



Removal of coliform bacteria from industrial wastewaters using polyelectrolytes/silver nanoparticles self-assembled thin films



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ABSTRACT

The aim of this work was to prepare and characterize thin films obtained from weak polyelectrolytes with silver nanoparticles and crosslinked with glutaraldehyde for the development of a new material to be used as a bactericidal agent in the treatment of industrial wastewaters. To evaluate the efficiency of these materials, the thin films were used for the treatment of an industrial wastewater sample. It was observed that, after 360 min of treatment, the thin films which presented the better inhibitory activity against *Escherichia coli* in the preliminary microbiological tests decreased by up to 93% the total coliforms present originally in the industrial wastewater sample. Considering the ease of application, low environmental impact, and satisfactory inhibiting action, the thin films developed in this work have a great potential to be used as a means of enhancing the removal of coliform bacteria from industrial wastewaters.

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Introduction

Industrial activities are the largest and primary source of water pollution due to the generation of effluents with distinct characteristics [1]. The pollutants commonly found in water include pesticides, volatile organic contaminants, heavy metals, nitrates, and radionuclides [2]. Moreover, water may contain many disease-causing organisms such as bacteria, viruses, and parasites which must be removed or inactivated for the safety of people who will eventually come into contact with the water during reuse operations. Several treatment techniques are used for this purpose, such as advanced oxidation processes [3,4], assisted peroxidation by ultraviolet radiation [5], ozonation [6,7], membrane bioreactors [8,9], hybrid treatment systems [10] and chlorination [11–13]. Chlorination is still the most widely used technique for water disinfection due to its ease of use and the wide range of products that are available [14–16]. However, the use of chlorine may result in the formation of toxic by-products, such as trihalomethanes and

haloacetic acids [17–23], which are potentially carcinogenic. Another disadvantage of the use of chlorine and its derivatives is the incomplete inactivation of protozoa, such as *Cryptosporidium* sp., which is responsible for several outbreaks of cryptosporidiosis worldwide [24–27].

Thus, the need for technological innovation to enable integrated water management cannot be underestimated. In addition, the development of new methods and strategies for the disinfection and reuse of industrial wastewater must be able to reduce or eliminate the chemicals used and minimize the amount of waste generated. The challenges of achieving appropriate disinfection without forming harmful disinfection byproducts via conventional chemical disinfectants and meeting the growing demand for decentralized or point-of-use water treatment and recycling systems calls for new technologies for efficient disinfection and microbial control. Although some nanomaterials have been used as antimicrobial agents in consumer products, including home purification systems, their potential for disinfection or microbial control in system-level water treatment has not been carefully evaluated [28–30].

Accordingly, nanotechnology holds great potential in advancing water and wastewater treatment to improve efficiency and to augment water supply through the safe use of unconventional

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water sources. The highly efficient, modular, and multifunctional processes enabled by nanotechnology are envisaged to provide high performance, affordable water and wastewater treatment solutions that rely less on large-scale infrastructures. Furthermore, the use of nanotechnology in water and wastewater treatment promises not only to overcome the major challenges faced by the existing treatment technologies but also to provide new opportunities for treatment that could allow for the economic use of non-conventional water resources, with the goal of expanding the water supply [31].

The potential of nanotechnology in disinfection and microbial control, including the use of nanoparticles such as Ag, ZnO, MgO, TiO₂, Ce₂O₄, carbon nanotubes, and fullerenes, has been studied by several authors [32–34]. Silver nanoparticles (AgNPs) are the most widely used antimicrobial nanomaterial because they exhibit activity against a wide range of micro-organisms, including fungi and viruses [35,36]. The inhibitory effect of silver is probably the sum of distinct mechanisms of action and is still partially understood [37]. A number of studies suggest that silver ions react with thiol groups of proteins [38] and play an essential role in bacterial inactivation [39]. Moreover, these ions have been reported to uncouple respiratory electron transport from oxidative phosphorylation, which inhibits respiratory chain enzymes or interferes with membrane permeability to protons and phosphate [40]. They can also interact with nucleic acids that probably results in impairment of DNA replication [41].

Silver ions and AgNPs also have inhibitory and lethal effects on Gram-negative and Gram-positive bacterial species, such as *E. coli* and *Staphylococcus aureus*, respectively [42]. The comparison of the antimicrobial effect of silver ions and AgNPs is an interesting field of research and some studies were performed in this direction. Morones et al. [39] showed that the overall effect of the AgNPs was different from the effect of silver ions alone. When nanoparticles were used, no evidence was found for the formation of a low density region as reported previously by Feng et al. [41] for silver ions. Instead, a large number of AgNPs were observed inside the bacteria. Jung et al. [43] also suggested that the thickness of the peptidoglycan layer of Gram-positive bacteria may prevent to some extent the action of silver species, since they found a higher inhibitory activity against *E. coli* than against *S. aureus* which also corroborates the conclusion of Feng et al. [41].

Although the applications of nanoparticles are increasing in many fields, concerns about their environmental and health impacts remain unresolved. The use of nanomaterials, such as AgNPs, is also likely to result in their release into aquatic ecosystems and may pose serious risks to the environment [44]. Even in its bulk form, silver is extremely toxic to fish [45], algae, some plants [46], crustaceans, and bacteria like nitrogen fixing heterotrophic and soil forming chemolithotrophic bacteria [47].

In order to reduce their mobility and prevent their appearance in the environment, the nanoparticles can be immobilized on different types of organic and inorganic matrices [48], which enables the development of various antibacterial materials for applications in water treatment devices [2,49,50]. The combination of AgNPs with magnetic nanoparticles in a core-shell structure for water purification is an interesting approach which allows recovering the nanoparticles from the system by applying an external magnetic field at the end of the treatment [51]. Other approaches have also been proposed in the literature, being the most of the studies related to the incorporation of AgNPs into polymeric membranes [52], ceramics [53], and thin films [54–57].

The layer-by-layer (LbL) self-assembly process is an efficient method for constructing thin films [58–66] and is a simple method to immobilize AgNPs with controlled size, as well as the amount of material deposited or introduced within the polymeric network. Furthermore, one of the most important advantages of these

materials is the possibility of its reuse in the process involving the disinfection of water.

In this regard, there are few studies in the literature focusing directly on polyelectrolyte's thin films produced by LbL technique with antimicrobial properties. Most studies found focuses on the application of these thin films as coatings for medical devices. Grunlan et al. [54] evaluated the effect of silver nitrate and cetyltrimethylammonium bromide (CTAB) on the antimicrobial efficacy of thin films of polyethyleneimine (PEI) and poly(acrylic acid) (PAA) prepared by LbL technique. The combination of the two antimicrobial agents provided a large scale bactericidal function on short time periods and an extended-release over longer time scales. Dvoracek et al. [56] also used CTAB in the preparation of multilayer polyelectrolyte thin films, together or not with poly(diallyldimethylammonium chloride) (PDDA) as the cationic layer, and poly(acrylic acid) (PAA) as the anionic layer. The influence of the thickness of the films, temperature and time of exposure on the formation of a zone of inhibition (ZOI) was evaluated. Recently, Kruk et al. [57] used negatively charged AgNPs nanoparticles in the preparation of multilayer polyelectrolyte films of polyethyleneimine (PEI). Their results indicated a relationship between the number of bilayers and the position of AgNPs in the film on the bacterial adhesion on the thin films.

In view of all these facts, the goals of this work are: (i) prepare thin films from weak polyelectrolytes solutions containing poly(allylamine hydrochloride) (PAH) and poly(acrylic acid) (PAA) in the presence of AgNPs through the LbL technique, and (ii) use these thin films for the treatment of an industrial wastewater. The obtained thin films were characterized by applying several instrumental techniques, including ultraviolet and visible (UV-vis) spectrophotometry, glow discharge optical emission spectroscopy (GD-OES), Rutherford backscattering spectrometry (RBS) and transmission electron microscopy (TEM).

2. Materials and methods

2.1. Preparation of thin films

To perform this work, glass substrates (VWR, Cat # 48312-027) were used as a support for the growth of the thin films. These substrates were cleaned in a neutral Extran solution (5% v/v) for 30 min at 60 °C and then were soaked in a solution of acetone: methanol (1:1 v/v) for 10 min. At the end of this procedure, the substrates were washed with deionized water (resistivity of <18.2 MΩ cm) obtained from a Milli-Q system (Millipore) and were left to dry at room temperature.

The thin films were prepared with NanoStracto Sequence equipment by sequential dipping of the glass substrates into aqueous solutions of polyelectrolytes with rinsing between each deposition step. PAH ($M_w = 70,000 \text{ g mol}^{-1}$, Sigma-Aldrich) was used as the polycation, and PAA ($M_w = 90,000 \text{ g mol}^{-1}$, Polysciences) was used as the polyanion. All polyelectrolytes were used as received, without further purification, and were prepared as 0.01 mol L^{-1} solutions (based on the repeat-unit molecular weight). The pH values of the polyelectrolyte solutions were adjusted to the desired pH with a 0.10 mol L^{-1} solution of HCl or NaOH.

The glass substrates were initially immersed in the cationic system (PAH) for 15 min. This immersion was followed by rinsing in deionized water for 2, 1 and 1 min to form the first monolayer. Next, the substrates were immersed in the anionic system (PAA) using the same procedure described above to form the second monolayer. This process was repeated until the desired number of layers was formed.

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