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journal homepage: www.elsevier.com/locate/apcatbAtomic interactions of two-dimensional PtS₂ quantum dots/TiC heterostructures for hydrogen evolution reactionSangmin Jeong^a, Hien Duy Mai^a, Tri Khoa Nguyen^a, Jong-Sang Youn^b, Ki-Hun Nam^c, Cheol-Min Park^{c,d,**}, Ki-Joon Jeon^{a,e,*}^a Department of Environmental Engineering, Inha University, Incheon, 22212, Republic of Korea^b Department of Energy and Environmental Engineering, The Catholic University of Korea, Bucheon, 14662, Republic of Korea^c School of Materials Science and Engineering, Kumoh National Institute of Technology, 61 Daehak-ro, Gumi, Gyeongbuk, 39177, Republic of Korea^d Department of Energy Engineering Convergence, Kumoh National Institute of Technology, 61 Daehak-ro, Gumi, Gyeongbuk, 39177, Republic of Korea^e Program in Environmental and Polymer Engineering, Inha University, Incheon, 22212, Republic of Korea

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ABSTRACT

Two-dimensional quantum dots (2D QDs) comprising PtS₂ with low Pt loading (0.002 wt.%) distributed on a distinctive CVD-grown titanium carbide substrate (PtS₂/TiC) was successfully synthesized and employed for a hydrogen evolution reaction (HER). Notably, despite the low loading of the former component, PtS₂/TiC showed excellent HER activity with a superior overpotential (55 mV at 10 mA/cm²) to that of commercial Pt/C (50 mV at 10 mA/cm²). The Faraday efficiency of PtS₂/TiC was found to be 92.5 %, revealing the superior properties of hydrogen production. The In-situ Raman spectra reveal the important role of S atoms in PtS₂ as the active sites for HER, as evidenced by S—H bonding formation at 2532 cm^{−1} during the HER process. This study provides a fundamental understanding essential for the design of more efficient catalysts in the field of electrochemical applications.

1. Introduction

Research on 2D quantum dots (QDs) has recently garnered considerable interest due to their unique structure-dependent and electrical properties [1–4]. 2D QDs have a higher surface-to-volume ratio, which can overcome the issues of low metal utilization efficiency in conventional electrocatalysts, wherein only the surfacial atoms participate in the catalytic process while a majority of the bulk atoms state inactive [1, 2]. However, adverse aggregation of 2D QDs into larger crystals during the synthesis and operation due to the high surface energy remains problematic, necessitating the dispersion of the catalysts firmly on a conductive and stable matrix to achieve a high hydrogen evolution reaction (HER) performance. Transition metal dichalcogenides (TMDs) with the formula MX₂ (where M = group-4 to group-10 transition metals, X=S, Se, Te) are promising catalysts owing to their structure and layer-dependent electrochemical properties [5]. Despite tremendous research effort intensively devoted toward group-6 (e.g., MoS₂, WS₂) [6–8], only a few reports have revealed the electrocatalytic behaviors of

other groups in the TMD family (e.g., group-10 TMDs) [9–12]. Similar to MoS₂, PtS₂ with band edge energies near the redox potentials of hydrogen evolution (H⁺/H₂) is considered an efficient HER electrocatalyst among group-10 TMDs [13]. However, research on PtS₂ as HER catalysts is very scarce, and the reported HER activity is far below that required to replace commercial Pt/C [11]. Inspired by the high potential of PtS₂ for HER, we strive to explore PtS₂ QDs deposited on suitable support materials. Titanium carbide (TiC) is a promising electrocatalyst or supporting material for electrochemical applications because of its high electrical conductivity and rigidity as well as exceptional chemical and thermal stability [14,15]. Furthermore, the orbital hybridization between d-orbitals of transition metals and p-orbitals of carbon leads to a strong interaction between TiC and noble metal atoms (e.g., Pt, Pd, Au), thus improving the stability and the electrocatalytic performance [15–17]. Powder-like can be synthesized by direct carbothermal reaction and molten salt-assisted reduction. Nevertheless, most of these materials often undergo harsh reaction conditions (i.e., high temperature and prolonged reaction time) or involve the excessive use of

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