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Destabilizing surface bubbles with excessive bulk oversaturation

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ABSTRACT

The response and dynamics of surface-attached bubbles in gas-oversaturated environments have practical implications for industrial processes such as photocatalytic water splitting. Surprisingly, the behavior of microbubbles and nanobubbles depends rather strongly on the nucleation techniques, e.g., solvent exchange gives rise to stable bubbles, while other methods like electrochemical water splitting produce unstable ones. By experimentally investigating a prototypical system of bubble nucleation, we show how these outcomes are determined by a competition between gas oversaturation and contact line friction. We derive a stability line in the oversaturation-radius parameter space, which not only agrees with our experiments but also correctly predicts the outcome of previous experiments across five orders in oversaturation and bubble radius.

1. Introduction

Gas-evolving reactions on liquid-immersed surfaces are ubiquitously encountered in the energy or manufacturing industries, e.g., chlor-alkali and aluminum production [1,2]. Growing fears about the catastrophic effects of global warming have stimulated recent interest in green technologies such as electrolysis and photocatalytic water splitting for the production of hydrogen fuel [3–5]. As a product of gas-evolving reactions, bubbles inhibit future activity by blocking active sites on working surfaces [6–8]. Examples are electrocatalytic reactions [9], electrochemical water splitting [10], and the charging process of zinc-air batteries [11], where bubbles generated during reactions can

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Fig. 1. Experiment setup and dynamic changes of the surface bubble. (a) Schematics of bubble growth mode and experiment setup. Upper panel: the surface bubble first nucleates (dashed line) and grows with constant contact angle growth mode (left), and then changed to constant footprint radius growth mode (right), in which the bubble grows with a constant contact radius when pinned to the contact line. Lower panel: the experimental setup. A coverslip coated with a 50 nm platinum layer serves as a substrate where the decomposition of hydrogen peroxide occurs and generates bubbles, where R presents the curvature of a bubble, L is the footprint radius, and θ is the contact angle at the gas phase. A 100 µL aliquot of 1% hydrogen peroxide was placed on a coverslip sputtered with platinum. The videos were captured through a 60× objective using a high-speed camera from below the cover slip plane at a frame rate of 500 fps. (b) - (i) Dynamic changes of surface bubble nucleation, growth, and contact line pinning. Upon nucleation, the bubble grows proportionally on the surface before stopping its advancement at about $t \sim 1$ s (g). The three-phase contact line of the bubble is highlighted in the dashed yellow line in (f). The scale bar is 50 µm.

hinder active sites and cause overpotentials, further significantly affecting the efficiency of these reactions [12]. Moreover, bubbles covering solid surfaces inevitably affect light absorption in photocatalysis. The attached bubbles reduced the average light absorption on silicon electrodes by 18% [13]. An understanding of how bubbles survive, grow, or dissolve on catalytic surfaces and electrodes is clearly valuable for the design of key green technologies. Contemporaneously, the last few decades have seen intense research activity on nanobubbles, long-lived and small (10–100 nm) surface-attached gas domains [14–16]. The remarkable stability and dynamics of nanobubbles are especially pertinent to gas-evolving reactions since the critical radius of nucleation becomes nanometric once the dissolved gas concentration exceeds several times the saturation concentration [17], a condition easily satisfied by typical gas-evolving reactions.

The fate of nucleated bubbles depends on how they are produced. Bubbles produced by exchanging organic solvents with water are notoriously long-lived and stable, exceeding theoretical expectations by orders of magnitude [18-20]. However, electrochemically generated nanobubbles produced on nanoelectrodes immersed in concentrated electrolyte [21–24] do not survive beyond the application of potential. These observations raise the question of what physical mechanisms determine the fate of bubbles in oversaturated solution. Luo and White simulated a single hydrogen nanobubble nucleated on a platinum nanoelectrode and proposed that the nanobubble is dynamically stabilized when the dissolution driven by the Laplace pressure is balanced by the electrogenerated hydrogen at the partially exposed Pt surface [25]. This modeling is plausible but can hardly apply to catalysts with large active surface area that generate more gases. Ma and coworkers developed a dynamic equilibrium model [26], similar to Brenner and Lohse's previous work [27], based on the balance of influx and outflux, where the influx is driven by gas oversaturation, though the quantity of the oversaturation was not given. Sverdrup et al. also simulated surface nanobubbles with a dynamic equilibrium model that nanobubbles are stabilized under an oversaturation up to ~ 5 [28]. However, the real local oversaturation in chemical reactions would be far more than 5. Yang et al. combined the diffusion theory and the molecular-kinetic model to simulate the origin and growth of gas bubbles on solid surfaces in a supersaturated solution [29]. Yang's simulation aligns somewhat with Chan's [30] report that surface nanobubbles can be stable at contact angles at gas phase smaller than 90 °, but unstable at contact angles larger than 90 °. This work has claimed that the classical Epstein-Plesset diffusion theory can satisfactorily predict the growth of surface bubbles, while the molecular-kinetic model can only fit the floating and expansion stages but not transition stages. More work needs to focus on these transitions.

In this article, we investigated the decomposition of hydrogen peroxide using a noble metal platinum catalysis. This chemical reaction is ubiquitous in nature and was widely used for eliminating sources of hydroxy radicals for water purification [31]. It has been also applied for bubble assisted propulsion of micro- or nanorobots as sensors [32]. To gain insight into the nucleation and further dynamics of bubbles generated during these chemical or electrochemistry reactions, we utilized a flat platinum surface and visualized the *in situ* dynamic changes with high-speed imaging. We hypothesize the existence of a stability line in the oversaturation-radius parameter space that partitions stable from unstable bubbles. We then proceed to prove its existence by experimentally visualizing electrochemically generated bubbles as they transition between the two states.

2. Results and discussion

The dynamics of a single bubble in a typical experiment in 1% hydrogen peroxide is shown in Fig. 1. At the highest framerates investigated, we observe that the bubble's smallest size is at least one pixel, indicating that the critical size at the point of nucleation is smaller than the ~ 300 nm resolution of our $60 \times$ objective, i.e. the nucleate is a nanobubble. After a few frames, a dark and approximately circular ring [highlighted in yellow in Fig. 1(f) t = 0.5 s] can be seen inside the transmission image of the growing bubble. This dark ring is the three-phase contact line of the bubble and the fact that it is smaller than the bubble width indicates that it possesses a contact angle θ exceeding 90°; this allows us to track R and thus the corresponding three-dimensional bubble shape in time. We have triplicated the experiment at the same nucleation site and observed at 3 different spots on the same substrates. The recorded videos did not show much difference at the same position after 5 trials.

Upon nucleation, the bubble experiences a phase of diffusion-limited growth. Here, its footprint radius L advances in the highly gas over-



Fig. 2. Growth of a single bubble in 1% hydrogen peroxide solution. (a) Evolution of the bubble's radius of curvature *R* and footprint radius *L*. (b) The experimental growth curve [black dots; every fifteenth point from (a) is selected] is well-fitted to the Epstein-Plesset equation [red line], with implied oversaturation $\zeta \sim 36.6$. (c) Evolution of R^2 and L^2 against time *t*. The deviation of L^2 from linearity indicates a transition to line pinning. (d) The contact angle θ of the bubble. At early times, the contact angle fluctuates about $\theta \sim 132 \pm 3^{\circ}$ in the gas phase due to several instances of line depinning on surface defects. At t = 1 s, θ starts to rise, indicating the onset of a dynamical transition from constant angle to constant footprint radius dynamics.

saturated environment while its contact angle, calculated by $\sin\theta = \frac{L}{R}$, remains approximately constant, see Fig. 2(d). To relate bubble shape to the evolution of gas we fit R(t) to the solution of the Epstein-Plesset equation [33]

$$\frac{dR}{dt} = \frac{Dc_s\zeta}{\rho} \left(\frac{1}{R} + \frac{1}{\sqrt{\pi Dt}}\right) \tag{1}$$

where *D* is the diffusion coefficient of the gas in the

liquids, c_s is the saturation concentration, and ζ is the gas oversaturation

$$\zeta = \frac{c_{\infty}}{c_s} - 1 \tag{2}$$

through a curve fitting process. Although the Epstein-Plesset equation specifically models a spherical bubble, it can also describe a sessile bubble on a plate once an O(1) correction factor [34,35] has been introduced. Here, the oversaturation ζ is defined in terms of the bulk concentration c_{∞} and solubility c_s . Since the critical nucleation radius of each bubble is smaller than our imaging resolution of 300 nm, the diffusion timescale $\tau \sim l^2/D \sim 1\mu s$ is far smaller than the experimental timescale $t \sim 1$ s. By taking the long time limit $1/\sqrt{\pi Dt} \rightarrow 0$, the experimental data in Fig. 2 can be modelled by an equation of the form

$$R(t) = A\sqrt{t}.$$
(3)

In the case of the bubble in Fig. 1 and its size changes in Fig. 2(a), the least squares fit to Eq. (3) yields an implied oversaturation $\zeta \approx 36.6$. As seen in Fig. 2(b) the fitting is excellent; all the other bubbles we imaged



Fig. 3. Normalized Curvature radii of bubbles $(R(t)/\sqrt{A})$ versus growing with time t. The data are from 12 other randomly selected bubbles in the experiment that showed similar contact line pinning growth.

are similarly well-described by Eq. (3). Fig. 3 shows profiles of normalized bubble curvature radii, calculated by $R(t)/\sqrt{A}$, versus experimental time *t*. It indicates a general agreement among different data sets, a consensus fitting of oversaturation ζ for different bubbles.

After the diffusion-limited growth phase, in which both R^2 and L^2 grow linearly with *t*, L^2 begins to deviate from linearity and the contact



Fig. 4. A stability line in the bulk oversaturation ζ against radius R parameter space predicts the stability of bubble nucleation. From a dynamic balance of oversaturation driven inflation and contact line friction, we hypothesize a stability line [black lines for friction coefficients $\lambda = 0.1$, 0.15, and 0.2 Pa s]. Our stability line not only correctly predicts that our experiments [blue points] achieve stability, but also correctly distinguishes between experiments producing unstable bubbles by electrochemistry [23,24] and nanocatalysts [21], and those producing stable surface-attached bubbles [green squares] characterized by AFM [42,47–49] and optical microscopy [30,50,51].

line comes to a halt. Before its contact line becomes stationary, the bubble's contact angle $\theta = \sin^{-1}(L/R)$ fluctuates about 132° several times, corresponding to 4–5 events during which the contact line jumps over weak surface defects. However, at $t \sim 1$ s the contact angle stops fluctuating about a mean value and abruptly rises. These features characterize a dynamical change from a 'constant angle' mode (i.e. the bubble grows proportionally in shape), to a 'constant footprint radius' mode, in which the contact line is stationary.

Surface nanobubbles formed in stable nucleation experiments are known to observe the latter 'constant radius' mode [36]. Although the onset of contact line pinning is a complex topic on its own right, we will now propose a relatively simple method of predicting the onset of pinning from a dynamical balance at the contact line. Contact line motion is driven by gas influx into the bubble from the supersaturated external environment. From the Epstein-Plesset equation [33] we have contact line velocity

$$u_g = \frac{dL}{dt} \sim \frac{A}{2\sin\theta\sqrt{t}} \tag{4}$$

where A is the proportionality constant from Eq. (3). Eq. (4) implies that the contact line never reaches zero. However, we argue that the contact line will be effectively immobilized if, at some future time t_e , the contact line velocity becomes comparable to the characteristic molecular kinetic theory (MKT) velocity [37]

$$u_i \sim \frac{\gamma}{\lambda} \tag{5}$$

where λ is an experimentally measurable coefficient of friction, which has the units of viscosity (Pa s), and γ is the surface tension. At the MKT velocity, any advancement of the contact line becomes a thermally activated process as opposed to being driven by hydrodynamics. At time t_e , the EP contact line velocity equates to the MKT one.

The dynamical balance outlined above allows us to rationalize the fate of bubbles produced across a parameter space spanning five orders of magnitude in oversaturation ζ and bubble radius R, see Fig. 4. The arguments above allow us to construct stability lines that mark out the expected equilibrium radius of the bubble $R_e = A\sqrt{t_e}$ after it experiences diffusive growth. Direct experimental measurements using independent methods [38–41] suggest $0.1 < \lambda < 0.2$ Pa s in inviscid fluids.

Our proposed stability line successfully partitions a wide variety of

previous experiments across five orders of magnitude in the (ζ , R_e) parameter space, as shown in Fig. 4. While nucleation experiments with Atomic Force Microscopy (AFM) and fluorescence microscopy are known to produce stable surface nanobubbles whose footprint radii do not change after nucleation [42,43], nanobubbles produced by other techniques are generally unstable, including electrocatalysis on Pt nanoelectrodes [21,44]. We have selected only experiments in which the oversaturation ζ can be calculated from tables of standard values [45], or in which ζ has been estimated by the authors. Further, our comparisons consider only experiments on smooth surfaces, omitting studies in which nucleation occurs on structured surfaces. We also neglect the large size limit [46], in which the bubble's buoyancy force $F_b \sim \rho R^3 g$ overcomes substrate pinning, causing it to detach from the substrate altogether.

By observing bubbles as they transition from unstable growth to stable equilibrium, our experiments directly corroborate the existence of this stability line. Each of the bubbles in our experiments starts in the unstable portion of the phase diagram, at an initial radius that is smaller than the optical limit of 300 nm; this is marked with a horizontal black line in Fig. 4. A crude estimate of the critical radius R^* as a function of ζ is Ward's criterion [17,52]

$$R^*(\zeta) \approx \frac{2\gamma}{P_0\zeta} \tag{6}$$

derived by rigorously calculating the chemical potential balance during the nucleation of a single-component gas bubble in a dilute solution; this curve is marked in a dashed line in Fig. 4. The critical radius for bubble nucleation at equilibrium is derived by taking the differential of the Helmholtz free energy to zero, i.e., the gradient of concentration is zero. Thus, we anticipate that, at the point of nucleation, each bubble occupies the part of (ζ, R) phase space bounded by the optical limit and Ward's criterion. In other words, under specific gas supersaturation, only gas nuclei nucleated larger than the critical size at certain oversaturation can continue to grow and remain stable under the action of the contact line pinning. Therefore, based on the stable phase from Fig. 4, the control of the surface bubbles generated can be achieved by adjusting the gas saturation in the solution. Sustained bulk oversaturation can potentially improve the performance of heterogeneous catalysts by eradicating gaseous bubbles from surfaces, thereby preventing the blocking of active sites on the catalyst surface. Bubbles that form during gas-evolving reactions can inhibit catalytic activity by physically blocking the access of reactants to the catalyst surface. By maintaining a high concentration of dissolved gas in the bulk solution, sustained bulk oversaturation can prevent the formation and growth of bubbles on the catalyst surface, allowing for continuous and unimpeded catalytic reactions. Heterogeneous nucleation of bubbles often dominates and arises from sites of impurities or structures with small free energy barriers on surfaces. In this work, the Pt-coated surface exhibited a surface roughness of approximately 1 nm. According to Fig. 4, coupling larger friction coefficients into the model demonstrated that equilibrium stability lines were more easily achieved, indicating that friction forces opposing bubble expansion would alter the dynamic equilibrium of the gas bubble at the contact line. Therefore, bubbles nucleated at sites having the lowest free energy when dissolved gas is oversaturated, and surface bubble nucleation induced by this oversaturated solution on uneven surfaces is more stable than on smooth surfaces.

We have also carried out preliminary experiments on surfaces of differing wettability. In a separate experiment, we pre-treated the platinum-sputtered coverslip with oxygen plasma, upon which a droplet of our hydrogen peroxide solution has a contact angle of $15 \pm 3^{\circ}$ in dense phase. Interestingly, the bubble's growth rate on the hydrophilic surface is approximately 20% faster than on the untreated platinum surfaces (contact angle of $90 \pm 4^{\circ}$). On that hydrophilic surface, once nucleated, bubbles are usually evolved into constant pinned footprint growth when

the EP contact line velocity u_g equates to the MKT velocity u_i . When two bubbles merged into a bigger one, they lost their original position, were unpinned and detached from the surface. The hydrophilic property of surface results in a relatively small pinned radius and a small pinning force so that the detachment easily occurs while the bubble is growing big enough. On the other hand, bubbles nucleated on an untreated platinum surface preferred to grow with constant contact angles. When two bubbles merged, they did not detach due to the large contact area and thus, the large pinning force. Interestingly, we also observed that the bubble growth mode changed from constant footprint to constant contact angle when the fresh hydrophilic surface was used for ~ 1 hour for an unknown reason. The afterwards contact angle measurement shows that they became hydrophobic. Probably, oxygen generation passivates the surface and presents it hydrophobic, on which we are more often to observe a bubble growing in the constant contact angles.

3. Conclusions

In summary, we have experimentally investigated a prototypical electrochemical system in which gas bubbles nucleate from electrodes and observe the transition of each bubble from unstable growth to a stable equilibrium. We hypothesized the existence of a stability line in the oversaturation-radius parameter space based on a dynamic balance between the contact line speed induced by ambient gas oversaturation and contact line friction.

The stability line not only correctly anticipates that the bubbles in our experiments will attain stability but is also consistent with a large number of previous experiments. A notable practical consequence of our findings is that virtually all gaseous bubbles can be eradicated from surfaces by the application of sustained bulk oversaturation, providing a path towards improving the performance of heterogeneous catalysts. We note that this condition is achievable, for example, by carrying out gas evolving electrochemical or stoichiometric reactions in concentrated electrolytes. A cursory inspection of our phase diagram in Fig. 4 reveals that in the large oversaturation limit the dynamic balance between inflation growth and contact line friction is no longer viable at any size. Sessile bubbles subject to excess bulk oversaturation should gorge gas much too quickly and detach, leaving behind a bare surface.

4. Experimental

To produce bubbles, a 50 nm layer of platinum was sputtered onto a clean glass coverslip (no. 1, Menzel-Gläser, Germany). A stock solution (30%, Chem-supply, Australia) was diluted with deionized water into different concentrations of 0.5%, 1%, and 3%. Then, a droplet of diluted hydrogen peroxide solution was deposited onto the Pt-coated coverslip. This experiment was conducted at a laboratory ambient temperature of 20 °C. As the substrate is catalytically active, the hydrogen peroxide solution spontaneously decomposes, producing a large concentration of dissolved oxygen at the coverslip, which, in turn, drives continuous nucleation of oxygen bubbles across the surface. The gas oversaturation is tuned in our experiment by adjusting the concentration of the solution. After the pretrial experiment, we used the optimal concentration of 1% of hydrogen peroxide. Based on our observation, in the 0.5% group, the bubble grew very slowly, and it took a long time to capture the video. In the 3% group, bubbles grew quickly, coalesced, and detached rapidly. Once floating in the solutions, these bubbles block the illumination light, challenging the image analysis as the footprint became blurred. We chose a concentration of 1% to elaborate on bubble dynamics. To capture the contact line dynamics of the bubble, we use a 60 \times oil immersion objective with a numerical aperture of 1.49 (Olympus, Japan), and image the resulting bubbles on an inverted microscope (IX71, Olympus) through a high-speed camera (SA5, Photron, U.S.) at frame rates ranging from 500 to 20,000 fps. The experiment was replicated at least for 3 times at the same nucleation site and 3 different spots on the same substrates for each group. More than 3 Pt-coated surfaces were



Fig. 5. The scheme of the experiment setup. 100 μ l of a certain concentration of hydrogen peroxide solution was placed on a coverslip coated with a 50 nm platinum layer. An Olympus APO N $60\times/NA1.49$ oil immersion TIRF objective lens was used to capture the nucleation and growth of surface bubbles equipped with a high-speed camera.

used in the experiments. The scheme of the experiment setup is shown in Fig. 5.

CRediT authorship contribution statement

Qingyun Zeng: Writing – review & editing. Beng Hau Tan: Conceptualization, Formal analysis, Methodology, Writing – review & editing. Hongjie An: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Supervision, Visualization, Writing – original draft, Writing – review & editing. Nam-Trung Nguyen: Resources, Writing – review & editing. Lingxi Ouyang: Data curation, Methodology, Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Hongjie An reports financial support was provided by Australian Research Council. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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References

- R.K. Karlsson, A. Cornell, Selectivity between oxygen and chlorine evolution in the chlor-alkali and chlorate processes, Chem. Rev. 116 (2016) 2982–3028.
- [2] G.G. Botte, Electrochemical manufacturing in the chemical industry, Electrochem. Soc. Interface 23 (2014) 49.
- [3] J. Zhu, L. Hu, P. Zhao, L.Y.S. Lee, K.-Y. Wong, Recent advances in electrocatalytic hydrogen evolution using nanoparticles, Chem. Rev. 120 (2019) 851–918.
- [4] K. Maeda, K. Domen, Photocatalytic water splitting: recent progress and future challenges, J. Phys. Chem. Lett. 1 (2010) 2655–2661.

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- [5] S.S. Kumar, V. Himabindu, Hydrogen production by PEM water electrolysis A review, Mater. Sci. Energy Technol. 2 (2019) 442–454.
- [6] A. Angulo, P. van der Linde, H. Gardeniers, M. Modestino, D.F. Rivas, Influence of bubbles on the energy conversion efficiency of electrochemical reactors, Joule 4 (2020) 555–579.
- [7] R. Iwata, L. Zhang, K.L. Wilke, S. Gong, M. He, B.M. Gallant, E.N. Wang, Bubble growth and departure modes on wettable/non-wettable porous foams in alkaline water splitting, Joule 5 (2021) 887–900.
- [8] X. Zhao, H. Ren, L. Luo, Gas bubbles in electrochemical gas evolution reactions, Langmuir 35 (2019) 5392–5408.
- [9] A.E. Angulo, D. Frey, M.A. Modestino, Understanding bubble-induced overpotential losses in multiphase flow electrochemical reactors, Energy Fuels 36 (14) (2022) 7908–7914.
- [10] Y. Chen, J. Chen, K. Bai, Z. Xiao, S. Fan, A flow-through electrode for hydrogen production from water splitting by mitigating bubble induced overpotential, J. Power Sources 561 (2023) 232733.
- [11] Y. He, Y. Cui, W. Shang, Z. Zhao, P. Tan, Insight into the bubble-induced overpotential towards high-rate charging of Zn-air batteries, Chem. Eng. J. 448 (2022) 137782.
- [12] A.R. Zeradjanin, P. Narangoda, I. Spanos, J. Masa, R. Schlögl, How to minimise destabilising effect of gas bubbles on water splitting electrocatalysts?, Current Opinion in, Electrochemistry 30 (2021) 100797.
- [13] A. Bhanawat, K. Zhu, L. Pilon, How do bubbles affect light absorption in photoelectrodes for solar water splitting? Sustain. Energy Fuels 6 (3) (2022) 910–924.
- [14] B.H. Tan, H. An, C.-D. Ohl, Stability of surface and bulk nanobubbles, Curr. Opin. Colloid Interface Sci. 53 (2021) 101428.
- [15] B.H. Tan, H. An, C.-D. Ohl, Surface nanobubbles are stabilized by hydrophobic attraction, Phys. Rev. Lett. 120 (16) (2018) 164502.
- [16] B.H. Tan, H. An, C.-D. Ohl, Stability, dynamics, and tolerance to undersaturation of surface nanobubbles, Phys. Rev. Lett. 122 (13) (2019) 134502.
- [17] C. Ward, A. Balakrishnan, F. Hooper, On the thermodynamics of nucleation in weak gas-liquid solutions, J. Basic Eng. 92 (1970) 695.
- [18] D. Lohse, X. Zhang, Surface nanobubbles and nanodroplets, Rev. Mod. Phys. 87 (2015) 981.
- [19] J.R. Seddon, D. Lohse, Nanobubbles and micropancakes: gaseous domains on immersed substrates, J. Phys. Condens. Matter 23 (2011) 133001.
- [20] B.H. Tan, C.-D. Ohl, H. An, Transient solubility gradients mediate oversaturation during solvent exchange, Phys. Rev. Lett. 126 (2021) 234502.
- [21] Al.M. Soto, S.R. German, H. Ren, D. Van Der Meer, D. Lohse, M.A. Edwards, H. S. White, The nucleation rate of single O2 nanobubbles at Pt nanoelectrodes, Langmuir 34 (2018) 7309–7318.
- [22] M.A. Edwards, H.S. White, H. Ren, Voltammetric determination of the stochastic formation rate and geometry of individual H2, N2, and O2 bubble nuclei, ACS nano 13 (2019) 6330–6340.
- [23] S.R. German, M.A. Edwards, Q. Chen, H.S. White, Laplace pressure of individual H2 nanobubbles from pressure–addition electrochemistry, Nano Lett. 16 (2016) 6691–6694.
- [24] V.B. Svetovoy, R.G. Sanders, M.C. Elwenspoek, Transient nanobubbles in shorttime electrolysis, J. Phys. Condens. Matter 25 (2013) 184002.
- [25] L. Luo, H.S. White, Electrogeneration of single nanobubbles at sub-50-nm-radius platinum nanodisk electrodes, Langmuir 29 (35) (2013) 11169–11175.
- [26] Y. Ma, Z. Guo, Q. Chen, X. Zhang, Dynamic equilibrium model for surface nanobubbles in electrochemistry, Langmuir 37 (8) (2021) 2771–2779.
- [27] M.P. Brenner, D. Lohse, Dynamic equilibrium mechanism for surface nanobubble stabilization, Phys. Rev. Lett. 101 (21) (2008) 214505.
- [28] K. Sverdrup, S.-J. Kimmerle, P. Berg, Computational investigation of the stability and dissolution of nanobubbles, Appl. Math. Model. 49 (2017) 199–219.

- [29] S. Yang, S. Bao, C. Liu, D. Yuan, W. Huang, An analytical model of the growth of invisible bubbles on solid surfaces in a supersaturated solution, Chem. Eng. Sci. 215 (2020) 114968.
- [30] C.U. Chan, M. Arora, C.-D. Ohl, Coalescence, growth, and stability of surfaceattached nanobubbles, Langmuir 31 (25) (2015) 7041–7046.
- [31] A.A. Solovev, S. Sanchez, M. Pumera, Y.F. Mei, O.G. Schmidt, Magnetic control of tubular catalytic microbots for the transport, assembly, and delivery of microobjects, Adv. Funct. Mater. 20 (15) (2010) 2430–2435.
- [32] J.G.S. Moo, H. Wang, G. Zhao, M. Pumera, Biomimetic artificial inorganic enzymefree self-propelled microfish robot for selective detection of Pb2+ in water, Chem. – A Eur. J. 20 (15) (2014) 4292–4296.
- [33] P.S. Epstein, M.S. Plesset, On the stability of gas bubbles in liquid-gas solutions, J. Chem. Phys. 18 (11) (1950) 1505–1509.
- [34] T. Yamashita, K. Ando, Aeration of water with oxygen microbubbles and its purging effect, J. Fluid Mech. 825 (2017) 16–28.
- [35] O.R. Enríquez, D. Lohse, A. Prosperetti, C. Sun, D. van der Meer, The quasi-static growth of CO2 bubbles, J. Fluid Mech. 741 (2014) R1.
- [36] X. Zhang, D.Y. Chan, D. Wang, N. Maeda, Stability of interfacial nanobubbles, Langmuir 29 (2013) 1017–1023.
- [37] D. Duvivier, T.D. Blake, J. De Coninck, Toward a predictive theory of wetting dynamics, Langmuir 29 (2013) 10132–10140.
- [38] A. Carlson, G. Bellani, G. Amberg, Universality in dynamic wetting dominated by contact-line friction, Phys. Rev. E 85 (2012) 045302.
- [39] A. Carlson, G. Bellani, G. Amberg, Contact line dissipation in short-time dynamic wetting, Europhys. Lett. 97 (2012) 44004.
- [40] N. Kumari, V. Bahadur, S. Garimella, Electrical actuation of dielectric droplets, J. Micromech. Microeng. 18 (2008) 085018.
- [41] Q. Vo, H. Su, T. Tran, Universal transient dynamics of electrowetting droplets, Sci. Rep. 8 (2018) 836.
- [42] J. Qian, V.S. Craig, M. Jehannin, Long-term stability of surface nanobubbles in undersaturated aqueous solution, Langmuir 35 (2018) 718–728.
- [43] H. An, B.H. Tan, C.-D. Ohl, Distinguishing nanobubbles from nanodroplets with AFM: the influence of vertical and lateral imaging forces, Langmuir 32 (2016) 12710–12715.
- [44] Q. Chen, L. Luo, H. Faraji, S.W. Feldberg, H.S. White, Electrochemical measurements of single H2 nanobubble nucleation and stability at Pt nanoelectrodes, J. Phys. Chem. Lett. 5 (2014) 3539–3544.
- [45] R. Battino, T.R. Rettich, T. Tominaga, The solubility of nitrogen and air in liquids, J. Phys. Chem. Ref. Data 13 (1984) 563–600.
- [46] P. Lv, H. Le The, J. Eijkel, A. Van den Berg, X. Zhang, D. Lohse, Growth and detachment of oxygen bubbles induced by gold-catalyzed decomposition of hydrogen peroxide, J. Phys. Chem. C. 121 (2017) 20769–20776.
- [47] H. An, B.H. Tan, Q. Zeng, C.-D. Ohl, Stability of nanobubbles formed at the interface between cold water and hot highly oriented pyrolytic graphite, Langmuir 32 (2016) 11212–11220.
- [48] M. Liu, W. Zhao, S. Wang, W. Guo, Y. Tang, Y. Dong, Study on nanobubble generation: saline solution/water exchange method, ChemPhysChem 14 (2013) 2589–2593.
- [49] L.-M. Zhou, S. Wang, J. Qiu, L. Wang, X.-Y. Wang, B. Li, L.-J. Zhang, J. Hu, Interfacial nanobubbles produced by long-time preserved cold water, Chin. Phys. B 26 (2017) 106803.
- [50] S. Karpitschka, E. Dietrich, J.R. Seddon, H.J. Zandvliet, D. Lohse, H. Riegler, Nonintrusive optical visualization of surface nanobubbles, Phys. Rev. Lett. 109 (2012) 066102.
- [51] N. Hain, D. Wesner, S.I. Druzhinin, H. Schönherr, Surface nanobubbles studied by time-resolved fluorescence microscopy methods combined with AFM: the impact of surface treatment on nanobubble nucleation, Langmuir 32 (2016) 11155–11163.
- [52] C. Ward, A. Tucker, Thermodynamic theory of diffusion- controlled bubble growth or dissolution and experimental examination of the predictions, J. Appl. Phys. 46 (1975) 233–238.