Retrieving back plastic wastes for conversion to value added petrochemicals: opportunities, challenges and outlooks

Manish Kumar [a], Shiv Bolan [b,c], Lokesh P. Padhye [d], Muxina Konarova [e], Shin Ying Foong [f], Su Shiung Lam [f,g], Stuart Wagland [h], Runzi Cao [i], Yang Li [i], Nuno Batalha [j], Mohamed Ahmed [d], Ashok Pandey [k,l,m], Kadambot H.M. Siddique [b,c], Hailong Wang [n], Jörg Rinklebe [o], Nanthi Bolan [b,c,*]

[a] School of Environmental Sciences, Jawaharlal Nehru University, New Delhi 110067, India
[b] UWA School of Agriculture and Environment, The University of Western Australia, Perth, WA 6009, Australia
[c] The UWA Institute of Agriculture, The University of Western Australia, Perth, WA 6009, Australia
[d] Department of Civil and Environmental Engineering, Faculty of Engineering, The University of Auckland, Auckland, 1010, New Zealand
[e] School of Chemical Engineering, The University of Queensland, Brisbane, Australia
[f] Higher Institution Centre of Excellence (HICoE), Institute of Tropical Aquaculture and Fisheries (AKUATROP), Universiti Malaysia Terengganu, 21030 Kuala Nerus, Terengganu, Malaysia
[g] Center for Transdisciplinary Research, Saveetha Institute of Medical and Technical Sciences, Saveetha University, Chennai, India
[h] School of Water, Energy and Environment, Cranfield University, Cranfield, Bedfordshire MK43 0AL, UK
[i] Key Laboratory of Water and Sediment Sciences of Ministry of Education, State Key Laboratory of Water Environment Simulation, School of Environment, Beijing Normal University, Beijing 100875, China
[j] Institut de Recherches sur la Catalyse et l'Environnement de Lyon (IRCELYON), UMR5256 CNRS-UCB Lyon 1, 2 Avenue Albert Einstein, 69626, Villeurbanne Cedex, France
[k] Centre for Innovation and Translational Research, CSIR-Indian Institute of Toxicology Research, Lucknow 226 001, India
[l] Sustainability Cluster, School of Engineering, University of Petroleum and Energy Studies, Dehradun 248 007, Uttarakhand, India
[m] Centre for Energy and Environmental Sustainability, Lucknow 226 029, Uttar Pradesh, India
[n] Biochar Engineering Technology Research Center of Guangdong Province, School of Environmental and Chemical Engineering, Foshan University, Foshan, Guangdong 528000, China
[o] University of Wuppertal, School of Architecture and Civil Engineering, Institute of Foundation Engineering, Water- and Waste-Management, Laboratory of Soil- and Groundwater-Management, Pauluskirchstraße 7, 42285 Wuppertal, Germany

HIGHLIGHTS

- Petrochemicals are the primary feedstocks for the manufacture of plastics.
- The plastic wastes can be retrieved back to value added petrochemicals.
- Plastic-based petrochemicals include crude oil, gases, hydrogen and aromatic char.
- Plastic conversion technologies include thermochemical, catalytic and chemolysis.
- Environmental impacts and economic of the processes need further evaluation.

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ABSTRACT

Plastic production and its unplanned management and disposal, has been shown to pollute terrestrial, aquatic, and atmospheric environments. Petroleum-derived plastics do not decompose and tend to persist in the surrounding environment for longer time. Plastics can be ingested and accumulate into the tissues of both terrestrial and aquatic animals, which can impede their growth and development. Petrochemicals are the primary feedstocks for the manufacture of plastics. The plastic wastes can be retrieved back for conversion to value added petrochemicals including aromatic char, hydrogen, synthesis gas, and bio-crude oil using various technologies including thermochemical, catalytic conversion and chemolysis. This review focusses on technologies, opportunities, challenges and outlooks of retrieving back plastic wastes for conversion to value added petrochemicals.

* Corresponding author at: UWA School of Agriculture and Environment, The University of Western Australia, Perth, WA, 6001, Australia.
E-mail address: nanthi.bolan@uwa.edu.au (N. Bolan).

The review also explores both the technical and management approaches for conversion of plastic wastes to petrochemicals in regard to commercial feasibility, and economic and environmental sustainability. Further, this review work provides a detailed discussion on opportunities and challenges associated with recent thermochemical and catalytic conversion technologies adopted for retrieving plastic waste to fuels and chemicals. The review also recommends prospects for future research to improve the processes and cost-efficiency of promising technologies for conversion of plastic wastes to petrochemicals. It is envisioned that this review would overcomes the knowledge gaps on conversion technologies and further contribute in emerging sustainable approaches for exploiting plastic wastes for value-added products.

1. Introduction

More than 200 million tonnes of plastic are consumed yearly worldwide, making plastic the third most desirable petroleum product. The primary component of plastics is a polymer, which is derived from both non-renewable and renewable sources [1]. The non-renewable sources of polymers applied for plastic production are produced from crude oil, natural gas, and coal, while the renewable sources include plant- and microbial-derived polysaccharides/polymers [2–4]. The first step in the processing of plastic from non-renewable sources is cracking, which turns either the crude oil-based product (naptha) or the natural gas-based product (ethane) into ethylene, a starting point for a variety of chemical products including polymers [5]. There are two major sources of renewable components of plastics that include polylactic acid (PLA) derived from plant-based polysaccharides (i.e., starch and sugars) [6], and polyhydroxyalkanoates (PHAs) derived from microorganisms [7].

Plastic production, as well as its inappropriate disposal and management, adversely impact marine, terrestrial, and atmospheric ecosystems [8]. Synthetic plastic derived from petroleum industries do not easily biodegrade and persist in the environment for longer time [9]. Plastics in the environment can occur in a range of physical size fractions covering macro-, micro- and nano-plastics [10]. Although the term 'microplastics' is used extensively to represent plastic contamination, while the term 'particulate plastics' used to represent the range of plastic contamination in all the environmental components which include air, water, and soil [11,12]. These particulate plastics present in aquatic and terrestrial ecosystems are becoming a major source of pollutants [13].

There are two key routes/sources of particulate plastics in terrestrial and aquatic ecosystem: (i) primary particulate plastics are produced as a direct result of human activity using materials derived from plastic (e.g., microbeads in cosmetics); and (ii) secondary particulate plastics are break down products of larger plastic fragments [8]. These both types of plastic fragments persist in the terrestrial (i.e., soil) and aquatic (i.e., marine) environments. Plastics can be entered and ingested into the tissues of marine and terrestrial living well-being because they do not decompose easily [4]. The majority of the particulate plastics that are still in the aquatic environment come from terrestrial environment and were carried by the means of soil erosion or sediment transfer [14,15].

Many countries (China, United Kingdom (UK), Germany) have various adopted technologies such as sanitary landfilling, composting, and incineration to manage the solid waste contains plastics [16]. However, generation of secondary pollutants, such as particulate plastics and gaseous emission are major hurdle in its wider application, considering its environmental and health impacts [17]. Hence, upcycling is considered as more effective approach in management and valorization of plastic waste. Numerous research conducted recently to investigate the viability of adopted upcycling methods on a wider scale to transform plastic waste into products with better economic value [18–20]. The objective of utilizing plastic waste s to synthesize novel value-added products by adapting a circular economy approach has obtained the attention of investigators worldwide [21,22]. Closing the loop of a circular economy requires the development of effective recycling and upcycling techniques that convert plastic waste into other useful and less perilous products [17].

Petrochemicals are the primary feedstocks for the manufacture of plastics. The plastic wastes can be retrieved back for conversion to value added petrochemicals including bio-crude oil [23], synthesis gas [24], hydrogen [25] and aromatics [26] using various technologies including thermochemical [17], catalytic conversion [19], chemical recycling, and chemolysis [27]. Although, several previous researches and reviews have highlighted the utilization of plastic waste as a potential feedstock for synthesis of value-added fuels and chemicals [17,28–32]. For instance, Zeng et al. [26] investigated microwave catalytic co-pyrolysis of waste cooking oil and low-density polyethylene (LDPE) to yield monocyclic aromatic hydrocarbons and effect of various catalysts and pyrolysis conditions. Putra et al. [17] comprehensively discussed the application of microwave pyrolysis as a viable and sustainable plastic waste utilization technique. Barnard et al. [28] broadly discussed the application of chemocatalytic technologies in depolymerisation of polyethylene terephthalate (PET). Similarly, Rahimi and García, [31] critically discussed the chemical recycling methods of plastic waste to fabrication of new plastic products. Still, as per the author's best knowledge, a comprehensive discussion on various plastic waste management and upcycling technologies for synthesis of value-added chemicals and fuels have not been thoroughly summarized at one place in the earlier published literature.

Hence, this review focusses on technologies, opportunities, challenges and outlooks of retrieving back plastic wastes for conversion to value-added petrochemicals. An effort has been made to censoriously analyse the previous published literature on the conversion of plastic wastes into petrochemicals using several conversion technologies. Specifically, we determine (1) novel insights on environmentally sustainability of plastic wastes valorization to petrochemicals (2) economic feasibility as well as environmental impacts imposed by the plastic waste upcycling technologies (3) the knowledge gaps in the literature in regards to current commercialized technologies for large scale conversion of plastic wastes to value added petrochemicals. Moreover, increased knowledge about conversion of plastic wastes to petrochemicals will assist the plastics and petrochemical industries to discover the opportunities for beneficial upcycling of plastic wastes. The prospect for future research in the use of various technologies for conversion of plastic wastes into petroleum chemicals are also proposed.

2. Review methodology

Understanding what has been described on a subject/topic and finding issues or questions that need further research are both important outcomes of literature reviews. The synthesis of quantitative and qualitative evidence from literary data can be done by a number of routes, including frequency analysis, meta-analysis, grounded theory, narrative analysis, and meta-ethnography. Quantitative analysis is helpful when conducting a descriptive review, and qualitative investigation is effective when conducting narrative reviews. We conducted a quantitative investigation of literature data for this study. By using the keywords "Plastic wastes," "Petrochemicals," "Pyrolysis," "Gasification," "Liquefaction," "Chemolysis," "Catalysis," and "Plastic waste life cycle analysis", appropriate literature was examined through the databases of Web of Science, ScienceDirect, Scopus, Google Scholar, and other different sources. Nevertheless, it was surprising that majority of literatures dealing with plastic pollution, microplastic, their environmental and

health impacts, reuse and recycling of plastic instead of upcycling and/or thermochemical/catalytic conversion of plastic waste. Therefore, we manually segregated and mainly included those literatures which were mostly covered the information related to plastic upcycling along with management.

Hence, this review tries to present the most recent findings from plastic management and valorisation technologies. This review tries to close knowledge gaps about the environmental effects of these technologies by presenting the most recent, cutting-edge research on solutions for managing and valorizing plastic waste. This rigorous analysis will assist in creating a sustainable method for managing plastic waste generated by anthropogenic activity. This knowledge is essential for the creation of new measures that lessen the adverse impact imposed by the plastic waste and its particulates on living well-being and the environment. This literature analysis delivers an outline for recognizing important research domains to fill up the research gaps on the management of plastic waste and its efficient use. Furthermore, the industrial sectors and ultimately human society will profit from increased knowledge regarding the production of valuable products using plastic waste as a feedstock. The handling of plastic waste and its associated contaminates sustainably are also suggested as future research outlooks.

3. Manufacturing of plastics

3.1. Polymer sources and polymerisation reactions

There are two major sources of polymers used in plastic manufacture, which include non-renewable (i.e., synthetic) and renewable (i.e., natural) sources [1,6]. Synthetic polymers are produced from crude oil, natural gas, and coal, and some of the synthetic polymers include teflon, polyethylene (PE), polyester, and nylon. Natural polymers are derived from plants and microorganisms, and some of the natural polymers include rayon and polyester [33,34]. It is imperative to make a note on similar polymer products (e.g., polyester) used for plastic manufacture can be derived from both natural and synthetic sources [35]. Polymers, which are the primary components of plastics, are formed through polymerisation reactions of monomers. Polymerisation requires either the processes of addition or condensation reactions to occur [36], which involve three basic steps: initiation, propagation, and termination. For the process of addition polymerisation, the last 'mer' in the chain-sequence becomes the binding site for the monomers.

The addition polymerisation process can result in the formation of PE, polystyrene (PS), and acrylic plastics. The thermoplastic properties of polymers derived from the addition polymerisation process allow the heating to soften them, and then the process of hardening takes place when they are cooled [37]. During condensation polymerisation, a smaller molecule such as water (H_2O) or alcohol is discharged as the monomers adhere. In order for the chemical reactions to be continue during condensation polymerisation, the elimination of undesirable by products is a requirement for the synthesis of useful chemicals [2]. The strength and endurance of the resulting plastic is the primary distinction between addition and condensation polymerisation reactions. Generally, condensation polymers are relatively more readily degradable compared to addition polymers. Monomers can be combined in diverse configurations to synthesise plastic resins with variable properties and characteristics [38]. Combining the same monomers together produces a homogenous polymer referred to as a homopolymer, while, co-polymers refer to plastics where multiple monomers are used [2].

3.2. Types of plastics

According to the origin of polymers, plastics can be grouped into natural (derived from animals and plants) and synthetic (artificially synthesised by chemical processes) [1,6]. For example, cellulose polymer is naturally occurring and used for making stick y tape, whereas nylon is a synthetic polymer made in a factory. Because the majority of

plastics are synthetic, the groupings based on stability during heating and the sources of polymers are more frequently used in the applications of plastics [39]. The behaviour of plastics when heated allows the differentiation into two broad categories, namely: thermoplastics and thermosets.

Thermoplastics make up the majority of plastics with approximately 92 % of plastics falling into this category [40]. Thermoplastic polymers when subjected to heat soften due to weak secondary bonding forces that hold together the molecules of the polymer plastic. When reverted to ambient temperature these polymers return to their original orientation; thus, when softened, thermoplastic can be shaped by extrusion, molding, or pressing processes allowing its adaptability for a vast array of applications [41]. Examples of thermoplastics include PE or polythene (plastic bottles and sheets), PS (packaging material), polypropylene (PP) (plastic ropes), polyvinylchloride (PVC) (toys and drainage pipes), polycarbonate (PC) (plastic windows and car headlamps), and polyamides (PAs) (nylon purposed for stockings and swimming wear).

A thermoset is a plastic polymer that solidifies or "sets" irreversibly when heated or cured [42]. Larger polymer chains are used to make thermoset plastics than thermoplastics. During the initial synthesis operation, dense assemblies with strong cross-links bind long molecular chains together via heating and compression [43]. These connections between carbon atoms lattice together to form two and three-dimensional frames as opposed to one-dimensional chains. These thermoset plastics are not meltable and hence have specific values for their durability and strength. Common examples of thermosets include melamine (hard plastic crockery), polytetrafluoroethylene (PTFE) (non-stick coatings), polyurethane (PUR) (insulating material in buildings), and epoxy resin [2].

More than 90 % global plastic products are derived from these non-renewable sources, however, in recent years, production of plastics/bioplastics are gaining attentions from renewable sources. The term 'bioplastic' broadly represents plastic substances that are derived wholly or in part from biomass-based feedstock rather than petroleum-based feedstock material [44,45]. Most of the bioplastics are biodegradable, and there are three broad descriptors of bioplastics [1]. (i) Bio-based plastics, which include all plastic materials, derived wholly or in part from plant-based material. Cellulose and starch are the two most common renewable feedstocks utilized to synthesise bioplastics. (ii) Degradable plastics, which include only those that degrade within a relatively short duration. Bioplastics that do not degrade within a short period are sometimes called 'durable plastic'. (iii) Compostable plastics, which include those that undergo the process of biological decomposition in a specialised compost site and break down to CO_2, H_2O, inorganic mixtures, and biomass, at a rate similar with other identified compostables (e.g., cellulose).

3.3. Processing of plastics

Plastic polymers known as resins are utilised in the synthesis of various plastic-derived products. Some of the major plastic processing

Table 1
General characteristics of plastic processing methods [2].

Process	Characteristics
Injection	Thin walls, high production speeds, pricey tooling, complex shapes of diverse sizes, and excellent dimensional accuracy.
Extrusion	Wide tolerance, continuous and consistently solid or hollow cross sections, high production rates, reasonably affordable tooling.
Calendering	Continuous process; high output; capacity to handle low melt strength; well-maintained thickness; smoothed surface.
Spinnerate	Fine fibres; high precision and high production cost; relatively high tooling and maintenance costs
Blow molding	High production rates, various-sized bottles with hollow, thin walls, and relatively inexpensive tooling.

and shaping methods include extrusion, injection, blow molding, calendering, and spinnerating [45,46] (Table 1).

Plastic granules, pellets/powder are loaded into a hopper before being fed into a heated extruder where they are transported by the mechanism of a continually rotating screw in the continuous extrusion process [47]. The heat from the extruder wall and mechanical action of the screw combine to melt the plastic. Following this procedure, the molten plastic is extruded through a small hole known as a die to shape the final product. In injection molding, which is not a continuous process, the plastic material is fed into a hopper, and an extruder screw advances the plastic through the heating chamber, which melts the material [48]. At the end of the extruder, the molten plastic is pressed at high pressure into a closed cold mold. The plastic then cools to a solid state, the mold opens and the finished product is cast out. A technique for producing hollow plastic pieces is called blow moulding [49].

Extrusion blow moulding, injection blow moulding, and injection stretch blow moulding are the three basic types of blow moulding [50]. The melting of the plastic and formation of a parison are the first steps in the blow moulding process. The parison is a plastic tube with a hole in one end that allows compressed air to pass through. The air pressure pushes the plastic out to match the mould. The final product is released from the mould once the plastic has cooled and become rigid. Calendering is a continuous operation applied for high output and the capacity to deal with low melt strength [51]. The polished rollers maintain the thickness and make the surface smooth. Spinnerating is an extruding process in which the melted plastic is enforced through a microscopically small sieve, called a spinneret, to make thin fibers used for various products including toothbrushes and nylon stockings [2].

A number of additives are added either during the manufacture of plastics or during processing plastics into consumer products [13,52] (Table 2).

Improvements to the basic chemical, physical and mechanical properties can be done through the incorporation of additives. These also have protective effects, reducing degradation of the polymer from light, heat, or bacteria [2]. They have an impact on viscosity and melt flow in polymer manufacturing. Moreover, additives give products their colour and other unique qualities including increased surface properties, less friction, and flame retardancy [8].

4. Environmental challenges with plastic wastes

The environmental challenges with plastic wastes are dominated by their impact on (i) Land [9], water [53] and air pollution [12] and (ii) Human and animal health [54].

4.1. Land, water and air pollution

Pollution by plastic wastes can harm and pollute the terrestrial

Table 2
Additives used during plastic manufacture and processing [2].

Types of additives	Functions
Antioxidants	Weather resistance is required for outdoor applications and plastic processing.
Colorants	For colouring plastic products.
Foaming agents	Building board and PUR carpet underlayment are suitable for expanding plastic items, such as PS cups.
Plasticizers	Utilized in wire insulation, floors, gutters, and various films to slow down the breakdown of materials due to light.
Lubricants	To produce flexible plastics; used for making fibers and squeeze bottles.
Anti-statics	To reduce dust collection by static electricity attraction.
Antimicrobials	To control biofilm development; applied for shower curtains and wall coverings (Bithionol).
Flame retardants	To increase the security of cultured marble and wire and cable covers.

ecosystem and can be later transported to the aquatic ecosystem, as roughly 80 % of plastic waste existing in marine environment comes terrestrial environments [9]. Two criteria are often adhered in order to restrict waste plastic pollution. One is to stop using plastic altogether and stop producing plastic garbage; the other is to boost recycling and waste treatment [16]. Considering the results of the earlier investigations, the 1st principle appears difficult to put into practise immediately. Regarding the 2nd principle, it is commonly accepted that improper plastic waste management, including dumping and littering, is the main cause of plastic waste entering the environment [55], whereas managed plastic waste treatment methods, like sanitary landfilling, biological treatment, and incineration, can reduce or completely eradicate the risks imposed by plastic waste on the environments. However, Recent researches indicates that existing plastic waste management approaches may leads to release of microplastics into the environment. The abundance of microplastics in Chinese MSW landfill leachate lies in the range of 0.42–24.58 particle/L [56], demonstrating that landfilling is a possible source of microplastics.

According to Su et al. [57], the average abundance of microplastics in garbage from Chinese landfills was 62 particle/kg. Microplastics have been reported to enter the terrestrial environment through compost/digested produces from organic household waste through anaerobic digestion and composting [58]. In Germany, compost is thought to emit between 35 billion to 2.2 trillion microplastic particles into the environment each year [59], with the presence of microplastics ranging from 14 to 895 particle/kg. It is generally known that incineration is a permanent solution for handling and management of plastic waste. However, a solid residue/plastiglomerate generated after incineration contains 360–102,000 microplastic particles/MT, are able to release the microplastic particles in the environment [16]. For instance, plastiglomerate produced by incinerating plastic debris on the beach have been discovered in numerous places, including Ontario, Canada, Madeira, Portugal, and Bali, Indonesia [60]. The findings showed that the plastic waste did not result in a plastic pollution catastrophe by being burned, composted, or dumped in landfills.

Additionally, additives used in plastic, such as stabilisers, colourants, plasticizers, and heavy metals, have the potential to leak into the environment and contaminate soil and water [10,11,13]. Chlorinated plastics have the ability to release hazardous chemicals into the soil, which can then leak into nearby aquatic systems or underground water, damaging the ecosystem [8]. Dumping and landfilling of plastic wastes on land leads to weathering, as well as abiotic and biotic degradation. Methane (CH_4), a potential greenhouse gas (GHG), which is released during the microbial biodegradation of plastics and significantly contributes to global warming [1].

Because to its fast colonisation by marine animals (i.e., the plastisphere) and long-term existence on the oceanic surface, floating plastic fragments may serve as a vector for the spread of 'alien' or non-native species [61]. Pollutants from microplastics are bioavailable for several marine organisms owing to their existence in the pelagic and benthic environments and its tiny sizes. In the marine environment, microplastics can accumulate and sorb pollutants discharged from various sources [11]. Apart from marine pollution, atmospheric pollution can result from the release of GHGs during the degradation and weathering of plastics, and also from the emission of various compounds including particulate matter during the incineration of plastic wastes [62]. CO_2 and CH_4 are emitted into the atmosphere when plastic wastes are disposed in landfills [1]. Open burning and incineration of plastic waste emits contaminates such heavy metals, dioxins, polychlorinated biphenyls (PCBs), and furans that, when breathe in, can have adversely affect human health, particularly respiratory problems [63].

4.2. Human and animal health

Routes of human exposure of plastics include oral uptake through drinking water and diet containing particulate plastics, inhalation of

microplastics from the air, and dermal uptake of nano-plastics and the associated chemicals derived from cosmetics [8]. For example, seafood consumption represents one of the major pathways for particulate-plastic exposure to human [15]. Plastics impact human health at all stages of their lifecycle. They arise from the release of toxic chemicals during feedstock material extraction and plastic manufacturing, exposure to various toxic additives during use, and the contamination of the environment resulting from the plastic waste stream [52]. Extraction of fossil fuels, such as crude oil and gas that provide the feedstock materials for plastic production, releases significant volumes of toxic chemicals into the air, soil, and water [64]. A number of chemical additives, including antioxidants, colourants, foaming agents, plasticizers, lubricants, and flame-retardants, are added during the manufacture of plastics from fossil fuels can also adversely affect the health of the environment and living well-being [13]. Utilisation of plastic-based products leads to inhalation and ingestion of both micro- and nano-plastic particles and the associated toxic substances [53]. Plastic waste management approaches such as, incineration, gasification, and pyrolysis, also result in the emission of hazardous chemicals [65]. The respiratory, neurological, and gastrointestinal systems as well as sensory organs are all affected by these hazardous chemicals [66]. For instance, significantly higher levels of dioxins have been discovered in the tissues of people living close to incinerators in the UK, Spain, and Japan [63]. According to studies performed during the COVID-19 pandemic, poor air quality caused by open burning of plastic and medical waste may be associated with a higher fatality rate in the United States and the Netherlands that ranges from 8 % to 21.4 % [67].

Marine faunas are exposed to plastic fragments mainly via entanglement and ingestion; nevertheless, ingestion is more often than entanglement [8]. Majority of marine animals ingest plastic waste as food by mistake [11]. Additionally, entanglement with plastic wastes such as nets, can leads to injury and harm, even some time death to marine faunas. Marine plastic pollution mainly distresses sea turtles and other marine species whose primary food are jelly fishes since they frequently confuse waste plastic bags for jelly fish [68]. Sea birds frequently make the same mistake, mistaking microplastics for fishes. Ingestion of plastic debris leads to impediment and physical injury to bird's digestive system, impair the digestive capacity of the system resulting malnutrition, starvation, and ultimately death [69].

According to Suman et al. [70], both chronic and acute exposures to polystyrene microplastics at 1 and 100 mg/L, respectively, resulted in the deformation of epithelial cells in the midgut. In an investigation, Chen et al. [71] observed that redclaw crayfish (*Cherax quadricarinatus*) were exposed to various concentrations (0, 0.5, and 5 mg/L) of 200 nm-sized polystyrene microspheres for 21 days. The microplastics were found in the intestine and hepatopancreas after ingestion and inhibited *Cherax quadricarinatus* growth. Freshwater microalgae, Euglena gracilis, were subjected to 1 mg/L of polystyrene microplastics for 24 h [72]. Microalgae had their vacuoles triggered, and their pigment levels had dramatically decreased (p 0.05). Therefore, in consideration with the adverse impacts caused by the plastic pollution on the health of environment and living well-being, its management and upcycling strategies need to be prioritized, which we are comprehensively discussed below.

5. Retrieving plastic wastes for petrochemicals

Globally, around 450 million tonnes of plastics are produced [73] and an estimated 9 % are recycled whilst 22 % are mismanaged [74]. This presents an opportunity, globally, to recover better value from plastic wastes either through more effective recycling or through chemical recycling and conversion of plastics into value-added products [30] Plastic products at the end of use are deemed a waste, however the way in which they are managed varies significantly depending on plastic types and geographical location. Plastics become waste as the end-user deems the material to be of no value to them, nevertheless the recovery of plastics for reuse or recycling is driven by the demand for such

material [75]. As such the value of discarded plastic must be of a certain point to make it worthwhile for recovery, which can either be through the formal or informal resource management sectors.

Recovery of plastics from waste commences at the waste generation sites, thus depends on the availability of a waste collection service, whether or not a source-segregated collection scheme is available and the available market for recovered polymers. Where source-segregation of wastes occurs, enabling the collection of co-mingled recyclables (paper, card, aluminium and plastics etc), the collected materials can be processed to recover target materials. Mixed wastes (i.e. not source-segregated recyclables) can also be managed to extract recyclable materials, however this is often associated with higher levels of contamination [76]. Post-consumer plastic wastes can be separated from co-mingled recycling or from mixed wastes using mechanical processes [32,77,78], often involving an element of hand sorting, in facilities termed as material processing facilities (MRFs). MRFs vary in complexity [77], depending on their location, target input material and the intended separation effectiveness.

Plastic wastes ultimately end up either being recycled in some form (including reuse), thermally treated to recover energy, disposed of in landfills/dumpsites or discarded to the environment [9]. Formal collection processes and mechanical facilities are a crucial component of plastic waste recovery, however the role of the informal sector in developing economies is significant [79]. In the informal sector, components of waste are either collected from the streets or reclaimed from open dumpsites [80,81]. Plastics are recovered through informal processes, thus can be recycled in similar processes as plastics recovered from formal waste management operations.

Reprocessing plastics into useful products is achieved through recycling, which can be categorised as primary, secondary, tertiary or quaternary recycling [32,82]. Primary recycling is a mechanical process whereby plastic waste is returned back into its original product [32,53]. Secondary recycling is similar; however, the result is a downgraded product [82]. Tertiary recycling includes chemical recycling processes, producing chemicals and fuels by breaking the polymeric chains [31 53 83]. Quaternary processes involve the recovery of energy from plastic wastes through thermochemical processes [84]; whilst not yielding a physical product, energy is produced and landfill disposal is avoided [85]. An overview of the pathways of managing post-consumer plastic wastes is shown in Fig. 1.

Mechanical processes enabling plastic recycling may include physical sorting, washing and grinding of the material. Mechanical processes to separate and sorting of plastic wastes is summarized in Table 3.

Mechanical processing for recovery of plastics plays a crucial role in enabling chemical recycling of plastics, as conversion processes are designed for a specific polymer or polymer blend [30,32,84]. Alternatively, it may be necessary to remove certain polymers such as PVC due to the impacts on the quality of the chemicals and fuels being produced [77]. Therefore, for the conversion of plastics to valuable products through chemical recycling, the quality of the feedstock is critical necessitating a controlled system to recover plastics suited to the thermochemical conversion. Advanced thermal methods, such as gasification and pyrolysis, have been studied for the purpose of chemical recycling for the production of valuable products [87–89], showing the importance and realize to understand the full system of plastic waste generation, collection, separation and conversion.

6. Processes of conversion of plastic wastes to petrochemicals

6.1. Thermochemical processing

6.1.1. Pyrolysis

Pyrolysis has appeared as a promising technology to convert plastic waste into valuable products including petrochemicals. It is a thermochemical process that breaks down long-chain polymeric materials into smaller molecules under an inert or limited oxygen environment
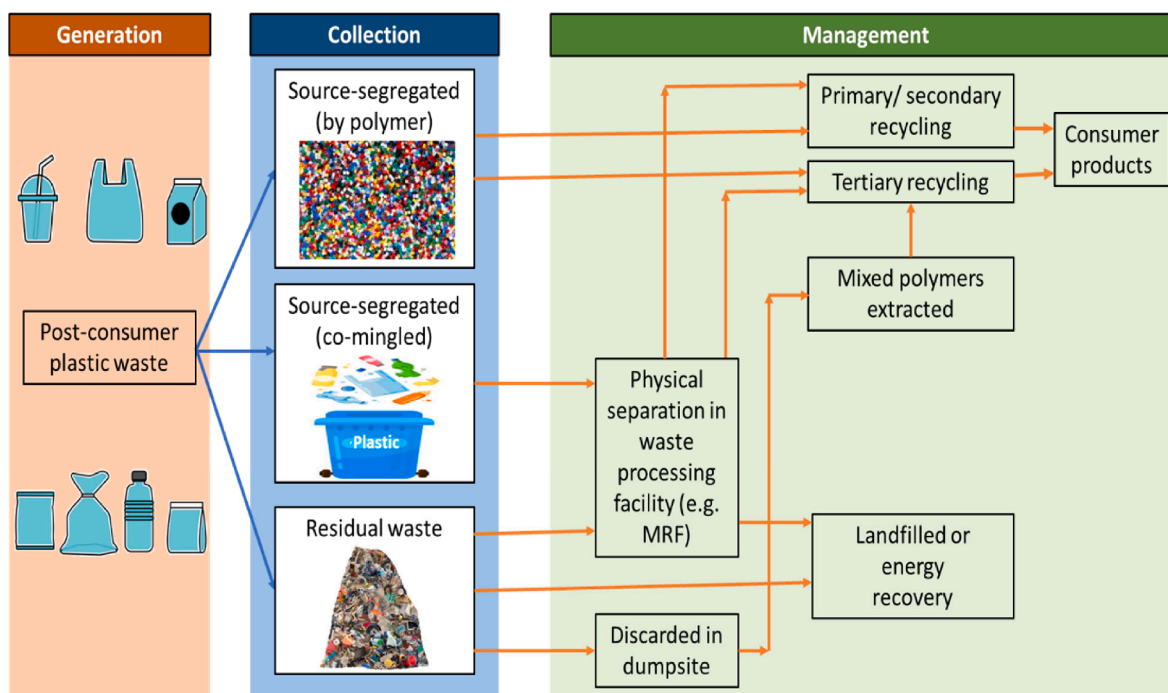
Fig. 1. Overview of managing post-consumer plastic wastes.

Table 3
Overview of mechanical processes to separate and sort plastic wastes [77,86].

Process	Function	Objective	Role in plastic separation
Screening	Sorting waste by size	Separate large and small components	Plastics remain in larger fraction
Air separation	Use of blowers to separate by density	Separates heavy and light fractions	Plastics remain in the light fraction
Ballistic separation	Vibration	Separates by size and can sort rigid and flexible plastics	Plastics are sorted into rigid and flexible fractions
Hand sorting	Manual sorting of waste, typically using a conveyor belt	Laborers selectively separate target materials	Plastics are sorted into flexible and rigid fractions. Plastics may also be sorted into polymer types.
Drum washing and floatation	Use of water to separate and/or clean dense contamination from waste	Removal of contaminants	Plastics are separated by floatation. Contaminants are removed.
Sensor-based sorting	Optical and near-infrared sensors detect polymer types and activate air jet to sort	Separate plastics and sort by polymer types	Target polymers are sorted

[90,91]. Fundamentally, four different mechanisms such as random-chain scission, end-chain scission or depolymerization, cross-linking, and chain stripping, take place during the pyrolysis of plastic waste [92]. Before the process of pyrolysis, the compositions of waste plastic must be subjected to proximate analysis in order to determine their moisture, fixed carbon, volatile matter, and ash contents [55]. As a result, the main variables affecting pyrolysis yields are the volatile matter and ash contents. The high ash content in the plastic waste leads to reduction in yield of liquid oil and, as a result, increased gas yield and char formation. While, the quantity of volatile matters favored plastic oil production (17).

The products produced from plastic waste pyrolysis are liquid hydrocarbon and incondensable gaseous, which can then be converted into petrochemical or fuel products [93]. Pyrolysis conversion method for plastic waste is preferable over incineration and landfill because it does not cause air pollution (from incineration) and water pollution (from landfill) as pyrolysis is performed in an enclosed system. In addition, high costs for plastic separation processes can be avoided as the input waste for pyrolysis can be a mixture of recyclable and non-recyclable plastic waste. Conventional pyrolysis of plastic wastes utilized an electrical oven/furnace as the heating source, with a heating rate of up to 10 °C/min and a maximum temperature at 600 °C, while inert gases such as nitrogen are used as carrier gas to create an inert environment

for pyrolysis processes to occur [92]. The conversion rate of plastic waste into liquid oil is up to 80 wt% and the yield can be affected by the pyrolysis parameters, including heating rate, temperature, time, feedstock composition, types of reactors and also the presence of catalyst [17].

Among them, the quality and quantity of the pyrolytic products are strongly influenced by temperature since it has an impact on the cracking reactions to different fuels and gases, but has minimal effect on the generation of char. Due to the rapid heating rate, it can vaporize feedstocks to produce gases at high temperatures which can further condensed to form liquid products, while reduce the yield of chars [94]. Temperature and heating rate not only influences the yield; it can also influence the compositions and quality of the fuel oils. When PP and PE were pyrolyzed at various temperatures, PP produced 53 wt% oil, primarily benzene, toluene, and xylene (BTX), whereas PE produced 32 wt% BTX fractions [95]. In the pyrolysis of polyolefins, Diels-Alder reactions afterwards dehydrogenation result in the generation of aromatics. Primary and secondary radicals are first produced in PP by a random chain scission mechanism, afterwards intramolecular radical transfer reactions that result in tertiary radicals. Propene is ultimately produced when the tertiary radicals are cleaved at the β-position. Among the BTX aromatics, benzene has been produced in both the PP and PE fractions with a notable yield.

The pyrolysis procedure and the end products are also affected by retention time and feedstock compositions. However, they have less of an effect than temperature. Miandad et al. [96] described the synthesis of aromatic hydrocarbons at shorter retention durations, particularly when the feedstock is PS. At 350 °C, pyrolysis of mixed polymers comprising PS, PP, and PE produces 60 % of aromatic hydrocarbons [92]. It was concluded that the retention time had a negligible impact on the smaller fractions of carbon chains, since essentially identical fractions that contained smaller carbon chains were found at each temperature despite varying the retention time. In contrast to this, fractions with more than C_{13} chain length were seen as the temperature increases. This is because plastic and its derivatives degraded over a longer retention time in the reactors, producing large carbon chain products in contrary to light carbon chain products, which produced over a shorter retention time and high temperature [97]. Therefore, the temperature is the key bottleneck in conventional pyrolysis as the heat loss and inefficient heating during the pyrolysis resulting a rise in production cost, thus, microwave heating has been introduced as an alternative to conventional heating for application in pyrolysis [98].

Unlike conventional heating, microwave heating is a result of the molecular interaction between the material's particle and electromagnetic field via mechanisms such as dipole polarization and ionic conduction [90]. Microwave energy can penetrate the inner part of the material being heated, where heating occurs from the core of the material toward the surrounding environment, and in turn providing a rapid (up to 50 °C/min) and uniform heating, short process time (less than 30 min) and produce high oil (81 wt%) and gas (18 wt%) yield [99]. The rapid heating reduces the processing time required and thus lowering the energy consumption. Pre-treatment such as drying and particle size reduction can also be excluded from microwave pyrolysis [100]. Thus, the overall production cost for conversion of plastic waste via microwave pyrolysis is relatively reduced as compared to conventional pyrolysis. Unfortunately, plastic waste has poor microwave absorption, leading to a futile transfer of microwave energy and faster thermal runaway. Therefore, efforts have been made in assisting the heating processes in microwave pyrolysis of plastic waste by adding microwave adsorbers such as activated carbon or biochar that may improve the energy adsorption. Table 4 summarizes the major products obtained from microwave pyrolysis at different parameters.

A review was performed by Putra et al. [17] on the microwave pyrolysis of plastic waste as a feasible waste management method. The review reported that microwave pyrolysis can be improved by modifying the reaction parameters such as microwave power, temperature, reactor design, adsorber and catalyst input. The liquid by products produced during microwave pyrolysis of plastic waste were reported to show fuel properties that are comparable to diesel or gasoline, which can possibly be used as a fuel blending component [17]. Instead of adding microwave absorbent or catalyst, some researchers performed microwave co-pyrolysis, where the plastic is mixed with other feedstock during microwave pyrolysis processes [23]. For instance, mixing of waste cooking oil and LDPE can enhance the production of monocyclic aromatic hydrocarbon in the pyrolytic oil [26], and a mixture of hospital plastic waste with waste vegetable oil or palm kernel shell produced high yield liquid oil (78.2–80.5 wt%) with low nitrogen and oxygen content [104], mixing of PET and biomass such as rice husk produced mainly monoaromatic oxygenates (52 %) and biphenyl hydrocarbon (29 %) [105].

Modifications have been performed on the conventional microwave technology, such as introducing vacuum conditions to replace the need for nitrogen as carrier gas during pyrolysis [23]. Pyrolysis performed under vacuum conditions was also reported to enhance the thermal cracking of feedstock, producing high bio-oil yield and increasing the final temperature of the processes, which in turn improving the energy efficiency and reducing the production cost [23,99]. Replacing N_2 with CO_2 in microwave pyrolysis of plastic waste also shows positive synergy where the carbon reallocation ability of plastic waste was changed, resulting in an increased production of syngas yield with a controlled H_2/CO ratio [106]. However, there is still some research gap to be filled in microwave-assisted pyrolysis. These include in-depth research on advanced pyrolysis technology (e.g., vacuum, co-pyrolysis and catalytic pyrolysis) and a systematic review of specific advanced pyrolysis techniques in the valorization of plastic waste. Besides that, no research was performed on the life-cycle assessment (LCA) and techno-economic analysis (TEA) of microwave pyrolysis of plastic waste [107,108], where the environmental impact and the operating cost for a microwave pyrolysis plant for plastic valorization remain unknown, hence the challenge to commercialize this technology in near future.

6.1.2. Gasification

Gasification is a thermal process utilizing gasifying agent (e.g., steam, air, and oxygen) to convert plastic waste into a gaseous mixture of CO_2, H_2, CO and CH_4 via partial oxidation at high temperatures (500–1300 °C) [24,98]. Gasification is one of the emerging and promising approaches in thermochemical recycling where most of the plastic waste can be gasified even when the polymer is contaminated, thus limiting the need for pre-treatment. The gasification of plastic waste also shows several benefits over incineration or landfills. For instance, the emission of harmful gases such as dioxins, SO_2, NO_x, HCl and HF are

Table 4

Microwave pyrolysis of various types of plastic at different parameters and its major products.

Feedstock	Adsorbent/catalyst	Microwave power (W)	Temperature (°C)	Products	Reference
HDPE	ZSM-5	1000	500	98.9 % C_5-C_{12} hydrocarbons, where 55.6 % are C_5-C_{12} olefins, 28 % are aromatics	[101]
HDPE, LDPE and PP	Fe/Ni-CeO$_2$@CNTs	1000	700–800	91.5 vol% H_2 (50.2 mmol/g)	[25]
HDPE	NiFe$_2$O$_4$	1000	400	93.0 wt% H_2 (57.0 mmol/g)	[102]
PS	SiC	650	460	98.8 wt% oil yield, where 64.8–85.9 % are C_8-C_{16}	[103]
LDPE + waste cooking oil	HZSM-5	1400	500	48.0 wt% oil yield, BTX content are 500.2 mg/mL	[26]
LDPE + waste vegetable oil	Activated carbon	700	450	73.7–80.5 wt% bio-oil yield, with low composition of aromatics	[104]
LDPE + palm kernel shell	Activated carbon	700	450–550	64.5–78.2 wt% bio-oil yield, domain by alkenes, alcohols and aromatics	[104]
PET + rice husk	Graphite	450	600	29 wt% syngas yield, biocrude contained mainly monoaromatic oxygenates (52 %) and biphenyl hydrocarbon (29 %).	[105]
LDPE + seaweeds	SiC	700	500	High bio-oil yield (68.9 w. %) and contain mainly hydrocarbon (51.2 %)	[23]
HDPE + waste cooking oil	Activated carbon	800	402–470	High bio-oil yield (62 wt%) with high heating value (49 MJ/kg)	[99]

HDPE: High density polyethylene; LDPE: Low density polyethylene; PP: Polyethylene; PS: Polystyrene; PET: Polyethylene Terephthalate; BTX; Benzene, Toluene, and Xylene.

reduced via gasification under a controlled quantity of oxidant used and at a lower temperature compared to incineration [109]. Syngas (H_2 + CO) is the major product of gasification and the quality of syngas highly depends on the H_2/CO ratio, where it can be converted to methanol (catalytic hydrogenation) or produce fuels and chemicals via Fischer-Tropsch processes [56]. Theoretically, the addition of steam (Eq.1) in gasification processes produces a higher hydrogen yield compared to pure air or oxygen (Eq.2) [55].

$$C_xH_y + xH_2O \rightarrow xCO + \left(\frac{y}{2} + x\right)H_2 \tag{1}$$

$$C_xH_y + \frac{x}{2}O_2 \rightarrow xCO + \left(\frac{y}{2}\right)H_2 \tag{2}$$

Recently, numerous efforts have been made by the researcher to introduce microwave heating in gasification processes. Ellison et al. [110] have performed a study to compare the conventional gasification and microwave gasification of different types of coal. The result reported that utilizing microwave as a heating source has successfully decreased the coal gasification temperature to 700 °C (conventional heating required more than 700 °C), and has greater selectivity of syngas species, as well as increasing the yield of non-condensable gaseous. Coal with microwave adsorption properties has created different hotspots within the reaction region, then heats the reaction region in a short period, resulting in a higher decomposition rate of the coal, and thus producing a higher yield of gaseous products. Another research was performed by Li et al. [111] where microwave radiation and catalyst (red mud) were utilized in spirit-based distillers' grain gasification. An increase in syngas yield and total gas yield was experienced in the presence of a catalyst. This is because the use of microwave heating has created a higher surface area for the feedstock, thus increasing the amount of catalyst inherent on the surface, which in turn improves the gasification reaction. According to a review performed by Chan et al [112], the types of feedstocks were limited to oil palm shell, pistachio nutshell [113,114] and coconut shell [115]. Microwave gasification of plastic waste has a huge research gap that needs to be filled as there was no research reported on this topic. The potential of this combined technology in plastic waste valorization is yet to be thoroughly examined.

6.1.3. Hydrothermal liquefaction

Hydrothermal liquefaction (HTL) is a thermochemical technology to convert feedstock such as plastic waste into crude-like oil within a temperature and pressure range of 250–450 °C and 4–22 MPa, respectively, with the presence of a solvent [116]. A reaction environment with high pressure and temperature reduces the dielectric constant and density of water, resulting in the water molecules being less polar and thus can dissolve the organic components in HTL conditions. Besides that, the less polar water molecule has a more evenly shared electron between H and O atoms, resulting in an increasing dissociation of the water molecules into H^+ and OH^- ions, suggesting that it is a good medium for based- or acid-catalyzed reactions [116]. In comparison with pyrolysis, the products produced from HTL pose higher heating values and lower moisture and oxygen content, and the wet and unsorted feedstocks (e.g., plastic) can be processed directly [117]. Previous research confirmed that the optimum temperature for HTL processes for most plastic falls between 350 and 450 °C [116].

Microwave hydrothermal liquefaction (MA-HTL) has recently been explored for its potential to reduce reaction time, accelerate the reaction, and enhance the product yield and quality as compared to conventional hydrothermal liquefaction [118]. Similar to microwave pyrolysis and microwave gasification, the heating source of the processes is replaced with microwave heating owing to its unique heating mechanism. Different feedstocks were selected including macroalgae [119], and lignocellulosic and non-lignocellulosic biomass [120]. A review performed by Gao et al. [121] on MA-HTL of biomass summarized that the optimum reaction conditions of MA-HTL are moderate temperature ranging (180–250 °C) and pressure (2–10 MPa), reaction time ranging from 20 to 60 min, while the presence of a catalyst, solid to water ratio, microwave power and biomass particle size need further experiment to determine the optimum condition. It is believed that MA-HTL treatment is an up-and-coming method in biomass/waste valorization. However, no research has been performed using this technology in plastic waste valorization, which is a big research gap that is yet to be filled.

6.2. Catalytic conversion

These days, with the expansion of agriculture and economy, the global demand for plastics has continued to increase, and most of the plastics are accumulated in landfills or left in the environment, resulting in serious plastic pollution and resource loss [22]. Environmental pollution and energy shortages have gradually become common challenges facing the world. Therefore, methods for the catalytic conversion of plastics, such as electrocatalysis and photocatalysis, have gained increasing consideration, through which plastic waste can be converted into value-added chemicals and fuel, or materials with additional commercial benefits [19,122].

Electrocatalysis can be driven by renewable energy (solar, wind, and hydro), which produces clean H_2 or fuel from the cathode under mild conditions and value-added oxygenates through the oxidative decomposition of organic compounds at the anode [123,124]. This method has gradually become a sustainable strategy for the catalytic conversion of plastics, which has made great progress in the efficient and selective conversion of various organic compounds [125,126]. Zhou et al. [127] investigated the conversion of waste PET into value-added products through an electrocatalysis method. Fig. 2 represents the conventional route for PET recycling and well as electrocatalytic PET upcycling and its TEA at different current density.

PET is hydrolyzed in KOH solution into two monomers, terephthalic acid (PTA) and ethylene glycol (EG). In the membrane-electrode assembly reactor, the carbon fiber paper anode can efficiently catalyze the oxidative cleavage of C–C bonds of EG into formate, and at the cathode, it has excellent hydrogen evolution activity. Thereafter, formic acid is utilized as an acidifying agent for PET electrolysis to recycle plastic monomer PTA. The resulting liquid stream is converted to high value-added solid product potassium diformate (KDF) by concentration and crystallization. The estimated net income of 1 ton plastic waste is about 350 dollars using this electrocatalysis technique. In addition, some researchers applied an external voltage (0.55 V) in H_3PO_4 solution at 200 °C to successfully convert polyvinyl alcohol (PVA) to clean energy H_2 (9.5 μmol/min) [125]; In addition, electrocatalytic technique has also been used to degrade PVC and form industrial raw materials CH_3COOH (75 %) at 100 °C on a C cathode (−0.7 V) [126].

As one of the emerging strategies for solar fuel conversion, photocatalytic technology has been extensively studied recently owing to its environmental friendliness and low cost, which is an ideal solution to energy shortage problems. Photocatalysis technology mainly utilizes the catalyst to generate electrons (e^-), holes (h^+) and reactive oxygen radicals (ROS) under the irradiation of light sources with energy greater than its band gap [128,129]. Holes and ROS oxidatively degrade microplastics into small organics and CO_2, and electrons reduce water molecules and CO_2 into H_2 and fuel [129]. The technology can be performed at room temperature and pressure, and can directly use clean solar energy to transform plastics into high value fuels and other small-molecule chemicals [128].

Researchers have developed photocatalysts for the oxidative degradation and reduction of microplastics in terms of productivity [128,129]. Titanium dioxide (TiO_2) doped with metallic platinum (Pt) particles can degrade PE, PVC, and polyvinyl alcohol under UV light irradiation, while generating H_2 (5.7–15.0 μmol/g_{cata}/h). But the modified TiO_2 catalyst has narrow photoresponse range and low productivity [130]. To overcome these deficiencies, researchers have
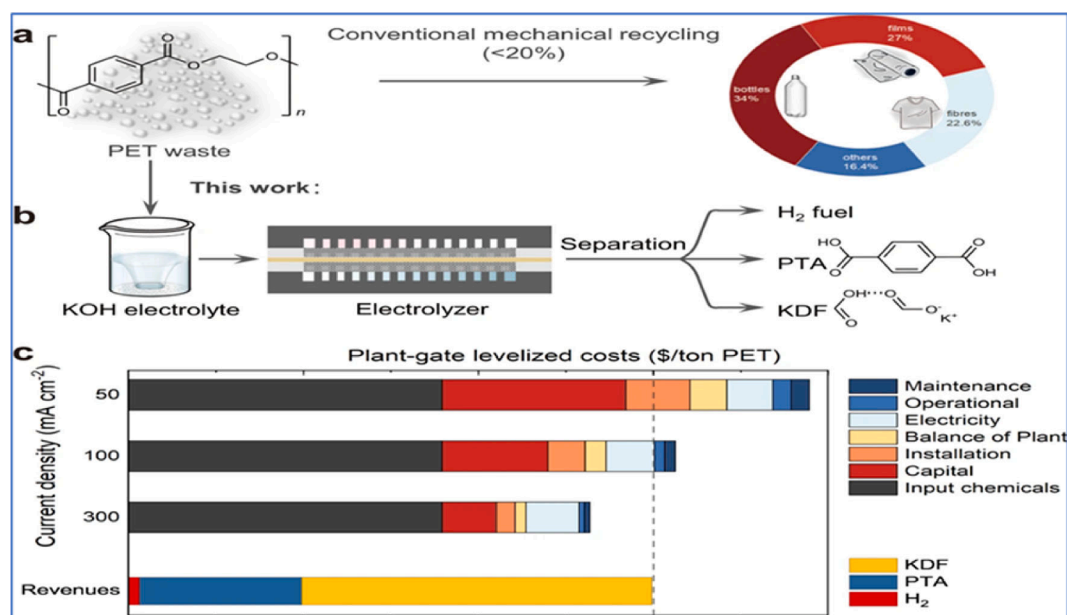
Fig. 2. (a) Conventional route for PET recycling. (b) Electrocatalytic PET upcycling to commodity chemicals and H_2 fuel (Route I). (c) Techno-economic analysis (TEA) of Route I at different current density. Copyright 2022, nature communications [127].

developed CdS/CdO_x quantum dots and carbon nitride/nickel phosphide (CN_x/Ni_2P) nanocomposite catalysts. These photocatlysts can convert PLA, PET, plastics such as PU to H_2 by visible light catalytic reaction under alkaline (1–10 M NaOH) and room temperature conditions. However, CdS/CdO_x quantum dots and CN_x/Ni_2P nanocomposite has disadvantages such as toxic Cd^{2+} release, low photon yield (less than15 %) and low H_2 yield (0.85–64.3 mmol·gCdS/CdO$_x$/h, 25.8–55.6 µmol·gCN$_x$/Ni$_2$P/h) [128,131].

In addition, disposable bags, disposable food containers, food packaging films with main components of PE, PP and PVC were photocatalyzed converted to CH_3COOH for the first time through a two-step conversion route of continuous light-induced C–C splitting and coupling when Nb_2O_5 atomic layer catalysts were used as photocatalysts under under xenon lamp irradiation to simulate natural environment conditions [125]. The results show that PE is photodegraded to CO_2 by Nb_2O_5 atomic layer within 40 h, and the generated CO_2 is further photoreduced to industrial raw materials CH_3COOH by C–C bond coupling [129].

First, the photocatalytic oxidation half-reaction mineralizes the plastic to CO_2 under the action of ·OH and O_2 (Eq. (3); then CO_2 is under the action of electrons to generate a ·COOH intermediate, which generates fuel CH_3COOH through a C–C coupling and continuous protonation (Eqs.3–6), but the fuel yield was too low (18.8–47.4 µg/gcata/h) [129]. How to increase fuel production will be the focus in the futher research.

$$polymer \xrightarrow{\cdot OH, O_2} CO_2 \tag{3}$$

$$CO_2 + e^- + H^+ \rightarrow \cdot COOH \tag{4}$$

$$COOH + \cdot COOH \rightarrow HOOC - COOH \tag{5}$$

$$HOOC - COOH + 6e^- + 6H^+ \rightarrow CH_3COOH \tag{6}$$

6.2.1. Catalytic upgrading of mixed plastics waste containing heteroatoms

The transformation of mixed waste plastic that doesn't contain heteroatoms is considered significantly easier compared with the feedstock that has these additives. Thermochemical processes of mixed plastic waste to petrochemical conversion are faced with great challenges related to the presence of heteroatoms in some plastic types such as PVC,

polyacrylonitrile (PAN), PUR, and PET. This detrimentally affects the quality of the final products by retaining chlorinated, nitrogenated and oxygenated compounds, hence leading to a dropping in useful hydrocarbon yield. The addition of heteroatoms was purposed to improve the polymers' resistivity to combustion and corrosion [132]. These essential virtues increased the demand of modified plastics to around 24.8 % of the total plastics demand [133]. The easy yet expensive solution to retrieve pure hydrocarbons is sorting the plastic waste to exclude the heteroatoms-containing plastic and process the remainders such as PE, PP and PS. Alternatively, utilizing an efficient sorbent/catalyst would be a more realistic and cost-effective solution to selectively eject the heteroatoms while providing valuable petrochemicals with no additional cost or pre-treatments. Additionally, the catalyst also plays an essential role in lowering the activation energy and thus reducing the reaction temperature and energy consumption. Moreover, catalytic materials such as zeolites allow directing product selectivity toward valuable compounds by harnessing their distinctive pore architecture. Hence, catalyst presence in any thermochemical process seems very crucial to ensure the ultimate exploitation of plastic waste as represented in Table 5.

6.2.2. Catalytic pyrolysis of mixed plastics waste

Dealing with heteroatoms present in mixed plastics waste seems inevitable; therefore, developing a catalyst that can limit the possible corrosive and toxic emissions (HCl, HBr, and HNO_3) became the scope of many studies. The release of these heteroatoms to the gas or liquid stream is considered very harmful to the reactor setup and oil quality. This suggests that the most adequate way to contain these additives is to capture them in the solid phase. Lopez-Urionabarrenechea et al. [134] tried to overcome retaining chlorine in the liquid stream by implementing two-step catalytic pyrolysis. The first step was meant to dechlorinate the feedstock thermally by heating up to 300 °C for 60 min forcing the chlorine to be rejected first in the presence of ZSM-5 catalyst. The second step was elevating the temperature to 440 °C to allow the remained feedstock to pyrolyze. The study reported that the dechlorination step succeeded in removing 75 % of the chlorine in the liquid phase. However, the dechlorination negatively affected the catalytic activity of the catalyst resulting in longer-chain hydrocarbons and less aromatic yield. The group solved this drawback by adding the catalyst after the thermal dechlorination, consequently, that restored the catalyst

Table 5

Summary of catalytic conversion of various types of plastics and their dichlorination efficiency.

Plastic feedstock	Catalyst/Process	Dichlorination efficiency	Reference
PE, PP, PS, PET and PVC	ZSM-5/semi-batch reactor (stepwise dechlorination)	75 % of chlorine in the oil was removed.	[134]
HDPE, PP, PS, PET and PVC	Reused ZSM-5/semi-batch reactor	–	[135]
PE, PP, PS, PET and PVC	ZSM-5 and red mud/semi-batch reactor	Red mud captured 42-time chlorine higher than ZSM-5	[135]
PE/PVC PP/PVC PS/PVC	Goethite FeOOH, hematite Fe_2O_3, and magnetite Fe_3O_4, mixed with silica_alumina/semi-batch reactor	90 % of chlorine was captured over Fe3O4.	[136]
PE/PVC PP/PVC PS/PVC	(α-FeOOH), (α-Fe_2O_3), and (Fe_3O_4)/semi-batch reactor	Maximum dechlorination of 96.5 % was achieved over Fe_3O_4.	[137]
PE/PVC PP/PVC PS/PVC	Red mud, γ-Fe_2O_3 + silica alumina /semi-batch reactor (stepwise dechlorination)	77.65 % of the chlorine was captured over red mud.	[138]
PE, PP, PS, PET and PVC	$CaCO_3$/closed-batch reactor	35 % of the total chlorine was captured over the catalyst	[135]
PP, PE, PS, PVC, HIPS-Br	$CaCO_3$ over carbon/semi-batch reactor	94 % of bromine and more than 99 % of chlorine have been removed from the oil.	[139]
PE/PVC PP/PVC PS/PVC	Al-Zn composite	91.1 % of the total chlorine was captured over the catalyst	[140]
LDPE, PP, PS, PVC	Al–Mg composite	91–93 % of the total chlorine was absorbed over the catalyst.	[141]
LDPE, PP, PS, PVC	Ni over $CaCO_3$	99.2 % of HCl in the gas stream was removed.	[142]

PE: Polyethylene; PP: Polyethylene; PS: Polystyrene; PVC: Poly vinyl chloride; HDPE: High density polyethylene; LDPE: Low density polyethylene; PET: Polyethylene Terephthalate;

performance by maintaining its active sites. The two-step pyrolysis gave a good result in removing chlorine but this is only practical for the semi-batch process [134]. One of the biggest concerns when using zeolite as a catalyst in any reaction is the possible poising due to the irreversible interaction with the reactants. Lopez et al. [135] in another study demonstrated that the presence of PVC in the pyrolysis feedstock doesn't cause permanent damage to the catalyst, hence ZSM-5 was reusable many times after proper regeneration and performed as good as the fresh catalyst. This indicates that chlorine is not detrimental to the zeolite's active site.

In another study by Lopez et al, a comparison between ZSM-5 and red mud performance in mixed plastics waste pyrolysis was conducted [135]. The feedstock contained PVC which directly influenced the resultant liquid and solid compositions. Apparently, the red mud required a higher temperature of 500 °C to play a role in pyrolysis while ZSM-5 showed great improvement at only 440 °C. The authors related the humble performance of red mud to the poor surface area and low acidity. It is worth noting that, the chlorine amount on the solid product of red mud is higher than thermal and ZSM-5 runs. On the other hand, the chlorine content of the liquid obtained from red mud was less than its counterpart from ZSM-5 experiment. This is likely because CaO phase that exists in red mud promoted the chlorine capture.

Seemingly not only CaO in the red mud is a major player in absorbing HCl, but hematite also can actively capture the chlorine during the pyrolysis of MWP. In one of these studies led by [136] they examined the efficiency of three types of iron oxides, goethite FeOOH, hematite Fe_2O_3,

and magnetite Fe_3O_4, physically mixed with silica-alumina catalyst. Interestingly, FeOOH and Fe_3O_4 exhibited excellent performance as they removed more than 90 % of the chlorine meanwhile the efficiency of Fe_2O_3 didn't exceed 50 %. These findings suggest that the hematite phase present in red mud can work as a sorbent for chlorine however other forms of iron oxide could give better performance. The study also verified the formation of ferrous chloride over the spent catalyst; however, this new phase didn't form crystals as the XRD results didn't detect additional peaks [136]. Surprisingly, in another study by Shiraga et al. [137] revealed that all the active iron sorbents, except Fe_2O_3, were eventually converted to Fe_3O_4 phase after they were used in mixed plastic waste degradation and regenerated. In an article on exploring the effect of Fe in capturing chorine, Yanik et al. [138] compared the results of mixed plastic waste pyrolysis using three catalysts namely; red mud, γ-Fe_2O_3 and silica-alumina. Confirming the conclusion drawn by López et al. [135] red mud showed no effect on plastic degradation at a low temperature of 430 °C. However, it exhibited excellent chlorine fixation ability as discussed before. Passing nitrogen as a carrier gas helped in suppressing the reaction of chlorine with other plastic intermediates, hence that led to the decrease of chlorinated organics. Unexpectedly, the study reported that red mud increased the chlorine in the oil phase because the trapped chlorine over the red mud interacted with the volatiles derived from other plastic. This conclusion raises a concern that red mud could do an excellent job in the dechlorination of the gas stream but due to the existence of several metals, it also induced the reaction between the captured chlorine and other hydrocarbons [138].

In another direction of removing heteroatoms during mixed plastic waste pyrolysis, López et al. [135] investigated the use of $CaCO_3$ as an alkaline adsorbent for the selective capturing of HCl. The carbonate reacted with HCl vapors resulting $CaCl_2$ which remained with the solid residue. The proposed method recovered a great amount of HCl from the gas stream, however it resulted in more chlorine in the liquid stream. The group compared this method with the stepwise pyrolysis and they concluded that the stepwise route is more efficient as it dechlorinates the gas and liquid stream and enhances the oil products [135]. Beyond the dechlorination role, $CaCO_3$ exhibited further catalytic activity as it increased the gas yield and aromatic in the liquid phase. The use of alkaline carbonate for removing the chlorine attracted more attention as a different study was performed by Bhaskar et al. [139]. The feedstock they used contained, besides the polyolefins, PCV and high-impact PS with brominated flame retardant (HIPS-Br). The catalyst preparation included mixing the $CaCO_3$ with phenol resin followed by thermal treatment to obtain carbon composite which had a surface area of 40 m^2/g. The developed composite was very effective as it removed 94 % of the bromine and 99 % of the chlorine present in the oil. This result on oil dehalogenation is comparably better than the previous study by López et al. [135], even though both studies used the same $CaCO_3$. The way the catalyst composite was prepared by Bhaskar et al. [139] offered an extra surface area which might be the reason that this catalyst accommodated more solid residue.

The formation of a composite consisting of both absorbent and acidic catalyst became a promising approach for upgrading mixed plastic waste. Tang et al. [140] prepared an Al-Zn composite by coprecipitation using hydroxides precursor. The new catalyst lowered the boiling point of the liquid products as well as decreased the chlorine content compared with when Al_2O_3 and ZnO were used as catalysts individually. Similarly, Zhou et al. [141] prepared an Al-Mg composite that captured 91.1 % of the chlorine utilizing the excellent absorbent ability of Mg. On the other hand, the sole MgO catalyst only trapped 67.4 % of the chlorine in the solid phase. This enhancement, offered by Al-Mg catalyst, perhaps because the combination of MgO and Al_2O_3 provided an extra surface area that accommodated more chlorine [141].

Typically, mixed plastic pyrolysis leads to the formation of light and non-condensable alkanes and alkenes. These useful hydrocarbons could be further utilized for the synthesis of valuable petrochemicals. However, unfortunately, the gas stream also contains alkynes and HCl which

poison the catalyst. Hence, Veksha et al. [142] proposed loading Ni on alkaline carbonate and forming a bifunctional catalyst to remove the chlorine and promote the cracking of alkynes to hydrogen and carbon nanotubes simultaneously [142]. The developed catalyst worked perfectly as it assisted in converting 99 % of unsaturated hydrocarbons to hydrogen and carbon nano-tube (CNT). Additionally, it also promoted dechlorination with the help of $CaCO_3$, however, that relatively declined the catalytic activity of Ni due to the poisoning.

In a summary, chlorine can be removed efficiently in the stepwise heating process, however, that results in producing significant amounts of HCl which likely corrode the equipment. Alternative capturing options such as zeolites seem not promising because they tend to increase the chlorine content in the oil. This occurs because they allow a longer contact time between chlorine and other hydrocarbons. Moreover, conventional zeolites lack suitable active sites for trapping chlorine, therefore that influenced negatively on their performance despite the excellent degradation and aromatization properties. On the other hand, other metals like Fe, Ca, Mg, and Zn exhibit impressive chlorine-capturing capabilities reaching 99.2 %. Red mud contains two of these active metals, Fe and Ca, hence it showed remarkable chlorine fixation without a need for additional modifications. The efficiency of chlorine capturing relies largely, besides the active sites, on the availability of surface area, hence designing appropriate support is very critical step.

6.3. Chemolysis

The fundamental concept behind chemolysis is to trigger a reversal of the condensation polymerisation process. This technique aims to decompose condensation polymers into individual monomers or chemical compounds that can be further utilized. Various reactions, such as glycolysis, hydrolysis, methanolysis, and aminolysis, are employed with the assistance of specific solvents like ethylene glycol, water, methanol, and ethanolamine, respectively, to achieve this goal [28]. Condensation polymers can be considered as products in equilibrium, which can be reversed by adding other condensation products, like methanol or ethylene glycol, and heating the mixture. The chain scission reactions of these polymers can be precisely regulated by utilizing solvents that initiate nucleophilic attacks on the $C = O$ bond. The operating temperature for chemolysis usually falls within a range of 200–450 °C. Due to such non-extreme operating temperatures and the end products with higher values, this method is favoured over thermochemical or mechanical methods [18].

Condensation polymers, such as PET, PE, PAs, and PC, have been shown to be suitable for chemolysis [18]. PAs and PCs are polar condensation polymers that are very commonly used in several applications owing to their excellent mechanical and thermal properties. However, their non-degradable nature poses a challenge in terms of disposal and recycling. Depolymerisation of PA is challenging due to their stable amide bonds, which contribute the toughness. Catalytic hydrothermal depolymerisation of PA6 using phosphotungstic heteropoly acid as a catalyst has been reported to achieve a high yield of ε-caprolactam [143]. Subcritical water has also been applied to transform PA 6 into ε-caprolactam and 6-aminocaproic acid [143]. Hydrophilic ionic liquid and microwave irradiation with N,N-dimethylaminopyridine as an organocatalyst have also been used for depolymerizing PA6 [20,144]. The breakdown of PA12 into methyl ω-hydroxydodecanoate and ω-hydroxy alkanoic acid has been studied in supercritical methanol media with glycolic acid as a catalyst [145].

PCs are another type of condensation polymer with carbonate linkages. So far, hydrolysis, alcoholysis, and aminolysis have been studied to recycle PCs chemically [146]. The depolymerisation of bisphenol A polycarbonate (BPA-PC), the most economically relevant PC, results in the generation of bisphenol A (BPA) and CO_2 [18]. Secondary reactions can also occur. Various conditions have been studied to enhance the depolymerisation of BPA-PC, including high temperatures, the addition of salt or acetic acid, and the use of ionic liquid catalysts [27]. The latter

has the advantage of nearly full conversion at low temperatures, but the cost of ionic liquids is prohibitive. A bifunctional acid/base catalyst (zinc oxide and tetrabutylammonium salts) has also been used to depolymerize BPA-PC to obtain various carbonates or urea in excellent yields (greater than97 %) [147]. The presence of additives such as flame retardants and plasticizers can impact the depolymerisation, with the former accelerating hydrolysis and the latter having an inhibiting effect [148].

The use of chemolysis to enhance the utilisation of recycled PET goods has been investigated, as the conventional mechanical recycling process typically leads to downcycling with a moderate yield, ranging from 50 to 77 % [28]. Depolymerisation of PET is carried out with various chemical agents and reaction conditions, resulting in monomers such as bis(hydroxylethylene) terephthalate (BHET), dimethyl terephthalate (DMT), terephthalic acid (TPA), and EG [28]. Hydrolysis is the most commonly studied method, but it requires a large amount of concentrated sulfuric acid or alkaline solution for effective depolymerisation and results in low-purity TPA because water is a weak nucleophile. Aminolysis with amine-based aqueous solutions produces BHET, while methanolysis with methanol produces DMT and EG. However, separating methanolysis products can be expensive. Alcoholysis with ultrasmall ZnO nanoparticles has been proposed as a solution, as these nanoparticles accelerate depolymerisation and can be recovered after the reaction [149]. Glycolysis, which uses glycols to convert PET into BHET, is considered the most promising method due to mild reaction conditions and low volatility solvents [150]. However, glycolysis is a slow reaction and often requires the use of metal-based transesterification catalysts to improve its efficiency. Many catalysts have been applied in glycolysis, including metal derivatives, zeolites, and ionic liquids, with zinc and manganese derivatives showing excellent performance [150]. However, homogeneous catalysts like heavy metal salts have toxicity and recovery issues, leading to the study of environmentally friendly and efficient catalysts like nanocomposites, zeolites, and ionic liquids [150]. Recent research has focused on easily recoverable catalysts for glycolysis, such as magnetic materials that can be magnetically separated. Although nanocomposites still require harsh conditions, ultrasmall cobalt nanoparticles synthesized with a capping agent have been shown to improve glycolysis efficiency [151].

PUR is a commonly used polymer, which can be classified as flexible (53 %) and rigid (42 %) foams, with the rest (5 %) being durable elastomers [152]. PUR's chemical structures are complex due to the various monomers used, including polyol and isocyanate, as well as added surfactants, catalysts, blowing agents, and other functional additives, making recycling challenging [152]. PUR waste poses a substantial environmental apprehension owing to its persistence and recalcitrance. To minimize its adverse impact, extensive research has been conducted on recycling PUR. Thermoplastic PUR (TPUR) can be remelted and recycled using mechano-thermal processes. On the other hand, thermoset PUR, which constitutes the majority of PUR, cannot be reprocessed and is usually reused into low-value products. Chemolysis is one of the potential means of PUR recycling [152].

PUR can be hydrolyzed to yield diamines and original diols by breaking down the urethane linkages within PUR [153]. Glycolysis of PUR breaks down PU at 200 °C through a transesterification reaction, and the choice of catalyst dictates the resulting products. Catalysts can be categorized into hydroxides of alkaline metals, acetates, metallo-organic compounds, and amines [154]. For example, a Zn/Sn/Al hydrotalcite catalyst has been applied to recover polyol from PUR flexible foam [155], while diethylene glycol has been utilized as a glycolysis agent to produce polyols with similar properties to commercial polyols. PUR elastomer can be broken down using the $ZnCl_2/H_2O$ system [156], while a DES containing choline chloride and urea has been used for PC-based PUR. However, selective cleavage of the C-N versus C-O bond in the urethane unit and the complexity of commercial PUR preparations continue challenges for recycling.

To sum up, chemolysis is a potentially promising but still evolving

method for reducing plastic waste, with both benefits and challenges to consider. The yield and efficiency of chemolysis can be affected by various factors, such as plastic types, catalyst types, and the reaction conditions. Since most studies report the yield in the controlled environment at the laboratory scale, further research is needed to optimize the chemolysis and make it more practical for large-scale recycling of plastic waste.

7. Economic analysis of retrieving plastic wastes for petrochemicals

While plastic pyrolysis, gasification, and hydrothermal liquefaction of plastic are all technologies with a high technology readiness level (TRL greater than 7), most studies in the literature focus on pyrolysis. Indeed, companies such as Plastic Energy (pyrolysis) [157], Mura Technology (hydrothermal liquefaction) [158], and Enerkem (gasification) [159], among others, have all announced ambitious projects in 2022 for the chemical recycling of plastic waste at an industrial level. Unfortunately, no reports were found in the literature on the TEA of the hydrothermal liquefaction of plastic waste for the production of petrochemicals.

The overview of the TEA, published in the last 5 years, focused on converting plastic waste through gasification and pyrolysis into petrochemicals, is represented in Table 6.

All studies considered a central facility converting polyolefin plastic wastes, which in some cases included other plastic components, such as PVC or PET. The final product depended highly on the process configuration. Plastic waste gasification focused on producing methanol or hydrogen locally, as syngas has no market value [21]. Simultaneously, pyrolysis yielded a range of products that required further

transformation, like olefins, naphtha, and waxes, or purification, such as a mixture of aromatic complexes.

In the specific case of pyrolysis, the large range of products can significantly increase the complexity of the process. For example, Gracida-Alvarez et al. [160] have performed the TEA for waste high density polyethylene (HDPE) refinery with the individual recovery of ethylene and propylene, as well as other light hydrocarbons, aromatics, and heavy hydrocarbons (naphtha + wax) that will be further processed in petrochemical and refinery facilities. While the authors found the pyrolysis reactor to be the most expensive unit, the purification of ethylene and propylene represented 47.5 % of the total investment (118.5 M USD). Indeed, a similar study by Yadav et al. [163] resulted in the equivalent conclusions. Similarly, Larrain et al. [162] also observed an increase in the investment cost when comparing a pyrolysis process configuration optimized to maximize naphtha (high-pressure pyrolysis) against another that produced both naphtha and wax.

Despite being capital-intensive, the purification of various value-added products can remarkably improve the economic feasibility of plastic waste pyrolysis. When comparing a process configuration optimized for naphtha production to another producing naphtha and wax, Larrain et al. [162] observed that, on average, producing wax and naphtha led to a higher return investment despite the higher capital cost of this configuration. Indeed, purifying the product stream leads to a higher yield of higher-valuable products, i.e., olefins, naphtha, aromatics, and wax. For example, in the study by Larrain et al. [162] the configuration of the amount of product produced from plastic was increased from 0.69 kg$_{product}$/kg$_{feedstock}$, for the naphtha-only design, to 0.75 kg$_{product}$/kg$_{feedstock,}$ when naphtha and wax were produced. Yadav et al. [163] and Gracida-Alvarez et al. [160] also showed the credits of the overall products and by-products after purification were essential for

Table 6

Summary of techno-economic analyses (TEA) for various plastic conversion processes using data from previous studies.

	Gasification		Pyrolysis				
Author	[21]	[21]	[160]	[161]	[162]	[162]	[163]
Technology	Steam gasification + Water-Gas Shift	Steam gasification + Methanol synthesis	Non-catalytic Pyrolysis	Pyrolysis using molten salt	High-pressure pyrolysis	Non-catalytic Pyrolysis	Catalytic Pyrolysis
Hydrotreatment	N.A.	N.A.	Yes	No	Yes	Yes	Yes
Feedstock	Mixed plastic waste	Mixed plastic waste	HDPE waste	Mixed plastic waste including 11.5 wt% PVC and 5.3 wt% PET	Mixed polyolefins	Mixed polyolefins	Mixed plastic waste including 4 % PVC
Target Product	Hydrogen	Methanol	Ethylene, propylene, aromatics, light hydrocarbons, heavy hydrocarbons	Naphtha + Wax mixture	Naphtha	Naphtha, Wax	Aromatics
By-products	–	–	methane	light Gas	Olefins, light Gas	Olefins, light Gas	Naphtha, olefins, light Gas
Feedstock cost (USD/ton)	0.60	0.60	0.22	0.65	0.57 [g]	0.57 [g]	0.60
Feed scale (tpd)	240	240	500	10	360	360	240
Product yield (kg$_{product}$/kg$_{feedstock}$)	0.29	1.45	Ethylene: 0.188 PP: 0.134 Aromatics: 0.037 Light hydrocarbons: 0.54 Heavy hydrocarbons: 0.054	0.79	0.69	Naphtha: 0.408 Wax: 0.342	0.22
Energy supply (MJ/kg$_{product}$)	62.6	15.2	11.2 [a]	0.0 [b]	−2.6 [b]	3.9 [b]	14.3
TIC (million USD)	145	149	118.5	4.4	91 – 148 [c]	103 – 194 [c]	107
MSP (USD/kg)	3.39	0.69	–	0.32	0.57	Wax:0.45 [c] Naphtha: 0.57 [c]	1.07 [d]
Market price (USD/kg)	1.15	0.30	–	0.72	0.72	0.72	0.69

N.A.: Not applicable.

[a] Average energy supply per kg of product.

[b] Includes credit from electricity or heat produced by burning the light gases from pyrolysis.

[c] 1 EUR (2020) = 1.14 (USD (2020).

[d] Includes credit from by-products, including olefins, Gas, naphtha, and C$_9$-C$_{12}$ aromatics.

the overall process economics.

Light gases, which can include H_2, CH_4, light alkanes, light olefins (if not separated), and CO, CO_2, and HCl, if polymers other than polyolefins, e.g., PET and PVC, are present in the feedstock are also a by-product from the pyrolysis process. While light gases have low or no market value, this product stream is commonly used to produce energy, particularly electricity, to be used in the process. The impact of the energy generated from the light gas combustion on the overall process energy consumption can be very significant. For instance, Jiang et al. [161] and Larrain et al. [162] which included a power turbine for light gases, estimated an energy supply for the process between -2.6 MJ/$kg_{product}$ to 3.9 MJ/$kg_{product}$, against an energy consumption of 11.2–14.2 MJ/$kg_{product}$ [142,145]. Furthermore, Larrain et al. [162] reported a sharp reduction in energy consumption, from 3.9 MJ/$kg_{product}$ to -2.6 MJ/$kg_{product}$ when the gas yield increased from 0.15 kg_{gas}/$kg_{feedstock}$ to 0.19 kg_{gas}/$kg_{feedstock}$. Thus, heat integration or electricity production is an alternative to full-gas purification, particularly when the process plant is not located near a petrochemical complex (consuming point) or the production scale does not justify the purification.

Unlike pyrolysis, the gasification of plastic can be selectively converted to multiple products with petrochemical value, including hydrogen, methanol, synthetic paraffins and olefins through the conversion of the syngas by water–gas shift, methanol synthesis, and Fischer-Tropsch processes. Singh et al. [21] compared the TEA of methanol and hydrogen production from the gasification of mixed plastic waste. In terms of capital investment, both pathways were very similar (145–149 M USD), with the main expenses being related to the methanol synthesis process (49 % of TCI) or water–gas shift (37 % of TCI) and pressure swing adsorption (17 % of TCI) processes when producing H_2. Additionally, independently of the final product, the gasification section of the plant was only responsible for 8 % of TCI, while the syngas cleaning represented around 21–22 %. By opposition, the two pathways significantly differ in the production capacity, with the pathway for hydrogen production yielding 0.29 kg_{H2}/$kg_{feedstock}$ and the methanol pathway 1.45 kg_{CH3OH}/$kg_{feedstock}$. It should be mentioned that the substantial difference between the two pathways' product yields is like the nature of the reactions taking place since the maximum possible of hydrogen and methanol from plastic waste are ≈ 0.4 kg_{H2}/$kg_{feedstock}$ and ≈ 2.67 kg_{CH3OH}/$kg_{feedstock}$, respectively. The substantial difference in yield can also explain the much higher energy consumption per H_2 produced, i.e., 62.6 MJ/kg_{H2} vs. 15.2 MJ/kg_{CH3OH}.

By comparing the minimum selling price (MSP) of the petrochemicals produced from fossil sources (Table 6), e.g., natural Gas and crude oil, it becomes clear that wax and naphtha production from mixed plastic waste is economically competitive over the equivalent fossil streams, i.e., MSP < market price. In opposition, aromatics, methanol, and hydrogen MSP are above the fossil equivalents, suggesting the design of plants dedicated explicitly to producing these chemicals might not be economically feasible. Independently of the process, the cost associated with the feedstock (0.22–0.65 USD/kg), i.e., mixed plastic waste, is the main responsible for the MSP, representing around 70 % of OPEX in the gasification processes [21]. Indeed, gasification, especially pyrolysis processes, cannot process all plastic waste. For instance, common polymers, like PVC and PET, are transformed into problematic by-products, e.g., HCl and terephthalic acid, which significantly affect the operation of the reactors and subsequent purification equipment. Indeed, a recent report on the feedstock quality guidelines for the pyrolysis of plastic waste states a minimum composition of 85 % of polyethylene and polypropylene is required [164]. Hence, a pre-treatment stage must be added to ensure the feedstock meets specifications, increasing its cost. Furthermore, the feedstock specification requirements lead to another important factor, often overlooked by TEA studies: feedstock availability. Most TEA studies focus on large processing plants, which favour the economic viability of the process. For instance, Larrain et al. [162] estimated a minimum operating capacity of

70 kt/year (210 tpd) and 115 kt/year (345 tpd) would be necessary for a pyrolysis plant producing wax and naphtha and exclusively naphtha, respectively, to achieve economic feasibility. Yet, Europe's average plastic consumption per capita is 112 kg, with only 43 wt% of those plastics being polyethylene and polypropylene [165,166]. Hence, a 70 kt/year plant would require at least the equivalent of 1.2 million people generating waste to provide enough feedstock for operation, which indicates the conversion of plastic waste into petrochemicals might be more suited to largely populated areas. It should be noted Jiang et al. [161] demonstrated that small pyrolysis plants might also be economically. However, the authors did not include the necessary hydrotreatment of the naphtha and wax products, which prevents a definitive conclusion on the economic viability of the process.

8. Life cycle analysis of retrieving plastic wastes for petrochemicals

Life Cycle Analyses (LCA) is a tool for analysis that determines environmental impacts of a material by taking into account production, processing, conversion, and disposal of the product [167]. LCA have been used to evaluate the consequences of various production and disposal methods in order to better comprehend the scope of the environmental waste accumulation problem [168,169]. This approach has already been utilised to comprehend the implications of creating specific unique polymers, including bio-based polymers [170–172]. There are few LCA studies that examine the possibilities of recycling, converting to fuel and energy, and disposing of synthetic polymers.

To further these efforts, Khoo, [173] used LCA to analyse the mechanical recycling of plastics in Singapore, evaluating the process' effects and calculating the benefits of transforming the recovered plastics into useful forms of energy. The study stressed the value of recycling facilities for managing significant amounts of plastic garbage in locations where landfill space is scarce. Kreiger et al. [174] investigated whether 3D printing may be an effective way to mechanically recycle HDPE waste in the US. This investigation came to the conclusion that by using this technology, 1 kg of HDPE could be produced with 89 % less energy. However, there is an inadequate information available concerning the environmental costs and energy requirements of various polymer processing stages. Furthermore, pyrolysis of plastic waste has not gained much consideration in comparison to the large number of LCA studies that focused on waste incineration [175].

However, the WasteBusters project used the LCA technique to examine the climatic implications associated with the pyrolysis of plastic trash [176]. In the study, the applied LCA technique was partially framed by Oasmaa et al. [177]. A screening carbon footprint assessment was completed using the key data from the pyrolysis and pretreatment phases of the pilot tests. Literature, data from accessible databases, and expert opinion served as additional data sources. The system under study aimed to utilize plastic waste as a feedstock for the manufacturing of polymers and fuel. Diesel and PE in quantities comparable to those synthesized during the pyrolysis process, which was the functional unit used in the study, were produced after 1 kg of plastic waste was treated. The analyses' results exhibited that the pyrolysis of plastic waste had a significantly lower carbon footprint than direct combustion of plastic waste and producing fuel and polymers from virgin materials. The alternative pyrolysis scenarios in the examined scenarios had carbon footprints that were 15–60 % lower than the business-as-usual scenario. Similar results were found by Perugini et al [178], which showed that, compared to plastic incineration in the baseline scenario, low temperature pyrolysis/hydrocracking combined with mechanical recycling of plastic packaging waste reduced the carbon footprint of landfilling plastic by 67 % and by 76 %.

Foolmaun and Ramjeeawon [179] explained the environmental impact of numerous manufacturing and processing methods in Mauritius, such as 100 % flake production, 100 % landfilling, 34 % flake production with 66 % landfilling, 100 % incineration with energy

recovery, and 50 % incineration paired with 50 % landfilling. It has been asserted that incineration with energy recovery was the most environmentally beneficial option compared to landfilling, which was the least preferred. This method is typically thought of as one of the most important and widely used ways to compare different environmental performances of disposal systems and recycling processes. Additionally, LCA study observed that, among the technologies, landfill gas production has the most significant carcinogenic gas emissions [29]. The importance of pyrolysis for the reduction of GHG emissions is shown by Demetrious and Crossin [180], who compared and analysed the uses of plastic waste in the process of pyrolysis, gasification, incineration, and landfilling. However, landfilling needs less energy in comparison to gasification-pyrolysis system. At the end, their analysis provided a blueprint for changing plastic waste management policies. The UK-based Syngas Products Gas Ltd seeks to produce renewable gas with a high calorific value from the transformation of plastic waste feedstock into energy through the combined use of pyrolysis and gasification [29]. The plastic management unit in Dorset can generate power using 0.8 MWe of input and 10 ktpa of plastic trash as a feedstock. Therefore, the type of the input fuel is the only factor that truly distinguishes combustion-based energy producing units from waste to energy conversion methods economically.

The sensitivity analysis carried out during the numerous LCA case studies revealed that the modelling approach employed, such as the kind of electricity and heat substituted, or relying on the correctness of the data set, may cause a significant change in the results [176,177]. The results are influenced by the energy generation profiles, expected yields for mechanical recycling, and speculation for the incineration of plastic waste. Therefore, even while the results appear encouraging, it is not viable to draw general perceptions based on only a few case studies [167]. Future research should also focus on gathering and using more precise data for the processes under study. The study's scope should be expanded to include other emissions and environmental implications in addition to climate change impacts, starting with categories for air emissions and resource depletion [11]. Potential effects related to toxicity should be taken into account if dangerous compounds are predicted in the raw material [13]. The pyrolysis product must adhere to the GHG reduction standards outlined in the recast of the Renewable energy directive (REDII) for recovered carbon fuels if it will be used to produce transportation fuel.

9. Opportunities and challenges in retrieving plastic waste

9.1. Opportunities in retrieving plastic waste

The overabundance and continue release of waste plastic in the environment and their adverse health impacts are significant issue today [10,16]. In order to avoid landfill or incineration, the circular economy is therefore being seen as a more favourable choice from an environmental, economic, social, and legal standpoint [58,181]. The circular economy concept emphasises resource recovery through reuse, recycling, and upcycling while integrating operational waste management activities into the forward supply chain [182]. By creating new industries and jobs, lowering emissions, and improving the efficient use of natural resources in a closed-loop supply chain, the circular economy model also helps responsible producers. Most crucially, the circular economy model fills a gap between managing waste and circulating material resources by treating waste as a resource [169]. However, in order to support the shift to a circular economy, precise activity planning is essential, and this planning cannot ignore knowledge of the techniques for retrieving plastic waste [17].

Although, treatment and management of plastic waste is highly challenging, however, high-value materials can be obtained by upcycling of plastic waste through pyrolysis, gasification, and carbonization. Since 2000, Japan has successfully processed plastic waste in coke ovens using a commercialized large-scale waste plastic carbonization process.

The plastic waste is heated along with coal at a very high temperature of 1100 °C inside the coke oven to generate value-added products like coke, hydrocarbon oils, and coke oven gas (COG). PVC waste generates high volumes of chlorine, which can be trapped using ammonia liquor and used to cool the COG. Another carbonization approach involves blending of mixed plastic waste (PVC, PP, PS, PE, and PET) with montmorillonite (organically modified) and heating at 700 °C, followed by the addition of KOH at 850 °C to yield carbon nanosheets with an activated porous structure. These nanosheets can be utilized for CO_2 capture and H_2 storage [183].

Chemical upcycling techniques can synthesize value-added products such as vitrimers from not only covalent adaptable networks [184] and liquid alkanes [185] but also from mixed polyolefin waste [186]. Recycled plastic wastes can also serve as adsorbents to eliminate co-pollutants [187], or as 3D printing filaments with possible upcycling applications [188], which expand the scope of research in this field. Another thermochemical process involving two-step pyrolysis of plastic waste can synthesize high-value materials like graphene nanosheets [189]. The two-step slow process includes initial pyrolysis with nano-clay at 400 °C (forms the backbone for graphene nanosheets by eliminating the oily hydrocarbons) and final pyrolysis at a higher temperature of 750 °C to generate the final graphene nanosheets. Their applications include drug delivery, fuel cells, energy storage, and supercapacitors [190, 191].

Recently, catalytic hydrocracking might emerge to be an efficient treatment method for plastic waste because, at a temperature of 225 °C, polyolefin waste gets converted to liquid fuel [192]. Hydrocracking processes can be facilitated by blending platinum-deposited tungstate-zirconia ($Pt/WO_3/ZrO_2$) with HY zeolite under mild conditions. Mixed plastic waste (PP, PS, PE) or polyolefin waste can be converted to lubricants and fuel-range hydrocarbons by hydrocracking. Similarly, LDPE undergoes hydrogenolysis within an hour in the presence of $Pt/W/\beta$-catalyst at 3.0 MPa H_2 and 250 °C, yielding 63.6 wt% (94.0 wt% of C_1–C_{13} light alkanes) of valuable C_5–C_{12} gasoline alkanes [193]. Moreover, under the same conditions, a high yield (50.8–73.5 wt%) of valuable gasoline alkanes (C_5–C_{12}) can be obtained by the hydrogenolysis of HDPE, PP, and light low-density polyethylene (LLDPE). These all studies revealed the successful utilization of plastics and/or mixed plastic waste as feedstock for generation of value-added fuels and chemicals at large scale. Nevertheless, several challenges are still persisting in retrieving plastic waste to fuels and chemical which we are discussed below.

9.2. Challenges in retrieving plastic waste

Pyrolysis is a commonly used process that can be applied to carbonize plastic waste to get value-added products (such fuels and carbon materials). Such procedures are, however, economically unsustainable because to the high industrial setup costs and intense temperatures in the range of 600–1000 °C [194]. Additionally, it is typically not practical to reuse the costly catalysts used in the pyrolysis and carbonization processes [195]. As, economic feasibility is required for industrial applications, so only profitable and cost-effective technologies are considered. Microwave irradiation is considered as efficient approach that has been used in several investigations since it can be performed at room temperature and has a relatively quick reaction time [196]. However, the effectiveness at a commercial (plant-size) scale is questionable and needs further study. Additionally, the byproducts of the reaction may cause the catalysts employed to treat plastic waste to become inactive [197]. Therefore, the issue of catalytic stability must be addressed in any technical innovation or futuristic design for the upcycling of plastic waste.

The emission of volatile organic compounds (VOCs) throughout the processing steps is another significant obstacle to commercializing these thermochemical upcycling processes [198]. The mechanisms and potential reactions that result in the production of these VOCs or other

harmful compounds need to be thoroughly studied in order to reduce the potential environmental risks. The lack of effective and practical waste segregation, drying, and cleaning techniques limits these upcycling technologies and processes [199]. Electrostatic separation [200] can be used to separate plastic polymers with similar densities (like PP and PE), however it is inefficient and difficult to manage the material. Another method to separate mixed plastic waste depending on polymeric density is the sink-float method [199]. An auto-sorting system can use near-infrared sorting of mixed plastic waste to separate various polymers that absorb near-infrared radiation at different wavelengths [67]. These automated, near-infrared online sorting systems successfully distinguish PS, PP, and PE polymeric blends. Numerous studies have concentrated on the viability and practical efficacy of various plastic waste sorting methods [200,201]. Although research in this area has grown, the industrial application of an upcycling or recycling technology can only be aided by future study that takes into account practical and affordable sorting methods for mixed plastic waste of unknown composition.

A significant obstacle is the presence of numerous hazardous chemicals and colourants in the plastic waste that is currently accessible as feedstock [13]. Additionally, plastic waste involves post-consumer contamination, which is a serious problem in underdeveloped countries because the segregation and purification steps incur additional costs to the process and make it unprofitable [10]. However, to make up for that cost element, businesses and other organizations are concentrating on innovative uses of the gathered plastic waste to create marketable products. For instance, a German business called Rewindo GmbH has been collecting PVC trash from outdated or demolished buildings by setting up a national transportation network, and the waste has been used to create new building materials [199]. Single-use plastic packaging materials are being collected, cleaned, separated, and sorted in India by a handloom business called "EcoKaari" in order to produce marketable woven yarn-strips. To repurpose PET waste with methanol, Teijin Fibres Ltd. of Japan developed a high-pressure, high-temperature methanolysis process [202]. Even while there are a few such approaches are commercialized globally, their number is still insignificant when compared to the yearly global plastic waste release in the environment.

10. Conclusions and outlooks

The problem of plastic waste mismanagement and environmental accumulation has become a global concern due to impending, potential impacts on human health and environmental sustainability. Biodegradable plastics are being proposed as potential alternatives to conventional plastics. However, many studies have concluded that conventional biodegradable plastics derived from PLA and polyhydroxy butyrate (PHB), could also be as hazardous to the environment as the non-biodegradable plastics. Adapting to a circular economy, sustainable management approaches are needed to tackle the environmental issues of synthetic plastic wastes. Hence, reuse, recycling, and plastic waste valorisation have attracted significant attention in recent years. Novel approaches, like plastic waste conversion to value-added products including petrochemicals are widely proposed as promising valorisation techniques.

In comparison to uncontrolled incineration and landfilling practises, the conversion of plastic wastes into liquid oil (fuel) and other valuable by-products like char and gases using thermochemical technology is thought to be a comparatively environmentally friendly process. Numerous process variables, including temperature, catalyst selection, heating rate, carrier gases, retention time, type of plastics, reactor, and pressure, affect the thermochemical products. From thermoplastic waste, up to 80–90 % of fuel oil by weight can be recovered. The liquid oils that are produced are either diesel or petrol fractions with properties similar to those of commercial diesel or petrol fuels. Additionally, the gases recovered from the thermochemical conversion of waste plastics are predominantly composed of C_1–C_4 compounds for the polyolefins. Although, high retention time and temperature are key parameters

which affect economy of the process, and that need to be optimised to make the procedure more cost-effective. Plastic chars produced by the pyrolysis of plastic waste can also be activated for use as a raw material in the manufacturing of other materials as well as for the absorption of heavy metals and gaseous pollutants from different environmental matrices.

The quality of liquids produced by the thermochemical process has been improved through the study of various catalysts. More study is specifically needed to explore and make use of less expensive naturally occurring catalysts such natural zeolites. Additionally, further consideration is needed for catalyst modification to enhance their functionality and optimise the thermochemical conversion process. It is important to examine the conversion efficiency and selectivity of nanoscale zeolite in conversion of plastic waste to fuel. It should also be researched how to include and combine catalysts into bimetallic, trimetallic, and other systems for application in catalytic conversion. In addition to using the most recent technologies, such as microwave irradiation, plasma, and continuous systems to provide feasible and marketable thermochemical technologies, efforts should be made to lower the process temperature in order to reduce energy consumption. Further, the exact thermochemical reaction mechanism of the polymers should be investigated using computer-based technologies and models.

Various chemical methods offer promising upcycling prospects for plastic wastes. But scale-up, collection systems and starting materials are a few of the many constraints linked with chemical upcycling. Therefore, owing to the rapidly growing plastic waste pollution and the need for economically and environmentally sustainable solutions, immediate attention and research are required in this field.

At present, a few developed countries have implemented legislative and policy approaches to reduce plastic manufacture, consumption, and waste generation and to increase their reuse. However, advocating these strategies for combined plastic waste management and formulating a universal approach to address different types of plastic wastes are challenging. Thus, potential plastic waste management strategies to address the global plastic pollution problem must begin with a sustainable infrastructure development, followed by socially acceptable and economically feasible valorisation technologies or policy regulations.

CRediT authorship contribution statement

Manish Kumar: Conceptualization, Validation, Visualization, Writing – original draft, Writing – review & editing, Investigation, Methodology. **Shiv Bolan:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Lokesh P. Padhye:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Muxina Konarova:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Shin Ying Foong:** Writing – review & editing. **Su Shiung Lam:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Stuart Wagland:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Runzi Cao:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Yang Li:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Nuno Rocha Batalha:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Mohamed Ahmed:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Ashok Pandey:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Kadambot H.M. Siddique:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Hailong Wang:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Jörg Rinklebe:** Validation, Visualization, Writing – original draft, Writing – review & editing. **Nanthi Bolan:** Conceptualization, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing – original draft, 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

No data was used for the research described in the article.

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