

**Fluoride-specific fluorescence/MRI bimodal probe based on a  
Gadolinium(III)-flavone complex: Synthesis, mechanism and bioimaging  
application *in vivo***

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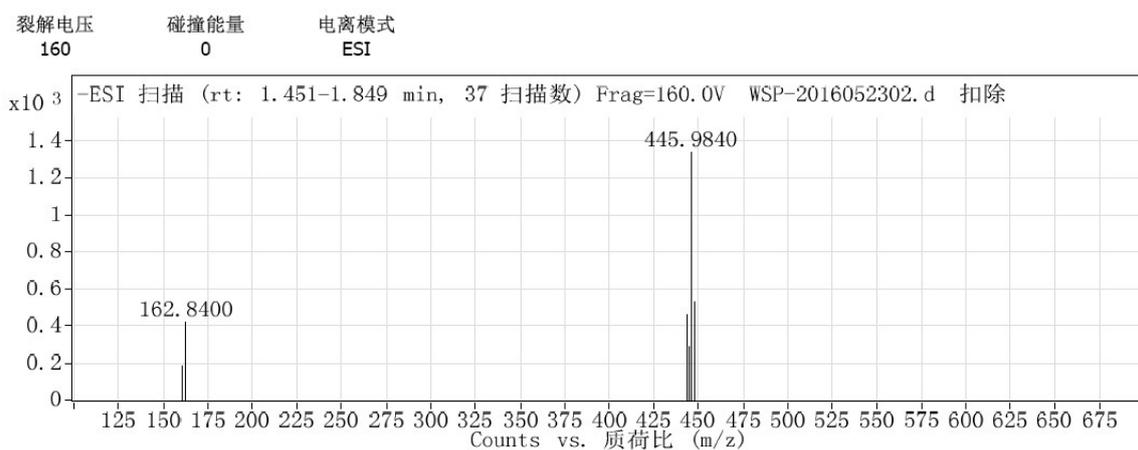
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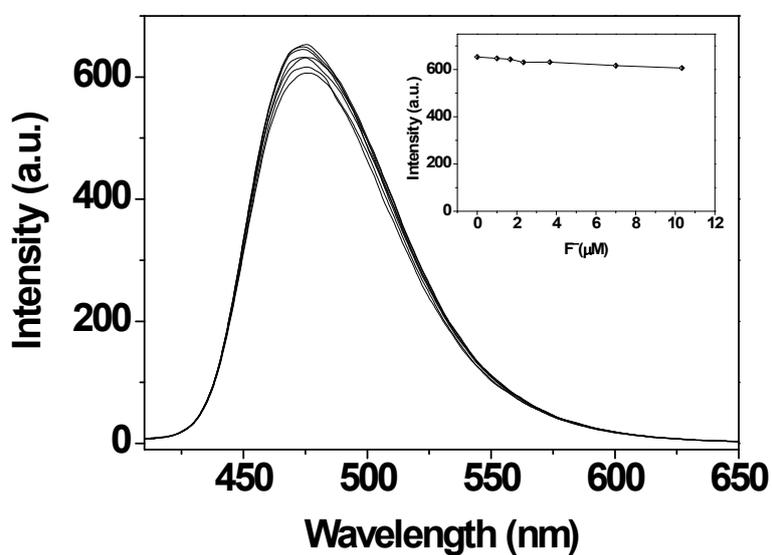
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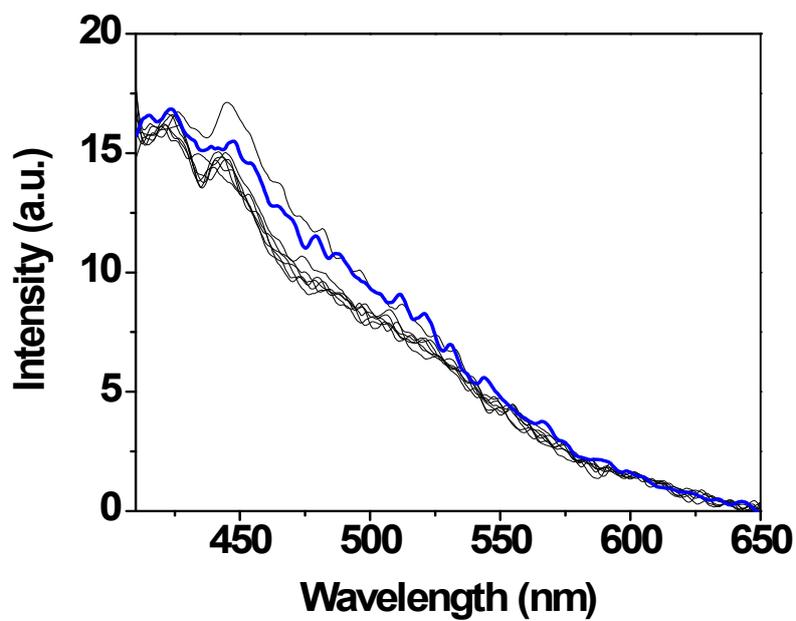
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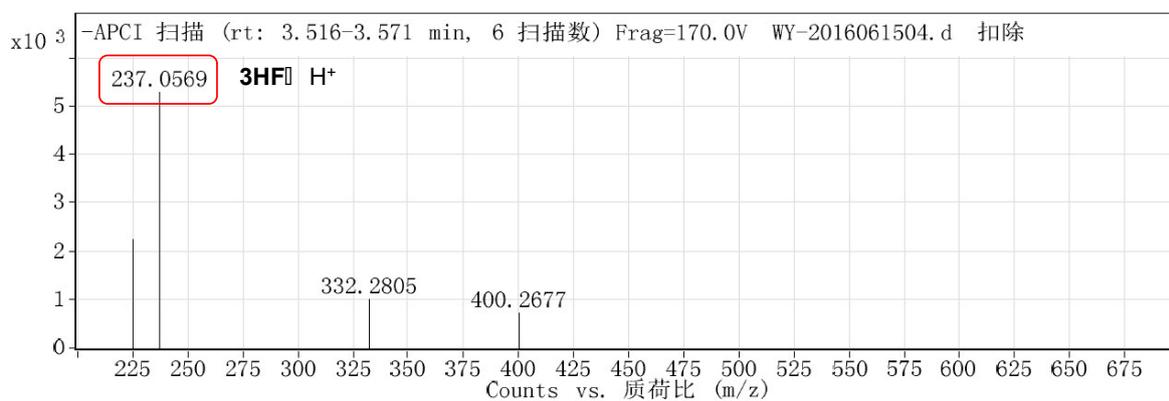
**Fig. S1.** ESI-MS spectrum of [EDTA-Gd]<sup>-</sup>



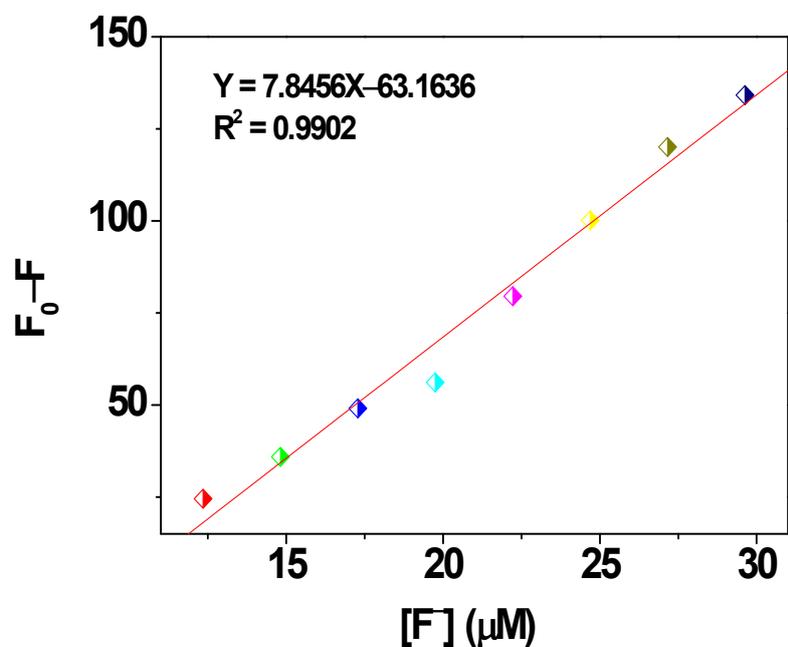
**Fig. S2.** Fluorescence spectra of EDTA-Gd-**HF** (10  $\mu\text{M}$ ) in the presence of increasing amount of fluoride ions (0–10  $\mu\text{M}$ ) in aqueous ( $\text{C}_2\text{H}_5\text{OH}:\text{H}_2\text{O} = 3:7$ ). Inset: plot of the fluorescence intensity changes of EDTA-Gd-**HF** (10  $\mu\text{M}$ ) observed at 476 nm versus fluoride ions concentration. Excitation was performed at 400 nm.



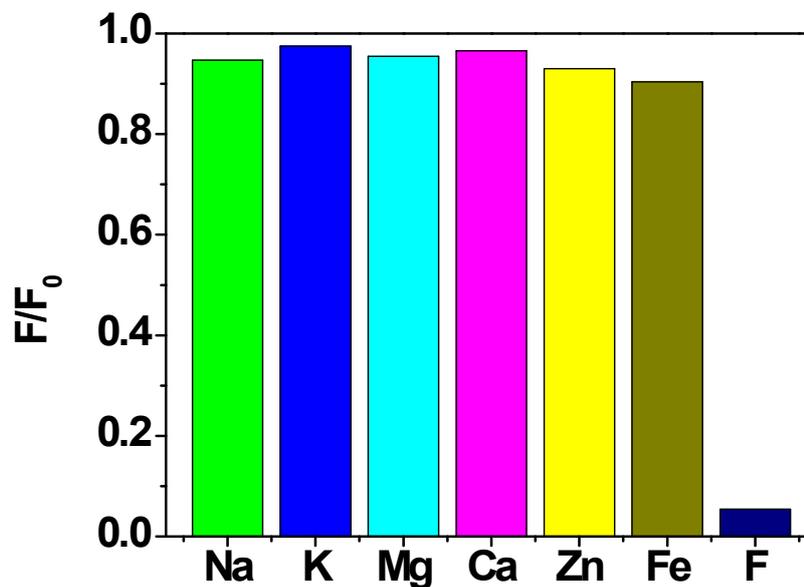
**Fig. S3.** Fluorescence spectra of **HF** (10  $\mu\text{M}$ ) in the presence of increasing amount of fluoride ions in aqueous ( $\text{C}_2\text{H}_5\text{OH}$ :  $\text{H}_2\text{O}$  = 3:7). Excitation was performed at 400 nm.



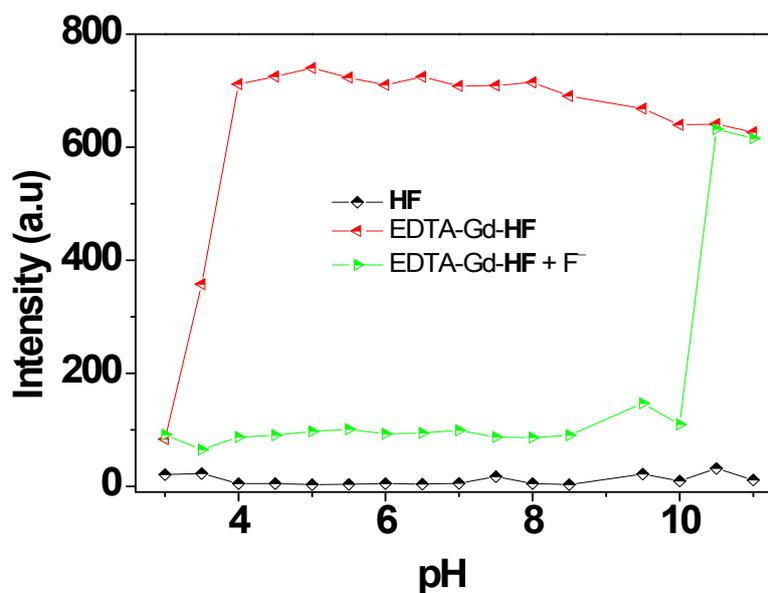
**Fig. S4.** ESI-MS spectrum of EDTA-Gd-**HF** in presence of fluoride ions in aqueous



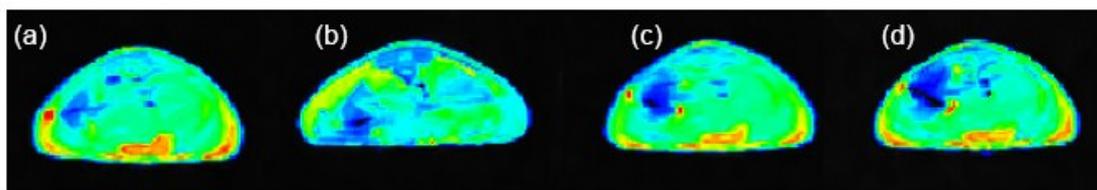
**Fig. S5.** Linear relationship between fluorescence intensity of EDTA-Gd-HF (3 μM) at 476 nm versus the concentration of fluoride ion (10–30 μM) in aqueous (CH<sub>3</sub>OH: H<sub>2</sub>O = 3:7). Excitation was performed at 400 nm.



**Fig. S6.** Fluorescence responses of EDTA-Gd-HF (10 μM) in the presence of various biological cations (0.5 mM) in aqueous (C<sub>2</sub>H<sub>5</sub>OH: H<sub>2</sub>O = 3:7). The intensities were recorded at 476 nm. Excitation was performed at 400 nm.



**Fig. S7.** Influence of pH on the fluorescence intensities of **HF**, EDTA-Gd-**HF** and EDTA-Gd-**HF** in the presence of fluoride ion. The intensities were recorded at 476 nm. Excitation was performed at 400 nm.



**Fig. S8.** T<sub>1</sub>-weighted MR transection images of living white mice after injection of: (a) EDTA-Gd-**HF** (0.2 mL, 0.2 mM), (b) 3.5 mM fluoride ions only, (c) 3.5 mM fluoride ions followed by injection of 0.2 mL EDTA-Gd-**3HF** (0.2 mM), and (d) further injection of 3.5 mM mixed anions into (c).