Contents lists available at ScienceDirect

Chemical Engineering Research and Design

journal homepage: www.elsevier.com/locate/cherd

Electrogenerated bubbles induced convection in narrow vertical cells: A review

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ARTICLE INFO

Article history:
Received 17 February 2015
Received in revised form 18 May 2015
Accepted 24 May 2015
Available online 30 May 2015

Keywords:
Electrochemical reactors
Electrogenerated bubbles
Electrolysis
CFD
Hydrodynamics
Metrology

ABSTRACT

The present paper focuses on Vertical Plane Electrode Reactors with Gas Electrogeneration, VPERGEs, which are involved in various industrial applications in large scale productions. It is well known that the efficiency of such electrochemical reactors is closely linked to the hydrodynamics of the electrogenerated bubbles which act as moving electrical insulators. Therefore, an accurate understanding of the two-phase flow hydrodynamics in these reactors is of prime importance to optimise their geometry and operation.

However, the small inter-electrode space in VPERGEs is problematic for both experimental and numerical (CFD) investigations. In fact, because of the narrowness of the gap separating the electrodes, many key aspects of the multiphase flow field are inaccessible to experimental measurements, while on the other hand, CFD models typically used nowadays are at the limit of validity.

This paper presents a review of articles dealing with experimental and numerical investigations of the flow in VPERGE reactors that were classified into four reactors type depending on their geometry and on the operating conditions. Such classification is necessary to enable extracting general information on the flow features. Focus is put in particularly on the advantages and drawbacks of the experimental techniques and numerical approaches encountered in the literature.

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http://dx.doi.org/10.1016/j.cherd.2015.05.035

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1. Introduction

Electrochemical processes are involved in various industrial applications of increasing importance such as electroplating, production of valuable gases e.g., hydrogen, chlorine and fluorine (Kuhn, 1971; Kreysa and Kuhn, 1985), electrochemical neutral pickling (Ipek et al., 2008), and more recently carbon dioxide-free metal reduction from ores such as iron electrolytic preparation (Allanore et al., 2010). In many electrochemical systems, gas is generated at a single or at both electrodes, either as the desired product or by undesired side reactions. In the case of vertical electrodes, gas is released into the electrolyte solution as dispersed small bubbles, whose behaviour and significance in the vicinity of the electrode depend on the current density.

The mechanism of bubbles electrogeneration is the following: the electroactive species (e.g., H^+ or OH^-), being an ion or a molecule, is transferred to the electrode, and upon charge transfer, is converted into dissolved molecular species (e.g., H_2 or O_2) which is gaseous under usual conditions. When its local concentration exceeds the saturation level (i.e., supersaturation), the produced molecule undergoes a phase transition into gas state. The heterogeneous nucleation occurs on preferential sites where pre-existing gas nuclei are trapped into the imperfections of the electrode surface. Due to the dissolved gas transfer from the surrounding supersaturated electrolyte, these nuclei grow into gas bubbles adhering to the electrode. When a bubble reaches a sufficient size, it departs from the solid surface, leaving some residual gas that keeps the nucleation site active. The detached bubbles rise up due to buoyancy force and form a two-phase layer in the vicinity of the electrode (Fig. 1a) that is referred to as bubbles' curtain. Generally, plane electrodes are oriented vertically so as to prevent bubbles accumulation. Such electrochemical cells are referred to here as Vertical Plane Electrode Reactors with Gas Electrogeneration, VPERGEs. It should be underlined that gas bubbles behaviour in electrochemical reactors has numerous common features with boiling, in spite of existing differences between the two phenomena, as discussed by Vogt et al. (2004).

The efficiency of electrochemical processes is closely linked to the hydrodynamic behaviour of the bubble curtain(s). First, bubbles accelerate the electrolyte flow near the electrode and enhance agitation; they have therefore a pronounced impact on the convective transport of electrochemically active species (as well as on heat transfer). Secondly, bubbles act as moving electrical insulators, thus affecting the current density distribution (Fig. 1a) and increasing the ohmic drop across the reactor. To limit this side-effect, several electrode configurations allowing an efficient bubbles' disengagement have been proposed (Kreysa and Kuhn, 1985; Ullmann, 1998; Jüttner, 2007). Other alternatives consist in increasing the operating pressure to reduce the gas's volume and increase its solubility, or in circulating the electrolyte using a mechanical pump. This last option is adopted in most industrial electrolysers since the forced convection also increases mass and heat transfer efficiencies.

Studies dealing with the hydrodynamics in VPERGEs have considered different operating conditions and reactor configurations, what makes it difficult to extract general information on the flow characteristics. Moreover, in most papers, the influence of the reactor configuration on the hydrodynamics in the reactor is not recognised in the literature survey part, which often leads to systematic assessment errors. Therefore, since the electrochemical cell's geometry has a strong impact on the flow field, we have classified the different reactor configurations encountered in the literature into four groups as illustrated in Fig. 1. The first one, referred to here as Single Electrode Configuration (SEC, Fig. 1a), consists of a single electrode, i.e. it is supposed that the counter electrode is positioned sufficiently far away. It has been employed for numerical flow investigation by Dahlkild (2001) for simplification purposes. The second design – Forced Convection Design, FCD, Fig. 1b – is the most representative of industrial processes: the electrolyte is circulated in the electrode gap by means of an external pump. In the third configuration, called here Free Convection Induced Circulation Configuration, FCICC (Fig. 1c), a net flow is generated in the electrode gap due to the rising bubbles induced convection. This geometry corresponds to important electrochemical applications, for example the chlorate process where the overall flow is generated by the aforementioned airlift effect (Boissonneau and Byrne, 2000). In the fourth design – No Net Flow Configuration, NNFC (Fig. 1d) – the free surface of the liquid prevents the occurrence of a net liquid flow in the cell: near the electrodes, the electrolyte is dragged upward by the gas bubbles and moves downward near in the central part of the cell, forming two recirculation loops. This configuration is not encountered in industrial processes, but has often been addressed in order to characterise more precisely some fundamental aspects of the flow, for example spreading of the bubble curtain(s) (Fig. 1). In fact, under forced convection, this effect generated by the bubble curtain can be masked by the turbulent dispersion or by the intense convection of bubbles.

In an attempt to summarise the advancements in the domain, this paper presents a literature survey on flow phenomena in VPERGE cells. The literature coping with the experimental and numerical investigations of the flow in VPERGEs is reviewed in Sections 2 and 3, respectively. Attention is focused on the advantages, the drawbacks and the limits of the various experimental techniques and numerical approaches encountered in the literature. It should be underlined that in this particular context, a strong complementarity