



Modeling of electrochemically generated bubbly flow under buoyancy-driven and forced convection



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ABSTRACT

This work is devoted to the modeling of two phase flows arising in typical electrolysis devices. A numerical mixture model is used in order to resolve the two dimensional bubble plumes evolving along the electrodes. Plumes thickness sensitivity is studied for various parameters, such as bubble diameter, electrolyte viscosity, electrochemical cell geometry and current density. Using thermal buoyancy driven flow analogy, a dimensionless *Rayleigh*-like number $R\alpha_{f,e}$ is defined to predict the behavior of the wall-bounded gas convection between two vertical facing electrodes. Different bubbles dispersion mechanisms are observed depending on two-phase flow dynamics and physical properties of the mixture. The effect of forced convection in the channel is also investigated. A scaling law for plume thickness evolution for a large range of *Prandtl*-equivalent number values is proposed. These results show that the bubble plume can be efficiently controlled by an imposed electrolyte velocity.

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1. Introduction

The efficiency of industrial electrochemical processes strongly depends on mass transfer. For example, electrochemical reactions can be enhanced by pumped electrolyte flow, which increases mass transfer at the electrode surface. Alternatively, a flow can be induced by the electrochemical bubble production. Therefore, the cell efficiency of many electrochemical industrial processes is affected by bubble existence, such as chlorate process, where hydrogen bubbles are produced at the cathode [1]. In Hall–Héroult process for the production of aluminum [2], carbon monoxide and carbon dioxide are produced at the bottom of the anode. Hence, bath/metal interface is affected by gas bubbles. Concerning zinc–air fuel cells, the performance is determined by active surface area, of which morphological modifications during charge and discharge are impacted by convection, among other mechanisms [3]. Furthermore in the energy field, Zeng and Zhang [4] showed that the additional resistance arising from partial coverage of the electrodes by the bubbles was critical in alkaline water electrolysis efficiency. A better understanding of bubble behavior would provide a scientific guidance to minimize this resistance and contribute to the development of hydrogen production. Generally, at

gas-evolving electrodes, the actual distribution of current density differs from its nominal value. According to Vogt [5], empirical descriptions may be used in order to describe the gas evolving electrodes but are restricted to some bubble coverage ranges and particular operating points.

Nagai et al. [6] have shown that the decrease of electrolysis efficiency occurs by the increase of void fraction between electrodes. Due to the coupling between the electrochemistry and the two-phase flow, process performances are dependent on numerous experimental parameters such as electrode and reactor geometries, current density, electrochemical kinetics and electrolyte concentration. Even though the combined influence of all these factors might be complex to predict, the fact remains that optimum operating conditions can be found from experimental results together with a physical model of void fraction evolution. Furthermore, extrapolating results from laboratory to pilot and industrial scale requires careful consideration. Dimensionless parameters and correlations can be used to design and characterize industrial electrochemical cell, as for example, in the case of filter-press reactors [7].

However, in many industrial electrochemical applications (e.g. chlorate process, waste treatment, redox flow battery and electrolysis) there is a lack of practical tools to characterize, optimize and scale-up reactors.

The purpose of this paper is to describe how the mechanisms of bubble dispersion affect the global flow by means of a *mixture* (also

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Nomenclature

A	void fraction scale
D	non-dimensional hydrodynamic diffusion coefficient
D_H	hydraulic diameter (m)
e	half inter-electrode gap (m)
\vec{e}_x, \vec{e}_y	horizontal and vertical unity vectors
f	hindering function
F	Faraday constant (C mol ⁻¹)
g	gravitational constant (m s ⁻²)
H	inter-electrode gap (m)
i	current density (A m ⁻²)
i_0	exchanged current density (A m ⁻²)
i_{av}	average current density (A m ⁻²)
I	identity matrix
K_α	void fraction dispersion coefficient (m ² s ⁻¹)
L	electrode length (m)
L_{in}	laminar entrance length (m)
p	absolute pressure (Pa)
P	reduced pressure (Pa)
Pr_α	Prandtl-equivalent number
q	mass-averaged velocity (m s ⁻¹)
r_b	bubble radius (m)
R	universal gas constant (J mol ⁻¹ K ⁻¹)
Re_{D_H}	channel Reynolds number
$R\alpha_{f,e}$	Rayleigh-equivalent number
T	operating temperature (K)
U	superficial velocity (m s ⁻¹)
U_{Hdiff}	hydrodynamic self-diffusion (m s ⁻¹)
U_{H2}	hydrogen flux generation (m s ⁻¹)
U_g	gas flux generation (m s ⁻¹)
U_{mig}	shear-induced migration (m s ⁻¹)
U_{O2}	oxygen flux generation (m s ⁻¹)
U_{Saff}	Saffman lift (m s ⁻¹)

U_{Sdiff}	shear-induced diffusion (m s ⁻¹)
U_{Stokes}	Stokes flux (m s ⁻¹)
U_r	void fraction relative flux (m s ⁻¹)
v	phase velocity (m s ⁻¹)
v_{Stokes}	Stokes velocity (m s ⁻¹)
V	vertical velocity scale (m s ⁻¹)
W	horizontal velocity scale (m s ⁻¹)
x	horizontal position (m)
y	vertical position (m)

Greek symbols

α	void fraction
β	non-dimensional shear-induced diffusion coefficient
$\dot{\gamma}$	shear rate (s ⁻¹)
δ_α	void fraction boundary layer thickness (m)
δ_H	hydrodynamic boundary layer thickness (m)
δ_T	thermal boundary layer thickness (m)
η	overpotential (V)
κ	non-dimensional shear-induced migration coefficient
μ	dynamic viscosity (Pa s)
ν	kinematic viscosity (m ² s ⁻²)
ρ	density (kg m ⁻³)
σ	electrolyte conductivity (S m ⁻¹)
τ	shear stress (Pa)
ϕ	electrolyte potential (V)

Subscripts

C	continuous phase
D	dispersed phase
x	horizontal component
y	vertical component

called *drift-flux model*. While an Eulerian–Eulerian two-fluid model (2FM) [8] or an Eulerian–Lagrangian liquid-bubble representation [9] would presumably be both more rigorous in the description of the dispersed phase, they would induce higher numerical costs. The *mixture model* – simpler in its formulation and resolution – is well suited for the present study due to its good efficiency in the case of unidirectional flows and small bubbles [10]. A new boundary layer model is derived from this *mixture* formulation and with it, a *Rayleigh-like* and a *Prandtl-like* dimensionless numbers to characterize bubble-driven convection and plume behavior. It is worth mentioning that the analogy between two-phase flows and heat transfer can be found in the literature by using for instance the Rayleigh–Benard instability analogy [11–13] or nucleate boiling heat transfer properties for describing mass transfer at the electrodes [14].

2. Model description

2.1. General description

Fig. 1 shows an illustration of a standard electrolysis set-up and represents the configuration of the model. The facing electrodes are vertical plates with infinite depth (2D approximation). x and y stand respectively for the horizontal and vertical directions. The cathode is located at $x = 0$, the anode at $x = 2e$ and $y = 0$ is set at the entrance of the channel. The electroactive length is noted L and the inter-plate gap is then $2e$. Generated bubbles evolve in plumes along the walls. The electrolyte flow can either be forced

into the channel or be induced by buoyancy forces arising from the bubbles creation.

2.2. Mixture model

In the present study, the two-phase flow dynamics is modeled by the *mixture formulation* as expressed by Ishii [15]. The quantities \vec{v}_D and \vec{v}_C are respectively the dispersed and the continuous phase velocities. \vec{q} and \vec{U} are respectively the *mass-* and *volume-averaged* (also called *superficial*) velocity fields of the mixture and

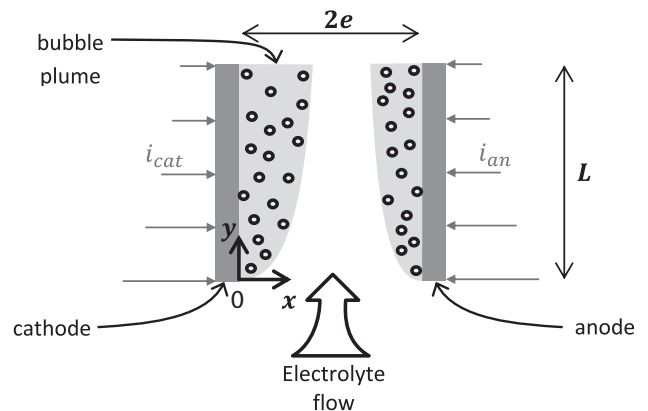


Fig. 1. Schematic representation of the electrochemically generated bubbly flow.

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