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METALLURGY AND CERAMICS

NONDESTRUCTIVE MEASUREMENT OF THE URANIUM CONTENT OF URANIUM-ALUMINUM ALLOYS

by

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ABSTRACT

An instrument is described which measures the uranium content of a 0.080-inch thick plate of uranium-aluminum alloy. The content is measured by gamma-ray transmission to an accuracy of ± 0.0006 gram per square centimeter over the range of 0.085 to 0.115 gram per square centimeter of contained uranium.

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NONDESTRUCTIVE MEASUREMENT OF THE URANIUM CONTENT OF URANIUM-ALUMINUM ALLOYS

INTRODUCTION

A nondestructive method for the measurement of uranium content in uranium-aluminum alloys was required. The alloy was 0.080 inch thick and was clad on both sides with 0.030 inch of aluminum. The uranium content was on the order of 0.100 gram/cm^2. This report describes the development of a suitable instrument.

SUMMARY

An instrument was developed that utilizes gamma-ray transmission to determine the amount of uranium in a uranium-aluminum alloy which is 0.080 inch thick. The extent to which the alloy attenuates the gamma radiation from an americium-241 source is determined with a scintillation detector. The attenuation is nearly a linear function of the uranium content between 0.085 and 0.115 gram/cm^2. The instrument measures the average uranium content of an area of 0.72 cm^2 to an accuracy of ± 0.0006 gram/cm^2, in less than one second.

DISCUSSION

A nondestructive method was needed to measure the uranium content of uranium-aluminum alloy. The alloy contains nominally 0.100 gram/cm^2 of uranium. Gamma-ray transmission was considered to be the most applicable technique.

In choosing a source of gamma radiation, several factors need to be taken into consideration; namely, the optimum quantum energy for maximum sensitivity, the specific activity of the source, the half-life of the source, the absence of undesirable quantum energies, cost, and availability.

The first consideration was to determine a gamma energy that would optimize the sensitivity of the instrument to variations in uranium content.

The transmission of monoenergetic radiation is given by the equation

$$I = I_o e^{-\mu X} \qquad (1)$$

where

I is the transmitted intensity

I_o is the initial intensity

μ is the linear absorption coefficient in cm^{-1}

X is the sample thickness in cm

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The usual transmission analysis by optical or X-ray methods involves the use of a radiation source with such a large output of photons that only a small percentage of the initial intensity need be transmitted to be detected. In this situation, an energy of radiation is chosen that gives a large percentage change in transmitted intensity per unit change in sample thickness. Relative sensitivity is defined as the fractional change in transmitted intensity per unit change of thickness, and from Equation 1 is written

$$\frac{\partial I/I}{\partial X} = \mu \qquad (2)$$

This equation shows that the best relative sensitivity can be obtained by making μ as large as possible, which can be achieved by selecting a source with a quantum energy such that μ is near an absorption edge for the material of concern.

In the case of transmission analysis by gamma rays, however, the photon output of the source is much smaller than in the case of X-rays so that the number of photons that can be expended is limited. In general, the previous sensitivity criterion no longer is applicable. For the present problem it was planned to measure the transmitted intensity with a scintillation counter, modified to give a current rather than a pulse output, to "buck out" part of the current with a battery, and to present the resulting current on a micro-microammeter. It was desirable, therefore, that the absolute change in transmitted intensity, as detected by the modified scintillation counter, be as large as possible for a given change in the uranium content of the uranium-aluminum alloy. The absolute sensitivity can be defined as $\frac{\partial I}{\partial X}$, and from Equation 1 it follows that

$$\frac{\partial I}{\partial X} = -\mu I_o e^{-\mu X} \qquad (3)$$

By differentiating this equation with respect to μ and equating to zero, Berman and Harris[1] find for the maximum absolute sensitivity,

$$\mu X = 1 \qquad (4)$$

For the case of approximately 0.100 gram per square centimeter of contained uranium, the optimum absorption coefficient corresponds to a photon energy of 56 Kev. Americium-241 was chosen as a suitable gamma source. Its principal gamma radiation is emitted at 59.7 Kev. Americium-241 is also attractive in other respects. It has a half-life of 470 years and an activity of approximately 3.3 curies per gram[3].

DESCRIPTION OF THE INSTRUMENT

A schematic diagram of the instrument is shown in Figure 1. The 0.15-curie americium source is mounted in an aluminum capsule that has a thin window, 0.010 inch thick. The transmitted gamma radiation is converted into light pulses by a thallium-activated sodium-iodide crystal, 2 mm thick. These light pulses are detected

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and integrated by a phototube whose d.c. output current is proportional to the transmitted radiation. The current is measured with a modified Beckman Model V micromicroammeter. This detector system permits the use of a source producing a count rate greater than 10^7 counts per second, with the resulting advantages of fast meter response and low statistical error.

The Phototube

The detector is a converted 6292 photomultiplier. The tube is connected as a simple phototube, using only the cathode and first dynode. The magnitude of the output current of the phototube is of the order of 10^{-10} ampere. For such low values of current, leakage at the base of the phototube is troublesome. The leakage was reduced by eliminating the base of the tube and by cleaning the exposed glass surface around the leads. The crystal was mounted on the tube with "Dow Corning 200" fluid of 10^6 cs. viscosity. The phototube and crystal assembly were mounted in a brass framework (Figure 2); then the glass surface around the leads was cleaned again. The entire assembly was mounted inside a light-tight steel cylinder (Figure 3). Connections were made to pins 14 and 1 of the tube, the cathode and the first dynode respectively, with a triaxial cable.

The Beckman Micromicroammeter

The small d.c. output current of the phototube is measured with a Beckman micromicroammeter, Model V. Since it is only necessary to measure small changes in uranium concentration about a standard value, a part of the phototube current is balanced out by placing a bucking voltage in the feedback loop of the Beckman (Figure 4). This permits the Beckman to be operated on a lower current range so that changes in the differential current may be read with greater accuracy. The bucking voltage may be adjusted to compensate for long-term drifts, if required. No short-term drift was experienced.

A cam, mounted on the shaft of the range-selector switch, actuates a microswitch which connects the bucking voltage in the circuit on the 10×10^{-11} ampere range and removes it on all other ranges. This prevents damage to the meter when the range selector is turned from the 10×10^{-11} ampere position to the 3×10^{-9} ampere position while changing samples.

PERFORMANCE OF THE INSTRUMENT

It was not possible to obtain a series of samples with known values of uranium content to evaluate the instrument. However, since the transmission of a sample is a function only of μX, it was possible to produce equivalent changes by varying the thickness X of a sample of known uranium content. This procedure involved the following steps: (1) Values of μX were calculated as a function of

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uranium content for an alloy sample of constant thickness. (2) Values of μX were calculated as a function of sample thickness for an alloy of known uranium content. (3) Instrument readings were taken as the sample thickness was varied by machining away successive increments. It was then possible to obtain a graph of instrument reading as a function of uranium content, since the thickness variations could be converted to an equivalent variation in uranium content. Details of this procedure are given in the Appendix.

A plot of Beckman reading vs. uranium content is shown in Figure 6. The graph indicates an instrument sensitivity of 0.80 division per 0.001 gram/cm² change in uranium content.

The required measurements are made by moving the source collimator assembly and detector along the sample while maintaining the alignment of the two units. Any extraneous movement causing a change in geometry between collimator and detector produces a change in detector current. With a 0.360-inch aperture in the lead collimator and a 1.5-inch diameter sodium-iodide crystal, the collimator detector geometry is much more critical in the axial direction than in the transverse direction. A movement of ± 0.010 inch in the axial direction gives a change in the Beckman reading of ± 0.2 division. This amount of movement can be tolerated and, coupled with ± 0.25 division of circuit "noise", allows an accuracy of ± 0.45 division or ± 0.0006 gram/cm².

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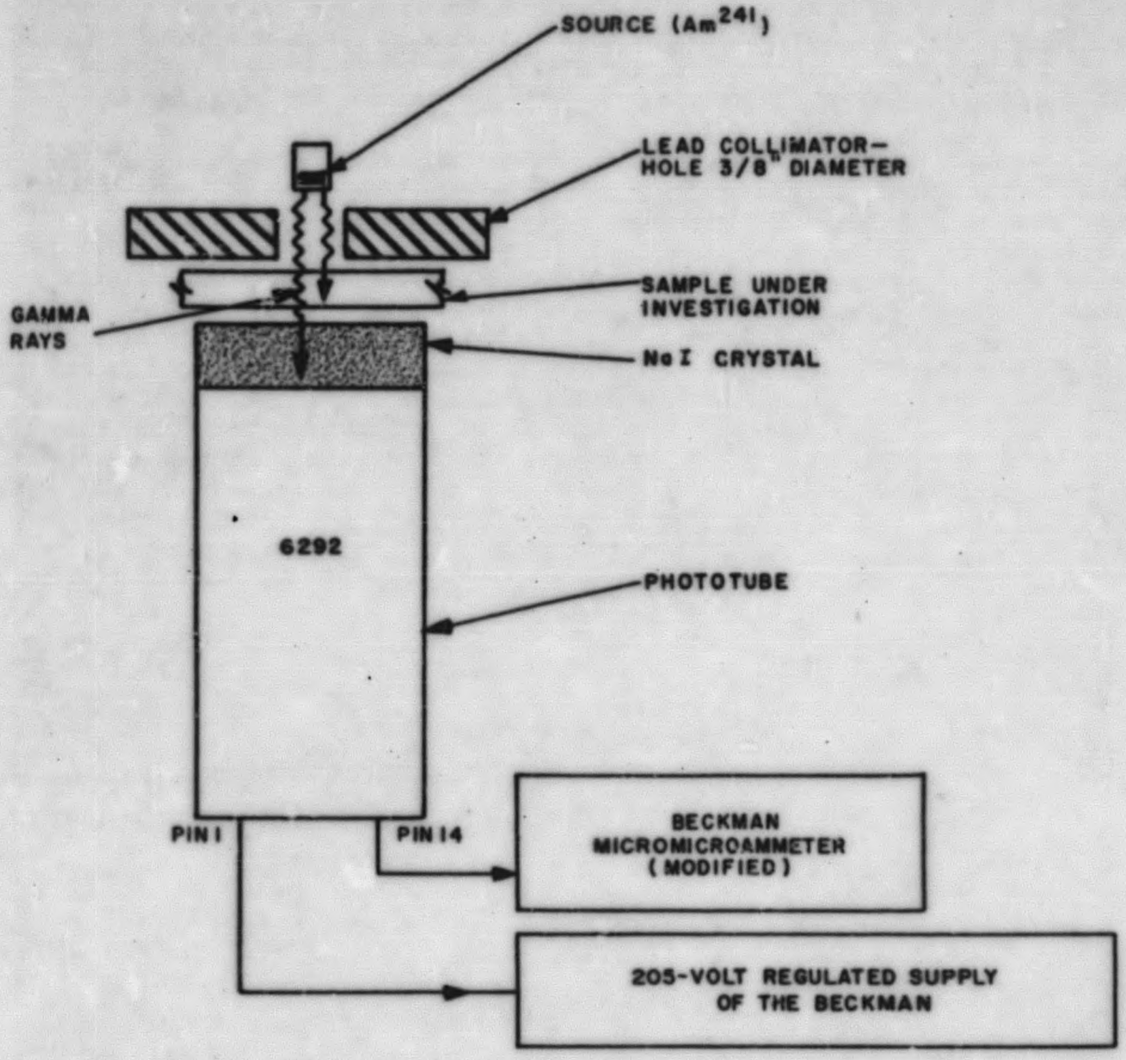
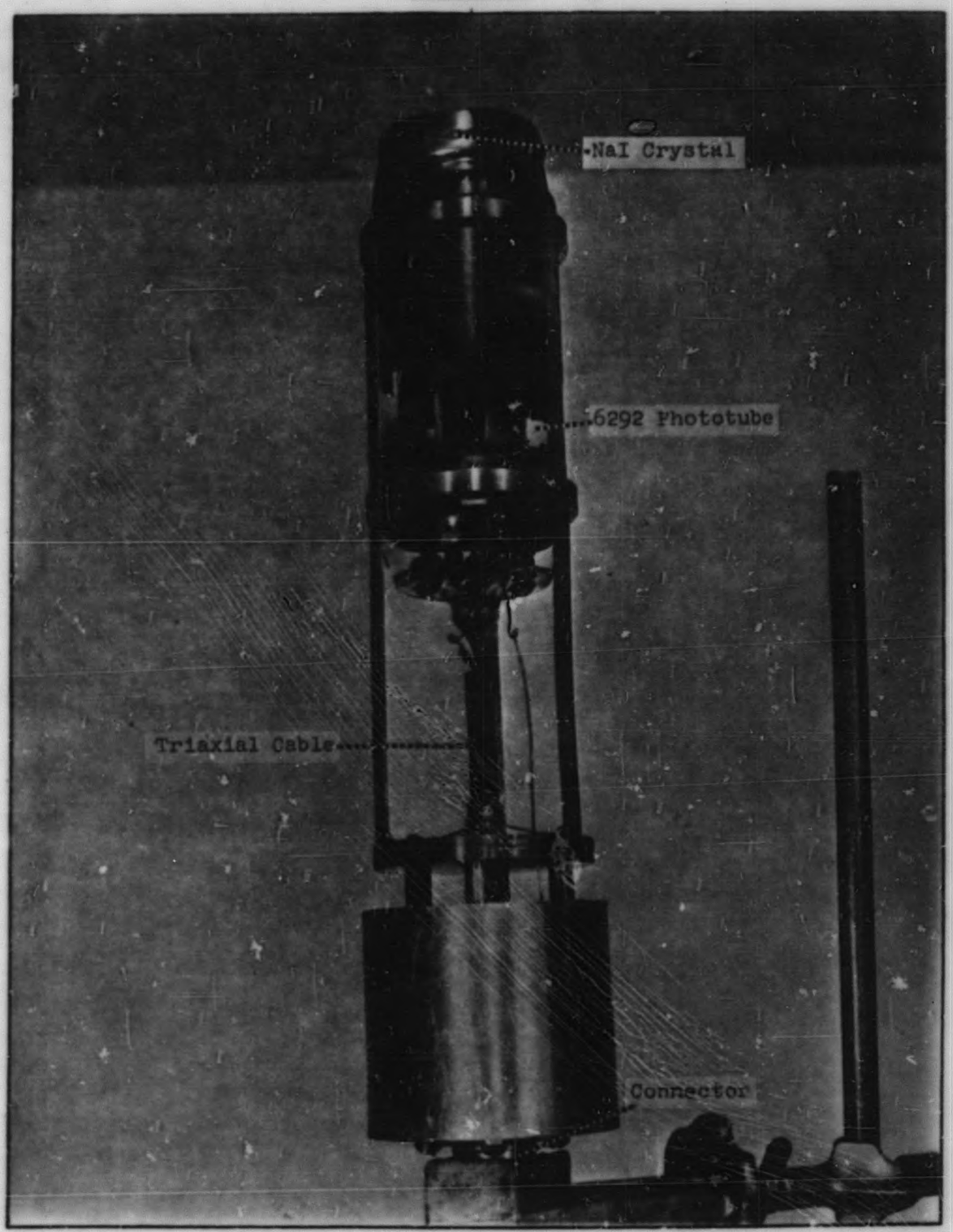
FIGURE 1



SCHEMATIC VIEW OF THE SOURCE-DETECTOR ARRANGEMENT

FIGURE 2



MOUNTING OF PHOTOTUBE AND CRYSTAL

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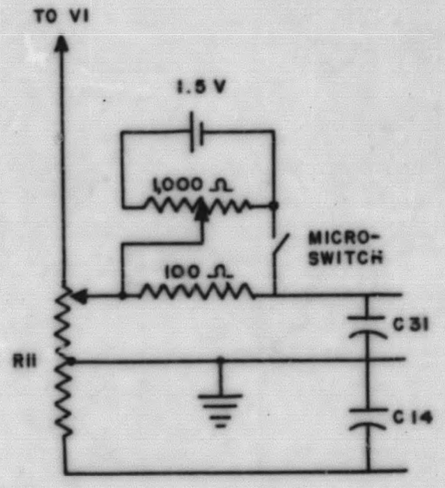
FIGURE 3

THE INSTRUMENT

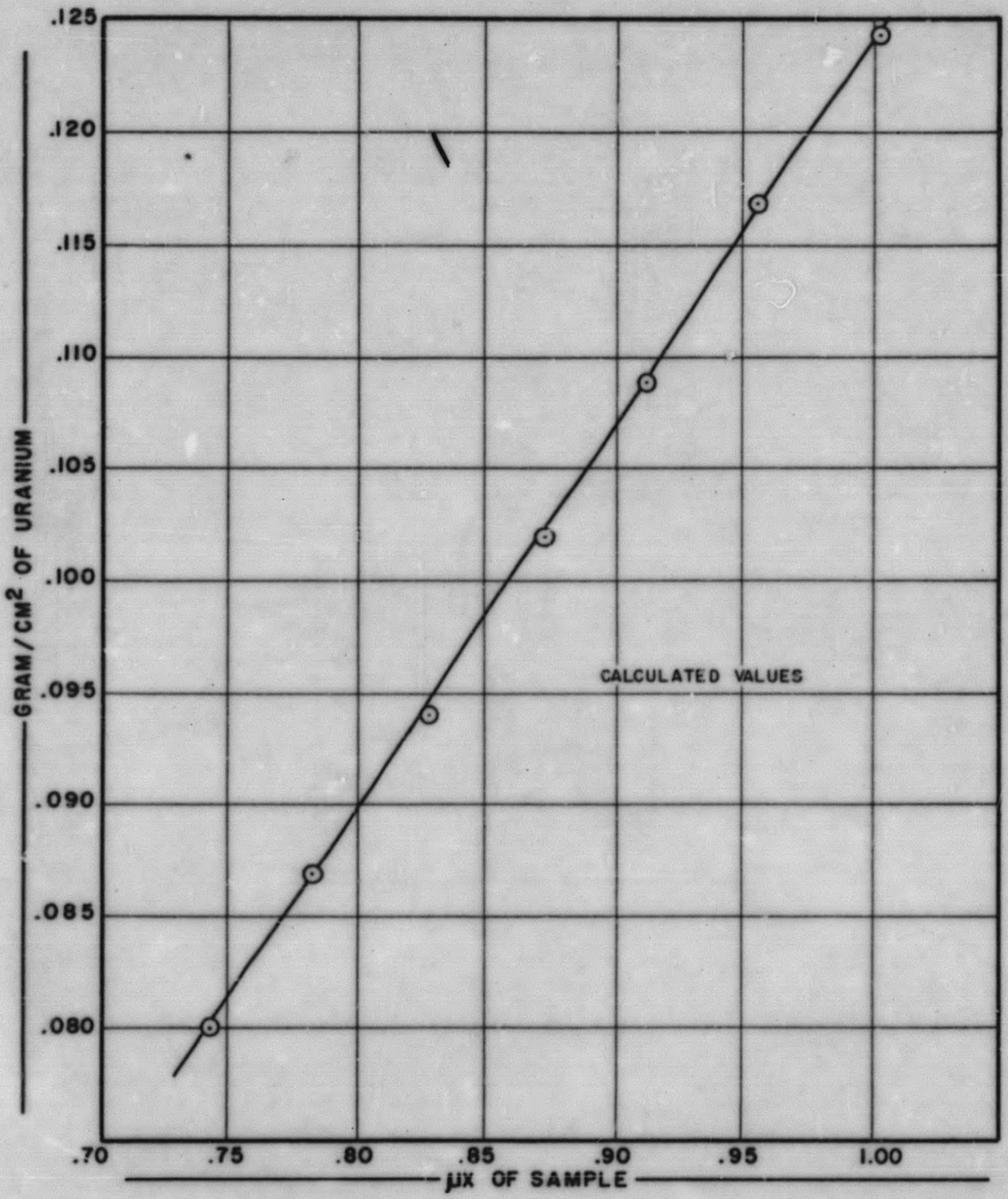


NOTE: V1, R11, C31, C14, REFER TO COMPONENTS OF THE CIRCUIT SUPPLIED BY THE MANUFACTURER.

FIGURE 4

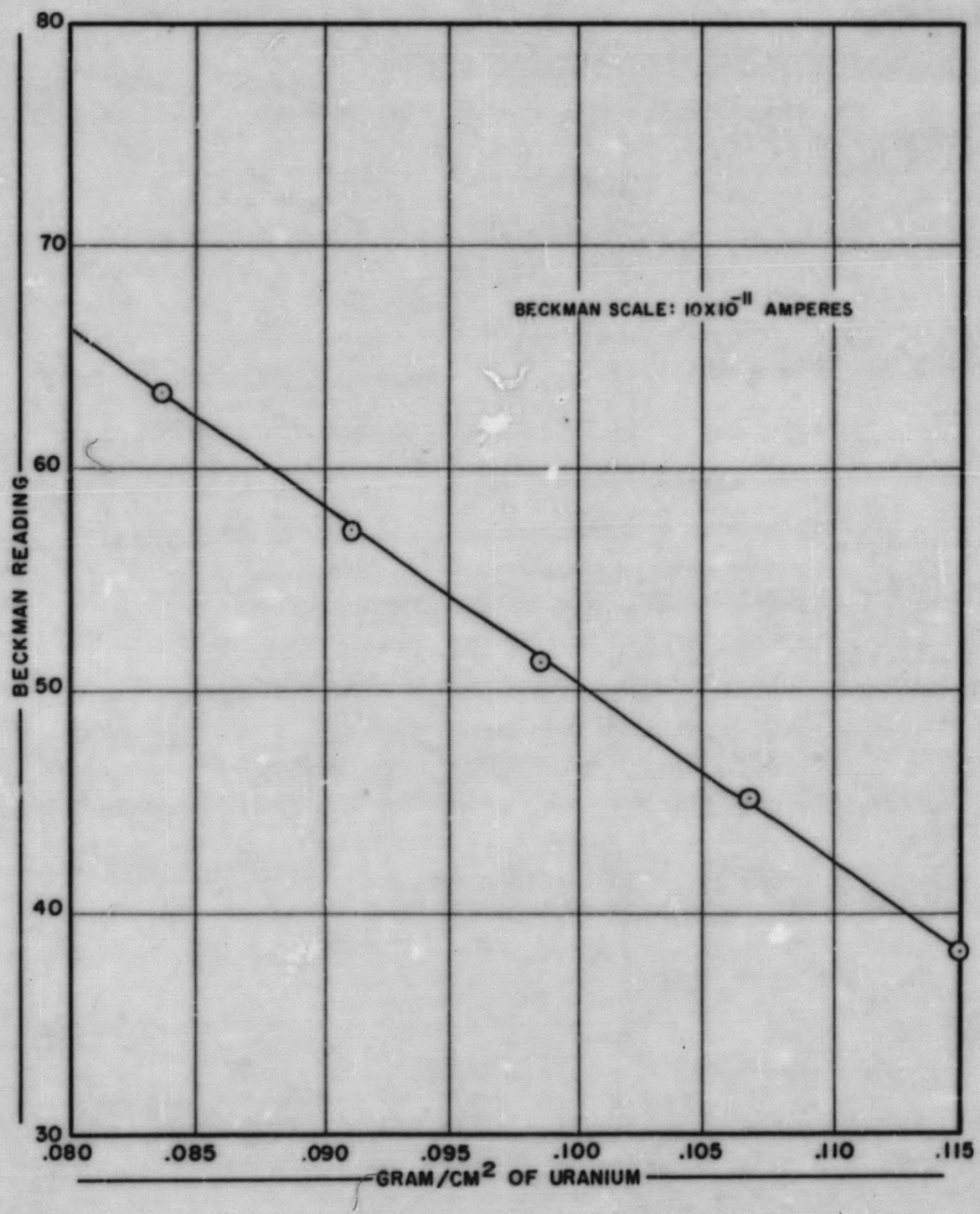
BUCKING VOLTAGE CIRCUIT
(MODIFICATIONS MADE TO THE MODEL V MICROMICROAMMETER)

FIGURE 5



GRAPH OF GRAMS/CM^2 OF U vs. μX OF SAMPLE

FIGURE 6



GRAPH OF BECKMAN READING vs. GRAMS/CM² OF URANIUM

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APPENDIX

CALIBRATION

Calculation of μX vs. Uranium Contents

The transmission of monoenergetic radiation is given by the equation

$$I = I_o e^{-\mu X}$$

The exponent μX for a clad alloy sample is given by

$$\mu X = \mu_{alloy}\, t_a + \mu_{cladding}\, t_c$$

where t_a and t_c are the thickness of the alloy and cladding respectively. The quantities μ_{alloy} and $\mu_{cladding}$ may be written as

$$\mu_{alloy} = \mu_{Al} + \mu_U = \mu_{m(Al)}\rho_{Al(a)} + \mu_{m(U)}\rho_{U(a)}$$

$$\mu_{cladding} = \mu_{m(Al)}\rho_{Al}$$

where $\mu_{m(Al)}$ is the mass absorption coefficient of aluminum
$\mu_{m(U)}$ is the mass absorption coefficient of uranium
$\rho_{Al(a)}$ is the density of aluminum in the alloy
$\rho_{U(a)}$ is the density of uranium in the alloy
ρ_{Al} is the density of aluminum = 2.70 grams/cm³

For 59.7-Kev gamma radiation the quantities $\mu_{m(Al)}$ and $\mu_{m(U)}$ are 0.281 and 5.96 cm²/grams, respectively. The quantities $\rho_{Al(a)}$ and $\rho_{U(a)}$ may be calculated to a good approximation from the equations

$$\rho_{Al(a)} = \frac{\% \text{ Al by weight}}{\frac{\% \text{ U by weight}}{\rho_U} + \frac{\% \text{ Al by weight}}{\rho_{Al}}} = \frac{\% \text{ Al by weight}}{D}$$

$$\rho_{U(a)} = \frac{\% \text{ U by weight}}{D}$$

where

ρ_U = density of U = 18.7 grams/cm³

From these values, μ_{alloy} and $\mu_{cladding}$ may be calculated. The values of t_a and t_c are 0.203 cm and 0.152 cm, respectively. The table below was obtained for various uranium concentrations, $\rho_{U(a)}t_a$, by substituting the appropriate values in the equation for μX.

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% U by Weight	$\rho_{U(a)}t_a$	μX
13	0.080	0.744
14	0.087	0.784
15	0.094	0.828
16	0.102	0.872
17	0.109	0.914
18	0.117	0.959
19	0.124	1.00

From the above table, a plot of μX vs $\rho_{U(a)}t_a$ was made as shown in Figure 5.

Calculation of μX vs. Sample Thickness

Initially, an unclad alloy sample used in calibrating the instrument contained 0.128 gram/cm^2 of uranium. The μ_{alloy} of the sample was calculated to be 3.74. Using this value of μ and various values of sample thickness, the calculated values of μX in the following table were obtained.

Sample Thickness (cm)	μX
0.254	0.951
0.241	0.902
0.228	0.853
0.216	0.808
0.204	0.764

Instrument Reading vs. Sample Thickness

The alloy sample was machined down in the successive steps given in the table. After each of the machining operations, the sample was placed between the source and detector and the Beckman reading was recorded. The results are included in the table below.

Sample Thickness (cm)	Beckman Reading
0.254	38.4
0.241	45.2
0.228	51.2
0.216	57.3
0.204	63.4

For the μX values in the table, corresponding values of $\rho_{U(a)}t_a$ were found from Figure 5 and plotted against the Beckman reading. This plot, a graph of Beckman reading as a function of uranium content, is shown in Figure 6. The graph shows that the instrument has a sensitivity of approximately 0.80 division per 0.001 gram/cm^2 change in uranium concentration. The fluctuation in meter reading due to "noise" corresponds to about ± 0.0004 gram/cm^2.

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