Conference on Formation and Properties of Gas Bubbles

THE PROPERTIES AND BEHAVIOUR OF GAS BUBBLES FORMED AT A CIRCULAR ORIFICE

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Introduction

There is a variety of operations and processes in industry in which gas as a dispersed phase plays an important part; and it is the purpose of this symposium to bring together such data as are available upon the properties and behaviour of such a phase and to indicate by some practical examples its role in mechanical, physical and chemical processes. In order to define the scope of the subject more closely, it will be convenient at the outset to adopt a rough classification based upon the functon of the dispersed phase in various types of system, as follows:

Dispersed Phase.

- *Liquid.* Foam.
- *Gas.* Bubbles.
  - *Mechanical.* Air Lift Pump. Air Agitator.
  - *Physical.* Ore Flotation. Aeration. Liquid Separation by phase change.
  - *Chemical.* Gas Separation by chemical reaction. Gas Absorption and Reaction (*e.g.*, Hydrogenation).

As a general rule, in operations employing gas as a dispersed phase, the size and shape of bubbles, the properties of interfaces and the relative motion of the two phases are matters requiring consideration; and, in this, the introductory paper of the Symposium, two examples will be discussed in which mechanical work and gas absorption processes, respectively, are involved.

Probably the best-known practical application of a dispersed gas phase for performing work is that represented by the air-lift pump. Although this simple device was first described some 200 years ago, it was not until the spectacular growth of the oil industry about the middle of the 19th century that it came into extensive use as a convenient and economic method of pumping large quantities of viscous liquid. An account of its operation is to be found in an early patent by Frizell[1] (1880) and a more detailed discussion of the underlying theory in a later patent by Pohle[2] (1886). Since that date, numerous investigations have been carried out with a view to improving its design and performance.

In principle, the pump consists of a U-tube, one arm of which contains homogeneous liquid and the other, the rising main, a less dense two-phase system of air and liquid. When the two columns are balanced, there will be a difference of level in the arms due to the difference in density. The aeration of the liquid is effected in a footpiece which in most modern design takes the form of a perforated cylinder or cone inserted into the rising main.

It will be evident that to design the footpiece so as to give an optimum size and distribution of bubbles a knowledge is required of the mechanism of bubble formation, the velocity with which they rise in a column of still liquid as a function of their size and the properties of the liquid and the slip between bubble and liquid when both are travelling in the same direction. Data of a similar kind are needed for the design of air-agitating plant and for carrying out the various physical and chemical processes mentioned above.

Whilst a considerable amount of data exists on the properties of bubbles, it is by no means complete or even concordant and it has, therefore, been found necessary in some instances to repeat the earlier work and to extend the range of observations.

1. The Mechanism of Bubble Formation

(*a*) *The relation between the volume of a bubble and the size of the orifice at which it is formed*

The formation of a bubble is associated with the growth of an interface in an environment subjected to the hydraulic pressure of a head of liquid; the gas under pressure is usually introduced through a conduit, which in the simplest case terminates in a circular orifice, and forms a succession of bubbles which break away from the solid—liquid—gas interface and thereafter travel as separate entities in the liquid. There are two cases requiring consideration, namely, (1) that in which the bubble is formed at such a rate that its buoyancy can be balanced against the surface forces tending to hold it on the orifice, and (2) that in which the velocity of the gas passing through the orifice is such as might give rise to a pressure within the bubble at the moment of release greater than that corresponding with the hydrostatic head of the liquid and its surface tension. In the first case, there will be a simple relationship between the sizes of bubble and orifice; in the second, the size of the bubble may depend *inter alia* upon the velocity of the gas stream and upon viscosity, momentum and frictional effects.

Case I—Low gas velocity

The measurement of the maximum pressure required to liberate a bubble from a capillary orifice has been used as the basis of a method for determining the surface tension of liquids and accounts of the theory and application of the method have been given by Cantor, Ferguson, Schroedinger, Sugden and others. (For references see Sugden, J.C.S., 1922, 858.) In carrying out a determination, the orifice is orientated vertically downwards and the hydrostatic head of liquid is kept small. In these circumstances, the bubbles from orifices of even small diameter tend to be distorted by buoyancy forces holding them against the walls and corrections are necessary for their departure from sphericity. In the air-lift pump the arrangements are somewhat different, the orifices normally facing sideways or vertically upwards and the hydrostatic head of liquid being large. Bubbles formed under these conditions tend to preserve a spherical shape at the moment of detachment up to much larger sizes than in the previous case (*cf.* Plate 1 (*d*)). It is thus possible to obtain a simple approximate relationship between the volume of bubble and the size of orifice.

If the bubble is assumed to be spherical and of radius R at the moment of release, then, if the axis of the circular orifice at which it is formed is vertical, the buoyancy force acting upon it is given by $\frac{4}{3}\pi R^3\rho g$, where ρ is the density of the liquid (or more accurately the difference between the densities of liquid and gas). This force is balanced by a surface tension force, $2\pi r\gamma(\cos\theta) f\left(\frac{r}{a}\right)$, where r is the radius of the orifice, γ is the surface tension of the liquid, θ is the angle of contact at the triple interface and $f\left(\frac{r}{a}\right)$ is a shape factor which, for a sphere, has the value 1. (*Note :* a is the square root of the capillary constant $\left.\frac{2\gamma}{\rho g}\right)$.

If it be further assumed that there is perfect wetting of the orifice by the liquid, *i.e.*, $\theta = 0$, the two forces may be equated :

$$\frac{4}{3}\pi R^3\rho g = 2\pi r\gamma$$

from which

$$R = \left(\frac{3}{2}\frac{r\gamma}{\rho g}\right)^{1/3} \quad \ldots\ldots\ldots\ldots (1)$$

In the case of a system air-water at 20° C.

$$r = 9{\cdot}05\ R^3$$

$$\text{or } \frac{V}{D} = 0{\cdot}231$$

where V is the volume of the bubble and D the diameter of the orifice.

At this point, attention may be drawn to two further assumptions implied in Equation (1), namely, that no forces other than buoyancy contribute to the release of the bubble and that the surface tension of an expanding surface is approximately that of the corresponding static or equilibrium value. It may also be noted that the bubble does not necessarily rest on the upper edge of the orifice until the moment of release but may develop a neck which retreats into the gas channel somewhat as shown in Plate 1. (*a*) and (*b*) show two stages of bubble growth and release.

It is apparent that, in cases in which the liquid wets the material from which the orifice is constructed, the thickness of wall of the orifice will be without influence on the size of the bubble produced.

There is little experimental data in the literature to permit of a rigorous test of Equation (1). Maier[3] has carried out a number of experiments on small orifices, usually not exceeding 0·07 cm. in diameter, and Owen[4] and Swindin[5] record a few measurements for orifices of diameters in the range 0·03—0·95 cm. The results, where comparison is possible, differ so widely as to make a repetition of the work desirable. We have, therefore, carried out a series of experiments with orifices of diameter from 0·022—0·519 cm., thus covering the range of sizes used by the workers mentioned above. The orifices employed consisted of selected glass capillary tubing cut and ground flat, but not polished. Each orifice was carefully examined and only those showing a circular section free from irregularities were employed. The volume of a bubble was obtained by collecting and measuring the gas from a given number of bubbles. The values so obtained were checked by a photographic method in the case of bubbles small enough to be taken as spherical. Before each series of determinations, gas was bubbled through the liquid for at least 1 hour to ensure equilibrium in the liquid phase.

The results are summarised in Table I, which also contains, for comparison, the values recorded by Maier, Owen and Swindin. They are also shown graphically in Fig. 1.

TABLE I

Showing relation between the sizes of bubbles and the orifice at which they are formed

Diameter D (cm.)	Volumes (ml.) Maier[3]	Owen[4]	Swindin[5]	Present work	$\frac{V}{D}$
·0118	·0022				0·18
·022	·0052			·0038	0·23/0·17
·030		·0011			0·04
·0349	·0023				0·07
·04				·013	0·43
·043	·0445		·0036		1·0/0·09
·070	·0602				0·8
·095				·032	0·34
·132				·045	0·34
·179				·054	0·30
·211				·064	0·30
·296				·100	0·34
·345		·0168			0·05
			·0220		0·06
·450				·140	0·31
·519				·228	0·44
·538		·134			0·24

It will be observed that, whereas the results of previous workers give widely varying values for the ratio $\frac{V}{D}$, those by the authors give an approximately constant ratio, the average value being 0·33 as compared with 0·231 from Equation (1).

The orientation of the orifice.—It will be recalled that in the derivation of Equation (1) it was assumed that the axis of the nozzle was vertical. If the axis be inclined to the vertical, the surface tension forces are operative only around a portion of the perimeter, the buoyancy forces tending to drag the bubble upwards across the plane of the orifice. The effect is shown in Plate I (*c*) and (*d*), which show two stages in the growth of the bubble. It would be expected, therefore, that the size of bubble from a vertically orientated orifice would be greater than from a similar inclined orifice, and that this is the case may be seen from the curves on Fig. 2, which show bubble size as a function of orifice diameter for horizontal and vertically orientated orifices.

The shape of gas bubbles formed at an orifice and rising in a column of liquid

Before considering Case II, attention may be drawn to some properties of bubbles as a function of their size. The following phenomena are noticed when streams of bubbles are produced from orifices orientated vertically upwards and of progressively increasing diameters :

(1) For circular orifices up to 0·04 cm. diameter, the bubbles are substantially spherical and after an initial acceleration on release travel upwards at a uniform velocity, following a vertical path.

(2) For orifices between 0·04 and 0·4 cm. diameter, the bubbles are spherical at the orifice, but on release rapidly assume an ellipsoidal shape with the longer axis horizontal. In this form they travel upwards, following a zig-zag path.

Miyagi[6] disagrees with this observation and assigns the bubbles a helical path which he attributes to harmonic oscillations within the bubble producing periodic changes in the inclination of its axis as it passes through the static spherical configuration. The observation made by the authors is that once the bubble has assumed the stable ellipsoidal form, its shape does not change appreciably, but it rocks to and fro about its shorter axis during ascent. The constancy of shape may be seen from Plate I (*e*), which shows a photograph of a stream of bubbles of air from an orifice 0·388 cm. in diameter, rising through a column of glycerine at a rate of 25 bubbles per minute.

(3) With orifice diameters exceeding 0·4 cm., the bubbles become unstable. They may assume a symmetrical saucer shape as in Plate I (*f*), which relates to an orifice of 0·432 cm. diameter, the bubbles rising in water at the rate of 25 per minute ; or, more frequently, an unsymmetrical shape as in Plate I (*g*). In the latter case, the size of bubble varies in a random manner, and their path becomes irregular ; for the larger sizes, disruption may occur, with the formation of small satellite bubbles.

(4) When the ratio, diameter of bubble : diameter of liquid column exceeds 0·75, the bubbles assume a cylindrical shape with an ogival head and a flat tail. The condition then approaches that seen in the Pohle air-lift, in which alternate pellets of gas and liquid rise in the eductor.

The limiting conditions for the three characteristic modes of bubble ascent are dependent upon the properties of the liquid and, in particular, its viscosity. Thus, for very viscous liquids, the bubbles tend to preserve symmetrical shapes up to much larger sizes than is the case with water and their path tends to remain vertical.

As the bubbles rise through the liquid, they expand with diminution of the hydrostatic pressure and, in consequence, a bubble having a stable spherical or ellipsoidal form near the orifice may, on rising, expand into the region of instability.

Case II.—High gas velocity

As the rate (and pressure) of gas supply to an orifice is increased, the rate of bubble formation also increases and it might be anticipated that momentum and frictional effects would then become important and that the size of the bubble formed would, in consequence, be some function of the gas pressure. Maier, indeed, has concluded, from experimental observations covering a wide range of gas pressures, that there is no definite bubble size for a given orifice. This statement appears to require modification, both in the light of his results and of our own experiments. Maier has, in fact, postulated a form of equation for bubble size which requires it to pass through a maximum or minimum value with increasing rate of formation. Eversole, Wagner and Stackhouse[7] have also suggested that a part of the kinetic energy of the gas stream emerging into an expanding bubble may be converted into pressure energy which would help to inflate the bubble.

We have found that, for orifices of diameters between 0·04 and 0·4 cm., the size of bubbles formed at first diminishes with increasing rate, passes through a minimum and thereafter slowly increases up to a rate at which random size distribution sets in. This critical rate is approximately 500 bubbles per minute for orifices up to 0·04 cm. diameter and about one-half this rate for orifices of 0·04—0·4 cm. diameter. The magnitude of the effect may be seen from the data given in Table II, which relates to orifices of four different sizes and rates of from 1 to 125 bubbles per minute. Maier's results are included in the Table for comparison.

It has also been noted that for bubbles in this range of sizes

TABLE II

Effect of rate of bubbles/min. on the volume at different nozzle diameters

Liquid column = 4·44 cms. in diameter.
Hydrostatic head = 121·9 cms.
Liquid = water.

	Diameter of nozzle = 0·022 cm.			Diameter of nozzle = 0·036 cm.			Diameter of nozzle = 0·141 cm.			Diameter of nozzle = 0·388 cm.		
Number of bubbles/min.	Volume of bubble × 10 − 4 (cc.)	Rate of airflow × 10 − 4 (cc./sec.)	Reynolds number	Volume of bubble × 10 − 4 (cc.)	Rate of airflow × 10 − 4 (cc./sec.)	Reynolds number	Volume of bubble × 10 − 4 (cc.)	Rate of airflow × 10 − 4 (cc./sec.)	Reynolds number	Volume of bubble × 10 − 4 (cc.)	Rate of airflow × 10 − 4 (cc./sec.)	Reynolds number
1	24·3	0·40	1·21	81·0	1·35	6·75	307·5	5·13	60·12	995·2	15·92	514
5	10·8	0·90	2·70	62·1	5·17	25·88	307·0	25·59	302·60	990·6	82·60	2667
9	11·7	1·75	5·26	43·1	6·46	32·28	304·9	45·71	537·00	990·1	148·60	4797
25	17·7	7·38	22·13	58·0	24·15	120·80	262·2	109·10	1282	983·5	410·20	13240
60	36·1	36·13	108·10	69·0	69·02	345·10	277·3	277·3	3258	941·6	941·90	30430
90	40·0	59·98	179·90	82·1	123·01	615·20	281·4	421·80	4955	963·1	1443	46670
125	48·6	101·25	303·40	99·2	206·50	1033·00	291·5	606·10	7129	982·5	2046	66070

Maier's Results

	Diameter of nozzle = 0·023 cm.
Number of bubbles/min.	Volume of bubble (cc.)
8	·0091
15	·0042
32	·0067
38	·0130

there is a rapid acceleration of velocity immediately on release to a value in excess of the terminal velocity, after which a retardation occurs. This phenomenon has not been investigated and further work is required in order to determine its cause.

In a further series of experiments, the results of which are summarised in Table III, it will be seen that hydrostatic head has very little influence on the relation between rate of formation and the size of bubble.

TABLE III

Volume of bubbles as a function of rate of formation at various hydrostatic heads

Orifice diameter = 0·141 cm.

Number of bubbles formed per min.	$10^{-4}\times$ volume of bubble (cc.) at the orifice for hydrostatic heads of:			
	cms. 213·3	cms. 121·9	cms. 91·22	cms. 60·95
1	308·6	307·5	305·2	306·8
5	307·3	307·0	308·2	305·4
9	300·1	304·9	303·3	301·6
25	268·9	262·2	263·8	266·3
60	271·3	277·3	281·9	278·6
90	283·9	281·4	286·0	282·7
125	300·1	291·5	293·4	308·2

The Pressure inside a bubble at the moment of release

The pressure, P_1, inside a bubble at any moment during its formation at a capillary orifice will be related to the pressure, P_2, of the gas supply, to the pressure, P_3, in the liquid at rest at the orifice and to the pressure, P_4, of the liquid undergoing acceleration at the interface.

The following relations are found to hold, assuming perfect wetting:

(*a*) when the meniscus is inside the capillary orifice

$$P_2 - P_3 < \frac{2\gamma}{r} \quad \text{.....(2)}$$

(*b*) when the bubble is expanding outside the orifice

$$P_1 - P_3 > \frac{2\gamma}{R} \quad \text{.....(3)}$$

$$P_1 - P_3 = \frac{2\gamma}{R} + (P_4 - P_3) \quad \text{.....(4)}$$

In general, the term $(P_4 - P_3)$ can be neglected.

During expansion, $\frac{2\gamma}{R}$ will increase as R diminishes, whilst P_1 will differ from P_2 by the pressure drop in the capillary. When the bubble has a hemispherical shape $(R = r)$, $\frac{2\gamma}{R}$ will be a maximum and the condition will be one of instability. It can be seen from Plate I (*b*) that, during the period of growth of the bubble and whilst $(P_1 - P_3)$ is increasing, the bubble develops a concave neck which eventually collapses and releases the bubble from the orifice. The exact relationship between γ and the maximum pressure in a bubble of shape other than spherical is given by Sugden (*loc. cit.*).

The Influence of surface tension on bubble formation

From Equation (1) we can write:

$$\frac{V}{D} = \frac{\pi\gamma}{\rho g} \quad \text{.....(5)}$$

and hence the volume of a bubble formed at a submerged orifice should, *ceteris paribus*, increase with the surface tension of the liquid.

In order to test this relationship experimentally, it is necessary to find a series of liquids differing widely in surface tension but having approximately the same densities. For this purpose, we have employed water, ethyl alcohol and various aqueous alcohol mixtures, with results recorded in Table IV.

TABLE IV

Relation between diameter of orifice and volume of bubble as a function of surface tension

Rate = 25 bubbles/minute; liquid head = 150 cms. water; temperature = 12–16° C.

Corrected at normal pressure.

D = nozzle diameter.
V = volume of bubble at the tip.

Properties of Medium			Dia. of nozzle = ·036 cm.		Dia. of nozzle = ·060 cm.		Dia. of nozzle = ·097 cm.		Dia. of nozzle = ·141 cm.		Dia. of nozzle = ·198 cm.		Dia. of nozzle = ·270 cm.		Dia. of nozzle = ·388 cm.		Dia. of nozzle = ·432 cm.		Dia. of nozzle = ·520 cm.		Dia. of nozzle = ·630 cm.		$\frac{V}{D}$ calculated
Percentage of ethyl alcohol in water (by weight)	Surface tension (dynes/cm.)	Density (gm./cc.)	Volume of bubble (cc.)	$\frac{V}{D}$ observed	Volume of bubble (cc.)	$\frac{V}{D}$ observed	Volume of bubble (cc.)	$\frac{V}{D}$ observed	Volume of bubble (cc.)	$\frac{V}{D}$ observed	Volume of bubble (cc.)	$\frac{V}{D}$ observed	Volume of bubble (cc.)	$\frac{V}{D}$ observed	Volume of bubble (cc.)	$\frac{V}{D}$ observed	Volume of bubble (cc.)	$\frac{V}{D}$ observed	Volume of bubble (cc.)	$\frac{V}{D}$ observed	Volume of bubble (cc.)	$\frac{V}{D}$ observed	by $\frac{V}{D} = \frac{\pi\gamma}{\rho g}$
0 (*i.e.*, water)	73·10	·9997	·0072	·200	·0132	·210	·0198	·210	·0292	·200	·0399	·210	·0630	·220	·0984	·240	·0994	·224	·0988	·190	·1071	·170	·234
1·96	64·6	·9954	·0070	·203	·0110	·184	·0160	·170	·0237	·170	·0360	·183	·0600	·210	·0960	·230	·0980	·220	·0960	·190	·0990	·160	·207
9·09	49·96	·9844	·0030	·090	·0060	·100	·0140	·144	·0199	·141	·0330	·162	·0570	·204	·0930	·220	·0955	·212	·0940	·180	·0985	·155	·162
33·30	32·39	·9518	·0022	·065	·0040	·070	·0070	·072	·0129	·089	·0261	·120	·0520	·190	·0920	·215	·0935	·210	·0920	·160	·0960	·150	·109
100·00	22·88	·7936	·0018	·053	·0020	·043	·0040	·042	·0126	·088	·0230	·110	·0480	·160	·0890	·210	·0920	·190	·0910	·154	·0955	·140	·092

c

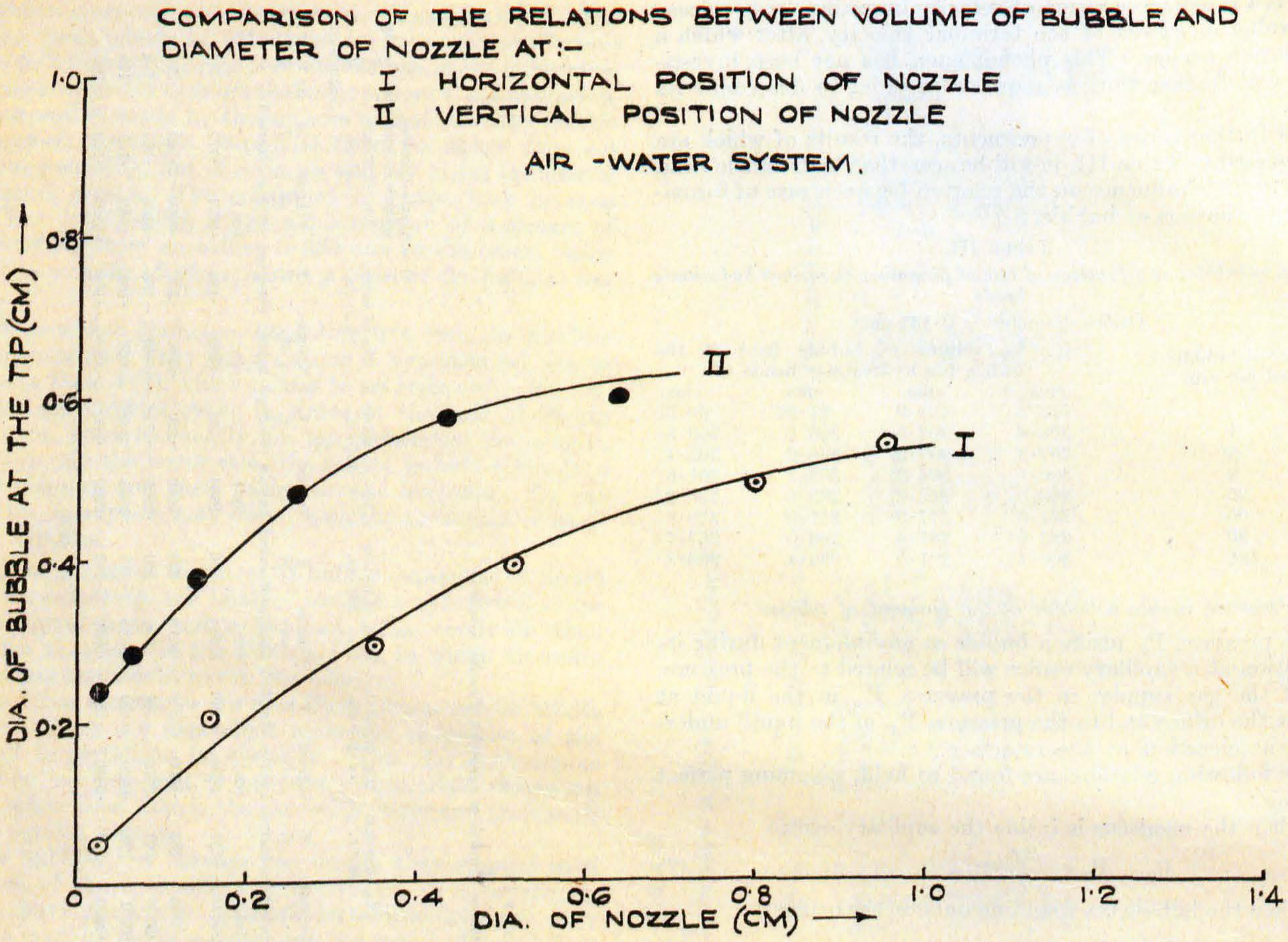


FIG. 1

It will be noted that the values of $\frac{V}{D}$ are reasonably constant for all sizes of orifice and increase with γ in accordance with Equation (5). The relationship between bubble volume and surface tension may be seen from Fig. 2 to be nearly linear over the whole range of surface tensions.

The Influence of viscosity on bubble formation

A number of investigations upon the influence of viscosity on the size of bubbles formed at orifices have been described. Schnurmann[8] measured the size of bubbles produced from porous earthenware and carbon filters immersed in a variety of liquids, including alcohols, acids, sugar solution, and certain electrolytes, and concluded that viscosity was the principal factor determining size. He failed, however, to take account of surface tension variations which have been shown to be an important contributory factor.

Halberstadt and Prausnitz[9], on the other hand, experimenting with ether, aqueous glycerine, aqueous alcohol, Turkey red oil, toluene and other liquids, concluded that surface tension, rather than viscosity, was the property determining bubble size.

We have re-examined the problem and, using a series of aqueous glycerine solutions having a wide range of viscosities, have shown that viscosity, *per se*, plays very little part in determining bubble size. For orifices of diameter 0·036–0·63 cm., a hundredfold increase in viscosity causes a diminution in volume of about 10 per cent.

The experimental data are summarised in Table V.

The Terminal velocity of bubbles

It might be expected that no great difficulties would be encountered in measuring experimentally the terminal velocities of bubbles rising in a column of liquid. We shall see, however, that, in spite of numerous investigations, the

TABLE V.

The Influence of viscosity on the size of bubbles formed at submerged orifices

Properties of liquid medium				Volume of bubble (cc.) formed at orifices of diameter :									
Percentage of Glycerine (by weight) in water	Viscosity (poise)	Surface tension (dynes/cm.)	Density (gms./cc.)	·036 cm.	·060 cm.	·097 cm.	·141 cm.	·198 cm.	·270 cm.	·388 cm.	·432 cm.	·520 cm.	·630 cm.
0	·012	72·8	·9994	·0072	·0132	·0198	·0292	·0399	·0630	·0984	·0994	·0988	·1071
65·0	·154	68·3	1·1700	·0700	·0110	·0170	·0250	·0350	·0570	·0870	·0900	·0900	·0950
70·5	·235	67·6	1·1850	·0700	·0100	·0170	·0245	·0340	·0550	·0870	·0870	·0880	·0930
79·0	·497	66·4	1·2100	·0068	·0100	·0150	·0231	·0332	·0550	·0850	·0850	·0860	·0930
65·0	1·108	65·7	1·2200	·0068	·0080	·0150	·0220	·0320	·0540	·0840	·0850	·0860	·0900

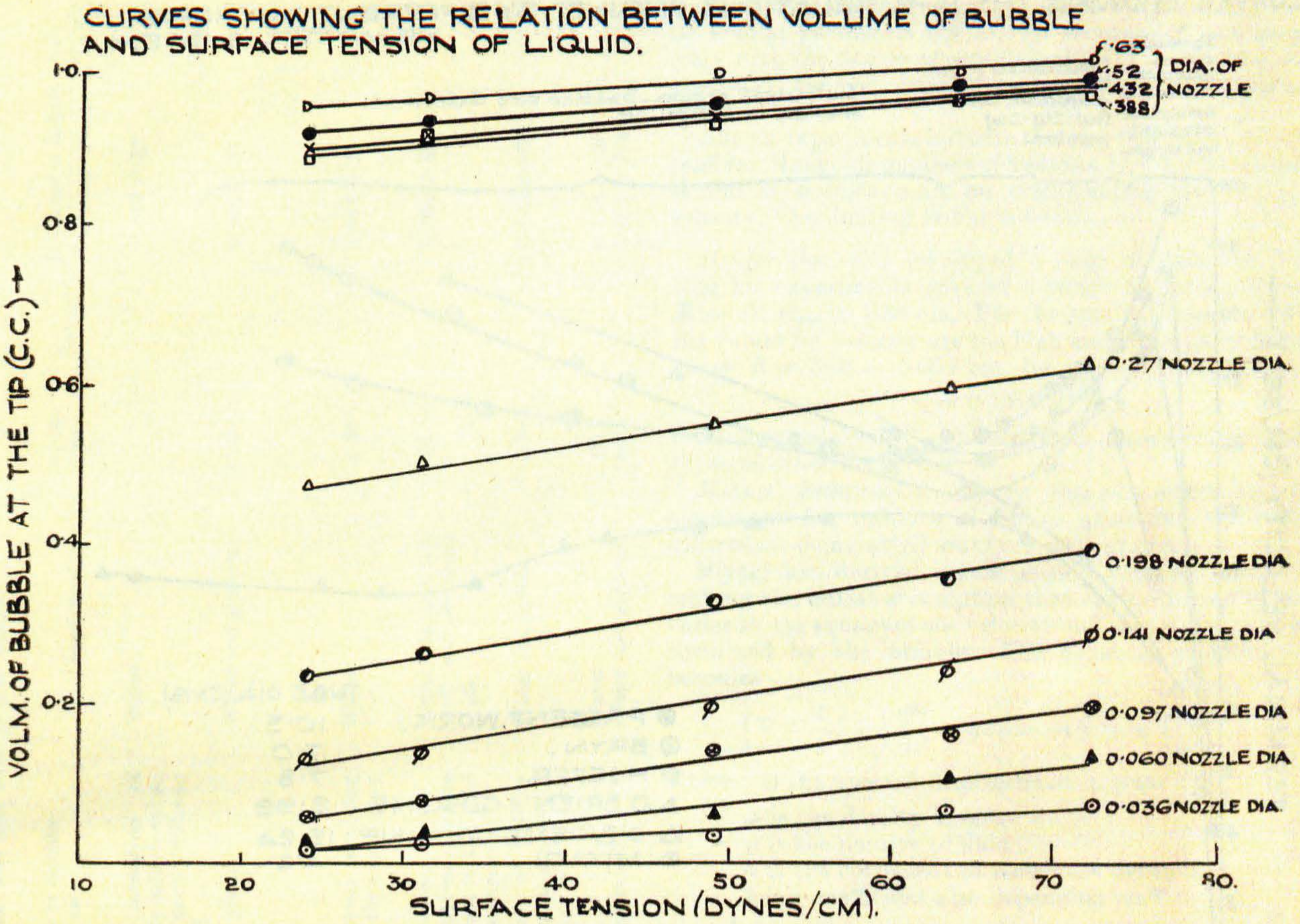


FIG. 2

results recorded in the literature show a variation greater than can be accounted for by the experimental errors inherent in the methods employed.

Experimental results

Measurements of terminal velocities have been made by Allen[11], Bryn[12], Hoefer[13], Luchsinger[14], Miyagi[6], O'Brien and Gosline[15], and the authors, under the conditions shown in Table VI.

The results are shown graphically in Fig. 3. It will be noted that, except in the case of O'Brien and Gosline's results, the values of v_t at first increase rapidly with bubble size to a maximum, diminish to a minimum and thereafter increase slowly with increasing bubble size. The position of the maximum varies somewhat, both in respect of velocity and size in the different investigations.

The factors which may be expected to influence the position and shape of the v_t — R curve are:

(1) *Temperature*, which alters the viscosity and surface tension of the liquid. Thus, for example, in the case of water, the viscosity between 10° C. and 20° C. diminishes by about 23%.

(2) *Wall effect*. There is abundant evidence that the ratio $\frac{d}{De}$ has a marked effect on velocity. It will be seen from Table VI that, in the investigations referred to the diameter of the liquid columns employed varied over a wide range.

(3) *Turbulence*. The curves in Fig. 3 cover ranges of streamline and turbulent motion, the boundaries of which cannot be accurately demarcated. An attempt has, however, been made on the diagram to indicate approximately the regions corresponding with the two types of motion.

(4) *Measurement of size of bubble*. There is reason to suppose that accuracy in the measurement of bubble size has not always been achieved. Thus, Hoefer (*loc. cit.*) and Owen (*loc. cit.*) have employed methods which are not capable of very great precision and the scatter of Hoefer's points in the range of small bubble size is probably attributable to errors arising from this cause. There is also evidence to suggest that, during the passage of bubbles up the column of liquid, some solution of gas may have occurred. Allen took special precaution to saturate the liquid before taking observations. Other workers do not always appear to have paid much attention to this factor.

(5) *Velocity*. No great difficulty appears to have been met with in the determination of velocity by direct timing of the bubbles. There is, however, an error in this measurement arising from the fact that, except in the case of very small and very large sizes, the bubbles do not rise vertically in the liquid column, but take a zig-zag or spiral path. The instantaneous velocities are, therefore, usually greater than the average velocity which is recorded.

The following observations relate to the several investigations:

Allen (*loc. cit.*) employed a tube of diameter 3 cm., and the ratio $\frac{\text{Diameter of bubble}}{\text{Diameter of tube}}$ included in his experiments covered the range ·005 — ·03. Over this range the bubbles appeared to have a rigidity similar to that of a solid particle.

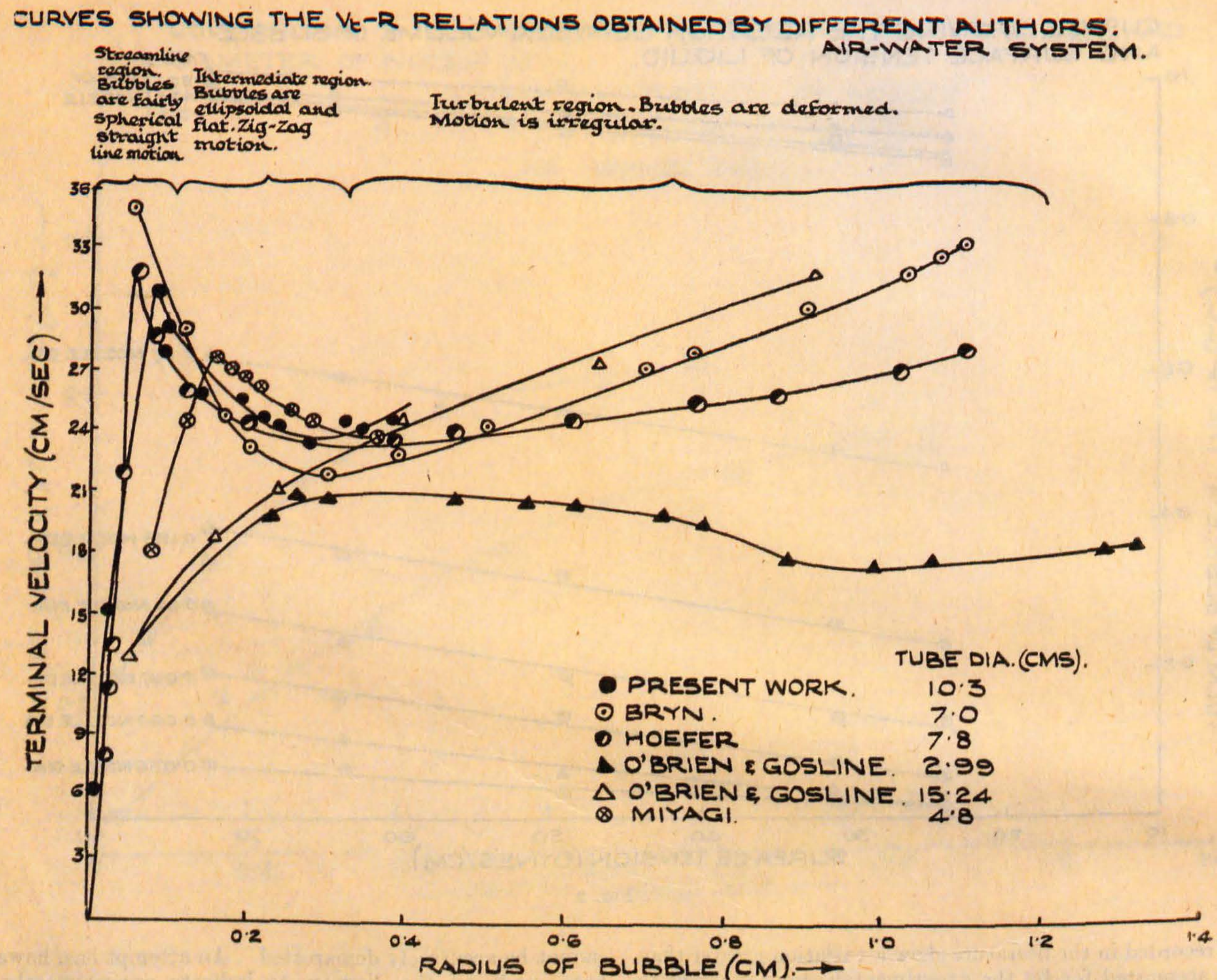


Fig. 3

In the derivation of Stoke's equation, it is assumed that there is no slip at the surface of a solid sphere falling through water. In the case of a gas bubble rising through a liquid, there will, in general, be a finite velocity of the liquid on the outside of the boundary envelope due to entrainment, and this will result in an increased value of the terminal velocity. According to Hadamard[16] and Rybczynski[17], the true terminal velocity will be given by

$$v_t = \frac{2}{9}R^2 g \frac{(\rho - \rho')}{\eta}\left[\frac{3\eta' + 3\eta}{3\eta' + 2\eta}\right] \quad \ldots\ldots\ldots\ldots\ldots\ldots(6)$$

the liquid and gas being considered as isotropic media of viscosities, η and η', respectively. ρ' is the density of the gas.

The expression in squared brackets corrects for the effect on velocity of internal circulation due to viscous drag, has a value of approximately 1·5 when η/η' is small and is independent of the radius of the bubble.

Allen proposed a somewhat similar modification of Stoke's equation, namely :

$$v_t = R^2 g \frac{(\rho - \rho')}{\eta}\left[\frac{\beta R + 3\eta}{\beta R + 2\eta}\right] \quad \ldots\ldots\ldots\ldots\ldots(7)$$

where β = a coefficient of sliding friction.

The expression in squared brackets has a value of approximately 1 for large values of β and a value of 1·5 when $\beta = 0$.

Bouissinesq[18], starting with the assumption that the viscosities of the gas and liquid are not isotropic in the neighbourhood of the boundary envelope and that there is a surface viscosity, η_s, which causes a resistance to motion in the surface, deduced the equation :

$$v_t = \frac{2}{9}\frac{R^2 g(\rho - \rho')}{\eta}\left[\frac{\eta_s + R}{\eta_s + R}\left(\frac{3\eta' + 3\eta}{3\eta' + 2\eta}\right)\right] \quad \ldots\ldots(8)$$

According to this equation, the expression in squared brackets will change with increasing R from 1 to 1·5. The effect of surface viscosity would be to cause a thin boundary layer of water to travel upwards with the bubble and in the case of small bubbles would prevent relative motion in the boundary layer.

Bond and Newton[20] have tested Equation (6), using viscous liquids, such as water-glass and golden syrup, and comparatively large bubbles, and have found that the expression in brackets has a value of unity for small bubbles and increases with R to an asymptotic value of about 1·43. They point out that, when η/η' is small, allowance should be made for the surface energy effect[19], which is related to the dimensionless quantity $\frac{(\rho - \rho')\, g\, R^2}{\gamma}$ and that the smaller the ratio η/η' the smaller is the value of the latter required to give transition from solid to fluid characteristics.

TABLE VI

Comparison of experimental conditions with those of other authors

Author	Diameter of liquid column (cm.)	Length of column (cm.)	Periodicity of bubbles of air	Liquid used	Temperature °C.	Ratio of $\frac{\text{Diameter of bubble}}{\text{Diameter of tube}} = \frac{d}{De}$
Hoefer	7·80	Maximum of 100	Very slow formation of bubbles. (No definite values are given under different conditions.)	Water		0·003–0·32
Bryn	(i) 7·00 (ii) 15 × 15 square section (equivalent diameter = 16·9)		No indication given.	Water and glycerine	17–20·5	0·006–0·35
Miyagi	(i) 2·41 (ii) 4·80	About 23–25	Does not indicate anything except that the rate is very slow.	Water	12–26	0·013–0·28
O'Brien and Gosline	(i) 2·99 (ii) 5·69 (iii) 15·24	150–550	(i) 1–3 bubbles/sec. (ii) For big bubbles only one bubble existed in the tube during measurement.	Water, mineral, seal oil and livestock oil	20	0·008–0·87
Allen ... . (for small bubbles)	3·00	72	Slow formation of bubbles and only one at a time for measurement.	Water and aniline	7·8–8·6	0·005–0·03
Luchsinger	16·00	250	No indication.	Water	20	0·006–0·030
Present investigation . ..	10.3 and 4·44	30–460	20–80 bubbles/minute.	Water and glycerine	18–21	0·002–0·074

The radius at which a gas bubble in water ceases to behave as a solid particle is not known accurately* and may well vary with the degree of contamination of the surface and the resultant changes in surface viscosity as postulated by Raleigh[12].

Allen's experiments indicate that, up to a radius of 0·04 cm. (and for Reynolds numbers of less than 1), the relation between R and v_t is linear and, on extrapolating his data to zero velocity, the limiting radius is 0·0035 cm.

Hoefer (*loc. cit.*) employed a tube of diameter 7·8 cm. and his experiments covered a range of bubble sizes from R = ·01 cm. to 1·34 cm. For the smaller diameter bubbles, his values for velocity are too high and are represented in the range R = 0·01 — 0·055 cm. by the empirical relationship

$$v_t = 0{\cdot}7256\ R^{1\cdot4} \quad \ldots\ldots\ldots\ldots\ldots(9)$$

for higher values of R, his results are generally confirmed by those of other workers.

Miyagi, Bryn and Luchsinger (*loc. cit.*) report data in fair agreement for bubbles of radius exceeding 0·1 cm., any differences being attributable to wall effects.

Miyagi has derived a relationship between v_t and R by making the initial assumption that the resistance to motion varies as the square of the velocity and that a mass of water is entrained by the bubble. The equation of motion then becomes

$$\left(V + \frac{1}{k}\rho\, V\right)\frac{d^2z}{dt^2} = g\,(\rho - \sigma)\,V - \phi\left(\frac{dz}{dt}\right)^2 \quad \ldots\ldots(10)$$

where z is the upward displacement in time, t ;

σ is the density of air ;
ρ is the density of fluid ;
ϕ is the coefficient of resistance depending on V ;
k is a coefficient also depending on V ;

and $\frac{1}{k}\rho V$ is the mass of water entrained by the bubble.

As the terminal velocity is constant, it must be independent of t and is given by

$$v_t = \frac{g\rho V}{\phi} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(11)$$

which in C.G.S. unit, gives

$$\phi = 4110\,\frac{R^3}{v_t^2}$$

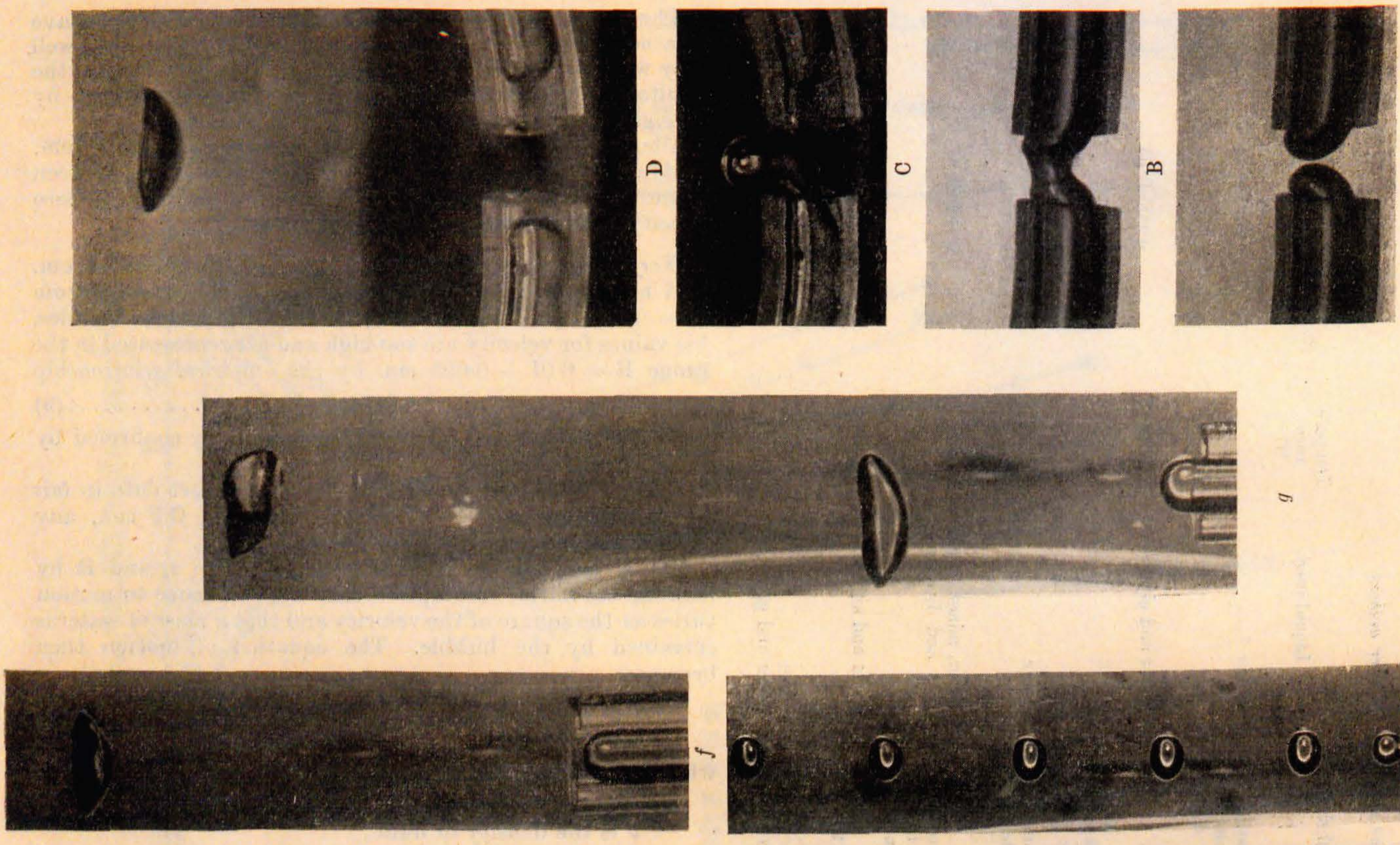
The coefficient of resistance is found to increase with the radius of the bubble as follows :

R	0·025	0·12	0·176	0·205	0·265	0·334	cms.
v_t	9·85	24·5	26·75	25·5	23·75	23·35	cms./sec.
$\phi^{\frac{1}{2}}$	0·257	0·1087	0·1779	0·2378	0·368	0·5295	

Miyagi has also shown that kR^2 is a constant having a value 0·054 when R is in cms., from which it follows that the mass of water carried up by a bubble is proportional to its surface area.

O'Brien and Gosline (*loc. cit.*) carried out experiments in tubes of diameters from 2·99 cms. to 15·24 cms. and showed that the wall effect in the case of narrow tubes tended to exert a retarding effect on the rate of rise of bubbles. Thus, in a 2·99 cm. tube, the velocity for a bubble of radius 0·4 cm. was 21·5 cms./sec., whilst, for a bubble of radius 1·1 cm.,

* *Note added in proof.*—Exception has been taken by the Referee to the above statement on the grounds that surface viscosity is not considered in the hydro-dynamic theory which serves to explain Bond and Newton's experimental results. It may be observed on the other hand, that Boussinesq's analysis implies that the static surface tension is not dynamically effective and that, as suggested by Klemm[22], Bond and Newton's results may equally well be explained by a dpendence of surface viscosity and surface tension.

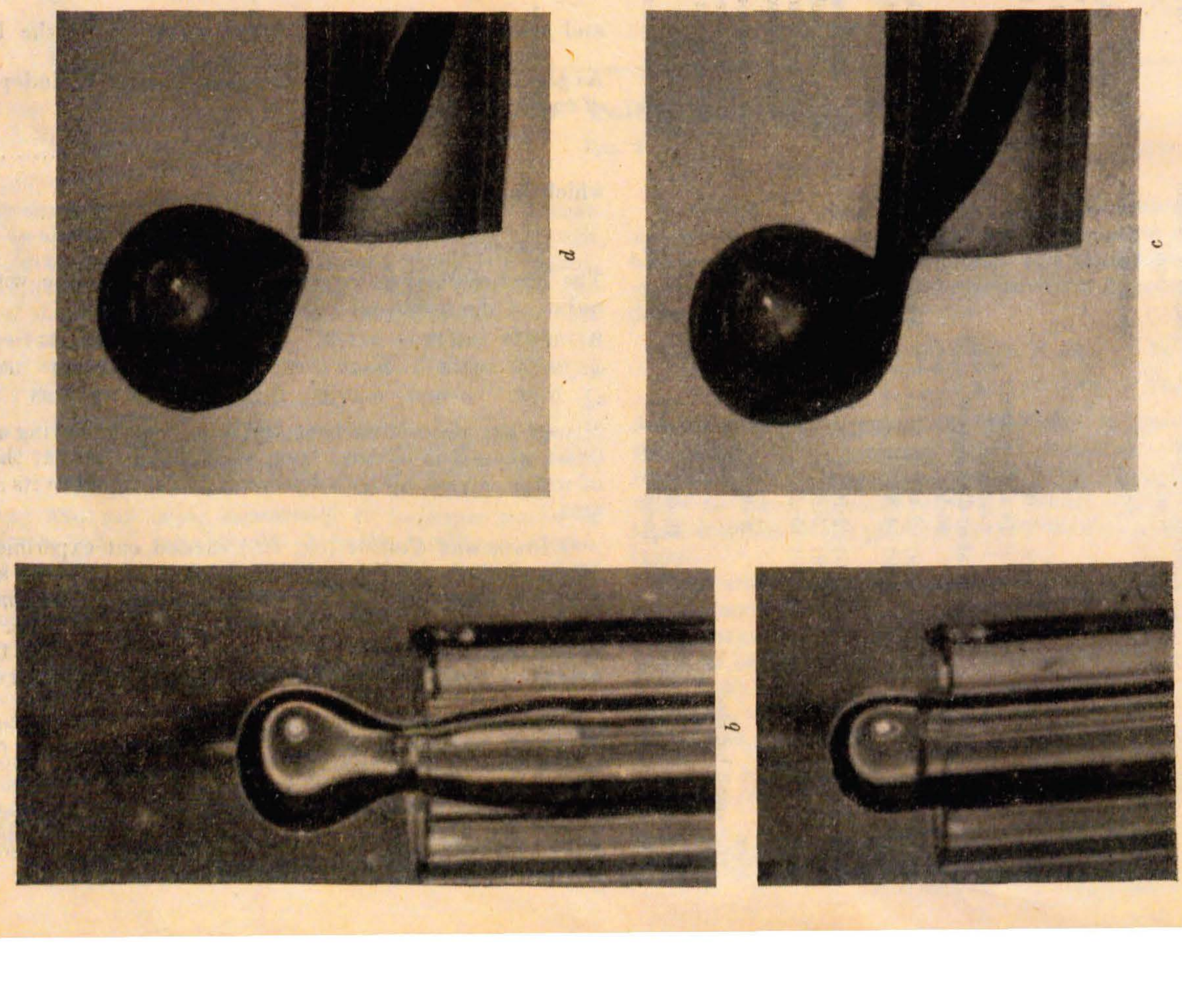


A B C D

g

e f

PLATE II



a b c d

PLATE I

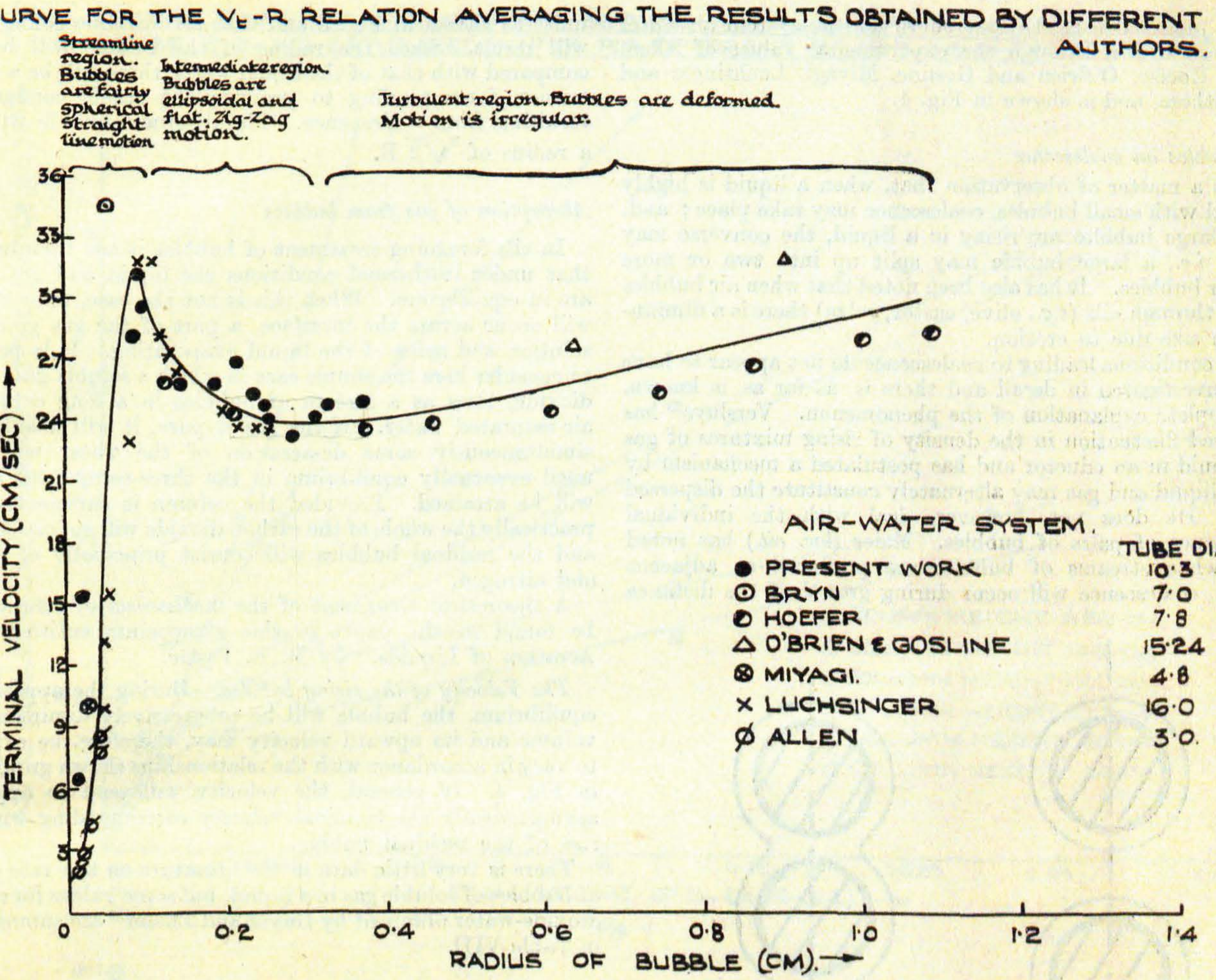


FIG. 4

it had fallen to 19 cms./sec. In a tube of diameter 15·24 cms., over the same range of bubble sizes, the velocity increased from 24·5 to 35 cms./sec.

Most of their experiments were carried out with comparatively large bubbles rising in water, mineral, seal oil and livestock oil, and their results are in reasonable agreement with those of Hoefer, Miyagi and Luchsinger for the larger-diameter tubes. It will be noted, however, that in no case do they obtain a clearly defined velocity maximum and no explanation can be found for this abnormality in their observations.

Results of present investigation

We have carried out a series of observations in a column 450 cms. high and 10·3 and 4·44 cms. in diameter, using an air-water system in which the water was saturated with air before velocity measurements were made. The bubbles were produced at glass capillary tubes as previously described and the velocities were measured by a stop watch.

The results are recorded in Table VII and those for the 10·3 cms. tube shown graphically in Figs. 3 and 4.

It will be seen that our results confirm those of Allen over the first part of the curve and those of Luchsinger, Hoefer and Miyagi in the region beyond the maximum. They also show that the linear relationship found by Allen for bubbles up to 0·039 cm. radius may be extended to 0·064 cm. radius.

TABLE VII

Relation between terminal velocity and radius of bubbles rising in water columns

Bubble radius R cms.	Observed velocity (v_t) cms./sec.	Reynolds number
Diameter of column = 4·44 cms.		
Temperature of water = 12·15° C.		
0·029	5·9	28
0·031	6·3	32
0·040	8·2	55
0·048	9·9	79
0·056	11·4	106
0·062	12·7	131
0·064	13·1	140
0·073	20·6	250
Diameter of column = 10·3 cms.		
Temperature of water = 18–21° C.		
ρ = 0·9983 gm./cc.		η = 0·0102 poise
0·013	6·25	15·3
0·025	15·4	75·4
0·089	28·0	479
0·090	31·0	546
0·097	29·3	556
0·140	26·1	715
0·198	25·9	1004
0·220	27·0	1133
0·240	25·0	1175
0·250	24·2	1185
0·288	23·5	1325
0·314	24·4	1500
0·322	24·6	1551
0·379	24·7	1833

For practical use, the best curve for the system air-water has been drawn through the experimental values of Allen, Bryn, Hoefer, O'Brien and Gosline, Miyagi, Luchsinger and the authors, and is shown in Fig. 4.

Some notes on coalescence

It is a matter of observation that, when a liquid is highly aerated with small bubbles, coalescence may take place ; and, when large bubbles are rising in a liquid, the converse may occur, *i.e.*, a large bubble may split up into two or more smaller bubbles. It has also been noted that when air bubbles travel through oils (*e.g.*, olive, castor, colza) there is a diminution in size due to erosion.

The conditions leading to coalescence do not appear to have been investigated in detail and there is, as far as is known, no complete explanation of the phenomenon. Versluys[23] has discussed fluctuation in the density of rising mixtures of gas and liquid in an eductor and has postulated a mechanism by which liquid and gas may alternately constitute the dispersed phase. He does not, however, deal with the individual coalescence of pairs of bubbles. Maier (*loc. cit.*) has noted that, when streams of bubbles are produced at adjacent orifices, coalescence will occur during growth if the distance

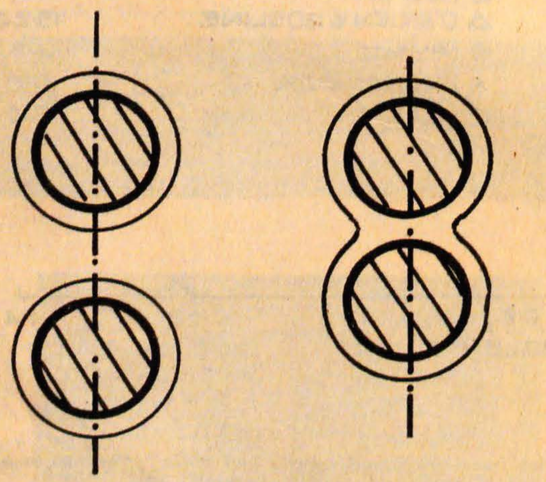


FIG. 5

between the orifices is less than the diameter of the bubble. The effect is shown in Plate II (*a*), (*b*), (*c*) and (*d*). Hoefer, Miyagi, Luchsinger and Allen do not refer to the phenomenon.

We have also observed the phenomenon when a stream of bubbles is moving freely in a column of liquid. The condition under which coalescence occurs most freely is when the bubbles are of such size as to be substantially spherical in shape and to travel along a vertical path ; in these circumstances, a bubble may be observed to be sucked into the slip stream of the bubble immediately ahead of it, the two remaining in proximity for a short interval and then coalescing. The essential condition appears to be the existence of some force, such as that due to the slip stream, holding the two bubbles in contact for a short time ; thus, when sideways contact is made, coalescence seldom occurs.

It has already been mentioned that a small gas bubble behaves like a solid sphere and this is interpreted to mean that it is surrounded by a rigid envelope or film of liquid which travels with it. Morley has calculated the thickness of this film, in the case of solid spheres, and Miyagi the mass of water entrained by a gas bubble rising in a liquid. When two bubbles are forced into contact and are held together, it may be assumed that their respective liquid envelopes will unite as shown in Fig. 5 and that an unstable configuration will result. Since the radius of the bubble will be large compared with that of the liquid waist, there will be a surface tension force tending to produce the stable configuration resulting from coalescence. The resultant bubble will have a radius of $\sqrt[3]{2}$ R.

Absorption of gas from bubbles

In the foregoing treatment of bubbles it has been assumed that under isothermal conditions the liquid and gas phases are in equilibrium. When this is not the case, mass transfer will occur across the interface, a part of the gas going into solution and some of the liquid evaporating. It is proposed to consider here the simple case in which a soluble gas, carbon dioxide, rises as a stream of bubbles in a long column of air-saturated water. If the gas is pure, it will dissolve and simultaneously some de-aeration of the water will occur until eventually equilibrium in the three-component system will be attained. Provided the column is sufficiently long, practically the whole of the carbon dioxide will go into solution and the residual bubbles will consist principally of oxygen and nitrogen.

A theoretical treatment of the mechanism of solution will be found in the paper in this symposium entitled "The Aeration of Liquids," by R. E. Pattle.

The Velocity of the rising bubbles.—During the approach to equilibrium, the bubble will be progressively diminishing in volume and its upward velocity may, therefore, be expected to vary in accordance with the relationships shown graphically in Fig. 4. In general, the velocity will tend to approach asymptotically the terminal velocity corresponding with the size of the residual bubble.

There is very little data in the literature on the rate of rise of bubbles of soluble gas in a liquid, but some values for carbon dioxide-water obtained by Guyer and Pfister[24] are summarised in Table VIII.

TABLE VIII

Bubble velocity as a function of bubble size for carbon dioxide-water at 20° C. (Guyer and Pfister)

Initial volume of bubble (ccs.)	Radius R (cm.)	Average velocity of rising bubble (cm./sec.)
0·005	0·105	35·0
0·01	0·135	31·5
0·02	0·178	28·2
0·03	0·195	26·8
0·04	0·21	25·6
0·06	0·25	24·5

These average velocities for the smaller-sized bubbles, when compared with those for an air-water system, are higher than would be expected ; and it is probable that, since they were measured in a column under 40 cms. long, end effects have masked the velocity-size relationship. Guyer and Pfister give no data on the variation of the size and velocity of the bubbles with distance from the orifice. We have measured the average velocities of ascent and the final bubble volume for a range of initial bubble sizes in columns 10·3 cms. diameter and of lengths 30, 60, 80, 120, 145 and 456 cms. In all cases the column of water was initially saturated with air. The bubbles were produced at a glass capillary orifice immersed beneath a layer of mercury at the foot of the column, so as to minimise absorption during the time the bubble is forming in water. The initial size of bubble was determined for each orifice by measurement in CO_2-saturated water. The results are summarised in Table IX.

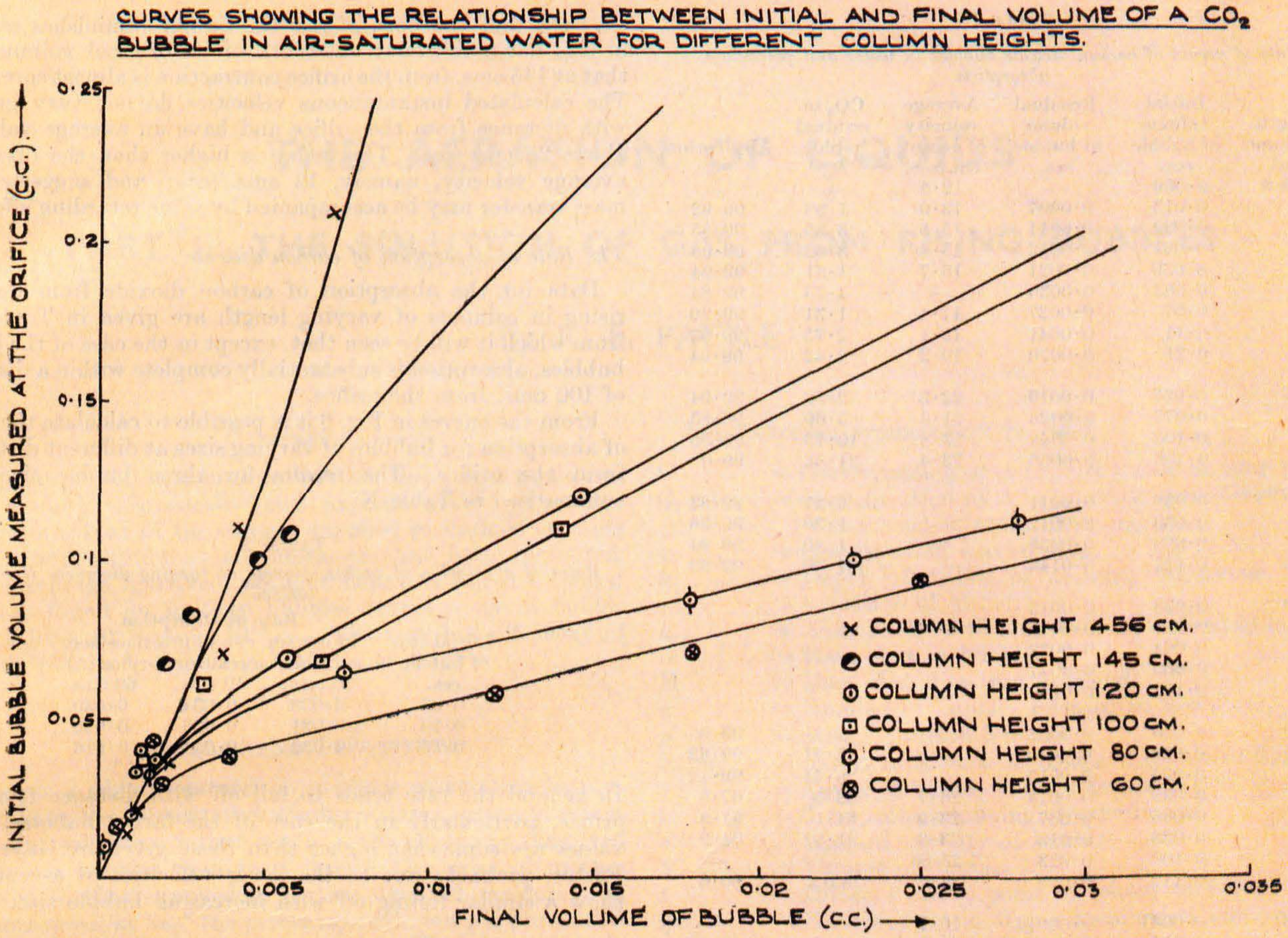

CURVES SHOWING THE RELATIONSHIP BETWEEN INITIAL AND FINAL VOLUME OF A CO_2 BUBBLE IN AIR-SATURATED WATER FOR DIFFERENT COLUMN HEIGHTS.
INITIAL BUBBLE VOLUME MEASURED AT THE ORIFICE (c.c.)
0·25
0·2
0·15
0·1
0·05
0·005
0·01
0·015
0·02
0·025
0·03
0·035
FINAL VOLUME OF BUBBLE (c.c.)
× COLUMN HEIGHT 456 CM.
COLUMN HEIGHT 145 CM.
COLUMN HEIGHT 120 CM.
COLUMN HEIGHT 100 CM.
COLUMN HEIGHT 80 CM.
COLUMN HEIGHT 60 CM.

Fig. 6

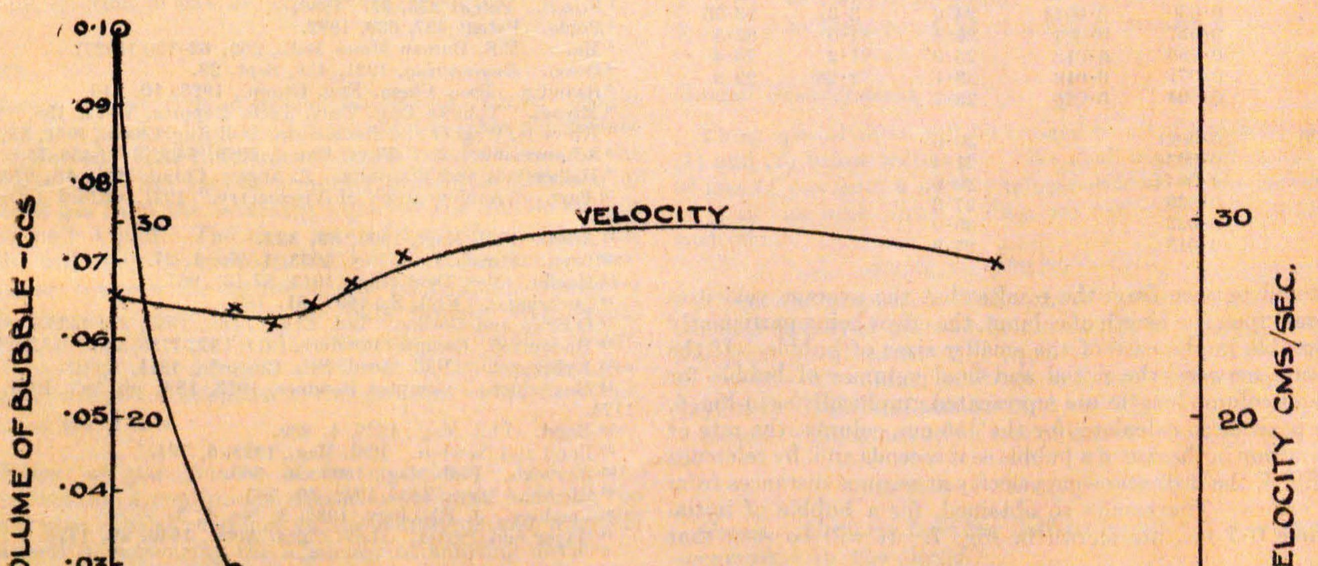

VOLUME OF BUBBLE - CCS
0·10
·09
·08
·07
·06
·05
·04
·03
·02
·01
0
30
20
10
VELOCITY
VOLUME
100
200
300
400
500
HEIGHT FROM NOZZLE - CMS.
VELOCITY CMS/SEC.
30
20
10

Fig. 7

TABLE IX

Rate of ascent of carbon dioxide bubbles in water and percentage absorption

Length of column cms.	Initial volume of bubble ccs.	Residual volume of bubble ccs.	Average velocity of ascent cm./sec.	CO_2 in residual bubble %	Absorption %
456·5	0·009	—	12·5	—	—
	0·013	0·0007	13·9	1·98	99·92
	0·020	0·0011	15·5	3·85	99·85
	0·031	0·0012	15·8	3·63	99·90
	0·036	0·0021	15·7	1·51	99·94
	0·062	0·0030	—	4·75	99·84
	0·07	0·0037	17·4	1·31	99·90
	0·11	0·0041	19·1	2·75	99·93
	0·21	0·0070	19·2	2·42	99·94
145	0·067	0·0019	22·3	2·52	99·94
	0·077	0·0026	21·6	5·60	98·83
	0·100	0·0049	23·8	10·73	99·56
	0·106	0·0056	23·4	21·52	99·0
120	0·030	0·0011	—	2·31	99·83
	0·036	0·0012	—	1·30	99·96
	0·068	0·0056	—	1·80	99·94
	0·122	0·0145	—	61·30	92·80
100	0·038	0·0013			
	0·058	0·0022			
	0·064	0·0032			
	0·069	0·0047			
	0·113	0·0140			
80	0·010	0·0002	—	2·91	99·95
	0·020	0·0010	—	3·71	99·83
	0·035	0·0013	—	25·71	99·12
	0·039	0·0014	26·7	63·0	97·9
	0·065	0·007	23·9	81·1	91·9
	0·088	0·018	23·6	81·9	84·2
	0·100	0·023	25·8	—	—
	0·112	0·028	24·2	83·2	80·6
60	0·0041	0·00014	10·7	—	—
	0·015	0·00056	20·7	1·2	96·4
	0·022	0·0073	22·2	67·8	93·3
	0·030	0·0024	24·0	8·3	93·75
	0·037	0·004	26·9	87·5	82·5
	0·056	0·012	25·0	91·2	78·4
	0·071	0·018	23·1	2·28	99·9
	0·094	0·025	23·7	—	—
30	0·0021		30·0		
	0·0034		21·4		
	0·0044		20·0		
	0·039		27·3		
	0·023		30·0		
	0·013		27·3		

It will be seen from the results that the average velocities depend upon the length of column, the effect being particularly noticeable in the case of the smaller sizes of bubble. If the relation between the initial and final volumes of bubble for various column lengths are represented graphically as in Fig. 6, it is possible to calculate, for the 456 cm. column, the rate of diminution in the size of a bubble as it ascends and, by reference to Fig. 4, the instantaneous velocity at various distances from the orifice. The results so obtained, for a bubble of initial volume 0·1 cc., are shown in Fig. 7. It will be seen that over the first 100 cms. of rise the bubble diminishes rapidly in size down to about one-tenth of its original volume and that at 145 cms. from the orifice contraction is almost complete. The calculated instantaneous velocities do not vary greatly with distance from the orifice and have an average value of about 25 cms./sec. This value is higher than the observed average velocity, namely, 19 cms./sec.; and suggests that mass transfer may be accompanied by some retarding effect.

The Rate of absorption of carbon dioxide

Data for the absorption of carbon dioxide from bubbles rising in columns of varying length are given in Table IX, from which it will be seen that, except in the case of the larger bubbles, absorption is substantially complete within a distance of 100 cms. from the orifice.

From the curves in Fig. 6 it is possible to calculate the rates of absorption for bubbles of varying sizes at different distances from the orifice. The results for three bubble sizes are summarised in Table X.

TABLE X

Rates of absorption of carbon dioxide at varying distances from the orifice

Initial size of bubble ccs.	Rate of absorption in ccs./sq. cm. of interface/sec. at distances from orifice: 20 cms.	40 cms.	60 cms.
0·05	0·033	0·034	0·030
0·10	0·031	0·026	0·020
0·15	0·032	0·024	0·016

In general the rate tends to fall off with distance from the orifice, particularly in the case of the larger bubbles. Our values are somewhat higher than those given by Guyer and Pfister, probably due to the lower velocities of ascent, but show a similar falling off with increasing bubble size.

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