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Recycling of polyolefins using recycled PVC under sunlight

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The recycling of polyolefin plastics is often hindered by high energy consumption and low economic efficiency. In particular, the recycling of poly(vinyl chloride) is especially challenging. Here, we present a protocol to recycle poly(vinyl chloride) into a photothermal agent for the depolymerization of polyolefins under sunlight. By dechlorinating the poly(vinyl chloride), we harness its unique ability to convert sunlight into the energy required to break C-C bonds in polyolefins. This protocol allows for the transformation of low-density polyethylene, high-density polyethylene, and polypropylene into waxes enriched with terminal olefins. Furthermore, it can also enable the conversion of polystyrene, poly(methyl methacrylate), poly(α-methyl styrene), styrene-acrylonitrile copolymer, and acrylonitrile-butadiene-styrene into their respective monomers. This broad applicability makes our protocol suitable for recycling of a wide range of post-consumer plastics and their mixtures under ambient environment. This work provides an effective strategy for polyolefins recycling using waste plastics and solar energy.

Plastic is a highly prevalent material in everyday use due to its low cost, durability, and versatility. Globally, the production of plastic is steadily increasing, with annual output over 360 million tons[1,2]. Polyolefins, such as high-density polyethylene (HDPE), low-density polyethylene (LDPE), poly(vinyl chloride) (PVC), polypropylene (PP), and polystyrene (PS), constitute over half of the total plastic production[3]. Despite their widespread use, the recycling rate of most polyolefin plastics remains low[4]. After use, these plastics are predominantly landfilled, incinerated, or mismanaged (Fig. 1a), leading to significant environmental pollution[5,6]. This is due to the chemical stability of polyolefin plastics, which means that they can take hundreds of years to decompose naturally in the environment[7].

An ideal solution to tackling the problem of plastic waste lies in recycling it for circular development. The methods for recycling plastics encompass both mechanical and chemical processes[8]. However, due to the limitations of sorting technology and the degradation of material properties, mechanical recycling is not an ideal method for recycling large amounts of plastic wastes[9,10]. Chemical recycling and upcycling approaches, which convert polyolefins into monomers or value-added chemicals for reuse, are envisioned as promising solutions to deal with high volume post-consumer wastes[9,11–13]. Recently, several chemical methods have been developed for the conversion of polyolefins, such as pyrolysis[14–16], catalytic cracking[17,18], hydrocracking and hydrogenolysis[19–22], olefin metathesis[23–25], oxidation[26–36], etc. Nevertheless, the high bond dissociation energy (BDE) and low polarity of the chemical bonds in polyolefins (e.g., BDE: ~85 kcal/mol for the C–C bond in PE)[37,38] necessitate substantial energy input for their chemical conversion, impeding plastic recycling efforts (Fig. 1b)[39]. Therefore, the utilization of renewable energy to facilitate the conversion of polyolefins is in urgent demand[40]. Photochemical conversion, which could harness solar energy directly, provides an alternative solution to plastics recycling by avoiding high consumption of fossil fuels, rendering it an optimal choice for facilitating polyolefin conversion[41–46].

Another challenge associated with polyolefin reuse is the recycling of PVC. In the recycling process, it is difficult to avoid mixing PVC with other plastics. However, PVC wastes typically contain a high level of plasticizers, which will affect the recyclability of both PVC itself and

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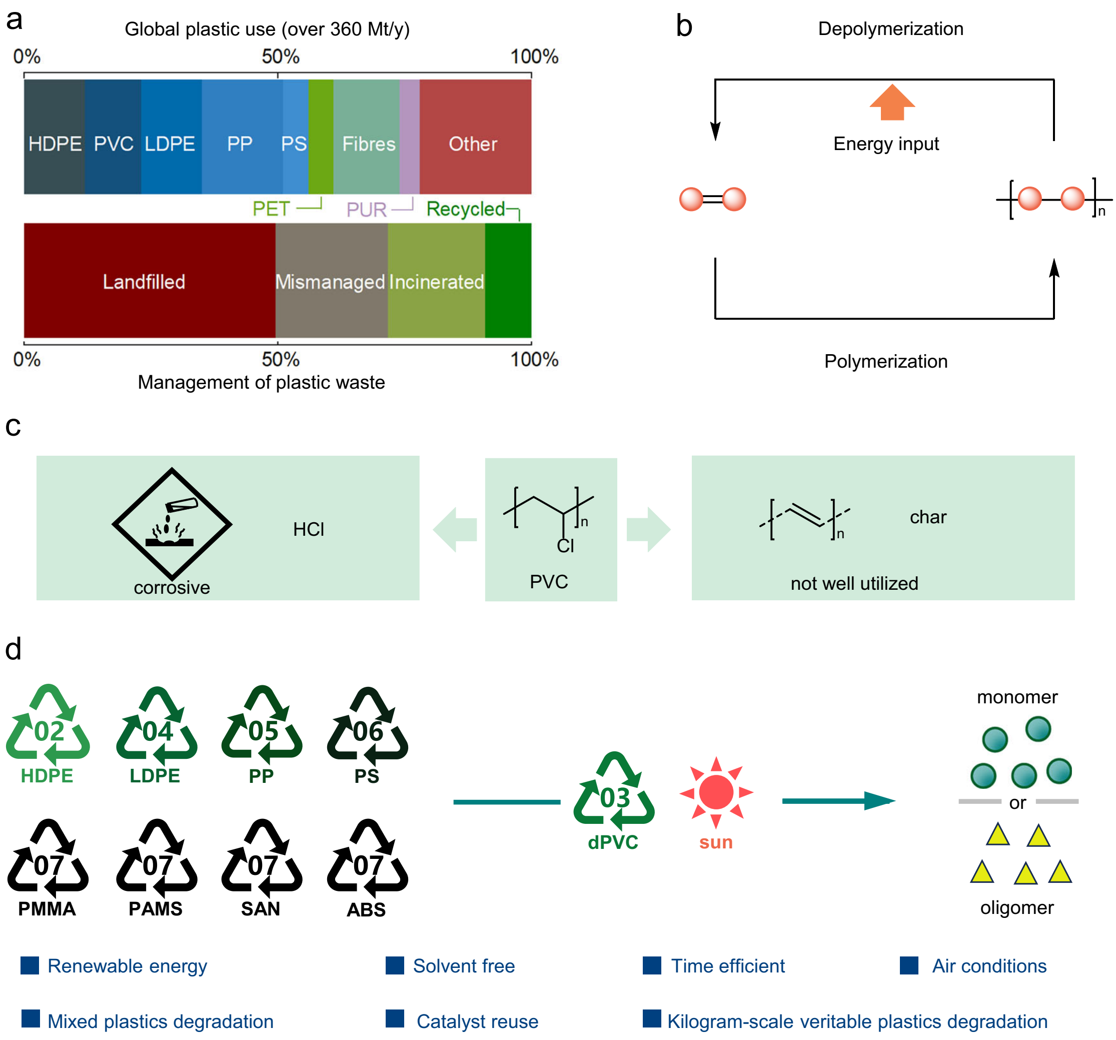

Fig. 1 | The use and recycling of polyolefin plastics. a Global plastic use and management of plastic waste. **b** Closed-loop chemical recycling of polyolefins. **c** Challenges in PVC recycling. **d** This work: A strategy for recycling of polyolefins using recycled PVC under sunlight.

the mixed plastics[47]. Furthermore, PVC is prone to undergoing elimination reactions to generate hydrogen chloride during the recycling process, leading to reactor corrosion and catalyst poisoning[8]. A promising strategy to address this issue is to utilize the chlorine in PVC for other reaction processes that require chlorine. For instance, recent studies have reported using PVC for the chlorination of arenes[48,49] and ketones[50]. The carbon chain, apart from chlorine, is another valuable component in PVC, but it has not been well utilized after dechlorination (Fig. 1c). We hypothesize that PVC after the removal of hydrogen chloride possess abundant conjugated bonds, which exhibits photothermal effects through the thermal vibration mechanism and can be potentially applied to transformation of polyolefins.

Here, we have presented an innovative polyolefin recycling method that utilizes waste PVC and solar energy (Fig. 1d)[51]. This protocol is highly efficient in terms of both time and material usage, capable of converting most of the common conventional plastics covering resin codes 2–7 under ambient pressure and air conditions with sunlight exposure. Using dechlorinated PVC (dPVC) as a reusable photothermal agent, the recycling process of polyolefins was completed within just a few minutes. During this process, HDPE, LDPE, and PP were depolymerized into value-added olefins, while PS, poly(methyl methacrylate) (PMMA), poly(α-methyl styrene) (PAMS), styrene-acrylonitrile copolymer (SAN), and acrylonitrile-butadiene-styrene (ABS) were recycled back into their corresponding monomers. Moreover, this protocol has been successfully applied to the recycling of post-consumer plastic wastes, including single plastics, mixed plastics, and even kilogram-scale plastics. The efficient conversion process utilizing solar energy would facilitate polyolefins recycling and upcycling, reducing white pollution and greenhouse gas emissions.

Results

Polyolefins recycling

Initially, a photothermal agent was obtained from PVC. In order to achieve the comprehensive valorization of PVC, a bromopropane-catalysed chlorine transfer protocol was adopted to enable the efficient reuse of chlorine in PVC[50]. Chlorinated ketones derived from ketones can be used in the synthesis of the nonsteroidal anti-inflammatory drug naproxen. Another raw material, PVC, upon dechlorination, transformed from its original white solid state into an insoluble black polymer. After washing, drying, and grinding, a black powder of dPVC was obtained with a yield of 72 wt% relative to PVC (Fig. 2a). To investigate the capability of dPVC in utilizing solar energy for the conversion of polyolefins, the chemical conversion of PE and PP was first attempted. According to Liu's work[16], PE and PP can be converted into olefins under heating conditions, which can then be oxidized to synthesize fatty acids for the production of surfactants. Utilizing solar energy as the driving force for this transformation will make the approach more practical. A mixture of polyolefin

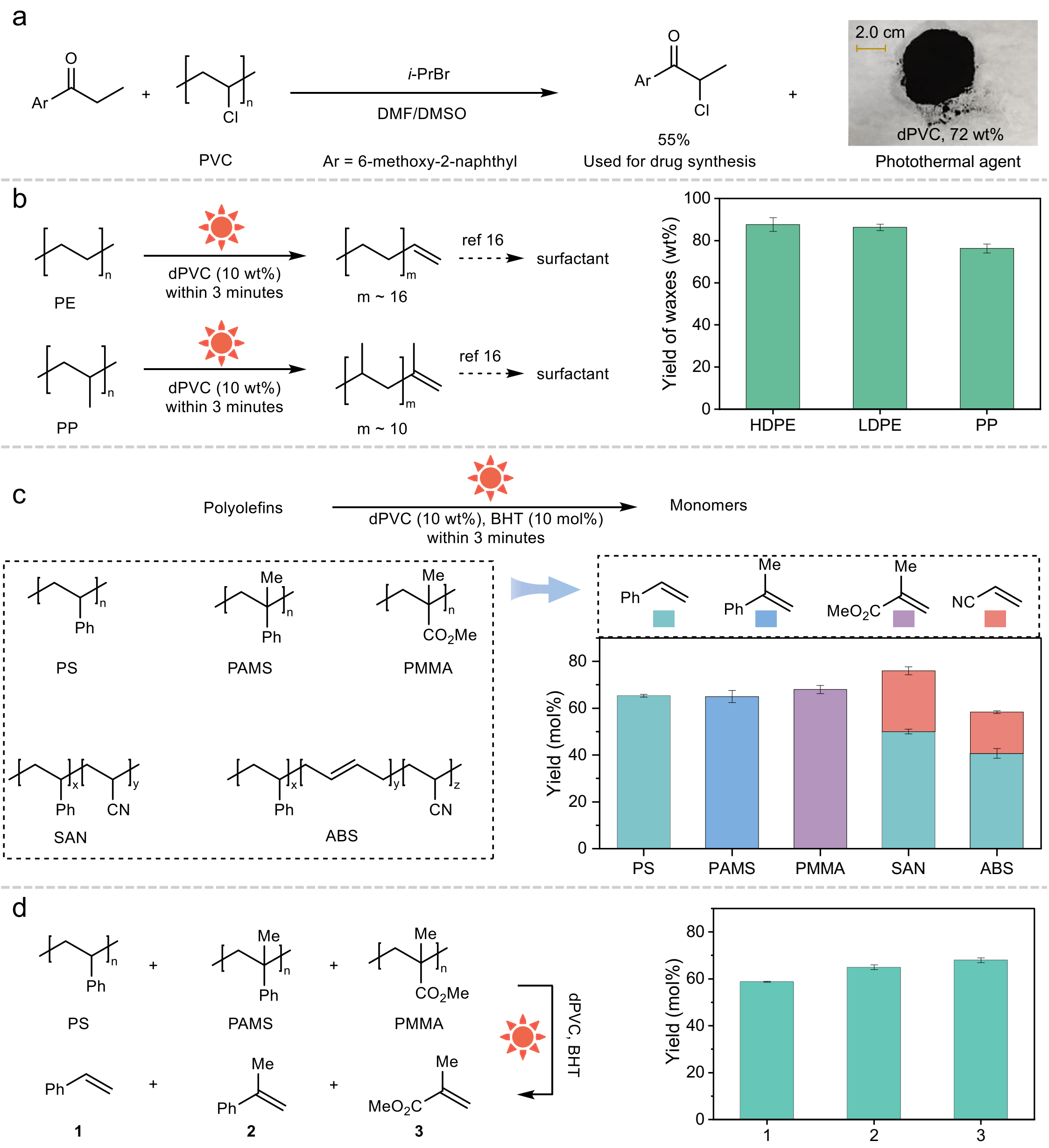

Fig. 2 | Recycling of polyolefins under sunlight. a Synthesis of photothermal agent. Conditions: 6-Methoxy-2-acetyl naphthalene (10.0 mmol), PVC (20.0 mmol repeat unit), *i*-PrBr (20 mol%), DMSO (10.0 mL), DMF (90.0 mL), 120 °C, 12 h. **b** Degradation of PP and PE. Conditions: PE (60.0 mg) or PP (100.0 mg), dPVC 10 wt%). **c** Recycling of polyolefins into their monomers. Conditions: Polyolefin (1.0 mmol repeat unit), dPVC (10 wt%), BHT (10 mol%). The yields were determined by GC-FID with mesitylene as the internal standard. **d** Recycling of mixed polyolefins. Conditions: PS (0.50 mmol repeat unit), PAMS (0.50 mmol repeat unit), PMMA (0.50 mmol repeat unit), dPVC (10 wt%), BHT (10 mol%). The yields were determined by GC-FID with mesitylene as the internal standard. All reactions were conducted under air atmosphere. The depolymerization of polyolefins was performed by focusing sunlight onto the mixture of polyolefins and dPVC using concave mirrors with a diameter of 30 cm. The error bars represent the standard deviation of yield in 3 parallel experiments.

and dPVC (10 wt% with respect to the polyolefin) was placed in a quartz crucible within a flask (Supplementary Fig. 1). When sunlight (100-110 mW cm^{-2}), focused by a concave mirror (with a diameter of 30 cm), was directed onto the reactants in the crucible, white smoke was immediately generated. This smoke likely consisted of fragments from the broken polyolefin that, after escaping the crucible, condensed onto the colder walls of the flask. With continued irradiation, the polyolefin in the crucible gradually disappeared, and waxes accumulated on the flask wall. The entire process, from the start of irradiation to the complete disappearance of the polyolefin in the crucible (the end of the reaction), took only 2-3 minutes. The isolated yields of waxes generated from LDPE (M_n, 51 KDa), HDPE (M_n, 46 KDa), and PP (M_n, 99 KDa) were 86, 88, and 76 wt%, respectively (Fig. 2b). By installing a balloon on the reaction device, it was observed that only a negligible amount of gaseous products might be generated. This phenomenon was presumably due to the fact that after the polyolefin broke down into waxy products, these products immediately escaped from the crucible, thereby preventing the transformation into products with lower molecular weights.

Nuclear magnetic resonance (NMR) spectroscopy was initially used to characterize these waxy products, revealing that the olefins in the waxes were predominantly terminal olefins, and was consistent with Liu's work. The ^{1}H NMR also indicated that there were small amount of aldehydes and aromatic hydrocarbons in the products of

LDPE and HDPE (Supplementary Figs. 2 and 4). Among them, the aldehydes were likely the result of the reaction between the polyolefins and oxygen, while the aromatic hydrocarbons were the result of cyclization and dehydrogenation. Aldehydes and aromatic hydrocarbons were hardly observed in the products of PP, which might be due to the branched-chain structure of PP. Additionally, gas chromatography–mass spectrometry (GC-MS) showed that the waxes were predominantly composed of olefins, with a small amount of alkanes (Supplementary Figs. 8–13). This is likely because the reaction system provided a sufficiently high temperature, which facilitated the β-scission of the radical intermediates, resulting in the formation of olefins[16]. Due to the inability of GC-MS to detect low-volatility heavy hydrocarbons, the waxes were further characterized by Fourier-transform ion cyclotron resonance mass spectrometry (FT-ICR MS) to determine the molecular weight distribution. Based on the spectra, the number-average molecular masses of the waxes from HDPE, LDPE, and PP were 501, 478, and 472, respectively, corresponding to olefins with carbon numbers of 36, 34, and 34 (Supplementary Figs. 15, 17, and 19). It's worth noting that in the absence of dPVC, neither PP nor PE could generate waxy products after being irradiated under concentrated sunlight for three minutes (Supplementary Table 5).

On the basis of the above results, the process using solar energy was applied to other commercially available and high-volume polyolefin recycling. Homopolymers PS, PAMS, and PMMA, when mixed with dPVC and butylated hydroxytoluene (BHT), decomposed into corresponding monomers and condensed on the flask walls under concentrated sunlight. As analyzed by gas chromatography (GC) with mesitylene as the internal standard, the monomer yields were determined to be 65%, 65% (isolated yield: 65%), and 68%, respectively. However, they were failed to yield monomers without dPVC (Supplementary Table 6). BHT was added to prevent the monomers from repolymerizing. When BHT was absent, the yield of monomer was decreased (Supplementary Table 2, entry 1). The commonly used binary copolymer SAN in daily life also decomposed into styrene and acrylonitrile, with yields of 50% and 26%, respectively. Terpolymer ABS, under the combined action of dPVC and sunlight, could yield 41% styrene and 18% acrylonitrile (Fig. 2c). Given the prevalence of mixed plastic waste in everyday life, achieving degradation of mixed plastics can effectively avoid the difficulties in plastic recycling caused by classification[52]. To our delight, mixtures of polyolefins with different monomers could also be depolymerized using dPVC as a photothermal agent and sunlight as an energy source. The yields of styrene (59%), α-methylstyrene (65%), and methyl methacrylate (68%) were comparable to those of single polyolefin depolymerization monomers (Fig. 2d). These results indicate that the dPVC can depolymerize various polyolefins without interference, suggesting significant application potential for this protocol in the field of mixed plastic recycling.

Post-consumer plastics recycling

Post-consumer plastics often contain additives such as dyes, antioxidants, and plasticizers, whose presence may hinder the depolymerization process, and therefore pose significant challenges for the recycling and upcycling of polyolefins[53]. To further explore the potential practical applications, it was attempted for the recovery of post-consumer plastic waste from daily life. The initial attempt involved recycling PVC waste plastics to synthesize photothermal agents, which were then used for the recovery of other polyolefin plastics. PVC gloves (PVC, 57 wt%, M_n, 92 kDa; DOTP, 41 wt%), PVC tubings (PVC, 63 wt%, M_n, 53 kDa; DEHP, 35 wt%), and PVC mats (PVC, 66 wt%, M_n, 57 kDa; DPHP, 34 wt%), after undergoing chloride transfer, all successfully generated photothermal agents. Despite the presence of significant amounts of plasticizers and varying molecular masses of PVC in these plastic products, the resulting agents were all capable of depolymerizing PS under sunlight exposure. Depolymerization experiments under sunlight showed that the yields of styrene were 58%, 56%, and 61%, respectively (Fig. 3a). These results indicate that the synthesis of photothermal agents from PVC waste plastics has good applicability. Subsequently, dPVC synthesized from discarded PVC gloves was used to attempt the recycling of common polyolefin plastic products in daily life (Fig. 3b). Under sunlight irradiation, the HDPE plastic bucket was recycled to yield waxes containing terminal olefins with 83 wt% isolated yield. The LDPE plastic bag and PP food container underwent chemical conversion to produce waxes with isolated yields of 84 wt% and 70 wt%, respectively. A PS cup was subjected to sunlight irradiation to recover 60% styrene. An identical trial with a used plastic sheet made from PMMA provided 80% methyl methacrylate. These results indicate that the method is well-suited for the recycling of real polyolefin plastics in everyday life.

Considering that daily waste recycling usually collects polyolefin mixtures, which are extremely difficult to separate. Finding a generic and profitable method to recycle polyolefin mixtures is thus imperative. To this end, a recycling scheme for mixed plastics is proposed (Fig. 3c). After mixing commercial plastics, including HDPE, LDPE, PP, PVC, PS, and PMMA, a chlorine transfer reaction occurred in a mixed solvent of DMF and DMSO, where the PVC was utilized to produce α-chloroketone and photothermal agent. Methanol was then added to the system to precipitate out the unreacted polyolefins and dPVC. The filtered mixture of dPVC and polyolefins was directly irradiated under concentrated sunlight, ultimately yielding 71 wt% waxes, 42% styrene, and 58% methyl methacrylate. This approach can avoid the difficult sorting tasks in plastic recycling and prevent the generation of large amounts of hydrogen chloride when recycling PVC-containing mixed plastics, promoting the recovery of daily plastics. To further demonstrate the practicality of this reaction, a kilogram-scale polyolefin depolymerization experiment was attempted using a distillation apparatus (Supplementary Fig. 43). By employing batch feeding, the decomposition of one kilogram of PS cups was achieved with only loading 0.1 wt% of dPVC under sunlight, yielding 0.97 kilograms of liquid product (Fig. 3d). The primary component of this liquid product was styrene, accounting for 57 wt% of the mixture, with a recovery rate of 55%. Additionally, the liquid product contained other valuable chemicals such as toluene, ethylbenzene, and α-methylstyrene (Supplementary Fig. 44). The above experimental results fully demonstrate that the protocol for converting polyolefins by dPVC under sunlight has great practicality in the recycling and upcycling of PVC, HDPE, LDPE, PP, PS, and PMMA waste plastics.

Photothermal effect and recycling test of dPVC

In order to evaluate the solar-thermal conversion ability of dPVC, the light absorption ability of dPVC and recycled dPVC (recycled from the depolymerization experiment of PS) were measured with a UV-Vis-NIR spectrophotometer. As shown in Fig. 4a, the black dPVC powder exhibited excellent absorption of sunlight over the whole solar spectrum. In the visible light region (400-700 nm), which accounts for 45% of the total energy of sunlight, the light absorption rate of dPVC exceeded 90%. Even in the short-wavelength near-infrared region (700-1100 nm), dPVC still showed good light absorption, but the absorption rate gradually decreased with increasing wavelength in long-wavelength near-infrared region (1100-2500 nm). After the depolymerization of PS, the recycled dPVC maintained its light absorption capability in the visible light region, and its light absorption capacity in the infrared region significantly increased. This may be due to partial unremoved chlorine undergoing further removal under sunlight, resulting in a higher degree of conjugation by generating more double bonds. This suggest that the dPVC may exhibit improved photothermal conversion ability after use.

Under concentrated sunlight exposure, dPVC absorbed sunlight and converted it into thermal energy, rapidly raising the temperature of the reaction system to 470 °C within 1 minute and maintaining this temperature until the reaction concluded (Fig. 4b). The entire process

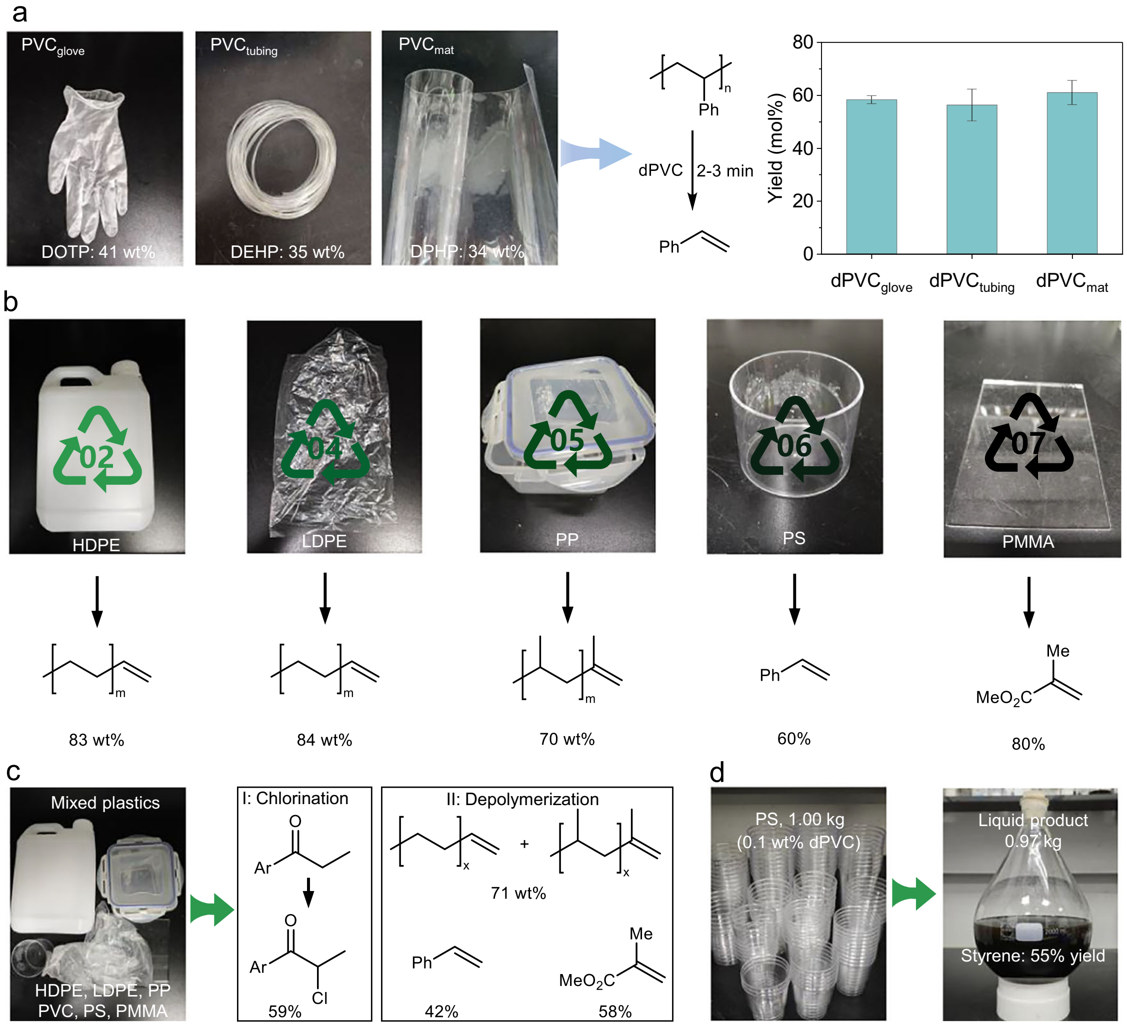

Fig. 3 | Recycling of real-life plastic waste under sunlight. a Comprehensive valorization of PVC waste. The yields were determined by GC-FID with mesitylene as the internal standard. **b** Post-consumer plastic waste depolymerization. **c** Recycling of mixed post-consumer plastic waste. **d** Kilogram-scale degradation of PS cups. DOTP = di(2-ethylhexyl) terephthalate. DEHP = di(2-ethylhexyl) phthalate. DPHP = di(2-propylheptyl) phthalate. The error bars represent the standard deviation of yield in 3 parallel experiments. All reaction conditions are available in Supplementary Information.

from initial illumination to reaction completion took approximately 2 minutes. Since the temperature increase was a result of dPVC converting light energy into heat, the high temperature was concentrated only in the crucible where the photothermal agent was present, while on the flask walls, the temperature remained at 75 °C (Fig. 4c). This created a temperature gradient, preventing further reaction of the depolymerization products and enhancing the yield of the products. Thermogravimetric analysis (TGA) was used to estimate the stability of dPVC. The thermogravimetric curve indicated that weight loss began at 126 °C and reached 42% by 380 °C. As the temperature increased, further weight loss occurred until the weight stabilized at 650 °C. In contrast, the recycled dPVC (recycled from the depolymerization experiment of PS) exhibited significantly enhanced thermal stability, with only 6% weight loss by 330 °C, possibly due to adsorbed water or residual solvent. Even at 680 °C, 76% of the material's mass was retained. The enhanced thermal stability is presumably ascribed to the removal of residual chlorine in dPVC under solar irradiation. This chlorine elimination was confirmed in subsequent experiments. Both UV-Vis-NIR absorption spectroscopy and TGA indicate that the structure of dPVC had undergone changes after participating in the reaction. An experimental investigation was conducted to explore whether its photothermal conversion has also changed. Experiments demonstrated that, without additional supplementation of dPVC, the photothermal agent consistently achieved the efficient depolymerization of PS over 10 successive reuse cycles (Fig. 4d). The yield of styrene in some experiments fluctuated, possibly due to occasional cloud cover blocking the sun. These results suggest that the dPVC underwent structural changes during its first exposure to sunlight for the depolymerization of polyolefins, increasing its light absorption capacity. However, its ability to depolymerize polyolefins under sunlight had not been lost, and thus it could be reused in the recycling of polyolefins.

Characterization of dPVC

Scanning electron microscopy (SEM) images (Fig. 5a–b) presented the microscopic morphological features of dPVC. It can be seen from Fig. 5a that the dPVC powder were predominantly in the form of irregular particles with diameters of 10–30 μm. These particles had rough surfaces, which enhance light absorption through multi-scattering effects. The recycled dPVC remained as irregular particles with rough surfaces (Fig. 5b). Elemental mapping images obtained through X-ray energy dispersive spectroscopy (EDS) showed a uniform distribution of characteristic elements, indicating the PVC after chlorine transfer was primarily composed of carbon, retained a portion of chlorine, and

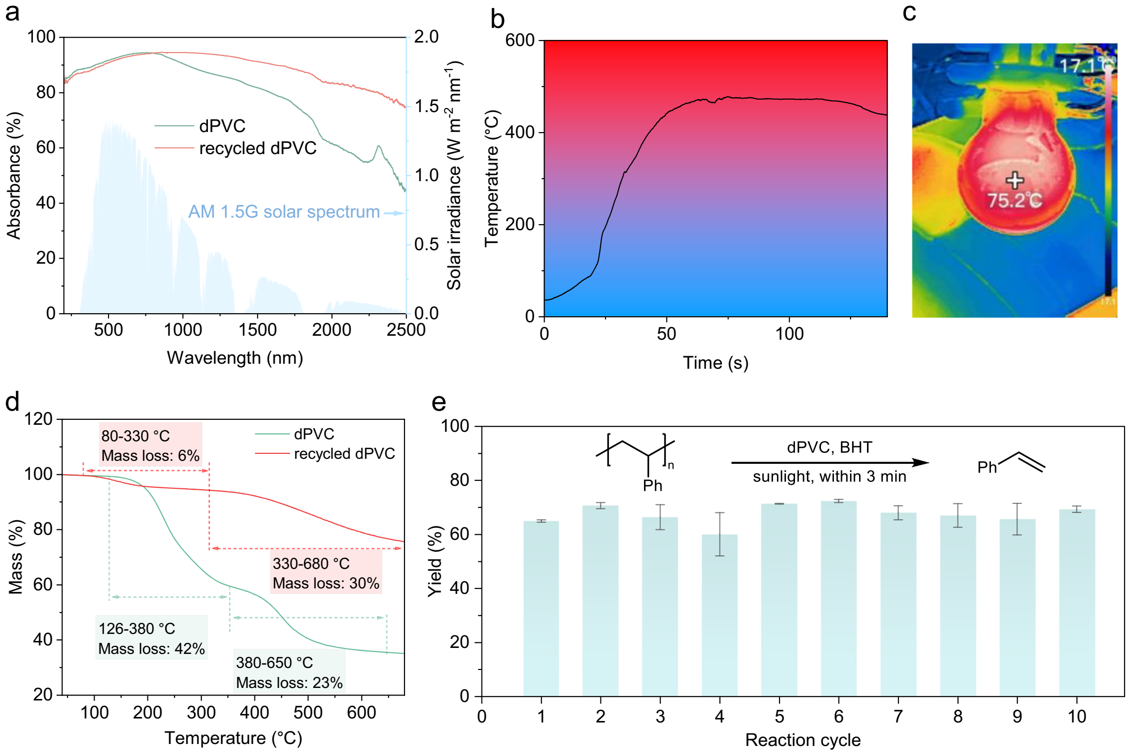

Fig. 4 | Photothermal effect and stability of dPVC. a UV-Vis-NIR spectra of dPVC. **b** Temperature curve of PS depolymerization. **c** Infrared thermal image of the reactor. **d** Thermal mass loss profile of dPVC. **e** Reuse of dPVC. The yields were determined by GC-FID with mesitylene as the internal standard. The error bars represent the standard deviation of yield in 3 parallel experiments.

introduced a small amount of oxygen (Fig. 5c). After participating in the depolymerization of PS under sunlight, chlorine element in dPVC was almost completely eliminated, while carbon and oxygen elements were retained (Fig. 5d). No chlorinated products were detected in the degradation products, and hardly any were generated even with the additional introduction of PVC (Supplementary Table 11). Moreover, given that the chlorine content in dPVC is extremely low relative to that of the recycled polyolefin plastics, the migration of residual chlorine from dPVC to the products is negligible. To further identify the chemical structures of the photothermal agent, ^{13}C CP/MAS NMR was conducted on dPVC. As shown in Fig. 5e, a prominent signal corresponding to sp^2 C was detected at 135 ppm, which suggested a significant presence of C = C bonds within dPVC. Additionally, dPVC exhibited strong signals indicative of sp^3 C (57, 46 ppm), implying that not all hydrogen chloride in PVC was eliminated to form double bonds.

This observation was further substantiated by Fourier transform infrared spectroscopy (FT-IR) analysis for investigation on the structural changes in dPVC after being used in depolymerization of polyolefins (Fig. 5f). The FT-IR spectra of dPVC and recycled dPVC both showed weak C(sp^2)-H stretching vibrations from 3000-3050 cm^{-1} and corresponding bending vibrations in the range of 685-760 cm^{-1}. In the FT-IR spectrum of dPVC, peaks at 2908 cm^{-1} and 1426 cm^{-1} represented the stretching and bending vibrations of C(sp^3)-H, respectively, but these signals became very weak in the recycled dPVC. This indicated a transformation of C-H bonds in dPVC. With respect to C = C bonds stretching vibrations, the relative peak intensity at 1580-1600 cm^{-1} in the recycled dPVC was stronger compared to that in dPVC, indicating an increase in the number of double bonds in the recycled dPVC. Additionally, strong C-Cl bond stretching vibration (607 cm^{-1}) was only detected in dPVC, further confirming the elimination of chlorine during the reaction, which resulted in the formation of more double bonds. Moreover, peaks at 1724 cm^{-1} ascribed to the stretching vibration of C = O and at 1240 cm^{-1} belonging to C-O stretching vibrations in both dPVC and recycled dPVC indicated that oxygen was bonded to carbon in the forms of C = O and C-O in dPVC. Raman spectrum was used to further characterize the chemical structure of the dPVC (Fig. 5g). The D band (at 1360 cm^{-1}) and G band (at 1590 cm^{-1}) reflected the sp^3-hybridized amorphous carbon structure and sp^2-hybridized C = C structure in dPVC respectively. Figure 5g presented that there were two bands within both samples, showing the occurred graphitization process from disordered structure to graphitic structure[54,55]. To investigate whether the graphitized dPVC possesses photocatalytic activity, its photocurrent response was tested. Under xenon lamp irradiation, the photocurrent of dPVC was extremely weak, which was much lower than that of graphitic carbon nitride (g-C_3N_4), a commonly used photocatalyst. Electrochemical impedance spectroscopy (EIS, Supplementary Fig. 57) demonstrated that dPVC exhibited a significantly higher charge transfer resistance compared to g-C_3N_4. Collectively, these results indicate that dPVC lacks efficient photocatalytic activity. Therefore, dPVC primarily functions as a photothermal conversion agent in the degradation of polyolefins.

Comparison of dPVC with other photothermal agents

Compared with some photothermal agents reported in the literature[14,56], dPVC exhibits certain advantages in terms of efficiency, stability, or cost-effectiveness for polyolefin recycling. To verify this, the efficiency of PS conversion to styrene was compared in the presence of different photothermal agents under sunlight irradiation (Supplementary Table 8). Specifically, dPVC exhibited higher monomer generation efficiency than carbon black (CB). Carbon nanotubes (CNTs) showed comparable performance to dPVC, but their high cost limits practical applicability. Carbon quantum dots (CQDs) also effectively facilitated the conversion of PS to monomers. However, they exhibited poor stability at high temperatures, making them

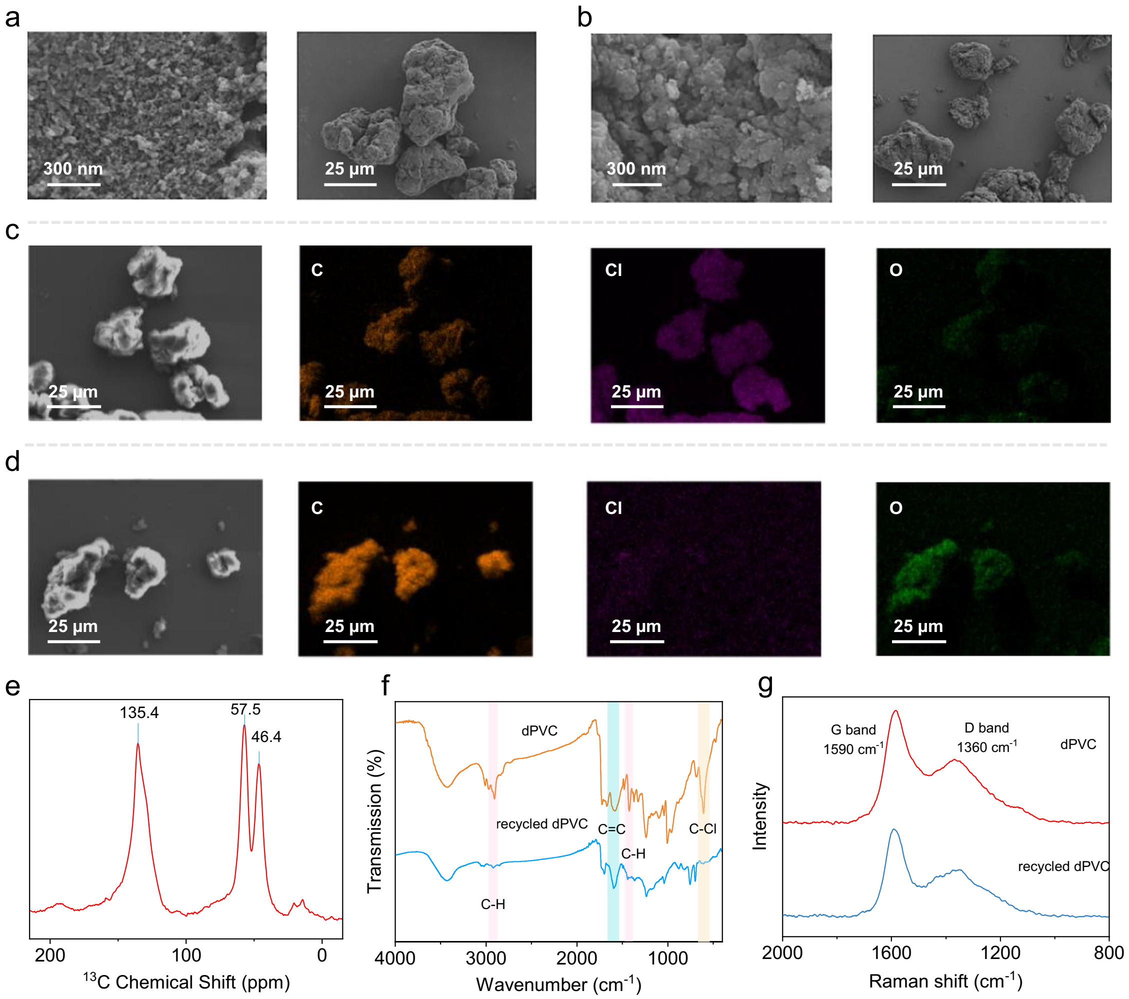

Fig. 5 | Characterization of dPVC. a SEM images of dPVC. **b** SEM images of recycled dPVC. **c** SEM image and corresponding energy dispersive X-ray spectroscopy elemental mapping images of dPVC. **d** SEM image and corresponding energy dispersive X-ray spectroscopy elemental mapping images of recycled dPVC. **e** ^{13}C NMR spectrum of dPVC. **f** FT-IR spectra of dPVC and recycled dPVC. (**g**) Raman spectra of dPVC and recycled dPVC.

challenging to reuse. These experimental results collectively demonstrate that dPVC is an excellent photothermal reagent for polyolefin recycling. Furthermore, the synthesis process of dPVC enables the full utilization of PVC, and it can be used to recycle PVC-containing mixed waste plastics without generating large quantities of hydrogen chloride.

Discussion

In conclusion, we have established a protocol for the synthesis of photothermal agents derived from waste PVC plastics, which possess the remarkable ability to facilitate the recycling of polyolefins when exposed to sunlight. This approach enables the chemical transformation of various polyolefins, including LDPE, HDPE, PP, PS, PMMA, SAN, and ABS under atmospheric pressure and ambient air conditions, solely utilizing solar energy. Moreover, this photothermal system can be extended to real-life plastic recycling scenarios. With the aid of just 0.1 wt% photothermal agent, the successful depolymerization of kilogram-scale PS further demonstrates the practicality of this approach. The key to the process is that after chlorine transfer in PVC, the resulting dPVC characterized by a rough surface microstructure and a multitude of conjugated bonds, thereby enhancing its capacity for sunlight absorption and conversion. Notably, dPVC exhibits increased stability throughout its utilization, while maintaining consistent photothermal efficiency, allowing for its repeated use. This research effectively repurposes waste plastics to facilitate their own recycling. By harnessing solar energy, this approach circumvents the reliance on fossil fuels, holding the promise of reducing greenhouse gas emissions and improving the economic viability of plastic recycling initiatives. It is anticipated that this advancement will substantially enhance the development of plastic recycling facilities and contribute to the reduction of environmental pollution attributable to plastic wastes.

Methods

Synthesis of dPVC

Typically, 6-methoxy-2-acetyl naphthalene (2.14 g, 10.0 mmol), *i*-PrBr (188 μL, 2.0 mmol, 20 mol%), PVC (1.25 g, 20.0 mmol repeat unit, 2.0 equiv.), DMSO (10 mL) and DMF (90 mL) were added to a 250 mL round bottom flask equipped with a Teflon coated magnetic stir bar and a condenser. Under vigorous stirring, the reaction mixture was heated at 120 °C for 12 h. After cooling to room temperature, the yield of *α*-chloroketone was determined by HPLC analysis of the crude product mixture using naphthalene as internal standard, and then the reaction mixture was poured into 100 mL methanol while stirring to precipitate the polymer. The insoluble polymer was collected by filtration and then ultrasonically cleaned in 100 mL methanol, followed by filtration. This process was repeated until no more ketones were detected in the solution by TLC. Finally, the polymer was collected by filtration and dried under vacuum overnight. The mass of dPVC for this reaction was 900 mg (yield: 72 wt%). Prior to its use as a catalyst, dPVC should be ground for at least 5 minutes using a ceramic mortar.

Depolymerization of PP and PE

Typically, depolymerization of PP, PE and their post-consumer plastics under air is described as follows. The polyolefin (60.0 mg) and dPVC (6.0 mg, 10 wt%) powders were weighed into a 4 mL sample vial. The mixture was stirred with a metal spatula until a homogeneous color was achieved, and then transferred to a crucible (8 × 8 × 6 mm) (Supplementary Fig. 1c), which was then positioned inside a 25 mL round-bottom flask (Supplementary Fig. 1b) equipped with an air condenser and secured to an iron stand. A concave mirror with a diameter of 30 cm was used to concentrate sunlight (100-110 mW cm^{-2}) onto the quartz crucible (Supplementary Fig. 1a). When only a small amount of black powder remained in the crucible, the light exposure ceased, marking the end of the reaction (Supplementary Fig. 1f). After cooling to room temperature, the white smoke dissipated, and waxy substances condensed on the walls of the flask and the condenser. The waxes were dissolved in hot petroleum ether, filtered to remove the dPVC, and the solvent was removed by rotary evaporation. The resulting product was vacuum dried.

Depolymerization of PS, PAMS, PMMA, SAN, and ABS

Typically, degradation of PS, PAMS, PMMA, SAN, ABS and post-consumer PS and PMMA plastics under air are described as follows. The polyolefin (1.0 mmol repeat unit), dPVC (10 wt%) and butylated hydroxytoluene (BHT, 10 mol%) powders were weighed into a 4 mL sample vial. The mixture was stirred with a metal spatula until a homogeneous color was achieved, and then transferred to a crucible (8*8*6 mm), which was then positioned inside a 25 mL round-bottom flask equipped with an air condenser and secured to an iron stand. A concave mirror with a diameter of 30 cm was used to concentrate sunlight (85-110 mW cm^{-2}) onto the quartz crucible. When only a small amount of black powder remained in the crucible, the light exposure ceased, marking the end of the reaction. After cooling to room temperature, the crude products were dissolved in ethyl acetate (EA). The yields were determined by GC-FID analysis of the crude product mixture using mesitylene as internal standard.

Depolymerization of mixed post-consumer plastics

Depolymerization of mixed post-consumer plastics under air is described as follows. A sealed tube (15 mL) was charged with HDPE bucket (50.0 mg), LDPE bag (50.0 mg), PP food container (50.0 mg), PS cup (50.0 mg), PMMA sheet (50.0 mg), PVC glove (50.0 mg), 6-methoxy-2-acetyl naphthalene (42.9 mg, 0.20 mmol), *i*-PrBr (3.8 μL, 0.04 mmol, 20 mol%), DMSO (0.4 mL) and DMF (3.8 mL) at room temperature. The reaction tube was sealed with a Teflon screw cap. Then, the reaction mixture was stirred at 120 °C for 12 hours. After cooling to room temperature, the yield of α-chloroketone was determined by HPLC analysis of the crude product mixture using naphthalene as internal standard. Then, the reaction mixture was poured into 50 mL methanol while stirring to precipitate the polymer. The insoluble solid was collected by centrifugation (3700 × g, 6 min) and then ultrasonically cleaned in 50 mL of methanol, followed by centrifugation. This process was repeated until no more ketones were detected in the solution by TLC. Finally, the solid was collected by centrifugation and dried under vacuum overnight. The black solid was placed in a quartz crucible (10*10*10 mm), which was then positioned inside a 25 mL round-bottom flask equipped with an air condenser and secured to an iron stand. A concave mirror with a diameter of 30 cm was used to concentrate sunlight (113 mW cm^{-2}) onto the quartz crucible When only a small amount of black powder remained in the crucible, the light exposure ceased, marking the end of the reaction.

Depolymerization of kilogram-scale PS plastic

The depolymerization scheme for kilogram-scale PS is as follows. The scheme employed a distillation setup and utilizes solar panels to power the condensation water circulation. 1.00 kg post-consumer PS (*M*n, 116 kDa; *M*w, 234 kDa) cups was pulverized, and then 200.00 g of the PS powder, along with 22.00 g of BHT and 1.00 g of dPVC made from PVC gloves, were mixed until a homogeneous color was achieved, and added to a 500 mL quartz three-necked flask. Under a nitrogen atmosphere and with continuous condensation water circulation, sunlight (111 mW cm^{-2}) was concentrated at the bottom of the flask using a concave mirror with a diameter of 1.5 meters. The styrene generated was condensed in the condenser and collected. After the polymer in the flask had mostly disappeared, the light was paused (this process would take about 30 minutes), and after the flask cooled, 200.00 g of PS was added for reaction. This process was repeated until 1.00 kg of PS completely reacted. The yield of styrene was determined by ^{1}H NMR analysis using 1,3,5-trimethoxybenzene as the internal standard.

All methods are included in the Supplementary Information.

Data availability

All data generated in this study are provided in the Article and its Supplementary Information. Raw FT-ICR MS, GC-MS, FT-IR, Raman, SEM, TGA, UV-Vis-NIR, and temperature curve data are available via Figshare at https://doi.org/10.6084/m9.figshare.30231052 (ref. 57). All data are available from the corresponding author upon request.

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Author contributions

Q.-A.C. conceived and supervised the project. Q.-A.C. and H.L. designed the experiments. H.L., S.-Y.X., and Z.-H.W. performed the experiments and analyzed the data. D.-W.J. and S.-Y.G. reviewed and edited the paper. All authors discussed the results and commented on the manuscript.

Competing interests

The authors declare no competing interests.

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