

Deposition of Nanodiamond Films on Titanium Substrates by Coaxial Arc Plasma Deposition and Their Application to Dental Implant

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Name

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(チタン基板上への同軸型アークプラズマ堆積法によるナノダイヤモンド薄膜の堆積と歯科インプラントへの応用)

Title

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論文内容の要旨

Thesis Summary

Coaxial arc plasma deposition (CAPD) is a type of physical vapor deposition (PVD) that uses a high-current arc discharge to vaporize graphitic target materials. The vaporized carbon from the graphitic materials is then transported to a substrate by a plasma stream, and it is deposited in a thin film. CAPD is a versatile technique that can be used to deposit quenched-produced diamond (Q-dia) films with nanodiamond grains of less than 10 nm in the amorphous carbon matrix. CAPD is a versatile technique that can be used to deposit diamond films on a wide range of substrates with a relatively low-cost process and without a wet seeding process making it an attractive option for a variety of biomedical applications. One of the key advantages of CAPD is that it can be used to deposit films at high deposition rates. This is due to the high energy of the plasma stream, which can transport the vaporized material to the substrate very quickly. CAPD can also be used to deposit films with very good adhesion. This is because the high energy of the plasma stream can activate the surface of the substrate, making it more receptive to the deposited material.

In this study, we took advantage of the ability to control the deposition parameters of CAPD such as substrate temperature and negative bias applied to substrates, which results in significant manipulation of the mechanical and structural properties of the deposited Q-dia films. Moreover, the thickness of the deposited film was controlled by adjusting the time that the substrate is exposed to the plasma stream.

Chapter 1 describes an overview of the research motivation and challenges of titanium dental implants. It summarizes carbon-based nanomaterials and their structure, as well as the breakthroughs in the discovery of new techniques for growing polycrystalline diamond films with grain sizes ≤ 10 nm. These breakthroughs have led to a significant advancement in materials science. Although different synthesis techniques of diamond films and their properties have been introduced, such as high-pressure high-temperature, chemical vapor deposition methods including microwave and hot filament plasma, and physical vapor deposition including pulsed laser deposition, this chapter focuses on nanocrystalline diamond films synthesized by coaxial arc plasma deposition (CAPD).

Chapter 2 presents the experimental setup employed for the synthesis of Q-dia films using CAPD. The chapter highlights the morphological characterization of the films, which was carried out through 3D measuring and field effect laser scanning microscopies (FE-SEM). In addition, the chemical and microstructural properties of the films were analyzed using techniques such as energy dispersive X-ray (EDX), confocal Raman spectroscopy, X-ray photoelectron spectroscopy, and near-edge fine absorption spectroscopy. In addition, the mechanical properties of the films were evaluated by scratch and nanoindentation tests, providing valuable insights into their durability and strength.

Chapter 3 describes the effects of various pretreatment methods on the adhesion of Q-dia films to titanium substrates through CAPD. The growth of Q-dia films on titanium substrates at room temperature is challenging due to the presence of a native oxide layer. However, the use of hydrofluoric acid etching and the introduction of a titanium carbide interlayer were found to be effective in removing the native oxide layer and promoting adhesion between Q-dia films and titanium at room temperature. However, complete removal of the oxide layer is difficult to achieve with these ex-situ pretreatment methods, because reoxidation is likely to occur after the pretreatment. The presence of residual atmospheric oxygen at the interfaces limits the adhesion strength of the Q-dia films on titanium after both ex-situ pretreatments.

Chapter 4 presents an innovative approach to achieve room-temperature adhesion of Q-dia films on titanium substrates. This breakthrough is achieved by a hybrid configuration of an ion etching gun (IG) and CAPD, which has been redesigned for this purpose. Among the various pretreatment methods discussed in Chapter 3, in-situ Ar^+ plasma etching has proven to be the most effective in completely removing the native oxide layer. By manipulating the substrate morphology and terminating the superficial oxide layer, in-situ Ar^+ plasma etching enables the instantaneous deposition of high-quality Q-dia films by CAPD at room temperature. The Q-dia films grown using this hybrid system (IG/CAPD) demonstrate superior mechanical and structural properties, as further discussed in the chapter.

Chapter 5 describes the remarkable success in achieving adhesion of high-quality Q-dia films on titanium substrates at room temperature using IG/CAPD. This chapter explores the significant advances made by the negative bias-enhanced nucleation bias enhanced growth (BEN-BEG). BEN-BEG pushes the boundaries of the structural and mechanical properties of Q-dia films on Ti, offering unprecedented improvements. By varying the bias voltages from 0 V to -100 V, the optimal negative bias voltage for Q-dia film growth was determined to be -40 V. At this optimal bias voltage, the films exhibited exceptional properties, including a strong adhesion strength of 48 N and an impressive superhardness value of 96 GPa equivalent to single crystalline diamond. These outstanding properties were achieved through a synergistic effect involving the selective etching of predeposited sp^2 -C bonded atoms to leave a nucleus of sp^3 -C bonded sites, sub-implantation of C^+ ions, and further growth of compressed sp^3 -C bonded atoms. This chapter highlights the potential of optimizing BEN-BEG for Q-dia films, demonstrating their ability to improve structural and mechanical properties and making them highly promising for the next generation of biomedical dental implants.

Chapter 6 investigates the biocompatibility of Q-dia films for dental implant applications through in vitro culture of epithelial cells on the Q-dia films. The influence of the Q-dia films roughness on the adhesion of the epithelial cells was investigated and revealed highest affinity of the epithelial cells to adhere to smooth surfaces of the Q-dia films rather than bare Ti substrates. Further investigation was carried out the adhesion of the epithelial cells to the optimal bias voltage Q-dia films with smooth and extremely smooth surfaces. The uniformity of the Q-dia films were altered through mechanical polishing of the films in attempt to remove amorphous carbon contribution to the surface. The epithelial cells seemed to have preferential affinity to uniform surfaces which promotes their proliferation and differentiation.

Chapter 7 summarizes the main objectives of this work. It highlights the diverse methodologies used to achieve room temperature adhesion of Q-dia films on Ti substrates using the CAPD technique. The *ex situ* HF acid etching for removing the native oxide layer

from the Ti substrate, as well as the insertion of a TiC intermediate layer to create a chemical composition gradient, were insufficient to establish strong adhesion of the Q-dia film to Ti. The presence of oxide residue, either in the roughened grooves after HF acid etching or at the interfaces due to the TiC intermediate layer, limited the mechanical and structural properties of the Q-dia films on Ti. On the other hand, the *in situ* elimination of the oxide layer demonstrated by modifying the CAPD into a hybrid configuration of IG/CAPD proved to be effective in excluding the oxide layer and depositing a high-quality Q-dia film on Ti at room temperature without the need for chemical pretreatment or interlayer insertion. The performance of the hybrid configuration was further enhanced by applying a conjugated negative bias during the film growth. An optimal bias voltage enabled the synthesis of superhard and highly adherent Q-dia films on Ti at room temperature.

Lastly, the biocompatibility of the developed Q-dia films was assessed by incubating oral epithelial cells obtained from Wistar rats. The cells exhibited a higher affinity towards the Q-dia films compared to bare Ti substrates, indicating great potential for dental implant applications.