

Cyclic voltammetric response of boron-doped homoepitaxially grown single crystal and polycrystalline CVD diamond

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Abstract

Homoepitaxial single crystal and polycrystalline boron-doped CVD diamond films were grown by a microwave plasma process using a methane and hydrogen gas mixture at a pressure of 33 Torr over a Type IIa (orientation: 100) and mechanically damaged, cleaned molybdenum substrate, respectively. Doped diamond was grown on both substrates in a single experiment to maintain constant doping levels in the as-deposited films. Boron-doping of diamond was achieved in situ by using a solid disc source of B_2O_3 . Doped diamond was evaluated for background response in 0.5 M NaCl and 0.5 M HCl solutions by cyclic voltammetry. We have observed a wide potential range over which negligible background current was observed for polycrystalline and single crystal diamond. Decomposition of water occurs electrochemically and evolves O_2 during positive (anodic) polarization and H_2 evolution during negative (cathodic) polarization. All experimental measurements were corrected for uncompensated IR drop and have been compared with the results obtained without the correction for uncompensated IR drop. Scanning electron microscopy has been used to study the morphology of diamond before and after electrochemical characterization and we conclude that the morphology was unchanged. We have investigated the redox kinetics of ferri-ferrocyanide ($Fe(CN)_6^{3-} + e^- \rightleftharpoons Fe(CN)_6^{4-}$) in 0.5 M NaCl solution at the homoepitaxial boron-doped diamond electrode. The reaction is reversible at the diamond electrode, but the redox kinetics is sluggish. The heterogeneous electron transfer rate constant has been determined using the experimental data and COOL algorithm. The rate constant was found to be from 6.62×10^{-5} to 1.41×10^{-4} cm s⁻¹ and the kinetics were considered to be sluggish at the diamond electrode/solution interface. © 1998 Published by Elsevier Science S.A. All rights reserved.

Keywords: Cyclic voltammetry; Boron-doped polycrystalline diamond; Boron-doped single crystal diamond; Ferri-ferrocyanide; Microwave plasma

1. Introduction

Diamond is a unique material for innumerable applications because of its unusual combination of physical and chemical properties [1]. Several potential applications can be anticipated for diamond in electronics, optics, protective corrosion resistant coatings, electrochemical sensors, toxic metal ion detection, detection of neurotransmitters, electroanalytical chemistry.

Glassy carbon, highly oriented pyrolytic graphite (HOPG), platinum (Pt) and the dropping mercury electrodes (DME), carbon paste, carbon nanotube,

graphite, gold (Au), silver (Ag) are the materials of general interest for electroanalysis, since they all have a large usable potential range in acidic solutions [2]. Glassy carbon and other solid electrodes will normally be deactivated when they are exposed to electrolyte solutions for a long time. Glassy carbon may be reactivated by using several approaches, such as mechanical and electrochemical polishing, plasma, heat or annealing treatments. Fagan et al. [3] have described the activation of glassy carbon electrodes using vacuum heat treatments and the references therein described other activation procedures. Since HOPG is very soft, Pt and Au are very expensive and Hg is considered as toxic, a search for alternative electrode materials for electroanalytical applications is required.

Cyclic voltammetry is the most useful electroanalytical technique for the study of electroactive species and it is often the first experiment performed in an electro-

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chemical evaluation of an electrode surface. The redox reactions usually provide information about the concentration, reaction mechanism, kinetics, chemical status and other behavior of the species in solution. The working electrode plays a critical role in the electrochemical experiments. Reactions of interest take place at the working electrode and that is usually constructed of chemically inert and possibly catalytic material.

There have been some reports on the characterization of diamond films by electrochemical techniques. Ramesham et al. [4] have demonstrated the growth of polycrystalline diamond over the glassy carbon and graphite electrode materials to employ in applied electrochemistry and electroanalytical chemistry. Swain and Ramesham [5], Swain [6], Alehashem et al. [7], Zhu et al. [8] have reported the cyclic voltammetric behavior of ferri-ferrocyanide using boron-doped diamond films. Ramesham and Rose [9] have recently shown that the diamond electrode material is stable over a wide potential range for the decomposition of H_2O to occur and evolve hydrogen and oxygen during cathodic and anodic polarization, respectively in various electrolytes. Miller et al. [10] have reported the cyclic voltammetric behavior of ferri/ferrocyanide using cobalt ion implanted diamond electrode. Ramesham and Rose [11,12] have reported the electrochemical behavior of doped-polycrystalline diamond in 0.5 M NaCl solution using Electrochemical AC impedance spectroscopy and DC polarization techniques. Voltammetric features for Ce^{3+}/Ce^{4+} were observed at the diamond electrode and redox process was found to be completely irreversible [13].

Good quality diamond films (rough polycrystalline and smooth single crystal quality) have been grown using less than 1% of methane in hydrogen at a pressure of 30–40 Torr [14]. Boron-doped diamond film is electrically conducting [15,16] and therefore, its cyclic voltammetric behavior is of significant practical interest in the field of electroanalytical chemistry. In this paper we will demonstrate the similarity of electrochemical voltammetric response for homoepitaxial and heteroepitaxial CVD diamond films by cyclic voltammetry in 0.5 M NaCl and 0.5 M HCl solutions. To our knowledge this is the first report on the comparison of electrochemical voltammetric response for polycrystalline and single crystal diamond. We will also report the redox kinetic behavior of ferri-ferrocyanide at the boron-doped single crystal diamond electrode in 0.5 M NaCl supporting electrolyte.

2. Experimental details

Surface damaging is a necessary process to nucleate CVD diamond on non-diamond substrates. Molybdenum substrates have been mechanically pretreated with

Table 1

Experimental parameters to deposit very low resistive boron-doped CVD diamond by microwave plasma

(a) Mechanically damaged and cleaned molybdenum substrate	
Pressure (Torr)	34.3 (~4.562 kPa)
H_2 flow (sccm)	500
CH_4 flow rate (sccm)	3.6
% CH_4 in CH_4+H_2	0.71
Substrate temperature ($^{\circ}C$)	894–919
Substrate area (cm^2)	4.4
Growth time (h)	30.5
Growth rate ($\mu m h^{-1}$)	0.5–1
Forward power (W)	750
Reflected power (W)	<2
Substrate base (area)	B_2O_3 disk (~9 cm^2)
(b) Type IIa (100) substrate	
Pressure (Torr)	32.9 (~4.376 kPa)
H_2 flow (sccm)	500
CH_4 flow rate (sccm)	3.6
% CH_4 in CH_4+H_2	0.71
Substrate temperature ($^{\circ}C$)	894–919
Substrate area (cm^2)	0.0231
Growth time (h)	30.5
Growth rate ($\mu m h^{-1}$)	did not measure
Forward power (W)	750
Reflected power (W)	<2
Base for type IIa (area)	CVD Diamond/Mo (4.4 cm^2)
Base for CVD diamond/Mo	B_2O_3 disk (~9 cm^2)

0.25–60 μm diamond paste and washed with tap water, acetone, methanol and deionized water [14–16]. We have cleaned the single crystal (highly electrically resis-

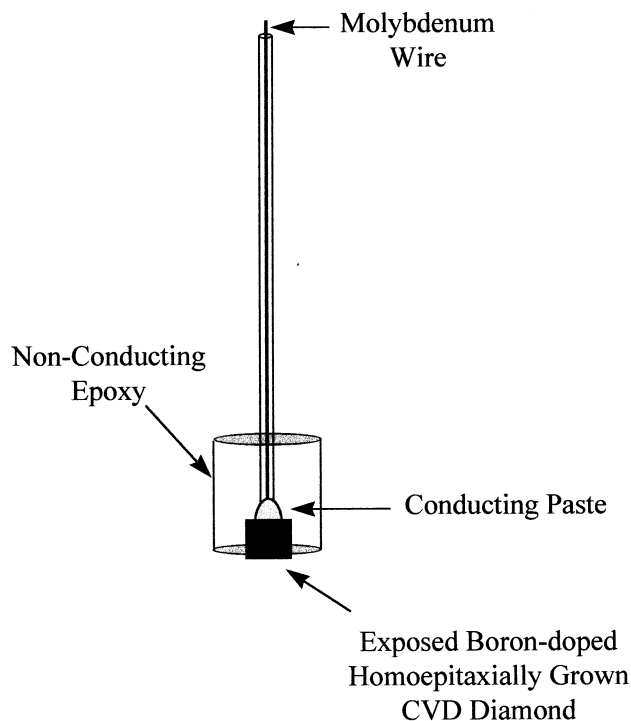


Fig. 1. Schematic diagram of the single crystal diamond electrode prepared for voltammetric studies.

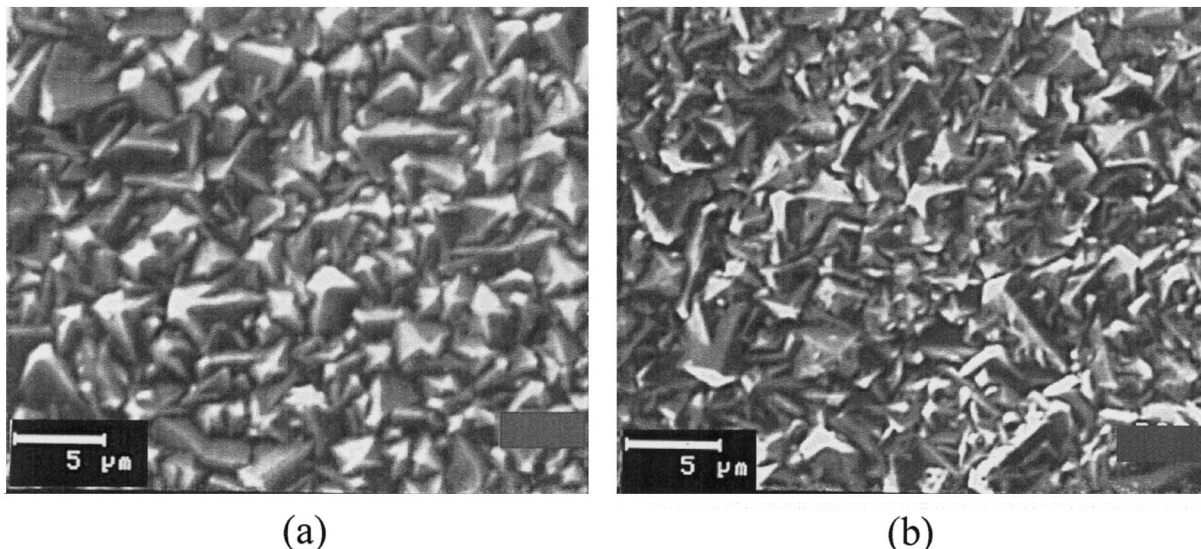


Fig. 2. Scanning electron micrographs of (a) as-deposited boron-doped polycrystalline diamond film grown over the molybdenum substrate and (b) electrochemically tested boron-doped diamond in 0.5 M NaCl supporting electrolyte solution.

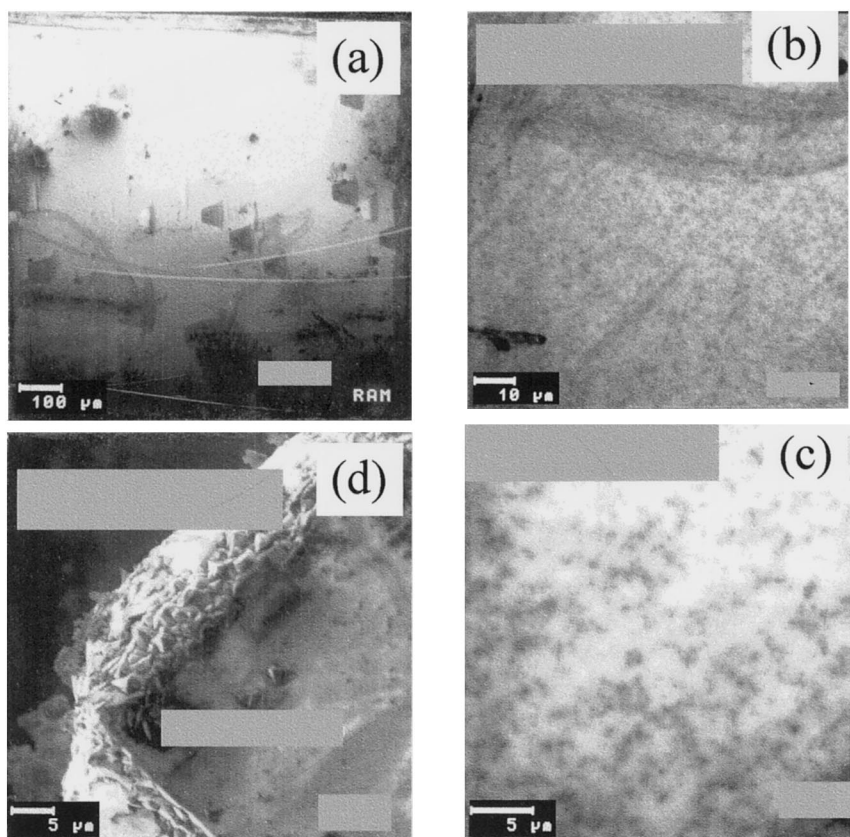


Fig. 3. Scanning electron micrographs of homoepitaxially grown boron-doped microwave plasma deposited CVD diamond over type IIa single crystal diamond. (a) Typical morphology of the diamond away from the edge of the crystal (b and c) magnified view of 'a' and (d) morphology of the diamond on the edge of the crystal.

tive, undoped) type IIa diamond (orientation: 100) substrate using acetone, methanol and deionized water. The single crystal undoped diamond substrate was not given any chemical pretreatment and we have deposited boron-doped diamond homoepitaxially.

A microwave plasma (2.45 GHz) assisted CVD system (ASTeX) has been used to grow diamond films. A schematic diagram of the diamond deposition system has been described earlier [14]. The substrate was placed at the center of the stage that was then loaded

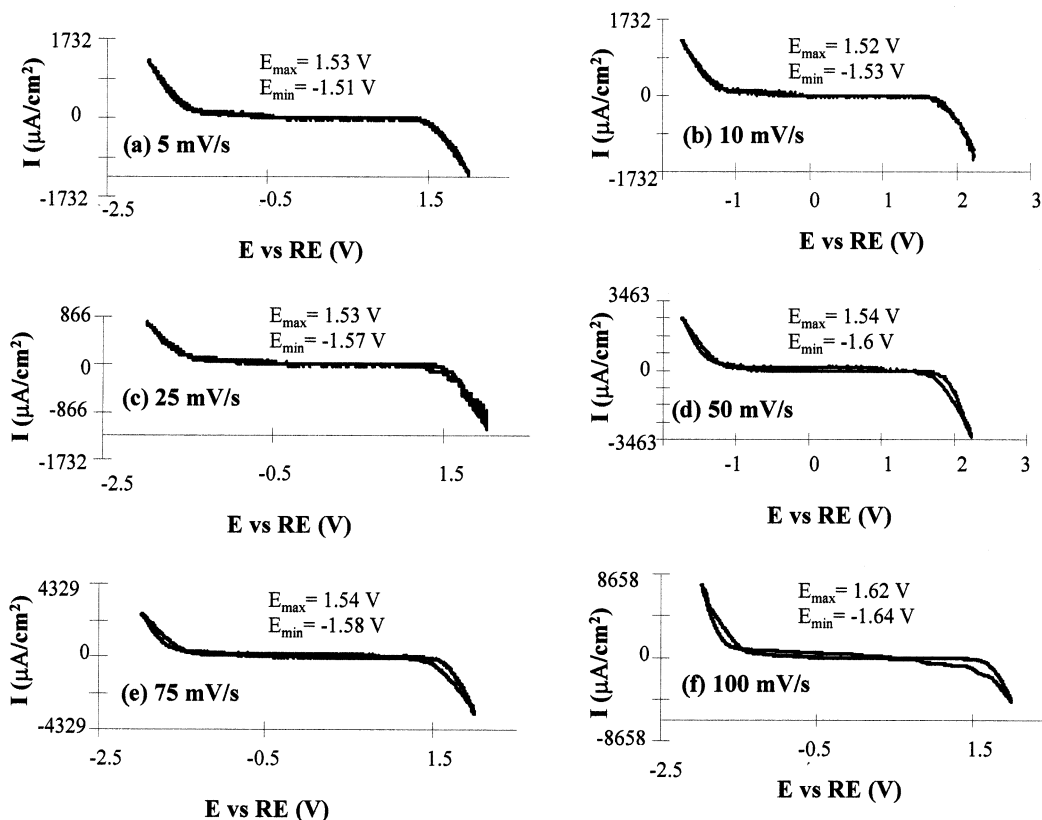


Fig. 4. Background cyclic voltammetric behavior of highly conducting boron-doped homoepitaxially grown CVD diamond over Type IIa diamond in a supporting electrolyte of 0.5 M NaCl with correction for uncompensated IR drop at various scan rate. (a) 5 (b) 10 (c) 25 (d) 50 (e) 75 and (f) 100 mV s^{-1} . Apparent area of the electrode 0.0231 cm^2 . Reference electrode: $\text{Ag}|\text{AgCl}|0.5 \text{ M NaCl}$.

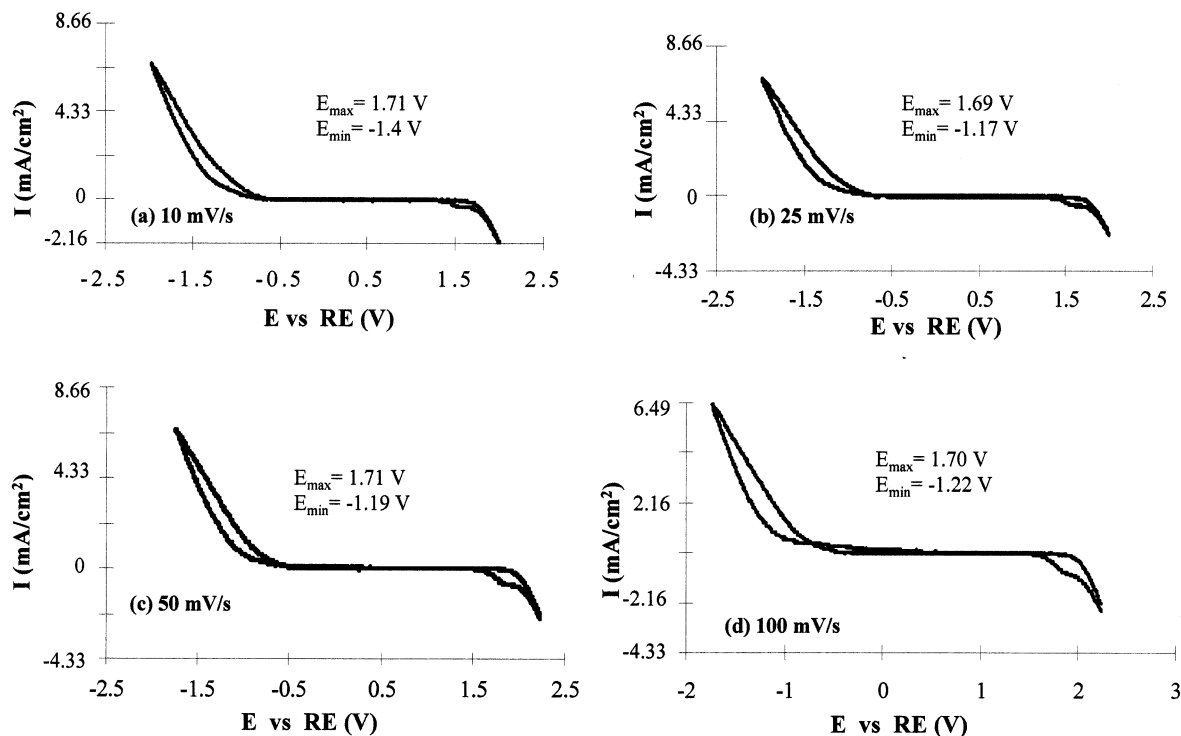


Fig. 5. Background cyclic voltammetric behavior of highly conducting boron-doped homoepitaxially grown CVD diamond over Type IIa diamond in a supporting electrolyte of 0.5 M HCl with correction for uncompensated IR drop at various scan rate. (a) 10 (b) 25 (c) 50 and (d) 100 mV s^{-1} . Apparent area of the electrode 0.0231 cm^2 . Reference electrode: $\text{Ag}|\text{AgCl}|0.5 \text{ M NaCl}$.

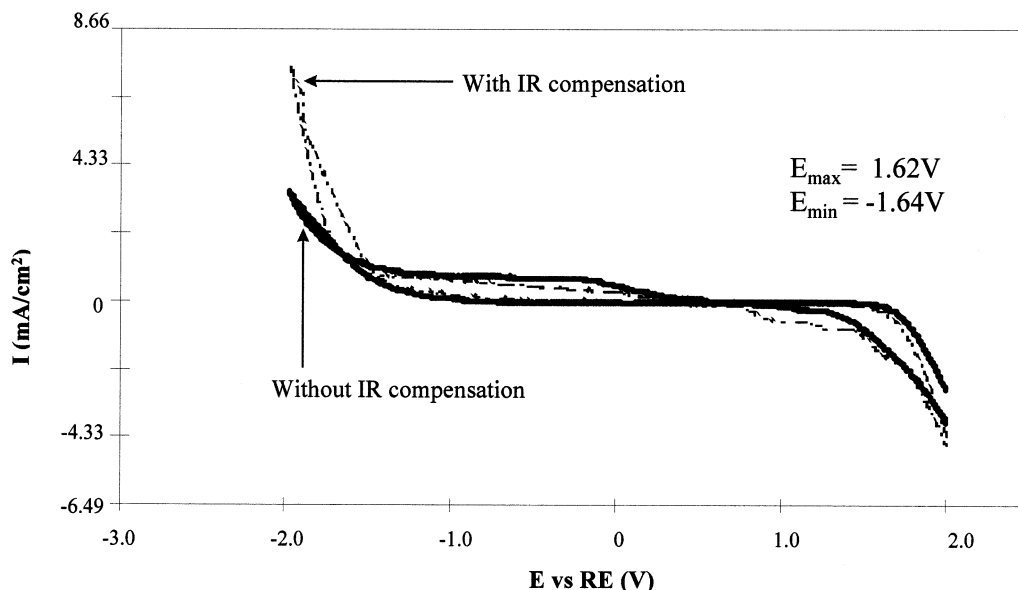


Fig. 6. Background cyclic voltammetric behavior of highly conducting boron-doped homoepitaxially grown single crystal CVD diamond in a supporting electrolyte of 0.5 M NaCl. Scan rate: 100 mV s^{-1} . (a) Without correction for uncompensated IR drop, (b) with correction for uncompensated IR drop. Apparent area of the electrode 0.0231 cm^2 . Reference electrode: $\text{Ag|AgCl|0.5 M NaCl}$.

into the quartz bell jar reactor. The reactor was evacuated to a base pressure of 10^{-4} Torr. A plasma was obtained by adjusting the pressure in the chamber, the hydrogen flow rate, microwave power and wave guide tuning. The substrate was heated in-situ by the microwave plasma to attain the desired substrate temperature before initiating the growth of diamond. Ultra high purity grade hydrogen and research grade methane was used in our experiments. The temperature was monitored remotely by an optical pyrometer. Diamond deposition was started by injecting methane into the system when the substrate reached the desired temperature. The deposition rate under the typical conditions provided in Table 1 was normally $1\text{--}0.5 \mu\text{m h}^{-1}$. A continuous film of diamond was usually obtained after 10–20 h of growth.

Boron doping of diamond film was achieved by using a solid disk source consisting of B_2O_3 and other oxides. The solid disk source of boron has been used as a base to the substrate during growth of in-situ doped diamond at a substrate temperature of $960\text{--}997^\circ\text{C}$. The size ($30 \times 30 \text{ mm}$) of the boron source disk is greater than that of the substrate ($21 \times 21 \text{ mm}$) where the doping source disk can be exposed to the hydrogen and methane microwave plasma to raise the temperature to have sufficient vapor pressure of atomic boron around the disc to yield in-situ doping of diamond. Typical deposition parameters are given in Table 1 [16]. Electrical resistivity of boron-doped diamond sample was measured to be $0.001\text{--}0.01 \Omega \text{ cm}$ by four-point probe technique. We did not measure the resistivity of single crystal boron-doped diamond. We have assumed the resistivity is similar to the polycrystalline or even lower resistivity since it has minimum number of defects in the film.

Initially, polycrystalline diamond was grown over the molybdenum substrate for 12.5 h and unloaded the sample and laid a single crystal substrate over the polycrystalline diamond and deposition was continued further for 9.25 h. At this stage the deposition was stopped, the chamber was evacuated and the single crystal diamond was flipped over. Deposition of boron-doped diamond was then continued for another 9 h. This procedure will allow the growth of doped diamond around the crystal and this will aid significantly in taking an electrical contact during the electrochemical measurements. The single crystal diamond substrate was attached to a molybdenum wire with conducting paste to have an electrical contact with the diamond. The molybdenum wire and single crystal diamond (all the sides and the back), conducting paste were completely covered with a non-conducting paste. Only the front side of the crystal was exposed to the solution to perform electrochemical measurements. Several precautions were taken during the diamond deposition and mounting the crystal to perform electrochemical experiments successfully. Fig. 1 shows the schematic diagram of the mounted boron-doped single crystal diamond electrode for electrochemical measurements.

All solutions were prepared using reagent grade chemicals in deionized water. Cyclic voltammograms were obtained at various scan rates of $100\text{--}5 \text{ mV s}^{-1}$. The exposed apparent area of the boron-doped diamond is 0.7122 cm^2 for polycrystalline diamond and 0.0231 cm^2 for single crystal diamond. True area of the electrode surface that is corrected for the surface roughness is 0.819 cm^2 [17]. The reference electrode, $\text{Ag|AgCl|0.5 M NaCl}$ for single crystal diamond and

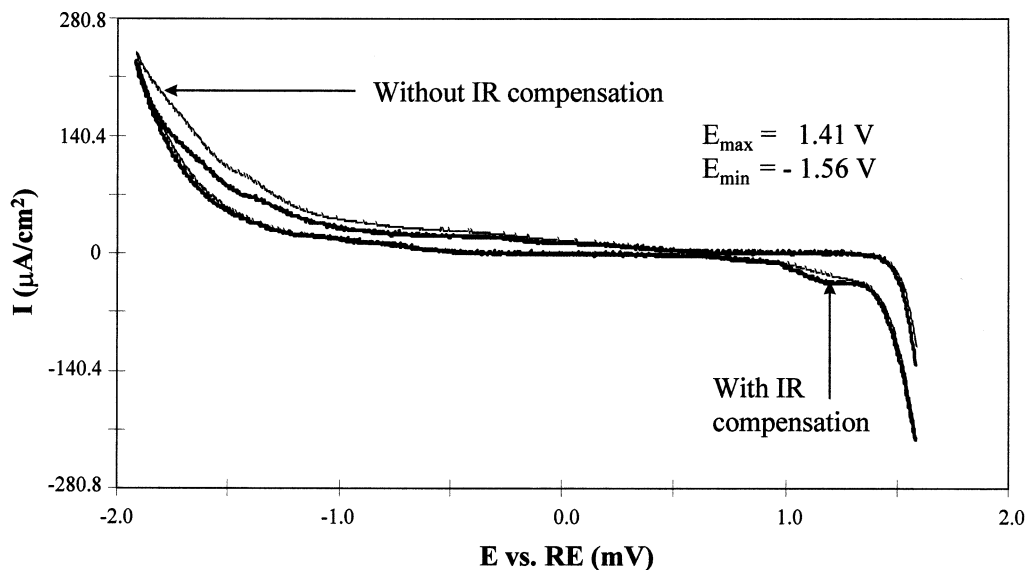


Fig. 7. Background cyclic voltammetric behavior of highly conducting boron-doped polycrystalline diamond in a supporting electrolyte of 0.5 M NaCl. Scan rate: 100 mV s^{-1} . (a) Without correction for uncompensated IR drop, (b) with correction for uncompensated IR drop. Apparent area of the electrode 0.7122 cm^2 (true area: $\sim 0.819 \text{ cm}^2$). Reference electrode: $\text{Ag}|\text{AgCl}|\text{satd. KCl}$.

$\text{Ag}|\text{AgCl}|\text{saturated KCl}$ for polycrystalline diamond, was used in this study. The potential of the reference electrode for 0.5 M NaCl and saturated KCl solution with respect to standard hydrogen electrode (SHE) is ~ 0.240 and 0.197 V versus NHE, respectively. A platinum mesh or foil counter electrode was used during all the electrochemical measurements. All the electrochemical experiments were conducted at room temperature, 25°C . The solution was open to the air during electrochemical measurements. A simple three electrode (test electrode, counter electrode, reference electrode) and single compartment electrochemical cell was used in this study. A potentiostat (EG and G Model 273) was used to perform cyclic voltammetry experiments. We have corrected the experimental data for uncompensated IR drop. The solution was not stirred during the cyclic voltammetry experiments.

3. Results and discussion

The morphology of the diamond films is very well faceted according to scanning electron microscopy. The films grown by microwave plasma CVD reactor under similar experimental conditions have been analyzed by Raman spectroscopy earlier [4,14–16]. As-deposited CVD diamond films are rough and the roughness factor varies from 0.1 to $1 \mu\text{m}$ as per profilometry [14]. Martin et al. [17] have recently reported the measurement of surface roughness of hot filament CVD diamond films by atomic force microscopy (AFM). A 15% area correction was found for roughness on the scale of the diamond crystallite dimension of $0.1 \mu\text{m}$. Therefore, we

are assuming the surface area of our films to be of the same order since the films are grown by microwave plasma CVD of similar morphology. Apparent area of the electrode 0.7122 cm^2 (true area corrected for surface roughness: $\sim 0.819 \text{ cm}^2$). Area of the electrode is constant in all the experiments reported in this paper. Fig. 2 shows the typical morphology of polycrystalline diamond grown over the molybdenum substrate before and after electrochemical characterization. The nature of the diamond has not changed after several electrochemical experiments. This is a significant observation although it was expected for the diamond.

Fig. 3 shows scanning electron micrographs of homoepitaxially grown boron-doped microwave plasma deposited CVD diamond over type IIa single crystal diamond. (a) Typical morphology of the diamond away from the edge of the crystal, (b and c) magnified view of 'a' and (d) morphology of the diamond on the edge of the crystal. Fig. 3 (a, b and c) show that the surface is smooth when compared the morphology at the edge of the crystal and polycrystalline diamond morphology shown in Fig. 2. The film that is deposited away from the edge is homoepitaxial in nature. We have not analyzed this crystal for Raman.

Figs. 4 (a–f) and 5 (a–d) show the background voltammetric response of the boron-doped homoepitaxially grown single crystal diamond in 0.5 M NaCl and 0.5 M HCl solution, respectively at various scan rates ranging from 5 to 100 mV s^{-1} . The data shown in Figs. 4 and 5 are corrected for uncompensated IR drop. Doped single crystal diamond is electrochemically stable over a potential range from $+1.5$ to -1.5 V versus R.E (R.E. Potential: 0.24 V versus NHE) in 0.5 M

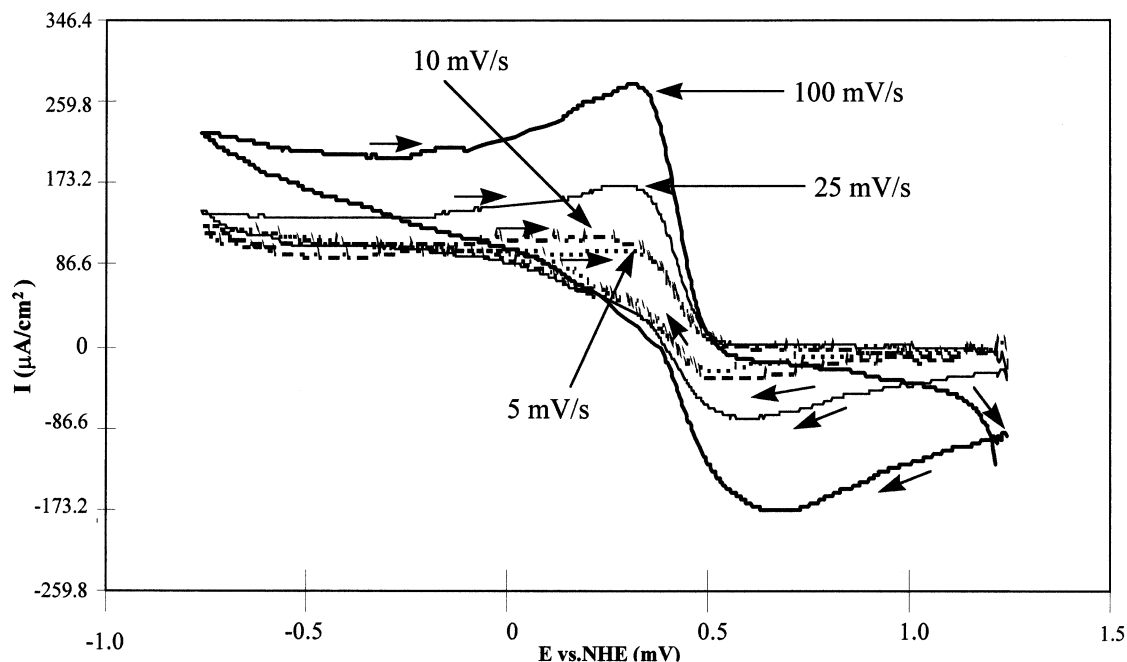


Fig. 8. Cyclic voltammetric behavior of single crystal homoepitaxially grown boron-doped CVD diamond in 0.5 M NaCl solution containing 4 mM $\text{K}_3\text{Fe}(\text{CN})_6$ at a scan rate of 100, 25, 10, 5 mV s^{-1} . Area of the electrode is 0.0231 cm^2 .

NaCl solution as shown in Fig. 4. Insignificant decomposition of water has occurred over a wide potential range. This characteristic may be used to investigate the redox kinetics in the electroanalytical chemistry. To our knowledge this is the first report on the voltammetric response at the single crystal diamond electrode/solution interface. Similarly, the diamond is very stable over a potential range from +1.7 to 1.2 V versus R.E. (R.E. Potential: 0.24 V versus NHE) in 0.5 M HCl solution. The potential ranges (E_{max} and E_{min}) provided in this paper is approximate values within $\pm 50 \text{ mV}$. The E_{max} (anodic side of the equilibrium potential or positive) and E_{min} (cathodic side of the equilibrium potential or negative) are potentials corresponding to the initiation of oxygen and hydrogen evolution, respectively, on the boron doped diamond electrode. The potential shift towards positive direction by 59 mV pH^{-1} according to Nernst equation. Therefore, a shift of 0.413 V in the positive direction as the supporting electrolyte changed from 0.5 M NaCl solution to 0.5 M HCl solution. We did not measure the pH of the solution. Positive voltammetric limit for single crystal diamond is higher than in 0.5 M NaCl solution.

Figs. 6 and 7 show the cyclic voltammetric response of the single crystal diamond and polycrystalline CVD diamond in 0.5 M NaCl solution, respectively, with and without correction for uncompensated IR drop. The stable voltammetric response over the wide potential range is almost similar but the rate of faradaic reaction is higher during O_2 and H_2 evolution while the data corrected for uncompensated IR drop. The potential

range is independent of the correction for uncompensated IR drop at least in this study. All the experimental data reported in this paper is corrected for uncompensated IR drop unless it is specifically stated. The voltammetric limits are slightly different for the polycrystalline and single crystal diamond as shown in Figs. 6 and 7 and that could be due to the nature of surface, grain boundaries, relative amount of non-diamond carbon, metallic impurities present in the polycrystalline diamond. Polycrystalline diamond was grown on non-diamond substrate and this could cause some contamination of deposited CVD diamond film.

Fig. 8 shows the cyclic voltammetric behavior of conducting boron-doped homoepitaxially grown CVD diamond at a scan rates of 100, 25, 10 and 5 mV s^{-1} in 0.5 M NaCl solution containing 4 mM $\text{K}_3\text{Fe}(\text{CN})_6$. ΔE_p is defined as separation of anodic and cathodic peaks in a cyclic voltammogram. The ΔE_p has decreased as the scan rate decreases and this value is higher than expected. This could be due the slow electron transfer reaction at the diamond electrode and also could be due to the higher electrical resistivity of diamond. The higher electrical resistivity of diamond may yield a lower faradaic current and also higher polarization resistance. Therefore, the potential of the electrode will be difficult to control. Such electrode system may be equivalent to a passive system or an irreversible system. The electrochemical irreversible system usually yield high ΔE_p values. This may be a plausible explanation to explain high ΔE_p values observed in this study. The data shown in the Fig. 8 was corrected for uncompen-

Table 2

Kinetic analysis results for boron-doped diamond electrode material in 0.5 M NaCl solution containing 4 mM ferricyanide

Scan rate (mV s ⁻¹)	t_p (s)	α	$k_{\text{het}} (\alpha = 0.5)$ (cm s ⁻¹)	α	$k_{\text{het}} (\alpha = \text{variable})$ (cm s ⁻¹)
100	0.03	0.5	6.07×10^{-5}	0.62	6.62×10^{-5}
25	0.12	0.5	2.06×10^{-4}	0.59	2.13×10^{-4}
10	0.30	0.5	1.41×10^{-4}	0.46	1.42×10^{-4}
5	0.60	0.5	1.50×10^{-4}	0.48	1.50×10^{-4}

sated IR drop and subtracted for the background current. The number of electrons transferred in the redox reaction studied is one; this can be determined from the separation between the peak potentials in the cyclic voltammogram. Inset in Fig. 8 shows the ΔE_p values that are significantly higher than the predicted value such as 59 mV. This indicates the occurrence of slow electron transfer kinetics at the diamond electrode surface. ‘Electrochemical irreversibility’ has caused the peak separation, ΔE_p , to increase. The higher value of peak separation could also be due to the high resistivity of diamond film grown by CVD processes [11,12]. The ratio of the peak currents is greater than unity, which indicates the influence of other chemical reactions, coupled to the electrode process and also the irreversibility of the electrode process itself. Highly conducting boron-doped diamond is necessary in order to use the diamond as an electrode material in electroanalytical chemistry and electrochemical sensor applications.

The heterogeneous electron transfer rate constant, k_{het} , was determined using the cyclic voltammetric data shown in Fig. 8 and EG and G’s Model 271 COOL Kinetic Analysis Software (v 1.23) [18]. Alpha (α) is the charge transfer coefficient associated with the quasi-reversible redox process. Simulation was performed using both constant and variable α values. The α -value affects the shape of the curves observed in cyclic voltammetric experimental data. The diffusion coefficient for ferricyanide (D_o) is 7.6×10^{-6} cm² s⁻¹ and for ferrocyanide (D_R) is 6.3×10^{-6} cm² s⁻¹. Table 2 shows the kinetic analysis results for boron-doped diamond electrode material in 0.5 M NaCl solution containing 4 mM ferricyanide. The observed heterogeneous rate constant varied from 1.41×10^{-4} to 6.07×10^{-5} cm s⁻¹ where α was equal to 0.5. The observed heterogeneous rate constant varied from 1.42×10^{-4} to 6.62×10^{-5} cm s⁻¹ where α was floating (0.46–0.62). The rate constant is approximately two orders of magnitude lower than the rate constant observed at the glassy carbon electrode. Therefore, the redox kinetics of ferri/ferrocyanide is considered sluggish at the boron diamond electrode when compared to the glassy carbon and other electrodes [18].

4. Conclusions

We have reported the growth of boron doped diamond over the molybdenum and type IIa substrate by microwave plasma CVD process using hydrogen along with methane as a carbon source. Background cyclic voltammograms for polycrystalline and single crystal diamond showed that the diamond electrode material is stable in supporting electrolytes such as 0.5 M NaCl and 0.5 M HCl over a wide potential range. The level of background current observed for single crystal diamond is lower than the polycrystalline diamond. To our knowledge this is the first report on the comparison of background current for polycrystalline and single crystal doped diamond. It may qualitatively be concluded that based on the stability characteristics observed in background cyclic voltammetry studies, diamond electrodes should have a significant application in electroanalytical chemistry. We have investigated the redox kinetics of ferri-ferrocyanide at the boron-doped single crystal diamond and the peak separation was observed to be in the range of 177 mV at a scan rate of 5 mV s⁻¹. This indicates the electrochemical irreversibility or slow electron transfer kinetics at the diamond/solution interface. The heterogeneous electron-transfer rate constant was observed to be in the range from 6.07×10^{-5} to 1.42×10^{-4} cm s⁻¹.

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Biography

R. Ramesham is working as a Senior Member of Key Engineering Staff at the Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA. His present research work focus on the reliability of pack-

aging and interconnects associated with the microelectromechanical systems (MEMS) applications. He also works on the application of polycrystalline synthetic diamond for MEMS, electrochemical, electroanalytical and corrosion resistant coating applications. He worked for Auburn University for little over 9 years. His research work at Auburn University addressed the fundamental issues involved in diamond processing techniques, heat dissipation techniques and electrochemical applications of diamond. He has extensive experience in diamond film growth, photolithography, thin films, semiconductor processing, characterization, microstructure fabrication, electrochemical evaluation of diamond coatings, etc. He has received an outstanding research performance award from the electrical engineering department of Auburn University. Dr. Ramesham has worked for the NASA's JPL, Caltech, Pasadena, CA, as a NRC RRA from October 1985 to October 1988 in Microdevices Laboratory. He has received three awards from NASA for the research work he performed. He has published over 86 refereed journal and proceedings articles and has made 58 national and international conference presentations. He has given invited presentations at the national and international conferences. He received the best research paper award from the IEEE Alabama Section. He has two patents and one is pending and two NASA technical disclosures. He has offered a short course on 'Fabrication of Thin Film Diamond Microstructures' at the First international Conference on the Applications of Diamond Thin Films and Related Materials, 17–22 August 1991, Auburn, AL. Dr. Ramesham received his Ph.D. from the Indian Institute of Science, Bangalore, India. He received a fellowship from the DoE, India during graduate studies. He has obtained 2nd rank at the M.S. and received a county first rank at the B.S. level. He received a National Merit Scholarship during his M.S. studies.