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Performance of a PEM water electrolyser using a TaC-supported iridium oxide electrocatalyst

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ABSTRACT

Polymer electrolyte membrane (PEM) water electrolysis is an attractive way of producing carbon-free hydrogen. One of the drawbacks of this method is the need for precious metal-based electrocatalysts. This calls for a highly efficient utilization of the precious metal, which can be obtained by dispersing the precious metal compound onto a catalyst support. Electrocatalysts with 50, 70 and 90 wt.% of IrO₂ on a TaC support were tested in a laboratory PEM water electrolyser and compared with pure IrO₂. The temperature was set at 90, 110, 120 and 130 °C respectively and the cell voltage was varied between 1.4 and 1.8 V. The load characteristics and electrochemical impedance spectra were obtained and compared for a range of electrocatalysts. The highest current densities and the lowest charge transfer and cell resistances were found for the 70 wt.% IrO₂ electrocatalyst. By contrast, the pure IrO₂ electrocatalyst showed the lowest current densities and the highest charge transfer and cell resistance. For example, the relative difference in current densities between the 70 wt.% IrO₂ and the pure IrO₂ electrocatalyst attained 36% at 130 °C and 1.7 V. All of the supported electrocatalysts showed a higher efficiency of utilization of the precious metal than the pure IrO₂.

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1. Introduction

Proton exchange membrane (PEM) water electrolysis is attracting considerable attention as an environmentally friendly way of producing hydrogen. However, this technology is expensive because precious metal electrocatalysts [1,2] are required on both electrodes. Therefore, the loading of the electrocatalyst has to be reduced to make the process economically viable. The use of an electrocatalyst support is a typical approach to solving this problem. The action of the

support is morefold: first, it provides a high number of crystallisation sites for the formation of the active phase during electrocatalyst synthesis, which leads to a more uniform distribution of electrocatalyst particles and ultimately to an increase in the specific surface area [3,4]. Second, synergies can arise between the electrocatalyst and the support, leading to enhanced electrocatalyst activity [5]. The third effect, specific to anode application for PEM water electrolysis, is the increase in the size of the electrocatalyst particles. Larger particles of a supported electrocatalyst do not penetrate deep into the gas diffusion layer (GDL) during deposition. Since only

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the particles in contact with both the membrane and the current collector can catalyse the reaction [6], the reduced penetration depth results in a higher degree of utilization of the electrocatalyst.

Various forms of carbon are generally used as electrocatalyst supports in PEM fuel cells [7] and can also be used on the cathode of a PEM water electrolyser (PEMWE). Carbon would, however, corrode quickly on the anode because of the strongly oxidative environment. Since there is no commonly accepted anodic electrocatalyst support, research efforts are currently focussing on finding one.

Researchers have investigated various oxidic materials for use as anodic electrocatalyst supports; of these, one of the most commonly investigated materials is SnO₂. While some researchers found performance improvement [4,8], others reported mixed results [9,10]. The biggest drawback of SnO₂ is the added ohmic resistance since it is actually an insulator. To mitigate the lack of electrical conductivity, doping with antimony is a common approach. Some researchers reported that iridium oxide supported on antimony-doped tin oxide performs better than an unsupported electrocatalyst [11–13]. Xu et al. [13] performed a longer-term stability test and reported that the support itself endured for the experiment duration of 760 h. Among other oxides, Ta₂O₅ has been investigated, but it did not display any improvement in performance over a mixed Ir–Ru oxide electrocatalyst [14]. Conductive titanium suboxides, commercially known as Ebonex (Atraverda Ltd, UK), were investigated in another study, but the active part was platinum and, therefore, the results are of limited applicability to PEMWE anodes [15]. In our previous study, various types of TiO₂ were tested as an IrO₂ support. It was found that the anode performance in a PEMWE was significantly better over an unsupported IrO₂ when TiO₂ with a relatively low specific surface area was used [16].

Carbides represent an alternative to metal oxides. Many of them are conductive and thus do not contribute significantly to the ohmic resistance of the electrocatalyst layer. It has been reported that carbide-supported electrocatalysts perform better than unsupported electrocatalysts [17,18]. In our previous work, TaC was studied as a potential new electrocatalyst support [19]. Cyclic voltammetry and Tafel analysis indicated that the performance of electrocatalysts with ≥50 wt.% of IrO₂ was similar to that of pure IrO₂. At that stage, however, the promising results were not confirmed by tests in an actual PEMWE. It is the aim of this study to fill this gap and to provide information on the performance of IrO₂ supported on TaC in a laboratory PEMWE.

2. Experimental

2.1. Materials

TaC-supported IrO₂ electrocatalysts were prepared by a modified Adams fusion method [20]. A detailed description of the procedure can be found in Ref. [19]. The three electrocatalysts indicated in Ref. [19] as the most promising ones were used in this study, viz 50, 70 and 90 wt.% IrO₂ on TaC. A pure IrO₂ electrocatalyst synthesized by the same method was used as a reference. While the IrO₂ had a specific surface area

of 121 m² g⁻¹, the TaC had only 2.4 m² g⁻¹. The specific surface area of the mixtures was found to be a linear combination of IrO₂ and TaC [19]. The crystallite size of IrO₂ was around 5 nm in the supported catalysts and 8.3 nm in the pure IrO₂ (Tab. 1).

2.2. Gas-diffusion electrode preparation

Gas-diffusion electrodes (GDEs) for the PEMWE were prepared by spraying an electrocatalytic ink onto a titanium felt (Fumatech) serving as a GDL. A SEM image of a freshly-prepared electrode activated with IrO₂ shows the uniform coverage of the titanium felt by the electrocatalyst (Fig. 1a). A cross-section of a used electrode with the remains of IrO₂ is shown on Fig. 1b. Most of the IrO₂ originally present on the electrode sticks to the membrane. The used electrode thus shows less IrO₂. The electrocatalytic ink consisted of the electrocatalyst and a Nafion dispersion in isopropanol. The isopropanol mass was 30 times the mass of the electrocatalyst. The ink was ultrasonically homogenized for 30 min beforehand. Nafion serves both as a binder and a proton-conductive phase. Its concentration in the dispersion was such as to obtain 15 wt.% of Nafion in the final electrocatalytic layer. The dispersion was sprayed in several layers on the top of the titanium felt to yield an IrO₂ loading of 1.3 mg cm⁻².

2.3. PEM water electrolyser setup

The prepared anodes were tested in a laboratory single-cell PEMWE at temperatures of 90, 110, 120 and 130 °C. To keep the water liquid and thus ensure a sufficient membrane swelling, both anodic and cathodic compartments of the PEMWE were equally pressurized to 300 kPa. These conditions were found to be suitable in the previous study [16]. Two tantalum-coated stainless steel flow-plates with parallel flow channels were used as the endplates. The active area was 4 cm². A commercial gas-diffusion electrode HT-ELAT (BASF) with a load of 0.5 mg Pt/cm² was used as the cathode. The polymer electrolyte membrane was a Nafion 117 (DuPont). Prior to the application, the membrane was activated by the following procedure: First, the sample was kept in deionized water at 80 °C for 60 min. Then, it was immersed in 3 wt.% H₂O₂ solution at 60 °C for 30 min. Subsequently, it was transferred to the H⁺ cycle by exposure to 0.05 mol dm⁻³ H₂SO₄ at 60 °C for 30 min. Finally, excess H₂SO₄ was removed from the sample in deionized water at 60 °C for 120 min. The PEMWE cell was fastened with 8 M6 bolts at a constant moment of force of 5 Nm. Deionized water with conductivity less than 1.0 μS cm⁻¹ was preheated

Table 1 – Crystallite sizes of IrO₂ and specific surface area of the supported electrocatalysts. Data taken from Ref. [19].

Catalyst, wt.% IrO ₂	Crystallite size, nm	BET specific surface area, m ² /g
100	8.3	121
90	4.3	112
70	4.6	86
50	5.2	60

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