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Author: Peiqiang Li Jinfeng Xu Hua Jing Chenxiao Wu Hui

Peng Jing Lu Hongzong Yin

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ACCEPTED MANUSCRIPT

Wedged N-doped CuO with More Negative Conductive Band and Lower Overpotential for High Efficiency Photoelectric Converting CO₂ to Methanol

Peiqiang Li*, Jinfeng Xu, Hua Jing, Chenxiao Wu, Hui Peng, Jing Lu, Hongzong Yin*

College of Chemistry and Material Science, Shandong Agricultural University, 271018, People's Republic of China

Abstract: One dimensional wedged N-doped CuO has been in situ prepared on Cu substate by anodization method. The as-prepared material with a length of 786 nm and width of 143 nm presents uniform wedged structure. The energy band gap and conductive band is 1.34 eV and -1.03 eV, respectively. The carrier concentration of wedged N-doped CuO (7.5×10⁵ m⁻³) is about 10⁸ times that of CuO film (4.8×10⁻³ m⁻³). The as-prepared material promotes the separation of photoelectrons and holes efficiently to achieve the excellent photocatalytic reduction property. For the electrochemical properties aspect, the electrochemical adsorptive active site for CO₂ on the as-prepared material (25 nmol) is 252 times that of CuO film (99 pmol). And the overpotential shifts 0.17 V positively relative to CuO film. Furthermore, it shows outstanding electrocatalytic property for CO₂ reduction. In the process of photoelectrocatalytic reduction CO₂, the predominant product is methanol, the current efficiency on wedged N-doped CuO electrode (84.4 %) is 14.5 times that of CuO film (5.84 %), the methanol output $(3.6 \text{ mmol L}^{-1} \text{ cm}^{-2})$ is 139 times that of CuO film (0.026 mmol L⁻¹ cm⁻²). In addition, it shows that the methanol output in the photoelectrocatalytic process is 1.3 times of the simple addition of photocatalytic process and electrocatalytic process, which indicates the distinct 1+1 > 2 synergistic effect between electrocatalytic reduction and photocatalytic reduction.

Key words: Wedged N-doped CuO; Photoelectrocatalytic; Carbon dioxide; Methanol; CuO film

1. Introduction

Global warming and resource shortage are the problems the society facing, the CO_2 emission has reached 31.6 billion metric tons on the world in 2012, which causes a series of environmental problems. As we known, CO_2 is also a very precious C1 resource, so the conversion of CO_2 to organics is not only benefit to the CO_2 emission reduction, but also contributed to the energy

^{*} Corresponding author: College of Chemistry and Material Science, Shandong Agricultural University 61 Daizong Road, Tai'an, Shandong 271018(PR China), Tel: 86-0538-8249017.

⁻ E-mail addresses: chem_carbon@outlook.com; pqli@sdau.edu.cn

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