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GROWTH RATE OF A GAS BUBBLE DURING ELECTROLYSIS IN SUPERSATURATED LIQUID

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Abstract — The growth of a spherical gas bubble on a horizontal plane has been studied both experimentally and theoretically. Experiments with a miniature electrode have been carried out in a uniformly (super)-saturated solution of oxygen in water, with and without electrolysis. All experiments have been done at a temperature of 291 K and at (sub)atmospheric pressure(s). An analytical expression has been derived for bubble growth in a uniformly supersaturated (or superheated) liquid, which holds for all Jakob numbers. For the more complicated case of simultaneous electrolysis a power series expansion has been found suitable. The experimental results were reproducible and in all cases the agreement with theoretical models was excellent.

NOMENCLATURE

A, surface area of the bubble [m^2];
a, $= JaD$ [$m^2 s^{-1}$];
a_n, coefficient of the nth term in the power series of equation (12);
B, $R/t^{1/2}$, independent of t [$m s^{-1/2}$];
b, $= Ja\,(3D/\pi)^{1/2}$ [$m^2 s^{-1}$];
C, local concentration of gas in liquid [$kg m^{-3}$];
C_b, initially uniform concentration of gas in bulk liquid [$kg m^{-3}$];
C_e, local concentration of gas in liquid due to the process of electrolysis [$kg m^{-3}$];
C_s, saturation concentration of gas in liquid [$kg m^{-3}$];
ΔC, uniform supersaturation concentration of gas in liquid [$kg m^{-3}$];
c, $= (V_m I/4\pi qeN) = (\hat{R}TI/4\pi pqF)$ [$m^3 s^{-1}$];
D, diffusivity of gas in liquid [$m^2 s^{-1}$];
e, elementary charge [C];
F, Faraday constant [$C kmol^{-1}$];
I, electrical current [A];
Ja, $= \Delta C/\rho_2$, Jakob number;
K, C/p, equilibrium constant in Henry's law [$kg m^{-3} Pa^{-1}$];
N, Avogadro's number [$kmol^{-1}$];
p, pressure [Pa];
Δp, excess pressure [Pa];
q, number of electrons involved in the formation of one molecule gas during electrolysis;
R, bubble radius [m];
$\dot{R}$, dR/dt [$m s^{-1}$];
$\hat{R}$, gas constant [$J K^{-1} kmol^{-1}$];
r, radial distance [m];
S, $= (a/c)R$, dimensionless bubble radius;
T, temperature [K];
t, time [s];
V_m, kilomolar volume [$m^3 kmol^{-1}$].

Greek symbols

α, $= b/a^{1/2}$;
β, dimensionless bubble growth constant;
ρ_1, liquid density [$kg m^{-3}$];
ρ_2, gas density [$kg m^{-3}$];
σ, surface tension [$kg s^{-2}$];
τ, $= (a^3/c^2)t$, dimensionless time.

Numerical values

$C_s(O_2, H_2O) = 4.37 \times 10^{-2}\ kg m^{-3}$ (293 K, 101.3 kPa [19]);
$D(O_2, H_2O) = 1.95 \times 10^{-9}\ m^2 s^{-1}$ (293 K [19, 20]).

1. INTRODUCTION

THE BEHAVIOUR of gas bubbles on electrodes is an important subject in bubble dynamics [1]. In general, each bubble is influenced by its neighbours in close proximity, which makes it a rather complicated problem to deal with. A more simple problem, which has to be clarified first, is the behaviour of a single separate bubble during electrolysis. The aims of the present study are to obtain reliable experimental results on the growth of a single gas bubble during electrolysis in a solution of well-defined uniform concentration and to develop theoretical models to explain these results.

An experimental set-up has been chosen, in which the dimension of the electrode surface is small compared to the departure diameter of the bubble. Some experiments, with set-ups similar to the one described in this paper, have been carried out previously [2–6]. Darby and Haque [2] applied high electrical current densities, but the liquid in their cell had an unknown gas concentration. Westwater *et al.* [3–5] pioneered in this field, with remarkable results. However, gas bubbles were also studied during electrolysis in a solution of unknown, non-uniform gas concentration [3, 4]. Also they did not succeed in developing a device

to obtain predictable nucleation [5]. On the other hand, the dissolving of stationary gas bubbles into pure degassed liquids has been investigated extensively, giving reproducible results [7]. Generally speaking, the existing theory was confirmed [8–12]. As pointed out by Cable [11], it appears to be much easier to obtain reasonable agreement between theory and experiment for dissolving bubbles than for growing ones.

A similarity exists between gas bubble growth due to supersaturation and vapour bubble growth due to superheating [1]. Quite a few papers have been published on experiments concerning the growth of a vapour bubble in a uniformly superheated liquid [13–15]. Experiments performed in the so-called diffusion-controlled region show good agreement with theory [16–18]. However, these experiments on boiling have been carried out for Jakob numbers, which are at least of an order of magnitude larger than those involved in this paper.

2. THEORY

2.1. *Pure gas bubbles*

During electrolysis gases may be produced at both electrodes. Initially, before bubbles are gene.ated, an electrolytically developed gas dissolves into the liquid adjacent to the electrode. However, most gases have low solubility in liquids and, in addition, diffusion coefficients are small. As a consequence the adjacent liquid will rapidly become supersaturated. If the local supersaturation exceeds the required value, nucleation takes place on cavities at the electrode surface and a gas bubble starts to grow. Due to a concentration gradient around the bubble wall, it will grow by mass diffusion. The mass balance for a growing bubble is given by

$$\rho_2 4\pi R^2 \dot{R} = D \int\int \left(\frac{\partial C}{\partial r}\right)_{r=R} \mathrm{d}A. \tag{1}$$

If C_e denotes the concentration due to the process of electrolysis, and C_b the initial concentration of gas in the bulk liquid, equation (1) is replaced by

$$\rho_2 4\pi R^2 \dot{R} = D \int\int \left[\left(\frac{\partial C_b}{\partial r}\right)_{r=R} + \left(\frac{\partial C_e}{\partial r}\right)_{r=R}\right] \mathrm{d}A. \tag{2}$$

The surface area of the miniature electrode is small compared to the cross-section of the bubble during most of its growth time. Therefore, it is assumed that all the gas produced at the electrode is transported directly to the bubble. In addition, it is assumed that the concentration gradient $(\partial C_b/\partial r)_{r=R}$ of the gas initially present in the liquid is uniform around the bubble wall. Strictly speaking, this holds only for a free and non-translating bubble. If the current is kept constant, equation (2) can be rewritten as follows

$$\dot{R} = \frac{D}{\rho_2}\left(\frac{\partial C_b}{\partial r}\right)_{r=R} + \frac{V_m I}{qeN4\pi R^2}, \tag{3}$$

with the boundary condition $R = 0$ at $t = 0$.

Surface tension forces as well as viscosity and inertia effects are neglected here. The following three cases are considered separately:

(i) $I \neq 0$, and $(\partial C_b/\partial r)_{r=R} = 0$, i.e. the liquid has been saturated with the same gas as the gas produced electrolytically at the miniature electrode. The solution of equation (3) is [1, 2]

$$R(t) = \left(\frac{3V_m I t}{4\pi qeN}\right)^{1/3} = \left(\frac{3\hat{R}TIt}{4\pi pqF}\right)^{1/3}. \tag{4}$$

(ii) $I = 0$, and $(\partial C_b/\partial r)_{r=R} \neq 0$, which means an initially uniform supersaturation of gas in the liquid. In the absence of electrolysis, a solution for the concentration gradient at the bubble wall is given by

$$\left(\frac{\partial C_b}{\partial r}\right)_{r=R} = \Delta C\left[\frac{1}{R} + \frac{1}{\left(\frac{\pi}{3}Dt\right)^{1/2}}\right]. \tag{5}$$

A similar solution was derived by Fricke [8], and Epstein and Plesset [9]. However, in equation (5) the radial convection due to the bubble growth is incorporated. Combination of equation (5) with equation (3), with $I = 0$, and subsequent substitution of $R = Bt^{1/2}$ in the resulting differential equation, yields

$$R(t) = \left\{\left(\frac{3}{\pi}\right)^{1/2} Ja + \left(\frac{3}{\pi}Ja^2 + 2Ja\right)^{1/2}\right\}(Dt)^{1/2}. \tag{6}$$

In the limits of the Jakob number, $\Delta C/\rho_2$, this expression resolves into the following well-known equations [16–18]:

$$R(t) = \left(\frac{12}{\pi}\right)^{1/2} Ja(Dt)^{1/2}, \quad \text{for } Ja \gg 1, \tag{7}$$

$$R(t) = (2JaDt)^{1/2}, \quad \text{for } Ja \ll 1. \tag{8}$$

Scriven [18] calculated the diffusion-controlled bubble growth numerically and obtained the expression $R = 2\beta(Dt)^{1/2}$, where $\beta = \beta(Ja)$. Comparison of the analytical solution, equation (6), with the numerical calculations [18] shows that equation (6) is a good approximation, with a maximal deviation of 2.3% (Fig. 8).

(iii) $I \neq 0$ and $(\partial C_b/\partial r)_{r=R} \neq 0$, i.e. electrolysis in a uniformly supersaturated solution. In this case, equation (3) can be replaced by

$$\dot{R} = JaD\left[\frac{1}{R} + \frac{1}{\left(\frac{\pi}{3}Dt\right)^{1/2}}\right] + \frac{V_m I}{4\pi qeNR^2}, \tag{9}$$

i.e.

$$\dot{R} = \frac{a}{R} + \frac{b}{t^{1/2}} + \frac{c}{R^2}, \tag{10}$$

where $a = JaD$, $b = Ja(3D/\pi)^{1/2}$ and $c = V_m I/4\pi qeN$ are taken constant. Equation (10) becomes dimension-

less by introducing $S = (a/c)R$ and $\tau = (a^3/c^2)t$, which results in

$$\frac{dS}{d\tau} = \frac{1}{S^2} + \frac{1}{S} + \frac{\alpha}{\tau^{1/2}}, \tag{11}$$

where $\alpha = b/a^{1/2} = [(3/\pi)Ja]^{1/2}$. An analytical solution is not attainable here. In the two preceding cases solutions were obtained in the form of $R \sim t^{1/3}$, equation (4), and $R \sim t^{1/2}$, equation (6), respectively. Consequently, $S(\tau)$ is expanded into the following infinite series:

$$S(\tau) = \tau^{1/3} \sum_{n=0}^{\infty} a_n \tau^{n/6}. \tag{12}$$

After substitution of equation (12) into equation (11), the values of a_n are obtained

$a_0 = 1/3$, $a_1 = 6\alpha/7$, and (for $n \geq 2$),

$$a_n = \left\{\left(\frac{n}{6} + 1\right)a_0^2\right\}^{-1}\left(a_{n-2} + \alpha a_0 a_{n-1} + \sum_{k=1}^{n-1}\left[\alpha a_k a_{n-k-1} - \tfrac{1}{6}a_{n-k}\left\{2a_0 a_k + (n-k+2)\times\left(\sum_{p=0}^{k} a_p a_{k-p}\right)\right\}\right]\right). \tag{13}$$

Under the conditions corresponding to the experiments described in Section 3, the series converges rapidly and the dimensionless radii of convergence turn out to be much larger than the dimensionless maximal time considered experimentally. If only the first five terms are used a maximal error of only 0.1% is made compared to the first 25 terms.

If $R \ll (\pi Dt/3)^{1/2}$, a solution in the form of a power expansion series can be avoided. Equation (10) simplifies to

$$\dot{R} = \frac{a}{R} + \frac{c}{R^2}, \tag{14}$$

and the solution is now given by

$$t = \frac{R^2}{2a} - \frac{cR}{a^2} + \frac{c^2}{a^3}\ln\left(1 + \frac{aR}{c}\right). \tag{15}$$

Equation (15) is equivalent to the solution of equation (11) with $\alpha = 0$. So, all solutions can be drawn in one dimensionless plot.

2.2. *The effect of water vapour in the gas bubble*

A growing gas bubble in water will contain water vapour. If it is assumed that thermodynamic equilibrium exists continuously between gas and liquid phase during bubble growth, the relative humidity inside the bubble approximates to 1. This means, that the partial pressure of the gas in the bubble equals the ambient pressure in the system minus the saturation vapour pressure corresponding to the temperature. This results in a faster bubble growth due to the simultaneous evaporation of the liquid. The theoretical equations of Section 2.1 have to be corrected for this phenomenon for comparison with experimental results at relatively low pressures or at relatively high temperatures. This can be done for all equations, using the partial pressure of the gas instead of the total pressure of the system. Substitution of the partial pressure in, for example, the denominator of equation (4) leads to an expected increase in bubble growth rate.

3. EXPERIMENTAL RESULTS

All experiments are carried out on oxygen in distilled water. Oxygen has been chosen because it is one of the gases which evolve during water electrolysis and also because it is less dangerous than hydrogen if large quantities are used to saturate the liquid.

3.1. *Experimental set-up*

The experimental set-up has been drawn in Fig. 1. One of the major problems was to avoid natural convection. To avoid local heating of the liquid in the test-cell, illumination was permitted only during exposure. The infrared rays of the bulb were absorbed in a water bath with a thickness of 10 cm. The bubble was generated on a platform turned away from the window to minimize convection near the top of the miniature electrode. Experiments were also carried out using a needle-shaped electrode. However, these results were not reproducible due to convection. All outlets of the stainless steel pressure vessel were interconnected to prevent the occurrence of pressure differences. Bubble formation due to spontaneous nucleation never occurred. Sometimes two or more bubbles appear on the electrode after switching on the electric current. The experiments were carried out on a normal bench in a remote area in the laboratory.

To saturate the water, oxygen was pressurized through porous glass, while stirring the liquid magnetically. A swarm of tiny bubbles produced a high surface/volume ratio. On the other hand, the radius of these bubbles exceeded the nucleation radius substantially, hence a liquid supersaturation is avoided. Experiments introducing bubbles in the liquid, showed saturation was achieved after 4 h. A uniform supersaturation was established by applying a negative pressure step. The pressure in the vessel could be kept at a constant value between 4 and 100 kPa using a

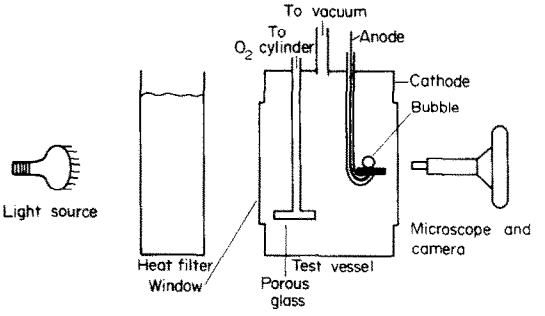

FIG. 1. Experimental set-up.

pump, in combination with a manostat and a pressure gauge.

Finally, the miniature electrode, a cut platinum wire, carefully sealed in soft glass and having a geometrical surface area of 0.79×10^{-8} m^2, was polished extensively to reduce both the number and the size of the cavities on the surface. However, the result was disappointing, because generation of more than one bubble even occurred at relatively low current densities. Instead of polishing, the electrode was treated with a non-wetting agent (liquid paraffin) before each experiment, resulting in the appearance of just one bubble. Another advantage was that the bubble departure time increased, which resulted in a longer observation time.

Generation of a bubble was simply caused by applying a short electric current pulse. A calculation showed a negligible influence of the pulse on the concentration field of the gas in the liquid.

Periodically, photographs were taken through a microscope and the elapsed time was recorded using a stopwatch. Larger bubbles showed deviations from the spherical shape. This has been corrected for by the use of an equivalent bubble radius, i.e. the radius of a sphere having the same volume as the bubble. All experiments have been carried out at a temperature close to 291 K, and at (sub)atmospheric pressure(s). The electric current, if used, has been kept constant by using a high voltage in combination with a relatively large resistor (10 MΩ), connected in series with the cell.

3.2. *Electrolysis in saturated liquid*

First the procedure for saturating the liquid took place. Then the electric current was switched on. If the bubble did not appear immediately it was generated by applying a current pulse. Currents were varied between 2.00 and 5.00 μA. Higher values resulted in more bubbles on the electrode. Some results are shown in Figs. 2 and 3. Experiments are in good agreement with theory.

Similar experiments have been carried out at different pressures ranging from 2 to 8 kPa. These values slightly exceed the saturated water vapor pressure at the given temperature. The experimental results clearly show that the relative humidity inside the bubble approximates to 1 (Figs. 4 and 5).

3.3. *Gas bubble growth in uniformly supersaturated liquid without electrolysis*

The liquid has been saturated at atmospheric pressure p_1. Next, the pressure in the cell was suddenly decreased by an amount Δp, to the value p_2. A lower saturation concentration corresponds to this value according to Henry's law, $C = Kp$. So, a supersaturation ΔC has been established: $\Delta C = K(p_1 - p_2) = K \cdot \Delta p$. Accordingly, the Jakob number equals $Ja = \Delta C/\rho_2$. In the experiments carried out, this number has been varied between 10^{-2} and 1. The bubble was always generated on the electrode due to an electric current pulse. Results and a comparison

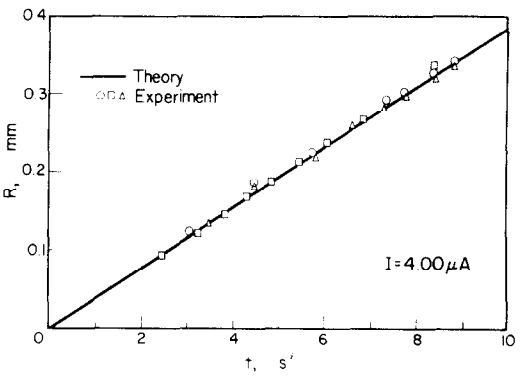

FIG. 2. Dependence of bubble radius on the one third power of time during electrolysis at a constant electric current on a miniature electrode. Different symbols denote different runs. (O_2/H_2O, p = 101 kPa, T = 291 K).

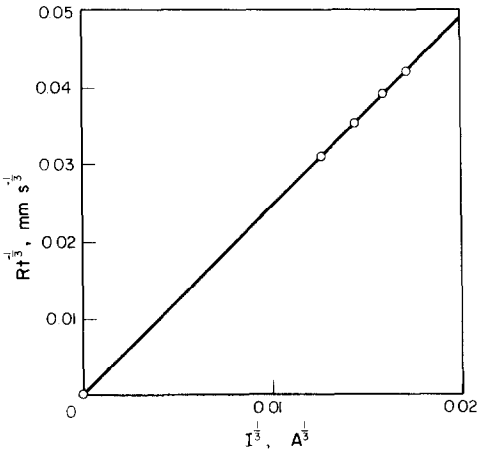

FIG. 3. The slopes of the R vs $t^{1/3}$ curves as a function of the one third power of the electric current. The symbols are average experimental values (O_2/H_2O, p = 101 kPa, T = 291 K). —— Theory according to equation (4).

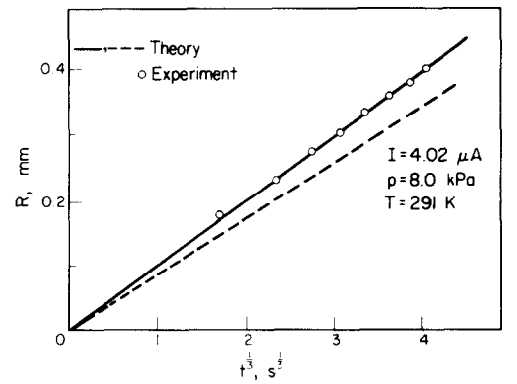

FIG. 4. Dependence of bubble radius on the one third power of time, during electrolysis at a miniature electrode, at a subatmospheric pressure (O_2/H_2O, T = 291 K). Theory according to equation (4): —— with relative humidity 1; – – – with relative humidity 0.

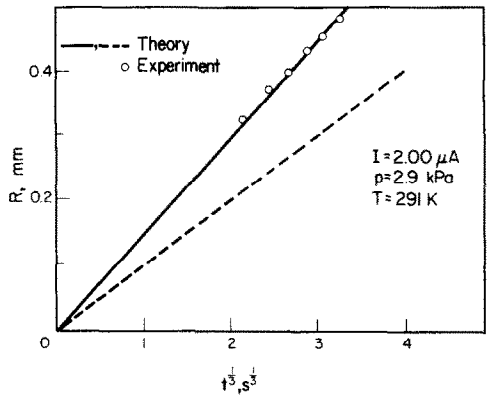

FIG. 5. Dependence of bubble radius on the one third power of time, during electrolysis at a miniature electrode, at a subatmospheric pressure. (O_2/H_2O, $T = 291$ K). Theory according to equation (4): —— with relative humidity 1; – – – with relative humidity 0.

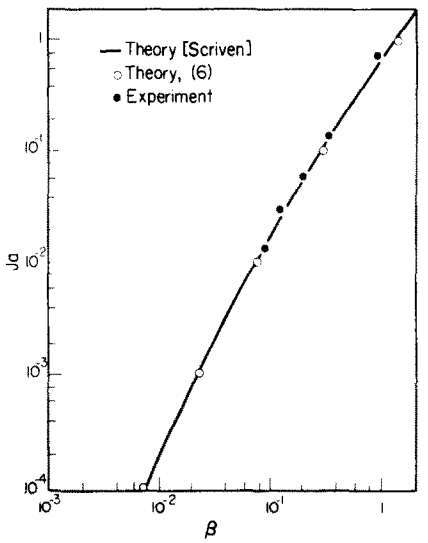

FIG. 8. Experimental and theoretical dependences of the growth constant β on the dimensionless Jakob number.

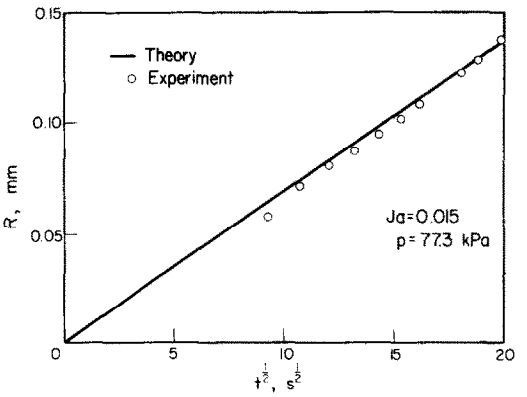

FIG. 6. Bubble radius vs square root of time for bubble growth on a horizontal plate in an initially uniformly supersaturated liquid (O_2/H_2O, $T = 291$ K).

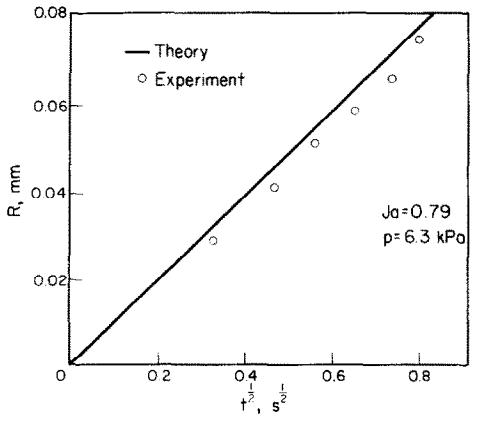

FIG. 7. Bubble radius vs square root of time for bubble growth on a horizontal plate in an initially uniformly supersaturated liquid (O_2/H_2O, $T = 291$ K).

with theory are shown in Figs. 6 and 7. In general, the bubble could be observed during a longer time period than indicated by those figures. However, departure times and departure radii are not given here because of insufficient reproducibility in this type of experiment. Preliminary experiments resulted in a large number of figures, in which the corresponding growth curves showed an increasing slope. This is attributed to the effect of convection, which decreases the average diffusion layer around the bubble causing an increase of the bubble growth rate. The absence of this effect in the final experiments proves the reliability of the results.

Figure 8 shows the experimental values of the growth constant $\beta = R/2(Dt)^{1/2}$ as a function of the Jakob number. The agreement with Scriven's theory [18] and with equation (6) is good. Most experimental values of β, however, are below the theoretical ones. Apparently, the reason for this is the influence of the solid wall which is not incorporated in either theory. Of course, the wall diminishes the upward gas flux to the bubble. Buehl and Westwater [5] gave theoretical predictions for the growth rate of a spherical bubble tangent to a wall. Actually, the present experimental results show the predicted trend, cf. Fig. 2 of [5].

3.4. *Electrolysis in a uniformly supersatured liquid*

These experiments are a combination of those described in Sections 3.2. and 3.3. After saturating the liquid and performing a negative pressure step, a constant electric current is switched on. The resulting Jakob number varied from 0.02 to 0.14, the pressures from 20 to 60 kPa. Experiment and theory are compared in Fig. 9. At low Jakob numbers, the analytical equation (15) agrees with the experimental data. However, at high Jakob numbers the relative error in the calculated radius increases drastically and the

series expansion, equations (12) and (13), has to be applied showing precise coverage of the data.

4. CONCLUSIONS

Because of the relatively low Jakob number, the growth rate of a gas bubble is small and the diffusion layer thickness is large. Therefore, in studying a growing gas bubble, extreme caution should be taken to avoid unknown convective currents. If also a small electrode, pretreated with a non-wetting agent, is used to prevent nucleation of other bubbles, reproducible bubble growth rates can be obtained.

In the experiments described above the relative humidity approximates to 1. So, the statement that thermodynamic equilibrium exists at the interface during gas bubble growth under the mentioned conditions, seems to be justified.

In uniformly supersaturated water, the Jakob numbers were varied from 10^{-2} to 1. The experimental results could not be explained by the well-known diffusion equation for high Jakob numbers, equation (7), showing clearly the limitations of boundary layer theory by experiment.

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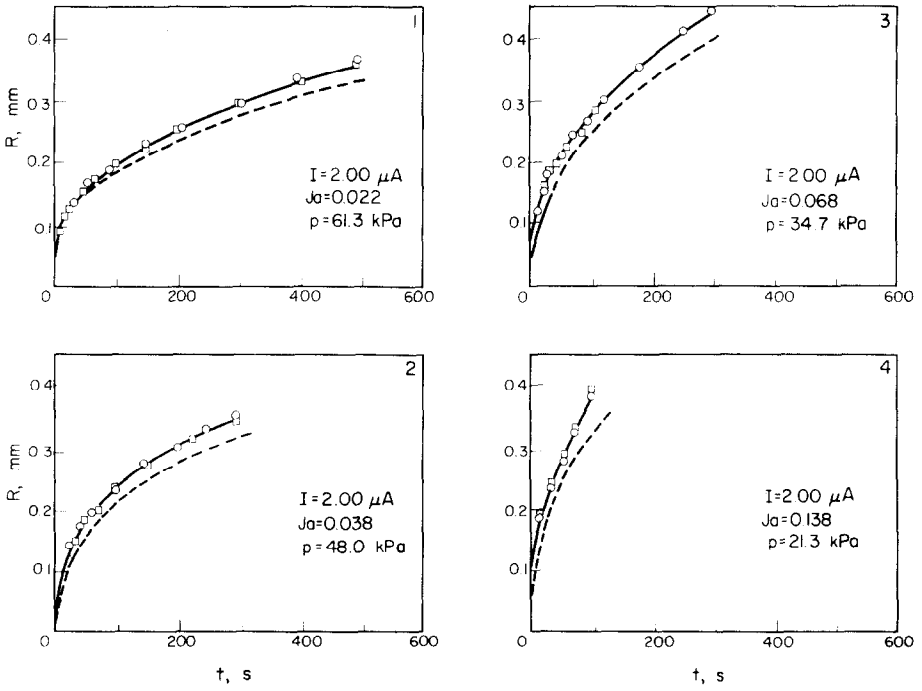

FIG. 9. Bubble radius vs time, during electrolysis in initially uniformly supersaturated liquid (O_2/H_2O, $T = 291$ K). The pressure decreases and the Jakob number increases, Figs. 9.1–9.4. —— Theory according to equation (12), $n = 0, 1, 2, 3, 4$; - - - Theory according to equation (15).

CROISSANCE D'UNE BULLE DE GAZ PENDANT L'ELECTROLYSE DANS UN LIQUIDE SURSATURE

Résumé — On étudie expérimentalement et théoriquement la croissance d'une bulle de gaz sphérique sur un plan horizontal. Des expériences avec une électrode miniature ont été conduites dans une solution sursaturée d'oxygène dans l'eau avec et sans électrolyse. Toutes les expériences correspondent à une température de 291 K et à des pressions inférieures ou égales, à la pression atmosphérique. Une expression analytique a été obtenue, pour la croissance de bulle dans un liquide sursaturé (ou surchauffé), valable pour tous les nombres de Jakob. Pour le cas plus complexe d'une électrolyse simultanée on a trouvé un développement en série. Les résultats expérimentaux sont reproductibles et l'accord avec les modèles théoriques est excellent.

WACHSTUMSGESCHWINDIGKEIT EINER GASBLASE WÄHREND DER ELEKTROLYSE IN ÜBERSÄTTIGTER FLÜSSIGKEIT

Zusammenfassung — Das Wachstum einer runden Gasblase auf einer waagerechten Ebene wurde sowohl versuchsmäßig als auch theoretisch untersucht. Versuche mit einer Kleinstelektrode wurden in einer gleichmäßig übersättigten Lösung von Sauerstoff in Wasser durchgeführt, und zwar mit und ohne Elektrolysevorgang. Alle Versuche wurden bei einer Temperatur von 291 K und bei Normalluftdruck (und darunter liegenden Drücken) gemacht. Für das Blasenwachstum in einer gleichmäßig übersättigten (oder überhitzten) Flüssigkeit wurde ein analytischer Ausdruck gefunden, der für alle Jakob-Zahlen gilt. Für den recht schwierigen Fall der gleichzeitigen Elektrolyse wurde eine Potenzreihenentwicklung als geeignete Darstellungsform gefunden. Die Versuchsergebnisse waren in allen Fällen reproduzierbar, und die Übereinstimmung mit den theoretischen Berechnungen war ausgezeichnet.

СКОРОСТЬ РОСТА ГАЗОВОГО ПУЗЫРЬКА ПРИ ЭЛЕКТРОЛИЗЕ В ПЕРЕНАСЫЩЕННОЙ ЖИДКОСТИ

Аннотация — Экспериментально и теоретически исследуется рост сферического газового пузырька на горизонтальной поверхности. Опыты с миниатюрным электродом проводились в равномерно насыщенном и перенасыщенном растворах кислорода в воде при наличии и отсутствии электролиза. Опыты проводились при 291 К и атмосферном и пониженном давлениях. Дано аналитическое описание роста пузырька в равномерно перенасыщенной (или перегретой) жидкости, справедливое для всех значений числа Якоба. В более сложном случае, с учётом электролиза в жидкости, удобнее пользоваться степенным разложением. Во всех случаях получены хорошие воспроизводимость опытов и их согласование с теорией.