Electronic Supplementary Information (ESI)

Size and density adjustment of nanostructures in nanochannels for screening performance improvement

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EXPERIMENTAL SECTION

Materials. Polydopamine (PDA) and gold (III) chloride hydrate (HAuCl4) are obtained from Sigma Aldrich. AAO membranes with average pore diameters of 85 ± 15 nm and average thickness of 60 ± 5 µm are purchased from Pu-Yuan Nano Technology Co., Ltd. (Hefei China). AAO membranes are prepared by classical anodic oxidation and gentle acidic etching treatment. All other chemicals with analytical grade are provided by Sinopharm Chemical Reagent Co., Ltd. (China) used without further purification. All solutions are prepared with deionized water (resistivity = $18.2 \text{ M}\Omega \cdot \text{cm}$) from a Millipore system.

Preparation of AAO membranes decorated by AuNPs. In a typical process, the AAO membrane was firstly immersed into the HCl solution ($V_{37wt\% HCl} : V_{H2O} = 1 : 2$) with ultrasonic treatment for 2 minutes, then washed with deionized water for 3 times and dried under nitrogen gas. The purified AAO membrane was impregnated with the PDA aqueous solutions with 2 mg·mL⁻¹ by using suction filtration with different filtration duration. Then, the as-prepared AAO membrane was submerged into the HAuCl₄ (1 mM) solution and undergone an ultrasonic treatment for 5 minutes, and followed by laying aside for 30 minutes. After this processing, AuNPs were synthesized in the inner surface of AAO channels by the adhesive and reducing property of PDA. The resultant sample was washed with deionized water for 3 times and dried by nitrogen flow. The ambient temperature of the whole experiment process was 25 °C. Special note: In this paper, the statistics for the size, number and distribution of AuNPs was

manually summed by using the "Nano Measurer" software developed by Fudan University.

Changing the PDA concentration. The specific experimental steps are the similar as above-mention process in the second section. Just changing the PDA concentration as 0.2, 2 and 20 mg \cdot mL⁻¹ respectively, with fixing the other factors.

Regulating the ambient temperature. The specific experimental steps are the similar as those described in the second section. The difference is that the PDA concentration remained at 2 mg mL⁻¹, and the ambient temperature of the whole experiment process was hold as 5, 15, 25 and 35 °C, respectively.

Changing the filtration duration. The same experimental steps as those described in the second section. Only kept the same PDA concentration as 2 mg mL⁻¹, and plus an operation, just after the AAO membrane was coated with PDA by the suction filtration, hold suction filtration for another period as 0, 30 minutes and 2 hours respectively.

Changing the hydrochloric acid concentration. The specific experimental steps are the same as those described in the second section. Just kept the same PDA concentration as 2 mg mL⁻¹, and regulate the HCl concentration which be used to the pretreatment for AAO membrane, as the $V_{37wt\%HCl}$: $V_{H2O} = 1 : 2$, $V_{37wt\%HCl}$: $V_{H2O} = 1 : 10$ and $V_{37wt\%HCl}$: $V_{H2O} = 1 : 20$.

Using EIS to characterize the variation of steric hindrance in AAO channels. The electrochemical characterization of the AAO and its derivative products were performed by using two-electrode cells in a 10 nm tris solution (pH=7.4, 500 mM NaCl,

1 mM MgCl₂) as electrolytes. The symmetric Ag/AgCl electrodes were used as working and counter electrode. Solartron electrochemical station (Solartron Analytical) was used to measure the electrochemical responses. EIS measurements were performed in a frequency range from 1 MHz to 10 MHz at the open circuit potential with an AC perturbation of 5 mV. Before EC measurement, each dry membrane was immersed in 10 nm tris buffer overnight to reduce the test error coming from inadequate impregnation.

Using UV to characterize the concentration of material in the permeate compartment. Use 10 nM MB or $C_{20}H_{23}N$ as feed solution, then apply a +2 V drive voltage to the permeate and the feed compartment, and the directionality drive lasted 40 min. Test the UV absorption of MB or $C_{20}H_{23}N$, and use standard concentration curve of MB or $C_{20}H_{23}N$ solutions to calculate the absorption values.

Conditions	PDA concentration (mg·mL ⁻¹)			Ambient temperature (°C)			Filtration duration (min)			HCl concentration $(V_{37_{Wt}\% HCl}:$ $V_{H2O})$		
PDA concentration	0.2	2	20		25			0			1:2	
Ambient temperature		2		5	15	25		0			1:2	
Filtration duration		2			25		0	30	120		1:2	
HCl concentration		2			25			0		1:20	1:10	1:2

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Table S1. The experimental conditions.

Table S2. Sequences of oligonucleotides.

Туре	Sequence			
Sequence 1	5'(SH)-TTTTATAAGTTTGGTGGTTAGAGAGTG-3'			
Sequence 2	5'-TTTTATAAGTTTGGTGGTTAGAGAGTG-3'			
PM	3'-C ₁ ACTCTCTAACC ₁₂ ACCAA ₁₇ -5' (Perfect match)			



Figure S1. SEM images of Au-decorated AAO membrane from sectional view. (A) is the SEM images of the top picture for Au-decorated AAO membrane; (B) is the SEM images of the buttom picture for Au-decorated AAO membrane; (C) is the SEM images of Au-decorated AAO membrane from sectional view, and all AuNPs in the SEM image are punctuated by red circles.



Figure S2. SEM images of AuNP-decorated AAO membranes from sectional views. A) Schematic illustration for the PDA sucking filtration through an AAO membrane. B) Typical SEM images with different magnifications (intercepted from c-III images in Fig. 1C). C) SEM images of the corresponding regions from I (top) to V (bottom) shown in Fig 1A with a scope of 15 μ m × 14 μ m (named as *x*-*I*, *x*-*II*...), where x represents the PDA concentration: (a) 0.2, (b) 2 and (c) 20 mg·mL⁻¹. The I-V array is in accordance with the direction of suction filtration for PDA. All SEM images in Figure R1C with a same scale bar of 4 μ m. AuNPs in SEM images are marked by red circles.



Figure S3. (A) Schematic illustration of AuNPs by regulating the PDA concentration. (B-E) the AuNP size of the AAO membranes decorating AuNPs under different PDA concentrations corresponding to (B) $0.2 \text{ mg} \cdot \text{mL}^{-1}$, (C) $2 \text{ mg} \cdot \text{mL}^{-1}$, (D) $20 \text{ mg} \cdot \text{mL}^{-1}$ from their statistical results based on ~2000 AuNPs in SEM images, insets are the SEM images of channels where AuNPs are marked by red circles. (E) the statistical plot of the AuNP number density, along the direction of the suction filtration from top to bottom. (F) the statistical results of the size and numbers of AuNPs. The total surface area of AAO inner wall is 8 ×10⁻¹⁰ m², which are calculated by SEM.



Figure S4. (A) Schematic illustration of AuNPs by regulating ambient temperature. (B,C and D) the AuNP size decorating in AAO channels under different ambient temperature corresponding to (B) 5 °C, (C) 15 °C, (D) 25 °C form their statistical results based on ~1500 AuNPs in SEM images, insets are the SEM images of channels where AuNPs are marked by red circles. (E) the statistical plot of the AuNP distribution density, along the direction of the suction filtration. (F) the statistical results of the size and numbers at 8 ×10⁻¹⁰ m² of AuNPs.



Figure S5. (A) Schematic illustration of AuNPs by regulating filtration duration. (B, C and D) the AuNP size decorating in AAO channels under different filtration duration corresponding to (B) 0 min, (C) 30 min, (D) 120 min form their statistical results based on the statistic ~1500 AuNPs in SEM images, insets are the SEM images of channels where AuNPs are marked by red circles. (E) the statistical plot of the AuNP number density, along the direction of the suction filtration. (F) The statistical results of the size and numbers of AuNPs in testing area (8×10⁻¹⁰ m²).



Figure S6. (A) Schematic illustration of AuNPs by regulating HCl concentration. (B, C, D) the AuNP size decorating in AAO channels under different HCl concentration corresponding to (B) 1 : 2, (C) 1 : 10, (D) 1 :20 form their statistical results based on ~1200 AuNPs in SEM images, insets are the SEM images of channels where AuNPs are marked by red circles. (E) the statistical plot of the AuNP distribution density, along the direction of the suction filtration. (F) the statistical results of the size and numbers at 8×10^{-10} m² of AuNPs.



Figure S7. (A) The scheme of two-electrode system in the present work. (B) Top figure is the equivalent circuit used to simulate the impedance spectra; bottom figure is the EIS sketch labeling the impedance elements in different frequency. The Nyquist plots of impedance data for the sequent modification in an AAO membrane. (C) the EIS of AAO membrane decorated with low density of AuNPs by pretreating with low HCl concentration and (D) the EIS of AAO membrane decorated with high HCl concentration. Insets in (C) and (D) are the magnified views of the high and low frequency domain.



Figure S8. Schematic illustration of electrochemical drive unit.



Figure S9. (A) UV absorption curves of MB solutions with different concentrations. (B) Standard concentration curve of MB solutions. (C) The permeate compartment concentration of MB through AAO membrane, AAO-A membrane and AAO-A- $C_{22}H_{46}S$ membrane.



Figure S10. (A) UV absorption curves of $C_{20}H_{23}N$ solutions with different concentrations. (B) Standard concentration curve of $C_{20}H_{23}N$ solutions. (C) The permeate compartment concentration of $C_{20}H_{23}N$ through AAO membrane, AAO-A membrane and AAO-A- $C_{22}H_{46}S$ membrane.



Figure S11. (A) UV absorption curves of FITC-BSA solutions with different concentrations. (B) Standard concentration curve of FITC-BSA solutions. (C) The permeate compartment concentration of FITC-BSA through AAO membrane, AAO-A membrane, AAO-A-D-Cys and AAO-A-L-Cys membrane.