

*Electronic Supplementary Information*

**An NIR-Sensitive Layered Supramolecular Nanovehicle for  
Combined Dual-Modal Imaging and Synergistic Therapy**

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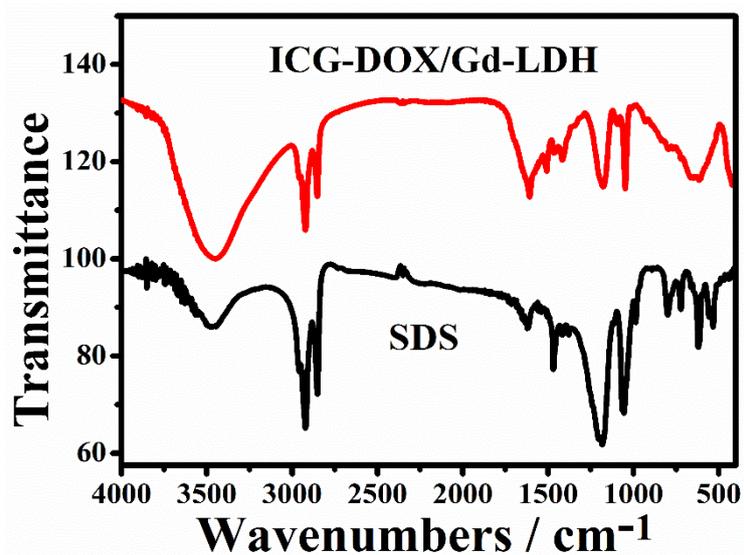
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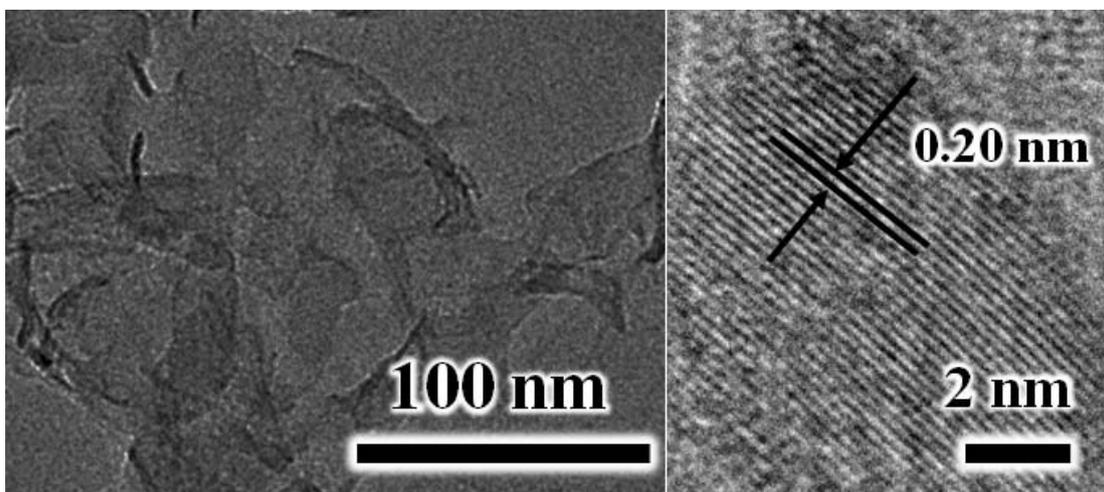
liangruizheng2000@163.com (R.L.)

**Table S1.** Chemical compositions of various composites.

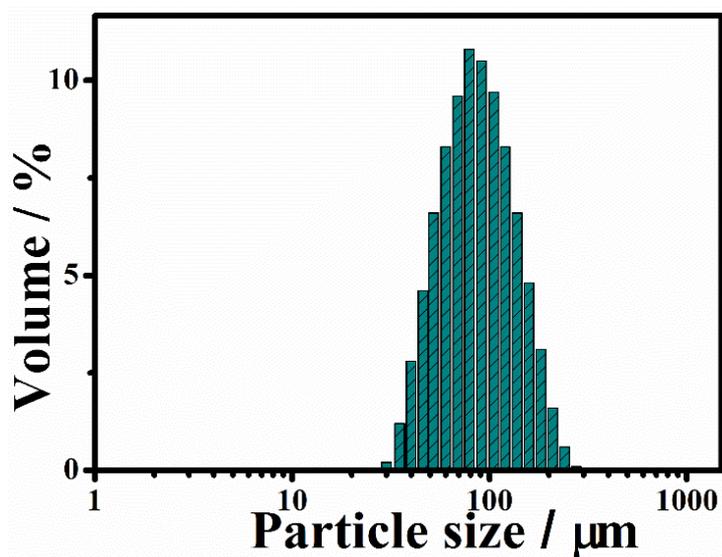
Sample	Chemical Composition	Mg <sup>2+</sup> /Al <sup>3+</sup>
Gd-LDH	Mg <sub>0.581</sub> Al <sub>0.366</sub> Gd <sub>0.053</sub> (OH) <sub>2</sub>	1.59
DOX/Gd-LDH	Mg <sub>0.602</sub> Al <sub>0.356</sub> Gd <sub>0.032</sub> (OH) <sub>2</sub> DOX <sub>0.00905</sub>	1.69
ICG/Gd-LDH	Mg <sub>0.674</sub> Al <sub>0.312</sub> Gd <sub>0.029</sub> (OH) <sub>2</sub> ICG <sub>0.024</sub> SDS <sub>0.0127</sub>	2.16
ICG-DOX/Gd-LDH	Mg <sub>0.641</sub> Al <sub>0.312</sub> Gd <sub>0.025</sub> (OH) <sub>2</sub> ICG <sub>0.0192</sub> DOX <sub>0.0153</sub> SDS <sub>0.0104</sub>	2.05



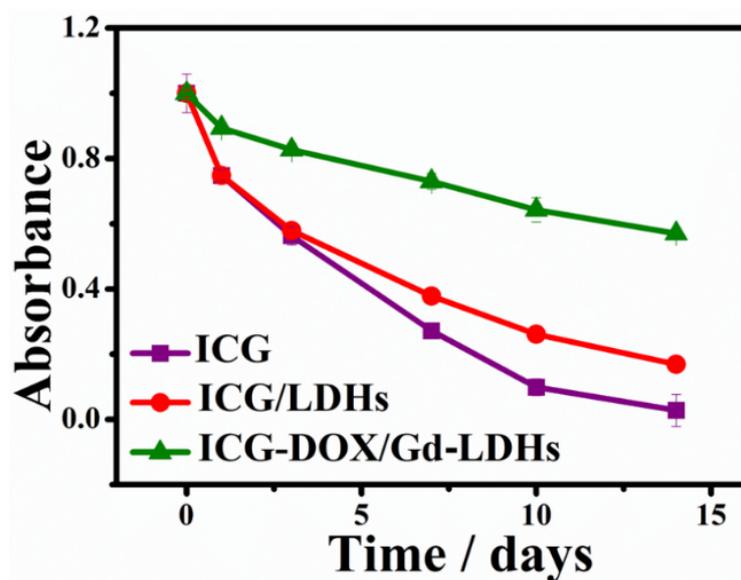
**Figure S1.** FTIR spectra of SDS and ICG-DOX/Gd-LDH, respectively.



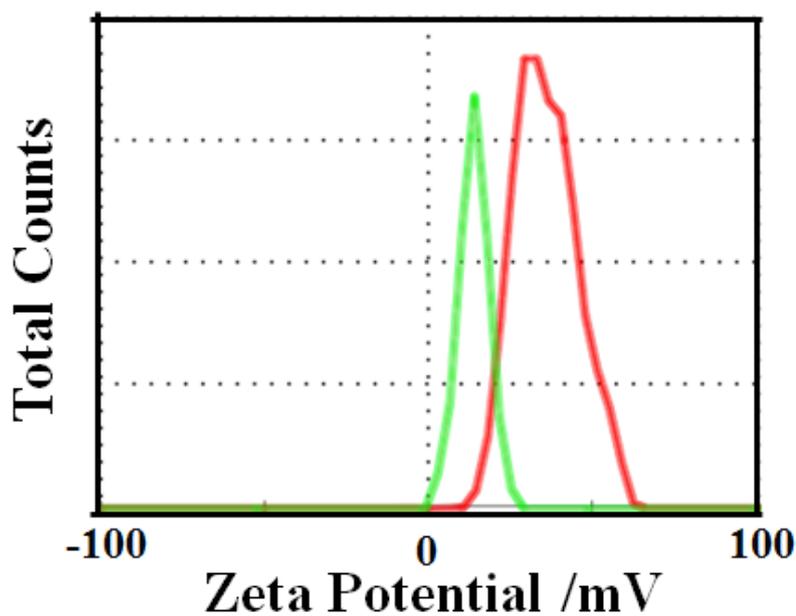
**Figure S2.** HRTEM images and lattice of the ICG-DOX/Gd-LDH.



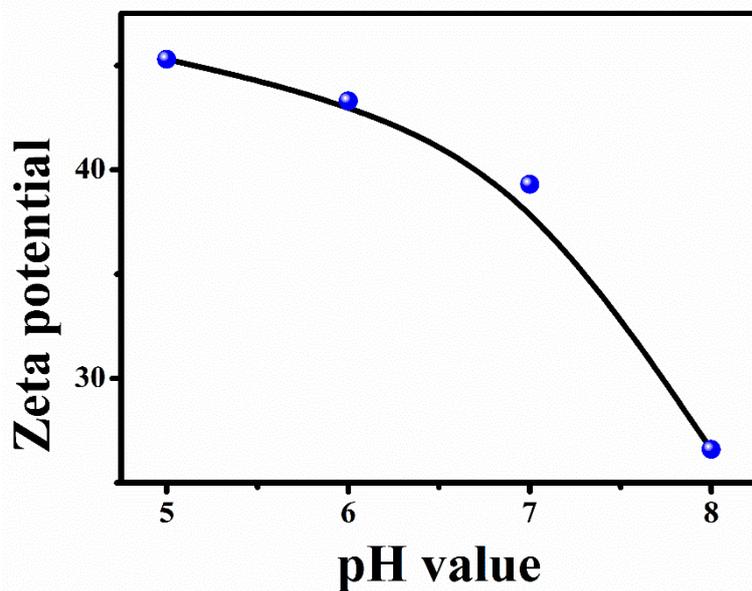
**Figure S3.** Particle size distribution of ICG-DOX/Gd-LDH determined by dynamic light scattering analyzer (DLS).



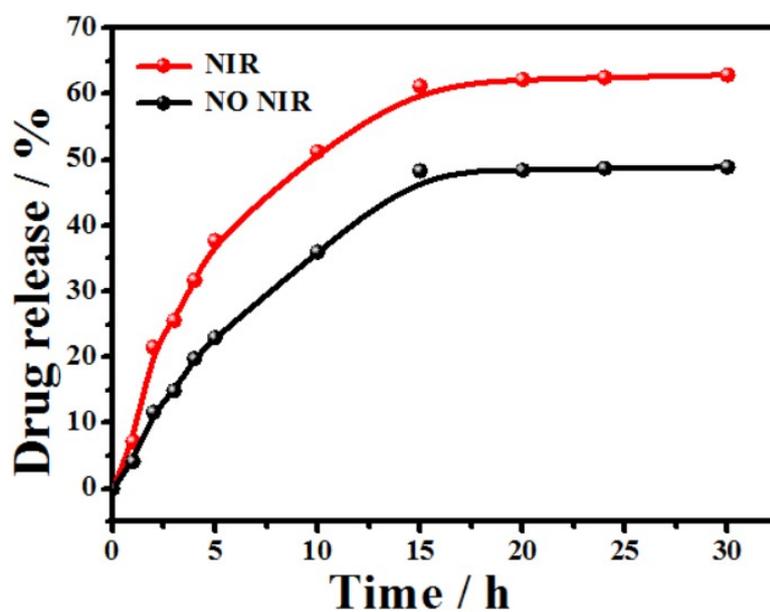
**Figure S4.** Stability test of ICG, ICG/Gd-LDH and ICG-DOX/Gd-LDH under the room temperature for 15 days.



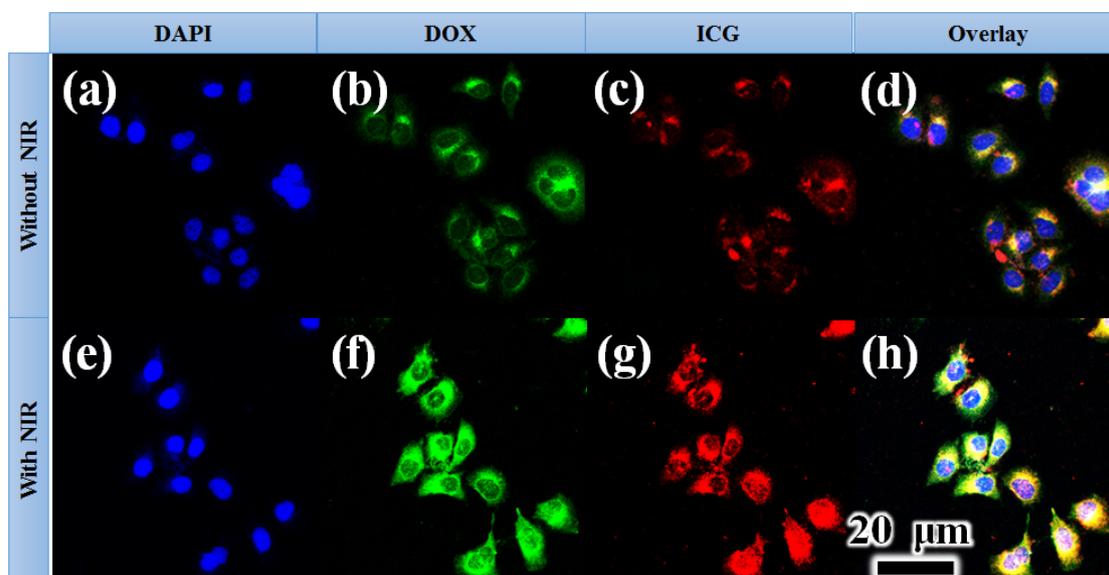
**Figure S5.** Zeta potential of ICG/Gd-LDH and ICG-DOX/Gd-LDH, respectively.



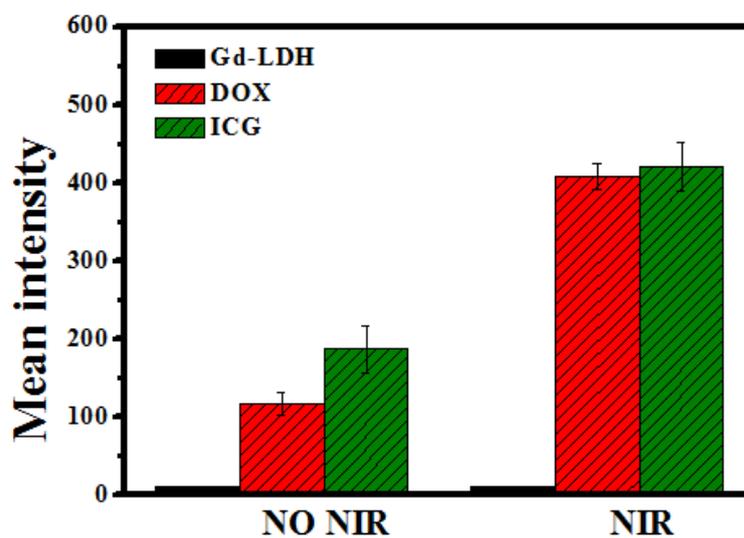
**Figure S6.** Zeta potential of ICG-DOX/Gd-LDH at various pH values.



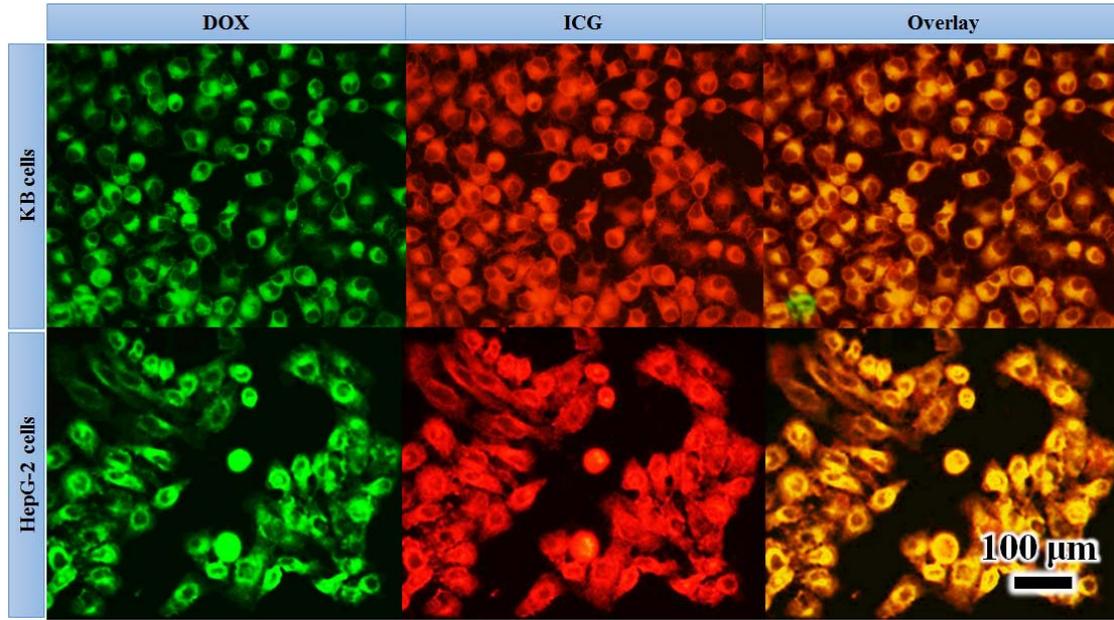
**Figure S7.** Cumulative DOX release from ICG-DOX/Gd-LDH under a simulant physiological condition (PBS buffer solution, pH=7.4) with and without NIR irradiation.



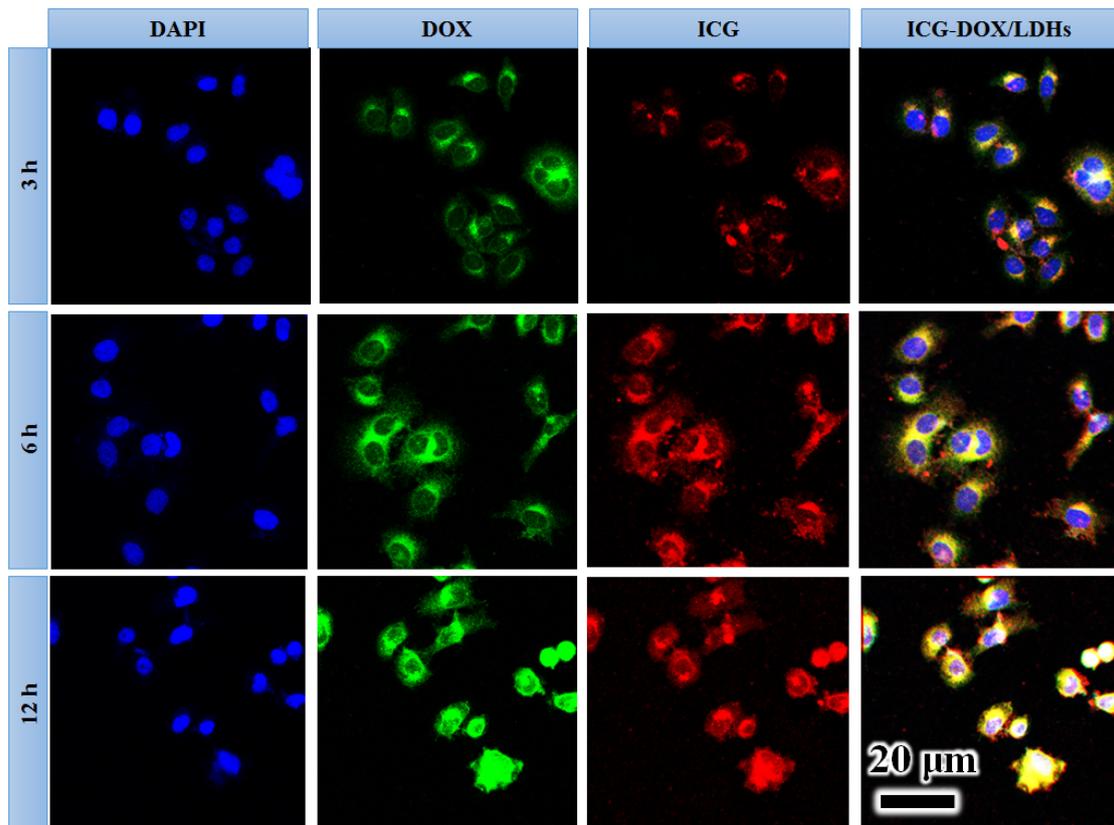
**Figure S8.** The confocal imaging photographs of ICG-DOX/Gd-LDH incubated with HeLa cells for 3h with and without NIR irradiation.



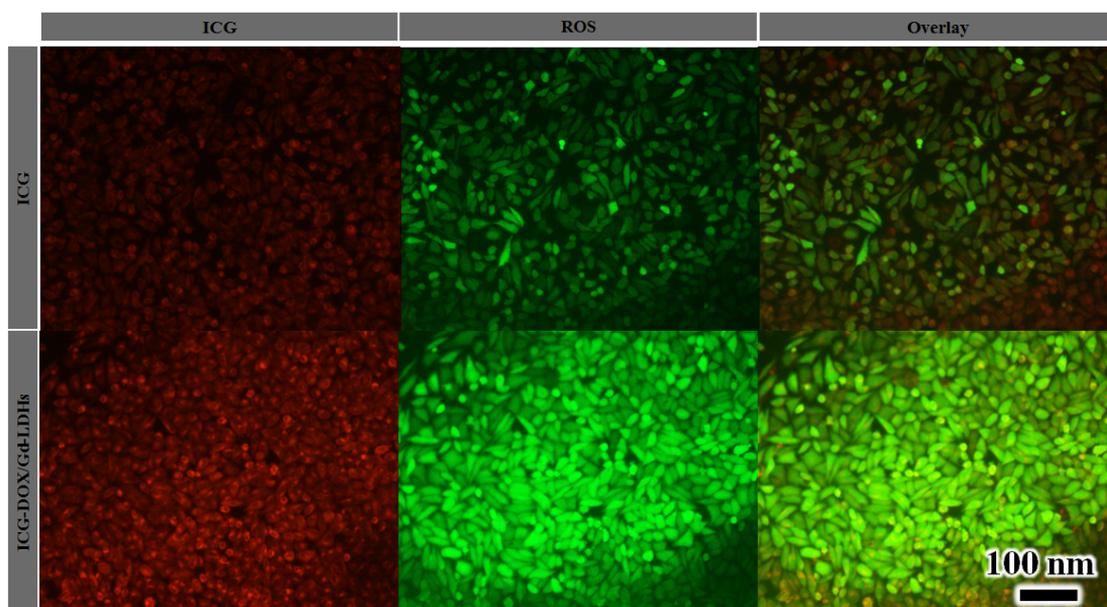
**Figure S9.** Fluorescence intensity of Gd-LDH, and ICG-DOX/Gd-LDH under dark and irradiation.



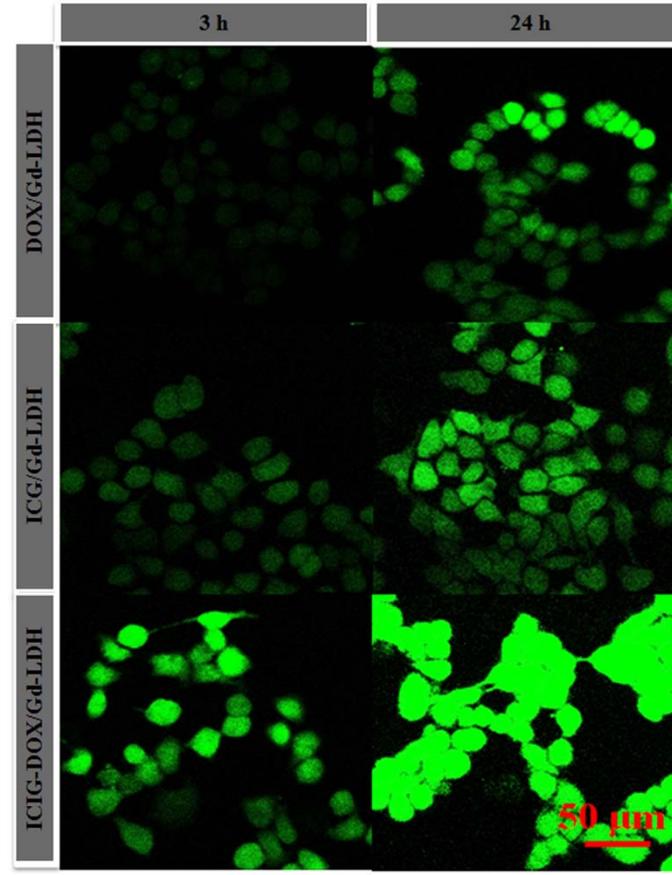
**Figure S10.** Fluorescence imaging of KB cells and HepG-2 cells incubated with ICG-DOX/Gd-LDHs, respectively.



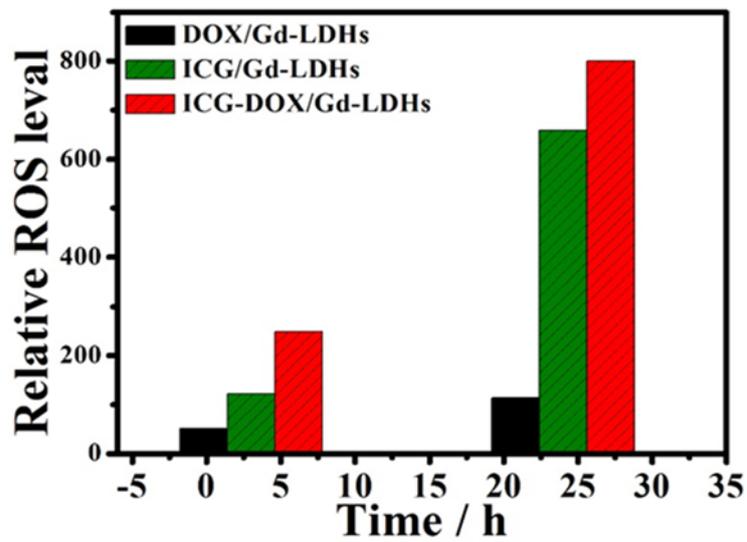
**Figure S11.** Confocal fluorescence images of HeLa cells incubated ICG-DOX/Gd-LDH for 3h, 6h and 12 h, respectively.



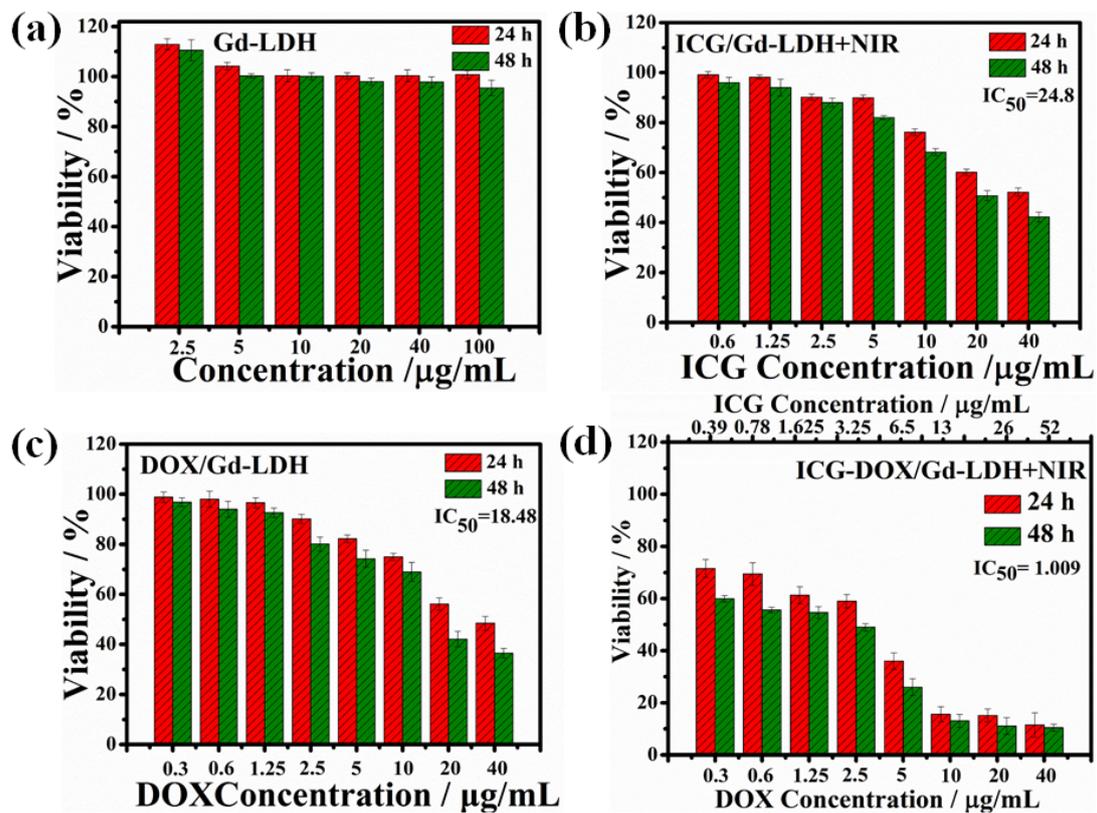
**Figure S12.** ROS images inside HeLa cells with the incubation with ICG and ICG-DOX/Gd-LDH after the NIR irradiation for 10 mins.



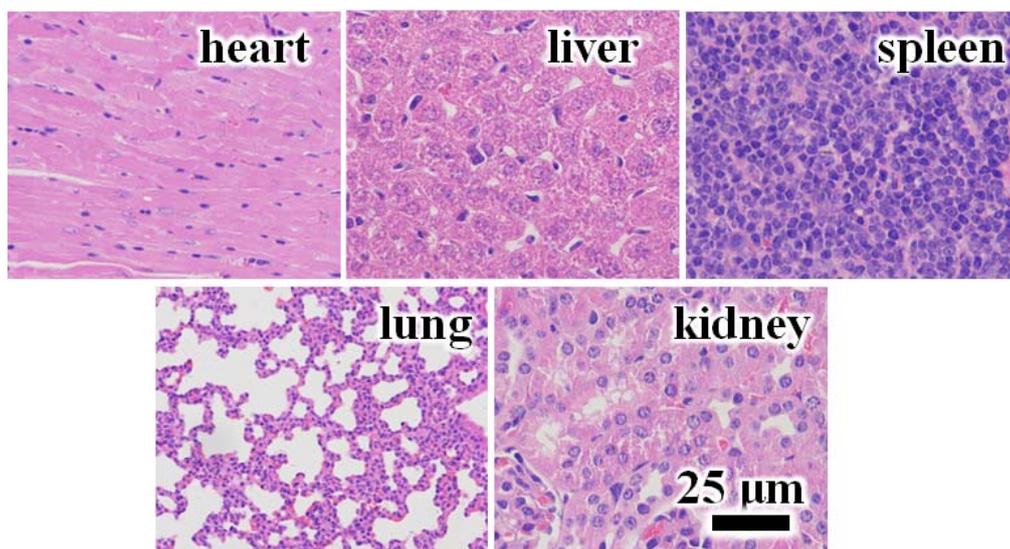
**Figure S13.** Time series of ROS images inside HeLa cells after treatment with various samples under NIR irradiation therapy.



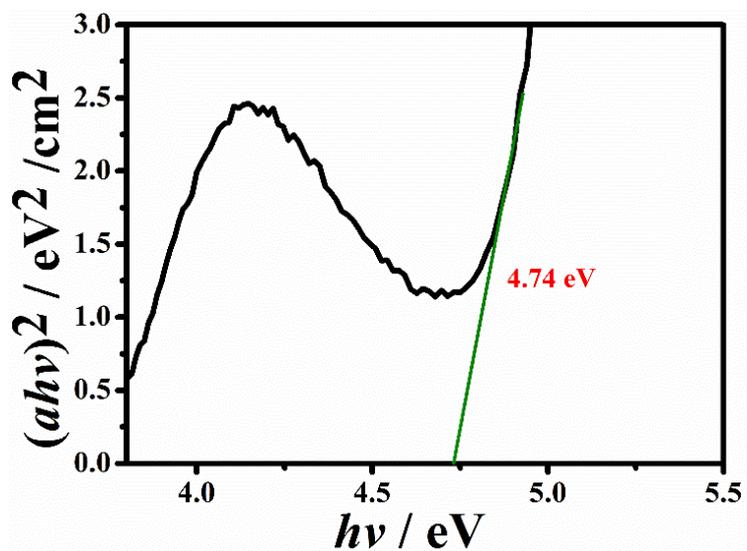
**Figure S14.** Time series of changes in the related ROS levels after treatment with various samples under NIR irradiation therapy.



**Figure S15.** (a) Viability of Gd-LDH incubated with Hela cell for 24 h and 48 h. (b) The viability of ICG/Gd-LDH (b) DOX/Gd-LDH (c) and ICG-DOX/Gd-LDH under dark and irradiation. The laser group were irradiated at 808 nm with the power of 1 W/cm<sup>2</sup>.



**Figure S16.** H&E-stained organs isolated from tumor bearing mice treated by ICG-DOX/Gd-LDH formulation.



**Figure S17.** UV-vis diffuse reflectance spectra of Gd-LDH.