

Quaternary Ammonium Salts of Poly(N,N-dimethylaminoethyl methacrylate) as an Efficient Antibacterial Agent for Polylactide Textiles

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Abstract- Polylactide nonwoven was modified by poly(N,N-dimethylaminoethyl methacrylate). The surface was quaternized with crosslinking. A two-step method was used: first spraying the polymer, then cross-linking with ethylene bromide. The modification was confirmed by infrared spectroscopy and solubility tests. The antibacterial activity of unmodified nonwoven, nonwoven with a deposited layer of PDMAEMA and nonwoven quaternized with PDMAEMA was evaluated against *Escherichia coli* and *Staphylococcus aureus*. The antibacterial results showed that these materials could achieve up to 100% efficiency.

Keywords: poly(N,N-dimethylaminoethyl methacrylate), antibacterial properties, surface modification, quaternary ammonium salt

I. INTRODUCTION

To improve competitiveness, the textile industry has focused its research on the development of multi-functional fabrics with high added value and applications in different areas. Example of such materials are biofunctional textiles, including antimicrobial products, obtained through special finishing applications or fiber modifications [1].

Textiles are excellent media for developing microorganisms, especially those made of natural fibers, which can act as substrates for the development of pathogenic or odour-generating bacteria and fungi. These materials offer an ideal environment for microbial growth because of their large surface area and their capacity to retain oxygen, heat and moisture, as well as nutrients from spillages and body exudates [2,3]. The prevention of microbial attacks on textiles has become increasingly

significant for their users. Quaternary ammonium salts (QASs) have been used as antiseptics since Domagk [4] established this class of antimicrobial agents. These compounds have a positive charge at the nitrogen atom and can affect microbes by causing damage to cell membranes, denaturing proteins and disrupting the cell structure [5].

Through the inactivation of bacterial cells, the quaternary ammonium group remains intact and retains its antimicrobial effects as long as the compound is attached to a textile [6]. QASs have great antimicrobial activities against bacteria, fungi and viruses, and some scientific applications of quaternary ammonium salts for textiles are currently being realized (Bioguard) [7-9].

Textile materials used in hospitals, such as gowns, drapes, masks, sheets, and pillows, can be the bases or media of cross-infections, and antimicrobial textiles should prevent or minimize infections or the transmission of disease [10].

Polylactide is a biodegradable material that is compatible with soft and hard tissue, and has applied to the design of various devices used in the biomedical field. Currently, it is known that the hydrophobic nature of polyesters allows the adhesion of pathogenic microorganisms onto materials prepared for them [11].

Poly(N,N-dimethylaminoethyl methacrylate) (PDMAEMA) has been the subject of research attention for a long time. PDMAEMA has excellent antimicrobial properties [1,12-14]. PDMAEMA is a linear molecule and is soluble in aqueous solutions. PDMAEMA has been used as a flocculant [15], a carrier for drug delivery systems [16], an ion exchange resin [17-19] and mordant for ink printing [17]. PDMAEMA can also be used to prepare composite membranes for blood purification [20], cationic/anionic mosaic membranes for desalination [21] and pervaporation membranes for the purification of ethyl tert-butyl ether [22]. Recently, it has been shown that PDMAEMA can be used to produce a nonwoven [23]. Regarding the structure of PDMAEMA, the polymer contains a tertiary amino group on the side chain of the

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monomer unit, which gives it a relatively high reactivity and allows it to be alkylated or quaternized to form a quaternary ammonium salt. Very recently, researchers have demonstrated an easily achieved method for the surface modification of polylactide nonwovens with a thin PDMAEMA film [12]. It has also been shown coating fibers with PDMAEMA result in the formation of a new material that possesses antimicrobial properties [12]. The application of quaternized PDMAEMA has only been described in a grafted form on silicon nanowires and resulted in reduced *Escherichia coli* adhesion and cell numbers [24].

PDMAEMA can also be autocrosslinked, due to chain transfers to the polymer; however, the autocrosslinking is too weak to prevent PDMAEMA from dissolution in aqueous solution. PDMAEMA can also be crosslinked by heat treatment. A previous study [20] described the preparation of a membrane, using PDMAEMA as the active layer and a cuprammonium regenerated cellulose hollow fiber as the substrate, and in a different patent [22], the authors prepared a composite membrane by depositing a PDMAEMA-water-ethanol solution on a porous polyacrylonitrile layer, which was further supported on a non-woven polyester fabric. Generally, the interest of the scientific community in PDMAEMA is increasing [25,26]. A frequent problem encountered when applying PDMAEMA modifiers is that this polymer is soluble in many common solvents (including water and ethanol). This study is focused on creating a crosslinked, quaternary, ammonium salt of PDMAEMA on the surface of polylactide nonwoven. Using the polymer in this form should result not only in better antibacterial properties but also in the insolubility of the external layer, which would increase the possible applications.

II. EXPERIMENTAL

A. Materials

Polylactide nonwoven (sample 0) was prepared using the needling technique at the Department of Material Science and Textile Metrology (Lodz University of Technology, Poland), with a surface weight of 39.1 g/m² and an average filament diameter of 26.5 μm.

PLA fibers were produced by FET (Taiwan) under the name PLA Type SLN-2660D, 0.66 tex, 64 mm. The average molecular weight of the PLA material was 83,000 g/mol ($M_w/M_n=1.4$).

PDMAEMA was prepared by the polymerization of dimethyl aminoethylmethacrylate (Aldrich), initiated with azobisisobutyronitrile (Aldrich). The average molecular weight ($M_w=63,000$ g/mol) was determined by gel chromatography.

B. Modification Procedures

B.1. PDMAEMA Deposition

The primary object of the modification was polylactide nonwoven (sample 0), which was modified by spraying a PDMAEMA water solution (1%) to obtain a deposition of 1.85% by weight (sample 1) or 3.71% of the introduced polymer (sample 2).

B.2. Quaternization with Crosslinking

Sample 3 was obtained by introducing sample 2 into an excess of ethylene bromide (10 w/w% ethanol solution) at room temperature for 24 h.

C. Characterization

C.1. Weight Tests

The amount of modifier in the nonwovens in the first step of modification (PDMAEMA deposition) was determined by the weight control method. In the second stage (PDMAEMA quaternization with crosslinking), solubility tests were performed to confirm the modification effect. All samples (approximately 1 g) were previously conditioned in a dry desiccator (0% humidity, room temperature) and then were introduced into distilled water for approximately 30 min, with gentle shaking. The samples were dried by storing overnight in room temperature conditions and then placing in a dry desiccator for 24 h. The sample weights were controlled before and after the procedure. The whole procedure was repeated at least ten times.

C.2. SEM Microscopy

Scanning electron microscopy was combined with energy-dispersive X-ray spectroscopy analysis. The chemical and morphological analyses of surface modified nonwoven fabrics were performed using a scanning electron microscope (SEM) Nova Nano-SEM 230 from FEI, Hillsboro, OR, USA (SE Detector, voltage: 15 kV, rough vacuum: 0.3 Torr).

C.3. FTIR Spectroscopy

The FTIR spectra were collected using Perkin-Elmer 2000 FTIR instrument. A Perkin-Elmer specular reflectance variable angle accessory (VASR) was used to gather the reflectance FTIR spectra at 60° (16÷700 scans). The unmodified nonwoven fabric was used as a background when creating the spectra of the modified nonwoven fabric.

C.4. Antibacterial Tests

The antibacterial activity was determined according to the ASTM E2149-10: Standard Test Method for Determining the Antimicrobial Activity of Immobilized Antimicrobial Agents under Dynamic Contact Conditions (International,

TABLE I
COMPOSITION OF DEPOSITED LAYERS

| Unmodified | | Type of layer (sample number) | | |
|------------|---------------------|-------------------------------|----------------------------------|--|
| 0 | 1 | 2 | 3 | |
| PLA | PLA-PDMAEMA (1.85%) | PLA-PDMAEMA (3.71%) | PLA-PDMAEMA (3.71%)+crosslinking | |

2010). The nonwoven samples were each added to 50 mL of bacterial inoculum and shaken for 1 h in Erlenmeyer flasks. After shaking the inoculum with the specimen for 1 h, the inoculum was immediately serially diluted, plated in Petri dishes, and cultured at 35 °C for 24 h to determine the number of colony forming units per mL (CFU mL⁻¹). The bacterial concentration of the solution at the “0” time point was also determined by performing serial dilutions and a standard plate count from an “inoculum only” sample flask. The results for the antibacterial test are reported as the percentage reduction (R%), calculated as:

$$R\% = \frac{B - A}{B} \times 10\% \quad (1)$$

Where, A is the CFU mL⁻¹ for the flask containing the modified nonwoven after the specified contact time and B is the CFU mL⁻¹ for the inoculum only flask after the specified contact time.

III. RESULTS AND DISCUSSION

A. Modifications and Confirmation of the Modified Structure

The modification process was a two-step procedure: in the first stage (PDMAEMA deposition), we used the spraying technique and we obtained physical bonding; and in the second stage (quaternization with cross-linking), we used deposition from a concentrated ethanol solution. Finally, we obtained nonwovens with different architectures (Table I).

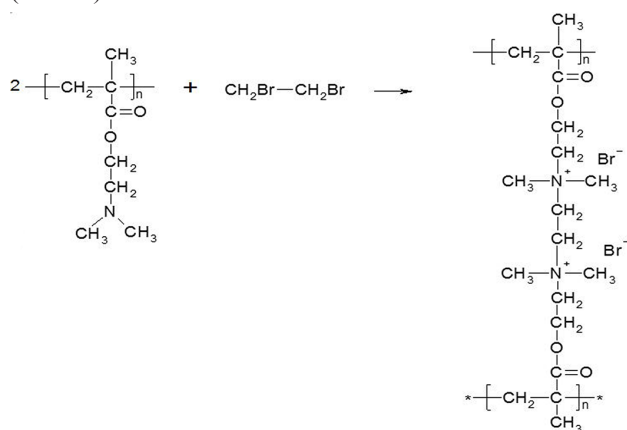


Fig. 1. PDMAEMA after crosslinking with quaternization.

After crosslinking with quaternization, we expected an insoluble structure on the surface, which is shown in the schematic below (Fig. 1).

According to the procedure proposed by us, quaternization with crosslinking is carried out *in situ* at the surface of the fiber. Bifunctional crosslinking compounds allow the formation of transverse bonds between macromolecules. As a result, an insoluble two-dimensional network is obtained. Textiles, however, are a flat product with an unstable, variable structure due to the existence of weaves that hold the fibers together. This clearly means that not all of the fiber surface is available for modification. In addition, during the use of the product, the mutual positions of the fibers change, causing a change in the availability. In the first stage of a two-step procedure, the linear polymer is introduced onto the fiber surface and temporarily bound by physical interactions. Then, the crosslinking procedure allows for the creation of a stable structure in places accessible to the polymer in the first stage of the process.

The structure of the modified samples was confirmed using reflectance FTIR spectroscopy, SEM microscopy and solubility tests. Introducing an external modifier at the 2-4% level could not provide a clear spectrum using transmission FTIR spectroscopy. Therefore, we used reflectance spectroscopy, which provided information regarding the external structure of the modified material. Parts of the reflectance FTIR spectra are shown in Fig. 2.

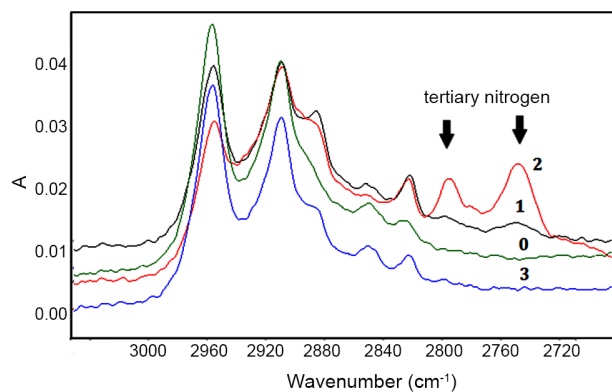


Fig. 2. Part of the reflectance FTIR spectra after different stages of modification (0-unmodified sample, 1 and 2-samples with different amounts of PDMAEMA, 3-sample after quaternization with crosslinking).

Regarding the spectra of the modified materials, important differences between the samples can be observed. For the samples with different amounts of deposited PDMAEMA (samples 1 and 2), a new double signal appears that is located near $2,800\text{ cm}^{-1}$ and is connected with the tertiary nitrogen $-N(\text{CH}_3)_2$, when compared with the unmodified polylactide (sample 0). In the spectrum of sample 2, this peak is larger because the amount of deposited polymer is larger. The spectrum of sample 3 shows the disappearance of the tertiary nitrogen signal,

which confirms its quaternization. The regularity of the modification and the roughness of the modified material is shown in Fig. 3.

SEM micrographs (Fig. 3) confirm that the fiber modification caused structural changes on the nonwoven surface. The surface of the unmodified nonwoven (sample 0) is very smooth, with a slight quantity of impurities. In the images of nonwovens with deposited PDMAEMA (samples 1 and 2), a clear change in the structures can be observed, due to the deposition of the new external layer.

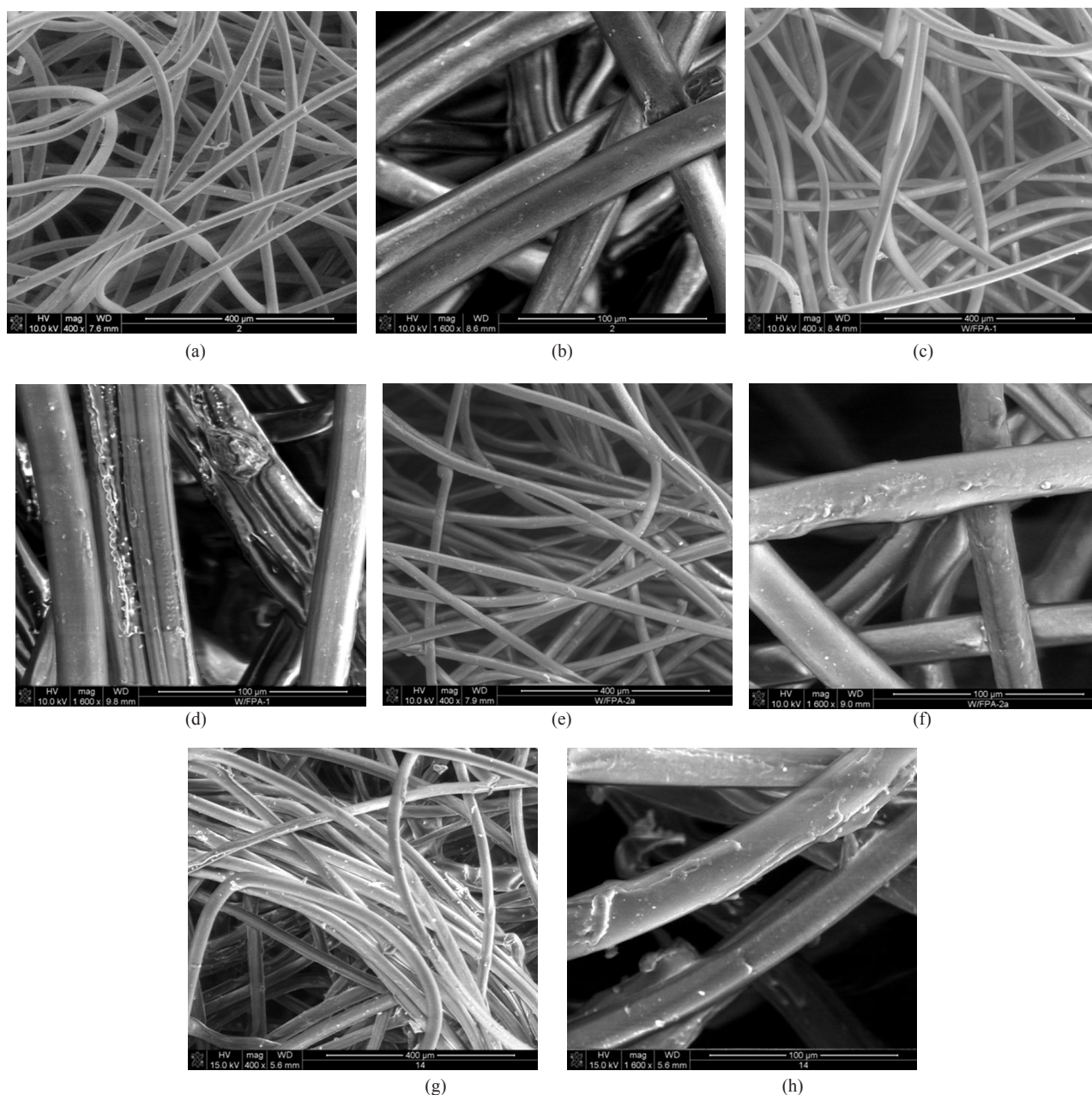


Fig. 3. SEM images of the unmodified PLA nonwoven (0) and following modifications with PDMAEMA (1-3) where: (a) sample 0, mag. x400, (b) sample 0, mag. x1,600, (c) sample 1, mag. x400, (d) sample 1, mag. x1,600, (e) sample 2, mag. x400, (f) sample 2, mag. x1,600, (g) sample 3, mag. x400, and (h) sample 3, mag. x1,600.

TABLE II
COMPARISON OF SAMPLE WEIGHTS BEFORE AND AFTER DISSOLVING TEST IN WATER

| Sample | Initial weight (g) | Final weight (g) | Weight change* (%) |
|--------------------------------------|--------------------|------------------|--------------------|
| 0 (PLA) | 1.015 | 0.966 | -4.83 |
| 1 (PLA-PDMAEMA 1.85%) | 1.005 | 0.952 | -5.27 |
| 2 (PLA-PDMAEMA 3.71%) | 1.094 | 0.999 | -8.68 |
| 3 (PLA-PDMAEMA 3.71% + crosslinking) | 0.996 | 0.990 | -0.60 |

*Estimated measurements error varies in the range 0.3±0.4%

The surface quality is changed, and the irregularities are visible. The pictures of samples after quaternization (sample 3) show some changes to the nonwoven surface. The surface structure becomes more nonuniform, showing local concentrations of the quaternized form. Furthermore, the quaternization of the modifier on the applied material results in the development of some conglomerates and microstructures that connect the fibers of the nonwoven.

The final, modified structure was also confirmed by controlling the solubility of the external layer. PDMAEMA, in its tertiary form, is soluble in a large number of solvents, including water. In contrast, the structures after crosslinking are completely insoluble. The results of solubility tests performed in water are shown in Table II.

From the data presented in Table II, it can be observed that even bathing the unmodified sample caused an almost 5% loss of sample weight. Considering the “hairy” structure of the nonwoven and its needle punch production method, this phenomenon can be expected; some weakly connected

fibers were removed. Sample 0 was analyzed in this test to determine out what percentage of weight loss was due to the wetting procedure alone. Sample 1 shows a larger level of weight loss because the water bath removed not only “free” fibers but also the introduced PDMAEMA layer. The bathing effect for sample 2 is reasonably larger because of the larger amount of modifier that could be removed during procedure. According to our expectations, sample 3 shows a much smaller amount of weight loss in this test. This result is due to the insolubility of the crosslinked external layer. Surprisingly, the bathing effect is even smaller than that of sample 0. This result suggests that the introduction of the external, crosslinked layer can stabilize the whole nonwoven architecture.

B. Antibacterial Properties of the Modified Nonwovens

The procedures outlined in the ASTM E2149–10 test method, the standard test method for determining the antimicrobial activity of immobilized antimicrobial

TABLE III
ANTIBACTERIAL RESULTS UNDER DYNAMIC CONDITIONS

| <i>Escherichia coli</i> | | | | |
|------------------------------|-------------------------------------|--------------------------------|------------------------------|-----------------------|
| Number | Composition | Initial CFU (cm ³) | Final CFU (cm ³) | Percent reduction (%) |
| 0 | PLA | 540,000 | 400,000 | 25.9 |
| 1 | PLA-PDMAEMA 1.85% | 1100,000 | 38,000 | 96.5 |
| 2 | PLA-PDMAEMA 3.71% | 520,000 | 12,000 | 97.7 |
| 3 | PLA-PDMAEMA 3.71% + crosslinking | 560,000 | 10,000 | 98.2 |
| <i>Staphylococcus aureus</i> | | | | |
| Number | Composition | Initial CFU (cm ³) | Final CFU (cm ³) | Percent reduction (%) |
| 0 | PLA | 304,000 | 254,000 | 16.4 |
| 1 | PLA-PDMAEMA 1.85% | 296,000 | 0,000 | 100.0 |
| 2 | PLA-PDMAEMA 3.71% | 300,000 | 0,000 | 100.0 |
| 3 | PLA-PDMAEMA 3.71% + crosslinking | 306,000 | 0,000 | 100.0 |

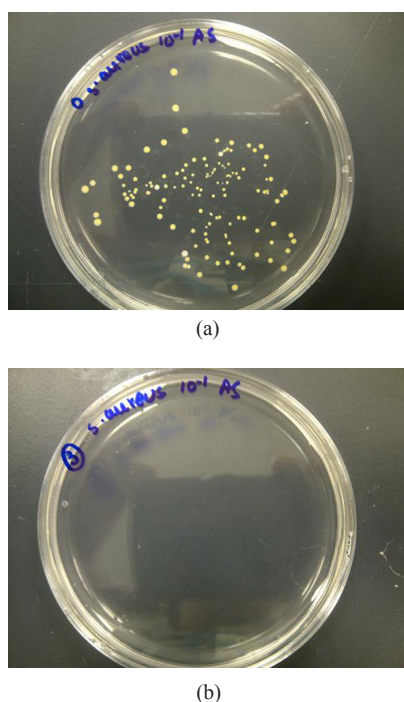


Fig. 4. Illustrative photographs of: (a) unmodified sample (0) compared with (b) sample with quaternized PDMAEMA.

agents under dynamic contact conditions, were employed for this assay. The test organisms used were *E. coli* and *S. aureus* (ATCC No. 6538). The results are shown in Table III.

The antimicrobial results obtained for samples with deposited tertiary PDMAEMA (samples 1 and 2) suggest that they are great antimicrobial agents against both types of bacteria, which is in accordance with our earlier investigations [10]. The samples with an external, quaternized and crosslinked PDMAEMA layer (sample 3) show excellent antimicrobial effects against *S. aureus* (100% reduction) and *E. coli* (more than 98% reduction). Unfortunately, even the quaternized samples did not achieve a level of 100% effectiveness against *E. coli* bacteria. Additionally, illustrative pictures demonstrating the antimicrobial activities of sample 0 (unmodified sample) compared to sample 3 (with the quaternized polymer) in the test performed against *S. aureus* are shown in Fig. 4.

Without complex cytotoxicity tests we cannot predict in vivo behavior of our polymer. However, considering the fact that, similar polymers are successfully applied in medical products and positively passing cytotoxicity tests, we suppose that they could be refer also to our product.

Antibacterial agents permanently attached to the surface of the fiber have barrier properties, protect the modified product against bacterial attack, while their effectiveness in regulating the level of microorganisms in the environment is small. Antibacterial agents can be bound to the fiber by

means of chemical (main) bonds, when there are suitable reactive groups in the modifier and fiber, and the process of their application takes place in appropriate conditions. In addition, the reaction mechanism between the two substances should be known (conditions, kinetics, catalysts, etc.). Antibacterial agents permanently bound to the surface to be modified are much more resistant to washing than agents that are released from the modified material. However, in this case, the antibacterial effect of the product is much shorter than the length of its overall life cycle. This is due to min. adsorption of dirt, dead microorganisms, or the formation of a complex between an antibacterial agent and detergents used in the washing process. By creating a durable PDMAEMA coating structure, it provides a durable modifier without the need to create chemical bonds between the fiber and the quaternary polymer. The above mechanism allows the use of this modifier to a wide range of fibers, including those that do not have reactive functional groups. The possibility of using PDMAEMA for different types of fibers and obtaining an effect in relation to a wide range of microorganisms are the main advantages of that polymer in comparison to other agents currently used in textile applications.

IV. CONCLUSION

The results shown in this work demonstrate that PDMAEMA deposited onto a polylactide nonwoven surface can be effectively crosslinked to obtain a quaternized form. In this way it is possible to obtain an external, insoluble layer on the surface of the fibers. The modified textiles with quaternary PDMAEMA layer show very good antimicrobial activity against *S. aureus* and *E. coli* bacteria. The antibacterial results can even achieve 100% efficiency. It is important to stress that the crosslinked derivative creates a strong, insoluble layer, in contrast with the tertiary polymer. This opens up a wide range of possible applications of polylactide textiles.

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