



Flowing electrolytic synthesis of fluorescent carbon nanoparticles and carbon nanosheets



Su Yao, Yufei Hu^{*}, Gongke Li^{*}

School of Chemistry and Chemical Engineering, Sun Yat-sen University, Guangzhou, PR China

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ABSTRACT

A flowing electrolytic device equipped with large electrode area and high electric field intensity was designed for the preparation of fluorescent carbon nanoparticles (CNPs) and carbon nanosheets (CNSs). Comparing with normal electrolysis, the flowing electrolysis showed higher efficiency for the preparation and separation of the products. In the high intensity electric field, the CNPs and CNSs were enriched to the flowing electrolyte and the anode surface respectively. The obtained CNPs were regarded as a fluorescent probe of Fe^{3+} , and the CNSs modified electrode was fabricated to be an electrochemical sensor of acetaminophen for its electrocatalytic properties. So the CNPs and CNSs were expected for the application of fluorescent and electrochemical detection.

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1. Introduction

Electrochemical method is a traditional route to produce carbon-based nanomaterials [1–5]. In the most cases, carbon-based materials such as graphite anode were electrochemically oxidized in the electrolyte. According to the literature [6,7], abundant carboxyl groups were generated in the anode surface. The carboxyl groups reduced the interaction of the graphite layers, and increased the hydrophilicity of the graphite layers. So, the oxidized graphite layers easily exfoliated from the graphite surface. The exfoliated graphite layers have hydrophilic carboxyl groups and hydrophobic graphite surface. For the π - π interaction and hydrophilic interaction, the small exfoliated graphite layers tended to wrinkle in the form of CNPs [8], the large exfoliated graphite layers tended to be the other products such as carbon nanoribbons, carbon nanosheets and graphene sheets. However, the carbon-based anode was not necessary. The fluorescent CNPs also could be prepared by the electrooxidation of ethanol with the inert electrode [9]. Compare with the other preparation methods such as carbonization synthesis [10–13], hydrothermal synthesis [14–17], microwave synthesis [18–20], ultrasonic synthesis [21–23], and chemical methods [24–26], electrochemical method shows some unique advantages such as simple operation and low cost.

However, the electrochemical method has also met some defects such as by-product and residual salt. In order to get enough products, the electrolysis were always sustained for several hours unless the color of the electrolyte turned to brown or black [27,28], and the product was the mixture of different carbon nanomaterials [29,30]. In addition, the salts in electrolyte were also kept in the product. So, it is necessary for the separation of the nanoscale products and the removal of the salts in the electrolyte by long time dialysis [6,7]. In our previous work [31], the sonoelectrochemical method was proposed for the simple preparation of pure blue fluorescent CNPs by combining the high intensity electric field and ultrasonic field. The complex purification was avoided in this method because the deionized water was used as electrolyte. But it was hard to enlarge the scale of the sonoelectrochemical preparation for the limited electrode area.

Flowing electrolysis is an efficient electrochemical method. In this method, the electrolyte flow through the electrode surface. According to its high mass transfer rate and refresh electrode surface, the flowing electrolysis has been used for waste water treatment [32,33], electrochemical detector [34,35] and reaction kinetics study [36–38]. Herein, a novel flowing electrolysis method was developed for the preparation of carbon nanomaterials. Compare to our previous work, the high intensity electric field was retained because it was necessary for the electrolysis in deionized water. But the ultrasonic electrolysis was replaced with flowing electrolysis. The special designed flowing electrolytic device has a large electrode area, and the flowing deionized water which was used as electrolyte kept washing the electrode surface. Two kinds of carbon nanomaterials with different morphology such as CNPs and carbon nanosheet (CNSs) were generated and separated in the

^{*} Corresponding authors. Tel.: +86 020 84110922.

E-mail addresses: huyufei@mail.sysu.edu.cn (Y. Hu), cesgkl@mail.sysu.edu.cn (G. Li).

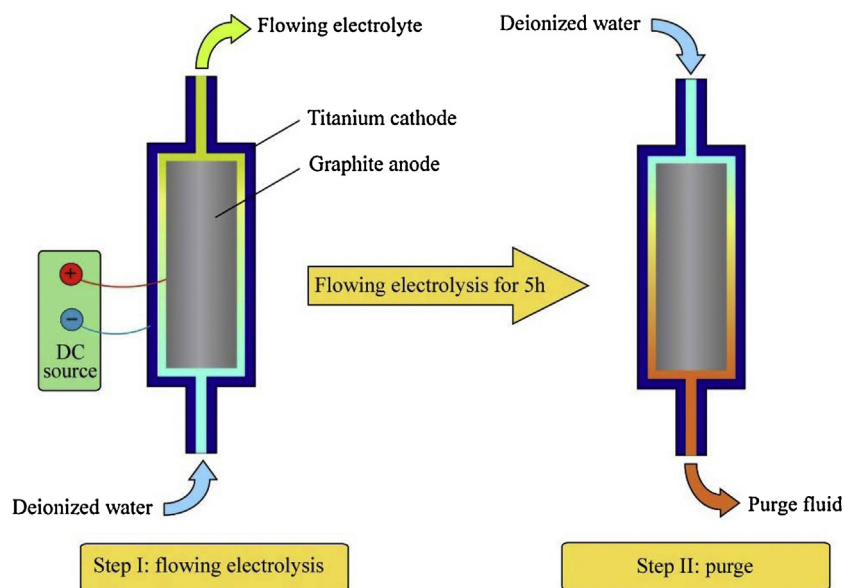


Fig. 1. Scheme of the flowing electrolytic device.

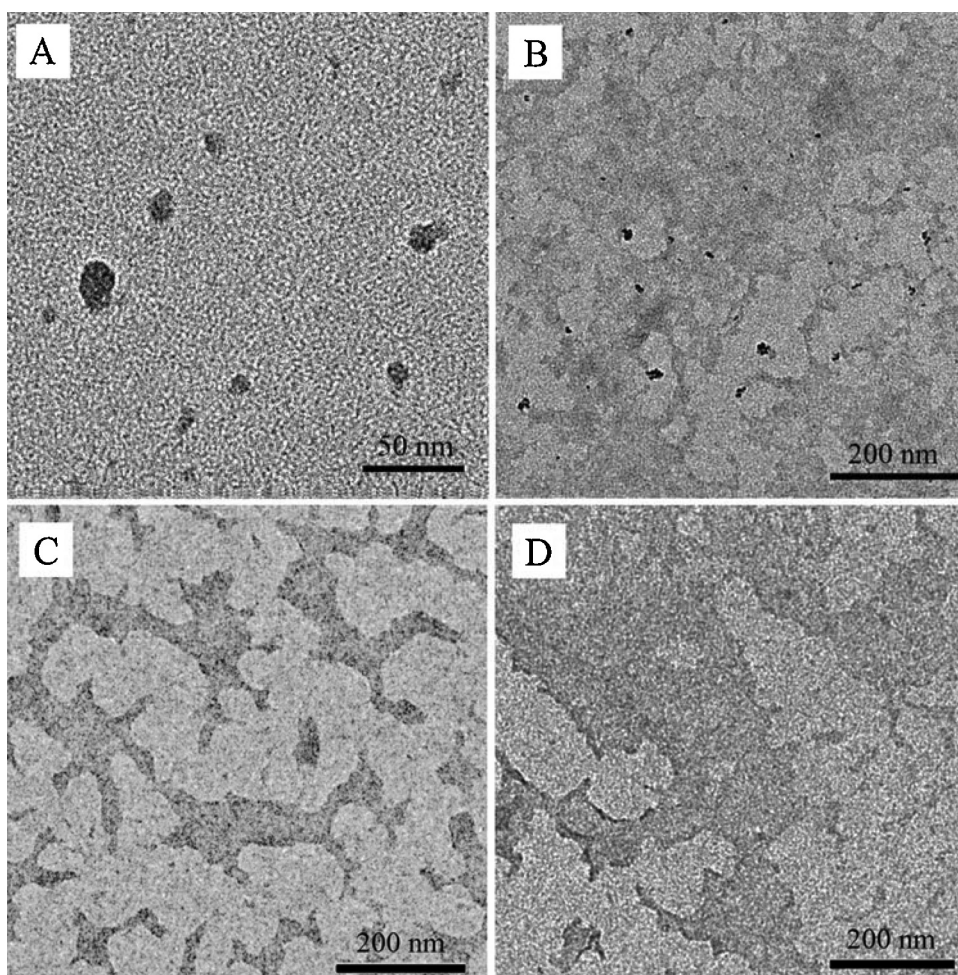


Fig. 2. The HRTEM images of flowing electrolyte (A) and purge fluid (B) which was obtained in the electrolytic current of 10 mA, and the HRTEM images of flowing electrolyte (C) and purge fluid (D) which was obtained in the electrolytic current of 160 mA.

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