Supporting Information

Durable Surfaces from Film-forming Silver Assemblies for Long-term Zero Bacterial Adhesion without Toxicity

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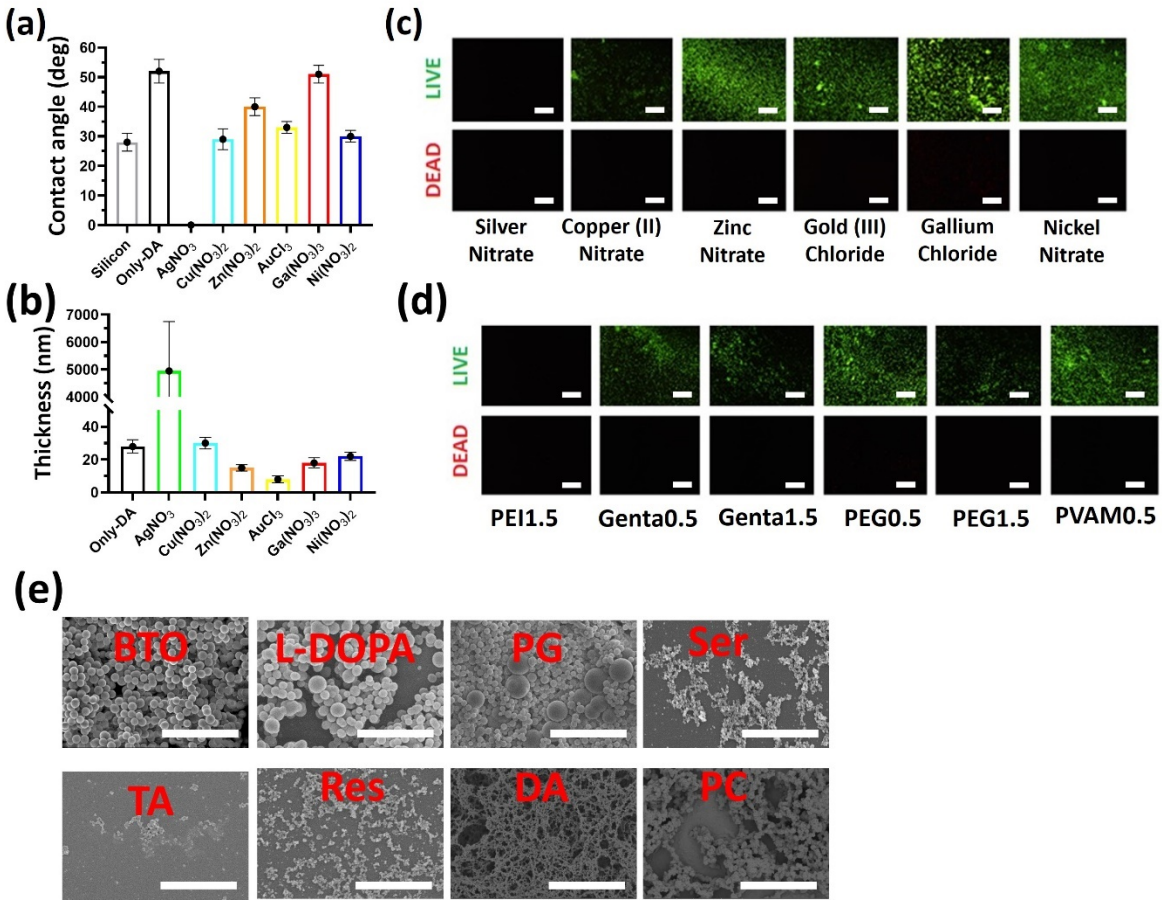
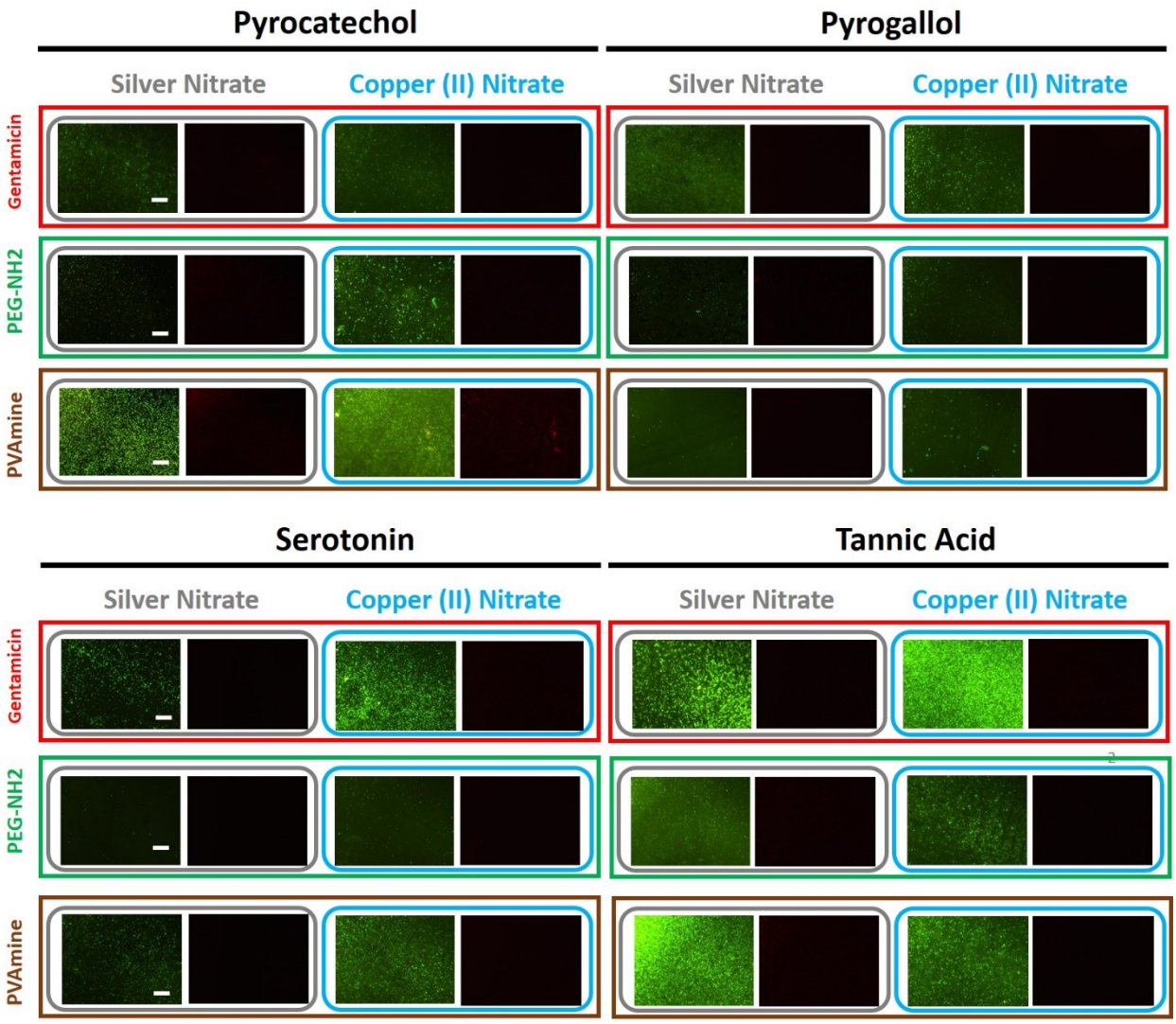
This file includes 21 figures and 2 tables

Supporting Figures



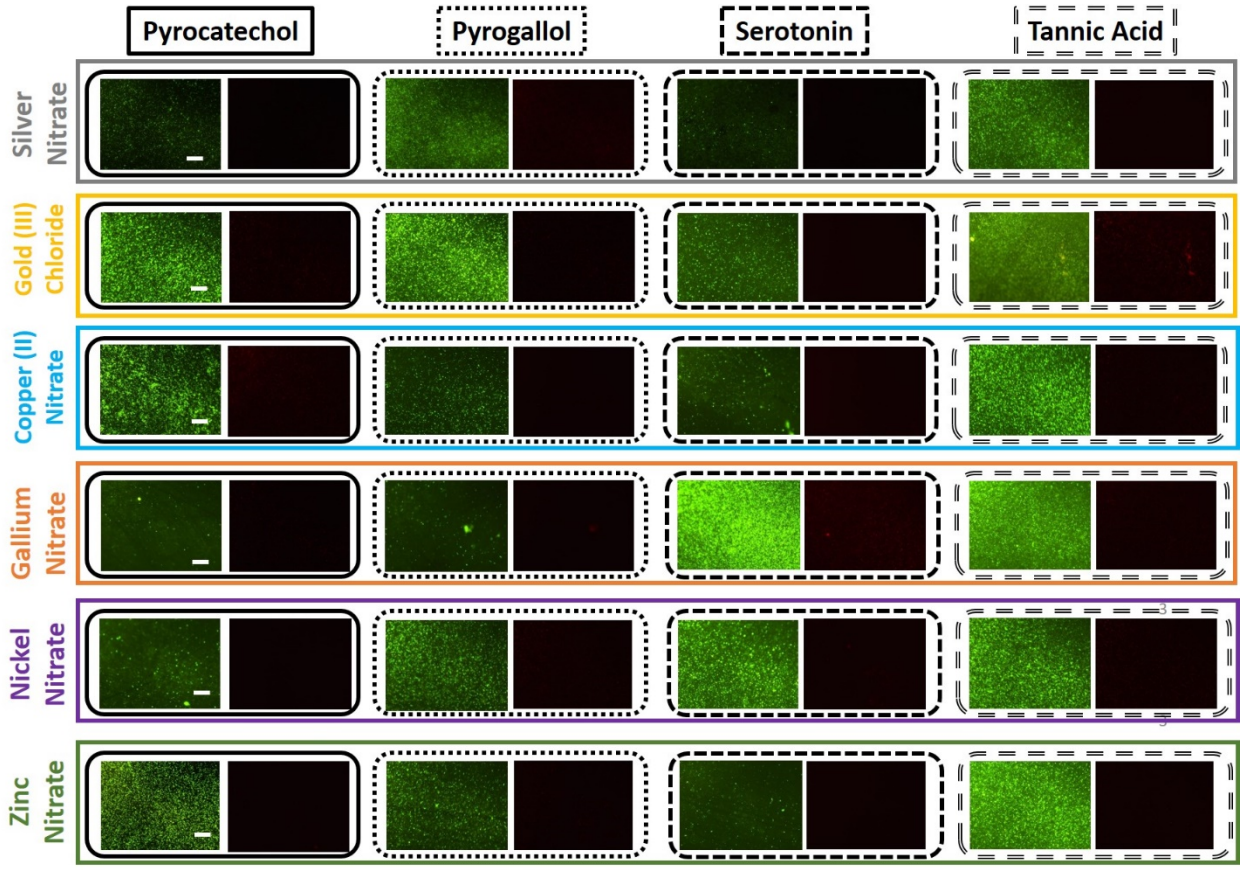
Supporting Figure 1. (a) Water contact angle image and **(b)** dry thickness of coatings based on compositions containing DA (2 mg/mL), L-PEI (1.5 mg/mL) and different salts (0.5 mg/ mL). **(c)** Fluorescence images of biofilm formation (green–live bacteria, red–dead bacteria) on the surface of silicon wafers treated with different three-component compositions containing DA (2 mg/mL), L-PEI (2 mg/mL) and metal salt (0.5 mg/mL) after 24 h incubation with *E. coli* (1×10^6 CFU/mL, LB). **(d)** Fluorescence images of biofilm formation (green–live bacteria, red–dead bacteria) on the surface of silicon wafers treated with different three-component compositions containing DA (2 mg/mL), cross-linking agent (1.5 mg/mL) and silver nitrate (0.5 mg/mL) after 7 days incubation with *E. coli* (1×10^6 CFU/mL, LB). The white scale bar is 100 μm. **(e)** SEM images of coatings

based on three-component compositions containing silver nitrate (0.5 mg/mL), L-PEI (1.5 mg/mL) and a catechol binder including: BTO, L-DOPA, PG, Ser, TA, Res, DA and PC. The Scale bar is 4 μm.

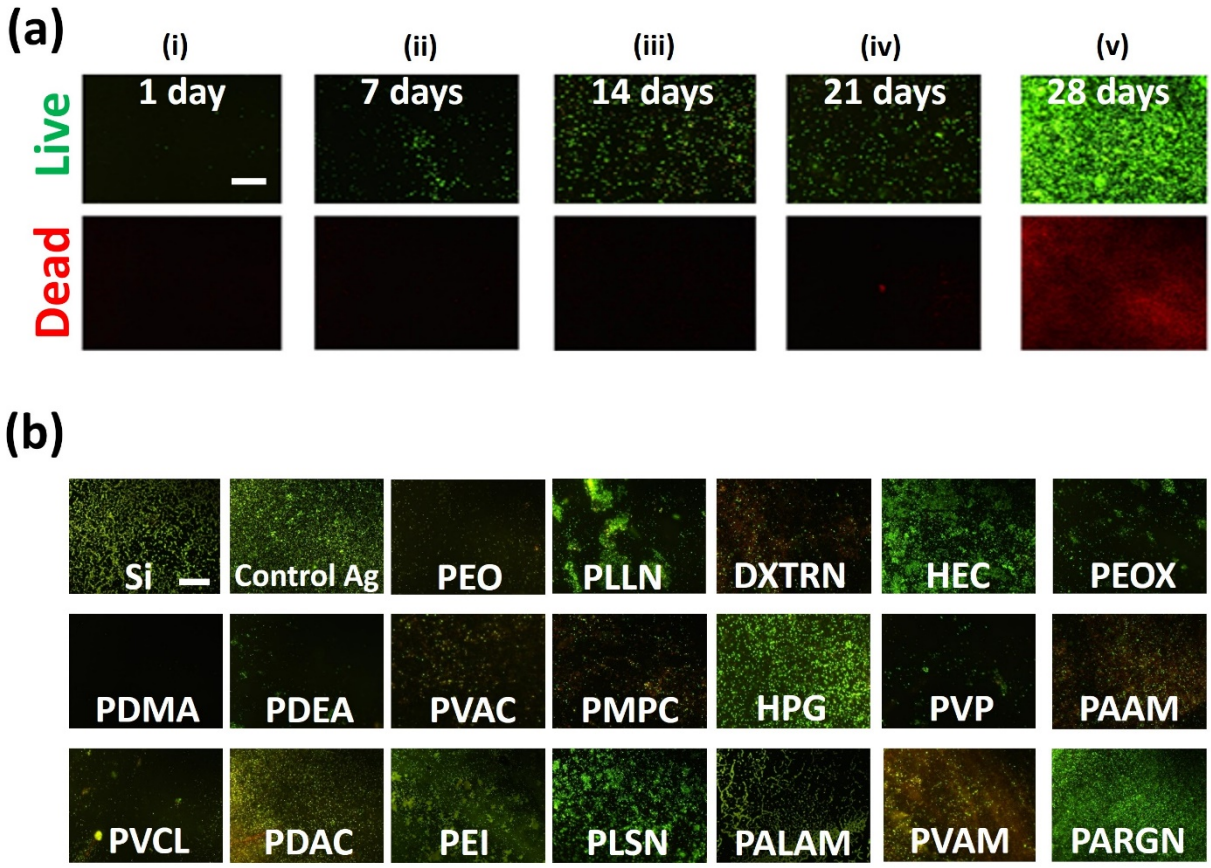


Supporting Figure 2. Fluorescence images of biofilm formed by *E. coli* (initial concentration: 1×10^6 CFU/mL, LB, 24 h) (green–live bacteria, red–dead bacteria) on the surface of silicon wafers treated with diverse compositions containing different catechol binders (pyrocatechol, pyrogallol, serotonin and tannic acid, 2 mg/mL), metal salts (silver nitrate and copper (II) nitrate, 0.5 mg/mL)

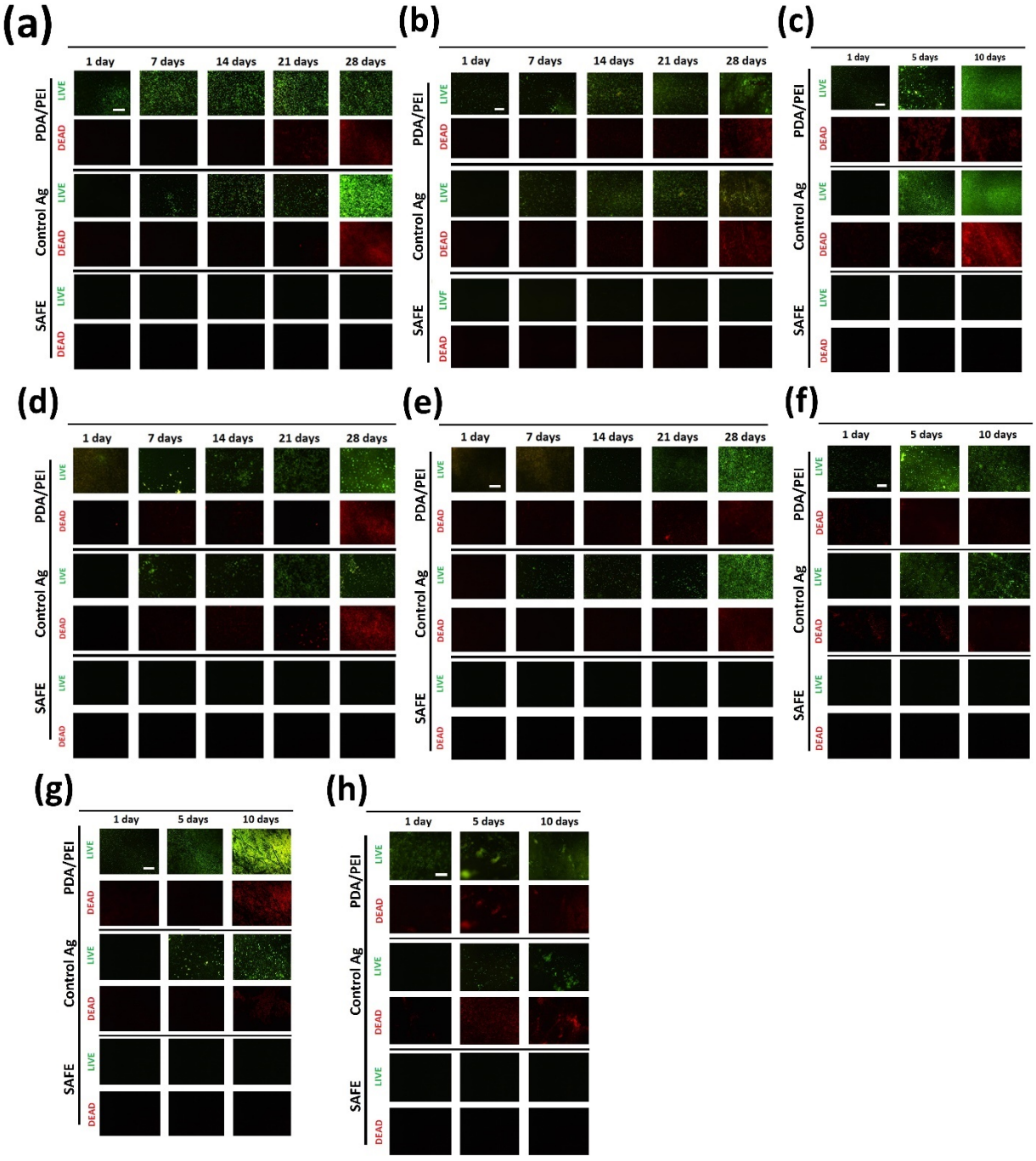
and cross-linking agent (gentamicin, amine-modified polyethylene glycol and poly (*N*-vinylamine), 1.5 mg/mL). The scale bar is 100 μm.



Supporting Figure 3. Fluorescence images of biofilm formed (green–live bacteria, red–dead bacteria) by *E. coli* (initial concentration: 1×10^6 CFU/mL, LB, 24 h) on the surface of silicon wafer treated with diverse compositions containing different catechol binders (pyrocatechol, pyrogallol, serotonin and tannic acid, 2 mg/mL), metal salts (silver nitrate, copper (II) nitrate, gold (III) chloride, gallium nitrate, nickel nitrate and zinc nitrate, 0.5 mg/mL) and polyethyleneimine as the cross-linking agent (1.5 mg/mL). The scale bar is 100 μm.

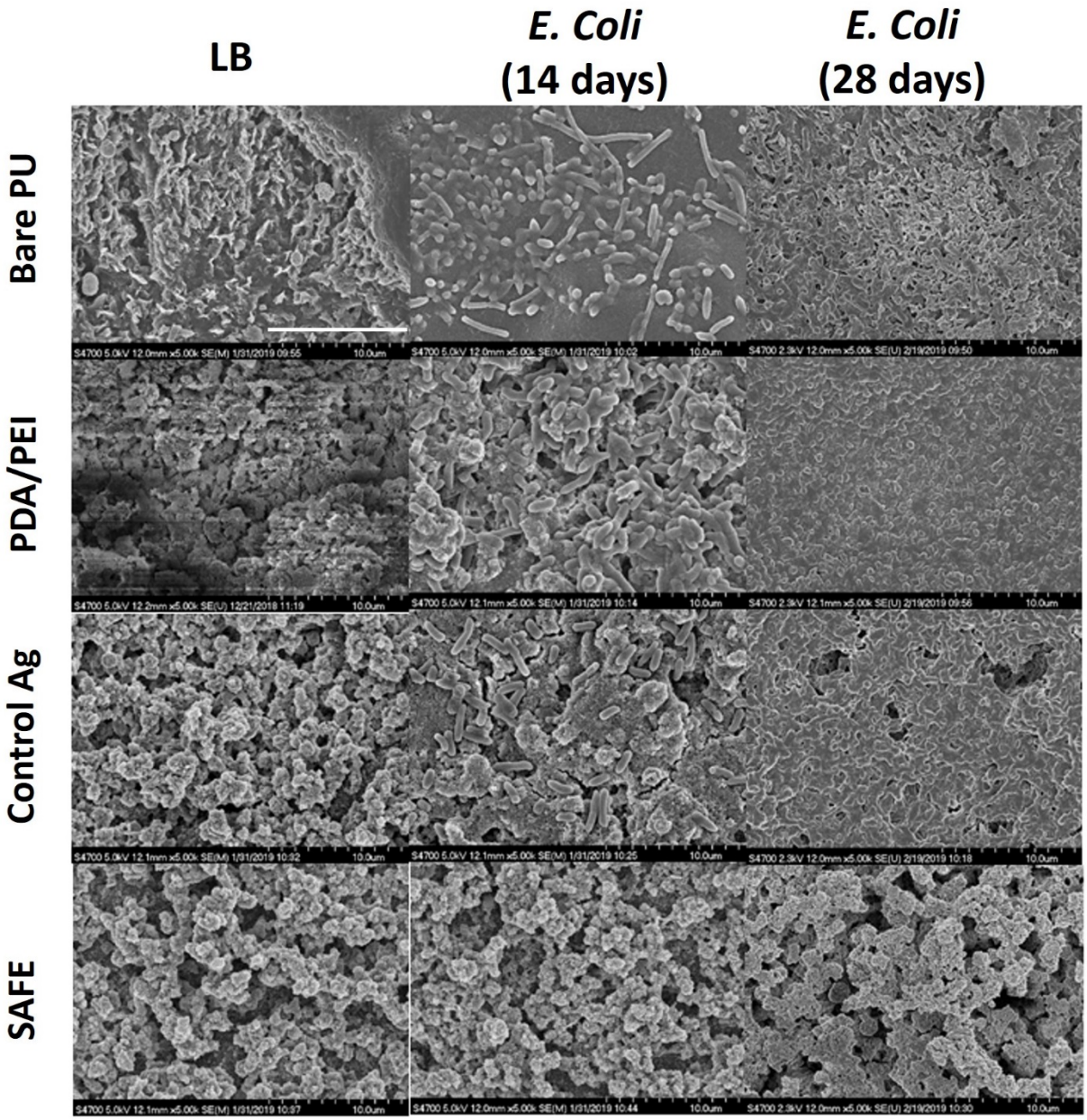


Supporting Figure 4. **(a)** Fluorescence images of biofilm formation (green–live bacteria, red–dead bacteria) on the surface of the control Ag exposed to *E. coli* (1×10^6 CFU/ mL, LB) for 28 days. **(b)** Fluorescence images of biofilm formation on the surface of the coatings based on diverse hydrophilic polymers after 7 days incubation with *E. coli* (1×10^6 CFU/mL, LB). The scale bar is 100 μm. The amount of silver released from the coatings based on different **(c)** DA: PDMA ratios and **(d)** molecular weights of PDMA after 21 days incubation with water.

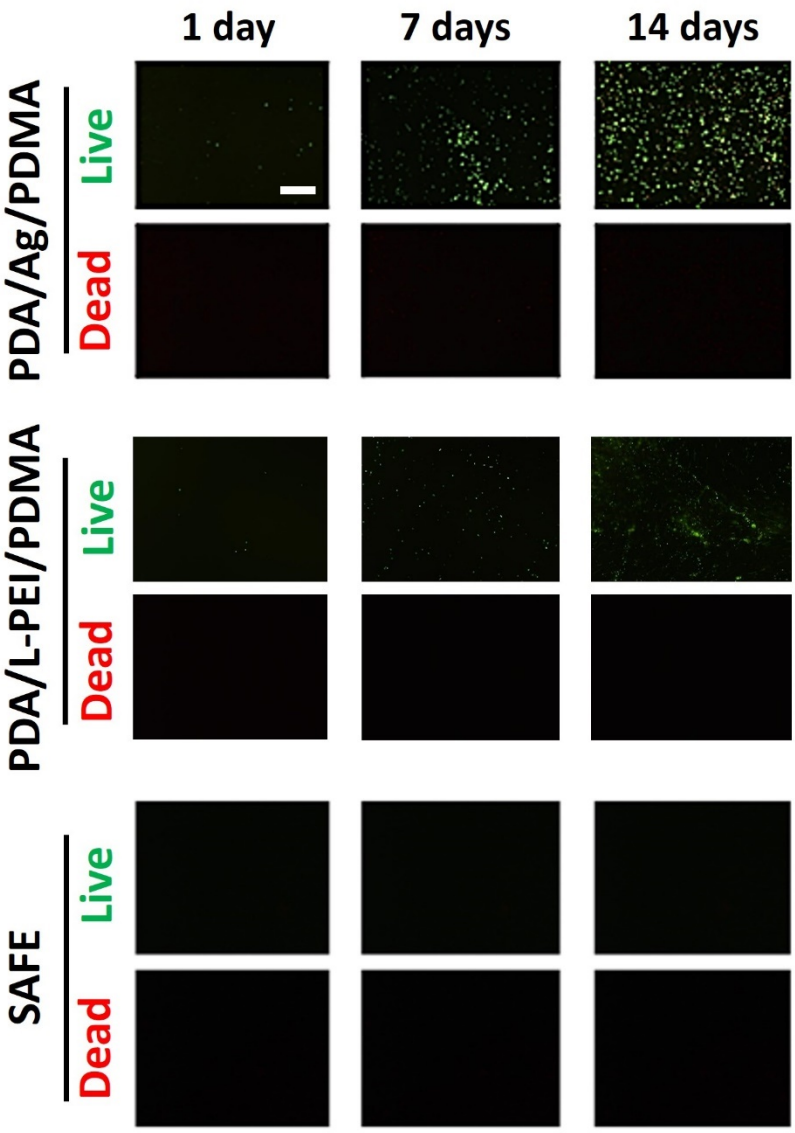


Supporting Figure 5. Fluorescence images of biofilm formation (green–live bacteria, red–dead bacteria) by **(a)** *E. coli*, **(b)** *P. aeruginosa*, **(c)** *E. faecalis*, **(d)** *S. aureus*, **(e)** *S. saprophyticus*, **(f)** MRSA, **(g)** *K. pneumonia*, and **(h)** *P. mirabilis* (1×10^6 CFU/mL, LB) on the surface of

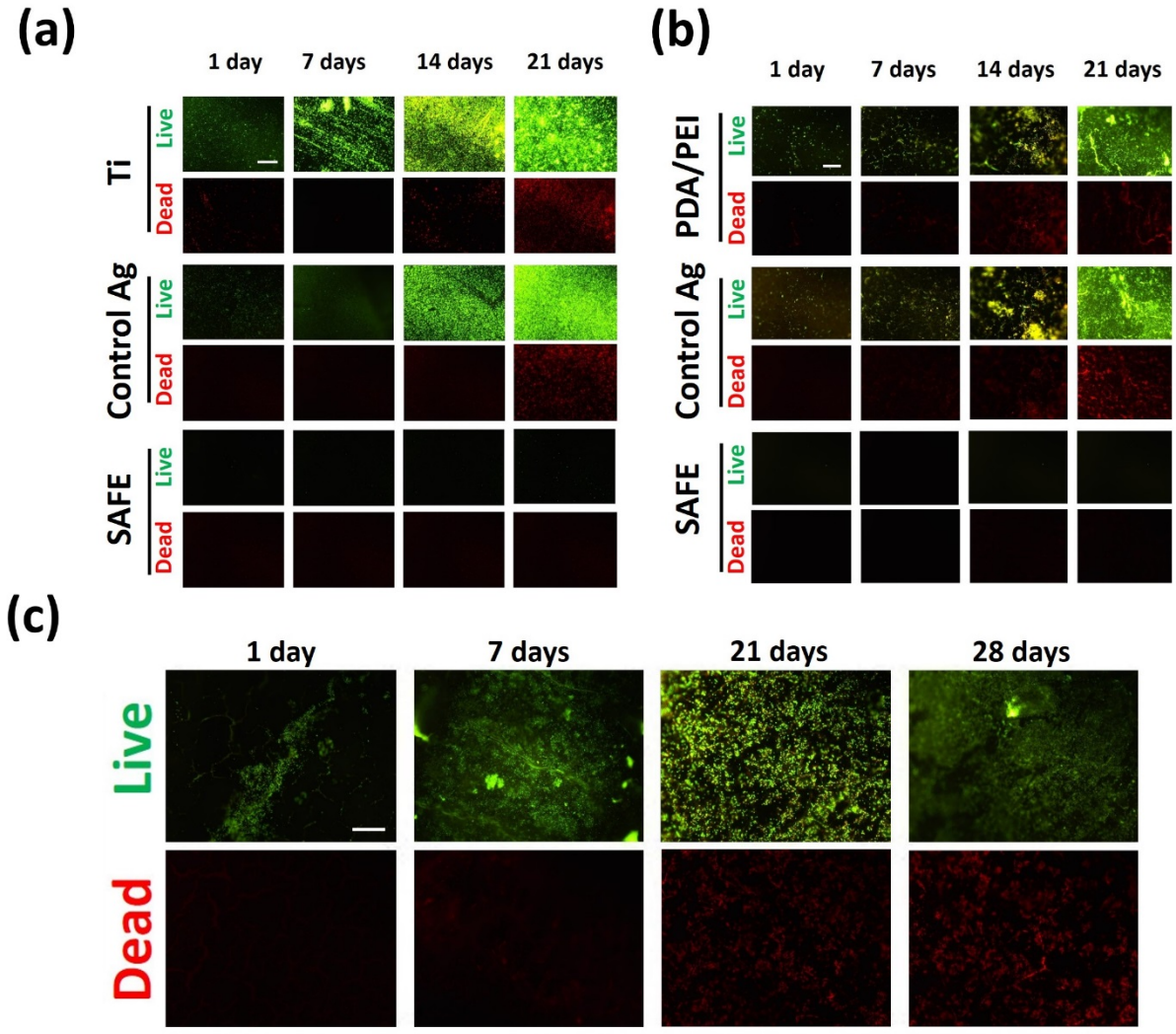
polyurethane substrates treated with different coatings (control PDA/PEI, control Ag and SAFE coating). The scale bar is 100 μm.



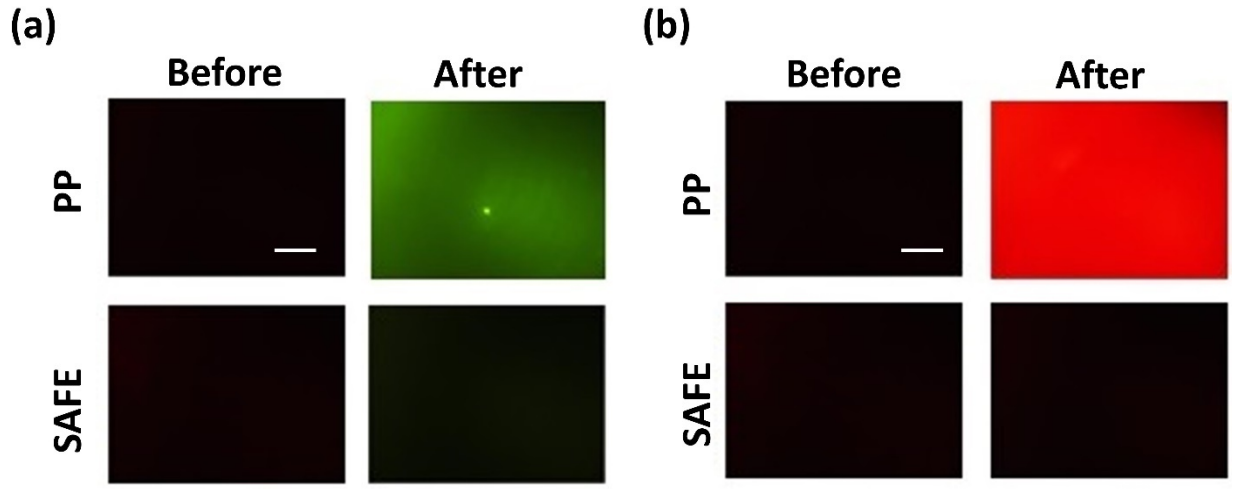
Supporting Figure 6. SEM images of biofilm formed by *E. coli* (1×10^6 CFU/mL, LB) on the surface of bare PU and different coatings (control PDA/PEI, control Ag and SAFE coating). The white scale bar is 10 μm.



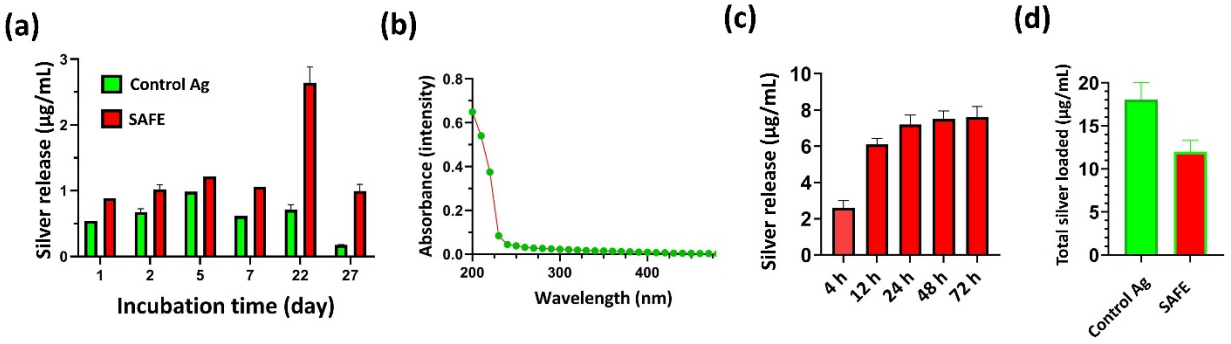
Supporting Figure 7. Fluorescence images of bacterial attachment to the surface of different coatings including control PDA/Ag/PDMA, PDA/L-PEI/PDMA and SAFE coating incubated with *E. coli* (initial concentration: 1×10^6 CFU/mL, LB) for 14 days. The scale bar is 100 μm.



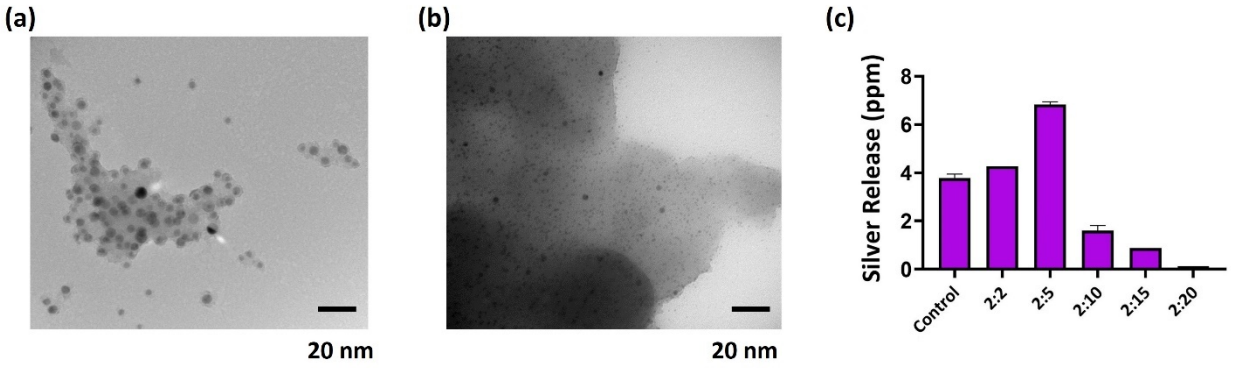
Supporting Figure 8. (a) Fluorescence images (green – live bacteria, red–dead bacteria) showing of the biofilm formation on the surface of the controls (uncoated Ti and the control Ag) and the SAFE coating on Ti in harsh conditions (>1×10^9 CFU/mL, *S. aureus*, in LB) for 21 days. Fluorescence images (green–live bacteria, red–dead bacteria) of the biofilm formation on the surface of the coated PU substrates exposed to stream of **(b)** *E. coli* fluid (> 1×10^9 CFU/mL, LB, 5 mL/min) for 21 days. **(c)** Fluorescence images (green – live bacteria, red – dead bacteria) of the biofilm formation on the surface of the control PDA/PEI coated PU substrate exposed to stream of *S. saprophyticus* fluid (>1×10^9 CFU/mL, LB, 5 mL/min) for 28 days. The scale bar is 100 µm.



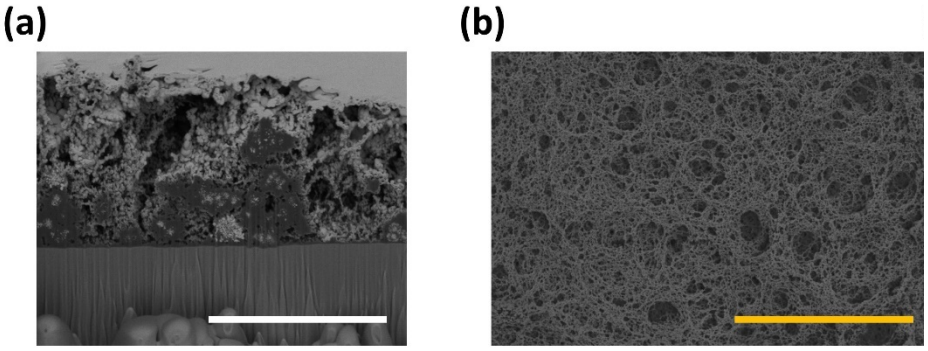
Supporting Figure 9. Fluorescence images of protein deposition on the surface of SAFE coated PP and uncoated PP after 1 h incubation with **(a)** FITC-BSA (1 mg/mL) and **(b)** Alexafluor 488-tagged fibrinogen (0.25 mg/mL) at 37 °C. The scale bar is 100 µm.



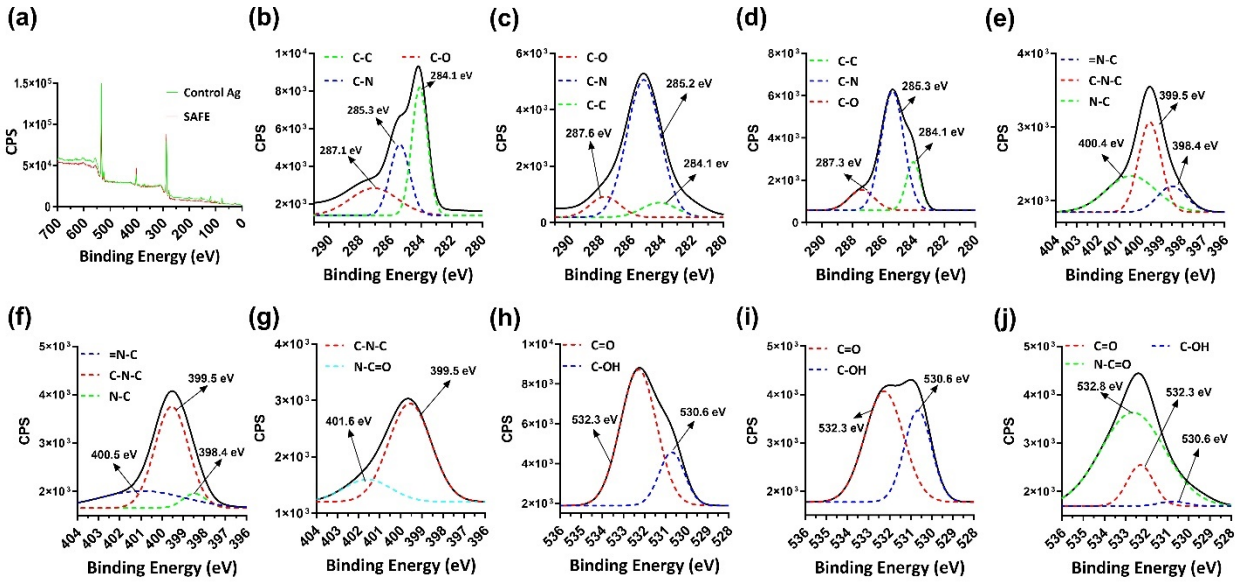
Supporting Figure 10. (a) UV-vis spectrum of water incubated with the SAFE coating for 24 h. **(b)** Non-cumulative silver release of the SAFE coating immersed in water for 28 days as a function of the immersion time. **(c)** Silver release of the SAFE coatings which prepared at different coating times over 28 days immersion in water. **(d)** Total concentration of silver incorporated into the control Ag and the SAFE coating.



Supporting Figure 11. TEM images of silver nanoparticles/assemblies incorporated into the SAFE coating **(a)** before and **(b)** after 28 days of immersion in water. The scale bar is 20 nm. **(c)** Silver release profile for coatings formed at different DA:PDMA ratios.

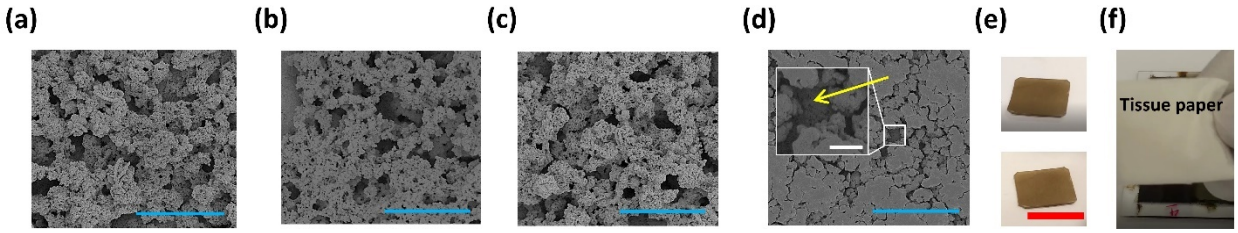


Supporting Figure 12. (a) SEM image of FIB-created cross-section of the control Ag on silicon wafer**.** The white scale bar is 5 μm. **(b)** SEM image of the control Ag coating. The yellow scale bar is 30 μm.

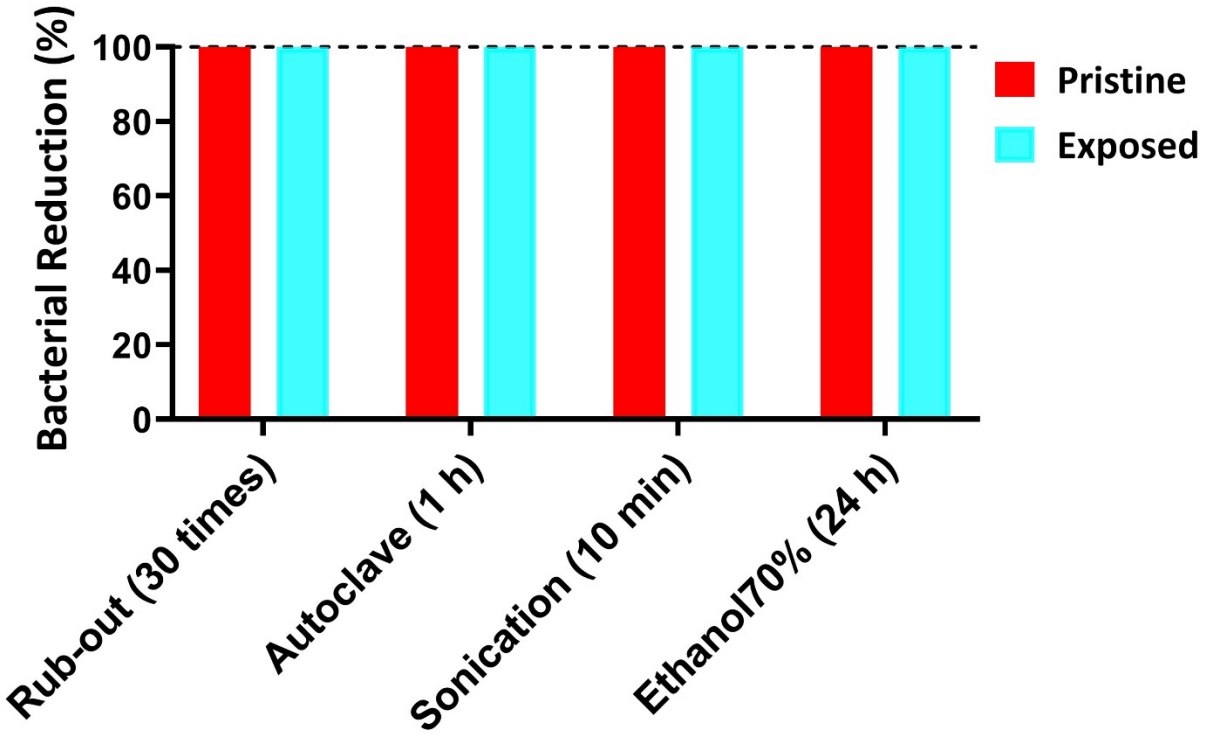


Supporting Figure 13. (a) XPS survey-scan of the control Ag coating and the SAFE coating. High-resolution spectra of C1s for, **(b)** PDA coating, **(c)** control Ag coating and **(d)** SAFE coating. High-resolution spectra of N1s for, **(e)** PDA coating, **(f)** control Ag coating and **(g)** SAFE coating. High-resolution spectra of O1s for, **(h)** PDA coating, **(i)** control Ag coating and **(j)** SAFE coating.

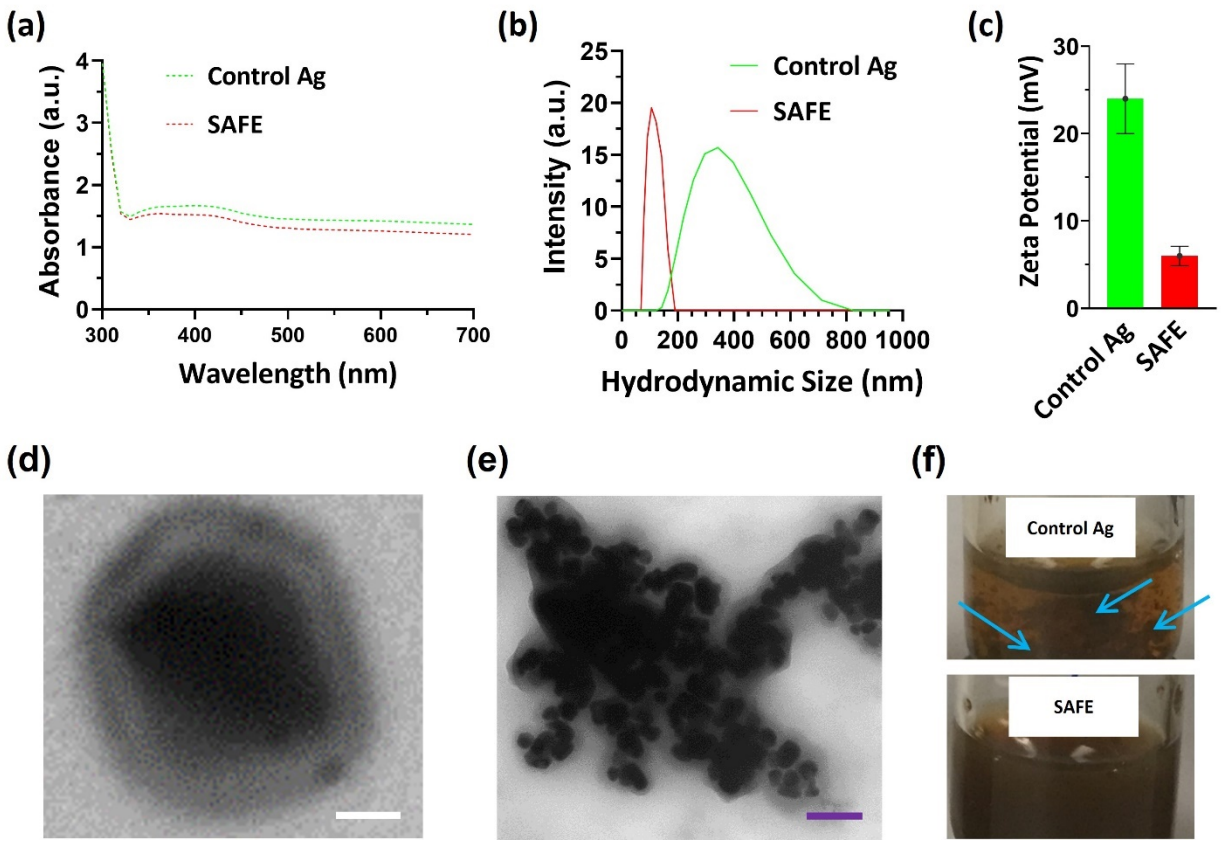
Supporting Figure 14. Water contact angle image of PDA, control Ag and SAFE coating.



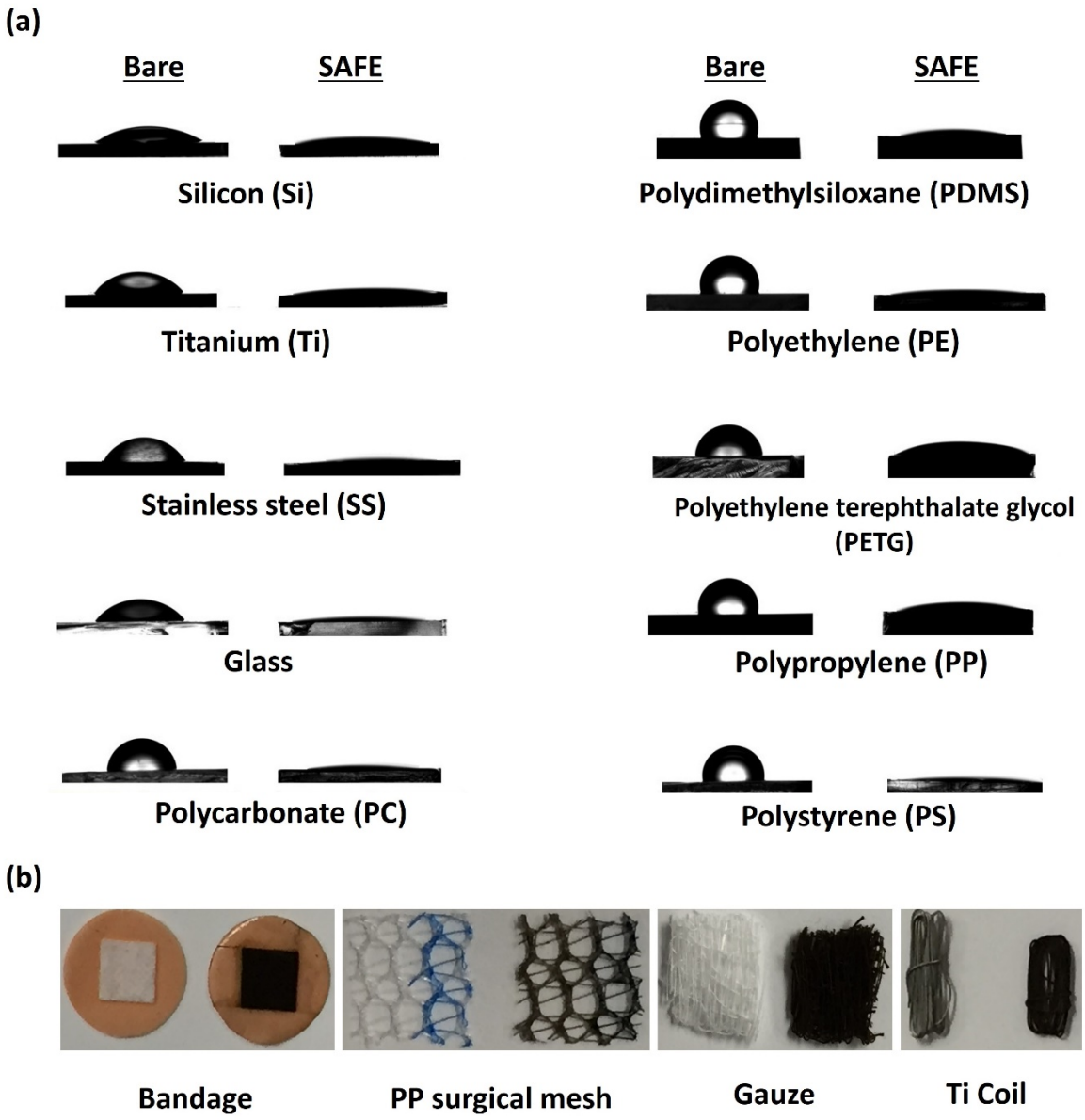
Supporting Figure 15. SEM images of the SAFE coating after exposure to different testing conditions: **(a)** sonication for 10 min, **(b)** immersion in ethanol 70% for 24 h, **(c)** autoclaving for 1 h at 120 °C and 15 psi and **(d)** back and forth rub-out (30 rubs). The white and blue scale bars are 500 nm and 5 μm. The yellow arrow pointed out the area that retained its original morphology and its great surface coveage upon the rub-out test. **(e)** Digital images of the SAFE coated PU sheet prior to (top image) and following (bottom image) 30 back and forht rubs with a piece of kimwipe tissue paper. The red scale bar is 2 cm. **(f)** Digital image of the finger-held kimwipe tissue paper rubbed out 30 times back and forth over the SAFE-sprayed piece of glass. The background of the image is the rubbed sample laid flat on the bench.



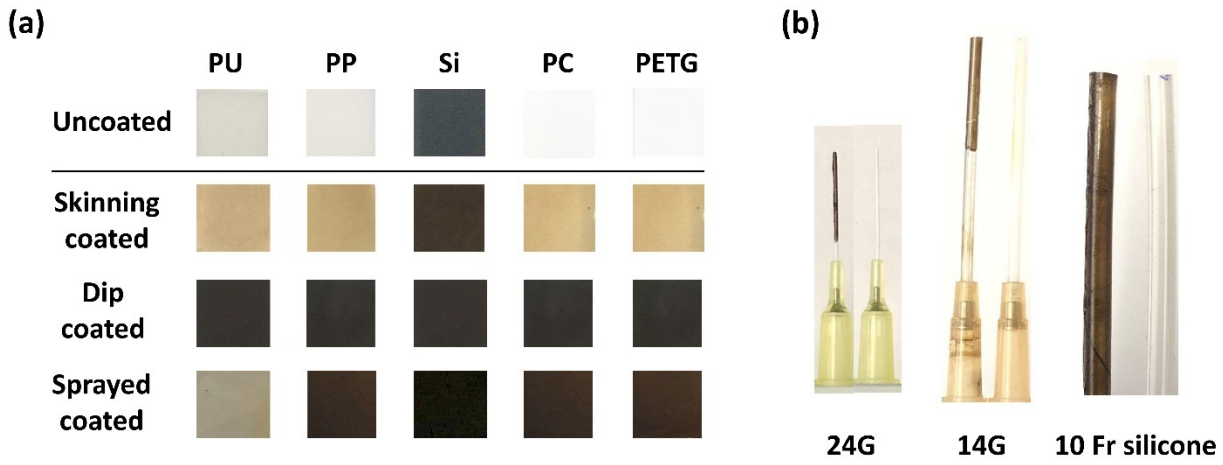
Supporting Figure 16. The percentage of reduction in bacterial attactment to the surafce of original and exposed SAFE coating after 7 days incubation with *E. coli* (1×10^6 CFU/mL, LB).



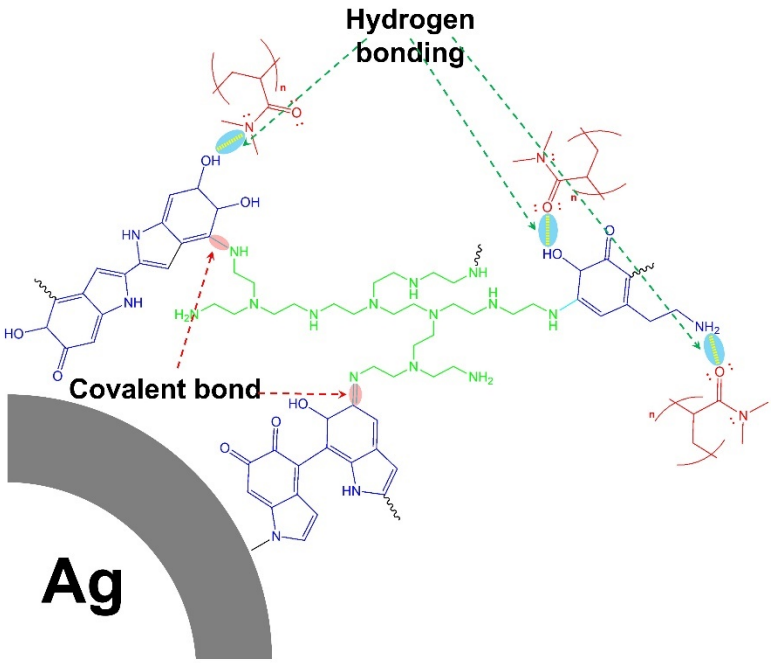
Supporting Figure 17. (a) UV-vis spectra of control Ag and SAFE solutions. **(b)** Hydrodynamic size and **(c)** zeta potential of control Ag and SAFE assemblies. **(d)** High magnification TEM image of an individual silver nanoparticle embedded in the SAFE assembly. The white scale bar is 3 nm. **(e)** TEM image of silver aggregates in the control Ag suspension. The violet scale bar is 50 nm. **(f)** Digital images of the control Ag suspension (top) and the SAFE suspension (bottom) after 60 days at room temperature. Blue arrows point out precipitates.



Supporting Figure 18. (a) Water contact angle images of diverse materials coated with SAFE coating. **(b)** Digital images of bandage, PP surgical mesh, gauze, and Ti coil; uncoated (left) and SAFE coated (right).

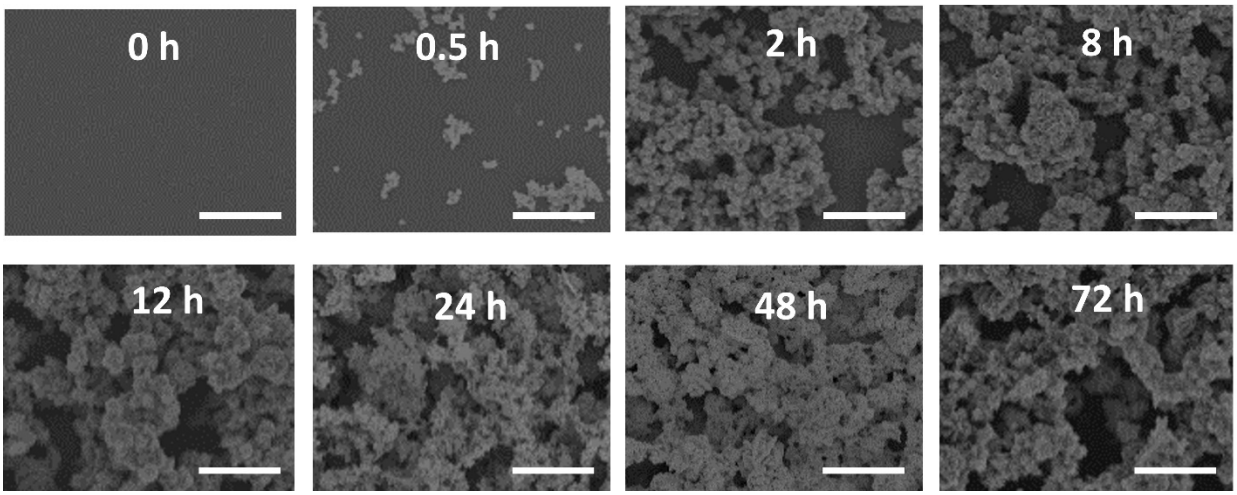


Supporting Figure 19. Digital images of **(a)** diverse materials coated with different SAFE coating methods including solution-skinning, dipping, and spraying. **(b)** Digital images of polymeric catheters coated with the SAFE coating utilizing the solution-skinning method. Uncoated catheters were also shown on the right side of images.



Supporting Figure 20. A schematic representation of the molecular architecture for the SAFE structure. The main intermolecular interactions involved in the SAFE coating formation include

covalent bonds, coordinate bonds and hydrogen bonding. PEI: green, DA/PDA: blue, PDMA: red, and a piece of the silver nanoparticle: grey.



Supporting Figure 21. SEM images of silicon wafers treated with the SAFE composition at different time points (0, 0.5, 2, 8, 12, 24, 48 and 72 h). The scale bar is 2 μm.

Supporting Tables

Supporting Table 1. Composition of diverse coating solutions tested

Color-map Cell	Catechol (mg^{-1})	LAP (mg/mL)	Metal Salt (mg/mL)
A1	L-DOPA (2)	Polyethylenimine (1.5)	Silver Nitrate (0.5)
A2	1,3,5- benzenetriol (2)	Polyethylenimine (1.5)	Silver Nitrate (0.5)
A3	Resorcinol (2)	Polyethylenimine (1.5)	Silver Nitrate (0.5)
A4	Dopamine (2)	Gentamicin (0.5)	Silver Nitrate (0.5)
A5	Dopamine (2)	Gentamicin (1.5)	Silver Nitrate (0.5)
A6	Dopamine (2)	Amine terminated 4-arm polyethylene glycol (0.5)	Silver Nitrate (0.5)
A7	Dopamine (2)	Amine terminated 4-arm polyethylene glycol (1.5)	Silver Nitrate (0.5)
A8	Dopamine (2)	Poly (*N*-vinyl amine) (0.5)	Silver Nitrate (0.5)
A9	Dopamine (2)	Polyethylenimine (1.5)	Silver Nitrate (0.5)
A10	Dopamine (2)	Polyethylenimine (1.5)	Gold (III) Chloride (0.5)
A11	Dopamine (2)	Polyethylenimine (1.5)	Copper (II) Nitrate (0.5)
A12	Dopamine (2)	Polyethylenimine (1.5)	Gallium Nitrate (0.5)
A13	Dopamine (2)	Polyethylenimine (1.5)	Nickel Nitrate (0.5)
B1	Dopamine (2)	Polyethylenimine (1.5)	Zinc Nitrate (0.5)
B2	Dopamine (2)	PVAm1.5	Silver Nitrate (0.5)
B3	Pyrocatechol (2)	Gentamicin (1.5)	Silver Nitrate (0.5)
B4	Pyrocatechol (2)	Gentamicin (1.5)	Copper (II) Nitrate (0.5 L)
B5	Pyrocatechol (2)	Amine terminated 4-arm polyethylene glycol (0.5)	Silver Nitrate (0.5)
B6	Pyrocatechol (2)	Amine terminated 4-arm polyethylene glycol (0.5)	Copper (II) Nitrate (0.5)
B7	Pyrocatechol (2)	Polyethylenimine (1.5)	Silver Nitrate (0.5)
B8	Pyrocatechol (2)	Polyethylenimine (1.5)	Gold (III) Chloride (0.5)
B9	Pyrocatechol (2)	Polyethylenimine (1.5)	Copper (II) Nitrate (0.5)
B10	Pyrocatechol (2)	Polyethylenimine (1.5)	Gallium Nitrate (0.5)
B11	Pyrocatechol (2)	Polyethylenimine (1.5)	Copper (II) Nitrate (0.5)
B12	Pyrocatechol (2)	Polyethylenimine (1.5)	Zinc Nitrate (0.5)
B13	Pyrocatechol (2)	Poly (*N*-vinyl amine) (1.5)	Silver Nitrate (0.5)
C1	Pyrocatechol (2)	Poly (*N*-vinyl amine) (1.5)	Copper (II) Nitrate (0.5)
C2	Pyrogallol (2)	Amikacin (1.5)	Copper (II) Nitrate (0.5)
C3	Pyrogallol (2)	Amikacin (1.5)	Copper (II) Nitrate (1.5)
C4	Pyrogallol (2)	Gentamicin (1.5)	Silver Nitrate (0.5)
C5	Pyrogallol (2)	Gentamicin (1.5)	Copper (II) Nitrate (0.5)
C6	Pyrogallol (2)	Amine terminated 4-arm polyethylene glycol (0.5)	Silver Nitrate (0.5)
C7	Pyrogallol (2)	Amine terminated 4-arm polyethylene glycol (0.5)	Copper (II) Nitrate (0.5)
C8	Pyrogallol (2)	Polyethylenimine (1.5)	Silver Nitrate (0.5)
C9	Pyrogallol (2)	Polyethylenimine (1.5)	Gold (III) Chloride (0.5)
C10	Pyrogallol (2)	Polyethylenimine (1.5)	Copper (II) Nitrate (0.5)
C11	Pyrogallol (2)	Polyethylenimine (1.5)	Gallium Nitrate (0.5)
C12	Pyrogallol (2)	Polyethylenimine (1.5)	Copper (II) Nitrate (0.5)
C13	Pyrogallol (2)	Polyethylenimine (1.5)	Zinc Nitrate (0.5)
D1	Pyrogallol (2)	Poly (*N*-vinyl amine) (1.5)	Silver Nitrate (0.5)

D2	Pyrogallol (2)	Poly (*N*-vinyl amine) (1.5)	Copper (II) Nitrate (0.5)
D3	Serotonin (2)	Gentamicin (1.5)	Silver Nitrate (0.5)
D4	Serotonin (2)	Gentamicin (1.5)	Copper (II) Nitrate (0.5)
D5	Serotonin (2)	Amine terminated 4-arm polyethylene glycol (0.5)	Silver Nitrate (0.5)
D6	Serotonin (2)	Amine terminated 4-arm polyethylene glycol (0.5)	Copper (II) Nitrate (0.5)
D7	Serotonin (2)	Polyethylenimine (1.5)	Silver Nitrate (0.5)
D8	Serotonin (2)	Polyethylenimine (1.5)	Gold (III) Chloride (0.5)
D9	Serotonin (2)	Polyethylenimine (1.5)	Copper (II) Nitrate (0.5)
D10	Serotonin (2)	Polyethylenimine (1.5)	Gallium Nitrate (0.5)
D11	Serotonin (2)	Polyethylenimine (1.5)	Copper (II) Nitrate (0.)
D12	Serotonin (2)	Polyethylenimine (1.5)	Zinc Nitrate (0.5)
D13	Serotonin (2)	Poly (*N*-vinyl amine) (1.5)	Silver Nitrate (0.5)
E1	Serotonin (2)	Poly (*N*-vinyl amine) (1.5)	Copper (II) Nitrate (0.5)
E2	Tannic Acid (2)	Gentamicin (1.5)	Silver Nitrate (0.5)
E3	Tannic Acid (2)	Gentamicin (1.5)	Copper (II) Nitrate (0.5)
E4	Tannic Acid (2)	Amine terminated 4-arm polyethylene glycol (0.5)	Silver Nitrate (0.5)
E5	Tannic Acid (2)	Amine terminated 4-arm polyethylene glycol (0.5)	Copper (II) Nitrate (0.5)
E6	Tannic Acid (2)	Polyethylenimine (1.5)	Silver Nitrate (0.5)
E7	Tannic Acid (2)	Polyethylenimine (1.5)	Gold (III) Chloride (0.5)
E8	Tannic Acid (2)	Polyethylenimine (1.5)	Copper (II) Nitrate (0.5)
E9	Tannic Acid (2)	Polyethylenimine (1.5)	Gallium Nitrate (0.5)
E10	Tannic Acid (2)	Polyethylenimine (1.5)	Copper (II) Nitrate (0.5)
E11	Tannic Acid (2)	Polyethylenimine (1.5)	Zinc Nitrate (0.5)
E12	Tannic Acid (2)	Poly (*N*-vinyl amine) (1.5)	Silver Nitrate (0.5)
E13	Tannic Acid (2)	Poly (*N*-vinyl amine) (1.5)	Copper (II) Nitrate (0.5)

Supporting Table 2. XPS data of control Ag and SAFE coating

	Si (%)	Cl (%)	C (%)	Ag (%)	O (%)	N/C	O/C
Control Ag	0.77	0.90	36.69	1.02	53.64	0.189	1.461
SAFE	0.01	0.02	48.29	0.03	37.77	0.288	0.783

Supporting Movies

Supporting Movie 1. Stability against rubbing. Movie showing rubbing out process on glass coated with SAFE coating. SAFE coating was prepared by a spray coating process.

Supporting Movie 2. Stability against rubbing. Movie showing rubbing out process on PP coupon coated with SAFE coating. SAFE coating was prepared by a dipping process

Supporting Movie 3. Stability against rubbing. Movie showing rubbing out process on PU coupon coated with SAFE coating. SAFE coating was prepared by a skinning process.

Supporting Movie 4. Movie showing the spray coating process using SAFE solution.

Supporting Movie 5. Movie showing the skinning coating process on PP coupon using the SAFE layer formed at air-liquid interface.

Supporting Movie 6. Movie showing the solution-skinning process on PDMS ball using the SAFE layer formed at air-liquid interface.