A Review Of Chemical Recycling Pathways For Waste Polyurethane

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Abstract

Polyurethane (PU) is a commonly utilized polymer due to its versatility and performance properties. However, its resistance to deterioration creates considerable hurdles for end-of-life care. This review gives a shortoverview of present recycling strategies for PU, with a particular emphasis on chemical recycling processes, including hydrolysis, alcoholysis, acidolysis, glycolysis, aminolysis, and phosphorolysis. These approaches enable the breakdown of PU into useful chemical constituents, facilitating potential reuse in polymer synthesis.This review highlights the mechanism, advantages, and limits of each method. Although additional optimization is required, chemical recycling offers a viable approach for enhancing the sustainability of PU material lifecycles.

Keywords: Polyurethane (PU), Chemical Recycling, Hydrolysis, Alcoholysis, Acidolysis, Glycolysis, Aminolysis, and Phosphorolysis, Rigid foam, Flexible foam.

1. Introduction

Polyurethane (PU) is the most versatile polymer renowned for its wide range of physical and chemical properties, making it a key material in the field of polymer science. Its performance and structural adaptability have added to its increasing importance in both the industrial and research sectors(1).The size of the worldwide polyurethane market was assessed at USD 87.10 billion in 2023 and is predicted to increase at a compound annual growth rate (CAGR) of 4.4% from USD 91.49 billion in 2024 to USD 135.08 billion by 2032(2,3).

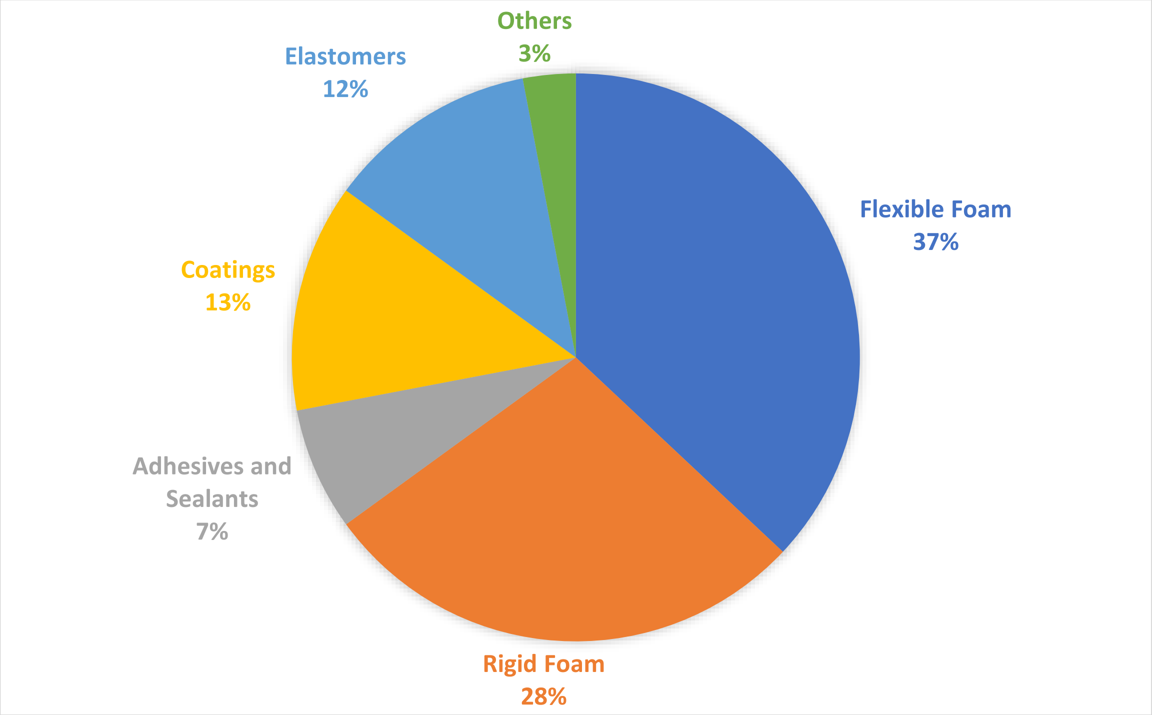


Fig. 1. Distribution of polyurethane usage across application sectors(4).

PU is the sole substance that provides a broad variety of chemical and physical qualities that may be tailored to the particular requirements of the final product or usage(5).PU is a comparatively high strength polymer which results in substantial use in construction and infrastructure industries. The most common uses of PU foams in building construction are for gap fillers around doors and windows and for insulated panels, such as wall and roof insulation. PU foams have several advantages when used as insulation, including improved mechanical and thermal performance, energy efficiency, and environmental benefits(6–8). PU is also frequently used as a protective coating in building and construction because of its quick-curing qualities (short gel time), glossy appearance, long product life, and great resistance to harsh weather conditions(6,9,10).PUs and PU-based composites have been effectively used in vehicle seats, bumpers, door panels, ceiling parts, and windows. Moreover, PU cushions are renowned for being comfortable for both drivers and passengers(11).With outstanding performance in biomedical applications, PUs has been a key component of numerous innovative medical devices. PU is the most promising option due to its exceptional mechanical and adhesive qualities, low cost, and exceptional antibacterial and biocompatibility performance. As a result, researchers are very interested in using them in a variety of biological applications, such as ureteral stents, PICC lines, endotracheal tubes, and catheters(12).In recent decades, polyurethane has rightfully dominated the CASE (Coating, Adhesive, Sealing and Elastomer) sector. While polyurethane-based sealants provide exceptionally tight sealing, PU-based adhesives have exceptional bonding strength(5,13).Over the past 20 years, a large number of studies on appropriate materials for coating purposes have been carried out, and polyurethane has continuously shown itself to be an excellent paint and surface coating material(5,14,15).Another significant material type is PU elastomers, which can be utilized for many practical applications, including goggles, ski boots, home goods, shoe soles, and sunglasses(16). They are also utilized to make high-performance industrial machinery parts including belts, gaskets, rollers, and pulleys, as well as thermal breaks in metal frames used in building construction(6).

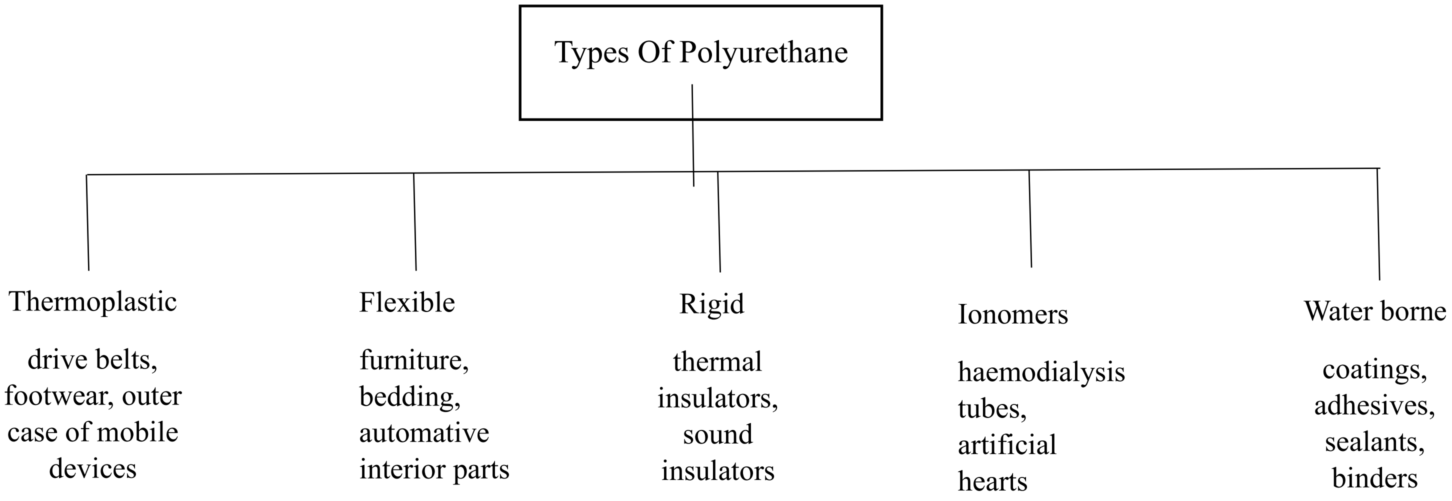


Fig.2. Types of Polyurethane and their applications.

1.1. Chemistry of polyurethane

The main ingredients used in the production of polyurethane are polyols anddisocyanates. The usual synthesis of a PU is based on the polyaddition reaction between a polyol, a molecule with two or more hydroxyl groups, and a different compound with two or more isocyanate groups.Organotin and compounds based on tertiary amines can accelerate this reaction through a step-growth method.In addition, a number of other components—such as blowing agents, surfactants, chain extenders, cross-linkers, fillers, can be added to the reaction system(17,18). The structure-property link between polyol and isocyanate is crucial for comprehending and constructing polyurethane products since changing either of these components can drastically alter the polyurethane's characteristics(5).

Polyurethane macromolecular structure comprises alternating soft and hard segments, each contributing distinct functional properties. The hard segments, primarily formed from polyisocyanates and cross-linking agents,serve as structural support points, enhancing the material's resistance to deformation.Conversely, the soft segments are composed of oligomeric polyols, and their molecular weight and degree of polymerization critically influence the flexibility, elasticity, and overall performance of the polyurethane matrix (19).The polyurethane's ability to be extremely adaptable and efficient for a wide range of applications is attributed to its combination of hard and soft segments(5).

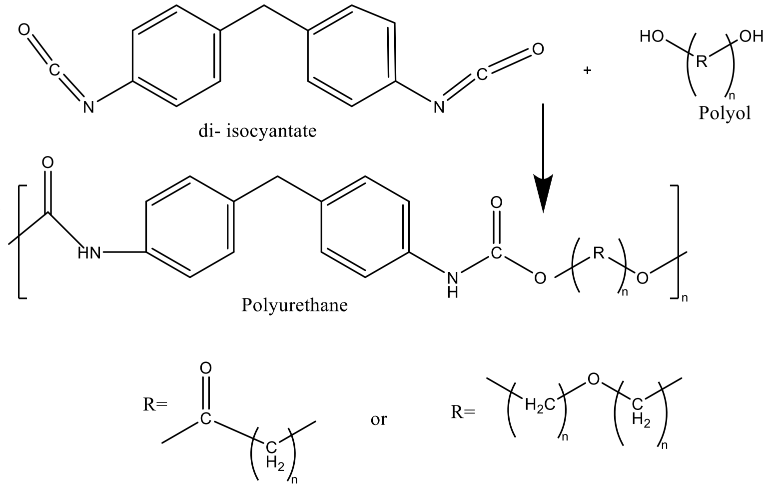


Fig.3:Reaction of polyurethane polymerization(20).

1.2. Environmental Impact of Polyurethane

Every year, more and more polyurethane waste is generated as a result of its many uses and economic success. These wastes include post-consumer (PC) and end-of-life (EOL) products, as well as leftovers from the production of polyurethanes. The latter arises from flaws in production and processing techniques and accounts for as much as 10% of PUs produced(20).Moreover, EOL and PC waste is a far greater issue because they are typically contaminated or distorted, making them less likely to be reused(20,21).

Currently, the most popular method for handling polyurethane waste is landfilling. The percentage of PUs disposed of in this manner approaches 50% of waste (post-consumer or post-production combined) (20,22)The largest issue is polyurethane foams, which also account for the largest portion of production. They have huge volumes because of their low apparent density. Additionally, a large volume of air inside foam cells can obstruct efforts to put out flames and supply oxygen to deep-seated fires. Toxic vapours released from the burning of polyurethane are another risk associated with landfill fires(20).Incineration, while reducing waste volume, releases substantial amounts of carbon and nitrogen oxides(23)Incineration holds a significant place in the recycling of waste made of polyurethane. However, inefficient combustion during the incineration process would result in the production of toxic gas, which will significantly contaminate the atmosphere(24).Beyond the effects that PU manufacture has on the environment, a sizable portion of the PU that are created wind up as improperly handled garbage. There are hazards to the environment and human health from this poorly PU garbage. There are still significant environmental effects from this garbage, even when it is effectively managed in landfills and incinerators(25).

PU presents environmental problems due of its resilience to deterioration and recycling.PU reduces the quantity of trash that is disposed of in landfills by recovering materials and recycling them as building blocks for fresh goods. This procedure lessens the environmental impact of their extraction and preparation while also reducing the requirement for new raw materials. Additionally, there may be chances for cost reductions and/or new sources of income through PU recycling(23).Both the recycling of waste polyurethanes and research aimed at understanding the degradability of polyurethanes are becoming increasingly significant.

2. Methods to recycle polyurethane

The process of recycling is influenced by multiple factors. The following categories ofdegradation are based on those factors:

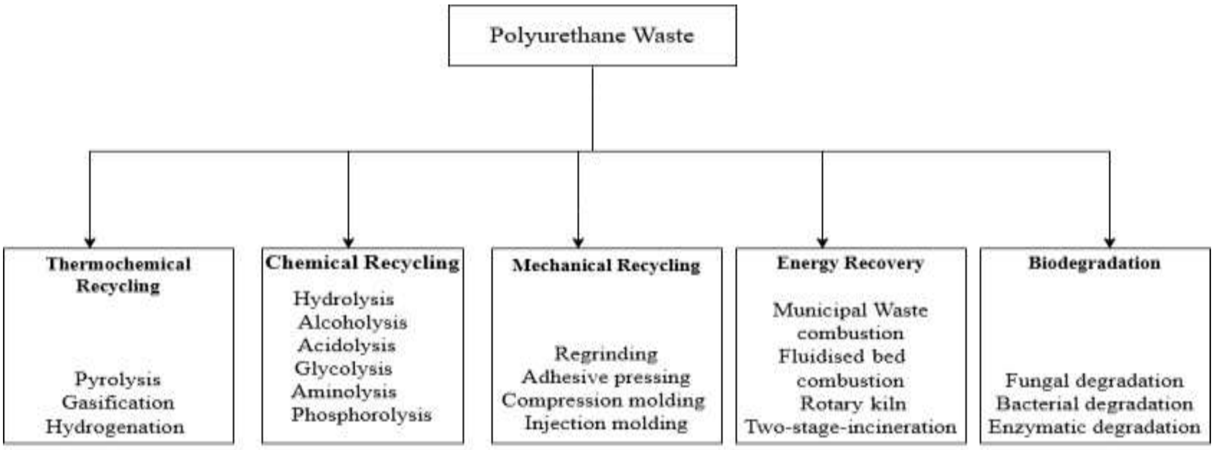


Fig.4. Different methods of polyurethane waste recycling(26).

2.1. Thermo-chemical Recycling:In the thermo-chemical recycling process the PU waste is thermally converted into chemicals for feedstock. The pyrolysis process consists of thermally decomposing the PU waste into gaseous, liquid and solid products in an oxygen-free (or oxygen-lean) environment(27).Partial oxidation is utilized in gasification to create syngas, which is mostly composed of H2, CO, and CO2. This syngas can then be properly scrubbed to yield useful chemicals. Although gasification technologies are widely used and technically advanced, they come with significant energy requirements and high operating expenses(28). Hydrogenation is a process that produces gaseous and liquid products by treating waste with heat and high-pressure hydrogen.These end products can be utilized as chemical feedstock and fuel for additional procedures. Between the effects of gasification (of high-pressure hydrogen) and pyrolysis (of heat), hydrogenation might be viewed as a compromise(29).

2.2. Chemical Recycling: When polyurethane depolymerizes completely to monomers or partially to oligomers and other chemicals, this is referred to as chemical degradation.PU contain a variety of organic and inorganic materials, they are readily broken down by reagents such as water, alcohols, acids, glycols, and amines(30).

2.3. Mechanical Recycling: The breakdown of polymers under mechanical stress is referred to as mechanical degradation. The mechanical deterioration is determined by four main routes. They are injection molding, compression molding, adhesive pressing, and regrinding(30). Polyurethane foams are mechanically recycled by being ground into powders, which enables them to be reused again as filler in new foam manufacture. Reusable techniques include compression molding, adhesive pressing, and flexible foam bonding(26).Adhesive pressing involves applying an adhesive binder to the surface of PU particles and bonding them in a hot press(30).Compression molding is a technique that creates neat particles without the use of extra binders by molding PU particles at high temperatures and pressures of 180°C and 350 bar. Automotive parts made from recycled PU using reaction injection molding have been successfully produced using this reaction(30,31).Through the use of injection molding, cross-linked polyurethane and polyurethane combinations with other plastics—preferably thermoplasts—can be recycled(27).

2.4. Energy Recovery: This recycling method is also known as incineration. In this process, PU waste is burned while oxygen is present. Since it recovers energy from the PU product, it is still preferable to disposing of the material in a landfill(32). Burning 1 kg of polyurethane has a calorific value of about 7000 kcal/kg, this can provide heat equivalent to the energy produced by the same weight of coal(24).Energy recovery is frequently seen as the only appropriate disposal technique regarding recovered substances which have no markets or cannot be made. This is especially true for waste that has PU laminates on wood, leather, textiles, or mixed materials. Additionally, recovery and recycling are extremely challenging for flame-retarded materials. When PU foam is incinerated, its volume is reduced by about 99%, which has a significant impact on lowering the amount of this material that is dumped in landfills while also eliminating CFCs and other dangerous foam blowing agents(33).

2.5. Biodegradation:In recent years, the biodegradation of polymers and the need for biodegradable polymers have arisen due to the overuse of polymer goods(34). The most common definition of biodegradation is "The process that can break down polymeric materials into carbon dioxide, methane, water, inorganic chemicals, or biomass with the aid of certain microorganisms." In biodegradation, fungi, bacteria, and algae are the most crucial species(35).

3. Chemical Recycling of Polyurethane

Chemical recycling alters the chemistry of material.The main goal of PU chemical recycling is to transform waste back into its original basic ingredients, particularly polyols as well as other valuable raw materials. The recuperation of monomers, the principal byproducts of the breaking of urethane bonds in a precisely controlled reaction conditions, is the major goal of chemical recycling.It refers to the complete or partial breakdown of polymers into oligomers and other chemical compounds(36).Two possible bases for depolymerization are the breaking or interchange of urethane bonds or the breaking or interchange of chemical bonds of the materials utilized to make polyurethane, such as the ester groups in polyol(37,38).The potential for recovering monomers and oligomers from polymers is closely linked to the experimental conditions used, including the choice of reactive organic compounds, temperature, pH, pressure, atmosphere, and the types of catalysts employed. The resulting depolymerized substances can be cleaned and distilled to produce monomers, which are basic hydrocarboned units that can be utilized as synthesis materials in the chemical and petrochemical industries. In this manner, high-value items can be produced(38,39).

Currently, mechanical recycling is still the mainstay of the plastic recycling sector today. The industry's interest in chemical recycling has grown in recent years. Plastics that cannot be recycled mechanically can be recycled using chemical recycling, which may also prevent impurity buildup, a common drawback of mechanical recycling(25).

The recycling route that is selected can result in the extraction of products with different physicochemical characteristics and functions.Since polymers are made up of a variety of organic and inorganic materials, reagents such as water, alcohols,acids,glycols, amines, esters of phosphoric and phosphonicacids canreadily cleave them.

Consequently, the chemical recycling is classified according to the reagents utilized as follows (36):

1. hydrolysis
2. alcoholysis
3. acidolysis
4. glycolysis
5. aminolysis
6. phosphorolysis

3.1. HYDROLYSIS

The process of hydrolysis entails adding water to an acidic, alkaline, or neutral environment to depolymerize the polymer. It is the first recycling method that Ford Motor Company has employed(30,40)It describes the process by which a polymer reacts with water, whether in the form of liquid or via steam, producing a variety of chemicals that include carbon dioxide, polyols, and amine intermediates(38).

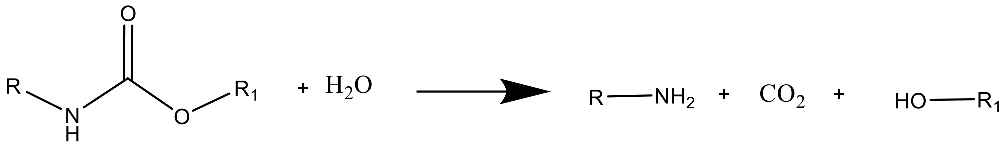


Fig.5. Hydrolysis reaction of polyurethane (20).

Hydrolysis can be carried out with bases (like sodium hydroxide).Waste polyurethane (PU) was efficiently degraded into polyols and benzene derivatives via NaOH-catalyzed hydrolysis in an EtOH-H_2O mixed solvent. A specific ethanol/water ratio (6:4) played a crucial role in both degradation efficiency and product separation. Complete degradation was achieved at 80 °C with a NaOH concentration of 0.2 g/mL. The optimized conditions enabled rapid and effective PU breakdown(41). Efficient hydrolysis of PU was achieved in high-temperature water under high CO_2 pressure, utilizing environmentally benign conditions. The in-situ formed carbonic acid from CO_2 and H_2O acted as an effective acid catalyst, promoting the reaction(42).These polyols can be used as fuel or added to the original polyol to make PU. Additionally, the recovery of the initial isocyanates is made possible by amine intermediates following phosgene treatment. Hydrolysed polyols and isocyanates react to produce the polyurethanes that comprise the inset(20)

Hydrolysis has disadvantages because it is performed under extreme operating conditions, such as high temperatures (even 300–400 °C) and pressures (typically 15–50 atm)(43). Compared to methanolysis and glycolysis, hydrolysis proceeds more slowly because water is the weakest nucleophile among the three depolymerizing agents (methanol, ethylene glycol, and water)(40).

Table 2:Overview of hydrolysis processes applied to various polyurethane (PU) wastes

Typeof PU waste	**Chemicals Used**	**Catalyst**	**Time**	**Temp(°C)**	**References**
Waste PU	Ethanol and water	NaOH	2h	40-80	(41)
PU	Water and High Pressure of CO2		24h	190	(42)
PU Foam	Superheated Steam		6h	288	(43)
PU Foam	Superheated Water		30min	249	(44)

3.2. ALCOHOLYSIS

It is the recyclingof polymers caused by low molecular weight alcohol and a catalyst. Polyurethane degraded into a low molecular mass liquid at a particular temperature.Here, the short reaction time and low temperature result in a high deterioration efficiency. The most crucial aspect is that the items don't require complicated processing afterward. It can be utilized straight away to make PU items as a raw material(24). The basic premise is that the alcohol hydroxyl group and polyurethane chain segment on the ammonia ester bond alcohol exchange process cause the polyurethane molecular segment to "crack" into relatively small molecular weight alcohol substances (regenerated polyether polyol)(45).

PU foam waste was recycled via alcoholysis using ethylene glycol (EG) and propylene glycol (PPG), with various catalytic systems. A synergistic system combining NaOH and duplex metal catalysts (DMC) showed the highest degradation efficiency (46). Another study shows, waste rigid polyurethane foam (RPUF) was efficiently recycled through alcoholysis using ethylene glycol and catalyzed by the metal–organic framework MIL-101(Fe). Under optimal conditions (195 °C, 4 h, 5 wt% catalyst), the process yielded up to 78.3% regenerated polyols with a hydroxyl value ≥270 mgKOH/g and stable viscosity around 670 mPa·s. MIL-101(Fe) facilitated selective urethane bond cleavage through domain-limited catalysis while minimizing side reactions. The recovered polyols were reused to synthesize RPUFs that exhibited excellent mechanical properties, including compressive strength between 0.257 and 0.355 MPa, low water absorption (0.03–0.08%), and good thermal stability. The catalyst showed high reusability, maintaining over 65% yield after nine cycles. These results demonstrate a promising and sustainable route for high-performance recycling of polyurethane waste (47).

Table 2:Overview of alcoholysis processes applied to various polyurethane (PU) wastes

Type of PU waste	Type of alcohol used	Catalyst	Time	Temp(°C)	References
Waste polyurethane elastomers.	Butanediol (BDO), propylene glycol (PG), ethylene glycol (EG), and monodiethylene glycol (DEG)	KOH	0.5-1 h	180	(49)
PU Foam	glycerol and butanediol	CsOH	2.5h	170	(50)
PU Foam	Ethylene glycol and propylene glycol	NaOH and DMC	2.5h	160	(46)
Rigid PU Foam	Propylene glycol and ethanolamine	KOH	4h	160	(51)
Rigid PU Foam	Ethylene glycol	MIL-101(Fe)	4h	195	(47)

Flexible polyurethane foam was recycled via split-phase alcoholysis using various alcoholyzing agents, including diethylene glycol, ethylene glycol, glycerol, diglycerol, and pentaerythritol. Diglycerol was identified as the most effective, achieving high purity(99%) and yield of recovered polyether polyol. The addition of 2-pyrrolidone as a co-reactant enhanced the depolymerization efficiency and allowed a reduced glycol-to-PU ratio of 0.5:1. The recycled polyol successfully replaced virgin polyol by up to 50% in new flexible PU foams without significant loss in mechanical or thermal performance (48).

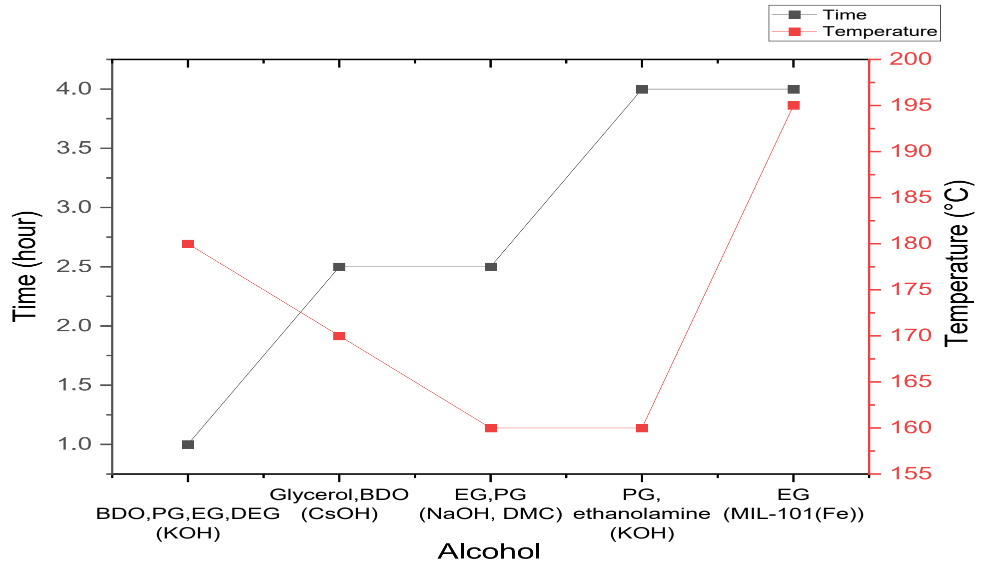
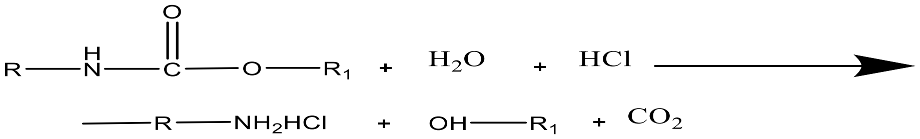


Fig.6.Comparison of reaction time and temperature for alcoholysis of different PU waste materials.

3.3. ACIDOLYSIS

Acidolysis by both organic and inorganic acids is another kind of reaction that has been tried in PU recycling procedures. Inorganic acids break down the urethane bonds in polyurethane, resulting in the formation of amine salts and polyols. This approach is a well-established analytical technique for decomposing polyurethane(30).Inorganic acids like HCl lead to the production of polyols, carbon dioxide, and amine salts. When organic acids, typically with dicarboxylic structures, react with urethane groups, they form amide groups. One of the main advantages of employing organic acids is that they cause little to no aromatic amines to develop in the finished product. For the production of recycled polyols with a low hydroxyl value, which are appropriate for flexible polyurethane applications, acidolysis is particularly helpful. However, glycolysis is a better technique than acidolysis for rigid polyurethane applications when a high hydroxyl value is needed(38,52).



$$R-\overset{H}{N}-\overset{\overset{O}{\|}}{C}-O-R_1 + H_2O + HCl \longrightarrow -R-NH_2HCl + OH-R_1 + CO_2$$

Fig.7Acidolysis of polyurethane by HCL(53)

Organic acids can be classified into two primary categories: unsaturated dicarboxylic acids, like fumaric or maleic acid, and saturated ones, such adipic or succinic acid. Due to saturated acids, chemical breakdown reactions can take place in milder environments with shorter reaction periods and lower temperatures (around 60 °C).This results in the formation of oligomers containing functional groups like amines, amides, urea compounds, or hydroxyl groups. In contrast, using unsaturated dicarboxylic acids leads to oligomers with double bonds, which are suitable for making adhesives(38).

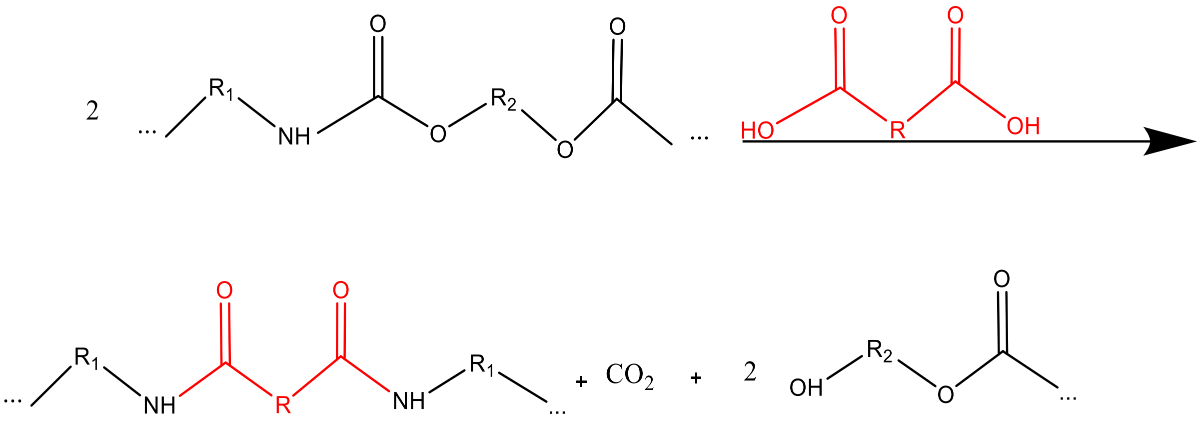


Fig.8.Acidolysis of polyurethane by dicarboxylic acid(29).

Acidolysis of polyurethane foam waste using succinic and phthalic dicarboxylic acids as cleavage agents was conducted. The reactions were conducted at 190 °C for 5 hours, using a PU to acid weight ratio of 4.6:1 for succinic acid and 3.0:1 for phthalic acid, with a conventional polyol as solvent. Succinic acid was found to be more efficient than phthalic acid as a cleavage agent, yielding polyols with higher hydroxyl values and significantly lower viscosities, which indicates a higher extent of depolymerization. The resulting products contained functional groups such as hydroxyls and aromatic moieties, making them suitable for use in high-performance polyurethane wood adhesives(54).

Another study examined how PU to succinic acid ratios (4:1 and 5:1), temperatures (190 °C and 200 °C), and times (4.5 and 5.5 h) affect polyol recovery via acidolysis. An increase in temperature and reaction time generally resulted in higher hydroxyl values (OH number) and lower acid values (AV), indicating more extensive depolymerization and consumption of the acid. Notably, a higher PU/acid ratio (i.e., using less acid) also led to higher OH numbers and lower AVs, which was contrary to initial expectations. This suggests that a smaller amount of acid is sufficient to effectively cleave the PU chains under the tested conditions. Among the variables, temperature had the most significant impact on the OH number, followed by the PU/acid ratio and reaction time. An inverse relationship was observed between hydroxyl and acid values (55).

Table 3:Overview of acidolysis processes applied to various polyurethane (PU) wastes

Type of PU waste	Type of carboxylic acid used	Ratio (PU/ Carboxylic acid)	Time	Temp (°C)	References
PUF Scraps	Succinic Acid	4:1, 5:1	4.5-5.5 h	190-200	(55)
PUF Scraps	SuccinicAcid/ Phthalic Acid *(R-100Suc-0Pht R-50Suc-50Pht R-0Suc-100Pht)	4.6:1 4.2:1 3:1	5 h	190	(54)
PUF	Maleic Acid	1:0.5	15 min	175	(56)
Flexible PUF	Adipic Acid	1.1:1 2.0:1 3.0:1	15-40 min	210-230	(57)

PUF	Succinic Acid (Zinc Acetate-Catalyst)	5:1	3 h	200	(58)

*(R-XSuc-YPht): X is the molar ratio of succinic acid utilized, and Y is the molar ratio of phthalic acid utilized (54)

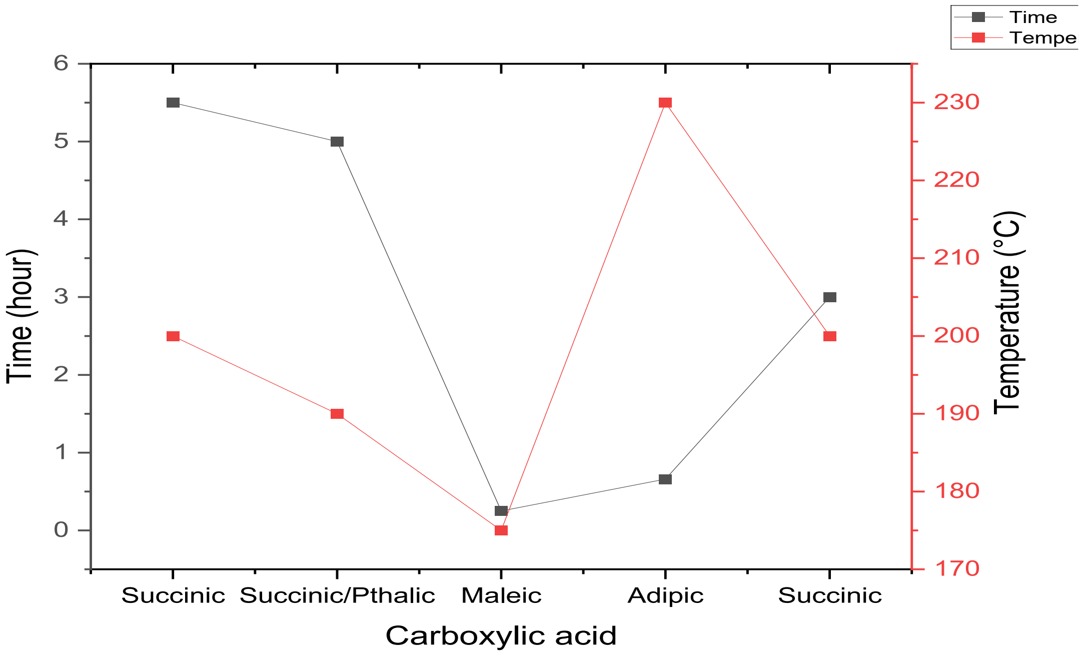


Fig.9.Comparison of reaction time and temperature for acidolysis of different PU waste materials.

3.4. GLYCOLYSIS

Glycolysis is the chemical recycling technique that is predominantly employed for polyurethane waste. Recovering the valuable monomers (polyols) from waste PU in order to produce fresh material is the main objective of this technique. Glycolysis is the process of heating ground polyurethane waste between 180 and 220°C when high-boiling point glycols, including diethylene glycol (DEG), and a catalyst are present.As destructive agents, glycols bind to the functional groups of urethane, break the bonds, and release amines and polyols(59).Glycolysis is based on a transesterification reaction, where a hydroxyl group from glycol takes the place of an ester group that contains a urethane bond's carbonyl carbon(20). The process involves two primary reactions: the breakdown of the crosslinking allophanate and biuret groups and the transesterification of urethane and urea linkages.Glycolysis of polyurethanes can yield the original polyols or glycolysates, which are oligomer molecules that resemble polyols in structure but frequently have a higher molecular weight and urethane links in their chain.Glycolysates include low molecular weight urea molecules, amines, mono- and dicarbamates, and urea oligomers according to their chemical structure(29).The following benefits make glycolysis the preferred method: low reaction temperature, short glycolysis duration, and the glycolysis product's potential for new PU production(45,60).

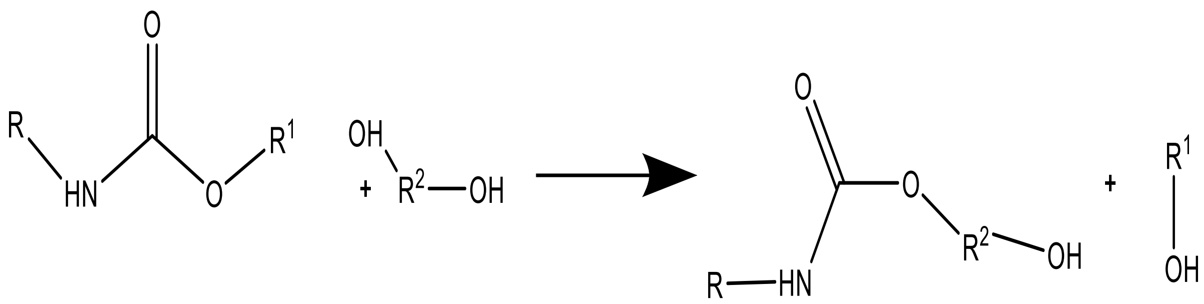


Fig.10. Reaction of glycolysis of polyurethane(3).

A single-phase or two-phase glycolysate is produced when glycol and foam waste are combined in the ideal ratio and heated to a high enough temperature. Glycolysates produced by the glycolysis process with mass excess of polyurethane waste are primarily homogeneous, purification and distillation of the excess glycol are not necessary. As part of the combination of polyols, the resulting semi-products can be used straight in the formation of polyurethanes. An adequate mass amount of glycolysis agent can limit the length of the oligomer chains since the molecular weight of oligomers decreases as the mass of applied glycol increases(29).Glycolysates produced by the glycolysis process with mass excess of glycol primarilyproduces two-phase glycolysate. The upper layer is mostly composed of recovered polyol, whereas the lower layer contains unreacted glycol and undesirable byproducts as urea, aromatic carbamates, oligomers containing urethane links and amines. Recovered polyol makes up the majority of the upper layer, while unreacted glycol and unwanted byproducts such amines, aromatic carbamates, ureas, and oligomers with urethane linkages are found in the lower layer. In certain instances, urea oligomers are also present in the third layer that forms(3).Following reaction completion, the recovered polyols need to be purified and can be directly used to produce new polyurethane materials.The applied glycol, which was used more than the mass of polyurethane waste, must be distilled. Sometimes the upper phase is refined further to concentrate the recovered polyol and reduce the amount of residual amines that affect the reactivity of the final polyol.Ethylene oxide (EO) and propylene oxide (PO) reactions with alkylene oxides are the most efficient way to lower the amine content. Additionally, there are less invasive techniques such as adding small amounts of isocyanate or using epoxidized natural fatty oils. Conversely, the lower phase is considered process waste and is rarely further processed, though it can be utilized in tiny amounts to produce hard polyurethane foams (29).

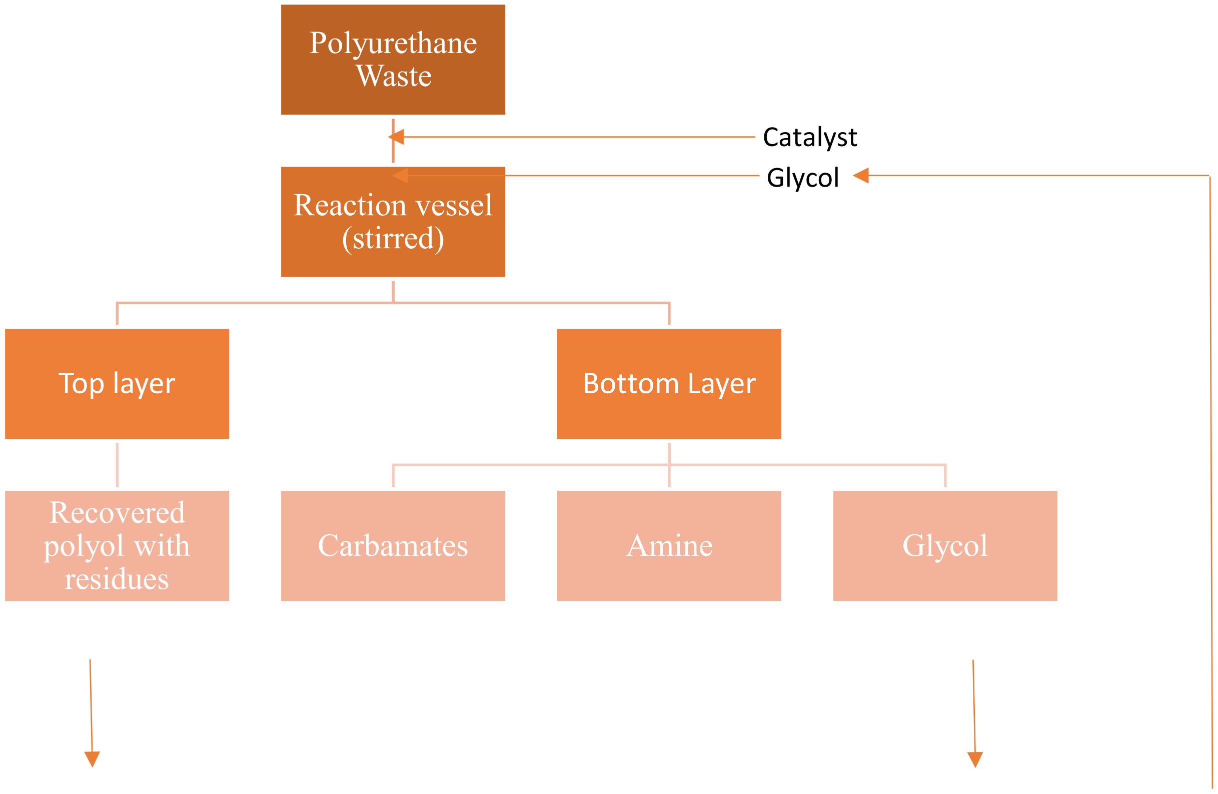


Fig. 11.Schematic representation of split-phase glycolysis.

3.4.1 PU Flexible foam glycolysis

Certain block copolymers make up flexible polyurethane foams, the phase separation between the hard and soft segments gives them their flexibility. While soft segments are pliable and increase the polymer's elasticity, hard segments are rigid structures that give the polymer its hardness(61).Flexible PUF's low-density cellular nature makes it ideal for cushioning, and its flexibility enables a variety of uses(33).

The study investigates the recovery of polyol-rich products through glycolysis of waste flexible polyurethane foam using potassium acetate and diethylene glycol (DEG). 90 minutes at 220°C, PU: DEG = 1:1.5, KAc = 1% by PU mass, and the ideal circumstances produced a single-phase liquid product with a high polyol content. The product's hydroxyl value ranged from 460 to 780 mg KOH/gand reached a stable state after 90 minutes, as validated by FTIR analysis, which also showed that the -NCOO- groups had degraded. Reusability in PU synthesis was shown by the recovered polyols' viscosity, which ranged from 633 to 3601 cps(62).Using DEG and DPG, the glycolysis of car seat polyurethane foam was maximized in this study at 220°C for 2.5 hours. DPG was more useful for reuse since it created a single-phase result, whereas DEG glycolysis produced two immiscible phases. SEC and UV measurements detected trace amounts of aromatic residues and verified the recovery of the original polyols. The DPG and upper DEG phase products have viscosities of about 2000 mPa·s and hydroxyl values of up to 923–1022 mg KOH/g, making them appropriate for the fabrication of new rigid PU foam without sacrificing material qualities(63).Anotherstudy describes a "split-phase" glycolysis of cold-cure polyurethane foam at 170–190 °C with atmospheric pressure, employing glycerin and NaOH (1% w/w) as a solvent and catalyst, respectively, in a 1:1 PU: glycerin ratio. The process yielded three phases, with the upper phase containing the recovered polyol. The structural similarity between recycled and virgin polyol was confirmed by FTIR, ^{1}H-NMR, and ^{13}C-NMR, with only a slight contamination from aromatic amines. The viscosity dropped from 9600 cPs to \~3400 cPs, indicating effective urethane bond breakdown, and the recycled polyol's hydroxyl values were 99 and 73 mg KOH/g after 1 and 3 hours, respectively. Glycerin is a useful glycolysis agent for flexible PU waste recycling because of its inexpensive cost (64).

3.4.2 PU Rigid foam glycolysis

The condensation polymers known as rigid polyurethane have a high degree of cross-linking(61).Rigid polyurethane foams are among the most widely used, adaptable, and energy-efficient insulations. It has been demonstrated that these foams work well as insulation materials that can be used for wall and roof insulationwindow insulation. (16).

A study uses EG, PG, and DEG at 190°C for 5 hours with sodium acetate acting as a catalyst to explore the desaminated glycolysis of MDI-based rigid PU foams. The procedure produced mono- and dicarbamate-type polyols, and ^{1}H-NMR confirmed that hazardous amines were transformed into OH-functional polyols through desamination with butyl glycidyl ether. The recovered polyols had hydroxyl values of 580–605 mg KOH/g, higher than the virgin polyols (420–490 mg KOH/g). PU foams reconstituted with up to 30% recycled polyol had comparable mechanical and physical properties, suggesting their potential for reuse(65).The glycolysis of rigid polyurethane foam (WPUR) from waste freezers is investigated in another study, which uses ethylene glycol (EG) as the ideal glycol and NaOH as the catalyst.A single-phase product was produced after two hours of reaction at 197.85°C under the ideal circumstances of EG/WPUR = 100% and NaOH/WPUR = 1%. The recovered mixed polyol had a hydroxyl value of 847–875 mg KOH/g,its viscosity was about 2100 mPa·s, and its molecular weight was similar to that of commercial polyether polyol. GB/T 26689-2011 criteria were met by regenerated polyurethane foams with up to 10% recycled polyol, exhibiting satisfactory thermal and physical characteristics appropriate for insulation applications(60).

Table 4: Overview of glycolysis processes applied to various polyurethane (PU) wastes

Type of PU waste	**Type of glycol used**	**Ratio (PU: Glycol)**	**Catalyst**	**Temp(°C)**	**References**
Flexible PU foam	Diethyleneglycol	1:1.5	Sn (Oct) $_2$	190	(66)
Flexible PU foam	Diethylene glycol (DEG), Dipropyleneglycol (DPG)	1:1	Diethanolamine (DEA)	220	(63)
Flexible PU foam	DEG	1:1.5	DEA, titanium (IV) butoxide, potassium octoate, Ca (Oc)$_2$	189	(67)
Flexible PU Foam	DEG	1:1.5	Potassium octoate, Ca (Oc)$_2$, Sn (Oct) $_2$	189	(68)
Flexible PU foam	Diethylene glycol	1:1.5	KAc	220	(62)
Flexible PU Foam	Glycerine	1:1	NaOH	170-190	(64)
Flexible PU Foam	Glycerine	1:1	NaOH KOH	Microwave irradiation	(69)
Flexible PU Foam	Crude Glycerol	1:1.5	Stannous octoate	190	(70)

Flexible PU Foam	Ethylene glycol	5:9	$FeCl_3$	160-200	(71)
Flexible PU Foam	HDO	1:1 - 10:1	KAc	230–245	(72)
Rigid PU Foam	Ethylene glycol Diethylene glycol	1:1	NaOH,NaAc, and triethanolamine	197.85	(60)
Rigid PU Foam	Diethylene glycol	1:0.6	Ethanolamine and Zinc stearate	160-210	(73)
Rigid PU Foam	Diethylene glycol	3:1	Ethanolamine and Zinc stearate	180	(74)
Rigid PU Foam	Ethylene glycol (EG) Propylene glycol (PG) diethylene glycol (DEG)	1:0.3-0.4	Sodium acetate	190	(65)
Rigid PU Foam	Ethylene glycol (EG) Propylene glycol (PDO) – Butylene glycol (BDO)	1:1	KOH $Ti(OCH_2CH_2O)_2$ (titanium–ethylene glycol)	180	(75)

3.5. AMINOLYSIS

Aminolysis is a process in which polyurethane foam can be chemically broken down by transesterification using aliphatic diamines or even polyamines with primary and secondary amine functionalities(38). The ester group from urethane is exchanged out for the amine group from ammonia or amine in this process. Depending on the reaction conditions, PUF aminolysis can result in either a complicated mixture in a single phase that is only suitable for less challenging purposes or a low-quality polyether polyol in a split phase that is only suitable for the formation of rigid PUFs(76).

Fig.12.Reaction of aminolysis of polyurethane(30).

Table 5: Overview of aminolysis processes applied to various polyurethane (PU) wastes

Type of PU waste	Type of chemical used	Catalyst	Time	Temp (°C)	References
PU foam	Mono ethanolamine (MEA)	None	3 h	120-150	(77)
Rigid PU foam	Diethylene triamine (DETA)	NaOH	70 min	180	(78)
PU Foam	Hexamethylene diamine	TBD: MSA	24h, 2h	160, 190	(79)
PU Foam	2-(methylamino) ethan-1-ol	TBD: MSA	7h	160	(79)
PU Foam	Diethylene triamine (DETA) Triethylene tetramine (TETA) Tetraethylene pentaamine (TEPA)	None	40 min - 150 min	150-180	(80)
Rigid PU foam	Ethylene diamine (EDA)	NaOH	3	120	(81)

PU shoe sole	1,3-propanediamine and 4,7,10-trioxatridecan-1,13-diamine	NaOH	3h	140	(82)

(MDI)-based polyurethane flexible foams were decomposed using monoethanolamine (MEA) and diethanolamine (DEA) at 150 °C without any catalyst.Two independent layers were formed by the breakdown products: a bottom layer that contained methylene diphenyl amine (MDA) and alkanolamine derivatives, and an upper layer that was composed of relatively pure polyether polyol. Compared to DEA, MEA decomposed significantly more quickly, taking two to three hours as opposed to more than ten hours. Steric hindrance and hydrogen bonding interactions were cited as the causes of this discrepancy(77).Low molecular weight oligomers and 4,4′-methylenedianiline (MDA) were produced by aminolyzing rigid polyurethane foam with diethylenetriamine (DETA) and sodium hydroxide (NaOH) at 180°C for 70 minutes. Increasing NaOH improved depolymerization but reduced MDA yield due to the secondary reactions. NaOH functioned both as a catalyst and reactant(78).Ethylene diamine (EDA) and sodium hydroxide (NaOH) were used to depolymerize waste polyurethane foam by both conventional heating and microwave-assisted aminolysis. Reaction time was greatly shortened (from three hours to thirty minutes) and side reactions were minimized using the microwave approach. Polyester amides were created by reacting the resultant product, which was abundant in hydroxyl and amine groups, with sebacic acid and BHET (from recycled PET). These were further cured with isocyanates (HDI biuret and IPDI) to produce coatings with good mechanical and chemical properties, demonstrating a sustainable route for PU foam recycling(81).

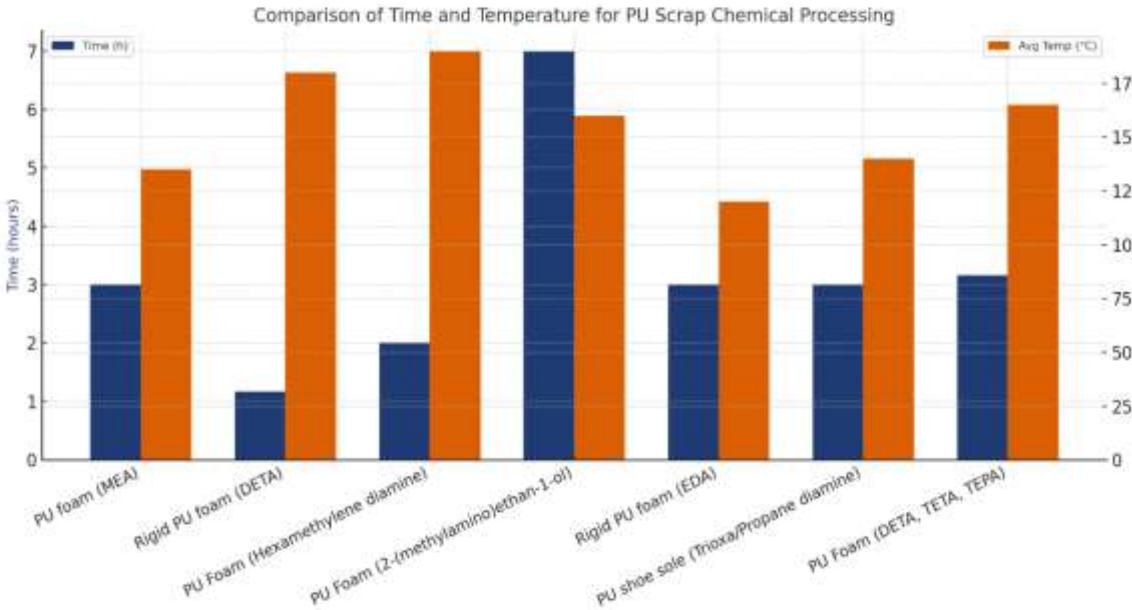


Fig.13. Comparison of reaction time and temperature for aminolysis of different PU waste materials.

3.6. PHOSPHOROLYSIS

Phosphorolysis is a process in which polyurethane waste is treated with esters of phosphoric and phosphonic acids. The wastes are transformed into a liquid product as the result of the interaction between the ester alkoxy group and the urethane group(20).A combination of oligourethanes containing phosphorus was obtained by this technique.New polyurethanes with enhanced UV resistance, adhesive qualities, and flame retardancy can be made with it. The procedure's drawback is that the input materials cannot be recovered, thus they are simply transferred to other production methods rather than being recycled back into the process(20,83)

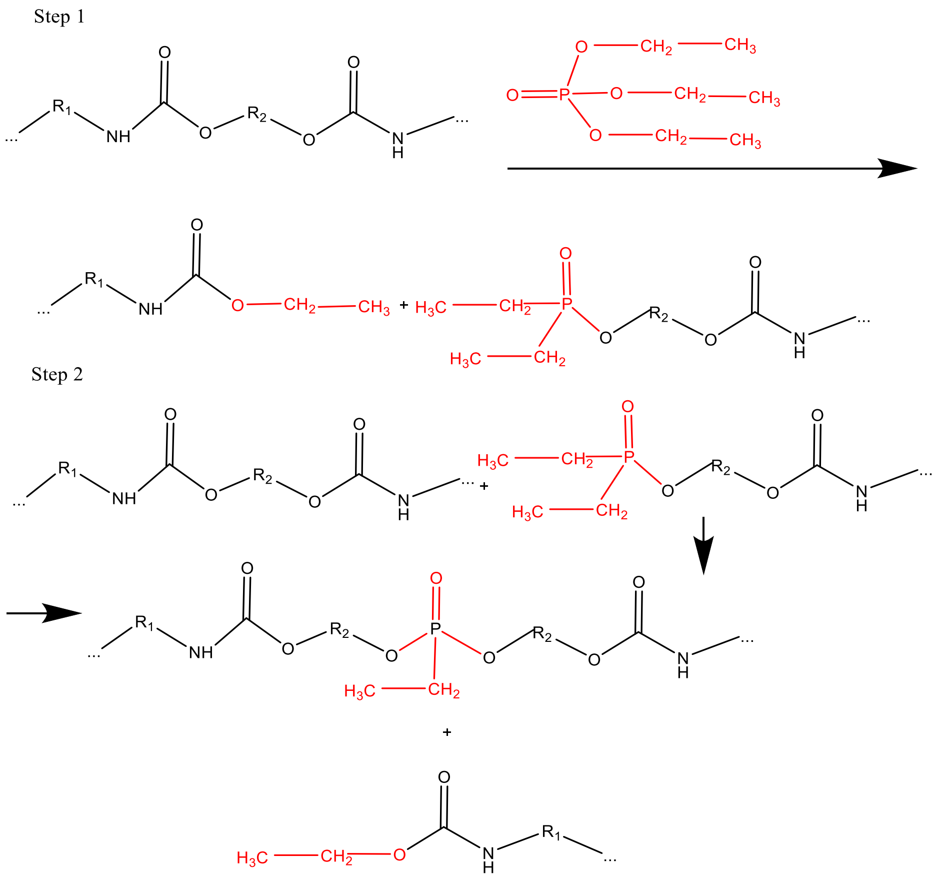


Fig.14.Reaction of phosphorolysisof polyurethane(29)

Table 6:Overview of phosphorolysis processes applied to various polyurethane (PU) wastes

Type of PU waste	Type of Phosphonate ester used(PE)	Ratio (PU:PE)	Time	Temp(°C)	References
Flexible polyether PU foam	Dimethyl phosphonate,diethyl phosphonate	1:3	1-3 h	160	(83)
Flexible polyether PU foam	Dimethyl phosphonate	1:3	1-3 h	160	(84)

Microporous Polyurethane Elastomer	Dimethyl phosphonate	1:2	45 min	142	(85)
Microporous polyurethane elastomer	Diethyl phosphonate and tris (1-methyl-2-chloroethyl) phosphonate	1:3	4-8 h	170, 180	(86)
Microcellular Elastomer Poly(ester-urethane)	Triethyl phosphonate, tris (2-chloroethyl) phosphonate	1:3	5-8 h	180	(87)

Dimethyl phosphonate is used to break down flexible polyether polyurethane foam at 160°C. Degradation produces two liquid layers with oligomers containing phosphorus. Polyether alcohols make up the majority of the upper layer, whilst aromatic and urethane-related chemicals are more abundant in the lower layer. There are nine different degradation products that contain phosphorus(84).Dimethyl phosphonate is used to chemically break down the microporous polyurethane elastomer at 142°C. A single uniform liquid layer including phosphorus-containing oligomers is the end outcome of the degradation. Four main phosphorus-containing compounds that resulted from exchange and alkylation reactions with urethane groups were found(85).

Diethyl phosphonate was used to chemically degrade microporous polyurethane elastomer for 4–8 hours at 170°C, and tris(1-methyl-2-chloroethyl) phosphate was used for the similar duration at 180°C.As a result of the deterioration, the elastomer completely liquefied and oligomers containing either phosphorus or phosphorus and chlorine were formed. The deteriorated components showed promise for repurposing in flame-retardant polymer formulations since they were soluble in ordinary organic solvents(86).

Table 7: Comparative analysis of different methods of PU chemical recycling

Category Of Comparison	**Hydrolysis**	**Alcoholysis**	**Acidolysis**	**Glycolysis**	**Aminolysis**	**Phosphorolysis**
Chemicals Used	superheated steam	low molecular alcohol	inorganic or organic acid	glycols	diamine or polyamine or alkanolamines	esters of phosphonic or phosphoric acid
Temperature(°C)	high temperature	low temperature	medium temperature	medium temperature	low temperature	low temperature
Pressure	high pressure	atm pressure	atm pressure	atm pressure	atm pressure	atmpressure

Product	Polyols, diaminead CO_2	polyols and aromatic compounds	polyols, amides, amine salts, and oligoureas	Polyols and aromatic amines	Polyols, polyamine compounds and aromatic compounds	phosphorus-containing oligourethanes
Large scale application	No	No	No	Yes	No	No

Conclusion

Polyurethane (PU) is a widely used polymer that plays an integral role in everyday applications due to its versatility and durability. Its widespread demand worldwide is a result of its durability and adaptability, but it has also led to a considerable increase in PU waste, raising serious environmental problems. Landfilling and other traditional disposal techniques are becoming less viable because of their ecological impact and incapacity to recover material value. As a result, recycling PU has become more and more important as a step toward the goals of the circular economy and sustainable materials management.

Chemical recycling has shown itself to be a particularly promising method among the other recycling techniques investigated since it can depolymerize PU into its original or modified monomers and oligomers. Several chemical recycling routes were reviewed in this review, includinghydrolysis, alcoholysis, acidolysis, glycolysis, aminolysis, and phosphorolysis. The type of PU waste, the intended end products, the reaction circumstances, and economic considerations are some of the variables that affect the specific reaction processes, benefits, and drawbacks of each approach. Glycolysis is noteworthy for its large-scale applicability and significant industrial implementation potential.
The scalability of chemical recycling solutions for PU nevertheless faces a number of obstacles, notwithstanding the advancements. The various chemical recycling methods need to be optimized for industrial scalability and economic viability, which will require more research. It could eventually emerge as a key tactic for attaining circular and sustainable polyurethane material use.

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