

## **Embedding heterostructured MnS/Co<sub>1-x</sub>S nanoparticles in porous carbon/graphene for superior lithium storage**

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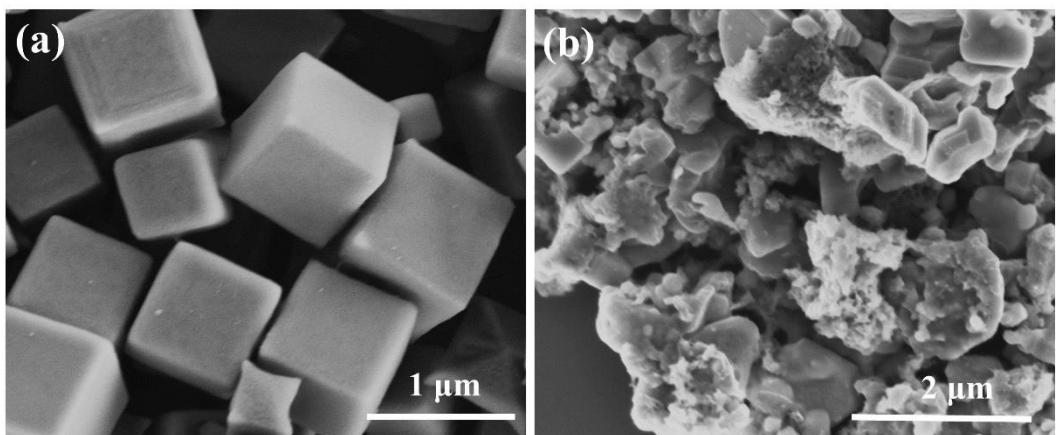
**Table S1.** Comparison of electrochemical performances of MnS/Co<sub>1-x</sub>S@C@rGO composites electrode with previously reported manganese- and cobalt-based electrodes.

<b>Products</b>	<b>Synthetic method</b>	<b>Electrode formulation<sup>a</sup></b>	<b>Cycling stability(A/B/n)<sup>b</sup></b>	<b>Ref.</b>
MnS/Co <sub>1-x</sub> S@C@rGO	Hydrothermal and thermal annealing	70:20:10	1018/1/500	This work
MnS@C	In situ co-pyrolysis	80:10:10	200/0.5/800	[S1]
coral-like α-MnS-NC	Hydrothermal and thermal annealing	80:10:10	699/0.5/500	[S2]
MnS/RGO	Hydrothermal	70:20:10	830/0.5/100	[S3]
MnS microboxes	Solvothermal and thermal annealing	70:20:10	495/0.2/100	[S4]
MnO/C microsheets	Thermal annealing	80:10:10	798/0.1/50	[S5]
Mn <sub>3</sub> O <sub>4</sub> tetragonal bipyramids	Hydrothermal	70:20:10 (CMC) <sup>c</sup>	822/0.2/50	[S6]
MnO/C-N	Immersion-annealing route	80:10:10	513/0.3/400	[S7]
CoS <sub>2</sub> /NCNTF	MOF-derivation	70:20:10	937/1/160	[S8]
Co <sub>9</sub> S <sub>8</sub> /N-C	MOF-derivation	70:10:20 (CMC) <sup>c</sup>	784/0.5/400	[S9]
NC/CoS <sub>2</sub>	MOF-derivation	70:20:10	560/0.1/50	[S10]
CNT@CoS@C	Thermal annealing	80:10:10 (CMC) <sup>c</sup>	1010/0.5/200	[S11]
hollow CoS <sub>2</sub> @C	Solvothermal and the calcination	80:10:10 (PTFE) <sup>d</sup>	730/0.5/200	[S12]

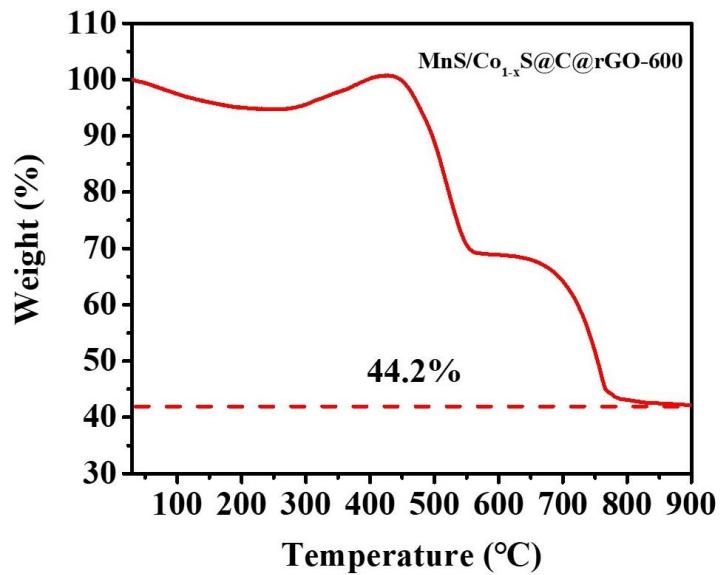
<sup>a</sup>means weight ratio of the active material, carbon and binder. PVDF was used as binder if not mentioned. Other values used were specified.

<sup>b</sup>A/B/n means the capacity of A (mAh g<sup>-1</sup>) at the certain current density of B (A g<sup>-1</sup>).

<sup>c</sup>CMC means carboxymethyl cellulose. <sup>d</sup>PTFE means polytetrafluoroethylene.

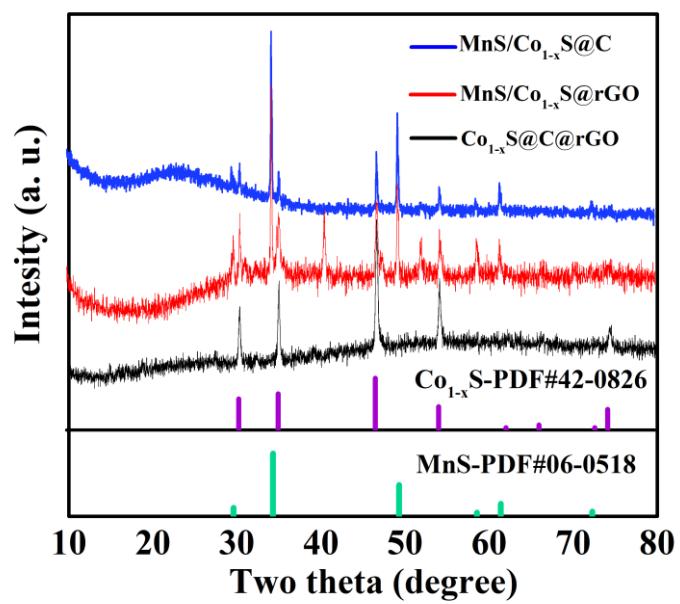


**Fig. S1** FESEM images of (a)  $\text{Mn}_3[\text{Co}(\text{CN})_6]_2 \cdot 9\text{H}_2\text{O}$  precursor and (b)  $\text{MnS}/\text{Co}_{1-x}\text{S}@\text{C}$ .

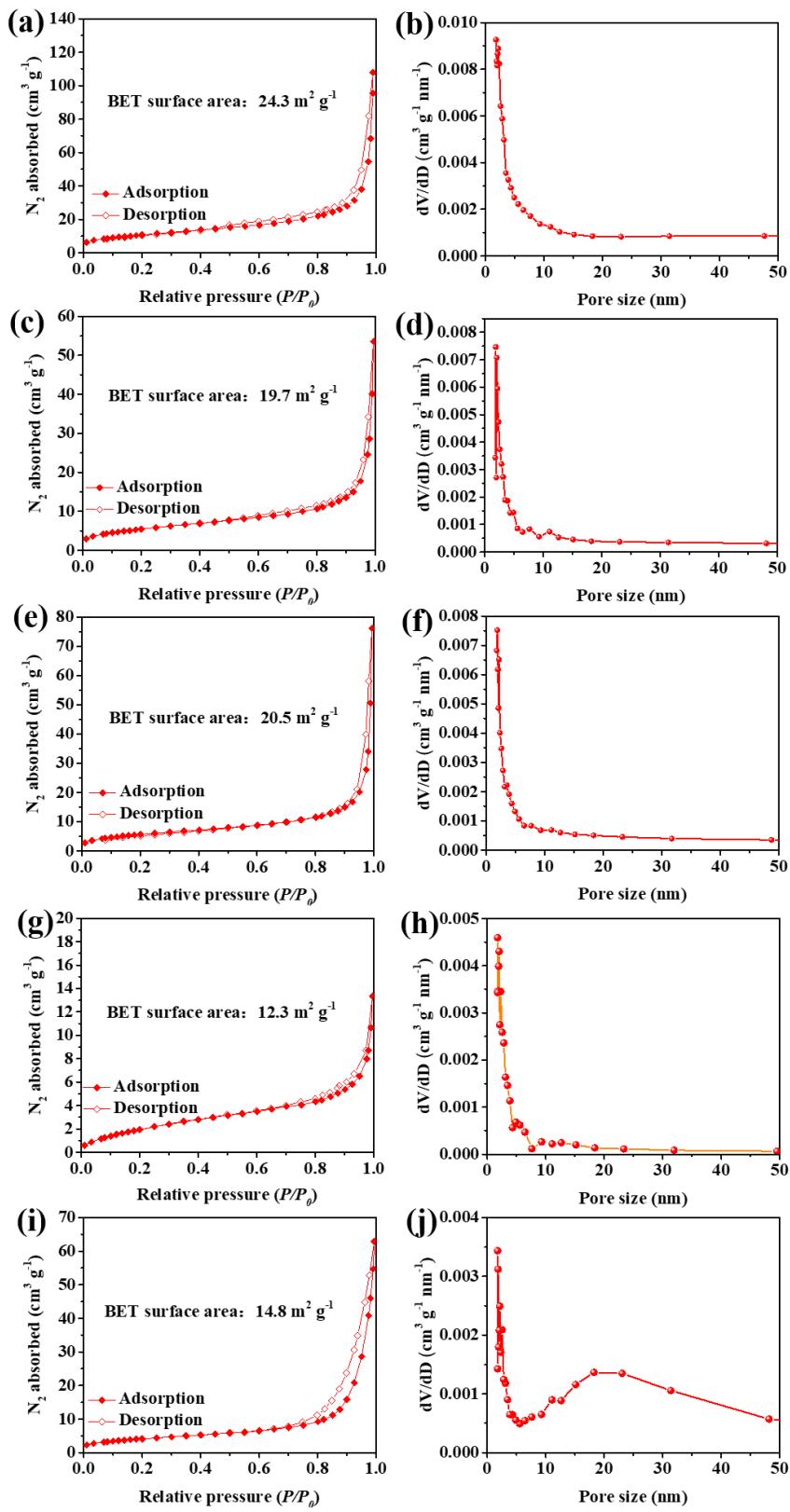


**Fig. S2** TGA curve of the as-prepared MnS/Co<sub>1-x</sub>S@C@rGO-600 composite.

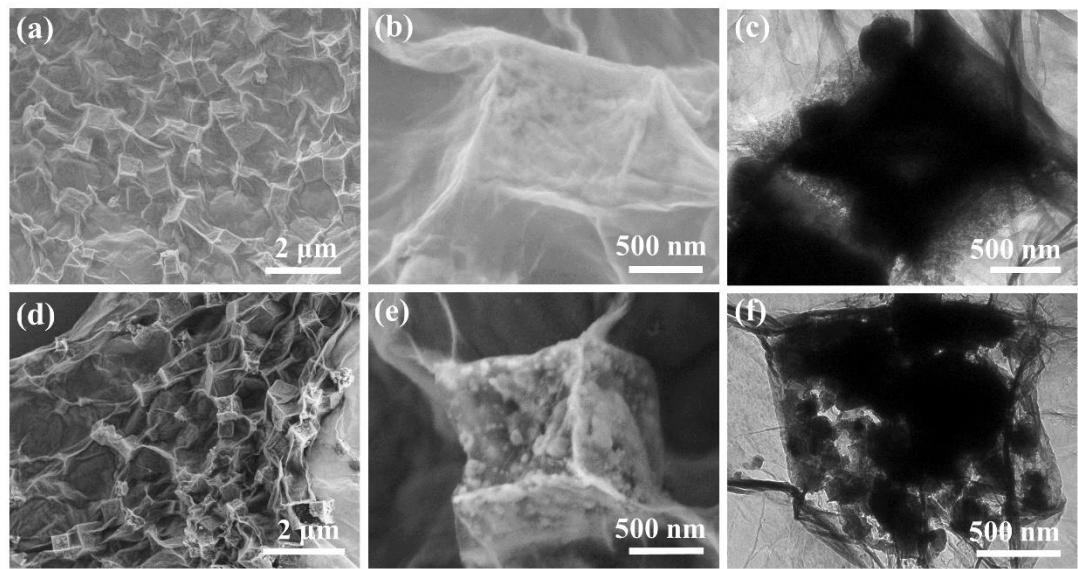
The atomic ratio of Mn, Co, S elements in MnS/Co<sub>1-x</sub>S@C@rGO-600 was determined to be about 1.5:1:2.6 by the ICP measurement.



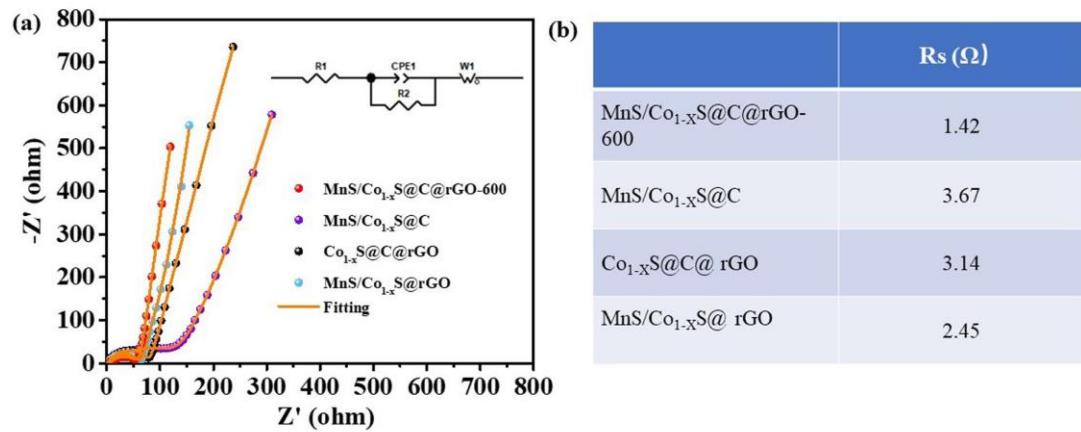
**Fig. S3** XRD patterns of  $\text{MnS}/\text{Co}_{1-x}\text{S}@\text{C}$ ,  $\text{MnS}/\text{Co}_{1-x}\text{S}@\text{rGO}$  and  $\text{Co}_{1-x}\text{S}@\text{C}@\text{rGO}$  composites.



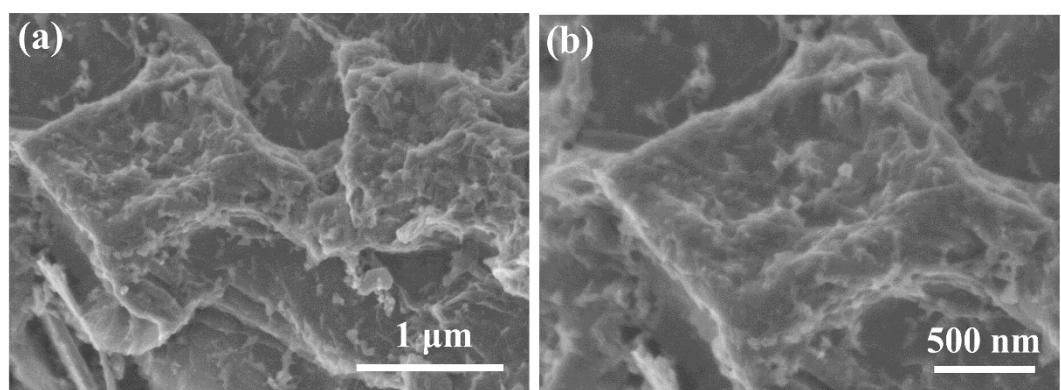
**Fig. S4** N<sub>2</sub> sorption isotherms and the corresponding pore-size distributions based on the Barrett–Joyner–Halenda (BJH) method (a,b) MnS/Co<sub>1-x</sub>S@C@rGO-500, (c,d) MnS/Co<sub>1-x</sub>S@C@rGO-700, (e,f) Co<sub>1-x</sub>S@C@rGO, (g,h) MnS/Co<sub>1-x</sub>S@rGO and (i,j) MnS/Co<sub>1-x</sub>S@C composites.



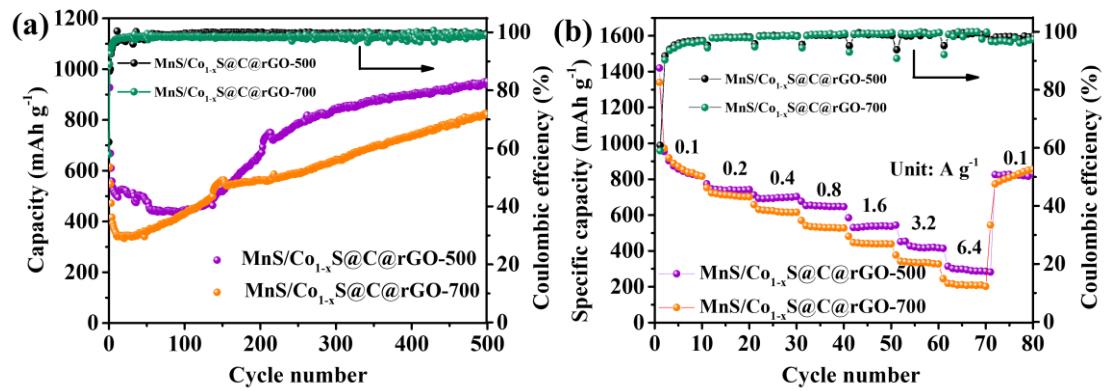
**Fig. S5** (a,b) FESEM images and (c) TEM image of MnS/Co<sub>1-x</sub>S@C@rGO-500; (d,e) FESEM images and (f) TEM image of MnS/Co<sub>1-x</sub>S@C@rGO-700.



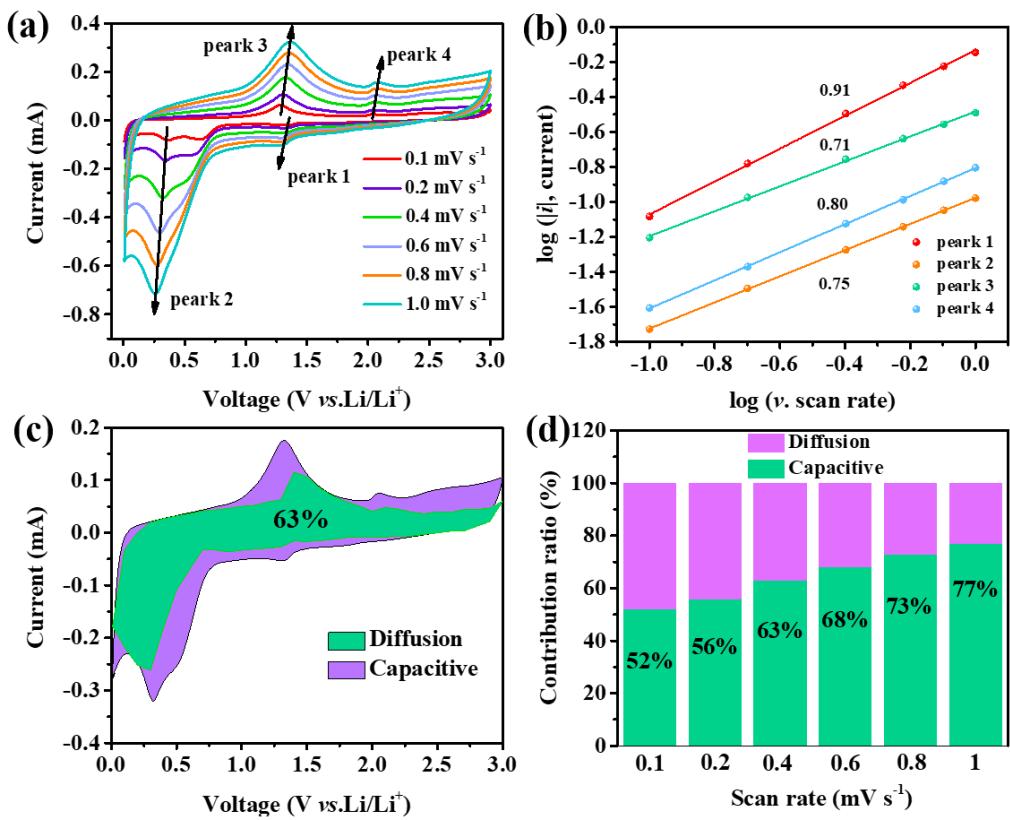
**Fig. S6** (a) The EIS curves and (b)  $R_s$  values of the MnS/Co<sub>1-x</sub>S@C@rGO-600, MnS/Co<sub>1-x</sub>S@C, Co<sub>1-x</sub>S@C@rGO and MnS/Co<sub>1-x</sub>S @rGO composites electrodes.



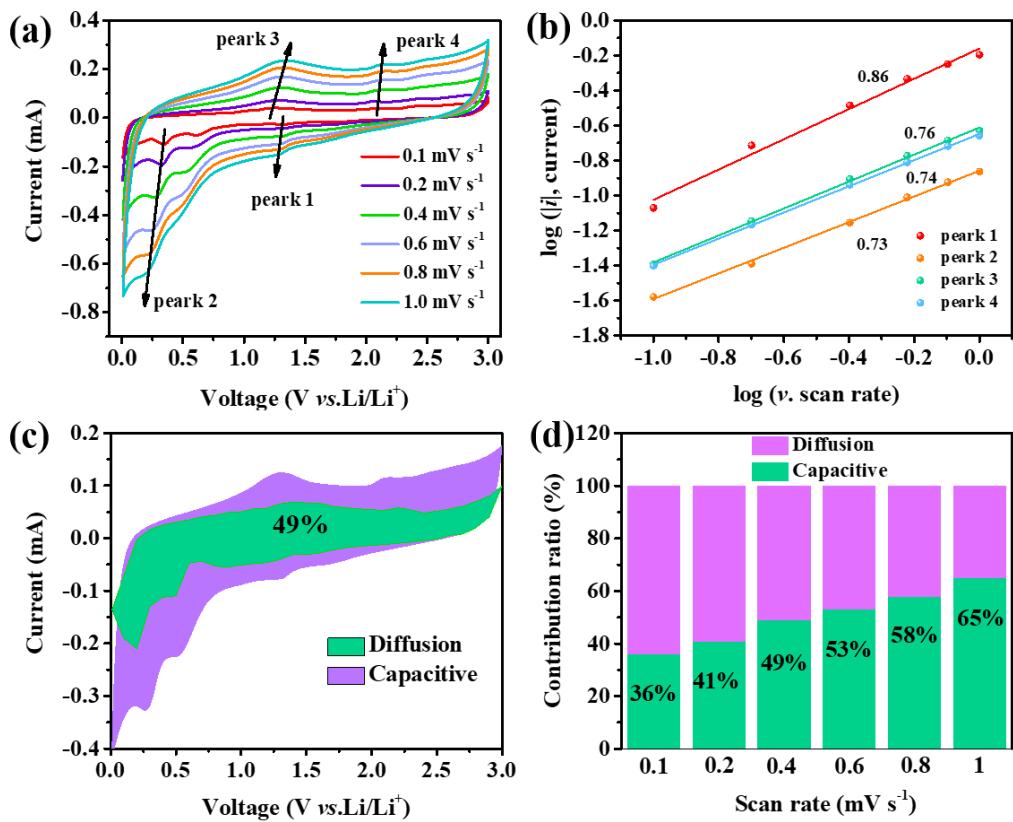
**Fig. S7** (a,b) FESEM images MnS/Co<sub>1-x</sub>S@C@rGO electrode after 500 cycles at a current density of 1 A g<sup>-1</sup>.



**Fig. S8** (a) Cycling performance at  $1 \text{ A g}^{-1}$  and (b) rate capabilities at various current densities of  $\text{MnS/Co}_{1-x}\text{S@C@rGO-500}$  and  $\text{MnS/Co}_{1-x}\text{S@C@rGO-700}$  electrodes.



**Fig. S9** (a) CV curves of MnS/Co<sub>1-x</sub>S@rGO electrode at various sweep rates, (b) log  $i/\log v$  plots at oxidized and reduced state of MnS/Co<sub>1-x</sub>S@rGO electrode, (c) the capacitive and diffusion-controlled contribution to all charge storage of MnS/Co<sub>1-x</sub>S@rGO electrode at the sweep rate of 0.4 mV/s, (d) the normalized contribution percentage from capacitive-controlled and diffusion-controlled behaviour for all charge storage at different sweep rates.



**Fig. S10** (a) CV curves of MnS/Co<sub>1-x</sub>S@C electrode at various sweep rates, (b) log i<sub>||</sub>/log v plots at oxidized and reduced state of MnS/Co<sub>1-x</sub>S@C electrode, (c) the capacitive and diffusion-controlled contribution to all charge storage of MnS/Co<sub>1-x</sub>S@C electrode at the sweep rate of 0.4 mV/s, (d) the normalized contribution percentage from capacitive-controlled and diffusion-controlled behaviour for all charge storage at different sweep rates.

## References

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