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Supporting Information

Photonic crystal barcodes assembled by dendritic silica nanoparticles for the multiplex immunoassays of ovarian cancer

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Reagents and chemicals

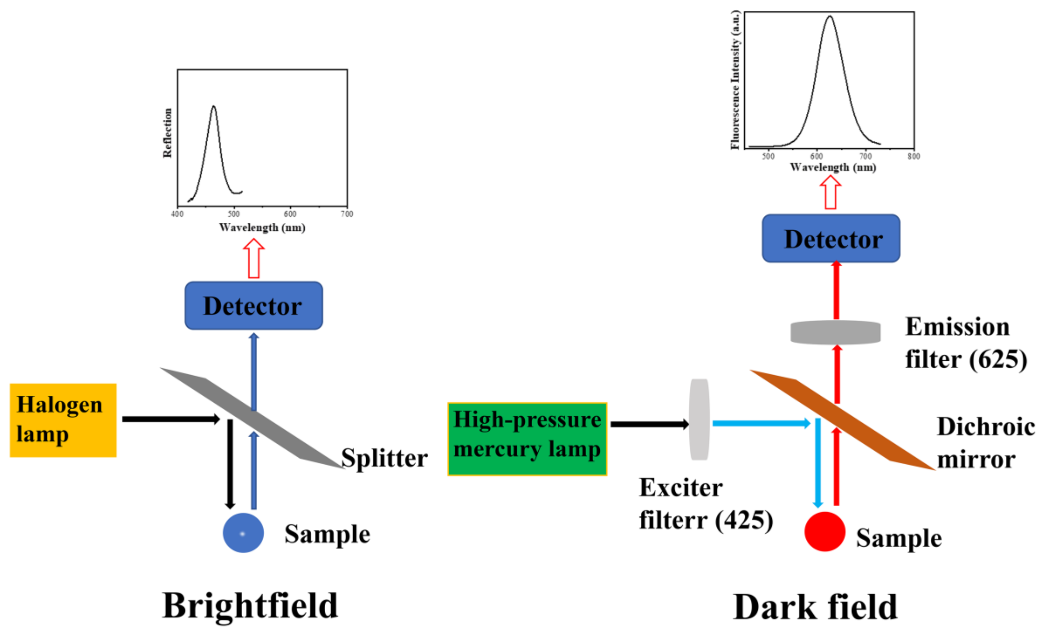
3-mercaptopropionic acid (MPA, ≥98%) was obtained from Aladdin Technology Co., Ltd. Tellurium (Te, 99.9%), sodium borohydride ($NaBH_4$, ≥96%), cadmium chloride($CdCl_2$•2.5H_2O, 99.0%), sodium hydroxide (NaOH, ≥96%), sodium dihydrogen phosphate dihydrate (NaH_2PO_4•2H_2O,≥99%) and disodium hydrogen phosphate dodecahydrate (Na_2HPO_4•12H_2O, ≥99%) were provided from Sinopharm Chemical Reagent Co., Ltd. Phosphate buffer saline (PBS, 0.1 M, pH 7.4), phosphate buffer saline tween-20 (PBST, 0.05% tween-20 in the PBS) were self-prepared. Double distilled water was used throughout the experiment. All reagents were purified without further purification.

Synthesist of CdTe QDs

The carboxylated CdTe quantum dots were prepared according to the literature. [1] Briefly, 67.0 μl of MPA and 91.3 mg of $CdCl_2$•2.5H_2O were dissolved in 40 mL of H_2O. The pH of solution was adjusted to 11.0 using 1.0 M NaOH. Under stirring with N_2, the 1.0 mL of 0.04 M NaHTe solution (produced by the reaction of oxygen-free $NaBH_4$ solution with Te power under N_2 atmosphere) was added to this solution at room temperature. Then the solution was refluxed at 100 ℃ for several hours. The obtained CdTe products were precipitated for three times by ethanol with centrifugation at 10,000 rpm for 10 min.

Bioconjugation of CdTe-labeled Ab_2

The preparation of CdTe QDs conjugated antibodies is not much different from previous reports.[2, 3] First, freshly prepared EDC (150 μL, 4.2 mg/mL) solution was used to activate the carboxyl groups on the surface of CdTe (150 μL, 5 mg/mL) (shaking for 15 minutes). Then, CEA-Ab_2 (500 μL, 26 μg/mL) was added to the above uniformly mixed solution and shaken for 2 hours in a dark environment at room temperature. To remove unconjugated QDs and byproduct, the resulting product was purified by ultrafiltration using a 50 KD filter (3000 g for 10 min). Finally, the non-specific active sites on the purified CdTe-CEA Ab_2 were blocked with 1% BSA in PBS (containing 2% NaCl). The preparation of CdTe QDs coupling CA125-Ab_2 (500 μL, 25 μg/mL) and AFP-Ab_2 (500 μL, 33 μg/mL) is the same as above.



Scheme S1 Schematic of collection of lights from the refection and fluorescence.

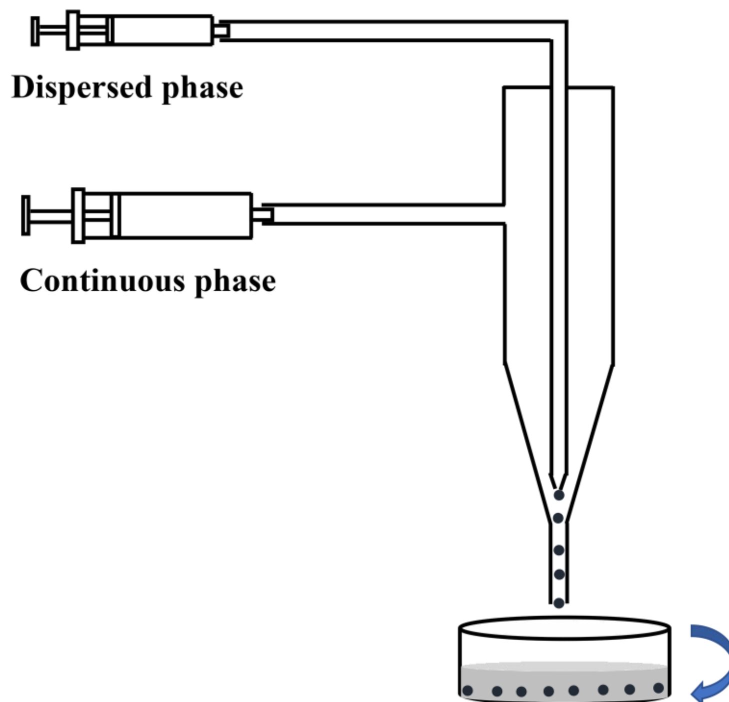


Fig. S1 Illustration of the T-shaped microfluidic device for the fabrication of photonic crystal microspheres. The dispersed phase was injected via a needle positioned along the main axis of the T-junction. The continuous phase was injected through the perpendicular inlet to the main channel of the T-junction.

Table S1 Particle size and pore size data of dendritic silica particles

	$dSiO_2$-1	$dSiO_2$-2	$dSiO_2$-3
Average particle size (nm)	242	274	330
Average pore size (nm)	22.45	17.91	24.38

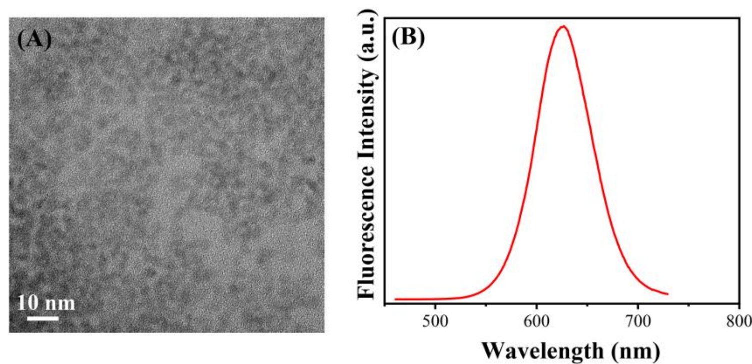


Fig. S2 (A) TEM image of CdTe QDs. (B) Fluorescence emission spectra of CdTe QDs (λ_{ex}=365 nm).

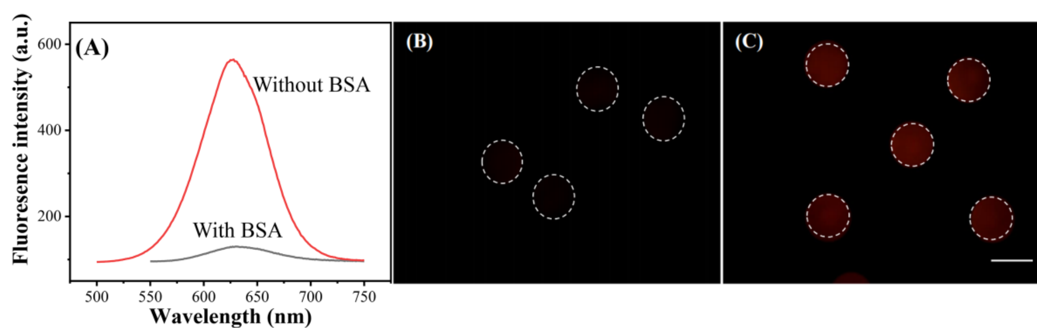


Fig. S3 The blocking effect of blocking agent. (A) Fluorescence signal of capture antibody (Ab_1) immobilized $dSiO_2$ PhC beads. (B) The $dSiO_2$ PhC beads modified with capture antibody (Ab_1) were incubated with 5% BSA in PBS for 2 h, and then incubated with CdTe-labeled Ab_2 for 25 min. The beads were thoroughly washed with PBS and PBST. (C) The capture antibody (Ab_1)-modified $dSiO_2$ PhC beads were directly incubated with CdTe-labeled Ab_2 for 25 min. The beads were thoroughly washed with PBS and PBST. The scale in the picture is 200 μm.

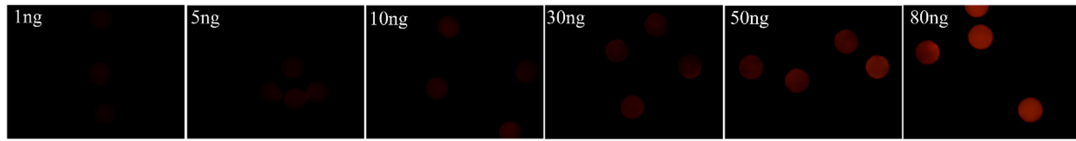


Fig. S4 Fluorescence resulted images at different AFP concentration using $dSiO_2$ PhC beads as carrier.

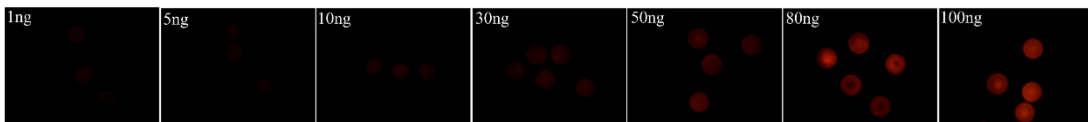


Fig. S5 Fluorescence resulted images at different CEA concentration using $dSiO_2$ PhC beads as carrier.

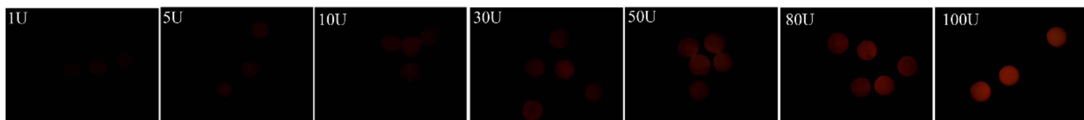


Fig. S6 Fluorescence resulted images at different CA125 concentration using $dSiO_2$ PhC beads as carrier.

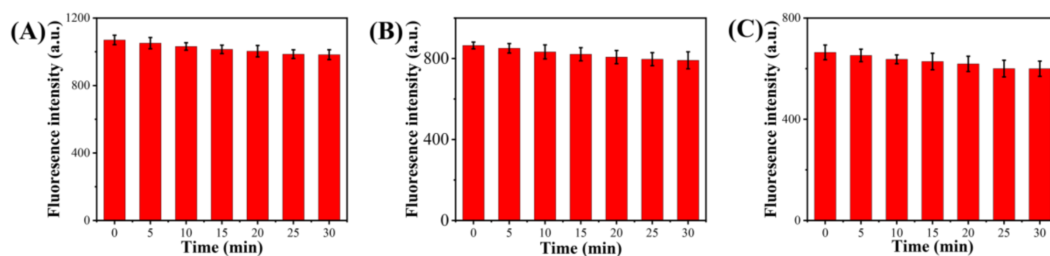


Fig. S7 Stability of (A) AFP, (B) CEA and (C) CA125 based on $dSiO_2$ PhC beads array. The error bars were obtained from three experiments.

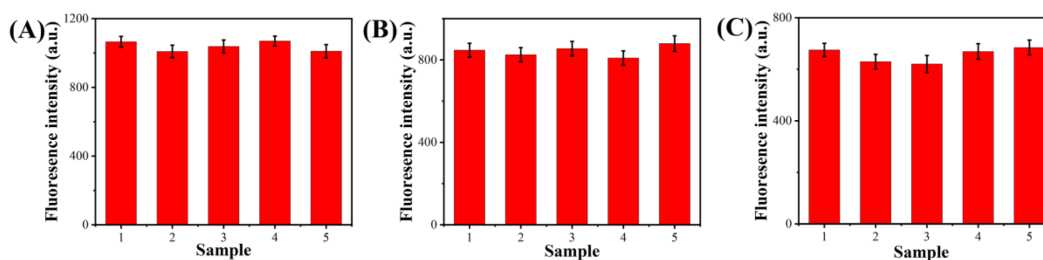


Fig. S8 Reproducibility of (A) AFP, (B) CEA and (C) CA125 based on $dSiO_2$ PhC beads array. The error bars were obtained from three experiments.

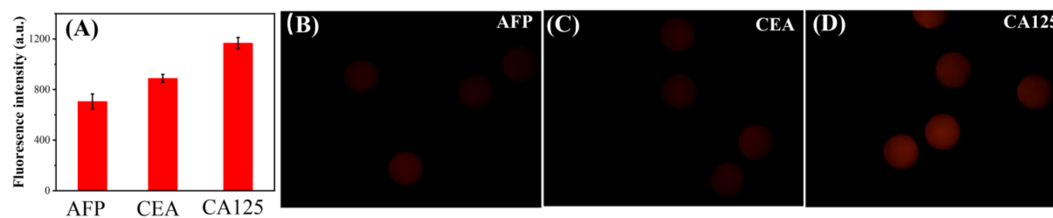


Fig. S9 The average fluorescence intensity of AFP, CEA, and CA125 from the three spiked sera. The error bars were obtained from three experiments.

References

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