



Antibacterial cotton fabrics based on hydrophilic amino-containing scaffolds

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ABSTRACT

A hydrophilic amino compound, 4,7,10-trioxatridecane-1,13-diamine, has been utilized in several chemical and biochemical studies. Among previous applications is its use as a flexible and economical spacer molecule to increase the length between two moieties of interest, one of which may be a solid-phase interface. In this study, we immobilized this molecule on cotton fabrics and showed that this modified surface (DA) exhibited significant antibacterial activities in both Gram-negative bacteria and a Gram-positive bacterium. Studies on the structure-activity relationship revealed that additional chemical modifications on DA usually led to lowered antibacterial activities, emphasizing an importance of having free amino groups. Further investigation by fluorescence microscope indicated that this modified surface likely interfered with the membrane integrity of bacteria, leading to cell lysis. In addition, this scaffold was also tested for its biocompatibility with mouse fibroblast cells, and exerted no detrimental effect to the cell growth, highlighting its potential as a practical antibacterial surface modifier.

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1. Introduction

Antibiotic resistance of bacteria has recently become a global health concern due to a number of reasons. While the failure of discovering new bioactive scaffolds to overcome resistance is a main contributor, the misuse of existing drugs is perhaps a larger, but preventable cause [1]. Therefore, preventative measures gained increasing attention as a more economical and practical way to help solve this antibiotic crisis. For example, the development of antimicrobial materials for settings with high risk of bacterial contamination, e.g., in hospitals, can greatly prevent the infection in the first place, thus avoiding the much higher cost of post-infection treatment and diagnosis [2,3]. As a result, there have been a number of antimicrobial surfaces reported to date, all of which can be broadly categorized into three main types, namely anti-adhesion, biocide-releasing, and contact-killing surfaces [4,5].

More attention has been put towards the latter two types, where the surfaces are designed to actively inhibit the growth of bacteria. While biocide-releasing materials offer some advantages such as the readily adaptable nature of the releasing biocide (no chemical functionalization needed), they can suffer from some drawbacks including the depletion of the active compound, and the leaching to the environment, which can be a serious matter in certain cases. Silver nanoparticles, for example, are a favorite biocide used in many biomaterials with success although there have been evidences that silver nanoparticles can pose some undesirable toxicity [6], or even lead to bacterial resistance [7].

As a consequence, contact-killing surfaces became more popular due to its non-leaching nature that consequently prevents the contamination of bioactive molecules into the environment. In most cases, contact-killing surfaces have permanently positive charges from quaternary ammonium groups [5,8–11], which are believed to attract to negative charges from the outer cell membranes of both Gram-positive (teichoic acid) and Gram-negative (lipopolysaccharides) bacteria. The destruction of the membrane integrity by the other parts of bioactive scaffolds was then followed, leading to the leakage of internal cell components and the eventual death [4,12–14]. This general concept has been successfully used to

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develop several antibacterial moieties. For example, Gultekinoglu et al. reported a design of antimicrobial polyurethane stents by coating the stents with brush-like polyethyleneimine (PEI), a scaffold known to be capable of forming positively charged quaternary ammonium groups [15]. Another example includes the work from Chaker and Boufi, which demonstrated that nanofibrillar cellulose (NFC) covalently modified with cationic glycidyltrimethylammonium chloride had high antibacterial activities [16]. Notably, while the structure-activity relationship is crucial for the development of potent antibacterial surfaces, the practicality of the fabrication should also be considered as an essential factor for more realistic applications. Scaffolds that are commercially available or require minimal synthesis are more likely to gain acceptance for a larger scale of fabrication.

In this study, we reported a new class of antibacterial scaffolds based on a hydrophilic amino-containing compound called 4,7,10-trioxa-1,13-tridecanediamine (“the spacer”), which was immobilized on cotton surface as a model. In contrast to positively charged quaternary ammonium compounds with hydrophobic moieties [10,15,17], this molecule is hydrophilic due to the presence of multiple ether functional groups. It is also commercially available, and not a polymer-based product, making the production of associated surfaces less complicated and more economical. After immobilizing on cotton surface, this amino-terminated surface was found to exert strong inhibitory activities in Gram-negative bacteria and mild inhibitory activity in a Gram-positive bacterium, with no toxicity to mammalian fibroblast cells. The fabrication, characterization, antibacterial studies, and biocompatibility studies were highlighted below.

2. Materials and methods

2.1. General information

Chemicals were purchased from Sigma-Aldrich, Acros, or Chemimpex Ltd. Solvents (Analytical grade) were purchased from RCI Labscan (Thailand) and used without further purification. Cotton fabric was purchased from Thai Textile Development and Finishing Co., Ltd. Bacterial media (Nutrient Broth and Agar powder) were purchased from Himedia (India). Washing, unless stated otherwise, was done with shaking at 200 rpm on an orbital shaker. UV absorbance data were collected from Cary 100 Bio UV-vis spectrophotometer. X-ray photoelectron spectroscopy (XPS) experiments were conducted on PHI 5000 VersaProbe III. Scanning Electron Microscopy (SEM) data were obtained from JEOL JSM-6480LV scanning electron microscope. Tensile properties were performed in a Hounsfield Universal Testing Machine (H10KM).

2.2. Immobilization of 4,7,10-trioxatridecane-1,13-diamine on cotton fabrics

Two-step modification was performed to create the spacer-modified cotton surface. First, unmodified cotton ($6 \times 6 \text{ cm}^2$) was washed with solvents (*N,N*-dimethylformamide (DMF), deionized water, methanol, and dichloromethane; 5 min each) to remove adsorbed impurities. The cotton sheet was then immersed in a solution of 2 M tosyl chloride in pyridine for 90 min. After washing the tosylated cotton sheet with DMF (5 min, $2 \times$), methanol (5 min, $2 \times$), dichloromethane (5 min, $2 \times$), and air drying, the sheet was placed in a container with 15 mL solution of 1 M 4,7,10-trioxatridecane-1,13-diamine in DMF. This container was then shaken for 18 h. Washing with 5 M NaOH in methanol (5 min), methanol (5 min, $2 \times$), DMF (5 min), deionized water (5 min, $4 \times$), and drying in an oven at 40°C for 18 h resulted in amino-terminated modified cotton fabrics (DA).

2.3. Further derivatization of DA surface

DA surface could be further functionalized by a variety of reactions as described below.

Fabrication of DA.CO.C3. DA surface was immersed into a solution of propionic anhydride (0.5 mL in 10-mL DMF). This solution was kept shaking at 160 rpm at room temperature for 15 min. After a series of washing (DMF (5 min, $2 \times$) and methanol (5 min, $2 \times$)), and air drying, DA.CO.C3 was obtained.

Fabrication of DA.CO.C10. 0.1 mmol of decanoic acid (19 μL) and 17 mg of 1-hydroxybenzotriazole (HOBt, 0.11 mmol) were added into 100- μL DMF. 17 μL of *N,N*-diisopropylcarbodiimide (DIC) was then added to this solution, and the final mixture was used immediately by spotting (2 μL) directly onto the surface of DA, and the cotton sheets were then incubated at room temperature for 10 min (although the spotting method was chosen to reduce the amount of chemicals needed, the solution usually spread through the entire cotton surface). Thereafter, the spotting process was repeated twice, totalling three spotting sessions. The sheets were subsequently washed in DMF (5 min, $2 \times$), methanol (5 min, $2 \times$), and dried under ambient air. DA.CO.C10 was thus obtained.

Fabrication of DA.C10. DA surface was immersed into 1 mL of bromodecane, and 0.80-mL pyridine in 10-mL DMF. The solutions were shaken at 160 rpm for 20 h. Washing with DMF, methanol, 0.1 M aq. sodium chloride solution, deionized water, methanol, hexane (5 min each) and air drying provided DA.C10.

Fabrication of DA.Me. DA surface was immersed into a solution of 310- μL iodomethane in 10-mL DMF (0.5 M). The solutions were shaken at 160 rpm for 20 h. Washing with DMF, methanol, 0.1 M aq. sodium chloride solution, deionized water, methanol (5 min each) and air drying provided DA.Me.

2.4. Characterizations of modified cotton surfaces

2.4.1. Loading determination of amino groups

Loading of amino groups was determined by treating cotton sheets with CI Orange II solution (immersing the sheets in 1.4 M solution of the dye in 50-mL water with 475- μL acetic acid) at 50°C for 30 min. The sheets ($2.5 \times 2.5 \text{ cm}^2$) were washed several times with deionized water. When ready, the attached dye was removed into the solution phase by immersing the sheets in 25% aq. pyridine for 15 min. Absorption measurement at 485 nm was used to determine the loading in comparison with the standard molar absorptivity ($\epsilon = 16.328 \text{ mM}^{-1} \text{ cm}^{-1}$).

For quaternary amine loading, another experiment utilizing fluorescein sodium salt was conducted. Cotton sheets ($2.0 \times 2.0 \text{ cm}^2$) were immersed in 26.5 mM solution of the dye (1 mL aqueous solution) for 5 min. Washing with deionized water (3 min, $5 \times$) was subsequently performed. When ready, the attached dye was removed into the solution phase by immersing the sheets in 0.25% aqueous solution of cetyltrimethylammonium bromide (CTAB) for 15 min. Absorption at 501 nm was used to determine the loading in comparison with the standard value ($\epsilon = 42.797 \text{ mM}^{-1} \text{ cm}^{-1}$).

2.4.2. XPS experiments

The surface elemental compositions of modified sheets were measured using PHI 5000 VersaProbe III with a monochromatic Al K α X-ray source ($h\nu = 1486.6 \text{ eV}$). Most spectra were conducted at a 45° take-off angle. Wide spectra were scanned at binding energies ranging from 0 to 1000 eV with a pass energy of 280 eV and the pass energy of individual element spectra were 26 eV (C 1s), 55 eV (O 1s), 140 eV (N 1s), 224 eV (Si 2p).

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