

New Method for Measuring the α -Particle Range and Straggling in Liquids

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(Received October 25, 1954)

The ranges of Po α particles in water and five other liquids have been measured with a new method involving a steel surface as support for the α source and a CaWO₄ crystal surface as detector for the α particles. The surfaces are optically plane and parallel and constitute the walls of a microcell for the liquid. The range obtained at 25°C in water was 39.9 microns, 2.7 percent longer than the value 38.8 microns, computed with the Bragg law from range measurements in gaseous hydrogen and oxygen. In the other five liquids the ranges obtained also were longer (2.3–4.6 percent) than the correspondingly computed Bragg law values.

THE range of RaC' α particles (energy: 7.68 Mev) in water was reported by Philipp¹ in 1923 to be 13 percent shorter than what could be computed from the measured range in water vapor. This was supposed to be caused by the strong association of the water molecules in the liquid state. Nonassociative liquids such as benzene and pyridine showed similar effects of only 5 percent when measured in the same apparatus. Michl² in 1914 reported measurements with a photographic method on Po α -particle (energy: 5.30 Mev) ranges in several liquids. The range obtained for water was 32 microns, which is 17 percent less than the value 38.7 microns given by the Bragg law when one uses for hydrogen the stopping power value 0.224 and for oxygen 1.060 relative to air³ and a mean range in air of 38.42 mm at 15°C and 1 atmos.⁴ More recent measurements by Appleyard⁵ and de Carvalho⁶ on Po α particles gave 33.2 and 38.1 microns respectively as the range in water. Appleyard's value is 14 percent and de Carvalho's value 1.5 percent smaller than that predicted by Bragg's law. A redetermination of the α -particle range in water and other liquids would therefore seem to be of interest.

The new method⁷ is built on the following principle. The liquid is placed between two plane, parallel surfaces constituting the walls of a microcell. One wall is the support of an α -particle source located at its center and of negligibly small thickness while the other is a scintillation crystal. Those α particles which are sent out within a certain angle to the normal reach the scintillation crystal and give light flashes which are counted by a photomultiplier.

The counting rate *versus* the distance between the

surfaces is given by the following formula:

$$N = \int_x^\infty \frac{N_0}{2\alpha\sqrt{\pi}} \exp[-(R-R_0)^2/\alpha^2] \left(1 - \frac{x}{R}\right) dR, \quad (1)$$

where N_0 is the total number of disintegrations per unit time in the source, x the distance between the surfaces, R_0 the mean range, α the straggling parameter, and R the variable in the (Gaussian) straggling function $(1/2\alpha\sqrt{\pi}) \exp[-(R-R_0)^2/\alpha^2]$ assumed to be valid.

In the limit as $\alpha/R_0 \rightarrow 0$, i.e., for a vanishingly small straggling, Eq. (1) reduces to

$$N = \frac{1}{2}N_0(1-x/R_0) \text{ for } x \leq R_0 \text{ and } N=0 \text{ for } x \geq R_0, \quad (2)$$

i.e., N as a function of x tends to a straight line intersecting the x -axis at R_0 . For values of α of one or a few percent of R_0 , the same will be very nearly true for (1) except for x -values within $R_0 - \alpha$ and $R_0 + \alpha$, where the sharp bend at $x = R_0$ is replaced by a gradual change in the slope of the curve. The value of N at $x = R_0$ is to a good approximation given by $(N_0\alpha/4R_0\sqrt{\pi})$, from which α may be obtained.⁸

The experimental arrangement is shown schematically in Fig. 1. A piston of 7-mm diameter was ground to a very good fit into a cylinder body, both being made from a type of steel used for gauge blocks. The end surfaces of the cylinder and the piston were ground plane⁹ to within about one-tenth of a micron and controlled in a

⁸ It may be remarked that instead of (1), one may write in general:

$$N = \int_x^\infty \frac{1}{2}N_0S(R) \left(1 - \frac{x}{R}\right) dR, \quad (3)$$

where $S(R)$ is any straggling function. With given $N(x)$, one obtains $S(R)$ from

$$S(R) = (2x/N_0)(d^2N/dx^2). \quad (4)$$

With the aid of a differential discriminator, one may also select pulses under a certain amplitude for counting, resulting in a relation between the count rate n and the distance x of the form

$$n = \int_x^\infty CS(R) \frac{x}{R^2} dR; \quad C = \text{a constant} \quad (5)$$

and

$$S(R) = \frac{1}{C} \left(n - x \frac{dn}{dx} \right). \quad (6)$$

Equation (6) has the advantage over (4) that the derivative is of only first order.

⁹ C. E. Johansson, AB, Eskilstuna, Sweden.

¹ K. Philipp, Z. Physik 17, 23 (1923).

² W. Michl, Sitzber. Akad. Wiss. Wien, Math. naturw. Kl. 123, 1965 (1914).

³ L. H. Gray, Proc. Cambridge Phil. Soc. 40, 87 (1944).

⁴ M. G. Holloway and M. S. Livingston, Phys. Rev. 54, 18 (1938).

⁵ R. K. Appleyard, Nature 163, 527 (1949); Proc. Cambridge Phil. Soc. 47, 443 (1951).

⁶ H. G. de Carvalho, Phys. Rev. 78, 330 (1950); H. G. de Carvalho and H. Yagoda, Phys. Rev. 88, 273 (1952).

⁷ G. Aniansson, Report at the Instruments and Measurements Conference, Stockholm, 1952 [Arkiv Fysik 7, 74 (1954)].

Michelson interferometer. The parallelepipedal ($3 \times 3 = 15$ mm) CaWO_4 scintillation crystal was ground plane on one side to the same tolerance.¹⁰ The crystal is slid on to the cylinder end surface to close contact which could be controlled by the light interference fringes that appear. The upper surface of the crystal is coarse-ground to diminish the trapping of light through total reflection. A glass-covered conical light collector brings the light to the photocathode of an EMI 6260 multiplier tube and serves as a container for the liquid which has free access to the space between the crystal and the piston (the piston diameter is 7 mm, the crystal breadth 3 mm).

The piston is moved by a lever connected to the cylinder and the piston by two plane metal springs. The displacement is read on a "mikrokator"¹¹ calibrated to within 0.05 micron.¹² The distance between the piston and cylinder end surfaces at the zero point of the "mikrokator" was determined with a second "mikrokator." The crystal, light collector and photo-multiplier were removed during this operation. The polonium α source was evaporated from a platinum support on to the center of the piston end surface. An autoradiograph showed that the polonium was evenly deposited on a circular area of about 1 mm diameter in the center of the piston end surface.

The pulses from the multiplier were amplified 30 times, passed through a discriminator which excluded pulses under a certain amplitude—among them thermionic pulses from the phototube—and counted with a scaler. The higher the discriminator level is, the longer must the α particle travel in the scintillation crystal to produce the necessary light amount. This results in a correspondingly smaller apparent range, and thus range measurements were made with different discriminator levels and the ranges obtained were extrapolated to zero discriminator level.

Figure 2 shows the results obtained with absolute ethyl alcohol with discriminator settings of 5, 10, 15, 20, and 40 volts. The curves are straight lines except in a small region near the apparent range as predicted by (1). The plot of apparent range *versus* discriminator level is also a straight line, which facilitates the extrapolation to zero level.

With the same crystal, light collector, and counting equipment, the apparent mean range in dry air was determined for different discriminator levels and extrapolated to zero level. The result, obtained at 25.0°C and 748.4 mm Hg, was 39.80 mm which at the normal temperature 15°C and pressure 760 mm Hg would correspond to 37.87 mm. This is 0.55 mm or 1.4 percent

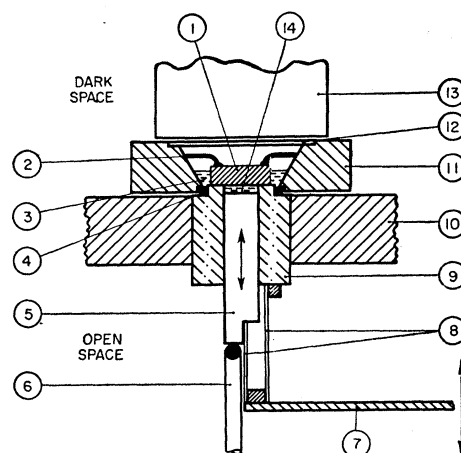


Fig. 1. The experimental arrangement. (1) Scintillation crystal, CaWO_4 , $3 \times 3 \times 15$ mm. (2) Metal spring pressing the crystal against the cylinder end surface. (3) Liquid. (4) Gasket. (5) Piston. (6) "Mikrokator" measuring arm. (7) Lever. (8) Plane metal springs. (9) Cylinder. (10) Base plate. (11) Light collector and liquid container. (12) Glass plate. (13) EMI 6260 photo-multiplier tube. (14) Po α source, diameter 1 mm.

shorter than the accepted standard value 38.42 mm obtained with ionization methods and is caused partly by the thin aluminum layer which for reasons given below was evaporated onto the middle part of the ground surface of the crystal. The rest of this discrepancy may be caused by a thin insensitive surface layer which has probably melted during the grinding process.

According to present theories^{13,14} of the scintillation process in solutions in liquid, the energetic particle gives off its energy to the solvent from which by some mechanism it is transferred to the solute molecules. If this transfer mechanism were efficient over distances of some tenths of a micron for the particular system,

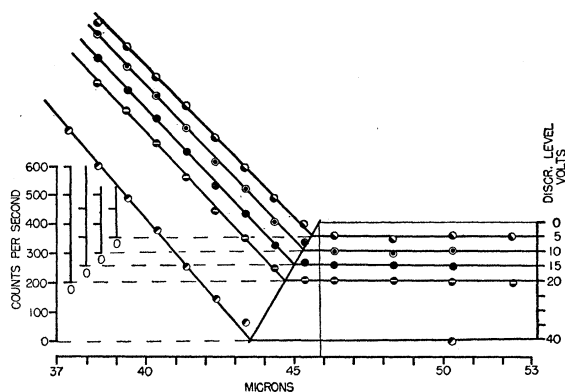


Fig. 2. Counting rate *versus* distance curves for absolute ethyl alcohol. The parallel, vertical displacements of the curves are proportional to the discriminator level.

¹⁰ Junger, AB, Stockholm, Sweden.

¹¹ Type 509-7, manufactured by C. E. Johansson, AB.

¹² When still higher accuracy is sought, interference or other optical methods may be used. The distance between the piston end and the crystal surface might for example be directly measured through the interference of monochromatic light rays reflected at the two surfaces.

¹³ S. C. Curran, *Luminescence and the Scintillation Counter* (Butterworths Scientific Publications, London, 1953).

¹⁴ J. B. Birks, *Scintillation Counters* (Pergamon Press, London, 1953).

TABLE I. Experimental values, corrections, and comparison with the Bragg law.

Liquid	Apparent mean range extrapol. to zero discr. level microns	Corrected mean range (average) microns	Density g cm ⁻³	Stopping power exptl	Stopping power theor ^a	$\frac{S_{\text{theor}}}{S_{\text{exptl}}} = \frac{R_{\text{exptl}}}{R_{\text{theo}}}$
Water	39.25	39.88	0.9969	1.469	1.508	1.027
	39.25					
	39.17					
Ethyl alcohol	45.85	46.53	0.7866	4.07	4.198	1.030
	45.90					
Benzene	43.40	44.09	0.8727	6.56	6.714	1.023
	43.55					
Pyridine	40.40	40.97	0.9775	6.39	6.585	1.031
<i>n</i> -heptane	49.90	50.48	0.6790	9.46	9.849	1.042
	49.65					
	49.75					
Iso-octane (2,2,4-trimethyl pentane)	49.40	50.27	0.6871	10.70	11.19	1.046
	49.75					

^a $s_0 = 1.060$, $s_H = 0.224$, and $s_c = 0.895$ according to reference 3.

^b From a different source.

liquid—CaWO₄, it would lead to a noticeable increase in the apparent range. Measurements were therefore made on two liquids, water and benzene, (a) with the crystal uncovered and (b) with the crystal covered by a thin but opaque aluminum film which would greatly reduce any energy transfer from the liquid to the crystal. The ranges found under (a) were 0.3 to 0.5 micron longer than those found under (b). Allowing for the contributing effect of the aluminum film, the differences become still smaller and completely within the limits of experimental error. For safety the film was, however, allowed to remain in the subsequent measurements and all the results reported have been obtained with this film on the crystal.

There is thus only one correction to apply to the "zero-level" ranges obtained, i.e., the one caused by the thin aluminum film and the insensitive layer on the crystal surface. In air the reduction in range caused by these layers is 1.4 percent and the same reduction applies for the ranges in the liquids provided the alteration with α -particle energy of the stopping power of the liquids relative to air may be neglected. There is no reason to take such an alteration into account in this case since the correction is almost as small as the experimental error.

In Table I are given the results obtained for six liquids at 25°C. The densities were determined with a pycnometer at the same temperature. The stopping power was computed from the formula

$$s = (38.42 \times 1.226 \times 10^{-3} / 14.56) \times (M / \rho R_0), \quad (7)$$

where 38.42 mm is the standard range in air of density 1.226×10^{-3} g cm⁻³ and mean atomic weight 14.56, M is the molecular weight of the liquid, ρ its density in g cm⁻³, and R_0 is the corrected range in the liquid in mm.

It is seen that the largest difference between any two experimental values for the range for the same liquid is less than 1 percent which is the assumed value for the uncertainty in the measurements.

The ranges are all *longer* than those computed from the Bragg law and measurements on gases, thus giving the impression of a general density effect.¹⁵ In contrast to this the aforementioned results by Michl, Philipp, Appleyard, and de Carvalho were all *lower* than the values predicted by the Bragg law. The differences between their results for water and that in the present work are 20 percent, 16 percent, 17 percent, and 4 percent respectively, clearly outside the assumed limit of accuracy of the present method.

Of the six compounds listed in Table I, only water has been subject to range measurements in the gaseous state with Po α particles. For water vapor, Förster¹⁶ obtained a 3 percent longer range than that predicted by the Bragg law, in agreement with the present result, whereas de Carvalho and Yagoda⁶ in a single measurement obtained a range 2.5 percent lower than that predicted by the Bragg law, in approximate agreement with Philipp's¹ result for RaC' α particles.

ACKNOWLEDGMENTS

The author wishes to express his thanks to the head of the division, Professor Ole Lamm, for suggesting and supporting this investigation. Thanks are also due to the members of the division for valuable discussions and other aid. The investigation was financed by the Swedish Atomic Committee.

¹⁵ R. M. Sternheimer, Phys. Rev. **93**, 351 (1954).

¹⁶ M. Förster, Ann. Physik **27**, 372 (1936).