Carbon-Based Electrocatalysts Derived From Biomass for Oxygen Reduction Reaction: A Minireview

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Oxygen reduction reaction (ORR) electrocatalysts derived from biomass have become one of the research focuses in hetero-catalysis due to their low cost, high performance, and reproducibility properties. Related researches are of great significance for the development of next-generation fuel cells and metal-air batteries. Herein, the preparation methods of various biomass-derived catalysts and their performance in alkaline, neutral, and acidic media are summarized. This review clarifies the research progress of biomass carbon-based electrocatalysts for ORR in acidic, alkaline and neutral media, and discusses the future development trends. This minireview can give us an important enlightenment to practical application in the future.

Keywords: oxygen reduction reaction, biomass-derived, electrocatalyst, fuel cells, hetero-catalysis

INTRODUCTION

The development of green energy has been urgent due to the increasingly international attention toward energy shortages and environmental pollutions. Among these energy storage devices, fuel cells have been considered as a promising alternative with clean, stable, and sustainable properties in order to meet the growing global energy demands (Dai et al., 2015). Therefore, rational design of low-cost oxygen reduction electrocatalysts is critical for the storage and electrochemical performance of renewable energy sources (Liu et al., 2015). As an ideal component of primary energy equipment, fuel cells using hydrogen or hydrocarbon fuels can directly convert chemical fuel into electricity through electrochemical processes and operate at ambient temperature (Winter and Brodd, 2004). Even though the amount of Pt is capable of achieving the desired catalytic effect by using Pt alloys (Stamenkovic et al., 2007; Jiang et al., 2009) or making core-shell nanostructures with supporting materials (Li et al., 2018), the high cost, insufficient durability, and unrefined technology still restrict the practical large-scale commercialization (Guo et al., 2013; Kaur et al., 2019).

Accordingly, to address these above issues, numerous non-Pt materials have been studied as cathode catalysts alternative to Pt-based catalysts for ORR (Banham et al., 2015; Shao et al., 2016). Cuurently, biomass-derived materials, such as active carbon (Deng et al., 2010), enzyme (Qiao et al., 2010), microorganism (Majidi et al., 2019; Papiya et al., 2019), transition metal porphyrins (Zheng et al., 2016), NiIn2S4/CNFs (Fu et al., 2019), and phthalocyanines (Kaare et al., 2016; Bhowmick et al., 2019) have potential capability to replace Pt. Therefore, the method of producing ORR catalyst from biomass has attracted extensive attention of researchers in many aspects (Liu et al., 2016; Sawant et al., 2016; Zhao et al., 2017). In this minireview, the catalytic mechanism for oxygen reduction reaction in different media is given, including a four-electron pathway and
a two-electron pathway. Thereafter, various biomass-derived carbons and composites for ORR in alkaline, neutral, and acidic media are summarized (Figure 1). The aforementioned biomass-derived carbons and composites exhibit outstanding electrochemical performance, which make them promising candidates for alternating Pt-based electrocatalysts. Moreover, we discuss the future research directions and challenges of biomass-derived carbon electrocatalyst for ORR in different media.

**CATALYTIC MECHANISM FOR OXYGEN REDUCTION REACTION**

Generally, the oxygen reduction reaction in an aqueous electrolyte can proceed via two types: a four-electron pathway and a two-electron pathway. The former method could directly reduce oxygen to water, which is preferable than the two-electron route using hydrogen peroxide as a reaction intermediate. The choice of overall pathway depends on the type of catalyst. So far, many literatures have reported the use of biomass and its derivatives as ORR catalysts in neutral or alkaline medium, the reaction mechanism can be described as follows:

Four-electron pathway:

\[
O_2 + 2H_2O + 4e^- \rightarrow 4OH^- \quad E = 0.401 \text{ V}
\]

Two-electron pathway:

\[
\begin{align*}
O_2 + H_2O + 2e^- & \rightarrow HO_2^- + OH^- \quad E = -0.065 \text{ V} \\
HO_2^- + H_2O + 2e^- & \rightarrow 3OH^- \quad E = 0.867 \text{ V} \\
2HO_2^- & \rightarrow 2OH^- + O_2
\end{align*}
\]

The accumulation of OH\(^-\) at the catalytic sites can lead to a considerable decline in the kinetic performance (Popat et al., 2012).

In an acidic medium, the mechanism can be described as follows:

Four-electron pathway:

\[
O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \quad E = 1.229 \text{ V}
\]

Two-electron pathway:

\[
\begin{align*}
O_2 + 2H^+ + 2e^- & \rightarrow H_2O_2 \quad E = 0.695 \text{ V} \\
H_2O_2 + 2H^+ + 2e^- & \rightarrow 2H_2O \quad E = 1.770 \text{ V} \\
2H_2O_2 & \rightarrow 2H_2O + O_2
\end{align*}
\]

**IN ALKALINE MEDIUM**

Catalysts with excellent ORR performance in an alkaline medium will play an important role in metal-air batteries. Currently, biomass-derived carbon electrocatalysts for ORR have been reported to be the most effective in alkaline medium. As illustrated in Figure S1A, natural cattail fibers are used to prepare porous nitrogen-doped carbon through direct chemical activation and heteroatom doping (Liu et al., 2019). The obtained graphene-like sheets from biomass pyrolysis are assembled into three-dimensional carbon frameworks, which exhibit a significant synergistic effect on the improvement of catalytic properties. Fu et al. successfully prepared NiIn\(_2\)S\(_4\) nanosheets supported on carbon nanofibers (Figure S1B). It was found that the performance of prepared catalyst is better than that of single metal Ni or In sulfides (Fu et al., 2019). Corn stover was also reported as a biomass precursor for the preparation of nitrogen, cobalt co-doped electrocatalyst (NCAC-Co) for ORR and aluminum-air batteries (Liu et al., 2018). The resulting porous biocarbon electrocatalyst not only exhibits the 4-electron oxygen reduction mechanism, but also displays excellent durability and stability. The author clearly demonstrated that the NCAC-Co has good prospects and is expected to become an economical and large-scale catalyst substitute for metal-air batteries. As illustrated in Figure S1C, the NCAC-Co electrocatalyst was prepared by two major steps of KOH activation and heteroatom doping. In addition to the examples mentioned above, other promising biomass precursors are dandelion seeds (Tang et al., 2019), shaddock peel (Lu et al., 2019), chitosan (Zhao et al., 2017), mulberry leaves (He et al., 2019), gelatin (Yang et al., 2019), and chitin (Wang et al., 2019) etc.

**IN NEUTRAL MEDIUM**

As the research on microbial fuel cells becomes more and more in-depth, the development of new high-performance biomass carbon materials has become increasingly crucial. Compared with the alkaline medium, biomass catalysts have fewer applications in neutral media because the ORR performances were slightly negative. However, biomass-derived electrocatalysts have a higher stability in a neutral medium. For instance, the sewage sludge-derived biochar was successfully prepared and employed as an excellent ORR electrocatalyst. As shown in
TABLE 1 | Typical examples of electrocatalysts derived from biomass.

| Electrolyte | Biomass precursor | Optimal preparation temperature | Specific surface area | Onset Potential of Catalyst/Pt/C |
|-------------|-------------------|---------------------------------|----------------------|----------------------------------|
| 0.1 M KOH   | Chitosan (Zhao et al., 2017) | 800°C                           | 543 m² g⁻¹           | −0.08/−0.055 V (vs. Ag/AgCl)     |
| 0.1 M KOH   | Corn Stovers (Liu et al., 2018) | 900°C                           | 1877.3 m² g⁻¹        | /                                |
| 0.1 M KOH   | Dandelion Seed (Tang et al., 2019) | 900°C                           | 1324.1 m² g⁻¹       | 0.83/0.85 V (vs. RHE)            |
| 0.1 M KOH   | Chrysanthemum Flowers (Xu et al., 2017) | 800°C                           | 810 m² g⁻¹          | 1.0/1.0 V (vs. RHE)              |
| 0.1 M KOH   | Malachium Aquaticum (Huang et al., 2016) | 900°C                           | 851.41 m² g⁻¹       | −0.053/−0.043 V (vs. Ag/AgCl)    |
| 0.1 M KOH   | Microalgae (Wu et al., 2019) | 1,000°C                          | /                    | 1.0/1.0 V (vs. RHE)              |
| 0.1 M KOH   | Lotus Root (Fajendran et al., 2019) | 700°C                           | 884 m² g⁻¹          | 0.84/0.92 V (vs. RHE)            |
| 0.1 M KOH   | Shaddock Peel (Lu et al., 2019) | 900°C                           | 548 m² g⁻¹          | /                                |
| 0.1 M KOH   | Coconut Shells (Borghei et al., 2017) | 1,000°C                          | 1260 m² g⁻¹        | −0.02/0.05 V (vs. Ag/AgCl)       |
| 0.1 M KOH   | Nιm2Sn/CNFs (Fu et al., 2019) | /                               | 196.3 m² g⁻¹        | 1.46/1.50 V (vs. RHE)            |
| 0.1 M KOH   | Cattail Fibers (Liu et al., 2019) | 900°C                           | 1773 m² g⁻¹         | 0.92/0.45 V (vs. RHE)            |
| 0.1 M KOH   | Mulberry Leaves (He et al., 2019) | 800°C                           | 1689 m² g⁻¹         | 0.86/0.88 V (vs. RHE)            |
| 0.5 M KOH   | Waste Leather (Alonso-Lemus et al., 2016) | /                               | 2100 m² g⁻¹        | 0.905/1.050 V (vs. RHE)          |
| 0.1 M KOH   | NCAC-Co (Liu et al., 2018) | 800°C                           | 1877.3 m² g⁻¹       | 0.795/0.760 V (vs. RHE)          |
| Phosphate Buffer | Corncob (Li et al., 2018) | 650°C                           | 655.89 m² g⁻¹       | −0.13/−0.05 V (vs. Ag/AgCl)      |
| Phosphate Buffer | Sewage Sludge (Yuan et al., 2013) | 900°C                           | 44 m² g⁻¹           | 0.11/0.09 V (vs. RHE)            |
| Phosphate Buffer | Egg (Lu et al., 2017) | 900°C                           | 703.47 m² g⁻¹       | 0.257/0.157 V (vs. Ag/AgCl)      |
| Phosphate Buffer | Bacteria (Ma et al., 2017) | 800°C                           | 1926.7 m² g⁻¹       | 1.01/1.01 V (vs. RHE)            |
| 0.1 M KOH   | Bagasse (Yuan et al., 2016) | 1,000°C                          | 1284 m² g⁻¹         | 1285/0.06 V (vs. Hg/HgO)         |
| 0.5 M H₂SO₄ | Enoki Mushroom (Guo et al., 2015) | 900°C                           | 305.3 m² g⁻¹        | 0.43/0.65 V (vs. Ag/AgCl)        |
| 0.5 M H₂SO₄ | /                   | /                               | 305.3 m² g⁻¹        | 0.94/0.98 V                      |
| 0.1 M KOH   | Sewage Sludge (Yuan and Dai, 2016) | 800°C                           | 265.05 m² g⁻¹       | 0.05/−0.08 V                     |
| 0.5 M H₂SO₄ | /                   | /                               | 265.05 m² g⁻¹       | 0.05/−0.08 V                     |
| 0.1 M KOH   | Corn Starch (Wang et al., 2012) | 500°C                           | 1568.85 m² g⁻¹      | −0.03/0.02 V                     |
| 0.5 M H₂SO₄ | /                   | /                               | 1568.85 m² g⁻¹      | 0.62/0.66 V (vs. RHE)            |
| 0.1 M KOH   | Starch (Yao et al., 2016) | 180°C                           | 976.6 m² g⁻¹        | 0.955/0.932 V                    |
| 0.5 M H₂SO₄ | /                   | /                               | 976.6 m² g⁻¹        | 0.840/0.930 V (vs. RHE)          |

Figure S2A, the structural change of as-obtained carbonized materials was clearly observed, which was detected by Raman spectroscopy (Yuan et al., 2013). In addition, Lu et al. developed a low-cost method to prepare egg-based heteroatom-doped carbon catalysts. The ORR catalytic activity of prepared electrocatalyst in a neutral medium is comparable to that of a commercially available Pt/C catalyst (Figure S2B, Lu et al., 2017). In the neutral medium, not only the above-mentioned biomass precursors are successfully used, the corn cob-derived catalysts synthesized by a simple pyrolysis method (Li et al., 2018) and mesoporous Fe-NC electrocatalysts prepared by activation of bacteria growing on Fe minerals (Ma et al., 2017) also delivered superior electrochemical performance.

IN ACIDIC MEDIUM

Owing to its low cost and abundant sources, biomass carbon materials can be used as excellent cathode catalysts to replace noble-metal electrocatalysts. One typical material is nitrogen-doped nanoporous carbon flakes extracted from low-cost bagasse (Yuan et al., 2016). Moreover, the enoki mushroom derived carbon electrocatalyst also possesses outstanding ORR activity and durability. The significant difference in ORR activity of the two carbon materials is shown in Figure S3A. Extraction of N-doped carbon nanomaterial from Nenoki mushroom biomass in a certain temperature is shown in Figure S3B (Guo et al., 2015). In addition to the above examples in an acid medium, sludge-based multi-doped electrocatalysts (Yuan and Dai, 2016) and corn starch derived nitrogen-doped carbon electrocatalysts (Wang et al., 2012) are also utilized for ORR in acidic medium.

As we discussed above, the biomass-derived materials not only have outstanding contributions in terms of catalytic performance, but also promote environmental improvement. Therefore, we establish a link between the electrolytes, biomass precursors, optimal preparation temperature, specific surface area, and onset potential of various ORR electrocatalysts, which are listed in Table 1.

CONCLUSIONS AND OUTLOOK

In recent years, there have been great progresses to develop biomass-derived carbon ORR electrocatalysts for meeting the requirements of high performance. Some materials, especially materials derived from biomass materials, have comparable or
superior ORR properties and better stability than commercial Pt/C. Thus, biomass-derived carbons have attracted particular interest as a potential substitute for commercial Pt/C due to their good activity, low-cost, and reproducibility (Gasteiger and Markovic, 2009). In addition to the biomass electrocatalysts mentioned in this minireview, there will be numerous high-performance biomass-derived electrocatalysts for practical application in the future.

Although biomass-derived carbon materials have the widest sources and the lowest price, the controllability toward distribution of active sites is very general, which depends on the composition and structure of the biomass itself. How to achieve large-scale production is also an urgent problem preventing industrialization. Up to now, most biomass-derived carbon materials are only suitable for catalyzing oxygen reduction reactions under alkaline conditions, and their performance is unsatisfying in neutral and acidic media, which seriously affects the large-scale application of fuel cells.

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AUTHOR CONTRIBUTIONS

MW, SW, and WK wrote the manuscript. HY and ZL modified the manuscript. SY and GL supervised the manuscript.

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SUPPLEMENTARY MATERIAL

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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