

Supporting information for

MoS₂ Architectures Supported on Graphene Foam/Carbon Nanotube Hybrid Films: Highly Integrated Frameworks with Ideal Contact for Superior Lithium Storage

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Table S1 A survey of electrochemical properties of MoS₂ and its hybrid composites.

Electrode description	1st capacity (mAh g ⁻¹)	Specific	1st Coulombic efficiency	Cycling stability	Rate performance
Worm-like MoS ₂ nanoarchitectures on GF/CNTs film (this work)	1568 mAh g ⁻¹ at 100 mA g ⁻¹		79.8%	1112 mAh g ⁻¹ after 120 cycles at 200 mA g ⁻¹	1368, 1140, 1064, 1006 and 823 mAh g ⁻¹ at 200, 500, 1000, 2000 and 5000 mA g ⁻¹
Honeycomb-like MoS ₂ nanoarchitectures on 3DGF ¹	1397 mAh g ⁻¹ at 100 mA g ⁻¹		82.9%	1100 mAh g ⁻¹ after 60 cycles at 200 mA g ⁻¹	1172, 1095, 1007, 966 and 800 mAh g ⁻¹ at 200, 500, 1000, 2000 and 5000 mA g ⁻¹
MoS ₂ @graphene nanocables ²	1150 mAh g ⁻¹ at 500 mA g ⁻¹			900 mAh g ⁻¹ after 700 cycles at 5 A g ⁻¹	1150 and 700 mAh g ⁻¹ at 500 mA g ⁻¹ and 10 A g ⁻¹
MoS ₂ -carbon nanofiber composite ³	1712 mAh g ⁻¹ at 100 mA g ⁻¹		74%	1007 mAh g ⁻¹ after 100 cycles at 1 A g ⁻¹	1095, 986, 768, 637, 620, 548 and 347 mAh g ⁻¹ at 0.5, 1, 5, 10, 20, 30 and 50 A g ⁻¹
MoS ₂ -graphene-carbon nanotube nanocomposites ⁴	949 mAh g ⁻¹ at 100 mA g ⁻¹			886 mAh g ⁻¹ after 100 cycles at 1 A g ⁻¹	949, 883, 858, 737 and 652 mAh g ⁻¹ at 100, 500, 1000, 5000 and 10000 mA g ⁻¹
Hierarchical C@MoS ₂ microspheres ⁵				750 mAh g ⁻¹ after 50 cycles at 100 mA g ⁻¹	500 mAh g ⁻¹ at 1000 mA g ⁻¹
MoS ₂ nanoflake array/carbon cloth ⁶	3.5 mAh cm ⁻² at a current density of 0.15 mA cm ⁻²		97.6%		3.26, 2.73, 2.39, 1.72, 1.24, and 0.85 mAh cm ⁻² at current densities of 0.15, 0.3, 0.75, 1.5, 2.25, and 3.0 mA cm ⁻²
MoS ₂ /3DGN ⁷	1222 mAh g ⁻¹ at 100 mA g ⁻¹		83.50%	877 mAh g ⁻¹ after 50 cycles at 100 mA g ⁻¹	849, 782, 692, 597 and 466 mAh g ⁻¹ at 100, 200, 500, 1000 and 4000 mA g ⁻¹
MoS ₂ /graphene nanosheet ⁸	2200 mAh g ⁻¹ at 100 mA g ⁻¹		59.10%	1290 mAh g ⁻¹ after 50 cycles at 100 mA g ⁻¹	1040 mAh g ⁻¹ at 1000 mA g ⁻¹

MoS ₂ /graphene composites ⁹	1462 mAh g ⁻¹ at 100 mA g ⁻¹	58.5%	1187 mAh g ⁻¹ after 100 cycles at 100 mA g ⁻¹	900 mAh g ⁻¹ at 1000 mA g ⁻¹
MoS ₂ /amorphous carbon ¹⁰	2100 mAh g ⁻¹ at 100 mA g ⁻¹	44.10%	912 mAh g ⁻¹ after 100 cycles at 100 mA g ⁻¹	
CNT@MoS ₂ ¹¹	1434 mAh g ⁻¹ at 100 mA g ⁻¹	60.01%	698 mAh g ⁻¹ after 60 cycles at 100 mA g ⁻¹	653, 459 and 369 mAh g ⁻¹ at 200, 500 and 1000 mA g ⁻¹
MoS ₂ /amorphous carbon ¹²	2108 mAh g ⁻¹ at 100 mA g ⁻¹	79%	755 mAh g ⁻¹ after 100 cycles at 100 mA g ⁻¹	850 mAh g ⁻¹ at 400 mA g ⁻¹
MoS ₂ /PS microspheres ¹³	1160 mAh g ⁻¹ at 100 mA g ⁻¹	68.20%	672 mAh g ⁻¹ after 50 cycles at 100 mA g ⁻¹	726, 581 and 353 mAh g ⁻¹ at 200, 500 and 1000 mA g ⁻¹
Graphene-network-ba cknosed MoS ₂ ¹⁴	1200 mAh g ⁻¹ at 600 mA g ⁻¹	68%	1200 mAh g ⁻¹ after 30 cycles at 600 mA g ⁻¹	620 and 270 mAh g ⁻¹ at 7200 and 84000 mA g ⁻¹
MoS ₂ nanoplates ¹⁵	1062 mAh g ⁻¹ at 1062 mA g ⁻¹	87%	907 mAh g ⁻¹ after 50 cycles at 100 mA g ⁻¹	790 and 700 mAh g ⁻¹ at 31.8 and 53.1A g ⁻¹
3D MoS ₂ flowers ¹⁶	869 mAh g ⁻¹ at 100 mA g ⁻¹	65.90%	633 mAh g ⁻¹ after 50 cycles at 100 mA g ⁻¹	848 and 740 mAh g ⁻¹ at 100 and 400 mA g ⁻¹
Mesoporous MoS ₂ ¹⁷	1052 mAh g ⁻¹ at 100 mA g ⁻¹	83.90%	876 mAh g ⁻¹ after 100 cycles at 100 mA g ⁻¹	903, 880, 845, 795, 748, 670 and 608 mAh g ⁻¹ at 100, 200, 500, 1000, 2000 and 5000 mA g ⁻¹
MoS ₂ /CNT network ¹⁸	1715 mAh g ⁻¹ at 200 mA g ⁻¹	76.10%	1456 mAh g ⁻¹ after 50 cycles at 200 mA g ⁻¹	1431, 1367, 1302 and 1224 mAh g ⁻¹ at 400, 600, 800 and 1000 mA g ⁻¹
MoS _x /CNT ¹⁹	1549 mAh g ⁻¹ at 50 mA g ⁻¹	74.80%	≥1000 mAh g ⁻¹ after 40 cycles at 50 mA g ⁻¹	1119, 904, 659, 358 and 197 mAh g ⁻¹ at 50, 200, 500 and 1000 mA g ⁻¹
MoS ₂ -CNT film ²⁰	1117 mAh g ⁻¹ at 100 mA g ⁻¹	73.40%	960 mAh g ⁻¹ after 100 cycles at 100 mA g ⁻¹	670 (3200) 670 mAh g ⁻¹ at 3200 mA g ⁻¹
Hollow MoS ₂ nanoparticles ²¹	1236 mAh g ⁻¹ at 100 mA g ⁻¹	74%	902 mAh g ⁻¹ after 80 cycles at 100 mA g ⁻¹	1030, 950, 910, 850 and 780 mAh g ⁻¹ at 100, 200, 300, 500

				and 1000 mA g ⁻¹
3D MoS ₂ assembly tubes ²²	1172 mAh g ⁻¹ at 100 mA g ⁻¹	68.30%	839 mAh g ⁻¹ after 50 cycles at 100 mA g ⁻¹	600 and 500 mAh g ⁻¹ at 1000 and 5000 mA g ⁻¹
MoS ₂ -graphene composites ²³	1367 mAh g ⁻¹ at 100 mA g ⁻¹	66.70%	808 mAh g ⁻¹ after 100 cycles at 100 mA g ⁻¹	571 mAh g ⁻¹ at 1000 mA g ⁻¹
PEO/MoS ₂ /graphene ²⁴	1150 mAh g ⁻¹ at 50 mA g ⁻¹	74%	≥1000 mAh g ⁻¹ after 180 cycles at 50 mA g ⁻¹	650 mAh g ⁻¹ at 200 mA g ⁻¹
MoS ₂ /polyaniline ²⁵	1460 mAh g ⁻¹ at 100 mA g ⁻¹	72.80%	953 mAh g ⁻¹ after 50 cycles at 100 mA g ⁻¹	1006 mAh g ⁻¹ at 200 mA g ⁻¹
MoS ₂ /C nanotube ²⁶	1320 mAh g ⁻¹ at 200 mA g ⁻¹	70.50%	776 mAh g ⁻¹ after 100 cycles at 200 mA g ⁻¹	450-600 mAh g ⁻¹ at 1000 mA g ⁻¹
MoS ₂ @carbon spheres ⁵	1020 mAh g ⁻¹ at 100 mA g ⁻¹	73.50%	750 mAh g ⁻¹ after 50 cycles at 100 mA g ⁻¹	500 mAh g ⁻¹ at 1000 mA g ⁻¹
MoS ₂ @carbon layer ²⁷	1251 mAh g ⁻¹ at 1000 mA g ⁻¹	90.70%	814 mAh g ⁻¹ after 100 cycles at 1000 mA g ⁻¹	600 mAh g ⁻¹ at 4000 mA g ⁻¹
MoS ₂ @CMK-3 ²⁸	1056 mAh g ⁻¹ at 250 mA g ⁻¹	78.03%	602 mAh g ⁻¹ after 100 cycles at 250 mA g ⁻¹	832, 774, 666 and 564 mAh g ⁻¹ at 250, 500, 1000 and 2000 mA g ⁻¹
Fe ₃ O ₄ /MoS ₂ ²⁹	1320 mAh g ⁻¹ at 100 mA g ⁻¹	81.74%	1200 mAh g ⁻¹ after 560 cycles at 500 mA g ⁻¹	1189, 943, 569, 362 and 270, 224 mAh g ⁻¹ at 1000, 2000, 4000, 6000, 8000 and 10000 mA g ⁻¹
MoS ₂ /TiO ₂ ³⁰	931 mAh g ⁻¹ at 100 mA g ⁻¹	74%	472 mAh g ⁻¹ after 100 cycles at 100 mA g ⁻¹	713, 636, 533 and 461 mAh g ⁻¹ at 100, 200, 500 and 1000 mA g ⁻¹

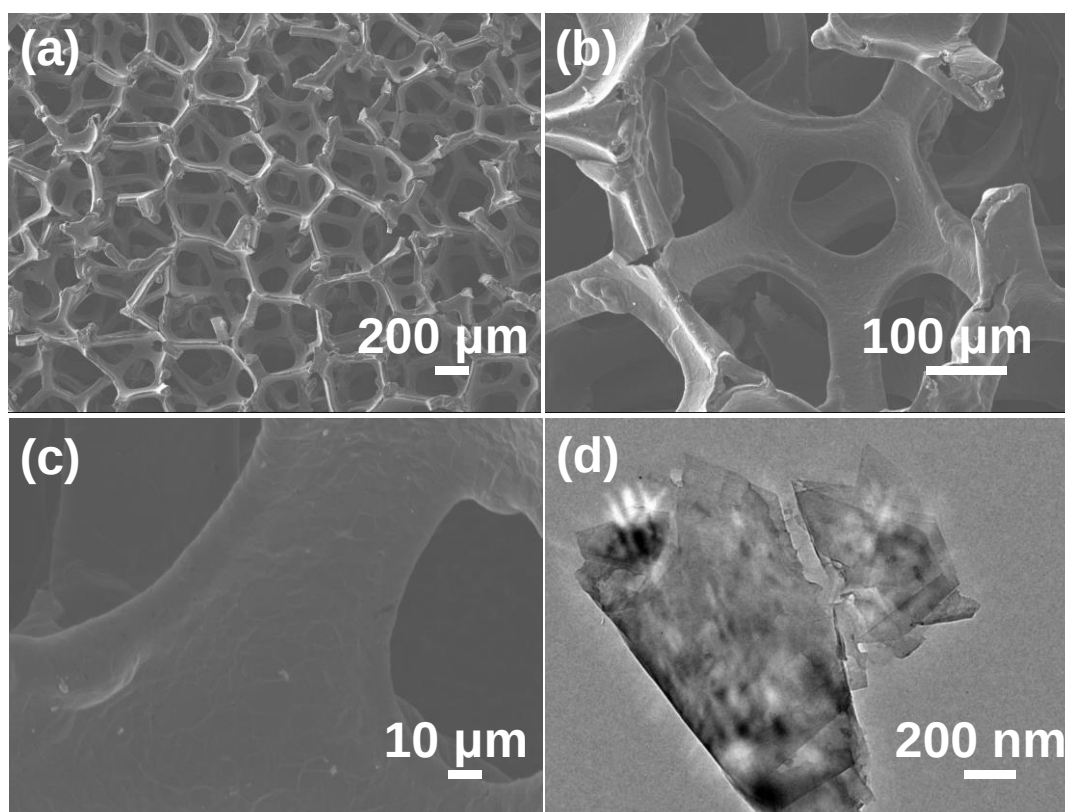


Figure S1 SEM images of GF (a), (b) and (c), and TEM image of GF.

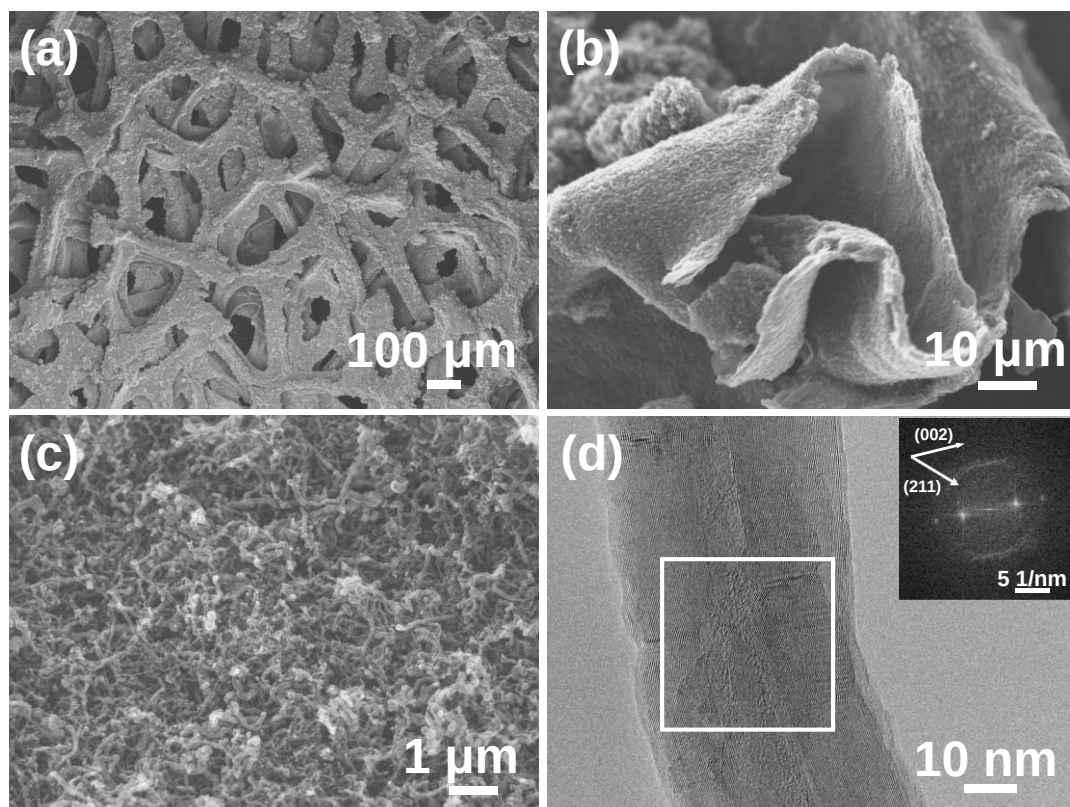


Figure S2 (a) Low-magnification SEM of the CNT-GF, (b), (c) and (d) TEM of the CNTs. Inset in (d) shows the FFT pattern taken from the marked area.

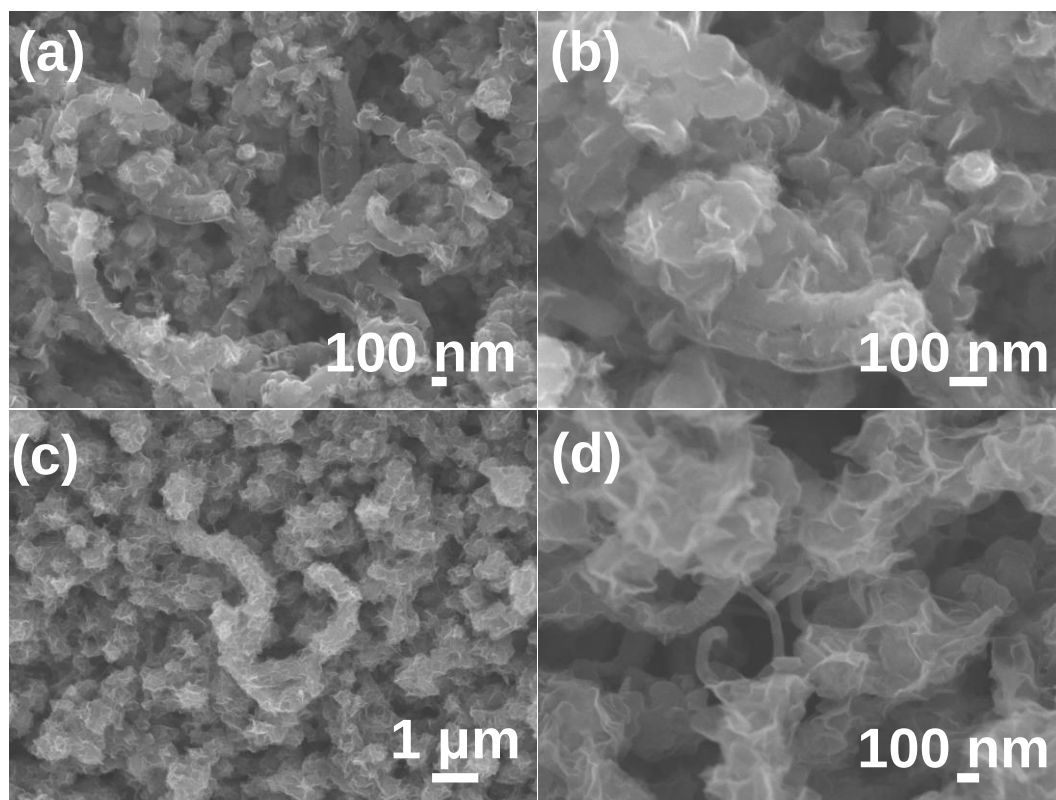


Figure S3 SEM images of samples prepared with different concentration of reactants: (a) and (b) the MoS₂@GF/CNTs sample prepared with 1.33 mg ml⁻¹ of TAA and 0.67 mg ml⁻¹ of sodium molybdate. (c) and (d) the MoS₂@GF/CNTs sample prepared with 2 mg ml⁻¹ of TAA and 1 mg ml⁻¹ of sodium molybdate.

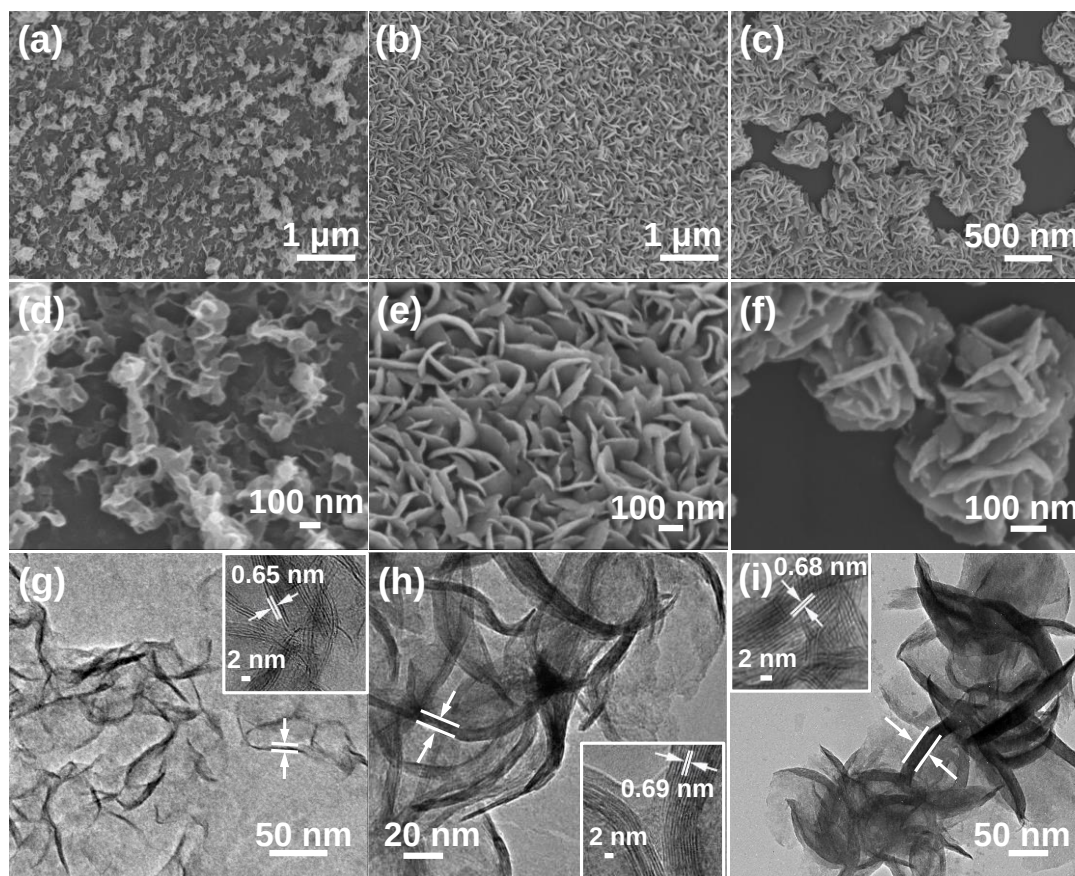


Figure S4 SEM and TEM images of samples prepared with different concentration of reactants: (a), (d) and (g) the MoS₂@GF sample prepared with 2 mg ml⁻¹ of TAA and 1 mg ml⁻¹ of sodium molybdate. (b), (e) and (h) the MoS₂@GF sample prepared with 3.33 mg ml⁻¹ of TAA and 1.67 mg ml⁻¹ of sodium molybdate. (c), (f) and (i) the MoS₂@GF sample prepared with 4 mg ml⁻¹ of TAA and 2 mg ml⁻¹ of sodium molybdate.

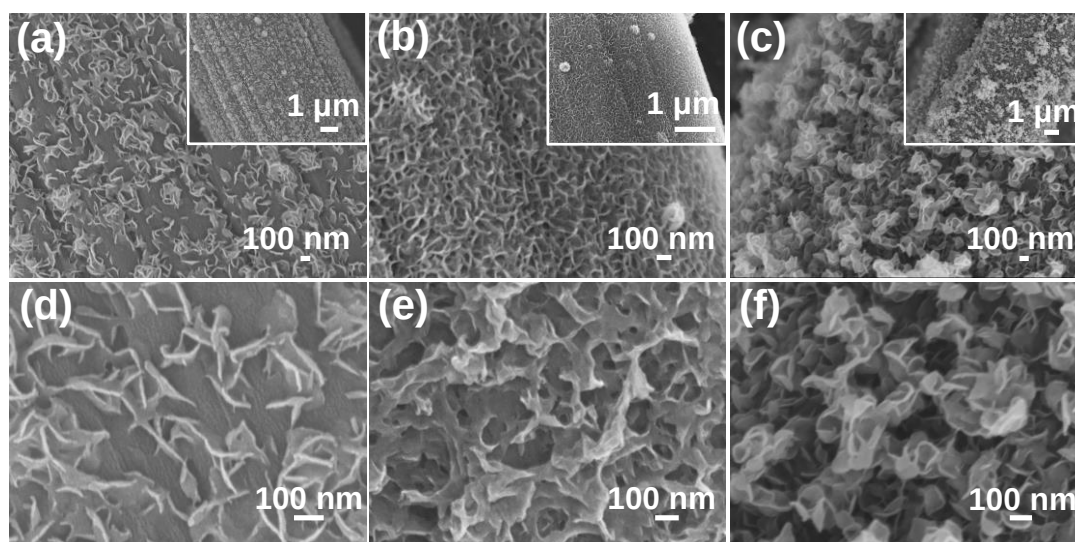


Figure S5 SEM images of samples prepared with different concentration of reactants: (a) and (d) the MoS₂@carbon cloth sample prepared with 1.67 mg ml⁻¹ of TAA and 0.83 mg ml⁻¹ of sodium molybdate. (b) and (e) the MoS₂@carbon cloth sample prepared with 2 mg ml⁻¹ of TAA and 1 mg ml⁻¹ of sodium molybdate. (c) and (f) the MoS₂@carbon cloth sample prepared with 3 mg ml⁻¹ of TAA and 1.5 mg ml⁻¹ of sodium molybdate.

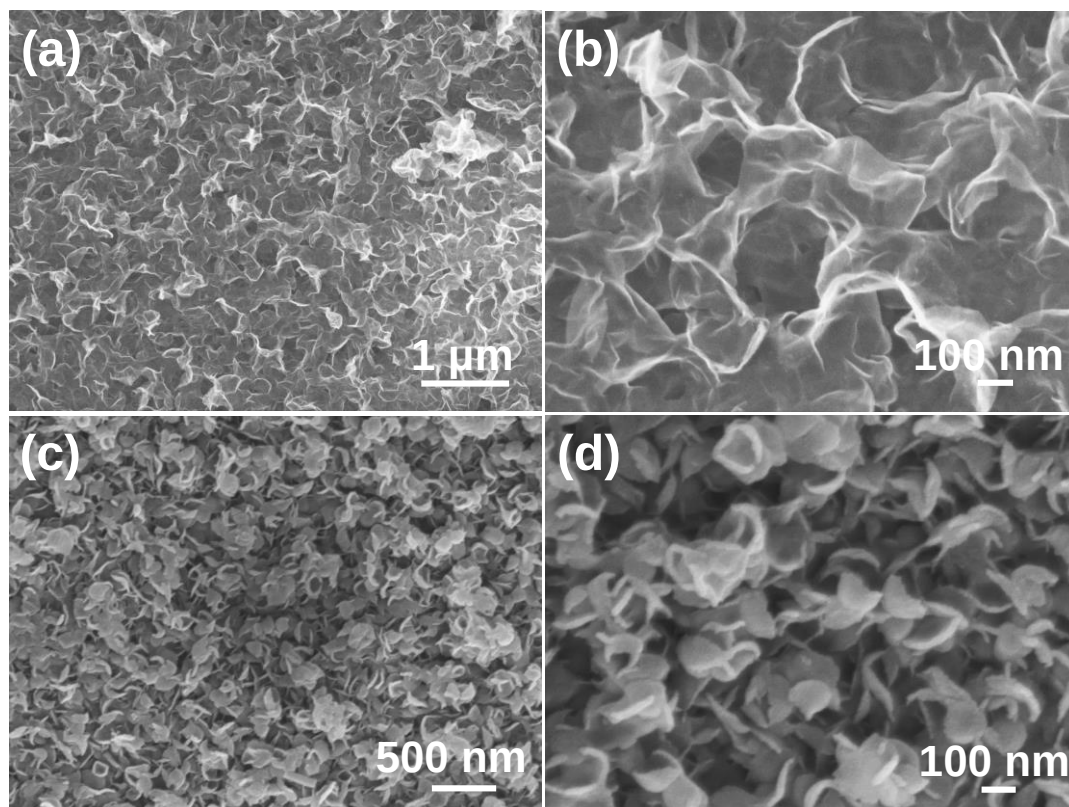


Figure S6 SEM images of samples grown on different substrates prepared with 3 mg ml⁻¹ of TAA and 1.5 mg ml⁻¹ of sodium molybdate: (a) and (b) on the Ti foil. (c) and (d) on the stainless steel.

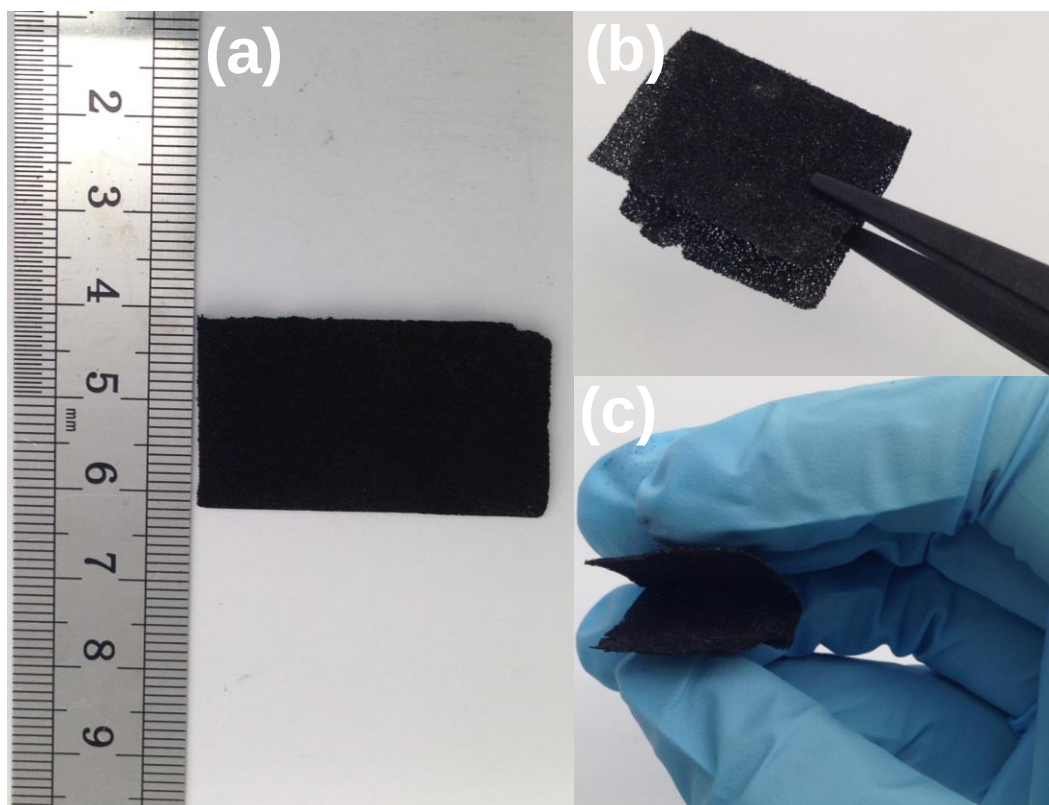


Figure S7 (a, b and c) Representative photograph showing the dimension and flexibility of the $\text{MoS}_2@\text{GF}/\text{CNT}$ electrode.

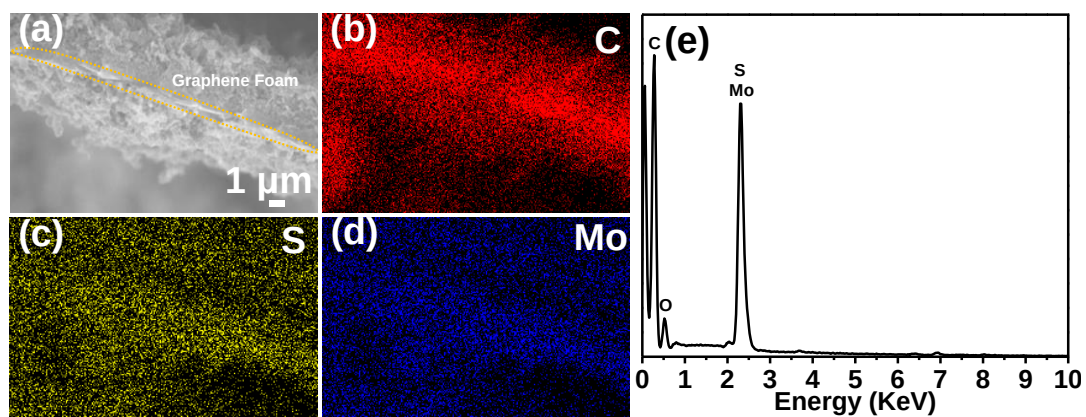


Figure S8 (a) SEM images of cross-sectional images of MoS₂@GF/CNT branch. Corresponding elemental mapping images of C (b), S (c) and Mo (d), indicating the uniform covering of MoS₂@CNTs on GF. And (e) EDS spectrum.

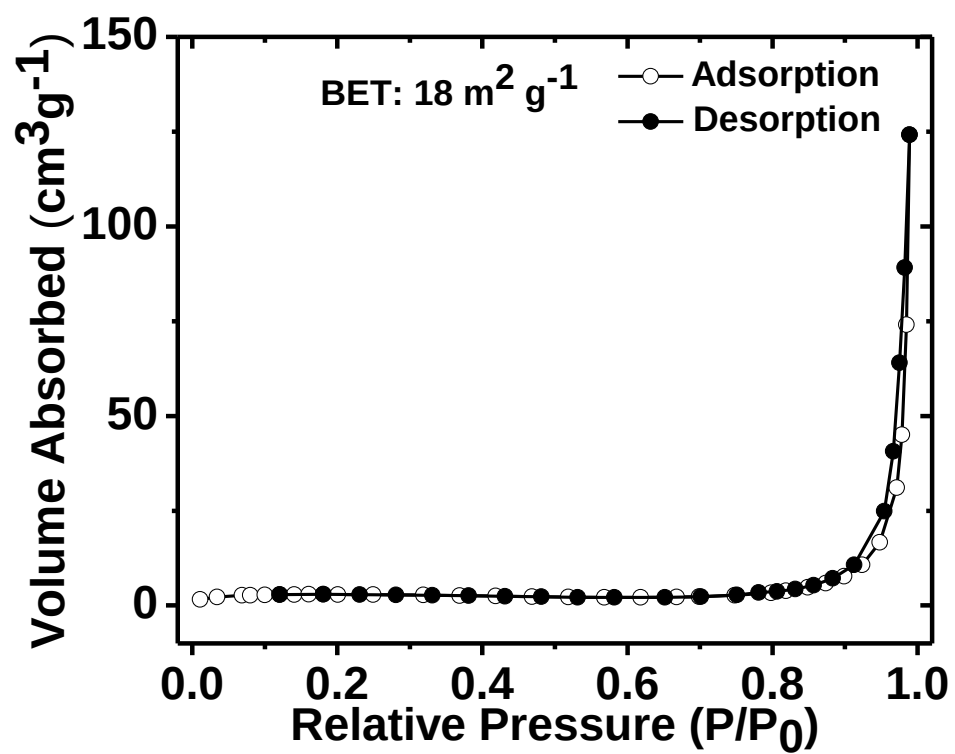


Figure S9 N_2 adsorption/desorption isotherms, and the insert corresponding pore size distribution of $\text{MoS}_2@\text{GF}$ composites prepared with 2.67 mg ml^{-1} of sodium molybdate and 1.33 mg ml^{-1} of TAA.

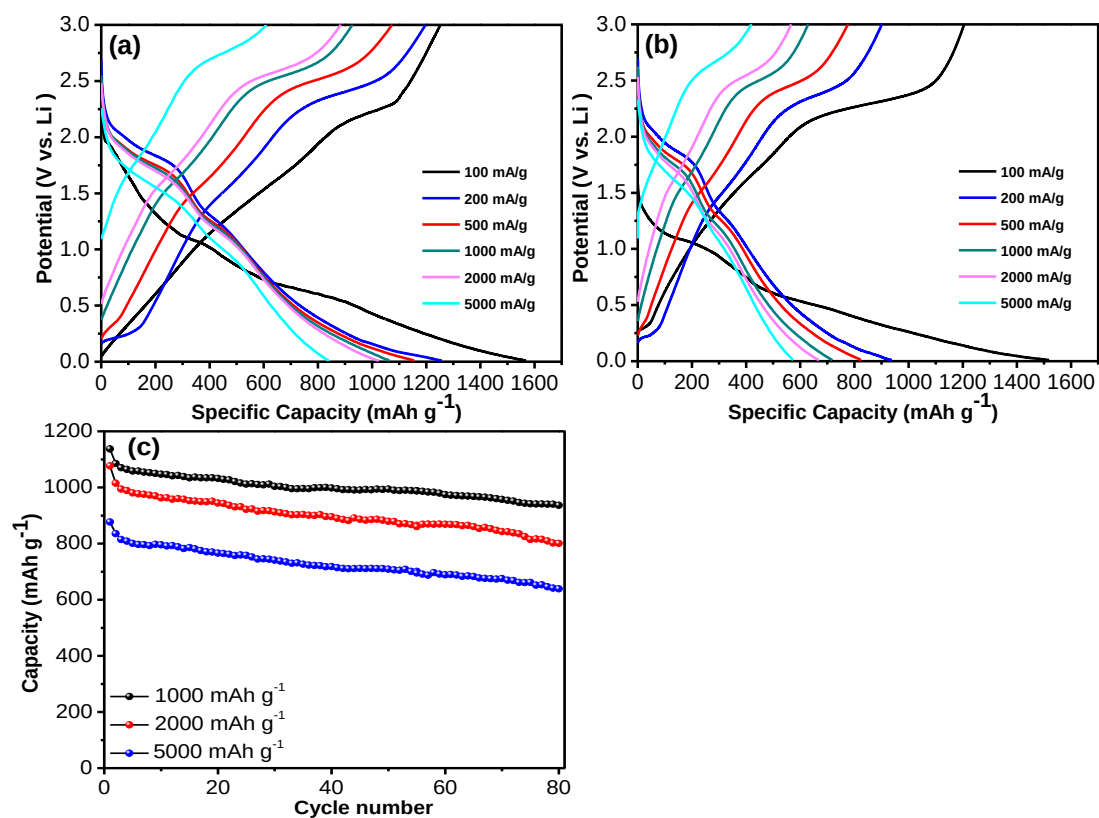


Figure S10 Charge and discharge curves of (a) MoS₂@GF/CNT and (b) MoS₂@GF at different C-rates. (c) Cycling behaviors of MoS₂@GF at current densities of 1000, 2000 and 5000 mAh g⁻¹.

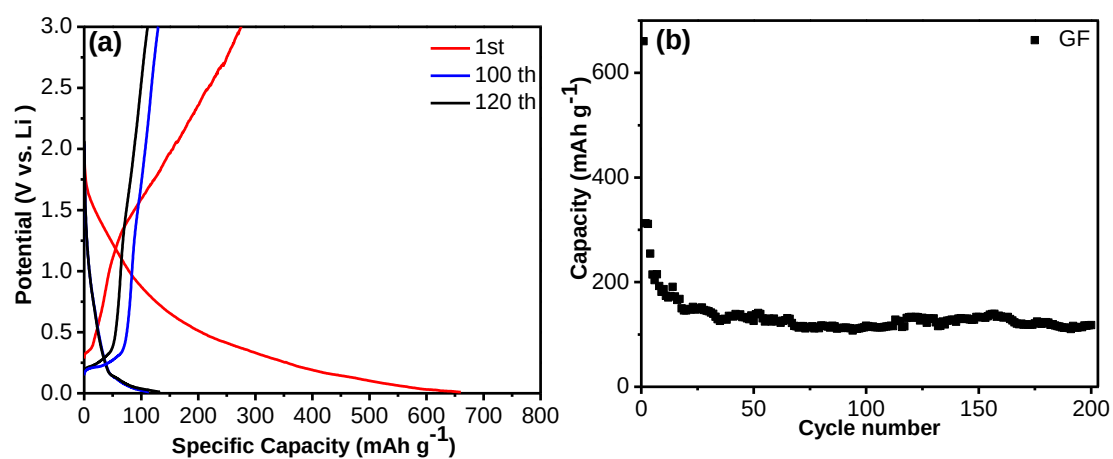


Figure S11 (a) Charge and discharge curves and (b) cycling behavior of pure GF at a current density of 200 mAh g⁻¹.

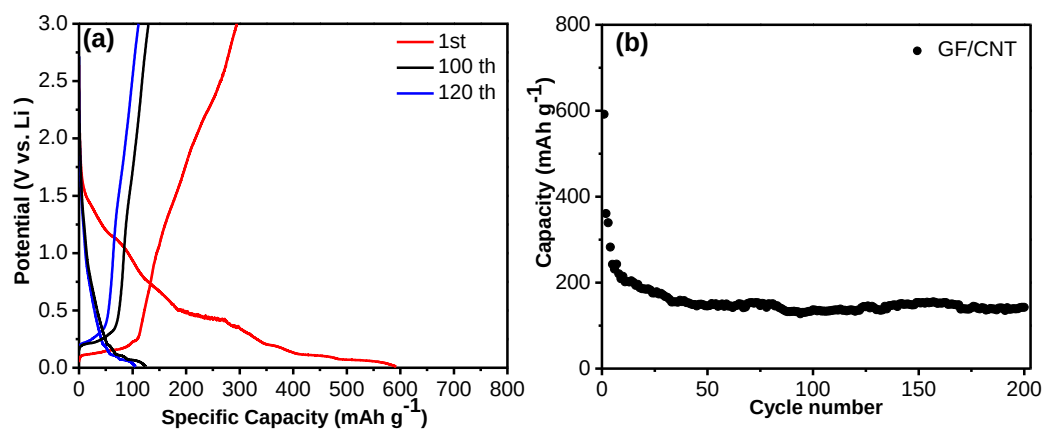


Figure S12 (a) Charge and discharge curves and (b) cycling behavior of pure GF/CNT at a current density of 200 mAh g⁻¹.

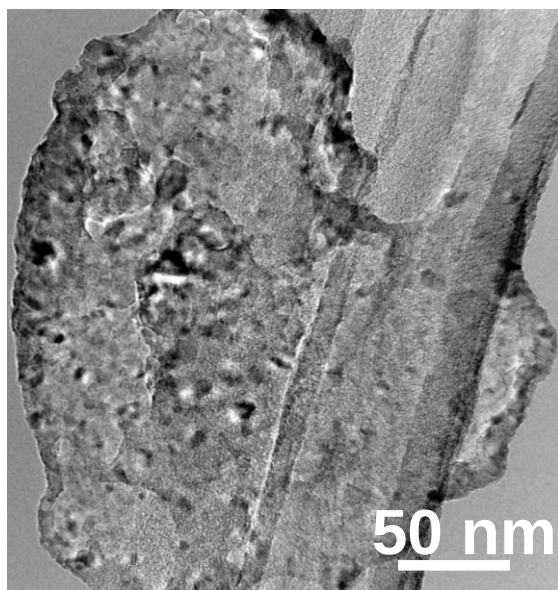


Figure S13 TEM images of MoS₂@GF/CNT electrode after cycling for 120 cycles.

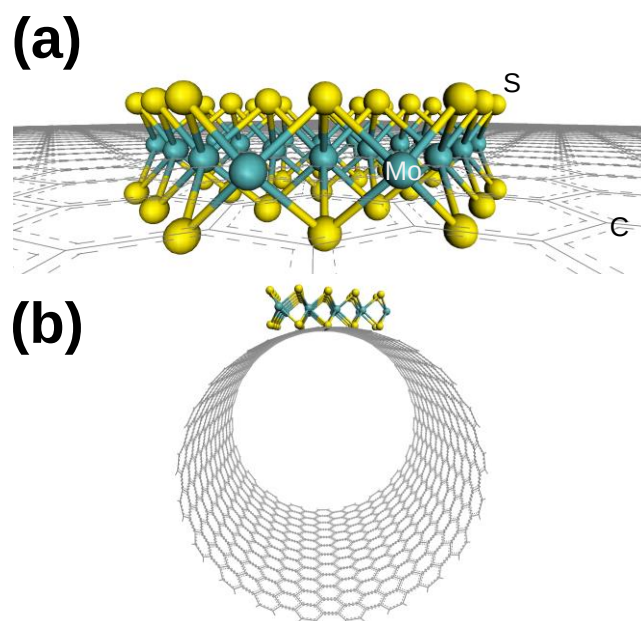


Figure S14 Cross-section illustration of MoS₂ cluster bonding with (a) graphene and (b) CNT.

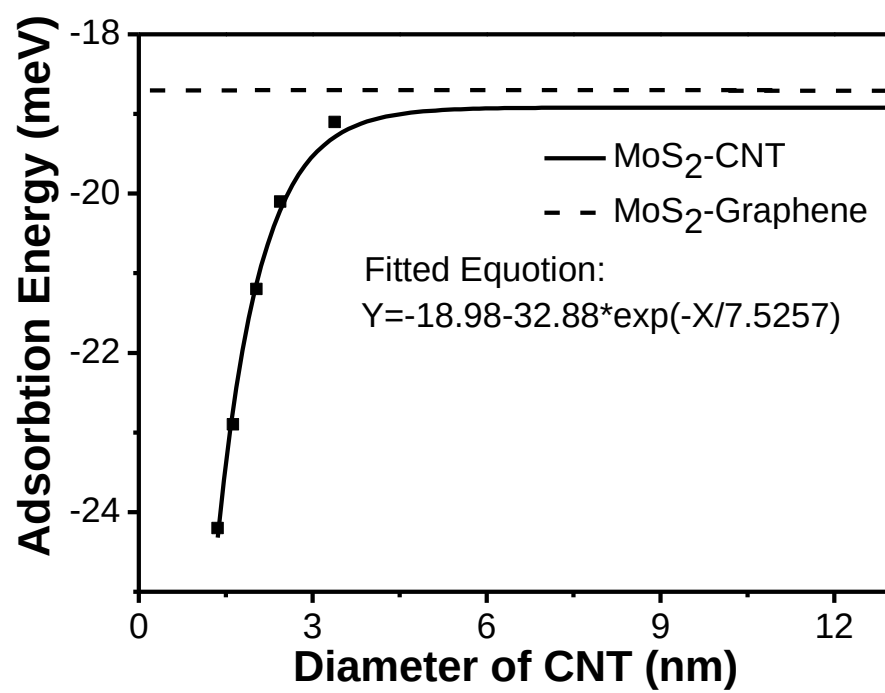


Figure S15 The relationship between the adsorption energies and diameters of CNTs.

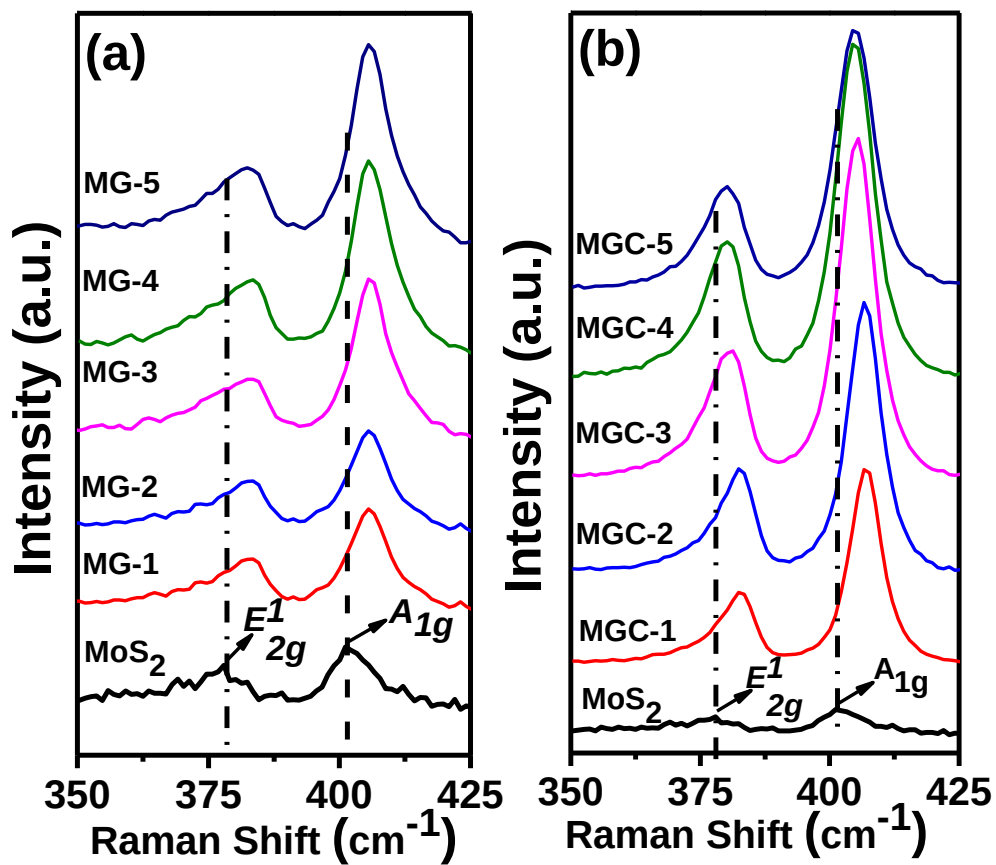


Figure S16 The Raman spectra of E_{2g}^1 and A_{1g} modes of (a) the MoS₂@GF taken at different five points and MoS₂ nanosheets. (b) the MoS₂@GF/CNT taken at five different points and MoS₂ nanosheets.

Table S2 The comparison of electrochemical performances for two electrodes, MoS₂@GF/CNTs, and MoS₂@GF.

Electrode description	1st Specific capacity (mAh g ⁻¹)	1st Coulombic efficiency	Cycling stability (%)	Rate performance
MoS ₂ @GF/CNTs	1568 mAh g ⁻¹ at 100 mA g ⁻¹	79.8%	1125 mAh g ⁻¹ after 80 cycles at 200 mA g ⁻¹	984, 927 and 820 mAh g ⁻¹ at 1000, 2000 and 5000 mA g ⁻¹
MoS ₂ @GF	1502 mAh g ⁻¹ at 100 mA g ⁻¹	65.2%	798 mAh g ⁻¹ after 80 cycles at 200 mA g ⁻¹	713, 664 and 565 mAh g ⁻¹ at 1000, 2000 and 5000 mA g ⁻¹

Table S3 Calculated adsorption energies of MoS₂ cluster at various active sites of Graphene and CNT with different diameters. For all calculated models, one unit contains 20 MoS₂ units and 216 carbon atoms.

Sample	Active site	Diameter (nm)	Adsorption energy E _{ab} (meV)
CNT	(10, 10)	T	1.3597
	(12, 12)	T	1.6272
	(15, 15)	T	2.0328
	(18, 18)	T	2.44
	(25, 25)	T	3.38
Graphene	H	–	-18.7

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