Supporting Information

Bio-inspired carbon nitride mesoporous spheres for artificial photosynthesis: photocatalytic cofactor regeneration for sustainable enzymatic synthesis**

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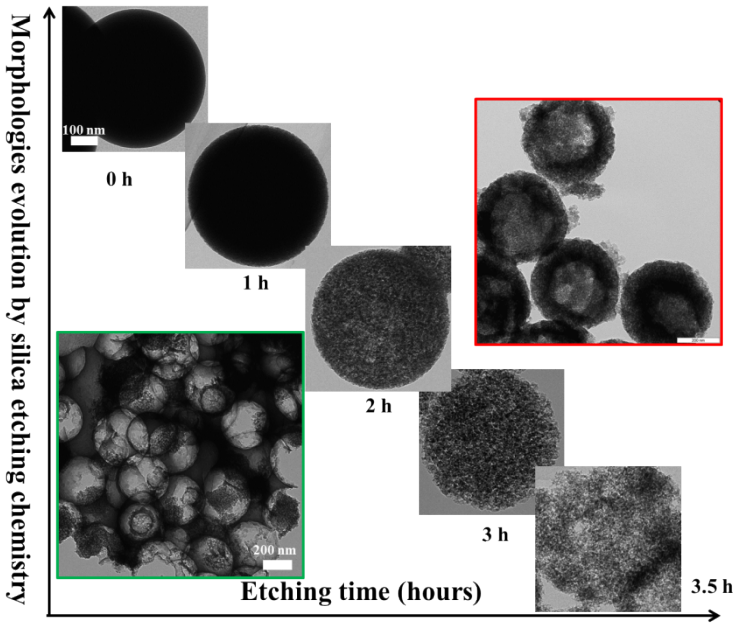


Figure S1. Morphologies evolution of SiO_2 spheres during different etching times. The inset image with green frame indicates the unsuccessful templating from 2 hours etched samples, while the image labeled with red square indicates the successful templating from 3 hours etched samples.

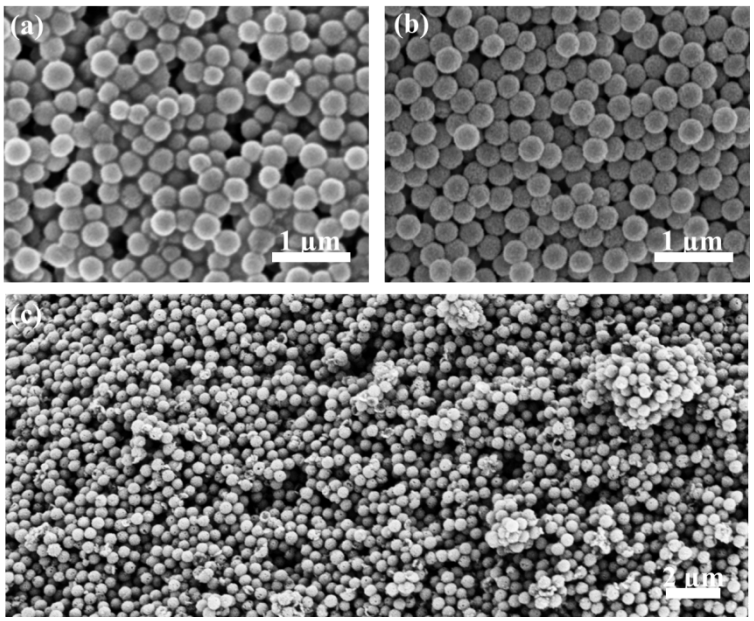


Figure S2. SEM images of silica before (a) and after etching (b). Image (c) illustrates the large area overview of CNMS.

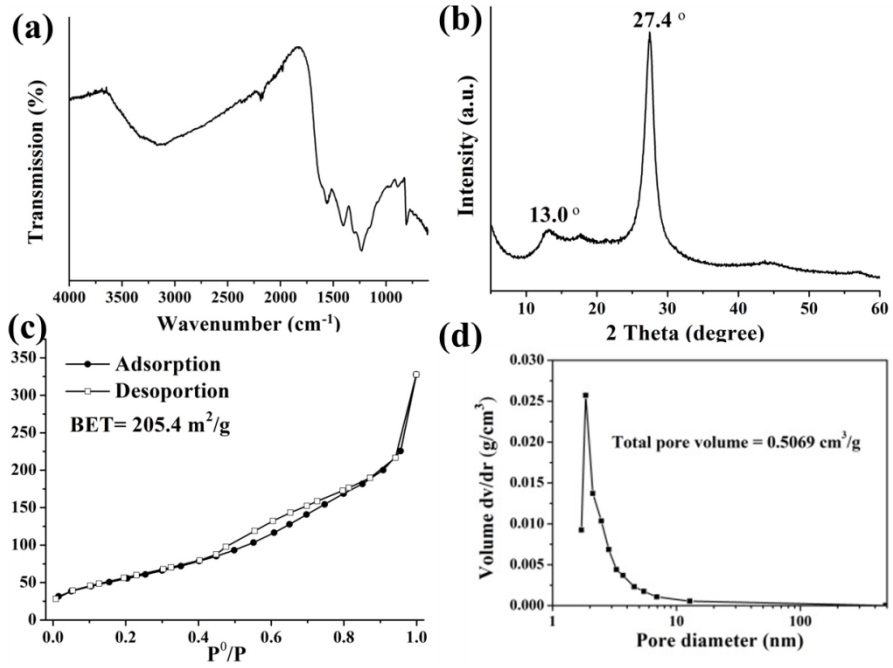


Figure S3. (a) FTIR spectrum of CNMS showing typical C-N heterocycle stretches in the 1100~1600 cm^{-1} spectral range and the breathing mode of the tri-s-triazine units at 810 cm^{-1}. (b) XRD spectrum of CNMS with two peaks at 13.0º and 27.4º ascribed to the in-planar repeat period and stacking of the conjugated aromatic system, respectively. (c) N_2 sorption isotherm of CNMS with the pore size distribution plot in (d).

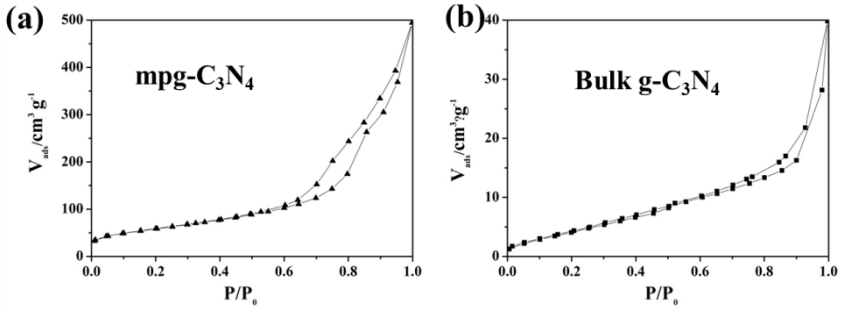


Figure S4. N_2 sorption isotherms of mpg-C_3N_4 (a) and bulk g-C_3N_4.

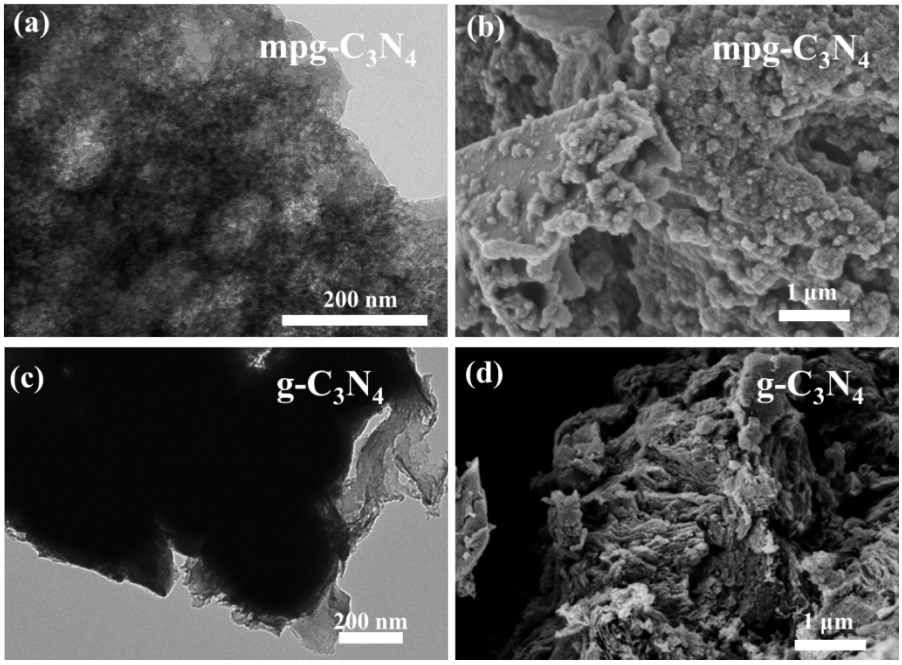
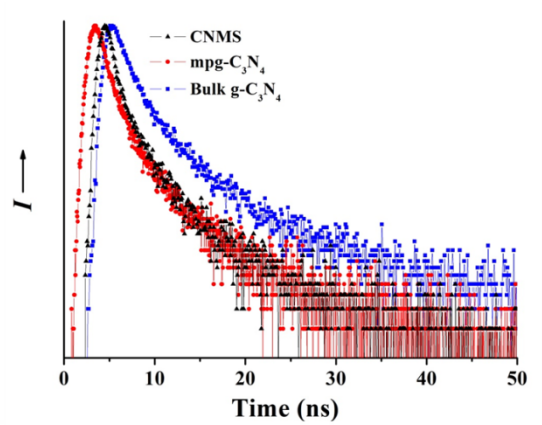


Figure S5. TEM and SEM characterizations of mpg-C_3N_4 (**a** and **b**) and g-C_3N_4 (**c** and **d**), respectively.



	Components	Lifetime (ns)	Rel%
Bulk g-C_3N_4	τ_1	1.5889	63.86
	τ_2	9.0553	36.14
mpg-C_3N_4	τ_1	0.8684	68.82
	τ_2	5.1380	31.18
CNMS	τ_1	0.5928	69.07
	τ_2	4.8226	30.93

Figure S6. Time resolved fluorescence decay spectra of CNMS, mpg-C_3N_4 and g-C_3N_4. The samples were excited by the incident light of 420 nm from a picosecond pulsed light-emitting diode.

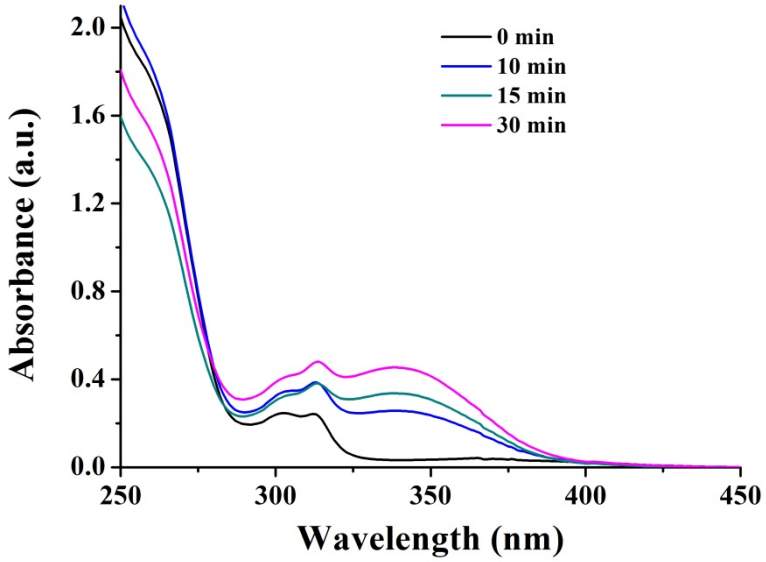


Figure S7. Spectral measurements of NADH concentration in mediator involved reaction solution by CNMS photocatalysis. β-NAD^+, 1 mM; **M**, 0.25 mM; TEOA, 15 w/v%; PBS buffer, 0.1 M, pH=8; CNMS, 3 mg.

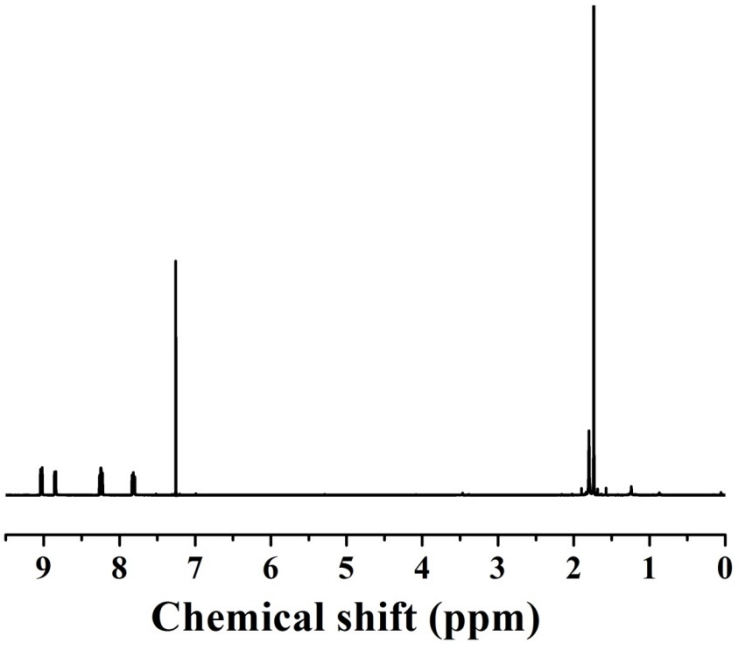


Figure S8. NMR of [Cp*Rh(bpy)Cl]Cl compound. H NMR (300 MHz, CDCl3): Cp*[Rh(2,2'-bpy)] δ(ppm) =9.02 (d, 2H, H-3, 3'), 8.84 (d, 2H, H-6, 6'), 8.24 (t, 2H, H-5, 5'), 7.81 (t, 2H, H-4, 4'), 1.74 (s, 15H, Cp*).

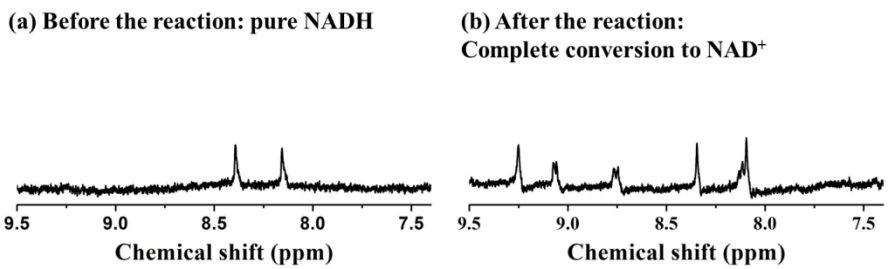


Figure S9. Partial HNMR spectra for the reaction between NADH and pyruvate in the presence of L-lactate dehydrogenase: (a) before the reaction; (b) after enzymatic reaction.

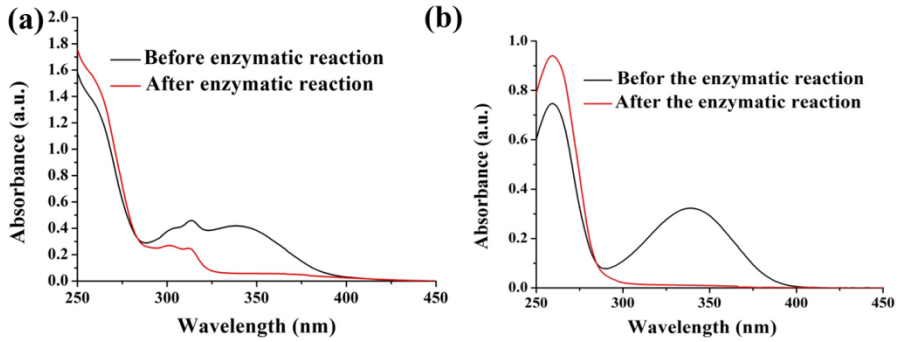


Figure S10. Spectral measurements of as-regenerated NADH (a) and commercial NADH (b) before (black plot) and after the enzymatic reaction (red plot), respectively.

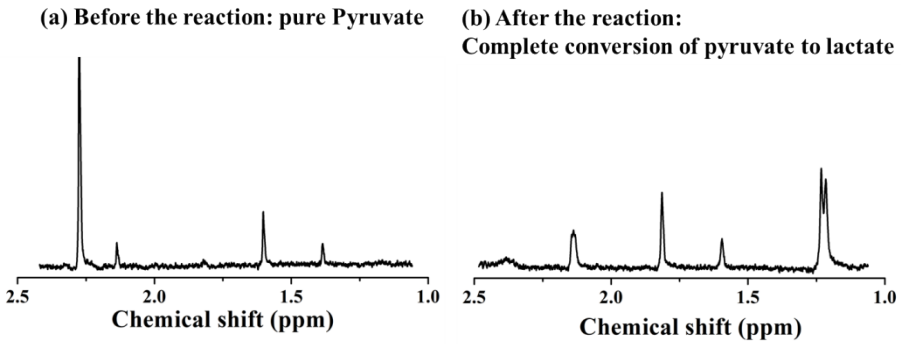


Figure S11. Partial HNMR spectra for the reaction between in-situ regenerated NADH and pyruvate in the presence of L-lactate dehydrogenase: (a) before the reaction; (b) after the enzymatic reaction.