

## Supporting Information for

# Fabrication of $\text{CN}_x/\text{C}$ Nanotube Arrays via Chemical Modification in Situ and Application in Electronic Nano-device

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### **Chemicals and reagents**

The aluminum sheets (thickness 0.1 mm; purity 99.99%) were obtained from Xinjiang Zhonghe Limited Corp. (Xinjiang, China). Analytical grade reagents of oxalic acid, phosphoric acid, perchloric acid, hydrofluoric acid, stannous chloride, ethanol, dopamine, polyethylene glycol (2000) were purchased from Sinopharm Chemical Reagent Co., Ltd..

### **Characterizations**

Scanning electron microscopy (SEM) images were acquired on a scanning electron microscope (S-4800 Hitachi, Japan) with a 8 mm working distance and 10 kV accelerating voltage. Laser scanning confocal microscopy (LSCM) images of double-layered PAMs labeled with FITC were obtained on a laser scanning confocal microscope (TCS SP5 C1-Z, Leica, Germany). Excitation wavelength was 488 nm, and emission wavelength was 510~530 nm. X-ray photoelectron spectroscopy (XPS) of

CN<sub>x</sub>/C nanotube arrays were measured using a X-ray photoelectron spectroscopy (K-Alpha, Thermo Fisher Scientific, America).

### ***Fabrication of PAMs and CN<sub>x</sub>/C Nanotube Arrays***

PAMs were prepared by a two-step anodic oxidation technique.<sup>1</sup> Aluminum sheets were cleaned in acetone and then electrochemically polished with perchloric acid/ethanol (1:9 v/v HClO<sub>4</sub>: EtOH) at a constant voltage of 10 V for 10 min to achieve mirror finished surface. The temperature of the electrolyte was maintained at 2 °C. The two-step fabrication approach is schematically shown in **Fig. S1**. Before performing the two-step anodization process, a pre-anodization process was introduced to obtain relatively well organized nanochannels. The pre-anodization process was performed in 0.3 M oxalic acid at 20 °C under the constant voltage of 50 V for 0.5 h (procedures a). Then, PAM was etched in an aqueous mixture solution of chromic acid (1.8 wt %) and phosphoric acid (5 wt %) to remove alumina formed in the pre-anodization (procedures b). The formed concaves are used as the nucleation sites for the following classic anodization process. The first anodization was carried out in the same condition as that in the pre-anodization. The only difference is that anodization time was 2 h (procedures c). Upon completion of the first anodization (procedure c), the fabricated PAM was dipped into the 5 mM dopamine buffer solution (Tris-HCl, pH=8.5) (procedure d).<sup>2</sup> After deposition for 12 h, the membrane was rinsed by de-ionized water. The second anodization (procedure e) was then performed to generate an additional porous layer below the modified porous one and anodization time was 2

h. Finally, free-standing PAM was formed by removing the remaining aluminum layer using  $\text{SnCl}_2$  solution followed by a bottom pore opening step carried out by immersing in 5 wt % phosphoric acid (procedure f).  $\text{CN}_x/\text{C}$  nanotubes in PAMs were fabricated via traditional CVD (procedure g).<sup>3</sup> The PAM was placed face down in a quartz tube heated by a tube furnace. Polyethylene glycol 2000 in a vessel beneath the PAM provides the main carbon source. Ar flowed through the quartz tube at 200 sccm during the preheating stage. When the temperature of the furnace reached 700 °C, the flow rate of Ar was decreased to 50 sccm and maintained for 30 min. For visualization, the as-synthesized CNTs/PAM was then dissolved in hydrofluoric acid, and the CNTs were collected by centrifugation.

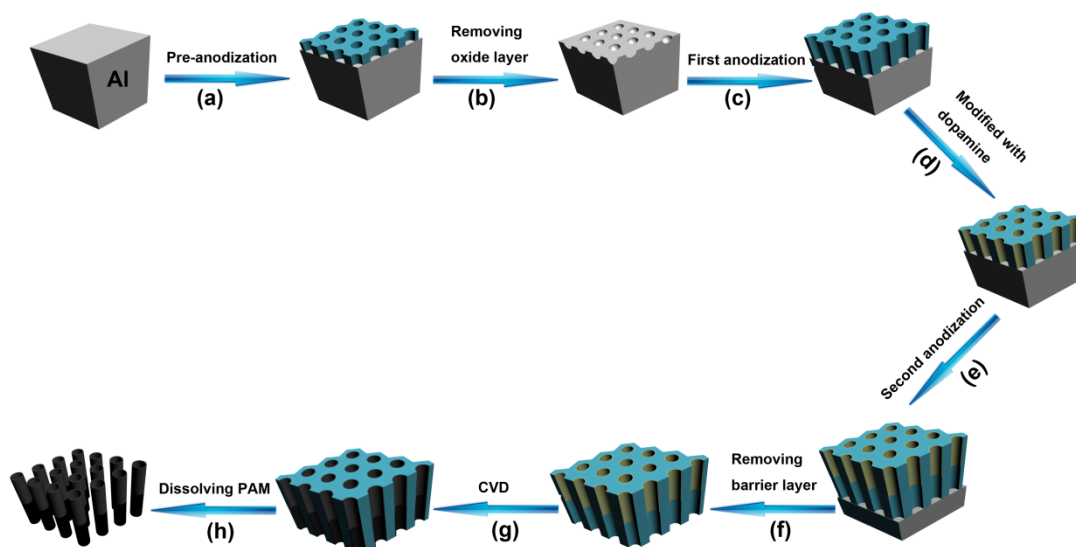


Fig. S1 Schematic illustration of the method for fabricating PAMs and  $\text{CN}_x/\text{C}$  nanotube arrays.

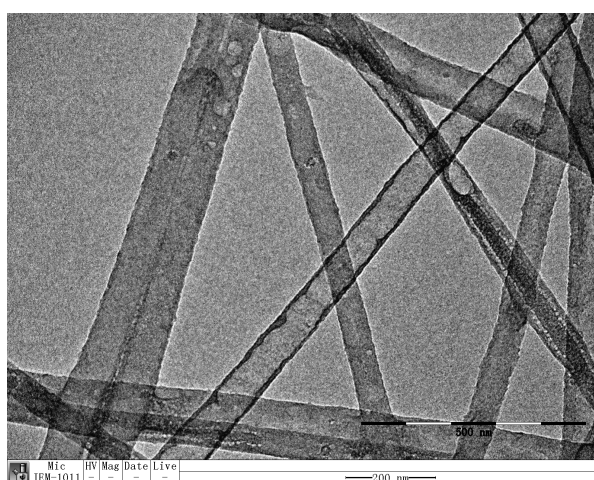
### *Fluorescence labeling*

Fluorescence labeling was performed by treating the membranes with 20  $\mu\text{g}/\text{mL}$  of fluorescein isothiocyanate (FITC) in 100 mM phosphate buffered saline (PBS)

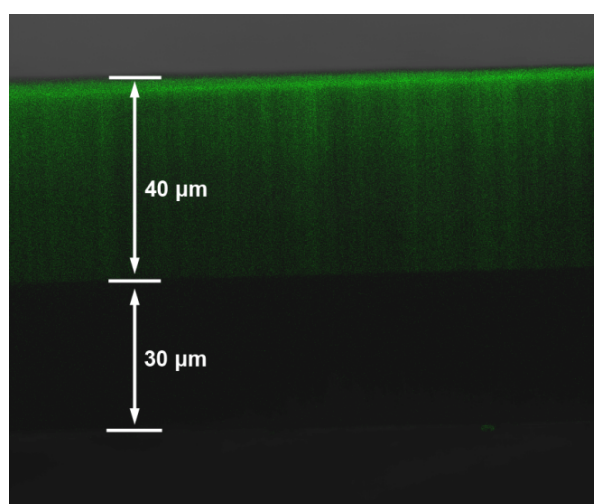
buffer (pH 7.2) at room temperature for 12 h. The unbounded dye was washed off by rinsing with large amount of PBS buffer.

### ***i-V Measurement***

Electronic properties were carried out on a prober station (Cascade EP6, Cascade Microtech, Inc). Linear sweep was measured by scanning the voltage from  $-5$  V to  $+5$  V.



**Fig. S2** TEM image of  $\text{CN}_x/\text{CNT}$  nanotubes after the PAM dissolved completely.



**Fig. S3** Cross-section LSCM image of the PAM fabricated at 50 V in 0.3 M oxalic acid solution. The anodization times for steps (c) and (e) were 4 h.

1. H. Masuda and K. Fukuda, *Science*, 1995, **268**, 1466.
2. H. Lee, S. M. Dellatore, W. M. Miller and P. B. Messersmith, *Science*, 2007, **318**, 426.
3. J. Y. Miao, Y. Cai, Y. F. Chan, P. Sheng and N. Wang, *J. Phys. Chem. B*, 2006, **110**, 2080.