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A novel strategy based on DNAzyme for electrochemiluminescence detection of Pb(II) with P-GO@QDs for signal amplification

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Abstract: A novel, simple, and versatile electrochemiluminescence (ECL) sensing platform for Pb(II) ions was developed based on DNAzyme with peroxidase-like activity to catalyze the reduction of hydrogen peroxide. Poly (diallyldimethylammonium chloride) (PDDA) with positive charge was adsorbed onto the surface of graphene oxide (GO) and then CdS quantum dots (QDs) with negative charge were enriched to the surface of GO by electrostatic interaction between CdS QDs and PDDA to prepare a composite (denoted as P-GO@QDs) which was immobilized on the surface of a glassy carbon electrode (GCE). When hydrogen peroxide was used as coreactant CdS QDs can produce ECL signal. G-rich DNA strand, *i.e.* T30695, can combine with Pb (II) ions to form a stable parallel G-quadruplex. The G-quadruplex can further combine with hemin to form DNAzyme which have catalysis towards the reduction of hydrogen peroxide causing the decrease of ECL signal. Amino-modified T30695 was attached to P-GO@QDs composite by the reaction between the amino group of T30695 and carboxyl group of CdS QDs. The ECL signal of the biosensor was linearly dependent on the logarithm of Pb(II) concentration from 1.0×10^{-14} to 1.0×10^{-11} M with a detection limit of 9 fM. An excellent performance of ECL biosensor indicated that it is promising for Pb(II) detection in real samples.

Keywords: DNAzyme; P-GO@QDs; lead ion; hydrogen peroxide

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1. Introduction

Lead ions that were toxic possess serious harm to ecosystem and human health, such as, nervous system, digestive system, blood system, especially for children whose brain system are in the sensitive period. Until now, many methods are used for the detection of Pb (II) ions, such as atomic absorption spectrometry [1, 2], inductively coupled plasma mass spectrometry [3, 4], fluorescence spectrometry[5] electrochemistry[6, 7] and electrochemiluminescence (ECL) [8]. Owing to the rapid response and simple use, electrochemistry and ECL have often been employed for the determination of lead ion.

In 1998, Sen et al reported that G-quadruplex/Hemin (DNAzyme) possessed peroxidase-like activity which consists of G-rich DNA sequence and some metal ions to form parallel, antiparallel or heterozygous G-quadruplex and embed hemin acting as cofactor [9]. T30695 containing G-rich DNA sequence could make up stable parallel G-quadruplex with Pb (II) even in the existence of K(I) and have a stronger affinity to hemin than other forms of G-quadruplex [10]. Li et al. constructed electrochemical sensor for Pb (II) based on DNAzyme with a detection limit of 5.0×10^{-10} M [7].

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