



## Short communication

## Enhanced activity of urea electrooxidation on nickel catalysts supported on tungsten carbides/carbon nanotubes



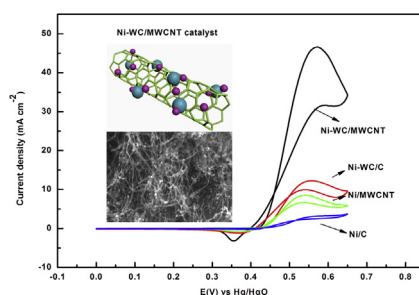
Lu Wang, Tingting Du, Jin Cheng, Xing Xie, Bolun Yang\*, Mingtao Li\*

Department of Chemical Engineering, State Key Laboratory of Multiphase Flow in Power Engineering, Xi'an Jiaotong University, Xi'an 710049, PR China

## HIGHLIGHTS

- The nanosized Ni were uniformly dispersed on WC/MWCNT by an impregnation method.
- The Ni-WC/MWCNT catalyst shows enhanced activity for the electrooxidation of urea.
- The MWCNT promote the electron and mass transfer of the urea electrooxidation.
- The combined effects on Ni, WC and MWCNT contribute to the urea electrooxidation.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

## Article history:

Received 8 September 2014  
 Received in revised form  
 29 December 2014  
 Accepted 22 January 2015  
 Available online 23 January 2015

## Keywords:

Nanostructured nickel-based electrocatalysts  
 Tungsten carbides  
 Multi-walled carbon nanotubes  
 Electrooxidation of urea

## ABSTRACT

Nickel nanoparticles with tungsten carbides supported on the multi-walled carbon nanotubes, noted as Ni-WC/MWCNT catalyst, is prepared through an impregnation method and used for the electrooxidation of urea in alkaline conditions. The micro-morphology and composition of the Ni-WC/MWCNT particles are determined by scanning electron microscopy (SEM), transmission electron microscopy (TEM) and X-ray diffraction (XRD) analysis. The electrooxidation activity and conductivity of the catalyst are investigated by cyclic voltammetry and electrochemical impedance spectroscopy, respectively. Characterization results indicate that the Ni nanoparticles are uniformly distributed on the WC/MWCNT framework, and the Ni-WC/MWCNT catalyst shows an improved activity for the urea electrooxidation. The current densities of Ni-WC/MWCNT are over 3 times and 15 times higher than those of the Ni-WC/C and Ni/C catalysts, respectively, and the electrochemical impedance also decreases markedly. The higher activity on Ni-WC/MWCNT is attributed to the support effect of MWCNT as well as the synergistic effect between Ni and WC.

© 2015 Elsevier B.V. All rights reserved.

## 1. Introduction

As a hydrogen carrier for long-term sustainable energy supply, urea ( $\text{CO}(\text{NH}_2)_2$ ) has been attracting increasing attention due to its stable, relatively non-toxic, non-flammable and renewable properties [1]. Recently, the electrooxidation of urea has been used as an

effective approach for hydrogen production as well as treatment of urea-rich wastewater [2]. Furthermore, the urea electrooxidation does not rely on the prohibitive noble-metal catalysts but a low-cost transition metal catalyst (nickel) in alkaline medium [3]. However, the current major challenge is to improve the catalytic activity and CO-tolerance of the catalyst.

During the electrooxidation of urea, nickel is oxidized to an active form ( $\text{NiOOH}$ ) which catalyzes the urea oxidation; however, the active nickel oxide state ( $\text{NiOOH}$ ) could be generated at

\* Corresponding authors.

E-mail address: [blunyang@mail.xjtu.edu.cn](mailto:blunyang@mail.xjtu.edu.cn) (B. Yang).

relatively high potential and be easily blocked by adsorbed CO, thus leading to low reaction rate and instability [4]. In our previous research, it has been found that the introduction of tungsten carbide (WC) as the co-catalyst in the nickel-based catalyst can improve the catalytic activity and stability [5]. The enhancement may be attributed to two reasons: i) the electron transfer from Ni to WC could facilitate the formation of NiOOH [5]; ii) the weak adsorption of CO on WC [6] would improve the CO-tolerance efficiently for Ni catalysts. Considering that the low specific surface area of WC is unfavorable for the dispersion of nickel particles [7], the activated carbon was used to improve the utilization of the active sites; however, the current density of urea electrooxidation is still limited by the relatively high electron and mass transfer resistance.

In order to further improve the electro-conductivity as well as the loading of the active sites, multi-walled carbon nanotubes (MWCNT) are used as the carrier in this work. Compared to the activated carbon, this type of nanotube-C possesses several unique features, such as highly graphitized tube-wall, nanosized channel and  $sp^2$ -C-constructed surface [8,9], which could provide high mass transfer [10] and electron transfer [11] in the catalysts. Therefore, a novel Ni-electrocatalyst with introduction of WC supported on MWCNT (noted as Ni-WC/MWCNT) was prepared and used for the urea electrooxidation in this study. The synergistic and structural effects on MWCNT, nickel and tungsten carbide could be expected to enhance the activity of urea electrocatalysis.

## 2. Experimental

### 2.1. Preparation of Ni-WC/MWCNT catalyst

The preparation of the catalyst is illustrated in Scheme 1. First, MWCNT (2.0 g,  $>200 \text{ m}^2 \text{ g}^{-1}$ , made by the CVD method, from TIME NANO Co. Ltd.) was incipiently impregnated in an ammonium paratungstate aqueous solution (containing 0.4 g  $(\text{NH}_4)_6\text{W}_7\text{O}_{24} \cdot 6\text{H}_2\text{O}$ ), followed by a temperature-programming reduction (TPR) treatment with  $\text{CH}_4$  and  $\text{H}_2$  to obtain WC/MWCNT framework (WC was 12.5 wt %). Then, these WC/MWCNT powders were added to a nickel nitrate aqueous solution (containing 2.8 g  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ) with the impregnation and another TPR process to produce the final catalyst containing 20 wt% Ni, 10 wt% WC and 70 wt% MWCNT (noted as Ni-WC/MWCNT). All the TPR procedures were as previously reported [5]. For comparison, Ni nanoparticles were supported on the WC/C, MWCNT or the activated carbon by the same procedures at the same loading, noted as Ni-WC/C, Ni/MWCNT or

Ni/C.

### 2.2. Structural characterization and electrochemical measurements

Field emission scanning electron microscopy (Hitach JSM-7000F, equipped with an energy dispersive X-ray spectrometer, EDS), transmission electron microscopy (TEM, Hitachi JEM-2100F) and X-ray diffraction (XRD, Rigaku D/max-2400,  $\text{CuK}\alpha$ ) were used to characterize the morphology and compositions of the catalysts. The activities and impedances were monitored using cyclic voltammetry and electrochemical impedance spectroscopy (EIS), respectively, in 1 M KOH/0.33 M urea solution (CHI760D instrument). The electrodes were prepared using previously reported procedures [5]. An Hg/HgO electrode and platinum wire were chosen as the reference and counter electrodes, respectively.

## 3. Results and discussion

### 3.1. Morphology and structure characterization of Ni-WC/MWCNT catalyst

The morphology and EDS images of the Ni-WC/MWCNT catalyst are shown in Fig. 1, which reveal that small and round grains are

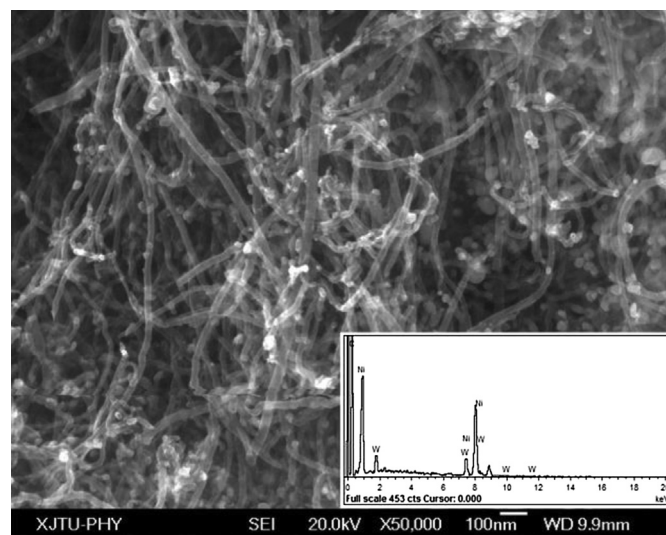
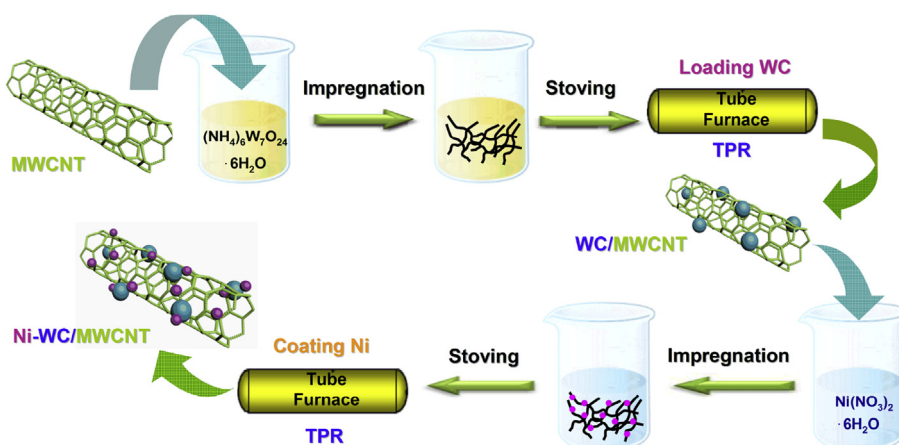


Fig. 1. SEM and EDS images of the Ni-WC/MWCNT catalyst.



Scheme 1. Schematic diagram of the Ni-WC/MWCNT catalyst preparation.

Download English Version:

<https://daneshyari.com/en/article/7733190>

Download Persian Version:

<https://daneshyari.com/article/7733190>

[Daneshyari.com](https://daneshyari.com)