

## Supplementary Information

### **MoS<sub>2</sub>-Quantum Dots Triggered Reactive Oxygen Species Generation and Depletion: Responsible for Enhanced Chemiluminescence**

Xiangnan Dou,<sup>ab</sup> Qiang Zhang,<sup>a</sup> Syed Niaz Ali Shah,<sup>a</sup> Mashooq Khan,<sup>a</sup> Katsumi Uchiyama<sup>b</sup> and Jin-Ming Lin<sup>\*a</sup>

*<sup>a</sup>Beijing Key Laboratory of Microanalytical Methods and Instrumentation, MOE Key Laboratory of Bioorganic Phosphorus Chemistry & Chemical Biology, Department of Chemistry, Tsinghua University, Beijing, 100084, China*

*<sup>b</sup> Department of Applied Chemistry, Graduate School of Urban Environmental Sciences, Tokyo Metropolitan University, Minamiohsawa, Hachioji, Tokyo 192-0397, Japan*

## Experimental Section

**1. Reagents.** All chemicals used in our work were of analytical grade. Hydrogen peroxide ( $\text{H}_2\text{O}_2$ ), Sodium hydroxide (NaOH), Hydrochloric acid (HCl), Ferrous sulfate( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ), Rhodamine B(RhB), Methylene blue (MB) were brought from Beijing Chemical Reagent Co. (Beijing, China). 2,2,6,6-tetramethyl-4-piperidine (TEMP), 5,5-Dimethyl-1-pyrroline N-oxide (DMPO) and Tetramethylbenzidine (TMB) were purchased from J&K Scientific. Ltd (Beijing, China). Hydrogen peroxide, NaOH and  $\text{Fe}^{2+}$  solution was prepared freshly before using.

**2. Synthesis of  $\text{MoS}_2$  QDs.**  $\text{MoS}_2$  QDs were prepared by top-down method as previously reported. First, 1g of bulk  $\text{MoS}_2$  powder was added in 100 mL of N,N-Dimethylformamide (DMF) and kept sonication for 12 h. The solution was put into flask and heated solvothermally at  $140^\circ\text{C}$  for 6 h with vigorous stirring. Afterwards, the resulting mixture was cooled down to ambient temperature naturally and centrifuged for 10 min at 10000 rpm. The  $\text{MoS}_2$  QDs was included in light yellow DMF supernatant. To obtain  $\text{MoS}_2$  QDs aqueous solution, DMF was removed via rotary evaporation method. Finally, the  $\text{MoS}_2$  QDs was dissolved by ultrapure water to a constant volume.

**3. Characterization of  $\text{MoS}_2$  QDs.** The UV-vis spectra were measured by UV-3900 spectrophotometer (Hitachi, Japan). Emission spectra were collected with F-7000 fluorescence spectrophotometer (Hitachi, Japan). The nanoparticle size was recorded by a JEM 2010 electron microscope (JEOL, Japan). Electron paramagnetic resonance (EPR) spectra were measured on a Model JES-FA200 spectrometer (JEOL, Tokyo, Japan). The fluorescence lifetime was recorded by FLSP920 (Edinburgh Instruments, Livingston, UK).

**4. Chemiluminescence analysis.** Batch CL experiments were carried out with a BPCL luminescence analyzer (Institute of Biophysics, Chinese Academy of Sciences, Beijing, China). The CL spectrum was obtained on the BPCL luminescence analyzer with high-energy cutoff filters from 400 to 640 nm between the flow CL cell and the PMT.

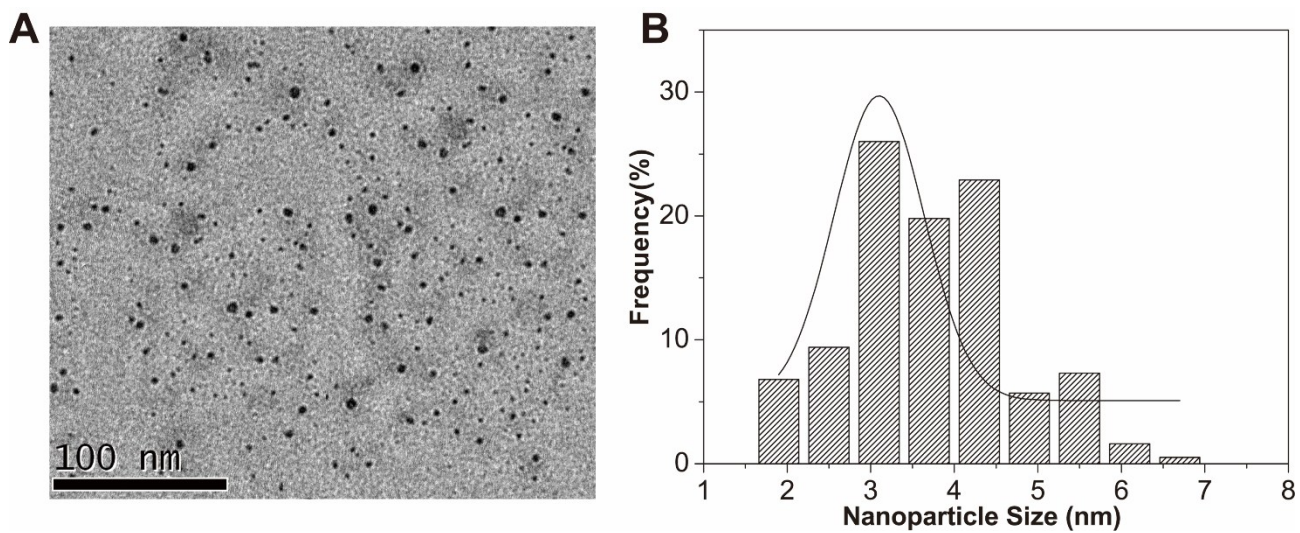


Fig. S1 (A)HRTEM image, and (B) size distribution of MoS<sub>2</sub>-QDs.

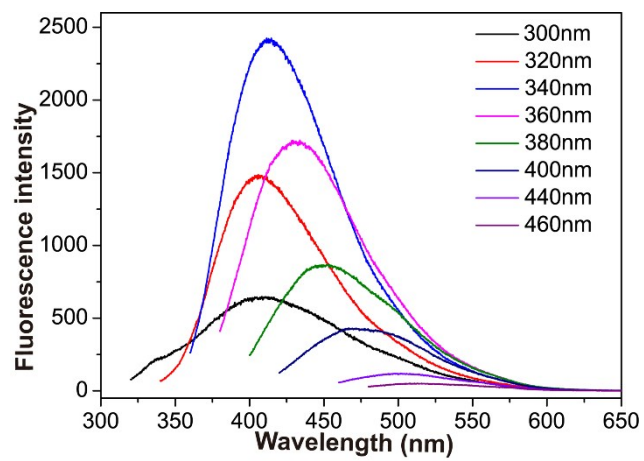


Fig. S2 The fluorescence spectra of MoS<sub>2</sub> QDs (Excitation slit: 2.5 nm, Emission slit: 5 nm; voltage: 700v )

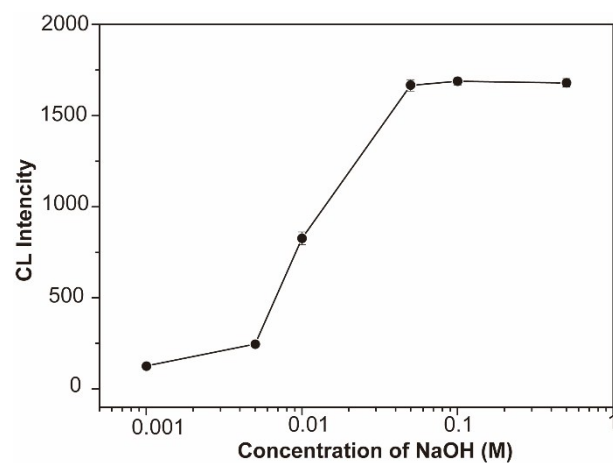


Fig. S3 The correlation of CL intensity with the concentration of sodium hydroxide (NaOH). The solution conditions were 50  $\mu\text{L}$  of 0.1 M  $\text{H}_2\text{O}_2$ , 50  $\mu\text{L}$  of 0.45mg/ml  $\text{MoS}_2$  QDs, and 50  $\mu\text{L}$  of sodium hydroxide with concentration of  $10^{-3}$ ,  $5 \times 10^{-3}$ , 0.01, 0.05, 0.1, 0.5M respectively. High voltage: 1300 V; interval time was set for 0.1 s.

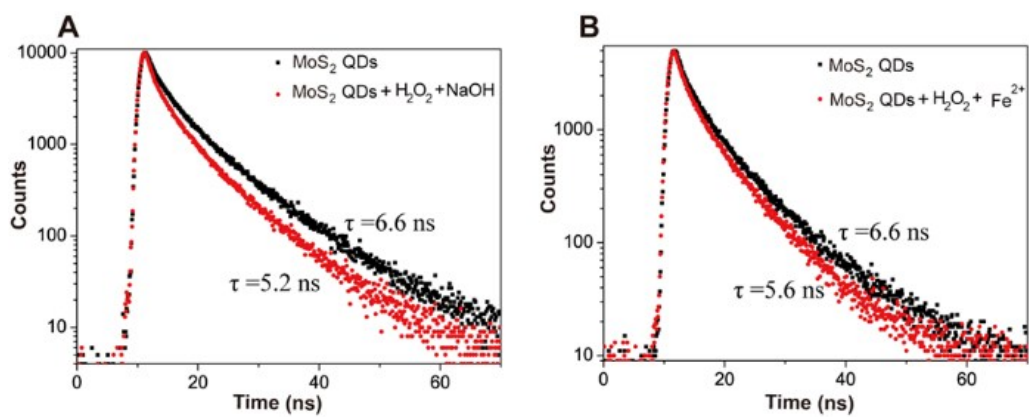


Fig. S4 (A) The fluorescence lifetime decay profile of MoS<sub>2</sub> QDs before (black) and after reaction with H<sub>2</sub>O<sub>2</sub>-NaOH; (B) The fluorescence lifetime decay curve of MoS<sub>2</sub> QDs before (black) and after reaction with H<sub>2</sub>O<sub>2</sub>-Fe<sup>2+</sup>. The solution conditions were 100  $\mu$ l of 0.1 M H<sub>2</sub>O<sub>2</sub>, 100  $\mu$ l of 0.45 mg/ml MoS<sub>2</sub> QDs, 100  $\mu$ l of 0.1 M NaOH and 100  $\mu$ L of 0.1 M Fe<sup>2+</sup>. The excitation wavelength: 325 nm, emission wavelength: 420 nm.

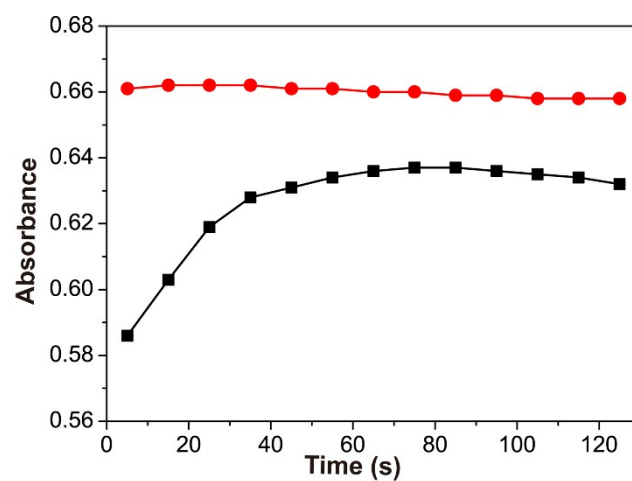
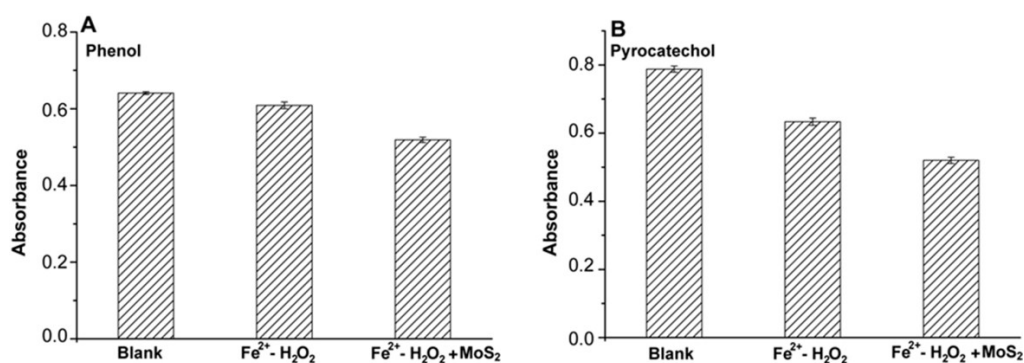
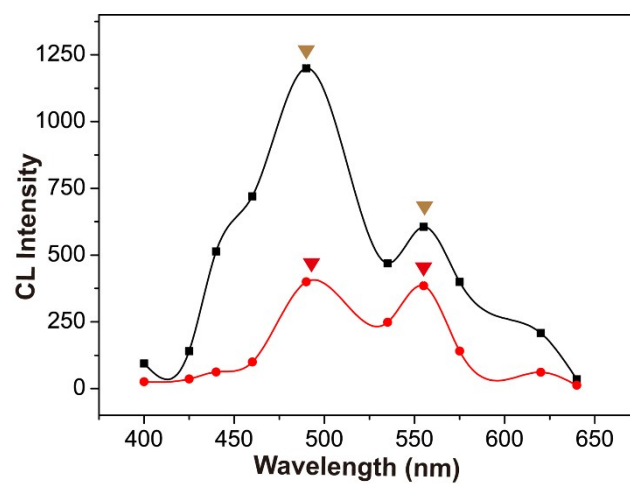


Fig. S5 The absorbance of TMB in  $\text{H}_2\text{O}_2\text{-Fe}^{2+}$  system(black) and  $\text{MoS}_2\text{ QDs-H}_2\text{O}_2\text{-Fe}^{2+}$  system. The solution conditions were  $100\mu\text{L}$  of  $0.01\text{ M H}_2\text{O}_2$ ,  $100\text{L}$  of  $0.45\text{mg/ml MoS}_2\text{ QDs}$ ,  $100\text{ul}$  of  $1\text{mM TMB}$  and  $100\mu\text{L}$  of  $1\text{mM Fe}^{2+}$ .

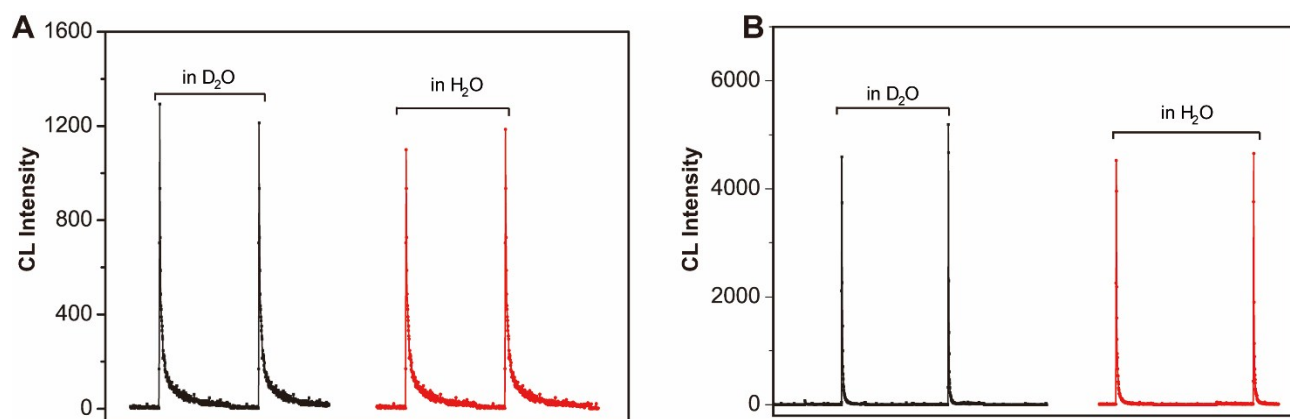


**Fig. S6** Comparison of phenolic compounds degradation in Fe<sup>2+</sup>-H<sub>2</sub>O<sub>2</sub> Fenton system and Fe<sup>2+</sup>-H<sub>2</sub>O<sub>2</sub> + MoS<sub>2</sub> QDs system. Phenolic compounds can react with 4-aminoantipyrine forming aminoantipyrine dye with absorbance at wavelength 530.5 nm. The initial concentration of phenolic compounds and final concentration after degradation by Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub> Fenton system and Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub>-MoS<sub>2</sub>-QDs system were measured by using 4-aminoantipyrine for the colorimetric determination. (A) The absorbance of 4-aminoantipyrine reacted with phenol after incubated in Fe<sup>2+</sup>-H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup>-H<sub>2</sub>O<sub>2</sub> + MoS<sub>2</sub>-QDs for 10min; (B) The absorbance of 4-aminoantipyrine reacted with pyrocatechol after incubated in Fe<sup>2+</sup>-H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup>-H<sub>2</sub>O<sub>2</sub> + MoS<sub>2</sub>-QDs for 10min. The solution conditions were 0.01 M H<sub>2</sub>O<sub>2</sub>, 0.45 mg/ml MoS<sub>2</sub>-QDs, 1mM Fe<sup>2+</sup>, 0.01 M K<sub>3</sub>Fe(CN)<sub>6</sub>, 1×10<sup>-4</sup>M phenol and 5×10<sup>-4</sup>M pyrocatechol.





**Fig. S7** The CL spectrum of MoS<sub>2</sub> QDs-H<sub>2</sub>O<sub>2</sub>-NaOH system(red) and MoS<sub>2</sub> QDs-H<sub>2</sub>O<sub>2</sub>-Fe<sup>2+</sup> system(black). The solution conditions were 50 $\mu$ L of 0.1 M H<sub>2</sub>O<sub>2</sub>, 50L of 0.45mg/ml MoS<sub>2</sub> QDs, 50ul of 0.1 M NaOH and 50 $\mu$ L of 0.1 M Fe<sup>2+</sup>.



**Fig. S8** (A) The comparison of CL of MoS<sub>2</sub> QDs-H<sub>2</sub>O<sub>2</sub>-NaOH system in D<sub>2</sub>O and H<sub>2</sub>O reagent respectively. (B) The comparison of CL of MoS<sub>2</sub> QDs-H<sub>2</sub>O<sub>2</sub>- Fe<sup>2+</sup> system in D<sub>2</sub>O and H<sub>2</sub>O reagent respectively.