Constructing Supports–Network with N–TiO₂ Nanofibres for Highly Efficient Hydrogen–Production of PEM Electrolyzer

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Abstract: Hydrogen production with a proton exchange membrane (PEM) electrolyzer utilized with renewable energy power is considered to be an efficient and clean green technique, but the poor oxygen evolution performance results in high energy consumption and low efficiency. In this work, a strategy is reported for the construction of a support network of the anodic catalyst layer to simultaneously ameliorate its sluggish reaction kinetics and mass transport in order to realize highly efficient hydrogen production of the PEM electrolyzer. After in situ synthesis of IrO₂ nanoparticles on N–doped TiO₂ nanofibers, the as–prepared IrO₂/N–TiO₂ electrode shows substantially enhanced Ir utilization and accelerated mass transport, consequently decreasing the corresponding cell potential of 107 mV relative to pure IrO₂ at 2 A cm⁻². The enhanced activity of IrO₂/N–TiO₂ could be due to the fact that the N–TiO₂ nanofiber support can form a porous network, endowing IrO₂/N–TiO₂ with a large reactive contact interface and favorable mass transfer characters. The strategy in this work supplies a pathway to develop high–efficiency interfacial reaction materials for diverse applications.

Keywords: catalyst; electrode; hydrogen; efficiency

1. Introduction

PEM electrolyzer technology is a promising alternative to splitting water into oxygen and clean hydrogen, especially as the intrinsic characteristics of a fast response and fluctuation adaptability of PEM electrolyzer are closely integrated with intermittent energy sources [1]. Unfortunately, the insufficient performance due to sluggish oxygen evolution reaction (OER) kinetics together with depressed mass transport gives rise to excessive consumption of electrical power and a high cost of hydrogen production [2,3]; consequently, research efforts devoted to the highly efficient anodic catalysts of PEM electrolyzer have gained attention. To date, numerous research works have been devoted to enhancing electrocatalytic performance, thus minimizing the overall voltage loss [4,5]. Among the modified methods, an attractive alternative that can be carried out to improve the efficiency of hydrogen production is the retention of Ir–based materials in nanoparticles using a support network; not only could this maximize Ir utilization to increase the number of active sites, but it could also realize fast mass transport via the construction of a mass diffusion pathway [6,7].

One–dimensional (1D) nanostructure materials are known for their unique physical and chemical properties, which have been widely applied in fuel cells, lithium–ion batteries, supercapacitors, etc. [8,9]. When employed as supports for a catalyst, 1D nanostructured materials can offer a large specific surface area and expedite electron (ion) transport, contributing to the high electrocatalytic performance of the catalyst–support system [10,11]. On the other hand, the 1D nanostructure could form a support network via the fabrication of the catalyst layer, bringing about the appropriate reaction and diffusion pathways for reactive intermediates and reduction products [12]. Inspired by the above discussion, a
well–designed support network is crucial for a catalyst to display robust reaction kinetics and rapid mass transfer. However, little research has paid attention to developing a 1D nanostructured support network for efficient PEM electrolyzer performance. Moreover, the relationship between the catalyst–support system, mass transport, and reaction kinetics of OER is rarely involved.

In this work, a N–doped TiO$_2$ (N–TiO$_2$) composite nanofiber is developed to construct a support network for IrO$_2$ nanoparticles in order to accelerate the OER kinetics and improve mass transport. IrO$_2$/N–TiO$_2$ is successfully prepared via the solvothermal method followed by heat treatment. SEM and TEM technologies have demonstrated that sub–2 nm IrO$_2$ nanoparticles are well dispersed on N–TiO$_2$ nanofiber support, and the support network of the anodic catalytic layer is effectively constructed. The IrO$_2$/N–TiO$_2$ electrode shows optimized OER activity and low cell potential of 107 mV at 2 A cm$^{-2}$ when compared to that of pure IrO$_2$. Electrochemical measurements confirm the increased active sites, accelerated OER kinetics, and favorable mass transfer. This could be assigned to the accelerated kinetics and rapid mass transport via the construction of the mass diffusion pathway.

2. Experiment
2.1. Preparation of N–TiO$_2$, IrO$_2$, and IrO$_2$/N–TiO$_2$

The hydrothermal method was employed to synthesize N–TiO$_2$ nanofibers. As shown in Scheme 1, 0.3 g TiO$_2$ (P25) and 0.078 g urea were dispersed into a 70 mL NaOH solution (10 M) and ultrasonicated for 30 min. Then, the suspension liquid was transferred to a hydrothermal reactor processed at 200 °C for 18 h. The white precipitate (Na–Ti–N–O) was washed to neutral with deionized water and added to a 500 mL HCl solution (0.1 M) for 24 h. After that, the product was washed to neutral with deionized water again and dried at 60 °C to obtain N–TiO$_2$.

IrO$_2$/N–TiO$_2$ was obtained with heat treatment after polyol reduction (Scheme 1). Briefly, 0.256 g of N–TiO$_2$ and 0.47 g of H$_2$IrCl$_6$ $6H_2$O were added to a 100 mL solution of isopropyl alcohol and water (vol% 1:1). The mixed solution was then heated at 80 °C for 6 h. After cooling down, the solution was filtrated and dried 60 °C. IrO$_2$/N–TiO$_2$ was finally achieved after calcining at 500 °C for 30 min. IrO$_2$ supported on the N–TiO$_2$ support was controlled at 80 wt.%. The IrO$_2$ sample was obtained in a similar manner to that for IrO$_2$/N–TiO$_2$ but without the addition of N–TiO$_2$.
2.2. Preparation of MEA

Nafion 117 membranes acted as the electrolyte and were pre–processed continuously in H₂SO₄ solution, H₂SO₄ solution, and deionized water at 90 °C for 1 h. Pt/C and IrO₂/N–TiO₂, acting as the cathode and anode catalysts, respectively, were sprayed on each side of the N117 membranes to prepare membrane electrode assembly (MEA). The loading mass of Pt and IrO₂ were controlled with 0.4 and 1.5 mg/cm², respectively. Regarding the catalyst ink, a certain amount of the catalyst and ionomer (Nafion solution) was added to the solution of isopropyl alcohol and water (vol% 1:1). The amount of Nafion was 25 wt% for the cathode and anode catalysts. The active area for the prepared MEA was 4 cm².

2.3. Physicochemical Characterization

An X–ray diffractometer (XRD, Bruker D8 Advance X–ray diffractometer) was used to detect the phase and crystal structure. A scanning electron microscope (SEM, FEI QUANTA250FEG*) and transmission electron microscope (TEM, JEOL JEM 2100F microscope) equipped with energy–dispersive X–ray (EDX) spectroscopy and high–angle annular dark–field imaging (HAADF) were used to record the microtopography features of the samples. A Kruss DSA100 system was employed to record the static contact angles of the prepared anodic catalyst layer by the drop shape method using water as the wetting liquid.

2.4. Electrochemical Analysis

Before electrochemical analysis, a homemade PEM single cell assembled with the prepared MEA was inputted into high–purity water for 24 h at a flow rate of 20 mL/min. Cyclic voltammetry (CV) curves of the prepared MEA were measured from the potential from 0 to 1.23 V with a scan rate of 50 mV/s. Polarization curves of the MEAs were performed on an incremental current density from 0.01 to 2 A cm⁻² with a DC supply. Electrochemical impedance spectroscopy (EIS) was tested at 1.5 V at a frequency range of 0.01 to 10 K Hz with an amplitude of 5 mV using a connection of a Solartron 1287 potentiostat and a 1260 frequency response analyzer. The high–frequency resistance values (HFR) of the PEM single cell were taken at the frequency of 10 kHz. All of the electrochemical tests of the PEM single cell were conducted at the conditions of 80 °C and flow rate of 20 mL/min.

3. Results

3.1. Materials Characterization

XRD patterns (Figure 1) were obtained to analyze the crystal structure of the prepared samples. For IrO₂, the peaks located at 28.05°, 34.71°, 40.06°, 54.02°, 57.94°, and 66.05° match well with the (110), (101), (200), (211), (220), and (112) planes, respectively, of the rutile phase IrO₂ (JCPDS 15–0870). The diffraction peaks of N–TiO₂ at about 25.28°, 37.80°, 48.05°, 53.89°, and 55.06° are attributed to the (101), (004), (200), (105), and (211) planes, respectively, of tetragonal anatase TiO₂ (JCPDS 21–1272). Regarding IrO₂, N–TiO₂, and IrO₂/N–TiO₂, it can be seen that no impurity phase was detected.

The SEM image (Figure 2a) confirms IrO₂/N–TiO₂ in the form of 1D nanofibers with an average diameter of 100 nm. TEM (Figure 2b and HRTEM). The images in Figure 2c confirm the well–dispersed IrO₂ nanoparticles on the surface of the N–TiO₂ nanofibers, and the particle size of IrO₂ is mainly less than 2 nm (obtained from the particle distribution statistics). Figure 2d shows the lattice fringe values of 0.352 and 0.258 nm that could be assigned to the (101) crystal plane of N–TiO₂ and the (101) crystal plane of IrO₂, respectively, which is consistent with the XRD results. IrO₂ nanoparticles uniformly supported on the surface of N–TiO₂ nanofibers were further verified by the HAADF image and the corresponding EDX mapping of Ir, Ti, O, and N elements (Figure 2e). The formed nanofiber is expected to supply more nucleation sites for Ir ions, realizing higher Ir utilization, which is favorable to the increase in active sites and enhancement of the mass activity of IrO₂.
Figure 1. XRD patterns of the prepared samples.

Figure 2. (a) SEM image; (b) TEM; (c,d) HR–TEM; (e) HAADF image; and the corresponding EDX mapping of Ir, Ti, O, and N elements of IrO$_2$/N–TiO$_2$.

Figure 3a,b reveal the SEM cross–sections of IrO$_2$ and IrO$_2$/N–TiO$_2$ catalyst layers, respectively. It can be seen that the average thickness of the IrO$_2$/N–TiO$_2$ catalyst layers is 4.85 µm, which is larger than that of IrO$_2$ (3.02 µm). IrO$_2$ catalyst layers show compact microtopography features (Figure 3c), while the IrO$_2$/N–TiO$_2$ catalyst layers reveal a porous network with more porosity (Figure 3d). Although the IrO$_2$/N–TiO$_2$ catalyst layers are thicker than those of IrO$_2$, it can be assumed that the formed porous networks endowed IrO$_2$/N–TiO$_2$ with a larger reactive contact interface and favorable mass transfer characteristics [13].
Figure 3. (a,c) SEM cross-sections and (b,d) SEM images of the IrO₂ and IrO₂/N–TiO₂ catalyst layers, respectively.

3.2. Electrocatalytic Performance Analysis

To explore the support effect of N–TiO₂ nanofibers on the activity of IrO₂, the electrocatalytic performance of the electrode was examined in a homemade PEM single cell (Figure 4a). Carbon paper and Ti mesh were used as the cathode and anode porous transport layers, respectively. Ti plates with flow fields were employed as bipolar plates. Figure 4b shows the measured polarization curves of pure IrO₂ and IrO₂/N–TiO₂ with potential from 0.01 to 2 A cm⁻² at 80 °C. The polarization curve could be divided into three regions: the kinetic-dominated region at the current density of less than 0.15 A cm⁻²; the ohmic resistance-dominated region at the current density of 0.15–1.8 A cm⁻²; and the region dominated by ohmic resistance and mass transport at the current density above 1.8 A cm⁻². Notably, IrO₂ shows a clear increased cell potential that is affected by mass transport when the current density is above 1.8 A cm⁻². The obtained cell voltage values of pure IrO₂ are 1.858 V at 1 A cm⁻² and 2.224 V 2 A cm⁻², while the cell voltage values of IrO₂/N–TiO₂ are 1.797 V at 1 A cm⁻² and 2.117 V 2 A cm⁻². Note that IrO₂/N–TiO₂ has superior electrocatalytic performance to that of IrO₂. Moreover, the electrocatalytic activity of the IrO₂/N–TiO₂ electrode is comparable to those in the recent literature. For example, Lim et al. [14] reported a novel IrO₂ nanoneedle prepared using the molten salt method in which the cell potential of IrO₂ nanoneedles reached >1.8 V at 1 A cm⁻² and 2.15 V 2 A cm⁻². IrOx/Nb–SnO₂ synthesized by Ohno et al. using the flame pyrolysis method showed a high cell potential of 1.91 V at 1 A cm⁻² [15]. Recently, Ramani’s group [16] prepared bifunctional Pt–IrO₂/RuTiO₂ using the co-deposition method; Pt–IrO₂/RuTiO₂ showed a cell potential of >2 V at 1.5 A cm⁻².
Figure 4. (a) Schematics of the home–made PEM single cell; reproduced with permission from Lv, H.; Wang, S.; Li, J.; Shao, C.; Zhou, W.; Shen, X.; Xue, M.; Zhang, C, Appl. Surf. Sci.; published by Elsevier, 2020 [17]; (b) polarization curves of IrO$_2$ and IrO$_2$/N–TiO$_2$ with the potential from 0.01 to 2 A cm$^{-2}$ at 80 ºC.

3.3. Origin of the Efficient Electrocatalytic Performances

To gain insight into the origin of the superior electrocatalytic performance of IrO$_2$/N–TiO$_2$, CV, Tafel slope, and EIS analyses were further carried out. It has been reported that the amount of electrochemically active sites is generally identified by CV curves. Note that IrO$_2$/N–TiO$_2$ shows a current density that is higher than that of IrO$_2$ (Figure 5a), indicating that the increase active sites due to the nanofiber could provide numerous nucleation sites for Ir ions. The ohmic resistance values (Figure 5b) of the PEM single cell at different densities were recorded at the frequency of 10 kHz. In terms of the poor conductivity of N–TiO$_2$, the ohmic resistance values of IrO$_2$/N–TiO$_2$ are higher than those of IrO$_2$. A decreasing trend of ohmic resistance values with the increasing current density could be ascribed to the occurring heat production at high current densities [18]. To explore the effect of support on the reaction kinetics of IrO$_2$, Tafel slope values were further calculated using Tafel’s equation (Equation (1)).

\[
\eta = a + b \log(j)
\]

where \(\eta\) is the overpotential and \(j\) is the current density. Figure 5c shows the calculated Tafel slope, and the slope values are 73 mV/dec and 48 mV/dec for IrO$_2$ and IrO$_2$/N–TiO$_2$, respectively, suggesting that substantially improved kinetics of IrO$_2$ were achieved after the use of supports. This is also verified by the EIS results. As shown in the EIS plots (Figure 5), the charge transfer resistance values for IrO$_2$ and IrO$_2$/N–TiO$_2$ are 71 and 54 m\(\Omega\), respectively. Thus, the increased active sites, smaller Tafel slope value, and charge transfer resistance of IrO$_2$/N–TiO$_2$ indicate that nanofiber supports could efficiently ameliorate the sluggish reaction kinetics of IrO$_2$, which is a benefit of enhancing the OER activity of IrO$_2$.

Figure 6a,b show the contact angle tests of IrO$_2$ and IrO$_2$/N–TiO$_2$ samples using deionized water as the wetting liquid. In addition, the contact angles of IrO$_2$ significantly decreased from 66º to 40º after N–TiO$_2$ introduction, which could be ascribed to fact that the hydrophilic TiO$_2$ nanofibers endow the catalyst–support system with a more hydrophilic surface. The more hydrophilic surface of IrO$_2$/N–TiO$_2$ indicates superior electrolyte penetration and gas adsorption–desorption. To verify the superior kinetics and mass transport of IrO$_2$/N–TiO$_2$, the voltage losses of kinetic losses, ohmic losses, and mass transport losses were estimated [19]. For an electrolysis cell, the cell voltage is the sum of thermodynamic voltage \(E^0(p, T)\), ohmic losses \(Rj\), kinetics losses, and mass transport losses (Equation (2)).

\[
E = E^0(p, T) + Rj + b \cdot \log(j) + \eta_{intx}
\]
Figure 5. (a) CV curves, (b) HFR resistance, (c) Tafel slopes, and (d) EIS of IrO$_2$ and IrO$_2$/N–TiO$_2$.

Figure 6. (a, b) Contact angles and (c, d) voltage losses of IrO$_2$ and IrO$_2$/N–TiO$_2$. 
In Equation (2), the thermodynamic cell voltage $E^0(p, T)$ is calculated from the Nernst equation (Equation (3)):

$$E^0(p, T) = E^0(T) + \frac{RT}{2F} \ln \left( \frac{a(H_2) \sqrt{a(O_2)}}{a(H_2O)} \right)$$  \hspace{1cm} (3)

The thermodynamic cell voltage value is calculated as 1.168 V at 80 °C and atmospheric pressure. Then, the three main potential losses are achieved, as shown in Figure 6c,d. Notably, the mass transport losses and kinetics losses at 2 A cm$^{-2}$ for IrO$_2$ are 12.1% and 71.4%, respectively, while for IrO$_2$/N–TiO$_2$, the mass transport losses and kinetics losses for IrO$_2$ are 9.8% and 70.6%, respectively. Thus, summarizing the above discussion, the increased electrocatalytic activity of IrO$_2$/N–TiO$_2$ could be assigned to the fact that the IrO$_2$/N–TiO$_2$ nanofiber support system can increase reaction kinetics and accelerate mass transport.

4. Conclusions

In summary, an IrO$_2$/N–TiO$_2$ anodic catalyst was prepared with sub–2 nm IrO$_2$ nanoparticles well dispersed on N–TiO$_2$ nanofiber support, and a porous network structure was constructed in an IrO$_2$/N–TiO$_2$ catalytic layer. The IrO$_2$/N–TiO$_2$ electrode showed cell potential of 2.117 V at 2 A cm$^{-2}$, which is lower than that of the IrO$_2$ electrode (2.224 V at 2 A cm$^{-2}$), suggesting the enhanced OER activity of IrO$_2$/N–TiO$_2$. In addition, IrO$_2$ the with N–TiO$_2$ support electrode had more active sites, a smaller Tafel slope value, and higher charge transfer resistance compared with IrO$_2$ without a N–TiO$_2$ support electrode. Moreover, a more hydrophilic surface was observed in IrO$_2$/N–TiO$_2$ after N–TiO$_2$ introduction, which indicates superior electrolyte penetration and gas adsorption–desorption. Therefore, the enhanced electrocatalytic performance of IrO$_2$/N–TiO$_2$ could be ascribed to the increased reaction kinetics and rapid mass transport via the construction of the mass diffusion pathway, as verified by the electrochemical measurements. Constructing support networks of the anodic catalyst layer was demonstrated as a simple and effective method to improve reaction kinetics and favorable mass transport for the highly efficient electrocatalytic performance of the PEM electrolyzer.

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