Asian Journal of Chemistry; Vol. 35, No. 10 (2023), 2301-2311

ASIAN JOURNAL OF CHEMISTRY

https://doi.org/10.14233/ajchem.2023.28206

REVIEW

# Zinc Catalysts: Unlocking the Sustainable Transformation of Post-Consumer PET Waste through Chemical Recycling

ANBARASU MURUGAN, VINITHA VISWANATHAN, DHANALAKSHMI RAVIKUMAR and SIVAMURUGAN VAJIRAVELU*

P.G. & Research Department of Chemistry, Pachaiyappa's College, Chennai-600030, India

*Corresponding author: E-mail: sivaatnus@gmail.com

*Received*: 29 June 2023; *Accepted*: 10 August 2023; *Published online*: 28 September 2023; AJC-21378

This review article focuses on the reclamation of polyethylene terephthalate (PET) waste accomplished through zinc catalyzed depolymerization. Polyethylene terephthalate (PET) is a polyester that can be depolymerized in a variety of ways, including hydrolysis, alcoholysis, ammonolysis and aminolysis. Among these methods, widely employed methods are glycolysis and aminolysis due to their simplicity and adaptability in synthetic processes. The review explores the role of zinc catalysts in the PET depolymerization, with an emphasis on the latest advancements in this field. Zinc catalysts, known for their Lewis acidic nature, are capable of activating ester bonds using ionic liquids, nanoparticles, complexes, salts, porous materials and nanocomposites. Both homogeneous and heterogeneous zinc catalysts are discussed briefly in this comprehensive review.

**Keywords: Polyethylene terephthalate, Chemical recycling, Zinc catalyst, Glycolysis, Aminolysis, Depolymerization.**

## INTRODUCTION

Polyethylene terephthalate (PET), a versatile thermoplastic polymer, plays a pivotal role in the packaging materials such as water, soft-drinks and other food containers manufacturing. The demand for PET has witnessed a substantial surge in recent years, driving significant growth in its global production. The key PET manufacturing hubs are primarily located in Asia, Europe and North America, with China, the United States and Europe emerging as major production centers. Derived from the monomers ethylene glycol and terephthalic acid, PET exhibits exceptional strength-to-weight ratio, transparency and moldability making it an ideal choice for the packaging industry. The consistent rise in PET production is propelled by the expanding packaging sector's need for lightweight and durable materials. Notably, Asia, led by China, has become the epicenter of PET production, with other prominent contributors including the United States, Europe and India. The PET production process encompasses multiple stages, involving monomer synthesis, polymerization and the eventual formation of resin pellets or flakes. These versatile pellets or flakes serve as building blocks for diverse products, including beverage bottles, food containers, films and fibers. The global PET production industry is marked by intense competition and market consolidation, with a few major players driving industry dynamics. Concurrently, endeavors are underway to develop innovative and sustainable production approaches, encompassing the use of recycled materials and alternative feedstocks [1,2].

India has emerged as a significant player in the steady increase in PET consumption over the past few years. The country boasts a well-developed packaging industry, with the rising demand for packaging materials acting as a catalyst for increased PET production. The proper disposal of waste generated from polyethylene terephthalate (PET) production and its associated products is of utmost importance in addressing environmental concerns. As PET remains a widely utilized material in packaging, the substantial quantities of waste produced during its manufacturing and usage necessitate careful management to mitigate potential environmental impacts. Waste PET can be either recycled or responsibly disposed of in landfills. Recycling PET is a well-established practice, with the material finding new life in a diverse array of products, including packaging materials, fibers and even construction materials. The recycling process involves efficient collection, sorting, cleaning and processing of waste PET to create recycled resin, which can be utilized for the production of novel items. Conversely, when PET is improperly disposed of in landfills, it can persist for hundreds of years, releasing harmful chemicals into the environment. To address this issue, numerous countries have implemented regulations and policies to promote PET recycling and ensure proper waste management. In short, the implementation of suitable waste management strategies and the prioritization of PET recycling are imperative measures in mitigating its adverse environmental consequences. Governments, businesses and individuals all have roles to play in fostering sustainable waste management practices and reducing the environmental footprint associated with PET production and usage [3].

There are several methods for disposing of PET waste, including **(i) Recycling:** This is the process of collecting, sorting, cleaning and processing waste PET to produce recycled resin, which can be used to make new products. Recycling is widely practiced and can help reduce the quantity of trash dumped as landfills along with conserving natural resources. **(ii) Landfilling:** This method involves burying waste PET in a landfill. Although landfilling is a common method for disposing of waste, it can have negative environmental impacts as PET takes hundreds of years to degrade and can release harmful chemicals into the environment. **(iii) Incineration:** This method involves burning waste PET to produce energy. However, incineration can release harmful pollutants into the atmosphere and is not a sustainable method for disposing of waste PET. **(iv) Conversion to fuel:** Waste PET can be converted into a synthetic fuel known as pyrolysis oil through a process called pyrolysis. This method can help reduce the plastic waste disposal and produce a valuable fuel that can be used to generate energy. It is important to note that each technique of disposal has its own set of advantages and disadvantages. In general, the plastic recycling by depolymerization beneficial and considered an extremely sustainable technique utilized in disposing of PET waste [4,5].

**PET wastes depolymerization: Tertiary recycling:** The tertiary recycling also known as chemical recycling of a plastic waste is a process by which waste PET is depolymerized to its constituent monomers (ethylene glycol and terephthalic acid) and then used to produce new PET products. This process is seen as a more sustainable option for the control of PET waste compared to mechanical recycling, as it can help reduce the need for petroleum-based raw materials, minimize green-house gas emissions and waste landfills. The chemical recycling process usually involves several steps, including shredding and washing the waste PET, depolymerizing the PET through a process known as cracking and purifying the resulting monomers. The purified monomers can then be used as feedstocks to produce new PET products. Another method is called polymer recycling, which involves breaking down the waste PET into a mixture of short-chain PET and ethylene glycol and then using this mixture to produce new PET. The interest has been in developing and commercializing chemical recycling technologies for PET, as it has the potential to significantly reduce the environmental impact of PET waste [6,7]. However, the research and development required for improving the efficacy and scalability of these processes and to ensure that they are economically viable and environmentally sustainable in the long term [8,9].

The chemical recycling process for PET involves:

(i) **Shredding and washing:** The shredding of PET waste and washing it to remove any contaminants, such as adhesives labels and food residues. The quality of the monomers obtained improved by the purity of the waste PET.

(ii) **Depolymerization:** The next step is to depolymerize the waste PET into its constituent monomers. This process is typically achieved through a process called cracking, which uses hydrothermal/dry thermal process to depolymerize the PET polymer into its component monomers.

(iii) **Purification:** After the depolymerization step, the resulting monomers are then purified to remove any impurities and contaminants. This step is crucial to ensure that the resulting monomers are of high quality and can be used as feedstocks for producing new PET products.

(iv) **Monomer production:** After the purification step, the purified monomers are then used to produce new PET products. This is typically achieved through a process called polymerization, in which the monomers are combined to form a new polymer.

(v) **Quality control:** The final step in chemical recycling is quality control, which involves testing the quality of the new PET products to ensure that they meet the desired specifications and are suitable for their intended applications. The chemical recycling of PET is still in its early development. However, the potential benefits of chemical recycling are significant, as it can reduce the impact of the plastic waste and conserve resources.

Few companies and organizations are working on the developing and implementing chemical recycling technologies for PET, for example, Loop Industries, a US-based company that specializes in the chemical recycling of PET and other plastics. Their process involves depolymerizing waste PET into its constituent monomers and then using these monomers to produce new, high-quality PET. Carbios, a French company is developing a technology for the enzymatic recycling of PET. Their process involves using enzymes to break down the PET polymer into its constituent monomers and then using these monomers to produce new PET. In other modus operandi, a US-based waste management company has developed PET polymer chemical or tertiary recycling methods. Their process involves waste PET into its constituent monomers and then using these monomers to produce new, high-quality PET. Evonik Industries, a German chemical company has developed a chemical recycling process for the PET waste. Their process involves depolymerizing the waste PET into its constituent monomers and then using these monomers to produce new PET products. Chemical Recycling Europe is an association based in Europe that works to encourage the creation of new chemical recycling technologies and their subsequent application. The organisation is trying to raise awareness of the benefits of chemical recycling and to stimulate the adoption of these technologies on a broader scale. In addition, the association is striving to encourage wider acceptance of these technologies. These are just a few examples of the companies and organizations that are working on developing and implementing chemical recycling technologies for PET. The field is rapidly evolving and many other companies and organizations are also making important contributions to this area of research [6,7].

In India, several companies and organizations are also working on developing and implementing chemical recycling technologies for PET. Here are a few examples: Jain Bridge India Polymers Ltd. (JBIPL) is developing the chemical recycling process for PET waste. Their process involves breaking down the waste PET into its constituent monomers and then using these monomers to produce new PET products. Blue Planet Environmental Solutions is another Indian company that specializes in the development of innovative waste management solutions. They have developed a chemical reprocessing of PET of waste, which involves degrading the destroyed PET polymers depolymerizing them into their constituent monomers and then using these monomers to produce new PET products. Reliance Industries is also actively involved in the development of chemical recycling technologies. These are just a few examples of companies and organizations in India that are working on developing and implementing chemical recycling technologies for PET. The Indian government is also supportive of these efforts and has implemented several policies and initiatives to promote the adoption of chemical recycling technologies in the country [10].

**Physical recycling methods:** The physical or recycling through mechanical means is typically referred to as a supplementary recovery method. This involves separating it from any pollutants through the use of mechanical means and then reprocessing it into usable granules. The procedures incorporate trash sorting and contaminant being removed, as well as minimization through crushing, melting and modification. In addition, the challenging is physically recycle contaminated and complicated thermoplastic trash. The primary challenge with this technology is the variability in complicated plastic waste and the degradation of physical and mechanical qualities during later recovery. An additional problem is the unattractive grey colours that result from the same sticky, but tougher coloured trash.

Physical recycling or secondary recycling of the plastic waste is cut, shredded, or washed into fine flakes or pellets for manufacturing [11,12]. Mechanical recycling steps, including the sorting and separation of wastes, removal of contaminants and reduction of size *via* breaking down and clenching and thermal the extrusion process and reformation [13]. The more polluted and intricate waste is, the more difficult it becomes to maintain physical recycling. The secondary (mechanical) regeneration produces components for multiple purposes that are distinct from the initial substance that was created from the greatest post-consumer products recycled [14]. Post-consumer polymers were infrequently reprocessed through mechanical technologies that have recently become accessible. Plastic materials have a thermally capacity of 36,000 KJ $kg^{-1}$ while mechanically recycled material has a thermodynamic value of 60,000 KJ $kg^{-1}$. Recycled waste made of plastic thus saves a greater amount of energy compared to combustion. Mechanical recycling of PET waste in melting and extrusion of PET waste into fibers produces the product with limited application. Mechanical recycling can be recycled only waste made of plastic represents 15-20% of all environmental waste. The physical recycling method entails transforming recyclable plastics into something of value having a comparable nature to that type of material made of virgin plastics [15-19]. Purification of post-consumer polyester is a challenging process in the mechanical recovery technique [20].

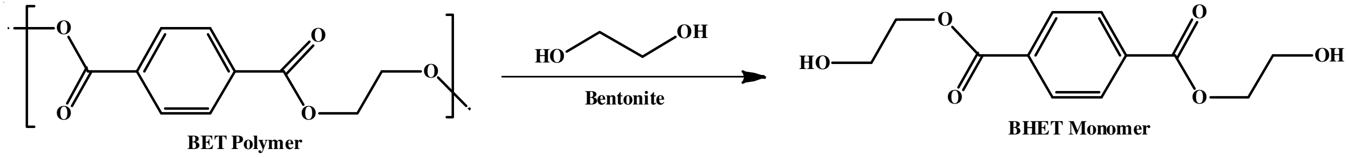
The most basic problem is dissolving any kind of polyester or other polymer waste material in the reaction medium by heat and hydrolysis of polymeric chains cause polymer degradation. The reprocessing of plastic waste by melting can produce low molecular weight oligomers, which can affect the quality of plastics such as printability, dying or any other applications. As impurity polyvinyl chloride, polyvinylidene chloride (PVTC), glue, ethylene-vinyl acetate (EVA), paper, *etc*. produced during the acid catalyst promoted hydrolysis of polyester by thermal reprocessing [21].

**Chemical recycling using zinc catalysts:** Zinc catalysts are a versatile class of catalysts with a wide spectrum of applications. One of the growing fields of study in organic transformation and environmental catalysis is the improvement of zinc catalysts with the goals of increasing their efficiency, specificity and durability. As technology advances, zinc catalysts are becoming more promising for a variety of applications. They are low-cost, environmentally friendly and highly active. As a result, they are becoming increasingly important in a variety of fields [22]. Zinc catalysts are being investigated as a potential way to manage plastic waste. Zinc catalysts can be used to break down plastic waste into its constituent monomers, which can then be reused or recycled [6,7].

The maximum recycling of PET trashes through chemical recycling (glycolysis, aminolysis, methanolysis and acidic/ alkylene hydrolysis) can be accomplished by commercially sustainable process with resource efficiency [10,19]. The post-consumer of PET waste chemical recycling method of glycolysis with the glycolytic agent of ethylene glycol (EG) in the presence of homogeneous or heterogeneous catalysts based on metal salts especially zinc-based metal oxides or salts at 180-240 ºC for 12-38 h to achieve 80-95% of monomer main product BHET in addition to other oligomers [23]. Tin (Sn) and silver (Ag) doped bimetallic ZnO nanoparticles investigated for glycolysis and aminolysis of waste from PET bottles to produce BHET and BHETA under microwave irradiation conditions [24,25].

The PET waste depolymerization through the glycolysis transesterification using bentonite clay impregnated with Lewis acidic metal such as Al(III), Fe(III) and Zn(II) explored as heterogeneous catalysts. The glycolysis reaction performed using ethylene glycol at 190-210 ºC under thermal conditions. The porous heterogeneous clay catalyst loaded with different mol% of metal ions showed an effect on surface area, crystalline nature of bentonite and pore volume of bentonite. Among the bentonite clay catalyst $Al^{3+}$ or $Zn^{2+}$ has shown significant BHET yields of up to 90% at 4 and 5% catalyst loading [19,26,27] (**Scheme-I**).

The PET chemical recycling involves ester bond cleavage and depolymerization through alkaline hydrolysis among the most appropriate under green chemistry perspectives due to mild reaction conditions break to the polymer chain. The method-



**Scheme-I:** Glycolysis of PET wastes using bentonite Zn clay catalyst

ology has produced the highest percentage of TPA up to 97% obtained. The post-consumer of PET glycolytic depolymerization using ethylene glycol on treated for very short reaction time and low temperature to obtain monomer up to 75% of BHET is mainly efficient amorphous morphology and its composed carbon and oxygen monomers are organic compounds [28]. The transition and alkali metals such as $Zn^{2+}$, $Pb^{2+}$, $Co^{2+}$, $Li^+$, $Na^+$ and $K^+$ have a significant impact role in glycolysis and aminolysis with zinc acetate being among the most advantages. Especially, the zinc material has excellent catalytic efficiency in glycolyzing PET and always potential coordination catalysis shines a light on the following research [29].

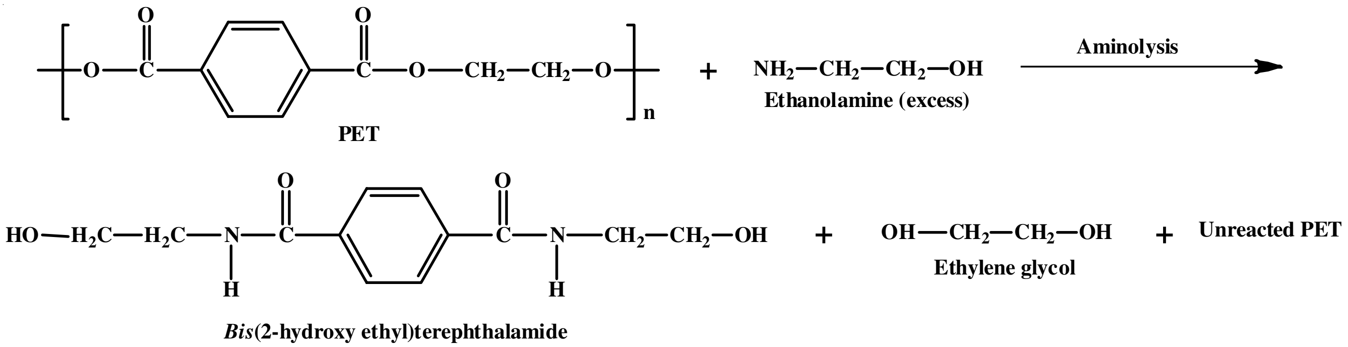
Under homogeneous conditions, zinc acetate catalyzed glycolytic depolymerization of PET using ethylene glycol under microwave irradiation have been explored. The 2% of weight of ratio of catalyst to PET weight and ratio of ethylene glycol to PET and 500 W (196 ºC) of microwave power optimized to achieved higher yield of BHET. The BHET monomer product is a 78% high percentage of yield to present. The main impact of irradiation parameters such as microwave power (MW), zinc catalyst to PET, reaction time and temperature, ethylene glycol to PET ratio and the stirring speed showed an effect on depolymerization of PET waste. In addition, the main concept of the polymer degradation is used for kinetic and potential efficiency is determined and the Arrhenius equation and find out to 36.5 KJ/mol lower than a similar procedure is researched using the conventional heating method [30]. The monomer BHET can be used as a raw material for PET manufacturing by combining with virgin BHET monomer. Furthermore, the monomer can also use to create unsaturated polyesters [31,32] alkyl resins and some other raw materials [33,34].

The aminolysis and glycolysis methods are widely used for the PET polymer depolymerization. The aminolysis reagent is amines of ethyl, propyl, butyl, ethanol, triethylene, tetramethylene, hexamethylene, benzyl, hydrazine monohydrate and some polyamines [35] and glycolysis (transesterification) reagents are ethylene glycol, diethylene glycol, propylene glycol. Both reactions can be catalyzed by zinc metal oxide, other metal oxides and ionic liquids. The aminolysis optimization temperature is (140-160 ºC) and the glycolysis temperature (180-240 ºC) is promoted by $Zn(OAc)_2$ as catalyst. The monomer obtained as a primary product using ethylene glycol are BHET, BHET dimer and oligomers and using propylene glycol produce BHPT, hydroxyl propyl terephthalate) and diethylene glycol produces oligo ester and diols. In aminolysis using ethanolamine produce a primary product BHETA, terephthalohydroxamic acid and *bis*-(2-aminoethyl) terehthalamide and secondary byproduct in the synthesis of polyurethane, bisoxazoline and nanoparticles [36-39] (**Scheme-II**).

Different types of PET trash, including double or multilayered PET, municipal/city sorting waste PET and highly coloured PET, were processed with ethylene glycol, which was then used as a reagent and solvent in the glycolysis process. The glycolysis reaction parameters time, heating rate, solvent ratio/quantity and catalyst amount [$Zn(OAc_2)$] has been significant considerations in glycolysis reaction. PET glycolysis has been carrying out by using Taguchi's measurement proposed method [40]. To optimizing parameters, (195 ºC, 150 min, 6 w/w PET and ethylene glycol, 0.2% w/w $Zn(OAc_2)$/PET) to produce yield of (86.82%, 84.52%, 82.65% and 76.93%) monomer product. The primary purpose of this study is to figure out ideal condition to achieve a better yield of BHET with optimizing and purification of monomer product. The PET degradation and depolymerization above all the parameters levels were selected based on the efficiency [40-46].

The glycolysis process has confirmed significant utilization of the waste management [47]. The optimum conditions to produce a high percentage of yield up to 78-80% of monomer (BHET). These have the largest potential to reduce the amount of PET trash transported to landfills or incinerated, and so have the greatest impact value for recycling non-mechanically recoverable PET waste [43].

**Zn-based catalyst-layered double catalyst:** Layered double hydroxides (LDHs) as well known as anionic clay [48] material



**Scheme-II:** Modified PET aminolysis reaction molecular formula is $[M(II)_{1-x}M(III)_x(OH)_2]^{x+}(A-)^{x/n}\cdot mH_2O$ [49]. The $M^{2+}$ divalent cations such as $Zn^{2+}$, $Ca^{2+}$, $Mg^{2+}$ and $Ni^{2+}$ and the ionic radii range is 0.65 to 0.80 Å. For $M^{3+}$ ($Al^{3+}$, $Cr^{3+}$, $Co^{3+}$, $Mn^{3+}$ and $Ga^{3+}$ and ionic radii is 0.62-0.69 Å. The X is mole fraction $M^{3+}/(M^{2+} + M^{3+})$ normal range is 0.17-0.33 [50]. In $A^{n-}$, n is an exchangeable interlayer $A^-$ is anions such as $CO_3^{2-}$, $SO_4^{2-}$, $NO_3^{2-}$ and $Cl^-$ and m is the number of co-intercalated of water molecules. This LDH clay material has made-up two-dimensional nanostructure [51]. The structures form layers and are called double metal hydroxide clays. LDH surfaces are formed up of particle that consists of bivalent and trivalent ions from metals as well as replaceable anions that exist between these layers [52]. The layered double hydroxide (LDH) contains diverse advantages such as high thermal and chemical stability, unique structure, simple synthesis, large specific surface area, homo-geneous metallic ions crystalline layer and presence of hydroxyl group high anion exchangeability [53]. The optimized for additional functional groups of LDHs nanocomposites were extensively fabricated and synthesized and characterized to a used variety of techniques such as ion exchange, hydrothermal, hydrolysis, dehydration and co-precipitation methods. The applications of LDHs hybrid nanostructures of materials to remove contaminated dyes, antibiotics, pharmaceuticals, heavy metals and organic compounds. LDHs have more interaction with pollutant water and the solution of pH parameter coexisting ions and the temperature was analyzed with the mechanism. The major application for LDH to advantages of the multi-pollutant system and removal for surface area augmentation, inactive microorganism's detrimental hormones all issued to be the clear role of LDHs [54].

The magnesium and aluminum layered double hydroxides (Mg/Al mole proportion is 3:1) were synthesized by the sol-gel method. The glycolysis of PET polymer to monomer, BHET have been investigated using magnesium and aluminum (Mg and Al) layered double hydroxides (LDH) contain 20% titanium dioxide nanoparticles. The thermal stability and catalytic properties of LDHs evaluated using the PET depolymerization into BHET monomer product to obtain layered double hydroxides (LDHs). The $TiO_2$ nanoparticles loaded LDHs have proved greater stability than unloaded LDHs. The enhancement of the catalyst activity of LDHs was achieved using freeze-drying and dielectric barrier discharge plasma then calcinated by thermal route. These highly efficient methods for control for features of LDHs and titania nanoparticles has highly efficient catalytic activity for the PET degradation process [55].

**Metal-organic framework (MOF):** Metal-organic frameworks (MOF) have coordination polymers together with metallic center ($Zn^{2+}$, $Fe^{3+}$, $Zr^{4+}$) and an organic multidentate ligand such as 2-methylimidazole, trimesic acid, terephthalic acid derivatives, *etc*. These materials have identified enhanced properties, enormous surface area, excellent thermal stability properties and resulting in widely used adsorbent material ($CO_2$, drugs, heavy metal ions and colourants). Catalysts act as both Lewis acid-base characters [56]. This investigation proposed a novel metal-organic framework (MOF) prepared using the hydrothermal method and the solvent is water. The polyethylene terephthalate treated with a metallic source must apply ($Zn^{2+}$/$Al^{3+}$) ions and organic ligands to achieve to glycolysis reaction obtained for the monomer product BHET. The BHET- Al-MOF product has better thermal conductivity than MIL53-Al and contains highly porous structure as compared to MIL53-Al micropores.

The novel MOF has been synthesized using BHET derived from the glycolysis of PET waste as an organic ligand and zinc & aluminum ions. It is an innovative concept to built BHET-MOF structure and properties particularly in comparison to representative MIL53-Al-MOF, a new MOF has been synthesized by hydrothermal method using zinc and aluminum, BHET produced from PET glycolysis and followed by the addition of terephthalic acid as organic ligand, nitrates as metallic resources. The XRD and ATR-FTIR were used to characterize MOFs. The advantages of using MOF for biomedical applications, according to the evidence presented since BHET is a new organic ligand obtained from PET glycolysis [57].

The MOFs are perovskite porous materials that could exhibit the unique properties of both functional and structural features. As finding results MOF innovation such as detectors or sensors [58], energy production [59], environmental monitoring [60] and magnetic materials [61]. The PET waste depolymerization by glycolysis reaction promoted by MOFs catalysts (ZIF8, ZIF67 and ZIF-5) used. The MOF based catalysts proved outstanding activity, especially ZIF-8 the PET has completely degraded PET and yield 76.75% of BHET obtained at 197 ºC. For PET glycolysis, the catalyst ZIF-8 has been shown the activation energy of 137.64 KJ/mol for the first-order kinetics. Moreover, its catalyst can be recycled and reused three times [62].

The investigation of MOF material, Zn-based catalyst ZIF-8/ZIF-67 and $CoFe_2$ composite catalyst used for glycolysis reaction. This catalyst has a significant impact with mixed PET/PBT with $CoFe_2O_4$ and ZIF-8/ZIF67 (0.05) and ethylene glycol (25 mL) was kept at 200 ºC for 1 h. The PET completely depolymerized into monomer product, BHET with yield of 84.3%. This catalyst component Zn and Co decreases less than 10% after recycling the catalyst for 5 times [63].

**Metal oxides:** Depending on Wurtzite structure and oxygen vacancies metals such as (Ni & Co) loaded on zinc oxide is a unique nanocomposite, while comparing to other metal and ZnO nanomaterials that exhibit a wide bandgap (3.37 eV). Due to its unique bandgap characteristics, ease of synthesis, low cost, great photosensitive reactions, optoelectronic features, superior biocompatibility, high electrochemical stability, *etc*. zinc oxide has garnered significant attention [22,64,65].

Zinc oxide nanoparticles (ZnO NPs) has prepared by the chemical co-precipitation technique by using $ZnCl_2$ as starting material and other metal salts as doping agents. The formation of ZnO nanoparticles is confirmed from the crystalline diffraction pattern obtained from the X-ray diffractometer method. The optical bandgap of doped ZnOs decrease while increasing the doping metal concentration. The doped ZnO nanoparticles crystalline average size is 6.50-7.70 nm. The results showed that the metal ions such as Ni and Co doped ZnO have found applications such as electrical devices, structural and optical analysis and have higher potential as optoelectronic devices [66].

The metal oxides such as Co/ZnO catalyst prepared by bio-hydrometallurgical method from the recycling of spent alkaline and lithium-ion batteries (LIBs) and recoverable zinc oxide and cobalt oxide could be achieved. These metal oxide catalysts were investigated for tertiary or chemical recycling of PET and showed complete conversion of PET into BHET up to yield of 80%. The efficiency of metal oxide catalyst throughout glycolysis reaction depends on the formation of acids and synergistic impact of mixed metal oxide nanoparticles has deduced from spent alkaline rechargeable batteries [67]. Zinc oxide containing ionic liquid catalyst investigated and showed unique properties like similar kind of ionic liquid catalyst that has shown better results while comparing to other type of ionic liquid catalyst materials [68-71]. It could be speculated that this catalyst might create hydrogen bonds with solvent ethylene glycol resulting in the superior catalytic activity [72].

Under mild reaction conditions, the polyoxometalates (POMs) with formula $K_6SiW_{11}MO_{39}(H_2O)$ ($M = Zn^{2+}$, $Co^{2+}$, $Cu^{2+}$ and $Ni^{2+}$) have shown significant catalytic activity for glycolysis of PET polyethylene terephthalate. The reaction parameters such as temperature, catalyst to PET ratio, solvent and reaction time were optimized to achieve maximum PET degradation and under optimized conditions depolymerization at 185 ºC for 30 min, afforded BHET in 84% using $SiW_{11}Zn$ catalyst. Zinc containing polyoxometalates (POMs) catalysts have shown higher catalytic efficiency, which can be applicable all types of PET polymer chemical recycing [73].

**Waste materials (spent batteries):** Spent batteries are hazardous waste since they contain existence of poisonous and pollute the environment with heavy metals ions [74-76]. Alkaline batteries, in specific, comprise of Zn & Mn is important for plants, animals, as well as humans, however in higher levels they seem to be poisonous among all living things [77-79]. Throughout many countries around the world, spent batteries were being generally thrown away as landfills as well along with waste disposal [80,81].

Metal recycling is beneficial not only to the environment but also to the economy. The recovering and reusing of Zn & Mn from spent batteries might be extremely beneficial [82] and there are several procedures have been developed for their maximum recovery from the storage batteries. The two most widely used methodologies for trying to recover Mn and Zn from spent alkaline and Zn/C batteries [83]. Using the recycling methods such as pyrometallurgical and hydrometallurgical process, the hydrometallurgical methodology seems to have environmentally advantages such as reduced gas emissions and energy consumption [84]. The solvents such as alcohol and ethylene glycol used in the investigation, might have been esterified with waste PET catalyzed by the acid produced from the alkaline batteries. In addition, the metallic zinc can catalyze the glycolysis, resulting in the transesterification of esters with alcohols, which utilize ZnO from recycled batteries as a catalyst for degradation of PET. The catalyst showed the higher PET conversion afforded 93.2% of BHET at 180 ºC in 4 h [85].

Zinc oxide is an important perovskite type material utilized for various applications, such as transparent conductive oxides [86], ultraviolet (UV) light blockers [87] and photocatalysts [84,88] and investigated using mixed zinc oxides as alkaline battery cells. After the bio-hydrometallurgical recycling treatment of alkaline and lithium-ion batteries, the oxide forms of zinc and cobalt (ReZnO and ReCoO) have been generated a mixed oxide (Co/RZnO). The mixed metal oxides investigated for the decomposition of PET plastics using ethylene glycol. The addition of Co/RZnO for a catalyst resulted in the higher BHET production (80%) [67].

To investigate the possibility, a spent alkaline battery-based catalyst for total oxidizing of hydrocarbons was developed using the carbon material recovered from spent alkaline battery. The investigation showed the effect of acid treatment on catalytic activities throughout the chemical synthesis technique, studied using five different acids such as HCl, $H_2SO_4$, $HNO_3$, $C_2H_2O_4$ (oxalic acid) and $H_3PO_4$. The volatile organic compounds (VOCs) used in the experimental studies were hexane, benzene, toluene and *o*-xylene (HBTX). Mn, Zn and Fe is abundant components in the SB400 catalysts and HBTX could be removed using SB400 catalysts between 340 and 410 ºC. As an outcome, an acid-treated SB400 catalyst could be suggested as an effective catalyst for the complete oxidation of VOCs [89].

**Deep eutectic solvents:** Under moderate reaction conditions, deep eutectic solvents (DESs) may be one of the most effective catalysts for the glycolysis of chemical reprocessing of PET into a monomer, allowing for a faster transformation and higher monomer specificity. DESs, being a substitute to ionic liquids, not only offer comparable properties to ionic liquids, but are additionally less expensive to make (due to reduced raw material prices), less hazardous and generally recyclable [90,91]. Due to these properties have led to their use in a variety of fields, including biocatalysis, extraction, $CO_2$ capture, biomedical applications and material synthesis [92,93]. More importantly, DESs derived from urea and metal salts can exhibit both strong hydrogen bond and coordination action. Furthermore, the catalytic activity of urea was greatly improved by the addition of a small amount of metal salt. Optimization conditions were discovered while studying the effects of technological parameters on PET degradation. The enhanced catalytic efficiency is due to mutually beneficial catalysis among H-bonds as well as coordination bonds produced between the DES catalyst and the ethylene glycol, according to the degradation mechanism study. The findings could serve as a valuable guide for the development of efficient catalysts for the waste from polymer decomposition along with additional chemical processes [94].

Zinc acetate complex of tropine is used as a novel catalyst of glycolysis depolymerization reaction. The tropine zinc acetate complex is synthesized under milder reaction conditions and its significantly different from basic DES. The glycolysis reaction optimized for different parameters at 190 ºC *viz.* ethylene glycol and PET (solvent & PET) ratio 5:1, tropine metal salt and PET (catalyst & PET) ratio 1:20, 100% of waste PET polymer was converted into 82.3% of monomer BHET product. The complex recovery and recycling are relatively simpler for Zn-tropine complex. These complexes are most probably to achieve coordination instead of hydrogen bonding, they could be referred to as coordination compounds of natural alkaloids and metal salts and their active improvement of green glycolysis catalysts [12]. Potassium carbonate and chemical recycling solvent ethylene glycol based deep eutectic solvent showed efficient glycolysis of PET wastes [95]. Choline chloride-$ZnCl_2$ as well as choline chloride-urea based DES are used for the aminolysis of PET wastes [96].

This study examined the sites that are active and impacting aspects of 1,3-dimethyl urea (1,3-DMU) depending on DESs in the polyethylene terephthalate (PET) glycolysis methodology. The active site of urea derivatives has been found to be the amino group, which is responsible for their basicity. However, the efficacy of catalyst is affected by steric hindrance. The final product, BHET compound was obtained to be 82% under the conditions of 5 wt% of 1,3-DMU/$Zn(OAc)_2$ in 20 mL of ethylene glycol in 20 min of glycolysis at 190 ºC. The glycolysis of PET catalyzed by 1,3-DMU/$Zn(OAc)_2$ followed the shrink-core model, with an apparent activation energy of 148.89 kJ/mol [97].

The glycolysis of PET in the presence of DES was investigated in comparison to other DESs, the DES produced from $K_2CO_3$ and ethylene glycol is superior and demonstrated higher catalytic activity. As a result, the parameterized investigation applied DES made from potassium carbonate and ethylene glycol. Since many different reaction temperatures, concentrations of DES and ethylene glycol amounts have influence on the reaction circumstances were analyzed for the PET glycolysis. The maximum BHET yield was achieved up to 88% at a reaction temperature of 180 ºC using chemical solvent and PET polymer (PET/EG) weight ratio of 1:5 and a PET/DES weight ratio is 1:6. Glycolysis was discovered to be an efficient method for the chemical recycling of PET and DES was found to be a catalytically effective catalyst for the depolymerization of PET. Moreover, PET was completely depolymerized at atmospheric pressure within relatively short reaction times [97].

**Magnetic catalyst:** Nanoparticles, particularly magnetic nanoparticles (MNPs), can be used not only as carriers but also have excellent catalytic performance, making them widely used in catalysis. PET and PBT glycolysis analyzed by utilizing $CoFe_2O_4$ and ZIF-8/ZIF-67 nanocomposites act as a heterogeneous catalyst. Since, the results showed $CoFe_2O_4$ and ZIF-8/ZIF-67 have an ideal catalytic activity in the plastic recycling method, particularly in PET and PBT mixed plastic glycolysis. For about 5 g of PET and PBT is glycolyzed using 0.05 g of $CoFe_2O_4$ & ZIF-8/ZIF-67 and added 25 mL of ethylene glycol were heated to 200 ºC for 1 h with PET wastes polymer conversion reaching 100% and yielding BHET in 84.3%. The external surface area of as-synthesized catalysts was found to have a significant impact on their catalytic performance. When the nitrogen adsorption isotherms and bimetallic Co-Zn-based ZIF with enhanced activity sites could improve glycolysis. The catalytic efficacy of $CoFe_2O_4$ and ZIF-8/ZIF-67 is significantly stable even after 5 times reaction cycles and the component concentration of Zn and Co in the catalyst was reduced by less than 10 percentage [98].

**Ionic liquids:** Ionic liquids have a wide variety of uses, and the breakdown of PET in excess chemical depolymerized with ethylene glycol has been catalyzed by ionic liquids. One example of the catalyst activity of these 1-hexyl-3-methylimidazolium (Hmim) halometallates such as [Hmim]$ZnCl_3$, [Hmim]-$CoCl_3$, [Hmim]$FeCl_4$ and [Hmim]$CuCl_3$. The advantages have influenced on catalytic activity by combination of [Hmim]-$ZnCl_3$ and [Hmim]$CoCl_3$ complexes during PET complete decomposition and complete conversion to BHET monomer product. Since, the BHET yield of 87.1% from PET polymer wastes catalyzed by an equal mole ratio of [Hmim]$ZnCl_3$ and [Hmim]$CoCl_3$ mixing is greater than that of any separate IL halometallate. The enhanced activity of catalyst [Hmim]$ZnCl_3$ accelerates the glycolysis chemical recycling reaction, resulting in the very highest percentage of PET convert to produced yield. Even so, the higher catalytic activity of $[ZnCl_3]^-$ will most likely increase the possibility of achieving BHET yield. Because $[CoCl_3]^-$ has a lower catalytic activity, chain scission will most likely be more selective. The PET glycolysis response might have been controlled among a high level of [Hmim]$CoCl_3$ for byproduct reduction and the high reactivity of [Hmim]$ZnCl_3$ for BHET production [99].

A variety of ionic liquids were synthesized and implemented as stabilizers to synthesize magnetic ionic liquids $CoFe_2O_4$ magnetic nanoparticles (MNPs). An alternative of $CoFe_2O_4$ MNPs was obtained having an average size of 7 nm and being so effectively disseminated in ethylene glycol that an ethylene glycol-based magnetic fluid was created optimized $CoFe_2O_4$ MNPs were employed as catalyst for PET glycolysis and displayed outstanding activity as a catalyst. Effects of ionic liquid as nanoparticle stabilizers have been studied and hypothetical variables on glycolysis chemical recycling method were utilized. PET conversion was 100% and BHET yield was 95.4% under optimal conditions of PET, 0.1 g of catalyst ($CoFe_2O_4$/C10-OAc) and 25 mL of ethylene glycol at 195 ºC for 2.5 h. Furthermore, the $CoFe_2O_4$ MNPs modified with ionic liquid were used as catalyst for the PET wastes depolymerized glycolysis method and demonstrated excellent catalytic efficiency. PET was completely dissolved and converted into 100% single monomer BHET yielded about 95.8% under optimal reaction conditions. Most efficiently this type of magnetic nanocomposites catalyst was quickly recovered by a applying magnetic field and can reusable subsequent reaction cycle [63].

**Metal salts:** According to the past 20 years, a significant number of metal salt catalysts, such as acetates [26,100-102], chlorides [103], hydroxides [104,105], carbonates [106], sulfates [107,108] and phosphates [109] are widely studied (Table-1). Most of these salts are soluble in ethylene glycol, but difficult to separate during separation process from the depolymerized products necessitate a supplementary component (distillation) mostly in the chemical reactions. Metal salts and oxides have numerous applications, ranging from the chemical sector (cata-lysis) to the electronics industry (electrodes and supercon-ductors). The utilization of metallic salts as catalysts in glyco-lysis processes may offer greater efficiency compared to the traditional catalysts. This is due to their ability to yield a higher percentage of single monomer compounds, resulting in supe-rior physical strength, increased melting & boiling points, and enhanced versatility in various applications. Additionally, metallic salts exhibit favourable

TABLE-1
METAL SALTS FOR PET WASTE GLYCOLYSIS

| Catalyst | BHET monomer yield (%) | Temp. (°C) | Time (min) | EG/PET ratio (mol/mol) | PET/catalyst weight ratio | Ref. |
|---|---|---|---|---|---|---|
| Zinc acetate | 85.60 | 196 | 180 | 5 | 0.01 | [26] |
| Zinc acetate | 62.80 | 200 | 150 | 2.77 | 0.003 | [109] |
| Titanium phosphate | 97.50 | 200 | 150 | 2.77 | 0.003 | [109] |
| Zinc acetate | 62.51 | 190 | 480 | 6 | 0.005 | [104] |
| Lead acetate | 61.50 | 190 | 480 | 6 | 0.005 | [104] |
| Sodium carbonate | 61.50 | 190 | 480 | 6 | 0.005 | [104] |
| Sodium bicarbonate | 61.94 | 190 | 480 | 6 | 0.005 | [104] |
| Acetic acid | 62.42 | 190 | 480 | 6 | 0.005 | [108] |
| Lithium hydroxide | 63.50 | 190 | 480 | 6 | 0.005 | [108] |
| Sodium sulphate | 65.72 | 190 | 480 | 6 | 0.005 | [108] |
| Potassium sulphate | 64.42 | 190 | 480 | 6 | 0.005 | [108] |
| β-Zeolite | 66.00 | 196 | 480 | 6 | 0.01 | [107] |
| γ-Zeolite | 65.00 | 196 | 480 | 6 | 0.01 | [107] |
| Zinc chloride | 73.24 | 197 | 480 | 10 | 0.005 | [111] |
| Lithium chloride | 59.46 | 197 | 480 | 10 | 0.005 | [111] |
| Didymium chloride | 71.01 | 197 | 480 | 10 | 0.005 | [111] |
| Magnesium chloride | 55.67 | 197 | 480 | 10 | 0.005 | [111] |
| Ferric chloride | 56.28 | 197 | 480 | 10 | 0.005 | [111] |
| Zinc oxide on silica nanoparticle | -85.00 | 300 | 80 | 11 | 0.01 | [111] |
| Magnesium oxide on silica nanoparticle | >90.00 | 300 | 80 | 11 | 0.01 | [111] |
| Ionic liquids | No data 100% conversion | 190 | 120 | 10 | 0.05 | [68,69] |
| [bmim] OH | 71.20 | 190 | 120 | 10 | 0.05 | [112] |

characteristics such as fluidi-zed accommodation, potential for recovery, ease of isolation and extended shelf-life [110].

The pure component oxides of metals such as ($Mn_3O_4$, ZnO and $Co_3O_4$) were isolated by utilizing a simple precipitation or co-precipitation process. The developed nanocomposites were used as transesterification catalysts in the PET glycolysis method. The combination of oxide spinels outperformed while comparing with the metal oxides in catalytic performance due to their larger porosity and greater acid site concentrations on the catalyst's interface. Ethylene glycol was applied as a transesterification reagent to depolymerize waste PET (mostly soft-drink bottles) to the monomer *bis*(2-hydroxyethyl)terephthalate (BHET). The impact of various parameters such as temperature, catalyst type, reaction time, EG/PET molar ratio and catalyst/PET weight ratio have been evaluated. The results revealed that the catalyst that produced the highest yield of BHET was 92.2 mol% [113].

Superheated PET can be used to create highly effective coordination compounds with zinc ions, which have been demonstrated to be more efficient than Ti, Fe and Co. Taguchi method for optimizing process parameters in the microwave-induced glycolysis of waste PET was chemical depolymerization. The effect of varying the amounts of ethylene glycol (EG) glycolytic solvent used on the number of catalysts and PET:EG ratio was researched. They optimized an outstanding reaction amalgamation of 1:3 with Taguchi within 0.5 h, EG with a 1% zinc acetate salt produces a 65% molar yield of BHET [114].

## Conclusion

The PET polymer wastes chemical or tertiary recycling was achievable with effectively managed by zinc-based catalysts. Since, zinc is a non-toxic, inexpensive and readily available metal that can be used to catalyze the glycolysis of PET in a more sustainable method. Both homogeneous and heterogeneous zinc catalysts can be used for the depolymerization of PET. Zinc-containing heterogeneous catalysts that are supported on a solid substrate, such as clay, zeolites or nanoparticles offer several advantages over homogeneous catalysts. Heterogeneous zinc catalysts are effective at depolymerizing PET to BHET with yields of over 90%. The use of heterogeneous catalysts can also make the process of chemical recycling more sustainable as the catalysts can be reused and the byproducts of the reaction can be recycled or disposed of safely. In addition to PET bottles, polyester textile waste can also be chemically recycled into value-added monomers. The use of zinc catalysts for the chemical recycling of PET polymer wastes is a promising new technology that has the potential to reduce the quantity of plastic waste that ends up in dumps into the seas. This technology is still under development, but it has the potential to make a significant impact on the sustainability of the plastic industry.

## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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