Supporting Information

Dual-potential Electrochemiluminescence from Black Phosphorus and Graphitic Carbon Nitrides for Label-Free Enzymatic Biosensing

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1. Experimental Section

1.1 Apparatus

For characterization of nanomaterials, transmission electron microscopy (TEM) was obtained on a JEOL-2100 transmission electron microscopy (JEOL, Japan). FL and UV-vis absorption spectra were recorded at Shimadzu UV-3600 spectrophotometer and RF-5301PC FL spectrophotometer, respectively. The reference electrode (SCE), the counter electrode (platinum wire), and the working electrode (the modified glassy carbon electrode (GCE, Φ = 3 mm)) made up the classical three-electrode system that was applied in the detection. The ECL monitoring and electrochemical characterization were performed on MPI-B ECL analyzer (Xi'an Remax Electronic science & Technology Co. Ltd., China) and CHI 760D electrochemical workstation (Shanghai Chenhua Instrument, China), respectively.

1.2 Preparation of BPNSs

The preparation of black phosphorus nanosheets is in accordance with the previous literature (Ding et al., 2019).[1] 5 mg of BP powder was added into 1 mL of double-distilled water in a mortar and then ground for 20 min. The mixture was transferred to a glass vial containing 3 mL of double-distilled water. After being sealed carefully, the vial was sonicated in an ice-bath for 8 h at a power of 100 W. The resulting dispersion was centrifuged at 11000 rpm for 20 minutes. The top 75% of the solution was collected as the sample of BPNSs.

1.3 Preparation of g-C_3N_4 nanosheet

The bulk g-C_3N_4 and the g-C_3N_4 nanosheets were prepared by direct pyrolysis of the mixture of thiourea and urea in a tube furnace according to previous work with some necessary revision (Niu et al., 2012).[2] In a typical process, 3 g thiourea and 3 g urea were mixed together in 20 mL deionized water, and ultrasonic treated for 5 min. Then, the mixture was put into the muffle furnace and calcined at 550 ℃ for 2 h with a heating rate of 5 ℃ min^{-1} for both of the heating and the cooling processes. The obtained yellow powder was the bulk g-C_3N_4, which was ground to powder for further use. In the next process, the bulk g-C_3N_4 powder was placed in an open ceramic container and heated at 500 ℃ for 2 h with a heating rate of 5 ℃ min^{-1} for both of the heating and the cooling processes. The light yellow powder of g-C_3N_4 nanosheets were finally obtained.

2. Results and discussions

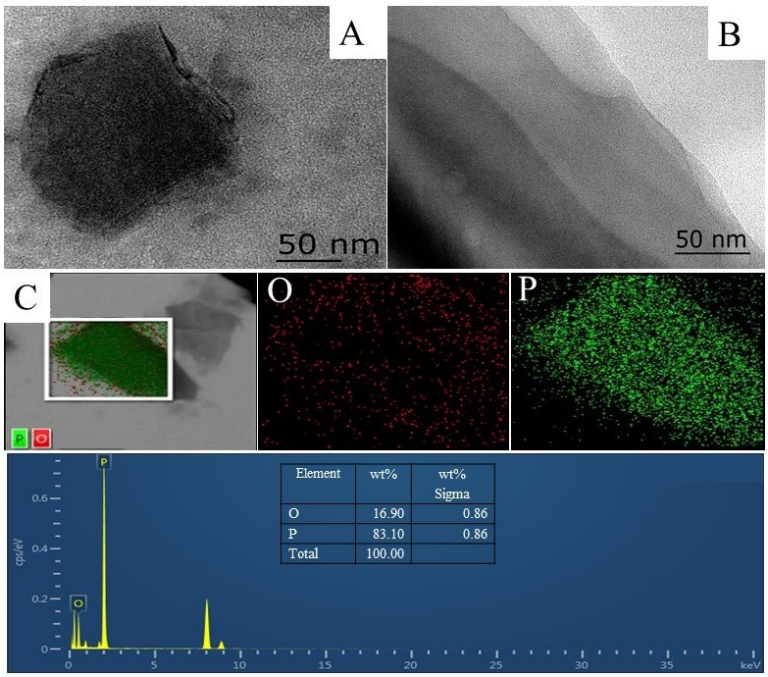

Fig.S1 (A, B) TEM images of BPNSs. (C) EDS and corresponding mapping analyses of BPNSs

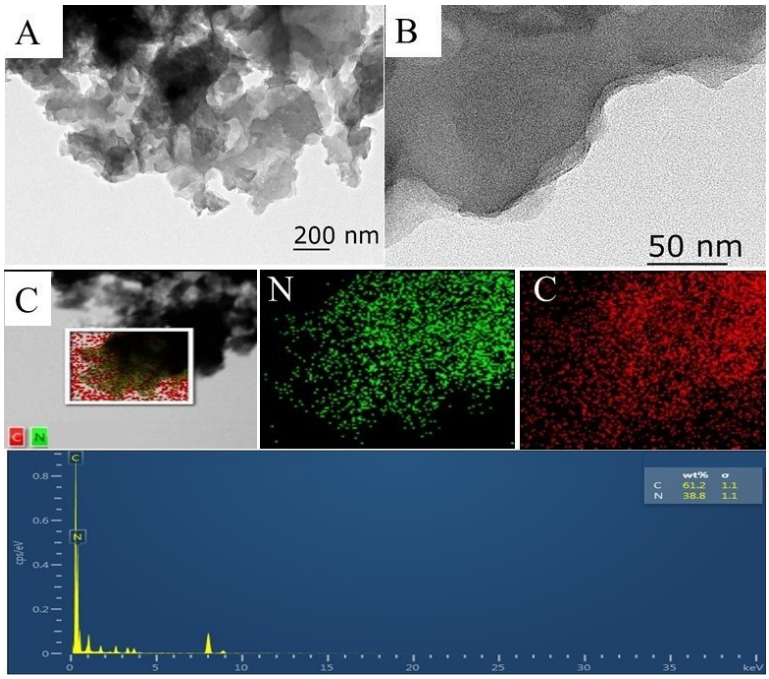

Fig.S2 (A, B) TEM images of g-C_3N_4. (C) EDS and corresponding mapping analyses of g-C_3N_4.

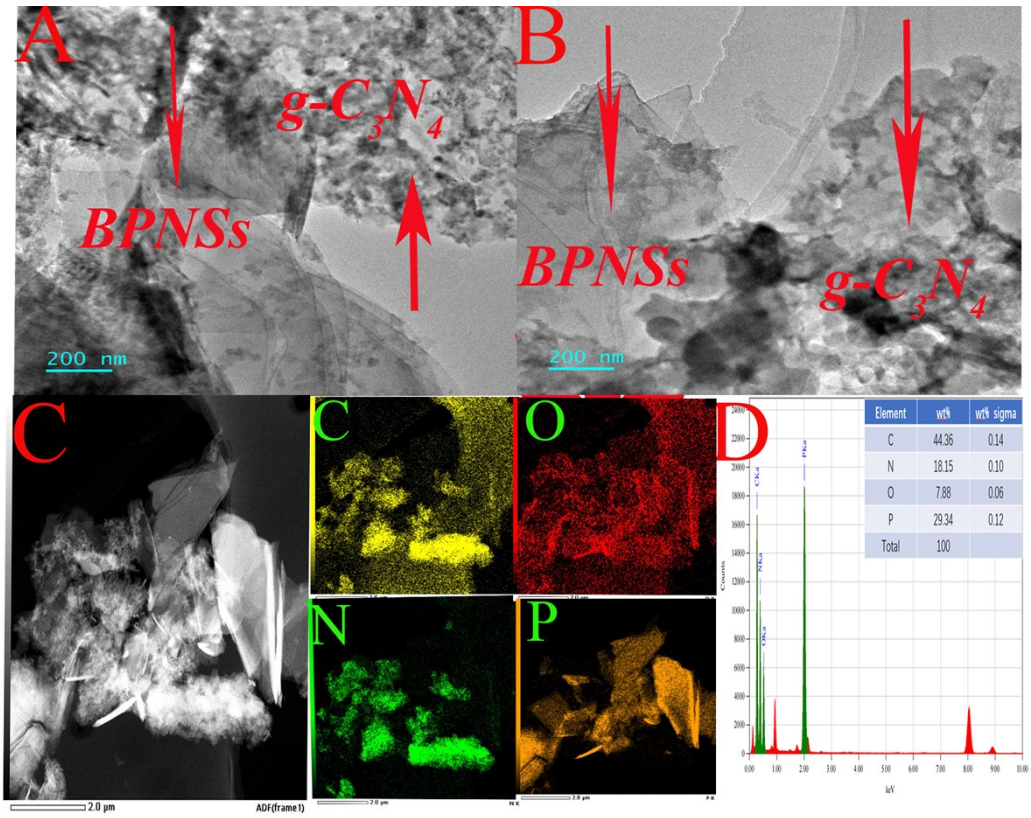

Fig.S3(A, B) TEM images of g-C_3N_4/BPNSs. (C, D) EDS and corresponding mapping analyses of g-C_3N_4/BPNS.

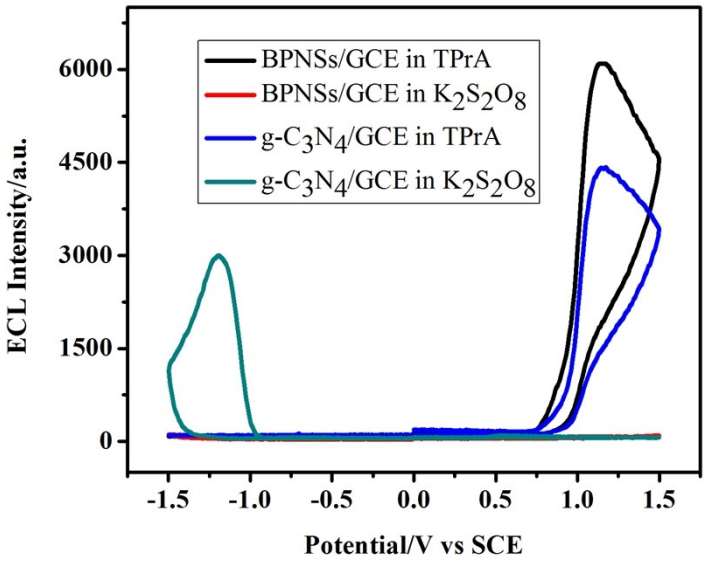

Fig.S4 ECL curves of a BPNSs/GCE and a g-C_3N_4/GCE in PBS with 0.1 mM $K_2S_2O_8$ or 1 mM TPrA; scan rate, 100 mV s^{-1}.

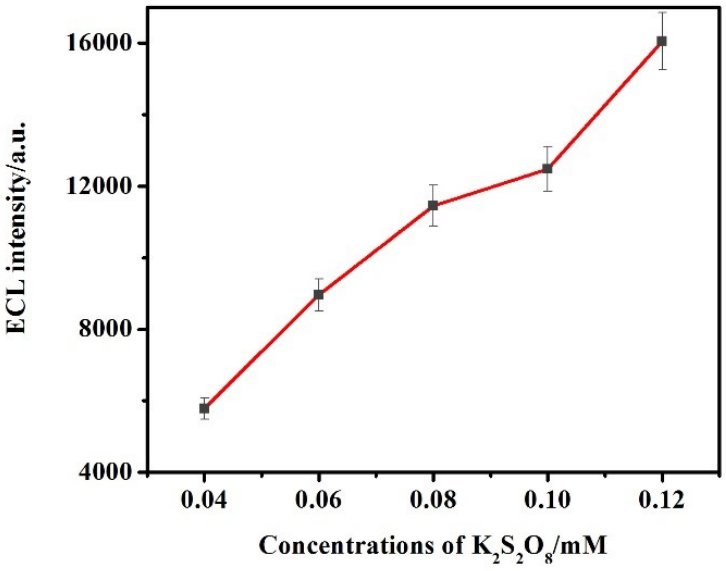

Fig.S5 Effect of the concentration of $K_2S_2O_8$ on cathodic-ECL intensity.

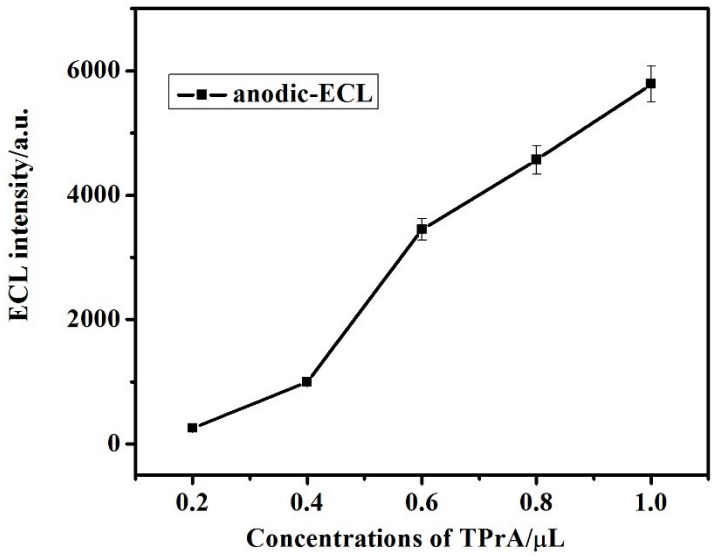

Fig.S6 Effect of the concentration of TPrA on cathodic-ECL intensity.

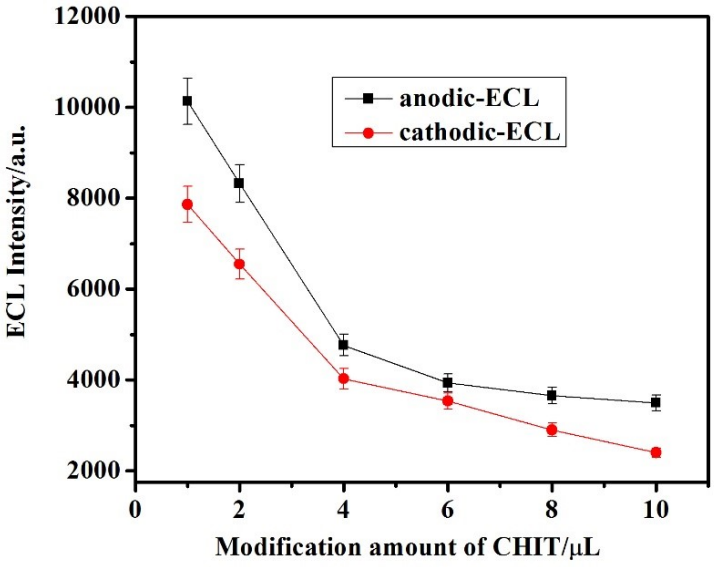

Fig.S7 Effect of the modification amount of CS on ECL intensity.

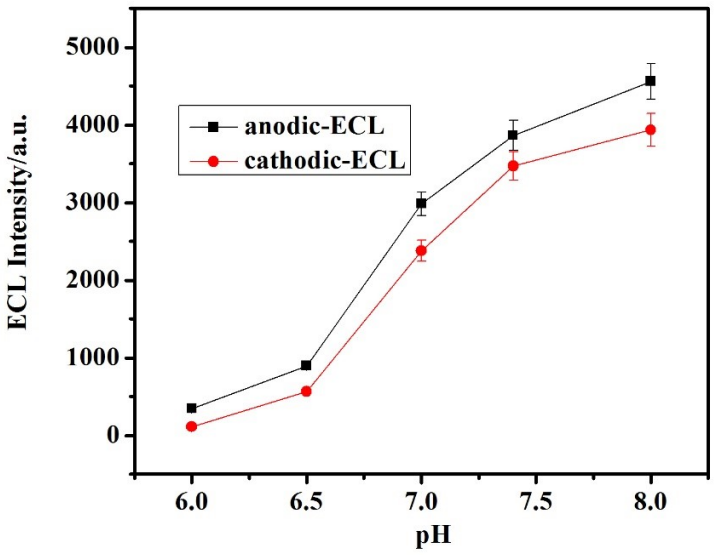

Fig.S8 Effect of the pH value on ECL intensity.

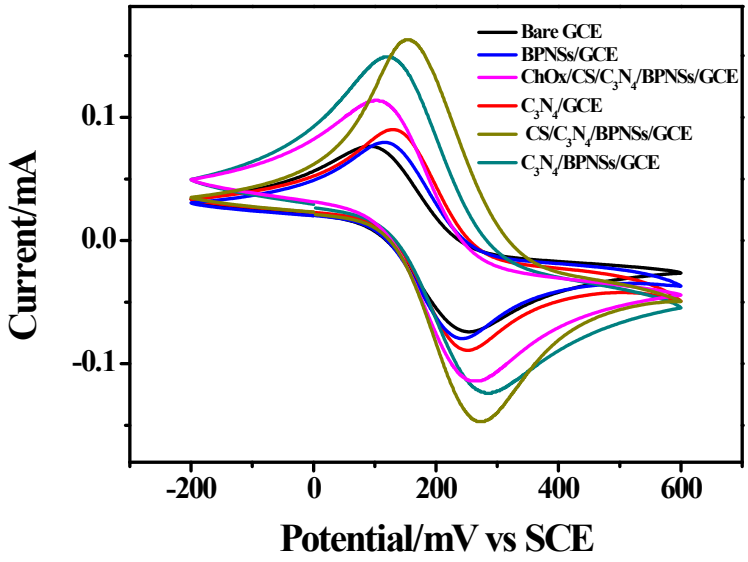

Fig.S9 CV curves of different stages of modified electrodes in 1 mM $K_3Fe(CN)_6$

Table S1. Comparison of different methods for determination of cholesterol.

Methods	Linear range/ mM	Detection limit/μM	Reference
colorimetry	0.1-1.5	40	3
DPV	0.01-5.0	4.3	4
Amperometry	0.0022-0.52	0.75	5
DPV	0.0001-9.331	0.021	6
ECL	0.0005 - 0.5	0.14	This paper

References

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