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Binary metal sulfide and polypyrrole on vertically-aligned CNTs array/carbon fiber paper as high-performance electrodes

Xiaoyi Cai,[a] Reinack V. Hansen,[c] Lili Zhang,[b] Baosheng Li,[d] Chee Kok Poh,[b] San Hua Lim,[b] Luwei Chen,[b] Jinglei Yang,[c] Linfei Lai,[a,*] Jianyi Lin,[a] Zexiang Shen[a,*]

a Energy Research Institute @ NTU (ERIAN), 1 CleanTech Loop, #06-04, CleanTech One, Singapore 637141

b Institute of Chemical and Engineering Sciences, A*STAR, 1 Pesek Road, Jurong Island, 627833, Singapore

c School of Mechanical & Aerospace Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798

d Key Laboratory of Flexible Electronics (KLOFE) & Institute of Advanced Materials (IAM), Jiangsu National Synergistic Innovation Center for Advanced Materials (SICAM), Nanjing Tech University (Nanjing Tech), 30 South Puzhu Road, Nanjing 211816, P. R. China

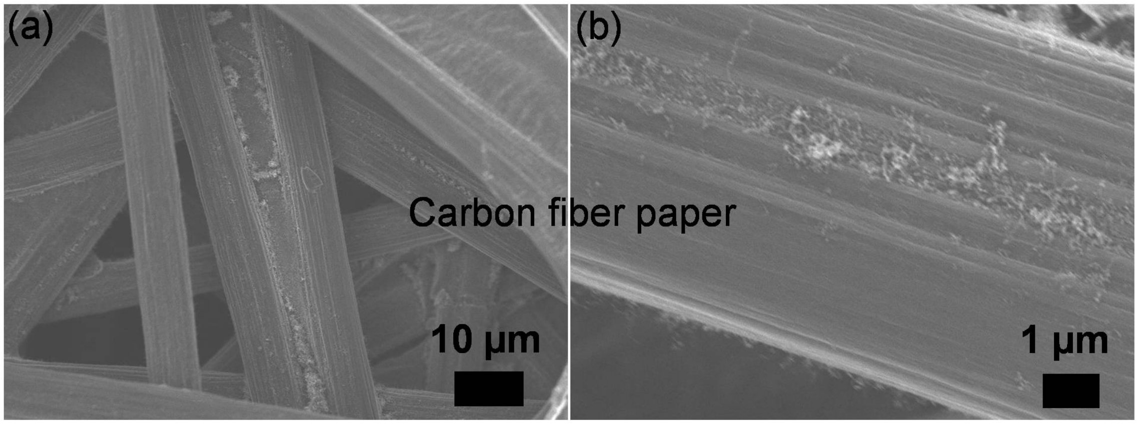

Figure S1: CNTs grown on CFP without coating of Al_2O_3 buffer layer.

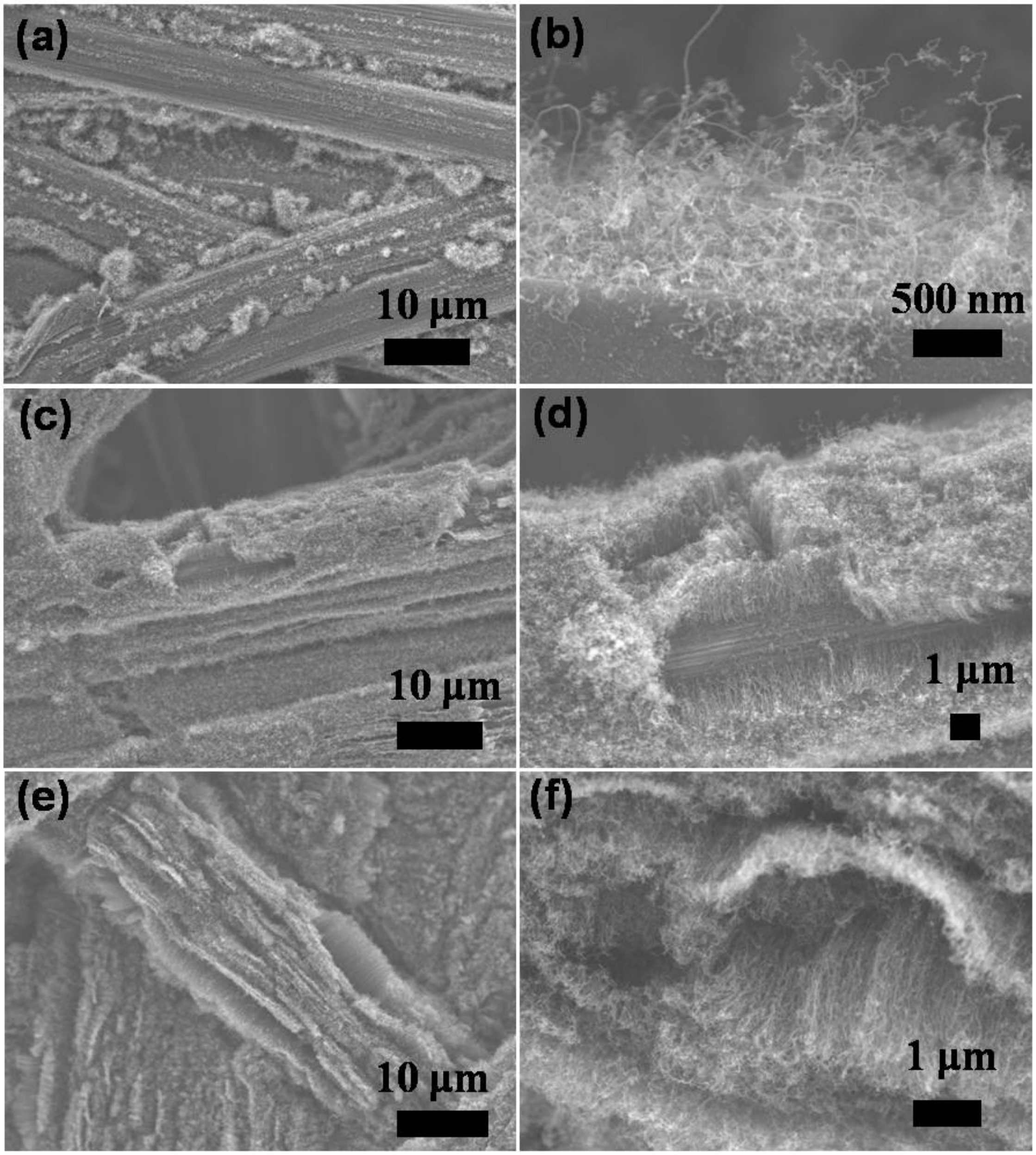

Figure S2: VA-CNTs array grown on CFP with 1 nm (a and b), 3nm (c and d) and 5 nm (e and f) of Al_2O_3 buffer layer.

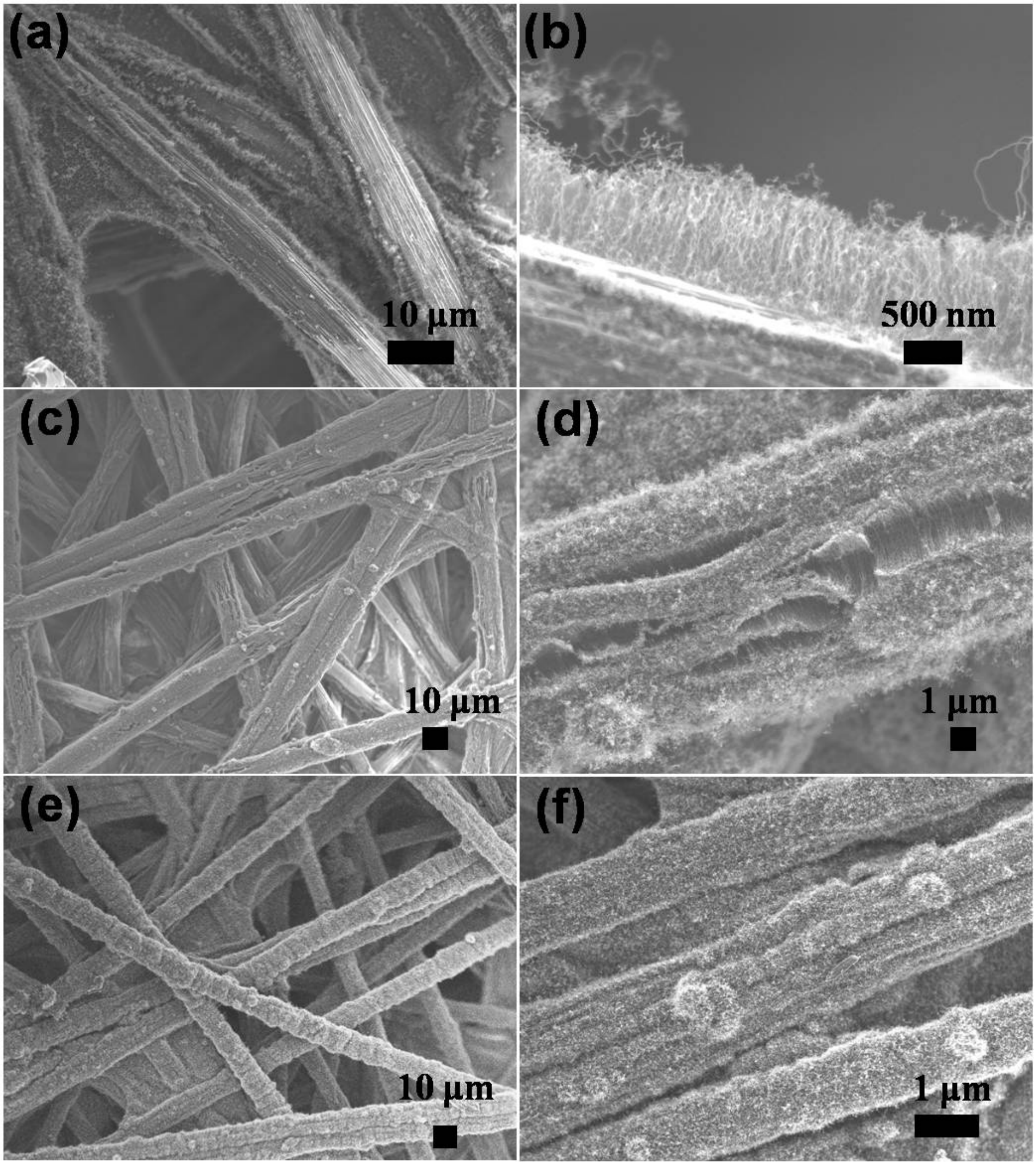

Figure S3: VA-CNTs array grown on CFP with various catalyst loading, from 0.1 (a and b), 0.5 (c and d) and 0.7 (e and f) mg mass per cm^2.

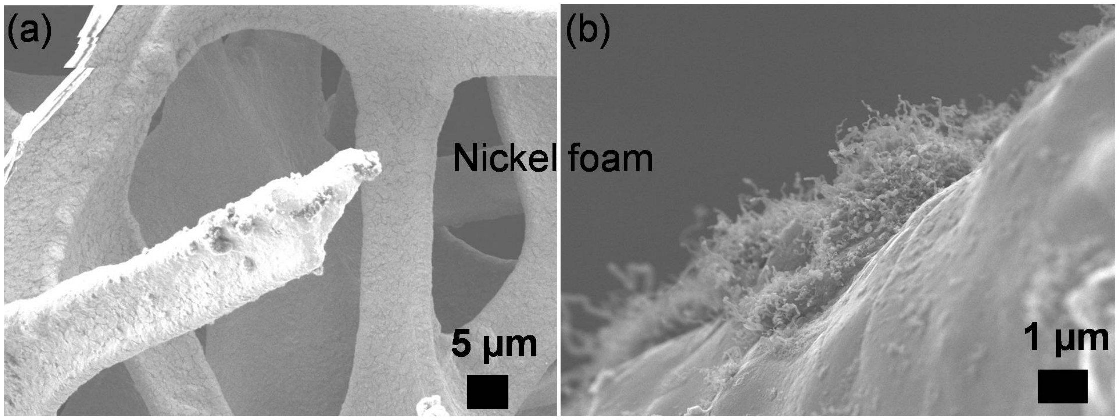

Figure S4: The SEM images of CNTs grown on 3 nm Al_2O_3 layer coated Ni foam with e-beam evaporation of 5 nm of Fe as catalyst.

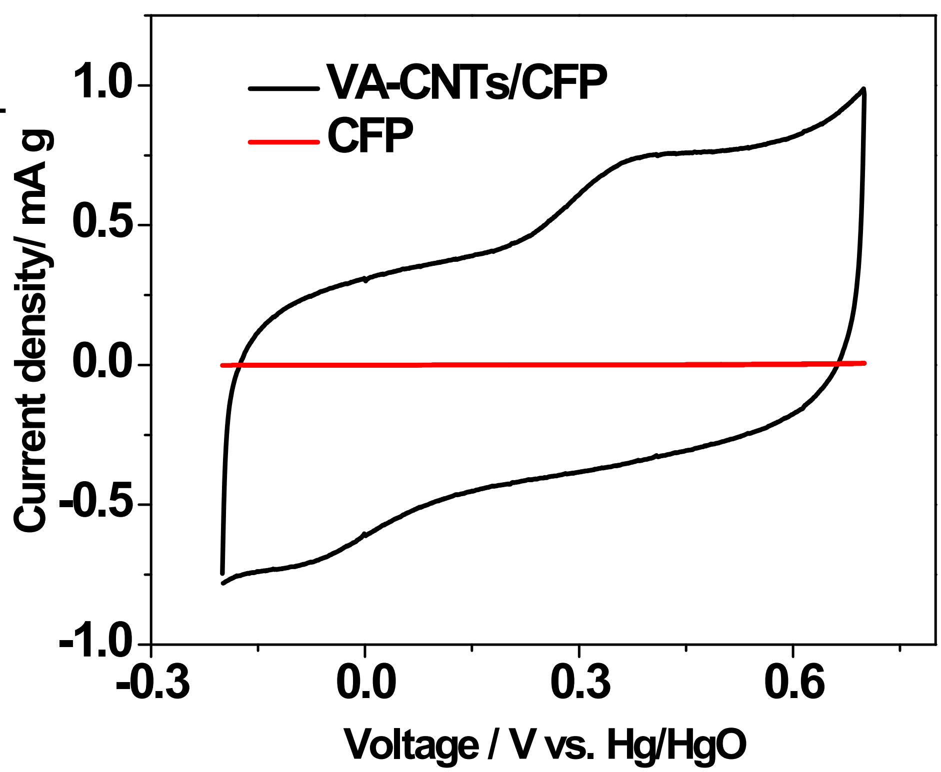

Figure S5: CV curves of CFP (red) and VA-CNTs/CFP (black) in 1M KOH at a scan rate of 50 mV/s.

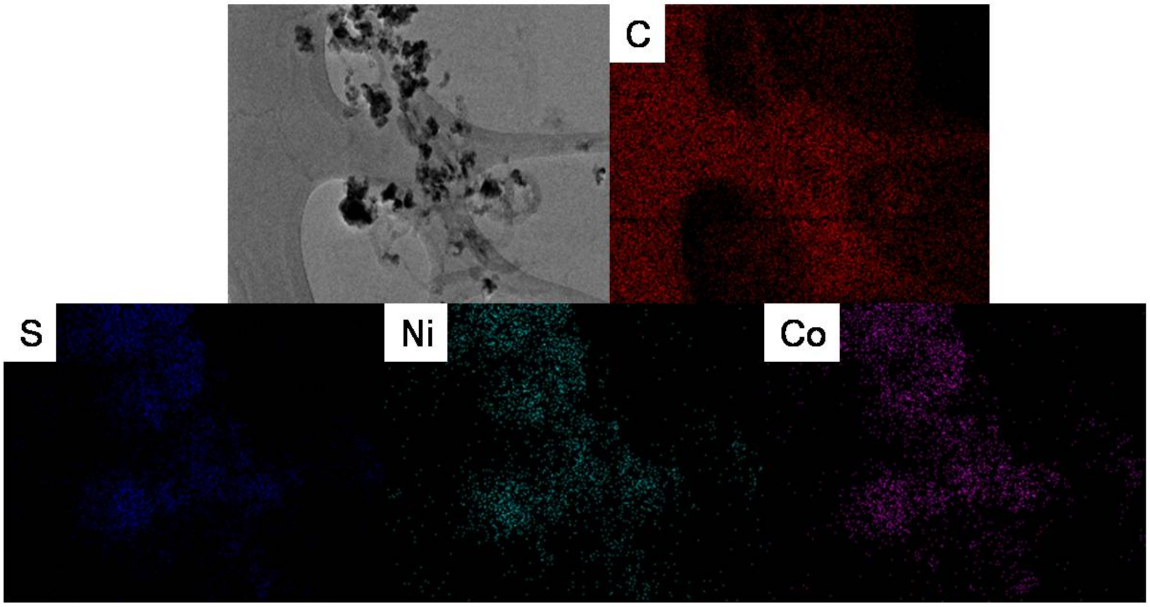

Figure S6: Energy-dispersive X-ray spectroscopy (EDS) elemental mapping of C, S, Ni and Co.

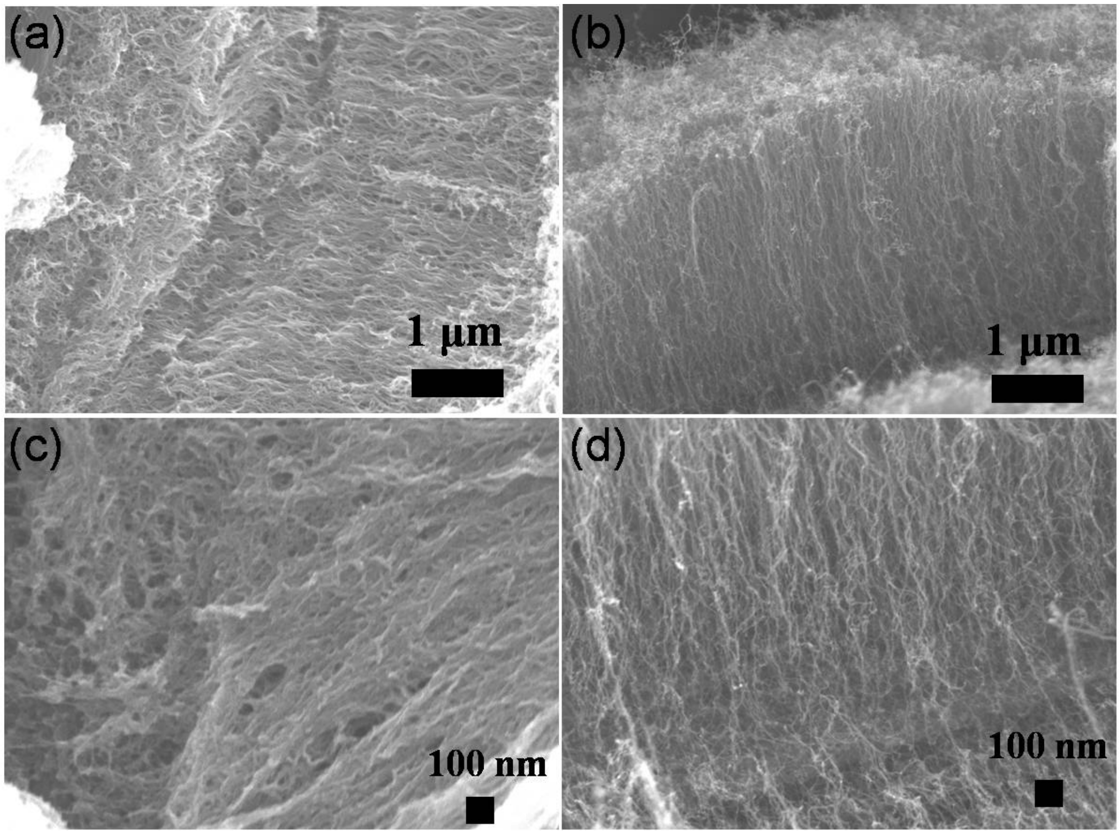

Figure S7: The SEM images of PPy/VA-CNTs/CFP (a and c) and VA-CNTs/CFP (b and d).

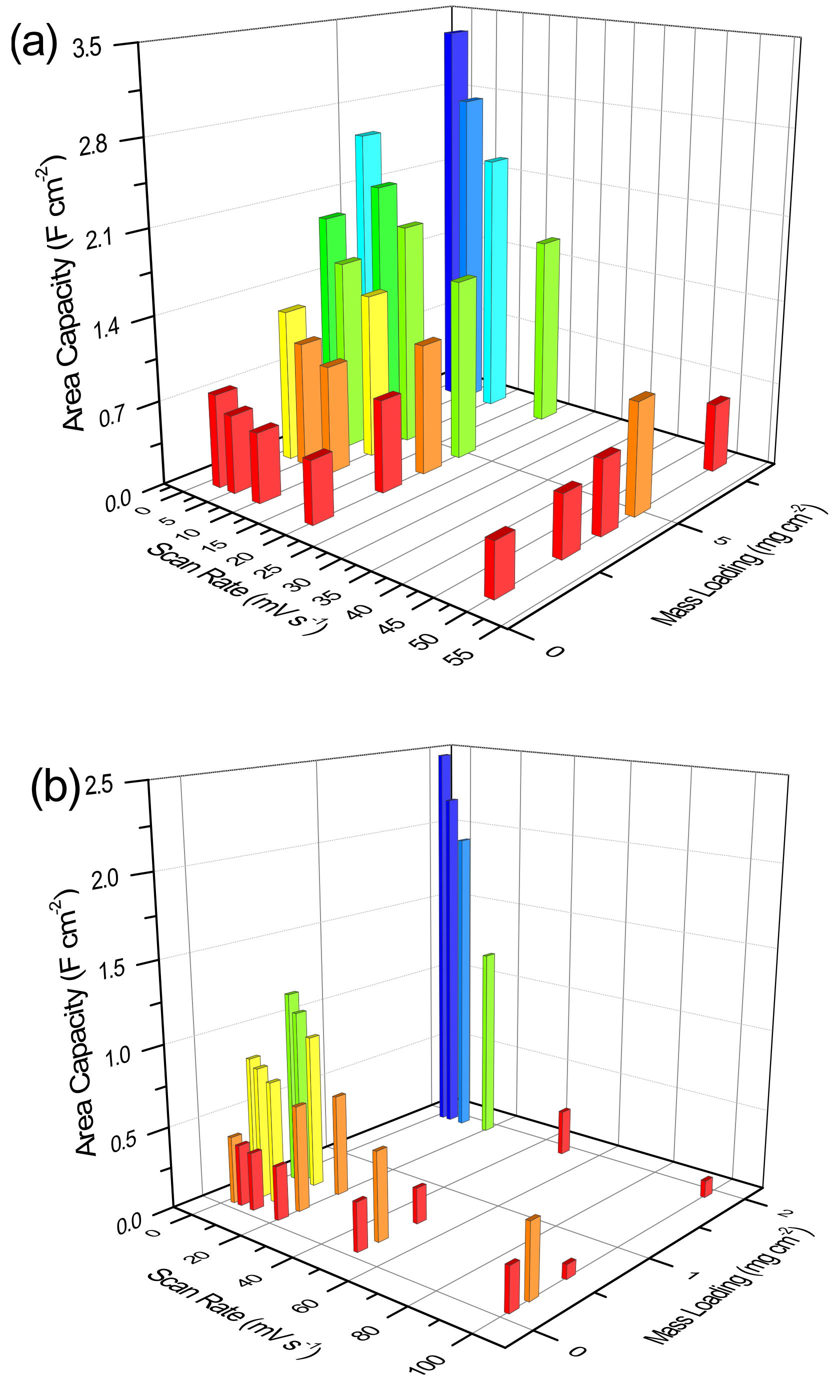

Figure S8. The specific areal capacitance values of PPy/VA-CNTs/CFP and Ni-Co-S/VA-CNTs/CFP with different mass loading of PPy (a) and Ni-Co-S (b) in mg per cm^2.

Table S1: Areal capacitance comparison of different electrode materials on various 3D substrates.

Electrode materials	Substrate	Areal capacitance
$CoxNi_{1-x}(OH)_2/NiCo_2S_4$[1]	Carbon fiber paper	2.86 F cm^2 at 4 mA/cm^2
NiO nanoflower[2]	Carbon fiber paper	0.93 F/cm^2 at 0.1 mA/cm^2
$NiCo_2O_4$ nanowires[3]	Carbon fiber paper	~1.64 F/cm^2 at 2mA/cm^2 for electrode
$(Co_xNi_{1-x})_9S_8$/carbon nanorod array[4]	Carbon fiber paper	1.32 F/cm^2 at 1mA/cm^2
PPy [5]	Carbon fiber paper	0.42 F/cm^2 at 1 mA/cm^2
PPy [6]	Carbon fiber paper	198.5 mF/cm^2 at 1 mA/cm^2
Ni-Co-S/VA-CNTs (this manuscript)	Carbon fiber paper	2.5 and 2.2 F/cm^2 at 2 and 5 mV/s
PPy/VA-CNTs (this manuscript)	Carbon fiber paper	3.3 and 2.7 F/cm2 at 2 and 5 mV/s
$NiCo_2O_4$ nanoneedles array[7]	Ni foam	3.12 and 1.44 F/cm^2 at 1.11 and 2.78 mA/cm^2
CoO nano array @PPy[8]	Ni foam	6.0 and 2.51 F/cm^2 at 1 and 5 mA/cm^2 for electrode
Ni-Co sulfide nanowire[9]	Ni foam	6.0 and 2.9 F/cm^2 at 2.5 and 30 mA/cm^2 for electrode
MnO_2-graphene[10]	Graphene/Ni foam	3.18 F/cm^2 (234.2 F/g)
Co_3O_4@MnO_2 core/shell nanowire array[11]	Stainless steel sheet (2D substrate)	0.56F/cm^2 at 11.25 mA/cm^2
TiO_2@PPy[12]	Carbon cloth	64.6 mF/cm^2 at 10 mV/s
RGO/PPy foam[13]	RGO foam	175 mF/cm^2 at 10 mV/s

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