

# Antimicrobial Aluminum Surfaces for Curbing Healthcare-Associated Infections—A Short Review <sup>†</sup>

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**Abstract:** Healthcare-associated infections are serious public health problem. Besides antibacterial, antifungal, and antiviral therapies, one potential strategy for breaking the chain of infection transmission is via the installation of antibacterial surfaces. Aluminum is an attractive material for fabricating frequently touched surfaces such as doorknobs, push plate, bedrails, etc. Recently, our research group and others have demonstrated that by utilizing appropriate surface treatment technologies, such as anodization, low-surface-energy passivation, and electrochemical surface modification, on an AA6061-T6 aluminum alloy, aluminum could be rendered antimicrobial. Such surface technologies can be efficient in antimicrobial activities, and they also show advantages in terms of robustness and durability. These novel surfaces have been shown to reduce the microbial burden of clinically relevant pathogens such as *Staphylococcus aureus*, *Pseudomonas aeruginosa*, and *Escherichia coli*.

**Keywords:** healthcare-associated infection; aluminum; surface modification; antimicrobial surfaces

## 1. Introduction

Healthcare-associated infections (HAIs) are serious public health problem [1]. They are the fourth leading cause of death after cancer, heart disease, and stroke [2]. In Canada, more than 200,000 patients contract HAIs yearly, out of which 8000–12,000 of the cases result in fatality [2]. Besides antibacterial, antifungal, and antiviral therapies, one potential strategy for breaking the chain of HAI transmission is to make frequently touched surfaces antimicrobial to curb the attachment of pathogens. However, the problems associated with existing antimicrobial solutions, such as a lack of long-term durable antimicrobial coating, the uncontrolled release of antimicrobial agents, and antimicrobial resistance problems [3], have necessitated the need for novel, stable, and durable antimicrobial surfaces in addition to an appropriate cleaning regime for the decontamination of frequently touched surfaces in hygiene-critical environments [4].

Owing to being lightweight and widely recyclable, aluminum (Al) is increasingly becoming the material of choice for engineering constructions. Many Al components are progressively adopted in the fabrication of medical devices [5] and are explored in fabricating frequently touched surfaces such as doorknobs, push plates, bedrails, over-bed tables, and countertops [6]. Aluminum has excellent physical, chemical, and mechanical properties such as a low density, good wear and corrosion resistance, and high elastic modulus and tensile strength [7]. Aluminum is the third most abundant element in the Earth's crust (after oxygen and silicon) and the most abundant metal on earth. It is a highly reactive metal and exists with a layer of passive aluminum oxide film on its surface. To render aluminum surfaces antimicrobial, this passive oxide layer needs to be modified with the appropriate technology. Our research group and others have demonstrated



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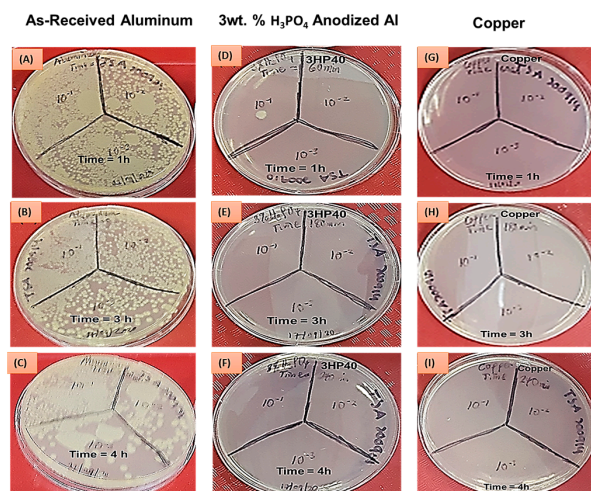
that by utilizing appropriate surface treatment technologies, aluminum can be rendered antibacterial.

This brief review presents the current progress which has been made in antimicrobial aluminum surface technologies achieved via (i) an anodization process to create a nanoporous topographical pattern, which kills bacteria on contact; (ii) passivating the nanoporous topographical pattern to create superhydrophobic properties to repel the initial attachment of bacteria; and (iii) immobilizing antimicrobial agents on anodized aluminum to kill bacteria on contact.

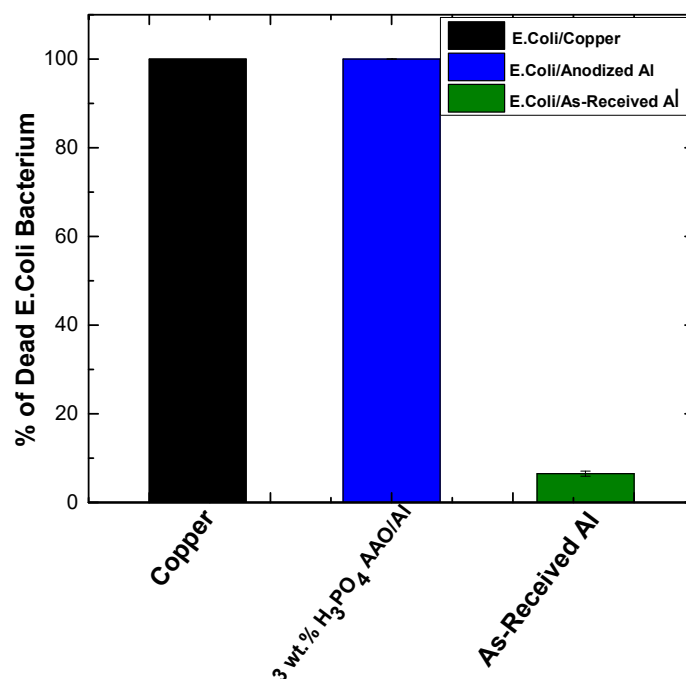
## 2. Mechano-Bactericidal Aluminum Surfaces

In nature, many living things, such as insects, plants, sharks, and geckos, have developed protective mechanisms towards pathogens, despite their ability to use complex mechanisms to colonize surfaces [8]. The former, on the other hand, exploit their micro- and nano-topographical features to induce antimicrobial activity. These surfaces that kill bacterial cells via topography are called mechano-bactericidal surfaces [8]. The first mechano-bactericidal surfaces mediated by the nano topography of cicada wings was reported by Ivanova et al. [9]. In this work, the authors showed that by the pure physical contact of *P. aeruginosa* (gram-ve) bacterium on the topographical rough surface of cicada's wings, *P. aeruginosa* died in 30 minutes. As a result, various surfaces on other wings, such as dragonfly and the skin of geckos, and their synthetic analogues, have been fabricated on many substrates, including aluminum alloys [10,11].

Hasan et al. [11] recently fabricated an antiviral aluminum surface using a chemical etching process. The aluminum surface was effective against SARS-CoV-2. In their other study [6], they reported the antimicrobial ability of a topography-mediated aluminum, fabricated using a chemical etching technique against common multi-drug-resistant pathogens such as *Pseudomonas aeruginosa* (P.A) and *Staphylococcus aureus* (S.A), and on respiratory viruses such as respiratory syncytial virus (RSV) and rhino virus (RV). While desirable nanoscale topography is achievable, the mechanical property of the nanoscale coating is rather weak [12]. In our research group, Agbe et al. [13] utilized anodization as an alternative surface treatment process to achieve antimicrobial aluminum surfaces with superior antimicrobial properties and mechanical robustness. In that work, a one-step hard aluminum anodization process was employed and anodization parameters, such as electrolytes concentration, current density, and anodization time, were optimized to obtain desirable surface morphology (with diameters  $151 \pm 37$  nm), which was effective in inactivating 100% *Escherichia-coli* (*E. coli*) bacteria (Figures 1 and 2).



**Figure 1.** Representative images of colonies of *E. coli* in contact with as-received aluminum from 1 to 4 h (A–C); 3 wt.%  $H_3PO_4$  anodized aluminum coupon-3HP40 from 1 to 4 h (D–F); and the reference copper samples from 1 to 4 h (G–I). Reprinted with permission from ACS Biomaterials Science & Engineering 8(3) (2022) 1087–1095. Copyright (2022) American Chemical Society.



**Figure 2.** Bactericidal efficiency of *E. coli* bacteria on 3 wt.% H<sub>3</sub>PO<sub>4</sub> anodized aluminum 3HP40 compared to control samples (as-received aluminum and copper) under 1 h of contact. Data represent three independent experiments. Reprinted with permission from ACS Biomaterials Science & Engineering 8(3) (2022) 1087–1095. Copyright (2022) American Chemical Society.

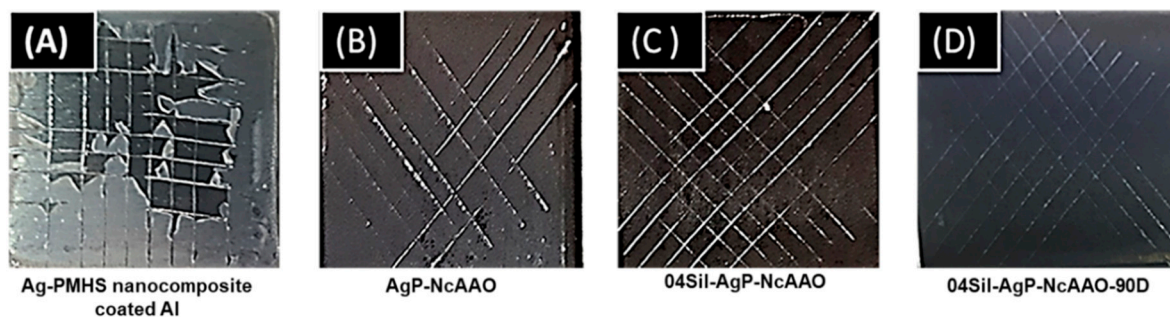
### 3. Superhydrophobic Antimicrobial Aluminum Surfaces

Scientists have discovered a unique water roll-off property in several plants, such as lotus leaves. These plants utilize their surface micro-nanoscale structures to adapt and inhibit colonization as an evolutionary surviving strategy [14]. The micro-nanostructures are made of long chain palmitic (hexadecanoic) and stearic (octadecanoic) fatty acids [15], which render them superhydrophobic. Inspired by nature, scientists have now fabricated superhydrophobic surfaces in the lab by patterning micro-nano structures, followed by passivation with low-surface-energy molecules. Various strategies such as photolithography, sol-gel, plasma etching, anodization, and chemical etching [16] have been used to fabricate superhydrophobic surfaces, which find wide applications as self-cleaning, anticorrosion, drag-reduction, and anti-biofouling surfaces of different components [17]. Of particular interest is in an anti-biofouling application, as superhydrophobic surfaces have the potential to prevent initial bacterial attachment and subsequent biofilm formation.

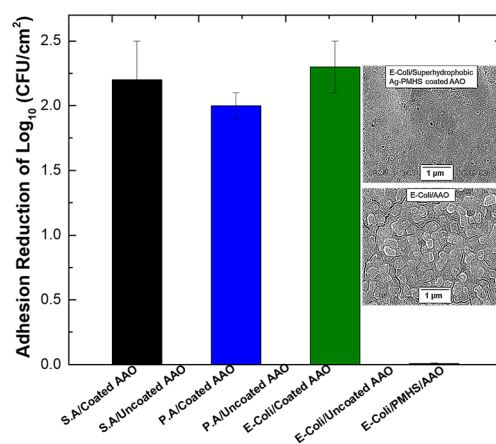
However, superhydrophobic surfaces fail under long-term exposure in humid environments [18]. Thus, incorporating bactericides in superhydrophobic surfaces provides the promise of not only integrating anti-biofouling and antibacterial properties, but also improving their longevity. Chung et al. [19] fabricated silver-perfluorodecanethiolate coatings on a silicon wafer with both superhydrophobic and antibacterial properties via the precipitation method, using perfluorodecanethiol and silver as fluorinated and metal-thiolate complex precursors, respectively. In another work, Wang et al. [20] fabricated superhydrophobic diamond films with both antibacterial and anti-biofouling properties using both hot filament chemical vapor deposition and a sol-gel perfluorodecyltrichlorosilane process. In spite of significant effort being made for the use of superhydrophobic coatings in the application of antibacterial surfaces, the bacterial-repellent performance was rather low [21,22]. Furthermore, the applicability of such coating is limited due to degradation with time and a lack of stability.

Novel superhydrophobic coatings, which anchor low-surface-energy molecules on aluminum substrates for enhanced durability and stability, appear increasingly attractive. In a recent work by Agbe et al. [23], a novel coating of silver-polymethylhydrosiloxane

(Ag-PMHS) nanocomposites (anchored on anodized Al) was fabricated via a sol-gel process for antibacterial and anti-biofouling applications. The Ag-PMHS nanocomposite coatings (anchored within an anodized Al oxide (anodized aluminum oxide)), led to enhanced adhesion, durability, and stability (Figure 3). The superhydrophobic Ag-PMHS nanocomposite coatings (achieved by combined effects of nano-rough porous anodized aluminum-Ag-PMHS nanocomposites, and low-surface-energy passivation), resulted in bacterial adhesion reductions of 99.0 %, 99.5 %, and 99.3 % for *Pseudomonas aeruginosa* (P.A.), *E.coli*, and *Staphylococcus aureus* (S.A.), respectively (Figure 4).



**Figure 3.** Digital images of scratch test based on American Standard Test Method (ASTM D 3359-02) showing the adhesion of Ag-PMHS nanocomposite coatings with a Ag<sup>+</sup>/Si-H molar ratio of 50:2 on: (A) as-received Al; (B) AAO/Al (AgP-NcAAO); (C) 0.4% *w/v* silicone incorporated in AgP-NcAAO (04Sil-AgP-NcAAO); and (D) 04Sil-AgP-NcAAO in 90 days of immersion (04Sil-AgP-NcAAO-90D). Reprinted with permission from ACS Applied Bio Materials 3 (2020) 4062–4073. Copyright (2020) American Chemical Society.



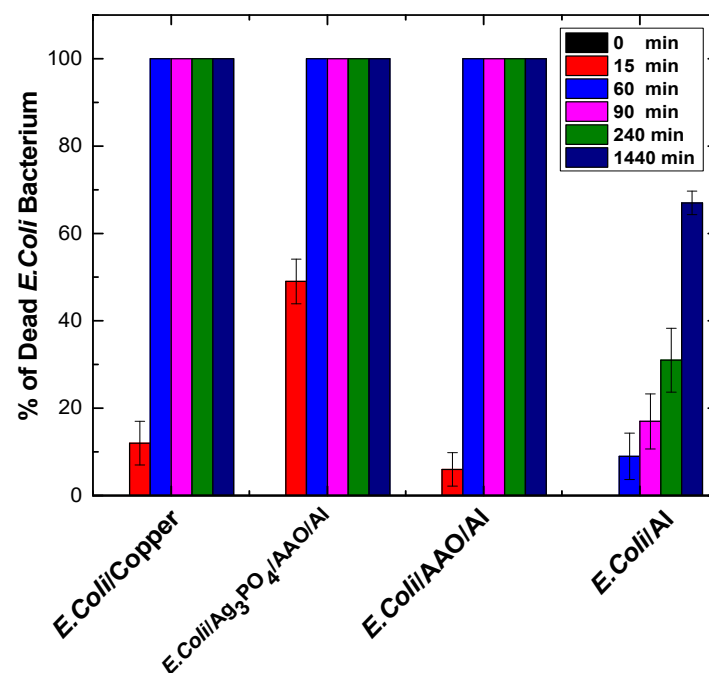
**Figure 4.** Adhesion reduction in bacteria (S.A, P.A, or *E-coli*) on superhydrophobic AAO/Al sample (04Sil-AgP-NcAAO) and control samples (AAO/Al and PMHS/AAO/Al). Data represent multiple independent experiments. Inset: SEM micrograph of *E-coli* biofilm on superhydrophobic AAO/Al sample (Top) and control sample (AAO/Al) (Bottom). Reprinted with permission from ACS Applied Bio Materials 3 (2020) 4062–4073. Copyright (2020) American Chemical Society.

#### 4. Hydrophilic Antimicrobial Aluminum Surfaces

Hydrophilic antimicrobial coatings are surfaces with a contact angle between 10° and 90°. Like superhydrophobic antimicrobial surfaces, the fabrication of hydrophilic antibacterial surfaces involves physical, chemical, and biological processes. These surfaces are particularly useful for orthopedic and medical implant devices as they enhance protein conditioning and cell tissue integration [24]. Various hydrophilic antimicrobial agents such as TiO<sub>2</sub>, ZnO, Ag, and Cu nanoparticles are commonly fabricated on metals and other substrates. However, silver-based hydrophilic antibacterial coatings fabricated via an electrochemical reduction process are further discussed.

As a widely used engineering material (for the fabrication of frequently touched surfaces), aluminum has become one of the generally used substrates for the electrodeposition of metallic coatings such as silver. For the electrodeposition of silver on anodized aluminum oxide (AAO/Al), an AC power source is recommended [25,26]. In a typical AC electrodeposition process, the barrier oxide layer acts as a rectifying p-n junction source. Therefore, AAO/Al templates can be used directly as the cathode during an electrodeposition process. Thus, the barrier layer becomes preferentially conductive during the cathodic half-cycle. This rectifying property allows for a reduction in metal ions in the pore while decreasing the oxidation rate of the deposited metal [26]. Chi et al. [25] deployed AC electrodeposition to deposit Ag for an antimicrobial application. Their results showed a 95% antimicrobial efficiency for *E. coli*, *P. aeruginosa*, *Streptococcus faecalis*, and *S. aureus*. On the other hand, for DC silver electrodeposition, it is necessary to separate the AAO/Al templates from the base aluminum metal by chemical etching, followed by the metallization of the former [27]; while DC silver electrodeposition on aluminum is feasible, in practice, it can be challenging due to the increased complexity of the overall electrodeposition process [28].

However, a novel approach was recently reported by Agbe et al. [29]. In their work, a two-step electrochemical process was deployed to fabricate  $\text{Ag}_3\text{PO}_4$  nanoparticles on anodized aluminum oxide for antimicrobial applications.  $\text{Ag}^+$  ion was first reduced to  $\text{Ag}^0$  metal at a reduction process, while the metallic  $\text{Ag}^0$  was oxidized again into  $\text{Ag}^+$  ion. The antimicrobial properties of the  $\text{Ag}_3\text{PO}_4$ -coated anodized aluminum ( $\text{Ag}_3\text{PO}_4/\text{AAO}/\text{Al}$ ) resulted in 100% *E. coli* bacteria inactivation (Figure 5).



**Figure 5.** Antimicrobial activity of *E. coli* bacteria on  $\text{Ag}_3\text{PO}_4$ -coated anodized AL ( $\text{Ag}_3\text{PO}_4/\text{AAO}/\text{Al}$ ) relative to the control samples (copper, AAO/Al, and as-received aluminum) under different contact times. Reprinted with permission from Surface and Coatings Technology 428 (2021) 127892. Copyright (2021) Elsevier B.V.

## 5. Conclusions

Multidrug-resistance pathogens such as *Staphylococcus aureus*, *Pseudomonas aeruginosa*, and *Escherichia coli* can survive on frequently touched surfaces, such as doorknobs, push plates, over-bed tables, and bed rails, in hygiene-critical environment. Such surfaces can become potential reservoirs for the transmission of healthcare-associated infections. Antimicrobial coatings have been recognized as an important strategy for reducing the microbial burden and breaking the chain of infections. Recent advances in antimicrobial aluminum

surface technologies have demonstrated that the utilization of novel surface engineering strategies, such as anodization, low-surface-energy passivation, and electrochemical surface modification of an AA6061-T6 aluminum alloy, has proved to be efficient in antimicrobial activities, and there are also advantages in terms of robustness and durability. These novel surfaces have shown the ability to reduce the microbial burden of clinically relevant and healthcare-associated pathogens such as *Staphylococcus aureus*, *Pseudomonas aeruginosa*, and *Escherichia coli*.

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