Chapter 14
Oxo-biodegradable Plastics: Who They Are and to What They Serve—Present Status and Future Perspectives

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The worldwide annual consumption of polymeric materials and relevant plastic items is nowadays well above 300 million Tons, of which less than 2 million Tons is represented by polymeric materials from renewable resources. These last materials, when converted to relevant plastic manufactured goods, are identified under the ambiguous chimeric "bioplastics" terminology as evidenced in official EU documents [1, 2].

The attribute "chimeric" that we did use in identifying the "bioplastics" is due to the fact that all plastic items are "man-made" whatsoever their ultimate service life as either commodities or engineering technoplastics, or thermosetting.

Beyond the suggestive ambiguity of the terminology, it is worthwhile highlighting that the term "bioplastics" is not even comprised in the EU Technical Report relevant to the nomenclature concerning degradable and biodegradable polymeric materials and relevant plastic items [3].

The data of plastic production mentioned in the first paragraph have been detected from the histograms reported in Fig. 14.1 where (a) section is referred to the trend of the worldwide production of polymeric materials and relevant plastic items recorded since the half of the past century [4] whereas in (b) section it is reported the growing trend of the production of polymeric materials from renewable resources convertible to the relevant "bio-based" plastic items since year 2008 [5].

It is to remark that about 40 % of the worldwide production of polymeric materials is converted to relevant plastic manufactured goods intended for short service life, which under the attribute of "commodities" are meant to satisfy the needs of packaging for applications in the food and non-food segments, including also the production of single-use items (disposables) as both flexible and semiflexible, and rigid items.

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Springer Series on Polymer and Composite Materials,
DOI 10.1007/978-3-319-25982-6_14

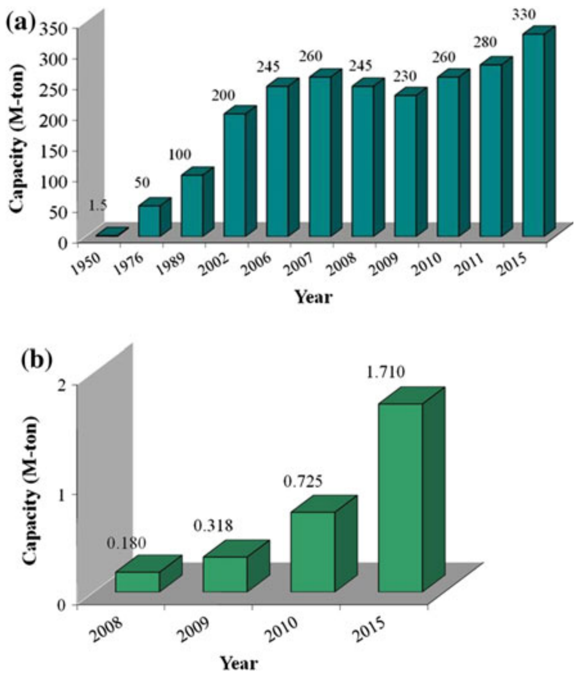

Fig. 14.1 **a** Global plastic production in 1950–2015 time frame. **b** Global production capacity of bio-based polymers in 2008–2015 time frame

Since the middle of the past century, after the various ages characterized by the use of inorganic raw materials (stone, iron, silver, and gold), we are nowadays right in the middle of the plastic age, that is based on the utilization of organic matter as raw materials susceptible to be used for the production of thermoplastic and thermosetting plastic items.

It is expected that the people have to live together for several years to come with plastic manufactures of broad use as they are nearly unsubstitutable, and permeate, with growing impact, all the types of anthropic activities which are expected to expand as a consequence of the worldwide population increase (Fig. 14.2).

This aspect, as easily understandable, implies an effective increase in the production of feed and food and hence of commodities, with all the associated risks of negative impact that may derive from their incautious release in the various environmental compartments (Fig. 14.3).

One cannot, however, ignore a serious problem relevant to the production of short-service-life plastics, mainly identifiable as commodities due to their progressive accumulation in the various environmental compartments, as not only for their incautious abandoning in the environment, but also as for the lack of an efficient capillary-organized collection of industrial and domestic waste. It cannot be also forgotten that the indicated problem is of global significance. In fact, a short-service-life plastic commodity item incautiously abandoned in a South African coastal area through free floating into marine compartments can end up on beaches of north European countries or North America [6, 7].

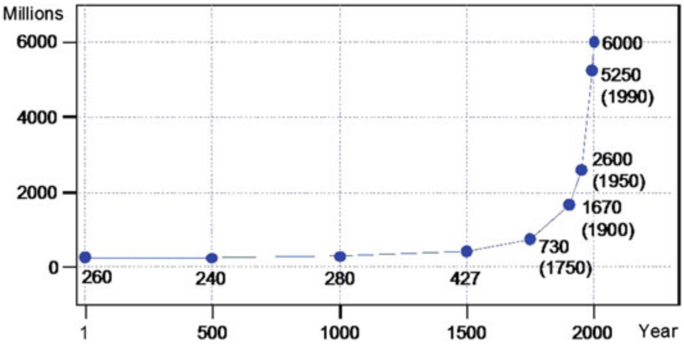

Fig. 14.2 Growth of world population

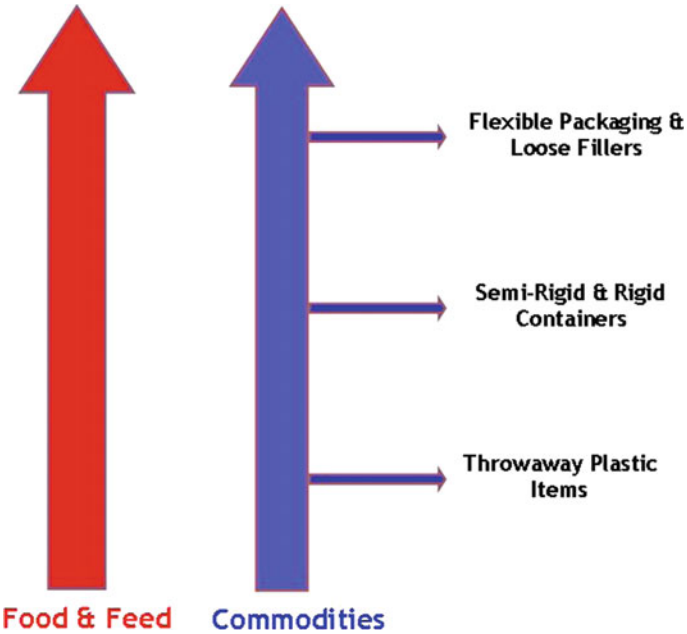

Fig. 14.3 Growing of needs versus growing of population

The plastic waste abandoned in the environment is holding a transnational citizenship, and hence, it is not easy to manage it in order to mitigate its potential deleterious effects on the environment.

The environmental accumulation of plastic waste in the last decades is simply due to the fact that the plastic commodities, as obtainable from the unsubstitutable polymeric materials based on the fossil fuel feedstock. These are consisting of full-carbon backbone macromolecules, which are characterized by degradation

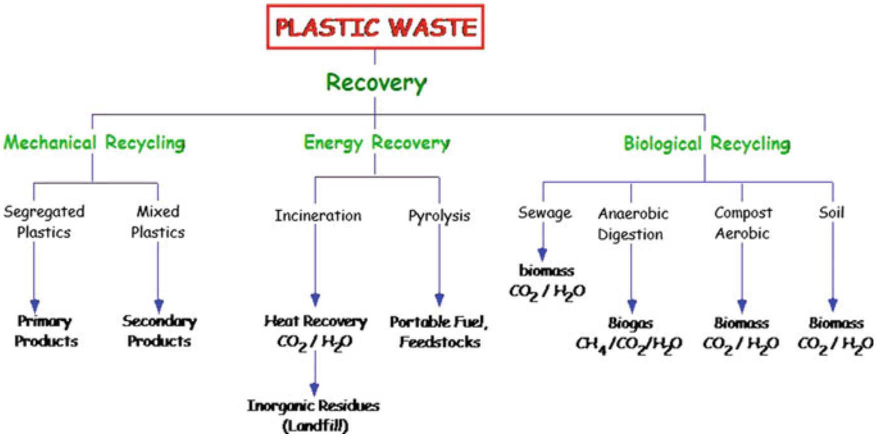

Fig. 14.4 Plastic waste management options

times and hence possibly of biodegradation in different environmental compartments, extremely long of the order of tens of decades if not of centuries [8].

Therefore, timely decisions are to be taken as aimed at promoting and approving political directives and supporting research activities focused on facing and solving the above-indicated issues by suggesting concretely viable solutions compatible with the socioeconomic development embracing effectively the worldwide global scenario of a sound plastic waste management with options as sketched in Fig. 14.4.

In this context, particular attention has to be paid to the developing countries, and countries in transition, as faced a few years ago in a program promoted and supported by the United Nation Industrial Development Organization (UNIDO) through the International Centre for Science and High Technology (ICS) established at Padriciano (Trieste, Italy) and implemented after the takeoff in year 1996 for more than a decade (Table 14.1) [9].

Even the present author (E.C.) has been pleased to contribute actively to the dissemination of the fundamental concepts relevant to the design and development of polymeric materials and relevant plastic items characterized by eco-compatibility and biodegradability under compelling suggestions on the correct management of postconsumer plastic commodities at the end of their service life.

An approach aimed at providing a viable solution to the issue of plastic accumulation in the environment has been focusing on the chimeric “bioplastics” as friendly substitutes of the commodity plastic items based on fossil fuel feedstock. In spite of the remarkable efforts spent in the design and production of plastics from renewable resources (Fig. 14.5), correctly identifiable as “bio-based” plastics, as anticipated their global production is, to be generous, extremely limited (around 0.5 %) of the worldwide plastic production based on fossil fuel feedstock.

As far as the production, consumption and management at the end of the service life of biodegradable plastics in general are concerned, from both petrochemical and

Table 14.1 Formative activity program on eco-friendly polymeric materials and biodegradable plastics promoted by ICS UNIDO in emerging and developing countries

December 1996	SPCM	Trieste, Italy	September 2002	EGM	Trieste, Italy
June 1997	TC	Alexandria, Egypt	October 2002	WSP	Bangkok, Thailand
November 1997	WSP	Pune, India	October 2002	WSP	Beijing, China
April 1998	EGM	Trieste, Italy	November 2002	WSP	Santiago, Chile
September 1998	WSP	Antalya, Turkey	July 2003	EGM	Trieste, Italy
November 1998	WSP	Campinas, Brazil	April 2004	WSP	Teheran, Iran
March 1999	WSP	Doha, Qatar	October 2004	WSP	Beijing, China
September 1999	WSP	Shanghai, China	November 2004	WSP	Kampala, Uganda
October 1999	WSP	Smolenice, Slovakia	December 2004	EGM	Trieste, Italy
March 2000	WSP	Sharjah, UAE	November 2005	WSP	San Jose, Costa Rica
September 2000	WSP	Seoul, Korea	December 2005	EGM	Trieste, Italy
November 2000	EGM	Trieste, Italy	February 2006	EGM	Trieste, Italy
June 2001	WSP	Lòdz, Poland	June 2006	WSP	Belgrade, Serbia
September 2001	WSP	Jakarta, Indonesia	December 2006	WSP	Pune, India
November 2001	WSP	Manama, Bahrain	October 2007	WSP	Beijing, China
December 2001	EGM	Trieste, Italy	March 2008	WSP	Cairo, Egypt

SPCM Scientific Planning Committee Meeting, *TC* Training Congress, *WSP* Workshop, *EGM* Expert Group Meeting

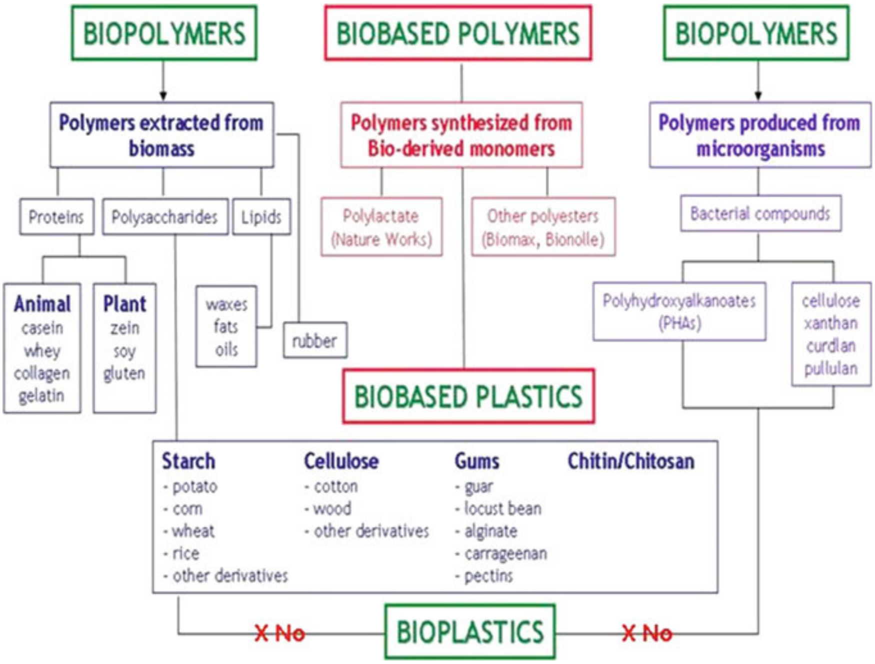

Fig. 14.5 Natural and derived polymers and relevant plastics—nomenclature

renewable resources, it is indeed worthwhile mentioning the "First International Scientific Consensus Workshop on Biodegradable Polymeric Materials and Plastics‶ [10]. That event dating back to the end of the 1980s recorded the international participation of researchers from a large number of countries, in an appreciable interdisciplinary expertise framework. The worldwide market of the biodegradable plastics from renewable resources is characterized by a productivity growth rate not commensurate, in our opinion, to the academic and industrial research investments aimed at their take-off and acquisition of reasonable quotas in the plastic commodity and disposable increasing demands.

Therefore, the outstanding recent efforts spent in industrially oriented programs aimed for the implementation of a "green chemistry" that the present authors would dare to define a reargued matter, it is not in the position to give fair answers to the growing needs for feed, food, and plastic commodities.

It is worthwhile mentioning that according to the statistic data issued by the IBAW Industrial Association, the production and consumption of "bio-based" plastics was 50 K Tons in year 2004, and two years later in spite of the rosy perspectives, only a production of 150 K Tons was recorded [11], and nowadays after a decade, it has reached a production level of one order of magnitude higher but maintaining, however, the same proportional gap with respect to the fossil fuel feedstock-based plastics [12] (Fig. 14.1).

As a case study aimed at mitigating the environmental burden caused by conventional polyethylene-based shopping bags, under this stimulus of the novel ecologist fervor born by the end of the 1980s in Italy, a decree with the force of the law was issued [13] and converted successfully in law [14], according to which a tax of 100 Italian Liras—equivalent to 5 €-cents—was charged on any conventional polyethylene (PE) shopping bag with the scope of limiting their consumption on the national territory.

Almost ten years later, that law was revoked in Italy by a decree with the force of the law issued by the Ministry of Environment [15]. The indicated legal positions gave a new input to the production of bio-based plastics as suited to cover the mercantile segments of massive consumption such as the packaging which comprises also the shopping bags.

According to the above considerations, we welcome polymeric materials and relevant plastics susceptible to be biodegraded under composting conditions, according to the Standard Norm EN 13432/02 [16] and relevant specification reported in the Norms EN 14995-06 [17] and ISO 17088 [18], provided they are derived for instance from agro-industrial waste and renewable resources that do not interfere with food and feed segments, such as agro-industrial waste, and are not intended to build up monopolies of materials and relevant manufactures against the elementary rules of commercial competitions without limiting the discretion liberty of the customers and refraining the free circulation of goods stated as safe for food contact.

It is then taken for granted that beside the biodegradable/compostable polymeric materials and relevant plastic manufactures, whatsoever the raw material used in their production, either fossil fuel feedstock or agro-industrial waste, it is necessary that plastic commodities' massive utilization derived from fossil fuel feedstock that

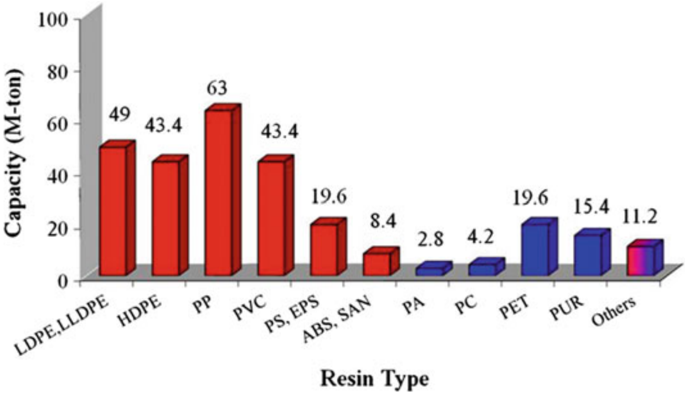

Fig. 14.6 Global plastic production in 2011 by type

continue to be produced eventually after a reengineering of their original formulations, in order to make them eco-compatible. In fact, if one makes an analytical assessment of the typology of the polymeric materials currently used for the production of massive plastic manufactured goods can realize that nowadays about 80–85 % is consisting of macromolecular components characterized by a full-carbon backbone structure (Fig. 14.6), and polyolefins are the major components of these productions.

These last materials and relevant manufactures are recalcitrant to the attack and digestion by microorganisms present in various environmental compartments, with the exception of microorganisms equipped with a genetic profile susceptible to exert a pro-oxidant activity such as those reported in Table 14.2 [20]. However, we have to take into account that due to the high molecular weight of the macromolecules, the oxidation rate of the carbon backbone in the formal average oxidation state of minus two (−2) and hence the following breakdown are extremely slow on a reasonable time frame that would refrain the accumulation in the environment.

Therefore, all the scientific and technological initiatives aimed at mimicking and hence speeding up the pro-oxidant outcome of the full-carbon backbone macromolecules, thus microorganisms can support the opening of the door to a chain breakdown to functional fragments, vulnerable to the microflora present in the different environmental compartments.

The oxo-biodegradation is then not a blasphemous attribute to be condemned like the “satanic verses,” and cannot even be set aside like a fictional word, because it is a reality to be boosted on behalf of environmental protection and freedom in competition for the production and commercialization of large consumption of plastic commodities and disposables.

The functional fragments attainable from full-carbon backbone polymeric materials as a consequence of the *tandem* action exerted by the pro-oxidant/pro-degradant additive (Fig. 14.7b), thanks to the presence of hydrophilic functional groups, become

Table 14.2 Bacteria and Yeasts capable of hydrocarbon oxidation

Bacteria	Yeast
Achromobacter	*Candida*
Acinetobacter	*Cryptococcus*
Actinomyces	*Debaryomyces*
Aeromonas	*Endomyces*
Alcaligenes	*Hansenula*
Arthrobacter	*Mycotorula*
Bacillus	*Pichia*
Beneckea	*Rhodotorula*
Brevibacterium	*Saccharomyces*
Corynebacterium	*Selenotila*
Flavobacterium	*Sporidiobolus*
Micromonospora	*Sporobolomyces*
Mycobacterium	*Torulopsis*
Nocardia	*Trichosporon*
Pseudomonas	
Spirillum	
Vibrio	

wettable, thus allowing for the attack by microbial consortia, ubiquitous in various environmental compartments, whose ultimate result is controlled safe digestion of the microbial fragments to water, carbon dioxide (and methane in semi-aerobic conditions), and cell biomass (Fig. 14.7a).

The described role of the pro-oxidant/pro-degradant additives allows us to generalize the concepts and the basic principles to support the propensity to biodegradation of polymeric materials and relevant plastic items bringing substantially all the matter to a class of materials and products identifiable as biodegradable under environmental conditions that have to be strictly defined.

In fact, the biodegradability is basically not an absolute attribute, because it is strictly connected to the environmental conditions and time frame in which the biodegradation process of either the material or the relevant products does occur.

For the sake of clarity and within the scope of substantiating and qualifying the biodegradable polymeric materials and relevant plastic items according to the key steps of the overall process, we can confine the materials and products into two classes, identifiable as "hydro-biodegradable" and "oxo-biodegradable" items, distinct for the first step of the overall process (Fig. 14.8). In both cases, the ultimate step is consisting of the microbial digestion to water, carbon dioxide, and cell biomass independent of the hydrolytic or oxidative pathways leading to the macromolecule carbon chain breakdowns.

The major difference between the two classes of polymeric materials is represented by the energetic profiles of the first step. In fact, as sketched in Fig. 14.9 the activation energy of the C–H bond that is the crucial point in the oxidative degradation step is higher than one order of magnitude with respect to the

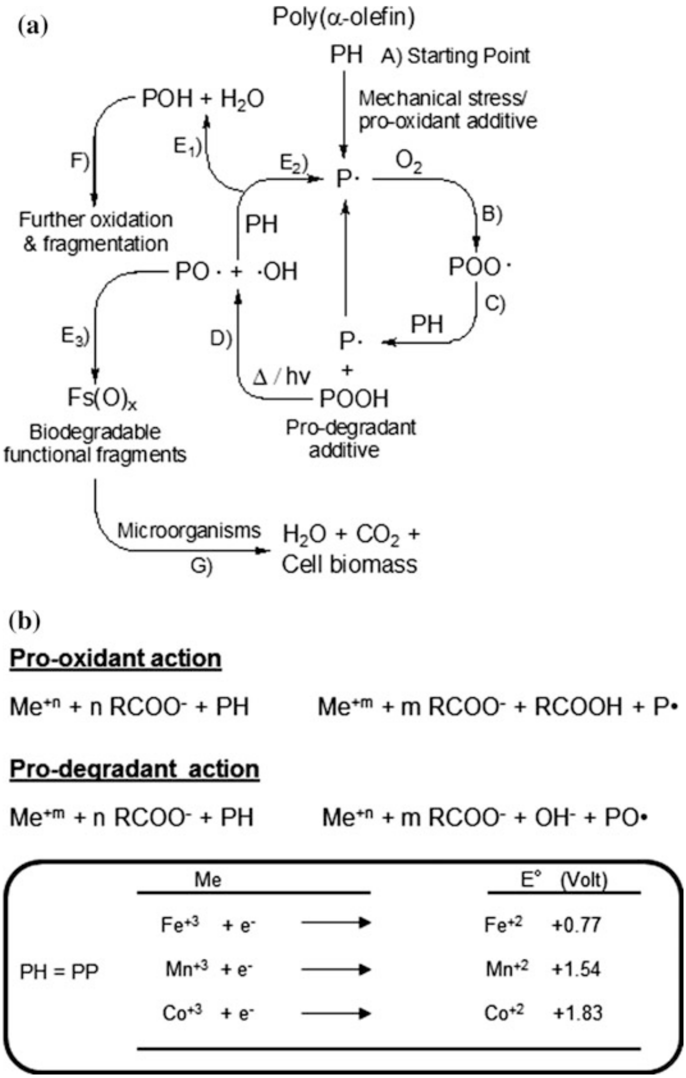

Fig. 14.7 Mechanism of oxidative degradation of full-carbon backbone polymeric materials. **a** General scheme including also the ultimate stage of biodegradation. **b** Specific transition metal salts able to promote oxidation followed by degradation of full-carbon backbone polymers in a tandem fashion action

hydrolytic step expected for the degradation of an heteropolymer such as polyesters. Therefore, in order to speed up the degradation rate of full-carbon backbone polymeric materials, a small amount of additive displaying catalytic activity in promoting the oxidation of C–H bond is needed.

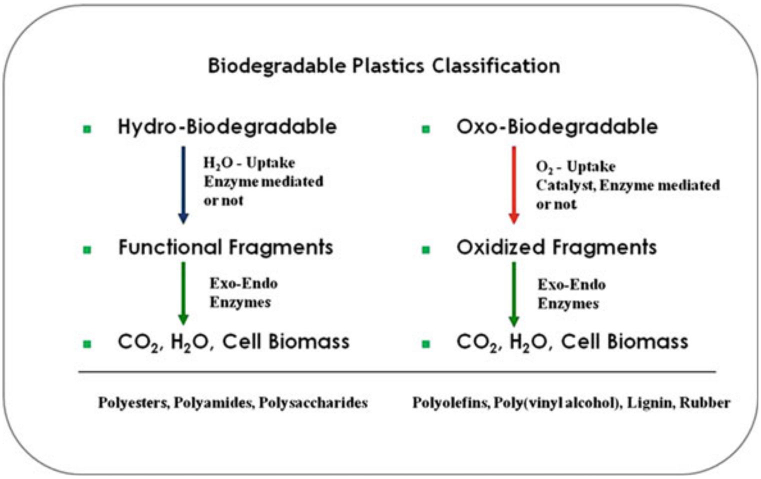

Fig. 14.8 Environmentally degradable polymers and plastics

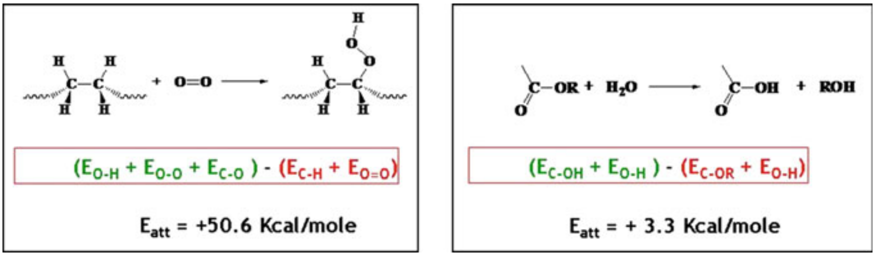

Fig. 14.9 Energetic profiles in oxo- and hydro-biodegradables' primary steps

In Fig. 14.10, a sketched representation of the effects caused by an oxo-biodegradable attack onto a low-density polyethylene (LDPE) macromolecular chain is shown. It includes at the bottom line the parameters that currently have to be monitored in order to gain undisputable experimental evidences on the various steps that start with the oxidation of the carbon backbone followed by the chain breakdown to functional fragments vulnerable by ubiquitous microorganisms in the different environmental compartments with ultimate conversion under aerobic conditions of water, carbon dioxide, and cell biomass [21–31].

It is worth mentioning that the positions taken by the unbelievers and disparagers of the oxo-biodegradable polymeric materials and plastics (OBPs) are often based on the following points:

(a) The toxicity of the pro-oxidant/pro-degradant additives due to the presence of heavy metals [absolutely false because the quoted additives are based on fatty acid salts of transition metals present as microelements in the soil and water (fresh and salty water) compartments].

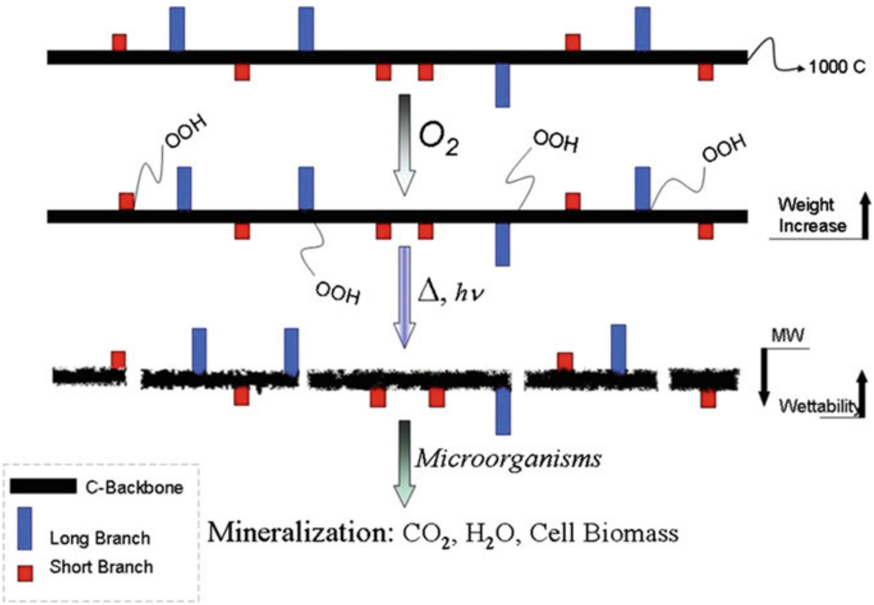

Fig. 14.10 Schematic representation of the oxo-biodegradation process of LDPE polymer chains

(b) The unsuitability of OBPs to be mechanically recycled as a second raw prime material (absolutely inconsistent, provided the recyclers do as usually a suited upgrading of the materials and manufactures submitted to a mechanical recycle).

(c) The accumulation of the oxidized fragments, derived from the first step of the process in the environment, with seriously deleterious effects on the flora and fauna present in the environmental compartments [a very debatable statement as we experimentally observed that the fragments derived from an inorganic (non-bionic step) attack can experience a further oxidative microbial attack (bionic step) in combination with the metabolic attack by the microorganisms with production of water, carbon dioxide, and biomass].

Non-comprehensive, but undebatable, evidences are reported in references [32–36].

14.1 Conclusions

OBPs based on a full-aliphatic-carbon backbone such as polyethylene (PE) and polypropylene (PP), when doped with relatively small amounts of pro-oxidant/pro-degradant additives (0.5–1 % by weight of fatty acid salts of transition metals such as Fe, Mn, or Co or their combination), become susceptible to an oxo-biodegradation process occurring at the end of their predetermined service life.

Table 14.3 List of more recent papers on oxo-biodegradable polymeric materials and relevant plastic items

M. U. de la Orden, J. M. Montes, J. M. Urreaga, A. Bento, M. R. Ribeiro, E. Pérez, M. L. Cerrada. "Thermo and photo-oxidation of functionalized metallocene high density polyethylene: Effect of hydrophilic groups", *Polym. Degad. Stab.*, 111, 78–88 (2015)
M. E. Boscaro, E. A. De Nadai Fernandes, M. A. Bacchi, S. M. Martins-Franchetti, L. G. Cofani dos Santos, S. S. N. S. Cofani dos Santos. "Neutron activation analysis for chemical characterization of Brazilian oxo-biodegradable plastics", *J. Radioanal. Nucl. Chem.,* 303, 421–426 (2015)
R. Vijayvargiya, A. K. S. Bhadoria, A. K. Nema. "Photo and biodegradation performance of polypropylene blended with photodegradable additive ferrocene (Part – I)", *Int. J. Appl. Sci., Eng. Res.,* 3, 153–170 (2014)
S. K. Samal, E.G. Fernandes, A. Corti, E. Chiellini. "Bio-based Polyethylene–Lignin Composites Containing a Pro-oxidant/Pro-degradant Additive: Preparation and Characterization", *J. Polym. Environ.,* 22, 58–68 (2014)
T. Muthukumar, A. Aravinthana, R. Dineshram, R. Venkatesan, M, Doble. "Biodegradation of Starch Blended High Density Polyethylene using Marine Bacteria Associated with Biofilm Formation and its Isolation Characterization", *Microb. & Biochem. Technol.,* 6, 116–122 (2014)
M. Takev, P. Velev, V. Samichkov. "Physicomechanical Properties of Biodegradable Composites, Based on Polypropylene and Paper From old Newspapers", *J. Chem. Technol. and Metall.,* 49, 363–369 (2014)
F. Masood, T. Yasin, A. Hameed. "Comparative oxo-biodegradation study of poly-3-hydroxybutyrate-co-3-hydroxyvalerate/polypropylene blend in controlled environments", *Int. Biodeterior. Biodegrad.* 87, 1–8 (2014)
S. T. Harini, S. Padmavathi, A. Satish, B. Raj. "Food compatibility and degradation properties of pro-oxidant-loaded LLDPE film", *J. Appl. Polym. Sci.*, 131, (2014)
M. M. Reddy, M. Misra, A. K. Mohanty. "Biodegradable Blends from Corn Gluten Meal and Poly (butylene adipate-co-terephthalate)(PBAT): Studies on the Influence of Plasticization and Destructurization on Rheology, Tensile Properties and Interfacial Interactions", *J. Polym. Environ.*, 22, 167–175 (2014)
L. S. Montagna, A. L. Catto, M. M. de Camargo Forte, E. Chiellini, A. Corti, A. Morelli, R. M. Campomanes Santana. "Comparative assessment of degradation in aqueous medium of polypropylene films doped with metal free (experimental) and transition metal containing (commercial) pro-oxidant/pro-degradant additives after exposure to controlled UV radiation". *Polym. Degrad. Stab.*, 120, 186–192 (2015)

That behavior is of utmost importance because it allows a consequent control of the accumulation onto the different environmental compartments of soft flexible plastic items identified as commodities eventually released in the environment as a consequence of impolite civil behavior of the end users.

In fact, depending upon the type and content of pro-oxidant/pro-degradant additives present in the doped plastic items and upon the environmental conditions recordable in the compartment where the postconsumer plastic items are either intentionally or accidentally abandoned, one can foresee what would be the reasonable time frame necessary for a safe, useful biodegradation, thus mitigating the accumulation of plastic waste in the environment.

Finally, regarding the future perspectives for the increasing production and consumption of oxo-biodegradable plastics based on full-aliphatic-carbon backbone polymeric materials, one can refer to the list of references (Table 14.3) relevant to the years 2014 and 2015 reporting on research activities ongoing in the field of products susceptible to oxidative degradation followed by production of functional fragments vulnerable to microflora present in various environmental compartments.

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