

**GEOPHYSICAL APPLICATIONS**

**OF**

**RADON MEASUREMENTS**

**Benjamin Holzman**

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TABLE OF CONTENTS

	Page
I. Abstract	I
II. Acknowledgements	II
III. Introduction	III

Part I.

IV. Methods of standarization for radium determination	
apparatus	1
A. The use of a radium standard	3
(1) Gamma ray method	3
(2) Emanation method	3
(a) Instability of radium solutions	4
(b) Liberation of radon from solutions	5,70
B. The use of uranium minerals	5
(1) Theoretical consideration of Ra/U ratio	5
(a) Conditions affecting Ra/U ratio	6
(2) Pitchblende	7
(3) Carnotite	8
(4) Emanating power	9
C. The use of the physical constants of the instruments	15
(1) Theoretical ionization for one curie of radon	15
(a) Initial ionization	15
(b) Maximum ionization	15
(2) Ionization in a gas	18
(a) $I = C \frac{dy}{dt}$	18
(b) Capacity determinations	20

\* Fig. 28, p. 76 has  
2 related maps of areas  
where surveys were made.

TABLE OF CONTENTS See pocket at end of Thesis

	page
(3) Duane and Laborde formula-----	22
(a) Theoretical consideration-----	25
(b) Applications-----	27
(c) Dependence on size of chamber-----	28
(a') Radon collecting equalizer-----	29
(b') Tap water as a source of radon-----	32
(d) Effect of saturation-----	39
(a') Calculating saturation effect-----	47
(e) Modified Duane and Laborde formula-----	52

### Part II.

V. Theory underlying geophysical applications of radon measurements-----	56
A. Radon and oil bearing region-----	60
B. Diffusion of radon-----	62
C. Radon and faults-----	64
(1) Magmatic influences-----	67

### Part III.

VI. Radon content of underground waters and its relation to the Raymond fault-----	69
A. Technique for the measurement of radon in solutions-----	69
(1) Theory of removal of radon from solutions-----	70
B. Raymond fault-----	74
(1) Radon content- surveys-----	74
VII. Conclusions-----	79

## ABSTRACT

In recent literature statements to the effect that a fault will be evidenced by a higher radon concentration in the overlying soil, that the radon content of soil in an oil bearing region is higher, and that soil formations appear to have more or less characteristic radon contents, have appeared.

Since radioactive measurements are concerned with extremely small quantities of the order of  $10^{-12}$  grams of radium per gram of rock, a study of the methods of standardization for radon apparatus is presented in an attempt to indicate the requirements for accurate and precise measurements. The use of a standard radium solution, uranium minerals, and the Duane and Laborde formula are considered.

An analysis of the theory underlying the geophysical applications suggested above indicates that soil formations are not expected to have a more or less characteristic radon content, that a high radon concentration in the overlying soil of an oil bearing region cannot be correlated genetically with the oil, and that it is possible to have a high radon concentration at a fault.

A simplified technique for the measurement of radon in solutions adaptable to field work is demonstrated. Surveys of the radon concentration of underground waters of Pasadena, indicated no systematic increase towards the Raymond Fault.

## ACKNOWLEDGEMENTS

I wish to express my thanks to Mr. R. Engel for introducing me into the field of radioactivity, and for his many valuable criticisms. Mr. Engel's active interest in the application of chemical and physical data to geology has been an actuating factor throughout this entire research .

The section of the paper on "Methods of Standardization for Radium Determination Apparatus" has been directly due to my work with Dr. R.D. Evans and to whom I owe many thanks for gaining a knowledge of the fundamentals of precise measurements in radioactive work.

I acknowledge my sincere indebtedness to Mr. R.W. Raitt for his ever helpful suggestions and unwearied assistance throughout this study. As a result of Mr. Raitt's analysis of the theory of removal of radon from solutions, has been developed a simplified technique for the measurement of radon in solutions adaptable to field work.

## INTRODUCTION

The relation of radioactivity to geology has in recent years been a subject of considerable interest. To such men as J. Joly, A. Holmes, T.C. Chamberlain, J. Barrell, L.H. Adams, and others who are want to delve into the intricacies of petrogenesis and physics of the earth, radioactivity offers a wealth of energy. Their views, regardless whether or not they are to be considered as precocious, have been received with zest in the geological realm and have invited a sincere effort for the search and study of corroborative data.

Of a more practical aspect has been the attempt to apply radioactive methods to geophysical prospecting, and this paper is presented as an effort to indicate the geological significance of these types of measurements.

The report has been divided into three sections. The first has to do with methods of standardization for radium determination apparatus and has been the result of a desire to obtain precise and accurate measurements; the second is concerned with a theoretical consideration of the principles underlying the geophysical applications of radon measurements; and lastly, the results of an investigation of the radon content of underground waters and their relation to faults are presented.

**Part I.**

**METHODS OF STANDARDIZATION FOR RADIUM DETERMINATION APPARATUS**

## INTRODUCTION

In all methods for the determination of radium the actual quantity of radium present is entirely dependent upon an accurate calibration of the electrometer by a radium standard. An International Radium Standard was adopted in 1910 at Brussels<sup>1</sup> wherein a quantity of radium in the form of  $\text{RaCl}_2$ , was carefully determined by analytical methods, and by which other radium containing substances may be standardized. Unfortunately at the present time no such absolute radium standard is available in this country.

With the large number of publications pertaining to the field of radioactivity, it becomes highly desirable that in reviewing the literature one may do so with a certain degree of surety that the various experimental data are comparable. This gains added importance in applying radioactivity to geology. Thus, when information concerning the radium content of rocks and waters are set forth in the literature, the intrinsic importance of such analyses lies not in the relative value, but in the absolute value. The relative value may have served the author's purpose, who, because of the localism of his problem may have derived justifiable conclusions; however, for a universal application the numerical data will have no significance.

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1

Rutherford, Chadwick, Ellis: Radiations of Radioactive Substances, 1930.



The relation of radioactivity to the earth's thermal history has suggested numerous theories of which a few, while theoretically sound, are perhaps premature and too positively asserted, since experimental evidence has yet to corroborate them. Not without due reason does A.C. Lane<sup>1</sup> state, "The uncertainty as to the average amount of radium in rocks is still of the order of 100 to 200 per cent." This of course is primarily due to a lack of data but an important factor contributing to the uncertainty is also due to the technique of measurement in which limitations of both gamma ray and emanation methods for the determination of radium become efficacious; pertaining to technique, standardization is of utmost importance.

The following study is presented to indicate the various precautions that one should observe in standardizing radium determination apparatus.

Essentially there are three procedures that one may follow, (a) The use of a standard that has been checked by an absolute standard, (b) The use of uranium minerals, (c) A calibration based on the physical constants of the instrument, i.e., capacity, voltage, etc.

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1

Report of the Committee on the Measurement of Geologic Time--  
A.C. Lane, chairman, April, 23, 1932 pp. 21

## METHODS OF STANDARDIZATION

## A. The Use of a Radium Standard

In the gamma ray method, if a known amount of a radium salt is available, the procedure for standardization offers no difficulties. The salt is usually in the form of a halide, although there has been an attempt to use a  $\text{RaBaCO}_3^1$  as a secondary standard. For the calibration of the instrument, the salt is placed in the proper position in the gamma ray set up and the rate of discharge is then observed.

The standardization in the emanation method, however, offers certain difficulties. As a rule the standard is in the form of a solution of known radium concentration. Since this method depends solely on the alpha ionization due to the gaseous emanation of radium, radon, it becomes necessary for an accurate calibration to free the entire or known amount of the radioactive gas from the solution.

It is now known that standard radium solutions are unstable unless protected by barium in an acid solution<sup>2</sup>. The instability has been clearly demonstrated in the literature when Rutherford and Boltwood<sup>3</sup> first presented the Ra/U ratio determined in uraninite from North Carolina as  $7.4 \times 10^{-7}$ . The following year they<sup>4</sup> found that a serious

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<sup>1</sup>  
M. Yovanévitch and M. Chamie: *Comp. Ru.* 266-268, (1922)

<sup>2</sup>  
J. Meran: *Trans. Roy. Soc. (Canada)*, 10, 77-84, (1916)

<sup>3</sup>  
Rutherford and Boltwood: *A.J.S.*, (4) 20, 55-6 (1905)

<sup>4</sup>  
Rutherford and Boltwood: *A.J.S.*, (4) 22, 1, (1906)

error had been made in the value of the radium standard solution caused by partial precipitation of the radium and accordingly the value of the ratio was changed to  $3.8 \times 10^{-7}$ . This involved an error of nearly 100 per cent.

The U.S. Bureau of Mines<sup>1</sup> advocates the following, "The latest practice in the employment of radium solutions as a standard indicates by a large excess of barium and by at least 5 per cent of halide acid, corresponding to the employment of the halide salt of radium, there is no diminution in the quantity of emanation that can be extracted from it."

In reference to the latter it is important that no sulfate ions be present for  $\text{RaSO}_4$  has the property of occluding radon.<sup>2</sup>

For the liberation of radon a frequent procedure has been to boil the solution. As elsewhere demonstrated, from a theoretical consideration such a procedure may involve a serious error if the liberation of radon is assumed complete. Since radon obeys Henry's law, merely boiling only changes the concentration to the gas between the two phases corresponding to a new distribution ratio at the temperature of the boiling solution. Thus at  $100^\circ\text{C}$ . where the distribution  $\frac{\text{(conc. in sol.)}}{\text{ratio (conc. in air.)}} = 0.107^3$ , unless the effective volume

1

U.S. Bureau of Mines, Bulletin 212

2

W.D. Urry: He and Ra content of some rocks of the Keweenaw series in Mich.- Report of Committee on the measurement of Geologic Time, April 23, 1932

3

St. Meyer and E. Schweidler: Radioaktivitat pp. 411.

above the solution is considered and a proper correction made for the amount of radon retained by the solution, an error of approximately 10 per cent may be made in the standardization. The magnitude of this error only applies to those methods which rely upon boiling for the liberation of radon.

#### B. The use of Uranium Minerals

If  $A \rightarrow B \rightarrow C \dots$  represent a radioactive disintegrating series, and  $N_a, N_b, N_c$ , etc., the total respective number of atoms in equilibrium, then,

$$\lambda_a N_a = \lambda_b N_b = \lambda_c N_c = \dots$$

where  $\lambda$  represents the total number of atoms which break up per second, or if  $N$  is the number of atoms present, and  $n$  of these disintegrate per second then  $\lambda$ , the disintegration constant =  $\frac{n}{N}$ .

Since  $1/\lambda$  may be considered the period of probable life of any radioactive substance which in turn is proportional to its half-life value, the following may be written

$$N_a : N_b : N_c : \dots = 1/\lambda_a : 1/\lambda_b : 1/\lambda_c : \dots =$$

$$L_a : L_b : L_c = T_a : T_b : T_c$$

Where  $L$  = probable life value

$T$  = half-life value

and are related by  $L = 1.443T = 1/\lambda$ <sup>1</sup>

Thus, when in equilibrium, the amounts of the different radioelements in a series are proportional to their half-life periods.

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<sup>1</sup>

A. Helms: Radioactivity and Geological Time  
National Research Council-Age of Earth, pp 130.

If now an uranium mineral is available in which the conditions throughout disintegration were<sup>d</sup> such as to insure equilibrium conditions, a careful analysis of the uranium content would immediately give the radium content. The conditions imposed upon the mineral involve its age and a geochemical consideration.

The time element is important in as much as the Ra/U ratio deviates considerably from the theoretical value in relatively recent minerals. There is a certain minimum age at which the Ra/U ratio becomes constant. See Fig. 1. In fact the deviation from the theoretical ratio has been made use of in age determinations of recent minerals whose life has attained the magnitude of only thousands of years. The Ra/U ratio becomes constant at approximately  $10^6$  years.

The geological environment may materially affect the Ra/U ratio. In regions where geochemical processes are active, replacement, alteration, impoverishment, and enrichment may all conceivably destroy equilibrium that time may have established between the uranium and radium. Uranium and radium may be both leached or secondarily enriched and in consequence an uranium analysis will not give the radium content since the Ra/U ratio will be unknown.

The Ra/U ratio in pitchblendes has been variously reported. Rutherford and Beltwood<sup>1</sup> reported the value as  $3.6 \times 10^{-7}$ .

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<sup>1</sup>

Rutherford and Beltwood: A.J.S., 22, 1, (1906)

$t$ (years)	$\frac{10^{-7} \text{ gm. Ra}}{1 \text{ gm. U}}$	$\left(\frac{R_a}{U}\right)_{t=0}$ $\left(\frac{R_a}{U}\right)_{t=\infty}$ EXPRESSED IN PER CENT
0.	0.0	0.0
100	.000095	0.0028
500	.00183	0.054
1,000	.0060	0.176
2,000	.0205	0.604
5,000	.0908	2.67
8,000	.1752	5.15
10,000	.2330	6.85
15,000	.3763	11.07
20,000	.5139	15.11
30,000	.7714	22.68
40,000	1.0055	29.57
50,000	1.2188	35.84
60,000	1.4132	41.53
70,000	1.5903	46.76
80,000	1.7514	51.50
90,000	1.8986	55.81
$1 \times 10^5$	2.0323	59.76
$2 \times 10^5$	2.8624	82.25
$3 \times 10^5$	3.1891	93.77
$4 \times 10^5$	3.3176	97.55
$5 \times 10^5$	3.3681	99.036
$6 \times 10^5$	3.3880	99.621
$7 \times 10^5$	3.3958	99.851
$8 \times 10^5$	3.3988	99.941
$9 \times 10^5$	3.4000	99.977
$10^6$	3.4005	99.991
$\infty$	3.4008	100

Fig. 1

Showing the dependence on time for the Ra/U ratio.

Starting with one gram of pure uranium, the

amount of radium produced increases with time

until  $10^6$  years. The theoretical Ra/U ratio equals

$3.4 \times 10^{-7}$  and is equal to a ratio of the half-lives of

radium and uranium.

Boltwood<sup>1</sup>, however, after redetermining the uranium content of the standard pitchblende (for which the value  $3.8 \times 10^{-7}$  was reported) lowered the value to  $3.4 \times 10^{-7}$ . Later in 1910, after the adoption of the International Radium Standard, Rutherford<sup>2</sup> made a comparison with his previously used radium standard, which indicated a change from 3.4 to  $3.23 \times 10^{-7}$  for the Ra/U ratio. In 1913 Heimann and Marckwald<sup>3</sup> examined eight specimens of pitchblende from the principal uranium deposits of the world and found the ratio to be constant within 0.4 per cent and reported the absolute value as  $3.328 \times 10^{-7}$ . Becker and Jannasch<sup>4</sup> reported an average value of  $3.399 \times 10^{-7}$ , obtained at the Radiological Institute at Heidelberg, using a standard radium salt from the Institute of Radium Research in Vienna. Lind and Roberts<sup>5</sup> using pitchblende from Colorado, state the following, "The results give as final average of 18 determinations  $3.40 \times 10^{-7} \pm 0.03$  for the ratio Ra/U."

With respect to the preparation of a standard radium solution from pitchblende, Lind and Roberts<sup>6</sup> cautions one of the sulfur impurities in the ore which enhances the difficulty of keeping radium in solution. This is probably due to the sulfate that may be formed in an oxidizing solution.

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1

Boltwood: A.J.S., 25, 296, (1908)

2

Rutherford: Phil. Mag., (6) 28, 320-7, (1914)

3

Heimann and Marckwald: Jahrb. Radioakt. Elektronik, 10, 299-323, (1913)

4

Becker and Jannasch: *ibid.*, 12, 1-34, (1915)

5

Lind and Roberts: J. Am. Chem. Soc., 42, 1170-7, (1920)

6

Lind and Roberts: J. Am. Chem. Soc., 42, 1170-7, (1920)

Another uranium mineral which has been used for standardization purposes is carnotite. Carnotite, whose formula corresponds to  $K_2O \cdot 2U_2O_3 \cdot V_2O_5 \cdot 3H_2O$ , occurs as a yellow crystalline powder, or in loosely cohering masses, intimately mixed with quartzose material, and is probably a secondary enrichment of an unknown vanadium mineral<sup>1</sup>. From the nature of its environment and its secondary affinities it might be justly concluded that the conditions under which the uranium disintegration may have taken place were variable and consequently no constancy in the Ra/U ratio should be expected. This is more or less the case. M. Curie<sup>2</sup> cites values for the Ra/U ratio in carnotites between  $2.3 \times 10^{-7}$  and  $3.6 \times 10^{-7}$ . Lind and Whitmore<sup>3</sup> found that in hand specimens up to a few pounds, the Ra/U ratio varied as much as  $2.48 \times 10^{-7}$  to  $4.6 \times 10^{-7}$ , but that in well sampled carload lots, a few hundred pounds to several tons, the ratio remained constant.

Because of its secondary nature there is also to be considered the time element upon which the value of the ratio is dependent. Since the minimum time at which the Ra/U ratio becomes constant is  $10^6$  years, the error involved in the standardization by using a relatively recent mineral may be considerable. See Fig. 1.

A very interesting property of uranium and radium minerals is their emanating power. The emanating power is defined as the fraction of the equilibrium quantity of radon that is associated

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<sup>1</sup> Lindgren: Mineral Deposits pp. 460

<sup>2</sup> M. Curie: Le Radium et les Radio-Elements, Bailliere, Paris pp. 147 (1925)

<sup>3</sup> Lind and Whitmore: U.S. Bureau of Mines, T.P. 88, (1915)



with the radium (time for this equilibrium approximately 30 days) which is given up to the air at specified conditions. The interest attendant to the emanating power of uranium minerals can be readily understood, for if the emanating power is constant under given conditions and is of the proper magnitude, a known amount of radon, assuming the theoretical Ra/U ratio, can be used for standardization and thus avoiding the disadvantages that the preparation of a standard solution may offer. One need only to seal a known amount of the uranium mineral within a tube, allow time for the radon to come in equilibrium with the radium, and the amount of radon that will be available after the proper time elapse will be,

if,  $U$  = amount of uranium in mineral

E.P. = emanating power expressed in %

then,  $\text{Curies} = U \times 3.4 \times 10^{-7} \times \text{E.P.}$

In general, however, it is rather inconvenient to wait thirty days so that by the employment of the following formula,

if,  $I_0$  = equilibrium ionization value of radon

$I_t$  = ionization value of radon at time  $t$

$t$  = time

$\lambda$  = disintegration constant for radon

then,  $I_t = I_0 (1 - e^{-\lambda t})$

and, the amount of radon available at time  $t$ , will be,

$\text{Curies} = U \times 3.4 \times 10^{-7} \times \text{E.P.} \times (1 - e^{-\lambda t})$

This, of course, assumes that the emanating power remains constant and is not function of time.

The emanating power of pitchblende amounts to only about 5-8 per cent, but in carnotite may be as high as 55 per cent. Undoubtedly the emanating power is a function of the surface exposed, but it is interesting to note that in rocks where the quantity of radium present is of the order of  $10^{-12}$  grams per gram of rock, grinding so that the rock will pass through 60 mesh does not perceptibly increase the emanating power.

J.H.L. Johnstone<sup>1</sup> has attempted to use carnotite as a radium emanation standard. He found that the temperature coefficient of the emanating power is considerably decreased by heating carnotite to 400°C. about 1/2 hour—that is for a range in temperature the emanating power for all practical purposes will remain constant. The emanating power of the mineral itself was decreased after such treatment.

In investigating the use of carnotite as an emanation standard, it was found that the emanating power was a function of temperature, pressure, and humidity. An increase in temperature increased the emanating power. Johnstone<sup>1</sup> has indicated that for normal carnotite between ranges of temperature of 20 - 130 degrees C., the emanating power is a linear function. A decrease in pressure decreased the emanating power. An increase in humidity increased the emanating power.

Fig. 2 represents diagrammatically the set up used. 2.5 grams of carnotite whose uranium content<sup>2</sup> determined as 1.59%,

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1

J.H.L. Johnstone: Royal Soc. of Canada, 15, 101-9, (1921)

2

Analysed by Ledoux & Co.

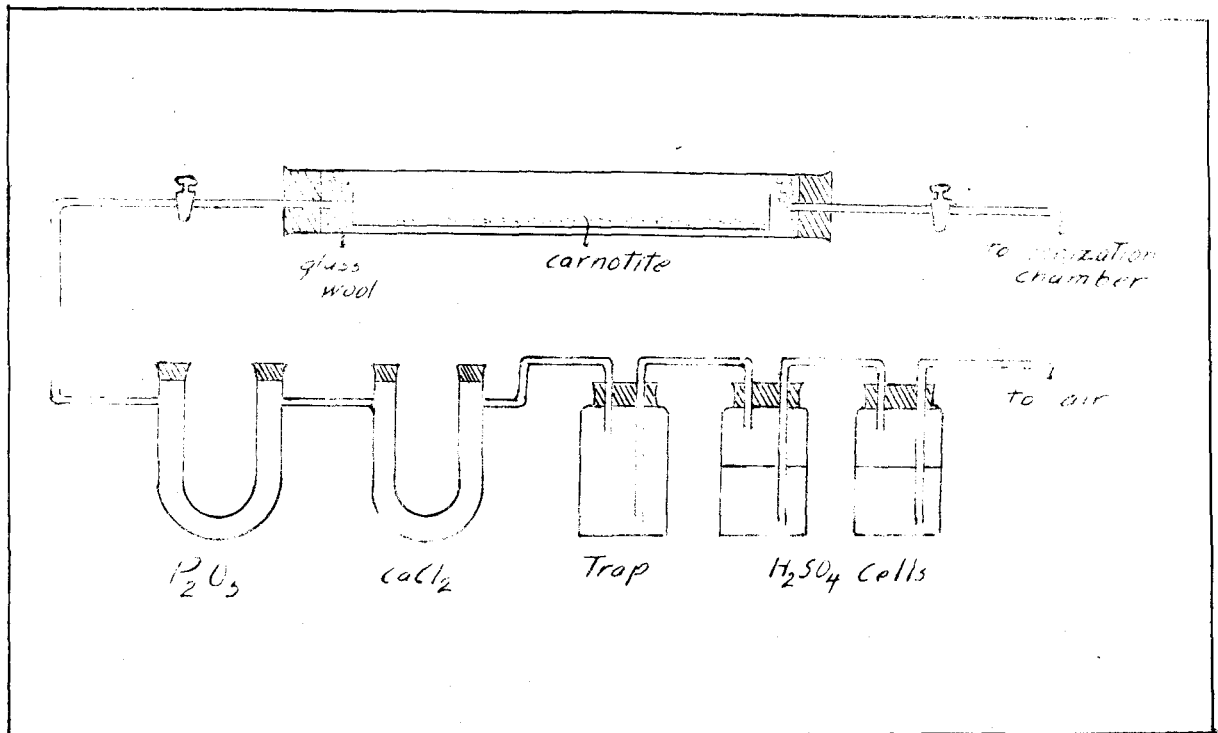


Fig. 2

Diagram of method for determining emanating power.

was spread evenly and in a thin layer in a long vessel and sealed in a glass tube. Dry air, obtained by passing through two sulfuric acid drying cells,  $\text{CaCl}_2$ , and  $\text{P}_2\text{O}_5$ , was used for sweeping the accumulated radon into the ionization chamber. This procedure was necessary since the effect of humidity as indicated is important.

Fig. 3 shows the variability of the emanating power of carnotite. It is interesting to note that these results do not confirm Johnstone's<sup>1</sup> conclusion that for preheated carnotite, the emanating power will remain constant through small changes in temperature. It is shown in Fig. 3 that the emanating power of carnotite is 1/10 of that of the unheated mineral. The heated carnotite exhibited a change in color from yellow to orange. This indicates a discontinuity in the emanating power-temperature relationship. A possible change in the physical chemical state of carnotite with a loss of the water of crystallization is the suggested explanation for this transformation. Rutherford has indicated a similar phenomenon for  $\text{RaCl}_2$ .  $\text{RaCl}_2$  with its water of crystallization loses a greater part of its emanation at a temperature considerably below that of fusion, but once the water of crystallization is removed, a much higher temperature at the second heating is required to drive off all of the emanation<sup>2</sup>.

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1

J.H.L. Johnstone: loc. cit.

2

Rutherford: Radioactive substances and their radiations. pp. 363

EMANATING POWER OF CARNOTITE						
Test	hours of accumulation	$D_0$ div/hr	$D$ div/hr	$D_{\text{equilibrium}}$ div/hr	emanating power in %	Remarks
1	47.3	1.1	235.0	783	52.9	normal carnotite used - radon accumulated in dry atmosphere
2	24.5	0.9	100.0	593	40.0	
3	24.0	0.9	113.6	686	46.4	
4	44.8	1.1	20.5	71.0	4.80	carnotite previously heated to 400°C for 1/2 hour used - radon accumulated in dry atmosphere
5	50.0	0.7	19.7	63.0	4.26	
6	46.0	0.9	16.7	57.0	3.85	
7	69.0	0.9	24.5	60.5	4.16	

Fig. 3

Indicating results obtained from set up in Fig. 2. It is interesting to note the variability of the emanating power.

### G. The Use of the Physical Constants of the Instruments

#### (1) Theoretical Ionization for one curie radon.

The value for the ionization in electrostatic units due to one curie of radon may be calculated from the number of alpha particles emitted, ion production, and the electronic charge.

Initial Ionization: One curie of radon emits  $3.7 \times 10^{10}$  alpha particles per second<sup>1</sup>. Each alpha particle from radon produces  $1.55 \times 10^5$  ion pairs<sup>1</sup>. Milliken's value for the electronic charge is  $4.77 \times 10^{-10}$  e.s.u. Then the ionization due to one curie radon is,

$$3.7 \times 10^{10} \times 1.55 \times 10^5 \times 4.77 \times 10^{-10} = 2.74 \times 10^6 \text{ e.s.u.}$$

This value may be compared with Mache and Meyer<sup>2</sup> who used slightly different constants.

$$3.4 \times 10^{10} \times 1.74 \times 10^5 \times 4.65 \times 10^{-10} = 2.75 \times 10^6 \text{ e.s.u.}$$

Maximum Ionization: In about three hours radon attains equilibrium with its decay products, which are RaA, RaB, RaC, RaC', RaC". Radium D may be considered for all practical purposes as the end product of this series, since its half-life value is 22 years. The alpha emitting substances of this series are only RaA, RaC, and RaC'. RaB and RaC emit beta and gamma radiations, and RaC" only beta radiation. The amount of ionization due to the alpha, beta, gamma rays is of the relative order of 10,000, 100, and 1<sup>3</sup>. Thus in an ionization chamber

<sup>1</sup> Rev. Mod. Ph. 3, 459, (1931)

<sup>2</sup> Mache and Meyer: Physik. Zeitschr., 13, 320, (1912)

<sup>3</sup> Rutherford: Radioactive Substances and their Radiations. pp. 117

where radon is in equilibrium with its decay products, the ionization, is almost totally caused by the alpha emitting substances, i.e., radon, RaA, RaC, RaC'. It can also be shown that the ionization caused by the alpha radiation from RaC is negligible, of a certain branching that occurs in the disintegration series at this point. See Fig. 4. Only 0.04 per cent of the radiation from RaC are alpha particles and consequently the ionization due to such may be neglected. RaA and RaC' produce  $1.70 \times 10^5$  and  $2.20 \times 10^5$  ion pairs respectively<sup>1</sup>.

Then the ionization due to one curie of radon in equilibrium with its decay products is,

$$3.7 \times 10^{10} \quad 1.55 \frac{1}{2} (1.70 \quad 2.20) \quad 10^5 \times 4.77 \times 10^{-10} = 6.19 \times 10^6 \text{ e.s.u.}$$

The factor of one half in the above expression is based on the fact that since a negatively charged electrode<sup>2</sup> will collect the radon decay products, one half of their energy must be imparted to the rod.

$6.19 \times 10^6$  may be compared with the value obtained by Mache and Meyer<sup>3</sup> who used slightly different constants.

$$3.4 \times 10^{10} \quad 1.74 \frac{1}{2} (1.87 \quad 2.37) \quad 10^5 \times 4.65 \times 10^{-10} = 4.10 \times 10^6 \text{ e.s.u.}$$

Schmidt and Nick<sup>4</sup>, Duane and Laborde<sup>5</sup>, Flamm and Mache<sup>6</sup>,

1

Rev. Mod. Phy. 3, 459, (1931)

2 Evans: Ph. D. thesis, C.I.T

Dedourian: A.J.S., 19, 16, (1905)

Kovarik and McKeehan: Bul. of Nat. Res. Council, 10, 140, (1925)

McGee: Phil. Mag., 13, 1, (1932)

3

Mache and Meyer: loc. cit.

4

Schmidt and Nick: Physik. Zeit., 13, 119, (1912)

5

Duane and Laborde: Comp. Rend., 1, 1421, (1910)

6

Flamm and Mache: Mitt. D. Inst. f. Radiumforschung XII, Weim. Ber. 121, (1912)

Physik. Zeit. 13, 321, (1912)

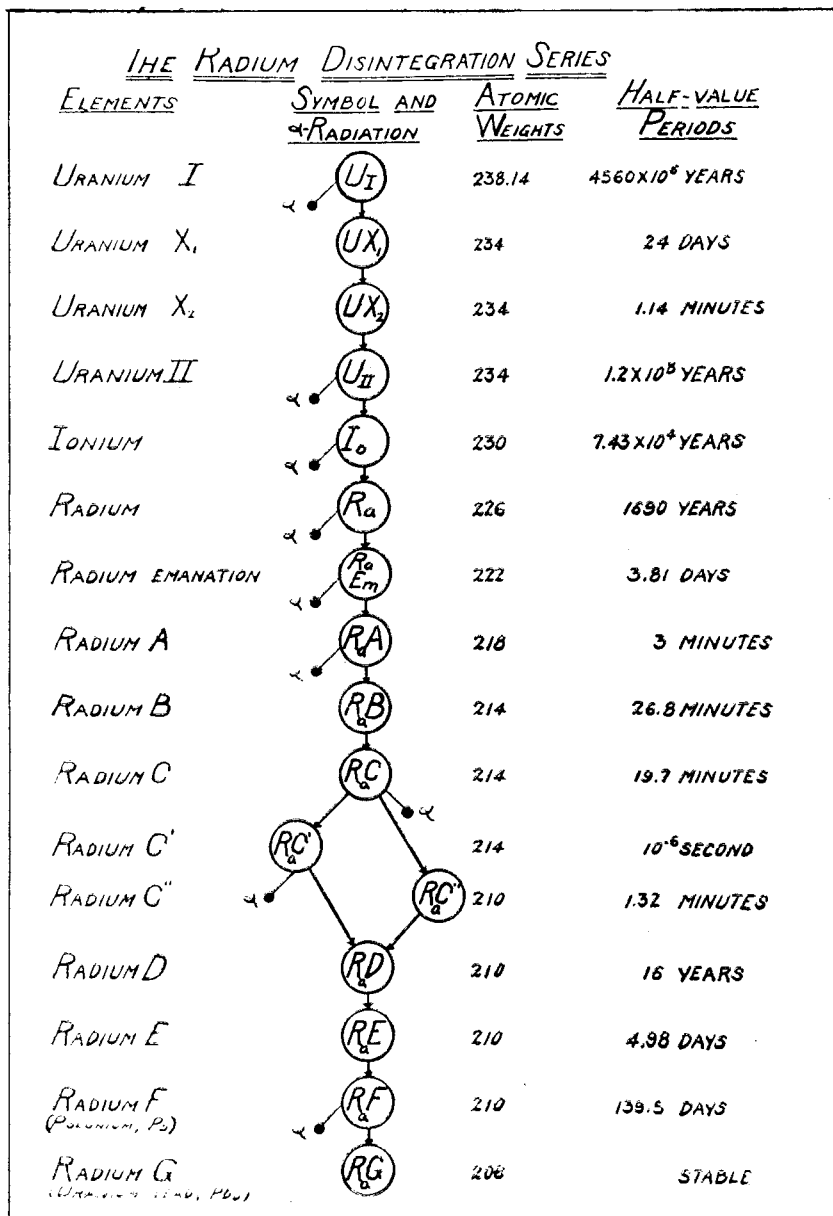


Fig. 4

Note branching of RaC. Only 0.04 per cent of the radiation from RaC are alpha particles and ionization from such may be neglected.



have published experimental values for the initial and maximum ionization due to one curie of radon. Some doubt, however, has been cast upon the standards used by Schmidt and Nick and their determinations are perhaps open to question. These values are summarized on Fig. 5.

(2) Ionization in a Gas.

If one curie of radon is introduced into an ionization chamber a certain theoretical ionization is produced in gas of the chamber, that may be calculated from the number of alpha particles that one curie of radon emits. This ionization is measured as a decrease in potential of a charged electrometer, and is related by the following formula,

if,  $I$  = ionization or current

$C$  = capacity of system (Cap. of ionization chamber plus capacity of electrometer)

$\frac{dy}{dt}$  = rate of decrease of potential

then,

$$I = C \frac{dy}{dt}$$

If it is assumed that all the alpha particles from the one curie of radon expend their energy in the production of ions and all of these ions are collected, the following may be written,

if,  $n$  = number of curies of radon

$C$  = total capacity in centimeters

$\frac{dy}{dt}$  = rate of decrease of potential e.s.u.

$I$  = theoretical ionization for one curie of radon in e.s.u.

then,

$$n = \frac{C \frac{dy}{dt}}{I}$$

## IONIZATION VALUES FOR ONE CURIE OF RADON

Values obtained by	Reference	$I_0 \cdot 10^6$ c.s.u.	$I_{max.} \cdot 10^6$ c.s.u.	Remarks
Experiment	Duane and Laborde	2.48	6.27	using various cylindrical chambers
	Flamm and Mache	2.67	6.02	using a plate condenser
	Schmidt and Nick	—	6.07	using a cylindrical chamber where $h = 25.2$ cm. $d = 7.1$ cm.
		—	6.36	using a cylindrical chamber where $h = 6.5$ cm. $d = 7.1$ cm
Theory	Mache and Meyer	2.75	6.10	based on Rutherford's constants
		2.74	6.19	based on constants in Rev. of Mod. Physics 2, 439, 1931

Fig. 5

This formula would immediately enable the standardization of an apparatus by merely determining the voltage calibration of the electrometer, and capacities of the electrometer and ionization chamber, but unfortunately the above two assumptions (wall effect and saturation effect) are not strictly valid and corresponding modifications must be made.

#### Capacity of Electrometer and Chambers.

The capacity of the electrometer was determined by the method of sharing of charges with a standard cylindrical condenser which is given by the expression,

if, C = Capacity

L = length of cylinder

b = radius of cylinder

a = radius of wire electrode

then,

$$C = \frac{L}{2 \log_{10} \frac{b}{a}}$$

The above expression for the capacity of a cylindrical condenser does not strictly hold because of the electrical field distortion at the ends, however, the formula was made to apply by making measurements on three cylinders of different lengths but the same diameters, and algebraically eliminating the end effects

The condensers were made of brass tubing and fitted with a collar having amber insulation for the center electrode.

(See description of ionization chamber by Evans<sup>1</sup>)

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<sup>1</sup>

Evans: Ph. D. thesis C.I.T.

The electrometer was charged to potential  $V_1$  and then connected with the standard cylindrical condenser by placing upon collar. The added capacity changed the initial potential to  $V_2$ .

Since a quantity of charge  $Q$ , equals the product of capacity  $C$  by voltage  $V_1$  or  $Q = CV_1$ , the following may be written,

if,  $C_e$  = capacity of electrometer

$C_s$  = capacity of cylindrical condenser

$V_1$  = initial voltage

$V_2$  = voltage after sharing charge with condenser

then,

$$C_e V_1 = (C_e + C_s) V_2$$

$$C_e = \frac{V_2}{V_1 - V_2} C_s$$

Then for a condenser where  $b = 2.435$  cm. and  $a = 0.04875$  cm.

$$\frac{C}{L} = \frac{1}{2 \log_e \frac{b}{a}}$$

$$\frac{C}{L} = 0.128 \text{ cm/cm.}$$

If  $l_1, l_2, l_3$  are the respective lengths of the shortest, intermediate, and longest condensers, the capacity of a condenser whose length is,

$$l_2 - l_1 = 0.128 \times 15.1 = 1.93 \text{ cm.} = C_x$$

$$l_3 - l_1 = 0.128 \times 30 = 3.84 \text{ cm.} = C_y$$

The following equations can then be obtained,

$$C_e = 1.145C_1$$

$$C_e = 0.703 (C_1 + C_x)$$

$$C_e = 0.507 (C_1 + C_y)$$

and solved for the electrometer capacity. See Fig. 6 for data.

It should be mentioned that the capacity of the electrometer is a function of the potential to which the fibers are charged. The above method for the determination of the capacity, essentially gives an averaged capacity for  $C_e$  between  $V_1$  and  $V_2$ .

The total capacity for the ionization chamber and the electrometer or the individual capacity of the chamber may be determined once the capacity of the electrometer is known. Merely charge the electrometer to potential  $V_1$ , and after sharing the charge with the chamber, the potential will be  $V_2$ . Then,

$$C_e V_1 = (C_e + C_{\text{chamber}}) V_2$$

Since we are only interested in the total capacity of the system,  $C_e + C_{\text{chamber}} = C_t$ ,

$$C_t = \frac{C_e V_1}{V_2}$$

A summary of this data is presented in Fig. 6 .

#### Formula of Duane and Laborde.

Consider an atom of radon in an ionization chamber; according to the laws that govern its probable life, an alpha particle eventually will be emitted. The range of an alpha particle emitted by radon in air is 4.122 cm. Thus the alpha

Test Cylinder	length cm.	dia. of cy. (inside) cm.	dia. of wire cm.	$V_1$ volts	$V_2$ volts	$\sqrt{\frac{V_2}{V_1 - V_2}}$	$C_E$	$C_E$
$C_1$	20.16	4.87	.0975	253.5	135.3	1.145	1.145 $C_1$	3.52
$C_2$	35.26	4.87	.0975	253.5	104.7	0.703	0.703( $C_1 + C_2$ )	3.495
$C_3$	50.16	4.87	.0975	253.5	85.3	0.507	0.507( $C_1 + C_2$ )	3.47

av. 3.50  
max. diff. from av. 0.85%

Fig. 6

Capacity determination data.

particle will travel this distance and will produce a certain number of ion pairs.

If, however,  $x$  curies of radon are contained in the chamber, the radon, being a gas, will be evenly distributed and consequently a certain fraction of the alpha particles will strike the walls of the chamber before they will have traveled their entire range. Thus upon striking the walls part of their energy is entirely absorbed, although secondary ions may be partially produced.

A modification of this formula,  $n = \frac{C \frac{dv}{dt}}{I}$ , presented on page 18 must be made as follows:

For a system of capacity,  $C$ , where the rate of discharge is  $\frac{dv}{dt}$ ,  $I$  equals the theoretical ionization of one curie,  $n$ , the number of curies will be,

$$n = \frac{C \frac{dv}{dt}}{I - \text{wall effect}}$$

Duane<sup>1</sup> in 1905 presented a formula for this effect in the following form:

if,  $I$  = theoretical ionization for one curie (as in free space)

$K$  = empirical constant

$S$  = surface of chamber

$V$  = volume of chamber

then,

$$I_{\text{obs.}} = I - K \frac{S}{V}$$

---

<sup>1</sup>

Duane: Jour. de Phy., 4, 605, (1905)  
Comp. Rend., 1, 581, (1905)

Although numerical values were presented for K and I in the early paper, these constants were not accurately determined until 1910 when Duane and Laborde<sup>1</sup> offered the following formulas:

if,  $I_0$  = initial ionization due to radon produced by 1 gm. radium in one second

$I_{\text{max.}}$  = ionization after three hours when equilibrium is established between Rn, RaA, RaC.

then,

$$I_0 = 5.19 \left(1 - 0.517 \frac{S}{V}\right)$$

$$I_{\text{max.}} = 13.15 \left(1 - 0.572 \frac{S}{V}\right).$$

#### Theoretical Consideration of Duane's Wall Effect.

The Duane and Laborde formula merely states that the wall effect or the energy lost by an alpha particle upon striking the wall, is roughly proportional to  $S/V$ . The formula, however, is an empirical one and can be shown to be invalid for small ionization chambers.

For if,

$$I_{\text{obs.}} = I \left(1 - K \frac{S}{V}\right)$$

$$\frac{S}{V} = \frac{2\pi rh + 2\pi r^2}{r^2 h}$$

$$= \frac{2(r+h)}{rh}$$

Thus for small chambers the  $K \frac{S}{V}$  factor becomes greater than one and the formula will give negative values for I.

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<sup>1</sup>

Duane and Laborde: *Comp. Ru.*, 1, 1421, (1910)



However, it is to be noted that the Duane and Laborde formula was applied to chamber whose heights are approximately twice their diameters, and also in the case where the height is only 6/10 the diameter. The dimensions<sup>1</sup>:

Chamber	Diameter	Height	$\frac{\text{Height}}{\text{Diameter}}$
1	18.5 cm.	37.5 cm.	2.02
2	11.92	26.75	2.24
3	6.70	12.5	1.87
4	18.0	11.0	0.61

The values 5.19 and 13.15 presented in the formulas on page 25 are for gm.-sec. units, that is, for the amount of radon that one gram of radium will produce in one second. These figures are to be multiplied by  $1/4 = 4.768 \times 10^5$  for radon in order to convert the values into the curie unit. The formulas then become,

$$I_o = 2.48 \left(1 - 0.517 \frac{S}{V}\right) 10^6 \text{ e.s.u.}$$

$$I_m = 6.27 \left(1 - 0.572 \frac{S}{V}\right) 10^6 \text{ e.s.u.}$$

The calibration formula based on Duane and Laborde work will then become,

If,  $n$  = number of curies of radon

$C$  = total capacity of system

$\frac{dv}{dt}$  = potential change three hours after introduction of radon in ionization chamber or when radon is in equilibrium with its decay products in e.s.u. per second.

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<sup>1</sup>

Duane and Laborde: Comp. Ru., 1, 1421, (1910)

then,

$$n = \frac{C \frac{dy}{dt}}{6.27 \left(1 - 0.572 \frac{S}{V}\right) 10^6}$$

#### Applications of Duane and Laborde Formula

The convenience and advantages offered by the use of a calibration formula based on physical characteristics of one's apparatus can readily be realized. The Duane and Laborde formula has been used in a various manner.

Hootman<sup>1</sup>, Hootman and Helms<sup>2</sup> have applied the formula in their work. In the latter paper the following statement was made, "The electroscopes were calibrated by means of the Duane and Laborde empirical formula, which was shown by Ramsay to give results of the same accuracy as those given by emanation standard E54." Geckel<sup>3</sup> has used the formula but has employed the constant  $6.02 \times 10^6$  e.s.u. for the initial term in the Duane and Laborde formula and has offered a temperature and pressure modification for the formula. Ramsay<sup>4</sup> compared the accuracy of the Duane and Laborde formula with emanation standard E54 which was certified by the Bureau of Standards to be accurate to within 3 per cent. He found at room temperature  $20^{\circ}\text{C}$ ., the

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1

Hootman: A.J.S., 22, 453, (1931)

2

Hootman and Helms: A.J.S., 21, 37, (1931)

3

Geckel: Die Radioaktivitat von Boden und Quellen; Sammlung Vieweg, pp. 102, (1914)

4

Ramsay: A.J.S., 40, 309, (1915)

formula to agree within 9 per cent. However, by using the value  $6.02 \times 10^6$  for the initial constant in the formula as is done by Geckel, he found the agreement to be within 4 per cent. Using the corrections for temperature and pressure as given by the same author, Ramsay found the accuracy of the Duane and Laborde formula to coincide with that of standard E54.

Geckel's temperature and pressure corrections are as follows:

if,  $I_c$  = corrected value for current  
 $I_{d\&l}$  = the Duane and Laborde value for current  
 $p$  = pressure in mm.  
 $t$  = temperature in C.

then,

$$I_c = I_{d\&l} \left[ 1 + 0.0007 (760 - p) + 0.002 (t - 15^\circ) \right]$$

Since Duane and Laborde only claimed applicability of their formula to chambers whose heights were approximately twice their diameters and in a single case where the height was only 6/10 of the diameter (See pp. 26), it was deemed advisable to investigate the pertinence of the Duane and Laborde formula to various sizes of ionization chambers.

#### Size of Ionization Chamber and Duane and Laborde Formula.

Four ionization chambers were constructed of brass. Since the center electrode, if charged negatively, will collect

the solid decay products from radon, the minimum radius dimension of these chambers was limited by the maximum range of subsequent alpha emitting solid on the center electrode.  $\text{RaC}^1$  has the longest range being 6.971 cm. in air. Two other brass chambers were available, but of the same dimensions, and had been constructed formerly, by the Geology Department especially for radium determination work. Thus a comparison of the ionization effects would be made with five chambers of different dimensions. See Fig. 7.

For comparison purposes, a known amount of radon must be inserted into each ionization chamber. An attempt to use carnotite as a source material for radon was made. Because of the variability of the emanating power of carnotite as demonstrated in Fig. 3., it was not to be expected that a procedure of merely sealing carnotite in a tube, allowing radon to accumulate for several days, and drawing the radon into an ionization chamber, could obtain duplicate readings even though all operations were exactly repeated.

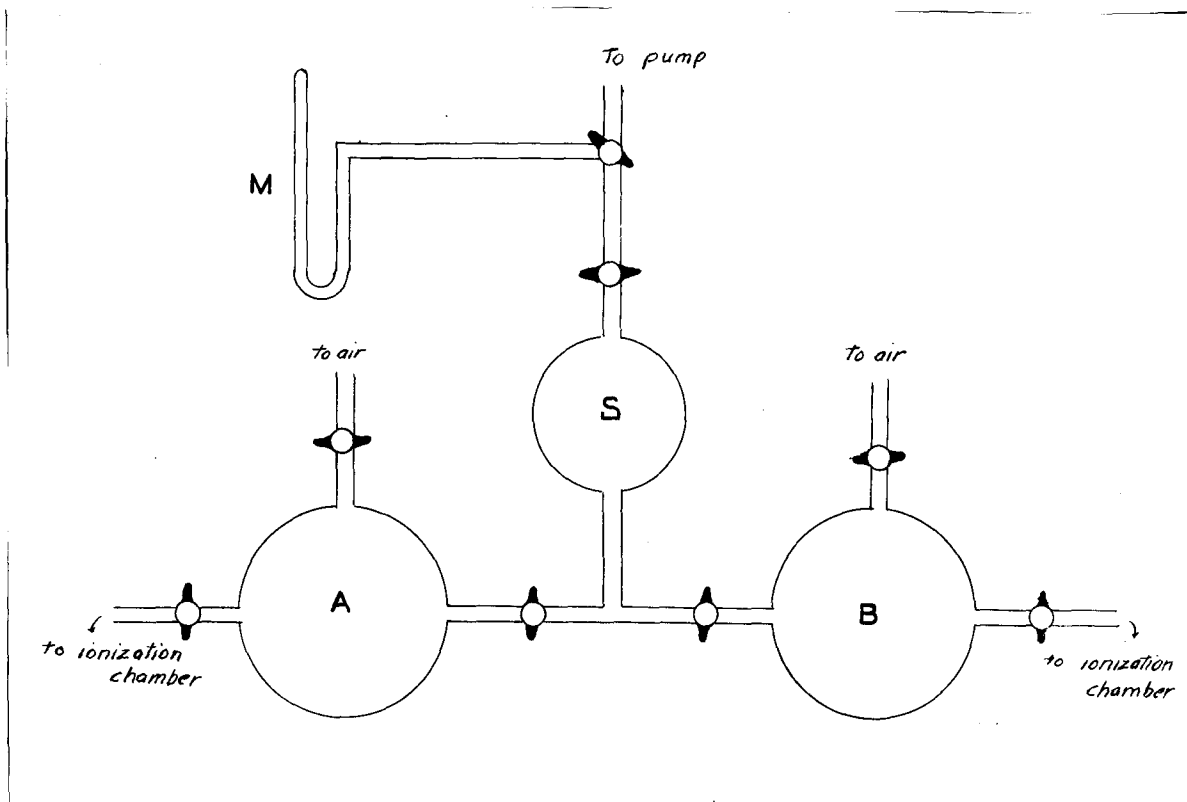
#### Radon Collecting Equalizer

A radon collecting equalizer was constructed of glass and is shown diagrammatically in Fig. 8. The apparatus was designed to divide the radon emitted from carnotite in source chamber S equally in bulbs A and B. Thus having two equal quantities of radon, comparison of the ionization could be made between

<i>Ionization Chamber</i>	<i>IC<sub>5</sub></i>	<i>IC<sub>6x6</sub></i>	<i>IC<sub>10x6</sub></i>	<i>IC<sub>6x12</sub></i>	<i>IC<sub>10x12</sub></i>
<i>height cm.</i>	19.05	15.24	15.24	30.45	30.45
<i>radius cm.</i>	7.3	7.62	12.70	7.62	12.70
<i>Total capacity (including electrometer)</i>	6.88	6.02	5.97	7.48	7.38
<i>Surface Volume</i>	0.378	0.393	0.288	0.328	0.223

Fig. 7

**Ionization chamber constants.**



**Fig. 8**

Diagram of radon collecting equalizer. M = manometer,  
 S = Source chamber containing carnotite, A-B = diffusion  
 bulbs.

two chambers of different sizes. The instrument was equipped with a manometer so that radon could accumulate under various conditions.

It was here again observed that an increase in the humidity at atmospheric pressures tended to increase the emanating power of carnotite. The emanating power was decreased considerably when accumulation of radon took place under reduced pressures such as three to six millimeters of mercury. However, the emanating power was increased somewhat at the latter pressures by insertion of a small crucible of water in the source chamber S.

Because of slight cross-sectional differences in the glass stopcocks the diffusion rates to the two bulbs were not the same, and unequal quantities of radon were collected in bulbs A and B. Since the diffusion of a gas is a function of time and concentration, this discrepancy could have been eliminated by performing the various experiments under equal times of diffusions. Under such conditions the ratio of concentrations of radon in bulbs A and B should be constant. However, after performing experiments under such conditions, it was found that this ratio did not remain constant, thus again indicating the variability of the emanating power of carnotite.. See Fig. 9. See previous discussion pp. 9.

#### Tap Water as a Source of Radon

A number of determinations of the radon content of ordinary tap water made and the results which show considerable

Test	Ionization Chamber IC dia x height	Background Leak Db in d/hrs	$D_0 - D_b = D$	# of hours radon accumulated	Ratio $\frac{D_1 \text{ d.b.}}{D_2 \text{ d.}}$
1	12x6	1.6	42.1	23	1.10
	6x6	1.5	37.9		
2	12x6	1.5	37.1	23.12	1.1
	6x6	1.8	50.2		
3	12x6	3.9	208.1	131	1.1
	6x6	3.0	188.5		
4	12x12	1.8	62.4	39.5	1.10
	5	0.5	60.7		
5	12x12	1.6	107.3	68.6	1.10
	5	0.5	99.1		
6	12x12	2.1	179.1	147.5	1.1
	5	2.7	162.3		
7	12x6	2.2	110.6	212.5	1.1
	5	0.7	86.8		
8	12x6	0.9	98.8	45.34	1.1
	5	2.2	82.1		
9	12x6	5.9	173.6	118.0	1.1
	5	2.5	139.8		
10	12x6	1.9	192.6	140	1.1
	5	0.6	159.4		
11	12x6	2.1	193.4	144	1.1
	5	0.7	160.9		
12	12x6	1.8	54.1	35.7	1.1
	5	1.0	20.5		
13	12x6	2.5	164.3	143.4	1.1
	5	1.0	140.8		
14	12x6	2.5	170.5	44	1.1
	5	0.7	146.3		
15	12x6	1.8	173.2	1.1	1.1
	5	0.9	134.1		
16	12x6	2.2	41.5	22.3	1.1
	5	1.1	31.5		

Fig. 9

Radon equalizer data.



variation from day to day are indicated in Fig. 10. The procedure for these determinations is described elsewhere.

See page 69.

The quantity of radon liberated from tap water produces a convenient rate of discharge of the electrometer and thus offered itself for use as a radon source. Two 2 liter flasks sampled directly from the tap within about five minutes were found to contain equal concentrations of radon. See Fig. 11.

The time required for the liberation of radon from a 2 liter flask is about one half hour, so both samples were run within one half hour of each other. The correction for the decay of radon in one half hour is negligible. Using this procedure excellent check values were obtained. Fig. 12 presents data of these experiments and percentage of differences for duplicate tests.

Fig 13 shows a comparison between the actually observed ionization effects for various sizes of chambers and that predicted by the Duane and Laborde formula for the maximum ionization for radon, i.e.,  $I = 6.27 (1 - 0.572 \frac{S}{V}) 10^6$  e.s.u. The chamber whose dimensions, height = 19.05 cm., radius = 7.3 cm., was designated as the standard chamber and all values are referred to this chamber for comparison. The values for the ionization effects are presented in ratios, i.e., ratio of chamber x to the standard chamber, since this form of presentation eliminates the error in absolute calibration of the standard chamber in terms of a radium standard and also individual

VARIATION OF RADON CONTENT OF TAP WATER				
Date	Observed radon concn. d.p.m./ltr	Background radon concn. d.p.m./ltr	$R_0 - R_b$ d.p.m./ltr	Calc. R.P. d.p.m./ltr
11-16-32	38.6	0.9	37.7	1.38
-17-	36.2	0.9	35.3	1.29
-18-	41.0	1.0	40.0	1.29
-19-	43.5	1.0	42.5	1.55
-20-	46.9	0.9	46.0	1.71
-21-	41.0	0.9	40.1	1.46
-22-	46.0	0.9	45.1	1.64
-24-	49.2	0.7	48.5	1.72
-25-	35.3	0.7	34.6	1.26
-27-	54.1	0.7	53.4	1.95
-28-	55.0	0.9	54.1	1.97
-30-	56.3	1.0	55.3	2.02
12-1-32	67.1	0.7	66.4	2.42
-2-	52.5	0.6	51.9	1.90
-3-	48.5	0.8	47.7	1.74
-4-	50.5	0.7	49.8	1.82
-5-	52.1	0.6	51.5	1.88
-25-	38.2	0.5	37.7	1.38
-26-	41.6	0.5	41.1	1.50
-27-	55.5	0.5	55.0	2.01
-30-	53.0	0.7	52.3	1.91
1-7-33	32.8	0.7	32.1	1.17

Fig. 10

Test	Ionization Chamber	Background Leak $D_b$	Observed Leak $D_o$	$D_o - D_b = D$	Curies per liter	Deviation in percent
A	$S_1$	0.7	47.2	46.5	$1.67 \times 10^{-10}$	0.54%
	$S_2$	0.9	46.9	46.0	$1.71 \times 10^{-10}$	
B	$S_1$	0.9	46.0	45.1	$1.64 \times 10^{-10}$	0.31%
	$S_2$	1.0	45.8	44.8	$1.53 \times 10^{-10}$	
C	$S_1$	0.9	40.7	39.8	$1.45 \times 10^{-10}$	0.34%
	$S_2$	1.0	41.0	40.0	$1.46 \times 10^{-10}$	

Fig. 11

Indicating the quantity of radon contained in tap water. Two samples taken within five minutes of each other contained virtually the same quantity of radon. Thus tap water offers a source of radon that may be used for comparing ionization effects.

Test	Ionization Chamber IC dia. (height width)	Background Leak $P_0$ in d/s/hr	Observed Leak $D_0$ in d/s/hr	$P_0 - P_{0.5}$ D	$\frac{L_{excess}}{D_{corrected}}$	Maximum Leakage
1	10x6	2.2	55.3	53.1	1.09	0.9%
	5	0.7	49.2	48.5		
2	10x6	2.2	61.7	59.5	1.1	
	5	0.9	55.0	54.1		
3	10x6	2.2	62.9	60.7	1.1	
	5	0.95	56.3	55.35		
4	6x6	1.33	58.3	57.0	1.1	
	5	0.6	52.5	51.9		
5	6x6	1.0	53.5	52.5	1.102	
	5	0.75	48.45	47.6		
6	6x6	1.1	55.4	54.3	1.09	
	5	0.7	50.5	49.8		
7	10x12	2.3	39.7	37.4	0.993	
	5	0.5	38.2	37.7		
8	10x12	2.3	43.2	40.9	0.995	
	5	0.5	41.6	41.1		
9	10x12	2.3	56.7	54.4	0.989	
	5	0.5	55.5	55.0		
10	10x12	2.5	36.6	34.1	0.986	
	5	0.7	35.3	34.6		
11	6x12	0.7	48.0	47.3	0.904	
	5	0.6	52.8	52.3		
12	6x12	0.7	29.1	28.4	0.907	
	5	0.5	32.6	32.1		

Fig. 12.

Indicating the ionisation differences due to various sizes of ionization chambers. Two 2 liter flasks of tap water sampled within five minutes of each other were used as sources of radon. Fig. 10 shows that equal quantities of radon are contained in the flask.

Ratio of Ionization Chambers	$\frac{6 \times 6}{5}$	$\frac{6 \times 12}{5}$	$\frac{10 \times 6}{5}$	$\frac{10 \times 12}{5}$
Ratio of Capacities $\frac{C_{dsh}}{C_s}$	0.875	1.088	0.869	1.07
Ratio of observed rates $\frac{D_{dsh}}{D_s}$	1.10	0.905	1.10	0.990
Ratio of observed Ionization $\frac{[D \times C]_{dsh}}{D \times C} = \frac{I_{dsh}}{I_s}$	0.962	0.985	0.956	1.06
Ratio of Duane and Laborde Ionization $\frac{I_{dsh}}{I_s}$	0.985	1.035	1.06	1.11
Percentage deviation from observed I.	2.6%	5.1%	12.1%	4.7%

Fig. 15

Indicating a comparison of observed ionization of various chambers with that given by the Duane and Laborde formula.

In Fig. 25 the ratios are corrected for saturation.

errors in the determination of the total capacity of each system.

Thus the observed ionization may be compared with that obtained by the use of the Duane and Laborde formula and it is apparent that the agreement is not satisfactory. In all cases the ratios obtained by the formula are high, and in a single case, see Fig. 13, as much as 12.1 per cent greater than the observed value.

It would seem from these results that the Duane and Laborde formula for the wall effect is unreliable but it will be shown that the data in Fig. 13, have to be modified because of "saturation", an effect which is fully as important as the wall effect and adds another element of complexity to the Duane and Laborde formula.

#### The Effect of Saturation

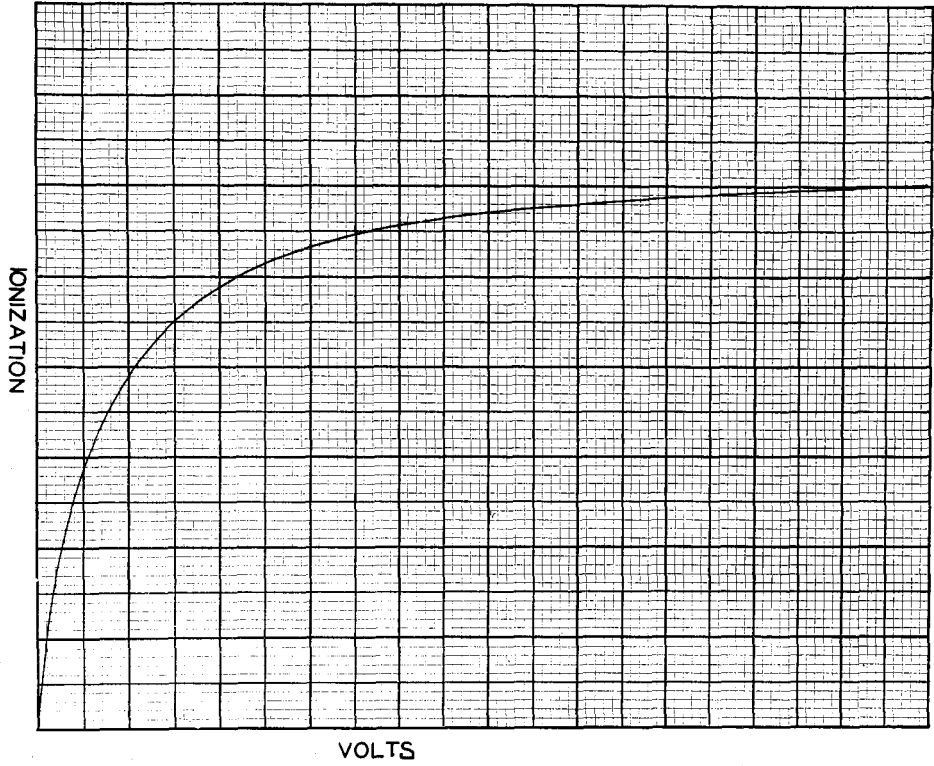
When an alpha particle expends its energy in a gas, a large number of ion pairs are produced. Were it not for an electrical field, these ions would merely recombine. However in spite of an electrical field a large number of these ions do recombine, and it is known that only at extremely high voltages can "saturation" or a full collection of the ions be expected.

A typical form of the saturation curve is shown in Fig. 14<sup>1</sup>.

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<sup>1</sup>

Rutherford: Radioactive Substances and their Radiations pp. 29



**Fig. 14**

**Saturation curve.**

A convenient form of plotting a saturation curve is to use current/voltage as the ordinate and the current as the abscissa. This has the advantage of keeping infinity on the paper. A typical curve of this type is depicted in Fig. 15<sup>1</sup>.

If the potential applied to the center electrode is the same in all cases, it is obvious that in comparing the ionization effects of various sizes of cylindrical ionization chambers, those chambers having the largest diameters would have a lesser field gradient and consequently a different number of ions will be collected, or in other words, a different percentage of saturation.

A number of experiments were performed applying various collecting potentials to the center electrode. A considerable quantity of radon was drawn into the chambers and readings were observed with the various voltages applied to the center electrode during the period when the maximum ionization for radon is attained (3-5 hours after chambers are filled.)

It was found that collecting potentials of 136 volts and 243 volts resulted in a difference of ionization of about 10 per cent for the standard chamber and was of a similar magnitude for the other chambers. The data obtained from these experiments are graphically represented by both suggested methods of plotting, and the percentage of saturation expected at 243 volts indicated. See Figs. 16, 17, 18 and 19.

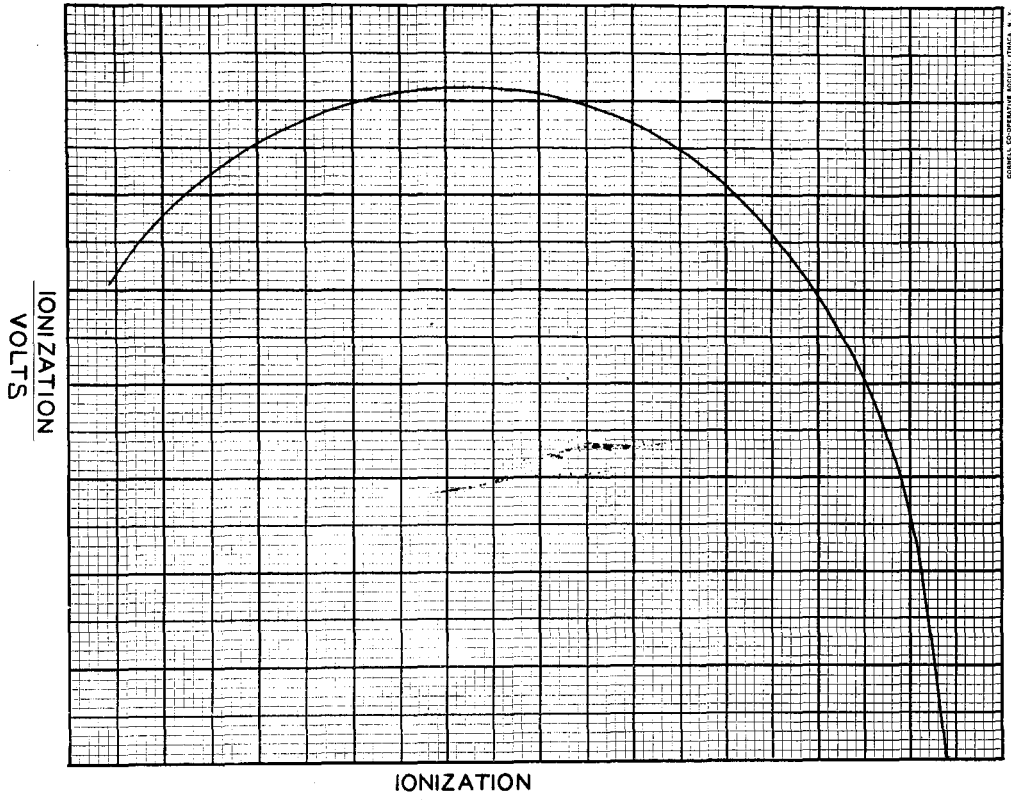
This method for determining the percentage of saturation

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<sup>1</sup>

Meyer and Schweidler: Radioaktivitat pp. 280.





**Fig. 15**  
**Saturation curve.**

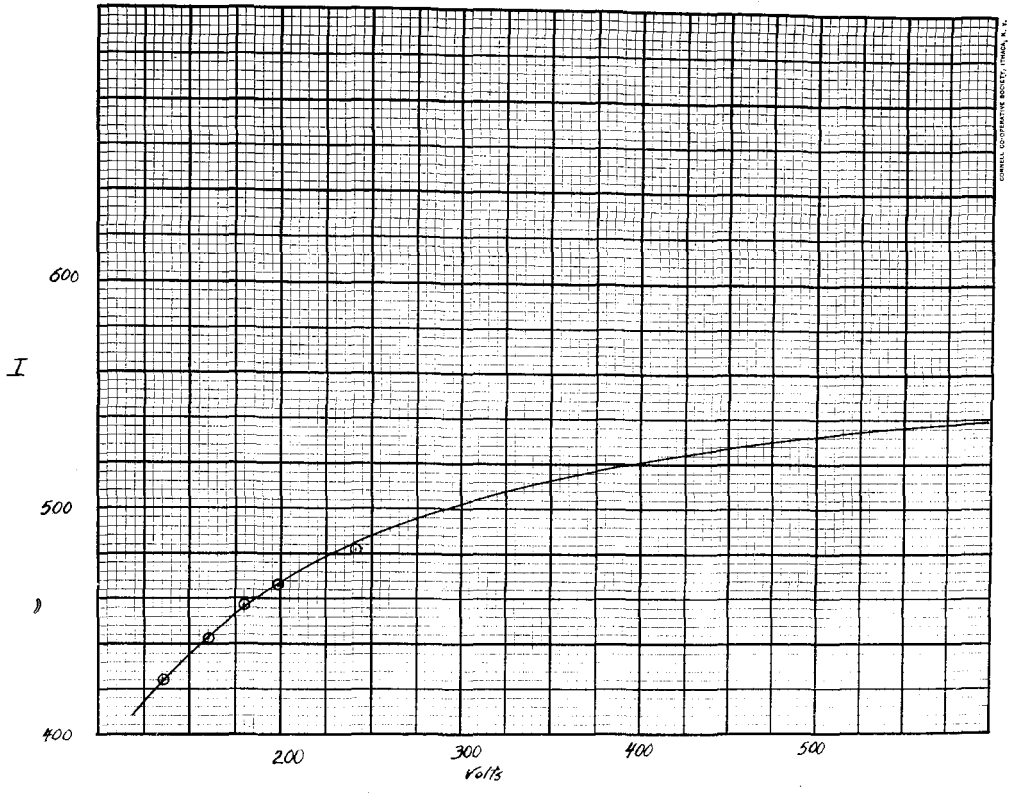


Fig. 16

% saturation at 243 volts for IC<sub>2</sub>

$$\frac{476}{540} \times 100 = 88.2\%$$

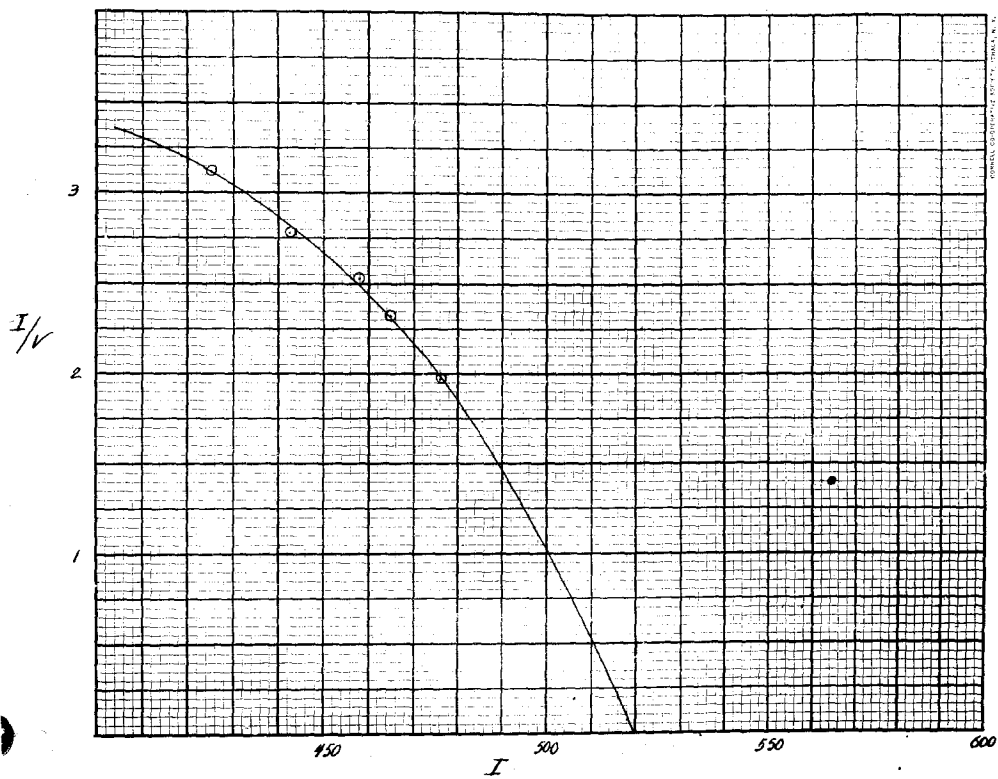


Fig. 17

\* saturation of 243 volts for  $IC_2$

$$\frac{476}{520} \times 100 = 91.5\%$$

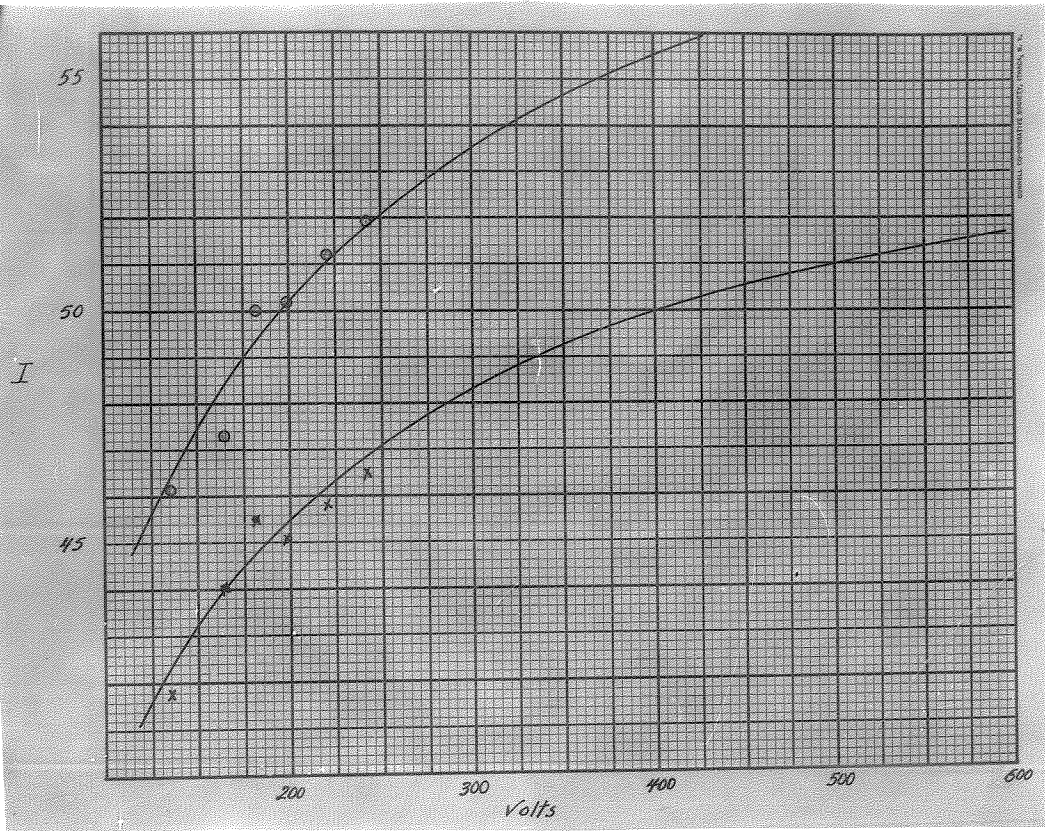


Fig. 18

Saturation curves

o = IC<sub>10x12</sub>x = IC<sub>6x12</sub>

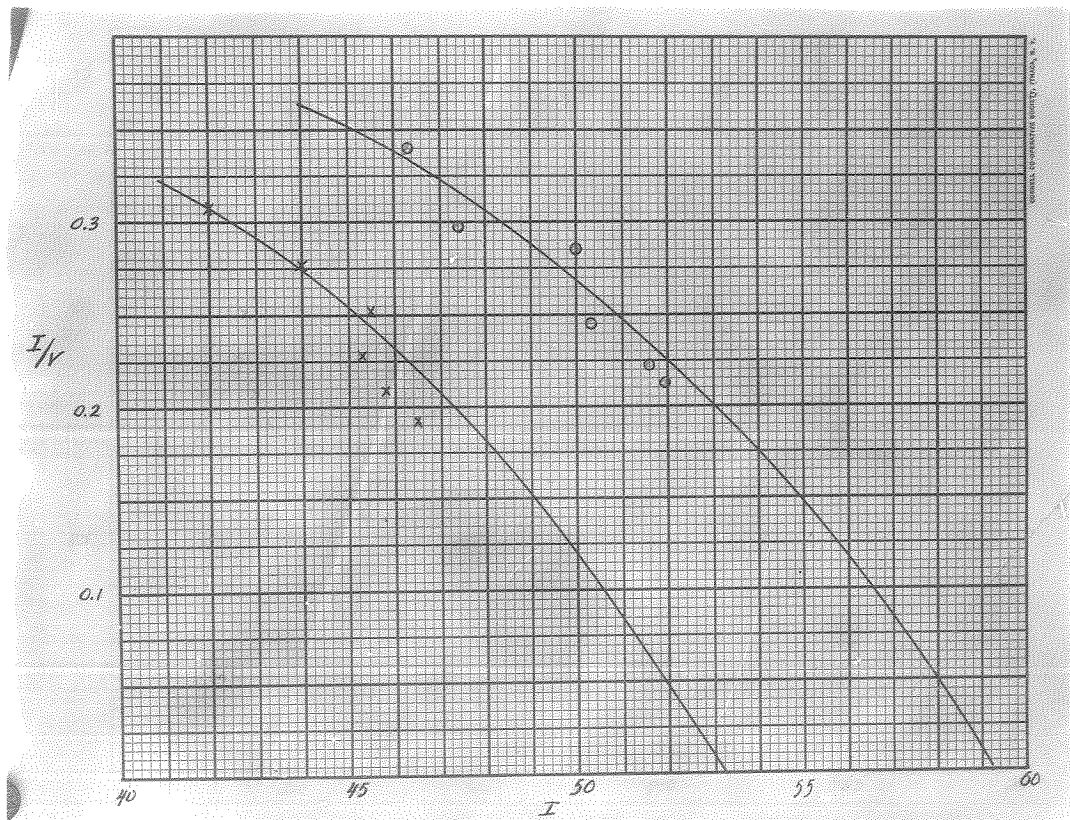


Fig. 19

## Saturation curves

$$\circ = IC_{10 \times 12} = \frac{51.9}{59.3} \times 100 = 87.5\%$$

$$\times = IC_{6 \times 12} = \frac{46.5}{53.3} \times 100 = 87.4\%$$

involves considerable inaccuracy because of the extra polation over a large distance, since the number of points obtainable were limited by the full scale potential of the electrometer which amounts to only approximately 300 volts. See Figs. 19

#### Calculation of Percentage of Saturation.

An approximate value for the percentage of saturation may be obtained in the following manner,

Assume that the electrical field of a cylinder is the same as if the cylinder were infinitely long.

if,  $a$  = radius of center electrode

$b$  = radius of cylinder

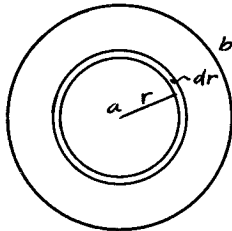
$V$  = voltage of center electrode

$X$  = field intensity (potential gradient) at distance  $r$   
from center electrode

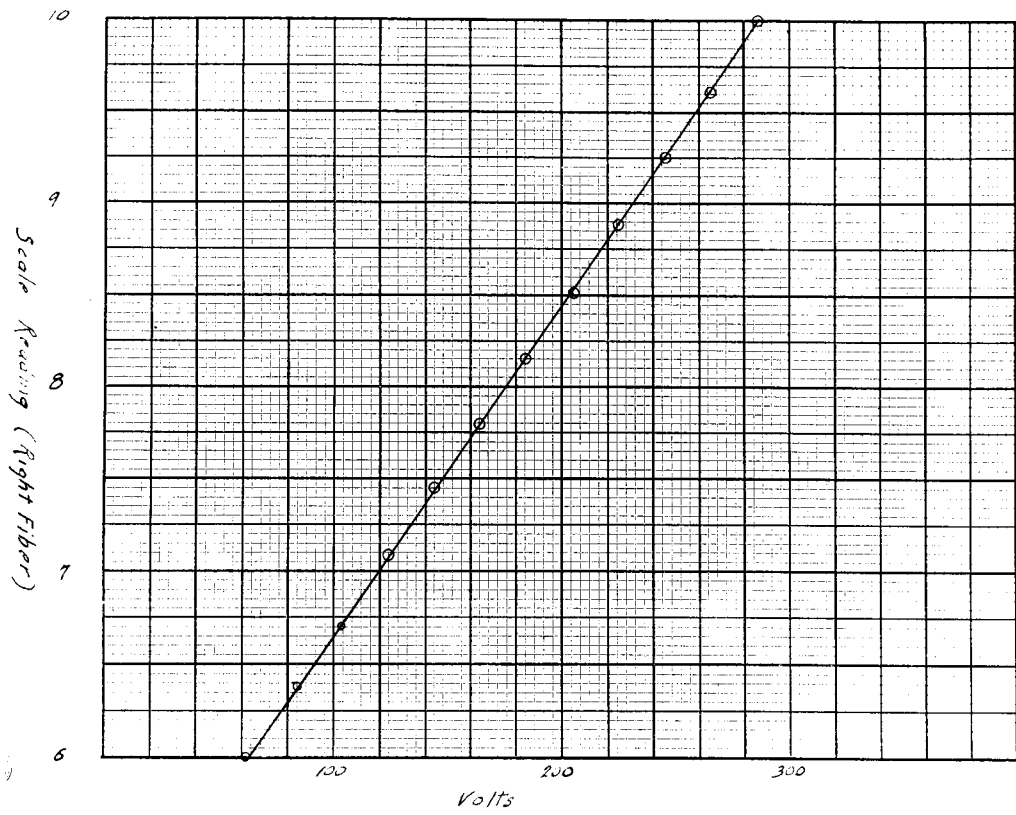
then,

$$X = \frac{V}{r \log_e \frac{b}{a}} \quad (1)$$

A summation of the total field intensity may be obtained by multiplying equation (1) by  $2\pi r dr$  and integrating from  $a$  to  $b$ .



$$\begin{aligned} \text{Total field intensity} &= \int_a^b \frac{V}{r \log_e \frac{b}{a}} \cdot 2\pi r dr \\ & \quad (\text{pot. grad.}) \\ &= \frac{2V}{\log_e \frac{b}{a}} (b - a) \quad (2) \end{aligned}$$

**Fig. 20****Voltage calibration of electrometer.**

By dividing (2) by the cross sectional area of the cylinder, an average field intensity is obtained.

$$\begin{aligned} \text{av. pot. grad.} &= \frac{2V(b-a)}{\log_e \frac{b}{a}} \cdot \frac{1}{(b^2 - a^2)} \\ &= \frac{2V}{\log_e \frac{b}{a} (b+a)} \end{aligned}$$

Then in the standard chamber where,

$$b = 7.3 \text{ cm.}, \quad a = .05 \text{ cm.}$$

$$\text{av. pot. grad.} = 13.3 \text{ volts per cm.}$$

This value of 13.3 volts per cm. corresponds to 80 per cent saturation. (Obtained by interpolating values given in Fig. 21. )

Then in chamber where,

$$b = 7.62, \quad a = .05$$

$$\text{av. pot. grad.} = 12.6 \text{ volts per cm.}$$

This value of 12.6 volts per cm. corresponds to 79.3 per cent saturation.

Then in chamber where,

$$b = 12.7, \quad a = .05$$

$$\text{av. pot. grad.} = 6.9 \text{ volts per cm.}$$

This value of 6.9 volts per cm. corresponds to 72.2 per cent saturation.

Since the formulas,

$$\begin{aligned} I_o &= 2.46 \left( 1 - 0.517 \frac{S}{V} \right) 10^6 \text{ e.s.u.} \\ I_m &= 6.27 \left( 1 - 0.572 \frac{S}{V} \right) 10^6 \text{ e.s.u.} \end{aligned}$$

are linear functions their curves are expressed as straight



Da die Korrektur für die mangelnde Sättigung infolge der Kolumnenionisation der  $\alpha$ -Bahnen in erster Annäherung unabhängig ist von der Stromstärke, kann hierfür die nachstehende Tabelle herangezogen werden, die aus den Angaben

Volt cm	$i/i_{\infty}$		Volt cm	$i/i_{\infty}$	
	dem Feld	$\perp$ dem Feld		dem Feld	$\perp$ dem Feld
5	0,66 <sub>5</sub>	0,72 <sub>5</sub>	200	0,93	0,99 <sub>5</sub>
10	0,72 <sub>5</sub>	0,83	300	0,95	0,99 <sub>7</sub>
15	0,75 <sub>5</sub>	0,87 <sub>5</sub>	400	0,96 <sub>5</sub>	0,99 <sub>8</sub>
20	0,77 <sub>5</sub>	0,91	500	0,97	0,99 <sub>9</sub>
30	0,80 <sub>5</sub>	0,94	750	0,98	1,00
40	0,82 <sub>5</sub>	0,95 <sub>5</sub>	1000	0,98 <sub>5</sub>	
50	0,84	0,96	1500	0,99	
75	0,86	0,97	2000	0,99 <sub>5</sub>	
100	0,88	0,98	30. 0	1,00	
150	0,91	0,99			

M. Moulins gewonnen wurde. In der ersten Kolonne steht das elektrische Gefälle in Volt pro cm; die zwei folgenden kennzeichnen den Grad der Sättigung für die Fälle, daß die  $\alpha$ -Bahnen parallel oder senkrecht zum elektrischen Feld liegen.

(Gute Werte bei parallelem Verlauf von Feld und Strahlenrichtung liefern Formeln von G. Jaffé<sup>3</sup>).

**Fig. 21**

**Percentage of saturation for given potential gradient**

**See Meyer and Schweidler pp. 281.**

lines shown in Fig. 22. It is to be noticed that when  $S/V$  becomes zero, the values for  $I_0$  and  $I_m$  should be equal to the theoretical values for the initial and final ionization for one curie of radon. These values are  $I_0 = 2.74 \times 10^6$ ,  $I_m = 6.19 \times 10^6$ . See Fig. 5. The Duane and Laborde formula gives the values  $I_0 = 2.48 \times 10^6$ ,  $I_m = 6.27 \times 10^6$ , when  $\frac{S}{V}$  equals zero.

The difference between  $I_m$  and  $I_{m \text{ D\&L}}$  is small whereas that between  $I_0$  and  $I_{0 \text{ D\&L}}$  is considerably larger. This discrepancy may be due to a saturation effect. On the basis of the theoretical ionization for one curie of radon, the Duane and Laborde formulas represent saturation values of  $\frac{6.27}{6.19} \times 100 = 101.5$  per cent and  $\frac{2.48}{2.74} \times 100 = 90.5$  per cent for  $I_m$  and  $I_0$  respectively.

#### Modified Duane and Laborde Formula.

It is obvious that the observed ratios for the various sizes of chambers presented in Fig. 13 must be corrected for the saturation effect before a true comparison can be made with Duane and Laborde ratios.

Since the electrometer is limited to about 250 volts of collecting potential, there is a good deal of uncertainty involved in the extrapolation of the curves of determining the percentage of saturation. It is therefore proposed to use the calculated values (see pp. 50) for the percentage of saturation. Although in calculating these values, the formula for the electrical field of a cylinder was used and cannot strictly

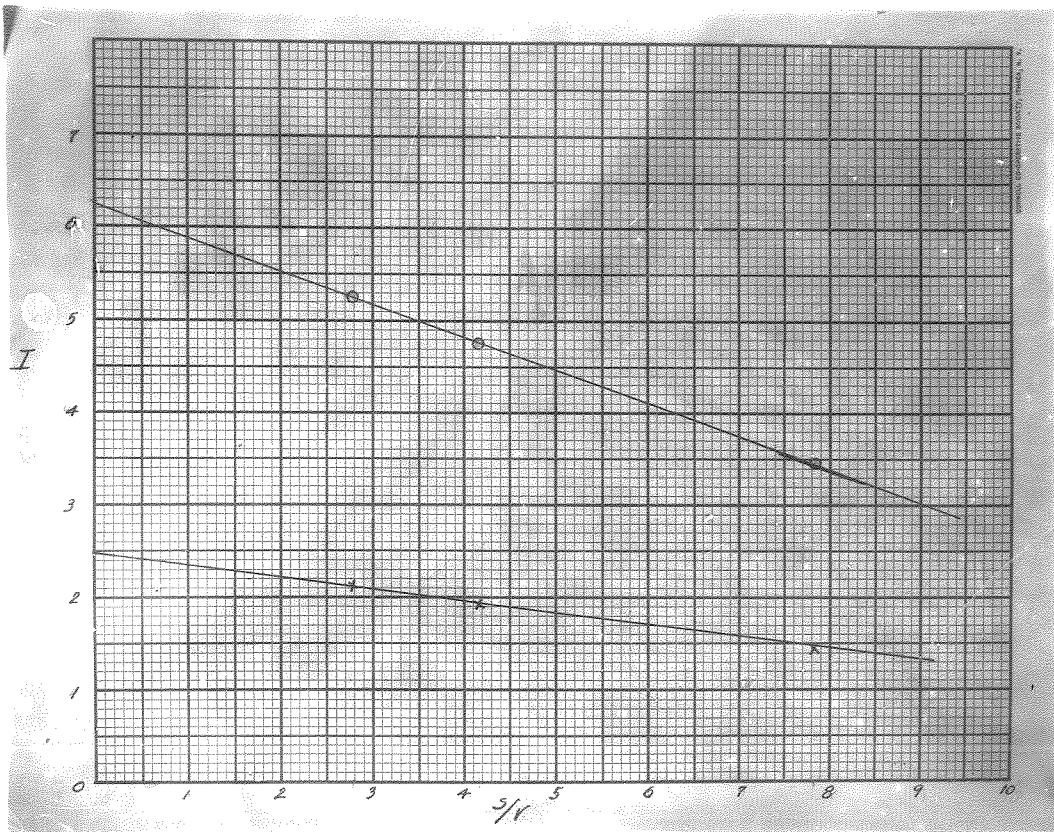


Fig. 22.

The Duane and Laborde formulas as linear functions.

$\circ$  = max. ionization  $x$  = initial ionization.

apply because of the field distortion at the ends of the ionization chambers, it is however, justifiable to use the calculated values because we are only concerned with their ratios.

After making the proper corrections as indicated in Fig. 23, it was found that the Duane and Laborde formula for ionization chambers whose dimensions are, diameter 6 inches, height 6-8 inches, and diameter 10 inches and height 6 inches was good to within 1.3 per cent.

Thus in calibrating an alpha ray set up for the determination of radium, the following formula should be used,

if,  $n$  = number of curies of radon

$C$  = total capacity of system

$\frac{dv}{dt}$  = potential change in e.s.u. per second 3 hours after

introduction of radon in ionization chambers, i.e.,

when radon is in equilibrium with its decay products.

$\phi$  = percentage of saturation

then,

$$n = \frac{\frac{dv}{C dt}}{6.27 (1 - 0.572 \frac{S}{V}) 10^6 \phi}$$

Ratio of Ionization Chambers	$\frac{6 \times 6}{5}$	$\frac{6 \times 12}{5}$	$\frac{10 \times 6}{5}$	$\frac{10 \times 12}{5}$
Ratio of Saturations $\frac{S_{dah}}{S_s}$	0.99	0.99	0.904	0.904
Ratio of observed Ionizations $\frac{I_{dah}}{I_s}$	0.962	0.985	0.956	1.06
Ratio of corrected Ionizations $\frac{I_{dah} \div \frac{S_{dah}}{S_s}}{I_s}$	0.972	0.995	1.06	1.17
Ratio of Duane and Laborde Ionization $\frac{I_{dah}}{I_s}$	0.985	1.035	1.06	1.11
Percentage deviation from corrected I.	1.3%	4.0%	0.0%	5.1%

Fig. 25

Indicating a comparison of the observed ionization of various chambers with that given by the Duane and Laborde formula. The ratios are corrected for saturation.

**Part II.**

**THEORY UNDERLYING GEOPHYSICAL APPLICATIONS**

**OF**

**RADON MEASUREMENTS.**

## THEORY UNDERLYING GEOPHYSICAL APPLICATIONS

## OF

## RADON MEASUREMENTS

Various assertions have been voiced with respect to the adaptability and geological significance of radioactive methods to geophysical prospecting. Without any clear indication as to their theoretical basis, statements have been made to the effect that a fault be evidenced by a higher radon concentration in the overlying soil<sup>1</sup>, that the radon content of soil in an oil bearing region is higher<sup>2</sup>, and that soil formations appear to have more or less characteristic radon contents<sup>3</sup>. It is obvious that if a theoretical explanation can be established for the above assertions, their value would be greatly enhanced and a more significant interpretation of radioactive data can be presented.

It is to be recognized that the measurements which have to do with the above suggested geophysical applications are concerned with radon, and bear only an indirect relationship to the actual radium content of rocks. Radon, the first decay product of radium, is a gaseous emanation of atomic weight 222, and half-life value of 3.82 days. Since radioactive substances disintegrate at a rate proportional to the quantity present, it can be shown that 30 days is required for the establishment of equilibrium between radium and radon, or in other words, that in 30 days just as much radon will decay as will be produced.

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1 F. Müller: Zeits. f. Geoph. 3, 330-6, (1927).

2 L. N. Begoyavlensky: Bull. Inst. Pract. Geoph. No. 3. 113-124, Leningrad, (1927).

3 Botset and Weaver: Physics, 2, 376-385, (1932).

If,  $t =$  time

$I_t =$  quantity of radon present at time  $t$

$I_0 =$  quantity of radon present at equilibrium

$\lambda =$  decay constant for radon

then,

$$I_0 = \frac{I_t}{1 - e^{-\lambda t}}$$

Thus in 30 days the decay factor  $e^{-\lambda t}$  becomes negligible, and which in turn indicates the time required for the establishment of equilibrium between radon and radium.

In general, all rock types with the possible exception of meteorites contain measureable quantities of radioactive substances. The radium concentration is, as a rule, of the order of magnitude of  $10^{-12}$  grams per gram of material. Visible evidence for the presence of radioactive elements and minerals often may be seen in thin sections of biotite, cordierite, tourmaline, chlorite, fluorite and others, where small circular darkened spots known as pleochroic halos are occasionally encountered. The halos are essentially due to the alpha radiation of minute crystalline inclusions usually zircon, allanite, or sphene. Careful scrutiny of the halos will sometimes reveal darkened concentric rings which correspond to the ranges of the alpha radiation.

Piggot<sup>1</sup> has ascertained in the case of a single granite with which mineral constituents the radioactive substances are associated. Using an aqueous solution of cadmium borotungstate (Kline's solution) he separated the various mineral grains of the

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1 Piggot: *A.J.S.*, 17, 13-34, (1929)



pulverized granite according to their densities as quartz, plagioclase, potash feldspar (mostly microcline), biotite, and muscovite. Radium analyses of the individual portions indicated concentrations of radium in ratios as quartz: plagioclase: microcline: biotite: muscovite: = 1 : 87 : 59 : 454 : 201. Other significant conclusions from the same experiment were that part of the radon may be leached out of a granite by water and that apparently some of the radioactivity is associated with the interstices between the grains, and with the surfaces of the grains of the constituent minerals, rather than with their interiors. The above results have immediate pertinency to the interpretation of radon measurements of sedimentary rocks.

Consider the genesis of a series of sediments. Assume in a nearby igneous mass the source material for the sediments. Sedimentary differentiation effected by the normal procedure of exogenous processes i.e., weathering, transportation of material, etc. will then act upon the igneous mass. The suite of sedimentary rocks, conglomerates, sandstones, and shales that will be consequent upon the erosion of the source mass will be composed of the ingredients of the source material, elutriatively selected, and of substances deposited through chemical agencies. Even granting a general constancy of the concentration of the radioactive substances in the igneous source, it is hardly conceivable that the resulting sedimentary rocks will maintain a similar uniformity. Variations might prevail during the conditions of deposition are too numerous to state.

W. Cogen<sup>1</sup> in a study of the lateral variation of heavy

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1 W. Cogen: Unpublished manuscript, C.I.T.

minerals in certain horizons has shown that differences in the concentration of heavy minerals as great as several hundred per cent in a single horizon within a mile may be expected. The composite minerals were also found to vary relative to each other. Since it has been indicated that radioactive substances are largely confined to the heavier minerals constituents, obviously then there is no more reason to believe that a sedimentary formation should have a more or less characteristic radium content than either titanium or zirconium content.

Indications from numerous analyses of the radium content of ocean sediments by Joly, Pettersson, and Piggot<sup>1</sup> likewise furnish further testimony substantiating the view that there may be considerable diversity in the distribution of radioactive constituents in sediments.

The fact that, in making radioactive determinations relevant to the geophysical suggestions offered in the introduction, measurements are made on radon adds further complexity to the problem. The amount of radon that will be available for measurement will be a function of the concentration of radium and its emanating power. The emanating power has been defined and discussed elsewhere (pp. 9 ) and it was concluded that temperature, pressure, humidity, may affect the amount of radon that will be emitted. Presumably the chemical and physical state of the rock minerals such as degree of alteration and particle size may also affect the emanating power.

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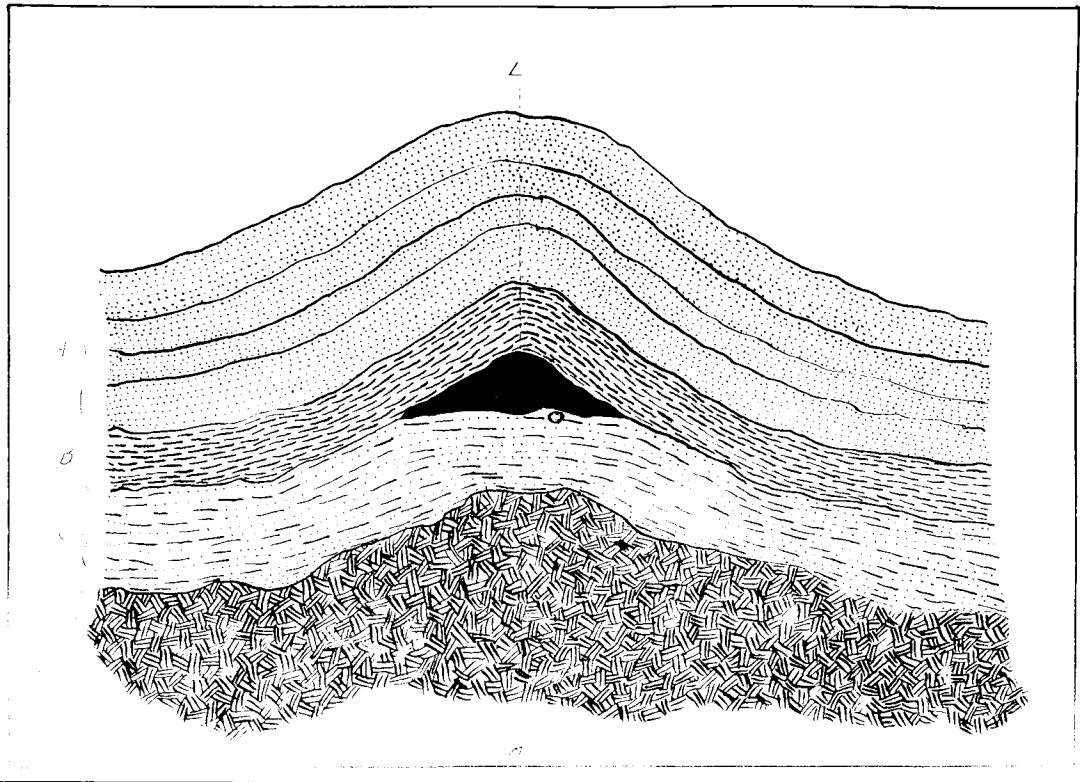
1 Piggot: A.J.S., 25, 229-238, (1933)

An attempt was made to determine the emanating power of a biotite granite when pulverized. The rock was screened to pass through 115 mesh and be retained on 250 mesh. 22.3 grams of the screen rock whose radium concentration determined by R.W. Raitt as  $1.6 \times 10^{-12}$  gm/gm were placed in test tube that contained 100 cc. of water and sealed. After shaking well the radon content of the water was determined and indicated an emanating power in the pulverized granite of less than one per cent.

In conclusion then, if a sedimentary formation does contain a constant radium concentration, there is no reason to believe that the emanating power of the rocks will be constant and in turn effect a constant radon emittance. Moreover with a heterogeneous distribution of the radioactive constituents, obviously there can be no correlation or simple geophysical interpretation of radon measurements.

#### Radon and Oil Bearing Region

With respect to the statement that an oil bearing region will be evidenced by a high radon concentration in the overlying soils, consider the simplest and most ideal conditions under which oil may accumulate. Given a symmetrical anticline with formations A B C D (fig. 24) of uniform radium concentration and emanating power. Assume for the moment that there has been no oil accumulation at O; then every sample on line L M in general should have the same radon concentration. With an accumulation of oil beneath the impervious layer S, radon will dissolve in the oil according to Henry's law.

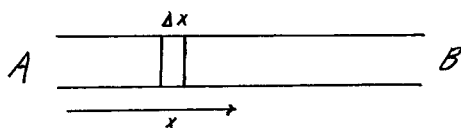


**Fig. 24**

The distribution ratio for radon between crude oil and air at 20°C is probably around 10  $\frac{\text{concentration in oil}}{\text{concentration in air}} = 10^1$  which merely indicates that during equilibrium conditions, there will be ten times as much radon in the oil as in the surrounding soil gas. This, however, will in no manner influence the radon concentration of the soil gas above or below the oil accumulation, and under the given conditions any sample along L M will have a constant radon concentration.

Assume some singular property of the oil accumulation such as a high radium concentration which will in turn effect a high radon concentration. Then as a consequence of the diffusion of radon, considered qualitatively, the radon concentration of the soil gas should increase approaching the oil body. Considered quantitatively, the unfeasibility of this though can be readily established.

#### Diffusion of Radon



In the above let A B be a cylinder of unit cross section filled with an inactive gas and radon from source where  $x=0$  is diffusing uniformly along the cylinder. Let D be the coefficient of diffusion of radon through the gas, and N be the number of atoms per cubic centimeter at a distance x from a fixed point along the axis of the tube. Then in consequence of the diffusion,

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1 Meyer and Schweidler: Radioaktivitat, pp. 142

$\frac{dN}{dt}$  the rate of increase in the number of atoms of radon per cc. will be,

$$\frac{dN}{dt} = \frac{\left[ -D \left( \frac{dN}{dx} \right)_x \right] + \left[ D \left( \frac{dN}{dx} \right)_{x+\Delta x} \right]}{\Delta x}$$

if  $\frac{dN}{dx} = y$

$$\frac{dN}{dt} = \frac{D(y_{x+\Delta x} - y_x)}{\Delta x}$$

$$= D \frac{dy}{dx}$$

$$= D \frac{d^2N}{dx^2}$$

but radon is decaying in accordance with its constant,  $\lambda$ , and therefore,

$$\frac{dN}{dt} = D \frac{d^2N}{dx^2} - \lambda N$$

because of the disintegration of radon there will be a point at which  $\frac{dN}{dt} = 0$  and,

$$D \frac{d^2N}{dx^2} = \lambda N$$

The solution of this differential equation is as follows,

$$N = a e^{-kx}$$

$$\frac{dN}{dx} = -ak e^{-kx}$$

$$\frac{d^2N}{dx^2} = ak^2 e^{-kx}$$

when,

$$x = 0$$

$$N = N_0$$

$$a = N_0$$

$$k = \sqrt{\frac{\lambda}{D}}$$

or

$$N = N_0 e^{-\sqrt{\frac{\lambda}{D}} x}$$

Using the values,  $\lambda = 2.1 \times 10^{-6}$  /sec. and  $D = 0.1$ , the point of which radon will fall to half its original value will be approximately 135 cm. This value has been calculated using the coefficient of diffusion for radon into air, and considering the through an impervious layer trapping an oil accumulation the estimate of 135 cm. will be considerably lessened.

It may be justly indicated therefore, the the conclusions deduced a priori from the report that the radon concentration of soil gas is high in an oil bearing region cannot genetically correlate the two and have a practical geophysical significance.

#### Raden and Faults

With respect to the statement that the radon content of the soil increases toward a fault, consider fig. 25 where formation A is of a uniform radium concentration, B is the basement complex also considered to have a uniform radium concentration, line C is a water table level and F is a fault with an indicated zone of brecciation. It may be entirely possible that on approaching the zone of brecciation, the radon content of the soil will increase. This can probably be explained by the logical assumption that the emanating power of the rocks increases due to their increased surface. In this respect it should be mentioned that grinding a rock to pass through 115 mesh did not perceptibly increase the emanating power. However because of the small quantity of material used, an increase of 0.1 per cent or 0.2 per cent might have been easily been obscured, and such an increase,

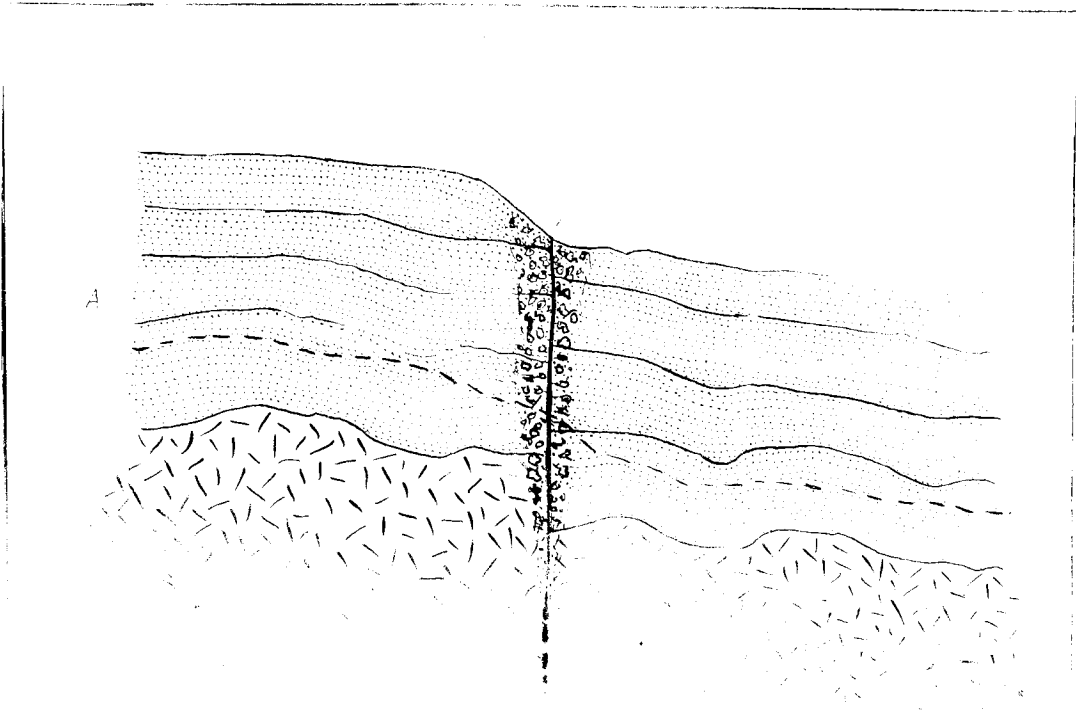


Fig 25



when the quantities involved are large as in a fractured fault zone, would be of considerable importance.

Raden soil measurements usually involved so many uncertain factors such as textural relation of the grains, degree and nature of cementation and effect upon porosity, difficulties in accurate sampling, etc., that it is highly improbable that a satisfactory explanation of these data can be obtained. It was suggested, however, by Mr. R. Engel<sup>1</sup> that radon measurements of underground waters would eliminate a number of these uncertainties and offer a more significant interpretation, especially in relation to faults.

In considering fig. 25 in regard to the radon content of underground waters, obviously because of the intimate contact of the water and grains as the water seeps through the soil in response to its gradient, a more representative radon measurement can be obtained. The absolute radon measurement of the soil will of course differ from the water measurement in accord with Henry's law. It should be here indicated that any radium concentration of the water which in turn would affect the radon content is probably negligible since the solubility of radioactive minerals is very small in water.

On approaching the brecciated zone, a greater dissolving action will be exerted, and a corresponding increase in the radon content should be expected at the fault. A similar increase is to be expected for any other chemical constituent that might be dissolved but because of the direction of flow and diffusion, these ions will soon become uniformly distributed, so that it

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1 R. Engel and L. J. Bohn: (abst.) G.S.A.B., 41, 154, (1930)

will be impossible to detect an increase toward the fault. Undoubtedly an important factor in this respect is the relative insolubility of rock minerals in ordinary waters, and the small differences in the concentration of the dissolved constituents at or near the fault even when diffusion and distribution effects have not acted would be extremely difficult to detect.

There will always be a high radon concentration at the fault in the above case since the disintegration of radon is more important than either diffusion or dissipation by seepage. In 3.82 days radon decays to half its value and in consequence as diffusion and dissipation play their part, the radon carried from the fault zone will have disintegrated to the normal concentration in water which will be in equilibrium with the surrounding rocks.

In the above discussion concerning the relation of radioactive content of underground waters to faults two fundamental assumptions have been made; first, a zone of brecciation must accompany a fault and second, that the distribution of radium constituents is homogeneous. Presumably neither of these conditions need necessarily be the case and therefore, a fault may or may not be revealed by high radon content.

An important consideration in the relation of radioactivity to faults has been purposely unincorporated in the above discussion and is concerned with the effect of waters of magmatic or juvenile origin. These waters, existing originally in the molten silicate solution constituting a magma, have been in intimate contact with

all of the various chemical constituents of the magma, and from the very fact that magmatic gases as helium, carbon dioxide, hydrogen sulfide and others, are contained in such waters, it may without undue reason be said that juvenile waters will contain a relatively high radon content. If such is the case, and provided the waters are able to ascend a fault plane or zone with sufficient rapidity so as to overcome the decay factor of radon, a fault will undoubtedly be detected by a high radon content in its immediate vicinity.

Work in progress by Mr. R. Engel will help to elucidate some of these factors.

**Part III.**

**RADON CONTENT OF UNDERGROUND WATERS**

**AND**

**ITS RELATION TO THE RAYMOND FAULT**

RADON CONTENT OF UNDERGROUND WATERS AND ITS RELATION TO THE  
RAYMOND FAULT

Technique for the Measurement of Radon in Solutions

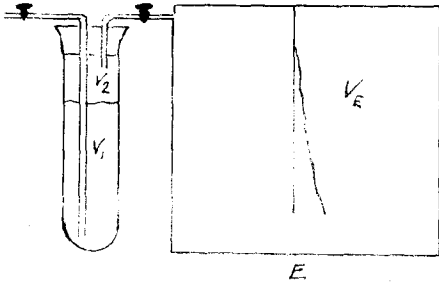
The method employed for the measurement of the radon content of solutions for the early part of the work was except for a single modification identically similar to that used by Evans<sup>1</sup>. The electrometer, reflux condenser, ionization chambers, etc., have been described in detail and will not be considered here. The modification has been to discard the displacing liquid and instead a volume of air equal to the volume of the evacuated ionization chamber is bubbled through the boiling solution for sweeping the radon into the chamber.

Because of the inconvenience of 2 liter samples for field work, a simplified was adopted for the latter part of the work. This procedure will be briefly described after the following consideration of the principles involved in the removal of radon from solutions.

Removal of radon from solutions: Radon is a gas and as such conforms to Henry's law and will be removed from solutions in obedience to this law.

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1 Evans: Ph. D. thesis C.I.T.  
Rev. Sci. Inst. 4, 216, (1933)



Let  $V_E$  = volume of electroscope

$V_1$  = volume of solution

$V_2$  = volume above solution

to stop-cock

$m$  = radon in solution

$m_t$  = total amount of radon

to stop-cock

$\alpha$  = distribution ratio for radon

Electroscope in evacuated and a slow stream of air is bubbled through the solution.

$dV$  admits  $dm_t$  of radon into E

$$\text{but } dm_t = -\frac{\alpha m}{V_1} dV$$

$$\begin{aligned} m_t &= m + C_2 V_2 \\ &= m + \alpha m V_2 \\ &= m \left( 1 + \alpha \frac{V_2}{V_1} \right) \end{aligned}$$

$$dm_t = \left( 1 + \alpha \frac{V_2}{V_1} \right) dm$$

$$\text{or } \left( 1 + \alpha \frac{V_2}{V_1} \right) dm = -\frac{\alpha m}{V_1} dV$$

$$\frac{dm}{m} = -\frac{\alpha dV}{V_1 + \alpha V_2}$$

$$\alpha = \frac{C_2}{C_1} = \frac{(\text{conc. in } V_2)}{(\text{conc. in } V_1)}$$

then integrating from  $m_0$  to  $m$  and from 0 to  $V_E$ , where  $m_0$  = original quantity of radon in solution

$$m = m_0 e^{-\frac{\alpha V_E}{V_1 + \alpha V_2}}$$

A simple experiment was performed to test the applicability of this formula. 2.926 grams of carnotite of uranium content 1.58 per cent by weight were placed in a test tube of 200 cc. capacity, dissolved in HCl, diluted to 150cc. with distilled water, and sealed. After 25 days the percent of removal of radon was tested by merely bubbling through the solution a volume of air equal to the standard ionization chamber (see pp. 30) and as soon as this operation was completed, a second standard chamber was similarly filled. The time difference between the filling of each chamber was ten minutes. The ionization was measured by the use of Wulf bi-filar electrometer<sup>1</sup> and the corresponding readings were 1545 div./hr. and 15.3 div./hr., indicating that the radon was 99 per cent removed from the solution.

Fig. 25 indicates diagrammatically the simplified set up employed. An electroscope was used and standardized against the Wulf instrument whose calibration factor had been accurately determined. The difference in sensitivity between the electroscope and Wulf electrometer is indicated by their respective calibration constants,  $11 \times 10^{-13}$  curies/div./hr. and  $73 \times 10^{-13}$  curies/div./hr. A fine hair was attached to the leaf to facilitate the reading of the electroscope.

Fig. 26 shows to what accuracy the duplication of results from similar samples may be expected by the method.

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1 Evans: loc. cit.

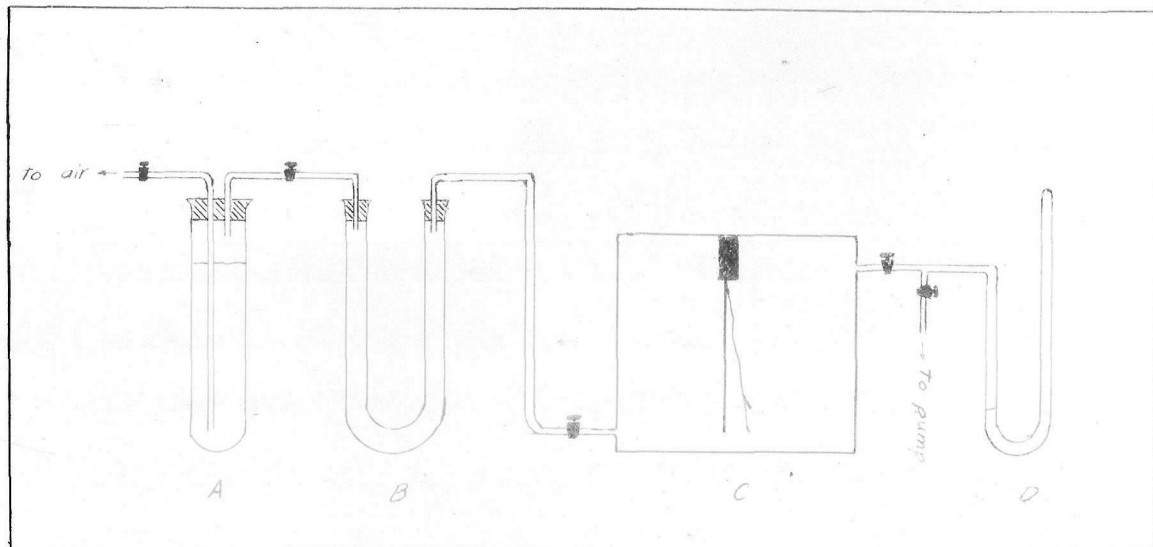


Fig. 26

Diagram of the simplified technique for the measurement of radon. A = sample, B =  $\text{CaCl}_2$ , C = electroscopes with fine hair attached to leaf to facilitate reading, D = manometer.



Test	volume of sample cc.	rate of discharge dir. / hr.	decay $e^{-\lambda t}$	radon content curies/liter $\times 10^{-10}$	deviation
A <sub>1</sub>	141	15.9	1	1.23	3.0%
A <sub>2</sub>	141	12.8	.834	1.19	
B <sub>1</sub>	146	24.2	1	1.81	3.2%
B <sub>2</sub>	154	24.8	.968	1.82	
B <sub>3</sub>	150	22.6	.863	1.90	
C <sub>1</sub>	152	17.9	1	1.30	This value is probably due to an error in sampling
C <sub>2</sub>	142	16.7	.966	1.34	
C <sub>3</sub>	146	13.0	.837	1.17	
C <sub>4</sub>	140	14.1	.810	1.36	
D <sub>1</sub>	142	20.1	1	1.55	0.65%
D <sub>2</sub>	135	15.4	.803	1.56	

**Fig. 27**

**Indicating the percentage of deviation of check samples by the simplified technique of measuring radon.**

The volume of the electroscopes in the simplified set up amounted to only 1250 cc. and although the formula indicates virtually complete removal of radon from a 150 cc. sample in a 200 cc. test tube, it would be perhaps advisable to have a larger volume for the electroscopes. This would act as a factor of safety against a too rapid bubbling which would not permit equilibrium in the bubble of air according to Henry's law. It should be pointed out that because of the small sample, the volume of the sample should be measured accurately, and a weighing method has been suggested by Mr. R. Engel. A number of check samples analyzed by the boiling method of Evans and by the simplified technique were found to be in excellent agreement.

#### Raymond Fault

A low but prominent escarpment delineating the Raymond fault traverses the southern portion of the city of Pasadena. It is relatively recent origin, faulting Miocene sediments, and is still active. See map. Numerous water wells are located north of the fault and with lesser abundance to the south of it, and offered an excellent opportunity to study the distribution of radon content of underground waters in relation to faults.

A number of wells were sampled and analyzed, attempting as much as possible to select a suite of wells in a linear profile perpendicular to the fault. The wells selected had to be sampled

the same day since the radon concentration varies from day to day. Only those wells that had been pumping for some time could be used as diagnostic samples, since stagnant well water will lose most of its radon either by diffusion into the atmosphere or by decay.

The results of these surveys are presented in figs. 27, 28 and 29. The importance of correct sampling is indicated by the analysis of well no. 227. At this particular well the water is pumped by means of an air lift and at the discharge the water is extremely well aerated; as a consequence an analysis for the radon content indicated a negligible quantity present.

No systematic increase of the radon content in the underground waters towards the Raymond fault was discovered.

sampled 12/10/32		sampled 12/20/32		sampled 4/12/33		sampled 4/17/33		sampled 4/22/33	
well	radon curies/liter x 10 <sup>-10</sup>	well	radon curies/liter x 10 <sup>-10</sup>	well	radon curies/liter x 10 <sup>-10</sup>	well	radon curies/liter x 10 <sup>-10</sup>	well	radon curies/liter x 10 <sup>-10</sup>
94	2.82	82	2.69	221	4.58	8	1.67	8	1.87
84	3.98	95	4.71	33	4.13	7	3.80	7	1.54
120	2.97	150	4.25	3a	1.92	24	1.18	24	1.85
227	0.003	110	4.60	2a	2.01	203	5.80	203	4.68
220	3.74	201	3.96	1a	4.24	81	2.57	81	5.35
110	2.14								
228	4.10								

Fig. 28

A summary of the surveys of the radon content of underground waters and their relation to the Raymond fault.

cf. maps # 1 & 2  
in pocket at end  
of Thesis

sampled 12/10/32

sampled 12/20/32

abscissa = perpendicular distance to fault

Radon Content  $\times 10^{-10}$  curies per liter

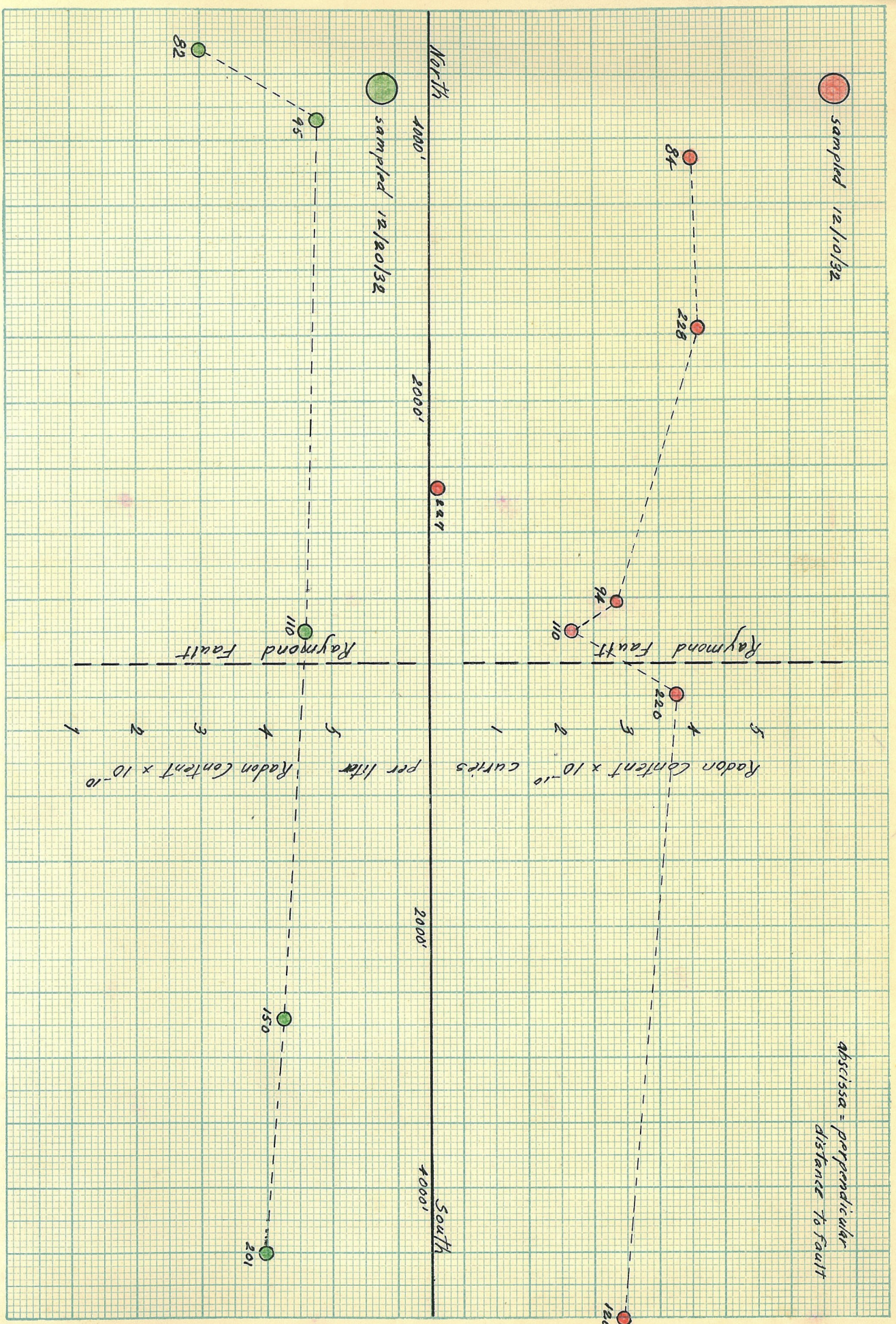


Fig. 29

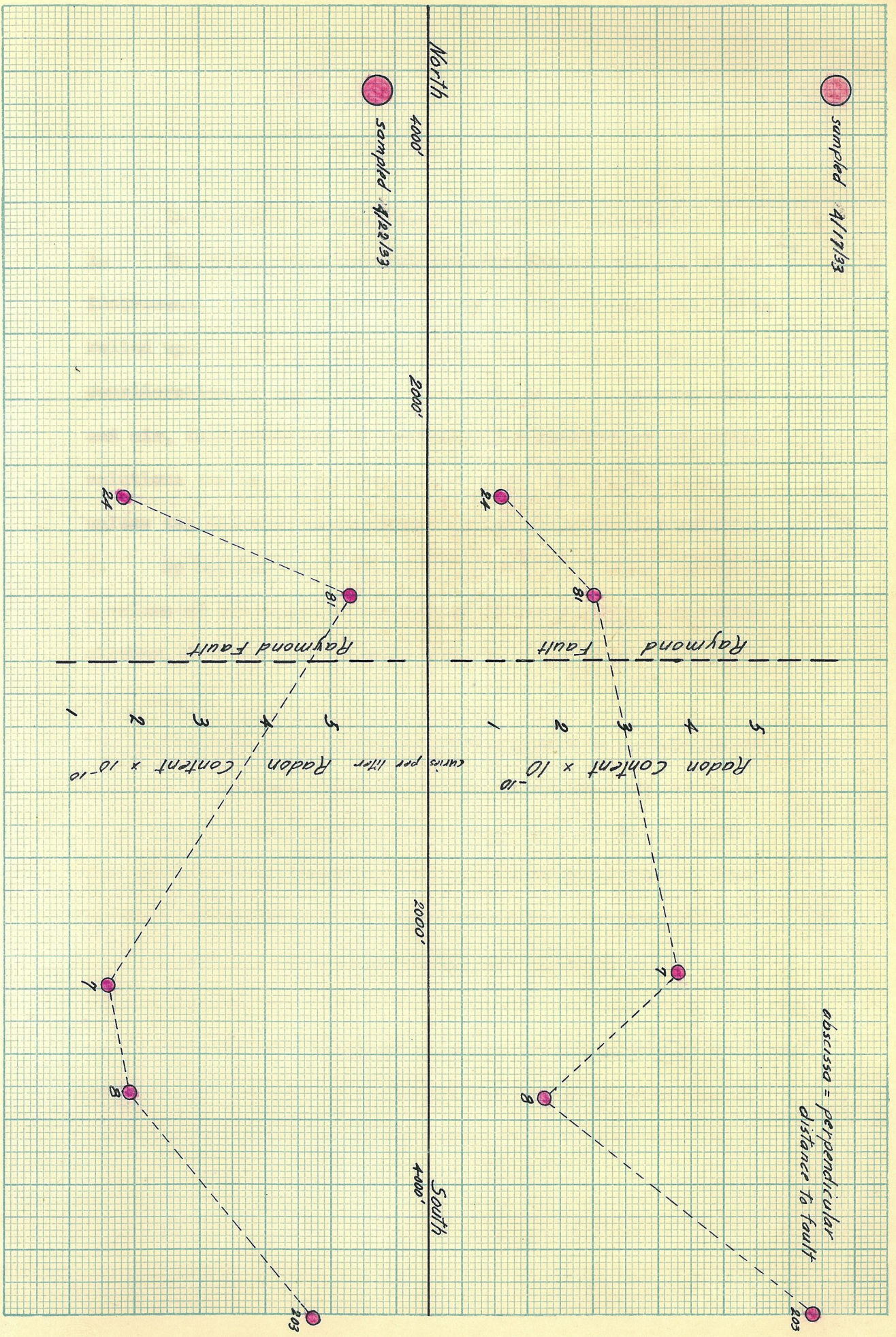


Fig. 30

## CONCLUSIONS

In summary, the following results are indicated:

1. In using a standard solution for the calibration of the instrument, a considerable error may be made if only boiling is relied upon to remove the radon from the solution. By consideration of the distribution ratio of radon between water and air, this error may be avoided. The instability of radium solutions may cause considerable error in the calibration, unless the solution is protected by barium in an acid solution.
2. By boiling a solution and moreover aspirating through it a volume of air equal to the volume of the standard ionization chamber, 3190 cc., it is shown that the removal of radon is complete.
3. The Ra/U ratio in radioactive minerals is a function of time and geological environment. In pitchblende the ratio is quite constant and may be taken as  $3.4 \times 10^{-7}$ . In carnotite the ratio deviates markedly from the theoretical value. Thus pitchblende may be used for standardization. In preparing a standard solution from pitchblende, oxidation of sulfur impurities should be avoided, for  $\text{RaSO}_4$  will occlude radon. (pp. 5-9)
4. The emanating power of carnotite is a function of humidity, temperature, and pressure. Within the limits of variation of these factors during the experiments, it was found that the

emanating power increased with these variables. (pp. 32) However by heating carnotite to 400°C for one half hour the emanating power was decreased to 1/10 of that of unheated carnotite, but the temperature coefficient of the emanating power was not noticeably changed. (pp. 11) This indicates a discontinuity in the emanating power - temperature relationship and may possibly be ascribed to a loss of water of crystallization. (fig. 3)

5. A modified Duane and Laborde formula was established for calibration,

if,  $n$  = number of curies of radon

$C$  = total capacity of system

$\frac{dv}{dt}$  = potential change in e.s.u. per second three hours after introduction of radon in ionization chamber, or when radon is in equilibrium with its decay products

$\theta$  = percentage of saturation

then,

$$n = \frac{C \frac{dv}{dt}}{6.27 (1 - 0.572 \frac{S}{V}) 10^6 \theta}$$

It was found that the above formula is reliable within an error of 1.3 per cent for ionization chambers whose dimensions are, diameter 6 inches, height 6-8 inches, and diameter 10 inches and height 6 inches. (fig. 23)

6. Because of a number of variables, as emanating power, heterogeneity of mineral constituents, textural relationships,



cementation, etc., a soil formation is not expected to have a uniform radon content.

7. A high radon concentration in the overlying soil of an oil bearing region cannot be correlated genetically with the oil.

(pp. 60)

8. The radon content of the soil and underground water in a fault region may be high in comparison with the surrounding region if there is a pronounced zone of brecciation. Magmatic influences such as juvenile waters of high radioactive content may be an important factor. (pp. 64-67)

9. A simplified technique for the measurement of the radon content of solutions has been found to be satisfactory and adaptable to field work. (fig. 25)

10. Surveys of the radon concentration of underground waters of Pasadena, indicated no systematic increase towards the Raymond fault. (figs. 27,28,29 )