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White-Light-Emitting Magnetite Nanoparticle-Polymer Composites: Photonic Reactions of Magnetic Multi-Granule Nanoclusters as Photothermal Agents

Yu Jin Kim,^{‡a} Bum Chul Park,^{‡b} June Park,^c Hee-Dae Kim,^d Nam Hoon Kim,^e Yung Doug Suh^{*e} and Young Keun Kim^{*b}

^aCenter for Creative Materials and Components, Korea University, 145 Anam-ro, Seongbuk-gu, Seoul 02841, Korea.

^bDepartment of Materials Science and Engineering, Korea University, 145 Anam-ro, Seongbuk-gu, Seoul 02841, Korea. E-mail: ykim97@korea.ac.kr; Fax: +82-2-928-3584; Tel: +82-2-3290-3281

^cUltra-Precision Optics Research Sector, Korea Photonics Technology Institute, Gwangju, 61007, Korea.

^dDepartment of Biological Sciences, Seoul National University, Seoul 08826, Korea.

^eResearch Center for Convergence NanoRaman Technology, Korea Research Institute of Chemical Technology, Yuseong, P.O. Box 107, Daejeon 34114, Korea. E-mail: ydsuh@kriect.re.kr; Fax: +82-42-860-7625; Tel: +82-42-860-7597

* Y.K.K and Y.D.S developed and supervised the project.

‡These authors made equal contributions to this study.

S1. Sample preparation

Au nanoparticles (NP) with a diameter of 100 nm were synthesized *via* a citrate method. Magnetic nanoclusters with a diameter of 100 nm were synthesized *via* a hydrolysis condensation and reductive polyol process. A known volume of poly(methyl methacrylate) (PMMA) solution (4 wt.% of PMMA dissolved in chlorobenzene) was added to prepare the Au NPs and the magnetic nanoclusters. We prepared a magnetic nanocluster-PMMA composite solution with different concentrations from 0.01 to 1 mg mL⁻¹. The mixtures were pipetted onto a cover glass, which can be used for optical measurements. Then, the samples were dried overnight in an oven.

S2. Optical measurements and analysis of the MGNC–polymer composites

Image acquisition and confocal fluorescence microscopy analysis

All images were acquired using a LSM 710 NLO confocal microscopy system (Carl Zeiss). An Ar laser (488 nm), a He-Ne laser (543 nm), and a Mai-Tai HP femtosecond laser (690–1040 nm, Spectra-Physics, USA) were used in this study. To quantify the obtained fluorescence signals, fluorescence intensity was measured in the regions of interest (ROI) using a ZEN2010 image analysis system (Carl Zeiss).

Irradiation of the MGNC-PMMA composite by confocal microscopy using a 780 nm multiphoton laser

The MGNC-PMMA composites were exposed to a multiphoton laser from Mai Tai HP. The multiphoton laser has a $\lambda = 780$ nm, repetition rate of 80 MHz, pulse duration of 100 fs, and a 1.2 mm beam size. The samples were exposed to pulse energies ranging from 0.375 to 37.5 nJ. Each spot of the MGNC-PMMA composite was irradiated with the focused beam for 3 μ s. The entire irradiation process was conducted using the focused beam spot by spot. The power of the multiphoton laser was controlled between 30 and 300 mW by measuring it with a power meter to ensure a consistent irradiation of the MGNC-PMMA composite.

Photoluminescence analysis

Photoluminescence spectra were measured at room temperature by using a 325-nm line source of a He-Cd laser. The laser power was maintained below 1 mW. The scattered light was first collected by a 15× objective lens, then dispersed with a 600 grooves mm^{-1} grating, and finally analyzed using a liquid N_2 -cooled Si CCD detector with a single-grating monochromator (LabRam HR800, Jobin Yvon Inc., France) having a focal length of 80 cm. The spectra were acquired in the 400–800 nm range.

S3. Results of photonic reactions for various particles at different 780 nm multiphoton laser powers

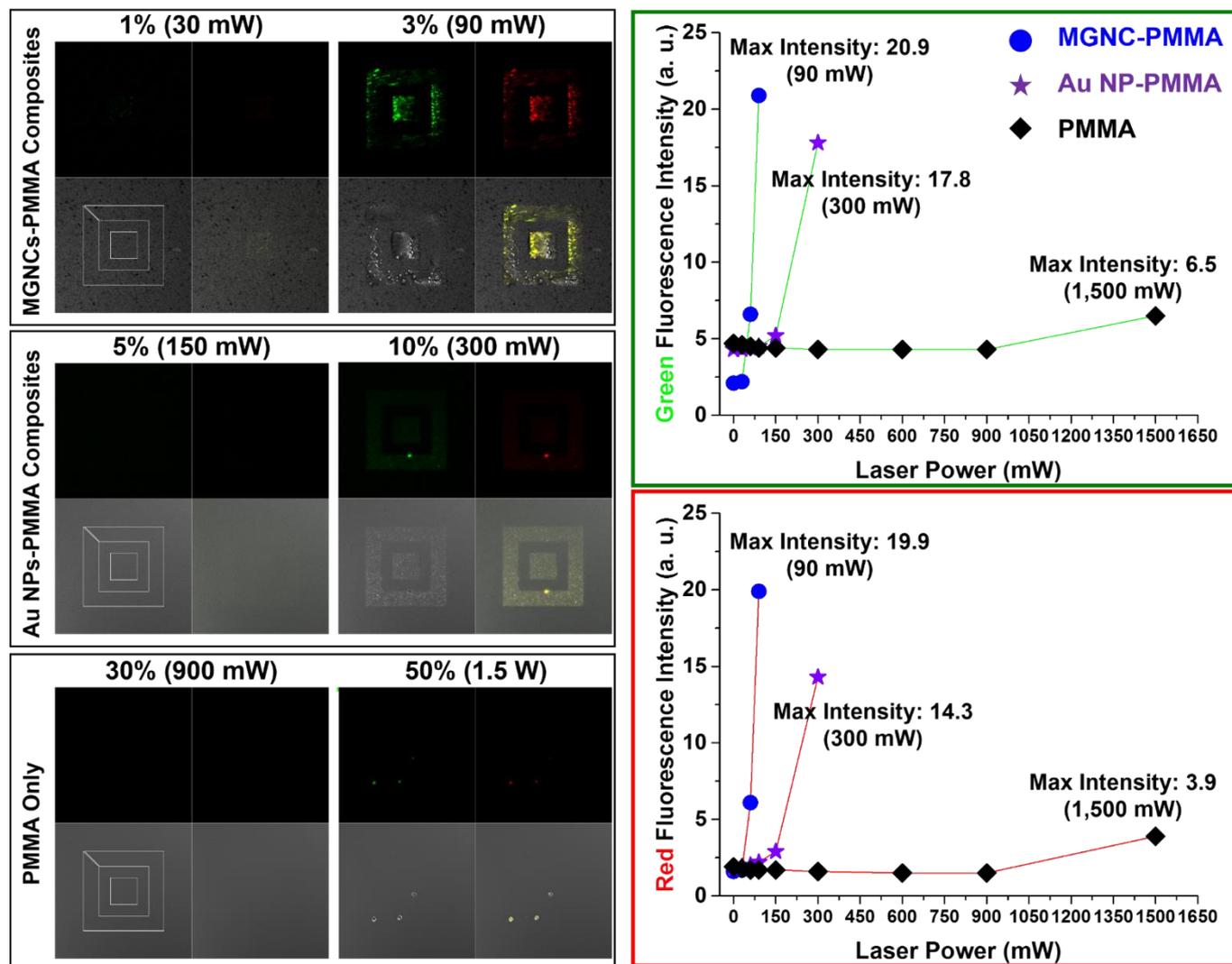


Fig. S1. Emission intensities for various particle types at different 780 nm multiphoton laser power values.

S4. π -conjugated polymer transition

Besides their conductivity, π -conjugated polymers are also characterized by absorption and emission characteristics related to visible light wavelengths. PMMA polymers can exhibit backbone chain transitions to π -conjugated polymers through thermal degradation and oxidation.

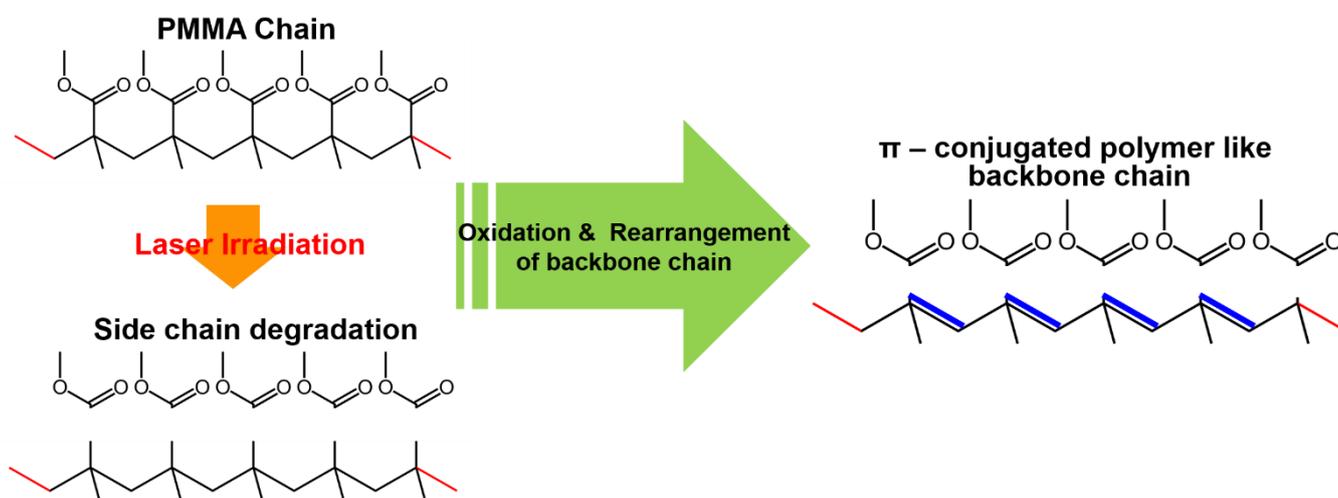


Fig. S2. Schematic representation of the PMMA polymer thermal degradation and oxidation by 780 nm multiphoton laser irradiation.

S5 CIE chromaticity diagram from the PL spectra

The corresponding CIE coordinates calculated with the PL spectrum are presented in Figure S3. It is worth noting that irradiated MGNC-PMMA composites exhibits the CIE coordinate of (0.293410132, 0.377475905). Even though slightly shifted to range of green color, it is close to the pure white light (0.33, 0.33) of the 1931 CIE. Likewise, the RGB merged image for irradiated MGNC-PMMA composites show the nearly white light emission.

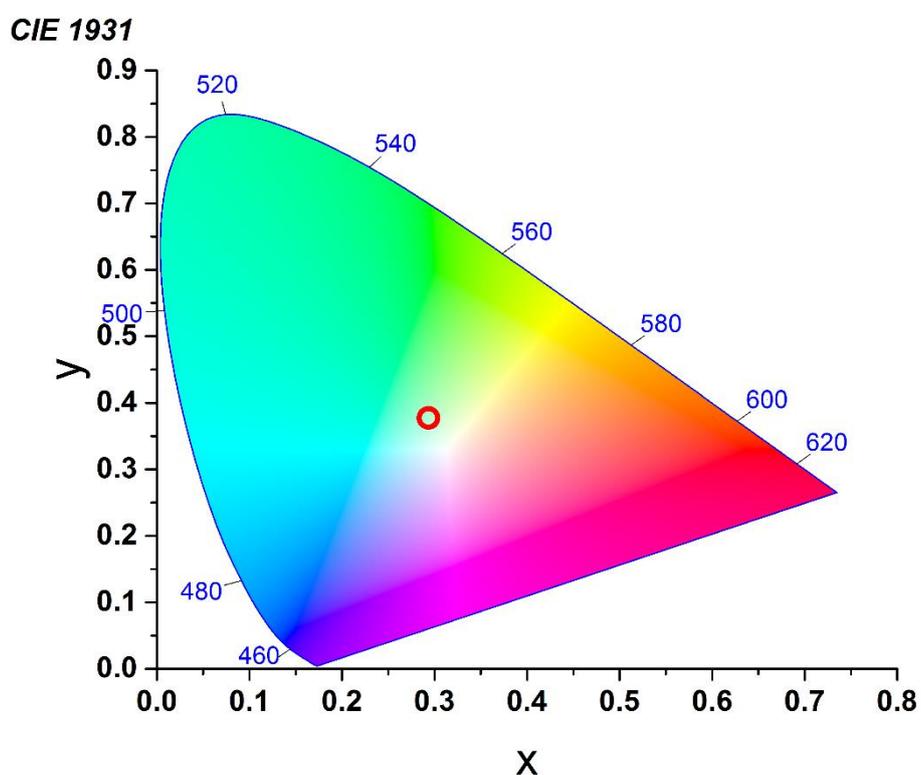


Fig. S3. Comparison of calculated CIE chromaticity coordinates corresponding to the PL spectrum of irradiated MGNC-PMMA composites at the excitation wavelength of 325 nm light source. (red circle)

Fig. S4 show the red, green and blue (RGB) fluorescence images of MGNC-PMMA composites. These confocal fluorescence microscope images measured after 3 times consecutive irradiation of multiphoton laser by tile scan mode. The merged image of RGB fluorescence represent slightly yellowish white light emission, relatively corresponding with CIE chromaticity.

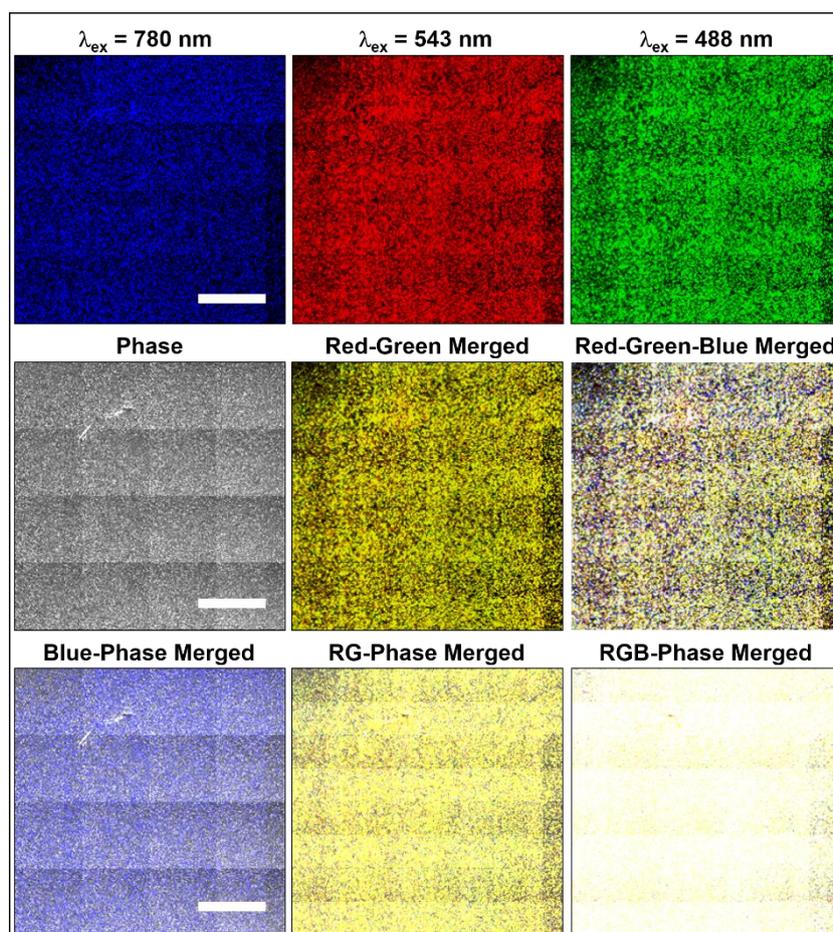


Fig. S4. RGB fluorescence images of MGNC-PMMA composites after laser irradiation. (scale bar: 400 μm)