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The oxygen evolution reaction mechanism at $\text{Ir}_x\text{Ru}_{1-x}\text{O}_2$ powders produced by hydrolysis synthesis

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Abstract

A mechanistic study of the oxygen evolution reaction (OER) has been performed for $\text{Ir}_x\text{Ru}_{1-x}\text{O}_2$, $x=1, 0.6, 0.3$ and 0 , prepared by the hydrolysis synthesis. The oxides were characterized by X-ray diffraction, cyclic voltammetry and steady state polarization measurements. The electrolyte pH was varied in order to study the reaction order with respect to protons. The polarization curves recorded could be well fitted to a model consisting of a series of concerted electron-proton transfer reactions (mononuclear mechanism) with either of the second, third, or fourth step being rate determining. The expected trends for this mechanism with respect to potential and pH were observed in the experimental data and are consistent with DFT results for the adsorption energies of the adsorbates [Rossmeis *et al.*, *J. Electroanal. Chem.* **607** (2007) 83 – 89] if the third or fourth step is rate-determining for RuO_2 and IrO_2 , respectively.

The fitting procedures also demonstrate the advantages of working with the full current-voltage expression when analyzing polarization curves, since Tafel behaviour may only prevail in a limited potential region.

Keywords: electrocatalysis, reaction order, water oxidation, iridium oxide, ruthenium oxide

2010 MSC: 00-01, 99-00

1. Introduction

The proton exchange membranes (PEM) water electrolyzer [1] is today considered to play a role in the integration of renewable energy sources. Conversion of electricity into the energy storage medium hydrogen gas can be conveniently and efficiently carried out in PEM water electrolyzers. Production of hydrogen by water electrolysis is traditionally carried out by alkaline water electrolysis (WE) which has an established technology and is widely used in the industry. However, PEM water electrolysis (WE) offers some advantages over alkaline WE, such as higher efficiency, higher current density, more compact design, can be operated under fluctuating current input and has higher purity of the hydrogen gas [2].

Critical to any efficient water electrolysis design are active catalysts for the anode and cathode reactions. Catalyst activities are frequently ranked in terms of the Sabatier principle and so-called volcano curves, as outlined by Parsons [3]. According to the Sabatier principle bonding is necessary for reactions (involving adsorbed intermediates) to proceed, but too strong bonding will block the surface [3]. The rate of an electrochemical reaction at a given potential is thus expected to depend on the binding energy of adsorbates, and the surface should bind these neither too weakly nor too strongly. The volcano curve correlates the bond strength of key adsorbed intermediates to the activity of the catalyst with respect to a certain reaction, and is usually shaped as the name implies as a volcano. For the hydrogen evolution reaction a volcano relationship thus exists between the activity of the catalyst and the bond strength of hydrogen to the metal surface [3], with Pt appearing close to the apex of the volcano.

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