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Antibacterial properties of fluorinated diamond-like carbon films deposited by direct and remote plasma

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ABSTRACT

The antibacterial activity and cell viability of fluorinated diamond-like carbon (F-DLC) films as bio coating were investigated. The F-DLC films were deposited on polycarbonate substrates by the radiofrequency (RF) plasma (direct mode) and microwave (MW) afterglow plasma (remote mode). The influences of RF and MW power on the fibroblast cell proliferation and antibacterial activity against *E.coli* were studied. Additionally the relevance of F atoms concentration, surface roughness and surface energy with the antibacterial activity was investigated. The fibroblast cell cultivation tests revealed that the F-DLC films have no toxicity for cells. Increasing the RF power increased the F concentration and surface roughness, lowered the surface energy and eradicated 70% of *E.coli* bacteria. A drop in this growing trend happened in the remote mode. As the MW power increased, the F content and surface roughness decreased, the films became hydrophilic, and only thirty percent of bacteria was killed.

1. Introduction

Nowadays there are increasing rate of implant devices inserted in the human body in several purposes from dental to cardiovascular and hip joints applications. The implant devices by themselves could make two main problems for the host body. The first one comes from the implant materials and the second problem is the bacterial infections. These infections cause local pain around the surgical area and additionally bone destruction which loses the implant typically leading to remove the device [1]. Consequently, to increase the lifetime of implants we have to overcome these two main problems.

Diamond-like carbon (DLC) due to its low friction coefficient, high hardness, wear resistant, chemical inertness, and biocompatibility is one of the most effective coatings for solving the first problem which increases durability of implants in body [2]. Although, DLC coatings have good mechanical properties but show no much antibacterial activity and the bacteria can attach to the surface of the DLC films [3]. Therefore, it is better to embed other elements in the DLC structure to improve its function as an antibacterial coating. DLC amorphous nature helps to embed other elements like F to its structure to improve its surface energy and biocompatibility for specific applications [4]. The objective of this paper is to evaluate the antibacterial activity of F-DLC coated polycarbonate (PC) biomaterial. Additionally, we investigate and compare the efficiency of remote mode (radio frequency plasma and microwave afterglow plasma, simultaneously) and direct mode (radio frequency plasma) processing on the antibacterial and cell viability of F-DLC coatings.

2. Experimental

The specifications of the direct and remote plasma deposition system were discussed in detail in [5]. The first section of the system, direct mode, includes a single-electrode capacitively coupled RF plasma system in which the generated plasma is in direct contact with the substrate. The door of the system was designed to be capable of adding surfaguide-produced MW plasma in which its afterglow is in contact with the substrate. 15 sccm of C_2H_2 and 10 sccm CF_4 as processing gases were injected to the system by mass flow controllers to stabilize the working pressure at 8 Pa. In the direct mode, both gases were introduced in to the RF section. In the remote mode, CF_4 plasma

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Table 1
Fluorine concentration, surface roughness, contact angle and surface energy of F-DLC films.

RF power (W)	MW power (W)	F	Roughness	Contact angle (degree)		Surface energy (mN/m)		
		(Atomic %)	(nm)	Water	Diiodomethane	Dispersive	Polar	Total
50	0	12.47	4.66	73.8	34.9	42.04	5.1	47.1
100	0	13.46	5.16	76.8	37.9	40.5	4.3	44.8
150	0	15.07	7.06	79.5	41.8	38.6	3.8	42.4
200	0	16.53	9.49	80.5	42.4	38.3	3.5	41.8
200	250	8.88	9.26	77.8	40.5	39.3	4.2	43.5
200	350	9.32	7.27	69.3	41	39	7.8	46.8
200	450	7.91	7.78	66.5	33.9	42.4	8.1	50.5

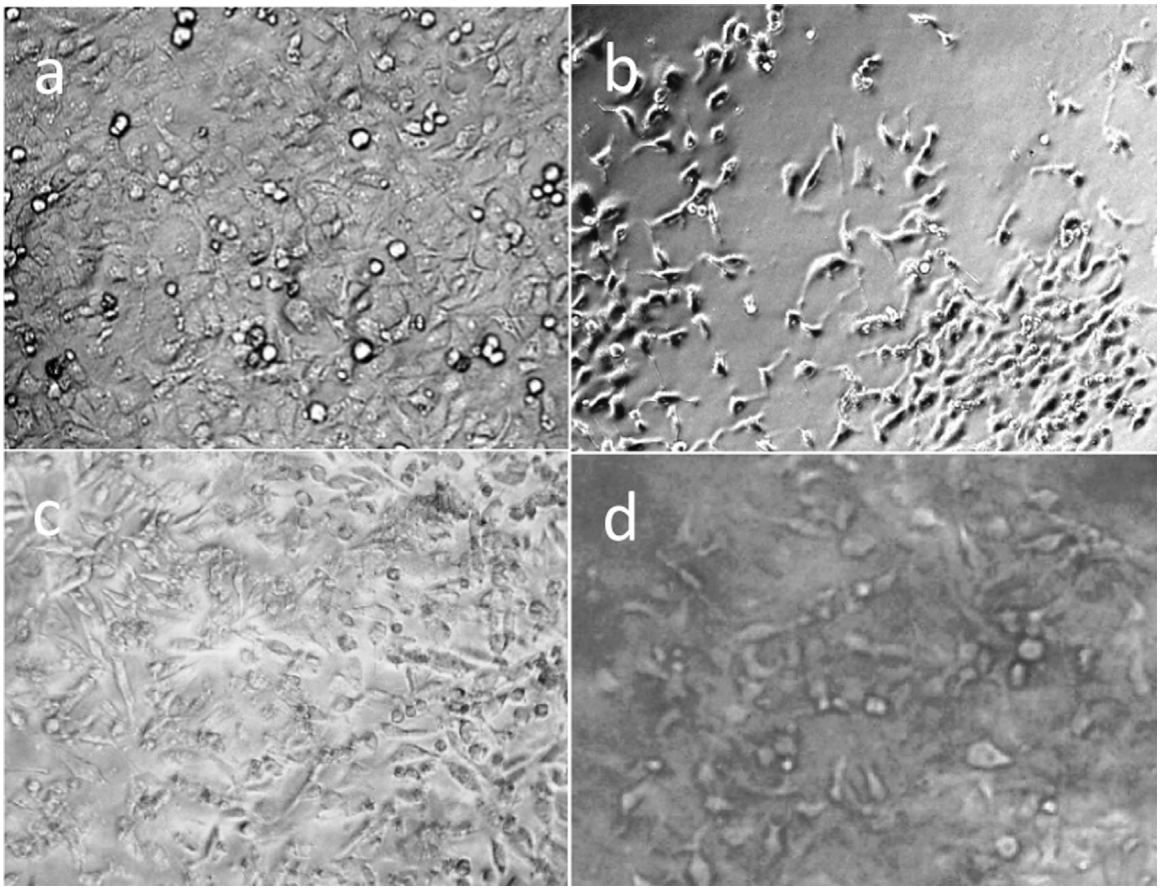

Fig. 1. Images of cultured cells on a) none coated PC and F-DLC coated PC in direct mode b) 50 W of RF power, c) 200 W of RF power and remote mode d) 200 W of RF power and 450 W of MW power.

(main source of fluorine atoms) was generated in the MW section.

In this work, we study the effects of RF power (50, 100, 150 and 200 W) in the direct mode and MW power (250, 350 and 450 W) with 200 W of RF power in the remote mode, respectively, on the cell viability and antibacterial activity of fluorinated DLC films. All of the experiments were done at room temperature in 90 s. The optical emission spectroscopy of the plasma and properties of F-DLC film such as fluorine concentration, deposition rate, surface roughness, and etc were presented in our last work [6]. However, some of them which help us to interpret the results of antibacterial activity of F-DLC films are summarized in Table 1.

In the direct mode, the thickness of the films grows from 117 nm to 234 nm with increasing the RF power from 50 W to 200 W which demonstrate the dominance of deposition to etching process. The similar trend exists in the remote mode in which the film thickness increases from 306 nm to 324 nm with increment of the MW power from 250 W to 450 W. Also the Raman spectroscopy of the films showed the G peak position shifts from 1541 toward 1553 cm^{-1} and I_D/I_G ratio increases from 1.3 to 1.52 as well as the growth of fluorine concentration in the film with increasing the RF power from 50 W to 200 W in direct mode. This is due to the fluorine termination of the C-C networks which result in the decrement of the cross linking, production of more open structural arrangements and the film density reduction [6,7]. In spite of the reduction of F concentration in the films in remote mode, almost the same tendency can be observed in the Raman spectra of the deposited F-DLC films in this mode. This is the result of more production of C_2 species compared to the F containing ones in the plasma which in turn causes the formation of more sp2 hybridization in the F-DLC matrix [8]. Also the XPS analysis of the other researches shows the presence of -C-C, -C-CF, -CF and $-CF_n$ chemical bonding in the F-DLC films [7].

To analyze the cell viability the HS-5 stromal cells were cultured on the F-DLC films and the [3,(4, 5-dimethylthiazol-2-yl)-5-(3-carboxymethoxyphenyl)-2-(4-)(sulfophenyl)-2H-tetrazolium] MTT assay was