Electronic Supplementary Information

Facile Assembly of Graphitic Carbon Nitride Film at Air/Water Interface for Photoelectrochemical NADH Regeneration

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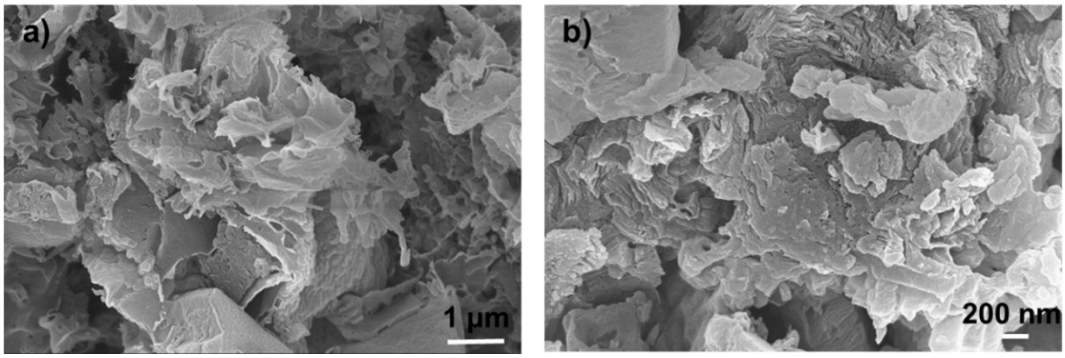

Fig. S1. SEM images of BCN before ball milling at a) low magnification, and b) high magnification.

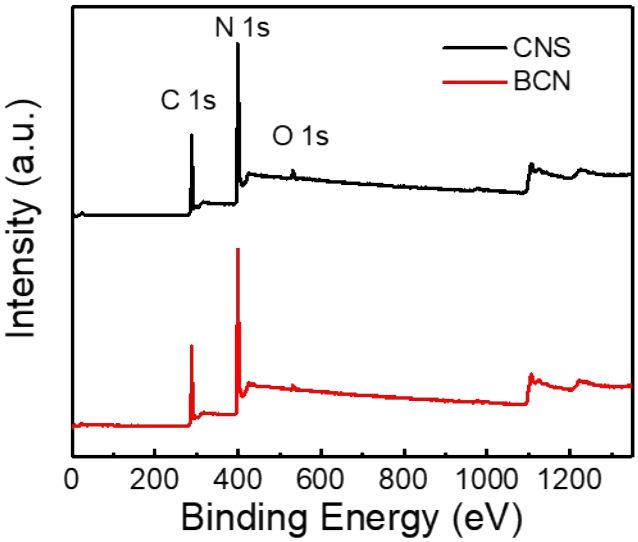

Fig. S2. XPS spectra (survey) of BCN and CNS, respectively.

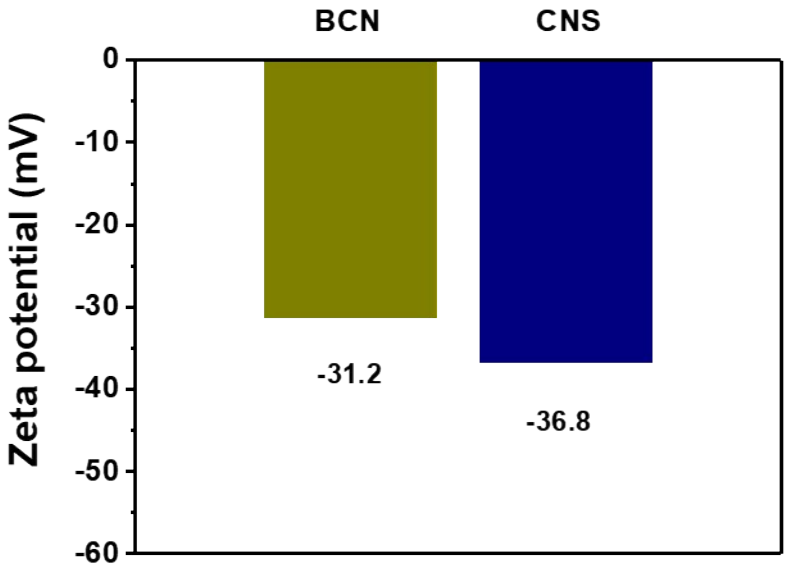

Fig. S3. Zeta potential of BCN and CNS measured in H_2O (pH = 7), respectively.

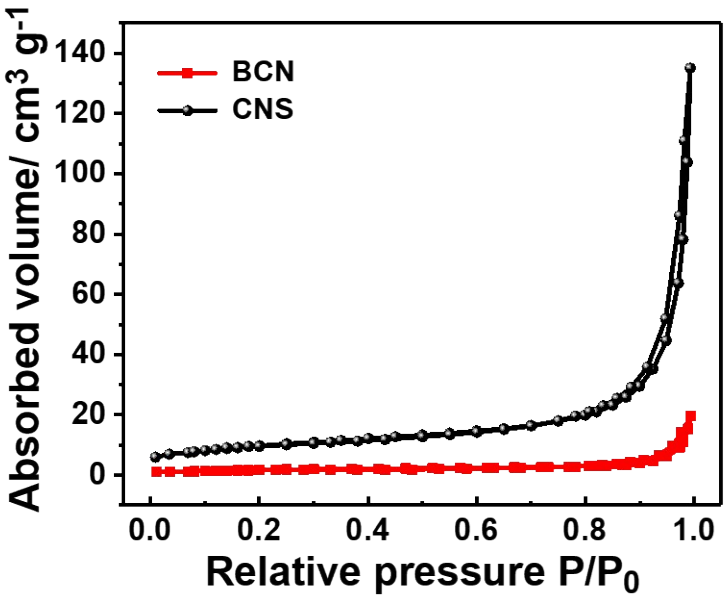

Fig. S4. N_2 adsorption-desorption isotherms plots of BCN and CNS, respectively.

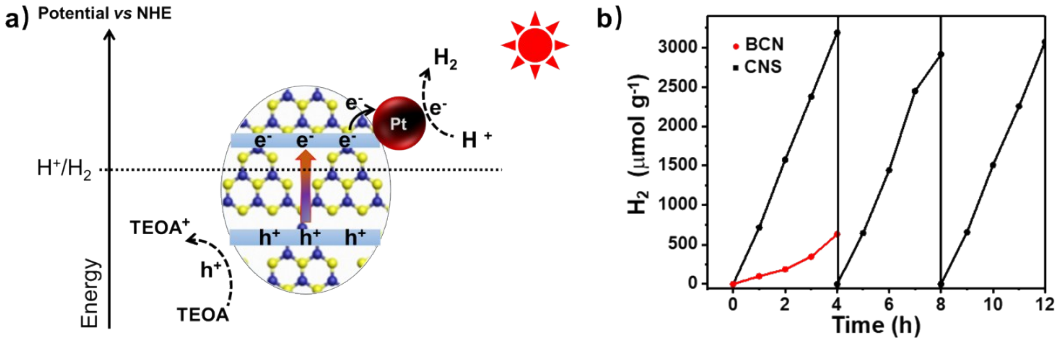

Fig. S5. a) Illustration of photocatalytic mechanism for H_2 evolution, b) photocatalytic H_2 evolution performances by BCN and CNS, respectively.

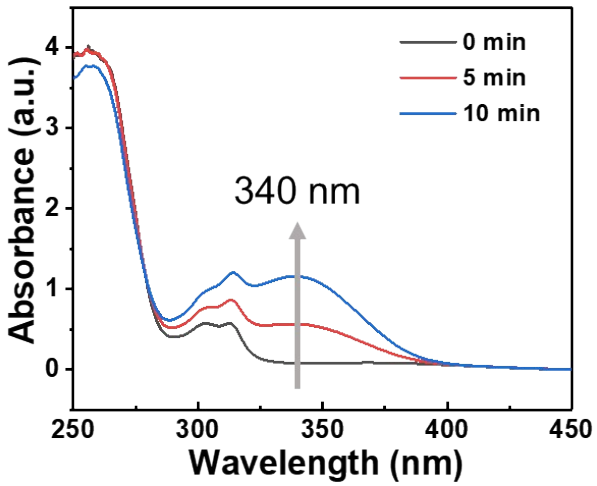

Fig. S6. UV-vis absorption spectra profile of NADH with CNS as a function of visible light (450 nm) irradiation time.

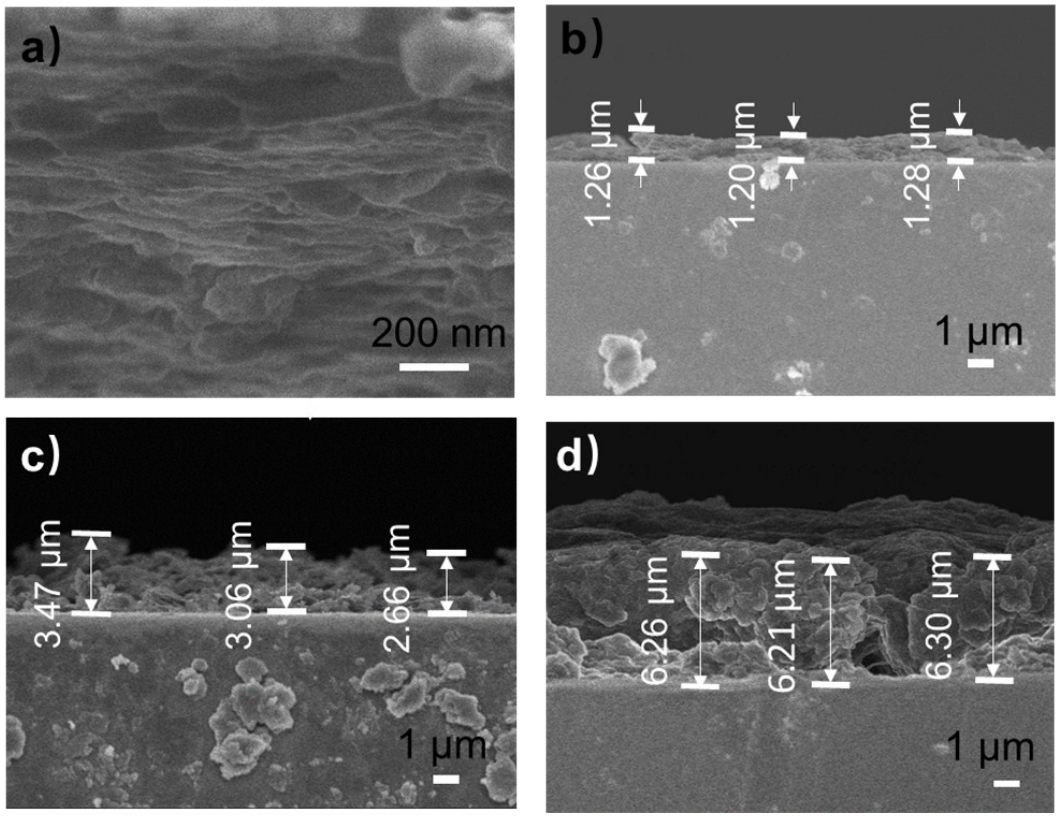

Fig. S7. Cross section of SEM images of CNS films: a) high magnification of the sample before annealing treatment, b) CNS-1-film, c) CNS-2-film, and d) CNS-3-film.

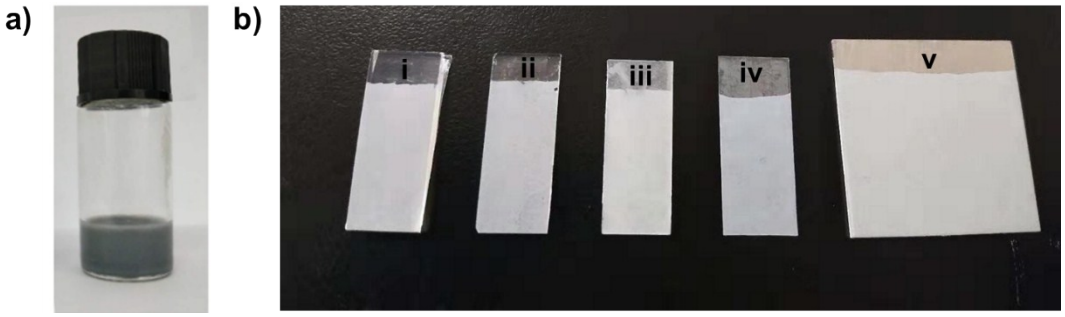

Fig. S8. a) Digital photo of CNS-Gr dispersion in ethanol solution for preparation film. b) CNS-Gr films on different substrates: i) FTO, ii) glass slide, iii) frosted glass, iv) flexible substrate (ITO/PET), v) titanium plate, respectively.

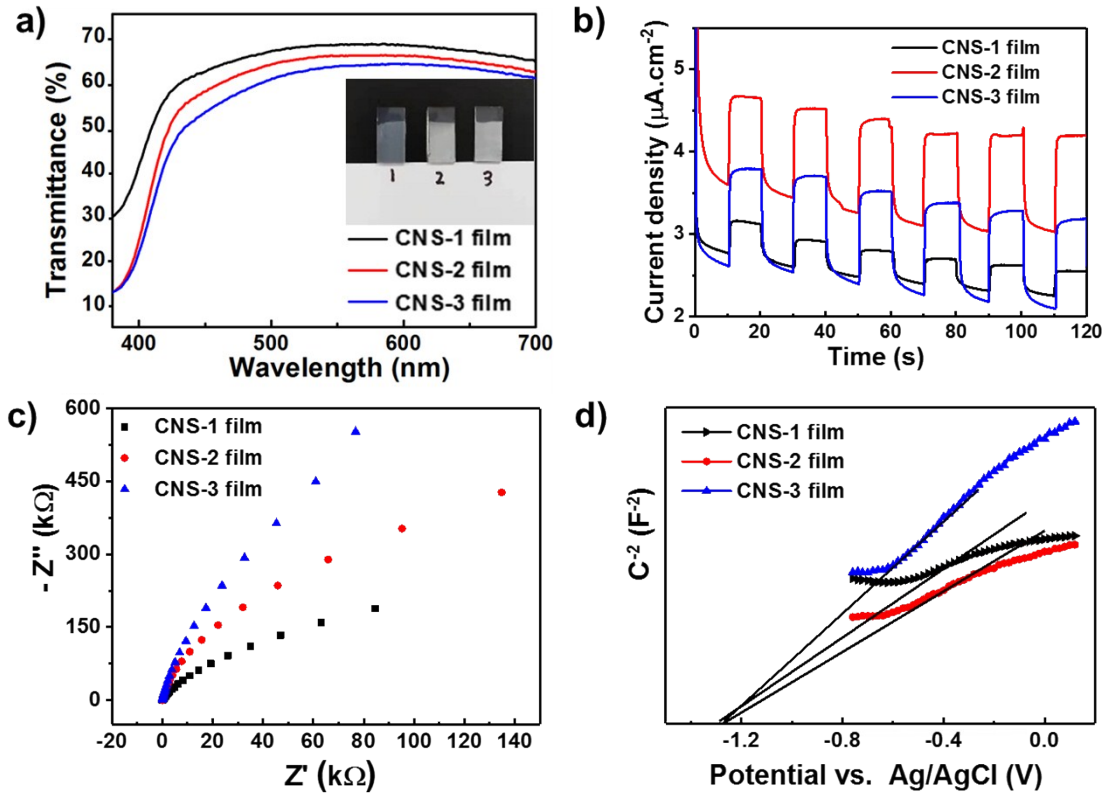

Fig. S9. a) Transmittance, b) photocurrent tests (in 0.2 M Na_2SO_4 aqueous solution detected under unbiased conditions), c) electrochemical impedance spectra, and d) M-S plots of CNS films with different transferring layers.

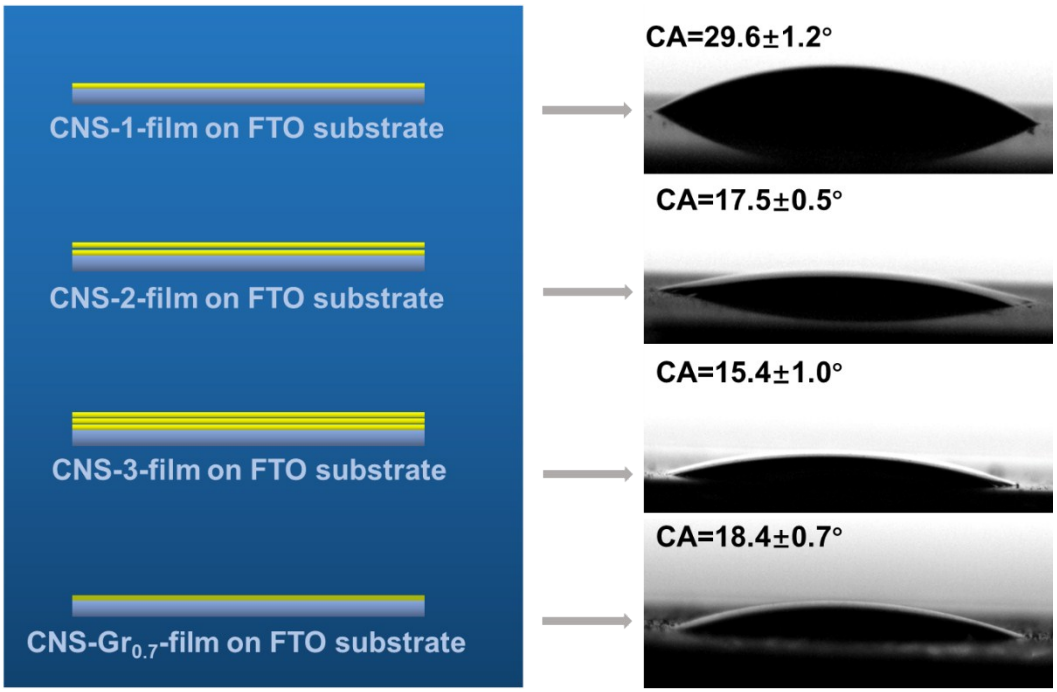

Fig. S10. Optical contact angle images of water on the CNS films with different transferring layers (CNS-1-film, CNS-2-film, and CNS-3-film) and CNS-$Gr_{0.7}$-film.

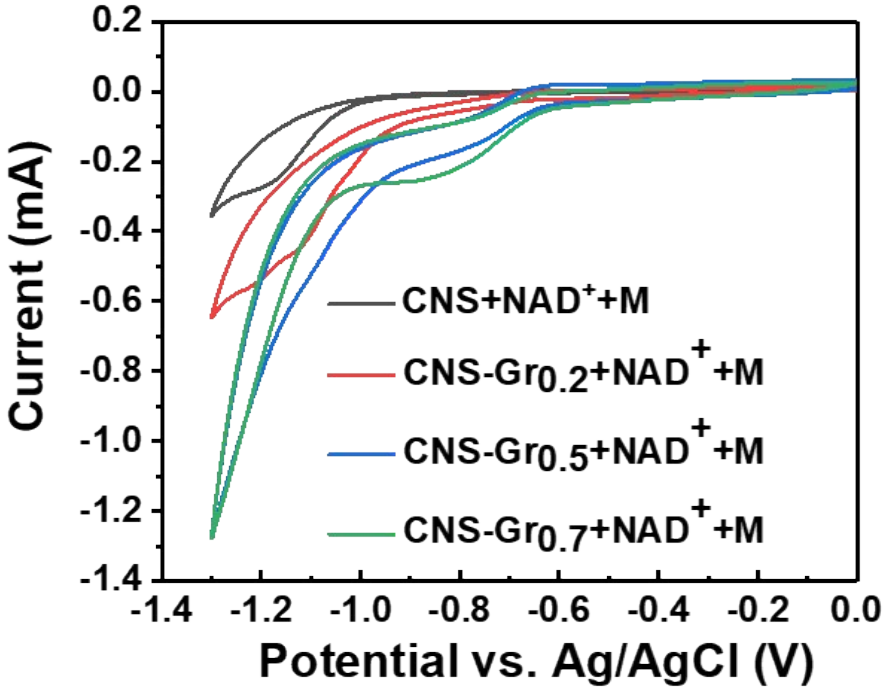

Fig. S11. Cyclic voltammograms of CNS assembling with different amount of graphene in phosphate buffer in the presence of **M** with NAD^+.

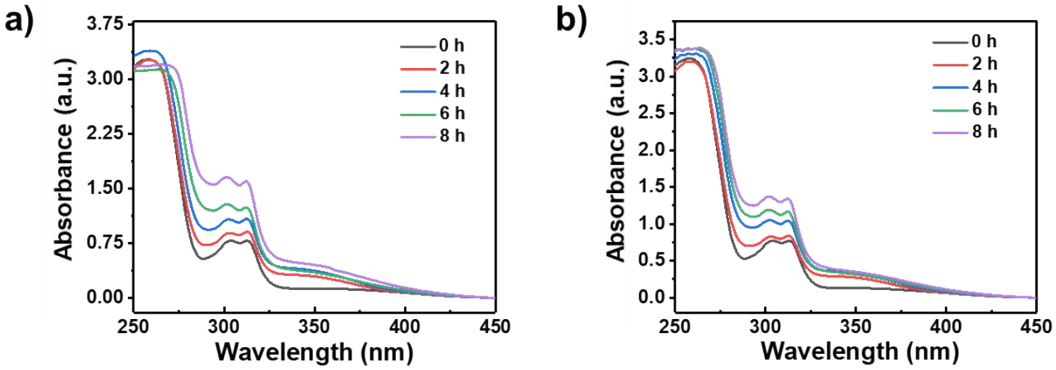

Fig. S12. a) Temporal changes of UV-vis absorption spectra of NADH regeneration with (a) CNS-$Gr_{0.7}$ and (b) CNS PEC electrodes as a function of visible light (≥ 420 nm) irradiation time.

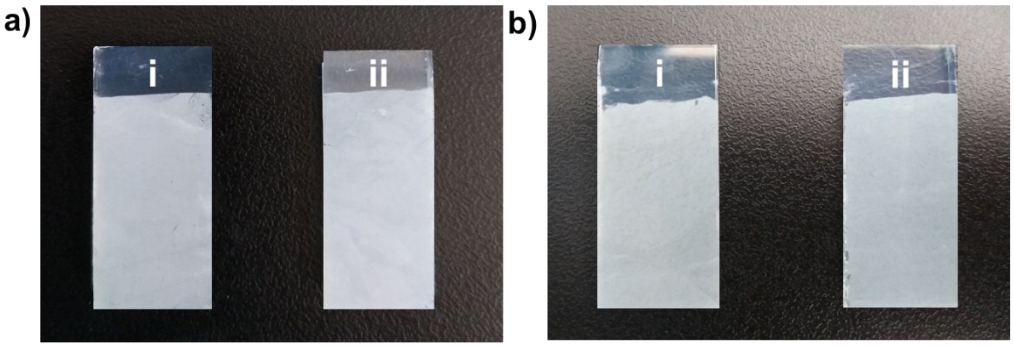

Fig. S13. Optical images of a) CNS film and b) CNS-$Gr_{0.7}$ film from the same batch before (i) and after (ii) immersed in water for 24 h, respectively.

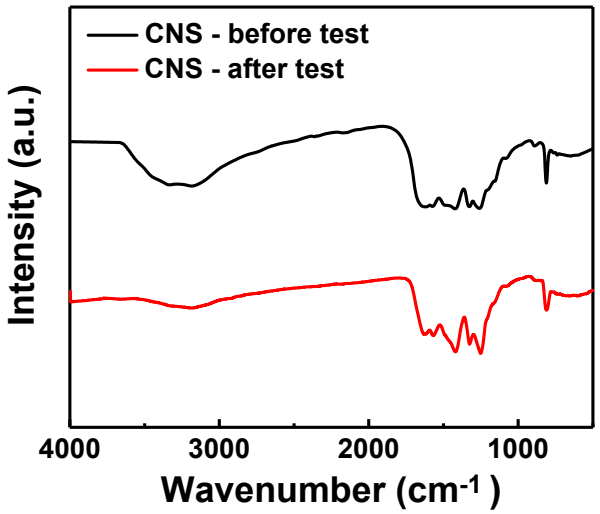

Fig. S14. FTIR spectra of CNS film before and after the PEC test.

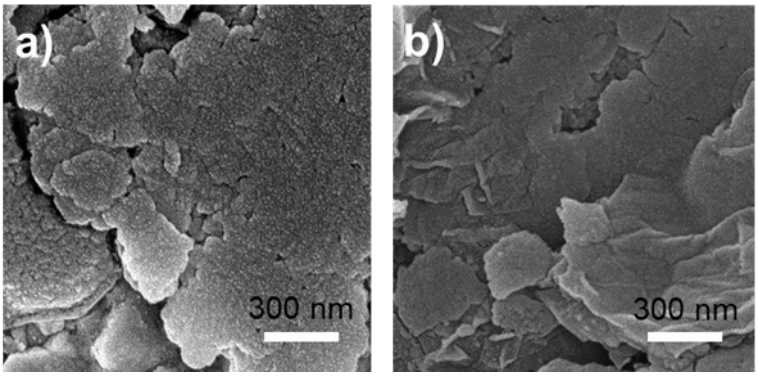

Fig. S15. SEM images of a) CNS film, b) CNS-$Gr_{0.7}$ film after the PEC reaction.

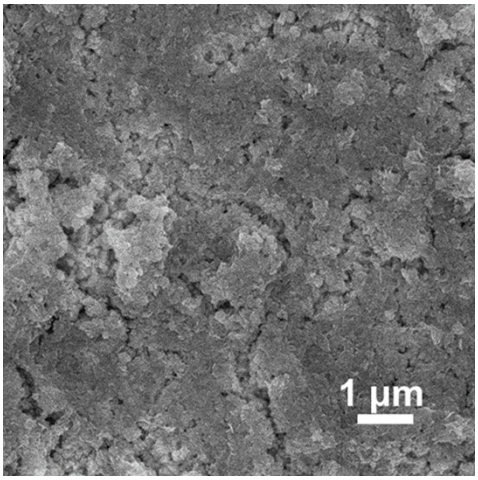

Fig. S16 SEM image of TiO_2 film fabricated by interfacial self-assembly method.

Table S1. Comparison of the concentration of the g-C_3N_4 nanosheet reported in the previous literatures.

sample	Concentration (mg mL^{-1})	Ref
ultrathin g-C_3N_4 nanosheet	0.15	[1]
few-layer g-C_3N_4 nanosheet	0.8	[2]
Porous g-C_3N_4	3	[3]

Table S2. Comparison of photoelectrochemical NADH regeneration yield using water as an electron donor

photoelectrode		incident light	illumination mode		applied	NADH regeneration	
Anode	Cathode	power (Xe lamp)	Anode	Cathode	voltage	yield in 2 h (%)	Ref
$FeOOH/Fe_2O_3$	Black silicon	450 W	Light	Light	1.2 V	~ 20	[4]
$FeOOH/Fe_2O_3$	Black silicon	450 W	Light	Dark	1.2 V	~ 5.6	[4]
$FeOOH/Fe_2O_3$	Black silicon	450 W	Dark	Light	1.2 V	~ 1.6	[4]
N/A	CNS	300 W	N/A	Light	-0.9 V	13	This work
N/A	CNS-$Gr_{0.7}$	300 W	N/A	Light	-0.9 V	19	This work

Supplementary reference

[1] X.D. Zhang, X. Xie, H. Wang, J.J. Zhang, B.C. Pan, Y. Xie, *J Am. Chem. Soc*. **2012**, 135, 18.

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