Supporting Information

Fabrication and application of flexible graphene silk composite film electrodes decorated with spiky Pt nanospheres

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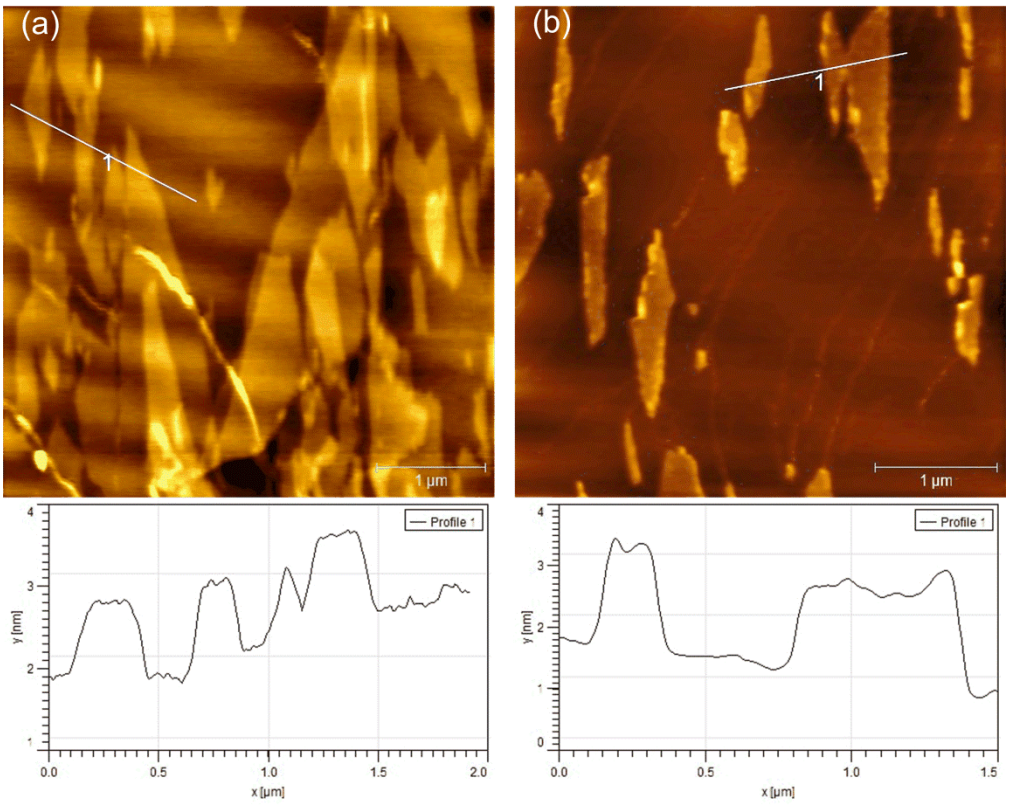

Figure S1. AFM images of GO spin-coating on mica substrates with solution concentrations of 0.5 mg/mL (a) and 0.05 mg/mL (b). The corresponding height profiles are shown at the bottom.

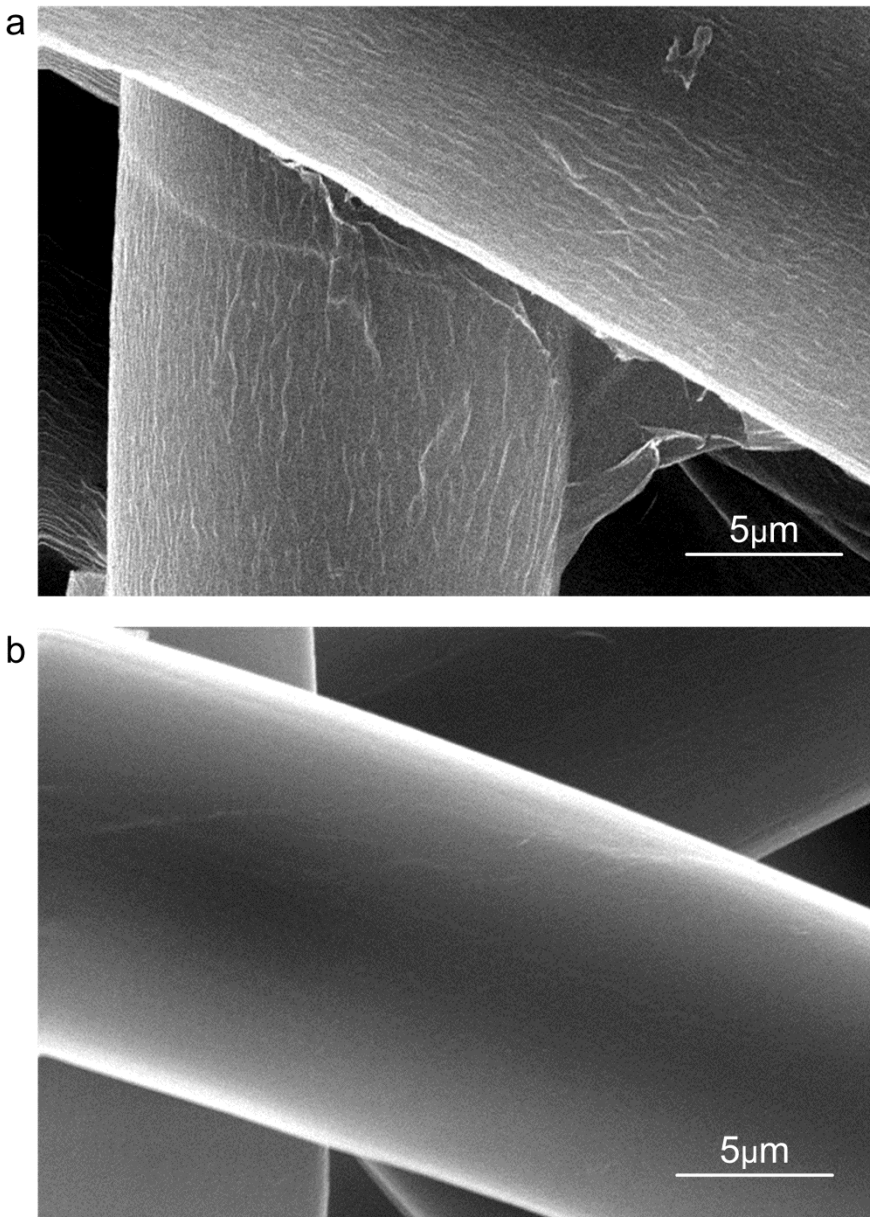

Figure S2. (a) SEM image of graphene coated silk fibre. (b) SEM image of pure silk fibre.

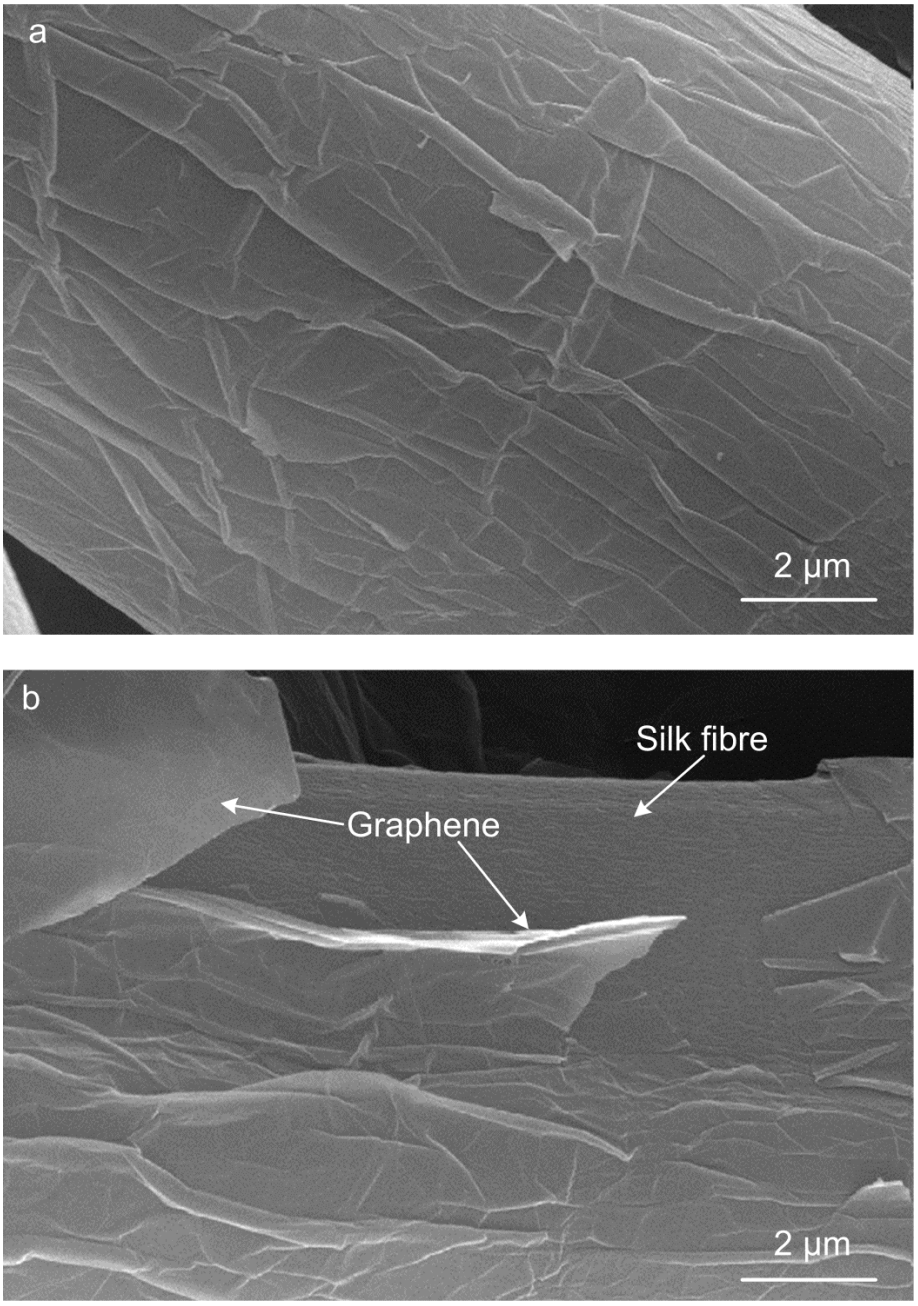

Figure S3. SEM images of GO coated silk fibres before (a) and after (b) chemical reduction.

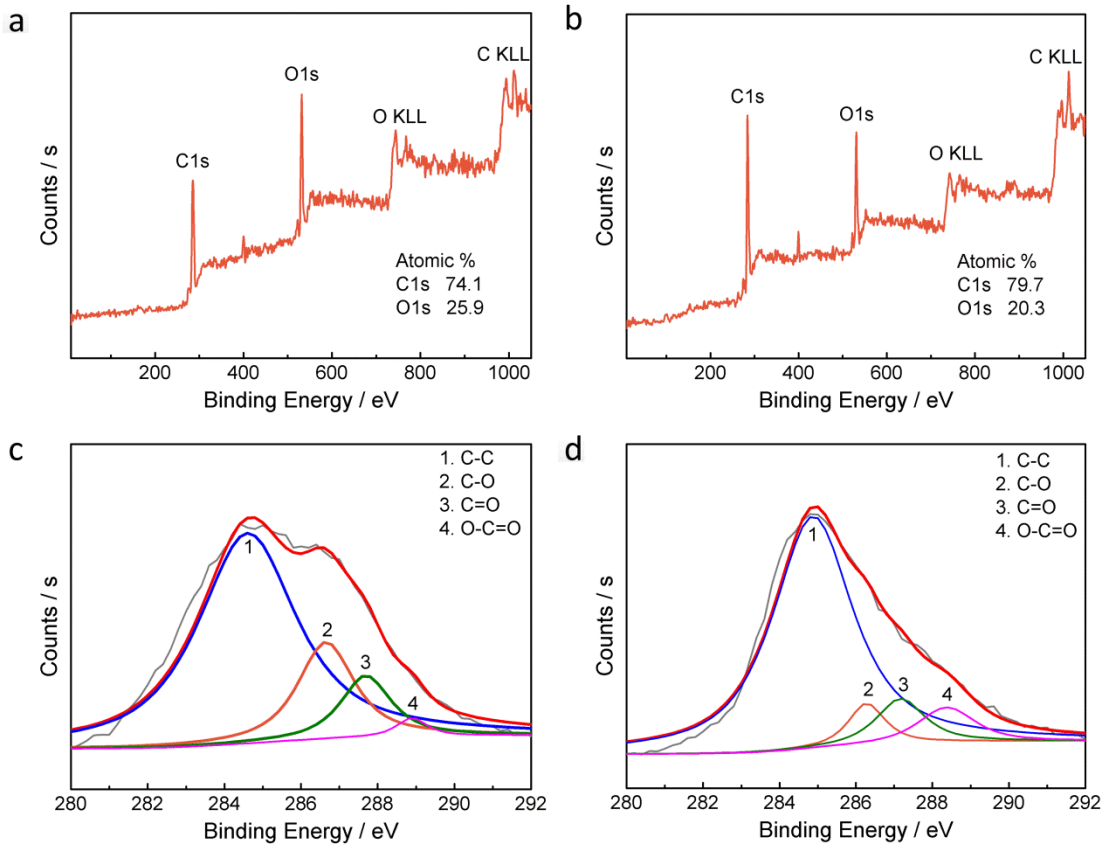

Figure S4. Full range XPS spectra and C 1s XPS spectra of GO before (a, c), and after chemical reduction (b, d).

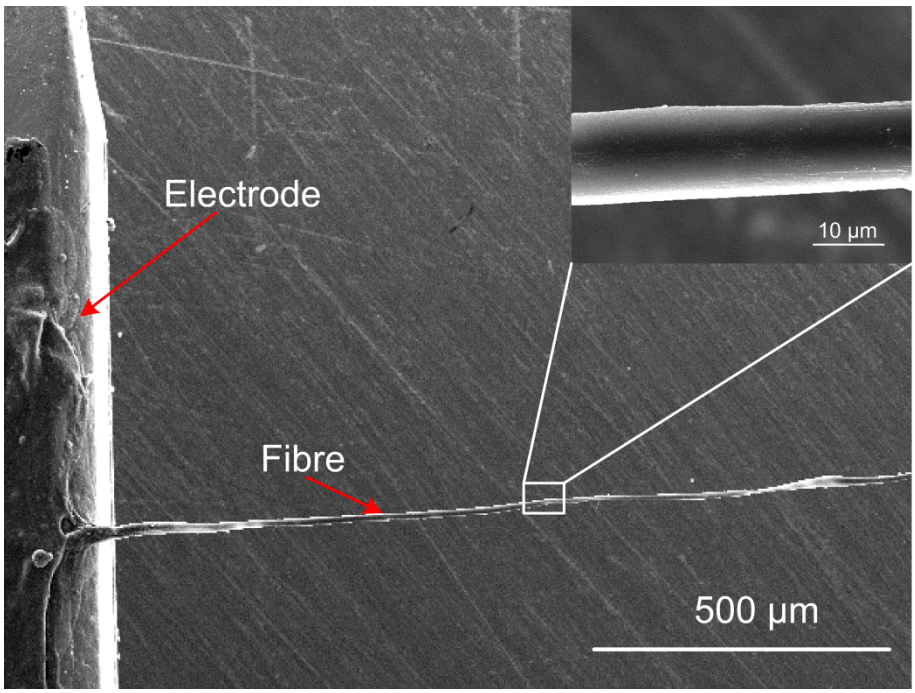

Figure S5. An SEM image of a single silk fibre coated with graphene bonding to an electrode with conductive carbon paint.

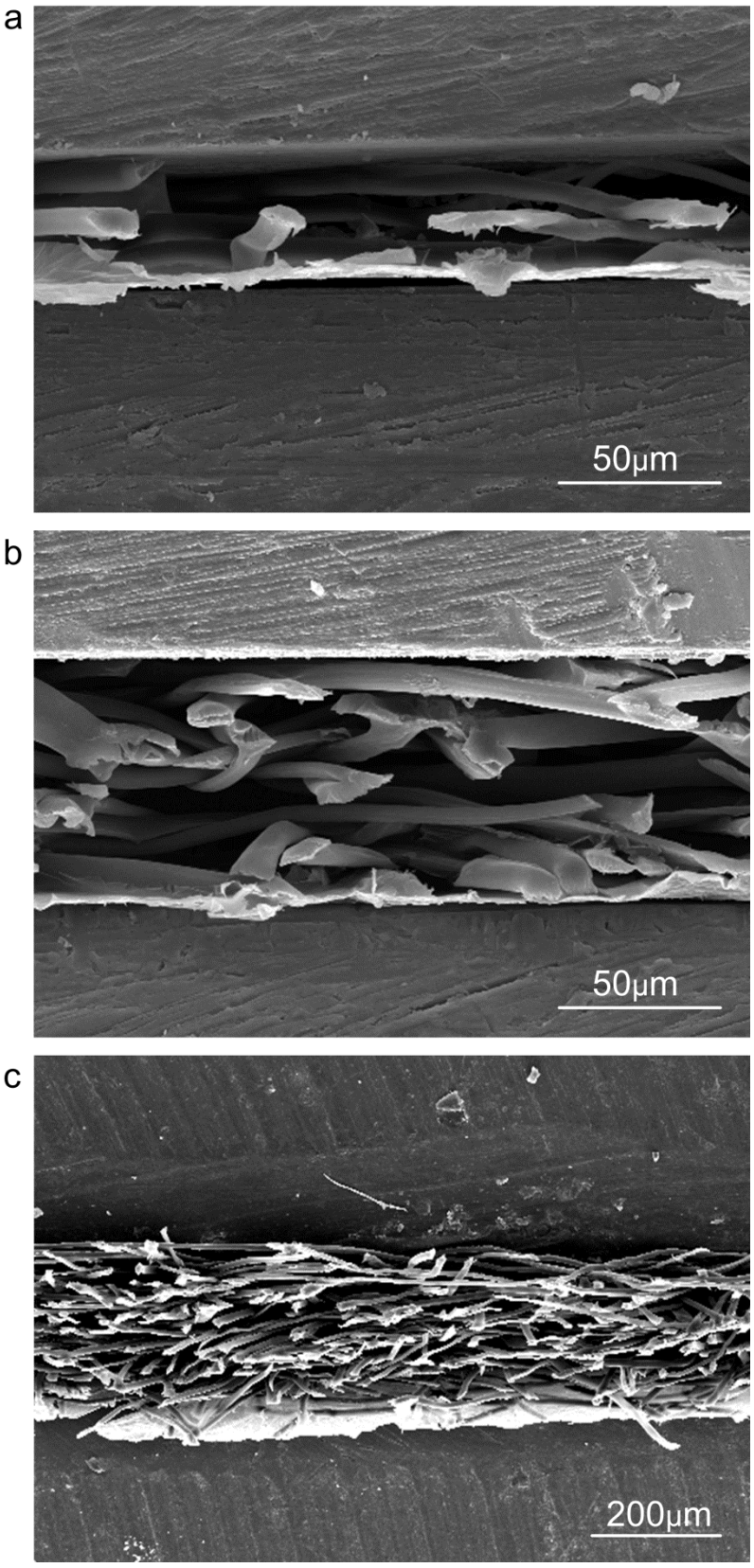

Figure S6. Side-view SEM images of G/S papers prepared with different amount of silk fibres: (a) 20 mg, (b) 50mg and (c) 100mg.

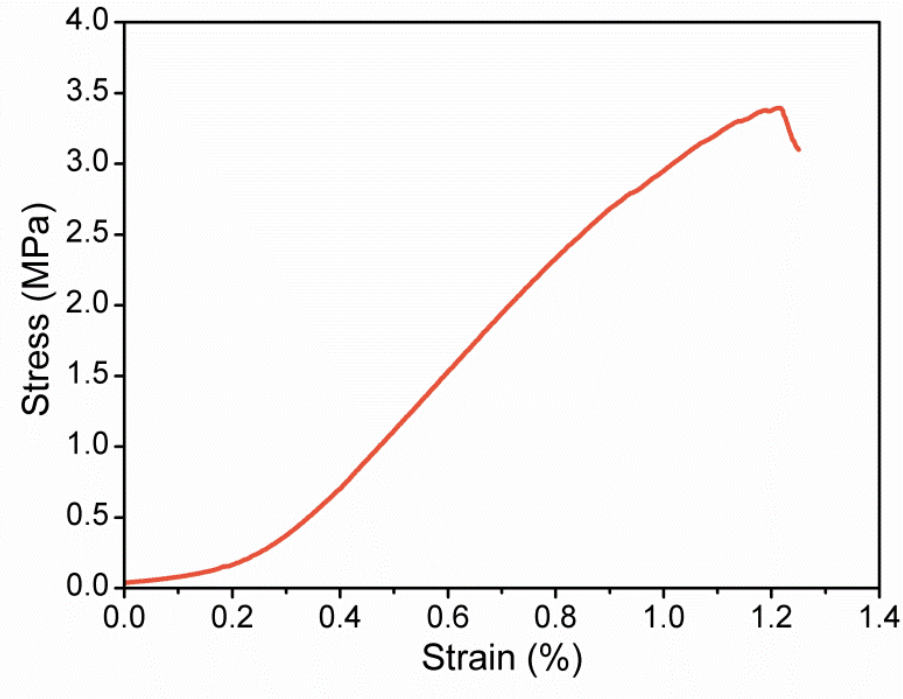

Figure S7. Stress–strain curve of the G/S film.

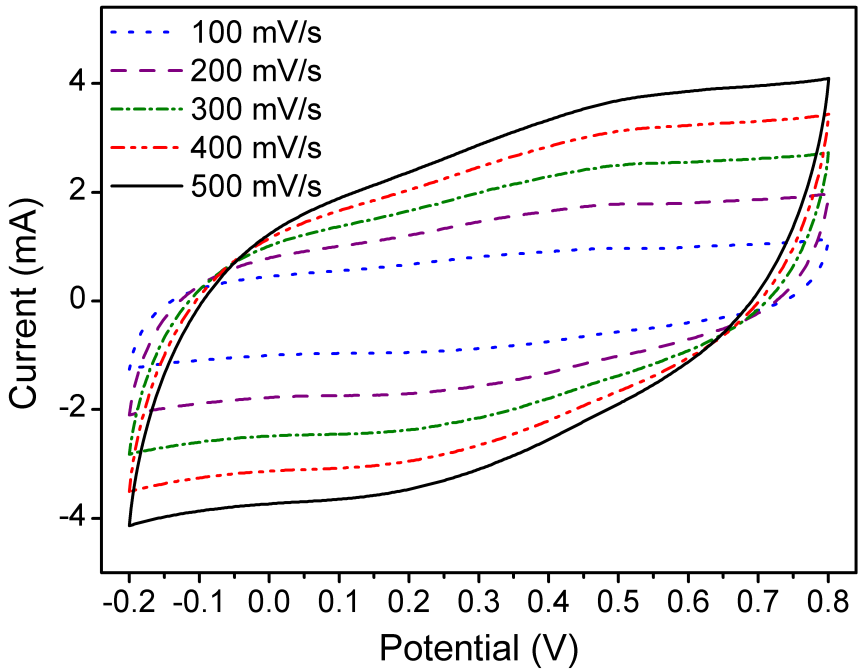

Figure S8. Cyclic voltammetry curves of a G/S film at different scan rates.

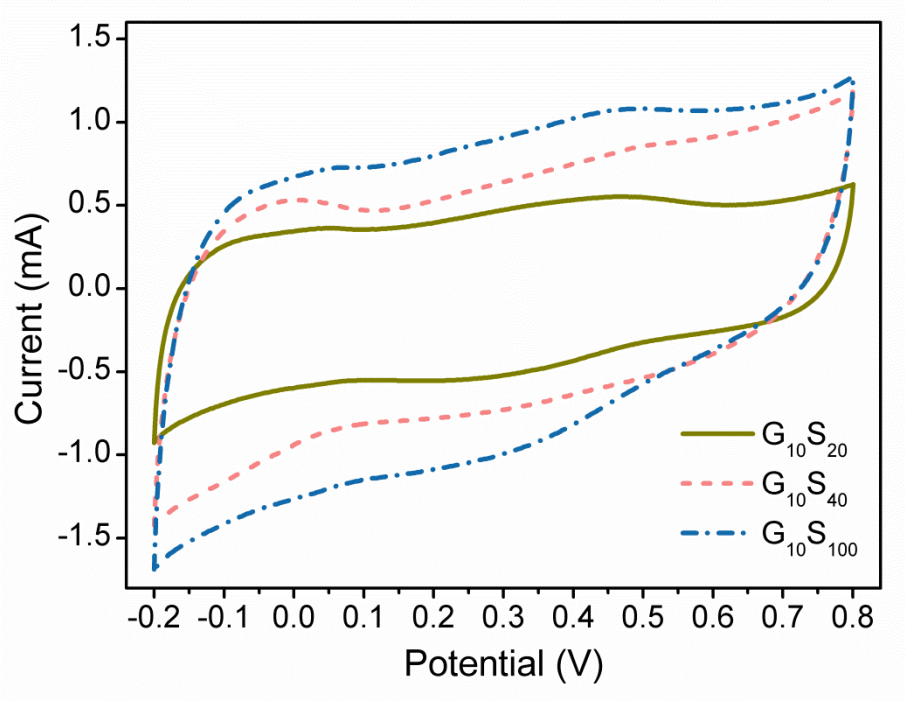

Figure S9. Cyclic voltammetry curves of G/S films prepared with 10 mg GO and 20 mg ($G_{10}S_{20}$), 40 mg ($G_{10}S_{40}$), and 100 mg ($G_{10}S_{100}$) silk fibres respectively.

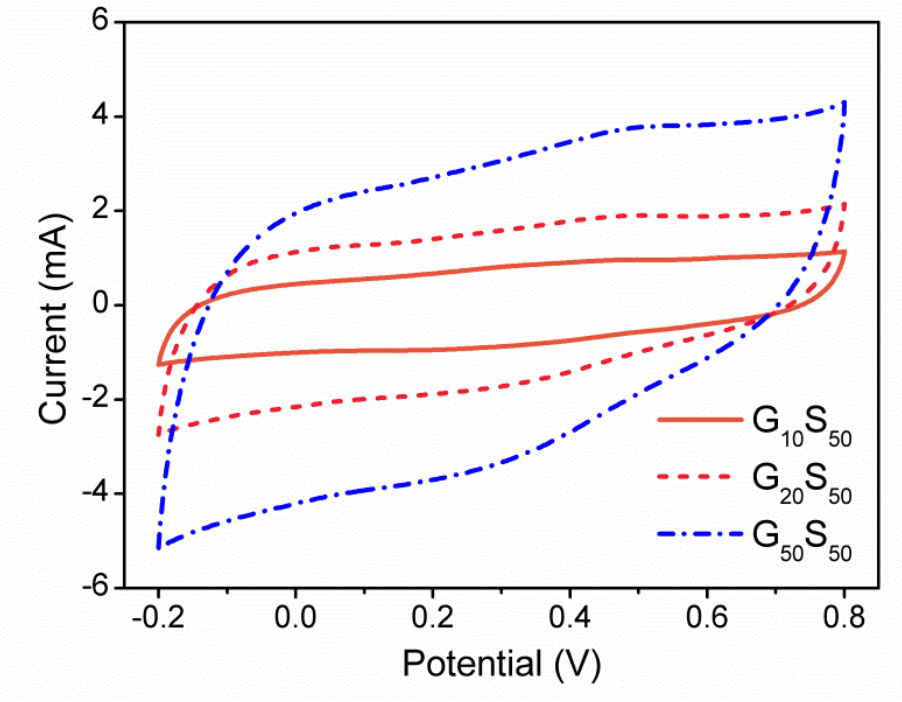

Figure S10. Cyclic voltammetry curves of G/S films prepared with 50 mg silk fibres and 10 mg ($G_{10}S_{50}$), 20 mg ($G_{20}S_{50}$), and 50 mg ($G_{50}S_{50}$) GO respectively.

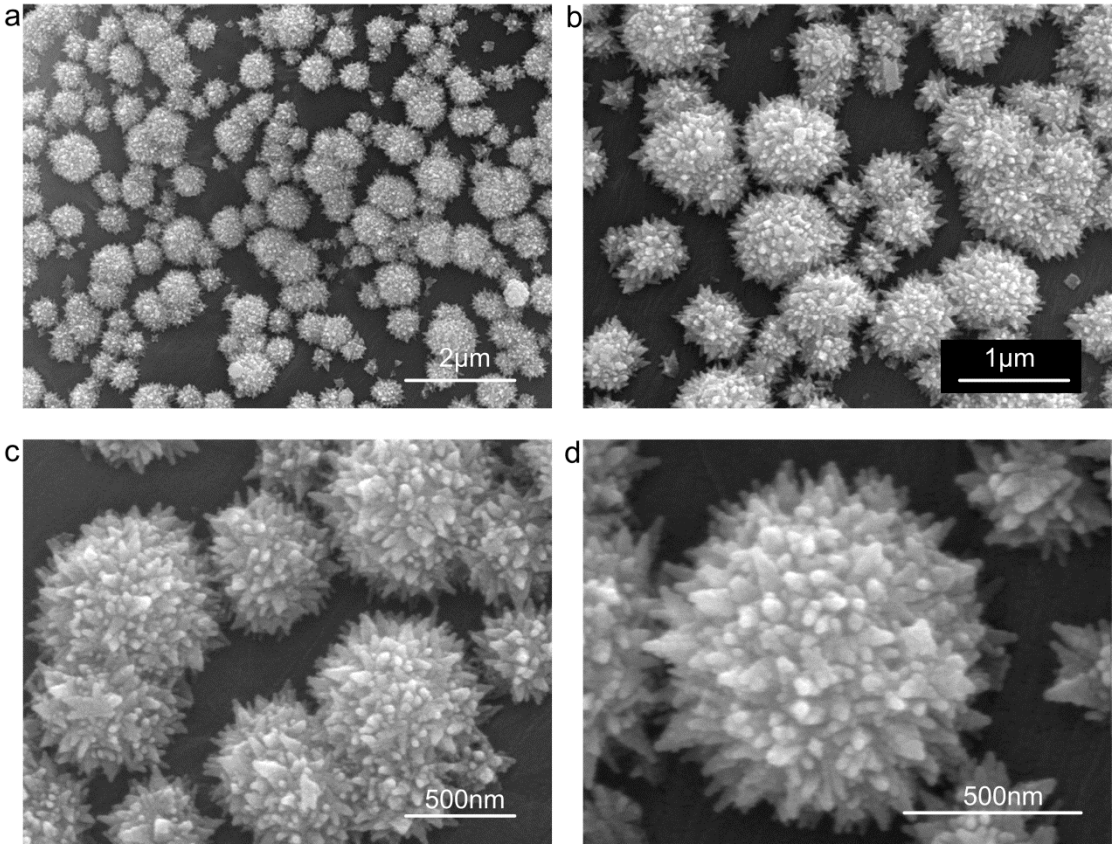

Figure S11. SEM images of spiky Pt nanospheres with different magnifications.

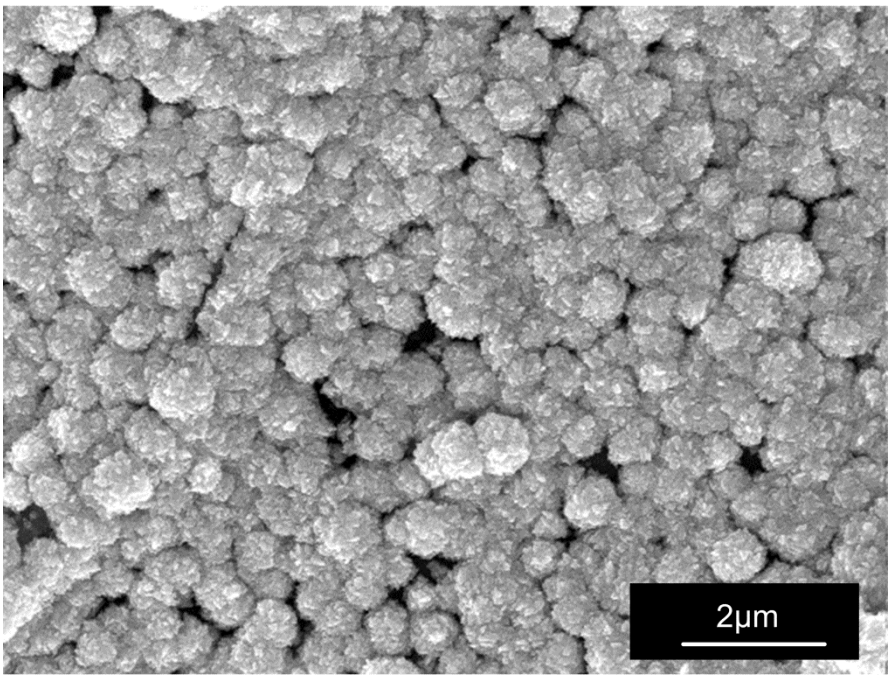

Figure S12. An SEM image of Pt nanoparticles deposited on the surface of a graphene film.

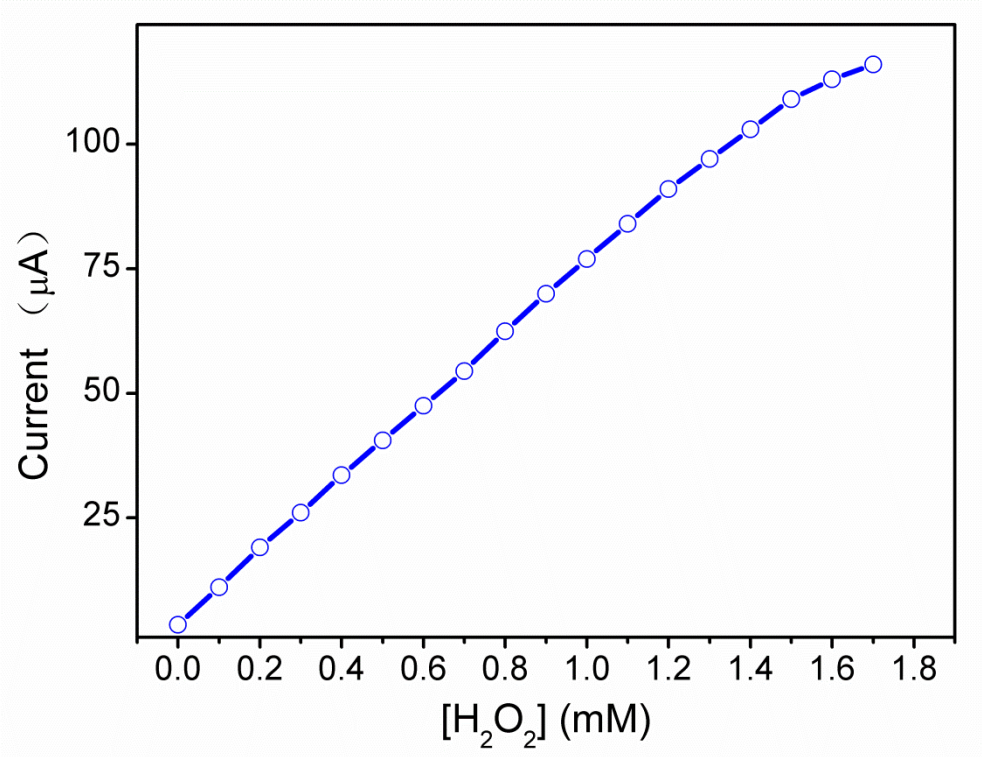

Figure S13. Calibration curve of H_2O_2 detection of the Pt immobilized graphene film at 0.65 V vs. Ag/AgCl.

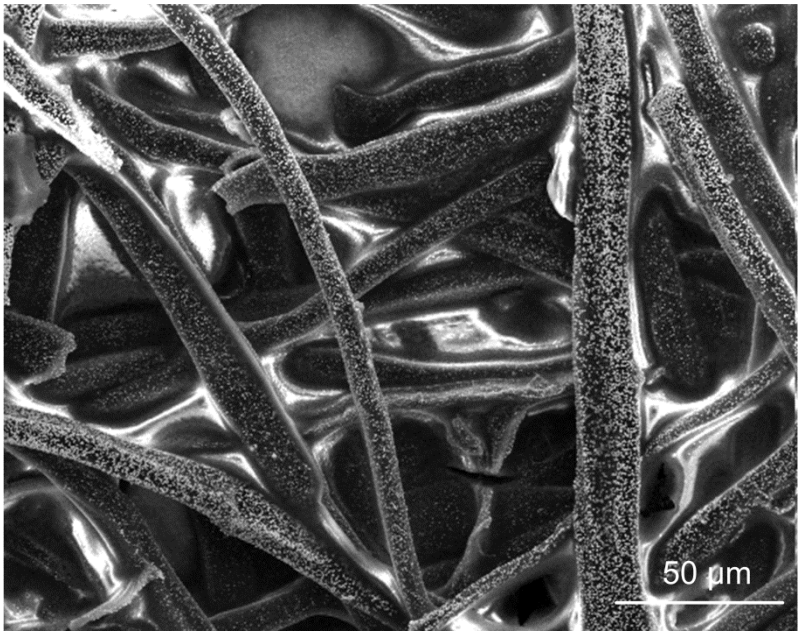

Figure S14. An SEM image of GOx enzyme immobilized G/S film.

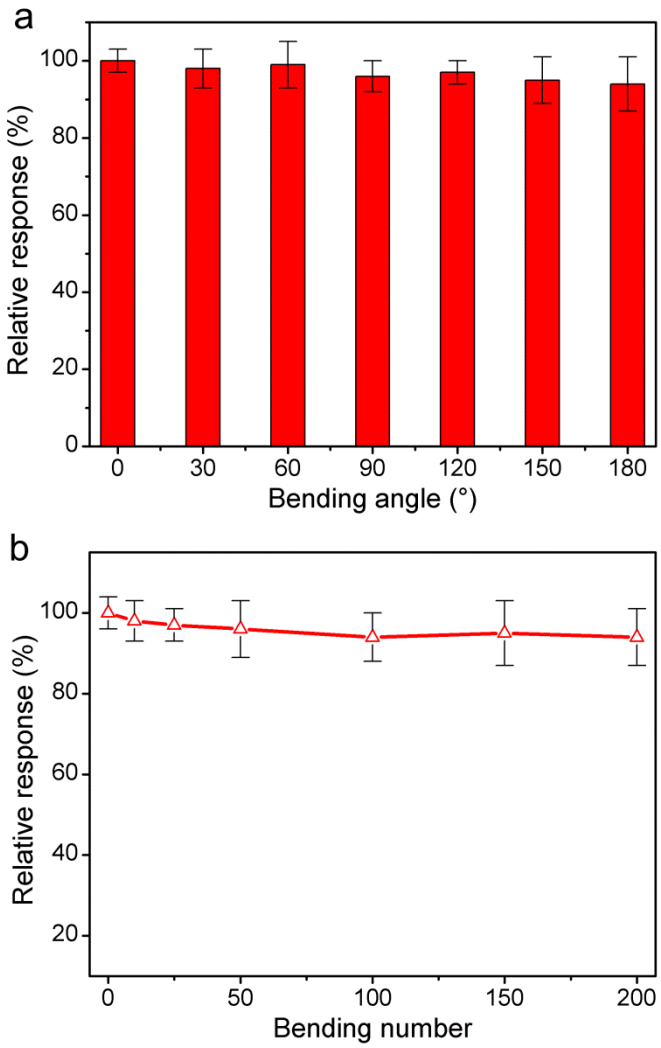

Figure S15. Effects of bending angle (a) and bending number (b) on the current response to 4.0 mM glucose of GOx immobilized G/S film electrodes.

Table S1. Comparison of the G/S film based glucose biosensor and other graphene based glucose sensors.

Working Electrode	Sensitivity ($\mu A\ mM^{-1}cm^{-2}$)	Linearity (mM)	LOD (μM)	References
PtAu-MnO_2/GP[1]	58.54	0.1 - 30	20	[1]
GOx-GR-AuNPs/GCE[2]	3.84	0.02 - 2.26	4.1	[2]
GOx-graphene-chitosan/GCE	37.93	0.08 - 12	20	[3]
GOx/GrOx/Pt-black[3]	78.0	0.001 - 2	1	[4]
GOx/PEI/PAA-graphene/PEI/GCE[4]	0.261	1 - 10	168	[5]
GOx/Pt/GS film	150.8	0.01 - 10	1	This work

[1] PtAu-MnO_2: PtAu alloy and MnO_2 integrated nanocomposites; GP: graphene paper.
[2] GR: graphene; AuNPs: gold nanoparticles; GCE: Glassy carbon electrode.
[3] GrOx: graphene oxide.
[4] PAA: poly(acrylic acid); PEI: poly(ethyleneimine).

References

[1] Xiao F, Li Y, Gao H, Ge S, Duan H. Growth of coral-like PtAu–MnO2 binary nanocomposites on free-standing graphene paper for flexible nonenzymatic glucose sensors. Biosens Bioelectron. 2013;41:417-23.

[2] Cao X, Ye Y, Li Y, Xu X, Yu J, Liu S. Self-assembled glucose oxidase/graphene/gold ternary nanocomposites for direct electrochemistry and electrocatalysis. J Electroanal Chem. 2013;697:10-4.

[3] Kang X, Wang J, Wu H, Aksay IA, Liu J, Lin Y. Glucose oxidase–graphene–chitosan modified electrode for direct electrochemistry and glucose sensing. Biosens Bioelectron. 2009;25:901-5.

[4] Shi J, Zhang H, Snyder A, Wang M-x, Xie J, Marshall Porterfield D, et al. An aqueous media based approach for the preparation of a biosensor platform composed of graphene oxide and Pt-black. Biosens Bioelectron. 2012;38:314-20.

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