Supporting Information

Magnetically Recoverable Photocatalyst Prepared by Supporting TiO₂ Nanoparticles on Superparamagnetic Iron Oxide Nanocluster Core@Fibrous Silica Shell Nanocomposite

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Figure S1. TEM image of the as-prepared superparamagnetic iron oxide nanocluster.

Synthesis Step	Sample Name	Nanoparticle	TEOS [µl]	CTAB [g]	TEA [g]	TBOT [µl]	HPC [mg]	Temp. (°C)	Reaction Time	Water	Solvent
1	SION ^{§§}	-	-	-	-	-	-	200	10 hour	-	-
2-1 (1 st FSS Generation)						-	-				274 ml 1-Octadecene [§]
2-2 (3 rd FSS Generation)	SION@FSS1	200 ml SION† (9.3mg/ml)	685^{\dagger}	92.9*	2.5*	-	-	60	12 hour	416 ml	274 ml Decalin [§]
2-3 (3 rd FSS Generation)						-	-				274 ml Cyclohexane [§]
3 (CTAB removal)	SION@FSS (powder) ^{¶¶}	(d ried in 60 °C oven	-	-	-	-	-	550	5 hour	-	-
4	SION@FSS _@ Am_ TiO _{2††}	110 mg SION@FSS	-	-	-	800	0.6	85	100 min	100 μl	200 ml Ethanol 100 μl
5	SION@FSS@A_ TiO _{2§§}	sıunwrss @Am_TiO ₂	-	-	-	-	-	180	1.5 hour	-	DI Water Ethanol

Table S1 Summary of synthesis conditions

* The reagent is added once in the 1st FSS layer generation and the remaining is used in the 2nd and 3rd FSS layer generation.

† The reagent is added in each FSS stratification step.

§ The reagent is added in each FSS stratification step and discarded at the end of each layer generation step

¶ Reaction in a 2L multi-neck flask with mechanical stirring.

†† In a 500 ml round bottom flask with reflux and mechanical stirring.

§§ Hydrothermal reaction

 $\P\P$ Annealed in a tube furnace

Synthesis Step	Sample Name	Controlled Parameter	Controlled Amount [ul]	Temperaure (°C)	Reaction Time	Other synthesis Parameters
2-1 (1 st FSS Generation)	SION@FSS	TEOS	4000-12.5	60	12 hour	Oil solvent: 1-Ocatadecene
2-3 (3 rd FSS Generation)	bioinerbb	TEOS	200-20	60	12 hour	Oil solvent: Cyclohexane

Table S2 Summary of synthesis conditions during the FSS thickness control of SION@FSS

Reaction in a 250ml round bottom flask with reflux mechanical stirring.



Figure S2. TEM images of the superparamagnetic iron oxide nanocluster core@fibrous silica shell nanocomposites after the first layer stratification synthesized with varying amounts of TEOS (a) 4 ml, (b) 1 ml, (c) 500 μ l, ((d),(e)) 250 μ l, ((f), (g)) 50 μ l and (h) 12.5 μ l for 6 h. (Refer to the synthesis step 2-1 in the **table S2** (ESI⁺) for the synthesis condition)



Figure S3 TEM images of the superparamagnetic iron oxide nanocluster core@fibrous silica shell nanocomposite at the end of the third stratification synthesized with varying amounts of TEOS (a) 50 μ l, (b) 100 μ l and (c) 200 μ l.

(Refer to the synthesis step 2-3 in the table S2 (ESI⁺) for the synthesis condition)



Figure S4 TEM images of SION@FSS@A-TiO₂ synthesized with varying amounts of TBOT (a) 10 μ l, (b) 50 μ l, (c) 800 μ l and (f) 2 ml.



Figure S5 Nitrogen adsorption desorption isotherms of (a) SION@FSS, (b) SION@FSS@Am-TiO₂ and (c) SION@FSS@A-TiO₂.



Figure S6 Energy-dispersive X-ray spectroscopy (EDS) spectrum of (a) SION@FSS@Am-TiO₂ and (b) SION@FSS@A-TiO₂.



Figure S7 Retention of the morphology of SION@FSS@A-TiO₂ after repetitive dye degradation cycles.



Figure S8 The Calibration curve of methylene blue and the linear fitting result.



Figure S9 The digital image of photo-reactor system. The system was covered with aluminum foil during UV-illumination to avoid exposure to UV light and shield plastic and rubber components.



Figure S10 MetB concentration of supernatant after stirring the MetB solution with $IO@FSS@TiO_2$ without UV illumination.



Figure S11 Photodecolorization test with reduced amount of MetB (3mg/L) shown in (a) absorbance *vs.* time, (b) Ln(C/CO) *vs.* time, (c) absorbance *vs.* wavelength, and (d) repeated trials of dye degradation test was performed using recovered SION@FSS@A-TiO₂ nanoparticles.



Figure S12 Photodecolorization test performed with reduced amount of MetB (7.5 mg/L) (a) with IO@FSS@TiO₂ and (b) P25.

Catalyst	Catalyst Concent ration [mg/ml]	Time required for reaching adsorption- desorption equilibrium [hour]	MetB Concen tration [mg/L]	Time required for complete dye removal [hour]	UV lamp Power [mW/ <i>cm</i> ² ,W]	UV wave length [nm]	ref
Rattle type $Fe_3O_4/SiO_2/TiO_2$	0.1	-	1	0.167 (10 min.)	800,-	254	[S1]
Fe ₃ 0 ₄ @rG0@TiO ₂	1.5	0.5	10	2	-,300	UV lamp with cut off filter	[s2]
sion@fss@a- ^{TiO} 2	0.1	2	3	3	50, 48	312	This work

Table S3 Comparison of MetB dye degradation using magnetically recollectablenanocomposites with TiO_2 nanoparticles.

References

- S1. S. Linley, T. Leshuk and F. X. Gu, ACS Appl. Mater. Interfaces, 2013, 5, 2540-2548.
- S2. X. Yang, W. Chen, J. Huang, Y. Zhou, Y. Zhu and C. Li, *Sci. Rep.*, 2015, **5**, 10632.