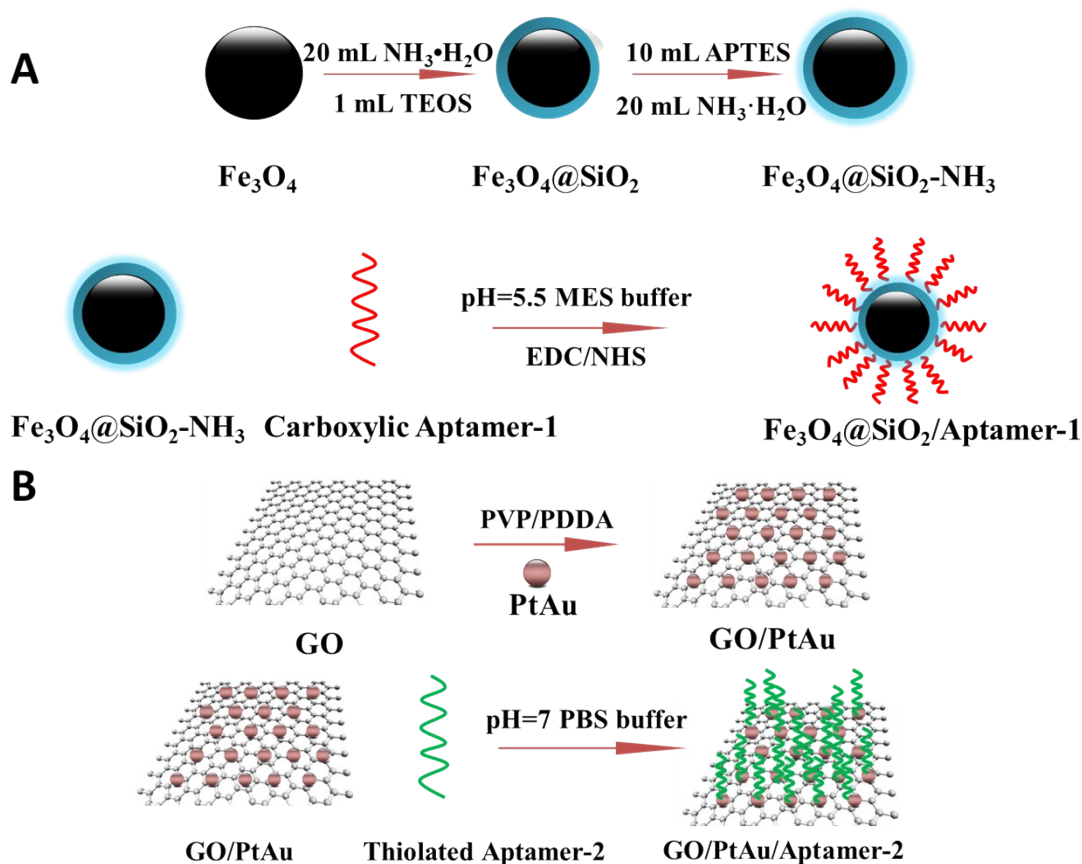


1 ethanol (80 mL) and ultrapure water (20 mL) under ultrasonication for 5 min.
2 Ammonia (1 mL) and TEOS (200 μ L) was added into the mixture solution under
3 mechanical stirring for 12 h. After being separated, the product mixed with APTES
4 (100 μ L) and ammonia (2 mL) under mechanical stirring for another 12 h. Finally, the
5 $\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-NH}_3$ was magnetic separated three times and re-dispersed in 5 mL
6 ethanol/ H_2O solution. The diameter of the $\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-NH}_3$ was about 200 nm
7 evaluated by TEM measurements.

8 The aptamer-1 functionalized $\text{Fe}_3\text{O}_4@\text{SiO}_2$ was prepared as follows.
9 $\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-NH}_3$ in 100 μ L ethanol/ H_2O solution was first mixed with 20 μ L Tween
10 20 (5% in ethanol solution) at room temperature. Then carboxyl modified aptamer-1
11 (final concentration of oligonucleotides 10 μ M) in 400 μ L 10 mM MES buffer with
12 200 mM EDC and 50 mM NHS was added into the $\text{Fe}_3\text{O}_4@\text{SiO}_2$ suspension. The
13 mixture solution was mechanical stirred at 37 $^\circ\text{C}$ for 16 hours. To remove excess
14 aptamer, these aptamer-functionalized $\text{Fe}_3\text{O}_4@\text{SiO}_2$ were magnetic separated and
15 washed three times using 10 mM Tris-HCl buffer containing 5 mM MgCl_2 and 15
16 mM KCl (pH=8.0). The functionalized $\text{Fe}_3\text{O}_4@\text{SiO}_2$ nanoparticles were stored in the
17 buffer solution.



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2 **Scheme S1.** (A) Schematic illustration of the preparation of aptamer-1 modified
 3 magnetic beads. (B) Schematic illustration of the preparation of aptamer-2 modified
 4 graphene oxide/platinum/gold nanoparticle hybrids

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6 2.5 Preparation of PDDA-Functionalized Graphene Oxide.

7 The graphene oxide was prepared from graphite powder according to the
 8 previous work². Then 80 mg of PVP was upon addition of 20 mL of 0.25 mg/mL
 9 obtained graphene oxide solution. After magnetic stirring 30 min, the dispersion was
 10 centrifuged separation three times and re-dispersed in 5 mL ultrapure water. The
 11 PDDA-functionalized graphene oxide was prepared by mixing 0.1 mL of 20 wt %

1 PDDA with 16.8 mL of 0.625 M KCl, followed by adding 4.2 mL of PVP-coated
2 graphene oxide. After sonication for 90 min, the PDDA/GO dispersion solution was
3 centrifuged separation three times and re-dispersed in 5 mL ultrapure water.

4 *2.6 Preparation of Aptamer-2 Modified Graphene Oxide/Platinum/Gold* 5 *Nanoparticle Hybrids.*

6 The Platinum/gold nanoparticle hybrids were synthesized with citrate reduction
7 according to previous work ³. A 100 mL sample of aqueous (0.9 mM) HAuCl₄ and
8 K₂PtCl₆ (0.1 mM) was prepared in a 250 mL round-bottom flask. The solution was
9 continuously heated to a boil while stirring, and 40 mM trisodium citrate solution (10
10 mL) was added. The boiling and stirring was continued for an additional 30 min after
11 the final color changing from pale yellow to deep red.

12 1 mL PDDA/GO dispersion solution (1 mg/mL) was added into 10 mL of
13 prepared PtAu_{NPs} under magnetic stirring. Then the dispersion solution was sonicated
14 for 10 min before overnight aging. The obtained product was washed three times and
15 dispersed in 2 mL of water.

16 The aptamer-2 functionalized GO/PtAu_{NPs} was prepared by mixing GO/PtAu_{NPs}
17 (2 mL) with thiol-aptamer (final concentration of oligonucleotides 10 μM) in PBS
18 buffer (0.3 M NaCl, 0.2 PBS, pH=7) under continuous shaking for 16 hours⁴. To
19 remove excess thiol-aptamer, these aptamer-2 functionalized GO / PtAu_{NPs} were
20 centrifuging for 45 min at 13000 rpm three times using 10 mM Tris-HCl buffer
21 containing 5 mM MgCl₂ and 15 mM KCl (pH=8.0). The functionalized GO / PtAu_{NPs}

1 nanoparticles were stored in the buffer solution. The synthesized nanomaterials were

2 shortly named as GO/PtAu_{NPs}/Aptamer-2.

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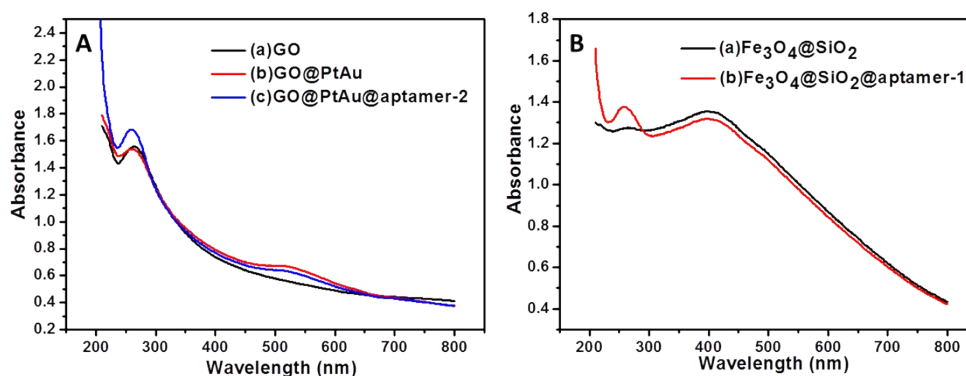
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2 **Fig. S1** (A) UV-Vis absorption spectra of (a) GO (10 $\mu\text{g/mL}$), (b) GO@PtAu (10
 3 $\mu\text{g/mL}$), and (c) GO@PtAu@aptamer-2 solutions (10 $\mu\text{g/mL}$). (B) UV-Vis absorption
 4 spectra of (a) Fe₃O₄@SiO₂ (0.06 mg/mL) and (b) Fe₃O₄@SiO₂@aptamer-1 solutions
 5 (0.06 mg/mL).

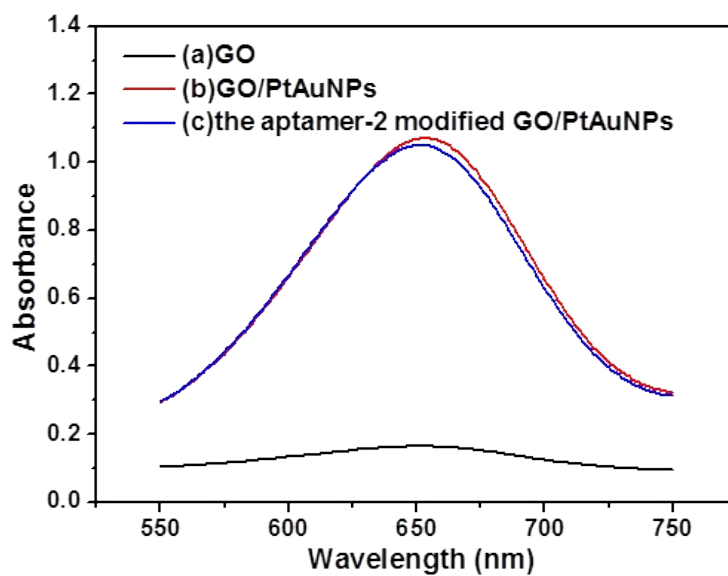
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7 As shown in Figure S1 (supporting information), the functionalized
 8 GO/PtAu_{NPs} and magnetic bead were characterized by measuring the UV absorbance
 9 spectra after removing excess DNA. Except for the feature peaks of GO at 270 nm
 10 and PtAu at 536 nm, a new and higher peak at 260 nm was detected for the
 11 GO/PtAu_{NPs}/aptamer-2 (curve c in Figure S1A), indicating the successful anchoring
 12 of aptamer-2 on the surface of GO/PtAu_{NPs}. A similar peak at 260 nm was observed
 13 for aptamer-1-modified magnetic beads (curve b in Figure S1B).

14 The absorbance value of the functionalized nanoparticles was used to estimate
 15 the amount of aptamers after deducting the absorbance value of naked nanoparticles at
 16 260 nm. According to oligonucleotide synthesis reports, the reciprocal of molar
 17 extinction coefficients for aptamer-1 and aptamer-2 are 7.0 and 3.9 nmol/OD,
 18 respectively. The amount of aptamer-1 on the magnetic beads was 10.94 nmol/mg

1 while the amount of aptamer-2 on the GO/PtAu_{NPs} was 57 nmol/mg according to
2 simple calculations.

3



4

5 **Fig. S2.** Peroxidase-like activity of (a)GO, (b) naked GO@PtAu and (c) the aptamer-2
6 modified GO/PtAuNPs on the colorimetric reaction of TMB in the presence of H₂O₂.

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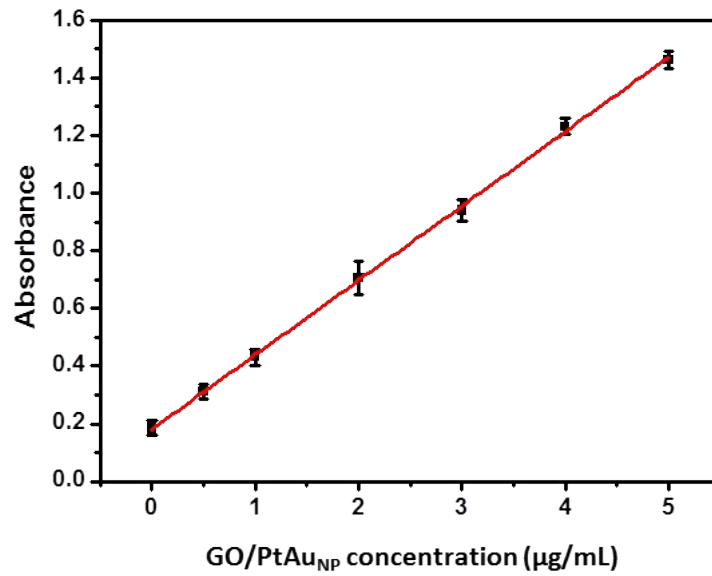
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2 **Fig. S3.** Linear relationship between absorbance value of the oxidized TMB product
3 at 652 nm and the concentration of GO/PtAu_{NPs}. The error bars represent the standard
4 deviation of three independent measurements.

5

6 Figure S3 provides the linear relationship between absorbance value of the
7 oxidized TMB product at 652 nm and the concentration of GO/PtAu_{NPs}. This research
8 shows that with the increasing concentration of the superior catalyst, the catalytic rate
9 of TMB is enhanced relatively.

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