Supporting Information

Bioinspired Electron Carrier Mediated Transmembrane

Photocatalytic Hydrogen Evolution in Silica Colloidosomes

Chengkun Bai, [a] Bingdi Wang, [a] Zhengshun Jiang, [a] Chunying Lv, [a] Zhenning Liu, [a] Shiyu Wang, [c] Songliang, *[a] Hongying Zang *[b]

a. Key Laboratory of Bionic Engineering (Ministry of Education), College of Biological and Agricultural Engineering, Jilin University, Changchun, 130022 P. R. China.

b. Key Laboratory of Polyoxometalate Science of the Ministry of Education, Faculty of Chemistry, Northeast Normal University, Changchun, 130024 P. R. China

c. Key Laboratory of High Performance Plastics, Ministry of Education, College of Chemistry, Jilin University, Changchun, 130012 P. R. China.

*Corresponding authors.

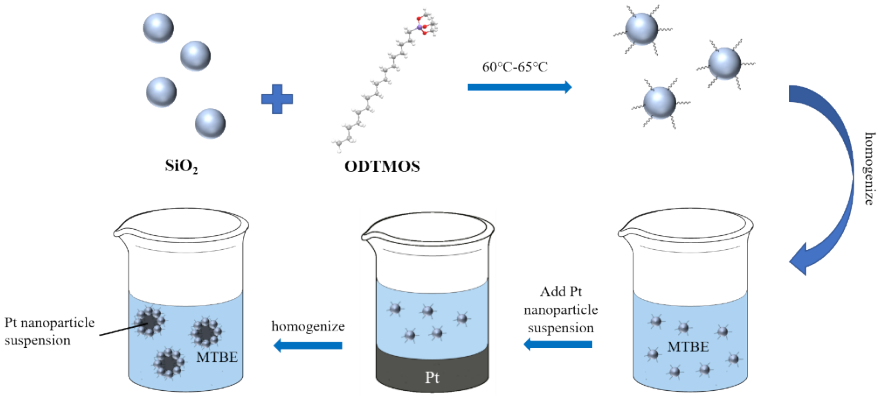

Fig. S1. Schematic illustration of the preparation process of Pt@OD-SiO_2.

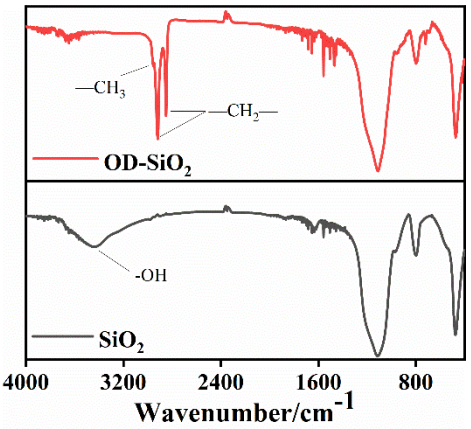

Fig. S2. Fourier transform infrared (FTIR) spectroscopy of SiO_2 and OD-SiO_2.

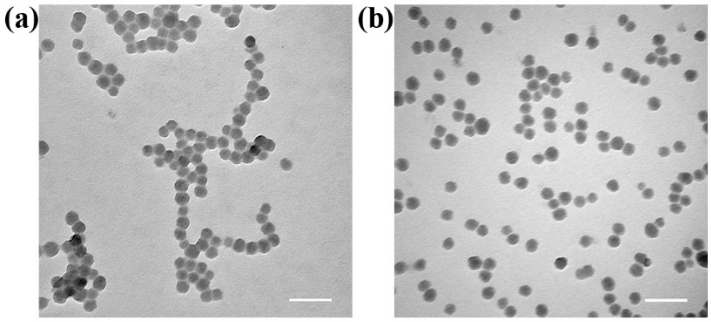

Fig. S3. TEM images of (a) SiO_2 and (b) OD-SiO_2. Scale bar:100 nm.

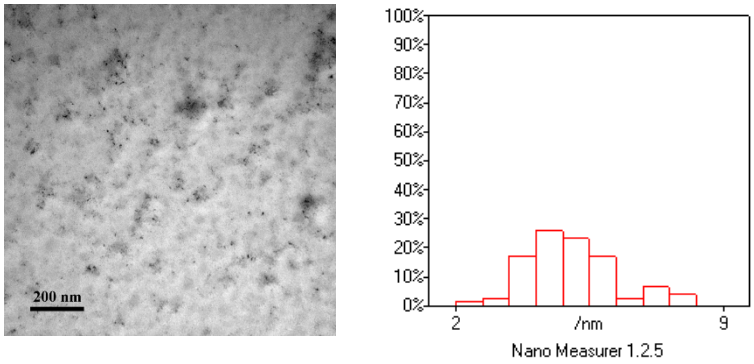

Fig. S4. TEM images of Pt nanoparticles.

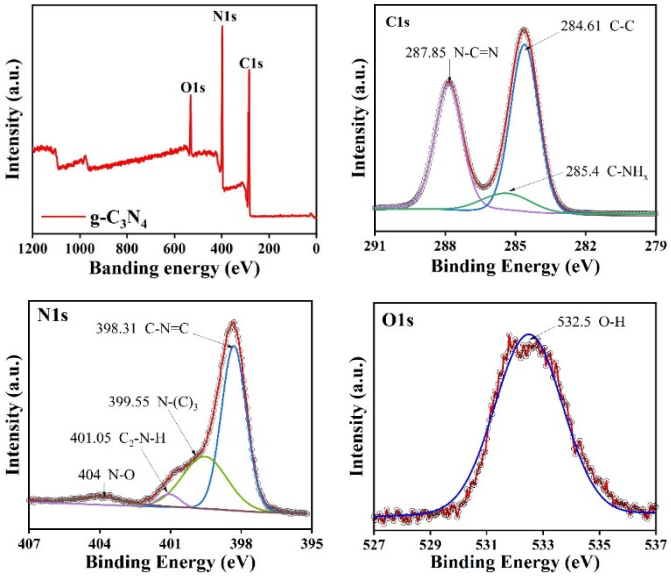

Fig. S5. The XPS images of g-C_3N_4

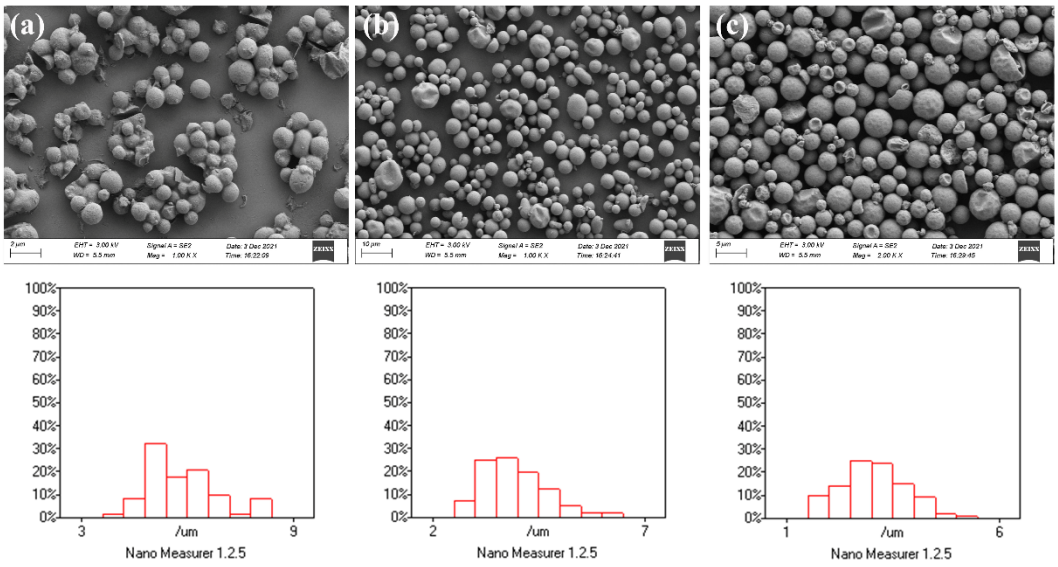

Fig. S6. The size distribution of colloidosomes with three different R values. (a) R = 0.05 g/ml, (b) R = 0.1 g/ml, (c) R = 0.2 g/ml.

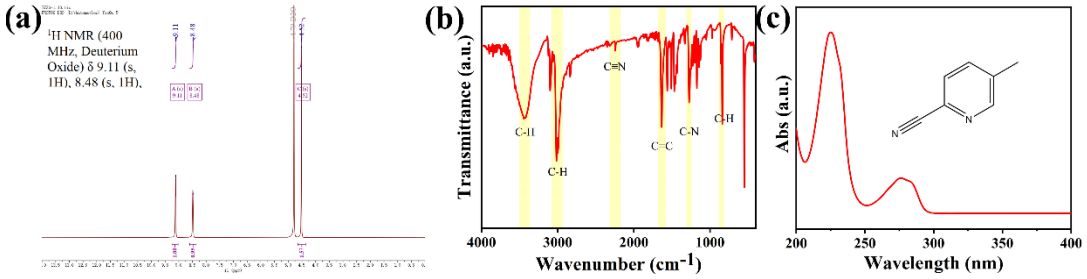

Fig. S7. The ^{1}H NMR spectra (a), FTIR spectra (b) and UV−vis absorption spectra (c) of MCP^{+}.

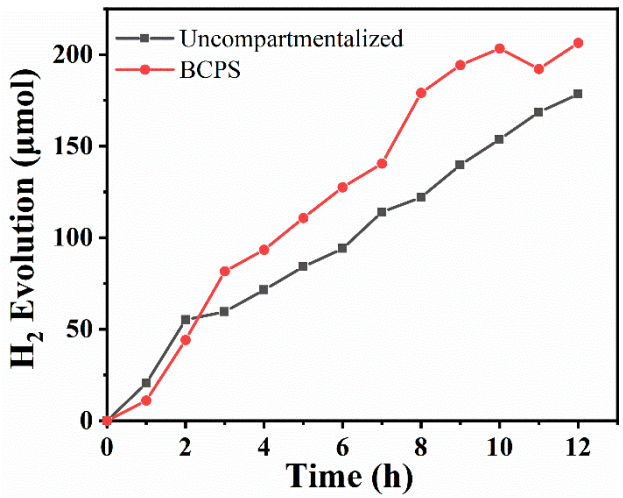

Figure S8. Hydrogen evolution of the BCPS and control group within 12 h.

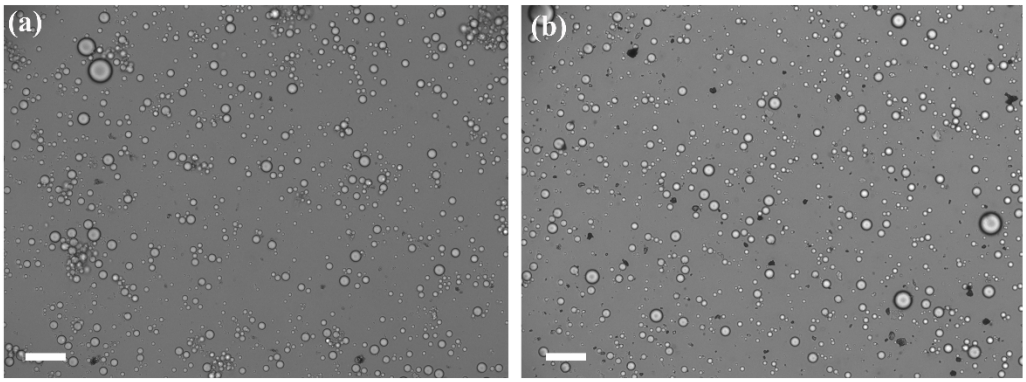

Figure S9. Microscopy images of the colloidosomes in the BCPS system before (a) and after (b) the reaction (Scale bar: 100 μm). The opaque sheet-like structures in (b) correspond to g-C_3N_4.

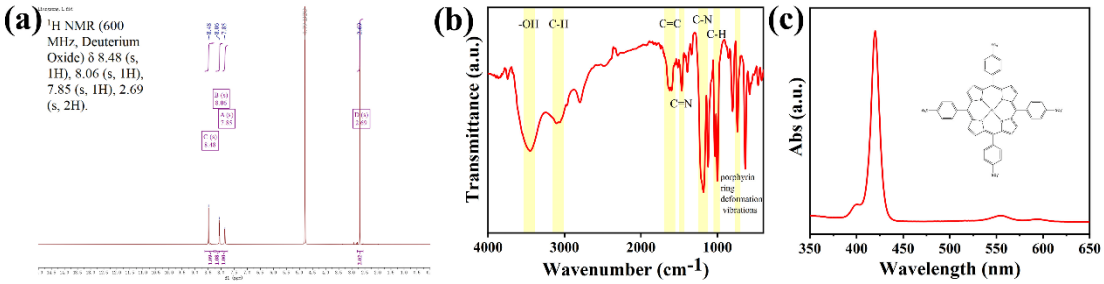

Fig. S10. The ^{1}H NMR spectra (a), FTIR spectra (b) and UV−vis absorption spectra (c) of ZnTPPS. The signal at δ 2.69 (s, 2H) originates from the -CH_3 group in zinc acetate

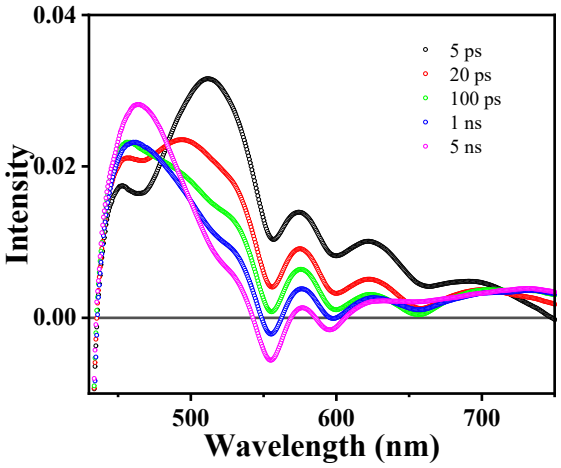

Fig. S11. Time-resolved transient fluorescence spectroscopy of ZnTPPS.

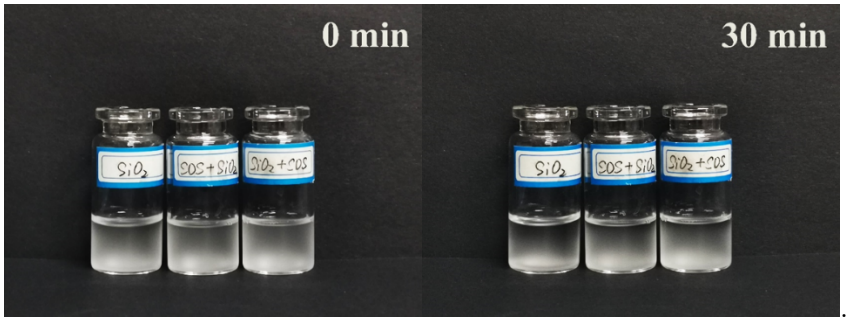

Fig. S12. Photographs of OD-SiO_2 colloidosomes and SDS-containing OD-SiO_2 colloidosomes after standing for 30 min.

Table 1 Summary of photocatalytic reactions in microreactors with different photocatalyst

Photocatalysts	Applications	Year	Ref.
PDA-TiO_2@clay	degradation of methylene blue and rhodamine B	2018	1
TiO_2	degradation of nitrobenzene	2020	2
TiO_2	degradation in continuous flow	2019	3
TiO_2	dye encapsulation and release	2020	4
Janus TiO_2	degradation of kerosene or nitrobenzene	2019	5
Janus TiO_2	degradation of dye	2020	6
Graphene oxide-Fe (III)	reduction of Cr (VI)	2020	7
Graphene oxide/TiO_2	degradation of 2-naphthol	2021	8
Ag_3PO_4/graphdiyne	degradation and oxygen evolution	2019	9
Ag_3PO_4@palygorskite	degradation of tetradecane	2020	10
Polymer particles	degradation of dye	2020	11
PbS QDs	degradation of methyl orange	2022	12
JMPSN/α-FeOOH	degradation of dye	2023	13
g-C_3N_4/P25	degradation of dye	2022	14
G-C_3N_4	treatment of oily sewage	2023	15
g-C_3N_4	degradation of dye	2022	16
Pt/TiO_2	H_2 production	2021	17
Ag_2O-TiO_2/SiO_2	H_2 production	2019	18
Janus TiO_2	H_2 production	2018	19
Co_3O_4/g-C_3N_4 nanovesicles	Catalytic hydrogenation	2018	20
algal/bacterial cell	H_2 production	2020	21

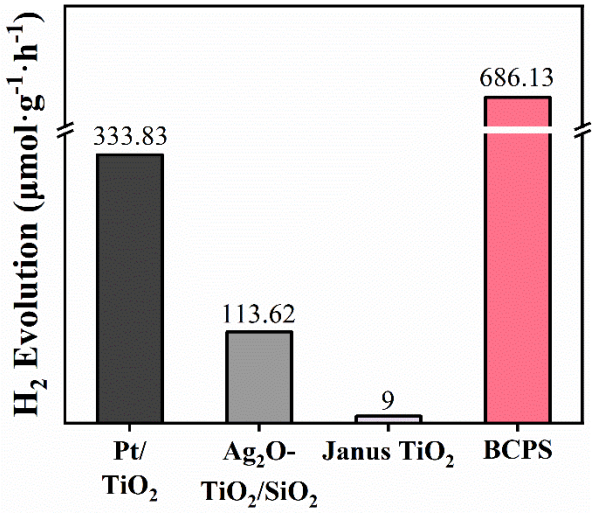

Figure S13. Comparison with other photocatalytic hydrogen production microreactors.

References

1. S. Lin, S. Sun, K. Shen, D. Tan, H. Zhang, F. Dong and X. Fu, Photocatalytic microreactors based on nano TiO2-containing clay colloidosomes, *Applied Clay Science*, 2018, **159**, 42-49.
2. N. Fessi, M. F. Nsib, Y. Chevalier, C. Guillard, F. Dappozze, A. Houas, L. Palmisano and F. Parrino, Pickering Emulsions of Fluorinated TiO2: A New Route for Intensification of Photocatalytic Degradation of Nitrobenzene, *Langmuir*, 2020, **36**, 13545-13554.
3. Q. Li, T. Zhao, M. Li, W. Li, B. Yang, D. Qin, K. Lv, X. Wang, L. Wu, X. Wu and J. Sun, One-step construction of Pickering emulsion via commercial TiO2 nanoparticles for photocatalytic dye degradation, *Applied Catalysis B: Environmental*, 2019, **249**, 1-8.
4. J. S. J. Tan, X. Cao, Y. Huang and Z. Chen, Janus-like particles prepared through partial UV irradiation at the water/oil interface and their encapsulation capabilities, *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 2020, **589**, 124460.
5. Y. Shi, Q. Zhang, Y. Liu, J. Chang and J. Guo, Preparation of amphiphilic TiO2 Janus particles with highly enhanced photocatalytic activity, *Chinese Journal of Catalysis*, 2019, **40**, 786-794.
6. Y. Zhou, F. Shen, S. Zhang, Q. Zhao, Z. Xu and H. Chen, Synthesis of Methyl-Capped TiO2–SiO2 Janus Pickering Emulsifiers for Selective Photodegradation of Water-Soluble Dyes, *ACS Applied Materials & Interfaces*, 2020, **12**, 29876-29882.
7. Z. Chen, X. Liu, C. Huang, J. Li and X. Shen, Artificial Cytochrome c Mimics: Graphene Oxide–Fe(III) Complex-Coated Molecularly Imprinted Colloidosomes for Selective Photoreduction of Highly Toxic Pollutants, *ACS Applied Materials & Interfaces*, 2020, **12**, 6615-6626.
8. Y. Lan, M. Yu, D. He, Y. Wang, Q. B. Meng, H. Huang, Y. Zhang, T. Ma and X.-M. Song, Pickering emulsion-embedded hierarchical solid-liquid hydrogel spheres for static and flow photocatalysis, *Journal of Colloid and Interface Science*, 2021, **589**, 587-596.
9. S. Guo, Y. Jiang, F. Wu, P. Yu, H. Liu, Y. Li and L. Mao, Graphdiyne-Promoted Highly Efficient Photocatalytic Activity of Graphdiyne/Silver Phosphate Pickering Emulsion Under Visible-Light Irradiation, *ACS Applied Materials & Interfaces*, 2019, **11**, 2684-2691.
10. C. Han, Y. Li, W. Wang, Y. Hou and D. Chen, Dual-functional Ag3PO4@palygorskite composite for efficient photodegradation of alkane by in situ forming Pickering emulsion photocatalytic system, *Science of The Total Environment*, 2020, **704**, 135356.
11. D. Kim, H. Kim and J. Y. Chang, Designing Internal Hierarchical Porous Networks in Polymer Monoliths that Exhibit Rapid Removal and Photocatalytic Degradation of Aromatic Pollutants, *Small*, 2020, **16**, 1907555.
12. Z. Huang, J. T. Koubek, A. Sellinger and M. C. Beard, Pickering Emulsions of Self-Assembled Lead Sulfide Quantum Dots with Janus-Ligand Shells as Nanoreactors for Photocatalytic Reactions, *ACS Applied Nano Materials*, 2022, **5**, 3183-3187.
13. B. Wang, K. Li, J. Yan and T. Zhou, Synthesis of functionalized janus hybrid nanosheets for one-step construction of pickering emulsion and selective photodegradation of water-soluble dyes, *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 2023, **664**, 131199.
14. S. Liu, S. Wang, C. Lei, R. Li, S. Feng and Q. Jin, Study of the efficiency of g-C3N4-loaded P25 for photocatalytic degradation of malachite green in aqueous and Pickering emulsion

systems, *Journal of Materials Science: Materials in Electronics*, 2022, **33**, 5846-5858.

15. B. Su, X. Ma, L. Ran, S. Wu and B. Hou, Improving the photodegradation efficiency of POPs in oily sewage: Design and synthesis of G-C3N4-based photocatalysts incorporating a pickering emulsion catalytic strategy, *Journal of Environmental Chemical Engineering*, 2023, **11**, 111297.
16. X. Zhang, H. Zhang, P. Chen, M. Liu, P. Wu, C. Liu and W. Jiang, One-step dye wastewater treatment by combined adsorption, extraction, and photocatalysis using g-C3N4 pickering emulsion, *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 2022, **644**, 128814.
17. C. Wang, W. Zhong, S. Peng, J. Zhang, R. Shu, Z. Tian, Q. Song and Y. Chen, Robust Hydrogen Production via Pickering Interfacial Catalytic Photoreforming of n-Octanol-Water Biphasic System, *Frontiers in Chemistry*, 2021, **9**.
18. E. Bu, Y. Chen, C. Wang, Z. Cheng, X. Luo, R. Shu, J. Zhang, M. Liao, Z. Jiang and Q. Song, Hydrogen production from bio-derived biphasic photoreforming over a raspberry-like amphiphilic Ag2O-TiO2/SiO2 catalyst, *Chemical Engineering Journal*, 2019, **370**, 646-657.
19. C. Wang, E. Bu, Y. Chen, Z. Cheng, J. Zhang, R. Shu and Q. Song, Enhanced photoreforming hydrogen production: Pickering interfacial catalysis from a bio-derived biphasic system, *Renewable Energy*, 2019, **134**, 113-124.