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Poly-L-lysine-modified boron-doped diamond electrodes for the amperometric detection of nucleic acid bases

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ABSTRACT

Boron-doped diamond (BDD) is a very promising supporting material used in the construction of biosensors for molecular recognition. The direct immobilization of structurally-organized huge molecules, such as poly-L-lysine (PLL) provides the possibility of determining organic molecules, e.g. nucleic acid bases (e.g. adenine, guanine) or peptides and proteins. This paper describes the direct method for chemical and electrochemical modification of the BDD electrode surface with organic molecules, including the potential application of such modified electrode for detecting selected nucleic acid bases, e.g. adenine and guanine.

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1. Introduction

In recent years, different kinds of chemically modified electrodes have been applied to investigate the direct electrochemistry of nucleic acids (NA) [1–3]. Both miniaturization of the electrochemical measuring systems and the application of advanced electrode materials belong to the key steps in the development of devices applicable to practical analyses such as genetic assays, DNA diagnostics, DNA damage monitoring, etc. [4].

Oliveira-Brett et al. [5,6] have firstly reported electrochemical oxidation mechanism of guanine and adenine at glassy carbon microelectrode and cyclic and differential pulse voltammetry. Furthermore, they demonstrated also application of ultrasound in combination with differential pulse voltammetry in a reliable analytical procedure for thymine and cytosine measurements avoiding electrode fouling and maintaining the electrode characteristics [7]. Finally, Oliveira-Brett's team [8] showed for the first time that equimolar mixtures of all DNA bases, nucleosides, and nucleotides could be quantified by differential pulse voltammetry

Teh et al. studied the electrochemistry of adenine on a sol–gel carbon composite electrode by differential pulse stripping adsorption voltammetry, a method useful for simultaneous analysis of adenine and guanine from denatured DNA [1]. Wu et al. investigated the direct electrochemistry of DNA, guanine and adenine at a nanostructure film-modified electrode, which resulted in the development of a sensitive electrochemical technique for measuring native DNA [9]. Moreover, Sun et al. [10] applied the carbon ionic liquid electrode for monitoring the direct electrooxidation behaviors of adenine and guanine in the thermally denatured single-stranded DNA. The simultaneous detection of guanine (G), adenine (A), thymine (T), and cytosine (C) was achieved at a multiwalled carbon nanotube (MWCNT)/choline (Ch) monolayer-modified glassy carbon electrode (GCE), with its output decreasing over time [11]. Wang et al. [12] developed reliable method based on adsorptive stripping at an electrochemically pre-treated glassy carbon electrode (GCE) for simultaneous or individual determination of G and A in DNA. Furthermore, the Nafion composite [13], graphene-COOH modified glassy carbon electrode (GCE) [14], multi-wall carbon nanotubes (MWNT) [15], b-cyclodextrin/MWNTs [16] or the porous structure of overoxidized polypyrrole/graphene electrochemically coated onto GCE [17] have been successfully utilized as efficient electrode material for the quantitative detection of the components of DNA and RNA. Moreover, a remarkable improvement in the kinetics of the electron transfer for guanine and adenine was shown by Wang et al. [18] on the surface of the silver decorated graphene quantum dots (AgNPs/GQDs) modified GC electrode, by shifting negatively in

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anodic peak potentials and significantly increasing the anodic peak current. Nevertheless, the fabrication of composite materials requires several separate steps or electrochemical pre-treatment, which creates difficulties with regard to the repeatable electrode production and scaling of this process.

The remarkable electrochemical properties of boron-doped diamond (BDD), e.g. low background current, wide potential window, high stability combined with biocompatibility and chemical inertness makes it a very promising material for the third-generation biosensors [19,20]. The fast response, sensitivity and selectivity of biosensors based on BBD electrodes aroused great interest among scientists [3, 21–26]. The BDD electrodes are mostly prepared by the microwave plasma enhanced-chemical vapor deposition (MW PE CVD) method, while the as-deposited surfaces of BDD are hydrogen terminated (H-BDD).

Hason et al. successfully applied the anodic stripping determination of purine bases in acid-hydrolysed DNA, which involved electrochemically controlled accumulation of purine-Cu(I) complexes [27]. The significant sensitivity improvements in DNA electrochemical sensing were obtained using vertically aligned diamond nanowires fabricated by reactive ion etching (RIE) with O_2 [28].

Recently, Prado et al. have demonstrated the possibility of detecting underivatized nucleic acids at the boron-doped diamond electrodes using cyclic voltammetry and square wave voltammetry [29]. Ivandini et al. [30] reported the importance of surface termination and ionic strength of electrolyte for electrochemical oxidation of nucleic acids at the diamond electrode. The authors have shown the linearity of current at concentrations between 0.1 and 8 μg mL^{-1} for both guanine and adenine residues at as-deposited BDD. As-deposited diamond film with predominantly hydrogen-terminated surface demonstrated superior performance in comparison to oxygen-terminated diamond in terms of sensitivity, but this feature was present only within the limited range of potential and pH [30].

The application of a bare BDD electrode for the determination of G and A (purine DNA bases) in biological samples was reported by Svorc et al. [31]. Low LOD values were obtained with the previously reported electrochemical methods for G and A detection. Compared to other electrodes, the bare BDD electrodes do not require a rather tedious modification process, however, they suffer from the interfering signals caused by common components such as non-target proteins or polysaccharides.

Different terminations on the surface of BDD electrode can be obtained by applying a fixed potential and pre-treatment [32,33]. The detection of a specific DNA nucleotide is strongly influenced by the presence of different functional groups on the BDD electrode surface [34,35].

Thus, there is a need to modify the surface of BDD electrodes because of the insufficiency of chemically reactive groups, which precludes the attachment of organic compounds to the electrode surface [36]. The most widely used approach to functionalizing diamond film surface with organic compounds is an amine functionality introduced to the surface. So far, several amination methods of diamond surface have been proposed [36–39]. In general, they require: (I) etching by NH_3 plasma in a specific reactor [39], (II) chemical modification with (3-aminopropyl) triethoxysilane [40], (III) photochemical reaction of amino molecules containing a vinyl group [41], or (IV) diazonium functionalization [42–44]. It is apparent that there is a need to develop a simple procedure with better sensitivity, and to achieve a good linear sensing range. Electropolymerized or chemically polymerized amine polymers seem to be attractive candidates to fulfill the aforementioned aims.

Gu et al. [45] demonstrated a one-step chemical modification of BDD by the thin polyaniline/poly (acrylic acid) (PANI/PAA) composite polymer film. The procedure did not show any non-specific DNA adsorption, and DNA probes immobilized on the BDD substrates were stable and selective to DNA sensing, while the oxidation peak potentials were lower than those reported for the oxidation of guanine and adenine on other carbon-based electrodes [22,23] and bare BDD [3,4]. However, the electroactivity (i.e. redox behavior) of polyaniline is strongly dependent on the pH of electrolyte, and is greatly weakened at pH > 4 [46]. Su et al. have fabricated the functionalized carboxyl graphene oxide (GO–COOH) at a glassy carbon electrode (GCE) and L-lysine was electropolymerized on the GO–COOH modified GCE by cyclic voltammetry (CV) [47].

Studies on the modification of BDD electrodes with poly-L-lysine, aimed at the detection of nucleotides, have not been published until now. Therefore, we report a simple and convenient one-step method to modify the surface of boron-doped diamond (BDD) by taking advantage of the conducting nature of BDD due to the presence of electropolymerized and chemically polymerized poly-L-lysine (BDD/PLL). The electrostatic interactions between the positively charged $–NH_2$ groups of PLL and negatively charged GO–COOH stabilize the thin film [48].

All prepared electrodes were characterized by various electrochemical systems such as $[Fe(CN)_6]^{3-/4-}$, Fe^{2+}/Fe^{3+} and (Q/H_2Q) in order to determine their electrochemical behavior. Furthermore, differential pulse voltammetry (DPV) was successfully utilized to investigate the interaction between BDD/PLL electrodes and adenine and guanine. This technique is suitable for studying biological systems since it is fast and highly sensitive.

One advantage of pulse techniques is that they are characterized by much better signal-to-noise ratio and, in many cases, by greater selectivity than steady-state techniques [49,50]. Moreover, the prepared PLL-modified BDD electrode was analyzed by means of X-Ray photoelectron spectroscopy (XPS) and Scanning Electron Microscopy (SEM).

2. Experimental

2.1. Si/BDD electrode deposition

BDD electrodes were deposited in an MW PA CVD system (Seki Technotron AX5400S, Japan) on p-type Si substrates with (111) orientation. Substrates were cleaned by sonication in acetone and 2-propanol for 5 min in each solvent. Next, the substrates were seeded by means of spin-coating in a nanodiamond suspension (crystallite size of 5–10 nm), and spun three times for 60 s at 4000 rpm [51]. The temperature of heating stage was kept at 700 °C during the deposition process. In the first step of the procedure, the substrates were etched in hydrogen plasma for 1 min. The optimized power of microwave plasma for diamond synthesis was kept at 1300 W. Excited plasma was ignited by microwave radiation (2.45 GHz). The total flow of gas mixture, containing 1% of the molar ratio of CH_4–H_2, was kept at 300 sccm. All samples were doped by using diborane (B_2H_6) dopant precursor; the [B]/[C] ratio in the plasma was 10,000 ppm (BDD10). The reactor chamber was evacuated to a base pressure of about 10^{-6} Torr, while the process pressure was kept at 50 Torr. The time of polycrystalline layer growth was 6 h, which resulted in the thickness of the deposited films of approx. 2 μm.

The four-step pre-treatment of the deposited Si/BDD electrodes was applied to obtain H-terminated surface and etch sp^2 phase impurities, as reported elsewhere [52–54]. For all Si/BDD samples, the diamond surface was cleaned with acids and hydrogen plasma. First, metallic impurities were dissolved in hot aqua regia (HNO_3: HCl/1:3), followed by the removal of organic impurities with hot “piranha” solution (H_2O_2: H_2SO_4/1:3) at 90 °C. Microwave hydrogen plasma treatment was performed using 1000 W of microwave power and 300 sccm of hydrogen gas flow for 10 min. Thus, the resulting BDD surface was predominantly hydrogen-terminated [53,54].

2.2. Si/BDD electrode modification with poly-L-lysine (PLL)

2.2.1. Reagents

Guanine, adenine and poly-L-lysine solution 0.1% (w/v in water) (PLL, MW 150 000–30 000) were purchased from Sigma–Aldrich and used without further purification. All other chemicals were of analytical

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