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Electrodeposited Nickel Cobalt Sulfide Flowerlike Architectures on Disposable Cellulose Filter Paper for Enzyme-Free Glucose Sensor Applications

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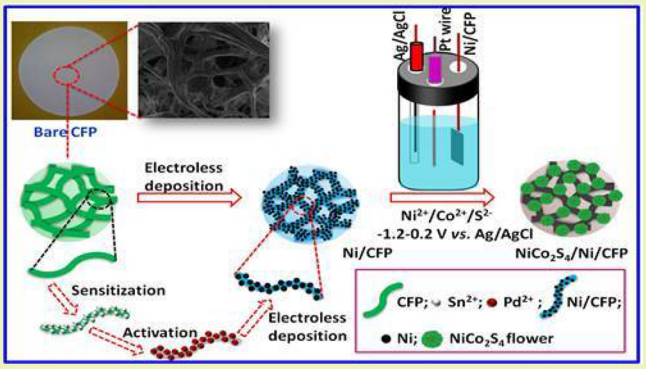
Supporting Information

ABSTRACT: Three-dimensional flowerlike nickel cobalt sulfide ($NiCo_2S_4$) nanostructures are electrochemically deposited over the Ni-modified cellulose filter paper (CFP). The fabricated $NiCo_2S_4$/Ni/CFP is explored as a binder-free and self-standing electrode for enzyme-free electrochemical detection of glucose in alkaline media, in which Ni/CFP and $NiCo_2S_4$ serve as the current collector and active material, respectively. Owing to the rich redox active sites, synergistic effect between Co^{4+} and Ni^{3+}, and rapid electron transportation tuned with the substitution of sulfur (S) with oxygen labile functional group, $NiCo_2S_4$/Ni/CFP exhibits considerable enzyme-free glucose-sensing performance with wide linear glucose concentration ranging from 0.5 μM to 6 mM, high sensitivity of 283 μA mM^{-1} cm^{-2}, and low detection limit of 50 nM. The developed $NiCo_2S_4$/Ni/CFP's sensing performances are reproducible, stable, and highly selective toward the analyte of interest in physiological interfering species. Thus, the self-standing, binder-free, inexpensive, easily disposable, and facile electrode fabrication process proposed in this study opens a new paradigm architecture for the development of efficient electrochemical sensor devices with affordable cost.



KEYWORDS: *Cellulose filter paper, Three-dimensional structure, Electrodeposition, Self-standing electrode, Chloride tolerance*

INTRODUCTION

The development of simple, reliable, cost efficient, and rapid glucose-monitoring devices has been effectively driven by rising demand in personal health care, diagnosis and management of diabetes mellitus, control of bioprocess, pharmaceutical analysis, and ecological and food monitoring.[1,2] An enzyme-free electrochemical glucose sensor (EFGS) is considered as the most convenient and effective tool as it provides the constructive features including excellent sensitivity, simple instrumentation, easy operation, low production cost, good reproducibility, and anti-interference abilities.[3,4] In general, EFGS is constructed with solid current collectors including glassy carbon electrode (GCE),[5] platinum (Pt),[6] and gold (Au).[7] However, the aforementioned electrodes require laborious polishing and modification procedures, which not only increase the cost of a sensor device but also preclude their practical utility. Hence, the fabrication of a self-standing, binder-free, easily disposable, and cost and time efficient electrode still receives a great deal of attention for the development of EFGSs. Recently, cellulose filter paper (CFP) has been considered as an ideal free-standing substrate due its widespread availability, low cost, light weight, extended porosity, interconnected scaffold architecture, and biodegradability.[8] The modification of CFP with highly conductive and catalytic active materials is logical for the development of EFGSs.

In this context, cobalt-based nanocatalysts including cobalt hydroxide ($Co(OH)_2$), cobalt oxide (CoO), and cobaltite oxide (Co_3O_4) have emerged as efficient electrochemical probes for EFGS owing to their low band gap (1.5−2.19 eV), low cost, and rational electrocatalytic activities.[9] Ding et al. reported Co_3O_4 nanofiber-based EFGS and observed the sensitivity and linear range of 36.25 μA mM^{-1} cm^{-2} and 0.97 μM, respectively, under alkaline conditions. The poor electrical conductivity (Co_3O_4, 10^{-5} S m^{-1}), easier agglomeration,

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