Supporting Information

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Fabrication of high-performance flexible photodetectors based on Zn-doped MoS₂/graphene hybrid fibers

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Experimental Section

Preparation of GO thin film and GO fibers

Graphene oxide (GO) was synthesized from natural graphite via a modified Hummer's method. Natural graphite flakes (Sigma Aldrich) were oxidized using NaNO₃, H₂SO₄, KMnO₄, and H₂O₂, and the as-synthesized GO was washed several times by centrifugation. The GO sheets were exfoliated by ultra-sonication. GO solution (20 mg) in 20 ml distilled water was spin-coated onto a SiO₂ substrate at 2000 rpm for 30s to prepare GO film. The reduced GO film was formed by heating at 600°C with a flow of Ar for 5 mins. For the preparation of GO fiber, a glass pipe with an inner diameter of 1.5 mm was utilized. The GO solution with a high concentration was injected into the glass pipe, which was then sealed at both ends with polyimide tape and heated to 80°C for 12 h. Water was then injected into the pipe to delaminate rGO fibers.

CVD Growth of MoS₂ Layer on GO thin film and GO fibers

A layer of *p*-THPP was deposited on the GO layer and GO fibers by thermal evaporation, and metalation of the *p*-THPP layer was carried out by introducing diethylzinc (DEZ) as a metal precursor in the atomic layer deposition (ALD) process. The Mo solution was prepared by dissolving 0.1 M ammonium heptamolybdate (Fluca, 99%) in 10 mL of distilled water. The solution was coated onto SiO₂ substrates by spin-coating at 2,000 rpm for 30 s. 0.1 g of sulfur powder (SAMCHUN, 98.0%) as the sulfur source was located upstream in the reactor. The distance between the sulfur and Mo sources was 19 cm. MOS_2 nanosheets were synthesized at 600°C under ~2 Torr while introducing Ar gas (500 sccm) for 5 min.

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Figure S2. AFM image of the spin-coated *p*-THPP layer.

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Figure S3. (a) Optical, (b) SEM, and (c) AFM images of GO/SiO₂, *p*-THPP/GO/SiO₂, and Zn-THPP/SiO₂, respectively.



Figure S4. Photograph of as-grown MoS_2 nanosheets on rGO film (4 x 4 cm²).

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Figure S5. (a) Cross-section SEM image of as-fabricated GO fiber. (b) Top view SEM image of as-fabricated GO fiber. (c) Real image of as fabricated GO fiber.



Figure S6. Raman spectra recorded from different positions of on (a) Zn-doped MoS_2 on rGO TF, and (b) Zn-doped MoS_2 on rGO fiber.



Figure S7. XPS depth profiles of the (a) Mo 3d, (b) S 2p, (c) Zn 2p, (d) C 1s, (e) O 1s and (f) Si 2p core level spectra for Zn-doped MoS_2 film as a function of etching time.



Figure S8. (a) Photoresponse characteristics of the device based on the Zn-doped MoS_2 -rGO (DEZ injection time : 80 s) over 9 junctions, (b) photograph showing 9 junctions in the Zn-doped MoS_2 -rGO/SiO₂-based device, and (c) plot of the drain current of junctions under light illumination.

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Figure S9. (a) XPS spectra of N 1s core level for (i) p-THPP thin films, Zn(II)THPP thin films formed by introducing Zn precursor for (ii-vi) 5, 20, 40, 80, and 120 s. (b) Plots for the peak area ratios of - NH-, -N= and -Zn-N- extracted from N 1s core level spectra as a function of DEZ exposure time. (c) The photocurrent of the devices based on Zn-doped MoS₂/GO nanosheets as a function of DEZ exposure time.

| | Year | Wavelength (nm) | Bias voltage (V) | Power density | Responsivity (A/W) |
|----------|------|--------------------|---------------------|--------------------------|-----------------------|
| Ref. 1 | 2014 | 632.8 nm | 0.1 V | 0.645 uW/cm ² | $10^4 \mathrm{mA/W}$ |
| Ref. 2 | 2014 | 850 nm | 0.1 V | 1 mW/cm ² | 8 mA/W |
| Ref. 3 | 2015 | 1440 nm | 2 V | 0.4 mW/cm^2 | 1.26 A/W |
| Ref. 4 | 2016 | 514 nm | -2 V | 3 mW/cm ² | ~25 mA/W |
| Ref. 5 | 2017 | 540 nm | 5 V | 0.1 mW/cm^2 | 40 mA/W |
| Our work | 2017 | visible | 2 V | 125.2 W/m ² | 5.73A/W |

Table S1. Photodetector performance for the Zn-doped MoS₂/graphene hybrid fibers to other references.

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