Fast and Facile Preparation of PEGylation Graphene from Graphene Oxide by Lysosome Targeting Delivery of Photosensitizer to Efficiently Enhance Photodynamic Therapy

Yiping Zeng,^{a1} Zhangyou Yang,^{a1} Shenglin Luo,^a Hong Li,^a Cong Liu,^a Yuhui

Hao,^a Jing Liu,^a Weidong Wang^{b,*} and Rong Li^{a,*}

 ^a State Key Laboratory of Trauma Burns and Combined Injury, Institute of Combined Injury, Chongqing Engineering Research Center for Nanomedicine, College of Preventive Medicine, Third Military Medical University, Chongqing 400038, China
^b Department of Radiation Oncology, Shanghai Sixth People Hospital, Shanghai Jiao Tong University, Shanghai 200233, China

E-mail: https://www.icea.com and www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com and www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.icea.com"/www.icea.com"/www.icea.com and <a href="https://www.ic

Supplementary information



Figure S1. The XRD spectrum of natural graphite and GO



Figure S2. (a) The photos of NGO-PEG samples prepared with small molecules after centrifugation at 7000 r/min for 3 min. (b) The photos of NGO-PEG samples prepared with different chain length after centrifugation at 7000 r/min for 3 min. (c) The photos of NGO-PEG samples prepared with different ratio of GO and KOH (w/w). (d) The photos of NGO-PEG samples prepared with different temperature.



Figure S3. The ¹H NMR spectra of (a) BPEI, (b) NGO-PEG, (c) NGO-PEG-BPEI.



Figure S4. The standard curve of measured the absorbance of cuparammonium complexes formed between BPEI and copper ion (II) at 630 nm at a series of PEI concentrations.



Figure S5. Zeta potentials of NGO-PEG, NGO-PEG-BPEI, NGO-PEG-Ce6 and NGO-PEG-BPEI-Ce6.



Figure S6. Fluorescence emission spectra of free Ce6 (a), NGO-PEG-Ce6 (b) and NGO-PEG-BPEI-Ce6 (c) in singlet oxygen sensor green (SOSG) solution at different laser irradiation time points.



Figure S7. Relative cell viability data obtained from the CCK-8 assay of HeLa cells after treated with NGO-PEG (a) and NGO-PEG-BPEI (b) without/with light irradiation by 662 nm laser, (c) Relative cell viability data obtained from the CCK-8 assay of HeLa cells after treated with Ce6, NGO-PEG-Ce6 and NGO-PEG-BPEI-Ce6 without light irradiation, the cell viability values were all normalized to control untreated cells. (d) Fluorescence image of Calcein AM /propidium iodide stained HeLa cells incubated with PBS (control group). Error bars were based on SD of triplicate parallel samples.



Figure S8. Fluorescent image (a, b) and phase contrast overlapped image (e, f) of Hela cells incubated with NGO-PEG-Ce6 (2 μ M Ce6) at 4°C for 2 h. Fluorescence image (c, d) and phase contrast overlapped image (g, h) of HeLa cells incubated with NGO-PEG-BPEI-Ce6 (2 μ M Ce6) at 37 °C for 2 h.



Figure S9. Localization image of Ce6, NGO-PEG-Ce6 and NGO-PEG-BPEI-Ce6 stained HeLa cells, incubated with 2 μ M free Ce6 or equivalent amount of NGO-PEG-Ce6 and NGO-PEG-BPEI-Ce6 (Mitotracker stained mitochondria).



Figure S10. Fluorescence image of determination ROS producing ability by DCFH-DA method with fluorescence microscope incubated with PBS (a) $1 \mu M$ free Ce6 (d, e), equivalent amount of NGO-PEG-Ce6 (b) and NGO-PEG-BPEI-Ce6 (c) for 24 h without irradiation



Figure S11. The magnification Fluorescence image of determination ROS producing ability of the cell treated with NGO-PEG-PEI-Ce6