Electronic supplementary information

Interface Nanostructured Array Guided High Performance Electrochemical Actuator

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Experimental

Materials. Natural graphite flake (325 meshes, 99.8%) was obtained from Sigma Aldrich. Potassium peroxydisulfate (97%) and Phosphorous oxide (98%) were obtained from Alfa-Aesa. $EMIBF_4$ (99%) obtained from Shanghai Cheng Jie Chemical Co. Thermoplastic polyurethane (TPU) was obtained from DuPont™. All other chemicals were obtained from Sinopharm Chemical Reagent Co., Ltd and deionized water was purified through Ultrapure Milli-Q system.

Preparation of actuators. GO was prepared by a modified Hummers method which has been described in our recent work.[1,2] The hybrid GO-MWCNTs was synthesized by dispersing MWCNTs in GO solution in ice water bath under 200 W 20 kHz horn sonication treatment in a program of 2 s on and 5 s off for 30 minutes. Then the GO-MWCNTs dispersion was reduced with N_2H_4 in a 90 ℃ oil bath overnight to form RGO-MWCNTs suspension. The obtained RGO-MWCNTs suspension was filtrated with deionized water and re-dispersed in the same sonication program in N-Methyl pyrrolidone forming gel-like suspension of 1 mg/mL RGO-MWCNTs. The as prepared 3 ml suspension was casted on glass substrate (2.5×7.5 cm^2 size) and heated at 80 ℃, obtaining dried RGO-MWCNTs film. Then we used tape to seal the edges of the as-prepared film and placed the film into inorganic salt solution (the molar rate of $NiCl_2$:NH_4Cl:NaOH is 1.15:6:2) and kept in an sealed container at 55 ℃ for 10 h, making $Ni(OH)_2$ vertically grew on the RGO-MWCNTs film. Further put the film into the tube furnace, annealing at 400 ℃ for 2hour with Argon protection and

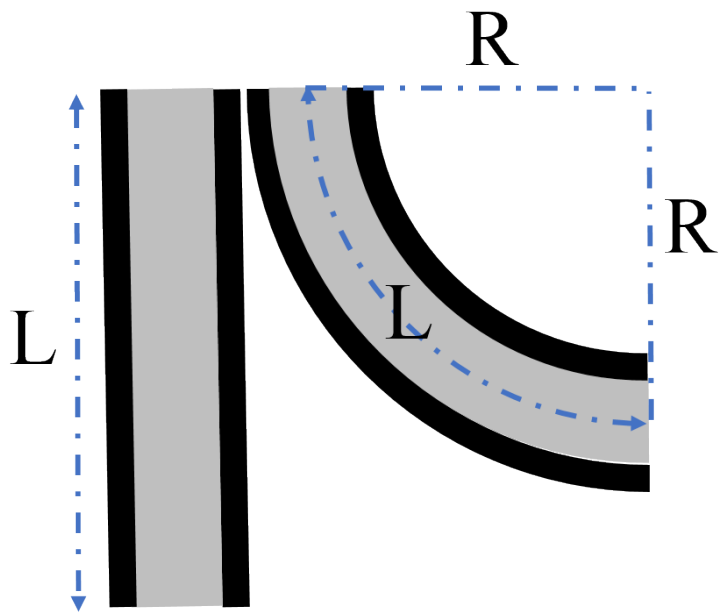
obtained the hierarchically nanocomposite VA-NiONWs@RGO-MWCNTs films. The disordered NiO was synthesized by electrochemical method, which was conducted under potentiostatic control (5 V) for 90 min in inorganic salt solution (the molar rate of $NaCl:NH_4Cl:NaOH$ is 10:1:1.25) and calcined at 400 ^oC for 2 h in air, forming disorder NiO. Then the NiO solution was casted on the on the half-dried RGO-MWCNTs film and heated at 80 ℃for 4 h, obtaining disorder NiO@RGO-MWCNTs film. In addition, 1 g TPU and 2 g $EMIBF_4$ was dissolved in 30 mL DMF in 80 ℃ overnight to obtain uniform solution. 2 mL $EMIBF_4$/TPU solution was casted on a glass substrate (2.5×7.5 cm^2 size) and dried at 80℃ for 3 days. Two piece of electrode films were laminated on TPU/EMIBF4 film and heat pressed at 150℃ for 3 hours and then 170℃ overnight, obtaining a bimorph membrane of about 60 μm thick, 5 mm width and 25 mm long actuator strip.

Actuator performance tests. The 25 x 5 mm sized actuator strips were all performed in a two-electrode configuration with 22 mm free length from electrode contacts. Actuation performance was tested by Multi-point step to the actuator strips using CHI760D electrochemical work station. The displacement (δ) of the actuator was measured by Keyence LK-G800 laser positioning system, where strain (ε), strain rate (ε_r) and stress rate (σ_r) generated in the electrode layers was estimated by the following equations:[3,4]

$$\varepsilon=\delta*d/(\delta^2+L^2),\ \varepsilon_r=4*\varepsilon*f,\ \sigma_r=\sigma*\varepsilon_r$$

Where d, δ, f, L are the thickness, displacement, frequency, free length of the actuator. For testing limited by Keyence LK-G800 laser positioning system, the large displacement can be accurate below 10 mm. So in this study, the displacements above 10 mm were recorded from photography by camera.

Other measurements. Sonication was achieved by using a Fisher Scientific model 500 digital sonic dismembrator equipped with a 12.5 mm diameter disruptor horn. Annealing treatment was processed using OTF-1200 tube furnace. FESEM was recorded by Hitach S-4800. Mechanical tests were performed using an Instron 3365 universal testing machine. All electrochemical characterizations were recorded by CHI760D electrochemical work station. Equivalent circuit analysis of impedance spectra data was carried out using electrochemical impedance software (Scribner Associates, Inc.) and the Chi-squared below 10^{-3}. The actuation process and electrochemical characterization were all performed in a two-electrode configuration. N2 adsorption/desorption analyses were carried out at 77 K using Micromeritics ASAP 2050.



Scheme S1. The relative parameters of a bending graphene strip for curvature calculation. Curvature=1/R.

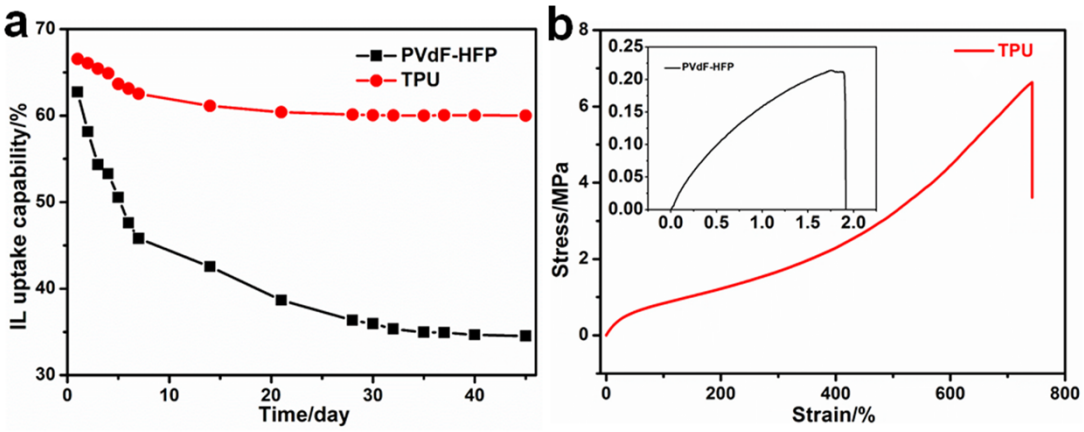


Fig. S1 (a) The uptake capability of ILs in PVdF-HFP and TPU film and the mass ratio of ILs and TPU (PVdF-HFP) is 2:1. (b) The strain-stress curve of PVdF-HFP and TPU supported ILs electrolyte layers.

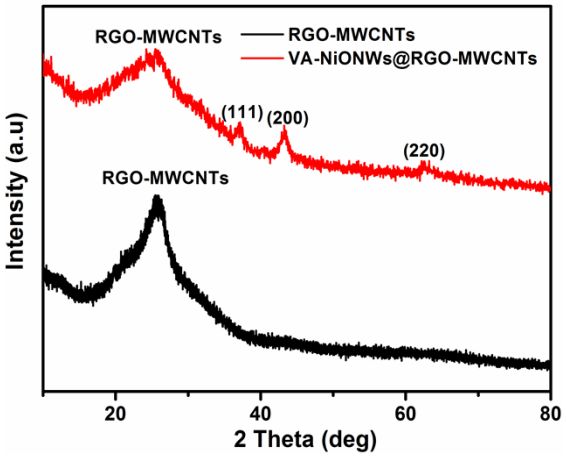


Fig. S2 XRD patters for RGO-MWCNTs and VA-NiONWs@RGO-MWCNTs films, respectively.

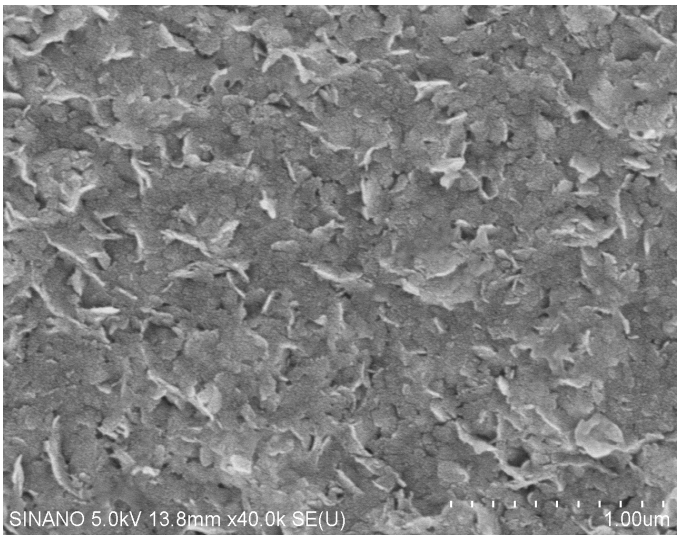


Fig. S3 Surface morphology of electrode. SEM image of disordered NiO@RGO-MWCNTs electrode.

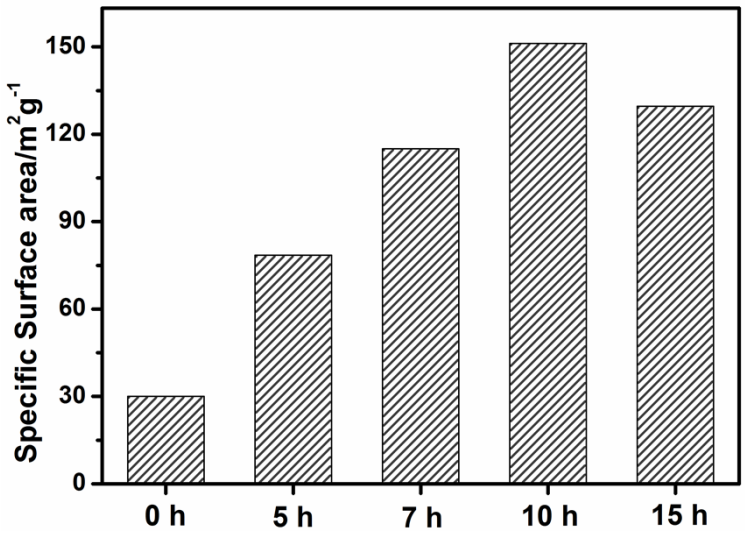


Fig. S4. The specific surface areas of different growth time based VA-NiONWs@RGO-MWCNTs electrode films.

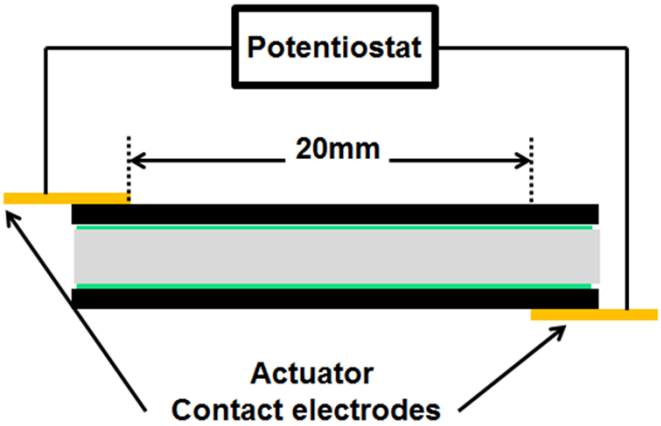


Fig. S5 Illustration for impedance characterization. Illustration for impedance measurement characterization of three kinds of electrodes based actuators.

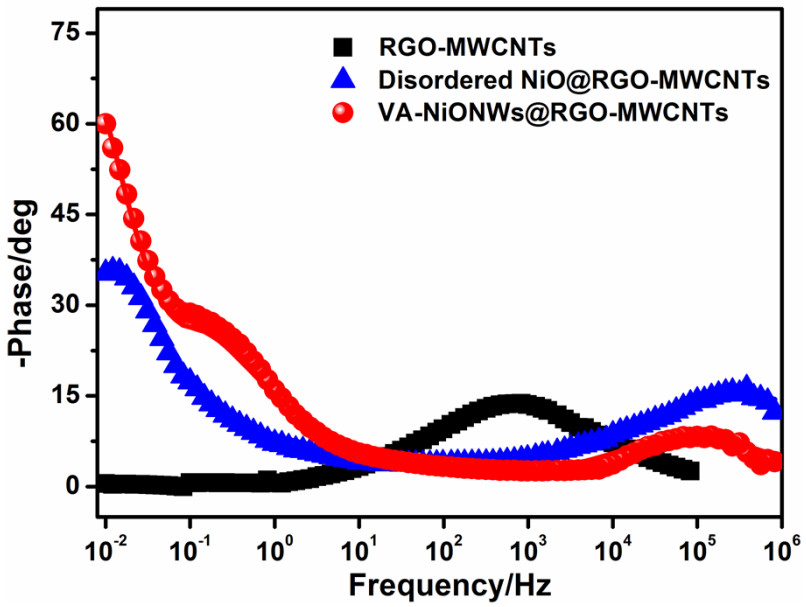


Fig. S6. Bode plots of RGO-MWCNTs, Disordered NiO@RGO-MWCNTs and VA-NiONWs@RGO-MWCNTs based actuators, respectively. Symbols denote experimental data, while the continuous lines represent the fitted data.

Table S1 EIS molding data. Parameter values from curve-fitting of the impedance results shown in Fig.5a by using the equivalent circuit described in inset of Fig. 5a.

	R_0/Ω	R_1/Ω	$Q_1/\mu F\ s^{n-1}$	n	R_2/Ω	Q_2/mF	Z_w
RGO-MWCNTs	207.4	343.5	$3.07x10^{-2}$	0.602	/	/	/
Disordered NiO@RGO-MWCNTs	202.3	545.9	$0.66x10^{-2}$	0.523	3526	6.04	4750
VA-NiONWs@RGO-MWCNTs	287	228.3	$3.35x10^{-2}$	0.59	1320	13.3	2305

References

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