Electronic Supplementary Information (ESI)

Chemically Bonded $NaTi_2(PO_4)_3$/rGO Microsphere Composite as High-Rate Insertion Anode for Sodium-Ion Capacitor

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Supplementary Table

	wt.% of elements from elemental analysis		
$NaTi_2(PO_4)_3$/rGO	Carbon	Hydrogen	Nitrogen
	2.06	0.02	0.11

Table S1. Elemental analysis of a 3D $NaTi_2(PO_4)_3$/rGO microsphere composite.

	R_s	R_{ct}
Fresh cell	8.07	20.85
After 250th cycle	9.48	23.09
After 500th cycle	9.15	22.89
After 1000th cycle	8.95	23.35
After 5000th cycle	8.59	24.36

Table S2. Fitting values of equivalent circuit elements of the $NaTi_2(PO_4)_3$/rGO microsphere composite//AC NHC electrode.

Supplementary Figures

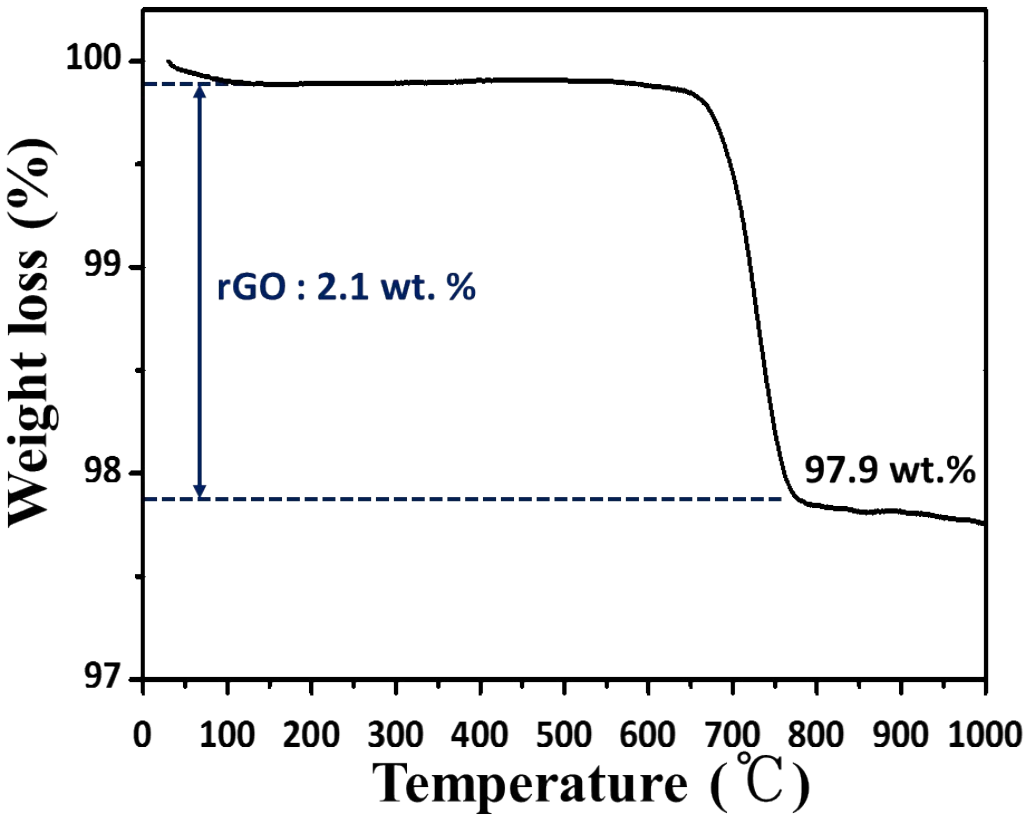

Fig. S1 TGA of $NaTi_2(PO_4)_3$/rGO microsphere composite, heated from room temperature to 1000 °C at 10 °C min^{-1} under air flow. From TGA the loading amount of $NaTi_2(PO_4)_3$ in the $NaTi_2(PO_4)_3$/rGO microsphere composite was determined to be 97.9 wt.%.

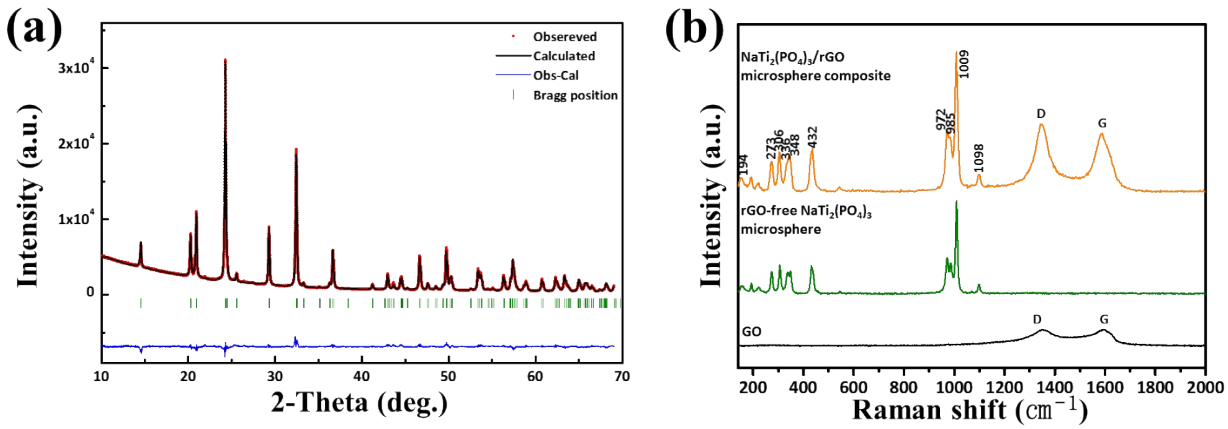

Fig. S2 (a) Rietveld refinement of the XRD pattern of $NaTi_2(PO_4)_3$/rGO microsphere composite with NASICON structure, (b) Raman spectra of GO, rGO-free $NaTi_2(PO_4)_3$ microsphere and $NaTi_2(PO_4)_3$/rGO microsphere composite.

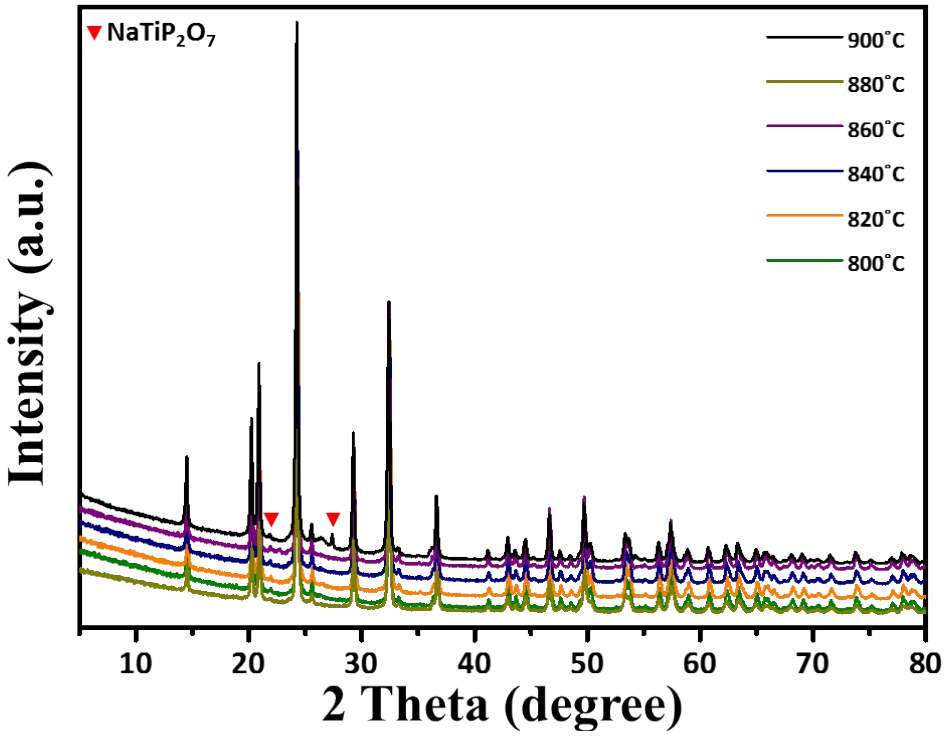

Fig. S3 X-ray diffraction patterns of $NaTi_2(PO_4)_3$/rGO microsphere composite at temperatures of 800, 820, 840, 860, 880 and 900 °C.

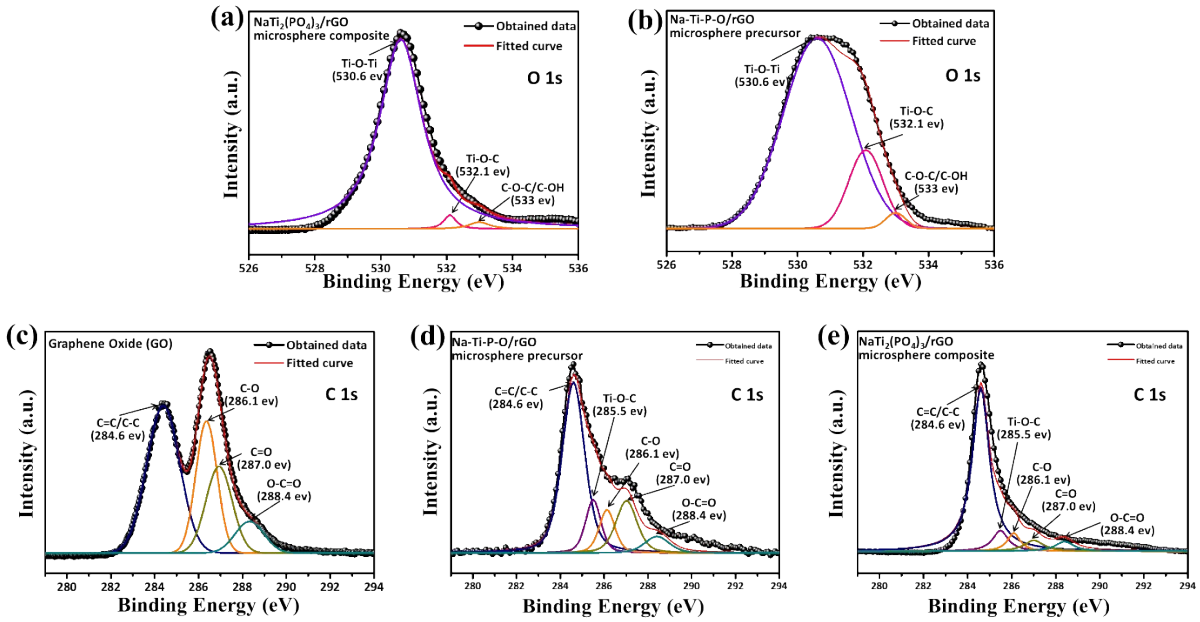

Fig. S4 XPS O 1s spectra of (a) Na–Ti–P–O/rGO microsphere precursor, (b) $NaTi_2(PO_4)_3$/rGO microsphere composite; XPS C 1s spectra of (c) GO, (d) Na–Ti–P–O/rGO microsphere precursor and (e) $NaTi_2(PO_4)_3$/rGO microsphere composite.

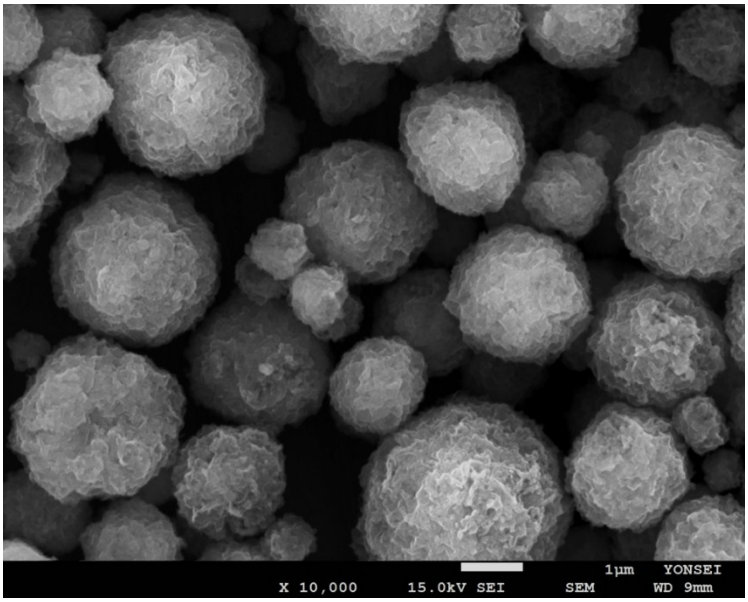

Fig. S5 Low-magnification SEM image of $NaTi_2(PO_4)_3$/rGO microsphere composite.

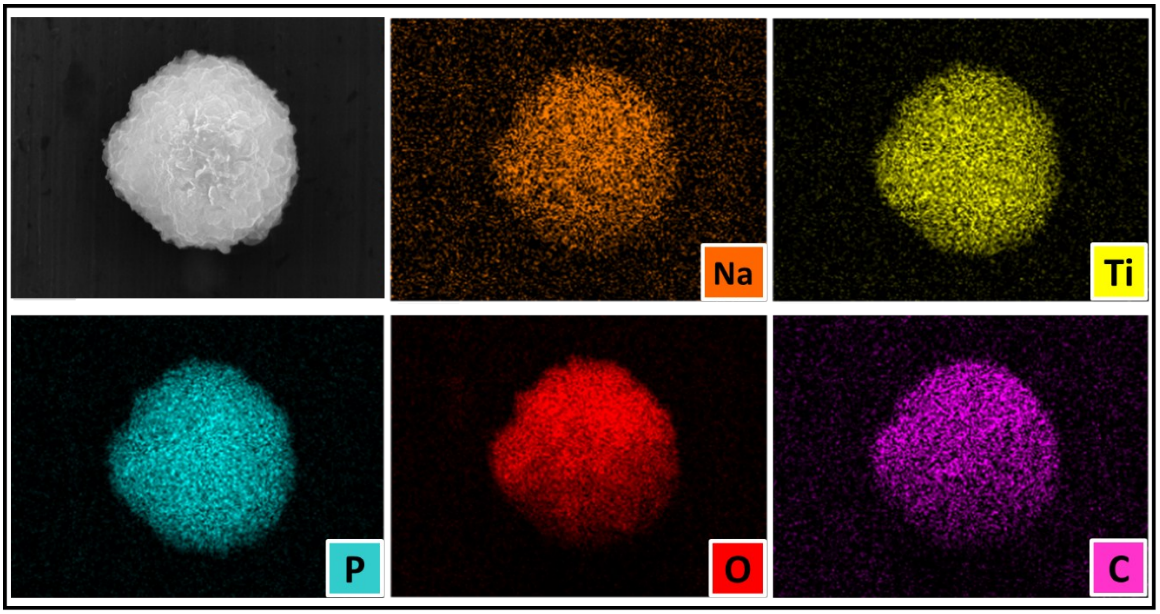

Fig. S6 Typical SEM image of $NaTi_2(PO_4)_3$/rGO microsphere composite and corresponding elemental mapping of sodium (orange), titanium (yellow), phosphate (blue), oxygen (red), and carbon (pink). EDS elemental mapping shows even distribution of Na, Ti, P, and O, suggesting that $NaTi_2(PO_4)_3$ particles are uniformly distributed in the composite.

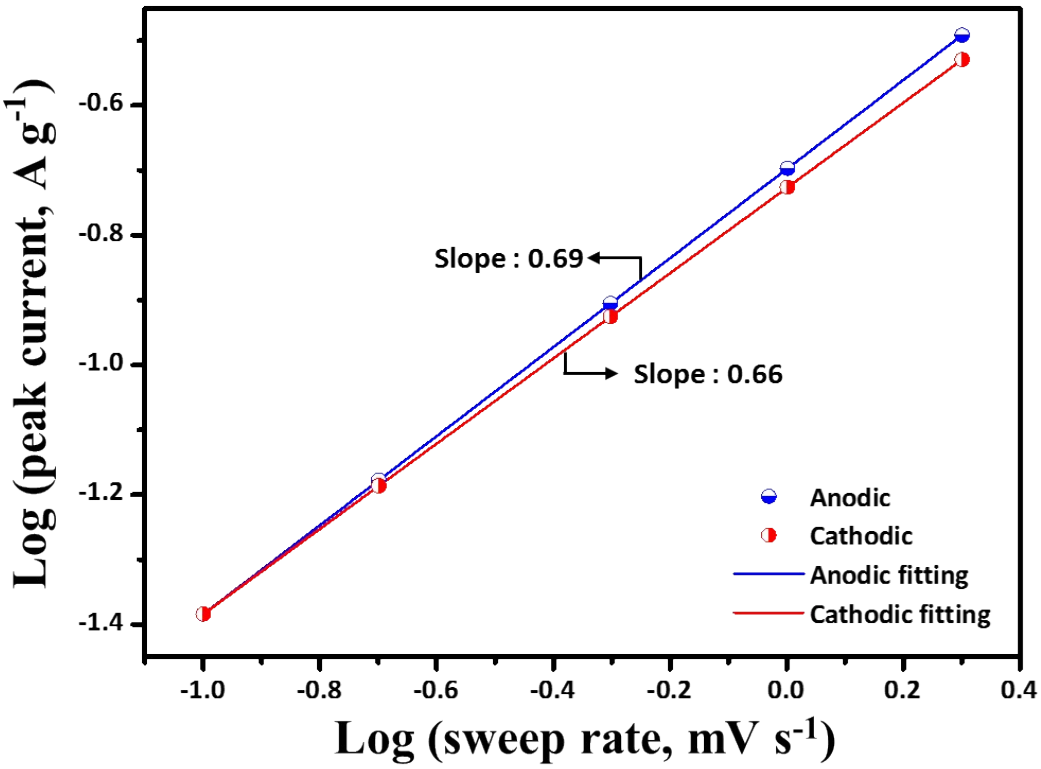

Fig. S7 A plot of log (peak current) vs. log (potential scan rate) for $NaTi_2(PO_4)_3$/rGO microsphere composite.

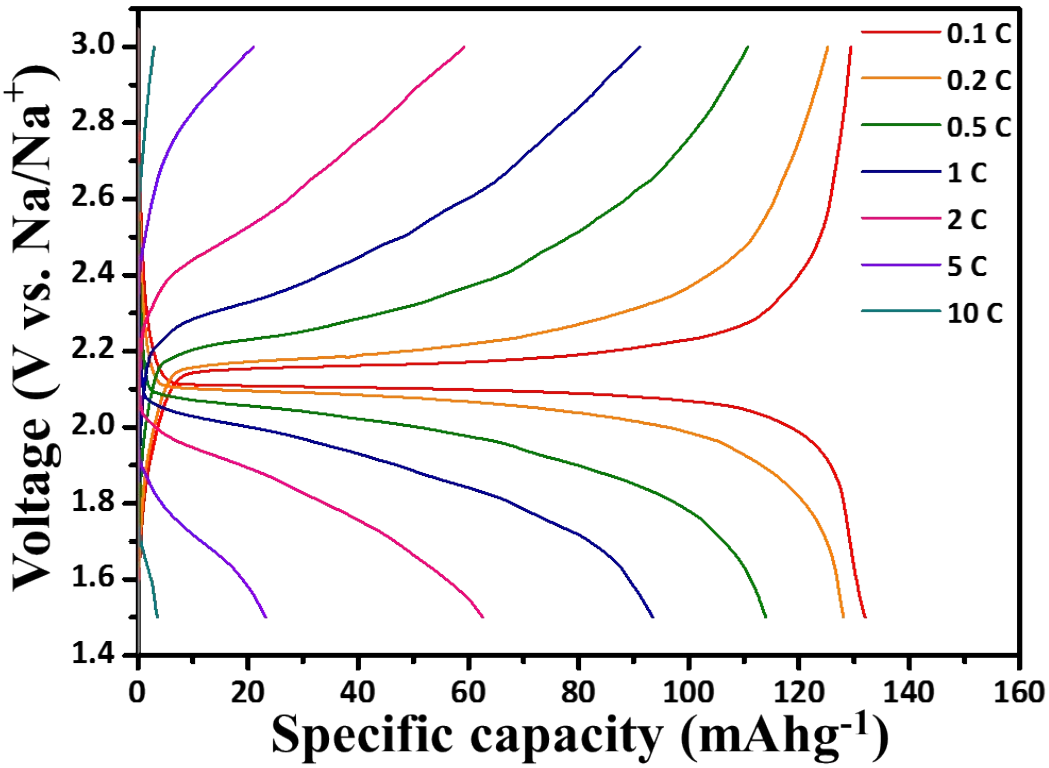

Fig. S8 Discharge/charge profiles of rGO-free $NaTi_2(PO_4)_3$ microsphere at increasing C-rate from 0.1 C to 10 C.

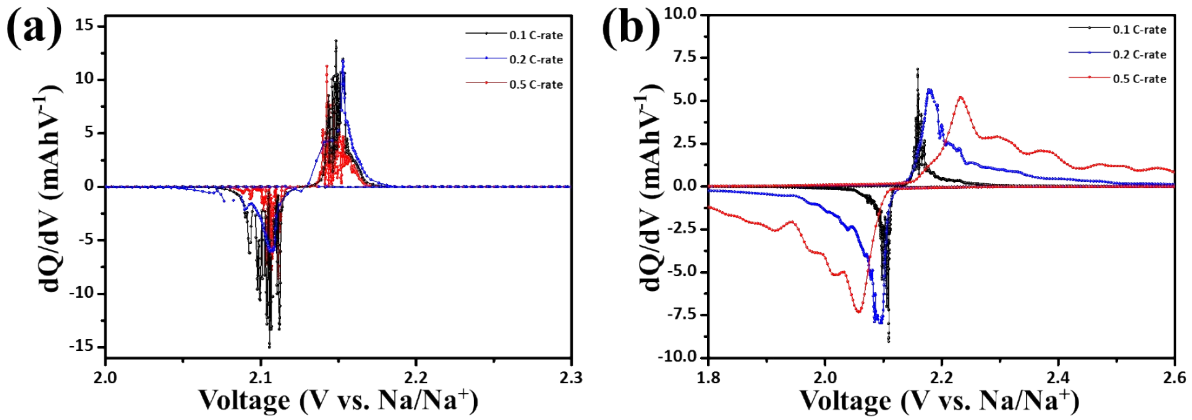

Fig. S9 Differential capacity (dQ/dV) vs. voltage profiles of (a) $NaTi_2(PO_4)_3$/rGO microsphere composite and (b) rGO-free $NaTi_2(PO_4)_3$ microsphere at 0.1, 0.2 and 0.5 C-rate.

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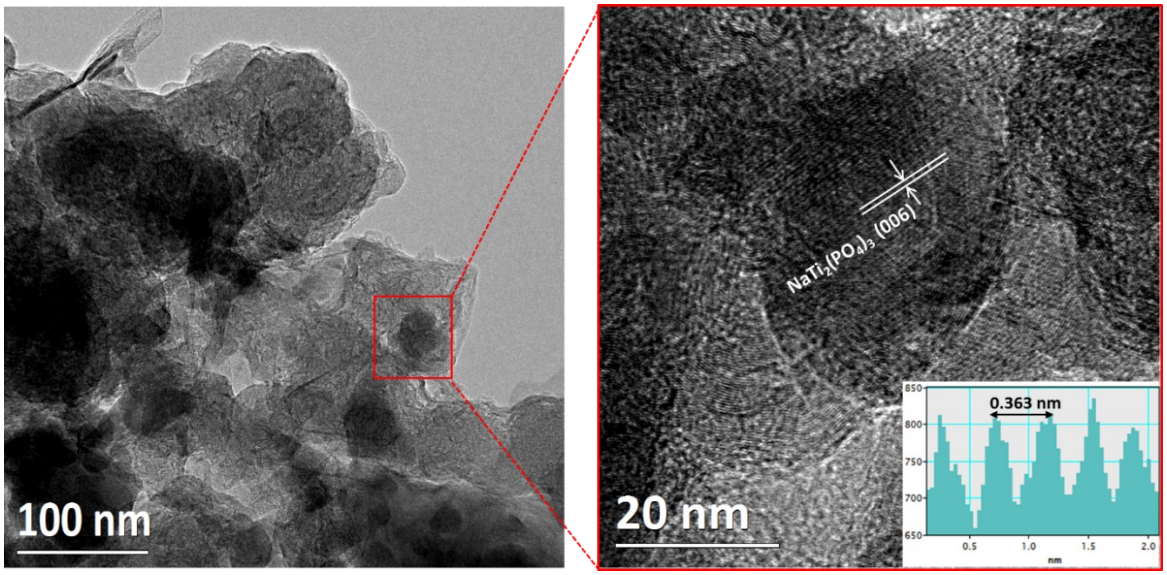

Fig. S10 TEM and HR-TEM images of $NaTi_2(PO_4)_3$/rGO microsphere composite after 200 cycles at 10 C. HR-TEM image shows clear lattice fringes with a d-spacing of 0.363 nm, which corresponds to the (006) plane spacing of $NaTi_2(PO_4)_3$, demonstrating that crystallinity of the $NaTi_2(PO_4)_3$ nanoparticles was well retained in the composite after 200 cycles at 10 C-rate.

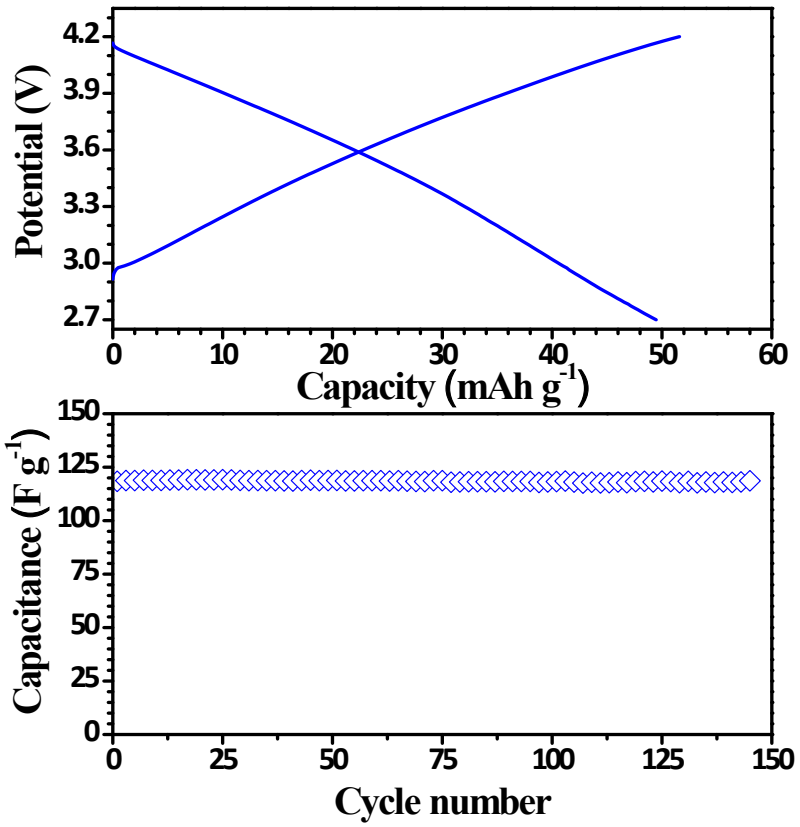

Fig. S11 Galvanostatic charge-discharge profiles and cycling stability of activated carbon (AC) measured between 2.7 and 4.2 V in 1 M $NaClO_4$ in PC with 5 wt.% FEC at a current density of 100 mA g^{-1}.

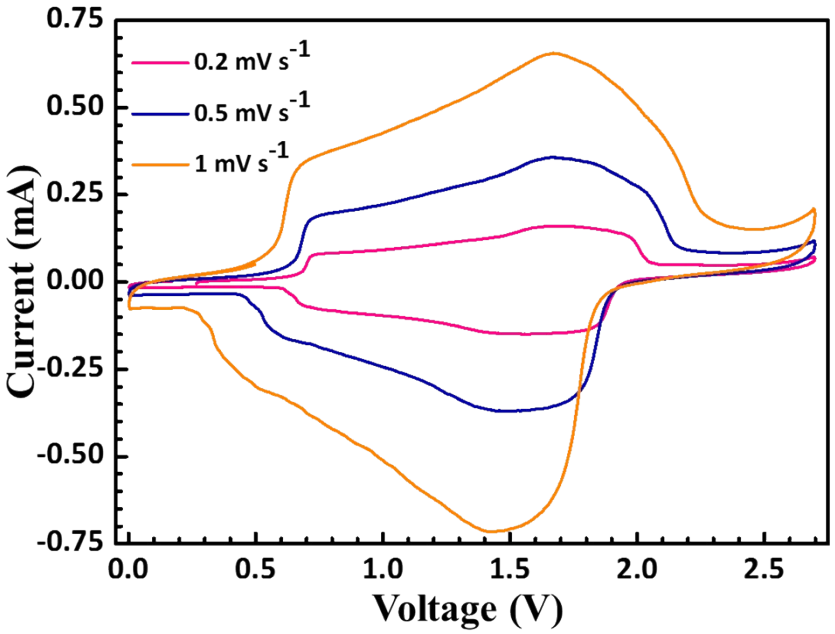

Fig. S12 Cyclic voltammograms of the $NaTi_2(PO_4)_3$/rGO microsphere composite/AC sodium-ion hybrid capacitor (NHC) at various scan rates.