

"The Disintegration of Lutecium of Mass 177".

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# TABLE OF CONTENTS

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FOREWORD AND ACKNOWLEDGEMENTS	-1-
SUMMARY	- <b>ii</b>
INTRODUCTION	1
1. Previous work	1
2. Beta Decay	2
3. Fermi Theory	3
4. Kurie Plot	5
5. Methods of Measurement	5
a) Beta Radiation	5
b) Gamma Radiation	8
c) Decay Scheme	8
d) Detectors	9
APPARATUS	11
1. Spectrometer	11
2. Current Regulator	11
3. Detectors	13
4. Alignment	15
5. Calibration	15
MEASUREMENTS ON Lu <sup>177</sup>	16
1. Source	16
2. Observations with Nylon Backing	17
3. Sources Mounted on Aluminum	18
4. Location and Origin of Conversion Lines	19
5. Kurie Plot	20
6. Gamma Ray Measurements	22
7. Half Life	25

# TABLE OF CONTENTS, cont'd.

	Page
DISCUSSION	26
SUMMARY AND CONCLUSIONS	32
BIBLIOGRAPHY	34

#### FOREWORD AND ACKNOWLEDGEMENTS

McGill's beta ray spectrometer, designed and built at the Chalk River Laboratories of the National Research Council of Canada, was installed here chiefly through the efforts of Dr. J.L. Wolfson assisted by Dr. L.G.S. Newsham. The writer had the opportunity of assisting Dr. E. Brannen in observing the spectrum of Hf<sup>181</sup>. During this time the desirability of improving the current regulation became apparent. The modifications effected by the author are described in the text. This does not imply any criticism of previous workers with this instrument, but is to be regarded as the normal progress of any project at a university.

Some attention has been paid to reducing the well known causes of errors in the low energy part of the spectrum. The author is grateful to Mr. J.H. Moon for procuring a suitable form of nylon and for assistance in developing the technique of making thin films. This has made possible the use of counter windows a tenth the thickness of those previously used.

Sincere thanks are herewith expressed to Professor J.S. Foster,
Director of the Radiation Laboratory, for his lively interest in this
work. The considerable assistance rendered by Mr. J.S. Fraser in discussions and actual experiments was greatly appreciated. Acknowledgements
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assisting in preparation of the diagrams.

#### SUMMARY

The beta and gamma spectra of 6.9 day lutecium have been measured using a thin lens spectrometer. Gamma rays of average energies  $112.2\pm0.6$  kev.,  $206.3\pm1.0$  kev., and  $317.3\pm1.5$  kev., have been found, the former two of comparable intensity and the latter much weaker. Characteristic X-radiation from Hf has also been observed. These energies are computed from four conversion lines on the beta spectrum at  $46.9\pm0.5$  kev.,  $101.8\pm0.6$  kev.,  $109.9\pm0.6$  kev., and  $141.4\pm0.8$  kev., as well as from photoelectron lines using lead, gold, and tin radiators. Beta end points based on a Kurie plot occur at  $169\pm10$  kev.,  $366\pm25$  kev., and  $495\pm5$  kev. A decay scheme is proposed consistent with these results. The great importance of avoiding source charging has been demonstrated by shifts of the conversion lines up to 20 kev. when a source mounting of thin nylon was used. Decay has been followed for 400 hours and the half period found to be  $6.98\pm0.10$  days.

#### INTRODUCTION

# 1. Previous Work on 6 day Lutecium.

Hevesy and Levi<sup>(1)</sup> first reported the lutecium activity under investigation in this thesis. They assigned to it a period of 5 days which they later corrected to 6 to 7 days<sup>(2)</sup>. They attributed this activity to Lu<sup>176</sup>, since their method of activation was neutron bombardment and Lu<sup>175</sup> was the only stable isotope of lutecium known at that time. A second isotope, Lu<sup>176</sup> occurring in natural lutecium with an abundance of 2.5% was discovered in 1939 by Mattauch and Lichtblau<sup>(3)</sup>. This isotope is naturally radioactive with a half period of the order 10<sup>10</sup> years<sup>(1)</sup>.

In 1943 Flammerfeld and Mattauch investigated the beta radiation of 6 day lutecium by absorption in aluminum and reported an end-point of 0.440 Mev. [5] Later Atterling, Bohr and Sigurgeirsson [6] made observations with a cloud chamber and found the end point to be 0.47 Mev. Bothe [7] reported the maximum energy to be 0.52 Mev. for the beta rays, a result also obtained by absorption in aluminum. He also found gamma radiation of energy 0.2 Mev. and a soft component which he called ytterbium X-radiation with an energy of about 60 Kev. He accounted for these X-rays by asserting that the 6 day lutecium activity decayed both by K capture and by negatron emission.

Bothe's explanation was plausible as long as the active isotope was assumed to be  $Lu^{176}$ . Inghram, Hayden and Hess<sup>(8)</sup> showed conclusively however that this activity must be assigned to  $Lu^{177}$ .

In the isotope tables of Seaborg and Perlman<sup>(9)</sup>, there is a reference to unpublished data by Wilkinson and Hicks in which a weak gamma ray of 1.3 Mev. is reported and the 0.2 Mev. gamma ray confirmed. Their value for the half life was 6.9 days to be compared with Bothe's

value of 6.8 days and 6.6 days obtained by Atterling et al<sup>(6)</sup> and Flammerfeld and Mattauch<sup>(5)</sup>.

# 2. Beta Decay.

Early in the history of radioactivity beta rays were identified as negative electrons. Studies of alpha active materials showed the existence of sharply defined energy levels in the nucleus and accordingly beta particles with definite energies were expected. In fact for some time it was thought that this was so until it was shown that lines in the beta ray spectrum are due to secondary electrons arising from gamma radiation. In all cases studied up to the present the primary beta spectrum is a continuum with a well defined upper limit.

The existence of this continuum presents a problem for which no completely satisfactory solution is available even at the present time. The difficulty arises from the fact that all known beta active materials are nuclei of a definite weight and in a definite energy state. This is likewise true for the product nuclei. The transition must therefore involve a definite energy loss and yet the experimental evidence points overwhelmingly towards the beta particle carrying away an amount of energy which varies from nucleus to nucleus. The difficulty cannot be avoided by arguing that the observed beta particles lost energy of varying amounts before detection. The total amount of beta energy from a known number of disintegrations when measured calorimetrically turns out to give each beta particle an average energy much closer to the average for the continuum than to the upper limit (10) To postulate the non-conservation of energy for an individual atomic process does not clear up the difficulty since conservation of momentum and statistics likewise do not hold.

On the other hand for cases where the difference in energy

corresponding to the upper limit of the continuum. If as suggested by

Pauli an undetected particle (called a neutrino) is emitted with the beta

particle the energy carried away by the two particles can for every process

equal the value required by the energy conservation law. Furthermore if

this new particle obeys Fermi statistics and has a spin \( \frac{1}{2} \) the remaining

difficulties are also removed. The neutrino can have no charge and possibly

no magnetic moment. Calculations show that its rest mass must be only a

few percent of that of the electron. It is to be expected that such a particle

would interact with matter only very slightly. Attempts to detect the

neutrino have usually involved the observations of recoils following orbital

electron capture. For these cases the neutrinos should be mono-energetic.

The difficulties involved in these and other neutrino experiments have been

recently reviewed by Crane(11).

## 3. Fermi Theory.

Fermi<sup>(17)</sup>developed Pauli's neutrino hypothesis by adding an assumed interaction term to the Hamiltonian for the nuclear system and making certain approximations. He arrived at the following distribution function for the electrons from a beta active nucleus:

$$P(W)dW = \frac{G^2}{2\pi^3} |M|^2 F(Z,W) pW(W_0 - W)^2 dW$$
...(1)

where PdW == probability per unit time for the emission of a beta particle in the energy range W to W + dW

G = Fermi constant of the order  $10^{-11}$  to  $10^{-12}$ 

M = matrix element for the transition

F = Fermi function

p = mmmentum of the beta particle in units mc.

W = energy of the beta particle in units  $mc^2$  including the rest energy.

The function F(ZW) expresses the effect of nuclear charge upon the distribution. The atomic number Z is to be taken positive for negative beta particles (negatrons) and negative for positrons. It has the effect of increasing the relative number of slow negatrons and fast positrons. Explicitly it is given as follows:

$$F(ZW) = 4(2pR)^{2s-2} e^{\pi \alpha ZW/p} \frac{\int \int (s+i\alpha ZW/p)^2}{\int \int (2s+1)^2} \dots (2)$$

with

R = nuclear radius

 $\alpha = 1/137$  (fine structure constant)

$$s = (1 - \alpha^2 z^2)^{\frac{1}{2}}$$

 $\Gamma$  = gamma function

Tables for F for both positive and negative Z in intervals of 5 units and for eight appropriately spaced values of W have been published by Flugge (12).

In the distribution eq..(1) M is expected to be constant for a restricted class of transitions called "allowed". For these  $\triangle I = 0$  where I is the angular momentum. Otherwise the transition belongs to one of the forbidden classes, the order depending on the number of terms in the expanded wave functions which must be retained for M to be other than zero.

For a more detailed account of the Fermi theory the reader is referred to a review article by Konopinski (13). In this paper a procedure is given for assignment of the order of forbiddenness of a spectrum through an examination of the half-life and the maximum beta energy.

Failure of an observed spectrum to agree with the distribution

(1) may be attributed to the presence of two or more distributions of which only the sum can be observed. If one is assured that the failure is not

instrumental in origin then it is possible to analyse the observed spectrum into its components. The specific procedure for doing this, first used by Richardson and Paxton<sup>(14)</sup> is outlined below. The present status of the Fermi theory however, is not sufficiently secure to be certain of complexity on this basis alone<sup>(15)</sup>.

# 4. Kurie Plot.

In most beta ray spectrometers the momentum interval observed is proportional to the momentum itself so that a plot of counting rate N against momentum p unduly emphasises the higher energies. This can be corrected by plotting N/p in place of N. Observing the relations

$$p^2 + 1 = W^2$$
, pdp = WdW

and noting that P(W)dW = P(p)dp one can re-write eq. (1) in the form

$$\sqrt[N]{\frac{N}{p^3F(p,Z)}} = k (W_0 - W)$$
 ....(3)

where k is independent of W.

A plot of the left hand side against the kinetic energy of the electrons should be a straight line intercepting the abscissa at the maximum beta energy. Should this plot depart from straightness when there is independent evidence for complexity one can obtain straight lines representing the other components by successive subtractions made in the appropriate manner. The intercepts of these represent the lower end points.

## 5. Methods of Measurements.

### a) Beta Radiation.

If sufficiently high specific activity is available it is possible to utilize an electric or magnetic field to focus the emitted electrons upon a detector remote from the source. The focal point is different for electrons

of different energies. The procedure is either to move the detector along the locus of foci and so obtain a curve of intensity against energy or to do this leaving the detector in a fixed position and focus electrons of different energies upon it by varying the field strength. Since electric focussing fields have rerely been used only magnetic instruments will be discussed.

In general, two geometric arrangements are possible. In the so-called 180° type, for focussing purposes use is made of the fact that circular electron trajectories of the same radius emanating from a common point will come to an approximate focus if they do not make large angles with each other. Refinements (16) of this early form of instrument have permitted larger angles of divergence from the source to be used without interfering with the sharpness of focus. Recently a modified form (18) has been designed and put into service in which focussing occurs in two dimensions. This has the advantage of increasing the effective solid angle of the detected electrons subtended at the source. The second arrangement utilizes the focussing effects of the field of a number of concentric turns of wire upon electrons from a source placed at a point on the axis. The turns may be concentrated in a coil of relatively short length or distributed in a solenoid over an extended region giving rise to the names "thin lens" and "thick lens" spectrometer, respectively. These instruments manifest a phenomenon analogous to chromatic aberration in an optical lens. In this case the locus of focal points for different energies is a portion of the axis itself. It is usually more convenient to vary the field strength than to move the detector in this type of instrument.

All magnetic spectrometers suffer the defect of utilizing only a small solid angle of radiation from the source. Their great advantage is high accuracy and good resolution. However for sources of low specific

activity they cannot be used. In this case the known manner in which electrons of different energies are absorbed by various materials (chiefly aluminum) is of service. The procedure is to set up a detector near a source and note the variation in detected radiation as thickness of absorber is increased. It is possible to analyse the curve so obtained and deduce the maximum electron energy from it with fair accuracy. Solid angles many times larger than are possible in a spectrometer can be utilized.

whatever experimental procedure is followed in obtaining the beta spectrum it is always highly desirable to reduce to the barest minimum the amount of material placed in the immediate vicinity of the source. One always strives therefore to obtain the source material in as concentrated a form as possible. The extent to which this is achieved is usually expressed by quoting the specific activity in millicuries per milligram. The supporting material or source backing must likewise be as light as possible. Plane sources are usually used and the thickness of source and backing expressed in weight per unit area.

The necessity for these precautions arises from the fact that in the low energy region the detector will always record electrons which have suffered one or more scattering collisions with material in the vicinity of their origin and have had their energy reduced to some extent. This effect can produce a more or less serious distortion of the lower part of the spectrum which may be wrongly interpreted as either complexity of decay or as failure of existing beta decay theory to apply to the nucleus being studied. Recently several reports have appeared in which special attention has been paid to the low energy region (19).

In reducing the source backing thickness to the extreme limit that appears to be necessary another difficulty arises which can produce very large errors. This is the effect of source charging (20) and arises because the backing materials most suitable in other respects are all very

good electrical insulators. This matter is discussed further on Page 17.

### b) Gamma Radiation.

Gamma ray energies are usually obtained by measuring the energies of secondary electrons produced by them. These secondaries may arise in the same atom as the gamma quantum itself. They appear then as internal conversion lines superimposed on the continuous beta spectrum. Internal conversion does not always occur and when it does one must decide what binding energy must be added to the energy of the observed electron to obtain the gamma ray energy. This cannot always be done with certainty. It is desirable therefore whenever source strength will permit, to perform supplementary experiments.

In these experiments a thin metallic foil (radiator) of high atomic number replaces the source in the spectrometer. Immediately behind this foil, as close to it as possible is placed a thin walled capsule containing the source material. The wall thickness must be just enough to stop the most energetic beta rays from the source. The gamma rays knock out photoelectrons from the foil and these pass through the spectrometer in the usual menner. The binding energies to be used are usually quite apparent from an inspection of the spectrum, the intensity being greatest for K electrons. If the intensity is sufficient an L line should follow each K line, separated from it by the known difference between the K and L binding energies for the element used in the radiator. This serves as a check in the correctness of identification. If ambiguity still exists this can nearly always be removed by performing another experiment using a different element for a radiator.

#### c) Decay Scheme.

After the beta and gamma spectra have been obtained an attempt is

made to determine the energy levels in the parent and daughter nuclei between which transitions occur. For this purpose a Kurie analysis of the continuous beta spectrum is helpful. If beta end-points are indicated whose differences are equal to observed gamma ray energies a decay scheme can be tentatively drawn-up. This, however, is subject to confirmation by suitable coincidence experiments. Instrumental effects such as scattering and absorption in the source can produce cerrors which taken with the uncertainty of the applicability of the Fermi theory, make it unwise to rely too heavily upon the Kurie Plot (15). The appropriate coincidence experiments to be performed depend upon the particular scheme proposed. Usually the eta -  $m{y}$  coincidence rate is observed and the ratio of this to the beta counting rate plotted against absorber thickness placed in front of the beta counter. If gamma rays in cascade are indicated by the decay scheme, gamma-gamma coincidences must be observed, taking into account the possible loss of observable gamma rays through internal conversion as indicated by the beta spectrum. coincidence method of analysis has been thoroughly discussed in numerous papers (21), (22), (23).

#### d) Detectors.

Geiger-Mueller counters are still the most widely used detectors. Without exercising elaborate precautions, reliable counters can be easily constructed. In fact the highly stabilized voltage supplies which are now available make it possible to use counters which a few years ago would have been useless. The criterion of excellence for a counter is the extent and slope of a relatively flat portion of a plot of counting rate for a given source and geometry against applied voltage.

When G.M. counters are used for beta detection they must be provided with an entrance window. If the source is placed in a region where

the pressure and kind of gas is different from that used in the counter (usually argon + 10% organic vapour, total pressure about 10 cm Hg.), the window must be closed by a membrane. This membrane must have the following characteristics: sufficient strength to withstand the pressure difference, impermeability to any gas in contact with it, stability in the presence of these gases, sufficient thinness to transmit the lowest energy electrons to be observed without less, uniform effective area for all electrons more energetic than this. The last point applies especially to windows provided with a supporting member for very thin membranes. This member must not itself be penetrable by the most energetic electrons observed.

Thin mice has long been used as the material for the window covering. It has the advantages of great strength, resistance to heat, stability in the presence of organic vapours. With a little practice there is no difficulty in obtaining membranes 1 mg/cm<sup>2</sup> thick which are satisfactory for measurements above about 40 Kev. For lower energies various organic materials such as zapon, nylon, formvar, and collodion are being widely used. These can be obtained thin enough to transmit 2 Kev. electrons.

For the 180° type of spectrometer the photographic method of detection has proved very useful for observing weak conversion lines. It is perhaps still the most precise way of obtaining conversion line energies.

Recently crystal counters used in conjunction with photomultiplier tubes have been adopted. These have a much higher gamma counting efficiency than G.M. counters. Very high counting rates can be used with negligible loss.

#### **APPARATUS**

#### 1. Spectrometer.

The McGill spectrometer has been described in detail previously (24). For the sake of completeness the summary of the more important dimensions will be repeated here.

The spectrometer is of the thin lens type (25) and is shown schematically in Fig. 1.

There are four sections to the magnet coil each consisting of 900 turns of No. 10 gauge double cotton-covered copper wire, sections being separated by  $\frac{1}{h}$  copper cooling tubes.

The spectrometer tube, about 40 inches in length and 8 inches in diameter, is held in place by six set screws attached to the drum on which the coil is wound. This allows flexibility for line-up and also permits the magnification to be varied by sliding the tube through the coil along its axis. The aluminum baffles can be interchanged to vary the transmission zone. A cylinder of lead about six inches long and two inches in diameter is supported on the axis at the centre to reduce the gamma background in the counter.

#### 2. Current Regulator.

The power supply consists of a 20 horse power,220 volt, 3-phase, 1200 r.p.m. induction motor coupled directly to a 12 horse power D.C. motor used as a generator. The output had previously been controlled through the generator field entirely, a deviation of the current from the desired value giving rise to a signal which produced a correcting change in the generator field. Owing to the long time constants involved such an arrangement is very susceptible to sustained oscillations (hunting). Through certain corrective measures which had been taken, a usable output had previously been obtained. However since a rough test later indicated that fluctuations as

great as one in two hundred sometimes occurred and that the regulation was even worse at high currents, the circuit previously used (24) was modified in some respects.

As a first step in reducing the delay time, the exciting generators were replaced by a power supply consisting of a Hammond Type 722 transformer with two Type 866 mercury vapour tubes connected as a full wave rectifier. The output of this supply was well filtered by two filter units each consisting of a 10 henry choke and 20 mfd. of capacity. In series with this supply and the generator field coils a bank of five 6A3 triodes in parallel was inserted. The amplified error voltage applied to the 6A3 grids changed the field current in a manner to correct the error. This arrangement, while eliminating some mechanical trouble spots did not show a marked improvement, indicating that most of the delay time had its origin in the generator and not the exciters.

A second feedback loop was then added to the system. This consisted of a bank of eight paralleled 6AS7 triodes in series with the magnet coil. The grids of these were supplied by the same voltage as that on the 6A3 grids. Since the eight 6AS7's could carry a maximum current of two amperes, these were shunted by a variable resistance which could be set at a suitable value. The setting usually chosen in operation was such that the triodes carried their maximum current. When the output network of the phase sensitive rectifier was chosen as shown this second loop was fast enough in its action to reduce hunting to imperceptible proportions (Fig.3) The percentage regulation can be estimated by observing the galvanometer. During operation this made slow random excursions seldom greater than 15 mm. which represents an error of 35 microvolts in amplitude across the standard resistance. The lowest voltage used in subsequent measurements

<sup>\*</sup> A circuit similar to this had previously been used by Mr. D.A. Anderson for his work with nuclear magnetic moments.

corresponding to the lowest energy observed was 60 milivolts corresponding to a regulation of about 0.6 per thousand. The galvanometer fluctuations were usually smaller for higher magnet currents resulting in considerably better regulation. This discussion does not include slow drifts which might have occurred if the system were left to itself. However, these were easily eliminated by occasional manual operation of a "variac" on the input of the No. 722 transformer. It can be said with certainty that errors and fluctuations in the current were entirely negligible in comparison with other errors.

In the present location of the instrument the earth's field has shown no appreciable change in direction over a period of six months. The current required in the system of compensating coils to minimize this field appears not to have changed either. While there is no single set of current values which will eliminate the earth's field entirely throughout the length of the tube, calibration tests show that the residual field cannot be very serious.

#### 3. Detectors.

A Geiger-counter was used as a detector throughout. This tube was a conventional point counter constructed as shown in Fig. 2(a). The window was 3/16 inch in diameter, covered with a nylon membrane. These were prepared following closely the directions of Saxon et al<sup>(26)</sup>

It should be noted that there are different types of nylon and that not all of these are soluble in isobutyl alcohol. The nylon used in this work was a sample supplied through the courtesy of Dr. Saxon.

A sheet of mica with a circular hole of a suitable size cut in it proved a very satisfactory frame for removing the films from the water surface. There was no difficulty in making the layers adhere to each other

and to the mica. After preparation the films were always left on the frame until used. The film was then carefully laid over the window which was previously painted with a thin layer of collodion. After the collodion had hardened the mica was cut off with a razor blade. In this way there was no wrinkling and less chance for collodion to get on the part of the film covering the window. Thinned collodion often ruptured the film. After removing the mica, collodion was painted liberally over the edge and top of the film up to the lip of the window. These operations nearly always produced a gas-tight seal. When used in this way the window could withstand the counter pressure for a few hours, but it stretched slowly and would usually be leaking when the counter was refilled for a second run. To overcome this a brass supporting member as shown in Fig. 2(a) was secured over the window with glyptal cement. After this was done windows 0.08 mg/cm<sup>2</sup> thick did not fail upon refilling the counter. A leaking window seldom increased the gas pressure in the spectrometer appreciably but a leak was always revealed by a counter characteristic which shifted continually towards lower voltages.

ethylene with a total pressure of 10 cm. to 12 cm. On two occassions when an attempt was made to use ethyl alcohol as a quenching agent the window ruptured shortly afterwards. The argon-ethylene mixture produced long slowly rising plateaus unless contaminated with air. However, even a poor characteristic was quite usable with the high voltage supply which was used. In any case during the course of all runs the counter was frequently checked by placing a Co<sup>60</sup> source in a standard position and observing about 8000 counts.

The counter was connected directly to a Model-1000 Berkeley Decimal Scaler\*. This instrument, which includes a high voltage supply, was convenient to use and, in general, quite satisfactory.

<sup>\*</sup> Berkeley Scientific Company, Richmond, California.

### 4. Alignment.

In order to realize the full resolving power of the instrument, the source and counter window must both be accurately centred along the axis of magnetic field. To do this an active deposit of RaTh was placed in the spectrometer as a source. The approximate location of the intense F line was known from previous work with this instrument. After aligning the tube visually as well as possible a run was taken on the F line. With the current in the coil set for the observed maximum counting rate, slight adjustments were made until the counting rate at the peak reached a maximum. This method of alignment gave a line width slightly less than that reported by previous workers. Later a check was made by replacing the counter by a piece of X-ray film.\* No further adjustment was found to be necessary.

#### 5. Calibration.

Several ThB sources were prepared during a period of six weeks. The location of the F line was observed with each of these and the peak was always found to be reproducible within less than 0.5%. The line width was about 1.7% at half maximum. After the technique of making nylon windows had been developed, the A line (24.7 Kev.) was also observed and its energy based on the F line calibration agreed with that given by Ellis (27) within 1%.

To check the linearity of the instrument in the high energy region a source of Co<sup>60</sup> was prepared in a capsule covered with a 28 mg/cm<sup>2</sup> lead radiator and the photoelectron lines due to the two gamma rays were observed. While their energies are not known with high accuracy, the values obtained here agreed within 0.5% with those of Jensen et al<sup>(28)</sup>. This established the linearity of the instrument within 1% over the range from very low energies to about 1 Mev.

The final calibration of the spectrometer used for all measurements reported here is based upon the value 1383.8 gauss:cm, for the F  $\lim_{\to}$  (29) and given by the equation p = 4762 V, where p is the momentum in gauss-cm, V the potential drop across the standard resistance S as read on the potentiometer, (see Fig. 3).

<sup>\*</sup> This was suggested by Dr. J.L. Wolfson, Chalk River.

# MEASUREMENTS ON Lu177

#### 1. Source.

The lutecium oxide used in these experiments, obtained from Johnson Matthey and Co., London, England was claimed to be "of a very high degree of purity". Thirty-five elements are listed for which no spectrum lines were observed in their analysis. These include all the rare earth elements with the exception of ytterbium for which one line at 3289.37 A.U. was "barely visible". Two or three lines from each of calcium, silver, and copper are said to be "faintly" or "very faintly" visible.

In attempting to decide whether any of the radiations observed from this sample when activated by slow neutrons are in fact due to the impurities, a careful examination of tables of isotopes (9),(30),(31) was made. Consideration of activation cross sections, half-lives, percentage abundances make it possible to dismiss all activities with the possible exception of those shown in Table I.

TABLE I

Contaminant and abundance	Thermal neutron cross section	Activity Produced.	Half Period	Energy of Radiation (Mev).		
Yb <sup>174</sup> (29.58%)	50	Yb <sup>175</sup>	4.2d.	0.5	0.35	
Ag <sup>109</sup> (48.65%)	97	Ag <sup>110</sup>	225 d.	0.59	1.4	(%)
					0.90	(47%)
					0.66	(44%)

In addition one must consider the possibility that elements with a large thermal neutron cross section may be detected more sensitively by gamma and beta ray spectroscopy of the activated sample than by optical spectroscopy. (32)

Two samples of this lutecium oxide were irradiated separately. The first (about half a milligram) was contained in a capsule of commercial grade aluminum and activated to a specific activity of 20 mc./mg. Sources for obtaining the beta spectrum were prepared from this. In order to avoid contaminating the lutecium oxide with aluminum and radioactive impurities in the aluminum, a drop of distilled water was put into the capsule and agitated slightly with a small glass rod. A dropper terminated by a capillary tube was then used to transfer a minute drop to the source backing. When this dried a slight stain having no visible irregularities appeared. Very strong sources could be prepared in this way. When handled carefully the activity appeared to be sufficiently adherent not to require coating. In view of this and uncertainty of the thickness of an applied layer of some suitable material such as collodion, sources were used without further treatment.

The details of the source mount are shown in Fig. 2(b). This arrangement allows a free space of two inches behind the source mounting.

# 2. Observations with Nylon Backing.

A source was prepared using as backing a 0.02 mg./cm<sup>2</sup> nylon film about half an inch in diameter, supported on a thin mics annulus (about 10 mg/cm<sup>2</sup>). Using a centering jig this was stuck with collodion to a second piece of mics with a half inch hole in it and of external diameter suitable for placing in the spectrometer. The beta spectrum was observed during about seven hours, spacing observations at currents corresponding to every 3 millivolts for the lower energies and every 5 millivolts for higher energies. Four conversion lines were found and the end point agreed approximately with that quoted in the literature where absorption methods were used. The next day further observations were made in the vicinity of the conversion lines. The peaks, particularly of the lowest energy line, appeared to have moved slightly towards lower energies. Since for the first run the statistics

were poor no very great importance was attached to the discrepancy. A second source several times stronger than the first was prepared, likewise using 0.02 mg./cm<sup>2</sup> nylon backing. In view of a report on the effects of source charging (20) this nylon film had a thin layer of aluminum evaporated upon it. The darkening was perceptible when viewed with transmitted light but the appearance was not metallic. No special precautions were taken to assure electrical contact between the spectrometer tube and the aluminum film, it being assumed that the mica would provide sufficient conductivity. The spectrum obtained from this source showed the same four lines but all at appreciably lower energies. A second run on the lowest energy line showed that it had moved to even lower energies. It was then quite apparent that the source was charging, and had reached a positive potential at least 10 kilovolts with respect to the spectrometer tube. The aluminum coating on the nylon provided a wholly inadequate conducting path for the source strength used. The rather short half life of the source made it inappropriate to investigate further at this time suitable means of grounding a source mounted on a thin nylon backing.

#### 3. Sources Mounted on Aluminum.

Aluminum foil 3 mg/cm<sup>2</sup> was available in the Laboratory and this was used as backing for the next attempt. As a preliminary test a run was taken on the lowest energy line and repeated 12 hours later. The two runs agreed remarkably well in the location of the peak and the shape of the line. This agreement made it quite safe to proceed. The whole spectrum was then observed checking the counter at hourly intervals against the standard cobalt source. In order to minimize decay corrections the observations were completed in less than 24 hours. A careful search was made for other conversion lines but none was found. Repeated observations at the peaks of the lines revealed no sign of drift.

Since a backing as heavy as 3 mg/cm<sup>2</sup> can produce appreciable distortion in the low energy region aluminum foil helf this thickness was procured commercially and etched to 0.5 mg/cm<sup>2</sup> chemically\*. A somewhat weaker source was prepared with this as backing and the spectrum observed. The result is plotted in Fig. 5. In the same figure the spectrum obtained with the second source (nylon backed) is also shown. The two curves were fitted at the high energy end. Of course, the curve obtained from the nylon backed source, does not correspond to a specific instant of time. The points were taken from left to right. The lowest energy line had certainly shifted even further to the left by the time the data for the highest energy line was taken. A considerable reduction in the intensity of the low energy line can be noted. This may be partially caused by a defocussing action of the electric field produced by the charged source.

The line width at half maximum was about 2.2% for all four lines in the spectrum using the 0.5 mg/cm<sup>2</sup> aluminum backed source. The width was greater by a small but perceptible amount when the 3.0 mg/cm<sup>2</sup> source backing was used.

### 4. Location and Origin of Conversion Lines.

The energies of the conversion lines and their origin are shown in Table II.

TABLE II
Conversion Lines of Lul77

Energy	Electron Shell	Gamma Energy
46.9 ±0.4	K	112.1
101.8 ±0.6	L	112.4
112.9 ±0.6	M	112.4
141.4 ±0.8	ĸ	206.6

<sup>\*</sup> This was done by Mr. John Fraser.

It is clear that within experimental error the separation of the first three lines indicate they arise from the same gamma ray. The fourth line may reasonably be assigned to a second gamma ray which is weakly converted in the K shell. The last column is the sum of the first and the corresponding binding energies of Hf\*. The treatment of the lines as shown in Table II is to be regarded as tentative pending confirmation by later exemination of the gamma spectrum.

### 5. Kurie Plot.

Kurie plots (Fig. 6) were made using the data obtained from the two aluminum backed sources\*\*. It is to be noted that the two sets of data agree well from the upper limit down to about 135 Kev. Below this point the source on thicker backing showed appreciably more electrons as would be expected. The curve is straight from the end point down to a point somewhat over 100 Kev.below the end point. The departure from straightness for lower energies can scarcely be ascribed to back scattering effects alone in view of the relatively small effect of a factor 6 in the backing thickness. Tentatively regarding the shape of the curve as an indication of complexity, two subtractions were made yielding two lower end points. The residue after the second subtraction still showed a curvature at the lower energy end. However the data was so meagre and subject to such large errors that a third subtraction was not warranted.

<sup>\*</sup> The binding energies used here were obtained from Compton and Allison (33), the M binding energy of Hf being an extrapolation of those for OS and W. The average of  $L_{\rm I}$ ,  $L_{\rm II}$ , and  $L_{\rm III}$  was used.

<sup>\*\*</sup>Owing to the lesser strength of the source mounted on 0.5 mg/cm<sup>2</sup> aluminum, the data has been multiplied by the factor 1.46.

A third set of points shown in Fig. 6 was obtained from the source with thinner backing. This data was taken with a view to improving the accuracy of determination of the upper end point.

The energies corresponding to these three end points are 169±10 kev., 366±25 kev., and 495±5 kev. The errors are obtained by considering possible variations in the placing of the straight lines. The reality of the lower end points is subject to confirmation by independent experimental data. Such data in fact has already been provided by the conversion lines. The sum of the gamma rays is equal to the difference between the extreme end points; and the lower gamma energy is equal to the difference between the two higher end points within experimental error. The possibility of this agreement being an accident cannot be excluded in view of the relatively large errors in the Kurie analysis. Appropriate coincident experiments could very well provide the necessary additional confirmation\*.

Assuming the lower end points to be real, distribution curves can be calculated from the straight lines by inverting the operations. These are shown in Fig. 7. The areas under the curves have been measured with a planimeter yielding relative intensities 65%, 16.7%, 18.3% with errors of about 10%. It is also possible to obtain from these areas the total number of electrons recorded in a given time interval. For a single distribution the number is given by

$$\int_{Q} \frac{N}{\Delta p} dp = \int_{Q} \frac{N}{Rp} dp = \frac{1}{R} \int_{Q} \frac{N}{V} dV = \frac{\text{area}}{R}$$

where p is momentum, R the resolving power (1.7%), N the recorded number of counts at a point, Ap the momentum interval at the same point and V the potentiometer voltage. Performing the operations the result 1500 electrons

<sup>\*</sup> The only coincidence mixer available failed after the source arrived and these important experiments could not be carried out.

per second is obtained. These are emitted from the source into a transmission zone subtending an angle  $6.7 \times 10^{-4}$  of the total solid angle

Correcting for the reduction in counter window area due to the supporting member, the figure 1500 should be increased to about 2000. This corresponds to a source strength of about 80 microcuries for the stronger source and 38 microcuries for the weaker. Considering the source area and using the specific activity 20 mc/mg with a decay factor, source thicknesses less than 0.1 mg/cm<sup>2</sup> are obtained.

#### 6. Gamma Ray Measurements.

After the beta spectrum had been observed an aluminum cup with wall thickness 0.030" and  $\frac{1}{4}$ " outside diameter was prepared and as much of the remaining activity as possible transferred to it. The cup was cemented in a central hole in a 1.25" aluminum disk and covered with an aluminum foil to assure electrical contact. Over the capsule was placed a 14.7 mg/cm² lead radiator. The counting rate with this source was low but well above background. Distinct photoelectron lines appeared which were immediately recognized as being due to the two gamma rays mentioned in Table II. This provided a satisfactory confirmation of the identification of the conversion lines.

The spectrum is shown in Fig. 8(a) and the K and L lines for each gamma ray marked. The K line for the low energy gamma appears very weakly. This is not surprising when one considers that these photoelectrons have an energy of only about 25 kev. so that the effective radiator thickness is only an outer layer about 1 mg/cm<sup>2</sup> thick.

In addition to the four lines mentioned above there appeared to be two other peaks of low intensity. These are in the places where L photo-electrons due to characteristic K radiation from the excited Hf atom should appear. This point is discussed further later.

In order to obtain better accuracy for the photoelectron energy, to examine the structure of the peaks attributed to X-rays in more detail as well as to ascertain whether other gamma rays existed, a second sample of  $\operatorname{Lu_2O_3}$  was prepared for irradiation. A capsule of 1S aluminum,  $\frac{1}{4}$  in diameter, was filled with 3 mg. of  $\operatorname{Lu_2O_3}$  and closed with a tapered plug. Since this capsule was to be irradiated and placed directly in the spectrometer, a dummy of the same weight was cut from the same piece of stock to be exposed with the capsule. The period of irradiation was two weeks giving an activity of 33 mc.

activity about one-seventieth of that of the capsule as measured with a Lauritsen Electroscope. Spectrometer observations began 24 hours later and continued for five days. The consistency of the results as well as the preferred geometric location of the Lu<sup>177</sup> activity make it unlikely that any of the photoelectron lines subsequently observed arise from gamma radiation of impurities.

A gold radiator ( $12 \text{ mg/cm}^2$ ) was used for the first run. The same set of lines (fig.8(a)) appeared as with the first gamma source. Those that had been identified as K lines were shifted about 7 Kev. towards higher energies while those identified as L remained in nearly the same locations. This is a satisfactory confirmation of the previous assignment.

In addition, there is some evidence for a weak additional line beyond the higher energy L line. This region was therefore examined with a 29 mg/cm<sup>2</sup> lead radiator. At the time of this measurement the contaminating gamma activity was estimated to be less than one four-hundredth of that due to lutecium. The region was swept over ten times, 50 second counts being taken at 14 appropriately spaced points. In this way the effect of instrumental drifts is minimized. A plot of these points is shown inset in Fig.  $\delta(s)$ 

A line is clearly indicated, shifted from the sharp drop in this region obtained with the gold radiator, as it should be if this is a K line. This is attributed therefore to a third gamma ray. Careful examination of the location of the line shows the gamma energy to agree remarkably well with the sum of the other two gamma ray energies. This is sufficient evidence for assigning it to lutecium.

In order to obtain further information about the X-ray lines as well as to obtain an accurate value for the lower energy gamma based on a K photoelectron line, the gamma spectrum was observed using a tin radiator (ll mg/cm<sup>2</sup>). This spectrum (Fig.8(b)) showed the K and L peaks for the two more intense gamma rays. No counts were taken beyond the upper L line.

The X-rays are energetic enough to produce K electrons from tin and these peaks were observed as well as the L peaks.

In a recent table of isotopes by Seaborg and Perlman<sup>(9)</sup>, a week gamma energy of 1.3 Mev. is reported, observed by absorption in lead. To check this an absorption experiment was performed in which a helf inch thickness of lead was placed between counter and source and the counting rate observed as additional layers of lead (4.2 g/cm²) were added. The counting rates indicated a weak high energy component which might have arisen from the contaminating activity in the aluminum. The aluminum dummy was then used in place of the capsule and the absorption experiment repeated. Subtraction of the corresponding counting rates revealed some evidence for a very weak gamma ray with energy of the order 1 Mev. It was estimated this gamma ray would produce a counting rate with a naked source well under a two-thousandth of the total gamma activity. Considering the greater efficiency of a gamma counter (copper cathode) for high energy gamma rays the ratio of intensities is even greater.

TABLE III

Absorption of Gamma Rays

Absorber Thickness	Net Counts in 500 Seconds				
g/cm <sup>2</sup>	Source	Dummy	Difference		
0	837	516	321		
4.2	639	428	211		
8.5	543	341	202		
12.8	397	239	158		
17.0	348	194	<b>1</b> 54		
21.3	<b>26</b> 2	168	94		

The geometrical arrangement used for this experiment is shown in Fig. 2(d).

#### 7. Half-Life.

A source was prepared by rubbing a dry glass rod against the inner well of the capsule containing the first activated sample. This rod was then touched lightly against a piece of mica taped to the tray under a Lauritsen Electroscope (Fred C. Henson Co.). The electroscope window was covered with a 0.35 mg/cm<sup>2</sup> formwar film. The time of discharge between the same two scale points was observed at intervals over a period of 400 hours. A plot of the natural logarithm of a number inversely proportional to the time of discharge, (corrected for background) against time is shown in Fig. 10. There is no evidence for any contaminating activity. The half life found from the slope of this curve is 6.98±0.10 days the error being obtained by drawing other straight lines through the points, differing slightly in slope.

#### DISCUSSION

The most clear cut part of this work is the determination of the gamma ray energies. This seems to leave little to be desired. Table IV summarizes the energy values obtained from the various lines for the three gamma rays. In averaging the results only values from K photoelectron lines have been used since the K absorption edge is single whereas L edges are triple. In addition the K lines are much more intense except where they occur at very low energies. The latter have not been used in the averaging process.

TABLE IV
Energy of Gamma Rays

Line	Location (Kev).	Added Binding Energy (Kev)	Energy (Kev)
Conversion	46.9	65.2 <b>(K</b> )	112.1 7/1
Conversion	101.8	10.6 (L)	112.4
Conversion	109.9	2.5 (M)	112.4
Conversion	141.4	65.2 (K)	206.6 <b>7</b> 2
Photoelectron (Au radiator)	125.3	80.9 (K)	206.2 <b>Y</b> 2
Photoelectron (Au radiator)	31.7	80.9 (K)	(112.6) <b>Y</b> 1
Photoelectron (Pb radiator)	229.1	88.2 <b>(</b> K)	317.3 $\gamma_3$
Photoelectron (Sn radiator)	82.6	29.2 (K)	111.8 <b>1</b> 1
Photoelectron (Sn radiator)	176.5	29.2 (K)	205.7 <b>Y</b> 2
Photoelectron (Pb radiator)	118.5	88.2 <b>(K)</b>	206.7 <b>γ</b> 2

The shapes of most of the photoelectron lines show that the radiators cannot be regarded as thin. The best energy value does not correspond to the peak of the line but is always greater than this. The effect of radiator thickness has recently been discussed in a paper by Jensen, Laslett and Pratt (28). Curves are presented showing the errors that can arise from thick

radiators if corrections are not applied. Perhaps the simplest way of applying a correction\* is to consider the line width as observed with a ThB Source. It is assumed then that the high energy side of a photoelectron line is unaffected by radiator thickness except near the peak. The true peak is taken as the half width (0.85%) measured in from the high energy side. This method of locating the peak has been used for all photoelectron lines. Strictly speaking a correction should also be applied to the conversion line peaks. However, these lines are not much wider than the ThF line and the correction is small.

The beta end point, 495 Kev., lies midway between the value 470 Kev., reported by Atterling, Bohr and Sigurgeirson (6) and 520 Kev. reported by Bothe (7). Flammerrfeld and Mattauch (5) obtained the value 440 Kev. The gamma energy 0.2 Mev. reported by Bothe (7) is in agreement with the value 206.3 Kev. obtained in this work.

The ft values (13) for the three components indicated by the Kurie analysis have been calculated and are shown in Table V. The partial half-lives depend on the relative intensities.

End Point (Kev)	Relative <u>Intensity</u>	TABLE V. Partial half-life t (sec)	<u>f</u>	<u>ft</u>	Group	Order
169	18.3	32.6 <b>x</b> 10 <sup>5</sup>	0.23	$7.5 \times 10^5$	113	lst Forbidden
366	16.7	35.7 x $10^5$	2.5	$8.8 \times 10^6$	1B	lst Forbidden
495	65	9.62 <b>x</b> 10 <sup>5</sup>	6.50	6.25 x 10 <sup>6</sup>	1B	lst Forbidden

The beta end points and gamma energies are consistent with the decay scheme shown in Fig. 10. There is no reasonable place on this diagram to put a very weak 1.3 Mev. gamma ray. If the lowest level shown is not in fact the ground state of Hf<sup>177</sup> one would expect to observe an additional 1 Mev. of gamma radiation as intense as the 206 Kev. gamma ray. A search was made with

<sup>\*</sup> This method was suggested to the author by Dr. J.L. Wolfson, Chalk River.

the spectrometer (using 12 mg/cm<sup>2</sup> gold as a radiator) and no other peak as intense as that of the 318 Kev. gamma ray was found corresponding to gamma energies up to 550 Kev. On the basis of this and absorption experiments of others ( ) it is fairly certain that this additional gamma radiation does not exist. In fact assuming Fig. 10 not to be correct and that the two gamma rays actually follow the high energy betas a total decay energy of 1.32 Mev. including the rest energy of the electron is obtained. It is scarcely possible to fit into this an alternative decay process involving a 1.3 Mev. gamma ray. It is therefore strongly suggested that this high energy gamma ray, in view of its very low intensity, erises from traces of impurities. In the sample used here it may have originated from the silver known to be present.

X-radiation is to be expected following orbital electron capture and following internal conversion of gamma rays. In the former case the X-rays should be characteristic of an atomic number Z-1, in the latter of Z or Z+1 depending on whether the gamma ray precedes or follows the beta decay. Z is the atomic number of the radioactive nucleus. It is interesting in the present case to see whether the resolution is sufficient to distinguish between the X rays of ytterbium, lutecium, and hafnium. Table VI shows the K lines for these three elements calculated from wavelengths given in Compton and Allison<sup>(33)</sup>. The lines fall into two groups corresponding to the L to K transitions and the M and N to K transitions respectively. Two rather broad photoelectron lines should be expected especially if arising from the multiple L shell of the radiator unless the resolution is adequate to resolve the fine structure. Of course with the radiator thickness used, broad photoelectron lines are to be expected even without fine structure.

TABLE VI

Energy of Characteristic K lines (Kev.) Calculated from Wavelengths given in

	Compton and Allison(33)			
Transition	<u>Yb</u>	Lu	<u>Hf</u>	
K-L <sub>II</sub>	51.44	53.07	54.72	
K-L	52.46	54.17	55.90	
K-M <sub>II</sub>	59.26	61.20	63.29	
K-M <sub>III</sub>	59.49	61.45	63.52	
K-N <sub>II</sub> , N <sub>III</sub>	60.99	63.08	65.09	

When lead and gold were used as radiators two peaks appeared which have energies corresponding respectively to the average of each of the two groups, diminished by the binding energy of the L shell (Fig. 8(a)). K electrons from gold and lead with binding energies of over 80 Kev. would not be expected to appear. It is not possible to state with certainty the origin of these x-rays on the basis of these spectra alone.

However the K binding energy for tin is only 29.2 Kev. making it possible for both K and L electrons to arise from each X-ray line. In addition the K photoelectron peak of the lowest gamma ray should be removed entirely from this region. The removal of this K photoelectron line and the greater complexity of the X-ray photoelectron lines is immediately evident in Fig. 8(b). Three peaks with possibly a fourth are visible. On the high energy edge of the lowest energy line a satellite (called Ai) can be seen. Calling these lines A, B, C, and D in increasing order of energy, Table VII shows their energies and the corresponding X-ray energies.

TABLE VII

Line	Energy	Binding Energy of Sn	X-Ray Energy
A	25.9	29.2 <b>(K</b> )	54.1
Al	24.9	29.2 (K)	55.2
В	33.7	29.2 (K)	62.9
C	49.9	4.2 (L)	54.1
D	58.5	4.2 (L)	62.7

Associating the lines A and Al with the two transitions  $K-L_{\rm II}$  and  $K-L_{\rm III}$  respectively the best fit occurs if these are Hf X-rays. The other lines rule out Yb but a choice cannot be made between Hf and Lu.

If the binding energies of lutecium are added to the conversion line energies one obtains the values 110.4 Kev., 112.0 Kev., 112.3 Kev., 204.9 Kev. for the gamma energies. The first three are not so self consistent as the values stated in Table IV and the agreement with the values obtained from the photoelectrons is clearly worse. This indicates that the gamma rays follow the beta rays as in fact, usually is the case.

Bothe (7) refers to a softer gamma component of about 60 Kev. which he attributes to Yb X-rays following K capture. As stated above there is good evidence that these are not Yb X-rays. Of course, Bothe was incorrect in the mass identification of the 6.9 day lutecium activity, attributing it to Lu<sup>176</sup>. (Seaborg classifies this activity among the "isotope certain" group.) Yb<sup>176</sup> is stable but Yb<sup>177</sup> is not. It is quite safe to assert that K capture does not occur. Bothe makes no mention of a gamma ray of the order 112 Kev. It apparently could not be resolved from the X-radiation. He reports coincidence experiments in which the softer component is coupled with the 0.2 Mev. gamma ray but not with the beta rays. This may be considered as consistent with the decay scheme (Fig. 10) suggested here when it is recalled that

according to it most of the Lu<sup>177</sup> nuclei decay directly to the ground state of Hf<sup>177</sup> and that the soft gamma rey is in cascade with the 0.2 MeV. gamma ray. The X-rays should also be coupled with the harder gamma ray since they follow internal conversion of the softer component.

## SUMMARY AND CONCLUSIONS

- 1. The current regulator for the McGill spectrometer has been improved by the insertion of control tubes in series with the coil of the magnet. Operation is now entirely satisfactory. The linearity of the spectrometer has been demonstrated by observation of a conversion line at 25 Kev. and photoelectron lines in the one Mev. region to be within 1%.
- 2. Thin nylon windows for the beta counter have been prepared and the low energy cut off for the spectrometer considerably reduced thereby.
- 3. The large errors that can be introduced by source charging have been demonstrated.
- 4. The gamma spectrum of Lu<sup>177</sup> has been observed through a study with the spectrometer of the photoelectrons ejected from lead, gold, and tin radiators. Three gamma rays have been found of energies  $112.2\pm0.6$  Kev.,  $206.3\pm1.0$  Kev.,  $317.3\pm1.5$  Kev. The former two are of comparable intensity, the latter much weaker.
- 5. X-rays from the source have been observed and their energies agree well with energies of characteristic K X-rays from Hf, not so well with those from Lu, and are clearly greater than those from Yb.
- 6. The beta spectrum has been studied with the same spectrometer and four conversion lines have been found with energies  $46.9\pm0.4$  KeV.,  $101.8\pm0.6$  KeV.,  $109.9\pm0.6$  KeV., and  $141.4\pm0.8$  KeV. The first three arise from the 112.2 KeV. gamma ray, the latter from the 206.3 gamma ray. There are clear indications that the gamma rays follow the beta rays.
- 7. A Kurie plot has been made and end-points at  $495\pm5$  kev.,  $366\pm25$  Kev.,  $169\pm10$  Kev. obtained corresponding to three beta groups of relative intensity 65%, 16.7% and 18.3% respectively. Evidence for the reality of the two lower energy groups is the agreement between the sum of

the two more intense gamma rays and the difference between the extreme end-points as well as the agreement between the difference between the two higher end points and the lowest energy gamma ray. This requires that these two gamma rays be in cascade. Strong evidence that this is so is provided by the existence of the third gamma ray of energy equal to their sum. If the decay scheme is assumed to be correct a more accurate estimation of the lower end points may be obtained by subtracting the appropriate gamma ray energy from the upper end point. The results are  $177 \pm 6$  Kev. and  $383 \pm 6$  Kev.

- 8. An energy diagram has been proposed consistent with these findings and apparently with coincidence measurements reported by Bothe (7).
- 9. The half life of Lu<sup>177</sup> has been measured by following the decay of a source for 400 hours. The value  $6.98\pm0.10$  days has been obtained, somewhat higher than the values reported by Bothe(7), Flammerfeld and Mattauch(5), and Atterling et al(6), but substantially in agreement with the value obtained by Wilkinson and Hicks(9).
- 10. An absorption experiment provides evidence for a very weak high energy gamma ray. Arguments are advanced to show that this gamma ray arises from a contaminating activity.

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## CROSS SECTION OF SPECTROMETER

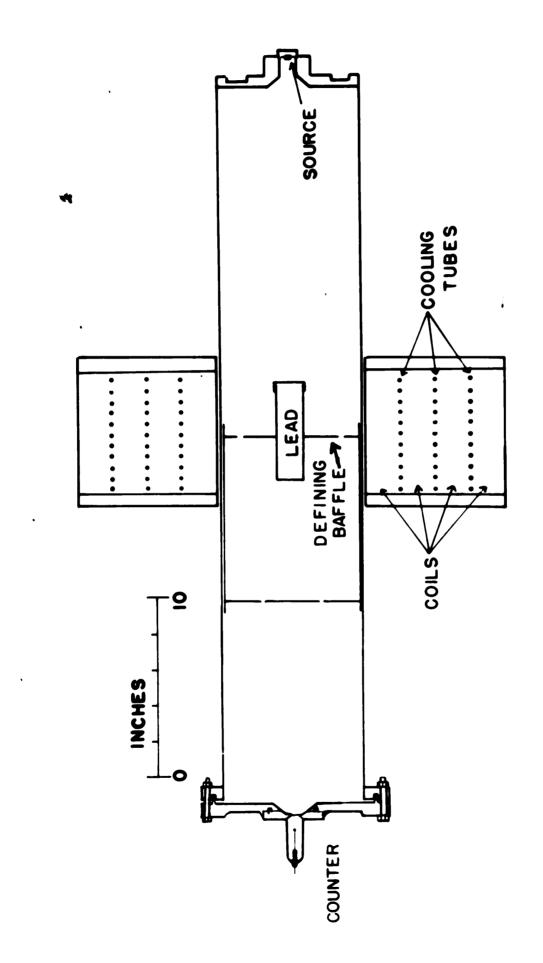
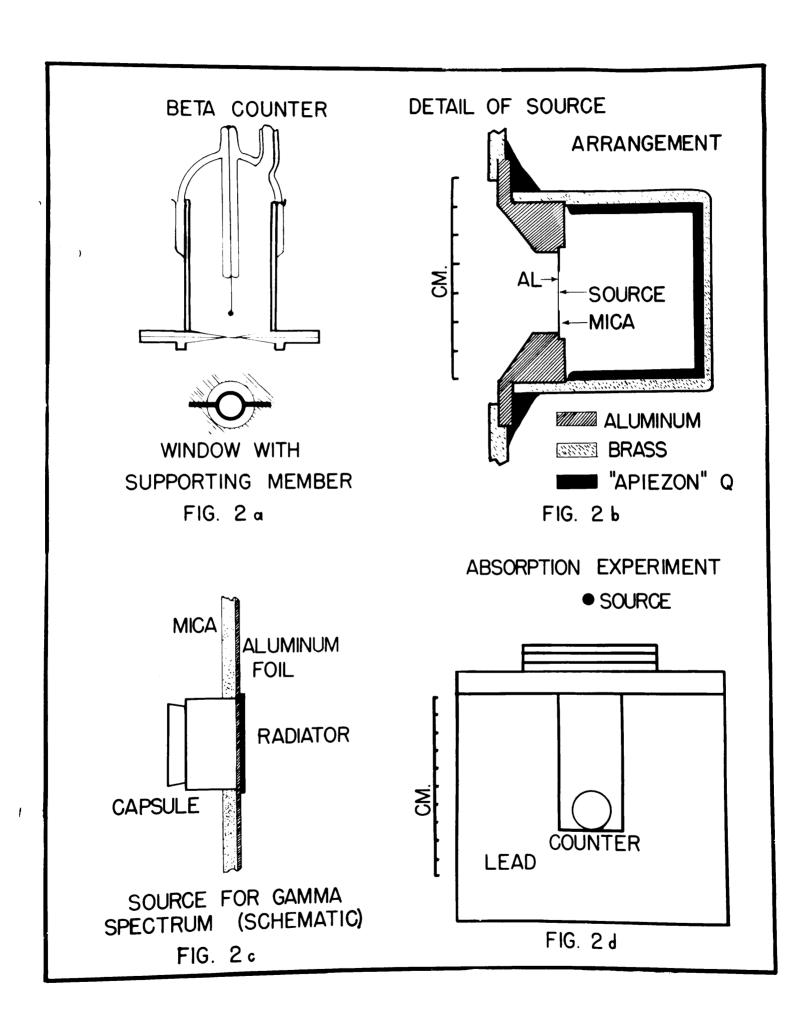
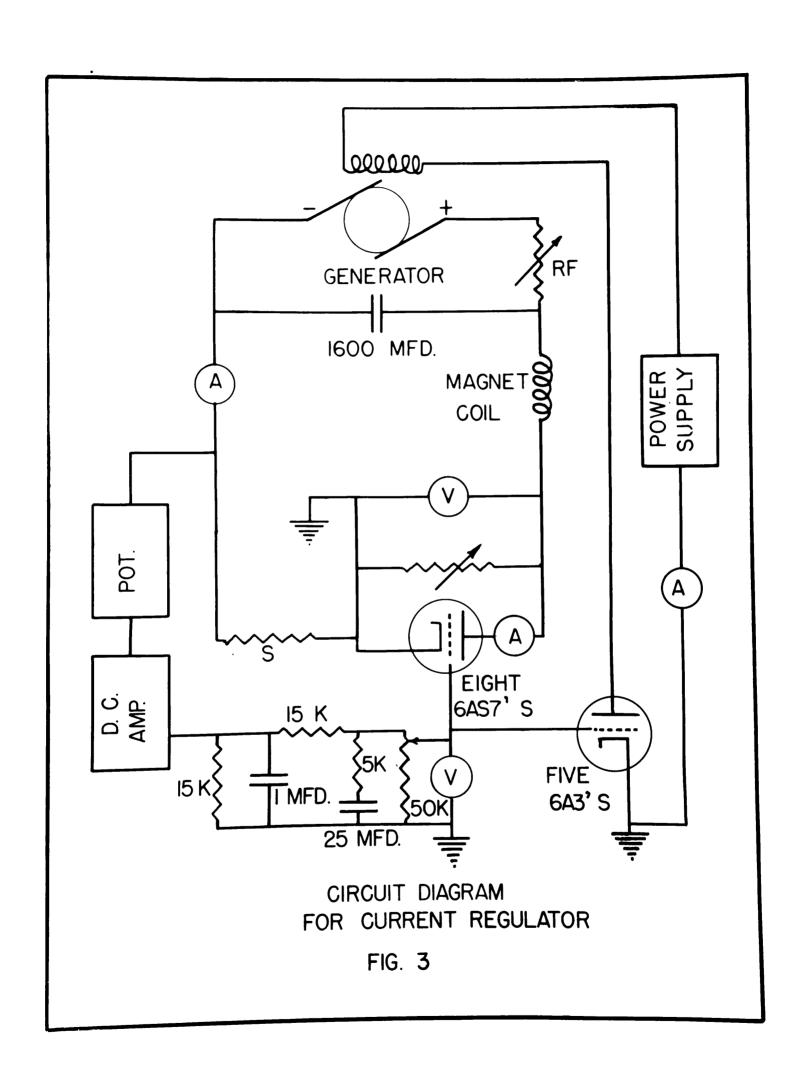
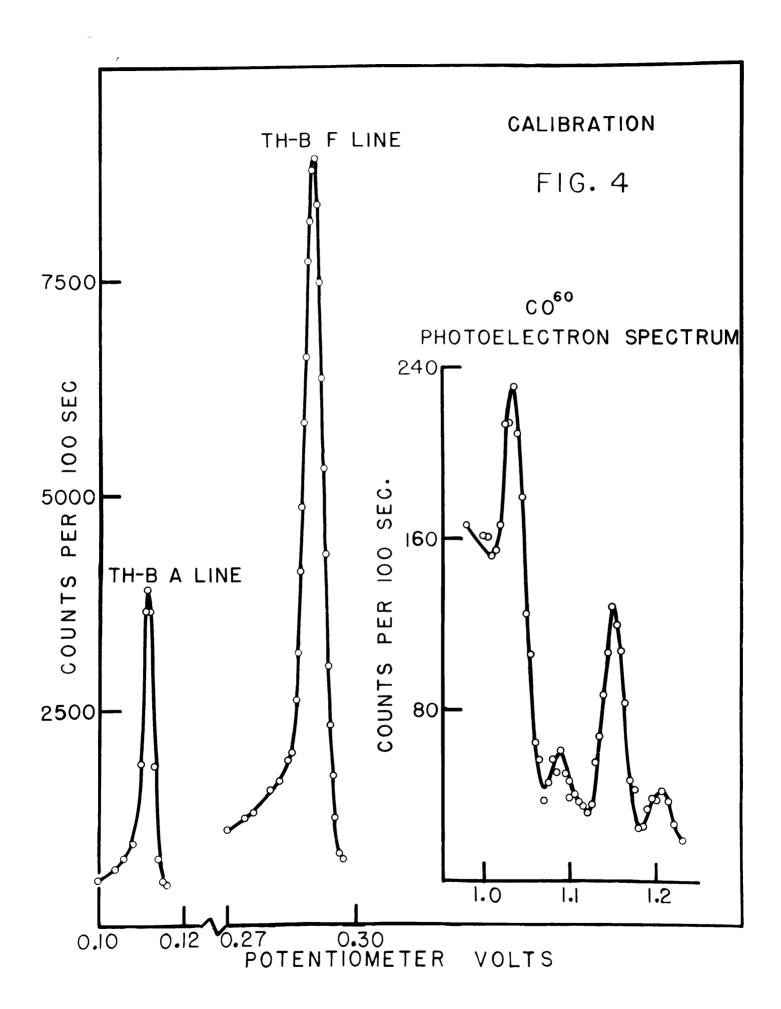


FIGURE 1.







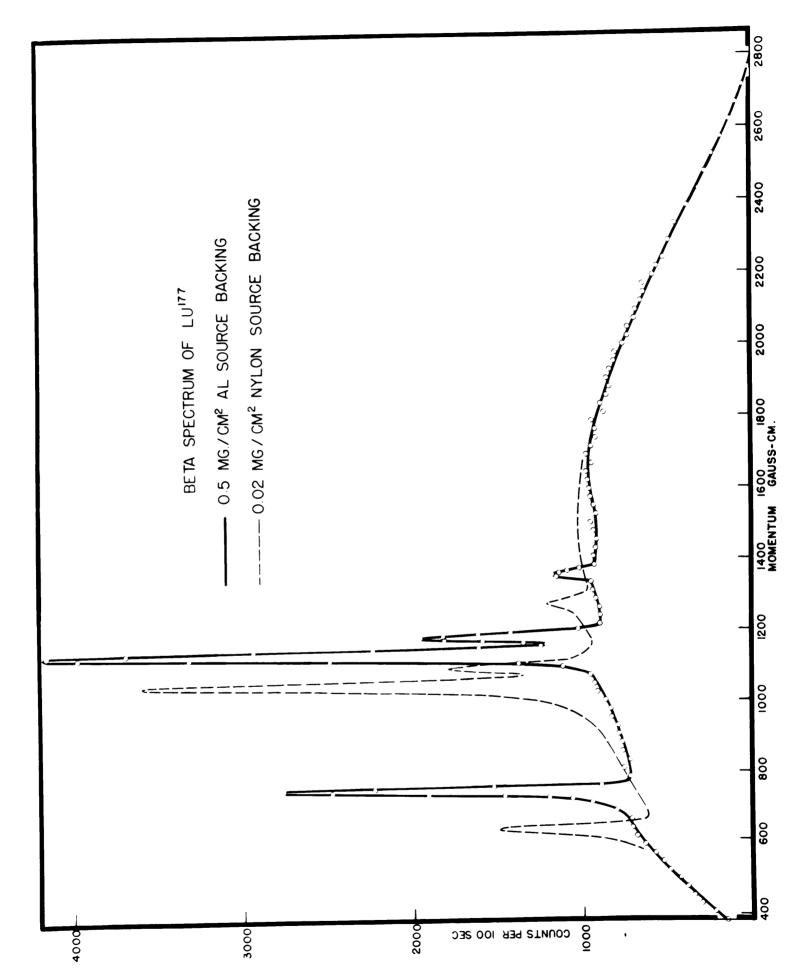
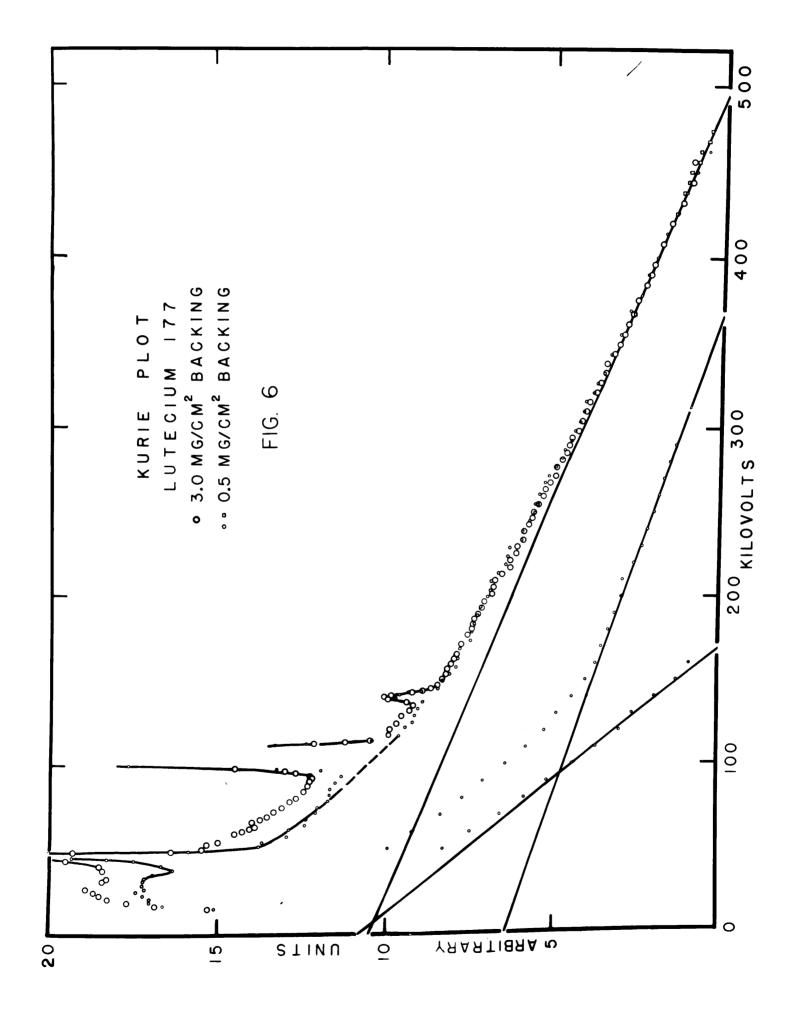
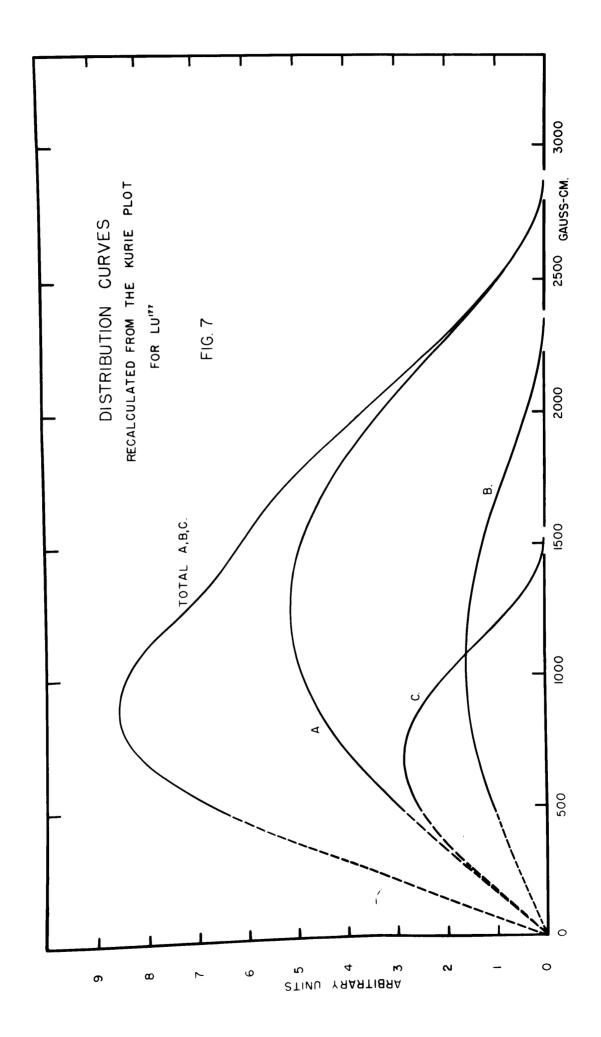
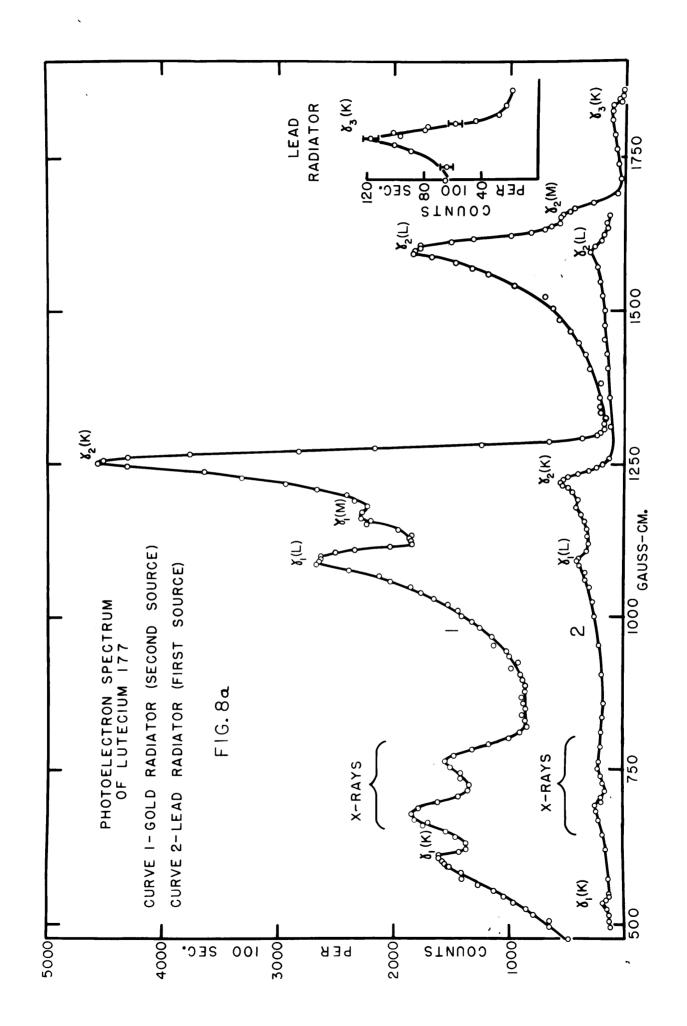
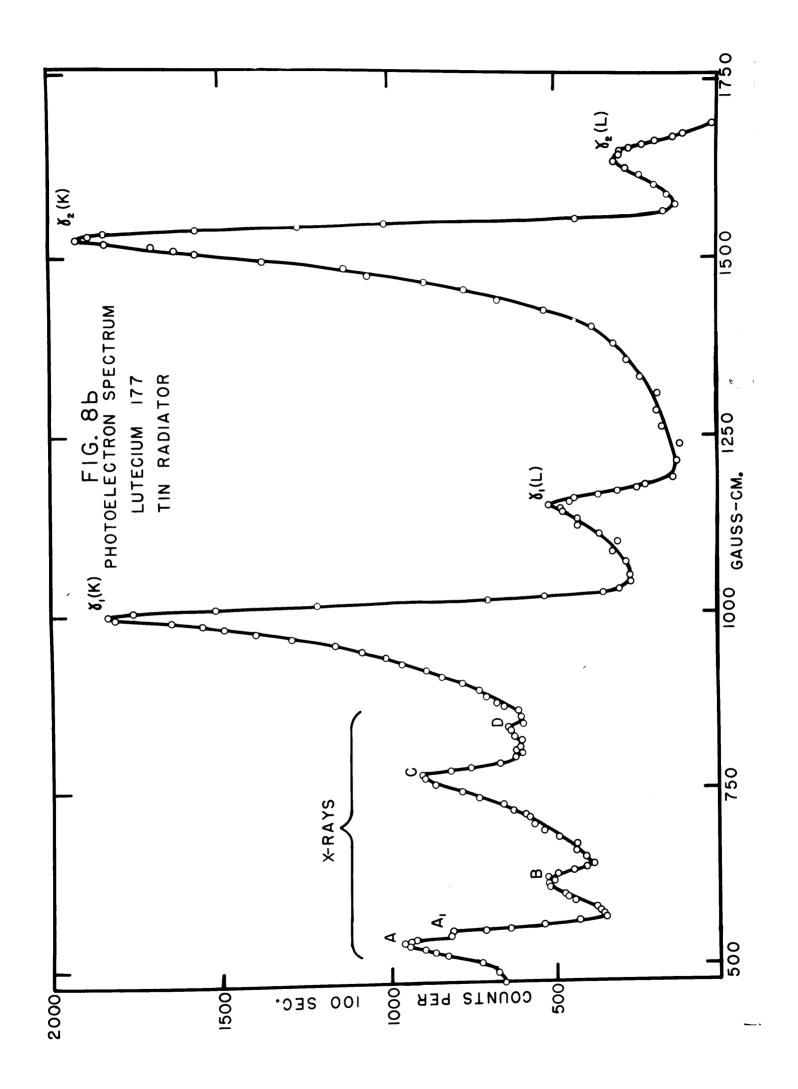


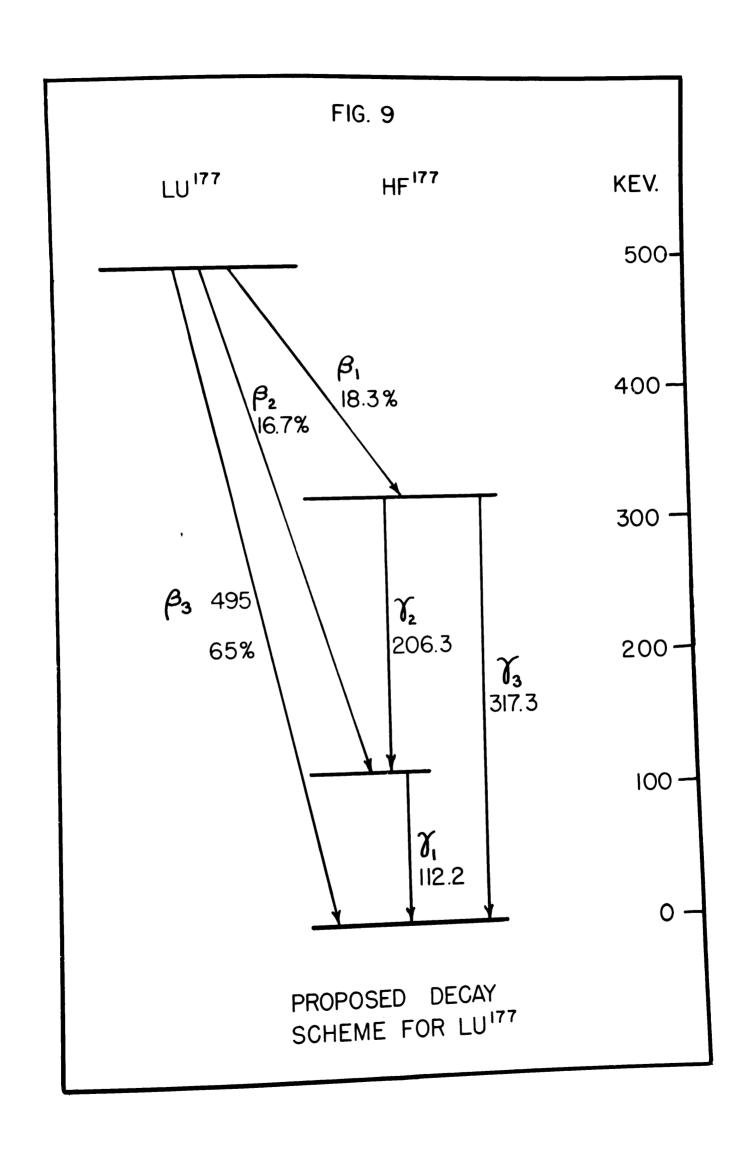
FIG. 5

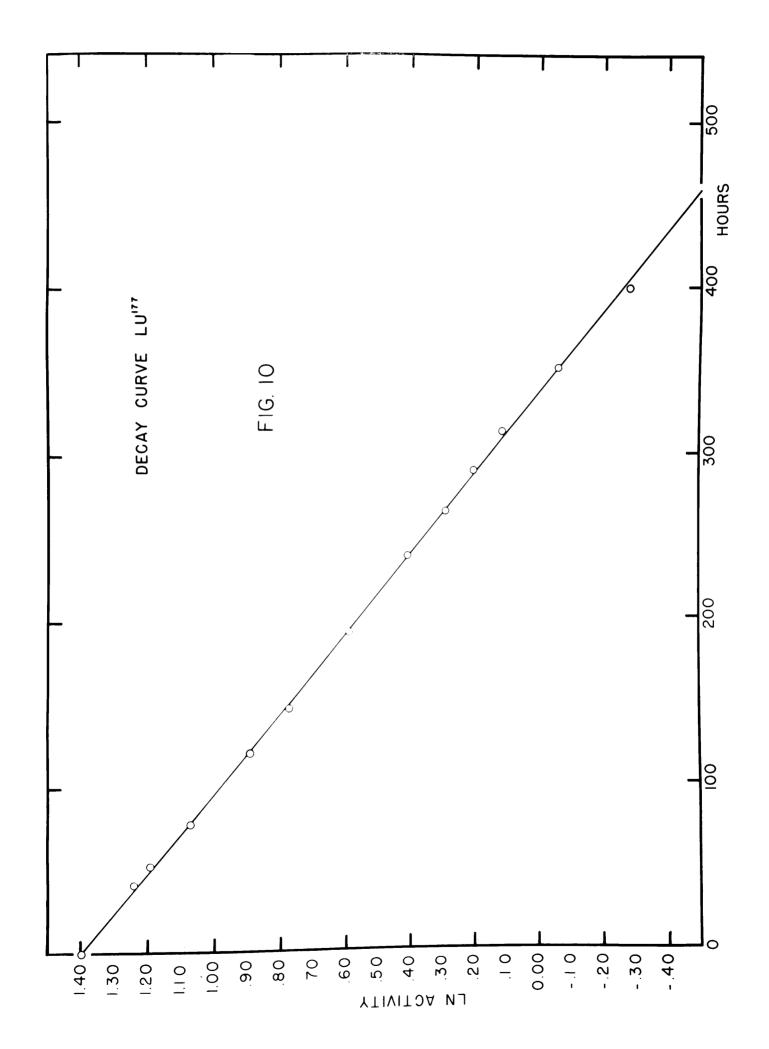












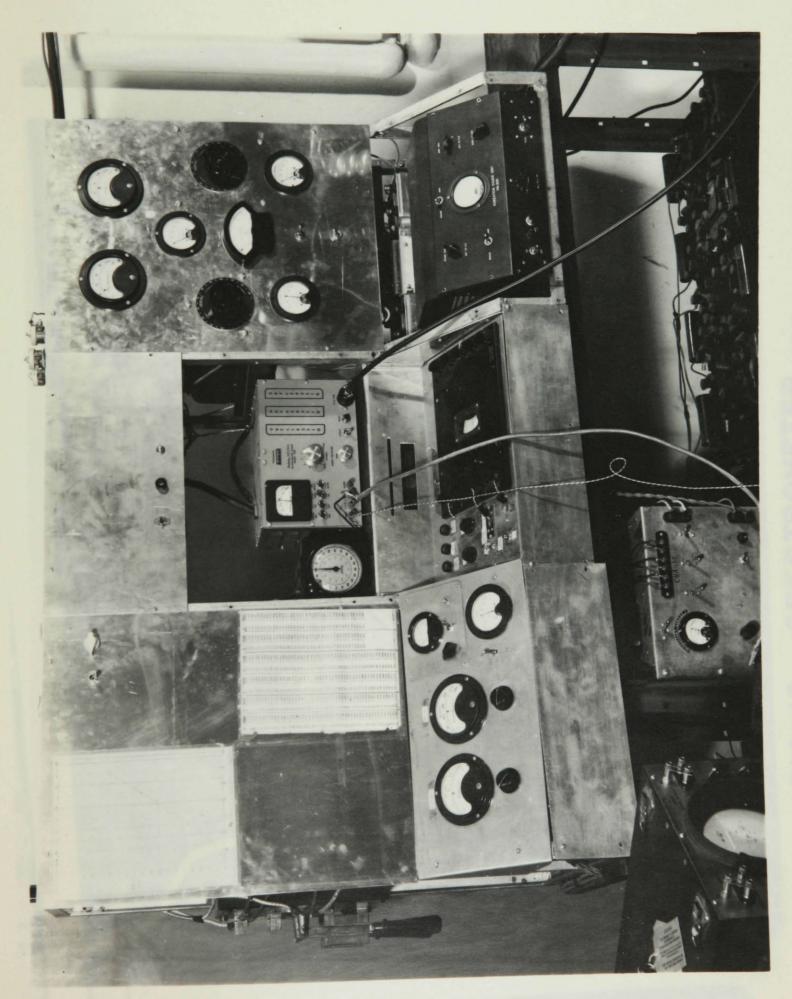


FIG. 12 SPECTROMETER

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UNACC.

