

Electrodeposition of nickel hexacyanoferrate/layered double hydroxide hybrid film on the gold electrode and its application in the electroanalysis of ascorbic acid



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ABSTRACT

Nickel hexacyanoferrate (NiHCF) and Ni-Al layered double hydroxide (LDH) hybrid film was fabricated on the gold electrode by a two-step electrodeposition method. Varying the strategies of electrodeposition of NiHCF, the hybrid films showed different morphology, composition and electrochemical behavior. As a demonstration, the hybrid film fabricated by using the absorbed Ni^{2+} on Ni-Al LDH modified gold electrode for the next electrodeposition of NiHCF was used for the electroanalysis of ascorbic acid (AA). The analytical performance of the proposed electrode was investigated by cyclic voltammetry and chronoamperometry. The linearity range of AA was from 0.1 to 3.5 mM with a detection limit ($S/N=3$) of 21 μM . The developed method was found to be linear, sensitive, selective, precise and accurate. The applicability of the method was confirmed by the satisfactory results in real sample analysis.

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1. Introduction

Transition metal hexacyanoferrates, an important class of inorganic coordination polymers, have recently attracted great attention for their open zeolite-like structure and interesting magnetic, optical, and electroactive properties [1]. They can be found wide applications in many fields such as ion-sieving membranes [2–4], hydrogen storage [5], magneto-optic devices [6], electroanalysis [7], electrochemical power sources [8,9], and electrochromic display [10,11]. Among the transition metal hexacyanoferrates, nickel hexacyanoferrate (NiHCF) is often selected as a model system due to the reversibility of its redox reactions in various alkali metal cations containing electrolytes [12]. NiHCF has been widely used to modify electrode surfaces as electron transfer mediator for analytical applications and electrochemical biosensors [13,14].

However, the stability and analytical performance of NiHCF and other transition metal hexacyanoferrates modified electrodes need to be further improved [15]. To achieve this aim, many efforts have been dedicated to fabricate the composites containing transition metal hexacyanoferrates. The hybrid can be the composite of hexacyanoferrates with different transition metal [16] or the heterostructure of transition metal hexacyanoferrates with other

kind of materials [17–24]. In terms of the role of the introduced materials, some increase the electrical conductivity of the hybrid, such as carbon nanotubes [17], some act as an immobilization matrix to increase the mechanical properties of the films, such as chitosan [18,19], histidine [20], and some offer both high conductivity and mechanical properties of the hybrid films, such as polyaniline [21,22], polypyrrole [23], and poly(1-naphthol) [24]. It has been shown that the hybrid films are superior to the single transition metal hexacyanoferrate in the stability and electrochemical properties of the electrodes due to their synergetic effect.

On the other hand, layered double hydroxides (LDHs), also well-known as anionic clays or hydrotalcite-like compounds, are a class of important lamella materials. Their structures are based on that of magnesium hydroxide in which divalent metal cations have been partly replaced by trivalent ions resulting in positive net charge in the layer which is compensated by interlayer anion along with water molecules [25,26]. In recent years, LDHs have attracted growing interest for their properties such as adjustable composition, anion exchange and adsorption ability [27–35]. In particular, LDHs are widely used as active electrochemical material in heterostructures [36–38]. Therefore, the introduction of LDHs into NiHCF as electrode materials may not only improve the stability of the modified electrode for LDHs' good adsorption ability but also enhance the electrocatalysis property for their possible synergetic effect. In addition, it is interesting to investigate the methods for preparing the hybrid of LDHs and NiHCF by utilizing the special structure and

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properties of LDHs. For example, NiHCF can be intercalated into the interlayer of the LDHs via anion exchange [39]. Certainly, this method is not preferred for its slow anion exchange rate.

In the present work, the hybrid film of NiHCF and Ni-Al LDHs was fabricated by a facile two-step electrochemical deposition method. Considering that the composition of NiHCF is prone to vary with fabrication process, different strategies were used in the step of electrodepositing NiHCF and the corresponding hybrids were compared in the respect of composition, morphology, and electrochemical behavior. To verify the applicability of Ni-Al LDH/NiHCF hybrid film, ascorbic acid (AA) was electrochemically determined at Ni-Al LDH/NiHCF hybrid film modified Au electrode.

2. Experimental

2.1. Chemicals and apparatus

$\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, $\text{K}_3[\text{Fe}(\text{CN})_6]$, NaNO_3 , KNO_3 and ascorbic acid (AA) were purchased from Chemical Reagent Company of Shanghai. All chemical reagents were of analytical grade and used without further purification. Milli-Q ultrapure water (Millipore, $\geq 18.2 \text{ M}\Omega \text{ cm}$) was used throughout.

The electrochemical measurements were carried out with CHI660C electrochemical workstation (Chenhua Instruments, Shanghai, China). A conventional three-electrode system was employed comprising a modified Au electrode ($\phi = 2 \text{ mm}$) as working electrode, a platinum wire as counter electrode, and an aqueous saturated calomel electrode (SCE) as reference electrode. All electrochemical experiments were performed in solutions deaerated by pure nitrogen at room temperature. Scanning electron microscopy (SEM) was conducted by EDS-integrated Hitachi S-4800 (Hitachi, Japan) for surface morphology observations and surface elemental composition analysis.

2.2. Electrodeposition of Ni-Al LDHs on the Au electrode

At first, the Au electrode was carefully polished with alumina powder (the grain size in the order of 0.3, 0.1 and 0.05 μm) on a chamois leather and then thoroughly cleaned ultrasonically in water. Prior to the modification, the Au electrode was electrochemically activated in 0.5 M H_2SO_4 solution by cycling the potential from 0 to 1.6 V at a scan rate of 50 mV s^{-1} for 20 circles [40].

The construction of Ni-Al LDH films on the activated Au electrode was carried out by a cathodic reduction in a solution (pH = 3) containing 0.12 M $\text{Ni}(\text{NO}_3)_2$, 0.04 M $\text{Al}(\text{NO}_3)_3$ and 0.15 M KNO_3 at a potential of -0.9 V vs. SCE. And the deposition time was limited to 30 s [41]. The modified electrode was denoted as LDH/Au electrode.

2.3. Electrosynthesis of NiHCF on LDH/Au electrode

The NiHCF was synthesized on the LDH/Au electrode by three different methods:

(1) The cyclic voltammetry (CV) was performed on the LDH/Au electrode in the solution of 1.0 mM $\text{K}_3[\text{Fe}(\text{CN})_6]$ and 1.0 M KNO_3 with the potential from 0 to 0.8 V at a scan rate of 100 mV s^{-1} for 20 circles. The LDH/Au electrode was pre-cleaned thoroughly with water to remove the absorbed- Ni^{2+} .

(2) The CV was performed as method (1), but the LDH/Au electrode was not pre-cleaned.

(3) The CV was performed on the LDH/Au electrode in the solution of 1.0 mM $\text{K}_3[\text{Fe}(\text{CN})_6]$, 1.0 mM $\text{Ni}(\text{NO}_3)_2$ and 1.0 M KNO_3 with the potential from 0 to 0.8 V at a scan rate of 100 mV s^{-1} for 20 circles. The LDH/Au electrode was pre-cleaned as method (1).

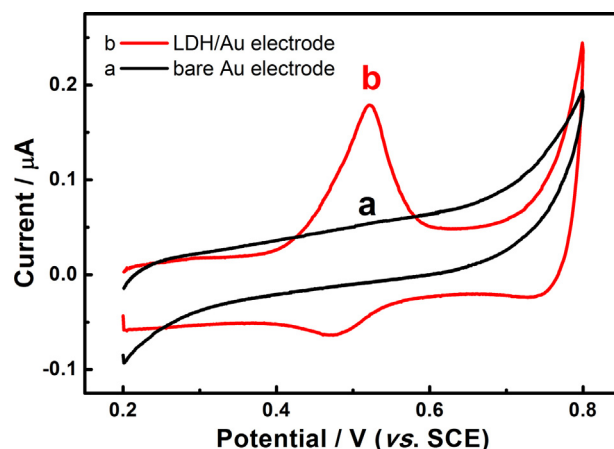


Fig. 1. Cyclic voltammograms of (a) bare Au electrode and (b) LDH/Au electrode in 0.1 M NaOH at a scan rate of 50 mV s^{-1} .

2.4. Determination of AA in vitamin C tablets

The vitamin C tablets were purchased from Nanjing Baijingu Pharmaceutical Co., Ltd. The determination process is described as follows.

0.1530 g of vitamin C was dissolved in water and then diluted to 10 mL in a volumetric flask. Quantitative determination of AA was carried out amperometrically in a PBS solution (pH 7.0, containing 1.0 M NaNO_3) and the potential of the electrode was kept constant at +0.35 V. The response was recorded by adding 80 μL sample out of the volumetric flask to the electrochemical cell with 5 ml PBS solution.

3. Results and discussion

3.1. Electrochemical characterization of Ni-Al LDH on Au electrode

Electrochemically deposited Ni-Al LDH on Au electrode was characterized by CV. In contrast to Au electrode which did not exhibit any electrochemical response (Fig. 1, curve a), LDH/Au electrode displayed a pair of redox peaks within a potential range from 0.4 to 0.6 V, which can be ascribed to the reversible one electron redox of $\text{Ni}^{\text{III}}/\text{Ni}^{\text{II}}$. The result is in good agreement with literature reports [41]. Therefore Ni-Al LDHs were successfully prepared on Au electrode.

3.2. Electrodeposition of NiHCF on LDH/Au electrode

The composition and structure of NiHCF strongly depends on the method of preparation [42]. In this contribution, the NiHCF was fabricated on LDH/Au electrode by three different strategies and their electrochemical behavior was discussed as follows.

Firstly, considering that there existed Ni^{2+} on the skeleton of LDHs, an interesting question was pointed out. That was whether these confined Ni^{2+} could cooperate with $[\text{Fe}(\text{CN})_6]^{3-}$ or $[\text{Fe}(\text{CN})_6]^{4-}$ and generate nickel hexacyanoferrate directly on the sheets of LDHs. Fig. 2A shows the cyclic voltammograms (CVs) recorded during the potential cycling of LDH/Au electrode in the solution containing 1.0 mM $\text{K}_3[\text{Fe}(\text{CN})_6]$ and 1.0 M KNO_3 . It can be seen that a pair of reversible redox peaks at 0.15 V and 0.24 V displayed in the curves and the peak currents had no change within 20 circles of potential cycling. The results showed that the process was only the reversible redox of $\text{K}_3[\text{Fe}(\text{CN})_6]$ and no NiHCF was deposited on the LDH/Au electrode. Namely, it was unsuccessful for Ni^{2+} on the skeleton of LDH to cooperate with $[\text{Fe}(\text{CN})_6]^{3-}$ or

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