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Current density distribution and gas volume fraction in the gap of lantern blade electrodes

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ABSTRACT

Electrochemical processes involving gas evolution at appreciable rates have been optimized in their design for the sake of reduced energy consumption. The present investigation was conducted in view to reducing the energy demand of a pilot process for electrolytic reduction of hematite particles to iron metal; attention was paid at the design of the lantern blade anodes where oxygen evolution occurs. An experimental cell consisting on two facing anodes and two remote cathodes has been designed and used for investigation of the gas behavior and current density distribution at the anode blades. The model for prediction of secondary distributions was validated by measurement of the currents at the segmented anodes and the effects of the average current density and the anode gap could be observed. The model was finally applied to the pilot cell for iron production; as expected, larger gaps allow more uniform current distributions at the anode, however without reducing the cell voltage. In contrast, blade lengths in the order of $10\text{--}15 \times 10^{-3}$ m only would allow visible reduction in the cell voltage.

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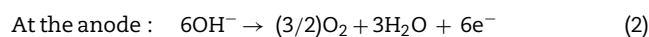
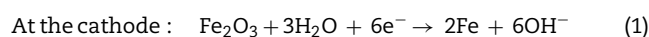
Keywords: Gas evolving electrodes; Current density distributions; Modeling; Segmented electrodes; Electrochemical iron production

1. Introduction

Electrochemical processes involving the production of gas have been optimized for decades in their design and operation for minimum energy consumption, by both improvement of the electrode materials and minimization of the ohmic drop, which is to be enhanced by the presence of non-conducting gas bubbles in the cell gap. The design of gas evolving electrodes has been the topics of numerous investigations; a possible design allowing prompt disengagement of gas bubbles from the solution has often consisted in lantern blade electrodes (Kuhn, 1971; Schmitt et al., 1995) formed by a series of inclined parallel blades. Bubbles evolving from the surface of the two faces of the blades leave the cell upward by Archimede's forces, with little influence of the possible convection of the fluid between the anode and the cathode.

In addition to usual electrochemical processes for gas production, e.g. water electrolysis or chlorine-alkali process, gas

evolution can occur at the counter electrode of cells designed for other applications such as electrosynthesis or metal deposition. In particular, an electrochemical process has been suggested years ago for production of steel and iron from iron ores such as hematite through their cathode reduction in a 50% NaOH solution at temperature near 110 °C (Allanore et al., 2007, 2010; Lavelaine and Allanore, 2008); this process far from commercialization had been proposed as an alternative from the conventional carbon-based reduction of iron oxide into iron with tremendous CO₂ emissions. As a matter of fact, in the current technology around 1.9 tons of CO₂ are produced per ton of iron (Worldsteel Association, 2012); 25% of CO₂ emissions from industrial activities originate from iron industry (International Energy Agency, 2007). The following reactions occur in the iron production cell:



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Nomenclature

A	Active area of electrode (m^2)
C_b	Concentration of the electroactive species in the bulk (mol m^{-3})
D	Diffusion coefficient of the electroactive species ($\text{m}^2 \text{s}^{-1}$)
e	Anode gap (m)
E_0	Equilibrium potential of the electrode (V)
F	Faraday constant ($96,485 \text{ C mol}^{-1}$)
I	Current (A)
i	Current density (A m^{-2})
i_0	Exchange current density (A m^{-2})
$i_{c,av}$	Average current density at the cathode (A m^{-2})
I_i	Current in the i th anode segment (A)
l	Anode-to-cathode gap (m)
L	Blade length (m)
n	coordinate perpendicular to the surface
n_e	Number of electrons involved in the reaction
Q	Total charge passed through the electrolyte (As)
T	Temperature (K)
t	Time (s)
U_c	Cell voltage (V)
U_{oc}	Reversible cell voltage (V)
V_g	Volume of gas phase (m^3)
V_l	Volume of liquid phase (m^3)
U_c	Cell voltage (V)
U_c^{eq}	Standard state cell voltage (V)
x, y, z	Cartesian coordinates

Greek symbols

β_a	Anode Tafel parameter (V^{-1})
ε	Precision of numerical calculations
ε_s	Surface fraction of the gaseous phase
ε_v	Volume fraction of the gaseous phase
η_a	Anodic activation overpotential (V)
η_c	Cathodic activation overpotential (V)
κ	Conductivity of gas-liquid mixture (S m^{-1})
κ_0	Conductivity of the solution (S m^{-1})
μ	Dynamic viscosity (Pa s)
φ	Potential (V)
$\Delta\varphi_{ohmic\ drop}$	Ohmic drop (V)

Subscripts

a	Anode
c	Cathode
M	Metal

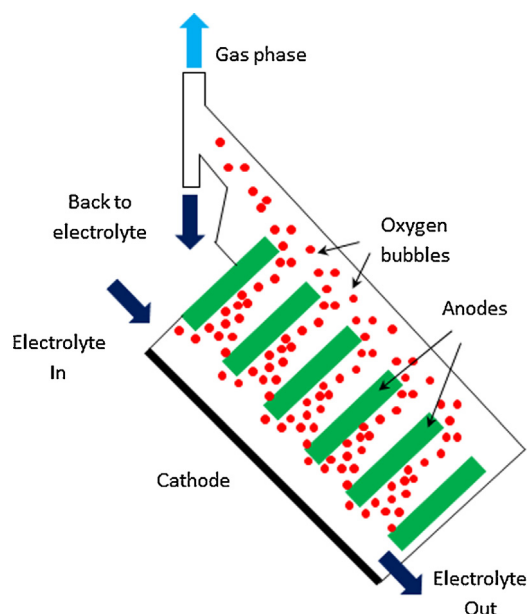
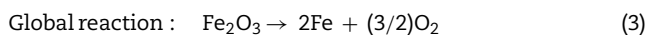


Fig. 1 – General scheme of the pilot cell for electrolytic reduction of hematite to iron metal (Lavelaine and Allanore, 2008).

deviation from the reversible voltage is due to overpotentials of oxygen evolution at the anode and deposition of iron metal at the cathode, and to the ohmic drop in the cell, in the order of 140 mV at 1000 A m^{-2} in the present conditions. Reducing the irreversibility sources in the cell is to reduce the cell voltage and the energy consumption as consequence. In addition to drastic reduction in CO_2 emissions, the electrolytic route is expected to allow iron production with an energy consumption of 3.7 MWh ton^{-1} , in comparison to coal-based processes which consume at least 4 MWh ton^{-1} .

A pilot cell for electrolytic reduction of hematite – a common iron ore – had been imagined and designed by ArcelorMittal for iron ore electrolysis (Allanore et al., 2007, 2010; Lavelaine and Allanore, 2008); the design has been inspired from current electrochemical processes for gas production (Kuhn, 1971; Schmitt et al., 1995). The industrial pilot cell consists in a flat plate inclined cathode and the anode formed by successive blades oriented perpendicular to facilitate the disengagement of oxygen bubbles formed at their surface in the gap, being a few millimeters broad. Both electrodes have been machined out of nickel. The cell is operated with continuous flow of hematite suspension from the top of the cell (Fig. 1). The inclination of the cell was chosen at 45° for regular transport of the hematite suspension in the cell with possible contact of the particles (with a diameter in the order of $10 \mu\text{m}$) at the cathode, and for easier evacuation of the electrogenerated gas. The fed suspension is composed of little soluble hematite particles, sodium hydroxide, and water, each of components at the same weight fraction as made in previous works (Allanore et al., 2007, 2010). The non-reacted part of hematite exits the cell by the bottom outlet with the liquid phase. Bubbles exit the cell by the top outlet of the cell as shown in Fig. 1. The dimensions of the various cell components had not yet been the topic of a full optimization. It can be imagined that long blades could favor gas-lift circulation of both the fluid and the gas; however, too long anode blades are to be avoided since the current density in electrode regions too far from the counter electrode is to be very low in comparison with that on the blade area facing the cathode.



From the differences in free energy and enthalpy of reaction (3), the reversible cell voltage and the thermoneutral voltage are calculated at 1.245 V and 1.422 V at 100°C respectively. The process still under development is a three-phase process, with the presence of hematite and iron as solids, sodium hydroxide solution, and oxygen bubbles as gas phase. In addition to strongly reduced CO_2 emissions – in particular when electricity is not produced by power plants – the electrochemical technique should allow appreciable reduction in the energy consumption; this can be attained by improved current efficiency at the cathode and reduced cell voltage, now in the order of 1.8 V at 1000 A m^{-2} (Lavelaine and Allanore, 2008). The

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