





Prooxidant-based polyolefins exhibiting no evidence of biodegradation under marine environments

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ABSTRACT

The use of oxo-biodegradable plastics has become increasingly controversial due to insufficient evidence supporting their environmental benefits. In particular, their biodegradability in marine environments remains poorly understood, sparking a global concern that the rapid abiotic fragmentation of these plastics could harm marine ecosystems. Here, we assessed the marine biodegradability of polyethylene and polypropylene films containing a commercial oxo-biodegradable prooxidant and compared them to their untreated counterparts using biochemical oxygen demand tests. Our findings revealed that while the prooxidant enhanced oxidative degradation and fragmentation of the polymers, it did not improve their marine biodegradability. Neither the initial nor the UV-exposed samples, with or without the prooxidant, exhibited significant marine biodegradability. The accelerated oxidative breakdown of these so-called oxo-biodegradable polyolefins, coupled with their limited marine biodegradability, may worsen the ocean microplastic pollution. Therefore, the potential environmental risks of using prooxidants should be carefully considered to prevent unintended negative impacts on the environment.

1. Introduction

Polyolefin-based plastic products are ubiquitous in our daily lives. Single-use items, such as packaging films and various disposable supplies, have become a significant part of the current throw-away culture, inevitably contributing to the global plastic waste crisis (Zhang et al., 2023; Wang et al., 2021; Thomas Allen, 2017). Because of their inherent chemical stability and bioinertness, discarded polyolefins tend to accumulate and fragment through weathering into microplastics (MPs), which persistently contaminate terrestrial and marine ecosystems (Wang et al., 2021; Ammala et al., 2011; Wiles and Scott, 2006; Scott and Wiles, 2001; Horton and Barnes, 2020; Colwell et al., 2023; An et al., 2024a, 2024b). This issue is of particular concern, because commodity polymers barely biodegrade in marine environments (Zhang et al., 2023; Wang et al., 2021; García Rellán et al., 2023; Erni-Cassola et al., 2019). It is estimated that over one-third of plastic waste in oceans consists of polyolefins, with polyethylene (PE) being the most prevalent

type of marine plastic debris (Wang et al., 2021; Hänninen et al., 2021). In response, significant efforts have been made to develop degradable polyolefins that retain desirable properties during their service life while biodegrading in disposal environments in a timely manner (Ammala et al., 2011; Wiles and Scott, 2006; Scott and Wiles, 2001; Abdelmoez et al., 2021).

Polyolefins are not hydrodegradable; their biodegradation requires an initial abiotic fragmentation into low molecular weight (M_w) fragments (<5000 Da) (Ammala et al., 2011) that can then be bioassimilated by microorganisms (Wiles and Scott, 2006; Scott and Wiles, 2001; Scott, 1976). This two-stage process forms the basis of the concept of oxo-biodegradable plastics, which undergo oxidative degradation, typically catalyzed by prooxidants, followed by biodegradation of the resulting oxidation products (Abdelmoez et al., 2021; Scott, 1976; Albertsson et al., 1995). Prooxidants are generally based on transition metal salts or complexes that decompose to generate free radicals, catalyzing the formation of carbonyl (C=O) compounds, which then

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undergo the Norrish type I and type II cleavages (Wiles and Scott, 2006; Scott and Wiles, 2001; Albertsson et al., 1987).

Although prooxidant-containing polyolefins have been marketed for more than four decades, uncertainty and debates surrounding their biodegradability and environmental impact continue to exist (Scott and Wiles, 2001; Sciscione et al., 2023). Moreover, concern has emerged that their accelerated breakdown may speed up the MP accumulation in terrestrial and freshwater ecosystems, which are primary sources of marine plastic debris, thereby aggravating the ocean MP pollution—one of today's most pressing environmental issues (Wang et al., 2021; Sciscione et al., 2023). Additionally, MPs can further fragment into nanoplastics, which are likely to pose the greatest ecological risk (Horton and Barnes, 2020). Consequently, there is a need for conclusive evidence regarding the marine biodegradation of these so-called oxo-biodegradable polymers. Herein, we focused on a biodegradable prooxidant additive known as P-Life (P-Life Japan, Inc.). Jakubowicz et al. reported that 91 % biodegradability of P-Life-based low-density PE (LDPE) films pre-oxidized at 65 °C for 240 h (M_w : 8800, M_n : 1700) has been observed after two years of the soil biodegradation test at 23 °C (Jakubowicz et al., 2011). Our previous research confirmed the enhanced photooxidative degradation of polyolefins containing P-Life through accelerated UV exposure tests (Padermshoke et al., 2022). However, direct experimental evidence regarding the marine biodegradability of P-Life-based plastics and similar materials is scarce (Abdelmoez et al., 2021; Sciscione et al., 2023; Zumstein et al., 2019). The impact of prooxidants on the marine biodegradation of polyolefins remains unclear. Therefore, assessing the marine biodegradability of these polymers will provide critical insights and significantly contribute to the growing body of evidence regarding their benefits and potential environmental risks.

In this study, we investigated the marine biodegradability of four polyolefin polymers containing P-Life, high-density PE (HDPE), LDPE, linear low-density PE (LLDPE), and isotactic polypropylene (*it*PP), and compared them with their neat counterparts, using biochemical oxygen demand (BOD) tests. The PE and PP polymers were chosen for their substantial contribution to marine plastic pollutants (Wang et al., 2021; Hänninen et al., 2021). P-Life masterbatch samples were subjected to real marine conditions through field tests conducted in the ocean, followed by chemical analysis using Fourier transform-infrared (FT-IR) spectroscopy. The effects of prior photooxidation on marine biodegradability were examined using UV-exposed polyolefin samples with and without P-Life.

2. Materials and methods

2.1. P-Life additive and polyolefin sample films

The additive masterbatches—P-Life Green20 (20/80 wt% prooxidant/bio-based LDPE) and P-Life PP20 (20/80 wt% prooxidant/bio-based PP)—were purchased from P-Life Japan Inc. and used as received. HDPE (melt mass flow rate (MFR) = 5.7 g/10 min) and LLDPE (MFR = 3.5 g/10 min) were supplied by Prime Polymer Co., Ltd.; LDPE (MFR = 7 g/10 min) was provided by Sumitomo Chemical Co., Ltd., and *it*PP (MFR = 11 g/10 min) was obtained from Japan Polypropylene Corporation. P-Life Green20 and P-Life PP20 were used to prepare oxo-biodegradable PE and *it*PP films of approximately 200- μ m thickness, respectively. Non-oriented films of the HDPE, LDPE, LLDPE, and *it*PP samples were prepared by hot pressing between steel plates with a 0.2 mm-thick spacer of tetrafluoroethylene. PE and *it*PP were melted at 190 and 210 °C, respectively, for 4 min. Then, they were compression molded at 10 MPa for 2 min and quenched at 20 °C for 2 min. The mixture of 2 wt% masterbatch and 98 wt% neat polymer was extruded using a single screw extruder with a screw diameter of 30 mm (Hoshi Plastics Co., Ltd.), resulting in a final concentration of 0.4 wt% prooxidant. Operating conditions were at 60-rpm screw rotation and a barrel temperature of 190 °C for PE and 210 °C for *it*PP.

2.2. UV exposure test

PE and *it*PP sample films (dimensions: 1×1 or 1×2 cm²), both with and without P-Life, were irradiated in a UV exposure test chamber (Q-SUN Xe-1-s/Q-Lab) with the irradiance (300–400 nm) and black panel temperature of 125 W/m² and 63 °C, respectively (Fig. S1(a)). The air temperature inside the UV test chamber measured approximately 40 °C. The UV exposure test periods were up to 4 weeks, where the integrated irradiation was 302.4 MJ/m², which is approximately equal to a 1-year UV exposure in the Pacific Ocean coastal region of Honshu, Japan (Tomita et al., 2006). The UV-exposed films were then subjected to marine biodegradation tests.

The extent of photooxidation of each sample was assessed using a carbonyl index, which is a measure of the generated C=O compounds, calculated from the IR spectra of the sample. Scanning electron microscopy (SEM) was employed to observe the surface morphologies of the initial and UV-exposed P-Life-containing samples. Details of the FT-IR spectral acquisition, carbonyl index calculation, and SEM experiment are given in the Supporting Information.

2.3. Marine biodegradation test

Accelerated biodegradation tests were performed in the laboratory using extracted seawater prepared according to the procedure proposed by Taguchi et al. (2021). Seawater and sediments were collected from Imazu, Nishi-ward, Fukuoka, Japan. The extracted seawater was obtained by mixing 600 ml of filtered seawater and 100 g of sediment, which was then ultrasonicated for 10 s and immediately filtered through a filter bag (pore size = 10 μ m). The viable cell numbers in fresh and extracted seawater were measured in a Fuchs–Rosenthal counting chamber using a 5-cyano-2, 3-ditolyl tetrazolium chloride (CTC) rapid staining kit and a fluorescence microscope (Nikon ECLIPSE E400). A mixture containing 75 ml of the extracted seawater and 15 mg of a sample were used for evaluating the marine biodegradability at 25 °C following the test method recommended by ISO 23977-1. Cellulose (filter paper 5C, ADVANTEC) was chosen as a reference because of its versatile biodegradability (An et al., 2023).

The marine biodegradation of the samples was assessed using the BOD tests. The test period was 60 d (June–August 2023), during which the consumed O₂ was measured using a BOD sensor (OxiTop IDS, WTW, Weilheim, Germany) (Fig. S1(b)). The biodegradation degree (%) was calculated from the BOD and theoretical O₂ demand (ThOD) according to the following equations (An et al., 2023):

$$\text{BOD} = (\text{O}_2 \text{ uptake sample (mg/L)} - \text{O}_2 \text{ uptake blank (mg/L)}) \quad (1)$$

$$\text{Biodegradation degree (\%)} = (\text{BOD/ThOD}) \times 100 \quad (2)$$

where O₂ uptake sample and O₂ uptake blank are the amounts of O₂ consumed by the samples and blank controls, respectively. The ThOD is the maximum theoretical amount of O₂ required to completely convert a polymer's carbon to CO₂, calculated from the molecular formula of the polymer.

Field tests of the PP20 masterbatch films (dimensions: $5 \times 5 \times 0.05$ cm³) were conducted at Ainan, Ehime, Japan (32.9644°N, 132.5045°E) from June 7 to July 7, 2023. A schematic of the sample setup and positions is shown in Fig. S2. Attenuated total reflection (ATR) FT-IR spectra of the field-test samples were collected with a Ge hemispherical crystal (incident angle: 30°) in the 4000–600 cm⁻¹ region at a resolution of 2 cm⁻¹ and 64 scans using a VERTEX 70 FT-IR spectrometer (Bruker). The marine biodegradation of the neat and P-Life-containing UV-exposed polyolefin films was evaluated using a titration method, as described in Fig. S3.

3. Results and discussion

3.1. Catalytic photo-oxidative degradation of polyolefins containing P-Life

P-Life was identified as a mixture of Mn palmitate and stearate (Padermshoke et al., 2022). The ability to switch between two oxidation states of the transition metals facilitates photooxidation of polymers via autooxidation process (Scott, 1976). The description and schematic of P-Life-catalyzed photooxidation of the polyolefin polymers are presented in Scheme S1. The formation of oxidation products in the UV-exposed polyolefin samples was monitored using FT-IR spectroscopy. UV-irradiation dependent transmission FT-IR spectra of HDPE and ATR FT-IR spectra of all the samples are shown in Figs. 1 and 2, respectively. The relatively low oxidation degree of HDPE allows for the observation of the transmission C=O stretching ($\nu_{C=O}$) bands throughout the 4-week UV exposure test period (Fig. 1). The $\nu_{C=O}$ (1780–1650 cm^{-1}), $\nu_{C=C}$ stretching ($\nu_{C=C}$) (1641 cm^{-1}), and $\text{H}_2\text{C}=\text{C}$ bending ($\delta_{C=C}$) (909 cm^{-1}) bands developed and continued to grow in intensity upon UV irradiation, suggesting the accumulation of various oxidation products formed via both the Norrish type I and type II cleavages (Albertsson et al., 1987). The ATR FT-IR spectra (Fig. 2) also revealed the presence of C=O compounds in all the UV-exposed polymers. As anticipated, the IR bands attributed to oxidation products intensified in the presence of P-Life as a result of the accelerated oxidation process.

The $\nu_{C=O}$ region contains several overlapped bands arising from different C=O compounds. Our previous work indicated that the relative contents of the oxidation products that are derived from ketones, e. g., esters (1735 cm^{-1}), carboxylic acids (1700 cm^{-1}), and lactones (1770 cm^{-1}), increased with UV exposure time (Padermshoke et al., 2022). Since the formation of these secondary products requires the photolysis of the ketonic C=O groups, the increase in their contents is indicative of the progressive chain scission and, thus, accumulation of low M_w C=O compounds in the polymers during prolonged UV exposure.

The extent of photooxidation of each polyolefin sample was determined using the carbonyl index. Fig. 3 compares the carbonyl index calculated for the surface (a) and bulk (b) regions of the UV-exposed polyolefin films, based on the UV-irradiation-dependent ATR and transmission FT-IR spectra, respectively. An increase in carbonyl index upon UV aging generally implies that the degradation of the polymers to lower M_w products is occurring (Ammala et al., 2011). P-Life significantly promotes oxidation, particularly in the bulk of all polyolefin samples, as indicated by the higher carbonyl index values observed in the P-Life-containing sample films. The increase in carbonyl index is particularly pronounced in the PE polymers, which are typically more

resistant to oxidation. In the transmission FT-IR spectra of the P-Life-containing LDPE and LLDPE films, the $\nu_{C=O}$ bands become saturated after 1–2 weeks of UV exposure, indicating the extensive formation of the C=O compounds due to the enhanced oxidation within the bulk of these polymers. The greater oxidation enhancements observed for LDPE and LLDPE, relative to HDPE, were largely due to their greater amorphous contents, i.e., lower degrees of crystallinity, as the mobility of O_2 , additives, and macroradicals is higher in amorphous regions than in highly ordered crystalline phases. Moreover, the greater degrees of branching made LDPE and LLDPE more liable to chemical attacks, further facilitating the degradation process (Padermshoke et al., 2022).

All UV-exposed samples exhibit severe surface deterioration, embrittlement, and disintegration (Fig. S4), reflecting a loss of mechanical properties due to the oxidative scission of the polymer main chains (An et al., 2024a; Padermshoke et al., 2024). Such damage promotes further degradation of the oxidized polymers through the increased exposed surface area; the low M_w oxidation products thus formed are subject to biodegradation, provided the receiving environment is microbially active.

3.2. Marine biodegradation of polyolefins containing P-Life

In this study, we investigated the marine biodegradation of HDPE, LDPE, LLDPE, and *it*PP films, both with and without P-Life, using the extracted seawater. The viable cell concentration in the extracted seawater (5.5×10^6 cells/mL) was markedly higher than that of fresh seawater (6.5×10^4 cells/mL), enabling a more rapid assessment of the marine biodegradability of the polymers.

Fig. 4 illustrates the biodegradation percentages for all the polyolefin samples, along with the cellulose reference. Cellulose gets readily biodegraded, displaying a linear increase in the biodegradation degree, reaching 32 % by the end of the test period. This confirms the suitability of the extracted seawater for evaluating the biodegradation of polymers in marine environments. In contrast, none of the polyolefin samples exhibit significant biodegradability in the extracted seawater. The biodegradation percentages of all the samples, both with and without P-Life, remain close to 0 % throughout the test period.

Furthermore, the field-test samples reveal a substantial loss of the P-Life additive from the sample surface, as shown in the ATR FT-IR spectra in Fig. 5. The IR bands associated with P-Life, such as carboxylate (COO^-), fatty acid CH stretching, and methylene band progression (1350–1180 cm^{-1}), decrease in intensity after the one-month field tests. Notably, the PP20-top (3) sample shows almost no IR spectral contribution from the P-Life additive, indicating a nearly complete leaching of P-Life from the sample surface into the ocean. This partly explains the lack of biodegradability of the P-Life-containing polyolefin films in the

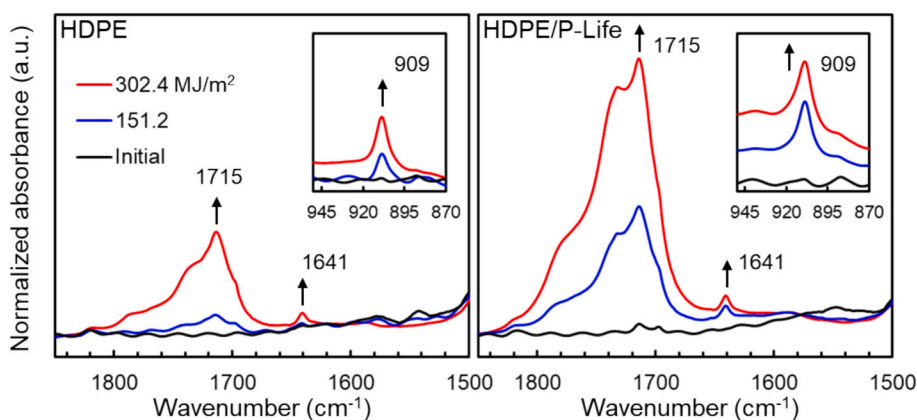


Fig. 1. Transmission FT-IR spectra in the $\nu_{C=O}$ (1780–1650 cm^{-1}), $\nu_{C=C}$ (1641 cm^{-1}), and $\delta_{C=C}$ (909 cm^{-1}) regions of HDPE without (left) and with (right) P-Life collected as a function of the integrated UV irradiation (0, 151.2 (2-week UV), and 302.4 (4-week UV) MJ/m^2). Arrows indicate the directions of spectral changes with the increasing integrated UV irradiation.

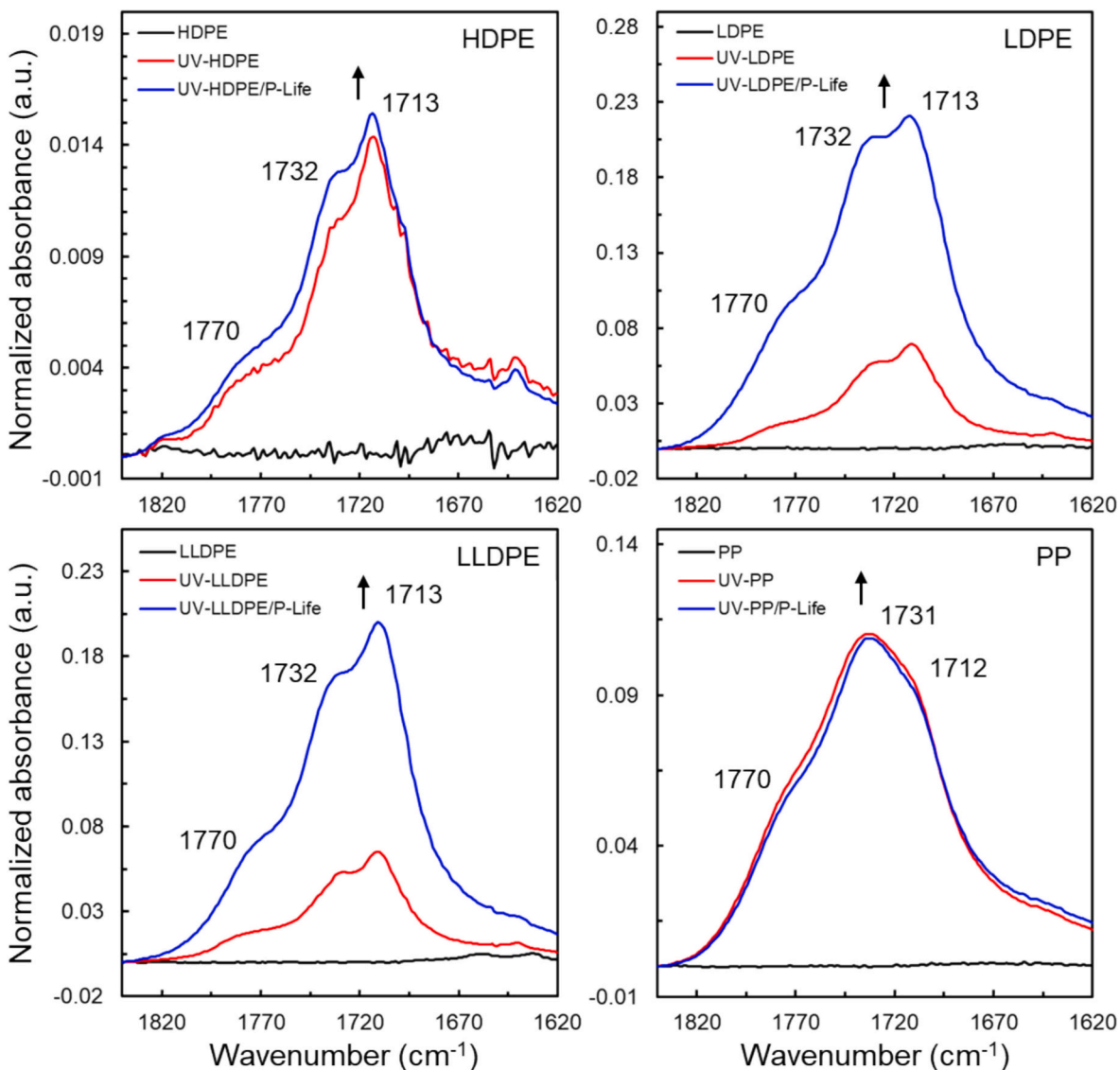


Fig. 2. ATR FT-IR spectra in the $\nu_{C=O}$ (1780–1650 cm^{-1}) region of the initial and 4-week UV-exposed HDPE, LDPE, LLDPE, and *i*tPP without (red) and with (blue) P-Life (integrated UV irradiation: 302.4 MJ/m^2). Arrows indicate the directions of spectral changes upon UV irradiation. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

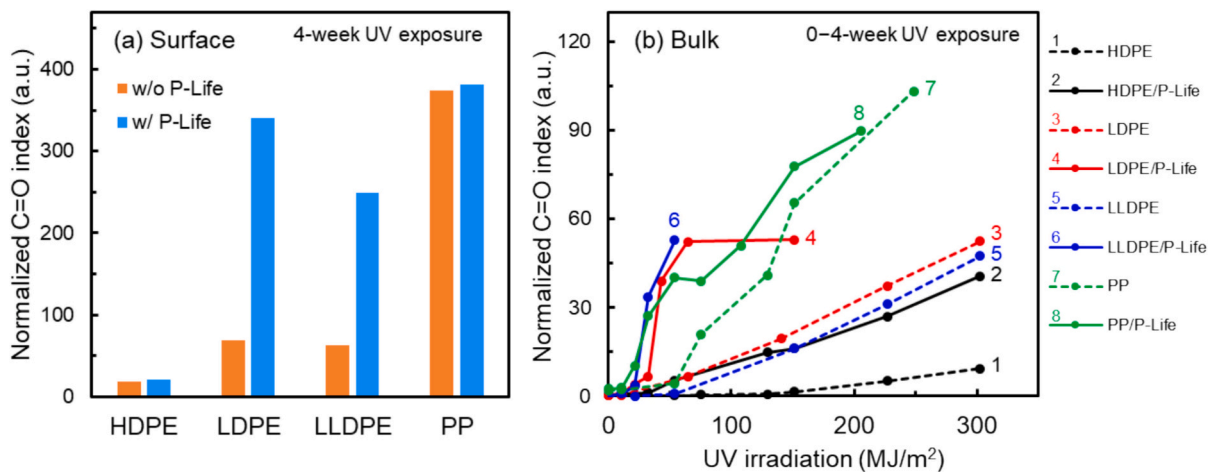


Fig. 3. Comparison of the surface (a) and bulk (b) carbonyl index of HDPE, LDPE, LLDPE, and *i*tPP films with and without P-Life calculated from the UV-irradiation dependent ATR and transmission FT-IR spectra, respectively (integrated UV irradiation: 0–302.4 MJ/m^2).

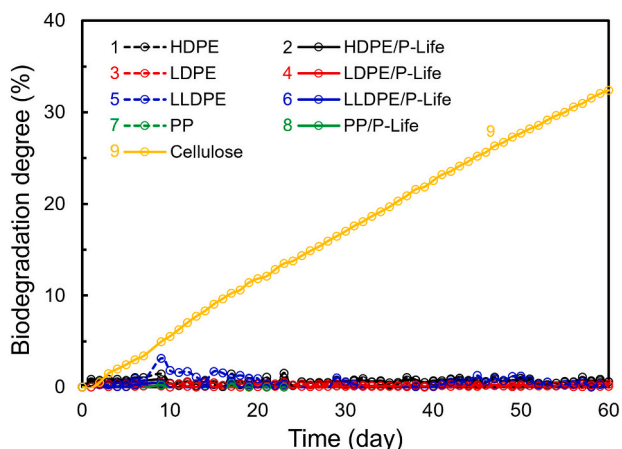


Fig. 4. Biodegradation degree (%) of HDPE, LDPE, LLDPE, and *it*PP films with and without P-Life obtained during the 60-d biodegradation test period using cellulose as a reference.

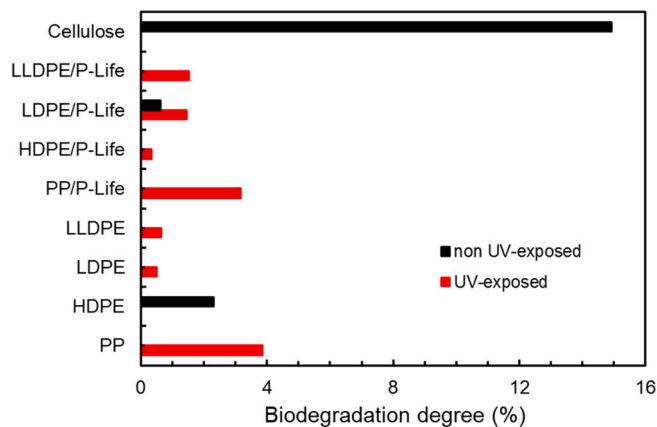


Fig. 6. Biodegradation degree (%) of initial and UV-exposed HDPE, LDPE, LLDPE, and *it*PP films without and with P-Life obtained during the 30-d biodegradation test period using cellulose as a reference (titration method).

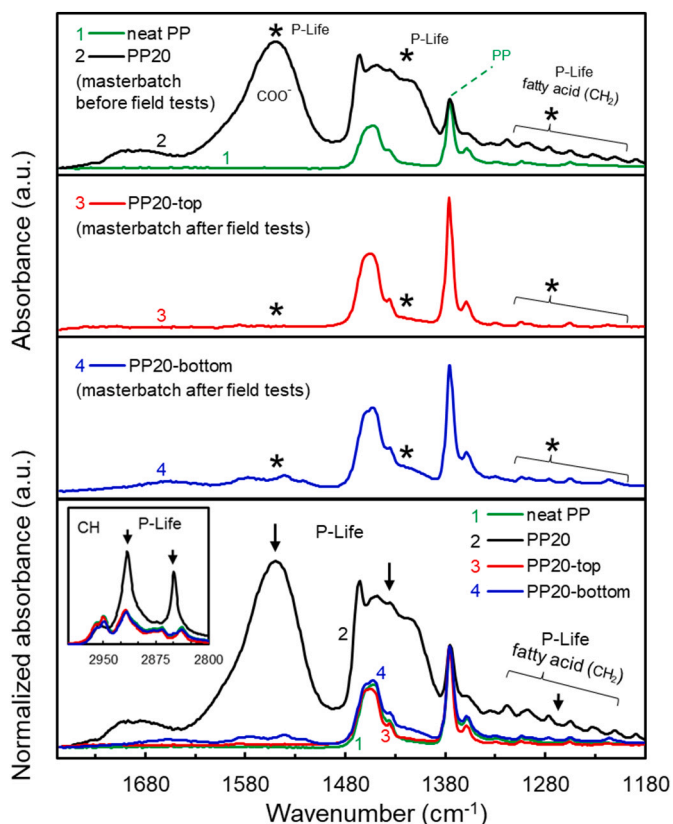


Fig. 5. ATR FT-IR spectra of the neat PP (1) and PP20 masterbatch films before (2) and after (3 and 4) the one-month field tests. The samples were positioned near the sea surface (PP20-top (3)) and at the bottom (PP20-bottom (4)) (approximate depth: 20 m). Asterisks denote IR band positions of P-Life. Arrows indicate the decrease in the intensities of IR bands of P-Life after the one-month field tests.

marine environment.

The marine biodegradability of the P-Life-containing polyolefin films with and without prior UV exposure was compared with that of their respective neat samples, as shown in Fig. 6. By the end of the 30-d test period, the biodegradation degree of cellulose reaches approximately 15 %, whereas all polyolefin samples exhibit minimal biodegradability (0–4 %). Notably, despite the significantly enhanced oxidative

degradation observed in the P-Life-containing polyolefin films, no sample shows improved marine biodegradability compared with that of the neat polymers. These findings indicate that both the initial and UV-exposed polyolefin films exhibit negligible biodegradation in the marine environment, regardless of the presence of P-Life.

Given that polymer biodegradation depends not only on material properties but also on the characteristics of the receiving environments (Zumstein et al., 2019), the poor marine biodegradability of P-Life-containing polyolefin polymers, in contrast to their biodegradability in soil (Jakubowicz et al., 2011), can likely be attributed to differences in environmental parameters such as temperature, salinity, nutrient availability, and microbial diversity between the two environments. Additionally, the leaching of P-Life from the sample surface into seawater, as noted in the field tests, may have reduced its catalytic efficiency in marine environments.

Even though enhanced biodegradation of prooxidant-based polymers in soil has been noted, there exists considerable uncertainty regarding the biodegradation rates and timescales required for complete mineralization (Abdelmoez et al., 2021; Sciscione et al., 2023; Selke et al., 2015; Napper and Thompson, 2019). These land-based polymer fragments can continue to disintegrate into MPs, posing a risk of contaminating marine ecosystems (Zhang et al., 2023) if biodegradation is not completed within an appropriate timeframe. Once distributed in the ocean, polyolefin MPs can accumulate toxic chemicals, such as polychlorinatedbiphenyls (PCBs) and dichlorodiphenyldichloroethylene (DDE) and transport them to marine food webs (Mato et al., 2001; Andradý, 2011). They are also a potential source of other ocean pollutants associated with plastic additives or their degradation products (Mato et al., 2001; Andradý, 2011). Therefore, the rapid abiotic disintegration of the prooxidant-containing polyolefins, combined with their lack of marine biodegradability, as evidenced here, emphasize the potentially harmful environmental impact of using oxo-biodegradable additives.

The present findings are consistent with previous studies that have revealed the lack of biodegradability of polyolefins marketed as oxo-biodegradable plastics in different simulated and natural environments, including open-air and marine settings (Selke et al., 2015; Napper and Thompson, 2019; Cheung and Not, 2024). The comparatively lower degradative stress present in natural environments, in contrast to the controlled conditions of laboratory testing, may primarily account for the observed limited biodegradability of the oxo-biodegradable-labeled plastics in real-world environments (Cheung and Not, 2024).

4. Conclusion

We evaluated the marine biodegradability of HDPE, LDPE, LLDPE,

and *it*PP films with and without the P-Life prooxidant in the extracted seawater using the laboratory BOD tests. The influence of photo-oxidative degradation on marine biodegradation was assessed using UV-exposed polyolefin films. None of the neat or additive-containing polyolefin samples demonstrated significant marine biodegradability under the test conditions, regardless of prior UV aging. This study revealed that, although P-Life effectively promoted the oxidative degradation and fragmentation of polyolefin polymers, it did not improve their marine biodegradability.

The present findings also highlight that commercial oxo-biodegradable polymers certified biodegradable under specific conditions may not biodegrade in different disposal environments. Moreover, the consequences of prooxidant leaching into the ocean require further clarification. Notably, the absence of conclusive evidence supporting the environmental benefits of oxo-biodegradable plastics has led the European Commission to restrict their use in the European Union (*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, n.d.*; *Study to provide information supplementing the study on the impact of the use of "oxo-degradable" plastic on the environment, n.d.*). Furthermore, over 150 organizations have endorsed an oxo-statement advocating a global ban on oxo-degradable plastic packaging (*Oxo statement, 2017*). This work demonstrates that marine ecosystems are not suitable environments for the biodegradation of prooxidant-based polyolefins. It underscores the importance of thoroughly evaluating the practical applicability and limitations of oxo-biodegradable additives to prevent unintended negative impacts on the global environment.

CRediT authorship contribution statement

Adchara Padermshoke: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Yingjun An:** Writing – review & editing, Investigation, Data curation. **Thin Van Nguyen:** Writing – review & editing. **Yutaka Kobayashi:** Resources. **Hiroshi Ito:** Writing – review & editing. **Atsushi Takahara:** Supervision, Resources, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Atsushi Takahara reports equipment, drugs, or supplies and travel were provided by Kyushu University. Atsushi Takahara reports a relationship with National University Corporation Kyushu University that includes: employment. If there are other authors, they 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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Appendix A. Supplementary data

Schematic of the UV exposure test and the accelerated marine biodegradation test, details of the FT-IR spectral acquisition and calculation of carbonyl index, schematic of the sample setup and positions in the field tests, photograph of the initial PP20 film and laser microscopy images of the field-test films, details of the accelerated marine biodegradation tests using a titration method, description and schematic of P-Life-catalyzed photooxidation of the polyolefin polymers, SEM images of the initial and UV-exposed polyolefin films with P-Life. These materials

are available online free of charge. Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2025.117697>.

Data availability

Data will be made available on request.

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