CANADA

THE RADIOMETRIC ANALYSIS OF URANIUM-BEARING STEELS

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MINES BRANCH

TECHNICAL BULLETIN

TB 25

Price 25 cents.

NOVEMBER 1961

Mines Branch Technical Bulletin TB 25

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by

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SYNOPSIS

Radiometric techniques which depend on counting either beta particles or low-energy gamma-rays provide a rapid, accurate method for estimating the uranium content of steel samples. With both methods uranium concentrations between 0.01 and 2% have been measured in less than 10 minutes, counting a 20-gram sample in a lead shield. Because the penetration of beta particles is limited to 1.4 mm of steel, the measurement is essentially that of the surface concentration; therefore, to eliminate serious error resulting from a non-uniform distribution of uranium in steel, samples should be prepared from fragmentary material, such as drillings or chips, which has been mechanically mixed and pressed into pellets to form an even surface for beta counting.

The measurement of gamma radiation from thorium-234, the daughter element of uranium-238, provides a result which is more representative of the uranium concentration throughout the whole sample. To obtain the required sensitivity for gamma counting, background effects are reduced by selecting radiation between 50 and 200 keV and by counting thin samples generally less than 6 mm thick to minimize self-absorption within the sample. Samples may be in the form of thin solid bars, fine drillings and chips, or fragments compressed into pellets as for the beta method. The last form is preferable; such samples can be prepared and counted in less than ten minutes. Details are given of the equipment required for both methods.

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Direction des mines

Bulletin technique TB 25

ANALYSE RADIOMÉTRIQUE D'ACIERS URANIFÈRES

par

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RÉSUMÉ

Les procédés radiométriques qui se fondent sur le dénombrement soit des particules bêta soit des rayons gamma dotés de peu d'énergie constituent des moyens rapides et précis d'évaluation de la teneur en uranium d'échantillons d'acier. A l'aide de l'un ou de l'autre procédé, on a pu mesurer en moins de 10 minutes des concentrations en uranium de l'ordre de 0.01 à 2 p. 100, un échantillon de 20 grammes étant placé derrière un écran de plomb. Du fait que la pénétration des particules bêta se limite à une couche d'acier de 1.4 millimètres, la mesure détermine essentiellement la concentration superficielle. En conséquence, afin d'éliminer une erreur grave qui pourrait résulter d'une répartition non uniforme de l'uranium au sein de l'acier, les échantillons doivent être préparés à partir de matériaux fragmentaires, comme par exemple des débris de forage ou des éclats, qui sont mélangés mécaniquement et comprimés en sphérules afin de constituer une surface uniforme pour fins de dénombrement des particules bêta.

Le dénombrement des rayons gamma émis par le thorium-234, produit de filiation de l'uranium-238, fournit un résultat encore beaucoup plus représentatif de la concentration en uranium dans l'ensemble de l'échantillon. Pour obtenir la sensibilité voulue pour le comptage des rayons gamma, on diminue les effets parasites en choisissant la radiation entre 50 et 200 keV et en procédant au comptage à partir d'échantillons minces, d'une épaisseur ordinairement inférieure à 6 mm, afin de réduire au minimum l'auto-absorption au sein de l'échantillon. Les échantillons peuvent se présenter sous forme de minces barres solides, de menus débris de forage et d'éclats, ou encore de fragments comprimés en sphérules comme il est indiqué plus haut. Cette dernière forme est préférable, car de tels échantillons peuvent être préparés et on peut procéder au comptage en moins de dix minutes. Le présent bulletin fournit des détails quant à l'outillage requis pour les deux procédés.

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INTRODUCTION

The effects of adding small amounts of uranium to metals have been receiving increasing attention recently. In such investigations it is important to know what proportion of the uranium added to a melt has remained in the alloy and how uniformly it is distributed. As a consequence there is a need for a simple, rapid method of analysis for uranium in metals so that a large number of determinations may be completed within a short period.

The technique to be described in this bulletin utilizes the radioactive properties of uranium to provide a comparison of the relative uranium content of steel samples. If a number of samples have been prepared from material previously analysed by a chemical method, they can serve as calibration standards that will enable the percentage of uranium in the unknown samples to be calculated. The accuracy of any determination depends very largely on the manner in which the sample has been prepared. With this method, it is generally possible to complete a uranium determination within five minutes, and by a suitable choice of counting equipment to have the counter read "per cent uranium" directly.

Samples can be assayed radiometrically in different forms: as solid samples, as loose drillings or chips, and as compressed fragments. The solid sample may consist of a small specimen of a

few grams in weight which can be placed in a lead shield close to a radiation detector, or it may be a large billet having a mass of several hundred kilograms. In the latter case the requirement may be to locate areas of high uranium concentration. This may be done conveniently with a probe containing a beta-sensitive Geiger tube, connected to a ratemeter for a continuous indication of the level of radioactivity, or connected to a scaling circuit with a timer if a numerical value is required (as in plotting the location of differing levels of activity). A Geiger tube having a thin window with a sensitive area of 10 cm^2 will give a useful indication of uranium concentrations of the order of 0.1% in steel. Because of the limited penetration of beta particles--about 1.4 mm in steel--a beta probe requires little shielding and is, therefore, easily handled; it is not affected to any significant extent by radiation from within the sample. It follows that the beta method should only be used for scanning surfaces and that no attempt should be made to interpret the readings as an indication of the uranium content of the whole volume unless the sample has been mixed mechanically.

For assaying the uranium content of a large volume of material, a gamma-ray method has been developed which utilizes the low-energy gamma-rays emitted by uranium metal. This method is also based on the use of comparison standards and has been found to be rapid and convenient.

SAMPLE PREPARATION

Samples in the form of small fragments can be readily mixed by stirring; however, such material generally contains particles of various sizes and shapes, rendering it virtually impossible to expose a uniform surface area to a detector. When using a gamma-ray detector it is difficult to control the density of such a porous structure adequately, although by using fine drillings or chips reproducible results can be achieved. Compression of the metal fragments into pellets containing a small amount of plastic binder has proved to be the best method of sample preparation to date. This technique provides better mixing of the sample, allows the dimensions and density to be controlled within the required limits, and produces a satisfactory surface for beta counting.

The first attempts to produce compressed pellets without adding a binding substance were not successful: the springy nature of the steel particles caused the pellets to flake apart when the pressure was removed. This problem was overcome by Mr. J.B. Zimmerman of the Extraction Metallurgy Division by coating the particles with a very thin layer of polystyrene. The metal particles were slurried with a very dilute solution of polystyrene in benzol and the solvent was evaporated under a heat lamp. This procedure has later been simplified by use of a plastic spray bomb. The particles

are spread out on an aluminum pie plate as shown in Figure 1, sprayed evenly but lightly with the spray and allowed to dry; drying can be accelerated by heating, but the sample should then be cooled. The coated metal fragments (20 grams has been found to be a convenient amount) are placed in a mould, 1.25 inches in diameter, and then pressed at room temperature under a pressure of 20,000 pounds per square inch.

DETERMINATIONS BY THE BETA METHOD

Uranium assaying of small samples by a shielded beta counter--consisting of an end-window Geiger tube and a scaler--was only attempted after it became possible to make satisfactory pellets with an even surface. The beta method will be described briefly here, before the other radiometric method, because it is relatively simple and requires less costly equipment than does gamma-ray counting.

Pellets were prepared as described, each weighing 20 grams. Their thickness was between 5 and 6 mm. Because of the limited penetration of the beta particles, any thickness greater than 1.5 mm constitutes an infinitely thick sample; if both surfaces of the pellet are counted, a thickness of 3 mm contributes to the measured count-rate. Precise thickness and weight are, therefore, not critical, although a thin pellet weighing less than 10 grams may tend to break unless handled carefully. A related factor is the

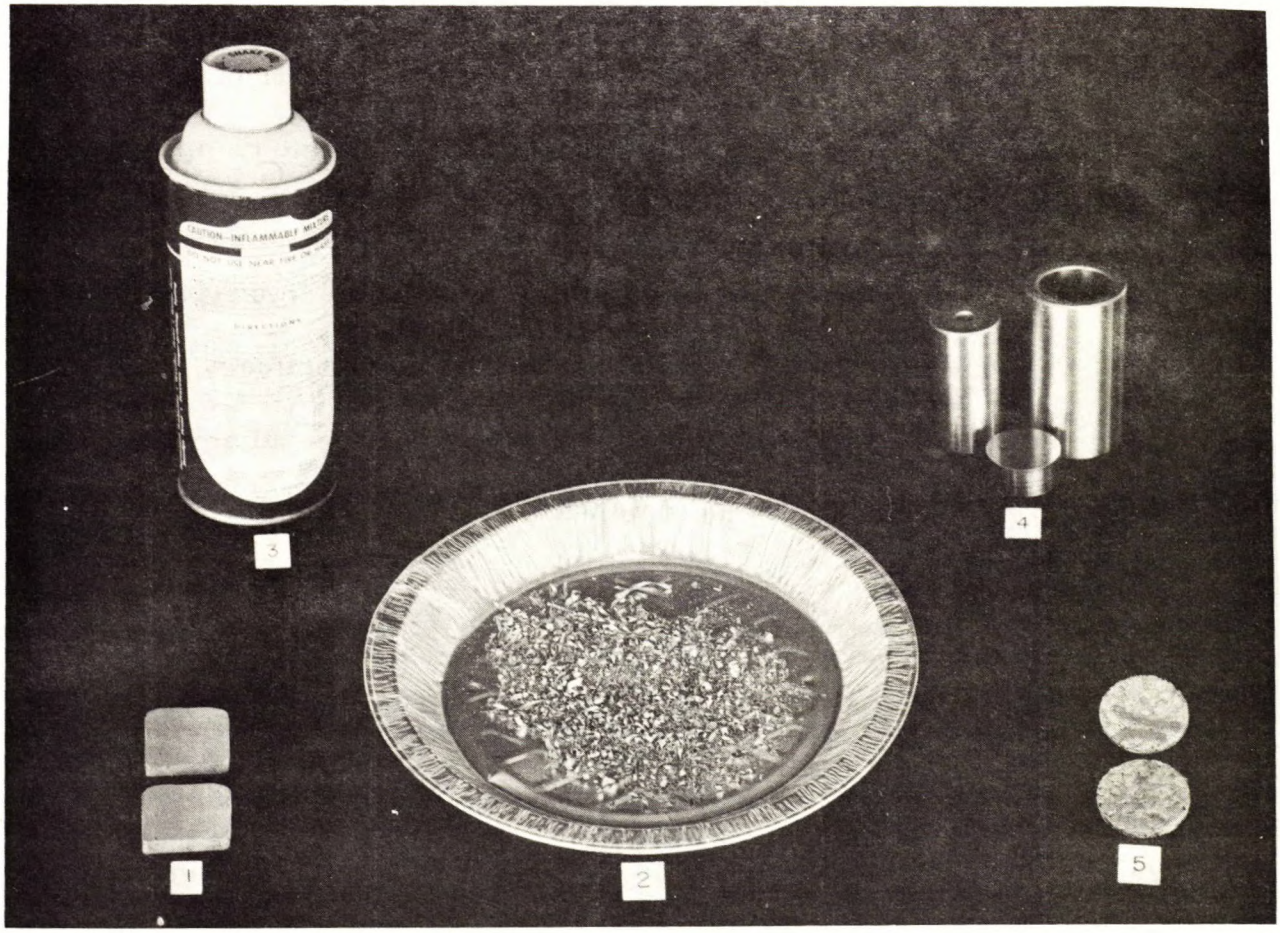

Figure 1 - Sample Preparation

(1) Sliced ingot. (2) Drillings ready for plastic spraying. (3) Spray bomb. (4) Typical mould. (5) Finished pellets.

distance between the surface of the pellet and the mica window of the Geiger tube. This distance should be kept constant; hence a major variation in sample thickness may require some compensation in this regard.

Some 20-gram pellets of differing uranium concentration were counted about 0.5 cm beneath a beta-sensitive, mica-window-type Geiger tube enclosed in a 2-inch lead shield. (This equipment is listed in Appendix 1.) Using a pellet 1.25 inches in diameter, the sensitivity was found to be about 2500 counts/minute/ per cent uranium with a background count, from a blank sample, of 19 counts/minute. Although the sensitivity could be increased by using a larger mould diameter, more sample material would be required in order to make a suitably strong pellet. The count-rate obtained from the 20-gram pellets will later be compared with the count-rate obtained by the gamma-ray method.

Direct reading of "per cent uranium content" can be achieved with the above-mentioned sensitivity as follows:

Sensitivity is $\frac{2500}{60}$ counts/second/per cent U;

using a scale of 100, $\frac{60 \times 100}{2500} = 2.4$ seconds.

Hence 2.4 seconds are required for one register count with a 1% sample. Similarly the background of 19 counts/minute is equivalent to $\frac{19}{60} \times \frac{2.4}{100} = 0.0076\%$ U. Therefore, count for a period of 240 seconds, then subtract a background equivalent to 0.0076% U from the dial reading that now indicates U values directly to

hundredths of a per cent. Repeat for the opposite face of the sample, and average. With a binary scale-of-64 unit, the corresponding counting period would be 154 seconds.

If a sufficient quantity of sample is available, more than one pellet could be made to obtain a more representative value for the uranium content. However, a gamma-ray determination would be more representative of the uranium content of the whole sample volume.

DETERMINATIONS BY THE GAMMA-RAY METHOD

Gamma radiation is most efficiently detected with a scintillation counter, which utilizes a thallium-activated sodium iodide crystal as a heavy, solid detector. Many people do not realize that purified uranium, from which radium and its daughter products have been removed, still possesses measurable gamma activity provided certain procedures are adopted in its detection. The gamma-ray spectrogram of purified uranium (Figure 2) shows that the strongest emission peaks occur below 200 keV; the radiation is therefore quite "soft", i.e., not penetrating. Accordingly, the counter should be operated with a low-energy threshold, about 60 keV, and the detector should be enclosed in a thin aluminum or magnesium cover only. The sample itself should be limited in thickness, preferably to less than a centimetre, to minimize internal absorption of this soft radiation within the sample. Under these conditions it is

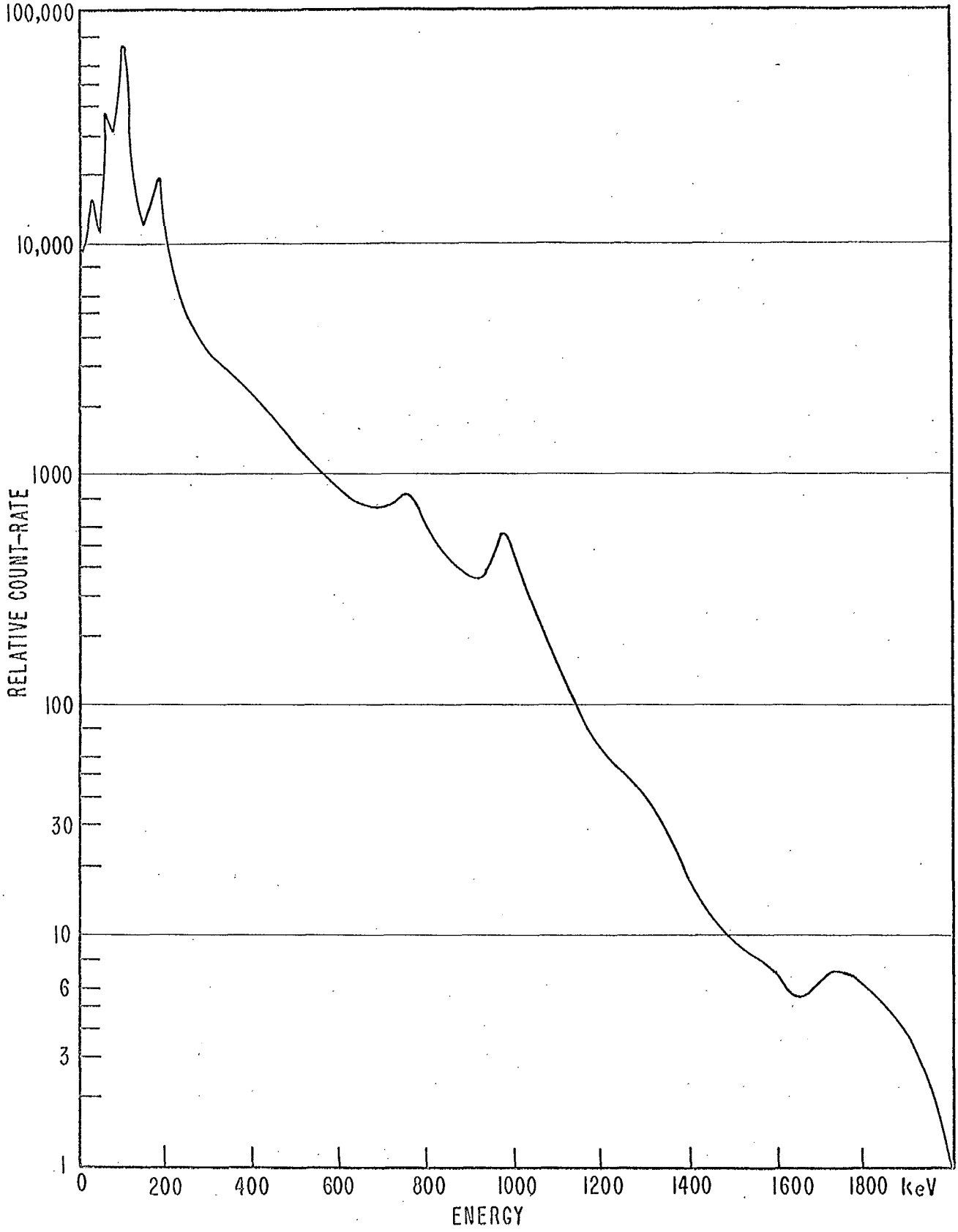

FIGURE 2 – GAMMA SPECTROGRAM OF REFINED URANIUM

possible to determine the uranium content of an unknown sample rapidly, with adequate accuracy, by comparing its gamma-ray emission within a selected energy interval, or channel, with that of a similar sample of known uranium content.

(a) Description of the Equipment

The gamma-ray detection equipment used was assembled in two versions: (1) a more complex one, shown in Figure 3, having a large lead shield with 4-inch-thick walls and a multichannel analyser and recording system; and (2) a system, shown in Figure 4, suitable for routine assay work, consisting of a small lead box with 2-inch-thick walls and a single-channel count recorder. The first type is a research instrument with a shield having the maximum practical thickness for the range of gamma-ray energies to be used in uranium determinations. It permits simultaneous investigation of these energies by recording the individual total counts in 100 energy intervals. The optimum energy range could thereby be determined for use of any single-channel analyser. Two sizes of detector and two thicknesses of shielding were used to establish minimum acceptable specifications for sensitive routine assay work.

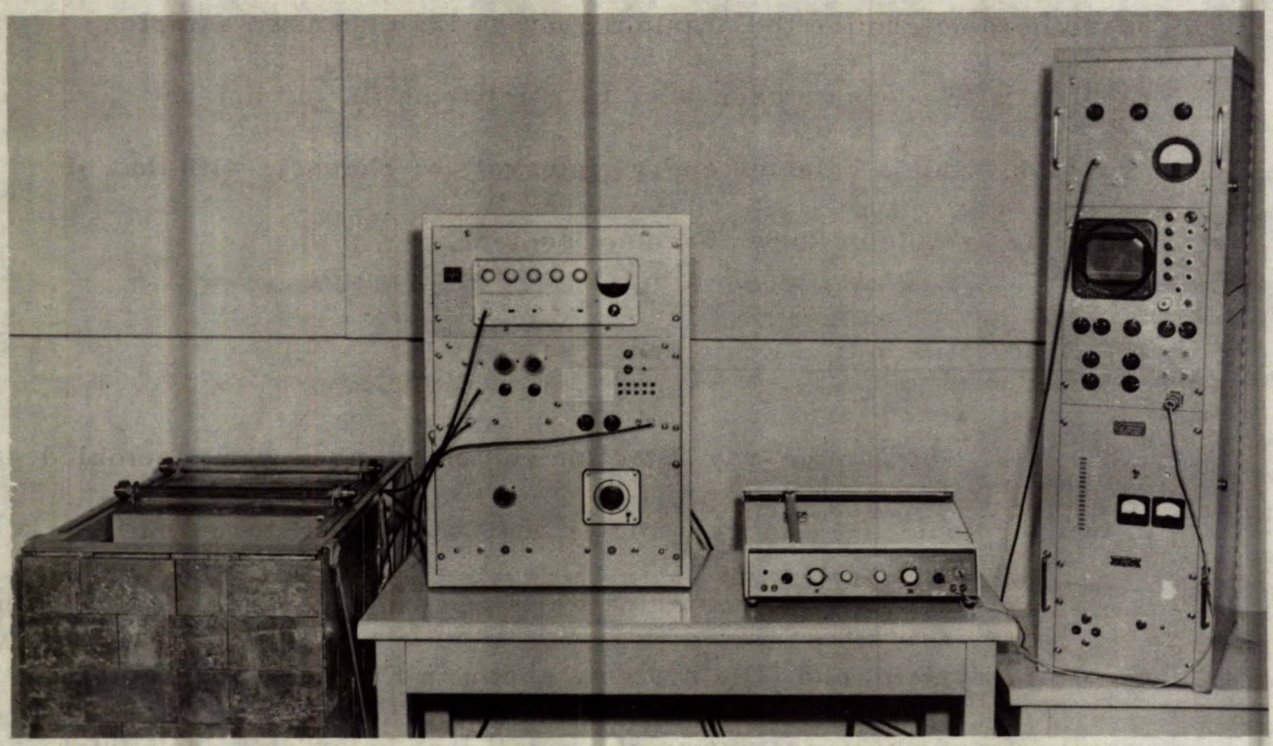

Figure 3 - View of the laboratory gamma-ray spectrometer.

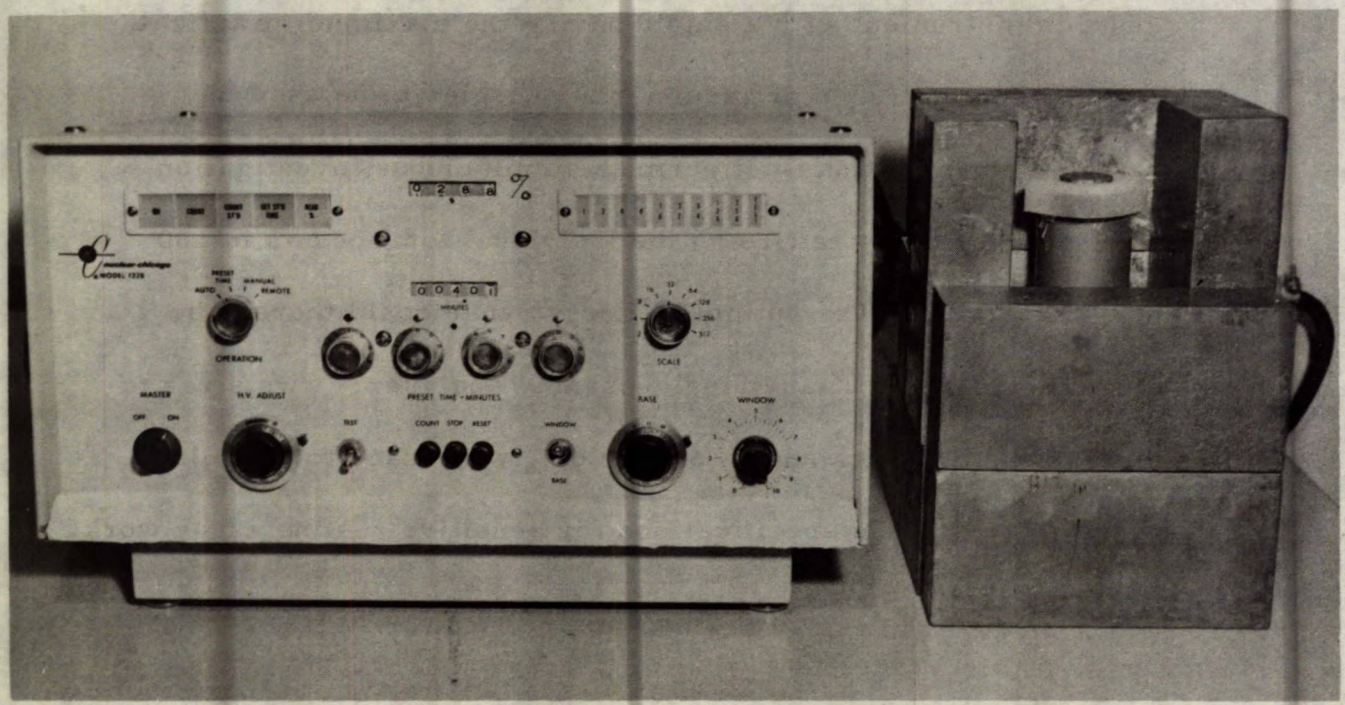

Figure 4 - View of the routine gamma assay equipment.

The principal components of this equipment are outlined by a block diagram in Figure 5 and are as follows:

(a) A sodium iodide crystal mounted against a photomultiplier tube to serve as a gamma-ray scintillation detector.

(b) A lead shield, enclosing detector and sample to reduce background radiation to an acceptable level.

(c) A stable, low-noise, high-voltage power supply to operate the photomultiplier tube.

(d) A linear pulse amplifier.

(e) A spectrum analyser and pulse counter.

In the routine model, (c), (d) and (e) are combined in a single unit, such as the Nuclear-Chicago Model 132B Analyzer-Computer, as indicated in the more detailed description in Appendix 2. The calibration procedure is described in Appendix 3.

(b) Nature of the Gamma-ray Spectra

Using the 100-channel analyser, the gamma-ray spectrum from uranium metal was examined; the resultant spectrogram, for energies up to 2MeV, is shown in Figure 2. The energies of the radiations from the related decay products are indicated in Table 1. Many of these radiations are only minor contributors to the overall spectrum (1). It will be noted from Figure 2 that the decrease in count-rate toward the higher energies is extremely rapid.

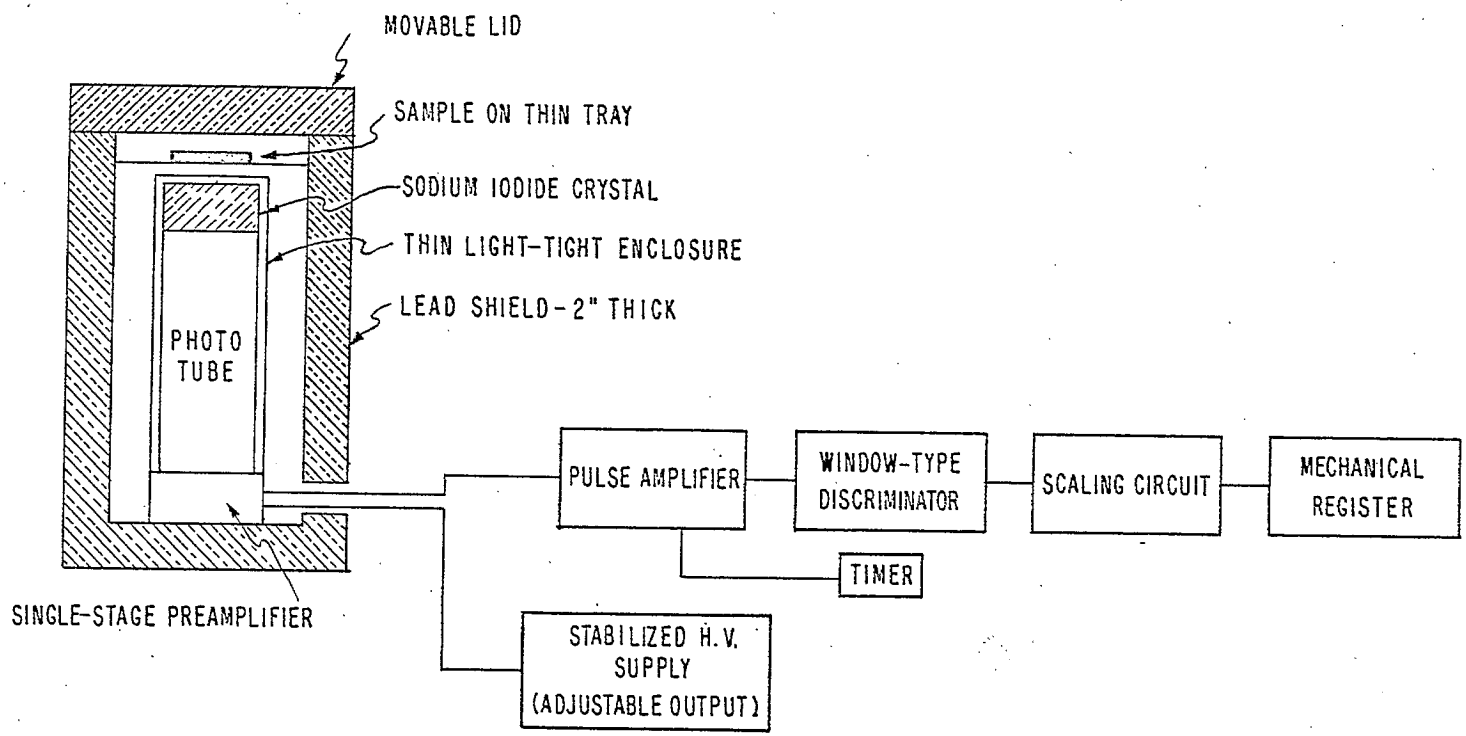

FIGURE 5 - BLOCK DIAGRAM OF THE PRINCIPAL UNITS OF THE ROUTINE GAMMA ASSAY EQUIPMENT.

TABLE 1*

Gamma-Ray Energies of U-238, U-235 and Their Immediate Decay Products

Nuclide	Half-life	Energy, in keV
U-238	4.5×10^9 y	48
Th-234	24.1 d	29, 63, 91
Pa-234	6.66 h	43, 99, 153, 225, 368, etc.
Pa-234m	1.17 min	1000 (37R), 760 (26R), 1840 (04R), 1490 (01R), 1700 (01R), etc.
U-234	2.5×10^5 y	53, 121
U-235	7.1×10^8 y	185 (55A), 143 (12A), 95 (9A), 110 (5A), 290 SUM, 390 SUM
Th-231	1.07 d	26 (13A), 84 (11A), 99 (2A), 146 (OA), 163 (OA)

Notes re Table 1:

In the "Energy, in keV" column, the principal peaks are underlined; A refers to absolute intensity, and R to relative intensity; (OA) means less than $\frac{1}{2}$% A; and SUM = summation peak.

* The data for this table were extracted from A.A. Madson's Appendix IV to "Applied Gamma-ray Spectrometry" (1960) (ed. by C.E. Crouthamel). See reference (1).

The low-energy portion of the spectrum is shown in greater detail in Figure 6. The effect of self-absorption within a sample may be observed by comparing the two curves. The solid curve was obtained from a steel sample containing 0.58% uranium and consisting of fine drillings occupying a cylindrical space 33 mm in diameter and 13 mm thick. The dashed curve is the spectrogram from a very thin (< 0.5 mm) sample in a similar container. When the two curves are normalized to the principal peak, just above 90 keV, an increased relative count is to be noted at lower energies in the thin sample. The curves shown in Figure 7, which have been calculated from Siegbahn's tables (2), indicate the effects of absorption in different thicknesses of solid steel and the dependence of self-absorption on gamma-ray energy; these curves apply more strictly to a collimated beam of radiation. In the case of drillings and other fragmented material, it would be necessary to consider an equivalent thickness of the solid material.

In addition to corrections for variations in density, there may be a further geometry correction factor involving the differences in distance from the various layers to the radiation detector. An empirical curve was determined which illustrates the effect of distance and is shown in Figure 8. Net count-rates from a very thin layer of uranium oxide powder were plotted for a number of air distances above the detector. If the diameter of the detector is large

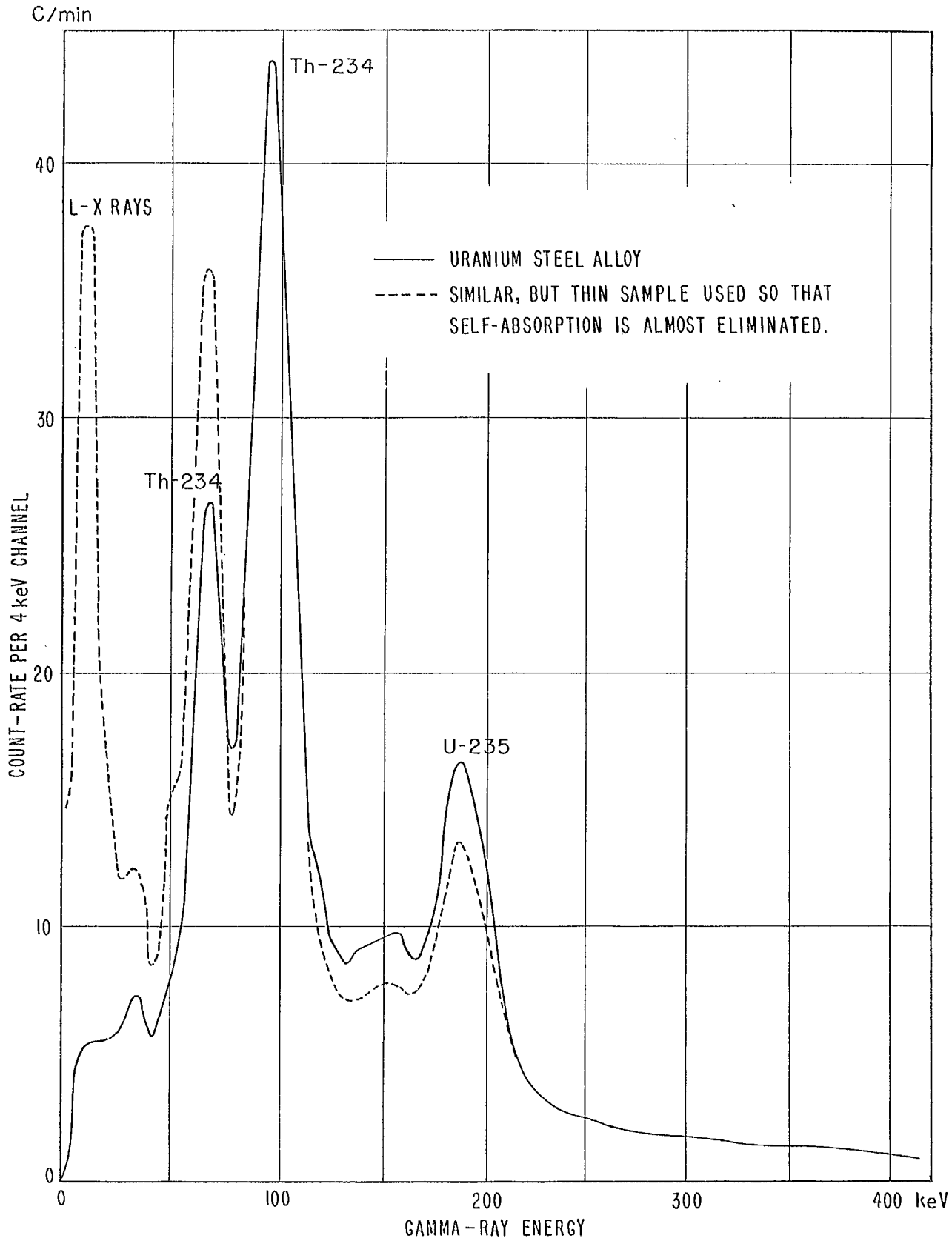

FIGURE 6 – GAMMA SPECTROGRAM OF URANIUM BELOW 400 keV.

URANIUM-STEEL ALLOY, 20 GRAMS – 0.58 % U.

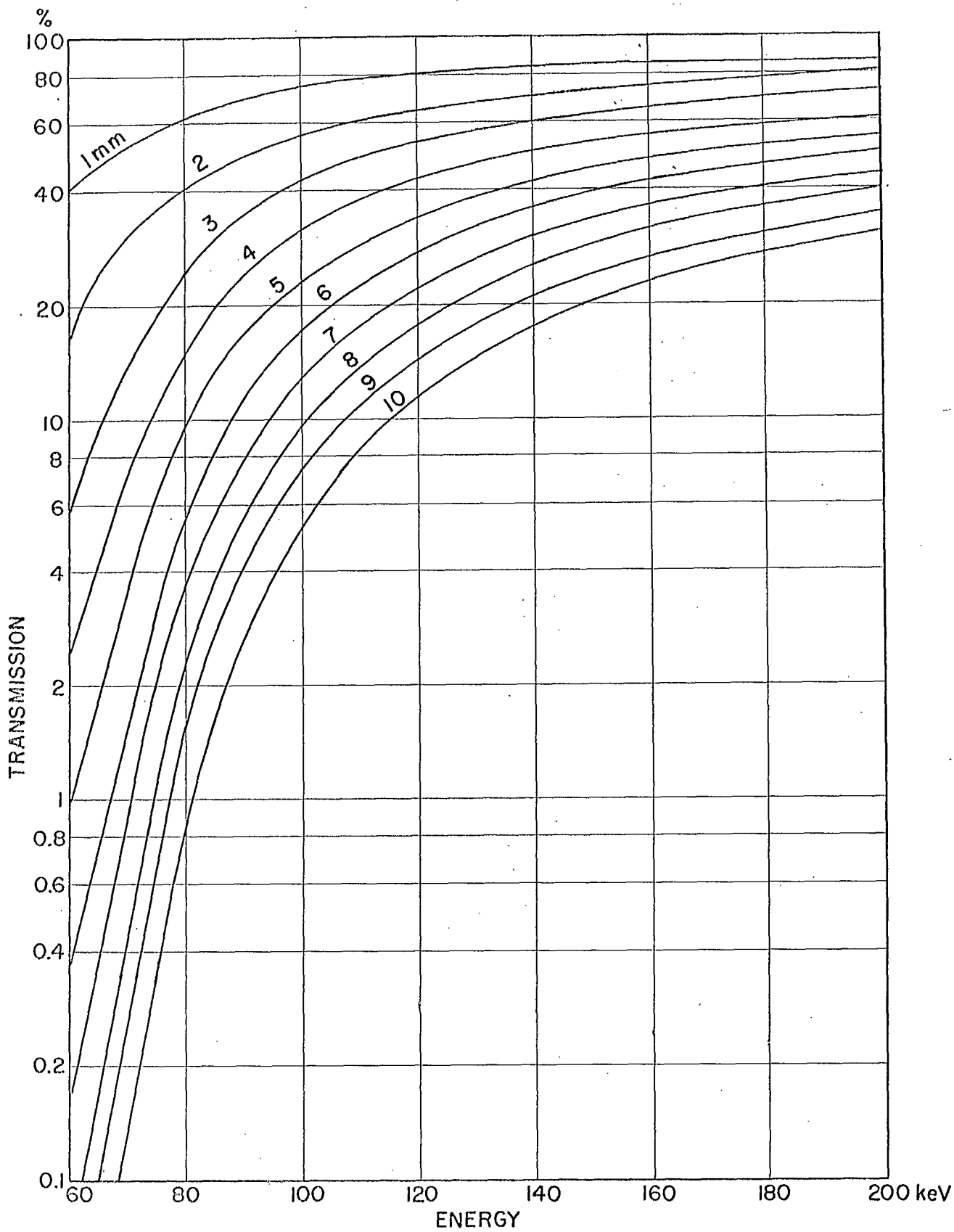

FIGURE 7 – THE TRANSMISSION OF SOFT GAMMA RADIATION THROUGH STEEL AS RELATED TO ENERGY AND THICKNESS.

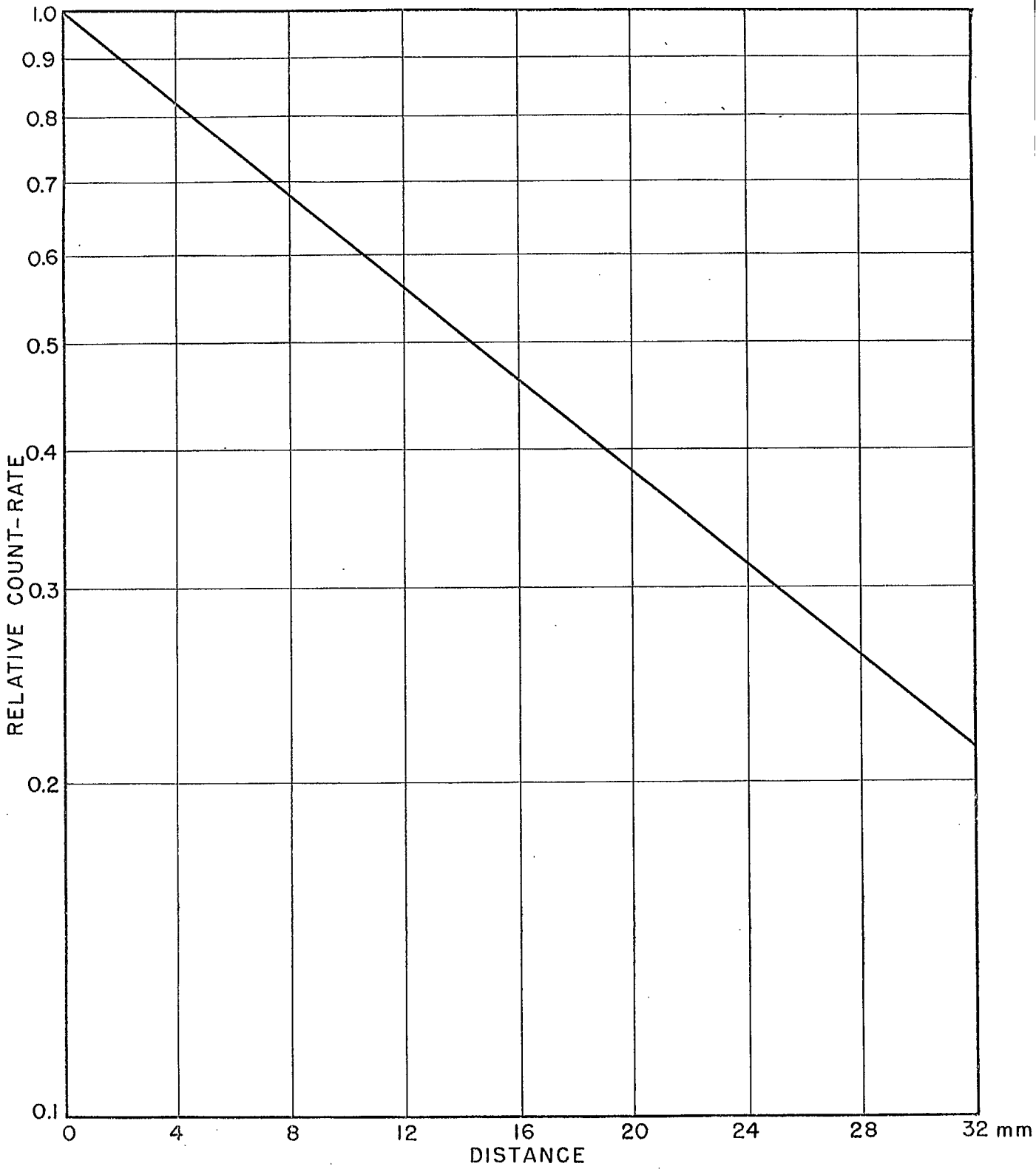

FIGURE 8 - THE COUNT-RATE FROM A THIN SOURCE RELATED TO AIR DISTANCE FROM THE GAMMA-RAY DETECTOR.

relative to that of the sample, edge effects will tend to reduce the absorption. Figures 7 and 8 are included as guides in choosing the best sample geometry but should not be used for purposes of calculation.

The benefits of good sample geometry are illustrated as follows: the sample material used to produce the spectrogram shown in Figure 6 (20 grams of drillings, 0.58% U) was counted under four different conditions of sample geometry, as shown in Table 2.

TABLE 2

Variation in Sensitivity with Sample Geometry

(Energy range, 75-125 keV; background, 25 counts/minute)

No.	Sample weight, grams	Diameter of container, mm	Sample thickness, mm (approx.)	Net counts per minute per gram
1	10	33	6.5	81.2
2	20	33	13	59.6
3	10	50	2.5	91.7
4	20	50	5	75.4

The results in Table 2 show not only that the 50-mm sample diameter produces a 13% higher sensitivity than the 33-mm sample of the same weight, but also that the dependence of sensitivity on sample weight is reduced; i.e., increasing the 50-mm sample from 10 to 20 grams results in an overall reduction in sensitivity of 17.8%, whereas for the 33-mm sample the reduction is 26.6%.

Another factor affecting the relative sensitivity of a detector is background radiation. Salmon (3), referring to the "figure of merit", F, of a detector where

$$F = \frac{(\text{counting efficiency})^2}{\text{background}},$$

compares a number of gamma-ray detectors at 100 keV energy as follows:

TABLE 3

Relative Merit of Various Gamma-Ray Detectors at 100 keV (3)

Counter	Maximum overall per cent efficiency	Background, counts/min	Relative F
Geiger counter	0.1	8	0.005
Proportional counter	0.2	15	0.01
Plastic scintillator, 1 in. x 0.05 in.	0.1	8	0.005
NaI (Tl) crystal, 1 in. x 1½ in.	15	10	90
NaI (Tl) crystal, 3 in. x 3 in.	30	50	70

The background spectrograms in Figure 9 were obtained in the lead cave (13 in. x 13 in. x 28 in. high inside) having walls 4 in. thick. The lead X-ray peak at 73 keV was prominent until a 0.25-in. lining of copper was added. If the inner surface is completely lined with copper, more than 95% of these X-rays will be removed. For uranium-steel assay work, limited to energies

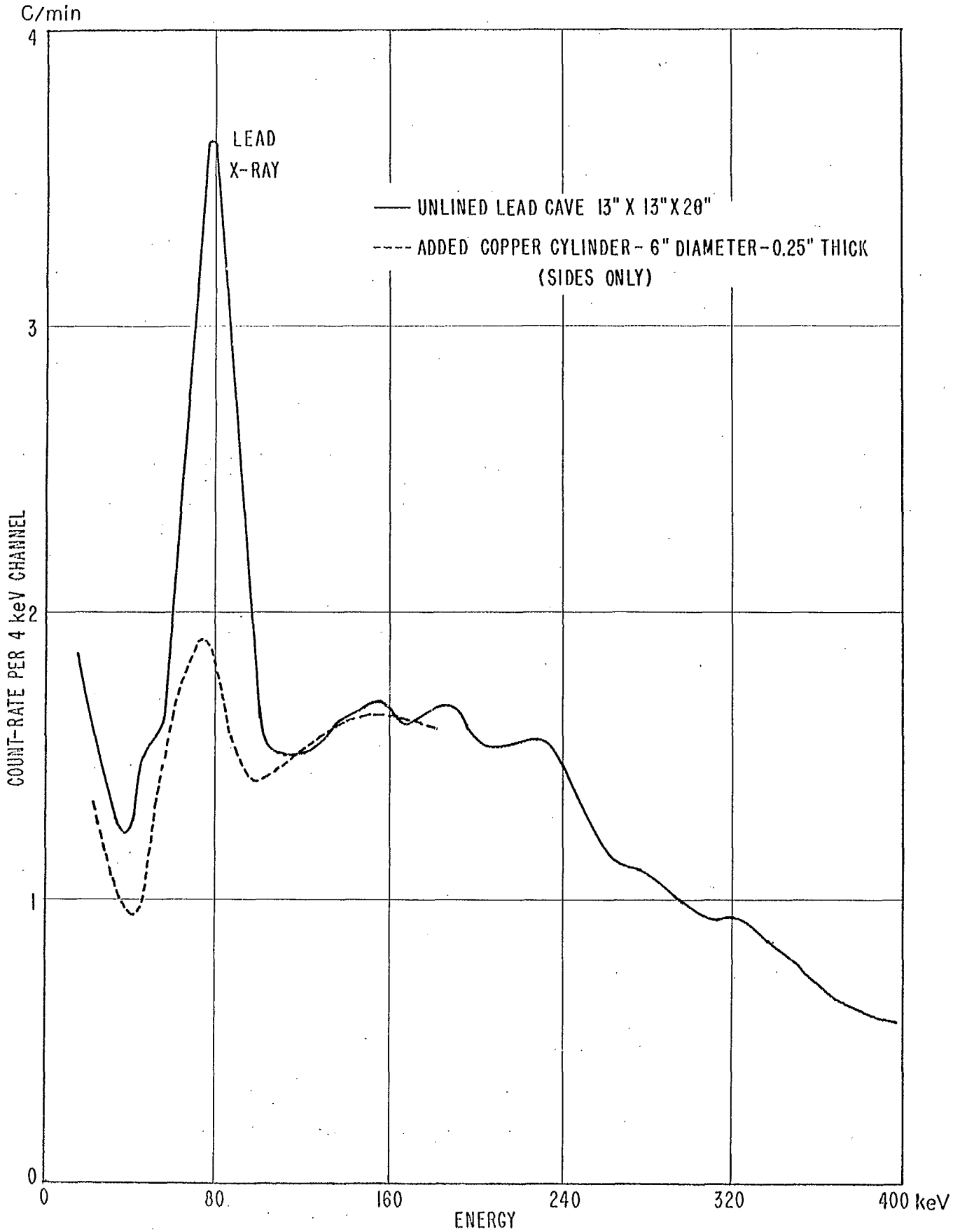

FIGURE 9 - BACKGROUND SPECTROGRAMS.

below 200 keV, a small, brass-lined lead shield having walls 2 to 3 inches thick is adequate. Much of the residual background will originate from weak, natural radioactivity in the materials of the detector assembly (4, 5), including potassium-40 occurring as an impurity in the sodium iodide crystal. The potassium background is proportional to the volume of the crystal, and may be minimized in this application by using a thin crystal (0.25 to 0.50 in. thick); this will also reduce the secondary effects from background radiation of higher energy.

If a detector unit is being especially constructed so that the components may be selected, it is worthwhile to use a plastic socket for the photomultiplier tube instead of the usual type made of mica-filled bakelite. Table 4 lists the background counts obtained under varied conditions of detector and shield thickness, relating these to the sensitivity to uranium and to the detection efficiency, i.e. $\frac{(\text{count-rate})^2}{\text{background}}$.

TABLE 4

Sensitivity to Uranium versus Background Related to Detector Size and Shield Thickness

Description of shielding	B = Background			C = Sensitivity, counts/min	Relative merit, $C^2/B \times 100$
	2 in. diameter crystal		Equiv. % U		
	$\frac{1}{2}$ in. thick	2 in. thick	*	*	
Small lead shield, 4 in. x 4 in. x 12 in. (inside measurements); walls 2 in. thick	32c/m (151)		0.011 (0.035)	15 (21.5)	700 (306)
		88c/m (367)	0.025 (0.060)	17.8 (30.8)	360 (260)
Lead cave, 13 in. x 13 in. x 28 in. (inside measurements); walls 4 in. thick	25		0.0083	15	900
		42	0.013	17.8	750
Same lead cave, but phototube used without base or socket	20 (estimate)		0.0067	15	1120

*For a 20-gram compressed sample, count-rates are those obtained between 65 and 175keV.

Figures in brackets are for total count above 65keV.

The detection efficiency (also referred to in Table 4 as "relative merit") was determined using the $\frac{1}{2}$-in.-thick crystal for several different intervals in the gamma-ray spectrum, between 50 and 220 keV, and for both the small 2-in. and 4-in. shields. Calculations based on these results indicate 50 to 200 keV to be the best energy interval for assay purposes; however, a threshold somewhat higher than 50 keV has been chosen to reduce the effects of sample absorption previously mentioned. The upper cut-off energy is less critical than the threshold value, and may be set between 160 and 220 keV; the values in brackets (where no upper cut-off was used) indicate a greatly reduced sensitivity relative to background.

Good counting conditions may, therefore, be summarized as follows:

(1) Use a thin sample--less than 6 mm in thickness, if possible--to minimize absorption and to keep the active material close to the detector. The sample area could ideally equal the cross-sectional area of the crystal.

(2) The detector should be a thin sodium iodide crystal, enclosed in a thin (<1 mm) aluminum container to reduce absorption.

(3) A plastic tube socket is preferable for the photomultiplier to reduce the background radiation from the detector assembly.

(4) A small, 2-in. lead shield is adequate for the sample and detector. Some reduction in background can be achieved with a copper or brass lining, 4-6 mm thick, to suppress 73 keV lead X-rays; this lining can also serve to support the sample tray.

(5) Samples are counted over an approximate energy range of 60 to 180 keV. This range may be determined for a particular scintillation counter using suitable radioactive isotopes; Cs-137 provides two calibration points for the base (or threshold) voltage control at 32 keV and 662 keV.

(c) Assays of Samples in Different Forms

This section describes the handling of various types of samples that have been counted: solid samples, coarse and fine turnings or drillings that were either loose or compressed in plastic vials, and compressed samples in which a small amount of a plastic binder was used. As mentioned before, the last of these is the most convenient; the others are described in case it is not possible

to produce pellets. Because the radiometric method is not an absolute one, frequent comparison should be made with identically-shaped samples whose uranium content has been determined by the fluorimetric (chemical) method.

The sample thickness has been limited to 6 mm in the case of solid samples, but has been somewhat greater in the case of loose turnings. With the latter type of sample, the thickness required was reduced by increasing the sample area, using thin plastic cups (about 50 mm in diameter) which are available at low cost.

Table 5 lists readings from a group of solid steel samples* having a uniform thickness of 6 mm and not varying too widely in weight. Their count-rates per gram are compared with the chemical assay values from two laboratories by means of the graph in Figure 10. It is seen that there is a good linear relationship over most of the range.

In Table 6, results are given for uncompressed drillings. The effect of variation in sample thickness, related to differences in sample weight and enclosed air spaces, was compensated for with an empirical weight correction curve, shown in Figure 11. The largest sample of the group was divided into a number of smaller samples representing the weight range required in the curve. This procedure could be repeated several times to minimize possible effects of segregation, provided these fractions were clean and free

* The solid steel samples were obtained from the Physical Metallurgy Division of the Mines Branch.

TABLE 5

Assays of Small Metal Bars of Uniform Thickness and Approximately the Same Area*

Sample No.	Weight, (grams)	Counts per 5 minutes		Total 10-minute count	Net count per 10 minutes	Net count per minute per gram	Uranium content (% U)		
		Top	Bottom				Chem. Lab A*	Chem. Lab B*	Average, A and B
A-1555	38.0	912	1006	1918	1575	4.2	0.045	0.0365	0.041
A-1403	40.5	1767	1732	3499	3156	7.8	0.084	0.072	0.078
A-1395	40.3	1990	1970	3960	3617	9.0	0.093	0.090	0.092
A-1373	34.7	2108	2179	4287	3944	11.4	0.10	0.12	0.11
A-1374	36.0	2396	2477	4873	4530	12.6	0.127	0.13	0.128
A-1518	45.5	3230	3118	6348	6005	13.2	0.127	0.14	0.134
A-1398	36.3	4310	4504	8814	8471	23.3	0.23	0.228	0.23
Background (steel only)	35	161	182	343					

*As supplied by the Physical Metallurgy Division.

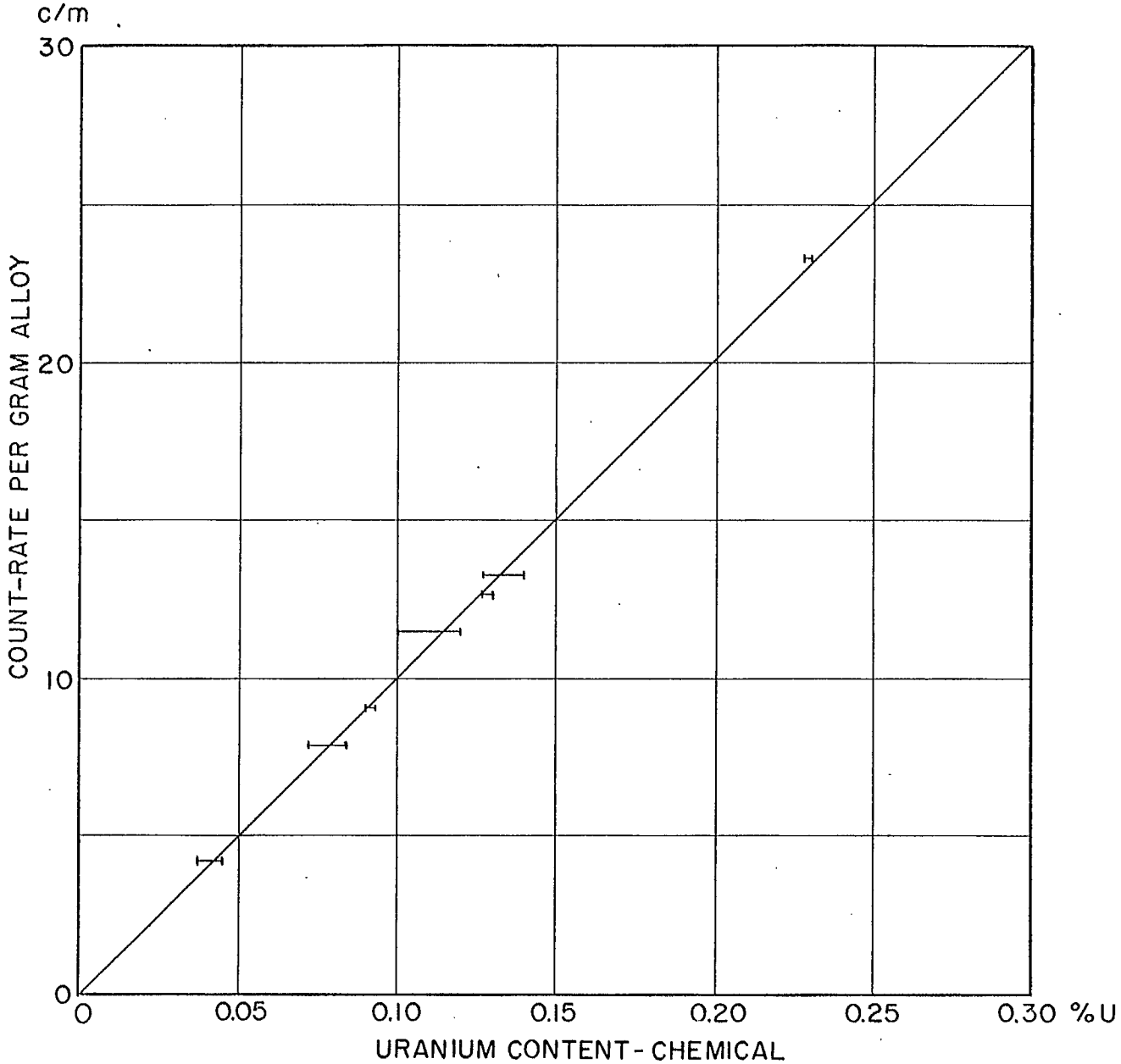

FIGURE 10 - COUNT-RATES FROM SMALL METAL BARS COMPARED WITH THE URANIUM CONCENTRATION AS DETERMINED BY CHEMICAL ANALYSIS

TABLE 6

Assays of Uncompressed Metal Drillings and Turnings

Sample No.	Weight, (grams)	Count per 10 minutes	Net c/min/g A	Weight factor B	Product A x B C	% U C x D*	% U_3O_8 (Radiom.)	% U_3O_8 (Chem.)
5214	26.7	28,435	104.4	1.46	152	0.70	0.83	0.78
"	25.0	27,020	105.6					
"	20.0	24,252	118.2					
"	15.0	20,248	130.9					
"	10.0	15,534	149.2	=1.000				
5208	17.4	658	2.4	1.19	2.9	0.013	0.015	0.010
5209	21.6	2427	8.38	1.30	10.89	0.050	0.059	0.063
5210	17.0	3355	16.11	1.18	19.0	0.087	0.103	0.10
5211	16.7	6463	35.0	1.17	41.0	0.19	0.22	0.22
5212	21.7	10,136	43.9	1.31	57.5	0.26	0.31	0.34
5213	21.0	14,963	68.3	1.29	88.1	0.41	0.48	0.53
5215	17.6	31,084	173	1.20	208	0.96	1.13	1.04
5216	21.7	31,028	140	1.31	183	0.84	0.99	1.09
5217	23.8	51,735	215	1.37	295	1.36	1.60	1.52
Background		616						

*D = calibration factor, 217 c/min/g per 1% U.

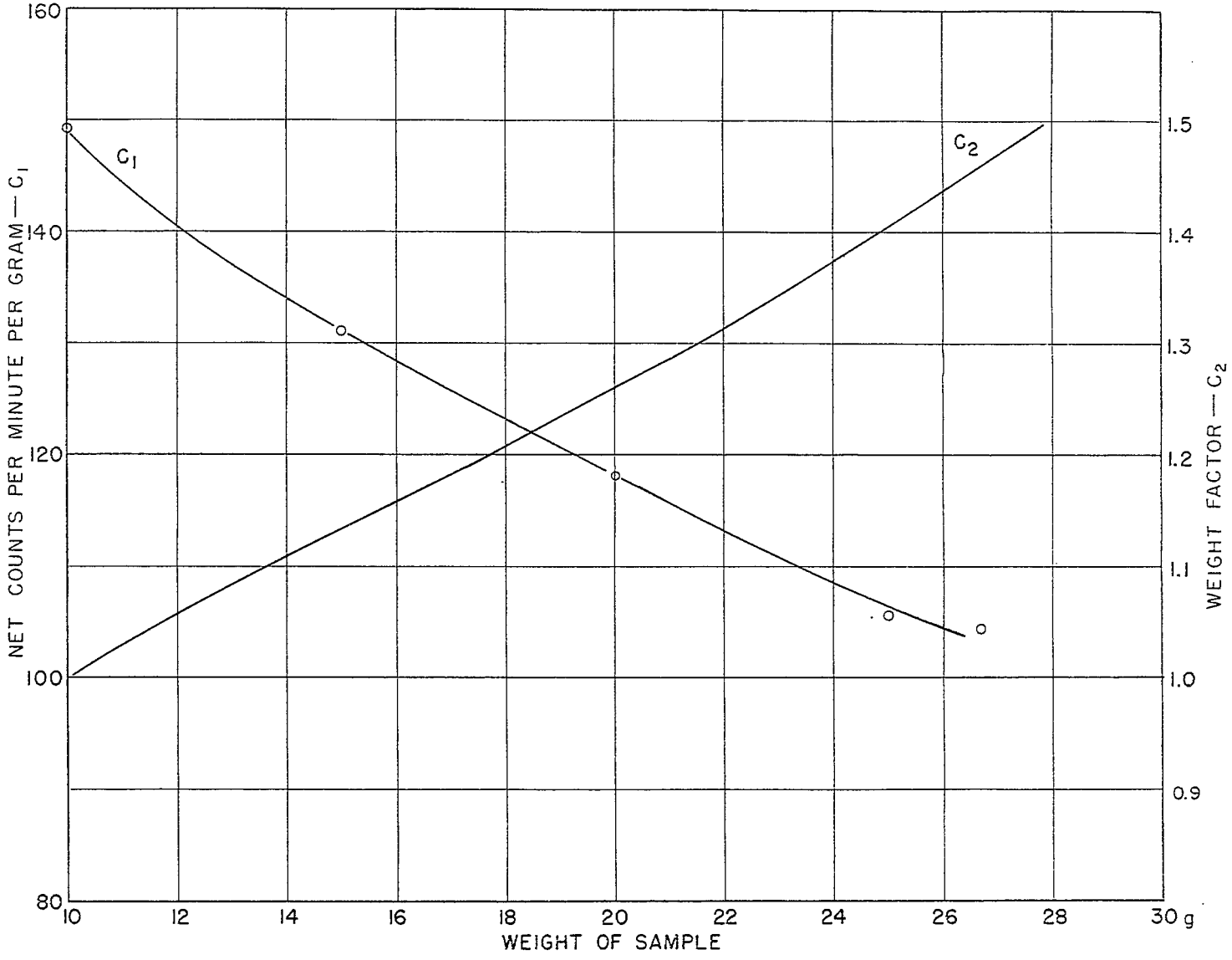

FIGURE II - EMPIRICAL CURVE TO CORRECT FOR WEIGHT VARIATION AMONG SAMPLES COMPOSED OF LOOSE DRILLINGS.

of particles more active than the sample as a whole. Because no compression was employed at this stage, the method was dependent on the accuracy with which the equivalent thickness could be estimated for a sample having a somewhat indefinite upper surface. It was found that this problem was not serious when the particles were small; it is, therefore, suggested that the drill bits used for sampling be ground at such an angle as to produce very fine drillings.

To provide a more definite sample thickness, and also to position the sample mass closer to the detector, a number of samples were compressed in round plastic vials 33 mm in diameter (wider vials, although preferable, were not immediately available), using a ram machined to a close fit in the plastic tube. The latter was supported on the outside by a metal ring also machined to a close fit.

The advantage of compressing samples made up of rather coarse drillings is illustrated by the following two examples:

Sample No.	Weight, (grams)	Uranium content (chemical), % U	Count-rate (c/min/mg U)	
			Before compressing	After compressing
5139	16.54	0.41	13.9	18.2
5140	16.38	0.50	17.0	18.9

The sample weights were almost equal, yet the thicknesses differed appreciably. The previous correction factor, based on weight for a more uniform material, could not be used here without an additional

thickness factor. However, compression reduced the thickness of the first sample from 31 to 7 mm, and that of the second from 12 to 8 mm. The values in the last column indicate the improved sample geometry in relation to the detector after compression. The difference in specific sensitivity was reduced to 3.8%, whereas before compression it had been 20%.

Comparative count-rates are shown in Table 7 for a number of samples pressed into pellets. No correction factors were required. The calculated uranium percentages were obtained by dividing the net average count-rate by 3000, which is the count-rate per minute for a 20-gram sample containing 1% U (counted between 65 and 175 keV, using a $\frac{1}{2}$-in. crystal). Because very small fragments occasionally become detached from these pellets, it is best to handle the pellets in thin flat plastic trays to avoid contamination of the shield cavity.

It is, of course, convenient to choose a counting period with the gamma-ray method so as to have the scaling circuit read "per cent uranium" directly, as in the beta method. The single-channel analyser used in these routine tests provides a particularly convenient timer for choosing non-integral, reproducible counting periods. It is a decade unit adjustable to hundredths of a minute. The correct counting interval may be found empirically without calculation, using known calibration standards; however, the "equivalent per cent uranium" of the background must still be

TABLE 7

Typical Assay Results for 20-Gram Pellets

Sample No.	Net count-rate, gamma			Net count-rate, beta			Uranium content (% U)		
	1st side	2nd side	Average	1st side	2nd side	Average	Calculated		Chem.
							Gamma	Beta	
1719	36.0	33.7	34.8	19.6	11.2	15.4	0.0116	0.006	0.010
1720	53.3	45.3	49.3				0.016		0.016
1721	164	140	152	120	121	120	0.050	0.048	0.051
1722	150	147	148				0.049		0.060
1723	382	380	381				0.13		0.14
1724	1067	1041	1054	785	817	801	0.35	0.32	0.39
1725	2378	2441	2410				0.80		0.98
1726	4771	4675	4723				1.57		1.64
1727	6021	5954	5988	5190	5337	5263	2.00	2.1	1.69
1728	4335	4318	4326				1.44		1.41
Background (blank sample)			33			19			

Counting intervals per side: Gamma - 3 minutes, Nos. 1719 to 1724
1 minute, Nos. 1725 to 1728
Beta - 5 minutes, Nos. 1719, 1721, 1724
1 minute, No. 1727

measured and calculated for the time interval, for subtraction from the gross count for the sample plus background. Very good results have been obtained for samples containing more than 0.03% uranium (20-gram pellets) with counting intervals as short as about one minute for each side of the sample; periods of 10 to 20 minutes will give higher accuracy if the investigation warrants this.

CONCLUSIONS

Stable radiometric equipment (operated without wide temperature variations near the detector) can provide a sensitive, rapid, non-destructive method for determining the uranium content of steel alloys. Beta determinations are possible with mixed samples having reproducible surface areas, such as drillings pressed to form pellets. Gamma determinations represent more accurately the uranium content throughout the whole sample, but the absorption of soft gamma radiation must be minimized by a proper choice of sample size and shape; the effect of background activity must also be minimized. For the gamma determination, solid, thin samples of uniform dimensions--e.g., bars--are suitable; in other cases, such as with drillings and other fragmented steel samples, the density throughout each sample should be made as uniform as possible by using fragments that are small, or preferably by mixing and compressing the samples into pellets. The accuracy of both counting techniques depends largely on good sample geometry.

Both types of equipment described herein for routine analyses can detect a content of 0.005% uranium in a 20-gram steel sample in a counting period of ten minutes. It is expected that if the uranium content of a sample is much above 1%, the self-absorption effects from this constituent will have to be corrected for. Such correction, however, has not been required for the samples received to date.

ACKNOWLEDGMENTS

Thanks are accorded to Mr. J.C. Ingles, Head, Control Analysis Section, and Mr. J.B. Zimmerman, both of the Extraction Metallurgy Division, for providing numerous samples and the results of their fluorimetric determinations. Thanks are also due Dr. G.P. Contractor, of the Physical Metallurgy Division, for providing the solid samples.

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APPENDIX 1

LIST OF SUGGESTED EQUIPMENT FOR THE BETA PARTICLE METHOD

(a) Probe and Ratemeter

Geiger-Müller tube:	Philips No. 18506*, end-window type, or Victoreen 1B85, axis parallel to surface of sample.
Ratemeter:	Nuclear-Chicago Corporation Model 1619A, or equivalent.

(b) Probe and Counter

Geiger-Müller tube:	Philips No. 18506*.
Scaler:	Nuclear-Chicago Corporation Model 151A, or equivalent.
Timer:	Nuclear-Chicago Corporation Model T 1, or equivalent. (Liebel-Flarsheim Model D 2)

(c) Shielded Assay Unit

Geiger-Müller tube:	Philips No. 18506*. (In some tests Anton No. 1001H was used.)
Scaler and timer:	As in (b).
Lead shield:	2 in. wall thickness; inner cavity approx. 3 in. x 3 in. x 5 in. high. Provide with a holder for the Geiger tube, and a tray system for samples. The shield should be lined with aluminum, copper, or brass.

* The Philips Type 18506 tube is suitable for use in a probe, as well as for mounting in a shield for assay work. A 10-megohm resistor must be connected in series with the anode, as recommended by the manufacturer. For a pellet 1.25 inches in diameter the sensitivity is 2000 counts per minute per per cent uranium, with a background rate (in a shield) of 13 counts per minute.

(d) Equipment for Preparing Samples
(also used in gamma method)

Hydraulic press, 24,000-lb capacity:	For example, the Carver Laboratory Press, Fred S. Carver Inc., 1 Chatham Rd., Summit, N.J.
Mould:	Carver or Buehler.
Plastic spray bomb:	Hughes-Owens "Crystal Clear".

Aluminum pie plates; fume hood; heat lamps or hot plate (optional).

APPENDIX 2

LIST OF SUGGESTED EQUIPMENT FOR THE (ROUTINE) GAMMA-RAY METHOD

(a)	Mounted sodium iodide crystal:	2 in. diameter x $\frac{1}{2}$ in. thick; Harshaw Chem. Co. No. 8D2.
(b)	Photomultiplier tube:	RCA type 6655.
(c)	Preamplifier:	Single-stage (anode follower for the 132-B Analyzer).
(d)	Single-channel analyser:	
	I. Pulse amplifier II. Single-channel discriminator III. Scaling circuit with mechanical register IV. Timer V. Regulated high-voltage supply VI. Cs-137 calibration standard (0.4 micro-curies, approx.)	I to VI supplied by Nuclear-Chicago Corporation as Model 132-B Analyzer-Computer.
(e)	Lead shield:	2 in. wall thickness; inner cavity, approx. 4 in. x 4 in. x 12 in. high; brass or copper lining, 1/8 to 1/4 in. thick; thin plastic tray above the crystal, to hold the samples; swinging or sliding door in the top of the shield.

APPENDIX 3

CALIBRATION OF THE GAMMA-RAY ASSAY UNIT

Two separate procedures are involved in ensuring that the gamma-ray counter will determine the uranium content of a sample:

(a) The relationship between the gain adjustment of the instrument and gamma-ray energy is determined so that the lower and upper limits of the required energy interval may be set.

(b) The physical conditions which affect the measured count-rate from a sample must be standardized so that a comparison of the count-rates of two samples will represent equally well their relative uranium content.

(a) Energy Calibration

The discriminator section of a single-channel analyser (see block diagram in Figure 5) usually has two controls: (1) a threshold, or base, control which permits the counter circuit to record electrical pulses whose amplitude exceeds V_B volts, and (2) a window-width control which rejects pulses whose amplitude exceeds $V_B + V_W$ volts. V_W is the window-width calibrated in volts. On the Nuclear-Chicago Analyzer the base control reads

0-1000, representating 0-100 volts; the window-width control is calibrated 0-10 volts. A standard calibration source, Cs-137, is supplied with this analyser, so that the gain of the photomultiplier tube and the gain of the pulse amplifier can be adjusted to have the base and window controls read known energy values. Cs-137 emits a gamma-ray of 662 keV energy.

To set the instrument to count between 65 and 185 keV:

(1) Set the amplifier gain to full (16X), the window-width to 10, and the base control to 502.

(2) With the Cs-137 sample near the detector, gradually increase the high voltage supplied to the photomultiplier tube from 500 V, until a peak counting rate is obtained. Suppose this happened near 730 volts, it would be advisable to take 1-minute counts at 726, 728, ------etc., until the maximum count was obtained. The base control now covers 0-1200 keV and the window control covers 0-120 keV above the base setting.

(3) Leaving the window control set at 10, reset the base control to 54 and remove the Cs-137 source.

(b) Calibration with Standard Samples
(This section is applicable to both beta and gamma methods)

Prepare a set of moulded 20-gram samples from drillings having known (chemically assayed) uranium content; two each of 0%, 0.02%, 0.05%, 0.10%, 0.35%, 1.0%, and 2.0% U are suggested as a convenient range of percentages.

Use the blank (0% U) samples to determine the background count-rate, counting for at least 10 minutes.

Count each side of the series of calibration samples, 5 to 10 minutes per side.

Subtract the background count and determine the net count-rate per minute for each sample.

Plot count-rate vs per cent U as a straight line through the origin and from the line determine count-rate per 1% U.

If it should prove necessary to measure samples whose masses differ from the usual 20-gram size, a range of samples weighing from 10 up to 50 grams may be moulded from a single bulk sample. The net count-rates obtained from these samples can then be used to produce a weight correction curve (Figure 12).

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JLH:(PES)DV

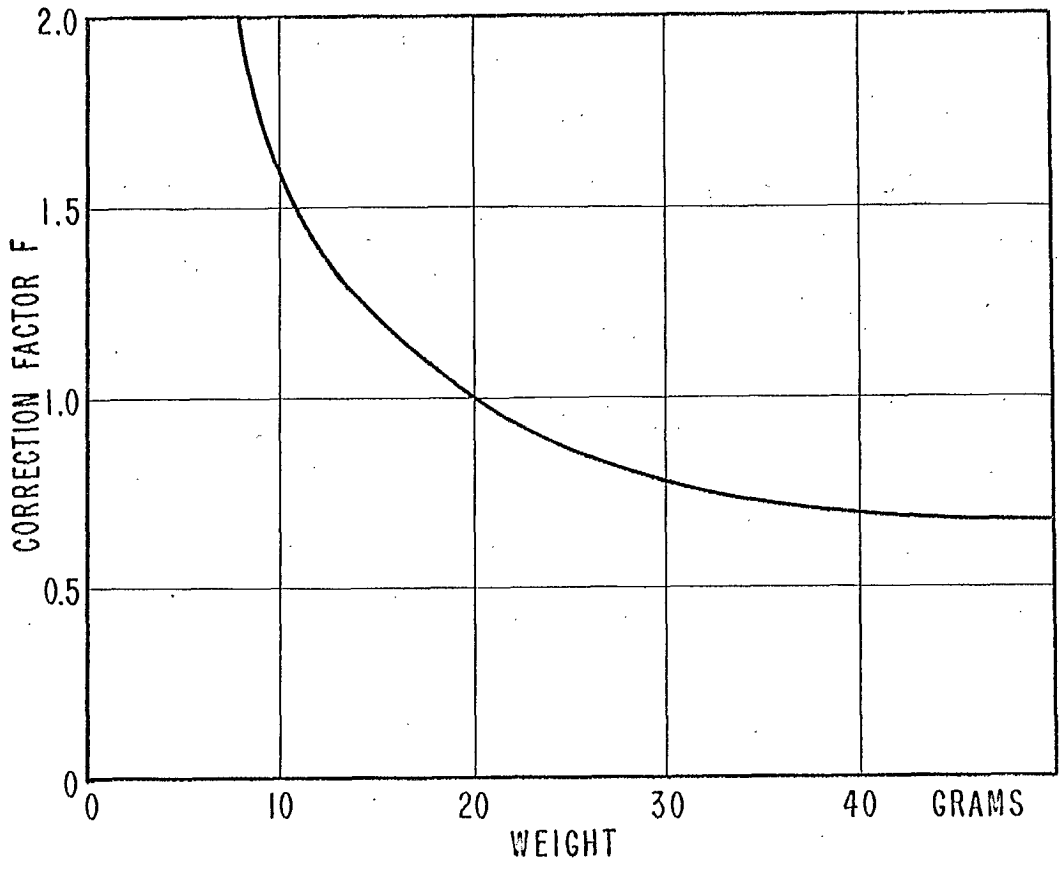

Figure 12 - Weight correction curve for pellets.

This is an empirical curve giving the required factors when the equipment is calibrated for 20-gram pellets.
Example: Suppose a pellet weighs only 16 grams.
Determine "per cent U" as in the case of a 20-gram pellet, subtracting the background as usual; then read the value of F corresponding to 16 grams, i.e. 1.16, and multiply by this factor to correct for the weight deficiency.

The reader is cautioned to determine his own curve for this purpose, because of possible variations in sample-detector geometry.