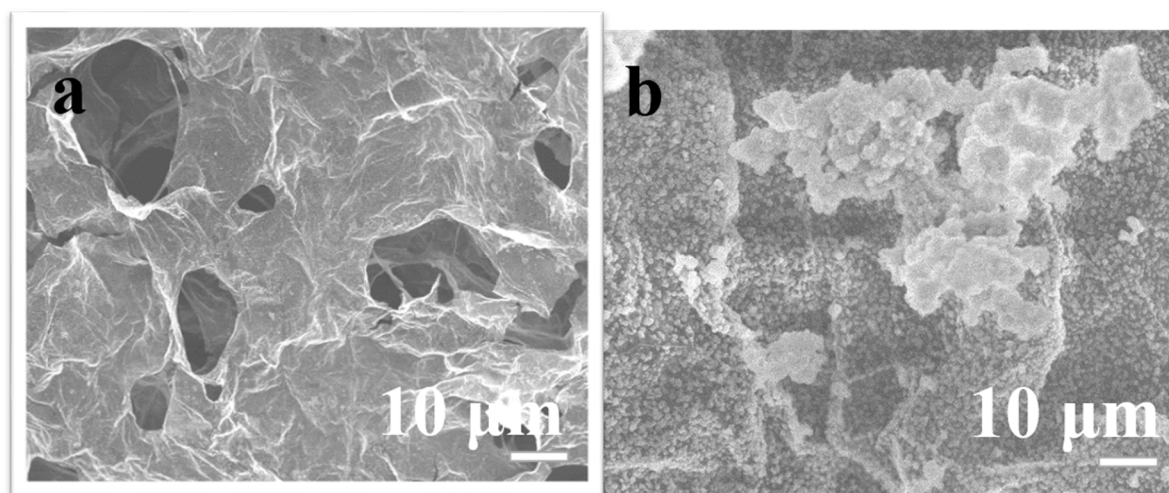


# Supporting Information: MnS-Nanoparticles-Decorated Three-Dimensional Graphene Hybrid as Highly Efficient Bifunctional Electrocatalyst for Hydrogen Evolution Reaction and Oxygen Reduction Reaction

Khalil ur Rehman, Shaista Airam, Long Song, Jian Gao, Qiang Guo, Yukun Xiao and Zhipan Zhang \*

Key Laboratory of Photoelectronic/Electrophotonic Conversion Materials, Key Laboratory of Cluster Science, Ministry of Education of China, School of Chemistry and Chemical Engineering, Beijing Institute of Technology, Beijing 100081, China; meer@bit.edu.cn (K.u.R.); shaistaerum25@yahoo.com (S.A.); longsongphd@gmail.com (L.S.); gaojian8908@163.com (J.G.); guoqiang15124780173@gmail.com (Q.G.); yukun.xiao6@gmail.com (Y.X.)

\* Correspondence: Correspondence to: zhipan@bit.edu.cn (Z.Z.); Tel.: +86-xxx-xxxx-xxxx



**Figure S1.** SEM images of SILAR-1 (a) and SILAR-10 (b).

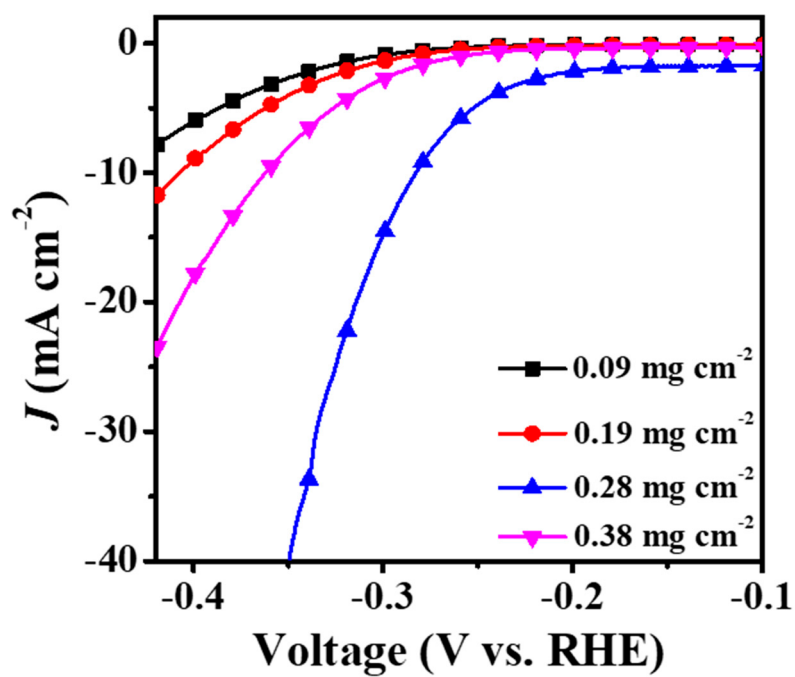


Figure S2. The Polarization curves of MnS-NPs@3DG with different loading weight.

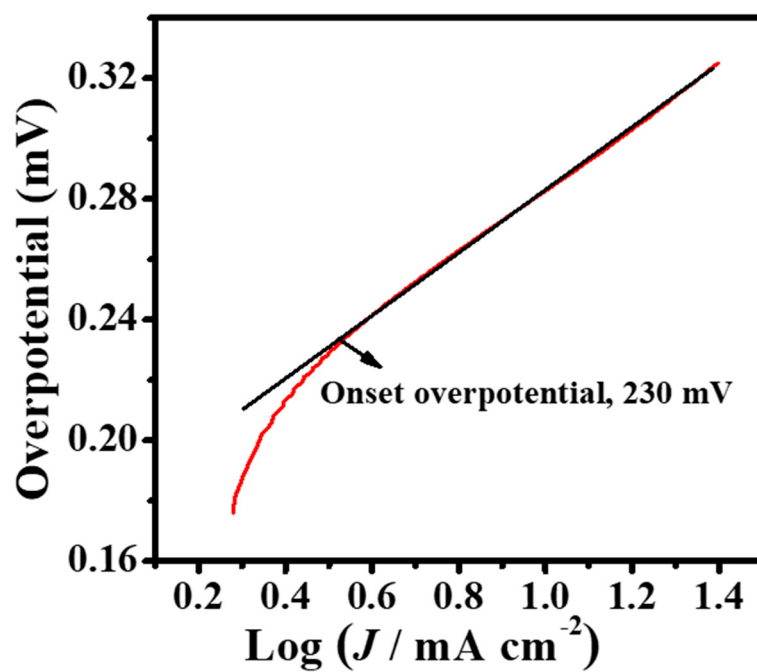
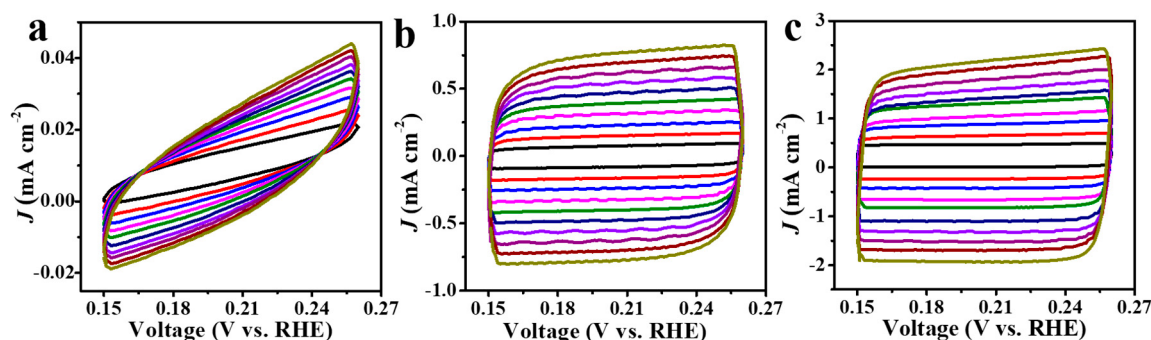


Figure S3. The Tafel plot of the MnS-NPs@3DG.



**Figure S4.** CV curves of bulk MnS (a), 3DG (b) and MnS-NPs@3DG (c) from 0.15 to -0.25 V vs. RHE at different scan rates.

**Table S1.** Comparison of recently reported highly active Mn-based HER electrocatalyst in the literature.

| Electrocatalyst                                       | Electrolyte | Mass loading (mg cm <sup>-2</sup> ) | Current density (mA cm <sup>-2</sup> ) | Overpotential (V vs. RHE) | Tafel slope (mV dec <sup>-1</sup> ) | Ref.      |
|-------------------------------------------------------|-------------|-------------------------------------|----------------------------------------|---------------------------|-------------------------------------|-----------|
| MnNi                                                  | 0.1 M KOH   | 0.28                                | 10                                     | 0.36                      | N/A                                 | [1]       |
| H-MnO <sub>2</sub> -MoS <sub>2</sub> nanohybrids      | 0.1 M KOH   | 0.27                                | 10                                     | 0.23                      | 66                                  | [2]       |
| Mn <sub>0.85</sub> Ni <sub>0.15</sub> PS <sub>3</sub> | 1 M KOH     | 1.25                                | 10                                     | 0.63                      | 178                                 | [3]       |
| MnO <sub>2</sub> /NiO@Ni                              | 1 M KOH     | N/A                                 | 10                                     | 0.23 <sup>+</sup>         | 38                                  | [4]       |
| Exf-MnPS <sub>3</sub>                                 | 1 M KOH     | 0.14                                | 10                                     | 1.09                      | N/A                                 | [5]       |
| Exf-MnPSe <sub>3</sub>                                | 1 M KOH     | 0.14                                | 10                                     | 0.99                      | N/A                                 | [5]       |
| MnS-NPs@3DG                                           | 0.1 M KOH   | 0.28                                | 10                                     | 0.27                      | 100                                 | This work |

**Table 2.** Comparison of recently reported highly active Mn-based ORR electrocatalyst in the literature.

| Electrocatalyst                          | Electrolyte | Mass loading (mg cm <sup>-2</sup> ) | E <sub>onset</sub> (V vs. RHE) | E <sub>1.0 mA cm<sup>-2</sup></sub> (V vs. RHE) | E <sub>reduction</sub> (V vs. RHE) | Ref.      |
|------------------------------------------|-------------|-------------------------------------|--------------------------------|-------------------------------------------------|------------------------------------|-----------|
| MnS-NT/rGO                               | 1.0 M KOH   | 0.10                                | 1.00                           | 0.94                                            | 0.61                               | [6]       |
| NCNT/Co <sub>x</sub> Mn <sub>1-x</sub> O | 1.0 M KOH   | 0.21                                | 0.97                           | 0.87                                            | N/A                                | [7]       |
| Mn <sub>3</sub> O <sub>4</sub> /C        | 1.0 M KOH   | 0.20                                | 0.92 <sup>a,b</sup>            | 0.82 <sup>a,b</sup>                             | 0.76 <sup>b</sup>                  | [8]       |
| MnO <sub>x</sub> /C                      | 1.0 M KOH   | 0.04                                | 0.93 <sup>a,b</sup>            | 0.84 <sup>a,b</sup>                             | 0.56 <sup>b</sup>                  | [9]       |
| MnS/S,N-3DG                              | 1.0 M KOH   | 0.42                                | 0.85 <sup>b</sup>              | 0.69 <sup>a,b</sup>                             | 0.63 <sup>b</sup>                  | [10]      |
| MnS/G (50%)                              | 1.0 M KOH   | 0.60                                | 0.83                           | 0.74 <sup>a</sup>                               | 0.6                                | [11]      |
| MnS-NPs@3DG                              | 0.1 M KOH   | 0.10                                | 0.9                            | 0.7                                             | 0.75                               | This work |

<sup>a</sup> Estimated value; observed values based on the Figures in the article; <sup>b</sup> Potential conversion of E<sub>vs</sub> (different reference electrodes) to E<sub>vs</sub>, RHE were shown in Table S2; E<sub>onset</sub>: Onset potential of the electrocatalysts for ORR; E<sub>1.0 mA cm<sup>-2</sup></sub>: Polarization potential of the electrocatalysts at 1.0 mA cm<sup>-2</sup> for ORR; E<sub>reduction</sub>: Reduction potential of the electrocatalyst for ORR.

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