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Reversible Superhydrophobic Coatings on Lifeless and Biotic Surfaces via Dry-Painting of Aerogel Microparticles

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Materials preparation

Aerogel microparticle samples (entry 1, 3 and 5 in Table S1) were steam sterilization at 115 ^{o}C for 10min, dried at 55 ^{o}C and finally dispersed in culture solution (RPMI-1640) for 24 h at 37 ^{o}C. The **incubated solution (A)** was used in the cell viability assay. The OD values of 96 well plates were read at 570 nm by Microplate Reader (Thermo MK3)

MTT Cell viability assay.

The cytotoxicity was studied using the 3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyl tetrazolium-bromide (MTT) assay. L929 cells (10^4 cell/ml) were seeded in 96-well plates and incubated in DMEM media containing 10% FBS at 37 ^{o}C in 5% CO_2/95% air atmosphere for 24 h. The media were replaced with fresh media containing sample **incubated solution (A)** and incubated for 68 h. Thereafter, MTT solution was added. The plates were incubated at 37^{o}C for another 4-6 h. The intracellular metabolized product MTT formazan was then retrieved by an addition of dimethylsulfoxide (DMSO). The plates were read at 570 nm by Microplate Reader (ThermoMK3), and the cell viability (CV) was calculated according to the following equation:

$CV=(A_s-A_b)/(A_0-A_b)\times 100\%$

where A_s are the absorption of sample group, A_0 is the absorption of control group, and A_b is the absorption of background (the plate).

The negative group used an incubated solution containing 80 μl double-antiserum in 720 μl RPMI-1640, and the positive group used an incubated solution by adding 50 μl

DMSO into 950 μl RPMI-1640. When the CV value is higher than 80%, it's considered nontoxic.

Tables

Table S1. Recipe, structure parameters, and CA of the SAMPs.

Entry[a]	Solvent[b]	Hydrophobic reagent[c]	Specific surface area (m^2/g)	Pore Volume (cm^3/g)	Density[d] (mg/cm^3)	Contact angle (°)
1	Hexane	HMDS	779.4 ± 5.8	2.0	89.0	155
2	Hexane	TMES	576.3 ±3.9	0.6	107.4	153
3	HMDSO	HMDS	769.4 ±6.8	1.6	84.9	151
4	HMDSO	TMES	576.4 ±5.1	2.4	112.5	150
5	Heptane	HMDS	709.0 ±5.9	1.4	174.5	145
6	Heptane	TMES	685.7 ±3.6	0.8	214.3	149
7	ethanol	HMDS	236.9 ±4.1	0.2	851.7	--

[a)] 5 ml CS sols were used for all the samples; [b)] 5 ml of the corresponding solvent that equal to CS sol were used; [c)] 1ml HMDS were used while 1.5 ml TMES were used. [d)] Tap density determined by dividing their weights by its volume.

Table S2. OD results, and the calculated RGR values of the samples and reference

	Control (A_o)	**Negative (A_s)**	**Positive (A_s)**	**sample1 (A_s)**	**Sample2 (A_s)**	**Sample3 (A_s)**	**Background (A_b)**
A	1.289	1.407	0.701	1.33	1.241	1.286	0.151
B	1.365	1.433	0.593	1.313	1.344	1.323	0.134
C	1.362	1.37	0.604	1.332	1.372	1.265	0.133
D	1.351	1.389	0.588	1.247	1.27	1.312	0.137
∑	5.367	5.599	2.486	5.222	5.227	5.186	0.555
Average CV (%)	--	104.8	40.2	97.0	97.1	92.2	--

OD values were collected at 570 nm

Figures

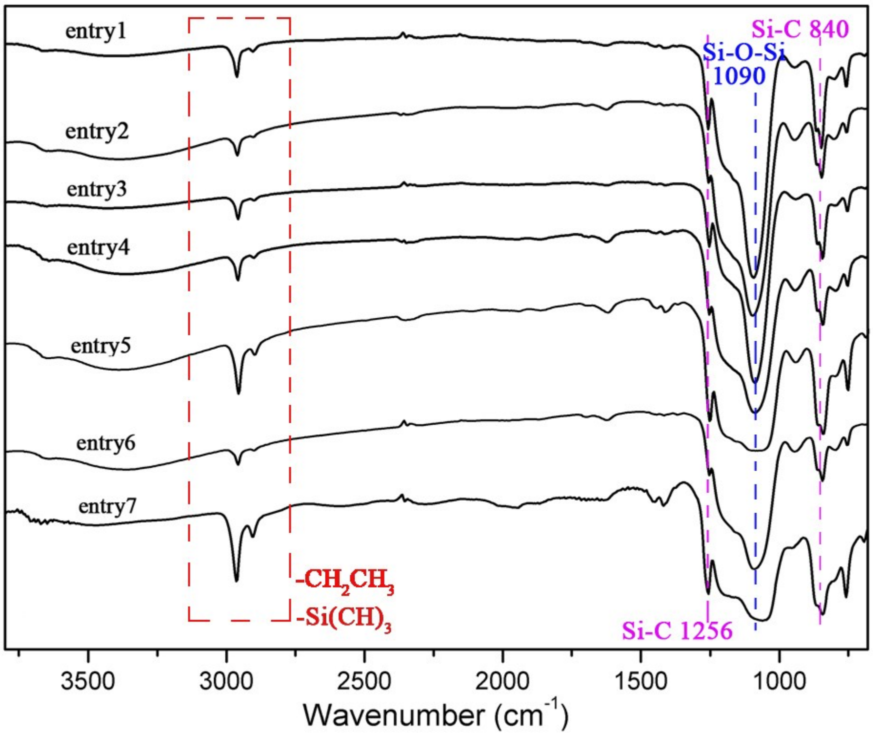

Figure S1. FTIR spectra of the SHB-SAMPs (entry 1-6) and xerogel (entry 7).

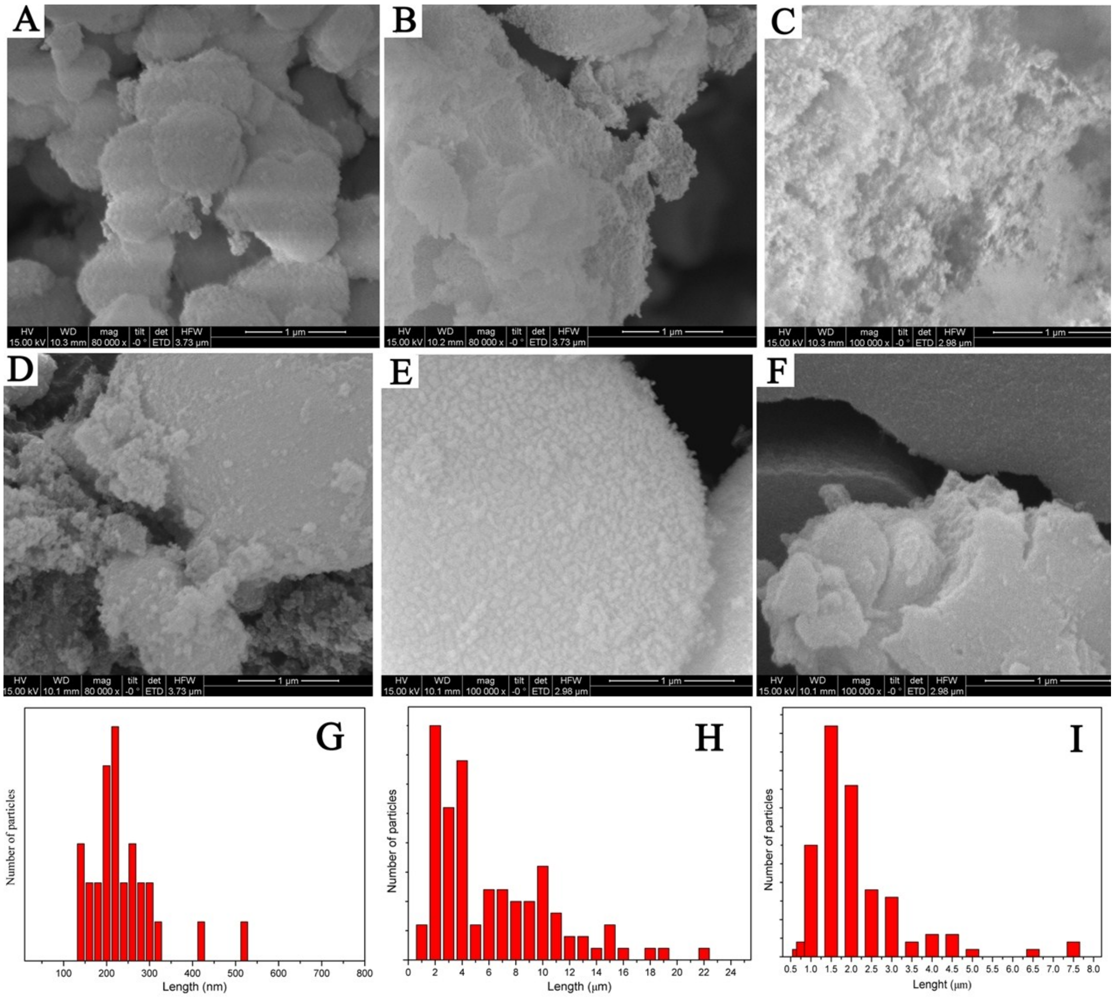

Figure S2. SEM images of the SHB-SAMPs: (A) entry 2; (B) entry 3; (C) entry 4; (D) entry 5; (E) entry 6; (F) entry 7. Particle size distribution of (G) entry 1, (H) entry 3, and (I) entry 5.

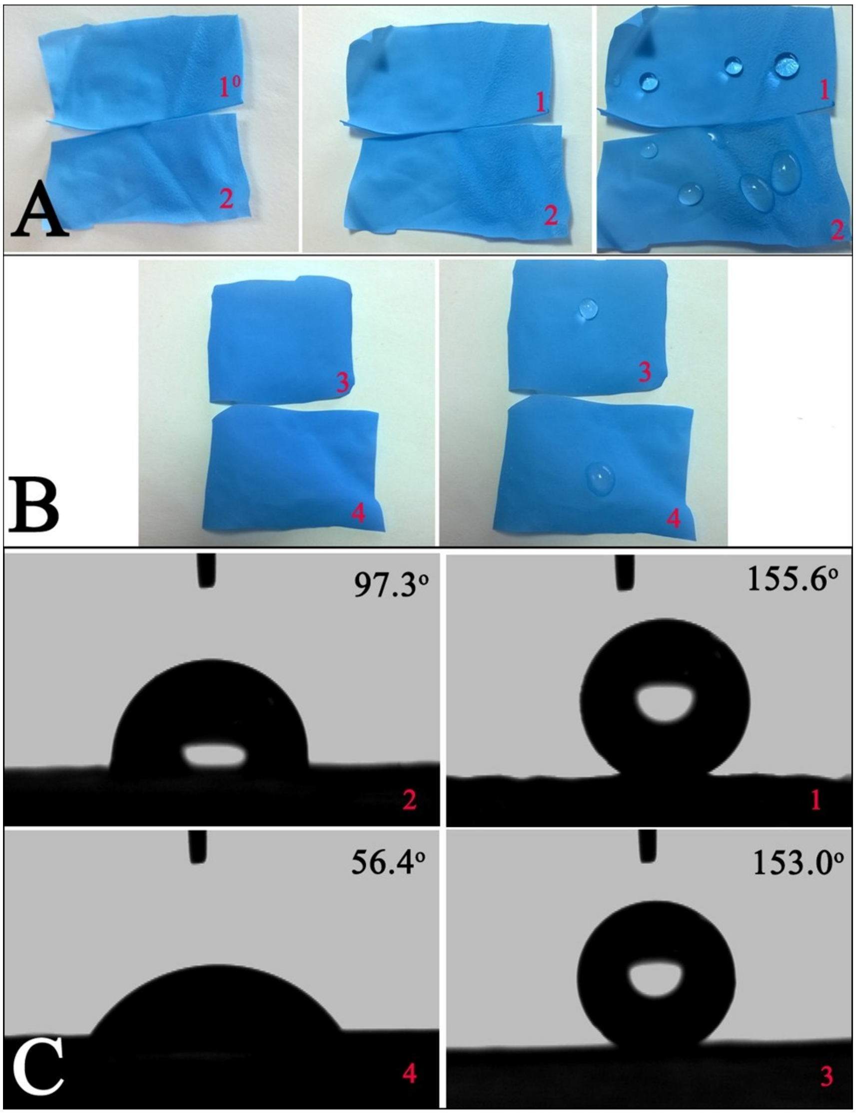

Figure S3. (A) Photo images of nitrile glove (outside) with and without SHB-SAMP coatings, no visible differences in appearance: *1⁰* and *2* is the native nitrile glove, *1* is the SHB-SAMP coated glove. (B) Photo images of nitrile glove (inside) with (*3*) and without coating (*4*). (C) Contact angle measurement of nitrile gloves: (*1*) after coated entry 1 on the outside surface; (*2*) native outside surface; (*3*) after coated entry 1 on the inside surface; (*4*) native inside surface.

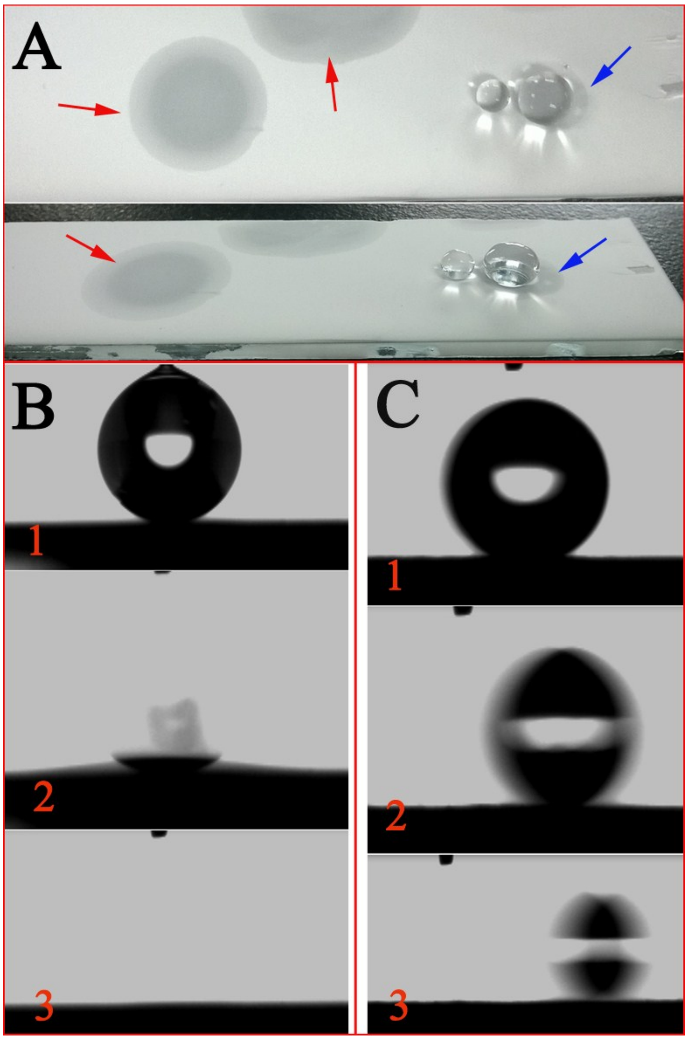

Figure S4. (A) Photo images of silica gel plate, red arrow shows the native surface is superhydrophilic and the blue arrow shows the entry 1 coated surface that become superhydrophobic. (B) Contact angle measurement of native silica gel plate, which completely absorb the water droplet. (C) Contact angle measurement of entry 1 coated silica gel plate, which is superhydrophobic and the water droplet rolled off.

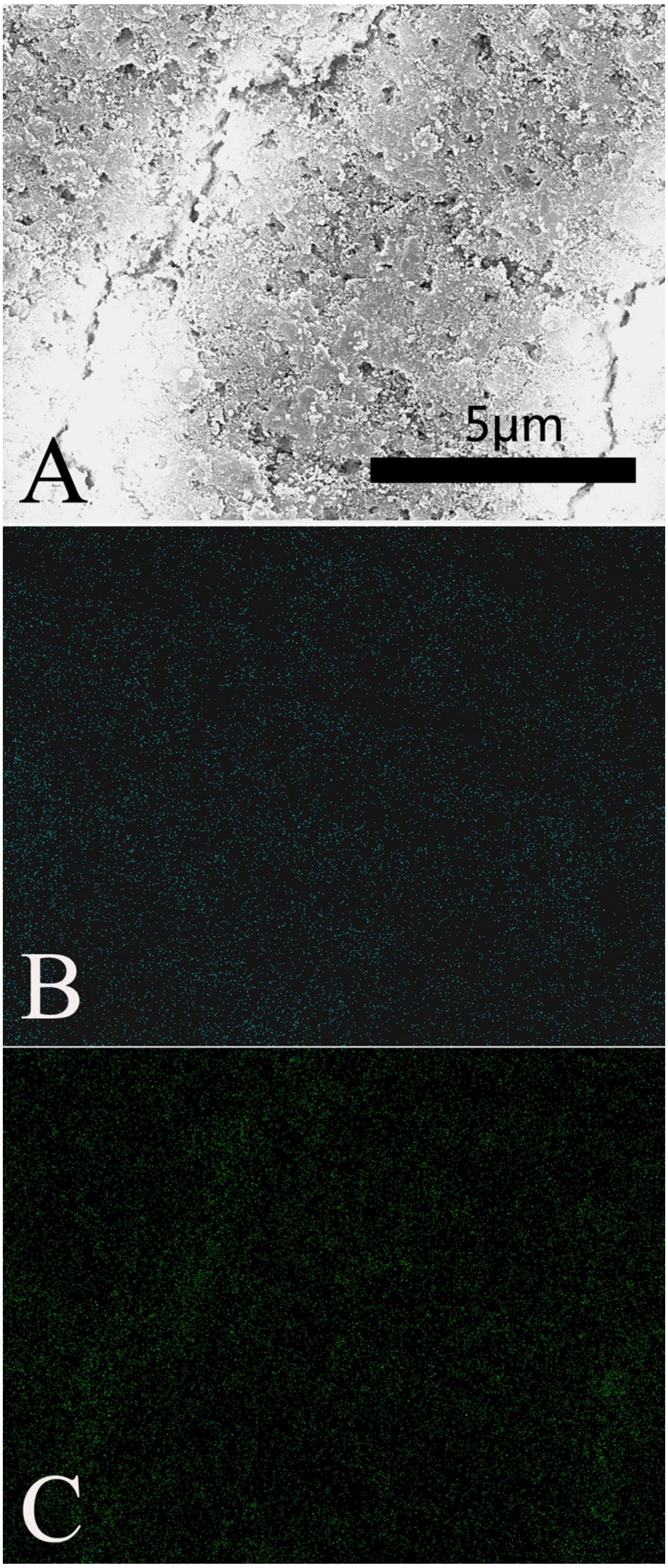

Figure S5. (A) SEM images of entry 1 coated glove surface. SEM-EDS spectra of A: (B) oxygen (blue) and (C) silicon (green)

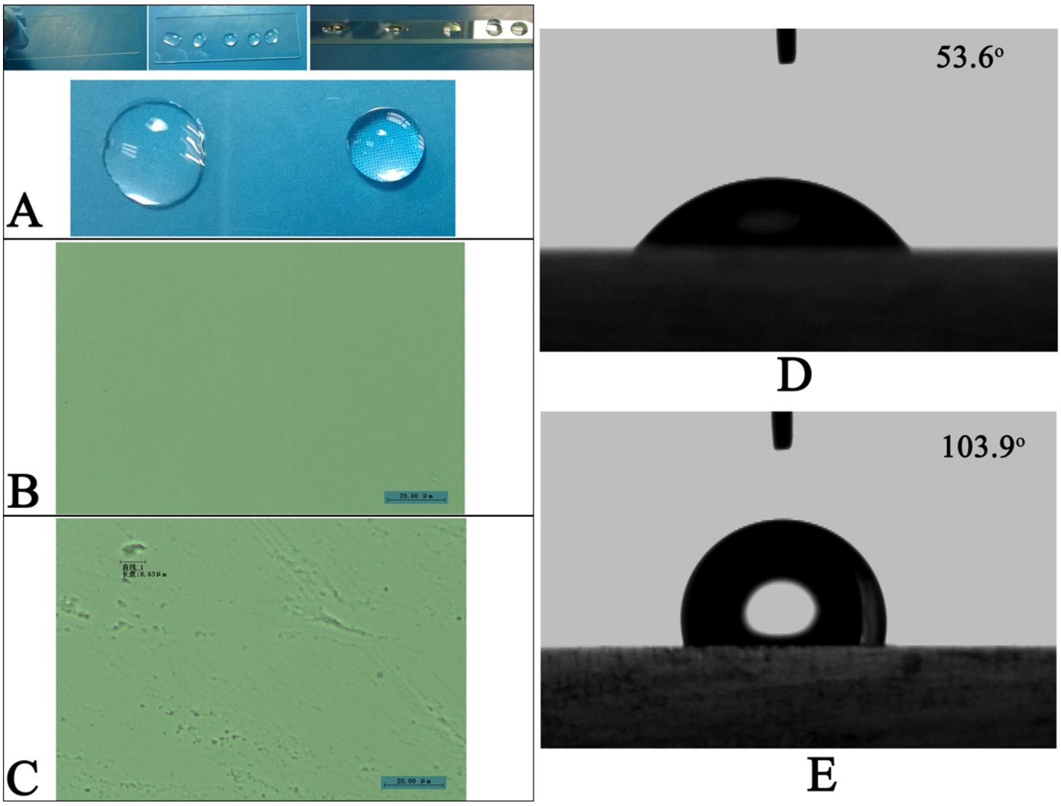

Figure S6. (A) Photo images of glasses: in the left side of all the images is the native surface and the right side is entry 1 coated surface, also no visible difference can be observed, but the contact angle is significantly increased from 53.6 ^{0} to 103.9 ^{0}. (B) Optical microphotograph of native glass, which is clear and smooth. (C) Optical microphotograph of entry 1 coated glass, silica microparticles can be observed inhomogeneous distributed on the surface. Photo images of water droplet on (D) native glass and (F) aerogel coated glass.

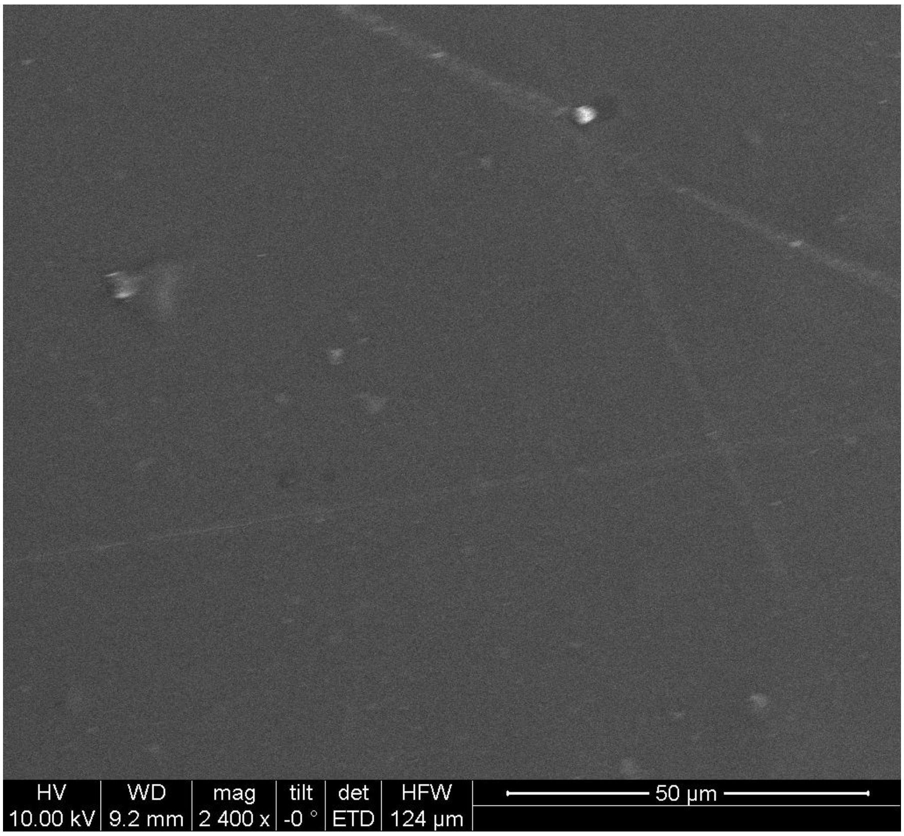

Figure S7. SEM images of PET film.

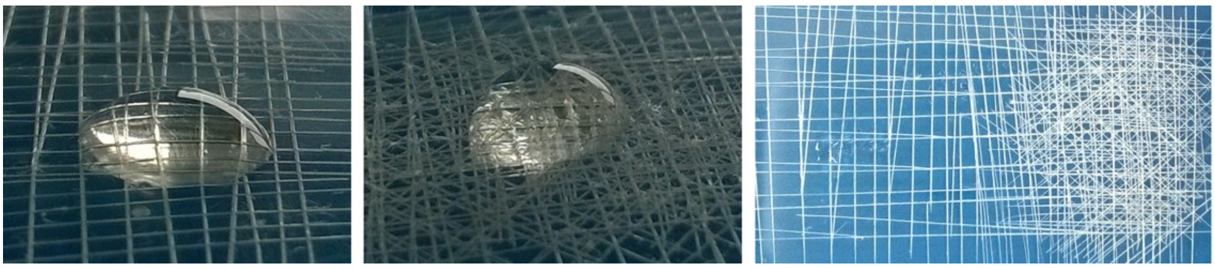

Figure S8. Photo images of water droplet on roughened PET (without aerogel coating).

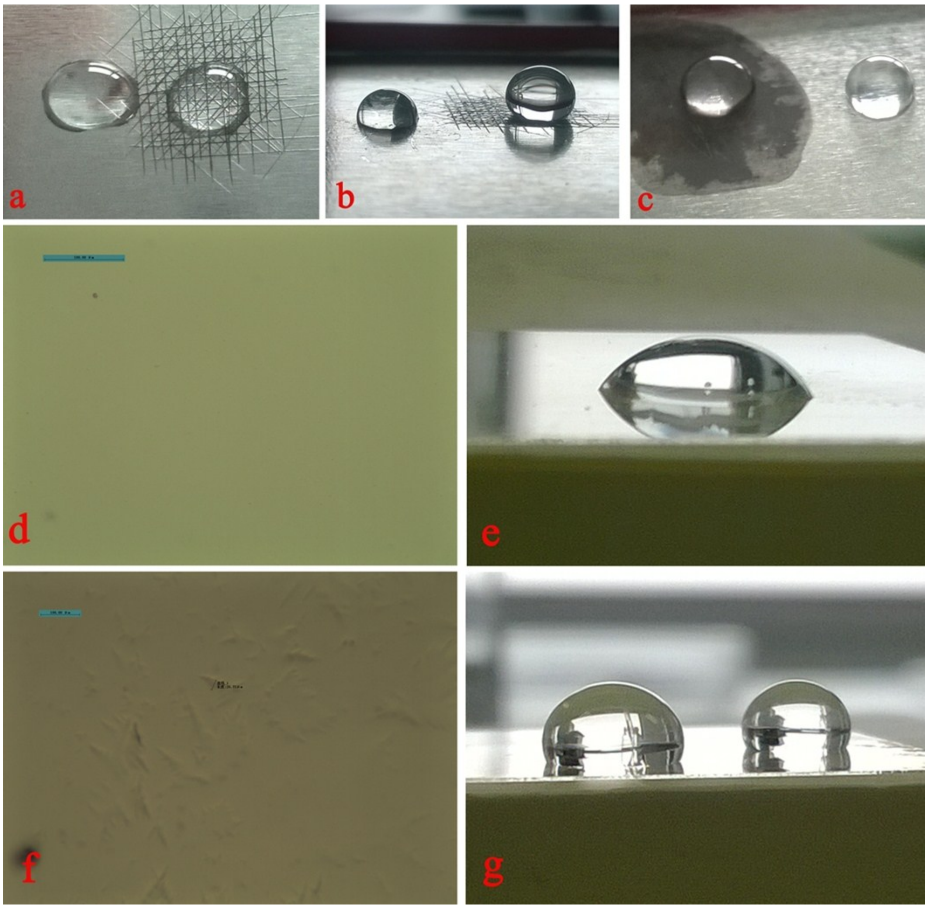

Figure S9. (a) water droplets on stainless steel. (b) and (c) water droplets on aerogel coated stainless steel. (c) stainless steel corrosive by HCl. (d) and (f) optical microphotograph of glass before and after corrosion. Water droplets on (e) aerogel coated glass surface, and (g) aerogel coated corrosive glass surface.

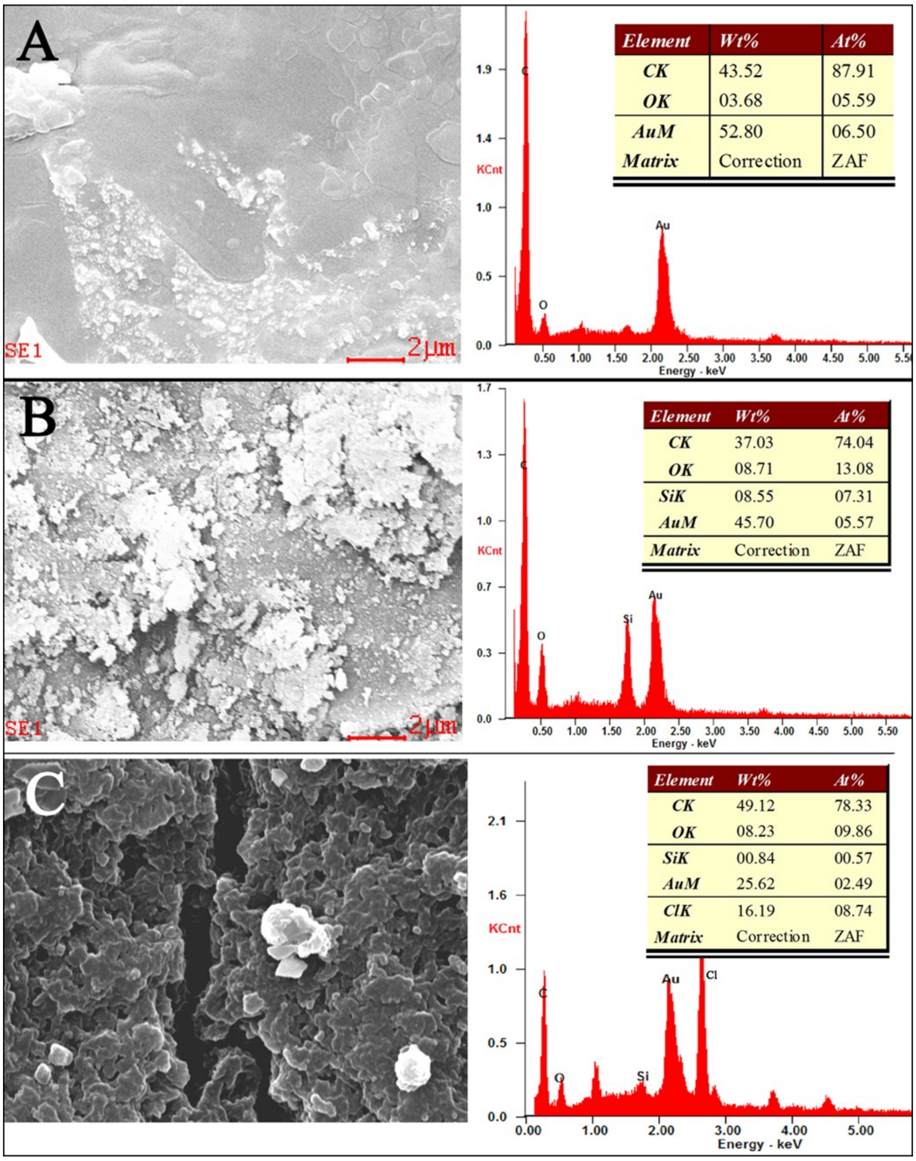

Figure S10. SEM-EDX spectra of (A) native nitrile glove, (B) entry 1 coated glove, (C) entry 1 coated glove after immersed in water for 10 days. The Si content in the native glove can be neglected, and that of aerogel coated glove is 8.55 (wt %), after immersed in water for 10 days, aerogels were detached from the glove by buoyancy, and only 0.84 (wt %) Si was left on the surface.

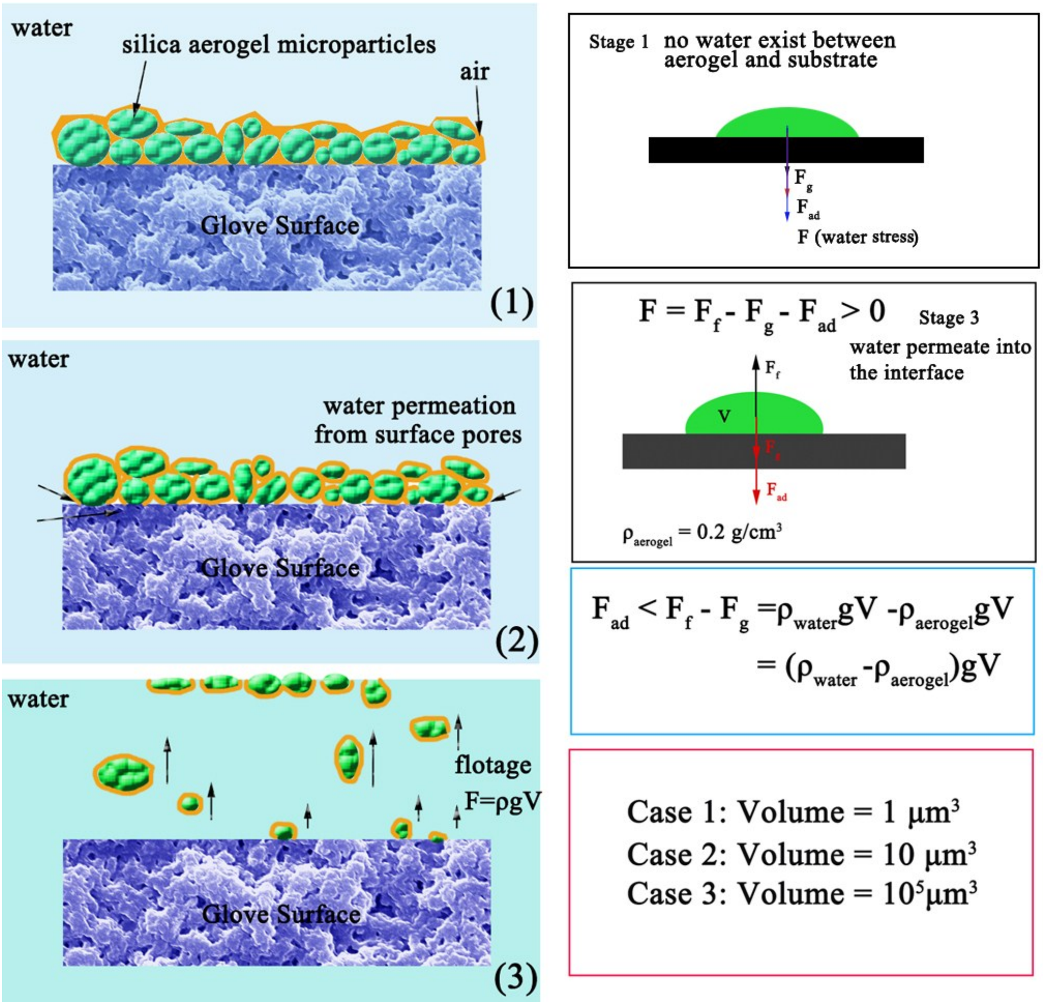

Figure S11. (1) – (3) Show different steps of the proposed mechanism for the detachment of aerogel particles: four kinds of forces were presenting on the aerogel particles, namely buoyancy (F_f), adhesion force (F_{ad}) between aerogel and the surface, water stress (F_{ws}) when there is no water exist in the interface, and its own gravity (F_g). Thus, as shown in the figure, when F_f - F_g - F_{ad} > 0 can the aerogels detached from the surface. At the early stage (1), there was no F_f exist on aerogels because no water exist between the interface of aerogel and glove surface. Then (2) water became permeate into the interface of aerogel and glove surface, but the volume exposed to F_f is small. With time goes by, (3) more and more water permeate into the interface and F_f is higher than F_g + F_{ad} (when the water permeate into the interface, water stress may be disappeared), and the aerogels became detached. For calculated values, we proposed three cases with volume of 1 (length × width × height=1×1×1), 10 (10×1×1), and 10^5 (100×100×10) μm^3 that the aerogel became detach, and the corresponding F_{ad} must be < 8 $\times 10^{-13}$, 8 $\times 10^{-12}$, and 8×10^{-10} N, respectively. In fact, when the volume of aerogel or their pack is 1 and 10 μm^3, they do not detach from the substrate. When the volume of aerogel reached to 10^5 μm^3, aerogel became detached. Thus, the F_{ad} might be in the range of 10^{-9} to 10^{-10} N and the value is in the range of Van der Waals force.

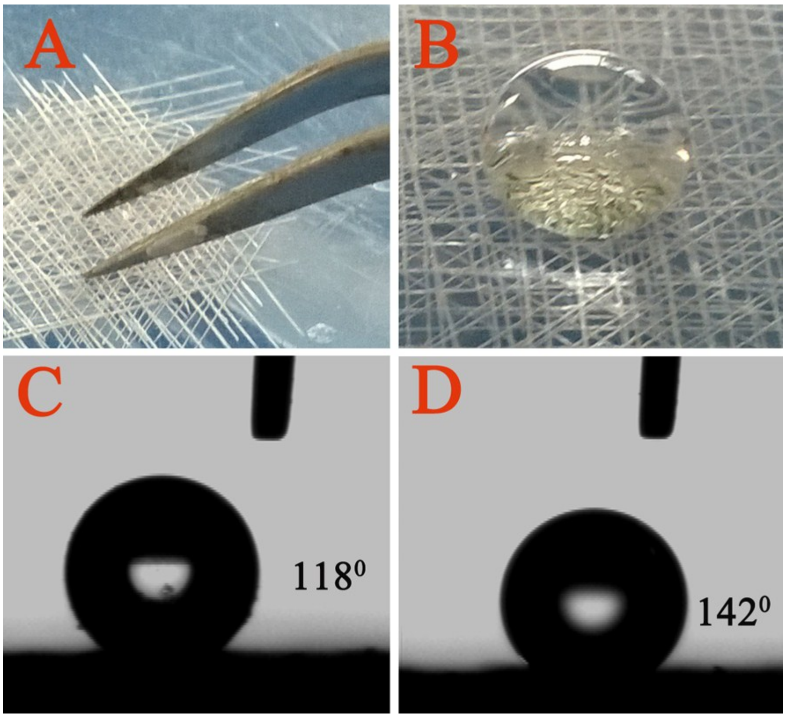

Figure S12. (A) Mechanical contact of the aerogel coated surface, crushing 10 times. (B) Photo image of water droplet on surface after mechanical crushing. Contact angle of aerogel coated surface after mechanical crushing: (C) slightly roughened surface, and (D) highly roughened surface shown in B.

Figure S13. Photo images of (A) invisible glove washed by running water, and (B) after 15 min washing, the invisible glove is still works as the formation of water droplet on the hand.

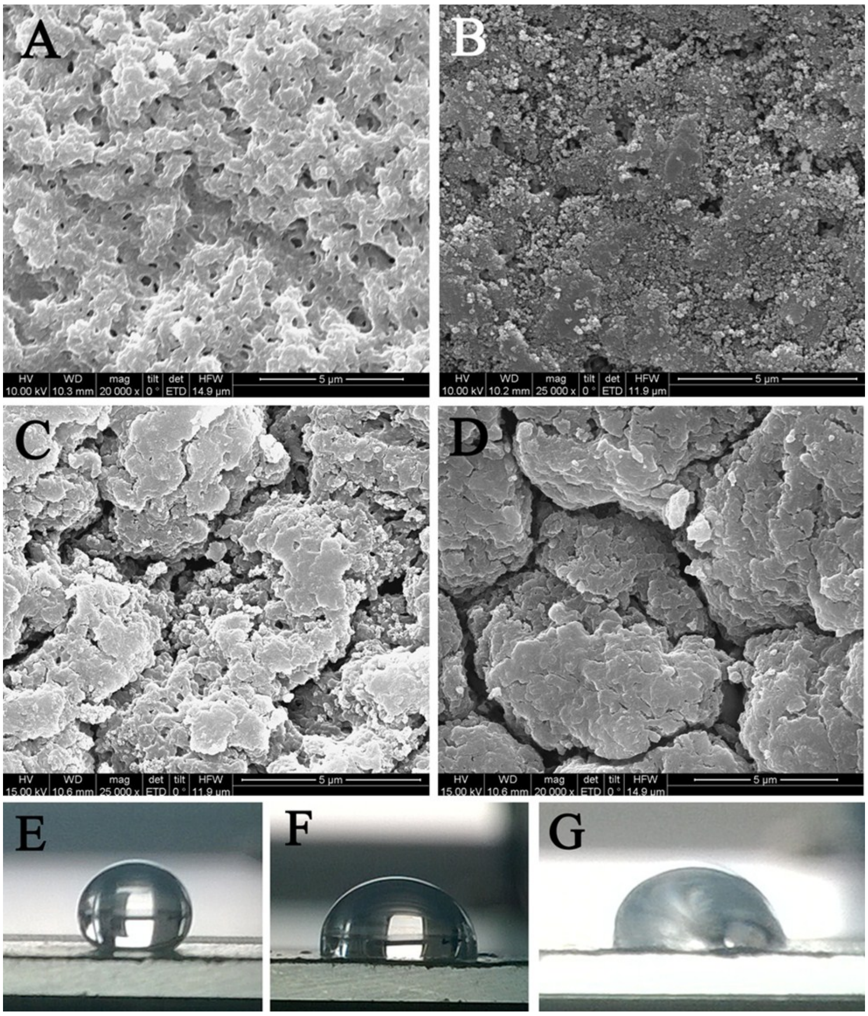

Figure S14. SEM images of (A) native nitrile glove, (B) aerogel coated surface, (C) the surface of B teared by adhesive tape for one time, (D) the surface of B teared by adhesive tape for two times. Water droplet on (E) aerogel coated surface, (F) teared by adhesive tape for one time, and (G) teared by adhesive tape for two times.