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Review Article

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A Review on Management and recycling of Post-Production and Post-Consumer Polyurethane Wastes

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Abstract

The huge accumulation of post-production and post-consumer polyurethane (PU) waste is responsible water, land and hazard to the environment. The landfills of PU waste releases toxic gases and carcinogens upon catching of fire. Therefore, there is the possibility of converting this waste into valuable products or feedstokes, which can be used for further production of raw materials. Polyurethanes are condensation products of polyisocyanates and polyalcohals commercially known as Polyols. The strength, adhesiveness and insulating properties of polyurethane make it wonderful materials. In this article, the latest developments in the recycling methods of Polyurethane waste were discussed. The study of various methods has shown that recycling of PU waste can be environmentally economically beneficial. Presently, mechanical recycling and glycolysis of PU waste are most accepted methods of processing the PU waste. A biological method of degradation polyurethane waste is most promising, since no harmful chemical are required in this process.

Keywords

Polyurethane,
MDI,
Polyols,
Glycolysis,
recycling

1. Introduction

In recent few decades, the usages of polyurethane (PU) have been increased dramatically due to the urbanization and improved living standards of populations. The physic-mechanical and chemical properties of polyurethane such as adhesion to the substrate, heat resistance, insulation, etc have boosted its market. The estimated demand of Polyurethane in India is about 565,000 million tons, which is increasing year by year with the growth rate of 10-12%, while the domestic production of polyurethane is 150,000 million tons (market and market, 2021). The polyurethane market size was 1779.3 million (USD) in the year 2021 with projected market size of 2619 million (USD) at the rate of 5.8% per year (stastista.com, 2022). According to a report (Lucia Fernandez, 2022) published by Lucia Fernandez in June 2022, the global Polyurethane market volume from 2015-2029 was reported 21.22 million metric tons in 2015 with the projected volume of 29.9 million metric tons in 2029 (figure1). The huge demand of polyurethane worldwide has influenced the world economy dramatically.

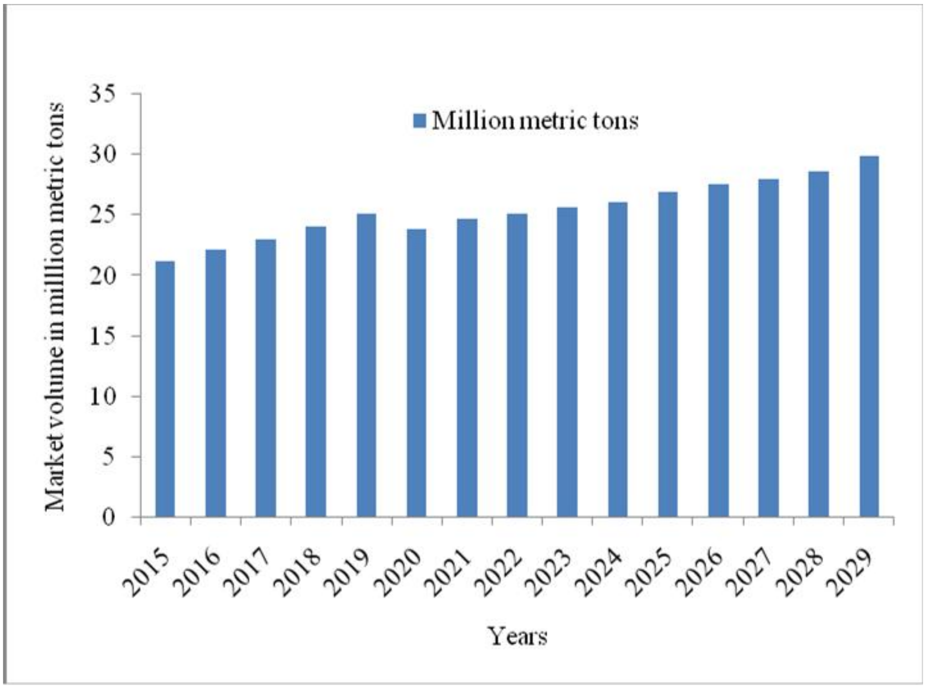

Figure 1. Projected Market volume in million metric tons

The value of global polyurethane market size was reported 72.82 billion USD in 2021 and the expected expansion in its valve is about up to 105.45 billion USD in 2030 at a compound annual growth rate of 4.3% from 2022 to 2030. Keeping the view of sustainability in mind, the insulation of building has escalated the demand of polyurethane during the forecast period. The concept of green buildings is growing rapidly in construction of residential and commercial buildings as the investment in smart energy-efficient systems (https://www.grandviewresearch.com, 2020). This concept has needed the higher demand of efficient insulating materials which can regulate heating, ventilation and air conditioning (HVAC). Polyurethane is best suited for utilization as efficient insulation materials in buildings (Witkiewicz W *et al*., 2006, Wu J W *et al*., 1999). The increasing demand of cold chain has boosted the polyurethane insulation market in India very rapidly. Without such faculties the farmers are dependent on the local market to sell their agro-products, due to which their products rapidly spoils and damaged in humid and hot climate and also the farmers have low price. Therefore, insulation of cold chain and insulated vehicles becomes more widely needed to save the food products and generated higher demands of polyurethane. The end user market of polyurethane includes bedding, furniture, insulation of buildings, automotive and transport industry, refrigerators, foot wear, constructions and others. According to figure 2, bedding (mattress) consumes about 40% of total produced flexible polyurethane foam (Pfa.org, 2021).

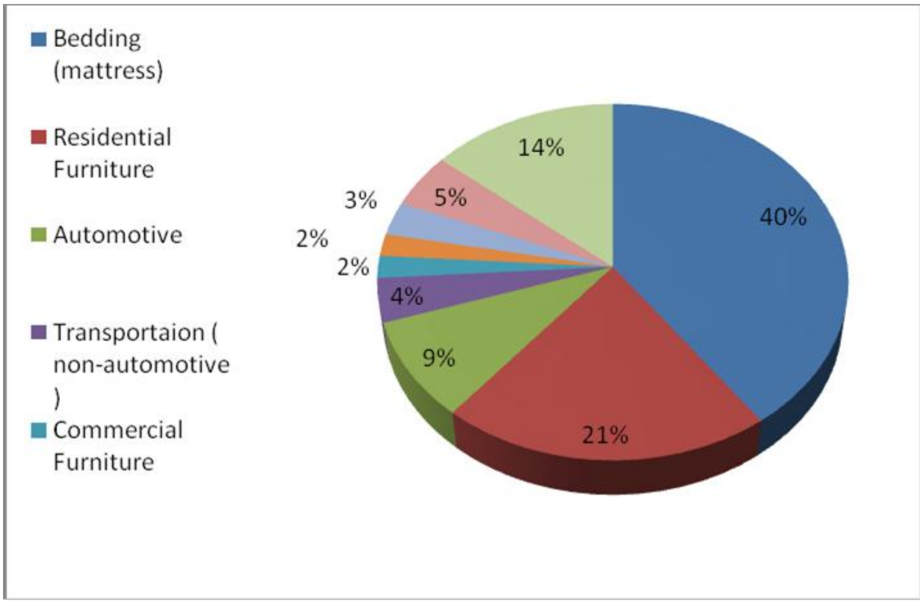

Figure 2. Flexible PU foam use by applications

The higher mechanical strength and easier manufacturing of PU foams make it very suitable to be used as sandwich panels (Gama NV *et al*., 2018). Low thermal conductivity and high durability of PU makes it more saving energy materials used in buildings (Report, 2006). Polyurethane constitutes about 9% share of total production of plastics (Palm, E *et al*., 2018). Due to the high demand of PU because of its versatile utility in various sectors, it generates significant amount of waste in the same ratio as production of PU. Unfortunately, landfilling is the first choice for disposal of PU waste (30.8%0, 29.7% of PU waste is recycled and 39.5% is recovered through energy recovery processes of total waste (Ravindra V. Gadhave *et al*., 2019). Keeping the concept of zero plastic waste and landfilling, the polymeric wastes are used as inclusion compounds as a substitute of aggregate and fine compounds. The polymeric wastes are also used for energy generation as an alternative route to their recyclization. The combustion of PU waste generates harmful gases, which are hazardous to human health and environment (Zia, K.M. *et al*., 2007).

2. Polyurethanes

While working on the polymer fibers, Otto Bayer, a German scientist, invented polyurethane in 1930s. In contrast to the other thermoplasts like polyethylene, polyurethanes includes no polymerization products but are classified as condensation polymers produced by the reaction polyhydroxyl compounds (polyether or polyester polyol; polymeric alcohalwith more than two reactive –OH groups per molecule), such as propylene glycol and polytetramethylene glycol commonly known as polyols and polyisocyanates such as 4,4-diphenulmethane diisocyanate (MDI) or toluene diisicyanate (TDI) in the presence of suitable catalysts, additivesand chain extenders **(Debuissy, T *et al*., 2017a, Debuissy, T. *et al*., 2017b, Simón, D. *et al*., 2018).** More than 70% currently used polyols for the manufacturing polyurethane, are polyether polyols and polyalcohals and these are obtained by the polymerization of propylene and ethylene oxides **(Austin, A *et al*., 2017, isopa.org, 2020).**

$$R-NCO + OH-R \longrightarrow R-NH-COOR$$

Due to the large variation in the starting of polyurethane, this group of polymers would exhibit various chemical characteristics. Therefore by adopting different chemical processing route numerous polymeric materials (such as polyurethane rigid and flexible foams, coating, adhesives, sealants and elastomers) (Magnin, A et al., 2020) can be synthesized which comes under the polyurethane family. Polyurethanes being a condensation polymer, they are not built by repeatable identical monomer units but made up of different segments and these segments are connected with each other's by various chemical bonds.

The PU foams made by water blown process consists of a significant number of urea linkages, formed by the chemical reaction between isocyanate moiety (–NCO) and water molecule, following the reaction between another –NCO and newly created amine group. The unique feature of PU is the presence of repetition of hard segment (isocyanate) and flexible (polyol) segments (Laurichesse, S et al., 2014). This property of PU gives versatility in its applications. The six main areas of their applications are given in table 1.

Table 1. Categories of PU application (Yimin Deng *et al*., 2020)

Categories	Application	Production (%)
Flexible foams	Automobile sector, seatings, mattresses	36
Rigid foams	Household appliances, packaging, insulation boards, etc	32
Elastomers	Medical utility, glues	8
Adhesives and sealants	Casting, sealants	6
coating	Vehicles	14
Binders	Wood board assembling, rubber, elastomeric flooring surfaces	4

The elastomeric PU is having most important commercial applications and it covers up to 70% global PU market while rigid PUF, covers around 26% of global PU market. The oligo-polyols used for production of rigid PU, are highly branched polyols having around 3-8 hydroxyl groups per molecule and the chain of one hydroxyl group is short i.e. equivalent weight is low . (D.J. Sparrow *et al*., 1987, T.H. Ferrigno , 1967, M. Szycher, 1999, W. Green *et al*., 1984). This is why; such polyols produces highly crosslinkedand very rigid PU by reacting with crude MDI or polymeric MDI (Mihail Ionescu, 2016).

The objective of this review article is to bring the attention on various technologies presently available and upcoming methods for the commercial application of recycling of post-consumer and post-production of polyurethane waste. An overview is given on the mechanical, chemical and biodegradation of PU waste in this article. A brief introduction on the usages of recycled PU is also given in this review.

3. Recycling and Waste Management of Polyurethane

Huge amounts of post-consumer and post-production of polyurethane waste are generated each year, as the demand of PU is increasing year by year, due significant application and commercial success of polyurethane. Post-production PU waste is the result of imperfection in processing methods and it make up to 10% of

PU produced (Simón, D *et al*., 2015). However post-consumer PU waste is big challenge to their disposal and recycling, because they are highly contaminated and less prone to being reused (Cregut, M *et al*.,2013).

3.1 Landfilling

Landfilling is first choice in many countries till now for the disposal of PU waste. As PU foam share major portion of polyurethane production, it creates big problem to the environment. They have large volume and acquire large area of land, due to their low density.

Because of cell structure in PU foam, the air contained in cells provides sufficient space to oxygen, which catches fire, which releases toxic fumes. The raw material TDI and MDI are themselves hazardous to the environment, as they release carbon monoxide and oxides of nitrogen. Therefore, landfilling is not a proper method for disposal of waste, it should be considered as temporary storage or stock for further recycling processes.

The recycling process of polyurethane can be divided into mechanical recycling, chemical recycling and energy recovery. Chemical recycling alters the chemical structure of the materials, which produces valuable materials. Energy recovery means incineration of PU waste, decomposition and the combustion of products.

3.2 Mechanical recycling

It is the easiest and most popular method of recycling and reuse of waste, in which collected solid waste is converted to flakes, powder or pellets. Mechanical recycling involves grinding, adhesive pressing, compression moulding and bonding of PU waste. The major sources of raw materials for mechanical recycling of PU waste includes post-production scrap of factories and also post-consumer discarded products. The fine powder can be produced by using two-roll mills. The powder produced by the mechanical recycling of PU waste can be used directly as filling for pillows, toys and can be used as filler for the production of polyurethane foams or elastomers by adding it into polyols. Different milling and knife cutting processes are employed to produce the particle size between 100 to200 µm. Shredded polyurethane foam (PUF) wastes can be rebound using pressure or heat. The density of the rebounded product is higher than the of original source material. This rebounded product is used as floor covering and sports matting due its flexibility and durability. About 90% floor covering are produced by this rebonding method (Nestle, D.E, 1994).

The PU waste can be rebounded without using any adhesive. In this process, fine particles of PU foam are condensed under high pressure in hot condition (180$^{\mathbf{0}}$C and 350 bar). This method is mainly employed on the recycling of rigid PU foam waste recovered from bumpers and side panel of automotive vehicles. These hot compressed newly formed materials have rigid structure and three- dimensional networks are formed when glassfiber is reinforced with it. The main disadvantage of this technique is that the raw materials painted and also it is difficult to shred very hard raw material.

3.3 Thermochemical Recycling

In thermochemical recycling of PU wastes, the waste stream is used as a source of energy, fuel or monomers of some value.

3.3.1. Pyrolysis

In this process, the thermal decomposition of polymeric compound into simpler molecules occurs in anaerobic condition at high temperature, producing oil, gas and ash[29]. The pyrolysis of polyurethane waste completes with the decomposition of two segments; decomposition of polyol and decomposition of isocyanate part. In the first stage the urethane bond breaks down and 50% mass of the polymer is lost and this process occurs between the temperature ranges from 100 to 300^{0}C. The decomposition of polyol parts is processed in the range 300 to 800^{0}C temperature. 3% ash of PU waste is remained in the pyrolysis of polyurethane waste (Jomaa, G *et al*., 2015).

3.3.2.Gasification

It is a highly exothermic reaction and produces a mixture of gases mainly carbon monoxide and hydrogen, heat and ashes. The gasification of polyurethane waste does not require segregation of waste. In this method, a waste stream is heated and passed air through it, which forms a mixture of carbon monoxide and hydrogen known as syngas. This syngas is used as source of energy and raw material for the synthesis of ammonia, methyl alcohol, carbohydrates, acetic acid, etc. However, the gasification of PU waste produces toxic gases like hydrogen cyanide and nitrogen dioxide, the emission of these gases can be reduce to some extent by using suitable catalyst (Guo, X *et al*., 2016).

3.3.3. Hydrogenation

Hydrogenation process is similar to both pyrolysis and gasification. In hydrogenation high pressure hydrogen gas is used while in pyrolysis inert gases are utilized. Hydrogenation process produces gases and oils with greater purity through the combination of heat, pressure and hydrogen as compared to pyrolysis. These products can be used as fuel and chemical feedstock.

3.4. Chemical or Feedstock Recycling

In chemical recycling, PU waste is converted into simpler molecules and other value added products, which are act as feedstock for the manufacturing of new products. The main aim of chemical recycling of PU waste is the recovery of monomers by breaking down the urethane linkage under controlled chemical reactions. Chemical recycling is more demanding technique in the comparison of mechanical recycling, due to the production feedstock and less energy is required in chemical recycling. It includes mainly hydrolysis, glycolysis, aminolysis, phosphorlysis of PU waste.

3.4.1. Hydrolysis

Hydrolysis process was started in the 1970s in USA for recycling of polyurethane waste especially flexible foam by applying superheated steam at 200^0C. This reaction occurs between PU wastes and water either in the form of liquid or steam resulting in the formation of polyols, amine and carbon dioxide (Gerlock, J.L *et al*., 1980). Figure 3 shows the schematic representation of polyurethane hydrolysis (Aleksandra Kemon *et al*., 2020).

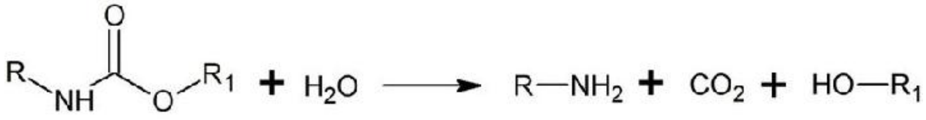

Figure 3. Schematic representation of polyurethane hydrolysis

This method may be employed for recycling of both post-production and post-consumer PU wastes (Zia, K.M *et al*., 2007). Hydrolysis of PU wastes is carried out in an anaerobic condition at elevated temperature (150-320^0C). The resulting polyols can be utilized as either additives to the original polyols or as fuel. The starting isocyanate is recovered after the phosgene treatment of amine intermediates produced in hydrolysis of PU waste. The requirement of high energy input either to heat up the batch or to apply high pressure, makes the hydrolysis of PU wastes uneconomical, this is why this technology has not started at commercial scale (Simón, D *et al*., 2018).

3.4.2. Hydroglycolysis

In this method of recycling of polyurethane wastes, water and glycols are combined under less demanding process conditions, in the presence of lithium hydroxide at 200^0C temperature.

The advantages of this technique are the contaminated and heterogeneous raw materials can be used for recycling purposes, but due high cost, this method is not employed industrially. The resulting polyols can be used in manufacturing of polyurethane using 50% virgin starting materials (Zia, K.M *et al*., 2007, Simón, D *et al*., 2018, Datta, J *et al*., 2017).

3.4.3. Aminolysis

In aminolysis method, an interchange is involved between the amine group from ammonia or amine and ester group from urethane. Therefore it is transesterifcation reaction as shown in figure 4 (Aleksandra Kemon *et al*., 2020). The mechanically grinded PU waste is dissolved in solvents like cyclic ether nitrogenous chlorinated hydrocarbon, which is subjected to aminolysis using a compound containing atleast one amino group at 80 to 190^{0}C temperature, in the presence of inert gases.

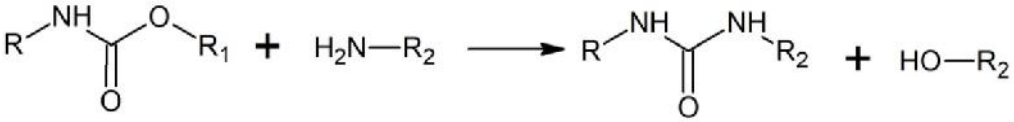

Figure 4. Schematic representation of Aminolysis of Polyurethane

Sodium hydroxide, sodium methoxide and aluminium hydroxides are generally used as catalyst in aminolysis of PU (Sheratte, M.B., 1978). Aminolysis is utilized for the recycling of PU foams and was not developed for the recycling of PU coating, adhesive, sealants and elastomers. This technology is limited to the only laboratory (Simón, D *et al*., 2018).

3.4.4. Phophorolysis

In this method, PU is processed with esters of phosphoric and phosphonic acids which converts wastes into liquid products through the reaction between the urethane group and ester alkoxy group. A mixture of phosphorus containing oligourethanes was obtained in this method as shown in figure 5 (Aleksandra Kemon *et al*., 2020). The new polyurethane with improved flame retardancy, adhesiveness and UV resistance can be obtained by this technology. There is no progress was reported since 2013 and no industrial applications are noticed (Simón, D *et al*., 2018).

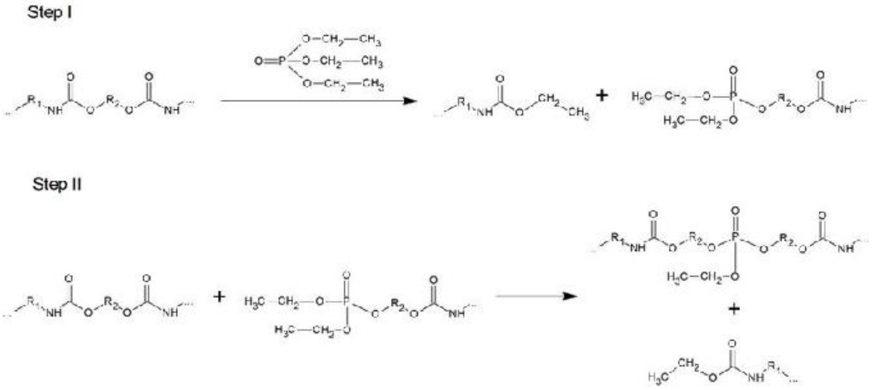

Figure 5. Schematic representation of phosphorolysis of polyurethanes

3.4.5. Glycolysis

Glycolysis is the most widely accepted method for chemical recycling of polyurethane waste either rigid or flexible foam. This technique was developed in the early 1980s mainly in USA and few countries of Europe with aim, the recovery of polyols from PU waste. In this process ester group containing carbonyl carbon of urethane linkage was repalced by hydroxyl group of glycol through the trans-esterification mechanism. The polyols produced in this reaction can be utilized in the manufacturing of polyurethanes, as they exhibited properties similar to the original polyol (Zevenhoven, R., 2004, Simón, D *et al*., 2015). In this method, pre grinded PU flakes were heated to 180-200^{0}C in high boiling glycols generally diethylene glycols with co reagent diethanol amine. At low temperature, the yield of reaction is very low, while at high temperature, above 220^{0}C the undesired side reactions towards amines are observed. Generally glycol/PU waste ratios 60:40 are taken for the glycolysis of PU. The glycolysis of PU can be studied through two approaches; a single polyol is recovered and second approach is that in which flexible and rigid polyol components are recovered (Scheirs, J., 1999). The second process is known as split-phase glycolysis (SPG), in which scraps of PU foam (MDI based) is reacted with diethylene glycol producing a mixture of having two phases. Schematic representation of SPG process for recycling of PU waste is given in figure 6 (Held, S. *et al*., 1999, Ron Zevenhoven, 2004). The lighter layer consists of flexible polyol, from the heavier layer MDI derived compound are recovered, which are converted into rigid polyol by using propylene oxide. This process is very sensitive to contamination of styrene-acrlonitrile (Scheirs, J., 1999, Held, S. *et al*., 1999).

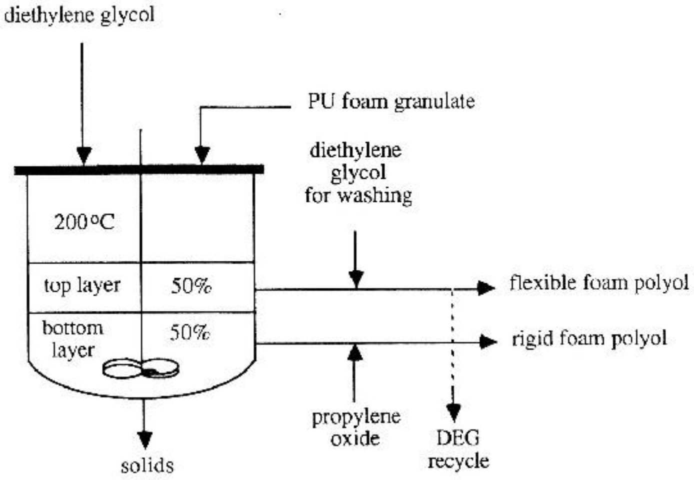

Figure 6. Schematic representation of SPG process for recycling of PU waste

3.5 Energy recovery

The PU wastes which are not suitable for any recycling process discussed above, can be processed for energy recovery by combustion and incineration processes. This method is employed in cases where PU wastes are contaminated, foam permanently linked with wood, leather or fabric and foam is free with Freon. The 99% volumes of landfill with PU waste are reduced by combustion and incineration.

The main hurdle in the energy recovery by the combustion process of PU waste is the presence of flame retardants, which is used in PU foam keeping in view the idea of costumer safety. Furthermore, at high combustion temperature PU releases toxic gases and carcinogens, because polyurethanes bear very much toxicity during thermal degradation in presence of air/oxygen (Paabo, M. *et al*., 1987).

In the next article the recycling of PU waste by biological degradation will be discussed. In the biodegradation process of polymers, the breakdown of complex polymeric chain into simpler molecules occurs by the action of living organism itself or by their enzymes under either aerobic or anaerobic conditions depending on the chemical composition and constituents of raw material.

4. Conclusion

There are many techniques are available to convert waste into useful material or feedstock for further manufacturing of new material, but they have a need of some improvements. Even today's landfilling is most applied way to dispose of PU waste, however, it is not environmentally friendly due to fire of waste, it consume a large volume of land and landfilling is also not suitable economically. The landfilling of PU waste should only be used as stock for short period, not for a permanent solution. Mechanical recycling is the second most accepted method for recycling of PU waste. It is not so costly method but have many limitations. The resultant products of mechanical recycling of PU waste is much cheaper than the original one. The glycolysis and gasification methods are implemented on industrial scale for chemical or feedstock recycling of PU waste while others are remained limited to only research stage in laboratory. Biological degradation does not require any harmful chemical but it is time taking process, but biodegradation is most promising due to the availability of wide range of modifications.

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