

## Electronic Supporting Information

### HP-DO3A-based amphiphilic MRI contrast agents and their relaxation enhancement through assembly with polyelectrolytes

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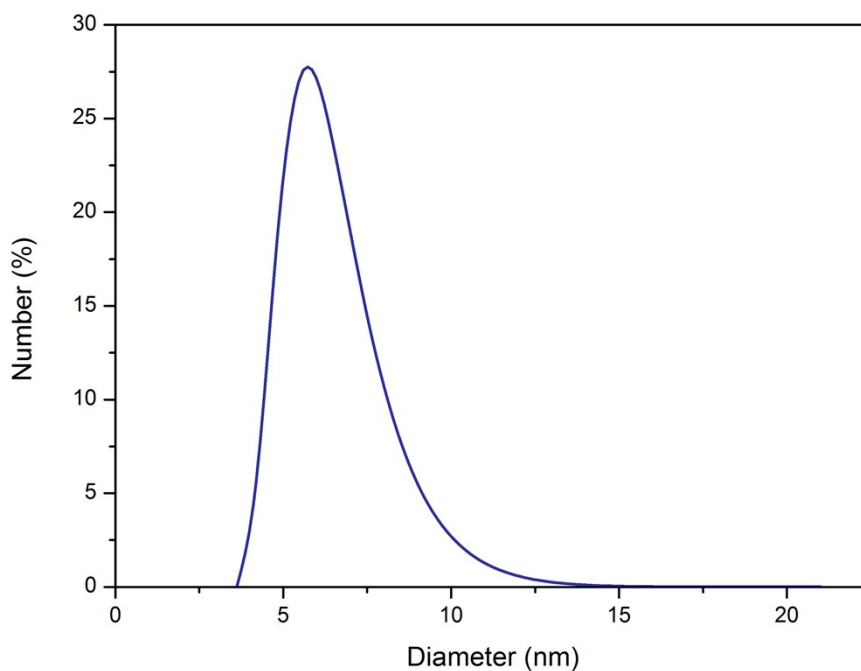


Fig. S1 Dynamic light scattering curve of GdL<sub>1</sub>.

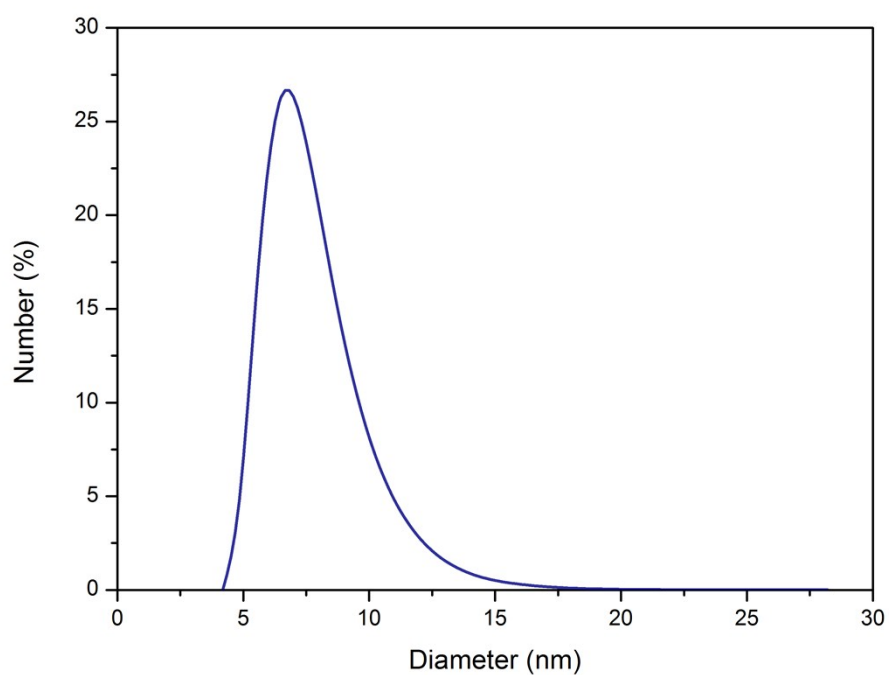


Fig. S2 Dynamic light scattering curve of GdL<sub>2</sub>.

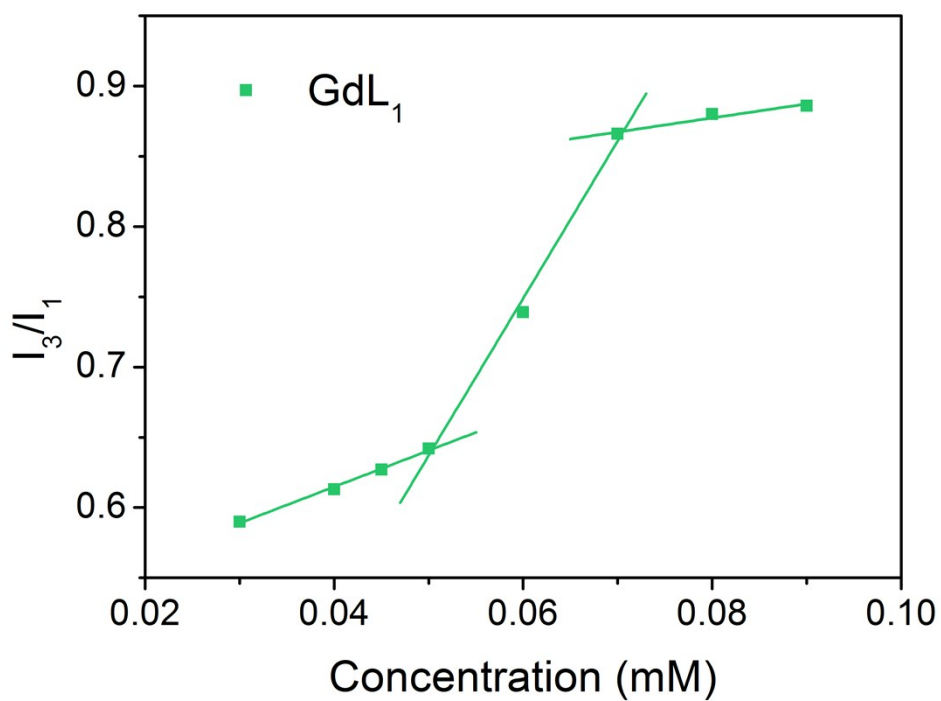


Fig. S3 Fluorescence intensity ratio ( $I_3/I_1$ ) of pyrene versus Gd concentration of GdL<sub>1</sub> (Excitation: 334 nm,  $I_3$ : 383 nm,  $I_1$ : 372nm). The first inflection point indicates the CMC<sup>1</sup>.

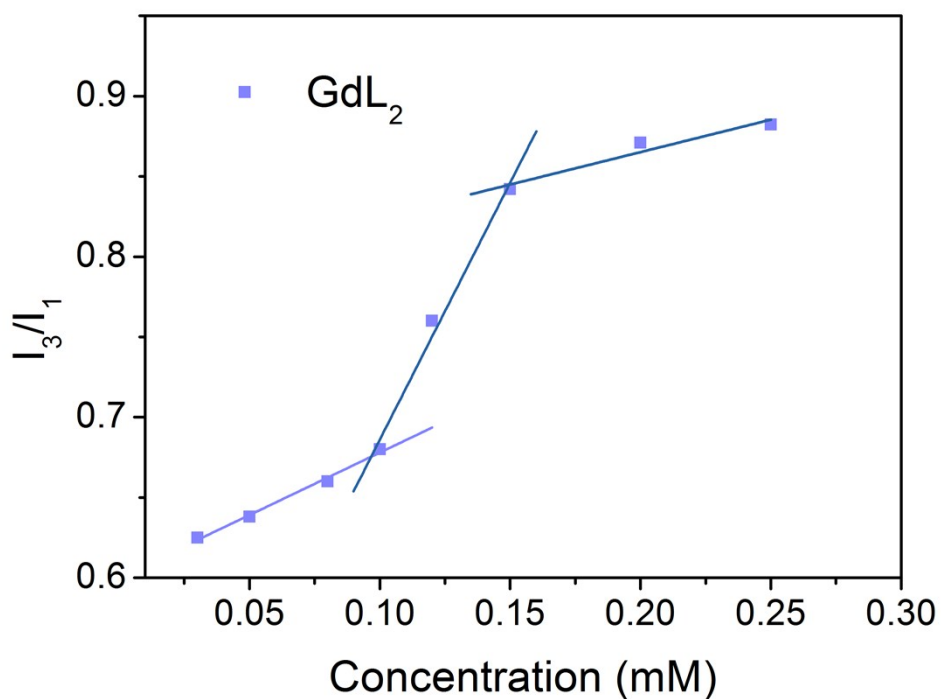


Fig. S4 Fluorescence intensity ratio ( $I_3/I_1$ ) of pyrene versus Gd concentration of  $GdL_2$  (Excitation: 334 nm,  $I_3$ : 383 nm,  $I_1$ : 372nm). The first inflection point indicates the CMC.



Fig. S5 Color photographs of xylene orange solution<sup>2</sup> at pH 5 in the presence of 20  $\mu\text{M}$  (a), 15  $\mu\text{M}$  (b), 10  $\mu\text{M}$  (c), 5  $\mu\text{M}$  (d), 1  $\mu\text{M}$  (e), 0  $\mu\text{M}$  (f) of Gd(III) ion and 1mM Gd chelates ( $GdL_1$ ) (g), respectively. The concentration of xylene orange solution is 12 mg/L.



Fig. S6 Color photographs of xylenol orange solution at pH 7.4 in the presence of 20  $\mu\text{M}$  (a), 15  $\mu\text{M}$  (b), 10  $\mu\text{M}$  (c), 5  $\mu\text{M}$  (d), 1  $\mu\text{M}$  (e), 0  $\mu\text{M}$  (f) of Gd(III) ion and 1mM Gd chelates ( $\text{GdL}_1$ ) (g), respectively. The concentration of xylenol orange solution is 12 mg/L.

## Reference

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2. A. Barge, G. Cravotto, E. Gianolio and F. Fedeli, *Contrast Media & Molecular Imaging*, 2006, **1**, 184-188.