

Review article

Recent developments in bio-based polyethylene: Degradation studies, waste management and recycling

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ABSTRACT

Nowadays, the tendency to replace conventional fossil-based plastics is increasing considerably; there is a growing trend towards alternatives that involve the development of plastic materials derived from renewable sources, which are compostable and biodegradable. Indeed, only 1.5 % of whole plastic production is part of the small bioplastics market, even when these materials with a partial or full composition from biomass are rapidly expanding. A very interesting field of investigation is currently being developed in which the disposal and processing of the final products are evaluated in terms of reducing environmental harm. This review presents a compilation of polyethylene (PE) types, their uses, and current problems in the waste management of PE and recycling. Particularly, this review is based on the capabilities to synthesize bio-based PE from natural and renewable sources as a replacement for the raw material derived from petroleum. In addition to recent studies in degradation on different types of PE with weight loss ranges from 1 to 47 %, the techniques used and the main changes observed after degradation. Finally, perspectives are presented in the manuscript about renewable and non-renewable polymers, depending on the non-degradable, biodegradable, and compostable behavior, including composting recent studies in PE. In addition, it contributes to the 3R approaches to responsible waste management of PE and advancement towards an environmentally friendly PE.

1. Introduction

PE is one of the most widely used thermoplastics and the highest-volume polymer in production and consumption. Its high toughness, ductility, excellent chemical resistance, low permeability and electrical conductivity, semicrystalline and ease of process make PE an attractive choice for various products and applications [1–4].

Although PE is considered an excellent material widely used in the areas of health care and food packaging, a considerable problem is the time required to achieve its complete degradation under uncontrolled conditions. For this reason, new tools and bio-based formulations must be formulated to reduce the material's degradation time and avoid contamination in landfills where it is discarded. Since most of the resources and raw materials used in PE production originate from oil, there is a predominant emphasis within the industry and scientific community on substituting these fossil-based materials with environmentally sustainable alternatives. Additionally, there is a significant focus on creating materials that can be effectively recycled or biodegraded after their product life

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cycle and bio-based compounds to produce new polymers and materials [5–8].

Some examples found in the literature are those in which agro-industrial residues such as garlic [9] and morning glory stem fibers [10] are used to elaborate dishes and cutlery, respectively. Besides, studies based on non-degradable polymers such as PE [11,12] and polyethylene terephthalate (PET) [13,14] by using bio-based sources to enhance the green behavior of the material, increase the possibilities of the inclusion in packaging with a more environmentally purpose.

This review addresses a compilation of PE types, their uses, and current problems in waste management and recycling; recent studies in degradation on different types of PE, weight loss ranges, the techniques used and the main changes observed after degradation. Particularly, the review is based on the capabilities to synthesize bio-based PE from natural and renewable sources. In addition, it contributes to the 3R approaches to responsible waste management of PE and advancement towards an environmentally friendly PE.

2. Types of polyethylene, structure, properties, and uses

PE is produced by free radical or addition polymerization of ethylene (or ethene) monomer. Ziegler-Natta, metal oxide, and metallocene catalysts are used to control linearity and density and thus obtain a type of PE with characteristics and specific properties [15–17]. In general, a PE molecule is formed by a long chain of repeating units of a pair of carbon atoms (-C-C-), covalently linked with two hydrogen atoms for each carbon (-CH₂-CH₂-), and the chain ends with a methyl group (R-CH₃) [2,15,16]. Among the different types of PE, more synthesized and with greater applications are (Fig. 1):

- High-density polyethylene (HDPE).
- Low-density polyethylene (LDPE).
- Linear low-density polyethylene (LLDPE).
- Ultra low-density polyethylene (ULDPE).
- Cross-linked polyethylene and polyethylene copolymers.

They are classified according to density and branching, and the most commonly utilized are HDPE, LDPE and LLDPE (Fig. 1).

The synthesis of these PEs depends on the method and the type of catalyst used, which will reflect in their physical and chemical properties, such as the chemical structure, density, degree of crystallinity, and thermal and mechanical properties, which will determine your final application. They are used in many applications: thin films, packaging, plastic containers, bottles, bags, plastic toys, wire and cable insulations, medical tubing, and injection molding, among others [1,2,15,16]. The main types of PE, their properties and their applications are presented in Table 1.

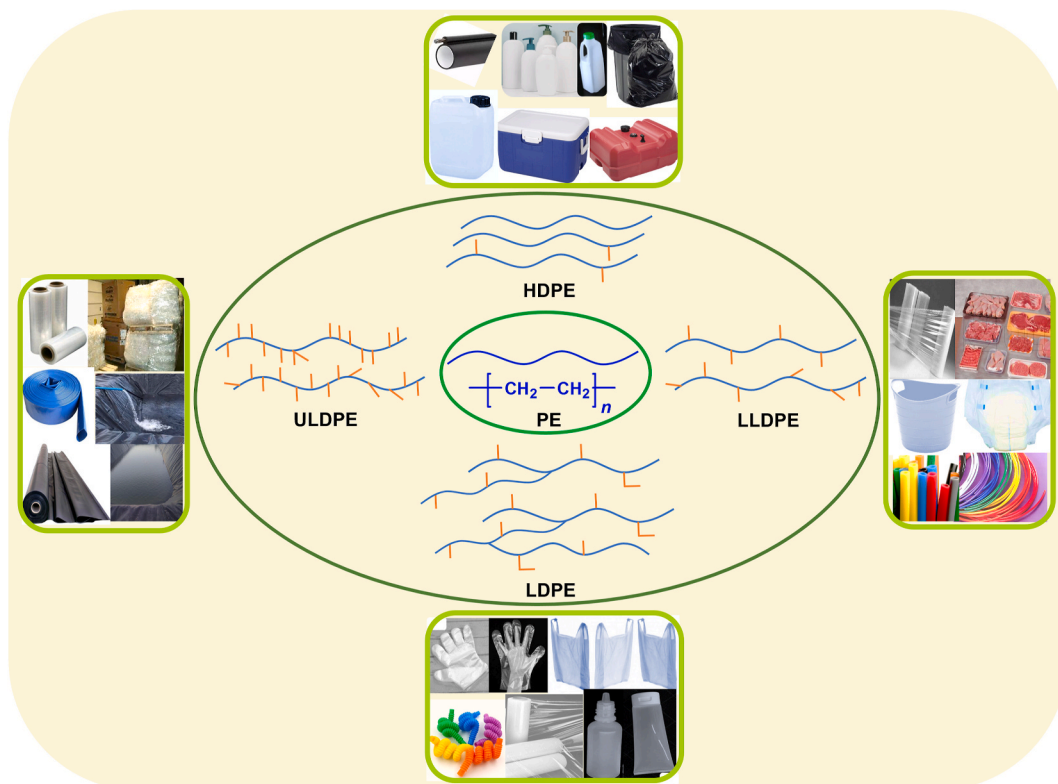


Fig. 1. Chemical structure of PE and its most used and commercial polymers for various applications.

Table 1
Properties, structure, and applications of different types of polyethylene.

Characteristics and properties	HDPE	LDPE	LLDPE	ULDPE
Name	High-density Polyethylene	Low-density Polyethylene	Linear Low-Density Polyethylene	Ultra Low-Density Polyethylene or Very Low-Density Polyethylene (VLDPE)
Structure	Linear polyethylene backbones (unbranched molecules with very few defects)	Molecules with a high degree of branching (the branches comprise ethyl and butyl groups and a few long-chain branches)	Linear polyethylene backbones with a high degree of short-chain branching (uniform branches)	Similar to LLDPE with a higher concentration of short-chain branching
Characteristics	Opaque white material, hard to semi-flexible, excellent chemical resistance, high resistance to solvents, high stiffness, high tensile strength, excellent electrical insulating properties and moisture barrier properties, very low water absorption	Translucent material, transparent in thin film form, high flexibility, excellent resistance to acids, bases, and vegetable oils, high resilience, high impact strength at low temperature, good barrier properties	Translucent material and natural milky color, very flexible with high impact strength, higher tensile strength, good impact resistance, higher puncture resistance, and good barrier properties	Transparent in thin film form, soft and flexible, low thickness, high deform, low tensile strength, low mechanical properties, good optical properties, and excellent sealability
Density (g/cm ³)	0.94–0.97	0.91–0.94	0.90–0.94	0.80–0.90
Degree of crystallinity (%)	High crystalline (>60 %)	Low crystalline and high amorphous (<40 %)	Semicrystalline (30–50 %)	Predominantly non-crystalline (<20 %)
Yield stress (Mpa)	17–35	8–19	8–29	<8
Elastic modulus (Mpa)	900–1400	150–350	250–900	<250
Hardness (Shore D)	66–73	44–50	55–70	25–55
Melting temperature (°C)	120–140	100–115	100–125	60–100
Fusion heat (cal/g)	38–53	21–37	15–43	<15
General Applications	Molded parts, milk and detergent bottles, chemical storage tanks, fuel tanks, load-bearing film, garbage bags, and supermarket packaging, food storage containers, housewares, ice boxes, toys, hollow plastic products and pipes, telecommunication cables	Thin films, packaging, plastic bags, garbage bags, diaper backing, dispensing bottles, squeeze bottles, food storage containers, housewares, flexible toys, agricultural films, cable jacketing, tubing, molded laboratory equipment, and coating for cardboard to create a waterproof and heat-sealable carton	Packaging, elastic films, meat packaging, fresh produce packages, garment packaging, diaper backing, agricultural film, food container lids, domestic trash cans, toys, wire and cable insulation, medical tubing, and injection molding	Thin films, packaging, health and hygiene purposes, irrigation pipes, geomembrane, corners, and joint sealing

Data were collected from Refs. [1,2,15–17].

3. PE production and current problems

The world plastics production in 2021 was 390.7 Mt, of which 90.2 % (352.3 Mt) was from fossil-based, 8.3 % from recycled, and only 1.5 % from bio-based plastics (Fig. 2a). For the 21st century and years to come, we can observe that the production of plastics from fossil resources prevails. Therefore, the development of bio-based plastics, their research, and scaling are necessary. Among the continents and countries that produced the most plastics by 2021 are North America (USA, Mexico, Canada) with 18 %, Europe with 15 %, and Asia with more than 50 %. China is the country that produces the most plastics, with 32 % (Fig. 2b). Of the most widely used plastics worldwide, the production was 26.9 % for the different types of PE, PET 6.2 %, PVC 12.9 %, PP 19.3 %, PS 5.3 %, and PU 5.5 % (Fig. 2c). The world's largest plastics markets were packaging, construction, and automotive applications [18,19].

PE is one of the most produced and used plastics, and the amount of recycled and reused is very low; this presents an environmental challenge for researchers and industries since once their life and use time are over, they are discarded, accumulating in landfills and as microplastics in the oceans. Because of this, further study and design of bio-based and biodegradable PE are needed to set standards on PE waste management and recycling globally and mainly in developing countries.

4. Waste management of polyethylene and recycling

Most of the discarded PEs are sent to landfills, which causes accumulation and contamination derived from plastics. PEs are non-biodegradable and contribute significantly to the world's plastic waste products [7,20]. The accumulation of PE and plastic waste in

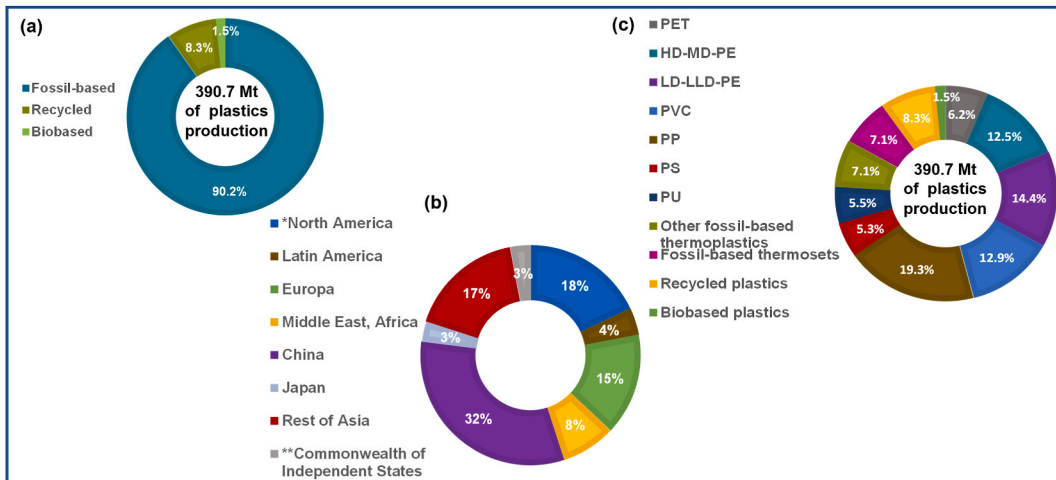


Fig. 2. (a) World plastics production, (b) continents and countries that produced the most plastics, and (c) plastics used worldwide. *Canada, Mexico, and the United States. **Azerbaijan, Armenia, Belarus, Kazakhstan, Kyrgyzstan, Moldova, Russia, Tajikistan, Turkmenistan, Uzbekistan, and Ukraine. Data were collected from Refs. [18,19].



Fig. 3. 3R approaches to responsible waste management.

landfills results in soil contamination, rendering it infertile. Besides, this plastic waste contains additives, retardants, plasticizers, and chemical compounds, which, when burned, release pollutants, toxic chemicals, and greenhouse gases [21,22]. A landfill does not offer suitable and fertile conditions to decompose PE and plastics, such as temperature, mineral medium, moisture, oxygen levels and pH [22,23]. So, even if PE wastes were biodegradable, they would not easily break down, be assimilated and decomposed in a landfill.

Since the problem caused by plastic pollution emerged, recycling has been promoted as one of the solutions to address it. Hand in hand with recycling, the 3R approach to responsible waste management, “reduce-reuse-recycle,” must be followed to contribute to waste management [24] and reduce the accumulation and environmental pollution caused by PE and plastics (Fig. 3). Of all the plastics and polymers produced and discarded; currently, only six plastics are classified and studied to be recyclable, including PE. According to the ASTM D7611, the International Resin Identification Code (RIC) for HDPE is number 2, and for LDPE is number 4.

In general, three stages are important to consider for the recycling and circular economy of PE and plastics [25–27]:

1. A system for collecting the specified PE or plastic.
2. A facility capable of processing the PE collected (industry or laboratory specialized in plastic recycling processes) into a form such as pellets, films, filaments, fibers, or ground, and manufacturers can utilize that to make a new PE or plastic.
3. New plastic products and PE made whole or in a percentage from recycled PE must be manufactured and sold.

Fig. 4 describes the PE product recycling process according to the three stages previously raised. PE is safe and non-toxic in solid form but could be toxic if inhaled and/or absorbed as a vapor or liquid (i.e., during extrusion and manufacturing processes).

5. Degradation of polyethylene

Polymers like PE can undergo degradation through various mechanisms such as chemical processes (oxidation, hydrolysis, or systems involving catalysts), thermal processes (involving high temperatures), mechanical processes (grinding or trituration), or physical processes (ultrasonic radiation, microwave, or sunlight) [6,28]. These processes have the potential to break down the original polymer into smaller molecular weights, generating monomers, oligomers and oils that could serve as potential fuel sources (Fig. 5) [26,29]. However, these degradation processes can come with certain drawbacks. These drawbacks encompass high energy consumption, the requirement of elevated temperatures and pressures, and the use of solvents and additives [6,30]. Additionally, in some cases where the isolation of degradation products is inadequate, polymer fragmentation can give rise to the formation of micro and nanoplastics [31].

One of the most promising and studied methods today is biological degradation (microorganisms) because it is a natural and sustainable process that could occur in the environment under certain conditions, compared to the other methods mentioned [32,33].

Biodegradation is affected by various factors, including the polymer nature, chemical composition, crystalline structure, molecular weight, conditions applied during the process and microorganism type [30,34–37].

PE is an extremely resistant and unreactive material, making it exceptionally challenging to degrade in the environment, even after being buried in a landfill for numerous years. PE resistance to biodegradation can be attributed to the presence of C–C backbone linear carbon atoms, C–C and C–H covalent bond stability, and the lack of functional groups; besides its high degree of hydrophobicity,

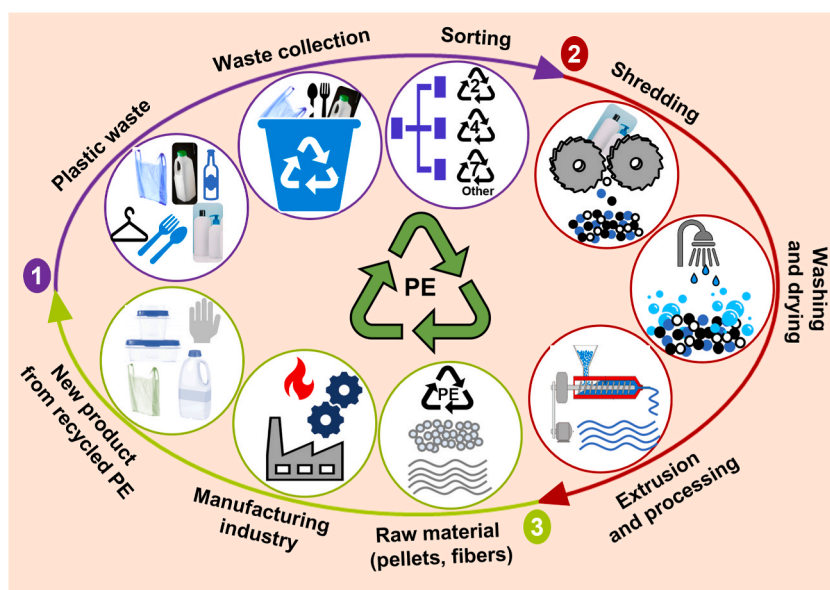


Fig. 4. PE product recycling process.

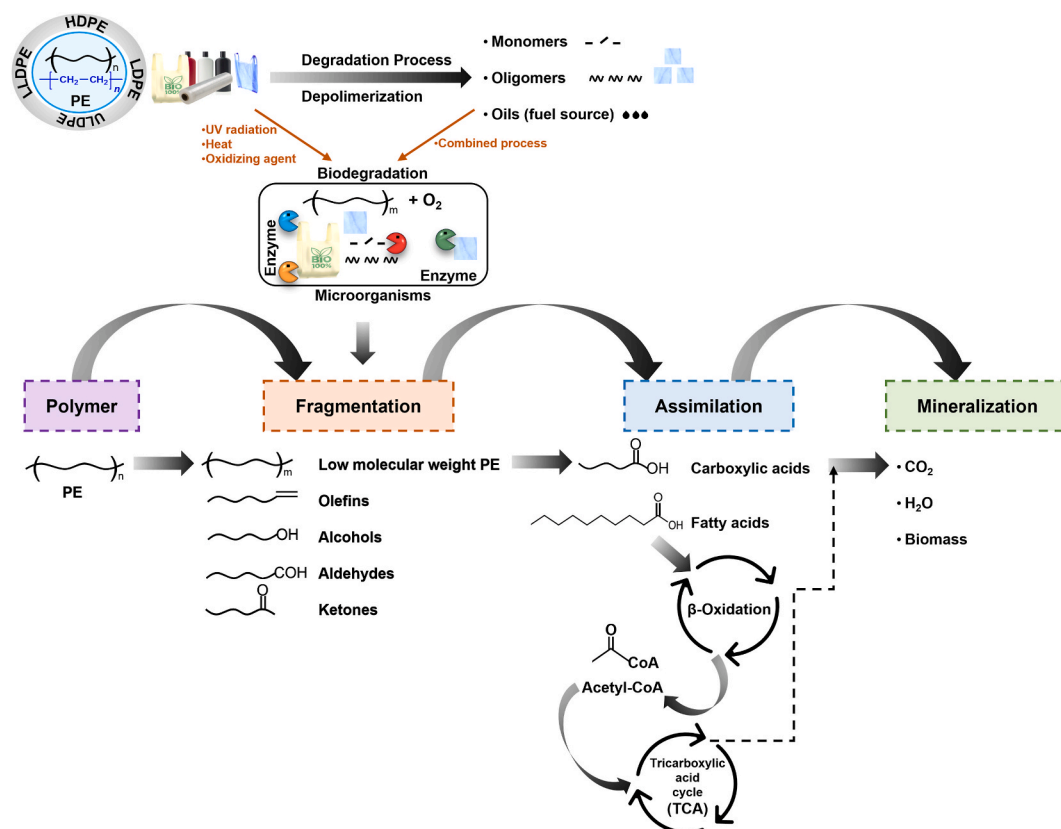


Fig. 5. Proposed mechanism of PE degradation process by abiotic and biotic systems. Four stages are involved in complete biodegradation: abiotic treatment/depolymerization, fragmentation, assimilation, and finally, mineralization.

degree of crystallinity, density and molecular weight and limited specific surface area [38,39]. The physical characteristics of PE can be altered by physicochemical or microbial processes or a hybrid process involving both methods. Physicochemical techniques encompass thermal treatment, UV exposure, oxidation and hydrolysis reactions. UV fragment carbon and hydrogen bonds, liberating free radicals and leading to the formation of smaller PE molecules. UV treatment, oxidizing agents, and hydrolysis reaction reduce the PE chain and form oxidized sites on the PE surface, such as hydroxyl (R-OH), carboxyl (R-COH), or carbonyl (R-CO-R) groups. These treatments alter the surface morphology, crystallinity, and polymer structure while enhancing its hydrophilicity and affinity for bacteria, facilitating polymer biodegradation [30,38–41]. The disadvantage of this hybrid process is that two or more methods must be combined, including biodegradation, which implies that the biodegradation process is no longer as sustainable and natural and difficult to scale.

Biodegradation is a promising method to mitigate plastic contamination and reduce waste, as long as it is used without combining other processes [42]. Still, PE biodegradation is difficult since it involves several biotic and abiotic systems. The collaborative interplay of these abiotic factors and microorganisms leads to PE fragmentation, thereby enhancing surface availability and allowing for the breakdown of bonds and functional group formation for biodegradation [38,39]. Enzymes that can break down phenolic and natural polymers that contain oxidants C-C bonds have played a role in the biodegradation of PE. These enzymes comprise laccases, manganese peroxidase, lignin peroxidases and alkane 1-hydroxylase (enzyme entry: EC 1.10.3.2, EC 1.11.1.13, EC 1.11.1.14 and EC 1.14.15.3, respectively) [43–45]. PE films were reported to undergo biodegradation by laccase derived from the *Rhodococcus ruber*, with a reduction of 15–20 % in the molecular weight, and was observed through FTIR analysis changes in the carbonyl groups; these changes were associated with the important role of the enzyme in the PE biodegradation [46]. The process of biodegrading PE involves several steps, such as dehydrogenation, oxidation and breaking C-C bonds. These steps form carboxylic and acetic acids, which then become integrated and transformed into the tricarboxylic acid cycle (TCA) [30].

In the suggested mechanism for PE degradation, including abiotic and biotic systems (Fig. 5), the complete biodegradation process comprises four stages: abiotic treatment/depolymerization, fragmentation, assimilation, and finally, mineralization. During the abiotic treatment/depolymerization, the PE is reduced to chains of lower molecular weight, unsaturation formations and oxidation (UV, heat, oxidizing agents) to be bio-fragmented, resulting in the creation of oxidized fragments with low molecular weight, like alcohols, aldehydes, and ketones. The enzymes initiate the cleavage of ester bonds by a nucleophilic attack on carbonyl atoms generated during oxidation reactions.

Break bonds of PE into smaller molecules (10–20 carbons) facilitate enzymatic activity and permit smaller fragments to pass through the cell membrane; this allows the intracellular metabolism and assimilation of carboxylic acids via β-oxidation and the

Table 2

Degradation studies of polyethylene, microorganisms, culture conditions, techniques used for characterization, and physical and chemical changes observed.

Type of PE	Microorganism	Source and culture conditions	Experiment duration	Level of degradation (%)	Techniques used ^a	Principal changes observed after degradation	Ref. ^b
HDPE (film)	<i>Bacillus paramycooides</i>	Isolated from a waste mulch recycling plant. Inoculated in an inorganic salt medium and cultured at 35 °C and 150 rpm	45 days	Weight loss of 12 %. Mw decreased 25.96 % and Mn 30.36 % by GPC	FTIR, SEM, XRD, GPC, DSC, WCA, XPS, PCR, gravimetric and 16SrRNA analysis	The surface hydrophobicity of the PE film decreased. Changes in functional groups. The relative crystallinity decreased	[54]
LDPE (film)	<i>Achroia grisella</i>	Waxworm larvae were fed with honeycomb wax and incubated at ambient conditions	30 days	Weight loss of 28.4 %	GPC, FTIR, XRD, SEM CO ₂ production and gravimetric analyses	Changes in functional groups. Reduction in tensile strength. Cavities with a depth of up to 1.2 μm.	[55]
HDPE (virgin film)	Environmental bacterial species	Bacterial isolates obtained from landfill waste. Cultivated on LB agar at 30 °C	30 days	Weight loss from 0.3 to 1.78 %	Gravimetric and GC/MS analyses	The hydrocarbons with single and double bonds were observed. GC/MS showed slow degradation	[56]
LDPE (film)	Bacteria strains	Plastic waste dumpsite soil. Inoculated into mineral salts medium at 35 °C and 150 rpm	56 days	Weight loss from 3.0 to 13.15 %	FTIR, SEM, EDX, and gravimetric analyses	Changes in functional groups. Reduction in the percentage of elemental carbon and an increase in the oxygen	[57]
LDPE LLDPE HDPE (powder)	<i>Tenebrio molitor</i> and <i>Tenebrio obscurus</i>	Yellow and dark mealworms. Liquid Agar and an artificial climate chamber at 25 ± 0.5 °C, 65 ± 5 % humidity	21 days	LDPE decreased between 25 and 28 % for Mn and Mw with both mealworms. The opposite of LLDPE and HDPE showed an increase	FTIR, ¹ H NMR, GPC GC/MS, XRD	The larvae consumed 1.8–2.5 g of PE. Biodegradation processes decrease in the order LDPE > LLDPE > HDPE. Formation of long-chain fatty acids. Low crystallinity and molecular weight and high branching promote biodegradation.	[58]
LDPE (Bag)	Fungal community	Pre-treatment of LDPE Plastic by UV. Fungal isolates from waste disposal soil were inoculated on agar (SDA and PDA) at 28 °C and 150 rpm	30 days	Weight loss from 16.1 to 22.9 %	FTIR, microscopic and gravimetric analyses	The C–H bond deformation in alkenes, ketones, and esters.	[59]
LDPE (sheet)	Marine bacterial consortium	Inoculated on Zobell's Marine agar medium (ZMA) and Minimal Salt Media at 30 °C and 150 rpm	120 days	Weight loss of 47.07 ± 6.67 %	FTIR, ¹³ C NMR, TGA, DSC, SEM, AFM, gravimetric and 16S rDNA analysis	Changes in the functional groups, crystallinity, and thermal properties. An increase in surface roughness and deformities on sheets	[52]
LDPE (film)	Fungal strains	Isolated from municipal landfill soils, mineral salt agar medium, incubated aerobically at 28 °C in static condition	90 days	Weight loss of 38.82 ± 1.08 %	Gravimetric analyses, pH, FE-SEM, FTIR, TGA	New functional groups. Changes in surface and thermal decomposition rates	[60]
LDPE (film)	Bacterial consortia	Bacterial consortia under H ₂ O ₂ biostimulation, mineral medium salt at 35 °C and 150 rpm	12 months	Weight loss from 19.8 to 22.5 %. Mn decreased from 20.1 to 49.5 % by GPC	FTIR, GPC, GC/MS, TGA, AFM, SEM, TOC, and gravimetric analyses	The addition of H ₂ O ₂ stimulated microbial activity. Reduction of Mn, Mz, and Mw by GPC. The morphology and structure of the LDPE films were significantly changed. The biodegradation intermediates were Trimethyldecane, Diethylphthalate, Heptacosane, Octadecane, Phthalic Acid, Butyl Tetradecyl Ester, Eicosane	[61]
LDPE (sheet)	Bacterial strains	Vermicompost, Mineral Salt Medium, incubated at 37 °C.	30 days	A negligible reduction in the weight of the LDPE (value not reported)	pH, FTIR SEM, gravimetric and 16S rRNA analysis	The peak changes (formation and shifts). Structure and surface changes	[62]
LLDPE (film)	Two <i>Pseudomonas</i> bacteria strains	Incubated in a rotary shaker at 28 °C, 180 rpm, pH 6.8–7.0	8 weeks	Weight loss from 3.62 ± 0.32 to 5.95 ± 0.03 %	FTIR, SEM, AFM, WCA, gravimetric and	Pits and wrinkles were observed. The hydrophobicity decreased,	[63]

(continued on next page)

Table 2 (continued)

Type of PE	Microorganism	Source and culture conditions	Experiment duration	Level of degradation (%)	Techniques used ^a	Principal changes observed after degradation	Ref. ^b
LDPE (film)	Fungal community	Plastic wastes from landfill soil, cultured on malt extract agar at 24 °C	6 months	Data not reported	16S rDNA analysis ATR-FTIR, SEM, PCR	and oxygen-containing functional groups were identified Strong oxidation phenomena and changes in the PE film morphology	[64]
LDPE (sheet)	<i>Ralstonia</i> sp. Strain SKM2 and <i>Bacillus</i> sp. Strain SM1	Municipal waste landfill soil, inoculated in broth media at 30 °C and 105 rpm	180 days	Weight loss of 39.2 % (SKM2) and 18.9 % (SM1)	FTIR, compound microscope, pH, PCR, 16S rRNA and gravimetric analyses	An irregular surface in LDPE, such as roughness, pits and cracks. Changes in the C–C bonds and functional groups.	[53]
LDPE (film)	Marine bacterial strains	Bushnell Haas (BH) medium with 3.0 % NaCl at 150 rpm and 30 °C	90 days	Weight loss from 0.97 to 1.72 %	ATR-FTIR, FE-SEM, AFM, TGA, carbon analysis, CO ₂ trapping, gravimetric analyses	An irregular surface in LDPE, such as roughness, pits and cracks. Changes in the chemical structure, shifting and decrease in absorbance of the peaks by FTIR. Consumption of carbon. CO ₂ evolution	[65]
HDPE (shopping bag)	Microbial strains	Microbial strains from soil, sludge, and worms. Incubated on a shaking incubator at 30 °C and 110 rpm	100 days	Weight loss from 2.5 to 5.5 %	FTIR, SEM, AFM, gravimetric and 16S rRNA analysis	Formation of functional groups (carbonyl and hydroxyl). Decolorization and surface deterioration	[66]
LDPE (particles)	<i>Microbulbifer hydrolyticus</i> IRE-31	Marine wastes. Maintained in a marine broth and incubated at 37 °C and 220 rpm	30 days	Data not reported	FTIR, SEM, 16SrDNA analysis	Morphological changes and formation of carbonyl groups	[67]
LDPE (foam)	<i>Galleria mellonella</i> Larvae	Wax Moth Larvae. The larvae were fed food containing mixed nutrients, including honey, beeswax, and bran, and placed in an incubator at 27.5 °C and with a humidity of 75 %.	21 days	Mass loss of 1.95 g	FTIR, GPC, GC/MS, TGA, gravimetric and 16S rRNA analysis	Formation of functional groups. Increase in the molecular weight (Mw, Mn) and formation of long-chain carboxylic acid esters	[68]

^a FTIR (Fourier transform infrared spectroscopy), GPC (gel permeation chromatography) Mn and Mw(number and weight average molecular weight by GPC), GC/MS (gas chromatography/mass spectrometry), NMR (nuclear magnetic resonance), SEM (scanning electron microscopy, AFM (atomic force microscopy), TGA (thermogravimetric analysis), DSC (differential scanning calorimetry), XPS (X-ray photoelectron spectroscopy), XRD (X-ray diffraction), TOC (total organic carbon), WCA (water contact angle), PCR (Polymerase Chain Reaction).

^b Data were collected from the last three years of publication.

tricarboxylic acid cycle. Finally, these compounds are mineralized into CO₂, H₂O and biomass [30,28,38,39,47–49].

Over the past few decades, more than 100 species exhibiting PE degradation activities have been isolated, encompassing bacteria, fungi, insects, and microbial communities [48]. Table 2 shows some of the latest research in the last three years on biodegradation studies in PE. We can observe that most of the studies are carried out with LDPE due to its properties, low density, crystallinity, and molecular weight, which allows this type of PE to be attacked by microorganisms, and a greater biodegradation occurs. We can see that the weight loss ranges from 1 to 47 %. But until now, it has not been possible to biodegrade more than 50 % or thoroughly the PE into biomass, CO₂, water and minerals by biological processes or what is established by the standards ASTM D6400, ASTM D5988, ISO 17556 and EN 13432 for biodegradable plastic materials [50,51]. Joshi et al. [52] reported a weight loss of 47 % for a biodegradation study of LDPE sheets using a Marine Bacterial Consortium for 120 days, observing changes in the crystallinity, functional groups, and LDPE surface (Table 2). Biki et al. [53] reported a weight loss of 39.2 % for LDPE sheets using *Ralstonia* sp. strain SKM2 from municipal waste landfill for 180 days, observing an irregular surface in LDPE such as roughness, pits and cracks.

Unlike LDPE, LLDPE and HDPE are more recalcitrant due to their chemical structures and properties. We can see in Table 2 that only some works have been reported on these types of PE. In addition, the mass losses are lower and range from 0.3 to 12 %. Most changes after biodegradation are associated with changes in functional groups and surface deterioration.

Yang et al. [58] studied the biodegradation processes of LDPE, LLDPE and HDPE using *Tenebrio molitor* and *Tenebrio obscurus* for 21 days and reported by GPC analysis that LDPE decreased between 25 and 28 % for Mn and Mw with both mealworms, the opposite of LLDPE and HDPE that showed an increase in Mn and Mw, this associated with the distribution of molecular weights and polydispersity indices during biodegradation.

The biodegradation decreased in the order LDPE > LLDPE > HDPE, in addition to the low crystallinity and molecular weight and high branching promoting PE biodegradation [58]. At present, eight different characteristics are usually monitored as changes to

establish the level of degradation of the PE [29,42,51]:

- Changes in functional groups
- Molecular weight (M_n , M_w and PDI)
- Degree of crystallinity
- Surface features
- Hydrophobicity/hydrophilicity
- Mechanical properties
- Thermal properties
- Polymer consumption (weight loss)

6. Bio-based polyethylene and Bio-PE composites

Since the uses and applications of PE are uncountable due to their well-known mechanical properties, chemical inert, and good resistance behavior [12], it contributes to large contamination in several environments such as soil [69], water [70], and even crops [14].

The difficulty associated with PE utilization often arises from its inclination toward single-use materials; this situation intensified when individuals prioritized these materials over reusable alternatives and environmentally friendly [12,14]. As a result, plastics began to proliferate in the environment since the 1970s, marked by a significant surge in the ratio between production and deposition; this led to an accumulation issue primarily stemming from PE's resistance to degradation under typical ambient conditions [69–71].

This accumulation problem originates from a new research line in the study of biomodified plastics using natural bio-based materials in preparing PE blends. The produced composite is well-known as “bio-based PE” since the main constituent for the reinforcement is the natural or biological materials from agro-industrial residues (Fig. 6). Besides, another group of composites named “bio-PE” refers to PE obtained from natural sources by anaerobic fermentation, distillation, and copolymerization to obtain PE [12,14,38]. This idea has viability since the degradation of PE could be increased using different polymer pre-treatments, such as thermal, photo and chemical oxidation [38].

However, another alternative to increase PE degradation in the natural environment involves grafting, copolymerization, and blending it with functional polymers and compounds [72]. The chemical interaction of PE between a few additives named compatibilizers with hydrophilic groups (i.e., stearic acid, maleic anhydride, PDMS) generates less hydrophobicity in the plastic and the biodegradation treatments could have a better susceptibility in the samples [73]. For bio-based PE, the most affordable approach is the microbial assimilation of the filler, starting the process of microbial attack in the hydrophilic part of the composite, which increases the availability of the synthetic material's surface area accessibility; this, in turn, renders it more susceptible to both abiotic and biotic oxidation [72,74]. After this, the main inert components of the composites should disintegrate and disappear, but only a few studies are based on these results [38,75,76]. Since the last decade, several polymers have been used as degradable fillers and structure modifiers with enhanced results in microbial biodegradation, like polyester, isolated natural compounds, such as cellulose, chitosan and starch, and synthetic plastics from oil, such as polylactic acid and polycaprolactone [14,77,78]. Despite several advantages such as good biodegradability, facile incorporation, easy isolation, and natural availability [38,79], the biodegradability and compostability

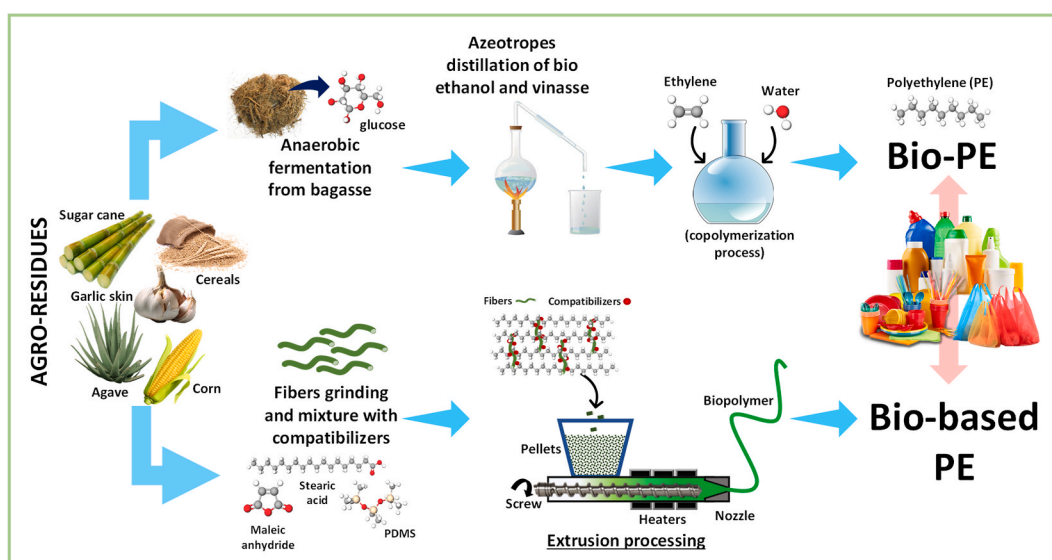


Fig. 6. Approaches of bio-PE and bio-based-PE from agro-industrial residues to the production of bioplastics in different disposable applications.

performance of the resulting composites are still unknown, and authors report only the enhancement in the physical and chemical properties, without a more detailed investigation of the environmental-friendly behavior of the material.

Some researchers have started implementing a conscious methodology to modify the PE chains chemically to reduce the biodegradation time of the material in ambient conditions [14]. This idea was proposed after the study of microbial degradation of LDPE using *Aspergillus clavatus* in controlled conditions, showing a noticeable reduction in the degradation time until 90 days [80]. However, the initiative became noteworthy when concern about the term 'biodegradability of polymer' emerged.

Polymer scientists defend the idea of degradability as the decrease or change in the chemical and physical properties; meanwhile, microbiologists associate degradation with the full mineralization of material. This discussion follows a new era in which both scientific groups start looking for convergence between both ideas but following a new principle of circular economic or environment-friendly materials that considerably reduce the emissions of contaminants to the environment [81]. Several researchers initiated the mixture of PE with bio-based reinforced materials to test the material's capabilities to become degradable without changing the intrinsic properties of the PE (Table 3). In the last decade, Brito et al. [82] investigated a PLA/Bio-PE blend prepared with two different compatibilizers for better cross-linkable properties, showing that both copolymers form a very stable ternary blend that shows an enhancement in mechanical performance and a noticeable decrease in the melt flow index which confirm the formation of PLA-compatibilizers copolymers. Castro et al. [83] and Kuciel et al. [84] reported the performance of bio-based PE after adding natural fibers to enhance the stiffness of the new materials. First, Castro et al. [83] evaluated the preparation of films using bio-HDPE obtained from sugarcane ethanol and curaua fibers. After mixing and thermopressing, the films were coupled with a compatibilizer agent (hydroxyl-terminated polybutadiene). The presence of the reinforced fibrillar material improved its strength and modulus. Finally, the authors explain that using different processes, like extrusion and injection molding, was better than mixer and thermopressing for film impact resistances. Second, Kuciel et al. [84] investigated bio-composite materials created from Bio-PE, which is synthesized using ethanol derived from sugarcane and reinforced with four distinct natural fillers (25 %w/w).

These composites were produced through two processes involving extrusion molding and injection molding. The authors demonstrated the incorporation of natural fillers into bio-PE allows the production of lightweight, structural, eco-friendly products with physical and economic improvements. More recently, Bello et al. [85] reported using wastepaper to formulate a biocomposite made with waste HDPE. The study evaluated the reinforced properties of PE with the wastepaper through physical, static, and dynamic mechanical properties. The authors reported that water absorption increment as the filler content was altered due to the filler material nature (hydrophilic); however, the mechanical properties (tensile stress and strain) were reduced, producing changes in the modulus of elasticity and the glass transition behavior. These results proved the new concept of changing properties by waste reuse and recycling to interesting sustainable engineering.

Table 3

Comparison of several bio-based PE preparations for blends and films using different reinforced materials.

Initial composition	Reinforced material	Compatibilizer and coupling agents	Temperature of processing	Processing of the composite	Applications	Reference
HDPE	Date palm flour and microcrystalline cellulose	N/A ^a	175–185 °C	Melt mixing and compression molding	Construction, automotive, sports, and other industries	[74]
LDPE	Polylactic acid (PLA)	Lotader™ AX8900 and AX8840	170–180 °C	Co-rotating twin-screw extruder and injection-molding	Optimization of plastic compatibilizers	[82]
HDPE obtained from sugarcane ethanol	Curaua fibers	Hydroxyl-terminated polybutadiene	(1) 160 °C (2) 160–190 °C	(1) Extrusion and injection molding (2) Mixer and thermopressing	Plastic production and optimization	[83]
Bio-PE from sugarcane bagasse	Wood flour, kenaf fibers, cellulose powder, and tuff particles.	N/A*	160–175	Extrusion and injection molding	Thermal resistance and the plastic industry	[84]
HDPE	Wastepaper	Silicon oil	150 °C	Carver compression machine	Thermal resistance materials	[85]
HDPE	Dried fibers from rice husk, cotton stalk, sugarcane bagasse, bamboo, and straw come from rice and wheat	Maleic anhydride, grafted PE and PA03 (lubricant)	155–165 °C	Extrusion processing	Construction and building materials.	[86]
Bio-PE	Corn cob residues	Stearic acid and PE wax. Polypropylene, grafted PE, maleic anhydride	140–175 °C	Co-rotating twin-screw extruder	The plastic industry with circular economy	[87]
HDPE	Eucalyptus fibers	Grafted PE and maleic anhydride	180 °C	Injection molding	Plastics, automotive, packaging, and construction industries.	[88]
LDPE	Jute, corn silk, and bagasse fibers	Maleic anhydride and ferric stearate (pro-degradant)	160–180 °C	Injection molding	Fire-resistant materials, packing	[89]

^a N/A: no applied.

Mu et al. [86] reported similar results using natural fibers such as rice husk, cotton stalk, sugarcane bagasse, bamboo, and straw come from rice and wheat; these fibers were chosen as reinforcement components in the manufacturing of a natural fiber/polymer composite via extrusion processing. The authors used a composite preparation methodology based on dried fibers, HDPE, a compatibilizer agent (maleic anhydride), and PA03 as a lubricant. The results indicated that incorporating natural fibers into HDPE enhances the mechanical properties of composites more than three times compared with raw PE. As previous authors, Bello et al. [85] proposed that agricultural residues as a potential material reinforce PE composites with probable uses in construction and building materials.

However, other authors try to improve the economic feasibility of some residues, as Chen et al. [87] reported for the corncob biorefinery process residues used as reinforcing materials in the preparation of bio-PE plastics employing a co-rotating twin-screw extruder. The authors prepared a composite with PE and corncob processing fibers, compatibilizer as stearic acid, and PE wax as coupling agents. The previous results proved that these works have potential applicability by using solid residues of biorefinery processes to make the waste profitable.

Abed et al. [74] presented a study of biodegradable PE-based biocomposites reinforced with date palm fibers and microcrystalline cellulose. The authors proposed a coupling process using melt mixing and compression molding to obtain the samples. The results reveal an environmentally friendly solution for reducing plastic pollution without using any compatibilizer of coupling agent.

Ouarhim et al. [88] used HDPE reinforced with natural fibers of eucalyptus. These fibers were treated with an alkali (NaOH and CH₃COOH), and the authors reported a notable improvement in the physical interaction with the matrix, the dispersion of fibers, and the transfer of interfacial stress in the material.

On the other hand, Elgamsy et al. [89] employed LDPE and different natural fibers (jute, corn silk, and bagasse) to synthesize a bio-based PE. They proved the usage of ammonium polyphosphates as a retardant agent to change not only the water absorption and mechanical properties but also the flammability, which was demonstrated in the final thermal properties of the materials. These authors used maleic anhydride as a coupling agent and injection molding to form bio-based PE composites.

7. Composting studies in polyethylene

Since the PE film only biodegrades over a long period through natural agents, other methods are available to increase the degradable character under specific conditions. Some of them could be sunlight, moisture, oxygen, and composting, which tend to enhance biodegradation by increasing the hydrophilic properties of the films and decreasing their hydrophobic characteristics [38,79,90]. However, a film's biodegradable and compostable behavior must be remembered when environmental problems must be quantifiable [14]. Particularly, a material is considered "biodegradable" when it partially degrades to a reduced form of the original material; this means some parts remain constant in the environment even when long composting periods are applied [38,91]. On the other hand, compostability emerges as the perfect definition of the actual bioplastics to be originated based on environmental needs. Nowadays, the UNE EN 13432:2001 standard norm (comparable with ASTM D6400) claims a "compostable" material only if the material accomplished four definitions: i) degradation of at least 90 % in weight must be achieved in an enhanced CO₂ environment for six months; ii) The size of the mass must be reduced by 90 % in three months if it is in contact with organic materials; also the mass fragments must be < 2 mm. iii) In the composting process, there should be no negative or secondary effects; iv) the limits of heavy metals present in the compost must be specified.

Composting is a dynamic process involving four complex phases. The changes in microbial growth and composition must be evaluated continuously to quantify the compostability process. The four phases are mesophilic, thermophilic, cooling, and maturation, in which the temperature changes and microbiota composition are the two variables that promote the degradation of the organic matter until a complete biosorption in the media [92,93]. Some of the most recent advances in the composting process for PE are found when the blend or the final product is a mixture of PE and some reinforced materials, which brings a more hydrophilic character to the non-degradable plastics. Recently, Singh et al. [94] evaluated the biodegradable behavior of commercial PE films in a natural composting environment over six months. The experiment was made in a compost burial test and monitored using spectroscopy and microscopy techniques. The authors reported that only a major change in the surface of PE was achieved after six months of compost exposure, with a total weight reduction of 17 %. From this study, the authors claimed that the compost burial test is a reliable method without ecological hindrances in natural ecosystems. Another example was found in the research of Vieyra et al. [95] for a biodegradable material prepared with PE and unripe banana flour; the blend exhibited a significant loss of weight, as well as a decrease in its tensile resistance and the moiety of 40 % of its weight in only 75 days of composting. The authors conclude that blends prepared with unripe banana flour and PE and controlled inoculated landfills with *Mortierella elongate* could be a promising technology to improve the big issue with plastic disposal in landfills.

Recently, Álvarez-Vega et al. [92] studied the PE changes in the chemical, physical and biological properties due to composting with rose waste. The samples were composted for 124 days, but the results were detected as insignificant. The authors confirm that small changes were observed since only mesophilic and not thermophilic fungi were found during the composting period. These results showed that the fungi population is a key factor influencing the slower degradation rate of total organic carbon and organic matter if not presented in the correct amount. Martínez et al. [96] reported a study using different types of commercial PE film in composting under domestic and ambient conditions. For the most reliable degradation analysis, the samples were exposed to radiation by UV-B. The authors explain that UV-B originated photo-oxidation into the material, allowing the formation of unsaturations and carbonyl groups, which causes the formation of polysaccharides after domestic composting. This change was corroborated by spectroscopy and microscopy techniques that confirmed the formation of a hydrophilic surface modification onto the samples. The authors claimed that PE biodegradability is enhanced caused by UV-B. However, despite this, the residue of the material persists in the environment as undesired polymer fragments, such as microplastics, thereby contributing to another environmental pollution issue. Besides that, the

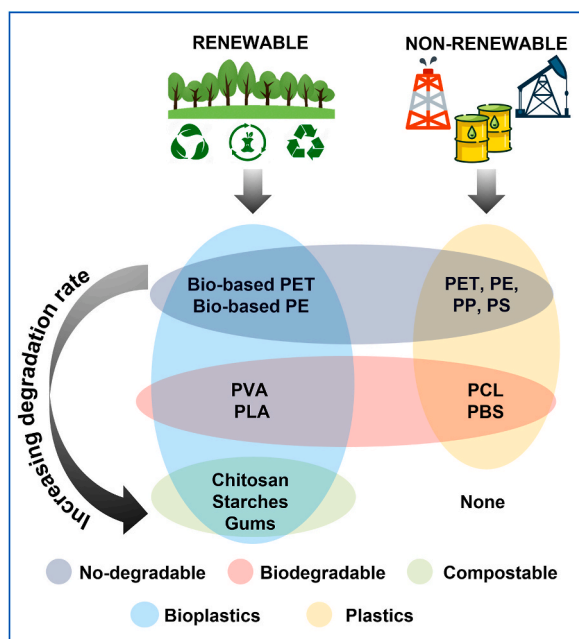


Fig. 7. Representation of the renewable and non-renewable polymers grouped depending on the non-degradable, biodegradable, and compostable behavior. The increasing degradation rate shows the tendency that the authors are following nowadays. PET: poly(ethylene terephthalate); PE: polyethylene; PP: polypropylene; PS: polystyrene; PVA: polyvinyl alcohol; PLA: polylactic acid; PCL: polycaprolactone; PBS: polybutylene succinate.

biodegradation of PE through domestic composting is conditioned by external factors like relative humidity and temperature.

Based on the results presented before, a profile of the composting experiments that could be implemented in the investigation of PE should be based on several parameters such as i) a group or individual microorganism that provokes numerous damage in the plastic; ii) a setting experiment with enough sunlight or artificial energy that generate a challenging environment for the degradation of the material; iii) the implementation of bio-PE or bio-based PE since it is well known that this material has more affinity to degradation in composting conditions; finally, iv) the uses of some material that improve the rates of degradation such as catalyst or compounds richer in the hydrophilic group as the compatibilizers explained before.

However, it is important to clarify that nowadays, efforts to find a perfect bioplastic that can be used in different applications, saving the environment without further damage to the ecosystem, are related to compostable materials from renewable sources [97–100]. Some can be found in cellulose, chitosan, starch and gums, exhibiting favorable environmental interactions and a promising and extensive application in bioplastics worldwide (Fig. 7).

8. Conclusions and perspectives

New trends in the development of polymeric materials with more environmentally friendly features are the joint objective of the development of bioplastics. However, a more environmentally friendly material must meet certain characteristics that allow its use without generating more contamination or avoiding its proliferation in clean or pure environments. The idea is to create less waste that pollutes the environment since it has been shown that microplastics are generated from non-degradable polymers that could harm health.

Transitioning to bioplastics is a prudent choice, given their renewable nature, degradability, eco-friendliness, and sustainable characteristics in contrast to plastics derived from petroleum sources. PE is non-biodegradable and contributes significantly to the world's plastic waste products. However, some factors could help to reduce the accumulation and contamination derived from PE and its degradation rates, such as the 3R methodology and recycling approaches, the chemical and structure modification, and the synthesis and use of bio-based PE and bio-PE; this will enable a new vision of bio-based-PE that is still being developed to reduce single-use petroleum-derived plastics.

Finally, considering that PE is an inert and highly recalcitrant material, further degradation and biodegradation studies of the different types of PE are needed since the most studied is LDPE because of its properties, low density, molecular weight and crystallinity. In addition to the fact that if a PE is bio-based, this does not make it biodegradable or compostable, requiring studies for its classification.

Perspectives:

- Standard norms that demand the use of bio-PE, bio-based PE or the incorporation of recycled PE for the production of single-use PE materials are needed. Since chemical modification and structure of PE could enhance the degradation level.

- Studies and techniques are needed to evaluate the formation of micro and nanoplastics released from degradation processes.
- Further research is required to explore reaction pathways and potential degradation products of PE and plastics under conditions that simulate the real-world environment.
- Additional research is necessary to explore the composting of PE under ambient conditions or in-home composting settings; this entails identifying the microorganisms and enzymes engaged in biodegradation across various compost types, decomposition time, and the byproducts formed.

Data availability statement

No data was used for the research described in the article.

Additional information

No additional information is available for this paper.

CRedit authorship contribution statement

Manuel Burelo: Conceptualization, Investigation, Project administration, Validation, Writing – original draft, Writing – review & editing. **Josué David Hernández-Varela:** Conceptualization, Investigation, Writing – original draft, Writing – review & editing. **Dora I. Medina:** Conceptualization, Investigation, Writing – original draft, Writing – review & editing. **Cecilia D. Treviño-Quintanilla:** Conceptualization, Investigation, Project administration, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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