Supporting information

Quantification of Cyclic DNA Polymerization with Lanthanide Coordination Nanomaterials for Liquid Biopsy

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1. Effect of PPi on the lifetime of Ce^{3+}.

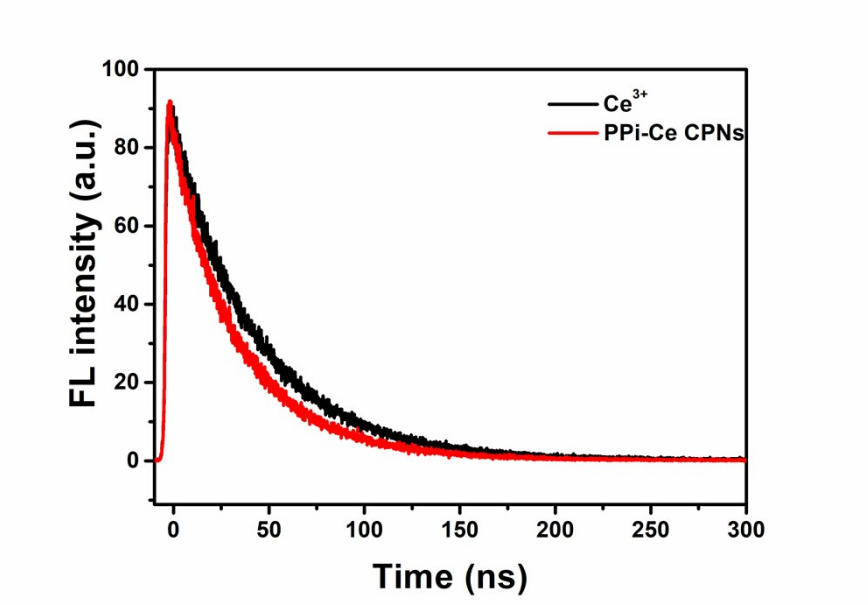

Fig. S1. Fluorescence decay curves of Ce^{3+} and PPi-Ce CPNs.

2. Effect of ratio of PPi to Ce^{3+} on fluorescence intensity of PPi-Ce CPNs.

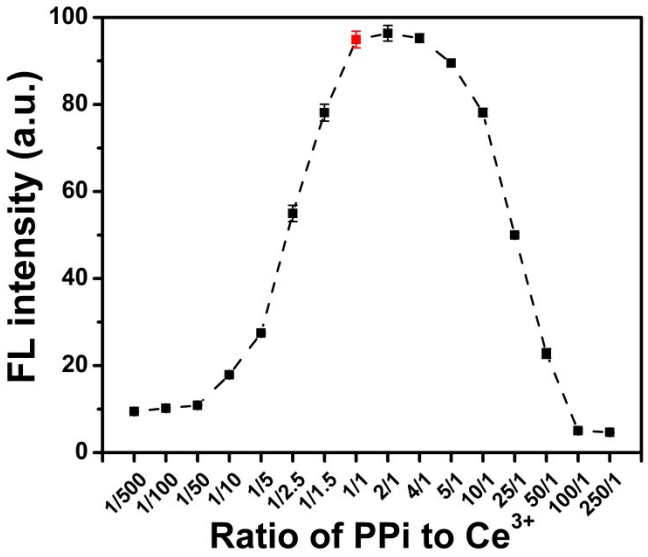

Fig. S2. Fluorescence intensity of Ce^{3+} mixed with different amounts of PPi.

3. Effect of pH on fluorescence intensity of PPi-Ce CPNs.

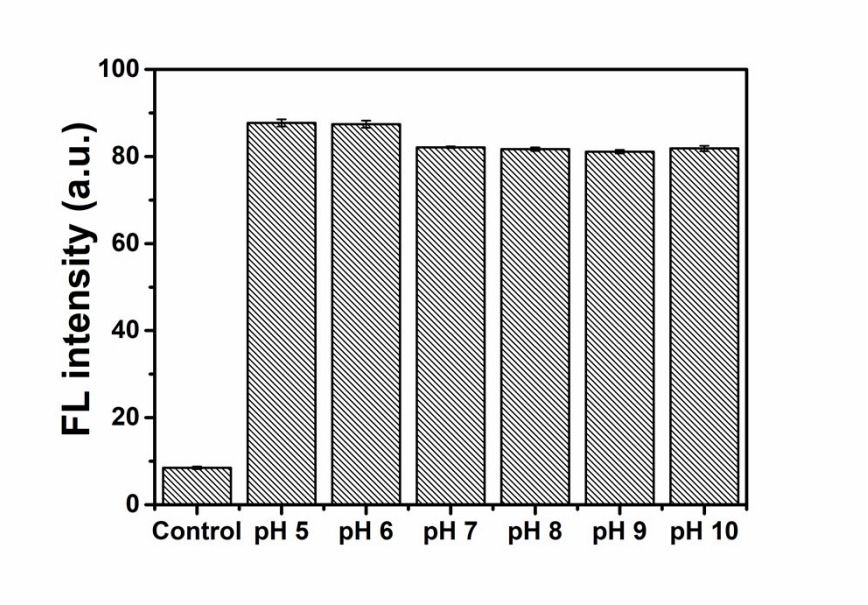

Fig. S3. Fluorescence intensity of PPi-Ce CPNs under different pH values.

4. Effect of PPi and ATP on fluorescence intensity of Ce^{3+}.

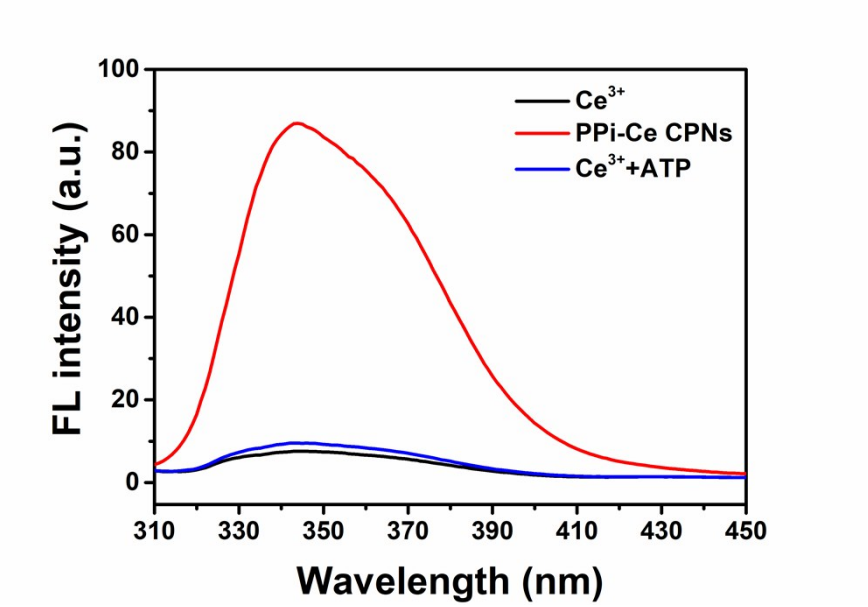

Fig. S4. Fluorescence intensity of Ce^{3+} in the presence of PPi and ATP.

5. Effect of PPi and dNTPs on fluorescence intensity of Ce^{3+}.

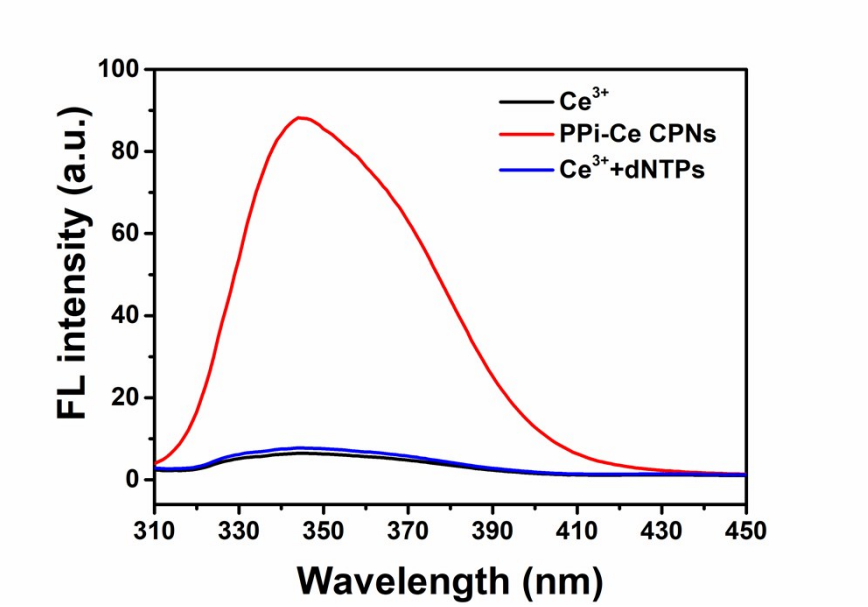

Fig. S5. Fluorescence intensity of Ce^{3+} in the presence of PPi and dNTPs.

6. Optimization of detection conditions.

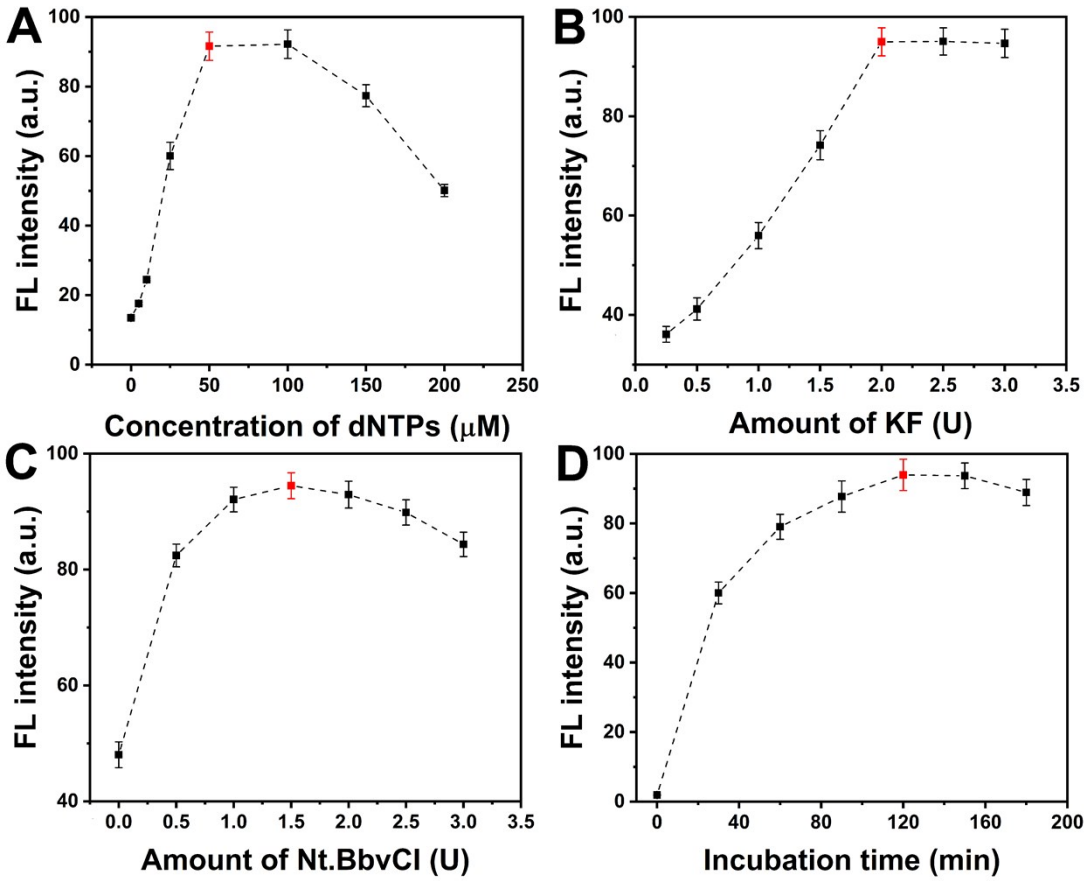

Fig. S6. Optimization of detection conditions of (A) concentration of dNTPs, (B) amount of KF, (C) amount of Nt.BbvCl, and (D) incubation time.

7. Fluorescence spectra obtained in different concentration of ctDNA.

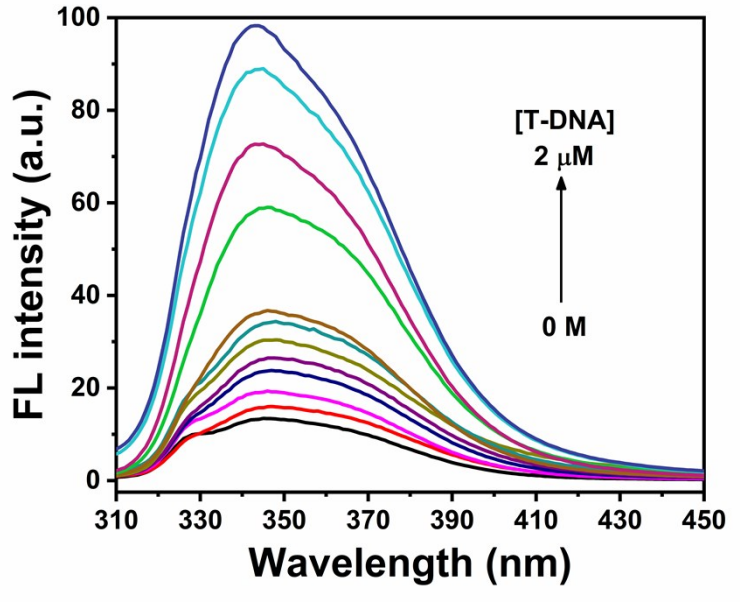

Fig. S7. Fluorescence spectra of PPi-Ce CPNs formed in the presence of various concentrations of ctDNA.

8. Comparison of different methods for the determination of ctDNA.

Table S1. Comparison of analytical performances of different ctDNA biosensors.

Method	Probe	Detection limit	Linear range	Instrument name	Ref.
Electrochemistry	DNA clutch probes	1 fg/μL of mutation DNA in 100 pg/μL of wild-type DNA	Not given	Bioanalytical Systems Epsilon Potentiostat	S1
Electrochemistry	Peptide nucleic acid probe	1 fg /μl	1 - 10^5 fg/μL	Bioanalytical Systems Epsilon Potentiostat	S2
Electrochemistry	Avidin-HRP molecules	0.1%	3×10^3 - 1.5×10^5 fg	Electrochemical Workstation (CHI650E)	S3
Colorimetry	G-quadruplex/hemin complex	100 fM	500 - 5×10^5 fM	UV-vis Spectrophotometer (UV-2550)	S4
Surface-enhanced Raman scattering	Single-walled carbon nanotubes	0.3 fM	10 - 10^6 fM	Confocal Microprobe Raman Instrument (Ram Lab-3010, Horiba Jobin Yvon)	S5
Surface-enhanced Raman scattering	Hairpin DNA-rN1-DNA probes	0.12 fM	0.1 - 107 fM	DXR Smart Raman Spectrometer (Thermo Fisher)	S6
Surface-enhanced Raman scattering	Fluorescence tags	51500 fM	Not given	Renishaw Raman Microspectrometer System (Gloucestershire)	S7
Localized surface plasmon resonance	Gold nanoparticles	50 fM	50 - 3.2×10^3 fM	Integrated System Includes Dark-field (Eclipse TE2000-U, Nikon) Microscope, Spectrograph (Microspec 2300i, RoperScientifics), and CCDcamera (PIXIS: 400B, Princeton Instruments)	S8
Inductively coupled plasma mass spectrometry	Fe-Au Nanoparticle-Coupling	0.1 fg/μL	0.1 - 10^4 fg/μL	Inductively Coupled Plasma Mass Spectrometry (Thermofisher X series 2)	S9
Fluorescence	Fluorophore-labeled DNA probe	4.9×10^4 fg	6.25×10^3 - 9.76×10^7 fg	Fluorescence Spectrophotometer (Model Cary Eclipse, Agilent Technologies)	S10
Fluorescence	PPi-Ce CPNs	0.16 fM or 7.16×10^{-4} fg/μL or 35.8×10^{-4} fg	0.2 - 10^9 fM or 8.94×10^{-4} - 3.58×10^6 fg/μL or 44.7×10^{-4} - 1.79×10^7 fg	Fluoromax-4 Spectrometer (Horiba Jobin Yvon)	This work

9. References

S1. J. Das, I. Ivanov, E. H. Sargent and S. O. Kelley, *J. Am. Chem. Soc.*, 2016, **138**, 11009-11016.

S2. J. Das, I. Ivanov, L. Montermini, J. Rak, E. H. Sargent and S. O. Kelley, *Nat. Chem.*, 2015, **7**, 569-575.

S3. X. Wang, F. Chen, D. Zhang, Y. Zhao, J. Wei, L. Wang, S. Song, C. Fan and Y. Zhao, *Chem. Sci.*, 2017, **8**, 4764-4770.

S4. R. Li, L. Zou, Y. Luo, M. Zhang and L. Ling, *Sci. Rep.*, 2017, **7**, 44212.

S5. Q. Zhou, J. Zheng, Z. Qing, M. Zheng, J. Yang, S. Yang, L. Ying and R. Yang, *Anal. Chem.*, 2016, **88**, 4759-4765.

S6. J. Zhang, Y. Dong, W. Zhu, D. Xie, Y. Zhao, D. Yang and M. Li, *ACS Appl. Mater. Interfaces*, 2019, **11**, 18145-18152.

S7. X. Li, T. Yang, C. S. Li, Y. Song, H. Lou, D. Guan and L. Jin, *Theranostics*, 2018, **8**, 1678-1689.

S8. A. H. Nguyen and S. J. Sim, *Biosens. Bioelectron.*, 2015, **67**, 443-449.

S9. P. Hu, S. Zhang, T. Wu, D. Ni, W. Fan, Y. Zhu, R. Qian and J. Shi, *Adv. Mater.*, 2018, **30**, e1801690.

S10. D. M. Kim, D. H. Kim, W. Jung, K. Y. Lee and D. E. Kim, *Analyst*, 2018, **143**, 1797-1804.