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USE OF A HIGH-PRESSURE IONIZATION CHAMBER IN ASSAYING UNCRUSHED ORE SAMPLES

by

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Radioactivity Division

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GENERAL

A method for the preliminary analysis of uncrushed ore samples for radioactivity is desirable for the ready elimination of specimens of no value with a minimum of sample preparation. Only gamma radiation has sufficient penetrating power to allow an assay of particles larger than very coarse sand. There are three possible types of gamma radiation detectors, namely, the high-pressure ionization chamber, a scintillation counter preferably arranged with coincidence circuits, or a geiger counter arranged similarly. The ionization chamber has proven to be the most effective, within limitations.

Although the scintillation counter seems to offer great promise, it is difficult to produce a large detector of this type. As a result, the number of counts from a given source diminishes rapidly with increasing distance from the scintillation crystal. This would be detrimental in the measurement of a large-sized sample. It would be necessary to use tray analysis with thoroughly mixed pulverized ore or to arrange about a large uncrushed sample a series of scintillation detectors, each with its own discriminator circuit.

A geiger counter with several tubes arranged in a coincidence circuit would diminish cosmic or other unwanted background radiation. It is doubtful, however, whether a group of geiger tubes could be used constantly for several months without change in sensitivity and the consequent need for recalibration.

THE HIGH-PRESSURE IONIZATION CHAMBER

In this chamber the number of ions, and hence the charge developed in the chamber, is proportional to the radiation intensity. The charge produced is measured by a direct-current amplifier or an electrometer. Commercially built electrometers are now available in which the drift or other instabilities are extremely low. The limiting factor with the ionization chamber is the fluctuation of the cosmic background. Shielding the chamber gives considerable improvement.

The present apparatus (Figures I and II) has been in daily use for the past twelve months. A block diagram of the circuit is given in Figure III. Prior to the arrangement of this apparatus several experimental ionization chambers were constructed using gas cylinders as casings. They were filled to a pressure of 1100 pounds / square inch with nitrogen, and, when it was established that no leaks were present, with argon. D-C amplifiers with electrometer tubes in a balanced input circuit functioned satisfactorily, although there was a slow drift as the batteries aged. It was shown that it would be possible to detect 0.05 per cent U_3O_8 (the minimum required) with approximately 200 grams of sample. Measurement of this percentage is now possible with 40 grams of sample.

The consistency of the present apparatus has been most gratifying. Over a period of one month the average variation in sensitivity was 0.5 per cent, the greatest departure being less than 2 per cent. The equipment remains on 24 hours a day, but is turned off on weekends, the required warm-up period being from one to two hours. Sensitivity is checked with a source made of powdered high-grade ore embedded in a thin layer of wax on a metal plate and protected by a paper envelope.

Designed and built by the National Research Council, Chalk River, Ontario, the chamber is a type T.P.J. (see report No. CR Tec-276 Design of the Chalk River Ion-Chamber, by H. Carmichael). The collecting volume is 996 c.c. enclosed in a cylinder 20 cm. long. The collecting electrode capacitance to the rest of the chamber is 28 m.m.f. The filling gas is argon of very high purity at a pressure of 600 pounds per square inch. As is shown in Figure III, the anode and cathode are both insulated from the wall, the latter serving as a guard ring and as an electrostatic shield. The leakage current is extremely small. Ionization currents below 10^{-14} amperes have been measured without difficulty. No change in sensitivity has been detected after a year's operation, as would occur if there were a leakage of the filling gas.

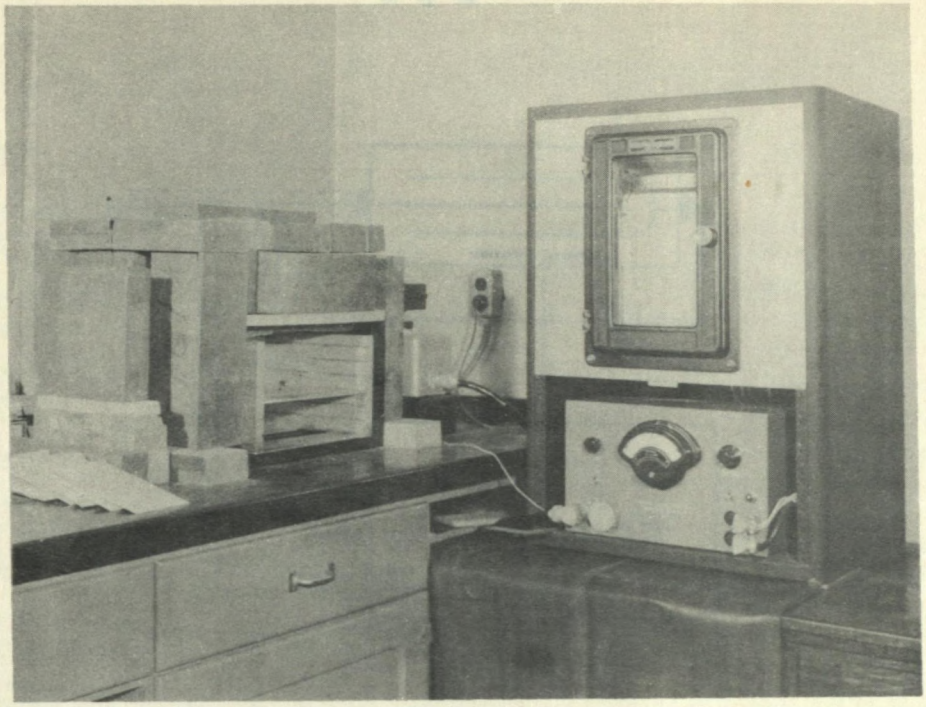

Figure I The complete apparatus; the lead castle shielding the chamber, the system of trays below the chamber, the head containing the vibrating reed condenser, the recording meter, and the main electrometer amplifier.

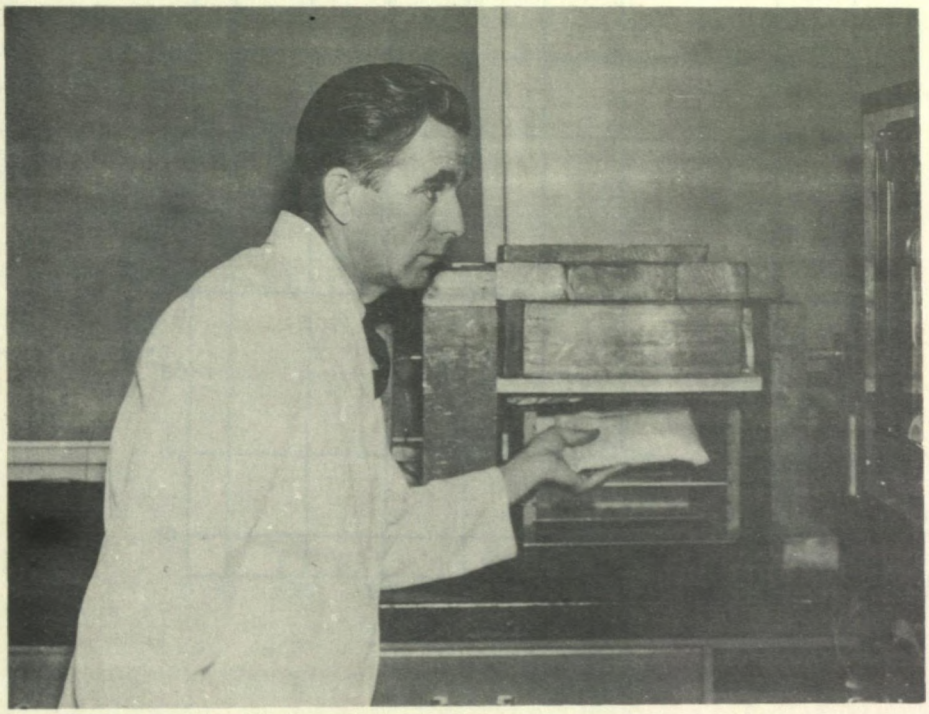

Figure II A sample is being placed on the tray in the D-position. Positions are A,B,C,D,E, and F each approximately one inch farther from the chamber.

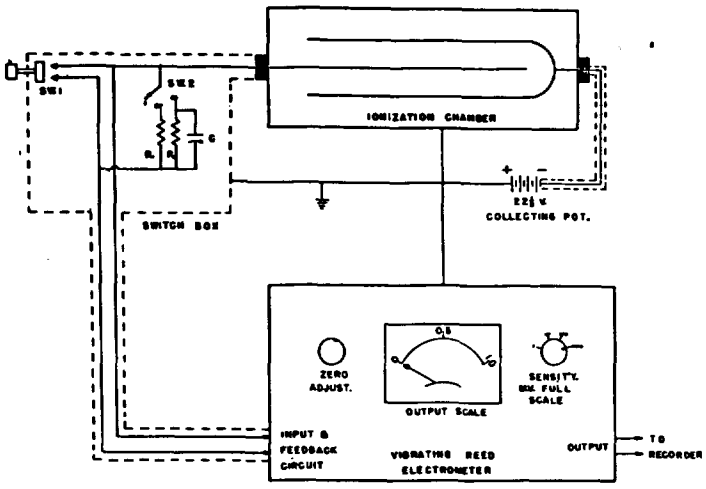

Figure III A block diagram of the apparatus, with the input circuit. $R_1 = 10^{11}$ ohms, $R_2 = 10^{12}$ ohms C = 10-100 mmf. Switch #1 operates for the charge time method when switch #2 is in position 1.

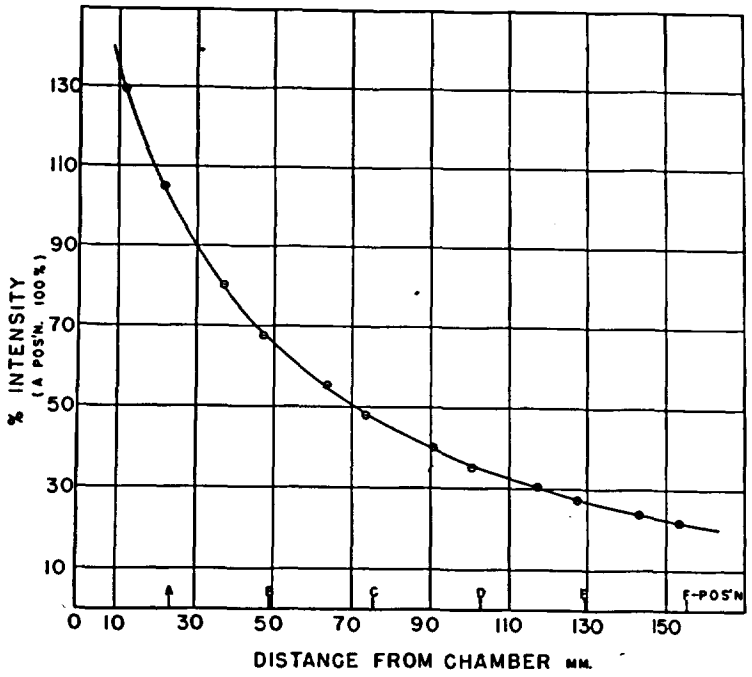

Figure IV A graph of the decrease in chamber output as a given source is placed in the various tray positions, A to F, away from the chamber.

With the chamber shielded above and around the sides by 7.5 cm. of lead, the background is reduced to about 40 per cent of the unshielded value and to 30 per cent of the unshielded value if 4 cm. of lead is used also beneath the chamber. Papers on cosmic rays state that about 10 cm. of lead will remove the low energy cosmic radiation, reducing the background to about 50 per cent, and that additional thicknesses of lead will not give significant improvement. The amount of lead used in this apparatus was limited by the strength of the floor.

Position of Tray and Sample versus Sensitivity

The system of trays under the chamber in the lead castle (Figures I and II) is used:

(a) to determine the relationship of distance versus sensitivity (Note the graph in Figure IV).

(b) to accommodate various sizes of ore samples.

(c) to allow for the placement of a weak sample very near the chamber and a strong one at a distance of several inches.

The anticipated decrease in sensitivity toward the edges of the tray was proven by using a "point" source of high-grade uranium ore. It was therefore necessary to place the samples in the same position on the tray and to use calibration standards of approximately the same size as the uncrushed samples.

This change in sensitivity over the tray surface is much less pronounced in the lowest position of the tray. It amounts to a decrease of about 20 per cent near the edge and only about 5-per cent in the area normally used. With a very large (2000-5000 grams) sample the operator increases the final result 1.05 to 1.20 times to reduce this error. This "sensitivity gradient" can be reduced also by placing a thin lead absorber against the chamber over the most sensitive part of the tray (a piece of lead 7.5 x 3.8 x 0.2 cm. gave some improvement). Placing the sample between two similar chambers should reduce the "change in sensitivity with distance" factor and render the placement of the sample and the correction for size and shape much less critical.

Although some gain in sensitivity could be achieved by placing small samples inside a hollow electrode within the chamber, the net advantage would be partly reduced in some circuits owing to the higher inter-electrode capacitance of this arrangement.

The Vibrating Reed Electrometer

The electrometer is shown in the cabinet below the recording milliameter in Figure I. The reed is in a separate head at the end of a three-foot cable, the head being connected to the chamber by the input circuit as shown in the block diagram. This circuit must be adequately protected from the effects of humidity. A tray containing a desiccant was placed in the case with the switches and the seams of the case were made airtight with melted wax. The condition of the desiccant was determined from the daily sensitivity test of the instrument, the sensitivity gradually decreasing as the desiccant became saturated. Even in very humid weather the drying agent did not require changing at less than six week intervals.

Briefly, the theory of operation of the vibrating reed electrometer is as follows: for a given amount of charge, Q coulombs, on the input circuit, the voltage V is dependent upon the capacitance, C farads; i.e. $Q = C \times V$. In this instrument C is varied sinusoidally at a constant frequency of about 1000 cycles/second. Thus V varies sinusoidally with its amplitude proportional to Q since the average value of C does not change. An A-C amplifier is used to amplify V. A detector, synchronized to the same frequency as the reed, provides an output voltage proportional to the charge Q on the input circuit. Four gain settings are provided whereby 1, 10, 100 or 1000 millivolts on the input circuit produces full-scale deflection of the output meter or of the recording meter. Only the 100- or 1000-scale settings are used with this chamber.

Systems of Measurement

The limit of detection is a function of the per cent U_3O_8 equivalent activity multiplied by the mass. Using the amount of lead shielding presently available with normal background fluctuations the limit of measurement is given by the following approximate rule:

$$\text{Per cent} = \frac{2}{M} \quad (M \text{ less than } 300 \text{ grams}) \qquad \text{or}$$

$$\text{Per cent} = \frac{6}{M} \quad (M \text{ greater than } 300 \text{ grams})$$

The limit of detection is sufficiently low for uncrushed hand samples to allow for the selection of all worthwhile specimens. A gamma measurement which proves to be much higher than the subsequent beta assay, when the two assays are given in terms of per cent U_3O_8 equivalent activity, suggests the presence of thorium. A more precise measurement of thorium and uranium content can be made using powdered ore with tray analysis.

Two systems of measurement are used:

(1) the Deflection Method - This method is used for very strong samples or where a continuous record of the sensitivity of the apparatus is required. A resistance of either 10^{11} or 10^{12} ohms is placed in the input circuit. The recording meter then gives a continous graph of the current flowing.

(2) the Charge Time Method - This method is used for rapid screening of samples where it is possible to measure thirty or more per hour, or where the sample is very weak. Down to 1/10 the strength of the background radiation has been reliably detected. The rate of charge built up in the input circuit may be measured by timing the rate of movement of the needle on the output meter between two convenient points on the scale.

Calculations are based on the following formulae:

$$Px = Ps \cdot \frac{Ms}{Mx} \cdot \frac{Rx}{Rs} \cdot K = \frac{K}{Mx} \cdot \frac{Rx}{S}$$

where:

Px..... per cent U_3O_8 equiv. of the unknown sample

Ps..... " " " " " standard

Mx..... mass of sample

Ms..... " " standard

Rx..... reading obtained for sample (see below)

Rs..... " " " standard (" ")

K..... correction factor based on differences in size and shape of sample and standard; also includes added percentage to compensate for absorption in the larger samples. K approaches unity where samples and standards are powdered and measured in the same container.

S..... Sensitivity number determined for a given tray position; it is a combination of Ps, Ms, and Rs. Several standards are used to obtain a good average value.

For Deflection Method using recording meter:

Rs = defection due to standard minus that due to background

Rx = defection due to sample minus that due to background

For Charge Time Method using stop watch:

$$Rs = \left(\frac{1}{T \text{ standard}} - \frac{1}{T \text{ background}} \right)$$

$$Rx \left(\frac{1}{T \text{ sample}} - \frac{1}{T \text{ background}} \right)$$

where T = seconds and $\frac{1}{T}$ = reciprocal of charge time.

Comparison of Methods

The deflection method with the recording meter is useful where a long measurement must be taken such as a survey of the changes in background or to show build up of decay products in a sample which is not in equilibrium. Little attention is required on the part of the operator, and the calculations are reduced to a minimum. The charge time method, however, is commonly used for screening samples because of its rapidity and its more accurate measurement of very low activities.

Compare the readings taken by both methods in the recordings shown in Figures V, VI, VII. As the charge time varies inversely with the radiation intensity, the reciprocal of the charge time of a sample minus the reciprocal of the

The chart readings reproduced below were made using an input resistance of 10^{12} ohms. The full-scale reading was 100 millivolts, or 2 millivolts per scale division.

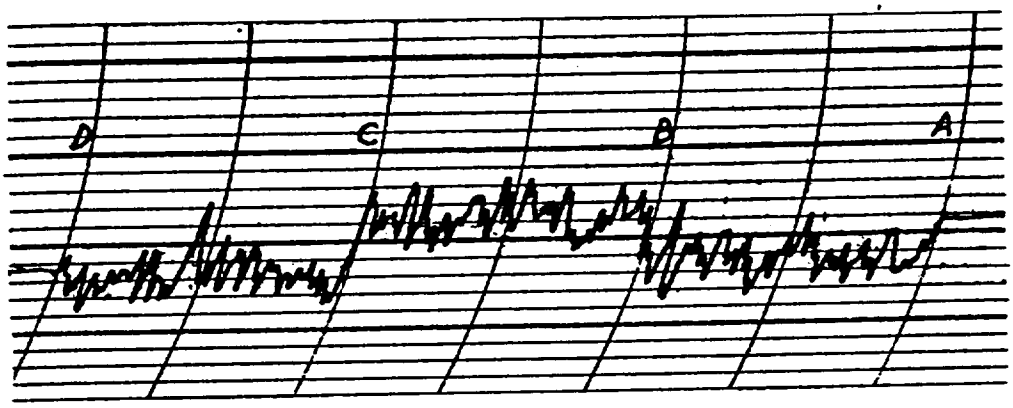

Figure V - Between A and D the apparatus recorded background intensity, except between B and C where there is the additional radiation from 150 grams of .0268 per cent U_3O_8 in the (A) tray position nearest to the chamber.

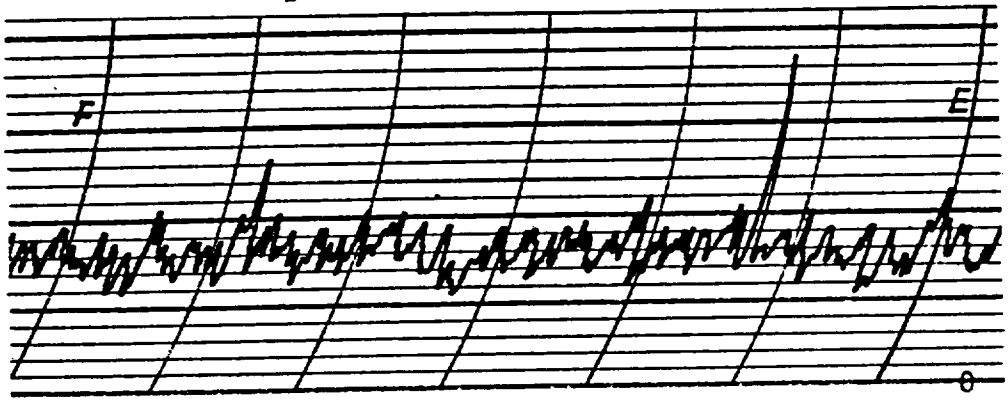

Figure VI - A forty-five minute background recording is shown between E and F. Note the cosmic ray burst.

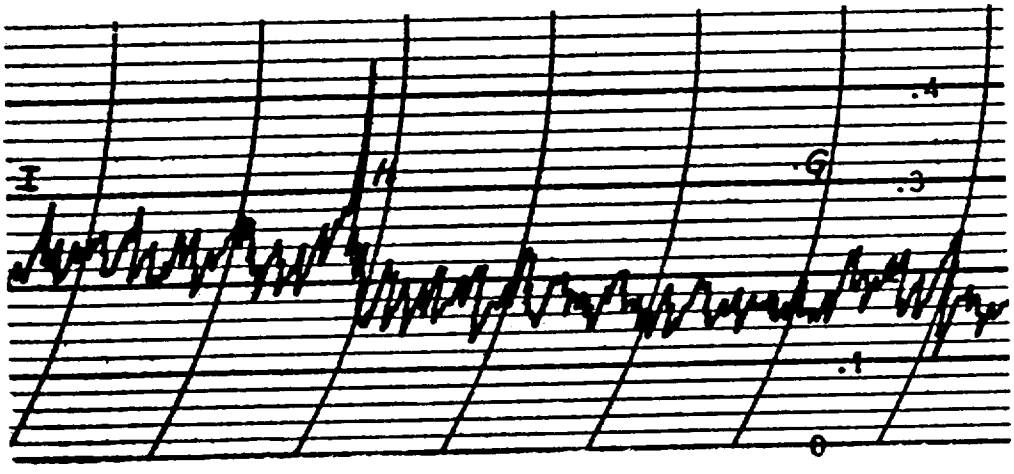

Figure VII - A further background reading is shown from G to H. The 1-inch lead floor below the chamber was removed between H and I with a resulting increase in background. Note the cosmic ray burst near H.

background charge time is a measure of the sample's activity. The resultant linear relationship would not have existed over the lower ranges had the leakage current been appreciable. The charge time results are shown in the last three sets of readings in Table IV.

Determination of Collecting Potential

Tests showed that a collecting potential of 22.5 volts is sufficiently large.

Table I

Determination of Collecting Potential

Collecting Potential (Volts, negative)	Ionization Current - Per Cent (Current at 22.5 volts = 100%)
22.5	100
12	100
6	100
3	98
1.5	90
1	83
.5	65
.3	47
.2	34
.1	14

Tests for Linearity

Prior to calibration the apparatus was tested for overall linearity using two methods:

(1) Known standards of various activities each of which was well mixed and contained 150 grams were measured in identical positions with respect to the ion chamber. The deflection per gram per 1.00 per cent was calculated for each standard. This latter quantity was found to be constant.

(2) With this method the values of the sources do not need to be known with much accuracy. The chamber is subjected to the required range of radiation intensities by placing source #1 at various distances from the chamber. By bringing source #2 from a "distant" point to any suitable point an increment may be added to each of the intensities. By observing that this increment remained constant at all points on the scale of the output meter, the apparatus was found to be linear.

Absorption effects were estimated by placing a small, strong, (point) source beneath various absorbing materials. The results outlined below are not entirely accurate because the rays from the source were not strictly parallel, and because of scattering from the lead shielding. The gamma rays were those produced by uranium ore.

Table II

Absorption Effects Due to Lead, Rock, Water

Material	Thickness required to reduce radiation by one-half
Lead	1.0 cms.
Metamorphosed sediment	7 cms.
Water	16 cms.

Scales were constructed to be held beside the sample to give directly the correction factor for both the approximate absorption and the fact that a large sample resting on a tray was closer to the chamber than a small one. When making an estimate by this means, some experience is required on the part of the operator to enable allowance for points and hollows on an irregular piece of ore. Each sample is turned to compensate for uneven distribution of active portions in the specimen. Here again the use of two chambers could make this operation much less critical.

The values of this preliminary analysis agree within ±5 to 10 per cent with the beta and chemical assays.

Table III

U_3O_8 Equivalent Activity Determined by Three Methods of Analysis

Gamma (Ionization Chamber)		Beta (Geiger Counter)	Chemical
Uncrushed	Crushed		
Per cent	Per cent	Per cent	Per cent
	.085	0.083	0.10
0.15		0.144	0.119
0.16		0.157	0.147
0.21		0.177	0.17
0.21		0.197	0.178
	0.25	0.202	
	0.28	0.257	
0.30		0.284	
	0.40	0.409	
0.69		0.615	
	0.80	0.80	0.80
0.82	1.0	1.03	1.17
	1.76	1.65	
	x1.92	1.40	0.77
	x2.13	1.28	0.50
—	2.1	2.25	2.15
5.6		5.75	
	5.97	6.28	5.48
7.1	—	7.09	6.09
	11.56	10.03	10.6
	16.51	14.2	15.6
	16.82	13.19	16.6

x The presence of thorium is indicated. The method of calculating the relative thorium to uranium content will be dealt with in a later report.

Readings and Calculations

All data were recorded on a printed form to speed up calculations. A sample form of typical readings and calculations is shown in Table IV.

TABLE IV.

Typical Readings and Calculations

Source		BKgnd	H-P		BKgnd	G902		BKgnd	.027%		BKgnd	.027%		BKgnd	.027%
Weight Gms.						732			150			150			150
Thickness						.85									
Position			D			D			A			A			A
Scale		M	M			M		C	C		C	C		C	C
Sensitivity			348			348			1060			1060			1060
Turned						*									
R	1	560	43.3		541	83.6		58.8	52.5		55.4	44.3		64.5	43.8
E	2		43.7			84.6		57.0	46.7		59.0	44.1		59.8	46.8
A	3		44.7			82.0		66.1	49.7		58.6	47.2		59.5	47.9
D	4		43.1					57.7	50.2		67.9	48.8		56.4	45.1
I	5		42.7			83.0*		62.1	45.4		59.0	47.1		57.6	50.8
N	6		44.3			80.6*		66.3	48.4		60.4	47.9		55.7	43.5
G	7		43.3			81.6*		53.4	48.6		54.6	47.7		56.4	49.5
S	8	576	42.9		533			57.0	45.7		63.8	54.2		58.6	48.6
Total		1136	348.0		1074	495.6		462.8	387.2		478.7	381.3		468.5	376.0
Average		568	43.50		537	82.6		60.43	48.40		59.84	47.66		58.56	47.00
Reciprocalx10^5		176	2299		186	1211		165.5	206.9		167.1	209.8		170.7	212.8
			2123			1025			41.9			42.7			42.1
Percent						0.34%									
Remarks		Sensitivity			Unknown	Sample		Low Activity		Sample Measured		on Three Different Days			
		= 348.2	x $\frac{2123}{2124}$												
		= 348													

The daily sensitivity measurement is shown in the first two columns of Table IV. The number 348 was determined for use in the day's calculations. It is 10^5 x reciprocal of charge time in seconds of an average standard minus the reciprocal of the background charge time, where the M(1000 millivolt) scale is used with a 1.00 per cent U_3O_8 standard of mass 100 grams in the D position. The actual standards used were 150 grams each and ranged in strength from 0.5 to 14 per cent. The resulting average was 348.2 per 100 grams per 1.00 per cent. A determination six weeks earlier was 348.1 using the same standards. The sensitivity standard H-P was run at the time of calibration. Its reciprocal was 2124. This standard is used for daily performance checks.

The next two columns show the reading taken for a solid specimen #G902 weighing 732 grams. The thickness correction .85 was read from a scale held beside the sample.

$$\text{Per cent } U_3O_8 \text{ equiv.} = \frac{\text{Thickness factor}}{\text{Mass in hundreds of grams}} \times \frac{\text{corrected reciprocal}}{\text{sensitivity}}$$

$$\text{for G902 Per cent} = \frac{.85}{7.32} \times \frac{1025}{348} = 0.34 \text{ per cent}$$

The last three determinations were made using a standard of low activity and illustrate the method followed with samples just measureable above the background. The C (or 100 millivolt) scale was used and eight readings were taken alternately for both sample and background. A simple average only was found sufficient. However, an occasional cosmic burst may be indicated by a reading significantly lower than the others in which case it is replaced by an additional one.

The lowest current that it has been possible to detect is calculated below to determine the feasibility of using the relatively inexpensive electrometer tubes in place of the dynamic condenser electrometer.

$$\frac{DQ}{DT} = C \frac{DV}{DT}$$

where $\frac{DQ}{DT}$ = the rate of change of charge on the input circuit expressed in coulombs per second.

C = the capacity of the input circuit in farads = approx. 50×10^{-12}

$\frac{DV}{DT}$ = the rate of change of potential in the input circuit in volts per second.

$\frac{DQ}{DT}$ = the ionization current, I amperes.

$\frac{DV}{DT}$ = 1.00 volts per 600 seconds for average background radiation.

$\therefore I = 50 \times 10^{-12} \times \frac{1}{600}$ = approx. 8×10^{-14} amperes.

Ionization producing a change in background current of 1/16 is generally detectable.

$\therefore$ I (Minimum detectable) = 5×10^{-15} amperes.

The useful current range of electrometer tubes is determined at the lower limit by the amount of grid current flowing. This latter quantity is about 10^{-14} amperes for triode electrometer tubes and about 10^{-15} amperes for the tetrode types. The ionization currents from this chamber can therefore be measured with the most sensitive electrometer circuits.

Conclusions

The ionization chamber, once set up, has proven capable of consistently reliable results within the limitations imposed by cosmic radiation. One thousand samples have been analysed without interruption in a six months' steady performance. A single complete calibration has been sufficient during this time. Room temperature variations have had no observable effect. The stop watch used should be of good quality as it is subjected to several thousand operations per month.

ACKNOWLEDGMENTS

The authors wish to express their thanks to Dr. F.E. Senftle, formerly of the Mines Branch and now at the Massachusetts Institute of Technology, Boston, U.S.A., who suggested the use of the high-pressure ionization chamber and dynamic condenser electrometer for this work.

APPENDIX

Reporting of Uranium or Thorium Determinations

Under the Atomic Energy Regulations of Canada the results of an assay or analysis of a mineral that indicates a content of more than 0.05 per cent by weight of uranium or thorium are to be reported forthwith to the Director of the Geological Survey, Department of Mines and Technical Surveys, Ottawa, for the Atomic Energy Control Board, together with full particulars relating to the material assayed or analysed, including the name and address of the person from whom such material was received, the purpose of the assay or analysis, and the origin of the material so far as known to the person making the report. This requirement does not apply to assays or analyses made for persons operating under orders of the Board which provide for periodical reports.

Copies of the Regulations may be obtained on application to the Secretary, Atomic Energy Control Board, Ottawa.