Recycling of Polyolefins Using Recycled PVC under Sunlight

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ABSTRACT

The recycling of polyolefin plastics is often hindered by high energy consumption and low economic efficiency. In particular, the recycling of poly(vinyl chloride) (PVC) is especially challenging. Here, we present an innovative protocol to recycle discarded PVC into a photothermal catalyst for the depolymerization of polyolefins under sunlight. By dechlorinating the PVC, we harness its unique ability to converts sunlight into the energy required to break C-C bonds in polyolefins. This protocol allows for the transformation of low-density polyethylene (LDPE), high-density polyethylene (HDPE), and polypropylene (PP) into waxes enriched with terminal olefins, which can be further utilized for the synthesis of surfactant. Furthermore, this protocol enables the conversion of polystyrene, poly(methyl methacrylate), poly(α-methyl styrene), styrene-acrylonitrile copolymer, and acrylonitrile-butadiene-styrene (ABS) into their respective monomers. This broad applicability makes our protocol suitable for the recycling of a wide range of real-life plastic products and their mixtures under ambient environment. Notably, with the aid of just 0.1 wt% catalyst, a successful depolymerization of kilogram-scale PS waste, such as plastic cups, using solar energy has been achieved. Additionally, the catalysts derived from PVC can be reused, thereby enhancing the sustainability and cost-effectiveness of the depolymerization of polyolefins. Our study presents a promising solution to the recycling of PVC and polyolefin plastics, leveraging solar energy to create a sustainable and efficient recycling process.

INTRODUCTION

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 (*1, 2*). Polyolefins, such as high-density polyethylene (HDPE), low-density polyethylene (LDPE), poly(vinyl chloride) (PVC), polypropylene (PP), and polystyrene (PS), make up 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 they can take hundreds of years to decompose naturally in the environment (*7*).
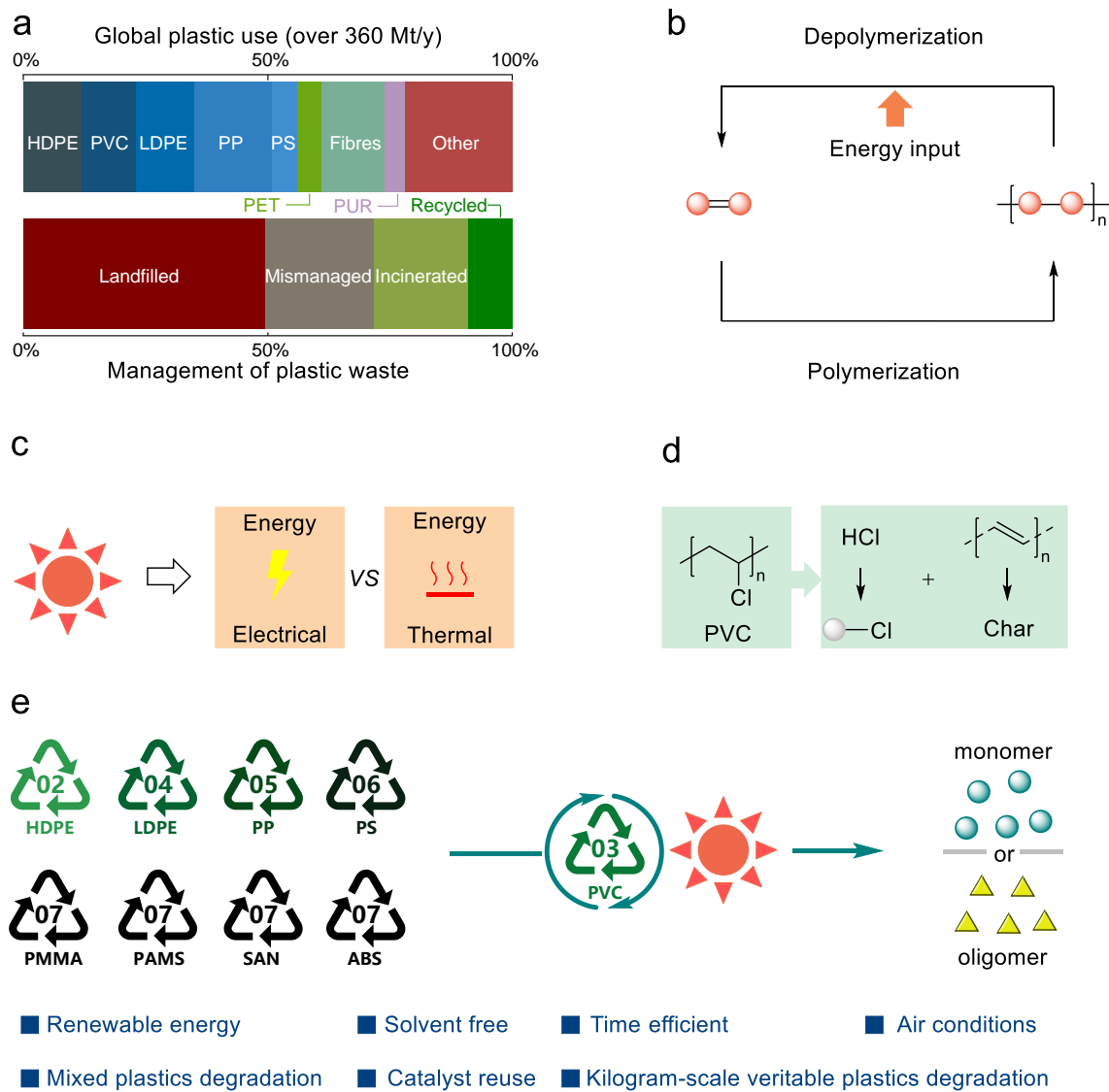
Fig. 1 The use and recycling of polyolefin plastics. (a) Global plastic use and management of plastic waste. (b) Closed-loop chemical recycling of polyolefins. (c) Solar energy conversion. (d) Recycling of PVC. (e) This work: recycling of polyolefins using recycled PVC under sunlight.

An ideal solution to tacking the problem of plastic waste lies in recycling it for circular development. The methods for recycling plastic 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 convert polyolefins into monomers or value-added chemicals for reuse, are envisioned as promising solutions to deal with high volume postconsumer 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 (*26-31*), and so on. Nevertheless, the high bond dissociation energy (BDE) and low

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polarity for the chemical bonds in polyolefins (e.g., BDE: ~85 kcal/mol for the C–C bond in PE) (*32, 33*) necessitate substantial energy input for their chemical conversion (Fig. 1b) (*34*). Consequently, the use of conventional energy sources for the chemical recycling and upcycling of polyolefins is economically inefficient and will lead to significant greenhouse gas emissions, thereby impeding plastic recycling efforts. Therefore, the utilization of renewable energy to facilitate the conversion of polyolefins could greatly propel the advancement of plastic recycling.

Solar energy, an abundant, clean, and renewable resource, provides an alternative solution to plastics recycling, by avoiding high consumption of fossil fuels (*35-37*). Currently, solar energy is commonly transformed into electricity and thermal energy for various applications (Fig. 1c). Although converting solar energy into electricity represents a valuable method for energy utilization, it faces conversion efficiency limitations, with the theoretical Shockley-Queisser maximum efficiency limit of p-n junction solar cells is approximately 33% (*38*). Consequently, solar energy cannot be fully utilized for high-energy-consumption plastic recycling. In contrast, the transformation of solar energy into heat does not encounter such constraint (*39, 40*), rendering it an optimal choice for facilitating polyolefin conversion.

Another challenge associated with the reuse of polyolefins is the recycling of PVC. Specifically, PVC plastics often contain high levels of plasticizers, which compromise their recyclability (*41*), Furthermore, PVC is prone to undergo 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 (*42, 43*) and ketones (*44*). The carbon chain, apart from chlorine, is another valuable component in PVC, but it has not been well utilized after dechlorination (Fig. 1d). We hypothesize that PVC after the removal of hydrogen chloride possess abundant conjugated π bonds, which exhibits photothermal effects through the thermal vibration mechanism and can be potentially applied to photocatalytic transformation of polyolefins.

Here, we have presented an innovative polyolefin recycling method that utilizes waste PVC and solar energy (Fig. 1e). 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 catalyst, 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 real-life polyolefin plastic wastes, including

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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.
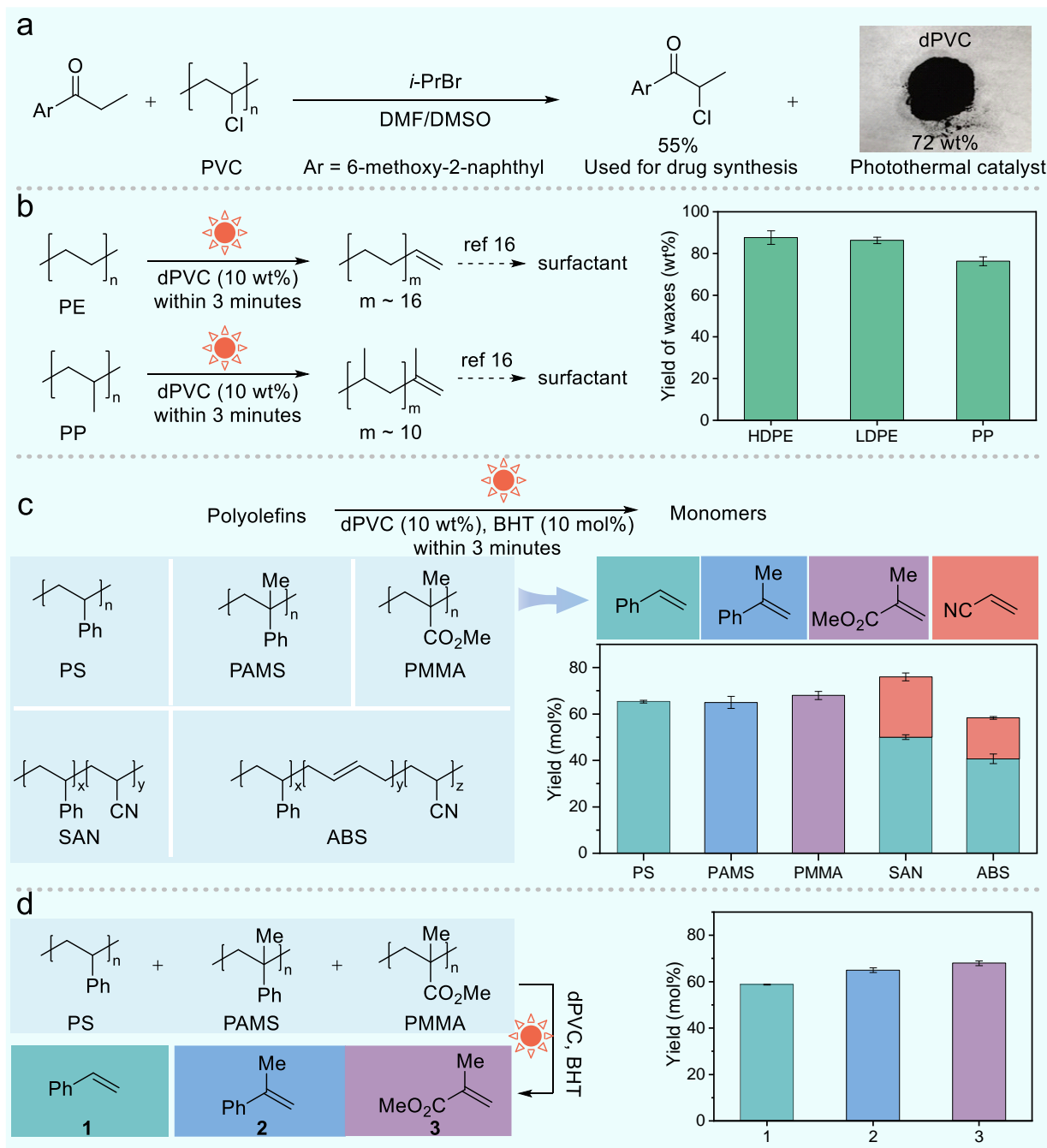

Fig. 2 Recycling of polyolefins under sunlight. (a) Synthesis of photothermal catalyst. (b) Degradation of PP and PE. (c) Recycling of polyolefins into their monomers. (d) Recycling of mixed polyolefins.

Initially, a photothermal catalyst was attempted to synthesize from PVC. Through a chlorine transfer process catalyzed by 2-bromopropane, PVC reacted with 6-methoxy-2-acetyl naphthalene in the mixed solvent of DMSO and DMF. Chlorinated ketones derived from ketones can be used in the synthesis of the nonsteroidal anti-inflammatory

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drug naproxen (*44*). 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 the PVC (Fig. 2a). To investigate the capability of dPVC in utilizing solar energy for the catalytic degradation of polyolefins, the chemical conversion of PE and PP was first attempted. According to the 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 and dPVC (10 wt% with respect to the polyolefin) was placed in a quartz crucible within a flask (Figure S1). 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 consists 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 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). Nuclear magnetic resonance (NMR) spectroscopy was initially used to characterize these products, revealing that the olefins in the waxes were predominantly terminal olefins, consistent with Liu's work. Additionally, gas chromatography–mass spectrometry (GC-MS) showed that the waxes contained a small amount of alkanes in addition to olefins (Figures S8-S13). 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 (Figures S15, S17, and S19).

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. The yields of monomers were 65%, 65%, and 68%, respectively. The commonly used binary copolymer SAN in daily life also decomposed into styrene and acrylonitrile under these conditions, 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 (*45*). To our delight, mixtures of polyolefins with different monomers could also be depolymerized using dPVC as a photocatalyst and

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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 catalyst can depolymerize various polyolefins without interference, suggesting significant application potential for this protocol in the field of mixed plastic recycling.
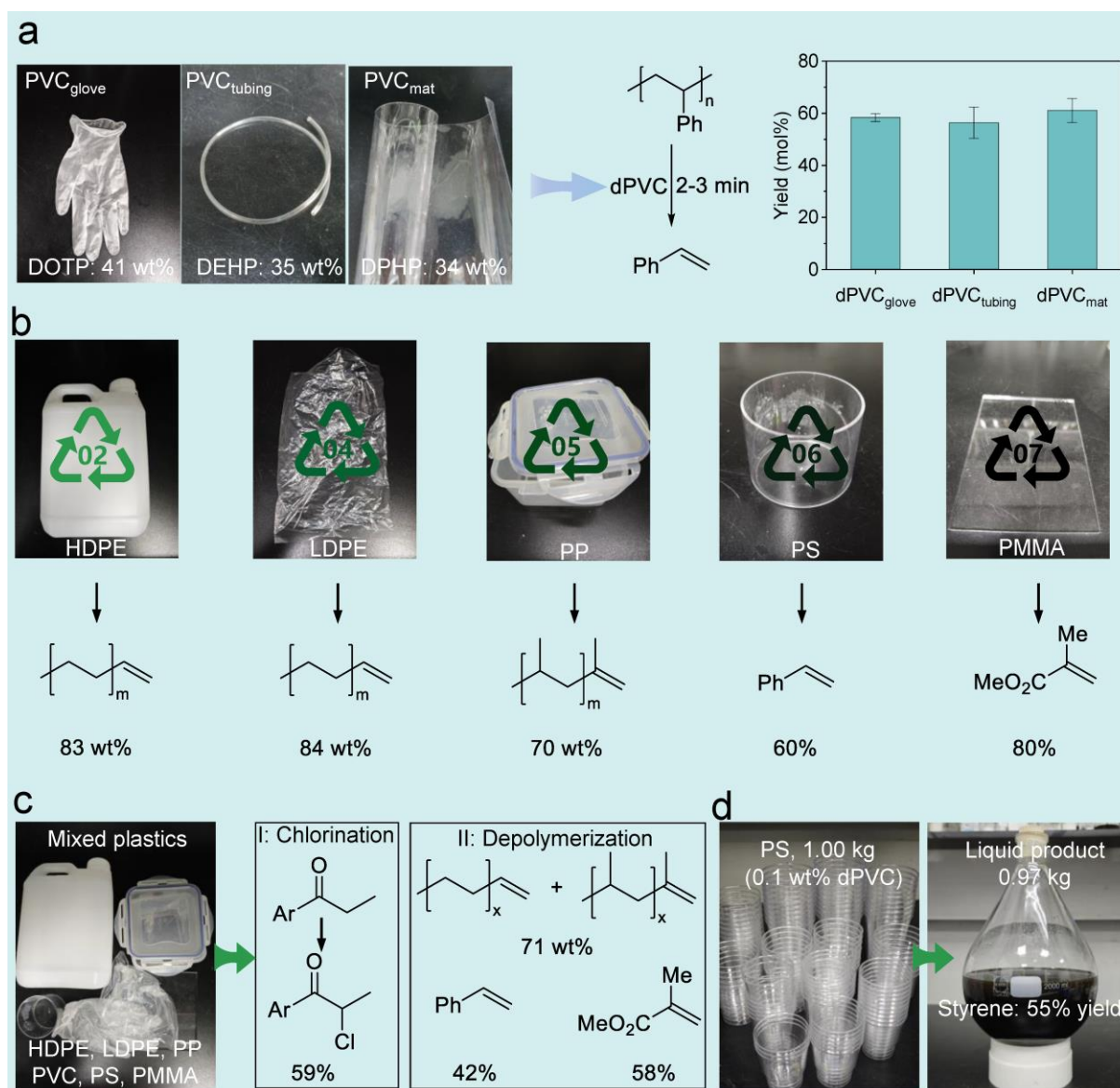

Fig. 3 Recycling of real-life plastic waste under sunlight. (a) Synthesis of photothermal catalyst from PVC waste. (b) Real-life polyolefin plastic waste depolymerization，all depolymerizations were completed within 3 minutes. (c) Recycling of mixed real-life plastic waste. (d) Kilogram-scale degradation of PS cups. DOTP = di(2-ethylhexyl) terephthalate. DEHP = di(2-ethylhexyl) phthalate. DPHP = di(2-propylheptyl) phthalate.

Postconsumer Polymer Recycling. Real-life plastics often contain additives such as dyes, antioxidants, and

plasticizers, whose presence may hinder the depolymerization process, therefore posing significant challenges for the recycling and upcycling of polyolefins (*46*). To further explore the potential practical applications, it was attempted for the recovery of real plastic waste from daily life. The initial attempt involved recycling PVC waste plastics to synthesize photothermal catalysts, 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 catalysts. Despite the presence of significant amounts of plasticizers and varying molecular masses of PVC in these plastic products, the resulting catalysts 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 catalysts from PVC waste plastics has good applicability. Subsequently, dPVC synthesized from discarded PVC gloves was used as a photocatalyst 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% yield. The LDPE plastic bag and PP food container underwent chemical conversion to produce waxes with 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 catalyst. 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, 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 (Figure S39). 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 (Figure S40). The above experimental results fully demonstrate that the protocol

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for converting polyolefins catalyzed by dPVC under sunlight has great practicality in the recycling and upcycling of PVC, HDPE, LDPE, PP, PS, and PMMA waste plastics.

In order to evaluate the solar-thermal conversion ability of dPVC, the light absorption ability of dPVC and recycled dPVC 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 photocatalyst dPVC may exhibit improved photothermal conversion ability after use.
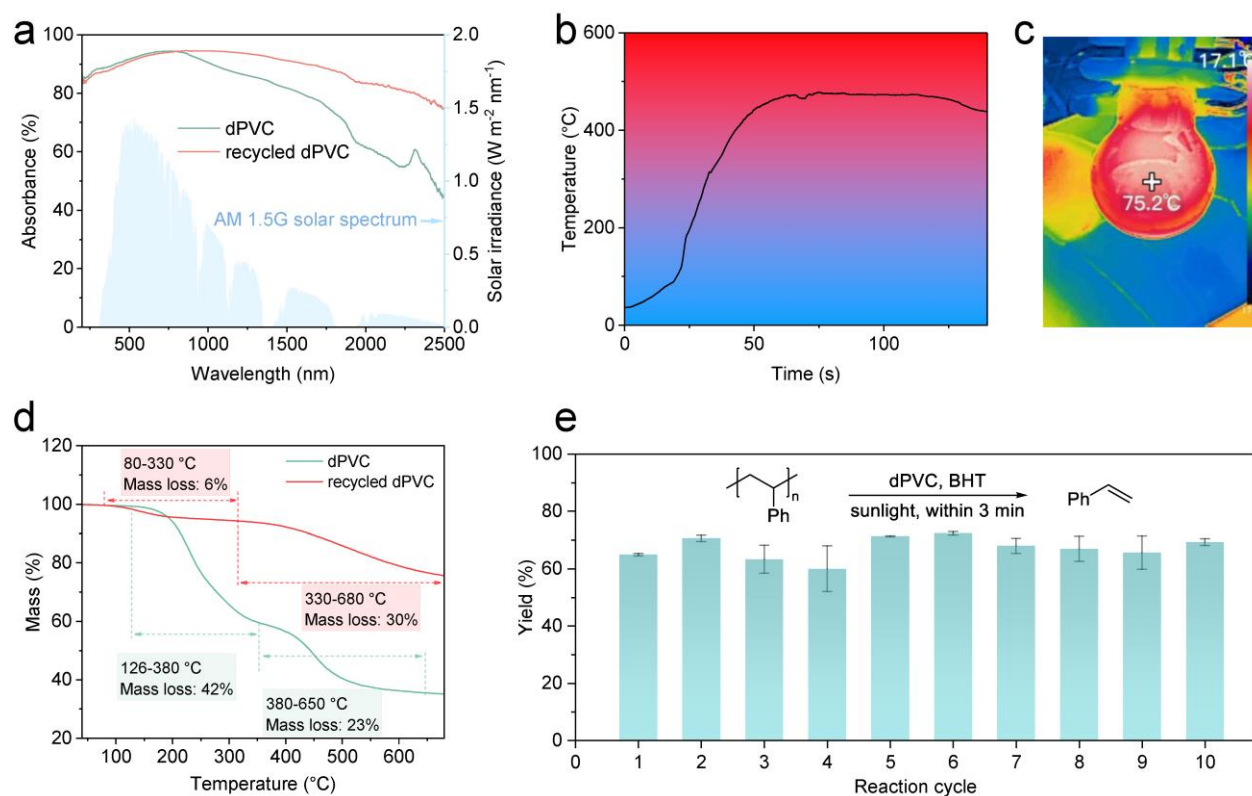

Fig. 4 Photothermal effect and stability of dPVC. (a) UV-Vis-NIR spectra of dPVC. (b) Temperature curve of PS depolymerization. (c) Infrared thermal image of the reactor. (d) Thermal mass loss profile of dPVC. (e) Investigation of catalyst stability.

Under concentrated sunlight exposure, dPVC absorbed sunlight and converted it into thermal energy, rapidly

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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 from initial illumination to reaction completion took approximately 2 minutes. In contrast, the reaction did not proceed in the absence of dPVC (Table S7). 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 catalyst 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. The high temperature of the reaction system poses stringent demands on the stability of the catalyst. 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 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. Both UV-Vis-NIR absorption spectroscopy and TGA indicated that the structure of dPVC had undergone changes after participating in the reaction. An experimental investigation was conducted to explore whether its catalytic activity has also changed. Experiments showed that the catalyst's ability to catalyze the depolymerization of PS remained unchanged over 10 cycles of reuse (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, this did not affect its catalytic capability, and thus it could be reused in the recycling of polyolefins.

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 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). To further identify the chemical structures of the catalyst, ^{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.
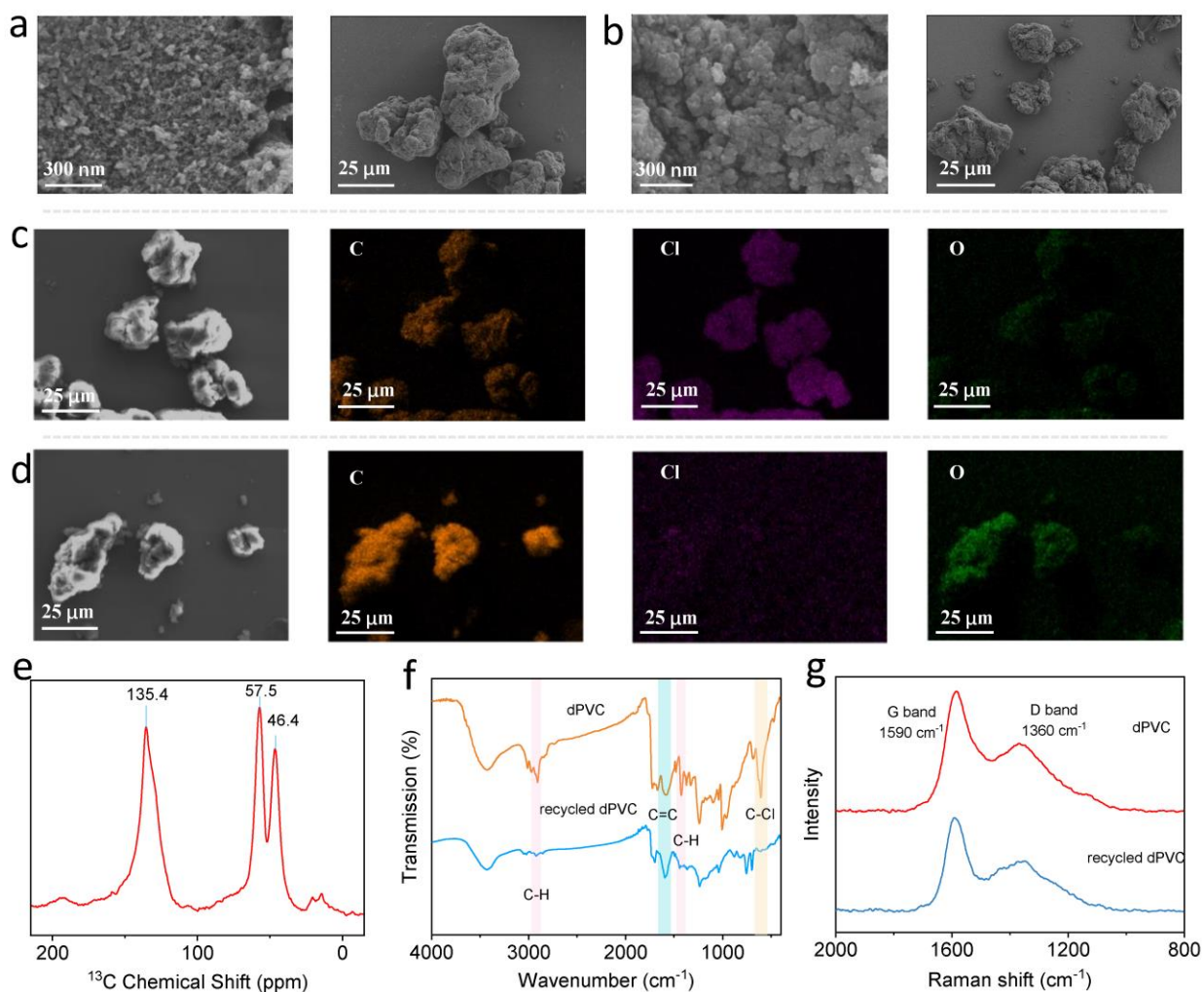
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Fig. 5 Characterization of dPVC. (a) SEM images of dPVC. (b) SEM images of recycled dPVC. (c) SEM image and corresponding energy dispersive X-ray spectroscopy elemental mapping images of dPVC. (d) SEM image and corresponding energy dispersive X-ray 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.

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 catalytic 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

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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. 5f). 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. Fig. 5f presented that there were two bands within both samples, showing the occurred graphitization process from disordered structure to graphitic structure (*47, 48*).

In conclusion, we have established an innovative protocol for the synthesis of photothermal catalysts 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 catalytic system can be extended to real-life plastic recycling scenarios. With the aid of just 0.1 wt% catalyst, the successful depolymerization of kilogram-scale polystyrene 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 catalytic 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.

REFERENCES AND NOTES

1. R. Geyer, J. R. Jambeck, K. L. Law, Production, use, and fate of all plastics ever made. *Sci. Adv.* **3**, e1700782 (2017).

2. B. Zhao *et al.*, Catalytic conversion of mixed polyolefins under mild atmospheric pressure. *The Innovation* **5**, 100586 (2024).

3. OECD, *Global Plastics Outlook: Economic Drivers, Environmental Impacts and Policy Options*. (OECD Publishing, Paris, 2022).

4. A. Rahimi, J. M. García, Chemical recycling of waste plastics for new materials production. *Nat. Rev. Chem.* **1**, 0046 (2017).

5. OECD, *Global Plastics Outlook: Policy Scenarios to 2060*. (OECD Publishing, Paris, 2022).

6. A. Stubbins, K. L. Law, S. E. Muñoz, T. S. Bianchi, L. Zhu, Plastics in the Earth system. *Science* **373**, 51-55 (2021).

7. A. Chamas *et al.*, Degradation rates of plastics in the environment. *ACS Sustain. Chem. Eng.* **8**, 3494-3511 (2020).

8. R. Cao, D. Xiao, M. Wang, Y. Gao, D. Ma, Solar-driven photocatalysis for recycling and upcycling plastics. *Appl. Catal. B Environ.* **341**, 123357 (2024).

9. G. W. Coates, Y. D. Y. L. Getzler, Chemical recycling to monomer for an ideal, circular polymer economy. *Nat. Rev. Mater.* **5**, 501-516 (2020).

10. J. M. Garcia, M. L. Robertson, The future of plastics recycling. *Science* **358**, 870-872 (2017).

11. L. D. Ellis *et al.*, Chemical and biological catalysis for plastics recycling and upcycling. *Nat. Catal.* **4**, 539-556 (2021).

12. I. Vollmer *et al.*, Beyond mechanical recycling: giving new life to plastic waste. *Angew. Chem. Int. Ed.* **59**, 15402-15423 (2020).

13. F. Zhang *et al.*, From trash to treasure: Chemical recycling and upcycling of commodity plastic waste to fuels, high-valued chemicals and advanced materials. *J. Energy Chem.* **69**, 369-388 (2022).

14. L. H. Kugelmass, C. Tagnon, E. E. Stache, Photothermal mediated chemical recycling to monomers via carbon quantum dots. *J. Am. Chem. Soc.* **145**, 16090-16097 (2023).

15. H. Li *et al.*, Hydroformylation of pyrolysis oils to aldehydes and alcohols from polyolefin waste. *Science* **381**, 660-666 (2023).

16. Z. Xu *et al.*, Chemical upcycling of polyethylene, polypropylene, and mixtures to high-value surfactants. *Science* **381**, 666-671 (2023).

17. U. Kanbur *et al.*, Catalytic carbon-carbon bond cleavage and carbon-element bond formation give new life for polyolefins as biodegradable surfactants. *Chem* **7**, 1347-1362 (2021).

18. W. Zhang *et al.*, Low-temperature upcycling of polyolefins into liquid alkanes via tandem cracking-alkylation. *Science* **379**, 807-811 (2023).

19. S. Chen *et al.*, Ultrasmall amorphous zirconia nanoparticles catalyse polyolefin hydrogenolysis. *Nat. Catal.* **6**, 161-173 (2023).

20. S. Liu, P. A. Kots, B. C. Vance, A. Danielson, D. G. Vlachos, Plastic waste to fuels by hydrocracking at mild conditions. *Sci. Adv.* **7**, eabf8283 (2021).

21. A. Tennakoon *et al.*, Catalytic upcycling of high-density polyethylene via a processive mechanism. *Nat. Catal.* **3**, 893-901 (2020).

22. F. Zhang *et al.*, Polyethylene upcycling to long-chain alkylaromatics by tandem hydrogenolysis/aromatization.

Science **370**, 437-441 (2020).

23. R. J. Conk *et al.*, Catalytic deconstruction of waste polyethylene with ethylene to form propylene. *Science* **377**, 1561-1566 (2022).

24. R. J. Conk *et al.*, Polyolefin waste to light olefins with ethylene and base-metal heterogeneous catalysts. *Science* **385**, 1322-1327 (2024).

25. N. M. Wang *et al.*, Chemical recycling of polyethylene by tandem catalytic conversion to propylene. *J. Am. Chem. Soc.* **144**, 18526-18531 (2022).

26. Z. Huang *et al.*, Chemical recycling of polystyrene to valuable chemicals via selective acid-catalyzed aerobic oxidation under visible light. *J. Am. Chem. Soc.* **144**, 6532-6542 (2022).

27. C. Li *et al.*, Upcycling of non-biodegradable plastics by base metal photocatalysis. *Chem* **9**, 2683-2700 (2023).

28. S. Oh, E. E. Stache, Chemical upcycling of commercial polystyrene via catalyst-controlled photooxidation. *J. Am. Chem. Soc.* **144**, 5745-5749 (2022).

29. K. P. Sullivan *et al.*, Mixed plastics waste valorization through tandem chemical oxidation and biological funneling. *Science* **378**, 207-211 (2022).

30. Q. Zhang *et al.*, Oxidative upcycling of polyethylene to long chain diacid over Co-MCM-41 catalyst. *Angew. Chem. Int. Ed.* **63**, e202407510.

31. B. Zhao *et al.*, Selective upcycling of polyolefins into high-value nitrogenated chemicals. *J. Am. Chem. Soc.* **146**, 28605-28611 (2024).

32. L. Gan *et al.*, Beyond conventional degradation: catalytic solutions for polyolefin upcycling. *CCS Chem.* **6**, 313-333 (2024).

33. X. Luo *et al.*, Circularly recyclable polymers featuring topochemically weakened carbon–carbon bonds. *J. Am. Chem. Soc.* **144**, 16588-16597 (2022).

34. L. Wimberger, G. Ng, C. Boyer, Light-driven polymer recycling to monomers and small molecules. *Nat. Commun.* **15**, 2510 (2024).

35. J. Lv *et al.*, Solar utilization beyond photosynthesis. *Nat. Rev. Chem.* **7**, 91-105 (2023).

36. Y. Miao *et al.*, Photothermal recycling of waste polyolefin plastics into liquid fuels with high selectivity under solvent-free conditions. *Nat. Commun.* **14**, 4242 (2023).

37. F. Eisenreich, Photocatalysis as an effective tool for upcycling polymers into value-added molecules. *Angew. Chem. Int. Ed.* **62**, e202301303 (2023).

38. W. Shockley, H. J. Queisser, Detailed balance limit of efficiency of p‐n junction solar cells. *J. Appl. Phys.* **32**, 510-519 (1961).

39. S. Jaisankar, J. Ananth, S. Thulasi, S. T. Jayasuthakar, K. N. Sheeba, A comprehensive review on solar water heaters. *Renew. Sustain. Energy Rev.* **15**, 3045-3050 (2011).

40. P. Tao *et al.*, Solar-driven interfacial evaporation. *Nat. Energy* **3**, 1031-1041 (2018).

41. S. Xu *et al.*, Upcycling chlorinated waste plastics. *Nat. Rev. Methods Primers* **3**, 44 (2023).

42. D. E. Fagnani, D. Kim, S. I. Camarero, J. F. Alfaro, A. J. McNeil, Using waste poly(vinyl chloride) to synthesize chloroarenes by plasticizer-mediated electro(de)chlorination. *Nat. Chem.* **15**, 222-229 (2023).

43. M. Liu, X. Wu, P. J. Dyson, Tandem catalysis enables chlorine-containing waste as chlorination reagents. *Nat. Chem.* **16**, 700-708 (2024).

44. H. Liu *et al.*, Repurposing of halogenated organic pollutants via alkyl bromide-catalysed transfer chlorination. *Nat. Chem.* **16**, 1505-1514 (2024).

45. M. Arifuzzaman *et al.*, Selective deconstruction of mixed plastics by a tailored organocatalyst. *Mater. Horiz.* **10**, 3360-3368 (2023).

46. J. Zheng, M. Arifuzzaman, X. Tang, X. C. Chen, T. Saito, Recent development of end-of-life strategies for plastic in industry and academia: bridging their gap for future deployment. *Mater. Horiz.* **10**, 1608-1624 (2023).

47. Y. Qian, P. Wei, P. Jiang, X. Zhao, H. Yu, Synthesis of a novel hybrid synergistic flame retardant and its application in PP/IFR. *Polym. Degrad. Stab.* **96**, 1134-1140 (2011).

48. F. Tuinstra, J. L. Koenig, Raman spectrum of graphite. *J. Chem. Phys.* **53**, 1126-1130 (1970).

DATE AVAILABILITY

Data relating to the characterization data of materials and products, general methods, experimental procedures are available in the Supplementary Information.

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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.

ADDITIONAL INFORMATION

Supplementary information includes Supplemental Experimental Procedures can be found with this article online.