Shielding surfaces from viruses and bacteria with a multifunctional coating

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Abstract

The spread of viral and bacterial pathogens mediated by contact with surfaces is a leading cause of infection worldwide. COVID-19 as well as the continuous rise of deaths associated with antibiotic-resistant bacteria highlights the need to impede surface-mediated transmission. We report a sprayable coating with an intrinsic ability to resist the uptake of bacteria and viruses from surfaces and droplets, such as those generated by sneezing or coughing. Our coating also provides an effective microbicidal functionality against bacteria, providing a dual barrier against pathogen uptake and transmission. This antimicrobial functionality is fully preserved following scratching and other induced damage to its surface or 9 days of submersion in a highly concentrated suspension of bacteria.

Introduction

COVID-19 has highlighted the global risk associated with the transmission of pathogens, and its rapid and severe impact on human health and the global economy ¹⁻³. There is broad evidence that surfaces facilitate pathogen dissemination, leading to widespread transmission throughout the population ⁴. Surface contamination also plays a major role in the evolution of antibiotic-resistant bacterial strains ⁵⁻⁷. Alternatives to traditional surface disinfection approaches are necessary as these are presenting declining efficacy and are often laborious and ineffective as recontamination is triggered episodically ^{8,9}.

Alternative approaches to prevent bacterial contamination of surfaces relies on the use of bactericidal coating materials including, silver, copper, zinc and TiO₂ along with other inorganic nanoparticles and quantum dots ^{10,11}. Among these, silver nanoparticles are the most widely used bactericidal agent. Recent reports highlight their limitations, including the significant variations in potency between microbial species and harmful side effects due to the accumulation of high concentrations of silver ions within tissue ^{12,13}. Furthermore, bactericidal agents fail at protecting surfaces from contamination by viral pathogens.

Recently, the use of water repellent coatings that can be deployed on high touch surfaces, such as hospital infrastructure, has shown promising efficacy in preventing bacterial contamination ¹⁴. Bacterial contamination of surfaces starts with the adhesion of few planktonic cells ¹⁵. Engineering of water repellent surfaces can prevent this initial step by the formation of an air layer between the contaminated liquid and solid surface. This provides a steep mechanistic barrier to bacterial adhesion and subsequent biofilm formation ¹⁶⁻¹⁹.

Inspired by these properties, recent studies (**Table S1**) have begun to investigate bacterial colonization of water repellent surfaces²⁰. While an initial reduction of bacterial growth was observed on such surfaces, to date there is a lack of direct evidence on the mechanism preventing bacterial adhesion and growth. Furthermore, the observed bacteria inhibition is short-lived with bacteria colony densities converging toward those on non-water repellent surfaces within 4-6 h of immersion in a bacterial suspension. Water repellent coatings also often provide insufficient robustness for real-world applications, due to the intrinsic fragility of the nanoscale roughness required for attaining a Cassie-Baxter water repellent state²¹.

Here, we report a multifunctional coating able to shield surfaces from both viral and bacterial pathogens. We gain the first direct insights on the ability of water repellent surfaces to prevent bacterial adhesion and on the eventual failure of this shielding mechanism, amid prolonged immersion in contaminated liquids or due to physical damage to the coatings. We use these learnings to engineer a coating that provides enhanced and prolonged resistance to pathogen contamination via a dual surface shielding mechanism. Our coating features both an enhanced and robust water repellency, based on a multi-scale roughness, and a highly effective localized microbicidal action triggered by a stimuli-responsive ion release mechanism. This coating structure can provide 99.94% and 99.85% initial reduction in, gram-positive and gram-negative, bacterial and an immediate 11-fold decreased in viral pathogen contamination. Notably, it also retains its antimicrobial performance following either significant coating damage or extended submersion in a bacterial suspension for more than 9 days. This is more than 22 times longer than the best performing water repellent coating reported. Our coating shielding mechanism is effective in both dry and wet environments, providing means to inhibit contamination of hospital infrastructure, air and water filters and others surfaces at high risk of mediating pathogen transmission.

Results And Discussion

Engineering a dual functional coating for shielding surfaces from bacteria and viruses

Our coating consists of two nanostructured components providing a pathogen repellent and microbicidal function, respectively (**Fig 1, SI Note 1 and 2**). Contamination by pathogen contained in liquid environments, such as respiratory droplets, is prevented by a water repellent superhydrophobic layer of fluorinated silica nanoparticles, robustly anchored on an underlying interpenetrated polymer network (IPN) film (see Materials and Methods and Note 3 in SI).

Effective microbicidal action is imparted by the incorporation of nanoscale grains of Zeolitic Imidazolate Framework-8 (ZIF-8) that act as a localized stimuli-responsive source of Zn ions (**Fig 1b, SI Note 2**). ZIF-8 is a zinc-based metal organic framework (MOF), resembling the zeolite topology, with 2-methylimidazole (HmIm) as the linker²². The absence of hydrophilic functional groups in the HmIm makes ZIF-8

inherently hydrophobic, contributing to the coating superhydrophobicity²³. The spontaneous release of Zn^{2+} ions from ZIF-8, due to its gradual decomposition in a broad range of pH values bears promise for its use as an antimicrobial agent^{24,25}. Here, the presence of ZIF-8 is aimed at killing bacteria that may adhere to the coating due to damage to its structural integrity, and thus localized loss of superhydrophobicity, evaporation of their liquid shell environment before removal from the surface, or loss of the air layer due to prolonged immersion in a contaminated liquid (**Fig 1b**). For further discussion on the surface protection mechanism please see **Note 5 in SI**.

Protecting surfaces from viral contamination

We first explored the performance of our water repellent surface structure in avoiding viral contamination. We used a steel rod with significant surface roughness (**Fig S15**) and tested its ability to mediate transmission of viruses in its uncoated state, or coated with our water repellent coatings, with or without ZIF-8 nanoparticles (referred to as water repellent and dual functional coatings, respectively; **SI Note 4, Fig S15**). For each test, the rod was dipped into a high titre solution of an EGFP-expressing lentiviral vector as a surrogate for infectious virus. The rod was dipped in a series of inert washing solutions (fresh Dulbecco's Modified Eagle's Medium (DMEM)) to determine the level of initial contamination and ease of cleaning (**Fig 2a, Fig S15**). Virus was quantified in the washing solutions by incubation on a monolayer of susceptible cells followed by fluorescence microscopy to detect and count infected cells (**Fig 2b**).

The first washing solution represents the capacity of the rod to resist viral contamination upon exposure to a high concentration lentiviral (i.e. human immunodeficiency virus) solution (1×10^7 TU/mL). Analysis of the first washing solution reveals a significant capability of the coatings to prevent viral contamination of the rod ($p=0.05$, **Fig 2c**). More than an 11-fold decrease in cell infection was observed from the bare rod (9.4% ($\pm 0.7\%$)) to the ones coated with a water repellent surface (0.8% ($\pm 0.3\%$)). The dual functional coating was also effective in preventing viral contamination, with a roughly 3-fold decrease in cell infection compared with bare steel. Remarkably, no virus was detected upon the first wash on the rod coated with the water repellent coating and only a very small amount of virus was detected for the dual functional coating, that was eliminated after the 2nd wash. The reduction in virus between the first two steps (contaminated surface to wash 1) for the dual functional coating was *ca.* 11-fold, after which detection of virus became sporadic. By contrast, the bare steel carried virus through all four wash steps showing a linear decline, with infectivity reducing by around a half with each successive wash. More details of the specific coating performance are provided in Note 4 in SI.

Bacterial Pathogen Shielding Performance and Mechanism

We used bacteria to evaluate the shielding performance against larger and heavier pathogens than virus, and to gain direct insights into the shielding kinetics of our coatings. **Fig S16** shows a schematic representation of the initial interface between bacteria suspended in a liquid and the coating surface, and its time evolution. A continuous air film, denoted as plastron layer, is initially formed at the solid-liquid interface preventing contact between bacteria and the coating surface. Over time parts of the plastron layer are removed via the formation and release of air bubbles, induced by the liquid pressure above the coating. These newly immersed coating surface becomes available for bacteria adhesion and eventually result in the formation of a biofilm. To investigate the specific shielding kinetics of our multi-scale rough coating, a droplet of bacterial suspension, in its exponential growth phase, was placed and kept on the coating surface and on other non-coated materials (bare steel) used as a comparison (**Materials and Methods in SI, Fig S 4(a)**).

Fig 3(a,b) shows in-operando in-situ confocal microscope images of a green fluorescent, bacteria droplet suspension on a bare steel plate and a steel plate coated with the dual functional coating. On the bare steel surface, the bacteria immediately settle and start the surface adhesion process (**Fig 3(b)**). In stark contrast, on our coating the bacteria suspension keeps floating above the plastron layer (white horizontal line), without being able to reach the underlying surface²⁶.

To gain further insights on the surface colonization mechanism, a droplet of bacterial suspension was kept on a bare and a coated steel surface and the cell behaviour was tracked with a confocal laser scanning microscope (CLSM) (**Movie 1-2, Materials and Methods in SI, Fig S4(a)**). Bacteria possess hair-like structures on their cell wall, denoted as flagella, which aids them in exploring microscale topography and facilitates adhesion to a surface. On the bare steel surface, the bacteria displayed nanoscopic vibrations around an equilibrium position (**Movie 1&2**). This is characteristic of a surface-bound motility behaviour referred as swarming, which is usually observed upon bacterial adhesion to a surface²⁶. More information regarding mobility and subsequent characterisation is described in Materials and Methods in SI.

Fig 3(c-e) and **Fig 3(g-i)** show a time sequence of 15 min of CLSM tracking (**Fig S4(a), Movie 1-2**) highlighting the dynamic response of the bacteria droplet suspensions on the bare steel and dual functional coating. Adhered bacteria were identified by lack of displacement and are false coloured in yellow. The average motion of the bacteria on both the surfaces were quantified by their average displacement between each time point (**Fig 3(f, j)**). Notably, bacteria cells on the bare steel surface had significantly ($p < 0.001$) lower average velocity of $4.09 (\pm 1.42) \mu\text{m}/\text{min}$ than those on the water repellent surface ($9.66 (\pm 1.43) \mu\text{m}/\text{min}$).

Biofilm formation on an abiotic surface, such as the bare steel here, starts with a reversible primary adhesion stage followed by an irreversible secondary adhesion stage. The primary stage is greatly influenced by hydrophobic interactions between the cell and the surface²⁷. Our in-situ analysis (**Movie 1**) indicates that several bacterial cells on the bare steel surface have already entered the secondary stage, showing no translational movement, and firmly adhering to the surface. In contrast, on the water repellent surface, no bacteria cells lacking motion could be identified, indicating prevention of the primary adhesion stage.

To attain a quantitative assessment of the effectiveness of our surface structure in preventing bacterial adhesion, bare and coated steel substrates were immersed in a bacterial culture and stamped on agar plates (**Fig S5**), with or without a prior washing step in Phosphate Buffered Saline (PBS). **Fig 4(a,c)** shows axio-observer microscopy of the agar plates stamped with the washed and the unwashed bare steel substrates. Notably, both samples resulted in the formation of a square-shaped layer of green fluorescing bacterial colonies, too densely packed to be counted. In stark contrast, the agar plates stamped with the un-washed water repellent coatings show only $12 (\pm 5)$ colonies (**Fig 4(b)**). Stamping the water repellent coatings after washing resulted in very few bacterial colonies, in the range of $2 (\pm 2)$ (**Fig 4(d)**). We attribute the small number of bacterial colonies, transferred from the un-washed coatings to the agar plate, to loosely adhered bacteria micro/nanodroplets. This is confirmed by the significant decrease in bacteria colonies upon a gentle washing step (**Fig 4(d)**), indicating the absence of biofilm formation (more discussion in Materials and Methods in SI).

To further quantify the efficacy in preventing bacterial adhesion, serial dilution of the residual PBS used to wash the samples after exposure to bacteria in their log growth phase was undertaken (**Fig S5**). **Fig 4(e)** shows that the bare steel samples resulted in $2.1 \times 10^5 (\pm 2.1 \times 10^4)$ and $1.4 \times 10^5 (\pm 3.1 \times 10^4)$ CFUs/mL colonies for gram-negative and gram-positive bacteria strains, respectively. Our coating successfully prevented up to 99.85% and 99.94 % of the gram-negative and gram-positive bacteria strain growth (significantly different with a p-value of less than 0.01 in a one-way ANOVA test) with only $297 (\pm 56)$ and $89 (\pm 24)$ CFUs/mL, respectively. To validate the essential role of the water repellent texture, control samples consisting of the same polymer coating without the fluorosilica layer were also investigated. The latter resulted in slightly higher CFUs/mL values than the bare steel surface, confirming that the polymer layer does not contribute to a decrease in bacterial adhesion.

Role of micro-scale coating defects on pathogen adhesion

To determine the role of micro-scale coating defect sites on pathogen adhesion, the water repellent fluorosilica layer was removed in a quasi-elliptic area of $62.8 \mu\text{m}^2$, exposing the underlying steel surface. A droplet of bacterial suspension was kept over the defective region of the coating and its intact water repellent surrounding (**Fig S4(a)**). The interface between the microscale defect and the water repellent region was continuously in-situ imaged by CLSM. **Fig 5(a)** shows an orthogonal projection of the droplet-substrate region. Over the intact water repellent surface (region A), a plastron layer with an above floating cloud of bacteria is observed. The plastron layer disappears on the defective coating area. The latter area consists of two parts (B,C), corresponding to the border with the intact region and the bulk of the defected area, respectively.

Bacteria over regions A and B exhibit a swimming behaviour whereas in region C, bacteria encounter the surface, due to the lack of the plastron layer. **Movies 3 & 4** show the time-lapse view from the image plane 1 and 2 in **Fig 5(a)**. **Fig 5(b,c)** shows static top view images taken after 14 min on these areas. Analysis of the velocity profiles in regions A and B (**Fig 5(d)**) confirm that the bacteria float over the intact surface and over the interface with the defected area with average velocities between $10.9 (\pm 1.6)$ and $11.5 (\pm 2.3) \mu\text{m}/\text{min}$, respectively. Whereas C has much smaller average values of $4.0 (\pm 1.6)$, in line with the analysis of the bare metal and water repellent surfaces above (**Fig 3(i, m)**). We also performed SEM analysis of such a defected area challenged with a bacterial culture for 10 min (**Fig 5(e-g)**). Bacteria began to colonise the defective area of the sample (**Fig 5(f)**), whereas there was no evidence of bacteria adhered to the neighbouring non-defective regions of the sample (**Fig 5(g)**). These results show that any microscale defects can act as a colonization site for bacteria. This may explain the observed few isolated colonies of bacteria observed on the agar plates stamped with the unwashed water repellent surfaces (**Fig 4(b)**).

Failure kinetic of the water repellent shielding mechanism

In addition to the presence of defects on the coating, the long-term stability of the plastron layer is a key factor to prevent bacteria and other pathogen adhesion to the surface. Previous studies report that the plastron layer on super-hydrophobic surfaces is lost within 1- 1.5 h of continuous immersion in water (**Table S1**). Here, to investigate the stability of the plastron layer on our surface structure, coated steel substrates were immersed in a liquid bacterial culture at a depth of 2.5 cm, and the water-surface interface was continuously imaged by CLSM over a period of 8 h (**Fig S4**).

Fig 6(a-h) show images with green fluorescent bacteria on agar plates stamped with the water repellent steel substrates as a function of their immersion time from 1 to 8 h. Notably, for up to 4 h of continuous immersion very few bacterial colonies are able to adhere to the substrate (**Fig 6(a-b)**). For exposures of 4 to 6 h, a rapidly increasing number of bacteria colonies was observed (**Fig 6(c-e)**). While longer time

results in complete colonization of the bacteria-challenged area (**Fig 6(f-h)**). For the full characterization of the failure kinetics of the water repellent shielding mechanism please refer to **Note 5 in SI**.

To gain insights on the mechanism determining the longer stability of the plastron layer on our water repellent surfaces, in situ studies were conducted by immersing our coating in a column of deionized water coloured with rhodamine B (**Fig 6(j-m)**, **Fig S4(b)**, **Movie 5**). The collapsing of the plastron layer was captured by the formation of a series of sequential contact point with the surface structure. This process rapidly accelerates after 6 h where the formation of bubbles supported by the rough surface features is observed. Subsequently, the bubbles are released resulting in the wetting of the surface as highlighted by the dye accumulation and the correlated increase in fluorescence intensity (**Fig 6(l, m)**). This lifetime of the plastron layer is attributed to the multiscale surface roughness of our coatings (see Note 5 in SI for more characterisation and discussion)

Secondary surface shielding mechanism

Our coating provides protection against both virus and bacteria until failure of the plastron layer. However, despite providing a longer stability than previous reports on water-repellent surfaces (> 4 h of continuous submersion), we acknowledged that the eventual dissolution of the plastron layer limits the breath of applications, for instance, in marine environments, membrane systems, or bathrooms, where an antibiofouling surface that could tolerate extended submersion is required. To this end, we have exploited the antimicrobial and hydrophobic properties of ZIF-8 nanograins (**Fig S11**) to impart a secondary shielding mechanism to the water repellent coatings. For material property optimization and wetting performance of the dual functional coating with ZIF-8 please see **Note 2 in SI**.

The capability of this surface composition to eliminate bacteria proliferation even in the event of dissolution of the plastron layer was demonstrated by long-term immersion of coated and bare steel substrates in a 5 cm column of solution containing 10^4 - 10^5 bacteria cells/mL (**Fig 6(n)**). After 1 day of immersion, there was no statistical difference between the bare and coated substrates. However, after 5 days of immersion, all the ZIF-8 containing coatings revealed a significant reduction in the cell counts of 2-3 orders of magnitude. More specifically, while on the bare substrates the bacteria colonies increased from 6×10^3 ($\pm 7.5 \times 10^2$) to 2.1×10^5 ($\pm 7.5 \times 10^4$) with immersion time increasing from 1 to 5 days, on the dual functional coatings the bacteria colonies decreased from 6.1×10^3 ($\pm 2.6 \times 10^2$), 5.4×10^3 ($\pm 3.7 \times 10^2$), 6.7×10^3 ($\pm 8.2 \times 10^2$) to 1.8×10^3 ($\pm 8.4 \times 10^2$), 6.9×10^2 ($\pm 2.1 \times 10^2$), 1.1×10^3 ($\pm 2.6 \times 10^2$) in the same time interval for the 5, 10 and 15 wt% ZIF-8 content, respectively.

The antimicrobial effect of the ZIF-8 became more accentuated with increasing immersion time with bacteria colonies further decreasing on the dual functional coatings after 9 days of continuous immersion. At such prolonged interaction between bacteria and coated surfaces, the impact of the ZIF-8 content became measurable, with the 15 wt% ZIF-8 containing coatings showing the smallest number of colonies of 3×10^1 ($\pm 1.8 \times 10^1$) versus the 1.6×10^2 ($\pm 6.9 \times 10^1$) of the 5 wt% ZIF-8 containing coatings and the 5.9×10^5 ($\pm 2.8 \times 10^5$) of the bare surfaces.

We further evaluated the efficacy of this antimicrobial mechanism to prevent bacteria adhesion in case of damage to the coating and subsequent loss of its water repellent functionality. A scratch was imparted on the pure fluorosilica water repellent coatings and the dual functional coatings with 15 wt% ZIF-8. A droplet containing bacteria culture in log phase was left on the defect (**Fig S7**), upon which the samples were washed with PBS and then incubated for 24 h. Subsequently the samples were stamped on agar plates, then incubated and analysed by microscopy (**Fig 6(o-r)**). The bare steel samples, used as controls, shows significant bacteria adhesion (**Fig 6(o)**). The non-scratched water repellent coatings had no bacterial adhesion (**Fig 6(p)**) in line with the plastron layer-based bacterial repulsion mechanism. The scratched water repellent coatings made of pure fluorosilica particles resulted in bacteria adhesion in proximity of the scratched region (**Fig 5(q)**). Notably, the scratched dual functional coatings containing ZIF-8 nanoparticles resulted in no bacteria adhesion despite the loss of their water repellent functionality (**Fig 6(r)**).

Conclusions

Realizing surfaces that are inherently able to resist pathogen contamination is central to healthcare strategies aimed at preventing pandemics. We introduced a dual functional coating capable of both shielding the surface from virus and bacteria. Our dual functional coating decreased the adhesion of bacteria by 99.8%, even when continuously immersed for 9 days in highly concentrated bacteria solutions. Importantly, the water repellent mechanism was also functional against virus, decreasing uptake by 11 times. Complete removal of the virus only required a gentle, single wash, while it was not possible to completely remove the same viruses from an uncoated steel surface even after 4 washes. The ease of fabrication of our coating by a rapid spray process and its compatibility with most materials makes it suitable for preventing pathogen contamination of a broad range of high-touch surfaces in public settings.

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Declarations

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Competing interest's statement

D.R.N and A.T. have patent coverage of the work presented.

Figures

Figure 1

Illustration showing the applications and mechanism of the dual functional coating for shielding potential high contamination surfaces, both in dry and wet environment from bacteria and viruses (a) Mechanism of the dual functional coating preventing surface contamination by microbes contained in liquid environment , (b) Mechanism of the localized loss of super-hydrophobicity of the dual functional coating and antimicrobial effect of ZIF-8 nanoparticles

Figure 2

Evaluation of viral pathogen shielding performance (a) Schematic of the experiment, where a metal rod is first dipped in a solution of an EGFP-expressing lentiviral vector (1×10^7 TU/mL) and then through a series of wash solutions (fresh DMEM). The washes were then tested for virus by incubating with cells and detecting infection by fluorescence microscopy. (b) Representative images of cells after incubation with the washes as labelled. Green indicates infection, blue is DAPI stained nuclei, scale bar= 100 μ m. (c) Quantification of numbers of infected cells as counted by microscopy, representing levels of virus contamination. Each replicate is shown as a dot ($n=9$), the bars and error are mean and SEM, respectively; *, $p < 0.01$.

Figure 3

Surface shielding mechanism. (a-b) CLSM orthogonal projection of bacteria on steel and water repellent surfaces, respectively. On the water repellent coating the plastron layer prevents bacteria from reaching and adhering to the surface. (f) Plot showing the velocity profile of bacteria on the bare steel surface and (c-e) screenshots from Movie 1 showing the behaviour of bacteria on this surface. Adhering bacteria are false coloured to yellow. (g-i) Screenshots showing the behaviour of bacteria on the water repellent coating and (j) plot showing the velocity profile of bacteria on these surfaces.

Figure 4

Surface shielding performance against bacteria (a-d) Axio-observer images of the agar imprints of control-steel and water repellent surfaces showing qualitatively the relative extent of adhesion under unwashed and washing conditions. (e) Plot showing the serial dilution data. The water repellent surfaces resisted 99.99% of bacteria compared to the steel surface. (* represents $p < 0.01$ in a one-way ANOVA test, $n = 9$).

Figure 5

Bacterial adhesion on point defects (a) Orthogonal projection of the coating-defect interface with bacteria on it. The two image planes are marked. The behaviour of cells over time on these planes are shown in Movies 3 & 4. (b-c) Screenshots of the coating-defect interface captured from Movies 3 & 4. A, B, C represents three regions where the cell-surface interactions are studied. (d) Velocity profiles of bacteria on regions A, B and C. (e-g) SEM images of a defective coating after exposure to bacterial culture for 10

minutes; (f) the defect is colonised by bacteria, whereas (g) the non-defective area has no bacteria attached to it.

Figure 6

Failure kinetic of water-repellent shielding mechanism and performance of the secondary shielding mechanism (a-h) Axio-observer images of the agar plate imprints corresponding to water repellent surfaces submersed in bacteria solutions for 1 hour to 8 h. Up to < 6 h immersion, a decrease in colonization with respect to the bare steel coating is observed. (i) Serial dilution data of the long

immersion experiment. (j-m) Screenshots from Movie 5 showing the plastron layer dissolution. (n) Plot showing the serial dilution data of the dual functional coatings containing ZIF-8 nanograins after 1, 5, and 9 days continuous immersion in a highle concentrated bacteria solution (** represents $p < 0.01$ and *** represents $p < 0.001$ in a one-way ANOVA test, $n=6$). Axio-observer images of the agar imprints; (o) control-steel, (p) pristine water repellent surfaces, (q) scratched water repellent surfaces, and (r) scratched water repellent surfaces containing 15 wt% of ZIF-8

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