

# Supporting Information

## **A DNzyme sensor that uses chemiluminescence resonance energy transfer for rapid, portable and ratiometric detection of metal ions**

Jiao Zheng,<sup>†,§</sup> Jing Luen Wai,<sup>†,#</sup> Ryan J. Lake,<sup>†</sup> Siu Yee New,<sup>\*,#</sup> Zhike He,<sup>\*,§</sup> Yi Lu<sup>\*,†,‡</sup>

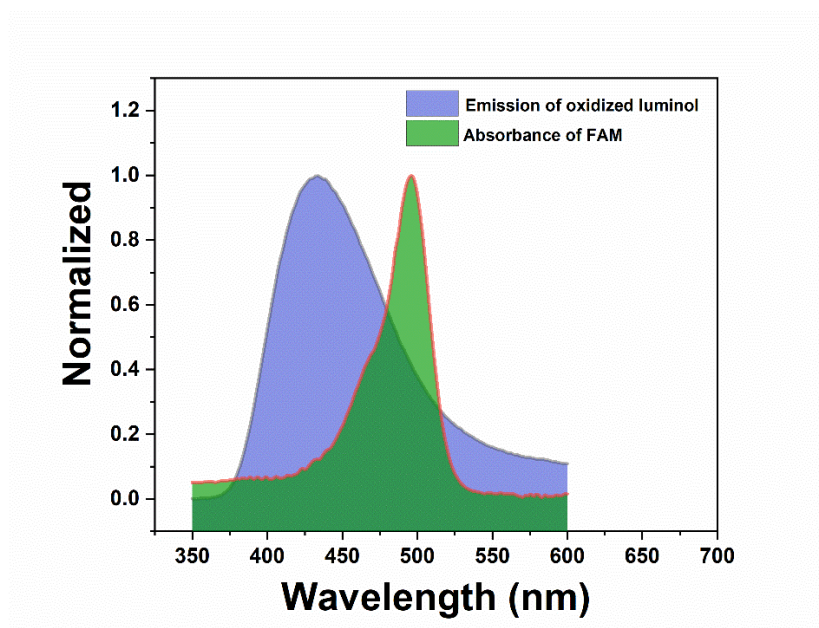
<sup>†</sup>Department of Chemistry, <sup>‡</sup> Department of Biochemistry, University of Illinois at Urbana–Champaign, Urbana, Illinois 61801, United States; Email: [yi-lu@illinois.edu](mailto:yi-lu@illinois.edu)

<sup>§</sup>Key Laboratory of Analytical Chemistry for Biology and Medicine (Ministry of Education), College of Chemistry and Molecular Sciences, Wuhan University, Wuhan 430072, China; Email: [zhkhe@whu.edu.cn](mailto:zhkhe@whu.edu.cn)

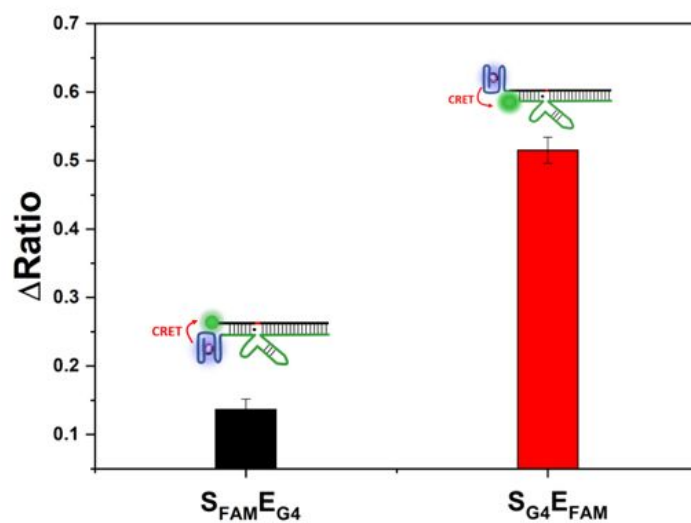
<sup>#</sup> School of Pharmacy, University of Nottingham Malaysia, Jalan Broga, Semenyih, 43500, Selangor, Malaysia; Email: [New@nottingham.edu.my](mailto:New@nottingham.edu.my)

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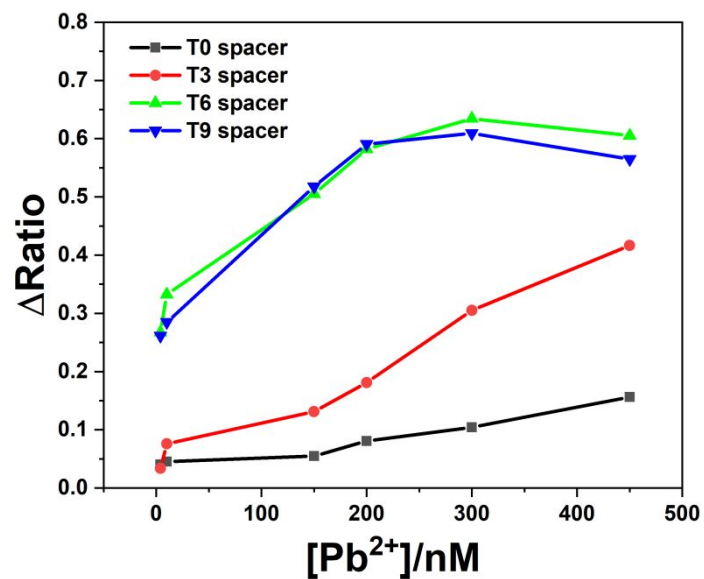
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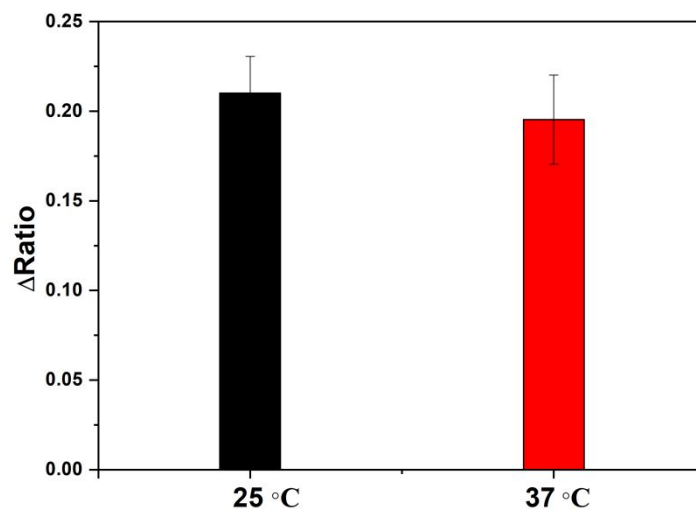
**Figure S1.** Normalized emission (Em.) spectra of oxidized luminol (donor, blue) and normalized absorbance (Abs.) spectra of fluorescein (acceptor, green) with their spectral overlapping area colored dark green.



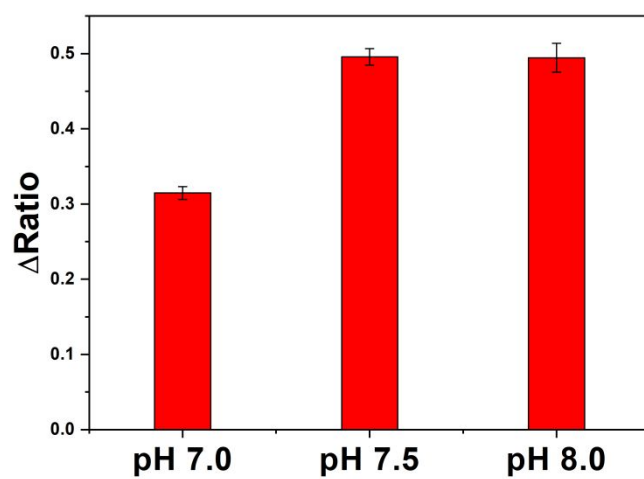
**Figure S2.** The chemiluminescence intensity of designed DNAzyme-CRET probe before addition of  $Pb^{2+}$  ions under varying positions of G4 sequence and fluorescein. Comparing  $S_{FAM}-E_{G4}$  probe and  $S_{G4}-E_{FAM}$  probe, the CRET signal of the latter combination was drastically increased, suggesting the incompatibility of both G4 sequence and catalytic core sequence to be presented on E strand simultaneously.



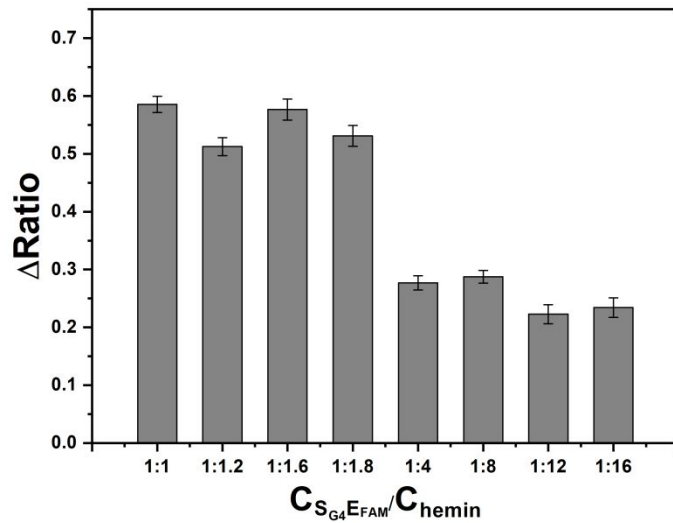
**Figure S3.** The changes of CRET ratio on DNAzyme-CRET probe in respond to increasing  $\text{Pb}^{2+}$  concentrations at different lengths of T-spacer. By increasing the T-spacer from 0 to 9 nucleotides, the CRET signal was drastically increased until a stable  $\Delta\text{Ratio}$  was reached, indicating that the proximity of G4 could interfere with the subsequent sensitivity towards  $\text{Pb}^{2+}$  ions.



**Figure S4.** The effect of varied reaction temperature on DNAzyme-CRET probe after addition of  $\text{Pb}^{2+}$  ions. The CRET signals of the DNAzyme-CRET probe obtained either at 25 °C or 37 °C were indistinguishable.

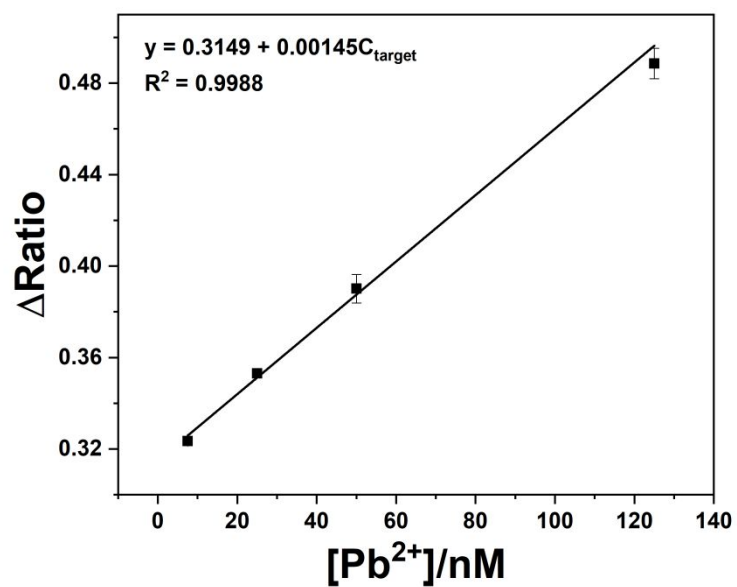


**Figure S5.** The effect of varying reaction pH on DNAzyme-CRET probe after addition of  $\text{Pb}^{2+}$  ions. After an initial sharp increase of  $\Delta\text{Ratio}$ , there is negligible difference between pH 7.5 and pH 8.0, indicating an optimal reaction pH was achieved at pH 7.5.

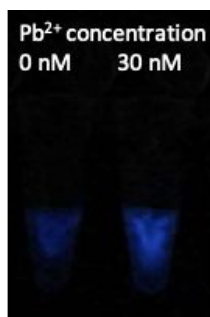


**Figure S6.** Optimizing catalytic oxidation efficiency of luminol by tuning the concentration of hemin. The effect of varying molar ratio between  $S_{G4}-E_{FAM}$  and hemin. With the overall decreasing trend of normalized chemiluminescence intensity, the optimal molar ratio was identified at 1:1.

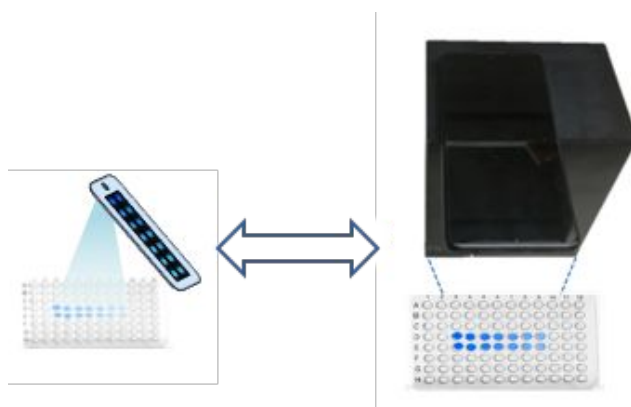




**Figure S7.** Calibration curve achieved by the DNAzyme-CRET sensor for Pb<sup>2+</sup> detection in buffer by fluorometer without an excitation light source. The data collecting procedure was conducted according to the manufacturer's protocol. The linear response range for Pb<sup>2+</sup> is from 7.5 nM to 125 nM.



**Figure S8.** Photographed image for DNAzyme CRET sensor in Tris buffer with addition of 0 and 30 nM  $\text{Pb}^{2+}$  ions respectively.



**Figure S9.** Real working process for smartphone measurement.

**Table S1. Comparison of the reported sensors using DNazymes for Pb<sup>2+</sup>**

NO.	Detection method	Portability	Dynamic range	Limit of detection	Ref.
1	Colorimetric	yes	0.4-2 $\mu$ M	not mentioned	1
2	FRET	no	1-50 nM	0.2-0.5 nM	2
3	Colorimetric	yes	120 nM-20 $\mu$ M	3 nM	3
4	Fluorescent	no	1 nM-1 $\mu$ M	300 pM	4
5	Fluorescent	no	10 nM-4 $\mu$ M	10 nM	5
6	Photoelectrochemical	no	0.1-50 nM	0.05 nM	6
7	CRET	yes	7.5-125 nM	5 nM	this work

## References

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