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### REVIEW ARTICLE

## OVERVIEWS ON RECYCLING OF POST-CONSUMER PET BOTTLES AND APPLICATIONS OF rPET

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### Abstract

In this review the various aspects related to the recycling of Waste PET bottles, their adequate methods of disposal and management are focused. PET is not as harmful as other polymeric waste but its huge volumes is a big threatening to environment, due non-biodegradability of it, contaminates soil, water and also becomes habitat of rodents/insects. These problems make necessity to recycled PET. Advanced technologies and methods of recycling of Post-consumer PET bottles, and their utilization after recycling, are discussed. This review summarizes the efforts of researchers on the proper utilization of rPET in road pavement, textile, concrete and feedstocks of many valuable chemicals, which has significant effect on environment pollution. The industrial utilization of rPET is cost effective, it is cheap as compared to virgin PET.

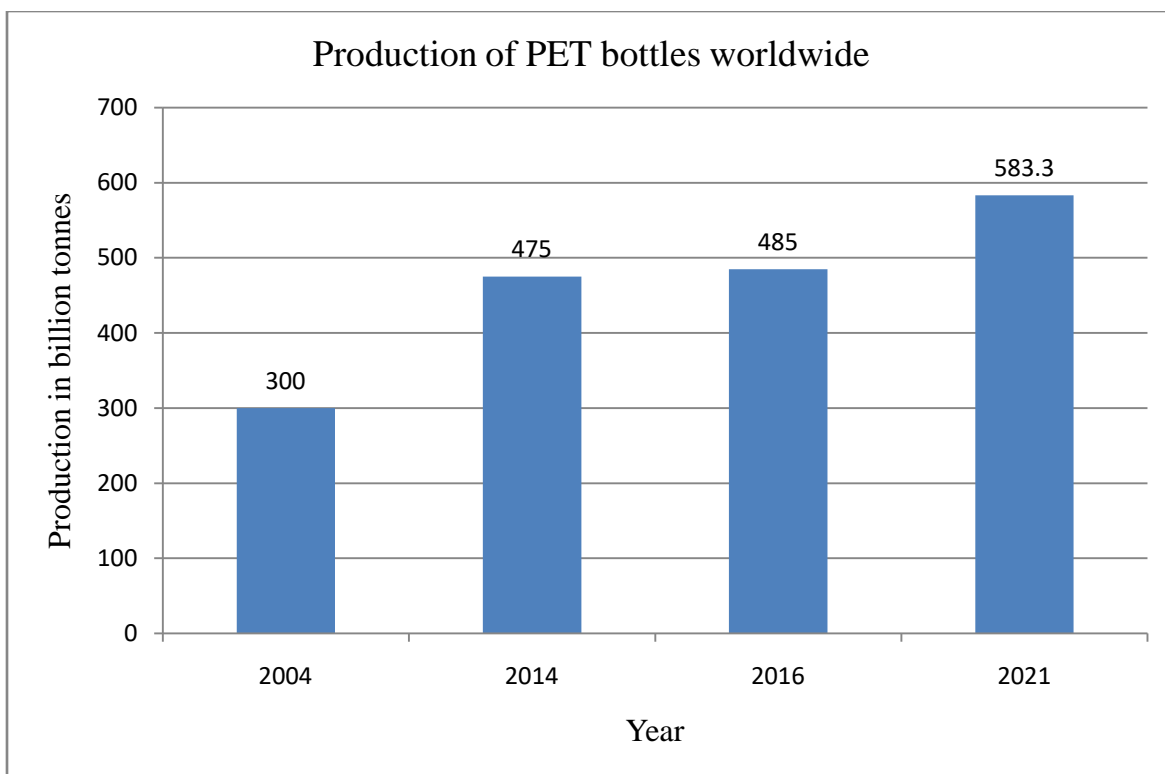
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### Introduction:-

The global production of polyethylene terephthalate (PET) was 485 billion tonnes in 2016 and the expected increase of their shall be reached up to 583.3 billons by the year 2021 as per the statistics published by Ian Tiseo<sup>1</sup>, Global PET bottle production 2004-2021 as shown in figure 1. Hygienic concern after covid-19 pandemic, led the 4% growth in 2020 in packaging industry as compared to 2% growth in 2019 as shown in figure 2<sup>2</sup>. Worldwide 7.27 % plastic wastes are generated by plastic bottles, food and beverages packages generate 31.1%, 11.8% generated by plastic bags and 15.5% plastic wastes are generated by the caps of bottle and containers.

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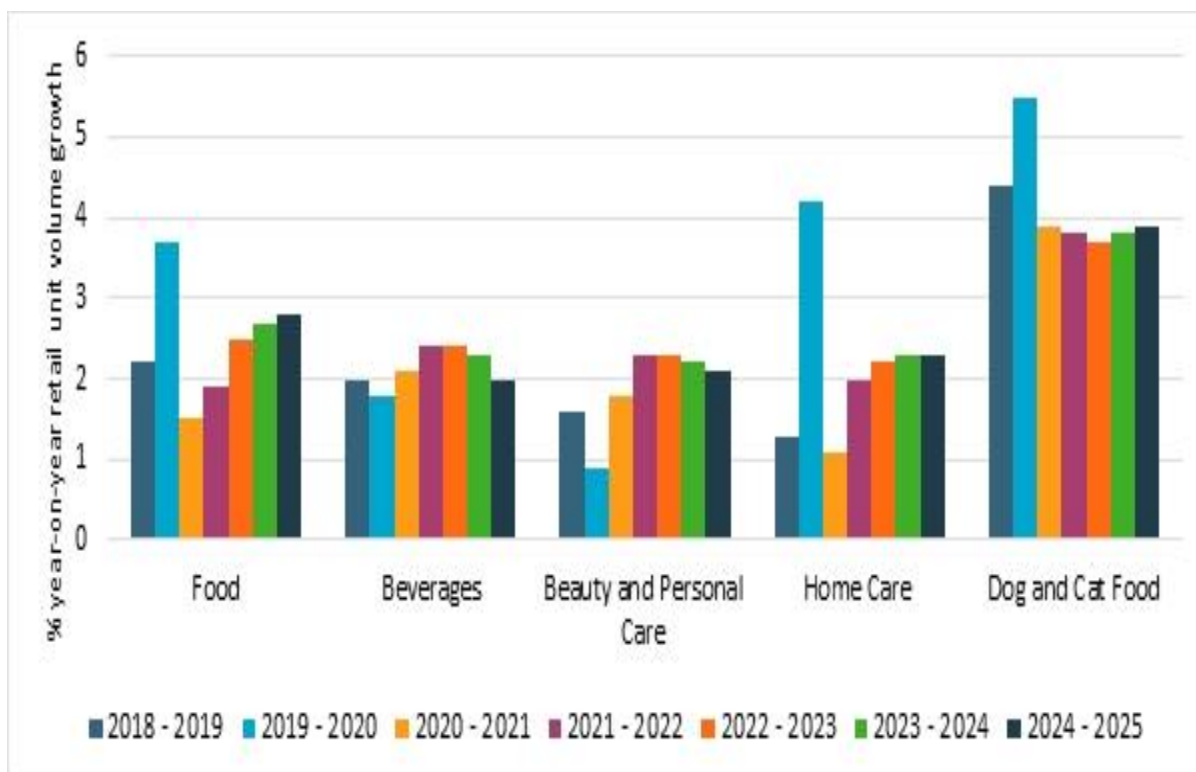
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**Fig.1:-** Depicts the worldwide production of PET bottles from 2004 to 2016 and forecasted for 2021.

India has import 12000 tonnes of PET bottles and flakes in 2016-17, which was increased to 48,000 tonnes in financial year 2017-18 at the growth of 290%. India consumes about 13 million tonnes of plastic, out of which only 4 million tonnes are recycled; this lack is mainly due to the mismanagement of plastic waste collection and an efficient waste segregation. Besides increasing the number and efficiency of domestic recycling and collection units, the government had banned the import of plastic waste, particularly PET bottles in 2015. In 2016, such imports were allowed only for industries which are situated in special economic zones. After that an expert committee of the ministry of Environment had recommended to a firm for importing of PET bottles and flakes up to 50% of their production capacity. In India, 15,342 tonnes of plastics waste is generated every day, out of which 6,000 tonnes of plastics waste remains uncollected and littered inadequately<sup>3</sup>. In India, 65% of PET bottles are recycled by registered firms, 15% are recycled by unorganised sectors and 10% bottles are reused at homes, this recycling rate is much higher than recycling rate for PET in Japan (72.1%), 31% in USA and 48.3% in Europe. By 2025, it is expected that the worth of India's waste management sector will be US\$13.62 billion with an annual growth rate of 7.17 percent<sup>4</sup>. Plastic materials are having very diversifying applications such as packaging, health products, building and construction, agricultural sectors, leisure and sports equipment or medical products, electrical and electronic equipment, automotive and aeronautics, and many other sectors<sup>5</sup>. Containers and packaging sectors consume highest portion of plastic production but they are also used in durable products like furniture, building materials, etc<sup>6</sup>.

The diversity in the application of plastic is due to their special features like low density, light weight, easy processing, chemical resistance, good mechanical properties, electric insulation and low cost. These properties of plastic make them highly competitive in the market and increasing the production rate day by day. These rapid productions of plastics are making abundance of post-production and post-consumer plastic products which led to the threatening of environment with both production and disposal. The production of plastic involves huge consumption of non renewable fossil fuels and high energy, while the abundance of post-production and post-consumer plastic products leads to disposal problems causing hazards to the environment. A plastic persists in the environment for hundreds of years and pollutes the water, soil, air, ocean and others. With increasing of demand of plastics, the issues related to plastic wastes will remain unresolved and growing which necessitate the improvement in product design and waste management in order to overcome the both problem: plastic waste generation and their disposal<sup>7</sup>.



**Fig.2:-** Consumer packing growth by industry 2018-2025<sup>2</sup>

However, two popular models for plastic waste management are; use of plastic waste as a source of energy like burning in cement kilns and power plants and recovering polymers from plastic wastes. In table 1 shows the quantities and types of polymers littered in municipal solid waste (MSW) annually in the United States and this data also shows that the main plastic wastes in MSW are thermoplastic polymers polyethylene, polypropylene and PET.

**Table 1:-** Types and quantity of Plastic in MSW in US<sup>6, 8</sup>

| Type of Plastic                  | Quantity (in million tons) |
|----------------------------------|----------------------------|
| Low density polyethylene (LDPE)  | 5.01                       |
| High density polyethylene (HDPE) | 4.12                       |
| Polypropylene (PP)               | 2.58                       |
| Polystyrene (PS)                 | 1.99                       |
| PET                              | 1.70                       |
| Others                           | 3.13                       |

Approximately 40 million tons of MSW is generated annually, which is growing at the rate of 1.5 to 2% each year. 12.3% of these 40 million tons of MSW consist of plastic wastes. PET bottles constitute most of the fraction<sup>9</sup>. PET has a good chemical resistance and also bears gas barrier properties<sup>10</sup> because this it is used for refilling of soft drinks and beverages. It belongs to the thermoplastic category of polymers and exhibits excellent thermo-mechanical properties<sup>11</sup>. It covers around 18% of the total polymers produced in the world and about 60% of total production of PET is used for making bottles and synthetic fibres<sup>12</sup>. Waste PET may be obtained mainly from three type of sources; bottles, foils and cord from tyres. Waste tyre rubber can be cheap and valuable source of PET. Waste tyre cord contains 77.6% PET, 18.7% polyamide (PA) and 3.7% polypropylene (PP). Depending on its processing and thermal history, polyethylene terephthalate may exist both as an amorphous (transparent) and as a semi-crystalline material. The semicrystalline material might appear transparent (particle size < 500 nm) or opaque and white (particle size up to a few microns) depending on its crystal structure and particle size. Its monomer (bis-B-hydroxyterephthalate) can be synthesized by the esterification reaction between terephthalic acid and ethylene glycol with water as a byproduct, or by transesterification reaction between ethylene glycol and dimethyl terephthalate with methanol as a byproduct. Polymerization is through a polycondensation reaction o f the monomers (done

immediately after esterification/transesterification) with ethylene glycol as the byproduct (the ethylene glycol is directly recycled in production).

PET is commonly recycled, and has the number "1" as its recycling symbol. Intrinsic viscosity of bottle grade PET is 0.70 - 0.78 dL/g Water bottles (flat) and 0.78 - 0.85 dL/g of Carbonated soft drink grade bottle.

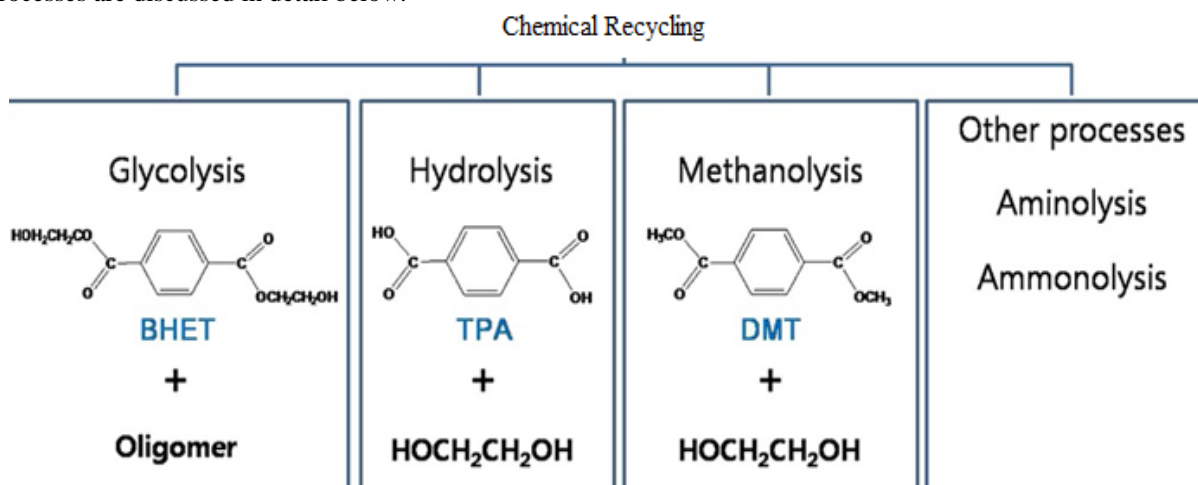
**Table:-** Physico- chemical properties of PET<sup>13</sup>

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|--|--|
| Molecular formula (C <sub>10</sub> H <sub>8</sub> O <sub>4</sub> ) n | Molecular formula (C <sub>10</sub> H <sub>8</sub> O <sub>4</sub> ) n |
| Density amorphous 1.370 g/cm <sup>3</sup>                            | Density amorphous 1.370 g/cm <sup>3</sup>                            |
| Density crystalline 1.455 g/cm <sup>3</sup>                          | Density crystalline 1.455 g/cm <sup>3</sup>                          |
| Young's modulus (E)  | 2800-3100 MPa  |
| Tensile strength   | 55-75 MPa  |
| Elastic limit  | 50-150%  |
| notch test   | 3.6kJ/m <sup>2</sup>   |
| Glass temperature 75 °C  | Glass temperature 75 °C  |
| melting point  | 260 °C   |
| Vicat B  | 170 °C   |
| Thermal conductivity   | 0.24 W/(m-K)   |
| linear expansion coefficient   | linear expansion coefficient (α) 7x10 <sup>-5</sup> /K               |
| Water absorption (ASTM)  | 0.16   |
| Refractive Index   | 1.5750   |
| Specific heat  | 1.0 kJ/kg <sup>-K</sup>  |

The established polyester recycling industry has three major sections; PET bottle collection and waste separation—waste logistics, Production of clean bottle flakes—flake production and Conversion of PET flakes to final products—flake processing. The intermediate product of first section contains PET content more than 90%. In the second section the collected bottles are converted to clean PET bottle flakes. This step can be more or less complex and complicated depending on required final flake quality. During third step PET bottle flakes are processed to desired products like fibre, bottles, film, filament, strapping or intermediates like pellets for further processing and moulding plastic products. PET flakes can be converted into PET fibres by mainly three processes; re-extrusion, mechanical recycling and chemical recycling. Mechanical recycling is the preferred method over chemical recycling of PET due to reduction of processing costs, reduction in the use of non-renewable energy, abiotic exhaustion, acidification, and also reduction in human and water toxicity<sup>22, 26</sup>. The mechanical recycling of PET involves collection of waste, washing, shredding or grinding to obtain flakes, dehydrating, drying and remoulding<sup>16, 18, 19, 21, 24, 25</sup>.

In the chemical recycling, depolymerisation of PET is carried out by chemical process like hydrolysis, glycolysis and methanolysis<sup>16-21</sup> and aminolysis reaction<sup>35, 36</sup>, it produces superior quality products but consumes highly labour and power, so this technique is costly<sup>22-24</sup>. Petrochemical components can be recovered in chemical recycling process, which can be used further for manufacturing of PET products or other synthetic chemicals<sup>27</sup>, but the recovery of these byproducts have not been cost effective because availability of low priced petrochemical feedstocks compared with processing cost for production synthetic chemicals or oligomers from PET waste<sup>28</sup>. In chemical recycling the PET polymer is converted into either its monomers or oligomers and other petrochemicals<sup>29-34</sup>. It is more costly process than the mechanical recycling; because of higher production cost of chemically recycled PET as compared to the cost of virgin PET therefore it needs to be implemented at a large scale, so that it becomes feasible. The

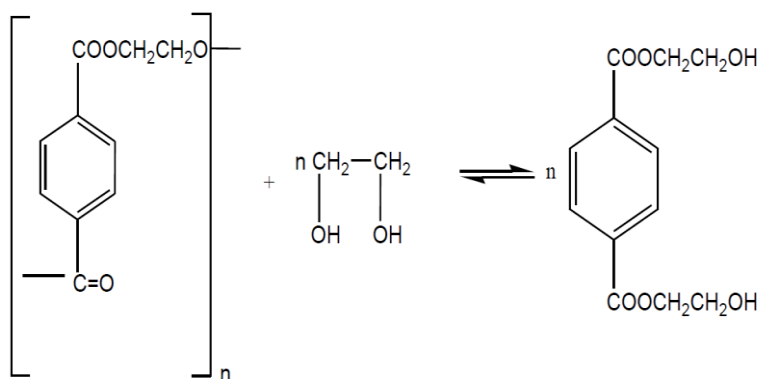
reaction products of each chemical recycling reaction of PET are given in figure 3. These chemical recycling processes are discussed in detail below.



**Fig. 3:-** Classification of the chemical recycling methods with the products of each reaction<sup>27</sup>.

### Glycolysis

This type of chemical recycling of PET widely accepted for commercial production of recycled products since 1965, firstly developed by McDowell et al<sup>37</sup>. In glycolysis reaction of recycled PET, polymer chain is break down by glycols in the presence of transesterification catalyst, yielding Bis(2-hydrxyl) terphthalate (BHET)<sup>38</sup> for polyester condensation as shown in figure 6. The produced BHET can be mixed with the virgin BHET, which can be utilized in production of different PET products. The chemical equation of glycolysis of PET is given in figure 4. As shown in figure 5, along with BHET other oligomers are also produced, so production of specific product is impossible<sup>39</sup>. The complete depolymerisation of recycled PET flakes is not possible due slow rate of glycolysis of PET flakes<sup>40, 41</sup>. Therefore it is field of investigation of methods for increasing the rate of glycolysis using suitable catalyst and optimizing the reaction conditions.



**Fig. 4:-** Equation of glycolysis of PET.

The metallic catalyst enhances the transesterification of PET<sup>42, 43</sup>. The reaction rate of glycolysis PET flakes and yield of BHET depends on various factors such as PET/ethylene glycol ratio, types and amounts of catalyst and PET/ethylene glycol ratio.

### Hydrolysis

In hydrolysis of PET, ethylene glycol and terephthalic acid (TPA) are formed under high pressure (1.4-2MPa) and at the temperature ranges 200-250°C, and it takes long time for depolymerization. Hydrolysis of PET can be proceeds by the addition of water in alkaline, acidic or neutral medium.

The acidic hydrolysis of PET is performed in the presence of sulphuric acid or nitric acid giving rise to TPA, however this process is not cost effective because separation of acid from ethylene glycol is not easy as well as high

price of Sulphuric acid again costly<sup>46</sup>. Sodium hydroxide and water or steam is used for alkaline and neutral medium respectively for hydrolysis of PET flakes<sup>47</sup>. The product obtained after the hydrolysis of PET may be utilized to produce virgin PET or oxalic acid<sup>45</sup>. This method is generally not used commercially, because the purification issues of TPA are associated with this technique<sup>44</sup>.

### Methanolysis

In methanolysis process PET is dissolved in methanol and converted into dimethyl terephthalate (DMT) and ethylene glycol (EG). The produced DMT is purified by distillation and crystallization<sup>48</sup>. Postconsumer films, fiber waste, plant waste, and bottle scraps are main PET feedstocks for methanolysis.

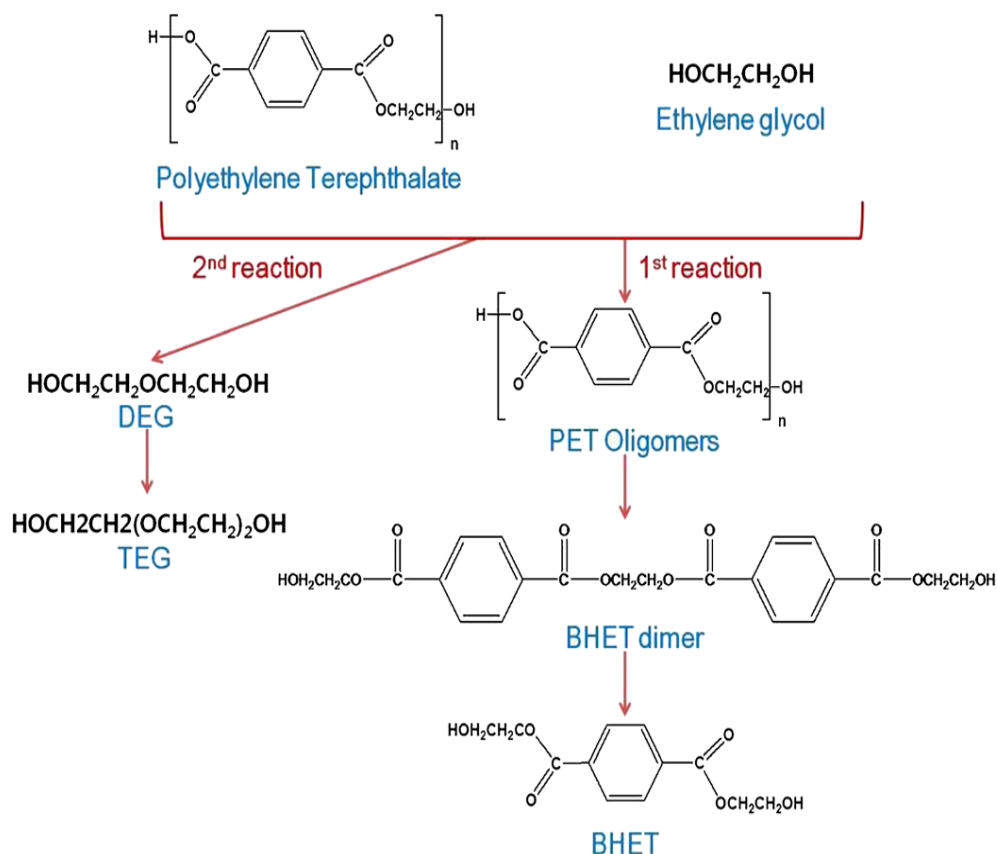


Fig. 5:- Glycolysis reaction scheme<sup>38</sup>

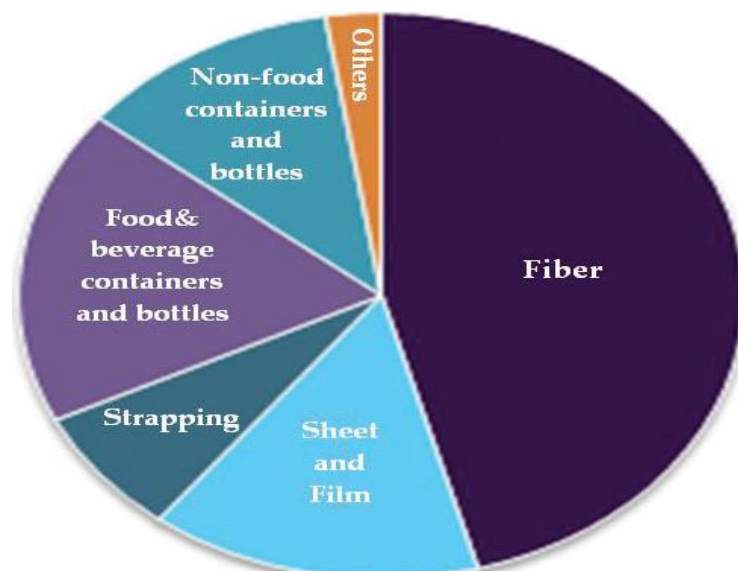
The costly separation and refining of reaction products makes this process disadvantageous. Also if water reaches the reaction, water poisons the catalysts and forms azeotropes.

### Aminolysis

Aminolysis of PET is the depolymerization of PET waste using different amines such as polyamines, hydrazine, allylamine and morpholine and produces terephthalamide<sup>49</sup>. The depolymerisation of PET by aminolysis process is not utilized commercially. However, partial aminolysis is applied in the improvement of PET quality in the production of fibres<sup>50</sup>.

### Application of recycled PET (rPET)

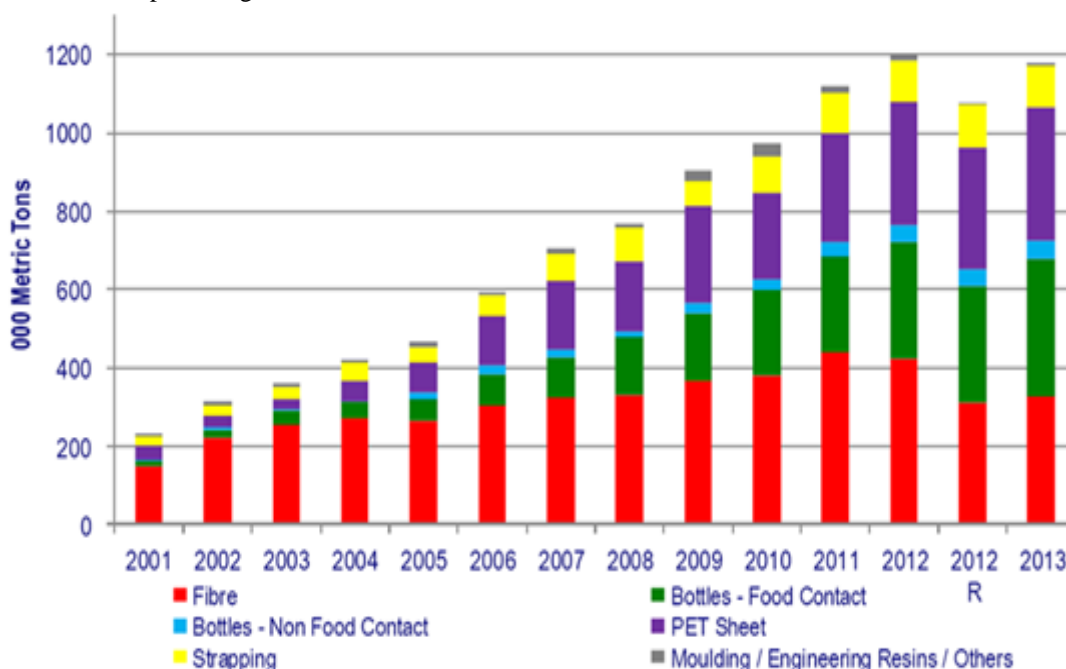
Application of recycled PET (rPET) can be studied into two headings: applications of fibres derived from mechanically rPET and applications of chemically rPET. Figure 6 shows various application of PET flakes<sup>14</sup>. The fibre obtained from PET flakes contributed about 44% proportion among end users of total market share in 2016. In Europe, recycled PET is near about equally utilized in making fibres, sheets and drinking bottles as shown in figure 7.



**Fig. 6:-** Global annual r-PET market volume by end-use in 2016<sup>14</sup>

The main product of mechanically rPET is fibre, which has several applications in various area such as textile industry, coating industry, food packaging, pavement industries, bottles used for different purposes, etc. In 1993, a technology was developed to obtain PET fibre much finer than 3denier, and this fibre has been used commercially<sup>39</sup>. To obtain these fibres the consistent intrinsic viscosity of post-consumer PET flakes should be around  $0.7\text{dlg}^{-1}$ .

The PET flakes can be converted into fibres by two methods: PET flakes are directly extruded into fibres and in the second method PET flakes are processed into pellets and then melt extruded into rPET fibres. Many studies are focused on the use of rPET staple fibre in the area of textile applications. Investigation are done on the properties of yarns and knitted fabrics obtained from virgin PET (vPET) and rPET and cotton fibres as virgin and recycled<sup>51</sup>. It is found that yarns obtained from rPET fibres had better yarn unevenness, better quality index and decreased yarn imperfections. Recycled yarn also exhibited lower knitted fabric burst strength as compared to virgin yarn<sup>51</sup>. Telli and Özdil studied the performance of blended yarns of r-PET and v-PET with cotton at different blend ratios 30/70, 50/50, 70/30 and 100 percentages<sup>52</sup>.



**Fig. 7:-** End use pattern of rPET in Europe<sup>53</sup>

One of the applications of rPET is the use of it as modifier of bitumen binder used in pavements<sup>54,55</sup>. Mohammed A. Hussein Alzuhair et al investigated the use of waste PET bottles as additive of bitumen for improving pavement performance in the doses of 2, 4, 6, 8, 10 and 12 wt%. These compositions were tested through the examination of FTIR, ductility and penetration tests. By increasing the content of PET in bituminous mixture, the flexibility, resistance to permanent deformation have increased which led to better engineering properties<sup>56</sup>. rPET are also used in asphalt concrete<sup>57</sup>. Mohamed Meftah Ben Zair et al also observed that the modification of asphalt with PET enhanced the mechanical properties, resistance to rutting and fatigue, durability, and also long-term sustainability of the pavement. Finally, the rPET modified asphalt mixture could serve as a wonderful and as an environmentally friendly material which can solve the problem of disposal of PET by producing high quality pavements<sup>58</sup>.

The improvement in the tensile strength, adhesion to the aggregate and other engineering properties have been reported by addition of rPET in concrete mix<sup>59, 60</sup>. D Alighiri et al observed that brick quality has been improved. It has compressive strength 60-100 kg/cm<sup>2</sup> and 20% water absorbency<sup>61</sup>.

### Applications for chemically recycled PET

Chemically recycled PET has been and having a very wide range of applications, such as manufacturing of polyurethane<sup>62, 63, 64</sup>, which itself is of great importance like foam, insulation, coating and artificial leather<sup>68</sup>, and unsaturated polyester resin<sup>65-67</sup>. The synthesis of new urethane oil from glycolysis-treated PET wastes was reported by Mercit<sup>69</sup>. The unsaturated polyester resins derived from rPET have been utilized for manufacturing of high performance fibre reinforced polymer composites<sup>66</sup>.

### Conclusions:-

The high strength of PET as compared to its light weight make it more energy efficient material. In the past few decades it becomes most demanding industrially used material. But as the demands of PET is growing time to time, in the almost same quantity of waste are generated. In this article, the strategies for PET recycling and their economic and environmental aspects are discussed. The advantages and disadvantages of mechanical and chemical methods of recycling of waste PET in industrial context are also discussed in this review. The fibres produced by mechanically recycled PET are mainly used in textile industry, while rPET produced from chemical recycling process are utilized as feedstock for manufacturing of polyurethane and unsaturated polyester resins. The main challenge before the researcher is develop an efficient, eco-friendly, less energy consuming and sustainable technology for management and recycling of waste PET.

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