Electronic Supplementary Material (ESI) for Nanoscale.
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# Electronic Supplementary Information (ESI)

# Multicatalytic Colloids with Highly Scalable, Adjustable, and Stable Functionalities in Organic and Aqueous Media

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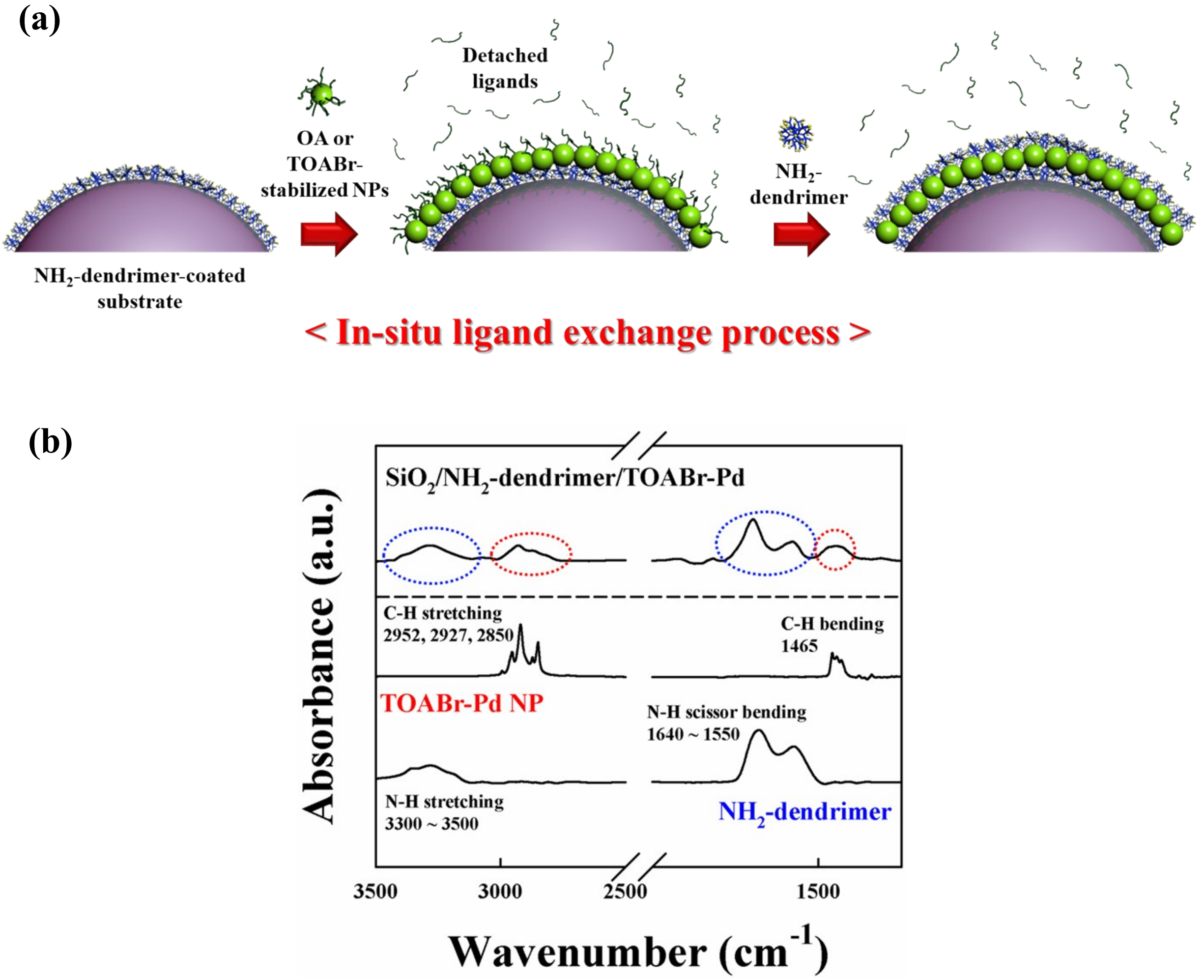
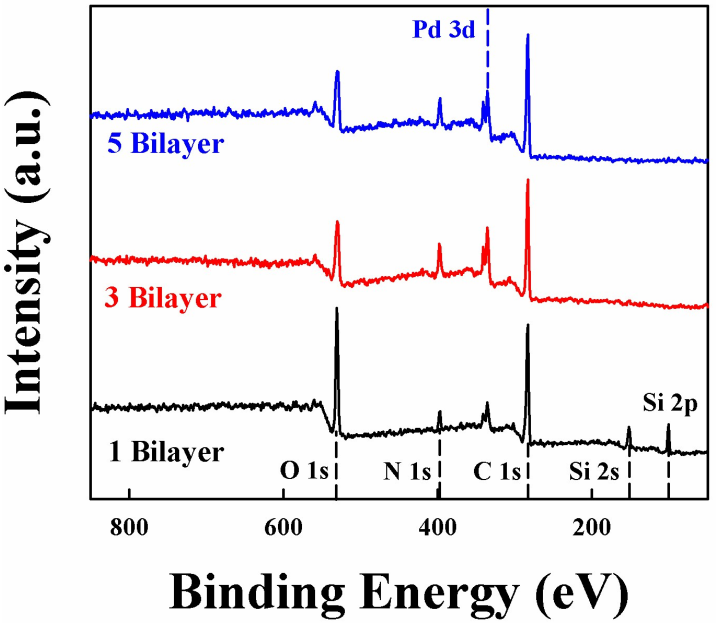


**Figure S1.** (a) Scheme of the *in-situ* ligand exchange between $NH_2$-dendrimers and TOABr (or OA)-stabilized metal (or metal oxide) NPs for the fabrication of multilayers. (b)ATR-FTIR spectra of pristine $NH_2$-dendrimers, TOABr-Pd NPs, and $SiO_2$/$NH_2$-dendrimer/TOABr-Pd NPs.



| | C 1s (283 eV) (%) | Pd 3d (336 eV) (%) | N 1s (397 eV) (%) | O 1s (531 eV) (%) |
|---|---|---|---|---|
| 1 bilayer | 59.79 | 1.27 | 6.08 | 32.87 |
| 3 bilayer | 60.91 | 2.46 | 9.89 | 26.73 |
| 5 bilayer | 62.07 | 2.53 | 10.45 | 24.95 |

**Figure S2.** XPS spectra and elemental composition of the ($NH_2$-dendrimer/TOABr-Pd NP)$_{n=1,3,\text{ and }5}$ multillayer-coated colloids.

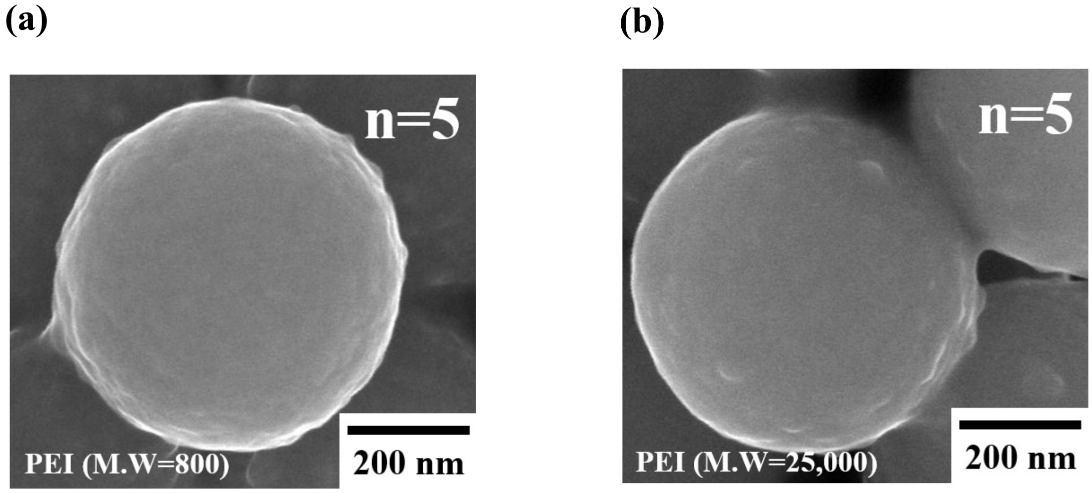


**Figure S3.** SEM images of (PEI/TOABr-Pd NP)$_5$ multilayer-coated colloids prepared from two different $M_w$ of PEI: $M_w$ of PEI = (a) 800 and (b) 25,000.

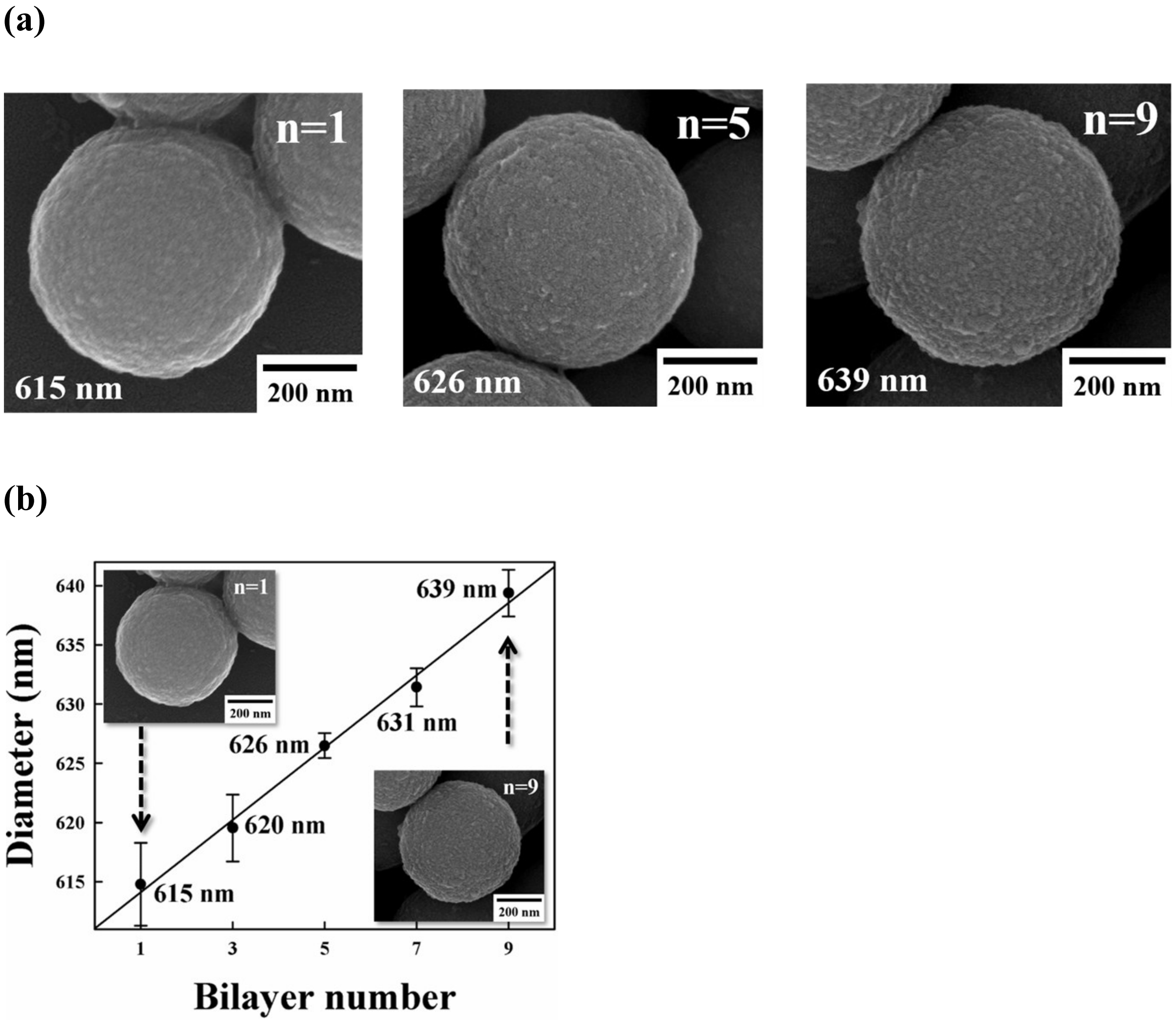


**Figure S4.** (a) SEM images of (cationic Pd NP/anionic polyelectrolyte)$_n$ multilayer-coated colloids with increasing bilayer number (n). (b) Size change of (cationic Pd NP/anionic polyelectrolyte)$_n$ multilayer-coated $SiO_2$ colloids with increasing bilayer number (n). Cationic Pd NPs in aqueous solution were prepared using the phase-transfer method of TOABr-Pd NPs synthesized in toluene. For the phase transfer of TOABr-Pd NPs from toluene to water, an aqueous 0.1 M 4-dimethyl-aminopyridine (DMAP) solution (80 mL) was added to the toluene solution (80 mL) containing TOABr-stabilized Pd NPs. After three hours, the Pd NPs dispersed in toluene were completely phase-transferred to the aqueous phase, and the separated toluene phase was removed. The resulting cationic Pd NPs dispersed in the aqueous phase were stabilized by the DMAP ligands. Additionally, poly(styrene sulfonic acid, sodium salt) (PSS) was used as a anionic polyelectrolyte. In this case, 0.5 M NaCl salt was added to the PSS solution.

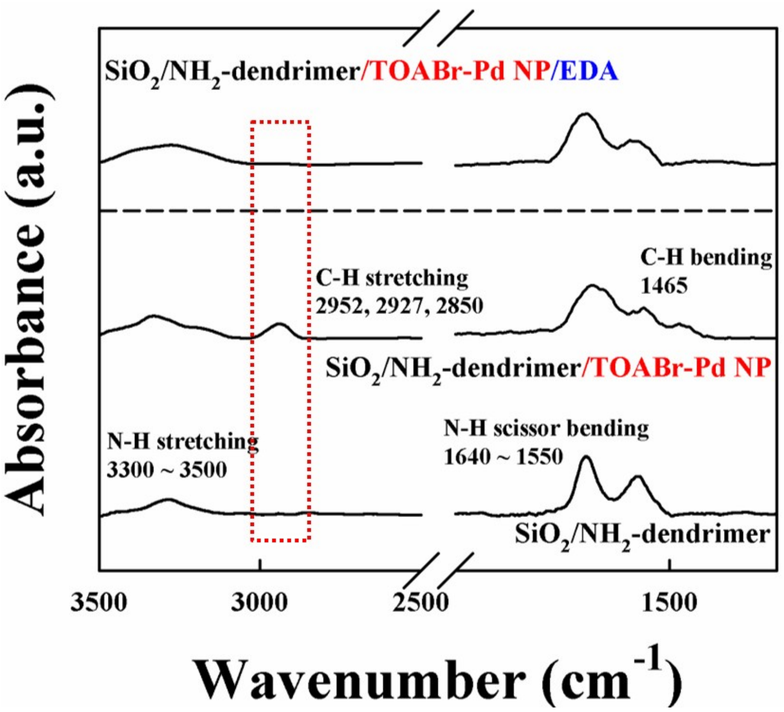


**Figure S5.** FT-IR spectra of $SiO_2$/$NH_2$-dendrimer, $SiO_2$/$NH_2$-dendrimer/TOABr-Pd NPs, and $SiO_2$/$NH_2$-dendrimer/TOABr-Pd NPs/EDA.

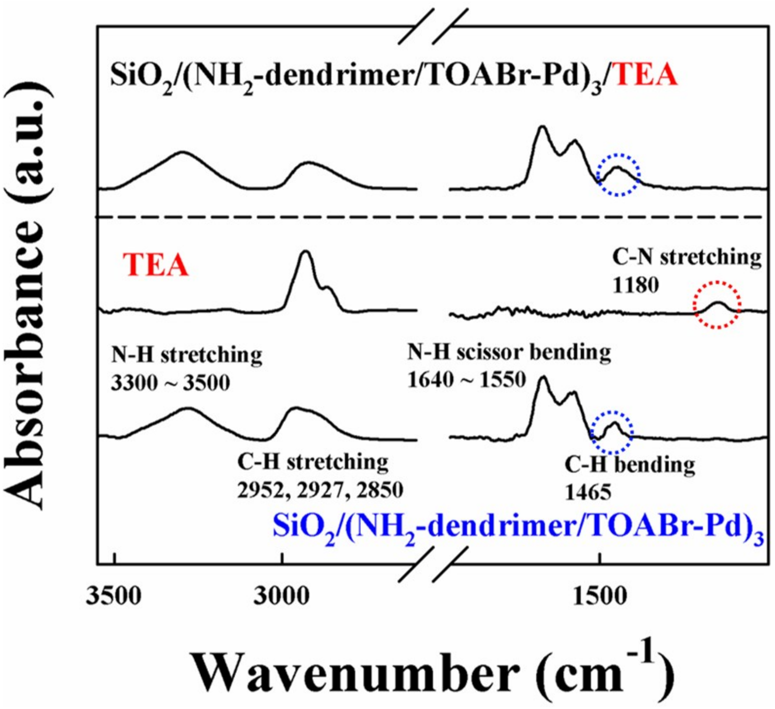


**Figure S6.** FTIR spectra of SiO$_2$/(NH$_2$-dendrimer/TOABr-Pd NPs)$_3$, triethylamine (TEA), and SiO$_2$/(NH$_2$-dendrimer/TOABr-Pd NPs)$_3$/TEA (i.e., the (NH$_2$-dendrimer/TOABr-Pd NPs)$_3$ multilayer-coated colloids treated in the TEA solution for 30 min). In the case of SiO$_2$/(NH$_2$-dendrimer/TOABr-Pd NPs)$_3$/TEA, we did not observe the C-N stretching peak (at 1180 cm$^{-1}$) occurring from TEA, and additionally the outermost TOABr ligands (*i.e.*, C-H bending peak at 1465 cm$^{-1}$) of SiO$_2$/(NH$_2$-dendrimer/TOABr-Pd NP)$_3$ were still observed after the TEA treatment.

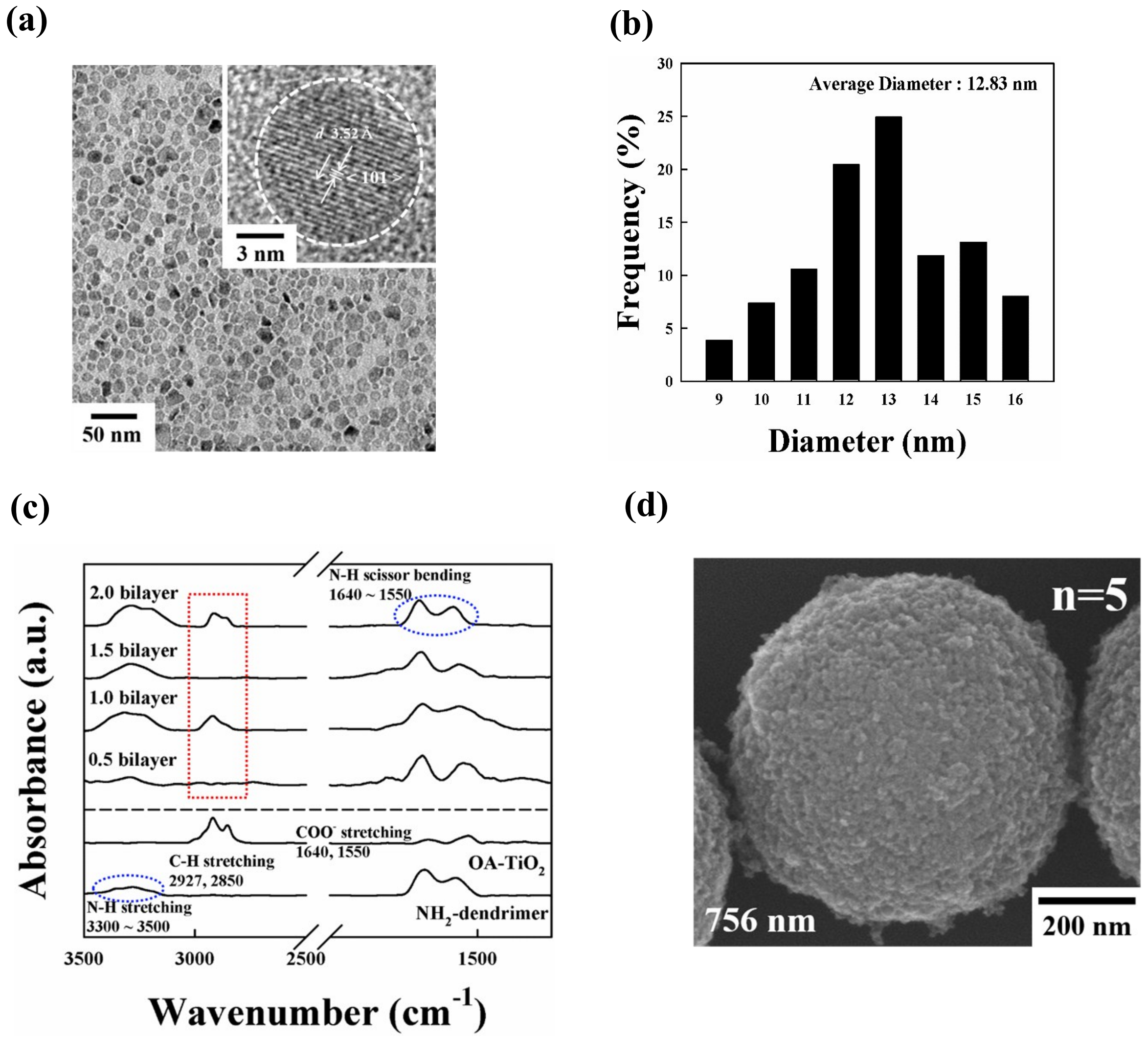


**Figure S7.** (a) HR-TEM image of 12.8 nm-sized OA-$TiO_2$ NPs. (b) Size distribution histogram of OA-$TiO_2$ NPs measured from HR-TEM images. (c) ATR-FTIR spectra of ($NH_2$-dendrimer/OA-$TiO_2$ NP)$_n$ multilayer films as a function of layer number (n), pristine OA-$TiO_2$ NP layer, and $NH_2$-dendrimer layer. OA-$TiO_2$ NPs were also deposited onto the $NH_2$-dendrimer-coated substrate via ligand exchange reaction between OA ligands bound to the OA-$TiO_2$ NP and amine moieties of the dendrimer. The characteristic C–H stretching peaks (at 2927 and 2850 cm$^{-1}$) by the long aliphatic chains of the OA ligands were generated by the deposition of OA-$TiO_2$ NPs as an outermost layer. However, they almost completely disappeared when the outermost layer was changed from OA-$TiO_2$ NPs to $NH_2$-dendrimers. These phenomena were repeated according to the alternating deposition of OA-$TiO_2$ NPs and $NH_2$-dendrimers, which implied the successful replacement of ligand between OA groups of $TiO_2$ NPs and $NH_2$ groups of dendrimers such as an adsorption mechanism shown in the ($NH_2$-dendrimer/TOABr-Pd NP)$_n$ and ($NH_2$-dendrimer/OA-$Fe_3O_4$ NP)$_n$ multilayers. (d) SEM image of ($NH_2$-dendrimer/OA-$TiO_2$ NP)$_5$ multilayer-coated colloid. Similar with the ($NH_2$-

dendrimer/TOABr-Pd NP$)_n$ multilayer-coated colloids, the ($NH_2$-dendrimer/OA-$TiO_2$ NP$)_5$ multilayers were homogeneously adsorbed onto the $SiO_2$ colloids without colloidal agglomeration.

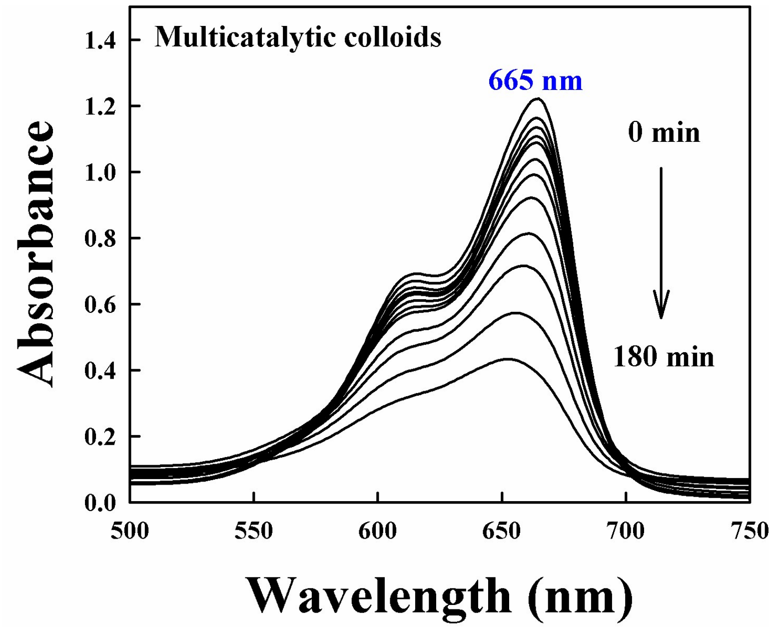


**Figure S8.** UV-Vis spectra of MB solution photodegraded by $SiO_2$/($NH_2$-dendrimer/OA-$Fe_3O_4$ NP$)_3$/($NH_2$-dendrimer/TOABr-Pd NP/$NH_2$-dendrimer/OA-$TiO_2$ NP$)_3$/$NH_2$-dendrimer from 0 to 180 min.

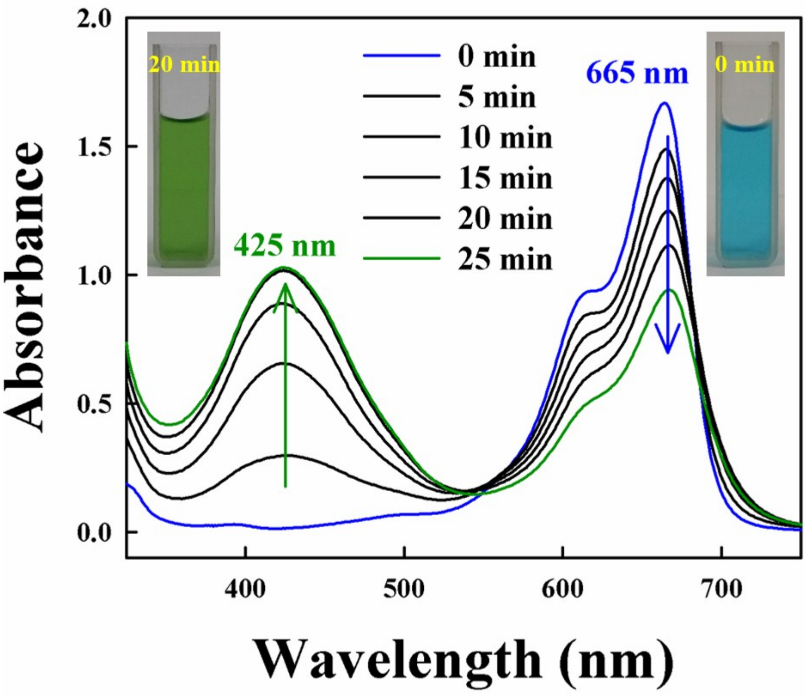


**Figure S9.** Cooperated multicatlaytic effects of multicatalytic colloids (*i.e.*, $SiO_2$/($NH_2$-dendrimer/OA-$Fe_3O_4$ NP)$_3$/($NH_2$-dendrimer/TOABr-Pd NP/$NH_2$-dendrimer/OA-$TiO_2$ NP)$_3$/$NH_2$-dendrimer) in aqueous mixture solution containing MB (15 ppm) and OPD (0.3 mM) in the presence of 100mM $H_2O_2$.

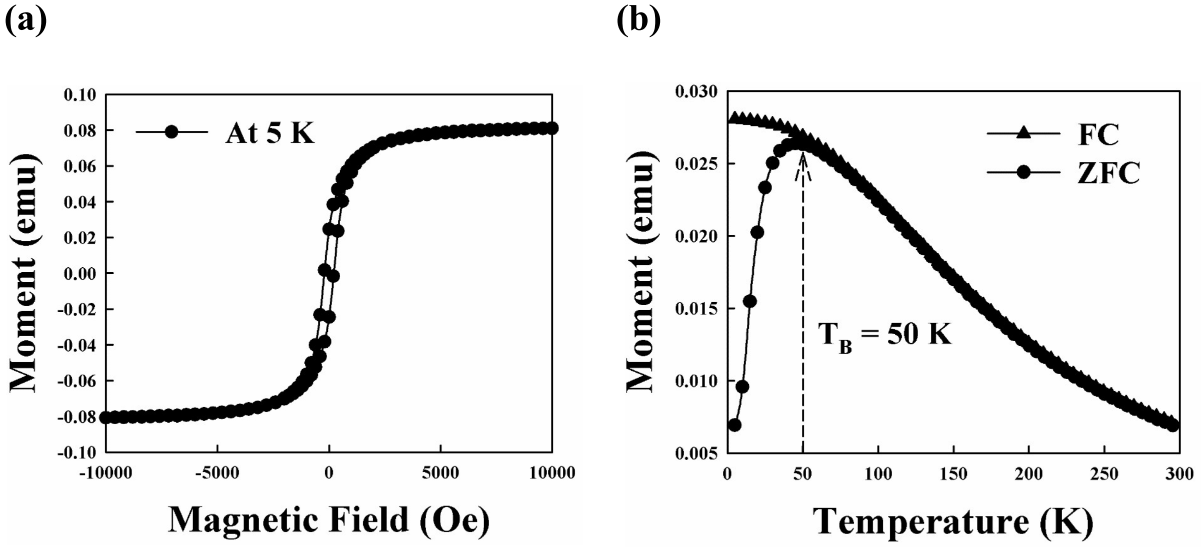


**Figure S10.** (a) Magnetism curve of multicatalytic colloids with magnetic properties measured at 5 K. At liquid helium temperature (T = 5 K), the thermally activated magnetization-flipping properties of the colloids revealed frustrated superparamagnetic properties. (b) Temperature dependence of zero-field-cooling (ZFC) and field-cooling (FC) magnetization measured using 150 Oe. The blocking temperature ($T_B$), which begins to show deviation between zero-field-cooling (ZFC) and field-cooling (FC) magnetization, was fixed to approximately 50 K. It has been reported that $T_B$ of 8-nm $Fe_3O_4$ NPs is approximately 50 K.[S1]

Ref. [S1] J. Park, K. An, Y. Hwang, J.-E. Park, H.-J. Noh, J.-Y. Kim, J.-H. Park, N.-M. Hwang, T. Hyeon, *Nat. Mater.* **2004**, *3*, 891.