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Biodegradation of polyethylene and polypropylene

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Polyethylene and polypropylene are the two polyolefins with wide ranging applications. They are recalcitrant and hence remain inert to degradation and deterioration leading to their accumulation in the environment, and, therefore creating serious environmental problems. In this review, biodegradation of these two polymers under *in vitro* conditions is reported. An attempt has been made to cover the mechanism of biodegradation, the various bacterial and fungal organisms that have been reported for the same, methods adopted for the studies and different characterization techniques followed to measure the extent of degradation

Keywords: polyethylene, polypropylene, biodegradation, *in vitro*

Introduction

The myriad applications of polymers in almost all the fields ranging from sophisticated articles such as, prosthetic hips and knee joints to disposable food utensils implies their significance and importance in our day to day life. Thus, enormous production and utilisation of polymers lead to their accumulation in the environment. Since not easily degraded by microorganisms, today they have become a serious source of pollution affecting both flora and fauna.

Polyolefins or saturated polymers have a broad range of applications. Polypropylene (PP) and polyethylene (PE), expressed as C_nH_{2n}, are most widely used linear hydrocarbon polymers. The versatility of these polymers arises from the fact that they are made from cheap petrochemical feed stocks through efficient catalytic polymerisation process and their ease of processing to various products. The range of their applications include, food packaging, textiles, lab equipments, and automotive components. PP has a methyl group instead of one of the hydrogens present in PE, on every other carbon, which gives rise to the existence of three stereoisomeric forms namely, atactic, isotactic, and syndiotactic[1]. This stereoregular polymer was first synthesised by Ziegler and Natta with propylene as the monomer. Metallocene catalysts can also be used for its synthesis. Industrially applicable PE was first synthesised in 1933 by Eric Fawcett and Reginald Gibson at ICI chemicals[2]. PE is totally linear and available with varying range of densities from 0.91 to 0.97 g/cm^3. Low density PE has branching at random places leading to low packing of the polymer chains, whereas the high density PE is more linear with minimal branching leading to high packing density[1].

As reported by American Plastic Association, percentage distribution of PP, high density polyethylene (HDPE), linear low density polyethylene (LLDPE) and low density polyethylene (LDPE) are 18.4%, 17.4%, 12.1% and 8.2%, respectively in terms of sales and use in the year 2004 in the United States, Canada, and Mexico[3]. Non-degradable plastics accumulate in the environment at a rate of 25 million tonnes per year[4]. Extensive use of non-biodegradable thermoplastics and the rate at which they accumulate in the environment, makes the humankind to realise the necessity to find its environmental impact. As the polymer usage is unavoidable, ways have to be found to (1) Enhance the biodegradability of the polymers by blending them with biodegradable natural polymers such as starch[5-19] or cellulose[20] etc; (2) Mixing with prooxidants[5,21,22] so that they are easily degraded and (3) Isolate[23] and improve microorganisms that can efficiently degrade these polymers. In order to attempt the third option the mechanism of biodegradation should be understood.

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Overview of Biodegradation of Polymers

A general overview of biodegradation of polymers over a period of time is schematically represented in Fig. 1. Polymeric materials released into the environment can undergo physical, chemical and biological degradation or combination of all these due to the presence of moisture, air, temperature, light (photo-degradation), high energy radiation (UV, γ-radiation) or microorganisms (bacteria or fungi). The rates of chemical and physical degradation are higher when compared to that of biodegradation. Also, physical and chemical degradation facilitates microbial degradation and complete mineralisation of the polymer happens due to biodegradation, which is generally the final step[24,25].

Mechanism of Biodegradation

Biodegradation of polymers involves following steps:

1. Attachment of microorganism to the surface of the polymer
2. Growth of microorganism utilising the polymer as the carbon source
3. Primary degradation of the polymer and
4. Ultimate degradation

Microorganisms can attach to the surface, if the polymer surface is hydrophilic. Since PP and PE have only CH_2 groups, the surfaces are hydrophobic. Initial physical or chemical degradation leads to the insertion of hydrophilic groups on the polymer surface making it more hydrophilic (insertion of hydrophilic groups also decreases the surface energy). Once the organism gets attached to the surface, it start growing by using the polymer as the carbon source. In the primary degradation, the main chain cleaves, leading to the formation of low-molecular weight fragments (oligomers), dimers or monomers[24]. The degradation is due to the extra cellular enzymes secreted by the organism. These low molecular weight compounds are further utilised by the microbes as carbon and energy sources. Small oligomers may also diffuse into the organism and get assimilated. The ultimate products of degradation are CO_2, H_2O and biomass under aerobic conditions. Anaerobic microorganisms can also degrade these polymers under anoxic conditions. The primary products then are CO_2, H_2O, CH_4 and biomass under methanogenic condition or H_2S, CO_2 and H_2O under sulfidogenic condition. The environmental conditions decide the group of microorganisms and the degradative pathway involved. Ultimate degradation of recalcitrant

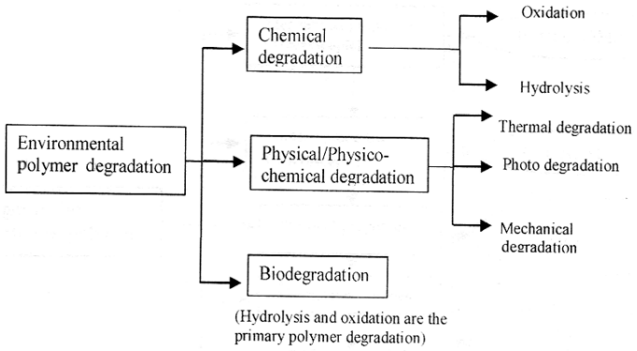

Fig. 1—Overview of degradation of polymers (Adapted from Vasile).

synthetic polymers may take several hundred years[24-28]. Additives, antioxidants and other stabilisers added to commercial polymers may be toxic to the organisms or may slow down the rate of biodegradation.

Strategies used to Characterize Biodegradability of Polymers

As mentioned before, the high molecular weight polymers are degraded first into oligomers, some of which might be water soluble and then they are further broken down into organic intermediates. The intermediate products may be acids, alcohols, ketones, etc. The following strategies are used to assess and monitor the biodegradation of the polymers:

1. Accumulation of biomass (experimentally determine the growth rate of microorganisms with the polymer as the sole carbon source)
2. Oxygen uptake rate
3. Carbon dioxide evolution rate
4. Products of reaction using chemical analysis
5. Surface changes
6. Changes in the mechanical and physical properties of the polymer[8]

Analytical Techniques

Several analytical techniques have been used to monitor the extent and nature of biodegradation (Fig. 2). These characterisation techniques are meant to study the mechanical, chemical, and physical properties of the polymer before and after degradation, which will help in understanding the extent as well as the mechanism of degradation. The study of mechanical properties comprises measuring of the tensile strength, elongation at fail and modulus of the polymer by using Instron. The physical properties of the polymers monitored are: morphology (microcracks, embrittlement using SEM, transmission optical microscopy), density, contact angle, viscosity,

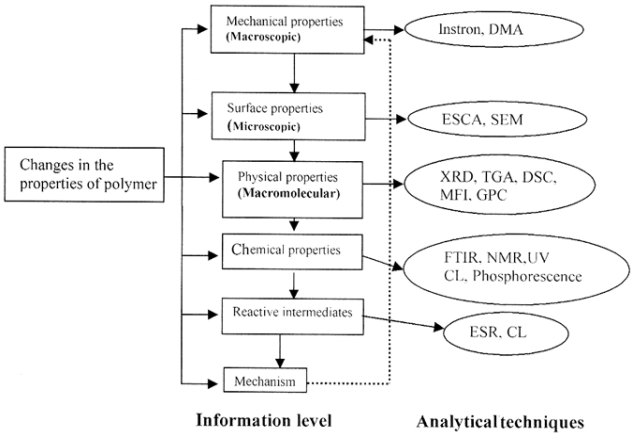

Fig. 2—Different levels of investigations on polymer biodegradation.

molecular weight distribution (using GPC), melting temperature (T_m), glass transition temperature (T_g) (doing TGA and DSC) and changes in the crystalline and amorphous regions (X-ray diffraction, SAXS and WAXS). The changes in the chemical properties that could be measured include formation or disappearance of functional groups as determined by FTIR. The molecular weight and molecular weight distribution of the degraded products or intermediates are characterised by techniques such as TLC, GC, GCMS, CL, MALDI-TOF, NMR (Fig. 3)[5,23]. The level of information derived from each technique, as shown in Fig. 2, increases as one moves downwards thereby understanding the mechanism of biodegradation. CO_2 evolution is measured by using GC[50], titrating with barium hydroxide[41]. Biofilm studies can be carried out using the acridine orange or BacLight bacterial viability kit[57]. The metabolic activity of the cells in the culture as well as in the biofilm can be done by ATP assays[22], protein analysis and FDA analysis[28]. Thermally stimulated current spectra obtained from electret-thermal analysis reveals the electric polarization properties of polymer which is used for investigating biodegradation. Corona discharge pretreatment of polymers showed better results compared to UV treatment[13,27].

Factors Affecting Biodegradability

Biodegradability of the polymer is essentially determined by the following important physical and chemical characteristics:

1. Availability of functional groups that increases hydrophilicity
2. Size, molecular weight and density of the polymer

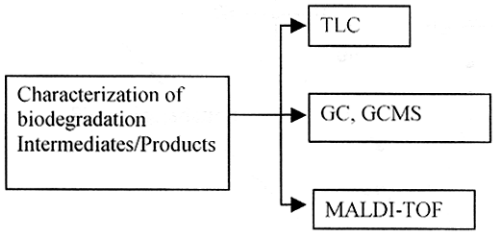

Fig. 3—Techniques used to characterize the degraded products.

3. Amount of crystalline and amorphous regions
4. Structural complexity such as linearity or presence of branching in the polymer
5. Presence of easily breakable bonds such as ester or amide bonds as against carbon-carbon bonds
6. Molecular composition (blend) and
7. Nature and physical form of the polymer such as whether it is in the form of films, pellets, powder or fibres[8,27]

Mechanism of Biodegradation of Polyolefins

In general, polyolefins are inert materials not susceptible to microbial attack because of the following reasons:

1. Hydrophobic backbones consisting of long carbon chains that give high resistivity against hydrolysis[3]
2. Addition of antioxidants and stabilisers during their manufacture which keeps polyolefins from atmospheric oxidation[3]
3. High molecular weight (from 10,000 to 40,000)
4. High packing density[8]

Even though PP is a polyolefin and prone to oxidative degradation similar to PE, the substitution of methyl in the place of hydrogen in the β position makes it more resistant to microbial attack, as already discussed in the factors affecting biodegradability (namely structural complexity)[8].

The decreasing order of susceptibility of polymers to degradation in soil mixed with municipal refuse was PE>>>>LDPE>HDPE as revealed by analysing the weight loss of samples, CO_2 evolution, changes in tensile strength, changes in FTIR and bacterial activity in the soil[12].

Studies reported on biodegradation of PP are given in Table 1. As evident from the table, the work carried out in this area is scarce. Apart from fungal species (*Aspergillus niger*), microbial communities such as the species of *Pseudomonas* and *Vibrio* have been reported to biodegrade PP[23]. A decrease in viscosity and

Table 1—Various literature reports on biodegradation of polypropylene and its blends

Title of the paper	Polymer	Organism	Conditions	Analytical techniques	Observation	Reference
Isotactic polypropylene biodegradation by microbial community	Isotactic polypropylene	Microaerophilic microbial community	Mineral medium containing sodiumlactate & glucose	IR, NMR, GC-MS	Organism & mycelia with known adaptability & metabolic flexibility can degrade isotactic PP	23
UV-Irradiated biodegradability of ethylene-propylene copolymers	Ethylene-propylene copolymers	Fungal species	Composting & culture environments	FTIR, SEM, VISCOSITY	Viscosity decrease & increase in carbonyl/hydroxyl region in FTIR	31
Biodegradation of γ-sterilised biomedical polyolefins	Isotactic polypropylene	Fungal species	Composting & culture environments	FTIR, SEM, VISCOSITY	Viscosity decrease & increase in chain scission	32
Blends						
Calorimetric & thermogravimetric studies of UV-irradiated polypropylene/starch-based materials aged in soil	Polypropylene/ starch	Soil	Soil burial tests	DSC and TGA	Biodegradation not affects the thermal stability, photooxidation decrease the thermal stability of the mixture	18
Effect of short wavelength UV-irradiation on ageing of polypropylene/cellulose compositions	polypropylene/ cellulose	Soil	Composted in garden soil	ATR-FTIR, TENSILE & SEM	Significant mechanical and surface changes found	20
Mechanical behavior of biodegradable polyolefins	(HDPE)/ polypropylene (PP)	Soil	Soil burial tests	DMM, VISCOELASTIC & DSC	A significant change in mechanical behaviour observed	36
Structure & properties of degradable polyolefin-starch blends	polyolefin-starch	*Phanerochaete chrsosporium*	Liquid fungus culture & soil burial test	Tensile DMTA, GPC, intrinsic viscosity, FTIR, & optical microscopy	Increased susceptibility to biodegradation	6
Enzymatic degradation of plastics containing polycaprolactone	PCL/PP	*Rhizopus arrhizus* lipase	Enzymatic condition	SEM & SPECTRO-METRIC	Blends of PCL and LDPE or PP retained high biodegradability of PCL	33
Thermal degradation of polypropylene/starch based materials with enhanced biodegradation	Polypropylene/ starch based materials	Soil	Soil burial tests	TGA, FTIR	Biodegradability observed more in starch based material rather than PP matters	15
Characterization by thermal analysis of HDPE/PP blends with enhanced biodegradation	Blends of HDPE/PP with different biodegradable additives	Soil	Soil burial tests	TG, DSC and dynamic-mechanical spectroscopy	Additive more affected by degradation than the polymeric matrix. Changes both in the crystalline morphology and activation energies of relaxation processes happens at different time & depends on the additives used	52

formation of new groups namely carbonyl and hydroxyl were observed during the degradation process[32,33]. Except for one report[23], all the studies deal with degradation of pretreated PP. The pretreatment techniques reported range from UV-irradiation[17,20,32], γ-sterilization[33] and thermal treatment[14]. These pretreatments either decrease the hydrophobicity of the polymer thereby making it more compatible with the organism or introduces groups such as C=O or –OH, which are more prone to degradation. It is reported that UV-treated PP sample is more susceptible to degradation than LDPE[32]. Biodegradation of polypropylene/starch or polypropylene/cellulose blends has been reported using soil organisms. It is observed that the organisms easily degrade starch or cellulose leaving behind the polymer. These carbohydrates or fillers also increase the adhesion of the organisms to the surface of the polymer[5-20]. Polycaprolactone (PCL) blended PP has also been reported to degrade in the presence of lipase[34]. PCL is an ester and since lipase is well known to degrade ester linkages, degradation of this polymer is facile. Lipase cannot affect the carbon-carbon present in PP. There are no reports available on the effect of tacticity on the nature and rates of biodegradation as well as on the use of marine organisms to achieve biodegradation.

Biodegradation of isotactic polypropylene without any pretreatment is reported with one of the community designated as 3S among the four microbial communities (designated as 1S, 2S, 3S and 6S) adapted to grow on starch containing polyethylene obtained from enrichment culture. *Pseudomonas chlororaphis*, *P. stutzeri*, and *Vibrio* species were identified in the community 3S. TLC, GC-MS, FTIR and NMR analysis of dichloro methane extracted products confirmed the mixtures of hydrocarbons with different degrees of functionalisation along with aromatic esters, which are added to the PP as a plasticiser. Sodium lactate and glucose had a co-metabolic effect. Starch enhances the adhesion of the microorganisms and also acts as a co-metabolite[23].

The degradability of PCL blends such as PCL with polystyrene (PS), poly-ethylenetelephthalate (PET), and polyhydroxybutyrate (PHB) were less when compared to the degradability of PCL blended with LDPE or PP. This was due to the miscibility of PCL with conventional plastics such as polyolefins. High biodegradability of PCL was observed with PCL-LDPE and PCL-PP blends[33].

Outdoor soil burial tests were done on the samples of a HDPE and PP blend with different biodegradable additives. DSC analysis of these polymers with different additives after a year showed no change in melting temperature and fraction of crystalline region. Therefore, it was concluded that the biodegradation begins at the amorphous region rather than at the crystalline region. Biodegraded HDPE/PP blends were more brittle in nature compared to non-degraded[36].

Mechanical, rheological and susceptibility for natural degradation of polymer starch blends mainly depends upon the content, properties of starch, kind and concentration of additives added with the plastics. LDPE demonstrated lower degradability as compared with polypropylene in the presence of epoxidised rubber. The biodegradation of polymer along with the starch phase was observed in few cases[6].

The biodegradability of the UV-irradiated films of isotactic polypropylene (i-PP), ethylene-propylene copolymer and LDPE was studied in composting and *A. niger* culture. Increase in the rate of carbonyl and hydroxyl groups, decrease in the intrinsic viscosity and increase in chain scission after UV-irradiation has been reported. Decrease in the carbonyl region in FTIR was confirmed by the utilization of oxidized polymers by the microorganisms. The copolymer EPF-30R (having 7.7% ethylene) degraded faster than EPQ-30R (having 15.1% ethylene) demonstrating the effect of the composition of copolymer on biodegradability. PP was found to be more susceptible to microbial attack than LDPE. Weight loss and surface erosion were also reported.[31] Additives are more susceptible to degradation rather than the HDPE and PP in HDPE/PP blends in outdoor soil burial test. Changes in the crystalline morphologies and activation energies of the relaxation process were confirmed by thermal analysis[52].

Accelerated photo- and bio-degradations were reported with PP/cellulose blends when compared with pure PP in garden soil compost[20]. γ-Sterilization of PP, LDPE and E-P copolymers were reported to have the same kind of effects as mentioned for UV-irradiated films[32]. Colorimetric and thermogravimetric studies on photo-degradation of polypropylene and a starch biodegradable additive mixture showed decrease in the crystallinity content due to free radical assisted chain scission, followed by biodegradation in soil, which later increased crystallinity due to the break down of chains in the amorphous region of the starch[18].

Studies carried out on polyethylene bio-degradation have been mentioned in Table 2. Unlike

Table 2—Various reports on biodegradation of polyethylene and its blends

Title of the paper	Polymer	Organism	Analytical techniques used	Observation	Reference
Biodegradation of thermally oxidized polyethylene	LDPE	Fungi *Aspergillus niger, Penicillium funiculosum, Paecilomyces variotii, & Cliocladium virens* Bacteria *Streptomyces badius, S. setnii & S. viridosporous*	DSC, FTIR, GPC & SEM	Molecular weight reduction, increase in carbonyl double bond groups, erosion on the surface of polyethylene is due to the microorganism	34
Degradation product pattern and morphology changes as means to differentiate abiotically and biotically aged degradable polyethylene	LDPE/ starch	*Arthrobacter paraffineus*	Gas chromatography-mass spectrometry, X-ray diffraction, size exclusion chromatography, FTIR, UV-Vis spectroscopy, DSC and SEM	Decrease in value of crystallinity, microorganism consumes carboxylic acids (carbon) evidenced by gas-mass spectrometry product	5
Biodegradation of octanoated starch and its blends with LDPE	LDPE/ starch blends	Soil microorganisms, sludge microorganisms	Tensile strength, elongation, weight loss & SEM	85% percentage of elongation and 50% weight loss in 6 months	7
Biodegradation of disposable polyethylene by fungi and *Streptomyces* species	Polyethylene	Fungi *Mucor rouxii* & Bacterium *Streptomyces* spp.	Tensile strength	Heat treatment 70°C for 10 d samples showed 60% elongation reduction in *Streptomyces* sp & 46.5% in fungi	40
Mechanical behavior of biodegradable polyolefins	HDPE/PP/ blends	Soil microorganisms	DSC, viscoelastic properties	Under soil burial conditions HDPE/PP blends altered mechanical behaviours	36
Physical structure of polyolefin-starch blends after ageing	LDPE blends	Soil organisms	DSC	48% increase in crystallinity index	8
Surface changes brought about by corona discharge treatment of polyethylene film and the effect on subsequent microbial colonization	LDPE	Fungus	Contact angle and FTIR	Formation of carbonyl groups by oxidative process	27
Enhancement of biodegradability of disposable polyethylene in controlled biological soil	LDPE/12% starch blend LDPE	Fungus *Phanerocheate chrysoporium*	Viscosity, percentage of elongation, CO_2 evolution, FT-IR	Molecular weight reduced from 90,000 to 50,000 in 6 months. FT-IR showed strong absorbance in the region 1650-1860 cm^{-1}, 56% percentage of elongation in 3 months, increases CO_2 evolution after 45 d of incubation	41
Thermally treated low density polyethylene biodegradation *Penicillium pinophilium* and *Aspergillus niger*	LDPE	*Penicillium pinophilium* & *Aspergillus niger*	DSC, X-ray diffraction, FTIR & SEM	Mineralization was evaluated and observed as 0.64% for *P. pinophilium* and 0.57% for *A. niger*. Decreases crystallinity, crystalline lamellar thickness. Increases carbonyl index incubation of 31 months samples	42

Contd.:

Table 2—Various reports on biodegradation of polyethylene and its blends: *Contd.:*

Title of the paper	Polymer	Organism	Analytical techniques used	Observation	Reference
Studies on biodegradability, morphology and thermo mechanical properties of LDPE/modified starch blends	LDPE/starch blends/starch phthalate	Soil organisms	Mechanical properties, DSC, melt flow index & SEM	Tensile strength & elongation at break increased in LDPE/starch phthalate blends compared to the LDPE/starch blends	11
Degradation of polyethylene by a fungus *Penicillum simssimum*	LDPE	Fungus *Penicillum simssimum*	HT-GPC, FT-IR	HT-GPC & FT-IR results showed double bonds of PE cut by Fungus *P. simssimum*	44
Experimental analysis and numerical simulation for biodegradability of polyethylene	Polyethylene/ wax	Microbial consortium	GPC	Weight loss was 31.5%	45
Evaluation of degradability of biodegradable polyethylene (PE)	Polyethylene	Soil microorganisms	Bioassmilations of product was evaluated	60% bioassimilation after 180 d	37
Biodegradation of thermally–oxidized, fragmented low density polyethylenes	LDPE pro-oxidant additives	Soil microorganisms	Co_2 Evolution, NMR, FTIR and SEM.	Increased 60% CO_2 evolution in 18 months, carbonyl & double bond relative intensities of the carbonyl bond at 1715 cm-1 & double bond at 1650 cm-1	38
Environmental Biodegradation of Polyethylene	Degradable polyethylene (EPI TAPA)	Bacteria *Rhodococcus rhodochrous, Cladosporium cladosporoides, Nocardia asteroides*	Epiflurocent microscopy, SEM, FT-IR	Increased absorbance of carbonyl groups & double bond formation in 6 months. 60% mineralization produced in 6 months	39
Biodegradation of synthetic polymers. II. A limited microbial conversion of ^{14}C in polyethylene to $^{14}CO_2$ by some soil fungi	Polyethylene	Unidentified three white rot fungi & *Fusarium redolens*	CO_2 Evolution	Increases 0.5% CO_2 evolution in 2 years of incubation	45
Biodegradation of synthetic polymers.III. The liberation of $^{14}CO_2$ by molds like *Fusarium redolens* from ^{14}C labeled pulverized high density polyethylene	HDPE	*Fusarium redolens, Acremonium kiliense, Aspergillus vesicolor* & *Verticillium lecanii*	CO_2 Evolution	Mixed culture of organism showed more degradation compared to single pure culture by estimation of CO_2	46
Biodegradation of plastic compost bags under controlled soil conditions	HDPELDPE and 9% starch polyethylene	Heterotrophic bacteria	Weight loss, tensile strength, carbon dioxide production & IR	Starch PE - 82.76% loss of tensile strength, HDPE-5.33% and LDPE 13.04%. 36% weight loss in starch blend PE	12
Biodegradation of physicochemically treated by a consortium of filamentous fungi	LDPE	*P. pinophilium, A. niger, Gliocladium virens & P. chrysosporium*	DSC, FTIR & SEM	Thermal treated PE samples decreasing melting point and relative crystallinity. Degradation products were carbonyl & double bonds groups	47

Contd.:

Table 2—Various reports on biodegradation of polyethylene and its blends: *Contd.:*

Title of the paper	Polymer	Organism	Analytical techniques used	Observation	Reference
Electret-thermal analysis to assess biodegradation of polymer composites	LDPE/starch	Bacteria *Baccilus, Clostridium & micrococcus* Fungi *Aspergillus, Penicillum & Mucor*	DSC, FTIR, SEM & Physico-Mechanical testing	Biological erosion of polyethylene by oxidative process	13
DSC, FTIR characterization of biodegradation of polyethylene	Polyethylene	Fungi *A. niger*	DSC & FTIR	Decreased amorphosity of the sample and relative intensity of carbonyl bond formation	48
Colonization, biofilm formation and biodegradation of polyethylene by a strain of *Rhodococus rubber*	LDPE blends	*Rhodococus rubber*	FTIR, SEM & weight loss	Carbonyl index reduced 66%, enrichment medium supplement with 2% mineral oil showed 50% degradation after 30 d incubation	28
Synergistic effect of combining UV sunlight-soil burial treatment on the biodegradation rate of LDPE/starch blends	LDPE/starch blends	Soil organisms	DSC, FT-IR, tensile strength & SEM	Starch blend PE exposed UV radiation & soil burial samples showed 66% degradation	14
Biodegradation of polyethylene by the thermophilic bacterium *Brevibacillus borstelensis*	LDPE	*Brevibacillus borstelen*	DSC, FT-IR	31% Molecular weight reduction in 30 d	51
Study and development of LDPE/starch partially biodegradable compounds	LDPE/starch blends	Sludge microorganisms	Tensile strength & SEM	Reduction in tensile strength & elongation properties, LDPE degraded in the amorphous region responsible for oxidative process	16
Acquired biodegradability of polyethylene containing pro-oxidant additives	LDPE HDPE/blends	*R. rhodochrous, N. asteroids, Aspergillus flavis, C. cladospoides*	ATP, ADP assays, Size exclusion chromatography,Micr oscopy techniques & NMR	*R. rhodochrous & N. astroides* found to be most active for molecular weight reduction	22
Effect of compatibiliser on the biodegradation and mechanical properties of high content starch/low density polyethylene blends	LDPE/starch blends	Soil organisms	Mechanical properties, weight loss, melt flow index & SEM	65% weight loss increase in 14 d	17
Polyethylene biodegradation by developed *Penicillium-Bacillus* biofilm	Polyethylene	*P. frequentans B. mycoides*	Microscopy, weight loss, gas chromatography	Weight loss of preheated polyethylene treated with fungi showed 7.150% & without preheating treated with showed 6.657%	50
Photo biodegradation of low density polyethylene/banana starch films	LDPE/starch blends	Soil microorganisms	FTIR, tensile strength, elongation & weight loss	Increased carbonyl index & Tensile strength & elongation at break increased in LDPE/starch blends	19
Biodegradation potential of some barrier-coated boards in different soil environments	Polyethylene & Polyester	Soil microorganisms	DSC & FTIR	Under soil burial condition PE/Polyester blends affect mechanical behaviors	49

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Table 2—Various reports on biodegradation of polyethylene and its blends: *Contd.:* -

Title of the paper	Polymer	Organism	Analytical techniques used	Observation	Reference
Modification of polymers by protein hydrolysate-A way to biodegradable materials	mLLDPE blend with HP	*Aspergillus oryzae*	Mechanical strength properties	Polymer blend with 20% HP (Protein hydrolysate) shows 35% biodegradation with acceptable range of mechanical strength whereas polymer with 40% HP shows 50% biodegradation with poor mechanical strength properties	40

polypropylene, more research articles are published on studies relating to biodegradation of PE. Fungi that include *A. niger, Penicillium funiculosum, Fusarium redolens,* and *A. vesicolor,* and soil microorganisms (mixed culture as well as *Rhodococcus rhodochrous, Cladosporium cladosporoides*) have been reported to degrade neat PE[8,11,19,34-39]. DSC or FTIR and other mechanical and physical techniques such as weight loss, changes in tensile strength have been the commonly used analytical techniques to monitor the nature of biodegradation. Thermal, UV, photo and corona treated PE has been found to degrade faster than the untreated polymer. Biodegradation of starch blended and modified PE with protein hydrolysate has also been studied[40]

Photooxidation is the triggering step in the oxidative degradation of polyethylene. UV radiation leads to radical formation, followed by the absorption of oxygen resulting in end products with carbonyl groups. Additional UV exposure causes the carbonyl group to undergo Norrish type I and/or Norrish type II degradation which leads to the cleavage of C-C bond and thus leading to the formation of oxidised low molecular weight fragments. Ultimately, photooxidation leads to the formation of low molecular weight fragments and thus increases the hydrophilicity of the polymer[14,18-20,25,26,29-31]. The photooxidation mechanism shown in Fig. 4, comprises both the formation of carbonyl group as well as Norrish type I and type II. Thus, photooxidation enhances the susceptibility of the polymer to microbes. The resulting carboxylic acid from the photooxidation and ω-oxidation of long chain hydrocarbons (similar to the biotic degradation of paraffin-C_{10-20}) enters the β-oxidation pathway as shown in Fig. 4. Later, the two carbon acetyl CoA, enters the TCA cycle and gets completely converted into carbon dioxide and water[5,26,27,30,31]

Cell homogenates from *P. putida* and *Bacillus brevis* were found to degrade PE films by oxidative degradation resulting in the formation of terminal hydroxyl, ketone and ester groups. The presence of alcohol dehydrogenase was confirmed indirectly in the degradation reaction by inhibition studies[55]. The known lignin degrading bacteria *S. virdosporos* T7A, *S. badius* 252, and *S. setonni* 75vi2 and the fungus *Phanerochaete chrysosporium* were used to assess their ability to degrade biodegradable polyethylene (polyethylene with 6% starch and pro-oxidant). The authors observed the accelerated pro-oxidant activity by heat treatment and UV treatment with different time period. Reduction in polydispersity and tensile strength were observed in biodegradable PE with bacterial treatment and not with the fungus[53]. The veratryl alcohol lignin peroxidase activity was confirmed[54].

A. niger has been reported to degrade commercially available PE. DSC analysis showed reduction in the amorphous region of the polymer[48]. Biodegradation of LDPE was enhanced with Tween 80 in the presence of *P. aeuroginosa*. This study explains the role of nonionic surfactant in biofilm formation, as explained before it is a prerequisite for biodegradation process[56]. Biodegradation of thermally oxidized LDPE with fungal cultures of *A. niger, Pencillium funicalosum, Paecilomyces variotii* and *Gliocladium virens* was marked by the gradual decrease in carbonyl region (1715 cm^{-1}) in FTIR[34]. Disposable polyethylene bags with 6% starch were subjected to biodegradation for a period of four weeks by eight different species of *Streptomyces* and the fungi, *Mucor rouxii* and *A. flavus*. Weight gain was seen after degradation with few *Streptomyces* species, whereas a slight loss of weight was observed with *S. aburaviensis, S. parvullus, S. nigellus* and *A. flavus*. Reduction in percentage elongation with *Streptomyces* and fungal cultures were 28.5% and 46.5%, respectively. Thermally treated film incubated with *Mucor* had 60% reduction in tensile strength[40].

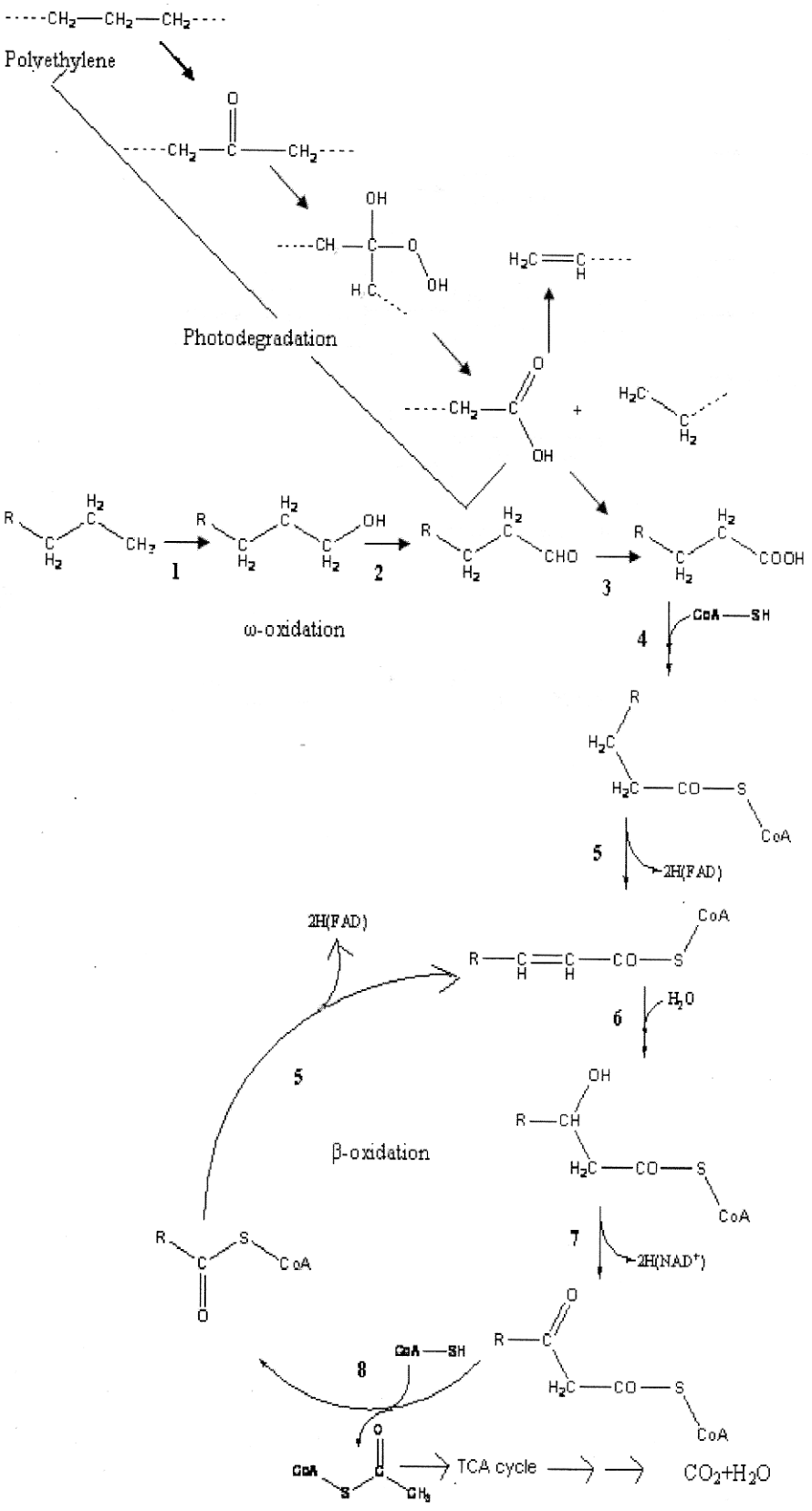

Fig. 4—Mechanism of biodegradation of polyethylene (Adapted from Vasile).

The bacteria, *Arthobacter parraffineus* was found to degrade LDPE in three years by utilising carboxylic acid formed during thermal oxidation. The utilization was through the β-oxidation mechanism that yields the degradation products like acetyl coA and propionyl CoA. 3- methyl-3-octanol and 1-hexadecanol were detected in biotic environment with series of n-alkanes such as C21-26. These were microbiologically metabolised by the oxidation of carboxylic acid through β-oxidation[5].

Rate of degradation of octonated starch is slower than pure starch. OCST-LDPE blend and octonated starch was subjected for six month soil burial test, which showed weight loss and reduction in mechanical properties. SEM analysis of OCST-LDPE blend showed the presence of holes on the surface, which confirmed the degradation of OCST region in blend[7].

Corona discharge treatment was found to be more effective towards colonisation of microorganisms on food packaging grade LDPE films with little effect on the mechanical properties as compared to UV treatment. This suggests that corona discharge treatment is affecting the hydrophobicity of the surface of the polymer and not penetrating it. A reduction in hydrophobicity of the LDPE from (92° to 66.6°) was also reported[27]. The *p*H of *Phanerochaete chrysosporium* inoculated soil with polyethylene decreased at a faster rate. Biomass, biological activity and CO_2 evolution was higher in inoculated soil. Analysis of the mechanical properties showed that decrease in the percentage elongation is faster in the inoculated soil compared to the uninoculated soil. Viscosity analysis of the polymers with regular intervals also showed the same trend[41]. A similar study was performed by Yamada-Onodera *et al* using Triton X-100. Improvement was observed in the growth of *Penicillium simplicissimum* YK, however, there was no utilisation of Triton X-100. FTIR analysis confirmed the utilisation of polyethylene by the fungus[44].

Thermal treatment of LDPE-TDPA (Pro-oxidant additive) in aerobic conditions showed substantial polymer fragmentation with loss of mechanical properties in 11 d. 26% of biodegradable and solvent extractable fraction was obtained after thermal oxidation for 20 d. 50-60% carbon dioxide evolution was observed in 18 months of further treatment with soil microorganisms[38]. Temperature is the crucial factor in determining the rate of thermo-oxidation whereas the effect of concentration of oxygen on the rate of thermo-oxidation is insignificant[37]. *A. niger, G. virens, Penicillium pinophilum, Phanerochaete chrysosporium* showed biodegradation on thermally treated or accelerated ageing treated (AAT) LDPE in 9 months. The biodegradation was evaluated by observing decrease in the onset of melting temperature (T_0) and melting temperature T_m and relative crystallinity. Highest mineralization (3.26%) values were obtained with AAT. Superficial growth of microorganisms occurred and penetration of hyphae was observed in the oxidised sample[47]. Synergistic effect of combining UV treatment and soil burial test was reported by Abd El-Rehim *et al*[14]. Electret-thermal analysis used in the electric polarization of dielectrics was used to investigate biodegradation of LDPE- starch blended polymer in 6 months. These studies were based on the assumption that biodegradation process of polymer material can cause transformation in their electrically non-equilibrium structure. Thermally stimulated current spectra (TSC) of PE films exposed to various ageing conditions in soil were reported. After ageing, new peaks were detected on spectra. FTIR results showed formation of functional groups. Reduction in melting was reported in DSC analysis. The degree of biological damage of the films was a function of starch content of the composites. The predominant microbial taxa in composites were *Bacillus*, *Clostridium*, *Micrococcus*, *Aspergillus*, *Penicillum* and *Mucor*[13].

Rhodococcus ruber C208 was isolated from the surface of the PE in polyethylene waste burial site by two step culture-enrichment protocol. Weight loss of 8% of photo-oxidised PE was observed in four weeks. This is higher than the rates already reported (3.5% to 8.4% after 10 years)[30]. In contrast to Albertsson's report, increase in the terminal double bond after photooxidation was observed. This could be explained by Norrish type I degradation of the carbonyl residues. They have reported that the double bonds were observed after the biodegradation of short PE oligomers produced during photooxidation. The analysis of extracellular polysaccharides in the biofilm of C208 was 2.5 folds higher than protein, suggesting its role in biofilm formation. Biofilm showed higher viability even after 60 d of incubation. Cell surface hydrophobicity of *R. ruber* was studied by SAT (salt aggregation test) and BATH (bacterial adhesion to hydrocarbon) tests. Addition of mineral

oil to this culture enhanced the degradation of the PE film by about 50% after four weeks of incubation. SEM photomicrographs of the bacterial biofilm showed some localized degradation of the PE around the bacteria. Protein assay and FDA hydrolysis by extracellular esterases showed increase in the biofilm formation for the first 2 d of assays followed by a sharp decrease in biomass density. The authors have hypothesised a low cell population with a low growth rate consisting of cells that are able to utilise PE as a carbon source[28,57].

Brevibacillus borstelensis, a thermophillic bacterium, was found to degrade polyethylene better than *R. rubber*, although the biofilm forming capacity of the former was not found to be as good as of the latter. Still it was able to show reduction in mass and molecular weight by 11 and 30%, respectively for UV irradiated polyethylene[51]. The LDPE and HDPE films after photo-oxidation and thermal oxidation corresponding to three years of outdoor weathering were incubated with *R. rhodochrous* and *Nocardia asteroids*. ATP assay was done to see the metabolic activity of the cells in culture and those adhered to the surface of the polymer. There was fast growth of microorganisms in the initial phase due to the availability of the low molecular weight oxidised products, which was followed by stabile metabolic activity. This was maintained for several months by the organisms utilising the polymer. The NMR analysis of the photo- and thermo-oxidized LDPE/HDPE aqueous extract revealed the presence of ethanol and formate, which are the end products of PE oxidation. This evidence supports the initial fast growth of microorganisms observed by ATP analysis. *Nocardia* formed dense filamentous mycelium on the surface. The size exclusion chromatographic analysis of the LDPE/HDPE after biotic and abiotic treatment showed no change in the molecular weight distribution indicating that the microbial attack was only on the surface of the polymer. The degradation due to both biotic and abiotic factors depended on the thickness of the polymer[22].

Studies on biofilm formation by *Penicillium frequentans* and *Bacillus mycoides* showed that *P. frequentans* formed a network of mycelia on degradable polyethylene (DPE–chemical or photoinitiator added polyethylene), which was colonised by *B. mycoides*. The biofilm formation increased the biodegradability of *P. frequentans* by 14 folds. In general, homologous gene has been found in the genome of some *Bacillus* species that produce alkane monooxygenase. The degradation was checked with weight loss, microscopic studies to visualise biofilm formation and CO_2 production using GC[50].

Conclusions

This review discusses the literature on biodegradation of PE and PP. Most of the examples deal with fungi and bacterial based degradation. Pre-treated polymers degrade more easily than the untreated polymers. Also, degradation is more facile with starch and cellulose blended polymers. Cell surface hydrophobicity and addition of surfactants showed an important role in biofilm formation, which is prerequisite for biodegradation. Degradation leads to decrease in molecular weight, tensile strength and viscosity, formation of new functional groups such as carbonyl, hydroxyl, etc. Based on the literature one could conclude that in order to enhance biodegradation of PP or PE the following approaches could be adopted:

I. Modify the polymer for microbial utility by the (i)Addition of natural polymers and/or prooxidants to PP; (ii) Modification of polymers by protein hydrolysates; and (iii) Pretreatment of the polymer.

II. Modify the microbes to utilise the polymer by (i) Modifying medium composition, and thus enhancing the utilisation of polymer; and (ii) genetically modify the microorganism to utilise the polymer.

III. Overexpress the enzyme, which is responsible for degradation and purify it and utilise for this purpose. Strategies II and III require the understanding of mechanism of microbial degradation of these polymers.

APPENDIX – ABBREIVIATIONS

AAT	-Accelerated ageing treatment
ATP	-Adenosine TriPhosphate
ATR-FTIR	-Attenuated total reflectance - Fourier Transform Infrared Spectroscopy
CL	-Chemilluminesence
DMA	-Dynamic Mechanical Analysis
DPE	-Degradable Polyehtylene
DSC	-Differential Scanning Calorimetry
ESCA	-Electron Spectroscopy for Chemical Analysis

ESR -Electron Spin Resonance
FDA -Fluorescien DiAcetate
FTIR -Fourier Transform Infrared Spectroscopy
GC-MS -Gas Chromatography - Mass Spectrometry
HDPE -High Density Polyethylene
HTGPC -High temperature gel permeation chromatography
i-PP -Isotactic Polypropylene
LDPE -Low Density Polyethylene
MALDI-TOF-Matrix Assisted Laser Desorption/Ionisation - Time of flight
MFI -Melt Flow Index
NMR -Nuclear Magnetic Resonance Spectroscopy
NY -Nylon
OCST -Octonated starch
PCL -Polycaprolactone
PET -Polyethylenetelephthalate
PHB -Polyhydroxy butyrate
PS -Polystyrene
SAXS -Small Angle X-ray Scattering
SEM -Scanning Electron Microscopy
Tg -Glass Transition temperature
TGA -Thermogravimetric analysis
TLC -Thin Layer Chromatography
Tm - Melting temperature
TSC -Thermally Stimulated Current Spectra
UV -Ultra Violet Spectroscopy
WAXS -Wide Angle X-ray Scattering
XPS -X-ray Photoelectron Spectroscopy
XRD -X-Ray Diffraction

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