

Energy storage through CO₂ electroreduction: A brief review of advanced Sn-based electrocatalysts and electrodes

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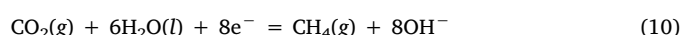
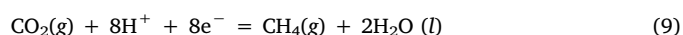
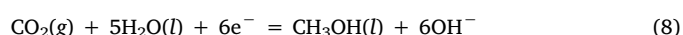
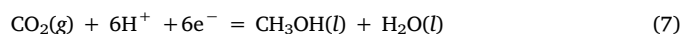
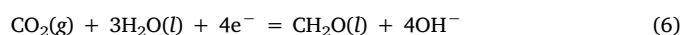
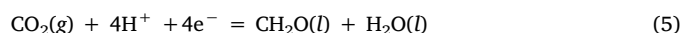
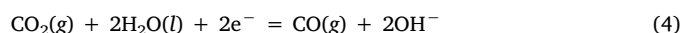
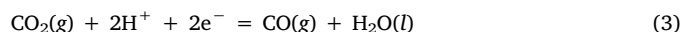
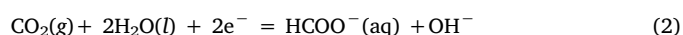
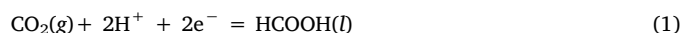
ABSTRACT

As fossil fuel usage continues to increase on a global scale, effective CO₂ conversion methods become increasingly important as many different avenues are being actively explored. In the case of CO₂ reduction, Sn-based electrocatalysts and their associated electrodes have shown great promise. In this review, recent progresses in Sn-based electrocatalysts and their associated electrodes for CO₂ reduction are briefly reviewed with a focus on catalyst synthesis, electrode fabrication, and electrocatalytic performance. Sn-based catalytic performance is greatly affected by the structure, morphology and composition of the catalyst, which in turn are strongly dependent on synthesis strategies and processes. Here, three catalyst synthesis methods: hydrothermal, electrodeposition and plasma treatment methods will be discussed and analyzed. Furthermore, electrode fabrication methods using Sn-based catalysts for CO₂ electroreduction will also be discussed to optimize catalytic activity, stability, and product selectivity so as to maximize CO₂ electroreduction performance. Finally, several technical challenges are analyzed and future research directions are proposed to facilitate further research and development toward the practical usage of Sn-based catalysts.

1. Introduction

With increasing energy demands, fossil fuel consumption has steadily increased, releasing more and more carbon dioxide (CO₂) into the atmosphere, causing global warming and dangerous climate change [1]. Because of this, CO₂ sequestration, storage, and conversion are becoming vital. In regards to CO₂ conversion, chemical, electrochemical [2–9], photochemical [6,10–16], photoelectrochemical [17–19] and biological methods [20,21] have all been explored as potential solutions. Among these methods, the electrochemical method; in which renewable or abandoned electrical energy can be utilized to electrolyze CO₂ into low carbon fuels, has shown great promise [4] and is recognized as a feasible solution for energy conversion and storage. There are major two approaches to CO₂ electrochemical reduction in which one operates at low-temperatures (less than 100 °C) using fuel-cell typed electrolyzers, and the other operates at high-temperatures (600–900 °C) using solid oxide electrolysis cells. In the case of low-temperature operations, the electrolyzer cathodes require an electrocatalyst to catalyze the CO₂ electroreduction process in which different catalysts generally lead to different and/or mixed products, which can be C1 products such as formic acid/formate and carbon monoxide

(formed via 2 e[−] process), formaldehyde (4 e[−]), methanol (6 e[−]), methane (8 e[−]), and sometimes C2 (e.g. C₂H₄) [4,22], which are formed as below:



HCOOH is thought to be more practical and desirable product. The advantage and benefits of its production over other C1 products are

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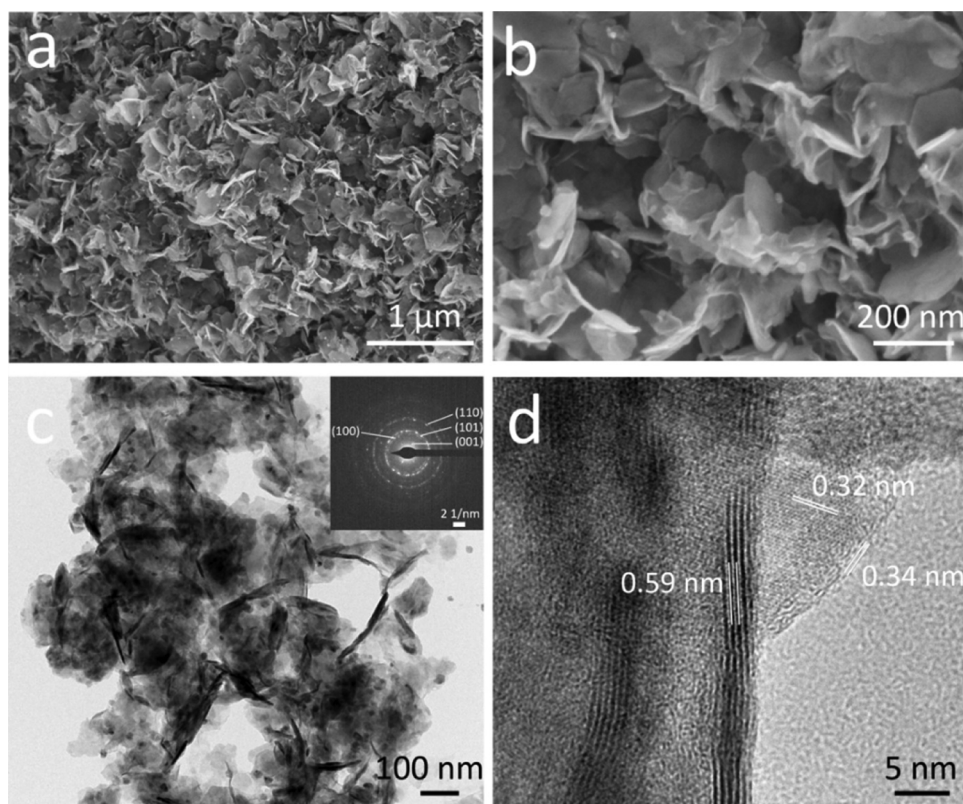


Fig. 1. Morphology characterizations of SnS₂/rGO. (a, b) SEM and (c, d) TEM images. Inset in (c) is the corresponding SAED pattern. [32].

obvious. First, formic acid is one of the attractive candidates as a liquid fuel for the hydrogen economy because of its high volumetric hydrogen density of 53 g of H₂/L, nontoxicity, safety, and transportability [23–26]. It can easily be converted into H₂ under ambient conditions with the development of an efficient formic acid dehydrogenation catalyst [27–29]. It is also a useful energy carrier for fuel cell applications [30–32]. Second, HCOOH / HCOO[−] is an important raw materials for the production of various organic agents through thermal treatments or catalytic reactions [33–36]. The largest users of formic acid have been for long time the leather and tanning industries which, in 2009, were surpassed by silage preservation (cereals and meat) and animal feed additives [34,37]. Third, since the ERCO₂ to the reduction products except for HCOOH / HCOO[−] and CO deal with slow kinetics of multiple electron transfer, it is difficult to achieve a high FE [38]. Thus, the predicted feasibility of the products decreases according to the series CO ≈ HCOOH > CH₃OH ≫ C₂H₄ > CH₄, and the commercialization of a CO₂-to-formate process is the most feasible and the most likely profitable route, compared to processes that produce CH₃OH, CO or long-chain hydrocarbons [39,40]. Fourth, it should be mentioned that both HCOOH and CO sell for near \$1200 per ton of product and require approximately 2500 kW h/ton for their production via electrochemical CO₂ conversion. Other products, such as CH₄, require nearly 40,000 kW h/ton for conversion, and would only achieve \$200–\$300 per ton on the market [41].

In the industrial production of HCOOH, BASF technology is popularly adopted [34], which uses both methanol and CO as raw materials: 1) CH₃OH + CO → CH₃OCHO; 2) CH₃OCHO + H₂O = CH₃OH + HCOOH. The net reaction is H₂O + CO = HCOOH. However, the separation of formic acid from the reaction mixture requires a dedicated extraction and brought about an economic and energetic cost. Formic acid can also be produced from biomass and biomass-derived products through hydrolysis and oxidation processes [42]. Anyway, this CO₂-based approach often requires various raw materials such as H₂ and NH₃, and severe reaction condition. Compared to them, the ERCO₂ is

conducted by simpler technology under milder conditions.

To promote CO₂ electroreduction, great efforts have been made to develop highly active, stable, and selective electrocatalysts, with many earlier results revealing that single metals such as Cu [43–45], Zn [9], In [46,47], Sn [48–59], Pb [60–62] and Bi [63–68] can selectively catalyze CO₂ electroreduction into low-carbon fuels. Among these metals, Sn and Sn-alloy catalysts have attracted great attention because of their promising properties [69–71]. For example, Hori et al. [72] reported a single metal Sn electrode that can produce a high FE_{formate} (~88.4%) at an electrode potential at -1.1 V vs. reversible hydrogen electrode (RHE) for CO₂ electroreduction. Some other researchers [53,73] also reported that if the electrode potential was controlled at -0.66 V vs. RHE, their Sn oxide electrode can provide a FE of over 30% as compared with their single metal Sn electrode, which could only provide a FE of only 12%, suggesting that trace amounts of tin oxide on the surface of Sn electrodes can significantly impact catalytic performances for CO₂ electroreduction reactions [55,74,75]. In recent decades, nanotechnologies have also been widely employed for electrocatalyst synthesis and electrode fabrication and significant progresses have been made [76–78].

To facilitate the research and development of high performance catalysts and electrodes for CO₂ electroreduction, this article will review the recent progresses using single metal, oxide, and alloy Sn-based electrocatalysts for the ERCO₂ to produce HCOOH/ HCOO[−] as examples. Technical challenges will also be analyzed and future research directions and solutions will be proposed.

2. Development of sn-based catalysts and electrodes

Single metals, oxides and alloys of Sn can be applied as desirable catalysts for CO₂ electroreduction [4] in which Sn-based materials can generally be used as bulk electrodes [50,52,79,80] and/or supported catalysts. Support materials include metal plates [48,55,56,81,82], meshes [49,83], graphite plates [84,85], carbon papers/fibers/cloths

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