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Recycling of poly(ethylene terephthalate) – A review focusing on chemical methods

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Abstract. Recycling of poly(ethylene terephthalate) (PET) is of crucial importance, since worldwide amounts of PET-waste increase rapidly due to its widespread applications. Hence, several methods have been developed, like energetic, material, thermo-mechanical and chemical recycling of PET. Most frequently, PET-waste is incinerated for energy recovery, used as additive in concrete composites or glycolysed to yield mixtures of monomers and undefined oligomers. While energetic and thermo-mechanical recycling entail downcycling of the material, chemical recycling requires considerable amounts of chemicals and demanding processing steps entailing toxic and ecological issues. This review provides a thorough survey of PET-recycling including energetic, material, thermo-mechanical and chemical methods. It focuses on chemical methods describing important reaction parameters and yields of obtained reaction products. While most methods yield monomers, only a few yield undefined low molecular weight oligomers for impaired applications (dispersants or plasticizers). Further, the present work presents an alternative chemical recycling method of PET in comparison to existing chemical methods.

Keywords: *recycling, poly(ethylene terephthalate), degradation, monomers, oligomers*

1. Introduction

From the late 1990s to the year 2011, the worldwide amount of poly(ethylene terephthalate) (PET) increased rapidly from approximately 14 to 60 million tons [1, 2]. Correspondingly, equivalent amounts of PET-waste are generated. Since life cycle assessment studies showed that re-utilization of PET has a positive effect on energy balance and the reduction of CO_2 emissions, for ecological reasons, the need of an appropriate PET-recycling is greater than ever [3–7]. Thus, several PET-recycling methods have been developed, which were partially reviewed in the literature [8–15].

One of these methods is the incineration of the PET-waste using the released heat of combustion (direct energy recovery), which amounts to about 46 $MJ\cdot kg^{-1}$ [13, 14]. Another method, the pyrolysis of the PET-waste is applied to produce a substitute of coal (carbonization) or aromatic and aliphatic compounds as an alternative for fossil fuels (indirect energy recovery). These applications are classified as energetic recycling, since they both use the released thermal energy either directly from incineration of the PET-waste or indirectly from combustion of pyrolysis products [16–31].

For material recycling, the PET-waste is used as an additive in crushed form. In this application, it acts as a partial substitute of natural raw materials such as sand and other natural aggregates and helps reducing the consumption of such resources. It is mixed into composite materials (e.g. asphalt, mortar or concrete) or other polymers to improve their mechanical properties [32–65].

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Thermo-mechanical recycling represents another method of re-utilization of PET-waste. Generally, the PET-waste is re-melted without additives. Since this procedure leads to a downcycling of the material due to discoloration or thermal degradation, it is also re-melted with specific additives (complementary colors, inorganic materials or epoxy-chain extenders) to prevent the downcycling [66–94].

In this review, downcycling is used in various meanings. In a strict sense, downcycling means the deterioration of the material properties of PET by damaging or shortening the polymer chains of the polyester. However, processes leading to the utilization of PET in low value applications such as a filler material where the material potential of PET is not fully used can also be considered as some kind of 'downcycling' in a wider sense. Ultimately, thermal degradation processes like incineration, pyrolysis or carbonization which lead to monomers and hence a total loss of efforts for synthesis for virgin PET can also be considered as 'downcycling'. Hence, most energetic and material recycling methods ultimately lead to a downcycling of PET.

In contrast, chemical recycling methods offer the possibility of re-introduction of PET into the material cycle without loss of quality by de-polymerizing PET-waste into monomers. These monomers are used for re-polymerization. Most frequently applied methods use water (hydrolysis), glycols (glycolysis), amines (aminolysis) and alcohols (alcoholysis) for de-polymerization of PET-waste under various reaction conditions. However, these methods require high temperature and high pressure conditions as well as considerable amounts of solvents and degrading agents for de-polymerization [95]. Further, chemical recycling methods generally entail separation and purification steps for product recovery. Thus, this kind of chemical recycling imposes toxic and environmentally hazardous issues.

In the present review a comprehensive survey of PET-recycling methods is given, reviewing both conventional and exceptional methods (carbonization of PET, use of castor oil or ionic liquids for PET de-polymerization). This review focuses on chemical recycling methods with respect to the yields of the obtained reaction products and their usability for value added applications.

2. Classification of recycling poly(ethylene terephthalate)

2.1. Energetic recycling

2.1.1. Pyrolysis

Pyrolysis of PET-waste was first described in 1982 by Day *et al*. [96]. It is an alternative to PET disposal in landfills. In general, PET waste is pyrolysed without further purification of the plastic waste. Pyrolysis is carried out at temperatures between 200 and 900 °C for 0.5 hours to 1hour [16–19, 21, 24–31, 97–101]. The majority of pyrolyses were conducted to produce aliphatic and aromatic hydrocarbons as an alternative for fossil fuels or as a source for chemicals [17, 19, 21, 24–31]. Other research on pyrolysis was done to either model degradation kinetics or to use PET-wastes in the production of coke for steel making process [98, 100–103]. Finally, Urbanova *et al*. [104] studied the influence of IR laser irradiation for PET pyrolysis. As in conventional pyrolysis, they obtained aliphatic and aromatic hydrocarbons.

2.1.2. Carbonization

Carbonization is a second method of pyrolysing PET-waste. It is carried out at temperatures between 350–1550 °C for 0.5 to 18.5 hours [105–111]. The major application of carbonization is the production of active carbon as adsorbent materials for either waste water or as CO_2-scavenger [105, 108, 110, 111]. Carbonized PET-waste is also used as slag foaming agent and as a substitute of coal in steel making process [106, 107].

2.2. Sorting

Since PET-waste is often supplied in mixtures with other polymers, PET has to be separated from these polymers prior to re-processing. Therefore, several methods have been developed and are described in the literature. These methods comprise froth flotation, wet shaking table, swelling or thermo-mechanical procedures [112–123].

2.2.1. Application of PET as additive

A further use of recycled PET-waste after sorting is its use as additive in stone mastic asphalt, cementitious materials, mortars or concrete composites. The PET-waste is mixed in crushed shape in the corresponding materials to improve mechanical properties of these composites. Moreover, this method is intended to reduce the weight of such composites, when being used as material for the construction of large buildings [32–64]. A second possibility of using PET-waste as an additive is described by de Mello *et al*. [65], who applied PET particles as reinforcing component in polyurethane foams to improve the mechanical strength of the material. Finally, Zou *et al*. [124] studied the use of recycled PET as additive for paper coatings. Although proposing an eco-friendly alternative to energetic recycling methods and landfilling disposal, this method remains a downcycling application for PET-waste in the wider sense since the potential of the material PET is not used to its full extent in this low value application.

2.3. Thermo-mechanical recycling

2.3.1. Re-melting without additives

The simplest way of thermo-mechanical recycling is re-melting the sorted PET-waste. This method is applied in bottle-to-bottle technologies, where sorted PET-bottles are re-melted in crushed shape and reprocessed to bottles as beverage packaging. Several studies have been conducted on thermal re-processing PET. During this process, the polymer is exposed to high temperatures, shear forces and pressures. Thus, thermal degradation of PET occurs. As a consequence, reduced thermal and mechanical properties of the re-processed material were the main findings of these investigations (downcycling) [66–78]. Hence, a repeated thermal re-processing of PET-waste finally leads to a downcycling of the material.

2.3.2. Re-melting with additives

To make re-utilization of recycled PET feasible, the application of additives to PET-waste has emerged. Since the collection of PET-waste entails generally a mixture of differently colored PET-materials, such thermally re-processed material leads to undesired coloration of recycled PET. Therefore, the addition of complementary colors to PET-waste has been applied to mask discoloration [79]. Although being an approved procedure, this method probably limits the use of the recycled PET. Likewise, additives were developed to improve viscosity and impact strength of recycled PET, but the recyclate was excluded from food packaging in Europe [80].

Besides such additives, the addition of other polymers (e. g. polyethylene, polypropylene) or inorganic materials (clay minerals) has been applied for thermo-mechanical PET recycling to improve mechanical properties of the recyclate [81–94]. In addition to blending PET with a second polymer, the use of compatibilizers such as ethylene vinyl acetate, ethylene-butyl acrylate-glycidyl methacrylate copolymer, poly (styrene-ethylene/butylene-styrene) and epoxy-chain extenders has been applied to further improve mechanical properties [82–86]. Although the use of additives to PET can improve properties of thermo-mechanically recycled PET, it ultimately leads to a downcycling of the material, since this material becomes increasingly difficult to be recycled again due to the heterogeneous and inherent composition of PET with the other components.

2.4. Chemical recycling

2.4.1. Ionic liquids

The application of ionic liquids for de-polymerization was first described in the year 2000 by Adams *et al*. [125]. This method was developed to avoid the drawbacks of former methods like methanolysis (high pressure and temperature), glycolysis (heterogeneous reaction products) or acidic and alkaline hydrolysis (pollution problems) to provide an eco-friendly degrading agent for polymers and to enable degradation under moderate reaction conditions. However, no application of the obtained reaction products was described. The general reaction scheme is depicted in Figure 1a [126].

Wang *et al*. [126] depolymerized PET with an excess of the ionic liquid at 120– 200 °C for 6–10 hours. After the reaction was finished, the residual PET was removed, the degradation product precipitated by the addition of water and steam extracted for purification. They used different ionic liquids such as 1-butyl-3-methylimidazolium tetrachloroaluminate ($[\text{bmim}]^+\text{AlCl}_4^-$), 1-butyl-3-methylimidazolium chloride ($[\text{bmim}]^+\text{Cl}^-$), 1-butyl-3-methylimidazolium bromide ($[\text{bmim}]^+\text{Br}^-$), 1-ethyl-3-methylimidazolium bromide ($[\text{emim}]^+\text{Br}^-$), 1-butyl-3-methylimidazolium tetrafluoroborate ($[\text{bmim}]^+\text{BF}_4^-$), 1-butyl-3-methylimidazolium hexafluorophoasphate ($[\text{bmim}]^+\text{PF}_6^-$), 1-butyl-3-methylimidazolium trifluoroacetate ($[\text{bmim}]^+\text{CF}_3\text{COO}^-$), 1-butyl-3-methylimidazolium acetate ($[\text{bmim}]^+\text{CH}_3\text{COO}^-$) (Figure 1b). Since the ionic liquid $[\text{bmim}]^+\text{Cl}^-$ was most stable and PET was less soluble in the other liquids, only the reaction with $[\text{bmim}]^+\text{Cl}^-$ was studied. Further, they studied the use of zinc acetate, tetrabutyl titanate and solid superacid as catalysts on the solubility of PET in the ionic liquid, but found that solubility was decreased

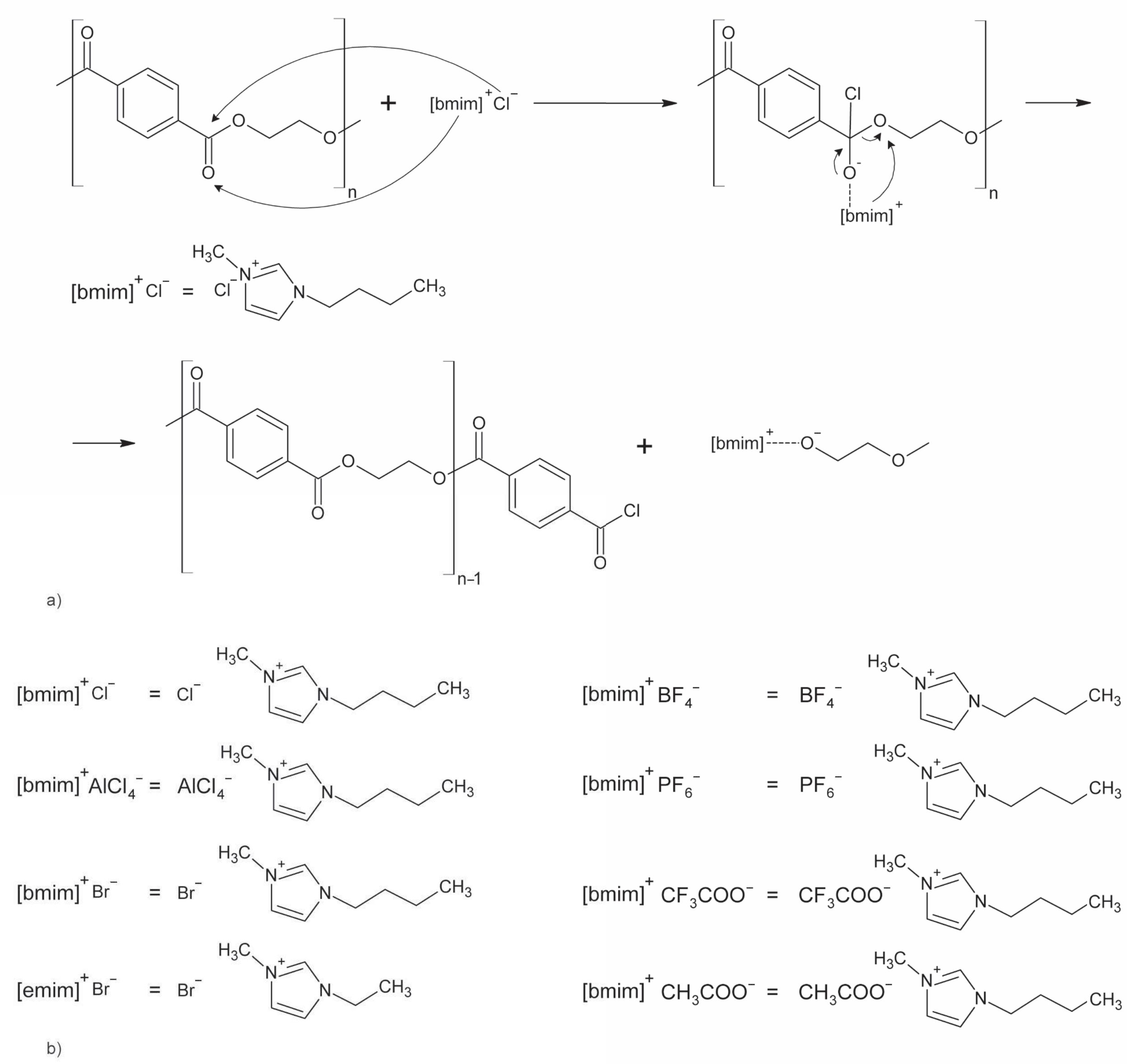

Figure 1. Reaction of the ionic liquid 1-butyl-3-methylimidazolium chloride ($[bmim]^+Cl^-$) with PET (a), Applied ionic liquids (b) [126]

independent of the applied catalyst. As expected, they found an increased degradation rate with increasing reaction temperature. The molecular weight (characterized by GPC) of the undefined oligomers was in the range of 777 to 790 $g\cdot mol^{-1}$. The yield of these products was not determined. The main objectives of their research were the study of the reaction kinetics and the recyclability of the ionic liquids for the de-polymerization reaction of PET in terms of an eco-friendly degrading agent as mentioned above.

2.4.2. Castor oil

The application of castor oil for de-polymerization was first described in the year 1999 by Kržan [127]. This method was developed to provide a renewable substitute of petrochemical agents (for example, glycols) for PET de-polymerization. After de-polymerization, the reaction products were aimed for the preparation of polyurethane systems. The general reaction scheme is depicted in Figure 2 [128].

Beneš *et al.* [128] depolymerized PET with castor oil at 230–240 °C for 0–2 hours assisted microwaves. After the reaction was finished, solid residues were removed by vacuum filtration. Further, they applied zinc acetate, sodium carbonate and sodium hydrogen carbonate as catalysts. They found that the optimum reaction temperature range is between 230 and 240 °C, whereas below 230 °C almost no reaction was observed. In contrast, reaction temperatures above 240 °C lead to undesired side reactions of the castor

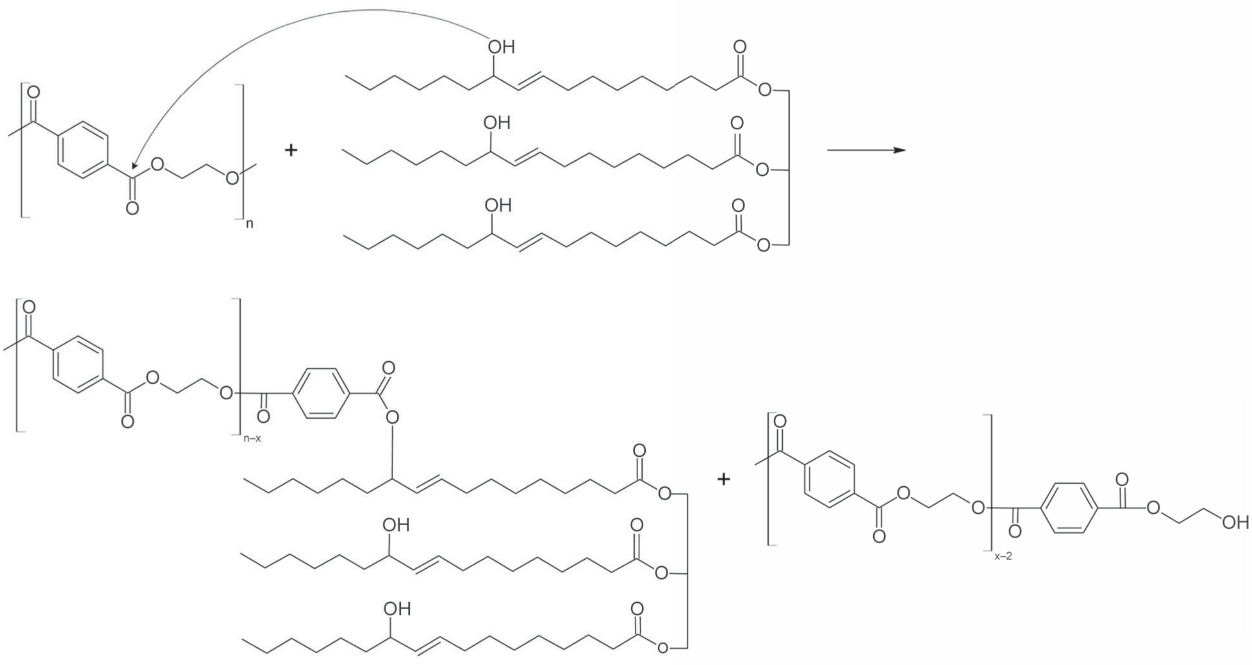

Figure 2. Reaction of castor oil with PET [128]

oil (dehydration, hydrolysis and transesterification). Moreover, zinc acetate seemed to be an efficient catalyst compared to sodium carbonate or sodium hydrogen carbonate. However, it was difficult to determine characteristic molecular weights (characterized by GPC) due to the excessive amount of applied castor oil, but it was stated, that no higher molecular weight PET-oligomers were obtained. Even with precise control of the reaction temperature a heterogeneous mixture of reaction products were obtained.

2.4.3. De-polymerization of PET using enzymes

The degradation of polymers using enzymes was first described in the year 1977 by Tokiwa and Suzuki [129]. As the use of ionic liquids and castor oil, this bio-chemical method was developed to provide an eco-friendly procedure of polymer recycling in contrast to conventional chemical recycling methods like methanolysis (high pressure and temperature), glycolysis (heterogeneous reaction products) or acidic and alkaline hydrolysis (pollution problems). The aimed application of reaction products after de-polymerization is the surface functionalization of polyester materials. Generally, PET was incubated in the enzymatic solution at temperatures between 30 to 60 °C for a period of time ranging from 3 to 14 days. Residual PET and solution were separated for product characterization [130–133].

As shown in Table 1, different enzymes such as *saccharomonospora viridis* cutinase polyesterase, *thermobifidia fusca* hydrolase, cutinase and lipase were applied for PET degradation. Mueller discussed the general applicability of *thermobifidia fusca* hydrolase for PET de-polymerization without mentioning a main degradation product or its yield [131].

Donelli *et al*. [132] only studied the surface morphology of PET treated with cutinase and did not make a qualitative or quantitative statement of possible reaction products. In contrast, Zhang *et al*. [133] studied the application of lipase as degrading agent for diethylene glycol terephthalate (DGTP) and PET at 30 °C and 14 days. They found that lipase is capable of fully converting DGTP to terephthalic acid (TPA), whereas the degradation of PET to TPA was negligible.

Finally, Kawai *et al*. [130] used *saccharomonospora viridis* cutinase polyesterase for degradation of PET at reaction conditions of 63 °C and 3 days. With this enzyme it was possible to obtain 10–27% of TPA form PET degradation.

In these references, the authors conclude, that depolymerization of PET using enzymes is generally possible as an eco-friendly alternative to conventional chemical recycling methods, since the latter require high pressure and temperature equipment and considerable amounts of toxic as well as hazardous chemicals. However, efficiency is rather low with respect to complete de-polymerization of PET and hence quantitative recovery of homogeneous reaction products for re-use is not possible.

Table 1. Reaction conditions and yields of enzymatic degradation of PET

Reaction product	Yield [%]	Enzyme	Reaction temperature [°C]	Reaction time [days]	Reference
Monomer, TPA[a]	10 to 27	Saccharomonospora viridis cutinase polyesterase	63	3	[130]
Monomer, TPA[a]	negligible	Lipase	30	14	[133]
Not given	not given	Thermobifidia fusca hydrolase	55	not given	[131]
Not given	not given	Cutinase	40	0.1	[132]

[a]TPA: Terephthalic acid

2.4.4. Alcoholysis

Alcoholysis for de-polymerization of PET was first described in the year 1991 by Wang *et al*. [134]. This method was developed to avoid the drawbacks of the methods like glycolysis (heterogeneous reaction products) or acidic and alkaline hydrolysis (pollution problems) to provide a renewable and eco-friendly degrading agent for polymers [135]. The general reaction scheme is depicted in Figure 3a.

Generally, PET is de-polymerized with an excess of an alcohol to yield corresponding esters of terephthalic acid and ethylene glycol [136].

Fávaro *et al*. [135] completely de-polymerized PET with excess supercritical ethanol at 255 °C and 116 bar for 30 to 120 minutes. They obtained diethylene terephthalate with 80% yield (Table 2) as monomer for PET synthesis. However, the obtained ethylene glycol was only suitable for an impaired application as cooling liquid due to its reduced purity.

Mendes *et al*. [34] used pentaerythrytol (PENTE) for alcoholysis of PET at 250 °C for 10 minutes. They melt-mixed PET with different concentrations of PENTE and found with increasing amount of PENTE the trend of formation from branched, undefined low molecular weight oligomers to the monomer bis(trihydroxy neopentyl) terephthalate (BTHNPT). This monomer could be used as additive for asphalt or as adhesives.

Nikje and Nazari studied alcoholysis of PET using 1-butanol, 1-pentanol and 1-hexanol. They refluxed PET with excess alcohol under microwave irradiation to accelerate complete de-polymerization of PET and obtained terephthalic acid with high purity in yields between 84 and 96% [137]. The use of microwave irradiation provided short reaction times and no further oxidation of the side product ethylene glycol. In this case, the aimed application of reaction products after de-polymerization was the synthesis of virgin PET.

Likewise, Liu *et al*. [136] used excess 1-butanol for alcoholysis of PET and studied the influence of different catalysts and their re-useability on PET conversion, but did not mention any aimed application of obtained reaction products after de-polymerization. Reaction was carried out at 205 °C for 480 minutes to give the monomer dibutyl terephthalate (DBTP). Highest yield of 95% was obtained using (3-sulfonic acid)-propyltriethylammonium chlorozincinate as catalyst.

In contrast to the previous alcoholytic methods, Dutt and Soni applied excess 2-ethyl-1-hexanol for PET-alcoholysis to produce plasticizers for nitrile rubber

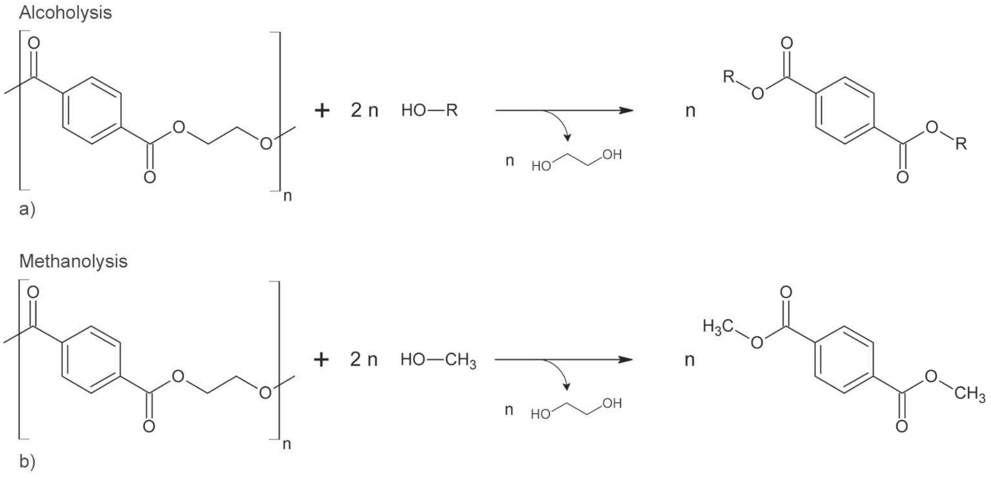

Figure 3. Chemical recycling methods of PET ((a) Alcoholysis, (b) Methanolysis)

Table 2. Reaction conditions and yields of alcoholysis, methanolysis and hydrolysis of PET

Reaction product	Yield [%]	Reagent	Reaction temperature [°C]	Reaction time [min]	Pressure [bar]	Catalyst	Reference
Monomer, DETP[a]	80	Ethanol	255	30–120	116	none	[135]
Monomer, TPA[b]	84	1-Hexanol	100	not given	1[f]	Potassium hydroxide	[137]
Monomer, DBTP[c]	95	1-Butanol	205	480	1[f]	Acidic ionic liquids	[136]
Monomer, TPA[b]	96	1-Butanol	100	not given	1[f]	Potassium hydroxide	[137]
Monomer, TPA[b]	96	1-Pentanol	100	not given	1[f]	Potassium hydroxide	[137]
Monomer, BTHNPT[d]	not given	Pentaerythrytol	250	10	1[f]	Zinc acetate	[34]
Oligomers, [250–1430 $g\cdot mol^{-1}$]	not given	Titanium alkoxide	270–280	1 to 20	1[f]	none	[139]
Oligomers, [450–900 $g\cdot mol^{-1}$]	not given	2-Ethyl-1-Hexanol	170–190	600–720	1[f]	none	[138]
Monomer, DMTP[e]	60	Methanol	270	0–90	1–150	none	[142]
Monomer, DMTP[e]	80	Methanol	300	0–90	9.8	none	[141]
Monomer, DMTP[e]	80	Methanol	300–350	2–120	200	none	[144]
Monomer, DMTP[e]	80	Methanol	300	2–120	200	none	[145]
Monomer, DMTP[e]	88	Methanol	200	120	not given	Aluminium triisopropoxide	[146]
Monomer, DMTP[e]	98	Methanol	300	0–90	147	none	[141]
Monomer, DMTP[e]	98	Methanol	330	0–90	1–150	none	[142]
Monomer, DMTP[e]	60–95	Methanol	250–270	0–60	85–140	Zinc acetate	[148]
Monomer, DMTP[e]	not given	Methanol	160–200	0–60	16	Zinc acetate	[147]
Monomer, TPA[b]	85	H_2O, Sodium Hydroxide	99	150	1[f]	none	[166]
Monomer, TPA[b]	90	H_2O	115–145	0–420	1[f]	$[(CH_3)_3N(C_{16}H_{33})]_3[PW_{12}O_{40}]$	[150]
Monomer, TPA[b]	90	H_2O	250–420	0–60	480	none	[154]
Monomer, TPA[b]	91	H_2O	220–300	6 to 60	32	Zinc acetate	[152]
Monomer, TPA[b]	96	H_2O	200	30–240	16	none	[153]
Monomer, TPA[b]	96	H_2O, Sodium Hydroxide	Molten state[g]	6	1[f]	none	[168]
Monomer, TPA[b]	98	H_2O, Sodium Hydroxide	120–150	60–420	1[f]	none	[163]
Monomer, TPA[b]	98	H_2O, Sodium Hydroxide	70–95	300–360	1[f]	Trioctyl ammonium bromide	[165]
Monomer, TPA[b]	99	H_2O	205	6–240	16	none	[155]
Monomer, TPA[b]	99	H_2O	190	10	1[f]	Hydrotalcite	[156]
Monomer, TPA[b]	99	H_2O, Sodium Hydroxide	90–98	0–60	1[f]	Tetrabutyl ammonium bromide	[164]
Monomer, TPA[b]	99	H_2O, Sodium Hydroxide	90	600–4200	1[f]	Tetrabutyl ammonium iodide	[167]
Monomer, TPA[b]	100	H_2O, Sodium Hydroxide	180	30	1[f]	Trioctylmethyl ammonium bromide	[157]
Monomer, TPA[b]	100	H_2O, Sulfuric Acid	150	60–360	1[f]	none	[161]
Monomer, TPA[b]	not given	H_2O	100–250	120	Autogen	none	[158]
Monomer, TPA[b]	not given	H_2O, Nitric Acid	Reflux	not given	1[f]	none	[160]
Not given	not given	H_2O	140–180	not given	10	none	[151]
Not given	not given	H_2O, Sulfuric Acid	30	6–120	1[f]	none	[159]
Not given	not given	H_2O, Sodium Hydroxide	Ambient temperature	15	1[f]	none	[162]
Not given	not given	H_2O, Sodium Hydroxide	120	not given	1[f]	none	[169]
Not given	not given	H_2O, Sodium Hydroxide	250[h]	not given	1[f]	none	[170]
Oligomers, [540 $g\cdot mol^{-1}$]	97	H_2O	170	180	1[f]	Zinc acetate	[149]
Oligomers, [2047 $g\cdot mol^{-1}$]	99	H_2O	170	180	1[f]	Potassium hydroxide	[149]

[a]DETP: Diethyl terephthalate. [b]TPA: Terephthalic acid. [c]DBTP: Dibutyl terephthalate. [d]BTHNPT: Bis(tri-hydroxy neopentyl) terephthalate. [e]DMTP: Dimethyl terephthalate. [f]No pressure was given in the experimental section, thus, atmospheric pressure was assumed. [g]Reaction was carried out in a microwave reactor and reaction mixture was heated until the mixture was completely molten. [h]Temperature during a differential scanning calorimetry measurement.

and nitrile rubber polyvinyl chloride (PVC) blends. The reaction was carried out at 170–190 °C for 10–12 hours. Complete alcoholysis of PET was reached after 12 hours [138]. However, the composition of the obtained reaction mixture with respect to monomers or oligomers is not clarified. Although a molecular structure of low molecular weight oligomers (450–900 $g\cdot mol^{-1}$) is postulated, it is not clear whether and how many monomers were obtained, since reaction was complete after 12 hours.

Chabert *et al*. [139] used titanium tetra-n-butoxide and titanium tetra-n-propoxide for PET de-polymerization. Concentration of titanium tetra-n-butoxide and titanium tetra-n-propoxide were 22 and 50% in the PET-mixture, which was extruded at 270–280 °C. They found that chain scission of PET proceeded faster using titanium tetra-n-propoxide compared to titanium tetra-n-butoxide. They further studied the number of active alkoxide-groups in corresponding titanium tetra-n-alkoxides and found that mainly two alkoxide-groups of the titanium tetra-n-alkoxide were involved in the chain scission reaction of PET. Mixtures of undefined low molecular weight oligomers in the range between 250 and 1430 $g\cdot mol^{-1}$ were obtained. However, they did not mention any aimed application of reaction products after de-polymerization. This procedure is advantageous in contrast to prior described solvent based procedures, which required additional separation and purification steps. However, functionality of PET-oligomers was limited due to alkyl end-groups.

Among the alcoholysis methods, reaction with methanol has gained special importance because of the low price and the availability of methanol. Methanolysis for de-polymerization of PET was first described in the year 1962 by Heisenberg *et al*. [140]. The application of the reaction products after de-polymerization was to provide the monomers for synthesis of virgin PET. The general reaction scheme is depicted in Figure 3b.

Generally, PET is de-polymerized with an excess of methanol at 160–350 °C (Table 2) to yield the monomer dimethyl terephthalate (DMTP). To increase the yield of DMTP and to shorten reaction times high pressures (9–200 bar, Table 2) are applied [141–148].

Genta and coworkers [141, 142] studied the effect of supercritical methanol compared to vapor methanol for PET de-polymerization. They found that de-polymerization proceeds faster in supercritical methanol than in vapor methanol. The reaction product consisted of a mixture of bis(hydroxy-ethylene) terephthalate (BHET), methyl-2-hydroxy ethylene terephthalate (MHET) and dimethyl terephthalate (DMTP). The yield of the main product DMTP was around 80%. Furthermore, the energy consumption of the supercritical methanolysis ($2.35\cdot 10^6$ $kJ\cdot kmol^{-1}$) was lower than of the vapor methanolysis ($2.84\cdot 10^6$ $kJ\cdot kmol^{-1}$). Goto reviewed the general applicability of supercritical fluids for de-polymerization, including methanol. Although supercritical methanol was suitable for PET de-polymerization and yielded DMTP as the main reaction product (yield 80%), the reaction mixture also consisted of BHET and MHET and hence entailed further purification steps [143].

Goto and coworkers [144, 145] studied the reaction kinetics during supercritical methanolysis of PET. Reaction was carried out at 300–350 °C at 200 bar (Table 2). Beside terephthalic acid monomethyl ester (TAMME), MHET and BHET, they obtained DMTP as main product in yields of 80%.

Likewise, Yang *et al*. [148] studied the effect of reaction temperature and time during supercritical methanolysis of PET (Table 2). Excess methanol was used for PET de-polymerization. They found that the extent of the reaction increased with increasing temperature and time. The reaction product consisted of DMTP, MHET, BHET and their dimers, which entailed further purification steps of the monomer DMTP. The yield of DMTP ranged from 60–95%.

Kurokawa *et al*. [146] studied the effect of aluminium tri-isopropoxide (ATIP) as catalyst and the solvent mixture of methanol and toluene on methanolysis of PET. Reaction was carried out at 200 °C without high pressure conditions (Table 2). With the application of the catalyst ATIP, the yield of the main product DMTP was raised to 67%. Finally, the application of ATIP combined with the solvent mixture methanol and toluene, the yield of DMTP was maximized to 88%.

Beside the use of supercritical methanol or additional application of a catalyst for methanolysis, Siddiqui *et al*. [147] studied the influence of microwave assisted methanolysis of PET in the temperature range from 160 to 200 °C with the addition of zinc acetate as catalyst. They found that the amount of de-polymerized PET increased with increasing temperature and increasing microwave power from 50 to 200 W. The characterized main product was DMTP, which was not quantified by the authors.

2.4.5. Hydrolysis

Hydrolysis of PET can be classified into neutral, acidic and alkaline hydrolysis (Table 2). The general reaction scheme of neutral and acidic hydrolysis is depicted in Figure 4. Generally, the main application of obtained reaction products after de-polymerization was to provide monomers for the synthesis of virgin PET. Neutral hydrolysis of PET is generally conducted with excess water at high temperature ranges between 115–420 °C and high pressure ranges from 10 to 480 bar. Correspondingly, reaction times are up to 7 hours [149–158].

Güçlü *et al*. [149] found that the solvent xylene provided greater de-polymerization of PET, whereas the catalysts zinc acetate or potassium hydroxide had negligible effect. They obtained low molecular weight oligomers (540+ 2047 $g\cdot mol^{-1}$, Table 2) with yields of 97+99%.

Other studies investigated the effects of different reaction parameters on neutral hydrolysis of PET, such as catalysts (zinc acetate, potassium hydroxide, phase transfer catalyst, hydrotalcite) [151, 153, 156], steam or plasma treatment [154, 155] and microwave irradiation [157]. Although having different foci in these papers, the monomer TPA was obtained as reaction product in yields of 90–100% (Table 2).

In contrast to neutral hydrolysis, acidic hydrolysis of PET is generally conducted with excess acid (sulfuric or nitric acid) at ambient temperature and atmospheric pressure. Reaction times are in between 6 and 360 minutes [159, 160]. Yoshioka *et al*. [161] conducted acidic hydrolysis at 150 °C. de Carvalho *et al.* [159] completely de-polymerized PET to yield TPA. Kumar and Rajeswara Rao studied kinetics of acidic hydrolysis and Kumar and Rao [160] and Yoshioka *et al.* [161] examined the re-usability of dilute sulfuric acid for the de-polymerization of PET. In both studies, TPA was obtained. Although neutral and acidic hydrolysis of PET gave high yields of the monomer TPA, purification of the reaction product is solvent consuming, and hence imposes environmental issues. For the alkaline hydrolysis of PET-waste metal hydroxides are used. This method yields the corresponding metal salts of terephthalic acid. By a subsequent acidification of the formed salt, pure terephthalic acid is obtained. Alkaline hydrolysis of PET-waste is generally conducted with excess metal hydroxide (sodium or potassium hydroxide) at temperature ranges between 70–150 °C. Reaction times are between 6 minutes and 70 hours (Table 2). The general reaction scheme of alkaline hydrolysis is depicted in Figure 4 [162–171].

Caparanga *et al*. [162] used alkaline washing as pretreatment of PET-waste before actual recycling. Karayannidis *et al*. [163] completely de-polymerized PET to obtain pure TPA in yields of 98%.

Kosmidis *et al*. [165], Mishra *et al*. [166], Karayannidis *et al*. [170] studied degradation kinetics of alkaline hydrolyis of PET. Kosmidis *et al*. [165] used a catalyst (trioctyl ammonium bromide) and obtained a TPA yield of 98%, whereas Mishra *et al*. [166] did not use a catalyst and obtained a TPA yield of 85%. Karayannidis *et al*. [170] did not determine any yield of any reaction product in alkaline PET hydrolysis.

Additionally to conventional alkaline hydrolysis, Khalaf and Hasan [164], Paliwal and Mungray [167], Shafique *et al*. [168] additionally used microwave or ultrasound irradiation for de-polymerization of PET. Although Khalaf and Hasan as well as Paliwal and Mungray used a catalyst (tetra butyl ammonium bromide, tetra butyl ammonium iodide), their TPA yield of 99% was not significantly greater than the yield of Shafique *et al*. [168] with 96%.

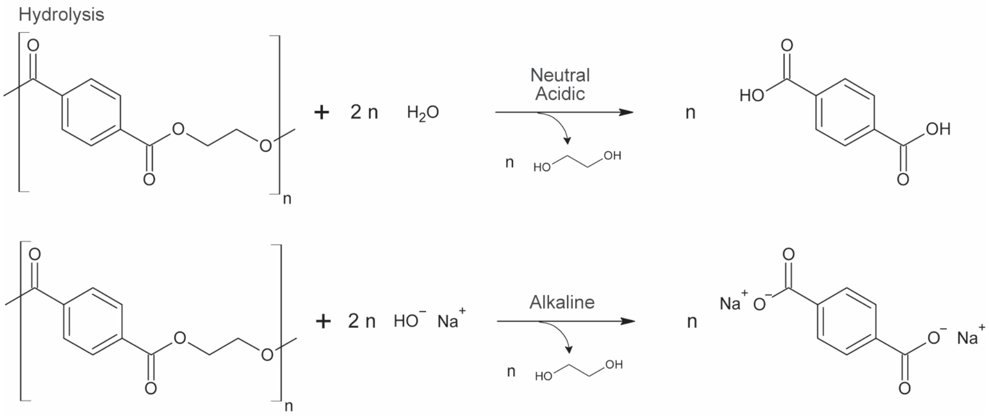

Figure 4. Chemical recycling methods of PET (Neutral, acidic and alkaline Hydrolysis)

Finally, Rosmaninho *et al*. [169] used alkaline hydrolysis for surface modification of PET not to yield PET monomers but to synthesize a cation exchange material as adsorbent for cationic contaminants. They compared this procedure with an acid surface modified PET. They found that acid modification generates more efficiently carboxyl-groups as potential cation exchange positions.

In most cases of the above described procedures, reaction products needed to be separated and purified after complete reaction.

2.4.6. Glycolysis

The general reaction scheme of PET glycolysis is depicted in Figure 5. Glycolysis of PET leads to the formation of PET-monomers and low molecular weight PET-oligomers (Table 3). Thus, a main application of the reaction products after de-polymerization was to provide monomers for the synthesis of virgin PET. The main process parameters of glycolysis are reaction temperature (110–270 °C) and reaction time (up to 15 hours) [99, 172–199]. In contrast to methanolysis, there is only little use of high pressure for glycolysis of PET [10, 180, 200].

Generally, the degrading agent was used in excess for de-polymerization of PET. Predominantly, ethylene glycol was used as degrading agent to give mainly the PET-monomer bis(hydroxy-ethylene) terephthalate (BHET) in yields between 46 to 100% [10, 11, 99, 184, 185, 187–189, 191–193, 195, 198, 200, 201–214]. Saint-Loup and co-workers also used ethylene glycol in reactive extrusion to produce low molecular weight oligomers (1450+1800 g·mol^{-1}) without quantification of their yields for synthesis of PET-polycarbonate polyesters. However, these oligomers had to be separated and purified for further processing, since the crude reaction product consisted of a heterogeneous mixture of BHET-analoga [215–218].

The second most common degrading agent for glycolysis was diethylene glycol. Here, both BHET and low molecular weight oligomers (dimers to hexamers of BHET) were obtained, but no quantification of the yield was made [64, 173, 174, 177–179, 190, 181, 219, 220].

Further, but less applied chemicals for PET-glycolysis were propylene glycol, diethanol amine and triethanol amine. Using propylene glycol, BHET-analogues were obtained, but not quantified, since these intermediates were directly used for synthesis of unsaturated polyester resins [186, 190, 221]. Application of diethanol amine and triethanol amine yielded low molecular weight oligomers (900+1130 g·mol^{-1}, not quantified) for the use as dispersants or synthesis of epoxy resin. Again, additional separation and purification of the reaction products were necessary [176, 196, 197].

The least applied diols were BHET, neopentyl glycol (NPG), tetraethylene glycol (TEEG), poly(ethylene glycol) (400 g·mol^{-1}), poly(tetramethylene oxide) (650 g·mol^{-1}) and terpoly[poly(oxyethylene)-poly-(oxypropylene)-poly(oxyethylene)] (1100 g·mol^{-1}). In case of NPG, the corresponding monomer bis(neopentyl ethylene) terephthalate was obtained in a yield of 70%. With the other diols low molecular weight oligomers were obtained, since de-polymerization agents were already of low molecular weight. These reaction products were used for the synthesis of copolymers containing polyester species. Additionally, a consecutive separation and purification of the obtained products was necessary [175, 182, 194].

Another parameter for PET-glycolysis is the application of a catalyst. A various number has been used for PET-glycolysis and is listed in Table 3. The most important catalysts were zinc acetate and manganese acetate [64, 173–175, 182–186, 192, 193, 200, 202–204, 206, 208, 210, 211, 213, 219]. Further catalysts, but less used metal acetate catalysts, than the aforementioned ones, are cobalt and lead acetate [192, 205, 211]. Baliga and Wong studied the influence of the cation zinc, manganese, cobalt and lead on the catalytic effect of corresponding metal acetates on PET-glycolysis. They found that de-polymerization increased in the order $Pb^{2+} < Co^{2+} < Mn^{2+} < Zn^{2+}$.

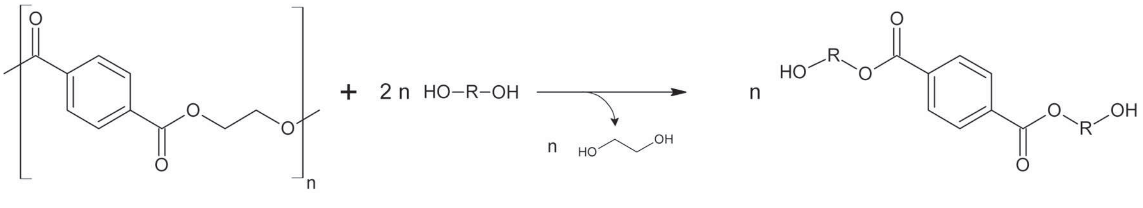

Figure 5. Chemical recycling methods of PET (Glycolysis)

Table 3. Reaction conditions and yields of PET glycolysis

Reaction product	Yield [%]	Reagent	Reaction temperature [°C]	Reaction time [hours]	Pressure [bar]	Catalyst	Reference
Monomers	0.3	EG[a]	300	0.4–0.8	11	none	[10]
Monomers	2.5	EG[a]	350	0.4–0.8	25	none	[10]
Monomers	25	EG[a]	80–200	15	1[q]	$ZnSO_4$	[192]
Monomers	46	EG[a]	150–190	1 to 4	1[q]	$Cu(OAc)_2$-[Bmim][OAc][r]	[194]
Monomers	54	EG[a]	150–190	1 to 4	1[q]	$Zn(OAc)_2$-[Bmim][OAc][s]	[194]
Monomers	57	EG[a]	196	9	1[q]	$FeCl_2$	[190]
Monomers	60	EG[a]	196	8	1[q]	Na_2SO_4	[186]
Monomers	60	EG[a]	196	8	1[q]	Na_2SO_4	[187]
Monomers	60	EG[a]	196	8	1[q]	Zeolithe	[186]
Monomers	61	EG[a]	196	1	1[q]	Na_2CO_3	[203]
Monomers	61	EG[a]	196	9	1[q]	LiCl	[190]
Monomers	61	EG[a]	196	9	1[q]	$MgCl_2$	[190]
Monomers	63	EG[a]	230–300	0.7–1.3	1[q]	Co_3O_4	[11]
Monomers	64	EG[a]	50–175	0.5–2	1[q]	[bmim]Br[t]	[213]
Monomers	65	EG[a]	80–200	15	1[q]	Zinc stearate	[192]
Monomers	67	EG[a]	230–300	0.7–1.3	1[q]	ZnO	[11]
Monomers	68	EG[a]	190	8	1[q]	none	[188]
Monomers	70	EG[a]	196	1 to 8	1[q]	Zinc acetate	[205]
Monomers	70	EG[a]	196	1 to 8	1[q]	Na_2CO_3	[205]
Monomers	70	NPG[b]	200–220	6	1[q]	Zinc acetate	[181]
Monomers	72	EG[a]	196	9	1[q]	Didymium chloride	[190]
Monomers	73	EG[a]	196	9	1[q]	$ZnCl_2$	[190]
Monomers	74	EG[a]	196	1	1[q]	Zinc acetate	[171]
Monomers	74	EG[a]	196	1	1[q]	Na_2CO_3	[171]
Monomers	74	EG[a]	196	1	1[q]	$NaHCO_3$	[171]
Monomers	74	EG[a]	196	1	1[q]	BaOH	[171]
Monomers	74	EG[a]	230–300	0.7–1.3	1[q]	Mn_3O_4	[11]
Monomers	75	EG[a]	198	10	1[q]	Zinc acetate	[191]
Monomers	75	EG[a]	198	10	1[q]	Lead acetate	[191]
Monomers	75	EG[a]	198	10	1[q]	Manganese acetate	[191]
Monomers	75	EG[a]	198	10	1[q]	Cobalt acetate	[191]
Monomers	75	EG[a]	180	8	1[q]	Zinc acetate	[183]
Monomers	75	EG[a]	180	8	1[q]	Lead acetate	[183]
Monomers	75	EG[a]	180	8	1[q]	Manganese acetate	[183]
Monomers	75	EG[a]	180	8	1[q]	Cobalt acetate	[183]
Monomers	76	EG[a]	196	1	1[q]	Zinc acetate	[203]
Monomers	78	EG[a]	190	3.5 hours	1[q]	TBD[u]	[197]
Monomers	78	EG[a]	190	not given	1[q]	Cyclic amidine	[200]
Monomers	78	EG[a]	80–200	15	1[q]	Zinc acetate	[192]
Monomers	79	EG[a]	196	1.50	1[q]	Zn/Al-hydrotalcite	[208]
Monomers	80	EG[a]	165–196	0–10	1[q]	Na_2CO_3	[211]
Monomers	80	EG[a]	165–196	0–10	1[q]	$NaHCO_3$	[211]
Monomers	80	EG[a]	165–196	0–10	1[q]	Na_2SO_4	[211]
Monomers	80	EG[a]	165–196	0–10	1[q]	K_2SO_4	[211]
Monomers	80	EG[a]	195–220	2.5–3.5	1[q]	Zinc acetate	[212]
Monomers	80	EG[a]	196	not given	1[q]	Zinc acetate	[202]
Monomers	80	EG[a]	196	not given	1[q]	Na_2CO_3	[202]
Monomers	81	EG[a]	230–300	0.7–1.3	1[q]	$ZnCo_2O_4$	[11]
Monomers	89	EG[a]	230–300	0.7–1.3	1[q]	$CoMn_2O_4$	[11]
Monomers	90	EG[a]	300	0.6–1.3	11	$\gamma\text{-}Fe_2O_3$	[199]
Monomers	92	EG[a]	197	3 to 4	1[q]	Zinc acetate	[206]
Monomers	92	EG[a]	230–300	0.7–1.3	1[q]	$ZnMn_2O_4$	[11]
Monomers	94	EG[a]	450	0.4–0.8	153	none	[10]
Monomers	98	EG[a]	198	0.5–2.5	1[q]	Zinc acetate	[210]
Monomers	98	EG[a]	198	0.5–2.5	1[q]	Lead acetate	[210]
Monomers	98	EG[a]	198	0.5–2.5	1[q]	Manganese acetate	[210]

Table 3. Reaction conditions and yields of PET glycolysis (1. continue)

Reaction product	**Yield [%]**	**Reagent**	**Reaction temperature [°C]**	**Reaction time [hours]**	**Pressure [bar]**	**Catalyst**	**Reference**
Monomers	98	EG[a]	198	0.5–2.5	1[q]	Cobalt acetate	[210]
Monomers	100	EG[a]	reflux	0.5	1[q]	Zinc acetate	[209]
Monomers	not given	DEG[c]	200–220	4	1[q]	Manganese acetate	[176]
Monomers	not given	DEG[c]	180	0.1	1[q]	Manganese acetate	[219]
Monomers	not given	DEG[c]	240	2	1[q]	Zinc acetate	[173]
Monomers	not given	EG[a]	reflux	1	1[q]	Zinc acetate	[172]
Monomers	not given	DEG[c]	240	0.02	1[q]	Zinc acetate	[218]
Monomers	not given	EG[a]	200	not given	1[q]	none	[77]
Monomers	not given	EG[a]	110	not given	1[q]	Cobalt acetate	[204]
Monomers	not given	EG[a]	110–190	0–2	1[q]	Manganese acetate	[99]
Monomers	not given	EG[a]	220	7 to 8	1[q]	Zinc acetate	[201]
Monomers	not given	EG[a]	198	3	1[q]	Zinc acetate	[207]
Monomers	not given	EG[a]	198	3	1[q]	Manganese acetate	[207]
Monomers	not given	EG[a]	196	not given	1[q]	$NaHCO_3$	[202]
Monomers	not given	EG[a]	196	not given	1[q]	Na_2SO_4	[202]
Monomers	not given	EG[a]	196	not given	1[q]	K_2SO_4	[202]
Monomers	not given	EG[a]	196	1 to 8	1[q]	$NaHCO_3$	[205]
Monomers	not given	EG[a]	196	1 to 8	1[q]	Na_2SO_4	[205]
Monomers	not given	EG[a]	196	1 to 8	1[q]	K_2SO_4	[205]
Monomers	not given	EG[a]	190	8	1[q]	Zinc acetate	[184]
Monomers	not given	DEG[c]	190	8	1[q]	Zinc acetate	[184]
Monomers	not given	PG[d]	190	8	1[q]	Zinc acetate	[184]
Monomers	not given	PG[d]	235	5	2 to 5	none	[179]
Monomers	not given	PG[d]	190+210	6+1	1[q]	Tetrabutoxy titanium	[220]
Monomers	not given	PG[d]	190	8	1[q]	Zinc acetate	[185]
Not given	not given	DEG[c]	210	not given	1[q]	Zinc acetate	[64]
Not given	not given	PG[d]	210	not given	1[q]	Zinc acetate	[64]
Not given	not given	DPG[e]	210	not given	1[q]	Zinc acetate	[64]
Not given	not given	BG[f]	210	not given	1[q]	Zinc acetate	[64]
Not given	not given	DEG[c]	200–220	6	1[q]	Manganese acetate	[180]
Not given	not given	PG[d]	200–220	6	1[q]	Manganese acetate	[180]
Not given	not given	TEG[g]	200–220	6	1[q]	Manganese acetate	[180]
Not given	not given	EG[a]	190	8	1[q]	Zinc acetate	[182]
Oligomers [2840 $g\cdot mol^{-1}$]	17	PTMO[h]	200–270	8	1[q]	Titanium tetra isopropoxide	[193]
Oligomers [1380 $g\cdot mol^{-1}$]	18	PEG[i]	200–270	8	1[q]	Titanium tetra isopropoxide	[193]
Oligomers [1120 $g\cdot mol^{-1}$]	20	TEEG[j]	200–270	8	1[q]	Titanium tetra isopropoxide	[193]
Oligomers [2300 $g\cdot mol^{-1}$]	23	Pluronic L31[k]	200–270	8	1[q]	Titanium tetra isopropoxide	[193]
Oligomers [1131 $g\cdot mol^{-1}$]	not given	TEA[l]	190–200	3	1[q]	Manganese acetate	[175]
Oligomers [1360 $g\cdot mol^{-1}$]	not given	BHET[m]	250	2	1[q]	Zinc acetate	[174]
Oligomers [1450 $g\cdot mol^{-1}$]	not given	EG[a]	270	not given	1[q]	none	[214]
Oligomers [1450 $g\cdot mol^{-1}$]	not given	EG[a]	270	not given	1[q]	none	[215]
Oligomers [1450 $g\cdot mol^{-1}$]	not given	EG[a]	270	not given	1[q]	none	[217]
Oligomers [1648 $g\cdot mol^{-1}$]	not given	PEG[i]	190–200	8	1[q]	none	[189]
Oligomers [1800 $g\cdot mol^{-1}$]	not given	EG[a]	270	not given	1[q]	none	[216]
Oligomers [210–595 $g\cdot mol^{-1}$]	not given	EG[a]	170+190	1 to 6	1[q]	Zinc acetate	[199]

Table 3. Reaction conditions and yields of PET glycolysis (2. continue)

Reaction product	Yield [%]	Reagent	Reaction temperature [°C]	Reaction time [hours]	Pressure [bar]	Catalyst	Reference
Oligomers [300–500 g·mol^{-1}]	not given	DEG[c]	200–220	4	1[q]	Manganese acetate	[177]
Oligomers [450–510 g·mol^{-1}]	not given	DEG[c]	210	4	1[q]	Manganese acetate	[178]
Oligomers [497 g·mol^{-1}]	not given	BD[n]	not given	not given	1[q]	none	[221]
Oligomers [534 g·mol^{-1}]	not given	DEG[c]	190–200	8	1[q]	none	[189]
Oligomers [881 g·mol^{-1}]	not given	TEG[g]	not given	not given	1[q]	none	[221]
Oligomers [900 g·mol^{-1}]	not given	DEA[o]	170–210	3 to 4	1[q]	Manganese acetate	[195]
Oligomers [900 g·mol^{-1}]	not given	TEA[l]	170–210	3 to 4	1[q]	Manganese acetate	[195]
Oligomers [900 g·mol^{-1}]	not given	DEA[o]	180–210	3 to 4	high pressure	Manganese acetate	[196]
Oligomers [900 g·mol^{-1}]	not given	TEA[l]	180–210	3 to 4	high pressure	Manganese acetate	[196]
Oligomers [900 g·mol^{-1}]	not given	DEG[c]	180–210	3 to 4	high pressure	Manganese acetate	[196]
Oligomers [900 g·mol^{-1}]	not given	TMP[p]	180–210	3 to 4	high pressure	Manganese acetate	[196]
Oligomers [957g·mol^{-1}]	not given	DEA[o]	190–200	3	1[q]	Manganese acetate	[175]

[a]EG: Ethylene glycol. [b]NPG: Neopentyl glycol. [c]DEG: Diethylene glycol. [d]PG: Propylene glycol. [e]DPG: Dipropylene glycol. [f]BG: Butylene glycol. [g]TEG: Triethylene glycol. [h]PTMO: Poly(tetramethylene oxide). [i]PEG: Polyethylene glycol. [j]TEEG: Tetraethylene glycol. [k]Pluronic L31: Terpoly[poly(oxyethylene)-poly-(oxypropylene)-poly(oxyethylene)]. [l]TEA: Triethanol amine. [m]BHET: Bis(hydroxy ethylene) terephthalate. [n]BD: Butanediol. [o]DEA: Diethanol amine. [p]TMP: Trimethylol propane. [q]No pressure was given in the experimental section, thus, atmospheric pressure was assumed. [r]$Cu(OAc)_2$-[Bmim][OAc]: 1-butyl-3-methylimidazolium acetate-promoted copper acetate. [s]$Zn(OAc)_2$-[Bmim][OAc]: 1-Butyl-3-methylimidazolium acetate-promoted zinc acetate. [t][bmim]Br: 1-butyl-3-methylimidazolium bromide. [u]TBD: 1,5,7-triazabicyclo[4.4.0]dec-5-ene.

These results were confirmed by Ghaemy and Moassaddegh as well as Goje and Mishra [184, 192, 211]. Pingale *et al*. [191] also studied the influence of different cations, namely zinc, lithium, didymium, magnesium and iron, on the catalytic effect of respective chlorides in the glycolysis of PET. They found zinc chloride to be the most effective catalyst yielding 73% BHET, followed by didymium chloride, magnesium chloride, lithium chloride and ferric chloride. In contrast, Carné Sánchez and Collinson [193] studied the catalytic effect of zinc catalysts on PET-glycolysis with different anions, namely acetate, stearate and sulfate. They found zinc acetate to be the most effective catalyst (Table 3, 78% BHET), followed by zinc stearate (Table 3, 65% BHET) and finally zinc sulfate (Table 3, 25% BHET). Analogously, Pingale and Shukla [172], Duque-Ingunza and coworkers [203, 204, 206, 212] studied PET-glycolysis using different sodium catalysts with different anions (carbonate, bicarbonate and sulfate). The effectiveness of the sodium catalysts on glycolysis yielding BHET decreased in the following order: sodium bicarbonate > sodium carbonate > sodium sulfate.

Finally, there are only few reports about the use of very special catalyst for glycolysis. Al-Sabagh *et al.* [195] and Alnaqbi *et al.* [214] applied ionic liquids (1-butyl-3-methylimidazolium bromide or 1-butyl-3-methylimidazolium acetate as co-catalyst) for complete glycolysis of PET. Further, Fukushima and coworkers [198, 201] used cyclic amidine catalysts (e.g. 1,5,7-triazabicyclo[4.4.0]dec-5-ene, TBD) to de-polymerize PET].

Other authors used very specific catalyst like tetrabutoxy titanium, titanium tetraisopropoxide, zeolite, Zn/Al-hydrotalcite, ZnO, metal oxide spinels (Co_3O_4 and Mn_3O_4), mixed metal oxide spinels ($ZnMn_2O_4$, $CoMn_2O_4$, $ZnCo_2O_4$) and γ-Fe_2O_3 as alternatives to common catalysts [11, 187, 200, 210, 221].

In addition, glycolysis leads to the formation of undesired cyclic oligomers [128]. The monomers and low molecular weight oligomers obtained from glycolysis of PET were generally used for impaired applications, such as modifiers for PVC or other polymer composites [183, 190, 208, 222]. Further, these glycolyized products were applied for synthesis of co-polymers of undefined composition [174, 179, 181, 182, 194, 219].

2.4.7. Aminolysis/ammonolysis

Aminolysis and ammonolyis were developed, since the reactivity of the amine-group is higher than the hydroxyl-group in glycols or alcohols used in glycolysis or alcoholysis of PET [223, 224]. Further, drawbacks of other conventional chemical recycling methods (hydrolysis, methanolysis) like high temperature and high pressure conditions were aimed of being avoided by aminolysis and ammonolysis. After de-polymerization, in general, the reaction products were applied as curing agents for epoxy resins, as components for polyurethane synthesis or as plasticizers [225–228].

The general reaction scheme of aminolysis and ammonolysis is depicted in Figure 6. Here, corresponding diamides of terephthalic acid are obtained. Aminolysis and ammonolyis are generally conducted at temperatures between 25 and 190 °C. Generally no high pressure is applied and reaction time may vary from few hours to several days (Table 4). The chemicals such as alkyl amines or liquid ammonia are used in excess for PET de-polymerization (Table 4). Aminolysis and ammonolysis mainly yielded the corresponding monomeric amides of terephthalic acid [223, 226–234].

The de-polymerization agent, which was mainly applied, was ethanol amine (EA, Table 4). EA was used to synthesize the bi-functional monomer bis(2-hydroxyl-ethylene) terephthalamide (BHETPA) in yields between 62 to 91% for further polymer syntheses (Table 4) [223, 224, 230, 232]. Further, but less used chemicals for aminolysis or ammonolysis of PET are methyl amine (MA), liquor ammonia (NH_3) and ethylene diamine (EDA). As in case of EA, these chemicals lead to the formation of corresponding bi-functional monomers of terephthalic acid, which could be applied as curing agents for epoxy resins [227, 228, 233]. Hoang and Dang used excess EDA to de-polymerize PET to low molecular weight oligomers (250–820 $g \cdot mol^{-1}$, Table 4). Removal of solid residues by filtration and additional purification gave these oligomers in yields of 30%. These were assumed to be used for synthesis of polyamides or polyimides [235]. Only little use of hydrazine hydrate, triethylene tetramine, tetraethylene pentamine, allylamine, di- or triethanol amine for aminolysis of PET was made. With these chemicals corresponding diamides of terephthalic acid were obtained, which were consecutively applied as additives in concrete mixture or used for further synthesis of antibacterial chemicals [230, 233, 234].

As for glycolysis, different catalysts were also applied for aminolysis, ammonolysis. Mainly metal acetates were used (zinc acetate, sodium acetate, potassium acetate,Table 4) [226, 227, 231, 232]. Shukla and Harad studied the effect of sodium-, potassium acetate and acetic acid on efficiency of PET-aminolysis. They found sodium acetate as the most efficient catalyst, followed by potassium acetate and acetic acid [232]. More *et al*. [224] compared zinc acetate and sodium acetate for PET-aminolysis and found (as in case of glycolysis) zinc acetate to be more efficient for aminolysis reaction. Mittal *et al*. [228] compared aminolysis and ammonolysis using cetyl am-

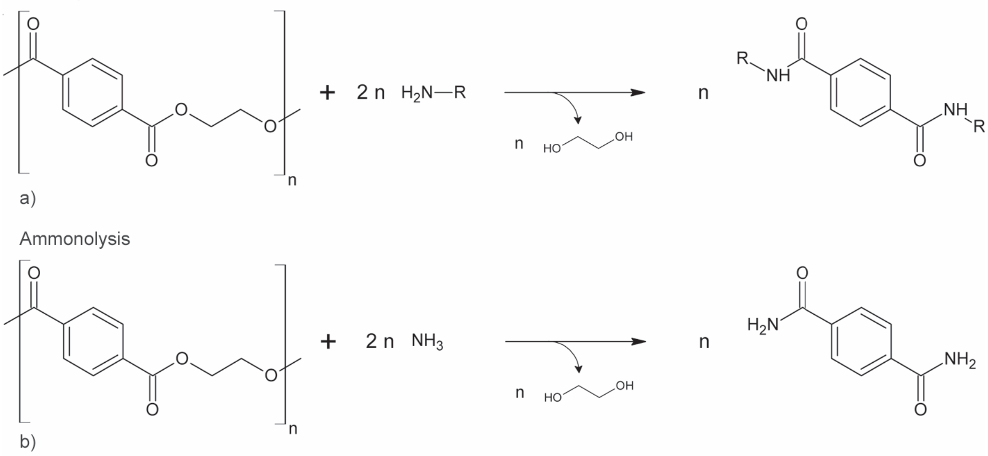

Figure 6. Chemical recycling methods of PET ((a) Aminolysis, (b) Ammonolysis)

Table 4. Reaction conditions and yields of PET aminolysis, ammonolysis

Reaction product	Yield [%]	Reagent	Reaction temperature [°C]	Reaction time [min]	Pressure [bar]	Catalyst	Reference
Monomers	38	NH_3	40	180–2700	1[h]	Cetyl ammonium bromide	[227]
Monomers	62	EA[a]	190	2 to 12	1	Dibutyltinoxide	[222]
Monomers	75	EA[a]	160	2 to 12	1[h]	Sodium acetate	[223]
Monomers	76	EA[a]	172	18	1[h]	Acetic acid	[231]
Monomers	81	EA[a]	160	2 to 12	1[h]	Zinc acetate	[223]
Monomers	85	EA[a]	172	18	1[h]	Sodium acetate	[228]
Monomers	86	Hydrazin hydrate	114	20	1[h]	none	[230]
Monomers	87	EA[a]	172	18	1[h]	Potassium sulphate	[231]
Monomers	91	EA[a]	172	18	1[h]	Sodium acetate	[231]
Monomers	94	MA[b]	40	180–2700	1[h]	Cetyl ammonium bromide	[227]
Monomers	not given	Allylamine	180	not given	15–20	none	[233]
Monomers	not given	EA[a]	180	not given	15–20	none	[233]
Monomers	not given	DEA[c]	180	not given	15–20	none	[233]
Monomers	not given	TEA[d]	180	not given	15–20	none	[233]
Monomers	not given	EDA[e]	Ambient temperature	0–300	1[h]	none	[225]
Monomers	not given	MA[b]	40	180–2700	1[h]	Cetyl ammonium bromide	[232]
Monomers	not given	NH_3	40	180–2700	1[h]	Cetyl ammonium bromide	[232]
Monomers	not given	NH_3	Ambient temperature	180–2700	1[h]	Zinc acetate	[226]
Monomers	quantitative	TETA[f]	130–140	18	1[h]	none	[229]
Monomers	quantitative	TEEPA[g]	130–140	18	1[h]	none	[229]
Oligomers [250–820 g·mol^{-1}]	30	EDA[e]	100	42	1[h]	none	[234]

[a] EA: Ethanol amine. [b] MA: Methyl amine. [c] DEA: Diethanol amine. [d] TEA: Triethanol amine. [e] EDA: Ethylene diamine. [f] TETA: Triethylene tetramine. [g] TEEPA: Tetraethylene pentamine. [h] No specific pressure was given in the experimental section, thus, atmospheric pressure was assumed.

monium bromide as catalyst. Generally, both reaction types yielded less monomeric reaction product without catalyst. Using the catalyst, they found a higher yield for aminolysis (94%, Table 4) than for ammonolysis (38%, Table 4). Compared to the aforementioned catalysts, di-butyl tinoxide used by Tawfik and Eskander [223] was less efficient for aminolysis of PET (Table 4).

All described chemical recycling methods require either high pressure apparatus (hydrolysis, methanolysis) or toxic chemicals (alkyl amines, glycols, methanol, sulfuric acid, nitric acid) to yield mainly monomers of PET. In case of glycolysis precise process control is of crucial importance either to prevent re-polymerization or other side-reactions [8, 9, 148, 153, 226].

2.4.8. Controlled de-polymerization using blocking chain scission

This method was developed by Geyer *et al.* [95] as an alternative chemical recycling method which prevents uncontrolled de-polymerization. Furthermore, it overcomes some of the drawbacks of the methods described in earlier sections: The need of high temperature and high pressure conditions, the use of toxic and environmentally problematic chemicals (either the de-polymerization agents or the catalysts) and considerable amounts of solvents. The general reaction scheme of blocking chain scission is depicted in Figure 7.

PET was melt-mixed with stoichiometric amounts of adipic acid to yield tailored oligomers of defined molecular weight. With this approach defined oligo-

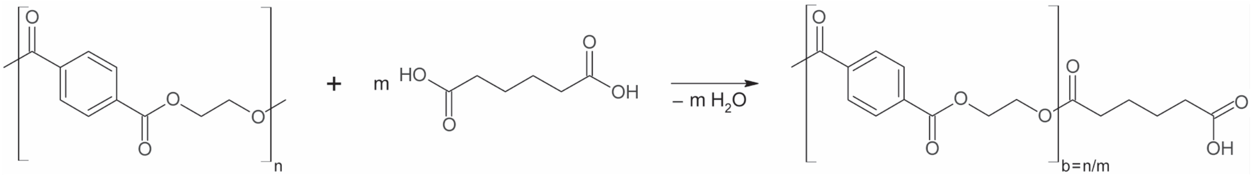

Figure 7. Blocking chain scission of PET using adipic acid [95]

mers of molecular weights in the range of 960–23 500 g·mol^{-1} were readily prepared, which was not possible with former chemical recycling methods [95]. However, this method requires sorted PET-material, which has to be free of contaminants or other polymer species.

Geyer *et al*. [236] used these defined building blocks to synthesize novel block-co-polyesters with tailored surface properties. By the combination of defined PET- and PEN-oligomers PET-PEN-block-co-polyesters with tailored block segment composition were prepared. In dependence of the block segment composition it was possible to control the dispersive surface energy and specific desorption energy of these block-co-polyesters. By tailoring these surface properties, these PET-PEN-block-co-polyesters could be applied as compatibilizing agent to prepare transparent blends of PET and PEN. Since PET and PEN are originally immiscible, leading to opaque products, in contrast, such blends of PET and PEN made transparent could now be used for high-value added applications. For example, as encapsulating material for organic photovoltaics, which require maximum transparency for optimum efficiency of the solar cell. Furthermore, such blends exhibit an improved storage modulus and higher glass transition temperatures compared to pure PET, which makes them especially interesting as materials for hot re-fillable or pasteurizable food packaging [236, 237].

3. Conclusions

A thorough review of conventional and exceptional PET-recycling with a special emphasis on on chemical methods has been given. Energetic recycling methods like incineration or pyrolysis lead to a downcycling of PET (due to thermal degradation), using its reaction products either directly (use of the released heat of combustion) or indirectly (use of obtained chemicals as alternative for fossil fuels) for energy recovery. Although carbonization produces coal, active carbon or adsorbents for chemicals as more eco-friendly applications, again, due to thermal degradation, this method remains a downcycling of PET. On the other hand, thermo-mechanical recycling of PET leads to a downcycling of the material either, since thermal conditions caused thermal degradation of PET and reduced physical and mechanical properties of the recycled PET. In contrast, chemical recycling methods, which provided complete de-polymerization of PET yielding monomers, enabled value-added re-usability of reaction products like the synthesis of virgin PET. However, these methods require high temperature and high pressure apparatus. Further, large amounts of chemicals are consumed for de-polymerization and consecutive separation as well as purification steps. Thus, such chemical recycling methods impose toxic and ecological issues. The alternative of producing PET-oligomers with chemical recycling methods is not well solved either. Although glycolysis is generally conducted without high pressure conditions, again, considerable quantities of chemicals are required for partial de-polymerization, separation and purification steps of the reaction products. Hence, as in case of complete de-polymerization of PET, partial de-polymerization imposes toxic and ecological issues either. Further, these less well-defined low molecular weight oligomers are within a heterogeneous mixture of by-products such as monomers, di- or trimers. Moreover, these heterogeneous reaction products are generally used for impaired applications such as dispersants or plasticizers. An alternative chemical recycling of PET was given by the controlled de-polymerization of PET using blocking chain scission with defined amounts of the de-polymerization agent. This method produced PET-oligomers of well-defined molecular weights in a greater range than existing chemical methods (like glycolysis). These building-blocks enabled the synthesis of tailored block-co-polyesters as potential compatibilizers to produce transparent PET-PEN-blends, which are suitable as an encapsulation material for photovoltaic cells or for hot refillable and pasteurizable food packaging.

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