

Supporting Information

Enabling Colloidal Synthesis of Edge-Oriented MoS₂ with Expanded Interlayer Spacing for Enhanced HER Catalysis

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Statistical Analysis of MoS₂ Nanosheets in the MoS₂/rGO Composite Samples

The SEM images of the MoS₂/rGO composite samples are used to estimate the density (i.e., number of MoS₂ nanosheets per unit of area) and projected edge length of the MoS₂ nanosheets anchored on the rGO nanosheets. Because the MoS₂ nanosheets protrude out of the basal surface of the rGO nanosheets, the edges of the MoS₂ nanosheets exhibit much brighter imaging contrast compared to the underneath rGO nanosheets. The stark imaging contrast enables us to count the number of MoS₂ nanosheets in the defined interesting regions. We first draw a square of 1 $\mu\text{m} \times 1 \mu\text{m}$ over a SEM image and count the number of bright lines (corresponding to MoS₂ nanosheets), resulting in the density of MoS₂ nanosheets (in a unit of sheets/ μm^{-2}). Five squares are drawn and the corresponding densities are averaged and plotted in Figure 2D.

Because of the mechanical flexibility and somehow random orientations of individual MoS₂ nanosheets, it is unrealistically difficult to precisely measure either surface areas or edge lengths of the as-grown MoS₂ nanosheets. Since there is no specific structural relationship between the MoS₂ nanosheets and the supporting rGO nanosheets (i.e., no epitaxial relationship between MoS₂ and rGO), it is reasonably considered that the growth of the MoS₂ nanosheets is isotropic. Therefore, the overall length of the peripheral edges of each MoS₂ nanosheet is proportional to its projected edge length in the SEM image. For example, a MoS₂ nanosheet grows from one nucleation site and expands to a half circle with a radius of r through isotropic growth. The overall peripheral edge length is $2r + \pi r$ while the projected length is $2r$. The ratio between the projected edge length and the overall edge length is $2/(2 + \pi)$, which is a constant independent of size of the nanosheet, indicating the proportional relationship between the projected edge length and the overall peripheral edge length. It is reasonable to use the projected edge length to study the relationship of catalytic performance on the overall peripheral edge length of the MoS₂ nanosheets. The averaged projected edge lengths of the MoS₂ nanosheets synthesized at the different temperatures are presented in Figure 2D.

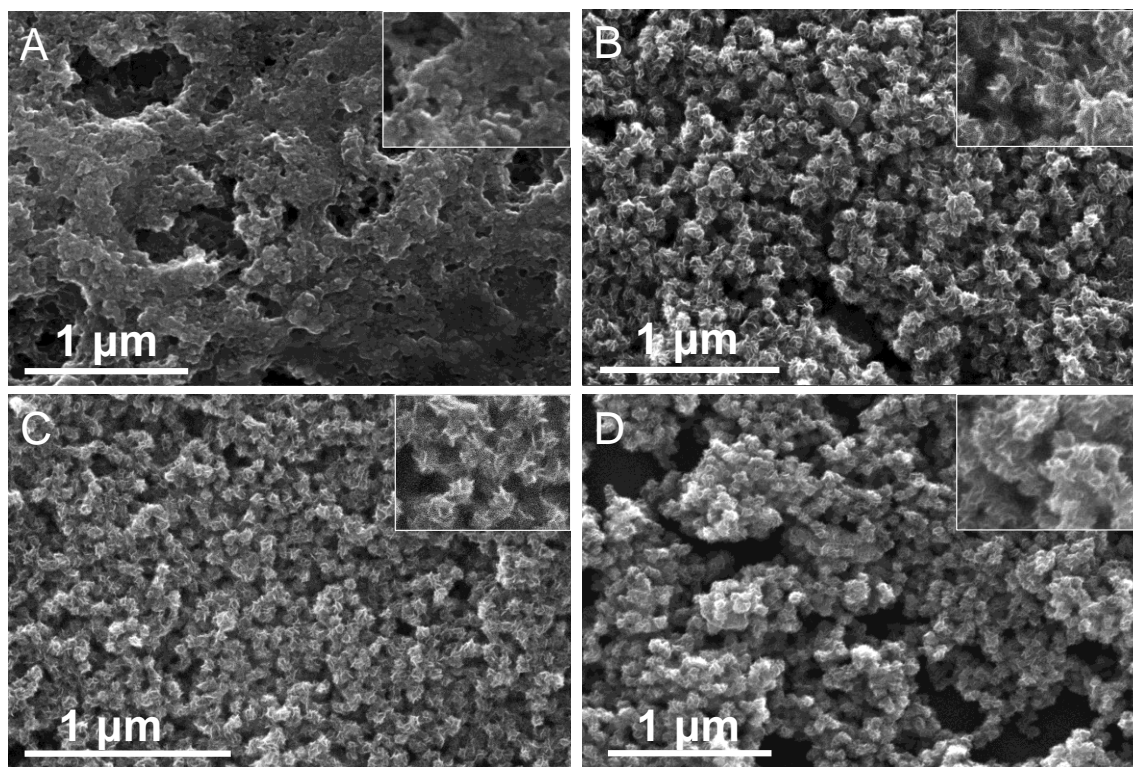


Figure S1. SEM images of IE MoS₂ nanostructures synthesized from reactions at (A) 200 °C, (B) 220 °C, (C) 240 °C, and (D) 260°C in the absence of GO sheets.

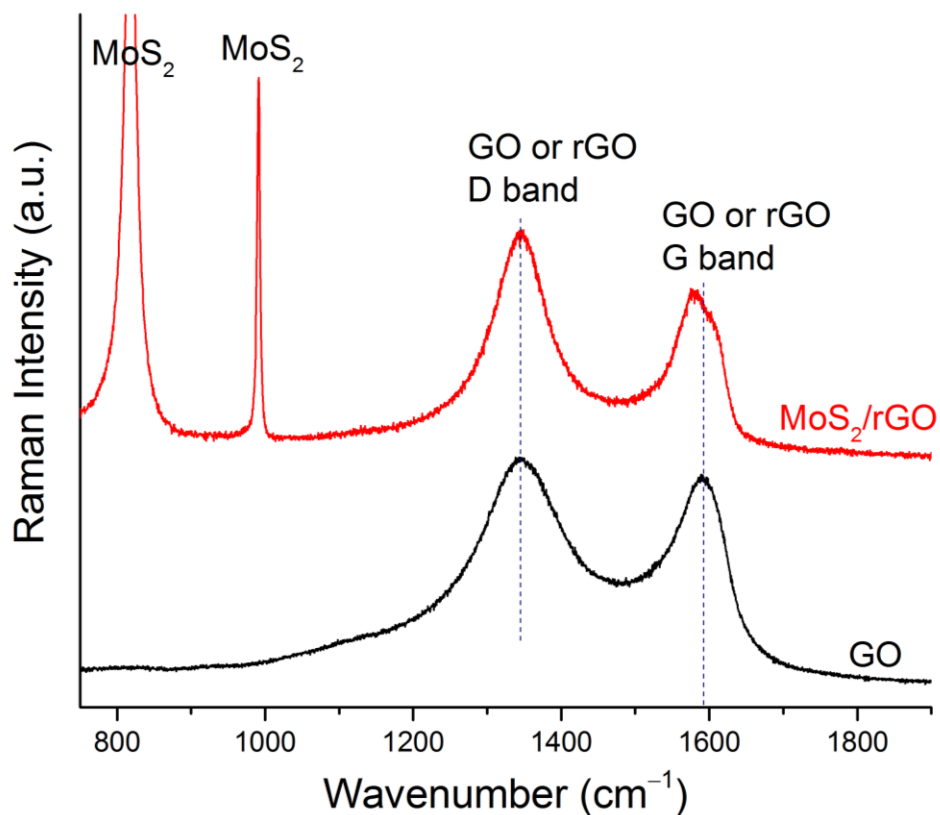


Figure S2. Raman spectra of the GO nanosheets and the MoS₂/rGO composite synthesized at 260 °C (as that shown in Fig. 1). The G band of the GO nanosheets exhibits a well-defined peak at 1591 cm⁻¹. The G band of the rGO nanosheets in the MoS₂/rGO composite becomes asymmetric with the major peak shift to a lower energy of 1578 cm⁻¹ along with the shoulder peak. Such variation of the G band is consistent with the reduction of GO to rGO.

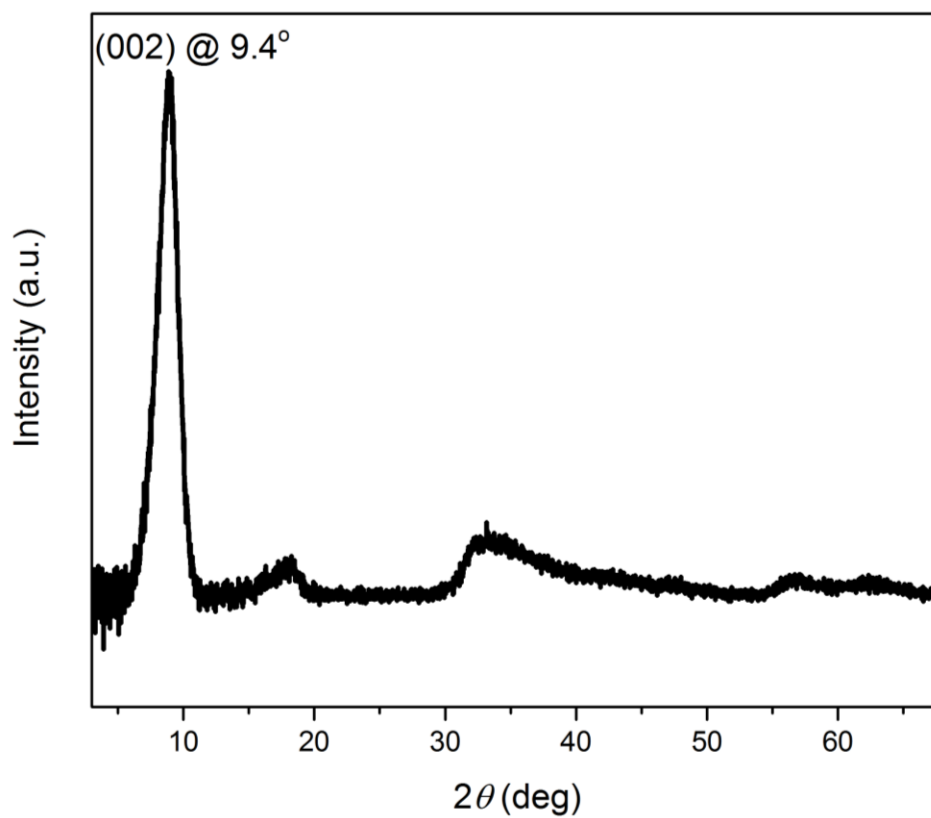


Figure S3. XRD pattern of the MoS₂ flowers synthesized in the absence of GO nanosheets. The sample is as shown in Figure S1D.

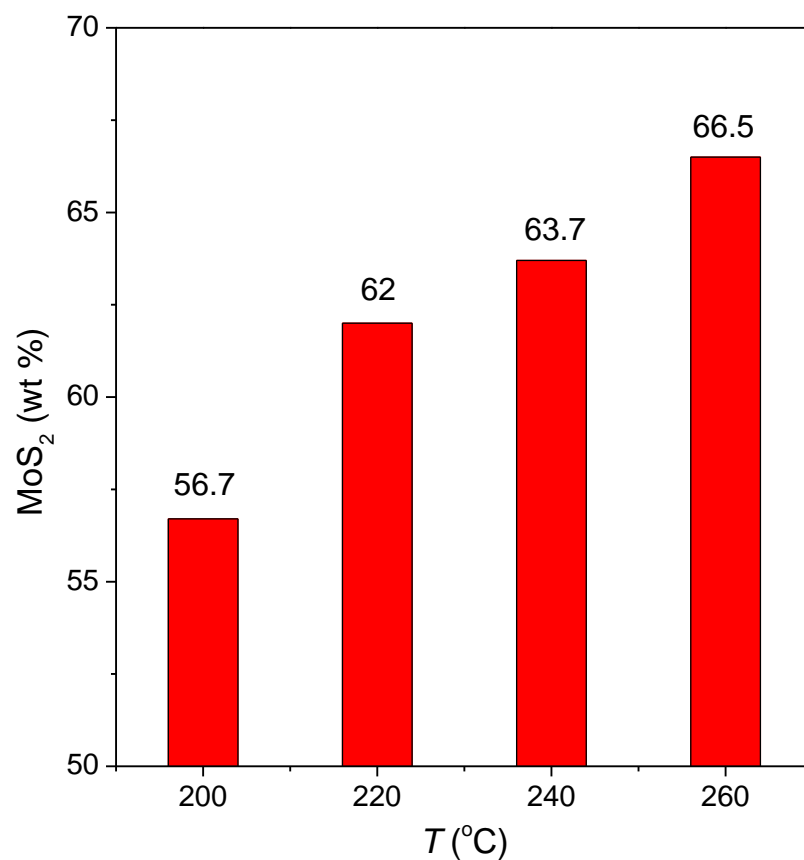


Figure S4. Weight percentage of MoS_2 in the EO&IE MoS_2/rGO samples synthesized from reactions at different temperatures.

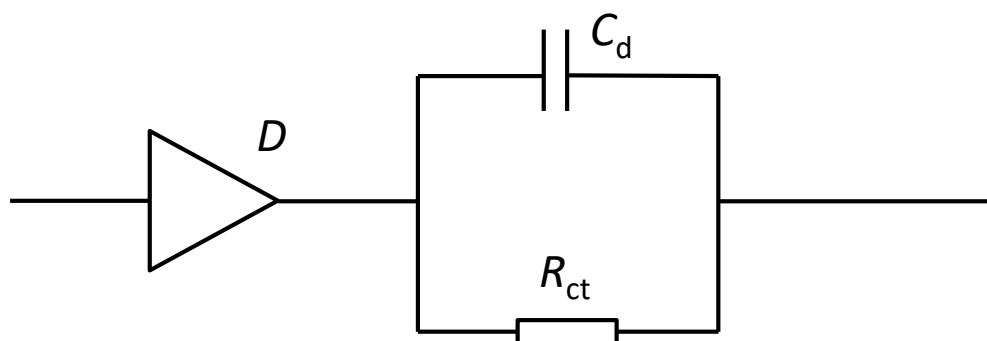


Figure S5. Equivalent circuit corresponding to HER process on the EO&IE MoS_2/rGO catalysts.