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Exploring more for magnetic nanoparticles: separation/concentration-signal-generation in one method for biosensing

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Electrochemical biosensors using magnetic nanoparticles (MNPs) for separation and enrichment significantly improve detection performance and simplify the operation. However, these studies mainly focused on the magnetic properties of MNPs, leaving their roles as signal labels rarely exploited. Here, by exploring electrochemical properties of MNPs, we propose a novel magnetic-separation/collection-signal-amplification in-one electrochemical biosensing strategy for chloramphenicol (CAP) detection. In the design, not only the magnetic properties of MNPs were fully explored, but also a new way to generate a signal through the electrochemical conversion (ECC) of MNPs to electrochemically active Prussian blue (PB) was developed, thus the generation and amplification of signals without any additional labels were realized. The biosensors exhibited satisfactory performance for the detection of CAP with a detection limit down to 1 ng mL^{-1}, the detection range is from 1 ng mL^{-1} to 10000 ng mL^{-1} as well as good specificity and stability. Moreover, the biosensors worked well for the detection of CAP in milk and river-water samples.

Introduction

Antibiotics residue has been one of the most serious threatens for food safety, and the rapid detection of antibiotics is needed but remains challenging.

For biosensing, the separation/concentration of targets from samples and the following labelling procedure are generally independent, which increases the complexity of operations and the cost.

Method

CAP aptamer hybridized with its complementary DNA chain (S1) was modified on MNPs (MNPs-Apt/S1). In the presence of CAP, aptamer combines with CAP and detaches from MNPs. Subsequently, the MNPs-S1 composite was conjugated to Au electrode through the DNA hybridization between the S1 probe and a strand of complementary DNA (S2), followed by ECC of MNPs labels to electrochemically active PB. Finally, the anodic and cathodic currents of PB were taken as signal.

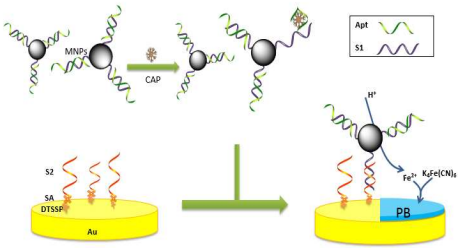

Fig. 1. Illustration of the procedures and detection mechanism of the proposed biosensor.

Results and Discussion

Through quantifying the residual aptamer in supernatant, we found out the successful conjugation of aptamer on MNPs (Fig. 2A), as also demonstrated by FTIR (Fig. 2B).

The successful layer-by-layer modification of the electrode was monitored by cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) (Fig. 3).

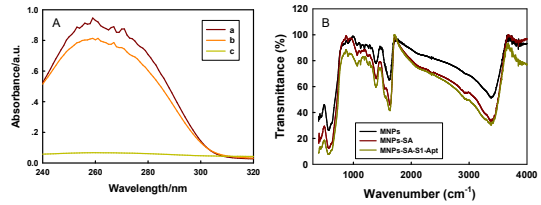

Fig. 2. (A) UV-vis spectra of the supernatants of the solutions before (a) and after (b) the aptamer conjugation, and the control (c). (B) FTIR spectra of three kinds of magnetic composites.

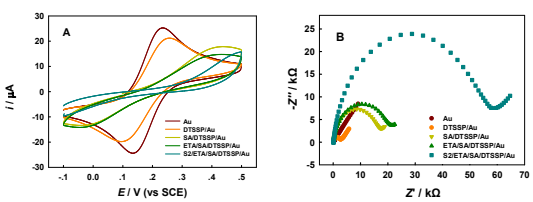

Fig. 3. CV (A) and EIS (B) of the layer-by-layer modification of electrodes.

During the ECC process, the applied high potential oxidized the species on the surface of the electrode and generated oxygen bubbles, both leading to the refreshment of the electrode surface, which was significantly beneficial to the amperometric signal readout, as proved by the CV and EIS (Fig. 4).

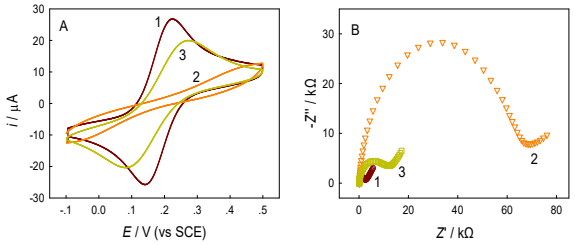

Fig. 4. CV (A) and EIS (B) of a bare gold electrode, MNPs-Apt-conjugated gold electrodes before and after ECC process (curves 1, 2 and 3 represent Au, Au-MNPs-Apt and Au-MNPs-Apt after ECC, respectively).

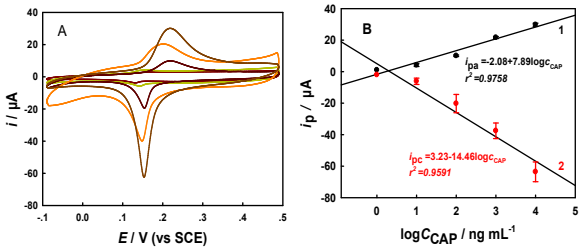

Fig. 5. (A) CV responses of biosensors to CAP at different concentrations (from top to bottom: control, 1, 10, 100, 1000, 10000 ng mL^{-1}). (B) The calibration plot of oxidative (1) and reductive (2) peak currents to CAP concentrations.

Under optimized conditions, the biosensors exhibited satisfactory performance for the detection of CAP with a detection limit down to 1 ng mL^{-1}, the detection range is from 1 ng mL^{-1} to 10000 ng mL^{-1} (Fig. 5) as well as good specificity and stability. Moreover, the biosensors worked well for the detection of CAP in milk and river-water samples.

Conclusion

We have developed a novel magnetic-separation/collection-signal-amplification in-one electrochemical biosensing strategy for CAP detection with satisfactory performance.

The proposed method might create new direction for the label-free, rapid and sensitive biosensing technologies, and open up new opportunity for the simplification of electrochemical instrument for various applications.

Reference

Q. Zhang, L.Y. Li, Z.H. Qiao, C.Y. Lei, Y.C. Fu, Q.J. Xie, S.Z. Yao, Y.B. Li, Y.B. Ying, Anal. Chem., 2017, 89, 12145.

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