

Highly selective upcycling of plastic mixture waste by microwave-assisted catalysis over Zn/b-ZnO

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7 billion of 9.2 billion tons of plastic produced becomes waste while conventional catalytic plastic recycling methods are vulnerable with degraded performance and intensive energy input. Here, a hybrid Zn/b-ZnO catalyst, together with the specially-designed microwave reaction system, has achieved fast plastic waste upgrading under atmospheric pressure without using H₂. Bifunctional ZnO acts as a microwave absorber and substrate catalyst, and in-situ formed Zn clusters promote C-C bond cleavage and nearly 100% upcycle landfilled plastic mixtures into lubricant base oil precursors and monomers. Unprecedented turnover number (250 g_{plastic} g⁻¹_{catalyst}) of plastic depolymerisation and long-time stability over 50 successive cycles have been demonstrated, together with 8-time higher energy efficiency compared with conventional catalysis, indicating this strategy is an economical approach to efficient upcycling of plastics towards valuable products. Moreover, the catalyst can tolerate high contaminants, even the landfilled plastics can still be converted to lubricant base oil precursors, which has never been reported before.

Plastic, the most representative artificial material of post-industrialisation, has permanently changed the lives of humanity with its high modifiability and accessibility^{1–4}. In 2019, over 360 million tons of plastic were produced globally, and after use, most became waste, ending up in landfills or being dumped⁵. Plastic pollution can alter habitats and natural processes, reduce ecosystems' ability to adapt to climate change, and directly affect millions of people's livelihoods in terms of food production capabilities and social well-being. Common treatments such as incinerating and landfilling can result in more and long-term pollution, which is not sustainable. Mechanical recycling is an essential plastic recycling method but often requires a combination of multiple separation methods to separate a sufficiently single and

pure polymer from the post-consumer plastic waste stream, which is challenging to recycle complex mixed plastics and multi-layer packaging materials, but chemical recycling is much superior^{3,6}. Thus, an economically feasible technology to use the plastic waste is much sought-after, and chemical recycling^{3,7} of plastics to yield value-added chemicals such as H₂⁸, alkenes^{9,10} (ethylene, propylene, etc.), fuel^{11–13} and lubricant base oil¹⁴ has been recognised as a promising solution. Polyolefins, including high-density polyethylene (HDPE), low-density polyethylene (LDPE), and polypropylene (PP), account for over 50% of all plastics produced¹⁵. However, their recycling poses a significant challenge due to the thermodynamically inert C-C bonds that make them difficult to activate or break. Consequently, a considerable

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amount of post-consumer polyolefin waste ends up being dumped in the environment¹⁶. Furthermore, these plastic wastes are often physically mixed and contaminated with various foreign materials, further complicating recycling efforts. Additionally, the plastic could be composited by several polymer and additive components, such as multilayer packaging, making recycling even more challenging¹⁷. Unfortunately, there is a scarcity of applicable catalysts that can meet the stringent requirements for recycling these complex and multilayer mixtures. Therefore, it is of utmost importance to develop an efficient method for the smart recycling of polyolefins, whether as single plastics or mixed plastics, in order to build an environmentally friendly society.

The existing methods have not adequately addressed these challenges. For example, conventional plastic recycling techniques, such as pyrolysis and gasification, often require high operating temperatures (700–1300 K)¹⁸, which demand significant energy and lead to intricate side reactions. Although recently emerged methods, such as catalytic hydrogenolysis and tandem catalytic alkane metathesis, exhibit better product selectivity to some extent, they have to work under high-pressure conditions (8–50 bar H₂, O₂, or ethylene), use precious metals catalysts (Pt, Ru, Ir, etc.), and relatively prolonged reaction durations (6–48 h), resulting in limited practical application^{10,12,19}. These catalysts also are very prone to contaminants which are always present in the used plastics. Very recently, two benchmark works employed a pyrolysis method to convert plastic into olefin-rich oils or waxes^{20,21}. Subsequently, these products were further processed into aldehydes or fatty acids through hydroformylation or oxidation. Based upon these successes, there is a pressing need of further advances for the plastic recycling, involving the efficient catalytic process operated under milder conditions and generating short-chain valuable molecules²². Therefore, it is necessary to develop a smart plastic recycling pathway with high selectivity to value-added products like lubricant-based oil precursors. Importantly, the selective heating by microwaves can facilitate process electrification, intensify chemical recycling of plastic and reduce energy consumption.

Herein, an efficient Zn/b-ZnO catalyst has been developed through in-situ construction using microwave irradiation to upcycle plastic waste and selectively depolymerise polyolefin waste into lubricant base oil precursors. In this process, powdered polyolefin is mechanically blended with bifunctional ZnO (b-ZnO) prior to expose to microwave radiation. The reaction temperature and time are set at 280 °C and 30 min, respectively. Following the microwave radiation, a high yield of hydrocarbon oil, mainly lubricant base oil precursors is produced. Notably, this in-situ constructed catalyst exhibits remarkable stability over 50 cycles, with an oil yield range of 70 wt% to 84 wt%, thereby maximising the production of valuable lubricant base oil precursors. The gaseous byproducts, predominantly C₂–C₄ monomers which exhibit a selectivity of 60–80%, further enhance the commercial value. Moreover, an exceptionally high turnover number (250 g_{plastic} g⁻¹ catalyst⁻¹) for plastic depolymerisation is achieved with this Zn/b-ZnO catalyst. Importantly, when applied to a mixture of plastics waste dug out from landfill sites, the Zn/b-ZnO catalyst still efficiently generates lubricant base oil precursors. Through comparative experiments and detailed characterisation of bifunctional catalysts at different usage stages, a plastic upcycling mechanism has been proposed. Overall, this work holds significance in advancing plastic upcycling.

Results

Microwave catalytic upcycling of plastic

The b-ZnO was prepared via a controlled precipitation method. Zn(CH₃COO)₂·2H₂O was dissolved in deionised water, and then aqueous ammonia was added dropwise into the solution. The resulting mixture was precipitated in an 85 °C water bath to obtain the b-ZnO precipitate. Such lab-prepared ZnO catalyst is found to own complicated microstructures, more defects, excellent catalytic activity and

excellent microwave absorption ability compared with commercial ZnO. Therefore the synthesised ZnO is defined as bifunctional ZnO (b-ZnO). (Supplementary Figs. 1–3 and Supplementary Table 1). The plastic depolymerisation process was conducted in a specially-designed microwave reaction system that utilised a 2.45 GHz magnetron multimode microwave reactor (Supplementary Fig. 4). Firstly, different microwave power such as 240 W, 320 W and 400 W were evaluated (Supplementary Table 2 and Supplementary Fig. 5), along with electric field strength and distribution simulations and discussions (Supplementary Figs. 6–8). Results show that the oil yield increases from 29.9 wt% to 31.4 wt% as the microwave power increases from 240 W to 320 W, while further increasing the microwave power to 400 W, the oil yield decreases to 26.0 wt%. Moreover, the C₂–C₄ monomers present the highest accounting in the gaseous products when the microwave power setting is at 320 W. Therefore, the optimal microwave irradiation power is determined to be 320 W. Next, the mass ratio of plastic to the catalyst is optimised to be 4:1 or 5:1. When the mass ratio is smaller than 4, the heating capacity of the mixture under microwave irradiation is enhanced. In this case, (i) the mixture can powerfully transform the incoming microwave electromagnetic energy into heat, providing sufficient energy to break the C–C bonds and may induce excessive depolymerisation; (ii) the plastic and products have more chance to contact with the ‘catalyst’ or ‘active sites’, causing secondary cracking or side reaction, which can be proved by Supplementary Fig. 9. In contrast, when the mass ratio of plastic to catalyst increases to a proper range, although the heating efficiency of the mixture is slightly reduced, the heat generated selectively on the catalyst can be better allocated to the plastic, forming a solid-liquid contact state and reducing the contact opportunity between the ‘catalyst’ or ‘active sites’ with the plastic intermediates, facilitating to reduce excessive depolymerisation and obtain oil products²³ (Supplementary Fig. 10, Supplementary Table 3 and Supplementary Movie 1). However, when the mass ratio of plastic to catalyst is set as 6:1, the reaction is prolonged from 30 min to 45 min (Supplementary Fig. 11). Furthermore, the in-situ construction (self-activation) of the catalyst was also investigated. Results indicate that under the optimal mass ratio of plastic to the catalyst, the catalyst is activated within the first five cycles, which significantly increases the oil yield to more than 70% and maintains stability in subsequent cycles (Supplementary Figs. 12–14, Supplementary Tables 4–6). Importantly, the catalyst reconstruction rates can be accelerated by adjusting the mass ratio of plastic to the catalyst (Supplementary Table 3).

The above-selected conditions were then employed in the subsequent investigation of microwave catalytic plastic depolymerisation. Figure 1 shows the results of the microwave catalytic depolymerisation of mechanically pulverised agricultural film plastic (dug out from landfill site, ~6.4 wt% impurities, Supplementary Fig. 15) using activated b-ZnO as the catalyst at a reaction temperature of 280 °C and a reaction time of 30 min. A typical cycle includes mixing LDPE feedstock pulverised from agricultural film with activated b-ZnO catalyst particles and then irradiating the mixtures by microwave in a special reactor (Fig. 1a and Supplementary Movie 2). For upcycling agricultural film plastic waste (polyethylene), the gas yield (Yield: 27–37 wt%) and oil yield (Yield: 61–70 wt%) remain relatively stable over ten cycles (Fig. 1b and Supplementary Table 7), despite the presence of high impurities in this plastic waste. Gas chromatography was used to analyse the byproducts, particularly the volatile products. It was found that between 56–70% of the gaseous species produced from LDPE depolymerisation are ethylene, propylene, and butylene (Fig. 1c and Supplementary Fig. 16), which can be utilised in the resynthesis of plastics.

The excellent catalytic activity is further highlighted when compared with the representative plastic upcycling catalysts (Fig. 1d and Supplementary Table 8). The oil yield per g catalyst per hour (g g⁻¹ cat h⁻¹) over Zn/b-ZnO surpasses the majority of benchmark catalysts by a significant margin, about several times higher activity even

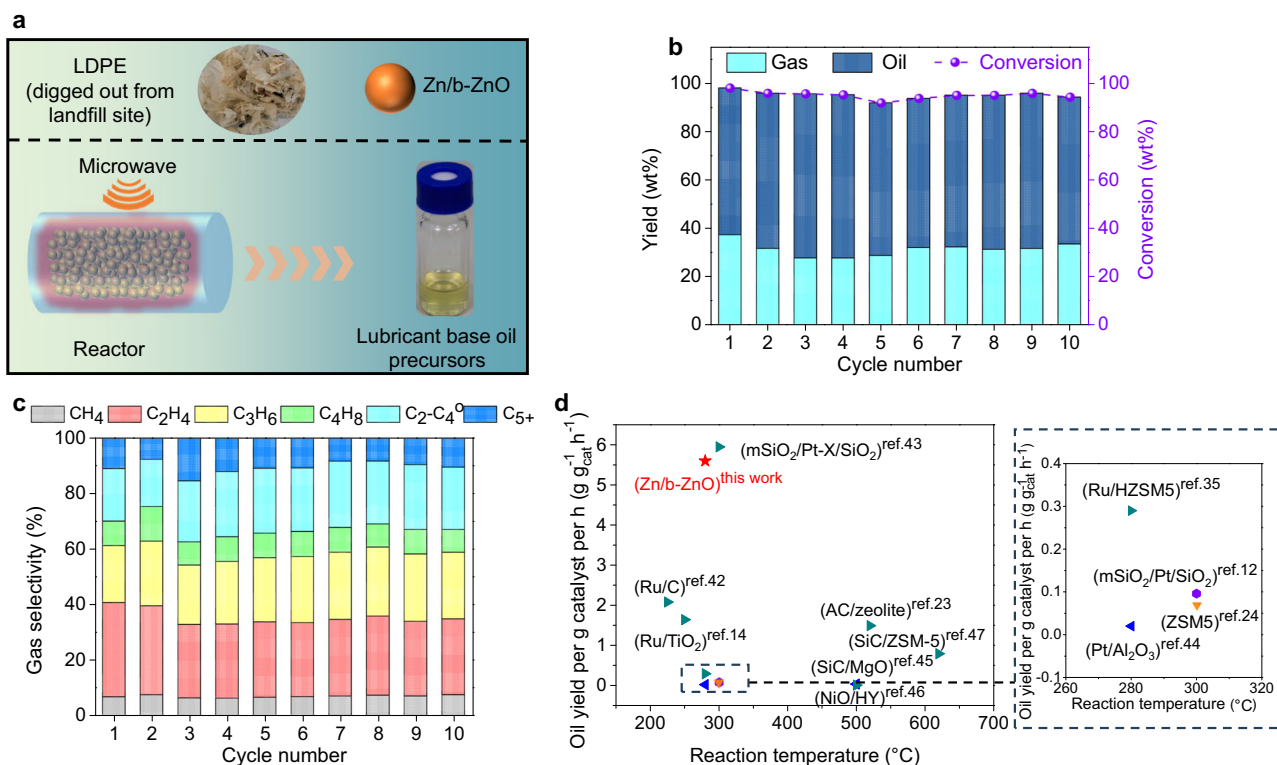


Fig. 1 | Successive cycles of microwave catalytic depolymerisation of LDPE (digged out from landfill site). **a** Microwave catalytic depolymerisation process. **b** Products yield and conversion. **c** Gas product selectivity. The mass ratio of the catalyst to the plastic was: Zn/b-ZnO: Plastic = 10 g: 40 g. C₂H₄, C₃H₆, C₄H₈ represent ethylene, propylene, butene, and C₂-C₄ means ethane, propane, and butane. **d** Summary of oil yield per g catalyst per hour (g g⁻¹ cat h⁻¹) in different reaction temperatures achieved in this work and other representative studies (mSiO₂/Pt/

SiO₂ (autoclave reactor, ref. 12), Ru/TiO₂ (autoclave reactor, ref. 14), Ru/HZSM5 (autoclave reactor, ref. 35), Ru/C (autoclave reactor, ref. 42), mSiO₂/Pt-X/SiO₂ (autoclave reactor, ref. 43), Pt/Al₂O₃ (autoclave reactor, ref. 44), ZSM5 (microwave reactor, ref. 24), SiC/MgO (microwave reactor, ref. 45), NiO/HY (microwave reactor, ref. 46), AC-zeolite (microwave reactor, ref. 23), SiC/ZSM-5 (microwave reactor, ref. 47)).

though many of those catalysts consist of noble metals known for their high catalytic activity. Therefore, this highly active Zn/b-ZnO catalyst holds the potential to substantially enhance the oil production while concurrently reducing energy consumption.

To assess the catalyst's robustness and stability against different plastic streams, mixed plastics waste (digged out from landfill site, ~10 wt% impurities, Supplementary Fig. 17) collected from a waste disposal site in Britain and single-used lunch boxes (PP) were selected as representative materials (Fig. 2a). For upcycling mixed plastics waste, the gas yield (Yield: 32–44 wt%) and oil yield (Yield: 47–62 wt%) exhibit slight variations over five cycles, reflecting the complexity of the mixed plastics waste (Fig. 2b and Supplementary Table 9). In addition, 50 cycles of PP depolymerisation were evaluated under microwave radiation to assess the long-term stability of the catalyst. As discussed above, the first five consecutive cycles of the catalytic reactions are the self-activation of the b-ZnO, the gas yield decreases from 66 wt% to 25 wt%, while the oil yield increases from 30 wt% to 70 wt%, after which the yields stabilise in the following 45 cycles, the oil yield on average reaches 73 wt%, as shown in Fig. 2c and Supplementary Table 10. The selectivity of olefins (ethylene, propylene, and butylene) from mixed plastics waste and PP is around 65–70% and 60–80%, respectively, and the overall selectivity to C₂-C₄ monomers is also comparable to some previous work²⁴ (Supplementary Figs. 18–19a). Olefins dominate over alkanes in the gas products (Supplementary Figs. 18–19b), especially for depolymerisation of PP, the ratio of propylene is 21 times higher than propane, accounting for around 50% of the gaseous species, indicating a relatively high purity of olefins. This high purity is advantageous for further refining these compounds into industrial monomer materials²⁵. Additionally, the

production rate of CH₄ and H₂ are relatively low (Supplementary Figs. 18–19c). This is beneficial as excessive generation of methane and hydrogen leads to the aggregation of carbon deposits, which can block active sites on the catalyst^{7,23,24}. By reducing methane and hydrogen production, the selectivity to high value-added products and the lifetime of the catalyst can be dramatically improved^{26,27} (Supplementary Fig. 20).

Compared with the gaseous product, it is more appealing that oil is produced from LDPE (digged out from landfill site), PP and mixed plastics waste (digged out from landfill site) depolymerisation, which was analysed by simulated distillation gas chromatography, GC-MS, ¹H-NMR and ¹³C-NMR. Simulated distillation curves indicate the LDPE oil products mainly consist of hydrocarbons with boiling points around 280–650 °C (molecular weight around 200–1000 Da), which can be used as lubricant base oil precursors¹⁹ (Fig. 3a and Supplementary Table 11). GC-MS reveals that oil products have carbon numbers ranging from C₈-C₄₀ and a high account of up to 65% for alkanes, with C₁₆-C₄₀ representing >74% of the hydrocarbons (Fig. 3d-f and Supplementary Figs. 21–25), in consistence with simulated distillation results (Fig. 3a). Furthermore, the carbon number distribution of the oil products is relatively narrowed with the increase of catalyst cycles, making the oil products more suitable to be used as the lubricant base oil precursors. In addition, the ¹H and ¹³C NMR spectra indicate that the oil products derived from LDPE plastic waste depolymerisation consist of linear-chain compounds. As the boiling point/molecular weight of the components increases, the composition shifts to more linear chain and fewer complex isomers in the oil products, especially for the hydrocarbons with carbon number >C₁₆. This implies that this portion of the product is obtained directly through C-C bond cleavage without

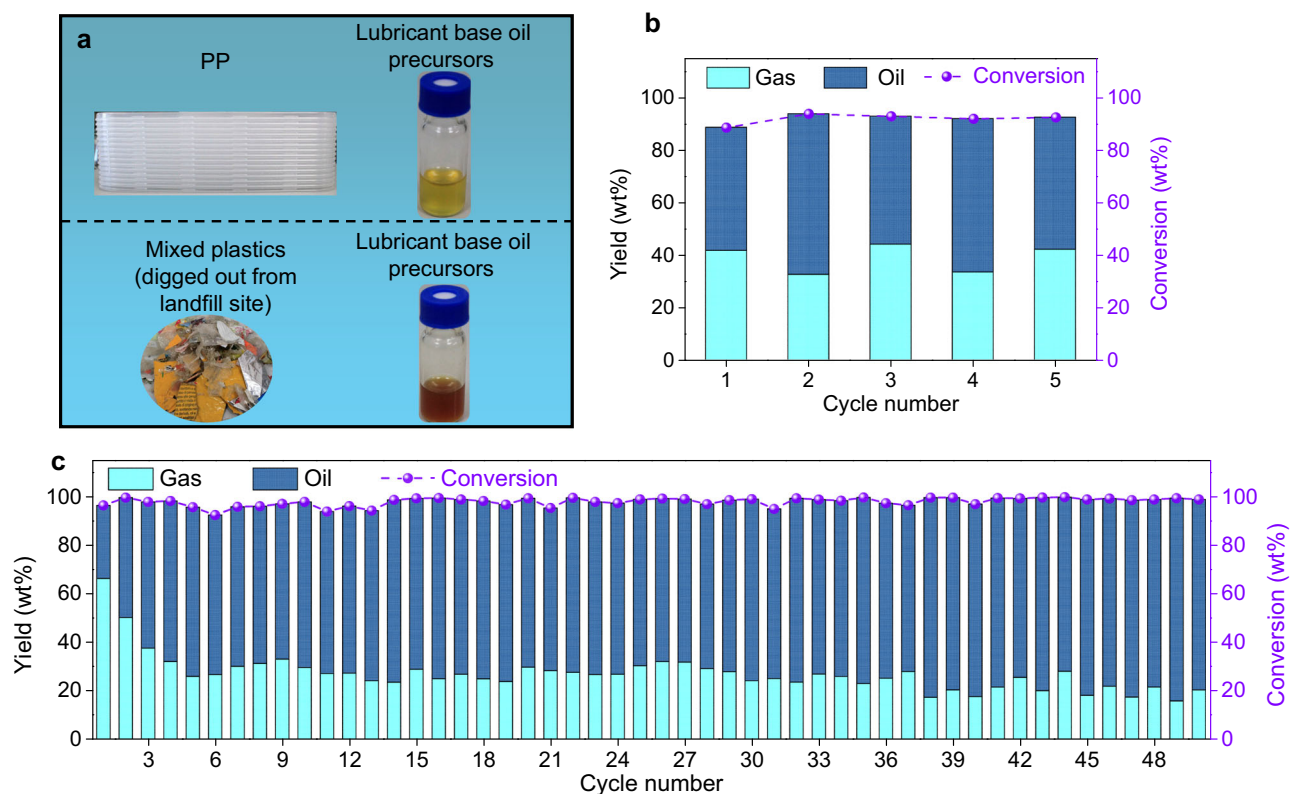


Fig. 2 | Microwave catalytic depolymerisation of PP and mixed plastics (dug out from landfill site). **a** Images of plastic waste and corresponding oil products. **b, c** Products yield and conversion of mixed plastics and PP, respectively. The mass

ratio of the catalyst to the plastic was: Zn/b-ZnO: mixed plastics = 10 g: 40 g, b-ZnO: PP = 10 g: 50 g.

undergoing further isomerisation (Supplementary Fig. 26). However, for lubricant production, the highly branched structure is more conducive to improving the fluidity of the lubricant and reducing the pour point. Polypropylene plastics have a rich branched skeleton structure. Therefore, the catalytic depolymerisation of polypropylene or mixed plastics waste is expected to obtain better lubricant base oil precursors.

Similar boiling points and carbon number distribution of oil products are observed in the upcycling of mixed plastics and PP waste. The results from simulated distillation gas chromatography reveal that the predominant components of the oil products are composed of lubricant base oil precursors fractions (Fig. 3b, c). This finding is further corroborated by the GC-MS results. Furthermore, the oil products obtained from the first cycle exhibit a broader distribution, while the other cycles produce a relatively uniform distribution of oil (Fig. 3g–n) and Supplementary Figs. 27–38). These results suggest that the catalyst's capacity for selectively cleaving C–C bonds progressively improves over the course of the reaction. Furthermore, the catalyst remains effective even after 50 cycles of PP depolymerisation, enabling the continuous depolymerisation of LDPE (specifically, plastic straws), resulting in the production of C_{20} -centred distributed oils (Supplementary Figs. 39–40) with a yield of 76.9 wt%. This suggests that the designed catalyst possesses a lifespan exceeding 50 cycles and can efficiently cleave the C–C bond to obtain the narrowly distributed oil products. Conventionally, such narrow distribution oil products are commonly achieved through high-pressure (10–50 bar H_2) hydrogenolysis or long-term reaction durations^{12–14,28} (Supplementary Table 12).

Moreover, lubricant base oil precursor properties are important in the evaluation of the potential application of the oil products obtained in this work in the lubricant industry. Therefore, the pour point, kinematic viscosity, and viscosity index (VI) of oil products were analysed and listed in Supplementary Table 13. Results indicate that

the VI of oil products obtained from microwave catalytic upcycling of LDPE, mixed plastics and PP are 141.3, 123.4 and 122.2, while the pour point is 39.7 °C, 40.9 °C and 41.2 °C, respectively. Typically, the VI below 120 corresponds to Group I–III mineral oil and higher than 120 belongs to synthetic lubricants^{29,30}. Compared to commercial lubricants, liquid oil products show lower kinematic viscosity and higher pour points, as microwave catalytic depolymerisation proceeds under hydrogen-free and atmosphere pressure without undergoing hydrogenation, isomerisation, and cracking, so it is defined as lubricant base oil precursors. Importantly, the lubricant base oil precursors play a vital role in the production of lubricants (lubricants are composed of 80% to 90% base oils and 10% to 20% additives), which can be post-processed to produce lubricants through industrially available processes such as hydrogenation, branching and refining³¹.

Overall, based on the above results, the catalyst has demonstrated remarkable efficiency in depolymerising complex plastics, showcasing notable robustness and stability. This holds promising potential for the efficient upcycling of plastic mixtures, eliminating the need for any pre-treatment, and yielding high-value-added monomers and lubricant base oil precursors since the relatively weak acidity of Zn/b-ZnO and selective heating characteristics under microwave irradiation contribute to obtaining these valuable products (Supplementary Fig. 41). Furthermore the valuable cyclic (C_6 – C_{14}) hydrocarbons can also be selectively improved by mixing a small amount of ZSM-5 with Zn/b-ZnO during microwave catalytic depolymerisation of plastic (Supplementary Figs. 42–43). Therefore, the product distribution from depolymerisation of plastics can be flexibly controlled by selecting a catalyst with suitable acidity.

The excellent performance of the synthesised b-ZnO catalyst in upcycling plastic waste can further be highlighted by comparing it with various catalysts used in microwave catalytic depolymerisation of polyolefin. The synthesised catalyst exhibits a significant increase in

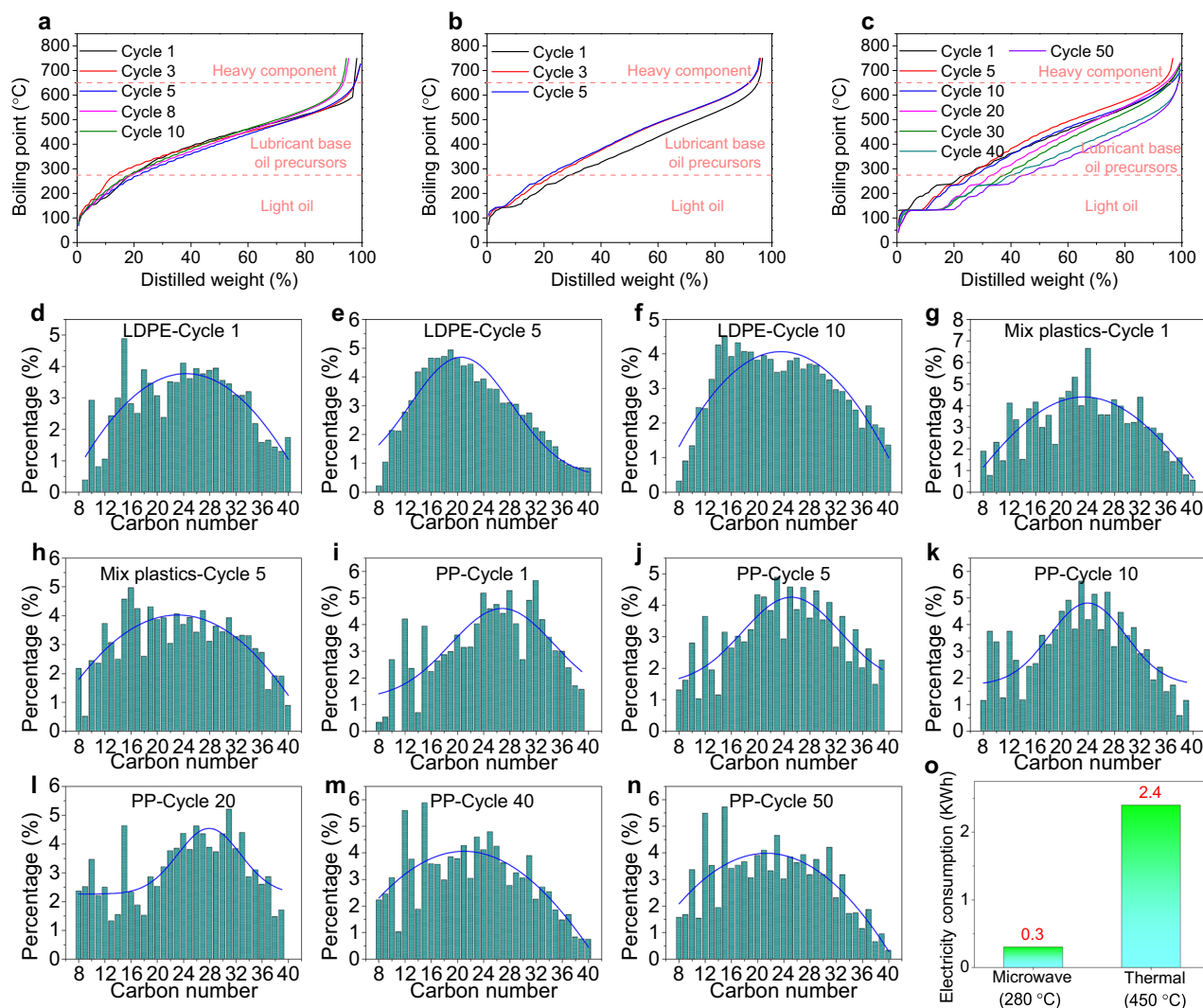


Fig. 3 | Microwave catalytic upcycling results and the process energy efficiency. Simulated distillation gas chromatography results of oil products from the microwave catalytic upcycling of (a) LDPE (dugged out from landfill site), (b) mixed plastics (dugged out from landfill site), and (c) PP at different cycles, respectively.

Carbon number distribution of oil products from the microwave upcycling of LDPE (dugged out from landfill site) (d-f), mixed plastics (dugged out from landfill site) (g-h), and PP (i-n) at different cycles, respectively. o Electricity consumption of microwave (280 °C) and conventional thermal (450 °C) catalytic processes.

the yield of monomers and lubricant base oil precursors, surpassing the performance of other catalysts, including carbon materials and commercial ZnO as summarised in Supplementary Fig. 44 and Supplementary Table 14. This superiority can be attributed to the structural characteristics of the hybrid catalyst, together with the unique microwave heating (Supplementary Figs. 1–3, Supplementary Fig. 10 and Supplementary Table 1). Another well-known microwave-absorbing carrier, e.g. activated carbon was added to b-ZnO to enhance its microwave absorption. However, this process often leads to undesired side reactions³², ultimately resulting in lower added value of the final product (Supplementary Table 15). Thus the synthesised b-ZnO catalyst does not require such additional measures to strengthen microwave absorption and can appropriately harness the microwave energy for efficient depolymerisation, thereby achieving higher product yields and maintaining the value of the end product.

To illustrate the superiority of the Zn/b-ZnO catalyst for microwave-assisted catalytic depolymerisation, a comparison of product distribution and electricity consumption between microwave and traditional thermal catalysis was conducted. The traditional thermal catalysis process under a series of reaction temperatures (400 °C, 450 °C, 500 °C and 600 °C) presents that the oil distribution obtained by

the depolymerisation of PP with thermal catalysis has specific differences compared with microwave-assisted catalytic depolymerisation of PP, which also reflects the uniqueness of microwave-assisted catalytic depolymerisation of plastics (Supplementary Fig. 45). Moreover, the energy efficiency of the overall PP depolymerisation reaction was estimated using the electricity metre. Figure 3o illustrates the electricity consumption of the designed Zn/b-ZnO catalyst in PP upcycling by microwave and traditional heating methods. The electricity consumption of the microwave-assisted catalytic depolymerisation method is only 0.3 kWh when 10 g catalysts were used with 50 g plastic per cycle. In comparison, the traditional thermochemical method reaches 2.4 kWh (450 °C when the oil products distribution is similar to microwave-assisted catalytic to some extent), significantly higher than microwave-assisted catalytic depolymerisation. This improvement can be attributed to the bifunctionality of the designed Zn/b-ZnO catalyst, which acts as an excellent microwave absorber, efficiently converting electromagnetic energy into hot spots (Supplementary Fig. 46), and faster catalytically depolymerising plastic to generate terminal products. In contrast, the plastic catalytic depolymerisation by conventionally thermal catalysis is also much slower than that of the microwave-assisted catalysis due to the overall heating process,

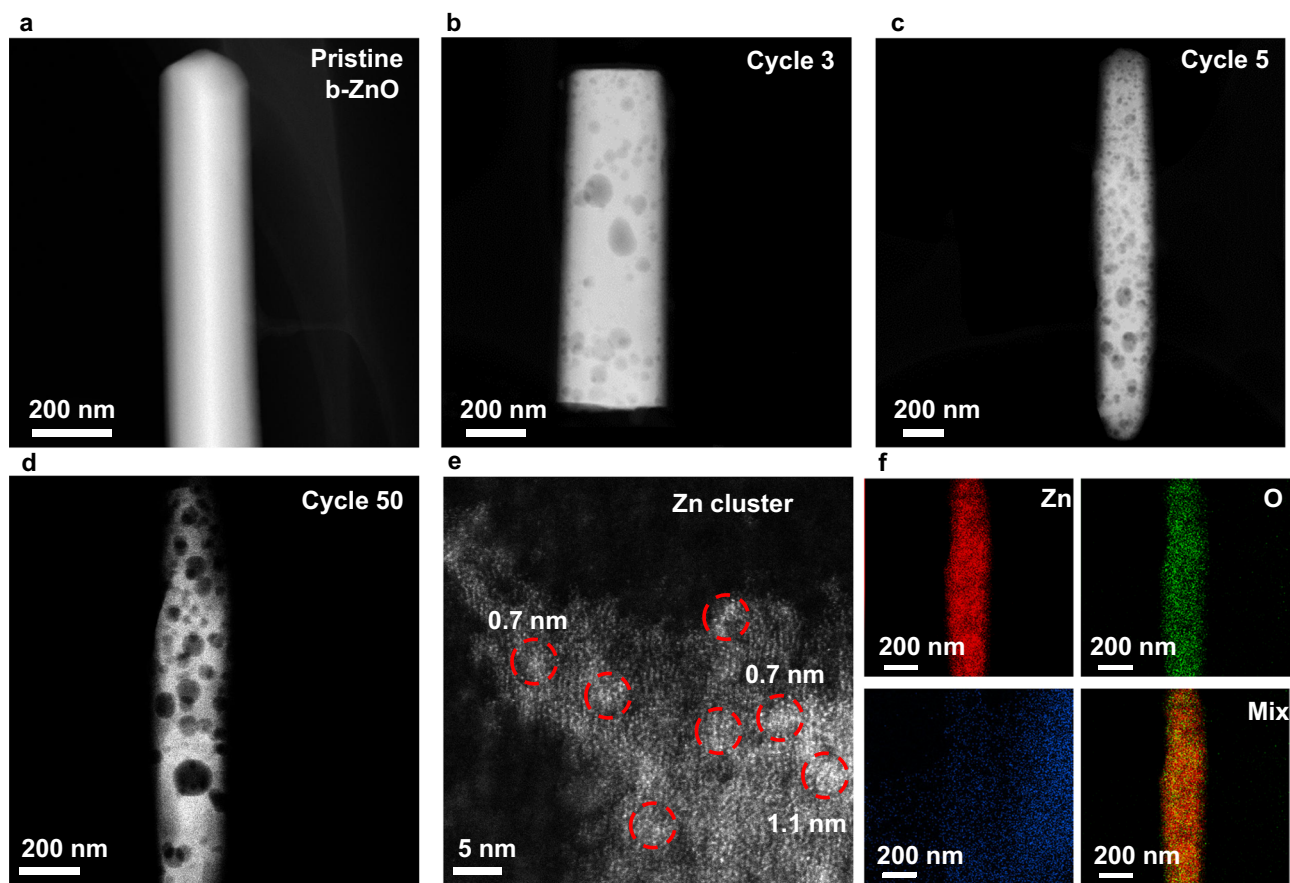


Fig. 4 | Characterisations of the used b-ZnO catalyst after different cycles of plastic depolymerisation. a–c HAADF-STEM images of pristine and used b-ZnO and **(d, e)** HAADF-STEM images of b-ZnO after 50 cycles, the Zn clusters on b-ZnO marked by red circles. **f** Corresponding element mapping (red for Zn, green for O and blue for C).

leading to higher energy consumption and the formation of numerous undesired side reactions, reducing the yield of high value added lubricant base oil precursors and increasing the complexity of the product¹⁸. Notably, the rapid depolymerisation reaction and short residence time of the microwave-assisted process greatly reduce the energy consumption and suppress side reactions of the reaction^{8,24}. Therefore, the microwave-assisted catalytic depolymerisation approach using the designed Zn/b-ZnO catalyst presents remarkable energy efficiency and excellent product distribution in the depolymerisation of plastic.

The characterisation of the active catalyst

To investigate the evolution of product distribution from 1st cycle to 5th cycle, the improvement in catalytic efficiency, and the long-term stability of high catalytic activity throughout the subsequent 45 cycles, the catalysts used at different stages were characterised to comprehensively monitor the changes occurring in the catalyst during the extended cycling process. Firstly, the aberration-corrected high-angle annular dark field scanning transmission electron microscope (HAADF-STEM) and energy dispersive X-ray spectroscopy (EDX) mapping were carried out to investigate the morphology of the catalysts. The pristine b-ZnO exhibits a uniform, smooth rod-like structure, while the used b-ZnO surface shows an increasing number of elliptical-shaped holes, indicating structural modifications with prolonged cycling (Fig. 4a–d) and Supplementary Fig. 47). This is due to the generation of hydrogen during the depolymerisation reaction of plastics, which could initiate the reduction of b-ZnO, forming etching holes.

Furthermore, an in-depth analysis of a 50-cycle used catalyst sample reveals that the recycled b-ZnO sample exposes abundant Zn

clusters (0.7–1.1 nm, Fig. 4e). The EDX mapping further confirmed that the existence of etching holes and Zn clusters, providing additional support for the aforementioned procedure (Fig. 4f and Supplementary Figs. 48–49). The related information can also be derived from the X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), X-ray absorption spectroscopy (XAS), thermogravimetry (TGA) and H₂-TPR as discussed in the Supplementary Information (Supplementary Figs. 50–58, and Supplementary Table 16).

Proposed reaction pathway

Plastic materials, due to their low dielectric loss and essentially microwave transparency, are inefficient in converting electromagnetic wave energy into heat (Supplementary Fig. 10), which hampers their ability to initiate chemical reactions. As a result, it is often necessary to combine them with materials that possess high microwave energy conversion capacity to induce reactions. In this process, b-ZnO serves as a catalyst that rapidly converts microwave energy into local heat (hot spots), ensuring selective heating of the catalyst particles (Supplementary Figs. 46, 59a, b). The generated heat is then transferred to the plastic, causing it to melt quickly and transition into a solid-liquid contact state which facilitates the plastic depolymerisation reaction (Supplementary Movie 1).

During the reaction, a notable shift in product compositions towards more valuable products is observed in the early stages, indicating an intrinsic transforming of the catalyst. The production rate of lubricant base oil precursors gradually increases while gaseous products exhibit a gradual decrease and eventually reach a plateau after five cycles. Similar trends are also observed in the catalytic depolymerisation of other plastic waste (Supplementary Figs. 12–14,

Supplementary Tables 4–6 and Supplementary Figs. 60–69). The STEM, XAS, XRD, TGA, H₂-TPR and XPS results of the used catalyst suggest the b-ZnO catalyst progressively undergoes reduction and transformation, i.e. a self-activation process, forming a highly reactive and selective Zn cluster/b-ZnO hybrid catalyst (Supplementary Figs. 50–58).

The self-activation process can be validated by adjusting the mass ratio of the plastic feedstock to the catalyst, resulting in controlled activation. When the mass ratio of the plastic to the catalyst is set to 1:1 or 2:1 in the first cycle with the same radiation power, the catalyst takes only one cycle to be fully activated. Upon readjusting the mass ratio to 5:1 for this activated catalyst, it directly attained an oil production rate of 70–74 wt% for the subsequent cycles. This acceleration is attributed to the lower ratio of the plastic amount to the catalyst, which promotes the fracturing of the C-H bond and facilitates H₂ generation (Supplementary Table 3). Furthermore, the presence of hot spots during the reaction likely enhances the dissociation of H₂ into atomic hydrogen. The atomic hydrogen, in turn, leads to the breaking of Zn-O bonds and the conversion of b-ZnO into Zn clusters (Supplementary Fig. 59(c–f)). Additionally, this process generates -OH (or H₂O) and creates pits on the catalyst surface, known as hydrogen etching^{33,34} (Fig. 4).

Particularly, the HAADF-STEM results provide insights into the etching process (Fig. 4, and Supplementary Fig. 49), which exhibits a gradual transfer process of the b-ZnO catalyst from a uniform, smooth rod-like structure to elliptical-shaped holes structure. The formation of holes is conducive to exposing the active site and increasing the specific surface area of the catalyst. Moreover, this etching process facilitates the b-ZnO reduction into Zn clusters³³ and the transformation of the b-ZnO substrate catalyst into a Zn/b-ZnO hybrid catalyst that exhibits extraordinary catalytic activity and selectivity towards value-added monomers and lubricant base oil precursors.

Based on the above analysis, the reaction mechanism for the plastic upcycling over Zn/b-ZnO hybrid catalyst under microwave irradiation was studied using n-hexadecane as a model compound. The product distribution after being catalysed by Zn (80 nm), b-ZnO and Zn (80 nm) /b-ZnO was studied to explore the potential catalytic reaction mechanism. It is worth noting that since Zn (80 nm) cannot be heated under microwave irradiation, therefore to ensure as much consistency as possible in the reaction conditions, the catalyst and n-hexadecane were mixed at a mass ratio of 1:5 and placed in a reaction vessel, which was further placed in a quartz sleeve filled with graphite. Under microwave irradiation, graphite converts electromagnetic wave energy into heat and heats the reactive material through heat conduction, realising the catalytic depolymerisation process of n-hexadecane (Supplementary Fig. 70).

Supplementary Fig. 71 presents the carbon number distribution of oil products from n-hexadecane depolymerisation. Interestingly, the oil products are mainly olefins and alkanes when Zn and b-ZnO or Zn/b-ZnO are used as catalysts, respectively, which indicates the dehydrogenation process is efficiently promoted on Zn sites, just like the function of previously reported metal catalysts^{13,35} (e.g. Ir, Rh), additional evidence can be found from a time-on-stream study (Supplementary Fig. 72). Moreover, XRD results can further demonstrate the superior dehydrogenation properties of Zn as the diffraction peaks at the 2θ of 18.1°, 23.4°, 27.3°, 31.7°, 34.4°, 41.3°, 47.8°, 56.8°, 62.9°, 68.4° are detected in the catalyst samples (especially the used Zn catalyst) after catalytic depolymerisation of n-hexadecane, which corresponds to the characteristic diffraction peaks of ZnH₂ and ZnH_{1.96}. While the diffraction peaks at the 2θ of 43.2° and 54.3° are detected in the used b-ZnO catalyst, corresponding to Zn (Supplementary Fig. 73).

Based on a comprehensive series of control experiments, a depolymerisation mechanism of plastics is proposed (Fig. 5). For the b-ZnO catalyst, the plastic reacts with it through initial C-H bond activation to form an intermediate (x1), in which the hydrogen is transferred from the plastic to the O atom and the alkyl chain is

bonded to the Zn atom (a0-a1). Afterward, the β-alkyl chain is eliminated, resulting in the C-C bond scission, forming alkene and a new intermediate (x2) (a1-a2). The produced x2 intermediate can undergo a hydrogen transfer process to obtain alkane compounds, where the hydrogen is transferred from the O atom to the alkyl chain and accompanied by the metal-alkyl bond broken, releasing the b-ZnO catalyst (a2-a3). Additionally, the β hydrogen elimination reaction can also occur, where the hydrogen is transferred from the alkyl chain to the O atom and following the metal-alkyl bond broken, producing alkene and ZnOH₂ (a2-a4). Finally, the ZnOH₂ is decomposed into Zn and H₂O (a4-a5)^{28,33,36}.

The Zn generated via the a4-a5 process also plays an essential role in plastic depolymerisation. The C-H bond activation is the initial step in the plastic depolymerisation reaction, both the hydrogen atom and alkyl chain are shifted and bonded with the Zn atom, forming a y1 intermediate (b0-b1). In the second step, β alkyl elimination reaction causes the C-C bond to break and forms alkene compounds and y2 intermediate (b1-b2). Finally, β hydrogen elimination process facilitates the cleavage of the metal-alkyl bond, resulting in the acquisition of alkene and ZnH₂ (b2-b3).

Eventually, this in-situ conversion of b-ZnO into Zn/b-ZnO catalyst achieves an efficient upcycling of plastics, gradually depolymerising macromolecular polymers into small value-added chemicals through a synergistic catalytic cracking process. Owing to the difficulty of obtaining pure zinc hydride, it is difficult to explore its functional role alone, but it is undeniable that the formation of zinc hydride likely plays a crucial role in breaking C-C bonds and acquiring saturated alkanes^{13,28}.

Further evidence regarding the influence of the Zn and supported catalyst can be observed through comparative experimental results (Supplementary Fig. 74 and Supplementary Table 17). Before the reaction test, the b-ZnO catalyst was pre-reduced in a tubular furnace at 550 °C for 0/1/2/3 h in a 10% H₂/Ar gas mixture (heating rate 10 °C min⁻¹), and then the pre-reduced b-ZnO catalyst mixed with PP with a mass ratio of 1:5. Subsequently, the samples were catalytically depolymerised at 320 W microwave power for 30 min. The gas yield decreases from 66.3 wt% to 35.7 wt% and the oil yield increases from 30.2 wt% to 61.1 wt% as the reduction time of b-ZnO increases from 0 h to 3 h, indicating that b-ZnO loaded with the reduced Zn can selectively improve the oil yield (Supplementary Fig. 75). Moreover, in order to provide more straightforward evidence, Zn/b-ZnO and Zn/AC catalysts were constructed by loading with 1 wt% Zn on b-ZnO and activated carbon (AC) support, respectively. The oil yields of 70.2 wt% and 55.3 wt% are achieved when applying Zn/b-ZnO and Zn/AC catalysts for microwave-assisted catalytic depolymerisation of PP, higher than b-ZnO and AC. These results further proved that the in-situ formed Zn is the active site, and the excellent activity of the Zn/b-ZnO catalyst (Supplementary Table 18).

Discussion

This work presents an in-situ generated Zn/b-ZnO hybrid catalysts with remarkable efficiency, exhibiting outstanding catalytic performance and stability in selectively depolymerising diverse polyolefin plastic waste into valuable hydrocarbons. Leveraging a microwave-assisted catalytic process, this approach facilitates swift depolymerisation, converting over 92 wt% of PP into monomers and high-quality lubricant base oil precursors. Notably, the resultant lubricant base oil precursors represent the predominant product, accounting for 60–85% of the oil products and the corresponding yield reaches 46–60 wt% in all products. Remarkably, this process achieves similar lubricant base oil precursors production when depolymerising unprocessed agricultural film plastic and mixed plastics waste digged out from landfill sites, despite their complex compositions and elevated impurity levels. Crucially, this selective upcycling of the plastic occurs under mild conditions, at 280 °C, with a fast reaction time of just 30 minutes. The



All materials were of analytical grade and used as received without further purification. Commercial ZnO, Zinc acetate dehydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$), and Aqueous ammonia solution (NH_4OH) were purchased from Aladdin Chemical Reagent Co., Ltd (Shanghai, China). Plastic waste such as plastic straws (LDPE), drug bottles (HDPE),

single-used lunch boxes (PP), and car bumpers plastic (PP) are collected from supermarkets. Agricultural film plastic (LDPE) (digged out from landfill site) and mixed plastics waste (digged out from landfill site) are collected from landfill sites in UK.

Preparation of ZnO catalyst

The synthesis process of the b-ZnO substrate catalyst follows: 44 g Zn (CH_3COO)₂·2H₂O were weighed in a container and dissolved in 1800 mL deionised water. The mixture was stirred at room temperature until fully dissolved into a transparent solution. Then aqueous ammonia was added dropwise into the solution. With the dropwise addition of aqueous ammonia, a white precipitate is initially observed, which subsequently dissolves into a clear solution. The clear solution was heated under 85 °C for 6 h to obtain the b-ZnO precipitate. The b-ZnO precipitate was filtrated out and stoved in the oven at 80 °C for 5 h to obtain the catalyst.

The synthesis process of Zn clusters-carbon composite catalysts follows: the made b-ZnO catalyst was mixed with plastic in a ratio of 1:2 and proceeded by different standard reaction cycles. The resulting mixture was washed with 1 M sulphuric acid 3 times and dried under 80 °C for 5 h to obtain the Zn clusters-carbon composite catalysts.

The procedure of microwave catalytic depolymerisation of the plastic

After mixing the catalyst and the plastic, transferring it into a quartz bottle, the filled bottle was placed in the centre of the microwave cavity of a 2.45 GHz magnetron, multimode microwave reactor, the maximum output power was 800 W (Qingdao Microwave Applied Technology Co., Ltd). The sample temperature was measured using an infra-red probe. Although previous works have demonstrated that the temperature measured by the infra-red camera can be significantly lower (−100–300 °C) than the actual bed temperature measured in the centre of the reactor^{37,38}, this depends on the composition of the system and is unlikely to occur during microwave-assisted catalytic depolymerisation of plastics³⁹. Therefore, it is reasonable to use infra-red temperature measurement for the reaction temperature in this work. Argon flow was applied for 30 min at a rate of 100 ml min^{−1} to purge oxygen from the reaction system before the microwave-assisted catalytic depolymerisation of the plastic. Then, the samples were catalytically depolymerised at 320 W microwave power for 30 min. Two-stage cold traps (0 °C) were employed to collect oil products, and non-condensable products were collected in an airbag. The compositions were analysed by gas chromatography (GC-7820), and the mass of each gaseous species was calculated by molar fraction.

$$\text{Gas yield} = \frac{m_{\text{gas}}}{m_{\text{plastic}}} \times 100\% (\text{wt}\%) \quad (1)$$

$$\text{Oil yield} = \frac{m_{\text{reactor}} + m_{\text{cold trap collects}}}{m_{\text{plastics}}} \times 100\% (\text{wt}\%) \quad (2)$$

$$\text{Gas selectivity} = \frac{X_{\text{product } x} \cdot N_{\text{product } x}}{\sum_{\text{product } 1}^{\text{product } n} X_{\text{product } x} \cdot N_{\text{product } x}} \times 100\% (\%) \quad (3)$$

$$\text{TON} = \frac{n \cdot m_{\text{plastic}}}{m_{\text{catalyst}}} \quad (4)$$

Where the m_{gas} , m_{plastic} , m_{reactor} , $m_{\text{cold trap collects}}$ refers to total mass of gaseous products, mass of plastic substrates, mass of oil collected in the reactor, and mass of oil collected in the cold trap, respectively. And

$X_{\text{product } x}$, $N_{\text{product } x}$, n , m_{catalyst} refers to the molar fraction, the carbon number of product x ⁴⁰, cycle number of catalyst, and mass of catalyst, respectively. TON refers to the turnover number of plastic depolymerisation, which is underestimated as we consume that all molecules of the catalyst are active sites.

Electric field strength and distribution simulation

The electric field strength and distribution in the microwave reactor were simulated using COMSOL v.5.6 software. The complex permittivity used for plastic in this simulation is $\epsilon = 2.35 - j \cdot 2 \times 10^{-4}$ ⁴¹, and the complex permittivity for the b-ZnO catalyst is 12.4·j^{1.18}, which is obtained from the vector network analyser results (2.45 GHz, Supplementary Fig. 2a, b).

Characterisation and analysis

The crystalline structure of the catalyst before and after use was characterised by XRD on a Bruker D8-advance diffractometer using Cu K α ($\lambda = 1.54056 \text{ \AA}$). Morphologies of the catalyst before and after use were examined by QUANTA FEG 250 field emission scanning electron microscope (SEM). High-angle annular dark-field scanning TEM (HAADF-STEM) images were captured by an Aberration-corrected transmission electron microscope (JEM-ARM300F) operated at an accelerating voltage of 300 kV. X-ray absorption near-edge spectra (XANES) and extended X-ray absorption fine structure spectra (EXAFS) measurements of Zn were performed at the beamline 1W1B of the Beijing Synchrotron Radiation Facility (BSRF), Institute of High Energy Physics (IHEP), Chinese Academy of Sciences (CAS) in a transmission mode. The carbon residues were measured by in Via-Qontor Raman spectroscopy with a 532 nm laser. A gas chromatograph coupled to a high-resolution mass spectrometry (GC-MS QP2010 Ultra) with an Rtx-5MS column ((5%-phenyl)-methylpolysiloxane, 30 m × 0.25 mm) was employed to analyse the oil product. The compositions of gas products were analysed by gas chromatography (GC-7820, Shandong Huifen Instrument Co., Ltd.), permanent gases were detected by thermal conductivity detectors (TCD) with a TDX-O1 carbon molecular sieve column (3 mm × 2 m), and hydrocarbons were detected by a flame ionised detector (FID) with an HP-PLOT-Q column (30 m × 0.25 mm). Thermogravimetric analysis (TGA) was used to characterise the carbon yield of used catalysts, heated from room temperature to 900 °C with a heating rate of 10 °C min^{−1} in an air atmosphere. High-temperature gel permeation chromatography (GPC) was performed on an Agilent PL 220 equipped with a refractive index (RI) detector and a PL-Gel Mixed B columns, the samples were dissolved in 1,2,4-trichlorobenzene (TCB) containing di-tert-butylhydroxytoluene (BHT, 0.01 wt%) and heated at 150 °C at least 1 h to ensure the sample was completely dissolved. Then, GPC columns were eluted at 1.0 mL/min with TCB (with BHT) at 150 °C. The elemental valence state information of catalysts' surfaces before and after use was obtained by X-ray photoelectron spectroscopy (XPS ThermoFisher ESCALAB 250Xi). ¹H and ¹³C NMR spectrum was recorded with Ascend 600 MHz spectrometer at room temperature, CDCl₃ was used as the solvent. Simulated Distillation Gas Chromatography Flame Ionization Detection (SimDist GC-FID) was performed on an AC Agilent 6890 gas chromatograph equipped with a split/splitless injector and a flame ionisation detector (FID). H₂-TPR was obtained on AutoChem II 2920. Before measurements, the catalyst sample was flushed with 99.99% N₂ at 120 °C for 0.5 h to remove traces of water and naturally cooled down to 50 °C. After that, the temperature was increased from 50 to 700 °C with a heating rate of 10 °C min^{−1} in a 10% H₂/N₂ atmosphere.

Data availability

The authors declare that all data are available in the main text or the supplementary information. Source data are provided with this paper.

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Author contributions

W.M., T.X. and J.T. supervised the entire project. J.Z., B.L., W.M., T.X. and J.T. conceived and designed the project. J.Z. designed and developed the experiments and evaluated and analysed the performance data. L.X. and W.L. designed and performed the X-ray absorption spectroscopy measurements and analysis of the data. J.Z., D.W., and L.X. drafted the original manuscript. All authors, including L.J., J.Y., P.W., S.Z. and P.E. contributed to the analysis, interpretation and discussion of results and the writing and revisions of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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