Supporting Information

Meta-Aerogel Electric Trap Enables Instant and Continuable Pathogen Killing in Face Masks

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Supplementary Methods

Preparation of o-PAN Nanofibers: The typical procedure for preparing carbon nanofibrous membranes was as follows. Firstly, a 12 wt% PAN solution was prepared by dissolving PAN powder in DMF by magnetic stirring for 12 h. The solution was following an electrospun with an applied voltage of 23 kV and a constant feed rate of 1.5 mL h\(^{-1}\) at ambient conditions. The as-spun nanofibers were collected on a grounded nonwoven-covered metallic rotating roller, followed by drying at 80 °C under vacuum for 3 h. Finally, the nanofibers were heated in air to 155 °C for 1 h and followed by the oxidation process at 280 °C for another 1 h with a heat ramp rate of 1 °C min\(^{-1}\) to obtain o-PAN nanofibers.

Bacteria and virus culture and concentration determination: E. coli (ATCC 25922), S. aureus (ATCC 25923), and coliphage (SHBCC D24291) were chosen to carry out the experiments. Both bacteria were incubated in LB broth at 37 °C for 24 h, harvested by centrifugation, washed twice with PBS, and dispersed in deionized water to obtain suspensions with a concentration of ~10^6 CFU mL\(^{-1}\) for the following experiments. The coliphage was grown and amplified with the host E. coli (TG1) in LB broth on a shaker set to 100 rpm at 37 °C for 24 h. The culture was frozen, defrosted, centrifuged, and filtered to obtain phage suspension (~10^9 PFU mL\(^{-1}\)), followed by diluting in deionized water for a contaminated sample with ~10^6 PFU mL\(^{-1}\) of coliphage. For bacterial concentration determination, 100 μL of the eluent samples and their 10-fold diluent were respectively spotted in the four quadrants on LB agar plates and then incubated at 37 °C for 24 h to evaluate bactericidal efficacies by plate count method. The concentrations of virus samples were tested according to a standard double agar layer method.

Long-term bioprotective performances of masks: The mask samples were worn on the head model as shown in Figure 4c and tested for 4 h per day continuously for 7 d to simulate an actual usage pattern. After testing, a 3×3 cm section of the mask sample, which was in close proximity to the “mouth”, was taken for bacterial concentration testing. The cut sample was
immersed into a sterilized centrifuge tube containing 20 mL of PBS and vortexed for 2 min to wash off the bacteria from the mask. Subsequently, the bacterial concentration in the eluate was measured and approximated as the bacterial concentration retained on the mask.

**SEM and TEM observation of microbes:** For SEM imaging, the METs with harvested bacteria after disinfection were fixed with 2.5 wt% glutaraldehyde for 1 h and then successively dehydrated using aqueous ethanol solution with graded concentrations (50, 70, and 90 wt%) and tert-butanol. Ultimately, the aerogels were dried through a freeze-drying procedure. For the TEM imaging, phages were harvested from the effluent by a centrifuge and then resuspended and fixed in 2.5 wt% glutaraldehyde solution. A 10 μL of the samples were pipetted on a TEM grid and dried at room temperature for 15 min. 2% phosphotungstic acid solution was used to stain phage samples for 1 min and then removed and air-dried for TEM characterization.

**Supplementary Discussions**

**Regulation of the 3D network architecture of METs:**

The structural regulation of METs was carried out by adding different CNT content of 40, 60, and 80 wt% of CNFs, namely, MET-40, MET-60, and MET-80, respectively. Figure S1 depicted the SEM images of the as-prepared METs. With a low CNT content of 40 wt%, only a few sheet-like CNT nanonets were observed sparsely anchored on the junctions of the carbon nanofibrous scaffold. The limited amount of CNTs resulted in an insufficient formation of an interconnected self-knotted network structure. However, as the CNT content increased to 60 wt%, the CNTs gradually filled the carbon fibrous frames, yielding a secondary network structure. It can be inferred that the strong π–π interactions between CNFs and CNTs facilitated the attachment of CNTs and reinforced the attractive interactions between adjacent nanotubes. Further increasing the CNT content up to 80 wt% led to the aggregation of CNTs and a dense CNT film formed among the CNFs.
Subsequently, we tested and compared the mechanical properties, filtration performances, and antibacterial activities of these samples with different CNT contents, as shown in Figure S2. Firstly, the aerogels demonstrated favorable mechanical properties at a compressive strain of 60%, with an increase in compressive stress corresponding to higher CNT content. This can be attributed to the formation and expansion of a self-knotted CNT network, gradually filling the nanofibrous framework. Consequently, the aerogel acquired robust structural support and numerous pathways for energy dissipation, resulting in enhanced resistance to compression deformation. Secondly, in terms of filtration performances (under a flow rate of 1 m s\(^{-1}\)), the increased CNT content caused a significant rise in pressure drop, leading to substantial energy loss. On the other hand, the amplification of filtration efficiency was limited with the further increase of CNT content from 60% to 80%. Thirdly, the antibacterial activity of the EDU enhanced as the CNT content increased from 40% to 60%. This can be attributed to the presence of more antibacterial sites (tips of CNTs) on the cell walls, augmenting the acquisition probability of microbes. However, when the CNT content was further increased to 80%, the disinfection efficiency plateaued as the antibacterial sites became saturated.

In conclusion, we opted for an aerogel sample with a CNT content of 60% for subsequent testing and characterization in this study.

**MD simulation details:**

We performed the calculations using the Forcite module as implemented in the Materials Studio software. An equilibrated (298 K) diamond cell with a cubic edge length of twice the cut-off radius of the van der Waals force (12.5 Å) was heated to 5000 K in the NVT ensemble with a total simulation time of 500 ps. After the equilibration of 5000 K, it was rapidly cooled to 298K in 0.001 ps and equilibrated. This annealing treatment of alternate circles of heating and cooling was repeated 3 times. The final structure obtained is the equilibrium configuration of the amorphous CNF (Figure S5). An armchair double-walled CNT (6, 6) (diameter = 8.1 Å, length=24.6 Å, wall separation=3.3 Å) was constructed as a module to decrease the counting
amount (Figure S6). The adsorption behavior of the CNT on the CNF surface was investigated by MD simulations in the NVT ensemble at 298 K using a time step of 1 fs. The temperature was regulated using a Nosé-Hoover thermostat. The COMPASSII force field was used to describe energy interactions. The trajectories of the system (200 frames) were recorded from runs of 500 ps after the geometry optimization.

The quantitative calculation of disinfection efficiency:

Considering that aerogels can efficiently capture bacteria, the concentration of trapped bacteria should be taken into account when calculating the disinfection efficiency of the EDU. To verify the applicability of resuspending the MET electrode into PBS for measuring the bacteria concentration, a controlled experiment was conducted. Firstly, the bacteria concentration in the exhaust PBS (C₁) was measured without employing the EDU in the chamber. Then, with the EDU in use, we measured both the bacteria concentration in the PBS (C₂) and METs served as positive and negative electrode respectively (C₃⁺ and C₃⁻). The operation time was fixed at 5 min without voltage applied. It was found that C₁ ≈ C₂ + C₃⁺ + C₃⁻. As a result, the calculation of disinfection efficiency should include the quantitative assessment of bacteria killed on the electrode.

The simulation for disinfection mechanism:

The motion behavior of bacteria was simulated using finite element simulation under the coupling fields of ‘creeping flow’, ‘electric currents’, and ‘particle tracing for fluid flow’. A simplified two-dimensional model of tortuous channels profile was constructed, based on the microscopic structure of METs (Figure S11). The thickness of the cell wall was enlarged to 1 μm, in order to be visible, and the interval was 20 μm. The dots in the middle of the channels represented the CNFs that support the layers. An electrostatic potential (10 V) was applied to the cell walls and CNFs, while the top boundary was treated as the ground. Other boundaries were set as electric insulation. The bacteria were simplified as particles that carried 100 elementary charge units (1.6×10⁻¹⁷ C). The air flowed into the top of the simulation domain.
and flowed out through channels at an initial speed of 1 mm s\(^{-1}\). The stationary study aimed to show the inhomogeneous distribution of the electric field and the velocity distribution of the flow, whereas the time-dependent study aimed to trace the particles (Movie S3).

Verification of disinfection mechanisms:

During the electroporation process, the formation of locally concentrated charge density at the sharp tips of CNTs allowed the migration of free electrons and promoted the electrochemical redox for the in situ generation of reactive species.\(^1\)\(^2\) Electrochemical oxidation was evaluated by adding radical scavengers to eliminate the pathogen-killing effects of generated oxidative species (mainly \(\cdot\text{OH}\) and \(\cdot\text{O}^2^-\) as the DI water used in this work). Isopropanol (IPA; \(\cdot\text{OH}\) scavenger; \(\sim 1\) mM) and benzoquinone (BQ; \(\cdot\text{O}^2^-\) scavenger; \(\sim 1\) mM) were selected and added to the \(E.\ coli\) suspensions, respectively, and included into the bioaerosol generated by the nebulizer.\(^3\) Interestingly, disinfection efficiency remained unchanged after passing through the EDU at a flow rate of 1 m s\(^{-1}\) with an applied voltage of 10 V, indicating the absence of electrochemical oxidation during the EDU disinfection process (Figure S12a). Regarding the mechanical damage and inactivation of microbes caused by sharp tips of CNTs, it is commonly known that a relatively long contact time, typically minutes or even hours, is necessary. To investigate this, we cultured the bacteria with METs for a certain period of time and measured the bacterial concentration in the suspension before and after culture (Figure S12b). After 5 min of interaction between aerogels and bacteria (the running time of the disinfection system), the bacterial concentration in the suspension showed no significant change. Only after 30 min of exposure did a slight decrease occur. It is proved that the direct physical interaction of CNTs in this EDU disinfection process is negligible due to the extremely short contact time (0.016 s).

In summary, grounded upon the microstructure investigations of microbes and relevant experimental verification, the primary cause responsible for microbial inactivation in our EDU is confirmed to be electroporation.
REFERENCES

