



# One-pot microwave synthesis of hierarchical C-doped CuO dandelions/g-C<sub>3</sub>N<sub>4</sub> nanocomposite with enhanced photostability for photoelectrochemical water splitting

Seyed Morteza Hosseini H.<sup>a,b</sup>, Roozbeh Siavash Moakhar<sup>a,b</sup>, Foad Soleimani<sup>a</sup>,  
Sayed Khatiboleslam Sadrnezhaad<sup>a,\*</sup>, Saeid Masudy-Panah<sup>c,d</sup>, Reza Katal<sup>e</sup>, Ashkan Seza<sup>a</sup>,  
Navid Ghane<sup>a</sup>, Seeram Ramakrishna<sup>f,\*</sup>

<sup>a</sup> Department of Materials Science and Engineering, Sharif University of Technology, Azadi Ave., Tehran, P.O. Box: 11155-9466, Iran

<sup>b</sup> Institute of Materials Research and Engineering, A\*STAR (Agency for Science, Technology and Research), 2 Fusionopolis Way, Innovis, Singapore 138634, Singapore

<sup>c</sup> Low Energy Electronic Systems (LEES), Singapore-MIT Alliance for Research and Technology (SMART) Centre, Singapore

<sup>d</sup> Department of Electrical and Computer Engineering, National University of Singapore, Engineering Drive 2, Singapore 119260, Singapore

<sup>e</sup> Department of Civil & Environmental Engineering, National University of Singapore, Engineering Drive 2, Singapore 119260, Singapore

<sup>f</sup> Centre for Nanofibers and Nanotechnology, Department of Mechanical Engineering, National University of Singapore, 9 Engineering Drive 1, Singapore 117576, Singapore

## ARTICLE INFO

### Keywords:

Cupric oxide (CuO)  
Graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>)  
Microwave-assisted method  
Nanostructured thin film  
Photoelectrochemical water splitting

## ABSTRACT

Cupric oxide (CuO) is a semiconductor of choice for photocathode in photoelectrochemical (PEC) applications due to its great sunlight absorption capability. However, photocorrosion is the main drawback of CuO. Herein, CuO/graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) with a unique microstructure, enhanced PEC performance, and considerable photostability is synthesized under microwave irradiation. A facile, one-pot method is utilized to directly deposit the nanocomposite onto fluorine-doped tin oxide from a solution containing copper precursor and urea. Possible mechanism of CuO/g-C<sub>3</sub>N<sub>4</sub> formation through this novel method is investigated. It is elucidated that controlled amounts of urea critically determine the morphological evolution of CuO, while its excess quantities convert to g-C<sub>3</sub>N<sub>4</sub> in the presence of CuO as the catalyst. Through an appropriate heat treatment, carbon is doped into CuO lattice. The obtained C-doped CuO/g-C<sub>3</sub>N<sub>4</sub> demonstrates 227% enhancement over CuO in photocurrent density and ~80% photocurrent retention. The enhanced photoelectrocatalytic activity is mainly attributed to unique morphology of CuO, effective separation of charge carriers, and formation of heterojunction. These characteristics manifest the superiority of this approach over many other chemical-based methods. The nanocomposite synergistically integrates the advantages of both the constituents, offering a low-cost, efficient photocathode for PEC water splitting, photocatalytic hydrogen evolution, and degradation of pollutants.

## 1. Introduction

Achieving a sustainable energy supply has been the motivating force behind numerous efforts in concern with the high demand for energy and scarcity of natural resources [1–3]. Solar light-driven water splitting and hydrogen production systems are among the promising candidates to supply solar fuel as an abundant and renewable green energy source in lieu of fossil fuels [4–6]. They generally consist of an electrode coated by photoactive materials and/or semiconductors with specific characteristics and properties [4,7]. The wise selection of materials with controlled morphologies and engineered bandgap structures is

highly demanded to optimize the solar light-harvesting, charge transfer, and electron-hole separation capabilities [8,9].

As an inexpensive, abundant semiconductor, cupric oxide (CuO), which possesses a relatively small bandgap energy of around 1.2–1.8 eV and consequently high sunlight absorption, has shown promising applications in PEC water-splitting [10,11], pollutants removal [12], and lithium-ion battery making [13]. Nevertheless, drawbacks such as high electrode photocorrosion, fast recombination of photoexcited charges, and low charge transfer across the photoelectrode–electrolyte restrict the application of CuO as the hydrogen evolution reaction (HER) catalyst in a PEC cell [8,14]. Several attempts have been conducted to

\* Corresponding authors.

E-mail addresses: [s.m.hosseini.sharif@gmail.com](mailto:s.m.hosseini.sharif@gmail.com) (S.M. Hosseini H.), [roozbehsiavash@gmail.com](mailto:roozbehsiavash@gmail.com) (R. Siavash Moakhar), [foadsoleimani@gmail.com](mailto:foadsoleimani@gmail.com) (F. Soleimani), [sadrnezhaad@sharif.edu](mailto:sadrnezhaad@sharif.edu) (S.K. Sadrnezhaad), [seeram@nus.edu.sg](mailto:seeram@nus.edu.sg) (S. Ramakrishna).

<https://doi.org/10.1016/j.apsusc.2020.147271>

Received 2 May 2020; Received in revised form 21 June 2020; Accepted 29 June 2020

Available online 17 July 2020

0169-4332/ © 2020 Elsevier B.V. All rights reserved.