

REVIEW

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Poly (ethylene terephthalate) recycling for high value added textiles

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Abstract

This study reviews the problems in the use and disposal of poly (ethylene terephthalate) (PET) and includes the concise background of virgin and recycled PET as well as their possible applications. The current state of knowledge with respect to PET recycling method is presented. Recycling of PET is the most desirable method for waste management, providing an opportunity for reductions in oil usage, carbon dioxide emissions and PET waste requiring disposal because of its non-degradability. Advanced technologies and systems for reducing contamination, mechanical and chemical recycling, and their applications are discussed, and the possibility of diverting the majority of PET waste from landfills or incineration to recycling is suggested.

Keywords: Polyethylene terephthalate; Mechanical recycle; Chemical recycle

Introduction

Poly (ethylene terephthalate) (PET), commonly referred to as 'polyester' in the textile industry, is considered to be one of the most important thermoplastic polyesters (Incornato et al. 2000). It is widely used for various applications such as bottles, fibers, moldings, and sheets because of its excellent tensile and impact strength, clarity, processability, chemical resistance, and thermal stability (Pawlak et al. 2000; Kong and Hay 2003; Avila-Orta et al. 2003). The PET fiber patented originally by DuPont (DuPont, 1997) dominates over 50% of the world synthetic fiber market. In the late 1950s, PET film was developed and used for X-ray, photographic, and cassette recording films as well as flexible packing materials. Blow molding techniques began to be used to stretch PET, which produced three dimensional structures, resulting in the rapid growth of light and unbreakable PET bottle manufacturers in the early 1970s (Wyeth and Roseveare 1973).

These developments in the PET manufacturing process resulted in a diversity of industrial applications, leading to increased consumer demand for PET, and many global companies produced PET with different trade names (Carraher 2000). The total world production capacity of PET amounted to 64,400 kilotons per year (kt/year) in 2008. Polyester fibers and bottles accounted for most of the total world production, representing 63.5% and 30.3%, respectively, while the production of polyester film and engineering resin accounted for 6.2% (Cischem. Com Co., Ltd. Publication 2010). PET goods such as fibers and bottles have become a major part of human life, and their production and consumption have increased continuously.

PET, unlike natural polymers, is a non-degradable polymer in the natural environment (Edge et al. 1991), leading to environmental pollution when it is discarded after use. Procedures to enable biological degradation of PET are both complicated and expensive (PACIA 2002). Therefore, PET waste management with upcycling concept has become an important social issue because of PET's increasing consumption and non-biodegradability. The upcycling concept by recycling is shown in Figure 1. Extensive utilization of PET results in waste management difficulties. Landfill of PET is undesirable because of space limitations and ground pollution. Incineration is also not the preferred option, because of the inevitable emission of toxic gases that are generated from the decomposition of PET molecular chains and residual additives, although incineration produces energy.

Because of this problem, recycling of PET is considered to be the best upcycling way to manage PET waste economically (Farahat et al. 2000). The recycling of PET is also an important concept with regard to the awareness of environmental concerns and conservation of scarce energy and feedstocks, which is related to national economy. The cost effective technology for PET recycling should be developed because virgin PET has a low and steady price.

Since the first recycling of PET in 1977 (Miller 2002), many studies have been conducted to investigate PET recycling methods (Helwani et al. 2009; Patterson 2007; Yoshioko et al. 1994, 2001), and the recovery rate of consumed PET has been consistently increasing. The world consumption of PET packaging is expected to about 19.1 million tons by 2017, with 5.2% increase per year between 2012 and 2017 (Smithers Pira 2012). The majority of the recycled PET was converted into fibers as shown in Figure 2 and based on the report by Noone (Noone 2008). There are two kinds of PET fibers, partially oriented yarn (POY) and staple fibers. They are utilized for various end-use applications with different material properties. Therefore, the recycled PET staple fibers and POY are

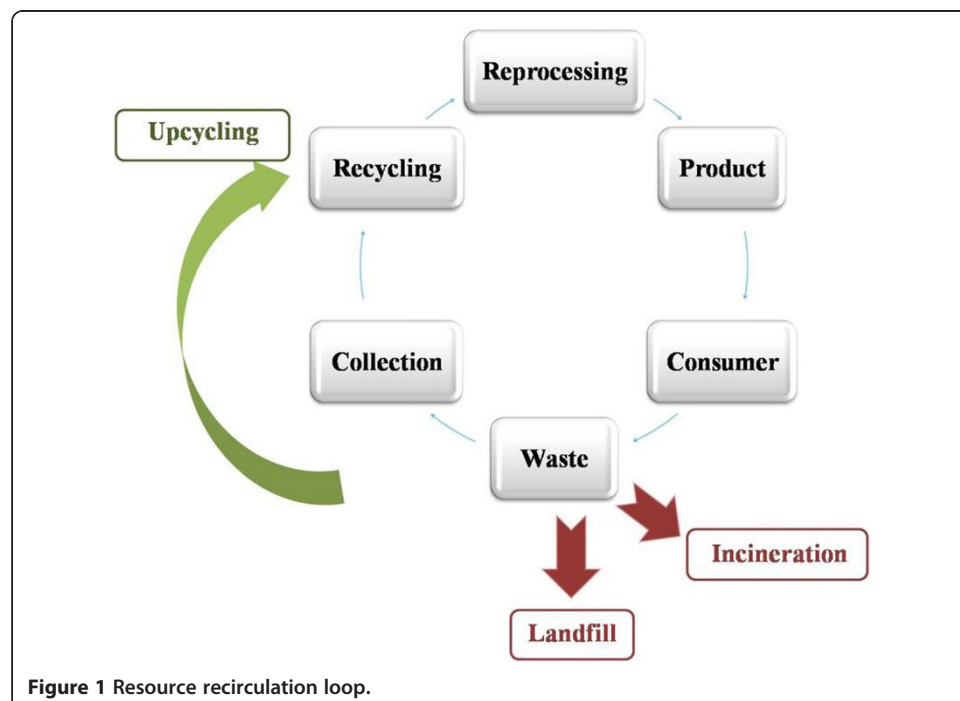
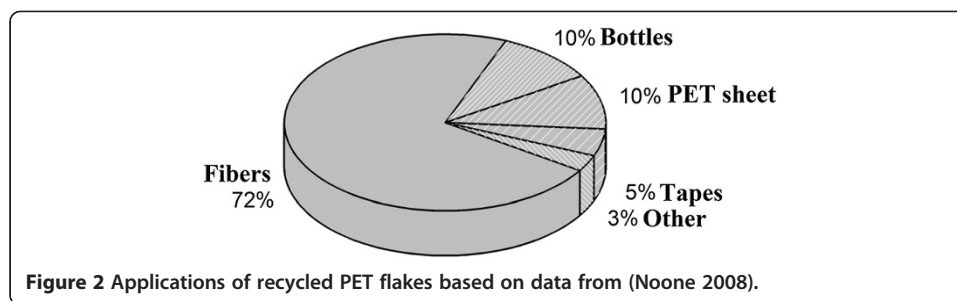


Figure 1 Resource recirculation loop.



very important because they play a crucial role as intermediate products in the nonwoven and textile industries.

PET waste can be recycled in many ways including chemical and mechanical methods. Hopewell (Hopewell et al. 2009) has summarized various terminologies used for plastic recycling. The primary recycling method is re-extrusion, the classical method of recycling PET. This method involves recycling of scrap materials to form original products. However, because various polymers and other materials such as paper, pigments, metals, and adhesives are used as plastic packing materials, this makes recycling by this method difficult. Although this method has the advantages of simplicity and low cost, it is not a popular method that requires uncontaminated scrap with only a single type of waste.

The secondary recycling is the mechanical recycling method, commercialized in the 1970s. This method is carried out by reprocessing PET waste to granules after separation of the polymer from contaminants. It is also called 'materials recycling,' which includes sorting and separation of waste, washing for removal of dirt and contaminants, grinding and crushing, and reprocessing process (Aguada and Serrano 1999). The heterogeneity of the PET waste is the main issue for mechanical recycling because the complexity and contamination of the PET waste makes it difficult to recycle mechanically. The decline in the product quality is also the main disadvantage of mechanical recycling because the heat of fusion causes photo-oxidation, and mechanical stress results from the inverse reaction. Therefore, this recycling method is not used for making products that require high quality standards.

The third recycling, also called chemical recycling, has great potential because of the severe limitations of the mechanical recycling of PET wastes. Chemical recycling enables recovery of the petrochemical constituents of PET wastes, which can be utilized for remanufacturing PET products or other synthetic chemicals. It has not been cost effective to use this method because of the lower price of petrochemical feedstocks compared with the process prices for production of PET monomers or oligomers from PET waste (Patel et al. 2000). Chemical recycling has the advantage of reversing the energy-intensive polymerization performed during the original manufacture of PET, which is important in terms of life-cycle assessment. PET wastes can be depolymerized by hydrolysis, methanolysis and glycolysis (Chen et al. 2001a; Kosmidis et al. 2001; Yamaye et al. 2002; Goje and Mishra 2003), and converted into PET resin or other unsaturated polyesters (Sinha et al. 2010), providing raw materials for a large variety of products.

The fourth recycling is the energy recovery method, which involves incineration of the PET waste. PET waste management by incineration generates thermal energy by recovering the chemical energy stored in PET wastes. It is considered to be an undesirable

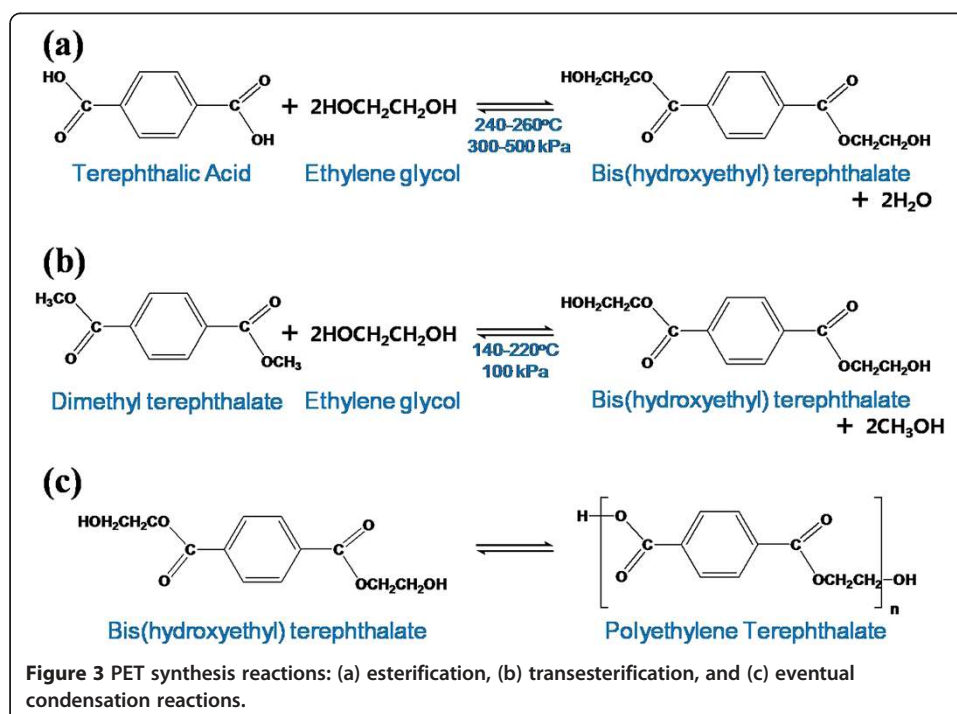
method because it causes air pollution and health risks from toxic gases generated during the incineration of PET wastes. Therefore, the discussion in this study focuses only on the mechanical and chemical recycling methods relevant to the management for PET wastes.

In this research, the current recycling systems and technologies for PET waste management are reviewed, and the manufacturing process for preparation of recycled PET fibers is discussed in terms of a sustainable textile industry.

Poly (ethylene terephthalate) (PET)

PET has been one of the most widely used engineering polymers during the past two decades because of its lightness, cheap and readily available feedstock, and low energy requirements for processing and fabrication. The two PET grades (fiber-grade PET and bottle-grade PET) are dominant in the global market. These two PET grades have different molecular weight or intrinsic viscosity (IV), optical properties and production recipes, which differ mainly in quantity and type of comonomers, stabilizers, metal catalyst, and colorants (Rieckmann and Volker 2003).

PET is produced by step-growth polycondensation from terephthalic acid (TPA) and ethylene glycol (EG), and the synthesis of PET involves two different starting reactions. One is the esterification reaction in which TPA is reacted with EG, forming bis (hydroxyethyl) terephthalate (BHET) as a prepolymer. The other is the transesterification reaction in which dimethyl terephthalate (DMT) is reacted (Ravindranath and Mashelkar 1986). These two different starting reactions are shown in Figure 3. The transesterification process was much preferred in the early years of PET production because of the easy purification of DMT by distillation. However, the majority of processes for PET production have shifted from DMT to TPA as the feedstock since highly purified TPA has been obtained for the first time at an industrial scale by recrystallization in the late



1960s. The PET, up to this stage, is appropriate for applications such as fibers and recording tapes for which high molecular weight or viscosity is not required. PET with high molecular weight and viscosity for bottles or technical yarns is produced by further polycondensation through the solid-state polymerization process.

Processed PET has been used in a wide range of applications because of its low cost (Thompson et al. 2009), remarkable tensile strength, chemical resistance, clarity, and reasonable thermal stability (Caldicott 1999). More than 60% of the PET produced worldwide is applied in the textile industry. Other applications include recording tapes, X-ray films, and thermoformed products such as housewares, automobiles, lighting products, sporting goods, and food packaging (Carraher 2000; ILSI Europe Report Series 2000). In the food packaging industry, PET has mainly been utilized for beverages because of its glass-like transparency and adequate gas barrier properties. Its toughness also leads to lightweight and unbreakable containers with large capacity (Welle 2011).

This widespread application of PET results in the unavoidable creation of large amounts of PET waste, creating serious difficulties in maintaining a clean environment. With increasing environmental awareness in our society, the recycle method is the most viable option for PET waste management. The recycling of PET waste can be carried out in many ways as described in the next chapter.

Recycled PET

The extensive use of PET products leads to waste management difficulties. PET products have a slow decomposition rate (Edge et al. 1991), creating ongoing environmental pollution problems after use. There are many studies describing methods for successful PET recycling, and the minimum requirements that recycled PET flakes should meet are reported in Table 1 (Miller 2002; Awaja and Pavel 2005; Pawlak et al. 2000). Recycled PET should fulfill the following quality objectives for suitability in high-value applications. The quality objectives are color separation, minimum contamination, intrinsic viscosity close to the original values, and consistent batch-to-batch quality.

Contamination

Contamination of postconsumer PET is the major factor that contributes to the deterioration of its physical and chemical properties during reprocessing (Giannotta et al. 1994).

Table 1 Minimum requirements for PET flakes to be reprocessed (Awaja and Pavel 2005; Miller 2002; Pawlak et al. 2000)

Properties	Values
Intrinsic viscosity	> 0.7 (dlg ⁻¹)
Melting temperature	> 240 (°C)
Water content	< 0.02 wt%
Flake size	0.4 mm < suitable value < 8 mm
Dye content	< 10 ppm
Yellow index	< 20
Metal content	< 3 ppm
Polyvinyl chloride content	< 50 ppm
Polyolefin content	< 10 ppm

In general, contamination cannot be admitted for fiber or bottle applications because of fiber breakage and aesthetic problems. Minimization of contamination leads to better quality of recycled PET (Torres et al. 2000).

Acids contamination

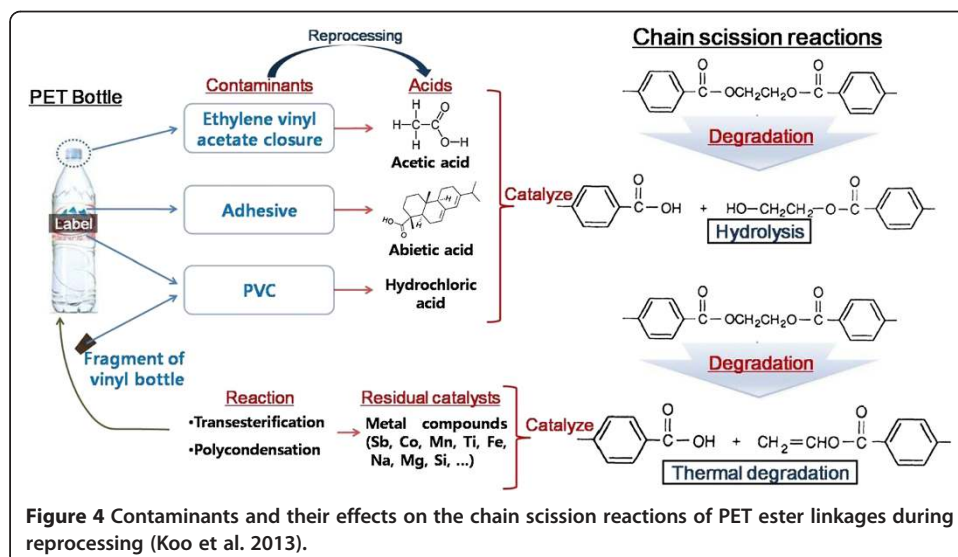
The chain scission reactions of the ester linkages of PET are catalyzed by acidic compounds, generated from contaminants at the high temperatures used during extrusion in the reprocessing. Typical contaminants such as PVC, glue, dirt, ethylene vinyl acetate (EVA), and paper, and the acidic compounds that catalyze the hydrolysis of the PET ester linkages, are shown in Figure 4 (Koo et al. 2013). The most harmful acidic compounds are acetic acid (produced by EVA from the cap liners), rosin acid and abietic acid ($C_{19}H_{29}COOH$; produced by adhesives from the label), and hydrochloric acid (produced by PVC flakes from PVC bottles). These acids play a role as catalysts for PET chain scission reactions during reprocessing (Scheirs 1998; Cardi et al. 1993; Paci and La Mantia 1998).

Moisture

Moisture contamination must be below 0.02% to prevent molecular weight reduction by hydrolysis (Scheirs 1998). Seo and Cloyd (Seo and Cloyd 1991) reported that there were two degradation stages by moisture contamination during reprocessing. The first stage of degradation was attributed to hydrolysis from residual water in the recycled PET, while the second stage was attributed to thermooxidative chain scission with a lower degradation rate than the first stage. The conventional solution is to dry the PET before reprocessing by using special driers such as the Sikoplast™ T800 drier. Commercial driers are available for successful PET drying at 150°C for 4 hours (Scheirs 1998).

Labels and adhesives

Ultrathin polyethylene (PE) films are used for PET labels without any adhesive assistance, and they are removed by flotation during the separation process. The separation



of PE films becomes difficult when heavy overprinting is applied, resulting in an increase in the label density.

Three types of adhesives (Scheirs 1998) are utilized for unmodified labels instead of water-soluble adhesives such as dextrin, starch, and casein glues, which have poor adhesion to PET plastics. The first type is synthetic glues based on polyvinyl acetate or ethylene vinyl acetate, and these glues are broken down in water during the recycling process. Some glue may be retained in the PET and be incorporated into the recycled resin. The second type is thermal adhesives based on ethylene vinyl acetate, which are completely uninfluenced by the washing process. These glues can soften during shredding or the label removal process because of their low T_m , resulting in glue residue being attached to the PET. The last type is alkali-soluble glue. When it is exposed to NaOH solution (2 wt%), this glue crumbles and separates from the PET. This glue is the preferred option except for its higher cost compared with traditional adhesives.

Coloring contaminations

Dyes for color bottles cause undesirable coloring during reprocessing, and printed ink labels can also discolor PET flakes in the washing process. Progress in sorting and washing techniques may reduce this color contamination in bottle recycling (Awaja and Pavel 2005).

Metal interactions

In general, recycled PET includes various metal catalyst residues that can be used for the PET manufacturing process, as summarized in Table 2 (Richard et al. 1992). These metal ions, which promote transesterification and polycondensation reactions, lead to chemical heterogeneity of the recycled PET that affects the melt rheological behavior.

Acetaldehyde

Acetaldehyde is the major by-product of PET degradation reactions. The reaction by the recombination of vinyl ester and hydroxyl endgroups restores the molecular weight of the PET; however, this reaction also unfortunately produces vinyl alcohol, which forms acetaldehyde by tautomerization. This acetaldehyde can migrate into the final product, which is a major concern for food applications. Fortunately, acetaldehydes are highly volatile, so their production can be minimized by processing under vacuum or drying. Another method for the minimization of acetaldehyde production was reported by Villain et. al. (Villain et al. 1995). Various stabilizers such as 4-aminobenzoic acid,

Table 2 Various metal ions in recycled PET and their origins (Richard et al. 1992)

Metal ions	Concentration (ppm)	Origin
Sb	220-240	Polycondensation catalyst
Co	50-100	Polycondensation catalyst
Mn	20-60	Transesterification catalyst
Ti	0-80	Polycondensation catalyst
Fe	0-6	Incorporated during washing
Na, Mg, Si	-	Food additives

diphenylamine and 4,5-dihydroxybenzoic acid have been incorporated into PET to minimize the amount of acetaldehyde formation.

Other contaminants

People use PET bottles for storing dangerous materials such as detergents, fuel, and pesticides, which can be a hazard to public health if these materials remain after PET recycling (Demertzis et al. 1997). An increase in public awareness about the dangers of storing these materials in PET bottles is required for application of recycled PET.

Mechanical recycling

Mechanical recycling, also known as material recycling, involves many treatments and operations. The steps in mechanical recycling include separation of waste, washing to remove dirt and contaminants, crushing and grinding to decrease PET particle size, reextrusion, and reprocessing for production of new PET goods (Babinchak 1991). It is possible to recycle thermoplastic PET mechanically, while it is impossible to recycle thermoset PET because it cannot be remolded by heat. It is difficult to recycle contaminated PET waste mechanically because of the complexity of the waste stream. The major issues for mechanical recycling are the heterogeneity of the PET waste and deterioration of the product properties each time it is recycled. Small amounts of another polymer incorporated in the PET matrix may significantly change the properties of PET, disturbing its possible use in conventional applications. In addition, PET, like most polymers, is degraded during use because of various factors such as temperature, ultraviolet radiation, oxygen, ozone, and mechanical stresses, leading to altered properties and reduced performance of recycled PET compared with virgin PET. Another difficulty with recycled PET is an undesirable grey color, resulting from the presence of PET waste made from the same resin (Bartolome et al. 2012). High-quality recycled PET is achieved when successful separation is conducted prior to the remolding step.

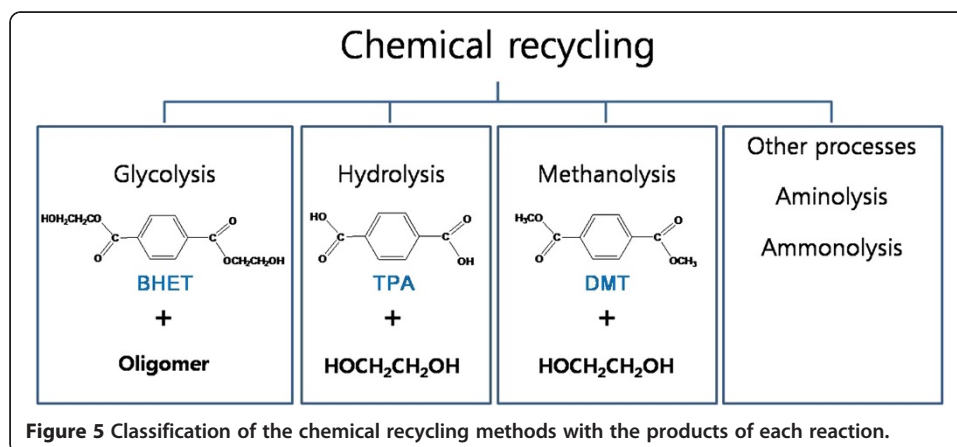
PET flakes can be reprocessed into fibers by the melt extrusion system. Currently, there are two methods for the production of recycled PET fibers from mechanical recycling. One is the more common method where PET flakes are extruded directly into fibers, and the other is the conversion method where PET flakes are reprocessed into granules or pellets and then melt extruded into fibers. Compared with chemical recycling, the mechanical recycling of PET has many advantages, such as the relative simplicity of the process, low investment cost, utilization of readily available equipment, flexibility of feedstock supply, and little negative environmental effect. However, mechanical recycling can degrade the printability or dyeability of the final product because of the cyclic and linear oligomers generated (Dulio et al. 1995). The yellowing of mechanically recycled PET is also a significant problem because of the intramolecular cross-linking and oxidation reactions (Edge et al. 1996). Above all, the major disadvantage of mechanical recycling is the reduction of the molecular weight or intrinsic viscosity, by thermal and hydrolytic degradation. Both the academic and industrial fields have explored methods for maintaining the molecular weight or intrinsic viscosity during the mechanical recycling process of post-consumer PET.

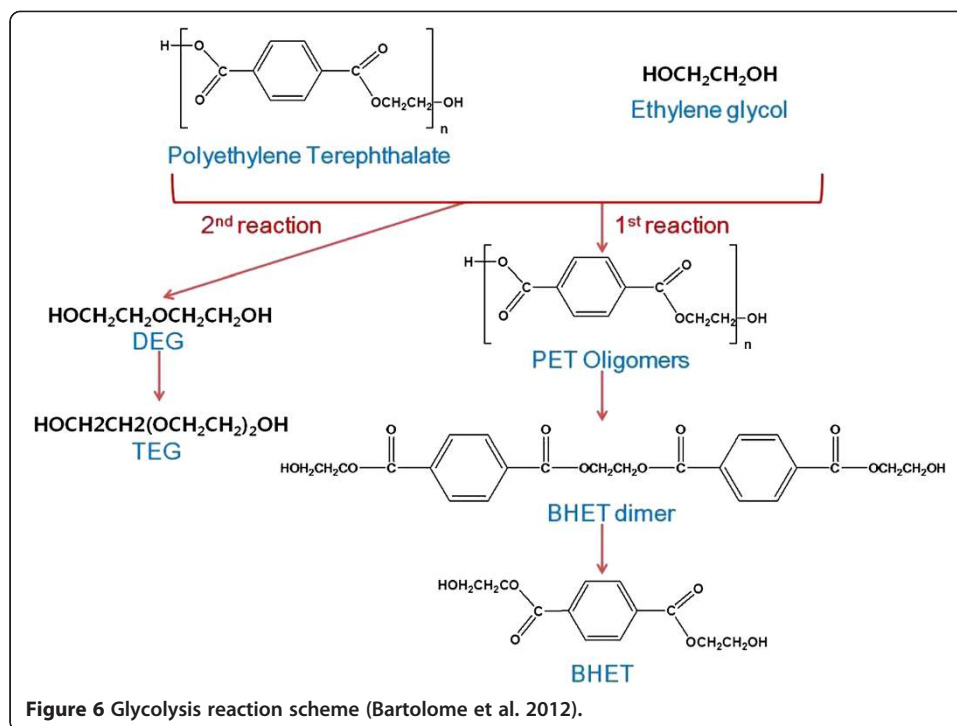
Chemical recycling

Chemical recycling is accompanied by transformation of the PET chain. By means of polymer chain scission, the PET polymer can be broken down into either its monomers or its oligomers and other chemicals (Chen et al. 2001b; Kosmidis et al. 2001; Yamaye et al. 2002; Goje and Mishra 2003; Kao et al. 1998; Carta et al. 2003). It is more cost consuming than the mechanical recycling, and there is no economic incentive because of the higher manufacturing cost of chemically recycled PET compared with virgin PET. Chemical recycling needs to be implemented at a large scale to become economically feasible. Compared with other recycling methods, chemical recycling is the only method that conforms to 'sustainability' principles (Bartolome et al. 2012), because it produces original raw materials. In addition, extra resources for producing PET are unnecessary in the chemical recycling of PET (Achilias and Karayannidis 2004). The commercially available chemical recycling technologies contain glycolysis (Ikladios 2000; Chen et al. 2001b; Pardal and Tersac 2007; Kosmidis et al. 2001), hydrolysis (Carta et al. 2003; Goto et al. 2002; Kurokawa et al. 2003; Genta et al. 2007), methanolysis (Goto et al. 2002; Kurokawa et al. 2003; Genta et al. 2007), and aminolysis (Spychaj et al. 2001; Shukla and Harad 2006) reactions. The classification of chemical recycling with each reaction product is shown in Figure 5. These chemical recycling technologies are discussed in detail below.

Glycolysis

Since the chemical recycling of PET by glycolysis was first developed by McDowell et al. in 1965 (McDowell and Klusio 1965), this process has been widely utilized on a commercial scale. The PET polymer is decomposed through the glycolysis reaction by glycol in the presence of transesterification catalysts. This reaction mainly yields BHET for polyester condensation, and the glycolysis reaction scheme is shown in Figure 6 (Bartolome et al. 2012). Numerous studies have investigated glycolysis of PET because of the advantages of this process, including simplicity, flexibility, and low capital costs. Another advantage is that the glycolysis process can be easily applied to conventional PET production plants (Scheirs 1998). The recovered BHET can be incorporated into virgin BHET, and this mixture can be utilized for two different PET production processes (DMT-or TPA-based). Chemical recycling by glycolysis uses ethylene glycol (Baliga and Wong 1989; Chen et al.





1991), diethylene glycol (Karayannidis et al. 2006; Vaidya and Nadkarni 1987a, 1987b), propylene glycol (Vaidya and Nadkarni 1987a, 1987b, 1987c; Guclu et al. 1998), or triethylene glycol (Ozturk and Guclu 2005). Glycolysis cannot produce clear virgin PET because it is a partial depolymerization to the intermediate BHET, and colorants or dyes are not removed. Glycolysis yields not only BHET but also significant amounts of other oligomers (Scheirs 1998), and this makes it difficult to apply for synthesis of specific desired products. Furthermore, it is known from studies on PET glycolysis kinetics that the glycolysis rate is very slow without a catalyst, and complete depolymerization of PET to BHET is impossible (Campanelli et al. 1994a; Chen and Chen 1999). Therefore, many researchers have investigated methods for enhancing the glycolysis rate and BHET monomer yield by application of efficient catalysts and optimization of the reaction conditions. Another approach is the development of products made from glycolysis without the separation oligomer (Grzebierek and Wesolowski 2004). Since the beginning of glycolysis research, the BHET monomer yield and reaction time have improved continuously from just 65% yield over 8 hours reaction time to at least 90% with the remarkably reduced reaction time of about 30 minutes (Bartolome et al. 2012).

The most intensively studied method for increasing glycolysis rate is transesterification catalysis. The metal-based catalyst promotes the glycolysis reaction mechanisms (Shukla and Harad 2005; Pingale et al. 2010). The free electron pair of the ethylene glycol attacks the carbonyl carbon of the PET ester group, and then the covalent bonding between the hydroxyethyl group of ethylene glycol and the carbonyl carbon of PET is formed, which leads to breakage of the long chain into short chain oligomers and creation of BHET. The glycolysis rate and BHET monomer yield are dependent on various reaction parameters such as temperature, PET/ethylene glycol ratio, and type and

amount of catalysts. A number of studies are summarized in Table 3, detailing the optimum glycolysis conditions and reaction parameters obtained (Dulio et al. 1995; Pingale et al. 2010; Xi et al. 2005; Troev et al. 2003; Shukla and Kulkarni 2002; Shukla et al. 2008; Imran et al. 2011; Wang et al. 2009; Yue et al. 2011).

Hydrolysis

The next chemical recycling method is hydrolysis, which produces TPA and ethylene glycol under high-pressure (1.4-2 MPa) and high-temperature (200-250°C) conditions with a long depolymerization time. In general, hydrolysis is not used commercially in food-grade production, because of the cost associated with purification of the recycled TPA (Sinha et al. 2010). The hydrolysis of PET can be performed as acid hydrolysis, alkaline hydrolysis, and neutral hydrolysis.

Acid hydrolysis is carried out by using sulfuric or nitric acid to yield TPA. This process becomes very costly because of the necessity of recycling large quantities of concentrated sulfuric acid and purifying ethylene glycol containing sulfuric acid. Yoshioka (Yoshioko et al. 1994) reported an effective hydrolysis process using 7 M sulfuric acid, resulting in good yields of TPA and ethylene glycol. The hydrolysis reaction using nitric acid also produces TPA and ethylene glycol, and the ethylene glycol is simultaneously oxidized to oxalic acid, which is more expensive than TPA and ethylene glycol.

Table 3 Various reaction parameters for PET glycolysis ((a) Pingale, et al. 2010; (b) Xi, et al. 2005; (c) Troev et al. 2003; (d) Shukla and Kulkarni 2002; (e) Shukla et al. 2008; (f) Imran, et al. 2011; (g) Wang et al. 2009; (h) Yue et al. 2011)

Catalysts	BHET yield (%)	Temp. (°C)	Time (min)	Ethylene glycol /PET mol ratio	PET/Catalyst weight ratio	Refs
Lithium chloride	59.46	197	480	10	0.005	(a)
Zinc chloride	73.24					
Didymium chloride	71.01					
Magnesium chloride	55.67					
Ferric chloride	56.28					
Zinc acetate	85.6	196	180	5 (w/w)	0.01	(b)
Zinc acetate	62.8	200	150	2.77	0.003	(c)
Titanium phosphate	97.5					
Zinc acetate	62.51	190	480	6	0.005	(d)
Lead acetate	61.65					
Sodium carbonate	61.5					
Sodium bicarbonate	61.94					
β-zeolite	66	196	480	6	0.01	(e)
γ-zeolite	65					
Zinc oxide on silica	~85	300	80	11	0.01	(f)
Magnesium oxide on silica nanoparticle	>90					
Ionic liquids	-	190	120	10 (w/w)	0.05	(g)
[bmim] OH	71.2	190	120	10	0.05	(h)

Alkaline hydrolysis has the economic advantage of using cheap NaOH as the catalyst for hydrolytically recycled PET, with no high-pressure equipment necessary. However, recycled PET by alkaline hydrolysis contains up to 40 wt% impurities, while mechanical and other recycling methods contain 1–10 wt% of contaminants. Alkaline hydrolysis also converts ethylene glycol to CO₂ and oxalic acid, which adds value and makes the process economically feasible because oxalic acid is more valuable than ethylene glycol. Furthermore, the final product is colorless even when using green PET bottle feedstock, implying that this process effectively oxidizes the green dye in the PET feedstock.

Since the development of hydrolytic techniques under alkaline or acidic condition, hydrolytic scission of PET under neutral conditions has been developed. The PET can be depolymerized in a high-pressure autoclave with excess water, resulting in high-purity TPA and ethylene glycol. The complete depolymerization of PET by this method is performed within 2 hours at 265°C (Cardi et al. 1993), and advanced methods (Campanelli et al. 1994b) using metal salts as catalysts are also being investigated.

Methanolysis

Methanolysis is a simple alcoholysis treatment, which produces DMT and ethylene glycol. The PET is dissolved and partially glycolyzed by the methanolysis, resulting in DMT and ethylene glycol, and then DMT is purified by crystallization and distillation (Scheirs 1998). Typical PET feedstocks for methanolysis are postconsumer films, plant waste, fiber waste, and bottle scraps. Because it is much easier to purify DMT compared with BHET, PET feedstocks with lower quality are acceptable in the methanolysis process as compared with the glycolysis process, and other products such as ethylene glycol and methanol are easily recycled and recovered (Goto et al. 2002; Kurokawa et al. 2003; Genta et al. 2007; Guclu et al. 2003). Although methanolysis has high processing costs, its relatively low feedstock costs compensate for the relatively high processing costs because methanolysis is more tolerant of contamination. However, the main feedstock for PET production is purified TPA, not DMT, which is the major product of methanolysis. The conversion of DMT to TPA by hydrolysis adds considerable expense to the methanolysis process. A novel methanolysis technique, which has lower conversion costs and high yields, has been developed by Eastman Kodak and DuPont (Scheirs 1998).

Aminolysis

Aminolysis of PET involves the reaction of PET with amine, and terephthalamide is the product obtained. There are many studies concerning the depolymerization of PET waste using different amines such as allylamine, morpholine, hydrazine, and polyamines (Spychaj et al. 2001; Shukla and Harad 2006). This process is not commercially utilized in PET recycling. However, partial aminolysis has found an application in the improvement of PET properties in the manufacture of fibers (Collins et al. 1991). The reaction was generally conducted using primary amine aqueous solutions such as methylamine, ethylamine, and ethanolamine at 20–100°C (Collins et al. 1991). Anhydrous *n*-butylamine was also used as the aminolytic agent (Collins et al. 1991).

Application of recycled PET

Fiber applications for mechanically recycled PET

There are several major application areas for the PET textile industry, such as staple fibers, filament, nonwoven fabric and fiberfill. Generally, recycled PET has been utilized for fibers of more than 6 denier, while the largest market for the PET fiber industry (clothing and apparel applications) is in the 1.5-3 denier range. A novel processing technology was developed in 1993, which enabled recycled PET to be used in fibers much finer than 3 denier, and this fiber obtained from recycled PET has been commercialized (Scheirs 1998). These fibers require high-quality feedstock, and postconsumer PET flakes used for these fibers should have consistent intrinsic viscosity around 0.7 dl g^{-1} .

Traditionally, recycled PET has not been widely used for filament production because of contaminants leading to breakage of the filament. Melt filtration of the PET is carried out to achieve high quality of the resin. Nonwoven fabrics, used as filters and absorbents, are prepared by spun bounding technology. In the production of recycled PET, nonwoven, cleaned PET bottle flake is dried, crystallized and incorporated into the extruder. The melt-spun filaments are spread and distributed on a screen belt under strong vacuum, and the fibrous materials are compressed into a mat for web-forming. Fiberfill is utilized for various applications such as filling materials and insulant in sleeping bags, pillows, and bedding. These applications allow the use of colored PET flake and require PET with intrinsic viscosity in the range of $0.58\text{-}0.65 \text{ dl g}^{-1}$.

Applications for chemically recycled PET

Chemically recycled PET can be used for applications in the manufacture of polyurethane (Lee et al. 1994, 1995; Vaidya and Nadkarni 1990) and unsaturated polyester resin (Abdel-Azim et al. 1994; Lu and Kim 2001; Suh et al. 2000). Polyurethane is a widely used polymer for various applications such as insulation, seating materials, and artificial leather (Park et al. 2013). The oligoesters obtained from PET glycolysis can be further reacted with aliphatic diacids to form polyester polyols, which can be utilized as the starting material for polyurethane synthesis (Nikles and Farahat 2005). The PET oligoesters can be directly reacted with diisocyanate for preparation of polyurethane (Ikladious 2000; Chen et al. 2001b; Pardal and Tersac 2007; Kosmidis et al. 2001). Mercit (Mercit and Akar 2001) reported on the synthesis of new urethane oil from glycolysis-treated PET waste. The glycolysis of PET using different diol compounds produces different glycolysis products, leading to a broad spectrum of physical properties of the polyurethane obtained from these products.

Unsaturated polyester resins have been used extensively as a matrix for high-performance fiber-reinforced polymer composites, and they can be synthesized from virgin material or from recycled PET (Lu and Kim 2001). Pimpan et al. (Pimpan et al. 2003) investigated the synthesis and curing process of unsaturated polyester resins from PET waste bottles. This study (Pimpan et al. 2003) discovered that the type of glycol, such as ethylene glycol, propylene glycol, or diethylene glycol, had a crucial effect on the properties of the glycolysis products. The results from this study (Pimpan et al. 2003) also showed that the type of bottle did not significantly affect the properties of the unsaturated polyester resins, implying that no separation bottle is needed.

Conclusion

In summary, PET recycling is one strategy for PET waste management with economic and environmental advantages. Extensive studies concerning the synthesis, properties, and processing of virgin PET as well as their composites for high performance are reported in this paper. In addition, publication reviews as described above indicate that many scientific technologies have been developed in the field of PET recycling. In particular, this review has focused on the advantages and disadvantages of mechanical and chemical recycling methods with an emphasis on industrial applications. It is possible to use mechanically recycled PET for various applications in the textile industry, and the products obtained from chemical recycling of PET can be also used as feedstocks for polyurethane and unsaturated polyester resins. With these efforts to increase the use and specification of recycled grade PET as a replacement for virgin PET, the recycling of PET waste is an effective method for enhancing the environmental performance of the PET fashion and textile industries.

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