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Supporting Information

PEGylated Reduced-Graphene Oxide Hybridized with Fe₃O₄ Nanoparticles for Cancer Photothermal- Immunotherapy

Lu Wang,^{a,b} Meng Wang,^{b,c} Benqing Zhou,^{b,c} Feifan Zhou,^{c,b} Cynthia Murray,^b Rheal
A. Towner,^d Nataliya Smith,^d Debra Saunders,^d Gang Xie^a and Wei R. Chen^{b*}

^a Key Laboratory of Synthetic and Natural Functional Molecule Chemistry of
Ministry of Education, College of Chemistry & Materials Science, Northwest
University, Xi'an, Shaanxi 710127, P. R. China

^b Biophotonics Research Laboratory, Center for Interdisciplinary Biomedical
Education and Research, College of Mathematics and Science, University of Central
Oklahoma, Edmond, Oklahoma 73034, United States

^c Key Laboratory of Optoelectronic Devices and Systems of Ministry of Education
and Guangdong Province, College of Optoelectronic Engineering, Shenzhen
University, Shenzhen, Guangdong 518060, P. R. China

^d Advanced Magnetic Resonance Center, Oklahoma Medical Research Foundation,
Oklahoma City, Oklahoma 73104, United States

* Corresponding Author: Wei R. Chen

Email: wchen@uco.edu

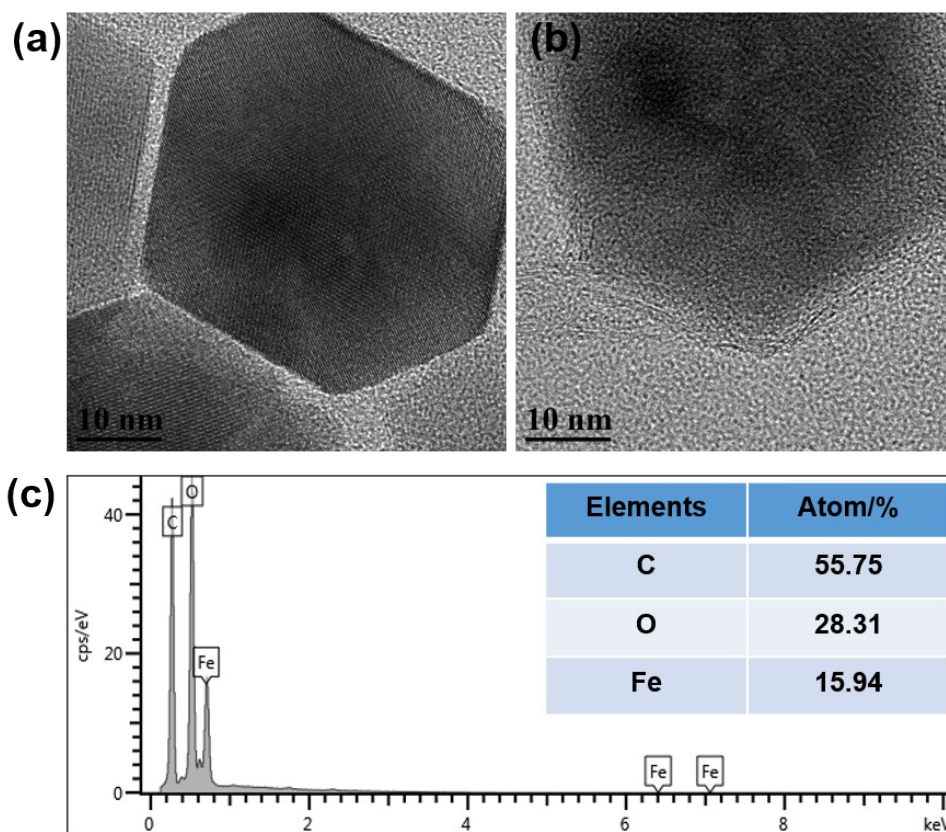


Fig. S1. Morphology and composition characterization of FNPs/rGO-PEG. High Resolution Transmission Electron Microscope (HRTEM) images of (a) FNPs and (b) FNPs/rGO-PEG. Scale bar = 10 nm. (c) Energy dispersive X-ray spectrum (EDS) analysis of FNPs/rGO-PEG, which reveals the existence of Fe, O and C elements in FNPs/rGO-PEG.

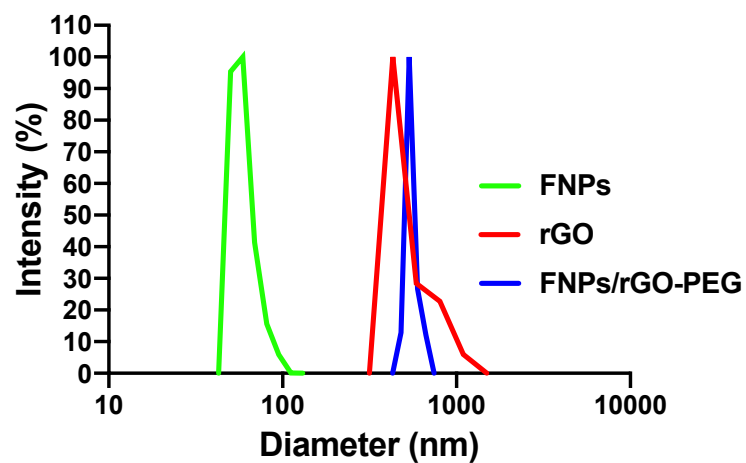


Fig. S2. Hydrodynamic diameter distribution of the FNPs, rGO-PEG and FNPs/rGO-PEG. The hydrodynamic diameter of the FNPs (58.96 nm) is in agreement with their TEM images. The hydrodynamic diameter of FNPs/rGO-PEG increased to 534.87 nm, which similar with the hydrodynamic diameter of rGO, indicated that the FNPs were successfully encapsulated in rGO-PEG.

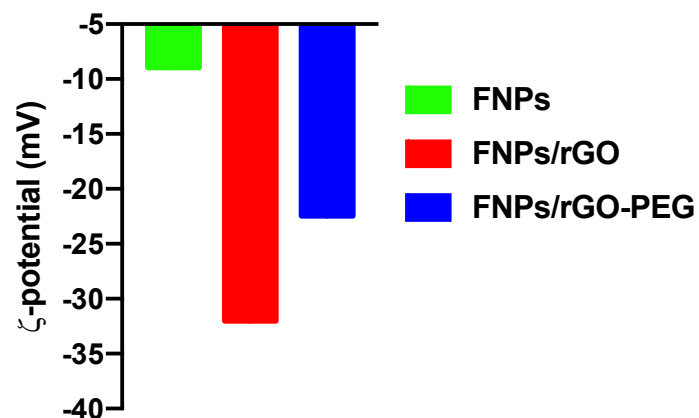


Fig. S3. Zeta-potentials of FNP, FNP/rGO and FNP/rGO-PEG obtained from the DLS measurement. After the encapsulation of the rGO, the zeta-potential of the FNP changed from -8.96 mV to -32.028 mV (pH 7.4), which verifying the successful conjugation of rGO to the surface of FNP.

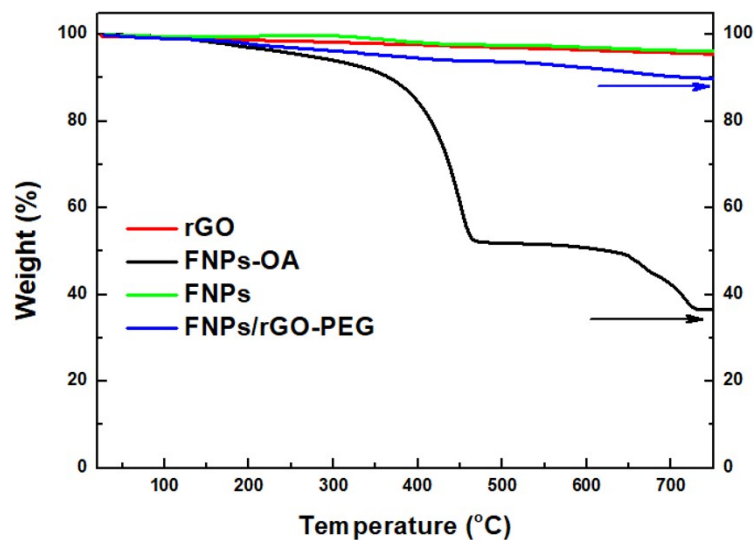


Fig. S4. The oleate-capped FNPs display a large weight loss of 62.83% with a decomposition temperature of 794 °C. For comparison, the FNPs show a much smaller weight loss of 3.78%, thus re-confirming the successful removal of oleate ligand from the surface of the FNPs. In comparison with FNPs and rGO, the FNPs/rGO-PEG show a much larger weight loss of 10.47%, thus re-confirming the successful modification of the PEG-NH₂ on the surface of FNPs/rGO.

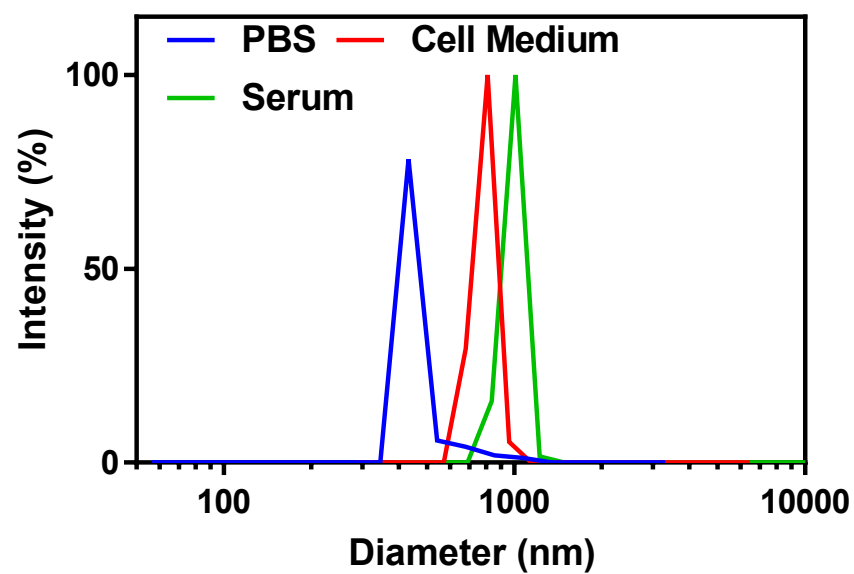


Fig. S5. Hydrodynamic diameter distribution of FNP/rGO-PEG in PBS, Cell Medium and Serum.

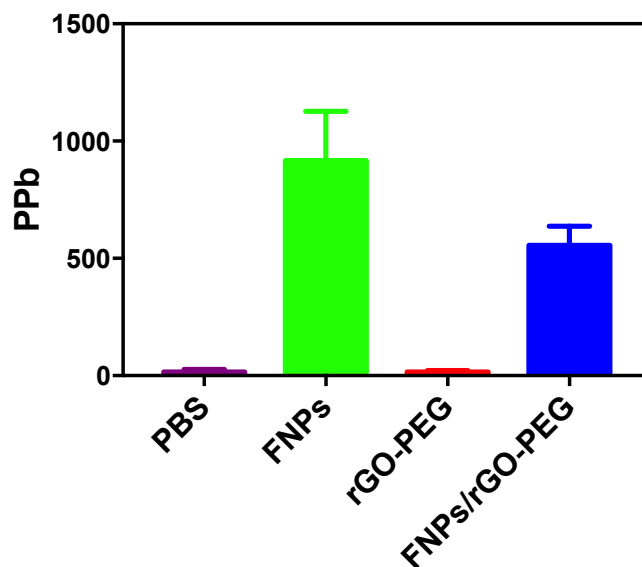


Fig. S6. Fe contents in 4T1 tumor cells after treatment with PBS, FNP, rGO-PEG and FNP/rGO-PEG. (n = 4). 4T1 tumor cells were incubated with PBS, FNP (500 $\mu\text{g mL}^{-1}$), rGO-PEG (40 $\mu\text{g mL}^{-1}$) and FNP/rGO-PEG (500 $\mu\text{g mL}^{-1}$) for 4 h. After 4 h incubation, the cells were collected and washed twice with PBS. Then the different cell samples were analyzed via Inductive Coupled Plasma Emission Spectrometer (ICP). Data are presented as the mean $\pm$ S.E.M.

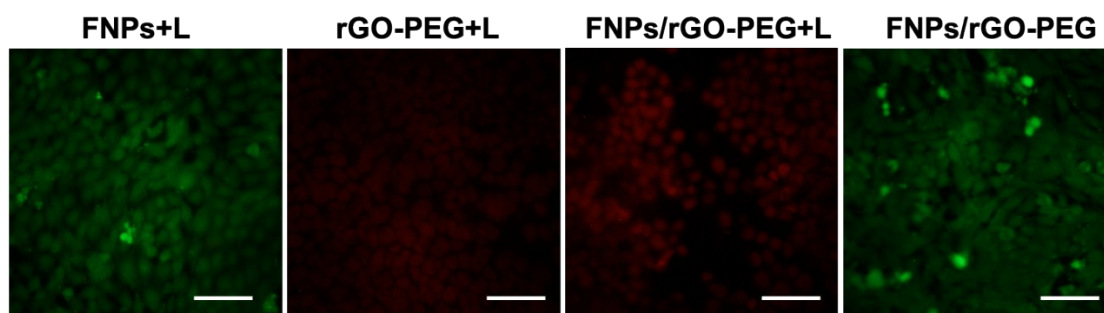


Fig. S7. Fluorescence images of 4T1 tumor cells incubated with FNPs, rGO-PEG and FNPs/rGO-PEG after irradiation by the 805-nm laser (1 W cm^{-2}) for 5 min. The cells were co-stained by calcein AM (green, live cells) and propidium iodide (red, dead cells) before imaging. Scale bar = $100 \text{ }\mu\text{m}$.

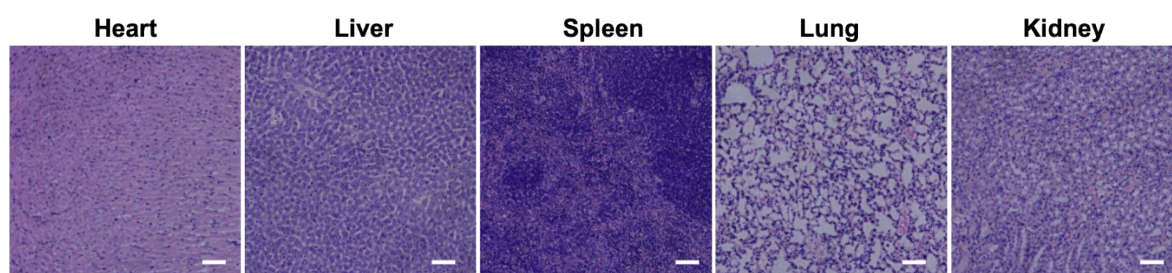


Fig. S8. Representative H&E-stained images of major organs from the FNP/rGO-PEG treated mice. Scale bar = 100 μm .

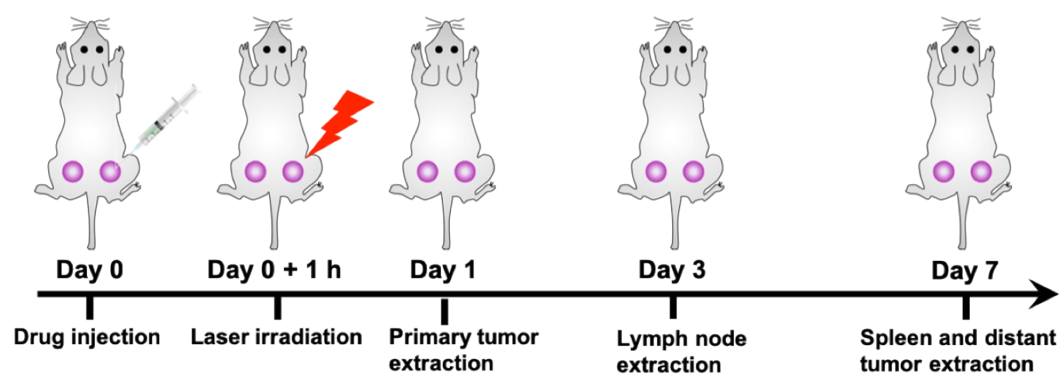


Fig. S9. Schematic depiction of tumor-specific abscopal effect.

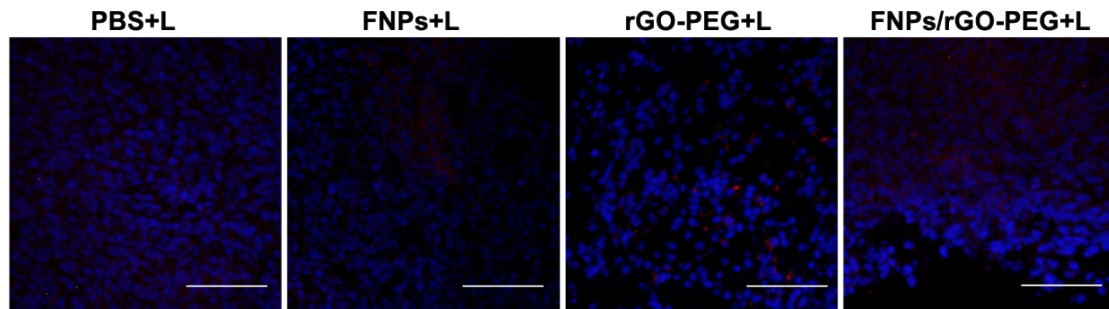


Fig. S10. Histochemical images of tumor cells collected 24 h after different treatments. Immunofluorescence of the tumors showed infiltrated CD11c⁺ DC cells. Scar bar = 200 μ m.

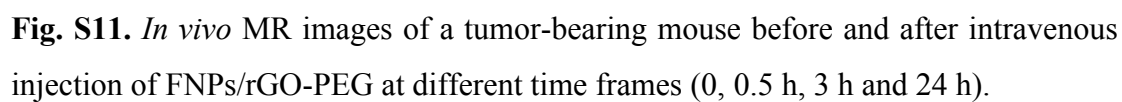


Fig. S11. *In vivo* MR images of a tumor-bearing mouse before and after intravenous injection of FNPs/rGO-PEG at different time frames (0, 0.5 h, 3 h and 24 h).

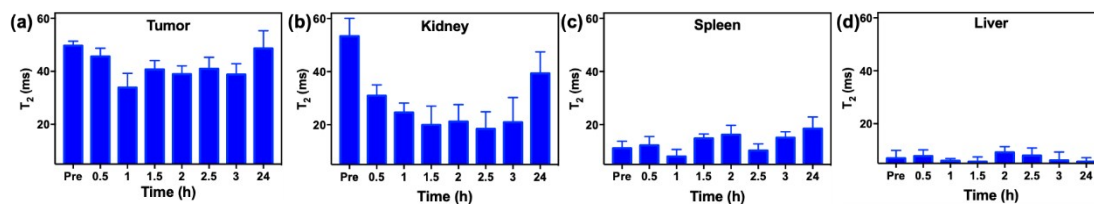


Fig. S12. Corresponding representative T2*-weighted signals of tumor (a), kidney (b), spleen (c) and liver (d) at different time points after intravenous injection of FNP/rGO-PEG.