

Review

Biodegradability of bioplastics in different aquatic environments: A systematic review

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ARTICLE INFO

Article history: Received 8 March 2023 Revised 8 June 2023 Accepted 8 June 2023 Available online 19 June 2023

Keywords: Biodegradation Bioplastic Plastic Natural environment Plastic waste

ABSTRACT

Bioplastics were first introduced as environmentally friendly materials, with properties similar to those of conventional plastics. A bioplastic is defined as biodegradable if it can be decomposed into carbon dioxide under aerobic degradation, or methane and CO₂ under anaerobic conditions, inorganic compounds, and new cellular biomass, by the action of naturally occurring microorganisms. This definition however does not provide any information on the environmental conditions, timescale and extent at which decomposition processes should occur. With regard to the aquatic environment, recognized standards have been established to assess the ability of plastics to undergo biodegradation; however, these standards fail to provide clear targets to be met to allow labelling of a bioplastic as biodegradable. Moreover, these standards grant the user an extensive leeway in the choice of process parameters. For these reasons, the comparison of results deriving from different studies is challenging.

The authors analysed and discussed the degree of biodegradability of a series of biodegradable bioplastics in aquatic environments (both fresh and salt water) using the results obtained in the laboratory and from on-site testing in the context of different research studies. Biochemical Oxygen Demand (BOD), CO₂ evolution, surface erosion and weight loss were the main parameters used by researchers to describe the percentage of biodegradation. The results showed a large variability both in weight loss and BOD, even when evaluating the same type of bioplastics. This confirms the need for a reference range of values to be established with regard to parameters applied in defining the biodegradability of bioplastics. © 2024 The Research Center for Eco-Environmental Sciences, Chinese Academy of

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https://doi.org/10.1016/j.jes.2023.06.013

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Introduction

A large percentage of plastic waste is abandoned in the environment by humans, first reaching urban waterways, such as rivers and canals, which act as "plastic highways" in transporting the waste from the cities to the sea. Approximately 80% of marine plastic pollution derives from terrestrial litter (Canal and River Trust and Coventry University, 2019; GESAMP, 2019; Munari et al., 2021).

The most of fossil-based plastics persist in the environment for long periods due to their high resistance to microbial degradation (Andler et al., 2022). When dispersed in the aquatic environment, plastics may float and accumulate generating plastic islands; they may be mechanically broken down into micro- or nano-plastics by the action of UV radiation, wind and waves; they may be ingested by living organisms, bioaccumulating in the food chain or causing their death; and they may become a carrier of disease (Shruti and Kutralam-Muniasamy, 2019).

Despite this daunting picture, however, polymeric materials are essential for many applications and indispensable in numerous industries. Possible solutions might be the reduction of plastic use by redesigning the entire products supply chain, pursuing a vision of sustainable design (eco-design), or by replacing it with other bio-derived materials. Extended producer responsibility policies might be a further option to limit single-use products, which significantly contribute to the problem of marine litter.

Over the past decades, various typologies of bioplastics have been introduced with the aim of partially replacing plastic products, featuring different properties for a series of possible applications (e.g. Peñalva et al., 2020). According to European Bioplastics (2021), a polymer is defined as a bioplastic if it is either bio-based, biodegradable, or features both properties (Fig. 1).

Therefore, the mere use of a "bio" prefix does not necessarily imply that the plastic is biodegradable, thus leading to confusion (Aluffi, 2020).



Fig. 1 – Different groups of bioplastics based on the origin of the raw material and environmental fate.

In view of the exponential expected growth in production of these biopolymers (it is estimated that the global bioplastics production capacity will increase from 2.08 million tons in 2020, to 7.54 million tons in 2026 (European Bioplastics, 2022)), it is of fundamental importance that a series of misconceptions related not only to the term "bioplastic" but also to the terms "biodegradable" and "compostable" should be overcome (Harrison et al., 2018).

Several definitions for "biodegradable" are available, although none establish the required criteria for labelling a substance as "biodegradable". The most frequently used definition states that a substance is "biodegradable" if it can be decomposed into carbon dioxide under aerobic degradation, or methane and carbon dioxide under anaerobic conditions, inorganic compounds and new cellular biomass, by the action of naturally-occurring microorganisms. However, no information is provided on the environmental conditions (temperature, presence of microorganisms etc.), timescale and extent at which the decomposition process should occur (Harrison et al., 2018). Without any timescale specification, all materials are therefore inherently biodegradable, whether it takes a few weeks or a million years to break down into water, carbon dioxide and methane. Moreover, the term biodegradable is often used interchangeably with degradable and disintegrable, resulting in misleading terminology.

Further confusion derives from the common belief that "biodegradable" and "compostable" are synonymous. The term "compostable" refers to the ability of an organic material to biodegrade and turn into compost (Lavagnolo et al., 2020). The ISO 17088:2021 defines compost as an "organic soil conditioner obtained by biodegradation of a mixture consisting principally of vegetable residues, occasionally with other organic material and having a limited mineral content". Unlike the definition of "biodegradable", the definition of "compostable" contains requirements to be met to define a plastic as "compostable". These are set by European standards EN 13432 and EN ISO 14995 as follows:

- Biodegradability: within six months 90% of the material must be assimilated by microorganisms and then converted into CO₂;
- Disintegrability: within three months 90% of the material must consist of fragments smaller than two millimetres;
- Absence of negative effects on the composting process;
- Low levels of heavy metals and no toxicity on final compost.

As a result, not all biodegradable bioplastics are necessarily compostable.

Therefore, when dealing with industrialized environments under controlled conditions (such as composting plants), a clear definition of the term "biodegradable" is found in different specification standards (such as ISO 18606:2013, EN 13432:2000 and EN 14995:2006). On the other hand, when it concerns natural environments, the biodegradation standards are testing standards, i.e. they describe the procedure to be followed for executing the test methods and the way in which to measure the results of the test without

Table 1 – Standards on the biodegradability of plastics in an aquatic environment (OECD standards are not included in the table below as they mainly relate to organic chemicals soluble in liquids).

| Standards | Description | Publication date |
|-------------------|--|------------------|
| EN ISO 14851:2019 | Determination of the ultimate aerobic biodegradability of plastic materials in an aqueous medium. Method by measuring the oxygen demand in a closed respirometer. | 1999 |
| EN ISO 14852:2021 | Determination of the ultimate aerobic biodegradability of plastic materials in an aqueous medium. Method by analysis of evolved carbon dioxide. | 1999 |
| EN ISO 18830:2017 | Plastics - Determination of aerobic biodegradation of non-floating plastic materials in a seawater/sandy sediment interface -Method by measuring the oxygen demand in closed respirometer. | 2016 |
| EN ISO 19679:2020 | Plastics - Determination of aerobic biodegradation of non-floating plastic materials in a seawater/sediment interface. Method by analysis of evolved carbon dioxide. | 2018 |
| EN ISO 22404:2021 | Plastics. Determination of the aerobic biodegradation of non-floating materials exposed to marine sediment. Method by analysis of evolved carbon dioxide. | 2019 |
| ISO 15314:2018 | Plastics. Methods for marine exposure. | 2005 |
| EN 14047:2002 | Packaging. Determination of the ultimate aerobic biodegradability of packaging materials in an aqueous medium. Method by analysis of evolved carbon dioxide. | 2002 |
| EN 14048:2002 | Packaging. Determination of the ultimate aerobic biodegradability of packaging materials in an aqueous medium. Method by measuring the oxygen demand in a closed respirometer. | 2002 |
| ASTM D6691-17 | Standard test method for determining aerobic biodegradation of plastic materials in the marine environment by a defined microbial consortium or natural sea water inoculum. | 2010 |
| ASTM D5209 – 92 | Standard test method for determining the aerobic biodegradation of plastic materials in the presence of municipal sewage sludge. | 1992 |
| ASTM D5271 – 02 | Standard test method for determining the aerobic biodegradation of plastic materials in an activated-sludge-wastewater-treatment system. | 1993 |
| ASTM D7991-22 | Standard test method for determining aerobic biodegradation of plastics buried in sandy marine sediment under controlled laboratory conditions. | 2015 |
| ASTM D7081-05 | Standard specifications for non-floating biodegradable plastics in the marine environment. | 2005 |
| JSA-JIS K 6950–94 | Plastics - Testing method for aerobic biodegradability by activated sludge. | 1994 |
| DIN V 54900–98 | Determination of the aerobic biodegradability of polymeric materials in aquatic batch tests. | 1998 |

providing any threshold values to be reached (Folino et al., 2023).

With regard to aquatic ecosystems, many different internationally recognized standards have been established to assess the biodegradability of plastics (Table 1); however, they all envisage standard procedures without providing clear criteria to be reached for the substance to be defined as "biodegradable" in this type of environment. Moreover, standardized methodologies leave the user with an extensive leeway in choosing process parameters (such as temperature, timeframe, inoculum concentration) and bioplastic shape and size. For these reasons, the comparison of results deriving from different studies is challenging.

Table 1 lists three different types of standards:

- EN ISO standards: this type of standards was initially issued by the International Organization for Standardization (ISO), becoming ISO standards only and subsequently implemented, at a European level, by the European Committee de Normalisation (CEN), becoming EN ISO standards.
- ISO standards: these standards were issued by the International Organization for Standardization (ISO). No implementation has been undertaken at European and national level.
- EN standards: these standards were issued by the European Committee de Normalisation (CEN). No internationally or nationally implemented version exists.

- ASTM-D standards: these standards were issued by the American Society for Testing and Materials (ASTM). The D20.96 Committee investigated test methods for waterinsoluble polymers and plastic materials.
- JSA-JIS standards: these standards were issued by the Japanese Industrial Standards Committee (JIS).
- DIN standards: these standards were issued by the German Institute for standardization (Deutsches Institut f
 ür Normung, DIN).

The oldest standards are EN ISO 14851 and EN ISO 14852, both established for a freshwater environment. The first evaluates biodegradation by measuring Biochemical Oxygen Demand (BOD), whilst the second evaluates biodegradation by measuring CO_2 produced.

EN ISO 18830 is based on EN ISO 14851 but substituting the test medium from freshwater to saltwater. Similarly, EN ISO 19679 is based on EN ISO 14852, but using salt water instead of fresh water. Both EN ISO 18830 and EN ISO 19679 are used to evaluate the biodegradation of plastic at the interface between seawater and bottom sandy marine sediments.

Procedure-wise, EN ISO 14852 was equivalent to ASTM D5209. It evaluates biodegradation by measuring the variation of carbon dioxide over time, residual polymer weight and Soluble Organic Carbon (SOC) content. ASTM D5271 was equivalent to EN ISO 14851 and assesses biodegradation by measur-

ing BOD and residual polymer weight. The test medium used in both standards is freshwater.

ASTM D6691 evaluates biodegradation by measuring the variation of CO_2 over time from a tested material immersed in saltwater medium.

The only standard that does not establish a test method, but rather the requirements to be met in labelling a product as "marine disposable", in accordance with guidelines issued by the Federal Trade Commission, is ASTM D7081. Regarding biodegradability requirement, the standard states that "a product is considered to have demonstrated inherent biodegradability if:

- a. 30% or more of the organic carbon is converted to carbon dioxide using Test Method D6691 (within 180 days at 30°C), when compared to a positive control. Furthermore, 90% biodegradation in an active environment such as compost must be demonstrated, in accordance with Test Method D5338.
- b. Following the Test Method D5338, to fulfil the requirements of this section plastics should achieve one of the following ratios of conversion to carbon dioxide found in I-III, within the time periods specified in IV or V:
 - I. For products consisting of a single polymer (homopolymers), 60% of organic carbon must be converted to carbon dioxide by the end of the test compared to positive control.
 - II. For all other polymers and substrates, 90% of organic carbon must be converted to carbon dioxide by the end of the test period compared to positive control.
 - III. For products consisting of more than one polymer, each individual polymer present at a concentration of more than 1% must achieve the 60% specification for homopolymers, as described in I.
 - IV. For materials that are not radiolabelled (referring to any compound that has been associated with a radioactive substance) the test period shall be no greater than 180 days.
 - V. If radiolabelled materials are used, the test period shall be no greater than 365 days".

A mark of conformity has also been developed for products that can be labelled as biodegradable in seawater ("Vinçotte OK Biodegradable MARINE"). The required percentage of absolute or relative biodegradation for a material/product to be labelled as "marine biodegradable" is equal to 90% within 6 months of testing. In addition, Vincotte introduced an "OK Biodegradable WATER" mark of conformity based on EN ISO 14851 and EN ISO 14852 standards, to label a product as "degradable in natural freshwater environment". If a product degrades at a rate of 90% over a period of 56 days incubation at a temperature of 20-25°C, it can be marked. These marks are not widely used, as they hold little appeal for either the producer or the consumer. In these cases, the requirements have been defined only to label the products and not to define biodegradability of the polymer used. In addition, these standards are difficult to meet as the product needs to be tested without any size reduction.

With the aim of collecting information on the types of biodegradability tests performed by different research labora-

tories and extrapolating range of parameters deemed useful in defining biodegradability of the different groups of bioplastics in aquatic environment, the authors analysed and discussed the literature results obtained for both fresh and salt water, also highlighting any knowledge gaps to be remedied to better address the research.

1. Materials and methods

The literature review was performed using Scopus and Web Of Science (WOS) databases, using the following construction for the three keyword strings formulation (Eq. (1)):

(1)

BP + BIODEGR + AE

where:

- BP = "bioplastics" or its synonyms. Subsequently, for more specific research, the name of the most widely-used biopolymers (PLA, PHA and PHB and PHBV, PBS, PES, PBSA, PGA, PCL, PBAT, PVA, TPS and cellulose) was used. BIODEG = "biodegradable" or its synonyms.
- AE = "aquatic environment", either salt- or fresh-water. Synonyms such as "fresh water", "river", "lake", "eutrophic reservoirs", "sea", "marine environment", "salt water" and "ocean" were also used.

Twenty-four strings were used: twelve for fresh water and twelve for salt water. The research was conducted on title, abstract and keywords. All scientific review papers thus obtained were automatically removed from the selected articles. In this scientific review only research papers were accepted.

This search found a total of 527 articles for fresh water and 993 articles for salt water. Subsequently, these articles underwent two consecutive screenings.

The first screening was focused on the exclusion or acceptance of an article based on analysis of title and abstract. The total number of articles that progressed to the second screening were 87 for freshwater and 222 for saltwater.

In the second screening, all articles accepted in the first screening were read in their entirety. At the end of this stage, 25 articles were accepted for freshwater and 67 articles for saltwater.

The reasons for exclusion applied in the second screening, divided by fresh and salt water, are illustrated in Fig. 2.

The same reasons were applied both freshwater and saltwater, although at different percentages. The two overriding reasons for exclusion, both in fresh and salt water, lay in the use of unconventional units of measurement, and the presence of review articles, which were not considered for the purpose of this scientific review. The absence of use of a clear standardized procedure in carrying out biodegradability tests on plastics placed the burden of decision on those performing the tests, thus giving rise to a plethora of results expressed with different units of measurement. This complicated the comparison of results obtained in the different biodegradability tests.

The most frequently used test medium was saltwater (76.60%) (Fig. 3), suggesting a greater interest in ascertain-



Fig. 2 - Reasons for exclusion applied in the second screening for saltwater (a) and freshwater (b).



Fig. 3 – Percentage of biodegradation test carried out in salt and fresh water and subdivision of tests according to the group of bioplastics tested in each of the two mediums. Blue indicates saltwater, green freshwater.

ing the fate of bioplastics, likewise also reflecting public concern over the presence of plastics in the food chain, starting from the ingestion of plastics by marine invertebrates in seas/oceans.

The most widely-tested amongst the different groups of bioplastics, was the bio-based group (55.60% in FW and 63.60% in SW) (Fig. 3). Within this group, the PHA and PLA families were found to be the most popular (Fig. 4). This result is in line with the finding that the most-widely produced bioplastics belong to these two families (European Bioplastics, 2022). Furthermore, this review highlights the presence on the market of an extremely large number of different bioplastics, a number which is expected to continue growing in the future. Given this broad panorama, the definition of a biodegradability standard is highly complex.

2. Results and discussion

The biodegradability experiments reviewed, previously grouped according to the test medium (fresh and salt water) and origin (biobased, partially biobased and fossil-based polymers), were further grouped according to test temperature, considered one of the most important parameters regulating the biodegradation process (Folino et al., 2020; Zhu and Wang, 2020) and test location (on site and off site).

The majority of freshwater tests were performed off-site (68.00%), while saltwater tests were largely performed on-site (55.60%). A preference for on-site tests in the case of saltwater might be due to the difficulty of reproducing the marine environment in the laboratory (temperature, water composition, microbial community) and to a wish to understand what happens to bioplastics if released into an actual natural environment.

The temperatures tested in both salt and fresh water, both as range and average values, in situ and off-site are summarized in Table 2.

The results obtained revealed a wide variability in tested temperature values The average temperature obtained in offsite tests in both salt and freshwater was approximately 25°C, and was used as a reference value to distinguish tests into two groups: tests carried out at a temperature below 25°C, and tests carried out at a temperature greater than or equal to 25°C. Data were then divided and represented according to the indicators used to evaluate degree of biodegradability of the bioplastic, such as:

- Weight Loss (WL) [% of initial weight];
- Biochemical Oxygen Demand (BOD) [% of ThOD];
- Biochemical Oxygen Demand (BOD) [mg/L];
- Carbon dioxide evolution (CO₂) [% of ThCO₂];



* includes: PHB, PHB/PHV, BIOPOL, P(3HB-co-4HB), P(3HB-co-3HV-co-3HB), PHBHH, ° includes: PLA, PLLA, PDLA, PLLA/PDLA

Fig. 4 – Number of times each type of polymer was tested, divided according to test medium and origin (biobased, partially biobased and fossil based). Acronyms provided by European Bioplastics have been used (European Bioplastics, 2022).

Table 2 – Temperature range and average of on-site and off-site tests carried out in salt and fresh water (in the case of tests carried out on-site, it proved impossible to calculate the average temperature due to the periodic variations of the operating temperature; N/A indicates non-available).

| | Freshwater | | Saltwater | | |
|--|---------------|-------------|----------------|----------------|--|
| | ON-SITE | OFF-SITE | ON-SITE | OFF-SITE | |
| Temperature range (°C) Average Temperature (°C) | 3.5–31 N/A | 20–60 26 | -1.7–32 N/A | 10–60 24.50 | |

• Surface Erosion (SE) [% of initial surface].

Table 3 provides a quantitative indication of the occurrence of different indicators (used to describe biodegradation) in the selected tests.

The most frequently used indicator is WL, which provides information relating to weight reduction of the bioplastics during the experiments. This feature is commonly associated with material degradation (including both biotic and abiotic degradation contributes), but has not been standardized in the norms mentioned above.

In some tests, biodegradability results are expressed both as WL and SE, both of which indicate disintegration of the material. Although SE is one of the parameters taken into consideration when assessing compostability of any given material (EN 13432:2000), it is not mentioned in the definition of biodegradability in the same norm. The second most commonly used indicator is BOD (%), together with CO₂, the reference indicator for biodegradability assessment in standard ISO 14851:2021.

As WL and BOD (%) were the most commonly used indicators, literature results have been illustrated in Fig. 5 in terms of WL and BOD (%). In the same figure, two different graphical representations were used: boxplots for statistically relevant results (occurrence more than 8 times) and dots in the other cases (occurrence less than 8 times).

The results obtained in terms of both WL and BOD (%) were affected by a wide variability. The majority of tests investigated biobased biodegradable bioplastics under all different environmental conditions (freshwater and saltwater, temperature higher and lower to 25°C). Then majority of tests on partially-biobased and fossil based biodegradable bioplastics were performed in saltwater, confirming the increased interest in investigating the fate of bioplastics in marine environments.

In Fig. 5, average value is illustrated by the inner horizontal line in the box plots. In general, lower temperatures were associated with a lesser degree of biodegradation, in terms of both WL and BOD consumed. This is due to the correlation between temperature and biological kinetics as illustrated by Miksch et al. (2022). Moreover, graphs (a1), (a2) and (b2) indicated a higher degree of biodegradation (both in terms of WL and BOD) within the same temperature range, in freshwater. This is justified by the fact that a marine environment may ••

c1. :

| | | | WL | BOD (%) | BOD (mg/L) | CO ₂ | SE | References |
|-----------------------|----|--|--------|---------|------------|-----------------|----|---|
| BIOBASED | FW | T < 25°C | 15 | - | 4 | - | - | (Volova et al., 2007), (Brandl and Piichner, 1991), (Wang et al., 2005), (Yadav and Hakkarainen, 2022), (Olewnik-Kruszkowska et al., 2020), (Walczak et al., 2015) |
| | | $T \ge 25^{\circ}C$ | 18 | 8 | - | - | - | (Ho et al., 2002), (Salim et al., 2012), (Yadav and Hakkarainen, 2022), (Lopez-Llorca et al., 1994), (Brdlík et al., 2022), (Ikejima et al., 1999), (Kusaka et al., 1999), (Abe and Doi, 1996), (Koyama and Doi, 1996), (He et al., 2020) |
| | SW | T < 25°C | 69 | - | - | 1 | 12 | (Huang et al., 2020), (Delacuvellerie et al., 2021), (Beltrán-Sanahuja et al., 2020), (Pelegrini et al., 2016), (Chaabane et al., 2022), (Gerritse et al., 2020), (Tsuji and Suzuyoshi, 2002b), (Kumar et al., 2022), (Rutkowska et al., 2008), (Briassoulis et al., 2019), (Thellen et al., 2008), (Wang et al., 2005), (Nakayama et al., 2019), (Seggiani et al., 2017), (Liu et al., 2022), (Hu et al., 2022a), (Wojciechowska et al., 2011), (Yadav and Hakkarainen, 2022), (Mazzotta et al., 2022), (Seggiani et al., 2018), (Wang et al., 2018), (Mukai and Doi, 1995) |
| | | T ≥ 25°C | 45 | 15 | 2 | 2 | - | (Niu et al., 2021), (Chen et al., 2011), (Chen et al., 2020), (Tsuji and Suzuyoshi, 2002a), (Tsuji and Suzuyoshi, 2003), (Volova et al., 2010), (Volova et al., 2011), (Lopez-Llorca et al., 1994), (Ding et al., 2021), (Kim et al., 2021), (Yadav and Hakkarainen, 2022), (Imam et al., 1999), (Wang et al., 2021), (Sashiwa et al., 2018), (Zahir et al., 2021a), (Zahir et al., 2021b), (Nakayama et al., 2019), (Al-Salem, 2022), (Suzuki et al., 2017), (Tachibana et al., 2013), (Tran et al., 2020) |
| PARTIALLY BIOBASED | FW | $\begin{array}{l} T < 25^{\circ}\text{C} \\ T \geq 25^{\circ}\text{C} \end{array}$ | - | - 19 | 2 - | - - | - | (Olewnik-Kruszkowska et al., 2020) (Ikejima et al., 1999), (Koyama and Doi, 1996), |
| | SW | T < 25°C | 54 | - | - | - | 2 | (He et al., 2020) (Huang et al., 2020), (Delacuvellerie et al., 2021), (Briassoulis et al., 2019), (Nakayama et al., 2019), (Liu et al., 2022), (Huang et al., 2022), (Guzman-Sielicka et al., 2012), (Wang et al., 2019), (Hu et al., 2022b), (Hu et al., 2021), (Pauli et al., 2017) |
| | | $T \ge 25^{\circ}C$ | 19 | 9 | 5 | 4 | 1 | (Janik et al., 2018), (Chen et al., 2020), (Wang et al., 2019), (Lu et al., 2021), (Imam et al., 1999), (Sashiwa et al., 2018), (Zahir et al., 2021a), (Zahir et al., 2021b), (Nakayama et al., 2019), (Tran et al., 2020), (Tosin et al., 2012), (Briassoulis et al., 2020), (Accinelli et al., 2012) |
| FOSSIL BASED | FW | T < 25°C T > 25°C | 1 2 | - 5 | _ | - 4 | _ | (Heimowska et al., 2017) (Wang et al., 2021), (Marušincová et al., 2013), |
| | | - | | | | | | (Ikejima et al., 1999), (He et al., 2020), (Hoffmann et al. 2003) |
| | SW | T < 25°C | 35 | - | - | - | - | (Tsuji and Suzuyoshi, 2002b), (Nakayama et al., 2019), (Shaiju et al., 2020), (Liu et al., 2022), (Huang et al., 2022), (Huang et al., 2019), (Heimowska et al., 2017), (Krasowska et al., 2016), (Guzman-Sielicka et al., 2012), (Hu et al., 2022a), (Hu et al. 2021) |
| | | $T \ge 25^{\circ}C$ | 8 | 8 | 2 | - | - | (Tsuji and Suzuyoshi, 2002a), (Tsuji and Suzuyoshi, 2003), (Lu et al., 2018), (Wang et al., 2021), (Tachibana et al., 2013), (Zahir et al., 2021a), (Zahir et al., 2021b), (Nakayama et al., 2019), (Suzuki et al., 2017), (Tran et al., 2020) |

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Fig. 5 – Results of the biodegradability of bioplastics (biobased (a), partially-biobased (b), fossil based (c)) expressed as weight loss (1) and BOD (%) (2). Boxplots represent statistically relevant results (occurrence higher than 8 times) where the inner horizontal lines indicate the average values; dots represent values occurring less than 8 times.

prove to be more inhospitable for the microbial community than a freshwater environment (Chen et al., 2019).

Table 4 illustrates the duration of tests in days, both as range and average. Duration of the test was characterized by a wide variability with average values varying between 21 and 365 days. This reflects the lack of an unequivocal definition of biodegradability in an aquatic environment. As a results, the definition of test duration was, in some cases, based on maximum duration of the tests provided by standards, whilst in others on the definition of biodegradable material provided by composting standards: conversion of 90% of material into CO₂ within a time frame of six months. The average value obtained for each biodegradability indicator for each type of biopolymer was compared with average value of the same indicator obtained for the group to which the biopolymers belonged (biobased, partially-biobased, fossil based) to provide a qualitative assessment (Fig. 6). The polymers chosen were those for which a larger number of results had been obtained from the literature.

Biodegradable bioplastics purpose-designed as biodegradable (partially biobased and fossil-based bioplastics) showed a lower degree of biodegradation compared to average biodegradation rates observed for their respective category, this was particularly true for partially biobased bioplastics. Table 4 – Test duration in days for each of the five indicators (dash indicates that the indicator was not used in those tests). BB indicates "biobased", PBB indicates "partially biobased", FB indicates "fossil based".

| | | | WL | | BOD (%) | | BOD (mg/L) | | CO ₂ | | SE | |
|-----|----|----------------------|------|--------|---------|-------|------------|--------|-----------------|---------|------|--------|
| | | | mean | range | mean | range | mean | range | mean | range | mean | range |
| BB | FW | $T < 25^{\circ}C$ | 180 | 22–480 | - | - | 25 | 14–28 | - | - | - | - |
| | | $T \geq 25^\circ C$ | 74 | 15–365 | 28 | 21–30 | - | - | - | - | - | - |
| | SW | $T < 25^{\circ}C$ | 217 | 36–600 | - | - | - | - | 195 | 195 | 59 | 22–120 |
| | | $T \geq 25^{\circ}C$ | 193 | 30–365 | 31 | 25–82 | 30 | 30 | 280 | 200–360 | - | - |
| PBB | FW | $T < 25^{\circ}C$ | - | - | - | - | 28 | 28 | - | - | - | - |
| | | $T \geq 25^{\circ}C$ | - | - | 29 | 28–30 | - | - | - | - | - | - |
| | SW | $T < 25^{\circ}C$ | 259 | 42-440 | - | - | - | - | - | - | 365 | 365 |
| | | $T \geq 25^{\circ}C$ | 234 | 21–392 | 28 | 28 | 154 | 30–236 | 280 | 200–360 | 90 | 90 |
| FB | FW | $T < 25^{\circ}C$ | 294 | 294 | - | - | - | - | - | - | - | - |
| | | $T \geq 25^\circ C$ | 42 | 27–57 | 30 | 30 | - | - | 21 | 21 | - | - |
| | SW | $T < 25^{\circ}C$ | 104 | 35–440 | - | - | - | - | - | - | - | - |
| | | $T \geq 25^\circ C$ | 78 | 21–364 | 28 | 25–28 | 30 | 30 | - | - | - | - |



Fig. 6 – Qualitative representation of the biodegradability of specific biopolymers compared to the average of each of the three groups of bioplastics, considering the different indicators. Green dots indicate that polymer biodegradation rate exceeds the average obtained by group of origin; yellow dots indicate that polymer biodegradation rate is equal to the average obtained by group of origin; red dots indicate that polymer biodegradation rate is lower than the average obtained by group of origin.

Of all the biobased bioplastics, PHB was confirmed as the most readily biodegradable of its category both in FW and SW. Despite PLA being the most widely used and produced bioplastic, as mentioned previously, this bioplastic appears to be burdened by a higher number of criticalities.

With regard to partially biobased biodegradable bioplastics, literature results relating to tests conducted in salt water, indicated how starch-based bioplastics alone were capable of any significant level of biodegradation.

Among the fossil-based group, PVA demonstrated a higherthan-average degree of biodegradation (although only tests carried out in fresh water were detected in the literature), whilst PCL seemed to degrade adequately in saltwater, but insufficiently in freshwater and PBS was only tested in saltwater and the expected results were not yielded.

3. Conclusions

The findings of evaluations conducted on articles identified in our literature search confirmed the wide heterogeneity of methodologies used to verify the biodegradability of bioplastics.

The lack of a standard establishing parameters to be met in labelling a polymer as "biodegradable" has led to the utilization of various test methods with different test durations, temperatures, test medium composition, tested material forms and sizes to assess the degree of biodegradability. This has impeded comparison of the degree of biodegradability achieved by different bioplastics and complicated the definition of degree of biodegradability amongst similar categories of bioplastics.

Test methods endorsed by existing standards (for both fresh and salt water) fail to establish a value for parameters to be met to clearly define a substance as "biodegradable". Moreover, numerous biodegradation tests present in literature fail to follow these test methods, thus resulting in highly variable and at times discordant results.

The duration of test methods may significantly underestimate the time frame required for polymer biodegradation within natural ecosystems. In the available standards, the maximum duration of the test alone is indicated, thus leaving it up to the individual scientists to identify an appropriate duration. Test duration, temperature, and initial shape of the tested bioplastic substantially influence the results obtained in biodegradation tests. It is therefore crucial that test methods specifically define all these parameters to enable the obtaining of comparable results.

According to the published norms, to facilitate biodegradation, tested materials should be reduced in size, or powdered prior to being immersed in the test medium; however, no specifications of the final millimetres or centimetres to be achieved are provided. Indeed, as plastic goods are usually integral on arrival in an aquatic environment, the shredding process alters the true fate of the product. The "OK Biodegradable MARINE" and "OK Biodegradable WATER" marks of conformity established by Vinçotte, the only marks certifying biodegradability of a product in water, refer to ISO and ASTM standards which report ambiguity relating to size.

Various standards set test temperature in the range of 20– 25°C, which is frequently not a temperature range representative of a natural aquatic environment (sea/ocean water temperature is usually below 13°C). Although test temperature was used to aggregate the data, a wide variability of results has been found in the literature; moreover, the fact that they have been expressed largely as weight loss (without specifying initial size) hampers identification of the effective degree of biodegradability of the bioplastics.

In an appropriate standard relating to the biodegradability of bioplastics in an aquatic environment, the authors suggest including ecotoxic analysis of by-products in the presence of incomplete biodegradation.

Although bioplastics are largely perceived, particularly by the population, to be a biodegradable material, the large amount of biodegradable biopolymers on the market are characterized by a widely varied degree of actual biodegradability. PHB appear to represent the biobased bioplastics that can be most readily biodegraded under a series of different conditions (temperature, size, different degradation environments), whilst starch based bioplastics are the most biodegradable of the partially biobased group, and PVA and PGM of the fossilbased bioplastics. These conclusions were reached following careful analysis of literature data obtained, although a considerable lack of homogeneity in testing these materials was highlighted.

Declaration of Competing Interest

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

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