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Antibacterial nanocomposite coatings produced by means of gas aggregation source of silver nanoparticles



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ABSTRACT

Silver nanoparticle-based antibacterial nanocomposite coatings were fabricated by means of gas aggregation source of Ag nanoparticles and plasma-enhanced chemical vapour deposition of matrix material. Combination of these techniques makes it possible to independently control the amount of Ag nanoparticles in the nanocomposites, as well as properties of matrix material, such as its chemical composition or wettability. This subsequently enables to tune kinetics of silver ion release and hence the antibacterial performance of produced nanocomposites. Based on detail measurements of silver ion release from Ag/plasma polymerized hexamethyldisiloxane and Ag/SiO_x nano-composites, it may be concluded that the release rate is strongly dependent both on matrix material and amount of Ag nanoparticles present in the nanocomposite. These measurements are furthermore accompanied with tests focused on the evaluation of antibacterial potency of produced nanocomposites.

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1. Introduction

Several hundreds of millions of surgical operations are performed worldwide each year (e.g. 51.4 million inpatients procedures were performed only in the U.S.A. in 2010 [1]). These procedures involve direct or indirect contact between patient's tissues and various medical instruments, devices or accessories and thus are connected with high risk of introduction of pathogens into a human body and post-operational complications. In addition, in implant surgery device related infection often results in necessity of re-operation that increases risks for patients weakened by previous operation and increases the costs to health care systems [2]. The number of these events could be largely reduced by improved sterilization and decontamination procedures adopted nowadays in hospitals. However, in the case of implants the late-stage infections are still reported. These events are often connected with the development of bacterial biofilm on the implant surface. This not only may decrease the implant acceptance but also provide a reservoir of infection from which bacteria can enter in the vascular system and be disseminated throughout the body.

The common strategy to treat bacterial infections is of course the application of antibiotics. However, due to changed physiology and the presence of protecting matrix, bacteria in biofilm are highly resistant to

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conventional antibiotic treatments. Also the general increase of multidrug resistance in many bacterial species, especially those involved in nosocomial infections, presents a serious actual problem of healthcare. Therefore an urgent demand to develop alternative bactericides exists. From this point of view materials that experience gradually increasing attention are nanocomposites that contain silver whose antibacterial properties are known since ancient times [3].

Recently, various nanocomposite coatings consisting of silver nanoparticles (Ag NPs) inside a matrix of plasma polymers and derived materials have been intensively investigated [4–10]. In this case, the matrix serve as protective coating that fixes Ag nanoparticles on a surface and prevents direct interaction of Ag NPs with bacteria as well as with human cells (e.g. accumulation of Ag NPs on the cell membrane and uptake within the cell that can induce cytotoxic or genotoxic effects) or release of Ag NPs into the surrounding environment reducing thus potential adverse effects on the environment. The nanocomposites then act as a reservoir for the out-diffusion of silver ions that are produced in aqueous environment by oxidative dissolution process involving protons and dissolved oxygen [11]:

$$2Ag_{(s)} + \frac{1}{2}O_{2(aq)} + 2H^{+}_{(aq)} \rightarrow 2Ag^{+}_{(aq)} + H_{2}O.$$
 (1)

Such ions subsequently interact with microorganisms and may inhibit their growth by impairing the function of their enzymes, damaging nucleic acids or by irreversible destruction of the cell membranes [12–14].

The advantage of use of plasma polymers as matrix material is 1) their very good adhesion to various substrates and 2) the possibility to regulate Ag⁺ ion release by properties of plasma polymerized matrix, e.g. its cross-link density, wettability or chemical structure [2].

Numerous ways were developed to produce Ag containing nanocomposites by plasma based techniques. The most common strategies are co-sputtering or sputtering combined with plasma-enhanced chemical vapor deposition (PE-CVD) [6–8]. However, these approaches are known for their limited ability to control independently the amount of NPs, their size distribution and the properties of embedding matrix. This arises from the fact that the process of NP formation from atomic fluxes is strongly coupled with the plasma polymerization process, both running on the substrate surface. In order to overcome this obstacle, other procedures were proposed in which the Ag NPs are produced independently of the matrix material. These methods were based for instance on addition of pre-fabricated Ag NPs or metal salts into the working gas flow [10,15], Ag NPs deposition by "wet" chemistry [8,16] or by immersion of substrates into the Ag NPs containing solution [17], which was followed by subsequent overcoating of such created Ag NPs films by a thin film of plasma polymers.

In this work, we study an alternative, fully vacuum based method that was previously employed for the fabrication of surfaces with controllable roughness or wettability [18,19]. This method is based on the combination of PE-CVD process used for production of matrix material with the deposition of Ag NPs by means of a gas aggregation nanocluster source. The NPs are created inside the gas aggregation source and reach the substrate in the form of beam of already formed nanoparticles. The size of produced NPs is thus determined solely by the operational parameters inside the gas aggregation source (e.g. pressure, power, gas flow, aggregation length), which enables to vary the amount of deposited NPs keeping their size distribution constant simply by increasing the deposition time. Such created NPs films may be subsequently overcoated by matrix material with required chemical composition or thickness. The principal advantage of this strategy is complete separation of production of NPs from the production of matrix.

The main aim of this study is to introduce this strategy as simple, but valuable approach that makes it possible to tailor the silver ion release, and hence antibacterial potency of produced nanocomposites, in wide range. In order to reach this aim the main attention is devoted to the evaluation of the effect of the amount of Ag NPs in produced nanocomposites, role of matrix material and influence of architecture of produced coatings on kinetics of silver ions.

2. Experimental details

2.1. Deposition of thin and multilayer coatings

Deposition system that was used in this study consisted of high vacuum chamber pumped by diffusion and rotary pumps, RF planar electrode operated at frequency 13.56 MHz, gas aggregation source (GAS) and load-lock system for introduction of samples into the deposition chamber (see Fig. 1). In the case of matrix deposition the precursor hexamethyldisiloxane (HMDSO, Sigma) was used. It was thermally stabilized and vaporized outside the apparatus and introduced to the deposition chamber at flow 0.25 sccm. Pressure during the depositions was 4 Pa and applied RF power was 40 W. In order to evaluate the effect of chemical composition of matrix material on the kinetics of Ag⁺ ion release and on the antibacterial properties, the HMDSO was used either alone or in a mixture with oxygen (HMDSO/O₂ ratio 1:60). According to the analysis of properties of thin films performed in previous study [19] coatings deposited using pure HMDSO (denoted as pHMDSO) have the structure typical for plasma polymerized organic films with high atomic concentration of carbon (57%), relatively low O (16%) and Si (27%) atomic concentrations and high water contact angle $(101.4 \pm 4^{\circ})$. In contrast, coatings prepared using O₂:HMDSO 60:1 mixture exhibited chemical structure close to stoichiometric SiO₂ (the atomic concentration of Si was 32%, O 64% and only 4% of carbon was observed) and are highly wettable with contact angle of water $15 \pm 5^{\circ}$.

Gas aggregation source (GAS) was used for the deposition of silver nanoparticles (Fig. 1b). It consisted of water cooled aggregation chamber ended with a conical lid with an orifice 2 mm in diameter. Inside the gas aggregation chamber was placed a DC planar magnetron equipped with silver target. The pressure inside the gas aggregation chamber was 30 Pa, magnetron current was 0.1 A. No mass or size filtration was applied in this study. More details related to the deposition of Ag NPs can be found in the previous study [20].

Sequential deposition of layers of plasma polymers and Ag NPs was used for the fabrication of nanocomposite coatings on glass disks (12 mm in diameter). Sandwich structures with up to 5 inter-layers of Ag NPs were produced. The first, inner and the last layers were always either pHMDSO or SiOx. The thickness of the first layer on the glass disk was 40 nm and the thicknesses of individual pHMDSO or SiOx layers as well as the thicknesses of the top-most layers were kept constant and equal to 10 nm. The thickness of pHMDSO or SiOx layers was determined by means of variable angle spectroscopic ellipsometer (Woolam M-2000DI) in the wavelength range of $\lambda = 192-1690$ nm at an angle of incidence AOI = 55-75° in air and at room temperature.



Fig. 1. Experimental set-up used for deposition of a) thin films and b) Ag nanoparticles.

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