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REVIEW

Bio- and oxo-degradable plastics: Insights on facts and challenges

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Abstract

The global accumulation of single-use plastic bags made from nonbiodegradable plastics is the most concerning environmental issue nowadays. The utilization of biodegradable materials is a choice to reduce the environmental impact resulting from the use of plastic products. The utilization of renewable resources to produce fully biodegradable plastics is among the technologies used to overcome petroleum plastic's negative impact. On the other hand, the utilization of oxo-biodegradable plastics where prodegradant additives are incorporated in conventional plastics to promote their degradation under certain conditions has recently received much attention. This review discusses the types and challenges that face the implementation of biodegradable plastics technology that uses renewable resources. This review also covers the debate addressed in the literature about the biodegradability fate of oxo-biodegradable plastic in the air, compost, soil, landfill, and marine. A comparative study included the potential published literature in the last 10 years was performed. Based on the discussed evidence in this review, it can be concluded that all literature agrees that the addition of pro-oxidant/prodegradants can accelerate the degradation of oxo-plastics to small fragments. However, the complete biodegradation of oxo-plastics by microorganisms remains in doubt. On the other hand, biopolymers produced from natural resources seem to be the future direction for plastics manufacturing especially single-use plastic bags.

KEYWORDS
environmental pollution, oxo-biodegradable, poly-lactic acid, polyolefins, starch

1 | INTRODUCTION

Currently, the primary concern of municipal solid waste is plastic bags made from nonbiodegradable polymers, which raised many concerns about environmental pollution. Plastic bags are considered the primary litter source; it poses a significant challenge to control waste generation. Polyethylene is one of the most known petroleum plastic used in packaging.[1] As the biodegradation of polyethylene (PE) or the other petroleum-based plastics is very slow on coast or land, their disposal is complicated; this results in various toxic chemicals. Currently, attention was given to produce polymers and polymers' additives from renewable resources; this is also tended by the global environmental awareness.[2-7] Bioplastics undergo biological degradation by the action of microorganisms (algae, bacteria, and fungi). The main resulted degradation products are methane, carbon dioxide, and biomass.[8]

There are many natural resources where renewable-based polymers originate from. These include cellulose, starch, chitosan, and proteins from animal and plant origins. Because of their green features, such natural polymers are considered attractive alternatives for petroleum-based nonbiodegradable plastics. Cellulose and starch are potential and abundant raw materials for producing bioplastics. These natural bioplastics become increasingly common owing to their biocompatibility and biodegradability features, environmentally friendly, renewability, and low cost.[9,10]

One of the most potent challenges is that most biopolymers are not utilized alone for plastic bags manufacturing. Most of them are brittle with low elongation at break and hydrophilic. Thus, incorporating other materials such as plasticizers has resulted in accepted materials with improved mechanical properties. However, the high cost of natural-based plastics compared to petroleum-based is the major challenge along the path to its complete commercialization in the world.

Oxo-biodegradable plastics are composed of petroleum-based polymers such as PE incorporated with selected additives (metal salts) that give the final product the degradability feature. The materials used as pro-oxidants are typically transition metals like nickel, iron, manganese, and cobalt.[11] These materials' principal task is to break down the large polymer molecular weight chain into smaller fragments, so the microorganisms can process and convert it to CO_2 and biomass. There is no doubt that the incorporation of oxo-additives in the nonbiodegradable polymer chain produces new plastics that, if exposed to moisture and sunlight, fragments into fine pieces (abiotic process). The degradation (fragmentation) time is sometimes unpredicted as it depends on the climate factors such as temperature and intensity of solar radiation. After fragmentation, the plastic may become invisible (but still exist in the environment). These tiny fragments are also concerned to impact the environment. After the first step of fragmentation (abiotic), the second step of biodegradation (biotic) comes; in this step, the microorganism processes the degradation products.[12,13] Recently, after the EU parliament's ban decision, many concerns have been raised about whether oxo-degradable plastics are fully biodegradable as per the international standards EN14855, D5338, and EN13432. The global oxo-degradable additives manufacturers were and still do their best to prove to the world the positive environmental footprint and applicability of these materials. Such great effort from the big players is accompanied by extensive research supporting or opposing oxo-biodegradable plastics. Roy et al[14] and Fontanella et al[15] raised concerns about the environmental fate of the polymer residues after fragmentation. It was also introduced that oxo-degradable plastics are not suitable for composting media and are not recyclable. The lifetime of the recycled products is determined by the presence of pro-oxidants in the original oxo-products.[14] Investigating these and other concerns about oxo-biodegradable plastics has become a necessity before accepting oxo-plastics as eco-friendly materials.[16] In this review, we have discussed the potential published work about oxo-biodegradable plastics in the last 10 years. The confidence of finding/recommendations of each published work was then demonstrated based on the author H index, journal impact factor, and publisher to give a clear overview about the value of data presented. This work has also reviewed the emerging technologies along with the challenges and opportunities of bioplastic come from renewable resources.

2 | OXO BIO-DEGRADABLE PLASTICS

The consumption of polyolefins, the primary raw materials for plastics-based products, has increased because of their superior mechanical properties, low cost, durability, and lightweight. Such properties made polyolefins the material of choice in most applications. On the other hand, plastic disposal has become a problem everywhere, especially for single-use plastic bags. Conventional disposal ways involve landfill, incineration, recycling, and composting. However, these bags' degradation may take about 300 years to entirely degrade in soil.[17,18]

To manage such environmental issues, finding degradable polyolefin has become a significant target of research. The main purpose of designing biodegradable polyolefins is to keep the functionality of the plastic products as a commodity with improved disposal solutions for the environment. Recently, it was found that polyolefins can be fragmented into tiny pieces by incorporating pro-oxidants (most likely are transition metals) within the plastic formula. The resulted plastic combination is called oxo-biodegradable plastic.[17,18]

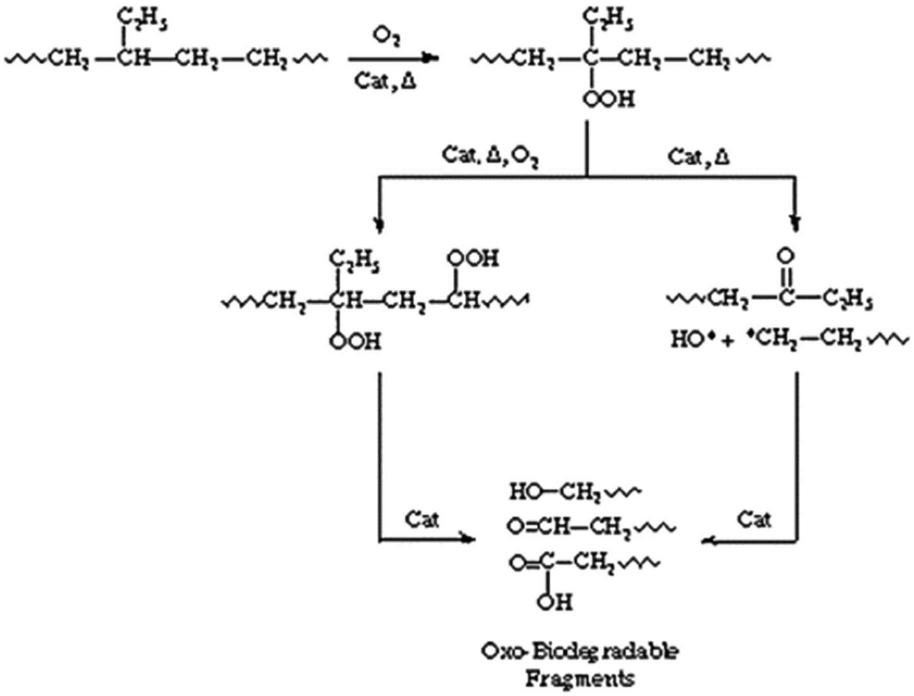
In oxo-biodegradable plastics, two degradation stage process occurs, namely abiotic and biotic. The abiotic process is composed of oxidative degradation that occurs by pro-oxidants' action.[17,18] The second step is the biotic process where microorganism converts the oxidation products resulted from the abiotic process into CO_2 and biomass. This degradation mechanism is called oxo-degradation. Pro-oxidant/prodegradants are accelerating the rate of abiotic oxidation by catalyzing chain breakdown by heat and/or light, producing free radicals, as shown in Scheme 1.[19] Consequently, oxidation products with low molecular weight are rapidly formed as shown in Figure 1A-C.[20,21]

2.1 | Common commercially oxo-biodegradable plastics manufacturers

For most polyolefins, transition metals represent the main percentage of commercially existing pro-oxidants. The leading common prodegradant producers are listed in Table 1.[17]

EPI Company claimed that the polyolefins containing their TDPA pro-oxidants are compostable and can be recyclable before degradation starts. The company stated that low percentages from 2 to 3 wt% of the TDPA additive to the traditional polyolefin would give degradation in the period between 20 and 36 months when disposed of in a suitable environment.[17] Renatura is a patented additive of iron stearate (prodegradant) and UV absorbers sold as a masterbatch for PE and PP. A loading percentage of about 2 wt% is enough for manufacturing degradable polyolefins. The resulting plastic blend can be processed in standard production processes (casting, extrusion, injection molding, and blow molding). The producer has also claimed that this oxo-biodegradable additive is a recyclable material, where the recyclability can be performed before the initiation of degradation. Reverte is supplied as masterbatches blended with PP and PE. The company claimed that it produced the

SCHEME 1 Oxidation mechanism of polyethylene[19]



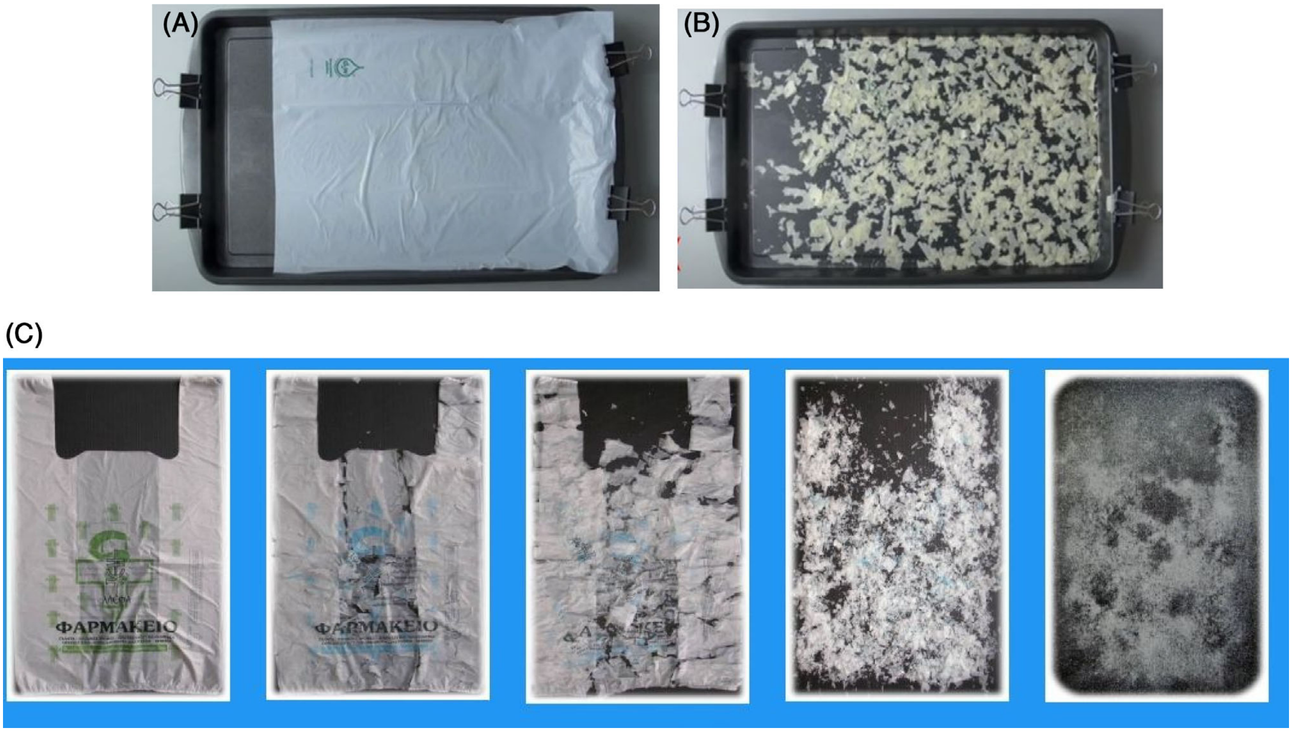


FIGURE 1 Low-molecular-weight oxidation products are rapidly formed: (A) oxo-degradable plastic bag at time zero, (B) oxo-degradable bag after 24 months.[20] The study of Revert single-use bag (C)[21]

first oxo-biodegradable additive for polyethylene terephthalate (PET) beverage bottles.[17]

One of the common patented additives that serves the single-use plastic bags production is AddiFlex. It is explained as oxo-thermal materials work in synergy with calcium carbonate ($CaCO_3$). The existence of $CaCO_3$ can support the increase of UV degradation by up to 66%; hence, less additive is required. d2W is another common prodegradant produced by Symphony Environmental. The company claimed that the additives are biodegradable but not suitable in composting medium as they are not suitable in anaerobic parameters.

TABLE 1 General information on transition metal-based prodegradants commercially available[17]

Trade name	Manufacturer	Active compounds	Loading (wt%)	Degrading conditions		Polymer types
				No light	anaerobic	
TDPA	EPI	Metal stearates (Fe, Ce, Co) and citric acid (typically co)	2–3	Yes	Yes	PP, PE, PS
Renatura	Nor-X industries	Iron stearate and combination of stabilizers/antioxidants	2	Yes	Possibly no	PP, PE
Reverte	Wells plastic limited	Undisclosed photo-inhibiting package, metal ion prodegradant package, and biodegradation promotors (micronized cellulose)	1–5	Yes	Possibly no	PP, PE, PS, PET, ABS
Addiflex	Add-X-biotech	Metal carboxylate (Fe, Mn, Cu, Co, Ni), starch, $CaCO_3$: manganese stearate has been identified for Addiflex HE	10–20 (Addiflex A) 3–6 (Addiflex HE) 1.5–6 (Addiflex HES)	Yes	Possibly no	PP, PE, PS
d2W	Symphony environmental	Metal stearates and stabilizers (typically Mn)	1–3	No	No	PP, PE

Abbreviations: ABS, acrylonitrile butadiene styrene; PE, polyethylene; PET, polyethylene terephthalate; PP, poly propylene; PS, polystyrene.

Only a percentage of 1–3 wt% is enough to produce degradable polymers. The lifetimes of biodegradation differ according to the type of metal salts and stabilizers the material contains.[17]

Before the European Parliament ban on Oxo-degradable Plastics last 2019, some consumables utilizing d2W were from Pizza Hut, Nescafe, KFC, Tiger Brands, Tesco Barclay, and Walmart. However, after this ban, the situation became different where there are few countries still use oxo-biodegradable plastics such as the United States of Emirates, Saudi Arabia, Bahrain, and Jordan. The big manufacturers of oxo-biodegradable still support their products and claim that they are friendly to the environment, and they reach full biodegradation after reaching the environment. Recently, the d2w oxo-plastics have been approved by the Saudi Arabian Standards Organization, SASO, and Emirates Authority Standardization Metrology, ESMA. Since April 2017, Saudi Arabia uses oxo-plastics in all single-use plastic bags used in shopping, trash, and clothing. The county planned to expand utilizing the oxo-plastics in phase 2 and phase 3 in April 2020. Recently, it postponed applying the phases may be due to the effect COVID-19 pandemic on the country. Phase 2 and 3 will include bubble wrap and cushioning packaging, postal use carrier bags, mail order bags, disposal tableware such as plates, spoons, and cups, and even plastic bags used with food (bakery items). The big manufacturers also see that the European Parliament ban on Oxo-degradable plastics is a political move that ignores the established science.[22]

The report from the commission to the European Parliament and the council on the impact of the use of oxo-degradable plastic, including oxo-degradable plastic carrier bags, on the environment stated the following aspects:

- Lack of evidence that oxo-degradable plastics are fully biodegradable in a reasonable time.
- Oxo-degradable plastics are not appropriate for recycling, long-term use, or composting.
- There is a particular risk that small plastics pieces will not completely biodegrade. Subsequently, microplastics can be accumulated in the marine environment.

2.2 | Oxo-plastics “are they degradable or biodegradable?”

In recent studies, there were many claims about oxo-plastics biodegradation. All agree about the fact of polymer fragmentation into small parts. However, some claims have raised the fact that oxo-plastics are not biodegradable. Because the fragmentation step does not produce enough small plastic molecular weight that the microorganisms can process and convert to CO2 and biomass.[12,16] There is a clear debate in the literature that addresses the biodegradation of oxo-plastics. Oxo-plastics biodegradation by the action microorganisms is well-reported, and potential scientific papers emphasized that after fragmentation, the fragments (small pieces of plastics) can be consumed by microorganisms.[23-28] However, many other publications have figured out that the biodegradation level can differ a lot under different experimental parameters[29,30] reflecting that environmental parameters are also different. Various biodegradation rates are discussed in the literature, that is, between 5 and 60%.[31-33] Such difference can be explained by the variation of the protocols utilized to determine the degradation, the experimental designs, and use of poorly controlled biotic and abiotic parameters.[24]

Many papers address the biodegradability of polyolefins involving prodegradants in the presence of selected microbial species in a controlled medium under controlled laboratory parameters and microorganism exists in the environment. Now, it seems to be accepted that high-molecular-weight polyolefin abiotic degradation into low-molecular-weight materials is necessary to make the action of enzymes and/or microorganisms possible.[24] The most used assessment methods to determine the biodegradation of polymer involve carbonyl index, loss of molecular weight, SEM analysis of film surface, elongation, tensile strength, and so on. Also, it has been reported that microorganisms can adhere to the polymer's surface to grow on it and synthesize biosurfactants that are probably important to degrade the oxidation products.[24]

2.2.1 | Selected Studies support the biodegradation of oxo-plastics

Many researchers studied the biodegradation of oxo-plastics.[31,34,35] Jakubowicz et al[35] tested the abiotic and biotic degradation of 15-mm thick low density poly ethylene (LDPE) film containing a prodegradant system. After elapsing 2 years, results revealed that high biodegradability levels of 91% in the soil environment were achieved, compared with only 43% in the compost environment at 58°C. This outstanding finding reveals the possibility of making LDPE that will achieve complete biodegradation in soil within 2 years. It also reports that the plastic fragments risk that remains in soil is shallow. Another important conclusion for this research is that the rate compost environment seems not suitable for oxo-plastics biodegradation. This is, maybe, due to the different mineralization, temperature, fungal, and bacterial strain in the compost environment compared to that of soil. In the same context, Chiellini et al[31] obtained a biodegradability of about 48% in soil compared with about 26% in the compost after 14 months. This achievement comes into agreement with data obtained by Jakubowicz et al.[35]

Gomesa et al[34] determined the effect of prodegradant additive and accelerated aging on the polyethylene film degradation in simulated soil, in line with ASTM G160-03. The films were presented for 30, 60, and 90 days in simulated soil under moisture and pH control. The results showed an increase in carbonyl index in samples with pro-oxidants and after 30 days of exposure. A decrease, after 60 and 90 days, indicates the uptake of material oxidation by-products by microorganisms.

Contat-Rodrigo[18] reported the abiotic and biotic degradation of PP samples with prodegradant materials. Abiotic degradation was carried out by exposing the samples of oxo-PP to UV radiation. He reported that significant changes in the thermal and morphological properties were noted. This result proves a higher level of oxidation in these samples. On the other hand, in the pristine control samples, changes in the thermal properties and morphology were detected in previously photo-oxidized PP samples with pro-oxidants when subjected to a subsequent soil burial test. Such findings suggest that previous abiotic degradation plays a vital role in soil microorganisms to do biodegradation. It is worth to mention that samples containing prodegradants but not previously oxidized showed almost no biodegradation when aged in soil. The previous results suggested in this work confirm the potential of utilizing pro-oxidants in producing biodegradable plastics through the combined effect of abiotic and biotic processes.[18] Ammala et al[17] have evaluated the photocatalytic oxidation performance of Reverte additives. The experiments were carried out in-house in the absence of light but in the presence of heat and UV radiation. A prompt rate of degradation (determined by carbonyl index) was observed under UV light for LDPE films. The molecular weight has arrived at a value of less than 10,000 Da. When samples were incubated for 5 weeks with fungi, they showed worth noting biodegradation. Figure 2A shows the virgin LDPE film that did not show any biodegradation signs. After incubation for 5 weeks, fungal growth was observed throughout the film of LDPE as shown in Figure 2B,C. SEM analysis also showed that a surface biofilm was observed, owing to the fungal growth and cracks within the LDPE film as shown in Figure 2D. Further approval of LDPE biodegradation was examined in the compost test (lab scale). The results suggested that 60% of biodegradation was obtained at 50°C after 400 days. Va'zquez-Morillas et al,[33] examined the biodegradation of different plastics (natural and petroleum) by conducting a controlled composting test in the laboratory for 180 days. The biodegradation percentages differed between the materials where the following percentages were obtained: 41% of PLA; 32.24% of printed oxo-degradable polyethylene, 25.84% of oxo-degradable polyethylene; 18.23% of printed polyethylene; and 13.48% of polyethylene, a sample of cellulose was utilized as a control, and it was mineralized in 58.45%. The authors reported that under appropriate waste management conditions, oxo-based plastics could be utilized as an alternative to reduce the impact of plastic films on the environment.

Corti et al[36] reported that an accelerated oxidative degradation of LDPE-containing prodegradant can be achieved within outdoor exposure. It was found that the oxidized fraction (fragments) started to disappear, which indicated that the biotic biodegradation process is working. This research concludes that oxo-degradable plastics are capable of performing both degradation and biodegradation. Khajehpour-Tadavani et al[37] reported that presence of isotactic polypropylene in HDPE matrix containing prodegradants additives could enhance the oxo-biodegradability of HDPE.

New enhanced nanoparticles as prodegradants to accelerate biodegradation of LDPE was suggested by Zhang et al.[38] The suggested martial is polymethyl methacrylate (PMMA)-grafted TiO_2. The results indicated a molecular weight reduction of 96.95% and a loss of weight of 39.6%. The authors' conclusion reported that PMMA grafting could improve the LDPE biodegradation. Mittal and Patwary[39] have developed biodegradable nanocomposites of polypropylene by blending prodegradant and other fillers such as graphene silica. The results suggested controlled biodegradation. Konduri et al[40] used (manganese, titanium, iron, cobalt) stearate as prodegradant to treat LDPE, followed by irradiation with UV and incubation with *A. oryzae*. The results suggested a loss in tensile strength by 51, 45, 40, and 39% for manganese stearate, titanium stearate, iron stearate, and cobalt stearate.

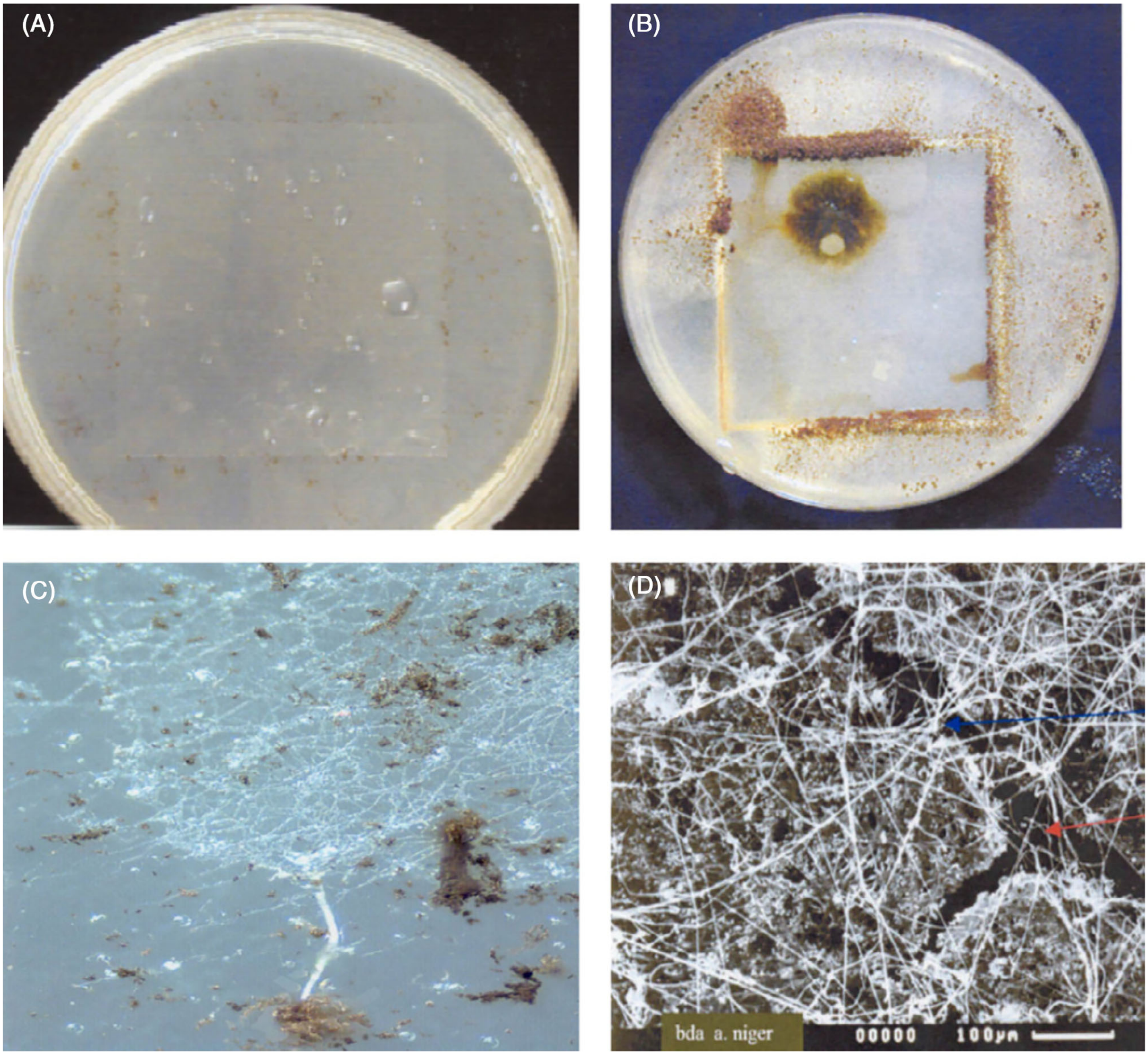


FIGURE 2 Polyolefins microbial biodegradation of LDPE containing Reverte additives. (A) virgin LDPE, (B) after 5 weeks of incubation, (C) evidence of fungal growth, and (D) significant fungal growth and cracks within the film were observed[16]

Montagna et al[41] has done a biodegradation test for PP samples at 58°C and 120 days. The results suggested that the modified PP exhibited an improved degradation through increased generation of CO_2 that was evidenced by weight loss during incubation. The thermal analysis also showed a decrease in the melt temperature and an increase in the degree of crystallinity. SEM analysis showed surface deterioration and holes appearance. Sable et al[42] suggested that the biodegradability of oxo-plastics is lower than that of PLA. However, it significantly is much higher than pure PP. The results indicated that incorporating pro-oxidants together with PLA throughout PP could accelerate the biodegradability. The results also revealed that cobalt stearate showed superior biodegradation improvement compared to calcium stearate.

Very few results addressed the biodegradation of oxo-plastics in the marine environment.[43] One of the few studies was conducted by Abed et al[43] in Arabian Gulf. The authors examined the degradation of PE, PET, and oxo-based PE in the marine environment at different depths of 2 and 6 m. SEM showed the remarkable formation of a fissure on oxo-based PE that indicated physical degradation. Regarding chemical degradation, carbonyl bonds, and hydroxyl groups were detected on oxo-based plastics by FTIR. Bacteriodetes, Proteobacteria, and Planctomycetes were found on all types of plastics. But, sequences of Zoogloea and Alteromonas were oxo-based PE specifically, indicating a possible involvement of such bacterial genera in the degradation of oxo-based PE. As a conclusion for this research, it was suggested that oxo-degradable plastic showed many degradation signs with time because of a combination of abiotic and biotic processes. Also, it should be mentioned that the degradation was less in the planktonic than in the benthic zone. Figure 3 shows that all plastic types of PE, PET, and oxo-based PE developed communities of

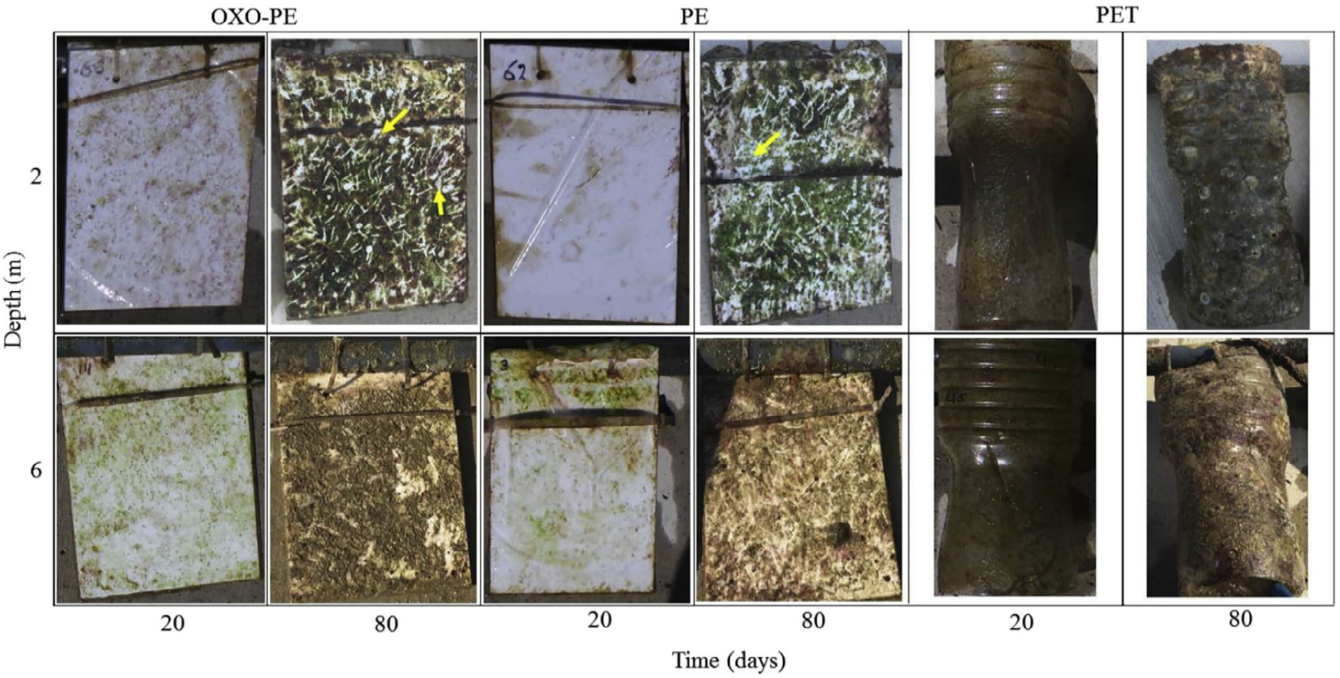


FIGURE 3 Communities of biofouling development on films of oxo-based PE, PE, and PET bottles after 20 and 80 days, submerged in seawater at a depth of 2 and 6 m.[43] PE, polyethylene; PET, polyethylene terephthalate

biofouling at both depths and at a time ranging from 20 to 80 days; however, red algae were observed more on oxo-based PE.

2.2.2 | Selected studies oppose the biodegradation of oxo-plastics

Al-Salem et al[44] investigated companies' claims that supply oxo-biodegradable plastic bags under the so-called "environmentally friendly." The commercial products are composed of polyethylene with prodegradant compounds. Aging tests performed revealed that UV radiation controlled the mechanism of biodegradation. After exposure to the weathering test, 50% of weight loss was reached, which is evidence of plastic film fragmentation. Also, in soil burial, a weight loss of some 83% was determined after 12 months. Based on the results of both thermal stability and mechanical properties, it can be said that weathering was more severe than soil burial. The authors reported that considering such commercial products as eco-friendly is in doubt as none of such showed significant biodegradation evidence even after being evaluated in harsh conditions.

Musioł et al[12] have determined the degradability in the biotic and abiotic medium of oxo bio-degradable polyethylene commercial bags taken from the Polish market. Samples were exposed to hydrolytic conditions and industrial composting for up to 364 days with modest structure change. The average molecular weight reduction was 47%. The results suggested that part of the PE in the environment may crosslink and remain in the environment after degradation. The main finding of this work is that products labeled as oxo-biodegradable that are present in the market fragmented and degraded very slowly in the compost medium in the tested period of 1 year. As a conclusion, the authors reported that these bags should not be labeled as environmentally friendly and should be further tested to avoid the persistent artificial materials spread in nature. Portillo et al,[16] reported that oxo-degradant can promote the polyethylene samples degradation using UV irradiation. This degradation produces reduced polymer molecular weight, especially at the early stages of the biotic degradation. The conclusion of their research revealed that despite oxo pro-oxidants additives promotes the photodegradation of PE, this degradation is not enough, neither in the more severe condition of irradiation (CI 0.59 for PE and 5.6, for PEþAD), to produce a decrease in the molecular weight that enables composting.

Kumar Sen and Raut[30] have suggested that oxo-biodegradable plastics do not conform to the compostability requirements set out in various established standards. (Home and/or industrial). The authors suggested that oxo-degradable plastic bags are not biodegradable but are designed to break down into small ferments after exposure to oxygen. Such smaller plastic pieces represent a severe problem if animals, fish, consume them or if they are scattered over the ground. Finzi-Quintao et al[45] were among the few researchers who studied the biodegradability of oxo-based plastics in the landfill. It was suggested that despite utilizing oxo-biodegradable compounds known in the literature, the composition of the bags did not permit for biodegradation in Brazilian landfill because landfills have an acid environment with no light or oxygen to permit the process of biodegradation for the studied material.

Yashchuk et al[46] evaluated the d2w prodegradant additives found in the local market along with traditional PE bags. The samples were thermally aged with UV light. Then to determine the biodegradability, the samples were put in a bioreactor in a controlled compost environment. It was observed that in the first 30 days of the biodegradation process, a higher CO_2 production rate was detected. After 90 days of incubation, 24% of oxo-based plastics were biodegraded which is not enough to yield mineralization of polyethylene in a complete form.

Briassoulis et al[47] evaluated the biodegradation of LDPE/prodegradant in the case of buried plastic bags in the soil for 8.5 years. Up to the knowledge of the authors of this review, this is the longest biodegradation time that a published work investigates. It was observed that the degradation was abiotic and proceeded slowly. Then, with time, the degradation was increased, and it was found that the samples were turned into infinitesimally very small and invisible micro-fragments (diameter of 1 mm). The authors suggested that the presence of such residues on the environment and the soil can be judged; however, it is also not possible to deny the degradation of these bags by adding oxidants and exposure of LDPE films to UV irritation. Figure 4 shows the progress of degradation for preaccelerated samples using oven against UV after different time intervals. The clear microfragments that exist in the photos indicate clearly that oxo-materials are not degradable even after 8.5 years of the real test. Such very tiny microfragments reveal a major risk on the respiratory system of animals or humans.

2.2.3 | What could be concluded from this debate?

As per the last studies, all agree that the addition of pro-oxidant/prodegradants can accelerate the degradation of oxo-plastics to small fragments “abiotic degradation.” However, the debate remains in the following question “Does the molecular weight of these small fragments is enough for the microorganism to do a complete biodegradation?” Answering this question is challenging because the efficiency of the degradation process depends on many factors such as temperature, intensity, duration of sunshine, the geographic place where the plastic bag is, marine degradation compared to outdoor (air) degradation, and type and dose of pro-oxidant/prodegradants. Furthermore, to the moment, it is not obvious how much polymer fragmentation is needed to let microorganism biodegrade the fragments completely. Haines and Alexander[48] reported that only alkanes with molecular weights of <620 Da can be biodegraded. Restreopo-Folrez et al[49] suggested that the molecular weight should not exceed 50 carbons.

Yoon et al[50] pointed out that PE with a molecular weight of less than 1700 in presence of *Pseudomonas* sp. *E4* underwent biodegradation arriving about 30% after 80 days. Generally, the molecular weight should be much smaller than the virgin plastics. This means that a drastic decrease in the molecular weight of polyolefins must happen as a condition for later biotic mineralization.[51-54] It is worth mentioning that the fragmented material biodegradation fate is a great concern because they may undergo other structure changes like crosslinking, resulting in a lack of biodegradation by the microorganism. Thomas et al,[13] therefore, complete biodegradation of plastic bags in the open atmosphere, soil, and composting environment remain in doubt. However, it can be concluded that the presence of pro-oxidant/prodegradants accelerates the plastic fragmentation

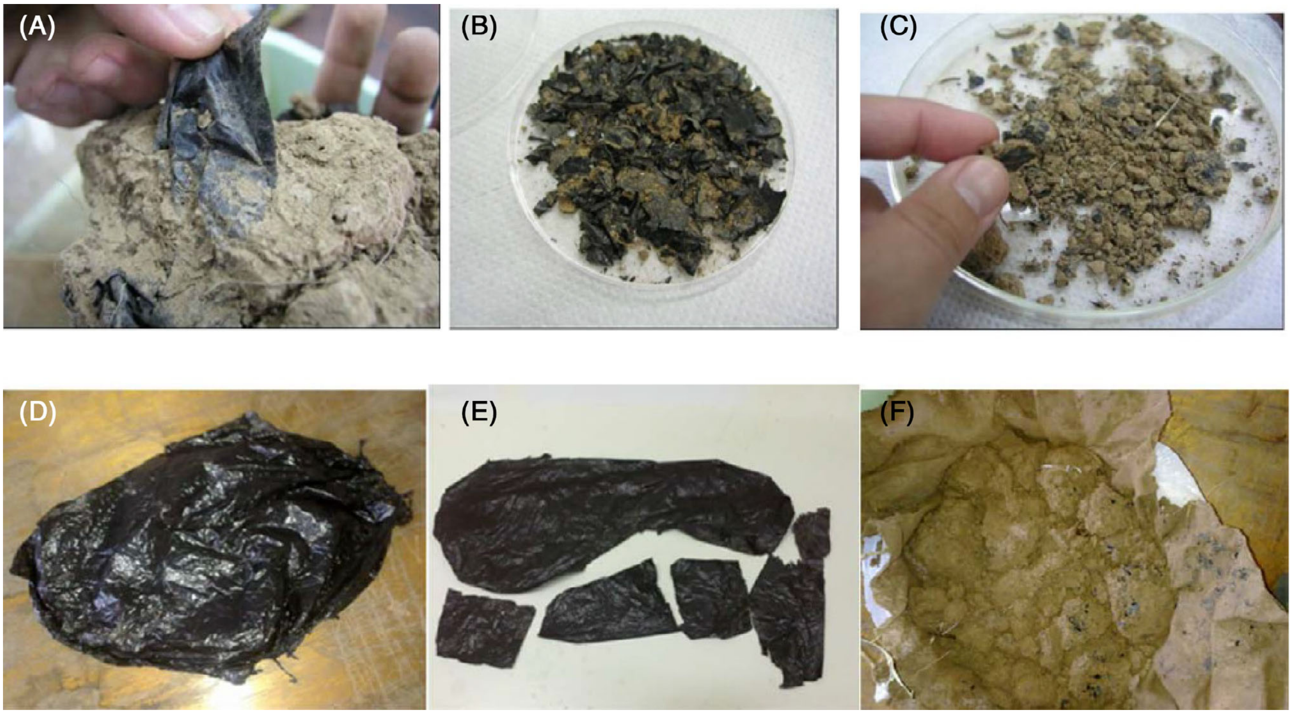


FIGURE 4 (A) Intact part extracting of the oven-treated “oxo-LDPE” sample after 31 months of soil burial; (B) UV-treated pieces of LDPE film collected in a petri dish (31 months); (C) solid aggregates of soil and fragments (31 months); (D) untreated sample oxo-LDPE inside the pouch and after washing and drying; (E) oven-treated sample of oxo-LDPE; and (F) UV-treated sample of oxo-LDPE, after 69 months in conditions of soil burial[47]

TABLE 2 A comparative study between published work that supports and opposes oxo-degradable plastics

#	Corresponding author/first author			Journal	Publisher	Impact factor (2020)	Main conclusion of paper	References
	Name	Country	Scopus H index (2020)					
Selected papers support oxo-plastics								
1	Emo Chiellini	Italy and Canada	51	*Polymer Degradation and Stability*	Elsevier	3.78	Prodegradants used in the formulation of LDPE film samples were effective.	36
2	Anne Ammala	Australia and Malaysia	10	*Progress in Polymer Science*	Elsevier	24.5	Oxo-plastics undergo changes in chemical structure as a result of oxidation in air, thus causing the breakdown of the molecules into small fragments that are then bioassimilated.	18
3	Contat-Rodrigo	Spain	10	*Polymer Degradation and Stability*	Elsevier	3.78	Pro-oxidant/pro-degradant addition promotes the whole degradation of the PP samples, confirming the potential of this additive in producing environmentally degradable polypropylene via combination of abiotic and biotic oxidizing agents.	19
4	Lucas Bonan Gomes	Brazil	4	*Materials Research*	Universidade Federal de Sao Carlos	0.84	The use of polyethylene and pro-oxidants could lead to manufacture biodegradable films in soil environments.	49
5	Ignacy Jakubowicz	Sweden	12	*Polymer Degradation and Stability*	Elsevier	3.78	After 2 years in the soil mineralization experiment, 91% biodegradability was achieved without reaching a plateau phase.	40
6	Maria Catarina Megumi Kasuya	Brazil	24	*PLoS ONE*	Public Library of Science	2.77	P. ostreatus is capable of degrading oxo-biodegradable plastic and producing mushrooms using the plastic waste without any prior physical treatment.	56
7	Emo Chiellini	Italy	51	*Journal of Polymers and the Environment*	Springer	2.77	The obtained results have significance in achieving effective and sustained biodegradation of LLDPE.	41
8	Vikas Mittal	UAE	27	*Polymer Engineering & Science*	Wiley	1.92	The pro-oxidant was successful in attaining controlled degradation.	44
9	Gholam-Reza Nejabat	Iran	8	*Journal of Applied Polymer Science*	Wiley	2.18	The presence of isotactic polypropylene in HDPE matrix containing oxo-compound can improve HDPE oxo-biodegradablity.	59
10	Mortaza Gholizadeh	Iran	13	*Asia-Pacific Journal of Chemical Engineer*	Wiley	1.520	Degradation of PE was increased after adding starch, oxo-material, PLA mixture.	58
11	Chaoqun Zhang	China	6	*Polymer Composites*	Wiley	2.010	Adding poly (methyl methacrylate) grafted TiO_2 nanoparticles enhanced degradability of LDPE.	43
12	Mohan K. R. Konduri		8	*Applied Polymer Science*	Wiley	2.18	Pro-degradant additive (PSH) to LDPE enhanced the degradation of LDPE.	45
13	Briassoulis, Demetrios	Greece	29	*Applied Polymer Science*	Wiley	2.18	By using oxidation followed by exposure to ultraviolet radiation and burial of low-density polyethylene, LDPE can degrade into fine fragments.	52

(Continues)

TABLE 2 (Continued)

	Corresponding author/first author							
#	Name	Country	Scopus H index (2020)	Journal	Publisher	Impact factor (2020)	Main conclusion of paper	References
14	Ruth Santana	Brazil	16	*Applied Polymer Science*	Wiley	2.18	Adding organic additive can improve degradation of PP.	46
15	Raquez, Jean Marie	Belgium	41	*Applied Polymer Science*	Wiley	2.18	Mixing oxo-LDPE with TPPS can improve degradation of LDPE.	57
16	Sanjeev Ahuja	India	4	*Journal of Environmental Management*	Elsevier	4.865	The biodegradability results of films with/without pro-oxidants show that their biodegradability is significantly lower than cellulose but significantly higher than pure PP. The biodegradability studies demonstrate that filling pro-oxidants to the PP and PP/PLA blends accelerate their decomposition.	47
17	B. Eyheraguibel	France	8	*Chemosphere*	Elsevier	5.108	Oxo-polymers degadation in nature is strongly dependent on environmental conditions. In nature, the biodegradability of oligomers could result from processes occurring both at the molecular (oxidation) and the macromolecular (diffusion and release) levels.	25
18	Raeid M.M. Abed	Sultanate of Oman	30	*Marine Pollution Bulletin*	Elsevier	3.782	We conclude that OXO-PE shows increased signs of degradation with time owing to the combination of abiotic and biotic processes, and its degradation is higher in the benthic than in the planktonic zone.	48
	Total		351			72.42		

	Corresponding author/first author							
#	Name	Country	H index	Journal	Publisher	Impact factor	Main conclusion of paper	References
Selected Papers oppose oxo-plastics								
1	Sultan Al-Salem	Kuwait	33	*Journal of Environmental Management*	Elsevier	4.87	The claims by the manufacturing companies which provided the original specimens under an environmentally friendly pretense is disputed due to the fact that none of the products actually showed evidence of major fragmentation or deterioration after exposure to harsh environments.	49
2	Marta Musioł	UK and Poland	11	*Waste Management*	Elsevier	5.43	The work suggests that these materials should not be labeled as biodegradable and should be further analyzed in order to avoid the spread of persistent artificial materials in nature.	13
3	Elida Beatriz Hermida	Argentina	12	*Polymer Testing*	Elsevier	2.94	The molecular weight reduction in compost was not enough to reach the maximum biotic degradation level established by international standards for biodegradable materials	17

TABLE 2 (Continued)

#	Corresponding author/first author Name	Country	H index	Journal	Publisher	Impact factor	Main conclusion of paper	References
4	Sangeeta Raut	India	7	*Journal of Environmental Chemical Engineering*	Elsevier	CiteScore: 4.09	The oxo-degradable plastic bags are not biodegradable but are designed to break down into small pieces after exposure to oxygen. The smallerpieces may lead to environmental problems if they are consumedby animals or if the small pieces are scattered over the ground	[35]
5	Cristiane M. Finzi-Quintao	Brazil	3	*Macromolecular Symposia*	Wiley	0.68	Landfills have an acid environment with no oxygen or light to allow the biodegradation process of oxo-plastics.	[50]
6	O. Yashchuk	Argentina	7	*Procedia Materials Science*	Elsevier	-	The additive promoted degradation by abiotic factors and increased the microbial activity in the early stages of the biodegradation. However, the additive is not sufficient to produce a complete mineralization of polyethylene.	[51]
	Total		73			18		

without a doubt. Also, in most reported papers, biodegradation process (in air/soil) happens slowly and can achieve high biodegradation parentage. It should be also concluded that oxo-plastics are poorly biodegraded in a compost environment.

A comparison of the potential of the published works can be shown in Table 2. We herein introduce a new debate analysis method by presenting the potential published works for both debated parties (Only Elsevier and Wiley) in the last 10 days. Both papers that support and/or oppose the oxo-plastics are reported. As the survey done by the authors of this review, it could be seen that the number of published papers that supported oxo-plastics was higher that oppose with a total h index of authors of 332 and 102, respectively, as shown in Table 2. After 2017 and especially after banning oxo-plastics in Europe, the number of published papers that oppose oxo-plastics increased. It is also expected that the opposing status becomes the trend in the future. Also, the big players of oxo-plastics manufacturers likely continue to prove their products' biodegradability nature scientifically. It should be mentioned that before banning the oxo-plastics in Europe, remarkable consumers were utilizing d2W oxo-plastics such as Nescafe, Tiger Brands, Barclay, Pizza Hut, KFC, Tesco, Unilever, Pepsi, and Walmart. After banning the oxo-plastics in Europe, most of these common consumers stopped using oxo-plastics in their products in all countries all over the world even if those countries have not banned oxo-plastics; the authors think that there is a reason beyond the environmental pollution one. This behavior could be due to the logistics challenges at these consumers' side. In other words, it could be flexible at the multinational companies' side if adjacent countries use the same type of plastics instead of managing to supply different kinds of plastics for other countries.

3 | BIO-BASED PLASTICS

The word biopolymer includes naturally occurring polymers but also high-molecular-mass molecules polymerized by chemical and biological methods. The biopolymers are not always biodegradable. Biodegradable plastics are primarily produced through aerobic synthesis from renewable raw materials and through composting and anaerobic digestion from waste management. Biopolymers are categorized according to their structure or biodegradability. They are derived directly from biomass (polysaccharides and proteins).[8]

3.1 | Starch

Increasing awareness in biopolymers and renewable feedstock has brought about the development of numerous options to traditional plastics. The oil crisis in 1970s prompted the investigation for alternative compounds to substitute fossil plastics.[55] Starch is one of the most interesting materials; it is the maximum renewable and abundant plant polysaccharides.[56] It is a biodegradable material, and it can be produced in huge masses at a relatively low fee, treated without problems, and produces film products with low oxygen permeability. But, native starches show negative mechanical properties, thermal stability, and brittleness. Those limit its diverse applications in its local form and its use for the manufacturing of food packaging and plastic bags.[57] To improve bioplastics' functionality, numerous plasticizers (sorbitol, glycol, glycerol,) are utilized to transform the starch into thermoplastic starch (TPS) by subjecting shear and heat over extrusion techniques.[58,59]

3.2 | Polylactic acid

Ployactic acid (PLA) is a polyester (synthetic biopolymer) that can be derived from starch materials. PLA originates from PLA, so its backbone is hydrolyzable and is ready for biodegradation.[60] The mechanical properties of PLA are much similar to many petroleum-based plastics such as PET and polystyrene (PS); that is why PLA has received much attention.[61,62] It also has many other inserting advantages such as biocompatibility, no toxicity, environmentally friendly, ease of fabrication, consumes less fossil energy, compostable, and high thermal plasticity.[63,64] Because of PLA is already synthesized from renewable and natural resources, so the expansion in its use could reduce the consumption of fossil energy and subsequently reduce the greenhouse gas emissions.[64] The Food and Drug Administration (FDA) declared that PLA to be employed for materials production that come into contact with food. Production of lactic acid is carried out by the following process steps: carbohydrates fermentation, protein and cell mass removal, recovery and purification, concentration, and removal of color (Henton et al., 2005).[65]

Despite that PLA disposal through composting, combustion, and so on, generates $CO_{2,}$ which is a greenhouse gas. It can be said that the impact of this generation is much lower than that of generated from petroleum polymer disposal. Unlike petroleum plastic disposal, the CO2 generated from PLA disposal is balanced during the feedstock plant growth by an equal amount (Henton et al., 2005). Many industrial and academic initiatives concentrated on enhancing PLA efficiency, especially in the single-use plastic bags sector. Efforts included blending PLA with other materials such as copolymerization with biopolymers.[66]

Industrially, PLA modification by melt blending is interesting as it is a simple process, economic, and readily available technology at the industrial level.[67] Generally, the blend component compatibility affects the final material physical properties such as melting point, glass transition temperature, morphology, and crystallinity. Such properties identify the final material performance, such as rigidity, processability, degradation behavior, barrier properties, tensile, and impact strength.[68]

3.3 | Polycaprolactone

Polycaprolactone (PCL) is biodegradable thermoplastic polyester with superior heat processability, low viscosity, and low melting point. PCL is produced by ε-caprolactone polymerization. Since PCL has a low melting point, poor mechanical, and barrier properties, PCL application alone (without blending) as a biodegradable polymer in the packaging industry is rare due to the weak barrier properties and the weak mechanical properties of the PCL. Usually, PCL is combined with other materials such as cellulose acetate sulfate, polyacetic acid, and cellulose propionate. These additives enhance the stress crack resistance, adhesion, and dyeability of plastic.[69]

3.4 | Polyhydroxyalkanoates

Polyhydroxyalkanoates (PHAs) are R-hydroxyalkanoic acids, which are thermoplastic polyesters. PHAs are produced and deposited in gram-positive and gram-negative bacteria as intracellular carbon and energy reserves under the nutrient limitation of oxygen, phosphorus, nitrogen, or after shifts of pH.[70] PHAs are stored in cytoplasmic granules with a diameter usually of 0.2–0.5 m. Visualization of these granules can be performed using staining dyes such as Sudan Black Band Nile red or by phase contrast microscopy due to their high refractivity. If the cell is supplied with the limiting nutrient, such energy-stored compounds are decomposed and utilized as a carbon source for bacterial growth.[71]

Polyhydroxybutyrate (PHB) is a popular member of PHA with a high crystallinity degree. Maurice Lemoige discovered PHB in 1926 in the *Bacillus megaterium* bacterium, which exhibited intracellular granules.[72] The oil crisis in 1970s prompted the investigation for alternative compounds to substitute fossil plastics.[55] After its discovery by 50 years, PHB was produced on an industrial scale. PHAs composition varies based on the microorganism, the culture conditions, and the carbon source (Table 3).

PHB exhibits remarkable mechanical properties such as polyethylene when crystallinity becomes up to 70%. In addition, PHB is appropriate for the applications of food packaging due to its lamellar structure, which contributes to its useful properties as an aroma barrier with a permeability of water vapor. PHB has a close melting point compared to PLA, allowing both polymers to be mixed in their melted form. However, PHB's melt processing behavior and poor mechanical efficiency, that is, high fragility, complicated processing along with inadequate barrier properties, and low thermal stability, restrict its use.[73] Dozens of attempts are being made to enhance its packaging technology properties.[74,75] Melt mixing formulated plasticized PLA/PHB blends mixed with catechin exhibited improved mechanical properties, which showed potential as food bio-based active packaging material.[8]

Life cycle assessment represents that the energy requirements for PHB synthesis are lower than the needs of energy for traditional polyethylene and polypropylene. Furthermore, PHB production is more favorable for preventing ozone layer, global warming, abiotic depletion, and toxicity levels.[76,77]

3.5 | Blending starch with biopolymers

Starch mixes have also been investigated with biologically degradable synthetic polymers. Examples of biodegradable synthetic polymers

TABLE 3 Production of polyhydroxyalkanoates by diverse bacteria

Bacterial strain	Carbon source	PHA
Burkholderia sp. DSMZ 9243	Gluconate or sucrose	PHB P(3HPE)
Ralstonia eutropha H16	Fructose, Glucose, valeric acid, acetic acid	PHB PHV
Burkholderia sacchari IPT 189	Propionic acid	P(3HB-CO-3 HV)
Burkholderia cepacia ATCC 17759	Levulinic acid, xylose,	P(3HB-CO-3 HV)
Burkholderia xenovorans LB400	Glucose	PHB

Note: ND: not determined. 3-Hydroxy-4-pentanoic acid. Ratio (wt/vol in % 2.2: (0.07–0.52). Ratio (wt/wt) 10:1, 19:1, 30:1, 61.5:1.

involve polyvinyl alcohol, polyester carbonate, tetraphthalate, polylactic acid, polycaprolactone, polyethylene modified, and other aliphatic polyesters. Such products are mixed with starch for cost reduction because they are relatively expensive.

3.5.1 | Starch/PLA blend

The blend of PLA with starch could reduce cost and improve composite biodegradation but not without some changes on the properties of composite like a slash in mechanical properties such as elongation and tensile strength and an increase in water sensitivity owing to the hydrophilic nature of native starch.[78,79] It is known that with increasing starch content in the starch/PLA composite enhances the biodegradability; however, the incompatibility problem between PLA matrix and starch granules is still weak due to their polarities difference. For this reason, compatibilizers are added, such as acrylic acid,[79] methylene diphenyl diisocyanate,[80] and maleic anhydride.[81] Such hybrid composition has exhibited better interfacial bonding between PLA matric and stash granules.

3.5.2 | Starch/PCL blend

In an analogous improvement, Shin et al[82] noted good compatibility between PCL and plasticized corn starch; however, they figured out that the blend was thermodynamically immiscible, owing to the glass transition temperatures and a melting point of PCL remained unchanged with the addition of TPS. They reported that the formation of hydrogen bonds between the starch hydroxyl groups and the ester carbonyl groups of PCL might be the reason of bad compatibility. In conclusion, as TPS percentage increased, both elongation and tensile strength decreased while an upward trend of modulus was observed.

3.6 | Feasibility of bioplastics implementation, future aspect, and recommendations

Now, the majority of the available bioplastics are produced from agricultural feedstocks. These feedstocks are not considered optimum solutions for aligning with the UN's sustainable development goals because of their competition of arable land, high water consumption, and competition with food production.[83] Such reasons are probably the main reasons for the high cost of bioplastics compared to petroleum plastics. Other feedstocks alternatives have been reported in the literature. One of the promising solutions is the microalgae-based bioplastic. Microalgae can be grown in nonarable land, use saline, and/or wastewater and enable useful nutrients (such as nitrogen and phosphorus) recycling in the agriculture system. This behavior would remarkably reduce the need for chemical fertilizers.[84] Another direction to produce cleaner bioplastic is from agricultural wastes. This direction allows the production of a value-added product with reduced waste volume. Many candidates of agriculture residues are available and used by many authors to produce value-added bioplastics such as rice straw,[85] tea waste,[86] sugarcane bagasse fiber,[87] crop waste of date palm fruit,[88] cotton, and coffee wastes.[89]

4 | CONCLUSION

The main aim of designing a biodegradable polyolefin as commodity plastic is to retain functionality for the required job life but degrade to nontoxic end materials in a disposal environment. Based on the extensive survey performed in this review, it is undisputed that oxo-degradable plastic, including plastic carrier bags, degrades quicker in the open environment than conventional plastic. Complete biodegradability of oxo-biodegradable plastics probably occurs once the molecular weight is below 5000. The molecular weight can reach at this range if the plastic remained for not less than 1 year in the environment, which has warmth, bacterial activity, and moisture. At this level, the polymer no longer exists and is a wide range of discrete oxidized species. Bacteria and fungi find these species suitable for a carbon source that can be assimilated into the lifecycle resulting in the formation of biomass and CO2 under aerobic conditions. The process of biodegradation is challenged by molecular weight reduction, since this process cannot be guaranteed. If an oxo-biodegradable bag reaches the soil before molecular weight reduction, it may remain for decades before being biodegraded. Also, conventional plastics (without oxo additives) remain more if they reached the soil. When it comes to composting, many previous publications stated that the biodegradation in the compost environment is very slow, which then produces compost with plastic residual. Also, the research published on the biodegradation of oxo-biodegradable plastic in the marine environment is rare. There are concerns from reaching the oxo-biodegradable microfragments to the marine because of the risk of plastic uptake by fish increases when plastic sizes become smaller. Therefore, more studies should consider the biodegradation of oxo-products in the marine environment. It can be concluded from this review that there was no previous study had reported a 100% complete biodegradation of oxo-biodegradable plastic. Such fining accompanied with banning the oxo-plastics in Europe suggest the restricted use of oxo-degradable plastics in the future. Bioplastics instead seem to occupy a higher market share in the near future due to their eco-friendly nature.

CONFLICT OF INTEREST

The authors declare no potential conflict of interest.

DATA AVAILABILITY STATEMENT

As this is a review article, data sharing is not applicable here.

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