



Ordered mesoporous ruthenium oxide with balanced catalytic activity and stability toward oxygen evolution reaction

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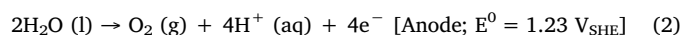
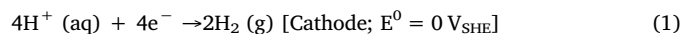
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ABSTRACT

A unique ordered mesoporous RuO₂ (Meso-RuO₂) catalyst with improved activity and stability is synthesized using the nano replication method. The initial activity for oxygen evolution reaction (OER) of the synthesized Meso-RuO₂ catalyst is compared with that of the state-of-the-art catalyst (IrO₂) and a high surface area RuO₂ catalyst synthesized by the well-known Adams method (Ad-RuO₂), respectively. The synthesized Meso-RuO₂ catalyst exhibited lower overpotential of 266 mV at 10 mA cm⁻² than IrO₂ (308 mV) or Ad-RuO₂ (326 mV). Furthermore, the overpotential difference in the OER before and after the accelerated stability test for Meso-RuO₂ was only 26 mV, indicating tolerable electrochemical stability. The enhanced activity and stability for the OER of the Meso-RuO₂ could be attributed to the effect of the ordered mesoporous structure of RuO₂ and the good electronic conductivity index (ratio of the electrochemical surface area and specific surface area). This study provides significant insights into the relationship between the OER performance and physicochemical properties of the catalysts.

1. Introduction

Increasing energy demands and the intermittency and instability of renewable energy generation by wind and photovoltaic devices have led to an increasing shift toward the use of polymer electrolyte membrane water electrolyzers (PEMWEs) as a means of energy storage. The primary reaction in the PEMWE is the electrolysis of water, which is simultaneously accompanied by the hydrogen evolution reaction (HER, Eq. (1)) and oxygen evolution reaction (OER, Eq. (2)) [1]:



Commercialization of the PEMWE is challenging, because the sluggish kinetics of the OER at the anode demands more energy [2–7], resulting in low efficiency and poor durability. OER catalysts under acidic conditions based on metals such as Ir and Ru have been extensively investigated to address this challenge [8–25]. However, most materials with excellent OER activities are unstable [26–28]. For example, the activity of metals in the OER decreases in the order Os >

Ru > Ir > Pt > Au, while the stability of the metals decreases in the order Au > Pt > Ir > Ru > Os. Thus, to achieve a balance between the catalytic activity and stability, the mixed metal oxide strategy, wherein a more active metal (e.g., Ru) and a relatively stable metal (e.g., Ir) are mixed, have long been investigated [10,29–33]. Several studies have focused on improving the catalytic performances of such materials by strengthening the metal/metal-oxide support interaction [34–41] and by intentionally implementing the leaching of transition metals [11,12,42–49]. Nonetheless, due to the dissolution of the metals, there is a great demand for the development of mixed metal oxide catalysts with desirable stability [1,28]. In addition, the extreme scarcity of Ir makes its use very expensive [2]. Hence, it is necessary to develop non-Ir based OER catalysts to reduce the system cost. For example, CeO₂-supported Ru could improve the mass activity of the OER catalyst [15].

In addition, to improve the utilization of Ru-containing catalysts, a film consisting of three-dimensional mesoporous RuO₂ with an ordered mesoporous structure was introduced recently [16]. In particular, it was suggested that during the electrochemical evaluation, the ordered mesoporous structure could improve the diffusion of reactants and

Abbreviations: AST, accelerated stability test; BET, Brunauer-Emmett-Teller; DIW, de-ionized water; ECSA, electrochemical surface area; EIS, electrochemical impedance spectroscopy; HER, hydrogen evolution reaction; HMDS, hexamethyldisilazane; OER, oxygen evolution reaction; PEMWE, polymer electrolyte membrane water electrolyzer; PSD, pore size distribution; RDS, rate determining step; TEM, transition electron microscopy; XRD, X-ray diffractometry

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