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From trash to treasure: Chemical recycling and upcycling of commodity plastic waste to fuels, high-valued chemicals and advanced materials

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ABSTRACT

Of all the existing materials, plastics are no doubt among the most versatile ones. However, the extreme increases in plastic production as well as the difficulty of the material for degradation have led to a huge number of plastic wastes. Their recycling rate after disposal is less than 10%, resulting in a series of serious environmental and ecological problems as well as a significant waste of resources. Current recycling methods generally suffer from large energy consumption, the low utilization rate of recycled products with low added value, and produce other waste during the process. Here, we summarized recently-developed chemical recycling ways on commodity plastics, especially new catalytic paths in production of fuels, high-valued chemicals and advanced materials from a single virgin or a mixture of plastic waste, which have emerged as promising ways to valorize waste plastics more economically and environmentally friendly. The new catalyst design criteria as well as innovative catalytic paths and technologies for plastic upcycling are highlighted. Beyond energy recovery by incineration, these approaches demonstrate how waste plastics can be a viable feedstock for energy use with the generation of clean H_2, high-quality liquid fuels and materials for energy storage, and help inspiring more catalytic process on plastic upcycling to overcome the economical hurdle and building a circular plastic economy.

1. Introduction

Plastic are one of the most versatile materials and have become an indispensable part of our life. They are critical for various economic and society fields such as food preservation, health care products and parts of electronics etc. Billions of tons of plastics have been made over the last seventy years, from only 1.8 million tons in 1950 to 465 million tons in 2018 [1]. In fact, their production is forecast to be more than double by 2050 [2]. However, beyond this rapid and extreme increase in plastic production, most of them (7 billion of the 8.8 billion tons produced globally over the whole period to 2017) have become waste [3]. According to recent statistics, 175 Mt plastic waste enters landfills and the natural environment every 12 months, and this amount could rebuild the Great Wall of China (all 6000 km currently standing) every year [4]. Even worse, the stability of most plastic materials, originating from their chemical inertness, makes them hard to broken down and extremely slow to be degraded.

As a result, if the plastic waste cannot be treated reasonably, it will raise a number of serious global issues from climate change, environmental pollution especially marine pollution, ecological crisis and increasing concerns on human health. The CO_2 emission during the full life cycle of plastics is tremendously huge. By incineration, up to 10 tons of CO_2 are released per ton of plastic waste [5]. It is reported that plastic waste embodies potential CO_2 emissions up to 0.86 gigatons in 2019 and will reach 2.80 gigatons in 2050, which further accelerate climate change [6]. When plastic wastes are buried in landfill, the environmental leakages are detrimental for ecosystems. The amount of plastics trash that flows into the oceans was estimated to be 8 million metric tons annually in 2015 [7], and is expected to be more than 29 million metric tons by 2040. The massive amount of plastic waste, from big pieces to micro-plastics, threatening and contaminating the entire marine lives, from whales to aquatic organisms, and will embedded in

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the ocean's wildlife eventually forever if we could not get the plastic pollution under control. Recently, microplastics are even found in human placenta [8]. Considering these microplastics might carry other chemicals, such as environmental pollutants and additives, it raises great concerns for their potential harmful effects for human being.

The consolation news is that more and more efforts and ambitious commitments by government and industry are made to reduce plastic pollution, increase plastic recyclability, and minimize contamination of plastic waste. Here, we post the citing results on "poly* recycl*" by web of science, as shown in Fig. 1, the number of publications on the topic of polymer recycling has increased by more than 10 times from 2000 to 2020, and the sum of times cited of the publications has increased by more than 100 times in recent 20 years, demonstrating "polymer recycling" as a fast-growing field.

In general, there are four types of recycling processes: namely primary recycling (in-plant recycling), secondary recycling (mechanical), tertiary recycling (chemical recycling by pyrolysis and solvolysis), and quaternary recycling (incineration with energy recovery) [9]. In-plant and mechanical recycling have limited applicability to a certain number of plastic products and materials, respectively; quaternary recycling applicable for almost all the plastics could save some energy, but lead to more CO_2 emissions compared with other three recycling processes. Chemical (tertiary) recycling, with the main advantage to treat heterogeneous and contaminated plastics with help of some pretreatment, has attracted much attention. It is also known as depolymerization to monomers or pyrolysis to chemicals with different carbon numbers, which have been useful for production of fuels or feedstock to produce new chemicals or plastics.

Beyond the four recycling mode of plastics discussed above, there are two recycling concepts arising wide research interest: closed-loop recycling and upcycling. When a recycling process is costly, energy-intensive or deteriorates the properties and appearance of recovered materials, the value is largely lost. It is generally viewed as the dilemma of plastic recycling. Both the new concepts are developed to solve it. Closed-loop recycling is a process whereby the recovered material could be recycled immediately back into the same production cycle and their useful properties could be fully maintained through multiple recycling loops. A widely acknowledged example of this is collecting and reprocessing PET bottles, which is also one of the excellent in-plant and mechanical recycling schemes. Designing new polymers that could be easily recycled to monomers has emerged as a potential chemical way to achieve closed-loop recycling. In the past few years, a series of new polymeric materials designed and synthesized based on this concept, such as polyester [10,11], polydealdehyde [12], polybutydiene [13], polyolefin-like materials [14], etc. Helms and co-workers designed and synthesized a new kind of cross-linked polymers using covalent diketoenamine bonds, which allows the recovery of monomers even in the presence of mixed plastic waste [15].By avoiding single use material deposit and pollution to the environment, synthesizing new polymers based on closed-loop recycling concept are critical to the sustainable development of plastics recycling. However, for most commodity plastics, especially polyolefins, to achieve their closed-loop recycling remains a huge challenge.

On the other hand, chemical upcycling strategy, where plastic waste is converted into value-added chemicals and materials, is more promising to solve the environmental and economic impacts of the plastics problem. Unlike traditional pyrolysis path, the chemical upcycling strategy is more selective and less energy intense. Although most of the chemicals and materials based on this process may still relatively expensive compared to petrochemical feedstock, considering petroleum is a non-renewable fossil resources, plastics could be viewed as valuable resources at the end of their life. Additionally, newly developed process and technologies have emerged as promising ways to valorize waste plastics more economically [5,16,17]. Additionally, the number of publications about the topics of "polymer upcycle" and "polymer upgrade" has increased from 25 in 2000 to 452 in 2020 at an amazing rate. With the increasingly growing concern of environmental impact of plastic waste, plastic upcycling has become a young but very important research field and attracted more and more focus.

After a short description of traditional pyrolysis path, this review focuses on recent chemical recycling and upcycling paths on commodity plastics in production of fuels, high-valued chemicals and materials from a single virgin or a mixture of plastic waste. To clarify the uses of each path on commodity plastic recycling, we classified the commodity plastic into three catalogs: (1) polyolefin (PE and PP); (2) polymers containing aromatic group (PET, PS, PC and PUR et al.); (3) other commodity plastics (PVC and PLA). We highlighted the recent representative cases using newly developed catalytic paths as well as recent innovative technologies for plastic catalytic upcycling. The aim of this review is to help inspiring more catalytic process on plastic recycling and upcycling to overcome the economical hurdle and building a circular plastic economy.

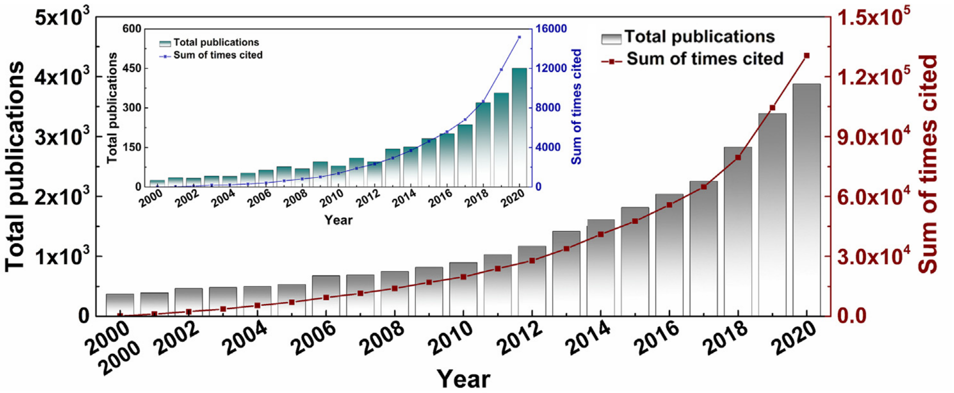

Fig. 1. Total publications and sum of times cited per year of the "poly* recycl*" and the "poly* upcyl* or poly* upgrad*" topics (showing in the inset) searched from web of science core collection between 2000 and 2020 (Indexes: SCI-EXPANDED).

2. Traditional thermochemical recycling route on commodity plastics

2.1. Thermolysis or thermal cracking

Thermolysis or thermal cracking is a thermochemical process that converts waste plastics into a mixture of gaseous, liquid and solid hydrocarbon products at high reaction temperature (usually above 500 °C) in the absence of oxygen. The composition, proportion and properties of the products depends mainly on the nature of the plastic waste and process conditions [18]. Williams et al. reported that thermal cracking of polyolefin plastics, including high-density polyethylene (HDPE), low-density polyethylene (LDPE) and polypropylene (PP) at 700 °C to produce the aliphatic composition dominated by alkenes, alkanes and alkadienes with the negligible concentrations of aromatic species, while thermolysis of polystyrene (PS) or poly-(ethylene terephthalate) (PET), which have aromatic groups in their structures, showed a more aromatic composition [19]. Angyal et al. studied the effect of the concentration of feeding polymers on the properties of degradation products and found that the yield of light products and aromatic content in the naphtha-like fraction was increased with the concentration of PS by thermal cracking of the mixtures of PP and PS at the temperature of 510 ~ 520 °C [20]. Lee and co-workers investigated production of hydrogen chloride, benzenes, and polycyclic aromatic hydrocarbons (PAHs) by thermolysis of polyvinyl chloride (PVC) in N_2. Meanwhile, the usage of CO_2 as reaction medium for thermolysis of PVC can decrease the concentration of benzene derivatives and PAHs at temperatures higher than 480 °C [21].

2.2. Traditional catalytic pyrolysis and hydro-cracking

Compared with thermolysis, catalytic pyrolysis of waste plastics could undergo at relatively lower temperature with shorter reaction time, yielding products of a relatively narrower molecular distribution at the same time [22]. Lee et al. studied the degradation of PS at 400 °C for 2 h over a natural clinoptilolite zeolite catalyst, which shows a good catalytic performance with over 99% of selectivity for aromatic liquid oils of C_5–C_{12} observed and much less formation of residues or cokes [23]. Wang's group reported that Al-Zn and Al-Mg composite catalyst could accelerate the degradation rate of PVC-containing mixed plastics at the temperature of 360 °C ~ 420 °C, and obtained liquid products of a decreased boiling point and a narrow molecular distribution. For example, the distribution of liquid hydrocarbon products was decreased from C_4-C_{25} to C_4-C_{19} over Al-Mg catalyst for pyrolysis of LDPE/PVC [24,25]. They also found that for catalytic degradation of LDPE, the modified ZSM-5 by desilication and incorporation of lanthanum could increase the degradation rate with higher selectivity of olefin and isoparaffin in liquid products compared with the parent ZSM-5 zeolite [26,27]. Hydro-cracking of plastic under high hydrogen pressures can obtain a high-quality gasoline starting from a wide range of waste plastic feeds. Ding et al. systematically studied different hydrocracking behaviors of HDPE over many bifunctional catalysts, and successfully converted HDPE at 375 °C within 1000 psig H_2 over the Ni/HSiAl catalyst into a clean and gasoline-like product, which is even of a better quality than the commercial premium gasoline [28].

3. More selective thermochemical recycling and upcycling route on commodity plastics

Traditional catalytic pyrolysis or hydrocracking of plastic waste at relatively higher temperature always suffer from the low selectivity of products and the poor stability of the catalyst, which hampered further practical applications. A wide variety of more selective thermochemical paths and new catalysts are developed in the last ten years to obtain products with high value under the relatively milder reaction condition. Table 1 summarizes the latest examples within six strategies including hydrogenolysis, oxidation, solvolysis, other C–H functionalization path, tandem process and carbonization that are employed in commodity plastic recycling and upcycling to synthesize fuels, high-value chemicals and materials in recent five years, with the main advantage and challenging of each strategy in our understanding shown in Table 2. All the abbreviations used and their full name are listed in Table 3.

3.1. Hydrogenolysis

Compared to pyrolysis, hydrogenolysis is a catalytic chemical reaction operated in the conditions of a hydrogen rich atmosphere to break the C–C or C–O bonds in plastic waste (PP, PS, PET or the mixed polymers) and add hydrogen to produce solid, liquid and gaseous hydrocarbons and other chemicals. The presence of hydrogen can improve the product quality significantly, i.e. a higher H/C ratio, lower aromatic content and narrower molecular distribution of products, and reduce the production of toxic products [22,29]. Up to now, the hydrogenolysis of plastic waste is still in early development stages and most of the cases are on lab scale investigations. For industrial large-scale application, it is necessary to design more stable catalysts and avoid using high operating pressures.

3.1.1. Hydrogenolysis of polyolefins

Along with the traditional thermochemical studies in the last decade, Dufaud and Basset proposed a different path to convert PE and PP at the moderate temperature of 150 °C and 190 °C, respectively, into diesels or lower alkanes by catalytic hydrogenolysis over a silica-alumina supported zirconium hydride (Zr-H) [30]. Compared with the random cleavage of C–C bond in pyrolysis, the new path has a much higher selectivity due to the β-alkyl transfer. However, the efficiency of conversion was low. After 15 hours of hydrogenolysis of polypropylene (M = 250 000) at 190 °C, only 40% of the starting polypropylene had been converted into light alkanes. Subsequently, they designed another kind of metal hydrides (Hf-H) catalyst with a relatively higher activity compared to Zr-H, which was active in polyolefin depolymerization (PE, PP and polyisobutene) at 150 °C under hydrogen [31]. However, both grafted metal hydride catalysts were unstable and the light alkanes products can be produced much more cheaply in alternative ways from petroleum and other feedstocks.

Catalytic hydrogenolysis of polyolefin plastics waste to liquid fuels over noble catalysts under a relatively mild condition is becoming more popular in recent years. Nakaji and co-workers described a way to convert various polyolefins including LDPE and HDPE over Ru/CeO_2 into valuable chemicals such as liquid fuels (C_5-C_{21}) and waxes (C_{22}-C_{45}) in high yields (83–90%) [32]. Lin and coworkers developed a more efficient proposes to convert HDPE at 220 °C for 1 h by liquid phase hydrogenolysis using Ru/C catalyst [33]. The products are hydrocarbons in the jet fuel and lubricant range, with maximum yields of 60.8 wt% and 31.6 wt%, respectively. Kots et al. discovered PP could be converted selectively to lubricant-range hydrocarbons by Ru/TiO_2 at 250 °C for 16 h, while a much higher fraction of gas product (>95 wt%) was produced over Ru/CeO_2 and Ru/C under the same condition [34].

Beyond screening the support effect, there are more ways toward rational catalyst design. Celik et al. developed an innovative catalyst by dispersing Pt nanoparticles on $SrTiO_3$ perovskite nanocuboids by atomic layer deposition (Fig. 2a) [35]. PE or a single-use plastic bag can be completely transformed into narrowly distributed high-quality liquid products over this catalyst

Table 1

Representative systems for recycling and upcycling of different commodity plastics through newly developed or more selective thermochemical routes.

Plastic	Catalyst	Chemical involved	Product	Yield (%)	Energy use and other application	Temperature (℃)	Time (h)	[Ref.]
Hydrogenolysis								
Polyolefin								
PE	Zr-H	H_2	lower alkanes (C_1 to C_9)	84	gas fuels	150	5	[30]
	Hf-H	H_2	alkanes (C_1 to C_n, $n \approx 30$)	N/A	gas fuels, liquid fuel (jet fuels, diesel fuels) lubricants	150	17	[31]
	Ru/CeO_2	H_2	HCs (C_5-C_{45})	92	gas fuels, liquid fuel, lubricants, waxes	240	8	[32]
	Ru/C	H_2	alkanes (C_5-C_{45})	92	jet fuel, lubricant	220	1	[33]
	$Pt/SrTiO_3$	H_2	liquid HCs(Mn: 820 Da, Mw: 960 Da, Đ: 1.2)	99	motor oil, lubricants, waxes	300	96	[35]
	$mSiO_2/Pt/SiO_2$	H_2	liquid HCs (C_{12}-C_{17})	76	fuels, solvents, lubricants	250	48	[36]
PP	Zr-H	H_2	lower alkanes (C_1 to C_7)	40	gas fuels	190	15	[30]
	Ru/TiO_2	H_2	branched alkanes (Mn: 0.90 kDa, Mw: 0.68 kDa, Đ: 1.3)	66	liquid oil, lubricants	250	16	[34]
Polymers containing aromatic group								
PET	C/MoO_2	H_2	TPA/C_2H_4	87/<5	synthesis of new PET	260	24	[38]
	[Ru(triphos-xyl) tmm] + $HNTf_2$	H_2	1,4-benzene dimethanol and EG	>99		140	16	[37]
PC	[Ru(triphos) tmm] + $HNTf_2$	H_2	bisphenol A and methanol	>99	synthesis of new polymers	140	16	[37]
mixed plastics (PET, PS, PC, PPO)	Ru/Nb_2O_5	H_2	arenes	78.9	chemical feedstock	320	16	[39]
Other commodity plastics								
PLA	$[Ru(triphos)tmm]^+$ $HNTf_2$	H_2	PG	>99	synthesis of linear and cyclic acetals	140	16	[37]
	Ruthenium(II) PNN pincer complexes	H_2	PG	>99	chemical feedstocks	160	24	[40]
	Ru-MACHO-BH complex	H_2	PG	>99	Platform chemical	140	3	[41]
Oxidation								
Polyolefin								
PE	N/A	O_2、NO_x	dicarboxylic acid	65	high-valued platform chemicals	170	16	[47]
	N/A	HNO_3	dicarboxylic acid	71		180	2	[49]
	$[Ni(Me_4Phen)_3](BPh_4)_2$	^{m}CPBA, DCE	hydroxylated PEs	88	macroinitiators	80	3	[55]
	Ru(TPP)Cl		Ox_LDPE	53	adhesives	120	2	[56]
PP	Mn_2O_3	O_2	acetic acid, formic acid	N/A	high-valued chemicals	135	24	[48]
Polymers containing aromatic group								
PS	Transition metal salt	O_2	benzoic acid	N/A	high-valued chemicals	N/A	N/A	[57] [58]
Other commodity plastics								
PVC	Pd/C	NaOH, H_2O_2	organic acid	N/A	high-valued chemicals	180	24	[60]
Solvolysis								
Polymers containing aromatic group								
PET	ultrasmall ZnO nanoparticles	methanol and EG	DMT	95	synthesis of new PET	170	0.25	[68]
	K_2CO_3	methanol and DCM	DMT	93	hydrogenation into CHDM for modification of PET resins	25	24	[69]
	TBD	EG	terephthalamide	89	synthesis of new polymers	110	1	[71]
	TBD:MSA	EG	BHET	91		180	<2	[74]
BPA-PC	TBD:MSA	EG	BPA/ethylene carbonate	89/83		130	4	[75]

Table 1 (*continued*)

Plastic	Catalyst	Chemical involved	Product	Yield (%)	Energy use and other application	Temperature (℃)	Time (h)	[Ref.]
Other commodity plastics								
PLA	N/A	H_2O	lactic acid	>99	synthetic polylactic acid	170–200	1.5	[79]
	[Bmim][OAc]	H_2O	lactic acid	94		120	2	[80]
	KF	CH_3OH	methyl lactate	98	high-valued chemicals	180	<0.2	[81]
	TMC	CH_3OH	methyl lactate	100		50	1	[82]
	TBD	ethanol	ethyl lactate	90		room temperature	0.05	[83]
Other C–H functionalization path								
Polyolefin								
PE	TBADT	DIAD, 1,1,2,2-tetrachloroethane	PE-graft-DIAD		compatibilizers	110	48	[86]
*i*PP	thiocarbonylthio amide reagents	DCP	xanthylated *i*PP	–	functionalized iPP with improved adhesion properties	180	0.5	[85]
Polymers containing aromatic group								
PPE	$Ir(OMe)Cod_2'$	$B_2(pin)_2$	PPE-Bpin	–	can be further functionalized into the epoxy adhesive	80	4	[90]
PS, PSU, PET	Au	methyl propiolate, $AgSbF_6$, $C_2H_4Cl_2$	phenyl rings with 1,2-substituted methyl acrylate functional groups	–	modified physical properties	25	12	[91]
Tandem process								
Polyolefin								
PE	Ru/Ru-Ni	H_2O_2	H_2	51.52	fuel	200	1	[94]
	$Zr(CH_2CMe_3)_2@SiAlO_x$	Al*i*Bu$_3$, O_2	oil containing fatty alcohols and alkanes	31	biodegradable surfactant and detergents	200	12	[95]
Various grade of PE	Ir pincer complex and ReO_x/Al_2O_3	pentane, petroleum ether	liquid alkane products	51–86	liquid fuels	150	24–72	[96]
Various grade of PE	Pt/Al_2O_3		liquid products with a high selectivity to long-chain alkylaromatics	55–80	high-valued chemicals	280	24	[98]
Various polyolefin	$Pt/WO_3/ZrO_2$ and HY zeolite	H_2	branched hydrocarbons	85	liquid fuels	250	4	[97]
Polymers containing aromatic group								
PC	carbonate salt	bis (aryl fluoride)s	PSU	94	reverse osmosis, water purification membranes, medical equipment and high temperature applications	190	18	[99]
PET	Ru/Nb_2O_5		BTX	91	chemical feedstock	220	12	[100]
Other commodity plastics								
PVC	$NaHCO_3$	NaOH, hydrogen carbonate, water	formate	16	platform chemicals	300	8	[101]
PLA	Ru/TiO_2	ammonia	alanine	77	high-valued chemicals	140	94	[102]
Carbonization								
Polyolefin								
PE	–[a]	KOH	HPC	45	supercapacitors	700	1	[104]
			spherical carbon particles	50	lithium-ion batteries	700	3	[108]
	$Co(C_2H_3O_2)_2$		CNT	40	lithium-ion batteries	700	2	[108]
Polymers containing aromatic group								
PS	$AlCl_3$	CCl_4	HPC	40	lithium-ion batteries	900	3	[109]
PET	ferrocene		magnetic fullerene nano-composites	N/A	adsorbent materials	800	24	[110]
Other commodity plastics								
PVC		$Mg(OH)_2$, MnOx	nanoporous carbon	N/A	supercapacitor	700	2	[113]
PTFE		$CaCO_3$, $CO(NH_2)_2$	nanoporous carbon spheres	N/A	supercapacitor	700	2	[114]

(*continued on next page*)

Table 1 (*continued*)

Plastic	Catalyst	Chemical involved	Product	Yield (%)	Energy use and other application	Temperature (℃)	Time (h)	[Ref.]
Mixed plastic								
mixed PP/PE/PS	Ni_2O_3 + CB		CNTs	31.6	supercapacitors	700	N/A	[114]
Mixed Plastics (PS, PP, PE, PVC)	MgO/Fe(acac)$_3$		HCS/PCF	>70	lithium-ion batteries	700	2	[117]
commercial plastics (HDPE, PP, PS)	microwaves and $FeAlO_x$		Hydrogen/ MWCNTs	–[b]	clean H_2 fuel and value-added carbon products	ca. 400	3–5 min	[118]
Real wastes (PP, fabrics, mask wastes, etc.)	–[c]		gases (H_2, CO, C_2H_4, C_3H_6, CH_4, etc.), inorganic minerals and solid carbon	N/A	gases fuel and artificial “coal”	ca. 3000	a few second	[119]
Mixed Plastics (PE, PP, PS, PVC, PET)	–[d]		flash graphene	N/A	bulk composites of plastic, metals, plywood, concrete and other building materials	ca. 2700	one second	[120]

(N/A: not available; –[a]: autogenic pressure carbonization followed by KOH activation process; –[b]: the yield of hydrogen is 55.6 mmol $g_{plastic}^{-1}$, and the yield of MWCNTs is above 92% of the carbon production; –[c]: carbon foam microwave plasma process; –[d]: flash joule heating.)

at 300 °C and 170 psi of H_2, such as lubricants and waxes. The ordering and distances of the Pt site were hypothesized to influence the product selectivity (Fig. 2b). Subsequently, collaborators from Ames national lab incorporated catalytic Pt at the base of an ordered and mesoporous silica pores to achieve selective hydrogenolysis of HDPE into a narrow distribution of short-chain products via a processive mechanism [36]. The processive mechanism is similar to the mechanism of enzyme catalysis, which the polymer threads and binds into the catalytic pore and depolymerized at the active site, releasing a low molecular mass fragment. Selective formation of the products with specific chain lengths was achieved through the design of the active sites and the construction of the reaction environment, providing in sight into for selectively deconstructing the plastic to higher value-added short-chain products.

3.1.2. Hydrogenolysis of polymers containing aromatic group

Hydrogenolysis reaction was also employed to convert polymers containing aromatic group to get their monomers or other higher value-added products. Westhues and co-workers presented a molecular ruthenium catalyst with selected triphos ligands, which can selectively hydrogenolysis of various polyester- and polycarbonate-based materials into the respective value-added diols (Fig. 3) [37]. For example, PET flakes can be fully converted to the diols with excellent selectivity (>99%) using the reported catalyst at 140 °C under 100 bar H_2 for 16 h, and the products can be reused to yield polymeric materials with comparable quality or new value-added polymeric products. Though this approach can provide a novel strategy for efficient polymer recycling, the homogeneous catalyst system in most cases need high H_2 pressures and long reaction times (16 h).

Alternatively, Kratish et al. developed a more air- and moisture-stable heterogenous catalyst, carbon-supported single-site Mo-dioxo complex (C/MoO_2), to selectively hydrogenolysis of PET to terephthalic acid (TPA) and ethylene under 1 atmosphere of H_2 without solvent. Such C/MoO_2 was an attractive catalyst for depolymerizing of polyesters as a consequence of its selectivity, stability, non-toxicity and recyclability [38]. Despite the remarkable progress of designing efficient catalyst for hydrogenolysis of plastic waste, much attention had been paid on the selective conversion of single-component plastic. To broaden the range of degradation substrates, recently, a significant work collaborated by Wang's and Yan's groups showed catalytic hydrogenolysis of various aromatic plastic waste and simultaneously conversing the mixture to arenes via the precise cleavage of C–O and/or C–C linkages over a Ru/Nb_2O_5 catalyst with 75–85% yield (Fig. 4) [39]. This catalytic system offered a new option in transforming aromatic plastic waste into value-added arenes, and these high value-added products can be converted back into the supply chain of plastic production.

3.1.3. Hydrogenolysis of other commodity plastics

Hydrogenolysis of polylactic acid (PLA) can lead to the production of diols. Krall et al. demonstrated that the hydrogenative depolymerization of PLA using ruthenium(II)-PNN pincer complex as a catalyst offers the promise of converting PLA into propylene glycol (PG) [40]. By abstraction of the Cl ligand using KOtBu, the active species is generated in situ with quantitative yield within 24 h at 160 °C. Subsequently, Kindler and coworkers used a commercially available Ru-MACHO-BH complex to hydrogenatively depolymerize PLA into 1,2-propanediol at milder reaction conditions with lower reaction tempreture of 140 °C and shorter reaction time of 3 h. The employed catalyst was reported to be tolerant to the presence of dyes and additives as well [41]. These methods were also effective for accessing useful small molecules of polyesters or polycarbonates as mentioned above.

Table 2
Main advantages and challenges of the selective thermochemical route.

Catalytic path	Advantages	Challenges
Hydrogenolysis	Aims for high quality liquid hydrocarbon products; Produces less toxic products; Tolerant with a certain amount of additives and impurities	Requires hydrogen under high operating pressures; Operates under relatively high temperatures
Oxygenation	Operates under mild reaction temperature; High reaction efficiency within less reaction time; Tolerant with a certain amount of additives and impurities	Usually needs harsh oxidant; Difficult to control the degree of oxidation; Less selective with cross-linking and carbonization product and a certain amount of CO_2
Solvolysis	Operates under mild reaction temperature; High selectivity to monomers and a certain kind of chemicals based on monomer conversion from condensation polymers	Usually needs large amount of solvents; Applicable only to condensation polymers
Other C–H functionalization	Direct transformation of polymers into the value-added materials; Potential to discover new properties within recovered materials with better atomic economy	Generally less selective with a random reaction along the polymer backbone; Requires complex chemical reactant
Tandem process	Increased selectivity control to target product that are difficult to be formed within one step; Open new possibility to produce complex value-added chemicals and materials	Include more chemical operations with potentially higher cost; Requires good compatibility of catalysts used in one pot reaction
Carbonization	Applicable for various types of plastics and mixed plastics; Tolerant with large amount of additives and impurities; Easy for a large scale formation of functional carbon material	Requires high temperature and energy input; Limited to carbon-based materials production

3.2. Oxidation

Compared with thermal/catalytic cracking and hydrocracking, oxidation can transform commodity plastics into various higher value-added compounds due to the introduction of oxygen elements. Considering most of the oxidation process are exothermic, the oxidation path acquires the reaction temperature significantly lower than that of thermal/catalytic cracking. Most commodity plastics could be aged and decomposed with exposure to air, water, or ultraviolet radiation in the natural environment, although some of the process is extremely slow. With help of novel catalysts and selected oxidizing agents, there are big opportunities to achieve selectively cleavage or functionalization at mild reaction conditions either through oxidation of the C–C bond or C–H bond. The biggest challenge involved is to control the oxidation at a certain content, avoiding crosslinking and carbonization side reactions as well as generation of CO_2, which results in a waste of carbon resources.

3.2.1. Oxidation of polyolefins

The most common approaches of oxidation cleavage in polyolefin have been reported using oxidants such as O_2 [42], NO_2 [43-46], NO [47], HNO_3 [45]. Pioneering studies have been carried out by Ogihara et al. with the oxidation cleavage of PE and PP in the presence of NO_2. According to infrared spectroscopy, the hydrogen abstraction reaction could not happen with NO_2 as oxidants at 25 °C, except that the temperature is higher than 100 °C [43,44]. To completely degrade PE into new molecules, such as dicarboxylic acids, which can be used as platform chemicals in turn, it generally requires harsh conditions and a relatively long reaction time. PE was converted into dicarboxylic acid under the oxidation of NO and O_2 at 170 °C for 16 h with 65% yield [47]. Elmanovich et al. used nano Mn_2O_3 to catalyze oxidative degradation of PP using O_2 as the oxidants in supercritical CO_2, and finally obtained acetic acid, formic acid, propionic acid and acetone, among which the selective performance of acetic acid reached 97% [48]. Therefore, it is highly desirable to develop a mild and effective method for oxidation cleavage of polyolefin. Transforming LDPE plastic waste to low molecular weight products via microwave-assisted oxidative process was investigated to be much more effective. In 0.1 g/mL HNO_3 solution, LDPE was degraded to dicarboxylic acid by microwave-assisted circulation for 3 h, and the yield of succinic acid was 36%. The carbon efficiency and total yield were 37% and 71%, respectively [49]. The group subsequently applied the same method to transform HDPE into short dicarboxylic acids with higher selectivity. The yields of succinic acid, glutaric acid, and adipic acid were 49%, 39%, and 12%, respectively [50]. Besides, directly using the obtained mixture to synthesize oligoesters has been demonstrated to be effective plasticizers for polylactic acid (PLA) (Fig. 5).

In addition, a greener and milder oxidant H_2O_2 could be employed in oxidative degradation of polyolefins. Zhou et al. obtained C_1-C_5 fatty acids such as acetic acid and propionic acid by hydrothermal oxidation of H_2O_2 at 300℃ for 30 min without any catalyst, with a total yield of 20% [51]. By sulfonating PE with chlorosulfonic acid firstly and complexing it with Fe^{2+}, Chow et al. developed a highly efficient oxidative method to convert the pretreated PE into succinic acid and other small acid molecules with H_2O_2 at room temperature within 1 h. Fe^{2+} on pretreated PE catalyzed H_2O_2 in situ to produce hydroxyl radicals, which are facilitating to attacked the carbon–carbon double bond nearby [52].

The oxidative functionalization of C–H bond is an attractive approach to introduce polar functional groups, which is another viable path to upcycling waste plastics. Compared to oxidation cleavage, the oxidation of C–H bond can transform polyolefins into value-added polymers or new functional materials without changing the molecular weight, mechanical properties as well as thermal properties. Early reports on the C–H functionalization of polyolefins by free radicals could lead to side reactions of scission and cross-linking of the chains due to excessive oxidation. As an alternative to free-radical oxidation, more selective approaches of metal-catalyzed oxidation of C–H bond have been developed. Boaen and co-workers employed Mn-catalysts for the functionalization of poly(ethylene-alt-propylene) (PEP) using oxone (potassium peroxymonosulfate) as the terminal oxidant (Fig. 6b). In this reaction, hydroxyl groups and carbonyl groups were installed at the tertiary carbon sites and secondary carbon atoms, respectively [53]. Subsequently, Bae et al. reported the Rh-catalyzed borylation of polyolefins (Fig. 6a). The hydroxyl group appears on the alkyl side chain [54]. To improve the selectivity of carbon atoms, Bunescu and co-workers reported Ni-catalyzed C–H oxidation, which occurred at the secondary C–H bonds of polyethylene

Table 3
Abbreviations.

Abbreviation	Full name
Plastic	
Polyolefin	
PE	Polyethylene
HDPE	High-density polyethylene
LDPE	Low-density polyethylene
PEP	Poly(ethylene-alt-propylene)
PP	Polypropylene
iPP	Isotactic polypropylene
Polymers containing aromatic group	
BPA-PC	Bisphenol A based polycarbonate
PC	Polycarbonate
PET	Poly (ethylene terephthalate)
PPE	Poly (phenyl ether)
PS	Polystyrene
PSU	Poly (aryl ether sulfone)
Other commodity plastics	
PLA	Polylactic acid
PTFE	Polytetrafluoroethylene
PVC	Polyvinyl chloride
Catalysts	
C/MoO_2	Carbon-supported single-site Mo-dioxo complex
EG	Ethylene glycol
$Fe(acac)_3$	Iron (III) acetylacetonate
$HNTf_2$	Bis(trifluoromethanesulfonyl) imide
^{m}CPBA	*meta*-chloroperoxybenzoic acid
MSA	Methane sulfonic acid
Ni/HSiAl	Ni and NiMo sulfides loaded on a hybrid support
$SrTiO_3$	Strontium titanate
TBADT	tetrabutylammonium decatungstate
TBD	Triazabicyclodecene
Zr-H	Silica-alumina supported zirconium hydride
ZSM-5	Zeolite socony mobil-5
Products	
BHET	Bis(2-hydroxyethyl) terephthalate
BPA	Bisphenol A
BTX	Benzene, toluene and xylene
CB	Carbon black
CDs	Carbon dots
CNMs	Carbon nanomaterials
CNTs	Carbon nanotubes
DMT	Dimethyl terephthalate
FMFNc	Functionalized magnetic fullerene nanocomposites
HCs	Hydrocarbons
HCS	3D structured hollow carbon sphere
HPC	Hierarchical porous carbon
HQLs	High-quality liquid products
LIBs	Lithium-ion batteries
Me-LA	methyl lactates
MWCNTs	Multi-walled carbon nanotubes
PAHs	Polycyclic aromatic hydrocarbons
PCF	Porous carbon flake
PG	Propylene glycol
PPE-Bpin	Boronate ester functionalized PPE
PTA	Terephthalic acid
TMAD	Trimethylolpropane allyl ether diol
TMC	Tetramethylammonium methyl carbonate
TPA	Terephthalic acid
Others	
APR	Aqueous phase reforming
$B_2(pin)_2$	Bis(pinacolato)diboron
CAM	Cross alkane metathesis
CHDM	1,4-cyclohexanedimethanol
DCE	Dichloroethane
DCP	Dicumyl peroxide
DIAD	diisopropyl azodicarboxylate
FG	Flash graphene
FJH	Flash joule heating
MWD	Molecular-weight distribution
THF	tetrahydrofuran

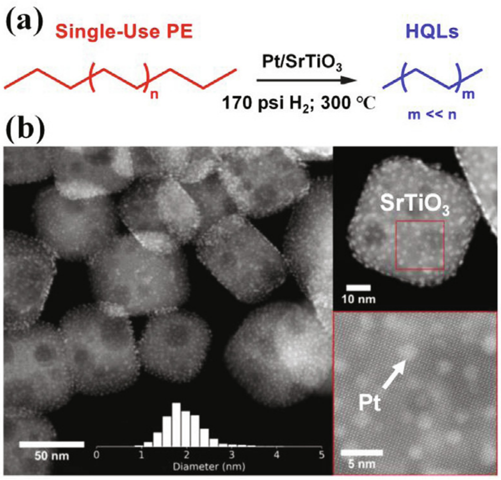

Fig. 2. (a) Hydrogenolysis of PE into high-quality liquid products (HQLs). (b) Electron micrographs of Pt NPs with an average size of 2.0 ± 0.5 nm deposited on $SrTiO_3$ nanocuboid supports. Reproduced from Ref. [35] with permission from American Chemical Society.

under mild conditions (Fig. 6c) [55]. However, this catalyst could incorporate chlorine due to poor chemoselectivity. Furthermore, the use of a strong oxidant (^{m}CPBA, *meta*-chloroperoxybenzoic acid) might produce esters as a side product. More recently, Chen and co-workers reported Ruthenium-catalyzed oxidation of C–H bonds using milder oxidants to avoid over oxidation, which had high turnovers and selective oxidation of varying architectures PE without chlorine and side reaction to form esters could be realized (Fig. 6d). Remarkably, the functionalized PE exhibited strong adhesive properties and could be painted with common waterborne latex paint, which makes it possible to upcycle existing commodity plastics into more valuable and potentially more sustainable materials [56]. However, these methods for the oxidation of polyolefin had not been widely used due to its high cost of the used homogeneous metal complex catalyst and the low efficiency. There is still a huge opportunity to develop effective heterogeneous catalytic system and more efficient and sustainable oxidation method to convert polyolefin into high-valued chemicals and materials.

3.2.2. Oxidation of polymers containing aromatic group

There are patents reported on the use of manganese, vanadium, chromium, iron, zirconium, molybdenum and other heavy metal salt as catalysts to obtained benzoic acid by catalytic oxidative degradation of PS [57,58]. Feng et al. introduced sulfonic acid group to the benzene ring of PS and complexed with Fe^{2+} under a similar strategy as mentioned above for PE. More than 99% of the pretreated PS was oxidized and decomposed into small molecules containing benzene ring with H_2O_2 reaction [59].

3.2.3. Oxidation of other commodity plastics

There are few studies on oxidation of PVC into valuable products. In most cases, total oxidative degradation of PVC into CO_2 is reported. Strikingly, a two-step hydrothermal dechlorination/oxidative degradation of PVC using NaOH as base, H_2O_2 as oxidant and Pd on activated carbon (Pd/AC) as catalyst has been reported. Along with a dechlorination rate up to 90%, water-soluble organic acids has been confirmed [60].

3.3. Solvolysis

Solvolysis, including hydrolysis, aminolysis, ammonolysis, alcoholysis (glycolysis and methanolysis) and phosphorolysis, is employed as a more selective and efficient way to depolymerize of plastics compared to the pyrolysis, which can be used to selectively cleave the ether, ester and acid amide bonds with the production of monomers [61,62]. Considering the monomers could be re-polymerized to obtain new polymers, solvolysis is viewed as a closed-loop recovery method. However, the applicability of solvolysis is less extensive than that of hyderogenolysis or oxidation as mentioned above. Solvolysis is only suitable to condensation polymers, and doesn't work for polyolefins and PVC. In addition, for an industrial application in large scale, it is necessary to reduce the amount of solvent used, to optimize the route of product separation and purification, or even to achieve self-separation of products and solvent.

3.3.1. Solvolysis of polymers containing aromatic group

Karayannidis et al. depolymerized the PET with aqueous NaOH solutions (1.125 M) at 200 °C with a TPA yield of 98% in 1 h [63]. Mancini and Zanin acquired the TPA monomer from PET through acid hydrolysis process using aqueous solutions of sulfuric acid (7.5 M) and the reaction extent reached 90% in 5 h at 135 °C [64]. To avoid the corrosion by the strong acid and alkali, Liu et al. employed the supercritical methanol to decompose PET at 298 °C in 112 min, which can produce the dimethyl terephthalate (DMT) with high yield of 99.79% [65]. However, the need of the conditions of high temperature and high pressure as well as the

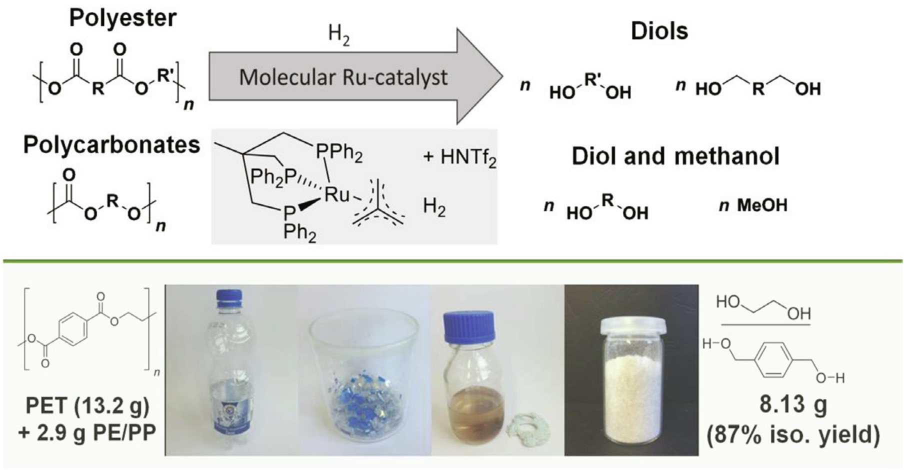

Fig. 3. Reductive depolymerization of polymeric waste using [Ru(triphos)tmm] or [Ru(triphos-xyl)tmm] and bis(trifluoromethanesulfonyl) imide ($HNTf_2$) as catalyst system. Reproduced from Ref. [37] with permission from American Association for the Advancement of Science (AAAS).

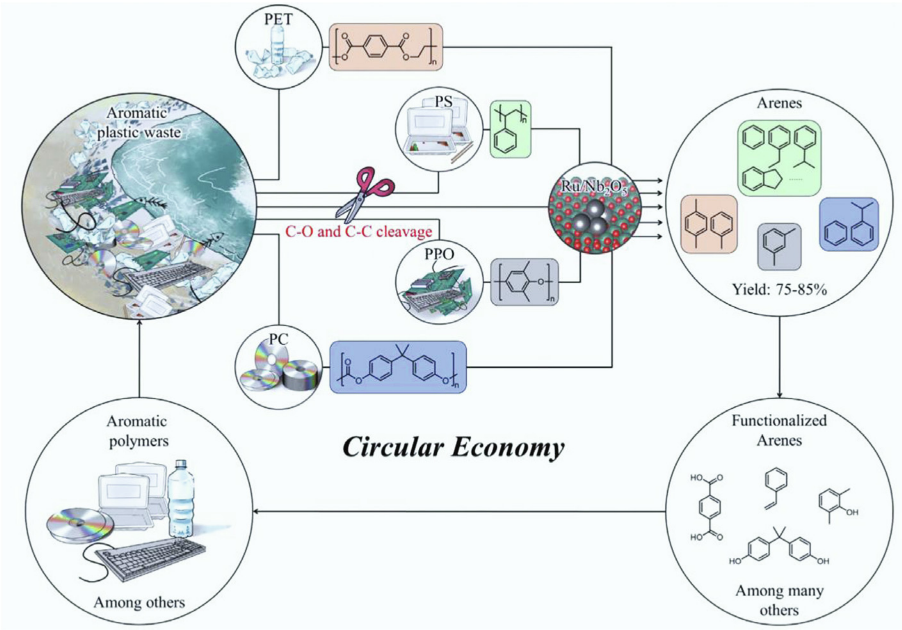

Fig. 4. The integration of a C–O and C–C bond cleavage catalyst into the circular plastic economy, and conversion of various aromatic plastic waste and mixtures thereof into arenes over a Ru/Nb_2O_5 catalyst. Reproduced from Ref. [39] with permission from Wiley-VCH.

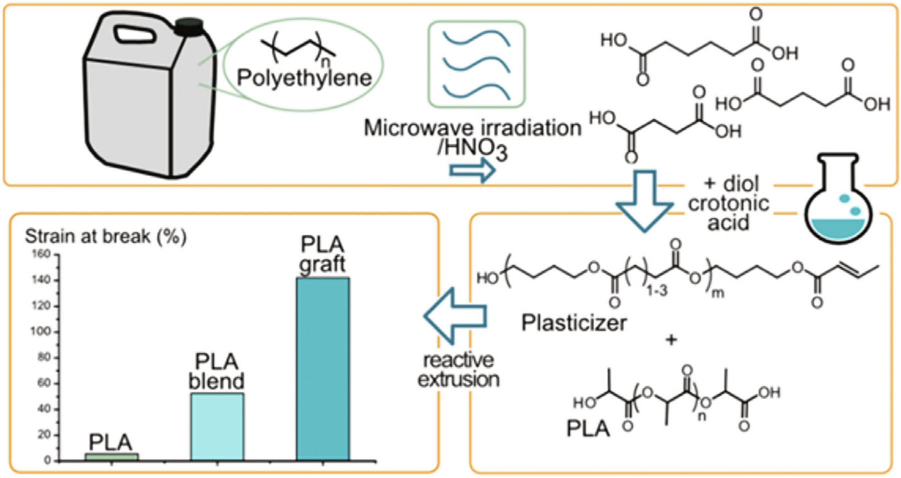

Fig. 5. The waste polyethylene was transformed into covalent polylactic acid plasticizer under the assistance of microwave. Reproduced from Ref. [50] with permission from American Chemical Society.

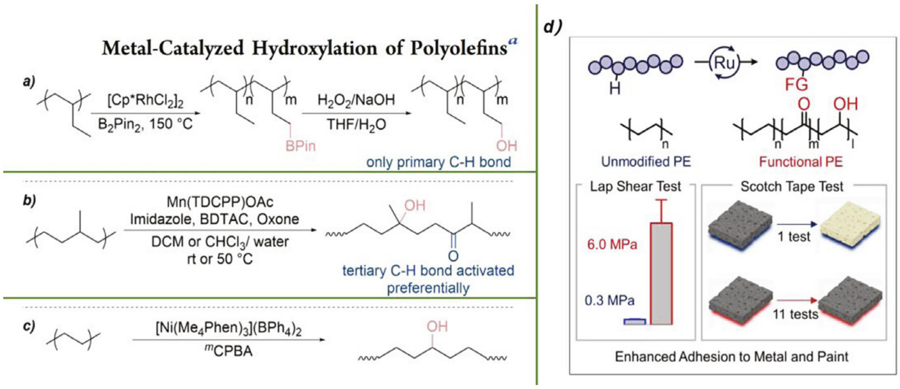

Fig. 6. (a) Rh-catalyzed, (b) Mn-catalyzed and (c) Ni-catalyzed the C–H oxidation. Reproduced from Ref. [55] with permission from American Chemical Society. (d) Ru-catalyzed the selective oxidation of C–H bonds on polyethylene. Reproduced from Ref. [56] with permission from Elsevier.

operating costs of these approaches greatly limit the large-scale or industrial applications [66,67]. In order to lower the energy consumption, Du et al. adopted methanol dispersions of ultrasmall ZnO nanoparticles (with a size of about 4 nm) as pseudohomogeneous catalysts for alcoholysis of PET, and the yield of DMT can reach 95% at 170 °C after 15 min [68]. Pham and Cho developed another novel catalytic route of methanolysis worked even under ambient conditions, efficiently converting a single layer of waste PET bottle flakes into DMT with the yield reaching 93.1% in 24 h using K_2CO_3 and dichloromethane as catalyst and cosolvent, respectively [69]. Nevertheless, the separation and purification costs become much larger than the depolymerization steps, and the residual catalysts, some unwanted by-products or other contaminants would lower the quality of the recycled materials and reduce the value of products [70].

Recently, some new organic catalysts have been developed to "solvolysis" of polymer waste in the absence of solvents. Fukushima and co-workers demonstrated that the introduction of a potent organic triazabicyclodecene (TBD) catalyst can promote the aminolytic depolymerization of waste PET in the absence of solvents, resulting in a broad range of crystalline terephthalamides products [71]. Compared to the conventional solvonolysis, such path shows no requirement for the harsh reaction conditions and unrecyclable metal catalysts (e.g., various metal salt forms such as metal acetates, metal oxides and metal hydroxides) [72,73]. Furthermore, Jehanno et al. prepared an innovative protic ionic salt organocatalyst by mixing an equimolar quantity of TBD and methanesulfonic acid (MSA), which can completely glycolysis of PET into highly pure bis(2-hydroxyethyl) terephthalate (BHET) monomer at high yields (over 90%) without solvent [74]. This catalyst showed extremely high thermal stability, which can be easily reused and recycled for several reactions. Subsequently, the authors employed the TBD:MSA organocatalyst for catalytic depolymerization of bisphenol A based polycarbonate (BPA-PC) to access the high-valued functionalized cyclic carbonates [75].

Although the individual depolymerization of plastic of different nature have been achieved, how to simultaneously solvolysis of the mixed plastic waste is still a big challenge. Lately, Sardon's group took advantage of the energetic differences between the glycolysis of PET and BPA-PC which is emphasized by their computational investigations, explored the selective and sequential depolymerization of different BPA-PC/PET mixtures in the presence of a protic ionic salt TBD:MSA organocatalyst (Fig. 7a) [76]. High added-value products including the starting monomers, BPA, BHET or functionalized cyclic carbonates can be obtained through this methodology. Moreover, selective depolymerization of different BPA-PC/PET mixtures, such as commercial pellets and real aromatic plastic waste, was achieved using trimethylolpropane allyl ether diol (TMAD) as nucleophile under inert atmosphere at 130 °C. BPA and functionalized carbonate were produced with high yield without depolymerization of PET, which providing great potential for selective recycling mixtures of waste plastics and commercial blends (Fig. 7b).

3.3.2. Solvolysis of other commodity plastics

PLA hydrolysis to lactic acid is known to proceed at relatively high temperature (120–250 °C) due to its inherent insolubility in water [77]. As hydrolysis proceeds, the pH of the solution decreases, which results in an rate acceleration [78]. Villani et al. observed a complete hydrolysis of PLA within 90 min at reaction temperatures (170–200 °C) [79]. To overcome the challenge of PLA insolubility in water, Song et al. employed ionic liquids, [Bmim][OAc], as a catalyst in water to achieve a high yield of lactic acid up to 94% within 2 h under a relatively mild condition at 120 °C [80]. However, the high catalyst loading and price limited their further scalable application.

PLA alcoholysis to alkyl lactates (lactate esters) arises as another attractive path beyond hydrolysis, which could be achieved using a simple metal salt or a cheap organocatalyst. Recently, Alberti et al. used a alkali metal halide salts, KF, as a cat-

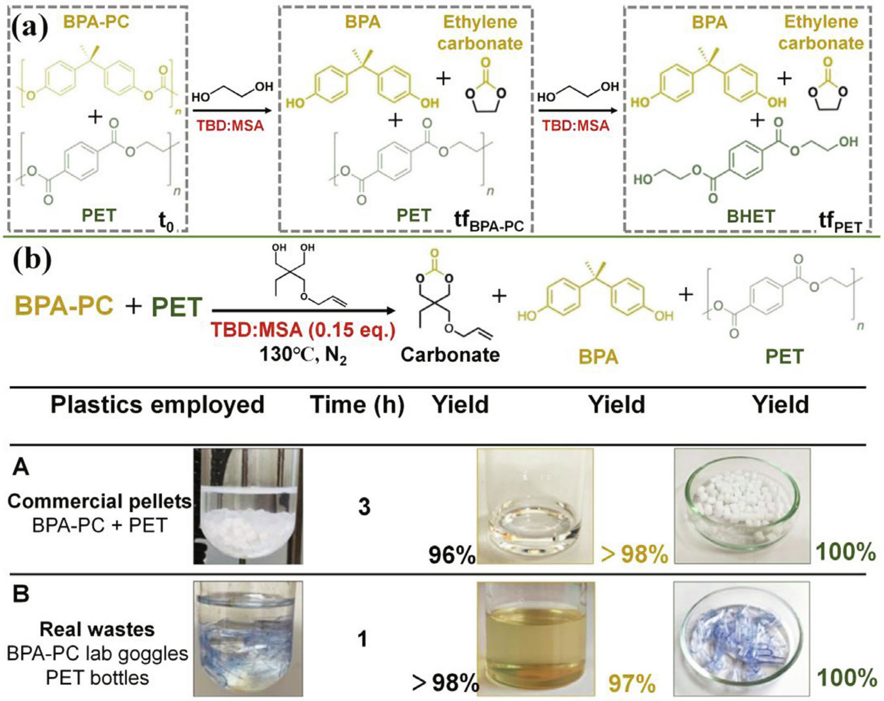

Fig. 7. (a) Scheme of the selective glycolysis of BPA-PC and PET over a protic ionic salt TBD:MSA organocatalyst. (b) Selective depolymerization of BPA-PC/PET mixtures with (A) commercially available BPA-PC and PET pellets and (B) BPA-PC from laboratory goggles. Reproduced from Ref. [76] with permission from Wiley-VCH.

alyst to facilitate PLA methanolysis at 180 °C, and obtained high yields of methyl lactates (Me-LA) up to 98% within 10 min [81]. This process could be more efficient using an organocatalyst. McKeown et al. used tetramethylammonium methyl carbonate (TMC) as a simple and cheap organocatalyst for PLA methanolysis and achieved a 100% Me-LA yield within 1 h at 50 °C in tetrahydrofuran (THF) [82]. Notably, the catalyst loadings could be as low as 0.5 mol% to retain their high activity, which is a promising feature among organocatalysts, especially compared ILs catalysts as mentioned above. Another representative example reported by Leibfarth et al. was TBD as an extremely efficient catalyst for PLA ethanolysis, achieving >90% Et-LA yield within 3 min at room temperature. The remarkable activity was proposed through a simultaneous activation of both the carbonyl group of PLA and incoming ethanol via H-bonding with TBD, and could be extended to a range of primary alcohols [83].

3.4. Other C–H functionalization

Except for the widely used hydrogenolysis, oxidative degradation and solvolysis, other C–H functionalization ways are proved as alternative methods to convert plastic waste to high value-added products. Recently, Williamson et al. had summarized these works and reviewed the lasted development in the C–H functionalization of polymers [84], which inspired chemists develop new methods to reuse post-consumer plastic waste by polymer C–H functionalization. Here, we listed some representative new examples to further demonstrate the advantage and challenging of this strategy.

3.4.1. Other C–H functionalization on polyolefins

As a representative new work of this field, Williamson developed a metal-free postpolymerization modification approach to functionalize the isotactic polypropylene (iPP) using thiocarbonylthio amide reagents (Fig. 8) [85]. Such thermally initiated C–H xanthylation of iPP could occur without coincident chain scission. The mechanistic hypothesis by systematic experimental and kinetic study revealed that the amidyl radical could abstract hydrogen atom and the thiocarbonyl group of the reagent subsequently trap the polymer-centered radical. The obtained functionalized iPP, while maintaining the tensile properties, showed that the adhesion strength to polar substrates was twice that of commercially available iPP. Kong et al. grafted the diisopropyl azodicarboxylate (DIAD) onto the PE at 110 °C using tetrabutylammonium decatungstate (TBADT) as an organocatalyst [86]. The polarity of functionalized PE can be adjusted by changing the amount of DIAD or TBADT used in the reaction, and the resultant PE-graft-DIAD can be used as compatibilizers to blend polyolefin plastics and starch. Besides, there are many studies on the C–H functionalization of commodity polymers including aromatic polymers, polyolefins, aliphatic polyethers and polyesters.

3.4.2. Other C–H functionalization on polymers containing aromatic group

Polymers containing aromatic group can be modified by C–H functionalization of the aromatic rings, and the most widely applied methods are Friedel-Crafts alkylations and acylations [84]. In addition, halomethylation, sulfonation, alkyl-Li metalation, perfluoroalkylation and Ir-catalyzed borylation can be applied for the polymers with aromatic rings to create the value-added materials [87-89]. Feng et al. employed the C–H borylation reaction to convert poly (phenyl ether) (PPE) into the borylated analog (PPE-Bpin), which can be further functionalized into the epoxy adhesive without destruction of the polymer's backbone [90]. The obtained white adhesive products possess an adhesion energy of 12 kJ/m^2, which correlated to an adhesive strength of 5 MPa at room temperature. However, these approaches in most cases are nonselective, and need strict conditions as well as complex fabricating procedures which resulted in deleterious side reactions and cumbersome purification. Very recently, King et al. demonstrated the first catalytic approach using homogeneous gold (Au) catalysis to functionalize polymers containing aromatic group [91]. The catalytic application of homogeneous gold complexes between methyl cc and nucleophilic arenes within PS, PSU, and PET resulted in their direct functionalization with methyl acrylate functional groups without side reactions. The reactivity and functionalization depended on the catalyst structure and the reactions provide a platform to chemically transform aromatic polymers in a mild, chemoselective and practical way.

3.5. Tandem process

Most plastics are widely used due to their chemical inertness and their structural diversity. There is a huge challenge for their chemical decomposition at a molecular lever, and it is even more difficult to achieve it using a single path. A tandem process combined with two or more reactions at the same time or in a sequence could more selectively depolymerize and functionalize polymers, including polyethylene, the most difficult one to do so.

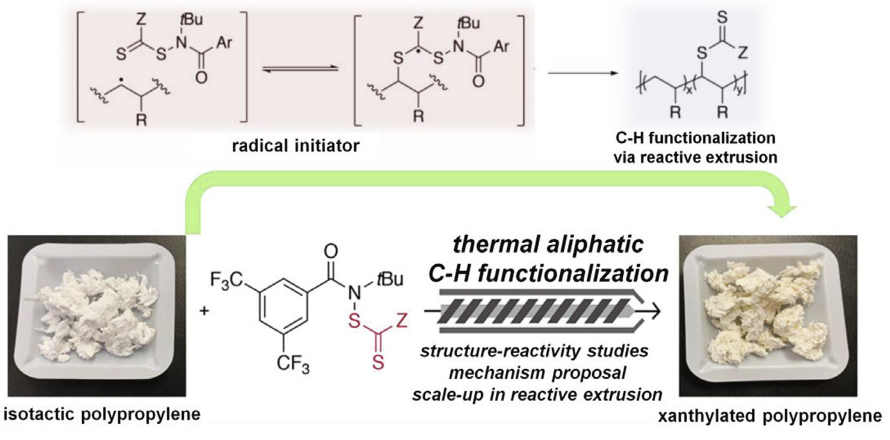

Fig. 8. Transformations of isotactic polypropylene into xanthylated polypropylene though thermal aliphatic C–H functionalization. Reproduced from Ref. [85] with permission from American Chemical Society.

3.5.1. Tandem process on polyolefins

Usually, polyolefin plastics need harsh condition to be converted to hydrogen, and the techniques include high-temperature pyrolysis (700–900 °C) [92] or supercritical water gasification ($T > 374$ °C, $P > 22.1$ MPa) [93] Recently, Su et al. developed an innovative two-step process integrating hydrothermal oxidation pretreatment and catalytic aqueous phase reforming (APR) to convert waste PE into H_2 under mild hydrothermal conditions [94]. Rapid oxidation of waste PE with diluted H_2O_2 into small acid molecules was firstly achieved, and reforming of the aqueous phase derived from the first step process was proceeded over a Ru or a Ru-Ni catalyst, which can make 10.83 moles of hydrogen per kilogram of plastic, with a hydrogen molar fraction of more than 50%. Beyond H_2, there are large opportunities to obtain high-valued chemicals from tandem process on polyolefins. Kanbur et al. developed a new catalytic method to transform polyolefins into biodegradable fatty alcohols and fatty acids (Fig. 9) [95] This newly reported catalytic reaction process involving a sequence of zirconation via C–H bond activation, β-alkyl elimination for C–C bond cleavage and heterobimetallic alkyl group exchange for C–Al bond formation, and the resultant aliphatic alkylaluminum species can be further converted into high-value fatty acids or alcohols.

The tandem process could proceed in one pot using a mixture of two or more different kind of catalysts. For example, a new distribution of alkane products could be achieved by cross alkane metathesis (CAM) over a combination of catalysts work for a sequence of dehydrogenation–metathesis–hydrogenation reactions [96]. Considering PE as ultra-long chain alkanes, inspired by the previous work on small molecules, Jia et al. firstly developed a catalytic system for selective degradation of PE into liquid fuels and waxes at mild conditions (150 °C) with assistance of light alkanes reactants (Fig. 10a). A mixed Ir-pincer metal complex and supported rhenium oxide catalysts worked for dehydrogenation/hydrogenation and metathesis reaction (Fig. 10b), respectively, which converted PE to short hydrocarbon segments after multiple cycles of CAM with light alkanes. Although the use of precious metals (Ir and Re) and the challenging synthesis of the designer ligands is difficult to implement on a practical application, such study with a tandem catalytic system represent a great proof-of-concept. Another example was recently reported by Liu and Vlachos et al. Using a mixture of $Pt/WO_3/ZrO_2$ and HY zeolite, polyolefins could be selectively converted to branched, liquid fuels including diesel, jet, and gasoline-range hydrocarbons, with high yield up to 85% at 225 °C. The process was proposed via tandem catalysis with initial activation and subsequent cracking over the acid sites of WO_3/ZrO_2 and HY zeolite, isomerization over WO_3/ZrO_2 sites, and hydrogenation of olefin intermediates over Pt [97].

The tandem process could also proceed in one pot using one multifunctional catalyst. Based on a simple Pt/γ-Al_2O_3 catalyst, Zhang et al. pioneered a tandem catalytic hydrogenolysis and aromatization strategy to upgrade PE of various grade to long-chain alkylaromatics with a high selectivity and narrow distribution [98]. Since aromatization is strongly endothermic at moderate reaction temperatures, the key to the formation of alkylaromatics is the coupling of the difficult reaction with hydrogenolysis, which is much easier and provides an internal source of energy. Control experiments with $C_{30}H_{62}$ showed a significant decreased amount of alkylaromatics and proved PE with long-chain structure as a better feedstock within the tandem reaction system (Fig. 11). Further-

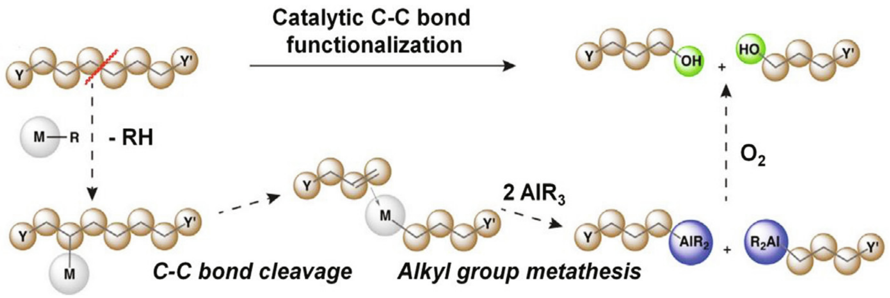

Fig. 9. Catalytic conversation of saturated hydrocarbons into biodegradable surfactants via C–C bond alumination for tandem carbon–carbon bond cleavage and carbon–aluminum bond formation. Reproduced from Ref. [95] with permission from Elsevier.

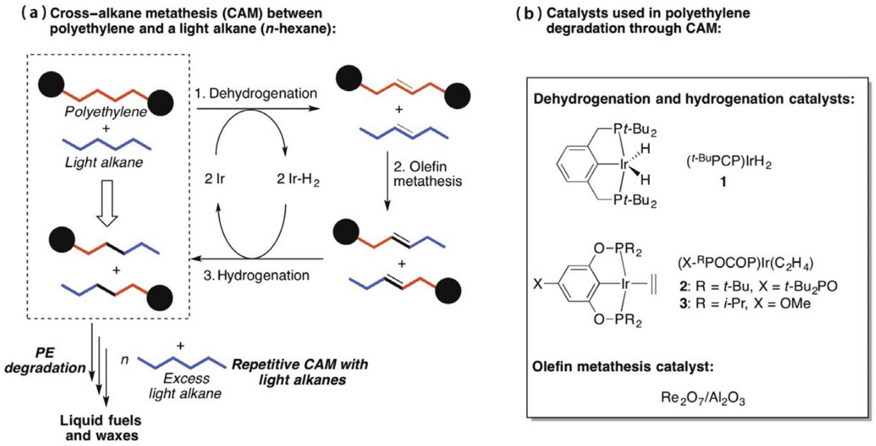

Fig. 10. (a) Degradation of PE through catalytic CAM with light alkanes. (b) Structures of the dehydrogenation and olefin metathesis catalysts. Reproduced from Ref. [96]with permission from AAAS.

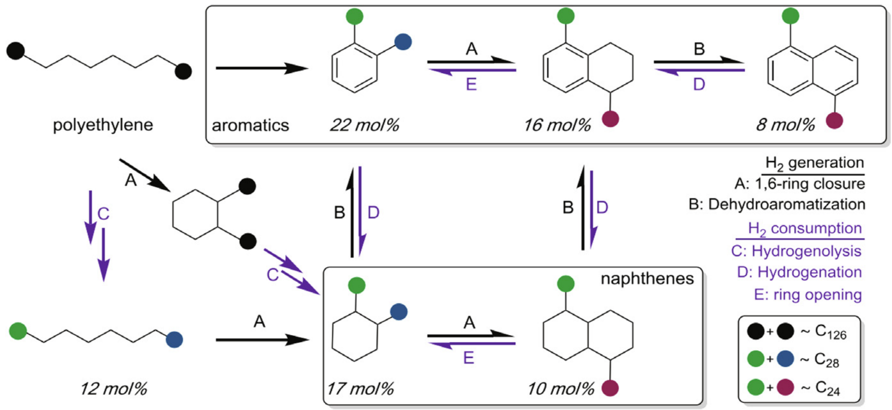

Fig. 11. Overall PE conversion to alkylaromatics and alkylnaphthenes, and proposed mechanism of tandem polyethylene hydrogenolysis/aromatization via dehydrocyclization. Reproduced from Ref. [98]with permission from AAAS.

more, the long chain alkylaromatics could be further sulfonated to form anionic surfactants and are of higher value compared to saturated alkane products in the most hydrogenolysis systems as well as the above CAM case.

3.5.2. Tandem process on polymers containing aromatic group

As an alternative of depolymerizaton of aromatic plastic waste into value-added monomers, directly obtaining value-added polymeric materials from plastic waste could be achieved by a tandem process based on solvolysis and following reaction. Jones et al. proposed a one-pot strategy to form high-value PSUs from the depolymerization of PCs in the presence of a carbonate salt and bis(aryl fluoride)s (Fig. 12) [99]. The generated PSUs could be separated according to their thermal properties. The carbonate salt played two important catalytic roles in the decomposition of PC and formation of PSUs products: decomposition of the PCs by nucleophilic attack as well as promoted the reaction of phenolate dimers with the aryl fluorides. This method represents a convenient and direct way to transform one class of polymer to another.

Recently, Wang's group developed the H_2-free PET conversion to benzene, toluene and xylene (BTX) using hidden structural hydrogen from ethylene glycol as the initial depolymerized product over Ru/Nb_2O_5 catalyst. The whole process including hydrolysis, reforming and hydrogenolysis/decarboxylation. Compared with $Ru/NiAl_2O_4$, Ru/Nb_2O_5 exhibited superior hydrogenolysis because of the strong oxygen affinity of NbO_x species for C–O bond activation, and showed poorer decarboxylation performance attributed to less metallic zero-valent Ru species on Ru/Nb_2O_5. These two key roles contributed to the distinct selectivity to alkyl aromatics among BTX, providing new options in the conversion of PET waste [100].

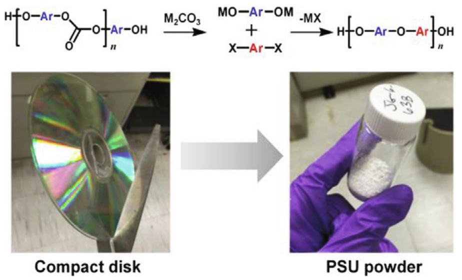

Fig. 12. Formation of poly (aryl ether) via the repurposing of poly(carbonate)s in the presence of a carbonate salt, and compact disk (left) and PSU powder (right) are shown before and after the depolymerization/repolymerization process. Reproduced from Ref. [99] with permission from National Academy of Sciences.

3.5.3. Tandem process on other commodity plastics

Lu et al. demonstrated an interesting strategy to reduce hydrogen carbonate to formate with PVC as a hydrogen source in high-temperature water (Fig. 13). Complete dechlorination of PVC at 300 °C was achieved accompany with a 16% yield of formate based on hydrogen carbonate. The proposed mechanism suggested that the substitution reaction of chlorine in PVC by OH^- to form CH–OH group carried out initially, and then the formate formed via a redox reaction between the CH–OH group and HCO_3^-. Moreover, the selection of water as the reaction medium played a key role in the reduction of HCO_3 to formate using PVC [101].

Recently, Tian et al. reported for the first time on a new one-pot catalytic process to convert PLA into alanine (Fig. 14). Using a Ru/TiO_2 catalyst with a simple ammonia solution heating treatment, up to 77% yield of alanine could be achieved at 140 °C. It has been verified that lactamide and ammonium lactate are the initial intermediates by ammonolysis and following hydrolysis, respectively. Metal catalysts play a key role in the activation and subsequent amineization of α-H of ammonium lactate, and the reaction follows the dehydro-amine-rehydrogenation route. The efficiency of this process was assessed using 5.0 g commercial PLA straw (approximately 83% PLA) as reactant and obtained 3.0 g of pure alanine. The new process demonstrated here could expand the application of PLA waste and inspire new upcycling strategies for different plastic wastes [102].

3.6. Carbonization

Carbonization reaction refers to the conversion of organic substances into carbon material by cracking or dry distillation, which is widely used to convert various waste plastic polymers into valuable nanocarbon materials. The prepared carbon nanomaterials (CNMs) contains carbon dots (CDs), carbon nanotubes (CNTs), graphene and carbon nanosheet or nanohorns, which could be further used as a series of energy storage materials of great performance, such as supercapacitors, anode materials of lithium-ion batteries (LIBs) and materials of other energy-related applications [103].

3.6.1. Carbonization of polyolefins

Recently, Zhang et al. synthesized a hierarchical porous carbon (HPC) from LDPE through an autogenic pressure carbonization-KOH activation process (Fig. 15) [104]. The obtained HPC possessed a micrometer-scale carbon sphere morphology with hierarchical pores, exhibiting an excellent capacitive performance with a speci-

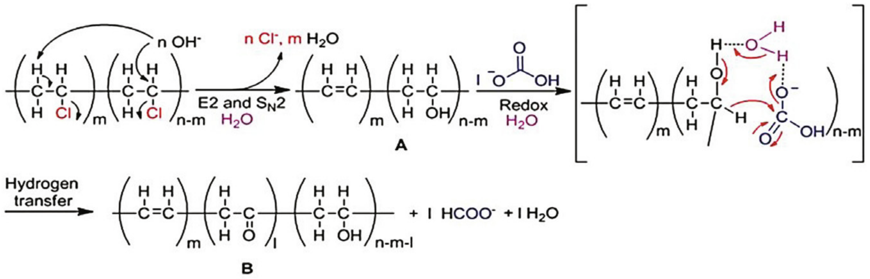

Fig. 13. Proposed mechanism of HCO_3^- reduction to formate using PVC. Reproduced from Ref. [101] with permission from Royal Society of Chemistry.

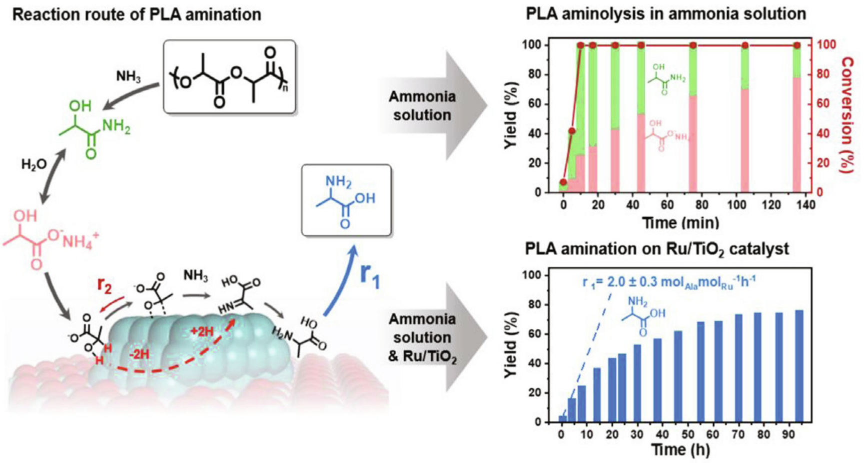

Fig. 14. Catalytic transformation of PLA to alanine, and the proposed reaction mechanism of PLA amination on a Ru/TiO_2 catalyst in ammonia solution. Reproduced from Ref. [102] with permission from American Chemical Society.

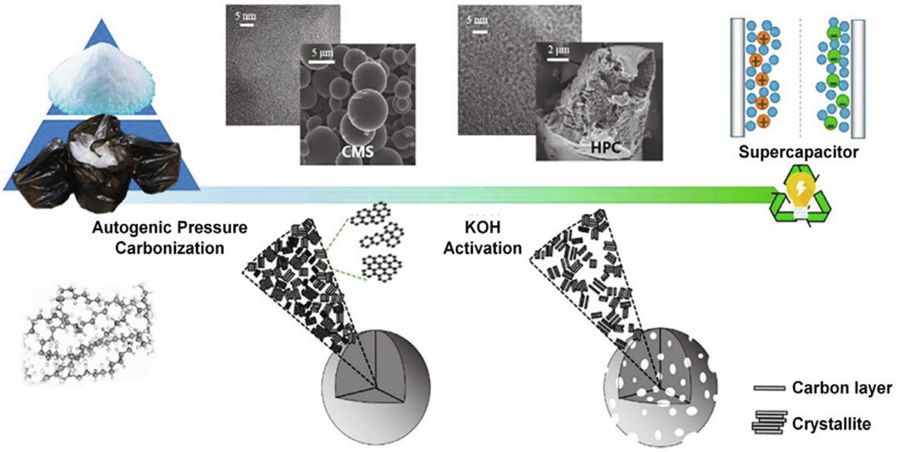

Fig. 15. Schematic illustration of the synthesis of HPC through autogenic pressure carbonization followed by KOH activation. Reproduced from Ref. [104] with permission from American Chemical Society.

fic capacitance of 355 F g^{-1} at a current density of 0.2 A g^{-1} in 6 M KOH electrolyte when acting as supercapacitor electrode material. Owing to the porous structure and good conductivity, the carbon materials prepared by the decomposition and carbonization of waste polymers can also be applied in LIBs [105-107]. Pol et al. prepared the spherical carbon particles and carbon nanotubes by the thermal decomposition of PE, and the obtained materials can be used as negative electrodes in lithium electrochemical cells, showing a stable capacity of approximately 240 mA h g^{-1} between 1.5 V and 5 mV for hundreds of cycles [108].

3.6.2. Carbonization of polymers containing aromatic group

Yang and coworkers synthesized porous carbon materials with hierarchical macro-meso-microporous structure by carbonization of PS at 900 °C, and the products acting as high-rate anode electrodes in LIBs showed a stable capacity of 410 mA h g^{-1} for over 100 cycles [109]. Elessawy et al. synthesized functionalized magnetic fullerene nanocomposites (FMFNc) via one-step catalytic thermal decomposition of PET using ferrocene as a catalyst and precursor of magnetite nanoparticles at 800 °C for 20 h [110]. The obtained FMFNc with mesopores and micropore volumes had relatively high surface area, and showed a good performance in the adsorption of cationic methylene blue dye and anionic acid blue 25 dye.

3.6.3. Carbonization of PVC and other halogen-containing polymers

Heteroatom-containing polymers inevitably exist in waste plastics, and how to convert the halogen-containing plastics into valuable products is challenging [111,112]. Cheng et al. selected PVC as a carbon source to prepare nanoporous carbon by a facile and efficient template carbonization approach, in which $Mg(OH)_2$ served as a hard template [113]. The nanoporous carbon product exhibited large BET surface area of 958.6 $m^2\ g^{-1}$ and high pore volume of 3.56 $cm^3\ g^{-1}$. By incorporating with MnOx nanoparticles, the electrochemical performance of the product was further improved with the specific capacitance elevated up to 751.5 F g^{-1} at 1.0 Ag^{-1}. This approach provides a proper method to dispose waste PVC into nanoporous carbon, which can be applied in supercapacitor. Jiang et al. presented a $CaCO_3$-assisted template carbonization method to transform the polytetrafluoroethylene (PTFE) waste into nanoporous carbon spheres [114]. The resulting carbon materials displayed 179.9 F g^{-1} of specific capacitance when measured at 1 A g^{-1} using 6 M KOH as electrolyte, and the specific capacitance can be improved to be 237.8F g^{-1} when adding $CO(NH_2)_2$ as a nitrogen source to dope carbon materials, showing good behaviors for supercapacitor application.

3.6.4. Carbonization of mixed plastics

Different from most other transformations to convert a certain kind of polymer, the carbonization of polymers provides an attractive avenue to upcycle plastic waste mixture into a high value-added product, which is viewed as its biggest advantage. For example, the mixed PP/PE/PS were used to synthesized CNTs by controlling the degradation behavior of polymers and the vapor depositions of degradation products following a strategy of combined catalysts to regulate the carbonization proposed by Tang et al. They applied the nanosized carbon black (CB) to catalyze the degradation of mixed plastics to produce more aromatic compounds, while Ni particles together with CB subsequently catalyze the dehydrogenation and reassembling of the products [115]. The CNTs were obtained with high yields (31.6 wt%) and relatively larger BET specific surface areas (447 m^2/g), and displayed a high specific capacitance of 190 F g^{-1} in 6 M KOH electrolyte at the scan rate of 5 mV s^{-1} used for the electrode of supercapacitor [116].

Recently, Min et al. established a general template based on magnesium oxide and iron (III) acetylacetonate to carbonize the plastic waste (PS, PP, PE, PVC, and their mixture) into hollow carbon sphere/porous flakes (Fig. 16) [117]. Benefiting from their 3D structured, the products exhibited ideal electrochemical performance in LIBs (802 mA h g^{-1} after 500 cycles at 0.5 A g^{-1}).

Compared with conventional heating, the energy provided by microwaves can be better absorbed by polymer chains, thereby significantly improves the degradation efficiency. Microwave-assisted catalytic polyethylene degradation over a FeAlOx catalyst was published by Jie et al [118]. In a very short time (30–90 s), commercial plastics (HDPE, PP, PS) were converted to multi-walled carbon nanotubes with a much higher carbon yield of 1560 mg $g^{-1}_{catalyst}g^{-1}_{plastic}$. In addition, hydrogen was prepared with a yield of 55.6 mmol $g^{-1}_{plastic}$ in one step. Recently, Xu et al. developed a carbon foam microwave plasma process which can generate high surface temperatures exceeding ~3000 K locally in a N_2 atmosphere [119]. This process can transform real plastics (PP, fabrics, mask wastes, etc.) into gas fuels containing H_2 (gas yield of up to 43 vol%) and useful H-bearing small gas molecules (CH_4, C_2H_4, C_3H_6 et al), with excess high-quality graphitic carbon grafted on the pre-existing carbon form as a self-perpetuating catalyst, which can be finally buried as artificial "coal". Meanwhile, Flash Joule Heating (FJH) is also an efficient heating method. Luong and co-workers used FJH technology to convert different cheap carbon sources such as coal, petroleum coke, biochar, carbon black, waste food, rubber tires, and mixed plastic waste into gram scale of flash graphene (FG) within one second [120]. In subsequent work, they specifically explored the conversion of various plastic waste (PET, PVC, HDPE, LDPE, PP, PS) into FG [121]. The higher the stability of the plastic, the higher the FG yield, with the maximum yield up to 25 wt% for PE and PP.

4. Emerging electrochemical and photocatalysis path

Beyond the newly-developed and more relative thermochemical routes on waste plastics recycling and upcycling as mentioned above, there are a series of appealing and promising technologies emerged, such as mechano-chemical, biotechnological, electrolysis and photocatalysis paths using alternative energy sources. Several reviews have been published by Weckhusyen's group [5], Beckham's group [122] and Soo's group [123,124] recently, focusing on these non-thermal processes for plastic deconstruction and upcycling. Here, we listed some representative new cases on commodity plastic upcycling by electrochemical or photocatalysis way at mild conditions, even at ambient temperatures and pressures for some cases, demonstrating the advantages of the new strategies compared with their thermochemical counterparts as well as their challenging for a scalable application.

4.1. Electrocatalytic upcycling of commodity plastics

Electrolysis, generally conducted in aqueous solvents at ambient temperatures and pressures, is viewed as a clean and sustainable alternative path to thermal degradation of polymers. There has been work on applying electrocatalysis to transform lignin into chemical feedstock [125,126]. However, there has been little research on electrocatalytic conversion of commodity plastic, possibly due to the low solubility of most plastic in aqueous solutions.

Last year, Myren et al. carried out the electrochemical depolymerization of PET in a methanol/water system with 0.1 M NaCl as a supporting electrolyte within an H-cell [127]. At a potential of −2.2 V, 17% yield of terephthalic acid (PTA) was produced via electrocatalytic hydrolysis at 21 °C, but CO_2 was formed as an undesirable byproduct at the same time. Recently, Zhou et al. demonstrated a remarkable electrocatalytic strategy to convert PET into high valued commercial chemicals of potassium diformate and PTA using a dual-function $CoNi_{0.25}P$ electrocatalyst in a potassium hydroxide electrolyte solution, and to produce a valuable by-product, hydrogen, at the same time (Fig. 17a) [128]. It could be viewed as a tandem process of hydrolysis of PET and electrocatalytic HER and EG oxidation. The EG intermediate formed by KOH-catalyzed PET hydrolysis, underwent C–C cleavage to formate with a high selectively (>90%) over anodic CoNi0.25P catalyst in electrolyzer, accompany with the H_2 formation over the same catalyst at cathode. This research may open a way to profitably produce value-added commodity chemicals and clean H_2 from PET, demonstrating a sustainable way to implement upcycling of plastic waste.

4.2. Photocatalytic upcycling of commodity plastics

Photocatalytic upcycling of commodity plastics is appealing and viewed as a more sustainable approach since sunlight is used as the only energy source. Unlike photodegradation, photocatalytic upcycling strategy can potentially convert plastic waste into value-added products, such as H_2, short chain hydrocarbon fuels and value-added oxygenates. In 2018, Reisner and co-workers

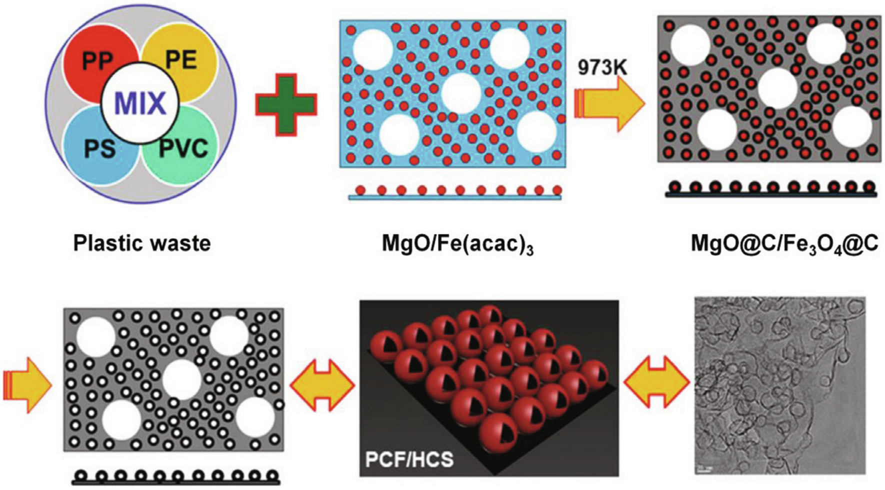

Fig. 16. Schematic illustration of the formation of a 3D hollow carbon sphere/porous carbon flake hybrid nanostructure from carbonization of plastic waste. Reproduced from Ref. [117] with permission from Royal Society of Chemistry.

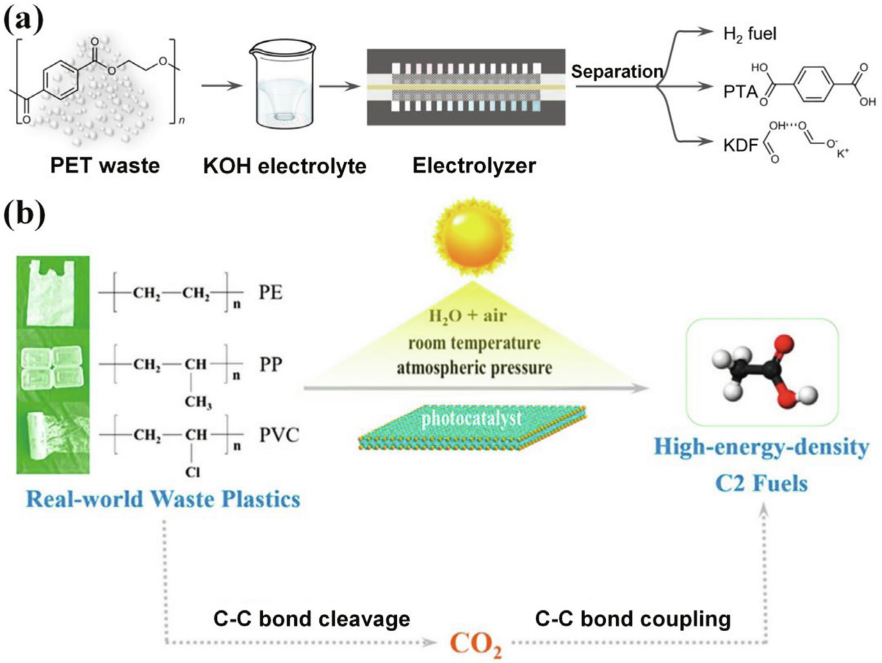

Fig. 17. (a) Electrocatalytic PET upcycling to commodity chemicals and H_2 fuel. (b) Converting various waste plastics into C2 fuels over a photocatalyst by a designed two-step pathway under simulated natural environment conditions. Reproduced from Refs. [128,132] with permission from Springer Nature and Wiley-VCH.

developed a photoreforming strategy to produce H_2 from various types of plastics by CdS quantum dots without using the sacrificial reagents [129]. Various small oxygenates are formed alongside the H_2 generation, which could potentially be recovered and add benefit to the process, instead of just CO_2 formation. The catalytic system was further improved using a Cd-free carbon nitride / nickel phosphide catalyst to avoid using the toxic Cd materials [130,131]; however, it still suffers from low productivity and poor selectivity towards a single high-value oxidation product.

A real problem to face in most upcycling path developed is the variable composition of polymers in the plastic waste feed, which cannot be easily separated and would further hamper their valorization based on the path designed for a certain kind of polymetric material. Recently, Jiao et al. firstly reported a new photocatalytic path to selective conversion of various waste plastics, including PE, PP, PVC as well as real disposable food containers and food wrap films, into liquid products of acetic acid without using sacrificial agent [132]. It was achieved by a universal photoinduced sequential C–C bond cleavage and coupling pathway over the single-unit-cell thick Nb_2O_5 layers under simulated natural environment conditions (Fig. 17b). Although the efficiency of C2 fuels production is difficult to implement on a large scale, it is inspiring more efficient photocatalytic tandem systems to be involved in upgrading of real waste plastics.

5. Conclusion and outlook

A variety of thermochemical plastic recycling paths have been reported in the last decades, while pyrolysis is mostly used undoubtedly [3]. However, considering that thermal or catalytic pyrolysis always takes place at relatively higher temperature (>400 °C) through a random scission mechanism, the obtained products are usually a mixture of gas, liquid fuels, and waxes. The newly developed ways on chemical recycling or upcycling of commodity plastic waste, operative under mild condition, are aiming for a more selective process to either achieve monomer recovery on a large scale or transform the virgin polymers into fuels, high-valued chemicals or materials. According to the recent developments on recycling and upcycling methods on existing commodity plastics, three directions are proposed to be of particular interest in the field.

Firstly, to meet economic requirements, the key of a useful newly-developed method on plastic upcycling should be focus on efficiently forming recovered products of great performance and sizable market value. Beyond fuels and energy storage materials, conversion of commodity plastics into high-value chemicals and functional materials is also a good option in the plastic upcycling field, such as adsorption material, adhesive, and oil–water separation material [133-136], etc. This could be very challenging for some kind of commodity plastics, like polyolefin. Especially, the recycled or upcycled materials must possess good mechanical and thermal properties to meet the requirement as drop-in options to the polymetric materials currently used. To achieve the goal, new polymeric materials are encouraged to be designed with recyclability from the beginning, which is a determinant factor as important as good mechanical and thermal properties. With a sustainable supply of the new monomers and a decreasing cost comparable to the usual polymers, these new polymers may replace some commodity plastic with poor recyclability long-term. Recently, Haussler et al. used long chain aliphatic acid and diol as monomers to synthesize polyester material, which not only has similar properties to polyethylene, but also can be recycled in solvent with a recovery rate of 96%. Moreover, the materials' high-performance properties (like those of high-density polyethylene) of the recycled polymer are fully retained [14].

Another promising direction is using real plastic waste as feedstock rather than model polymetric compounds, as a clear mismatch in the research could hinder their further application. Due to the high cost involved in the separation of different compound or elimination of impurities within the plastic waste, a commercial success of these developed catalytic upcycling paths may depend not only on the value added of the products but also on the ability to be well used with the high contaminant contents in the feed. The feedstock problem is becoming one of the biggest bottlenecks in plastic recycling. How to employ a recycling or upcyling strategy that works well for post-consumer plastic waste of mixed kinds of polymers with impurities? How to deal with the additives such as compatibilizers or flame retardants during the recycling or upcycling process? In the existing chemical upcycling methods, there are a few cases using hydrogenolysis, carbonization and photoreforming strategy to convert mixed commodity plastic into hydrocarbon fuels, carbon materials and clean H_2, demonstrating their potential to deal with large scale of real plastic waste feedstock. In addition, there might be some smart cases in the future to use these additives (inorganic fillers or organic flame retardants within the polymer) as catalysts or reactants to convert commodity plastics into higher-valued products.

Last but not least, it is very important to design new or improved catalysts of higher stability during the process or easily to be regenerated. Noble metal catalysts (Pt, Ru et al) are less prone to aggregation at reaction condition compared with non-noble metal catalyst (Fe, Co, Ni et al), and are widely used in traditional pyrolysis [137] and new chemical upcycling paths [35,36,98]. However, considering their high price and the amount of waste plastic to deal with, developing precious metal-substituted nanocatalysts are of particular interest in this field. Metal carbides, which show catalytic activity similar to those of noble metals in various catalytic reactions, have attracted much attention both as catalyst support [138-140] or catalyst directly used [141-144]. Especially, supported molybdenum or tungsten carbide catalysts have been used for the biomass conversion to high-value small-molecular chemicals [145-147] and other biomass product upgrading conversion [148,149]. Supported iron catalysts are also of high interest. Previously, carbon supported iron catalyst show similar catalytic activity to its platinum counterparts in conversion of polyethylene to aromatics at 450 °C in Ar [150]. Recently, graphene confined single iron atom structures ($Fe\text{-}N_4$) has shown as efficient active site for catalyzing C–H oxidation to oxidize benzene to phenol [151] or to directly convert methane to C1 oxygenated products [152] at room temperature, which inspires its future application to polyolefin upgrading. Finally, catalytic pyrolysis of waste polyethyelene into aromatics was recently achieved by a non-metal H_3PO_4 activated carbon catalyst [153]. Although the activity and selectivity of these new non-noble metal or non-metal catalysts are possibly not as good as traditional zeolites or supported noble catalysts at hand, there is a bright future for chemists to find new and improved ones for chemical upcycling of various polymeric materials.

In the end, it is expected to see more sustainable, creative and highly efficient approaches to chemical recycling and upcycling of plastic waste in the future, and further expand this young but fast-growing field.

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.

Acknowledgments

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