DOI: 10.32347/2412-9933.2021.48.160-164

Cheng Weihua
Profession: Chemical Engineering, *orcid.org/0000-0003-2681-2341*
Yancheng Polytechnic College, School of Medicine and Health, China

GREEN PREPARATION OF SINGLE-WALLED CARBON NANOTUBES/GOLD NANOCOMPOSITES AND STUDY ON THE DETECTION OF INDIGO BLUE

Abstract. *Single-walled carbon nanotubes (SWNTs) have attracted more and more attention due to their unique structure and excellent properties. In this paper, single-walled carbon nanotubes /gold (SWNTs/Au) nanocomposites were prepared by a green and simple method, and a highly sensitive indigo blue sensor was constructed based on the nanocomposites. The results showed that the optimum pH of indigo blue (IB) was 5 and the optimum enrichment time was 270s. The reaction was controlled by surface adsorption. The unique feature of this method is that no toxic reducing agents or surfactants are added during the preparation process, which is environmentally friendly and sensitive to the detection of IB based on the nanocomposite modified electrode. Compared with gold nanomaterials, single-wall carbon nanotube-gold nanocomposites are more sensitive to IB. Comparing the current response signals of different pH values, 0.1mol/LPBS solution with pH=5 was selected as the test solution. When the enrichment time reaches 270s, the IB adsorbed on the electrode surface tends to be saturated, so the optimal enrichment time is 270s. The above experiments show that the nanocomposite material has a good electrocatalytic ability for IB, and the detection limit (3S/N) is 0.02μmol/L. The modified electrode has a short response time, a wide linear range, a low detection limit, good selectivity and high stability.*

Keywords: Single-walled carbon nanotubes; Gold nanoparticles; Indigo blue; Electrochemical sensor

Introduction

As one of the thinnest known materials with relatively large specific surface area, strong electrical conductivity and super high strength, carbon nanotubes have brought revolutionary progress in the semiconductor industry, Li ion batteries, aerospace and so on [1-4]. The physicochemical properties, preparation methods and nanomaterials of carbon nanotubes have attracted more and more attention [5 – 9]. The research on carbon nanotubes in China mainly focuses on the preparation and application of single-walled carbon nanotubes composites [10 – 12]. One or more efficient, environmentally friendly and low-cost production methods provide an important direction for the research of single-walled carbon nanotubes and their composites.

This paper mainly focuses on the preparation of single-walled carbon nanotubes/gold nanocomposites by a new green reduction method. The composite was prepared under mild experimental conditions. Instead of the traditional toxic hydrazine reduction at 24h, the mixture reaction of chlorogold acid and single-walled carbon nanotubes was carried out under alkaline heating condition, and a new single-walled carbon nanotubes/gold nanocomposites was prepared by this method. The electrochemical behavior of the nanocomposites was analyzed. Cyclic voltammetry (CV) was used to optimize the experimental conditions and discuss the optimal reaction conditions and mechanism. The results show that the nanocomposite has good electrocatalytic ability for IB, a food additive.

Experimental Part

Preparation of single-walled carbon nanotubes/gold nanocomposites

21mg single-wall carbon nanotubes were accurately weighed with an electronic balance, and 10mL deionized water was added into a round-bottom flask for 5min of ultrasonic oscillation. Add 20μL 0.024mol/L HAuCl4 solution, adjust pH to 13 with 0.5mol/L NaOH solution; The prepared solution was heated in an oil bath for about 3 hours at a temperature of 98-100℃.After the oil bath, take out the flask and conduct ultrasonic oscillation to prepare the required product SWNTs/Au compound, and pour it into 10mL plastic tube for reserve. If the product is too thin, centrifuge concentration can be carried out.

Preparation of modified Electrode

First, glassy carbon electrode (GCE) with a diameter of 3mm was polished on suede, and then polished to mirror surface on Al2O3 of 1.0 and 0.3μm. Ultrasonic treatment was conducted in water and ethanol for 2min, respectively, and nitrogen was used to blow dry. Finally, 5μ l 0.1MgML-1 single-walled carbon nanotubes/gold nanocomposite dispersed droplets were coated on the polished electrode and dried at room temperature to obtain GCE modified by single-walled carbon nanotubes/gold nanocomposite. In order to

conduct comparative experiments, single-walled carbon nanotubes and gold nanoparticles modified electrodes were also prepared by a similar method. The modified electrode was used to detect (IB) electrochemically.

Analysis Methods

Electrochemical Test Analysis The electrochemical experiment was conducted with a three-electrode system: glassy carbon electrode as the working electrode, platinum electrode as the counter electrode, and Ag/AgCl as the reference electrode. Cyclic voltammetry (CV) is one of the methods used in electrochemical testing.

Results and Discussion

UV Characterization

Figure 1 shows the UV-visible absorption spectra of SWNTs, SWNTs/Au composite and chlorauric acid. Curve a shows the absorption spectra of SWNTs dispersion with a strong absorption peak at 210nm. Curve b shows the absorption spectra of SWNTs/Au composite. Curve c is the absorption spectrum curve of $HAuCl_4$ dispersion. From the comparison of curves a and b, it can be seen that the absorption peak of the prepared SWNTs/Au composite appears at about 270nm, indicating that the reduction of single-walled carbon nanotubes is complete. In addition, curves b and c have a relatively wide absorption peak near the wavelength of 270nm, which is consistent with the conclusion of the absorption peak of chlorauric acid at about 280nm in the literature, indicating that the single-walled carbon nanotubes/gold nanocomposites have been successfully prepared.

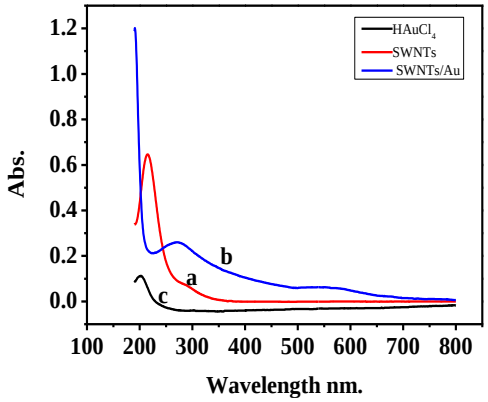

Figure 1 – UV-vis absorption spectra of SWNTs (a), SWNTs/Au composites (b) and $HauCl_4$ (c)

Electrochemical Behavior of modified Electrodes

The electrochemical properties of modified electrodes with different materials were studied by cyclic voltammetry. Figure 2 shows the cyclic voltammetry of the composite modified electrode in blank and 0.1mol/LPBS buffer containing 50μmol/LIB (pH7.0). As can be seen from the comparison in Figure 2, SWNTS-Au /GCE did not produce any peak for the blank solution. After IB was added, a large reversible reduction peak appeared at 0.77V, which could be used to quantify the concentration of IB. Figure 3 shows the cyclic voltammetry of different modified electrodes in 0.1mol/LPBS buffer solution (pH7.0) containing 50μmol/L indigo blue (IB). The peak current of Au/GCE (Curve b) is almost zero, and the peak current of SWNTS-Au /GCE (Curve c) is the largest. This is because SWNTs has certain defects on its surface, so it is easy to chemically modify the surface of gold nanoparticles and form a strong force between each other. This interaction and the good dispersion of gold nanoparticles on the surface of SWNTs make SWNTs/Au complex have good electrocatalytic performance for IB, which also indicates the successful preparation of SWNTs/Au nanomaterials in the experiment.

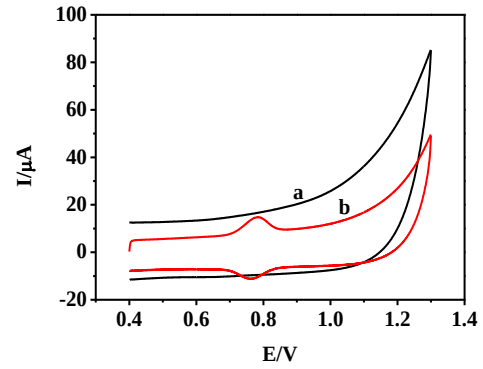

Figure 2 – SWNTS-Au /GCE is not in the cyclic voltammetry of 0.1 M PBS (buffer (cyclic diagram in pH 7.0) with IB(a) free and with 50 μM IB(b)

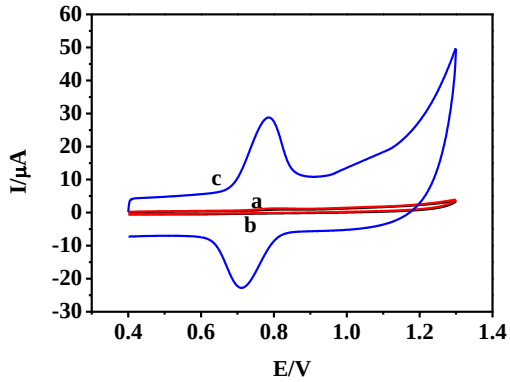

Figure 3 – GCE(a), Au/GCE(b), SWNTS-Au /GCE(c) contained in a 0.1m PBS flush containing 50 μM IB, sweep rate: 100 mV/s

Influence of pH

Figure 4A is the cyclic voltammetry of SWNTs/Au/GCE in PBS buffer solutions with different pH, and Figure 4B is the relationship between peak current and pH. It can be seen from the figure that pH has A great influence on the electrochemical determination of IB.

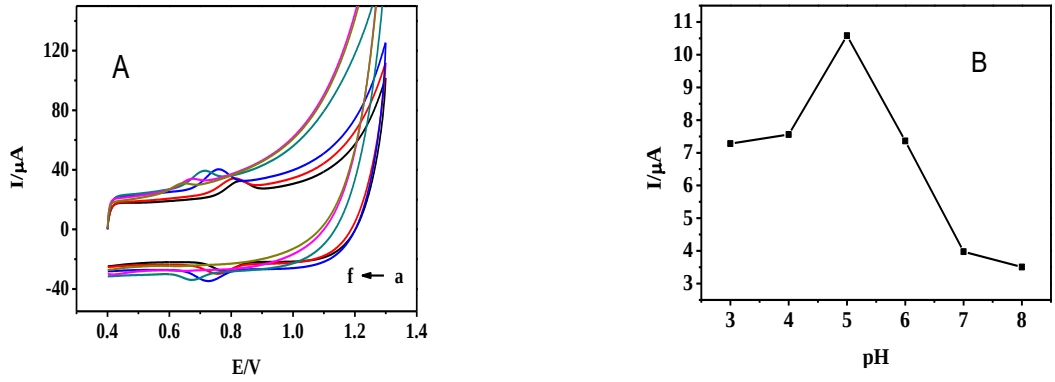

Figure 4 – (A) 5μL SWNTS-Au /GCE in 0.1 mol/L PBS at different pH: a → F (3.0, 4.0, 5.0, 6.0, 7.0, 8.0), the cyclic voltammetry of 50 μmol/L IB was measured in buffer solution (cyclic voltammetry obtained by detection; (B) peak current the change of peak current relative to pH

The peak potential of IB shifted negatively with the increase of pH, while the peak current increased with the increase of pH. When pH exceeded 5.0, the peak current decreased with the increase of pH. Therefore, PBS with pH5.0 was selected as the buffer solution.

Influence of scanning speed

Figure 5 (A) is the cyclic voltammetry diagram of 5μL SWNTS-Au /GCE in PBS buffer solution pH5.0 at different scanning rates. It can be seen from the figure that the peak current of IB increases with the increase of scanning speed, and the peak current is linear with scanning speed in the range of scanning speed 5-500MVS-1. The linear equation is y=0.64+0.118x, and the linear correlation coefficient R=0.9946 (Figure 5B), indicating that the reaction is controlled by surface adsorption.

Influence of interset shadow enrichment time

Figure 6 (A) is the cyclic voltammetry of 5μL SWNTS-Au /GCE buffer solution with 0.1MPBS (pH5.0) at 50μmol/LIB at different enrichment times, and (B) is the curve of peak current with enrichment time .It can be seen from the figure that the electrochemical behavior of the modified electrode is affected by the enrichment time, and the peak current reaches the maximum when the enrichment time is 270s.

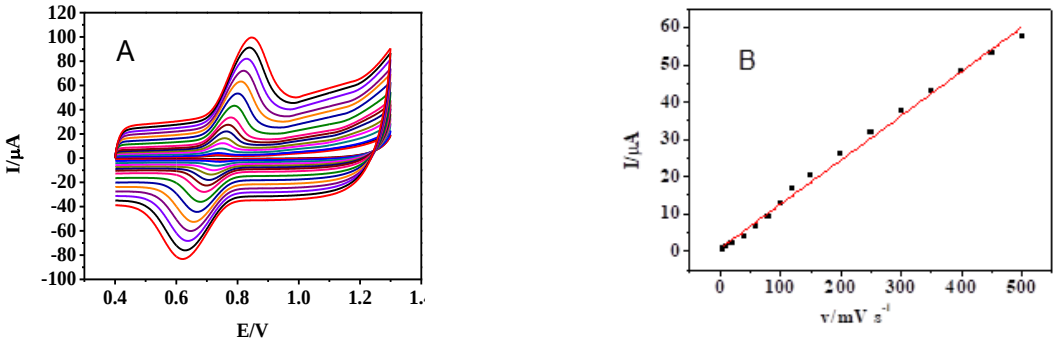

Figure 5 (A) 5μL SWNTS-Au /GCE unscanned cyclic voltammetry of 50 μmol/L IB rows in 0.1M PBS (medium solution, pH 5.0) at different scanning rates from to, and scanning speed from inside to outside:5, 10, 20, 40, 80, 100, 120, 150, 200, 250, 300, 350, 400, 450, 500mV s-1; (B) electrical scanning peak current changes with the scanning speed

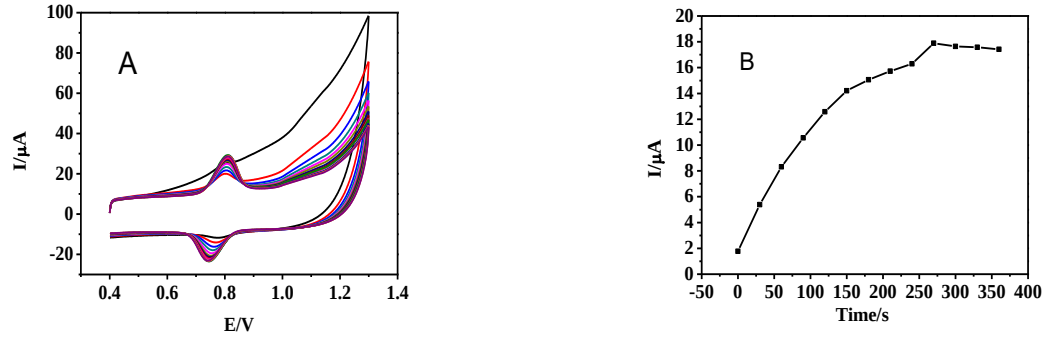

Figure 6(A) – Cyclic voltammetry of 5 μL SWNTS-Au /GCE in 0.1 mol/L PBS (medium solution, pH 5.0) in 50 μmol/L IB with cyclic ampere:, sweep speed: 100 mV S-1 rich from:, enrichment time from inside to outside: Peak current of 0, 30, 60, 90, 120, 180, 210, 240, 270 s; B peak flow rich curve with enrichment time curve

When the enrichment time exceeds 270s, the peak current tends to be stable, indicating that the IB adsorbed on the electrode surface reaches saturation. Therefore, the optimal enrichment time selected in the experiment is 270s.

Sample analysis and recovery experiment

In order to evaluate the practical application value of the method, the modified electrode was used to measure IB in real fruit grains and the experiment of standard recovery was carried out. In table, the concentration of IB measured was 8.5mmol/L, which was close to the 6.2mmol/L indicated by IB in the purchased real fruit. It can be used for the detection and quality control of food additives.

Table – ***Determination of IB in carbonated beverages (Yanzhong)***

Sample, μL	Add, μmol/L	Found, μmol/L	Recovery
5	10	14.51	96.73%
	20	24.44	97.76%
	30	35.16	100.05%
	40	44.75	99.44%
	50	55.19	100.03%

Conclusion

In this paper, a novel single-walled carbon nanotube/gold nanocomposite was prepared using single-walled carbon nanotube as the starting point. The electrochemical analysis of the prepared nanocomposite was carried out, and the highly sensitive IB electrochemical sensor was constructed by modification of the nanocomposite onto the electrode surface.

1. Compared with gold nanomaterials, single-wall carbon nanotube-gold nanocomposites are more sensitive to IB;

2. Comparing the current response signals of different pH values, 0.1mol/LPBS solution with pH=5 was selected as the test solution;

3. When the enrichment time reaches 270s, the IB adsorbed on the electrode surface tends to be saturated, so the optimal enrichment time is 270s.The above experiments show that the nanocomposite material has a good electrocatalytic ability for IB, and the detection limit (3S/N) is 0.02μmol/L. The modified electrode has a short response time, a wide linear range, a low detection limit, good selectivity and high stability.

References

1. Cheng, H M, Li, F, Su, G et al. (1998). Large-scale and low-cost IBnthesis of single-walled carbon nanotubes by the catalytic pyrolysis of hydrocarbons. *[J]. Appl Phys Lett,* 72(25), 3282–3284.

2. Journet, M. K., Bernier, C. (1997). Large scale production of sing lewalled carbon nanotubes by the electricarc technique. *[J]. Nature,* 388, 756.

3. Iijima, S. (1991). Helical microtubules of graphitic carbon. *[J]. Nature,* 354–356.

4. Thess, A., Lee, R., Nikolaev, P., Dai, H. (1996). Crystalline Ropes of Metallic Carbon Nanotubes. *[J]. Science,* 273(5274), 483–487.

5. Colomer, J. F., Bister, G., Willems, I. et al. (1999). Ibn thesis of single-welled carbon nanotubes catalytic decomposition of hydrocarbons. *[J]. Chem. Commun.,* 1343–1344.

6. Sun, L. F., Mao, J. M., Pan, z. W. et al. (1999). Growth of straight nanotubes with a cobalt-nickel catalyst by chemical vapor deposition. *[J]. Appl. Phys. Lett.,* 74, 644–646.

7. Chang, T. (2008). Dominoes in Carbon Nanotubes. *[J]. Phys. Rev. Lett.,* 101(17), 5501.

8. Terrones, M., Hsu, W. K., Hare, J. P. (1996). Metal particle catalysed production of nanoscale BN structures. *[J]. Chem Phys Lett,* 259, 568-573.

9. Li, Y. L., Yu, Y. D, liang, Y. (1997). A novel method for IBnthesis of carbon nanotubes: Low temperature solid pyrolysis. *[J]. J Mater Res,* 12, 1678–1680.

10. Vander Wal, R L., Ticich, T M., Curtis, V E. (2000). Diffusion flame IBnthesis of single-walled carbon Nanotubes. *J. Chem Phys Lett,* 323(3), 217-223.

11. Conway, B. E. (1999). Electrochemical supercapacitors, scientific fundamentals and technological applications. Kluwer Academic Publishers, Plenum Press, New York, 321, 56–60.

12. Stoller, M. D., Park, S., Zhu, Y., An, J., Ruoff, R. S. (2008). Graphene-based ultracapacitors. *[J]. Nano Lett.,* 8, 3498–3502.

Received 28.11.2021

Ченг Вейхуа
Професія: хімічна інженерія, *orcid.org/0000-0003-2681-2341*
Політехнічний коледж Яньчен, Школа медицини та здоров'я, Китай

ЗЕЛЕНА ПІДГОТОВКА ОДНОСТІННИХ ВУГЛЕВОВИХ НАНОТРУБОК / НАНОКОМПОЗИТІВ ЗОЛОТА ТА ДОСЛІДЖЕННЯ НА ВИЯВЛЕННЯ СИНЬОГО ІНДИГО

Анотація. *Одностінні вуглецеві нанотрубки (ОСВНТ) привертають все більше уваги завдяки своїй унікальній структурі та чудовим властивостям. У цій роботі нанокомпозити з одностінними вуглецевими нанотрубками / золотом (SWNTs / Au) були виготовлені стійким і простим методом, а на основі цих нанокомпозитів створено високочутливий датчик синього кольору індиго. Результати засвідчили, що оптимальний pH синього індиго (ІВ) становив 5, а оптимальний час збагачення становив 270 с. Реакцію контролювали поверхневою адсорбцією. Унікальна особливість цього методу полягає в тому, що в процесі приготування не додаються токсичні відновники або поверхнево-активні речовини, що є екологічно чистим і чутливим до виявлення індиго синього на основі модифікованого нанокомпозитним електродом. У порівнянні з наноматеріалами золота, одностінні вуглецеві нанотрубки-золоті нанокомпозити більш чутливі до синього індиго. Порівнюючи поточні сигнали відповіді різних значень pH, як досліджуваний розчин було вибрано 0,1 моль / LPBS розчин з pH = 5. Коли час збагачення досягає 270 с, ІВ, адсорбований на поверхні електрода, має тенденцію до насичення, тому оптимальний час збагачення становить 270 с. Наведені експерименти доводять, що нанокомпозитний матеріал має добру електрокаталітичну здатність щодо індиго синього, а межа виявлення (3S/N) становить 0,02 мкмоль/л. Модифікований електрод має короткий час відгуку, широкий лінійний діапазон, низьку межу виявлення, добру селективність і високу стабільність.*

Ключові слова: одностінні вуглецеві нанотрубки; наночастинки золота; синій індиго; електрохімічний датчик

Link to the article

APA Cheng, Weihua. (2021). Green preparation of single-walled carbon nanotubes/gold nanocomposites and study on the detection of indigo blue. *Management of Development of Complex Systems*, 48, 160–164. dx.doi.org\10.32347/2412-9933.2021.48.160-164.

ДСТУ Ченг Вейхуа. Зелена підготовка одностінних вуглевових нанотрубок / нанокомпозитів золота та дослідження на виявлення синього індиго. *Управління розвитком складних систем.* Київ, 2021. № 48. С. 160 – 164, dx.doi.org\10.32347/2412-9933.2021.48.160-164.