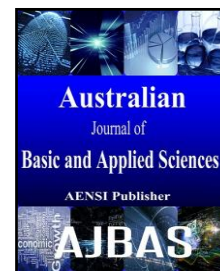




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Current Scenario of Biodegradation of Plastics – Review

¹G. Gnanavel, ²M. Thirumarimurugan and ³V.P. Mohana Jeya Valli¹Department of Biotechnology, Karunya University, Coimbatore, Tamil Nadu, India.²Department of Chemical Engineering, Coimbatore Institute of Technology, Coimbatore, Tamil Nadu, India,³Tamilnadu Pollution Control Board, Coimbatore, Tamil Nadu, India.

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ABSTRACT

Plastic, any organic material with the ability to flow into a desired shape when heat and pressure are applied to it and to retain the shape when they are withdrawn. Based on the sources of plastics, it can be classified in to two major groups. This review paper is mainly focus on the degradation of synthetic and natural plastics using microorganisms and controls the plastics waste. Isolated fungal strains were identified as *Aspergillus niger*, *A. japonicus*, *A. terreus*, *A. flavus* and *Mucor sp.* Predominant fungal strains *Aspergillus niger* and *A. japonicus* were selected for polythene degradation under laboratory conditions. Their effectiveness on the degradation of commercial polythene carry bags of low density polyethylene (LDPE) was studied over a period of 4 weeks in shaker culture under laboratory conditions. Biodegradation was measured in terms of mean weight loss, which was nearly 8 to 12% after a period of 4 weeks. Further SEM (Scanning electron microscopy) analysis confirmed the degradation by revealing the presence of porosity and fragility of the fungal degraded polythene surface. *Aspergillus japonicus* showed 12% degradation potential when compared to *A. niger* of 8% degradation in one month period.

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INTRODUCTION

Plastics are one of the most engineered materials, and have experienced spectacular growth in both usage and adaption. The raw materials used for plastics manufacture have also changed over time from coal, cellulose to petroleum and the latter is currently the main raw material. Plastics are available in the form of bars, tubes, sheets, coils, and blocks, and these can be fabricated to specification. (R.C. Thompson *et al.*, 2009). Plastics are usually classified by their chemical structure of the polymer's backbone and side chains. Some important groups in these classifications are the acrylics, polyesters, silicones, polyurethanes, and halogenated plastics. Plastics can also be classified by the chemical process used in their synthesis, such as condensation, poly-addition, and cross-linking. (Andrady & Neal 2009)

However, plastic articles are commonly manufactured from plastic powders in which desired shapes are fashioned by compression, transfer, injection, or extrusion moulding. In compression moulding, materials are generally placed immediately in mould cavities, where the application

of heat and pressure makes them first plastic, then hard. The transfer method, in which the compound is plasticized by outside heating and then poured into a mould to harden, is used for designs with intricate shapes and great variations in wall thickness. Injection-moulding machinery dissolves the plastic powder in a heating chamber and by plunger action forces it into cold moulds, where the product sets. (Hosler *et al.* 1999). The operations take place at rigidly controlled temperatures and intervals. Extrusion moulding employs a heating cylinder, pressure, and an extrusion die through which the molten plastic is sent and from which it exits in continuous form to be cut in lengths or coiled. (Nigel j. Mills, 2005)

2. Sources of plastics:

a). Synthetic:

Synthetic plastics are chemically manufactured from:

- Crude oil
- Coal
- Natural gas

b). Natural:

Natural sources of plastics include:

- *Plants - from which cellulose can be extracted*
- *Trees - from which latex, amber and resin can be extracted*

- *Animals - from which horn and milk (used to make glues) are obtained*
- *Insects - from which shellac (used to make polish) is obtained*

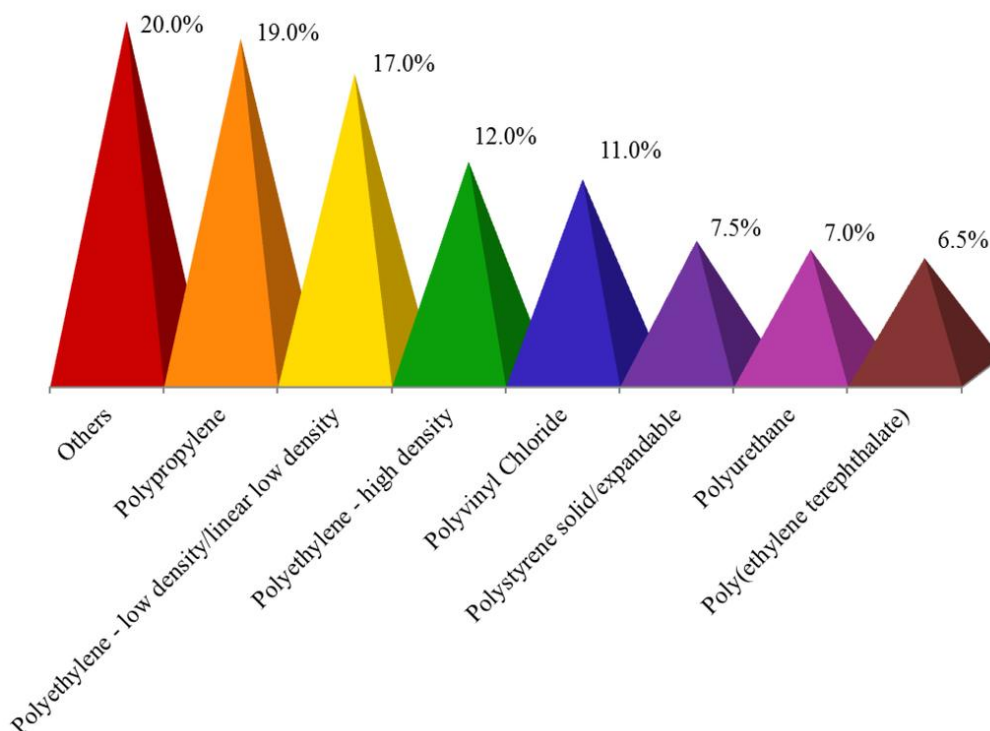


Fig. 2.1: World Plastics demands Plastics Europe. Plastics – The Facts 2012 (Hayden K. Webb, 2013).

2.1. Synthetic Plastics:

The Society of the Plastics Industry Inc. (SPI) introduced the Resin Identification Code (RIC) system in 1988 at the urging of recyclers around the country. The code was developed to meet recyclers' needs while providing manufacturers a consistent, uniform system that could apply nationwide. Because municipal recycling programs traditionally have targeted packaging - primarily containers - the coding system offered a means of identifying the resin content of bottles and containers commonly found in the residential waste stream. The majority of plastic packaging is made with one of six resins: polyethylene terephthalate (PETE); high density polyethylene (HDPE); polyvinyl chloride (PVC or vinyl); low density polyethylene (LDPE); polypropylene (PP); or polystyrene (PS). The RIC assigns each of these resins a number from 1 to 6 (See L. K. Arnold, 1968), (J. H. DuBois, 1972).

Indian Petrochemicals Industry:

The Indian basic petrochemicals market [including end products market which includes polymers, synthetic fibers, elastomers and surfactants] the total petrochemical market has growth at a CAGR of 8.1% from USD 13 billion in FY₂₀₀₆ to USD 19.3 billion in FY₂₀₁₁.

By global standards, its contribution to global market size is not very large, primary reason being low per capita consumption of polymers in India, only ~ 7 kgs, compared to world average of ~ 25 kgs.

1. *Polymers:* Polymers are popularly known as plastics, Polyethylene, Polystyrene, Polypropylene and Polyvinyl chloride are major types of polymers. Consumption of polymers has increased from 61% to ~70% of total volume of major end products petrochemicals between the period FY₀₆ and FY₁₁.

2. *Synthetic Fibers:* Synthetic fibers account for about half of all fiber usage, with applications in every field of fiber and textile technology. The market share of synthetic fibers has decreased from 28% in FY₀₆ to ~ 22% in FY₁₁.

3. *Elastomers:* Elastomers are polymers with elastic properties. Elastomers have declined from 5% in FY₀₆ to 2% in FY₁₁.

4. *Surfactants:* Surfactants stabilize mixture of oil and water. Its share has remained same in overall market at 6%.

The figure below gives the domestic demand and capacity of these segments for 2012 as well as future projections.

2.2. Natural Plastics:

Bioplastics are plastics derived from renewable biomass sources, such as vegetable fats and oils, corn

starch, pea starch or microbiota (Hong Chua *et al.* 1999). Bioplastic can be made from agricultural byproducts and also from used plastic bottles and other containers using microorganisms. Biodegradable bioplastics can break down in either anaerobic or aerobic environments, depending on how they are manufactured. Bioplastics can be composed of starches, cellulose, biopolymers, and a variety of other materials. Bioplastics are used for disposable items, such as packaging, crockery,

cutlery, pots, bowls, and straws (Chen, G, & Patel, M.2012). These plastics are also used in non-disposable applications including mobile phone casings, carpet fibres, insulation car interiors, fuel lines, and plastic piping. New electroactive bioplastics are being developed that can be used to carry electrical current (Suszkiw, Jan, 2005). Biopolymers are available as coatings for paper rather than the more common petrochemical coatings. (Khwalidia *et al.* 2010).

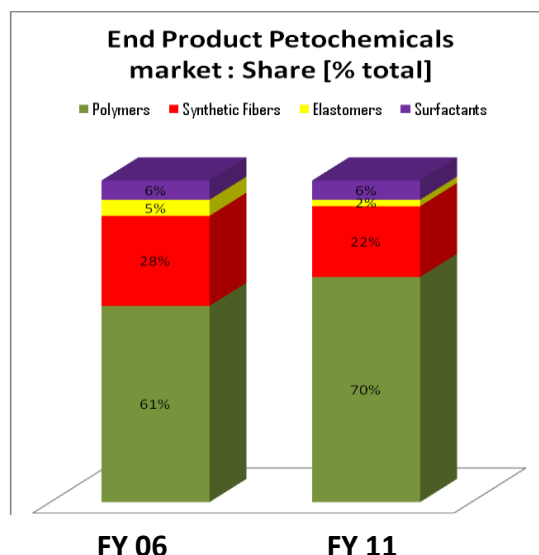


Fig. 2.2: Percentage of end product petrochemical market.

Table 2.1: Different segments of polymers.

| Segments | Major products | Main applications |
|------------------|--|--|
| Polymers | Poly ethylene, Poly styrene, and Poly vinyl chloride Poly propylene | Packaging, carrier bags, extrusion coating |
| Synthetic fibers | Polyester, nylon, acrylic fiber, purified terephthalic acid | Fiber and textile technology |
| Elastomers | Styrene butadiene rubber, poly butadiene rubber , plasticized PVC | Tyres, toys, consumer items |
| Surfactants | Linear alkyl benzene and ethylene oxide | Detergents, emulsifiers, foaming conditioning agents |

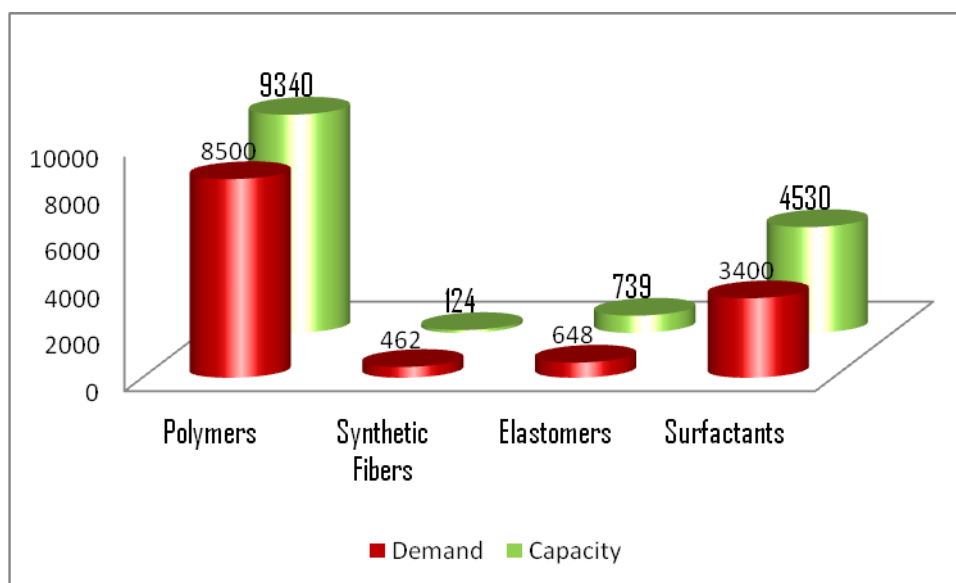


Fig. 2.3: Domestic demand and capacity of polymers.

Biobased: The term “biobased” means that the material or product is (partly) derived from biomass (plants). Biomass used for bioplastics stems from e.g. corn, sugarcane, or cellulose. **Biodegradable:** Biodegradation is a chemical process during which micro-organisms that are available in the environment convert materials into natural substances such as water, carbon dioxide, and compost (artificial additives are not needed). The process of biodegradation depends on the surrounding environmental conditions (e.g. location or temperature), on the material and on the application.

Bioplastics types:

The family of bioplastics is roughly divided into three main groups:

1. Biobased or partly biobased non-biodegradable plastics such as biobased PE, PP, or PET (so-called drop-ins) and biobased technical performance polymers such as PTT or TPC-ET
2. Plastics that are both biobased and biodegradable, such as PLA and PHA or PBS
3. Plastics that are based on fossil resources and are biodegradable, such as PBAT.

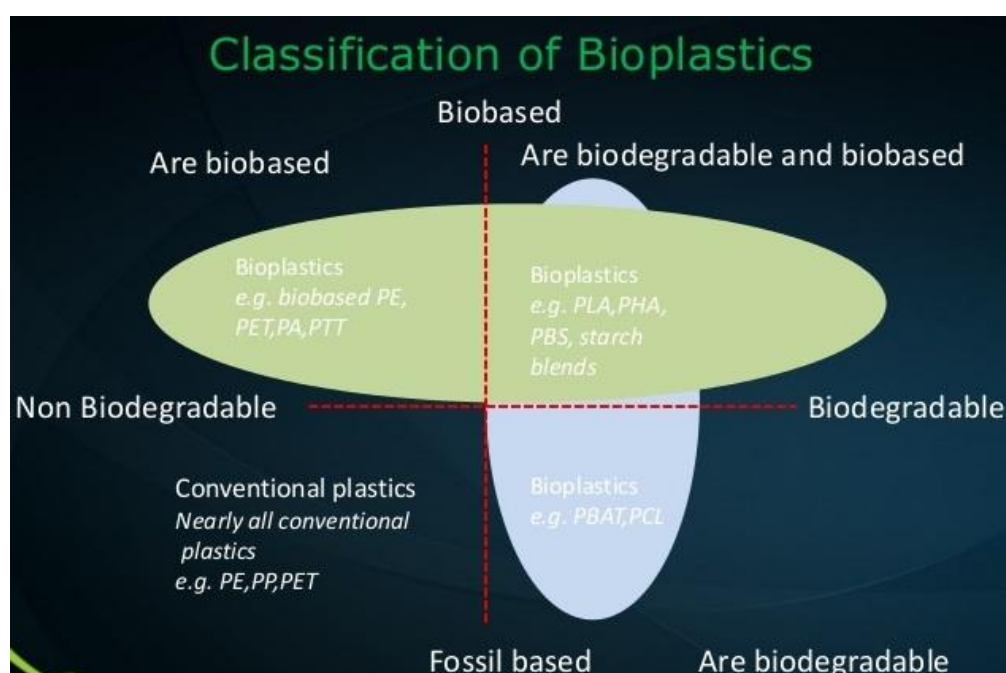


Fig. 2.4: Classification of bio-plastics.

Benefits of bioplastics:

In search of new material solutions and keeping an eye on the goal of sustainable production and consumption, bioplastics have several (potential) advantages. The use of renewable resources to produce bioplastics is the key for:

- increasing resource efficiency
- The resources can be cultivated on an (at least) annual basis;
- The principle of cascade use, as biomass can first be used for materials and then for energy generation;
- A reduction of the carbon footprint and GHG emissions of some materials and products
- Saving fossil resources, and for substituting them step by step.

3. How to reduce the plastic waste:

Material Classes Disposed in the Waste Stream:

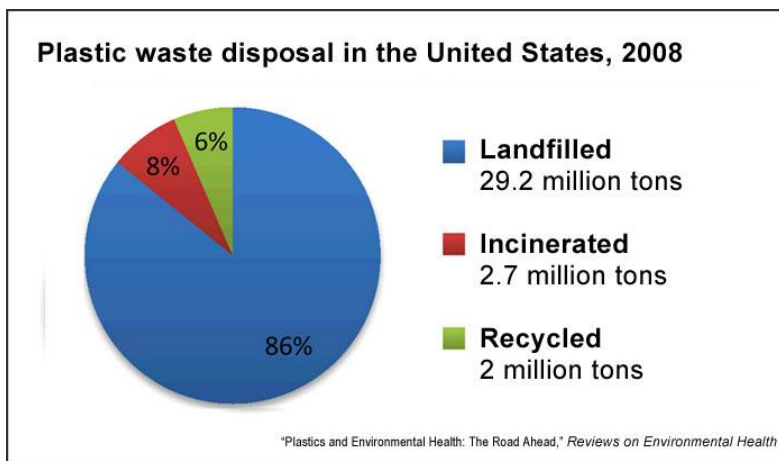
Waste characterization means finding out how much paper, glass, food waste, etc. is discarded in your waste stream. This typically involves collecting,

sorting, and weighing waste generated at schools and district facilities. Waste characterization information helps in planning how to reduce waste, set up recycling programs, and conserve money and other resources. (Kathy Frevert, Cara Morgan, 1999). Three ways we can reduce the plastics waste. i). Recycling method ii). Plastics additives iii). Plastics to fuel conversion.

On average, 300 million tons of plastic are produced around the globe each year. Of this, 50% is for disposable applications such as packaging. In 2008, 34 million tons of plastic was disposed in the United States. Of this, 86% ended up in landfills. However, “disposal of plastics in landfills is ultimately unsustainable and diminishes land resources fit for other uses of higher societal value. Incineration results in the release of carbon dioxide, a greenhouse gas, and of other air pollutants, including carcinogenic polycyclic aromatic hydrocarbons (PAHs) and dioxins.” (North *et al.*, 2014).

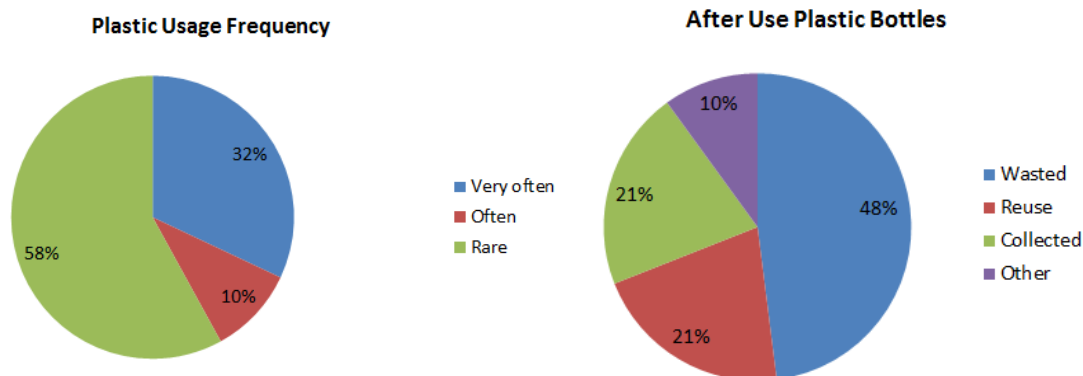
Table 3.1: School Waste Characterization Data.

| Material Type | Percent (%) of Waste Stream |
|-----------------------------------|-----------------------------|
| Construction and Demolition Waste | 1.0% |
| Glass | 0.6% |
| Household Hazardous Waste | 0.0% |
| Metal | 4.3% |
| Mixed Residue | <0.8% |
| Organics | 48.3% |
| Paper | 33.4% |
| Plastic | 11.1% |
| Special Waste | 0.5% |

**Fig. 3.1:** Plastics waste disposal in USA.

This survey was about plastic issue that will be brought by ITB_Indonesia team to iGEM 2014 competition in Boston. This survey depicted that 57% respondent used plastic in their daily activity and 48% of the respondent discarded those plastic

wastes. The awareness to recycle plastic wastes were very low and may cause accumulation of plastic wastes, especially bottle packaging. Therefore, such an environmentally degradation system was inevitably required.









**Fig. 3.2:** Plastics usage.

3.1. Recycling method:

Compared with lucrative recycling of materials, such as glass and metal, plastic polymers recycling is often more challenging because of low density and low value. There are also numerous technical hurdles to overcome when recycling plastic. Before recycling, most plastics are sorted according to their resin type. In the past, plastic reclaimers used the resin identification code (RIC), a method of categorization of polymer types, which was

developed by the Society of the Plastics Industry in 1988. The quantity of post-consumer plastics recycled has increased every year since at least 1990, but rates lag far behind those of other items, such as newspaper (about 80%) and corrugated fiber board (about 70%). Overall, U.S. post-consumer plastic waste for 2008 was estimated at 33.6 million tons; 2.2 million tons (6.5%) were recycled and 2.6 million tons (7.7%) were burned for energy; 28.9 million tons, or 85.5%, were discarded in landfills.

Table 3.2: Plastics Recycling code and applications.

| THE PLASTIC IDENTIFICATION CODE | | | | |
|--|---|---|---|--|
| Symbol | Type of Plastic | Properties | Common Uses | Recycled in: |
|  PET | PET Polyethylene Terephthalate | Clear, tough, solvent resistant, barrier to gas and moisture, softens at 80° | Soft drink and water bottles, salad domes, biscuit trays, salad dressing and containers | Pillow and sleeping bag filling, clothing, soft drink bottles, carpeting, building insulation |
|  HDPE | HDPE High Density Polyethylene | Hard to semi-flexible, resistant to chemicals and moisture, waxy surface, opaque, softens at 75° C, easily coloured, processed and formed | Shopping bags, freezer bags, milk bottles, ice cream containers, juice bottles, shampoo, chemical and detergent bottles, buckets, rigid agricultural pipe, crates | Recycling bins, compost bins, buckets, detergent containers, posts, fencing, pipes, plastic timber |
|  PVC | PVC Unplasticised Polyvinyl Chloride PVC-U Plasticised Polyvinyl Chloride PVC-P | Strong, tough, can be clear, can be solvent welded, softens at 80° C Flexible, clear, elastic, can be solvent welded | Cosmetic containers, electrical conduit, plumbing pipes and fittings, blister packs, wall cladding, roof sheeting, bottles Garden hose, shoe soles, cable sheathing, blood bags and tubing | Flooring, film and sheets, cables, speed bumps, packaging, binders, mud flaps and mats, new gumboots and shoes |
|  LDPE | LDPE Low density Polyethylene | Soft, flexible, waxy surface, translucent, softens at 70° C, scratches easily | Cling wrap, garbage bags, squeeze bottles, irrigation tubing, mulch film, refuse bags | Bin liners, pallet sheets |
|  PP | PP Polypropylene | Hard but still flexible, waxy surface, softens at 140° C, translucent, withstands solvents, versatile | Bottles and ice cream tubs, potato chip bags, straws, microwave dishes, kettles, garden furniture, lunch boxes, packaging tape | Pegs, bins, pipes, pallet sheets, oil funnels, car battery cases, trays |
|  PS  PS-E | PS Polystyrene PS-E Expanded polystyrene | Clear, glassy, rigid, opaque, semi-tough, softens at 95° C. Affected by fat, acids and solvents, but resistant to alkalis, salt solutions. Low water absorption, when not pigmented is clear, is odour and taste free. Special types of PS are available for special applications. | CD cases, plastic cutlery, imitation glassware, low cost brittle toys, video cases! Foamed polystyrene cups, takeaway clamshells, foamed meat trays, protective packaging and building and food insulation | Coat hangers, coasters, white ware components, stationery trays and accessories, picture frames, seed trays, building products |
|  OTHER | OTHER Letter below indicate ISO code for plastic type e.g. SAN, ABS, PC, Nylon | Includes all resins and multi-materials (e.g. laminates). Properties dependent on plastic or combination of plastics. | Automotive and appliance components, computers, electronics, cooler bottles, packaging | Automotive components, plastic timber |

3.2. Degradation of Plastics using additives:

Plastics are rarely used by themselves; typically, the resins are mixed with other materials called 'additives' to enhance performance. These may include inorganic fillers (e.g. carbon or silica) to reinforce the plastic material, thermal stabilizers to allow the plastics to be processed at high temperatures, plasticizers to render the material pliable and flexible, fire retardants to discourage ignition and burning, and UV stabilizers to prevent degradation when exposed to sunlight. Colorants, matting agents, opacifiers and lustre additives might also be used to enhance the appearance of a plastic product. Additives are often the most expensive component of a formulation, and the minimum quantity needed to achieve a given level of performance is generally used. The additives are intimately mixed with the polymer or 'compounded' into a formulation that is processed into the shape of

the final product. The potential adverse health issues associated with the use of specific additives, such as phthalate plasticizers, have been raised (for recent reviews, see Koch & Calafat 2009; Meeker *et al.* 2009; Oehlmann *et al.* 2009; Talsness *et al.* 2009).

3.3. Conversion of plastics to fuel:

American Plastics Council (APC) plastic can be converted totally to liquids and gases at 375°C in the presence of a hydrocracking catalyst, Ni/HSiAl or KC-2600. The quality of the liquid products obtained was close to that of a commercial premium gasoline. Catalytic hydrocracking of DSD plastic at the same conditions was found to be more difficult than that of APC plastic. This may be due to the negative effects of some impurities contained in DSD plastic, such as chlorine, ash, and paper. At 400°C, Duales System Deutschland (DSD) plastic can be nearly totally converted to gaseous and liquid products with 40%

KC- 2001 catalyst.(Weibing Ding *et al.*). Pyrolysis plants at commercial scale have promising economic results with return on investment being 2 to 5 years. Pyrolysis could be one of the solutions to problems in the future such as scarce land-fill space and lack of energy sources.(Antony Raja *et al.* 2011)

4. Testing procedure:

Industrial compostability – EN 13432, ASTM D6400:

Many starch-based plastics, PLA-based plastics and certain aliphatic-aromatic copolyester compounds, such as succinates and adipates, have obtained these certificates. Additive-based bioplastics sold as photodegradable or Oxo Biodegradable do not comply with these standards in their current form.

Compostability – ASTM D6002:

Which is capable of undergoing biological decomposition in a compost site such that the material is not visually distinguishable and breaks down into carbon dioxide, water, inorganic compounds and biomass at a rate consistent with known compostable materials.

Biobased – ASTM D6866:

The percentage of renewable carbon in a material (solid or liquid) can be measured with an accelerator mass spectrometer.

5. Polymer degradation:

Polymer degradation is a change in the properties -tensile strength, color, shape, etc.-of a polymer or polymer-based product under the influence of one or more environmental factors such as heat, light or chemicals such as acids, alkalis and some salts. These changes are usually undesirable, such as cracking and chemical disintegration of products or, more rarely, desirable, as in biodegradation, or deliberately lowering the molecular weight of a polymer for recycling.

Types of degradation:

1. Photoinduced degradation
2. Thermal degradation
3. Chemical degradation
 - a. Solvolysis
 - b. Ozonolysis
 - c. Oxidation
 - d. Galvanic action
 - e. Chlorine-induced cracking
4. Biological degradation
5. Stabilizers

Photo induced degradation:

Most polymers can be degraded by photolysis to give lower molecular weight molecules. Electromagnetic waves with the energy of visible light or higher, such as ultraviolet light,

(R.V.Lapshin, 2010), X-rays and gamma rays are usually involved in such reactions.

Thermal degradation:

Chain-growth polymers like poly(methyl methacrylate) can be degraded by thermolysis at high temperatures to give monomers, oils, gases and water.

Solvolysis - Step-growth polymers like polyesters, polyamides and polycarbonates can be degraded by solvolysis and mainly hydrolysis to give lower molecular weight molecules.

Ozonolysis- Cracks can be formed in many different elastomers by ozone attack.

Oxidation- The polymers are susceptible to attack by atmospheric oxygen, especially at elevated temperatures encountered during processing to shape. Galvanic action- polymer degradation may occur through galvanic action similar to that of metals under certain conditions and has been referred to as the "Faudree Effect".

Chlorine-induced cracking - highly reactive gas is chlorine, which will attack susceptible polymers such as acetal resin and polybutylene pipework.

Stabilizers- Hindered amine light stabilizers (HALS) stabilize against weathering by scavenging free radicals that are produced by photo-oxidation of the polymer matrix.

Biodegradation:

Plastics can be biologically degraded by microorganisms to give lower molecular weight molecules. Microorganisms can degrade plastic over 90 genera, from bacteria and fungi, among them; *Bacillus megaterium*, *Pseudomonas sp.*, *Azotobacter*, *Ralstonia eutropha*, *Halomonas sp.*, etc. (Chee *et al.*, 2010). *Phanerochaete chrysosporium* has been found to degrade LDPE blended with starch incubated in soil. *P. chrysosporium* was able to degrade LDPE more than 50%, then *P. chrysosporium* grown on media other than soil degradation occurs only 12% (Orhan and Buyukgungor 2000). Polyethylene has a high molecular weight can be degraded by fungi that degrade lignin under conditions of limited nitrogen and carbon limited and by manganese peroxidase. Fungi such as *Mucor rouxii* NRRL 1835 and *Aspergillus flavus* and some *Streptomyces* strains capable of degrading polyethylene containing 6% starch (Premraj and Mukesh, 2005). Biodegradation of plastics take an active part in different soil conditions according to the nature of the bacteria, because the microorganisms responsible for the degradation different from each other and have optimal growth of bacteria in the soil. It is also based on research (Katherisan, 2003) these bacteria *Pseudomonas sp.* able to degrade the plastic by 8.16% and was able to degrade the polythene by 20.54% within one month incubation anaerobically.

While this type of fungi *Aspergillus Glaucus* able to degrade the plastic by 7.26% and was able to degrade the polythene by 28.80% within one month incubation anaerobically.

6. Biodegradation studies:

Soil burial test:

Different types of plastics like synthetic plastics–Poly ethylene [PE] and natural plastics Poly (3-Hydroxybutyrate), [PHB] films were buried in soil for 3months, 6 months and 12months at room temperature. The availability of mineral salts and moisture and structure change in the polymer was analysed after soil burial. The plastics films were taken out and rinsed with sterilized distilled water and analyzed further to check the breakdown of functional groups and also change in surface morphology by FTIR and SEM respectively.

Plate assay:

This assay was carried for natural plastics [PHB] films and enrichment media were further tested for their ability to degrade the polymer by clear zone test. For clear zone tests (Augusta *et al.*, 1993) PHB powders were added to mineral salt medium at a final concentration of 0.2% and 0.3% w/v respectively, and the mixture was sonicated for 1 hour at 90 duty cycles using a Branson sonifer.

Sturm test:

Sturm test for the analysis of biodegradation by CO₂ evolution as a result of synthetic plastics [PE] and natural plastics [PHB]. The pieces of plastics films were added to the culture bottle containing 300 ml of mineral salts medium without any other carbon sources. About 5% inoculums of selected bacterial and fungal microorganisms were used for the biodegradation of different polymers. Spore suspension of *Fusarium* sp. was prepared in sterilized saline and was used to inoculate the test and the control bottles, containing mineral salt medium to study the degradation of synthetic plastics [PE].

7. Analytical methods:

Light microscopy:

The washed pieces of polymer buried in soil were taken. The plastic pieces were washed with sterilized distilled water and examined through microscope to observe the microbial attachment to plastic surface.

Scanning Electron Microscopy (SEM):

The surface morphology of synthetic plastic [PE] and natural plastic [PHB] films was thoroughly washed with sterilized distilled water and analyzed through Scanning Electron Microscopy to check for any structural changes after soil burials.

Fourier Transform Infra Red (FTIR) spectroscopy:

FTIR spectroscopy analysis was done to detect the degradation of synthetic plastic [PE] and natural plastic [PHB] after culturing in liquid media, on the basis of changes in the functional groups.

8. Conclusion:

Aspergillus niger showed degradation of low density polyethylene up to 5.8% in one month while *A. japonicus* showed more capability to degrade low density polyethylene up to 11.11% in one month under laboratory conditions. SEM analysis was done at different magnification. The control polythene strips displayed a normal surface view but the polythene strips treated with *A. niger* and *A. japonicus* showed appreciable surface corrosion, folding and cracks. (Lee, B *et al.* 1991), (N. Raaman *et al.*, 2012)

FTIR spectroscopy showed that synthetic plastic [PE] regions 3100 – 2700 cm⁻¹ and 1400 -1700 cm⁻¹ and natural plastic [PHB] regions 1400 – 1600 cm⁻¹ indicates the formation of C=C due to the breakdown of C-H bonds. Scanning Electron Microscopy (SEM) Few structural changes and erosions on the surface of synthetic plastic [PE] and natural plastic [PHB] pieces were observed, Sturm test: In synthetic plastic [PE], The total amount of CO₂ produced was 1.85 g/l, whereas in control it was 1.45 g/l and natural plastic [PHB] 7.85g/l whereas in control it was 0.94g/l.

Microorganisms play a significant role in biological decomposition of materials, including synthetic polymers in natural environments. High-density and low-density polyethylenes are the most commonly used synthetic plastics and they are slow in degradability in natural environments, causing serious environmental problems. In this regard, there is a growing interest in non-degradable synthetic polymer biodegradation using effective microorganisms. The potency of degradation by *A. japonicus* is twice than that of *A. niger* i.e., *A. japonicus* degraded 11.11% per month while *A. niger* degraded 5.8% per month. (N. Raaman *et al.*, 2012) (Lee, B *et al.* 1991) (Gu *et al.* 2000).

Fungal strains *Aspergillus niger* and *A. japonicus* were selected for polythene degradation under laboratory conditions. Their effectiveness on the degradation of commercial polythene carry bags of low density polyethylene was studied over a period of 2 and 4 weeks. Biodegradation was measured in terms of mean weight loss, which was nearly 8 to 12% after a period of 4 weeks. Further, SEM analysis confirmed the degradation revealing the presence of porosity and fragility of the fungal degraded polythene surface. *Aspergillus japonicus* showed 12% degradation potential when compared to *A. niger* of 8% degradation in one month period. (N. Raaman *et al.*, 2012).

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