Electronic Supplementary Information (ESI)

Improved Capacitive Deionization by Using 3D Intercalated Graphene Sheet-

Sphere Nanocomposite Architectures

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2. MATERIAL AND METHODS

2.3. Characterization Techniques

The morphology of the all samples were observed by using field emission scanning electron microscopy (SEM, JEOL JEM-700F) and transmission electron microscopy (TEM, JEOL JEM-200CX) at an accelerating voltage of 200 kV. High-resolution transmission electron microscopy (HR-TEM) images were characterized by using JEOL JEM-2010F. X-ray diffraction (XRD) were recorded on Rigaku D/MAX-RB X-ray diffractometer using crystal monochromatic Cu K α radiation. X-ray photoelectron spectroscopy (XPS) data was taken on a Perkin-Elmer PHI 5000C ESCA system equipped with a dual anode Mg Ka (1253.6 eV) X-ray source. Specific surface area and macro and mesopore size distribution was calculated by using Brunauer-Emmett-Teller (BET) with a Micromeritics ASAP 2010 analyzer and Barrett-Joyner-Halenda (BJH) model. Raman spectra were carried out on Raman spectrometer (JY H800UV) (INVIA) with an optical microscope. The surface wettability of the samples was calculated by dynamic contact angle analysis was used to calculate by using (Krüss DSA100). A 1 μ L drop of water with a rate of 1 μ Ls⁻¹ was placed on the surface of electrode and series of images were taken as function of contact time.

2.4. Electrochemical property measurements. The cyclic voltammetry (CV), galvanostatic charge-discharge (GCD) and electrochemical impedance spectroscopy (EIS) were performed in 0.5 mol L⁻¹ NaCl salt solutions by using three-electrode cell configuration that consisted of calomel reference electrode, working electrode and graphite electrode. The working electrodes were fabricated by mixing sample material (90%) and polytetrafluoroethylene (10%) to make homogenous pastes. The slurry was pressed on graphite sheet and then dried at 110 °C for 12 hr. The frequency range for EIS was 10 mHz to10 kHz and the amplitude of the applied voltage was 5 mV for all electrodes. The CV measurement was conducted between -0.5 to 0.5 V and specific capacitance (C) were calculated from the CV results using eqn (S1):

$$C = \frac{\int I dV}{2v\Delta Vm}$$
(S1)

Where C is the specific capacitance, dV is the potential window, I is refer to response current, v is the scan rate and m is refers as the total mass of the sample electrode.

The GCD measurement was carried out by using automatic LAND battery test system in order to assess the charge-discharge ratio.



Figure S1 TEM image of (a) GBs, (b) GSSNA-21 and (c) GSSNA-12.



Figure S2. XRD patterns of the GSSNA/Ni nanocomposites before etching.



Figure S3. (a) XRD patterns and (b) Raman spectra of GSSNA-21 and GSSNA-12.

Samples	BET $(m^2 g^{-1})$	$V_{tot} (cm^3 g^{-1})$	D _{ave} (nm)
GSSNA-11	664	3.227	4.897
GSSNA-21	597	2.208	3.739
GSSNA-12	435	2.295	3.418
GBs	522	1.658	2.393
GR	330	2.536	4.076

 Table S1. Textural properties of various samples.



Figure S4. Nitrogen sorption isotherms and pore size distribution profiles (inset) of GSSNA-21 and GSSNA-

12.



Figure S5. Nitrogen sorption isotherms and pore size distribution profiles (inset) of (a) GBs and (b) GR.



Figure S6. (a) The survey XPS spectra of GSSNA-11; and (b) High resolution C 1s spectra of GSSNA-11.



Figure S7. Dynamic contact angle of GSSNA-11, GBs and GR electrodes as a function of contact time.



Figure S8. (a) CV curves of GSSNA-21 and GSSNA-12 at a scan rate 5 mVs⁻¹ in a 0.5 M NaCl solution; and
(b) specific capacitance of GSSNA-21 and GSSNA-12 at different scan rates.



Figure S9. CV curves of GSSNA-11 at different scan rates from 1mVs⁻¹ to 50mVs⁻¹ in a 0.5M NaCl solution.



Figure S10. iR drop of GSSNA-11, GBs and GR at different discharge current density.



Figure S11. Plots of the solution conductivity vs time for graphite collectors in a 500 mg L^{-1} NaCl solution at 1.2 V.



Figure S12. SAC curves and Ragone plots of GSSNA-11 electrodes (a, b) in different NaCl concentration, (c, d) at different cell voltage; and (e, f) with different flow rates.



Figure S13. Current transient and charge efficiency for GSSNA-11 electrodes in different concentration of

NaCl solution at 1.2 V.

Electrode	Specific capacitance	BET	Cell Voltage	NaCl Conc.	SAC (mg g ⁻¹)	References
materials	(F g ⁻¹)	$(m^2 g^{-1})$	(V)	(mg L ⁻¹)		
Graphene/Metal	119.7	187.60	1.2	500	15.5	1
Oxide Hybrids						
3D Graphene	200	425	1.6	500	15	2
Architectures						
RGO-PPy-MnO ₂	356	331	1.2	500	18.4	3
Graphenic	323	589	1.2	500	13.1	4
Fibers						
N-doped	178	453	1.2	500	13.71	5
Carbon Spheres						
Porous Carbon	129	606	1.2	500	13.86	6
Polyhedra						
Nitrogen-doped	150	813	1.2	500	16.63	7
Porous Carbon						
3D Hierarchical	142	2061	1.2	500	17.83	8
Carbon						
GSSNA-11	249	664	1.2	500	22.09	This work
GSSNA-21	179	597	1.2	500	17.28	This work
GBs	147	522	1.2	500	12.35	This work

 Table S2. Comparison of the electrosorption capacity of various CDI electrode materials.

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