



Recent progress on parylene C polymer for biomedical applications: A review



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ABSTRACT

Parylene C films have numerous advantages. The versatility of parylene C coatings makes them useful in a broad range of biomedical applications. The coatings are widely used commercially because of the unique combination of their physicochemical properties, i.e. flexibility and dielectricity. The chemical vapour deposition process enables parylene C conformal coatings to be applied to even the most complex medical devices. The coatings can be customised for the desired application using surface modification methods, which alter surface chemistry and topography. In this review, we summarise the last ten years (2008–2018) of research on parylene C for biomedical applications. We discuss how parylene C properties can be modulated through surface and bulk modifications to improve its key functions, i.e. anticorrosive, biocompatible, anti-infection, and therapeutic functions. We emphasise current and potential biomedical applications and finally highlight the advantages and limitations of the coatings, pointing out the perspectives and the most promising research trends.

1. Introduction

The application of biomaterials significantly contributes to human health; this has motivated extensive scientific research on these materials. The field is constantly growing along with the aging population and the increasing standard of living. The development of new generation biomaterials has led to multidisciplinary collaboration between scientists, engineers, and medical doctors. This is because biomaterials are rarely used alone. In most cases, they are integrated into hybrid biomedical devices, which consist of different classes of biomaterials, e.g. hip endoprostheses are fabricated from a stainless steel or titanium alloy (stem), a ceramic (head), and a polymer (insert).

Metal alloys are the most common class of biomaterials used for the fabrication of biomedical devices such as stents, knee and hip implants, pacemakers, and heart valves [1]. For such devices, corrosion rate is inhibited by a surface oxide film with a thickness of up to 100 μm , which is referred to as a passive layer. As biomaterials are in constant contact with body fluids at almost 37 $^{\circ}\text{C}$, corrosion is a major concern in biomaterials science. Metal surfaces undergo physicochemical destructive processes owing to the presence of inorganic ions, such as Na^+ , K^+ , Ca^{2+} , Mg^{2+} , OH^- , Cl^- , and PO_4^{3-} , in the surrounding. These processes are dominated by the kinetics of the dissolution of implant metal ions (accelerated by Cl^-) and repassivation through the

regeneration of a protective layer (quenched by Ca^{2+} and PO_4^{3-}) [2].

Damaging processes occur at a metal implant–body fluid interface after implantation. These processes are persistent, and they cannot be stopped through conventional surface engineering, as reported in numerous scientific papers [3–6]. Therefore, the deposition of a protective coating, independent of the class of biomaterial used, is a promising solution to stop the damaging processes Fig. 1.

There are several strategies for improving the corrosion resistance of metal–implant surfaces by forming a protective layer of hydroxyapatite [7,8], titania [7], zirconia/polymer hybrid coatings [9], and polymeric coatings [10,11]. Among these, one of the most explored strategies is the coating of metal implants with synthetic polymers, which are stable in physiological conditions. Irrespective of the polymer used, the imposed requirements are the same, i.e. effective protection against corrosion, high durability of coatings, and a simple manufacturing method with relatively low cost.

The classes of polymers commonly used as biomaterials are polyamides, polyolefins, acrylates, poly(methylmethacrylate) (PMMA), polyesters, and fluorocarbon polymers. As different polymers exhibit a wide variety of properties, they are used in a broad range of applications. Hard, rigid, and biostable polymers, such as PMMA, are used as bone cement, while flexible and fatigue-resistant polymers, such as poly(dimethylsiloxane) (PDMS), are successfully applied in ear and nose

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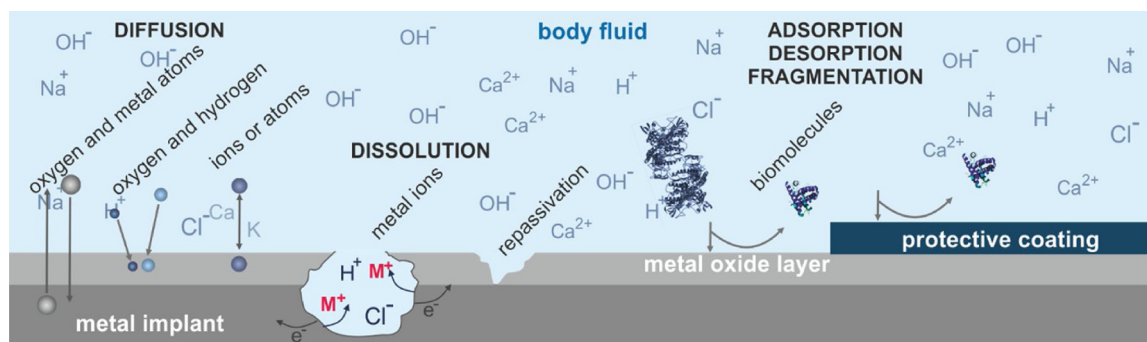


Fig. 1. Summary of basic processes occurring at a metal implant–body fluid interface with and without protective coating.

reconstruction. A wide variety of polymeric coatings have been investigated for the purpose of corrosion protection, i.e. polyaniline [12], ultrafine polytetrafluoroethylene [13], poly(L-lactic acid) [14], and parylene [15]. The selection of a polymer protective coating is based on the requirements and coating performance in a biological environment. One of the general strategies focuses on the applications of polymer coatings at implant surfaces to fabricate a robust and biocompatible interface. For these reasons, thin-film coatings have been extensively investigated for forming conformal barriers, protection enhancement, and surface functionalization.

Parylene is a generic name for the members of a xylylene family of polymers. They are classified as thermoplastic polymers formed on substrate surfaces using vacuum deposition polymerisation. They are linear and polycrystalline in nature. Parylenes have exceptional properties such as being stress free, truly conformal deposition, chemical inertness, bioacceptability, and optical transparency (index of refraction 1.6) [16].

Parylene (poly(para-xylylene)) was discovered by Michael Szwarc in late 1940s after he heated multiple gaseous compounds, including toluene and xylylene, and observed the degradation of the products and their decomposition rates as a function of temperature. In the case of xylylene, a film formed downstream from the high-temperature zone in the cooler extremities of glassware. The film could be removed as a tube-shape skin, which was described as ‘the skin of a small snake’. The film was formed by the polymerisation of the reaction product p-xylylene, which is currently known as parylene N. The process was commercialised in 1965 by W. Gorham and Union Carbide Company. Since then, Union Carbide developed over 20 types of parylenes, but only three of them are commercially valuable, i.e. parylene N and its halogenated derivatives and parylenes C (p-xylylene substituted with a single chlorine molecule) and D (substituted with two chlorine molecules). Parylene N is primarily used as a dielectric barrier, and it has a strong ability to prevent crevice penetration. The major drawback of this type of parylene is its deposition rate, which is the slowest among parylenes. Parylene C (poly(2-chloro-p-xylylene)) is widely used for biological applications because it was the first variant to attain the ISO 10933, USP class VI rating (the highest biocompatibility rating for plastics) and has excellent water and gas barrier properties compared to parylene N [17]. In recent years, fluorinated parylene HT (also named AF-4, SF) has gained more interest because it has the lowest coefficient of friction and highest penetrating resistibility compared to other commercially available parylenes [18]. The chemical structures of commercially valuable parylenes are summarised in Fig. 2, along with the overview of the scientific papers on a different type of parylenes published over the last 10 years. As illustrated in the pie chart, parylene C is the predominantly used (over 80 %) type of parylene, probably owing to its widespread availability and physicochemical properties.

It is essential for implant coatings to have a long-time resistance to corrosive body fluids, electrolytes, proteins, enzymes, and lipids. Therefore, parylene C coatings are ideal candidates for such applications because of their bioinertness and bulk barrier properties, which

are among the best organic polymer coatings. Parylene C is an organic coating free from solvents, catalysts, and plasticisers with a deposition layer thickness controlled down to 500 Å [19–21]. The bulk resistivity of parylene C is advantageously high because of its purity, low moisture absorption, and lack of trace ionic impurities. Apart from the chemical structure, the properties of parylene C are the consequence of its deposition method, i.e. chemical vapor deposition (CVD), which is described in detail below. This is a relatively simple and tunable process with numerous variants. Currently, this process is being extensively investigated to achieve novel parylene C films in terms of structure and surface morphologies [22–24].

In recent years, over 1000 papers have been published on parylene C, most of which are in the area of materials science (biomaterials). Parylene C is widely used as a biomaterial because of the unique properties of CVD-produced polymers, which can be applied to almost any substrate (Fig. 3).

The presented review addresses academia and industrial communities, covering fundamental and applied perspectives. It spans over several physicochemical issues related to the bulk and surface properties of parylene C. We summarise and discuss the last ten years (2008–2018) of research on parylene C in the context of biomedical applications. It is worth emphasising that the proportion of papers on parylene C as a biomaterial increased from 25 % in 2008 to 50 % in 2018. Despite the rapid development of this polymer in biomaterials science, there is lack of a state-of-the-art review of parylene C biomaterial coating applications. In this review, we collect, analyse and discuss the recent progress in the knowledge on this superior material with the aim of covering this gap.

2. Fabrication

Fabrication processes and their parameters and conditions strongly influence the final physicochemical properties of parylene C and thus its specific applications. In this section, we focus on several CVD-based variants of the fabrication method, which allows for the control of the desired properties of parylene C coatings. The methods were developed and discussed by leading groups in the field from Pennsylvania State University [22] and the University of Tokyo [25,26].

2.1. Chemical vapor deposition and its variants

In general, CVD polymerisation involves the following three key steps: the sublimation of a solid dimer and subsequent pyrolysis leading to gaseous monomer molecules, the transport of reactants from the vapor phase to a substrate, and polymerisation to thin and solid macromolecular films (see Fig. 4). The classic Gorham process allows for the full conversion of the dimer into the polymer thin film without any by-products. The mechanism of parylene C thin film CVD involves free radical polymerisation and consists of four sequential stages, i.e. monomer formation, chain initiation, propagation, and termination, which are described below.

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