



Control of the pore size distribution inside the RuO₂ catalyst by using silica nanosphere particle for highly efficient water electrolysis

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ABSTRACT

As state-of-the-art catalysts for polymer electrolyte membrane water electrolysis (PEMWE), IrO₂ and RuO₂ have been extensively developed to accelerate the oxygen evolution reaction (OER) and to achieve high stability under acidic conditions. In this study, RuO₂ catalysts with controlled pore size distributions, particularly with a large number of macropores, are synthesized using a modified Adams method with silica nanosphere particles (SNPs) as a template. The specific surface areas of the prepared catalysts are 240–270 m² g^{−1} regardless of the sizes of the SNP template. Also, all RuO₂ catalysts have a rutile structure suggested from X-ray diffraction. The RuO₂ catalysts with large portions of macropores (more than 20% of the total pore volume) exhibited greater OER activity and stability than the catalysts prepared using the conventional Adams method. These enhancements are attributed to fast mass transport during the reaction and the 50% increase in the electrochemically active surface area (ECSA) of the RuO₂ catalysts prepared from the modified method over those prepared by the conventional Adams method. In addition, the enhanced catalytic activity (overpotential of 263 mV at 10 mA cm^{−2}) of RuO₂ with a large portion of macropores is revealed at the single-cell level in PEMWE.

1. Introduction

As evidenced by their growing share in total energy generation, renewable energy sources such as photovoltaic and wind power are promising alternatives to fossil fuels [1–4]. However, the supply uncertainty and instability of wind and solar power [4–7] have inspired a search for eco-friendly power storage measures such as power-to-gas systems, which use surplus energy to produce hydrogen by electrolysis [1,8,9]. As hydrogen is a zero-emission energy carrier with advantages of long-term and large-scale storage [8,10–13], many studies have focused on its production by polymer electrolyte membrane (PEM) water electrolysis owing to high current operation and capability of intermittent operations [14–16].

Although IrO₂ is commonly used as a stable oxygen evolution reaction (OER) catalyst at the anode of PEM electrolyzers, its scarcity and high overpotential preclude its large-scale application [17–20]. Thus, there is a demand to develop non-Ir catalysts (such as ruthenium dioxide (RuO₂)) with secured activities and stabilities. Studies in this field have mostly been focused on the formation of binary oxides such as Cr_{0.6}Ru_{0.4}O₂ and IrRuO_x or the structural- control of RuO₂ to provide a large surface area and porous structures [19,21,22].

For several decades, the nano-template method was studied in an attempt to control the nanostructures of catalysts and reveal the effects of each pore size during catalytic reactions [23–29]. For OER catalysts, mesoporous and macroporous metal oxides were prepared using ordered mesoporous silica such as KIT-6 and SBA-15 [25,26] and micro-sized templates such as silica spheres or polymer beads [27–29]. Although those mesoporous and macroporous structures enhanced OER activity and stability, owing to high surface areas and fast mass transport, the nano-template method developed only specific pore sizes due to the uniform and ordered structure of the templates and there was no attempt to synthesize metal oxides with various pore sizes using the nano-template method for OER, to the best of our knowledge [23, 25–29].

The Adams fusion method is a simple and commonly used method to synthesize metal oxides with large specific surface areas and micropore-dominant structures [17]. For further structural modification, several types of chelating agents are applied [30,31]. For example, Li et al. prepared micro/mesopore developed IrO₂ using ammonia as a chelating agent and porogen via the Adams fusion method [30]. The developed micropores facilitated mass transport by channeling the active sites to the mesopores. In our previous study, Meso-RuO₂ (synthesized using the

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