Supporting Information

Acid-resistant bilayer MXene/attapulgite-g-C_3N_4/BiOBr aerogel for high-efficiency solar-driven interfacial evaporation and photocatalytic water treatment

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1. Synthesis and preparation of materials

1.1 Purification and acidification of attapulgite

First, 15.00 g of PAL was dispersed in 300 mL of deionized water, followed by the addition of 0.75 g of $(NaPO_3)_6$ with continuous stirring for 12 h. Upon cessation of agitation, the resulting suspension was permitted to settle until phase separation was achieved, after which the lower sediment layer was carefully decanted and discarded. The remaining suspension was slowly acidified with 7.5 mL of HCl and maintained at room temperature for 13 h under static conditions. The treated suspension was then centrifuged at 5000 rpm for 10 min to separate solid components from the liquid medium. The collected solids underwent repeated washing with deionized water until the supernatant reached neutral pH (pH≈7.0, measured by pH meter). Finally, the purified material was dried in an oven at 68°C for 24 h and mechanically ground in an agate mortar to achieve particle sizes ≤75 μm (corresponding to 200-mesh sieve specification), yielding the attapulgite product.

1.2 Preparation of the MXene

Ti_3C_2 powder was synthesized via hydrofluoric acid etching using the following optimized procedure. Initially, 15.0 mL of HCl and 5.0 mL of deionized water were transferred into a 50 mL centrifuge tube, which was continuously agitated at 41°C. Subsequently, 1.00 g of LiF was gradually introduced into the centrifuge tube through sequential batch additions. Next, 1.00 g of Ti_3AlC_2 precursor was slowly added to the reaction mixture to avoid excessive gas evolution and temperature rise. The centrifuge tube was sealed with a pierced cap to allow gas release during the 48 h reaction period.

After completion of the etching process, the resulting suspension was centrifuged at 9000 rpm, followed by repeated washing with deionized water until neutralization (supernatant pH = 7), yielding delaminated MXene powder.

1.3 Preparation of g-C_3N_4

Urea and melamine in a mass ratio of 3:1 were thoroughly ground and homogenized, followed by thermal treatment in a muffle furnace at 550°C for 4 h. After the reaction system cooled to room temperature, g-C_3N_4 was obtained via centrifugation.

1.4 Preparation of the BiOBr

A certain amount of $Bi(NO_3)_3 \cdot 5H_2O$ was dissolved in $C_2H_6O_2$ (ethylene glycol) to form solution A. Meanwhile, KBr was dissolved in $C_2H_6O_2$ to form solution B. Solution A was added dropwise into solution B under stirring. After complete addition, the mixture was transferred into an autoclave and reacted at 120 °C for 8 h. The resulting product was dried to obtain BiOBr[1].

1.5 Preparation of the CN/BiOBr

Initially, 0.60 g of $Bi(NO_3)_3 \cdot 5H_2O$ was dissolved in 50.0 mL of deionized water to form Solution A. Subsequently, 0.32 g of g-C_3N_4 was dispersed in 50 mL of deionized water, followed by the addition of 0.73 g of $C_{19}H_{42}BrN$, resulting in Solution B. Solution A was then added dropwise into Solution B under continuous stirring. The mixture was heated in an 80°C oil bath for 3 h. Thereafter, the product was isolated via vacuum filtration, thoroughly washed three times with deionized water and EtOH, and dried in an 80°C oven to yield the CN/BiOBr composite.

2. Material characterization and performance evaluation of bilayer MXene/PPAL-CN/BiOBr aerogel

2.1 Material characterization

The morphology of the samples was observed using scanning electron microscopy (SEM, TESCAN MIRA LMS, Czech Republic). Fourier transform infrared (FT-IR) spectra of the samples were recorded in the range of 400-4000 cm^{-1} using an FT-IR spectrometer (Nexus 670, Nicolet). X-ray diffraction (XRD) analysis was performed on the samples using an X-ray diffractometer (D8 Advance, Bruker, Germany). Thermal conductivity was measured using a thermal conductivity meter (TC3200, XIATECH, China). Longitudinal compression tests of the aerogels were conducted using a universal testing machine (Jinan Lianguo Testing Technology Co., Ltd., China) at a compression rate of 10 $mm \cdot min^{-1}$. The wetting behavior of the samples was observed and recorded using a contact angle meter (OCA20). Pore characteristics were analyzed using an automatic mercury porosimeter (AutoPore IV 9500, Micromeritics). Diffuse reflectance measurements of the samples were performed using a UV-Vis-NIR spectrophotometer (Cary 5000 UV-Vis-NIR) equipped with an integrating sphere. The elemental composition and chemical states were analyzed using X-ray photoelectron spectroscopy (XPS, PHI 5000 Versaprobe III, Japan).

2.2 Performance evaluation of bilayer MXene/PPAL-CN/BiOBr aerogel

2.2.1 Evaluation of solar interfacial evaporation performance

A laboratory-built solar simulator-based interfacial evaporation setup was used to evaluate the performance of the solar evaporator, as shown in Figure S1. The setup

included a solar simulator lamp to ensure uniform illumination, a precision analytical balance connected to a computer for real-time recording of water mass changes, and an optical power densitometer to adjust the sample height to achieve standard light intensity. An infrared camera was employed to monitor the evaporator temperature for thermal behavior analysis.

After turning on the xenon lamp, the light intensity was set to 1 kW·m^{-2}. The photothermal material was placed on the water surface inside a fixed-height glass tube, while the analytical balance and computer synchronously recorded mass changes, and an infrared thermal camera tracked the temperature. The ambient temperature ranged from 19 to 26 °C, with a relative humidity of 15-28%. All data were obtained under a light intensity of 1 kW·m^{-2}.

The energy efficiency (η) of solar evaporation is calculated using the following formula:

$$\eta = \frac{m h_{LV}}{C_{opt} q_i} \quad (1)$$

Where η is the solar-to-thermal energy conversion efficiency, m is the mass change rate of water, h_{LV} is the total enthalpy (including both latent and sensible heat), q_i is the solar radiation intensity under one sun illumination, and $C_{opt} q_i$ is the total power density of solar irradiation.

$$h_{LV} = \lambda + C\Delta T \quad (2)$$

Where λ is the phase change latent heat of water, $C = 4.2\ \mathrm{kJ\ kg^{-1}\ K^{-1}}$ is the specific heat capacity of water, and ΔT is the change in surface temperature of the

sample before and after the experiment.

The efficiency is calculated by multiplying the absolute value of the slope of the water mass change versus time curve during evaporation by the duration of the evaporation experiment.

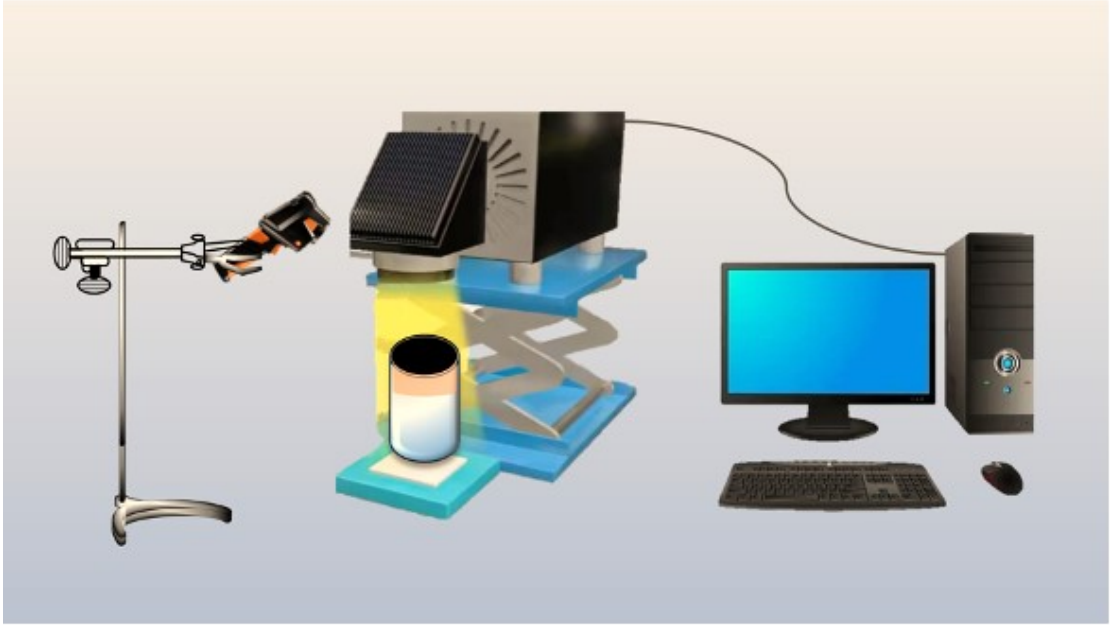

Figure S1. The schematic diagram of the evaporation system

2.2.2 Evaluation of interfacial evaporation performance in acidic wastewater

For the interfacial evaporation test in acidic wastewater, the pure water used in the solar interfacial evaporation test was replaced with acidic wastewater prepared by diluting concentrated H_2SO_4 to concentrations of 3%, 5%, and 10%.

2.2.3 Photoelectrochemical performance measurement

Photoelectrochemical characterization was performed using a CHI660E electrochemical workstation (Shanghai Chenhua) with a standard three-electrode system. First, 9 mg of the material was dispersed in 240 μL of ethylene glycol and 10 mL of Nafion membrane solution (5 wt%), followed by ultrasonication for 30 min.

Then, 50 μL of the resulting catalyst ink was drop-cast onto the surface of conductive glass (active area of 2 cm^2), which served as the working electrode. Ag/AgCl and a platinum sheet were used as the reference and counter electrodes, respectively, with 0.5 M Na_2SO_4 as the electrolyte. A 50 W LED lamp was used as the light source ($\lambda \geq 420$ nm). Photocurrent measurements were conducted at room temperature under open-circuit potential conditions. Electrochemical impedance spectroscopy (EIS) was performed in the frequency range of 10 mHz to 100 kHz with an AC amplitude of 5 mV at open-circuit potential. Mott–Schottky plots were recorded at open-circuit potential with an initial potential of –1 V and a final potential of 0.5 V.

2.2.4 Photocatalytic performance test

The photocatalytic degradation of tetracycline (TC) was evaluated at room temperature using a 50 W LED lamp ($\lambda \geq 420$ nm). Typically, 0.4 g of the aerogel (or, for CB/BiOBr, 100 mg based on the solid mass fraction of MXene/PPAL-CN/BiOBr50) was placed in 100 mL of a 20 $mg \cdot L^{-1}$ TC solution. To eliminate the effect of adsorption, the suspension was magnetically stirred in the dark for 40 min to achieve adsorption–desorption equilibrium before irradiation. After the photodegradation reaction, the catalyst was separated using a polypropylene syringe filter.

Under visible light irradiation, the light absorption of the pollutant follows the Lambert-Beer law, and the degradation efficiency can be calculated using the following equation:

$$= \left(1 - \frac{c}{c_0}\right) \times 100\% = \left(1 - \frac{A}{A_0}\right) \times 100\% \quad (3)$$

Where E is the degradation efficiency, A_0 is the absorbance of the initial solution, and A_0 is the absorbance of the solution at different time intervals.

2.2.5 Cyclic performance test

For the cyclic stability test, 0.4 g of the aerogel was added to 100 mL of a 20 mg·L^{-1} TC solution. After 40 min of dark adsorption, the suspension was irradiated under visible light for 80 min to ensure complete removal of TC adsorbed on the photocatalyst. The photocatalyst was then collected by centrifugation with anhydrous ethanol, washed with deionized water, and freeze-dried for 12 h before being reused in the next cycle.

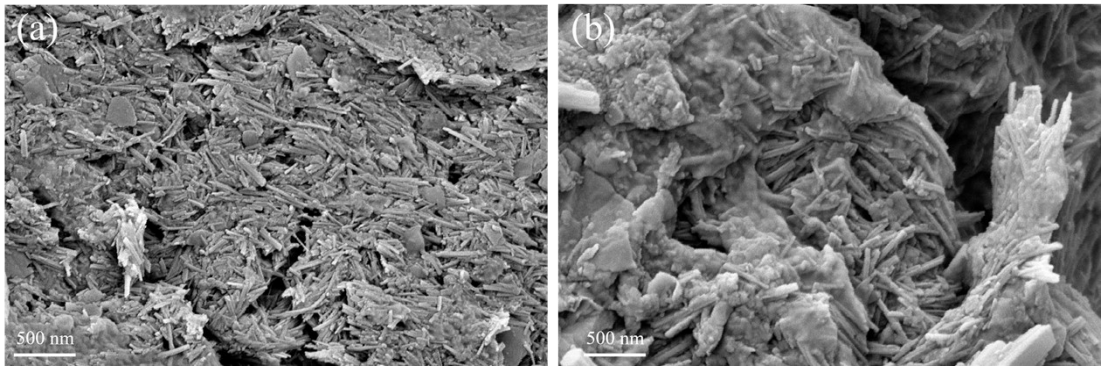

Figure S2. SEM images of MXene/PPAL-CN/BiOBr30 before and after exposure to 5% H_2SO_4 for one hour: (a) before acid exposure, (b) after acid exposure.

References

1 L. Xie, P. Zhu, J. Xu, M. Duan, S. Zhang and X. Wu, *Langmuir*, 2022, 38, 9532–9545.