INVESTIGATION INTO THE BACTERIAL REPELLING BEHAVIOR OF SUPERHYDROPHOBIC MODIFIED METAL SURFACES FOR FOOD SAFETY AND HYGIENE

A Thesis

by

SHUHAO LIU

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Chair of Committee,	Mustafa Akbulut
Committee Members,	Svetlana Sukhishvili
	Zhengdong Cheng
Head of Department,	Ibrahim Karaman

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ABSTRACT

As a result of frequent outbreaks occurring due to poor hygiene and improper sanitation of processing environments, there has been an increasing demand for the development of food-contact surface materials that intrinsically inhibit and reduce the likelihood of potential microbial adherence and biofilm formation. Herein, we report the synergistic utilization of surface nanotexturing and chemical modifications with nonpolar functional groups on aluminum surfaces to produce coatings having bacterial super-repellant and mud anti-fouling characteristics. Using these coatings, the attachment of *Salmonella* Typhimurium LT2 and *Listeria* innocua as pathogen surrogates was reduced more than 99.0%, compared to the bare aluminum surfaces. In addition, the coating strongly resisted the adhesion of mud, showing a 10-fold reduction in the area of mud adhesion upon submerging in mud solution. Moreover, this method is both versatile and scalable, involving inert, non-leaching, and biocompatible building blocks. Overall, this study contributes to the field of food safety through the design and development of novel coatings for achieving improved food safety and hygiene.

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Contributors

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NOMENCLATURE

CDC	Centers for Disease Control and Prevention
THFS	Trichloro(1H,1H,2H,2H-heptadecafluorodecyl)silane
DI	Deionized
SEM	Scanning electron microscopy
ATR-FTIR	Attenuated total reflectance-Fourier transform infrared
TSA	Trypticase Soy Agar
TSB	Tryptic Soy Broth
FDA	Food and Drug Administration

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1. INTRODUCTION*

1.1 Outbreak of foodborne illness

All countries throughout the world are suffered a significant threat to public health security due to a large number of illnesses and deaths caused by unsafe food. A big range of the burden and cost of the foodborne illnesses due to the pathogenic bacterial, parasitic microorganisms, and food contamination are regarded as the substantial. (Kuchenmüller et al., 2009). The outbreak of foodborne illness related to contaminated food can make a big affection on the trust of society and economic harm on the countries (Kirk, Ford, Glass, & Hall, 2014). Shannon estimated that around 93.8 million cases of gastroenteritis were caused by *Salmonella* species and 80.3 million of these were foodborne illness, which includes 39,000-303,000 deaths every year globally (Majowicz et al., 2010).

Many countries have estimated the incidence of foodborne diseases. Around 25% of gastroenteritis in Australia was due to the contaminated food, and about 4.1 million foodborne gastroenteritis occurred in 2010 (Kirk et al., 2014). Also, the outbreak of foodborne illnesses every year represents a significant health problem in the United States. According to the table from the Centers for Disease Control and Prevention (CDC) survey, an estimated 9.4 million illnesses were caused by 31 identified pathogens annually. About 56000 of these incidences lead to hospitalization, and 1350 of them caused death. Moreover, another 38.4 million illnesses, 72000 hospitalizations and 1700 deaths were resulted by unspecified agents per year in the United States (Angelo, Nisler, Hall, Brown, & Gould, 2017; Ebel et al., 2016). Recent research reported that

foodborne illnesses lead to an annual burden to society of approximately 36 billion dollars, with an average identified illness estimated to reduce quality-adjusted life days by 0.84, which is monetized and included in the average cost of burden per illness at $3,630 (Minor et al., 2015). Efforts of food safety take place during the entire food chain from production to retail level. However, these processes could not guarantee that the potential of pathogenic bacteria are free in food products (Whitney et al., 2015).

1.1.1 Foodborne illnesses

A large fraction of foodborne illnesses is resulted by several bacteria pathogens, including *Salmonella* enterica, *Listeria* monocytogenes, *Escherichia* coli, *Staphylococcus* aureus, *Campylobacter* spp, and *Yersinia* enterocolitica. These strains of the foodborne pathogen can survive on different food contact surface for periods from several hours to several days and form biofilms (Brooks & Flint, 2008; Osimani, Garofalo, Clementi, Tavoletti, & Aquilanti, 2014). Some of these foodborne pathogens are transmitted to humans by foods which can be contaminated at any procedure in the farm to table continuum like in human's kitchen (Nyachuba, 2010).

Salmonella, all members of which are Gram-negative enteric bacteria, are considered to be human pathogens(Jay, 2000). Nontyphoidal *Salmonella* is a significant reason for diarrheal disease globally which causes 155,000 diarrheal deaths every year. HIV-infected, malaria-infected person and malnourished children in many countries are suffering increasing risk for this disease (Majowicz et al., 2010). *Salmonella* spp. causes more than 1.2 million illnesses in which around 23000 hospitalizations and 450 death per year. It is the primary cause of foodborne illnesses in the United States. *Salmonella* enteric serotype has been reported as most common serotype among

more than 2500 serotype of *Salmonella* (Yishan Yang, Kumar, Zheng, & Yuk, 2015). *Salmonella enterica* serovar Typhimurium (*S.* Typhimurium) is one of the most common serotypes which results of nontyphoidal *Salmonella* infections in human beings(Won & Lee, 2017). As well as contaminated egg, poultry, meat, fruits and nuts, the contaminated water, personal contact, equipment are other routes to cause the transmission of *Salmonella* in the life (Alexander et al., 2016).

Listeria monocytogenes (*Listeria*), Gram-positive ubiquitous foodborne bacterium, is a widespread one of the most virulent foodborne pathogens cause the infection listeriosis (Ramaswamy et al., 2007). Since *Listeria* monocytogenes was first discovered as foodborne pathogen in the 1980s, it has been identified as a significant cause of the outbreak of the foodborne diseases. Most bacteria grow poorly under 4 °C while *Listeria* can survive in a bigger range of temperature from -7 °C to human body temperature. On the other hand, *L.* monocytogenes can grow at the environment with low moisture and high salts, in comparison of many other foodborne pathogens, which allows this pathogen grow well in soil, dust, sewage, silage and animal feeds (Lamont et al., 2011). Therefore, *Listeria* can be growing at both ready-to-eat food which has been kept refrigerated and food contact surface, which makes the *Listeria* more challenging to be controlled (de Noordhout et al., 2014). Some investigation of outbreak showed that *Listeria* in 11% of all food samples retrieved from refrigerators which were contaminated and 64% of the refrigerators of infected cases contained at least one contaminated food item(Lamont et al., 2011). Although women with pregnancy, the elder people and immunocompromised people are at more risk of clinical listeriosis, healthy people can also get a febrile gastroenteritis due to the infections of *Listeria*(de Noordhout et al., 2014).

1.1.2 Contamination/cross-contamination

Contaminated or insufficiently sanitized food-contact surfaces are one of the main pathways for bacterial contamination and cross-contamination leading to the foodborne diseases (Scott & Bloomfield, 1990). For instance, past research has indicated the ability of coring knives to transfer *E.* coli O157:H7 to lettuce heads after contact with pathogen-contaminated soil. Coring knives can also be contaminated with pathogens through contact with workers' gloves (Taormina et al., 2009). Another recent study has stated that the knife or grater was contaminated by Escherichia coli O157:H7 or *Salmonella* enteric after slicing inoculated carrots, tomatoes, honeydew melons, strawberries, cucumbers, and cantaloupes (Erickson, Liao, Cannon, & Ortega, 2015). Moreover, produce minimal processing operations such as washing/sanitizing, cutting, and storing provide numerous opportunities for cross-contamination(Matthews, Sapers, & Gerba, 2014). *E.* coli O157:H7 transfer from equipment surfaces to fresh-cut leafy greens was reported to occur during processing (Buchholz, Davidson, Marks, Todd, & Ryser, 2012). *Listeria* monocytogenes contamination was studied in a poultry slaughter by Daniela F.Schäfer. Their results showed that 95% of the contamination with *L.* monocytogenes occurred before the chilling process and the contamination will linearly increase after the cutting process in the slaughterhouse (Schäfer et al., 2017). Metal bench surfaces were identified to be other important facilitators of pathogenic cross-contamination in fresh produce (Axelsson et al., 2013; Kusumaningrum, Riboldi, Hazeleger, & Beumer, 2003; Ryu & Beuchat, 2005). Lehto et al. (Lehto, Kuisma, Määttä, Kymäläinen, & Mäki, 2011) analyzed six fresh-cut vegetable processing plants and found that the levels of total aerobic microbial counts on cutting and peeling machines were more than 20 CFU/cm2 on all surfaces and 18% of surfaces in contact with product, other than machines, were moderately contaminated with

a mean of 2–10 CFU/cm2. Aside from metal surfaces, there have been numerous studies investigating the dynamics and mechanisms of interactions of bacterial pathogens with various natural and synthetic food-contact surfaces, including ceramics(J. K. Oh, Perez, et al., 2015), glass (Chia, Goulter, McMeekin, Dykes, & Fegan, 2009), paper (Jin, Jiang, Niu, & Huang, 2012), textiles (Perelshtein et al., 2008), and plastics (Faille et al., 2002).

1.1.3 Prevention for the contamination of foodborne bacteria

It is clear that various types of food-contact surfaces, including equipment, tools, containers, and accessories used at the storage, sorting, and processing stages can contribute to cross-contamination/recontamination by pathogens and recontamination of food and compromise their microbiological safety, thereby increasing the risk of foodborne illness for consumers. As such, a large body of work has focused on the development of versatile, effective sanitization approaches for improving the microbiological safety of food contact surfaces (Duncan, 2011; Llorens, Lloret, Picouet, Trbojevich, & Fernandez, 2012). These strategies tend to involve the utilization of heat; pressure or vacuum; liquid, gas, or aerosolized chemical disinfectants; cold and atmospheric plasma; ionizing or non-ionizing radiation (Neal, Cabrera-Diaz, Marquez-Gonzalez, Maxim, & Castillo, 2008; S. Oh, Gray, Dougherty, & Kang, 2005; Park et al., 2012; Wan, Coventry, Swiergon, Sanguansri, & Versteeg, 2009; M. Zhang et al., 2015; M. Zhang, Oh, Cisneros-Zevallos, & Akbulut, 2013). Some of these techniques face difficulties in sanitizing pathogens trapped between abraded sections, crevices, pits, cracks, and rough sections of food contact surfaces (Awad, Asker, & Hatton, 2018; Moerman & Kastelein, 2014). In addition, the cost associated with the frequent and repeated applications of sanitizers constitutes a large expense, which needs to be spent to ensure the sufficient hygiene of food-contact surfaces. Therefore, research on

complementary approaches, such as those relying on the prevention of bacterial contamination via smart surface design, has gained traction in the food safety community at large. Surface functionalized by antibacterial agents have been reported to prevent the contamination and cross-contamination of bacterial pathogen. (J. K. Oh et al., 2016). For instance, the antibacterial property against *E.* coli O157:H7 and *S.* Typhimurium of food package with the carvacrol nano-emulsion incorporated the chitosan coating was investigated on green beans (Severino et al., 2015). Good antibacterial property of silver zeolite contained polymer coating on steel surfaces was evaluated by Griffith (Griffith, Neethirajan, & Warriner, 2015). Although the material with antimicrobial agents achieved the goal of reducing bacterial growth and improve food safety, there still are many issues that still need to be overcome like the antibacterial resistance, dead bacteria films may provide a platform for bacterial attachment (Lu et al., 2016; J. K. Oh et al., 2016). Another type of surfaces, which reduced the contamination by developing anti-biofouling and anti-adhesive properties, attracts attention these years.

1.2 Superhydrophobic surface and its application

Superhydrophobic surfaces were represented the surfaces which have a higher than 150° of static water contact angle (WCA) and sliding angle less than 10°. Recently, researches on superhydrophobic surfaces have attracted significant attention and grown dramatically since 2004 due to the high performance of water repealing. (Darband, Aliofkhazraei, Khorsand, Sokhanvar, & Kaboli, 2018; Hu et al., 2013). Bagheri et al. (Bagheri, Aliofkhazraei, Forooshani, & Rouhaghdam, 2018) have already fabricated superhydrophobic nanocomposite coating. The water can jump more than three times and remain good shape of a water droplet on the surface in the bouncing test, and the water jet will reflect from the surface at an elevation angle of 26° in water

-jet evaluation test. The highest water contact angle of their coating achieved 161° which performed excellent self-cleaning properties. Because of the advanced water-repellent properties of superhydrophobic surfaces, this type of surfaces have significant potential for practical application, including self-cleaning (Bixler & Bhushan, 2014), anti-bioadhesion (Hu et al., 2013), anti-fouling(Xue, Guo, Ma, & Jia, 2015), anti-icing (Jung, Tiwari, Doan, & Poulikakos, 2012), drag reduction(Martell, Perot, & Rothstein, 2009), good boiling heat transfer (Takata, Hidaka, & Kohno, 2012), anti-corrosion(Z. Zhang, Ge, Men, & Li, 2016), oil and water separation, anti-bacterial surfaces, and medical applications (Darband et al., 2018). Fig.1 shows good anti-fouling and self-cleaning properties of the superhydrophobic coating on polymer surfaces. For example, Zhang et al. (Lv & Zhang, 2014) have fabricated the superhydrophobic surfaces on aluminum alloy substrates which delay the freezing time of water droplets by more than 20 minutes and reduce the freezing temperature to −11.9℃. Dong et al. have developed a self-cleaning coating on a model boat, the results showed that the coating took good effect in decreasing drag force from the water to the boat at a high speed (Dong, Cheng, Zhang, Wei, & Shi, 2013). These studies on superhydrophobic surfaces shows a good potential practical use in the life such as airplane, boat, industry processing and even food safety.

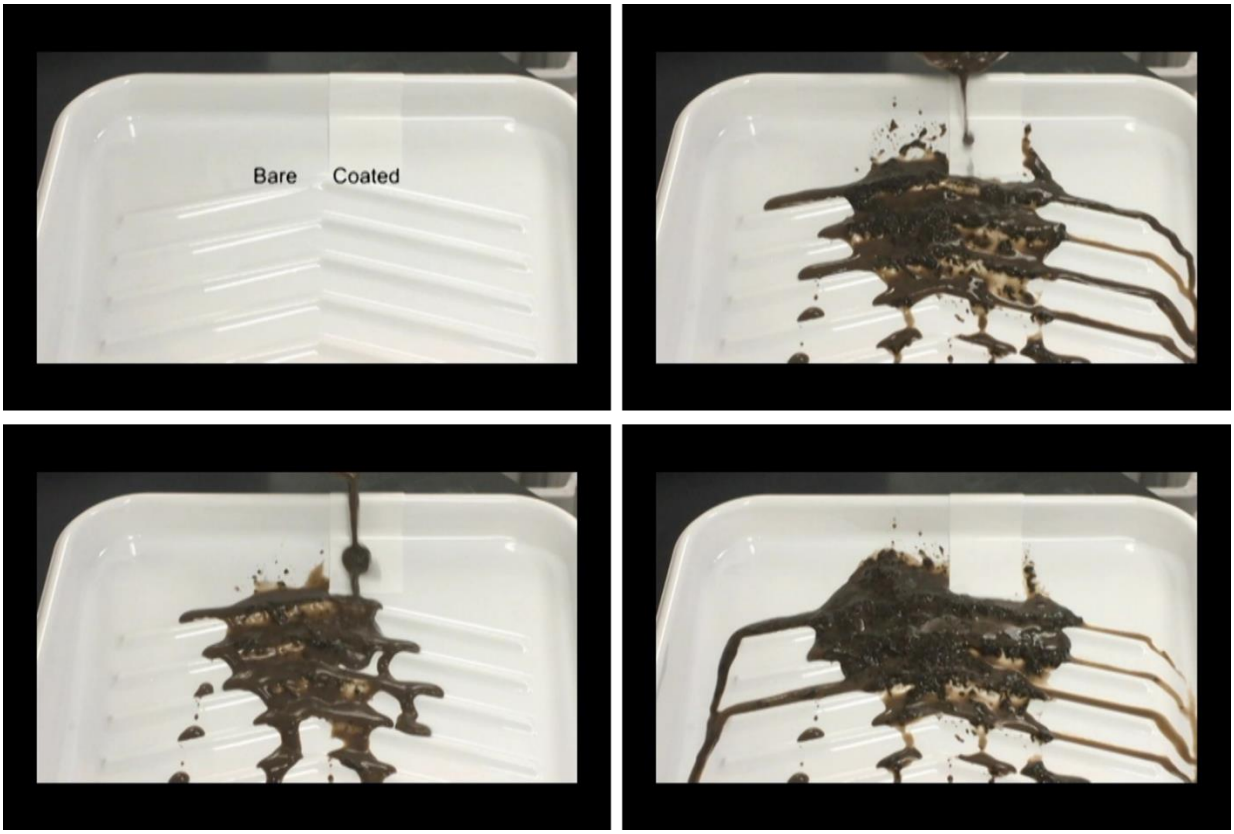

Fig. 1 Anti-fouling property of superhydrophobic surfaces: mud solution flow down from superhydrophobic plastic surfaces and bare plastic surfaces

1.2.1 Silica nanoparticles and long-chain fluorinated silane

Silica particles were chosen as nanotexturing entity because silica (silicon dioxide) is a Food and Drug Administration (FDA)-approved food additive (Tang, Li, & Chen, 2012). Yang et al. (Yi-Xin Yang et al., 2014) have evaluated the gastrointestinal toxicity of silica nanoparticles to the human body and the human gastrointestinal tract in their studies. They induced the silica nanoparticles into human gastric epithelial cell GES-1 and colorectal adenocarcinoma cell Caco-2 to evaluate the safety level. The results of cell uptake, cellular reactive oxygen species (ROS) level, cell cycle and apoptosis indicated that silica nanoparticles are safe as food additive. The

interaction between silica nanoparticles and food matrices, such as saccharides, proteins, lipids, and minerals were investigated by Go et al. (Go, Bae, Kim, Yu, & Choi, 2017). Their results illustrated that the silica interacts with only a small portion of food, and the interaction are not strong, which may be useful for predicting the toxicity of silica nanoparticles combine the food components. Further, silica-based materials have been used in several food industries (e.g., food packaging materials, food processing material) (Marsh & Bugusu, 2007; J. K. Oh, Perez, et al., 2015), biomedical (e.g., food biosensors)(Baby & Ramaprabhu, 2010; Sanchez, Belleville, Popall, & Nicole, 2011), and healthcare (e.g., food service water filtration systems) (Neofotistou & Demadis, 2004; Yu, Xu, Shen, & Yang, 2009) applications, owing to their biocompatibility, low toxicity, and relatively low cost.

The superhydrophobicity of surfaces depends on two primary factors including surface microstructure (surface roughness) and chemical composition. Regarding the chemical composition on surfaces, it determines the surface energy of the surfaces which affects the wettability significantly. Superhydrophobicity can be achieved by introducing the component which has low surface energy on a surface with specific nanotexture or microstructure (Darband et al., 2018; Islam, Akter, & Karim, 2010). For chemical modification of silica nanoparticle coating which has proper surface microstructure, silane has high affinity to silica. In addition, fluorinated polymers like poly (vinylidene fluoride) (PVDF) and polytetrafluoroethylene (PTFE) or fluorinated chemicals like fluorinated silane was commonly used to produce superhydrophobic surface due to trifluoromethyl (-CF3) group which has better nonpolar property over other groups. In comparison of trifluoromethyl group with methyl group, surface energy decrease as hydrogen(H) atoms are replaced by fluorine(F) atoms (J. K. Oh et al., 2016). For instance, Jeong

et al. (Jeong et al., 2018) reported their superhydrophobic surface fabricated by grafting heptadecafluoro-1,1,2,2,-tetra-hydrodecyl)trichlorosilane (FTS) on PVDF nanostructure. The PVDF-FTS film developed in this work showed good stability by remaining a high contact angle surface after against organic solvent. Moreover, Jun Kyun Oh et al. (J. K. Oh et al., 2016) has already prepared anti-bioadhesion gloves by dipping the gloves into the fluorinated silane grafted silica nanoparticles solution. These gloves with superhydrophobic property exhibited 99% percentage reduction of *S.* Typhimurium LT2 and *S.* aureus attachment, compared with the bare gloves.

1.2.2 Anti-biofouling and anti-adhesive surfaces

Among various types of smart protective coatings for food-contact surfaces; anti-biofouling and anti-adhesive materials have received growing attention over the past decade. These surfaces rely on superhydrophobic effect which repels aqueous residues, spills, and droplets carrying bacterial suspensions. The synergistic combination of nanotexturing and surface functionalization (i.e., chemical modification) is required to produce superhydrophobic coatings (X.-M. Li, Reinhoudt, & Crego-Calama, 2007; J. K. Oh, Lu, Min, Cisneros-Zevallos, & Akbulut, 2015). The schemes for fabricating superhydrophobic anti-fouling/anti-adhesive coatings include photolithography (Roach, Shirtcliffe, & Newton, 2008), etching (Xue et al., 2015), chemical vapor deposition(R. Yang, Xu, Ozaydin-Ince, Wong, & Gleason, 2011), and layer-by-layer deposition (Bastarrachea, Denis-Rohr, & Goddard, 2015). Some of these nanotechnology-based approaches have been taken advantage of in food safety applications in the form of protein-repellent surfaces(M. Zhang et al., 2014), zwitterionic surfaces (Cheng, Zhang, Chen, Bryers, & Jiang, 2007), stimuli-responsive polymers (Cunliffe, de las Heras Alarcón, Peters, Smith, & Alexander,

2003), biomimetic materials (Bixler & Bhushan, 2014), polyethylene glycol (PEG)-based polymer coatings (G. Li et al., 2008), stimuli-responsive polymers (so-called "smart polymers") (Shivapooja et al., 2013), heparin-based layer-by-layer coatings (Fu, Ji, Yuan, & Shen, 2005), and amphiphilic coatings (Krishnan et al., 2006).

Although, Hizal has already developed superhydrophobic surface by with 162° water contact angle by coating Teflon on the aluminum surface treated by the anodizing and post-etching processes. Their surfaces decrease the adhesion of *E.* coli K12 and *S.* aureus ATCC12600 more than 99.5% in comparation to bare surfaces (Hizal et al., 2017). The key challenge in achieving translation of these novel, advanced technologies to large-scale use in food industry is the need for complex procedures and multiple steps in preparation and high-cost of intermediate chemicals and functional agents.

1.3 Objective

In this study, we describe anti-adhesive aluminum food-contact surfaces based on the scalable, economic dip-coating method of ultrafine silica particles and the sequential surface modification with nonpolar fluorine-containing terminal groups. Bacterial anti-fouling/anti-adhesion properties of the developed coatings was investigated using microbial enumeration and scanning electron microscopy (SEM) techniques, using *Salmonella* Typhimurium LT2 and *Listeria innocua* as surrogates for human pathogens. The physicochemical characterization of the developed smart coating was performed via attenuated total reflectance-Fourier transform infrared (ATR-FTIR) spectroscopy and by measurement of contact angle using sessile drop techniques. In addition, ATR-FTIR was also used for assessing the chemical stability and leaching resistance of the coating over prolonged period of time.

2. MATERIALS AND METHODS**

2.1. Preparation of aluminum surfaces

Aluminum sheets (thickness: 0.8 mm, ASTM B209; Metals Depot, Winchester, KY, USA) were cut into 10 mm × 10 mm pieces and then washed with Milli-Q water (resistivity of 18.2 MΩ·cm) and left to dry in air at room temperature (23 °C). Subsequently, the aluminum pieces were then soaked with isopropanol and acetone before a final rinsing step in Miili-Q water.

2.2. Preparation of silica nanoparticle-coated aluminum surfaces

First, a suspension of silica nanoparticles (an average diameter of ca. 200 nm, silica, fumed powder; MilliporeSigma Corp., St. Louis, MO, USA) was prepared to 0.5 wt% in deionized (DI) water by rigorous stirring. Then, the solution was placed in a probe-type ultrasonic homogenizer (SJIA-2000W; Ningbo Haishu Sklon Electronic Instrument Co., Ltd., Ningbo, China) and sonicated for 20 min until the particles became fully suspended (confirmed by dynamic light scattering). The prepared alumina sheets were further cleaned and compatibilized with silica nanoparticles via by oxygen plasma treatment (PDC-32G (115 V); Harrick Plasma, Ithaca, NY, USA) for 45 sec. The treated alumina surfaces were submerged in silica suspension to induce the deposition and physisorption of silica particles on them (Fig. 2). The deposition step involves dipping the surfaces into the suspension for 1 min and sequentially air-drying for 1 min for four times repeatedly. Then, the silica nanoparticle-coated aluminum surfaces were then left to air dry

at room temperature for 12 h. Finally, silica on alumina was permanently fused via sintering for 2 h at 600 °C.

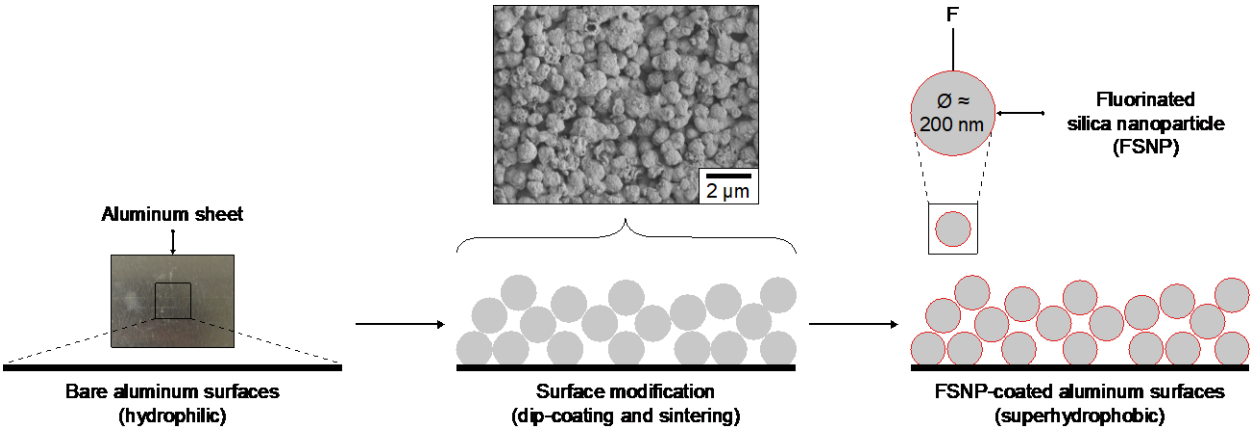

Fig. 2. Surface modification of aluminum surfaces with "fluorinated silica nanoparticle" (FSNP) to display desired bacterial anti-adhesion properties. SEM micrograph shows surface morphology of the aluminum surfaces after dip-coating and sintering processes.

2.3. Affixation of nonpolar groups to nanotexture

A 5 mM of trichloro(1H,1H,2H,2H-heptadecafluorodecyl)silane (THFS, purity: >96.0%; TCI America, Portland, OR, USA) was added dropwise into 20 mL of hexane (ACS grade; Avantor Performance Materials, LLC, Center Valley, PA, USA). The solution was covered with an aluminum foil and secured with a plastic paraffin film to reduce evaporation. The mixture was sonicated in a sonication bath for 5 min to throughly solubilize the silane in hexane. The silica nanoparticle-coated aluminum surfaces were placed in the silane solution and covered again with an aluminum foil and a plastic paraffin film to ensure the silane molecule to chemically react with the silica nanoparticles. After 2 h, the pieces could be removed from the silane solution and then left to air dry at room temperature for 30 min, resulting in fluorinated silica nanoparticle (FSNP)-coated aluminum surfaces.

2.4. Physicochemical characterization of the coating

The chemical composition of the resultant coating was analyzed by attenuated total reflectance-Fourier transform infrared (ATR-FTIR) spectroscopy using an IRPrestige-21 (Shimadzu Corp., Kyoto, Japan). The IRsolution software version 1.40 (Shimadzu Corp., Kyoto, Japan) was used to identify and analyze the functional groups and chemical elements.

To determine the surface wetting (i.e., hydrophilic or hydrophobic) characteristics of the aluminum surfaces, static water contact angle was measured by using a sessile drop technique (the volume of a water droplet was fixed at 5 μL)(Kwok, Gietzelt, Grundke, Jacobasch, & Neumann, 1997). The ImageJ software (National Institutes of Health (NIH), Bethesda, MD, USA) was used to analyze the contact angle by using a plug-in named Low-Bond Axisymmetric Drop Shape Analysis (LBADSA) (Stalder et al., 2010). Reported contact angles are the average of at least five measurements at room temperature.

2.5. Bacterial cultures

Two bacterial isolates were used in this study, gram-negative *Salmonella* Typhimurium LT2 (ATCC 700720) and gram-positive *Listeria* innocua (NADC 2841). Fig.3 shows the procedure of the bacterial cultures. Working cultures of *S.* Typhimurium LT2 and *L.* innocua were prepared by transferring a loopful (10 μL) of culture from tryptic soy agar (TSA; Becton, Dickinson and Co., Franklin Lakes, NJ, USA) slants to culture tubes containing 9.0 mL of tryptic soy broth (TSB; Becton, Dickinson and Co., Franklin Lakes, NJ, USA) and 9.0 mL of TSB containing 0.6% yeast extract (TSB-YE; Becton, Dickinson and Co., Franklin Lakes, NJ, USA), respectively. The tubes

of all strains were incubated aerobically at 37 °C for 24 h without shaking. A second transfer was completed by transferring a loopful culture to fresh culture medium (i.e., TSB and TSB-YE) and then incubating under the same conditions. The final concentrations of both bacteria in the culture media ranged from 8.8 to 9.2 log10 CFU/mL.

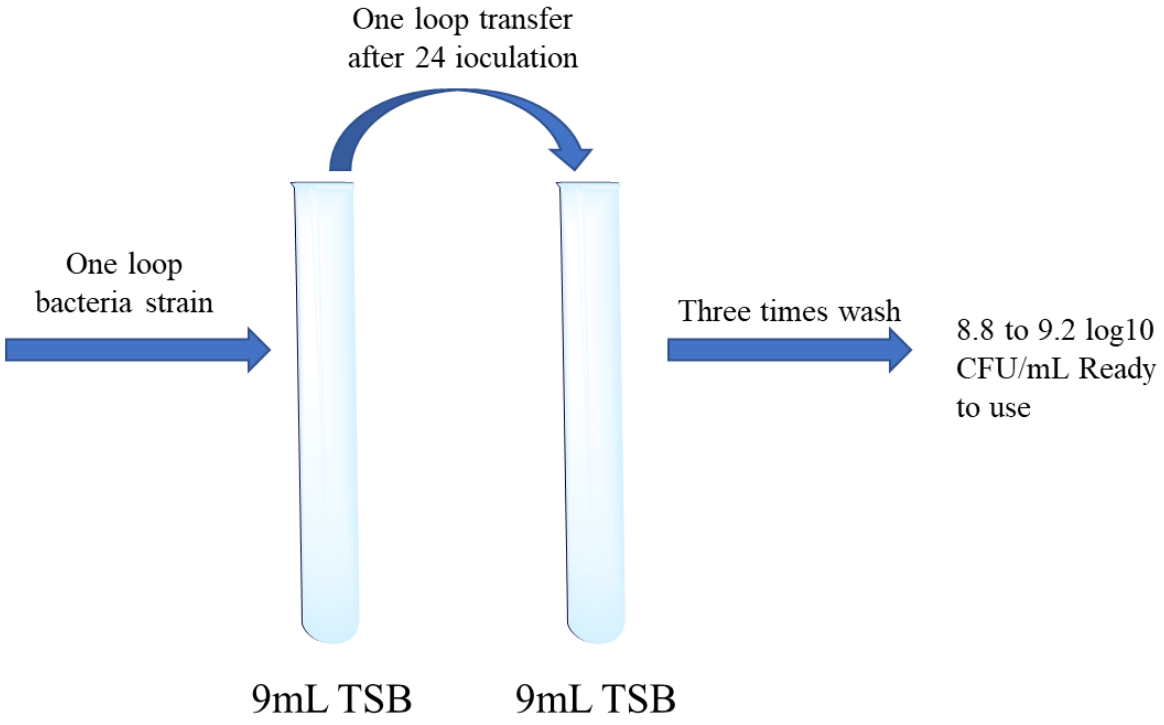

Fig. 3 The scheme of bacterial culture preparation process.

2.6. Surface inoculation

To inoculate the surfaces, the bare and functionalized, nanotextured aluminum coating were submerged in 9.0 mL of bacterial suspension (bacterial population density: 8.8–9.2 1og CFU/mL) at room temperature for 1 day and 7 days, respectively. For 7 days-exposed samples, the bacterial suspension was replaced to fresh medium every 24 h. Later, the samples were removed from the

bacterial suspension and rinsed gently with Milli-Q water to dislodge loosely attached bacterial cells. Remaining bacteria which were not removed by rinsing were regarded as attached bacteria.

2.7. Bacterial adhesion assay

Bacterial adhesion on the bare and the coated aluminum surfaces were enumerated on Petri plates containing TSA, and by counting attached bacteria using SEM. Fig.4 shows the procedure of preparing the plating containing TSA. In plate counting studies, aluminum pieces which were inoculated for 1 day were vortex-mixed vigorously in 0.1% peptone water (Thermo Fisher Scientific, Waltham, MA, USA) for 5 min to detach the bacteria from the material surfaces. Afterwards, 0.1% peptone water containing detached bacteria were decimally diluted serial times and plated on the TSA Petri plates. The bacterial population densities of detached bacteria from the bare and FSNP-coated aluminum surfaces were determined after 24 h of aerobic incubation at 37 °C in atmospheric conditions. All experiments were replicated three times.

The direct count of attached bacteria on the bare and the coated aluminum surfaces was performed on aluminum pieces inoculated with bacteria for 1 day and 7 days using SEM. Prior to SEM imaging, in order to inactivate bacterial strains completely, bacteria were exposed to small amounts (at concentrations above 10 mg/m3) of acrolein (MilliporeSigma Corp., St. Louis, MO, USA) vapor. Next, palladium and platinum (Pd/Pt) alloy was coated on the sample surfaces with a film thickness of 15 nm to reduce the charging effects, followed by inactivation. At least ten different selected areas were observed and analyzed to quantify the attachment of *S.* Typhimurium LT2 and *L.* innocua.

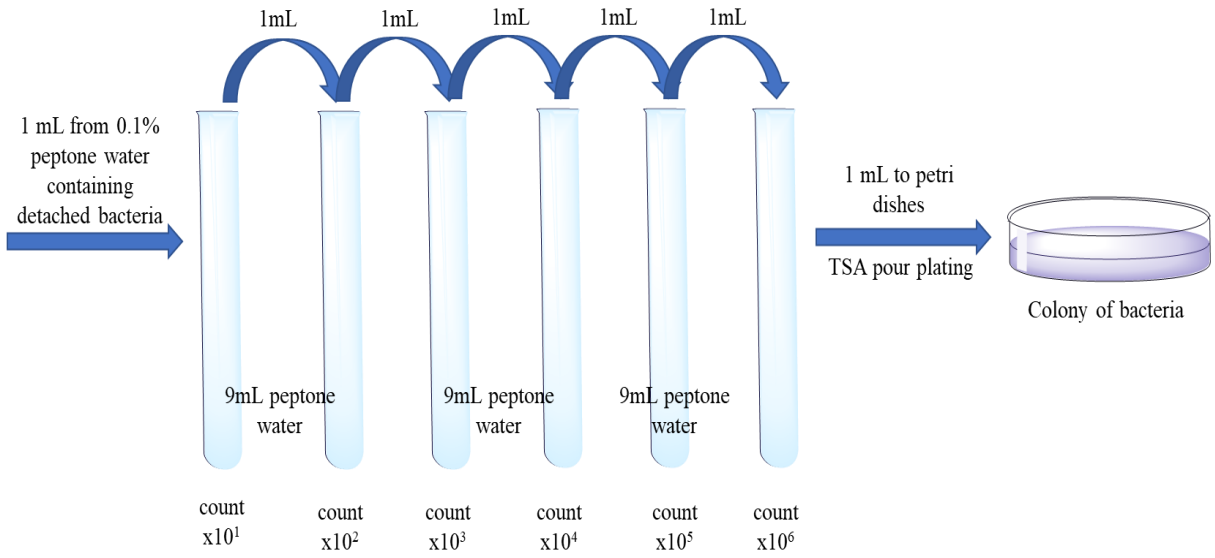

Fig. 4 The scheme of dilution process of the pour plating test

2.8. Determination of mud adhesion

Mud solutions with different viscosities were prepared by mixing dry soil with water in different ratios to achieve viscosities in the range of 0.1 Pa.s to 100 Pa·s. The viscosity of the mud solution was measured using a rotational rheometer (HAAKE RheoStress 1, Thermo Fisher Scientific, Waltham, MA, USA). Afterwards, the bare and the coated aluminum surfaces were completely submerged in mud solution for 1 min. The aluminum pieces were vertically removed from the mud slurry in a single-continuous motion. The images of the exposed surfaces were captured using a high-resolution digital camera. The area of mud remaining on the aluminum surfaces was analyzed using Gwyddion software version 2.31 (Czech Metrology Institute, Brno, Czech Republic) in order to determine the percentage mud coverage of the samples.

2.9. Chemical stability and leaching resistance test

The leaching resistance of the coated aluminum surfaces was determined in deionized (DI) water as a function of time. Fig.5 shows the procedure of the chemical stability and leaching resistance test. It was achieved by taking and analyzing the small volume aliquots (50 μL) collected from the samples, i.e., the coated aluminum surfaces submerged in DI water at various time intervals (1 day and 7 days). The analysis was done using the ATR-FTIR spectroscopy that can reveal any chemical leaching from the surfaces to solution with a detection limit of 1.0 ppm.

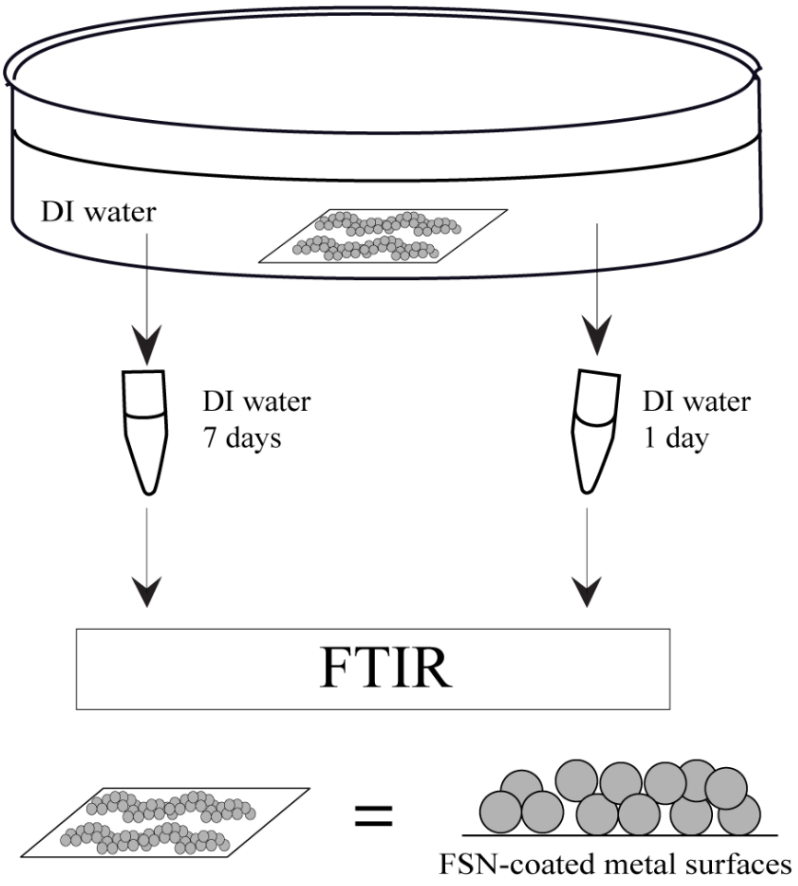

Fig. 5 The scheme of the chemical stability and leaching resistance test of the coated aluminum surfaces

2.10. Statistical analysis

Bacterial population data have been transformed using the logarithm function prior to the statistical analysis. One-way analysis of variance (ANOVA) with Tukey's post-hoc test was used to detect statistically differing plate counts in pathogen surrogates for bare and coated surface samples. Likewise, SEM-derived microbial counts were compared between the bare and FSNP-coated aluminum surfaces for each bacterium by one-way ANOVA and Tukey's post-hoc test. The p-value for statistical difference between means was set at $p = 0.05$. All statistical analysis was performed by using Analysis ToolPak in Excel software (Microsoft Corp., Redmond, WA, USA).

3. RESULTS AND DISCUSSION***

3.1. Characterization of the coating on aluminum surfaces

Fluorinated silane (i.e., THFS) compounds tend to have a high affinity toward the silica surfaces (Vansant, Van Der Voort, & Vrancken, 1995). c(Nishino, Meguro, Nakamae, Matsushita, & Ueda, 1999). To confirm the reaction between the THFS-functionalized silica nanoparticles and aluminum surfaces, the ATR-FTIR spectroscopy was used to determine the chemical groups on the surfaces of the samples. Fig. 6 shows the ATR-FTIR spectra of the bare and FSNP-coated aluminum surfaces. The bare aluminum surfaces had no peaks between 700 cm-1 to 1450 cm-1, which is the region containing peaks associted with C–F stretching (J.-K. Chen, Ko, Hsieh, Chou, & Chang, 2004). There was a strong peak at 1050 cm-1, which corresponds to an overlap between the Si–O–Si bond and –CF3 symmetric stretching for the FSNP-coated aluminum surfaces (Zeitler & Brown, 1957).

To determine the influence of nanotexturing and chemical modification of aluminum surfaces, we compared the wetting properties of the bare and FSNP-coated aluminum surfaces by measuring the contact angle of water droplets (Fig. 7). The static contact angle of water droplets on bare aluminum surfaces was $\theta = 74° \pm 1.3°$, indicating surfaces were highly hydrophilic, which is in agreement with previous experimental data (Jhee, Lee, & Kim, 2002). In comparison, the water contact angle of the coated aluminum surfaces is, $\theta = 161° \pm 0.6°$, showing a transformation from a hydrophilic behavior to superhydrophobicity.

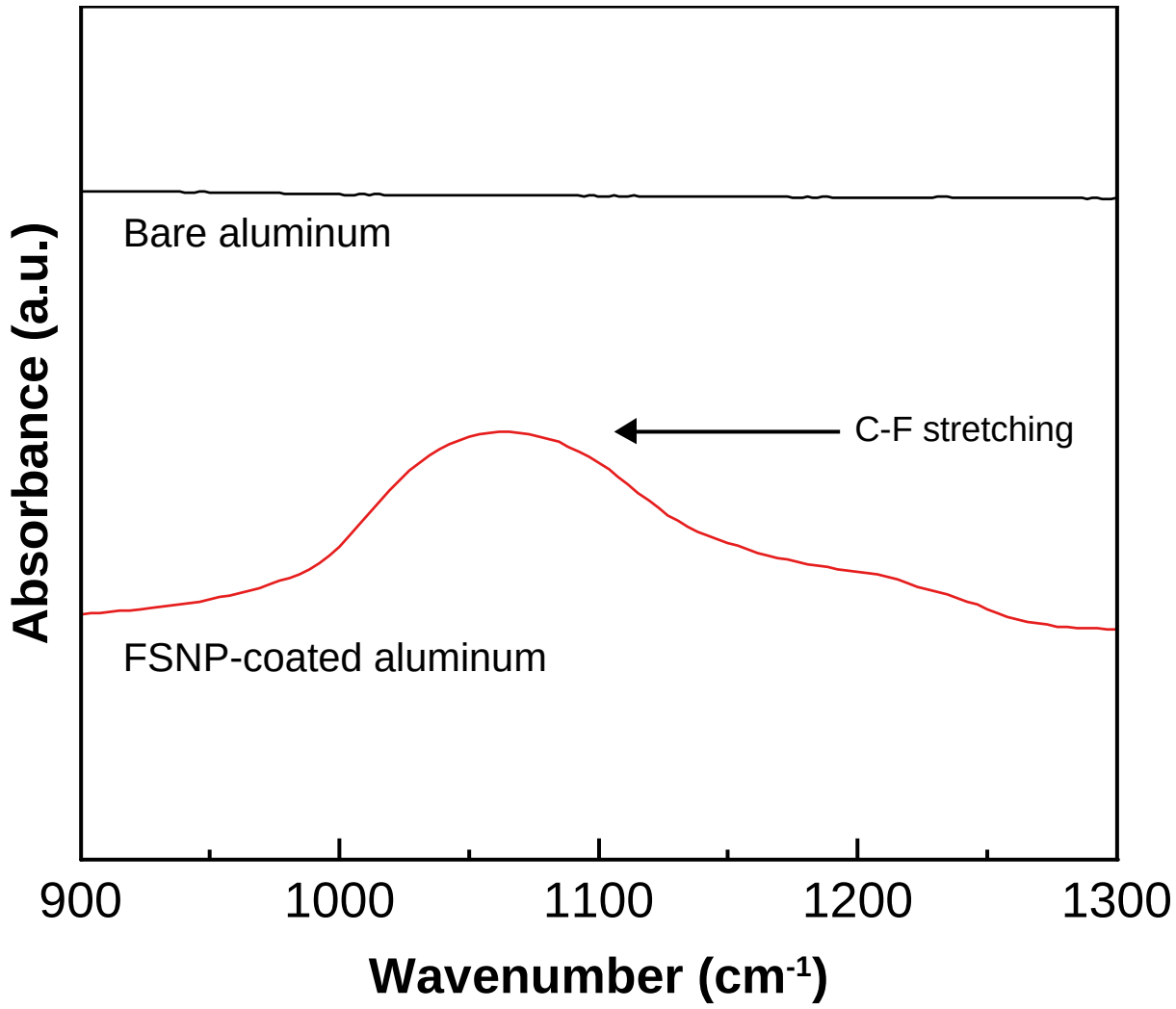

Fig. 6 ATR-FTIR spectra of the bare aluminum surface and fluorine-functionalized FSNP-coated aluminum surface. The emergence of a peak around 1050 cm^{-1} associated with C–F stretching indicates the presence of a fluoro-compound chemically bound on the alumina surface.

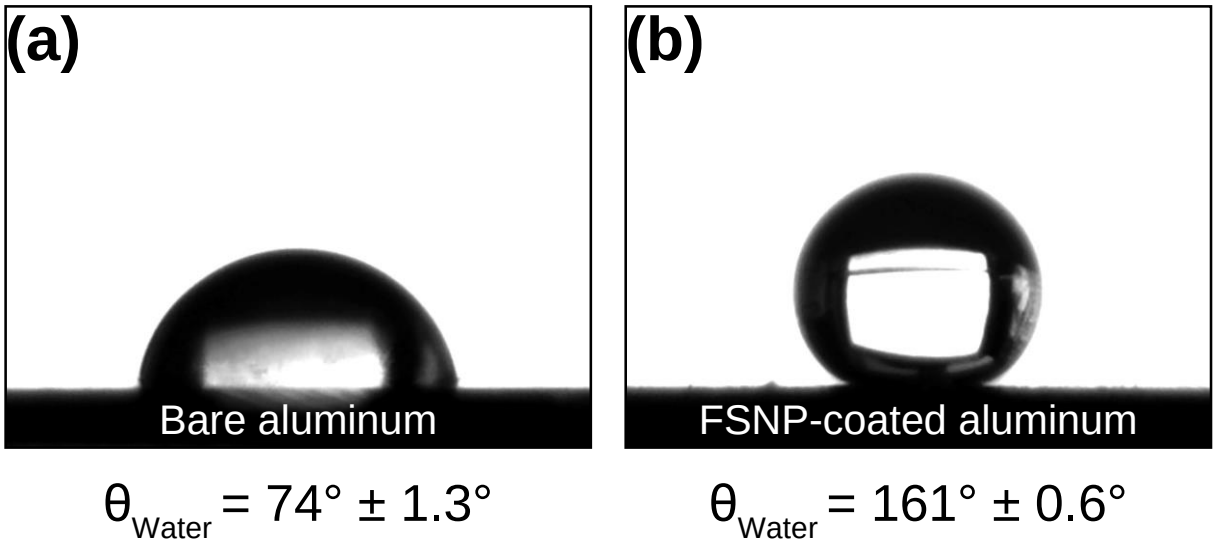

Fig. 7 The static contact angle of water droplets on the (a) bare and (b) FSNP-coated aluminum surfaces. The clear transition from a hydrophilic state to a superhydrophobic was attributed to the coating involving a synergistic combination of nanotexture and nonpolar fluorosilanes.

3.2. Quantification of bacterial attachment by plate counting

After characterizing the surface interfacial properties of the coatings, we investigated the attachment behaviors of gram-negative *S.* Typhimurium LT2 and gram-positive *L.* innocua isolates. Fig. 8 depicts attachment of bacteria to the bare and coated aluminum surfaces, after 24 h inoculation. The population mean of attached *S.* Typhimurium LT2 on the bare aluminum surfaces was 5.3 ± 0.2 log10 CFU/mL, indicating that the bare aluminum surfaces supported bacterial adhesion of the gram-negative bacterium. On the other hand, the population mean of adhered *S.* Typhimurium LT2 on the coated aluminum surfaces decreased to 3.3 ± 0.1 log10 CFU/mL, a >99.1% reduction in bacterial adhesion on coated aluminum surfaces versus bare surfaces. One-way ANOVA and Tukey's means separation procedures determined the difference in the

attachment of *S.* Typhimurium LT2 between the bare and coated aluminum surfaces was statistically significant (p-value < 0.05). Similar experiments were also performed with *L.* innocua. While the population mean of attached *L.* innocua on the bare aluminum surfaces was 5.3 ± 0.1 log10 CFU/mL, a bacterial count of 3.3 ± 0.2 log10 CFU/mL was observed on the coated aluminum surfaces, corresponding to a >99.0% reduction in bacterial attachment. As in the case of *S.* Typhimurium LT2, the attachment of *L.* innocua on the bare versus coated aluminum surfaces was statistically differed (p-value < 0.05). Overall, plate counting experiments concluded that a reduction of 2.0 to 2.1 log10 CFU/mL could be achieved by coating aluminum surfaces with nanotextured and fluorosilanes for *S.* Typhimurium LT2 and *L.* innocua.

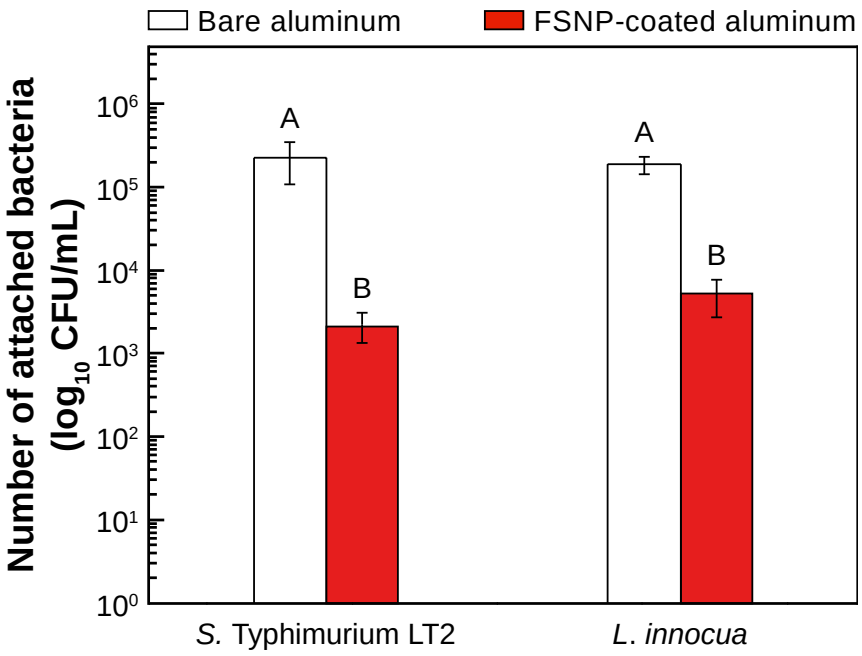

Fig. 8 The graph shows the number of attached gram-negative *S.* Typhimurium LT2 and gram-positive *L.* innocua to bare and FSNP-coated aluminum surfaces after 24 h exposure to the bacterial suspension. The letters A and B indicate statistically significant difference (p-value < 0.05) between means.

3.3. Quantification of bacterial attachment by electron microscopy

Additionally, to visually complement plating data, SEM was used to examine the counts and distribution of attached bacteria on the bare and coated aluminum surfaces. Fig. 9 shows the SEM micrographs of attached *S.* Typhimurium LT2 on bare and coated aluminum surfaces after 1 day and 7 days' inoculations. After a 1-day inoculation, the population mean of attached *S.* Typhimurium LT2 on the bare aluminum surfaces was 5.3 ± 0.2 log10 cells/mm^2. Conversely, the mean population of attached *S.* Typhimurium LT2 on the FSNP-coated aluminum surfaces was 2.9 ± 0.2 log10 cells/mm^2, which corresponds to a reduction of >99.0%. Likewise, while after 7 days' inoculation, the population mean of attached *S.* Typhimurium LT2 on the bare aluminum surfaces was 5.6 ± 0.1 log10 cells/mm^2, mean attached *S.* Typhimurium LT2 on the FSNP-coated aluminum surfaces numbered 3.1 ± 0.1 log10 cells/mm^2. This corresponds to a reduction of >99.5% in comparison to the bare aluminum surfaces. Essentially, numbers of attached *S.* Typhimurium LT2 to the superhydrophobically-modified coated aluminum surfaces were much less than on bare aluminum surfaces.

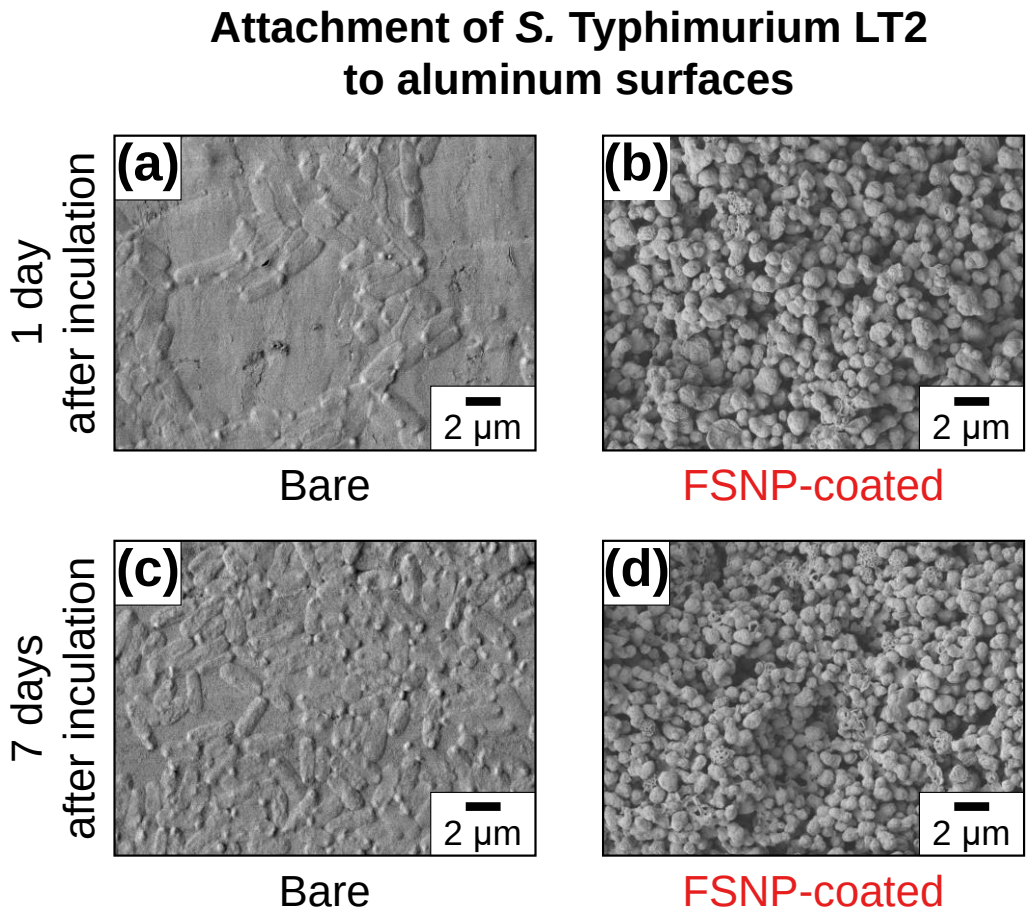

Fig. 9 High-resolution SEM micrographs of attached *S.* Typhimurium LT2 to the bare and FSNP-coated aluminum surfaces after (a),(b) 1 day and (c),(d) 7 days exposure to the bacterial suspension.

We also enumerated the numbers of attached *L.* innocua on bare and coated aluminum surfaces under the same inoculation conditions as for *S.* Typhimurium LT2. Fig. 10 demonstrates the SEM micrographs of attached *L.* innocua on these surfaces after 1 and 7 days' exposure to the bacterial suspension. After a 1 day inoculation, the population mean of attached *L.* innocua on the bare aluminum surfaces was 5.7 ± 0.1 log10 cells/mm^2, while the mean population of attached *L.* innocua on FSNP-coated aluminum surfaces was 2.8 ± 0.2 log10 cells/mm^2. Moreover, after 7 days of inoculation, the mean attached *L.* innocua on the bare aluminum surfaces numbered 6.2 ±

0.1 log10 cells/mm^2. Similar to *L.* innocua from 1 day inoculation experiments, the population mean of attached *L.* innocua on FSNP-coated aluminum surfaces was 3.1 ± 0.2 log10 cells/mm^2, corresponding to a >99.3% reduction in *L.* innocua adhering to FSNP-coated surfaces versus bare aluminum surfaces.

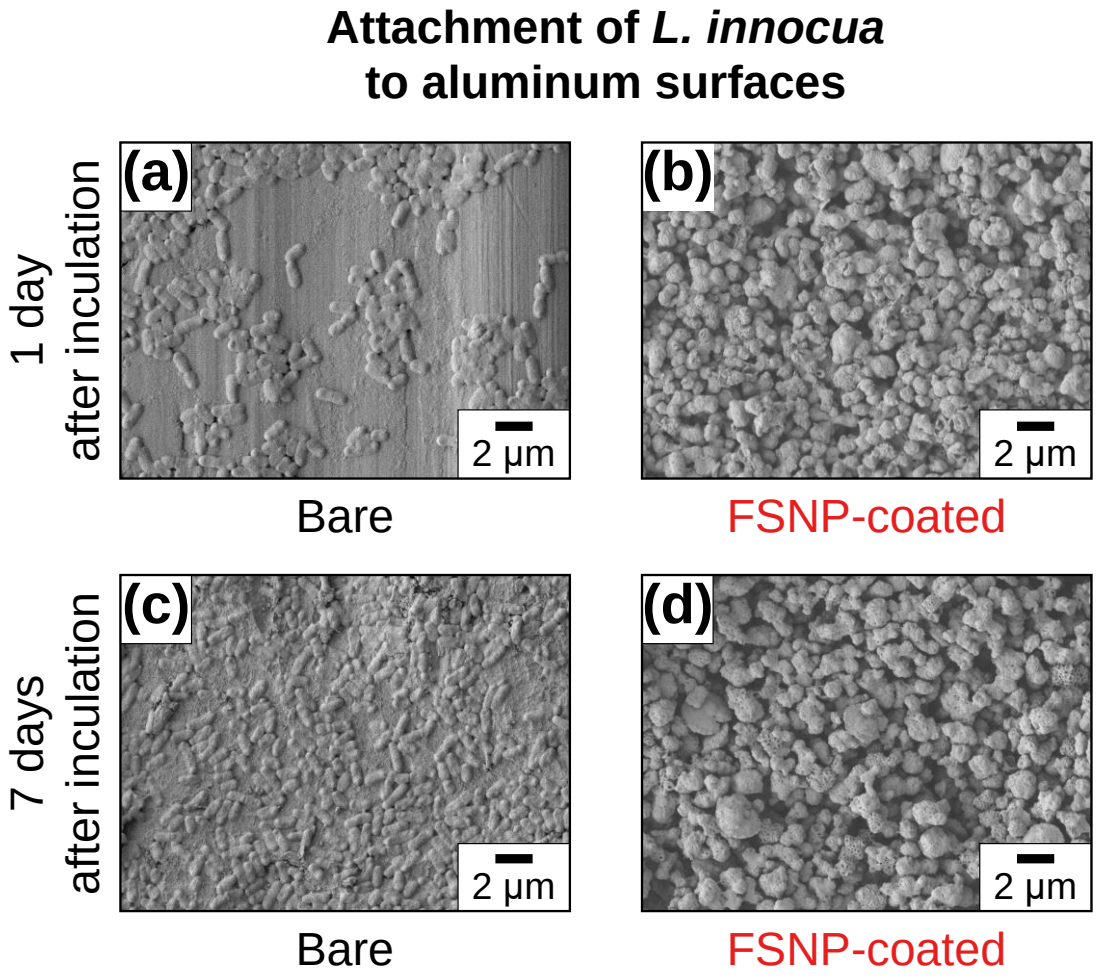

Fig. 10 High-resolution SEM micrographs of attached *L. innocua* to the bare and FSNP-coated aluminum surfaces after (a),(b) 1 day and (c),(d) 7 days exposure to the bacterial suspension.

Overall, both plate counting and SEM assays confirmed that the superhydrophobically modified aluminum surfaces strongly inhibit the bacterial attachment in comparison to the bare aluminum surfaces. Even after 7 days of exposure to the *S.* Typhimurium LT2 and *L.* innocua suspensions, bacterial attachment was significantly less on the coated aluminum surfaces, indicating that the nanotextured aluminum surfaces have the ability to repel the bacteria from the material surfaces effectively.

The reduced bacterial attachment may be explained by considering the wetting transition from the Wenzel model to the Cassie-Baxter model (i.e., air-pockets exist between the liquid and solid) due to the superhydrophobic characteristics of coating (L. Chen, Yang, & Wang, 2012) (Fig. 11). The transition to the Cassie-Baxter regime implies that the air-pockets form when water comes into contact with the coated aluminum surfaces, thereby decreasing the actual effective contact area between the bacterial suspension and coated aluminum surface (Crick, Ismail, Pratten, & Parkin, 2011). The observed phenomenon can be also explained in terms of the superhydrophobic effect (Southall, Dill, & Haymet, 2002) due to highly nonpolar fluorine groups (Dalvi & Rossky, 2010).

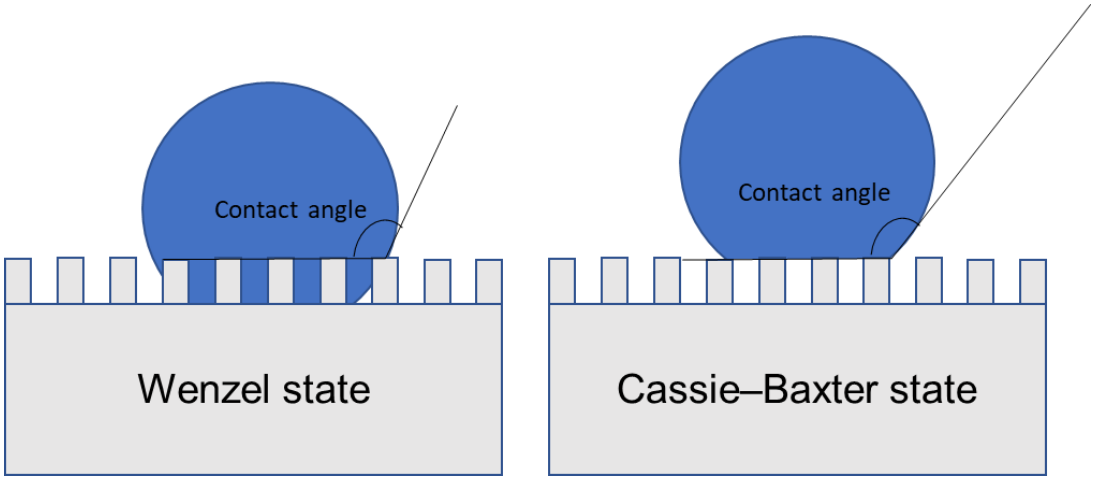

Fig. 11 Droplet behavior on a nanostructure surfaces

3.4. Mud adhesion assays

As soil is a source of bacterial contaminants, we tested the adhesion behavior of mud on the developed coatings using muds of varying viscosities (0.1–100 Pa·s). Fig.13 represent the comparison of dipping the bare metal surface and coated aluminum surfaces into the mud. After keeping the surface samples in the mud and then carefully removing them, we quantified the mud adhesion (Fig. 12). It was found that the mud coverage on the FSNP-coated aluminum surfaces was very low (1.2–3.3%), even at high mud viscosities. On the other hand, the mud remaining on the bare aluminum surfaces was much higher, in the range of 33.3 to 94.1% depending on the mud viscosity.

These results indicate the potential of these surfaces for contributing sustainability considering that all equipment, tools, containers, accessories, and surfaces used for storage, sorting, and processing of food need to be routinely and in some cases frequently washed, cleaned, and sanitized, representing a large requirement for potable water for food safety purposes. Due to

rising costs, changes in public preferences regarding water allocation among competing uses, increasing budget scrutiny in the national and state legislatures, and increasing awareness of climate, there is a growing need to improve water-use efficiency. Because of their self-cleaning nature, these coatings will require a smaller amount of water use for cleaning and sanitization, potential contributing the water efficiency and sustainability in the food industry.

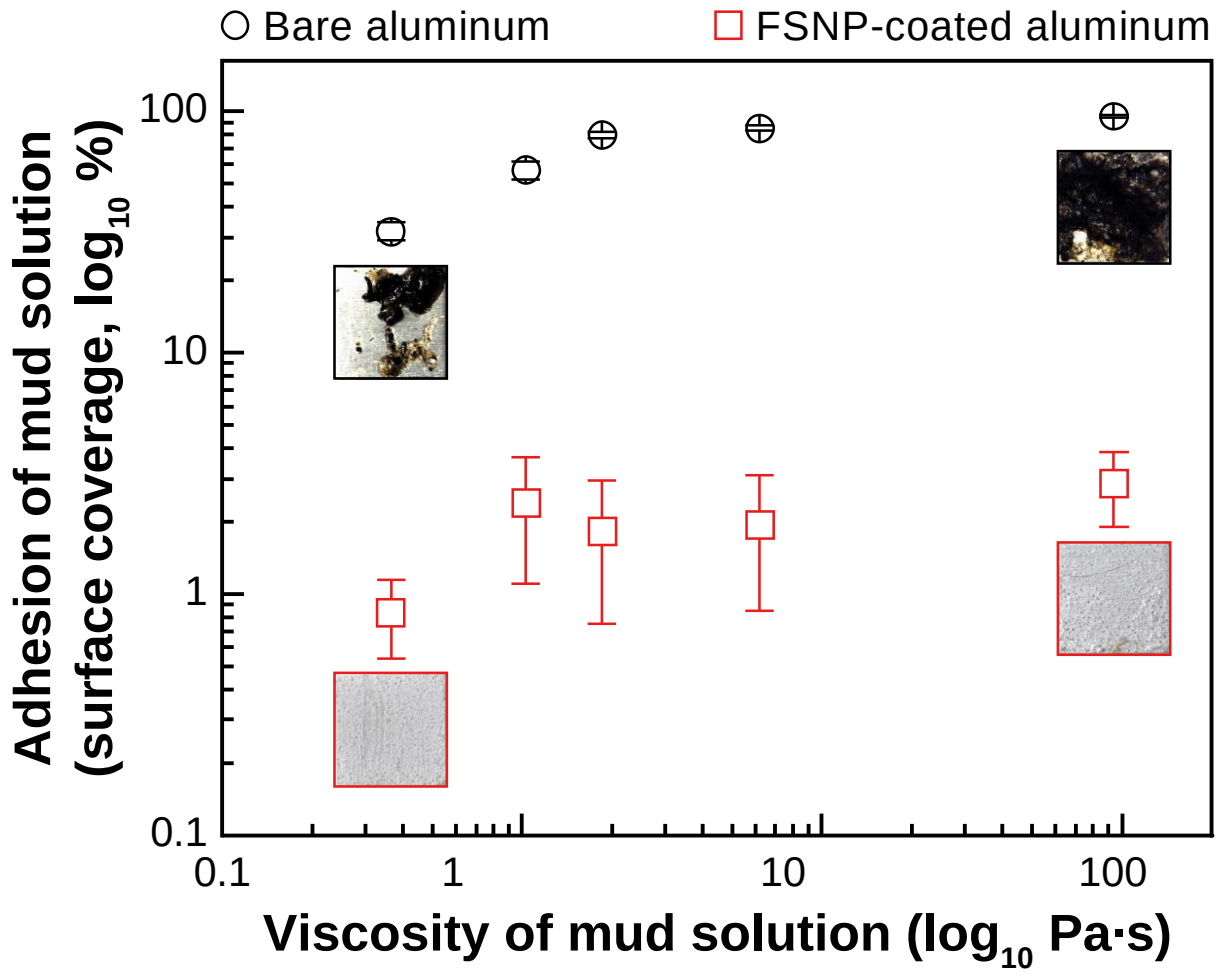

Fig. 12 A plot of viscosity versus surface coverage of mud solution for the bare (black circle) and FSNP-coated (red square) aluminum surfaces. (For a linear scale plot, see Supplementary Data).

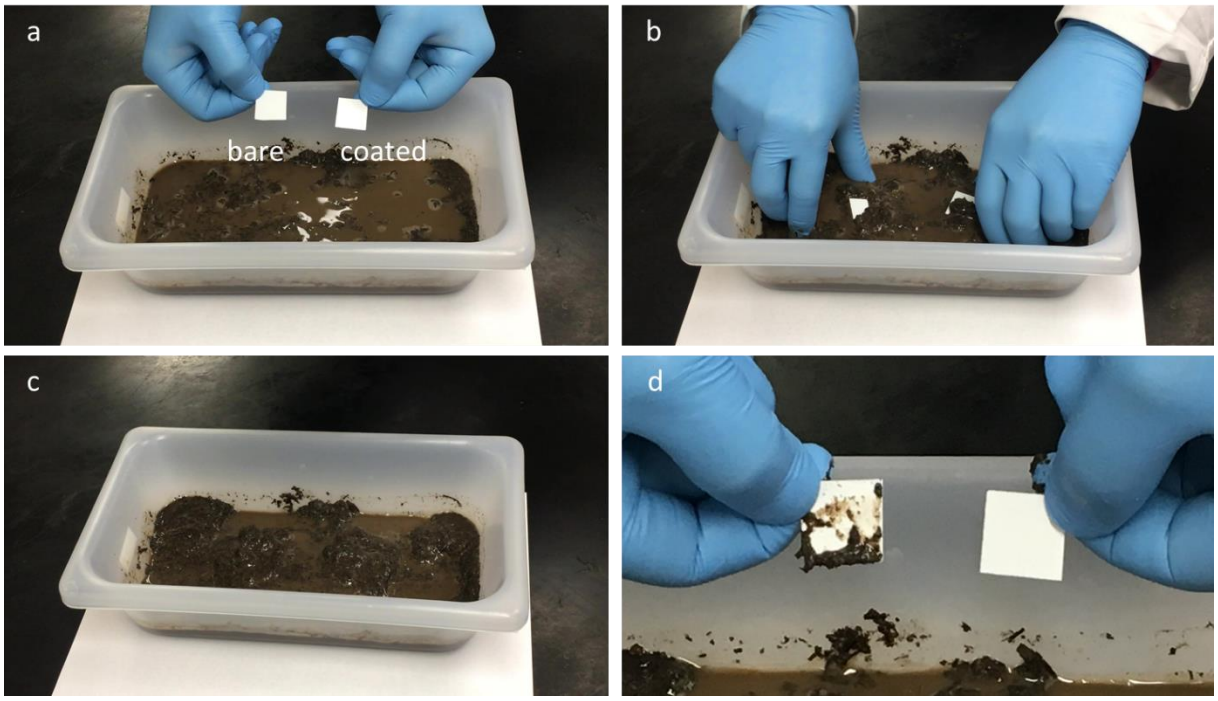

Fig. 13 Full process of the mud test

3.5. Leaching durability of coating

To examine if there is any chemical leaching possibility from the coated aluminum surfaces, the degradation or detachment dynamics of fluoro-groups was monitored using ATR-FTIR spectroscopy. The data revealed that for the solution containing the FSNP-coated samples, there was no asymmetric and symmetric C–F stretching peak between 1000 cm-1 to 1250 cm-1 during the time frame of these experiments (up to 7 days). These findings indicate the absence of unbound chemicals from the coated aluminum surfaces and no leaching at a detection limit of 1.0 ppm.

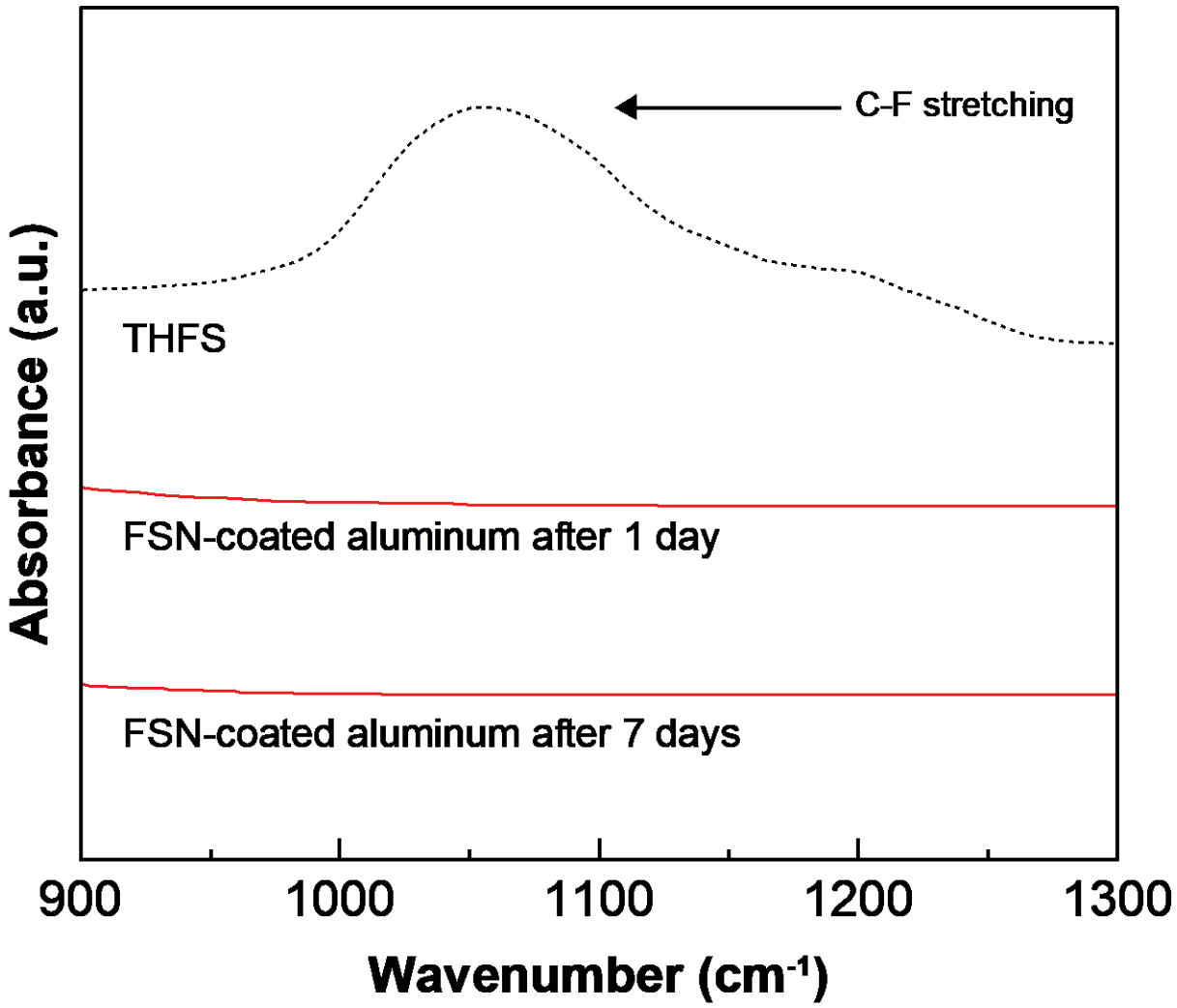

Fig. 14 ATR-FTIR spectra of aliquots collected from the samples (i.e., FSNP-coated aluminum pieces) submerged in DI water for 1 day and 7 days. The results indicate the chemical stability (no leaching) of functional groups on the FSNP-coated aluminum surfaces, confirmed by the absence of C–F stretching absorption in solution.

4. CONCLUSIONS[****]

This work demonstrates the proof-of-concept that the nanotexturing and chemical functionalization of aluminum surfaces with a combination of ultrafine silica particles and nonpolar hydrocarbons can incorporate bacterial anti-adhesive and mud-repellent properties on aluminum food-contact surfaces. The bacterial super-repellency of the coated aluminum was confirmed against *S.* Typhimurium LT2 and *L.* innocua. Furthermore, the mud adhesion was reduced by 10-fold on coated aluminum surfaces compared to the bare aluminum surfaces. Along with superior bacterial anti-adhesion properties, versatile applicability, and scalability, easy-to-clean characteristics make this functionalization process useful for food-contacting surfaces and sanitary design of food processing equipment. Overall, we have developed advanced bacterial anti-adhesive materials for controlling bacterial attachment and reducing resultant scenarios of cross-contamination and transmission of pathogens and foodborne diseases.

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