



Superhydrophobic coatings for food packaging applications: A review

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ABSTRACT

Food waste is a serious problem in our modern era, causing economic loss and exacerbating issues like hunger, environmental pollution, and water shortage. Residual food is one main culprit that can be easily eliminated by proper packaging. Advanced packaging techniques with self-cleaning and anti-fouling capabilities are critically important to tackle this issue. In this regard, superhydrophobic coatings are emerging as an innovative approach to address many critical issues in the food industry. Superhydrophobic coatings can prevent fouling and contamination of food packages. An additional capability is the minimization of food waste and improving consumer experience due to the easy sliding of food from the inner side of the package. In this article, we provide an overview of recent studies on the application of superhydrophobic coatings and surfaces for food packaging applications, with a focus on studies aimed at reducing residual food waste via superhydrophobic coatings prepared from edible, nontoxic, and ecofriendly materials.

1. Introduction

Noble laureate Seamus Heaney wrote “A rat-grey fungus, glutting on our cache. The juice was stinking too. Once off the bush, the fruit fermented, the sweet flesh would turn sour.” He was describing spoiled blackberries. Indeed, food waste is an everyday occurrence in our modern life with significant repercussions on society, the economy, and the environment (Munesue et al., 2015; Roodhuyzen, Luning, Fogliano, & Steenbekkers, 2017). For example, a recent United Nations report estimates the wastage of 931 million tons of food globally in 2019 alone. Considering the fact that globally there are 690 million people without enough food to survive, the amount of waste is disquieting (Forbes et al., 2021). Furthermore, wasted food means wastage of water and land (Vilariño et al., 2017). Therefore, reducing food waste is a rational approach to address urgent issues such as food insecurity, water scarcity, global warming, and pollution (Marston, Read, Brown, & Muth, 2021; Poyatos-Racionero, Ros-Lis, Vivancos, & Martínez-Máñez, 2018).

A convenient approach to reduce food waste is via smart packaging (Goddard & Herskovitz, 2020). Food packaging conventionally has been used as a physical barrier to protect and preserve key qualities of food in the farm-to-fork processes (Brody, Bugusu, Han, Sand, & McHugh, 2008; Cataldi et al., 2019; Duncan, 2011; Mustafa & Andreescu, 2020). Smart

packaging, on the other hand, incorporate added functionalities such as antioxidant, antibacterial properties, and sensing capabilities to indicate parameters like freshness, pH, etc. while also able to actively affect those parameters to conserve food quality and extend shelf life (Baek, Maruthupandy, Lee, Kim, & Seo, 2020; Heredia-Guerrero et al., 2018; Jo et al., 2020; Park et al., 2020; Romero, Sharp, Dawson, Darby, & Cooksey, 2021; Vilas et al., 2020). Another functionality that can be easily added to packaging is water and grease repellency, which also brings about improved barrier property and enhanced humidity resistance (Balasubramaniam et al., 2020; Lee & Robertson, 2021; Samyn, 2013; Zhang et al., 2019). Here, grease and water repellent packaging are beneficial for reducing residual food waste, especially viscous foods such as honey and syrup that stick to the container, leaving up to 15% of content stuck to the packaging material surface (Zhang et al., 2019). Since packaging-related food waste constitutes the major component of total food waste, especially in households, it is important to target (Manzocco, Alongi, Sillani, & Nicoli, 2016; Silvenius et al., 2014). Besides extending shelf life and optimizing packaging material, size, and design to facilitate opening and re-closing, making packaging material repellent to various foods is considered as an important factor relevant to household food waste reduction (Silvenius et al., 2014; Wikström, Williams, Trischler, & Rowe, 2019). In this respect, superhydrophobic

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coatings are emerging as an innovative approach to tackle many critical issues in food packaging (Ghasemi & Niakousari, 2020; Li et al., 2018).

Superhydrophobic coatings are extremely repellent to liquids and thus the solid-liquid contact area is minimal, and adhesion is weak. Initially developed to impart surfaces with special functionalities such as self-cleaning (Bhushan & Jung, 2011; Celik, Torun, Ruzi, Esidir, & Onses, 2020), anti-adhesion/anti-fouling (Bixler & Bhushan, 2012; Celik et al., 2021; Marmur, 2006), and anti-icing properties (Farhadi et al., 2011; Kreder, Alvarenga, Kim, & Aizenberg, 2016), superhydrophobic coatings found wide application varying from antibacterial surfaces (Heinonen et al., 2014; Shateri Khalil-Abad & Yazdanshenas, 2010), to enhancing molecular sensing (Korkmaz et al., 2021; Liang et al., 2017) and energy harvesting (Hu et al., 2020; Ipekci et al., 2021). In recent years, researchers are starting to exploit the novel properties of superhydrophobic coatings and surfaces in the food industry and packaging applications (Ghasemi & Niakousari, 2020).

In this article, we review recent studies on the application of superhydrophobic coatings in food packaging, with a particular focus on the fabrication techniques that utilize eco-friendly materials and methods. This topic is motivated by the fact that the widespread usage of hydrophobic molecules (such as fluorocarbons) has started to raise concerns about environmental pollution and human health, due to their toxicity, persistence, and bioaccumulation (Bayer, 2020; Glenn et al., 2021). Since packaging materials directly contact food, it is important to eliminate or reduce the consumption of toxic chemicals (Pinto et al., 2021; Terzioğlu, Güney, Parin, Şen, & Tuna, 2021). Indeed, several cities and countries already enacted laws to ban or limit the usage of perfluorinated chemicals in food packaging (NY State Senate, 2020; US Congress, 2021). Therefore, this article focuses on superhydrophobic coatings that are ideal for food packaging applications due to their non-toxicity and environmental friendliness (Wang, Zhao, Han, & Tam, 2022). The latter concept includes the usage of eco-friendly materials, including solvents that are commonly used for preparing coatings. This is significant since solvents are generally the main component of typical coatings, oftentimes constituting more than 90% of the dispersion. However, upon treatment of the desired surfaces, the solvent evaporates, resulting in significant material waste and potential environmental problems (Prat et al., 2015). Furthermore, some of the commonly used solvents pose safety and health risks (Joshi & Adhikari, 2019).

This article is organized as follows. First, we briefly introduce wetting phenomena and superhydrophobicity. We then present recent studies on the fabrication of superhydrophobic coatings that apply to any material surfaces and are prepared using water or other nontoxic chemicals as the solvent, employing eco-friendly materials and techniques. Afterward, methods for superhydrophobic modification of food packaging materials will be presented. This section focused on intrinsically hydrophilic metal and cellulose based materials, whereas hydrophobic polymeric food-packaging materials were excluded. In the end, we provide some prospects on the field and future directions to enable the practical application of superhydrophobic coatings in the food packaging industry. Readers interested in the broader topic of superhydrophobicity can refer to previous review articles focused on the fundamentals (Drelich & Marmur, 2014), fabrication techniques (Simpson et al., 2015), and practical applications (Zhang, Wang, Sun, Song, & Deng, 2021).

2. Wetting property of materials

Wetting is the ability of a liquid to spread on a solid surface that depends on the properties of both liquids and solids. A small liquid droplet placed on a solid surface can completely wet the surface, forming a thin layer of liquid film, or can take a spherical cap shape, forming a finite angle with the solid surface (Drelich, 2019; Gao & McCarthy, 2009; Marmur, 2009; Quéiré, 2008). The wetting ability of liquids on solid surfaces is typically characterized by the measurement of contact angles (CA): the angle formed between the liquid and the

vapor interface tangent, and the solid surface (Kwok & Neumann, 1999; Marmur, 2009). The contact angle of a liquid droplet formed on an ideal flat surface is called Young's CA, in honor of Thomas Young (Young, 1805). The Young's CA can be related to the interfacial tensions via Young's Eq. (1):

$$\cos(\theta) = (\gamma_{sv} - \gamma_{sl})/\gamma_{lv} \quad (1)$$

where θ is the contact angle, γ_{sv} is solid-vapor interfacial tension, γ_{sl} is solid-liquid interfacial tension, and γ_{lv} is the liquid-vapor interfacial tension (Marmur, 2009). Note that γ_{lv} is typically referred to as surface tension of liquids and γ_{sv} is the surface energy of solids. Interfacial tension can be defined as force per unit length or the unit energy to generate an interphase with common units of millinewtons/meter (mN/m) or millijoules/square meter (mJ/m²), respectively (Drelich, 2019). Among common liquids, the surface tension of water is high, with a value of 72.8 mN/m. Conversely, the surface tension of liquid alkanes, such as n-hexane (18.4 mN/m), is small. The surface energy of fluorinated hydrocarbons, which are conventionally used for hydrophobic treatment, is very low. For example, the surface energy of atomically flat C₂₀F₄₂ is 6 mJ/m², which is the lowest known to mankind (Nishino, Meguro, Nakamae, Matsushita, & Ueda, 1999). The surface tension of common edible liquids is given in Table 1. It should be noted that the surface tension may depend on the brand and type of liquids. Here, they are listed to show the range of values.

Real surfaces can be rough and heterogeneous, therefore, the contact angle of liquids on such surfaces deviates from the prediction of Young's equation (Vuckovac, Latikka, Liu, Huhtamäki, & Ras, 2019). Accordingly, the measured contact angle can be termed apparent CA, and unless stated otherwise, this is the angle that is generally reported in the literature and simply called contact angle or at times static contact angle. Other important terms concerning wetting are illustrated in Fig. 1 and include sliding angle (SA), advancing (θ_{ac}) and receding (θ_{rc}) contact angle, and contact angle hysteresis (CAH), which is the difference between advancing and receding contact angle (Huhtamäki, Tian, Korhonen, & Ras, 2018; Liu, Vuckovac, Latikka, Huhtamäki, & Ras, 2019). On a liquid repellent surface, depending on the microstructure of the surface and its chemistry, a droplet generally exists in one of the two distinct states; Cassie-Baxter state where the liquid droplet stays on top of the rough surface where the air is trapped between the micro-asperities, as illustrated in Fig. 1 (Cassie, 1948; Long et al., 2015). The other state is called the Wenzel state where the droplet impales the micro-cavities (Wenzel, 1936; Whyman et al., 2008). The difference between the two states manifests itself on the CA and the CAH; the CA of a droplet on a Cassie state is high while the hysteresis is low. A water droplet on superhydrophobic surfaces generally takes the Cassie state

Table 1
The surface tension of common edible liquids.

Liquid type	Surface tension (mN/m)	Reference
Water	72.0	Wang and Guo (2020)
Orange juice	69.3	Zhao et al. (2018)
Orange juice	30.5	Wang and Guo (2020)
Lipton green tea	69.0	Li et al. (2018)
Gatorade	68.0	Li et al. (2018)
Apple juice	57.0	Li et al. (2018)
Tea	53.8	Wang and Guo (2020)
Wine	53.0	Li et al. (2018)
Energy drink	52.6	Wang and Guo (2020)
Beer	52.0	Li et al. (2018)
Cola	57.9	Sahin et al. (2021)
Cola	48.5	Wang and Guo (2020)
Whole milk	48.0	Li et al. (2018)
Yogurt	46.3	Wang et al. (2020)
Yogurt	40.4	Wang and Guo (2020)
Coffee	46.0	Li et al. (2018)
Coffee	36.8	Wang and Guo (2020)
Sunflower oil	30.6	Celik et al. (2021)

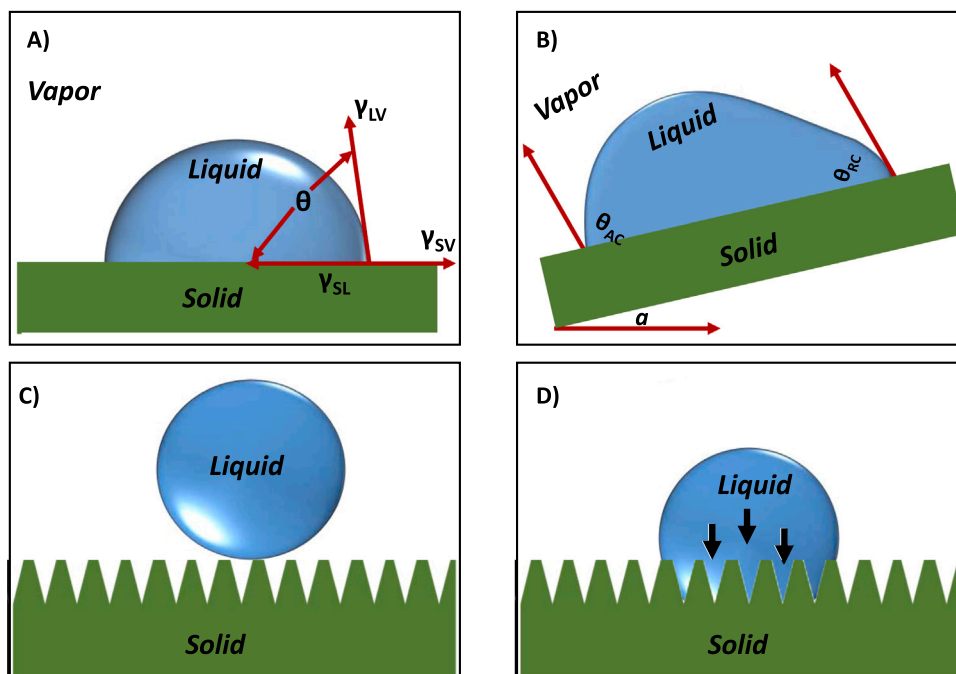


Fig. 1. Contact angles. A) Young's CA on a flat surface and its relation to the surface tension of the solid, liquid, and vapor. B) Illustration of advancing and receding CA. The sliding angle (SA) is represented with α . Also shown are the illustrations of a liquid droplet on a rough surface on the C) Cassie-Baxter state and D) Wenzel state.

Table 2

Materials used to fabricate superhydrophobic surfaces and type of tests conducted to evaluate their durability.

Material type	Materials	Wetting Characteristics	Tests	Reference
Natural	Waxes	CA = 161°, SA = 4°	Cell viability	Zhang et al. (2019)
		CA = 165°, CAH = 2°	Thermal and chemical stability Water impact	Milionis et al. (2019)
		CA = 153°, SA = 2°	Mechanical durability Biodegradability	Wang et al. (2020)
		CA = 155.8°, SA = 1.5°	Water stability Mechanical stability	Liu, Xue, An, Jia, and Xu (2019)
Clay	Clay	CA = 169°, SA = 3°	Mechanical, thermal, chemical stability, food residue test	Sahin et al. (2021)
		CA = 151.4°	Chemical stability, SERS antibacterial, water impact test, food residue test, leaching test	Li et al. (2019)
		CA = 153°	Water and air permeability test, flame retardancy test, water-oil separation, Mechanical stability	Wu, Pickett, Panchal, Liu, and Lvov (2019)
		CA > 170°, SA < 5°	Mechanical abrasion Washing test, bacterial resistance	Baidya, Das, Ras, and Pradeep (2018)
Synthetic	PDMS	CA > 160°, SA < 5°	Antibiofouling Thermal and Chemical stability	Razavi et al. (2019)
		CA > 150°	Mechanical durability	Li et al. (2021)
		$\theta_{adv} = 155^\circ, \theta_{rec} = 154^\circ,$ CA > 166.7°	Mechanical durability, adhesion, antibacterial, food residue test, cell viability	Liu et al. (2020)
		CA ≈ 160°, SA < 10°	Mechanical durability, underwater stability	Balu et al. (2008)
Alkyl ketene dimer	Alkyl ketene dimer	CA = 159°, SA = 5°	Tape test	Wang and Zhao (2021)
		CA = 159°, SA = 5°	Thermal and mechanical stability	Davis, Surdo, Caputo, Bayer, and Athanassiou (2018)
		CA = 159°, SA = 26°	Abrasion, chemical & UV stability, tape peel, antifouling	Ge et al. (2020)
		CA = 152.33°, SA = 8.25°	Laundry, abrasion chemical stability, self-healing	Wang, Chang, Ma, Qiu, and Tian (2021)
Alkyl ketene dimer	Alkyl ketene dimer	CA = 166°	Food residue test, mechanical and chemical stability	Arminger, Gindl-Altmuttner, Keckes, and Hansmann (2019)
		CA = 150°, SA = 17°	Color change, thermal stability	Reverdy et al. (2018)

where the water CA is larger than 150° and the sliding angle or CAH is below 10° . On a Wenzel state, on the other hand, the CAH is high. Therefore, to achieve superhydrophobicity, the surface roughness should be optimally engineered to make sure that the desired liquid droplets stay in the Cassie state (Bormashenko, 2015).

3. Applications in food packaging

The basic requirement to achieve superhydrophobicity is simple where one needs a rough-textured surface and low surface energy (Bhushan & Jung, 2011). Table 2 shows some of the commonly used eco-friendly materials that have significant food packaging-related applications. Sometimes it is possible to combine the two requirements where a textured surface of a low surface energy molecule can lead to superhydrophobic behavior. For example, the surface structure of the lotus leaf can be transferred to soft polymers like polydimethylsiloxane (PDMS) which is also a low surface energy molecule, resulting in an artificial superhydrophobic surface (Sun et al., 2005). In another recent report, Milionis et al. (2019) have shown a novel combination of molded cellulose micropillars and spray-coated carnauba wax.

Therefore, the strategy of combining low surface energy with texture has been utilized to prepare superhydrophobic coatings over the last decade (Teisala & Butt, 2019). In this section, we present an overview of the literature that has been published in recent years on superhydrophobic coatings aimed for food packaging and preservation, paying special attention to applications in reducing food waste using non-toxic, eco-friendly materials, such as plant-based waxes (Bayer, 2020; Saji, 2020; Wang et al., 2022). This aspect is important since any materials in contact with food may migrate into the food. Therefore, studies that did not directly demonstrate food packaging-related applications were excluded, as well as coatings that involve fluorocarbons and other chemicals that may arouse health concerns.

3.1. Materials and methods suitable for food packaging applications

Colloidal nanoparticles mixed with low surface energy molecules are presumably the simplest way to impart superhydrophobicity to a surface. In this approach, colloidal nanoparticles are deposited by spray-coating, drop-casting, or dip-coating (Bai, Zhang, Shao, Zhang, & Zhu, 2021; Hooda, Goyat, Pandey, Kumar, & Gupta, 2020; Sahoo, Yoon, Seo, & Lee, 2018). In the colloidal dispersion, the solvent is generally the major component, typically constituting more than 90% of the suspension. Following the deposition, the solvent would be lost due to evaporation. Therefore, reducing solvent usage or replacement with cheap, harmless, and eco-friendly solvents is an important measure that can be taken (Tian, Mendivelso-Perez, Banerjee, Smith, & Cademartiri, 2019). Water, without a question, is the best solvent in that regard; however, low surface energy molecules, are generally hydrophobic and do not disperse well in water. Thus, various processes have been developed to disperse these molecules in water. Davis et al. (2018) prepared PDMS-in-water emulsion by slowly (~ 10 droplets/min) adding water into PDMS elastomer (Sylgard 182) while under mechanical stirring, leading to a white creamy emulsion. The authors used hydrophilic SiO_2 nanoparticle dispersion in water as a mold, cured the PDMS, and evaporated water, leading to micro/nanotextured superhydrophobic monolith. Ge et al. (2020) and Zhang et al. (2020) tackled this problem by first plasma treatment of PDMS in air, producing OH groups that facilitate dispersion in water. Aided by ultrasonication and heating at 40°C , a 0.9% PDMS-in-water dispersion led to a stable and translucent emulsion with an average particle size of 719 nm. The authors dip-coated a cotton fabric with the dispersion followed by drying at 80°C for an hour. The coated fabric exhibits a water ($6\ \mu\text{L}$) contact angle of 159° and a sliding angle of 26° , respectively. One of the most significant results is the ability to self-heal where damages caused by mechanical abrasion or laundering can be reversed by annealing the damaged fabric at 80°C for 30 min or just leaving it to heal at room

temperatures for 24 h. The mechanism of self-healing, as hypothesized by the authors, is that the plasma-treated water-soluble PDMS penetrated the amorphous region of cellulose fibers and can diffuse out onto the surface to replenish the damaged or removed PDMS and orientate the hydrophobic CH_3 groups to decrease the surface energy. The applications in oil/water separation and self-cleaning are demonstrated. This study is an important step forward in fabricating environment-friendly superhydrophobic surfaces for real-world application, even though the step involved in plasma treatment may increase cost and hinder large-scale production.

Choi, Yoo, Park, and Kim (2017) proposed an ingenious method to prepare PDMS based superhydrophobic surface. They spread micro-sized ($< 63\ \mu\text{m}$) salt particles onto a PDMS surface before curing, leading to a rough-textured surface upon curing. The salt particles are dissolved by dipping the cured PDMS into the water, assisted by sonication. The result is a superhydrophobic surface with a contact angle of 151° and a sliding angle of 6° . The authors also demonstrated a simple and cost-effective way to fabricate a patterned (minimum pattern size of $500\ \mu\text{m}$) superhydrophobic surface for droplet manipulation. The superhydrophobic surface showed good resistance against acid/alkali solutions without showing any corrosion while dipped in acidic/basic solution for 12 h. Its resistance against linear abrasion and water droplet impact, though, appears to be weak, as acknowledged by the authors. Concerning practical application, the work by Cai et al. (2018) is a significant step where they used all water-based and commercially available PDMS or paraffin wax emulsion with water-based SiO_2 nanoparticle dispersion to impart extreme water repellency to polyester fabric, achieving water contact angle of 145° and sliding angle of 1.5° , respectively.

Aqueous dispersions of commercially available polyolefin have been used to fabricate superhydrophobic coatings. In a study, Schutzius, Bayer, Qin, Waldroup, and Megaridis (2013) prepared superhydrophobic coatings by mixing ammonium stabilized aqueous solution of exfoliated graphite nanoplatelets (2 nm thick, size $< 2\ \mu\text{m}$) with polyolefin, where the dispersion can be spray-coated onto aluminum, paper, and glass. After drying, the coated surface exhibits superhydrophobicity with a contact angle of 154° and contact angle hysteresis of 9° . The stability of the water-based dispersion is attributed to the electrostatic repulsion of charged particles. The prepared superhydrophobic surface seems to have good stability against mild abrasion and water jet impact, as well as mild chemical stability against acid/alkali solutions.

In another effort using electrostatic interactions for stabilizing hydrophobic particles in water, Lozhechnikova, Bellanger, Michen, Burger, and Österberg (2017) prepared carnauba wax-in-water dispersion (concentration 1 g/L $-10\ \text{g/L}$) by a sonicating mixture of water and carnauba wax at a temperature close to the melting point of carnauba wax. This heating was followed by rapid cooling in an ice bath and filtering through a fine filter (pore size $40\text{--}100\ \mu\text{m}$). The obtained dispersion is stable for at least three weeks, the stability of which was attributed to the large zeta potential ($-53\ \text{mV}$) that provides electrostatic double-layer repulsion and thus the stabilization. Commercially available ZnO nanoparticle dispersion was used to introduce nanoscale roughness, in addition to the positive surface charge of ZnO that can enhance binding with the negatively charged carnauba wax. The authors dip-coated wood by consecutively immersing it in the ZnO dispersion, and then the carnauba wax dispersion, followed by drying. This layer-by-layer process is repeated to obtain the most hydrophobic samples where the coated wood surface displays a water contact angle of 155° and a sliding angle of 24° . Armingier et al. (2019) used similar processes to prepare alkyl ketene dimer dispersion in water and obtained superhydrophobic wood surface by spray-coating. They showed that rapid cooling of the molten wax and water mixture is essential for obtaining a dispersion that is stable for one week.

The ideal materials and processes to obtain superhydrophobicity should resort to materials procured from renewable sources and involve

mild procedures. The work by [Morrissette et al. \(2018\)](#) used plant-based or commercially available materials to prepare water-based superhydrophobic coatings. They used carnauba wax or beeswax as a hydrophobic agent and lycopodium pores to bring about roughness, assured due to the hierarchical structure comprised of micro-scale pores (20 μm) and nanoscale polygons. The coatings are sprayable, and after application and drying, the surface displays a water contact angle of 160° and contact angle hysteresis of 7°. The coatings are not only made from plant-based materials but also require only moderate annealing to melt the waxes and cover the surface of lycopodium to impart superhydrophobicity.

3.2. Fabrication methods

3.2.1. Substrate independent superhydrophobic coatings

This type of coatings includes dispersions and pastes, which are generally fabricated by dispersing low surface energy molecules and nanoparticles in an organic solvent ([Celik, Kiremitler, et al., 2021](#); [Nguyen-Tri et al., 2019](#); [Park et al., 2020](#)). These types of coatings are easy to fabricate, inexpensive, and can be applied to almost any surface, thus having large commercial potential. Plant waxes have been an attractive low surface energy natural material. For example, [Wang et al. \(2016\)](#) prepared a sprayable superhydrophobic coating using US FDA approved, plant-derived carnauba wax and beeswax by dispersing them

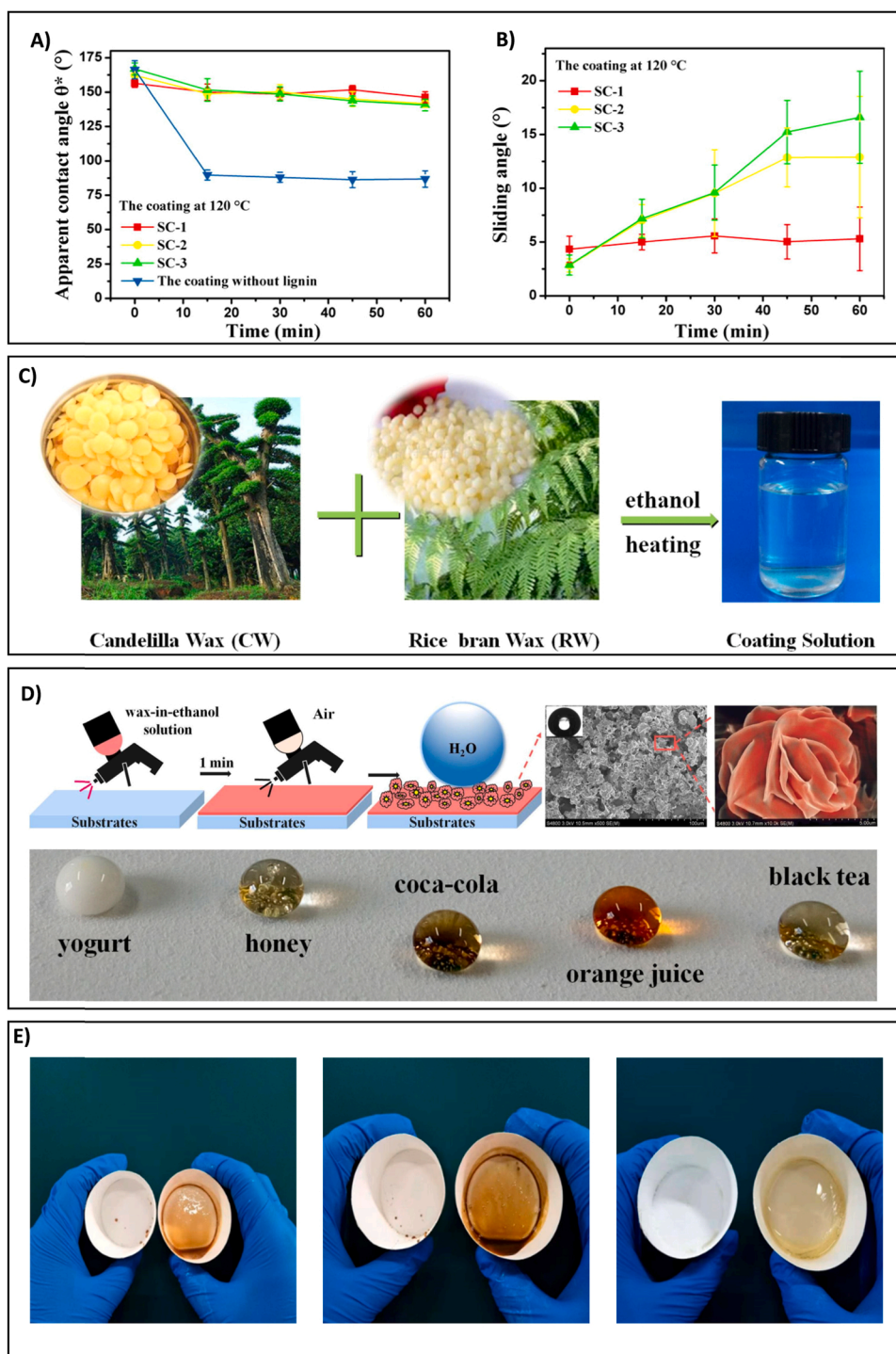


Fig. 2. Eco-friendly preparation of superhydrophobic coatings. A, B) Thermal durability of superhydrophobic coating made from coffee lignin extract and beeswax (lignin/wax ratio is 2, 1, 0.5 for SC-1, SC-2, and SC-3, respectively) measured as the change of A) contact angle and B) sliding angle. C) fabrication of superhydrophobic coating from candelilla wax and rice bran wax and D) illustration and demonstration of liquid food repellency. E) comparison of residual liquid food left on a superhydrophobic coated cup (left) and ordinary cup after emptying. The liquids are milk tea, chocolate syrup, and honey from left to right.

Sources: A) and B) are reprinted from [Zhang et al. \(2019\)](#) with permission from Elsevier. C) and D) are reprinted from [Liu et al. \(2019\)](#) with permission from Elsevier. E) is from [Shen et al. \(2020\)](#).

in acetone (concentration 20 mg / mL). Glass slides coated with the dispersion exhibited superhydrophobicity for liquids with surface tension higher than 45 mN / m. Furthermore, the authors demonstrated usage of the coating for reducing residual liquid food, where sticky liquids such as honey and syrup can be completely poured out from a coated polystyrene cup. Additionally, the toxicity and cell viability of the coatings was evaluated using 3T3-J2 murine embryonic fibroblasts where the results indicated that cell viability was not affected up to carnauba wax concentration of 500 mg / mL. In another study, Y. Zhang et al. prepared an edible superhydrophobic coating from beeswax and lignin extracted from coffee beans by dispersing them in a mixture of acetone and n-hexane (Zhang et al., 2019). The coating exhibited superhydrophobicity for several food liquids, such as milk, tea, cola, and so on. Besides, the coating does not negatively affect cell viability, as evaluated using mouse osteoblast cells. A unique aspect of the coating was its thermal stability up to 120 °C (up to 60 min of annealing), well above the stability of beeswax, which was attributed to the thermal stability enhancement of the lignin component (Fig. 2A & B). Besides carnauba and beeswax, even though rare, other plant-based waxes have also been used. For instance, B. Y. Liu et al. (Liu et al., 2019) used plant-derived candelilla wax and rice bran wax and prepared a superhydrophobic coating by dispersing the waxes in hot (85 °C) ethanol, which is an eco-friendly solvent that can also be made from plants (Fig. 2C & D). The authors spray-coated (density 0.49 mg / cm²) the hot ethanol dispersion on polypropylene (PP) substrate and the substrate showed superhydrophobicity with water CA of 156° and SA of 2° after drying of the solvent quickly. The fabrication process was illustrated in Fig. 2D. The superhydrophobic coating showed excellent repellency against common liquid foods. Furthermore, the coated PP substrate showed excellent durability against cyclic bending, retaining superhydrophobicity even after 1200 cycles of bending. In another study, Shen et al. prepared a superhydrophobic coating by dispersing soybean wax in ethanol (Shen, Fan, Li, Xu, & Fan, 2020). After coating, the surface exhibited water CA of 159° and SA of 7°. A paper cup with the coating can eliminate residual liquid foods such as syrup and honey (Fig. 2E). Furthermore, the coatings exhibited good durability, as evaluated using sandpaper abrasion test, and cyclic immersion and retrieval test in yogurt. In a relevant study, Cheng et al. prepared a biodegradable superhydrophobic coating using sustainable materials (Cheng, An, Li, Huang, & Zeng, 2017). Specifically, they treated surfaces with a water solution of epoxidized soybean and ZnO NPs (50 nm), followed by dip coating with stearic acid. The coating was stable in water for at least 7 days. Moreover, a filter paper treated with superhydrophobic coatings was used for the separation of oil from water with an efficiency of more than 97%.

An interesting choice of material is clay, a naturally abundant material. In a study, Razavi et al. (Razavi et al., 2019) used sepiolite clay and treated it with plant-derived and thus environmentally friendly material, such as cinnamic acid or myristic acid, to impart hydrophobicity to the naturally hydrophilic clay, as illustrated in Fig. 3A. Afterward, the hydrophobic clay was mixed with a commercial fluoroalkyl polymer solution (Capstone ST-100) to get the final dispersion (Fig. 3B). The authors demonstrated the versatility of the coating concerning the substrate where substrates such as aluminum, paper, and glass can be dip-coated with the dispersion, resulting in superhydrophobicity with water CA of 160° and CAH of 5°. Furthermore, the superhydrophobic coating demonstrated anti-biofouling ability, where the attachment of *Staphylococcus epidermidis* and *E. coli* bacteria were as low as 9% and 3%, respectively.

One important aspect of fabricating superhydrophobic coatings is to use eco-friendly, non-toxic, and cheap materials. Bayer and coworkers have done several important contributions to the field (Bayer, 2020). In one example (Naderizadeh et al., 2020), agricultural waste bio-resin (polyfurfuryl alcohol), along with fluoro acrylic copolymer (Capstone St-100) and hydrophobic SiO₂ NPs (7–40 nm) were used to fabricate an acetone dispersion that can be spray coated onto aluminum foil,

aluminum plate, and glass slides (Fig. 3C). The superhydrophobic coating was thermally stable up to 200 °C and showed good viability for human cell lines (HeLa cells) within 120 h of test duration. Furthermore, the coating showed good anti-adhesion performance against *E. coli*, *Staphylococcus*, and biofilm-forming bacteria *Pseudomonas aeruginosa*. In another study, D. Wang et al. prepared a superhydrophobic membrane using materials from tomato waste (pectin and cutin), followed by spray coating with beeswax dispersion in ethanol (Wang et al., 2020). The membrane showed good anti-fouling activity against several food liquids. Importantly, the superhydrophobic membrane showed a good barrier property against water and oxygen, enabling the usage of the membrane to wrap apple and delay browning (Fig. 3D and E).

An important step towards smart packaging is to use edible materials. In this regard, an edible superhydrophobic coating was prepared by J. Li et al. (Li et al., 2021) by the reaction of egg lysozyme with cysteine, which resulted in phase transitioned lysozyme (PTL) with rough nano/microscale texture, followed by spin coating with a hot hexane dispersion of carnauba wax. Besides spin coating, the superhydrophobic coating can be applied to almost any material using spray coating and dip coating (Fig. 4A). In another study, Y. Li et al. prepared an edible superhydrophobic film by casting dispersion of Arabic gum, gelatin, and glycerine in water, followed by drying and spray-coating with beeswax dispersion (Li et al., 2018). The edible film showed superhydrophobicity with water CA of 158° and SA of 7°. The film was flexible, able to withstand at least 50 cycles of folding. Notable, the coating showed no observable cytotoxicity towards mouse osteoblasts within 72 h period of testing. In addition, the coatings showed excellent repellency towards common food liquids (Fig. 4B).

3.2.2. Metallic substrates

Aluminum and stainless steel dominate the application of metallic materials in food packaging. Cans used in soft drinks, ready-to-cook soup, and tomato paste are the main food types that use metallic packaging materials. Besides, it should be noted that metals, especially stainless steel, are also widely used in food manufacturing processes, like cutters, mincing devices, and heat exchangers. Lithographic/laser patterning and electro/chemical etching are the two main approaches to fabricate textured surfaces on metals, which will become superhydrophobic upon treatment with hydrophobic materials (Gong et al., 2016; Long et al., 2015; Tripathy et al., 2017; Zheng et al., 2021). In a study, Pan et al. (Pan, Cao, Xue, Zhu, & Liu, 2019) fabricated superhydrophobic stainless steel using picosecond laser texturing, followed by treatment with a stearic acid solution, resulting in superhydrophobicity with a water CA of 163° and SA of 0.5°. Importantly, the superhydrophobic steel surface showed more than 90% adhesion resistance against *E. coli* and *S. aureus* bacteria. In another study, Barish and Goddard used a different approach. Here, the authors prepared anti-fouling stainless steel employing a Nickel plating technique (Barish & Goddard, 2013). They first cleaned the surface with alkali detergent at 85 °C, then with HCl acid (50%), followed by depositing a nickel phosphate layer. The prepared surface was hydrophobic with water CAH of 7.6° and able to reduce fouling by 97% in a heat exchanger (operated at 85 °C) used for raw milk processing. Employing a more convenient approach, Yoon et al. (Yoon, Rungraeng, Song, & Jun, 2014) prepared antibacterial superhydrophobic stainless steel by first spin coating with a dispersion of TiO₂ nanoparticles (20–30 nm), followed by spray coating with carbon nanotube solution, and again spin coating with a dispersion of PTFE (polytetrafluoroethylene) micro-particles. In the end, the coated multi-layer stainless steel was annealed at 360 °C for 1 h. The superhydrophobic stainless steel displayed water CA of 155° and exhibited good resistance against *E. coli* adhesion and biofilm formation under dynamic conditions, reducing bacterial adhesion by 80%.

Using a systematic approach, Liu et al. reported imparting superhydrophobicity to aluminum substrates employing several different approaches (Liu et al., 2020, 2021; Oh et al., 2019). In one study, they prepared a mechanically robust superhydrophobic surface that showed

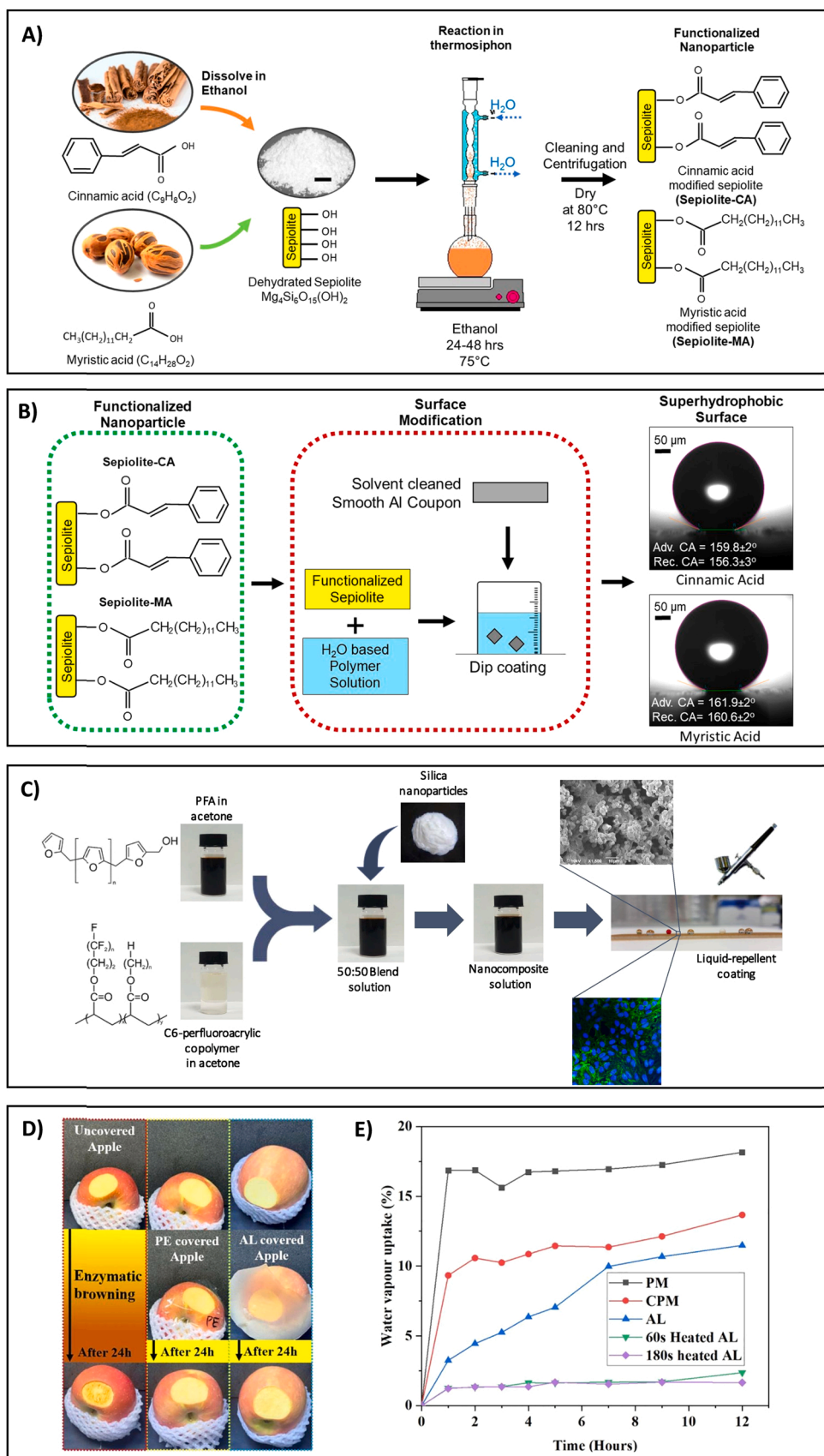


Fig. 3. Illustration of A) hydrophobic treatment of nanoparticles with natural products and B) fabrication of superhydrophobic surface via dip-coating, showing pictures of a water droplet on the coated surface. C) Illustration of the fabrication process of the superhydrophobic coating using natural bio-resin polyfurfuryl alcohol and commercial product, showing surface morphology and antibacterial activity via fluorescence imaging. D) Demonstration of enhanced oxygen barrier of the superhydrophobic artificial lotus leaf (AL) prepared from tomato leaf extract (cutin and pectin) and beeswax, showing suppression of the AL membrane of the enzymatic browning of cut apples. E) Water uptake characterization of the pectin membrane (PM), cutin-pectin membrane (CPM), the superhydrophobic AL surface, and after heated for 60 s and 120 s, respectively.

Sources: A and B) are adapted with permission from [Razavi et al. \(2019\)](#). Copyright 2019, American Chemical Society. C) is reprinted from [Naderizadeh et al. \(2020\)](#), with permission from Elsevier. D and E) are adapted from [Wang et al. \(2020\)](#) with permission from Elsevier.

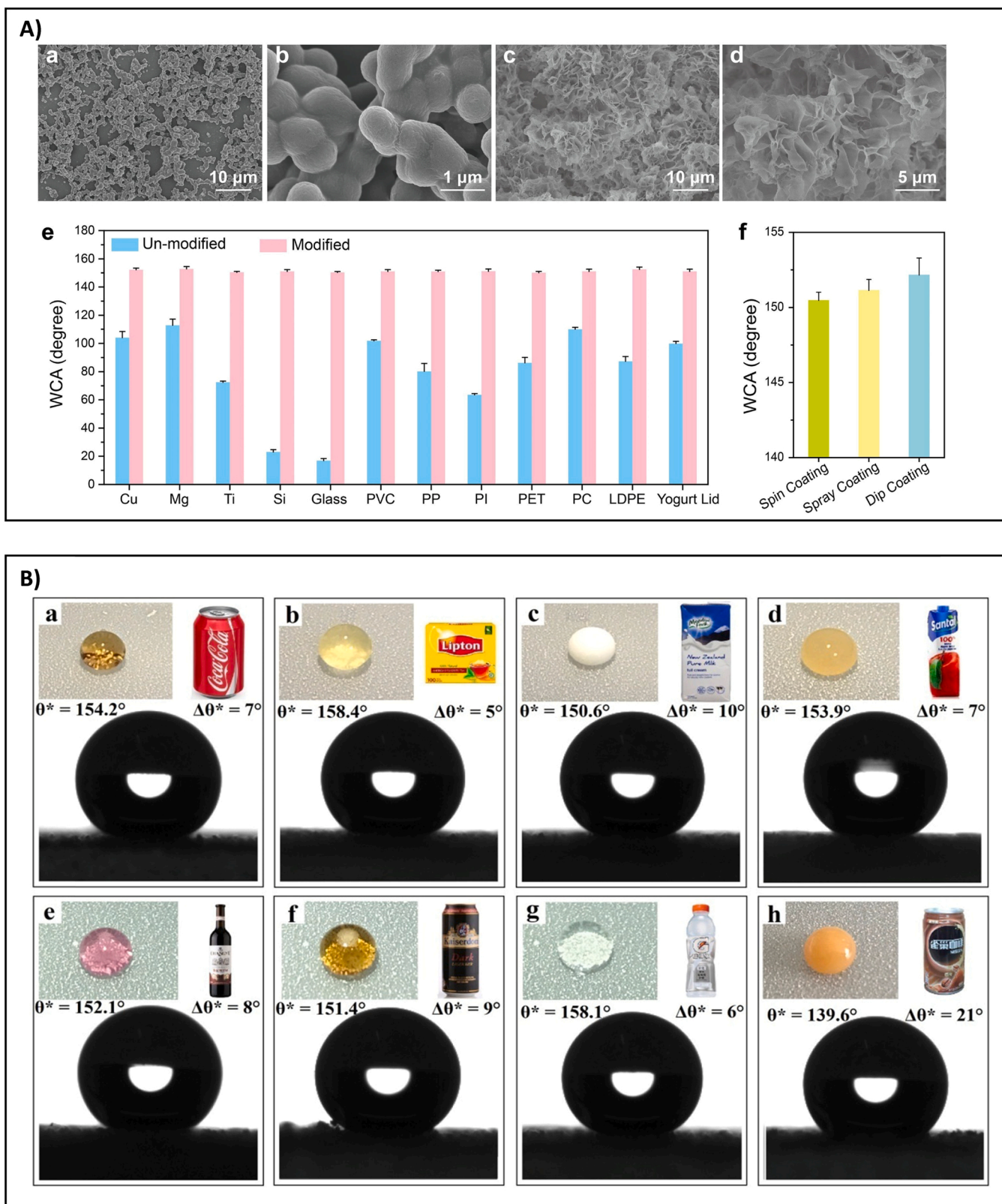


Fig. 4. Characterization of various edible superhydrophobic coatings. A: SEM images of the a-b) PTL coating and c-d) the superhydrophobic coating prepared by coating the PTL with carnauba wax. e) water contact angle (WCA) of various substrates before and after modification. f) Effect of various coating processes on WCA. B: a-h) Contact angle of various liquid food on the surface of the coated superhydrophobic surface. Sources: A is adapted from Li et al. (2021) with permission from Elsevier. B) is reprinted with permission from Li et al. (2018). Copyright 2018, American Chemical Society.

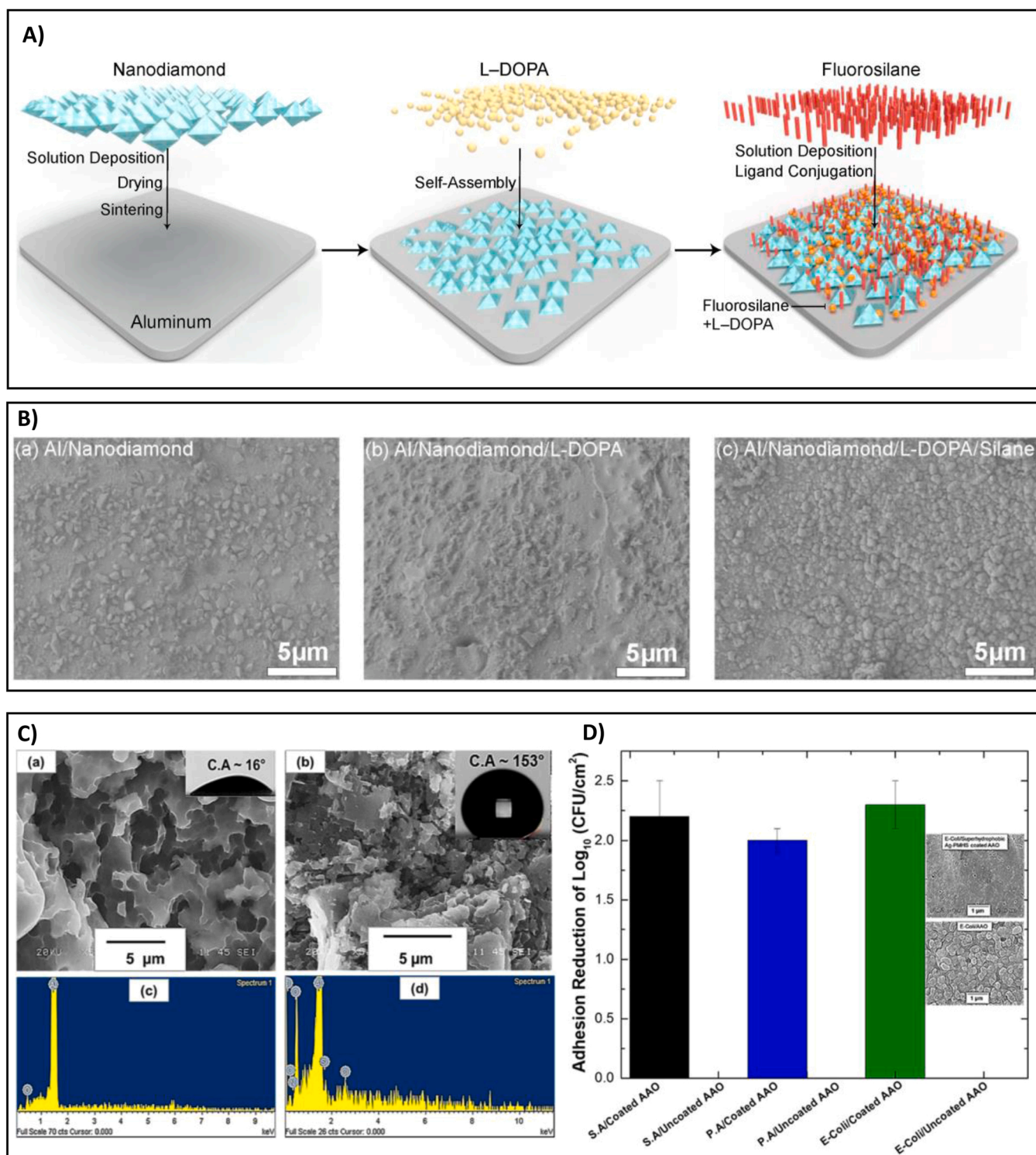


Fig. 5. Preparation and characterization of antibacterial superhydrophobic surfaces. A: Illustration of the fabrication process of nanodiamond enhanced superhydrophobic aluminum surface. B) SEM images of the corresponding surfaces. C) SEM images and EDX analysis of the etched aluminum and the superhydrophobic modified aluminum. D: Antibacterial efficacy of the Ag-PDMS coated and uncoated anodized aluminum oxide (AAO) surface against *Staphylococcus aureus* (S.A), *Pseudomonas aeruginosa* (P.A), and *E. coli* bacteria.

Sources: A and B are reprinted from Liu et al. (2021), with permission from Elsevier. C is from Agbe, Sarkar, and Chen (2020). D is reprinted with permission from Agbe, Sarkar, Chen, et al. (2020). Copyright 2020, American Chemical Society.

excellent resistance against *E. coli* and *S. aureus* bacteria, reducing bacterial attachment by > 99% when compared to bare aluminum (Liu et al., 2021). The robust durability of the superhydrophobic surface was attributed to the fabrication method, as illustrated in Fig. 5A & B. First, the authors deposited hard material, nanodiamond (0–500 nm), and

sintered at high temperatures (650 °C), achieving the infusion of the nanodiamond into the aluminum plate. Then, the nanodiamond-infused aluminum plate was dip-coated with an adhesive layer, namely L-dopamine (L-DOPA), followed by coating with a hydrophobic layer by immersing it in a trichlorofluoroalkyl silane solution. The anti-adhesion

superhydrophobic aluminum plate is mechanically durable, retaining superhydrophobicity and anti-adhesion property even after 10,000 cycles of scratching against a Nylon abrader under the load of 10 mN – 50 mN. Using a similar high-temperature sintering approach, the same authors imparted superhydrophobicity to aluminum via sequential deposition of SiO₂ nanoparticles (200–300 nm), lysozyme, and -fluorosilane (Liu et al., 2020).

The ease of reactivity of aluminum with acids and basis can be used to introduce roughness on an aluminum substrate, like the work done by Agbe et al. (Agbe, Sarkar, & Chen, 2020). Here, the authors fabricated a superhydrophobic antibacterial aluminum plate by etching aluminum in HCl and then immersing it in an ethanolic solution of octyltriethoxysilane, followed by drop-wise addition of quaternary ammonium solution. The etched aluminum exhibited roughness of 6.2 μm and CA of 16°. After drying the treated aluminum plate at 100 °C for 2 h, superhydrophobicity was achieved with a water CA of 153° (Fig. 5C). Additionally, the superhydrophobic aluminum plate showed more than 99% anti-biofouling activity against *Staphylococcus aureus*, *Pseudomonas aeruginosa*, and *E. coli* bacteria. The same authors (Agbe, Sarkar, Chen, et al., 2020) prepared a similar anti-biofouling superhydrophobic surface by anodizing aluminum followed by coating it with Ag-decorated polysiloxane (Fig. 5D). Here, the superhydrophobic coating decorated with Ag was able to reduce adhesion of both gram-positive (*Staphylococcus aureus*) and gram-negative (*Pseudomonas aeruginosa* and *E. coli*) bacteria by at least 99%.

3.2.3. Cellulose materials

This category of materials includes raw materials that are derived from plants, where the main constituent is cellulose, and modified and refined materials, such as crystalline nano cellulose and cellulose esters. The main usage of cellulose-based materials is in making paper, cardboard, and various wraps (Glenn et al., 2021). Due to the hydrophilic nature of these types of materials, they are very vulnerable to water and moisture, resulting in loss of strength and shape (Cataldi et al., 2019). Therefore, cellulose-based materials generally require hydrophobic/superhydrophobic treatment to increase water barrier property and stability against water immersion. Even though there are many reports of imparting superhydrophobicity to paper and related materials (Balu et al., 2008; Barona & Amirfazli, 2011; Esmaeili et al., 2020; Hu, Zen, Gong, & Deng, 2009; Koşak Söz et al., 2018; Long et al., 2022; Ogihara et al., 2013; Werner, Quan, Turner, Pettersson, & Wågberg, 2010; Zhang, Lu, Qian, & Xiao, 2014), only a handful of them investigated food packaging-related applications. These are our focus here. But before delving into that, we like to highlight a recent paper by Esmaeili et al. (Esmaeili et al., 2020) where they imparted superhydrophobicity to paper by using alky ketene dimer, a synthetic wax that is already widely in use in the paper industry. The process of using widely available materials, melting them at mild temperatures (40 °C), followed by dipping paper into the molten wax and etching with eco-friendly solvent ethanol, is especially suited and has great potential in being adopted by industry in large scale roll-to-roll fabrication.

In food packaging applications of superhydrophobic cellulose materials, Li et al. (Li et al., 2019) used a layer-by-layer coating approach: they first immersed cellulose fibers into an acetic acid solution of chitosan to create a positively charged surface, then sequentially immersed in carrageenan solution, chitosan solution, clay (Montmorillonite) dispersion, and again in chitosan solution, for 20 mins each, followed by making into sheets (80 g / m²). At the last step, the treated cellulose sheet was dip-coated with carnauba wax emulsion, followed by drying at 105 °C for 30 mins. The fabricated paper exhibited superhydrophobicity with water CA of 151.4°, and good air and moisture barrier property, while retaining similar tensile strength when compared to the original paper. Furthermore, a bag was fabricated from superhydrophobic paper and used to store strawberries, where the results indicate good moisture and acidic content preservation ability of the coatings. In another study, Zhao et al. (Zhao et al., 2020) prepared omniphobic paper using

commercial papers by first oxygen plasma treatment, followed by vapor phase deposition of 1,3-dichloro-tetramethyl disiloxane. It is believed that the siloxane is chemically grafted to the surface of the paper and displays a ‘polymer brush’ like structure, therefore exhibiting negligible resistance against the movement of liquids and small CAH (6°). The modified paper is omniphobic in that it is repellent towards even liquids with low surface tension, such as olive oil, hexadecane, and ethanol. The modified paper exhibited excellent water repellency, as well as high grease resistance and is thermally stable up to 200 °C. A different approach was taken by I. Torun and M. S. Onses (Torun & Onses, 2017) where they used a simple approach: they deposited hydrophobic silica nanoparticles onto paper, which after drying of the solvent, exhibited superhydrophobicity towards liquids with surface tension higher than 45 mN / m. The coated paper exhibited good durability when compared to coated flat surfaces, which the authors attributed to the rough structure of the paper where the cellulose microfibrils protect the nanoparticles. Furthermore, the authors demonstrated self-cleaning of the surface against ketchup and used the superhydrophobic paper to wrap chicken breast, which showed anti-freezing behavior (- 30 °C) as well as non-stickiness to the chicken meat.

4. Conclusion and perspectives

Since the discovery made by Wenzel some 70 years ago about the extreme repellency of surface-modified textiles against water, superhydrophobic surfaces have been used for various purposes such as self-cleaning, anti-fouling, and anti-corrosion. These same properties enable the application of superhydrophobic surfaces for food packaging for reducing residual food waste and minimizing cleaning and disinfecting costs of food production equipment and containers. Therefore, most if not all food packaging-related studies employing superhydrophobic surfaces and coatings are focused on reducing food waste, with some studies also investigating reducing bacterial adhesion capability of the superhydrophobic surfaces. The main reason for this focus is the convenience of demonstrating reducing food waste, which can be performed by either simply placing tested liquid food on a tilted superhydrophobic surface and showing complete removal or by emptying the food content of a superhydrophobic container. As such, most of these studies are qualitative. Furthermore, very few studies tested packaging-related properties such as strength, elasticity, heat resistance, air and humidity permeability, chemical and mechanical durability, and toxicity of the materials used.

In conclusion, the application of superhydrophobic coatings for food packaging is gaining attraction among scientists and engineers for their promising capabilities, resulting in the publication of a few dozen research articles in the last few years, with the focus on anti-fouling of food handling equipment and reducing residual food waste. However, to be applicable in real-life scenarios, further research needs to be done. One of the most serious issues of superhydrophobic coatings and surfaces is their weak durability. In practical applications, a superhydrophobic packaging material should have enough mechanical strength to hold the food and be able to withstand forces that may be exerted during transportation and handling. However, very few studies report the mechanical property (Young modulus, tensile strength, etc) of the superhydrophobic packaging materials. Once the superhydrophobic packaging material has enough mechanical strength, then the superhydrophobic layer must be strongly adhered to the underlying material and withstand mechanical abrasion, some folding and twisting forces. Yet measurement of adhesion property of superhydrophobic coatings is rare. At last, but not least, the evaluation of the interaction of food content with the superhydrophobic material under realistic conditions is rare. Here, the most important property is the safety of coating material which leaches into the food content. Any superhydrophobic coating with potential food packaging application should be subjected to detailed studies on the long-term leaching behavior and biocompatibility, not only under ideal conditions (pure water or solutions with various pH

values) but also using real food, which is typically mixtures of various natural and synthetic chemicals.

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