High-performance solar vapor generation of Ni/carbon nanomaterials by controlled carbonization of waste polypropylene

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ABSTRACT Solar vapor generation is emerging as a promising technology using solar energy for various applications including desalination and freshwater production. However, from the viewpoints of industrial and academic research, it remains challenging to prepare low-cost and high-efficiency photothermal materials. In this work, we report the controlled carbonization of polypropylene (PP) using NiO and poly(ionic liquid) (PIL) as combined catalysts to prepare a Ni/carbon nanomaterial (Ni/CNM). The morphology and textural property of Ni/CNM are modulated by adding a trace amount of PIL. Ni/CNM consists of cup-stacked carbon nanotubes (CS-CNTs) and pear-shaped metallic Ni nanoparticles. Due to the synergistic effect of Ni and CS-CNTs in solar absorption, Ni/CNM possesses an excellent property of photothermal conversion. Meanwhile, Ni/CNM with a high specific surface area and rich micro-/meso-/macropores constructs a three-dimensional (3D) porous network for efficient water supply and vapor channels. Thanks to high solar absorption, fast water transport, and low thermal conductivity, Ni/CNM exhibits a high water evaporation rate of $1.67 \text{ kg m}^{-2} \text{ h}^{-1}$, a solar-to-vapor conversion efficiency of 94.9%, and an excellent stability for 10 cycles. It also works well when converting dye-containing water, seawater, and oil/water emulsion into healthy drinkable water. The metallic ion removal efficiency of seawater is 99.99%, and the dye removal efficiency is >99.9%. More importantly, it prevails over the-state-of-art carbon-based photothermal materials in solar energy-driven vapor generation. This work not only proposes a new sustainable

approach to convert waste polymers into advanced metal/carbon hybrids, but also contributes to the fields of solar energy utilization and seawater desalination.

Keywords: solar vapor generation, waste polymer, controlled carbonization, Ni/carbon nanomaterial, synergistic effect, photothermal materials

INTRODUCTION

During the last two decades, the shortage of freshwater resources has become a global problem, which affects human production and life. Currently, the major methods to increase freshwater supply include desalination and sewage treatment. As a clean energy source, solar energy can be converted into heat for evaporating seawater to obtain fresh water [1–4]. Traditional evaporation is achieved by heating bulk water, which however is prone to heat loss and leads to low solar-to-vapor conversion efficiency. Recently, photothermal materials, which are located at the interface of water and air, have drawn a great deal of attention for vapor generation. It reduces heat loss in the bulk water by localized heat, so that the solar-to-vapor conversion efficiency is promoted. To realize a high photothermal conversion efficiency at the water-air interface, photothermal materials should show broadband sunlight absorbability, open porosity for rapid water molecule transportation, and low thermal conductivity. Various photothermal materials have been ex-

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pored, including metal plasmonic nanoparticles (e.g., Au and Ag) [5–8], and carbon materials (e.g., graphene, carbon nanotube (CNT) and carbon fiber) [9–12].

Under resonant illumination by light, the plasmon-excited electrons of metal nanoparticles are non-radiatively damped *via* the Landau damping mechanism and redistribute their energy through electron-electron and electron-phonon scattering processes to generate heat [13]. While the solar absorption of carbon materials involves the excitation of electrons and their subsequent relaxation, and the thermalization of optically excited electrons is also caused by the scattering of electron-electron and electron-phonon. The previous achievements lay a solid foundation for the solar vapor generation to obtain clean water. However, to further improve the photothermal conversion efficiency and the performance of solar vapor generation, some basic questions are urgent to be addressed. Firstly, could metal nanoparticles and carbon materials show a synergistic effect in solar vapor generation considering their similar photothermal conversion mechanisms? Secondly, the relationship between the morphology of carbon materials and solar vapor generation performance is ambiguous. Could the solar vapor generation performance be improved by optimizing the morphology and microstructure of carbon materials? Finally, the high cost of precious metals limits the application in solar vapor generation; consequently, a facile sustainable strategy is highly desirable to prepare low-cost photothermal materials for utilizing solar energy.

Waste polymers have caused terrible environmental issues such as “White Pollution”, because most waste polymers are not biodegradable and need hundreds of years to be degraded naturally. Taking waste plastics as an example, the quantity of worldwide waste plastics has reached 6.4 billion tons since the 1950s. Sustainable development calls for low-environmental-impact technologies to recycle waste polymers in place of the current practices of landfilling and incineration [14,15]. Since polymers generally contain rich carbon element, converting waste polymers into high value-added carbon not only contributes to the reutilization of waste polymers, but also provides a low-cost way to prepare functional carbon materials for diverse applications [16–21]. For example, Williams’ group [22,23] catalyzed gasification of waste polypropylene (PP), polyethylene (PE) and polystyrene into CNTs. Levendis’ group [24,25] reported the synthesis of CNTs from recycled PE using a pyrolysis-combustion technique. We put forward the controlled carbonization of polymers to prepare CNTs and carbon

nanosheets using combined catalysts or active templates [26–28]. Unfortunately, in most of these above cases, the corrosive acids or alkalis are required to remove the remained catalysts or templates for purifying carbon products, which makes the production process cumbersome. Besides, there have been no reports yet on solar vapor generation from seawater to obtain fresh water using low-cost waste polymers-derived carbon materials.

Poly(ionic liquid) (PIL) refers to a subclass of polyelectrolytes that feature an ionic liquid species in each monomer repeating unit, connected through a polymeric backbone to form a macromolecular structure [29]. It usually contains conjugated or incorporated heteroatoms in the molecular structure, which brings PILs versatile properties. In this contribution, we report the combined catalysts of halogen-containing PIL (PIL-X)/NiO to achieve the controlled carbonization of PP into Ni/CNM by a one-pot approach. The morphology, microstructure and textural structure of Ni/CNM are precisely regulated by using a trace amount of PIL-X. The synergistic effect of Ni and CS-CNTs in Ni/CNM is proved to improve the solar absorption in photothermal conversion. Meanwhile, Ni/CNM bearing small and long CS-CNTs as well as high porosity forms a three-dimensional (3D) porous network to accelerate water transport. Such combined features endow Ni/CNM with excellent performance in solar vapor generation which exceeds most of the previous carbon-based photothermal materials.

EXPERIMENTAL SECTION

Materials

PP (weight-average molecular weight = 3.07×10^5 g mol⁻¹, trademark M16) pellets were purchased from Shandong Usole Co. Waste PP cups were collected from the local restaurant (Wuhan). Ethanol, methylene blue (MB), Rhodamine B (RhB), dimethylpolysiloxane, and hydrochloric acid were purchased from Sinopharm Chemical Reagent Co., Ltd. NiO nanoparticles were prepared by a sol-gel combustion method. Briefly, Ni(NO₃)₂·6H₂O (64.5 g) and citric acid (13.5 g) were dissolved in 300 mL of deionized water. The solution was evaporated at 80°C until forming gel, which was heated at the temperature of self-ignition in air. The obtained NiO powder was calcined at 300°C for 2 h in a muffle oven and used as the carbonization catalyst. The sizes of NiO nanoparticles were mainly in the range of 10–18 nm (Fig. S1). Seawater was obtained from South Sea (near to Hainan). Three PIL-X (Fig. 1), namely, PIL-Cl, PIL-Br and PIL-I, were synthesized according to the work by Yuan *et al.* [30–32].

Figure 1 Schematic diagram of the controlled carbonization of PP by combined catalysts of NiO/PIL-X to prepare Ni/CNM for solar steam generation.

Preparation of Ni/CNM from the carbonization of PP by NiO/PIL-X

Firstly, PP pellet (10.0 g) was mixed with NiO (750.0 mg, 7.5 wt%) and a designed amount of PIL-X, i.e., PIL-Cl=189.0 mg (1.89 wt%), PIL-Br=34.0 mg (0.34 wt%), and PIL-I=19.0 mg (0.19 wt%). The amount of PIL-X was determined according to our previous work [26], in which the optimal halogen content for the carbonization of PP in the presence of NiO was found to be 7.28 $\mu\text{mol}/100\text{ g PP}$ for Cl, 1.52 $\mu\text{mol}/100\text{ g PP}$ for Br, and 0.52 $\mu\text{mol}/100\text{ g PP}$ for I, respectively. The mixture was denoted as PP/NiO/PIL-Cl, PP/NiO/PIL-Br, and PP/NiO/PIL-I, respectively. For comparison, PP/NiO mixture (NiO=7.5 wt%) was prepared, and W-PP/NiO/PIL-I mixture was obtained by mixing waste PP (10.0 g) with NiO (750.0 mg) and PIL-I (18.9 mg).

Secondly, Ni/CNM was obtained by heating the PP/NiO/PIL-X mixture in a crucible at 750°C for 8 min. The resultant Ni/CNM was slowly cooled to room temperature, weighed and finally denoted as Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I, respectively. Likewise, Ni/CNM-0 and W-Ni/CNM-I (Fig. S2) were obtained from the carbonization of PP/NiO and W-PP/NiO/PIL-I mixtures, respectively. The yield of Ni/CNM was calculated by dividing the amount of residue by that of mixture, and the carbon conversion was obtained by dividing the amount of carbon (i.e., the amount of residue after subtracting the amount of residual catalyst) by that of carbon in PP. Besides, Ni/CNM-I was treated by HCl aqueous solution (0.5 mol L⁻¹) for 2 days at room temperature to remove the Ni catalyst, and the product was denoted as CNM-I.

Characterization

The morphology of Ni/CNM was observed by means of a

field-emission scanning electron microscope (SEM, S4800ESEM-FEG). The surface element distribution was analyzed using an energy dispersive X-ray spectrometer (EDX, Genesis 2000). The microstructure of Ni/CNM was investigated by using a transmission electron microscope (TEM, Tecnai G2 F30) at an accelerating voltage of 100 kV and high-resolution TEM (HRTEM) on a Tecnai G2 F30 field-emission transmission electron microscope operating at 200 kV. For preparing TEM sample, 1 mg of Ni/CNM-I was dispersed in 10 mL of ethanol by sonication (100 W) for 10 min, and 50 μL of the dispersion was dropped on a copper mesh bearing a carbon membrane and naturally dried at room temperature. The textural structure of Ni/CNM was measured based on N₂ adsorption/desorption isotherms at 77 K by using a surface area analyzer (Micromeritics ASAP 2460). Before the measurement, the sample (ca. 100 mg) was dried at 150°C under vacuum of 10⁻⁶ bar for 24 h. The pore size distribution was obtained by the quench solid density functional theory (DFT) and Barret-Joyner-Halenda (BJH) method. Raman spectrum was collected by using a confocal Raman microscope (inVia Reflex, excitation beam wavelength=532 nm). The phase structure of Ni/CNM was analyzed by X-ray diffraction (XRD, SmartLab-SE) with Cu K α radiation operating at 40 kV and 200 mA. The element content of Ni/CNM was determined by an Elemental Analyzer (Vario EL III). The surface element composition of Ni/CNM was characterized by means of X-ray photoelectron spectroscopy (XPS) carried out on a VG ESCALAB MK II spectrometer. The thermal stability of Ni/CNM was measured by thermogravimetric analysis (TGA) by using TA Instruments SDT Q600 in air atmosphere from 30 to 800°C at a heating rate of 10°C min⁻¹, and the mass of sample was ca. 20 mg. The functional group of Ni/CNM was characterized by

Fourier transform infrared spectroscopy (FT-IR, BRUKER Vertex 80 FT-IR, Germany). The absorption spectrum of Ni/CNM was tested using a UV-Vis-NIR spectrophotometer (Lambda 750 S) with an integrating sphere. The thermal conductivity of Ni/CNM-I was measured by using a thermal conductivity Instrument (Hot Disk, TPS 2500, Sweden). Water contact angle measurement was carried out on a micro optical contact angle measurement (Dataphysics OCA15EC, Germany) by using an 18 μL droplet of water as an indicator.

Solar vapor generation experiment

A solar light simulator (CEL-S500L) was used to carry out the vapor generation experiment (Fig. S3). Firstly, Ni/CNM membrane was prepared by vacuum filtration. Typically, 6.0 mg of Ni/CNM-I was added into 24 mL of ethanol and sonicated for 5 min. The dispersion was filtered onto polyvinylidene fluoride (PVDF) membrane (diameter=50 mm, pore size=0.22 μm) by a vacuum pump. The resultant membrane was dried at room temperature and employed as the absorber in solar vapor device. Our previous results showed that the amount of Ni/CNM-I affected the solar vapor generation, and the best result was obtained by adding 6.0 mg of Ni/CNM-I (Fig. S4) under the humidity of 30%. Afterwards, the resultant Ni/CNM membrane with a diameter of 26 mm was floated on the surface of a 2 cm-thick polystyrene foam. The surface temperature of water and membrane was monitored by an infrared thermal imaging camera (DM-I220, Dongmei). Infrared images were also captured by using this camera. The mass change of water was measured by an electronic balance (JA2003, Sotop). The evaporation rate (m , $\text{kg m}^{-2} \text{h}^{-1}$), solar-to-vapor conversion efficiency (η , %), and enhancement factor (EF) were calculated by Equations (1–3), respectively

$$m = \frac{\Delta m}{S \times t}, \quad (1)$$

$$\eta = \frac{m' \times h_{\text{LV}}}{P_{\text{in}}}, \quad (2)$$

$$\text{EF} = \frac{m(\text{absorber})}{m(\text{blank})}, \quad (3)$$

where Δm is the mass change of water in 1 h (kg), S is the area of Ni/CNM membrane (m^2), t is the time of solar irradiation (1 h), m' is the evaporation rate after subtracting the evaporation rate in dark ($\text{kg m}^{-2} \text{h}^{-1}$), h_{LV} is the latent heat of vaporization of water (kJ kg^{-1}), and P_{in} is the incident light power on the solar absorber (kW m^{-2}). Solar vapor generation without absorbers was studied as reference experiment, noted as blank.

For the water purification experiment, seawater, dye-containing water (i.e., RhB with the concentration of 20 mg L^{-1} and MB with the concentration of 20 mg L^{-1} , respectively), and oil/water emulsion (i.e., dimethylpolysiloxane with the concentration of 1 wt%) were prepared. UV-Vis absorption spectra of the dye-containing water and the condensed water were recorded on a UV-6100 spectrometer (METASH). The metallic ion concentrations of the seawater and the condensed water were determined by using inductively coupled plasma-optical emission spectrometry (ICP-OES, JY 200-2, HORIBA Scientific). The optical images of the oil/water emulsion and the condensed water were observed by using an optical microscope (Mshot, MS60). Moreover, the solar vapor generation performance of Ni/CNM-I in dye solution (MB=20–80 mg L^{-1} or RhB=20–80 mg L^{-1}), salt solution (0.8–10 wt%) or oil/water emulsion (0.5–2.5 wt%) was also investigated.

To study the effects of morphology and microstructure of Ni/CNM on the evaporation rate, a model experiment was conducted. Briefly, a piece of Ni/CNM membrane was put on the surface of water, and the diffusion of water on the membrane was recorded by a camera. The kinetics of diffusion process was analyzed by the Weibull distribution function as follows:

$$D = 1 - e^{-\frac{(t-\alpha)^k}{\beta}}, \quad (4)$$

where t represents the diffusion time (s), D (%) is the degree of water diffusion in t , and α is the position parameter, indicating the lag time. Since there is no lag time in the process of water diffusion, the α value is 0. β is the scale parameter which influences the size of the curve. k is the shape parameter which determines the shape of the curve. The parameters k and β play the major roles in the process of water diffusion. The deformation equation of the Weibull function is shown in

$$\ln\left(\frac{1}{1-D}\right) = k \ln t - \ln \beta. \quad (5)$$

It is a linear equation in which $-\ln \beta$ represents the intercept and k is the slope. Thus, β and k are calculated by linear fitting of Equation (5).

RESULTS AND DISCUSSION

Yield of Ni/CNM

The effect of PIL-X on the yield of Ni/CNM from the carbonization of PP at 750°C was explored (Table S1). When the carbonization of PP is conducted in the pre-

sence of PIL-X alone, the yield of carbon is ca. 0.33, 0.14 and 0.12 wt% using PIL-Cl, PIL-Br and PIL-I, respectively. While the yield of carbon using NiO alone is 9.5 wt%. It is indicated that PIL-X or NiO alone could not effectively catalyze the carbonization of PP. By contrast, the yield of carbon materials using combined NiO/PIL-X catalysts is 23.5 wt% for NiO/PIL-Cl, 25.3 wt% for NiO/PIL-Br, and 42.9 wt% for NiO/PIL-I, obviously higher than that using PIL-X or NiO alone. Therefore, NiO/PIL-X shows a synergetic effect on the “controlled carbonization” of PP into Ni/CNM, and the catalytic efficiency of the combined catalysts follows the sequence: NiO/PIL-Cl < NiO/PIL-Br < NiO/PIL-I. The combined catalysts of NiO/PIL-I also convert waste PP into carbon product (W-Ni/CNM-I) with a high yield of 35.5 wt%.

Morphology and microstructure of Ni/CNM

To investigate whether PIL-X had any effects on the morphology and microstructure of Ni/CNM, SEM, TEM and HRTEM observations were performed. Ni/CNM-0 is composed of granular carbon with a size range of 90–150 nm, while Ni/CNM-Cl, Ni/CNM-Br, and Ni/CNM-I are mainly in the form of carbon filaments (Fig. 2 and Figs S5–S7). Interestingly, the mean diameter of carbon filaments in Ni/CNM-I (24.2 nm) is smaller than that of Ni/CNM-Cl (48.2 nm) or Ni/CNM-Br (37.4 nm), while the carbon filaments of Ni/CNM-I (875.7 nm) are longer than those of Ni/CNM-Cl (317.7 nm) or Ni/CNM-Br (446.7 nm). Hence, Ni/CNM-Cl and Ni/CNM-Br bear shorter and bigger filamentous carbon, while Ni/CNM-I contains relatively longer and slimmer filamentous carbon. The voids or macropores among the filamentous carbon increase obviously from Ni/CNM-Cl and Ni/CNM-Br to Ni/CNM-I. These voids or macropores can accelerate the transportation of water by providing more channels due to capillary force. W-Ni/CNM-I also contains a large amount of filamentous carbon. The diameter and length of filamentous carbon are 36.9 and 220.6 nm, respectively (Fig. S8). Furthermore, the elemental mapping based on EDX (Fig. S9) portrays the homogeneous distribution of carbon, oxygen and nickel elements in Ni/CNM-I.

The resultant long filamentous carbon has a tubular-like form, which is the characteristic of CNTs (Fig. S10). The graphene layers in the CNTs are oblique to the CNT axis in a range of 25°–31° (Fig. 2f). Hence, the obtained CNTs are identified as CS-CNTs. The graphene layer space of CS-CNTs was calculated by fast Fourier transform to be ca. 0.34 nm (Fig. 2f), which agrees with that of graphite. To clarify the growth mechanism of CS-CNTs,

Figure 2 SEM images of (a) Ni/CNM-0, (b) Ni/CNM-Cl, (c) Ni/CNM-Br, and (d) Ni/CNM-I. (e, f) HRTEM images of Ni/CNM-I.

the morphology of metallic Ni catalyst in CS-CNTs was observed. Interestingly, most of the metallic Ni catalysts (20–30 nm) are pear-like and embedded on the tip of CS-CNTs (Fig. 2e). The graphene layers near the surface of the pear-shaped Ni catalyst are almost parallel to the surface of catalyst, implying that CS-CNTs grow up from the tip of catalyst. The above results suggest the reconstruction of NiO nanoparticles into the pear-like NiO nanoparticles before the growth of CS-CNTs.

Phase structure and thermal stability of Ni/CNM

XRD patterns of Ni/CNM are displayed in Fig. 3a. Characteristic diffraction peaks of graphite ($2\theta=26.1^\circ$ (002)) and metallic Ni ($2\theta=44.8^\circ$ (111) and 52.3° (200)) are observed, confirming the reduction of NiO into metallic Ni during carbonization. Furthermore, the full width at half maximum (FWHM) of graphite (002) diffraction peak decreases from Ni/CNM-0 (3.8°), Ni/CNM-Cl (3.3°) and Ni/CNM-Br (3.1°) to Ni/CNM-I (2.7°), indicating that PIL-X facilitates the formation of better graphite structure. Fig. 3b shows Raman spectra of Ni/CNM. The peak at ca. 1350 cm^{-1} (D band) is associated with the vibration of carbon atoms with dangling bonds in the plane terminations of disordered graphite [33]. The

Figure 3 (a) XRD patterns, (b) TGA curves, (c) Raman spectra, (d) N_2 adsorption-desorption isotherms at 77 K, and pore size distribution plots using (e) DFT model and (f) BJH model of Ni/CNM.

G band at $\text{ca. } 1580 \text{ cm}^{-1}$ is owing to an E_{2g} mode of hexagonal graphite and related to the vibration of sp^2 -bonded carbon atoms in a graphite layer. Raman curve fittings of G and D peaks are presented in Fig. S11. The I_G/I_D is calculated by the area ratio of the G band to the D band. A larger I_G/I_D ratio indicates a higher degree of structural ordering for graphite [34]. The I_G/I_D ratio increases from 0.34 for Ni/CNM-0 to 0.37 for Ni/CNM-Cl, 0.50 for Ni/CNM-Br, and 0.58 for Ni/CNM-I. Thereby, PIL-X promotes the formation of Ni/CNM with fewer defects inside the graphite sheets and less amorphous carbon.

Furthermore, TGA was conducted to study the thermal stability of Ni/CNM and measure the contents of Ni and

carbon (Fig. 3c). The weight loss over the range of 450–600°C is caused by the oxidation of carbon [35]. The residue at 750°C is 61.9 wt% (Ni/CNM-0), 31.6 wt% (Ni/CNM-Cl), 31.6 wt% (Ni/CNM-Br), and 13.3 wt% (Ni/CNM-I), respectively. Since metallic Ni is oxidized to NiO, the content of Ni is calculated to be 48.5, 24.8, 24.8, and 10.4 wt%, respectively. The derivative TGA curves of Ni/CNM are displayed in Fig. S12. Ni/CNM-0 shows the maximum oxidation temperature at 448°C, which is lower than that of Ni/CNM-Cl, Ni/CNM-Br or Ni/CNM-I. Therefore, PIL-X facilitates the formation of graphitic carbon in Ni/CNM, consistent with the results of XRD and Raman.

The textural property of Ni/CNM was investigated by

N_2 adsorption/desorption measurement at 77 K. Ni/CNM presents the combined type I/IV physisorption isotherms (Fig. 3d). The type-H4 hysteresis loop occurs at a relative pressure (P/P_0) ranging from 0.6 to 0.9, which results from the filling and emptying of mesopores by capillary condensation. Meanwhile, the maximum adsorption amount at $P/P_0=0.1$ gradually increases from Ni/CNM-0 to Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I, implying the presence of many micropores. The pore size distribution plots in Ni/CNM were calculated using DFT (Fig. 3e) and BJH (Fig. 3f) models. With the addition of PIL-X, the micropores in Ni/CNM increase as expected. Particularly, Ni/CNM-Br and Ni/CNM-I possess abundant micropores centered at 0.4 and 1.3 nm.

According to the result of BJH model, Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I contain more mesopores and macropores than Ni/CNM-0, and Ni/CNM-I bears larger macropores than Ni/CNM-Cl and Ni/CNM-Br. Macropores or large mesopores are related to the voids among the CS-CNTs as shown in the SEM image (Fig. 2), and affected by the length and diameter of CS-CNTs. Fig. S13 shows the Brunauer-Emmett-Teller (BET) specific surface area (S_{BET}), specific surface area of micropores (S_{micro}), and specific surface area of mesopores and macropores ($S_{meso+macro}$) of Ni/CNM. Obviously, the addition of PIL-X improves the S_{BET} of Ni/CNM, and Ni/CNM-I shows the highest S_{BET} , S_{micro} , and $S_{meso+macro}$. Furthermore, when waste PP was used as carbon source, the carbon product W-Ni/CNM-I displays a S_{BET} of $225.3 \text{ m}^2 \text{ g}^{-1}$ (Fig. S14). Besides, the Ni/CNM bears rich oxygen-containing

functional groups (Fig. S15), which are believed to facilitate the transportation of water molecules.

Based on the above results and our previous work [26,27,36], the possible mechanism on the controlled carbonization of PP into Ni/CNM is shown in Fig. 4. Firstly, PP fragment radicals are generated by the cleavage of C-H and C-C bonds on PP backbone due to the heat induction (steps 1 and 2 in Fig. 4a). Meanwhile, PIL-X is decomposed into halogen radical (step 3 in Fig. 4a), which promotes the dehydrogenation and aromatization of PP fragment radicals. After further hydrogen transfer, dehydrogenation, cyclization, isomerization and aromatization, light hydrocarbons and aromatics are formed (steps 4–8 in Fig. 4a). The combination of halogen radical with hydrogen radical into halogen hydride (step 4 in Fig. 4a) is the rate determining step. Since the thermal stability of halogen hydride decreases in the sequence: chlorine hydride > bromine hydride > iodide hydride, the catalysis efficiency of PIL-X for the degradation of PP fragment radicals into light hydrocarbons and aromatics increases in the following consequence: PIL-Cl < PIL-Br < PIL-I. Hence the catalytic efficiency of combined catalysts follows the sequence: NiO/PIL-Cl < NiO/PIL-Br < NiO/PIL-I. The resultant light hydrocarbons and aromatics are carbon feedstocks for the growth of Ni/CNM. They firstly promote the reconstruction of NiO nanoparticles into pear-shape (step 9 in Fig. 4b), which are then reduced to metallic Ni prior to the growth of CS-CNTs. Subsequently, light hydrocarbons and aromatics are catalyzed into graphene nanosheets on the surface of

Figure 4 (a) Scheme showing the effect of PIL-X on the degradation of PP. (b) Possible mechanism for the growth of CS-CNTs from the degradation products of PP by NiO.

pear-shaped Ni catalyst (step 10 in Fig. 4b). These graphene nanosheets are parallel to the surface of pear-shaped Ni catalyst and assembled to gradually construct the CS-CNTs structure (step 11 in Fig. 4b). As a result, graphene layers in the CS-CNTs are oblique to the CS-CNT axis. Finally, CS-CNTs grow up from the tip of pear-shaped Ni catalyst.

Synergy effect of Ni and CS-CNTs on the solar vapor generation

In the above results, NiO/PIL-X was found to promote the controlled carbonization of PP into Ni/CNM. It is very attractive for academic and industrial communities to explore their potential applications in energy storage and utilization, and environmental remediation, etc. Here, Ni/CNM was utilized for solar vapor generation. As well-known, the ideal photothermal materials for solar vapor generation should show broadband sunlight absorbability, open porosity for rapid water transportation, low thermal conductivity, and high energy conversion efficiency.

Firstly, the solar vapor generation performance of Ni/CNM floating on the water surface was quantitatively

investigated by recording the mass change of water as the solar irradiation time. As shown in Fig. 5a, the water mass decreases approximately linearly with irradiation time. Ni/CNM-I presents an extremely high evaporation rate of $1.67 \text{ kg m}^{-2} \text{ h}^{-1}$, which is 2.7, 2.3 and 1.3 times as that of pure water ($0.62 \text{ kg m}^{-2} \text{ h}^{-1}$), PVDF ($0.72 \text{ kg m}^{-2} \text{ h}^{-1}$) and Ni/CNM-0 ($1.28 \text{ kg m}^{-2} \text{ h}^{-1}$), respectively. As shown in Fig. 5b and c, the solar-to-vapor conversion efficiency of Ni/CNM-I (94.9%) exceeds that of PVDF (31.9%) and Ni/CNM-0 (67.7%). Meanwhile, the enhancement factor of Ni/CNM-I (2.69) oversteps that of PVDF (1.16) and Ni/CNM-0 (2.14). These results indicate that Ni/CNM-I from the controlled carbonization of PP by NiO/PIL-X exhibits better performance in solar vapor generation than Ni/CNM-0 obtained by using NiO alone. The reason for the different performances between Ni/CNM-I and Ni/CNM-0 is summarized in two aspects. Firstly, Ni/CNM-I consisting of carbon filament structure possesses a higher S_{BET} than Ni/CNM-0 with a granule structure (Figs 2 and 3). The micropores, mesopores and macropores of Ni/CNM-I provide more channels to ensure that vapor passes through the membrane quickly. Secondly, the macropores or voids among the CS-CNTs in Ni/

Figure 5 (a) Cumulative mass changes of water vs. time under various conditions: water in dark (dark); water under 1 kW m^{-2} solar irradiation (blank); water with PVDF, Ni/CNM-0, Ni/CNM-I and CNM-I under 1 kW m^{-2} solar irradiation. (b) Evaporation rate and solar-to-vapor conversion efficiency of water, (c) enhancement factor of water, and (d) UV-Vis-NIR absorption spectra of Ni/CNM-0, Ni/CNM-I and CNM-I.

CNM-I (Fig. 2d) reduce the reflection of solar and promote the solar absorption. As a proof of concept, the solar absorption of Ni/CNM-I reaches to 98.9%–99.9%, higher than that of Ni/CNM-0 (95.6%–98.6%, Fig. 5d). Good solar absorption property results in the higher surface temperature of Ni/CNM-I (48.3°C) than that of Ni/CNM-0 (45.9°C, Fig. S16) in vapor generation, which can transform more water molecules to vapor and ultimately enhance the evaporation rate.

To study the synergy effect of Ni and CNM-I (i.e., CS-CNTs) on the performance of solar vapor generation, metallic Ni of Ni/CNM-I was removed by using hydrochloric acid to prepare CNM-I (i.e., CS-CNT, Fig. S17). CNM-I displays the evaporation rate of $1.51 \text{ kg m}^{-2} \text{ h}^{-1}$, the solar-to-vapor conversion efficiency of 84.0% (Fig. 5b) and the enhancement factor of 2.43 (Fig. 5c), which are lower than those of Ni/CNM-I ($1.67 \text{ kg m}^{-2} \text{ h}^{-1}$, 94.9%, and 2.69, respectively). Metallic Ni thus plays an indispensable role in solar vapor generation. To reveal the role of metallic Ni, the UV-Vis-NIR absorption spectrum of CNM-I was measured. CNM-I shows a broad-band absorption of solar irradiation with the optical absorption around 94.0%–96.5% (Fig. 5d), but lower than that of Ni/CNM-I (98.9%–99.9%), indicating that metallic Ni promotes the absorption of solar irradiation.

Fig. 6a shows the surface temperatures of water, PVDF, CNM-I and Ni/CNM-I in air. The temperature of water and PVDF is only 33.2 and 44.0°C, respectively, after 10 min under 1 kW m^{-2} solar light irradiation. For CNM-I and Ni/CNM-I, the surface temperature raises rapidly in the first 2 min. The temperature of Ni/CNM-I begins to stabilize at 3 min, but the temperature of CNM-I starts to stabilize until 7 min. After 10 min irradiation, the surface temperature of CNM-I (72.4°C) is lower than that of Ni/CNM-I (75.1°C). Additionally, CNM-I cools faster than Ni/CNM-I. Previous work showed that metallic particles could enhance the absorption of carbon materials to solar irradiation [37–39]. Here Ni/CNM-I is thus heated up rapidly to a higher temperature under light irradiation and slowly cools without solar irradiation. The surface temperatures of CNM-I and Ni/CNM-I on the water surface were also measured (Fig. 6b). The average temperatures of Ni/CNM-I and CNM-I are 48.3 and 45.0°C, respectively, which confirms the important role of Ni in photothermal conversion. Fig. 6c shows the infrared images of Ni/CNM-I and CNM-I under 1 kW m^{-2} solar irradiation. The experimental results reveal that the synergy effect of Ni and CNM-I (i.e., CS-CNT) endows Ni/CNM-I with an excellent photothermal conversion

property (Fig. 6d). In addition, we calculated the heat loss of Ni/CNM-I and CNM-I in the water evaporation process (for detailed information, see Note 1 in Supplementary information). Ni/CNM-I shows a low heat loss of 15.3%, including radiation (7.2%), convection (5.0%) and conduction (3.1%). This value is close to the heat loss of CNM-I (14.8%). Besides, Ni/CNM-I shows a low thermal conductivity of $0.119 \text{ W m}^{-1} \text{ K}^{-1}$. As a result, Ni/CNM-I shows high performance in the solar vapor generation (the inset in Fig. 6f, and Video S1). More importantly, it surpasses most of previously reported photothermal materials [5,6,40–43] (Fig. 6 and Table S2), and the whole cost of Ni/CNM-I is calculated to be ca. ¥ 0.86/g (Table S3). Besides, W-Ni/CNM-I exhibits good performance in solar vapor generation. The evaporation rate, solar-to-vapor conversion efficiency and enhancement factor are $1.52 \text{ kg m}^{-2} \text{ h}^{-1}$, 84.9% and 2.45, respectively (Fig. S14).

Effect of the morphology of Ni/CNM on the solar vapor generation

To interpret the effect of the morphology of Ni/CNM on the solar vapor generation, Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I were employed as absorbers (Fig. 7). The mass loss of water increases gradually from Ni/CNM-Cl and Ni/CNM-Br to Ni/CNM-I (Fig. S18a). The evaporation rate, solar-to-vapor conversion efficiency and enhancement factor are $1.33 \text{ kg m}^{-2} \text{ h}^{-1}$, 71.9% and 2.14 for Ni/CNM-Cl, respectively, and $1.55 \text{ kg m}^{-2} \text{ h}^{-1}$, 87.0% and 2.50 for Ni/CNM-Br, respectively (Fig. 7a and S18b), revealing that Ni/CNM-I shows higher performance in solar vapor generation than Ni/CNM-Cl and Ni/CNM-Br. Consequently, the morphology of Ni/CNM indeed affects the performance of solar vapor generation. However, whether in the air or on the water surface, the surface temperature of Ni/CNM-I is close to that of Ni/CNM-Cl or Ni/CNM-Br (Fig. 7d), meaning that they show similar solar absorption properties.

It is acknowledged that the broadband sunlight absorbability and open porosity for rapid water molecule transportation are crucial for solar vapor generation. To explain the different performances of Ni/CNM in solar vapor generation, we conducted a model experiment to explore the water transportation in Ni/CNM (inset in Fig. 7c). Briefly, Ni/CNM membrane with a diameter of 26 mm was placed on a polystyrene foam which was wrapped by hydrophilic silk and floating on the water surface. The diffusion of water in the Ni/CNM was characterized by observing the cumulative diffusion area of water *versus* time (Fig. 7c). At first, the diffusion of

Figure 6 Surface temperature of water (blank), PVDF, CNM-I and Ni/CNM-I under 1 kW m^{-2} solar irradiation (a) in air and (b) on the water surface. (c) Infrared images of CNM-I and Ni/CNM-I under 1 kW m^{-2} solar irradiation. Schematic diagrams of photothermal conversion mechanisms of (d) CNM-I and (e) Ni/CNM-I. (f) Comparison of solar vapor generation performance of Ni/CNM-I with previous photothermal materials (Table S2). The inset shows the screenshot of the video about the solar vapor generation using Ni/CNM-I (Video S1).

water in Ni/CNM is rapid, then slows down, and finally stabilizes since Ni/CNM is completely infiltrated. There are some differences in diffusion-time curves (e.g., shape and size). The water diffusion in Ni/CNM-Cl begins to slow down after 40 s, and the membrane is completely infiltrated at 80 s. While the water diffuses slowly in Ni/CNM-Br after 35 s and completely covers the membrane at 60 s. In contrast, water infiltrates ca. 80% area of Ni/CNM-I membrane at 20 s, and completely diffuses on the surface of membrane at 55 s. In addition, the fitting results by using Weibull model (Fig. 7d) show that the value of shape parameter (k) in Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I is 1.12, 1.14 and 1.27, respectively. The higher

k value of Ni/CNM-I proves the faster water diffusion in Ni/CNM-I than Ni/CNM-Cl and Ni/CNM-Br. Meanwhile, the value of scale parameter (β) decreases from Ni/CNM-Cl (39.65) and Ni/CNM-Br (29.67) to Ni/CNM-I (27.39), suggesting the decreasing size of diffusion-time curves in Ni/CNM in the same sequence. Due to a certain area of membrane (i.e., diffusion=100%), the smaller size of the diffusion-time curve means the shorter time to complete the diffusion on the surface of the membrane. These results above illustrate that the water transportation capability of Ni/CNM follows the sequence: Ni/CNM-Cl < Ni/CNM-Br < Ni/CNM-I, which agrees with the results of SEM and N_2 physisorption. It is thus supposed

Figure 7 (a) Evaporation rates and solar-to-vapor conversion efficiencies of water with Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I. (b) The average surface temperature of Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I under 1 kW m^{-2} solar irradiation in air and on the water surface. (c) The diffusion-time curves showing cumulative diffusion area of water vs. time in Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I; the inset displays the scheme of water diffusion experiment. (d) The fitting results of the water diffusion using Weibull model in Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I. (e) Schemes of water diffusion in Ni/CNM-Cl, Ni/CNM-Br and Ni/CNM-I.

that the water diffusion in Ni/CNM is associated with the morphology of Ni/CNM (Fig. 7e). The diameter of CS-CNTs decreases, following the sequence: Ni/CNM-Cl < Ni/CNM-Br < Ni/CNM-I (Fig. 2), but the length of CS-CNTs increases, which improves their porous structures (e.g., macropores). In solar vapor generation, pores are conducive to water diffusion and transportation to the surface of the absorber by capillary force, and enhance the evaporation rate.

In addition, water contact angle of Ni/CNM was measured to investigate the water wettability (Fig. S19). Overall, Ni/CNM exhibits the Wenzel's wetting behavior [44]. Ni/CNM-0 has an initial high contact angle (146.2°) which decreases to 0° in 43.5 s, suggesting its poor wettability probably due to the low S_{BET} and micron-scale structure. Comparatively, the initial contact angle of Ni/CNM-Cl, Ni/CNM-Br or Ni/CNM-I is lower and rapidly decreases to 0° . Moreover, the initial contact angle and

infiltration time decrease in the sequence: Ni/CNM-Cl (43.9° and 4.6 s)>Ni/CNM-Br (22.1° and 1.5 s)>Ni/CNM-I (21.6° and 0.9 s), which agrees with the results of water transportation experiment. Overall, the performance of Ni/CNM in solar vapor generation is closely related to their water transportation capabilities and morphologies.

Solar vapor generation of Ni/CNM using different kinds of water

The application of Ni/CNM-I in the generation of clean

water from the simulated industrial dye wastewater, oil/water emulsion, and seawater was also investigated. MB and RhB aqueous solutions were used as the simulated industrial dye wastewater samples. After purification using Ni/CNM-I under solar irradiation, the condensed water is colorless with naked eye and shows the UV-Vis absorbance value of near to zero (Fig. 8a). The purification result of seawater is shown in Fig. 8b. The concentrations of Na^+ , K^+ , Mg^{2+} and Ca^{2+} are remarkably reduced and far below the standards of drinkable water as defined by the World Health Organization (WHO) and

Figure 8 (a) UV-Vis absorption spectra of the simulated dye wastewater before and after purification; insets show photographs of the simulated dye wastewater and the condensed water. (b) Concentrations of Na^+ , K^+ , Mg^{2+} and Ca^{2+} in the seawater and the condensed water; the dashed lines in (b) indicate the WHO and EPA standards for the healthy drinkable water. Optical images of oil/water emulsion before (c) and after (d) purification; insets show photographs. (e) Stability of Ni/CNM-I in solar vapor generation in water for 10 cycles under 1 kW m^{-2} solar irradiation.

U.S. Environmental Protection Agency (EPA). Besides, no oil droplets are found in the condensed water compared with the oil/water emulsion (Fig. 8c and d). The inset in Fig. 8c shows that light in oil/water emulsion exhibits the obvious Tyndall effect, which is not observed in the condensed water (the inset in Fig. 8d). The evaporation rates in different systems above are close to that of pure water (Fig. S20). The concentration of solution has no significant effects on the evaporation rate (Fig. S21), indicating that Ni/CNM-I possesses a good applicability. The stability of Ni/CNM-I was evaluated by measuring the evaporation rate in deionized water and seawater, respectively. The evaporation rate shows no significant differences after 10 cycles in water (Fig. 8e) or seawater (Fig. S22), demonstrating the good stability of Ni/CNM-I in solar vapor generation.

CONCLUSIONS

In summary, the controlled carbonization of PP by using NiO and PIL-X as combined catalysts has been proposed to prepare Ni/CNM. The yield, morphology and textural structure of Ni/CNM are modulated by adding a trace amount of PIL-X. Ni/CNM-I is obtained by using NiO/PIL-I, and consists of pear-like Ni nanoparticles and CS-CNTs. Due to the synergistic effect of Ni and CS-CNTs in photothermal conversion, Ni/CNM-I possesses an excellent absorption of solar spectrum and a high surface temperature in solar vapor generation. Moreover, Ni/CNM-I constructs the 3D porous network for water supply and vapor channels. Taking advantages of high solar absorption, fast water transport, and low thermal conductivity, Ni/CNM-I exhibits a high evaporation rate of $1.67 \text{ kg m}^{-2} \text{ h}^{-1}$ and a solar-to-vapor conversion efficiency up to 94.9%. More importantly, Ni/CNM-I prevails over the state-of-art carbon-based photothermal materials. This work not only contributes to the field of seawater desalination, but also proposes a new sustainable approach to convert waste polymers into high value-added carbon nanomaterials. We hope this work can inspire the research on the carbonization of waste polymers into advanced carbon-based materials for a wide range of fields such as energy storage and conversion, and environmental remediation.

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Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Supporting data are available in the online version of the paper.

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可控碳化废弃聚丙烯制备镍/碳纳米材料用于高效光蒸气转换

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摘要 利用太阳能实现光蒸气转化是一项极具前景的技术, 可应用于海水脱盐和淡水制备等领域. 然而, 从工业的角度来看, 制备低成本、高效率的光热材料仍具有挑战性. 本文利用聚离子液体(PIL)和氧化镍(NiO)作为复合催化剂, 实现了聚丙烯(PP)的可控碳化, 并制备了镍/碳纳米材料(Ni/CNM). 研究表明, 加入微量的PIL可实现对Ni/CNM形貌和织态结构的调控. Ni/CNM由杯状碳纳米管(CS-CNT)和梨形镍纳米颗粒组成, 二者在太阳光吸收上的协同作用使得Ni/CNM具有优异的光热转换性能. 此外, Ni/CNM具有较高的比表面积和丰富的微/介/大孔, 其构建的三维多孔网络可为水和蒸气的高效传输提供通道. 光吸收高、水传输快和热导率低等优势, 使Ni/CNM的水蒸发速率高达 $1.67 \text{ kg m}^{-2} \text{ h}^{-1}$, 光-蒸气转换效率高达94.9%, 且重复使用10次后性能依然保持稳定. 该材料同时适用于染料废水、海水和油/水乳液等水质的纯化. 其中, 海水中金属离子的去除效率高达99.99%, 染料去除率>99.9%. 更重要的是, 材料的光蒸气转换性能优于最新报道的碳基光热材料. 此工作不仅提出了一种可将废弃聚合物转化为先进的金属/碳杂化物的可持续方法, 同时也有助于太阳能利用和海水淡化领域的进一步研究.