## **ELECTRONIC SUPPLEMENTARY INFORMATION**

## Sulfur-doped Graphene / Transition Metal Dichalcogenide Heterostructured Hybrids with Electrocatalytic Activity Toward the Hydrogen Evolution Reaction

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**Figure S1**. Thermogravimetric analysis graphs for (a) SG/MoS<sub>2</sub> (3:1) (red), SG/MoS<sub>2</sub> (1:1) (green), and MoS<sub>2</sub> (black), and (b) SG/WS<sub>2</sub> (3:1) (purple), SG/WS<sub>2</sub> (1:1) (cyan), and WS<sub>2</sub> (grey), obtained under nitrogen.



**Figure S2**. (a) EEL spectrum of material displayed in Figure 4c, showing the C-K, S- $L_{2,3}$  and Mo-M<sub>4,5</sub> edges. (b1, b2) EEL spectra of material displayed in Figure 4f, showing the C-K, S- $L_{2,3}$  and W-O<sub>2,3</sub> edges.



Figure S3. Representative SEM image of SG.



**Figure S4.** SEM images for (a)  $MoS_2$ , (b) SG/MoS\_2 (1:1), and (c) SG/MoS\_2 (3:1). EDS for (d)  $MoS_2$ , (e) SG/MoS\_2 (1:1), and (f) SG/MoS\_2 (3:1). The presence of Si is due to the sample holder.



**Figure S5.** SEM images for (a)  $WS_2$ , (b) SG/WS<sub>2</sub> (1:1), and (c) SG/WS<sub>2</sub> (3:1). EDS for (d)  $WS_2$ , (e) SG/WS<sub>2</sub> (1:1), and (f) SG/WS<sub>2</sub> (3:1). The presence of Si is due to the sample holder.



**Figure S6.** Low magnification TEM images of (a) SG/MoS<sub>2</sub> (1:1), and (b) SG/WS<sub>2</sub>, (1:1). Typical MoS<sub>2</sub> and WS<sub>2</sub> features are highlighted with yellow arrows. The scale bar is 200 nm.



**Figure S7.** Cyclic voltammetry curves of (a)  $MoS_2$ , (b) SG/MoS<sub>2</sub> (1:1), (c) SG/MoS<sub>2</sub> (3:1), (d) WS<sub>2</sub>, (e) SG/WS<sub>2</sub> (1:1), (f) SG/WS<sub>2</sub> (3:1) and (g) SG measured in a nitrogen saturated aqueous 0.5 M H<sub>2</sub>SO<sub>4</sub> electrolyte, at a rotation speed of 1,600 rpm and scan rates from 50 to 500 mV/s. Inset: images of the scan rate dependence of the current densities for the corresponding materials.

![](_page_8_Figure_0.jpeg)

**Figure S8.** LSV for HER of (a) SG/MoS<sub>2</sub> (3:1) (red), SG/MoS<sub>2</sub> (1:1) (green), MoS<sub>2</sub> (black), SG/WS<sub>2</sub> (3:1) (purple), SG/WS<sub>2</sub> (1:1) (cyan), WS<sub>2</sub> (grey), SG (pink) and Pt/C (blue) (solid lines) before after 1,000 cycles (dashed lines). The LSV polarization curves were obtained in a nitrogen saturated aqueous 0.5 M H<sub>2</sub>SO<sub>4</sub> electrolyte, at a rotation speed of 1,600 rpm and scan rate of 5 mV/s.