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Graphical Abstract

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Unraveling mysteries of hydrogen electrooxidation in anion exchange membrane fuel cells

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Review Article

Unraveling mysteries of hydrogen electrooxidation in anion exchange membrane fuel cells

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Anion exchange membrane fuel cells (AEMFCs) can potentially revolutionize the energy generation market; however, to be seriously considered as a real alternative to the mainstream fuel cell technology, complete removal of previous metal electrocatalysts needs to be achieved. While in cathode electrodes platinum can be easily substituted, the electrochemical hydrogen oxidation reaction (HOR) in the AEMFC anodes currently involves prohibitive overpotential losses, making the removal of platinum extremely challenging. Understanding the HOR in AEMFCs will facilitate the path to overcome the challenge and finally develop and demonstrate platinum-free high-performance AEMFC devices.

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Introduction

There is an increasing worldwide interest in anion exchange membrane fuel cells (AEMFCs), as the technology promises significant reduction in costs by avoiding the use of platinum as electrocatalysts [1–4]. Very recently, extensive research in hydrogen AEMFCs led to significant progress in cell performance of this technology [5–10], and to an important increasing in the understanding of the main current challenges – membrane degradation/performance stability [11–16] and CO₂ effect on AEMFC performance [17–24]. This recent progress and the numerous achievements reached in the past years in the technology, suggest that AEMFC can now be considered as a serious alternative to the counterpart proton exchange membrane fuel cells (PEMFCs),

but potentially without expensive electrocatalysts in their cells. In this front however, while for oxygen reduction reaction there are already several potentially good non-precious metal electrocatalyst alternatives under study [25–29], for the hydrogen oxidation reaction (HOR) at the anode of AEMFCs it seems that the challenge is considerably higher, and the way to achieve high anode performance based on low cost, non-PGM catalysts is still very long [30]. The ability to understand and resolve the challenge of hydrogen electrooxidation in AEMFC, in order to develop electrocatalysts highly active towards HOR, is crucial for the future of the AEMFC technology [5,30,31].

About the challenging HOR in AEMFC alkaline environment

The electrochemical reactions of hydrogen oxidation differ, depending on the medium, as shown below. In acidic and alkaline environment, the HOR is given by:

The HOR in the acidic medium (Equation (1)) has been well studied, as this electrochemical reaction is of fundamental importance in electrochemistry and electrochemical devices, including PEMFCs. Numerous studies showing different aspects of this reaction have been published [32–34]. Researchers found that the kinetic rates and activities of different electrocatalytic materials towards HOR in acidic medium, are strongly correlated to the hydrogen energy bonding (HEB) of the adsorbed hydrogen (H_{ad}) onto the catalyst surface [33–35].

In contrast, the HOR in alkaline medium (Equation (2)), although also a reaction of central importance in the electrochemical field, has been scarcely studied. A recent comprehensive review of the current understanding of HOR electrocatalysis in basic medium describes in details the research studies done in this field, highlighting the challenges involved in developing new materials with high activity in this medium [30]. In their review, the authors also focused on the kinetics and the reaction mechanisms of HOR in alkaline medium. Several research

Figure 1

Measured polarization curves for pH=1–13 for (a) Au(111), (a), Pt(111) (b) and Ir-poly. Adopted from [39**].

studies have shown that the kinetics of the HOR in alkaline media is much slower than in acidic solutions [36,37*]. For instance, the HOR activity of carbon-supported noble metals (Pt, Pd and Ir) decreases by around two orders of magnitude when transitioning from low to high pH [38]. The HOR kinetics changes with the pH of the medium were clearly shown by Strmcnik *et al.* [39**]. The authors presented a series of experimental measured polarization curves for the HOR on different catalytic materials in a wide range of pH ($1 < \text{pH} < 13$), as shown in Figure 1. Clear differences between the measured current densities at $\text{pH} < 3$ and $\text{pH} > 11$ can be seen.

The authors also performed a simulation of the polarization curves using a simple set of electrochemical equations, confirming the experimental results shown in Figure 1. Results revealed that in strong acidic medium ($\text{pH} < 3$, corresponding to acidic environment of PEMFCs) the HOR proceeds through the direct oxidation of hydrogen to H^+ (Equation (1)); while in strong alkaline medium ($\text{pH} > 13$, corresponding to acidic environment of AEMFCs), pure diffusion-limiting currents are also observed. As mentioned by the authors, this suggests that in alkaline medium, the supply for both hydrogen and hydroxide anions (both reactants in the HOR in AEMFCs, see Equation (2)) are sufficient to maximize the electrochemical oxidation of hydrogen to water. We can then also say that as the pH in the environment is increased, an increase in the diffusion-limiting currents is obtained. In other words, as the OH^- concentration in the fuel cell is higher, purer diffusion-limiting currents will be observed, indicating then that the OH^- plays an important role in the HOR for AEMFCs. What is the best way to quantify this role of the OH^- is not yet clear.

Is HBE descriptor good enough to evaluate HOR catalyst for AEMFCs?

As shown in Equation (2), in AEMFCs the electrochemical oxidation of hydrogen involves two reactants –

hydrogen and hydroxide anions – instead of only one reactant for the acidic counterpart (Equation (1)) – hydrogen. As hydrogen is a reactant for both acidic and alkaline fuel cells, the differences in the kinetics rates for the HOR on different electrocatalysts in both systems are sometimes attempted to be explained by the variation in HBE values [40]. Although differences in HBE mostly succeed to explain kinetics variations for PEMFCs, in the alkaline counterpart the situation seems to be more complex [37*].

The best catalysts for HOR in acidic PEMFCs are those with an optimal interaction between the catalytic substrate and the H_{ad} species, that usually is quantified by the HBE. Similarly, it was proposed that the role of the OH^- in the HOR in alkaline medium can be related to the OH_{ad} properties of the catalyst [39**]. In other words, it was suggested that fine-tuning of the adsorption energy of OH_{ad} could be a new way to quantify HOR in AEMFC systems [39**]. To quantify this energy, Davydova *et al.* [30**] proposed to calculate a new descriptor called OHBE – OH_{ad} binding energy – and compare it to the HBE for each electrocatalyst system. Similar to what has been suggested by Koper [41], for some electrocatalytic systems, both HBE and OHBE are expected to be interconnected, as the presence of OH_{ad} is probable to improve the reactivity of the adsorbed hydrogen intermediate H_{ad} , enhancing the overall reactivity of HOR, and vice versa. In other words, if this basic idea is correct, it means that the best electrocatalysts for HOR in alkaline AEMFCs are those with an optimal interaction between the catalytic substrate and both the H_{ad} and OH_{ad} species, evaluated by the HBE and OHBE descriptors, respectively. What exactly is the relationship between HBE and OHBE, and how close they are interconnected each other, is currently an unknown.

Very recently, Davydova *et al.* [42] first attempted to evaluate both HBE and OHBE descriptors on their HOR electrocatalysts for AEMFCs (see Figure 2). The authors

Figure 2

Correlation between the experimentally measured exchange current density values of the Ni and Ni-metal catalysts, with the theoretically predicated HBE and OHBE values for (111) and (200) facets (Adopted from [42]).

used density functional theory to calculate the HBE and OHBE values for bare Ni and Ni-metal catalysts, for a few metals (Cu, Co, Fe), and suggested that the dopant metal onto Ni electrocatalyst have a significant effect onto the HBE and OHBE values, showing that these can be useful descriptors to evaluate different catalysts and eventually, relate these values to the exchange current densities. However, this is a first attempt, while this idea is waiting for further exploration, to finally unravel the full role of the hydroxide anions in the HOR.

Ideally, to find the best HOR electrocatalysts for AEMFCs we should then evaluate both HBE and OHBE and correlate the interaction between them and the exchange current densities. This ideal situation is schematically illustrated by a 'volcano plot', where both HBE and OHBE descriptors are simultaneously plotted for different electrocatalysts, as shown in Figure 3. The interaction between the electrocatalytic substrate and the H_{ad} species, quantified by the HBE, is for instance, illustrated for a series of electrocatalysts with different OHBE values by a series of yellow curves. Similarly, interaction between the electrocatalytic substrate and the OH_{ad} species, quantified by the OHBE, for electrocatalysts with different HBE values is shown by the green curves in Figure 3. The optimal fine-tuning of the adsorption energy of H_{ad} and OH_{ad} , which may lead to the optimal HOR electrocatalysts can then be represented by a combination and simultaneous optimization of both HBE and OHBE descriptors. In that case, different fine-tuned HBE/OHBE combinations can lead to electrocatalysts highly active towards HOR for AEMFCs.

Does water play a role in the AEMFC HOR?

During operation of an AEMFC, the higher overpotential of the HOR at high pH anode electrode is also aggravated by the fact that water needs to be quickly removed from the catalyst layer. In contrast to the acidic counterpart, AEMFC operation involves generation of water as a product of the electrochemical HOR. Figure 4(a) shows a schematic diagram of an anode catalyst layer of AEMFCs. The porous of the anode catalyst layer needs not only to allow the hydrogen transport through the entire layer thickness, but also to 'accommodate' the water generated and allow its efficient removal. Two water molecules per reactant hydrogen molecule are generated (see Equation (2)), in absolute contrast to the HOR in acidic PEMFCs, where water is not taken direct part in the electrochemical reaction. This is a significant amount of water that is generated at the AEMFC anode, twice as much as water generated in the cathode of PEMFCs, per hydrogen molecule (or per electron produced), during the reduction of oxygen. In addition to that, in practical fuel cell systems running under standard gas stoichs, the volumetric flow of hydrogen is around 5 to 10 times lower than that of air. Therefore, with these extreme low hydrogen flows and the high amount of water generated by the HOR, water removal from AEMFC anodes is extremely challenging.

The accumulation of water in the AEMFC anode itself represents also a challenge for the supply of the reactants that as previously discussed, are sufficient to maximize the electrochemical HOR in the cell. Due to this fact, the electrolyte/electrode interface of AEMFCs can behave very differently from other similar electrochemical devices. The characterization of interfacial structure and understanding of fundamental characteristics of the AEMFC electrode/electrolyte interface may help shed

Figure 3

Schematic representation of an ideal simultaneous optimization of HBE and OHBE to evaluate and select electrocatalysts highly active towards HOR for AEMFCs.

light on designing and developing highly active non-precious metal HOR electrocatalysts.

Figure 4(b) shows a schematic comparison of the triple-phase boundary (TPB) for the electrooxidation of hydrogen for both acidic and alkaline fuel cell environments. As mentioned above, for AEMFCs the supply of hydrogen as well as the supply of OH^- reactants to the TPB are critical for the HOR. Two OH^- anions (of larger ion radius as compared with the H^+ cation radius in the acidic counterpart) should be transported through the solid anion-conducting electrolyte to react with the hydrogen in the TPB. In addition, as HOR occurs, water is generated in the surroundings of the catalyst. If water is not quickly removed from the TPB, a thin film of water will eventually cover the catalyst surface, creating additional resistance to the diffusive transport of the hydrogen to the TPB (see bottom Figure 4(b)). As previously discussed, removal of this water film from the fuel electrode is challenging, therefore we expect this film to be of significant thickness, affecting then the overall HOR kinetics. We can also assume that

this water film affects the properties of the catalyst, for instance, affecting the HEB and maybe the OHBE and in turn, the HOR activity. Moreover, as the current density in the AEMFC device increases, the amount of water accumulated on the catalyst surrounding increases, generating thicker water films around the catalysts, which in turn are translated to increased resistance of the hydrogen transport through the water film. Taken into account all these effects water has onto the HOR electrocatalysis and AEMFC anodes, would provide a better understanding of the requirements for designing better hydrogen electrooxidation electrocatalysts for this technology.

Concluding comments

The field of AEMFCs has never been more active than in the past couple of years. However, to become a real alternative to the counterpart acidic fuel cells, AEMFCs should be able to equal PEMFC performance while based on affordable PGM-free electrocatalysts. In this work, we have remarked the challenges of HOR in AEMFCs. We highlighted the prohibitive overpotential

Figure 4

Schematic diagrams of the (a) AEMFC anode catalyst layer; and (b) a zoom-in into the triple-phase boundary of HOR, showing the differences between acidic and alkaline fuel cell environments.

of standard electrocatalysts towards HOR, the need to have two descriptors to achieve a better understanding of this reaction, and the importance and challenge of water in the AEMFC anodes. It is crucial to understand all these relations in order to develop highly active HOR PGM-free electrocatalysts for AEMFCs. In particular, increasing the understanding of the mechanism of reaction as well as the relationship between the HOR current density and the descriptors HBE and OHBE, will be of significant help in solving this major barrier in the technology, aiming for cells based completely on precious metal-free AEMFC electrodes.

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