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Antibacterial silver treatments on polymeric membranes for fouling control and disinfection in water filtration

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ABSTRACT: Over the past several decades, a lot of emerging contaminants have been detected in water and wastewater effluents. Their release should be minimized since their presence in the environment can result in toxic effects for water and human life. Many different technologies have been used to remove contaminants from drinking water; among them, filtration is one of the most commonly used methods. This study investigated the antibacterial capability of silver water filters and their potential application in the reduction of bacterial fouling and proliferation in water treatment. Poly(ether sulfone) membranes commonly used in water filtration were coated with silver nanoparticles synthesized via the *in situ* photoreduction method. The morphology of the coating and the distribution of silver clusters were studied by scanning electron microscopy. The amount of silver on the surface was quantified by thermogravimetric analysis, and the silver released from the substrate was analyzed through inductively coupled plasma mass spectrometry. The antibacterial capability of the silver-treated filters was demonstrated through microbiological tests defined for the specific application on *Escherichia coli*, as the representative coliform bacterium and pathogenic microorganism commonly associated with contaminated drinking water. © 2016 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 43848.

KEYWORDS: coatings; functionalization of polymers; properties and characterization

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INTRODUCTION

The presence of contaminants, in surface water and wastewater, represents a serious concern in relation to their toxic effects on the environment and human health. The World Health Organization and UNICEF have reported that 1.1 billion people do not have access to improved drinking water sources.[1,2] About 80% of communicable diseases in the world are waterborne.[3] In order to reduce the incidence of waterborne diseases and to meet the increasing need for potable water, the removal of pathogenic organisms is absolutely necessary. Various technologies are used to remove bacteria from drinking water, such as the addition of chemicals, coagulation, flocculation, sedimentation, filtration, and disinfection.[4] The use of nanosilver particles in drinking water treatment can play an important role in reducing microbial growth.[5] The properties and the antibacterial activity of silver have been known since ancient times. Silver ions are used as biocides for cleaning and disinfecting drinking water storing products, and modern medicine adopts silver as an antibacterial agent on various supports, for local application in treating wounds, burns, eye-infection prophylaxis, dentistry, and so on.[6,7] Silver as an antibacterial agent has numerous advantages, such as its high biocompatibility, excellent resistance to sterilization conditions, effectiveness on different bacteria, and the long-term durability of its antibacterial effects.[8] Silver ions interact with bacterial proteins by combining the thiol groups to cause structural changes in the cellular wall and bacterial membranes, which finally leads to the destruction and death of the bacteria.[9]

Poly(ether sulfone) (PES) filters are widely used in the form of membranes because of their high glass-transition temperature and their high mechanical, thermal, and chemical resistance.[10] This polymer contains repeated ether and sulfone linkages alternating between aromatic rings, which provide a high degree of molecular immovability, creating creep resistance, high rigidity, superior strength, and dimensional stability.[11] The use of PES membranes in water-treatment applications has been widely established and is nowadays considered as the standard technology.[12]

During filtration, smaller suspended particles and dissolved macromolecules pass through the membranes, while the bigger molecules are mostly rejected.[13] The deposition of some of these rejected molecules, such as colloids or bacteria, on the

Additional Supporting Information may be found in the online version of this article.

membrane surfaces causes considerable fouling of the membrane.[14] In particular, biofouling is caused by the attachment and growth of bacteria and accumulation of the bacterial metabolic products such as extracellular polysaccharides, proteins, and lipids on the membrane surface.[15] For this reason, the filters can represent a reservoir of bacteria and a potential risk for contamination in water filtration. Membrane fouling results in deterioration of membrane performance and ultimately reduces membrane life.[16]

In this work, silver-treated filters have been developed to reduce biofouling formation and the risk of bacterial infections, commonly resulting during bathing, washing, and drinking and in the preparation of food. Poly(ether sulfone) filters were treated with silver nanoparticles through a deposition technology based on an *in situ* photoreduction reaction. Previous works demonstrated durable antimicrobial properties and the ease of technology transfer on an industrial scale.[17–19] Particularly, great attention has been paid to the application of antimicrobial silver treatments to indoor environments,[18–20] for the development of antibacterial natural leather and air filtration units for the prevention of cross transmission and the reduction of bioaerosolization of human pathogens into environments such as in public places, trains, and buses.[19] Environmental aspects were also the focus of a previous work where high-density polyethylene (HDPE) nets for agriculture were deposited with silver coatings using the same silver-deposition technology, in order to protect plants and crops from diseases associated with microorganisms.[21] The potential of the technology for the specific application of water filtration with a PES membrane as substrate has never been investigated before and required an optimization to define the proper process parameters. Then the efficacy of the adopted technology was verified through agar diffusion tests and bacterial enumeration of *Escherichia coli*, selected as an indicator of fecal contamination of water. To confirm the bactericidal effect of the silver-treated filters and to reproduce the real conditions, the bacterial suspension was filtered through the silver-coated PES membrane. The silver-treated filters were also characterized through scanning electron microscopy, thermogravimetric analysis, and inductively coupled plasma mass spectrometry to examine the stability of the silver coating and the silver release.

EXPERIMENTAL

Silver Deposition on PES Filters

Poly(ether sulfone) membranes were treated with silver according to a patented technique developed at the University of Salento (Lecce, Italy) and based on the *in situ* synthesis and deposition of silver nanoparticles.[22] The silver treatment was obtained by impregnating PES membranes (molecular weight cutoff of the membrane 1000 Da to 300,000 Da; molecular weight of PES 58,000 gr/mol; tensile strength 94 MPa; tensile modulus 2.6 GPa) in an alcohol solution containing a photoreducing agent (methanol 99.5%) and a silver precursor (silver nitrate 0.5 wt/v %). Afterward, the wet substrates were exposed to ultraviolet (UV) irradiation ($\lambda = 365$ nm) for 20 min on each side, in order to induce the formation of silver particles on the surface of the material. The concentration of the silver precursor (0.5 wt/v %) and the UV exposure time (20 min) were defined through the optimization study reported in the Supporting Information, by evaluating the effect of the three percentages of silver that were tested (0.1, 0.5, and 1 wt/v %) and the three UV exposure times (5, 10, and 20 min) on the antibacterial capabilities of the samples.

The photoreduction process is strictly related to the presence of methanol, which is used as both solvent and reducing agent. However, silver solutions also containing water can be adopted according to the nature of the material in order to reduce the treatment costs.

After the treatment, the substrates were washed carefully with deionized water and wiped before testing to remove any trace of unreacted silver nitrate.

Silver Release

The release of silver from the substrates was calculated using inductively coupled plasma mass spectrometry ICP–MS (iCAP Q, Thermo Scientific, Waltham, Massachusetts, United States), in order to assess the adhesion of the coating to the filter and to verify that silver was not released into the environment. For this characterization, the silver-treated samples (0.02 g) were immersed in tap water (1 ml) and incubated at 37 °C up to 720 h. Then 500 μl of water was extracted at selected time points with a sterilized syringe and were analyzed after acid digestion with nitric acid 1% (v/v). This was followed by appropriate dilution of the test samples, using silver solutions with known concentrations as standards.

Antibacterial Activity

As part of the optimization study performed to define the process parameters for the silver treatment, antibacterial tests were performed through agar diffusion tests on samples treated with different concentrations of silver and different UV exposure times (Supporting Information). Then, the antimicrobial capability of the selected silver-treated PES membranes was determined through further microbiological characterizations, aiming to investigate the efficacy of the silver coating in reducing bacterial fouling on the surface of the filter and also in reducing the presence of bacteria in the filtered water. For this purpose, the following microbiological tests were performed for the specific application. The antibacterial capability of the silver coating deposited on the filters was evaluated through agar diffusion tests; the efficacy of the silver coating in reducing the presence of viable bacteria in the filtered water was quantified through both bacterial enumeration and optical density (OD) readings; the capability of the materials to reduce the viability of bacteria adhered on the filters was evaluated through macroscopic growth on agar plates and bacterial enumeration after water filtration; the durability of the silver coating was finally analyzed through antibacterial tests on the samples used for ICP–MS, after incubation in tap water at 37 °C.

Qualitative Analysis. Agar diffusion tests were performed according to Standard SNV 195920-1992 on *Escherichia coli*, using Luria Bertani (LB) as the bacterial growth medium. After UV sterilization for 15 min for each side, the samples were placed in contact with bacteria on agar plates and incubated overnight at 37 °C. The measurement of the antibacterial capability, related to the width of the area of inhibition to bacteria growth observed around the samples, was defined according to

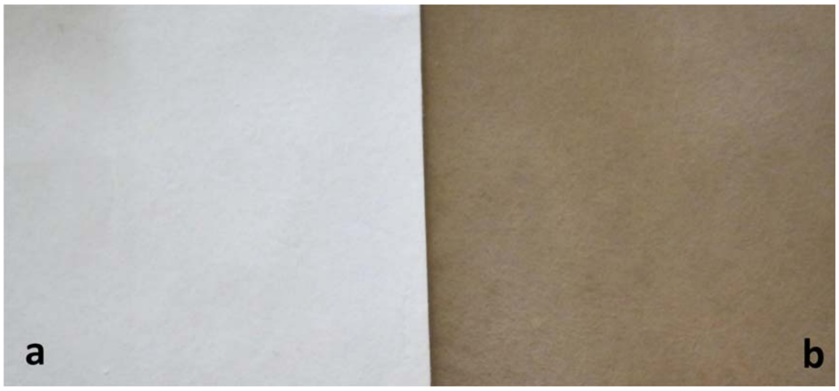

Figure 1. Visual comparison of (a) untreated PES membrane and (b) PES membrane treated with 0.5% Ag. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the antibacterial levels defined by the standard. An area of inhibition to bacterial growth larger than 1 mm indicated good antibacterial capability; in contrast, samples fully covered by bacteria indicated insufficient activity.

Filtration Experiment. A quantitative analysis was also conducted on *E. coli* to measure the ability of the treated sample to reduce the bacterial concentration in the filtered solution. In order to examine the bactericidal effect of the silver-treated filters, 1 mL of bacterial suspension (inoculating cell density 3.9×10^6 Colony Forming Units [CFU] per mL) was filtered through the silver-coated PES membrane. The filtered bacterial suspensions were used to perform both the optical density (OD) measurement ($\lambda = 600$ nm) and the bacterial enumeration through the serial dilution method. A suspension filtered with an uncoated PES membrane was used as control. The OD measurement was performed through spectrometric analysis (Visible spectrophotometer V-1200; VWR, Radnor, Pennsylvania, United States), and the absorbance measured was related to the number of bacteria (1 OD = 10^9 CFU/mL). For the serial dilution method, the samples of filtered water were diluted in phosphate-buffered saline, and 100 µL of each dilution was plated on LB plates. After incubation overnight at 37 °C, the bacterial colonies were counted in order to quantify the concentration of bacteria and the reduction of their amount that was due to the presence of silver.

Quantitative Evaluation of Cells Retained by the Filter. This experiment was performed to verify whether the filter material retained viable microbial cells. As describe above, 1 mL of bacteria suspension was filtered through the PES membranes. In order to quantify the presence of bacteria retained on or inside the filter, samples were placed in a liquid medium (LB) after the filtration experiment, vortexed for 1 min, and incubated at 37 °C overnight. After incubation, the concentration of bacteria detached from the filters was determined through the bacterial enumeration test. As reported in the Supporting Information, a qualitative evaluation was also performed by placing the silver-coated and uncoated filters onto uninoculated agar plates after filtration.

Antibacterial Durability. The samples used for ICP–MS after incubation in tap water at 37 °C were analyzed at the time points defined for ICP–MS through agar diffusion tests in order to verify the durability of the silver coatings. The samples were placed on agar plates and, after incubation, the area of inhibition to bacterial growth around and beneath the samples was evalutated.

SEM/EDX Analysis

Silver-treated samples were analyzed by scanning electron microscopy (SEM, Zeiss Evo, Oberkochen, Germany) to study the morphology of the silver coating and the distribution of silver clusters. Energy dispersive X-ray spectroscopy (EDX, Bruker, Billerica, Massachusetts, United States) was used to confirm and quantify the presence of silver particles on the surface of the samples.

Thermogravimetric Analysis

Thermogravimetric analysis (TGA, Mettler Toledo, Columbus, Ohio, United States) was adopted to measure the amount of silver deposited on the substrate. The samples were heated with a heating rate of 10 °C/min from 25 °C to 1200 °C in a nitrogen flow (50 mL/min). The solid residue at the end of the test represented the incombustible silver coating, and it was quantified with respect to the untreated sample. Samples were tested in triplicate, and the results are expressed as mean ± standard deviation.

RESULTS

Silver Deposition on PES Filters

The PES membranes treated with silver were characterized by an evident change in color from white to brown that was due to the presence of uniformly distributed silver clusters. As shown in Figure 1, the comparison between untreated [Figure 1(a)] and silver-treated PES membranes [Figure 1(b)] was characterized by the typical darkening associated with the presence of silver.

Silver Release

The release of silver calculated through ICP–MS is reported in Figure 2, where the maximum value was 0.2461 ± 0.003 ppb. The results were consistent with the average silver concentrations reported for natural waters (0.2–0.3 µg/L) and are well below the concentrations reported for water treated with silver (50 µg/L or higher).[23]

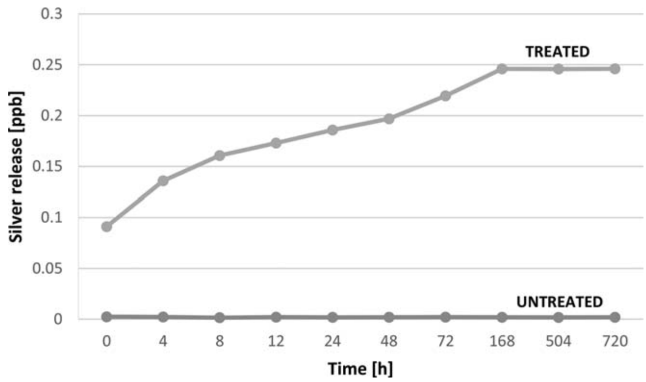

Figure 2. Release of silver analyzed by inductively coupled plasma mass spectrometry (ICP–MS).

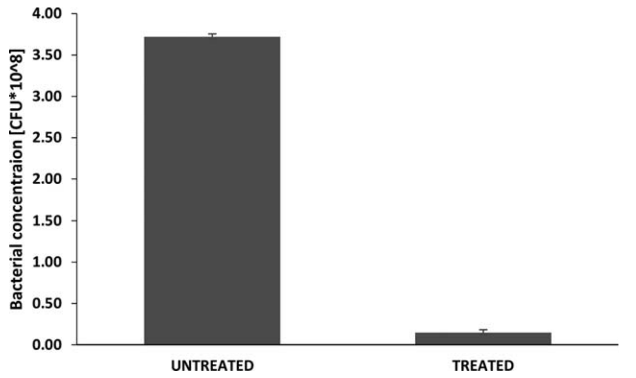

Figure 4. Quantitative analysis of the bacterial concentration of a solution after filtration with an untreated and a silver-treated filter. In presence of the silver-treated sample, a significant reduction of bacterial proliferation can be observed.

Antibacterial Capability

Qualitative Analysis. The results obtained by the optimization study on the definition of the process parameters are reported in the Supporting Information (Figure S1), where different antibacterial capabilities can be observed as a function of the different silver concentrations and the UV exposure times tested.

In Figure 3, a clearly defined bacterial-free zone larger than 1 mm can be observed around the selected sample treated with 0.5 wt/v % of silver and exposed to UV for 20 min. The width of the inhibition growth area confirmed the good antibacterial effect, according to the levels of antibacterial capability reported in the standard.[8]

Filtration Experiment. The qualitative analysis of the disk diffusion test was confirmed by the bacterial enumeration reported in Figure 4, where the concentration of bacteria was $3.72 \times 10^8 \pm 0.003$ and $0.15 \times 10^8 \pm 0.002$ CFU/mL, respectively, for the untreated and silver-treated samples. The optical density reading demonstrated a bacterial reduction of 100% for the treated sample and of 7% for the untreated sample, thus confirming the data obtained by bacterial enumeration. The quantification of the bacterial concentration, evaluated after filtration, demonstrated that the silver-treated filter is able to reduce the amount of bacteria in the filtered suspension.

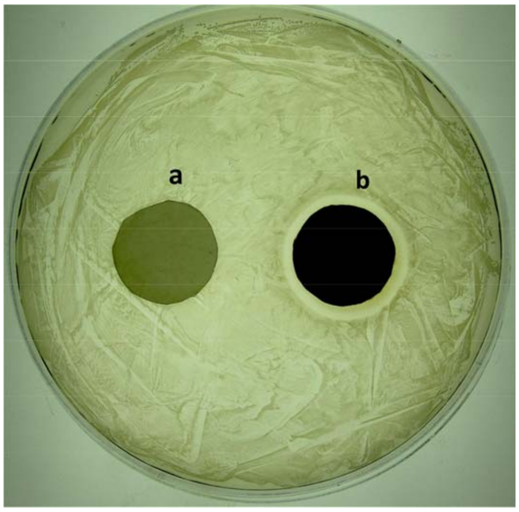

Figure 3. Agar diffusion tests on *Escherichia coli*: (a) untreated sample; (b) sample treated with 0.5% Ag. The well-defined area of inhibition to bacterial growth can be observed around the silver-treated sample. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Quantitative Evaluation of Cells Retained by the Filter. The bacterial enumeration test demonstrated a reduction of 70% in the amount of bacteria retained by the treated filter, compared to the untreated sample. The results obtained by the qualitative tests are reported in the Supporting Information (Figure S2), where no bacterial growth can be observed in the presence of the silver-treated sample.

Antibacterial Durability. The antibacterial durability of the silver-treated PES membranes was tested through agar diffusion tests on samples subjected to silver-release experiments. In Figure 5, the permanence of a zone of inhibition of bacterial growth can be observed around the silver-treated samples, even after long-term incubation in water.

SEM/EDX Analysis

The distribution of the silver particles on the water filters was analyzed by scanning electron microscopy. The results of the SEM analysis are reported in Figure 6, and the presence of silver particles visible on the treated samples [Figure 6(b), arrows] was confirmed by further EDX analysis. The relative percentage of silver calculated by EDX in relation to the chemical composition of the substrate deposited was 1.37% on the treated sample (Figure 7).

Thermogravimetric Analysis

TGA was performed in order to quantify the amount of silver deposited on the treated samples. The average amount of silver was calculated as the difference between the solid residues obtained for the silver-treated sample and the untreated one. The samples were tested in triplicate, and the percentages of silver deposited were $1.61 \pm 0.01\%$.

DISCUSSION

Industrial development has had profound negative effects on the environment because of the presence of contaminants in the

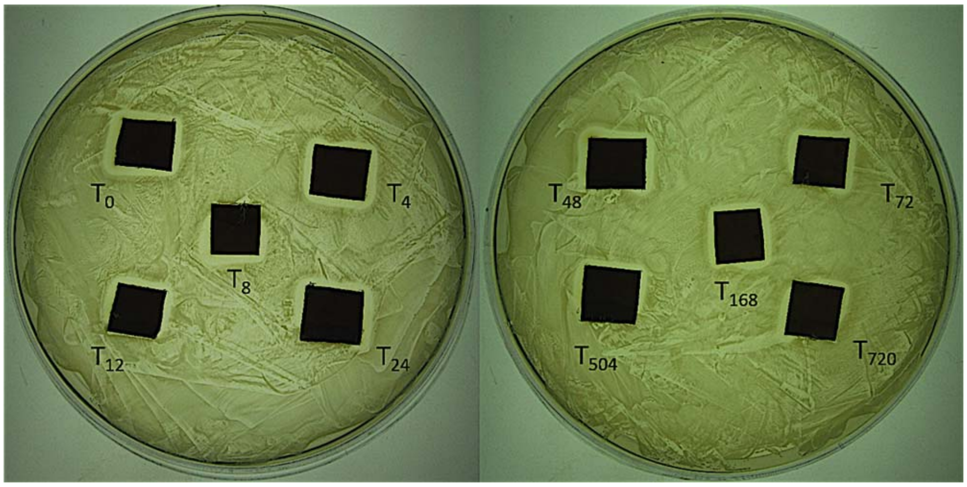

Figure 5. Agar diffusion tests on *Escherichia coli* on silver-treated samples after incubation in water at different time points. Around all the samples a well-defined area of inhibition to bacterial proliferation can be observed. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

effluents of urban wastewater purification plants.[24] During the past few decades, several investigations have been carried out concerning the use of different types of water filtration to promote water disinfection. The ineffectiveness of conventional purification systems entailed the need to implement systems with advanced treatments and to improve the removal of contaminants. However, advanced treatment processes for wastewater are rarely used because of the excessive investment costs.[25] Some authors have reported the possibility of modifying PES membranes for fouling control with carbon nanotubes or ZrO_2.[26,27] The current work aims to provide a good strategy for developing antibacterial water filters through an innovative and low-cost silver-deposition technology without adverse consequences for humans and the environment.

The use of silver to control the contamination of liquids and to reduce the spread of diseases can be traced to ancient times, though only in recent decades have studies revealed the biochemical reactions of silver in the inactivation of bacteria.[28,29] Today, the increasing health-care costs and the resistance demonstrated by many microorganisms to multiple antibiotics have encouraged the application of silver in the biomedical field for the production of antibacterial products.[30,31] In environmental applications, the use of silver has huge potential in the control of pollution thanks to the broad-spectrum antimicrobial activity demonstrated against a wide range of microorganisms and the possibility of reducing the use of chemical products in disinfection. In addition, an important adavantage of the deposition technology adopted here is the excellent adhesion of the silver coating to the PES substrate, which avoids the dispersion of silver in the environment. Moreover, the silver-deposition treatment does not affect the bulk properties or morphology of the substrate and does not affect its porosity, thus also indicating that the filtration properties of the material remain unchanged. In order to develop a product with good antibacterial properties and to contain the production costs, the process parameters of the silver-deposition technology were defined for the specific application and the polymeric substrate through an optimization study focused on the definition of the concentration of the silver precursor and the UV exposure time. The choice of the process parameters was made by evaluating their effect on the antibacterial capability of the samples. Thus, samples treated with 0.1, 0.5, and 1 wt/v % of silver were exposed for the same amount of time to UV light (20 min), and then lower UV

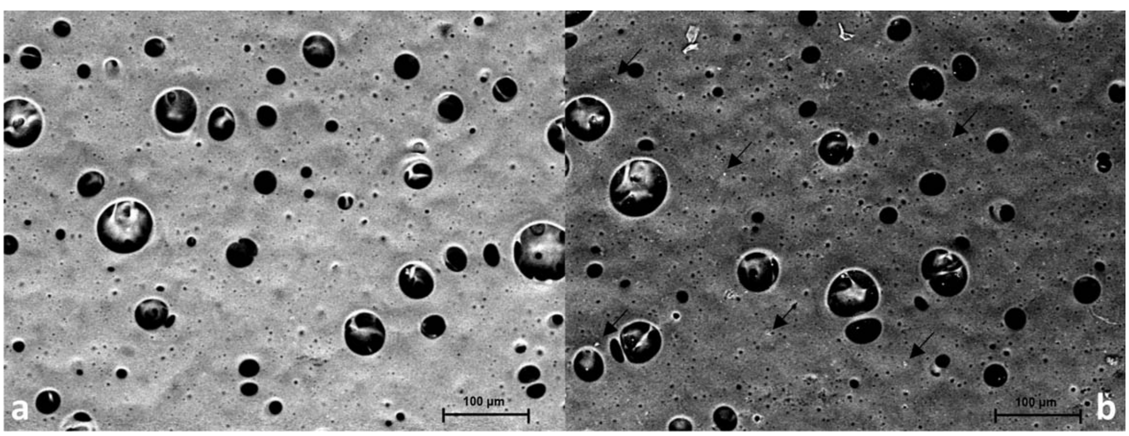

Figure 6. SEM images of the PES membranes: (a) untreated sample; (b) treated sample. The arrows indicate some silver clusters.

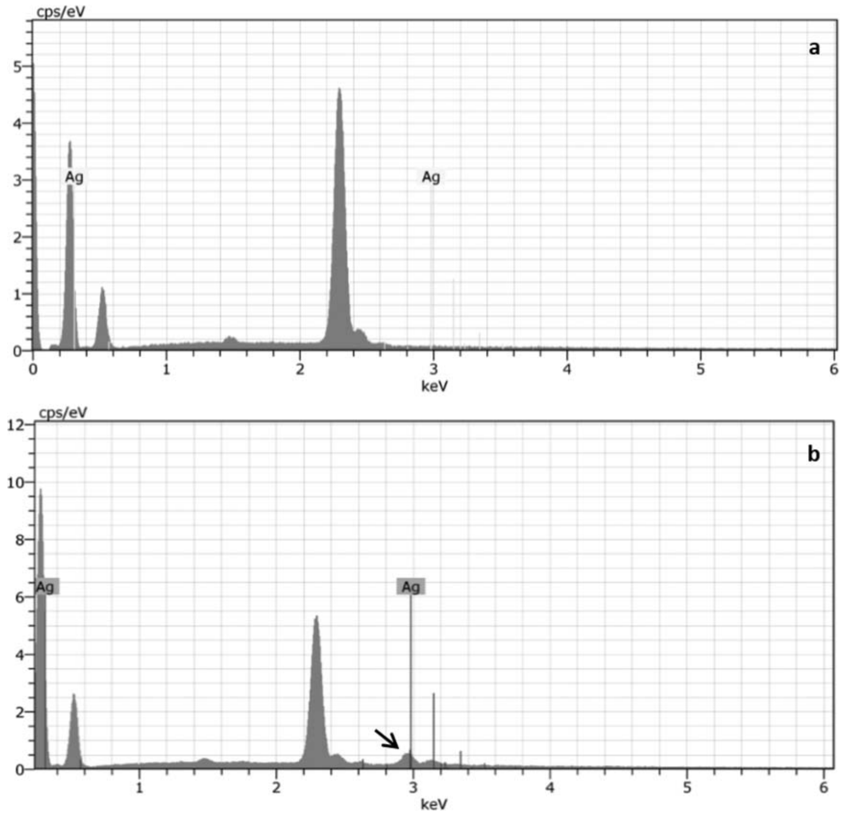

Figure 7. EDX analysis of (a) untreated and (b) silver-treated PES filters.

exposure times were tested (5 and 10 min). As reported in the Supporting Information (Figure S1 and related comments), the best results in terms of costs and effectiveness were obtained by the samples treated with 0.5 wt/v % $AgNO_3$ and exposed to UV for 20 min, and, for this reason, they were adopted for further analyses (Figure 1). The samples were characterized using ICP–MS, and the silver release in water was evaluated for up to one month. Interestingly, the results obtained indicated very low values of silver release (Figure 2), which was consistent with the percentages of silver calculated in natural water and was lower than the values recommended by the regulations.[23] Moreover, after the time point corresponding to one week (168 h), the values of silver release remain stable, thus confirming the excellent adhesion of the coating to the substrate and suggesting the development of an environmentally friendly product.

As the most significant microorganism responsible for water contamination and waterborne disease, *Escherichia coli* has been selected for the microbiological characterization, with the aim of determining the efficacy of the treated filter in removing bacteria from water and onto the surface of the filter. The agar diffusion tests reported in Figure 3 demonstrated the capability of the coating in inhibiting the bacterial growth. The area of inhibition to bacterial growth around the silver-treated sample was larger than 1 mm and, according to Standard SNV 195920-1992, indicates a good antibacterial capability. On the other hand, the untreated samples were completely colonized by bacteria, as expected. This result was confirmed by the bacterial enumeration test and optical density readings on the bacterial suspension filtered through the silver-treated and untreated samples. The results demonstrated an impressive reduction of the bacterial concentration (Figure 4), and the evaluation of the bacterial adhesion onto the polymeric substrate also indicated significantly decreased biofouling in the presence of the silver treatment (Supporting Information, Figure S2).

The long-lasting antibacterial activity was demonstrated by agar diffusion tests performed on samples incubated in water up to one month. As visible in Figure 5 by observing the width of the area of inhibition to bacterial growth, all of the samples maintained the antibacterial capability for the whole experiment. These results were consistent with those obtained by ICP–MS analysis and confirmed the excellent adhesion of the coating onto the substrate and the durable antibacterial capability of the silver coating.

In terms of the presence and distribution of the silver particles on the polymeric membrane, SEM–EDX (Figures 6 and 7) and TGA analyses confirmed the presence of silver deposited on the PES membrane. Along with the porous structure of the membrane, silver nanoparticles were observed on the surface by SEM [Figure 6(b), arrows], and the peak for silver can be observed in the EDX graph [Figure 7(b)]. Previous studies on the technology adopted in this work have investigated the size of the particles deposited on the silver-coated copper grid using transmission electron microscopy (TEM), which demonstrated the presence of either single silver nanocrystals with diameters below 2 nm and aggregates of a few nanocrystals.[32]

CONCLUSIONS

This study investigated the antibacterial capability of silver-treated polymeric membranes used in water filtration. Poly (ether sulfone) filters were treated with silver by using a technology based on the photochemical deposition of silver particles. The process, involving the impregnation of the materials in a silver solution and subsequent UV illumination, was effective in preventing water contamination during water filtration, and the retention and viability of bacteria on the filter surface were also significantly reduced. As an important aspect of the study, ICP–MS measurements indicated extremely low values of silver release into the water even after one month, thus suggesting no significant effects for human health and the environment.

The adopted surface-engineering process may be useful for the production of water filters able to reduce the risk of contamination and the dissemination of waterborne diseases. The results obtained, along with the simplicity of the adopted technology and the ease and the low-cost aspects of production, suggest that the materials developed are an interesting alternative to conventional treatments used in water disinfection, with no dangerous effects for human health and the environment.

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