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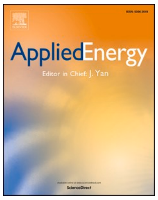

Recent advances in plastic waste pyrolysis for liquid fuel production: Critical factors and machine learning applications

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HIGHLIGHTS

- Conversion of plastic waste into biofuel by pyrolysis was compressively reviewed.
- Effects of conditions, catalyst, plastic, and reactor type on biofuel were discussed.
- The state-of-the-art of machine learning in plastic pyrolysis was reviewed.
- A critical discussion of recent challenges with future perspectives was presented.

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ABSTRACT

It is urgent to promote the technology development of plastic waste (PW) valorization to mitigate the present serious plastic pollution. Pyrolysis has been widely used to convert PW into high-value-added products, especially liquid fuels, in a sustainable manner. However, achieving a high yield of liquid fuel with good quality remains challenging due to the difficulty in optimizing pyrolysis process and synthesizing outstanding catalysts to narrow the product distribution. This work comprehensively reviewed PW pyrolysis from both technical and computational (machine learning modeling) aspects, with a critical discussion of recent challenges to find new insights for improving the conversion efficiency and promoting commercialization. Results indicated that the impacts of various factors, including PW type, process condition, catalyst, and reactor type, on the PW pyrolysis were extensively investigated by the research community. Machine learning methods have also been frequently applied to predict, interpret, and optimize the PW pyrolysis. However, more efforts can be made in the future regarding catalyst synthesis, selection of co-pyrolysis additives, mechanism of catalyst deactivation, and design of renewable energy supply system for PW pyrolysis plants. Additionally, more attention should be paid to enlarging the data size, improving model interpretability, and exploring innovative ways of machine learning application (e.g., active learning) in process optimization and catalyst design for PW pyrolysis.

1. Introduction

Nowadays, plastic pollution has become a significant global environmental challenge due to the high consumption and weak biodegradability of plastics. The global plastics consumption has exceeded 380 million tons and continues to rise annually [1]. The outbreak of Covid-19 has further exacerbated this issue, generating massive plastic waste (PW) from the consumption of medical plastics, masks, and takeaway boxes. In 2016, nearly 23 million tons of PW entered aquatic ecosystems (e.g., rivers and oceans) [2], posing a severe threat to the growth of aquatic organisms (Fig. 1). Researchers cataloged more than 12 million pieces of trash in and around rivers, oceans, coastlines, and seabeds, which suggested that more than 40% of the plastic was associated with takeaway food and beverages [3]. Ultimately, plastics may

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spread and accumulate through the food chain in the form of microplastics, posing a potential threat to human health.

On the other hand, plastic-related emission has accounted for 4.5% of global greenhouse gas emissions [4]. The production of 1 ton of plastics from using traditional fossil resources will generate about 5 tons of equivalent carbon emissions [5], while recycling 1 ton of plastics by chemical and physical methods generate only 1.0 and 1.5 tons of equivalent carbon emissions, respectively [6]. But so far, only 21% of PW has been recycled or incinerated worldwide [7], and the recycling rate in China is only about 30% [8], since most of them are still treated by landfilling which would lead to waste of resource and greenhouse gases emissions [9-11]. To control plastic pollution and encourage the high value-added utilization of PW, new policies have been introduced by the Development and Reform Commission and the Ministry of Ecology and Environment of China. Recycling and converting PW into renewable energy and sustainable resources can not only aid the control of plastic pollution but also substitute some fossil fuels to reduce carbon emissions, which is beneficial to achieving the goal of “double carbon” proposed by China. Different conversion technologies for PW valorization have been developed, including thermal [12], microbe/enzyme-aided [13-15], and mild catalyst-aided conversions [16]. Thermal conversion can efficiently deconstruct any kind of PW (unsorted and contaminated ones) with multiple products generated, such as carbon material, bio-oil [17], and syngas [18,19]. Although biological conversion is cost-saving, the conversion efficiency is too slow to handle the large generation of PW. Mild catalyst-aided conversions, such as

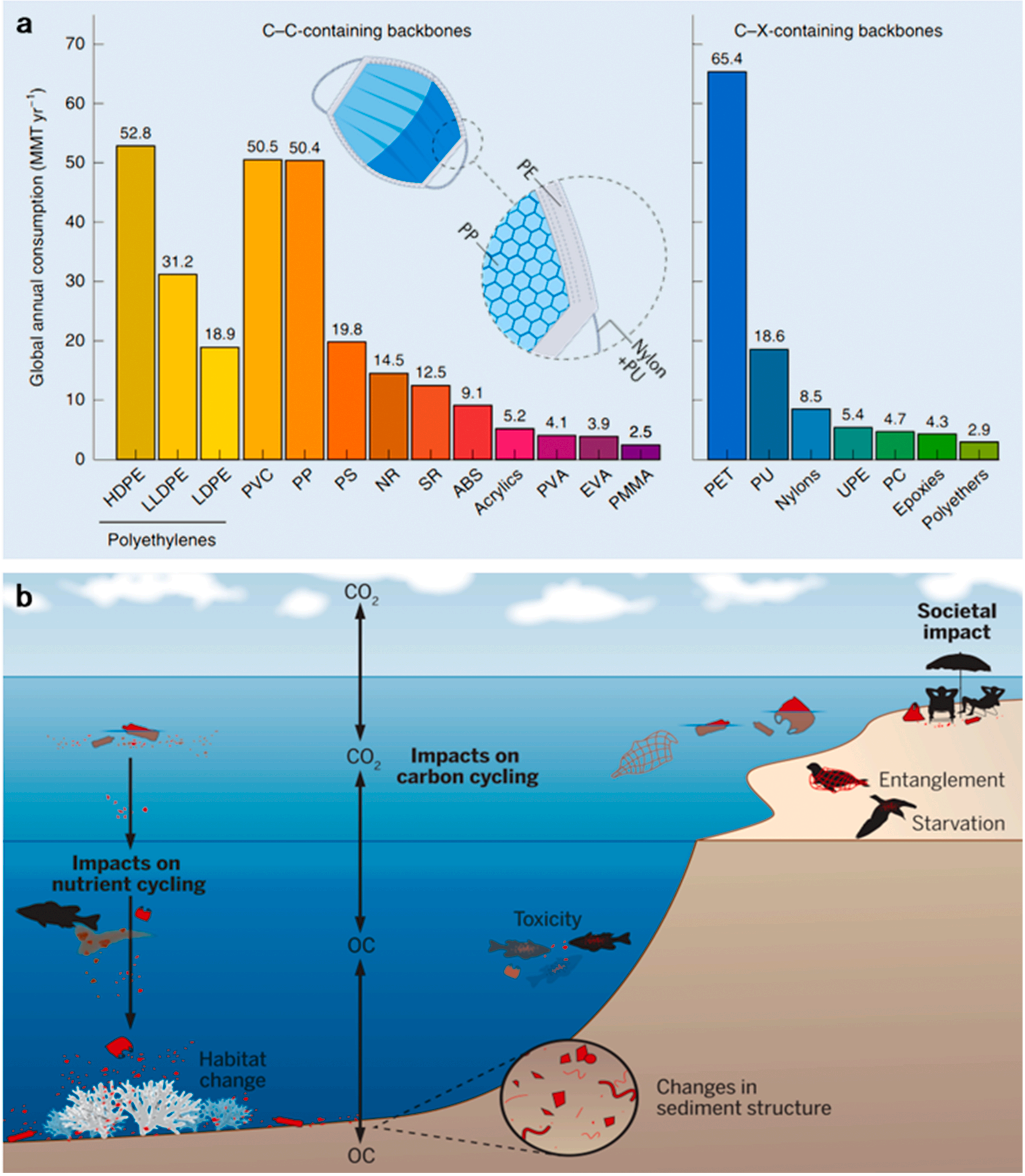

Fig. 1. (a) Global annual consumption amounts of different plastics [1] and (b) the impact of plastic waste on the marine ecosystem [35].

hydrogenolysis [20], photocatalysis [21], and electrocatalysis [22,23], could produce high-quality chemicals. However, developing outstanding catalysts for these processes requires significant investment in both labor and capital. Comparatively, thermal conversion is the most promising approach to be scaled up and commercialized for PW utilization due to its strong adaption to PW type and high conversion efficiency. Among thermal conversion pathways, pyrolysis is the most widely used one since it can produce a range of high-value-added fuels and chemicals by controlling the pyrolysis conditions [24,25]. Specifically, a high yield of liquid oil (up to 80 wt%) could be achieved in PW pyrolysis under a moderate temperature of about 500 °C [25].

Due to the merit of pyrolysis for PW utilization, significant efforts have been made to further upgrade this technology. Although pyrolysis is flexible to produce various products by manipulating and optimizing the process conditions, intensive labor, time, and capital are essential to design and conduct the intensive investigation [26]. To overcome this limitation, the research community has developed computational modeling approaches [27-29], such as machine learning (ML), computational fluid dynamics (CFD), and Aspen simulation, to explore pyrolysis and optimize process conditions for promoting its scale-up application. Recently, ML methods have gained more and more attention due to their advances in computational speed and robust adaption in different research domains [28,30,31]. ML, a data-driven approach, can learn from complex, highly dimensional, and large historical data to generate predictive models [32,33]. This means there is a strong flexibility in feature selection, allowing us to consider as many factors related to pyrolysis as possible to develop ML models [34]. This unique advantage of ML modeling compared to other computational approaches has led to a surge of interest in using ML methods. As a result, domain researchers have reported numerous studies on the ML modeling of PW pyrolysis.

To understand the recent advances in PW pyrolysis and corresponding ML applications, we conducted a systematic literature review from both technical and computational perspectives. This review is the first of its kind to report such a comprehensive review related to PW pyrolysis and ML application. It should be noted that the yield and quality of products from PW pyrolysis are highly dependent on several factors, including the feedstock type, pyrolysis reactor, catalyst, and pyrolysis conditions. All of these factors can also be considered in the ML modeling process to develop predictive models for specific purposes, such as predicting the distribution of three-phase products and pyrolysis kinetic parameters, characterizing and evaluating pyrolysis oil during production and application. Moreover, the ML models for PW pyrolysis are also applied to interpret the importance and correlations of input factors on the predicted targets and combine them with optimization algorithms to optimize the pyrolysis conditions for producing desired products by identifying individual objective functions. In general, this review focuses on PW pyrolysis from both technical and computational perspectives. Section 2 summarizes and discusses the technical aspect of PW pyrolysis with the consideration of various factors involved. The third section provides a detailed review of ML applications in PW pyrolysis based on different modeling purposes. Section 4 presents the current challenges and limitations on both the technical aspects and ML modeling of PW pyrolysis with proposed future directions.

2. Prominent factors in plastic waste pyrolysis

Pyrolysis is a thermal conversion in which the PW is heated and decomposed under inert gas or oxygen-deficient conditions [36]. It is considered one of the most promising technologies for PW valorization. The process produces three-phase products, i.e., liquid oil, char, and combustible gas. Among these products, bio-oil is regarded as the most valuable one due to its high yield and rich contents of various chemicals. High-quality bio-oil could be directly used as renewable energy to replace traditional fossil fuels and mitigate the greenhouse gas emissions for carbon neutrality [37]. To achieve a high yield of bio-oil with good quality, it is crucial to understand the impacts of the factors involved in the PW pyrolysis on bio-oil production. Therefore, a systematic literature review was conducted based on the factors, including types of feedstocks (types of plastics and other additives), pyrolysis conditions, catalysts applied in pyrolysis, and types of pyrolysis reactors (Fig. 2), to understand their impacts on the distribution of three-phase products and the components in bio-oil from PW pyrolysis, as shown in Table 1.

2.1. Effect of feedstock

Pyrolysis has proven to be capable of utilizing different types of PW as feedstock for renewable energy production [25]. The decomposition process, distribution, and compositions of products from PW pyrolysis vary depending on PW types being processed. Cepeliogullar and Putun investigated the thermal behaviors of both polyethylene terephthalate (PET) and polyvinyl chloride (PVC) plastics during pyrolysis [38]. They found that the maximum weight loss for PET and PVC occurred at around 400 °C and 285 °C, respectively. Moreover, fixed bed pyrolysis experiments indicated that the liquid/gas yield from pyrolysis of PET and PVC was 23.1 wt%/ 76.9 wt% and 12.3 wt%/ 87.7 wt%, respectively, under the conditions of 500 °C with a heating rate of 10 °C min^{-1}. Ahmad et al. studied the pyrolysis of polypropylene (PP) and high density polyethylene (HDPE) in a steel microreactor and found that a PP conversion of 98.66 wt% was achieved at 300 °C with 69.82 wt% liquid and 28.84 wt% gas products [39]. Similarly, a high conversion efficiency (98.12 wt%) of HDPE was obtained at 350 °C with 80.88 wt% liquid and 17.24 wt% gas products. Moreover, the analysis of the liquid fractions showed that naphtha range hydrocarbons were enriched from both PP and HDPE pyrolysis; however, the high proportion compounds in PP-derived liquid were C13-C16 and C6-C12 for HDPE. It also should be noted that the detailed chemical products of liquid fraction from pyrolysis of polyolefin and other plastics were different. For polyolefin plastic, the main components of liquid products were paraffin, olefins, and naphthene [39]. In the case of other plastics, such as PET, it was reported that half of the liquid product was benzoic acid which was unfavorable due to its corrosiveness [38].

Based on the above literature review, it was found that the type of PW had significant impacts on the decomposition process, distribution, and compositions of products from pyrolysis. These impacts were dominated by the different structures and compositions of plastics. For example, the possible reason why the temperature for the maximum weight loss of PET was higher than that of PVC could be due to the higher bond dissociation enthalpy of PET compared to PVC (Fig. 3a) [38,40]. In addition, the volatile matter (VM) content from the proximate analysis of plastics plays an important role in the generation of liquid oil from pyrolysis, where a higher VM content contributes to a higher yield of liquid oil [19]. The oil yield from the PET pyrolysis was found to be only 23.1 wt% which is significantly lower compared to that obtained from the PP pyrolysis [38,39]. The lower liquid yield could be explained by the lower content of VM in PET, as shown in Fig. 3b. It should be noted that despite the high VM content (97 wt%) of PVC, the oil yield from its pyrolysis is quite low owing to the high content of Cl in PVC, which would lead to the formation of abundant HCl instead during pyrolysis [41]. The generation of HCl is a disaster during pyrolysis due to its highly corrosive and toxic properties. It was reported that HCl formation was one of the main reasons for the shutdown of a pyrolysis pilot plant in Ebenhausen, Germany [42].

Co-pyrolysis of PW with other biomass wastes has also been frequently studied to evaluate the possible synergies and determine effects of different components on the process. Xu et al. studied the co-pyrolysis of macroalgae (Enteromorpha prolifera) and HDPE in the presence of the HZSM-5 catalysis [43]. The results showed that acids, oxides, and nitrogen-containing compounds decreased while the aromatic hydrocarbons and light oil increased during co-pyrolysis. Akancha achieved the highest liquid oil yield of 80.5 wt% during the co-pyrolysis of waste PP and rice bran wax at 550 °C with a blend ratio of 1:3 [44].

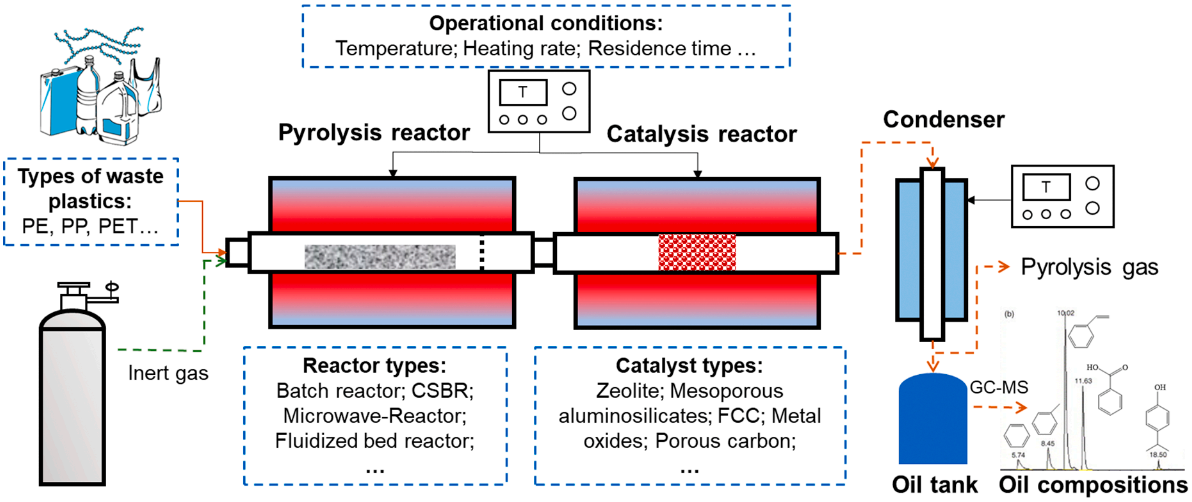

Fig.2. Basic flow and prominent factors of plastic waste pyrolysis.

The reduced oxygenates, higher aliphatic compounds, and fuel properties within the range of gasoline and diesel indicated good quality of the generated oil. Singh and Ruj studied the optimum pyrolysis temperature of the mixed PW (58.8 wt% PE (HDPE and low density polyethylene (LDPE), 26.9 wt% PP, 8.7 wt% polystyrene (PS), and 5.6 wt% PET) using thermogravimetric analysis (TGA) [45]. They found that the initial decomposition temperature decreased from 350 to 310 °C compared to individual plastic pyrolysis. López et al. further explored the impact of real packing waste composition on the characteristics of pyrolysis products at 500 °C for 30 min [46]. They stated that high paper content in feedstock would produce more water in the pyrolysis liquid and more CO and CO_2 in the gases; while higher PE film content could increase the viscosity of liquids. Moreover, the metal content in feedstock had catalytic effects to increase the proportion of aromatic hydrocarbons in pyrolysis oil.

2.2. Effect of pyrolysis conditions

Operation condition is another important factor in PW pyrolysis, with pyrolysis temperature, residence time, and heating rate being the three main parameters involved. Temperature is one of the most important factors in PW pyrolysis since it dominates the chain-cracking processes of plastic polymer. As the pyrolysis temperature increases, the vibration of molecules in plastics will become stronger to overcome the attraction of Van der Waals force. Once the vibration energy is higher than the enthalpy of the C–C bond in the chain, the carbon chain will be broken [47]. Gracida-Alvarez et al. investigated the impact of temperature on the products of HDPE micropyrolysis [48]. The decreased liquid yield from 62.7 to 48.8 wt% and increased gas product from 34.3 to 51.0 wt% were found as the temperature increased from 625 to 675 °C. Moreover, it was observed that the increase in temperature enhanced the degradation of diesel range oil (C11-C20) and waxes (C21-C31) with more aromatics and light aliphatics formation. For a two-step catalytic pyrolysis, the PW will be first decomposed, and then primary volatiles pass through a catalytic bed to be further reformed. The catalytic temperature is also a crucial parameter for catalyst selectivity and product distribution. Onwudili et al. found a gradual increase of C8-C16 straight-chain alkanes and monocyclic aromatics with the catalytic temperature from 430 to 571 °C during the pyrolysis-catalysis of a simulated mixed PW sample. They also obtained an increase of C1-C4 hydrocarbons when the catalyst bed temperature increased from 500 to 600 °C [49]. However, a further increase in catalytic temperature decreased the liquid yield due to its promotion of the secondary cracking of primary vapors.

Residence time is also an important factor in PW pyrolysis, although its effect is weaker than that of temperature. A long residence time can promote the secondary cracking of PW and result in a high gas yield in most cases. Miandad et al. investigated the role of residence time on PS pyrolysis and found that the gas yield increased constantly with the increase of residence time, while the yield of liquid oil remained unchanged when residence time further increased from 75 to 120 min [50]. They also emphasized the highest styrene yield (48 wt%) could be obtained when pyrolysis temperature and residence time were set at 450 °C and 75 min. Mastral et al. investigated the effect of residence time on HDPE pyrolysis at a two-stage micropyrolysis reactor [51]. The results suggested that the longer residence time tended to facilitate aromatics production. However, the effect of residence time was strongly limited comparing to the pyrolysis temperature and weakened further on liquid and gaseous yield when the temperature was over 685 °C.

Heating rate can make a difference to the pyrolysis-temperature pattern. For example, Cao et al. found a higher heating rate could delay the HCl primary release temperature from 270 °C at 10 °C/min to 370 °C at 60 °C/min during PVC pyrolysis [52]. Xu et al. found a higher heating rate could shift the initial, end, and peak temperatures to higher values during pyrolysis experiments in a thermal analyzer [53]. These phenomena could be explained by the large temperature difference between the furnace and the PW sample, which was attributed to the thermal lag and the limitations in heat transfer rate. On the other hand, the higher DTG peaks observed at higher heating rates indicated that the reaction rate would be increased by a higher heating rate, thereby speeding up the weight loss. Furthermore, the influence of heating rates on PP pyrolysis in a bench-scale reactor was found to be significant at high temperatures, where a fast-heating rate (180 °C/min) promoted the cracking reactions of volatiles released at 600 °C and formed more light compounds [54].

To sum up, the pyrolysis temperature plays the most crucial role in the C–C bond broken of PW, while residence time presents a significant effect on the product distribution and liquid composition at a considerably low pyrolysis temperature during PW pyrolysis. Higher temperature, longer residence time, and slower heating rate would enhance the secondary cracking of volatiles and promote the decomposition of polymers, resulting in a higher proportion of low-molecular-weight hydrocarbons in liquid oil and a higher gas yield. Moreover, the selectivity of aromatics is controlled by both the temperature and residence time, indicating that optimal products require careful selection in pyrolysis parameters. To achieve the maximum liquid oil recovery from PW pyrolysis, the pyrolysis temperature should be accurately controlled based on the PW type. A further elevation in reaction temperatures would result in the transformation of more volatiles into non-condensable gases.

Table 1
Summary of energy and resource production from pyrolysis of plastic waste.

Plastic types	Reactor types	Pyrolysis conditions			Catalyst (plastic to catalyst ratio)	Liquid yield (wt %)	Gas yield (wt%)	Solid yield (wt%)	Components in liquid	Ref.
		Temperature (°C)	Residence time (min)	Heating rate (°C/min)						
PET	Tandem μ-reactor	450;700 (catalysis)	20	10	ZnO (1:20)	/	/	/	83.9 wt% of aromatic compounds, 16.1 wt% of oxygenates	[63]
					MgO (1:20)				38.7 wt% of aromatics, 61.3 wt% of oxygenates	
					TiO_2 (1:20)				38.9 wt% of aromatics, 61.1 wt% of oxygenates	
					ZrO_2 (1:20)				43.9 wt% of aromatics, 56.1 wt% of oxygenates	
					CaO (1:20)				99.7 wt% of aromatics, 0.3 wt% of oxygenates	
PET	Cylindrical horizontal furnace	450	10; 0.3	45	Sulphated ZrO_2 (100:3)	48	35	17	27.5 wt% of benzoic acid, 13.9 wt% of other aromatic compounds	[84]
HDPE	Horizontal steel	350	30	20	/	80.88	17.24	1.88	59.70 wt% of paraffinic, 31.90 wt% olefinic, and 8.40 wt% of naphthenic hydrocarbons	[64]
HDPE	Semi-batch	450	25	/	FCC catalyst (10:1)	91.2	4.1	4.7	73.78 wt% of olefins, 14.95 wt% of paraffins, 5.84 wt% of aromatic compounds	[85]
HDPE	Fixed bed	600;600 (catalysis)	30	10	Ni/Mo/Fe/Ga/Ru/Co Y-zeolite (1:2)	31–43	29–43	14–24	Around 80–95 wt% of aromatics, 5–20 wt% of aliphatic hydrocarbons	[57]
HDPE	Fixed bed	500;500 (catalysis)	30	10	MCM-41: ZSM-5: HDPE = 1:1:1	83.15	16.85	/	97.72 wt% of gasoline range hydrocarbons, 95.85 wt% of aromatics	[59]
HDPE	Microwave-assisted reactor	500;450 (catalysis)	20	80–100	MgO (15:1)	37.00	60.85	2.15	36 wt% of C5-C12 aromatics, 38 wt% of C5-C12 alkenes, 11 wt% of C5-C12 alkanes	[86]
HDPE	Batch reactor	550	/	5	HZSM-5 (10:1)	17.3	72.6	10.1	6.7 wt% of paraffins, 42.7 wt% of olefins, 50.6 wt% of aromatics	[87]
					HUSY (10:1)	41.0	39.5	19.5	37.9 wt% of paraffins, 18.3 wt% of olefins, 43.8 wt% of aromatics	
PVC	Fixed bed	500	/	10		12.3	87.7	0	33.55% of naphthalene derives	[38]
LDPE	Batch reactor	550	/	5	HZSM-5 (10:1)	18.3	70.7	11	8.5 wt% of paraffins, 39.4 wt% of olefins, 52.1 wt% of aromatics	[87]
					HUSY (10:1)	61.6	34.5	3.9	34.8 wt% of paraffins, 33.9 wt% of olefins, 31.3 wt% of aromatics	
PP	Tube furnace	600	60	10	Kaolin clay (9:1)	17.8	64.8	17.4	30–40 wt% of aromatics, 25–35 wt% of straight alkenes, 5–10 wt % of cycloparaffins, 2–7 wt% of straight alkanes	[88]
PP	Tubular reactor	600	60	15	/	60.5	31.8	2.5	31.0 wt% of heavy fractions and 29.5 wt% of light fractions	[54]
PP	Auger reactor and fluidized bed reactor	720	/	/	/	18.48	81.10	0.17	0.9% aliphatics, 10.12% aromatics and 7.46% unknowns	[89]
PS	Fixed bed	450	75	/	Natural zeolite	54	12.8	33.2	91 wt% of aromatics, mostly ethylbenzene (60.8 wt%)	[90]
PS	Batch reactor	450	120	/	ZnO (5:1)	96.73	/	/	2.47 wt% toluene, 1.16 wt% ethylbenzene, 47.96 wt% styrene monomer and 1.90 wt% α-methylstyrenes	[91]
PE	Fixed bed	400–420	180–240		/	65	26	9	13.1% of hydrogen with a higher heating value of 45.31 Mj/kg	[92]
PP, PE, and PET	Batch reactor	550	174	/	/	86.04	8.11	5.85	94.8% non-aromatic products, 5.2 aromatic product	[93]
			128	/	HZSM-5 (9:1)	82.53	10.72	6.75	80.5% non-aromatic products, 19.5 aromatic product	
			146	/	/	84.24	9.01	6.75	92.6% non-aromatic products, 7.4 aromatic product	
			121	/	HZSM-5 (9:1)	71.63	19.82	8.55	94.8% non-aromatic products, 5.2 aromatic product	
PE, PP, and PS	Tubular furnace	550; 600 (catalysis)	40	20	Sewage sludge derived char (8:5)	41.3	48.8	9.9	47.4 wt% of Monocyclic aromatics, 24.5 wt% of PHAs, and 1.3 wt% of alkanes	[68]
PE, PP, and PS	Tubular furnace	550; 600 (catalysis)	40	20	Quartz sand (8:5)	46.6	51.0	2.4	48.6 wt% of Monocyclic aromatics, 19.8 wt% of PHAs, and 4.8 wt% of alkanes	[68]

(*continued on next page*)

Table 1 (*continued*)

Plastic types	Reactor types	Pyrolysis conditions			Catalyst (plastic to catalyst ratio)	Liquid yield (wt %)	Gas yield (wt%)	Solid yield (wt%)	Components in liquid	Ref.
		Temperature (°C)	Residence time (min)	Heating rate (°C/ min)						
PS and PP	Microwave oven	900 W	10	/	Activated carbon (10:1)	84.30	15.7	0	80.58 wt% of aromatic content (toluene (3.39 wt%), styrene (67.58 wt%)	[94]
LDPE	Microwave oven	500	/	/	NiO (in-situ) and HY (ex-situ)	56.5	41.8	1.7	93.8% of gasoline fraction with 6.1% fractions of C_{13+}	[95]

HDPE: High Density Polyethylene, LDPE: Low Density Polyethylene, PE: Polyethylene, PP: polypropylene, PS: polystyrene, PVC: polyvinyl chloride, PET: polyethylene terephthalate, PLA: polylactic acid.

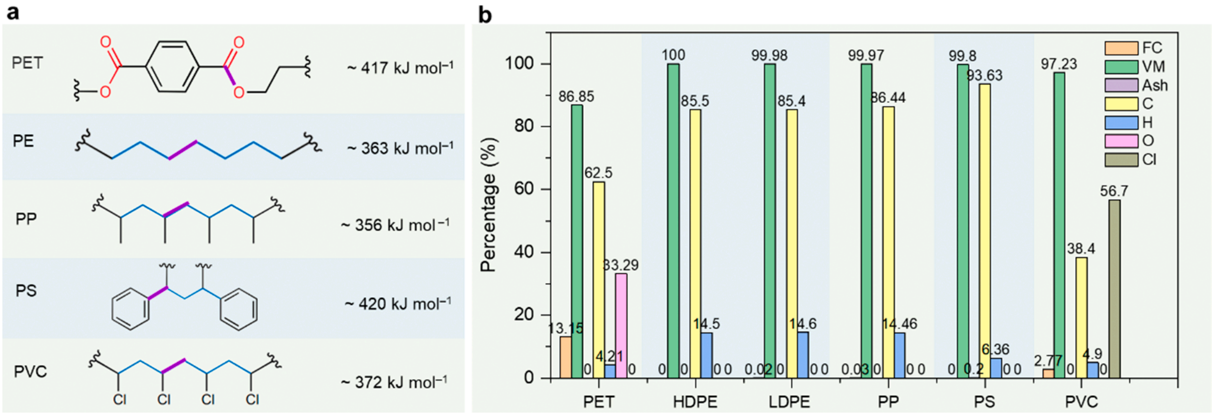

Fig. 3. The basic structure with bond dissociation enthalpies (a) and proximate and elemental analysis (b) of different types of plastics [19,40].

2.3. *Effect of catalyst in pyrolysis*

Catalytic pyrolysis can promote the rearrangement of randomly broken polymers into specific products, resulting in narrower product distributions. However, achieving a targeted conversion efficiency of PW into desirable products remains challenging, even with the adjustment of pyrolysis parameters to obtain a higher liquid oil yield with a high proportion of light fraction. Recently, various catalysts have been developed to narrow product distribution and improve product selectivity, and their catalytic effects are mainly dominated by acidity and pore structure [55]. Specifically, the addition of catalysts can reduce the energy demand for breaking C—C linkage, decreasing the decomposition temperature of PW in pyrolysis and accelerating the heat transfer rate to further increase the reaction rate [19].

Zeolites are widely investigated in PW pyrolysis due to their strong acidic properties, unique pore structure, and selectivity. The catalytic activity of zeolites is highly dependent on the SiO_2/Al_2O_3 ratio which dominates the acidic strength. Artetxe et al. studied the impact of the SiO_2/Al_2O_3 ratio of HZSM-5 zeolite on the product composition during HDPE pyrolysis [56]. The results showed that reducing SiO_2/Al_2O_3 from 280 to 30 increased the yield of light olefins and aromatics while decreasing the yield of heavy fractions of C12–C20. Moreover, zeolite catalysts have demonstrated high selectivity towards aromatics. Akubo et al. found that the aromatic proportion, especially mono-cyclic aromatics, exceeded 80% in the liquid oil from HDPE pyrolysis in a two-stage fixed bed with the catalysis of metal-loaded Y-zeolites, while the aliphatic hydrocarbons occupied over 99% in the non-catalytic process [57].

Mesoporous aluminosilicates are also suitable catalysts for PW pyrolysis due to their pores with a size of 1.5–30 nm which allows the direct entrance of pyrolysis volatiles and increases the interaction between plastic molecules with internal acid sites. Zhang et al. developed a shape-selective Al-SBA-15 catalyst that converted LDPE into gasoline [58]. Despite having weaker acid sites compared to ZSM-5, Al-SBA-15 (silicon/aluminum molar ratio = 5) achieved 50% conversion of the feedstock at a lower temperature, with enrichment of C4–C10 alkenes and alkanes rather than mono-aromatics. On the other hand, Ratnasari et al. achieved the conversion of HDPE into gasoline-range hydrocarbons through two-staged catalysis of MCM-41 and ZSM-5 (1:1) [59]. By combining the high surface area and large pore volume of mesoporous MCM-41 with the shape-selectivity of ZSM-5, they achieved a 97.72 wt% yield of gasoline range hydrocarbon production with 95.95 wt% aromatics.

Fluid catalytic cracking (FCC) catalysts, a combination of zeolite crystals and non-zeolite acid matrix, have been applied as PW pyrolysis catalysts due to their economical consideration for industrial application, as the spent FCC catalysts are easily available from commercial petroleum refineries. Kyong et al. studied the efficiency of spent FCC catalyst on HDPE pyrolysis and found that it could increase the oil yield from 75.5 to 79.7 wt% with a dramatic decrease of solid residue yield from 4.5 to 0.9 wt% [60,61]. Moreover, the initial temperature of oil formation was lowered with the assistance of spent FCC, demonstrating a significant improvement in the conversion rate of feedstock. In addition, the spent FCC was also applied to the thermal cracking of HDPE [62]. It was found that fewer aromatics but more olefins were produced in the naphtha fraction, which is attributed to the meso- and macroporous pores in the zeolite crystals that favor selectivity to light olefins.

In addition to the catalysts mentioned earlier, metal catalysts and porous carbon are also potential catalysts for PW pyrolysis for their multivalent nature and high specific surface area with rich functional groups, respectively [63-65]. Kumagai et al. investigated the tandem pyrolysis of PET, and the volatiles produced from the first reactor was catalyzed by CaO in the second reactor [66]. They found CaO enhanced the decarboxylation and increased benzene production, resulting in 83.6% selectivity for aromatics at 700 °C. Ji et al. found the addition of transition metal oxide during in-situ pyrolysis of PVC through Py-

GC–MS could reduce the initial decomposition temperature, expand the pyrolysis temperature range, and significantly decrease the proportion of aromatic compounds in products [67]. Additionally, porous carbon materials, such as biochar and activated carbon, with great surface properties and low production cost, are widely used in PW catalytic pyrolysis. Sun et al. utilized sewage sludge-derived char as a catalyst for PW pyrolysis, which successfully increased the aromatic selectivity in produced oil due to the enhanced dehydrogenation contributed by P, Fe, and S-containing active sites, achieving 75.3 wt% monocyclic aromatics production at 600 °C [68]. Mateo et al. synthesized a sulfonated activated carbon to promote the production of bio-jet fuel from co-pyrolysis of biomass and PW, achieving 50 wt% of bio-oil with a high proportion (97.51 wt%) of bio-jet fuels (aromatics and C9-16 alkanes) [69].

Recent studies have focused on the problem of catalyst deactivation during solid waste pyrolysis. This is an inevitable phenomenon during the catalytic pyrolysis of PW, especially for in-situ catalysis, as impurities in PW would limit the activity of the catalysts [19]. Moreover, the effective separation and regeneration of catalysts from solid residues are also a barrier. Meanwhile, in the pyrolysis-catalysis system, the deactivated catalyst caused by coke deposition and active site poisoning is also a tough concern.

2.4. *Effect of pyrolysis types with different reactor designs*

The pyrolysis types with different reactor designs play a crucial role in determining the mass and heat transfer efficiency during pyrolysis. This is particularly important for PW due to the low thermal conductivity of plastic materials. Variations in mass and heat transfer rates of different reactors can affect the PW conversion process, thus influencing the pyrolysis efficiency, product distribution and compositions. Batch reactors and semi-batch reactor (Fig. 4) are widely used for plastic pyrolysis on the lab and industrial scale. However, their disadvantages of poor heat transfer, high labor costs, intermittent operation, and difficulty in large-scale production limited its wider application [25,70].

To enable continuous pyrolysis, fluidized bed reactors (Fig. 4) were developed with high heating rates and short residence time for PW pyrolysis to minimize the secondary reactions of volatiles [71,72]. Park et al. designed a two-stage pyrolysis process containing an auger reactor and a fluidized bed (Fig. 4) for waste PE recycling, which achieved 74.6 wt% gas yield containing 34.5 wt% ethene with a higher proportion of aromatic hydrocarbons in the pyrolysis oil [73]. Despite these advantages, agglomeration of the molten feedstock and the corresponding defluidization are the main problems for fluidized bed reactors, particularly when alkali elements are contained in the feedstock [19]. In this regard, conical spouted bed reactors (CSBR) (Fig. 4) become a better option with good mixing ability of gas and large particle size solid phases, which can significantly improve heat transfer and avoid the defluidization. Elordi et al. conducted the catalytic cracking of HDPE in CSBR at 500 °C with spent FCC catalyst, achieving 50 wt% gasoline yield and about 28 wt% C2-C4 olefins production [74]. To further avoid bed defluidization and improve the efficiency of catalytic plastic pyrolysis in CSBR, Orozco et al. investigated the impacts of plastic type, the ratio of bed mass to plastic feed rate ($W_{bed}/Q_{plastic}$), spouting velocity and use of catalyst. Their results indicated lower temperature was required to avoid defluidization when $W_{bed}/Q_{plastic}$ and spouting velocity were increased [75].

Microwave-assisted pyrolysis (MAP) is a newly developed technology applied in organic solid pyrolysis [76-78]. It directly heats microwave-absorbing materials through dielectric heating provided by microwave irradiation. However, the low dielectric constant of plastics requires mixing with a high microwave-absorbent material during MAP. The application of MAP in PW recycling was first conducted by Undri et al. who found that tires and carbonaceous char, as microwave absorbers, significantly reduced the residence time for HDPE and PP pyrolysis [79]. Subsequently, the microwave power, absorber type, and pyrolysis catalyst became the keys for MAP to maximize oil yield. Zhou

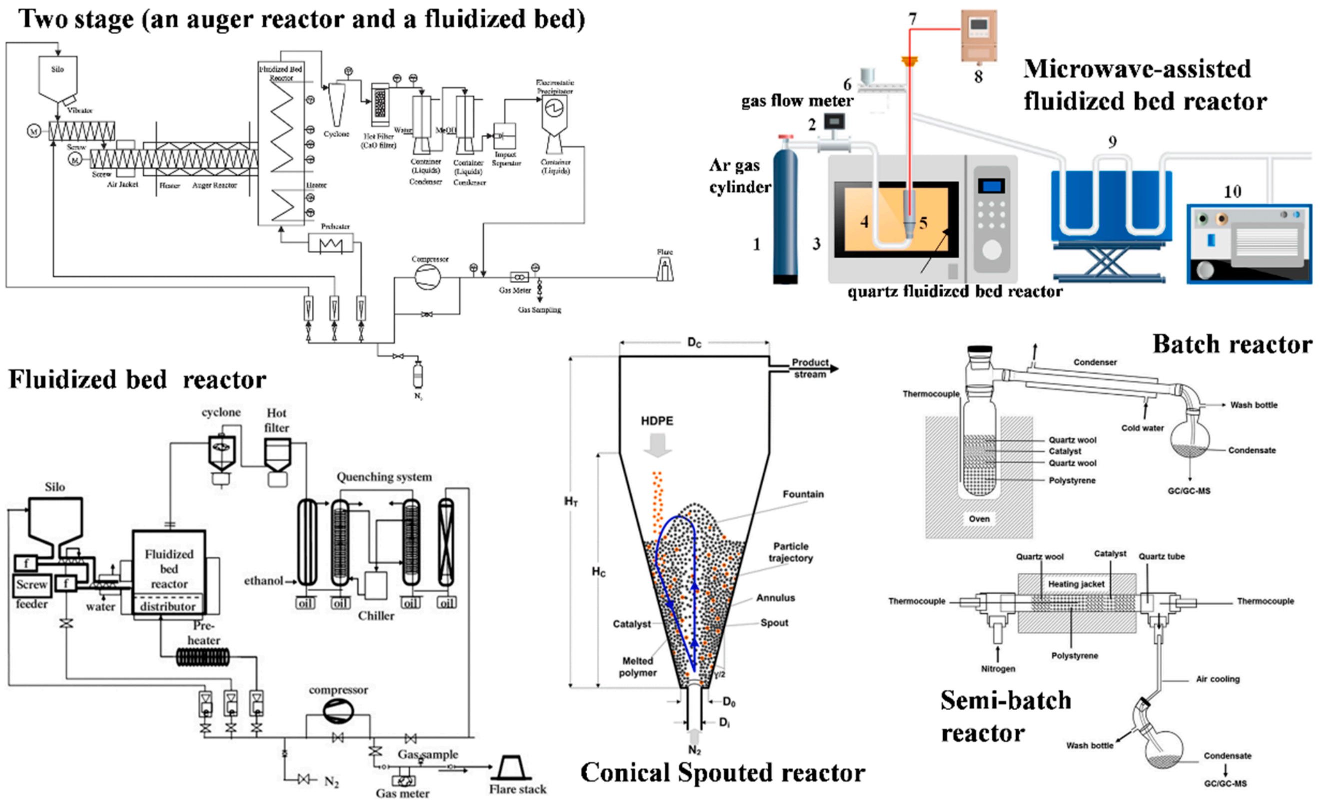

Fig. 4. Scheme of the pyrolysis process in batch and semi-batch reactor [70], fluidized bed reactor [71], conical spouted reactor [74], two-stage pyrolysis process (an auger reactor and a fluidized bed) [89], and microwave-assisted fluidized bed reactor[81] (Adapted with permission from Elsevier).

et al. studied the effect of temperature, plastic composition, and catalyst on the MAP of PW [80]. They found that Talc showed a high cracking activity as a plastic filler for PP pyrolysis. Moreover, the addition of a ZSM-5 catalyst can achieve a 48.9 wt% recovery of liquid oil with 73.5 wt% gasoline-range hydrocarbons. Recently, a microwave fluidizing bed reactor (Fig. 4) was developed by Cui et al. for the PP pyrolysis, which achieved an optimal syngas yield of 76.1 wt% with higher heating value of 51.8 MJ/m^3 when the pyrolysis temperature was 900 °C, fluidizing velocity was 2.36×10^{-3} m/s and microwave power was 800 W [81].

In addition, other advanced pyrolysis reactors have been developed by researchers, such as solar concentrator reactors and autogenic pressure autoclave reactors. Zeaiter et al. converted scrap rubber into gaseous and liquid products through an automated solar concentrator system to utilize solar energy for pyrolysis [82]. The reactor could be heated up to 550 °C within minutes, and the highest gas yield could reach 32.8 wt% with the catalysis of the H-beta catalyst. Zhang et al. conducted waste PET recycling through autogenic pressure pyrolysis in a steel autoclave reactor at 700 °C for 30 min, and the high proportion of methane (34.58 vol%) in the pyrolysis gas makes it an alternation to natural gas [83].

3. ML modeling of plastic waste pyrolysis

The previous section provided a comprehensive technical summary of PW pyrolysis to understand the effects of different important factors on the decomposition process, distribution, and compositions of products from pyrolysis. However, based on the literature review, there is still room for improvement in the yield and quality of the desired products. Co-pyrolysis with a synergetic effect, optimizing pyrolysis processes, synthesizing outstanding catalysts, and designing pyrolysis reactors with good heat transfer have the potential to improve the conversion efficiency, yield, and quality of desired products from PW pyrolysis. However, it is challenging to efficiently find the proper additives, optimal conditions, and effective catalysts for different PW pyrolysis scenarios from simple experimental investigations. In this context, researchers have proposed and applied many state-of-the-art computational technologies, especially emerging ML methods, to aid PW pyrolysis in promoting the sustainable production of desired products [96].

Table 2

Summary of machine learning application in plastic waste pyrolysis.

Feedstock type	Conversions	Input variables	Output targets	ML model	Data size	Best R^2	Ref.
LDPE	Microwave-assisted catalytic co-pyrolysis	Catalyst type, catalysis temperature, space time	C2-C4 olefins	Active learning	18	0.96	[97]
Rice husk and PS	Microwave-assisted co-pyrolysis	Feedstock ratio, heating rate	Liquid, char, and water yields, average heating rate, and conversion percentage	SVR	11	0.81–0.94	[98]
PS	Microwave-assisted pyrolysis	Mass of KOH and PS	Yield of char, gas, oil, pyrolysis time, heating rate, power, energy, and conversion efficiency of specific microwave, and conductive heat loss	SVR	13	0.98–1.00	[99]
Torrefied biomass and plastic wastes	Microwave-assisted catalytic co-pyrolysis	KOH mass and torrefied temperatures	Yield of char, oil, and gas, microwave heating rate, conversion efficiency, and susceptor thermal energy	Polynomial regression	9	> 0.98	[100]
PS with waste tea	In-situ catalytic co-pyrolysis	Mass of PS and waste tea	Yield of water, char, gas, oil, and average heating rate	SVR	11	0.85–0.93	[101]
Mixed plastics	Pyrolysis	Reaction temperature, catalyst ratio, and reaction time	Liquid and gaseous yield	NN, Least Square SVR	1048–1130	0.99	[102]
PP, PE, PVC, PS, PET	Pyrolysis and co-pyrolysis	Plastic type, ultimate analysis, particle size, feed intake, pyrolysis temperature, residence time	Yield of oil gas, char, liquid and gas compositions	SVR, DT, NN, GP	274–301	0.61–0.94	[103]
Biomass and plastics	Co-pyrolysis	Information of proximate and ultimate analysis, reaction temperature, time, heating rate, feedstock ratio	Yield of biochar and bio-oil	NN, XGBoost	94–96	0.85–0.96	[104]
Plastics and biomass	Co-pyrolysis	Ultimate and proximate analysis, pyrolysis conditions	Bio-oil yields and synergistic effects	XGBoost	360	0.86–0.90	[105]
Biomass and polymeric wastes	Co-pyrolysis	Ultimate analysis, proximate analysis, pyrolysis conditions	Yield of char, oil, and gas	NN, ELM, ANFIS, GAM, SVR, and GP	339	0.93–0.98	[106]
Mixture of HDPE, LDPE, PP, and PS	Co-pyrolysis	Ratios of different plastics	Yield of char, oil, and gas	NN	24	0.95–0.99	[107]
PE and PS	Co-pyrolysis	Temperature, PS mass fraction, and flow rate of carrier gas	Yield of char, oil, and gas, oil components and fractions	NN	22	0.87–0.99	[108]
Bamboo sawdust and LDPE	Co-pyrolysis	Signal intensity of representative products	Weight loss	RF, LSTM, MLR	80	0.90–0.99	[109]
Agricultural waste and HDPE	Co-pyrolysis	Pyrolysis conditions	Weight loss	NN	10,000	0.99	[110]
PLA and lignin	Co-pyrolysis	Mixed ratio, temperature, heating rate	Weight loss	NN	–	0.99	[111]
HDPE and oily sludge	Co-pyrolysis	Mixed ratio, temperature, heating rate	interactive effect and activation energy	NN	–	0.99, 0.92	[112]
Mixed plastics	Co-pyrolysis	Temperature, heating rate	Weight loss	NN	358, 752	0.99	[113]

LDPE: Low Density Polyethylene, DT: Decision tree, NN: Neural networks, SVR: Support vector regression, GP: Gaussian process, ELM: Extreme learning machine, ANFIS: Adaptive neuro-fuzzy inference system, GAM: Generalized additive model, PE: Polyethylene, PP: polypropylene, PS: polystyrene, PVC: polyvinyl chloride, PET: polyethylene terephthalate, PLA: polylactic acid.

3.1. *Comprehensive analysis of ML application in plastic waste pyrolysis*

To understand the significance of ML application in PW pyrolysis for aiding energy production, we conducted a comprehensive literature review and create a tabular summary, as shown in Table 2. It was found that ML methods have been widely applied in different types of pyrolysis, including mono and mixed PW pyrolysis, co-pyrolysis of PW with other biomass waste, catalyst-aided pyrolysis, and microwave-assisted pyrolysis. Detailly, co-pyrolysis was the most popular pyrolysis type modeled by ML methods, followed by micro-assisted pyrolysis and normal pyrolysis. Although various catalysts have been used in PW pyrolysis, ML methods have not been well applied to screen or aid in the synthesis of catalysts for catalytic pyrolysis of PW, since only three studies related to the catalyst-aided pyrolysis were modeled by ML algorithms by considering some simply process parameters, according to our literature review.

Regarding the ML model development, Neural Network (NN) and Support Vector Regression (SVR) methods were the most two welcomed algorithms used for modeling PW pyrolysis (Table 2). To develop ML models, the input and output variables need to be determined. For the input variables related to the PW pyrolysis, different inputs were considered in different ML works. Our literature review found that most of the data were from the authors' own experimental setup, which resulted in a wide variation in the number and variables of inputs and outputs, although all of them included the reaction temperature. When using their own experimental data for ML modeling, authors identified only two or three investigated factors as input variables, with a particular focus on the catalyst amount and pyrolysis conditions. However,

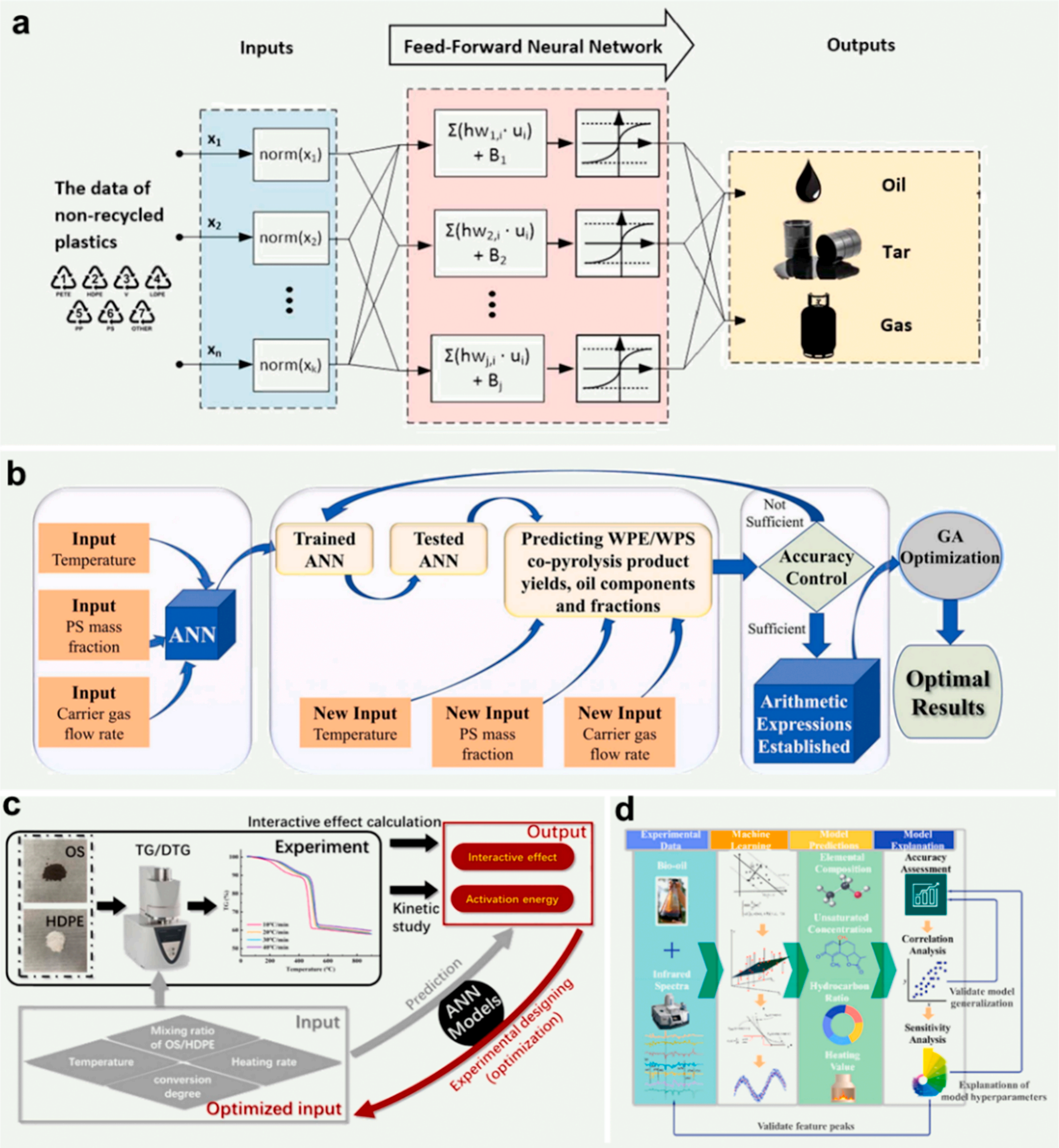

Fig. 5. Prediction of product distribution from pyrolysis of plastic waste by NN model (a) [107], the ML-based prediction and optimization of PE and PS co-pyrolysis for enhancing oil production (b) [108], the prediction and optimization of kinetic parameters (interactive effect and activation energy) in co-pyrolysis of oily sludge and HDPE (c) [112], and the ML-aided fast characterization of pyrolysis oil via combination (d) [120] (Adapted with permission from Elsevier).

when using data collected from the literature, rich inputs were considered, including information (proximate and ultimate analysis) on feedstock and pyrolysis conditions. For the output targets of ML modeling, it should be noted that ML methods have been applied in the PW pyrolysis for different purposes. According to our literature review (Table 2), the most common aim of ML application in PW pyrolysis is to predict the yield and distribution of three-phase products and optimize the pyrolysis conditions to produce desired products. Also, many ML modeling works focused on the pyrolysis kinetic to predict the weight loss and activation energy of PW during pyrolysis. Additionally, ML methods have been shown to be able to cooperate with some characterization instruments to identify the components in bio-oil generated from PW pyrolysis. More details about the ML application in PW pyrolysis are reviewed and discussed in the following sections.

With respect to the data size, the number of data points from the authors' own experimental setups was all less than 30 points, while the sizes of data sets compiled from the literature have significant differences, with the number of data points from hundreds to thousands. Surprisingly, all ML models developed from both small and large data sizes exhibited good prediction accuracy. All the prediction R^2 of the developed ML models were in the range of 0.81 to 1.00, with most of them over 0.90. It is reasonable to expect high prediction performance of an ML model trained based on their own experiments because their own data had high quality with consistent formats and experimental setups. This would help the ML algorithm learn useful information from the data easily to well adapt to the specific procedure. However, it should be addressed that the generalization and robust ability of these ML models with small data sizes from the authors' own experiments are limited.

3.2. ML prediction, interpretation, and optimization of the products from plastic waste pyrolysis

Recent studies have shown ML methods are capable of modeling PW pyrolysis for predicting product distribution from plastic waste (Fig. 5a). Cheng et al. employed four ML methods to predict product distribution, liquid and gas compositions from plastic pyrolysis. They found that the decision three (DT) model was the best one by comparing it with Supporting Vector regression (SVR), Neural Network (NN), and Gaussian Process (GP) models, with a testing $R^2 > 0.85$ for liquid yield production. Furthermore, the DT models were further trained to predict the properties of other products with a testing R^2 of 0.61–0.94 [103]. Sridevi et al. investigated the microwave-assisted co-pyrolysis of rice husk (RH) and polystyrene (PS) [98]. They found that PS enhanced the oil production with a yield as high as 70 wt% under the ratio of 5:1 for PS to RH. They also employed the ML techniques to analyze the role of feedstock blending ratios of PS: RH on the yield of bio-oil yield, biogas yield, biochar yield, water yield, average heating rate, and conversion percentage, achieving good prediction R^2 of 0.81–0.94. Yapıcı et al. developed SVR and GP models to predict gas yield from packaging waste pyrolysis with the consideration of waste types, temperature, heating rate, and type and amount of catalyst as inputs [114]. Good prediction performances from SVR and GP were achieved with R^2 values of 0.89 and 0.93, respectively.

Based on the developed ML models with good prediction performance, new insights into feature importance and correlations to the predicted targets could be unveiled through ML model-based interpretation. Prasertpong et al. developed the XGBoost model to predict the oil yields and synergistic effects of co-pyrolysis of biomass and plastic waste, achieving prediction R^2 values of 0.86 and 0.90 [105]. They used a synthetic minority over-sampling technique for regression with Gaussian noise to increase prediction accuracy by reducing the data imbalance. The SHapley Additive exPlanations method (SHAP) method was also applied to improve the interpretability of the ML mode, revealing that pyrolysis temperature and biomass-to-plastic ratio were the two most important features affecting oil generation. Alabdrabalnabi et al. collected about 95 data points involved in the co-pyrolysis of plastic waste and biomass for ML model development [104]. They employed two ML algorithms, e.g., NN and XGBoost, to simulate the co-pyrolysis process and predict the yields of biochar and bio-oil with high prediction accuracy achieved (R^2 as high as 0.96). They also applied SHAP to work with the developed ML models for understanding the influence of various parameters on the yield of biochar and bio-oil from the co-pyrolysis process, finding that the ratio of PW was the most important factor impacting the yields of both biochar and bio-oil.

Prediction and interpretation are the fundamental application of ML models. Combining ML models with other optimization algorithms can provide more practical insights to guide the experimental investigation (Fig. 5b). Pan et al. developed a hybrid NN-genetic algorithm (NN-GA) model to predict and optimize the oil production from co-pyrolysis of PE and PS [108]. They suggested that NN models could accurately predict the oil yield, components, and fractions with mean absolute errors of less than 8%. Moreover, based on the NN-GA optimization, the highest oil yield of 82.33 wt% and the highest styrene/aromatics ratio of 55.22 wt% were achieved under the optimal conditions of 525 °C with 10 wt% PS. They also claimed that low temperature, high PS mass fraction, and low carrier gas flow rate were preferred to obtain light oil from co-pyrolysis. Shahbeik et al. used evolutionary ML methods to model and optimize the co-pyrolysis of biomass and polymeric wastes [106]. They found that the GP model was the best one with testing R^2 values of 0.98, 0.93, and 0.95 for the prediction of yields of oil, char, and syngas, respectively. Furthermore, they applied a multi-objective particle swarm optimization algorithm to determine the feedstock composition and operating conditions with the objectives of maximizing pyrolysis oil yield and minimizing the char and syngas yield. Under the optimal conditions from the model optimization, they achieved a high oil yield of 70.9–75.3 wt% from co-pyrolysis. L. Quesada examined the mathematical modeling and optimization of pyrolysis of plastic film waste for biofuel generation [115]. They found that the neural fuzzy model with an R^2 of 0.95 was better than the polynomial model for fitting the experimental data. The optimum pyrolysis conditions for oil production were a temperature of 500 °C with a residence time of 120 min and a heating rate of 20 °C/min.

3.3. ML prediction and optimization of the kinetic of plastic waste pyrolysis

In addition to the ML application in PW pyrolysis for prediction, interpretation, and optimization to achieve a high yield of desired products with good quality, ML methods have also been used to investigate the kinetic process of PW pyrolysis [116]. Yin et al. investigated the thermal pyrolysis behavior of HDPE by thermogravimetric analysis (TGA) and NN modeling [117]. They found that the weight loss, activation energy, and pre-exponential factor could be accurately predicted with $R^2 > 0.99$ and a relative error of 6.8%. Dubdub developed an NN model based on the TGA data of mixed plastics, including the mixtures of PS + PP and PS + LDPE + PP, to study their co-pyrolysis behaviors [113]. The weight loss fraction of the mixed polymers could be well predicted with the temperature and heating rate as inputs by NN models, achieving testing $R^2 > 0.99$. Yang et al. investigated the co-pyrolysis of bamboo sawdust and LDPE by a photoionization mass spectrometer (PI-MS) and ML methods [109]. They found that the TGA curve of the mixture of bamboo sawdust and LDPE during pyrolysis could be well predicted by both RF and Long Short-Term Memory (LSTM) models with signal intensity (detected by the PI-MS) of representative products from the corresponding temperature. The prediction accuracies of the TG curve were over 0.98 and 0.90 from the LSTM and RF, respectively. This work provided a reliable tool for monitoring the co-pyrolysis of biomass and plastics and understanding the kinetics of this process with TG curve prediction.

Besides, ML-based optimization strategies have merits in maximizing the synergistic effect of co-pyrolysis, decreasing the activation energy, and promoting the production of desired products (Fig. 5c). Ai et al.

investigated the interactive effect and activation energy of co-pyrolysis between oily sludge (OS) and HDPE [112]. They found that co-pyrolysis of OS with HDPE had a synergistic effect that promoted conversion and accelerated degradation reactions, which may be caused by the abundant •CH_3 free radicals provided by HDPE in co-pyrolysis. Moreover, two NN models were developed to predict interactive effect and activation energy with testing R^2 values of 0.99 and 0.92, respectively. More interestingly, new pyrolysis conditions were optimized based on the developed NN model to acquire the best synergistic effect and decrease the activation energy. The model found that the best synergistic effect (90.61%) was acquired at a pyrolysis temperature of 480 °C, heating rate of 10 °C/min, and a biomass to plastic blending ratio of 0.7 with an experimental validation error of 6%. Dogu et al. developed a tree-based kinetic Monte Carlo with parameter tuning based on Bayesian optimization to model the pyrolysis pathways of PS [118]. The kinetic parameters from the scission pathway were used as inputs to predict the major product yields. It was found that the yield and selectivity of styrene monomer could be efficiently maximized in PS pyrolysis to be reused as a feedstock for styrene polymerization. The highest styrene yield of 77.3 wt% was achieved under the pyrolysis temperature of 600 °C.

3.4. ML-aided characterization and evaluation of pyrolysis oil

ML methods are powerful tools that can also aid the characterization and identification of products from PW pyrolysis, especially for the bio-oil with complex compositions (Fig. 5d). Sholokhova et al. managed to predict and identify the liquid product of plastic pyrolysis followed by ozone treatment using gas chromatography-mass spectrometry (GC–MS) and ML [119]. They trained NN models to predict the retention indices from spectrometry with standard polar and nonpolar phases as input variables. This could assist in determining the composition of the ozone-treated pyrolysis liquid, such as hydrocarbon and oxygen-containing compounds. Chen et al. proposed a fast characterization method that combined attenuated total reflection Flourier transformed infrared spectroscopy (ATR-FTIR) and ML to understand the indicators of bio-oil from pyrolysis [120]. To increase the prediction accuracy of the ML model (SVR algorithm was used), principal component analysis (PCA) was applied to reduce the dimensionality of the original ATR-FTIR data from 7469 to below 20. After the PCA pretreatment, the ML model performance significantly increased with the R^2 values increasing from 0.05 to 21 to 0.62–0.93 for the prediction of unsaturated concentration, effective hydrocarbon ratio, low calorific value, C content, H content, and O content of pyrolysis bio-oil. Moreover, the model-based sensitivity analysis indicated that PC6 and PC7 had the most significant impacts on the prediction performance, which indicated the PC6 and PC7-related peaks were most important for the identification of these bio-oil indicators.

In addition to the above-mentioned ML application in bio-oil characterization from PW pyrolysis, ML algorithms can also be employed to evaluate the potential application of bio-oil and its possible emissions. As if Afzal et al. blended the plastic pyrolysis oil and coconut oil with diesel fuel to evaluate their potential application in a diesel engine by developing an NN to predict the brake thermal efficiency, specific fuel consumption, and the emissions of CO, CO_2, and NO_x [121]. They found that the NN model after hyper-parameter tuning could predict the experimental results well with ratios varying from 90% to 93.5%. Such intelligent model could aid in optimizing engine performance and evaluating the emission features with the bio-oil from PW pyrolysis as energy supply.

4. Challenges and future direction

Pyrolysis is an effective and sustainable technology widely applied in the PW recycling for high-quality product production. The impacts of various factors (e.g., feedstock, process condition, catalyst, and reactors) on the PW pyrolysis were well investigated by the domain research community to improve the conversion efficiency and quality of converted products. However, there are still many challenges and limitations for the technical aspect of PW pyrolysis that need to be addressed in the future.

The complexity of pyrolysis oil products is restricted by plastic types and operation conditions, making it difficult for real application as gasoline, diesel, and other fuels. More studies should focus on developing outstanding catalysts and designing proper feedstock combinations for co-pyrolysis to improve target product selectivity without further downstream processes for oil upgrading. The deactivation and poisoning of catalysts increase the operation costs and raise another challenge for the pyrolysis formula and process design. Moreover, the mechanisms for catalyst deactivation, coke formation, and regeneration during catalytic pyrolysis are still unclear based on current research. Further studies should analyze changes in surface-active sites and internal pore structure of catalysts, as well as the intermediates generated during the process to determine reaction pathways during catalytic PW pyrolysis.

The delicate separation of single PW is a challenge in real industrial applications. Therefore, research focused on the pyrolysis of multi-component mixtures of PW needs to be emphasized in the future. It is of great significance to develop a robust catalyst that can adapt different types of waste for producing desired products. Furthermore, the mixture of feedstock should meet the real waste mixture compositions according to local municipal solid waste classification. The occurrence of PVC in mixed waste PW is inevitable in reality, which would pollute the product and damage reactors by producing corrosive hydrochloric acid during pyrolysis and application. Therefore, advanced techniques are required for separating PVC from the plastic mixture, removing chlorine from PVC during valorization, or getting rid of hydrochloric acid in the final product.

Additionally, the reported works are typically conducted at a gram scale or lower. To achieve industrial breakthroughs and provide practical references to commercial applications, it is essential to conduct pilot-scale studies based on the foundation of lab investigation. Furthermore, comprehensive system perspectives should be improved in the waste to energy [122]. For example, renewable energy supply systems with multiple energy sources (e.g., solar, wind, biomass, organic waste) should be designed for the energy supply of industrial PW pyrolysis with the aim of zero or negative carbon emissions. It should be noted that the required energy could be partially or completely replaced by the thermal energy from pyrolysis gases, oil, and solid residue. Thus, energy recovery and waste management of PW pyrolysis could be implemented as closed-loop recycling.

To promote the development of PW pyrolysis, ML methods have been widely used to predict and optimize the distribution and composition of converted products and the pyrolysis kinetics. They are also applied to cooperate with characterization equipment (e.g., GC–MS and ATR-FTIR) to identify chemical compounds in bio-oil from PW pyrolysis. However, there are still some research gaps and limitations that need to be addressed in the future.

More effort should be made to employ ML algorithms to aid the catalyst screening and synthesis for PW pyrolysis, as the catalyst is the key to increasing the product quality and decreasing the energy consumption during pyrolysis process. For example, our previous work explored the ML modeling of another similar thermal conversion, the gasification of waste with catalyst consideration [123]. A robust NN model was provided to adapt both non-catalyst and catalyst hydrothermal gasification processes of wet waste for the yield prediction of syngas composition (i.e., H_2, CH_4, CO_2, and CO). The robust model was further applied to understand the impacts of the feedstock property, gasification condition, and catalyst properties on syngas yield. Besides, we combined optimization strategy with the robust ML model to develop an inverse design framework for catalyst screening and the optimization of gasification conditions with the aim of H_2-rich syngas production.

Based on the tabular summary in Table 2, over 50% of the datasets have small sizes, with below 100 data points, among which most of them had less than 30 data points. Although these ML models have good prediction performance within their own experimental system, it is challenging to ensure the generalization and robustness of the developed model. Moreover, many of the reported works only considered 2–3 factors in PW pyrolysis as inputs to develop ML models. Other important factors that have been overlooked might play a crucial role in determining the yield and quality of the converted products. To develop a comprehensive ML model, data from different experimental setups and researchers should be collected, considering various types of input factors, including the PW type, catalyst, and operational conditions involved in the PW pyrolysis. However, in practice, it is quite difficult to extract all the input information from different publications due to the irregular format and inconsistency of data provided by researchers. The more factors we considered, the less data there is. Therefore, it is necessary to trade off the number of data points and the number of input features during the dataset-compiling procedure. Additionally, it would be beneficial for data collection if a database with a standardized format of dataset compiling in this domain was published by reputable organizations. This would provide a data pool for the research community to upload and download related data and raise their awareness of data sharing. Besides, traditional modeling approaches, such as Computational Fluid Dynamics and Aspen Simulation, could cooperate with ML to provide more data to overcome the sparse experimental data, such hybrid modeling approach was proposed and detailed introduced in our previous work [28].

For the ML modeling limited to the authors' own experimental data, more innovative ways could be explored by incorporating ML and experiments to promote the PW pyrolysis. One widely used approach is active learning which is based on roughly experimental data to develop a surrogate model first and then combine it with optimization algorithms to achieve goal-oriented reverse engineering practices for new experimental design. This approach can guide experimentalists to conduct further research in the lab, generating more data to update the original dataset and surrogate model. Finally, a model-based prediction-optimization-experiments close loop will be generated to improve the PW pyrolysis performance and achieve the final goal in an iterative way. Based on the literature review, only one similar work has been published in the PW pyrolysis domain. Ureel et al. developed an active-learning strategy that combined k-means clustering with the Expected Model Output Change acquisition function optimized for GP to investigate the catalytic pyrolysis of plastic waste for light olefins (C_2-C_4) production. They found that the active learning strategy achieved a 33%-reduction in experiments [97]. However, this work only optimized the catalysis temperature and time. More efforts should be made to simultaneously optimize the catalyst synthesis and PW pyrolysis in the future.

In addition, a weakness of pure data-driven black-box ML models is their weak interpretability. Although some ML algorithms (e.g., tree-based models) and model explainers (e.g., SHAP method) can provide insights on the feature importance and correlations to the predicted targets, as described in Section 3.2 for PW pyrolysis, such information can only tell us to what extent the input feature impact the prediction performance. It is still unclear why the input features have these impacts. In recent years, physics-informed or domain knowledge-guided ML modeling has been reported. To develop such ML model, domain knowledge or first principal rules are considered in the loss function for ML model training. For example, Zobeiry and Humfeld developed a physics-informed ML approach for solving the heat transfer equation by defining the activation function and loss function of NN based on errors to satisfy partial differential equations, boundary conditions, and initial conditions of heat transfer scenarios [124]. This approach can also be applied to assist the ML investigation of the heat transfer during PW pyrolysis to deeply understand the heating process and enhance the PW conversion.

5. Conclusions

This paper provides a comprehensive review of the state-of-the-art of pyrolysis for PW valorization from both technical and ML modeling aspects. Many studies have investigated the impact of PW type, process condition, catalyst, and reactor type on the yield and quality of products from PW pyrolysis. Pyrolysis temperature plays a critical role in breaking the C–C bonds of PW. Zeolite-related, metal-involved, and carbon-based catalysts are frequently applied in PW pyrolysis. Various reactors, including fluidized bed reactors, microwave-assisted reactors, solar concentrator reactors, and autogenic pressure autoclave reactors, have been developed to assist in PW pyrolysis. However, more efforts are still needed to design outstanding catalysts and select co-pyrolysis additives that can adapt to different PW types, especially the real unsorted PW, and improve the target product selectivity. The mechanism of catalyst deactivation, coke formation, and regeneration, as well as the thermochemical reaction pathways in PW pyrolysis, should be further investigated. To promote the commercialization of PW pyrolysis, more pilot-scale studies should be conducted. Furthermore, renewable energy supply systems with multiple energy sources should be designed for PW pyrolysis plants to achieve carbon neutrality.

In the case of ML application in PW pyrolysis, many studies have explored the potential of ML methods to predict, interpret, and optimize different PW pyrolysis scenarios (e.g., mono and mixed PW pyrolysis, co-pyrolysis of PW with other biomass waste, catalyst-aided pyrolysis, and microwave-assisted pyrolysis) for understanding the process and improving the production of desired products. Co-pyrolysis was the most popular pyrolysis type modeled by ML methods, followed by micro-assisted pyrolysis, and normal pyrolysis. Moreover, the most widely used algorithms for modeling PW pyrolysis were Neural Network (NN) and Support Vector Regression (SVR) methods. Also, there are many limitations that can be further improved in the future. The ML-aided synthesis of catalysts with high selectivity and activity for PW pyrolysis should be further explored in innovative ways, such as active learning with inverse design. More efforts, such as the development of a standardized dataset format and cooperation with traditional modeling, should be made to increase the size of datasets with more relevant factors considered. Last but not least, the interpretability of ML models can be improved by developing physics-informed or domain knowledge-guided models for PW pyrolysis.

CRediT authorship contribution statement

Jie Li: Conceptualization, Data curation, Formal analysis, Funding acquisition, Visualization, Writing – original draft, Writing – review & editing. **Di Yu:** Visualization, Writing – original draft, Writing – review & editing. **Lanjia Pan:** Writing – original draft, Writing – review & editing. **Xinhai Xu:** Writing – original draft. **Xiaonan Wang:** Writing – review & editing. **Yin Wang:** Funding acquisition, Project administration, Resources, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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