

## Electronic Supplementary Information

### High-efficiency and Safe Sulfur-Doped Iron Oxides for Magnetic Resonance Imaging Guided Photothermal/Magnetic Hyperthermia Therapy

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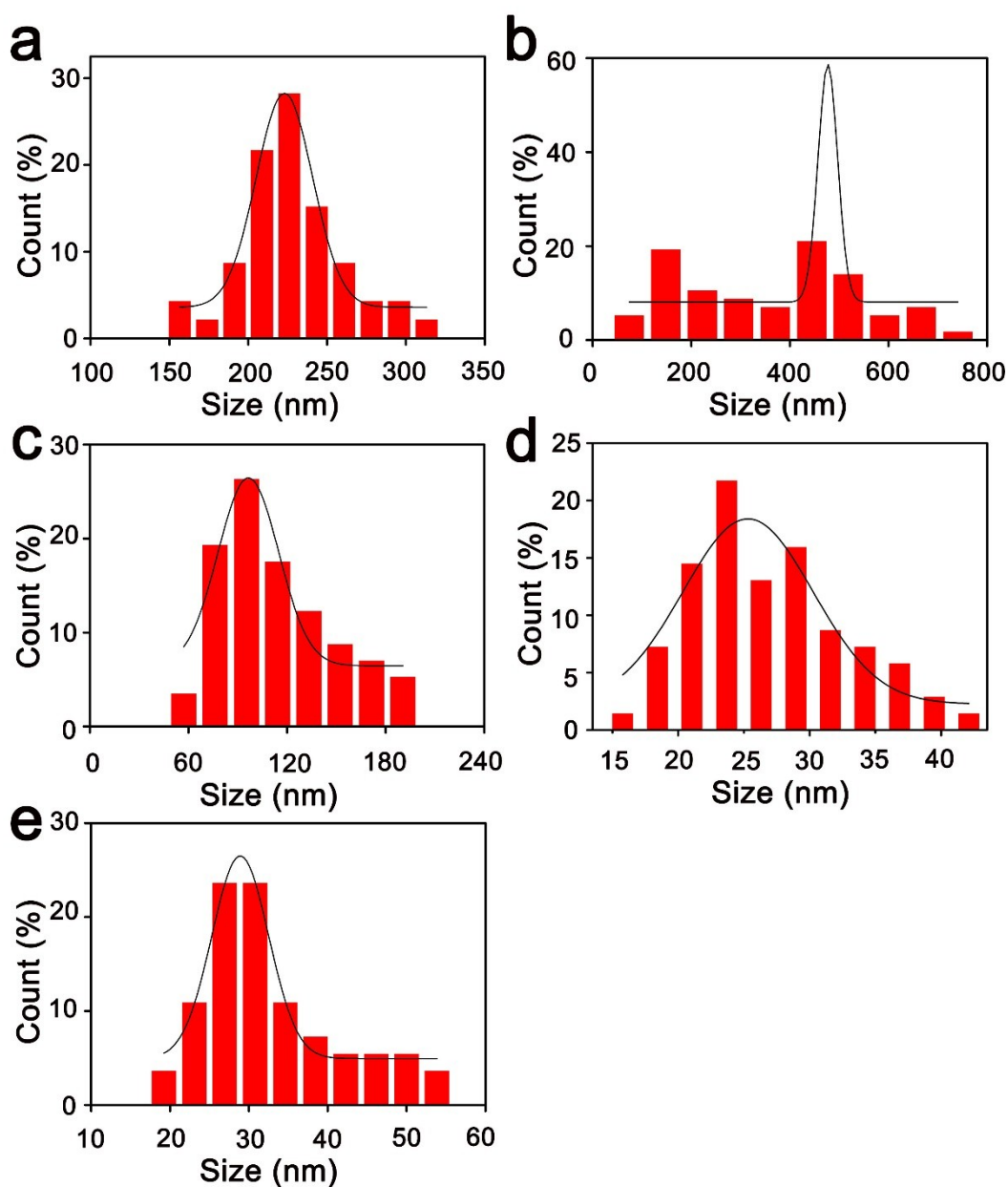
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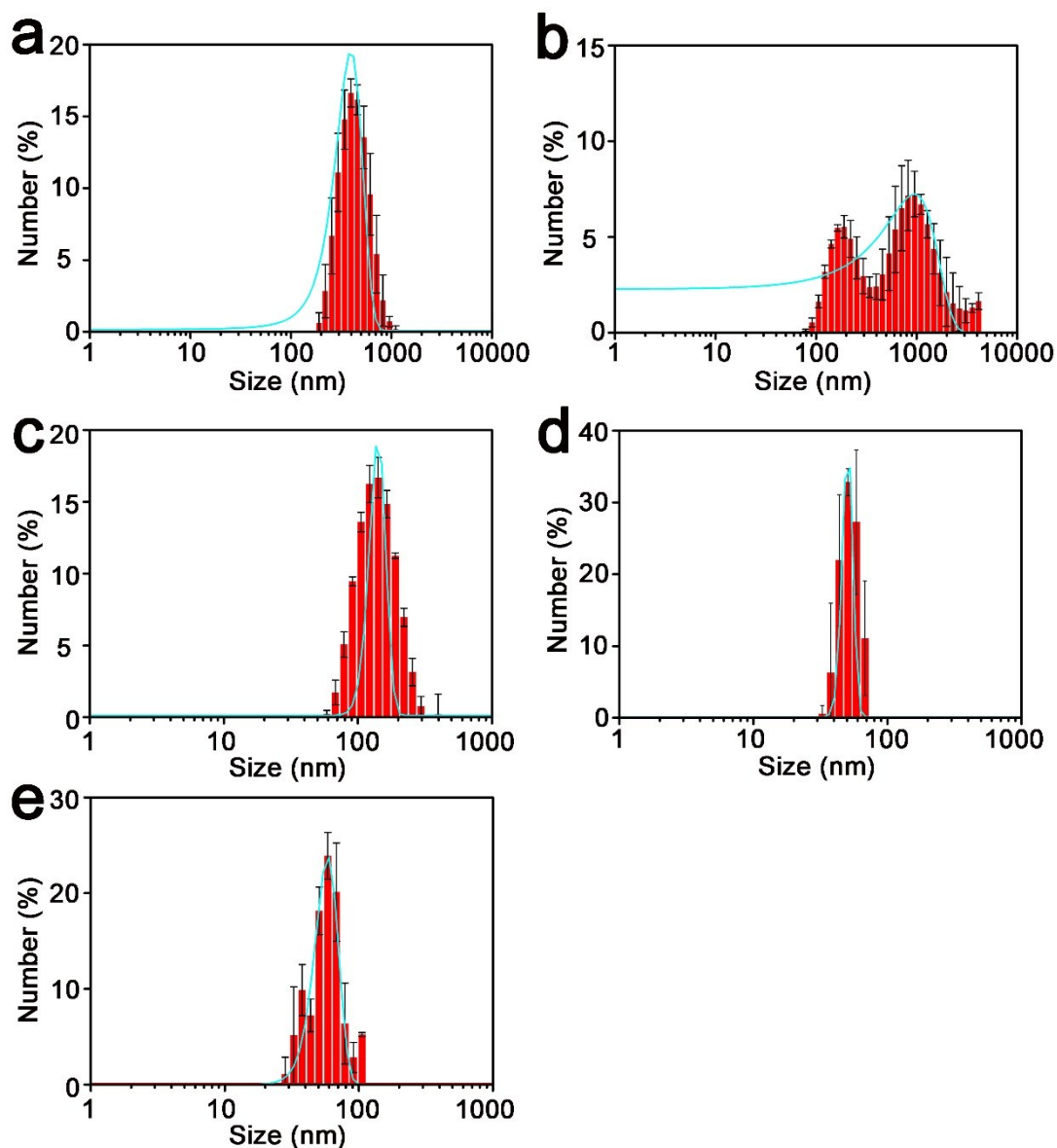
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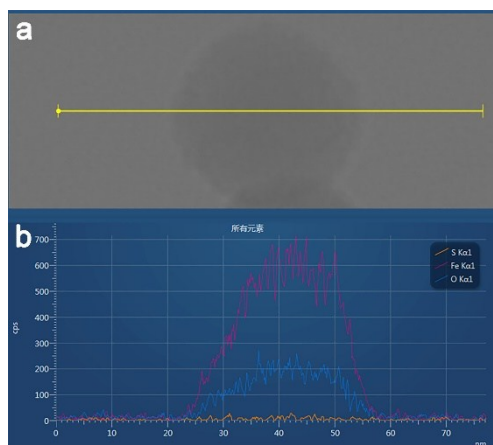
\* Corresponding Author.



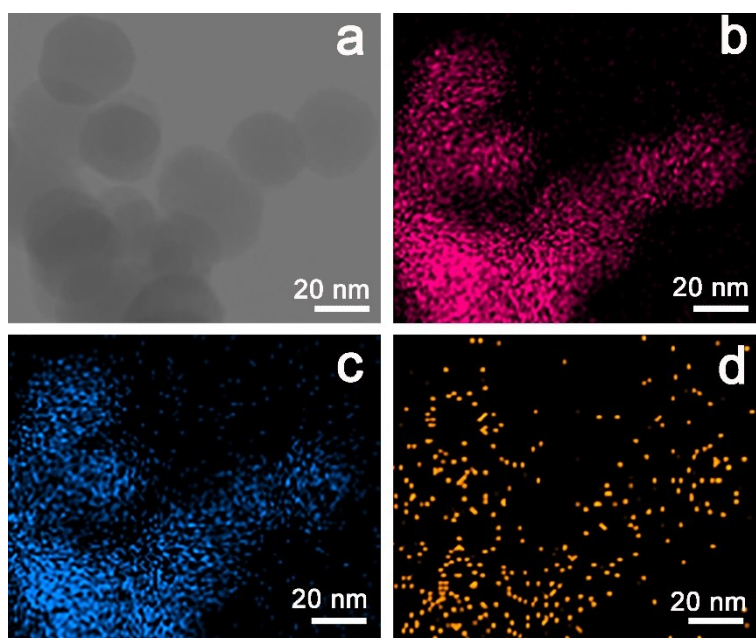
**Fig. S1** Size distribution histogram of over 100 randomly selected nanoparticles from the corresponding TEM images. (a) The long edge length of SDIO-0 is  $223.0 \pm 35.9$  nm. (b) The long edge length of SDIO-25 is  $477.3 \pm 39.8$  nm. (c) The mean size of SDIO-50 is  $96.4 \pm 36.7$  nm. (d) The mean size of SDIO-100 is  $25.3 \pm 9.8$  nm. (e) The edge length of SDIO-200 is  $28.9 \pm 7.1$  nm. Data is presented as the mean  $\pm$  s.e.m.



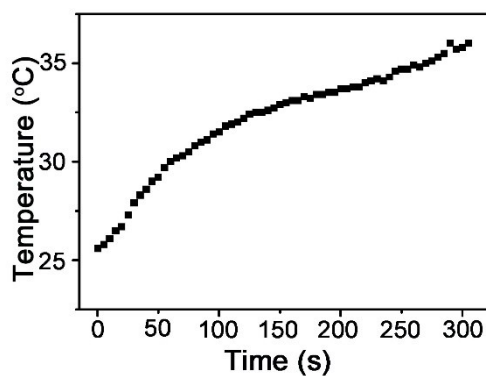
**Fig. S2** DLS of the SDIOs in PBS solution for three times. (a) The hydrodynamic diameter (HD) of SDIO-0 is  $396.1 \pm 240.4$  nm. (b) The hydrodynamic diameter of SDIO-25 is  $955.4 \pm 628.7$  nm. (c) The hydrodynamic diameter of SDIO-50 is  $141.8 \pm 39.6$  nm. (d) The hydrodynamic diameter of SDIO-100 is  $50.7 \pm 9.1$  nm. (e) The hydrodynamic diameter of SDIO-200 is  $58.7 \pm 11.6$  nm. Data is presented as the mean  $\pm$  s.e.m.



**Fig. S3** The corresponding EDX line scan profiles of SDIO-100 nanoparticle, indicating small amount of S doping in the iron oxide crystal.

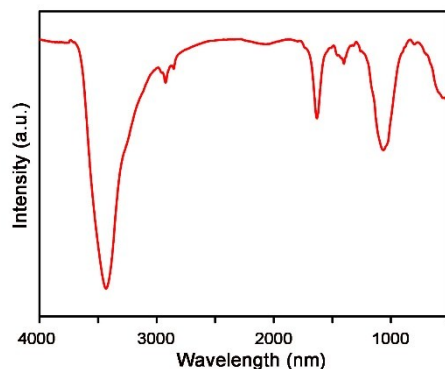


**Fig. S4** STEM-EDS elemental maps of the nanocrystals. (a) STEM image, elemental mapping (Fe: (b), O: (c), S: (d)) of the sample.

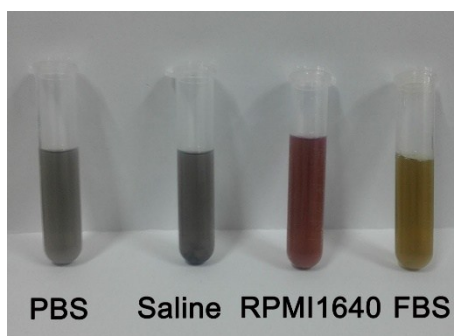


**Fig. S5** Heating curves of the IO-100 aqueous dispersion in an alternating magnetic

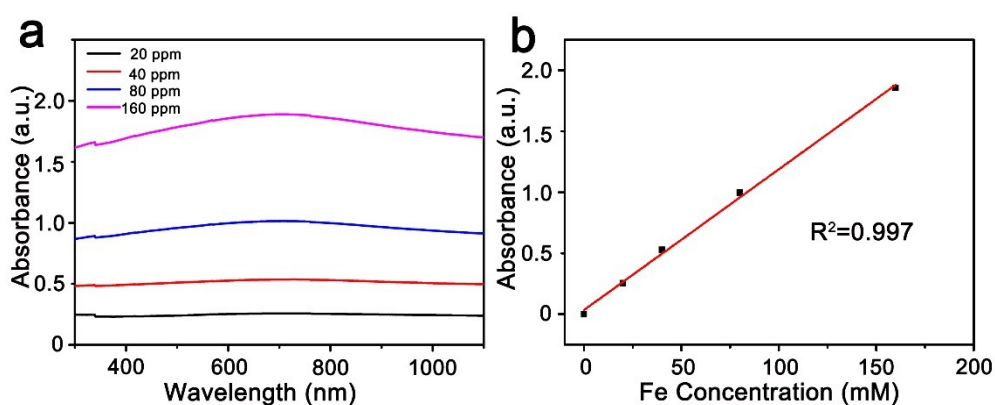
field for 300 s.



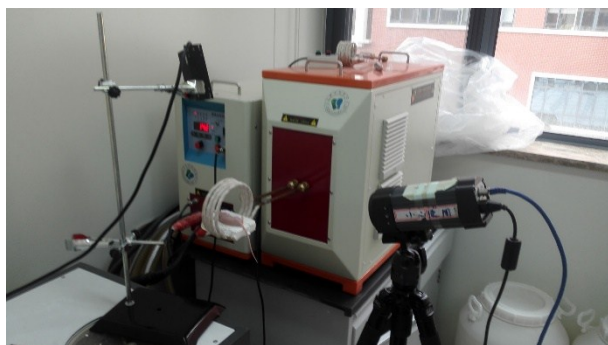
**Fig. S6** FTIR spectrum of the SDIO-100. The bands at 2850-2950  $\text{cm}^{-1}$  are due to the C-H stretching vibration from the PVP and the band around 1400  $\text{cm}^{-1}$  is assigned to the C-H deformation vibration in the PVP.



**Fig. S7** A camera image of the SDIO-100 dispersed in PBS, Saline, RPMI1640, FBS for over one day.



**Fig. S8** (a) UV-Vis-NIR spectra of the SDIO-100 aqueous solution at different Fe concentrations. (b) The corresponding linear relationship between the absorbance at 808 nm and the concentration.



**Fig. S9** The camera image shows the in vivo synergistic treatment combination between PTT and MH.

**Table S1.** The detailed peak positions for the fitted Fe peaks.

Samples	Fe <sup>2+</sup> 2p <sub>3/2</sub>	Fe <sup>2+</sup> 2p <sub>1/2</sub>	Fe <sup>3+</sup> 2p <sub>3/2</sub>	Fe <sup>3+</sup> 2p <sub>1/2</sub>
SDIO-0	710.1	723.7	712.0	725.5
SDIO-25	710.4	723.9	712.6	725.7
SDIO-50	710.2	723.8	712.2	726.0
SDIO-100	709.9	723.1	712.6	725.5
SDIO-200	709.9	722.9	712.9	725.1

**Table S2.** The detailed peak positions for the fitted O peaks.

Samples	Fe-O	chemisorbed O	S-O
Fe-0	529.4	530.9	532.2
Fe-25	529.2	530.3	532.7
Fe-50	529.2	530.7	532.0
Fe-100	529.0	530.7	532.3
Fe-200	529.1	530.3	531.6

**Table S3.** The detail peak positions for the fitted S peaks.

Samples	S <sup>2-</sup> 2p <sub>3/2</sub>	S <sup>2-</sup> 2p <sub>1/2</sub>	S=C	FeIII-S	Fe <sub>x</sub> O <sub>y</sub> S <sub>z</sub>	Fe <sub>x</sub> O <sub>y</sub> S <sub>z</sub>
Fe-0	161.8	162.3	163.2	165.3	168.0	169.4
Fe-25	161.8	162.2	163.0	165.4	168.0	169.3
Fe-50	161.7	162.5	163.4	165.2	167.8	168.5

Fe-100	161.2	162.3	163.1	164.9	167.9	168.8
Fe-200	161.8	162.8	163.4	165.0	167.0	168.5

**Table S4.** Determination of the mass ratio of S to Fe in the SDIO samples which the ethylenediamine with different volumes was added.

	SDIO-0	SDIO-25	SDIO-50	SDIO-100	SDIO-200
S/Fe (ICP)	16.89 %	7.14 %	1.04 %	0.87 %	1.69 %

**Table S5.** The specific absorption rate (SAR) values of SDIOs and IO-100.

Sample	SDIO-0	SDIO-25	SDIO-50	SDIO-100	SDIO-200	IO-100
SAR (W g <sup>-1</sup> )	334.8	368.3	493.8	1235.4	664.5	1004.4