Three dimensional carbon substrate materials for electrolysis of water

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ABSTRACT Water splitting is an important approach for energy conversion to obtain hydrogen and oxygen. Apart from solar water splitting, electrochemical method plays a key role in the booming field, and it is urgent to develop novel and efficient catalysts to accelerate water splitting reaction. Recently, newly emerging self-supported materials, especially three dimensional (3D) carbon substrate electrochemical catalysts, have attracted great attention benefiting from their fantastic catalytic performances, such as large surface area, enhanced conductivity, tunable porosity, and so on. This review summarizes the outstanding materials used for hydrogen evolution reaction and oxygen evolution reaction. And catalysts that acted as both anode and cathode in two-electrode systems for overall water splitting are introduced systematically. The opportunities and challenges of 3D carbon substrate materials for electrochemical water splitting are proposed.

Keywords: carbon substrate materials, hydrogen evolution reaction, oxygen evolution reaction, water splitting, electrocatalysis, energy conversion

INTRODUCTION

The renewable clean energy is highly demanded for the gradual consumption of fossil fuels, while the sustainable clean energy including solar, wind and tidal is intermittent and unpredictable. It is essential to develop the technology for energy conversion and storage, and the relative research has attracted much attention for decades and is still the focus [1–6].

As compressed gas energy storage, hydrogen, with high energy density and carbon-free production, has demonstrated its potential for sustainable energy economy [7,8]. Water splitting plays a key role to generate hydrogen, which can be obtained in a photochemical or electro-

chemical way. Solar water splitting is a hotspot and many researchers make great efforts to explore it [9–18]. However, as a traditional method to produce hydrogen, electrolysis of water is still flourishing with the development of various excellent catalysts [19–28].

Non-noble metal catalysts demonstrate high efficiency for hydrogen evolution reaction (HER), oxygen evolution reaction (OER) as well as overall water splitting. They are more favorable than traditional Pt-based catalysts (for HER), RuO₂ and IrO₂ (for OER) because of the low cost and good stability [29–33]. However, polymeric binders and extra conductive additives are usually needed when they are used under experimental conditions, which may hinder active sites and reduce the catalytic efficiency [34–36].

Recently, self-supported electrocatalysts exhibiting outstanding performance for HER and OER processes with many intrinsic advantages, such as binder-free, large electroactive surface areas, easy electrolyte penetration as well as enhanced long-term stability, have developed rapidly [37–42]. The electroactive species are able to grow *in situ* on different substrate to form diverse three dimensional (3D) materials *via* various synthetic strategies, such as hydrothermal processing [43], electrochemical deposition [44], solid-state thermolysis [45], and thermal evaporation [46]. The prepared 3D carbon substrate materials not only have the above merits, but also show cost-effectiveness, high conductivity and resistance to corrosion. There is no doubt that they are the shining star for water splitting at present and in the future. Other self-supported materials such as free-standing films [47,48], 3D carbon-free materials supported on nickel foam [49–54], copper foam [55], Ti mesh [56,57], as well as stainless steel [58], which can also be used as electrocatalysts for water splitting.

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This review summarizes the recent progress of 3D carbon substrate materials serving as catalysts for water splitting. Carbon substrate materials include carbon cloth (CC), carbon fibers (CFs), carbon nanotubes, graphene, metal-organic frameworks (MOF)-derived porous carbon. The electrocatalysts for HER and OER, before exploring the catalysts used as anode and cathode simultaneously in a two-electrode system for overall water splitting are also involved. Finally, the challenge and the development trend in this field are proposed.

HYDROGEN EVOLUTION REACTION

HERs go well in acidic electrolytes that can afford enough H^+ in the solution. Thus, non-noble metal carbide, sulfide and phosphide with high stability in strong acid are selected as catalysts supported on carbon matrix for HER. Graphene, with large surface area and excellent electrical conductivity, is a versatile supporting material for HER. For example, 3D hierarchical MoS_2 nanosheets self-assembled on graphene oxide (MoS_2/G) frameworks were synthesized *via* a simple one-step hydrothermal process [43]. The 3D frameworks exhibited efficient and stable electrocatalytic activity when MoS_2/G was used directly as HER electrodes without binder. Besides the effective charge transport in 3D graphene frameworks, the continuously interconnected pores with abundant nucleation sites made the MoS_2/G exhibit excellent stability and high activity for HER. Furthermore, 2D hydrogenated graphene (HG) can also be introduced into ultrathin and vertical MoS_2 nanosheets (MoS_2/HG) for the preparation of highly efficient 3D catalyst for HER [59]. The catalyst had more active sites due to the vertical MoS_2 nanosheets supported on the HG. In 0.5 mol L^{-1} H_2SO_4 solution, the optimized MoS_2/HG hybrid catalyst produced a current density of 10 mA cm^{-2} with a Tafel slope of 41.0 mV dec^{-1} . And there was no observable degradation for 24 h when operated at 30 mA cm^{-2} . Density functional theory (DFT) calculations unveiled the high performance of the MoS_2/HG catalyst for HER. The electronic structure of MoS_2 was modulated *via* the incorporated HG, and the numbers of active sites were increased and hydrogen adsorption energy at S-edge atoms was optimized simultaneously.

In addition to graphene, other carbon materials, such as carbon nanotubes (CNT), nanoporous carbon (NPC), ordered mesoporous carbon (OMC), have also been used as support materials for electrochemical reduction of water. For instance, Sun *et al.* [60] prepared a 3D flexible materials with MoS_2 and CNF (MoS_2/CNF) for HER electrode in acidic electrolyte, which exhibited excellent

catalytic activity. The overpotential was 230 mV when the current density was 16 mA cm^{-2} . This novel HER catalyst had an onset overpotential of 120 mV, a small Tafel slope (44 mV dec^{-1}), exchange current density (0.09 mA cm^{-2}), and a high Faradaic efficiency (nearly 100%). This work offers an attractive method to prepare flexible electrocatalyst for water splitting.

Moreover, different porous carbon materials have been synthesized by using MOF serving as precursor and template. In particular, MOF-derived 3D-NPC has been successfully obtained. For example, Yang *et al.* [61] constructed a 3D architecture *via* the confined growth of MoS_2 nanosheets in 3D-NPC. Benefiting from the unique structure, the prepared 3D materials presented enhanced conductivity as well as more exposed active sites and remarkable electrocatalytic activity for HER, which achieved a current density of 10 mA cm^{-2} with the overpotential of 210 mV. The onset overpotential was 160 mV and the Tafel slope was 51.0 mV dec^{-1} . The *in situ* confined growth in MOF provides a new approach to construct high-performance catalysts.

Apart from MoS_2 , molybdenum carbide is another attractive candidate for water reduction. Recently, Chen *et al.* [62] reported highly dispersed β - Mo_2C nanoparticles ($\sim 5\text{ nm}$) embedded in OMC ($Mo_2C@OMC$) for HER in both acidic and alkaline solution, as displayed in Fig. 1. The Mo_2C possesses approximately 26.5% active sites exposed, which facilitates the electrocatalytic activity. The optimized $Mo_2C@OMC$ delivers a current density of 10 mA cm^{-2} with overpotential of 160 mV in 0.5 mol L^{-1} H_2SO_4 and 175 mV in 1.0 mol L^{-1} NaOH. Besides, bio-carbon-fiber cloth decorated with Mo_2C (BCF/ Mo_2C), prepared by Wang *et al.* [63], shows an outstanding electrocatalytic performance for HER. It requires an overpotential of 115 and 88 mV to achieve 20 mA cm^{-2} in acidic and alkaline media, respectively.

Another 3D carbon substrate material used in both acidic and alkaline solutions was NiP_2 NS/CC, where NiP_2 nanosheet arrays are supported on CC [64]. The NiP_2 NS/CC cathode showed strong durability and attained 10 mA cm^{-2} with the overpotential of 75 mV in acidic electrolytes and 102 mV in alkaline solutions, respectively. It is worth noting that the NiP_2 NS/CC using Nafion was also active for HER, but the corresponding overpotential was much larger than that of NiP_2 NS/CC, because the binder-free 3D architecture of NiP_2 NS/CC improved the conductivity and exposed more active sites. Furthermore, Ni nanoparticles and reduced graphene oxide (rGO) electrodeposited on 3D nickel foam were obtained [65]. The catalyst exhibited excellent activity for

Figure 1 Synthetic procedures for Mo₂C@OMC nanocomposites. Reprinted with permission from Ref. [62]. Copyright 2018, the American Chemical Society.

Figure 2 Synthetic route for Co-C-N catalyst. Reprinted with permission from Ref. [66]. Copyright 2015, the American Chemical Society.

HER in alkaline medium with a low overpotential of only 36 mV to drive 10 mA cm⁻². This remarkable HER performance can be attributed to its excellent electrical conductivity, large surface area and superior adsorption/desorption of H⁺ (H⁺ represents H₃O⁺ gaining an electron) of Ni-rGO composite simulated by DFT calculations.

All the above mentioned carbon substrate 3D materials are used as electrodes for HER in acidic and/or basic medium; however pH-universal electrocatalysts is highly desirable for practical applications. Zhang *et al.* [66] reported a self-supported and 3D porous Co-C-N complex bonded carbon (only 0.22 at% Co), which exhibited catalytic activity with overpotential of 138 mV at 10 mA cm⁻² and the Tafel slope of 55 mV dec⁻¹ in 0.5 mol L⁻¹ H₂SO₄. Moreover, the catalysts can also be used in neutral and basic conditions for HER with 100% Faradaic efficiency. This work provides an excellent synthetic approach to prepare catalysts with little metal for electrochemical devices (Fig. 2). Recently, Shen *et al.*

[67] developed an active and robust HER catalyst, where vanadium-doped CoP grew on self-supported conductive and flexible CC (V-CoP/CC) *via* hydrothermal-phosphorization method. The V-CoP/CC electrode achieved a ultralow overpotential of 71.0 mV at 10 mA cm⁻² (in 1.0 mol L⁻¹ KOH), 123 mV (in 1.0 mol L⁻¹ PBS) and 47 mV (in 0.5 mol L⁻¹ H₂SO₄). This cost-effective material with excellent electrochemical performance broadens our vision to prepare non-noble metal catalysts for the important reactions in energy conversion and storage. And the representative 3D carbon substrate materials for HER are summarized in Table 1.

OXYGEN EVOLUTION REACTION

OER is kinetically sluggish with a high overpotential above its thermodynamic potential (1.23 V), leading to a low conversion efficiency of the overall water splitting process. OER catalysts are used to overcome such a barrier. Iron, cobalt, nickel and their corresponding alloys, oxide, selenide, sulfide, phosphide are usually used as

Table 1 Representative 3D carbon substrate materials for HER

Catalyst	Substrate ^a	Current density (mA cm ⁻²)	Overpotential (mV)	Solution	Ref.
MoS ₂	HG	10	41	0.5 mol L ⁻¹ H ₂ SO ₄	[59]
MoS ₂	CNF	16	230	0.5 mol L ⁻¹ H ₂ SO ₄	[60]
MoS ₂	NPC	10	210	0.5 mol L ⁻¹ H ₂ SO ₄	[61]
Mo ₂ C	OMC	10	160	0.5 mol L ⁻¹ H ₂ SO ₄	[62]
Mo ₂ C	OMC	10	175	1.0 mol L ⁻¹ NaOH	[62]
Mo ₂ C	BCF	20	115	0.5 mol L ⁻¹ H ₂ SO ₄	[63]
Mo ₂ C	BCF	20	88	1.0 mol L ⁻¹ KOH	[63]
NiP ₂	CC	10	75	0.5 mol L ⁻¹ H ₂ SO ₄	[64]
NiP ₂	CC	10	102	1.0 mol L ⁻¹ NaOH	[64]
Co-N-C	CP	10	138	0.5 mol L ⁻¹ H ₂ SO ₄	[66]
V-CoP	CC	10	47	0.5 mol L ⁻¹ H ₂ SO ₄	[67]
V-CoP	CC	10	123	1.0 mol L ⁻¹ PBS	[67]
V-CoP	CC	10	71	1.0 mol L ⁻¹ KOH	[67]

a) BCF represents biocarbon fiber and CP represents carbon paper.

catalysts for OER. They exhibit excellent electrocatalytic activity when supported on carbon substrate. For example, Sun *et al.* [68] explored the OER activity of hematite nanorods array anchored on CC (α -Fe₂O₃ NA/CC). It required an overpotential of 420 mV to attain the current density of 10 mA cm⁻². Besides, Fe-Ni₂P/CC was also used as OER catalysts in alkaline electrolyte. It demands an overpotential of only 215 mV to drive 50 mA cm⁻² [69].

It is highly desired to fabricate water oxidation catalysts with efficient activity under benign conditions. Sun *et al.* [70] reported a high-efficiency and durable iron-based material for OER in mild pH solutions based on oxidative polarization of FeP nanoarrays on CC in potassium borate (KBi). The 3D Fe-Pi-Bi/CC used as electrode for water oxidation exhibited high activity and strong electrochemical durability. To achieve a current density of 10 mA cm⁻², it demanded an overpotential of 434 mV in 0.1 mol L⁻¹ KBi and maintained its activity for over 20 h. This work not only offers an attractive low-cost catalytic 3D material for OER under benign conditions, but also provides us a universal method to prepare non-noble metal-based 3D catalysts for electrochemical application.

Besides, amorphous NiFe(oxy)hydroxide nanosheets electrodeposited on 3D partially exfoliated graphite (EG) foil *via* a facile one-step process was proposed by Liu *et al.* [71]. The obtained NiFe/EG composite possessed excellent electric conductivity, facile ion transport, efficient gas (O₂) diffusion due to its unique hierarchical structure (Fig. 3). The optimized electrode exhibited a high OER

efficiency within a wide current density range. It demanded a low overpotential of 214 mV to attain 10 mA cm⁻² in 1.0 mol L⁻¹ KOH. More importantly, the electrode presented superior performance at high current density (500 mA cm⁻²) with the overpotential of 251.0 mV for 48 h. The prominent OER activity of the 3D NiFe/EG material opens up a new way to large-scale practical applications for energy conversion. Furthermore, cobalt-based nanoparticles decorated on 3D porous carbon nanofiber networks (CNF@Co) were prepared [72]. The interconnected porous 3D networks provided abundant channels and interfaces, leading to efficient mass transfer and oxygen evolution. As a result, an overpotential of 314 mV was needed for CNF@Co of OER catalyst to achieve 10 mA cm⁻² in alkaline solution.

Furthermore, MOF-derived carbon materials are also outstanding candidates for the construction of OER catalysts. Our group prepared a free-standing Zn-doped CoSe₂ grown on the MOF-derived carbon fabric collector (CFC) catalyst for OER [73]. Compared with CoSe₂/CFC, the Zn-CoSe₂/CFC manifested an enhanced catalytic activity in 1.0 mol L⁻¹ KOH solution, showing an overpotential of 356 mV at 10 mA cm⁻² with a Tafel slope of 88 mV dec⁻¹. Recently, Dai *et al.* [74] introduced a new MOF-derived Co@N-C bifunctional material for OER and Zn-air batteries. The unique 3D hierarchical catalysts, comprising homogeneously distributed cobalt nanoparticles which was encapsulated by partially graphitized N-doped carbon rings, exhibited excellent electrocatalytic activities for OER. The optimal product showed a po-

Figure 3 Fabrication process for NiFe/EG. Reprinted with permission from Ref. [71]. Copyright 2017, the Royal Society of Chemistry.

Table 2 Representative 3D carbon substrate materials for OER

Catalyst	Substrate ^a	Current density (mA cm ⁻²)	Overpotential (mV)	Solution	Ref.
α -Fe ₂ O ₃	CC	10	420	0.1 mol L ⁻¹ KOH	[68]
Fe-Ni ₂ P	CC	50	215	1.0 mol L ⁻¹ KOH	[69]
Fe-Pi-Bi	CC	10	434	0.1 mol L ⁻¹ KBi	[70]
NiFe	EG	10	214	1.0 mol L ⁻¹ KOH	[71]
Co	CNF	10	314	1.0 mol L ⁻¹ KOH	[72]
Zn-CoSe ₂	CFC	10	356	1.0 mol L ⁻¹ KOH	[73]
Co	N-C	10	350	0.1 mol L ⁻¹ KOH	[74]
CoNi-NCNT	NPCN	10	360	0.1 mol L ⁻¹ KOH	[75]

a) N-C represents N-doped carbon.

tential of 1.58 V (overpotential 350 mV) at the current density of 10 mA cm⁻². Heteroatoms (N, P, S, and so on) doped in carbon skeleton can ameliorate the catalytic activity as well. Chen *et al.* [75] developed a strongly coupled 3D hybrid where N-doped CoNi alloy-encapsulating CNT (CoNi-NCNT) grew *in situ* on N-doped porous carbon nanosheets (NPCN) for OER in alkaline medium. The NPCN/CoNi-NCNT catalyst achieved a current density of 10 mA cm⁻² at 1.59 V (overpotential 360 mV). Wang *et al.* [76] prepared metal-free nitrogen-doped holey graphene nanocapsules (NHGNs) *via* a substrate-induced synthesis method for OER, and its Tafel slope was 158 mV dec⁻¹.

It is worth noting that recently our group synthesized a free-standing nickel foam supported Ni₃S₂@graphene@Co₉S₈ material [77]. The sandwich-structured Ni₃S₂@graphene@Co₉S₈ exhibited an enhanced catalytic performance due to the highly conductive graphene as an intermediary to accelerate the electron transfer. This 3D catalyst presented excellent electrocatalytic activities for OER, obtaining 10 mA cm⁻² with an overpotential of 210 mV. In particular, it manifested predominant durability of over 96 h in 1.0 mol L⁻¹ KOH (14 mA cm⁻² at constant overpotential of 215 mV). Besides, we also designed a free-standing 3D NiO@C nanobelt supported on nickel foam for OER in alkaline solution, achieving 20 mA cm⁻² at overpotential of 368 mV [78]. Some re-

presentative 3D carbon substrate materials for OER are summarized in Table 2.

OVERALL WATER SPLITTING

Compared with the two-half reaction of water splitting, namely HER and OER, the overall water electrolysis is more significant for electrical energy conversion from renewable resources into chemical fuels. For instance, Feng *et al.* [79] reported a ternary EG/Co_{0.85}Se/NiFe-LDH hybrid prepared with two steps. Firstly, cobalt selenide (Co_{0.85}Se) nanosheets were *in situ* grown on EG foil, and then NiFe layered-double-hydroxide (NiFe-LDH) was deposited *via* hydrothermal approach. The obtained 3D hierarchical catalyst possessed excellent HER and OER activity. A current density of 20 mA cm⁻² was achieved at a potential of 1.71 V when this material was used as both cathode and anode in 1.0 mol L⁻¹ KOH for overall water splitting. Supported on the flexible exfoliated graphene (FEG) foil, they later developed another free-standing 3D hybrid Co-N_x|P-GC/FEG with hydrothermal and pyrolysis method [80]. Large specific surface area with plenty of active sites and high conductivity guaranteed the prominent electrocatalytic performance for water splitting. It only required 1.6 V to attain 10 mA cm⁻² in alkaline solution. Moreover, this hybrid integrated onto a hematite electrode can be used as an efficient material for solar-driven water splitting.

Figure 4 (a) Photograph of overall water splitting device on working state. Reprinted with permission from Ref. [82]. Copyright 2017, the American Chemical Society. (b) Photograph of overall water splitting powered by an AAA battery with a nominal voltage of 1.5 V. Reprinted with permission from Ref. [83]. Copyright 2016, the American Chemical Society.

Besides, nickel phosphide (NiP_x) nanoflakes grown on the CF were prepared by Wang *et al.* [81]. As a flexible bifunctional gas-evolving electrode, the CF@NiP_x performed excellent electrocatalytic activity for both HER and OER in alkaline electrolyte owing to the unique porous structure, enhanced conductivity as well as the active Ni^{2+} and $\text{P}^{\delta-}$ with rich valences. With a cell voltage of only 1.61 V (iR uncorrected), the overall water splitting current density of 10 mA cm^{-2} can be achieved. Later, the same group fabricated fibrous Ni@NiO wrapped CF (CF@Ni@NiO) for HER and OER [82]. And the resulting flexible electrodes were used as both anode and cathode for overall water splitting (Fig. 4a). The current density of 20 mA cm^{-2} can be obtained at 1.73 V (iR uncorrected), which provided a facile strategy for preparing bifunctional gas-evolving fibrous electrode. Zhang *et al.* [83] also fabricated an integrated 3D electrode of CP/carbon tubes/cobalt-sulfide sheets (CP/CTs/Co-S) for overall water splitting. With an operating potential of 1.743 V, the electrodes afford a current density of 10 mA cm^{-2} in 1.0 mol L^{-1} KOH using a two-electrode configuration. Moreover, an AAA battery (nominal voltage of 1.5 V) can drive overall water splitting with obvious gas bubble release (Fig. 4b).

MOF-derived materials are versatile for water splitting. Our group developed MOF-derived CoSe_2 nanoparticles

anchored on CF, which manifested efficient and stable catalytic activity for HER and OER in alkaline medium (Fig. 5) [84]. When the electrolyzer was equipped with CoSe_2/CF anode and cathode, the current density of 10 mA cm^{-2} was achieved at a cell potential of 1.63 V for overall water splitting. Besides, Chen *et al.* [85] reported a Co-based hybrid electrocatalyst prepared with MOF template carbonization. Benefiting from the exposed active sites and conductivity, the catalyst can act as both cathode and anode for electrochemical water splitting, to achieve current density of 10 mA cm^{-2} with a voltage of 1.625 V. Moreover, MOF-derived porous Co-N-C flakes developed on CNF grafted with MoS_2 nanosheets were synthesized [86]. The binder-free catalyst exhibited a potential of 1.62 V to generate 10 mA cm^{-2} with remarkable stability during the electrolysis of water. Guo *et al.* [87] reported a 3D hierarchical material composed of FeCo@NC core-shell units which were dispersed on the flower-like architecture of N-doped carbon nanosheets (NCNS). Different ratio of Fe/Co source and calcination temperature were explored. The optimal $\text{Fe}_{0.5}\text{Co}_{0.5}\text{@NC/NCNS}$ required a cell voltage as low as 1.60 V to attain 10 mA cm^{-2} in a two-electrode system for overall water splitting. In addition, the catalytic activities of ultrafine Ni-Fe/Ni-Mo nanoparticles embedded in N-doped carbon were studied by Fan *et al.* [88]. The authors selected NiMo-PVP as cathode and NiFe-PVP as anode in a two-electrode system which delivered a current density of 10 mA cm^{-2} at 1.66 V with a Faradaic efficiency of nearly 100% in alkaline solution. Recently, Li *et al.* [89] reported graphdiyne-supported NiCo_2S_4 nanowires with fantastic electrocatalytic activity. As a highly active 3D bifunctional material, NiCo_2S_4 NW/GDF electrode needed only 1.53 and 1.56 V to perform a current density of 10 and 20 mA cm^{-2} for water splitting in alkaline solution, respectively.

However, the materials mentioned above were all used in alkaline medium for overall water splitting. Catalysts working within wide pH ranges are of great importance.

Figure 5 Synthetic procedures for CoSe_2/CF . Reprinted with permission from Ref. [84]. Copyright 2016, Springer Nature.

Figure 6 Overall water splitting electrolyzer powered by two Zn-air batteries (left) and the evolution of hydrogen and oxygen bubbles (right). Reprinted with permission from Ref. [92]. Copyright 2017, the Royal Society of Chemistry.

Naldoni *et al.* [90] prepared 3D flexible electrodes based on Al-doped Ni(OH)₂ wrapped with a CNT forest (CNT-F). This material not only showed an excellent OER activity under alkaline conditions, but also can be used as anode and cathode simultaneously for overall water splitting in 1.0 mol L⁻¹ KOH and 0.5 mol L⁻¹ H₂SO₄. It required a relatively high cell voltage of 1.96 and 2.04 V to generate 10 mA cm⁻² in alkaline and acidic solution, respectively. Moreover, Xu and Luque [91] designed the first metal-free 3D porous carbonaceous materials for overall water splitting *via* a scalable methodology and this unprecedented binder-free electrode was prepared by O, N and P tri-doped porous graphite nanocarbon (ONPPGC) directly grown on oxidized CC (OCC). ONPPGC/OCC offered outstanding catalytic performance and durability in pH-universal electrolyte. It needed 1.66 V to drive a current density of 10 mA cm⁻² in

1.0 mol L⁻¹ KOH solution. Recently, Wang *et al.* [92] reported a defect-rich porous carbon (DRPC), bifunctional metal-free water splitting electrocatalyst. The resultant catalysts displayed superb catalytic activities for HER and OER in alkaline solution and delivered a 10 mA cm⁻² overall water splitting current density with voltage of 1.74 V. Moreover, the electrochemical water splitting system was powered by Zn-air battery and the air electrode was made from the same materials (DRPC) for oxygen reduction reaction (Fig. 6). Representative 3D carbon substrate materials for overall water splitting are summarized in Table 3.

CONCLUSION

In this review, we have summarized the prominent 3D carbon substrate materials composed of earth-abundant elements for water splitting. Most of carbon materials and non-noble metal compounds are used as substrate and active substance, respectively. Although 3D metal-free carbon substrate have been reported, yet there needs to be further explored. Self-supported, binder-free as well as flexible catalysts are the development trend. Neutral media will be a promising electrolyte for HER and OER. It is very significant to develop pH-universal catalysts for overall water splitting.

Besides, the catalytic mechanism is not discussed here, but it is of great importance to employ computational simulation methods to elucidate. Moreover, it will be amazing if we can design the nanostructures of self-

Table 3 Representative 3D carbon substrate materials for overall water splitting

Catalyst	Substrate ^a	Current density (mA cm ⁻²)	Potential (V)	Solution	Ref.
Co _{0.85} Se/NiFe-LDH	EG	20	1.71	1.0 mol L ⁻¹ KOH	[79]
Co-N _x P-GC	FEG	10	1.60	1.0 mol L ⁻¹ KOH	[80]
NiP _x	CF	10	1.61 ^b	1.0 mol L ⁻¹ KOH	[81]
Ni@NiO	CF	20	1.73 ^b	1.0 mol L ⁻¹ KOH	[82]
Co-S	CP/CTs	10	1.743	1.0 mol L ⁻¹ KOH	[83]
CoSe ₂	CF	10	1.63	1.0 mol L ⁻¹ KOH	[84]
Co-NC	CNT	10	1.625	1.0 mol L ⁻¹ KOH	[85]
CoNC@MoS ₂	CNF	10	1.62	1.0 mol L ⁻¹ KOH	[86]
FeCo@NC	NCNS	10	1.6	1.0 mol L ⁻¹ KOH	[87]
NiCo ₂ S ₄	GDF	10	1.53	1.0 mol L ⁻¹ KOH	[89]
NiAl	CNT-F	10	1.96	1.0 mol L ⁻¹ KOH	[90]
NiAl	CNT-F	10	2.04	0.5 mol L ⁻¹ H ₂ SO ₄	[90]
ONPPGC	OCC	10	1.66	1.0 mol L ⁻¹ KOH	[91]
DRPC	DRPC	10	1.74	1.0 mol L ⁻¹ KOH	[92]

a) CNT-F represents carbon nanotube forest. b) *iR* uncorrected.

supported 3D materials under the guidance of computer and accomplish it through 3D-printing technology. However, there is still challenging to transfer the laboratory-scale research into industry. Even so, this booming field will continue to receive extensive attention in the future.

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Conflict of interest The authors declare no conflict of interest.

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三维碳基材料在电解水中的应用

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摘要 水分解制备氢气和氧气是能源转换的一种重要方法, 除光解水之外, 电解水也一直备受关注. 电解水过程中, 使用新型高效催化剂可以加快水分解并降低成本. 最近, 自支撑三维碳基材料由于其比表面积大、导电性强、耐酸碱性好等优点在电催化水分解领域引起研究者的广泛兴趣. 本文总结了近年来三维碳基材料在析氢、析氧领域的研究现状, 重点介绍了其在双电极体系中分解水同时制氢气和氧气的研究进展, 指出了三维碳基材料在电解水领域存在的问题和未来的发展方向.