



Research Paper

Cyclic computation of bubble growth in water electrolysis using a level-set approach

Jaesung Park, Gihun Son*

Research Institute for Smart Design & Manufacturing Technology, Department of Mechanical Engineering, Sogang University, 35 Baekbeom-ro, Mapo-gu, Seoul 04107, South Korea

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ABSTRACT

A level-set method is extended to resolve cyclic hydrogen bubble growth during water electrolysis by incorporating microlayer evaporation and interfacial mass transfer. The proposed formulation accounts for hydrogen transport from the cathode through the electrolyte and microlayer, as well as vapor phase change, while accurately capturing spatial and temporal variations in microlayer thickness. Repetitive nucleation, growth, and detachment processes are computed over successive cycles until a quasi-periodic state is reached. The developed numerical framework reproduces bubble growth behaviors consistent with experimental observations when a variable contact-angle model is applied. Cyclic simulations reveal that dissolved hydrogen accumulated during bubble growth and remaining after bubble departure significantly affects subsequent growth behavior when the electrode size exceeds a certain characteristic length scale, whereas its influence remains limited for small electrodes. The cyclic behavior of bubble growth is analyzed in detail, and the influences of key parameters — including electrode size, current density, wall temperature, and contact angle — on hydrogen production and hydrogen evolution efficiency are systematically investigated. The results indicate that cyclic computation combined with microlayer transport modeling is essential for quantitatively predicting electrolysis performance under practical operating conditions.

1. Introduction

Hydrogen has gained increasing attention as a clean and efficient energy carrier, owing to its carbon-free nature and high energy density. In the context of sustainable hydrogen production, water electrolysis has attracted significant attention as one of the most mature and carbon-free technologies available [1]. Electrolytic bubbles nucleate at the cathode surface and grow continuously during electrochemical reactions, thereby promoting the removal of hydrogen species dissolved near the active reaction zone [2], while also inducing micro-convective flow [3] that enhances electrolysis performance. However, these bubbles can also reduce electrochemical efficiency by covering active electrode areas and hindering ion and electron transport [4, 5]. Therefore, understanding bubble dynamics in water electrolysis, particularly in relation to mass transfer, is essential for improving overall conversion efficiency. Despite considerable interest in electrolytic bubble behavior, a general predictive model that accurately describes transient growth and departure of hydrogen bubbles under practical electrochemical conditions has not yet been established.

Most numerical studies [6–9] have investigated hydrogen bubble growth in water electrolysis using fixed-diameter bubble assumptions.

Weier et al. [6] modeled an electrolytic bubble as a rigid sphere positioned above a horizontal electrode under steady-state conditions and computed the induced flow field generated by an electrode-normal magnetic field. Liu et al. [7] introduced a pair of anchored bubbles with fixed diameters to simulate both the steady-state magnetohydrodynamic interactions among neighboring bubbles and their influence on bubble departure. Massing et al. [8] employed unsteady governing equations to investigate thermocapillary-induced flow generated along the surface of a stationary spherical bubble on a microelectrode surrounded by an insulating region. While these fixed-diameter models offer useful insights into localized transport behavior, they are inherently limited in capturing the continuous bubble growth driven by supersaturated dissolved hydrogen and the transient interfacial dynamics that govern bubble shape evolution and motion. [10,11].

Interface-tracking methods, particularly the volume-of-fluid (VOF) method [12,13], which are widely recognized for their capability to capture time-dependent interfacial dynamics, have been rarely applied to hydrogen bubble growth during water electrolysis [14,15]. Liu et al. [14] developed a VOF-based numerical approach coupled with Sherwood number correlations to model hydrogen diffusion toward the

* Corresponding author.

E-mail address: gihun@sogang.ac.kr (G. Son).<https://doi.org/10.1016/j.applthermaleng.2026.131570>

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Nomenclature**Variables**

a	sound speed
c	specific heat at constant volume
C_H	Henry's constant
$C_{1/2/3}$	Antoine's coefficients
D_l	diffusivity
F	Faraday's constant
G	mass flux
H_b	heaviside function
i_{lv}	latent heat
i_w	electric current density
M	Molar mass
m	mass
$\mathbf{n}$	normal vector
p	pressure
R	radius
R_g	gas constant
r	radial coordinate
S	sign function
T	temperature
t	time
$\mathbf{u}$	flow velocity
Y	mass fraction
y	axial coordinate

Greek Symbols

α	smooth step function
γ_a	accommodation coefficient
Δ	increment
δ_l	microlayer thickness
η	hydrogen evolution efficiency
κ	interfacial curvature
λ	thermal conductivity
μ	viscosity
ϕ	level-set function
ρ	density
σ	surface tension coefficient
τ^*	fictitious time step
τ	strain tensor

Subscripts

b, l	bubble, liquid
d	departure
e	electrode
f	fluid
g, h, v	gas, hydrogen, vapor
m	microlayer
o	initial
s	surface
sat	saturation
t	total
w	wall
∞	ambient

to capture the evolution and detachment process of an individual hydrogen bubble. However, these studies considered only the mass flux at the bubble interface and did not account for interfacial mass transfer occurring within the microlayer beneath the bubble—a feature well established in boiling and also reported in electrolysis [16].

Microlayer evaporation has been more extensively studied through numerical methods in boiling than in water electrolysis [17–20]. Son et al. [17] implemented a level-set (LS) framework to resolve bubble growth during boiling, explicitly modeling evaporation processes in the thin microlayer. Sato and Niceno [19] proposed a generalized model of nucleate boiling capable of dynamically predicting microlayer thinning and associated evaporation effect. Considering the analogy between boiling and electrolytic bubble growth [3], these studies provide valuable insight for modeling interfacial transport phenomena during electrolysis [10,11]. Zhan et al. [10] introduced a direct injection model that assumed a virtual microlayer and incorporated hydrogen influx across a thin liquid film to simulate electrolytic bubble growth. However, they did not consider phase-change heat transfer or vapor flux, whereas in electrolysis, both phase-change heat transfer and evaporative vapor flux play important roles in governing bubble dynamics [11]. Recently, Park et al. [11] developed a LS based approach that accounts for evaporation and hydrogen transport within the microlayer, utilizing the microlayer modeling framework proposed by Sato and Niceno [19]. They reported that evaporative heat transfer in the microlayer, along with the associated vapor phase-change flux, plays a critical role in influencing the bubble departure cycle and electrolysis performance. Under practical electrolysis conditions, bubble growth and detachment occur in a periodic manner. However, numerical models that simultaneously capture both microlayer evaporation and cyclic bubble dynamics are still extremely limited, hindering accurate performance prediction.

Cyclic simulations of electrolyte bubble growth [21,22] have been performed in recent years for water electrolysis. Van der Linde et al. [21] modeled the successive bubble growth generated at a hydrophobic pit located at the tip of a single micropillar, assuming a time-dependent spherical hydrogen bubble shape. Raman et al. [22] simulated seven successive hydrogen bubbles that grew and detached from a central hydrophobic pit positioned inside a ring-shaped microelectrode using the arbitrary Lagrangian-Eulerian method. The simulated bubble growth profiles were compared against their previously reported experimental data [23] under various constant current conditions. However, their numerical approach employed a cutting protocol that forced bubble detachment at a prescribed departure radius and did not account for microlayer evaporation, thereby limiting its applicability to spatially decoupled bubble growth on ring-shaped microelectrodes. To the best of our knowledge, a general numerical framework capable of simultaneously resolving microlayer evaporation and cyclic bubble dynamics in water electrolysis has not yet been established.

In this work, we extend our previous LS method for interfacial phase-change problems [11,24,25] to simulate cyclic hydrogen bubble growth during water electrolysis by incorporating microlayer evaporation. The proposed model accounts for hydrogen mass transfer from the cathode through the electrolyte and microlayer, as well as evaporative mass transfer, to realistically represent electrolysis conditions in which phase-change heat transfer must be considered. Our model is further extended to capture the repetitive sequence of bubble nucleation, growth, and departure, thereby enabling a more realistic representation of cyclic bubble dynamics in electrolysis. The cyclic behavior is analyzed in detail, and the resulting hydrogen production rate and hydrogen evolution efficiency are quantitatively evaluated. Parametric studies are further conducted to examine the influences of electrode size, current density, wall temperature, and contact angle on system performance. In particular, the wall temperature acts as a key parameter that simultaneously affects the bubble growth rate and internal composition. Therefore, incorporating phase-change heat transfer is essential for simulating high-temperature electrolysis and practical electrolysis systems with significant temperature variations.

bubble interface. Vachaparambil and Einarsrud [15] extended the VOF method within the OpenFOAM framework by incorporating electrochemical reaction kinetics and Fick's law-based interfacial mass transfer

2. Numerical analysis

The previously proposed LS method [11,24,25] is further developed to facilitate the analysis of cyclic bubble growth in water electrolysis by incorporating microlayer evaporation beneath the bubble. The bubble–liquid interface is captured using a signed-distance LS function, ϕ_b , which represents the shortest distance from the interface with its sign indicating the fluid phase [24,26], rather than using the VOF method commonly adopted for time-dependent interfacial flows. The LS method offers advantages in the smooth and accurate evaluation of interfacial geometrical quantities, such as the interface curvature and normal vectors, resulting in more reliable computation of surface tension forces. Accurate evaluation of these quantities is essential for resolving the balance between buoyancy and surface tension, which governs the bubble departure radius and growth cycle and therefore plays a critical role in cyclic electrolysis simulations and performance prediction. A hydrogen bubble undergoes growth and departure on a circular electrode with radius R_e , as illustrated in Fig. 1(a). An axisymmetric laminar flow is assumed, and the liquid phase is treated as incompressible, while the dissolved hydrogen mass fraction Y_l is resolved. The gas inside the bubble is treated as a spatially uniform binary ideal mixture, where the hydrogen mass fraction $Y_b = Y_b(t)$ evolves in time. The bubble pressure p_b is evaluated from a state relation derived in accordance with Dalton's law:

$$p_b = \rho_b [Y_b R_{g,h} + (1 - Y_b) R_{g,v}] T_b \quad (1)$$

where R_g denotes the specific gas constant, with subscripts h and v indicating hydrogen and water vapor, respectively.

2.1. Governing equations for hydrogen bubble growth

The governing equations for mass, momentum, and energy are formulated separately for the gas and liquid phases. In addition, the dissolved hydrogen mass fraction Y_l in the liquid phase is governed by a species-transport equation. These equations are summarized as follows:

$$\frac{\partial \rho_f}{\partial t} + \nabla \cdot \mathbf{u}_f \rho_f = 0 \quad (2)$$

$$\frac{\partial \rho_f \mathbf{u}_f}{\partial t} + \nabla \cdot (\mathbf{u}_f \rho_f \mathbf{u}_f) = -\nabla p_f + \nabla \cdot \boldsymbol{\tau}_f \quad (3)$$

$$\frac{\partial \rho_f c_f T_f}{\partial t} + \nabla \cdot (\rho_f \mathbf{u}_f c_f T_f) = \nabla \cdot \lambda_f \nabla T_f + s_{T,f} \quad (4)$$

$$\frac{\partial \rho_l Y_l}{\partial t} + \nabla \cdot (\mathbf{u}_l \rho_l Y_l) = \nabla \cdot \rho_l D_l \nabla Y_l \quad (5)$$

where

$$\boldsymbol{\tau}_f = \mu_f \left[\nabla \mathbf{u}_f + \nabla \mathbf{u}_f^T - \frac{2}{3} (\nabla \cdot \mathbf{u}_f) \mathbf{I} \right] \quad (6)$$

$$s_{T,f} = -\nabla \cdot (\mathbf{u}_f p_f) + \boldsymbol{\tau}_f : \nabla \mathbf{u}_f \quad (7)$$

Here, $f = b$ in the bubble region ($\phi_b \geq 0$) and $f = l$ in the liquid region ($\phi_b < 0$). Coupling between the bubble and liquid regions is enforced through interfacial conditions applied at the bubble boundary defined by $\phi_b = 0$, as described below.

$$\mathbf{u}_{ls} - \mathbf{u}_{bs} = \nu_{lb} G_t \mathbf{n}_l \quad (8)$$

$$\mathbf{n}_l \cdot [-(p_{ls} - p_{bs}) \mathbf{I} + \boldsymbol{\tau}_{ls} - \boldsymbol{\tau}_{bs}] = (-\sigma_{bl} \kappa_b - \nu_{lb} G_t^2) \mathbf{n}_l \quad (9)$$

$$T_{ls} = T_{bs} = T_s \quad (10)$$

$$Y_{ls} = \frac{C_H \rho_{bs} Y_b}{\rho_l} \quad (11)$$

where

$$\nu_{lb} = \rho_{bs}^{-1} - \rho_l^{-1} \quad (12)$$

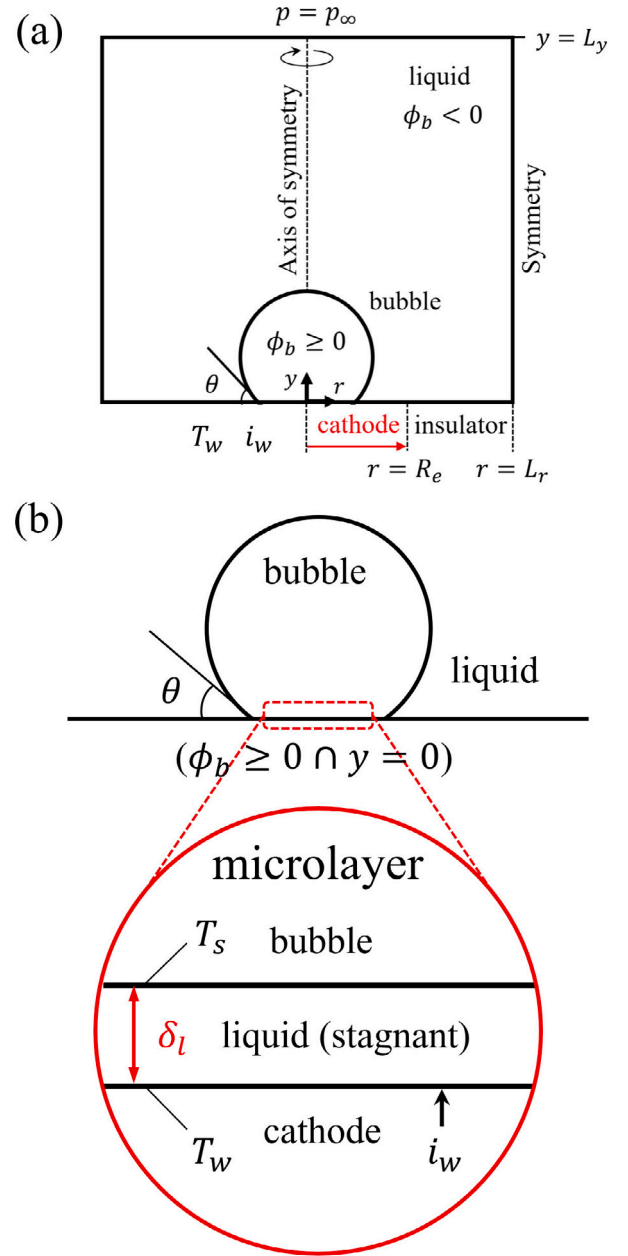


Fig. 1. Schematic of the simulation setup: (a) bubble evolution on a partially insulated electrode, and (b) microlayer formed beneath the bubble adjacent to the cathode.

$$\kappa_b = -\nabla \cdot \mathbf{n}_l \quad (13)$$

$$\mathbf{n}_l = -\nabla \phi_b / |\nabla \phi_b| \quad (14)$$

Here, κ_b is the bubble–liquid interfacial curvature computed from ϕ_b , G_t is the total mass flux, C_H is the Henry's law coefficient and the subscript s indicates the bubble surface. The bubble surface temperature T_s is iteratively calculated based on Eqs. (24) and (27), as described in Section 2.2. To determine Y_{ls} from Eq. (11), the time-dependent Y_b is updated over time using the hydrogen mass flux G_h as

$$\frac{d}{dt} \left(\int \rho_b Y_b dV_b \right) = \int G_h dA_b \quad \text{if } \phi_b \geq 0 \quad (15)$$

For the bubble–liquid two-phase regions, Eqs. (2) and (3) are reformulated considering compressibility in the bubble phase [24,27], as

$$\nabla \cdot \mathbf{u} = -\frac{H_b}{\rho_b a_b^2} \frac{Dp}{Dt} - v_{lb} G_t \mathbf{n}_l \cdot \nabla H_b \quad (16)$$

$$\begin{aligned} \frac{\partial \rho \mathbf{u}}{\partial t} + \nabla \cdot (\rho \mathbf{u} \mathbf{u}) &= -(\nabla p - \sigma_{bl} \kappa_b \nabla H_b + v_{lb} G_t^2 \nabla H_b) \\ + \nabla \cdot \mu \left[\nabla \mathbf{u} + \nabla \mathbf{u}^T - \frac{2}{3} (\nabla \cdot \mathbf{u}) \mathbf{I} \right] \end{aligned} \quad (17)$$

where

$$\rho = \rho_b \alpha_b + \rho_l (1 - \alpha_b) \quad (18)$$

$$\mu = \mu_b \alpha_b + \mu_l (1 - \alpha_b) \quad (19)$$

$$\begin{aligned} H_b &= 1 & \text{if } \phi_b \geq 0 \\ &= 0 & \text{if } \phi_b < 0 \end{aligned} \quad (20)$$

$$\alpha_b = \min \left[1, \max \left(0, \frac{\phi_b}{h} + \frac{1}{2} \right) \right] \quad (21)$$

Here, α_b is a smooth step function varying linearly across the interfacial thickness, in contrast to the discontinuous step function H_b , and h is the grid spacing.

The governing equations are discretized using a finite-volume framework, where spatial derivatives are evaluated on a staggered-grid system. Diffusion terms are discretized with second-order central differencing, while advection is handled using essentially non-oscillatory schemes. Temporal integration is performed in a fractional-step manner [24,25]. The fluid density ρ_f in the mass Eq. (2) is solved using the ghost fluid method [24,28] to accurately resolve discontinuities across the gas–liquid interface. Further details on the discretization and solution procedures are presented in our previous studies [24,25,29].

2.2. Mass fluxes of vapor and H_2 at the bubble interface

The hydrogen and total mass fluxes, G_h and G_t , at the interface given in Eqs. (15)–(17) can be expressed using the relation $G_t = G_v + G_h$ as

$$G_h = G_t Y_{ls} + \rho_l D_l \nabla Y_l \cdot \mathbf{n}_l \quad (22)$$

$$G_t = \frac{1}{1 - Y_{ls}} (G_v + \rho_l D_l \nabla Y_l \cdot \mathbf{n}_l) \quad (23)$$

Here, G_v denotes the vapor mass flux, which is evaluated from the local thermal balance at the phase boundary [24,25], as expressed in Eq. (24), and is governed by the bubble-surface temperature T_s . To determine both T_s and G_v , an additional mass-flux expression based on gas kinetic theory [30–32] is introduced and coupled, as follows:

$$G_v = \frac{1}{i_{lv}} [\lambda_l \nabla T_l - \lambda_b \nabla T_b] \cdot \mathbf{n}_l = \frac{2\gamma_a}{(2 - \gamma_a)} \frac{(p_{v,sat} - p_{vs})}{\sqrt{2\pi R_{g,v} T_s}} \quad (24)$$

where

$$p_{v,sat} = \exp \left(C_1 - \frac{C_2}{T_s + C_3} \right) \quad (25)$$

$$p_{vs} = \rho_{bs} (1 - Y_b) R_{g,v} T_s \quad (26)$$

where $p_{v,sat}$ denotes the vapor saturation pressure and p_{vs} corresponds to the interfacial vapor pressure. Note that both sides of Eq. (24) are implicit with respect to T_s . This implies that the phase-change model based on the heat flux balance formulation in Eq. (24), combined with the gas kinetic relation, provides a generalized framework applicable to evaporation processes in which the interfacial temperature deviates from the saturation temperature. However, it should be noted that the accommodation coefficient γ_a in Eq. (24) must be prescribed a priori, and its accurate determination remains challenging [33,34].

Once T_s is iteratively obtained from Eq. (24), G_v , G_t and G_h are subsequently calculated using Eqs. (22)–(24). In the present study, where the temperature field remains relatively smooth, the numerical results exhibit only weak sensitivity to the accommodation coefficient γ_a within the range $0.2 \leq \gamma_a \leq 0.8$, showing only minor differences in the predicted bubble growth behavior. Accordingly, an intermediate value of $\gamma_a = 0.5$ is adopted in this work. This value has also been widely employed in previous numerical studies involving various engineering applications, including boiling, electrolysis, and acoustic cavitation processes [11,30–32,35].

Similar to boiling phenomena [36], the liquid microlayer beneath electrolytic bubbles has been reported to contribute significantly to mass transfer [16]. To incorporate the effect of microlayer evaporation, as illustrated in Fig. 1(b), the concept of a depletable microlayer introduced by Sato and Niceno [19] is adopted. Assuming that the liquid in the microlayer region is stagnant [18,19], the microlayer vapor flux G_{vm} is modeled as:

$$G_{vm} = \frac{1}{i_{lv}} \left(\lambda_l \frac{T_w - T_s}{\delta_l} \right) = \frac{2\gamma_a}{(2 - \gamma_a)} \frac{(p_{v,sat} - p_{vs})}{\sqrt{2\pi R_{g,v} T_s}} \quad (27)$$

where T_w denotes the temperature of the cathode wall, and δ_l denotes the microlayer thickness. It should be noted that the microlayer region is assumed to remain stagnant even under cyclic bubble growth conditions. Unlike boiling phenomena, where vigorous bubble dynamics may challenge this assumption, the relatively weaker bubble motion and wake-induced convection under the present electrolysis conditions are not sufficient to overcome the strong viscous resistance within the thin microlayer formed between the bubble and the electrode surface. Since the microlayer thickness is typically on the order of only a few micrometers, the liquid film behaves as a highly resistive diffusion-dominated layer, thereby supporting the validity of the stagnant microlayer assumption [18,19,37,38]. To update the temporal evolution of δ_l , a mass balance is constructed over the liquid region confined beneath the bubble, as illustrated in Fig. 1(b), and is expressed as [11,39]:

$$\frac{d\delta_l}{dt} = -\frac{G_{tm}}{\rho_l} \quad (28)$$

Here, δ_{l0} denotes the initial microlayer thickness. In the present study, the predicted bubble growth behavior shows only minor differences within the range $1 \mu\text{m} \leq \delta_{l0} \leq 9 \mu\text{m}$. Accordingly, a value of $\delta_{l0} = 9 \mu\text{m}$ is adopted to minimize the possibility of microlayer dryout caused by continuous evaporation. This choice is consistent with previous experimental and numerical studies [9,40], which reported that the initial microlayer thickness is typically on the order of $1 \sim 10 \mu\text{m}$. G_{tm} denotes the total microlayer flux, which can be obtained by extending Eq. (23) as

$$G_{tm} = \frac{1}{1 - Y_{ls}} \left(G_{vm} + \frac{M_h}{2F} i_w \right) \quad (29)$$

where F is Faraday's constant, i_w is the current density, and M_h is the hydrogen molar mass. It should be noted that the second term on the right-hand side of Eq. (29) represents the hydrogen gas generated at the cathode surface and directly transported to the bottom bubble interface ($\phi_b \geq 0$ and $r \leq R_e$) through the liquid microlayer, where the electrochemical hydrogen generation rate is expressed as $-\rho_l D_l (\partial Y_l / \partial y)_{y=0} = M_h i_w / (2F)$, as presented in Eq. (36). Although the overall numerical formulation follows our previous work [11], the present framework differs in that cyclic bubble nucleation, growth, detachment, and subsequent re-nucleation are explicitly incorporated.

2.3. LS formulation

The LS function ϕ_b evolves according to the following equation,

$$\frac{\partial \phi_b}{\partial t} + (\mathbf{u}_l + \frac{G_t \mathbf{n}_l}{\rho_l}) \cdot \nabla \phi_b = 0 \quad (30)$$

Table 1

Thermophysical properties of water vapor(v), hydrogen gas(h), and liquid water(l).

Physical properties	Unit	Vapor(v)	Hydrogen(h)	Liquid(l)
$\mu_{v/h/l}$	Pa s	1.9×10^{-5}	8.9×10^{-6}	1.0×10^{-3}
$\lambda_{v/h/l}$	W/(m K)	1.89×10^{-2}	1.87×10^{-1}	6.0×10^{-1}
ρ_l	kg/m ³	–	–	1.0×10^3
$c_{v/h/l}$	J/(kg K)	1.92×10^3	1.43×10^4	4.2×10^3
$M_{v/h/l}$	kg/mol	18×10^{-3}	2×10^{-3}	18×10^{-3}
D_l	m ² /s	–	–	4.5×10^{-9}

and is subsequently reinitialized using the following formulation:

$$\frac{\partial \phi_b}{\partial \tau^*} = S(\phi_b)(1 - |\nabla \phi_b|) \quad (31)$$

where

$$S(\phi_b) = 0 \quad \text{if } |\phi_b| \leq \frac{h}{2} \quad (32)$$

$$= \phi_b / \sqrt{\phi_b^2 + h^2} \quad \text{if } |\phi_b| > \frac{h}{2}$$

where $S(\phi_b)$ is a sign function and τ^* denotes a fictitious time step. It is noted that the reinitialization described in Eq. (31) is applied only to regions where $|\phi_b| > h/2$ in order to better preserve the conservative properties of the LS function [24,29].

3. Results and discussion

Simulations are carried out in an axisymmetric domain formulated in cylindrical coordinates, covering $0 \leq r \leq L_r = 1.8 \text{ mm}$ and $0 \leq y \leq L_y = 3.6 \text{ mm}$, as depicted in Fig. 1(a). Fine uniform meshes with grid sizes of $\Delta r = \Delta y = 12.5 \text{ }\mu\text{m}$ are employed in the near-bubble region ($0 \leq r \leq 1.2 \text{ mm}$, $0 \leq y \leq 1.65 \text{ mm}$), while non-uniform coarser grids are applied in the remaining regions to improve computational efficiency. The computational domain size ($L_r \times L_y = 1.8 \text{ mm} \times 3.6 \text{ mm}$) was selected to be sufficiently large to avoid boundary effects, and further enlargement of the domain was found to have a negligible influence on the predicted bubble growth behavior. Initially, a spherical electrolytic bubble with a radius of $R_{bo} = 50 \text{ }\mu\text{m}$ is placed at $(r, y) = (0, R_{bo} \cos \theta)$, submerged in quiescent liquid water at $p_\infty = 1 \text{ atm}$ and $T_\infty = T_w$. The initial bubble pressure is given by $p_{bo} = p_\infty + 2\sigma_{bl}/R_{bo}$, and the initial hydrogen mass fraction inside the bubble is given by $Y_{bo} = (p_{bo} - p_{vo})M_h / [p_{vo}M_v + (p_{bo} - p_{vo})M_h]$, where $p_{vo} = p_{v,sat}(T_\infty)$ as defined in Eq. (25). The present study adopts the thermophysical properties of the liquid, vapor, and hydrogen phases summarized in Table 1. In addition, parameters required for the phase-change formulation and interfacial dynamics are specified as $i_{lv} = 2.38 \times 10^6 \text{ J/(kg} \cdot \text{K)}$ and $\sigma_{bl} = 7.2 \times 10^{-2} \text{ kg/s}^2$. At the bottom boundary ($0 \leq r \leq L_r$, $y = 0$), an isothermal wall condition and a contact-angle boundary condition are imposed, with the wall temperature T_w and contact angle θ applied as follows:

$$u = v = 0, \quad \frac{\partial p}{\partial y} = 0 \quad (33)$$

$$T(y = 0) = T_w \quad (34)$$

$$\left. \frac{\partial \phi_b}{\partial y} \right|_{y=0} = \cos \theta \quad (35)$$

Hydrogen is generated at the active electrode surface ($r \leq R_e$), where the electrochemical generation rate is given by

$$-\rho_l D_l \left. \frac{\partial Y_l}{\partial y} \right|_{y=0} = \frac{M_h}{2F} i_w \quad \text{if } r \leq R_e \quad (36)$$

$$= 0 \quad \text{if } r > R_e$$

Here, F denotes Faraday's constant, 96485 C/kmol . Along the outer radial boundary ($r = L_r$, $0 \leq y \leq L_y$) and the centerline ($r = 0$, $0 \leq y \leq L_y$), symmetry boundary conditions are imposed as

$$u = 0, \quad \frac{\partial v}{\partial r} = 0, \quad \frac{\partial p}{\partial r} = 0 \quad \text{if } r = 0 \cup r = L_r \quad (37)$$

At the top boundary ($0 \leq r \leq L_r$, $y = L_y$), a fixed-pressure condition is applied:

$$\frac{\partial u}{\partial y} = 0, \quad \frac{\partial v}{\partial y} = 0, \quad p = p_\infty \quad (38)$$

3.1. Variable contact angle model

Based on experimental observations [41] on water electrolysis indicating that the contact angle θ decreases with increasing bubble radius R_b , we employ the following contact-angle model:

$$\theta = \min \left[\theta_o, \max \left(\theta_1, \theta_o - (\theta_o - \theta_1) \frac{R_b - R_{bo}}{R_{b1} - R_{bo}} \right) \right] \quad (39)$$

Here, θ_o denotes the initial contact angle for $R_b \leq R_{bo}$, whereas θ_1 denotes the later contact angle for $R_b \geq R_{b1}$. In the present study, $R_{bo} = 0.1 \text{ mm}$ and $R_{b1} = 0.25 \text{ mm}$ are adopted. This formulation enables a smooth transition between the initial and later contact-angle regimes. In the present study, the operating conditions are set to $R_e = 0.1 \text{ mm}$, $i_w = 38.5 \text{ A/cm}^2$, $T_w = 25^\circ\text{C}$, and $\theta_1 = 14^\circ$, which are consistent with the experimental conditions reported in Ref. [41]. It should be noted that both the present numerical results and the experimental measurements correspond to a converged periodic state obtained after multiple continuous bubble growth-departure cycles. In particular, as shown in Fig. 6(b), for the validation case of $R_e = 0.1 \text{ mm}$, the bubble growth behavior from the second cycle onward exhibits negligible differences compared with the first cycle, indicating rapid convergence to a periodic steady state. For a constant contact angle condition ($\theta_o = \theta_1$), the computed bubble growth history deviates from the experimental data, as shown in Fig. 2(a). When $\theta_o = \theta_1 = 14^\circ$, the bubble radius R_b increases more slowly than in the experiment during the early growth stage; however, the growth rate gradually approaches the experimental trend as time progresses. Owing to this discrepancy in the early growth rate, bubble departure occurs at a delayed time of $t = 361 \text{ ms}$. Despite the delayed departure, the bubble departure radius R_{bd} remains comparable to the experimental value. When $\theta_o = \theta_1 = 45^\circ$, R_b increases more rapidly in the early stage and initially agrees well with the experiment; however, the deviation progressively increases during the later growth period. These results indicate that the constant contact-angle condition cannot simultaneously reproduce both the early and late growth behaviors observed experimentally. Specifically, the case with $\theta_o = 14^\circ$ shows better agreement during the later stage, whereas the case with $\theta_o = 45^\circ$ more accurately captures the initial growth trend. This observation suggests that a variable contact angle within the range $14^\circ \leq \theta \leq 45^\circ$ is more appropriate for verification against the experimental data. For the variable contact angle case ($\theta_o = 45^\circ$, $\theta_1 = 14^\circ$), the temporal evolution of R_b closely matches the experimental data. These results demonstrate that the proposed cyclic formulation is capable of accurately reproducing experimentally observed periodic bubble growth behavior, including the bubble departure radius and cycle characteristics, under electrolysis conditions. The predicted bubble departure size R_{bd} is only 3.7% larger than that obtained under the constant contact angle condition of $\theta_o = \theta_1 = 14^\circ$. This consistent departure radius can be interpreted based on Fritz's correlation, $R_{bd}^o = 0.0104\theta \sqrt{\sigma_{bl} / [(\rho_l - \rho_b)g]}$, for which the theoretical prediction at $\theta = 14^\circ$ differs from the computed result by only 6.4%. The bubble interface shapes at $t = 285 \text{ ms}$, immediately prior to departure, are shown in Fig. 2(b). Under the variable contact-angle condition, the bubble begins to detach from the electrode surface, whereas in the two constant-angle cases the bubble has not yet departed due to either the slower growth rate or the excessively large R_{bd} . In the present study, all subsequent simulations are performed using the variable contact angle formulation in Eq. (39). It should be noted that this formulation offers greater flexibility for electrolysis applications compared with that adopted in our previous study [11], where the contact angle was prescribed as a more complex function of R_b . To further highlight the role of microlayer evaporation in the present model, the microlayer evaporation rate $\dot{m}_{bm}$

is compared with the total evaporation rate $\dot{m}_b$ in Fig. 2(c). Here, $\dot{m}_b$ and $\dot{m}_{bm}$ denote the evaporation rates integrated over the entire bubble surface and over the microlayer region, respectively. During the early stage of bubble growth, when both the bubble size and contact angle change rapidly, $\dot{m}_{bm}$ accounts for more than 50% of $\dot{m}_b$. This behavior arises because the microlayer occupies a relatively large fraction of the total bubble surface area during this stage, leading to a dominant contribution from microlayer evaporation. Even after the contact angle stabilizes at $t = 67$ ms, $\dot{m}_{bm}$ still contributes approximately 20 ~ 30% of $\dot{m}_b$ and maintains a relatively steady trend. Near the bubble departure stage ($t \approx 280$ ms), however, $\dot{m}_{bm}$ decreases sharply due to the reduction in microlayer area as the bubble lifts off from the surface. These results demonstrate that microlayer evaporation contributes substantially to the overall phase-change mass transfer throughout most of the bubble growth process and therefore represents an essential component of the present numerical framework for electrolysis simulations.

To verify the convergence characteristics of the present numerical formulation, additional simulations were performed under the same operating conditions as those used in Fig. 2 with different spatial and temporal resolutions, as shown in Fig. 3. The spatial resolutions considered were $\Delta r = 1/40$ mm, $1/80$ mm, and $1/120$ mm, while the temporal resolutions were $\Delta t = 1$ μ s and 0.5 μ s. As the grid resolution is refined, the predicted bubble radius R_b gradually converges, as illustrated in Fig. 3(a). While the finer grid cases ($\Delta r = 1/80$ mm and $1/120$ mm) show only negligible differences, the coarser grid case ($\Delta r = 1/40$ mm) exhibits noticeable deviations. This tendency is further confirmed by the interface shapes at $t = 280$ ms, shown in Fig. 3(b), where the finer grid cases produce nearly identical interface profiles and similar bubble departure behavior. The temporal resolution test also indicates that the results obtained with $\Delta t = 1$ μ s closely agree with those obtained using $\Delta t = 0.5$ μ s, as shown in Figs. 3(c) and 3(d). Based on these observations, $\Delta r = 1/80$ mm and $\Delta t = 1$ μ s were selected as the reference resolutions, providing a good balance between computational accuracy and efficiency. Overall, the results demonstrate that the present numerical formulation achieves sufficient spatial and temporal convergence and provides reliable predictions of hydrogen bubble growth during electrolysis.

3.2. Cyclic simulation of bubble growth

Fig. 4 illustrates cyclic simulation of hydrogen bubble growth, departure, and re-nucleation at $R_e = 0.1$ mm, $i_w = 38.5$ A/cm², $T_w = 20^\circ$ C, and $\theta_1 = 14^\circ$. In the early stage, bubble growth on the electrode surface is observed at $t = 2$ ms. Continued growth is sustained by the consumption of dissolved hydrogen and the displacement of the surrounding liquid, while the bubble maintains a nearly spherical shape. As hydrogen is continuously generated at the electrode surface and supplied to both the electrolyte and the bubble, a slight accumulation of dissolved hydrogen develops near the electrode periphery, as observed at $t = 160$ ms, indicating that the local generation rate exceeds the mass transfer across the bubble interface. This accumulated hydrogen remains localized near the outer edge of the electrode for 160 ms $\leq t \leq 280$ ms, consistent with previous simulations [10] showing confined gas distribution near the electrode periphery. As the bubble enlarges, buoyancy gradually overcomes surface tension, causing the contact area with the electrode to shrink, as observed at $t = 288$ ms. After departure, a dissolved hydrogen region still remains near the electrode surface and is redistributed by the upward motion of the bubble and the induced electrolyte convection, forming a residual concentration distribution near the electrode along the central y -axis, as shown at $t = 294$ ms. To model bubble re-nucleation after sufficient dissolved-hydrogen accumulation, a small bubble acting as the nucleus for the subsequent cycle is placed on the electrode at $t = 295$ ms. Because the waiting period τ_w preceding re-nucleation at a given site cannot be uniquely determined, the waiting period was chosen as $\tau_w = 5$ ms, which is short but still long enough to prevent interference from the

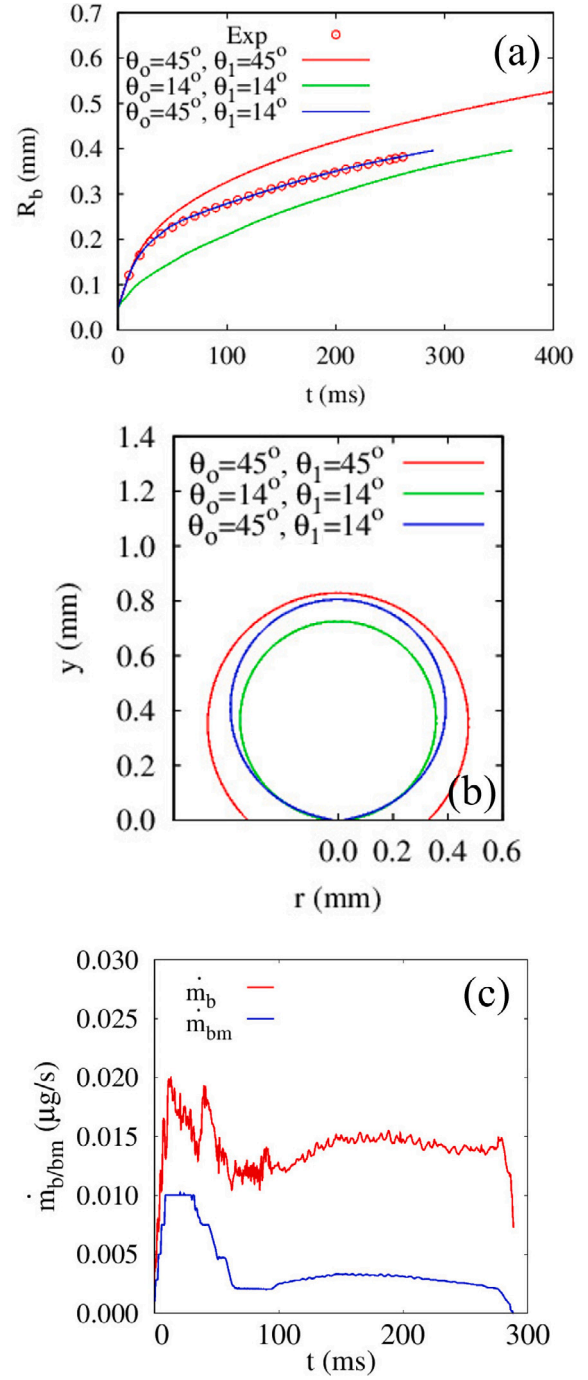


Fig. 2. Numerical results at $R_e = 0.1$ mm, $i_w = 38.5$ A/cm², and $T_w = 25^\circ$ C for different contact angles: (a) temporal evolution of the averaged bubble radius R_b , (b) bubble interface shape at $t = 285$ ms, and (c) the total evaporation rate $\dot{m}_b$ and the microlayer evaporation rate $\dot{m}_{bm}$ at $\theta_o = 40^\circ$ and $\theta_i = 15^\circ$.

preceding bubble. The second bubble grows over a period of $\tau_i = 287$ ms and reaches departure at $t = 582$ ms, which is nearly identical to the total growth period ($\tau_i = 288$ ms) observed in the first cycle. This suggests that a sufficiently developed dissolved hydrogen boundary layer after bubble departure is required to influence re-nucleation and subsequent growth dynamics; however, under the present small electrode condition ($R_e = 0.1$ mm), such an effect remains limited.

When the electrode radius increases to $R_e = 0.2$ mm, the corresponding liquid velocity and dissolved hydrogen concentration fields during cyclic bubble growth are shown in Fig. 5. Similar to the case

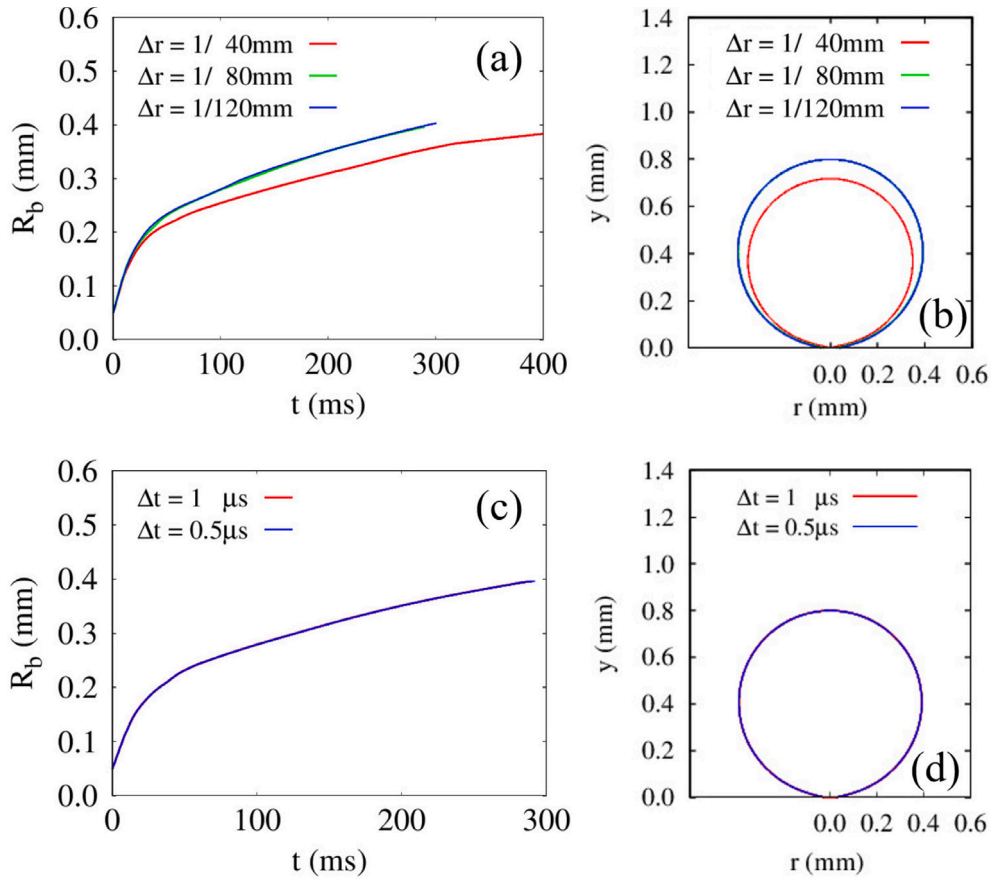


Fig. 3. Resolution tests for grid size (a–b) and time step (c–d) at $R_e = 0.1$ mm, $i_w = 38.5$ A/cm², $T_w = 25$ °C, and $\theta_1 = 14$ °: (a,c) averaged bubble radius, R_b ; (b,d) bubble interface at $t = 280$ ms.

shown in Fig. 4, the bubble initially grows within a dilute dissolved hydrogen field, as illustrated at $t = 2$ ms. However, a noticeable accumulation of dissolved hydrogen develops near the electrode surface at an earlier stage ($t \approx 30$ ms). As the bubble continues to grow, the dissolved hydrogen boundary layer progressively develops, and its thickness becomes more pronounced near the outer edge of the electrode. This indicates that hydrogen generated near the central region of the electrode is predominantly transported into the bubble, whereas a significant portion of hydrogen produced near the electrode periphery diffuses into the surrounding electrolyte. The bubble maintains a nearly spherical shape and grows until it departs at $t = 179$ ms. Bubble departure and the subsequent upward motion induce convective flow in the surrounding liquid, leading to the formation of a narrowly confined region of high hydrogen concentration along the central y -axis, as shown at $t = 185$ ms. Compared with the smaller electrode case in Fig. 4, the residual dissolved hydrogen boundary layer after departure is noticeably thicker and sufficiently developed to influence subsequent bubble nucleation and growth. Notably, the dissolved hydrogen distribution after bubble departure differs markedly from the initial distribution. After the bubble separates from the cathode, nucleation at the same site is observed at $t = 186$ ms within a denser concentration boundary layer than that in the first cycle shown in Fig. 4. This elevated local hydrogen concentration results from the residual hydrogen and its relaxation prior to re-nucleation, leading to faster bubble growth and a shorter growth period. At $t = 260$ ms, the bubble grown for 74 ms reaches a size comparable to that at $t = 100$ ms in Fig. 4, indicating a higher growth rate than in the first cycle. The bubble then grows over a period of $\tau_i = 149$ ms and departs at $t = 335$ ms, corresponding to a 16.7% reduction in the growth period compared with the first cycle. These results demonstrate that cyclic simulations incorporating

bubble departure and re-nucleation become physically meaningful in electrolysis systems with electrodes of $R_e \geq 0.2$ mm, where the residual dissolved hydrogen boundary layer attains sufficient thickness to alter subsequent growth dynamics.

The shortened growth period τ_i during the second cycle reduces additional hydrogen accumulation near the electrode during bubble growth, thereby moderating the growth rate in subsequent cycles. This counterbalancing effect between the growth period and residual hydrogen availability leads to a gradual convergence toward a repeatable profile. To ensure that the dissolved hydrogen concentration field at nucleation closely resembles the post-departure distribution, the simulation must be repeated over multiple cycles until the dissolved hydrogen boundary layer distribution reaches a quasi-periodic converged state. Such cyclic computation is essential for accurately resolving electrolytic bubble dynamics, although it has rarely been incorporated in previous numerical studies.

The cyclic bubble growth for $R_e = 0.1$ mm and 0.2 mm at $i_w = 38.5$ A/cm², $T_w = 20$ °C, and $\theta_1 = 14$ ° is summarized in Fig. 6. The bubble radius R_b is plotted only up to bubble re-nucleation. For $R_e = 0.1$ mm, the temporal evolution of R_b exhibits nearly identical patterns over successive cycles, as shown in Fig. 6(a). A direct comparison of R_b from the nucleation time t_{bo} to just before departure is provided in Fig. 6(b), where the 1st to 4th growth cycles almost overlap. This indicates that cyclic effects exert negligible influence on bubble growth dynamics under the small-electrode condition of $R_e = 0.1$ mm. In contrast, for $R_e = 0.2$ mm, the slope of the R_b evolution gradually changes with increasing cycle number, as observed in Fig. 6(c). The total growth period τ_i is shorter than that for $R_e = 0.1$ mm, owing to the increased hydrogen generation rate from the larger active electrode

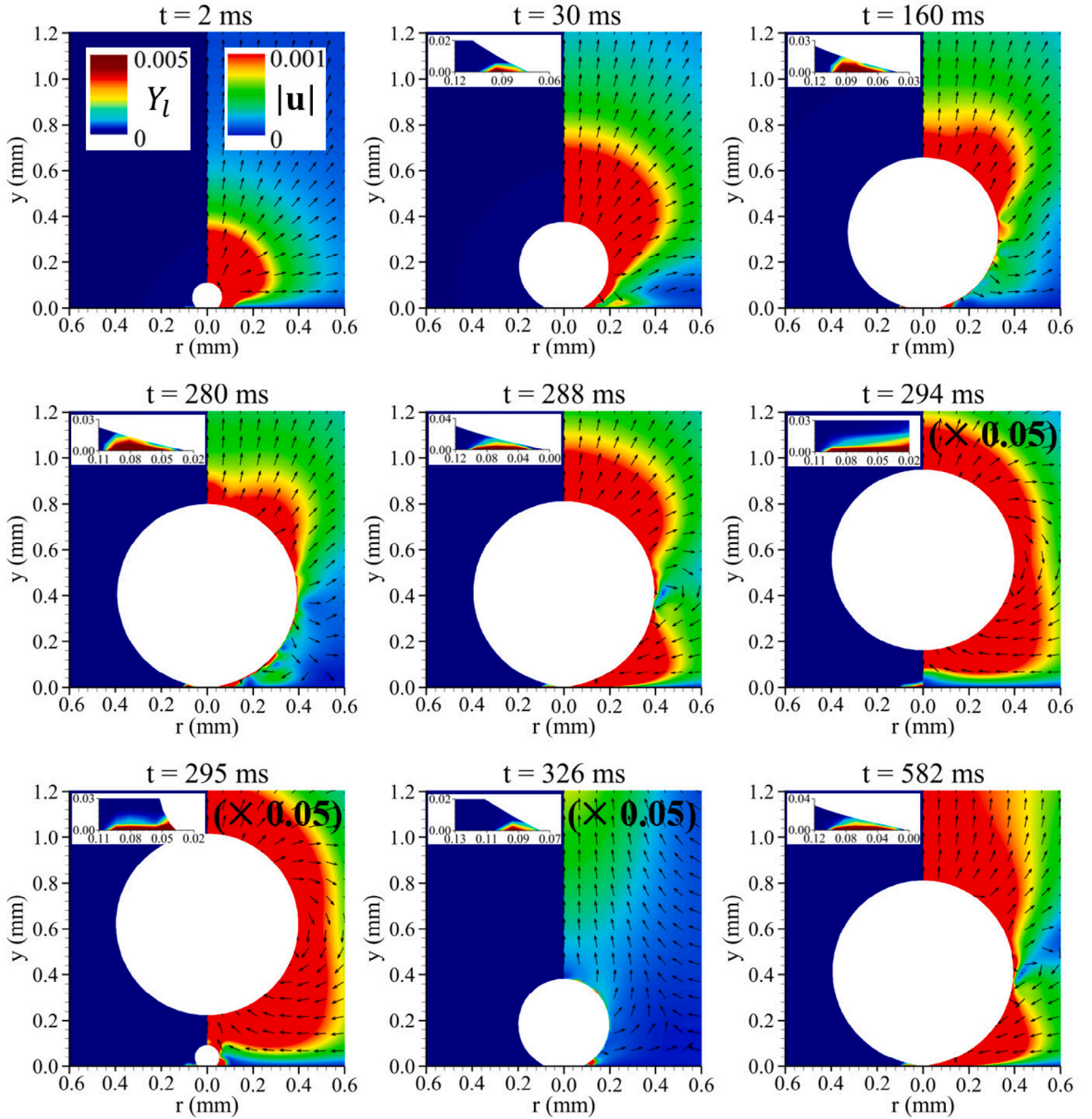


Fig. 4. Dissolved hydrogen concentration field (left) and liquid velocity field (right) around the growing bubble interface at $R_e = 0.1$ mm, $i_w = 38.5$ A/cm², $T_w = 20^\circ\text{C}$, and $\theta_1 = 14^\circ$. For clarity, the velocity vectors shown for $294\text{ ms} \leq t \leq 326\text{ ms}$ are scaled by a factor of 0.05. The insets in the upper-left corner provide enlarged views of the dissolved hydrogen concentration field near the bottom region of the bubble. The white region represents the bubble phase.

area, with τ_i remaining below 0.2 s. A direct comparison of R_b during the 1st to 5th is shown in Fig. 6(d). In the 1st cycle, R_b increases at a relatively slow rate, leading to the most delayed departure at $t - t_{bo} = 179$ ms. As high-concentration hydrogen accumulates near the electrode during the prolonged τ_i , R_b increases more rapidly in the 2nd cycle. The temporal evolution of R_b becomes progressively steeper over the subsequent cycles. To evaluate the number of cycles required for convergence, the bubble departure time after nucleation is adopted as the convergence criterion. Convergence is considered to be achieved when the relative difference in the departure time between two successive cycles becomes less than 1%. Based on this criterion,

convergence is generally attained from approximately the fourth cycle onward. This behavior is consistent with the trend observed in Fig. 6(d), where the growth rate of R_b also converges after approximately four cycles. These results further indicate that the dissolved hydrogen concentration fields in the post-departure state become nearly identical between successive cycles. This behavior qualitatively resembles the convergent trend toward a repeatable growth curve reported in the previous numerical study [22]. The observed convergence results from the counterbalancing effect between accumulated residual hydrogen and the variation in the overall growth period τ_i . These results suggest that, under the present simulation conditions, approximately 4 to 5

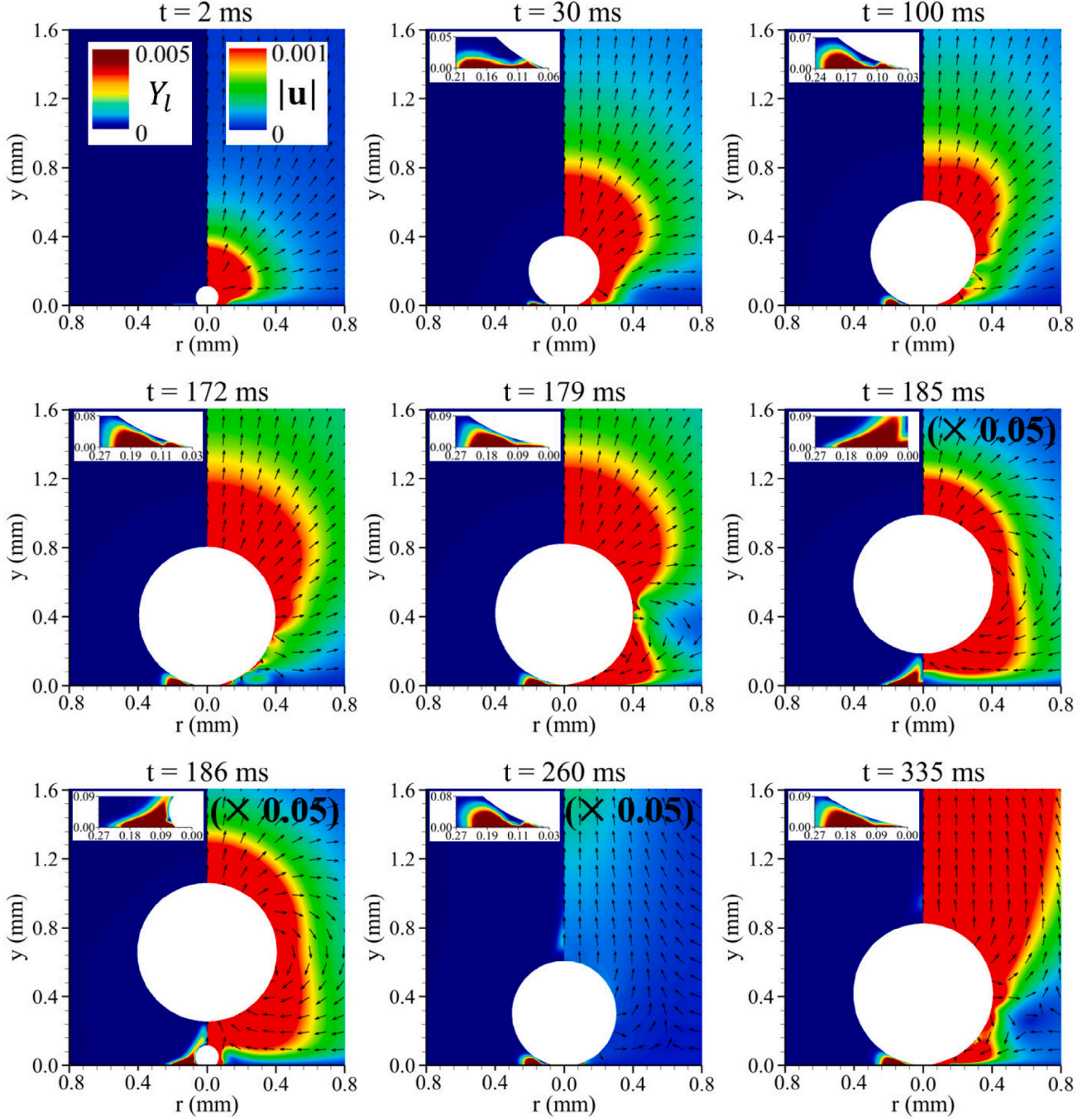


Fig. 5. Dissolved hydrogen concentration field (left) and liquid velocity field (right) around the growing bubble interface at $R_e = 0.2$ mm, $i_w = 38.5$ A/cm², $T_w = 20$ °C, and $\theta_1 = 14$ °. For clarity, the velocity vectors shown for 294 ms $\leq t \leq 326$ ms are scaled by a factor of 0.05. The insets in the upper-left corner provide enlarged views of the dissolved hydrogen concentration field near the bottom region of the bubble. The white region represents the bubble phase.

successive cycles are required for the bubble growth to stabilize into a quasi-periodic pattern, which serves as a necessary condition for reliably evaluating hydrogen generation performance and hydrogen evolution efficiency.

3.3. Effects of electrode size and current density

Fig. 7 illustrates the effects of the electrode radius R_e on the evolution of the averaged bubble radius R_b and the associated hydrogen production characteristics at $i_w = 38.5$ A/cm², $T_w = 20$ °C, and $\theta_1 = 14$ °. Here, the hydrogen bubble production rate $\dot{m}_{bh}$ and the hydrogen

evolution efficiency η are defined as

$$\dot{m}_{bh} = \frac{m_{bd} Y_{bd}}{\tau_t} \quad (40)$$

$$\eta = \frac{m_{bd} Y_{bd}}{(M_h/2F) i_w \pi R_e^2 \tau_t} \quad (41)$$

where $m_{bd} = \rho_{bd} \pi (4/3) R_{bd}^3$ and Y_{bd} denote the total bubble mass at departure and the hydrogen mass fraction inside the bubble, respectively. It should be noted that the hydrogen evolution efficiency is defined as the ratio of the actual hydrogen production rate to the theoretical hydrogen generation rate predicted by Faraday's law. Therefore, it primarily reflects mass transfer limitations and dissolved hydrogen

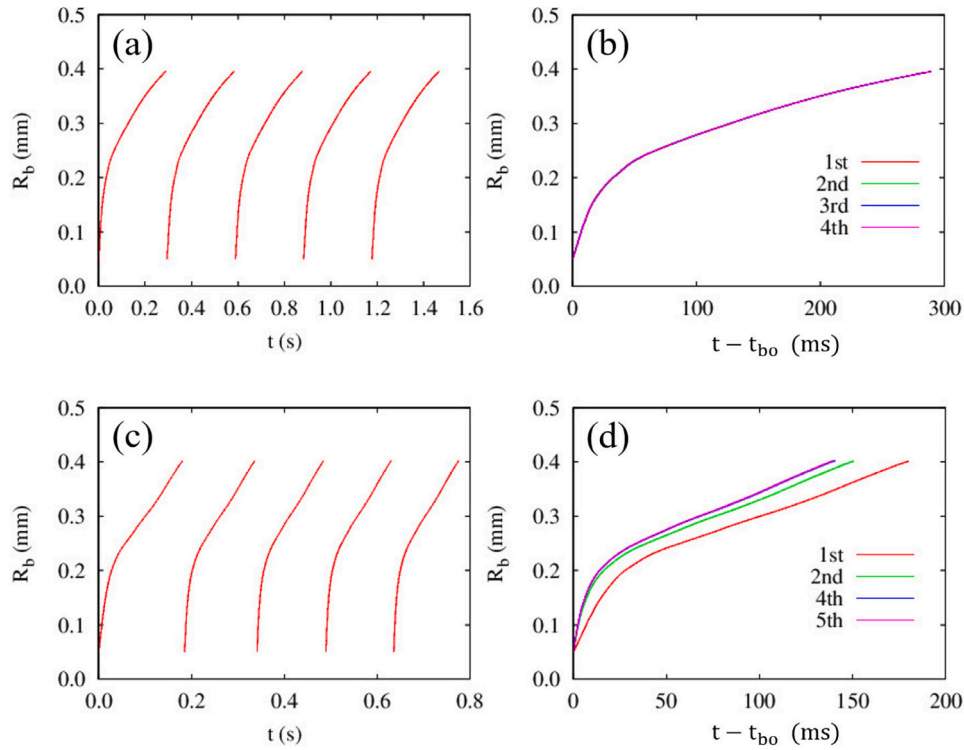


Fig. 6. Temporal evolution of bubble radius R_b over multiple cycles at $i_w = 38.5 \text{ A/cm}^2$, $T_w = 20^\circ\text{C}$, and $\theta_1 = 14^\circ$ for different electrode radii: (a), (b) $R_e = 0.1 \text{ mm}$ and (c), (d) $R_e = 0.2 \text{ mm}$. Panels (b) and (d) present the results plotted with respect to $t - t_{bo}$, where t_{bo} denotes the onset time of bubble nucleation.

losses into the electrolyte, rather than the overall thermodynamic or electrochemical energy efficiency of the system. As R_e increases from $R_e = 0.1 \text{ mm}$ to 0.5 mm , hydrogen generation near the catalytic interface is enhanced, thereby accelerating bubble growth, as depicted in Fig. 7(a). Here, the profile of R_b corresponds to the converged state obtained from cyclic simulations and is plotted from the nucleation time t_{bo} to the departure instant. For $R_e = 0.5 \text{ mm}$, the bubble detaches at $\tau_t = 45.2 \text{ ms}$, which corresponds to approximately 15.4% of the growth period observed for $R_e = 0.1 \text{ mm}$. The rapid bubble growth at $R_e = 0.5 \text{ mm}$ leads to a slightly larger departure radius R_{bd} approximately $25 \mu\text{m}$ greater than that at $R_e = 0.1 \text{ mm}$, owing to enhanced inertial effects in the surrounding liquid, as also evident in Fig. 7(b). In contrast, during the initial cycle, R_{bd} exhibits slightly smaller values for all R_e , which is attributed to the slower growth rate and the consequently weaker inertial effect in the first cycle. This tendency is also confirmed by the direct comparison of R_b among cycles in Fig. 6(d), where the first cycle shows the most gradual growth profile. For $R_e \geq 0.2 \text{ mm}$, R_{bd} and the corresponding τ_t for the initial cycle approach certain asymptotic values, whereas the cyclic simulation results for τ_t decrease markedly with increasing R_e , as shown in Fig. 7(c). This reduction in τ_t is attributed to the enhanced residual dissolved H_2 concentration after departure associated with the larger active electrode area, thereby increasing the hydrogen bubble production rate $\dot{m}_{bh}$ as plotted in Fig. 7(d). Note that, because τ_t appears in the denominator of Eq. (40), $\dot{m}_{bh}$ is primarily governed by the frequency of bubble departure when the H_2 mass inside the bubble at detachment remains comparable. The comparable H_2 mass in the departing bubble is demonstrated by the nearly identical $\dot{m}_{bh}$ for $R_e \geq 0.2 \text{ mm}$ observed during the initial cycle, consistent with the nearly constant values of τ_t and R_{bd} . In contrast to the enhanced hydrogen bubble production rate, the hydrogen evolution efficiency η decreases with increasing R_e , as shown in Fig. 7(e), due to the greater tendency of dissolved H_2 to diffuse away from the reaction zone over the enlarged active surface area. During the initial cycle, η is generally lower than that obtained from cyclic simulations, since although the instantaneous H_2 loss rate from the reaction zone may be

smaller, the prolonged growth period τ_t results in a larger cumulative amount of H_2 loss. This implies that evaluating η based solely on the initial cycle may significantly underestimate the performance compared with cyclic simulations that better represent realistic electrolysis conditions. These results underscore the importance of optimizing electrode size in electrolyzer design, particularly when hydrogen production rate and hydrogen evolution efficiency must be considered simultaneously. It should be noted that the present definition of the hydrogen evolution efficiency (η) accounts only for transport-related effects associated with bubble dynamics and mass transfer. Temperature-dependent electrochemical kinetics, overpotential, and catalytic activity are not included in the present model.

The effect of current density i_w on the evolution of R_b and overall system performance is presented in Fig. 8, with all other parameters fixed at $R_e = 0.2 \text{ mm}$, $T_w = 20^\circ\text{C}$, and $\theta_1 = 14^\circ$. As i_w increases from 28.9 A/cm^2 to 48.1 A/cm^2 , R_b increases at a progressively higher rate, as shown in Fig. 8(a). The enhanced growth rate with increasing i_w and the corresponding increase in liquid inertia around the bubble lead to a slight increase in the departure radius R_{bd} ; however, the increase remains modest, as shown in Fig. 8(b), due to the relatively similar slopes of R_b during the later growth stage. While R_{bd} remains nearly unchanged with increasing i_w , a substantial reduction in τ_t is observed in Fig. 8(c), indicating more frequent bubble growth-departure cycles at elevated i_w . This tendency is consistent with previous computational results [22], in which electrolytic bubble evolution and detachment were analyzed under different current densities using a moving-mesh framework. The shortening of τ_t plays a dominant role in enhancing $\dot{m}_{bh}$, as shown in Fig. 8(d). As i_w increases from 28.9 A/cm^2 to 38.5 A/cm^2 and 48.9 A/cm^2 , τ_t decreases by factors of $1/1.23$ and $1/1.44$, respectively, while $\dot{m}_{bh}$ correspondingly increases by factors of 1.23 and 1.46 . Considering Eq. (40), in which τ_t appears in the denominator and $\dot{m}_{bd}Y_{bd}$ in the numerator, these proportionalities indicate that the hydrogen mass inside the departing bubble remains nearly unchanged with varying i_w . In contrast, η gradually decreases as i_w increases, indicating that higher local dissolved H_2 concentrations promote diffusion

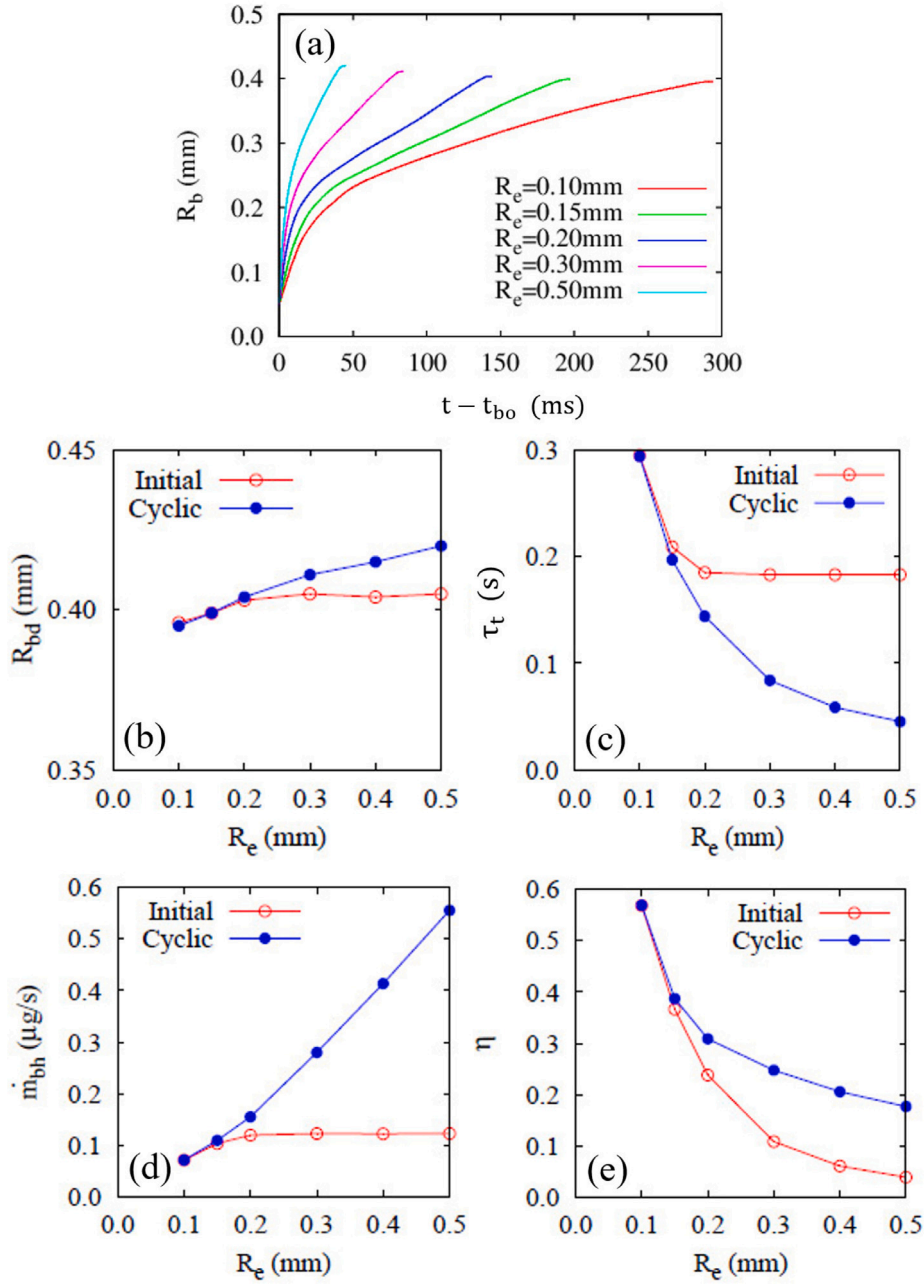


Fig. 7. Effect of electrode radius R_e on the numerical results at $i_w = 38.5 \text{ A/cm}^2$, $T_w = 20^\circ\text{C}$, and $\theta_1 = 14^\circ$: (a) temporal variation of the bubble radius R_b over a cyclic period; (b) bubble departure radius R_{bd} ; (c) total growth time τ_t ; (d) hydrogen bubble production rate $\dot{m}_{bh}$; and (e) hydrogen evolution efficiency η for the initial and cyclic periods.

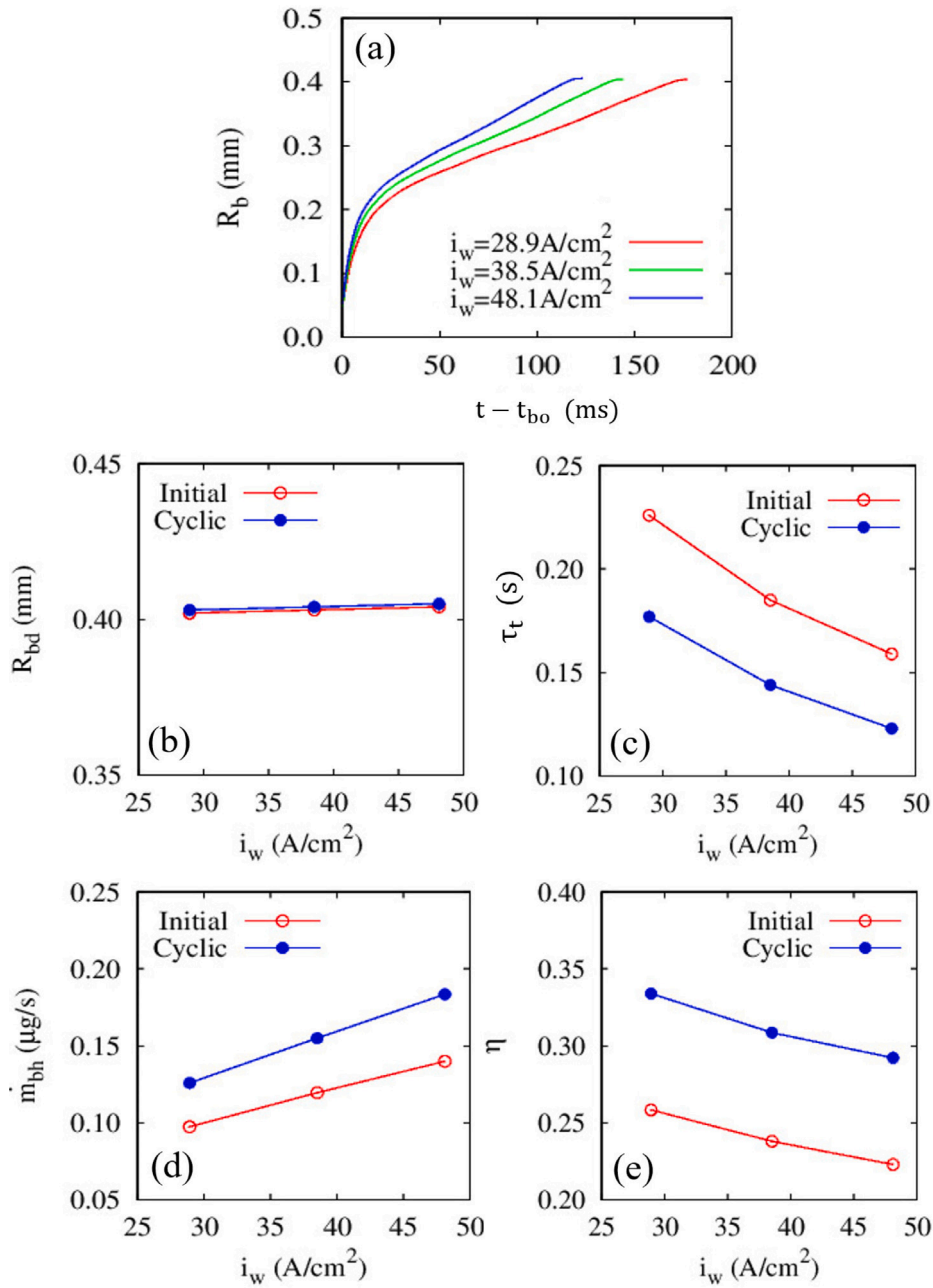


Fig. 8. Effect of current density i_w on the numerical results at $R_e = 0.2 \text{ mm}$, $T_w = 20^\circ\text{C}$, and $\theta_1 = 14^\circ$: (a) temporal variation of the bubble radius R_b over a cyclic period; (b) bubble departure radius R_{bd} ; (c) total growth time τ_t ; (d) hydrogen bubble production rate $\dot{m}_{bh}$; and (e) hydrogen evolution efficiency η for the initial and cyclic periods.

losses into the bulk liquid. Compared with the variation of η with R_e in Fig. 7(e), the sensitivity of η to changes in i_w is relatively weaker, because in Eq. (41) the product $\tau_t i_w$ appears in the denominator, and the increase in i_w is partially offset by the decrease in τ_t . This tradeoff between hydrogen production rate and hydrogen evolution efficiency underscores the importance of optimizing current density to achieve balanced performance in electrolysis system design.

Fig. 9 presents the effect of electrode radius on R_b and electrolysis performance under a constant applied energy condition of $R_e^2 i_w = 38.5 \text{ mA}$, with $T_w = 20^\circ\text{C}$ and $\theta_1 = 14^\circ$ fixed. As R_e increases from 0.1 mm to 0.15 mm, the growth rate of R_b gradually decreases, as shown in Fig. 9(a), exhibiting a trend opposite to that observed in Fig. 7(a) under constant current density. This behavior arises because the

enlarged active electrode area results in a reduced local current density, causing H_2 generation to be spatially distributed; although the total H_2 production remains constant, a larger fraction diffuses into the bulk electrolyte rather than being transported directly into the bubble. The decrease in the growth rate of R_b becomes less pronounced between $R_e = 0.15 \text{ mm}$ and 0.2 mm . This is attributed to residual dissolved hydrogen that is not immediately captured by the bubble because the gas generation sites are farther from the nucleation region in larger electrodes, and is subsequently redistributed toward the nucleation region by departure-induced convection. During the later growth stage, the slope of R_b becomes nearly identical across cases, particularly between $R_e = 0.15 \text{ mm}$ and 0.2 mm , resulting in an almost unchanged R_{bd} with respect to R_e , whereas τ_t exhibits similar values only for

$R_e = 0.15$ mm and 0.2 mm, as depicted in Figs. 9(b) and 9(c). When Y_{bd} remains nearly constant and R_{bd} varies little with R_e , the hydrogen mass inside the departing bubble becomes comparable across cases, and τ_i consequently governs $\dot{m}_{bh}$, as illustrated in Fig. 9(d). It should be noted that because $R_e^2 i_w$ is fixed in Eq. (41), η is primarily governed by τ_i under the present condition, as shown in Fig. 9(e). These results highlight that increasing R_e under constant total current reduces both $\dot{m}_{bh}$ and η , with a pronounced decline observed at $R_e = 0.15$ mm. Note that the contact radius at departure is approximately $R_{bd} \sin(14^\circ) \approx 0.1$ mm; once R_e exceeds this characteristic length scale, a portion of the generated hydrogen can escape the effective bubble capture region. For $R_e \geq 0.15$ mm, the decreasing trend becomes more gradual, indicating that part of the diffused hydrogen is subsequently re-entrained into the bubble growth process through cyclic accumulation and convection. These findings indicate that the departure contact radius may act as an effective threshold length scale beyond which diffusion losses become significant.

3.4. Effect of wall temperature

Fig. 10 illustrates the influence of wall temperature T_w on electrolysis performance under the conditions of $R_e = 0.2$ mm, $i_w = 38.5$ A/cm², and $\theta_1 = 14^\circ$. As T_w increases from 20°C to 60°C , bubble growth becomes more pronounced and detachment occurs after a shorter growth period, as shown in Fig. 10(a). This accelerated growth is mainly attributed to the enhanced liquid–vapor phase-change rate into the bubble at elevated temperatures. The increased vapor contribution at higher T_w reduces the hydrogen mass fraction Y_b , as shown in Fig. 10(b). Although vapor continuously enters the bubble throughout the growth process, the temporal variation of Y_b remains nearly constant, indicating that vapor and hydrogen gas inflow are thermodynamically balanced. The value of Y_b at detachment is measured as $Y_{bd} = 0.810$, 0.542 , and 0.256 for $T_w = 20^\circ\text{C}$, 40°C , and 60°C , respectively. These values, predominantly governed by T_w closely follow the theoretical equilibrium estimate $Y_{be}(R_{bd}, T_w) = (p_b - p_{v,sat}) M_h / [(p_b - p_{v,sat}) M_h + p_{v,sat} M_v] \approx Y_{be}(T_w)$, where $p_b(R_{bd}) = 2\sigma/R_{bd} + p_\infty \approx p_\infty$. The increase in vapor content enhances both the bubble growth rate and the internal bubble mass, leading to a larger R_{bd} and a shorter τ_i at $T_w = 60^\circ\text{C}$, as observed in Figs. 10(c) and 10(d). In contrast to previous cases, however, the reduction in τ_i has a limited impact on $\dot{m}_{bh}$ and η , as shown in Figs. 10(e) and 10(f). According to Eqs. (40) and (41), the numerator terms $m_{bd} = \rho_{bd} \pi (4/3) R_{bd}^3$ and Y_{bd} exhibit strong temperature dependence. For $T_w = 60^\circ\text{C}$, $\tau_i = 107$ ms, corresponding to 0.743 times that in the $T_w = 20^\circ\text{C}$ case, whereas m_{bd} and Y_{bd} increase to 2.18 and 0.316 times those in the $T_w = 20^\circ\text{C}$ case, respectively, thereby offsetting much of the reduction in τ_i and resulting in only a marginal decrease ($\approx 7.3\%$) in $\dot{m}_{bh}$ and η . In addition, the initial-cycle results underestimate the system performance parameters ($\dot{m}_{bh}$ and η) compared with those obtained from the cyclic simulations. These results indicate that the apparent efficiency trends with respect to wall temperature depend on whether cyclic behavior is considered in the analysis. It is noted that although elevated wall temperatures promote faster bubble growth, they do not necessarily enhance hydrogen bubble generation or improve hydrogen evolution efficiency. It should also be emphasized that the temperature effects discussed in this study are primarily associated with enhanced phase change and interfacial transport phenomena, whereas temperature-dependent electrochemical kinetics and overpotential effects are beyond the scope of the current formulation.

3.5. Effect of contact angle

Fig. 11 presents the liquid velocity and pressure fields around the bubble during the cyclic simulation for an increased contact angle of $\theta_1 = 20^\circ$, while maintaining $R_e = 0.2$ mm, $i_w = 38.5$ A/cm², and $T_w = 20^\circ\text{C}$. In the 1st cycle, the bubble initially grows with an

approximately spherical shape and generates a radially outward flow field in the surrounding liquid, as observed at $t = 60$ ms. The larger contact angle promotes lateral spreading of the bubble base, leading to an expanded coverage area and a wider microlayer region with reduced mass-transfer resistance, together with a locally concentrated distribution of dissolved H_2 near the electrode edge, as shown at $t = 150$ ms. Compared with the case of $\theta_1 = 14^\circ$ in Fig. 5, the bubble grows to a larger size before detachment, as seen at $t = 275$ ms. Detachment begins at $t = 288$ ms, during which significant interfacial motion along the electrode surface and the upward bubble movement induce a strong convective flow. The detachment size is substantially larger than that for $\theta_1 = 14^\circ$, consistent with Fritz's formulation, which indicates that the contact angle strongly governs the detachment size. After detachment, the residual dissolved H_2 remaining at the electrode surface is more weakly distributed than in the $\theta_1 = 14^\circ$ case and accumulates near the electrode center, as observed at $t = 294$ ms. This distribution results from convective transport induced by the preceding detached bubble, consistent with previous numerical study [15] on electrolytic bubble dynamics. A new bubble nucleates within this relatively depleted H_2 concentration boundary layer at $t = 295$ ms and begins to grow. The expanded coverage area and the subsequent dilute concentration field lead to a slower initial growth rate during the early stage of the following cycle. After a growth period of $\tau_i = 265$ ms, the bubble detaches again at $t = 570$ ms. The second growth period is approximately 8.0% shorter than the first, which is smaller than the 16.9% reduction observed between the first and second cycles for $\theta_1 = 14^\circ$. These results indicate that the contact angle critically controls both the overall growth period and the detachment radius, thereby exerting a strong influence on electrolysis performance.

When the contact angle further increases to $\theta_1 = 30^\circ$, the numerical results are presented in Fig. 12. As the bubble grows and the contact line between the bubble interface and the electrode surface recedes, the microlayer area gradually expands. The enlarged microlayer area accelerates bubble growth and induces a stronger radially outward liquid flow, as observed at $t = 30$ ms. The bubble eventually covers the entire electrode surface, as seen at $t = 87$ ms, and the hydrogen generated at the electrode is directly injected into the bubble through the microlayer, where mass-transfer resistance is minimal. Owing to the larger contact angle and the corresponding increase in surface tension force, the bubble remains attached to the electrode surface even at $t = 440$ ms and detaches at a significantly delayed time of $t = 492$ ms. Compared with the cases of $\theta_1 = 14^\circ$ and 20° , almost no residual dissolved H_2 is observed near the electrode surface after detachment, as shown at $t = 495$ ms. This behavior results from the fact that, during most of the growth period, the electrode active surface is covered by the bubble, and the generated hydrogen is transferred directly into the bubble without diffusing into the bulk electrolyte. The subsequently nucleated bubble grows under conditions nearly identical to the initial state and detaches again at $t = 987$ ms after a growth period $\tau_i = 491$ ms, which differs by less than 1% from that of the 1st cycle. These results indicate that, for microscale electrodes with large contact angles, the influence of residual hydrogen on subsequent bubble growth becomes negligible, and the deviation between initial and cyclic bubble-growth dynamics diminishes. It should be noted that, within the current microlayer model, surface coverage by the growing bubble does not necessarily hinder hydrogen evolution efficiency, as hydrogen mass transfer remains active beneath the bubble throughout its evolution.

The effect of θ_1 on the evolution of R_b and the electrolysis performance is presented in Fig. 13. For $\theta_1 = 14^\circ$, the initial growth stage exhibits the steepest slope of R_b , as shown in Fig. 13(a), due to the elevated residual H_2 concentration accumulated near the electrode surface, which accelerates early bubble growth. As growth proceeds, the rate of increase in R_b gradually decreases, and cases with $\theta_1 > 14^\circ$ display steeper growth during the intermediate stage owing to the wider microlayer region and enhanced mass transfer therein. Increasing

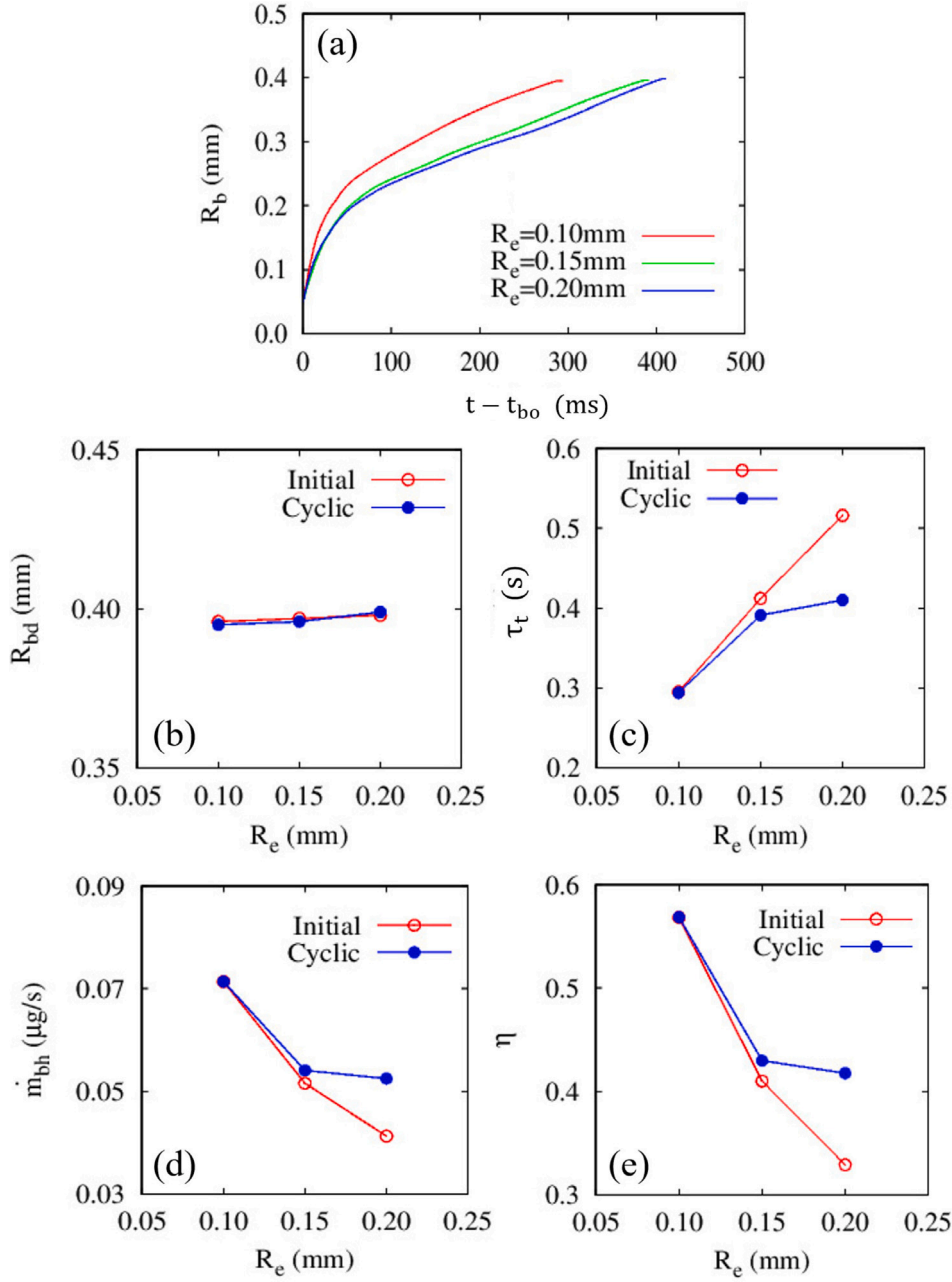


Fig. 9. Effect of electrode radius R_e on the numerical results at $R_{e,iw}^2 = 38.5 \text{ mA}$, $T_w = 20^\circ\text{C}$, and $\theta_1 = 14^\circ$: (a) temporal variation of the bubble radius R_b over a cyclic period; (b) bubble departure radius R_{bd} ; (c) total growth time τ_t ; (d) hydrogen bubble production rate $\dot{m}_{bh}$; and (e) hydrogen evolution efficiency η for the initial and cyclic periods.

θ_1 requires a larger buoyancy force for bubble detachment, resulting in both a larger R_{bd} and a longer τ_t , as shown in Figs. 13(b) and 13(c). As θ_1 increases from 14° to 30° , R_{bd} follows an approximately linear trend with respect to θ_1 , consistent with the theoretical prediction from Fritz's correlation. A monotonic increase in τ_t is also observed, indicating that larger contact angles induce slower bubble removal cycles during electrolysis; moreover, the discrepancy in τ_t between the initial and cyclic simulations is more pronounced at smaller θ_1 . Based on the computed values of R_{bd} and τ_t , $\dot{m}_{bh}$ and η were evaluated and are shown in Figs. 13(d) and 13(e). At $\theta_1 = 30^\circ$, the electrolysis performance reaches its maximum, with $\dot{m}_{bh} = 0.46 \mu\text{g/s}$ and $\eta = 0.92$. In Eqs. (40) and (41), R_{bd} and τ_t act as the dominant variables when Y_{bd} and ρ_{bd} remain nearly constant. The cubic dependence of R_{bd} outweighs the corresponding increase in τ_t leading to an overall

enhancement in electrolysis performance. This improvement is associated with microlayer-driven mass flux over a wider electrode coverage area, promoting rapid bubble growth. However, it should be noted that the enlarged departure radius at high contact angles may require careful consideration of electrode spacing in practical configurations. It is further noted that the deviations in τ_t , $\dot{m}_{bh}$ and η between the initial and cyclic simulations become significant only for $\theta_1 < 25^\circ$ and diminish beyond this value. For larger contact angles, the dissolved H_2 remaining near the electrode after detachment becomes sufficiently weak to influence subsequent nucleation and growth. These results indicate that cyclic effects on bubble growth dynamics and electrolysis performance are pronounced at small contact angles and can also become significant for larger contact angles when the electrode size is sufficiently large.

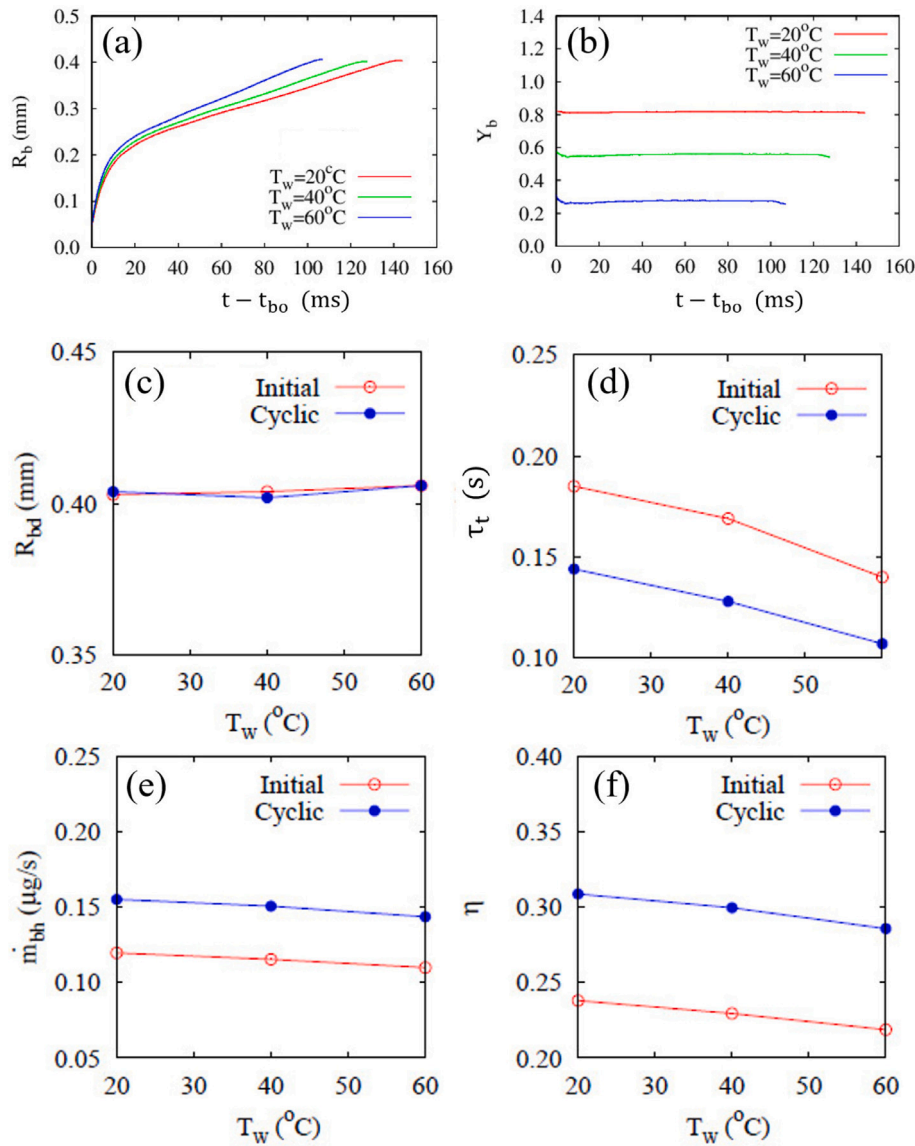


Fig. 10. Effect of wall temperature T_w on the numerical results at $R_e = 0.2$ mm, $i_w = 38.5$ A/cm², and $\theta_1 = 14^\circ$: (a) temporal variation of the bubble radius R_b over a cyclic period; (b) bubble departure radius R_{bd} ; (c) total growth time τ_t ; (d) hydrogen bubble production rate $\dot{m}_{bh}$; and (e) hydrogen evolution efficiency η for the initial and cyclic periods.

Fig. 14 presents the numerical results obtained for different waiting periods τ_w at $R_e = 0.2$ mm, $i_w = 38.5$ A/cm², $T_w = 20^\circ$ C, and $\theta_1 = 14^\circ$. In the present study, a fixed waiting period of $\tau_w = 5$ ms is adopted, corresponding to the minimum threshold required to maintain a periodic bubble growth-departure cycle without bubble merging. Since bubble coalescence represents a different growth regime, it is not considered in the present work and will be addressed in future studies. As τ_w increases from 5 ms to 20 ms, the bubble exhibits a slightly faster growth rate, as shown in Fig. 14(a), owing to the longer accumulation time of dissolved hydrogen near the electrode surface. However, the total cycle period increases to $\tau_t = 145.3$ ms, 147.7 ms, and 152.8 ms, indicating that the increase in waiting time outweighs the reduction in the bubble growth time. Consequently, the hydrogen production rate $\dot{m}_{bh}$ slightly decreases with increasing τ_w as shown in Fig. 14(b). In addition, the hydrogen evolution efficiency η , gradually decreases as τ_w increases because a larger amount of dissolved hydrogen diffuses into the electrolyte during the extended waiting period. Overall, although the waiting period tends to reduce both the hydrogen production rate and the hydrogen evolution efficiency, its influence remains relatively weak compared with other governing parameters such as electrode size,

current density, wall temperature, and contact angle. In practice, the waiting period may vary depending on the local dissolved hydrogen concentration and supersaturation level, and therefore may not remain constant under different operating conditions. This suggests that the incorporation of a variable waiting-period model would be an important extension of the present framework and will be considered in future work.

4. Conclusions

A level-set-based numerical framework was developed to compute cyclic hydrogen bubble growth in water electrolysis by incorporating microlayer evaporation and interfacial mass transfer. Successive nucleation, growth, and detachment processes were resolved until convergence toward a quasi-periodic regime was achieved. The results show that dissolved hydrogen accumulated during bubble growth and partially retained after departure influences subsequent bubble dynamics when the electrode size is sufficiently large, whereas cyclic effects become negligible for small electrodes or large contact angles. Consequently, evaluating hydrogen production rate and hydrogen

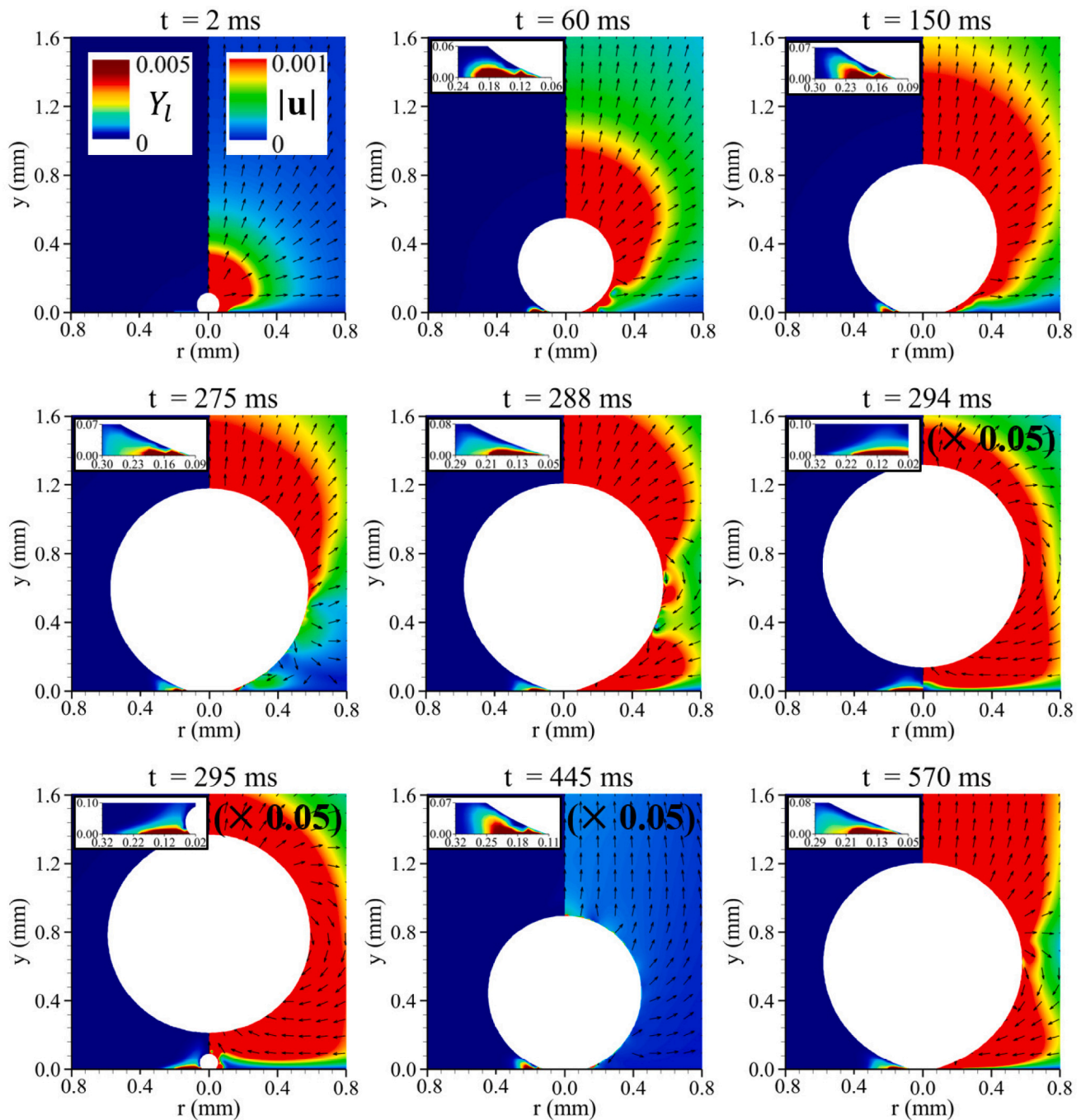


Fig. 11. Dissolved hydrogen concentration field (left) and liquid velocity field (right) around the growing bubble interface at $R_e = 0.2$ mm, $i_w = 38.5$ A/cm², $T_w = 20$ °C, and $\theta_1 = 20$ °. For clarity, the velocity vectors shown for $294 \text{ ms} \leq t \leq 326 \text{ ms}$ are scaled by a factor of 0.05. The insets in the upper-left corner provide enlarged views of the dissolved hydrogen concentration field near the bottom region of the bubble. The white region represents the bubble phase.

evolution efficiency based solely on the initial growth cycle may significantly underestimate realistic electrolysis performance. Increasing electrode size or current density enhances hydrogen bubble production rate primarily through reduction of the growth period, but hydrogen evolution efficiency decreases due to intensified diffusion losses over enlarged active regions. Under constant total current conditions, enlargement of the electrode beyond a certain length scale promotes diffusion losses and reduces overall performance. Elevated wall temperature accelerates bubble growth through intensified vapor phase change but does not necessarily improve hydrogen utilization efficiency, whereas larger contact angles increase departure size and can

enhance hydrogen production and evolution efficiency when the increase in bubble volume outweighs the accompanying extension of growth time. It should be noted that the present model incorporates temperature effects only through phase change and interfacial transport mechanisms. Temperature-dependent electrochemical kinetics, overpotential, and catalytic activity are not considered. Therefore, the reported hydrogen evolution efficiency, (η), should be interpreted within the framework of transport-related effects only. These findings demonstrate that accurate prediction of microscale electrolysis performance requires integrated consideration of microlayer transport and cyclic bubble dynamics.

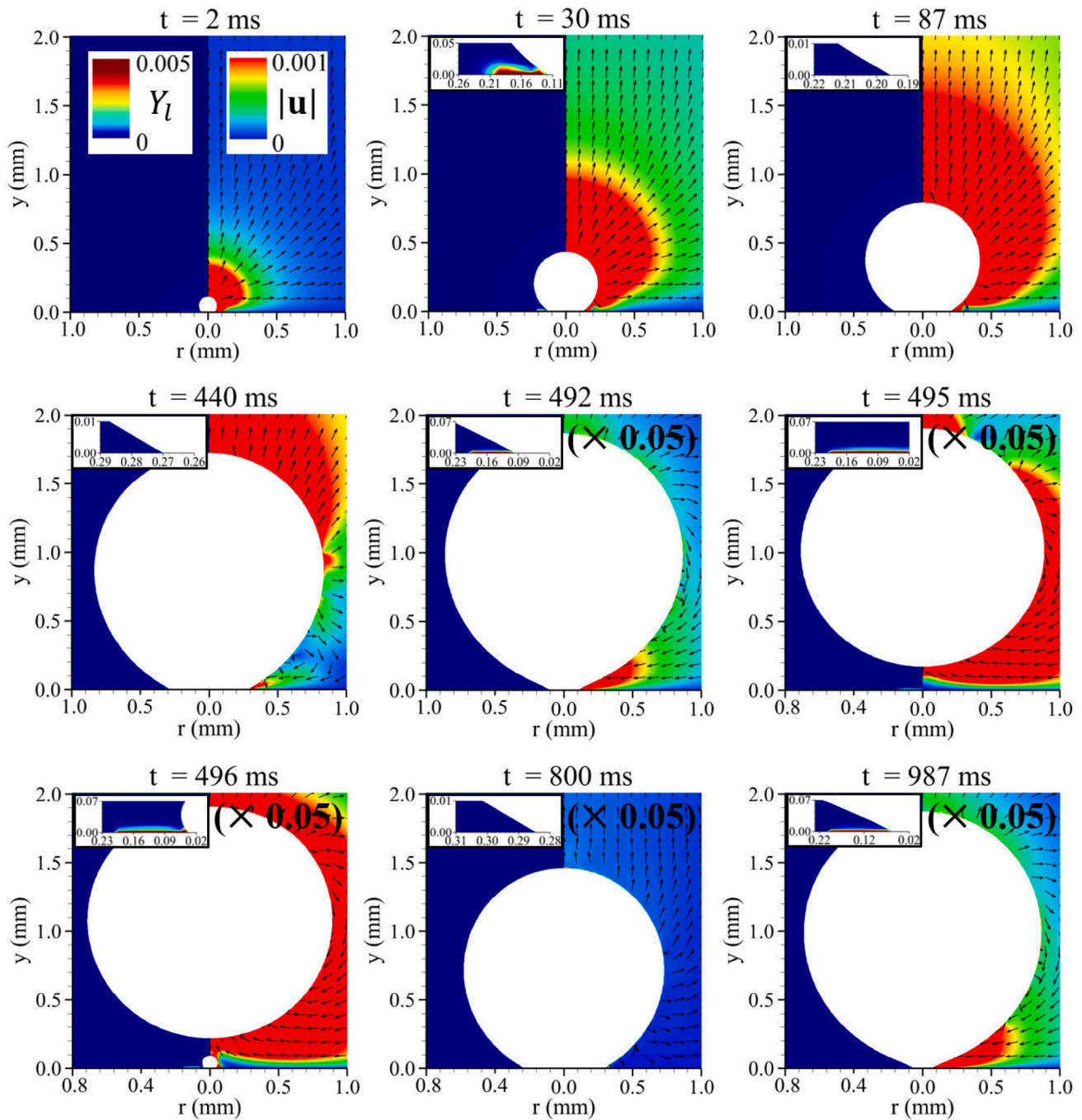


Fig. 12. Dissolved hydrogen concentration field (left) and liquid velocity field (right) around the growing bubble interface at $R_b = 0.2$ mm, $i_w = 38.5$ A/cm², $T_w = 20$ °C, and $\theta_l = 30$ °. For clarity, the velocity vectors shown for $294 \text{ ms} \leq t \leq 326 \text{ ms}$ are scaled by a factor of 0.05. The insets in the upper-left corner provide enlarged views of the dissolved hydrogen concentration field near the bottom region of the bubble. The white region represents the bubble phase.

Furthermore, the underlying mechanisms resolved in the present study — interfacial mass transfer, phase change, and bubble-induced transport — are not limited to electrochemical systems but are also relevant to a wide range of thermal processes. In particular, the proposed framework can be extended to phase-change heat transfer systems such as nucleate boiling and evaporation-driven cooling, as well as microscale thermal management applications. It is also applicable to gas-evolving electrochemical systems beyond water electrolysis and to coupled thermo-electrochemical processes, where temperature variations strongly influence interfacial transport, bubble dynamics, and overall system performance.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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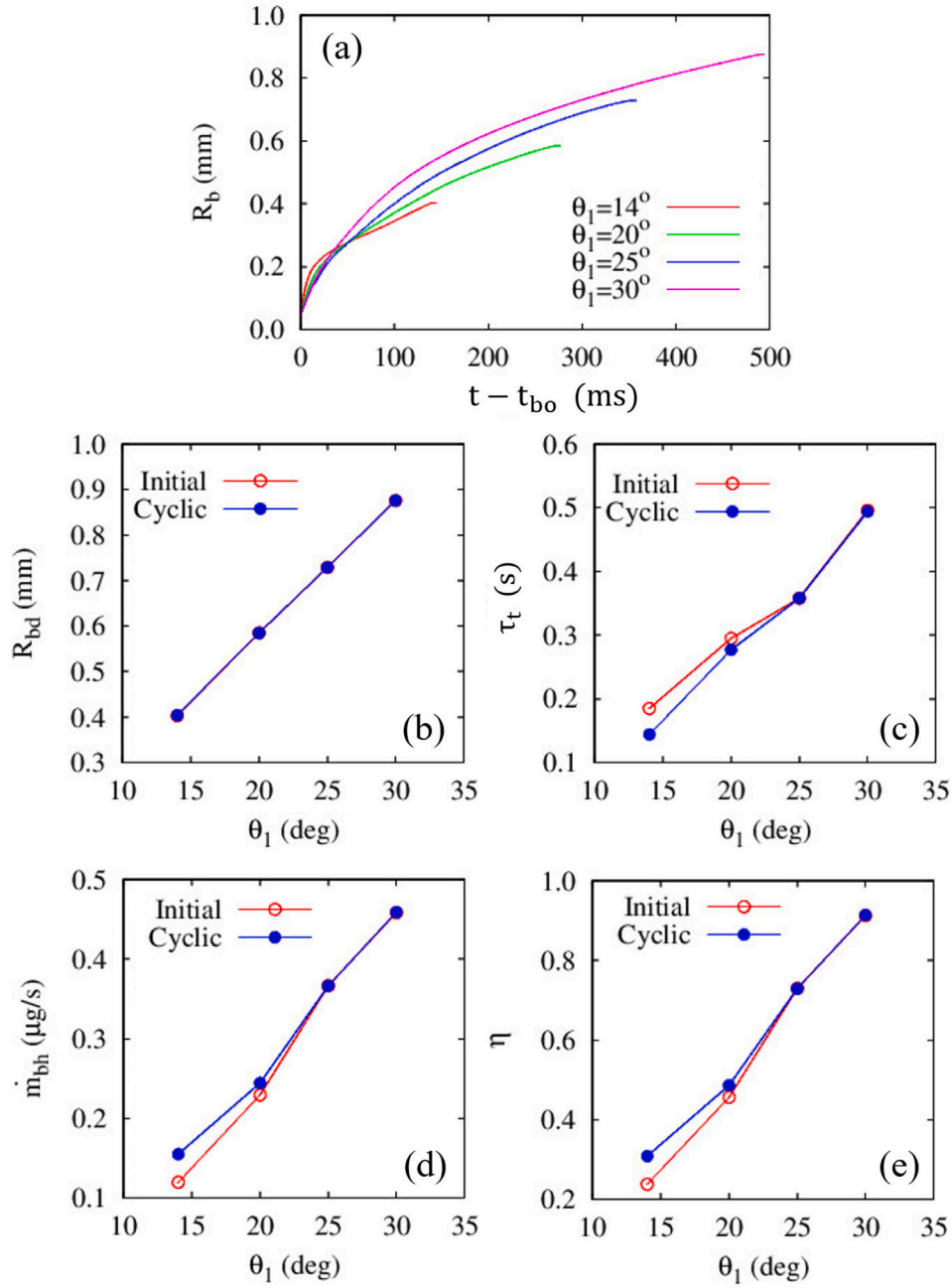


Fig. 13. Effect of contact angle θ_1 on the numerical results at $R_e = 0.2 \text{ mm}$, $i_w = 38.5 \text{ A/cm}^2$, and $T_w = 20^\circ\text{C}$: (a) temporal variation of the bubble radius R_b over a cyclic period; (b) bubble departure radius R_{bd} ; (c) total growth time τ_t ; (d) hydrogen bubble production rate $\dot{m}_{bh}$; and (e) hydrogen evolution efficiency η for the initial and cyclic periods.

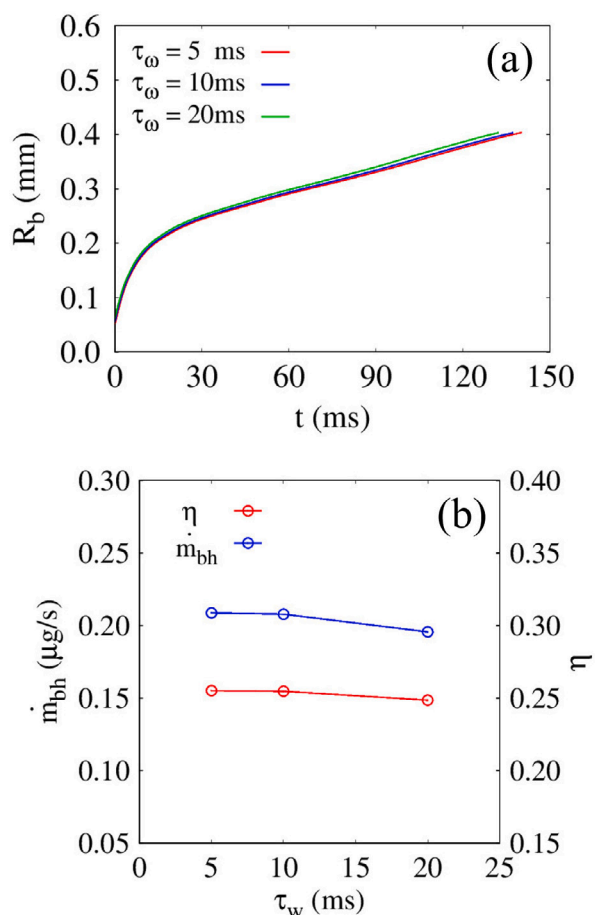


Fig. 14. Effect of the waiting period, τ_w , on (a) the averaged bubble radius, R_b , and (b) the electrolysis performance, $\dot{m}_{bh}$ and η , at $R_e = 0.2$ mm, $i_w = 38.5$ A/cm², $T_w = 20$ °C, and $\theta_1 = 14^\circ$.

Data availability

Data will be made available on request.

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