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CODEWORD
AN APPARATUS FOR

THE PURIFICATION OF RADON.
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As is well known, radium has been used for many years as a source of gamma radiation for therapeutic use. The method consists in placing needles of radium in suitable positions on and around the cancerous growth; dosage being controlled by the radium content of each needle, and its time of application. Such operations are extremely dangerous, in that the needles are small and easily lost; yet the radium decays very slowly so that the intensity of radiation emitted by the needles remains almost constant. The high cost of such needles also limits their use.

An alternative method of gamma ray therapy utilizes needles of radon, the radioactive gas which is the first decay product of radium. Radon has a half life period of 3.825 days, compared with that of radium, of 1580 years. Consequently its activity is appreciable only over its first few days, having fallen to one per cent of its initial value in 25 days. The danger factor is thereby largely eliminated, since lost radon needles would be quite safe, even if still inside the patient, after a month or so. Moreover, the dosage can be arranged so that the needles are left permanently in the patient, the dose integrating to the required amount in infinite time. In some cases, where the tumour is rather inaccessible, this method is very convenient.

A further advantage of radon lies in the fact that it is a gas, and can therefore be compressed to small sources of any required shape or size. Thus the radium from which it is prepared is rendered many times more useful, all types of needles being available from a common source. This extends its use to many cases not otherwise capable of treatment, as it will be appreciated that the cost of a complete stock of all types of radium needles is prohibitive. The more so, as many of them would remain out of use for years.

Radon has also been used recently in the preparation of a radioactive
ointment, emitting a weak alpha radiation. A solution of radon in vaseline is the usual type, and it appears to have a marked beneficial effect on certain radio necrosis cases, caused through excessive exposure to gamma radiation or X rays.

For these reasons, and certain others, it has been decided to construct a plant in this Dominion, for the supply of radon from radium. Such a plant was set up in Wellington some years ago, but met with an accident, and was then dismantled. A portion of the radium used in the above plant has been made available for the present purpose. The object of the present work is then, to construct an apparatus for the isolation and purification of radon from radium. The following is a brief summary of the relevant points:

(1) Some 300 milligrams of radium are available, as radium chloride, mixed with an unknown amount of barium chloride, and other impurities of an uncertain nature.

(2) The plant is to be permanently established, as a constant demand for the radon is anticipated.

(3) The radon must be obtained in sufficient purity for therapeutic use in needles, implants etc. Without going into details, it can be stated that 1% purity is adequate for this, but a higher degree of purity would be desirable for research purposes.

(4) In view of the danger of radiation, all operations must be effected with a minimum of exposure to the radon, and its parent radium. In the present work, the tolerance dose has been taken as 1 r. per week (1), and every effort has been made to keep well below this figure. An estimate of the "r" unit is gained from the following statement:

The dose received in one hour, at a distance of one centimetre from one milligram of radium, filtered by 0.5 mm. Pt, is 8.3 r.
THE RADIOACTIVITY OF RADON.

In this section some account will be given of the radiations met with in the radium series of radio-elements. A brief mathematical treatment will be necessary to explain the calculation of yields of radon from radium, and for other reasons which will be apparent.

A section of the radium series is shown below, together with the half lives of each member, and the nature of the radiation emitted in each transformation:

\[
\begin{align*}
\text{Ra} & \xrightarrow{\alpha} \text{Ra}_2 & \xrightarrow{\alpha} \text{Ra}_A & \xrightarrow{\alpha} \text{Ra}_B \\
\beta & \xrightarrow{\beta} \text{Ra}_C & \xrightarrow{\beta} \text{Ra}_D & \xrightarrow{\beta} \text{Ra}_E \\
\text{Ra}_2 & \xrightarrow{\gamma} \text{Ra}_C'' & \xrightarrow{\gamma} \text{Ra}_C' & \xrightarrow{\gamma} \text{Ra}_C \\
\text{Ra}_C & \xrightarrow{\gamma} \text{Ra}_C'' & \xrightarrow{\gamma} \text{Ra}_C' & \xrightarrow{\gamma} \text{Ra}_C
\end{align*}
\]

The short range of alpha and beta rays renders them harmless when filtered by a millimetre of lead or its equivalent. Gamma radiation is of primary importance, not only because of its penetrating power and consequent danger, but because it is this, alone, which is required for therapeutic use. Moreover, the present work utilises gamma radiation for measuring purposes.

Radium decays with the emission of \( \alpha, \beta \), and a very weak \( \gamma \) radiation, into radon. Radon disintegrates into Radium A, and both of these emit \( \alpha \), but no \( \gamma \) radiation. Radium B, formed by the decomposition of Ra A, emits an appreciable \( \gamma \) radiation, and Ra C, the next member displays an intense \( \gamma \) activity. Ra C decays by two mechanisms, forming 0.04% of Ra C'' to 99.96% of Ra C'. The \( \gamma \) activity of Ra C'' is negligible, while Ra C' emits only \( \alpha \) radiation. The later members are governed by the long half life of Radium D, and radiate only very weakly.

Thus the \( \gamma \) ray activity of a radon or radium preparation is controlled by the amount of Ra B and Ra C present in the system, and is in fact directly
proportional to these quantities.

Mathematical treatment.

Consider a system starting with \( N_0 \) atoms of radium, isolated from its mother element. After \( t \) seconds, let there be \( N_1 \) atoms of Ra present.

The rate of decay of these \( N_1 \) atoms will be given by the classical equation:

\[
- \frac{dN_1}{dt} = \lambda_1 N_1
\]

which integrates to

\[
N_1 = N_0 e^{-\lambda_1 t}
\]

where \( \lambda_1 \) is the decay constant of radium, related to its half life period, \( T \), by the equation:

\[
\frac{1}{\lambda_1} = \frac{\ln 2}{\log_e 2}
\]

Now let \( N_2 \) be the number of atoms of Ra present after \( t \) seconds.

This radon is being formed by the decomposition of Ra, and is decaying at a rate proportional to \( N_2 \).

Thus

\[
- \frac{dN_2}{dt} = \lambda_2 N_2 - \lambda_1 N_1
\]

This will be integrated in full, as it is required for further reference.

Substituting (2) in (4), and rearranging,

\[
\frac{dN_2}{dt} + \lambda_2 N_2 = \lambda_1 N_0 e^{-\lambda_1 t}
\]

Multiplying by the integrating factor \( e^{\lambda_2 t} \),

\[
e^{\lambda_2 t} \frac{dN_2}{dt} + \lambda_2 e^{\lambda_2 t} N_2 = \lambda_1 N_0 e^{(\lambda_2 - \lambda_1) t}
\]

\[
\therefore e^{\lambda_2 t} N_2 = \int \lambda_1 N_0 e^{(\lambda_2 - \lambda_1) t}
\]

\[\ldots\ldots(5)\]
A general integration of this may be written:

\[ N_2 = \frac{\lambda_1 N_0}{(\lambda_2 - \lambda_1)} \left\{ e^{(\lambda_2 - \lambda_1) t} - e^{\lambda_1 t} \right\} + K \] \hspace{1cm} \ldots \ldots \ldots (6)

Where \( K \) is an integration constant. Assuming that initially the system is free from radon (i.e., consists only of radium), then

\[ N_2 = 0 \quad \text{when} \quad t = 0 \]

so that

\[ N_2 = \frac{\lambda_1 N_0}{(\lambda_2 - \lambda_1)} \left\{ e^{\lambda_1 t} - e^{\lambda_2 t} \right\} \] \hspace{1cm} \ldots \ldots \ldots (7)

Since \( \lambda_1 \) is extremely small in the case of radium, the above equation may be written as the very good approximation:

\[ N_2 = N_0 \left( \frac{\lambda_1}{\lambda_2} \right) \left( 1 - e^{\lambda_2 t} \right) \] \hspace{1cm} \ldots \ldots \ldots (8)

This equation gives the theoretical yield of radon \( (N_2) \) formed after \( t \) seconds from \( N_0 \) atoms of radium. It is customary, however, to express the amounts of radon in curies. The curie is defined as that amount of radon present in a system in equilibrium with one gram of radium, isolated from its (radium's) mother element.

As \( t \) increases, in equation (8) above, the factor \( e^{-\lambda_2 t} \) becomes vanishingly small, so that the equilibrium yield of radon from \( N_0 \) atoms of radium will be given by

\[ N_2 \text{ equilib.} = N_0 \frac{\lambda_1}{\lambda_2} \] \hspace{1cm} \ldots \ldots \ldots (9)

and if \( N_0 \) is expressed in milligrams, then \( N_2 \) will necessarily be in milli-curie. Thus the theoretical yield, \( R \), of radon in milli-curie, from \( N \) milligrams of radium will be given by

\[ R = N \left( 1 - e^{\lambda_2 t} \right) \] \hspace{1cm} \ldots \ldots \ldots (10)
As may be anticipated, tables of the function \( (1 - e^{-\lambda_2 t}) \) are available for all values of \( t \), so that theoretical yields are readily calculated.

The \( \gamma \) ray activity of radon.

As has been stated above, it is proposed to measure the radon by means of its gamma radiation. Evidently some correlation of this property with the amount of radon present is required, not only for purposes of measurement, but as a guide to the proper safety precautions to be adopted in the operation of radon plants. The gamma radiation proceeds almost entirely from Ra B and Ra C as explained above, and expressions for the growth of these radio-elements will be given.

Consider a system starting with \( N_x \) atoms of Ra, isolated from its parent radium, and from its decay products. Let the number of atoms of Ra A, Ra B, Ra C, present after \( t \) seconds be \( N_A, N_B, N_C \) with decay constants \( \lambda_A, \lambda_B, \lambda_C \) as follows:

- **Initially** \( N_x \) atoms of Ra, decay constant \( \lambda_R \)

<table>
<thead>
<tr>
<th>After ( t ) seconds</th>
<th>Element</th>
<th>( N_x ) of atoms present</th>
<th>Decay constant</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ra</td>
<td>Ra A</td>
<td>Ra B</td>
</tr>
<tr>
<td></td>
<td>( N_R )</td>
<td>( N_A )</td>
<td>( N_B )</td>
</tr>
<tr>
<td>Decay constant</td>
<td>( \lambda_R )</td>
<td>( \lambda_A )</td>
<td>( \lambda_B )</td>
</tr>
</tbody>
</table>

Following the same procedure as before, the following equations will obtain:

\[
\frac{d N_R}{dt} = \lambda_R N_R \\
N_R = N_x e^{-\lambda_2 t} \\
.............(11)
\]

\[
\frac{d N_A}{dt} = \lambda_A N_A - \lambda_R N_R \\
.............(11A)
\]

\[
\frac{d N_B}{dt} = \lambda_B N_B - \lambda_A N_A \\
.............(12)
\]

\[
\frac{d N_C}{dt} = \lambda_C N_C - \lambda_B N_B \\
.............(13)
\]

Repeated integration and substitution evolves the following equation for \( N_B \) and...
\[
N(t) = \frac{N_0}{\lambda} \left( \frac{1}{\lambda - \lambda_0} \right) + \frac{N_0}{\lambda} \left( \frac{1}{\lambda - \lambda_1} \right)
\]

\[
N(t) = \frac{N_0}{\lambda} \left( \frac{1}{\lambda - \lambda_0} \right) + \frac{N_0}{\lambda} \left( \frac{1}{\lambda - \lambda_1} \right)
\]

While each member of these expressions is of significance for small values of \( t \), it is found that after some five hours, the terms \( e^{-\lambda \ast t} \), \( e^{-\lambda \ast t} \), become negligible in comparison with the term \( e^{-\lambda \ast t} \). This is, of course, due to the small half lives of the active deposits Ra A, Ra B, and Ra C compared with that of radon itself. After such five hours then, the number of atoms of Ra C present will be given by the equation,

\[
N(t) = \frac{N_0}{\lambda} \left( \frac{1}{\lambda - \lambda_0} \right) + \frac{N_0}{\lambda} \left( \frac{1}{\lambda - \lambda_1} \right)
\]

A similar form of expression holds for \( N_B \), so that both of these follow the half lives of radon. The \( \gamma \) ray activity is proportional to the amounts of Ra B and Ra C, so that this quantity must also follow the half life of radon itself after five hours. It is not possible to give an absolute value to this \( \gamma \) ray activity, but comparison may be made with standard radium preparations, and measurements are based on this comparison. Thus a sample of radon may have an activity equivalent to 10 milligrams of radium at mid-day on a certain date. This statement affords sufficient information for dosage calculations, allowance being made for the decay of the radon; for it is this \( \gamma \) activity which is
utilised in therapy, and is also that property of the radon which is quantitative-
ly measured.

It is not, however, strictly correct to state that, in the above case, the radon content, as distinct from its gamma ray activity, of the specimen is 10 millicuries at mid-day on the same date, although this is very nearly so, as the following calculation will show:

The γ activity is mainly due to Ra C, and this element will be considered. Equation (17) gives the number of atoms of Ra C which will be in transient equilibrium with an amount of Ra, which consisted initially of $N_x$ atoms. The term "transient" is applied since this type of equilibrium is rather different from the secular equilibrium encountered with radium. Considering any interval of $t$ seconds, then the radon present at this time, corresponding to $N_C$ atoms of Ra C, will be given by

$$N_C = N_x e^{-\frac{\lambda_R t}{\lambda_{Ra}}}$$

Now consider the same amount of radon in equilibrium with radium and its decay products. The number of atoms, ($P$) of radium, yielding the above amount of radon at equilibrium will be given by

$$P = N_x e^{-\frac{\lambda_R t}{\lambda_{Ra}}}$$

where $\lambda_{Ra} = \text{decay constant of radium.}$

This result is obtained from equation (9). A similar calculation to that used in equation (17) will give the following expression for the number of atoms ($N^0_C$) of Ra C in equilibrium with $P$ atoms of Ra:

$$N^0_C = (\frac{\lambda_R}{\lambda_{Ra}}) N_x e^{-\frac{\lambda_R t}{\lambda_{Ra}}} \left\{ \frac{\lambda_{Ra}}{(\lambda_C-\lambda_{Ra})(\lambda_A-\lambda_{Ra})(\lambda_B-\lambda_{Ra})} \right\}$$

In this case, however, since $\lambda_{Ra}$ is exceedingly small, the equation reduces to
\[ N_C^0 = N_C e^{-\lambda_A t} \left( \frac{\lambda_R}{\lambda_G} \right) \]

Comparing this with (17), it is found that the ratio \( N_C : N_C^0 \) is given by (4):

\[ \frac{N_C}{N_C^0} = \left( \frac{\lambda_A \lambda_B \lambda_G}{\lambda_C^0} \right) \left( \frac{\lambda_G}{\lambda_C^0} \left( \frac{\lambda_B}{\lambda_R} \right) \left( \frac{\lambda_A}{\lambda_R} \right) \right) \]

and substituting the actual values:

\[ \frac{N_C}{N_C^0} = 1.0089 \]

In other words, the amount of \( \text{Ra C} \) in transient equilibrium with a quantity of \( \text{Ra} \) isolated from its parent radium, is higher than the amount of \( \text{Ra C} \) in secular equilibrium with the same quantity of radon by a factor of 1.0089; "secular" implying that the radon is, in the second case, in equilibrium not only with its decay products but also with radium. The same result must hold for the \( \gamma \) ray activity of radon, and leads to the interesting conclusion that one millieur of radon, in transient equilibrium, has a higher \( \gamma \) ray activity than does one milligram of radium.

Thus in the case above cited, it is evident that the actual content of the radon sample measured would be, not 10 millieurcs, but very slightly less. However, the factor is very small, when account has been taken of \( \text{Ra B} \), and is customarily neglected, so that radon of activity equivalent to \( "n" \) milligrams of radium will be expressed directly as \( "n" \) millieurcs.

The behaviour of radon before it reaches its transient equilibrium is also of considerable importance. Equation (15) and (16) give expressions for the \( \text{Ra B} \) and \( \text{Ra C} \) contents of a system starting with \( N_x \) atoms of \( \text{Ra} \). Each of these expressions is exponential, passes through a maximum for a
certain value of \( t \), and then falls off with a half life similar to that of radon. The graphs in figure (1) are based on these equations, and express:

1. The growth of Ra B from Rn, expressing the amount present at any interval as a percentage of the maximum value.

2. The growth of Ra C from radon, similarly expressed.

3. The growth of \( \gamma \) ray activity in a radon tube, again expressed as a percentage of the maximum. In this case, the growth is that measured with an ionization chamber employing 1 cm. of lead filtration.

Since the \( \gamma \) radiation is mainly due to Ra C the third curve closely follows the second. The gamma ray activity may be accurately expressed as

\[
S = K (B) + (1-K) (C)
\]

where \( S \) = observed percentage of maximum \( \gamma \) ray activity,

\( B \) = percentage of maximum of Ra B accumulated,

\( C \) = percentage of maximum of Ra C accumulated.

\( K \) is an experimentally determined co-efficient, depending on the depth of filtration employed in measurement. Values of \( K \) have been determined by SIAMER, the value corresponding to one centimetre of lead being 0.040.

The radiation from Ra B is softer (i.e., less penetrating) than that from Ra C, so that the relative contribution of Ra B falls as the depth of filtration is increased.

If the time elapsing since sealing a radon container is accurately known, curve (3) above permits the calculation of its maximum \( \gamma \) ray activity from measurements made before it reaches its equilibrium. Thus it is possible to measure a radon sample at any time after sealing.

It will be noticed that the curves in figure (1) rise very steeply for small values of \( t \). This is important in the operation of radon plants, for the exposure to radiation can be considerably lowered by reducing the time of
operation. This reduction in time of operation from 60 to 30 minutes lowers the radiation exposure to $\frac{1}{5}$ of its value for 60 minutes. In general, it is advantageous to handle the radon, where necessary, when it is just freshly isolated from its decay products, for it then has a very low $\gamma$ activity.
ISOLATION OF THE RADON.

Turning now to the more chemical aspects of the problem, we find it somewhat complicated by the strong tendency of radon to become adsorbed on solids. Most dry radium salts exhibit this property strongly, so that it is impossible to obtain a good yield of radon merely by pumping the gas off dry radium chloride. The emanating power, or ease of removal of radon from radium salts, may be increased by heating the salt, by adsorption of the radium on a suitable carrier precipitate, or by solution:—

1. Fusion of radium compounds.

Kolowrat (6) has shown that in general, the amount of radon liberated from radium compounds depends not only on the temperature to which they are subjected, but also on the time of heating.

Some 3 or 4 hours at about 1000° C may be required for a good yield. This does not appear to be a very satisfactory method.

2. Adsorption on carrier precipitates.

In recent years, certain preparations have been investigated which display emanating powers of the order of 100%. (7) Thus if radium ions are adsorbed on an aged precipitate of Fe(OH)₃ in the presence of sulphate ions to prevent re-solution, the preparation, when dried, will give off some 95% of its theoretical yield of radon. Moreover, this result appears to be permanent.

Purification of the resultant radon is in this case, quite simple. The only impurities are vapours from the iron oxide, and possibly organic vapours from the drying agent used in the preparation. Werner (8) has developed an apparatus, yielding extremely high purity of radon, utilising, inter alia, calcium filings for adsorption of the gaseous impurities given off by the iron oxide. However, it appears that the radon emanating from such preparations can be adsorbed immediately in charcoal filled capillary in
sufficient concentration for therapeutic use. Such a method is used in
the "radon water," a device marketed by a German manufacturing organisation.

In view of this simplicity of purification, and the complete absence
of water in the resulting radon (this point will be discussed in the next
section), such a preparation was seriously considered. Unfortunately, the
quantity and uncertainty of the impurities in the available radium debar
the project. The preparation of the starting material would almost certainly
be upset by the impurities, and in any case the extensive handling of the
radium involves considerable risk.

(3) Aqueous solutions of radium.

The last stated method for obtaining high yields of radon depends on
its fairly low solubility in water. With the radium and its impurities
completely in solution, no solids are present on which the radon can be ad-
sorbed, save for the glass of the container. This adsorption on glass is
small but may be appreciable in time. As radon accumulates in aqueous
solution, it remains only partially dissolved in the water, and may be com-
pletely removed by pumping.

This method has the disadvantage of forming considerable impurities,
but appears to be the only one applicable in this case.

Impurities in aqueous solution of radon.

The word "impurities" is rather misleading, since the ratio of
impurities to radon may well be of the order of 500,000 to 1. A dis-
cussion of the various impurities, together with possible methods of removal,
follows:

Hydrogen and oxygen:

These are the major impurities, and are formed by decomposition of
water under the influence of α, β, and γ radiation. Duane and Scheuer
(12)
have shown that the effect is mainly due to alpha radiation, and is much more pronounced in the liquid phase of the system. The rate of formation of these gases then depends \(^{(13)}\) firstly, on the fraction of the radiation adsorbed by the water, and hence on the dimensions of the containing vessel; and secondly, on the relative volumes of solution and gas above it, for this determines the radon concentration in the liquid, and hence the alpha ray activity in it. The amount of radium present has of course a direct bearing on the rate, and this appears to be the only factor for which figures are available. Thus Gray and Ramsay \(^{(14)}\) state that the weekly yield of electrolytic gas from 21 gms. of radium in solution was 25 cc, at S.T.P. In the absence of determinations taking all the above factors into account, the above figures have been used as an approximate basis for calculations. The gases are not evolved in their stoichiometric ratio; an excess of hydrogen being present. This has been attributed to the formation of hydrogen peroxide \(^{(15)}\) under radiochemical influence, and certainly some \(\text{H}_2\text{O}_2\) appears in the solution. Livingston \(^{(16)}\) objects to this theory, however, since the excess of hydrogen is undiminished after years of use of such a solution. However this may be; the excess hydrogen presents a problem, for the gases cannot be completely eliminated by recombination.

Various methods of removal of \(\text{H}_2\) and \(\text{O}_2\) have been used, recombination as a preliminary step being common practice. Many types of apparatus \(^{14, 16-22}\) employ a spark for recombination purposes; hot metallic calcium \(^{(5)}\), or a hot platinum wire \(^{20, 23, 24}\) effect a less explosive action. Excess hydrogen may be removed by oxidation with hot copper oxide. \(^{12, 23, 24}\) This latter, in conjunction with copper itself \(^{22, 24}\) is a very convenient purifier, removing hydrogen and any hydrocarbons by oxidation to \(\text{CO}_2\) and \(\text{H}_2\text{O}\); and oxygen by reduction to \(\text{CuO}\). More usually however, copper oxide is used in conjunction
with platinum, either plated onto it \(^{(20)}\), or merely surrounding it. \(^{23, 24}\)

A further method for removing excess hydrogen utilises its high capacity for adsorption in hot palladium \(^{(20)}\).

Physical methods for the removal of hydrogen and oxygen will be discussed in detail later.

Carbon dioxide:

Alpha radiation has a very marked photochemical action on organic matter such as stopcock grease. In the presence of oxygen, the major decomposition product is carbon dioxide, and its removal is a matter of no little difficulty. Figures do not appear to have been published regarding the rate of this reaction, but it must be exceedingly fast, since appreciable amounts of \(\text{CO}_2\) are formed during only momentary exposures of greased stopcocks to radon. It will be appreciated that minute amounts of impurities are of importance in the present work, since the amount of radon concerned is so small. An average yield of radon might be 100 milli-curies; this represents only 0.066 cubic millimetres of gas at S.T.P. In an apparatus of two litres volume, this would imply a pressure of only \(2.5 \times 10^{-5}\) mm. of mercury.

\(^{(18)}\)

Rutherford has studied the removal of carbon dioxide, and finds that 24 hours treatment with potassium hydroxide is necessary for complete removal. Fractional condensation of the mixture of radon and \(\text{CO}_2\) was also tried, but the two gases have very similar volatilities. Although \(\text{CO}_2\) has the higher melting point, some \(\text{Rn}\) always condenses with it, and is released when the temperature is raised. Rutherford finally tried substituting phosphoric anhydride for tap grease, but with only partial success. This method is too messy for routine use, and does not permit of very good vacua.

\(^{(26)}\)

Wartenstein has investigated the subject further, and states that potassium hydroxide will rapidly (10 to 20 minutes) lower the pressure of
CO₂ in a glass-walled system to a fraction of a bar; but thereafter the action is slow, depending on the rate of removal of this strongly adsorbed gas from the glass walls of the system. Later work (11) has shown that by fusing the KOH, the reaction may be accelerated sufficiently for routine use, but the problem is not very satisfactorily solved, and the aim has been to avoid grease as far as possible in radon plants.

Water vapour:

Some water vapour accompanies the gases pumped off radium solution, and if a recombination process is used, further water is formed at this stage. This water is usually removed with phosphoric anhydride, but a very convenient method has been introduced by Allen and Franklin (27) who employed a trap cooled in a mixture of dry ice and acetone. It was found that at the temperature so obtained, approximately -70°C, no condensation of radon was observed, and very effective removal of water vapour resulted.

It might be argued that since the pressure of water vapour will not exceed some 20 mm. of mercury, its presence in the final product is no serious inconvenience, since the radon and impurities still remaining will be compressed to nearly atmospheric pressure. However, this vapour will be decomposed into N₂ and O₂ by radon, and if any condensed drops of water are present in the radon tube these will in time also be decomposed, building up a pressure in the system that may lead to breakage. If this condensed water were eliminated from the final container, the above effect would be negligible, since the increase in pressure on decomposition of water vapour would be only 50% of the original water vapour pressure.

Hydrogen chloride:

Traces of HCl vapour from the radium solution accompany the radon as it is pumped off. A little HCl is usually added to the radium solution, to aid in solution of the radium. Such vapour may be removed with potassium
hydroxide.  

Livingston has suggested that some chlorine is formed in the radium flask by radiochemical decomposition of this hydrogen chloride. Very little should result, and very little mention of this effect appears to have been made in available literature. Beyond a possible reaction with mercury used in the purification system, the chlorine would be of no significance, provided it is present in small enough concentration.

Organic vapours:

Traces of organic vapours may be present from top greases etc., and may be removed by oxidation with hot copper oxide, or by condensation in a dry ice trap.

Helium:

Helium will be formed to an insignificant extent during radioactive decay. It may be removed by physical methods, being still gaseous at the temperature of liquid air when radon will condense.

Considering the impurities as a whole, the methods of removal may be roughly classified into chemical and physical. Chemical methods require fairly lengthy exposure of the gases to such reagents as a hot copper plated platinum wire, potassium hydroxide, and phosphorous pentoxide.

Physical methods of fractional condensation are much quicker, but are subject, here, to certain limitations. Water vapour and organic vapours may be frozen out with a dry ice trap with no condensation of radon. Radon has a boiling point of -64°C at atmospheric pressure, but its condensation temperature for the minute partial pressures existing in the purification apparatus is much lower. Rutherford has measured this temperature to be of the order of -150°C, and has also given tables of its vapour pressure at various temperatures down to -127°C. Figures below this temperature are
not available, but it appears that even at \(-133^\circ C\), obtainable with a liquid air bath, a small but appreciable vapour pressure still persists. However, this temperature is not low enough to condense hydrogen, oxygen, or helium, so that these gases may be pumped out of the system while radon remains condensed in a liquid air trap. This trap should be as small as possible, and time of pumping kept to a minimum, so as to reduce the loss of radon due to its residual vapour pressure.

Carbon dioxide cannot be removed by physical methods, as explained above, since it condenses with the radon in the liquid air trap. Treatment with potassium hydroxide is necessary for this gas.

**Types of purification apparatus.**

It will not be possible to discuss all the purification apparatus encountered in the literature in view of their number and variety. The following may be considered representative of the various systems:

The Dave Apparatus

A diagram of the essential features of this apparatus is shown in figure (2). It employs a chemical purification system, consisting of a copper oxide wire gauze (10) electrically heated with an inner platinum wire. A P.O. tube (11), and a KOH tube (9) are also employed. Radon and its impurities accumulate in the radium flask (1), and are pumped into this purification unit when required with the teopler pump (4). This type of pump is standard in most plants, since the mercury used in its operation has a sufficiently low vapour pressure, and does not appear to interfere with the purification. The mechanism is as follows:-- The whole system is first evacuated as far as the teopler pump, when the mercury level in (4) is lowered to below the level of the by pass. Gases in the radium flask now share their volume with that of the whole system. The mercury is now raised again, so that the gases in
flask (4) become isolated from the radium solution and are forced right up through the 3 capillary (8) into the purification unit. Mercury is raised above this capillary and is then lowered again, such raising and lowering being controlled by varying the pressure in reservoir (7). As the mercury falls, a certain amount remains trapped in the capillary (8) and acts as a valve, preventing back diffusion of the gases pumped above this valve.

The process may be repeated at will, depending on the efficiency of extraction required. After sufficient time has been allowed for purification, the gases are transferred into a glass capillary above stopcock "B" with a second transfer pump (13) and reservoir (16).

The process requires some 30 minutes manipulation of stopcocks and switches in close proximity to the radon. Because of this, Mailla (24) has rendered it semi-automatic by the use of relay circuits, operated by the mercury. The plant is fairly complex however, affording ample opportunities for accident, and the presence of a greased stopcock after removal of the carbon dioxide is not very commendable.

The use of the weighty and time consuming stopcooler pumps has been eliminated by Allen (31), and Allen and Franklin (27). The latter employ a dry ice trap to freeze out water vapour in a side arm of the purification unit. When connection is made between this unit and the radium flask, the rush of vapour so induced carries over most of the radon and impurities with it. A specially designed mercury vapour pump transfers the purified gas from the purification unit into a large flask, from which it is forced into capillary tubing by displacement with mercury. Here again, the system is complex and liable to meet with accidents.

A greatly simplified design has been introduced by Henderson (32), who have dispensed with a second transfer pump in a "straight through" type of apparatus as in figure (3). The purification unit is so designed that mercury can be raised right through it, displacing the gases into the
Schematic Diagram of Apparatus. (Not to Scale.)

FIGURE 4
capillary tube above it.

While it is possible to fuse potassium hydroxide onto the glass walls of the purification tube, this does not hold for P₂O₅, and removal of water is therefore not complete in this apparatus. However, the authors claim that this is not necessary, and that no drops of water have ever been apparent on the mercury surface. Certainly the KOH will remove a little water.

A physical method of purification has been described by Oddie (11), whose apparatus seems to be a considerable advance on those discussed above. This apparatus is shown in figure (4). Two transfer pumps of large capacity are used to transfer the gases from the radium solution first to the water condensation trap C, and thence to the flask F and liquid air trap G. With the mercury level in flask F raised nearly to the base of the liquid air trap, the gases are left for 10 minutes for complete condensation of the radon, when residual gases are pumped out through the side arm H, and the liquid air is removed. With a trap of such design, it is now possible to displace all the radon into the tube K, which is coated with fused KOH, and thence into the capillary I, by raising the mercury through flask F.

This plant gives a very satisfactory performance. Yields of the order of 30% are obtained, at concentrations of 2 - 4% radon in 30 minutes operation. The impurities remaining have been claimed to be largely O₂ (33), but a considerable amount of H₂ and O₂ appears to be occluded with the condensed radon since concentrations of 1% radon are obtained on a second condensation of the radon in C.

The method is more convenient than others considered, in that the apparatus requires no renewal of chemicals or fittings in awkward places. The KOH in tube K is easily replaceable, since the whole tube may be removed at the ground joint H. Moreover, there is no possibility of clogging capillaries etc.
which is fairly common with some chemical units, e.g., those using hot copper oxide.

The above considerations have led to the development of a new apparatus, using Oddie's method of purification, but with certain modifications in the design, aiming primarily at the elimination of all grease from the path of the radon.
THE PURIFICATION APPARATUS.

In Oddie's plant, the purification is effected in two distinct steps, requiring the use of two rather bulky topler pumps. However the dry ice freezing unit is very efficient, and the gases are passed only once through it. It seems possible then, to redesign the plant on a "straight through" type of system, so that gases are merely run through the dry ice trap on their way to the liquid oxygen stage. Such a principle is employed in the present apparatus, shown in figure (5). Oddie's first topler pump has been eliminated, and a cut off system incorporated between the radium solution and purification unit.

On opening this cut off system, with the apparatus evacuated, gases from the radium solution in flask (1) diffuse into the dry ice trap (14); and thence into the topler pump (17) and liquid oxygen trap (20). Two strokes of the pump affect fairly complete transfer of the gases into the liquid oxygen stage. The pumping action is only partly due to the topler pump, however. As water vapour diffuses into the dry ice trap and condenses therein, it must also carry over other gases with it, in the same way as utilized in the pumping system of Allen and Franklin's apparatus. In their apparatus the permanent gases (H₂ and O₂) are slowly removed by combination as they diffuse beyond the dry ice trap, thus lowering their tendency to diffuse back into the radium flask. Since no such effect operates here, the back diffusion tendency has been prevented by making the system beyond the dry ice trap of large volume.

Two entrances to the liquid air trap have been provided. The first enables the topler pump to transfer its contents into the trap, but is necessarily rather constricted and circuitous. A more direct route, via the by pass (24), is opened during the short period when the cut off system is first opened. This permits of quick and efficient diffusion of the radon.
into its condensation stage, further contributing to a good yield of this gas.

By a rather extensive system of mercury sealing, all greased stopcocks have been protected from exposure to the radon; and in fact the radon encounters no grease at all in the whole system, except that which the mercury raises with it. Even this is largely trapped by the splashheads 35, 36, and 37.

A description of the routine procedure for extraction will make the above points clear:

Position after previous extraction:

The cut off system of magnetic vacuum valves (6) and (10) will have been closed during the previous extraction, so that the pressure in the purification system may be altered without affecting the radon accumulating in flask (1), and other apparatus beyond the cut off system. Mercury, contained in the steel reservoir (38), and auxiliary adjustable reservoir (41), will be at the levels indicated in the diagram. The apparatus is usually left evacuated as an additional precaution, to relieve the pressure on valve (10).

Preparation for extraction:

A gold capillary tube of suitable length must be fitted onto the KOH tube 22, and the latter filled with KOH. To this end, taps (26) and (39) are first opened. Gases now diffuse through the mercury reservoir (38) via the slow leak (40), and thence into the purification system until it reaches atmospheric pressure. A suitably prepared KOH tube with gold capillary attached is now substituted for the sealed off socket which takes its place when the plant is not in use. Preparing the KOH tube consists in introducing a few pellets of KOH into the section immediately opposite the figure (22) in the diagram. To ensure quick action, this KOH is melted onto the glass. The ground joint (21) is greased, but with a layer of grease in-
sufficient to reach the top of the joint, which is then sealed externally with soft wax for additional leakage protection.

Tap (39) is now closed, and the high vacuum pump switched on. Satisfactory vacuum has been obtained without the use of diffusion pumps, a mechanical rotary pump has been sufficient. Pressures of $10^{-3}$ mm. of mercury are generally obtainable, and occasionally as low as $10^{-4}$ mm., with new oil in the pump. The dry ice traps (14) and (29) are surrounded with dewar flasks of acetone - dry ice mixtures, the liquid air trap is filled, and tap (26) is closed. This tap is now sealed with mercury by opening stopcock (30), until the mercury rises to the required level above tap (26). Having adjusted the slow leak (40) to a pre-determined setting, the plant is now ready for extraction to proceed.

Procedure of extraction:

A detailed discussion of the cut off system has been reserved for the next section, for the present it will suffice to state that extraction begins by opening this system for some 10 seconds (T₁), after which magnets (6) and (10) are switched off again. The by pass tube (25) is closed by filling it with mercury with the aid of tap (31). Float valve (24) prevents the mercury from rising too far up the left hand tubing. As explained above, the only entrance to the liquid air trap is now through the toeppler pump.

This pump is now put in operation. The high vacuum pump having been left running, tap (39) is opened and the pump switched off some five seconds later. This lowers the pressure in reservoir (38) to such a level that tap (33) may be turned fully on, mercury now rising up the toeppler flask (17) at a rate depending on the velocity of diffusion of gas into the reservoir through the slow leak.

After opening tap 33, the toeppler pump is operated by remote control switching of the high vacuum pump, so that radiation exposure is reduced.
As the mercury rises, it closes float valve 16, which restricts its path to the tospler pump itself. When the mercury level reaches the point "X", the vacuum pump is switched on. This pump is of very high capacity and quickly lowers the mercury again, even against slow leak (40) which is continuously operating. The exact time at which to switch on the pump is so judged that mercury will fill the capillary 18 before it breaks at "X" and begins to fall again.

The pump is left on until the mercury falls just below the by pass (42), when it is switched off. In view of the large volume of mercury which must pass through tap (33), a state of equilibrium is never reached as regards pressure in flask (17) and the reservoir; so that mercury continues to fall for some seconds after switching off the pump. Moreover, the slow leak prevents a quick rise of pressure in the reservoir, so that some time elapses before the mercury level rises again. During the interval, the cut off system is opened for a further five seconds (T2), so that a large proportion of the remaining gases diffuse into the tospler pump, and are bubbled through the mercury in capillary (18) at the top of the second stroke of the pump. The cycle may be repeated if required, but two strokes of the pump are generally adequate. As the mercury reaches the capillary for the second time, it is allowed to continue filling into flask 19 until it reaches the neck of this flask below the liquid air trap. Tap (33) is now closed, and some ten minutes are allowed for condensation of the radon, during which time the operator retires to a discreet distance.

The mercury, sealing tap (26), is now lowered by pumping out reservoir (38), and opening tap 30 until the mercury falls to the desired level. Tap 39 is closed and the high vacuum pump left running. When it has evacuated the tubing leading to tap 26, the liquid air trap is refilled
with liquid to the brim, and tap 26 opened for twenty seconds. (p. 7). Having pumped out the permanent gases, tap 26 is closed and once more sealed with mercury. The liquid air is now removed from its container with a stream of warm air. This is provided with a water jet pump and heated copper spiral, not shown in the figure. Four minutes are allowed for complete removal of the liquid, and evaporation of the radon, when the mercury is raised right up to the tip of the gold capillary. Reservoir (38) is not of sufficient height to raise the mercury this far, so that the adjustable reservoir (41) is used in conjunction with tap (32) for the final operation. If required, the mercury may be held at a level just above the ground joint for some minutes, allowing time for further removal of carbon dioxide by the potassium hydroxide. For routine extractions this is not necessary, however. Finally, the gold capillary and its contents are cut off just above the base with a pair of blunted side cutters. The gold is of such fine internal bore, that it is autogenously welded as the side cutters pinch it off, and a perfectly leak-proof seal results.

Whenever possible, all handling of the gold capillary is carried out at this stage, since the enclosed radon has, as yet, only a low X activity. When time permits, the plant is returned to its initial position as follows:—

Return to initial position:

With the vacuum pump running, and tap (39) open, the mercury is run down through tap (33) until about one third of the teopler pump is emptied. Since there is no gas in the liquid air section as the mercury falls, it breaks at the point "x", leaving flask (19) half full. The mercury is lowered below tap (26) again, and air is admitted to the apparatus through this tap, having switched off the vacuum pump and opened tap (39). This drives the mercury out of flask (19) into the space at the top of the teopler pump left
for it. At this stage, it is well to pump out any radon that may have got
entrapped in the KOH tube. This is easily effected by evacuating the
system, filling it with air and evacuating a few times, to wash out any
absorbed gas.

The outlet from the high vacuum pump leads to the intake of a very
efficient ventilation system, so that the issuing gases will be withdrawn
from the laboratory. This ventilation system has its main intake directly
under the plant, radon being a heavy gas, and will have been in operation
throughout the extraction.

The KOH tube may now be removed, when a sealed off socket is sub-
stituted for it. The apparatus is once more evacuated, tap (26) is closed,
and by the use of taps (36) and (33) the mercury is lowered below float
valves (16) and (24) to its initial position. The dewar flask is removed
from trap (14), and the condensed water is distilled into trap (29) by
opening tap (26) with the vacuum pump running. Gentle warming of the
apparatus, especially trap (14), effectively removes all water vapour.

Tap (26) is closed, the pump switched off, and air admitted to the high
vacuum leads by means of tap (39). The dewar flask is removed from trap
(29), and its base section removed at the ground joint. Condensed water is
then removed from this trap, the latter cleaned and replaced, and the apparatus
is once more in its initial position.

A few remarks on the general running of the plant will be made at
this stage:

Measurement of pressures:

Throughout the present work, a high frequency spark discharge has
been used as a vacuum gauge. The colour and intensity of the glow produced
in the evacuated section affords an approximate measure of the pressure.
Although not very accurate, the method is exceedingly quick, and is highly
suitable for measuring the pressure of residual gases in the liquid air trap, as the hydrogen and oxygen are pumped off. Experience has shown that the pressure generally falls to $10^{-3}$ mm of mercury after about twenty seconds pumping, but a quick method of testing this is almost essential at this stage. The instrument has been calibrated against a Macleod gauge.

The same instrument has been invaluable as a leak tester, minute "pinhole" leaks being of common occurrence with the pyrex glass used. Routine practice has been to test the whole system in this way before extraction, with the exception of all parts in which radon is accumulating. It is only a matter of a few seconds.

**Filling and removal of mercury:**

Mercury is filled into the apparatus in the following manner:—

With all taps except (26) turned off, the apparatus is evacuated. Reservoir (41) is now filled with mercury, and this is run into the trap pump with the use of tap (34). Closing the slow leak (40), reservoir (38) is now evacuated, and a little mercury run in through tap (33), filling the connecting tubing. The connecting tubing on the other end of the reservoir is similarly filled, using taps (30), (31), and (32). With repeated fillings of reservoir (41) the whole system is now filled with mercury, thus ensuring that the correct amount is run in, and the mercury is then filled into reservoir (38) by use of the appropriate taps. Addition of a little more mercury to reservoir (41) completes the process.

Mercury may be emptied from the system by running it all into reservoir (38), and closing the rubber connecting tubing with clamps. This tubing is then cut off below taps (33) and (31) and the mercury run out by gradually opening the clamps.
Operation of Toepler pump.

The most time consuming part of the extraction is the operation of the Toepler pump, and for this reason a relay circuit was considered, to render the process automatic. However, the exact point at which the vacuum pump should be switched on at the top of the Toepler pump's stroke, depends on the pressure of gas in flask (19), and this varies with each extraction. Moreover, any relay circuit is liable to break down, and sufficient protection against radiation at this stage can be obtained by remote control switching. At a distance of only six feet from the pump, the gamma ray intensity is quite low at this stage, since the radon is only just isolated from its decay products.

Details of design and construction:

The design of the plant will now be considered in some detail, and constructional aspects pointed out where necessary. Factors relating to the volumes of each section will also be discussed.

The radium flask:

The volume required for this flask depends on several factors, the most important being the volume of water necessary to completely dissolve the radium salt and its impurities. This is very uncertain, because of the large quantity of impurities of questionable nature. It will be appreciated that an experimental determination of this volume is not possible until the whole apparatus has been prepared to receive the radium and its accompanying gases. An approximate figure for this minimum volume has been given as 150 cc. by the operator of the plant previously run in Wellington, but this, again, is very uncertain.

Other factors bearing on the volume of the flask are as follows:

(1) Since a certain amount of the water solvent is removed at each extraction as water vapour, hydrogen, and oxygen, the volume originally adopted for the
solution must be well above the minimum required for complete solubility.

Even so, additions of water may have to be made at intervals.

(2) To ensure a good yield of radon, the volume of gas in equilibrium with the solution must be sufficiently large. Kofler (5) has shown that the solubility of radon obeys Henry's law, and has expressed this solubility on a volume basis with the following equation:

$$\alpha = \frac{V_G R}{V_S}$$

where

- $V_G$ = volume of gas in equilibrium with the solution
- $R$ = amount of radon in the solution
- $V_S$ = volume of solution
- $E_G$ = amount of radon in the gaseous phase
- $\alpha$ = solubility coefficient.

Kofler has tabulated values of $\alpha$ at different temperatures. $\alpha$ will vary also, with the total salt concentration in the solution, being lowered as more salt is added. In the present case it has been impossible to determine the total amount of salt so that this factor has been neglected; and the value $\alpha = 0.26$, corresponding to 20°C has been adopted. With equal volumes of gas and solution, the radon would be 26% dissolved in the solution, while with a gas to solution ratio of 2:1, only 13% of the radon remains dissolved.

As the gas above the solution is removed during extraction of the radon, some radon will come out of solution in order to maintain the above equilibrium ratio. This effect will be accelerated by a large area of solution surface, but Robertson (33) and others have found that it is too slow to be of significance.

(3) Because of the necessity for screening those sections where radon accumulates, it will be advantageous to keep most of the accumulating gas in the radium flask itself, rather than in the connecting tubing and other sections up to the cut off system. The radium flask may be heavily screened with
lead, but such screening of glass tubing is neither easy nor convenient, as periodical inspections of such tubing will be necessary.

(4) All the above points favour a large volume for the radium flask, but the limiting factor lies in the pumping system. For efficient pumping, the volume of the teesler pump should be at least greater than that of the gas in the radium flask. Repeated strokes of the pump will ensure a good yield, but this is time consuming, while volumes of mercury in excess of one or two litres are dangerously heavy for accommodation in glass apparatus.

Taking the above factors into consideration, it was decided to allow for some 250 cc. of solution in one flask of 500 cc. capacity, having others in reserve to be substituted if necessary, when the radium solution was made up. The volume of connecting tubing etc. amounts to 170 cc., so that the gas to liquid ratio, based on above figures, is 420 : 250, resulting in a loss of only 15% of the radon due to solubility. Since 420 cc. of gas are to be pumped off at each extraction, a volume of one litre for the teesler pump is reasonable, especially as flask (19) contributes to the pumping action on the first stroke of the pump.

In point of fact, it was found possible to dissolve the available radium in 200 cc. of water, with the addition of about one tenth of a cc. of concentrated hydrochloric acid. This was poured into the above flask as explained below, and together with washing water, brought the total volume of solution up to 240 cc. The total radium was measured as 344 mgms. before solution, and the total small amount remaining undissolved amounted to 5 mgms. This was probably absorbed on insoluble impurities, the latter being filtered off before pouring in the radium solution.

Regarding constructional details, figure (6) presents a sketch of the radium flask and safe, drawn to one fifth natural size. The flask "A" is
seated inside a thick walled glass beaker "B" to retain the solution in
case of leaks or breakage. A loosely fitting tin lid, not shown in the
figure, is placed over this beaker for similar purposes. The beaker
stands on copper tubing spiral "E", intended as a cooling coil. So far,
this coil has not been used, but was intended to cool the radium solution,
when cold water would be run through it, in this way permitting the distil-
lution of water into the radium solution from more accessible positions.

Ample protection is provided by a lead cylinder "C", two inches in
thickness, and the whole is enclosed in a fire proof safe, "F". Access
to the radium flask is available by removing two blocks cut out of the
cylinder. These blocks have been so cut that at least two inches of
lead protection is maintained in all directions radiating from the bounds
of the solution, as will be seen from the cross sectional diagram at "A",
"B" and "J" are sections of the blocks. The lid "D" of the cylinder has
been cut similarly, along the line "G" in the figure, to render it removable,
again with no loss in protection. However, an unfiltered cone of radiation
gasses through the hole in the lid. It has not proved necessary to screen
this, as its intensity on the floor above is sufficiently low at all times.

A photograph of this section of the apparatus appears in figure (7)
opposite this page. The above description renders it self explanatory.

The connecting tubing is of 8 mm. internal diameter, and this width
has been maintained throughout the apparatus to ensure rapid diffusion of
gases. In some cases, shortage of glass has necessitated the use of 7 mm.
tubing, however. The platinum contacts (2) in figure (5) were provided
in case a small radium flask proved possible. In this case, the hydrogen
and oxygen would build up to a pressure greater than 6 em., which has been
shown to be (22) the minimum required for recombination by sparking, so
that a partial purification by sparking would be possible prior to extraction. Moreover, the water formed would be returned to the radium solution. Since 240 cc. of solution were required in the above flask, the total pressure will not build up to sparking range in less than about 9 days, on the basis of Grey and Ramsay's figures. This is much less than the average interval between extractions so that the process has not proved feasible.

The splash-head (3), prevents any bubbles of solution coming over with the gas as it is pumped off. The connecting tubing has been extended upward to the point "D" to facilitate the filling operation. This consists in breaking off the tip of the tubing at "D" and pouring in the solution, or water, with the aid of a long funnel extending below the side arm level.

The tube "E", sealed into the ground cone (4), is a safety device for the removal of accumulating gas if the cut off system suffers an accident and the tubing to the right of it has to be sealed off. In this event, a tube and tap may be fitted on at the ground joint, and gases pumped out through the tap. The tip of tube "E" will be broken off prior to this.

At the point "A", the connecting tubing diverges into two channels, leading to either of two duplicate units. The remainder of the apparatus has been constructed in duplicate, so that in case of breakage, a second purification unit will be able to handle the radon. One side, only, of the duplicate system will be considered, for each is independent of the other.

The trap (5) is intended to prevent any mercury reaching the safe, if a breakage in the cut off system should send it flying in that direction. The capacity of this trap is sufficient to hold all the mercury used in the cut off system, and the tube ending at "F" can easily be broken at its tip, enabling removal of the mercury.
MAGNETIC VACUUM VALVE

FIGURE 8
The cut off system:

The essentials of a cut off system in this case are that it shall be greaseless, since it is in constant contact with radon; and shall be capable of being opened against pressure differences which may arise in either direction.

The only simple solution consists of a mercury seal, of the type used to close the by pass (25) in figure (5). This is not very convenient when opened against large pressure differences however, and a sudden increase of pressure on one side could send the mercury flying.

An alternative solution is provided with the use of two magnetically operated vacuum valves, one of which is shown in detail in figure (8). This has been drawn to natural size. The valve consists of a ground joint "A", the socket portion of which is sealed onto a plunger "B", so that the joint may be opened by raising this plunger. A soft iron core "C" is sealed inside the plunger, tightly packed with asbestos "D", to ensure rigidity. This core enables the plunger to be raised magnetically when the electromagnet "K" is switched on. The valve case "E" has been extended beyond the normal range of the plunger to preclude any possibility of breakage if the plunger should "overshoot". When the valve is closed and not in use, the joint is sealed with mercury "F" as shown, to render it vacuum tight. The mercury system is shown in full in figure (5), consisting of a reservoir (8), which may be opened to atmospheric pressure, or to low vacuum leads, through the two way tap (9). Stopcock (7) controls the exact height to which the mercury is raised or lowered. Normally the valve remains closed and sealed with mercury, but immediately prior to extraction of radon, the mercury is lowered, a water jet pump affording ample vacuum for the purpose.

It will be seen that the valve shown in detail in figure (8) can only
resist a pressure difference higher on the side "X". If a greater pressure builds up in the tube "Y", it will force the plunger up, allowing gas to bubble through the mercury. For this reason, two such valves are used, (6) and (10) in fig. (5). These resist pressures from either direction provided that the section between them remains evacuated. Since the whole system is always roughly evacuated when the valves are closed, this condition is fulfilled in normal use.

If it is desired to keep the valve opened for long periods, a second plunger "H" may be slid under the plunger "E" while it is raised. A small iron core "J" is inserted for this purpose. A small permanent magnet easily affects the required movement.

There is necessarily a large gap between the iron core and electromagnet K, owing to several thicknesses of glass and asbestos. This considerably reduces the efficiency of the magnet, and a solenoid affording the maximum pull on the plunger has been designed especially for this purpose by Mr. Neale of the Engineering Department at Canterbury College. The solenoid will carry 4 amps for a few seconds, exerting a tension on the plunger more than sufficient to raise it against atmospheric pressure. It is iron clad, as shown in figure (7), and employs 2400 turns of number 24 S.W.G. enamelled copper wire. The position of maximum pull on the plunger has been found experimentally to be about 1 cm. above this plunger, as shown.

A switching circuit for the magnet has been developed experimentally, and is shown in figure (9). The power source is 230 volts D.C. from the mains: alternating current, as normally supplied, leads to overshooting of the plunger, as the period of one fiftieth of a second is of the same order as the time required for the raising of the plunger. The current passing through the magnet "M" is controlled by a master switch "S", and a rotary selection switch, which varies the resistance in series with the magnet.
A range of resistances has been chosen so that when the arm "PH" of the switch is at its extreme position "L", 4.4 amps are passing. This is the maximum current permissible for a few seconds, and is intended to raise the plunger against excessive pressure or frictional resistance. After raising the plunger, .65 amps suffice to hold it well above its socket, and this current is obtained at position "3". This plunger is lowered until nearly touching its socket by switching onto position "D", and the remaining positions afford slightly lower currents, lowering the plunger gently onto its socket. All jarring of the plunger is thereby avoided. Normally, the plunger can be raised with .65 amps, so that the higher currents are not required except under exceptional conditions.

There proved to be considerable sparking as the selecting arm moved from one terminal to the next, attributable mainly to the inductive affect of the magnet. This has been reduced to a permissible degree with condensers, as indicated in the figure.

A pilot light "N" has been included which glows when any current passes through the magnet. This is a \(\frac{1}{4}\) watt neon lamp, drawing only a minute current, and operating at varying voltages. The voltage across the points "X" to "Y" varies from nearly 230 volts when the arm is at "3" to 110 volts when the arm is at "L". A series resistance of 100,000 ohms, brings this within the range of the lamp.

These valves have been developed from a type used by Allen and Franklin \(^{(27)}\). A diagram of their valve is inset in figure (8), and is an accurate copy of the only available data on the subject. This valve remains continually sealed with mercury, and is thereby much more easily operated than the above type. However, experience has shown that when such a valve is opened against a pressure difference of a few centimetres of mercury, the sudden rush of gas so induced is sufficient to drive pellets of mercury out
of the valve. This occurs in the brief interval when the plunger has been raised just above the joint, but with its base still dipping in mercury. Moreover, pellets of mercury tend to adhere to the ground joint, preventing it's closing properly when the plunger is lowered again.

Before proceeding to the remainder of the apparatus, some mention should be made of the type of glass employed. All sections of the apparatus in which radon is permanently accumulating, i.e. the radium flask etc., have been made of soft soda glass, with a wall thickness of one millimetre. It is stated that pyrex glass devitrifies more readily than soda under the excessive alpha ray bombardment to which these sections are subject.

Macdonald and Margolese (22) discuss the question of glass very fully, and are in accord with the above view, but consider pyrex permissible for the purification system since it will receive only occasional alpha radiation. They stress the importance of using reasonably thin walled glass, since the strain set up by radiation is of no small account, and will easily cause cracks in thick walled joints that have not been perfectly annealed.

In view of the liquid air trap, pyrex has been used for the remainder of the system, being also more easy to work with. The change over from soda to pyrex is made between the valves (6) and (10). Graded seals have not been available, so that a ground cone of soda glass fitting into a ground socket of pyrex has been used. The joint has been sealed externally with picein wax, which unavoidably exposes a minute area of organic matter to the radon.

The purification system:

This system begins with the water vapour trap, (14). This reaches a temperature of about \(-70^\circ C\), causing very efficient freezing of water. For this reason, a fairly wide passage for gases has been maintained, equivalent to a cross sectional area of 0.4 cm\(^2\). Narrower paths for the gases
SAFETY VALVE

FIGURE 10
may possibly improve the efficiency, but also lead to blockage of the trap with the ice formed. Such blockages are occasionally met with in Oddie's apparatus.

Float valve (15) was set up before the lower valves (16) and (24) had been included. At this stage it was essential, and it has been retained as an additional safety device to prevent any mercury reaching the safe. The consequences of such would be so disastrous that the possibility of the lower valves not functioning has been provided for. The valve has been designed to afford a wide passage for gases when open, and to withstand a sudden uprush of mercury without breaking. Figure (10) presents a natural sized diagram of the valve. The plunger "A" will just float when mercury has filled the wider flask to the point "B". If and when sufficient mercury has entered the valve to fill it to this height, further filling of the valve forces the plunger into its ground socket "C", but reasonably slowly, since the mercury has a wide flask to fill at this stage. The mercury enters the valve mainly through "D", in such a direction as to avoid a sudden upward impact on the plunger, and it may be drained out through the capillary tube "E". The cross sectional views show the method of maintaining a wide passage when the valve is open.

The asppler pump (17) has been made of one litre capacity as explained above. The by pass (42) is a standard device, enabling gases to enter the pump via this tube, rather than bubble through a large volume of mercury from the base of the pump. The flask has been supported in a metal cup, packed with asbestos, and rigidly supported, as some 40 lbs. of mercury are involved.

The function of the capillary (18) has been explained. Its height,
the distance $X - Y$, must be greater than the optimum pressure of gas that is to be retained in flask (19). If too high, however, the mercury jars the glass as it falls, and 20 cms. was found to be permissible from this point of view. Accordingly, flask (19) has been made of sufficient volume to accommodate the maximum anticipated yield of gas at a pressure of less than 20 cms. Referring again to Gray and Ramsay's figures, it appears that a volume of 250 cc. should accommodate approximately a fortnight's yield at a pressure of 19 cms., and this has been the volume chosen. A larger volume has advantages, but involves an unnecessary weight of mercury.

Several designs have been considered for the liquid air trap; the one chosen, (20) in figure (5) has several advantages. It is of a "straight through" design, so that mercury can be raised through it after removal of the liquid. The latter operation is easily effected in 4 minutes with a stream of warm air. Moreover, it has the inestimable advantage of a small volume, so that no very high vacuum is required for removal of permanent gases. This means that the time of evacuation may be kept down without seriously effecting the resultant concentration of radon.

The KOH tube has been made of the form shown in (A), figure (11). The KOH can be retained in the blown out section, lowering the possibility of its getting below the ground joint if warmed too much. The method of waxing on the gold capillary is also illustrated.

(C) in figure (11) is a form of collecting tube used for sealing the radon into small glass seeds. These latter have proved the most satisfactory containers for radon intended for radon ointment. They can be easily broken under vaseline.

For determinations of concentration of radon, collecting vessels of type (B) have been used.

The system as a whole has been kept at as low a level as possible
with respect to mercury reservoir. (38) This permits all operations, except the final compression of the gas into its capillary, to be effected from this reservoir, instead of having to adjust reservoir (41) at various stages.

The high vacuum leads:

All tubing leading from tap (26) to the high vacuum pump is of 14 mm. internal diameter, or more, to facilitate quick evacuation. In no place has this wide passage been reduced, even in float valve (28). Stopcock (26) is only of 5 mm. bore, but was the only size available. The large volume trap (27), and float valve (28), both situated above barometric height, are intended to prevent any mercury reaching the pump.

Trap (29) protects the pump against condensable vapours, and maintains the wide passage pointed out above. The lower section of this trap is removable at the ground joint, so that it may be readily cleaned and dried.

At certain stages, gases enter the pump through slow leak (40). This is a metal needle valve, and is backed by a calcium chloride tube, not shown in the figure.

General:

In general, the apparatus has been made as compact as is consistent with reasonable ease of access. To ensure rigidity of construction it has been mounted on a piping framework, so that the weight of mercury and lead screening is easily taken. The radium safe is separately mounted on a solid block, and is bolted to the remainder of the system. A tray is mounted below the main system to catch any mercury that may break loose.

The accompanying photographs, figures (12) and (13) will clarify the above points. Unfortunately it has not been possible to secure a single photograph of the whole system since the space is so limited.
Certain sections of the duplicate unit are visible. These do not coincide exactly in all cases since other designs have been tried, and in fact, as yet only one system has been completed to the above description.

The more prominent sections are labelled as in figure (5). Hot air to bubble off liquid air is heated in the copper spiral (S), and filtered through (T). When required, the glass tube (V) is dipped into the liquid air trap, a short length of rubber tubing enabling this operation.

Most sections beyond the cut off system are screened by lead. Thus only the tip of trap (5) can be seen.
PERFORMANCE OF THE PLANT.

The performance of the plant may be discussed conveniently under the following headings:

1. Mechanical performance of the system
2. Yield of radon obtained
3. Purity of resultant radon
4. Time of operation, and radiation received during extraction of the radon.

Mechanical Performance.

The operation of the Toepler pump has been discussed, and has proved quite satisfactory except in one respect. The exact moment at which to switch on the vacuum pump at the top of the stroke is critical and hard to judge. If left too long, a large volume of mercury runs over into the capillary valve, and causes considerable bubbling on the next stroke of the pump. Pellets of mercury have often hit the liquid air trap.

To obviate this difficulty, the design of this section of the apparatus has been altered to that shown below:

![Diagram](attachment:image.png)

Any bubbling on the second and third strokes of the pump is now confined to the nearly horizontal tube A B, and does not permit the mercury reaching the liquid air trap.

The cut off system has given no trouble at all in the two and a half
months operation of the system so far recorded. With the exception of one very cold morning, when a current of 1 amp. was required to lift the valve plunger instead of the usual .65 amp., no sign of this plungers sticking has been observed.

The remainder of the system requires no special comment, and has behaved quite satisfactorily. None of the various safety devices have been required so far, but it is not felt that this detracts from their value.
Method of measurement of the radon:

The radon has been measured by virtue of the ionization it produces in an ionization chamber designed for the measurement of gamma radiation. It is not proposed to discuss in detail this ionization chamber, nor the method of amplification and measurement of the ionization current set up therein. Readings and calculations have been carried out by the writer, but the design and method of operation of the measuring system are due to (34) Mr. G. E. Roth. In brief, the method consists in measuring the ionization current caused by a known quantity of radium placed in the chamber, followed by a similar determination with the radon sample under like conditions. All due precautions have been taken to ensure that the current so measured is strictly proportional to the intensity of gamma radiation set up in the chamber. These may be summarised as follows:

1. The voltage across the ionization chamber has been sufficiently high (generally 1700 volts) to ensure that a saturation current is measured, i.e. that all the ions formed which are capable of being collected on the collecting electrode, are so collected.

2. To avoid errors due to fluctuations in the response of the amplification system, the current produced by the radium standard has been measured at frequent intervals during each series of determinations.

3. Zero errors due to varying background effects and other causes have been corrected by suitable adjustment before each measurement.

4. Where large amounts of radon are measured, a smaller resistance has been substituted for the normal one through which the ionization current is caused to flow. This lowers the voltage across this resistance to a value that is within the range of the amplification system.
(5) To avoid errors in reading the milliammeter employed in measurement, a mean of a series of readings has been taken, the readings being altered by varying the resistance in series with the meter. Correction for such resistance has been made in calculating the mean reading.

One further point should be noted:--

The ionization chamber is in the form of cylinder, with the radiating source placed inside in a smaller concentric cylinder of lead, to eliminate α and β radiation. Since that radiation proceeding from the ends of a long source must pass through this lead cylinder at an angle, in order to reach certain sections of the ionization chamber, the average filtration due to the above lead cylinder is greater for a long source than for a short one.

Thus the observed activity of a source of 10 cms. active length is smaller than that due to the same amount of radon enclosed in a source of active length 3 cms. An experimental correction curve has been drawn up to make approximate correction for this small effect, which varies with the active length of the source.

Observing the above precautions and corrections, such determinations afford a measure of the γ-ray activity of a radon sample at the time of measurement. For calculations of yield however, the radon content at the time of removal from the purification apparatus is required. The following examples illustrate the calculations involved. As explained previously, the radon may be measured before or after it reaches transient equilibrium with its decay products.

Measurement after reaching equilibrium:--

A certain radon sample was measured at 5 P.M. on the day of its extraction, by comparison with a radium preparation of content 9.96 mgs. This standard radium preparation was enclosed in a brass container, reducing its activity by 5%, as experimentally determined. It is thus equivalent to 9.46 mgs. of radium,
and its active length has been taken as approximately 7 mm.

The radon sample was enclosed in a gold capillary tube of .3 mm. wall thickness, equivalent to a 3% reduction in γ activity (as previously determined) and 8.5 cm. long. With the use of the active length correction curve, it is seen that if this radon were enclosed in a source 7 mm. long, and situated in the same position in the chamber as the standard, it would display 2% greater activity.

The ratio of the ionization current set up by the radon sample to that set up by the standard was 2.63. Taking the above corrections into account, the content of the sample at the time of measurement was:

\[
\frac{102}{100} \times \frac{102}{100} \times 9.46 \times 2.63 = 26.2 \text{ mcs.}
\]

The sample was extracted at midday on the same day, so that 5 hours have elapsed in which it has decayed. The radon content of the sample at midday will be given by equation (11 A):

\[ N_r = N_x e^{-\lambda t} \]

or, expressing the radon in millicuries:

\[ N_t = N_0 e^{-\lambda t} \]

where \( N_t = 26.2 \text{ mcs.} \)

\( t = 5 \text{ hrs.} \)

\( N_0 = \text{millicurie content of the sample at midday.} \)

Evaluating the factor \((e^{-\lambda t})\), we have \( N_0 = 27.2 \text{ mcs.} \)

As a proof that the observed activity of such radon samples does follow the above equation, the graph in figure (14) has been drawn up from a series of readings on one sample on successive days. The radon content in millicuries, at each observation, has been plotted against the time of
such observation, dating from the first reading. To bring out the exponential nature of the decay, logarithmic graph paper has been employed, and the observed points lie very closely on a straight line, as equation (11 A) would indicate. Further, the slope of this line is such that the radon has decayed to half its initial value in 3.82 days, which is in agreement with the accepted half life period of radon, of 3.825 days.

This degree of agreement is probably fortuitous since the experimental error is approximately 2%, but it does afford formal proof that the gas investigated was radon.

Measurement prior to reaching equilibrium:

Calculations made on these measurements must be based on equations (15), and (16), and their resulting graphs in figure (1). Such equations have been derived on the assumption that initially, the system consists only of radon, with none of its decay products. This is not strictly true in practice, since some radon will diffuse into the containing vessel some time before the bulk of the yield is compressed into it. This small amount will form active deposit on the container before it is sealed off, so that at the precise moment at which the mercury is raised to the seal off point, the containing vessel already contains some active deposit, and invalidates the above assumption. However, the volume of the container is generally less than 0.1 cc., so that the amount of radon in this volume is negligible compared with that in the 80 cc. of liquid air trap etc., into which the radon vapourises after condensation. As the mercury is raised in the final stages, however, even this small volume becomes appreciable, but only for about 10 seconds, this being the time required to raise the mercury over its last few centimetres. Thus the above quoted graphs should enable a cal-
calculation of radon content of such a container with a time error of about 10 seconds, dating from the moment at which the mercury reached the seal off point.

In cases where the mercury is lowered again before sealing off the radon, this method is not applicable, and measurements must await equilibrium. This former method involves a delay of some five hours, and also leads to greater radiation exposure, so that it is not so convenient as the one just discussed. An example of such a measurement is given below:

A certain radon container was measured at 12 hrs., on the day of its extraction, and after making all due corrections, its gamma activity was found to be equivalent to that from 16.4 mgs. of radium.

This sample was sealed off from the extraction plant at 10^5 hrs., but the radon in it had been compressed to the point of sealing off at 10^51 hrs., i.e. 72 minutes before the above measurement. Applying graph (3) in figure (1) it is seen that in 72 minutes a radon sample has reached 62.6% of its maximum activity, when measured with an ionization chamber employing 1 cm. of lead filtration, as in the present case. Further, this maximum activity is reached in 260 minutes, and in this time the radon has decayed to 96.8% of its initial value, as deduced from equation (11 A).

Thus the radon content of the sample at the time of removal from the plant was

\[ 16.4 \times \frac{100}{62.6} \times \frac{100}{96.8} = 27.0 \text{ mcs.} \]

As explained previously, graph (3) in figure (1) has been drawn up theoretically, using Slater's value for K in equation (28). To prove the validity of this graph, and in particular to ensure that the above time factor is of negligible account, a series of measurements has been made on a radon sample very soon after its removal from the plant.
The results obtained have been graphed in figure (15). The observed gamma activity, measured as outlined above, has been expressed as a percentage $^{\mathrm{18}}$ of the maximum value, found by graphical interpolation of values observed before and after this maximum. Time, has been dated from the moment at which the radon was compressed to the seal off point. The continuous curve in figure (15) is the theoretical plot of $P$ against time, and is of course identical with graph (3) in figure (1). Observed points are indicated by crosses, and these lie very closely on the curve. In particular, agreement is very good for small values of time, when any error in scaling off point would be most noticeable.

The above method is thereby proved for the case considered. Further, it has been routine practice to check such measurements with observations made on the same sample after reaching equilibrium. In all cases, the agreement has been within the limits of experimental error, and such checks have been discontinued recently in view of this.

In general, more than one set of determinations has been made on each sample, to preclude any possibility of error, and it may be said with some justification that whatever the method of measurement employed, the result is correct to within approximately two per cent of the stated value, for quantities in excess of ten millicuries. Below this value, the measuring system is not generally so accurate, but most determinations for the present work have kept above this limit.

Calculation of theoretical yield.

The following example will illustrate such a calculation, which has been explained in previous sections:

In one extraction, pumping operations ceased at $11^\text{h}$ hrs. on $11/6/47$. In the previous extraction, pumping operations were concluded at $10^\text{h}$ hrs.
on 9/6/47, i.e. 2 days 1 hr. 8 mins. previously. The total radium in solution has been estimated at 339 mgs.

Applying equation (10), we have the theoretical yield $R$ expressed as follows:

$$ R = N \left(1 - e^{-\lambda t}\right) $$

where, in this case, $N = 339$ mgs.

and $t = 2$ days 1 hr. 8 mins.

From graphical evaluation of the exponential term, we have

$$ R = 339 \times 0.303 = 104 \text{ mgs}. $$

**Observed Yields.**

The observed yield in any case would be expected to depend on the precise method of operation of the plant. The following factors should be of importance:

1. The longer the period of opening the cut off system ($T_1$ and $T_2$ in the description given), the more complete should be the diffusion of radon into the liquid air trap. However, too long a period may cause blockage of the dry ice trap (14) with condensed ice, and will certainly remove an undesirable amount of water from the radium solution.

2. Subject to the above limitations on the time of opening the cut off system, a third stroke of the toepler pump may increase the yield slightly, at the cost of a little more time spent in operation.

3. If the period of evacuation of the liquid air trap is prolonged, it may improve the vacuum, and hence the resultant concentration of the radon, but it should also cause a loss in yield owing to the small residual vapour pressure of radon at the temperature of liquid air.

4. Some radon will decay during the period of extraction, and this effect will vary with the time spent on extraction. This effect can be easily calculated, but it is so small, (generally less than 12%) that it has
been neglected.

(5) Since a 100% yield is hardly to be expected, the yield in any one case will depend on the amount of radon remaining in the radium flask after the previous extraction.

An attempt has been made to investigate each of the above factors by varying each in turn while maintaining the others constant. However, the time of extraction has not always been a matter of choice, so that factor (5) unavoidably varies in each case.

The observed results are set out in table I, under several headings. The period of accumulation, in the second column, is the time interval between extractions, i.e. \( t \) in equation (10). The third column shows the number of strokes of the toepler pump employed in each case, and the period during which the valves were opened at each stroke is given in the next column. The observed and calculated yields follow, together with the former expressed as a percentage of the latter. The column headed "A" will be explained later.
<table>
<thead>
<tr>
<th>Extraction Period of Accumulation</th>
<th>No. of strokes of taepler pump</th>
<th>Period of opening cut-off system</th>
<th>Theoretical Yield (Ao)</th>
<th>Observed Yield (Yobs)</th>
<th>Percentage of theoretical yield obtained</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 day 16.5 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
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<td>6%</td>
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<td></td>
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<td>6%</td>
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<td>3 days 22.1 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>4 days 25.8 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>5 days 27.5 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>6 days 29.1 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
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<td>7 days 29.9 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>8 days 30.7 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>9 days 31.9 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>10 days 32.6 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
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<tr>
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<td>11 days 33.5 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>12 days 34.7 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>13 days 35.5 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
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</tr>
<tr>
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<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
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</tr>
<tr>
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<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>16 days 37.8 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>17 days 38.7 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>18 days 39.5 hrs,</td>
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<td>00 00 00 00 sec, 00 sec,</td>
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<td>6%</td>
</tr>
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<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
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<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>21 days 40.9 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>22 days 41.3 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>23 days 41.5 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>24 days 41.9 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>25 days 42.1 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>26 days 42.3 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>27 days 42.5 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
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<td>28 days 42.7 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>29 days 42.9 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
<tr>
<td></td>
<td>30 days 43.0 hrs,</td>
<td>2</td>
<td>00 00 00 00 sec, 00 sec,</td>
<td>00 00 00 00 sec,</td>
<td>6%</td>
</tr>
</tbody>
</table>
Discussion of Results.

In general, the observed yield varies from 70% to 120% of the value deduced from equation (10). The few exceptions to this are all due to anomalous circumstances such as accidental breakage, leaks etc., and can all be satisfactorily explained as in extractions 2, 7, 9, 12, and 22.

The wide variations in observed and theoretical yields in other cases are largely attributable to factor (5) above. The following mathematical treatment of this point, although obvious, has not been found in any available literature on the subject:

Consider again equation (5) on page (4)

\[ e^{\lambda_2 t} N_2 = \int \lambda_1 N_0 \cdot e^{(\lambda_2 - \lambda_1)t} \]

.........(5)

which integrates, generally, to

\[ e^{\lambda_2 t} N_2 = \frac{\lambda_1 N_0}{(\lambda_2 - \lambda_1)} e^{(\lambda_2 - \lambda_1)t} + K \]

.........(6)

This equation is quite general, and makes no assumption as to the initial state of the system, except that it commences with \( N_0 \) atoms of radium, isolated from its mother element, but not necessarily from its decay products.

If now, it is assumed that initially no radon is present, but radium only, then we have \( N_0 = 0 \) when \( t = 0 \), and the resultant equation for the theoretical yield, "\( N_T \)" of radon from \( N \) milligrams of radium in time "\( t \)"

takes the form:

\[ N_T = N (1 - e^t) \]

Now suppose that the system consists initially of \( N_0 \) atoms of radium, and \( Y \) of radon. That is, suppose \( Y \) atoms of radon remain unremoved from the radium flask at the end of a certain extraction.
For the following extraction, then, we have \( N_0 = Y \) when \( t = 0 \), so that

\[
K = Y - \left( \frac{\lambda_1 N_0}{\lambda_2 - \lambda_1} \right) 
\]

which reduces, on approximation to:

\[
N_2 = N_0 \left( \frac{\lambda_1}{\lambda_2} \right) \left( 1 - e^{-\lambda_1 t} \right) + Y e^{-\lambda_1 t} 
\]

Following the same reasoning as in the derivation of equation (10), the expression \( N_0 \left( \frac{\lambda_1}{\lambda_2} \right) \left( 1 - e^{-\lambda_1 t} \right) \) will be equivalent to

\[
N \left( 1 - e^{-\lambda_1 t} \right) \text{ millicuries} \text{ if } N_0 \text{ atoms of } Ra \text{ are equivalent to } N \text{ atoms of } Ra. \]

Let \( Y \) atoms of \( Ra \) be equivalent to \( X \) millicuries, then the true theoretical yield \( "Y" \), in millicuries, will be given by

\[
Y = N \left( 1 - e^{-\lambda_1 t} \right) + X e^{-\lambda_1 t} 
\]

\[
= N_0 + X e^{-\lambda_1 t} 
\]

where \( N_0 \) is the theoretical yield explained and expressed above.

Values of \( e^{-\lambda_1 t} \) are easily read from tabulated values of the function \( (1 - e^{-\lambda_1 t}) \), so that \( Y \) may be calculated if \( X \) is known. Assuming that all the radon leaving the radium flask in any extraction is finally collected in the container and measured, then \( "X" \) for any one extraction may be obtained from the difference between observed and calculated yields in the previous case.
Further, it is reasonable to suppose that an almost constant percentage of the theoretical yield ",N", is obtained at each extraction, so that the observed yield, "Nobs.", may be expressed as

\[ \text{Nobs.} = A \left( \frac{N}{\theta} + X e^{-\lambda t} \right) \]

where \( A \) should be constant.

Values of \( A \) have been calculated for several extractions, as shown in the following table II.
<table>
<thead>
<tr>
<th>Retraction</th>
<th>Period of Accumulation</th>
<th>pH</th>
<th>X</th>
<th>$\lambda_X$</th>
<th>P</th>
<th>$\lambda_{poe}$</th>
<th>A</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1 day 16.5 hrs.</td>
<td>89% min.</td>
<td>0</td>
<td>0</td>
<td>89% min.</td>
<td>58% min.</td>
<td>.85</td>
</tr>
<tr>
<td>2</td>
<td>2 days 22.2 hrs.</td>
<td>73% min.</td>
<td>30% min.</td>
<td>16% min.</td>
<td>357% min.</td>
<td>130% min.</td>
<td>.70 (c.)</td>
</tr>
<tr>
<td>3</td>
<td>1 day 5.0 hrs.</td>
<td>66% min.</td>
<td>07% min.</td>
<td>30% min.</td>
<td>164% min.</td>
<td>75% min.</td>
<td>.77</td>
</tr>
<tr>
<td>4</td>
<td>1 day 19.6 hrs.</td>
<td>29% min.</td>
<td>27% min.</td>
<td>77% min.</td>
<td>215% min.</td>
<td>05% min.</td>
<td>.74</td>
</tr>
<tr>
<td>5</td>
<td>1 day 3.1 hrs.</td>
<td>70% min.</td>
<td>10% min.</td>
<td>257% min.</td>
<td>386% min.</td>
<td>.76</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>1 day 10.6 hrs.</td>
<td>96% min.</td>
<td>57% min.</td>
<td>41% min.</td>
<td>357% min.</td>
<td>97% min.</td>
<td>.77</td>
</tr>
<tr>
<td>13</td>
<td>5 days 0.0 hrs.</td>
<td>200% min.</td>
<td>36% (c.)</td>
<td>35%</td>
<td>746% min.</td>
<td>395% min.</td>
<td>.77</td>
</tr>
<tr>
<td>14</td>
<td>1 day 22.0 hrs.</td>
<td>700% min.</td>
<td>63%</td>
<td>43%</td>
<td>143% min.</td>
<td>99% min.</td>
<td>.64</td>
</tr>
<tr>
<td>15</td>
<td>1 day 0.0 hrs.</td>
<td>56% min.</td>
<td>40% min.</td>
<td>36% min.</td>
<td>94% min.</td>
<td>63% min.</td>
<td>.67</td>
</tr>
<tr>
<td>16</td>
<td>1 day 3.9 hrs.</td>
<td>227% min.</td>
<td>26%</td>
<td>9%</td>
<td>294% min.</td>
<td>186% min.</td>
<td>.79</td>
</tr>
<tr>
<td>17</td>
<td>1 day 27.3 hrs.</td>
<td>90% min.</td>
<td>46%</td>
<td>46%</td>
<td>92% min.</td>
<td>69% min.</td>
<td>.66</td>
</tr>
<tr>
<td>18</td>
<td>1 day 1.0 hrs.</td>
<td>127% min.</td>
<td>32%</td>
<td>15%</td>
<td>196% min.</td>
<td>154% min.</td>
<td>.77</td>
</tr>
<tr>
<td>19</td>
<td>1 day 1.5 hrs.</td>
<td>97% min.</td>
<td>50%</td>
<td>20%</td>
<td>157% min.</td>
<td>94% min.</td>
<td>.69</td>
</tr>
<tr>
<td>20</td>
<td>1 day 1.5 hrs.</td>
<td>97% min.</td>
<td>43%</td>
<td>35%</td>
<td>94% min.</td>
<td>59% min.</td>
<td>.65</td>
</tr>
<tr>
<td>24</td>
<td>5 days 0.2 hrs.</td>
<td>200% min.</td>
<td>30% (c.)</td>
<td>4%</td>
<td>204% min.</td>
<td>160% min.</td>
<td>.79</td>
</tr>
<tr>
<td>25</td>
<td>1 day 32.8 hrs.</td>
<td>100% min.</td>
<td>40%</td>
<td>40%</td>
<td>150% min.</td>
<td>99% min.</td>
<td>.76</td>
</tr>
<tr>
<td>26</td>
<td>1 day 32.0 hrs.</td>
<td>25% min.</td>
<td>40% min.</td>
<td>25% min.</td>
<td>79% min.</td>
<td>61% min.</td>
<td>.77</td>
</tr>
<tr>
<td>27</td>
<td>3 days 0.8 hrs.</td>
<td>140% min.</td>
<td>13%</td>
<td>20%</td>
<td>196% min.</td>
<td>130% min.</td>
<td>.85</td>
</tr>
<tr>
<td>28</td>
<td>3 days 1.1 hrs.</td>
<td>104% min.</td>
<td>22%</td>
<td>15%</td>
<td>133% min.</td>
<td>97% min.</td>
<td>.82</td>
</tr>
<tr>
<td>29</td>
<td>3 days 2.0 hrs.</td>
<td>54% min.</td>
<td>22%</td>
<td>10%</td>
<td>73% min.</td>
<td>59% min.</td>
<td>.83</td>
</tr>
<tr>
<td>30</td>
<td>3 days 3.0 hrs.</td>
<td>164% min.</td>
<td>14%</td>
<td>7%</td>
<td>171% min.</td>
<td>132% min.</td>
<td>.83</td>
</tr>
</tbody>
</table>
In the first extraction, $X = 0$, since the radium flask was evacuated immediately after it was filled with the freshly prepared radium solution. Thus, in this case, $P = N^*_0 = 89$ mcs. But the observed yield was 58 mcs., so that $A = 0.65$, and $X$ for the next extraction $= 31$ mcs. A value of $N_{obs.}$ has been calculated for the next extraction, in which the yield was dropped before it could be measured, using a mean value of 0.7 for $A$. This enables a calculation of $X$ for the next extraction, and thereafter the calculations are straightforward.

In view of the anomalous yield in extraction (62), a value of $X$ for the next case has been obtained from a calculated value of $N_{obs.}$ in extraction (62). Similarly, a calculated value of $X$ has been used in extraction (24), since extraction (22) was anomalous.

It will be seen that $A$ is not constant, but is much more nearly so than the ratio of $N_{obs.}$ to $N_p$. Thus, over the last seven extractions, when the operation has been fairly consistent, the mean value of $A$ is 0.800, with a maximum deviation of 0.65 in only one case where $A = 0.85$.

The mean ratio of $N_{obs.}$ to $N_p$, however, is 0.903 over the same extractions, and values vary from 0.80 to 1.15.

Such "mean" values of $A$ are obtained from the ratio of the sum of all $N_{obs.}$ values to the sum of all $P$ values over the same range, and similarly with $N_{obs.}: N_p$.

Thus it can be said that equation (35), gives a fairly good approximation to the observed yield in all cases. It is interesting to it is also true of extreme cases such as extractions 26 and the time interval since last extraction was short, and the $n$ of the $X e^{-A_t}$ term is quite large.

Yet of other factors on the yield, and hence on $A$ may now
be considered. For this purpose, values of A have been included in table 1.

The period of opening the valves appears to have only very little influence on the yield. Thus over extractions 3 to 6, the average total period was 51 seconds, with a mean A value of .747. Keeping other factors constant, this period was successively lowered, and over extractions 13 to 20 it averaged 22 seconds, with a resultant mean A of .700. This is slightly lower, but not very significantly so, since the yields were rather inconsistent at this stage.

The volume of ice formed was alarmingly large however, over the first few extractions, and the lowest possible period is advisable on this score. Over the thirty extractions so far carried out, some 22 cc. of water have been removed from the radium flask, as determined from the drop in solution level. The total period of opening the cut off system has been 931 seconds. Neglecting the loss due to hydrogen and oxygen, estimated at approximately one cc. of water per year, these figures imply a loss of 1.06 cc. of water per week of three extractions, each with a total period of 15 seconds. Reduction of this time to even 10 seconds appears possible as regards its effect on yield, so that the yearly loss of solution would be only 34 cc.

Over the last few extractions, a third stroke of the toepler pump has been made, keeping a constant valve period of 15 seconds. The mean value of A has thereby increased from .70 to .80 so that this procedure appears justifiable. The extra time involved is only 2.5 to 3 minutes, at a very low level of gamma activity.

On a few recent extractions, 28 and 30, the liquid air trap has been evacuated for total periods of one minute, in place of the normal constant figure of 20 seconds. It will be seen that no appreciable diminution in
A resulted, even although two condensations of the radon were employed. Thus where high concentrations of radon are required, this procedure is quite permissible.

Concerning comparisons with other plants, it appears to be customary to express the yield as a percentage of $M_0$, or better, as a mean percentage over several extractions. Over the last seven extractions this mean, as defined above, is 90.3% ranging from 80% to 115%. This compares not unfavourably with Oddie's figures (11) which range from 79% to 90%. It should be pointed out however, that the actual value of this ratio which is observed, depends to a great degree on the frequency of extractions. The ratio is always high for a short period of accumulation since the remains from the previous extraction have not had time to decay appreciably. Thus the observed mean of 90.3% could be raised to, say 95% if extractions were performed daily, which is the case with Oddie's plant, except over week ends.

Although the above $A$ values imply a yield of only 80%, this is based on the assumption that all the radon lost at each extraction is retained in the radium solution. This is not strictly true, since some little radon is lost through small factors such as blockage in the dry ice trap, absorption on the glass, and decay during extraction etc. In one case, the residue in the plant was exhaustively transferred to a second container with the teoppler pump, without opening the cut off system, and 1.7 milli-curies were obtained.

This result indicates the reasons for fluctuations in $A$, other than those due to the factors outlined above. Probably the actual yield is about 85% of the total radon available at each extraction, which is not inconsistent with a solubility loss of 13% in the radium solution.
PURITY.

Method of measurement.

Determinations of concentration of the radon required a knowledge of the total volume and pressure of the gas isolated. Such have been obtained by the use of a special container for the radon, shown in figure (11)B, opposite page (39). The capillary tubing used has been of uniform one millimetre bore, and has been sealed off as bluntly as possible at the end, so that the volume of gas isolated above the mercury level is very nearly cylindrical in shape.

Prior to sealing off the radon container, the mercury is raised above the constriction in this capillary tube, and reaches equilibrium with that in reservoir (41) in figure (5), top (32) being left open for this purpose. Under these conditions, the volume of gas so isolated will be equal to \( l \) where \( l \) is the length of the entrapped cylinder of radon, and \( s \) is its cross-sectional area.

Further, the pressure "\( P \)" of the gas in centimetres of mercury, will be given by the equation:

\[
P = A - (x + y)
\]

where \( A \) = atmospheric pressure in cms. of mercury

\( x \) = height of the mercury level in the capillary above that in the reservoir

\( y \) = the capillary depression of mercury in the capillary of one millimetre bore.

\( x \) has been obtained with the aid of a levelling device, and \( y \) has been calculated as \( +0.3 \) cms. To minimise errors, a series of corresponding readings of \( l \) and \( x \) has been made for each sample of gas. The following set
of readings will illustrate the method, and the degree of accuracy obtainable.

In this case, the gas to be determined was first compressed into a short length of capillary, with the mercury reservoir (41) raised fairly high. After the mercury had reached equilibrium, values of 1 and x were measured, and reservoir (41) then lowered a few centimetres. x and 1 were again measured, and the process was once more repeated, with the following results:

<table>
<thead>
<tr>
<th>1</th>
<th>x</th>
<th>y</th>
<th>A</th>
<th>P</th>
<th>F1</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>63.1</td>
<td>0.8</td>
<td>75.0</td>
<td>11.1</td>
<td>5.5</td>
</tr>
<tr>
<td>85</td>
<td>62.5</td>
<td>0.8</td>
<td>75.0</td>
<td>5.7</td>
<td>4.9</td>
</tr>
<tr>
<td>45</td>
<td>72.4</td>
<td>0.8</td>
<td>75.0</td>
<td>1.8</td>
<td>4.4</td>
</tr>
</tbody>
</table>

The product F1 should be constant for a gas obeying Boyle's law, but is higher for small values of 1 in this case, since the capillary was not strictly cylindrical in shape, but slightly tapered at its tip. On the other hand, measurements of very small values of P are likely to be in error, so a compromise has been made in taking a mean value for the three observations as

\[ F_1 = 4.9 \text{ cm}^2 \]

Assuming that the gas obeys Boyle's law, its volume \( V \) at 76 cm pressure will be given by the relation:

\[ F_1 x = 76 V \]

Substituting the above value of 4.9 cm\(^2\) for \( F_1 \), and taking \( x \) as 0.00785 for the capillary in use, we have

\[ V = \frac{0.000503 \text{ cm}^3}{0.003 \text{ cm}} = 0.503 \text{ mm}^3 \]

In this case, the yield of radon was 97 mcs., so that the concentration attained was 194 mcs. per cubic millimetre at 76 cm pressure and room temperature.
Results:

Such determinations of the total gas have been made at various stages of the purification, as shown in table III. The effect of continued exposure of the gas to KOH has been investigated, also double condensations with liquid air. These determinations have been possible only when the radon was not required for other purposes. Further, the radiation exposure involved has been very considerable, especially after second condensations, since the radon has by then reached an appreciable gamma activity. For these reasons, such determinations have been limited in number, and indeed undertaken with some trepidation.

Results have been calculated to three significant figures, but reduced to two figures in the table, as the accuracy of the measurements is not considered to warrant any more than this.
<table>
<thead>
<tr>
<th>Extraction No.</th>
<th>Yield</th>
<th>Stage of purification</th>
<th>Total time of exposure to KOH</th>
<th>Purity</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>75 ma.</td>
<td>1</td>
<td>9 minutes</td>
<td>9.1 mm.$^3$</td>
</tr>
<tr>
<td>4</td>
<td>85 &quot;</td>
<td>1</td>
<td>6 &quot;</td>
<td>4.9 &quot;</td>
</tr>
<tr>
<td>5</td>
<td>180 &quot;</td>
<td>1</td>
<td>5 &quot;</td>
<td>2.7 &quot;</td>
</tr>
<tr>
<td>6</td>
<td>93 &quot;</td>
<td>1</td>
<td>No KOH used.</td>
<td>6.6 &quot;</td>
</tr>
<tr>
<td>13</td>
<td>155 &quot;</td>
<td>1</td>
<td>3 minutes</td>
<td>15.0 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>6.2 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>5.9 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>5.8 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.30 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.70 &quot;</td>
</tr>
<tr>
<td>19</td>
<td>94 &quot;</td>
<td>1</td>
<td>0.5 &quot;</td>
<td>3.1 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2.5 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>4.5 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>7.5 &quot;</td>
</tr>
<tr>
<td>23</td>
<td>92 &quot;</td>
<td>1</td>
<td>0.5 &quot;</td>
<td>7.8 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3.5 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>7.5 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>8.0 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>12 &quot;</td>
</tr>
<tr>
<td>28</td>
<td>97 &quot;</td>
<td>1</td>
<td>0.5 &quot;</td>
<td>1.5 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2.5 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>4.5 &quot;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>5.0 &quot;</td>
</tr>
</tbody>
</table>

* in cubic millimetres at 76 cms. pressure and room temperature

† in millieuries per cubic millimetre at 76 cms. pressure and room temperature.
Discussion of results.

Results have been expressed in the conventional manner as millicuries per cubic millimetre at room temperature and 76 cm. pressure. 15.1 mcs. per mm.$^3$ is equivalent to a radon concentration of 1$\%$ of the total gas present, and this has proved very adequate for therapeutic purposes. With a few exceptions in earlier extractions, the observed results are well above this figure, so that the purification obtained has been sufficient for its intended purpose.

The comparatively poor concentrations attained in the first few extractions may be attributed to gaseous impurities in the radium flask which entered during filling operations, but could not be effectively pumped out in one extraction. Even so, the major constituent of the air which must have entered as above is nitrogen, and this would be removed in the purification process to which the radon is subjected.

Extraction (6) was undertaken with no KOH to test the quantity of CO$_2$ evolved. The resultant concentration of 13.8 mcs./mm.$^3$ is not very drastically below the previous figure of 48.3 mcs./mm.$^3$ obtained with the use of KOH, so that the amount of this gas formed in the purification process must be very small. This result affords some justification for the precautions taken to prevent the exposure of radon to grease. Silicone grease has also been used through out in hopes of reducing the CO$_2$, by this means, since it is lower in carbon content than the usual apiezon grease. Robertson has also used this grease, but with no definite results in its favour.

Extractions (13) and (23) show just how sensitive is the radon to any trace of grease. In both these cases, a little grease had penetrated above the mercury sealed area, and the resultant gas dropped considerably in amount
with successive treatments of the KOH, indicating that larger amounts of CO₂ had formed than in normal cases such as 19 and 23, where the decrease in total gas is only slight after prolonged KOH treatment.

The failure to reproduce the purity of extraction (5), of 48.3 mcs./mm.³ on the first condensation of the radon, has been explained in the case of extraction (6) since no KOH was used, but later results require further explanation.

It was thought that these comparatively poor results were due to incomplete evacuation of the liquid air trap, leaving a considerable residue of permanent gas. An investigation of the evacuation rate of the high vacuum pump followed after extraction 23; and proved quite definitely that this was so. For some reason, the efficiency of this pump had fallen to such a degree that vacua of the order of only 10⁻² mm. of mercury were obtainable, on 20 seconds evacuation of the liquid air trap. These began from initial pressures of 20 cms., as in working conditions. Such pressures in the 80 cc. volume of the "evacuated" section, yield values of (P₁) of the same order as those obtained in the presence of radon, being equivalent to one or two cubic millimetres of gas at 76 cms. pressure.

In view of these rather surprising results, the oil in the pump was changed, and evacuation was prolonged for one minute, with the results shown under extraction 28. Before discussing these, it should be stressed that this period of 20 seconds evacuation has been routine practice; and has been varied only where two condensations of the radon have been employed. In the first of these cases, extraction (13), the period was only 10 seconds at the first stage, and a further 10 seconds on second condensation. As such as 5 mm.³ of residual permanent gas on first evacuation is thus understandable,
so explaining the observed results of 5.8 mm.\(^3\) total gas. In extraction (23), the corresponding periods were 20 seconds on first condensation, followed by 10 seconds at the second stage. The large increase in concentration on such condensations is partly attributable to the above further evacuation.

To return, it was found that prolonged evacuation lowered the pressure to approximately 10\(^{-3}\) mm. of mercury, equivalent to 0.1 mm.\(^3\) of residual gas. The resulting increase in concentration in extraction 28 is now fully understandable; and the purity obtained of 60-100 mcs./mm.\(^3\) may be taken as the routine figure for the future. Such prolonged evacuation has been shown to have no appreciable effect on the yield, and in any case, 30 seconds evacuation is sufficient when the pump is in good order.

Even so, this figure has doubled on the second condensation, so that some 50% of the impurities present at the first stage are permanent gases, which must have become occluded with the radon as it condensed, in a kind of adsorption. The effect would be negligible on the second condensation because of the reduced pressure of such gases.

An analysis of the total gas isolated on such a second condensation is of necessity extremely approximate, but might run as follows, for a yield of 100 mcs.:

\[
\begin{array}{ccc}
100 \text{ mcs. of radon} & \ldots & \ldots & \ldots & \ldots & 1066 \text{ mm.}^3 & 13\% \\
\text{Permanent gases (H}_2\text{ and O}_2\text{)} & \ldots & \ldots & \ldots & 1 & 20\% \\
\text{incompletely evacuated,} & & & & & \\
\text{Other gas not estimated,} & \ldots & \ldots & \ldots & 334 & 67\% \\
\text{but assumed CO}_2 & & & & 500 \text{ mm.}^3 & 100\% \\
\end{array}
\]

There has been no evidence of the presence of water in the radon containers, nor have any drops of condensed water appeared on the surface of the mercury.
The above quoted results of 60-100 mcs./mm.\textsuperscript{3} on the first condensation, and 200 mcs./mm.\textsuperscript{3} on the second (230 mcs./mm.\textsuperscript{3} has been recorded) may be compared with Oddie's figures of 60 and 208 mcs./mm.\textsuperscript{3} respectively. The result depends on the time allowed for the purification, and if necessary, the above figures could be improved considerably.
TIME OF OPERATION.

The total time of operation of the plant, from the point at which the cut off system is opened, to the time of sealing off the radon container, has generally been 27 minutes for a routine extraction, involving 3 strokes of the teapot like pump, and one condensation of the radon. This figure is only slightly lower than Oddie's, of 30 minutes, under similar conditions. Since two pumping operations are involved in Oddie's plant, one such requiring 8 minutes in the plant described, it is felt that this total time should permit of further reduction.

However, of the above 27 minutes, 6 are expended on pumping operations, with remote control switching, and approximately 12 (to Oddie's 10) are allowed for complete condensation of the radon. During this time the operator receives virtually no radiation. A further 4 minutes are allowed for bubbling off liquid air, again with no appreciable radiation exposure, so that in all, only a brief exposure is involved.

Radiation exposure.

The dosage received by the operator has been measured daily with the aid of a Victoren dosemeter. This meter measures the discharge caused in a series of small condenser type of ionization chambers which have been carried on the body of the operator. Such chambers are charged to a constant potential before use, with the aid of the same dosemeter. The instrument is calibrated directly in r. units and affords a direct measure of the radiation received on a certain point of the operator's body.

The radiation received is due in part to the primary gamma radiation which penetrates the glass walls of the purification system. In addition, some primary beta radiation is received, issuing from Ra B and Ra C, since
the glass walls are not thick enough to absorb this radiation to any extent, although all alpha radiation is completely filtered. Further, a considerable amount of secondary radiation is received, being scattered by the body of the operator. All such radiation contributes to the ionization and consequent discharge set up in the above chambers, since their walls are of such thickness that beta radiation will penetrate them, and will also cause scattering comparable to that set up by the operator’s body.

Thus the dose recorded above is an approximate measure of the total radiation received.

For normal extractions of 100 milliuries, the total dose received has generally been 0.013 r., while for larger yields of 140 milliuries 0.03 r. have been received.

On second condensation of the radon however, at least a further 20 minutes are involved, with a very considerable increase in dose. This increase has never been less than 0.1 r., over the extra period involved, excepting that received whilst taking readings of concentrations. Oddie performs a second condensation in 15 minutes, allowing 10 minutes for the condensation itself, but it has not been possible to reduce the above 20 minutes to this extent. This may be due to the period of 4 minutes allowed for evaporation of the radon and bubbling off the liquid air, which may be excessive.

It will be seen that the time of operation is no insignificant factor, but is in fact of primary importance to the operator. Considerable satisfaction is felt with the small values of the dose received under normal conditions.
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