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Gold-supported magnetron sputtered Ir thin films as OER catalysts for cost-efficient water electrolysis

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ABSTRACT

This work presents a research on the preparation of thin composite catalytic films in which an essential part of the efficient but expensive Ir is substituted by Au sub-layer using the preparation method of direct current magnetron sputtering (DCMS). The aim is to investigate the influence of the Au sub-layer on the catalytic activity toward oxygen evolution reaction (OER) of water electrolysis. The properties of the sputtered films are studied using X-ray diffraction (XRD) and electrochemical methods of cyclic voltammetry, quasi steady state polarization and chronoamperometry. It is found that by proper variations in the films thickness it is possible to realize synergetic effects leading to essential decrease in Ir loading and the cost of catalysis without sacrifice in efficiency.

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Introduction

The electrolysis of water in cells with polymer electrolyte membrane (Polymer Electrolyte Membrane Electrolysis Cells, PEMEC) is an attractive and efficient method for producing hydrogen and oxygen at low temperature. The broad use of this otherwise very attractive technology is held back by the high cost of PEMEC components such as the proton exchange membrane and the precious metal electrocatalysts. Since there are still no non-noble metal electrocatalysts with satisfactory activity, it becomes vital to reduce the cost by

improving the specific performance and durability of the well-established noble metal catalysts and decreasing the catalyst loading. On the hydrogen side of the cell, platinum provides the best performance and is commonly used as PEMEC cathode. The most of the overpotential losses however, are related to the electrochemical processes at the anode, where the oxygen evolution reaction (OER) takes place. Therefore, the development and optimization of anode electrocatalysts is of great importance for PEMEC technology. In the past several decades, the electrocatalytic OER has been extensively studied and various catalysts have been designed to improve electrode kinetics and stability under different electrolyte

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environments. It has been proven that several factors influence the electrocatalytic evolution of oxygen including crystallinity and crystallite size, the crystal-field stabilization energy, dispersion, mixed and doped oxides, etc. [1].

Oxygen evolution occurs predominantly on noble metal catalysts (e.g., Pt, Au, Ir, Rh, Ru, Ag). Generally the oxides (e.g., RuO₂, IrO₂) are more active for this reaction than the pure metals. Therefore, the most extensively studied catalysts for OER are Ir, IrO_x, RuO_x [2–4]. IrO₂ and RuO₂ are well established in many industrial electrochemical processes in the form of dimensionally stable anodes [5,6]. IrO₂ exhibits high corrosion resistance but slightly lower electrocatalytic activity than RuO₂, while the latter suffers lower stability [7,8]. Both RuO₂ and IrO₂ however, as precious metal oxides have the drawback of high cost which retards their large scale application.

The establishment of appropriate method for catalyst preparation is essential for the effective utilization of the limited noble metal resources and for the overall OER efficiency. The DCMS is well established technique for thin film deposition, widely used for integrated circuit manufacturing. Recently, it has been also introduced as an alternative of the classical methods for catalysts preparation offering several advantages as: (i) homogeneous and uniform distribution of the metal particles, (ii) precise control of the catalyst loading (down to 10 μg cm⁻²), (iii) tailoring of microstructure and morphology (iv) simplicity and reliability of the preparation process. The sputter deposition has been investigated for the preparation of more effective electrodes in fuel and electrolysis cells [9–13]. The DCMS catalysts are deposited as thin compact (mono, bi- or poly-metallic, and/or oxide) films upon selected substrate material or even directly onto the polymer electrolyte membrane [14–17]. The advantages of the sputtering method compared to the state of the art chemical deposition techniques have been well demonstrated in number of publications. Srinivasan et al. [18,19] sputter deposited a thin Pt film onto the cathode active Pt/C layer and demonstrated that the localization of Pt on the top surface of the electrode results in improved kinetics, higher exchange current density and about 4-fold improvement of the current density. Alvisi et al. [20] studied the sputter deposition of Pt on non-catalysed gas diffusion electrodes and have shown that the sputtering method enables strict control of the Pt cluster size. A very uniform particle distribution was observed resulting in an increased electrochemically active surface and decreased Pt loading relative to a commercial chemically dispersed Pt catalyst. More recently Calliard et al. [21,22] investigated sputtered Pt/C films and found that although they possess lower catalytic activity compared to the chemically deposited carbon supported catalysts, their specific activity is much higher which in turn results in improved utilization. Makino et al. [23] declared an improved mass activity of sputter-deposited Pt cathodes and about 10 times higher utilization compared to the electrode with same Pt loading prepared by the paste method. Slavcheva et al. have investigated sputtered IrO_x and composite IrO_x-Pt-IrO_x catalysts and have reported excellent catalytic properties, mechanical stability, and high corrosion resistance under intensive oxygen evolution in aqueous solutions and in a laboratory electrolyser with polymer proton conductive electrolyte [13,24,25].

This work presents a research on the preparation and investigation of thin composite catalytic films deposited by DCMS in which an essential part of the efficient but expensive Ir is substituted by Au sub-layer. Recently, gold-based bimetallic catalysts have attracted much attention. These films are of interest for two main reasons. First of all a synergism between gold and iridium in bimetallic systems has been reported for heterogeneous catalytic reactions such as total decomposition of volatile organic compounds, CO and ethanol oxidation, etc. [26–29] which implies that in the sputtered Au–Ir films changes in the inter-atomic bonding and coordination of iridium particles, or change in the local atom distribution could be expected, resulting in improved OER performance. Secondly, the partial substitution of Ir by Au will improve the cost-efficiency of the catalysis since gold is cheaper and more abundant in the nature than iridium.

Experimental and procedures

The test samples are sputtered using a magnetron sputtering machine Nordiko 2550, equipped with titanium, gold and an iridium targets with high purity (99.999%). The functional catalytic films are deposited on carbon substrate (Torrax paper) upon 25 nm thick Ti sub-layer serving as an adhesion promoter. Two of the test samples are pure metal films - Au, and Ir with thickness of 250 nm. The Ir is deposited at previously optimized sputtering conditions (dc power of 100 W, total gas pressure in the reactor of 3.2×10^{-2} Torr, and 55 sccm Ar flow rate) ensuring high porosity (resp. high active surface per unit geometric area) combined with good mechanical stability of the films [13,30]. The Au film is much denser. It is deposited at cold sputtering regime in pure argon plasma applying an external electric field with dc power 75 W, total gas pressure in the reactor of 3.2×10^{-2} Torr, and 55 sccm Ar flow rate (parameters used for sputtering of Au contacts in the silicon microtechnology). The composite Au–Ir catalysts consisting of two consecutive functional layers are sputtered at conditions identical to those used for the single metal films. The film thickness is controlled by the deposition time. For the pure metal films it is fixed to 250 nm, while for the Au–Ir samples the thickness of each component is varied in the range 10–250 nm and is denoted in the corresponding sample name, for instance Au240–Ir10 is a sample with 240 nm thick Au sub-layer and a 10 nm thick Ir top layer.

The surface structure and morphology of the sputtered catalysts are studied by X-ray diffraction (XRD). The diffraction data are collected using X-ray diffractometer Philips ADP15 with Cu-K α ($\lambda = 1.54178 \text{ \AA}$) radiation at a constant rate of 0.20 s^{-1} over an angle range $2\theta = 10\text{--}90$. The size of Au and Ir crystallites are determined by Scherrer equation [31].

The catalytic properties of the sputtered Au–Ir films are investigated by conventional electrochemical techniques of cycling voltammetry and quasi-steady state polarization curves. The cyclic voltammograms (CV) are recorded in the potential range between hydrogen and oxygen evolution at scan rate of 100 mV s^{-1} . The quasi steady state polarization tests are carried out in a potentiodynamic mode with scan rate of 1 mV s^{-1} . All experiments are performed with Galvanostat/Potentiostat POS 2 (Bank Elektronik, Germany). The initial electrochemical characterization of the sputtered films is

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