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REVIEW

Thermochemical conversion of plastic waste to fuels: a review

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Abstract

Plastics are common in our daily lifestyle, notably in the packaging of goods to reducing volume, enhancing transportation efficiency, keeping food fresh and preventing spoilage, manufacturing healthcare products, preserving drugs and insulating electrical components. Nonetheless, massive amounts of non-biodegradable plastic wastes are generated and end up in the environment, notably as microplastics. The worldwide industrial production of plastics has increased by nearly 80% since 2002. Based on the degree of recyclability, plastics are classified into seven major groups: polyethylene terephthalate, high-density polyethylene, polyvinyl chloride, low-density polyethylene, polypropylene, polystyrene and miscellaneous plastics. Recycling technologies can reduce the accumulation of plastic wastes, yet they also pollute the environment, consume energy, labor and capital cost. Here we review waste-to-energy technologies such as pyrolysis, liquefaction and gasification for transforming plastics into clean fuels and chemicals. We focus on thermochemical conversion technologies for the valorization of waste plastics. This technology reduces the diversion of plastics to landfills and oceans, reduces carbon footprints, and has high conversion efficiency and cost-effectiveness. Depending on the conversion method, plastics can be selectively converted either to bio-oil, bio-crude oil, synthesis gas, hydrogen or aromatic char. We discuss the influence of process parameters such as temperature, heating rate, feedstock concentration, reaction time, reactor type and catalysts. Reaction mechanisms, efficiency, merits and demerits of biological and thermochemical plastic conversion processes are also discussed.

Keywords Waste plastic · Waste-to-energy · Pyrolysis · Liquefaction · Gasification

Abbreviations

$(C_{10}H_8O_4)_n$	Ethylene phthalate
$(C_3H_6)_n$	Polypropylene
$(C_8H_8)_n$	Polystyrene
°C/min	Degree Celsius per minute
°C/s	Degree Celsius per second
°C	Degree Celsius
·H	Hydrogen radical
·OH	Hydroxyl radical
Al_2O_3	Aluminum oxide or alumina
ASTM	American Society for Testing and Materials
$Ba(OH)_2$	Barium hydroxide
C_2H_2	Acetylene
C_2H_4	Ethene or ethylene
C_2H_6	Ethane
C_3H_6	Propene
C_3H_8	Propane
C_4H_{10}	Butane
C_4H_8	Butene
$Ca(OH)_2$	Calcium hydroxide
CeO_2	Ceric oxide or ceria
CH_4	Methane
Co/Al_2O_3	Cobalt on alumina
Co/CeO_2	Cobalt on ceria
$Co/CeO_2–Al_2O_3$	Cobalt on ceria–alumina
CO	Carbon monoxide
CO_2	Carbon dioxide
cP	Centipoise
Cu/Al_2O_3	Copper on alumina
FDA	United States Food and Drug Administration
Fe/Al_2O_3	Iron on alumina
Fe_2O_3/CeO_2	Ferric oxide on ceria
FHYD/CA	Ferrihydrite treated with citric acid

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g/cm^3	Gram per cubic centimeter
g/h	Gram per hour
h	Hour
H^+	Cationic hydron
H_2	Hydrogen
H_2S	Hydrogen sulfide
HDPE	High-density polyethylene
HNZ	Protonated natural zeolite
HZSM-5	Protonated Zeolite Socony Mobil-5
IUPAC	International Union of Pure and Applied Chemistry
kcal/kg	Kilocalorie per kilogram
kJ/kg	Kilojoule per kilogram
kJ/mol	Kilojoule per mole
KOH	Potassium hydroxide
kW	Kilowatt
LDPE	Low-density polyethylene
m^2/g	Meter square per gram
$Mg(OH)_2$	Magnesium hydroxide
min	Minute
MJ/kg	Megajoule per kilogram
mm	Millimeter
mmol/g	Millimoles per gram
MMT	Million metric tons
MPa	Megapascal
N_2	Nitrogen
NAFTA	North American Free Trade Agreement
Ni/Al_2O_3	Nickel on alumina
$Ni/SiO_2–Al_2O_3$	Nickel on silica/alumina
nm	Nanometer
NO_x	Nitrogen oxides
O_2	Oxygen
OH^-	Anionic hydroxide
P_c	Critical pressure
PET or PETE	Polyethylene terephthalate
pH	Potential of hydrogen
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinyl chloride
RIC	Resin Identification Code
Ru/Al_2O_3	Ruthenium on alumina
RuO_2	Ruthenium(IV) oxide
s	Second
SiO_2/Al_2O_3	Silica–alumina
SO_x	Sulfur oxides
T_c	Critical temperature
wt%	Weight percent
µm	Micrometer

Introduction

Plastics are a key element of our lifestyle and day-to-day activities. The lightness, dexterity, versatility and low production cost have made plastics one of the most desired materials for utility in several sectors by innumerable ways such as our standard of living, food and goods packaging, construction, electronics, biomedical, engineering, automotive, aerospace, transportation, leisure and many more. Plastics are synthesized from organic polymers or long chains of carbon atoms in addition to hydrogen, oxygen, nitrogen, sulfur, and chlorine. The long chains of polymers have several thousand repeating units of monomers, which makes plastic durable. This property of plastics makes it last in the environment for hundreds of years to degrade naturally. It should also be noted that fossil fuels account for 99% of the raw materials base for plastic production (British Plastics Federation 2020).

Approximately, 4% of the fossil fuels produced globally are used to manufacture plastics (Ahorsu et al. 2018) and another 4% is used to generate power for the plastic manufacturing industries (Gourmelon 2015). From 1950 to 2012, the worldwide production rate of plastics averaged 8.7% per year, booming from 1.5 million metric tons (MMT) to the nearly 335 MMT in 2016 and 359 MMT in 2018 (Statista 2020a). Plastics account for nearly 30% of packaging sales and makeup about 10% by weight of a typical US motor vehicle, i.e., 152 kg of plastic used per vehicle (Gourmelon 2015). This is in contrast to the mere 9% of plastics previously used in a single motor vehicle in the 1960s. The production of plastics requires less energy, i.e., 62–108 MJ/kg than that of producing silicon, i.e., 235 MJ/kg (Statista 2020a).

Enormous amounts of waste plastics end up in the municipal solid waste and are eventually disposed of in landfills and oceans, which causes serious environmental and ecological problems. Plastics are resilient to physical, chemical and biological degradation, thus they tend to be stable in the landfills, oceans and coastal seawater for decades to centuries. Each year, nearly 10–20 million tons of plastics end up in the oceans (Gourmelon 2015). It is estimated that more than 5 trillion plastic fragments of variable sizes or shapes and weighing a total of over 250,000 tons currently floating in the world's oceans (Eriksen et al. 2014). Because of this massive amount of plastic debris including microplastics in the oceans, annual expenditures approaching $13 billion in losses are encountered through ecological damages to marine habitats, financial damages to aquaculture, pisciculture and tourism as well as remediation technologies implemented for cleaning polluted water bodies (Gourmelon 2015).

The discarded plastic debris in the oceans degrades into mesoplastic and microplastic fragments that directly affect marine life and poses a direct risk to planktonic and invertebrate organisms and an indirect risk to human health as it enters the food web through aquatic animals (Kwon et al. 2014; Padervand et al. 2020). The typical average particle size of mesoplastics and microplastics are in the range of 1–10 mm and 1–1000 µm, respectively. Aquatic animals such as fishes, whales and dolphins as well as seabirds are highly vulnerable to microplastic fragments in the oceans as they can be entangled, trapped and smothered and even ingest the debris while feeding (Cole et al. 2011; Wright et al. 2013). Microplastics and fine plastic debris can serve as vectors for transporting persistent toxic substances including hazardous chemicals and pathogens, which could enter the food web through biomagnification. Microplastics are potentially toxic because of the unreacted monomers, oligomers, and chemical additives such as phthalates, polybrominated diphenyl ethers and bisphenol A, which can be absorbed by tissues of the host organism to interfere with the cell division, metabolism and physiology while leading to serious health implications (Smith et al. 2018).

Microplastics having a particle size less than 5 mm are usually pervasive in the air, water and soil for longer durations with higher stability and slower degradation rates (Padervand et al. 2020). Fragmentation, exfoliation, wear and tear, abrasion, and gradual degradation of plastic products under atmospheric or induced conditions tend to release microplastics into the environment (Zhang et al. 2018; Padervand et al. 2020). Owing to their ubiquitous nature, microplastics have marked their presence in industrial effluents and sediments (Deng et al. 2020), subtidal sediments (Graham and Thompson 2009), deep-sea sediments (Van Cauwenberghe et al. 2013), oceans (Law et al. 2014), freshwaters (Li et al. 2020), beaches (Bissen and Chawchai 2020), islands (González-Hernández et al. 2020), urban and rural areas (Yin et al. 2020), intertidal ecosystems (Mathalon and Hill 2014), and even in Arctic Ocean sea ice (Kanhai et al. 2020) and Antarctic terrestrial food webs (Bergami et al. 2020). Padervand et al. (2020) have comprehensively reviewed several techniques to sequester microplastics from the environment, a few of which include adsorption on green algae, membrane-based sorption, filtration, coagulation, electrocoagulation, sedimentation, ultrafiltration, agglomeration, photocatalysis and ingestion–retention–trapping–adhesion by marine organisms and microorganisms.

Al-Salem et al. (2017) have discussed different primary, secondary, tertiary and quaternary treatment routes for managing plastic wastes. In the primary route, waste plastics are re-introduced into the heating cycle of a polymer processing plant to increase its production rate. This route for reusing waste plastics can reduce the processing cost and close the recycling loop. Mechanical recycling is a secondary treatment where waste plastics are extruded and blended with virgin polymers to reduce the overall processing cost. However, the primary and secondary treatments are sensitive to the type of waste plastic being reprocessed with virgin plastic. The contamination of the virgin plastic with a different type of waste plastic and the presence of impurities such as dyes, chemical compounds and inert materials can alter the chemical properties, physical integrity and durability of the final recycled plastic products. Chemical treatment methods are categorized as tertiary methods for waste plastic management. Such methods involve chemical and thermal agents to alter the polymeric structure of the finished product. Last but not the least, energy recovery is a quaternary treatment method for using waste plastics for generating energy in the form of heat and electricity through combustion. This is usually performed for cogeneration or combined heat and power generation for the plastic producing and recycling industrial facilities.

The major setbacks in implementing most of the plastic waste management operations are the high demand for energy and associated utilities. Moreover, recycling of waste plastics has many impediments in terms of process constraints, high expenditures, energy intensiveness, labor intensiveness, separation problems, compromised recycled product quality and environmental pollution (Sharuddin et al. 2016). The environmental pollution associated with the industrial recycling of waste plastics is the massive emissions of greenhouse gases, toxic obnoxious gases and wastewater containing non-biodegradable polymeric compounds (Sharuddin et al. 2016). During recycling, waste plastics are subjected to heat treatment, which leads to molecular alterations such as chain scission, cross-linking, depolymerization and formation of double bonds in the plastic polymers, thereby reducing the durability and integrity of the recycled plastic product (Al-Salem et al. 2017). Therefore, owing to the rigorous quality requirements of plastic products in the consumer market, recycling waste plastics tends to be a challenging task. With the cumulating plastic wastes generated annually, significant amounts of fossil fuels are required to fulfill the recycling operations, which is environmentally undesirable. It is reported that around 4% of the world's crude oil supply is dedicated to plastic industries (Al-Salem et al. 2017).

Plastic polymers are in principle the derivatives of fossil fuels and petrochemicals. Hence, their decomposition through thermochemical and catalytic routes could generate green fuels and chemicals. However, there is a lack of available literature on the utilization of plastics for waste-to-energy conversion in a closed-loop system. There is an immediate need to explore alternative and eco-friendly methods, which could valorize waste plastics to generate value-added industrial products. With this objective, the current paper broadly reviews waste plastics as an attractive feedstock for alternative fuel and chemical production through thermochemical technologies, particularly pyrolysis, liquefaction and gasification. The paper systematically reviews the classification of different plastic wastes and their decomposition behavior via biodegradation and thermochemical conversion. The physicochemical properties of the fuel products derived from plastic wastes are discussed along with the reaction mechanisms, advantages and technical limitations of each conversion process.

Global scenario for plastic waste production and recycling

Rapid urbanization, economic development, population growth and demand for routine commodities have pointedly increased the generation of municipal solid waste. Municipal solid waste usually includes organic residues, i.e., kitchen and yard wastes, waste paper, plastic wastes, glass, metals and miscellaneous garbage. The miscellaneous garbage consists of pharmaceuticals, electronic wastes, construction, renovation and demolition wastes. The management practices for municipal solid waste consists of sequential platforms such as (i) generation and initial sorting of wastes at the origin; (ii) collection of waste through garbage collection trucks by the local municipality; (iii) transfer of wastes to material recovery facilities; and (iv) disposal, processing and recycling of wastes.

Every year, nearly 1.9 billion tons of municipal solid waste is generated worldwide, of which around 30% remain uncollected by municipalities (Waste Atlas 2019). The production of municipal solid waste is projected for escalation to 3.4 billion tons by 2050 (The World Bank 2019). Today, about 70% of solid waste is dumped in landfills, 19% is recycled and 11% is diverted to waste-to-energy conversion facilities for energy recovery. Much of the plastic waste is landfilled because of the low cost of disposal than recycling. Capital cost is involved in each step of plastic recycling programs such as plastic waste collection and handling, transportation and logistics, sorting and material recovery, classification, washing, shredding, electricity and heat energy input, cooling water and wastewater recycling. Depending on the municipality and country, the cost can vary from the US $65–400/ton (Kunwar et al. 2016). In the USA, the curbside pick-up of plastics can cost around $50–150/ton and the landfilling can cost nearly $25–150/ton. Because of the low cost, landfilling of waste plastics has been practiced for the last few decades than a diversion for energy and fuel recovery.

The composition of municipal solid waste typically consists of biodegradable and non-biodegradable materials from both organic and inorganic sources. Moreover, the composition of municipal solid waste largely differs among different municipalities across the world and depends on the socioeconomic profile of the residents, demographics and local geographical conditions. Figure 1 shows the typical composition of municipal solid waste between different socioeconomic or income groups at a global level. The proportion of organic wastes is found to be higher in the case of low and middle-income groups. On an average basis, municipal solid waste collected from rural areas with the majority of low-income groups contains nearly 40–85% of organic matter (Worldwatch Institute 2012). On the contrary, the high-income group demonstrates comparatively greater levels of waste paper, metals, glasses and miscellaneous garbage. Plastic wastes were found to be the highest in the middle-income group followed by the high-income group.

The heating value of municipal solid waste from developing countries with a majority of low and middle-income groups is relatively lower due to the higher composition of high moisture-containing organic wastes. For example, the heating value of municipal solid waste collected

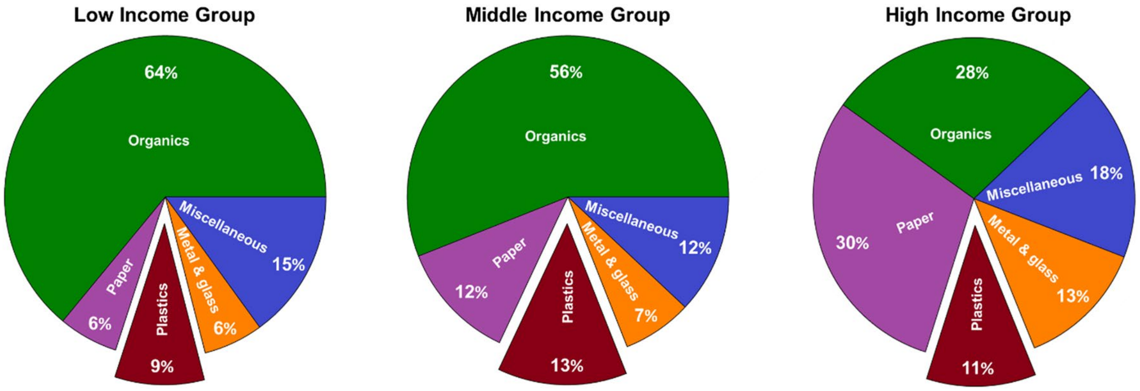

Fig. 1 Typical composition of municipal solid waste in different socio-economic groups worldwide. The higher proportion of organic wastes in low and middle-income groups should be noted. Additionally, paper waste, metal and glasses as well as miscellaneous wastes are in higher proportions in high-income groups. Data source: Kumar and Samadder (2017)

in a few developing countries decreased as: Malaysia, 1500–2600 kcal/kg > China, 1200–1600 kcal/kg > Thailand, 500–1500 kcal/kg > Sri Lanka, 950–1250 kcal/kg > India, 800–1100 kcal/kg > Bangladesh, 717 kcal/kg (Kumar and Samadder 2017). Conversely, plastic and paper wastes are dramatically higher in municipal solid waste generated in developed countries with a majority of the high-income group. The presence of plastics and paper results contributes to the comparatively higher heating value of municipal solid waste due to lower moisture content and greater carbon content. For instance, the heating value of municipal solid waste collected in a few developed countries decreased as: South Korea, 2600–3000 kcal/kg > the UK, 2200–3000 kcal/kg > Japan, 2000–2200 kcal/kg (Kumar and Samadder 2017). Developed countries are economically benefited due to the large amounts of plastic and paper wastes, metals and glasses in their solid waste stream. This is because the annual global market value for recycling paper wastes and scrap metal is over $30 billion (Worldwatch Institute 2012).

A few decades ago, plastics initially constituted less than 1% of municipal solid waste by weight in the USA and several other countries and with the years, this proportion has risen by an order of magnitude (Jambeck et al. 2015). By 2017, plastics made up at least 10% of municipal solid waste by mass as reported by The World Bank (2019). The worldwide production of plastics has dramatically elevated from 1.5 MMT in 1950 to whooping 359 MMT in 2018 (Statista 2020a). The demand for plastics has been increasing at a rate of 5% per annum (Kunwar et al. 2016). Figure 2 shows the trend in the production of plastics over the years. Nearly 8300 MMT of virgin plastics have been produced globally since the 1950s, of which 79% is landfilled, 12%, i.e., 800 MMT is incinerated and only 9%, i.e., 600 MMT is recycled (Geyer et al. 2017). It is projected that if the current patterns of plastic production, use and waste management continue, then by 2050, nearly 9000 MMT of plastic wastes will be recycled, 12,000 MMT incinerated, and 12,000 MMT discarded in the landfills (Geyer et al. 2017). Another report also suggests that about 8 MMT of plastics also end up in the oceans every year (Guern 2018). Every year, an average person in North America and Europe consumes about 100 kg of plastics in the form of packaging for commodities (Gourmelon 2015). The unremitting demand for plastics is leading to the intensifying accumulation of its wastes.

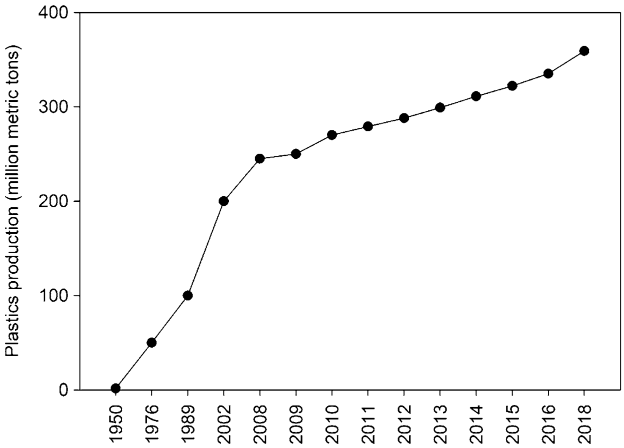

Fig. 2 Global plastic production over the years, in million metric tons. (Data source: Statista 2020a). This figure shows a trend of gradual increase in the production of plastics at a worldwide scale. The industrial production of plastics has increased by 80% and 33% since 2002 and 2010, respectively

The USA leads the world with its highest municipal solid waste generation rate of about 624,700 tons/day, whereas China ranks second with 520,548 tons/day of municipal solid waste (Hoornweg and Bhada-Tata 2012). Over the last 3 decades, China has landfilled more than 3 billion tons of municipal solid waste, whereas it currently dumps nearly 73% of its wastes in 547 operational landfills (Zhou et al. 2015). China is also one of the largest plastic producers in the world with more than a quarter share of global production (Statista 2020a). Nearly, 58 million tons of plastics were manufactured in China in 2012, which generated domestic revenues of 1.65 trillion Yuan (Statista 2018). With increasing exports to most of the developed countries, China is foreseen to scale-up its plastic production as more industries that are efficient continue to produce versatile high-quality plastic products.

Figure 3 depicts the trend in worldwide plastic production in 2016. The total worldwide production of plastics in 2016 amounted to some 355 MMT (Statista 2018). Of the total global plastic production, the North American Free Trade Agreement or NAFTA countries, which include the USA, Canada, and Mexico had 18% of the share. Europe produced 19% of the total plastics. It is reported that about 25.8 million tonnes of plastic wastes are generated in Europe of which 39% is landfilled, 26% recycled and 36% is utilized for energy recovery (Sharuddin et al. 2016; Al-Salem et al. 2017). Russian Commonwealth states, Japan, Latin America, Middle East and Africa demonstrated low plastic production. China exceeded all other nations in ranking as the topmost plastic producer with 29% of the global share, while the rest of Asia attributed to 17%.

Owing to the increasing plastic demands, China produces and exports much of its virgin plastic products to many developed countries. Besides, it also imports massive amounts of waste plastic from the USA, Europe and other countries, which accounts for about 56% of the total waste plastic imports worldwide (Gourmelon 2015). Incineration of plastics for energy recovery requires strict pollutant emissions controls as it produces toxic gases such as dioxins, furans, CO, CO_2, H_2S, SO_x, NO_x and other greenhouse gases, hazardous fly ash, coke and wastewater containing

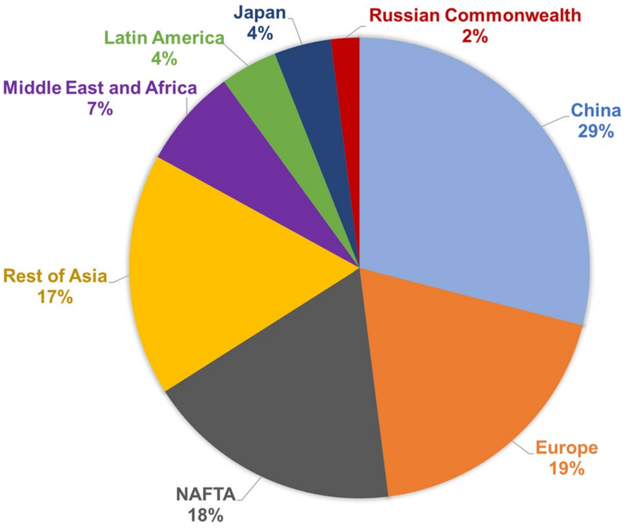

Fig. 3 Current global plastic production by region. The figure illustrates the dominance of China in plastic production followed by Europe; North American Free Trade Agreement countries, i.e., the USA, Canada and Mexico; and other collective Asian countries. Russian Commonwealth states reveal the lowest plastic production. Data source: Statista (2018)

polymeric degradation products. These environmental pollutants require effective remediation treatments before their disposal, which can considerably add to the overall process expenditures. Due to stringent environmental regulations in most developed nations for plastic disposal or incineration, much of the waste plastics collected for recycling is despatched to countries where environmental regulation controls are less implemented. However, recently in China and other developing countries, the government is devising and effectively implementing standard protocols and emission controls to reduce pollution because of plastic production and recycling.

End-of-life tires are also an integral component of solid wastes. Owing to rapid industrialization, tires are widely used in vehicles employed in several sectors such as commercial transportation, cargo, freight, agricultural practices, industries and aviation. The current estimated growth rate for the demand of tires worldwide is 4.3% per year (Machin et al. 2017a). The manufacturing of tires has been steadily increasing over recent years, for example, from 2.9 billion units in 2014 to 3.7 billion units in 2018 (Statista 2020b). According to the recent estimations, approximately 1.6 billion units of new tires are manufactured globally and nearly 1 billion units of waste tires are generated, of which the recycling industries process only 100 million units annually (Goldstein Research 2020). In the present scenario, the USA and Canada produce nearly 313 million and 35 million units of waste tires, respectively (Takallou 2015), whereas Brazil generates 20 million units (Machin et al. 2017a).

The durability and resistance of tires against wear and tear, fracture, friction, abrasion as well as variable pressures and temperatures is attributed to their composition, which includes synthetic rubber, steel wires, nylon, fabric, polymers, carbon black and some chemical additives (Hita et al. 2016). The rubbers widely used in tire manufacturing are styrene–butadiene, isoprene and polybutadiene (Seidelt et al. 2006). The rubber polymers are vulcanized with the application of cross-linking promoters, e.g., sulfur and accelerators at extreme temperatures to render strength and durable mechanical properties to the tires (Martínez et al. 2013; Labaki and Jeguirim 2017). Considering the non-biodegradable nature of waste tires, there are limited options for their disposal and recycling. While landfilling of scrap tires is not permissible, incineration is also restricted owing to the larger extent of toxic and greenhouse gas emissions. Scrap tires are mostly recycled to manufacture coatings materials for waterproof roofing of large infrastructures, road pavements and flooring of public arenas and sports fields. The typical calorific value of waste tires, i.e., 34,890 kJ/kg is equivalent to that of coals, i.e., 16,050–33,355 kJ/kg, suggesting their diversion to waste-to-energy conversion facilities as a feasible and benign alternative to fossil fuels (Nanda et al. 2019b).

From a global perspective, plastic wastes can be perceived to be valuable, low-cost and bountiful with a continual supply chain worldwide. Recovering plastics from the municipal solid waste stream for recycling and/or diverting to generate energy has the potential to minimize the problems of landfilling and ocean disposal. A circular economy approach of diverting the plastic wastes for alternative fuel and chemical generation is shown in Fig. 4. Such an approach can close the loop of a system in the efforts to prevent no outflow of the products and intermediates while, at the same time, achieving their direct or alternative utilization. One of the mature waste-to-energy conversion technologies, i.e., incineration has been widely applied to completely utilize municipal solid waste containing plastic wastes for reducing volume and meeting the rising energy demands. However, the massive amounts of resulting greenhouse gases are also a matter of serious environmental concern. On the contrary, the thermochemical waste-to-energy processes such as pyrolysis, liquefaction and gasification could potentially transform plastics among other organic wastes into fuel oil, combustible gases and aromatic char with high calorific values and superior material properties (Nanda et al. 2014, 2018b). These waste-to-energy conversion technologies can be a first-hand solution to the disposal issues of plastic wastes in landfills and oceans. The steady escalation of plastic wastes generation along with the requirement for eco-friendly waste management practices can contribute to exploring their thermochemical valorization pathways.

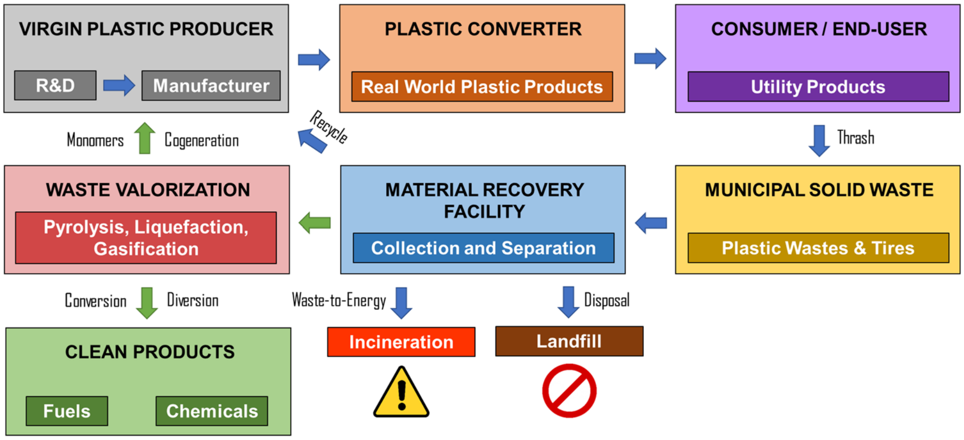

Fig. 4 Concept of a circular economy for plastic waste management. The circular economy model involving waste plastics as the feedstock can minimize its accumulation while enhancing its valorization as well as resource and value recovery. As per this concept, the disposal of plastics to landfills and oceans can be mitigated

Classification of plastics

The following basic steps are used to produce plastics: (i) synthesis of the basic polymer from the raw material, (ii) polymer compounding into a usable fraction, and (iii) molding the plastic. The additives, dyes, pigments and other compounds can be added during the polymer production process. There are several ways to classify the plastics in terms of their source, chemical structure, synthesis process, physical and mechanical properties, product design, degradability and recyclability. Based on the source, waste plastics can be classified into either industrial or municipal plastics (Kunwar et al. 2016). The industrial waste plastics are relatively more homogeneous with less contamination; hence, they are suitable for down-cycling into recovered byproducts and lower-grade plastics. In contrast, municipal waste plastics are largely heterogeneous containing more extraneous materials, which make the recycling process expensive and challenging. Therefore, a significant proportion of municipal waste plastics are landfilled.

Thermoplastics and thermosetting plastics are the most common classes of plastics based on their permeance of the form factor. Upon heat treatment, thermoplastics do not usually undergo chemical alterations in their composition and can be molded and reshaped again. On the contrary, thermosetting plastics, when heated, can undergo irreversible chemical reactions followed by melting and molding only once before solidification. In the thermosetting process, a chemical reaction occurs that is irreversible. While polyethylene, polypropylene, polystyrene and polyvinyl chloride are the examples of thermosetting plastics, rubber represents thermosetting plastics. Based on the degree of crystallinity and molecular structure, plastics can also be grouped into the following categories: (i) amorphous plastics, e.g., polystyrene and polymethyl methacrylate, and (ii) crystalline or semicrystalline plastics, e.g., polyamides, polyesters, polyethylene, polypropylene, polyvinyl chloride and some polyurethanes. Based on the degradability, plastics are also classed into biodegradable and non-biodegradable plastics. In contrast to conventional plastics that are made from petrochemicals, bioplastics made from plant residues and biopolymers are biodegradable. Bioplastics can degrade naturally or under induced conditions such as upon the exposure to sunlight or ultraviolet radiation, water, microorganisms, enzymes and oxygen levels (Kale et al. 2007). Recently, the growing interest in waste biomass for the production of bioplastics has resulted in its industrial production of 2.1 million tons in 2018 (British Plastics Federation 2020).

The most important of all categorization systems, the standard classification of mixed plastics are based on their identification mark or symbols administered by the American Society for Testing and Materials, i.e., ASTM International Resin Identification Coding, i.e., RIC System (ASTM 2014). The symbols or RIC identify the type of plastic resin in a plastic product. The classification of plastics was originally established in 1988 by the Society of the Plastics Industry, which is now known as the Plastics Industry Association. Since 2008, ASTM International has administered the plastic classification system with the issuance of the Standard Practice for Coding Plastic Manufactured Articles for Resin Identification, i.e., ASTM D7611. Table 1 summarizes the general properties of seven main classes of plastic resins such as polyethylene terephthalate, high-density polyethylene, polyvinyl chloride, low-density polyethylene, polypropylene, polystyrene and others, e.g., nylon, acrylonitrile butadiene styrene, polycarbonate, polylactic acid, and multilayered mixed polymers. Figure 5 illustrates the chemical structures of the above-mentioned plastic resins.

Polyethylene terephthalate has the International Union of Pure and Applied Chemistry or IUPAC name of poly(ethylbenzene-1,4-dicarboxylate), the chemical formula of $(C_{10}H_8O_4)_n$ and is characterized by the resin identification code 1. Among the polyester family, polyethylene terephthalate is the most widely applied thermoplastic polymer resin with applications varying from food packaging, e.g., bottled water, carbonated drinks, fruit juices and beverages

Table 1 Classification and properties of plastics

Plastic type	Resin identification code	Chemical and physical properties	Examples of final products
Polyethylene terephthalate	1 PETE	Recycling number or resin identification code: 1 Chemical formula: $(C_{10}H_8O_4)_n$ Density: 1.38 g/cm^3 Melting point: > 250 °C Boiling point: > 350 °C Lower heating value: 22.07 MJ/kg Resistant to high heat and solvent Barrier to gases and moisture	Potable water bottles Beverage bottles Food trays Medicine jars Clothing and carpet fiber Electrical insulations X-ray and photographic films
High-density polyethylene	2 HDPE	Recycling number or resin identification code: 2 Chemical formula: $(C_2H_4)_n$ Density: 0.941–0.96 g/cm^3 Melting point: 130 °C Lower heating value: 42.2 MJ/kg Relatively hard and strong Resistant to chemical and physical stress Barrier to moisture Permeability to gases	Three-dimensional printer filament Bottle caps Detergent and bleach bottles Milk bottles Buckets Coax cable insulation Electrical plumbing boxes Food storage containers Shoe last Plastic lumber Piping for water and sewer Plastic surgery Plant pots Compost containers Garden furniture Storage sheds
Polyvinyl chloride	03 PVC	Recycling number or resin identification code: 3 Chemical formula: $(C_2H_3Cl)_n$ Density: 1.38 g/cm^3 Melting point: 100–260 °C Lower heating value: 22.26 MJ/kg Resistant to chemicals Negligible permeability to gases	Credit card Vinyl records Wire rope Cable sheathing Construction flooring Door and window frames Plumbing pipes and fittings Ceiling tiles Home playground, toys, etc.
Low-density polyethylene	4 LDPE	Recycling number or resin identification code: 4 Chemical formula: $(C_2H_4)_n$ Density: 0.91–0.925 g/cm^3 Melting point: 115 °C Barrier to moisture Relatively soft and flexible	Shampoo bottles Detergent bottles Flexible bottles Edible oil containers Plastic cans Irrigation pipes Packaging bags Bubble wrap Shopping bags
Polypropylene	5 PP	Recycling number or resin identification code: 5 Chemical formula: $(C_3H_6)_n$ Density: 0.855 g/cm^3 Melting point: 130–171 °C Lower heating value: 41.04 MJ/kg Resistant to chemicals and heat Tough but flexible	Bottle caps Chips bags Biscuit wrappers Drinking straws Heavy-duty bags Plant pots Crates Chairs and desks File folders Tarpaulin Car bumpers

Table 1 (continued)

Plastic type	Resin identification code	Chemical and physical properties	Examples of final products
Polystyrene	6 PS	Recycling number or resin identification code: 6 Chemical formula: $(C_8H_8)_n$ Density: 0.96–1.04 g/cm^3 Melting point: 240 °C Flexible and brittle Less resistance to fats and solvents	Flexible plastics Packing peanuts Styrofoam Plastic cups Disposable cutlery Fast food trays Egg boxes Coat hangers
Others. This category includes polybutylene terephthalate, polycarbonate, polylactic acid, acrylic, acrylonitrile butadiene styrene, multilayered mixed polymers and nylon.	7 OTHER	Recycling number or resin identification code: 7 Difficult to recycle	Baby bottles Plastic lumber Safety shields Safety glasses Headlight lenses Compact discs, digital versatile discs, Blu-ray discs Automotive, aircraft and railway components Luggage casing Musical instruments Toys

References: Heikkinen et al. (2004), Wilkes et al. (2005), Batra (2014), Sharuddin et al. (2016), IFA (2018) and Lenntech (2020)

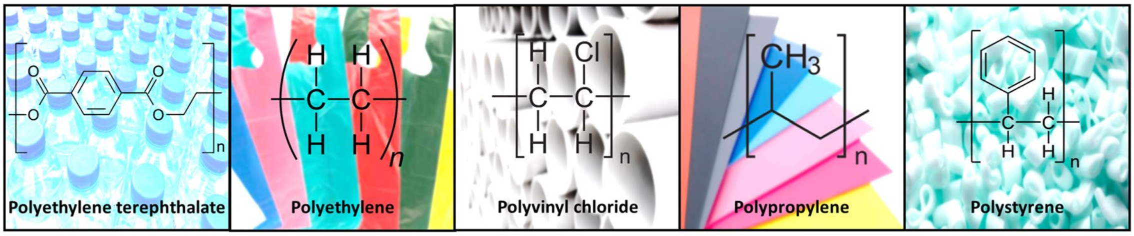

Fig. 5 Chemical structures and applications of some everyday utility plastic resins. As illustrated in this collage, some selected commercial applications of polyethylene terephthalate, polyethylene, polyvinyl chloride, polypropylene and polystyrene are found in the manufacturing of potable water bottles, shopping bags, plumbing pipes, file folders and styrofoam, respectively

to fibers for clothing and carpets. The high melting and boiling points of polyethylene terephthalate, as well as its resistance to heat and solvents, making it favorable for wide-scale applications. Polyethylene terephthalate is used to make many commercial items for household use such as mineral water bottles, beverage bottles, food trays, medicine jars, clothing and carpet fiber, electrical insulations, X-ray and photographic films. Although polyethylene terephthalate provides a certain barrier to gases and moisture, it could also absorb odors and flavors from foods and drinks. The items made from polyethylene terephthalate are recyclable but their extensive utility results in excessive accumulation of waste in the landfills (Sharuddin et al. 2016).

High-density polyethylene is thermoplastic with a long linear polymer chain that has a high degree of crystallinity, less branching, stronger intermolecular forces and greater tensile strength. The IUPAC name and chemical formula of its precursor polyethylene is poly(methylene) and $(C_2H_4)_n$. High-density polyethylene is identified by the resin identification code 2 and is commonly recycled. The slightly larger density of high-density polyethylene, i.e., 0.94–0.96 g/cm^3 than that of low-density polyethylene, i.e., 0.91–0.93 g/cm^3 makes it more rigid and less flexible. The large strength-to-density ratio has made high-density polyethylene an attractive material for many industrial, commercial, household and biomedical applications. A few of its applications include use in three-dimensional printer filament, bottle caps, detergent and bleach bottles, milk bottles, buckets, coax cable insulation, electrical plumbing boxes, food storage containers, shoe last, plastic lumber, piping for water and sewer, plastic surgery, plant pots, compost containers, garden furniture and storage sheds.

With such widespread utilities, high-density polyethylene contributes to nearly 18% of total plastics in municipal solid waste, thus making it the third-largest plastic waste category (Sharuddin et al. 2016). The highest consumer market for high-density polyethylene is the Asia-Pacific region with a global market share of 48% (Ceresana 2020a). Nevertheless, the worldwide demand for high-density polyethylene is expected to increase by 2.9% per annum until 2026. As one of the key applications, high-density polyethylene geomembranes are used in the bottom liners for sanitary landfills to create a homogeneous chemical-resistant barrier and prevent the permeation of highly contagious municipal waste leachate into the soil and groundwater (Rowe and Islam 2009).

Polyvinyl chloride is characterized by the resin identification code 3. Polyvinyl chloride is a polymer prepared from a vinyl chloride monomer, which reflects on its IUPAC name as poly(1-chloroethylene). After polyethylene and polypropylene, polyvinyl chloride is the third-most extensively produced synthetic plastic polymer at a global scale (Fischer et al. 2014). Polyvinyl chloride is used mostly in construction such as construction flooring, door and window frames, plumbing pipes and fittings, ceiling tiles, home playgrounds as well as vinyl records, wire rope, cable sheathing, toys and credit card. The mixing of additives during the production of polyvinyl chloride such as heat stabilizers, plasticizers, fillers and lubricants can affect its physicochemical and mechanical properties (Fischer et al. 2014). Polyvinyl chloride is manufactured usually from a combination of 43% carbon, i.e., ethylene or natural gas and 57% chlorine, which makes it resistant of chemicals, less permeable to gases and relatively resistant to heat and fire in contrast to polyethylene, polypropylene and polystyrene (Sharuddin et al. 2016).

Low-density polyethylene, distinguished by the resin identification code 4, has more branching than high-density polyethylene, which results in a weaker intermolecular force, lower degree of crystallinity, less tensile strength and hardness. However, more side branching and fewer crystallinity results in rendering low-density polyethylene better ductility and easy molding than high-density polyethylene (Sharuddin et al. 2016). Low-density polyethylene has offered an excellent barrier to water, which allows its largescale use in packaging industries, flexible bottles for shampoo and detergent, edible oil containers, plastic cans and irrigation pipes. In the packaging industries, low-density polyethylene is used in commercial products such as plastic bags, grocery bags, garbage bags, bubble wraps and wrapping foils.

The Asia-Pacific region also leads the global market share for low-density polyethylene processing of over 7.1 million tons in 2013. The multifarious use of low-density polyethylene plastics in our routine lives generates surplus wastes to rank it the second most plastic waste in municipal solid waste after polypropylene (Sharuddin et al. 2016). Nonetheless, with a current share of 41% of the total low-density polyethylene consumption by the Asia-Pacific countries, the usage of low-density polyethylene products increases at a rate of 2.5% per annum globally (Ceresana 2020b).

Polypropylene has the IUPAC name of poly(propene), the chemical formula of $(C_3H_6)_n$ and the resin identification code 5. It is a thermoplastic polymer belonging to a group of polyolefins and produced through chain-growth polymerization from the propylene monomer. Polypropylene is non-polar, partially crystalline and resistant to chemicals and heat. Compared to high-density polyethylene, polypropylene has a lower density of 0.855 g/cm^3 and a higher melting point of 130–171 °C, which makes it tough but flexible. The applications of polypropylene are found in the manufacturing of bottles, chips bags, biscuit wrappers, drinking straws, heavy-duty bags, plant pots, crates, chairs and desks, office folders, tarpaulin and car bumpers. Polypropylene contributes to the highest proportion of plastics in municipal solid waste, which is nearly 24.3% (Sharuddin et al. 2016). However, it is occasionally recycled.

Polystyrene is identified by the resin identification code 6 and has its chemical formula of $(C_8H_8)_n$ and the IUPAC name of poly(1-phenylethene). As the name suggests, polystyrene is a synthetic aromatic hydrocarbon polymer in solid or foam state produced from a styrene monomer. It has a poor barrier to moisture and gases and has less resistance to fats and solvents. However, the lightness of polystyrene offers benefits such as strength and durability. These properties make this plastic desirable for wide applications in the packaging of food and appliances, electronics and construction. Some notable uses of polystyrene can be found in the manufacturing of flexible plastics, packing peanuts, Styrofoam, plastic cups, disposable cutlery, fast food trays, egg boxes and coat hangers. The large-scale applications of polystyrene result in significant amounts of its residual waste generation in municipal solid waste at the consumer level.

Due to the low-density polystyrene foam, it is a tedious task to separate polystyrene efficiently and economically for recycling. With its profuse production globally, polystyrene wastes generate litter and foam particles, which biodegrade slowly and accumulate in the environment, especially in water bodies and soil. In the soil and water bodies, polystyrene debris undergoes hydrolysis, photodegradation and oxidation. More brittle particles are formed from polystyrene debris because of chemical weathering, traction and saltation (Kwon et al. 2014). These partially degraded microplastics could potentially enter the food chain and contaminate the soil and seawater ecosystems, thereby inhibiting the sustenance of burrowing animals, insects and aquatic flora and fauna.

The resin identification code 7 designates other miscellaneous plastic wastes such as polybutylene terephthalate, polycarbonate, polylactic acid, acrylic, acrylonitrile butadiene styrene, multilayered mixed polymers and nylon. The plastics in this category are difficult to recycle. Polycarbonates are thermoplastic polymers that contain carbonates. Being optically transparent, polycarbonates products are resistant to temperature and physical impact, which makes them desirable for many commodity and engineering plastic-based goods. Examples of polycarbonate products include compact discs, digital versatile discs, Blu-ray discs, baby bottles, electrical components, medical storage containers and components for construction, automotive, aircraft and railways.

Polycarbonates manufactured products also contain bisphenol A as a precursor monomer, which is a xenoestrogen. Xenoestrogens are a kind of xenohormones that exhibit estrogen-mimicking properties and often lead to most common implications to human health such as precocious puberty and reproductive disorders (Nikaido et al. 2004). Recently, the US Food and Drug Administration has amended its regulations not to use bisphenol A-based polycarbonates and epoxy resins in baby bottles, sippy cups and other food packaging products (FDA 2020). Following the regulations, the USA, Canada, the European Union and several other countries have either banned or restricted the use of bisphenol A-based polycarbonates in baby products.

Polybutylene terephthalate is a thermoplastic semicrystalline polymer in the resin identification code 7 category. This type of polyester is durable and resistant to solvents and moderate heat. With the addition of additives, the flammability properties of polybutylene terephthalate can be altered. Polybutylene terephthalate has profound applications in electrical components for household and automotive. Another candidate in the resin identification code 7 group, polylactic acid, is a biodegradable thermoplastic and an aliphatic polyester produced from renewable resources. Polylactic acid is a precursor to produce bioplastics, which are gaining popularity in research and commercial applications because of lower CO_2 footprint and biodegradability (Emadian et al. 2017). On the global scale, revenues generated from bioplastics are expected to exceed the US $4.4 billion by 2026 (Ceresana 2020c).

Acrylonitrile butadiene styrene is an amorphous thermoplastic terpolymer made through the polymerization of acrylonitrile and styrene in the presence of polybutadiene. Acrylonitrile butadiene styrene is lightweight but relatively stronger than pure polystyrene and has found applications in musical instruments, automotive parts, medical devices, electrical components, luggage cases and toys. Lastly, nylon is a thermoplastic silk-type synthetic polymer composed of aliphatic and semi-aromatic polyamides. Nylon is applied commercially in fabrics, apparel, fibers, carpet flooring and automotive furnishing Nylon can be recycled in injection molding machines. The utility of nylon is increasing momentously, which is evident by the steady increase in its market value from the US $20.5 billion in 2013 to the projected US $30 billion by the end of 2020 (Acmite 2014).

As discussed in this section, the classification of plastics is based on the physical and chemical similarities between the grades of polymers. For example, thermoplastics and thermosetting plastics are classified based on their permeance of the form factor upon heat treatment. Amorphous, crystalline and semicrystalline plastics are categorized by the virtue of their molecular framework. However, the ASTM International Resin Identification Coding system establishes a standard classification of plastics, which is followed in most of the countries.

Biological degradation of plastics

The high tensile strength and durability of plastics make them linger in the environment for centuries after their disposal in landfills. The most commonly used plastics in our everyday life such as polyethylene, polypropylene, polystyrene, polyvinyl chloride and polyethylene terephthalate are resilience to natural degradation and accumulate in the soil and water bodies. There are two ways to manage the eco-friendly remediation of plastics, which are: (i) exploring microorganisms effective in degrading polymers, and (ii) synthesizing polymers susceptible to biodegradation. The biodegradation of plastics by microorganisms occurs via the following stages: (i) pretreatment of the plastics for depolymerization, (ii) adherence of the microorganisms to the polymer, (iii) extracellular enzymatic activity for further depolymerization of the polymers to monomeric compounds, (iv) metabolism of the monomers by the microorganisms as the carbon source, (v) degradation of the depolymerized monomers, dimers and oligomers to innocuous components (Fig. 6).

Biodegradation of plastics is a heterogeneous process, which is facilitated by the enzymatic activities of certain microorganisms via chain cleavage of the polymer into monomers and oligomers. Both extracellular and intracellular enzymes, particularly depolymerases are involved in the biodegradation of plastics (Mohan and Srivastava 2010). Some extracellular enzymes such as depolymerases secreted by microorganisms also depolymerize the polymer resulting in the cleavage of oligopolymers to dimers and monomers, which reduces their molar mass to water-soluble intermediates (Alshehrei 2017). This helps for easy transport of the monomers into the microbial cells where they are metabolized.

Depending on the microbial respiration, both aerobic and anaerobic pathways can perform biodegradation of plastics. Aerobic metabolism usually results in CO_2 and water as the end products while anaerobic metabolism produced CO_2, CH_4, H_2S and water. The physicochemical factors such as temperature, pressure, pH, available water, irradiation, stretching and tearing cause mechanical damage to the polymer by increasing its surface area, thereby enhancing the access of biological forces such as enzymes and microbial

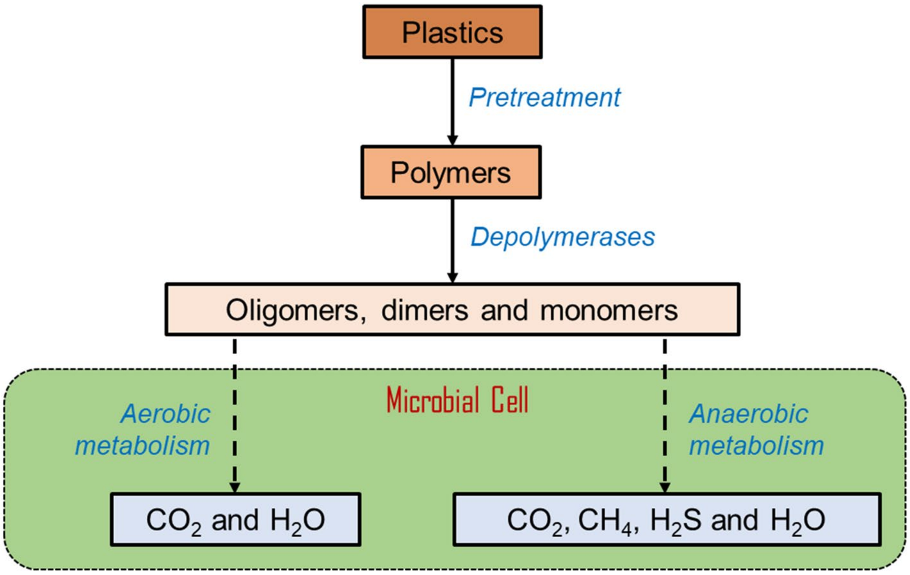

Fig. 6 Typical representation of biodegradation of plastics. This figure represents some basic steps in the microbial biodegradation of plastics, which include pretreatment and depolymerization of plastics; microbial colonization on depolymerized materials; extracellular enzymatic degradation to monomers; as well as microbial metabolic denaturation of monomers, dimers and oligomers to permanent gases and innocuous byproducts

metabolites to catalyze the degradation. Before microbial degradation, a few pretreatment steps are necessary to depolymerize the plastic polymers to enhance the enzymatic activities. Table 2 summarizes the list of some notable microorganisms capable of degrading plastic polymers such as nylon, polybutylene succinate, polyethylene succinate, polycaprolactone, polycarbonate, polyethylene, polyethylene adipate, polyhydroxyalkanoates, polyhydroxybutyrate, polylactic acid, polystyrene, polyurethane, polyvinyl alcohol and poly-β-propiolactone.

Certain microorganisms synthesize surface-active amphiphilic compounds known as biosurfactants, which can enhance the biodegradation of plastics. Biosurfactants are molecules that have both hydrophobic, i.e., polar and hydrophilic, i.e., nonpolar domains. The hydrophilic moieties comprise of peptides, proteins, monosaccharides, disaccharides or polysaccharides whereas the hydrophobic moieties consist of unsaturated and saturated fatty acids or fatty alcohols. A few notable microorganisms that are capable of secreting biosurfactants belong to the genera *Acinetobacter*, *Alcanivorax*, *Arthrobacter*, *Aspergillus*, *Bacillus*, *Capnocytophaga*, *Candida*, *Corynebacterium*, *Gluconobacter*, *Halomonas*, *Kluyveromyces*, *Lactobacillus*, *Mycobacterium*, *Nocardia*, *Penicillium*, *Pseudomonas*, *Rhodococcus*, *Rhodotorula*, *Saccharomyces*, *Serratia*, *Sphingomonas*, *Talaramyces*, *Thiobacillus*, *Tsukamurella*, *Ustilago* and *Yarrowia* (Santos et al. 2016). The amphiphilic molecular structure of biosurfactants results in a higher surface area of the hydrophobic water-insoluble substances, thereby increasing the water bioavailability and alterations to the bacterial cell surface properties (Vimala and Mathew 2016). Apart from its uses such as agriculture, food industries, cosmetics and pharmaceutics, biosurfactants have the potential for the degradation of oil and plastic polymers, e.g., polyolefins, polyethylene and polycarbonates (Ron and Rosenberg 2002; Artham and Doble 2008; Arkatkar et al. 2009; Vimala and Mathew 2016). Biosurfactants usually aid the biodegradation of polymers along with other pretreatment methods such as ultraviolet irradiation, gamma-irradiation, photo-oxidation and other physicochemical agents (Artham and Doble 2008).

As reviewed by a few authors (Mohan and Srivastava 2010; Alshehrei 2017), the biodegradation of plastics is influenced by several factors, a few of which include:

(i) Chemical properties of the polymer, e.g., molecular weight, density and crystallinity;
(ii) Physical properties of the polymer, e.g., pellets, powder, fibers, chips and films;
(iii) Mechanical properties of the polymer, e.g., tensile strength, stiffness, hardness, embrittlement and impact resistance;
(iv) Electrical properties of the polymer, e.g., dielectric strength, insulation resistance and arc resistance;
(v) Optical properties of the polymer, e.g., optical transmission and haze;
(vi) Molecular composition or polymer blend;
(vii) The ratio of hydrophobic and hydrophilic domains;
(viii) The ratio of crystalline and amorphous regions;
(ix) The structural complexity of the polymer, e.g., hydrocarbon chain linearity, coupling and branching;
(x) Type of pretreatment employed for depolymerization of plastics;
(xi) Extracellular enzymes;
(xii) Type of microorganisms used for biodegradation; and
(xiii) Process parameters employed in biological degradation

Table 2 Microorganisms capable of degrading plastic polymers

Polymer	Microorganism
Nylon	*Flavobacterium* sp. *Pseudomonas* sp.
Polybutylene succinate and polyethylene succinate	*Aspergillus clavatus* *Bacillus pumilus* *Excellospora japonica* *Excellospora viridilutea* *Microbispora rosea* *Paenibacillus* sp.
Polycaprolactone	*Aspergillus flavus* *Clostridium* sp. *Penicillium funiculosum* *Rhizopus arrhizus*
Polycarbonate	*Amycolatopsis* sp. *Candida cylindracea* *Chromobacterium viscosum* *Pseudomonas* sp. *Roseateles depolymerans*
Polyethylene	*Brevibacillus borstelensis* *Penicillium simplicissimum* *Phanerochaete chrysosporium* *Rhodococcus ruber* *Streptomyces badius* *Streptomyces setonii* *Streptomyces viridosporus*
Polyethylene adipate	*Achromobacter* sp. *Candida* sp. *Penicillium* sp. *Rhizopus* sp.
Polyhydroxyalkanoates	*Alcaligenes faecalis* *Aspergillus* sp. *Bacillus* sp. *Pseudomonas stutzeri* *Streptomyces* sp.
Polyhydroxybutyrate	*Acidovorax* sp. *Alcaligenes faecalis* *Aspergillus fumigatus* *Comamonas testosterone* *Pseudomonas lemoignei* *Pseudomonas stutzeri* *Streptomyces* sp. *Variovorax paradoxus*
Polylactic acid	*Amycolatopsis* sp. *Bacillus brevis* *Saccharotrix* sp. *Tritirachium album*
Polystyrene	*Arthrobacter* sp. *Escherichia coli* *Micrococcus* sp. *Pseudomonas* sp.
Polyurethane	*Aureobasidium pullulans* *Comamonas acidovorans* *Curvularia senegalensis* *Fusarium solani* *Rhizopus delemar*
Polyvinyl alcohol	*Alcaligenes faecalis* *Pseudomonas putida*
Poly-β-propiolactone	*Acidovorax* sp. *Rhizopus delemar* *Sphingomonas paucimobilis* *Variovorax paradoxus*

References: Shimao (2001), Tokiwa et al. (2009), Sivan (2011), Vimala and Mathew (2016) and Alshehrei (2017)

Advanced eco-friendly technologies have allowed the development of biodegradable plastics or bioplastics from natural products and renewable biomass sources such as agricultural crop residues, forestry biomass, vegetable extracts, food starch and fatty acid wastes. The bioplastics must retain the durability and mechanical properties similar to those of conventional plastics in addition to biodegradability. Bioplastics are made of biodegradable polymers, which when disposed of favorably in the environment, e.g., compost, soil or wastewater are acted upon by microorganisms as the sources of carbon and energy, thereby leading to their degradation (Calabrò and Grosso 2018). The examples of some common bioplastic polymers include starch plastics, cellulose esters, polylactic acid, polybutylene succinate, polyhydroxyalkanoate, bio-polyethylene, bio-polyethylene terephthalate and bio-polyamide (Emadian et al. 2017). The blends of biodegradable polymers may generally be regarded as safe but they might also release certain toxic byproducts during biodegradation under unfavorable composting conditions. Therefore, short-term and long-term environmental performance and economic indicators such as eco-efficiency assessment and end-of-life options in the bioplastic manufacturing system should be assessed to determine their sustainability (Morone et al. 2015; Changwichan et al. 2018).

The research on bioplastic is gaining momentum, which has a tendency to partially, if not fully, replace the use of conventional plastics to enhance their biodegradability in landfills and composting pits under natural conditions. Nevertheless, biodegradation of plastics is highly crucial for polar and non-polar polymers, monomers and oligomers, which cannot be recycled, while eventually biomagnifying in various ecosystems, food chains and food webs. Therefore, microbial biodegradation of plastics seems promising in this aspect. However, the interactions between the microorganisms and the natural or synthetic polymers largely determine the biodegradation of waste plastics.

Thermochemical processing of plastic wastes

Pyrolysis

Pyrolysis is a thermochemical biomass-to-liquid conversion technology, which operates in the absence of oxygen to produce liquid hydrocarbon fuels, i.e., bio-oil, char and producer gas. The product distribution from pyrolysis greatly depends on the reaction temperature, heating rate, residence time, reactor type (Nanda et al. 2013; Azargohar et al. 2013). The physicochemical properties of the feedstock such as moisture content, elemental composition and particle size also determine the yields of bio-oil, char and gases. Many reactor configurations such as ablative reactor, fixed bed, transported bed, vacuum moving bed, circulating fluidized bed, mechanically fluidized bed and rotating cone have been investigated for pyrolysis of biomass and other organic matter. Nevertheless, fluidized bed reactors are the most commonly used due to enhanced heat and mass transfer leading to higher thermal cracking and greater bio-oil yields.

Depending on the heating rate and vapor residence time, pyrolysis can be classified into slow, fast, flash and intermediate pyrolysis. High temperatures, fast heating rates and short vapor residence times are characteristic features for fast and flash pyrolysis. These attributes result in greater yields of bio-oil than char and permanent gases. Contrariwise, moderate temperatures, slow heating rates and longer vapor residence times are distinguishing properties of slow pyrolysis. These features improve the yields of char than bio-oil. A temperature range of 400–500 °C and a higher heating rate of 10–200 °C/s, as well as a vapor residence time of 30–1500 s typically characterize fast pyrolysis (Bridgwater 1999). On the other hand, slow pyrolysis is operated at a wider temperature range of 300–700 °C with a slower heating rate of 0.1–1 °C/s and a vapor residence time of 10–100 min (Bridgwater and Peacocke 2000). Flash pyrolysis prefers higher temperatures between 800 and 1000 °C with swift heating rates greater than 1000 °C/s and vapor residence time around 0.5 s (Maschio et al. 1992). Nevertheless, intermediate pyrolysis is performed at temperatures of 500–600 °C, a heating rate of 2–10 °C/s and moderate vapor residence time of 10–20 s (Ahmad et al. 2014).

The product distribution from pyrolysis is dependent on several process parameters as mentioned earlier, but the most important of all are temperature, heating rate and vapor residence time. Nanda et al. (2016) comprehensively reviewed several studies that have evaluated the variation in the quantity and quality of bio-oils concerning temperature and heating rate. It was reported that the average liquid product yield or bio-crude from pyrolysis increased with temperature as follows: 300–400 °C, 30 wt% < 400–500 °C, 42 wt% < 500–600 °C, 47 wt%. On the contrary, char yield decreased with rising temperature as: 200–300 °C, 77 wt% > 300–400 °C, 49 wt% > 400–500 °C, 34 wt% > 500–600 °C, 27 wt%.

During pyrolysis, high temperature initially leads to dehydration, depolymerization and fragmentation of biomass and other organics to produce volatile components. Upon condensation, the volatile components quench to produce bio-crude. It is the quenching process and residence time of the volatile vapors that determine bio-oil quality. Rapid quenching sequesters many degraded volatile compounds that can condense, cleave or react with other intermediate components at longer vapor residence time (Park et al. 2016). Gases such as H_2, CO_2, CO and CH_4 tend to be non-condensable and exit in the producer gas (Kanaujia et al. 2014). The char production is suppressed at high temperatures because of greater cracking reactions. The longer vapor residence time in the reactor causes a reduction in the condensation rates of volatile components, which favors secondary reactions in pyrolysis. The secondary reactions result in the generation of high molecular weight compounds such as tars and char. Therefore, char and tar are the products of secondary polymerization reactions.

Bio-crude, obtained as the liquid product of pyrolysis, is composed of organic and aqueous phases. The aqueous phase consists of water-soluble components such as acetic acids, carboxylic acids, alcohols, aldehydes, ethers, esters and ketones (Cardoso et al. 2016). On the other hand, the organic phase consists of carbonyls, phenolics, tar and heavy oil. The origin and composition of a feedstock, e.g., cellulose, hemicellulose, lignin, fats, lipids, mineral matter and moisture content have a strong influence on the chemical compositions of bio-oil, char and producer gas.

Unlike biomass-derived oils, oil generated from thermal cracking of plastics typically contains heavy oil, light oil, mid-distillates, naphtha and gases (Angyal et al. 2007). The naphtha obtained from polystyrene mostly contains styrene, benzene, toluene, ethylbenzene, xylene, izoproylbenzene, α-methylstyrene and other hydrocarbons. The naphtha-like fractions contain C_5–C_{15} hydrocarbons including paraffins, olefins and aromatics. The mid-distillates contain C_{12}–C_{28} hydrocarbons. The light oil consists of olefins and paraffins and has a boiling point in the range of 250–350 °C. Conversely, heavy oils have boiling points typically greater than 350 °C and contain wax-like materials such as paraffins, olefins, aromatics and high molecular weight components. The gas-phase comprises of butane (C_4H_{10}), butene (C_4H_8), propane (C_3H_8), propene (C_3H_6), ethane (C_2H_6), ethene (C_2H_4) and methane (CH_4) along with smaller amounts of hydrogen (H_2), carbon dioxide (CO_2) and carbon monoxide (CO) (Adrados et al. 2012). As reported by Toraman et al. (2014), the oils produced from pyrolysis of waste plastics contain numerous compounds that can be grouped under the following categories: (i) hydrocarbons, e.g., *n*-paraffins, iso-paraffins, olefins, naphthenes, mono-aromatics, di-aromatics, tri-aromatics, tetra-aromatics, naphthenoaromatics, naphthenodiaromatics and naphthenotriaromatics; (ii) nitrogenous compounds, e.g., nitriles, pyridines, quinolines, indole and caprolactam; (iii) sulfurous compounds, e.g., thiols, thiophenes, benzothiophenes and dibenzothiophenes; and (iv) oxygenated compounds, e.g., aldehydes, ketones, phenols, esters and ethers.

Pyrolysis oil requires upgrading to be used either as a fuel for vehicle engines and power plants or as a feedstock to synthesize value-added fine chemicals. Bio-oil can be upgraded through direct and indirect pathways. Since bio-oil is highly oxygenated, the oxygen content results in their lower calorific value and energy density, as well as lagging thermal stability. An upgrading technique selectively removes oxygen from the bio-oil is a contributing factor in its low stability and low heating value. The direct routes of bio-oil upgrading consist of zeolite cracking, catalytic hydrodeoxygenation and emulsification with diesel. Hydrodeoxygenation involves many sub-reactions such as decarboxylation, decarbonylation, dealkoxylation, dealkylation, hydrocracking, hydrogenation, hydrogenolysis and methyl transfer (Ruddy et al. 2014). The carbon loss from the bio-oil because of upgrading can be minimized based on the catalyst selectivity. Conversely, the indirect upgrading route involves liquid-to-gas and gas-to-liquid technologies. The bio-oil initially undergoes steam reforming or gasification to produce syngas, which is further transformed into long-chain hydrocarbons and higher alcohols such as green diesel, ethanol, butanol and methanol through the Fischer–Tropsch synthesis (Nanda et al. 2014).

Pyrolysis also results in char as the solid products, which is normally made of stable aromatic carbon. Sub-reactions during pyrolysis and gasification such as dehydration, deamination, decarboxylation and dehydrogenation of organics lead to aromatic char formation (Azargohar et al. 2014, 2019). The quality and applications of char are determined through its physicochemical attributes such as carbon content, degree of carbon crystallinity and amorphicity, elemental composition, mineral matter, specific surface area, porosity, pH, electrical conductivity and ion-exchange capacity (Nanda et al. 2018a). The original feedstock properties and pyrolysis conditions largely influence the physicochemical and structural features of char. Nanda et al. (2016) comprehensively reviewed the multifarious applications of char especially in the following sectors: (i) energy recovery as well as combined heat and power; (ii) fuel and chemical refineries, e.g., catalyst, catalyst support and fluidizing agent; (iii) manufacturing of specialty materials, e.g., activated carbon, carbon nanotubes, carbon nanohorns, carbon fibers, adsorbents and electrodes; (iv) agronomy, e.g., soil fertility, water retention and crop productivity; (v) environmental remediation, e.g., pollutant adsorption and carbon sequestration; and (vi) pharmaceuticals, e.g., toxin adsorption, fillers, additives and cosmetics.

Kim et al. (2002) reported catalytic thermal degradation of mixed plastics containing polypropylene and polystyrene in a semi-batch reactor at 400 °C with a heating rate of 30 °C/min for 2 h. The catalysts investigated for thermal degradation of plastics were silica-alumina, i.e., SiO_2/Al_2O_3 and proton-exchanged zeolites like protonated Zeolite Socony Mobil–5, i.e., HZSM-5 and a protonated natural zeolite, i.e., HNZ. The main degradation product of mixed plastics was oil containing gasoline-like hydrocarbons such as n-paraffins, i-paraffins, olefins, naphthenes and aromatics. The oil yields from polystyrene degradation varied as: SiO_2/Al_2O_3, 83.5 wt% > no catalyst, 81.7 wt% > HNZ, 81.5 wt% > HZSM-5, 75.1 wt%. Likewise, the oil yield from polypropylene degradation varied as: no catalyst, 75.3 wt% > SiO_2/Al_2O_3, 75 wt% > HNZ, 74.3 wt% > HZSM-5, 33.3 wt%. The gas yields from polypropylene were significantly improved with catalysts application such as: no catalyst, 13.7 wt% < SiO_2/Al_2O_3, 24.1 wt% < HNZ, 24.9 wt% < HZSM-5, 66 wt%. The rise in reaction temperature up to 450 °C increased the yields of styrene, methylstyrene, toluene, benzene, trimethylbenzene, xylene, indane, indene, C_{10} and C_{11}–C_{13} hydrocarbons while decreasing the yields of ethylbenzene and propylbenzene.

Williams and Slaney (2007) performed pyrolysis under N_2 atmosphere at 18 MPa and liquefaction under H_2 atmosphere of high-density polyethylene, polypropylene, polystyrene, polyethylene terephthalate and polyvinyl chloride at 500 °C with a heating rate of 5 °C/min for 1 h. Polyethylene terephthalate gave high yields of gases, i.e., 32 wt% both from pyrolysis and liquefaction, whereas polyvinyl chloride gave maximum gases, i.e., 38 wt% from liquefaction, although it produced only 2 wt% oil. The hydrocarbon composition in the pyrolysis gas of plastics included mostly alkane gases such as CH_4, C_2H_6, C_3H_8 and C_4H_{10} with lower concentrations of the alkene gases such as C_2H_4, C_3H_6 and C_4H_8 (Williams and Slaney 2007; Singh and Ruj 2016). Polyethylene, polypropylene and polystyrene gave greater oil yields of 93–95 wt%, 95 wt% and 71–77 wt%, respectively during pyrolysis and liquefaction (Williams and Slaney 2007). In contrast, polyethylene terephthalate and polyvinyl chloride revealed lower oil yields of 15–27 wt% and 2 wt%, respectively. However, pyrolysis and liquefaction of mixed plastics gave maximum oil yields with high concentrations of aromatic compounds than those obtained from individual plastic polymer. This suggests synergistic interactions when mixed plastics are processed together than separate.

Sophonrat et al. (2017) described the interactions between cellulose and various plastics, e.g., polypropylene, polyethylene and polyethylene terephthalate through their co-pyrolysis at 450–600 °C. The derived oil products from co-pyrolysis of cellulose and plastics exhibited greater yields of single and multi-ring aromatic hydrocarbons, aldehydes, alkylated benzene and styrene oligomers with reduced yields of lighter hydrocarbons such as aliphatics, alkanes, dialkenes, furans and pyrans. The reduced yields of lighter products, especially C_3–C_4 hydrocarbons and aliphatics was due to the interactions of the degradation derivatives of polypropylene with aromatic products from polystyrene to form alkylated benzene. Interestingly, the main degradation products from cellulose pyrolysis such as the volatile components were not affected by the co-pyrolysis with plastics. Therefore, the interactions between the degradation products of mixed plastics are more pronounced than the interactions between plastics and cellulose.

Wang et al. (2018) also studied the thermal behavior and reaction kinetics during transition metal-catalyzed co-pyrolysis of cellulose and polyethylene. Transition metals such as nickel, cobalt, iron and manganese promoted the thermal decomposition of cellulose and polyethylene by reducing the initial devolatilization temperatures by 91–136 °C and 8–15 °C, respectively. Furthermore, the transition metal catalysts also reduce the activation energy of cellulose and polyethylene in the mixture by 53.21–80.53 kJ/mol and 8.23–12.36 kJ/mol, respectively.

In a similar study, Miandad et al. (2017) studied the effects of different waste plastics, e.g., polystyrene, polyethylene, polypropylene and polyethylene terephthalate on the pyrolysis oil yield. The pyrolysis experiments were performed at 450 °C with 10 °C/min of heating rate for 75 min. Polystyrene produced maximum oil yields of 80.8 wt% along with the lowest yields of gases, i.e., 13 wt% and char, i.e., 6.2 wt%. The relatively simpler and less branched chemical structure of polystyrene resulted in its maximum degradation in comparison to other plastics. In contrast, polyethylene, when pyrolyzed separately, generated waxes instead of oil due to its long carbon chain structure. However, polyethylene produced 49 wt% oil upon co-pyrolysis with polystyrene and polypropylene. Polyethylene resulted in 40 wt% oil upon co-pyrolysis with polystyrene, polypropylene and polyethylene terephthalate. The differences in the oil yields were due to the production of free radicals from polystyrene that had distinctive synergistic effects. Moreover, the oils derived from all the plastics consisted of mostly aromatics with some alkanes and alkenes. Thermal cracking of polystyrene usually results in styrene monomer and dimer, cumene, α-methylstyrene, ethylbenzene and toluene at higher levels followed by smaller extents of benzene, xylenes and other aromatics (Angyal et al. 2007; Jung et al. 2013). The composition of the pyrolysis oil obtained from acrylonitrile–butadiene–styrene is similar to that of polystyrene, except that the former does not contain nitrogenous compounds (Jung et al. 2013).

Williams and Williams (1999) established the individual and combined effects of waste plastics during pyrolysis in an earlier study. The pyrolysis of high-density polyethylene, low-density polyethylene, polypropylene, polystyrene, polyvinyl chloride and polyethylene terephthalate was performed in a fixed-bed reactor at 700 °C with 25 °C/min of heating rate for 1 h. The gas products from pyrolysis predominantly comprised of H_2, CH_4, C_2H_4, C_2H_6, C_3H_6, C_3H_8, C_4H_8 and C_4H_{10}. While only polyethylene terephthalate produced CO and CO_2, polyvinyl chloride exclusively generated hydrochloric acid. The pyrolysis liquid from high-density polyethylene, low-density polyethylene, polypropylene mostly contained aliphatics, alkanes, alkenes and alkadienes, whereas polystyrene, polyvinyl chloride and polyethylene terephthalate produced aromatic hydrocarbons. The yield of oil and waxes from the plastics was in the order of: polyvinyl chloride, 31.7 wt% < polyethylene terephthalate, 41.3 wt% < high-density polyethylene, 79.7 wt% < low-density polyethylene, 84.3 wt% < polypropylene, 84.4 wt% < polystyrene, 83.8 wt%. In contrast, the gas yields showed the following trend: polyvinyl chloride, 2.5 wt% < polystyrene, 3.4 wt% < low-density polyethylene, 15.1 wt% < polypropylene, 15.3 wt% < high-density polyethylene, 18 wt% < polyethylene terephthalate, 38.7 wt%. Elevated gas yields of 13.8–33.8 wt% and reduced oil or wax yields of 49.5–83.6 wt% were obtained upon the blending of polystyrene at 1:1 ratio with all the plastics. Nearly 24 wt% of hydrochloric acid was produced only from the pyrolysis of polyvinyl chloride with polystyrene. The concentration of aromatic hydrocarbons and polycyclic aromatic hydrocarbons in the oils reduced in co-pyrolysis of plastic mixtures than that of separate pyrolysis of individual plastics.

Demirbas (2004) performed non-catalytic pyrolysis of municipal plastic wastes, e.g., polystyrene, polyethylene and polypropylene to produce gasoline-range hydrocarbons. Because of heating from 25 to 750 °C, the pyrolysis of plastic wastes gave liquid products rich in higher boiling point hydrocarbons. The pyrolysis of polystyrene gave higher liquid yields, whereas and polyethylene and polypropylene resulted in higher gas yields. The pyrolytic oil appeared to be an excellent resource for the recovery of industrially relevant chemicals such as benzene, toluene, styrene and condensable aromatic hydrocarbons. Sajdak and Muzyka (2014) also reported that with 30% polypropylene addition to alder wood and pinewood during co-pyrolysis decreased the yields of liquid products by 1.5–2.5%, consequently increasing the char and gas yields. The rise in biomass degassing due to polypropylene blending also affected the chemical properties of liquids, char and gases (Sajdak et al. 2015).

Chattopadhyay et al. (2016) demonstrated co-pyrolysis of waste paper and waste plastics, e.g., high-density polyethylene, polypropylene and polyethylene terephthalate in a fixed-bed reactor at 800 °C at a heating rate of 10 °C/min in the presence of cobalt/alumina, Co/Al_2O_3; cobalt/ceria, Co/CeO_2 and cobalt/ceria-alumina, Co/CeO_2-Al_2O_3 catalysts. The biomass/plastic mixture resulted in H_2 yields as high as 47 wt%, while the bio-oil contained greater levels of aromatics, e.g., benzene, ethylbenzene, toluene, xylene, naphthalene, methyl naphthalene and multi-methyl naphthalene as well as olefins, e.g., ethylene, propylene and butene.

Ephraim et al. (2018) studied the influence of the type of plastic on product yields and quality from co-pyrolysis of poplar wood with non-polyolefins plastics, e.g., polystyrene and polyvinyl chloride using a fixed-bed reactor at 750 °C with a heating rate of 20 °C/min. Maximum oil yields of 64.2 wt% were obtained from the co-pyrolysis of 30 wt% polyvinyl chloride with 70 wt% poplar wood. Conversely, greater oil yields of 78.6 wt% were obtained from the co-pyrolysis of 50 wt% polystyrene with 50 wt% poplar wood. Co-pyrolysis with polystyrene had positive synergies leading to the formation of H_2, CH_4, CO and CO_2 with lower impacts on C_xH_y compounds. However, co-pyrolysis with polyvinyl chloride resulted in the dissolution of hydrochloric acid in the aqueous fraction of the liquid product instead of chlorinated organic compounds. Typically, pyrolysis of mixed plastics containing polyvinyl chloride can retain inorganic and organic chlorine compounds in the oil and char. The presence of halogens such as chlorine more than 10 ppm in the oil limits its use as a fuel or chemical precursor (Bhaskar et al. 2005).

Sanahuja-Parejo et al. (2018) reported the co-pyrolysis of grape seeds and waste tires to produce drop-in fuels. The pyrolysis was performed in a fixed-bed reactor using calcined calcite at 550 °C with 100 °C/min for 30 min. With 40 wt% of waste tires in the grape seed feedstock, the organic phase yield in the pyrolysis oil increased up to 73 wt%, which was significantly greater than those obtained from the pyrolysis of individual feedstock, i.e., 61 wt% organic phase. Moreover, abridged oxygen content of 4.2 wt% and pH value or total acid number of 1 mg KOH/g for the oil reduced the chances of furnace corrosivity and instability. Co-pyrolysis of grape seeds and waste tires at a ratio of 60:40 wt% with calcined calcite catalyst resulted in the oil with a higher calorific value of 35.4 MJ/kg. In contrast, the oil produced from non-catalytic conventional pyrolysis of grape seed demonstrated a heating value of 15.3 MJ/kg. The oil had high levels of aromatic and cyclic hydrocarbons with lower phenolics and sulfur compounds. Calcined calcite promoted dehydration of acids and phenols to generate hydrocarbons while the radical interactions between the waste tire and grape seeds synergistically improved oil yields.

In a more recent study, Shah et al. (2019) pyrolyzed waste tire and cotton stalks in different blend ratios in a fixed-bed reactor at 550 °C with a heating rate of 20 °C/min. The investigated blend ratios for cotton stalks-to-waste tire were 1:0, 4:1, 3:2, 2:3 and 0:1. The blend ratio of 2:3 showed maximum oil yield of 48 wt% with 78% organic phase having high calorific value of 41 MJ/kg and significant levels of alkanes. Other components identified in the oils derived from co-pyrolysis of the cotton stalk and waste tire were benzene derivatives, phenols, terpenes, olefins, alkanes, aldehydes and ketones, polycyclic aromatic hydrocarbons, acids and acid derivatives. The group of researchers also performed another study on co-pyrolysis of sugarcane bagasse with scrap tires at a 1:3 blend ratio in a fixed-bed reactor at 500 °C with a heating rate of 20 °C/min (Ahmed et al. 2018). Co-pyrolysis of sugarcane bagasse and scrap tire produced 49 wt% of liquid products compared to 42.1 wt% obtained from sugarcane bagasse upon its separate pyrolysis. Through many synergistic effects scrap tires enhanced the quality of co-pyrolysis liquid product such as its carbon content of 85.3 wt%, calorific value of 41 MJ/kg, viscosity of 3.63 cP, specific gravity of 0.916, flash point less than 30 °C and pH of 3.48. In contrast, the bio-oil produced from pyrolysis of pure sugarcane bagasse had the following properties: carbon content of 52.8 wt%, calorific value of 19.1 MJ/kg, viscosity of 7.36 cP, specific gravity of 1.048, flash point less than 30 °C and pH of 3.02. The oil produced from co-pyrolysis also comprised of benzene derivatives, phenols, terpenes, olefins, alkanes, aldehydes and ketones, polycyclic aromatic hydrocarbons, acids and acid derivatives.

Lu et al. (2018) elucidated the synergistic effects of co-pyrolysis of pinewood, polyethylene and polyvinyl chloride on the resulting char and oil. The co-pyrolysis was performed in a fixed-bed reactor at 600 °C for 15 min. Compared to the theoretical results, the blending of pinewood and polyethylene had many synergistic effects on the co-pyrolysis such as: (i) increased weight loss rate of pinewood by 8–23.7%, (ii) lower char yields of 13.8–22.4%, (iii) higher oil yields of 3.7–4.4%. Likewise, the blending of pinewood with polyvinyl chloride had the following effects: (i) lowering of temperature by 47–51 °C leading to a maximum weight loss of pinewood, (ii) higher char yields of 15.5–27.9%, and (iii) lower oil yields of 7.2–14.4%. The interactions between biomass and plastics decreased the atomic H/C ratio of char, resulting in higher chemical stability and aromaticity. The surface area of char produced from pinewood blended with polyethylene and polyvinyl chloride was relatively higher, i.e., 192–201 m^2/g than that of pinewood char, i.e., 185 m^2/g. The oil produced from wood and plastics mixture had better fuel properties including higher atomic hydrogen/carbon ratio and lower atomic oxygen/carbon ratio compared to the theoretical results. When pyrolyzed separately, pinewood-derived oil contained oxygenated compounds such as phenols, polyethylene-derived oil revealed waxes, e.g., dienes, alkenes and alkanes and polyvinyl chloride-derived oil showed the presence of polycyclic aromatic hydrocarbons. The oil produced from pinewood and polyethylene mainly contained waxes, e.g., dienes, alkenes and alkanes of C_{10}–C_{24} hydrocarbons.

As mentioned earlier, the manufacturing of plastics requires petroleum and other fossil fuels. Therefore, the thermochemical conversion of waste plastics to liquid oil through pyrolysis has great potential. The pyrolysis oil generated from plastics not only has high calorific value but also other fuel chemistry comparable with the commercial fossil fuels. Many environmental benefits such as reduced carbon footprints and greenhouse gas emissions can be realized from the co-pyrolysis of plastics with lignocellulosic biomass and other organic residues.

Liquefaction

Liquefaction is another thermochemical biomass-to-liquid technology that transforms waste organics at high pressures usually in the presence of catalysts to energy-dense bio-oil.

Liquefaction typically requires temperatures of 250–450 °C and pressure in the range of 1–20 MPa (Huber et al. 2006). The employment of alkali catalysts such as sodium carbonate, potassium carbonate, sodium hydroxide and potassium hydroxide in liquefaction can enhance bio-oil yields and subdue char generation (Zhong and Wei 2004). On the other hand, acidic catalysts such as phosphoric acid, sulfuric acid and *p*-toluenesulfonic acid can lower liquefaction temperature and the overall reaction time (Mun and Hassan 2004). Apart from bio-oil, liquefaction is also employed to produce industrially relevant products such as epoxy resins, adhesives and polyurethane foams (Zhang et al. 2010).

Liquefaction leads to the thermal decomposition of sugars and other organics into their monomeric units. The resulting monomers re-polymerize to produce oils and some small amount of char through condensation. However, certain solvents can diminish the higher-order solid-state reactions and detrimental condensation reactions that otherwise lead to char formation (Yadav et al. 2019). Supercritical ethanol is an attractive solvent used in the liquefaction of biomass. Supercritical alcohols are beneficial in improving the solubility and cracking of organic components, which can result in better hydrogen-donating properties, higher bio-oil yield and superior phase separation because of the alcohol's low boiling point (Brand et al. 2014). The process is termed solvolysis as it uses solvents, e.g., alcohols, phenols, creosote oil and ethylene glycol at high pressures to increase the solubility of biomass in hydrothermal conditions during liquefaction (Huber et al. 2006).

Liquefaction can also be performed at high-pressure aqueous conditions. Water acts as the aqueous reaction medium in hydrothermal liquefaction, which is performed at 280–370 °C and 10–25 MPa pressure (Toor et al. 2011). The critical temperature (T_c), i.e., 374 °C and critical pressure (P_c), i.e., 22.1 MPa of water determines its application as a green solvent in hydrothermal conversion processes owing to its better solvation properties, low cost, non-toxicity and abundancy. Subcritical water and supercritical water are the two fluid states of water that have gained increased interest in hydrothermal liquefaction and gasification processes (Nanda et al. 2017a, b; Okolie et al. 2019). Supercritical water occurs at a temperature and pressure beyond the water's critical points, whereas subcritical water occurs near or below water's critical points. Hydrothermal liquefaction causes hydrolysis of biomass into low molecular weight compounds. The fragmented molecules tend to be unstable and reactive, which causes them to re-polymerize into high molecular weight compounds (Toor et al. 2011). During this process, oxygen is removed from biomass through dehydration, deoxygenation and decarboxylation.

Pei et al. (2012) conducted co-liquefaction of microalgae *Spirulina* with a synthetic polymer, i.e., high-density polyethylene in subcritical and supercritical ethanol. The bio-oils generated through the liquefaction of microalgae alone are comprised of oxygenated compounds such as carboxylic acids, esters and ketones. However, co-liquefaction of microalgae and high-density polyethylene produced bio-oil rich in aliphatic hydrocarbons. Besides, it was also reported that the extraction activity between the thermally decomposed components and subcritical or supercritical ethanol could be enhanced with increasing pressures. The elevating pressure could also check the condensation, cyclization and re-polymerization of the intermediates. High pressures during liquefaction can prevent the gasification of liquid products, thereby increasing their recovery or yield (Behrendt et al. 2008).

As one of the early studies, Feng et al. (1996) performed catalytic liquefaction of plastics directly and along with coal. The catalysts used for liquefaction included the protonated Zeolite Socony Mobil–5 catalyst, i.e., HZSM-5, ferrihydrite treated with citric acid, i.e., FHYD/CA, co-precipitated alumina-silica, i.e., Al_2O_3-SiO_2 and ternary ferrihydrite-Al_2O_3-SiO_2. The catalytic liquefaction studies were conducted on high-density polyethylene, polypropylene and coal-plastic mixtures in a fluidized sand bath reactor at variable temperatures of 420–460 °C, gas pressures of 0.7–5.5 MPa, reaction times of 20–60 min and solvents concentrations of 0–15 g. While the effect of pressure was found to be minimal on the oil yields from the plastics, yet the organic fraction was rich in gasoline, kerosene and heavy oil. The lighter hydrocarbons in the oil improved with increasing H_2 gas pressure. The use of waste oil and tetralin mixtures as solvents increased the yield of oil from plastics at 445 °C. The co-liquefaction of coil and plastics with HZSM-5 catalyst produced 75 wt% oil and gas at 430 °C, which was higher compared to the yield at 450 °C, i.e., 66 wt%. The results indicated that HZSM-5 had higher catalytic activity than alumina-silica catalysts.

Luo and Curtis (1996) studied the effect of reaction parameters and catalyst type on liquefaction of waste plastics mixture, i.e., 50% high-density polyethylene, 30% polyethylene terephthalate and 20% polystyrene directly as well as with coal at 400–440 °C with H_2 gas pressure of 5.6 MPa for 30–120 min with HZSM-5 and fluid catalytic cracking catalysts. Oils produced from individual plastics exhibited greater levels of hexane-soluble compounds, whereas the oil produced from individual plastic polymers had more tetrahydrofuran-soluble components. The liquefaction efficiency of plastics increased at low H_2 pressures and with the addition of aromatic, hydroaromatic, cyclohexane and straight-chain aliphatic solvents. Nevertheless, catalytic co-liquefaction of plastics and coal had better hydrocarbon conversion efficiency even without the addition of a solvent. During co-liquefaction, the addition of tetralin as a hydroaromatic solvent accelerated coal conversion but retarded plastic conversion. The presence of coal suppressed the reactivity of plastics during co-liquefaction compared to when both the liquefied separately.

Zmierczak et al. (1996) performed depolymerization-liquefaction of plastics and rubbers, particularly polystyrenes and styrene–butadiene copolymers in a microclave reactor at variable temperatures of 350–450 °C, H_2 pressures of 3.4–17.2 MPa and reaction times of 15–120 min in the presence of solid superacid catalysts Gasoline-type liquids were obtained from the liquefaction of plastics and rubbers. Liquid hydrocarbon products from liquefaction of polystyrene contained benzene, ethylbenzene, toluene, alkylbenzene, diphenylethanes, diphenylpropanes, diphenylbutanes, diphenylpentanes, diphenylhexanes, triphenylalkanes, terphenyls, quaterphenyls, indanes, naphthalenes and other hydrocarbons. With the increase in liquefaction temperature from 350 to 450 °C, the yields of benzene, ethylbenzene, toluene, C_3-alkylbenzene and terphenyls improved. Liquefaction of non-vulcanized styrene–butadiene co-polymer generated paraffins, cycloparaffins, benzene, ethylbenzene, toluene, styrene, alkylbenzene, alkyl and cycloalkylbenzenes, diphenylalkanes and other hydrocarbons. Higher liquid yield from the liquefaction of styrene–butadiene copolymer was obtained at 375 °C, i.e., 95.5 wt% compared to 450 °C, i.e., 85.7 wt%. Similarly, shorter reaction times of 15 min resulted in higher liquid yields of 99.2 wt% than that obtained at longer reaction times of 120 min, i.e., 95 wt%.

In a continuation study, Shabtai et al. (1997) reported depolymerization-liquefaction of high-density polyethylene, polypropylene and polybutadiene in a microclave reactor at variable temperatures of 350–450 °C, H_2 pressures of 3.4–13.8 MPa and reaction times of 30–180 min in the presence of solid superacid catalysts. Catalytic liquefaction of high-density polyethylene at 350 °C produced liquids rich in normal and branched paraffin and lower levels of cycloparaffins and olefins. The liquid products from polypropylene contained branched gasoline-like compounds of branched paraffin. The liquid product from polybutadiene at 415 °C comprised of paraffin, alkyl-substituted naphthenes, benzene, alkylbenzenes, tetralins, indanes and bicyclic arenes.

Wang et al. (2014) studied different blending ratios of lignite, wheat straw and plastic waste during hydrothermal co-liquefaction in subcritical water at 260–320 °C for 30 min. A blending ratio of 5:4:1 for lignite, wheat straw and plastic waste had synergistic effects on oil and gas yields. The oil yields were found to increase at higher subcritical temperatures of 280–320 °C. Tourmaline supplemented the yield and quality of oil and improved the feedstock conversion during co-liquefaction. Several reactions that occurred during co-liquefaction of lignite, wheat straw and plastic wastes included dehydration, dehydrogenation, deoxygenation, decarboxylation followed by condensation, cyclization, polymerization under inert atmosphere and hydrogenation in the presence of H_2. During co-liquefaction, pre-asphaltene and asphaltene are formed, which further decompose to produce oil at high reaction temperatures.

Wu et al. (2017) performed co-liquefaction of microalgae *Dunaliella tertiolecta* and polypropylene plastic in subcritical and supercritical water. The addition of polypropylene in co-liquefaction with microalgae influences the composition of bio-oil, especially by reducing its acid content. The bio-oil from the liquefaction of microalgae alone had an acid content of 18.7%, whereas co-liquefaction of microalgae and polypropylene at a ratio of 8:2 reduced the acid content of bio-oil below the detection limit of gas chromatography. Moreover, polypropylene also affected the transformation of carbohydrates in microalgae to produce more cyclopentenone derivatives and favored the Maillard reaction between carbohydrates and proteins.

In summary, liquefaction of plastics generates an oil product with less aqueous content compared to the highly oxygenated bio-oil produced from pyrolysis of lignocellulosic biomass and other biogenic residues. Moreover, low oxygen levels in the oils derived from the liquefaction of plastics also contribute to their heating value and fuel properties. Owing to the low moisture content in plastic-derived oils, the intensity of catalytic fuel upgrading technologies such as hydroprocessing, hydrotreating and hydrodeoxygenation are usually much lesser.

Gasification

Gasification is a thermochemical biomass-to-gas technology that converts organics to synthesis gas. Synthesis gas or syngas is predominantly a mixture of H_2 and CO along with CO_2, CH_4 and trace amounts of acetylene (C_2H_2), ethylene (C_2H_4) and ethane (C_2H_6). While gasification of coal is a mature technology to produce syngas at high temperatures, biomass gasification is relatively newer that can operate at lower temperatures because of its greater reactivity (Huber et al. 2006). Compared to other thermochemical technologies, gasification is considered attractive because it generates H_2 that can reduce exergy loss during combustion in power plants (Fushimi et al. 2003). Moreover, H_2 is a clean energy carrier because of its higher heating value of 141.7 MJ/kg and the lowest exergy rate among most hydrocarbon fuels.

Gasification can be performed in air, steam or aqueous media. While conventional gasification is operated in the presence of air or steam, hydrothermal gasification involves subcritical or supercritical water as the reaction media. The thermophysical properties of supercritical water provide optimal conditions for the oxidation of organic wastes. Supercritical water has viscosities like gases and densities like liquids rendering better mass transfer and solvation properties. Water acts as a solvent with weak polarity near its critical points, which opens numerous opportunities for hydrolysis of recalcitrant organic compounds in a single homogeneous phase (Reddy et al. 2014; Nanda et al. 2019a). Superheated compressed water is known to possess weak intermolecular and intra-molecular hydrogen bonding, which makes itself a partial source of H_2 during gasification (Correa and Kruse 2018).

Conventional gasification involves a series of thermochemical reactions, especially partial oxidation, pyrolysis and steam gasification. Pyrolysis operates in the absence of oxygen leading to the thermal cracking of biomass to generate bio-oil, producer gas and biochar. On the contrary, partial oxidation employs oxygen below its stoichiometric amount of combustion. Furthermore, steam reforming involves the reforming of organics in the water to produce CO, CO_2 and H_2. On the other hand, hydrothermal gasification typically involves several sub-reactions such as hydrolysis, steam reforming, hydrogenation, methanation and water–gas shift reaction (Okolie et al. 2020a). The water–gas shift reaction is weakly exothermic that is characterized by the reaction of CO with water to liberate H_2 and CO_2. In methanation and hydrogenation reactions, CO reacts with H_2 to produce CH_4 and water. Hydrogenation aids in the cleavage of long-chain polymeric hydrocarbons to lighter compounds through the free radical mechanism.

Near-critical or subcritical water gasification involves three main reaction mechanisms, which are: (i) de-polymerization of the macromolecular organic components, (ii) decomposition of organics through dehydration, bond cleavage, decarboxylation and deamination, and (iii) re-polymerization of highly reactive components to tar (Gong et al. 2017a, b). The tar and char generation are more evident in subcritical water gasification than in supercritical water gasification. Supercritical water gasification involves both ionic and free radical mechanisms. The high density of ionic components of water such as cationic hydron, i.e., H^+ and anionic hydroxide, i.e., OH^- are generated in low-temperature supercritical water gasification, which promotes the cleavage of polymeric compounds to form simple monomers (Guo et al. 2010; Nanda et al. 2015a). As the gasification temperature increases, supercritical water prefers free radical mechanisms involving hydrogen radical, i.e., ·H and hydroxyl radical, i.e., ·OH. The low density of supercritical water at high temperatures favor the generation of free radicals that lead to efficient conversion of organics to permanent gases (Susanti et al. 2012).

Hydrothermal gasification exhibits rapid feedstock hydrolysis, rapid degradation of organic polymers to monomers, enriched solubility of monomers, higher carbon conversion efficiency, increased syngas yields, suppressed char and tar generation, and lower chances of intermediates polymerization (Correa and Kruse 2018; Okolie et al. 2020b). Besides, supercritical water gasification is also advantageous in lowering the overall process expenditure due to the temperature requirement less than 600 °C than the thermochemical gasification, which operates at higher temperatures in the range of 600–900 °C. Owing to the high-pressure requirements, i.e., 23–30 MPa in supercritical water gasification, the resulting syngas is obtained at high pressures, which reduces the costs involved in its compression and storage. Greater yields of syngas are optimally found at high temperatures and pressures, longer reaction times and lower feed concentrations (Okolie et al. 2020c). Hydrothermal gasification also results in some liquid effluents that contain a broad range of products such as acetic acid, propionic acid, butyric acid, phenols, aldehydes, ketones, methanol, ethanol and furfurals (Nanda et al. 2015a, b, 2017a). Catalyst application in subcritical and supercritical water gasification can reduce the high-temperature requirement for efficient carbon conversion.

Onwudili and Williams (2016) performed catalytic supercritical water gasification of plastics, e.g., low-density polyethylene, high-density polyethylene, polypropylene and polystyrene in a batch reactor at 450 °C for 60 min. Hydrothermal gasification of plastics involved a series of reactions such as thermal degradation, steam reforming reaction, methanation and hydrogenolysis. The gasification of plastics produced H_2, CO, CO_2, CH_4 as well as trace gases, e.g., ethene, ethane, propene, propane, butene and butane. However, CH_4 was the predominant gas product with yields approaching 37 mol/kg from low-density polyethylene with 20 wt% ruthenium(IV) oxide/gamma-alumina, i.e., $RuO_2/\gamma\text{-}Al_2O_3$ catalyst.

Narobe et al. (2014) studied pyrolysis kinetics, thermodynamic equilibrium model and material balances during co-gasification of plastics with woody biomass in a 100-kW dual fluidized bed reactor, i.e., continuous stirred tank reactor. The modeling study results suggested that co-gasification of plastics with biomass could be an attractive technology for efficient thermochemical conversion as opposed to mono-gasification of biomass and plastics separately.

Acomb et al. (2014) have demonstrated the production of carbon nanotubes via simultaneous pyrolysis-gasification of waste plastics such as low-density polyethylene, polypropylene and polystyrene. In the first step, waste plastics were pyrolyzed under N_2 atmosphere at 600 °C to generate vapors and gases, which were later reformed with steam at 800 °C in the presence of a nickel/alumina catalyst, i.e., Ni/Al_2O_3. Three steam injection rates of 0, 0.25, 1.90 and 4.74 g/h were investigated. High-quality carbon nanotubes were generated from low-density polyethylene in the absence of steam and polypropylene and polystyrene at the lowest injection rate of 0.25 g/h. The carbon nanotubes produced from waste plastics were multiwalled, several microns long and with diameters ranging from 10 to 20 nm.

Zhang et al. (2015) also reported the production of carbon nanotubes and H_2 through a two-stage pyrolysis-catalytic reforming and gasification of waste tires at 600 °C. Among all the catalysts tested, i.e., cobalt/alumina, Co/Al_2O_3; copper/alumina, Cu/Al_2O_3; iron/alumina, Fe/Al_2O_3 and nickel/alumina, Ni/Al_2O_3, the latter resulted in the highest H_2 yield of 18.1 mmol/g was high-grade multi-walled carbon nanotubes. On the other hand, Co/Al_2O_3 and Cu/Al_2O_3 produced amorphous carbon, which repressed the quality of carbon nanotubes.

Alvarez et al. (2014) reported sequential pyrolysis-gasification of woody biomass and plastic, e.g., polypropylene, polystyrene, high-density polyethylene and real plastic waste mixtures with the aid of Ni/Al_2O_3 catalyst in a two-stage fixed-bed reactor. In the first stage, biomass-plastic blended feedstock was pyrolyzed at 600 °C to release the vapors. In the next stage, the released pyrolysis vapors passed into the gasification reactor maintained at 800 °C. Co-pyrolysis and co-gasification of plastics, i.e., 20 wt% of polypropylene with biomass showed synergistic effects by improving the gas yield of 56.9 wt% and H_2 production of 11 mmol/g. Ni/Al_2O_3 catalyst resulted in the cleavage of the carbon–carbon bonds, improved water–gas shift reaction and low coke formation, all of which led to high gasification efficiency and greater H_2 formation.

Kumagai et al. (2015) conducted successive pyrolysis-gasification of wood sawdust and polypropylene using a novel nickel-magnesium–aluminum–calcium catalyst synthesized by the co-precipitation method for H_2 production. The catalyst was investigated for steam cracking of hydrocarbons and water–gas shift reaction for in situ absorption of CO_2. The study emphasized on the catalyst activity and performance for thermal cracking of wood/polypropylene mixture. The synthesized catalyst calcined at 500 °C having a molar ratio for nickel-magnesium–aluminum-calcium as 1:1:1:4 gave the highest H_2 yield of 39.6 mol/g.

Parparita et al. (2015) conducted steam gasification of polypropylene and biomass waste composites in a dual-bed reactor at 700 °C with ferric oxide on ceria catalyst, i.e., Fe_2O_3/CeO_2. The yields of H_2 and heating values of the syngas products were synergistically superior in the case of co-gasification of polypropylene and biomass composites than those from mono-gasification of polypropylene and biomass separately. The results showed comparative decomposition of biomass and plastics during gasification. At low gasification temperature, biomass undergoes dehydration and decarboxylation to release moisture and volatile components followed by secondary reactions to generate char. Conversely, polypropylene, at low temperature, degrades into small molecular radicals and that take part in the gasification process.

Burra and Gupta (2018) have also reported the synergistic effects of the co-gasification of biomass and waste plastics. They performed steam gasification of wood pellets and plastics, e.g., polycarbonate, polyethylene terephthalate and polypropylene in a semi-batch reactor at 900 °C under atmospheric pressure. Compared to the separate gasification process, co-gasification of biomass and plastic mixture produced greater yields of total gases as well as H_2, CO and CO_2 yields. The devolatilization of biomass led to the release of volatile components. However, this was synergized by the thermal cracking of plastics, which generated methyl radicals to act as hydrogen donors to volatiles released from biomass degradation. Benzyl radicals enhanced solid-phase synergism causing higher carbon conversion to gas products. These interactions improved syngas yields, carbon conversion and energy efficiency.

Nanda et al. (2019b) performed subcritical and supercritical water gasification of waste tires to produce H_2-rich syngas. Several process parameters were investigated which include temperature, reaction time and feedstock concentration. Maximum syngas yield of 34 mmol/g was reported from the supercritical water gasification of waste tires at 625 °C in 60 min with 5 wt% feed concentration. Under these optimal conditions, the hydrogen yield was 14.4 mmol/g. Several homogeneous catalysts were also tested to enhance the hydrogen and total syngas yields from the hydrothermal gasification of waste tires. Among the catalysts examined, nickel on silica/alumina catalyst, i.e., Ni/SiO_2-Al_2O_3 was found to maximize the H_2 yields up to 19.7 mmol/g followed by ruthenium on alumina, Ru/Al_2O_3; barium hydroxide, $Ba(OH)_2$; calcium hydroxide, $Ca(OH)_2$; and magnesium hydroxide, $Mg(OH)_2$ that showed H_{-2} yields of 17.9, 16.9, 16.7 and 15.4 mmol/g, respectively. The study also reported many advantages of using catalytic supercritical water as the reaction medium for the gasification of waste tires, which includes high conversion rates, relatively lower temperature requirements, lower heat and mass transfer limitations, enhanced hydrogen yields and less hazardous byproduct formation.

Toledo et al. (2018) used a hybrid filtration reactor comprising of waste tires particles and alumina spheres under different gasifier agents, i.e., steam and air to produce syngas. The use of air as the gasifier agent resulted in the highest gasification temperature of 1294 °C, whereas varying the steam–air ratio from 0.95 to 2.85 resulted in relatively lower reaction temperatures due to endothermicity.

Machin et al. (2017a, b) performed the technical assessment of waste tires gasification for electricity and thermal power generation. The assessment of technical viability studies indicated that combustion of the waste tire derived-syngas in an internal combustion engine driving a generator could be an efficient route for electricity generation compared to the efficiency obtained by using the syngas in a gas turbine. Nevertheless, gasification stands out to be a promising route for valorization and energy recovery considering the complexity of waste tires and its heterogeneous composition consisting of rubber, elastomer, carbon black, metals, additives, textile, zinc oxide and sulfur.

Gasification of plastics under steam, air or hydrothermal conditions seems to be another promising valorization technology of waste plastics to produce predominantly H_2-rich syngas and aromatic char. However, the diversity, composition and concentration of these products depend on the physical chemistry of plastics and the gasification conditions such as temperature, feed concentration, residence time and catalyst loading. Several homogeneous and heterogeneous catalysts play distinctive roles in progressing the gasification of plastics through steam reforming, water–gas shift, hydrogenation, methanation and Boudouard reaction, thereby influencing the product gas yields.

Conclusion

Despite many social benefits of plastic products in our everyday lives, their adverse implications to the natural ecosystems associated with its durability and long-term stability in the environment should be evaluated. Reducing superfluous plastic consumption, using alternative eco-friendly packaging and innovating approaches to develop biodegradable plastics can address some challenges associated with plastics in the long-term. However, it is imperative to find solutions to manage the colossal plastic residues generated in the solid waste streams worldwide. After surveying the available literature on the statistical occurrence, classification, chemistry, recyclability and adverse environmental impacts of waste plastics, their alternative usage as an expedient and attractive feedstock to generate high-quality clean energy products through thermochemical conversion technologies can be proposed.

Valorization of plastics for fuel production can simultaneously address the issues of plastic waste management and alternative energy recovery. Pyrolysis is by far the most widely researched conversion technology compared to liquefaction and gasification. However, optimization studies pertinent to reaction temperature, heating rate, residence time, reactor design and choice of catalyst are crucial in determining the yield of gasoline and diesel grade fuels from plastics. Moreover, co-processing technologies such as co-pyrolysis, co-liquefaction and co-gasification, which involve the blending of biomass with plastics have tremendous environmental and economic advantages. There are many pronounced synergistic interactions between plastics and biomass, which result in the improvement of final product quantity and quality, lesser requirement of oil upgrading techniques and reduction in greenhouse gas emissions. Furthermore, the application of effective and selective catalysts in the thermochemical conversion of plastics can significantly lower the operating temperature requirement as well as enhance carbon conversion and process efficiency subsequently improving process-specific yields of either oil or gas. The sustainability of thermochemical valorization of waste plastics is undeniable considering their accumulation in the landfills and oceans as well as the potential to reduce the dependency on fossil fuel, increasing energy security and lowering environmental degradation by producing clean alternative fuels.

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