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Recent progress for microwave catalytic pyrolysis of plastic waste to valuable chemicals

Mudi Wu[a] and Mingwu Tan *[ab]

Plastic waste is accumulating in our environment, causing severe threats to the ecosystem and human health. Conventional recycling methods often suffer from low efficiency and limited product value, hindering their extensive application. By comparison, microwave catalytic pyrolysis of plastic waste is a promising route due to its rapid and uniform heating, high energy efficiency, and superior product selectivity. Here we review recent progress in applying microwave catalytic pyrolysis for plastic waste upcycling from the perspective of type, function, and synthesis strategies of catalysts. This review introduces the fundamental principles of microwave heating in the catalytic pyrolysis process, and the conversion pathway of plastic waste over different catalysts, emphasizing the relationship between catalyst properties and product selectivity. The high economic viability of this technology provides huge potential for its industrial application. Finally, challenges, including catalyst stability, mechanism study, and the gap between lab and real-world scenarios, are also critically discussed. Overall, microwave catalysis shows significant potential for plastic waste upcycling.

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1 Introduction

Over the last few decades, plastic has been an indispensable material in both daily life and industrial production.[1,2] The excellent properties, such as lightweight, durability, low cost, *etc.*, have driven a consistent increase in global demand when offering convenience to modern society.[3] However, its rapid growth also indicates a high environmental cost. To date, the annual production of plastics has reached 414 million tons in 2023 (Fig. 1a), and the cumulative production is estimated to be more than 10 billion tons.[4–6] When plastic waste is not managed properly, these once-beneficial materials will transition into persistent environmental pollutants, posing significant ecological and economic challenges to the ecosystem. Although some advanced recycling policies have been implemented in certain developed regions, the global community still lacks a sustainable and scalable solution to effectively mitigate the ever-growing volume of plastic waste.[7] It's reported that only 21.3% of the plastic waste was recycled, and the remaining was incinerated or discarded, including landfilling or disposal into the ocean or natural environment[8] (Fig. 1b and c). Taking polyethylene (PE) as an example, as the most widely produced plastic globally with a market share of approximately 30%, it exhibits extreme resistance to natural degradation, with a decomposition timeline exceeding 200 years under typical environmental conditions,[9] which are 450–1000 years for PET, 10–600 years for PP, 50–80 years for PS, and 50–150 years for PVC.[10,11] This persistence in our environment not only intensifies plastic accumulation but also contributes to long-term ecological harm, including microplastic pollution and habitat disruption, which will ultimately affect human health.[12–14] Given the low proportion of recycled plastic quantities compared to the amount produced, the trend will pose a serious and lasting threat.[15–17] Hence, developing a better and more efficient way to recycle plastic waste is urgent. Current pathways of landfill and incineration for plastic waste treatment have low utilization efficiency and may cause severe secondary pollution to our environment,[18,19] and therefore, many researchers have been trying to find alternative valorization strategies to achieve more productive and sustainable recycling of plastic waste. At present, the most widely applied thermal-chemical solutions for plastic waste utilization include catalytic cracking, gasification, and pyrolysis (Fig. 2).[20,21] Considerable efforts have been devoted to producing value-added chemicals and fuels from plastic waste.[22–24] Although these approaches could effectively valorize plastic waste, they also face critical limitations, including high energy consumption, poor heating uniformity, and slow temperature ramping rates, all of which hinder their industrial scalability and economic feasibility. Therefore, the development of alternative strategies that are both energy-

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[a] *School of Chemical & Biomedical Engineering, Nanyang Technological University, 62, Nanyang Drive, 637459, Singapore*
[b] *Institute of Sustainability for Chemicals, Energy, and Environment (ISCE^2), Agency for Science, Technology, and Research (A*STAR), 1 Pesek Road, Jurong Island, 627833, Singapore. E-mail: Tan_Mingwu@a-star.edu.sg*

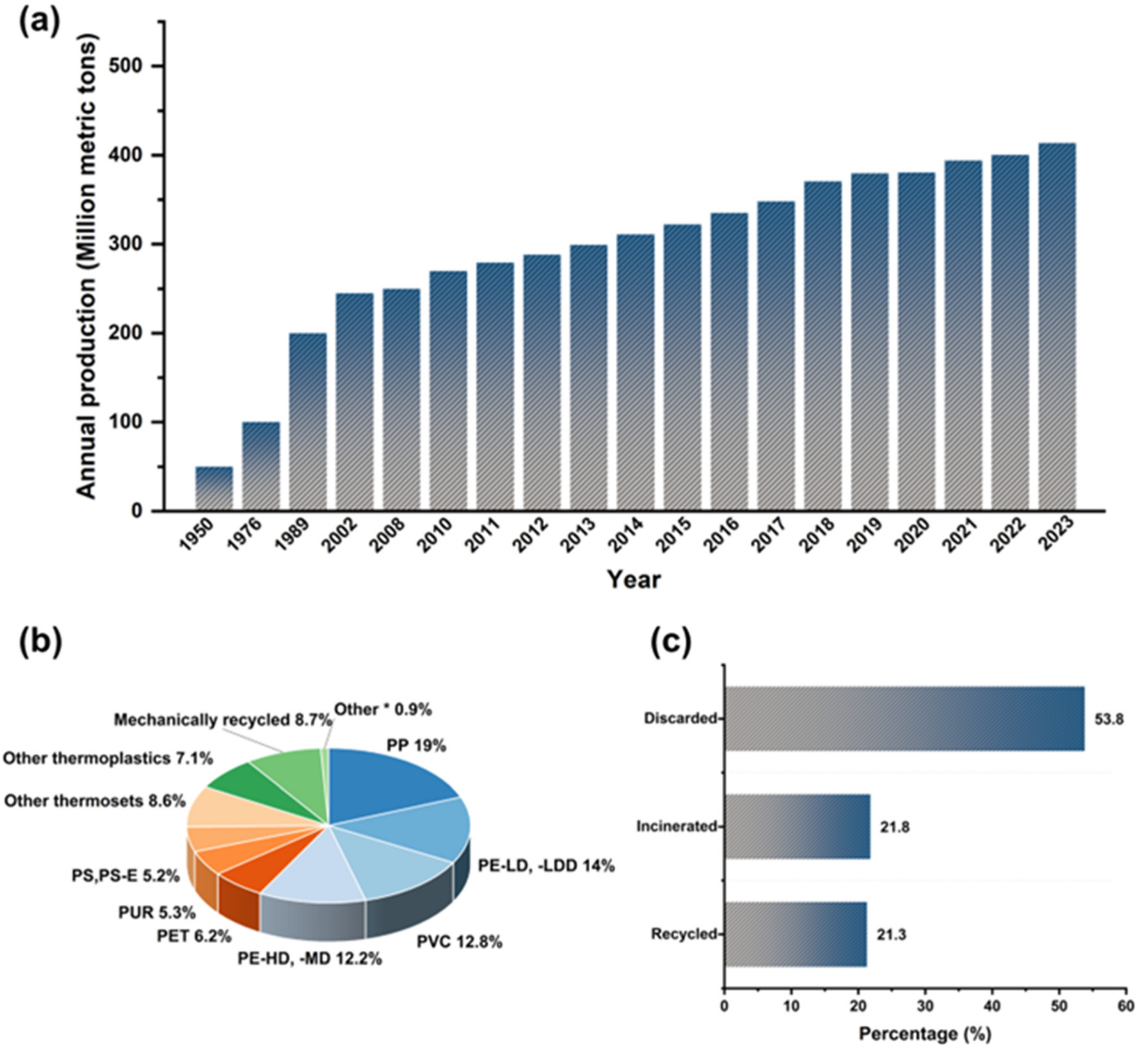

Fig. 1 a) Annual production of plastics worldwide from 1950 to 2023.[34] b) Distribution of plastic production worldwide in 2023, by type.[35] c) Distribution of plastic waste treatment worldwide in 2018, by method.[36]

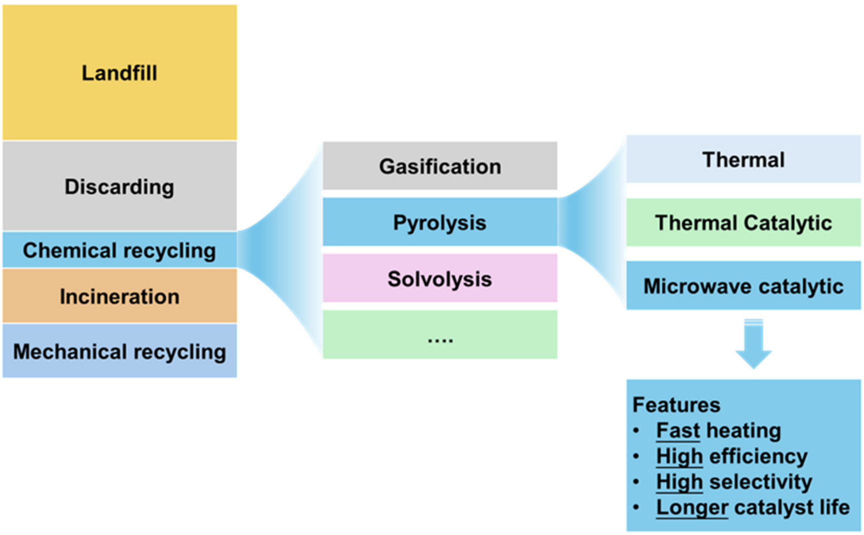

Fig. 2 Typical strategies for plastic waste treatment and diverse solutions for chemical recycling.

efficient and highly selective is urgently necessary. Microwave catalysis has recently emerged as a promising solution for plastic waste upcycling.[25–27] Because of the unique features of rapid and uniform heating at the molecular level, microwave catalytic processes could overcome many of the challenges inherent in conventional heating, including low energy efficiency and limited heat transfer. Since the energy is directly converted from electromagnetic waves to the absorbing material, it leads to a uniform heating of the substrate. This is superior to conventional heating strategies, which typically require an external heat source that may cause significant temperature gradients in both the axial and radial directions within the catalyst bed, and even at the intra-particle scale. Considering the superior ability in addressing the challenge of plastic waste, this route aligns well with the United Nations Sustainable Goals (UN SDGs), especially SDG 7 (Affordable and Clean Energy) and SDG 12 (Responsible Consumption and Production).[28,29] Consequently, the microwave technology has seen increasingly widespread application in recent years, from its initial domestic use to a wide range of industrial processes, including ceramic sintering,[30] nondestructive microwave testing,[31] and, more recently, the conversion of waste plastics and biomass.[32,33]

Given the ever-rapidly growing amount of plastic waste and the fast development of microwave catalytic technology, a timely and critical summary of the latest work is necessary. This work offers a comprehensive overview of recent progress in microwave catalytic pyrolysis of plastic waste from the perspective of application routes including H_2/carbon, monocyclic aromatic hydrocarbons (MAHs), liquid fuels, co-pyrolysis with biomass, CO_2 coupling & reforming (Fig. 3). Furthermore, a systematic and in-depth introduction from the microwave function mechanism, the type, design, and role of catalysts, and current applications and challenges are provided. We hope this work can help researchers get a clear insight into the current research landscape focus and a better understanding of the potential and challenges in microwave catalytic pyrolysis for plastic waste valorization.

2 Microwave catalysis mechanism

2.1 Microwave heating mechanism

Microwaves are a type of electromagnetic wave with high frequencies ranging from 300 MHz to 300 GHz and short wavelengths ranging from 0.001 to 1 m. For the heating of an irradiated object *via* microwave, its heating rate is highly dependent on the microwave absorption properties, and the absorption behavior follows the following equations.

The total power absorbed in a target material is shown in eqn (1):[37]

$$P_{\text{abs}} = 2\pi f \varepsilon_0 \varepsilon_r'' E^2 \quad (1)$$

where P_{abs} represents the local power density in the material (W m^{-3}), f is the frequency of incident microwave, ε_0 is the permittivity of free space ($\varepsilon_0 = 8.854 \times 10^{-12}$ F m^{-1}), and ε_r'' is the effective dielectric loss factor; E is the local value of the electric field strength.

The scalability of microwave-assisted synthesis is fundamentally constrained by the penetration depth (D_p), as sample volume increases, power attenuation limits heating uniformity and efficiency. The penetration depth follows eqn (2) and (3):[38]

$$D_p = \frac{\lambda_0}{2\pi\sqrt{2\varepsilon'}\sqrt{\sqrt{1+\tan^2\delta}-1}} \quad (2)$$

When the value of $\tan\delta$ is low, the equation could be simplified as below:

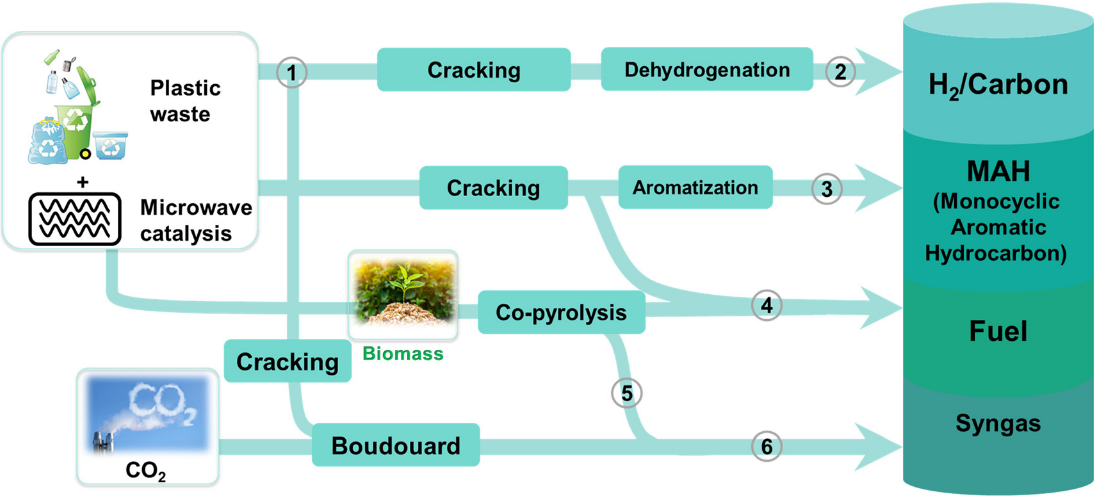

Fig. 3 Roadmap of recent advances in microwave-assisted catalytic pyrolysis of plastic waste: plastic waste upcycling routes: 1. primary catalytic cracking; 2. deep dehydrogenation & carbon growth; 3. intermediate aromatization to monocyclic aromatic hydrocarbon (MAH); 4. selective pyrolysis to liquid fuels; 5. synergistic co-pyrolysis with biomass; and 6. CO_2 coupling & reforming (Boudouard-driven).

$$D_\mathrm{p} = \frac{\lambda_0}{2\pi\sqrt{2\varepsilon'}} \quad (3)$$

where ε_r' is the relative dielectric constant, which is a measure of the ability of the dielectric material to store electrical energy.

Furthermore, the dielectric loss tangent, $\tan\delta$, is used to describe the interaction of a dielectric with microwaves, as shown in eqn (4):[39]

$$\tan\delta = \frac{\varepsilon_\mathrm{r}''}{\varepsilon_\mathrm{r}'} \quad (4)$$

In addition, the heating rate is given by eqn (5).[40]

$$\rho C_\mathrm{p}\frac{\mathrm{d}T}{\mathrm{d}t} = P_\mathrm{abs} \quad (5)$$

where C_p is the specific heat capacity of the material, $\mathrm{d}T/\mathrm{d}t$ is the temperature enhancement rate, and ρ is the material density.

In practical application, the heating pathway is highly related to the target material and the associated conversion process.[41] Dipolar polarization loss is one of the primary ways for microwave energy to convert into heat. Take polar liquids like water or alcohol as an example, molecules experience rapid reorientation under an alternating electric field, resulting in rotational motion. Correspondingly, this phenomenon would cause the friction between molecules, thereby facilitating the conversion of microwave energy into thermal energy.[42,43] Ionic conduction loss is another important way for the conversion of microwave energy into heat.[44] The charged ions will oscillate back and forth in the high-frequency microwave field and experience frequent collisions with adjacent atoms or molecules, resulting in the temperature increase as well. The resulting thermal energy contributes significantly to microwave heating in materials with high ionic conductivity. Together, these two pathways constitute the typical forms of dielectric loss. Aside from the above-mentioned two types of heating mechanisms, the generation of eddy or plasma over metal-based materials is another pathway that cannot be overlooked. In conductive materials, free electrons could also be driven by the alternating electric field, producing conduction currents. As these currents circulate, especially in materials with non-uniform conductivity, eddy currents will increase, which induce internal resistive heating accordingly.[45] In plasma or partially ionized gases, free electrons accelerate in the microwave field and collide with neutral atoms and ions, converting their kinetic energy into thermal energy. These effects of electrical conduction, eddy current, and collision within plasma systems together make this pathway a highly efficient way for converting microwave energy into thermal energy.

2.2 Catalyst classification

For most plastics, the melting temperature is below 200–300 °C, which would cause inhomogeneous spatial temperature gradients among reactants and catalysts, demanding precise regulation of the reaction process for optimal performance. Furthermore, the heat transfer difference between the catalysts and plastic would also aggravate uncontrollability, impeding the efficient valorization of plastic waste. Conversely, microwave heating, compared to conventional heating, would not wait for the heat-up of the whole system, and the inherent property of fast heating process from ambient temperature (T_ambient) to target temperature (T_target) of studied catalysts could avoid tremendous side reactions accordingly (Fig. 4).[46] Therefore, designing catalysts that could effectively adsorb or interact with microwaves is the key determinant for microwave catalysis processes. For example, aside from serving as an active site for C–C/C–H bond cleavage, iron oxide also exhibits high magnetic loss, allowing it to rapidly increase in temperature. In contrast, alumina (Al_2O_3) exhibits a weak microwave response but possesses high thermal conductivity and excellent structure stability, making it a suitable support for active site loading.[47,48] Furthermore, zeolites have been widely applied in microwave catalysis due to their tunable acidity, high pore volume, large specific surface area, and shape-selective pore structures.[49,50] These properties enrich the potential applications of plastic recycling technologies. Currently, composite catalysts such as Fe/ZSM-5 are being developed to offer enhanced functionality and higher efficiency in the microwave pyrolysis of plastic waste.[51] To date, four different types of catalysts have been developed for the microwave catalytic pyrolysis of plastics, as detailed below.

2.2.1 Type I: metal-based catalysts (*e.g.*, Fe, Co, and Ni). Transition metal species exhibit strong microwave responsiveness, and they can rapidly generate heat through magnetic hysteresis or eddy current losses upon microwave irradiation, accompanied by a fast temperature increase of the catalysts. Considering transition metal oxides could also catalyze the cracking process, microwave irradiation would facilitate the activation and cleavage of strong C–C and C–H bonds in plastic waste. Additionally, their high thermal conductivity helps maintain a uniform temperature distribution at the microscale, which could reduce side reactions accordingly.[52–54]

2.2.2 Type II: metal oxide-based catalysts (*e.g.*, MgO, ZnO, and Al_2O_3). Other metal oxides, such as Al_2O_3, SiO_2, and MgO, have weak intrinsic microwave absorption capability but show high thermal stability, mechanical strength, and chemical inertness, making them ideal components to serve as additives, supporting, or structural materials. These oxides could facilitate the homogenization of temperature distribution during microwave catalytic reactions and provide a stable framework for dispersing active metal species. For example, Al_2O_3 and MgO are widely used to prevent catalyst sintering at high temperatures and maintain an active surface area during long-term operation.[55,56] Furthermore, more roles have been reported for metal oxides in recent years. For example, Zn/ZnO is found to serve as an active site for C–C cleavage during plastic pyrolysis.[57]

2.2.3 Type III: zeolite-based catalysts (*e.g.* ZSM-5 and USY). Zeolites are a class of microporous or mesoporous

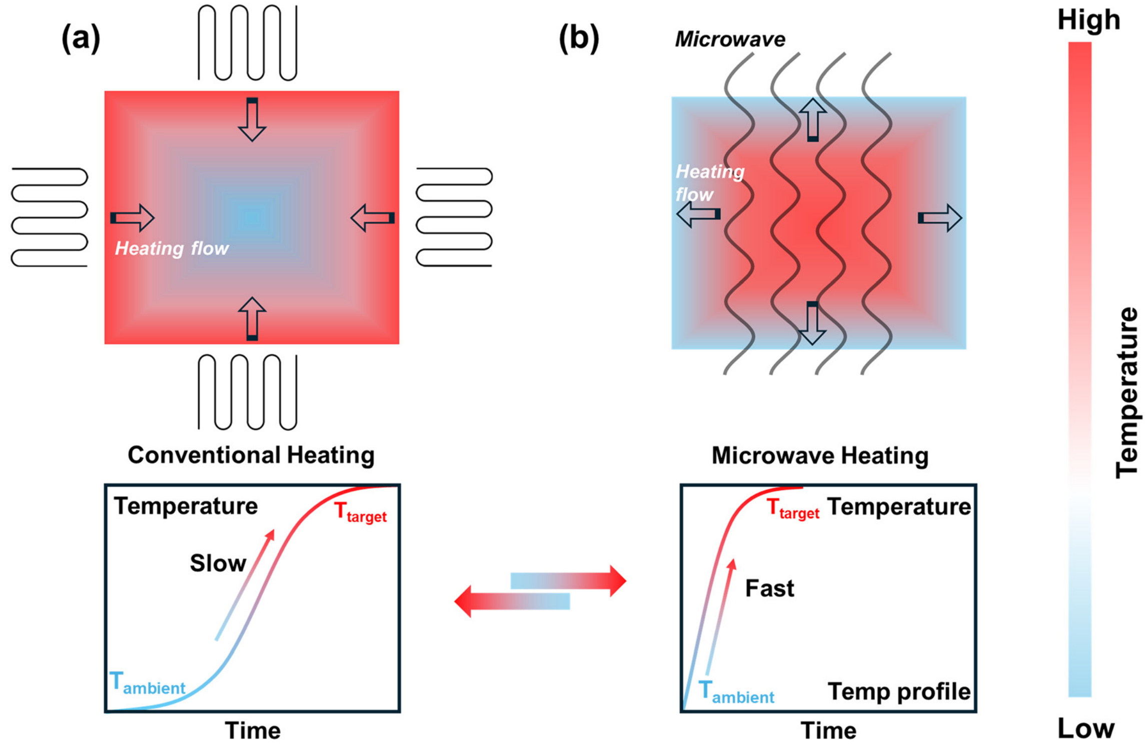

Fig. 4 Comparison of heat transfer between a) conventional heating and b) microwave heating.

aluminosilicates, which have moderate microwave responsivity and abundant acid sites. The unique pore structure could function as a shape-selective catalyst during the plastic cracking process.[58–60] These properties facilitate the selective cleavage of C–C bonds in polymer chains and thereby regulate product distribution toward target high-value products. In addition, the tunability of Brönsted and Lewis acid sites in zeolites further allows for precise control over reaction pathways, making them ideal candidates for valorizing plastic waste into high-value chemicals.

2.2.4 Type IV: other materials (*e.g.*, SiC and carbon). Silicon carbide (SiC) and carbon-based materials exhibit outstanding dielectric loss characteristics, which makes them excellent microwave absorbing materials.[61] Microwave irradiation over SiC could induce localized hotspots. This greatly increases the activity of loaded catalysts by accelerating reaction kinetics at the microscale. Moreover, its high thermal conductivity and structural stability make it a good choice for both support and microwave absorber. Similarly, carbon materials, such as activated carbon, biochar, and CNTs, are widely used due to their high microwave absorption, good thermal stability, and tunable surface properties (various surface functional groups). In addition, the large surface area could also facilitate the dispersion of active species,[62,63] and their electrical conductivity helps facilitate microwave absorption and charge transfer, thereby improving pyrolysis efficiency during the microwave catalytic degradation of plastic waste.

Currently, the commonly reported catalysts often comprise multiple types of materials to tune the synergy between catalysts, microwave irradiation and reactant environment to facilitate the selective and efficient conversion of plastic waste into high-value products, like carbon nanotubes, hydrogen, liquid fuels, monocyclic aromatic hydrocarbons (MAHs), *etc.* (Fig. 5). For example, transition metal oxides (Fe_2O_3, Co_3O_4, or NiO) are often incorporated into metal oxide supports (Al_2O_3, MgO), or within zeolite frameworks (ZSM-5 or Beta zeolite) to ensure high activity and good response to microwave. Hence, understanding the properties of these materials could help researchers to precisely tune the catalysts' properties in their own work, including acidity, redox capacity, pore structure, and microwave absorption capability. To illustrate that, the following sections discuss a series of representative cases to help understand the structure–performance relationships in microwave-catalytic plastic upcycling.

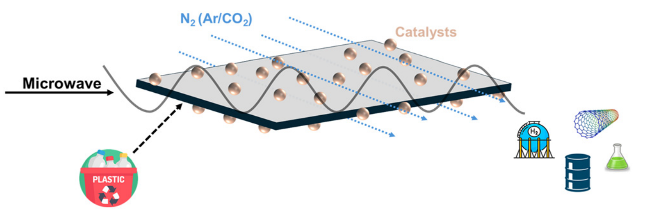

Fig. 5 Schematic illustration of microwave catalytic conversion of plastic waste into diverse products.

3 Applications in plastic valorization

3.1 Hydrogen and carbon material production

The direct pyrolysis of plastic waste to high-purity hydrogen, along with massive high-value carbon materials, has attracted extensive attention recently. Considering the simple composition of plastic, the direct separation of H/C elements from plastic waste to H_2 and carbon materials, simultaneously, is one efficient pathway that not only reduces pollution but also generates economic benefits with high selectivity. Furthermore, it's reported that the produced hydrogen is of high purity and can be easily purified, making it highly suitable for clean energy applications. As shown in Table 1, the catalysts based on Fe, Co, and Ni species are predominantly employed for the microwave pyrolysis of plastic into H_2 and carbon, due to their strong response to microwave and significant magnetic loss.[45,64] It's worth noting that the formed carbon material is mainly composed of high-value carbon nanotubes, accompanied by a high yield of H_2 (more than 97% hydrogen recovery). In recent years, one of the most significant advances for microwave catalytic pyrolysis of plastic waste is the one-step microwave catalysis of plastic waste over $FeAlO_x$ catalysts reported by Jie *et al.* in 2020. A high hydrogen yield of 55.6 mmol $g_{plastic}^{-1}$ is achieved over HDPE (high density polyethylene) under 1000 W microwave power, with more than 97% hydrogen recovery in 90 seconds, accompanied by the production of multiwalled carbon nanotubes.[26] The high magnetic losses of $FeAlO_x$ catalysts promote the efficient conversion of microwave energy into heat with rapid temperature increases, and the cleavage of the C–C and C–H bonds, leading to the efficient separation of hydrogen and carbon atoms. Moreover, iron could facilitate the dissolution and precipitation of carbon, therefore benefiting the carbon nanotube (CNT) growth. Similarly, Zhang *et al.* synthesized a two-dimensional porous $FeAlO_x$ catalyst by varying Fe/Al ratios and applied it in LDPE microwave pyrolysis.[65] The experimental results showed that the unique structure significantly enhances microwave capabilities to 90%, and $Fe_1Al_{0.5}O_x$ (molar ratio of Fe : Al = 1 : 0.5) could produce 60.2 mmol of H_2 per gram of plastic under a constant input power of 1000 W, with hydrogen accounting for 87.5% of the gaseous products by volume and ~84% of the theoretical H_2 production. Additionally, the residual carbon was converted into highly graphitized multi-walled carbon nanotubes (MWCNTs), with a yield of up to 571 mg g^{-1} of LDPE, even after three cycles. The mechanism study by DFT verified the high C–H bond cleavage activities of iron species. Zhou *et al.* treated $FeAlO_x$ catalysts with air plasma to introduce abundant oxygen vacancies and employed them for polyethylene (PE) degradation in combination with carbon fiber cloth, using a multi-mode microwave reactor.[66] The addition of carbon fiber cloth significantly enhances the microwave absorption capacity, accelerating both the initiation of heating and the overall reaction rate. The results showed that the introduction of a high density of oxygen vacancies in the $FeAlO_x$ catalysts enabled a record-high hydrogen yield of 67.3 mmol g^{-1} of plastic through the pyrolysis of PE waste. To further enhance its performance and optimize the product distribution, doping of a second metal, like nickel or cobalt, is always regarded as an effective strategy. The composite catalysts show higher dielectric loss and magnetic loss than iron-containing only catalysts, indicating the high energy conversion efficiency and faster temperature ramping.[67,68] Meanwhile, bimetallic components could improve the dispersion of active sites and stability by suppressing the aggregation phenomenon, thereby achieving superior reactivity.[69,70] For example, by doping cobalt into Fe–Al catalysts, Qian *et al.* synthesized $FeCo_1Al_2$ (cobalt-doped $FeAl_2O_4$) catalysts for LDPE pyrolysis, and the yield of hydrogen was reported to reach 61.39 mmol $g_{plastic}^{-1}$ (~86% of the theoretical H_2 production) under the microwave power of 700 W.[71] Compared to undoped Fe_1Al_2 catalysts, cobalt-doped Fe–Al catalysts exhibit higher H_2 yield and H_2 concentration in the gaseous product, accompanied by higher carbon yield of carbon nanotubes. The performance enhancement is attributed to the doping of cobalt, hindering the formation of Fe_3C, which changes the carbon diffusion pathway and promotes the formation of carbon nanotubes and hydrogen. Similarly, Zhang *et al.* synthesized

Table 1 Summary of microwave catalytic conversion of plastic waste to H_2 and CNTs

Feed	Catalyst	Reaction condition	H_2 yield-mmol $g_{plastic}^{-1}$	Carbon type (yield-%)	Ref.
HDPE	$FeAlO_x$	1 bar/N_2	55.6	CNTs (~70)	26
LDPE	$Ni_3Co_2O_x$	1 bar/Ar	63.5	MWCNTs (−)	52
Mixed plastic waste	$Ni_3Co_2O_x$	1 bar/Ar	44.1	MWCNTs (−)	52
HDPE	$Fe/FeAl_2O_4$	1 bar/Ar	47.03	CNTs (−)	78
LDPE	$FeNiAl_2O_4$	1 bar/N_2	52.1	CNTs (44.5)	73
PE	Fe/AC	1 bar/N_2	28.65	CNTs (−)	53
PP	Al-Fe	1 bar/N_2	44.07	Bamboo shape CNTs (−)	79
LDPE	2D $AlFeO_x$	1 bar/N_2	60.20	MWCNTs	65
LDPE	FeCoAl	1 bar/N_2	61.39	MWCNTs	71
PE	$FeAlO_x$	1 bar/N_2	66.4	MWCNTs (46.2%)	80
HDPE	Al_2O_3-$NiFe_2O_4$	1 bar/N_2	75 vol%	CNTs (78%)	47
Mixed plastic waste	$La_{0.6}Ca_{0.4}FeO_{3-\delta}$	1 bar/N_2	43.7	MWCNTs (−)	76
Masks	$FeAlO_x$-500	1 bar/N_2	29.03	MWCNTs (−)	77
Masks	FeCoAl	1 bar/Ar	38.66	CNTs (19.65%)	74

a series of NiFeAl composite catalysts and used them for LDPE microwave pyrolysis.[72] Through effectively tuning the metal–metal interaction, the microwave absorption and energy dissipation capabilities were increased remarkably, facilitating the cleavage of C–C and C–H during microwave irradiation. Compared to iron, nickel can facilitate bond scission, and the existence of multiple elements within the catalyst is beneficial in providing more active sites in successive tests. The as-synthesized catalysts NiFeAl-450 achieved a maximum H_2 yield of 60.5 mmol g_{LDPE}^{-1} with a H_2 concentration of 85.1 vol% in gas products (84.7% of the theoretical H_2 yield), alongside high-value MWCNTs. To verify the superiority of the bimetallic system, Chaouki *et al.* studied the performance of Fe, Ni, Co, and their mixture phase for LDPE valorization to carbon nanotubes and H_2.[73] The TEM study shows that the introduction of Ni or Co facilitates Fe dispersion, while the formation of the alloy phase promotes catalytic performance with a high yield of CNTs (44.5 wt%) with enhanced quality and superior H_2 yield (52.1 mmol $g_{plastic}^{-1}$, ~73% of the theoretical H_2 yield). Moreover, Zhang *et al.* introduced a secondary element, including cobalt and nickel, into $FeAlO_x$ catalysts to enhance their defect density and microwave absorption capability for deposed mask treatment.[74] During the pyrolysis process, 77 wt% gas and 20 wt% carbon nanotubes were produced within 14 minutes, with 90% of the gas phase composed of CO and H_2. The FeCoAl catalyst demonstrated a dehydrogenation efficiency of 38.66 mmol H_2 per gram of mask material. Instead of serving as a dopant, Co or Ni could serve as an effective active site itself, while nickel has been known as one of the most efficient active metals for catalyzing C–C bond dissociation, facilitating the carbon deposition and CNT growth accordingly.[52,54] In addition to exhibiting functions like Ni, cobalt could further improve the dehydrogenation capability and product selectivity. $NiCoO_x$ composite catalysts without iron also exhibit superior performance for microwave catalytic decomposition of LDPE.[52] The hydrogen yield reaches 63.5 mmol $g_{plastic}^{-1}$ with a high selectivity of 83.7 vol%, which is almost 2–3 times that of traditional thermal catalysis. Furthermore, bimetal catalysts possess much higher H_2 purity than that of single-metal catalysts, including Co_3O_4, NiO, or Fe_2O_3, due to the improved activity by the enhanced electron effect between bimetallic species. This observation underscores that dispersion of the active site is important for performance enhancement, which can maximize the atom efficiency of the metallic components.

In addition to the above-mentioned transitional metals, rare earth metals also serve as important promoters in microwave catalysis for plastic waste pyrolysis. For example, Li *et al.* synthesized a series of trimetallic catalysts by adding Pr, Ce, La, or Yb to a Co–Fe bimetallic catalyst. Catalyst Pr_1-$Co_1Fe_8O_x$ shows the highest yield of H_2, with a hydrogen efficiency as high as 95.56% (total H in H_2 and CH_4 counted) for pyrolysis of LDPE and few byproducts.[75] The promotion effect is attributed to the enhanced oxygen vacancy effect exerted by the Pr^{3+} cation. Similarly, perovskite oxide, a kind of rare-earth metal material, also shows potential for plastic degradation. For example, Chen and his group synthesized the $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ nanosphere catalyst and used it for mixed plastic waste decomposition to hydrogen and carbon materials.[76] The hydrogen yield reached 43.7 mmol $g_{plastic}^{-1}$, approaching ~80% of theoretical H_2 production, and the surface-exsolved iron species function as active sites for carbon nanotube formation, which have a longer length and smoother surface. Beyond microwave catalytic decomposition of single plastic waste, the treatment of mixed or real-world plastic waste is of greater practical significance. For instance, Liu *et al.* applied cobalt-doped $FeAlO_x$ catalysts to the treatment of disposable medical masks under the microwave power of 1000 W.[77] Results show that 78 wt% of the plastic waste was converted into gaseous products, and hydrogen is the main component, while the solid product is carbon nanotubes (CNTs). It's worth noting that carbon products evolve into bamboo-like CNTs by decreasing the reduction temperature of the iron catalyst. This study demonstrates the huge promise of microwave catalytic pyrolysis for converting realistic waste plastics into hydrogen and high-value carbon products. Similarly, Zhao *et al.* adopted $Ni_3Co_2O_x$ as catalysts for realistic plastic waste, and a high hydrogen yield of 44.1 mmol $g_{plastic}^{-1}$ is also achieved when the feedstock is extended to the landfill mixed plastics waste, further demonstrating the huge potential of microwave catalytic pyrolysis in practical scenarios.[52]

Overall, the research on transition metal-based catalysts, accompanied by morphology engineering, elemental doping (including rare earth metals), and defect engineering, has significantly enhanced catalytic activity and product selectivity for plastic waste upcycling. Extensive literature studies indicate that transition metal-based catalysts play a vital role in microwave catalysis pyrolysis to high-value carbon and high-purity H_2. Besides their ability to respond excellently to microwaves, Co, Fe, and Ni catalysts not only promote dehydrogenation and cracking of plastic waste but also favor the growth of high-value carbon materials. All these features would serve as a strong basis for new catalyst development.

3.2 MAH production

Monocyclic aromatic hydrocarbons (MAHs) are a fundamental chemical feedstock that is mainly obtained from the processing of petroleum, methanol, and coal tar. Compared to polycyclic aromatic hydrocarbons (PAHs), MAHs, including benzene, toluene, ethylbenzene, and xylene (collectively known as BTEX), offer higher economic value and broader industrial applicability. Hence, the catalytic upcycling of plastic waste into MAHs is also one of the most promising strategies for plastic waste upcycling.[81,82]

As shown in Fig. 6, the conversion from plastic waste to aromatics involves multiple steps: firstly, short-chain olefins are produced after thermal cracking of long-chain

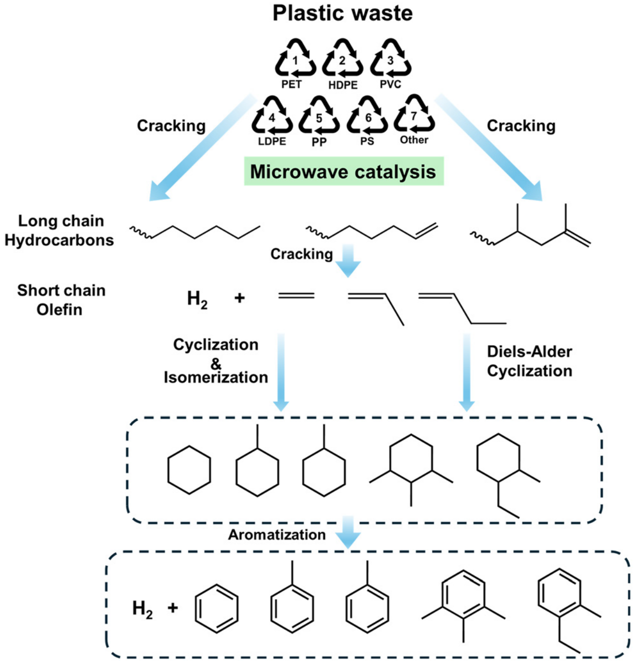

Fig. 6 General mechanism for the upcycling of plastic waste into monocyclic aromatic hydrocarbons (MAHs).

hydrocarbons; then, these olefins are converted into aromatic hydrocarbons *via* cyclization and isomerization, followed by dehydrogenation to produce monoaromatic hydrocarbons (MAHs). Additionally, polycyclic aromatic hydrocarbons may be formed by oligomerization and Diels–Alder reactions of olefins. The presence of strong acid sites promotes these transformations, accompanied by hydrogen release.[58,83] Table 2 lists the most current work about MAH production from plastic waste by microwave catalysis, and the yield could reach as high as 65%, illustrating the significant potential in industrialization. In the meantime, the most widely used catalysts for MAH production are zeolite-based, which provide abundant acid sites and shape selection functions that can, in turn, tune the MAH selectivity correspondingly. For example, Wang *et al.* compared the performance of different zeolites in LDPE microwave catalytic pyrolysis to MAHs, and they found that HZSM-5 exhibits the highest yield and the most centered product distribution.[58] The distinct catalytic effects were mainly attributed to the different pore structures and acid site concentration. More specifically, the cracking of LDPE mainly occurs inside HZSM-5, which has a pore size of around 0.5 nm in diameter and relatively high acidity. The smaller pore size can extend the residence time of C_3–C_5 products, and the abundant acid sites would facilitate secondary reactions, especially aromatization. Conversely, due to the large pore size and low concentration of acid sites, the main product obtained by MCM-41 was olefins. Hence, zeolite with a suitable pore structure and acidity is important for optimizing the product distribution. Further, Jiang *et al.* adopted zeolite USY as a catalyst for converting PS to MAHs.[84] Experimental results show that zeolite USY with a high surface area (734 $m^2\ g^{-1}$), large pore size (5.6 nm), and abundant strong acid sites (1.21 mmol g^{-1}) exhibits the most effective shape selectivity for ethylbenzene and benzene generation from PS. By reducing the Si/Al ratio, the acid sites increase, and more aliphatic chains break down to benzene and ethylbenzene. Furthermore, the type of plastic waste was also found to affect the production of aromatic hydrocarbons. Experimental results show that ethylbenzene production was effectively enhanced when using polystyrene as a feed, which is 54 times higher than that from polycarbonate. This underscores the importance of a suitable match between the zeolite catalyst and plastic type to achieve optimal efficiency. To produce more condensate fraction with higher selectivity for MAH products, Wang *et al.* adopted a dual-catalyst bed of MCM-41 and HY for continuous LDPE microwave pyrolysis.[82] Due to the larger pore size, MCM-41 helps crack long-chain polyolefin intermediates into shorter chains first, which can alleviate the steric and diffusional resistance while short-chain products pass through the micropore of the HY zeolite. Through such a strategy, the maximum liquid yield (63.75 wt%) and MAH selectivity (78.21%) were achieved at a temperature of 450 °C, feedstock to catalyst ratio of 8 : 3, and HY to MCM-41 ratio of 2 : 1. To further extend the application of microwave catalysis to practical environments, Wang *et al.* simulated a realistic scenario and studied the co-pyrolysis of waste edible oil (WEO) and LDPE together using HZSM-5 as a catalyst.[85] The strategy effectively promoted the formation of light aromatics, with the content of monocyclic aromatic hydrocarbons as high as 82.69% (LDPE/WEO = 1 : 3) and the

Table 2 Summary of microwave catalytic pyrolysis of plastic waste to MAHs

Feed	Catalyst	Reactor	Reaction conditions	MAH yield	Ref.
PS	USY	Batch	1 bar/He (650 °C)	65.1%	84
PS	HY	Batch	1 bar/He (650 °C)	58.1%	84
PS	Hβ	Batch	1 bar/He (650 °C)	40.6%	84
PS	ZSM-5	Batch	1 bar/He (650 °C)	22.7%	84
LDPE	MCM-41+USY	Continuous	1 bar/N_2 (450 °C)	49.9%	82
LDPE	MCM-41+USY	Batch	1 bar/N_2 (450 °C)	30.2%	82
LDPE+WEO	HZSM-5	Batch	1 bar/N_2 (450 °C)	47.5%	85
LDPE	HZSM-5	Batch	1 bar/N_2 (600 W)	28.12	49
LDPE+PET	HZSM-5/SiC	Batch	1 bar/N_2 (450 °C)	34.32	50
LDPE	Spent bleaching clay	Batch	1 bar/N_2 (450 °C)	54.92	86

BTEX compounds (benzene, toluene, ethylbenzene, and xylenes) up to 65.96%.

Overall, the production process of upgrading plastic waste into MAHs *via* microwave catalysis is strongly influenced by the interaction between catalyst properties and feedstock characteristics. The acidity, including the density and strength of acid sites, plays a decisive role in driving cracking, cyclization, and aromatization reactions. Medium acidity is beneficial for MAH formation while avoiding over-condensation. More acid sites could boost the cracking and dehydrogenation process. Hence, rational tuning of the acidity of the studied catalysts is of high importance. Pore structure could affect polymer chain accessibility and selectivity during the pyrolysis process. Smaller pores could provide spatial confinement and boost the cyclization and aromatization process, while larger pores could facilitate the cracking of long-chain molecules accordingly. In addition, the type of plastic feedstock significantly influences the product distribution.

3.3 Liquid fuel production

The catalytic conversion of plastic waste to liquid fuels, like gasoline, aviation fuel, or naphtha, is one of the most promising upcycling pathways.[87] In terms of sustainable fuel production, the conversion of plastic waste to gasoline is one viable alternative that can reduce CO_2 emissions compared to the conventional refinery route from crude oil.[88,89] Similarly, naphtha is generally a flammable liquid that is composed of the same carbon range (C_5–C_{12}) as gasoline and mainly paraffins and naphthene.[90] It's important for olefin production and fuel blending, because it could also serve as an effective high-octane gasoline component. Notably, the high paraffin content of naphtha is valued as it could serve as a feedstock for plastic production, which could reduce dependence on petroleum.[91]

As for naphtha production from plastic waste, Ruan's team has conducted extensive research and made significant progress. They first used high-silica ZSM-5 zeolite for HDPE microwave catalytic pyrolysis.[92] Results show that ZSM-5 with a high SiO_2/Al_2O_3 ratio (SiO_2/Al_2O_3 = 800) can effectively reduce the aromatic selectivity (28.5%) with high selectivity to C_5–C_{12} hydrocarbons (98.9%). After 8 successive cycle tests, the change in product distribution is minimal, exhibiting the potential for continuous production of low-aromatic liquid fuel. To further enhance naphtha production, they employed hierarchically structured ZSM-5 with micro-meso-macropores as a catalyst for microwave-assisted pyrolysis of plastic waste.[93] Compared to conventional ZSM-5, the hierarchical ZSM-5 possesses a larger average pore size with enhanced absorption capabilities, corresponding to enhanced diffusion capabilities. Besides the increase of aromatic selectivity from 6% to 73%, the enriched Brönsted acid concentration and hierarchical pore structure significantly elongated the catalyst's lifetime by more than 46 times. In addition, they also explored other materials and catalysis strategies to produce naphtha, especially with a low content of aromatics. Ruan *et al.* built up a coupled system with microwave catalytic pyrolysis and a tandem catalysis process for high-efficiency plastic waste upcycling, in which the first stage is for cracking waxes from plastic pyrolysis into short-chain hydrocarbons, and the second stage is for converting these unsaturated hydrocarbons into C_5–C_{12} paraffins by hydrogenation.[94] Firstly, they studied the product distribution over different porous materials, including SBA-15 (ordered mesoporous material) and zeolites (ZSM-5 and USY) for plastic pyrolysis. Although zeolites showed excellent cracking performance (>90% C_5–C_{12} hydrocarbons), the main components in the liquid oil were aromatic hydrocarbons, which are not desirable for cracking naphtha to plastic monomers. Next, by further screening loaded metals (Al/Zn/Ni/Zr/Rh/Pt/Pd/Fe/Mg/Mn/*etc.*…), they found that Zn/SBA-15 is the most effective catalyst for cracking HDPE into short-chain olefins with minimal aromatic selectivity (6.66% C_5–C_{12} aromatics). By comparison, Al/SBA-15 also exhibits higher cracking capability for HDPE decomposition, whereas the aromatic selectivity for this catalyst was also high, accounting for 45.9%. It could be due to the formation of proton acids that are bound to the silicon–aluminum bridge, resulting in high aromatic selectivity. Finally, by coupling with a hydrogenation reaction under microwave, the total paraffin selectivity in the oil products reached 50–71%, exhibiting applicability in real-world applications. Most recently, Vlachos *et al.* investigated the reactivity of Ni/BEA (beta zeolite) catalysts for polyethylene (PE) cracking into naphtha under microwave irradiation.[91] Their findings further contribute to the design framework of catalysts aimed at naphtha production. By tuning the acidity and metal loading ratio (MAB, metal–acid balance, equal to $n_{\mathrm{Ni}}/n_{\mathrm{BAS}}$), they found that metallic and acid sites must co-exist to facilitate hydrocracking, and the yield of naphtha increases with the MAB ratio. In addition, they found that the higher pore efficiencies of BEA(38) (Beta zeolite with SiO_2/Al_2O_3 of 38) enhance the reactivity by improving the internal mass transfer limitation. These findings further provide strong support for the following catalyst development to achieve more efficient naphtha production.

The effective microwave catalytic conversion of plastic waste to gasoline oil has also been receiving a lot of attention. Recently, Zhou *et al.* used ZSM-5 as a catalyst to investigate the pyrolysis behavior of linear LDPE in a continuously stirred microwave-assisted reactor.[59] The continuous system achieved 98.0% yield of gasoline-range products, with a mono-aromatic selectivity of 72.3%. Notably, the potential net energy gain was estimated at 34.16 MJ kg^{-1}, suggesting that the continuous system could be energy self-sufficient. By further coating catalysts onto structured SiC supports, Ruan *et al.* successfully synthesized ZSM-5/SiC foam for the catalytic pyrolysis of waste plastics under microwave irradiation.[95] Generally, the structured catalyst exhibits a heat transfer rate 2–5 times faster, slightly higher mass transfer coefficients, and over a 90% reduction in pressure drop when compared to conventional packed pellet beds, which were suggested to enhance mass and heat transfer within the reactor, potentially improving the catalytic activity and stability.[96,97] Experimental results show that the structured catalyst exhibited a stable selectivity to gasoline-range aromatic hydrocarbons of above 22% for 6 h at a reaction temperature of 450 °C and WHSV of 40 h^{-1}, outperforming conventionally shaped ZSM-5 catalysts by up to 37 times. Furthermore, the catalyst exhibits superior stability, and the performance shows a slight decrease after 5 regeneration–reuse cycles. This work demonstrates the potential for promoting the practical application of microwave catalysis. Aside from using pure zeolite as a catalyst, they also adopted NiO and HY as catalysts for LDPE conversion to gasoline *via* microwave catalysis.[98] The introduction of Ni species can effectively reduce the C–C and C–H bond dissociation energy, facilitating the chain-breaking accordingly. By suppressing the normal aliphatic hydrocarbon yield but promoting the generation of aromatics, the octane number of oil products increased effectively. Besides, co-pyrolysis is found to be an effective strategy for plastic valorization to fuel products. For example, Wang *et al.* adopted the strategy of co-pyrolysis for corncob and PS using the composite catalyst SiC@HZSM-5@$CoFe_2O_4$ as a catalyst, which has high microwave absorptivity and superior reactivity.[51] Experimental results show that the catalyst promoted the synergistic effect and improved the selectivity to renewable gasoline-range aromatics (47.2 area%). The enhancement may originate from the oxygen radicals generated during biomass cracking and the lattice oxygen from $CoFe_2O_4$, both of which can facilitate the decomposition of PS. Similarly, with the high demand for carbon neutralization in recent years, the selective conversion of plastic waste to sustainable aviation fuel is proposed to overcome the high CO_2 emissions by conventional aviation fuel.[99–101] To investigate the feasibility of the conversion of plastic waste to aviation oil, Zhang *et al.* focused on microwave catalytic pyrolysis of five kinds of plastic wastes (polypropylene, polystyrene, high-density polyethylene, low-density polyethylene, and polycarbonate) for cleaner aviation oil production under microwave.[102] The yield reached 98.78 wt% when using polystyrene as a feedstock, and C_8–C_{16} hydrocarbons are the main products. Similarly, Zhang *et al.* studied iron-based catalysts for converting polystyrene to aviation fuel.[103] Their results show that the oil yield could reach 97.67% with abundant C_8–C_{16} hydrocarbons of 52.04–77.88 area% under microwave heating. Recently, they compared the performance of microwave catalysis and conventional furnace heating for polypropylene converting to aviation fuel.[104] In the absence of a catalyst, microwave-assisted pyrolysis of PP produced the maximum oil yield at 70.8 wt%, whereas no oil was generated during conventional thermal pyrolysis. It could be due to the generation of local hotspots by microwave radiation that create high-temperature microenvironments, facilitating the cleavage of chemical bonds and the formation of free radicals, which were critical to molecular decomposition and transformation. The addition of catalyst FeNi/HY further increased the formation of naphthene, promoting the production of high-quality aviation fuel to 82.79 area% within the aviation fuel range.

Similar to the MAH formation pathway, the efficiency and product distribution of converting waste plastics into liquid fuels *via* microwave pyrolysis are primarily influenced by catalyst acidity, pore structure, and metal properties. The amount and strength of acid sites determine the efficiency of cracking, aromatization, and hydrocracking. Zeolites with a high Si/Al ratio or moderate acidity could decrease the selectivity to aromatics and increase the C_5–C_{12} paraffin/naphthenic hydrocarbon production. The pore structure could also affect the product distribution and long-term stability. Suitable pore size could serve the function of shape selection and benefit the production of gasoline range products. Hierarchical pore structure could improve mass diffusion and stability. Similarly, the introduction of active metal components could lower the activation energy barrier for C–C and C–H bond cleavage, effectively improving overall activity.

3.4 Other applications

Syngas production. Most of the studies on plastic degradation are operated under inert gas and use plastic waste as the only carbon and hydrogen source, which maximizes the potential of plastics but also brings significant stability problems due to the deactivation of the catalyst by coke deposition. To avoid this problem, converting plastic to syngas is one of the potential economic and alternative pathways. As an essential resource in the chemical industry, syngas serves as a versatile feedstock for producing valuable chemicals, fuels, and synthetic materials. The current solutions focus not only on catalysts but also on the entire pyrolysis process to achieve high-efficiency generation of syngas.[105–107] To achieve that, Liang *et al.* replaced the inert gas environment with a CO_2 atmosphere under microwave irradiation over $LaFeO_3$ catalysts for LDPE degradation.[108] The dehydrogenation of plastics was performed under microwave heating, and C–H bond selective scission took place due to the rapid activation of the catalysts. CO_2 was then activated and participated in the reaction through the Boudouard reaction. Compared to the low efficiency of conventional heating methods (carbon conversion lower than 10%, and CO_2 conversion lower than 1%), the carbon efficiency exceeds 60%, and CO_2 conversion reaches 41.3%. TGA results show that more than 93 wt% carbon is converted to CO effectively. Aside from the extremely high hydrogen extraction efficiency, the successive cycle test with no apparent decline further confirms the high stability in the long run. With the same strategy, Kwon *et al.* adopted Ni/Al_2O_3 catalysts for plastic bag waste pyrolysis, and they found that the existence of CO_2 could facilitate the cracking of HDPE and drive the conversion of long-chain hydrocarbons to shorter ones and boost the generation of massive carbon monoxide accordingly.[109] By simulating the turbine operation, the produced CO-rich syngas could help enhance the thermal efficiency of the gas turbine cycle significantly, demonstrating a more than two-fold increase compared to commercial natural gas. Besides taking CO_2 as an oxygen source, co-pyrolysis with biomass is another effective way to facilitate the production of syngas from plastic waste. Abedin *et al.* studied the catalytic effect of magnetite on microwave-assisted co-gasification of corn stover and plastic for syngas production using magnetite as a catalyst, which achieves a H_2 yield of 61 mmol $g_{feedstock}^{-1}$.[110] In addition, microwave catalysis effectively reduced the tar yield from 9% in the non-catalytic process to 2%.

Lubricant base oil precursors/aromatics production. The selective conversion of plastic waste to a series of chemicals or chemical precursors is also an important pathway. Tang *et al.* adopted microwave catalysis and used a type of hybrid Zn/ZnO as a catalyst for LDPE pyrolysis.[57] They found that the *in situ* formed Zn clusters could facilitate cleavage of the C–C bond, and near-complete upcycling (~100%) of landfilled plastic mixtures into lubricant base oil precursors and monomers is achieved. The high turnover number (250 $g_{plastic}$ $g_{catalyst}^{-1}$) for plastic depolymerization and excellent long-term stability over 50 successive cycle tests further demonstrate this strategy as a cost-effective and efficient approach for converting plastic waste into valuable products. Further, Pan *et al.* recently reported that the presence of CO_2 could facilitate the upcycling of polyolefin plastics to aromatics at 300 °C using the Pt/MnO_x-ZSM-5 catalyst. Aside from the 90% conversion of CO_2 to aromatic products when consuming 1 kg of polyethylene, this process could yield 0.63 kg aromatics, higher than 0.33 kg by the conventional pyrolysis process.[111] This strategy opens new avenues for plastic waste utilization while achieving effective CO_2 capture at the same time, and it provides great insight for microwave catalysis as well.

4 Techno-economic assessment

To further promote the application of microwave catalytic technology for plastic waste valorization in industrial scenarios, conducting an economic analysis of the route is necessary. Techno-economic analysis could help assess the economic viability of technology, primarily analyzing costs, benefits, and return on investment.[112,113] Recently, microwave catalytic technology for plastic waste upcycling has attracted tremendous attention due to its high energy efficiency, expansibility, and modular reactor design.[114] Taking microwave plastic pyrolysis to hydrogen gas and carbon nanotubes as an example (Fig. 7), microwave catalytic pyrolysis could achieve a net revenue of $577 per t plastic waste over 20 years with a high internal rate of return of 39% and a short payback period of 2.5 years; without catalysts, the direct microwave pyrolysis of plastic waste to H_2 and biochar or oil acquired the second highest net present value, but with a much lower internal rate of return (13%) and longer payback period (5.2 years). When it comes to the conventional pyrolysis process with catalysts, the net present value is negative because of limited high-value products, indicating the inviability of this route. Although the net present value could increase to positive with catalysts, the low internal rate of return (3%) and long payback period (13.7 years) reduce the possibilities of industrialization. Further sensitivity analysis indicates that carbon nanotube output and price are the primary impact factors of the net present value, followed by catalyst cost, hydrogen yield, and hydrogen price. For example, the calculation shows that a 20% increase in carbon nanotube production can increase the net present value to $30.45 million per year. Similarly, reducing catalyst cost could also enhance overall profit, reflecting the importance of catalyst stability and cost. However, comprehensive technical and economic analysis and life cycle assessment data for these systems are still limited, hindering broader comparison and industrial evaluation. In summary, the high economic profit, especially compared to the conventional heating strategy, effectively verifies the significant industrial potential of microwave catalytic pyrolysis for plastic upcycling.

5 Current reactor design

Microwave catalytic pyrolysis is mainly limited to lab-scale reactors, and thereby, the reported reactors are relatively

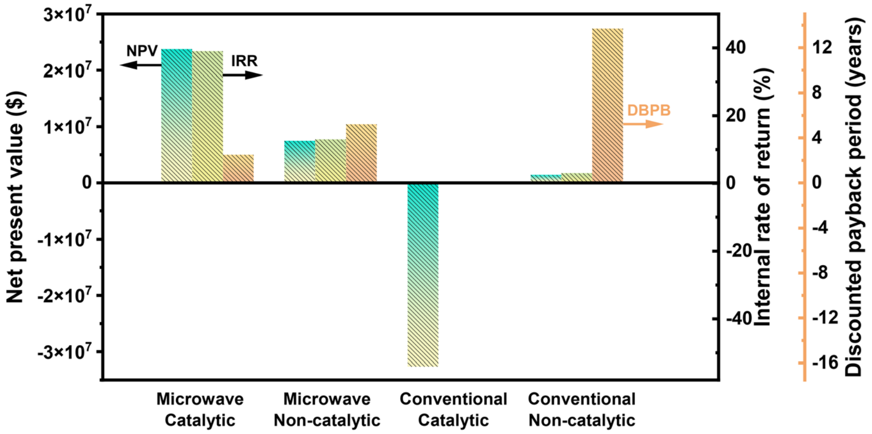

Fig. 7 The techno-economic assessment of microwave catalytic pyrolysis of plastic waste to hydrogen and carbon nanotubes.

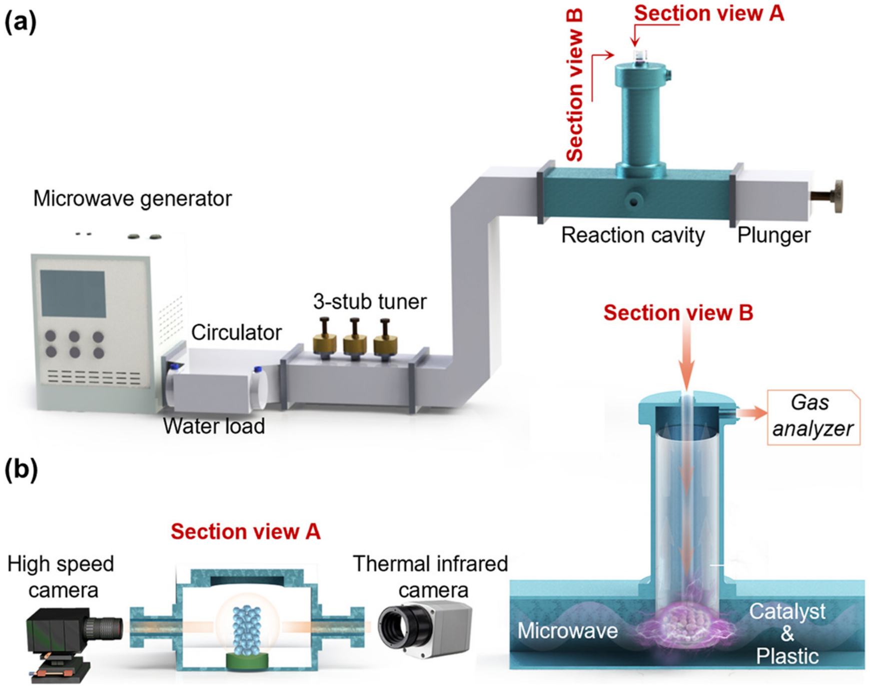

Fig. 8 Typical microwave reactor for plastic pyrolysis: a) device overview and b) cross-section view.

simple. As shown in Fig. 8, a typical single-model microwave reactor includes a microwave generator, reactor (reaction zone), temperature sensors (thermocouple or infrared thermometer for temperature measuring), and may include gas condensers (gas–liquid separation), cold traps (trap remaining liquid in gas), and liquid collectors (liquid products collection). Gas analyzers may also be installed for online gas analysis. At present, most reported work performed in microwave reactors is in the form of semi-continuous (batch) processes.[49,81,93,115] The low plastic and catalyst feed make it suitable for rapid catalyst screening. Recently, Fan *et al.* reported a continuous microwave reactor for plastic waste pyrolysis and compared the catalytic performance with that of a batch reactor system.[59] They found that more condensed liquid products are produced when using the continuous-stirred microwave pyrolysis (CSMP), and a positive potential net energy (~9.85 MJ kg^{-1}) is gained without catalysts. Under the same reaction conditions, a negative net energy is gained (~−42.86 MJ kg^{-1}) when using batch microwave pyrolysis (BMP). Then, the net energy gain of the continuous system after adding catalysts significantly increased to ~34.16 MJ kg^{-1}, while the net energy gain of the batch system remained negative (~−38.86 MJ kg^{-1}). It could be that the continuous system exhibits more uniform heat transfer and microwave distribution, hence gasoline product distribution is more concentrated. Zhou *et al.* developed a larger-scale continuous microwave-assisted pyrolysis system (CMAP). The system has a processing capability of 10 kg h^{-1} for plastic waste, and the overall energy efficiency is 90% with the total microwave power output of 9 kW.[116] They found that when using ZSM-5 as a catalyst, the yield of liquid products reaches 48.9%, in which the gasoline-fraction hydrocarbon ratio is approximately 73.5%. Therefore, the plastic microwave catalytic pyrolysis is of high promise for further scaling-up and industrialization.

6 Challenges

Although the application of microwave technology in plastic waste valorization exhibits huge potential for plastic waste treatment, including converting to H_2, carbon nanotubes, MAHs, and fuel, many key challenges remain unovercome that need to be addressed. Through analysis of recently reported work, some major limitations in microwave catalytic pyrolysis of plastic waste are proposed, including mechanism study, realistic plastic waste treatment, and reaction stability.

6.1 Mechanistic study

Generally, an in-depth understanding of reaction mechanisms is critical for guiding catalyst development. However, it's challenging for microwave catalysis of plastic waste, because of the long chain of plastic waste, tremendous intermediates and products, physical phase transitions of plastic waste, and chemical state change of catalysts. To track the evolution of plastic waste, high-frequency mass spectrometry could be an important tool to help analyze the product distribution and even capture the intermediates. It has been reported that time-of-flight mass spectrometry (TOF-MS) can enable soft ionization with minimal analyte fragmentation by coupling with proton transfer reaction ionization. The high mass resolution and sub-second response times enable this technique to effectively detect a wide range of compounds (mass range 0–500), making it a powerful tool for uncovering the underlying mechanism during microwave catalytic pyrolysis of plastic waste.[117] Furthermore, NMR (nuclear magnetic resonance), which could detect the resonance of atomic nuclei (such as ^{1}H and ^{13}C), has been increasingly applied in product analysis related to plastic upcycling. For instance, Ma *et al.* recently reported the successful application of in-line NMR for plastic separation and treatment monitoring process.[118] This achievement boosts the utilization of mixed plastics as feedstocks for treatment and shows huge potential. Ragauskas *et al.* adopted *in situ* NMR to trace the chemical structural changes of pyrolysis oil components during the accelerated plastic aging process to clarify the aging mechanism.[119] As for the catalyst physicochemical property characterization, microwave irradiation and surface property evolution are important for understanding the underlying reaction mechanism, which is also challenging. It's worth noting that the change in crystal phase and morphology with microwave irradiation may also change the active sites. Hence, to further distinguish the real active sites and clarify the interaction between microwave and the catalyst, *in situ* XRD or XAS can help understand the structure evolution during the reaction process. Furthermore, the surface chemical state transformation could be captured by NAP-XPS. Collectively, the detection of intermediates during the reaction and recognition of the surface-active sites could effectively help understand the catalytic conversion mechanism of plastic decomposition under a microwave environment.

6.2 Realistic plastic waste treatment

In most reported studies, the plastic feedstock used is typically single-type or simple plastic, which significantly differs from real-world plastic waste. This complexity discrepancy poses a major challenge when attempting to translate laboratory results into practical, large-scale applications. Massive inorganic or organic additives, colorants, and fillers make the practical microwave pyrolysis a much more complex process, which is an unavoidable problem in industrialization.[120] Common additives like $CaCO_3$ have been reported to reduce the yield of MAHs during the PP pyrolysis process by promoting cross-linking reactions that form macromolecules.[121] Furthermore, the existence of these additives may be detrimental to practical operation, such as the contamination of the target product, increasing separation cost, deactivating catalysts, and clogging the reactor in long-term operation.[122] As for heteroatoms, including N, S, P, Cl, and O, they may complicate the products' composition, reduce the value, and increase the pre-/post-treatment cost.[123] Therefore, from a practical perspective, more attention should be paid to the true composition of plastic waste. Additionally, the physicochemical properties of the various components should be carefully considered to optimize reactor design and process configurations to mitigate the adverse effects of additives or to exploit their potential for promoting plastic conversion.

6.3 Reaction stability

To achieve the effective upcycling of plastic waste to value-added products, stability is one of the non-negligible problems that needs to be overcome. Not to mention the impurity from practical plastic waste, like additives, contaminants, water, *etc.*, the catalysis process itself faces severe stability problems, like coke deposition, metal aggregation, and phase transition. For instance, during the conversion of plastic waste to H_2 and carbon nanotubes, massive carbon deposition over catalysts decreases the exposure of active sites, which would limit their capability of plastic pyrolysis in long-term operation.[26,124] Similarly, in the pyrolysis process of MAH production, the deactivation of acid sites and clogging of pores would accelerate catalyst deactivation.[50,125] Although addressing such a practical challenge is essential for verifying the viability of technology, relevant studies about the long-term stability and repeatability of catalysts are still limited. Therefore, establishing a standard benchmark for catalyst performance and stability evaluation is highly desirable, including using realistic plastics with specific compositions and concentrations to evaluate catalyst efficiency and stability, and conducting experiments under various conditions, especially harsh ones.

7 Conclusion and outlook

Microwave catalysis for plastic waste pyrolysis has recently attracted tremendous attention because of its high efficiency and rapid-heating features. This review summarizes recent progress in the microwave catalytic pyrolysis of plastic waste from the perspectives of catalyst design and reaction engineering, aiming to help promote its more extensive application and scaling-up. At present, multiple microwave catalytic pathways have been developed, including plastic waste converting to H_2 and carbon, MAHs, liquid fuels, syngas, *etc.*, and observable progress is obtained with higher yield and selectivity. The catalyst design, including composition, acidity, defects, microwave absorptivity, and morphology, has been tailored to enhance the overall catalytic performance. By tidying the relationship between catalyst properties and performance, this work could help deepen the understanding of the microwave catalytic process and provide clearer insights for catalyst development. Despite its advantages and potential, microwave catalytic pyrolysis for plastic waste valorization still faces several challenges before industrialization: for example, due to the macromolecular structure of plastic waste and tremendous intermediates during reaction, the in-depth mechanism study becomes quite difficult, further hindering the understanding of the reaction process at a molecular level and the following precise design of catalysts. Considering the catalysts would be applied in treating realistic plastic waste, more focus should be on the effects of additives, packaging, and contaminants to pave the road for industrialization; similarly, long-term stability requires attention as well, which is also an unavoidable challenge in the industrialization process. With the development of characterization techniques, the understanding of the underlying mechanisms, and the building of a standardized benchmark, microwave catalytic pyrolysis could be an ever-powerful technique for plastic waste upcycling. Further, scaling up of the microwave reactor to achieve higher treatment capability is also of high importance for its industrialization. Finally, we hope this review could offer researchers a systematic knowledge of the application of microwave catalysis in the treatment of plastic waste and boost its development and application in the near future.

Conflicts of interest

There are no conflicts to declare.

Abbreviations

Al_2O_3	Aluminum oxide
Fe	Iron
Co	Cobalt
Ni	Nickel
MgO	Magnesium oxide
ZnO	Zinc oxide
Zn	Zinc
SiC	Silicon carbide
SiO_2	Silicon dioxide
NiO	Nickel oxide
$FeAlO_x$	Iron–aluminum oxide
Co_3O_4	Cobalt oxide
Fe_2O_3	Iron oxide
Pr	Praseodymium
Ce	Cerium
La	Lanthanum
Yb	Ytterbium
N	Nitrogen
S	Sulfur
P	Phosphorus
Cl	Chlorine
O	Oxygen
H	Hydrogen
C	Carbon
$CaCO_3$	Calcium carbonate
BEA	Beta zeolite
MAB	Metal-acid balance
PE	Polyethylene
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinyl chloride
PET	Polyethylene terephthalate
C–C	Carbon–carbon bond
C–H	Carbon–hydrogen bond
CO	Carbon monoxide
CO_2	Carbon dioxide
Ar	Argon
N_2	Nitrogen
CNT	Carbon nanotube
MWCNT	Multi-wall carbon nanotube
MAH	Monocyclic aromatic hydrocarbon
PAH	Polycyclic aromatic hydrocarbons
BTEX	Benzene, toluene, and xylenes
HDPE	High-density polyethylene
LDPE	Low-density polyethylene
WHSV	Weight hourly space velocity
CMAP	Continuous microwave-assisted pyrolysis system
NMR	Nuclear magnetic resonance
XAS	X-Ray absorption spectroscopy
BMP	Batch microwave pyrolysis
CSMP	Continuous-stirred microwave pyrolysis
TOF-MS	Time-of-flight mass spectrometry
XRD	X-ray diffraction
BAS	Brønsted acid site

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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