



# Plastic Recycling of Polyethylene Terephthalate (PET) and Polyhydroxybutyrate (PHB)—a Comprehensive Review

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## Abstract

As an integral part of plastic waste, large amounts of polyethylene terephthalate (PET), polyhydroxybutyrate (PHB), and solid waste have led to serious environmental problems. In order to identify critical barriers to the reuse of plastic across a wide range of plastic values all over the world, we need to go through PET and PHB in terms of volume. This is a significant factor for the world and our global warming. The current uses of plastic through PET and PHB are significant. Through the recycling process of PET and PHB, the total volume of plastic uses will be decreased and reuse rate will be high by economic, social, and environmental factors. Key areas are identified by low demand due to price considerations, insufficient tracking and transparency of pricing transactions, and lack of common design. Integration of value chains is considered to be the most important intervention for respondents, following the need for increased investment and technology development. Recycling helps the environment and creates new economic opportunities. Reuse of plastics encourages businesses to develop new and innovative products. Recycling reduces through extraction (mining, quarrying, and logging), refining, and processing of raw materials which create significant air and water pollution. As renewable energy saving also reduces greenhouse gas emissions, which help to cope with climate change. This paper aims to raise awareness that increase the demand of recycling process of PET and PHB and to identify critical barriers of recycling throughout a series of plastic prices with the volume.

**Keywords** Polyethylene terephthalate · Polyhydroxybutyrate · Recycling methods · Catalysis · Waste management · Environment

## Introduction

Recently, there has been concern regarding plastics due to no degradability, the closing of landfill sites, and incrementing land and water pollution problems. Raw materials are

increasingly in short supply like crude oil for synthesizing plastics, and waste plastics recycling is becoming more vital (Howard 2002).

The word “plastics” generally denotes the synthetic polymers ubiquitous in the current society where every individual uses 68 kg annually in the USA and 50 kg in the EU. In 2016, the world produced 242 million tons of plastic waste—12% of all solid municipal waste. The waste comes from three regions—57 million tons from East Asia and the Pacific, 45 million tons from Europe and Central Asia, and 35 million tons from North America. In 2015, the volume of global waste reached nearly 6.3 billion metric tons. This figure is expected to grow to 12 billion tons by 2050. There are now 5.25 trillion pieces of plastic and microscopic plastic pieces in our ocean and 46,000 pieces in every square mile in the ocean, weighing up to 269,000 t. Every day, eight million pieces of plastic enter our ocean (Samavedi et al. 2020; Ghaderian 2020). Our daily life is pervaded by plastics in packaging, sports equipment, and

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clothing, biomedical tools, electronic components, as well as other applications. Unfortunately, most of the shared plastics with high markets are caused by using nonrenewable and ecologically devastating petroleum or natural gas feed stocks and processing methods (Thiounn and Smith 2020). Plastics are the mixtures of compounds with high molecular weights known as polymers including polypropylene, polystyrene, polyvinyl chloride, PTFE, and polyethylene. Numerous fossil-based plastics are utilized in manufacturing made of nylon, petroleum, and elastomer (natural rubber) releasing dioxins when incinerated. Thus, disposal is a vast problem generating enormous hazards for the environment (direct or indirect impacts on carbon footprint) and humankind (accumulating in the food chains). Plastics are not capable of degrading; therefore, recycling is performed in some cases to decrease such disposal problems although this cannot be a decent remedy. Several novel technologies are progressed to make plastic degradable such as photodegradable plastics; however, scientists look for a better substitute to resolve this problem and create a degradable substance or control lifetime (Varsha and Savitha 2011). The importance of access to novel technologies for repurposing and recycling plastics is clear considering their dogged environmental persistence and unsustainable origins on land and in the oceans (Thiounn and Smith 2020).

Over the previous years, the incrementing rate of global consumption and production of plastics was alarming. Accumulating the persistent and pervasive waste plastic has increased concomitantly in the environment and landfills. The economic, ecological, societal, and problems of plastic waste or pollution demand decisive and immediate actions. Only 9% of the plastic waste was successfully recycled in the USA, in 2015. The main present recycling processes concentrate on the plastic waste mechanical recycling; nevertheless, even this procedure is restricted by sorting or pretreatment of plastic waste and degrading plastics over the procedure. Chemical recycling is an alternative to the mechanical procedures for plastic waste. Effective chemical recycling would make it possible to produce the feed stocks for different utilization including chemical feed and fuels stocks for replacing petrochemicals (Silva and Wiebeck 2020). Plastic recycling is a huge challenge as an opportunity for the circular economy.

Based on the literature and as explained in the standard organizations, polymer recycling is categorized into 3 classes associated with the resource management level and reduced processing expenses. Thus, polymer recycling operations provide some preferences with rising order going from primary to quaternary (Silva and Wiebeck (2020)). These preferences are consisting of primary recycling or reusing that includes reprocessing procedures of industrial polymeric residues. Mechanical or secondary recycling, which contains transforming the polymeric residues into the

novel products with a suitable performance for its particular application utilizing thermomechanical and mechanical processing; chemical or tertiary recycling procedures where the polymer residues are transformed or modified chemically into small molecules (like hydrocarbons or oils, for polyolefins), oligomers, or monomers. Generally, plastic pollution creates significant economic and environmental burdens as a result of consuming other natural resources and natural fuels (energy). Moreover, their long shelf time debases the environment in several ways. To reduce using plastic and thus decreasing its production is the only approach to diminish the dangers of plastic pollution. Recycling and degradation steps after diminishing this plastic were not considered since it creates further chemical and mechanical stresses and also is expensive. Biodegradable plastics were considered as a main and eminent method for solving this issue since the 1970s. Polyhydroxybutyrate (PHB) obtained major prominence from the inventory of biopolymers performing the role of bioplastics due to the analogous behaviors of PHB and petro-based plastics. PHBs are the linear polymers created by the microbes for storing carbon and energy. The polyhydroxyalkanoate (PHA) production technique, current advances in creating, applications, and degradation of PHB elucidating the obtained importance in the industrial world, nowadays (Varsha and Savitha 2011).

Plastics cannot be infinitely recycled, not utilizing traditional methods at least. They are mainly given only one new lease of life prior to their ending up in the earth, an incinerator, or in the ocean. However, it is also hoped to provide various types of recycling called chemical recycling. Traditional mechanical or physical recycling characteristically divides a plastic into smaller sections that are combined and molded together then to produce lower grade plastic products. Moreover, in chemical recycling, the plastic is broken down to the molecular level, hence, “platform molecules” are become available that can then be utilized to create other substances. Although this idea is new, it could open up a complete range of opportunities. Previous researchers have focused on plastics containing the chemical bisphenol-A (BPA). At the same time, ordinary polyethylene terephthalate (PET) plastic water bottles retained their reputation as safe, at least in terms of human health. But new evidence suggests that recycled PET or polyethylene terephthalate may be very less harmful for the upcoming green world.

## Plastic Recycling Management

Plastics are an extensive class of substances known as polymers that are made from small “monomer” building block molecules composed typically from hydrogen and carbon. The challenges in chemically recycling plastic include finding the proper methods for breaking down and reconstituting the materials into various end products and minimizing

waste. Hence, economic, productive, carbon-neutral, and large-scale methods are required in this regard. The ultimate solution should make less harm compared to the problem of solving. The monomers making up plastics can take various sizes and shapes such as possessing straight lines, rings in branched mode. The material features of plastic are determined by the way of bonding together such as easiness for breaking them down, and their melting temperatures.

In common recycling, plastic is just broken into small pellets. For example, Stock/Shutter stock or breaking chemical bonds in the simplest terms all requires energy. Largely, plastics are very stable; hence, they require a good deal of energy for breaking down in general, typically in the form of heat for creating a process known as pyrolysis. Particular control is also possible over breaking down through the right catalysts sparking the chemical reaction from a particular position into the polymer chain (Fig. 1).

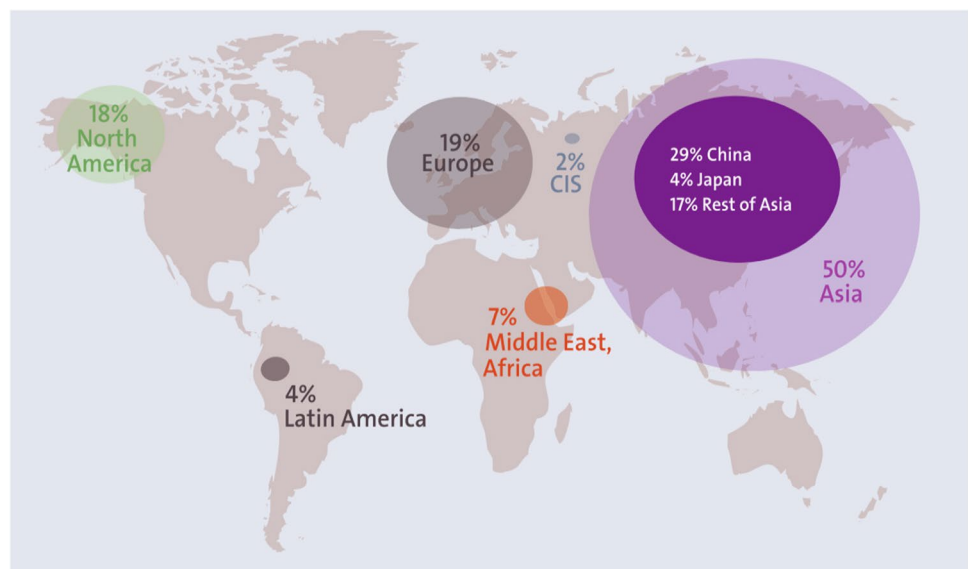
The biological molecule type, known as an enzyme is an example of a catalyst. These happen in living organisms and have a vital role in procedures in the body like digestion. Over 50 known plastivore microorganisms exist able to digest plastic due to the presence of enzymes helping for breaking it down. Biodegrading the plastics is a multifaceted procedure that contains numerous biotic and abiotic factors. By the cooperative action of microorganisms and abiotic factors, the bulk polymer fragmentation is caused to increment the available surfaces for biodegradation. More fragmentation of the polymers is conducted by some extracellular enzymes. Various lignin-degrading enzymes also take part in breaking down the polyethylene (PE) thermoplastic. Followed by the preliminary cleavage of the polymer into oligomers with 10–50 carbon atoms, they are conveyed into a cell for more metabolism. For instance, a lack of hydrolyzable functional groups in the

backbone restricts biodegrading the PE (Andler 2020). The hydroxyl and carbonyl groups created from different pretreatment like the addition of oxidizing agents or thermo-UV irradiation could be accepted for further stimulating of biodegradation. Therefore, the majority of the biodegradation works on PE have been conducted utilizing substrate peroxidation (Zimmermann 2020).

However, these natural procedures can be challenging to use since it is essential to maintain the biological organisms alive. Therefore, they need very precise circumstances like pH levels and temperature, and a long time is required for completing the procedure. Nevertheless, they might be commercially utilized in the future by more research.

Other catalysts can act quite rapidly. For instance, it was found that iron nanoparticles can be used to help turn black plastic (most difficult recycling) into carbon nanotubes in a matter of moments. Thus, this novel material can be used to create electrical components like data cables for transmitting information to a speaker system for playing music. There have been efforts in the world to establish new methods in this increasing field. Studies indicated that it is possible to recycle old cooking oil (a natural polymer) chemically into a biodegradable resin for use in three-dimensional printers. Other waste substances like rubber, food, and plastics can be utilized in quickly producing graphene (a type of carbon with the thickness of one atom). Scientists also established a method for repeatedly recycling bioplastics rather than leaving them to gradually biodegrading and releasing carbon dioxide. Procedures take expertise, money, and time. Nevertheless, until we stop utilizing plastics, this is an increasing investment field for developing a circular carbon economy as a result of plastic chemical recycling. Energy recovery or

**Fig. 1** The global plastics production. Source: Plastics Europe Market Research Group (PEMRG) and Conversio Market & Strategy GmbH (W. D'ambrières 2019)—reproduce permitted



quaternary recycling: technological solutions including the ignition of polymer waste for generating electricity, heat, and steam.

Chemical recycling could praise mechanical recycling, particularly for substances of the problem in physical recycling like microplastics and thin films. They are trapped in the grinding machinery as a result of the low strength and small size leading to getting stuck the whole system, slowing down, or even stopping completely and requiring cleaning. On thin films, the grinders cannot act even the microplastic materials with sizes hundreds of times smaller. Several methods have been found in the laboratory and now numerous companies exist performing this at a commercial level (Fig. 2; Table 1).

### Polymeric Mechanism of PET and PHB

PET as a semi-crystalline polymer belongs to the class of polyesters, with a melting point of ( $T_m$ ) about 250 °C and  $T_g$  of 70 °C. Despite the light weight of PET, its yield strength is about 40 MPa, and it has a tensile strength of about 170 MPa. Moreover, it has a very high resistance to wear and its flexural modulus is high. It is fairly impermeable to most liquids and gases, and this feature would be enhanced by an increase in its crystallinity or to copolymerize it with other monomers. After molding and setting, water is not absorbed by PET easily because of its crystalline nature. Besides, it has resistance to dissolution by common solvents. It has the biocompatibility feature, and FDA has confirmed it for repairing blood vessels and soft tissues (Samavedi et al. 2020). PET is the most popular material for packaging for drinking water. It has been about four decades that PET bottles have been marketed, and they are replacing glass bottles and polyvinylchloride (PVC) on the markets (Bach et al. 2012).

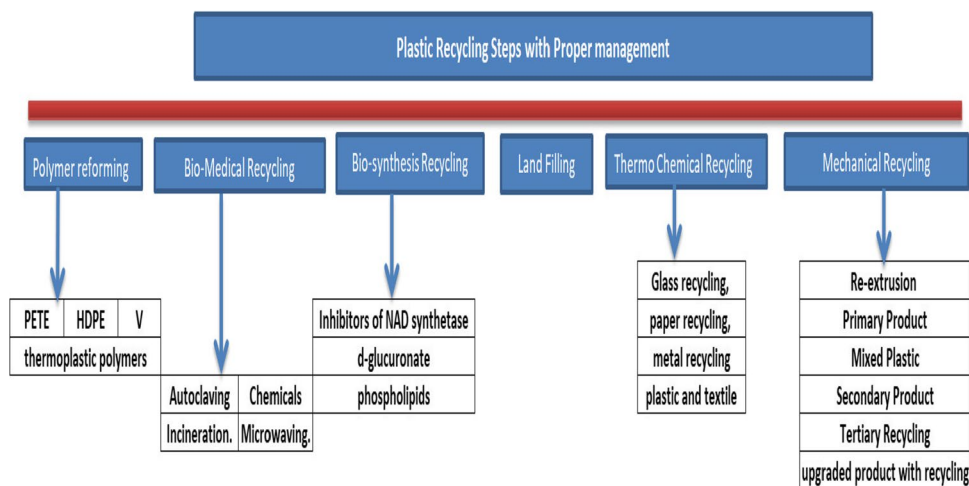
The first separated and recognized member in the family of PHA was PHB, which is considerably crystalline because

of its linear chain arrangement. It also includes both crystalline and amorphous phases. It is available as a virgin polymer or as part of blends and copolymers. Producers generate it as a carbon reserve used in various bacterial strains. Its industrial production is through bacterial fermentation. Moreover, PHB is more advantageous than synthetic polymers due to producing specific packaging applications. For example, PHB has a superior barrier permeability compared to polypropylene (PP) and polyethylene (PE). They also have higher rigidity and less flexibility compared to PP. In addition, PHB possesses great barrier characteristics compared to PVC and PET. The other significant property of PHB materials is their biodegradability, which occurs during a logical timescale when they have contact with degradation microorganisms in biologically active mediums, like freshwater, soils, and anaerobic and aerobic composting, and designate them as environment-friendly option substituting synthetic polymer (McAdam et al. 2020). The characteristics of PHB include possessing an ester linkage group ( $-COOR$ ) and a methyl functional group ( $CH_3$ ). These functional groups are in charge of the hydrophobic, thermoplastic, great crystallinity, and stiff features in the materials. The thermal features of semi-crystalline structures, like PHB and related by-products, are usually two primary temperatures: a  $T_m$  as the melting temperature for the crystalline stage and a  $T_g$  as the glass transition temperature for the amorphous stage. Besides, there exists  $T_d$  as the degradation temperature, which is the temperature when the material can initiate degeneration (McAdam et al. 2020) (Table 2).

### Advanced Recycling

Advanced plastics recycling, also known as chemical recycling indicates various technologies converting post-use plastics into their original building blocks, especially polymers, feed stocks for new fuels, waxes, plastics, and other valuable products (Fig. 3).

**Fig. 2** Different routes for plastic waste management

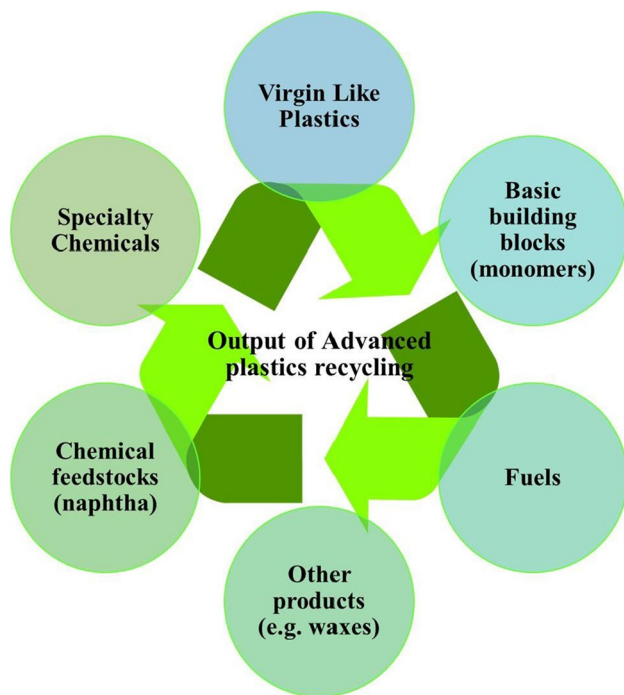


**Table 1** Plastic recycling for the different polymers and purposes in terms of environment

Symbol	Polymer name	Uses	Repurposed to make	Recycle to Environment
	Polyethylene Terephthalate (PETE or PET)	Soda bottles Water bottles Salad dressing bottles Medicine jars Peanut butter jars Jelly jars Combs Bean bags Rope Tote bags Carpet Fiberfill material in winter clothing	textiles, carpets, pillow stuffing, life jackets, storage containers, clothing, boat sails, auto parts, sleeping bags, shoes, luggage, winter coats.	Yes - reduce toxic emissions to air and water
	High-Density Polyethylene (HDPE)	Milk jugs Juice containers Grocery bags Trash bags Motor oil containers Shampoo and conditioner bottles Soap bottles Detergent containers Bleach containers Toys	Plastic crates, lumber, fencing	Yes - without recycle HDPE raises severe environmental issues, quite apart from toxic additives in PP such as lead and cadmium
	Polyvinyl Chloride (PVC)	Some tote bags Plumbing pipes Grocery bags Tile Cling films Shoes Gutters Window frames Ducts Sewage pipes	Flooring, mobile home skirting	Yes – without recycle toxic substances are harmful to the air quality of the immediate environment and can cause respiratory problems
	Low-Density Polyethylene	Cling wrap Sandwich bags Squeezable bottles for condiments such as honey and mustard Grocery bags Frozen food bags	Garbage cans, lumber	Yes – without recycle PE raises severe environmental issues, quite apart from toxic additives in PP such as lead and cadmium
	Polypropylene(PP)	Plastic diapers Tupperware Kitchenware Margarine tubs Yogurt containers Prescription bottles Stadium cups Bottle caps Take-out containers Disposable cups and plates	Ice scrapers, rakes, battery cables	It is susceptible to UV degradation onto our environment
	Polystyrene or Styrofoam (PS)	Disposable coffee cups Plastic food boxes Plastic cutlery Packing foam Packing peanuts	Insulation, license plate frames, rulers	In our environment, PS can harm wildlife and last for years and years.
	Miscellaneous Plastics (polycarbonate, polycyctide, acrylic, acrylonitrile butadiene, styrene, fiberglass, and nylon)	Plastic CDs and DVDs Baby bottles Large water bottles with multiple-gallon capacity Medical storage containers Eyeglasses Exterior lighting fixtures	Plastic lumber (which is often used in outdoor decks, molding, and park benches)	Plastic sticks around in the environment for ages, threatening wildlife and spreading toxins. Plastic also contributes to global warming

**Table 2** Summary of mechanical properties of and chemical structures of PET and PHB polymers

Mechanical property	PET	PHB
Degree of crystallinity (%)	7.97	50–60
Melting temperature (°C)	260	165–175
Glass Transition Temperature (°C)	67–81	5–9
Tensile modulus (GPa)	9.35	3–3.5
Tensile strength (MPa)	62	20–40
Elongation at break (%)	5–10	230
Chemical structure		

**Fig. 3** Advanced plastic recycling

We assist to preserve the original value of the plastics and conserve using the fossil-based resources through advanced recycling to create such outputs. Advanced recovery and recycling, chemical recycling, and transformational technologies are interchangeable. By such terms, advanced recycling is differentiated from the more extensively known recycling procedures using mechanical technologies for recycling utilized plastics.

Typically, in mechanical recycling, technologies are used for retaining the original physical type of utilized plastics. Hence, the plastic materials (resin) mainly remain the same over mechanical recycling. Normally, the utilized plastics are cleaned, sorted, melted, shredded, and then reextruded into the plastic pellets. Then, such pellets are utilized in the novel plastic packaging and other

products. The physical type of utilized plastics is altered by advanced recycling, either by dissolving the plastic with chemicals or through the heat for breaking down plastics into their original components. Hence, plastics are resultant in a purified form or feed stocks and chemical products that are then utilized for creating new plastics, other products, or fuels.

Advanced and mechanical recycling are complementary methods with various inputs and outputs. For instance, mechanical recycling acts well for particular forms, and resins such as bottles and containers are well-suited to create plastics for novel durable uses like pipe, pallets, and railroad ties. Nevertheless, sorting and processing some plastics are mechanically difficult. Moreover, it is more challenging to mechanically utilize recycled plastics in food contact packaging and applications. Furthermore, advanced recycling facilities can also set their outputs based on fluctuations in market demand.

The circular economy denotes an economic model where substances are repurposed not disposed of after use. A circular economy is different from the prevailing economic model where materials are largely disposed of after using that leads to numerous valuable substances either buried in landfills or littering the oceans and landscapes. Utilized plastics are repurposed and recovered in a circular sustainable model; however, it is not disposed of a model that contributes to retaining plastics out of the environment and connects their inherent value to make novel products.

Advanced and traditional recycling can significantly have a role in a circular economy by repurposing utilized plastics. Further types of post-use plastics are captured by emerging and ongoing advances in mechanical recycling; however, advanced recycling captures mainly utilized plastics that are not extensively recycled nowadays. By this mixture, will help to obtain America's Plastic Makers' objective of recovering and recycling 100% of utilized plastic packaging in the USA by 2040. There are enormous economic and environmental opportunities for advanced recycling. Advanced recycling can have a significant contribution to the circular economy where plastics are repurposed, not disposed, helping to keep plastics out of the environment or ocean and connects their inherent value for making novel valuable products. Based on a study from Argonne National Laboratory in 2017, for instance, making ultra-low sulfur diesel fuel from utilized plastics could reduce using water by 58% and using traditional energy sources by 96% (Ghatge et al. 2020). Moreover, advanced recycling can assist local and national economies, particularly in the regions with less infrastructure or no infrastructure for managing and recycling waste. A current report by Closed Loop Partners (CLP) (an organization investing in developing the circular economy) found that there are remarkable demands for the advanced recycling products. Three main classes of advanced recycling

technologies exist processing utilized plastics into novel products and plastics (Fig. 4).

Most of the technologies utilized in advanced recycling have been economically arranged in different industrial procedures such as producing industrial waxes. However, despite the technology, sorting, gathering, and processing utilized plastics provide several challenges the same as those characteristic in mechanical recycling. Nevertheless, considering the extensive range of outputs from advanced recycling, there are expansive markets for advanced recycling.

Advanced recycling technologies normally utilize thermal energy (heat); however, their physical forms are changed to create new chemicals, products, and fuels rather than combusting the plastics. Advanced recycling is an engineering procedure where processing the raw materials (utilized plastics) is performed to create novel products (chemicals, plastics, and fuels), similar to the other manufacturing facilities.

In the procedures utilized in advanced recycling of utilized plastics, the thermal or chemical procedures are utilized in the nonexistence of oxygen; hence, emissions from advanced recycling are normally regarded as very low.

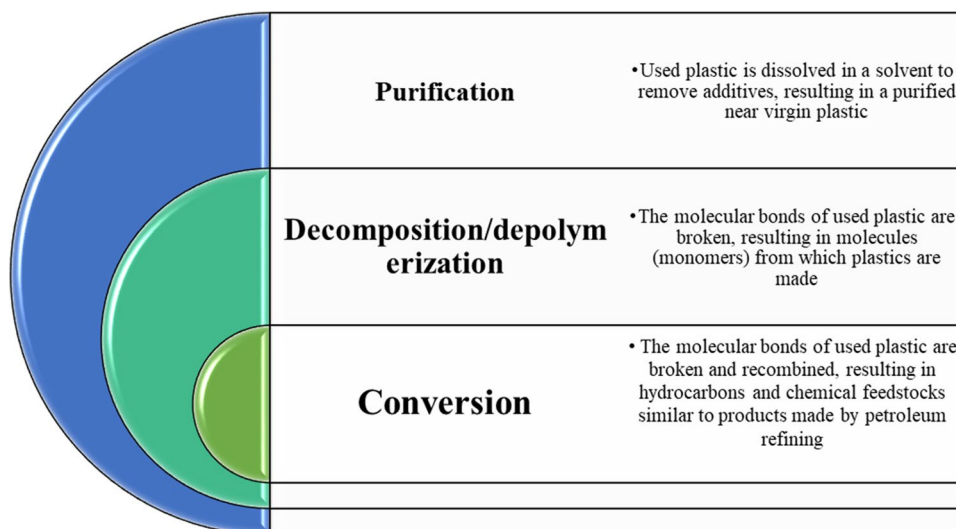
Although some manufacturing processes can lead to the environmental discharges, there were lower emissions from facilities using a usual advanced recycling technology (pyrolysis) in comparison to several commercial and industrial facilities like food manufacturing, universities, and hospitals. A non-hazardous by-product is created by this same technology, which can be controlled the same as other solid waste leading to a waste reduction of about 90% (Wang et al. 2019).

Although through thermal, chemical, and mechanical means, recycling plastic waste can be performed, they possess definite restrictions. In comparison to the chemical and mechanical recycling, plastic waste can be thermally treated while gaining more importance recently as a result

of the simple procedure and flexibility in the availability of the raw materials. The plastic waste is heated within the pyrolysis procedure, at a higher temperature at the range of 100–390 °C in the nonexistence of oxygen for a given period for breaking the polymer molecules into created vapors and monomers are condensed to attain gaseous and liquid fuel. High calorific-value natural gas and liquid fuel are generated by pyrolysis as the main products with char as a by-product. These can be used as a feedstock for solving the plastic waste problem and recovering high calorific value fuel from waste plastic (Ghamandea et al. 2019) (Table 3).

There has been a great contribution between different recycling technologies of plastic solid waste (PSW) such as primary (reextrusion), secondary (mechanical), tertiary (chemical), and quaternary (energy recovery) outlines and technologies and the waste management eco-image, mostly to PSW recycling, recovery, and treatment. The present situation may be beneficial for decreasing and reusing single-life polymeric materials. By starting the recycling loop in a processing line, it can be integrated with the procedure scrap reextrusion happening at various scales with various thermoplastics. Definite disadvantages are observed when selecting mechanical recycling as a route of recycling. The kinds of polymer-based plastic, its suitability, and the condition and the involved intense energy consumption are all the main issues regarding PSW. For successful practical use of such recycling approaches, it is worth noting that by-products caused by different mechanical treatments need to have similar features of commercial-grade plastics based on their monomer origin and type. A more sustainable solution is the tertiary treatment of waste plastic articles. Valuable petrochemicals are recovered as feedstock, providing a recycling route in the process. Moreover, it creates energy in the form of steam and heat. PSW (plastic solid waste) is derived from oil and possesses recoverable energy, in some cases

**Fig. 4** Three primary categories of advanced recycling technologies



**Table 3** New forms of waste recycling application

Industry	Materials	Waste	Properties	Ref
Civil	PET	Thermoplastic, elastomeric, thermosetting	Contributing to reducing energy consumption and natural resources  Improvement of tensile strength and chemical resistance, reduction of drying shrinkage, and also the possibility to decrease raw material costs	Jassim (2017)
Wood	PET and PHB	Thermoplastic	Loss of mechanical properties	Poletto (2017)
Segments of different industrial (the furniture and automotive ones)	PHB	Few thermoplastics (processing temperature < 200 °C)	Low cost, possibility of being recycled several times, and excellent processability by conventional polymer processing techniques	Borah and Kim (2016)
Other composite materials	PET	Thermoset composite	An alternative chemical recycling for fibers or fillers	Vercher et al. (2020)

compared to other energy sources. Certainly, direct incineration through one or two-stage combustion technologies can decrease the volume of PSW and the reliance on fossil fuels, which can result in better conservation of natural resources and combined waste management outlines. It is very significant to take into account energy recovery and recycling approaches in converting facilities and plastic manufacturing. Several quaternary and tertiary technologies seem to be strong in warranting more research and developing soon (Silva and Wiebeck 2020; Al-Salem et al. 2009). Table 2 obtains new forms of polymer waste recycling application.

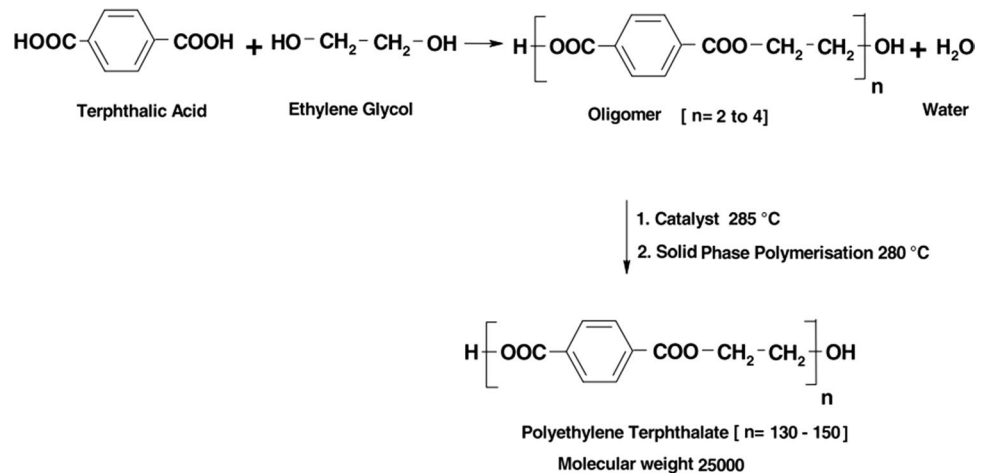
## Polyethylene Terephthalate

PET is a synthetic fiber that belongs to the polyester family (Sulyman et al. 2016). It is classified as thermoplastic, a type of plastic that becomes hard when cooled and soft when heated (Sulyman et al. 2016). The glass transition temperature of PET varies between 67 and 81 °C and has a melting point of 260 °C (Lanaro et al. 2018).

PET is a common name for a unique plastic family of polyester. PET polyester is composed of ethylene glycol (EG) and terephthalic acid (TPA), sometimes called “purified terephthalic acid” or PTA. The full chemical name for PET is polyethylene terephthalate. Polyethylene is an important member of the polyolefin framework. It is the most widely used plastic in the world, making products ranging from clear food wrappers and shopping bags to lubricants and car fuel tanks.

The intrinsic value of a standard bottle-grade PET is 0.80 dL/g (Padsalgikar 2017). PET is formed by heating ethylene glycol (EG) and terephthalic acid (TPA) together. When heating, a mixture consisting of the monomer bis(2-hydroxyethyl) terephthalate (BHET) and low molecular weight polymers is formed. Excess EG is then distilled and PET in the form of molten liquid is formed. It is then extruded and quenched with water to produce a glasslike amorphous material (Al-Sabagh et al. 2016) (Fig. 5).

Once the plastic with this above polymeric chain has been collected and sent to a recycling facility, it is usually separated by a variety of polymer types, separated

**Fig. 5** Molecular combination of assembling/PET



separately (and paper-like impurities are removed), and then melted back into the polymer pellets. These particles are marketed for use in new products. Simple plastic recycling processes involve collecting, filtering, grinding, washing, melting, and softening. Certain processes vary depending on the type of plastic or the type of plastic product. Thermoset plastics contain polymers that form chemical bonds that cannot be repaired and cannot be recycled, while thermoplastics can be melted and reconstituted.

Catalyst is used during the process in extremely low concentration to speed up the reactions and reduce cost. Antimony trioxide is the commonly used catalyst, with salts of titanium, cobalt, magnesium, germanium, manganese, and zinc being the other catalysts that can be used. The small amounts of catalyst remain in the polymer chain itself. PET can also be prepared in the laboratory by combining terephthaloyl chloride and ethylene glycol. Even though this is easier, it presents more risk as it is more dangerous (Al-Sabagh et al. 2016). The large aromatic ring in PET contributes to the stiffness and strength of PET which makes it used in many applications due to its strong and lightweight properties (Fig. 6).

Due to its wide usage, it is also the most recycled plastic. Recycling of PET will reduce the amount of space needed for landfill and also the amount of raw material required to produce new PET. Currently, the recycling rate in the USA for PET is 31% while the rate is 52% in Europe (Ghaderian 2020). There are two ways to recycle PET: physical and chemical recycling.

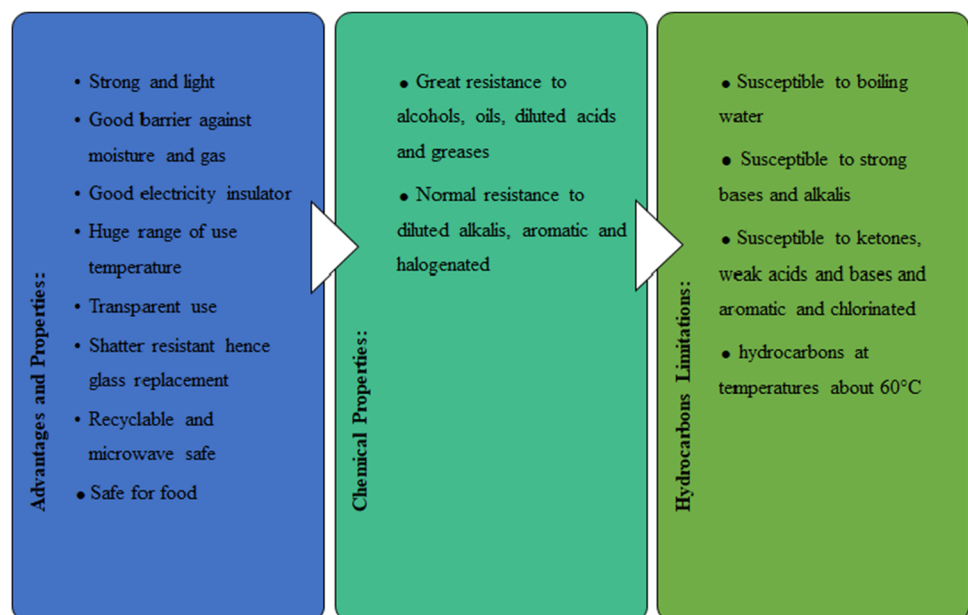
A recycling center, recycling facility, or multirecycle plant (MRF) is a specialized plant that acquires, classifies, and prepares recyclables for retail manufacturers. In

general, there are two distinct types: clean areas and access to building materials.

Waste enters the MRF when it is dumped down to the waist by collection trucks. Resources are collected at the top and placed on conveyor belts, which move it to the prebuilt location. Here, human workers remove other non-renewable materials, which will be sent to a landfill or to a heat sink. Potential hazards are also eliminated, such as lithium batteries, propane tanks, and aerosol cans, which can cause fires. Items such as plastic bags and pipes, which can hold recycling equipment, are also removed. From there, objects are transferred by another transmission band to a disk screen, which separates wide and flat objects such as soft cardboard boxes into items such as cans, jars, paper and bottles. Paved boxes mounted on the disk screen to the other side, while all other items fall to the bottom, where the paper is separated from the debris by a blower. The distribution of cardboard and paper is overseen by more than just human staff, who ensure that there is no plastic, metal, or glass. New or redesigned MRFs can use industrial robots instead of humans for prescreening and quality control. The metal is separated from the plastic and glass first by electromagnet, which removes the metal metals. Non-ferrous metals such as aluminum are removed with current eddy separators.

Glass and plastic streams are separated by continuous disc screens. The glass is crushed into a cullet for easy transport. The plastics are then separated by a polymer type, usually using infrared technology. Infrared light contrasts differently with different types of polymers; once identified, the airplane puts the plastic in the appropriate container. MRFs can only collect and recycle a few polymers in plastic, sending the rest to waste or hot dumps. Separated items are bet and shipped to the shipping port of the facility.

**Fig. 6** Advantages, properties, and limitations of PET



## Physical Recycling

Physical recycling of PET can be done through salvaging industrial scrap and physical reprocessing (Johnson et al. 2015). Salvaging industrial scrap involves the mixing of the recycled scrap with virgin material to maintain product quality. It can also be used itself as a second-grade material. Physical reprocessing involves grounding, removal of contamination, drying, and melting. Table 4 shows advantages and disadvantages for physical recycling of PET.

### Removal of Contamination

The PET waste is grounded into flaked or pelletized form after being collected (Paszun and Spychaj 1997). The flakes and pellets are then washed in 2 different ways. A hot 2% sodium hydroxide (NaOH) solution mixed with 80 °C detergent and a cold wash with water after. Another way consists of a solvent consisting of tetrachloroethylene (TCE).

### Drying

Drying reduces the amount of moisture in used PET flakes to reduce degradation due to presence of water. Thermoplastics, such as PET, are usually easier to reuse because the polymer chain drops to a very low temperature so there is no reduction in the polymer chain during the recycling process. Polymers are available. As a result, the entire recycling collection can be contaminated.

### Melting

The PET flakes can then be processed into granules through melt extrusion. Thermoplastic materials become liquids where they dissolve (approximately 260 °C in the case of PET). The great useful thing about thermoplastics is that it can be heated to its melting point, cooled, and heated again without much damage.

## Chemical Recycling

Chemical recycling is achieved through transforming polymer chain. The polymer backbone in PET is depolymerized (transformed into monomer units) during the recycling process or randomly broken into larger chain fragments.

Chemical recycling is done through solvolysis or pyrolysis. Solvolysis involves solvents to break down the PET while pyrolysis involves heating without the presence of oxygen or air to break down the PET. Monomers are obtained from chemical recycling and the monomers are used to manufacture polymers after being purified by distillation and drying. There are 5 different ways to recycle PET chemically: hydrolysis, aminolysis, ammonolysis, methanolysis, glycolysis (Fig. 7).

### Hydrolysis

This process hydrolyze PET into TA and EG. The high pressure (1.4–2 MPa) and temperature (200–250 °C), together with the long duration for complete depolymerization, make the process unideal. Due to the huge cost required to purify the recycled TPA, it is not often used for recycled food-grade PET. Hydrolysis can be performed via 3 different methods: (1) acid hydrolysis, (2) alkaline hydrolysis, and (3) neutral hydrolysis (Geyer et al. 2016).

### Alkaline Hydrolysis

Alkaline hydrolysis is usually conducted with either aqueous alkaline solution of NaOH or potassium hydroxide (KOH) of concentration 4–20 weight percent (wt%) (Al-Sabagh et al. 2016). The best results are obtained when the weight ratio of PET to NaOH is 1:20 at around 100 °C for 2 h. In reaction with PET, aqueous solution of NaOH is the least reactive and sodium ethoxide is the most reactive. Ether (dioxane, tetrahydrofuran, etc.) mixed with alcohol (methanol or ethanol) will speed up the reaction. With dioxane as a co-solvent, complete reaction (more than 96%) of solid PET and NaOH in 60 °C methanol will take 40 min. Without dioxane, the reaction will take 7 h (Fig. 8).

**Table 4** Advantages and disadvantages of physical recycling for PET

Advantages	<ul style="list-style-type: none"> <li>• Simple</li> <li>• Low investment cost</li> <li>• Flexible feedstock volume</li> <li>• Negligible impact on environment</li> </ul>
Disadvantages	<ul style="list-style-type: none"> <li>• Reduced melt viscosity due to hydrolytic and thermal degradation</li> <li>• Generation of cyclic and linear oligomers, affecting the printability and dyeability of the final PET product</li> <li>• Yellowing of PET due to oxidation and intramolecular cross-linking</li> <li>• Forming of acidic compounds due to contaminants speed up hydrolysis of the ester linkages in PET during thermal reprocessing</li> </ul>

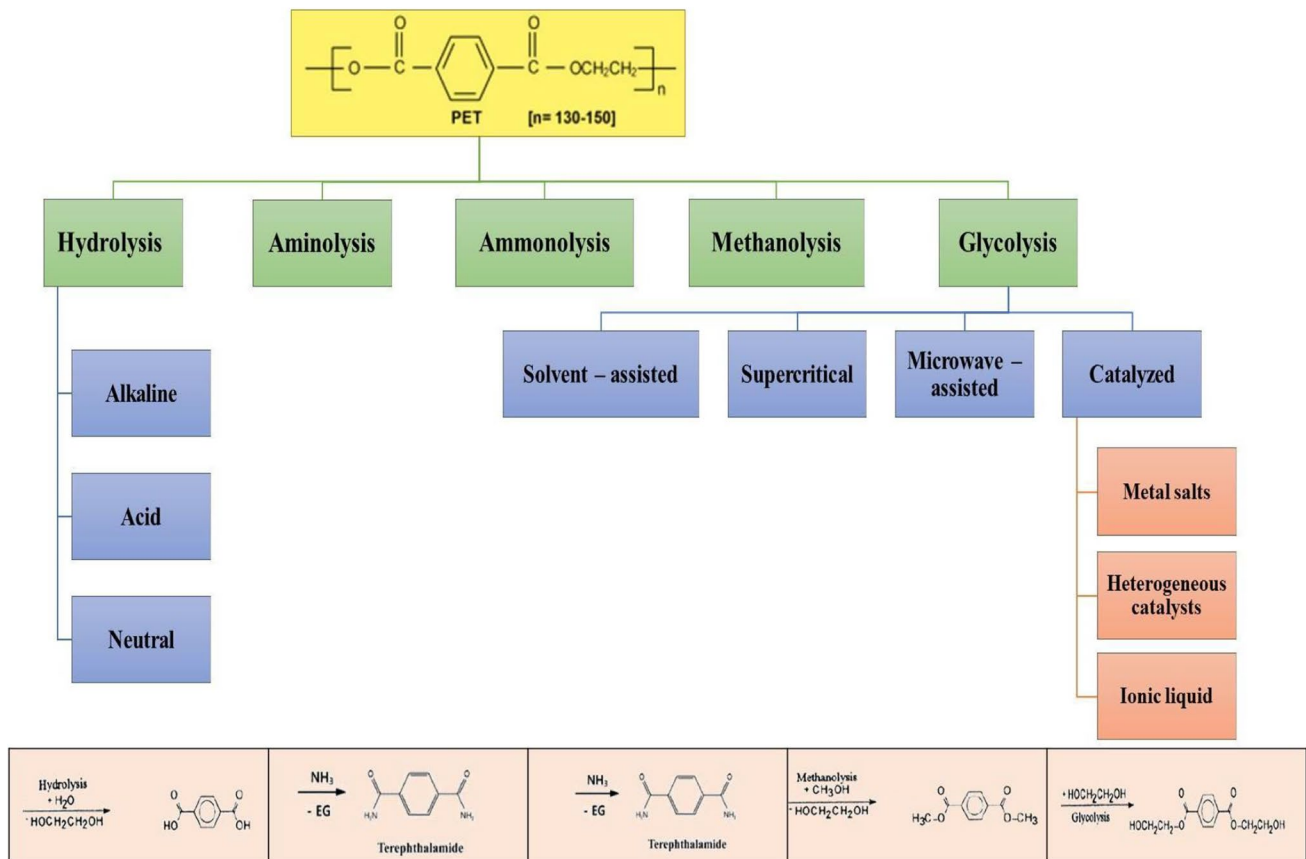


Fig. 7 Five different ways to recycle PET chemically

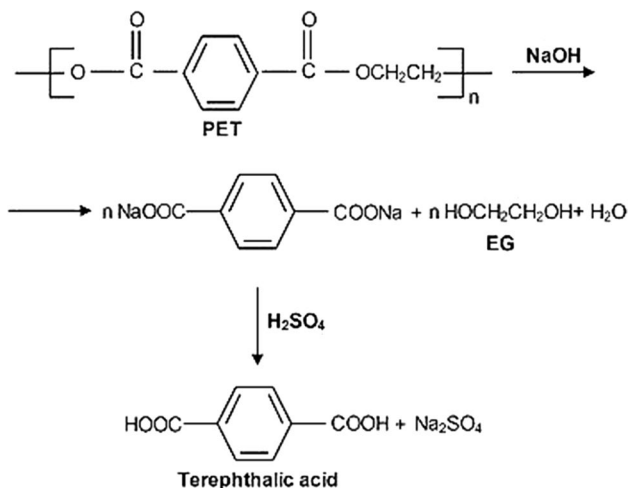


Fig. 8 Alkaline hydrolysis of PET with NaOH or KOH (Al-Sabagh et al. 2016)

For PET hydrolysis in non-aqueous alkaline solution, PET is mixed with non-aqueous ethylene glycol solution. Solid NaOH used in a mixer-extruder at 100–200 °C will

result in depolymerization. NaOH solution in methanol react faster with PET than analogous aqueous solutions (Karayannidis et al. 2002; Kumar and Guria 2005).

Catalyst can also be used to conduct alkaline hydrolysis of PET in low temperatures (below 100 °C) and atmospheric pressure. This catalyst is known as the phase transfer catalyst (PTC) (López-Fonseca et al. 2008). Initially, a mixture of aqueous NaOH and solid-organic phase of PET is formed. The catalyst, trioctyl methyl ammonium bromide, forms an ion pair with inorganic salt in aqueous form through an ion-exchange reaction. Due to the lipophilic nature of the ion pair, it can pass through the solid-liquid interface. The hydroxide anion is carried by the cation part of the catalyst to the surface. The PET macromolecule then depolymerize due to the attack of the hydroxide anion group. This process produces terephthalate anion, which forms disodium terephthalate salt with sodium ion. The reaction continues until PET is completely depolymerized to TPA- $\text{Na}_2$  and EG. The catalyst will remain in aqueous phase after the whole reaction (Langer et al. 2020).

## Acid Hydrolysis

The most common method of acid hydrolysis is using concentrated sulfuric acid. Other acids such as nitric or phosphoric acid have also been used. The use of concentrated sulfuric acid removes the need of high temperature and pressure. It is a costly process as large amount of concentrated sulfuric acid has to be recycled and removed from EG. TPA is either obtained from 60 to 93 °C concentrated sulfuric acid or 85 to 90 °C 90wt% sulfuric acid (Dutt and Soni 2013). The use of concentrated sulfuric acid leads to generation of huge amount of inorganic salts and aqueous wastes due to its high corrosive nature. Another way to depolymerize PET is the use of nitric acid. Nitric acid is mixed with PET powder at 70–100 °C for 72 h. This process produce both TPA and EG, and the EG is then oxidized to oxalic acid. The advantage of this method is the creation of oxalic acid, which is a value-added product.

## Neutral Hydrolysis

Neutral hydrolysis is conducted using hot water or steam. This process is conducted under the condition of 200–300 °C and 1–4 MPa. PET to water ratio varies from 1:2 to 1:12. Hydrolysis of molten PET is much faster than solid PET; hence, the temperature for depolymerization is recommended to be above 245 °C. At 275 °C, the time taken for complete depolymerization is roughly an hour. The depolymerization reaction is catalyzed by the carboxylic groups formed during the reaction. The TPA efficiency is above 90% when reaction temperature is above 420 °C while the EG efficiency of above 60% (maximum) is achieved when temperature is 300 °C.

Neutral hydrolysis does not produce large amount of inorganic salts and also does not require addition of acids and alkaline. However, the TPA produced contains impurities, which results in a less pure TPA obtained compared to acid and alkaline hydrolysis (Siddiqui et al. 2010; Kang et al. 2020).

## Aminolysis

Aminolysis of PET produce diamides of TPA and EG. This method is less explored as compared to other methods. Partial aminolysis is found to improve properties of PET fibers. This process is conducted in an aqueous or gaseous state at temperature range of 20–100 °C. The amines that are used in this process are usually ethylamine, methylamine, ethanalamine, butylamine, and triethylenetetramine. An aminolytic agent, anhydrous n-butylamine, with a temperature of 21 °C was also used (Padhan and Sreeram 2019).

Aminolysis of PET can also be done with excess ethanalamine. Catalysts to be used in this reaction are glacial

acetic acid, sodium acetate, and potassium sulfate. The product obtained from this reaction is bis(2-hydroxyethyl) terephthalamide at a 91% yield (Gupta and Bhandari 2019).

## Ammonolysis

Ammonolysis, the reaction between ammonia and PET under the presence of ethylene glycol, produces terephthalamide (Spychaj 2002). The pressure and temperature required for this process is 2 MPa and 120–180 °C, respectively. The process will last from 1 to 7 h, and the end product amide is filtered, washed with water, and dried at 80 °C. The purity of the product is a least 90% and has a yield of more than 90%. A low pressure method of ammonolysis can also be conducted by using zinc acetate of 0.05 wt% as catalyst and conducted at 70 °C. The ratio of PET to ammonia is 1:6. A yield of 87% of terephthalamide is produced.

## Methanolysis

Methanolysis refers to the degradation of PET with methanol at high pressures and temperatures, producing dimethyl terephthalate (DMT) and ethylene glycol (EG). This process is conducted at 180 to 280 °C and pressure of 2 to 4 MPa (Al-Sabagh et al. 2016). Catalyst is also used for this reaction, and the most common catalyst is zinc acetate. The catalyst has to be deactivated after the reaction to prevent the reduction of efficiency of DMT due to transesterification reaction with EG. Terephthalate produced in this reaction will be purified and used in the production of PET as a raw material (Pudack et al. 2020).

The equipment required for methanolysis are a reactor, crystallizer, centrifuge, and a facility to melt and purify DMT. For the process to run continuously, raw materials have to be transferred to a high-pressure reactor, resulting in a more complex system. A two-stage methanolysis process was designed by Hoechst which PET melted at 265 to 285 °C is passed as liquid form into the reactor. Preheated methanol at 190–210 °C is added into the reactor at a ratio of one-part PET to four parts methanol. In the first reactor, the percentage converted is 70 to 90%. In the second reactor, the reaction is continued at a lower temperature. The mixture is then cooled to 100 °C in the mixer and DMT is obtained through precipitation and purification (Vaidya and Nadkarni 1987).

Another way of methanolysis is done through the injection of mixture of PET and inert gas into the reactor with gaseous methanol. Before this process, PET is melted with superheated steam and solidified into powder form. The powdered PET is 1 mm in size, and the temperature in the reactor is kept at 250–300 °C (Sinha et al. 2010).

Another method of methanolysis is the treatment of PET with supercritical methanol. DMT, EG, and oligomers are

produced through the complete depolymerization of PET in this process at 300 °C and 11 MPa without a catalyst for 30 min. The use of supercritical methanol (239.15 °C and 8.09 MPa) allows faster reaction rate. There are also other ways of using supercritical methanol. DMT efficiency of 97% and 97.7% can be achieved at 300 °C and 310 °C, respectively. The most efficient reaction occurs at a temperature range of 259.85 °C to 270.85 °C, pressure range of 9 to 11 MPa, weight ratio of PET to methanol of 8:6 and duration of 40 to 60 min (Kárpáti et al. 2019).

Alcoholysis of PET through combination with trimethylolpropane produce PVC plastisols when mixed with PVC under the zinc acetate catalyst (Kárpáti et al. 2018). The main advantage of alcoholysis is that the process can be installed in the polymer production line as DMT produced is of the same quality as virgin DMT. EG and methanol can also be recovered and recycled easily, leading to the reuse of waste materials to produce PET (Karayannidis et al. 2002). However, the cost of separating and refining of reaction products (glycols, alcohols, and phthalate derivatives) from the mixture is high. Another disadvantage is the cost required to convert DMT to TPA due to the widely preferred TPA in PET production processes (Al-Sabagh et al. 2016).

## Glycolysis

Glycolysis is the most studied process for chemical recycling of PET. It works by breaking down the ester linkages in PET with glycol and produce bis(hydroxyalkyl) terephthalate monomer and a mixture of hydroxyl terminal groups with oligomers. The most common glycol used in glycolysis is EG. It is the cheapest, simplest, and oldest process. The main product from this reaction is bis(2-hydroxyethyl) terephthalate (BHET) with the excess usage of EG (Vinnakota 2018). During PET degradation, BHET monomer is transformed to a dimer and equilibrium is obtained with presence of polycondensation. Both processes are reversible and extending the reaction will reduce the amount of BHET monomer and increase the amount of dimer and oligomers (Langer et al. 2020). PET glycolysis without a catalyst is extremely slow and PET cannot be completely depolymerized. Furthermore, the products of the reaction contain very little BHET monomer, making it difficult to recover BHET monomer from the mixture of products. Therefore, to increase the amount of BHET monomer in reactions, catalysts have to be added. There are 4 different methods perform PET glycolysis: solvent-assisted glycolysis, supercritical glycolysis, microwave-assisted glycolysis, and catalyzed glycolysis (Yue et al. 2013).

## Solvent-Assisted Glycolysis

This PET degradation process by ethylene glycol involves the presence of a reaction medium in the form of a solvent. The addition of xylene to PET glycolysis reaction with the presence of zinc acetate as catalyst results to a higher BHET yield with and without the presence of xylene. Xylene was used to allow PET-glycol mixture to mix better. EG dissolves sparingly at 170 to 225 °C, while xylene dissolves readily. The glycolysis products can dissolve in xylene and the glycolysis products change to xylene phase as the reaction progress. The use of organic solvents is harmful to environment, and hence, massive adoption of the solvents is not recommended (Mejjatti et al. 2014; Vinnakota 2018).

## Supercritical Glycolysis

Supercritical glycolysis sets ethylene glycol to a temperature and pressure above its critical point. This process removes the need of a catalyst and is environmental friendly. In a study, using EG at 446.7 °C and 7.7 MPa in a process where the temperature is 450 °C and 15.3 MPa achieve a faster BHET-dimer equilibrium than subcritical processes that are conducted at 350 °C and 2.49 MPa and 300 °C and 1.1 MPa. The supercritical process only took 30 min and achieved a 93.5% BHET yield. The only downside to this process is the high temperature and pressure needed (Al-Sabagh et al. 2016).

## Microwave-Assisted Glycolysis

Microwave radiation as a heat source reduces the time required to complete from 8 h to 35 min but does not increase the yield of monomer. The addition of catalyst might be able to decrease the reaction time and increase the yield of BHET. In a study conducted, it is found that the presence of alkali catalyst in non-aqueous glycolysis with microwave irradiation is able to reduce the reaction time and come up with an affordable treatment process. Alkali concentration and irradiation were found to be the main factors affecting the treatment efficacy. Irradiation determines the temperature of the system. In another study, reaction time of PET with diol of different molecular weights catalyzed by zinc acetate (0.5%, w/w) under microwave irradiation can be reduced to 30 min as compared to the commonly used thermal glycolysis process that often takes 8 to 9 h minimally (Ikenaga et al. 2016). Recently, PET depolymerization using the microwave-assisted method via ZSM-5-based catalyst has been demonstrated (Kang et al. 2020).

## Catalyzed Glycolysis

Without catalyst, glycolysis is a very slow process. The most explored method to increase glycolysis rate is using catalyst. Since PET glycolysis is a transesterification reaction, transesterification catalysts are used to increase the rate of reaction. Out of all the catalysts, metal-based catalysts are the most popular (Hu et al. 2020).

In Fig. 9, the carbonyl carbon in the ester group of the PET is attacked by a free electron pair from the EG oxygen. The carbonyl carbon in polyester then forms a bond with the hydroxyethyl group in EG, with breaks the long-chain to short-chain oligomers then BHET (Al-Sabagh et al. 2016).

Rate of glycolysis is affected by a few parameters such as pressure, temperature, PET/EG ratio, type, and amount of catalyst used. The formation of BHET monomer through transformation of dimer is also a reversible process. Once equilibrium is achieved, prolonging of the reaction will result in a backward shift of the reaction. This will reduce the amount of BHET monomer and increase the amount of dimer. Thus, optimum condition must be understood for the reaction (Sheel and Pant 2019). A complex is formed between the carbonyl group and metal in metal-based catalysts. This complex controls the attack of EG on PET, resulting in the formation of BHET. Through different studies,

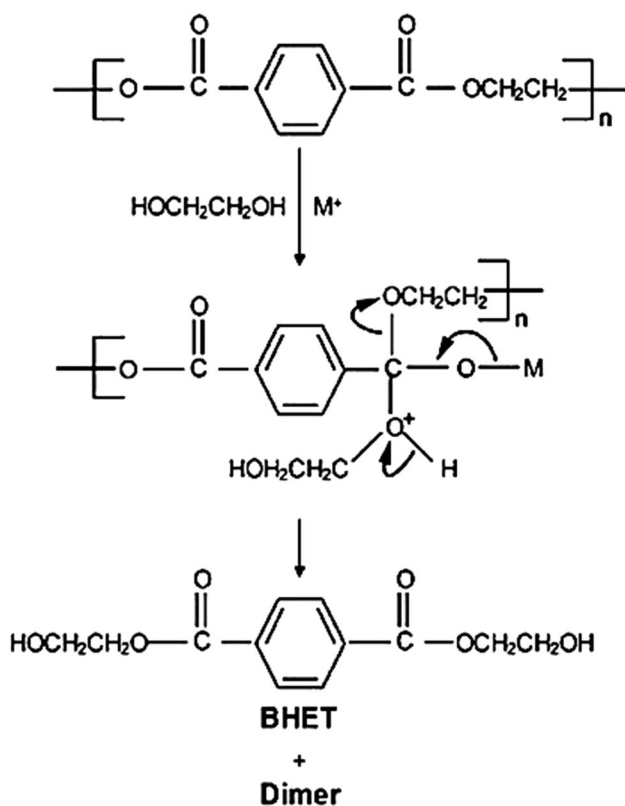


Fig. 9 Glycolysis with metal-based catalysts

there are plenty of catalysts found to be involved in glycolysis depolymerization process (Hu et al. 2020).

## Metal Salts

The first catalysts used for PET glycolysis are metal acetates, with zinc acetate being the first catalyst used to synthesize polyester polyols from waste PET (Al-Sabagh et al. 2016). In a study in 1989, it is found that out of all the metal acetates (zinc, cobalt, lead, and manganese), zinc acetate shows the greatest extent of depolymerization of PET. The reaction between BHET monomer and dimer reached an equilibrium after 8 h of reaction at a temperature of 190 °C. It is also found that the reactivity of catalysts goes from zinc being the most reactive, followed by manganese, then cobalt and lastly lead. With the use of zinc acetate as catalyst, equilibrium between the reaction of BHET monomer and dimer was achieved in 2 h as compared to 8 h (Al-Sabagh et al. 2016).

Another catalyst, titanium(IV) phosphate, was also discovered to be better than zinc acetate. The titanium(IV) phosphate catalyzed reaction at 200 °C and 0.003 catalyst/PET weight ratio for 150 min produce a yield of 97.5% BHET as compared to 62.8% BHET if the reaction was catalyzed by zinc acetate (Troev et al. 2003).

As heavy metals such as lead and zinc are well known for their negative impacts on the environment, alternative milder catalysts are being explored. Mild alkalies such as sodium bicarbonate and sodium carbonate are found to have similar monomer yields as zinc and lead acetate catalysts. Glacial acetic acid, sodium sulfate, lithium hydroxide, and potassium sulfate are also found to have similar yields as commonly used heavy metal catalysts. Zinc chloride was found to produce a BHET yield of 73.24% when used as a catalyst. Though the alternatives discovered are environmentally friendly, there are some drawbacks. Most of the catalysts are ethylene glycol soluble and hence, difficult to be separated from the reaction mixture. This then requires an additional step of distillation to remove the catalysts. The zinc salts are also ineffective when temperature goes above 245 °C (Terzopoulou et al. 2017).

## Heterogeneous Catalysts

Zeolites are explored as possible catalyst for glycolysis reaction due to the presence of mesopores and micropores. These mesopores and micropores provide a large surface area for reaction to occur. However, zeolites do not increase BHET yield as compared to other catalysts (Raheem et al. 2019).

Metal oxides are also explored to be used as catalysts (Imran et al. 2011). The metal oxides are impregnated with different types of silica support such as silica microparticles (SMPs) and silica nanoparticles (SNPs) to be used as catalysts for glycolysis. Four different catalysts—Mn<sub>3</sub>O<sub>4</sub>/SNPs,

Mn<sub>3</sub>O<sub>4</sub>/SMPs, ZnO/SNPs, and ZnO/SMPs—are used in reactions conducted at 11 MPa and 300 °C for 40 to 80 min at a PET to EG ratio of 1:11 and PET weight to catalyst ratio of 100:1. The results showed that Mn<sub>3</sub>O<sub>4</sub>/SNPs produce the higher monomer yield of more than 90%. This could be due to the irregular and porous structure of the catalyst, which provides many active sites for the reaction (Imran et al. 2011). Another type of metal oxide was also explored. Using precipitation, new metal oxide spinel catalysts such as ZnO, Co<sub>3</sub>O<sub>4</sub>, Mn<sub>3</sub>O<sub>4</sub>, ZnMn<sub>2</sub>O<sub>4</sub>, CoMn<sub>2</sub>O<sub>4</sub>, and ZnCo<sub>2</sub>O<sub>4</sub> were formed. Upon testing, it was found that at 260 °C and 5 atm, zinc manganite tetragonal spinel (ZnMn<sub>2</sub>O<sub>4</sub>) as a catalyst produce 92.2 mol% of BHET, the highest amount as compared to the other metal oxide spinel (Bahramian 2020). Using ultrasound, graphene oxide–manganese oxide nanocomposite (GO-Mn<sub>3</sub>O<sub>4</sub>) was produced. Using GO-Mn<sub>3</sub>O<sub>4</sub> as catalysts, it produces a BHET yield of 96.4% when conducted at 300 °C and 1.1 MPa for 80 min. Without graphene oxide (GO), a BHET yield of 82.7% is achieved (Al-Sabagh et al. 2016). Through coprecipitation, Mg–Al hydrotalcite was produced. Mg–Al hydrotalcite of different Mg/Al molar ratios is then used as catalysts for depolymerization of PET. To obtain Mg–Al mixed oxides, hydrotalcites have to calcinated. The Mg–Al mixed oxides display a higher catalytic activity as compared to their hydrotalcite counterparts. At 500 °C, Mg–Al mixed oxides offer the highest catalytic activity (Al-Sabagh et al. 2016).

In recent times, three solid catalysts SO<sub>4</sub><sup>2-</sup>/ZnO, SO<sub>4</sub><sup>2-</sup>/ZnO-TiO<sub>2</sub>, and SO<sub>4</sub><sup>2-</sup>/TiO<sub>2</sub> that have high catalytic activity are explored. Due to their ease of separation through filtration and ability to be recycled, they are preferred over homogeneous acids. High temperature and pressure condition is required for the 3 solid catalysts hence rendering it unsuitable to be used to produce BHET under mild conditions (Zhu et al. 2012).

### Ionic Liquid Catalyzed Glycolysis

Ionic liquids are salts in liquid state that have melting point of 100 °C and lower. They are considered for catalysts due to their thermal stability, electrochemical stability, low flammability, and strong ability to dissolve inorganic and organic compounds. The advantage ionic liquid has as a catalyst over metal acetates is that it is easier to purify the glycolysis products (Shuangjun et al. 2020).

In a study conducted, it is found that 100% PET conversion is achieved at 180 °C after 8 h with the use of 1-butyl-3-methyl-imidazolium bromide ([Bmim][Br]) as a catalyst. The study also found that ([Bmim][Br]) can be reused and requires an activation energy of 232.79 kJ/mol, which makes it possible to replace traditional organic solvents used in PET depolymerization (Wang et al. 2009). In a test conducted with some ionic liquids such as

1-butyl-3-methylimidazolium bicarbonate ([Bmim][Br]) and 1-butyl-3-methylimidazolium chloride ([Bmim]Cl), it is found that 1-butyl-3-methylimidazolium hydroxyl ([Bmim]OH) is the best catalyst for PET glycolysis. The condition required for [Bmim]OH to work well is 190 °C, ratio of mass of PET to EG of 1:10, 0.1 g of [Bmim]OH at 5 wt%, and a duration of 2 h (Wang et al. 2009).

A catalyst that is a better catalyst than [Bmim]Cl is 1-butyl-3-methylimidazolium tetra-chloroferrate ([Bmim]-FeCl<sub>4</sub>). However, the main monomer produced is easily contaminated by iron containing ionic liquids and has to be rinsed by deionized water, causing the loss of monomers and increase in water consumption. With 1 g of 1-butyl-3-methylimidazolium acetate ([Bmim][OAc]) as a catalyst, 20 g of PET and 20 g of EG, it takes 3 h for complete PET glycolysis at 190 °C and the BHET yield is 58.2%. This is due to the anion and cation in Lewis base ionic liquid [Bmim][OAc] producing a synergetic effect that results in the attack on carbon cation of the ester group by the oxygen in ethylene. Since [Bmim][OAc] can be reused up to 6 times, there is potential to use it for industrial level of PET glycolysis. The activation energy required for this reaction is 58.53 kJ/mol (Wang et al. 2010).

1-Butyl-3-methylimidazolium acetate-promoted zinc acetate (Zn(OAc)<sub>2</sub>-[Bmim][OAc]) and 1-butyl-3-methylimidazolium acetate-promoted copper acetate (Cu(OAc)<sub>2</sub>-[Bmim][OAc]) are also tested as catalysts for PET glycolysis. With a catalyst to EG to PET ratio of 1:20:3, complete PET glycolysis is achieved in 3 h at 190 °C. The BHET yield using Zn(OAc)<sub>2</sub>-[Bmim][OAc] and Cu(OAc)<sub>2</sub>-[Bmim][OAc] is 45.6% and 53.95%, respectively. The catalysts can also be reused up to 6 times without losing their catalytic performance. The catalyst works by forming a hydrogen bond between the C<sub>2</sub>-H in imidazolium ring in [Bmim][OAc] and carbonyl oxygen in the metal acetate. The metal acetate then transforms to a unidentate coordination structure from a bidentate one. Meanwhile, metal atom in the metal acetate of the catalyst then coordinate with the carbonyl oxygen on the PET molecule. The coordinated PET molecule is then attacked by the oxygen atom in the hydroxyl in EG. Both reactions are first order and the activation energy for Cu(OAc)<sub>2</sub>-[Bmim][OAc] and Zn(OAc)<sub>2</sub>-[Bmim][OAc] is 56.4 kJ/mol and 53.8 kJ/mol, respectively (Al-Sabagh et al. 2014).

### Upcycling Plastic

By using bioimaging, photocatalysis, biochemical sensing, and using CO<sub>2</sub> emission technology, we can make a barrier from air pollution as a preventive barrier in the following mechanisms.

- PET can be upcycled to porous carbon, which can be used to absorb tetrafluoromethane (CF<sub>4</sub>) (Yuan et al. 2020).
- PET can be upcycled into carbon dots (C-dots), which can be used as a material for sensors (Aji et al. 2018).
- PET fiber can be used to form strain-hardening cementitious composites (Lin et al. 2018).
- Synthetic graphite created from PET waste (Aji et al. 2018).
- PET fibers can be used in textile fabric production due to its properties (Majumdar et al. 2020).
- PET is upcycled to disodium terephthalate to store energy (Ghosh et al. 2020).

### Polyhydroxybutyrate

PHAs, such as PHBs, are defined as the biodegradable polymers generated by various microorganisms in stress conditions, including in the presence of surplus carbon or restricted nitrogen or phosphate (Du and Webb 2011). Polyhydroxybutyrate (PHB), as a natural microbial polyester, is generated by various archaea and bacteria from renewable sources. PHB is similar to some petrochemical plastics. However, it is biodegradable. Identifying appropriate microbial strains and developing processes with direct usage of starch from agricultural wastes without commercial amylase treatment is desirable.

PHB is a plastic that is biodegradable in nature and does not leave any harmful residues in the environment unlike conventional plastics (Sirohi et al. 2020). It is obtained through exposing microorganism (such as *Cupriavidus necator* and *Bacillus megaterium*) (Yousuf and Winterburn 2017) to different carbon sources such as volatile fatty acids, polysaccharides, and polyols. PHB has the potential to replace PET due to their similar mechanical properties but could not due to the cost of producing PHB being higher than PET (Naranjo et al. 2014). Cost of producing PHB is dependent on carbon and nitrogen sources, micro-organism yield and cost from pretreating and conditioning of raw materials, separation, fermentation and purification. Carbon source takes up to 25 to 45% of the production cost of PHB. The filtration and separation process also take up large amount of energy.

PHBs are gathered in various microorganisms as carbon- and energy-storage compounds under conditions with nutrient limitation. They are biodegradable polymers with the potential of replacement of fossil-derived polymers. Numerous studies have been conducted for constructing the PHB generation route in the recombinant *Escherichia coli* (*E. coli*) strains. Researchers have been successful in cloning the primary genes *phbB*, *phbC*, and *phbA* gained from different PHB natural producers, like *C. necator*, in the *E. coli* host. The enzymes PHB synthase, acetoacetyl-CoA reductase, and  $\beta$ -ketothiolase have been expressed functionally

in the recombinants for the conversion of sugar to PHB. As shown by fermentation results, PHB was gathered up to 90% of the dry weight of the cell. In addition to PHB, researchers synthesized co-polyesters, including poly(3HB-co-3HA) and poly(3HB-co-3HV), by mutant *E. coli* strains. There have been also other genetic engineering techniques useful for increasing acetyl-CoA and decreasing NADPH (nicotinamide adenine dinucleotide phosphate) availability in cells for accumulation of PHB (Du and Webb 2011).

### Recycling of PHB

This study focused on the ability of the PHB copolymer to be tested more frequently and to use different viruses to retrieve values. This work studied the effect of recurrence rates and temperature history on PHB copolymer materials. The remake was done for ten generations of retrieval and was a 7 regrind level lessons for virginity items. A 79% reduction in viscosity and a 10% reduction in the last solid strength were observed in the 10th generation of recycling. And a 5% decrease was observed in viscosity and maximum strength with a 50:50 virgin to regrind ratio.

High productivity and controlled conditions are provided by the fermentation process. It also provides uniform product quality and reduces the costs of investment. Despite the higher efficiency of continuous processes concerning production of PHB, they are generally vulnerable to pollution that can result in economic losses. Besides the selection of the process, there are other factors affecting mechanical and thermal characteristics of the PHB structures, including selection of the bacterial strain, fermentation device, parameters of agitation, carbon source, the retrieval approach, and secondary crystallization factors. The crystallinity degree of obtained PHBs and final molecular weights depend on the bacterial producing strain type. The product quality can be improved by development of recombinant strains and using genetic engineering approaches (Du and Webb 2011). Correct selection of carbon source and conditions of cultivation can improve the ultimate product features and minimize operating and capital costs, like energy consumption and substrate price. Economical substrates, like waste streams from various industries, would provide potentially ideal sustainable solutions, but only when the final product meets the quality necessities. The type of the solvent used and the downstream processing conditions could significantly influence thermal characteristics and elasticity of the recovered PHB. PHB properties also be changed by the use of different mechanical or chemical approaches, including obtaining copolymers, blending with other polymers, adding functional groups, inserting additives (nucleating agents, photostabilizers, plasticizers). In order to obtain the desirable features of material related to copolymer production, preparation of blends via compounding and processing



is a more attainable and quicker path. More enhancements provide a promising collection of technologies for the achievement of PHBs with features that suppress equivalent petroleum counterparts and fulfill circularity necessities (McAdam et al. 2020).

When the polymer chain initiates growing and becoming more hydrophobic, a granular structure is formed in the cell. Hydrophilic PHB synthase binds to chain ends, forming a coating surrounding granule, and separates polymer from aqueous cytosol. Then, polymerization goes on by withdrawing substrate from the cytosol and adding to chain through synthase. The growth of chains could be above 10,000 monomers in length. With lengthening the chains, the granules gain a denser packed form and fuse eventually as filling the cell. Until adequate substrate is present, it can be integrated into polymer, and cellular volume puts the limitation on production. Typically, *A. eutrophus* cells possess 8–13 granules, with the diameter of 0.2–0.5  $\mu\text{m}$ . PHB granules in the bacterial cell have a mobile amorphous state that provides simple accessibility for depolymerases. Hence, polymerization happens concomitantly with degradation. After extraction of PHB from the cells, irrevocable alteration to the crystalline happens because of the monomers' absolute stereoregularity (Hankermeyer and Tjeerdema 1999).

Generally, PHA degradation does not generate any destructive by-products intermediates. 3-Hydroxybutyrate is available in all higher animals as a middle metabolite and is found in fairly high amounts in blood plasma of human body. Therefore, PHAs have medical usages, like long-run drug administration, sutures, bone and blood vessel replacement, and surgical pins (Hankermeyer and Tjeerdema 1999).

### Using Lignocellulosic Material

Lignocellulosic waste consists of hemicellulose, lignin, and cellulose. These waste products have the potential to be made into value added products. For treatment of lignocellulosic wastes, enzymes, together with bacteria and fungi, are used to lower the cost and energy required for bioconversion and biodegradation. It is found that the bacteria *Ralstonia eutropha* works well in lignin derivatives. The amount of PHB it produced after 48 h of fermentation in rice paddy straw is 11.42  $\text{g L}^{-1}$ . In reaction with kenaf biomass that undergone various pretreatments, only 10.10  $\text{g L}^{-1}$  of PHB was produced. *E. coli* is found to be able to produce PHB in wastewater microalgae after hydrolysis and obtained PHB amounting to 32% of the dry cell weight of *E. coli*. Other media that are also used to produce PHB are sugarcane bagasse, date seed, and switchgrass. To reduce the cost of PHB, PHB can be produced together with other value added products. In an integrated algae-based biorefinery, biodiesel, PHB, and astaxanthin are produced. The cost of biodiesel was reduced to \$0.48 by selling PHB and

astaxanthin. Deoiled algae biomass residue was also used to produce biobased products. In this process,  $0.43 \pm 0.20$  g of PHB per gram of dry cell weight was produced (Sirohi et al. 2020).

### Using Starch-Based Material

Starch can be found in grains, tuber, and root crops. Before fermenting starch, it is hydrolyzed with enzymes such as amylase and glycoamylase into simple sugars. There are two steps in hydrolysis: liquefaction and saccharification. These two processes convert starch substrates into simple sugars. Using *Halomonas boliviensis*, 26  $\text{g L}^{-1}$  of PHB is obtained from cereal mash through conducting saccharification and fermentation simultaneously. This result obtained was 60% higher than if hydrolysis and fermentation were to be conducted separately. Genetically modified *E. coli* can hydrolyze starch and synthesize PHB at the same time. Starch 2% ( $\text{w v}^{-1}$ ) is also used a primary source of carbon for this process. PHB (1.24  $\text{g L}^{-1}$ ) is produced and it represents 57.4% of cell dry weight. Through testing different bacterial strain with different starch substrate, it is found that *Bacillus aryabhatai* can produce 75% of cell dry weight PHB with different sources of carbon (Sirohi et al. 2020).

### Using Sucrose-Based Material

A by-product produced in sugar production processes is molasses. It is a cheap and rich carbon source. In PHB production, the bacteria *Bacillus megaterium* BA-019 was used with sugarcane molasses and urea. Sugarcane molasses is a carbon source and urea is a nitrogen source. *Ralstonia eutropha* ATCC was found to produce biopolymer that is 97.8% of the biomass produced when added to a mixture of residual vinasse and sugarcane molasses. In the reaction between *C. necator* and sugarcane molasses and vinasse, vinasse was used as a source of water at the beginning of the process and molasses during the production stage. Maximum PHB produced was 11.70  $\text{g L}^{-1}$  with PHB accumulation of 56%. Nitrogen was found to raise the concentration of biomass, making it effective for PHB production. Desugared sugarbeet molasses mixed with *Bacillus megaterium* uyunni S29 can produce PHB with concentration of 0.6  $\text{g g}^{-1}$  and 16.7  $\text{g L}^{-1}$  cell dry weight after 24 h. The advantage of desugared sugar beet molasses is the ability to increase biomass production by 2 to 3 times as compared to mineral-based mediums (Sirohi et al. 2020).

### Using Dairy Industrial Waste

Whey from cheese factories can be used as a carbon source for PHB production. *Azohydromonas australica* DSM 1124 can depend on lactose sugar as carbon source to product

PHB. In 72 h,  $2.07 \pm 0.2 \text{ g L}^{-1}$  of PHB is produced at 2% (w v<sup>-1</sup>) lactose concentration. Other bacteria that can also produce PHB from dairy waste are *Bacillus subtilis*, *A. australica*, and *R. eutropha*. The combination of *Bacillus subtilis* EPAH18 and whey produce  $0.54 \text{ g L}^{-1}$  of PHB. The process is most efficient at 1000 t/h of whey feed (Sirohi et al. 2020).

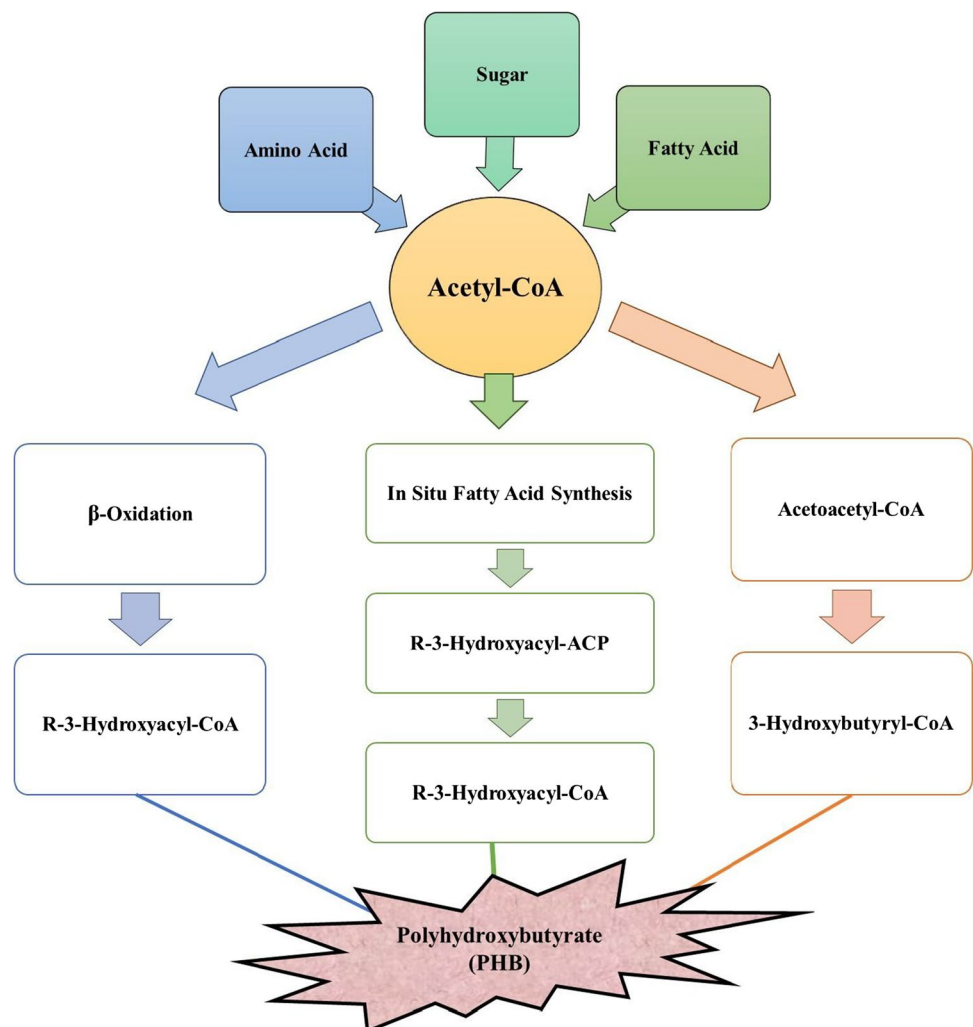
### Using Oil, Fatty Acids, Glycerol-Based Waste Materials

Crude glycerol and squander cooking oil are both classified as waste materials. Crude glycerol is derived as a by-product from biodiesel production. Squander cooking oil consists of plant, animal, and synthetic fat that are left over from cooking. *Bacillus megaterium* and glycerol reagent grade (GRG) will produce  $2.8 \text{ g L}^{-1}$  of PHB. The bacteria *Burkholderia thailandensis*, *Bacillus* sp., and *C. necator* are used with palm cooking oil, cooking oil, and glycerol to produce PHB effectively (Sirohi et al. 2020).

### Using Agro-industrial Waste and By-products

Food and agriculture industries produce plenty of by-products that are rich in carbon and have the potential to be used for value production. Some agro-industrial wastes include pretreated sugarcane tops, sugarcane bagasse, fruit peels, banana residues, deoiled cakes, and algal biomass. Sesame oil cake and ragi husk were used with *Bacillus megaterium* to produce  $0.61 \text{ g L}^{-1}$  of PHB. Used coffee ground was used with *Burkholderia capacia* to produce PHB. *Azohydromonas lata* DSM 1123 is mixed with by-products from the cane sugar industry to produce  $16.9 \text{ g L}^{-1}$  of PHB with 83.9% of cell dry weight. *Bacillus* sp. is used with sugarcane bagasse to produce PHB, which removes the problem of disposing the waste material. Wastewater from brewery can also be used to produce PHB, and the increase in concentration of activated sludge will increase the production rate by 39% (Hankermeyer and Tjeerdema 1999) (Fig. 10).

Fig. 10 Types of PHB production methods



## Recycling/Upcycling PHB

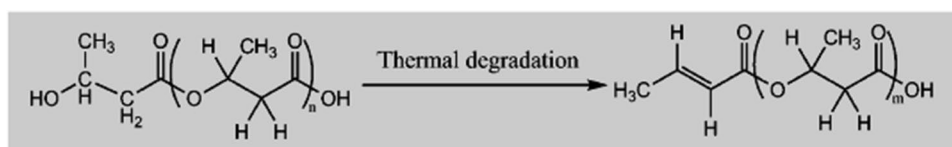
Instead of recycling PHB, PHB can be converted to biobased chemicals to create more value since cost of producing PHB is relatively higher than PET. PHB can be converted to methyl crotonate (MC) through a 2-step process. The first step thermolyze PHB into crotonic acid and the second step produce MC through esterification. The process is conducted at 200 °C and 18 bar, and 60% of MC is produced (Yousuf and Winterburn 2017). Plastic waste recycling is critically significant for having a sustainable environment since it creates novel sources for production of materials and decreases environmental contamination. The aim of polymer chemical recycling is fuel generation, development of novel functional chemical materials, and reproducing the original polymer by de-polymerizing and then repolymerizing to a material equivalent to the original one. It is possible to biologically recycled biodegradable polymers. However, energy-effective recycling to novel materials is a more economically and environmentally beneficial approach. PHAs are biodegradable and biobased polyesters generated as energy and carbon reservoirs in different microorganisms in the conditions when nutritional elements are limited and there are excessive carbon resources (Yousuf and Winterburn 2017). Earlier, PHAs were recycled chemically via thermal degradation or pyrolyzing to CA (crotonic acid) and oligomers with one carboxylic and one unsaturated acid chain end as well as cyclic oligomers. The general range of degradation temperatures was 170–300 °C. For instance, PHB was transformed to crotonic acid through catalytic thermal degradation in  $Mg(OH)_2$  or  $MgO$  presence at 240–280 °C. 12 3-Hydroxybutanoic acid, the possible monomer for synthesizing new PHB via a chemical path, is not generated in thermal degradation. It could be generated via PHB enzymatic hydrolysis. The usage of microwave energy has shown superiority compared to the traditional heating approach to accelerate chemical processes that lead to quick transformations, milder reaction conditions, and great yields. Various studies have used reactions assisted by microwave for chemical recycling of biomass and polymers. In recent years, microwave irradiation has been shown to an efficient method for generation of PHB oligomers in high yield via thermal degradation, leading to oligomers with different molecular weights and unsaturated chain ends. The temperatures used for the thermal degradation assisted by microwave has been at a range of 180 to 220 °C, with the degradation rate

as almost 100 times quicker compared to traditional thermal heating at the identical temperature (Yang et al. 2014). The focus of former works on PHB chemical recycling has been mostly on thermal recycling to unsaturated and CA oligomers. Although thermal pyrolysis has the capability of successful degradation of PHB to unsaturated oligomers and monomers, it is more favorable to recycle PHB at lower temperatures to monomers repolymerized to the equivalent degradable products. Our purpose is utilizing reactions assisted by microwave in water, green solvents, ethanol, and methanol with alkaline settings for fast hydrolysis of PHB at a lower temperature to functional monomers with hydroxyl and carboxyl or ethoxy and methoxy end groups rather than unsaturated end groups. Therefore, it is possible to exploit these monomers directly for synthesizing new PHB oligomers (Yang et al. 2014; Meerboer et al. 2020).

PHB can be obtained from renewable resources. However, the expensive production process is currently the major problem for the typical usage of this material. With growing the manufacturing market of PHB, more emerging and novel fermentation procedures would be applied in industrial situations. Continuous improvement of bio-engineering processes can create PHBs with high performance and efficient high yield, even from waste stream sources, which reduces expenses and deliver cost-effective PHBs with the qualified mechanical characteristics for broader market opportunities and new applications (McAdam et al. 2020). Degradation of PHB, PBL, and PLA in a controlled manner using different approaches, like thermal degradation, enzymatic and steam hydrolysis, etc., have been addressed in prior studies. The final products of degradation considerably depend on the employed approaches. As an example, PHB presents different degradation products, including monomers, like CA from thermal degradation and 3-hydroxybutyric acid from enzymatic hydrolysis; and cyclic oligomers, cyclic trimers, and linear oligomers from ring-closing depolymerization (Ariffin et al. 2010).

PLA and PHB have the possibility of biological recycling. However, recycling the materials would reduce desirable accessibility to useful intermediates and functional chemicals, resulting in more economical and eco-friendly solutions. With thermal degradation, PHB can be degraded to CA and oligomers with one carboxylic acid and one unsaturated chain end (Yang et al. 2015) (Fig. 11).

**Fig. 11** Thermal degradation of PHB to oligomers (Yang et al. 2015)



## Future Directions

The reason why recycling is so important is that it prevents land pollution, reduces the need to harvest new materials, saves energy, reduces emissions, saves money, reduces the amount of waste that ends up in garbage dumps, and allows for full product use. It prevents pollution by reducing the need to collect new raw materials. Increases economic security by tapping the source of household goods. By converting bottles, packaging and other plastic waste into new goods, recycling helps the environment and opens up new economic opportunities. Reuse of plastics keeps recyclables off-ground and encourages businesses to develop new and innovative products. The environmental effects of solid plastic waste are evident in the ever-increasing levels of plastic, land and sea pollution. But while there are significant economic and environmental incentives for recycling of plastics, alternative waste disposal treatment options for plastic waste are most effective. Recycling plastics before recycling is expensive and time consuming, recycling requires a lot of energy and often results in low-grade polymers, and current technology cannot be applied to many polymeric materials. Recent research points the way to chemical reuse methods for low energy requirements, compounding of plastic waste disposal to avoid the need for filtration, and increasing the recycling technology of non-recyclable polymers.

## Conclusion

Polymer plastics are synthetic, mainly made of petrochemical feedstock. Since the advent of the first plastic in the early 1900s, the number of plastics has increased dramatically, as has their use. As society has gradually increased its use of plastics, waste management in plastic has become a growing phenomenon around the world. Today, about 280 Mt plastics are produced annually. However, recycling of plastic is still limited compared to many other items in bulk. The rate of plastic reuse in the EU today is about 25%. The rate of discovery and recycling of plastics is growing in many countries around the world, and the international market for used plastics continues. Continuing to increase reuse, the industry faces many challenges. The biggest challenge is the quality of the findings and recycling. Plastics are easy to form, do not corrode or rot only slightly and characteristics can be modified to suit specific needs by using compounds or adding certain layer additives. These features are also a source of some of the problems associated with plastics. From the incorporation of chloro-containing plastic in the form of dioxin

in garbage in cinerators to plastic soups, the durability and widespread use of plastic play a significant role in important waste management problems. Reuse of plastics is one of the key ways to reduce the environmental problems associated with plastic.

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