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ENERGY LOSS OF DEUTERONS IN D₂O AT VERY LOW ENERGIES

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Introduction

In evaluating the cross section for a nuclear reaction from the observed values of thick-target yields, it is necessary to know the energy-range relation for the bombarding particles in the target material. It is the purpose of this paper to deduce from existing experimental evidence the rate of energy loss of deuterons in a D_2O (heavy ice) target, for the special case of very low bombarding energies (10 to 100 kev).

The data most pertinent to our problem are contained in several papers^(1,2,3,4) by Gerthsen and his co-workers, concerning the energy loss of slow protons in various media (air, hydrogen and celluloid). Their results are applicable to our case through the following assumptions:

- (a) That the rates of energy loss of a proton and a deuteron having the same velocity are identical in any given medium.
- (b) That the atomic stopping powers of H and D are identical.
- (c) That the molecular stopping power of D_2O can be found by adding the atomic stopping powers of two atoms of D and one atom of O.
- (d) That the stopping power of D_2O is independent of its physical state.

Although there has been no experimental verification of assumption

- (a) for low energies, it is a general principle that a proton and a deuteron having the same velocity will lose energy through electronic interactions at

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| (1) Chr Gerthsen | Ann. d. Phys. <u>5</u> , 657 (1930). |
| (2) " " | Phys. Zeits <u>31</u> , 448 (1930). |
| (3) A. Eckardt | Ann. d. Phys. <u>5</u> , 401 (1930). |
| (4) W. Reusse | " " " <u>15</u> , 256 (1932). |

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equal rates, whatever the exact nature of the energy loss process⁽⁵⁾. We shall therefore take it that this assumption is rigorously justified. Except where otherwise stated, values of energies mentioned in the text are those appropriate to protons; any relation arrived at for protons will be true for deuterons of twice the energy.

With regard to assumption (b), measurements have been made^(6,7,8) of the relative total amounts of ionization produced by protons and deuterons in hydrogen and deuterium gas. The results indicated that a slight difference existed between the total ionizations in H₂ and D₂, which would invalidate the assumption. The difference was the same whether the incident particle was proton or deuteron. The difference seemed, however, to arise only at very low incident-particle velocities ($6 \cdot 10^7$ cm/sec corresponding to 2-kev protons or 4-kev deuterons). Thus for proton or deuteron velocities greater than $6 \cdot 10^7$ cm/sec, one may consider the atomic stopping powers of H and D to be equal.

The basis for assumptions (c) and (d) is to be found in a paper by L. H. Gray⁽⁹⁾, where these matters are reviewed. The molecular stopping power of a chemical compound appears to be accurately equal (to about 2%) to the sum of the atomic stopping powers of its constituent atoms. Moreover, the limited evidence concerning the stopping powers of a substance in solid and gaseous forms suggests that there is no significant difference between them. It should be pointed out, however, that these results are based upon measurements with alpha particles, and at these high energies

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| (5) Livingston & Bethe | Rev. Mod. Phys. <u>9</u> , No. 3, p.271, (1937). |
| (6) R. Koops | Ann. d. Phys. <u>33</u> , 57 (1938). |
| (7) G. Joos | " " " <u>41</u> , 426 (1942). |
| (8) M. Jussuf | " " " <u>41</u> , 435 (1942). XXXXXXXXXX |
| (9) L. H. Gray | Proc. Camb. Phil. Cos. <u>40</u> , 72 (1944). |

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the difference in stopping power due to differences in chemical binding is expected on theoretical grounds to be less than 1%. The situation is markedly changed at the energies in which we are here interested, and the stopping power of heavy ice, as calculated from the observed atomic stopping powers of gaseous hydrogen and oxygen, may well be in error.

With this brief preamble we will proceed to a survey and analysis of the experiments of Gerthsen, Eckardt and Reusse.

The Loss of Energy of Slow Protons in Matter.

The experiments on the loss of energy of protons in matter can be classed into two types, integral and differential. In the first type of experiment, the total range in a gas of a proton of given initial energy is measured. This is the experiment described by Gerthsen⁽¹⁾. His results are shown in Figure 1. for the case of protons in air. The ranges R are expressed in cms of air at 1 mm pressure Hg, and the proton energies E in kev. Gerthsen found (cf. Figure 1) that R as a function of E could be very closely represented by the formula

$$R = \alpha E^{3/4} \quad (1)$$

(We have applied a least-squares analysis to his data, and have found that the results are better fitted if the power of E is 0.773; this difference, however, is scarcely significant). Now the quantity of interest in determining the cross section for a nuclear reaction is $-dE/dx$, the energy loss per unit distance in the target at a given energy. If we accept Gerthsen's formula, we have:

$$-dE/dx = \frac{4}{3\alpha} E^{1/4} = \text{const} \times v^{1/2}. \quad (2)$$

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dE/dx as a function of velocity is plotted in Figure 2.

In the differential experiments, the loss of velocity of protons in traversing a very small quantity of matter is directly observed. This is the type of experiment described by Eckardt⁽³⁾ and Reusse⁽⁴⁾. A proton beam was passed through celluloid films of various thicknesses Δx . For a given film, the energy loss ΔE in passing through it was measured as a function of the initial energy E . The results are shown in Figure 3. By considering the results for the various films, one can find the magnitude of $\Delta E/\Delta x$ as a function of Δx for a given E . The relation between them is found to be linear, of the form

$$-\frac{\Delta E}{\Delta x} = a' - b' \Delta x \quad (3)$$

By extrapolating the line to zero film thickness, one finds the value of dE/dx at the initial energy E . By performing this analysis for a series of values of E , one can obtain dE/dx as a function of velocity. We have treated all of the data of Eckardt and Reusse in this way, using least-squares solutions throughout, and find that they are consistent with a relation

$$-\frac{dE}{dx} = \beta (v - v_0) \quad (4)$$

This is plotted in Figure 4.

The discrepancy between the results of Gerthsen and of Eckardt and Reusse is very striking. From the one experiment one finds that dE/dx is proportional to the velocity, from the other to its square root. It is true that the values of $\Delta E/\Delta x$, obtainable from the measurements of Eckardt and Reusse, lead more directly to a value for dE/dx than do Gerthsen's measurements of total range, but both experiments are open to criticism and

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merit more detailed consideration.

The Experiment of Gerthsen

A proton beam generated in a canal-ray tube was magnetically analysed and passed through a thin celluloid window situated at the radius of curvature of a hemispherical ionization chamber. The thickness of the window was $80 \text{ m}\mu$ ($1 \text{ m}\mu \equiv 10^{-6} \text{ mm} \equiv 10 \text{ A.U.}$). The ionization chamber could be filled to any desired pressure (up to a few cms Hg) with air or H_2 . The positive ions produced by the protons in traversing the chamber were collected on an electrode which was made negative with respect to the walls of the chamber. The charge collected in a given time was registered on an electrometer; a second electrometer recorded the charge carried by the primary proton beam entering the chamber. The ionization current was found to increase steadily with increasing gas pressure up to a certain critical pressure p_c , and thereafter remained constant. At the pressure p_c the protons are just failing to reach the walls of the chamber, so that for this and higher pressures they lose their whole energy in the gas. If the radius of the chamber is ρ , the proton range r at unit gas pressure is ρp_c .

Gerthsen's experiment consisted in measuring p_c for air and H_2 for several values of E between 27 and 57 kev. He found that the range in H_2 at all energies was 2.50 times the corresponding range in air, so that the same form of the energy-range relation must hold for protons in air and in H_2 . It is necessary to point out that the value of E was measured before the protons passed through the celluloid window. Gerthsen assumed that the window was equivalent to the same thickness δ of air at all energies, and calculated δ by considering the celluloid to be composed of hydrogen and "air-like" atoms. In this latter category he placed the C, N and O atoms which composed 94.4% of the celluloid by weight. δ was then added to

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each observed value of r to give the total range R . \int was about 10% to 20% of R , so that any error in its value could materially affect the final results.

There exists an additional way of evaluating dE/dx from Gerthsen's experiment, which he himself describes⁽²⁾. When the pressure in the ionization chamber was very low, the ionization current was found to be a linear function of pressure. The slope of the current vs. pressure curve in this region gives directly the number of ion pairs, per mm pressure of gas in the chamber, produced by the whole proton beam. Since the primary proton beam was simultaneously recorded, it is possible to state the number, n , of ion pairs per cm of path, in gas at 1 mm pressure, produced by one proton of energy E' , where E' is the energy of a proton after it has lost energy ΔE in traversing the celluloid foil. With the assumed value of \int , ΔE and thence E' could be calculated for various values of E .

To translate n into a rate of energy loss, it is necessary to know the mean energy, W , required to produce an ion pair in the gas at the energy E' . Gerthsen could not determine W at a single energy, but by dividing the saturation current in the ion chamber by the current of primary protons, he could determine the total number N of ion pairs produced by a single proton of energy E' . The quotient E'/N then gave the average value of W over the energy range 0 to E' . This ratio was found to be independent of the value of E' ; it was therefore assumed that W itself was independent of energy, and equal to the constant quotient E'/N . (From our evaluation of the data we find $W = 38$ ev. Gerthsen gives $W = 35$ ev, but we have cause to suspect the values of E' on which this is based -- see below).

The rate of energy loss, $-dE'/dx$, of a proton of energy E' is then given by the product nW (key per cm of path at 1 mm pressure).

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Support for the assumed constancy of W is to be found in a paper of Joos⁽⁷⁾. He quotes an experiment in which the ratio E'/N for protons in H_2 is found constant for energies from 4 to 14 kev.

As Gerthsen has published his results, the values of dE/dx derived by this second method from his one experiment are very different from its values as derived by the first method mentioned earlier. (dE/dx appears to be proportional to v , not to $v^{1/2}$ as required by equation (2)). Upon careful re-evaluation of the results, we found that his values of ΔE , and hence of E' , were at variance with the integral energy-range relation. (equation 1). It would appear that the discrepancy may have arisen through numerical errors; in our own treatment of the data we have found that the values for dE/dx are essentially the same by either method of evaluation, and satisfy the relation expressed by equation (2). In determining ΔE , we took the mean of values obtained by three different methods. The first was that used by Gerthsen⁽¹⁾ (although our results differ from his, and satisfy the energy-range relation); the second method was to assume that the loss of velocity, Δv , in the celluloid foil was independent of the energy of incidence E (a result found by Eckardt and Reusse, v. inf.) and to accept Gerthsen's value for the air equivalent σ of the foil; the third method was to find directly from the data of Eckardt and Reusse the loss of energy suffered by a proton of various energies E in passing through a celluloid foil of 80 μ thickness.

The outcome of Gerthsen's experiment would thus appear to be that over the range 20 to 60 kev the rate of loss of energy of protons in air or hydrogen is proportional to the square root of the velocity. The actual magnitude of this loss, in kev per cm of path in either gas at 1 mm pressure, follows directly from the experimental results.

The Experiments of Eckardt and Reusse

A proton beam generated in a canal-ray tube was accelerated through an additional potential drop and was then magnetically analysed. Protons or H_2^+ ions of a specified velocity were thus selected. The range of proton energies covered was from 4 to 50 kev. After passing through a thin celluloid film the protons were subjected to a further magnetic analysis in order to determine their velocity upon emergence. There was of course a spread of energy in the emergent beam, but the velocity at the peak of the distribution was measured as the significant one. The experiment was repeated for celluloid films of various thicknesses (from 20 to 330 $m\mu$).

It may be noted that the celluloid films were extremely thin, the thickest being only about 3000 A.U. Thus it was impossible to measure the thickness through the use of interference fringes, and the method employed was to note the change of interference colour when one of the thin films was placed over a relatively thick film of celluloid. Unfortunately no details of the procedure are given in any of the published papers, and its accuracy must remain open to question.

The results given by Eckardt and Reusse are (a) that for a given incident velocity, the loss of velocity in passing through a foil is proportional to its thickness, and (b) that in passing through a given foil, the energy loss is a linear function of the incident velocity v . These results may be expressed by the following equations:

$$- \Delta v = a \cdot \Delta x \quad (5a)$$

$$- \Delta E = b \cdot v - c \quad (5b)$$

It will be noted that equations (5) are not the same as the

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equations (3) and (4) which we have used in evaluating dE/dx as a function of velocity. The two sets of equations may, however, be readily related. Suppose that the rate of energy loss is given by

$$- \frac{dE}{dx} = f(v) \quad (6)$$

i.e., $-mv \cdot dv = f(v) \cdot dx$, where m = mass of proton.

Then

$$\Delta x = -m \int_v^{v+\Delta v} \frac{v \, dv}{f(v)} \quad (7)$$

$$= +m [\phi(v) - \phi(v + \Delta v)] \quad \text{say}$$

If we accept equation (5a), we have $\Delta x = -\frac{1}{a} \Delta v$, and hence the identity:

$$-\frac{1}{a} \Delta v \equiv m [\phi(v) - \phi(v + \Delta v)] = -m \phi'(v) \cdot \Delta v + \dots$$

Consequently, $\phi'(v) = +\frac{1}{am} = +\frac{v}{f(v)}$ from (7)

so that $\frac{dE}{dx} = -amv$ (8)

Differentiating (8), $\frac{d^2E}{dx^2} = -am \frac{dv}{dx} = +a^2m$, from (5a) (9)

Now we may write $\Delta E \cong \frac{dE}{dx} \Delta x + \frac{1}{2} \frac{d^2E}{dx^2} (\Delta x)^2$, which by

(8) and (9) becomes $\frac{\Delta E}{\Delta x} \cong -amv + \frac{1}{2} a^2m \cdot \Delta x$

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$$\text{or } -\frac{\Delta E}{\Delta x} = \alpha_1 v - \alpha_2 \Delta x, \text{ where } \alpha_1 v = -\frac{dE}{dx} \quad (10)$$

Equation (10) may be seen at once to correspond to equations (3) and (5b) (to the former when v is fixed and Δx varies, to the latter when Δx is fixed and v varies).

The following table sets out some values of $\alpha_1 v (= -dE/dx)$, $\alpha_1 v/E^{\frac{1}{2}}$ and α_2 as we have calculated them by a least-squares analysis of the experimental data of Eckardt and Reusse:

TABLE 1

E kev	$\alpha_1 v$ kev/m μ of celluloid	$\alpha_1 v/E^{\frac{1}{2}}$	α_2
5.8	0.0395 \pm 0.0062	0.0164 \pm 0.0026	0.00018 \pm 0.00008
10.2	0.0621 \pm 0.0040	0.0195 \pm 0.0013	0.00019 \pm 0.00003
31	0.0890 \pm 0.0059	0.0160 \pm 0.0011	0.00009 \pm 0.00001
44	0.1313 \pm 0.0061	0.0198 \pm 0.0009	0.00017 \pm 0.00001

The fact that α_1 and α_2 are essentially independent of velocity may readily be seen.

In comparing equation (8) with equation (4), we see that they are in agreement only if v_0 is set equal to zero. The discrepancy probably arises because we have made a more careful analysis of the data, using least-squares solutions throughout, than did Eckardt and Reusse. In this analysis the parameter v_0 appeared, but it is doubtful that the experiments in themselves are accurate enough to give v_0 any significance. In any case it is clear that the energy loss cannot cease at and below v_0 , as equation

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(4) would demand. ($v_0 \sim 0.2$ kev proton energy).

The result of the experiments of Eckardt and Reusse is therefore that, for protons traversing celluloid, the rate of energy loss, $-dE/dx$, is a linear function of velocity. The result is of use to our problem only if we can derive from it the absolute value of the rate of energy loss in air or in hydrogen, and thus, using our initial assumptions, find the energy loss in a layer of ice. It is important to attempt this conversion, since the celluloid measurements extend to much lower energies (4 kev) than do the measurements on air and H_2 . We will therefore consider this matter in the next section.

The Energy Loss in Ice

As was stated at the beginning, we derive the rate of energy loss in D_2O , in either vapour or solid form, by adding the rates of energy loss (which are directly proportional to the stopping powers) of two atoms of H (or one molecule of H_2) and one atom of O (or half a molecule of O_2). That is:

$$\left(\frac{dE}{dx}\right)_{D_2O} = \left(\frac{dE}{dx}\right)_{H_2} + \frac{1}{2} \left(\frac{dE}{dx}\right)_{O_2} \quad (11)$$

Since no data exist at low energies for the energy loss in O_2 , we have taken a result⁽¹⁰⁾ which holds for alpha particles near the end of their range, namely:

$$\left(\frac{dE}{dx}\right)_{O_2} = 1.07 \left(\frac{dE}{dx}\right)_{air}, \quad \text{where} \quad \left(\frac{dE}{dx}\right)_{air}$$

refers to one "molecule" of air. Now Gerthsen's observation that the range in H_2 is always 2.50 times the corresponding range in air leads at once to the relation

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$$\left(\frac{dE}{dx}\right)_{H_2} = \frac{1}{2.50} \left(\frac{dE}{dx}\right)_{air} = 0.40 \left(\frac{dE}{dx}\right)_{air}$$

Using the above two equations, (11) becomes

$$\left(\frac{dE}{dx}\right)_{D_2O} = \left(\frac{dE}{dx}\right)_{air} [0.40 + 0.535] = 0.935 \left(\frac{dE}{dx}\right)_{air} \quad (12)$$

With the aid of (12), Gerthsen's values for $(dE/dx)_{air}$ (cf. Figure 2) can at once be converted into the corresponding values for heavy ice.

To convert $(dE/dx)_{celluloid}$ into $(dE/dx)_{D_2O}$, two alternative methods are possible. The first method is briefly as follows: Over the small range of velocities covered by Gerthsen, the plot of $(dE/dx)_{air}$ vs. v does not depart by more than about 10% from the linear relation expressed by equation (4). If one therefore draws a straight line through these points, with an intercept at v_0 on the v axis, its slope is not likely to be in error by more than about $\pm 10\%$. By comparing this slope with the slope of the corresponding line for the energy loss in celluloid (cf. Figure 4), one finds the number of cms of air at 1 mm pressure which are equivalent to 1 m μ of celluloid. The result of this comparison is:

$$-\left(\frac{dE}{dx}\right)_{air} = (0.40 \pm 0.04)(v \times 10^{-8} - 0.18), \quad (13)$$

where v is in cm/sec and $(dE/dx)_{air}$ is in kev per cm of air at 1 mm pressure.

The conversion to $(dE/dx)_{D_2O}$ then follows from (12).

The second method of converting the celluloid data into energy losses in D_2O is to assume, with Gerthsen, that celluloid may be considered as a combination of air and hydrogen. That is:

(10) Rutherford, Chadwick & Ellis, "Radiations from Radioactive Substances"
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$$\left(\frac{dE}{dx}\right)_{\text{celluloid}} = A\left(\frac{dE}{dx}\right)_{\text{air}} + B\left(\frac{dE}{dx}\right)_{\text{H}_2} = (A + 0.4 B) \frac{dE}{dx}_{\text{air}}$$

The constants A and B may be evaluated from the composition and density of celluloid. For the density, which is not stated by Eckardt and Reusse, we have assumed a value of 1.48 gm/cc, which lies midway between the accepted limits of 1.35 and 1.60. We thus subject ourselves to a possible error of $\pm 10\%$. Our estimate of $(dE/dx)_{\text{air}}$ by this means is

$$\left(\frac{dE}{dx}\right)_{\text{air}} = (0.37 \pm 0.04)(v \times 10^{-8} - 0.18) \quad (14)$$

The striking agreement between (13) and (14) must of course be considered fortuitous.

Energy Losses Below 20 Kev

The outcome of the above is that, over the energy-range which is common to both sets of experiments (20 to 50 kev), the value of $(dE/dx)_{\text{D}_2\text{O}}$ calculated in various ways is self-consistent to about $\pm 10\%$. The matter of its absolute accuracy is of course another question.

In evaluating $(dE/dx)_{\text{D}_2\text{O}}$ for lower energies (from 5 to 20 kev), the problem becomes more difficult. One must attempt to decide what is the form of the energy-range relation in this region of low velocities. It is true that the measurements of Eckardt and Reusse are the only ones actually carried out in this region, and lead to the relation expressed in equation (4), but the limited validity of any one formula for expressing the range-energy relation should be recognized.

To discuss this in more definite terms, suppose that the range-

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energy relation is described for a specified velocity v by the equivalent formulae

$$\left. \begin{aligned} R &= \alpha v^m \\ -\frac{dE}{dx} &= \beta \frac{v^{2-m}}{m} \end{aligned} \right\} \quad (15)$$

For regions of high energy, the well-known Geiger law asserts that $m = 3$. Gerthsen's experiment gives $m = 3/2$. The results of Eckardt and Reusse correspond to $m = 1$. It would therefore seem likely that m is a continuously changing exponent, and that although equation (4) may represent $-dE/dx$ adequately over the range 4 to 50 kev, it may well fail for higher or lower energies. In fact it almost certainly must. We have already remarked that equation (4) can scarcely continue to hold down to the velocity v_0 . At the high-energy end it is equally unsafe to extrapolate it. For proton energies of 200 kev or more, dE/dx can be calculated with some assurance on theoretical grounds⁽⁵⁾. In this region $-dE/dx$ is decreasing with increasing velocity, so that between 50 kev and 200 kev a maximum (corresponding to $m = 2$ in equation (15)) must occur. Thus at this higher energy end Gerthsen's result, with $m = 3/2$, should perhaps be given some weight.

In the light of the foregoing considerations we have decided that, in the region 5 to 20 kev, $-dE/dx$ is probably best expressed by combining equations (13) and (14), with the proviso that, at the smaller energies, the rate of energy loss thus derived may be somewhat lower than the true value. For energies from 20 to 50 kev, we think it best to take the mean of the values obtained from the two experiments. In Table 2, which follows, list the values of $-\left(\frac{dE}{dx}\right)_{n,n}$, for various proton and deuteron energies,

which come from following this procedure:

TABLE 2

E_p Proton en- ergy kev	E_d Deuteron energy kev	$-\left(\frac{dE}{dx}\right)_{D_2O}$ kev/cm	E_p Proton en- ergy kev	E_d Deuteron energy kev	$-\left(\frac{dE}{dx}\right)_{D_2O}$ kev/cm
5	10	0.29	30	60	0.84
10	20	0.43	35	70	0.89
15	30	0.55	40	80	0.94
20	40	0.72	45	90	0.98
25	50	0.78	50	100	1.03

These are plotted in Figure 5. $-\left(\frac{dE}{dx}\right)_{D_2O}$ is here expressed in kev per cm of D_2O vapour at 1 mm pressure at about $15^\circ C$. (We assume that 15° was the approximate temperature at which Gerthsen conducted his experiments on air and H_2). Now one cc of D_2O vapour under these conditions weighs 1.11 micrograms, so that if one accepts our initial assumption (d), the losses of energy listed above are those occurring in a layer of heavy ice of thickness 1.11 $\mu g/cm^2$. In this form they are immediately applicable to the results of thick target yields in the $D+D$ reaction.

Conclusion

We have offered what we consider to be a reasonable estimate of the energy loss of slow protons or deuterons in heavy ice. If one takes into account the extreme difficulty of performing experiments in this energy region, the differences between different types of experiment are not excessive. It is abundantly clear that the problem demands much closer investigation than it has hitherto received, and that until this has been done any statements on the energy-loss process are bound to be largely conjectural. Nevertheless

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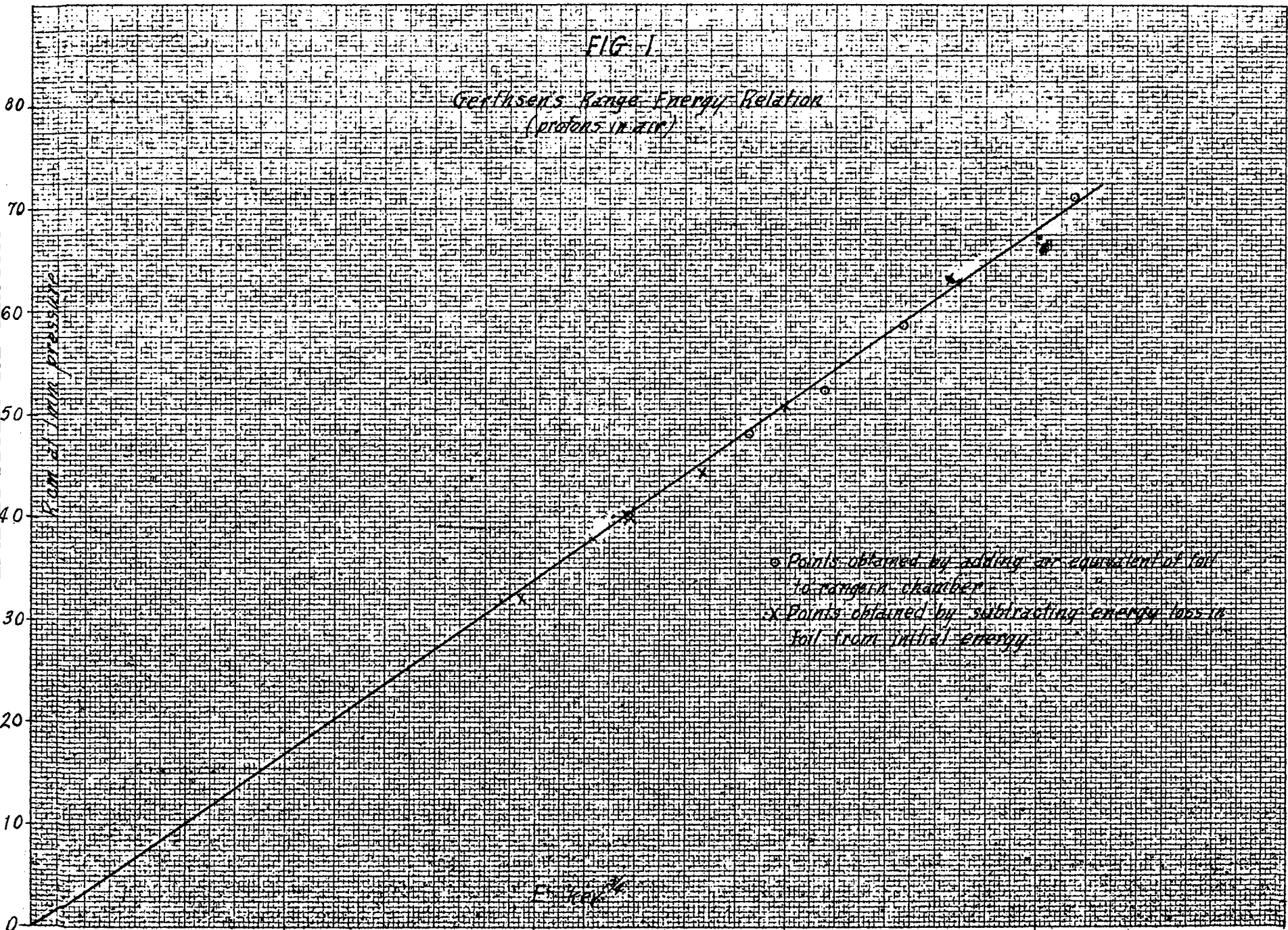
we hope that this account may prove a useful summary of the work that has so far been done in this field.

FIG-1

*Geithusen's Range-Energy Relation
 (protons in air)*

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Range in air (mm)



*o Points obtained by adding an equivalent of foil to range chamber
 x Points obtained by subtracting energy loss in foil from initial energy*

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FIG. 2

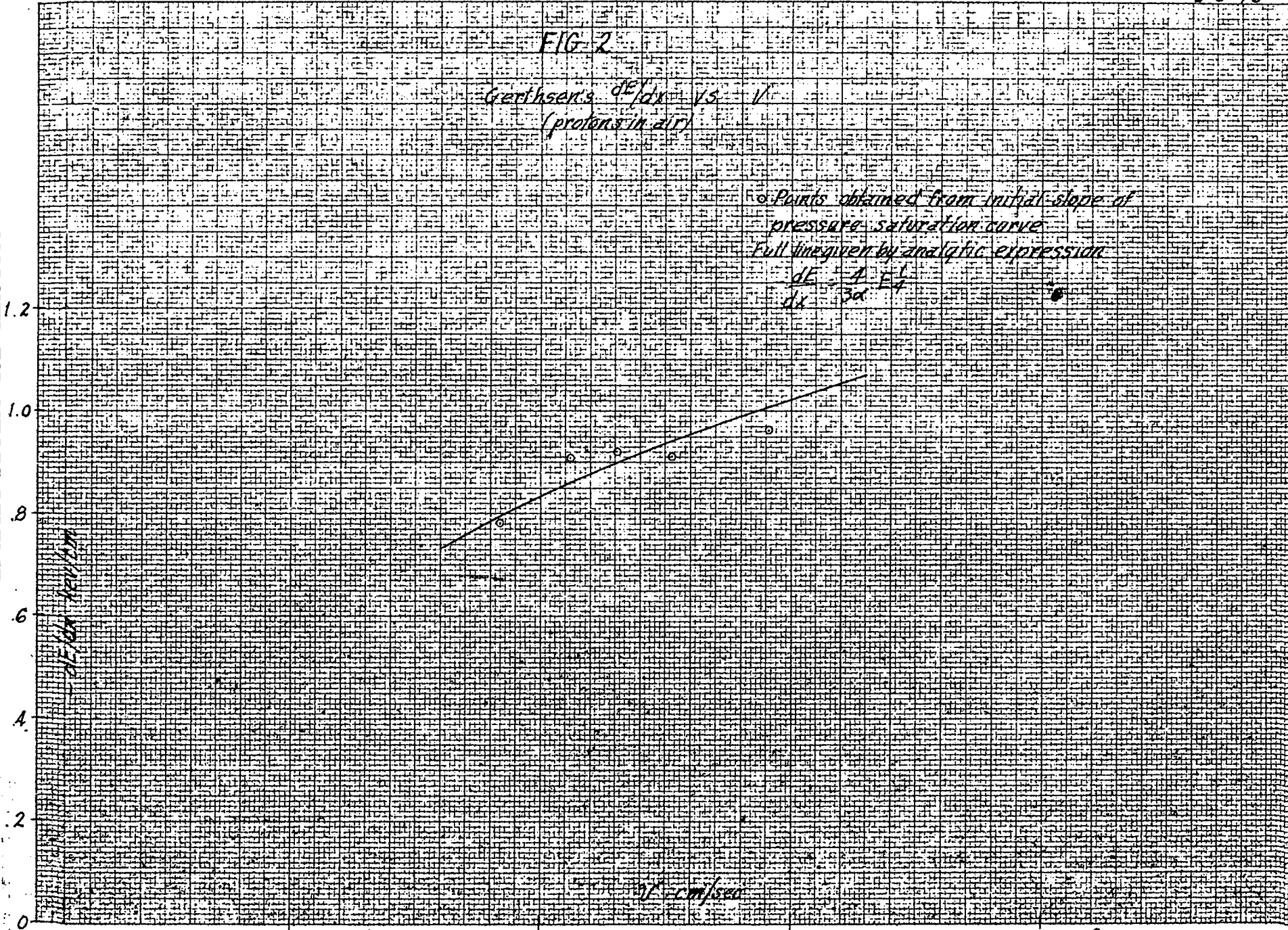
Gerthsen's $\frac{dE}{dx}$ vs V
 (protons in air)

o Points obtained from initial slope of
 pressure saturation curve
 Full line given by analytic expression

$$\frac{dE}{dx} = \frac{4}{3d} E^2$$

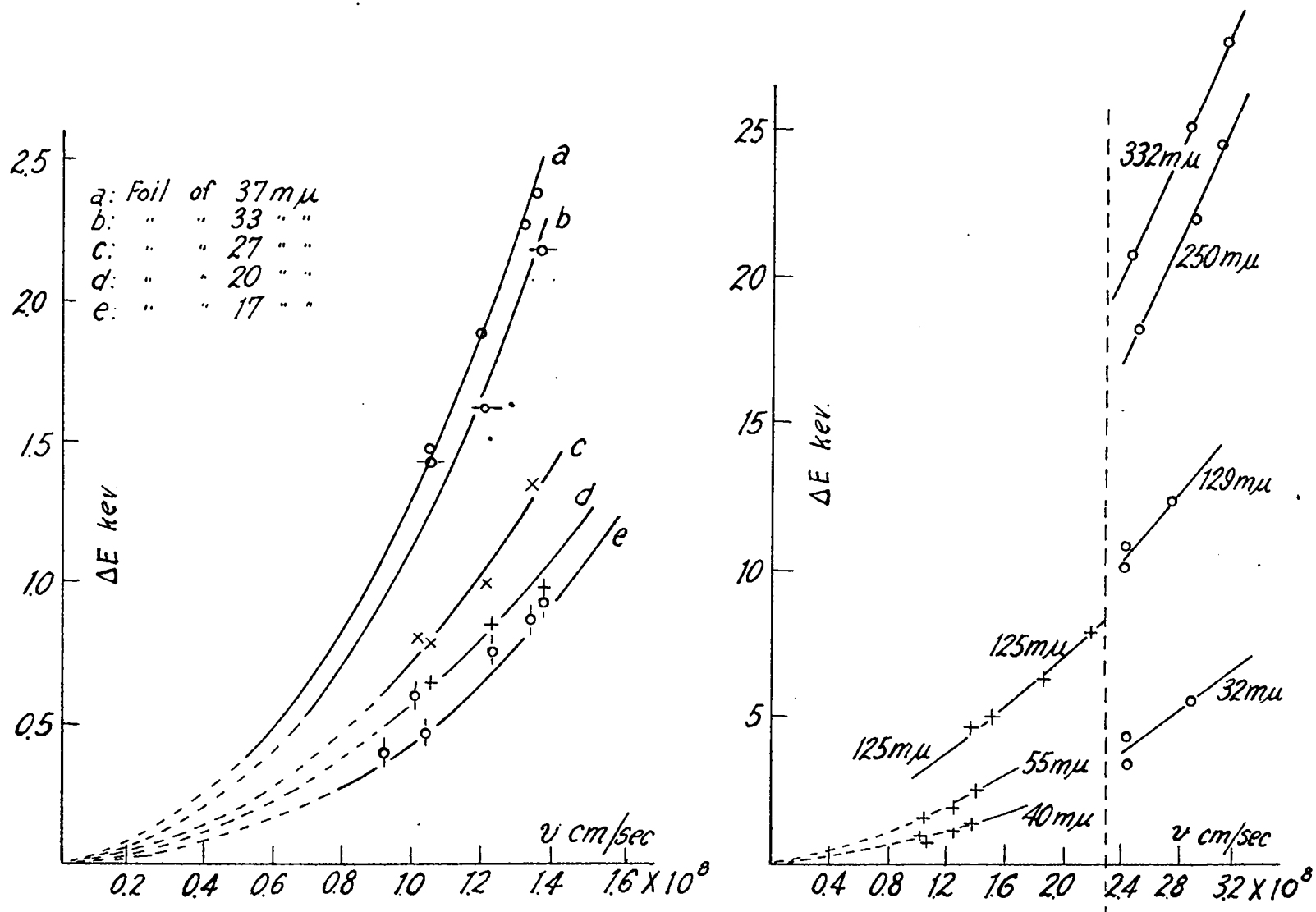
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31 cm/sec

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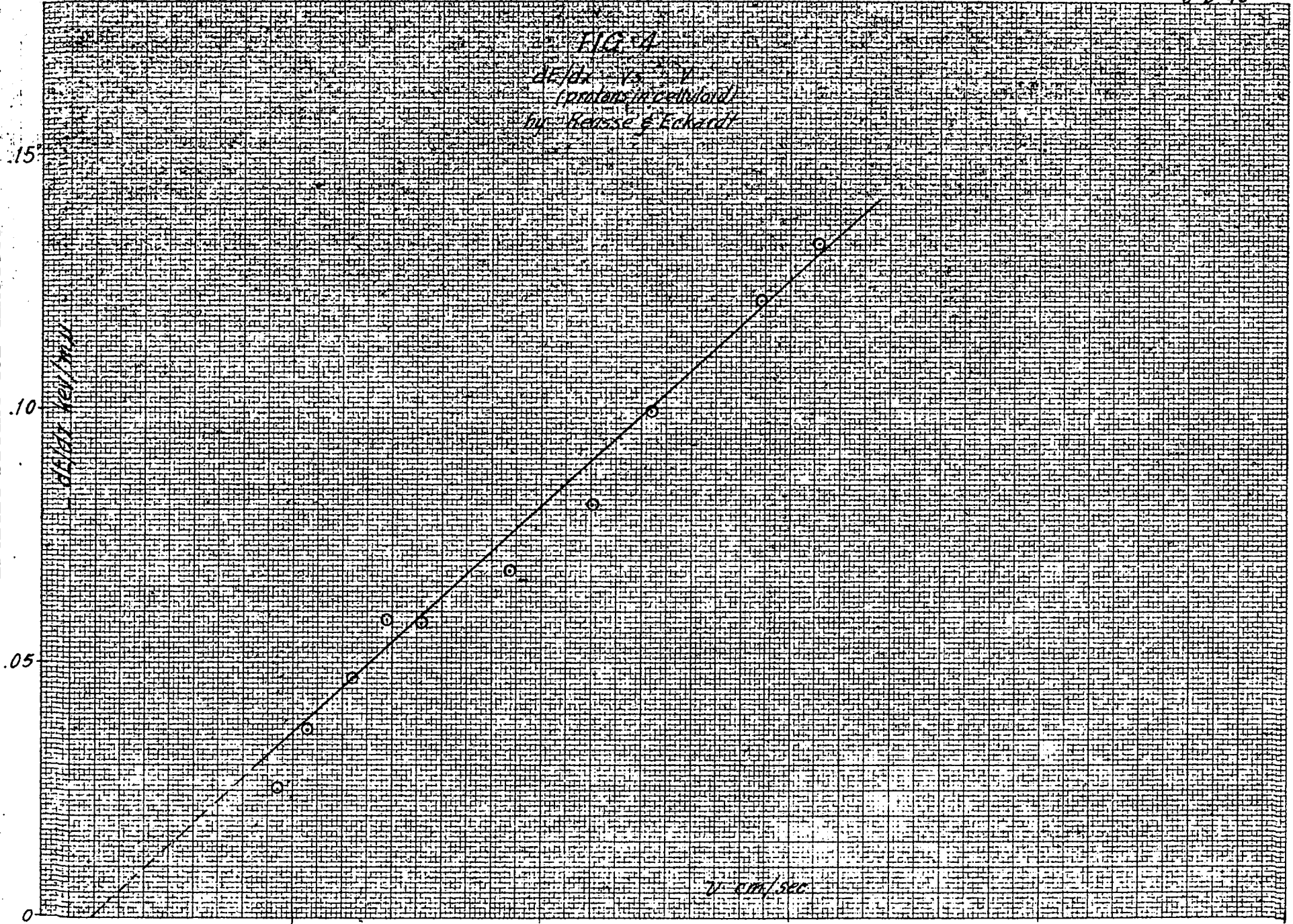


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FIG. 3
 Eckardt and Reusse
 ENERGY LOSS OF PROTONS IN CELLULOID FOILS

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FIG. 4
 $\Delta x/\Delta x_0$ vs v
(protons in celluloid)
by Keesse & Eckardt



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