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Polymeric stabilization at the gas-liquid interface for durable solar hydrogen production from plastic waste

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Heterogeneous photocatalysis offers substantial potential for sustainable energy conversion, yet its industrial application is constrained by limited durability under stringent photochemical conditions. Achieving high photocatalytic activity often requires harsh reaction conditions, compromising catalyst stability and longevity. Here we propose a strategy involving polymeric stabilization of photocatalytic centres uniquely localized at the gas-liquid interface, substantially enhancing both the catalytic activity and stability. Applied to the photocatalytic conversion of plastic waste into solar hydrogen, this approach maintained its catalytic performance over 2 months under harsh conditions. Using 0.3 wt% dynamically stabilized atomic Pt/TiO₂ photocatalysts and concentrated sunlight, we achieved a plastic reforming activity of 271 mmolH₂ h^{-1} m⁻². Scaling to 1 m² under natural sunlight yielded a hydrogen production rate of 0.906 l per day from polyethylene terephthalate waste. Economic analysis and extensive-scale simulations suggest this strategy as a promising pathway for high-performance, durable photocatalysis, advancing renewable energy conversion.

Heterogeneous photocatalysis¹ affords a compelling pathway for advancing sustainable industrial processes², leveraging its inherently clean energy sources³, scalability^{4,5} and low operational costs⁶. It is increasingly employed for hydrogen (H_2)⁷ and hydrocarbon⁸ production, carbon dioxide capture⁹ and conversion^{10,11}, and pollution mitigation². Despite its potential, the integration of heterogeneous photocatalysis into major energy sectors^{12,13} is hampered by its relatively lower catalytic efficiency compared with conventional thermo-^{14,15}, electro-^{16,17} and photoelectro-catalytic¹⁸ methods. This inefficiency primarily results from the low input energy density and thermodynamically constrained reaction mechanisms^{4,5}. To overcome these intrinsic limitations of photocatalytic systems, various strategies have been explored, including light concentration techniques^{7,19}, the application of external heat²⁰ and the addition of electrolytes²¹ (Extended Data Fig. 1). However, these methods result in harsh operating conditions such as elevated temperatures⁷, extreme pH levels²² and high electrolyte

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Fig. 1 | An overview of polymeric stabilization at the gas-liquid interface for advanced photocatalysis. **a**, A schematic illustration of the design of highperformance durable heterogeneous photocatalysis enabled by (i) polymeric stabilization and (ii) gas-liquid interface operation. D_{air} and D_{water} denote diffusion coefficients in air and water, respectively. hv and E_g denote light energy

and band gap energy, respectively. CB, conduction band; VB, valence band. **b**, A schematic illustration of photocatalytic waste upcycling using a floatable nanocomposite system and light concentrator. **c**, A comparative analysis of the stability times of plastic waste photo-reforming reactions reported in the literature. Number sign (#) means reference numbers.

concentrations²¹, which substantially compromise the durability of the catalysts⁷ (Extended Data Fig. 1).

To address these challenges, we introduce herein a strategic approach designed to enhance the performance^{23,24} and durability²⁵ of heterogeneous photocatalysis. Recent advancements underscore the potential of polymer networks^{26,27} for enhancing catalyst protection¹⁸, thereby significantly improving the durability²⁸ and product selectivity²⁹ of photoelectro-¹⁸ and electro-catalysis^{30,31}. However, a comprehensive understanding and the effective application of polymeric stabilization in heterogeneous photocatalysis are yet to be fully established. Additionally, the polymeric^{32,33} stabilization poses a challenge owing to its potential to constrain the reactivity of photocatalysts. Therefore, we employ innovative material engineering^{34,33} and optimize reaction conditions^{36,37} to simultaneously enhance the activity^{38,39} and ensure the long-term stability⁴⁰ of the photocatalysis.

Our process involves polymeric stabilization (Fig. 1a (i)) and gas-liquid interface⁴¹ operation (Fig. 1a (ii)), substantially boosting the catalytic activity while maintaining the stability. Specifically, we demonstrate this dual approach in the photocatalytic recycling of plastic waste²² into solar H₂ (refs. 42,43) using discarded polyethylene terephthalate (PET) and polylactic acid (PLA) (Fig. 1b). Importantly, the use of polymer networks^{44,45} enhances the overall stability of photocatalytic systems^{46,47}, even under harsh conditions (Fig. 1c), as corroborated by density functional theory (DFT) calculations. The scalability of our approach has been validated through outdoor experiments on a 1 m² scale, bolstered by economic analyses at a 10 m² scale and comprehensive simulation studies at a 100 m² scale. Together, these findings highlight the substantial potential of our innovative strategy as a promising avenue for scalable, durable and productive heterogeneous photocatalysis.

Results

Overview of polymeric stabilization at the gas–liquid interface Our strategy for achieving high-performance and durable heterogeneous photocatalysis incorporates polymeric^{48,49} stabilization and gas-liquid interface operation (Fig. 1a). Dynamically stabilized atomic (DSA) platinum/titanium dioxide (Pt/TiO₂) photocatalysts⁵⁰ (0.3 wt% Pt-DSA/TiO₂) were utilized for solar-driven upcycling of PET bottles and PLA into H₂ fuel (Extended Data Fig. 2), while macroporous hydrophilic polyurethane (HPU)-polypropylene glycol (PPG) nanocomposites were adopted for constructing effective polymer networks (Extended Data Fig. 3 and Supplementary Fig. 1). The Pt-DSA/TiO₂ catalyst is known to exhibit excellent H₂ production activity via photo-reforming of plastic wastes. This high performance can be attributed to the strong and specific interactions between the deposited Pt atoms and TiO₂, facilitated by surface oxygen vacancies (O_v) created through photochemical lattice O_v migration and the simultaneous stabilization of trap states (Supplementary Fig. 2 and Supplementary Note 1). Notably, the gas-liquid interface operation is designed to augment the catalytic activity even under polymeric stabilization (Fig. 1a (i)), by promoting the efficient separation of gaseous products and thus minimizing the likelihood of reverse reactions^{51,52} (Fig. 1a (ii)).

A practical application of our approach is demonstrated for the photocatalytic H_2 production from plastic wastes, specifically targeting PET bottles and PLA (Fig. 1b). These plastic wastes are first converted into their monomeric constituents, that is, ethylene glycol (EG) and terephthalic acid (TPA) from PET and lactic acid from PLA, through a robust alkaline pretreatment process. These monomers then serve as feedstocks for H_2 production through photo-reforming (Supplementary Fig. 3). Our system employs a unique floatable elastomer–hydrogel nanocomposite integrated with a light concentrator, optimizing the light absorption (Supplementary Fig. 4) and maximizing the photocatalytic activity.

The stabilization of the photocatalysts is achieved by embedding the catalytic nanomaterials within durable hydrogel–elastomer nanocomposite networks, which are designed to withstand harsh operational conditions such as high temperatures and strong alkaline environments. This stabilization process not only enhances the resilience of our catalytic system but also substantially extends the operational longevity of the photocatalytic system. The durability of our photocatalytic process for plastic waste upcycling is shown to be approximately four times longer than the durations reported in contemporary studies (Fig. 1c). This extended stability underpins the potential of our approach to deliver a sustainable and efficient solution for energy conversion through heterogeneous photocatalysis.

DFT calculations on polymeric catalyst stabilization

At the outset of this study, we first examined how polymer networks could enhance the stability of the photocatalysts by using DFT simulations. DFT simulations were conducted to assess the potential stabilization effects provided by the HPU–PPG nanocomposite to a Pt-DSA/TiO₂ system through the formation of surface O_v. Our calculations primarily focused on the impacts of polymeric encapsulation on the energy of formation of O_v and the work function (W_f) of the catalyst lattice (Fig. 1a (i)).

The interfacial charge transfer between the polymer and TiO_2 can substantially boost its photocatalytic stability, facilitated by proper band alignment and the formation of mid-gap trap states induced by the Pt atom (Fig. 2a and Extended Data Fig. 4). Bare TiO_2 , with its large W_f , effectively separates trap states from the valence band, localizing photogenerated electrons. However, the mid-gap trap states are located far from the valence band, which is inefficient for stabilizing oxygen vacancies and associated excess electrons. In contrast, the interaction with the HPU–PPG nanocomposite reduces the W_f of TiO_2 , creating trap states near the valence band maximum. This allows photogenerated electrons to bypass the shallow traps and reach the valence band directly, requiring further activation optimization. The proximity of these trap states to the valence band, however, enhances the stabilization of excess electrons from surface oxygen vacancies, which is crucial for the stable formation of Pt-DSA/TiO₂ (ref. 50). This active excess electron relief process is experimentally supported by electrochemical impedance spectroscopy (EIS) measurements, where the Nyquist circle of Pt-DSA/TiO₂ is increased after intercalation into the HPU–PPG nanocomposites, indicating a reduction in the charge transfer ability due to the stabilization of excess electrons in oxygen vacancies (Supplementary Fig. 5).

The spin density maps of Pt-DSA/TiO₂ with various monomers reveal that HPU and PPG monomers maintain the localization of photogenerated electrons at the Pt atom (Fig. 2b). However, the HPU-PPG nanocomposite does not retain this localization within a conventional photocatalyst model, highlighting the need for enhanced interface operations, as proposed in this study. The localization of photoelectrons in the HPU-TPA pair suggests that the sufficient adsorption of electron-donating OH terminal groups in the HPU-PPG nanocomposite synergistically facilitates charge recombination, contrasting with the electron-withdrawing COOH group in TPA. A strong linear correlation was observed between the electrophilicity of the polymer terminal groups and the changes in the $W_{\rm f}$ of TiO₂ (Fig. 2c). Adsorption of HPU modestly reduced the W_f of TiO₂ (from 4.83 eV for bare Pt-DSA/TiO₂ to 4.43 eV with HPU), while HPU-PPG co-adsorption substantially lowered it to 3.91 eV. The marginal effect of the TPA unit in the HPU-TPA composite (4.57 eV) underscores that the $W_{\rm f}$ of TiO₂ is mainly influenced by the adsorption of electron-donating OH groups with strong surface interactions.

Strong interactions between TiO₂ surfaces and the nucleophilic OH in HPU–PPG are further evinced by the shorter Ti–OH bond length in PPG (2.32 Å) and HPU (2.30 Å) compared with TPA (3.12 Å) and the stronger Gibbs free energy of adsorption for PPG (–1.23 eV) and HPU (–1.28 eV) compared with TPA (–0.30 eV) (Fig. 2d). The electron density difference by adsorption also confirms that the strong adsorption of HPU and PPG involves substantial electronic interactions while the weak adsorption of TPA barely shows electronic interactions with the TiO₂ surface (Fig. 2e).

While the HPU–PPG nanocomposite lowers the photocatalytic activity in conventional models, its notable advantage lies in facilitating favourable O_v formation (Fig. 2f). Pt-DSA/TiO₂ with the HPU–PPG nanocomposite exhibits the lowest O_v energy of formation (–0.77 eV with HPU–PPG), promoting stable Pt-DSA/TiO₂ formation. The propensity for O_v formation correlates closely with electron localization at the Pt atom. Less photoelectron localization at Pt correlates with a more positive Bader charge and linearly increases the favourability of O_v formation, enabling stable Pt-DSA/TiO₂ formation. Thus, the optimal design of Pt-DSA/TiO₂ can be suggested to be preemptive stabilization with the HPU–PPG nanocomposite followed by photocatalytic activation optimization through interface operation.

The long-term stability of the nanocomposite system

Consistent with theoretical predictions illustrated in Fig. 2, immobilization of the photocatalysts within hydrophilic HPU-PPG polymer networks notably enhances the long-term stability of the nanocomposite (Fig. 3 and Extended Data Fig. 5). Unlike conventional powder systems in suspension, the nanocomposite configuration effectively mitigates the degradation typically observed in photocatalysts exposed to highly alkaline solutions (Fig. 3a). After 8-week (56 days) immersion in a strongly alkaline solution (0.7 M potassium hydroxide (KOH)), where discarded PET bottles were processed, the composite system exhibited negligible changes, as evinced by consistent optical images, scanning electron microscopy (SEM) images, Fourier-transform infrared (FT-IR) results and Raman spectroscopy data (Fig. 3b,c). Further demonstrating the efficacy of this system, no catalyst leaching was observed from the device following 2-month immersion in 0.7 M KOH solutions that solubilized plastic wastes (Fig. 3d). In stark contrast, direct exposure of the photocatalyst to reaction solutions resulted in leaching of approximately 15% of the photocatalyst. Moreover, with the high pH sustained during plastic



Fig. 2 | **DFT calculations on polymeric catalyst stabilization. a**, A schematic illustration of the band alignments and DFT band structures of Pt-DSA/ TiO₂ with large W_f (bare TiO₂ surface) and small W_f (surface co-adsorbing the HPU-PPG nanocomposite) from left to right. The fatband indicates the Pt *d* states, with up and down spin states coloured in red and blue, respectively. The Fermi level is shown by a green line. *E* denotes energy level. **b**, The spin density of the Pt-DSA/TiO₂ surface without monomer adsorbates (bare) and with monomer adsorbates (HPU, PPG, HPU-PPG and HPU-TPA) in yellow (isovalue of 0.002 e Å⁻³). **c**, The W_f (eV) of the Pt-DSA/TiO₂ surface with various monomer adsorbate units as a function of the Bader charge of a monomer unit of the

polymer, q_{poly} . **d**, The Gibbs free energy of adsorption (ΔG_{ads}) of a monomer of polymers on the TiO₂ surface. **e**, The charge density difference by adsorption of polymers, $\Delta \rho = \rho(\text{total}) - \rho(\text{TiO}_2) - \rho(\text{polymer})$ (isovalue of 0.002 e Å⁻³). Electron accumulation and depletion are shown in yellow and cyan, respectively. In **b** and **e**, the balls in colours of light blue, red, brown and whitish dark blue denote Ti, O, C, H and N atoms, respectively. **f**. The O_v energy of formation (eV) of the Pt-DSA/TiO₂ surface with various monomer adsorbate units as a function of the Bader charge of a single Pt atom (q_{pt}). All properties were estimated after the photoelectron had been generated by light irradiation.

reforming (Supplementary Fig. 6), the composite system showed negligible degradation of the photocatalytic activity, maintaining its hydrogen evolution reaction performance even after 56 days. In contrast, the powder system exhibited a notable activity reduction of 30.9% (Fig. 3e,f). The exceptional durability of the nanocomposite system, surpassing the stability durations for plastic waste photo-reforming reported in recent literature by approximately four fold (Fig. 1c and Supplementary Tables 1 and 2), highlights the feasibility of our approach for advancing sustainable photocatalysis.

Interface operation for high performance

We next investigated the photocatalytic activity of a floatable elastomer–hydrogel nanocomposite compared with traditional sunken composites and powder systems within a slurry reactor, under identical light irradiation conditions. Utilizing conditions involving 17.4 mg ml⁻¹ of PET bottles and 25 mg ml⁻¹ of PLA, the floatable composite demonstrated H₂ production rates that were two to four times greater than the powder system and five to seven times higher than the submerged composite system under air mass (AM) 1.5G conditions with identical catalyst loading per area (Fig. 4b,c and Supplementary Table 3). Notably, the H₂ production activity of the sunken nanocomposite system was lower than that of the powder system owing to the constraint on reactivity imposed by polymer networks (Fig. 4a–c and Supplementary Fig. 7). These variations in activity correlate with the DFT calculations, which suggest a polymer-induced impact on the W_f of the catalysts, thus affecting the catalytic efficiency (Figs. 1a and 2).

The enhanced performance of the floatable composite, compared with both the sunken nanocomposite and powder systems, is attributed to its operation at the gas–liquid interface, which effectively captures gaseous products and minimizes back-oxidation, thus enhancing the overall catalytic activity (Fig. 1a (ii)). The composite also underwent durability tests over five cycles in solubilized PET and PLA solutions under simulated sunlight conditions, sustaining its activity (Fig. 4d). This setup maintained consistent H_2 production in diverse media such as tap water and seawater, where PET bottles were dissolved (Fig. 4e). Notably, the floatable composite system demonstrated an apparent



Fig. 3 | **The long-term stability of the nanocomposite system. a**, Schematic illustrations of the powder system (left), showing the degradation in catalytic activity due to (i) abrasion, (ii) aggregation and (iii) leaching, and the nanocomposite system (right), highlighting the long-term stability achieved through catalyst immobilization and stabilization. The magnified view of the red box denotes TiO₂ lattice and two polymer backbone monomers. **b,c**, Optical images and SEM images and FT-IR spectroscopy data (**b**) and

Raman spectroscopy data (**c**) of the nanocomposite system before and after 56 days. **d**, **e**, The leaching percentage of Pt-DSA/TiO₂ (**d**) and the maintenance ratio of H_2 production activity (**e**) of the composite system and powder system under solubilized PET bottles in 0.7 M KOH solution (emerald) and PLA in 1 M KOH solution (orange) after 56 days. **f**, The H_2 evolution rates by the floatable composite in an aqueous solution of solubilized PET bottles over time on days 0, 7,14, 21, 28, 35, 42, 49 and 56.





Inset: the surface temperature of the composite after 1 h. PU, polyurethane. **h**, The spectrum of the wavelength of sunlight: (i) the infrared region mainly contributes to the photothermal effect and (ii) the ultraviolet and visible light region is largely involved in photocatalysis. Photo. denotes the photocatalytic layer. **i**, The H₂ evolution rates (**i**) and the time course of H₂ production (**j**) of a floatable composite from solubilized PET solution under various sun intensities and temperatures: 0.2 sun (grey, n = 3), 0.4 sun (blue, n = 3), 1 sun at 25 °C (emerald, n = 3), 1 sun at 60 °C (mint, n = 3) and 19 sun (orange, n = 3), showing that (i) the H₂ production rate at 1 sun and 60 °C was 1.97 fold higher compared with 1 sun and 25 °C and (ii) the H₂ production rate at 19 sun intensity was 19.5 fold higher compared with 1 sun and 60 °C. Data are presented as mean ± s.d. **k**, The long-term time course of H₂ production in the solubilized solutions of PET bottle under concentrated sunlight with an intensity of 19 sun at days 0, 7, 14, 21 and 28.



plastic wastes. a, The Pt-DSA/TiO₂ nanocomposites within a steel cell reactor designed for 1- m^2 -scale outdoor solar waste upcycling. Inset: photographs depicting the N₂ carrier gas, gear pump and GC equipment connected to the reactor (left) along with a top view of the nanocomposites (right). **b**, A schematic illustration delineating the steel cell reactor facility. **c**, The variation in temperature (blue), solar radiation intensity (kW m⁻², orange) and accumulated production of H₂ (ml, green) by the 1-m²-scale nanocomposites measured on 21 and 29 March 2023. **d**, The daily average temperature (blue), daily average solar radiation intensity (orange) and accumulated H₂ produced during outdoor 1-m²-scale solar H₂ production over a month (3, 14, 17, 21, 27, 29 and 31 March and 8 April). **e**, A schematic illustration of large-scale outdoor solar H₂ production.

quantum yield (AQY) of 1.80% under irradiation with light at a wavelength of 370 nm (Supplementary Fig. 8).

The operational set-up for photocatalytic waste upcycling involved the floatable hydrophilic HPU-PPG nanocomposite device integrated with a light concentrator, a solar simulator and a reactor cell (Fig. 4f.g). When equipped with a light concentrator, the device achieved a remarkable hydrogen evolution reaction rate of 271 mmol h⁻¹ m⁻² under concentrated sunlight with an intensity equivalent to 19 sun in a solution derived from solubilized PET bottles (Fig. 4h-j and Supplementary Table 3). The surface temperature of the composite increased to approximately 60 °C owing to the infrared photothermal effect, enhancing the photocatalytic activity by 1.97 times (Fig. 4i, j (i)). Furthermore, the elevated light intensity, particularly in the ultraviolet region, led to a 19.5-fold increase in activity compared with that measured under 1-sun intensity irradiation at 60 °C (Fig. 4i, j (ii)). Consequently, the concentrated light conditions enhanced the H₂ production activity by 38.4 times relative to that under 1-sun intensity at ambient temperature. Moreover, the floatable composite system exhibits superior H₂ production activity compared with the powder system even with agitation, under 19-sun light irradiation (Supplementary Fig. 9). This enhanced performance highlights the unique advantage of gas-liquid interface operation, which effectively maximizes the photothermal effect (Fig. 4h). In contrast, the powder system fails to fully exploit this effect owing to the high specific heat capacity of water, thereby limiting its overall efficiency with light concentration. Over a period of 28 days, the floatable composite sustained H₂ production from PET bottles even under intensified sunlight with illumination levels elevated to 19 sun (Fig. 4k).

Comprehensive numerical simulations for laboratory-scale solar H_2 production using a 6-cm-scale setup corroborated the superior performance of the floatable composite relative to both the conventional powder system and the sunken composite (Extended Data Fig. 6). Additionally, the simulated concentration distributions of H_2 over time (for example, at 0, 10, 30, 60, 120 and 180 min) closely aligned with experimental results, reinforcing the validity and effectiveness of the floatable composite in high-performance photocatalytic applications (Fig. 4b,c and Extended Data Fig. 6).

Large-scale and long-term solar H_2 production

To validate the scalability of our nanocomposite system for H₂ production, we initiated a two-phase experimental process. The initial phase involved scaling up the nanocomposite from 28 to 144 cm², as detailed in Extended Data Fig. 7a–c. This expanded nanocomposite subsequently achieved an H₂ production rate of 0.264 l h⁻¹ m⁻² by processing solubilized PET bottles, demonstrating its potential for larger-scale applications (Extended Data Fig. 7d). Numerical simulations further confirmed the efficiency of the floatable system over traditional sunken reaction setups, highlighting its design superiority (Extended Data Fig. 8).

Subsequently, we constructed an arrayed nanocomposite system covering 1 m² and an outdoor H₂ production facility equipped with reactor cells, quartz windows, gas chromatography (GC) and a gear pump (Fig. 5a,b and Supplementary Video 1). The bulk synthesis of the Pt-DSA/TiO₂ photocatalyst was conducted by utilizing natural sunlight for the fabrication of the nanocomposite on a 1 m² scale (Extended Data Fig. 9). This large-scale set-up achieved daily H₂ production at the litre level under natural sunlight from discarded PET bottles (Fig. 5c and Extended Data Fig. 10). On 29 March, the system produced 0.9051 of H₂, under an average light intensity of 0.636 kW m⁻² and ambient temperature of 25.5 °C (Fig. 5c). The 1 m² device maintained a steady H₂ production rate of 12.3 mmol h⁻¹ and 0.906 l per day under standard 1-sun light irradiation (Fig. 5c).

Remarkably, this litre-level H_2 production was consistently sustained over a month, from 13 March to 8 April, cumulatively generating 5.07 l of H_2 from plastic waste (Fig. 5d). The scale-up process not only enhanced the catalytic activity but also preserved the long-term stability, supported by the immobilization within durable polymer networks. Computational simulations of H_2 production at a 1 m² scale for both floatable and sunken composites further underscored the advantages of the floatable system (Supplementary Fig. 10).

Utilizing plastic waste as a feedstock for H₂ production, our approach substantially mitigates greenhouse gas (GHG) emissions by diverting plastic waste from landfills (Supplementary Note 1). Compared with steam methane reforming, the predominant industrial method for H₂ production, our photocatalytic plastic waste upcycling could reduce GHG emissions notably (Fig. 5e, Supplementary Note 2 and Supplementary Fig. 11). An economic analysis demonstrated that H₂ production from plastic waste offers greater economic value than the separation and purification of derivatives for resale (Supplementary Fig. 12. Supplementary Note 3 and Supplementary Table 3). Furthermore, the analysis revealed that daytime H₂ production by a 10 m² device could potentially meet over 20% of per capita residential electricity consumption in various countries, with notable efficiency rates ranging from 50% to 90% for Great Britain, China and South Korea (Supplementary Note 4 and Supplementary Fig. 13a). An extended simulation for a 100 m² setup illustrated the transformative industrial potential of the nanocomposite system (Supplementary Fig. 13b-f).

Conclusions

This work presents a robust strategy for enhancing the performance and durability of heterogeneous photocatalysis, leveraging polymeric stabilization and gas-liquid interface operation. Our investigations focused on the photocatalytic upcycling of plastic waste, a pressing environmental issue. Through the strategic immobilization of catalysts within elastomer-hydrogel polymer networks, our approach ensures prolonged catalytic activity even under severe operational conditions. DFT calculations revealed the mechanistic basis of this stabilization, highlighting the alignment of the mid-gap trap state with the catalyst lattice valence band and the role of oxygen vacancies. In practical terms, the efficacy of our method was substantiated through large-scale experiments, where a nanocomposite system demonstrated its capability to reform plastic waste into H₂ gas at a scale of 1 m² under natural sunlight conditions. This system consistently achieved H₂ production at litre levels from discarded PET bottles. Additionally, economic evaluations at a 10 m² scale and comprehensive simulation studies at a 100 m² scale further affirm the economic viability and scalability of this photocatalytic approach. Collectively, these results underscore the transformative potential of our strategy, not only for advancing sustainable fuel production but also for contributing to an economically sustainable energy framework. This study paves the way for broader application and integration of heterogeneous photocatalysis in the energy sector, offering a viable path forwards in the transition to a greener and more sustainable energy landscape.

Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41565-025-01957-6.

References

- 1. Schultz, D. M. & Yoon, T. P. Solar synthesis: prospects in visible light photocatalysis. *Science* **343**, 1239176 (2014).
- 2. Hoffmann, M. R., Martin, S. T., Choi, W. & Bahnemann, D. W. Environmental applications of semiconductor photocatalysis. *Chem. Rev.* **95**, 69–96 (1995).
- Pinaud, B. A. et al. Technical and economic feasibility of centralized facilities for solar hydrogen production via photocatalysis and photoelectrochemistry. *Energy Environ. Sci.* 6, 1983–2002 (2013).

Article

- 4. Nishiyama, H. et al. Photocatalytic solar hydrogen production from water on a 100-m² scale. *Nature* **598**, 304–307 (2021).
- Goto, Y. et al. A particulate photocatalyst water-splitting panel for large-scale solar hydrogen generation. *Joule* 2, 509–520 (2018).
- 6. She, Z. W. et al. Combining theory and experiment in electrocatalysis: insights into materials design. *Science* **355**, eaad4998 (2017).
- 7. Zhou, P. et al. Solar-to-hydrogen efficiency of more than 9% in photocatalytic water splitting. *Nature* **613**, 66–70 (2023).
- 8. Zhou, B. et al. Light-driven synthesis of C_2H_6 from CO_2 and H_2O on a bimetallic Aulr composite supported on InGaN nanowires. *Nat. Catal.* **6**, 987–995 (2023).
- Kim, S. et al. Asymmetric chloride-mediated electrochemical process for CO₂ removal from oceanwater. *Energy Environ. Sci.* 16, 2030–2044 (2023).
- Chen, X. et al. Electrochemical CO₂-to-ethylene conversion on polyamine-incorporated Cu electrodes. *Nat. Catal.* 4, 20–27 (2021).
- Mariano, R. G., McKelvey, K., White, H. S. & Kanan, M. W. Selective increase in CO₂ electroreduction activity at grain-boundary surface terminations. *Science* **358**, 1187–1192 (2017).
- 12. Cargnello, M., Gordon, T. R. & Murray, C. B. Solution-phase synthesis of titanium dioxide nanoparticles and nanocrystals. *Chem. Rev.* **114**, 9319–9345 (2014).
- Gewirth, A. A., Varnell, J. A. & Diascro, A. M. Nonprecious metal catalysts for oxygen reduction in heterogeneous aqueous systems. *Chem. Rev.* **118**, 2313–2339 (2018).
- Banerjee, A., Dick, G. R., Yoshino, T. & Kanan, M. W. Carbon dioxide utilization via carbonate-promoted C–H carboxylation. *Nature* 531, 215–219 (2016).
- Huang, W. et al. Steam-created grain boundaries for methane C-H activation in palladium catalysts. Science **373**, 1518–1523 (2021).
- Kowal, A. et al. Ternary Pt/Rh/SnO₂ electrocatalysts for oxidizing ethanol to CO₂. Nat. Mater. 8, 325–330 (2009).
- Xiao, F. et al. Recent advances in electrocatalysts for proton exchange membrane fuel cells and alkaline membrane fuel cells. *Adv. Mater.* 33, 1–38 (2021).
- Tan, J. et al. Hydrogel protection strategy to stabilize water-splitting photoelectrodes. *Nat. Energy* 7, 537–547 (2022).
- Wang, Q., Pornrungroj, C., Linley, S. & Reisner, E. Strategies to improve light utilization in solar fuel synthesis. *Nat. Energy* 7, 13–24 (2022).
- 20. Xu, C., Tang, Q., Tu, W. & Wang, L. Photon and phonon powered photothermal catalysis. *Energy Environ. Sci.* **17**, 4461–4480 (2024).
- 21. Li, Y. et al. Electrolyte-assisted polarization leading to enhanced charge separation and solar-to-hydrogen conversion efficiency of seawater splitting. *Nat. Catal.* **7**, 77–88 (2024).
- Uekert, T., Kuehnel, M. F., Wakerley, D. W. & Reisner, E. Plastic waste as a feedstock for solar-driven H₂ generation. *Energy Environ. Sci.* **11**, 2853–2857 (2018).
- 23. Chen, C. et al. Highly crystalline multimetallic nanoframes with three-dimensional electrocatalytic surfaces. Science **343**, 1339–1343 (2014).
- 24. Zang, J. et al. Multifunctionality and control of the crumpling and unfolding of large-area graphene. *Nat. Mater.* **12**, 321–325 (2013).
- Povia, M. et al. Operando X-ray characterization of high surface area iridium oxides to decouple their activity losses for the oxygen evolution reaction. *Energy Environ. Sci.* **12**, 3038–3052 (2019).
- Kao, J., Thorkelsson, K., Bai, P., Rancatore, B. J. & Xu, T. Toward functional nanocomposites: taking the best of nanoparticles, polymers, and small molecules. *Chem. Soc. Rev.* 42, 2654–2678 (2013).

- Vargo, E. et al. Functional composites by programming entropy-driven nanosheet growth. *Nature* 623, 724–731 (2023).
- 28. Wang, D. et al. Structurally ordered intermetallic platinum-cobalt core-shell nanoparticles with enhanced activity and stability as oxygen reduction electrocatalysts. *Nat. Mater.* **12**, 81–87 (2013).
- 29. Lang, X., Chen, X. & Zhao, J. Heterogeneous visible light photocatalysis for selective organic transformations. *Chem. Soc. Rev.* **43**, 473–486 (2014).
- 30. Li, Z. et al. Iridium single-atom catalyst on nitrogen-doped carbon for formic acid oxidation synthesized using a general host-guest strategy. *Nat. Chem.* **12**, 764–772 (2020).
- Wang, J. et al. Synergistic effect of well-defined dual sites boosting the oxygen reduction reaction. *Energy Environ. Sci.* 11, 3375–3379 (2018).
- Oh, J. Y. et al. Intrinsically stretchable and healable semiconducting polymer for organic transistors. *Nature* 539, 411–415 (2016).
- Paxson, A. T., Yagüe, J. L., Gleason, K. K. & Varanasi, K. K. Stable dropwise condensation for enhancing heat transfer via the initiated chemical vapor deposition (iCVD) of grafted polymer films. *Adv. Mater.* 26, 418–423 (2014).
- Mao, X. et al. Self-assembled nanostructures in ionic liquids facilitate charge storage at electrified interfaces. *Nat. Mater.* 18, 1350–1357 (2019).
- Herrero, E., Buller, L. J. & Abruña, H. D. Underpotential deposition at single crystal surfaces of Au, Pt, Ag and other materials. *Chem. Rev.* 101, 1897–1930 (2001).
- Dash, S., de Ruiter, J. & Varanasi, K. K. Photothermal trap utilizing solar illumination for ice mitigation. Sci. Adv. 4, 1–7 (2018).
- Tang, Y., Zhang, Y., Li, W., Ma, B. & Chen, X. Rational material design for ultrafast rechargeable lithium-ion batteries. *Chem.* Soc. Rev. 44, 5926–5940 (2015).
- 38. Hochbaum, A. I. et al. Enhanced thermoelectric performance of rough silicon nanowires. *Nature* **451**, 163–167 (2008).
- Fabbri, E. et al. Dynamic surface self-reconstruction is the key of highly active perovskite nano-electrocatalysts for water splitting. *Nat. Mater.* 16, 925–931 (2017).
- Goodman, E. D. et al. Catalyst deactivation via decomposition into single atoms and the role of metal loading. *Nat. Catal.* 2, 748–755 (2019).
- 41. Dong, A., Chen, J., Vora, P. M., Kikkawa, J. M. & Murray, C. B. Binary nanocrystal superlattice membranes self-assembled at the liquid-air interface. *Nature* **466**, 474–477 (2010).
- 42. Wang, Y. et al. Current understanding and challenges of solar-driven hydrogen generation using polymeric photocatalysts. *Nat. Energy* **4**, 746–760 (2019).
- Moniz, S. J. A., Shevlin, S. A., Martin, D. J., Guo, Z. X. & Tang, J. Visible-light driven heterojunction photocatalysts for water splitting-a critical review. *Energy Environ. Sci.* 8, 731–759 (2015).
- 44. Li, C. H. et al. A highly stretchable autonomous self-healing elastomer. *Nat. Chem.* **8**, 618–624 (2016).
- Kim, Y., Yuk, H., Zhao, R., Chester, S. A. & Zhao, X. Printing ferromagnetic domains for untethered fast-transforming soft materials. *Nature* 558, 274–279 (2018).
- Zheng, Y., Liu, J., Liang, J., Jaroniec, M. & Qiao, S. Z. Graphitic carbon nitride materials: controllable synthesis and applications in fuel cells and photocatalysis. *Energy Environ. Sci.* 5, 6717–6731 (2012).
- Ran, J., Ma, T. Y., Gao, G., Du, X. W. & Qiao, S. Z. Porous P-doped graphitic carbon nitride nanosheets for synergistically enhanced visible-light photocatalytic H₂ production. *Energy Environ. Sci.* 8, 3708–3717 (2015).
- 48. Liu, W. et al. High-efficiency stretchable light-emitting polymers from thermally activated delayed fluorescence. *Nat. Mater.* **22**, 737–745 (2023).

Article

- 49. Dai, Y. et al. Stretchable redox-active semiconducting polymers for high-performance organic electrochemical transistors. *Adv. Mater.* **34**, 2201178 (2022).
- 50. Lee, C. W. et al. Photochemical tuning of dynamic defects for high-performance atomically dispersed catalysts. *Nat. Mater.* **23**, 552–559 (2024).
- Lee, W. H. et al. Floatable photocatalytic hydrogel nanocomposites for large-scale solar hydrogen production. *Nat. Nanotechnol.* 18, 754–762 (2023).
- Lee, W. H. et al. Highly efficient nitrogen-fixing microbial hydrogel device for sustainable solar hydrogen production. *Adv. Mater.* 35, 2306092 (2023).

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Methods Materials

Materials

HPU was purchased from Aekyung Chemical. PLA was purchased from Goodfellow. TiO₂ (anatase, <25 nm), chloroplatinic(IV) acid solution (H₂PtCl₆, 8 wt% in H₂O), 3-(trimethylsilyl)-1-propanesulfonic acid sodium salt (97.00%), dimethyl sulfoxide (99.90%) and deuterium oxide (D₂O, 99.00 at.% D) were purchased from Sigma Aldrich. Acetone, ethyl alcohol (EtOH), methyl alcohol (MeOH), KOH, sodium chloride (NaCl) and PPG were purchased from Samchun Chemicals.

Pretreatment of plastic wastes

After slicing and grinding of discarded PET bottles, the obtained PET flakes were subject to hydrolysis in 10.5 M KOH solution under ambient conditions with continuous agitation for 7 days. Subsequently, the resultant solution under went dilution with DI water, tap water and seawater to attain a concentration of 0.7 M KOH and 17.4 mg ml⁻¹. Similarly, PLA powder underwent hydrolysis in 1 M KOH solution, maintained at a concentration of 12.5 mg ml⁻¹, and subjected to stirring for 7 days.

Synthesis of Pt-DSA/TiO₂

Pt-DSA/TiO₂ was synthesized by a slightly modified procedure based on a previously reported method⁵⁰. Commercially available anatase TiO₂ (2g) was dispersed in 150 ml of 20% methanol aqueous solution within a 250-ml glass bottle, using sonication for 10 min. The opening of the glass bottle was then plugged with a rubber septum and tightly sealed using Teflon tape and parafilm. H_2PtCl_6 (1 ml) was mixed with 9 ml of H₂O to make 10 ml of aqueous Pt precursor solution. The TiO₂ dispersion and Pt precursor solution underwent 10-min purging with Ar gas. The TiO₂ dispersion was vigorously stirred while being exposed to ultraviolet-visible light for 1 h, with an intensity of 100 mW cm⁻², utilizing a xenon lamp light source (Newport). After the TiO₂ dispersion turned blueish, the Pt precursor solution was injected into it while vigorously stirring. Following 20 min of stirring, the reaction solution was centrifuged and then washed with water and ethanol. The product was dried overnight in an electric oven at 50 °C and then utilized for subsequent experiments.

DFT calculations

DFT calculations were performed using the Vienna Ab initio Simulation Package 5.4.4 (ref. 53). The Perdew–Burke–Ernzerhof functional⁵⁴ was employed to describe the reactions on the Pt-DSA/TiO₂ surface, and Grimme's D3 method⁵⁵ was applied for van der Waals correction. The projector augmented wave pseudopotentials⁵⁶ were used with a cut-off energy of 450 eV. All models were relaxed by applying **k**-points $(3 \times 2 \times 1)$ with the Monkhorst–Pack scheme. The vacuum slab was set as 25 Å thick. Both a bare slab and the corresponding reaction were optimized by fixing the first and second bottom layers of the Pt-DSA/TiO₂. The structures were minimized until the maximum atomic force became less than 0.1 eV Å⁻¹. The charge density difference was visualized by using VESTA 3.5.8 (ref. 57).

Material characterization

- Quantification of the Pt nanoparticle concentration was conducted at Seoul National University's National Center for Inter-University Research Facilities (NCIRF) by employing an inductively coupled plasma atomic emission spectrometer (ICPS, ICPS-8100).
- (2) Assessment of the supporting layer and bilayer density involved measuring the equilibrium density. The floatable composite, featuring a bilayer structure along with solely the supporting layer, was immersed in DI water, characterized by a density of 1 g ml^{-1} . Subsequently, acetonitrile with density of 0.786 g ml⁻¹ was incrementally added to the solution to achieve density adjustment. The point of hydrogel submersion denoted the

determination of hydrogel density, corresponding to the density of the ambient solution.

- (3) Evaluation of water mass transfer through the composite entailed quantifying water evaporation. The weight disparity of water, indicative of the evaporated amount, was determined by assessing the quantity of evaporated water through the composite within a vial after incubating for 24 h in a 70 °C oven. Additionally, control experiments involved measuring the quantities of evaporated water in both open and closed states, serving as positive and negative controls, respectively.
- (4) Analysis of the Pt leaching extent was conducted by measuring the Pt concentration in the solution after approximately 2 months. The floatable composite containing 150 mg of embedded catalyst, alongside the sunken catalyst, was separately immersed in 75 ml of a solubilized solution of PET bottle and PLA within glass vials. After 2 months, the solubilized Pt concentration in the solution was determined using ICPS-MS (PlasmaQuant MS Elite, analytik jena). Sample preparation involved neutralizing the solution with hydrochloric acid, filtering it through a 0.45-µm cellulose syringe filter to obtain a homogeneous solution and subsequently subjecting it to aqua regia treatment.

Atomic-resolution imaging for Cs-corrected STEM and EDS mapping measurements

Atomic-resolution imaging of the Pt-DSA/TiO₂ catalyst was performed at 200 kV with a spherical aberration-corrected JEM ARM-200F microscope (Cold FEG Type, JEOL) at the NCIRF. Energy-dispersive X-ray spectroscopy (EDS) measurements were carried out in scanning transmission electron microscopy (STEM) mode using a single drift detector (X-MaxN, Oxford Instruments).

X-ray absorption fine structure measurements

X-ray absorption fine structure (XAFS) data were collected at the 8C and 10C nano XAFS beamlines (BL8C and BL10C) of the Pohang Light Source (PLS-II) in the 3.0-GeV storage ring, operating at a ring current of 300 mA in top-up mode. A Si(111) crystal served as the monochromator, and the beam intensity was reduced by 30% to eliminate high-order harmonics. XAFS spectra were obtained using both transmission and fluorescence modes.

X-ray photoelectron spectroscopy measurements

The X-ray photoelectron spectroscopy (XPS) data for the Pt-DSA/TiO₂ photocatalyst were measured using the K-Alpha⁺ XPS system (Thermo Fisher Scientific) at the Busan Center of the Korea Basic Science Institute. Monochromated Al K α radiation served as the X-ray source.

Fabrication of the floatable photocatalytic elastomerhydrogel composite

The floatable composite comprised a bilayer structure consisting of the photocatalytic layer and the supporting layer. To fabricate the photocatalytic layer, EtOH was mixed with a previously prepared HPU solution (HPU in water, 50 wt%) to achieve a 10.5 wt% concentration of HPU. NaCl was added to the HPU solution to reach a NaCl concentration of 0.032 g ml⁻¹. Afterwards, Pt-DSA/TiO₂ photocatalysts were mixed into the HPU solution with vigorous stirring, followed by the addition of PPG, which was twice the volume of the HPU solution. A mixed sol–gel phase was prepared, and after 24 h, only the gel phase was isolated. This gel was then compressed into a mould and dried to evaporate EtOH and water at 60 °C for 1 day. The dried gel was then reswollen with water to remove NaCl inside the polymer network, which gives macroporosity to the elastomer–hydrogel hybrid. The supporting layer was fabricated by following the same procedures except for the addition of photocatalysts. Both layers were integrated by wetting the top side of the supporting layers with a few drops of EtOH. To remove EtOH through evaporation, the final integrated floatable composite was dried at 60 °C for 24 h and re-swollen in water. As the size expanded, the necessary materials also increased. The specific values are presented in Supplementary Table 5.

Electron paramagnetic resonance measurements

Electron paramagnetic resonance (EPR) measurements were performed at 150 K using a Bruker EMXmicro system with an X-band frequency of 9.42 GHz. The set-up featured a magnetic field modulation of 100 kHz and a modulation amplitude of 1.0 G. For the analysis, 20 mg of anatase TiO_2 and Pt-DSA/ TiO_2 powders were separately transferred into EPR tubes and measured at 150 K. To measure the signal from irradiated TiO_2 , 20 mg of anatase TiO_2 was placed in an EPR tube, purged with argon and securely sealed. After 30 min of light irradiation on the tube, an EPR measurement was performed at 150 K.

Photoluminescence spectroscopy measurements

Steady-state photoluminescence (PL) and time-resolved PL spectra were measured using a Floutime 300 spectrometer (PicoQuant) with an excitation laser wavelength of 265 nm. Time-resolved PL spectra were acquired at a wavelength of 480 nm. All measurements were conducted in an aqueous dispersion under an unaerated atmosphere, achieved by argon purging in gas-tight PL cuvettes.

¹H nuclear magnetic resonance measurements

The ¹H nuclear magnetic resonance spectra were recorded on a 600-MHz high-resolution nuclear magnetic resonance spectrometer (Bruker Avance 600) at the National Instrumentation Center for Environmental Management. Dimethyl sulfoxide and 3-(trimethylsilyl)-1-propanesulfonic acid sodium salt served as internal standards, with D_2O used as the solvent.

EIS measurements

EIS measurements were conducted using a PGSTAT302N potentiostat (Autolab). A standard three-electrode setup was employed. Specifically, nitrogen-saturated 0.1 M Na₂SO₄ was used as the electrolyte, with a platinum plate serving as the counter electrode and a silver/silver chloride (Ag/AgCl) electrode filled with 3 M potassium chloride (KCl) used as the reference electrode. To create the working electrode, 5 mg of the catalyst was mixed with 2.5 ml of ethanol, followed by the addition of 10 µl of 5 wt% Nafion solution. The mixture was sonicated for 15 min to ensure a smooth, consistent suspension. This catalyst ink was then applied onto a glassy carbon electrode (5 mm diameter) through a drop-casting method, achieving a mass loading of 0.040 mg cm⁻². Following drying at room temperature, EIS measurements were conducted at a bias potential of -1.3 V, spanning the frequency range from 10^{-1} to 10^{5} Hz in the dark.

pH measurements

The time course of the pH variation during H_2 production in the solubilized PET solution by the floatable composite under 1-sun irradiation was measured using a pH electrode (Sartorius, PB-30), with samples being taken and measured at 1-h intervals.

Photocatalytic measurements

General procedure for measurements of photocatalytic H₂ production. The measurements of photocatalytic H₂ production were conducted within a sealed quartz cell reactor. After purging the sealed reaction system with N₂ gas for 20 min, simulated sunlight was directed onto the upper surface of the floatable composite utilizing an ASTM E 927-05 solar simulator (Newport), operating at 20, 40 and 100 mW cm⁻² (AM 1.5G condition). The light intensity was quantified by employing a detector fitted with an F150A-BB-26 thermopile sensor (Ophir). Temperature regulation was maintained via a hot plate throughout the reaction. The quantity of H_2 generation was determined by utilizing an iGC7200A gas chromatography system (DS Science) equipped with a thermal conductivity detector and utilizing N_2 gas as the carrier gas. For the sunken composite, the assessment of photocatalytic H_2 production was executed under conditions analogous to the floatable composite, with the sole distinction being its complete submersion to the bottom of the solution. The catalyst loading for laboratory-scale experiments for both composite and powder systems was 5.31 mg cm⁻². The surface area and irradiation area of the composite system was 28.3 cm², and that of the powder system was 12.8 cm².

A Fresnel lens measuring $10 \times 10 \text{ cm}^2$ was employed to concentrate the simulated sunlight into a circular configuration with a diameter of 1.3 cm. The surface of the floatable composite was irradiated with concentrated simulated sunlight at an intensity of 1,900 mW cm⁻². The intensity was measured using an F150A-BB-26 thermopile sensor (Ophir).

Laboratory-scale PET photo-reforming measurements were conducted a minimum of three times, and the error bars were derived by averaging the results from these three separate measurements.

Measurements of photocatalytic H₂ production of the scalable-size floatable composite. A floatable composite of scalable dimensions was positioned atop a solution of solubilized waste derived from PET bottles within a reactor cell measuring $14.5 \times 14.5 \times 5$ cm³. After the placement of the floatable composite and the solubilized PET bottle waste solution into the reactor cell, the cell was sealed by affixing a quartz window utilizing a silicone sealant. Following this, the sealed reaction system underwent purging with N₂ gas for 1 h, after which simulated sunlight was directed onto the upper surface of the floatable composite, employing an ASTM E 927-05 solar simulator (Newport) operating at an intensity of 100 mW cm⁻² (AM 1.5G condition). The irradiated simulated sunlight was projected into a square shape with size of 8.5×8.5 cm² on the scalable-size floatable composite.

Measurements of photocatalytic H₂ production of the large-scale floatable composites. An array of 16 floatable nanocomposites, each measuring 25 × 25 cm², embedding Pt-DSA/TiO₂ photocatalysts with a cumulative area of 1 m² was placed on the solubilized waste solution of PET bottles inside a 113 × 113 × 7 cm³ steel cell reactor. After positioning the 16 nanocomposites and the solubilized PET bottle waste solution within the reactor cell, the cell was sealed by applying four quartz windows, each measuring 51 × 51 cm², utilizing a silicone sealant. Following this, the sealed reaction system underwent purging with N₂ gas for 6 h, after which the samples were subjected to irradiation with natural sunlight. Throughout the measurement process, fluctuations in solar radiation intensity and temperature were monitored by employing a Nova II energy meter (Ophir) and a Poseidon2 3266 automatic thermometer (HW group), respectively.

AQY measurements

The AQY measurements were conducted following the same procedure as the photocatalytic measurements, with the exception that the solar simulator was replaced by a Kessil PR160L LED light source (wavelengths of 370 nm, 390 nm and 427 nm). The AQY was calculated using the equation below, and the measurements were performed under a light intensity of 100 mA cm⁻². The equation for AQY includes the following parameters: $n(H_2)$ represents the number of moles of H_2 , N_A denotes Avogadro's number, h is Planck's constant and c represents the speed of light. *P* indicates the light intensity, and *S* is the illuminated area (28.3 cm²). t corresponds to the irradiation time, and λ represents the wavelength of the light.

$$AQY(\%) = \frac{2 \times n(H_2) \times N_A \times h \times c}{P \times S \times t \times \lambda} \times 100.$$

Numerical simulation

We set up a two-dimensional simulation domain, which has the same length scale as in experiments. In the simulation, H_2 is generated at a constant flux of 7.86 mmol $m^{-2} h^{-1}$ from the composite and 20 mmol $m^{-2} h^{-1}$ from the catalyst. For the composite, the H_2 is generated only at the top 0.1-cm-thickness box. The H_2 diffuses to water and air, following the equation

$$\frac{\partial c}{\partial t} = D\nabla^2 c + R,\tag{1}$$

where *c* is the H₂ concentration, *t* is time and *D* is the diffusion coefficient of H₂. The diffusion coefficient of H₂ is chosen as 5×10^{-5} cm² s⁻¹ in water⁵⁸ and 0.7 cm² s⁻¹ in air⁵⁹. *R* includes the generation and the reverse reaction rate of H₂. We assume that the reverse reaction rate is proportional to the H₂ concentration, $R = -k_rc$, where k_r is the rate constant of the reverse reaction and is chosen as 1×10^{-5} s⁻¹. Note that the generation of H₂ occurs at the composite or catalyst, and the reverse reaction occurs in the water.

The simulation domain is discretized into 8,000 elements using the commercial software COMSOL Multiphysics 5.4 (COMSOL). The conservation equation is numerically solved using the finite element method solver in COMSOL. A workstation with 2 octa-core processors (Intel Xeon CPUs E5-2687W 3.1 GHz) and 192 GB memory is used for the numerical calculation.

GHG emission calculation and economic analysis

GHG emissions stemming from the steam methane reforming and plastic waste photo-reforming processes were evaluated, incorporating the reaction mechanism governing EG decomposition ($C_2H_6O_2 + 2 H_2O \rightarrow 2CO_2 + 5H_2$) and TPA decomposition ($C_8H_6O_4 + 12H_2O \rightarrow 15H_2 + 8CO_2$). The total GHG emissions are computed by deducting the mitigated GHG emissions from the operational GHG emissions (Supplementary Note 1).

The data sources utilized for electricity consumption per capita are delineated in Supplementary Table 6.

Statistics

The experimental results including the porosity, catalyst leaching, elastic modulus, swelling ratio, contact angle and H₂ evolution rate were compared and evaluated using the independent samples *t*-test.

Data availability

All data that support the findings of this study are available from the corresponding authors upon reasonable request. Source data are provided with this paper.

References

 Kresse, G. & Furthmüller, J. Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set. *Comput. Mater. Sci.* 6, 15–50 (1996).

- 54. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized gradient approximation made simple. *Phys. Rev. Lett.* **77**, 3865–3868 (1996).
- Grimme, S., Ehrlich, S. & Goerigk, L. Effect of the damping function in dispersion corrected density functional theory. *J. Comput. Chem.* **32**, 1456–1465 (2011).
- 56. Blöchl, P. E. Projector augmented-wave method. *Phys. Rev. B* **50**, 17953–17979 (1994).
- Momma, K. & Izumi, F. VESTA3 for three-dimensional visualization of crystal, volumetric and morphology data. J. Appl. Crystallogr. 44, 1272–1276 (2011).
- Zhao, X. & Jin, H. Investigation of hydrogen diffusion in supercritical water: a molecular dynamics simulation study. *Int. J. Heat. Mass Transf.* 133, 718–728 (2019).
- Matsunaga, N., Hori, M. & Nagashima, A. Measurement of mutual diffusion coefficients of gases by the Taylor method: measurements on H₂-air, H₂-N₂, and H₂-O₂ systems. *Heat. Transf. Asian Res.* **31**, 182–193 (2002).

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Author contributions

W.H.L., H.P., C.W.L., H.K., M.K., T.H. and D.-H.K. conceived of the ideas, designed the experiments and wrote the paper. W.H.L., H.P., C.W.L., H.K., J.I.Y., J.H., G.D.C., M.S.B., B.-H.L., J.R., T.H. and D.-H.K. performed experiments and data analysis. J.H.J., S.-U.B., K.H.A. and M.K. performed computer simulations.

Competing interests

The authors declare no competing interests.

Additional information

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High-performance durable photocatalysis



Extended Data Fig. 1 | **Polymeric stabilization for durable photocatalysis.** Various strategies including light concentration technique, electrolyte addition, and external heating have been explored to enhance photocatalytic activity. However, these methods impose harsh operating conditions which substantially compromise catalyst durability. Our strategy improves both catalytic stability and activity through polymeric stabilization at the gas-liquid interface.

Article





 PtO_2 , and a reference Pt foil. **d**, XPS spectrum of Pt-DSA/TiO₂, with the pink line indicating Al 2p from residual Al in commercial anatase TiO₂. **e**, XANES spectra of Pt-DSA/TiO₂. The Pt loading amount is 0.3 wt% for all samples.



Extended Data Fig. 3 | **Fabrication process and material characterization of the composite. a**, Schematic illustrations (top row) and photographs (bottom row) of the composite fabrication process. (i) Formation of an HPU-PPG-NaCl gel embedding Pt-DSA/TiO₂ photocatalysts. (ii) Sequential drying and re-swelling procedures culminating in the establishment of a photocatalytic layer. The dissolution of NaCl granules in water facilitates the formation of macroporosity within the photocatalytic layer. (iii) Integration of the photocatalytic layer onto the prepared supporting layer to fabricate the bilayer composite. **b**, The density of the supporting layer (white; n = 3) and bilayer composite (blue; n = 3). Data are presented as mean values +/- standard deviation (SD). **c**, **d**, (**c**) Schematic illustrations and (**d**) graph delineating the residual water content after evaporation for 24 h within an oven set at 70 °C, contrasting between the open (n = 4) and closed (n = 5) systems (control groups) and the bilayer composite (experimental group; n = 5). Data are presented as mean values +/- SD. **e**, Porosity of the supporting layer (white; n = 3) and bilayer composite (blue; n = 3). Data are presented as mean values +/- SD.



Extended Data Fig. 4 | **Fatband structure of Pt d states in Pt-DSA/TiO₂ with a monomer unit of various polymer composites.** Band structures of Pt-DSA/TiO₂ with various monomer units on the surface: from left to right, bare, HPU, PPG, TPA, HPU-PPG, and PPG-PPG respectively. The fatband indicates the Pt d states with up and down spin states colored in red and blue, respectively. The Fermi level is shown in olive lines.



Extended Data Fig. 5 | **Long-term stable photocatalytic plastic waste upcycling. a**, Optical images of a floatable composite in a quartz cell connected to GC in the absence of simulated sunlight (left) and in the presence of simulated sunlight. **b**, **c**, (**b**) Long-term time course of H_2 production and (**c**) H_2 evolution rates in the solubilized solutions of PET bottle and PLA under one-sun irradiation (*for example*, day 0, day 7, day 14, day 21, and day 28).



Extended Data Fig. 6 | **Computational simulation of lab-scale H₂ production. a**, **b**, Schematic illustration of the lab-scale simulation domain of (**a**) floatable composite and (**b**) sunken catalyst. **c**, **d**, (**c**) The distribution of H₂ concentration during H₂ production and (**d**) H₂ concentration as a function of depth (z at various time points by floatable composite. **e**, **f**, (**e**) The distribution of H₂ concentration

during H₂ production and (**f**) H₂ concentration as a function of depth (z at various time points by powder system. **g**, **h**, (**g**) The distribution of H₂ concentration during H₂ production and (**h**) H₂ concentration as a function of depth (z at various time points by sunken composite.



Extended Data Fig. 7 | **Scalable photocatalytic upcycling of discarded PET bottles. a**, Optical image of the scalable-size floatable composite in a solubilized waste solution of PET bottles in a reactor cell. **b**, Optical image of a side view of the scalable-size floatable composite. **c**, Schematic illustration of an overview

of scalable H_2 -generating facility primarily comprising a reactor, gas, gas chromatograph, solar simulator, and computer. **d**, Time course of H_2 production of the scalable-size floatable composite in the solubilized waste solution of PET bottle.



Extended Data Fig. 8 | **Computational simulation of scalable H**₂ **production. a**, **b**, Schematic illustration of the scalable-size simulation domain of (**a**) floatable composite and (**b**) powder system. **c**, **d**, The distributions of H₂ concentration during H₂ production of (**c**) floatable composite and (**d**) powder system at various time points (*for example*, 0, 100, 500, 1000, and 1500 min). **e**, **f**, H₂ concentration

of (e) floatable composite and (f) powder system as a function of depth (z) at various time points: 0 min (black), 100 min (purple), 500 min (blue), 1000 min (green), and 1500 min (red). g, h, (g) The distribution of H₂ concentration during H₂ production and (h) H₂ concentration as a function of depth (z) at various time points by sunken composite.

Bulk synthesis of Pt-DSA/TiO₂ photocatalyst



Extended Data Fig. 9 | **Bulk synthesis of Pt-DSA/TiO₂ photocatalyst.** Optical images of the front view (top) and top view (bottom) of the outdoor setup for the bulk synthesis of Pt-DSA/TiO₂ photocatalyst in the quartz cell during (i) N_2 purging, (ii) irradiation of natural sunlight, and (iii) addition of Pt precursor.



Extended Data Fig. 10 | 1 m²-scale solar H₂ production. a – f, Variations in solar radiation intensity (orange), temperature (green), and the H₂ production rate by the 1 m²-scale floatable composite (navy blue) measured on (a) 13th, (b) 14th, (c) 17th (d) 27th, and (e) 31st of Mar. and (e) 8th of Apr. in 2023.