast plastic capsule with a 2-mm window. The combined filtrates of all operations were evaporated

and the resulting salt mixture was dried and sealed in a similar capsule. The count rate from the filtrate salts was about 12.4% of the count rate from the precipitate.

After evaluation of the millicurie promethium-147 tungstate source, a 1-curie promethium tungstate source was ordered from the Oak Ridge National Laboratory. The preparation of the tungstate and the encapsulation were performed by Mr. R. S Pressley of the ORNL staff. The details of the procedure are as follows. A solution containing 1387.5 mc of Pm-147 with an undetermined quantity of impurities was used to prepare a tungstate precipitate. All operations (precipitation, washing, drying, etc.) were carried out in a tared centrifuge tube. The weight of dry tungstate found by difference was 91.3 mg. As much of this dry material as possible was placed in a specially constructed stainless-steel capsule and sealed. The remaining tungstate adhering to the centrifuge tube walls was dissolved and combined with the filtrates and washes collected during precipitation. This combined solution was assayed and contained 362.5 mc of Pm-147. Therefore, the activity in the capsule was 1025 mc by difference.

It is difficult to determine the exact Nd:Pm ratio in the source, since the Nd resulted from fission product decay and its chemical atomic weight depends upon the reactor fuel history, which is unknown. A good approximation can be made using the atomic weight of natural neodymium. The weight ratio of the tungstates is perhaps a better measure, since the small differences in atomic weight have lower significance in the molecular weight of the tungstate.

As shown previously in Table I, pure promethium-147 has a specific activity of 938 curies/gm, or a curie weight of 1/938 = 0.00107 gm Pm/curie. Since $Pm_2(WO_4)_3$ has a molecular weight of 1037.8, and contains 2 Pm atoms per molecule, the weight of $Pm_2(WO_4)_3$ containing one curie of Pm is

$$\left(0.00107 \frac{\text{gm Pm}}{\text{curie}}\right)\left[\frac{1037.8}{2 \times 147} \frac{\text{gm } Pm_2(WO_4)_3}{\text{gm Pm}}\right] = 0.00376 \frac{\text{gm } Pm_2(WO_4)_3}{\text{curie}} .$$

The original source solution containing 1.3875 curies yielded 91.3 mg of mixed tungstates. The theoretical weight of the promethium tungstate is then

$$(1.3875 \text{ curies})\left[0.00376 \frac{\text{gm}}{\text{curie}} Pm_2(WO_4)_3\right] \cong 0.0052 \text{ gm, or } 5.2 \text{ mg.}$$

Then by difference, the weight of neodymium tungstate is 91.3 mg - 5.2 mg = 86.1 mg. The weight ratio of $Nd_2(WO_4)_3$ to $Pm_2(WO_4)_3$ is therefore 86.1/5.2 = 16.5. This implies that the same weight of material (91.3 mg) could contain (16.5)(1.3875) $\cong$ 23 curies of Pm-147, if no neodymium were present.

3. SPECTRAL METHODS

The spectra reported in this study were determined with an RCL Recording Spectrogammeometer* and a special probe containing a 1-1/2-in. diameter by 1/2-in.-thick NaI(Tl) crystal coupled by a lightpipe to a standard RCL Mark 10 Model 25 probe assembly.

The instrument was calibrated using iodine-131 and characteristic x-rays. Figure 11 is a plot of calibration curve showing the iodine peaks within the energy region studied, and Pb x-ray peaks. Other characteristic x-rays used, but not shown in the plot, were barium, tungsten, and neodymium. The curve was obtained with a gain setting of 64, high voltage of 1000 v, and a slit width of 10 v. All spectra were determined using these settings.

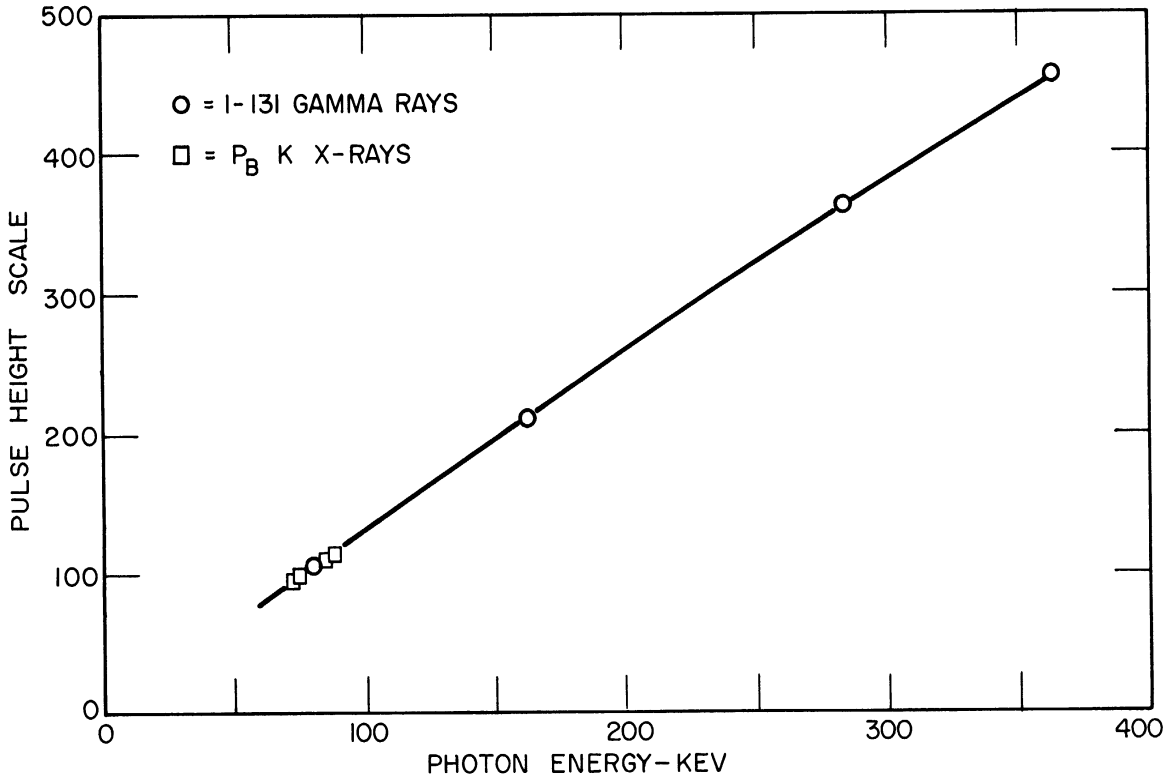


Fig. 11. Calibration curve for 1-1/2-in. x 1/2-in. NaI(Tl) crystal.

4. RADIOGRAPHIC METHODS

As much as possible, the radiographs were prepared using standard medical techniques. Kodak Blue-Brand X-Ray Film (Lot No. A5121-1668X27) was used with

*This instrument was used by arrangement with the Michigan Memorial-Phoenix Project.

Patterson Par Speed intensifying screens. Development was 6 min at 68°F in Kodak Medical X-Ray Developer. The usual stop-bath, hypo, washing, and drying procedures were employed. Due to the long exposure times required for the radiographs, special precautions were taken to minimize exposure to stray radiation. The film was stored and processed in a separate building, and cassettes were loaded immediately before exposure.

E. SUMMARY AND CONCLUSIONS

To be ideally suited as a portable radiation source for medical radiography, a radioactive isotope must have the following characteristics:

1) The spectral distribution of radiation from the source must lie in the diagnostic region, with no high-energy components which reduce radiographic contrast and increase shielding mass.

2) The radioisotope must be available at specific activities high enough to provide reasonable exposure factors for sources of small focal spot size.

3) The half-life of the radioisotope must be adequate to permit reasonable shelf-life and use-life, but not so long as to preclude high specific activity.

4) The radioisotope must be available in quantity, at reasonable cost, to permit wide-spread use in the medical profession.

None of the known radioisotopes completely meets these requirements. Although thulium-170 has a relatively short half-life (127 days) and yields flat radiographs because of higher-energy bremsstrahlung, it has been used to advantage in certain surgical procedures not requiring a high degree of radiographic definition.

Because portable radiation sources for medical radiography can extend the use of radiographic examination and diagnosis to areas of medical practice where it has been impractical, many attempts have been made to produce suitable sources. Aside from the question of reasonable half-life, all have been unsuccessful either because radiation intensity was inadequate to yield reasonable exposure factors, or because the spectrum of radiation contains undesirable higher-energy components.

The source-target mixture was chosen for study in hopes that these problems could be resolved. Although the results are discouraging from the standpoint of radiation intensity, the spectra of promethium source-target mixtures promise radiographs of higher quality than those thus far attainable with thulium.

There are several factors relative to the efficiency of promethium source-target mixtures that promise improved sources in the future. In 1958, the Oak Ridge National Laboratory expects to begin separating promethium-147 in quantity. With larger quantities available for purification, a sharper cut can be made from neodymium, and essentially pure Pm-147 is anticipated at a dollar a curie or less. This means an increase in specific activity of seventeenfold over the one-curie source studied. Assuming a beryllium-window capsule will provide a further twofold increase in source intensity, radiographs such as shown in Fig. 4b could be produced in about 30 minutes. For medical use, this exposure would be impractical, and an additional improvement factor of at least 60 will be required.

We have made no attempt in this study to maximize efficiency of the sources. Our primary objective has been to demonstrate that sources with suitable spectral distribution can be produced. However, the problem of efficiency has been considered and certain possible improvements are evident.

1) The promethium tungstate source-target mixture contains only about 28% promethium. When massive promethium metal can be prepared, sources with specific activities about four times higher will be possible. When the metallurgy of promethium can be studied, promethium-rich intermetallic compounds with targets such as bismuth may be discovered. These ordered structures can be expected to have higher bremsstrahlung efficiencies.

2) No attempt has been made to maximize image-production efficiency. The screen-film combination employed is far from the most efficient currently available. With the impetus recently arising in the various reports of biological effects of radiation, and the resulting public awareness of the desirability of holding radiation exposure to a minimum, it is reasonable to expect that commercial interests will provide new image-production systems which require an even lower radiation exposure. There has been no such impetus previously, and image production has been geared to electrical x-ray equipment capable of delivering high intensities.

It is thus concluded that medical radiographic equipment utilizing radioactive sources can become practical only if more efficient means of radiation production can be used with image-production systems of greater sensitivity.

REFERENCES

1. Carpenter, A. W., Heer, R. R., Jr., Krohn, L. H., Coleman, E. W., and Boyer, F. J., "Preliminary Report on a Complete Portable Field X-Ray Unit," AMRL Report No. 168, Ft. Knox, Kentucky, 1954.

2. Coleman, E. W., "A Portable Thulium-170 X-Ray Unit," Proc. of Health Physics Soc., June, 1956.

3. Kereiakes, J. G., and Krebs, A. T., "X-Radiography with Beta-Emitting Isotopes," AMRL Report No. 173, Ft. Knox, Kentucky, 1954.

4. Kereiakes, J. G., and Krebs, A. T., "Technical Radiography with Beta-Emitting Isotopes," AMRL Report No. 180, Ft. Knox, Kentucky, 1955.

5. Kereiakes, J. G., and Kraft, G. R., "Thallium-204 Beta-Excited X-Radiography," AMRL Report No. 251, Ft. Knox, Kentucky, 1956.

6. Liden, K., and Starfelt, N., "Gamma-Rays, X-Rays, and Bremsstrahlen from Tm-170 Sources," Arkiv for Fysik, 7 (9), 109, 1953.

7. Coleman, E. W., Brownell, L. E., and Purohit, S. N., Appendix I of "Studies on X-Rays and Bremsstrahlen from Source-Target Mixtures," Prelim. Report and Literature Survey, Ann Arbor, Univ. of Mich. Eng. Res. Inst. Report 2471-1-P, Oct., 1956.

8. Oak Ridge National Laboratory, "Catalog and Price List, Radioisotopes, Special Materials and Services," (revised) Jan., 1957.