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DOSE RATES OF RADIATION FROM THORIUM
AND FROM ENRICHED URANIUM

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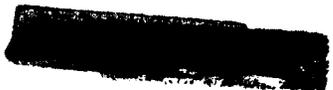
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Per letter instructions of

T. E. Bortner

and

H. K. Richards

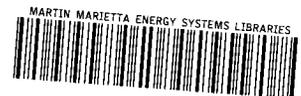
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Physics of Nuclear Radiation
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DOSE RATES OF RADIATION FROM THORIUM AND FROM
ENRICHED URANIUM

Abstract

Dose rates at a plane surface of a thick disc of thorium and of enriched uranium metal were measured with the extrapolation chamber. Using a carbon collector, the total dose rates were found to be, for thorium 7.2 esu/hr/cm^3 , and for the enriched uranium $1413.5 \text{ esu/hr/cm}^3$. The residual radiation (principally beta) with the alpha filtered out was found to be $0.096 \text{ esu/hr/cm}^3$ for thorium, and $0.208 \text{ esu/hr/cm}^3$ for the enriched uranium.

The total dose rate measured in rep/hr, of this sample of enriched uranium is 98.16 times that of natural uranium*. Calculations by H. K. Richards gave a ratio of 103.5 for enriched to natural uranium.

* ORNL-740, "Dose Rates of Radiation from Natural Uranium", T. E. Bortner

EXPERIMENTAL DETERMINATION OF DOSE RATES
FROM THORIUM AND FROM ENRICHED URANIUM

T. E. Bortner

Instrumentation

The extrapolation chambers used in this work are essentially the same as described by Sheppard and Abele, ORNL-265. Two chambers were used throughout. They are identical except for the area of the collector. In one this area is 3.1416 cm² and in the other it is 5.000 cm². There was no significant difference in the final results obtained with the two chambers.

A reed electrometer, built by the Applied Physics Corporation, was used as a null instrument with the output indicated on a Brown recorder. Condensers manufactured by John E. Fast Company were used to insure steady operation of the recorder; and resistors of 9.61×10^9 ohms and 991.5×10^9 ohms, manufactured by the Victoreen Instrument Company, and calibrated by the National Bureau of Standards, were used in the electrometer circuit. The dose rate, measured in esu/hr/cm³, was computed from the compensating emf, measured on a potentiometer, using the following formula:

$$\text{esu/hr/cm}^3 = 3 \times 10^{13} \times \frac{TE}{pRV};$$

where,

p = barometric pressure (mm of Hg)

T = absolute temperature (°K)

E = compensating emf (millivolts)

R = output resistance (ohms), and

V = collecting volume of air (cm³)

Methods and Results

Dose rates at plane surfaces of a thick disc of thorium metal and of enriched uranium metal were measured. The two discs were geometrically identical. Each was one-eighth inch thick and three inches in diameter, with the surface hand-lapped to a tolerance of 0.0001 inches. The following mass spectrographic assay was obtained for the enriched uranium:

U ²³⁴	1.08%
U ²³⁵	27.46%
U ²³⁸	71.46%

Nothing is known of the history of the thorium other than that it had been separated about two years previously.

In measuring the total surface dose rate, the source of radiation was used as an electrode of the chamber. The measurement of the dose rate due to radiation other than alpha was accomplished by filtering out the alpha radiation with a thin sheet of polystyrene coated with aquadag to make it an electrode. The surface density of the polystyrene used was 6.9 mg/cm². This thickness of polystyrene was used because preliminary examination made with a sensitive Poppy indicated that this weight of absorber would exclude all alpha radiation. Calculations pointed to a thinner absorber, but microscopic holes or other irregularities of thickness made greater weight seem advisable. The dose rate due to beta radiation was not measured separately from that due to gamma radiation, but it is known that the contribution of gamma radiation is relatively small.

Cyclical reversal of collecting voltage was used in all measurements to compensate for charge carried by radiation particles. Results given are averages

of the values thus obtained. For the measurement of the beta-gamma dose rate, two different extrapolation chambers were used with collecting electrodes of carbon. All measurements made with these chambers agreed within one-half of one percent.

Measured values of electrometer potentials resulting from total radiation are plotted against electrode separations for the enriched uranium in Figure 1, and for thorium in Figure 2. It will be observed that in the range of separations used, the experimental points fall on a straight line of which the slope corresponds to the total radiation. Figure 1 shows the total dose rate for the enriched uranium to be $1413.5 \text{ esu/hr/cm}^3$, and Figure 2 shows the total dose rate for thorium to be 7.2 esu/hr/cm^3 . Similar information on the beta-gamma radiations is given in Figures 3 and 4 where the dose rate from the enriched uranium is shown to be $0.208 \text{ esu/hr/cm}^3$, and that for thorium is $0.096 \text{ esu/hr/cm}^3$.

Dose rates in tissue, measured in reps per hour, corresponding to the above dose rates in air may be computed using relationships given by K. Z. Morgan: "The Use of the Roentgen Equivalent Physical", ORNL-783.

CALCULATION OF RELATIVE DOSE RATE FOR ANY ISOTOPIC
COMPOSITION OF URANIUM

H. K. Richards

It is desirable to calculate the alpha dose for any isotopic composition of uranium. When the composition and dose of natural uranium is known, this can be done with an approximation sufficient for health physics requirements.

To simplify the calculations, the constants for range, energy, decay, and ionization of U^{238} are taken as unity, and those of the other isotopes as relative values. The ionization produced by the alphas of each isotope will depend on:

- a. The maximum depth in the uranium sample from which the isotope contributes alpha particles.
- b. The average ionization produced by an alpha particle of the isotope.
- c. The concentration of the isotope.
- d. The decay constant of the isotope.

Since only relative values are needed, it is sufficient to know the penetration of the different alpha particles in uranium, setting that for U^{238} as one.

The penetration in air is given by:

$$(1) R \sim E^{3/2} \sim v^3$$

For solid materials the penetration L is given approximately by the

Bragg-Kleeman rule:

$$(2) L = 0.0003 \frac{R A^{1/2}}{\rho}$$

where

R = range in air

ρ = density of uranium for which variation with isotopic composition is neglected

v = velocity of the particle

A = atomic weight

Therefore, the composition of the penetrating particles will have the same relative velocity distribution as in air since A and ρ are constant.

The alpha energies, E , for the isotopes are given below, together with their ranges in air:

	<u>Mev</u>		<u>Range in Air</u>
(3) U^{238}	$E = 4.18$		$R_1 = 2.67$ cm
U^{235}	$E = 4.39$ "	} 80%	$R_{21} = 2.80$ "
	$E = 4.56$ "		} 20%
U^{234}	$E = 4.76$ "		$R_3 = 3.18$ "
			$R_2 = 2.84$ "

where R_2 is the average range for a particle from U^{235} . The indices 1, 2, 3, shall always refer to U^{238} , U^{235} , and U^{234} respectively.

Changing to relative values, dividing by the range for U^{238} , the ranges become:

$$(4) R_1 = 1.000$$

$$R_2 = 1.063$$

$$R_3 = 1.191$$

The alpha spectrum of the surface radiation for each isotope will consist of particles with an energy distribution ranging from zero to its maximum energy. The ionization produced by an alpha particle per cm of path is given by the approximate formula,

$$(5) \quad i = C(R-X)^{-1/3}$$

where R is the range of the particle at any point of its path and X is the distance measured from that point.

In order to compare the alpha ionization produced inside of the ionization chamber by different isotopes, it is required to know the ratio of their average ionization powers after leaving the uranium surface. Since the distance between the uranium alpha emitter and the collector was much less than 1 mm, it can be assumed that the average path is in the order of 1 mm; i.e., small compared with the remainder of the path length of the major number of alpha particles.

(5a) becomes, therefore, with good approximation inside of the chamber:

$$(5b) \quad i = CR^{-1/3}$$

The average ionization for the isotope of U^e is expressed (see Appendix) by $\bar{i}_e = 2 CR_e^{-1/3}$, or comparing two isotopes: $\frac{\bar{i}_e}{\bar{i}_m} = \frac{R_{em}^{-1/3}}{R_{mm}^{-1/3}} = \frac{i_{em}}{i_{mm}}$.

The ratio of the average alpha ionization/cm after leaving the uranium surface is equal to that of the ionization per cm before passing through the absorbing material. Therefore, (5b) can be used to compare the alpha ionization/cm for the different isotopes. Setting again the ionization of U^{238} as one, the values are given by:

$$(6) I_1 = 1.00 \quad \text{for } U^{238}$$

$$\left. \begin{array}{l} I_{21} = 0.983 \\ I_{22} = 0.963 \end{array} \right\} \text{ for } U^{235}$$

$$I_3 = 0.943 \quad \text{for } U^{234}$$

or $I_2 = 0.977$ as an average for U^{235}
where I_{21} contributes 80%
and I_{22} 20%

The indices 1,2,3, shall always refer to the same isotopes as in (6); i.e., U^{238} , U^{235} , U^{234} .

The exact values of the composition of natural uranium is well enough known for U^{238} and U^{235} , and so are the half-life times for these isotopes. However, for U^{234} these figures vary considerably. Since the contribution of U^{234} is by far the most important one for the enriched uranium, the uncertainty of its half-life time will introduce the major error.

The following values have been published recently:

	<u>Half-life</u>	<u>Abundance</u>	
For U^{234}	2.32×10^5 yrs.	0.0052%	Friedlander-Kennedy Radiochemistry, 1949
	2.522×10^5 "	0.00548%	Kienberger, Phys. Rev. Vol. 67, 1945, p. 336-39
	2.67×10^5 "	0.0058%	Godwin, Knight, et al. Phys. Rev. Vol. 67, 1945, p. 336-39
	2.33×10^5 "		E. Baldinger, P. Huber Helv. Phys. Acta 22, 1949 p. 365-8
U^{235}	7.07×10^8 "	0.719%	Friedlander-Kennedy
	8.91×10^8 "		W. H. Sullivan
U^{238}	4.51×10^9 "	99.28%	W. H. Sullivan

There are also different values for U^{235} , but they will not introduce a major error.

For an approximate estimation, the results of C. A. Kienberger in report K-329 will be used where K_1, K_2, K_3 is the relative abundance for each isotope when $\sum_1^3 K_i = 1$.

$$(7) \quad \begin{array}{ll} U^{238} & K_1 = 0.993 \\ U^{235} & K_2 = 0.00719 \\ U^{234} & K_3 = 0.0000548 \end{array}$$

The relative decay constants are inversely proportional to the half-life times of the isotopes. The radioactive equilibrium between U^{238} and U^{234} requires the $\lambda_1 K_1 = \lambda_3 K_3$, thus setting $\lambda_1 = 1$ gives $\lambda_3 = 1.812 \times 10^4$. Since the decay constants are inversely proportional to the half-lives, using the Friedlander-Kennedy value for the half-life of U^{235} , and the Sullivan value for U^{238} gives $\lambda_2 = 6.38$ when $\lambda_1 = 1$.

Thus, the relative values for the decay constants are:

$$(8) \quad \begin{array}{l} \lambda_1 = 1 \\ \lambda_2 = 6.38 \\ \lambda_3 = 1.812 \times 10^4 \end{array}$$

The ionization current will be proportional to $R_i I_i \lambda_i K_i$ for each isotope and concentration. The total ionization current I_t will then be:

$$(9) \quad I_t = \sum_1^3 R_i I_i \lambda_i K_i; \text{ setting the } R_i I_i \lambda_i = A_i$$

$$A_1 = 1.000 \text{ for } U^{238}$$

$$A_2 = 6.63 \text{ for } U^{235}$$

$$A_3 = 2.035 \times 10^4 \text{ for } U^{234}$$

For natural uranium:

$$(10) \quad A_1K_1 = 0.993$$

$$A_2K_2 = 0.048$$

$$A_3K_3 = 1.115$$

$$I_t = 2.156 \text{ relative units}$$

For enriched uranium of composition:

$$(11) \quad K_1 (U^{238}) = 0.715$$

$$K_2 (U^{235}) = 0.275$$

$$K_3 (U^{234}) = 0.0108$$

$$A_1K_1 = 0.715$$

$$A_2K_2 = 1.82$$

$$A_3K_3 = 219.95$$

$$I_t = 222.49 \text{ relative units}$$

From (10) and (11) the dose ratio of the enriched to natural uranium should be 103.3. Comparison of the measured dose rate of 1413.5 esu/hr/cm³ for the enriched uranium with the measured dose rate of 14.4 esu/hr/cm³ for natural uranium, reported by T. E. Bortner in ORNL-740, gives an experimental ratio of 98.2. This difference of 5% between the calculated and experimental ratios is probably largely due to uncertainty in the value of the disintegration constant of U²³⁴ which is the principal contributor even in low concentrations. While the Bragg-Kleeman Rule, Equation (2), is only an approximation, the conditions under which it is applied here are such that no large error in the calculated ratio is to be expected from its use.

Appendix

Average ionization of alpha particles after penetration of uranium layer:

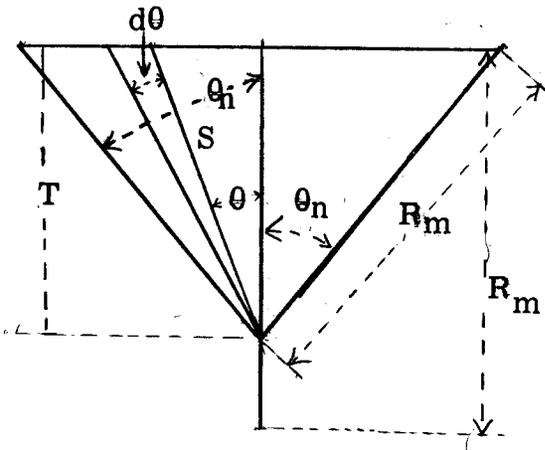


Diagram 1

Alpha Radiation of Uranium
Originating in Depth

R_m = maximum penetration depth
of alpha particles

$T = R_m \cos \theta_n$ = depth of alpha
source

$$dT = -R_m \sin \theta_n d\theta_n$$

S = path of alpha particles

$$S = R_m \frac{\cos \theta_n}{\cos \theta}$$

The ionization per cm per particle after leaving the uranium is given by

$$i = C(R-X)^{1/3}$$

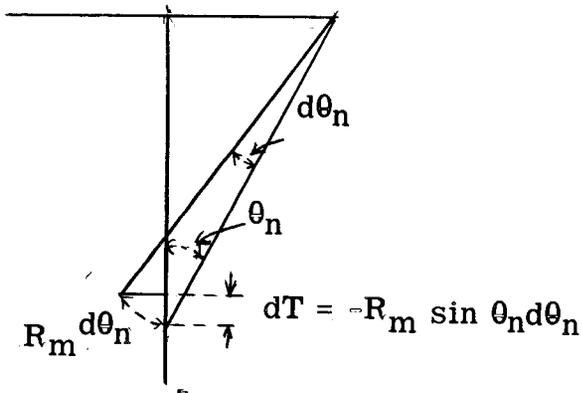


Diagram 2

If n is the number of alpha particles
originating per cm depth per cm^2 in
uranium, the number of produced at
depth T by an element dT is given by
 $n dT$. The number reaching the sur-
face from dT under an angle between

θ and $\theta + d\theta$ is given by:

$$n \frac{d\Omega dT}{4\pi} = \frac{n 2\pi \sin \theta d\theta}{4\pi} R_m \sin \theta_n d\theta_n =$$

$$dn(\theta, \theta_n)$$

These particles after leaving the uranium surface will produce in air an

ionization/cm of:

$$i(\theta, \theta_n) = \frac{n}{2} R_m C (R_m - R_m \frac{\sin \theta_n}{\sin \theta})^{1/3} \sin \theta_n \sin \theta \, d\theta \, d\theta_n$$

To find the average ionization per cm of path, the following integrations have to be made:

$$\bar{i} = \frac{\frac{n R_m C R_m^{-1/3}}{2} \int_0^{\theta_n} \int_0^{2\pi} \left(\frac{\cos \theta_n}{1 - \cos \theta} \right)^{-1/3} \sin \theta \sin \theta_n \, d\theta \, d\theta_n}{\frac{n R_m}{2} \int_0^{\theta_n} \int_0^{2\pi} \sin \theta \sin \theta_n \, d\theta \, d\theta_n}$$

The integral in the denominator is 1/2:

$$\bar{i} = 2 C R_m^{-1/3} \int_0^{\theta_n} \int_0^{2\pi} \left(1 - \frac{\cos \theta_n}{\cos \theta} \right)^{-1/3} \sin \theta \sin \theta_n \, d\theta \, d\theta_n = 2 i f(\theta, \theta_n)$$

The integral is independent of R_m and, therefore, identical for alpha particles of all isotopes.

If i_{em} and i_{mm} are the original ionizations per cm in air, and \bar{i}_e and \bar{i}_m the averages when leaving the uranium, the ratios of $\frac{\bar{i}_e}{\bar{i}_m}$ are given by:

$$\frac{\bar{i}_e}{\bar{i}_m} = \frac{R_{em}^{-1/3}}{R_{mm}^{-1/3}} = \frac{i_{em}}{i_{mm}}$$

Other relative quantities like alpha particle energy or the average depth contributing particles from each isotope can be calculated in the same way.

Since the dependence on θ or θ_n disappears for relative values, it can be shown easily that the average depth contributing particles at the surface is also a function of the maximum range R_m only, a result that has been applied to the previous calculations.

MEASUREMENT OF TOTAL RADIATION DOSE RATE
AT SURFACE OF ENRICHED URANIUM

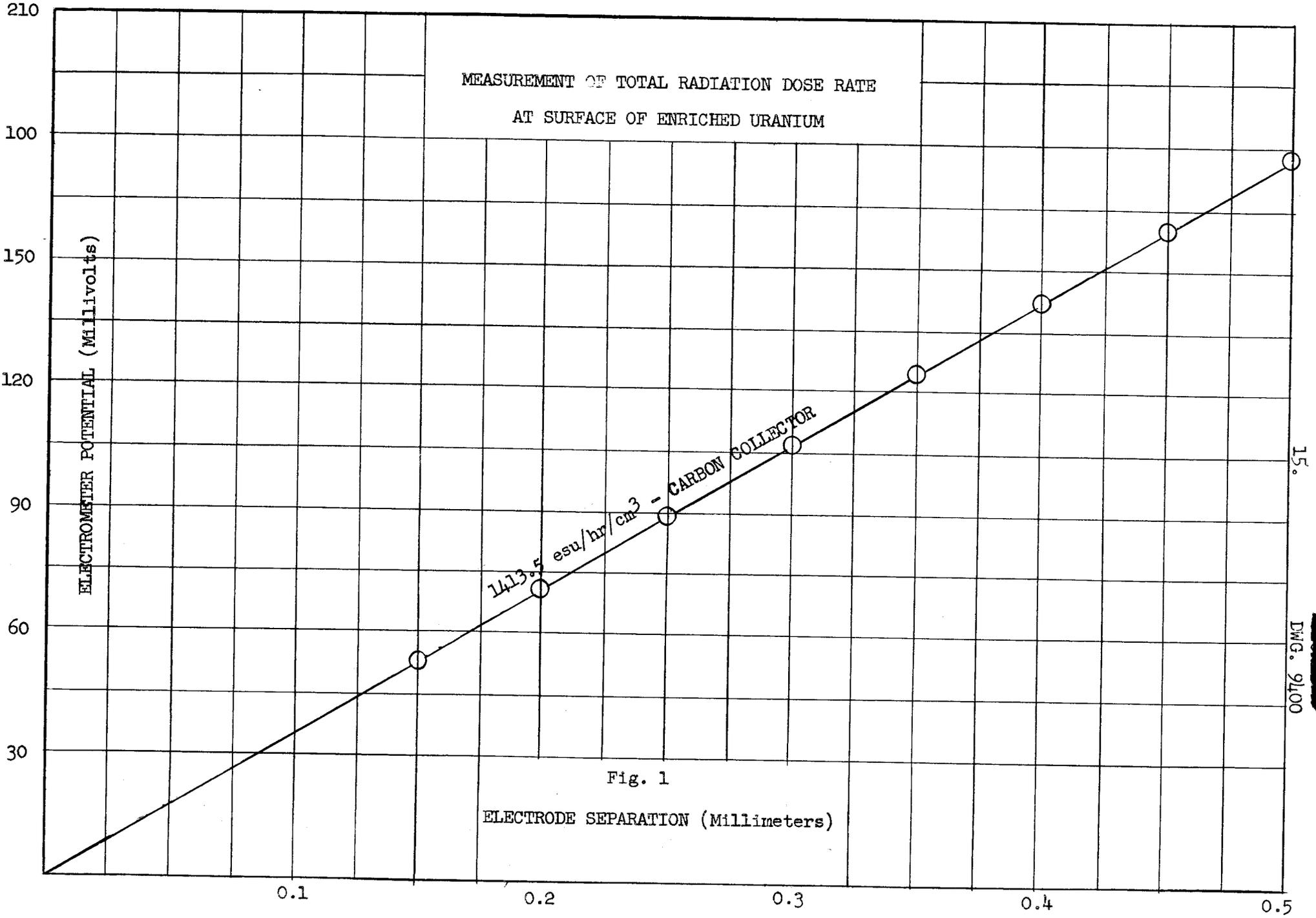
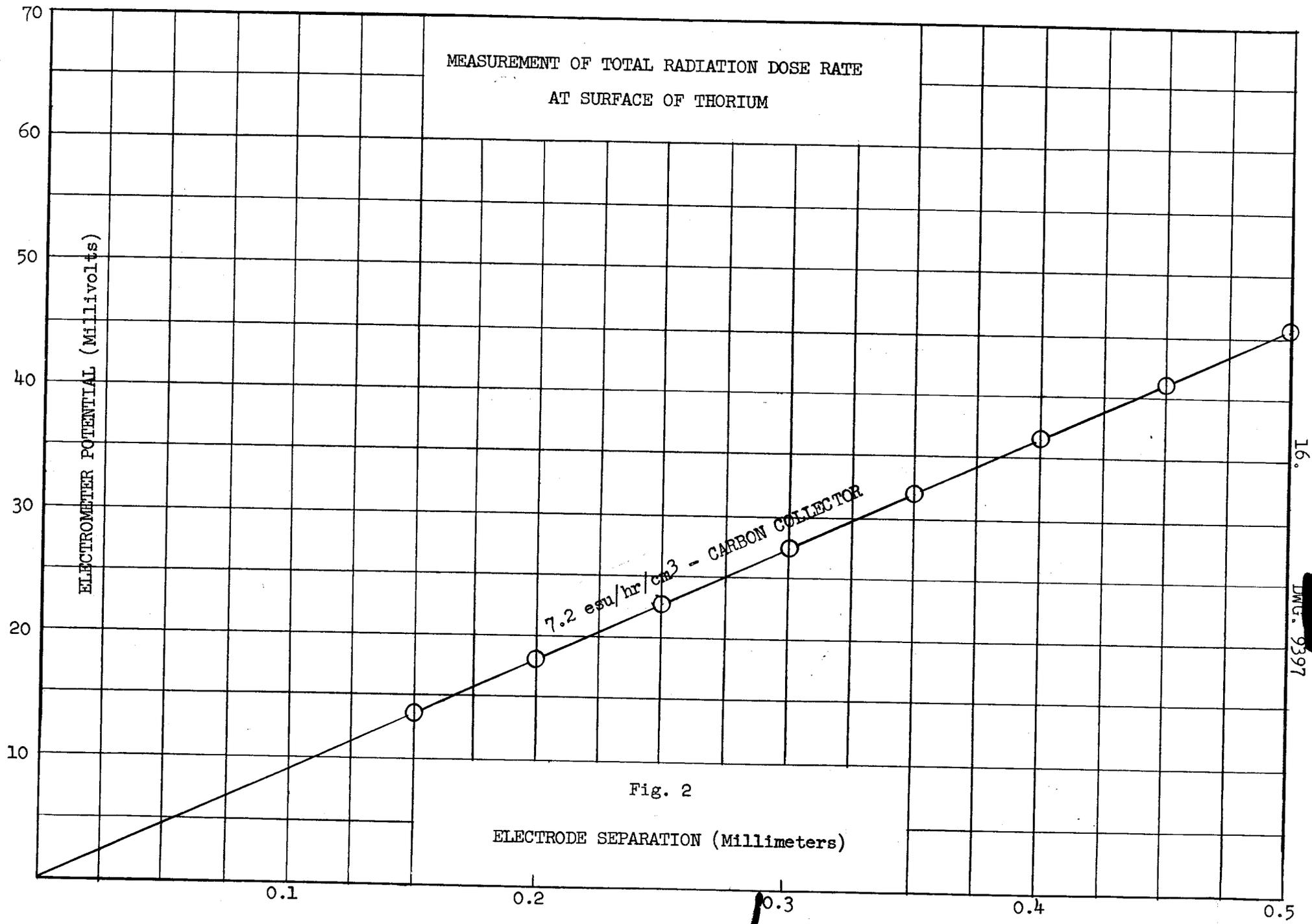
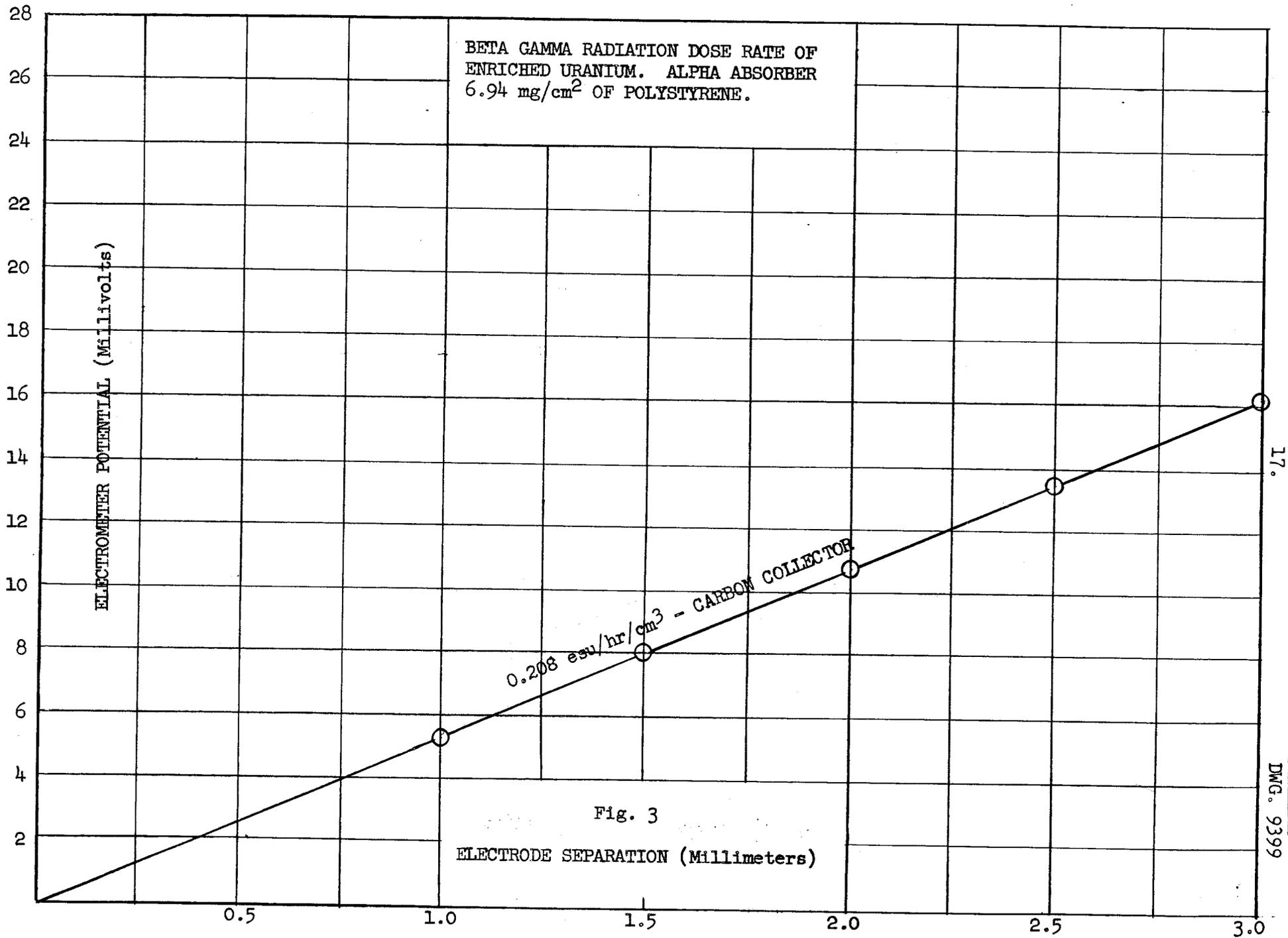


Fig. 1
ELECTRODE SEPARATION (Millimeters)

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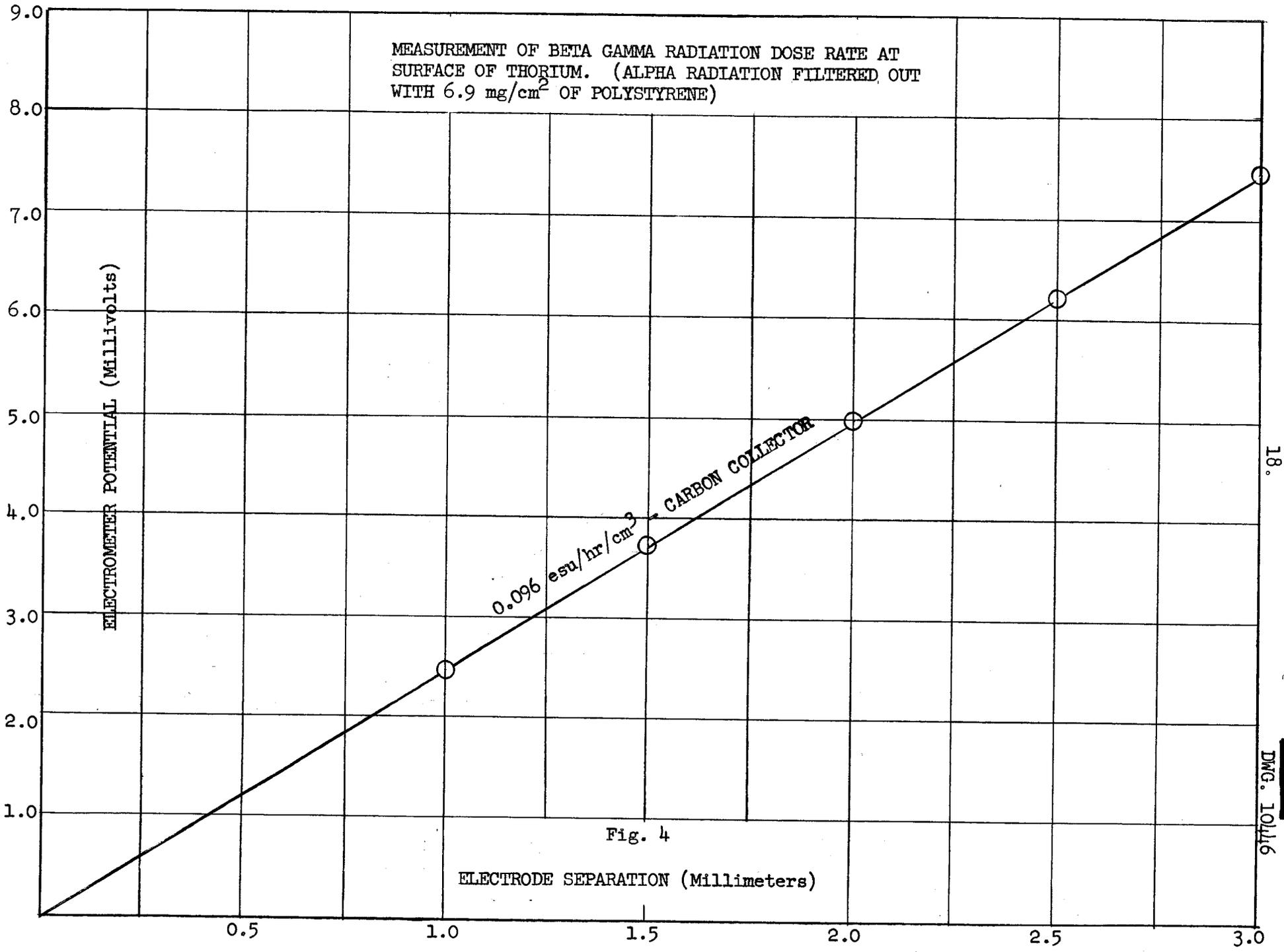


Fig. 4

18.

DWG. 101116