1 Supporting Information

² Facile Preparation of 3D MoS₂/MoSe₂ ³ Nanosheets-Graphene Networks as Efficient ⁴ Electrocatalysts for Hydrogen Evolution Reaction

5 Shengjie Xu[‡], Zhouyue Lei[‡] and Peiyi Wu^{*}

6

7 State Key Laboratory of Molecular Engineering of Polymers, Collaborative Innovation
8 Center of Polymers and Polymer Composite Materials, Department of Macromolecular
9 Science and Laboratory for Advanced Materials, Fudan University, Shanghai 200433, China.

10



11



13 The image (a) is referenced our previous work.¹



2 Fig. S2 TEM images of MoSe₂-graphene hybrid aerogel (MoSe₂-20). (a) smaller size of
3 MoSe₂ nanosheets or MoSe₂ quantum dots load on RGO sheets; (b), (c), (d) HRTEM images
4 of different area of (a). The inset of (a) is the corresponding SAED pattern.



7 Fig. S3 Nitrogen adsorption-desorption isotherms of pure GA, MoS₂-20, and MoSe₂-20.



- 2 Fig. S4 SEM image, elements mapping images, EDX pattern and the element weight contents
- 3 of MoS_2 -graphene hybrid aerogel (MoS_2 -20).
- 4



- 6 Fig. S5 SEM image, elements mapping images, EDX pattern and the element weight contents
- 7 of MoSe₂-graphene hybrid aerogel (MoSe₂-20).
- 8



Fig. S6 XPS spectra of MoS₂-20 (left column) and MoSe₂-20 (right column) composites. (a)
and (b) are XPS spectra; (c) and (d) are the high resolution O_{1s} XPS spectra; (e) and (f) are
the high resolution N_{1s} XPS spectra.



7 Fig. S7 FTIR spectra of (a) GO, (b) MoS₂-20 and (c) MoSe₂-20 composites.

6



1

Fig. S8 TGA curves of MoS₂-graphene hybrid aerogels (a) and MoSe₂-graphene hybrid
aerogels (b) prepared with different dosage of MoS₂ and MoSe₂. (I) 0 mL; (II) 1 mL; (III) 3
mL; (IV) 6 mL; (V) 10 mL; (VI) 15 mL; (VII) 20 mL; (VIII) 25 mL; (IX) 30 mL; 40 mL.



7 Fig. S9 TEM images of MoS₂-graphene hybrid aerogels prepared with different dosage of
8 MoS₂. (a) 1 mL; (b) 3 mL; (c) 6 mL; (d) 10 mL; (e) 15 mL. The inset is the corresponding

9 SAED pattern of each sample.



- 2 Fig. S10 TEM images of MoSe₂-graphene hybrid aerogels prepared with different dosage of
- 3 MoSe₂. (a) 1 mL; (b) 3 mL; (c) 6 mL; (d) 10 mL; (e) 15 mL; (f) 25 mL; (g) 30 mL; (h) 40 mL.
- 4 The inset is the corresponding SAED pattern of each sample.



- 7 Fig. S11 FESEM images of MoS₂-graphene hybrid aerogels prepared with different dosage
- 8~ of MoS_2. (a) 1 mL; (b) 3 mL; (c) 6 mL; (d) 10 mL; (e) 15 mL. The bottom row are the
- 9 magnified FESEM of top row.
- 10



2 Fig. S12 FESEM images of MoSe₂-graphene hybrid aerogels prepared with different dosage
3 of MoSe₂. (a) 1 mL; (b) 3 mL; (c) 6 mL; (d) 10 mL; (e) 15 mL; (f) 25 mL; (g) 30 mL; (h) 40



7 Fig. S13 XRD patterns (a) and Raman spectra (b) of GO (I) and graphene aerogel (II).



2 Fig. S14 Polarization curves of MoS₂ and MoSe₂ nanosheets.



5 Fig. S15 Nyquist plots of MoS₂-graphene hybrid aerogels (a) and MoSe₂-graphene hybrid
6 aerogels (b) prepared with different dosage of MoS₂ and MoSe₂. (I) 1 mL; (II) 3 mL; (III) 6
7 mL; (IV) 10 mL; (V) 15 mL; (VI) 20 mL; (VII) 25 mL; (VIII) 30 mL; (IX) 40 mL.

9 The charge-transfer resistances (R_{ct}) were determined by fitting the impedance spectra 10 using the Z-view software, and in all cases it achieved good agreement between the measured 11 and the fitted data. The charge-transfer resistances were summarized in Table. S1 (MoS₂-12 graphene hybrid aerogels) and Table. S2 (MoSe₂-graphene hybrid aerogels).



2 Fig. S16 Electrical conductivity of MoS₂-graphene hybrid aerogels (a) and MoSe₂-graphene

3 hybrid aerogels (b) prepared with different dosage of MoS_2 and $MoSe_2$.

4



Fig. S17 Typical CV curves (top row) and corresponding differences in the current density at
0.2 V plotted against scan rate (bottom row). (a) and (d) are corresponding to the graphene
aerogel, (b) and (e) are corresponding to MoS₂-20, (c) and (f) are corresponding to MoSe₂-20.

10 We calculated the electrochemical active surface areas of the as-prepared catalysts by 11 measuring their electrochemical double layer capacitances (C_{dl}) using a simple CV method 1 according to previous reports.^{2, 3} In brief, capacitance measurements were performed in a 2 potential range of 0.15-0.25 V vs RHE since there was no obvious electrochemical feature 3 corresponding to Faradic current observed in this region for each catalyst. And the capacitive 4 currents, i.e. $\Delta J@0.2$ V, were then plotted as a function of CV scan rate. It was observed a 5 linear relationship in which the slope twice larger than the C_{dl} Value. And C_{dl} and 6 electrochemical active surface area values were summarized in Table. S1 (MoS₂-graphene 7 hybrid aerogels) and Table. S2 (MoSe₂-graphene hybrid aerogels).

8

9 Table. S1 Summary of the charge-transfer resistance, C_{dl} and electrochemical active surface
10 area values of graphene aerogel and MoS₂-graphene hybrid aerogels.

Samples	Charge-transfer	C _{dl}	C _{dl} Electrochemical active	
	resistance (Ω)	(mF cm ⁻²)	surface area (cm ²)	
Graphene aerogel	25	5.4	19	
MoS ₂ -1	23	6.3	22	
MoS ₂ -3	20	8.9	31	
MoS ₂ -6	20	9.6	34	
MoS ₂ -10	19	11.1	39	
MoS ₂ -15	18	12.5	44	
MoS ₂ -20	17	15.8	56	

Samples	Charge-transfer	C _{dl}	C _{dl} Electrochemical active	
	resistance (Ω)	(mF cm ⁻²)	surface area (cm ²)	
MoSe ₂ -1	22	6.9	24	
MoSe ₂ -3	21	8.6	30	
MoSe ₂ -6	19	10.8	38	
MoSe ₂ -10	19	14.3	50	
MoSe ₂ -15	18	19.1	67	
MoSe ₂ -20	16	20.4	72	
MoSe ₂ -25	16	24.5	86	
MoS ₂ -30	15	28.4	100	
MoSe ₂ -40	14	31.5	111	

1 Table. S2 Summary of the charge-transfer resistance, C_{dl} and electrochemical active surface

\mathbf{a}	1	C M C	1	1 1 1	1
/	area values	OT MONES-	orannene	nynria	aerogeis
_		\mathbf{U}	graphene	II y UI I U	acrogois.
				2	0

3

4 References

- 5 1. S. Xu, D. Li and P. Wu, *Adv. Funct. Mater.*, 2015, **25**, 1127-1136.
- 6 2. Y. Zheng, Y. Jiao, L. H. Li, T. Xing, Y. Chen, M. Jaroniec and S. Z. Qiao, *ACS Nano*,
 7 2014, 8, 5290-5296.
- 8 3. E. Navarro-Flores, Z. Chong and S. Omanovic, J. Mol. Catal. A-Chem., 2005, 226, 179-197.