# A Novel Carbon Aerogel Enabling Respiratory Monitoring for Bio-Facial Mask

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Supporting Information

## **Experimental section**

#### Materials and chemicals

Bacterial cellulose was supplied by Hainan Laize Biochemical Co., China. The polymerization degree of the bacterial cellulose was around 5000. The potassium hydroxide (>98%, KOH), and thiourea (>99.5%,  $CN_2H_4S$ ) were purchased from Beijing Tongguang Chemical Research Institute, China. Zein protein from corn (95% purity) in this study is purchased from the Sigma-Aldrich Co. Ltd., USA. All other chemical reagents were analytical grade.

#### Preparation of hard carbon aerogels

The raw BC slices (5 cm  $\times$  5 cm  $\times$  0.5 cm) were immersed in 1 M KOH solution at 80 °C for 8 h and then rinsed in deionized water to remove the KOH. The BC slices were physically crushed (12 000 rpm/min) in 100 mL of deionized water before adding 100 mL of acetone. The mixture was maintained for 24 h . Then the BC slices were collected, then rinsed with ethanol alcohol and deionized water, respectively. Then, 3 ml thiourea solution (0.1, 0.5, 1, 2 M) were added into 15 mL BC suspension (11.5 mg/mL) by homogenizing the mixture for 10 min. Subsequently, the obtained dispersions were transferred to desired molds, degassed in a vacuum oven for 4 h, frozen liquid nitrogen (-196 °C) and then freeze-dried at -50 °C. Then, the dried BC aerogels were carbonized under N<sub>2</sub> flow with 5 °C/min to the final temperature in the range of 800 °C and held for 1.5 h. For comparison, pure BC carbon was prepared by the same carbonization procedure which was denoted as BCCA

#### Fabrication of protein nano-fabrics by electrospinning

Preparation of precursor solutions: zein solutions were prepared by dissolving zein protein powder with a solid weight ratio of 20% in ethanol solution (70% ethanol, 30% DI water). The solutions were treated by ultrasonication for 1 h at 90 °C.

Fabrication of protein nano-fabrics: The prepared precursor solutions were loaded into a syringe (Monojet Kendall) with a 21-gauge blunt-tip needle. A high-voltage power source (ES50P-5W, Gamma High Voltage Research) controlled the voltage at 20 kV for electrospinning. The extrusion of the solutions was controlled by a mono inject syringe pump (KD Scientific, KDS-100). The various plates including silica/polyamide as collectors were fixed on a grounded metal cylinder with the spinning rate of 100 rev/min to collect the nanofibers. The distance from the needle to the collector was controlled at 15 cm.

#### Sensor fabrication and tests

The resistive-type sensor was fabricated by sandwiching the composite films between two ultrathin polydimethylsiloxane (PDMS) substrates (30  $\mu$ m) with the Au (50 nm) electrodes, which are deposited on them with the size of 2 cm<sup>2</sup>. And then the foam-like hard carbon aerogels were assembled between the two electrodes using silver paste and annealing at 100 °C for 1 h. The sheet resistance of the sensor was measured using a four-point-probe method (4200-SCS, Keithley).

### Surface and structure characterization

The weight of samples and chemicals were measured using an ultra-micro balance (Mettler Toledo) with an accuracy of 10  $\mu$ g. Then the density (apparent density) of s-HCA were measured according to the ISO standard 845:2006. The compression tests were conducted

using a universal machine (Instron) with a 100-N load cell with a strain rate of 10 mm/min for  $\sigma$ - $\epsilon$  tests and 100 mm/min for cyclic fatigue tests. The microscopic architecture and chemical structure of samples were characterized by SEM (FE-SEM, Supra55, Carl Zeiss) with energy dispersive X-ray (EDX) spectroscopy, a high-resolution transmission electron microscope (HR-TEM, JEM-3010, JEOL), FT-IR spectroscopy (Nicolet 8700), Raman spectroscopy (Ar laser, wavelength: 532 nm, RM2000, Renishaw), thermal gravimetric analysis (TGA, TA-Q500), XPS (ThermoFisher scientific), and a D8 Advance Diffractometer (Bruker) with a Cu K $\alpha$  source. Nitrogen adsorption/desorption isotherms were collected at 77 K on a Micromeritics ASAP 2020 instrument. The specific surface area was obtained by the Brunauer-Emmett-Teller (BET) method. The pore size distribution was obtained from density functional theory (DFT) method. The wetting behaviors were characterized by OCA 15 plus contact angle analyzer.

#### Air-filtration and air-permeability tests

As mentioned in other reported work<sup>1-3</sup>, the particulate matter (PM) with particle size randomly ranging from 0.01 to 10  $\mu$ m were produced by burning incense. The formaldehyde (HCHO) was produced by formaldehyde solution (37% in H<sub>2</sub>O). The concentrations of PM and HCHO were diluted and stabilized in a glass bottle to a measurable level and then tested by a particle counter (CEM, DT-9881) and a formaldehyde meter (Omega, HHAQ-108). The air resistance was measured under various flow rates by utilizing a manometer (UEi, EM201-B). The filtration efficiency ( $\eta$ ) of PMs and HCHO was defined by following Equation (1):

where  $C_i$  and  $C_f$  are the initial and final pollutant concentration, respectively. The quality factor (QF) was determined by Equation (2) and (3):

$QF = -\ln(1-\eta)/\Delta P.\dots$	(2)	)
$QF_{normalized} = -ln(1-\eta)/\Delta P/\rho_a$	(3)	)

where  $\Delta P$  and  $\eta$  are the pressure drop and filtration efficiency of the namo-fabrics. The  $\rho_a$  is the areal density of nano-fabric.

Formaldehyde test gas mixtures were prepared using a formaldehyde permeation tube (Kin-Tek Analytical Inc). The output from the permeation tube was diluted with dry zero air flow to yield a 40 ppbv HCHO test gas mixture that was passed through the nanofiber filter. The filter was held in a Teflon filter holder. After determining HCHO concentrations in the air stream, the filters were put in place and downstream HCHO concentrations were continuously measured in real time using a proton transfer reaction mass spectrometer (Ionicon Analytik) to determine filter collection efficiency. Downstream measurements were made until the filter capacity was exhausted.



**Figure S1.** SEM images of (a) the nanofiber layers and (b) the single nanofiber layer in s-HCA.



Figure S2. The size distribution of carbon crystals in the carbonaceous structure.



Figure S3. (a) Nitrogen adsorption/desorption curves. (b) Pore size distribution.



**Figure S4.** (a) The fitted raman spectra of s-HCA. (b) The fitted XPS spectra of s-HCA. (c) The structural components of BCAG and s-HCA calculated for Raman spectra. (d) The carbonaceous structure of BCAG and s-HCA.



**Figure S5.** The morphologies of s-HCA (a) before and (b) after the sonification ( $\sim 20 \text{ kW/m}^3$ ) at room temperature.



Figure S6. The morphologies of s-HCA before (a and b) and after (c and d) cyclic compression.



Figure S7. (a) Specific signals according to the various words; (b) Long-term cyclic signals of various word.



Figure S8. The (a) airflow resistance and (b) filtration efficiency of zein NF with various areal densities.

	Filtration efficiency of HCHO (%)	
s-HCA	70% ± 1	
Zein NF	$45\% \pm 1$	
s-HCA/zein NF	$72\% \pm 1$	
Water contacting	Ethylene glycol contacting	
(a) (b) 138.5°	(b) 0.1 s 0.3 s	

Table S1. Porosity and corresponding filtration efficiency on formaldehyde (HCHO).

Figure S9. Surface contact angles of s-HCA with (a) deionized water and (b) ethylene glycol.



Figure S10. (a) The morphologies of zein NF and (b) magnified ribbon-like fibers.



Figure S11. The stability of composite air filters in the moisture condition (75%).



Figure S12. Long-term filtration stability of mask on  $PM_{2.5}$  in dry and humidified environment (flow rate: 4 L/min).



**Figure S13.** Compressive performance of s-HCA before and after long-term filtration (flow rate: 4 L/min).

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