

Supplementary Information

Bimetallic Pd-Pt Supported Graphene Promoted Enzymatic Redox Cycling for Ultrasensitive Electrochemical Quantification of MicroRNA from Cell Lysates

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Table S1. Sequences of Oligonucleotides Used in This Work

Name	Sequences(5'→3')
let-7b	UGA GGU AGU AGG UUG UGU GGU U
DNA1	HS-CGAAATTGT CACAACCACACAAC-OH
DNA2	Phos-CTACT ACC TCACACAGTA AAAGC-Biotin
let-7a	UGAGGUAGUAGGUUGUAUAGUU
let-7c	UGAGGUAGUAGGUUGUAUGGUU
let-7d	AGAGGUAGUAGGUUGCAUAGUU
miR-21	UAG CUU AUC AGACUG AUGUUGA

The Italic red letter refers to the mismatched base.

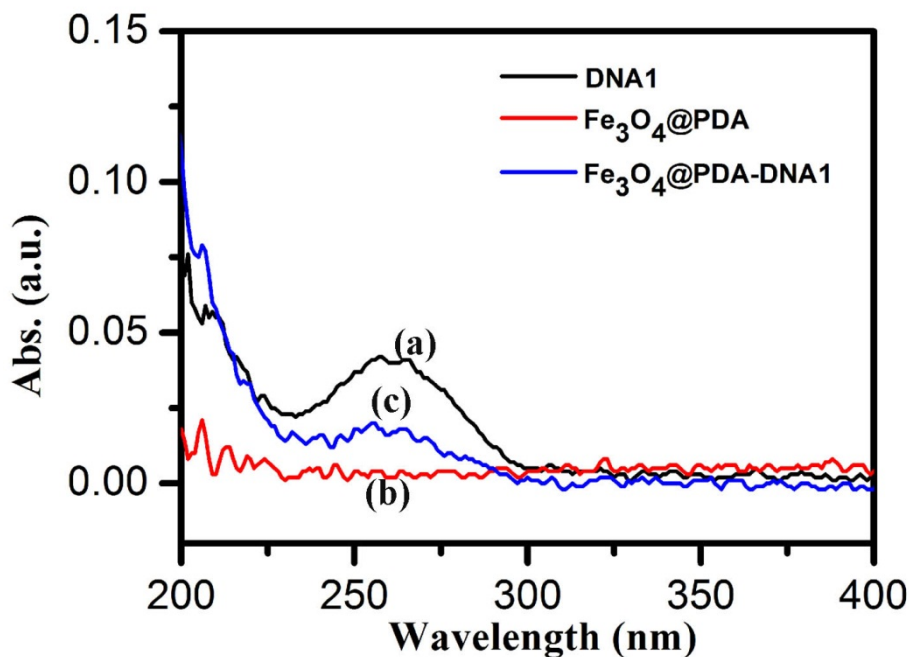


Figure S1. UV-visible spectra of DNA1 (a), $\text{Fe}_3\text{O}_4@PDA$ (b) and $\text{Fe}_3\text{O}_4\text{-DNA1}$ (c)

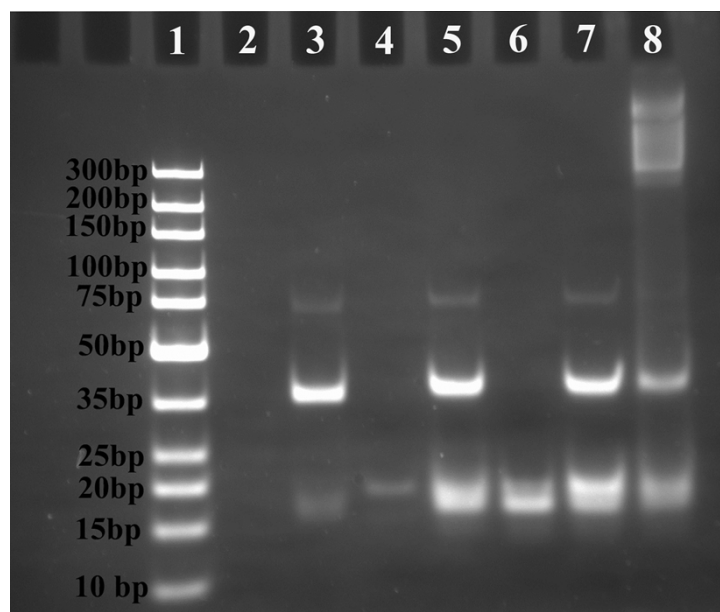


Figure S2 Polyacrylamide gel electrophoresis analysis of RNA hybridization process.

Lane 1: marker; lanes 2: let-7b; lane 3: DNA1; lane 4: DNA2; lane 5: DNA1+let-7b; lane 6: DNA2+let-7b; lane 7: DNA1+DNA2+let-7b; lane 8: DNA1+DNA2+let-7b+T4 DNA ligand.

As shown in Fig.S2, no band appeared in Lane 2 because the sequence of single-stranded let-7b is short and difficult to be dyed by EB. Two bands in Lane 3 assigned to DNA1 and disulphide DNA1, one band in Lane 4 assigned to DNA2. In the presence of let-7b (Lane 5), the emission bands between 15bp and 20bp is brighter than that of Lane 3, indicating that let-

7b is hybridized with DNA1 and forms DNA1/let-7b complex, the same as DNA2 in Lane 6. The band in Lane7 is no significant difference comparing to Lane 5. But in fact, the band at 20 bp is brightest. Moreover, in the presence of T4 DNA ligase (Lane 8), the band of DNA1/let-7b/DNA2 complex is shifted to the location of >300bp, and indicated the formation of DNA1/let-7b/DNA2 complex.

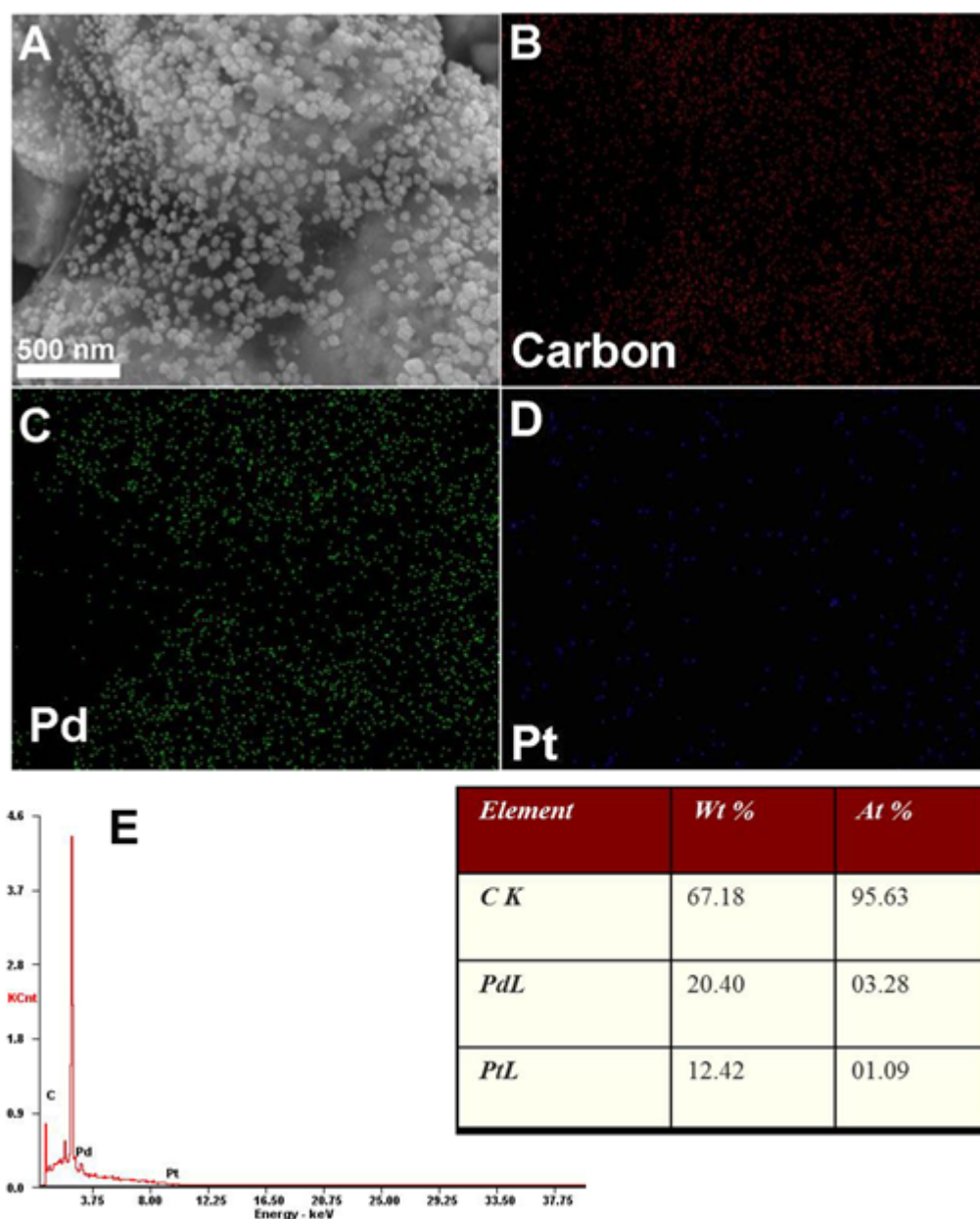


Figure S3. SEM images and EDX of Pt/Pd/RGO/SPGE electrode

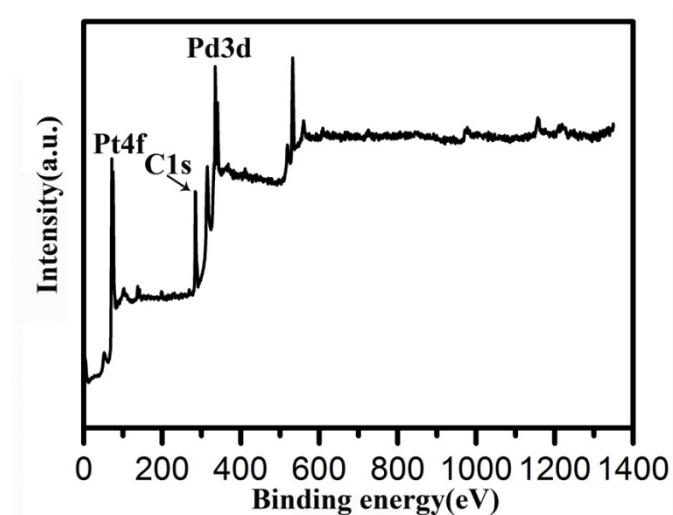


Figure S4. X-ray photoelectron spectroscopy (XPS) of Pt/Pd/RGO/SPGE electrode

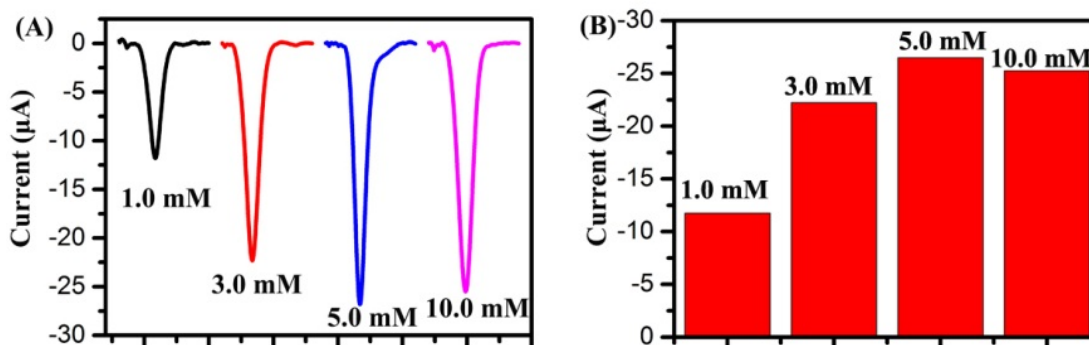


Figure S5A) DPV responses for p-NP after p-NPP solution of different concentrations was incubated with 10 μL, 0.01 mg mL⁻¹ SA-ALP for 15 min. B) The peak current value in Fig.S5A corresponding to the concentration of p-NPP

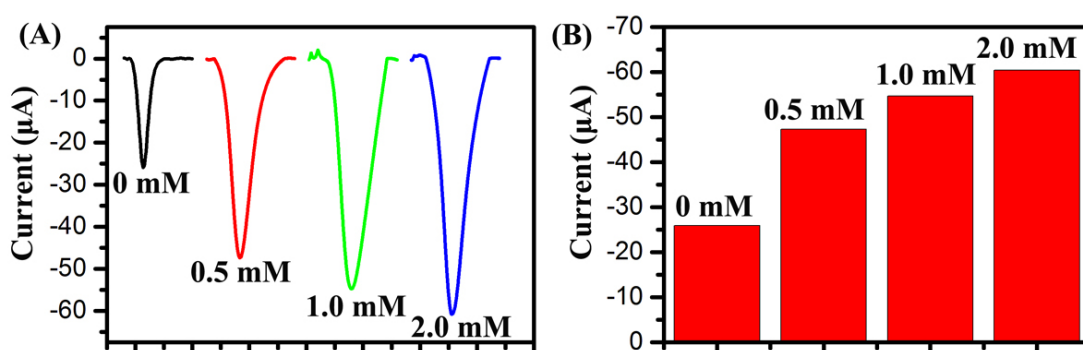


Figure S6. A) The DPV measurements for p-NP after p-NPP solution (5.0 mM) was incubated with 10 μL, 0.01 mg mL⁻¹ SA-ALP for 15 min in the presence of TCEP at a concentration from 0 mM to 2.0 mM using Pt/Pd/RGO/SPGE electrode. B) The peak current value in Fig.S6A corresponding to the concentration of TCEP

Optimization of assay conditions

This biosensor is based on using ALP to convert 1-naphthyl phosphate disodium salt (p-NPP) into electroactive 1-naphthol (p-NP), and tris(2-carboxyethyl) phosphine (TCEP) to regenerate p-NP for realizing a redox-cycling reaction. Therefore, the effects of substrate and TCEP concentration were investigated (Fig. S5 and S6). As shown in Fig. S5, with the substrate concentration increasing, the peak values tended to a steady value at the concentration of 5.0mM. In Fig.S6, with the TCEP concentration increased, the peak values tended to own a minor change at the concentration of 1.0 mM. Thus p-NPP of 5.0mM and TCEP of 1.0 mM were elected as optimal concentration for the following detection of let-7b.