

Supplementary Information

Highly dispersive Co_3O_4 nanoparticles incorporated in a cellulose nanofiber for a high performance flexible supercapacitor

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References

S1. Characterizations

The surface morphology of the as prepared (1D Co₃O₄@CNF hybrid, CNF and Co₃O₄) was analyzed using field emission scanning electron microscopy (FESEM; (JEOL JSM-6700F)) and transmission electron microscopy (TEM; (FEI Tecnai F30 S-TWIN TEM)). To TEM, sample were prepared by means of adding few drops of ethanol with samples and ultrasonically sonicated for about 1 min. Then, 0.3 μ/ml was dropped over carbon grid and arid into the vacuum oven for 1 hour. Compositional analysis was performed by using energy dispersive X-rays (EDX) technique operating at 5 kV and 10 keV. The sample was prepared for the EDX analysis by means of adding 0.3 μ/ml of diluted sample on the aluminum sheet and dried into the vacuum oven. X-ray diffraction (XRD; D/Max-2550 PC Rigaku Co., Korea, Cu Kα, λ = 1.5406 Å) and Raman spectroscopy (in Via-Reflex, Renishaw, Co., South Korea) analyses were employed to study the crystallinity structure and phase of all samples. X-ray photoelectron spectroscopy (XPS) were carried out using a Thermo Scientific ESCALAB 250Xi X-ray source to obtain the elemental composition, respectively. Moreover, the surface properties were measured using the Brunauer–Emmett–Teller (BET) N₂ adsorption-desorption isotherm by an automated adsorption system (ASAP-2020). The samples were first degassed followed by preconditioning the samples at 150 °C for 3 hours for the BET analysis. The powder sample was used for all techniques except of the EDX and TEM analyses.

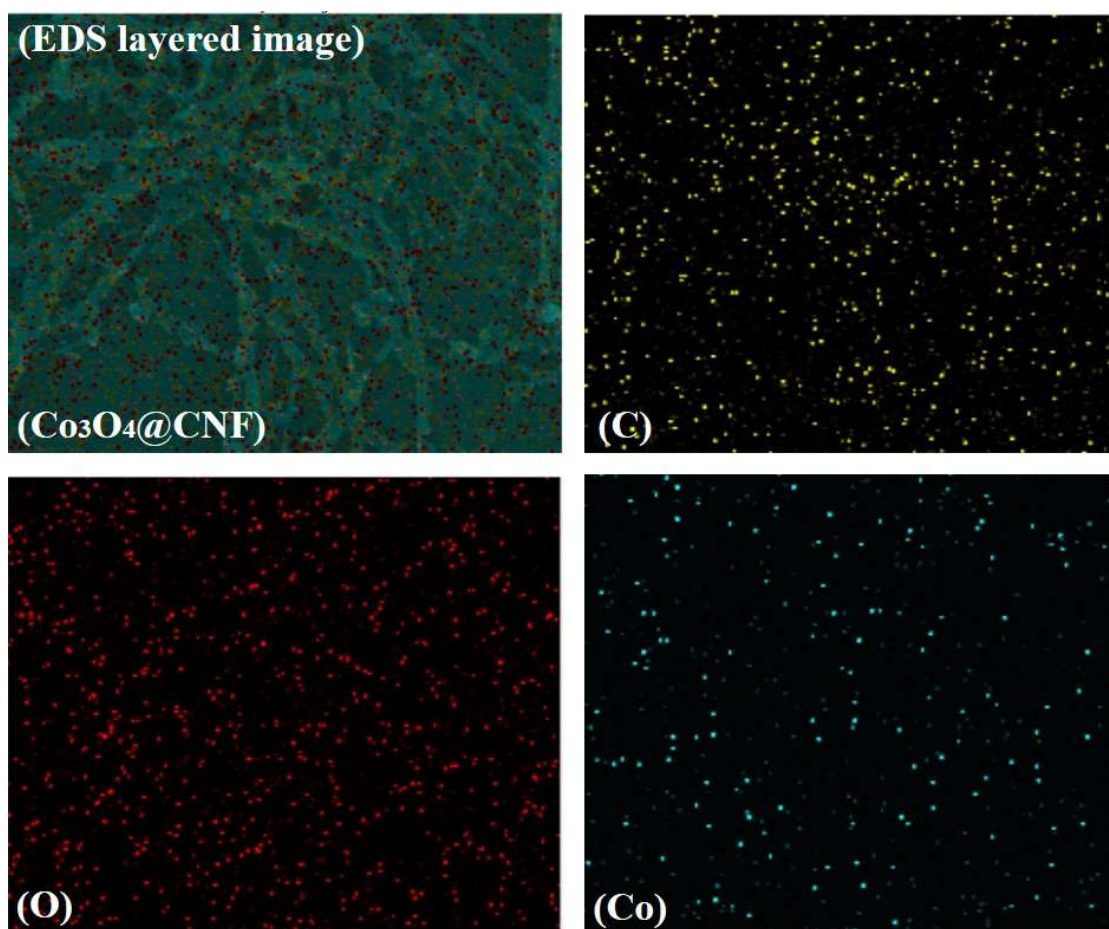


Figure S2. Elemental mapping for the 1D Co₃O₄@CNF hybrid.

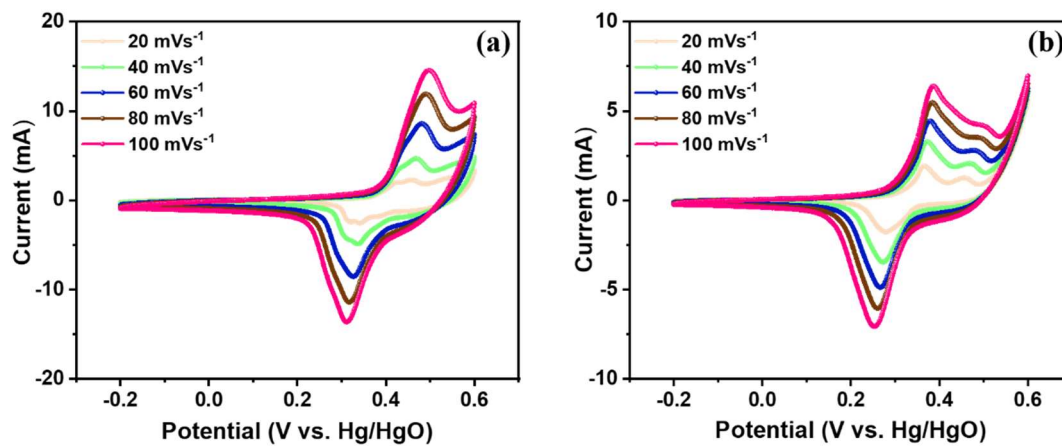


Figure S3. Cyclic voltammetry (CV) at different scan rates using 3 M KOH electrolyte within voltage window from -0.2 V to +0.6 V for **(a)** 1D Co₃O₄@CNF1 hybrid, **(b)** 1D Co₃O₄@CNF3 hybrid.

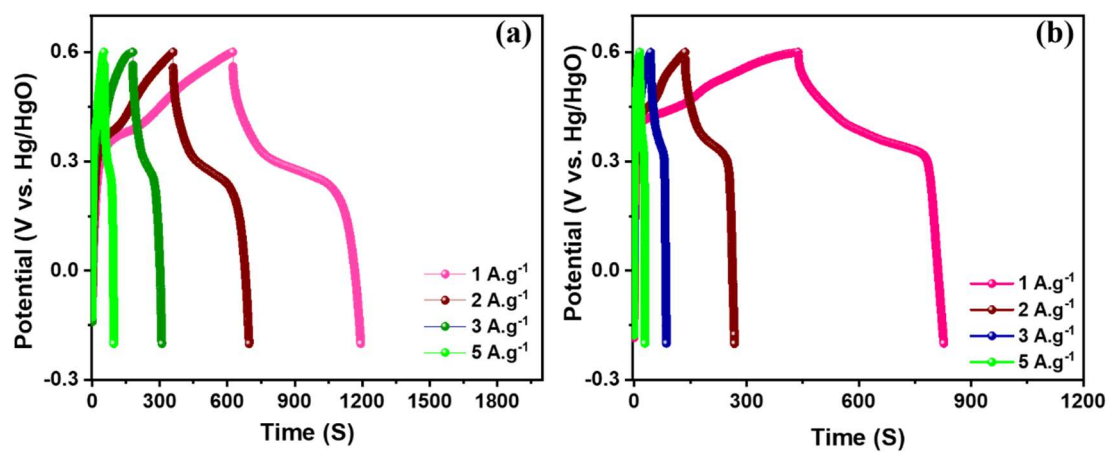


Figure S4. Charging/discharging (GCD) curves at current densities for (a) 1D Co₃O₄@CNF1 hybrid and (b) 1D Co₃O₄@CNF3 hybrid.

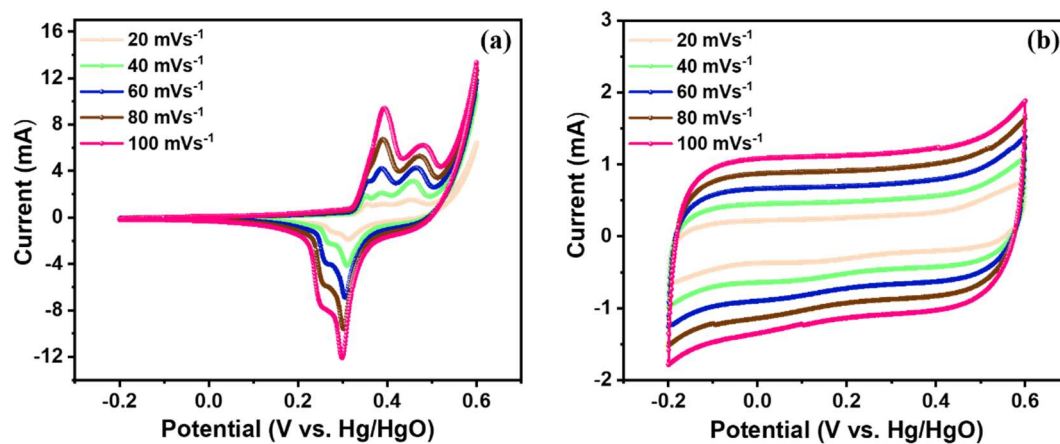


Figure S5. CV curves using 3 M KOH electrolyte at different scan rates within voltage window from -0.2 V to +0.6 V of **(a)** pristine Co_3O_4 nanoparticles **(b)** pristine CNF.

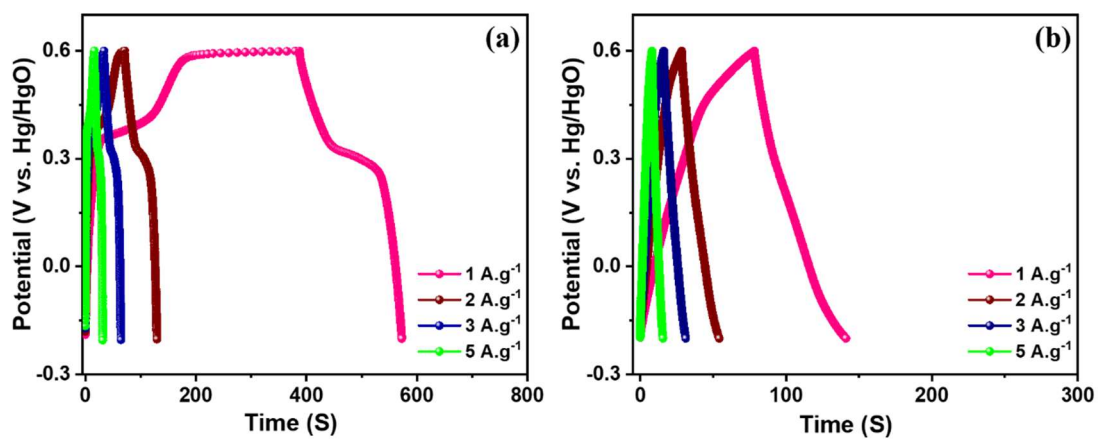


Figure S6. GCD curves at current densities with voltage window from -0.2 V to +0.6 V for **(a)** pristine Co_3O_4 nanoparticles **(b)** pristine CNF.

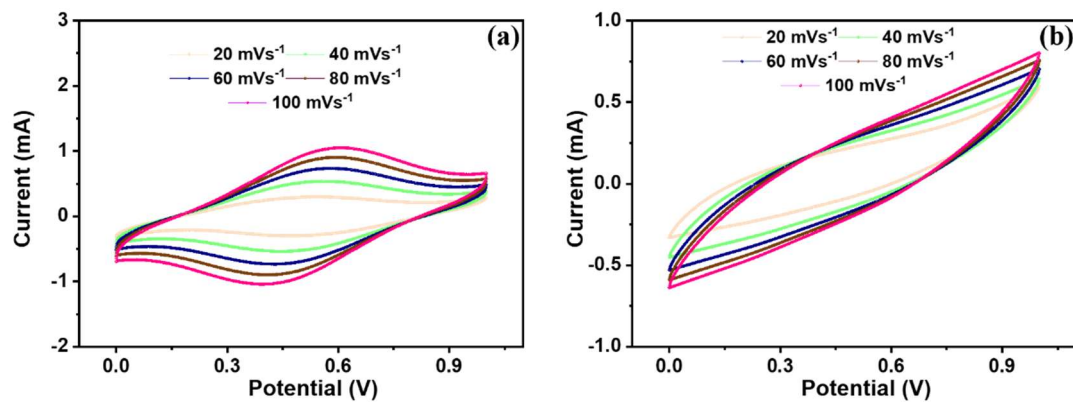


Figure S7. CV curves using 3 M KOH electrolyte at different scan rates within voltage window from 0 V to 1 V of **(a)** pristine Co_3O_4 nanoparticles and **(b)** pristine CNF.

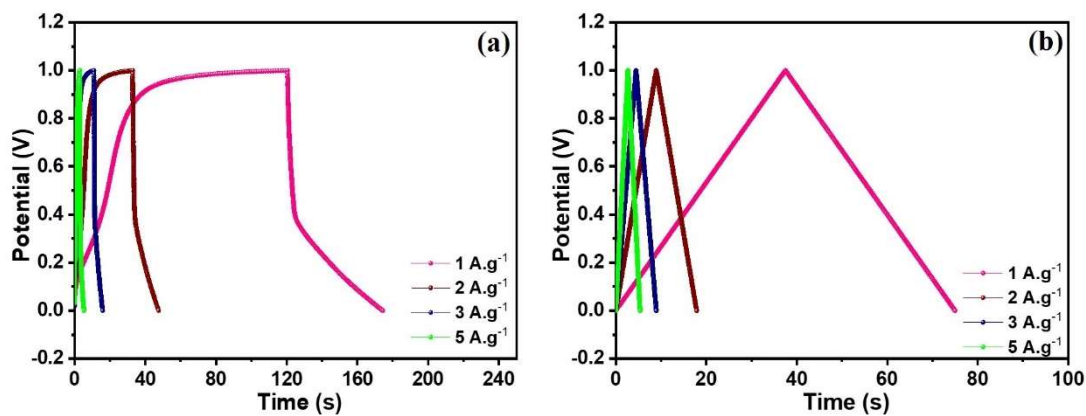


Figure S8. GCD curves at current densities with voltage window from 0 V to 1 V for **(a)** pristine Co₃O₄ nanoparticles and **(b)** pristine CNF.

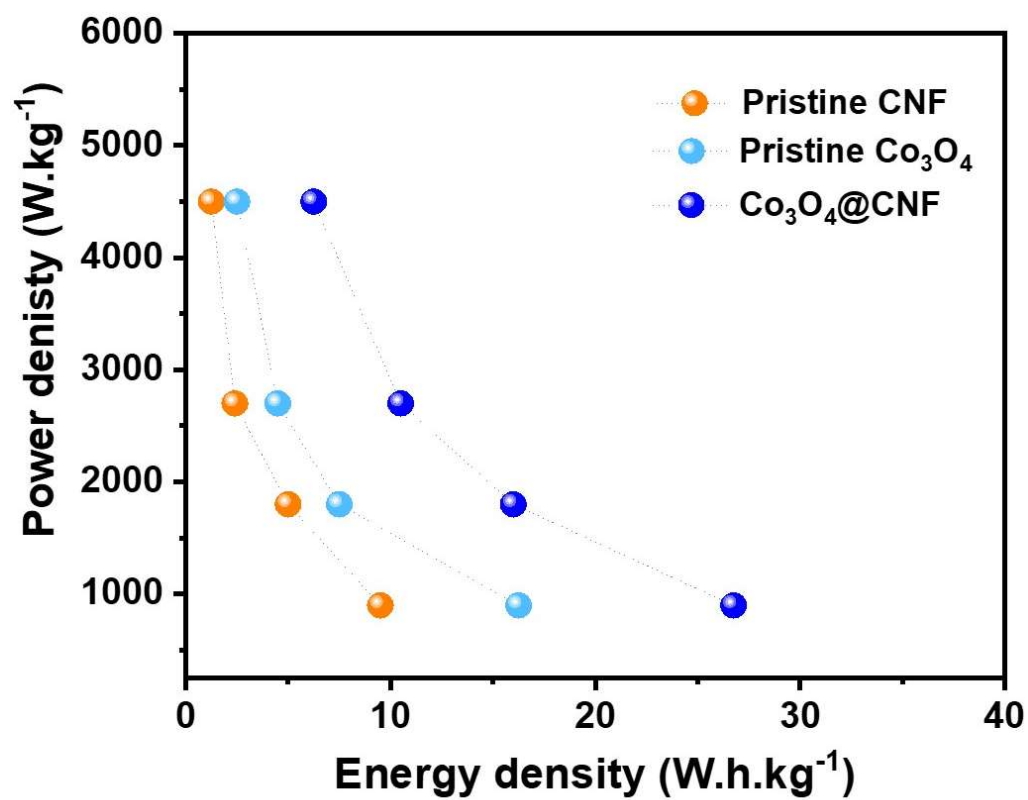


Figure S9. Energy density with respect to the power density plot at the different current sweep rates.

S10. Energy density and power density calculations by the non-linear discharging curve

The energy density (E), and power density (P) of SSCs cells were determined using the following equations:

$$E = \frac{1}{m} \int_0^{t_d} iV dt \quad \text{----- Equation 1}$$

$$P = \frac{E}{t_d} \quad \text{----- Equation 2}$$

Where ΔV is the voltage window, m is the mass of active electrode material, t_d is the discharging time, and i is the loading current density. We investigated the Ragone plot using the integrated discharging curve as can be seen below.

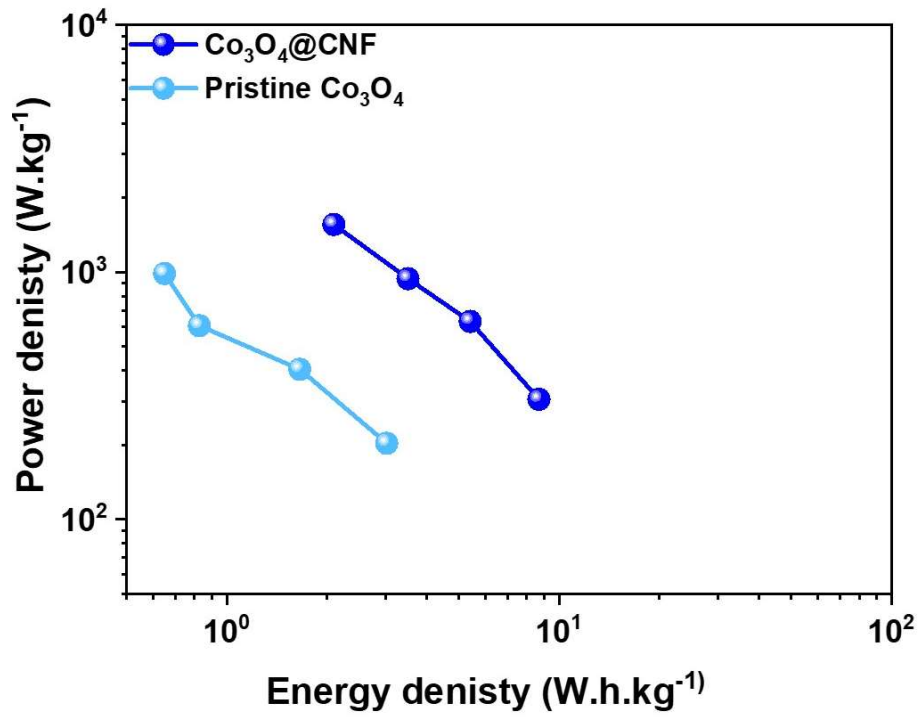


Figure S11. Ragone plot using integrated discharging formula.

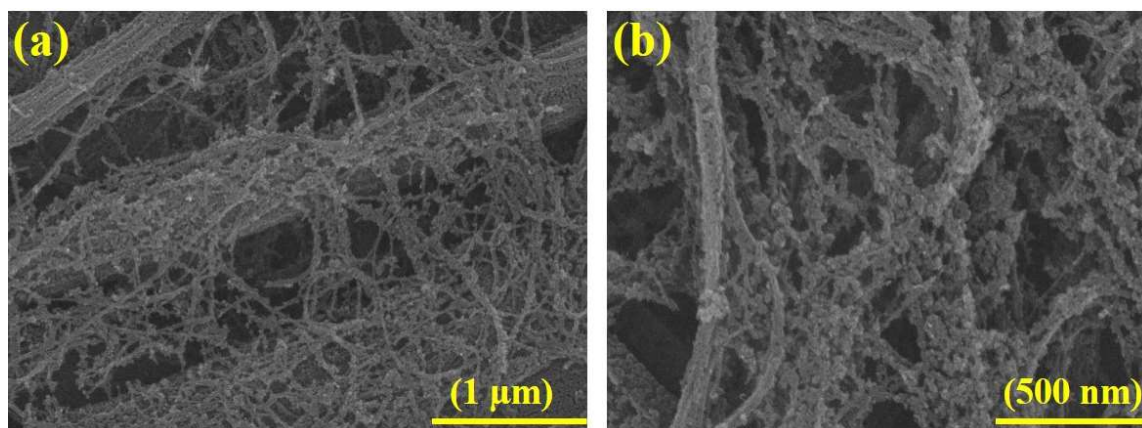


Figure S12(a-b). FESEM images of the 1D $\text{Co}_3\text{O}_4@\text{CNF}$ flexible paper like film.

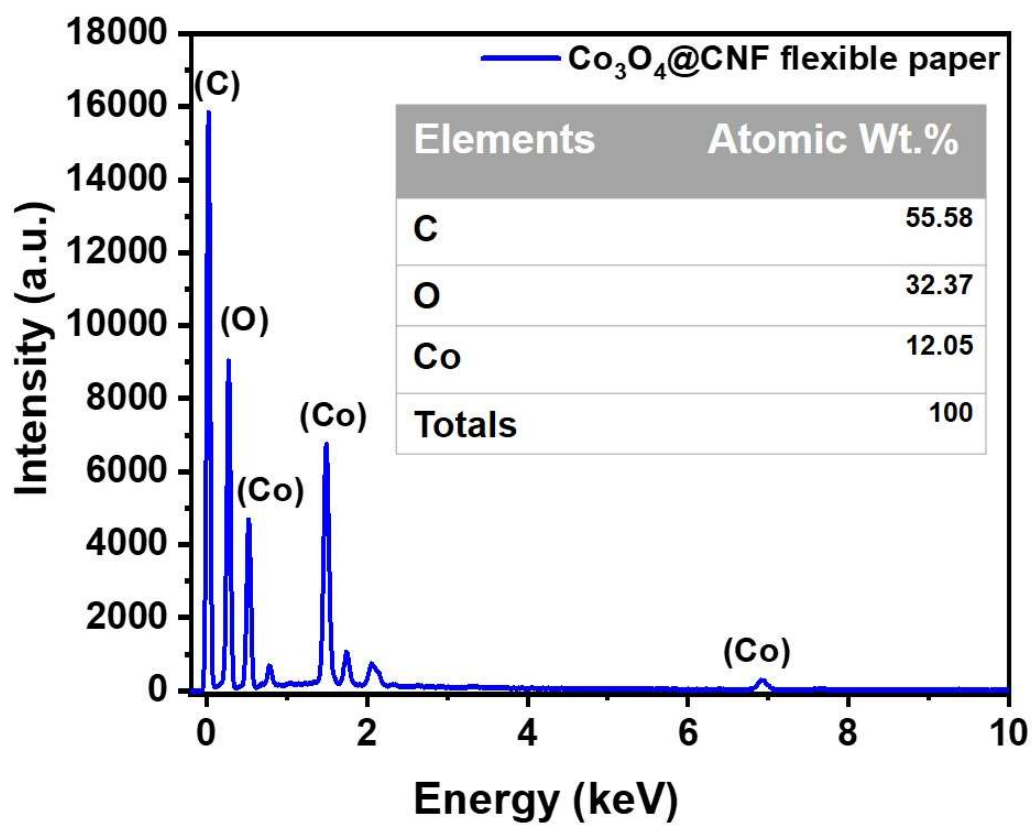


Figure S13. EDX spectrum of the 1D Co_3O_4 @CNF flexible paper like film.

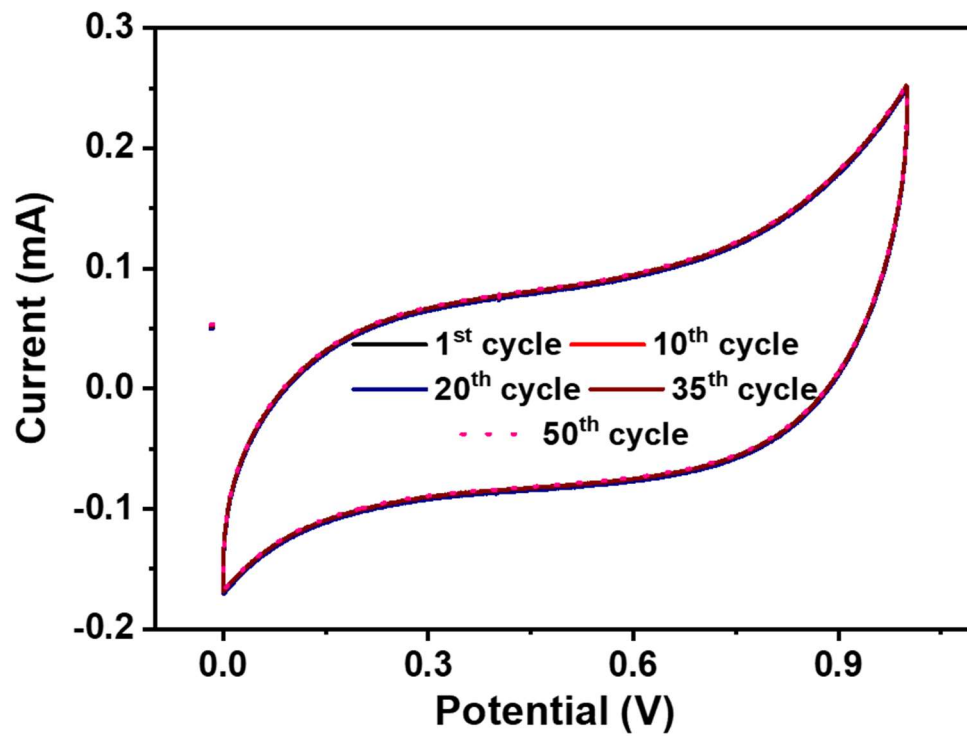


Figure S14. Stability curve of the 1D Co₃O₄@CNF flexible paper over the 50th cycle at the bending state.

Table S1. Comparative Electrochemical Performance of 1D Co₃O₄@CNF in the Three-Electrode System with Other Previously Reported Materials.

Electrode materials	Voltage window	Electrolyte	Specific capacitance	Ref.
1D Co ₃ O ₄ @CNF	-0.2 to 0.6 V	3 M KOH	789 F·g ⁻¹ at 1 A·g ⁻¹	present
yolk shell-CuCo ₂ Se ₄	-0.2 to 0.5 V	3 M KOH	512 F·g ⁻¹ at 1 A·g ⁻¹	[1]
Co ₃ O ₄ nanocrystals	-0.2 to 0.6 V	6 M KOH	284 F·g ⁻¹ at 1 A·g ⁻¹	[2]
Mn-Co sulfide nano sheets	0 to 0.65 V	2 M KOH	1.724 F cm ⁻² at 1 mA cm ⁻²	[3]
Porous MnCo ₂ O _{4.5}	-0.2 to 0.6 V	2 M KOH	342 F·g ⁻¹ at 0.5 A·g ⁻¹	[4]
porous Co ₃ O ₄	0 to 0.43 V	3 M KOH	342.1 F·g ⁻¹ at 1 A·g ⁻¹	[5]
Co ₉ S ₈ @N-C@MoS ₂	0 to 0.6 V	3 M KOH	410 F·g ⁻¹ at 1 A·g ⁻¹	[6]
Co ₃ O ₄ @N-rGO	-0.1 to 0.4 V	3 M KOH	450 F·g ⁻¹ at 1 A·g ⁻¹	[7]
NiCo ₂ S ₄	0 to 0.6 V	1 M KOH	464 F·g ⁻¹ at 1 A·g ⁻¹	[8]
NiCo ₂ O ₄	0 to 0.6 V	1 M KOH	173 F·g ⁻¹ at 1 A·g ⁻¹	[9]
MnO ₂ /MnCo ₂ O ₄	0 to 0.6 V	2 M KOH	497 F·g ⁻¹ at 1 A·g ⁻¹	[10]
NCA/Co ₃ O ₄	-0.05 to 0.45 V	6 M KOH	616 F·g ⁻¹ at 1.2 A	[10]
CuCo ₂ S ₄ /CNT/graphene	0 to 0.6 V	1 M Na ₂ SO ₄	504 F·g ⁻¹ at 10 A·g ⁻¹	[11]
CPSC-3rGO	-0.2 to 0.8 V	0.2 M Na ₂ SO ₄	446 F·g ⁻¹ at 1 A·g ⁻¹	[12]
Co ₉ S ₈ @Ni (OH) ₂	0 to 0.5 V	6 M KOH	149.44 mA h g ⁻¹ at 1Ag ⁻¹	[13]
CoS _x @carbon core-shell	0 to 0.4 V	6 M KOH	496 F·g ⁻¹ at 0.5 A·g ⁻¹	[14]
petal-like cobalt selenide	-0.1 to 0.65 V	2 M KOH	294 F·g ⁻¹ at 0.5 A·g ⁻¹	[15]
Co ₃ O ₄ nanoflakes@SrGO	-0.2 to 0.5 V	2 M KOH	406 F·g ⁻¹ at 1 A·g ⁻¹	[16]
CoMoO ₄ nanoclusters	-0.9 to 0.6 V	6.0 M KNO ₃	367 F·g ⁻¹ at 1.2 A·g ⁻¹	[17]
Ni-Co selenide	0 to 0.6 V	6 M KOH	742.4 F·g ⁻¹ at 1 mA cm ⁻²	[18]
NiCo ₂ O ₄	-0.2 to 0.6 V	6 M KOH	225 C. g ⁻¹ at 0.5 A g ⁻¹	[19]

Table S2. Comparative Electrochemical Performance of 1D Co₃O₄@CNF in the Two-Electrode System with Other Previously Reported Symmetric and Asymmetric Materials.

Electrode materials	Type	Electrolyte	Specific capacitance	Capacitance retention (%) / cycles	Ref.
Co ₃ O ₄ @CNF	Symmetric	3.0 M KOH	214 F·g ⁻¹ @ 1.0 A·g ⁻¹	94/5000	Present
CoNW/CF//CoNW/CF SSC	Symmetric	3.0 M KOH	517.33 mF/cm ³ @ 0.26 mA/cm ²	95/5000	[20]
NCOs	Symmetric	1.0 M KOH	89 F g ⁻¹ @ 0.23 A.g ⁻¹	-	[21]
CNF-RGO	Symmetric	H ₂ SO ₄ -PVA	203 F g ⁻¹ @ 0.7 mA/cm ²	99/5000	[22]
ZnO/Co ₃ O ₄ -450//AC	Asymmetric	1.0 M KOH	153 F g ⁻¹ @ 1 A.g ⁻¹	-	[23]
CC@NiC ₂ O ₄ //CC@NC	Asymmetric	6.0 M KOH	89.7 F g ⁻¹ @ 1 A g ⁻¹	86.7/20000	[24]
Co ₃ O ₄ @Ni ₃ S ₂	Asymmetric	3.0 M KOH	126 F g ⁻¹ @ 1 A.g ⁻¹	83.5/5000	[25]
Ag/NiO	Asymmetric	3.0 M KOH	204 C.g ⁻¹ @ 2.5 A.g ⁻¹	96/4000	[26]
Co ₃ O ₄ @Ni(OH) ₂ //AC	Asymmetric	6.0 M KOH	110 F.g ⁻¹ @ 2.5 A.g ⁻¹	86/1000	[27]
3D graphene-MoS ₂ hybrid	Symmetric	KOH/PVA	58.0F.g ⁻¹ @ 2 A.g ⁻¹	-	[28]
TaS ₂	Symmetric	PVA/LiCl	508 F/cm ³ @ 10 mV/s	92/4000	[29]
Cu ₂ WS ₄	Symmetric	PVA/LiCl	583.3 F cm ⁻³ @ 0.31 A cm ⁻³	95/3000	[30]
MoS ₂ -NH ₂ /PANI nanosheets	Symmetric	1 M H ₂ SO ₄	58.6 F g ⁻¹ @ 2 A.g ⁻¹	96.5/10000	[31]
MoS ₂ /CNS	Symmetric	1 M Na ₂ SO ₄	108 F g ⁻¹ @ 1 A.g ⁻¹	-	[32]
MoS ₂ /G nanobelts	Symmetric	1 M Na ₂ SO ₄	278.2 F.g ⁻¹ @ 0.8 A.g ⁻¹	-	[33]

MoS ₂ /rGO	Symmetric	1 M H ₂ SO ₄	306 F.g ⁻¹ @ 0.5 A.g ⁻¹	-	[34]
NiS/MoS ₂ @N-rGO	Symmetric	6 M KOH	1028 F.g ⁻¹ @1 A.g ⁻¹	94.5/50000	[35]
VSL-MoS ₂ @3D-Ni foam	Symmetric	Na ₂ SO ₄ / PVA	34.1 F.g ⁻¹ @ 1.3 A.g ⁻¹	82.5/10000	[36]
MoS ₂ /rGO	Symmetric	NaOH	323 F.g ⁻¹ @ 0.2 A.g ⁻¹	76.8/500	[37]
SS/MWCNTs/ MoTe ₂	Symmetric	PVA- LiClO ₄	68.01 F.g ⁻¹ @ 0.2 mA.cm ⁻²	94/2000	[38]
MWCNTs/MoSe ₂	Symmetric	PVA- KOH	27 F.g ⁻¹ @ 0.4 A/g	95/1000	[39]
MoS ₂ /carbon cloth	Symmetric	PVA- LiClO ₄	368 F.g ⁻¹ @ 5 mV/s	96.5/5000	[40]
MoS ₂ /NPG	Symmetric	1 M Na ₂ SO ₄	102.5 F g ⁻¹ @ 1 A g ⁻¹	91.67/5000	[41]
(Ni,Co) _{0.85} Se//porous graphene	Asymmetric	1.0 M KOH	529.3 mF c m ⁻² @ 1A g ⁻¹	85/10000	[42]
MoS ₂ /PEI-GO	Asymmetric	Na ₂ SO ₄	42.9 @ 0.5 A g ⁻¹	93.1/8000	[43]
MoS _{2-x} @CNTs/Ni	Asymmetric	1 M Na ₂ SO ₄	153.1F g ⁻¹ @ 1 A g ⁻¹	91/3000	[44]

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