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ACCEPTED MANUSCRIPT

Enzyme-triggered formation of enzyme-tyramine concatamers on nanogold-functionalized dendrimer for impedimetric detection of Hg(II) with sensitivity enhancement

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ABSTRACT

A new impedimetric sensing strategy based on enzyme-triggered formation of enzyme-tyramine concatamers on the nanogold-functionalized poly(amidoamine) (PAMAM) dendrimer was designed for sensitive detection of mercury(II) (Hg²⁺) ion, coupling with enzymatic biocatalytic precipitation towards 4-choloro-1-naphthol (4-CN) on thymine (T)-rich single-stranded DNA₁-modified electrode. Initially, nanogold-decorated PAMAM dendrimer (AuNP-PAMAM) was synthesized by the in-situ reduction method, and then functionalized with horseradish peroxidase (HRP) and another T-rich oligomer (DNA₂). Upon target Hg²⁺ introduction, probe DNA₂ on the AuNP-PAMAM bound to the DNA₁ on the electrode owing to the T-Hg²⁺-T coordination chemistry between the two DNA strands. Accompanying the AuNP-PAMAM, the carried HRP could trigger the formation of HRP-tyramine concatamer via the classical tyramine signal amplification strategy in the presence of HRP-tyramine conjugates and hydrogen peroxide. The concatenated HRP molecules in the concatamer catalyzed the 4-CN oxidation to produce an insoluble precipitation on the electrode, thereby resulting in the local alteration in the conductivity. Under optimal conditions, two signal-generation tags including HRP-AuNP-DNA2 and HRP-AuNP-PAMAM-DNA2 with or without tyramine signal amplification strategy (i.e., four schemes) were used for impedimetric detection of target Hg²⁺ on the basis of the same assay format. A low detection limit (LOD) of 0.4 and a wide dynamic working range of 0.001 - 100 nM Hg²⁺ by using рM

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