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Dario G Dekel

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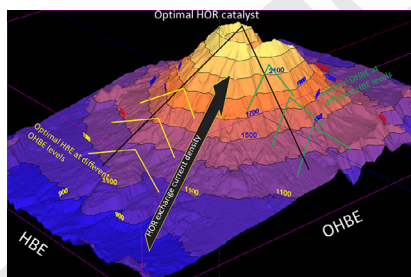
Graphical Abstract

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Unraveling mysteries of hydrogen electrooxidation in anion exchange membrane fuel cells

Dario R. Dekel*

The Wolfson Department of Chemical Engineering, The Nancy & Stephan Grand Technion Energy Program, Technion – Israel Institute of Technology, Haifa 3200003, Israel





ELSEVIER

Review Article

Unraveling mysteries of hydrogen electrooxidation in anion exchange membrane fuel cells

Dario R. Dekel*

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

Address

14 The Wolfson Department of Chemical Engineering, The Nancy &
15 Stephan Grand Technion Energy Program, Technion – Israel Institute of
16 Technology, Haifa 3200003, Israel

17 *Corresponding author: Dekel, Dario R. (dario@technion.ac.il)

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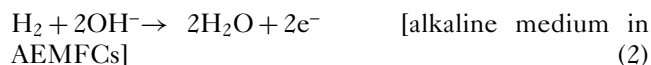
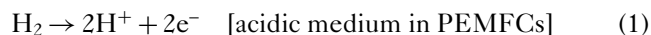
Introduction

18 There is an increasing worldwide interest in anion ex-
19 change membrane fuel cells (AEMFCs), as the technol-
20 ogy promises significant reduction in costs by avoiding
21 the use of platinum as electrocatalysts [1**–4]. Very re-
22 cently, extensive research in hydrogen AEMFCs led to
23 significant progress in cell performance of this technology
24 [5**–10], and to an important increasing in the un-
25 derstanding of the main current challenges – mem-
26 brane degradation/performance stability [11–16*] and
27 CO₂ effect on AEMFC performance [17–24]. This recent
28 progress and the numerous achievements reached in the
29 past years in the technology, suggest that AEMFC can
30 now be considered as a serious alternative to the coun-
31 terpart proton exchange membrane fuel cells (PEMFCs),
32

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 consid-
erably higher, and the way to achieve high anode per-
formance 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

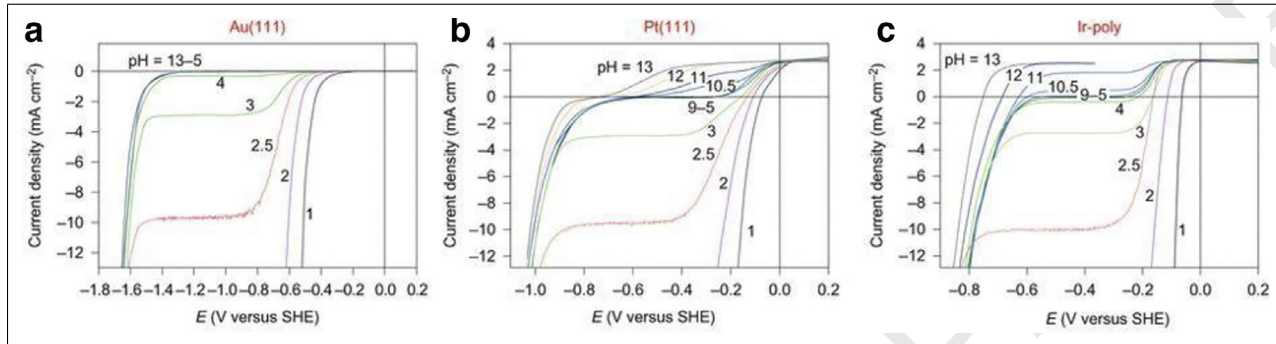
The electrochemical reactions of hydrogen oxidation dif-
fer, depending on the medium, as shown below. In acidic
and alkaline environment, the HOR is given by:



The HOR in the acidic medium (Equation (1)) has
been well studied, as this electrochemical reaction is of
fundamental importance in electrochemistry and electro-
chemical 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 de-
tails 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 au-
thors also focused on the kinetics and the reaction mech-
anisms 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**].

71 studies have shown that the kinetics of the HOR in alkaline media is much slower than in acidic solutions [36,37*].
 72 For instance, the HOR activity of carbon-supported noble metals (Pt, Pd and Ir) decreases by around two
 73 orders of magnitude when transitioning from low to high pH [38]. The HOR kinetics changes with the pH of
 74 the medium were clearly shown by Strmcnik *et al.* [39**].
 75 The authors presented a series of experimental measured polarization curves for the HOR on different catalytic
 76 materials in a wide range of pH ($1 < \text{pH} < 13$), as shown in Figure 1. Clear differences between the measured
 77 current densities at pH < 3 and pH > 11 can be seen.
 78

83 The authors also performed a simulation of the polarization curves using a simple set of electrochemical
 84 equations, confirming the experimental results shown in Figure 1. Results revealed that in strong acidic medium
 85 (pH < 3, corresponding to acidic environment of PEMFCs) the HOR proceeds through the direct oxidation of
 86 hydrogen to H^+ (Equation (1)); while in strong alkaline medium (pH > 13, corresponding to acidic environment
 87 of AEMFCs), pure diffusion-limiting currents are also observed. As mentioned by the authors, this suggests that in
 88 alkaline medium, the supply for both hydrogen and hydroxide anions (both reactants in the HOR in AEMFCs,
 89 see Equation (2)) are sufficient to maximize the electrochemical oxidation of hydrogen to water. We can then also
 90 say that as the pH in the environment is increased, an increase in the diffusion-limiting currents is obtained. In
 91 other words, as the OH^- concentration in the fuel cell is higher, purer diffusion-limiting currents will be observed,
 92 indicating then that the OH^- plays an important role in the HOR for AEMFCs. What is the best way to quantify
 93 this role of the OH^- is not yet clear.
 94

104 Is HBE descriptor good enough to evaluate 105 HOR catalyst for AEMFCs?

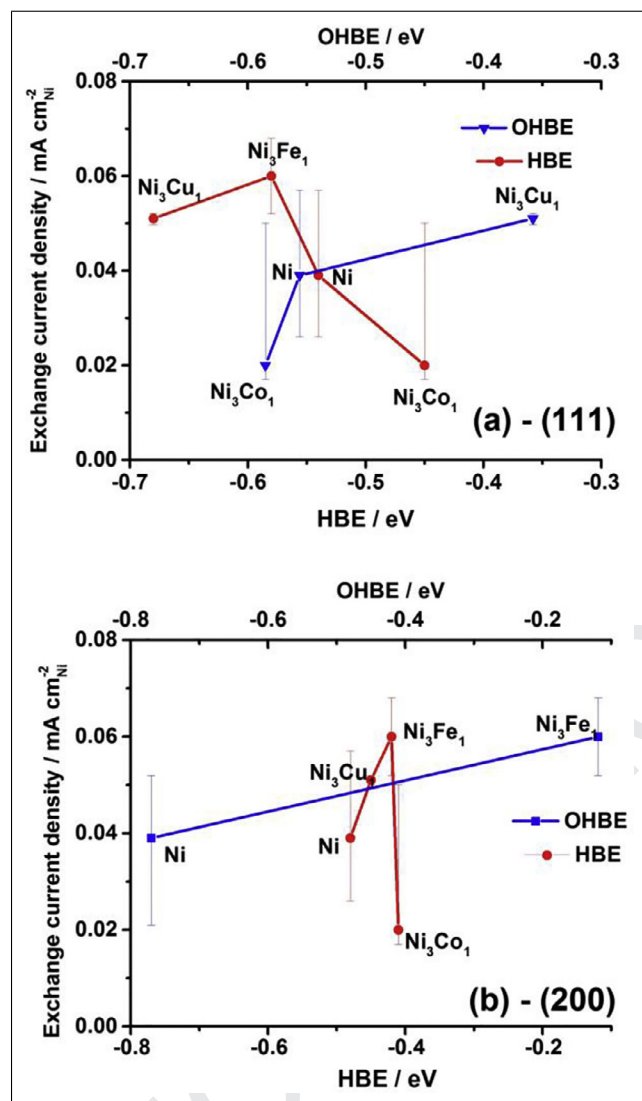
106 As shown in Equation (2), in AEMFCs the electrochemical oxidation of hydrogen involves two reactants –
 107

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 predicted 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.

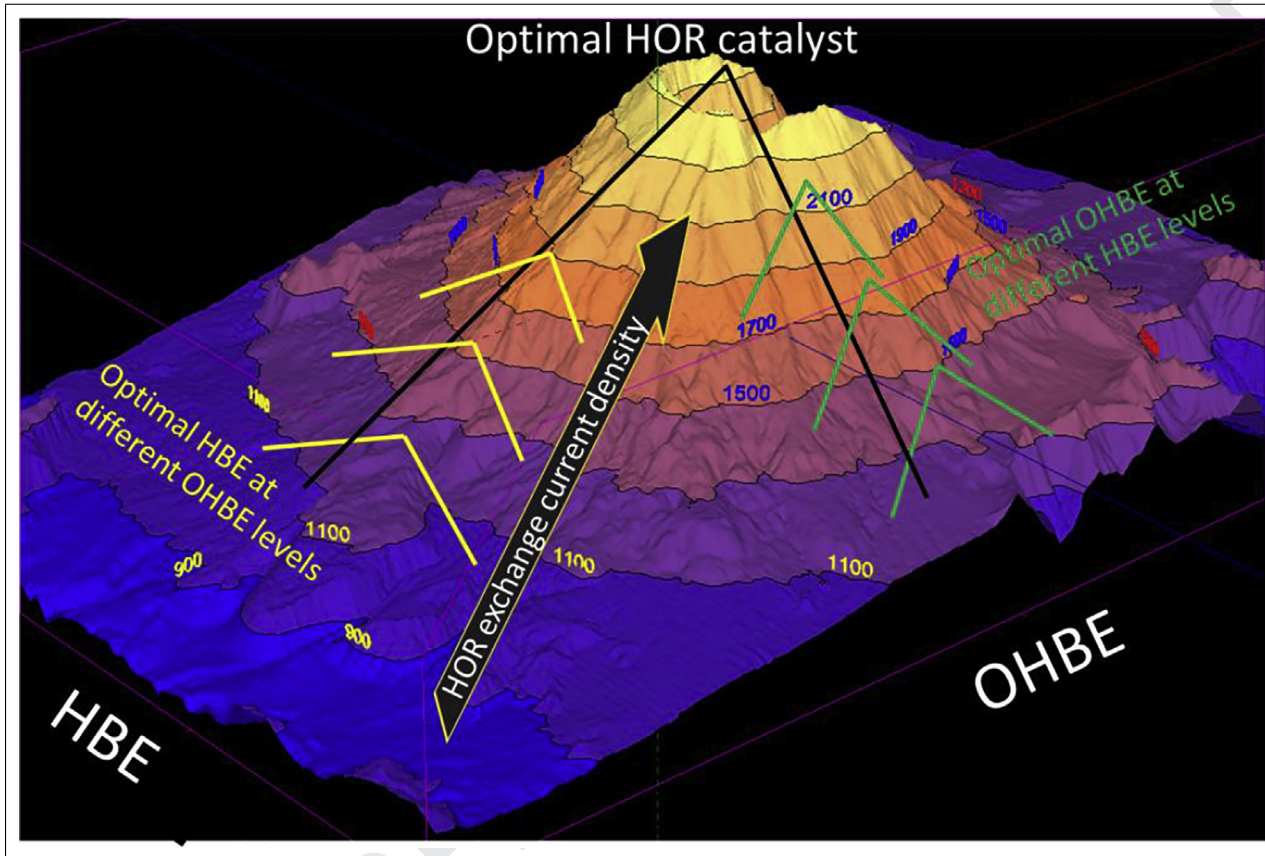
Ideally, to find the best HOR electrocatalysts for AEMFCs we should then evaluate both HBE and OHBE and correlate the interaction between them and the exchange current densities. This ideal situation is schematically illustrated by a ‘volcano plot’, where both HBE and OHBE descriptors are simultaneously 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.

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

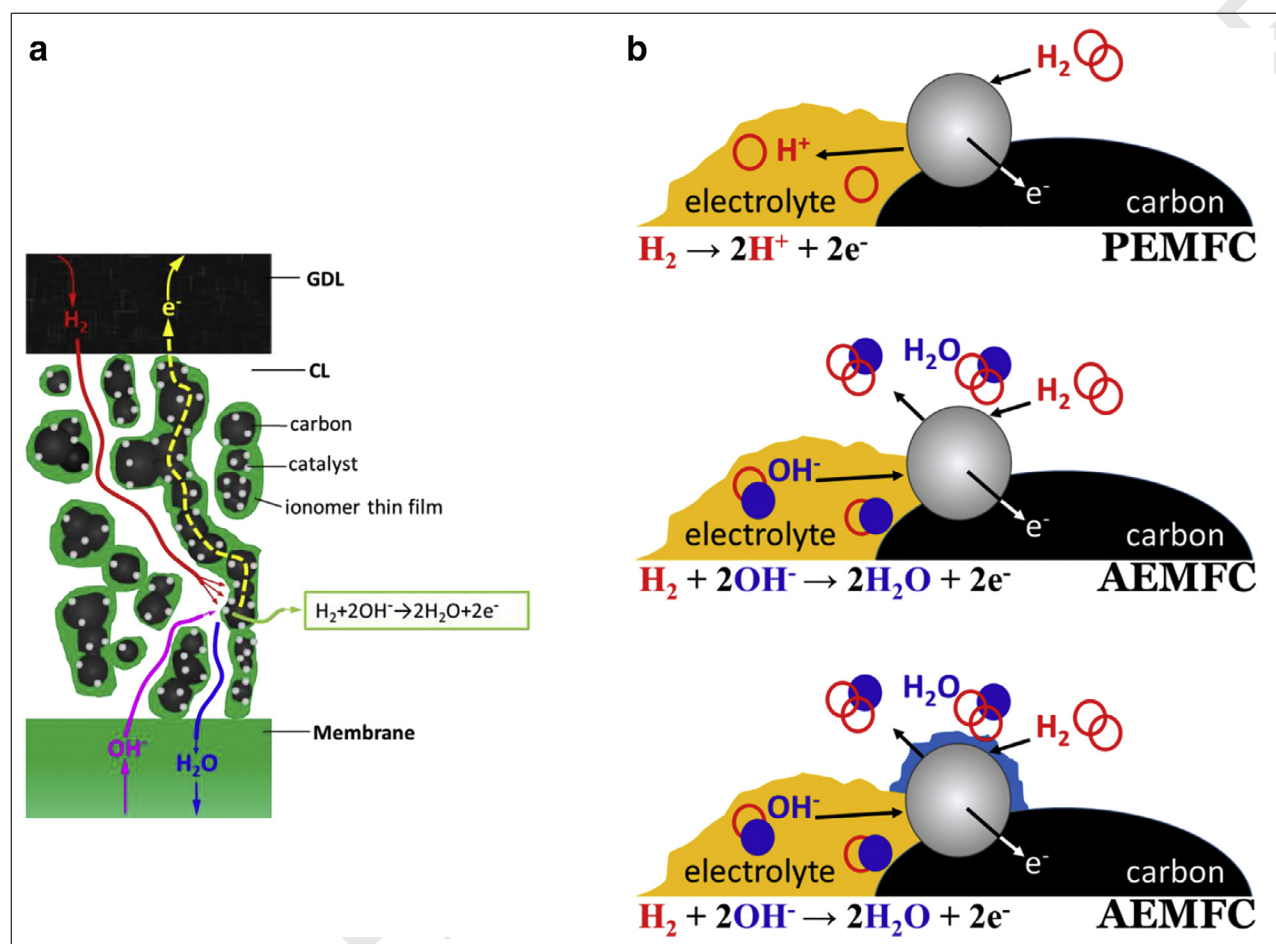
212 Figure 4(b) shows a schematic comparison of the triple-phase boundary (TPB) for the electrooxidation of hydrogen for both acidic and alkaline fuel cell environments. As mentioned above, for AEMFCs the supply of hydrogen as well as the supply of OH^- reactants to the TPB are critical for the HOR. 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

230 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

242 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.

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

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