The University of Glasgow

Thesis Entitled

The Effects of Alpha Particle Bombardment in Liquids

Presented

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by

E.W.T. Richards.

The Department of Natural Philosophy,
The University,
Glasgow.

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Contents.

Preface.

The work embodied in this thesis was performed at Glasgow University during the years 1947 - 1951. The work was concerned with the problems of ionization in liquids due to Alpha Particles. Initial results showed a disagreement with the generally accepted theory proposed by Jaffé. A new and original theory is porposed and experiments designed to substantiate the theory were performed.

All the experiments described, with the exception of the work on scintillations from thin films, were designed and conducted by the author. The work on thin films was carried out in conjunction with Mr. J. I. Cole of this Department.

The author wishes to thank Professor P. I. Dee for his continued interest and many useful discussion in connection with the work; also the Nuffield Trust whose generosity made the work possible.

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The Effect of Alpha Bombardment on Liquids.

The effect of bombarding a liquid with alpha particles was first studied about 1906. Since that date a considerable amount of work has been carried out on the various problems connected with this subject. From 1906 until 1919, Jaffé and his associates investigated the purely physical side of the problem, connected with the primary ionization produced. Since this period, comparatively little work has been effected on this aspect, although detailed research has been carried out on the chemical and biological effects. It is therefore not surprising that there appears to be considerable difficulty in correlating the results of these chemical and biological experiments with those obtained from the purely physical ones.

In gases, it has been shown that the chief method by which an alpha or beta particle loses energy is by the creation of ions and it is thought that this is also the case in liquids. However, unlike the gaseous case, no clear experimental evidence has been obtained that this is so. For the case of electrons created by means of X-rays, up to 80% of the theoretical ion current has been collected in a liquid. For alpha particles, only about 1/500th part of this theoretical current is available for collection. This result has been explained by means of a theory proposed by Jaffé in 1919, in which he assumes that the initial ionization is restricted to a small cylindrical volume surrounding the path of the ionizing particle. Due to normal diffusion processes, there is a high probability for recombination taking place and he calculated that half the ions would disappear in about 10^{-9} secs. This theory was supported by the results of the experiments of Taylor and Molher in 1934, who used X-rays as a source of ionization. The chemical experiments usually take the form of irradiating a suitable chemical solution in a reaction vessel and measuring the chemical change produced for a given dose of radiation. It is found that the yield is largely independent of the rate at which the dose is given, as well as the concentration of the solute in which the change takes place. It is therefore supposed that the reactions must take place through a "chain of events", in which the solvent molecules play an important part. Further, in the majority of reactions where alpha or beta particles are used, the number of molecules of the solute affected, is approximately equal to the number of ions or ion-pairs, which would be expected to be produced by theoretical considerations in the solvent. The physical properties of certain chemicals have also been studied when subjected to irradiation. In one case the viscosity of a fluid which increased under bombardment, is reported to have continued to increase for up to two minutes after the irradiating source had been removed. Hence it would appear that, either the ions themselves or some product, such as radicals or excited molecules, must persist for considerable periods.

Returning to the physical experiments and to the theory proposed by Jaffé, there are two main objections. Firstly, no account is taken of the electrostatic forces which must exist in the ion columns. Kramers has shown that field strengths of about 10^{7} volts/cms. will exist, since the diameters of the columns are assumed to be of the order of 10^{-6} cms. Such fields will still further increase the rate of recombination and so decrease the times for which the ions are present in the liquid, thus reducing the charge which can be collected. The second is based on the results of the chemical experiments. As previously indicated, these show that the ions or radicals must exist in liquids for considerable periods. If the ions combine in the time suggested by the theory, accompanied by the direct formation of radicals; then there appears to be no reason why these radicals should not recombine in a similar period due to a similar process. It would therefore appear necessary for some process to exist by which means the radicals would be formed in a lesser concentration than that outlined above.

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Ionization Experiments 1906-1919.

During this period a considerable number of experiments were performed by Jaffé and his collaborators, which resulted in the formation of the column theory of ionization. The early work was mainly devoted to the determination of the mobility of the ions and their coefficient of recombination.

In 1910 Jaffé measured the mobility of the ions produced in hexane by means of beta particles. He used two methods, one, a modification of the method used by Rutherford for gaseous ions, the other due to Langévin. The same apparatus was used in both sets of experiments and was later altered and used to determine the coefficient of recombination. The principle of Langévin's method was that ions were created between two plain, parallel electrodes, spaced accurately a known distance apart. During the period of irradiation and for a known time after the source had been removed, a known field was maintained between the electrodes. The field was then reversed and all the remaining ions collected and measured. By varying the time between the removal of the source and the reversal of the field, it is possible to calculate the mobility of the ions.

In Rutherford's method, the initial ionization is confined to a small volume around one of the electrodes. The source is then removed and a known field established for a known time. As in the previous method the field is then reversed and all the remaining ions collected and measured. In this manner it is possible to obtain a direct measurement of the velocities of the ions. The advantage of this method is twofold. In principle, only small fields need be used and the velocities of negative and positive ions are measured separately. However, it has the disadvantage that it requires dense local ionization which is difficult to obtain in a liquid.

The apparatus consisted of a vessel in which two parallel electrodes were spaced 1 mm. apart, and completely covered with liquid. One of these electrodes was connected to a sensitive electrometer, the other to a suitable source of potential. (Fig.1.). As a source of ionization, Jaffé used 20 m.g. of Radium Bromide. This was shielded with lead blocks so as to produce a 1 m.m. beam of electrons, which passed through the ionization chamber and between the electrodes. A system of electro-magnets was arranged so that the source could be drawn rapidly into a lead shielded cavity. With the source in this position, the ionization due to the γ radiation was negligible. It was found necessary to expose the liquids to the beam of ionization for a period of between 1 and 2 minutes before the start of the experiment, so that consistent results could be obtained.

The timing was carried out by means of a "Helmholtz-

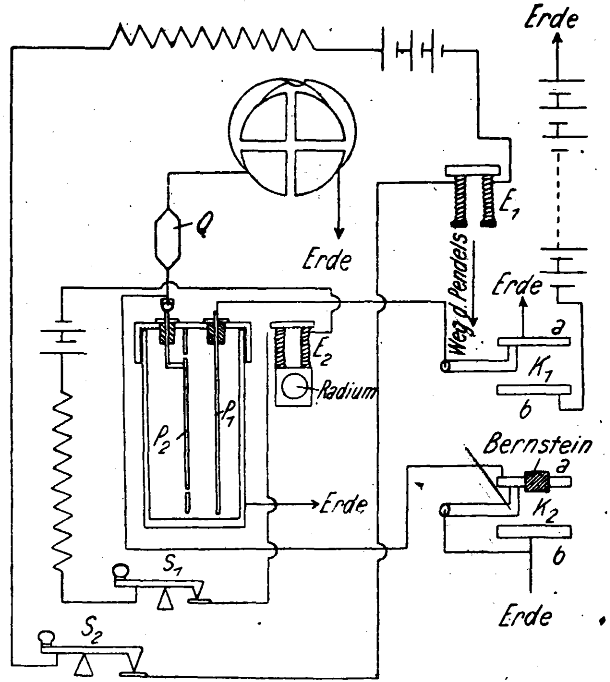


Fig. 1.

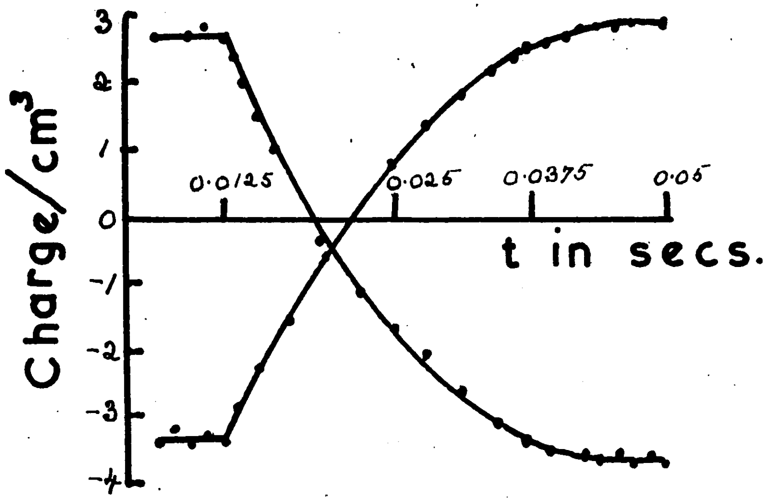
The Experimental Arrangement used by Jaffe and Van der Bijl

Edelmann" magnetic pendulum, which had a time constant of 6.84×10^{-4} seconds for one turn of the screw adjustment of the contact. The longest time which could be measured by this instrument was 8.55×10^{-2} seconds, so that an accuracy of one percent was possible. In liquids the mobilities of the ions are 3,000 times smaller than in a gas, so that to obtain the same order of collection times, 3,000 times the field strengths have to be used. This necessitates larger voltages, which might have caused sparking on the contacts and so invalidate the time measurements. Special precautions were taken to prevent this eventuality, such as splitting the voltage up into several sections and reversing each separately. The problem was further simplified by diminishing the spacing between the electrodes and in this way reducing the collection time. On the other hand, this lessens the accuracy, especially in the case of Rutherford's method, since the distance over which the initial ionization takes place becomes comparable with the electrode spacing. Accordingly spacings of between 1 and 2 m.m. were used. A further problem which arose in this connection was the "kick" received by the electrometer when the fields were reversed. In practise it was found that this did not materially affect the zero reading of the instrument.

Using apparatus of this type, Jaffé obtained the mobility of the ions produced in various liquids. (Table 1, Fig.2.). The two curves obtained are for fields in different directions. It will be noted that the curves are not symmetrical and that each has a "kink". This is caused by the fact that not all the ions have the same mobility. In particular, positive and negative ions have different velocities, but from these experiments alone it is not possible to determine which has the greater mobility. Jaffé resolved this point by the use of Rutherford's method, which has been previously described. For this experiment he increased the electrode spacing to 1.53 m.m. whilst retaining a 1 m.m. ionization beam, which passed across the face of one of the electrodes. A fairly large collecting field was used, so that the effects of recombination should be reduced to a minimum. This in turn means that small time intervals must be used with a consequent loss of accuracy. A typical set of results is given in Fig.3.

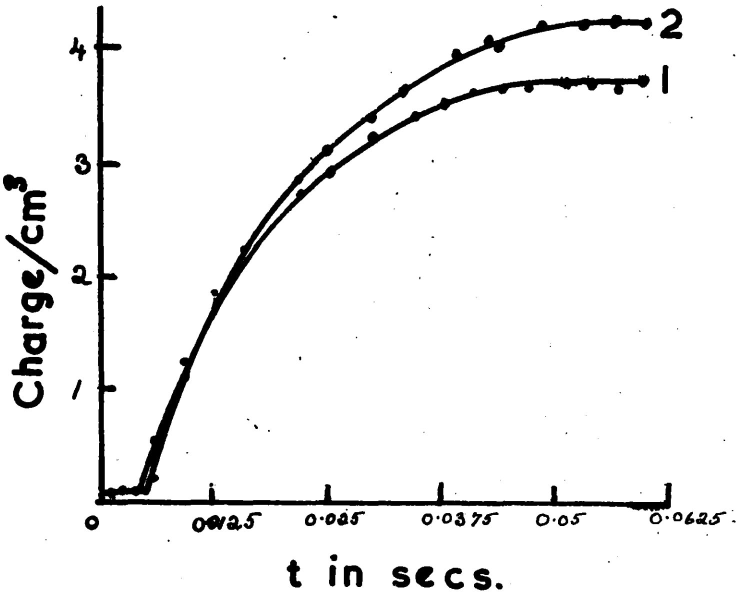
__Table 1.__ /

Fig. 2.



Ion Mobilities as Measured by Jaffe using Langevins Method. (Onecurve is taken with the field in each direction.)

Fig.3.



Ion Mobilities as measured by Jaffe using Rutherford's Method. (One curve is taken with the field in each direction.)

Table 1.

Condenser Spacing in m.m.	Intensity of Ionization in ESU/cm.	Field Strength Volts/cm.	Ion Mobilities. cm/sec/volt.	cm/sec/volt.
1.04	3.24 . 10	7550	5.92	3.77
1.04	3.22 . 10	6730	5.60	4.11
1.04	3.20 . 10	9430	7.00	4.47
1.53	4.12 . 10	6420	5.68	4.28
1.53	2.20 . 10	6420	5.94	4.24
		Average:	6.03	4.17

The mobilities of ions produced in Hexane measured by Langévin's Method. (Jaffé 1910).

Jaffé and Van der Bijl (1912) continued with similar experiments and succeeded in measuring the recombination rates of the ions. Van Bijl in his later experiments measured the charge collected on the electrometer, by a comparison with that produced by a crystal when subjected to pressure. By this method he was able to increase the accuracy of his results. Again two methods were used to determine the coefficient of recombination of the ions. The first was a direct measurement, and was accomplished by measuring the ions which remained in the liquid when no field was applied, at a known time after the ionization source had been removed. Time intervals of up to 120 seconds were used, and fairly large collection fields, approximately 2 K.V./cms., so as to minimise the effect of recombination during the time in which the ions are being collected. The coefficient of recombination was then calculated by means of the well known formula:

$$\frac{dn}{dt} = -\alpha n^2$$

A typical set of results are shown in Fig.4. and a value of 2.19 was obtained for .

The alternative method, utilised the fact that in the steady state, in a liquid subjected to ionization, the rate of the formation of ions is equal to the rate of loss. The general case of a liquid subjected to ionization is given by:

$$\frac{dn}{dt} = q - an^2$$

Where: n is the ion density,
α is the coefficient of recombination,
q is the rate of production of the ions.

Now in the equilibrium condition:

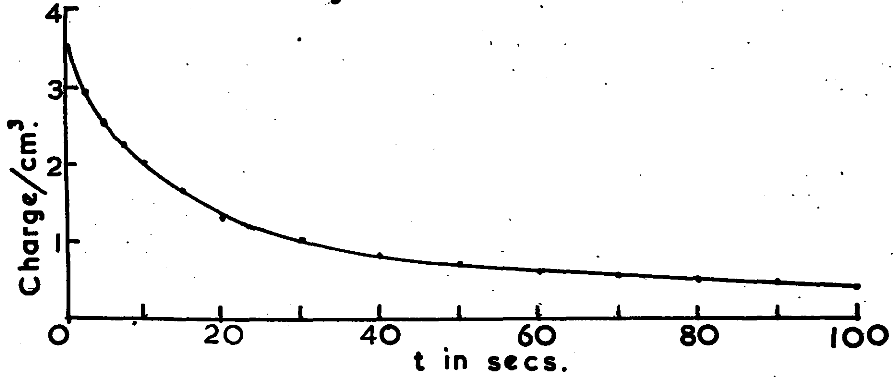
$$n^{\alpha} = q/\alpha$$

If it is assumed that the liquid is a perfect insulator when not subjected to ionization, the specific resistance of such a liquid under ionization is given by λ_o

$$\lambda_o = ne(u_+ + u_-)$$

Where e is the charge carried by each ion.
U_+ the mobility of the positive ions,
u_- the mobility of the negative ions.

Fig. 4.



Recombination Coefficients Measured by Van der Bijl. Direct Method.

Now the ionization current obtainable from a volume V of such a liquid, is given by I.

$$I = q\ e\ V$$

Combining these two expressions, we obtain an equation for the current recombination coefficient

$$a = e\ \frac{I}{V}\ \left(\frac{u_+ + u_-}{\lambda_o}\right)^2$$

The experiment consisted of measuring the ionization current and the specific resistance of the liquid, when subjected to irradiation, after equilibrium conditions have been reached. This was done both with a parallel plate electrode assembly and with a cylindrical arrangement. The results obtained are shown in Table 2. It will be seen that these results are in good agreement with those obtained by the previous method.

Table 2.

	Current in E.S.U.	S/V in E.S.U.	a
Parallel Electrodes	1.725 . 10	7.58 . 10	2.27
	1.42 . 10	5.18 . 10	2.26
	1.04 . 10	2.86 . 10	2.27
	7.48 . 10	1.48 . 10	2.27
Cylindrical Electrodes	5.31 . 10	7.34 . 10	2.20
	3.80 . 10	3.81 . 10	2.25
	9.67 . 10	2.60 . 10	2.30
		Average:	2.26

Values obtained for the recombination constant as measured by the indirect Method. Van der Bijl. (1912).

In the experiments so far discussed, the ionization was caused by beta particles. Experiments were also carried out in which alpha particles were used as a source of ionization. Unfortunately, in spite of the fact that these experiments would provide a more rigorous test of the "Column Theory", they are far more difficult to perform, and consequently no such thorough examination has been carried out. The experimental difficulties arise primarily from the short range of an alpha particle in a liquid. In the case of particles emitted from Polonium, the range in water is of the order of 32 microns. The problem which immediately arises is how to insure that at least some of the tracks occur between the electrodes. Two methods were evolved to overcome this difficulty. The first used by Jaffé (1914) was to dissolve an alpha source in the liquid in which the measurements were to be made. For this purpose he selected Radon as the source and Hexane as the liquid. This method has the disadvantage that it is impossible to collimate the alpha particles with respect to the collecting field.

In the experiments performed by Regner (1911) an attempt was made to overcome this difficulty. The source, in this case Polonium, was deposited on one of the electrodes. In this arrangement it is still impossible to collimate the particles, but this trouble was overcome to a certain extent by using different shaped electrode assemblies. Fig.5.

The results of both Jaffé's and Regner's experiments do not appear to give conclusive evidence with regard to the "Column Theory". In both experiments, (Fig. 6 and 7) the curves obtained are similar. In the majority of his experiments Jaffé unlike Regner did not obtain a saturation current. Both sets of results show fair agreement with the theory, but as it was not possible to collimate the particles, a rigorous test was impossible. However, it is of interest to note that as predicted the results obtained in various liquids are similar, as the ion mobility does not occur in the expression for the proportion of ions which are available for collection.

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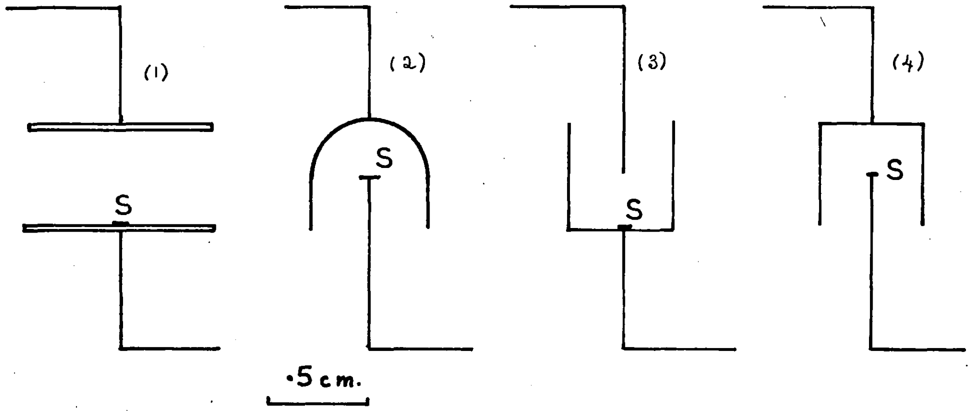
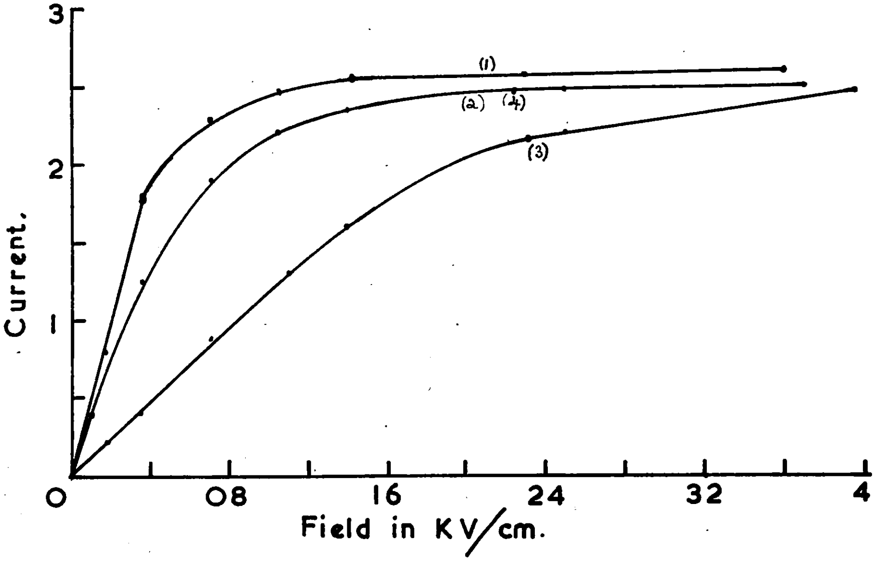
Fig. 5.



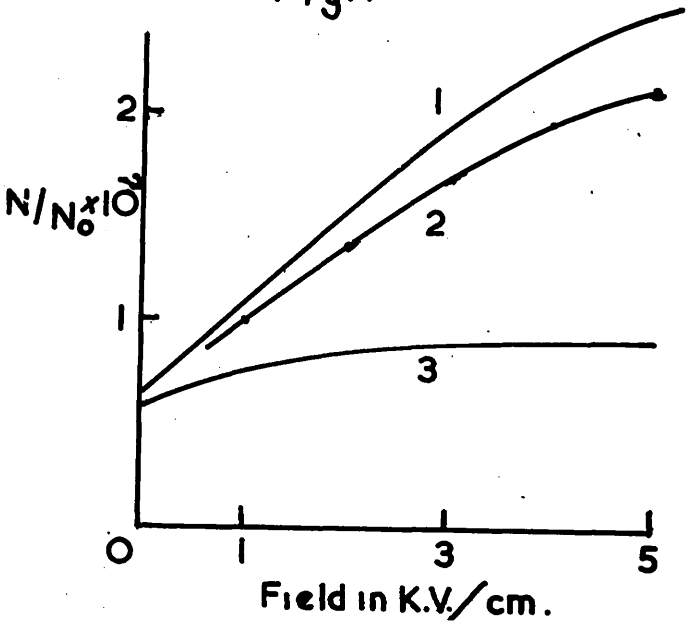
Electrode Designs used by Regner.

Fig.6.



Ionization Current Produced by Alpha Particles. Regner's Measurements.

Fig.7



Ionization Currents Produced by Alpha Particles compared with the Theoretical Currents, (Jaffe.)

Curve I Theoretical, Alpha Particles at Right Angles to the Field.

Curve 2 Theoretical, Alpha Particles Parallel to the Field.

Curve 3 Theoretical, Alpha Particles with Random Directions.

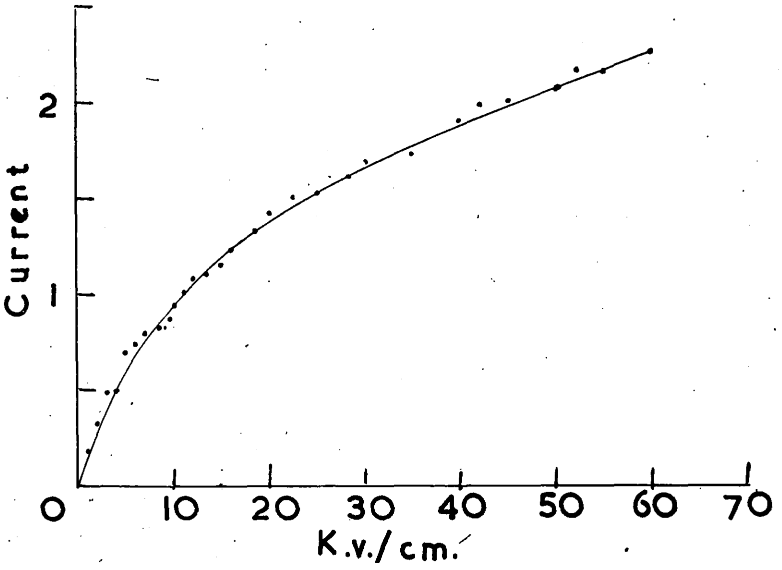
X-Ray Experiments. F.L.Mohler and L.S.Taylor.

Mohler and Taylor (1934) performed a series of experiments on the ionization produced in liquids by means of X-rays. In their experiments the liquid chosen was carbon bisulphide, one of the liquids used by Van Bijl. The liquid was very carefully purified, and in some of the experiments the apparatus was arranged so that the ion chamber could be filled from a vacuum distillation plant. In practise this procedure was abandoned since the extra precaution did not result in appreciably more stable conditions.

The electrode assembly consisted of two aluminium discs, 8 cms. in diameter, separated by a mica ring, 1 m.m. thick and 2 cms. wide. Aluminium was selected so as to reduce the "wall" effect to a minimum. The beam of X-rays was collimated so that it passed through the centre of the electrodes, so producing ionization in the enclosed liquid. Collection fields of up to 80 K.V./cms. were applied and the ionization current plotted as a function of the field strength. (Fig.8.). The maximum value of the collection field was not determined by the "breakdown" strength of the liquid, but rather by the electrical noise produced in the cell. It was found that this noise increased rapidly with the field applied, until at 80 K.V./cms. it became of the same order as the ion current.

The radiations were produced from a 120 K.V. X-ray tube,

Fig. 8.



X-ray Ionization Current, Mohler and Taylor.

special care being taken to ensure that the potential across the tube electrodes was constant. Intensities of 0.12 r/sec. were available, and the current produced in the liquid cell was compared to that produced in an exactly similar air ionization chamber. Currents of up to 2,500 times greater were measured in the liquid chambers, which represented an electrode efficiency of 80% of the current which would be theoretically expected to be produced by the initial ionization.

The results of these experiments were in accordance with Jaffe column theory. However, since the majority of the ionization produced by X-rays, is due to the secondary electrons liberated, this clearly provides no conclusive proof. It was suggested that this type of ion chamber might provide a suitable method of measuring X-ray dosage, but in view of the high field strengths required, it would seem to preclude such a development.

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More Recent Work.

In more recent years, considerable work has been devoted to the study of ionization phenomena in liquids at low temperatures. Since these experiments have often been carried out in the liquids obtained from noble gases it is not proposed to deal with these experiments in any detail. A number of these experiments have been devised with a view to producing a liquid proportional ion chamber or possibly a liquid "Geiger Counter".(1) These would be of great value because of the higher "stopping" power of a liquid as compared with a gas. Unfortunately, except in the case of the noble gases, no electron currents have ever been detected in a liquid. In the cases of liquid argon and neon, electron currents have been detected, and these currents have been multiplied by increasing the potential across the ionization chamber. In the case of helium, there is some evidence of an electron current being produced, but no amplication has been obtained. This is presumably due to the higher electron affinity which must exist in this case, so causing the trapping of the electrons to form negative ions. In all other cases no electron current is observed, this includes liquid nitrogen and hydrogen.

A.N.Gerriston (1948) performed a series of experiments on a number of liquified gases, including nitrogen and hydrogen. He obtained similar results to those from the earlier experiments, but like these no collimation of the alpha particles was possible. The source was mounted on one of the electrodes as in the experiments by Regner. So as to eliminate any trouble due to the noise produced in the cells when subjected to high field strengths, the experiments were repeated with the source partially covered with a foil. The apparatus used was similar in principle to that used in the previous experiments, the spacing between the electrodes being of the order of 1 m.m. The ion currents were measured by means of a Lutz-Edelmann electrometer, and care was taken in stabilizing the voltage between the electrodes.

As previously stated the general form of the results was the same as those obtained by Jaffé, (Fig. 9 and 10) but tended to confirm Kramers' modification to the "Column Theory". However, the results were not sufficiently precise to determine accurately the diameter of the ion columns.

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(1) See Bibliography. G.W.Hutchinson. ~~and~~ N.Davison and A.E.Larsh.

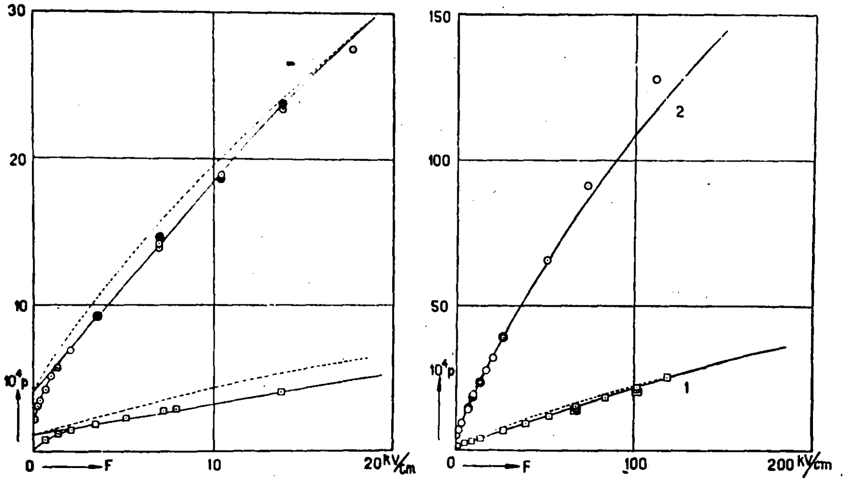

Fig 9 + 10 The ionization current in liquid nitrogen (77°K) as a function of the field, for weak and strong fields separately. The curves are calculated with Kramers's formula for p^* (dotted lines: $p = f\psi(f) + p_D$). Measured points: uncovered sample, positive (⊙) and negative (◎) ions; covered sample, positive (⊡) and negative (▣) ions.

The Column Ionization Theory.

In 1913 George Jaffé proposed his theory of ionization. This was that when a fluid is subjected to ionizing radiation, then columns of ions are formed along the tracks of the ionizing particles. The recombination rate of the ions so formed is largely due to the normal diffusion processes, and therefore to a first approximation is a function of the density in the columns. This is particularly true in the case of alpha particles where the track density is highest, the average ion spacing being of the same order as the molecular distances. Jaffé calculated the recombination coefficient for such formations and showed it to be in fair agreement with experimental results.

The recombination rate in one of these columns when first formed is given by:

$$(1) \quad \frac{\partial n}{\partial t} = D\left(\frac{\partial^2 n}{\partial r^2} + \frac{1}{r}\frac{\partial n}{\partial r}\right) - \alpha n^2$$

Where n is ion density.
D is the diffusion constant,
α is the coefficient of recombination,
r is the average distance between ions.

Unfortunately, the solution of (1) leads to no constructive results. The equation is therefore modified by omitting the last term, since this was considered not to be of great importance during the early stages of recombination. Considering, the case of a section of one of these columns, we can re-write (1) in the reduced form:

$$(1)' \quad \frac{\partial n'}{\partial t} = D\left(\frac{\partial^2 n'}{\partial r^2} + \frac{1}{r}\frac{\partial n'}{\partial r}\right)$$

Integrating (1) we obtain:

$$(2) \quad n' = \frac{N}{\pi(4Dt+b^2)}\, e^{-\frac{r^2}{4Dt+b^2}}$$

where b is a constant of integration, which is related to the width of the columns by the following relationship:

$$(3) \quad \bar{r} = \sqrt{\frac{\pi(4Dt+b^2)}{4}}$$

and N is the linear ion density.

At a time t = 0, b^2 is very small and the ion density is given by:

$$(2)' \quad n_0' = \frac{N}{\pi b^2}\, e^{-\frac{r^2}{b^2}}$$

with the average distribution given by:

$$(3)' \quad \bar{r}_0 = b\sqrt{\frac{\pi}{4}}$$

Now (2) is a solution of (1) and N is a variable with time. Substituting (2) in (1), we get:

$$(4) \quad \frac{dN}{dt}\,\frac{e^{-\frac{r^2}{4Dt+b^2}}}{\pi(4Dt+b^2)} = \frac{-\alpha^2 N^2}{\pi^2(4Dt+b^2)}\, e^{-\frac{2r^2}{4Dt+b^2}}$$

Integrating over a cylinder of unit length and for r = 0 to r = ∞, so as to get the total differential for N we have:

$$(5) \quad \frac{dN}{dt} = -\frac{\alpha^2 N^2}{2\pi(4Dt+b^2)}$$

Integrating:

$$(6)\quad N = \frac{N_0}{1 + \frac{\alpha N_0}{8\pi} \log\left(\frac{4Dt+b^2}{b^2}\right)}$$

and so as a solution to (1) we obtain:

$$(7)\quad n = \frac{N_0}{1 + \frac{\alpha N_0}{8\pi D} \log\left(\frac{4Dt+b^2}{b^2}\right)} \quad \frac{e^{-\left(\frac{r^2}{4Dt+b^2}\right)}}{\pi(4Dt+b^2)}$$

For t = 0, (7) reduces to the equation:

$$(8)\quad n = \frac{N_0}{\pi b^2} e^{r^2/b^2}$$

which is of the same form as (2).

So far no account has been taken for the interaction between different columns. If, as is generally the case, the column spacing is large compared to the diameter of the columns, then this can be considered as a problem in simple diffusion and the distribution of the ions taken as being random. The normal formula of recombination can then be applied.

Before the theory can be compared with experimental results, it must be decided at what radius these columns may be taken to cease to exist. Let us now consider the case of a column expanding from a radius C, to such a radius R.

From this definition we get N, the diffusion losses of the column:

$$(9)\quad N_1 = -\int_0^\infty 2R\pi D\left(\frac{\partial n}{\partial r}\right)_R dt = 4DR^2\int_0^\infty \frac{N(t)}{(4Dt+b^2)^2} e^{-\frac{R^2}{4Dt+b^2}}$$

Introducing another variable:

$$(10)\quad \xi = \frac{R^2}{4Dt+b^2}\ ;\quad \xi_0 = \frac{R^2}{b^2}\ ;\quad d\xi = -\frac{4DR^2}{4Dt+b^2}\,dt$$

so we obtain the ratio of the surviving ions:

$$(11)\quad \frac{N_1}{N_0} = -\int_{\xi_0}^{0} \frac{e^{-\xi}\,d\xi}{1+\frac{\alpha N_0}{8\pi}\log \xi_0/\xi}$$

In order to obtain a result correct to 0.5% it is only necessary to make $\xi_1 = 6$.

Before using equation (11) we must consider which root will satisfy equation (1). From differentiating (7), we obtain:

$$(1'')\quad \frac{\partial n}{\partial t} = D\left(\frac{\partial^2 n}{\partial r^2} + \frac{1}{r}\frac{\partial n}{\partial r}\right) - \alpha\,\frac{e^{\left(\frac{r^2}{4Dt+b^2}\right)}}{2}\,n^2$$

It will seem that the difference between this expression and (1) is in the second term. This now contains a second factor which is time dependent. This can be interpreted as a variation in the recombination coefficient due to the spreading of the columns.

For $\frac{\alpha N_0}{8\pi D} > 100$ which is normally the case, (11) can be simplified to:

$$(12)\quad \frac{N_1}{N_0} = 8\pi D\int_{0}^{\xi_0} \frac{e^{-\xi}\,d\xi}{\log \xi_0/\xi}$$

Let us now consider the effect of a field of strength E applied along the axis of such a column. In a time T the column will be lengthened by an amount d given by:

$$(13)\quad T = \frac{d}{2uE}$$

where u is the mobility of the ions.

In the part of the column in which the positive and negative ions are still in contact, the recombination will continue according to equation (6). In a short time dt, ions of each sign will be drawn away from the column the number of ions which will be liberated by the field is n_2 given by:

$$(14)\quad n_2 = 2uEN_0 \int_0^T \frac{dt}{1+\frac{\alpha N_0}{8\pi D}\log\left(\frac{4Dt+b^2}{b^2}\right)}$$

Assuming the original linear ion density to be N_0 and D to be the column length, then the proportion of ions which will be liberated by the field η_1:

$$(15)\quad \eta_1(E) = \frac{n_2}{N_0 d} = \frac{u}{2D}\eta y_1 e^{y_1}\left(li\, e^{y_2} - li\, e^{y_1}\right)$$

where $y_1 = \dfrac{8\pi D}{\alpha N_0}$

$y_2 = \dfrac{8\pi D}{\alpha N_0} + \log\left(\dfrac{4Dt+b^2}{b^2}\right)$

and $li\ e^x = \displaystyle\int_\infty^{-x} \frac{e^{-x}}{u}\,dx$

The second case of practical interest is that in which the field is applied at right angles to the axis of the columns. Let us assume that the columns are parallel to the z axis and the field lies along the x axis. If there are n positive ions and n negative, the problem reduces to solving the two following differential equations:

$$(16)\quad \frac{\partial n_\pm}{\partial t} = D\left(\frac{\partial^2 n_\pm}{\partial x^2} + \frac{\partial^2 n_\pm}{\partial y^2}\right) \mp uE\frac{\partial n_\pm}{\partial x} - \alpha n_+ n_-$$

Further from the theory, div E = 0.

Using the same method to solve (16) as was used for (1), we get:

$$(17) \quad n'_{\pm} = \frac{N}{\pi(4Dt+b^2)}\, e^{-\left(\frac{\{x \pm uEt\}^2 + y^2}{4Dt+b^2}\right)}$$

Equation (17) is the same as (2) except for a term which takes into account the widening of the columns at a rate given by 2uE.

Combining equations (17) and (16) we get an expression for the rate of recombination.

$$(18) \quad \frac{dN}{dt}\,\frac{e^{-\left(\frac{(x-uEt)^2+y^2}{4Dt+b^2}\right)}}{\pi(4Dt+b^2)} = -\frac{\alpha N^2}{\pi^2(4Dt+b^2)}\, e^{-\frac{2u^2E^2t^2}{4Dt+b^2}}\, e^{-\frac{2(x^2+y^2)}{4Dt+b^2}}$$

Integrating over a cylinder of unit length we get the differential equation:

$$(19) \quad \frac{dN}{dt} = -\frac{\alpha N^2}{2\pi(4Dt+b^2)}\, e^{-\frac{u^2E^2t^2}{4Dt+b^2}}$$

which has a solution:

$$(20) \quad N(t) = \frac{N_0}{1 + \frac{\alpha N_0}{2\pi}\int_0^t \frac{e^{-\left(\frac{2u^2E^2t^2}{4Dt+b^2}\right)}}{4Dt+b^2}\,dt}$$

The value of N given by (20), can be substituted in (19) and so we can obtain a solution to (16). Equation (20) gives the number of ions at a time t as a function of the field.

Writing N_∞, as the number of ions which can be collected, we get:

$$(21) \quad N_\infty = \frac{N_0}{1+F(E)}$$

where:

(22) $$F(E) = \frac{\alpha N_0}{2\pi} \int_0^\infty \frac{e^{-\frac{2u^2E^2t^2}{4Dt+b^2}}}{4Dt+b^2}\, dt$$

Writing:

$$4Dt + b^2 = y$$

and: $$\frac{2u^2E^2(y-b^2)}{16D^2y^2} = s$$

we can write F (E) in the form:

(23) $$F(E) = \frac{\alpha N_0}{8\pi}\sqrt{\frac{\pi}{\alpha}}\, S(z)$$

where: $$z = \frac{b^2u^2E^2}{2D^2}$$

and so we get:

(24) $$S(z) = \frac{1}{\pi}\int_0^\infty \frac{e^{-s}ds}{\sqrt{s(1-s/z)}}$$

Combining equations (24) and (23) we get an expression for the efficiency of the electrodes:

(25) $$\eta_2(E) = \frac{N_\infty}{N_0} = \frac{1}{1+\frac{\alpha N_0}{8\pi}\sqrt{\frac{\pi}{z}}\, S(z)}$$

Let us now consider the general case in which the field makes an angle φ with the axis of the column. The lengthening of the column, due to this field, in a time T is given by:

(26) $$T = \frac{d}{2uE\cos\varphi}$$

During this period the column has also widened by an amount 2 u E sin φ. The ions which can be collected due to the crosswise component of the field are given by equation (20). The total number of ions n_s which can be collected by such a field is therefore:

$$(27) \quad n_3 = 2uE\cos\varphi \int_0^1 N_1(t)\,dt.$$

and the saturation curves by:

$$(28) \quad n_3 = \frac{2uE\cos\varphi}{d}\int_0^T \frac{dt}{1+\frac{\alpha N_0}{2\pi}\int \frac{e^{-\left(\frac{2u^2E^2\sin^2\varphi\, t^2}{4Dt+b^2}\right)}}{4Dt+b^2}}$$

This function has no singular solution. For small values of E, the solution is given by (18), when E cosφ is the important term. For large values of E, by (25) when E sinφ is the important term, i.e.:

$$(29) \quad \eta_3(E) \geq \frac{2uE\cos\varphi}{d}\int_0^T \frac{dt}{1+\frac{\alpha N_0}{8\pi D}\sqrt{\frac{\pi}{\zeta'}}\,S(\zeta')}$$

where:

$$\zeta' = \frac{b^2u^2E^2\sin^2\varphi}{2D^2}$$

Solving and putting in the constants of integration, we get:

$$(30) \quad \eta_3(E) \geq \frac{1}{1+\frac{\alpha N_0}{8\pi D}\sqrt{\frac{\pi}{\zeta'}}\,S(\zeta')} = \frac{N_\infty}{N_0}$$

It is now possible to give an upper limit to N_3 (E). It is given when N$'$(t) tends to (N$'$), but a good approximation can be obtained if:

$$(31) \quad \frac{2u^2E^2\sin^2\varphi\, t^2}{4Dt+b^2} = 6$$

Writing the value of t, given by this expression as τ, we get:

$$(32) \quad \eta_3(E) \leq \frac{\tau}{T} + \frac{T-\tau}{T} \cdot \frac{N_\infty}{N_0}$$

By combining equations (30) and (32), we can write for most fields, i.e. when E and φ are not too small:

$$(33) \quad \eta_3(E) = \frac{1}{1 + \frac{\alpha N_0}{8\pi}\sqrt{\frac{\pi}{z'}}\, S(z')}$$

where:

$$z' = \frac{b^2 u^2 E^2 \sin^2\varphi}{2D^2}$$

Fig.7. shows the theoretical curves and a series of experimental points plotted together. It will be seen that the extremely good agreement is obtained if a random distribution of tracks is assumed.

As will be seen, no account has been taken in Jaffé's theory of the electrostatic forces which must exist in the ion columns. Kramers (Gerristen 1948) calculated that these were of the order of 10 V/cms. and would therefore play an important part in the recombination phenomena. Returning to equation (1) and inserting a term due to the collecting field, we get:

$$(34) \quad \frac{\partial n_\pm}{\partial t} = \pm E F \sin\varphi + D\left(\frac{\partial^2 n_\pm}{\partial r^2} + \frac{1}{r}\frac{\partial n_\pm}{\partial r}\right) - \alpha n_+ n_-$$

Grouping and substituting Langevin's (1902) expression for the recombination coefficient. $\alpha = 8\pi E^{2}$ where E is the electronic charge.

$$(35) \quad \frac{\partial n_{\mp}}{\partial t} = \pm F E \sin\varphi + k T \Delta n - 8\pi E^{2} n_{+} n_{-}$$

Writing this in the symbolic form:

$$(35') \quad \frac{\partial n_{\pm}}{\partial t} = [f] + [d] + [\tau]$$

which gives the following ratios, for the time t = 0.

$$(36) \quad \left[\frac{d}{f}\right]_0 \sim \frac{kT}{bEF}, \quad \left[\frac{f}{\tau}\right]_0 \sim \frac{Fb}{8E^{2}N_0}, \quad \left[\frac{d}{\tau}\right]_0 \sim \frac{kT}{8E^{2}N_0}$$

These ratios obviously differ greatly for gases and liquids, and are also temperature dependent. In gases the value of b is of the order of 10^{-3} cms. under normal conditions, whilst for a liquid it has a value of the order of 10^{-6} cms. In consequence the number of ions produced in a gas per cms. is about 10^{4} whilst in a liquid this figure becomes 10^{7}.

Hence the magnitude of the ratios are in the case of gases:

$$\left[\frac{d}{f}\right]_0 \sim 3.10^{-4}\frac{T}{F}, \quad \left[\frac{f}{\tau}\right]_0 \sim 3.10\,F, \quad \left[\frac{d}{\tau}\right]_0 \sim 8.10^{-3}\,T$$

and for liquids:

$$\left[\frac{d}{f}\right]_0 \sim 3.10^{-1}\frac{T}{F}, \quad \left[\frac{f}{\tau}\right]_0 \sim 3.10^{5}F, \quad \left[\frac{d}{\tau}\right]_0 \sim 8.10^{-6}\,T$$

Hence as soon as $3 \times 10^{-1} \ll F$ (F in e.s.u.) the influence of diffusion in a liquid is small compared with that of the field. This occurs for fields greater than 27 K.V./cms. for temperatures of 300 K. and 1.8 K.V./cms. for a temperature of 4 K. In gases the diffusion effects are negligible for fields above 27 v/cms. at 300 K.

The ratio (d/r) shows that at room temperature the influence of diffusion is 2.4 times that of recombination in a gas, whilst in liquids the recombination is 400 times greater at 300 K and 30,000 times at 4 K. Further from the ratio (f/r) we see that this has the magnitude 1 for fields of 10 v/cms. for gases and 10^7 v/cms. for liquids.

In view of these considerations he decided to solve equation (1) by first neglecting the diffusion term. Denoting the efficiency of ion collection by p , we get:

$$(37) \qquad p = f\,\psi(f)$$

where:

$$f = \frac{F \sin\varphi}{F_0}, \qquad F_0 = \frac{4\sqrt{\pi E N_0}}{b}$$

and $\psi(f)$ is given by:

$$\psi(f) = \frac{2}{\sqrt{\pi}}\,\mathcal{F}(-\ln f)$$

$\mathcal{F}(\eta)$ is the function defined as:

$$\mathcal{F}(\eta) = \int_0^\infty \frac{\sqrt{\xi}\,d\xi}{e^{(\xi-\eta)}+1}$$

ξ having the same meaning as in Jaffé's work.

The value of this integral has been calculated by McDougall and Stoner for $-4.0 \leq N \leq 20.0$.

Using the property of $\mathcal{J}(\eta)$ that for $n \rightarrow -\infty$ (or for $f \rightarrow +\infty$)

$$\mathcal{J}(\eta)_{n\rightarrow\infty} \longrightarrow \frac{\sqrt{\pi}}{2}\left(e^{\eta} - \frac{e^{2\eta}}{2\sqrt{2}}\right)$$

it follows that:

$$p(f)_{f\rightarrow\infty} \longrightarrow 1 - \frac{1}{2f\sqrt{2}}$$

or substituting for f:

$$p^2 = 1 - \sqrt{2\pi}\;\frac{e\pi}{b}\;\frac{1}{F\sin\varphi}$$

which is the same as Jaffé's result.

Introducing the diffusion correction Kramers deduced the approximate solution.

(38) $$p^* = f(\psi)(f^*) + p_0$$

with $$f^* = f\left(\frac{2}{\pi}\int_0^{cf} e^{-\xi^2}\,d\xi\right)^{-1}$$

and $$c = 4\,\theta\,\sqrt{\pi}\;\frac{E^2 N_0}{T}$$

where θ is a number between 1 and 5.

For large values of f the integral $\int_0^{cf} e^{-\xi^2}d\xi$ tends to $\sqrt{\pi}/2$ and f^* to f. Consequently p^* tends to p as given by (37).

This solution has been obtained by following the solution of (34), without diffusion only up to the moment when the diffusion term becomes comparable with the recombination trem. The number of ions collected after that moment is represented by p_D. In weak fields this quantity will soon increase to a constant value. Due to the uncertainty of the value of θ the value of p_D, is not accurately predicted.

For small values of f, f ψ (f^*), will be smaller than f (ψ) f and shows a linear behaviour for f sufficiently small indeed, when f tends to zero,   tends to infinity and F (η) to 2/3 $\eta^{2/3}$ so (37) becomes:

$$(39) \qquad p_3 = \frac{4}{3\sqrt{\pi}} f (-\ln f)^{3/2}$$

For $f \rightarrow 0$, $f^* \rightarrow \sqrt{\pi}/2c$ and (38) becomes:

$$p_8^* = \frac{4}{3\sqrt{\pi}} f \left(\ln \frac{8\theta E^2 N_0}{kT}\right)^{3/2} + p_D$$

or:

$$p_3^* = C_T f + p_D.$$

In Jaffé's solution the linearity is a result of omitting the influence of the diffusion trem, whilst in this case, it results in taking diffusion into account. It would appear that in the case of gases, Jaffé's theory would in general give satisfactory results, because of the comparatively large effects of diffusion. In liquids, it is to be expected that Kramers solution will give the more satisfactory agreement.

* * * * * * *

Chemical Experiments.

The fact that some forms of irradiation may cause chemical reactions has been known for a long time. However, it has only been in comparatively recent years that the effects of ionization has been studied in detail. As in the purely physical experiments, most of the work has been done using gaseous reagents, but even in this field there appears to exist contradictory evidence. Unfortunately, the results are further complicated by the fact that most of the reactions, that may be made to take place, are reversible. In spite of this, it appears, in the case of liquids, that when alpha or beta particles are used, one molecule is changed chemically for each ion or ion pair produced.

Water is the liquid on which perhaps most research has been done. It is also the substance from which most conflicting results have been obtained.(1) This may partially arise from the fact that pure air-free water appears to behave in an entirely different manner to that of pure water in which a little air has been dissolved. In the latter case, the experiments tend to show, that one molecule of Hydrogen Peroxide is produced for each ion pair created. When air-free water is used, the efficiency of production of Hydrogen Peroxide is much smaller for short periods of irradiation. However, as soon as a small

(1) See Bibliography. Effects of Radiations on Water.

quantity of Hydrogen Peroxide has been formed the rate of production rises until it has once again reached the efficiency of the former case. In these experiments the efficiency of alpha particles, is of the order of 85% of that of beta particles of the same energy. Therefore it appears reasonable to assume that the formation of the Hydrogen Peroxide is in some way directly connected with the primary ionization, also the chemical reaction requires some form of impurity in the water.

Another reaction which has been studied in considerable detail, is that between Hydrogen and Iodine or Bromine in aqueous solution. According to F.C.Lanning and S.C.Lind (1938), a yield of one molecule of Hydrogen Iodide is produced for each ion pair created, regardless of whether alpha or beta particles are used. Further this yield is largely independent of the concentration, and of the rate of irradiation. These experiments were performed in the usual manner. A known quantity of Radon was dissolved in water, free Iodine added and the mixture placed in a reaction vessel. After a known time the yield was measured by the normal titration method. The ionization was calculated assuming that the particles lost all their energy by this method and that 30 electron volts were required to create each ion pair. By these direct measurements, the value of M/N, the number of molecules of the resultant chemical to the number of ion pairs produced, was found to be 1.87. When Bromine was used to replace the Iodine a value of 1.187 for M/N was obtained.

Similar experiments were carried out to investigate the decomposition of ammonia in water and potassium permanganate solution. In these cases values of M/N of 0.87 and 0.95 were obtained respectively. From these results Lanning and Lind concluded that real values of M/N should be either 1 or 2. They explained the non-dependence on concentration by assuming that a chain reaction was taking place. The first process was the production of ions in the water. This was followed by the recombination of these ions with the production of hydrogen peroxide. This then diffused over the whole volume of the solution causing the chemical reactions which were measured. This view is further supported by the results of most of the other chemical experiments. In particular the oxidation of ferrous sulphate which has been studied by many people in great detail.

Unfortunately, little or no work has been done on the chemical effects of ionization in liquids when under the influence of electric fields. Williams and Essex (1948) studied the effect of an electric field on the decomposition of Nitrous Oxide. They concluded, that when beta particles were employed, collisions between ions of the opposite sign played little or no part in the decomposition. The greater part of this was due to the splitting of the molecules by electronic collisions. The application of a field above that required to obtain half-saturation current increased the yield. This was thought to be due to accelerating the secondary electrons so that they had sufficient energy to cause further decompositions. On increasing the field further no extra yield was obtained, this was explained by assuming that all the electrons had been trapped to form negative ions and so were not available to cause further dissociations. It is difficult to estimate how far these results can be applied to liquids, as except in liquids obtained from some of the noble gases, no electronic currents have been observed.

One of the problems connected with the simple type of explanation outlined earlier, is the "protective effect" investigated by Dale (1942). He found that many chemical reactions which could be initiated by irradiation, could be retarded by the addition of very small quantities of certain chemicals. This reduction of yield was considerably larger than that which could be expected if account is only taken of molecular size and concentration as will be readily seen from Table 3 . A surprising fact is, that if one of these protective agents is irradiated alone in solution, only one molecular change per ion produced is recorded.

Table 3./

Table 3.

Solute.	p_2/p_1
Glycerine	$1.8 \cdot 10^{-4}$
Na Oxalate	$1.5 \cdot 10^{-4}$
Na Nitrate	$1.8 \cdot 10^{-4}$
Leucyl Glycerine	$1.3 \cdot 10^{-3}$
K Ferrocyanide	$1.0 \cdot 10^{-3}$
K Ferricyanide	$1.0 \cdot 10^{-3}$
Glucose	$4.3 \cdot 10^{-2}$
Sucrose	$3.2 \cdot 10^{-2}$
Na Formate	$5.9 \cdot 10^{-2}$
Fructose	$1.4 \cdot 10^{-1}$
Na Nitrite	$1.8 \cdot 10^{-2}$
Alloxagin-adenino-denucleotido.	1.0

$$p_2/p_1 \text{, is the: } \frac{\text{concentration of the solution}}{\text{concentration of alloxagin-adenino-denucleotido.}}$$

Various theories have been put forward to explain these results. In this connection, it is perhaps of interest to note the problem of the mobilities of the hydrogen and the hydxoxyl ion in water. J.B.Bernal and R.H.Fowler (1933) proposed a theory based on the idea of "charge exchange" which resulted from their work on the crystaline structure in water. They assumed that a hydrogen ion consisted of a proton in association with a normal water molecule. This configuration gave place to a H_3O^+ ion in which the proton was loosely bound. When such an ion made a suitable contact with a neighbouring water molecule, the proton was able to be exchanged. In this manner it was found possible to explain the mobilities found experimentally. This type of process offers an explanation of some of the chemical effects of ionization. The ions instead of recombining by means of direct collisions, may be able to neutralise one another, by means of a charge exchange process, leaving an equal number of radicals or excited molecules which may then initiate the chemical reactions.

Another suggestion is due to Lind and Bardwell (1928). They proposed the "Cluster Theory", in which it is assumed that each ion is surrounded by 20 or 30 molecules of the solvent. This group is then free to move about the liquid as a unit and is capable of causing chemical changes. Garrison pointed out that in suitable organic materials, up to 6 dissociations may be connected with each ion pair created. Further, on the average 25-30 electron volts are required to form one ion pair, while only in the neighbourhood of 4.5 volts are required to cause a dissociation. From energy considerations, such reactions present no problem.

From these and similar experiments it appears necessary to assume that the chemical effects must take place through the medium of chain reactions. Further, either the ions themselves or some product must be able to exist in the liquid for considerable periods after the irradiation has ceased. This has been directly demonstrated in the case of certain organic liquids. In one case, the viscosity of such a liquid was found to increase when subjected to irradiation. This increase in viscosity was observed to continue for periods of up to 2 minutes after the irradiating source had been removed.

* * * * * * *

Conclusions.

From the experiments previously discussed, there appears to be no obvious connection between the results of the purely physical and the chemical experiments. In the cases when alpha particles were used, practically every molecule along the path of the particle was ionized, and therefore considerable field strengths must exist. For an alpha particle from Polonium, it has been calculated, that 90,000 ion pairs are formed along a 32 micron track. Assuming a track width as given by Jaffé's Theory, the average ion spacing is of the order of $3 \,.\, 10^{-6}$ cms. From such close spacings high fields will result, Kramers has shown that fields as high as 10^{7} volts per cms. are to be expected. This result is supported by the fact that no electronic current has been measured in a liquid except in certain liquids produced from noble gases. It therefore seems reasonable that the electrons ejected from the molecules by the alpha particles in the formation of positive ions, will be captured by other molecules in the immediate vicinity to give rise to negative ions. This process will be aided by the short mean free path of an electron in a liquid, and so it might be expected that all the electrons are captured within a distance of 10 molecular distances of the parent positive ions. The field strengths thus produced, appear to be at least of the order of the break down strength of the liquid, and therefore the configuration is presumably unstable. This in turn means that there would be fewer ions which could be collected by means of an electric field, and presumably fewer ions available for chemical effects.

Considering the results of the chemical experiments, from arguments concerned with the independence on concentration, it appears necessary that some effect of ionization should persist for considerable periods. If it is assumed that the original ions recombine by some means such as a charge exchange system, and that radicals are produced directly from the original ions, then there seems to be little reason why the radicals should recombine in a similar time to that calculated for the ions by Jaffé. It will be noted that in Jaffé's theory, no account is taken of any electrostatic forces which may exist, but only the influence of normal diffusion processes. This difficulty might be overcome, if the recombination of radicals required some special condition such as an energy consideration. In the case of water, the liquid used in most of the experiments, it appears difficult to explain the co-existance of hydrogen and hydroxyl radicals in the high concentrations which would be necessary in an alpha track. If, however, some process was involved by which the radicles were produced in columns of about ten times the diameter, this problem would be largely obviated.

Most of the chemical effects which can be caused by ionization can also be produced by exposure to ultra violet light. In a few cases extremely high quantum efficiencies are found, of the order of $10^3 - 10^4$ molecules per quanta.(1) Now, if the emission of quanta accompanied the recombination of the ions, this might offer an explanation of the various chemical experiments. It may be argued that no such emission takes place as no quanta are observed if water in bulk is subjected to bombardment by alpha particles. However, if the radiation emitted occurred in an absorption band of water such quanta might well be re-absorbed before they could escape from the liquid. Returning again to energy considerations, these quanta could well have sufficient energy to produce radicles, as about 30 electron volts are required to produce an ion in water, while only about 4.5 volts are required to produce a radical. These radicals would then be free to cause chemical changes. As has been pointed out, only comparatively small distances have to be traversed by these quanta, before they are absorbed in order to produce radicals with the required spacial distribution. Again, on theoretical grounds the emission of such quanta does not appear to be impossible, since the energy expended by an alpha particle in producing each ion pair is considerably higher than the theoretical

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(1) See Bibliography. Effect of Ultra Violet radiations on solutions.

ionization potentials, and therefore it is reasonable to assume that some of the ions produced are in a highly excited state. It may well be that on recombination taking place between two such excited ions quanta emission takes place. Another possibility of the emission of quanta arises from the fact that not always the lowest electron may be removed in the formation of a positive ion by an alpha particle. If one of the inner electrons are removed, radiation such as K or L X-rays may take place before or after recombination.

Against this type of argument it may be stated that if the chemical actions are primarily due to quanta emission due to the recombination of the ions, then far higher yields should result, more in agreement with those obtained from the experiments using ultra-violet light. However, the quanta efficiency of the recombination process may be low, and if this is not the case the number of quanta per unit volume surrounding the ion columns will be extremely large and some saturation effect would be expected. This may well take the form of a considerable degree of recombination of the radicals before any chemical effect has been effected.

From these considerations the following experiments were undertaken.

Firstly, it was considered desirable to repeat Jaffé's work on the physical side on the ionization of liquids. In order to test his theory, it was decided to measure the difference in the current obtained when the alpha particles travel at right angles and parallel to the collecting field. Further, by measuring the ionization current obtainable from the various parts of the alpha tracks,to try to establish the effect of the ion density on the proportion of those which can be liberated. It was felt that this could most readily be accomplished by making use of the variation in the specific ionization along the alpha track.

Experiments were also devised in order to try to establish if quanta emission accompanied ionization by alpha particles. In this connection experiments were performed using thin films of solids, because of the experimental difficulty involved in producing thin layers of liquids. Further, since a photo-multiplier was used to detect these quanta, it was considered that the quanta emitted by most liquids would be of too high a frequency to be observed. However, some experiments were performed using liquids and use was made of the fact that certain phosphors when radiated with ultra-violet light will emit in the visible region.

Finally, in an attempt to justify the theory previously outlined, experiments were carried out in which the solvent was separated from the substance in which the chemical change was to be detected. In these experiments a thin layer of the pure solvent was subjected to irradiation from powerful alpha source. Immediately below this layer and protected from the direct influence of the alphas by means of a suitable screen was placed a solution of the chemical under investigation. After a known period of irradiation, this sample was tested for any chemical reaction. It was considered that the whole theory could be tested in this manner.

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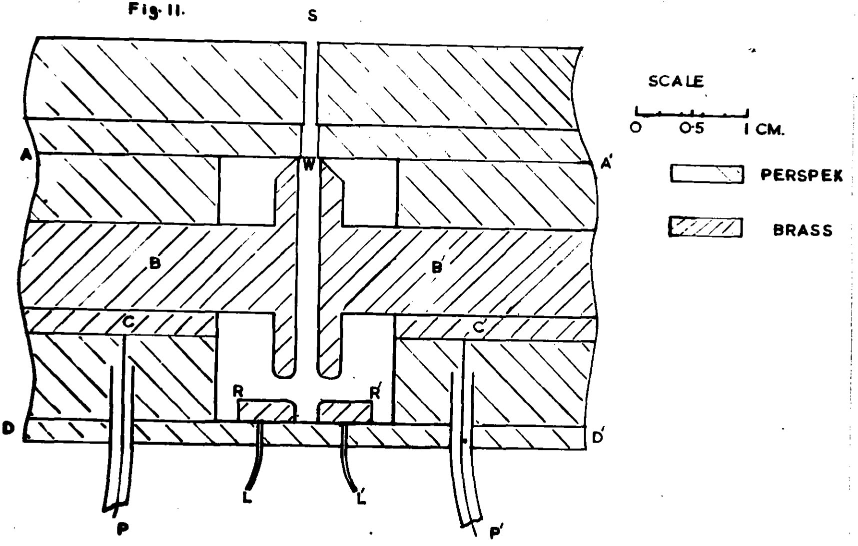
The Measurement of Ionization Currents in Liquids.

These experiments were designed to form a rigorous test of Jaffé's Theory when applied to liquids. For this purpose it was considered desirable to know the direction of the alpha particles relative to the applied field. The alpha particles were therefore colliminated. As the range in liquid is small, the collimination was carried out in air or vacuum before the particles were allowed to enter the ionization chamber.

For these experiments two types of ionization chambers were designed. The one for the case where the alpha particles travelled at right angles to the field and the other when they travelled along the field. In the former type, the electrode assembly was as shown in Fig.11. Various metals were tried in order to ascertain which was the most satisfactory material for use as electrodes. As no difference could be detected, brass was used for ease in manufacture. As will be seen in the diagram, provision was made to adjust both the height and the distance between the electrodes. These were mounted on perspex blocks, through which electrical connection was made by means of co-axial screened cables. Care was taken to ensure that at no point could the inner conductor act as an ion collector. The entire assembly was mounted in a perspex vessel. This material was chosen because of its good

Fig. 11.



The Electrode Assembly of the Liquid Ionization Chamber for Alpha Particles Right Angles to the Field.

A Liquid level. B Electrodes. C Brass slides.

D Chamber wall. L Leads to guard rings.

P Screened cables to electrodes. R Guard rings.

S Source, W Window

insulation properties. It was found that under certain conditions, conduction could take place along the surface of the perspex. This could be prevented by carefully washing the affected areas with chloroform. To further reduce the possibility of leakage currents, two guard electrodes were fixed to the base of the chamber directly under the collection electrodes. These were maintained at the same voltage as the electrodes, so that no field was applied across the perspex blocks on which these were mounted. The guard bars, also served to maintain a known field at the bottom of the electrode assembly. With this arrangement it was found possible to maintain an inter electrode resistance large compared to 10^{12} ohms.

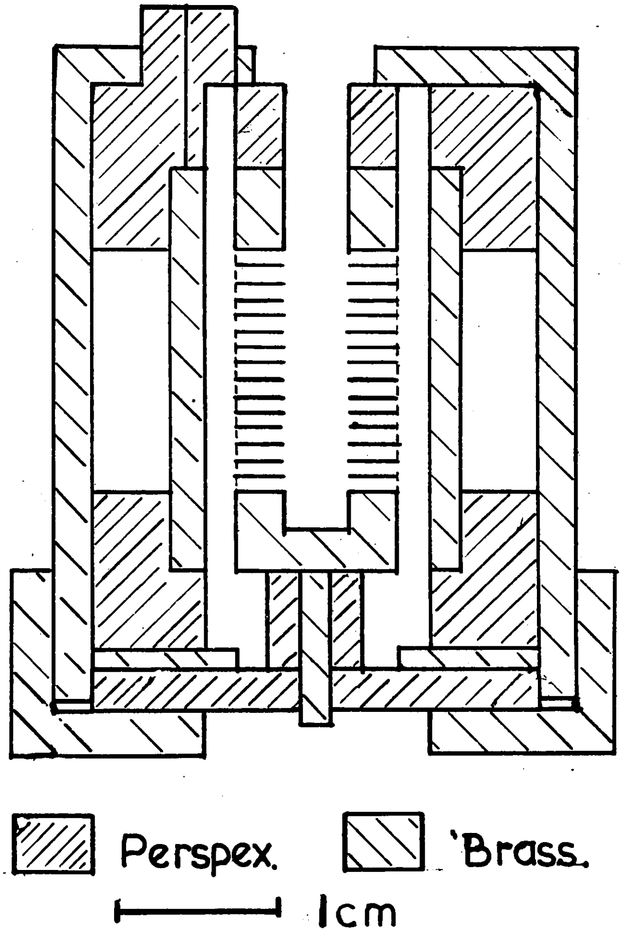
In the experiments, electrode spacings of between 0.5 and 2 mm. were used, the exact distances being measured by means of feeler gauges. The alpha particles were colliminated by means of a slit system, 0.5 mm. wide. This was made of perspex and mounted directly on top of the electrodes. It was found that if the particles were allowed to pass through a gas, that was in direct contact with the liquid, the ions formed in the gas were dissolved in the liquid and appeared as liquid ionization. This was presumably due to the image forces set up at the interface of the dielectricts. The currents due to this phenomenon were found to be up to 1,000 times as great as those due to ions produced in the liquid. Various systems of earthed screens and protective electrodes were tried unsuccessfully to prevent this effect. It was therefore decided that the liquid surface should be protected by means of a thin window. This caused considerable experimental difficulty, as the window had to withstand the full effect of the field and at the same time remain unaffected by the passage of the alpha particles. For this purpose, a window of thin pyrex glass, supported by means of a silk mesh, was used. Experiments were performed to determine the effect of the window by measuring the ionization current produced in apiezion oil, first with the window in place and then with no window, but with a vacuum above the oil surface. In this case, the oil level was carefully maintained at the same height as the top of the electrodes, and two guard electrodes were arranged 1 mm. above the liquid surface to collect any ions which might be formed in the vapour. No difference was detected in the ion current in the two cases, after account had been taken of the thickness of the window. In practise this was equivalent to about 0.5 cms. of air.

So as to protect the entire system from the influence of any stray electric fields, the outside of the chamber was coated with graphite and connected to earth. This also served, to protect the liquid from light. This was found to reduce the current passed by the liquid when subjected to an electric field without the presence of radiation.

The other type of ionization chamber was designed on the principle of two concentric cylindrical electrodes (Fig.12.) These were spaced 2 mm. apart by means of perspex rings. As in the former case, a guard ring was used to ensure that the insulators supporting the electrodes themselves were not subjected to any electric field. The centre portion of the inner electrode was fabricated from a series of rings and spacers. These formed a colliminator for the alpha particles, the source being suspended in the centre of the inner cylinder. The collimator was sealed by means of thin aluminium foil, which had an air equivalent of about 2 mm. In this manner, as in the former case, the liquid was prevented from coming in contact with gas through which the alpha particles had passed. The whole electrode assembly was enclosed in a brass cylinder which protected the system from the effects of stray electric fields. A larger chamber of this type was constructed so that ionization due to beta particles could be studied. In this case, thicker aluminium was used to seal the collimator, having a thickness equivalent to about 1 cm. of air.

The alpha sources used in these experiments was 20 milli curies of Polonium. This was mounted on a silver foil 2 mm. wide, 2 cm. long and 2 thou' thick. These sources were tested for contamination with other decay products of Radium, and were found to be free from beta

Fig. 12.



Electrode Assembly of the Concentric Ionization Chambers.

emitters.

The current measurements were made by means of a D.C. amplifier. This choice was largely conditioned by the comparatively large random current pulses which are observed when a liquid is subjected to high electric fields. These pulses, appear to be due either to the occurrence of local "break down" in the liquid or else to small quantities of impurities. They appear to travel with the same velocities as would be expected of liquid ions under similar conditions. This causes a high percentage of low frequency components in the current pulses observed. These pulses increase as the applied field is increased, until in these experiments at about 5 K.V./ cm. they became of the same order as the ionization currents. On integration these pulses give rise to what appears to be a steady background current which rises with the field until it reaches values in excess of the ion currents produced. It was therefore decided that a D.C. Amplifier in which this effect could be 'balanced out', would be more suitable than the older and simpler type of electrometer.

The circuit of the amplifier used was as shown in Fig. 13. The electrometer valve used was an E.T.I. manufactured by G.E.C. So as to maintain a constant potential across this valve, V_2 was connected as shown, as part of the anode load. This valve made it possible to obtain considerable amplification in the early stages of the amplifier, and so

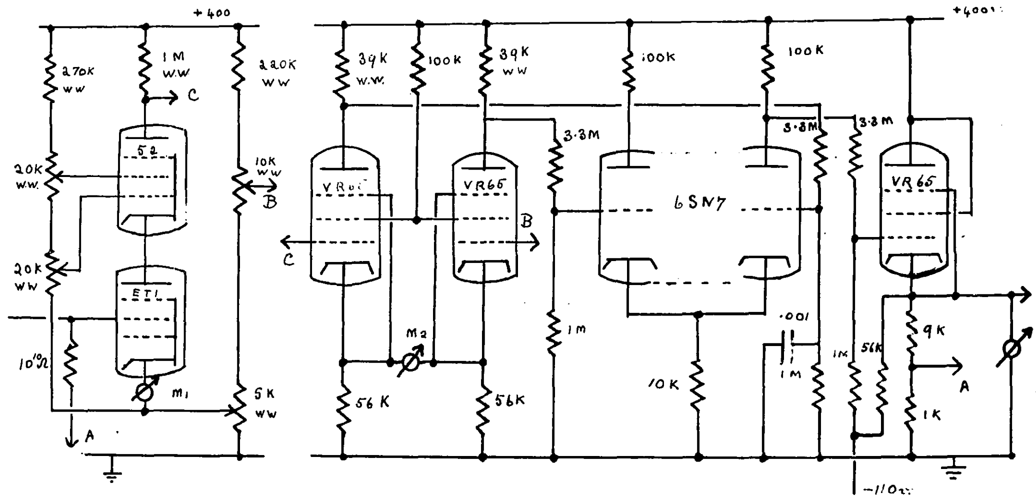


Fig. I3. D.C.Amplifier Circuit

tend to reduce the effect of any variation in the H.T. supplies on the apparent value of the ion current. The signals were then fed into a two stage D.C. 'push-pull' amplifier of conventional design. A 'feed back' circuit was arranged on the output of the 'push-pull' circuit, so that either the whole of the output or a portion of it could be fed to a tap-point on the input resistance. In this manner the gain of the amplifier could be adjusted. The input current was measured from the amplitude of the feed-back signals and the position of the tap-point on the input resistance. Such a system, whilst to a certain extent independent of small rapid changes in the various supply voltages, depends for long term stability on the constancy of these voltages. The amplifier was therefore supplied from two stabilized power packs of conventional design. So as to reduce this dependence to a minimum, it will be noted that all the bias potentials are taken from one chain of resistances. This also tends to eliminate the effect of any change in the value of these resistances. To reduce this possibility, all the important resistances used in the amplifier, were of the wire wound high stability type. With this apparatus, it was found possible to measure currents of 10^{-15} amps. using an input resistance of 10^{12} ohms. Unfortunately, due to the effects from the amplification of a field to a liquid, it was found impractical to use resistances greater than 10^{10} ohms. Consequently, the sensitivity of the amplifier was reduced.

The liquid used in most of the experiments was Hexane. This was chosen for two reasons. It was one of the liquids used in the early work on this subject, and it also possessed good insulation properties. It was found necessary to purify carefully the liquid used in the ionization chambers. (H.J.Plumley, 1941). Commercially pure Hexane was obtained and dried over sodium. The resultant liquid was then distilled and the chambers filled. Vacuum distillation directly into the ion chambers was tried, but no appreciable gain in the insulation properties was obtained. It was found that liquid thus purified could be used for a period of up to 24 hours after preparation. After this period sufficient moisture was absorbed to render it useless for ionization experiments. The electrical noise produced in the liquid could be reduced for short periods, if the liquid had been subjected to field strengths, in excess of that to be used, for a period of about 10 minutes immediately prior to the experiment. Use was made of this fact in all the experiments described. The Apezion oil used in some of the experiments was prepared in a similar manner to that used in the case of Hexane.

Experiments.

The first experiments carried out using these ion chambers were devoted to the determination of the effect of the collection field on the percentage of the ions which can be collected. Field strengths of up to 5 K.V./cms. were employed, this limit being set by considerations which have been stated earlier. Before each reading the liquid was stressed by a greater field than required, so as to reduce the electrical noise. Readings were taken of the current flowing, first with no radiation, and then with the source in position. In this manner it was found possible to reduce the effect of the noise produced in the liquid. In order to ensure that a steady condition had been reached, before the readings were taken the system was left under the experimental conditions required for a period of 10 minutes. Taking these precautions it was found possible to obtain reproducible results.

These experiments were repeated with both types of ion chambers, so as to obtain a direct comparison for the two cases. The amplifier was calibrated and a direct measure of the percentage of the ions which could be collected in a liquid found, by a comparison with an air ionization chamber. The same source and collimators were used both in the liquid and air ionization chambers. The ion currents were measured with the D.C. amplifier in both cases. This necessitated a change in value of the input resistance, due to the large increase in the value of the ion current in the latter case. The input resistances were compared in the usual manner, and the D.C. amplifier calibrated by measuring the ion current produced by a source of known strength.

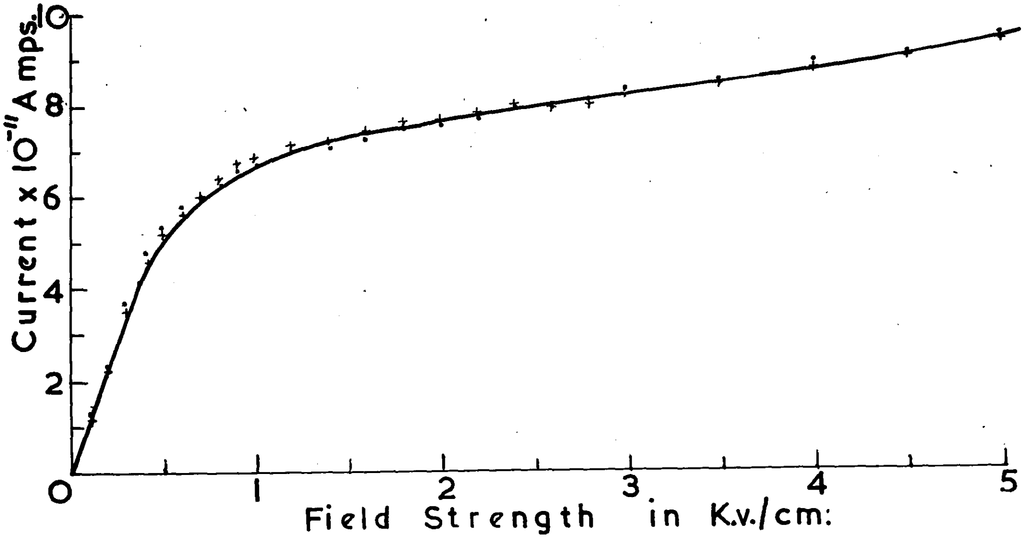
Experiments were performed to investigate the relationship between the ion density and the percentage of ions which can be liberated. For this purpose the variation in the specific ionization along the path of an alpha particle was used. In these experiments the cylindrical ionization chamber was used, because the placing of the absorbers between the source and the liquid would distort the field between the top of the electrodes in the other type of chamber. This is the most important region, since the range of alpha particles in hexane is short. For the particles emitted from a Polonium source, this is of the order of 32 microns. The primary ionization produced by an alpha particle in a liquid was assumed to obey the same law as in a gas. Therefore by reducing the energy of the incident particle, the ionization density should rise as indicated by the "Bragg Curve". In this manner the relationship was determined. The absorbers used were cylinders of thin aluminium leaf, having a thickness approximately equivalent to 0.8 mm. of air. Again, for each thickness of absorber a reading was taken, first without the source and then with the source in position.

The larger cylindrical chamber was used to perform similar experiments with beta particles. Again the source was mounted in a similar manner along the centre of the inner cylinder. The effect of the field strength was studied, and also the effect of placing absorbers between the source and the liquid. For these experiments several sources were used, including Thorium, Copper 64, and Rhubidium 85. Unfortunately a monoenergetic electron source was not available. The readings were taken in an exactly similar manner to that employed in the experiments in which alpha particles were used.

Results.

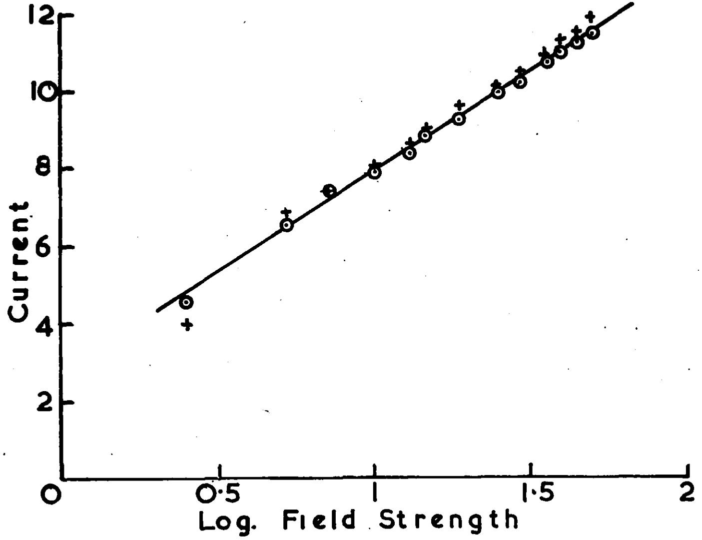
The results obtained from the experiments, when the alpha particles were travelling at right angles to the field, are given in Table 4 and Fig.14. It will be noted that the general form of the curve is in agreement with those obtained by Jaffé, as well as the percentage of the ions collected. However, Fig.14A. shows that relationship between the field strength and the current tends to obey a log. relation rather than an inverse function. In other words no linear portion is observed. This may be due to the fact that the field strengths were insufficient, but the results appear to be in good agreement with the logarithmetrical relationship. The results of the experiments when the alpha particle tracks were parallel to the applied field, are in complete agreement with the former case, Table 5, Fig.15. This may again be due to the limit imposed on the field strengths by the electrical noise level in the liquid, but at least in this region there appears to be no difference in the value of the ion

Fig. 14.



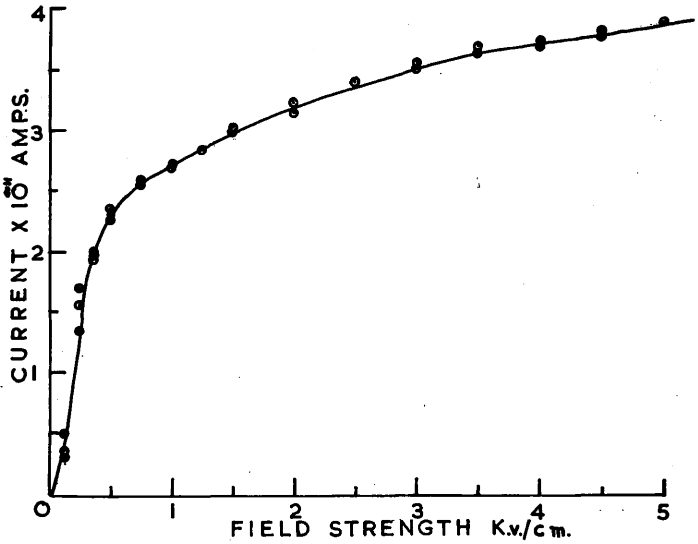
Field Strength- Current Relationship for Alpha Particles at Right Angles to the Field.

Fig. 14A.



Field Strength - Current Relationships for Alpha Particles at Right Angles and Parallel to the Field.

FIG. 15.

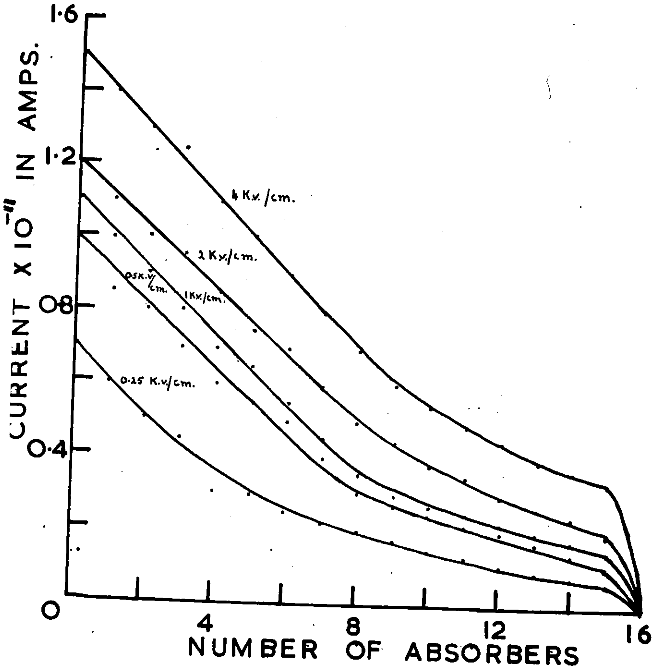


Field Strength - Current Relationship, Alpha Particles Parallel to the Field.

currents greater than the inaccuracies of the experiments.

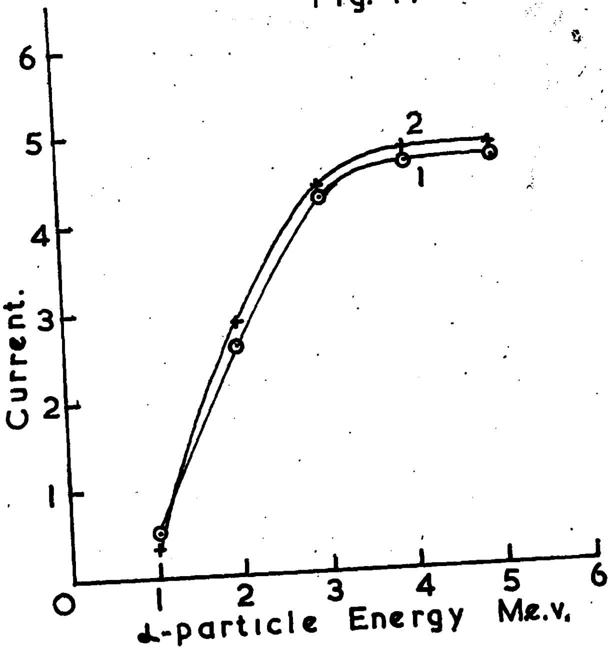
The results of the experiments, in which the relationship between the ion concentration and the proportion of the ion current which can be collected is shown in Table 6 and Fig.16. It will be noted that there is no apparent agreement between the curves obtained and the "Bragg Curve". Assuming that the ionization current collected was a direct function of the primary ionization produced by the alpha particles, it would appear that in a liquid the variation in specific ionization along the tracks did not obey the normal formula, as in the case of a gas. This is supported by the fact that corrections based on the variation in ion density would produce not less current from the portion of the tracks where the ion density was greatest. However, it will be noticed that there is apparently, at the extreme end of the tracks, a section where a considerable portion of the ions can be liberated. It would therefore appear that the ion current is not directly dependent on the primary ionization. If these results are compared with the theoretical curves for the ionization produced by means of high energy delta rays, some agreement is immediately apparent. This type of theory is supported by the fact that there is apparently no difference in current whether the collection field is applied at right angles or parallel to the direction of the incident alpha particles. Fig.17 shows the specific

FIG. 16.



Absorber thickness - Current Relationship, for Alpha Particles.

Fig. 17.



Curve I. Specific Ionization for Alpha Particles in Liquid.

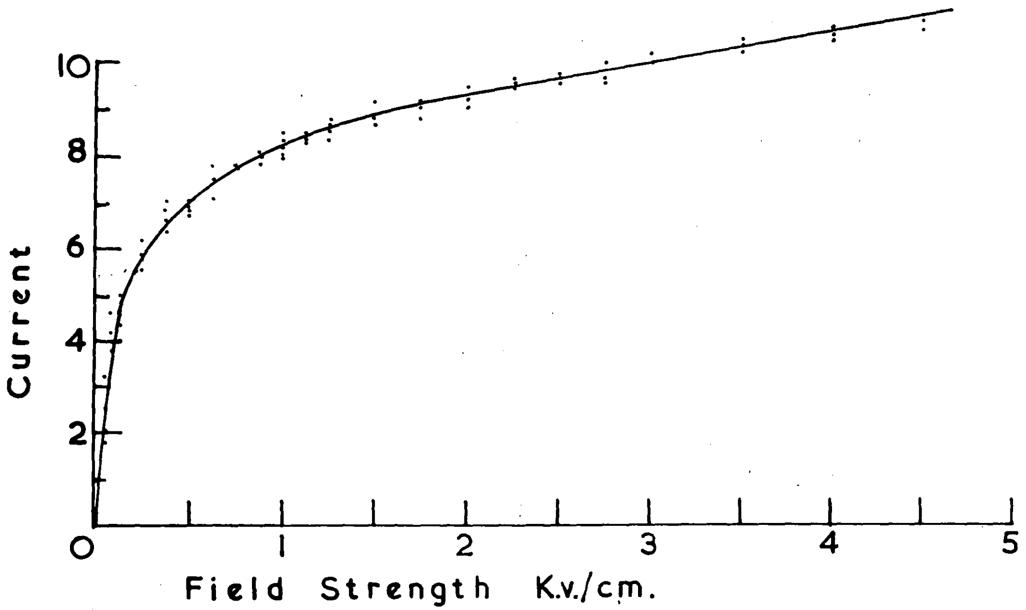
Curve 2. Ionization due to Delta Rays with more than I K.V. Energy.

ionization as measured in these experiments, plotted together with the ionization due to delta rays emitted with energy in excess of 1 K.V. per unit length of the track. It may be argued that even if the ionization current is solely due to these secondary delta rays, far higher currents should be observed. It has been shown that when beta particles are used to ionize a liquid, about 10% of the ions formed can be liberated. It might therefore be supposed that the same should be true in the case of these delta rays. However, account must be taken of the column of ionization formed by the alpha particle itself. Any ions formed by the delta rays in this volume will be subjected to the same probability of recombination as that of the primary ionization, and would therefore not be collected. In fact, it may well be the case that only the ions formed by the delta rays outside these columns can be collected. Some indication as to the size of the columns might be expected from these considerations, precise measurements are not possible because of the scattering of low energy electrons in the liquid. However, by estimating the minimum energy of the delta particles which contribute to the ion current, some idea of the diameter can be obtained. It appears likely that only delta rays having an energy of 1 K.V. contribute, giving a value of 0.05 u for the column diameter.

From the experiments using beta particles, the results are similar to those obtained from the alpha experiments (Table# 7 and Fig. 18 and 19). Unfortunately a mono-energetic beta source was not available, so no absorption measurements, comparable with those made in the former case, were possible. However, measurements using absorbers were made in connection with Copper 64 and Rhubidium 86. It will be seen that these results are very similar to those obtained using alpha particles. (Fig. 20 and 21). It is perhaps of interest that when a log. plot is made between the field strength and the absorber thickness, a linear relationship was found. It will be noticed that breaks occur when the 'end point' of one of the beta rays energies is reached. (Fig. 22 and 23). It is not suggested that this could be used to measure the maximum energies of beta spectra, but it does give some indication as to the behaviour of ions in a liquid. Fig.24 shows the corresponding logarithmetrical plots for alpha particles. It will be noted that again a linear relationship is obtained.

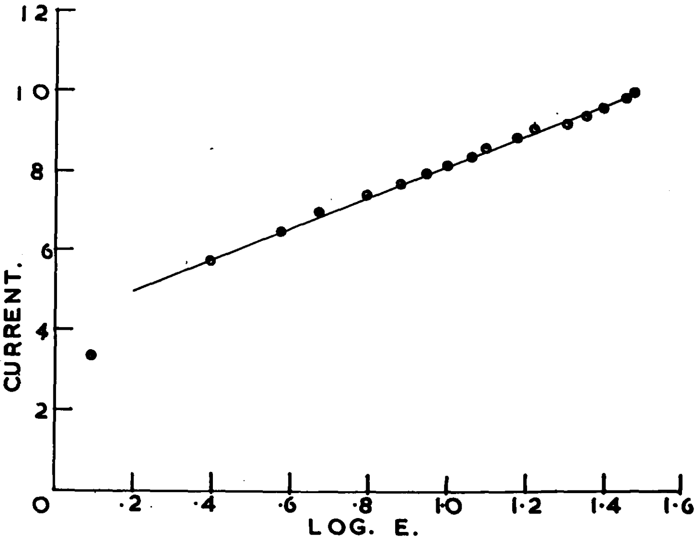
* * * * * * *

Fig. 18



Field Strenght - Current Relationship for Beta Particles.

Fig. 19.



Field Strength - Current Relationship for Beta Particles

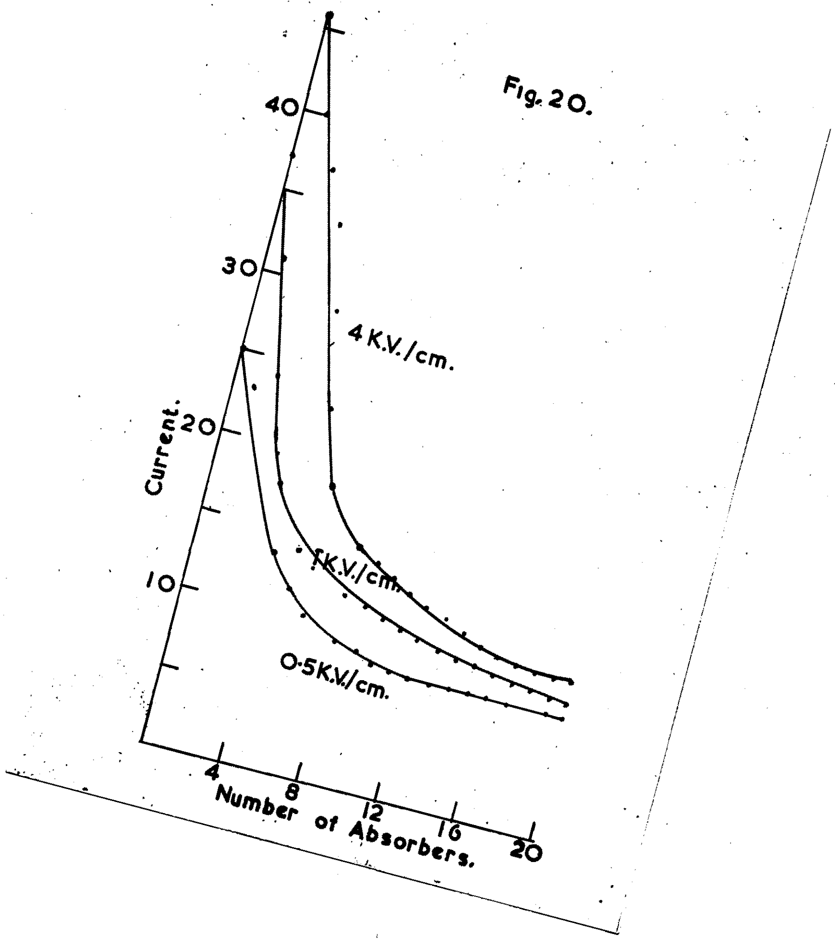
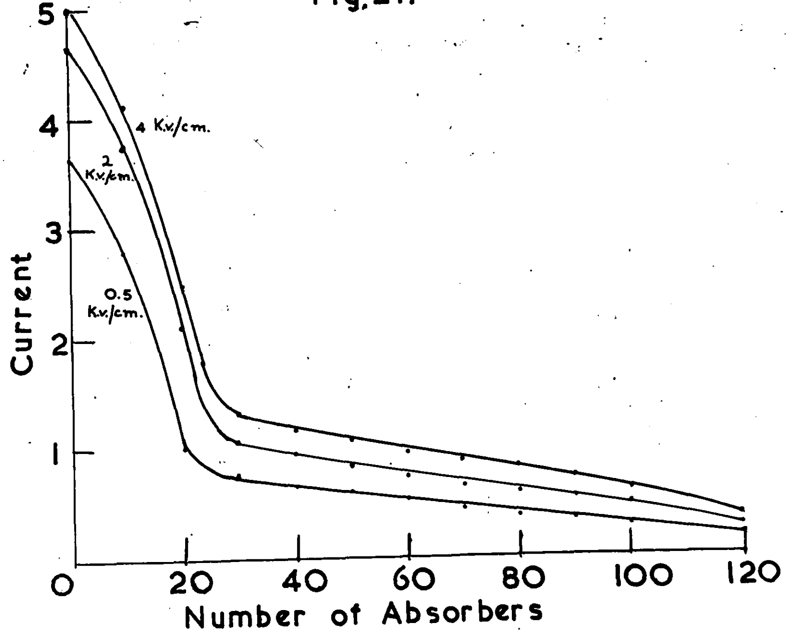


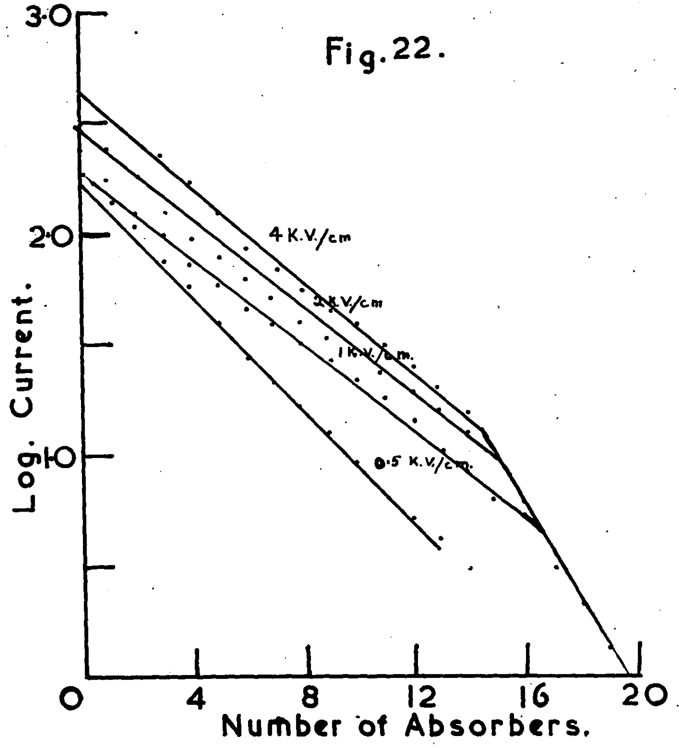
Fig. 20.

Field Strength - Current Relationship for Beta Particles from Copper 64.

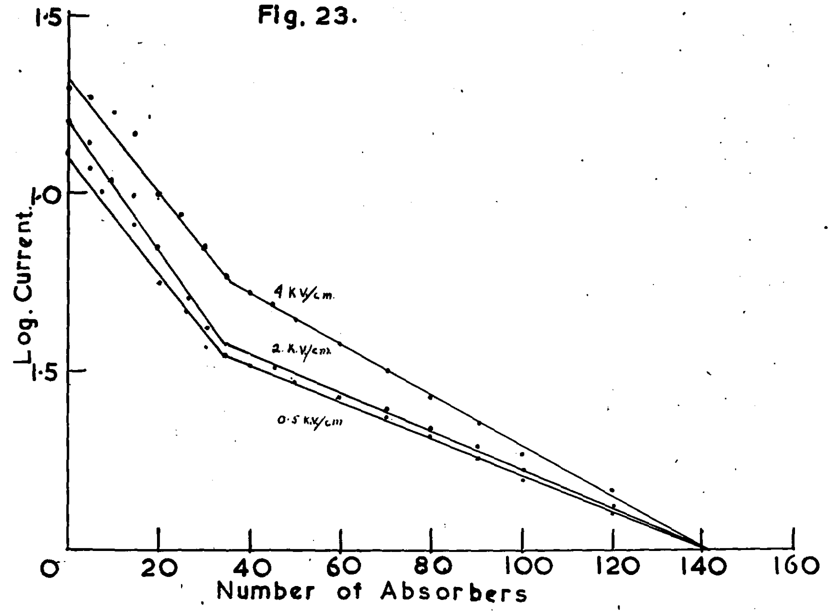
Fig. 21.



Field Strength - Current Relationship for Beta Particles from Rhubidium 86.

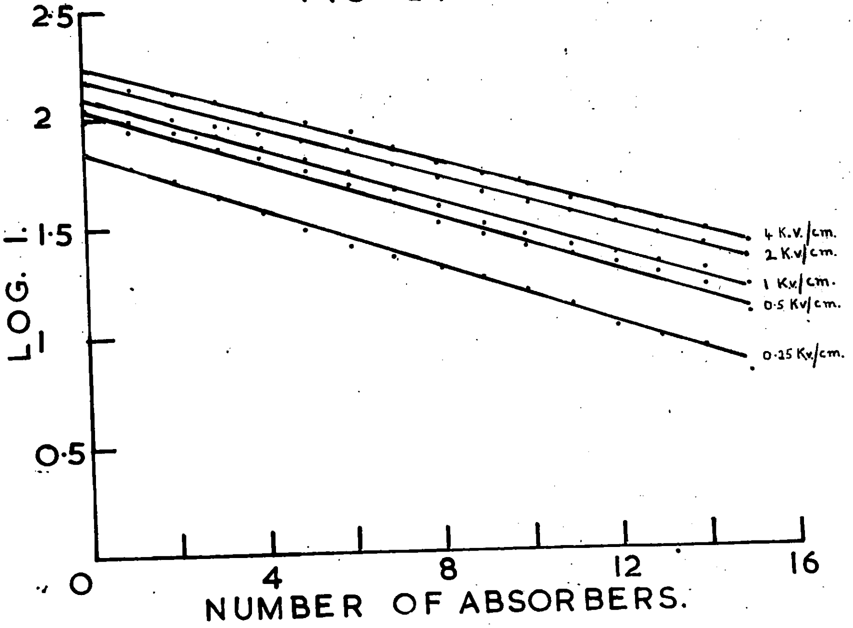


Absorber thickness - Current Relationship for Beta Particles from Copper 64. Break occurs at about 0.6 Mev.



Field Strength - Current Relationship for Beat particles from Rhubidium 86. Break occurs at about 0·7 M.e.v.

FIG 24



Field Strength - Current Relationshit for Alpha Particles

Table 4.

Current-Field Strength for Alpha Particles at at Right-angles to the Field.

Field Strength K.V./Cm.	Currents Amps. 10^{-11}.			
	a.	a.	b.	c.
0.25	2.60	2.85	2.2	2.1
0.5	5.40	5.35	5.5	5.4
0.75	7.65	7.55	7.65	7.5
1.0	7.95	8.4	8.0	7.8
1.5	8.70	8.65	9.0	8.7
2.0	9.60	9.7	9.7	9.5
2.5	9.80	10.0	10.0	9.8
3.0	10.60	10.45	10.45	10.3
3.5	10.80	10.75	10.9	10.7
4.0	11.2	11.3	11.3	11.0
4.5	11.6	11.5	11.7	11.4
5.0	12.0	11.9	12.0	11.8

Readings in columns "a" were taken using Hexane and a quartz window. Equivalent ionization current, using a gas filled chamber, was 8.5 10^{-8} amps. Column "b" was obtained using Apiazon oil in vacuo without a window. Column "c" was with Apiazon Oil and quartz window.

Table 5.

Current-Field Strength for Alpha Particles Parallel to the Field.

Field Strength. K.V./Cm.	Current. Amps 10^{-11}.		
0.125	0.1	0.05	0.1
0.25	1.7	1.3	1.55
0.5	2.35	2.30	2.30
0.75	2.60	2.60	2.58
1.0	2.70	2.70	2.70
1.25	2.85	2.85	2.85
1.5	3.02	3.00	2.95
2.0	3.15	3.15	3.10
2.5	3.40	3.40	3.40
3.0	3.55	3.55	3.53
3.5	3.70	3.65	3.65
4.0	3.75	3.70	3.72
4.5	3.85	3.80	3.82
5.0	3.90	3.90	3.90

Readings taken using the cylindrical ionization chamber and Po source. Liquid used was Hexane. Equivalent ionization current using a gas filled chamber was 2.63. 10^{-8} amps.

Table 6.

Specific Ionization for Alpha Particles in Hexane.

No.of Absorbers.	Current Amps 10^{-11}.					
	0.25	0.5	1	2	4	Field Strength K.V/Cm.
0	0.7	1.0	1.1	1.2	1.5	
1	0.6	0.85	1.0	1.1	1.4	
2	0.5	0.8	0.9	1.0	1.3	
3	0.45	0.7	0.8	0.9	1.25	
4	0.3	0.6	0.7	0.85	1.1	
5	0.3	0.55	0.65	0.75	0.9	
6	0.25	0.5	0.55	0.7	0.8	
7	0.22	0.4	0.45	0.6	0.7	
8	0.20	0.3	0.35	0.5	0.6	
9	0.18	0.28	0.30	0.45	0.5	
10	0.15	0.25	0.27	0.38	0.45	
11	0.13	0.22	0.25	0.35	0.4	
12	0.11	0.20	0.22	0.30	0.35	
13	0.09	0.18	0.20	0.27	0.3	
14	0.08	0.14	0.18	0.24	0.27	
15	0.06	0.11	0.15	0.20	0.24	
16	0	0	0	0	0	

Readings taken using the cylindrical ionization chamber and Po source. Liquid used was Hexane. Absorbers were aluminium leaf, having an air equivalent of 0.8 mm.

Table 7.

Current-Field Strength for Beta Particles.

Field Strength K.V./Cms.	Currents in arbitrary Units.					
	Th.Source.					Cu 64.
0.25	58	55	50	47.5	44	40
0.5	58	62.5	55	55	56	53.5
0.75	70	66.5	62.5	62.5	69	65
1.0	76	67	67.5	70	69	71.5
1.25	90	70	75	75	75	-
1.5	77	77	77.5	77.5	78	80.0
2.0	81.7	83.3	80.0	82.5	80	83.5
2.5	87.5	87.5	87.5	85	87	90.0
3.0	91.7	91.7	87.5	87.5	87	90.0
3.5	91.7	91.7	90.0	90.0	90	91.5
4.0	91.7	91.7	90.0	92.5	95	96.5
4.5	95.5	95.5	95.0	95.0	95	96.5
5.0	95.5	95.5	97.5	97.5	97	96.5
5.5	95.5	100.0	100.0	100.0	100	100.0
6.0	100.0	100.0	102.5	100.0	100	100.0

Readings taken using large cylindrical ionization chamber. Liquid used was Hexane.

Table 8.

Current to Absorber Thickness for Beta from Cu 64.

No. of Absorbers.	Current Amps 10^{-10}.					
	0.25	0.5	1	2	4	Field Strength K.V./Cm.
0	2.5	3.1	3.9	4.4	5.3	
1	2.0	2.3	2.7	3.1	4.0	
2	1.7	1.9	2.45	2.65	3.7	
3	1.4	1.5	1.95	2.0	3.4	
4	1.2	1.3	1.6	1.7	2.9	
5	1.0	1.1	1.4	1.5	2.3	
6	0.95	1.0	1.3	1.4	1.8	
7	0.92	0.95	1.24	1.32	1.6	
8	0.86	0.90	1.20	1.25	1.5	
9	0.84	0.87	1.16	1.20	1.45	
10	0.81	0.83	1.12	1.15	1.40	
11	0.79	0.81	1.08	1.10	1.30	
12	0.78	0.80	1.04	1.05	1.25	
13	0.78	0.79	1.00	1.02	1.20	
14	0.78	0.79	0.96	1.00	1.15	
15	0.77	0.78	0.94	0.96	1.10	
16	0.77	0.78	0.92	0.94	1.05	
17	0.77	0.77	0.90	0.91	1.03	
18	0.77	0.77	0.89	0.90	1.02	
19	0.76	0.77	0.88	0.89	1.01	
20	0.76	0.77	0.87	0.88	1.00	

Readings taken using the large cylindrical ionization chamber. Liquid used was Hexane.
Absorbers were Aluminium foil 0.002" thick.

Table 9.

Current-Absorber Thickness for Beta from Rb 86.

No. of Absorbers.	Current Amps 10^{-10} .				
	0.5	1	2	4	Field Strength K.V./Cm.
0	1.4	1.75	1.9	2.1	
5	1.3	1.50	1.75	2.0	
10	1.15	1.30	1.60	1.8	
15	0.90	1.10	1.45	1.6	
20	0.60	0.65	0.80	1.1	
25	0.50	0.55	0.70	1.0	
30	0.47	0.50	0.65	0.8	
35	0.45	0.47	0.60	0.7	
40	0.43	0.45	0.55	0.65	
45	0.43	0.43	0.50	0.60	
50	0.42	0.42	0.47	0.55	
60	0.40	0.40	0.45	0.50	
70	0.36	0.38	0.40	0.43	
80	0.31	0.33	0.35	0.38	
90	0.28	0.29	0.30	0.34	
100	0.25	0.26	0.27	0.30	
120	0.23	0.24	0.25	0.27	

Readings taken using the large cylindrical ionization chamber. Liquid used was Hexane. Absorbers were Aluminium foil 0.002" thick.

Scintillations from Thin Films.

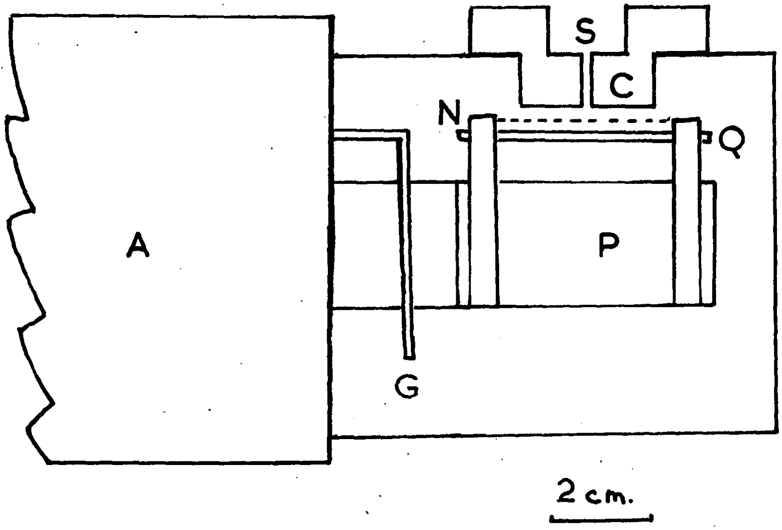
For reasons previously stated, it was thought that photon emission might be associated with the ionization produced by alpha particles in liquids, in particular when an alpha particle passes through water, since most of the chemical reactions which have been studied take place in aqueous solutions. If ionization due to alpha particles is produced in a large volume of water, such an emission is not observed. • This may well be due to the absorption of the radiation by the bulk of the water. This process would be extremely likely if a resonance phenomenon was responsible for the production of the radiation. It was therefore decided to study the effects produced by irradiating with alpha particles a water film, whose thickness was less than the range of the alpha particles. The production of such films proved to be very difficult, moreover, the detection of the radiated quanta presented serious problems. The latter arose from considerations as to the most likely frequencies to be involved. If the effect is produced by a resonance phenomenon, then the frequencies of the emitted quanta will presumably coincide with the absorption spectrum of water. Therefore they will occur in the extreme ultra violet or in the infra-red spectral regions. Unfortunately the photomultipliers available, were not sensitive in these regions. Experiments were performed in which a zinc sulphide screen was used to transform any quanta emitted in the ultra-violet regions to a suitable frequency for detection. However, owing to solid angle and efficiency considerations, it was not found possible to make quantative measurements. In view of these difficulties, it was decided to investigate the effect when films of solids were irradiated with alpha particles.

The experimental arrangement used for these investigations was as shown in Fig.25.

The film under investigation was placed about 1 cm. from a strong Polonium source, which was carefully collimated. Immediately below the film was a 1 mm. thick quartz slide, placed so as to prevent the glass of the 931A photomultiplier from being bombarded by the alpha particles. The multiplier was mounted in a perspex holder, fixed directly on to a head amplifier. The signals were then amplified in a type 1008 amplifier, designed by T.R.E. Malvern, and then fed through a discriminator unit to a Scalar Unit Type 1037.

Due to the low efficiency of the films for producing quanta, it was found necessary to take special precautions to reduce the noise level to a minimum. The 931A photomultipliers were specially selected, and a drying agent was enclosed with the tube to prevent the condensation of moisture on the valve base. As in the case of the liquid ionization chambers, leakage currents across the perspex

Fig.25.



Experimental Arrangement for the Investigation of Thin Films.

A	Head amplifier	G	Guard ring	C	Collimator
Q	Quartz slide	S	Po. source	N	Nylon film
P	931A Photomultiplier tube.				

valve holder were prevented by careful cleaning with chloroform. It was found that in some cases, considerable improvement of the noise level could be effected by cleaning the base of the multiplier itself. This was accomplished by placing the multiplier in a vessel which could be evacuated and leaving it under reduced pressure for a period of 24 hours. The multipliers were operated under their optimum conditions with regard to signal to noise ratio. It was found that the best results could be obtained when between 60 and 80 volts were applied across each stage, depending on the particular tube in use. A guard ring was used to obviate the possibility of collecting ions on the pins on the base of the photo-multiplier, so giving rise to spurious counts.

The circuit of the head amplifier was similar to that employed in the standard high frequency head amplifier designed for use with the 1008 amplifier. (Fig.26). Care was taken to ensure that the lead between the input and the photo-multiplier was as short as possible in order to reduce the input capacity to a minimum. The input resistance was reduced from 1000 Meg ohms to 10 Meg ohms, so as to shorten the input time constant, so reducing the effective width of the signals. In this manner it was found possible to reduce the noise from the first stage of the amplifier, and also to prevent the integration of the multiplier noise, hence improving the sensitivity of the system. It was

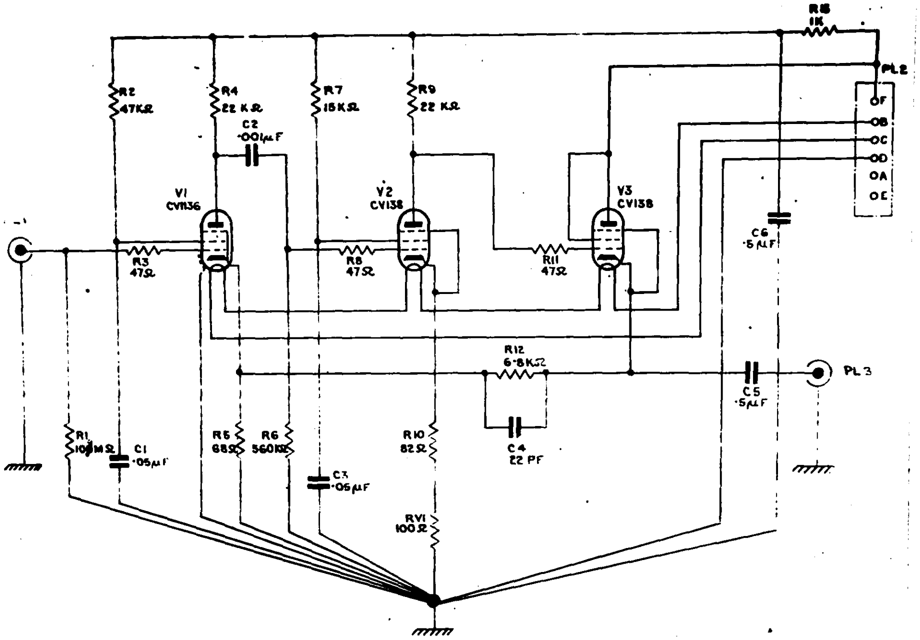


Fig. 26. Head Amplifier Circuit

found that such a head amplifier gave an improvement of up to 5 times the ratio signal to noise ratio, as compared to a simple cathode follower. This is due to the higher ratio between the input and output impedance obtainable from a ring of three valves compared with a single cathode follower. This is of particular importance when high frequency pulses, such as are produced in a photo-multiplier, are concerned. Under these conditions the normally accepted formulae for the operation of a cathode follower does not apply, since the stray capacities between the cathode and ground become of major importance. The valve is therefore required to pass comparatively large currents during the period for which the grid volts are rising, with a resultant drop in the grid impedance. (See Appendix 1).

The quartz slide served two purposes. It was found that if the glass envelopes of the photo-multipliers were bombarded with alpha particles, some scintillations were observed. The number could be considerably reduced by protecting the glass by means of a quartz slide. Also the majority of the films investigated were made by evaporating suitable materials on to quartz.

The films used could be divided into two types. Those which were self supporting and those deposited on to quartz. In the former category were Nylon, Collodion, Glass, Mica, etc. These were made in the usual manner, and in the case of Nylon, the thickness was determined by their stopping power for alpha particles. This was carried out using a Polonum source and an air ionization chamber in the usual way. The following technique was employed to obtain extremely thin Nylon films. A saturated solution of American Nylon was made by heating a mixture of Nylon chips and Isobutyl Alcohol in a water bath for three days. The undissolved Nylon was then filtered off and the clear solution used. It was found that on standing for periods of about 24 hours, that the solution became cloudy and tended to solidify. If the mixture was gently warmed in a water bath a few minutes, it became clear and could be used for the manufacture of films.

A small wire frame, about 5 cms. long and 3 cms. wide, on which the film was to be supported, was placed at an angle of approximately 10 degrees to the vertical, in a vessel containing clean water. The water level was so arranged that the top of the frame was about 0.5 cms. below the surface. The surface was swept clear of dust particles, by means of two waxed rods, in a similar manner to that employed in a Langmuir Trough. Two small drops of the nylon solution were then placed on the water surface by means of a pipette. After a period of 5 minutes the film was carefully cut away from the sides of the vessel, using a hot copper wire. The water level was then slowly lowered until the film just rested on the upper edge of the wire frame. A cut was made about ¼" from the frame and parallel to it. Two more cuts were made at right angles to the first, so as to produce an area of film about ¼" wider than the frame. The water was then slowly syphoned out of the vessel allowing the film to settle on to the frame. In this manner it was found possible to produce films having a thickness equivalent to 0.24 mm. of air.

The second class of films, which included metals, and ionic salts, were deposited on quartz slides which had been carefully cleaned. Wherever possible the materials were evaporated on to the slide in vacuo. This was possible in the case of all the metals, and some of the other materials. In the other cases, with the exception of carbon, the films were prepared by precipitation from dilute solutions in alcohol, or where this was not feasible, water. However, in these cases it was found impossible to produce films of constant thickness.

Experiments.

In all cases, irrespective of the material under investigation, the experimental procedure was the same. Because of the low efficiency of the films the background was measured immediately before and after each reading. This was done in two ways. First the film was removed and the background measured with a clean quartz slide in position. In the cases where the films were supported on quartz, the slide supporting the film was inverted. This precaution was taken so as to eliminate the possibility of the quartz becoming contaminated with grease or other materials during the preparation of the films.

In the case of Nylon, experiments were performed to determine the effect of thickness on this phenomenon. Nylon was chosen purely for the ease with which thin films of constant thickness could be obtained and not for any other property. A series of nylon films having a thickness equivalent to 0.48 mm. of air were used for this purpose. By bombarding various numbers of films under the conditions previously described, it was possible to determine the relationship between the number of counts recorded and the thickness. In a similar manner the effect of thickness on the pulse height distribution was investigated. (See Appendix 2). In order to determine the probability of the quanta being absorbed in nylon, specially thin films prepared in the manner previously described were used.

Experiments of a similar type were carried out using thin liquid films. The experimental arrangement was the same as in the previous experiments, with the exception that between the quartz slide and the photo-multiplier a zinc sulphide screen was placed. As previously, control experiments were carried out to ascertain the random background counts. Unfortunately it proved impossible to measure the thickness of the liquid films under investigation, and due to the difficulties discussed earlier, quantitative results were not possible. However, some evidence was obtained that quantum emission accompanied ionization produced by alpha particles.

When nylon chips were subjected to irradiation with ultra-violet light, a green fluorescence was observed. Similar effects were noted with some of the other materials used. Experiments were therefore undertaken in which an attempt was made to detect the quantum emission from large crystals of the materials. In the case of all the materials used, no quanta were observed when the thickness of the material used became large compared with the range of an alpha particle. Owing to experimental difficulties, it was not possible to observe the surface of the material which was under alpha bombardment, so that any quanta emitted from this surface could not be detected.

Similar experiments were carried out in which both thin and thick samples were irradiated with beta and gamma ray. No quantum emission was recorded in any of these experiments.

In order to obtain a standard for purposes of comparison, the experiments were repeated using a zinc sulphide screen.

Results.

The results of the experiments were very much as were expected. In all cases when thin films were used, quantum emission was detected under alpha bombardment. Also, the radiations were very highly absorbed by the material in which they were produced. The material used in these investigations included:

Sulphur.
Iodine.
Carbon.
Aluminium.
Copper,
Tungsten.
Iron.
Nickel Silphate.
Sodium Chromate.
Stannous Chloride.
Mercuric Iodide.
Nylon.
Glass.
Mica.
Collodion.

The relative efficiencies of the various materials can be seen from Table 10, and a typical pulse size distribution is given in Fig. ~~25~~ ~~and~~ 27. In none of the cases were the signals produced by the photo-multiplier greater than 4 times the noise level, and it will be seen that efficiencies of the various films are of the same order of magnitude. It was not practicable to measure the thickness of the films which were deposited on quartz slides, so that an exact comparison was not possible. However, this may be of secondary importance in view of the results obtained from the nylon experiments. The apparent reduction in efficiency observed in the case of ionic salts is probably due to imperfections in the films used. However, since it was not found possible to measure the frequency of the quanta emitted, this apparent drop may be due to the selectivity of the photo-multiplier.

In the case of liquids, far less conclusive results were obtained. However, when water and Hexane were irradiated,

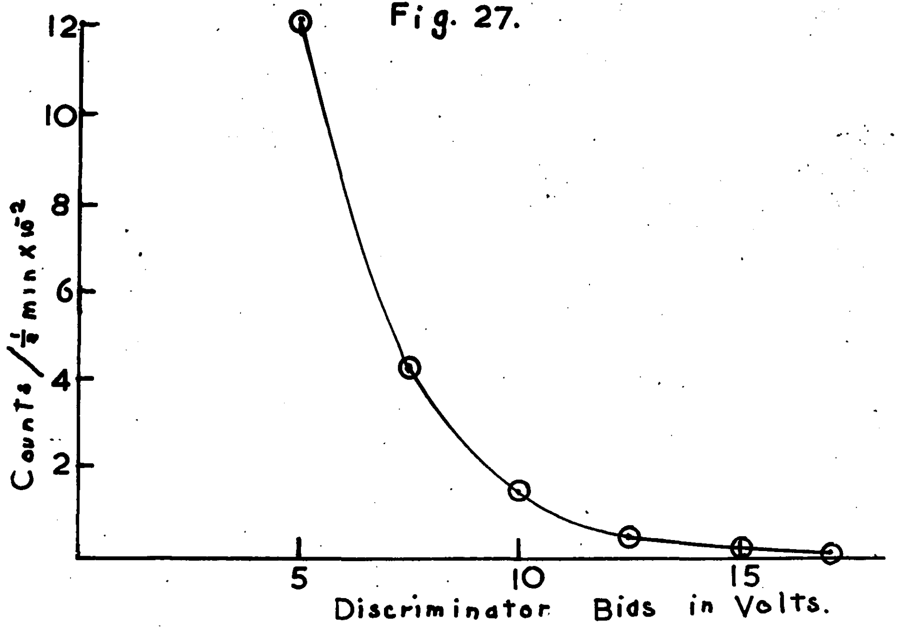


Fig. 27.

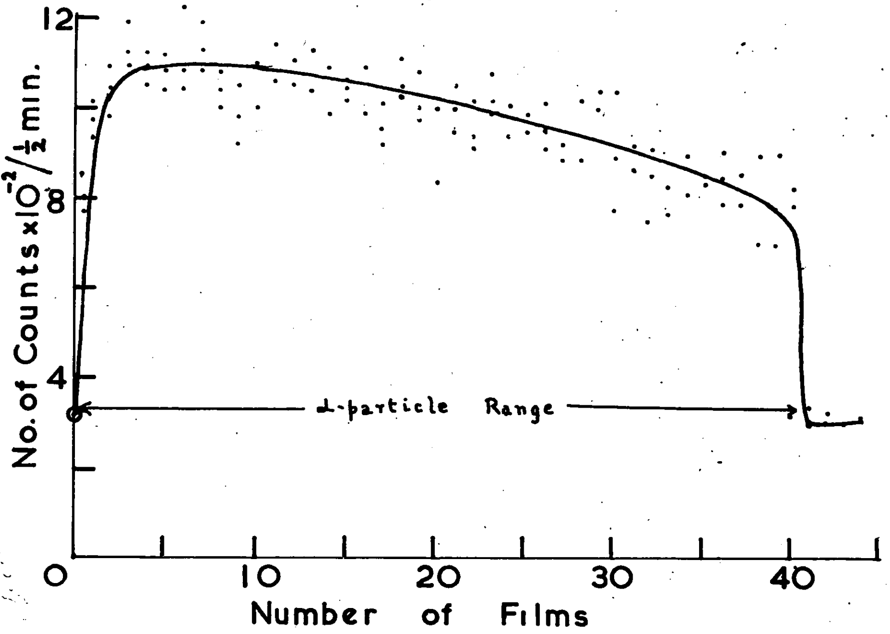
Pulse Height Distribution for a Single Nylon Film.

some indication of a similar effect was obtained. It was therefore concluded that quantum emission is associated with the dense ionization produced by alpha particles.

The results of the experiments, which were performed with nylon to investigate the effect of thickness, are given in Fig.28. It will be observed that the number of pulses is to a large extent independent of the thickness, until a value greater than the alpha range is reached. From the results obtained, using the very thin films, it appears that there is a fifty per cent probability of the quanta being absorbed in travelling about 200 molecular distances. However, as in the case of the specific ionization produced by an alpha particle in a liquid, it appears impossible to correlate the results with the normal "Bragg Curve". It will be noted that the number of quanta emitted per unit length of track decreases with the velocity of the alpha particle. If the effect was directly associated with the primary ionization, the reverse would be expected. It would therefore appear that some consideration, other than the ion density must be taken into account.

When an alpha particle produces ionization, considerably more energy is expended for each ion pair produced than would be anticipated, if the whole energy was used for this purpose. It appears reasonable to assume that this excess energy is used in producing either

Fig 28.



The Effect of Film Thickness on Nylon Film Efficiency.

excited molecules or excited ions. The latter may be produced by ionization in which other than the outermost electron participate. Perhaps the best known example of this type of phenomenon, is the production of "K" or "L" X-rays in metals. The penetration of such "K" radiation in the parent material, is of the same order of magnitude as the range of alpha particle from Polonium in the same metal. From energy considerations, the probability of producing such X-rays falls rapidly with the loss of energy of the ionizing particles. As the thickness of the metal films used in these experiments were not measured, the results obtained would not be at variance with the production of such radiation. On the other hand, when a metallic film comes in contact with air, there is a probability that an oxide layer will be formed. It is therefore possible that the entire effect in the case of metals is due to this layer. However, in the case of Tungsten, the probability of such a formation is greatly reduced, but results, using this material, were similar to those obtained when other metals were used.

In the case of non metallic materials, an explanation based on a similar theory is possible. In fact, for the non metallic elements, the theory offers the most plausible explanation. However, it should be noted that the radiations may not be "K" or "L" X-rays, but other characteristic radiations of the material concerned. In view of the fact, the effect was only detected under alpha particle bombardment, it is reasonable to assume that the phenomenon may be due to the recombination of the primary ions. When two excited ions recombine, there is a probability of quanta emission taking place. The number of excited ions produced in any section of an alpha track will be proportional to the number of and energy delta rays produced. Fig.29 gives the ionization produced by such delta ray per unit length of alpha track. It will be seen that the form of the curve is in good agreement with the number of quanta observed. With Nylon, there is a fifty per cent probability of the quanta being absorbed in travelling 200 molecular distances in Nylon. This probability is considerably greater than would be the case if the radiation took the form of "K" X-rays. It would appear therefore that in this case, at least, lower frequency radiation is concerned. It is unlikely that the quantum emission is caused by the delta rays themselves, since radiations are not observed when the films are bombarded with beta particles.

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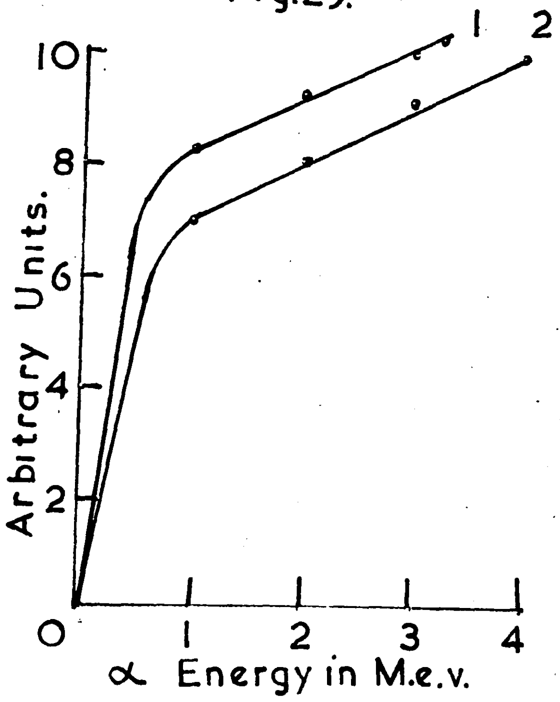
Fig.29.



Curve I.Efficiency - Alpha Energy.

Curve 2. Energy in Delta Rays.

Table 10.

Efficiencies of Thin Films as Scintillators.

Material.	Counts / $\frac{1}{2}$ min.			
	5	$7\frac{1}{2}$	10	Discriminator bias.
Sulphur	4800	450	125	
Iodine	3000	1200	700	
Carbon	1600	-	300	
Nickel Sulphate	2400	160	-	
Sodium Chromate	600	200	100	
Stanneous Chloride	620	50	-	
Mercuric Iodide	600	200	100	
Aluminium	9100	1600	300	
Copper	1400	250	-	
Tungsten	600	300	240	
Iron	900	225	120	
Collodion	800	200	110	
Mica	900	200	-	
Styrafoil	2020	200	62	
Nylon	1800	420	200	
Zinc Sulphide	6.10	6.10	6.10	

All readings taken using the apparatus shown in Fig. 25.

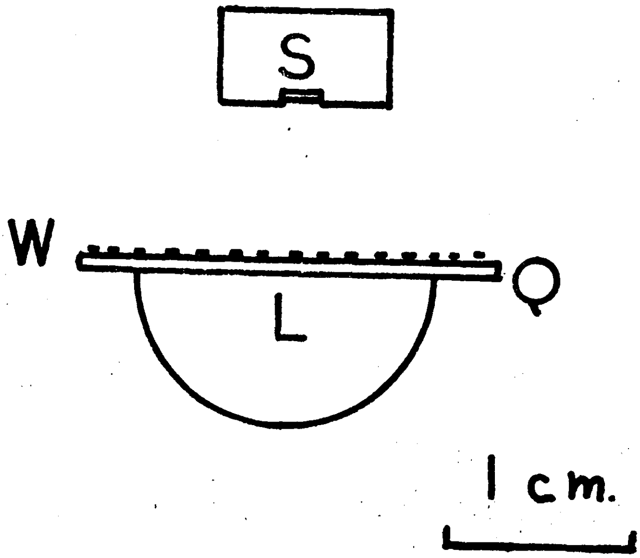
Chemical Experiments.

These experiments were designed in an attempt to correlate the results obtained in the purely physical experiments with those obtained by the chemists. From these, it appeared that either the ions produced by the passage of alpha particles, or some product, must be capable of existing in liquids for considerable periods. From arguments based on the probable concentrations of the ions and their products assuming that these are formed by direct action, it would appear unlikely that this would be the case. However, in view of the results obtained from the experiments performed with thin films, an explanation, based on the emission of quanta, would appear to be possible. It was therefore decided to carry out experiments in which the pure solvent was separated from the material in which the chemical change was to take place.

In these experiments the solvent chosen was water. A thin layer of water was formed on a quartz slide, 1 mm. thick, which covered a suitable chemical solution. The water film was then exposed to bombardment from a strong alpha source in such a manner that no alpha particles could enter the solution. After a period of time, the solution was tested for any chemical change. In this manner, it was hoped that the chemical action resulting from ionization due to alpha particles, was at least partly due to associated quanta emission.

The experimental arrangement was as shown in Fig.30. A 20 milli Curie Polonium source was placed about 0.5 cm. above the quartz slide supporting the water film. Considerable difficulty was experienced in obtaining a film whose thickness was less than the range of the alpha particles. As in the case of the experiments with thin films, the slide was carefully cleaned before the experiments. The slide was first boiled in carbon tetrachloride, in order to remove any grease. It was then washed with distilled water containing a small quantity of a suitable wetting agent. After this, it was boiled in a solution of chromic acid for 10 minutes, and again washed with distilled water. The slide was then washed with absolute alcohol and allowed to dry in a dust free atmosphere. The surface was finally cleaned by brushing with the discharge from a leak tester. In this manner it was found possible to produce a surface on which a thin layer of water could be formed. Unfortunately, it was not possible to measure the thickness of such layers, but from the results of the experiments on the emission of quanta from thin films, this may be of secondary importance. The slide so prepared, formed the top to a small container which held about 1.5 ccs. of solution of the chemical under investigation. Again this chemical had to be carefully purified as the yields of the experiments was extremely

Fig. 30.



The Experimental Arrangement used for the Chemical Experiments.

S Po source. W Water Film. Q Quartz plate

L Solution

small. Because of this, with each exposure a control experiment was carried out. This consisted of a similar quantity of the solution being placed in a similar container and kept under the same conditions as the sample being used in the experiment, with the exception that it was not subject to bombardment by alpha particles. In this manner it was found possible to eliminate the chemical changes which take place not associated with alpha ionization. This was especially important in the experiments where ferrous sulphate was used, as there is always some oxidation to ferric sulphate. Besides these control experiments, it was also necessary to show that the reactions were not caused by the irradiation of the quartz plate or by contamination present. For this purpose, a sample of the solution was placed under a clean dry slide and irradiated with alpha particles. The solution was then tested for chemical change as in the case when a water layer was present.

Because of the small yields obtained, the estimations were carried out using optical method. In the case of Ferrous Sulphate and Potassium Iodide colourmetric methods were employed. For the experiments in which Chloracetic Acid was used, the yield was determined by the scattering of light, due to the precipitate formed by the addition of silver nitrate to the solution.(1) For this purpose a 'Unicam' made by the Cambridge Instrument Company was

(1) See Bibliography. Estimation of Chlorine

employed in some of the experiments. In latter experiments more suitable apparatus was designed for this purpose. The arrangement used was as shown in Fig.31. One of the advantages for this design over the 'Unicam' is that greater path lengths through the liquid are obtainable for small volumes of solution. This is of the utmost importance when, as in these experiments, the yield is independent of the quantity of solution employed. It will be noted that the samples are contained in two thin glass tubes, through which a collimated beam of light can be passed. This beam, is interrupted by means of a revolving disc, which is slotted in such a manner that the light passes first through one tube and then through the other. The two beams are then reflected by means of two surface silvered mirrors, so that they impinge on the same part of the photo sensitive surface of a gas filled photo-cell. The output from the cell is then fed through an amplifier of conventional design, and displayed on a cathode ray osciliscope. Fig.32. In this manner it is possible to eliminate any effect due to the change of the amplifier gain, and to any change in the efficiency of the photo-cell. Also it is possible to obtain a direct comparison between an unknown solution and one of known concentration. Using this apparatus it was found possible to measure concentrations of free Iodine of the order of 10^{-6} Normal, which were liberated from Sodium Iodide when starch was used as an indicator.

Fig. 31.

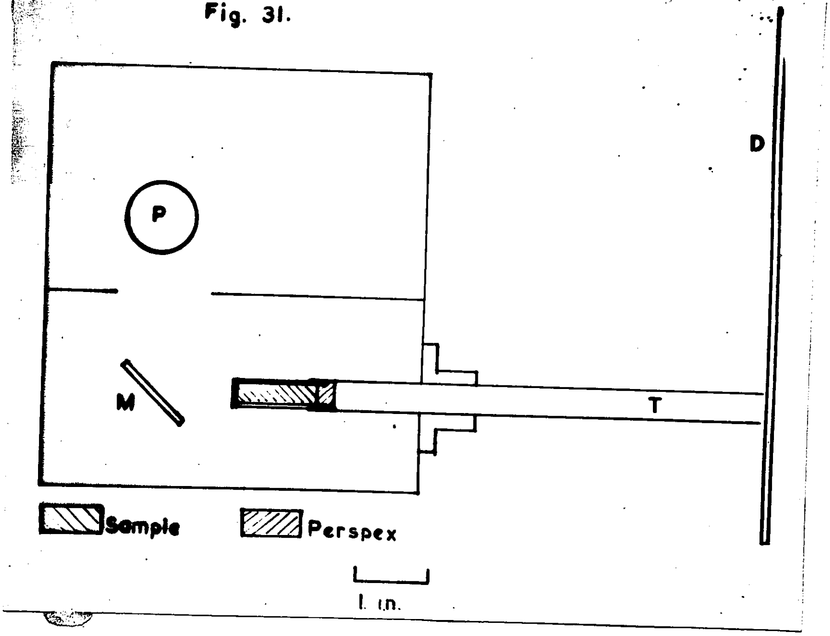


Photo-comparator for Measuring Chemical Yields.

D Revolving slotted disc M Mirror

S Sample in glass tube P Photo cell

T Collimating tube Second similar tube arrangement directly under that shown.

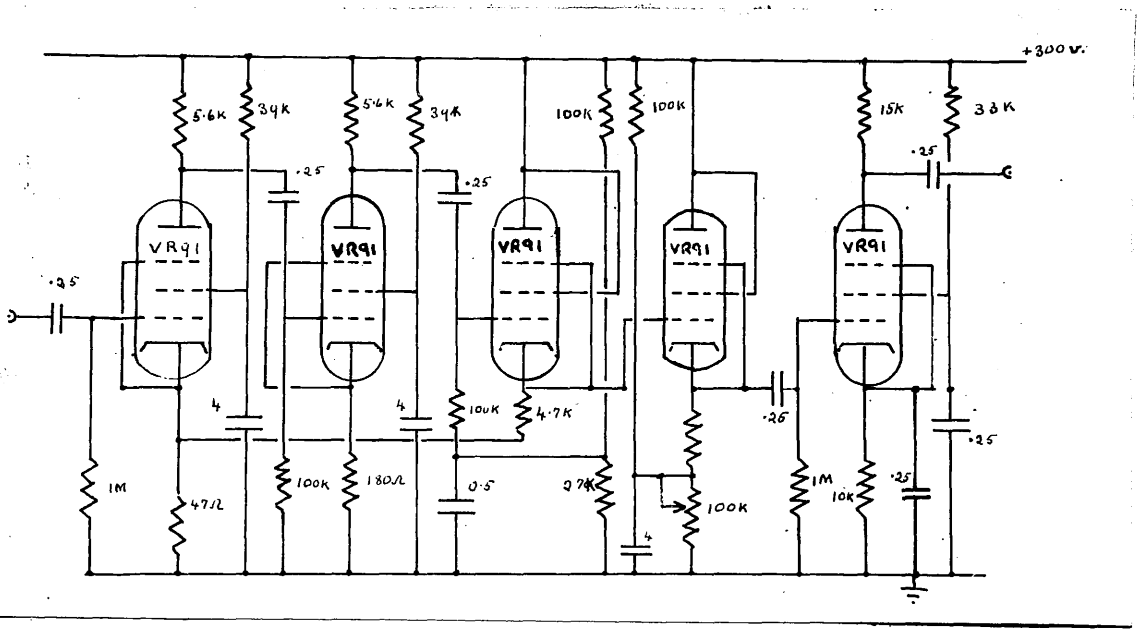


Fig 32. Circuit used with the Photo-comparator.

Experiments.

Three materials were used in the experiments. Ferrous Sulphate, Chloracetic Acid, and Potassium Iodide. The salt used in the first case was Ferrous Ammonium Sulphate, as it appeared to be more stable than Ferrous Sulphate. A commercially pure sample was taken, and so as to ensure the presence of as small an amount of the Ferric salt as possible, reduced by nascent hydrogen made by adding pure sulphuric acid and a little zinc. After reduction, the clear solution was filtered off, and made approximately normal by adding sulphuric acid. The two containers were then filled with solution, and covered by quartz plates. A thin film of water was then formed on one of the quartz slides and the samples were subjected to alpha bombardment for 24 hours. After this period, both samples were tested for the presence of Ferric Iron, by the addition of Potassium Thiocyanate. Conclusive results were not obtained owing to the production of Ferric Iron, due to the normal processes of oxidation. However, larger yields appeared to be obtained from the sample which was subjected to irradiation. As has been previously stated, a blank experiment was performed in which the quartz plate was irradiated, without the presence of a water film. In this case no yield in excess of the control experiment was recorded.

More satisfactory results were obtained when Chloracetic Acid was used. The acid was purified before the start of the experiments by fractional distillation, followed by distillation in vacuo. In this manner it was found possible to obtain samples which produced no precipitate on the addition of Silver Nitrate solution. As in the previous case, two solutions of the acid were taken and one subjected to irradiation. After a period of eleven days, the samples were tested for the presence of free chloride ions. This was done by adding to each solution 2 ccs. of 0.002 normal silver nitrate solution, which was decinormal in Nitric Acid. The solutions were then allowed to stand for 30 minutes in order to allow the precipitates to develop. Unfortunately it was found that the usual procedure of warming in a water bath to 40° Centigrade for 20 minutes was impracticable, because of the liberation of small quantities of chlorine ion from the unaffected Chloracetic Acid.(1) The samples were then compared either in a 'Unicam' or in the apparatus previously described. In this manner it was found possible to detect yields in the order of 10^{-5} normal. In practise the yields obtained from the irradiated solution were about 2×10^{-4} normal, whilst those from the control experiments about 4×10^{-5} normal. As in the case of Ferrous Sulphate,

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(1) See Bibliography. Estimation of Chlorine and Iodine.

the experiment was repeated, using a dry quartz plate. When this was done, yields comparable to the control experiment were obtained, i.e. 4×10^{-5} normal. In all cases the yields were measured immediately after the precipitates had developed, as it was found that if they were allowed to stand for periods of more than an hour, varying amounts of increased yields were recorded.

So as to obtain a direct comparison with the yields formed in this manner, with those obtained by the usual method, experiments were performed in which the alpha particles from the same source, were allowed to enter the Chloracetic Acid solution. In these experiments the arrangement was the same as before, with the exception that the quartz slide and water film were omitted. It was found that with exposures of 12 hours, yields of chloride ion of 4×10^{-4} normal were obtained. This gave a method of calibrating the effective strength of the source, since previous experimenters had determined the yield of free chloride ion to be one per ion formed in the solution by the alpha particles. It was therefore possible to obtain some idea as to whether the chemical action due to the quanta emission was of the same order of magnitude as that created by the passage of alpha particles through the solution itself.

Similar experiments were performed substituting sodium iodide for the chloracetic acid. It was noted that when solutions of sodium iodide were irradiated with ultra-violet light, then large yields of free iodine were produced. These could be readily detected with the use of starch. (Turner 1930). It was therefore decided that this would provide a suitable material for this type of experiment. The experimental procedure was the same as in the previous cases. This time, owing to the greater sensitivity of the method of estimation, periods of irradiation of 24 hours were used. In this time, yields of 10^{-5} normal were obtained from the radiated samples, whilst no free iodine was detected from the control experiments. As in the previous case, an experiment was carried out in which a clean, dry quartz plate was subjected to alpha bombardment over a sample of the iodide solution. Again no yield was detectable. Experiments were also performed in which the alpha particles were allowed to impinge directly on the iodide solution. As in the case of the Chloracetic Acid, larger yields were obtained than when a water film and quartz slide were used.

Some evidence of the frequency of the radiations emitted from the water film was obtained in the experiments in which the Chloracetic Acid was employed. From experiments using an ultra-violet lamp as a source of radiation, it was found that the effect required light of wave lengths less than 2,800 A. It would therefore appear probable that the wavelength of the emitted light must lie between this value and the absorption band in quartz, about 1,500 A. Now from the National Critical tables there would appear to be some evidence of an absorption band in water, due to hydroxyl ions at about 1,900 A. It suggested that this would be the most likely wave length for the quanta so produced.

Results.

The foregoing experiments indicate that at least some, if not all, of the chemical effects associated with the ionization produced by alpha particles in liquids, may be due to quanta emission resulting from the primary ionization. Unfortunately it was not found possible to maintain a water film of known thickness for a period of several days, so no experiments were conducted to determine the range of the quanta liberated in water. Consequently, it is impossible to make an accurate estimation of the chemical effects produced by this radiation. However, from the results obtained, it would appear that the total chemical yield produced by alpha particles could be due to this phenomenon. In fact, if the quanta in water have a similar probability of being absorbed, as that found in the case of Nylon, the yields obtained which are about 1/20 of that obtained when the particles are allowed direct access to the solution are considerably higher than would be anticipated.

In all the experiments the water films were maintained by performing the experiments in a saturated atmosphere. It is therefore possible that the presence of water vapour is essential to these experiments. However, in the experiments in which the alpha particles were allowed to enter the solutions in which the chemical actions took place, the conditions were the same in this respect. The yields obtained were in good agreement with those reported by previous experimenters, who introduced the alpha particles into the solutions, which allowed them to pass through air. It is therefore probable that the presence of water vapour has little or no effect and so the chemical change produced must be due to some form of radiation originating from the water films. Moreover, at least some of this radiation must be capable of penetrating a quartz slide 1 mm. thick. This condition would preclude the possibility of the reactions being caused by particle emission. Since if these were to be electrons, the velocities required would need to be very much in excess of the velocities of the bombarding alpha particles. Therefore the most likely process is that of quanta emission. As in the work carried out on the scintillations from thin films, no direct evidence was obtained as to the frequency of the quanta produced. For reasons stated, it appears likely that these have a wave length of the order of 1900A. However, there is little or no evidence to preclude the possibility of the emission of X-rays, as was suggested in the case of thin films.

This uncertainty in the frequency produces a corresponding uncertainty in the yield which can be produced in this manner. From experiments in which chemical actions are produced by ultra-violet irradiation, the quantum yield remains more or less constant once a certain threshold value has been reached.(1) In the case of chloracetic acid, quantum efficiencies of the order of 10 molecules per quantum have been recorded for ultra violet radiation. However the yield drops to the more normal value of 1 or 2 when X-rays or gamma rays are used.

Another quantity about which little or no evidence is available is the efficiency of water films for producing quanta when bombarded with alpha particles. Again, from the thin film experiments, it would appear that at least a 100 quanta are produced by each alpha particle. In the case of chloracetic acid, if a quantum efficiency of 10^3 is assumed, then the chemical yields which have been measured can be explained in terms of quantum emission. Similar explanations are possible in the case of sodium iodide.

For the majority of chemical reactions due to alpha particles, the quantum efficiency for the same reactions caused by ultra-violet light is over the order of 2 or 3 molecules per quantum. If this is the case then the argument stated would not explain the total yield. Now, the estimation of a 100 quanta per alpha particle was a

(1) See Bibliography. Chemical Reactions caused by Ultra radiations.

lower limit based on number detected in the thin film experiments. It would appear probable that quanta emission can take place whenever an excited ion recombines to form a neutral molecule. If an estimate of the excited ions produced is made, by estimating the number of primary ions formed with the emission of a fast delta ray, then between 10^3 and 10^4 such ions are produced per alpha particle. Therefore there is a possible of 10^4 quanta being emitted from an alpha track 32 microns long. In the case of chloracetic acid this would give rise to a possible yield of 10^7 molecules, which is in excess of that measured. But the concentration of quanta would be very much greater than that produced by an external ultra-violet source. Consequently, the efficiency of the chain reactions initiated may well be reduced, due to the recombination of the radicals produced. In fact the probability of the recombination of the radicals due to the normal diffusion process will be in accordance with the theory proposed by Jaffé for ions. Now if the range of the quanta is of the order of 200 molecular distances, then a reduction in yield by a factor of 10^2 would be anticipated. It will be noted that this value is the same as the one given by the previous theory. However, in view of the fact that the yields obtained from alpha bombardment are more or less independent of the quantum efficiencies measured using ultra-violet light, the latter explanation appears to be more plausible. Further evidence in support of this view is obtained from the fact that the yields obtained from the experiments using thin water films are greater than would be expected. This may well be due to a reduction in the concentration of the quanta density. However, the same effect would be observed if the probability of absorption was less than has been assumed.

* * * * * * *

Conclusions.

The experiments described were performed in an attempt to investigate the effects of the ionization produced in a liquid by alpha particles. The first series of experiments were devoted to the measurement of the ion current which can be collected due to ionization produced by alpha particles. In agreement with the results obtained by previous experimenters, it was found that only a small proportion of the primary ions produced, can be collected. However, contrary to the predictions of Jaffé's Theory, no difference in the ionization current was recorded whether the alpha particles were travelling at right angles or parallel to the collecting field. However, owing to the noise generated in liquids when subjected to electric fields, it was found impossible to use field strengths in excess of 5 K.V./cms. Kramers suggested that Jaffé's theory should be modified to take into account that the effects of the electrostatic forces present in the columns were more important then those due to diffusion. This modified theory still predicts a variation in the ion current available, depending on the angle between the alpha tracks and the collecting field. Moreover, the value calculated for the fields occurring in the ion columns is of the order of 10^7 volt/cms. which is at least of the same order of magnitude as the "breakdown strength" of liquids. It therefore appears likely that the only ions available for collection are those produced by delta rays at some distance from the centre of the ion columns. This view is supported by the results of the experiments in which attempts were made to measure the specific ionization at various points along the alpha tracks. Further, the relationship between the field strength and the ion current obeys a logarithmetical law, as would be expected. Some idea of the diameter of the ion columns can be obtained from the estimation of the minimum energy of the delta rays which contribute to the ion current. It is not possible to make an accurate determination owing to the scattering probability of such electrons. However a maximum of value of 0.05 microns appears probable.

An attempt was made to correlate the results of the purely physical experiments with those of the chemical experiments. Experiments were performed in which quanta emission, associated with the ionization produced by alpha particles, was detected. Owing to the high coefficient of absorption of most materials for the quanta so produced, these experiments where performed with films, then compared to the range of the alpha particles used. Due to the difficulty of producing suitable films of liquid, and to problems concerned with the detection of ultra-violet quanta, most of the experiments were performed using solid films.

In all cases, irrespective of the material used, quanta were detected. It was not found possible to measure the frequency of the radiations, so that no definite evidence was obtained as to the origin of the radiations. Large crystals of the same materials were also tested for scintillations, but with the experimental radiations used, no counts were recorded. Similar experiments were carried out using beta particles and gamma rays instead of alpha particles. No quanta were detected in any of these experiments. It therefore appears that the emissions observed were connected with the dense ionization associated with alpha particles. Experiments were carried out to determine the effect of thickness on this phenomenon. It would appear that quanta emission is a direct function of the density of the primary ionization produced, but a function of the probability of producing excited ions.

Experiments were performed to demonstrate that at least some of the chemical effects caused by alpha particles in liquids, were due to quantum emission associated with the dense ionization. Quanta emitted as a result of bombarding a thin film of water was used to initiate chemical changes in suitable solutions. The yields obtained from these experiments were in agreement with those calculated assuming the entire chemical effects of an alpha particle are due to this cause. From the known threshold frequency needed to cause one of the chemical reactions investigated it was possible to obtain some idea as to the frequency of the radiations from the water films. This tended to confirm the suggestion that any emission would take place in the ultra violet region.

The following conclusions were drawn from the results of the experiments:

(1) Only a small proportion of the number of ions produced by an alpha particle in a liquid can be collected.

(2) The majority of the ion current which can be collected is due to delta rays.

(3) Quanta emission is associated with the ionization produced by an alpha particle in a solid. In most material these radiations are highly self absorbed. There is some evidence to believe that a similar phenomenon takes place in liquids.

(4) Chemical reactions which take place in aqueous solution are at least partly caused by these quanta.

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Appendix 1.

There appears to be a considerable amount of misunderstanding with regard to the performance and limitations of a cathode follower. In calculating the performance of such a system it is not usual to take into account the limit imposed by the maximum current available, and the effect of grid current on the input circuit. In other words it is assumed that the valve has an infinite current amplification factor. In the majority of cases, this view is substantially correct, but where pulses of short duration are involved, the assumption can no longer be made. For pulses of one microsecond or less duration, because of the high frequency components, the stray capacities between the cathode and earth become of paramount importance, and the valve cannot be regarded as operating into a resistive load. The problem then arises of supplying the necessary current to charge the capacities in the desired period. This current may reach very large values when the signals are fed along a transmission line. In this case, the total capacity of the load and strays connected with the cathode, must be taken into account and the required current must be supplied by the valve. For short pulses, such as obtained from a photo-multiplier used for detecting scintillations, the peak current required may be in excess of that obtainable from the cathode of the valve. In any case, a phase lag will occur between the cathode and grid voltages, increasing the effective grid cathode capacity. This in turn will reduce the input impedance. The problem can be reduced to a certain extent by using a valve capable of providing large currents. However, in most cases such valves have low amplification factors. Now the output impedance is usually taken as being the inverse of the amplification factor, so that if such valves are used, the output impedance is increased. Moreover the amplification of a cathode follower is given by:

$$A = \frac{1}{1 + 1/\mu}$$

where A is the amplification, and μ is the amplification factor of the valve. It will be seen that loss of signal amplitude occurs if valves with small amplification factor are used. It must also be remembered that in most valves the value of the amplification factor drops large currents and it is this value which is important during the period in which the grid voltage is rising. Similar effects occur when the grid volts fall. In this case the output impedance tends towards the value of the resistance used as the cathode load since the D.C.Resistance of the valve increases with reduction in the current. This will easily be seen if the valve is regarded as a variable resistance whose value is determined by the grid cathode potential. However, by the use of specially designed valves, such as a C.V173, these problems may be considerably reduced. This type of valve is capable of giving large currents for short periods and at the same time has a high amplification factor. These qualities are obtained by using a close meshed grid close to a large cathode. As a result, the input impedance of such a valve is comparatively low.

There are two ways in which the ratio between the input and output impedances can be improved, both involving the use of more than one valve. The first, and more simple, is to use one cathode follower driving another. In this system the first, consists of a small high gain valve which is used to supply the power necessary to drive a valve capable of supplying the necessary current to feed the signals into the load. Such a system has the disadvantage that some loss in signal amplitude occurs since the voltage amplification of a cathode follower is less than one.

The other method consists of using three valves arranged as in the circuit shown in Fig.1A. V_1 and V_2 are two high gain valves connected as a voltage amplifier. The signals from the anode of V_2 which are in phase with the input, are then fed on to the grid of V_3 which is connected as a cathode follower. Part of the cathode load of V_3 also forms the cathode load of V_1 .

A portion of the output is therefore fed on to the cathode of the input stage, and any tendency for reduction in the amplification of the whole system due to a capatative load is compensated by an increase in the amplification obtained in V_1 and V_2 . The amplification of the system is given by the proportion of the signal which is "fed back" to the cathode of the input valve, or more accurately:

$$\frac{R_1+R_2}{R_2} \cdot \frac{1}{1+1/U_1.U_2} , \frac{1}{1+1/U_3}$$

The relation between the output and input impedance is given by:

where U_1 ,U_2 ,U_3 are the amplification factors of the valves used. However, there is still the limitation in the output impedance imposed by the maximum current obtainable from V_3 . Also the input impedance is limited by the grid impedance of V_1 . When such a system was used with a phot-multiplier, a gain in the signal to noise ratio of five was obtained, compared with that using a simple cathode follower.

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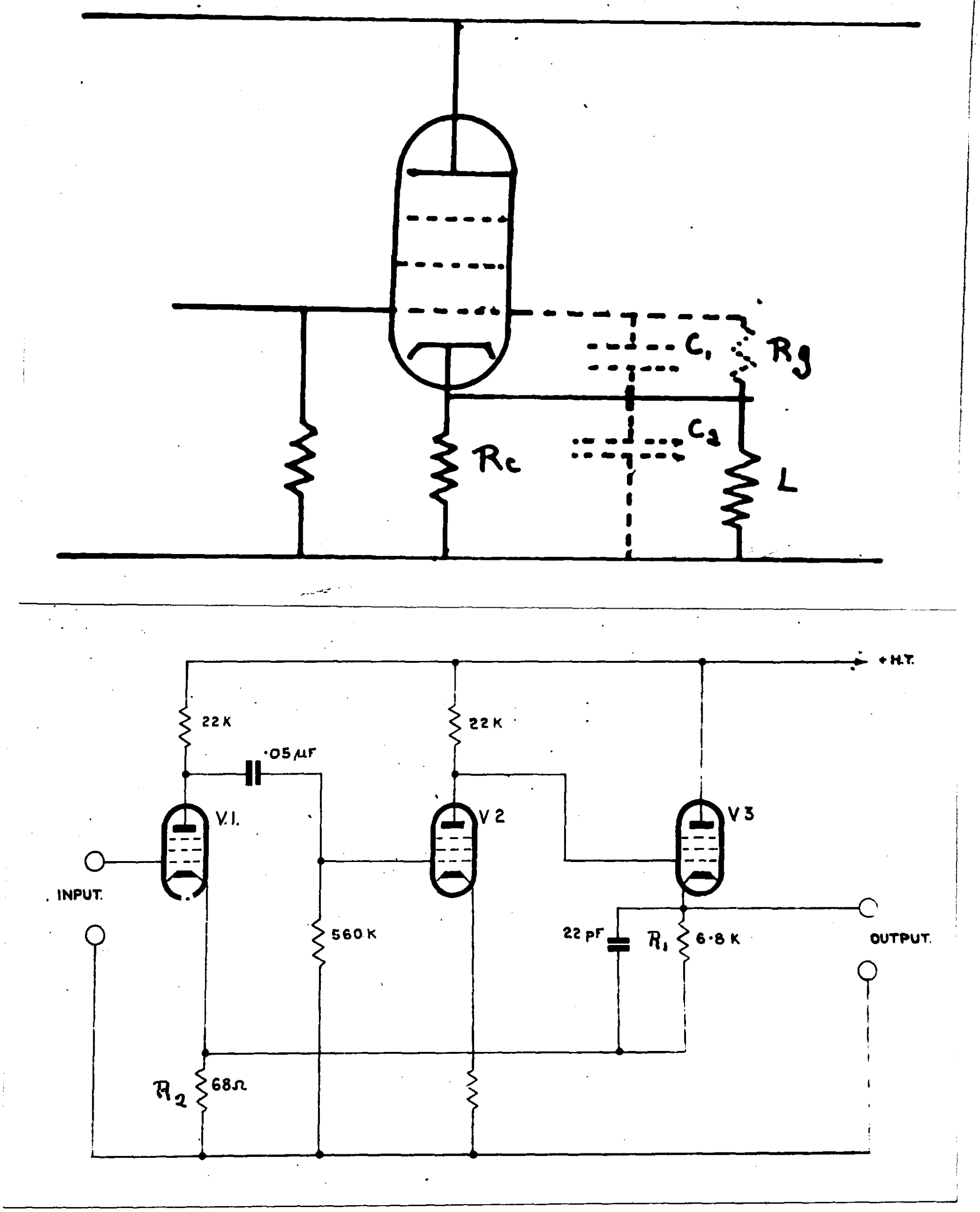

Fig Ia Basic Circuit Diagram For a Cathode Follower, and a 'Ring of Three!

Appendix 2.

In connection with the experiments performed on detecting the scintillations from thin films a single channel pulse amplitude analyser was designed. The circuit used was as shown in Fig. 2A.

The principle of the system was that the signals were split into two channels and fed into two discriminators, whose bias was held a known voltage apart. The signals produced in the discriminators were then fed into an anticoincidence circuit, the output of which was used to drive a scaling unit. Owing to the input signals having a finite rate of rise, it was necessary to delay the start of the signal produced by the discriminator which triggered at the lower voltage.

V_1 was a phase inverter, and it also served to prevent any effect of the discriminator circuits being fed back to the amplifier. The inverted signals were split into two channels and each section fed through a series diode, which were used as discriminators. This was accomplished by altering the bias in D_1 and D_2, that on D_2, being maintained at a higher value to that on D_1 by means of a grid bias battery. This voltage governed the width of the channel on which the signals were accepted. The signals from the diodes were then used to trigger the two flip-flops formed by V_2, V_3 and

V_4 V_6. The standard signal produced at the anode of V was fed on to the grid of V_6, which was normally in the non-conducting state maintained by the cathode bias applied. This valve was a V.R.116 chosen for its short suppressor bias, about 19 volts. The screen was connected to H.T. through a coil L_1, and also the suppressor by an 0.01 uF Condenser. When a positive signal was applied to the grid of V_6, the screen potential fell, due to the change in current through L_1, causing a corresponding reduction in the suppressor voltage. Hence no signals were produced across the anode load until the suppressor voltage returned due to the return of the screen potential. The time interval was controlled by the value of the inductance of L_1, and was so chosen to give a delay of about 2 microseconds. The delayed signals were inverted in V_7 and fed to the grid of V_8 which was also a V.R.116. The negative signals from the anode of V_5, one of the valves of the second flip-flop were fed to the suppressor of V_8. This valve was normal in a non-conducting state so that signals were only produced at the anode when a positive pulse was applied to the grid and there was no negative pulse on the suppressor. The anode signals were then inverted in V_9 and taken to a scalar unit type 1OB7.

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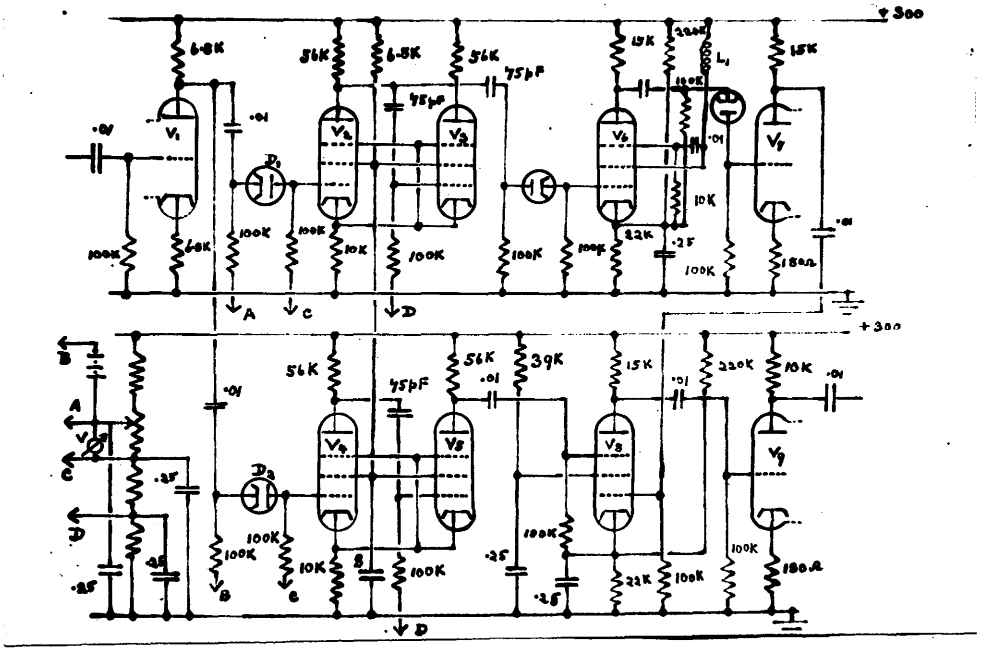

Fig 2a Circuit Diagram of the Pulse Analyser.

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