FINAL REPORT

BENEFITS AND CHALLENGES
OF BIO- AND OXO-DEGRADABLE PLASTICS

A COMPARATIVE LITERATURE STUDY

STUDY DSL-1

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TABLE OF CONTENTS

1. IDENTIFICATION OF THE STUDY

1.1. GENERAL INFORMATION

Title: Benefits and challenges of bio- and oxo-degradable plastics –
                 A comparative literature study

Study code: DSL-1

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O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 5 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

1.2. BACKGROUND AND OBJECTIVES

The inappropriate disposal of plastic waste represents a serious environmental issue that is today receiving increased attention. Incorrectly discharged plastic items tend to accumulate in nature, leaving behind an undesirable visual footprint and a potential risk to wildlife.

In an attempt to provide solutions (bio)degradable plastics have seen the light. They have been developed to solve specific problems related to the separate collection and treatment of food waste and not to provide a "short cut" to solve a bad behaviour like littering. To provide such a solution, those plastics should not only fragment into small pieces which are no longer visible to the naked eye, but should also be entirely consumed by bio-assimilation or conversion to CO_2 and H_2O. Since the introduction of "oxo-degradable" plastics, however, questions have arisen as to whether these plastics (bio)degrade according to the defined standards.

Two categories of plastics claim to be fully (bio)degradable: "biodegradable plastics" and "oxo-degradable plastics". The objective of this study is to review existing literature to evaluate the differences, advantages and disadvantages of "biodegradable plastics" versus "oxo-degradable plastics" for the purpose of creating a balanced scientific review of their properties.

To prevent confusion, the term "biodegradable plastics" is used to represent plastics which are ultimately biodegradable (as defined by EN 13432). The term "oxo-degradable plastics" is used for plastics which follow the degradation mechanism of "oxo-degradation" (as defined by CEN/TR 15351). However, it must be noted that the latter term is not standardized, only the process is standardized. More information on definitions is provided in Chapter 2.

Furthermore, it must also be noted that this study focusses mainly on "oxo-degradable plastics", or plastics containing inorganic additives that should cause the plastic to degrade by a process initiated by oxygen (see Chapter 6), and not on "enzyme-mediated degradable additives", or plastics containing organic additives that are claimed to be consumed by micro-organisms during which these excrete acids and enzymes that should break down the plastic (see Chapter 6.4.2).

2. DEFINITIONS

The term "biodegradable plastics" represents plastics which are ultimately biodegradable, as defined by the harmonized EU norm EN 13432. In other words, "biodegradable plastics" are plastics which degradation mechanism is characterized by the breakdown of the organic chemical compound by micro-organisms in the presence of oxygen to carbon dioxide, water and mineral salts of any other element present (mineralization) and new biomass or in the absence of oxygen to carbon dioxide, methane, mineral salts and new biomass.

The term "oxo-degradable plastics" was chosen to represent those plastics that follow the degradation mechanism of "oxo-degradation" as specified in CEN/TR 15351:

"Oxo-degradation": Degradation identified as resulting from oxidative cleavage of macromolecules

CEN/TR 15351 however only defines processes, not materials, which means that definition for "oxo-degradable plastics" is not provided. Nevertheless, for the purpose of this study, and for reasons of good readability, the term "oxo-degradable plastics" is used.

The term "oxo-degradable plastics" was actually introduced by the industry and more particularly by the "oxo-degradable plastics" industry. The term "oxo-degradable plastics" was introduced by several "oxo-degradable" additive producers to identify, advertise and sell their products. Besides the term "oxo-degradable plastics", the "oxo-degradable plastics" industry also introduced the term "hydro-degradable plastics", or "biodegradable plastics", to clarify that there is a difference between "oxo-degradable plastics" and "biodegradable plastics". In other words, the term "oxo-degradable plastics" is being used for commercial reasons, but is not (yet) standardized subject to criticism and not unanimously utilized or accepted in the industry.

3. LIST OF ABBREVIATIONS

ABA	Australasian Bioplastics Association
ACCC	Australian Competition and Consumer Commission
AD	Anaerobic digestion
ADP	Adenosine di-phosphate
AFNOR	*Association Française de Normalisation*, French normalization association
AMC	Advanced Materials Center
APR	Association of Postconsumer Plastic Recyclers
As	Arsenic
AS	Australian Standard
ASTM	American Society for Testing and Materials
ATP	Adenosine tri-phosphate
B-2-B	Business to business
B-2-C	Business to consumer
BBP	Belgian BioPackaging
BCPN	*Belangenvereniging Composteerbare Producten Nederland*, Dutch compostable products association
BNQ	*Bureau de normalization du Québec*, Normalization bureau of Québec
BPI	Biodegradable Products Institute
BPS	Biodegradable Plastics Society
BS(I)	British Standards (Institution)
C	Carbon
$CaCO_3$	Calcium carbonate
Cd	Cadmium

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 8 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Ce	Cerium
CEN	*Comité Européen de Normalisation,* European Committee on Normalization
CH_4	Methane
CIC	*Consorzio Italiano Compostatori*, Association of Compost Producers
Co	Cobalt
CO_2	Carbon dioxide
Cr	Chromium
CRU	Chico Research Foundation
Cu	Copper
DIN	*Deutsches Institut für Normung*, German Standardization Institute
ECAS	Emirates Conformity Assessment Scheme
EN	European Norm
EPA	Environmental Protection Agency
EPS	Expanded polystyrene
ESMA	Emirates Authority for Standardization and Metrology
EU	European Union
F	Fluorine
Fe	Iron
FTC	Federal Trade Commission
GEN	Global Ecolabeling Network
GMO	Genetically modified organism
H_2O	Water
HDPE	High Density Polyethylene
Hg	Mercury

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 9 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

IBAW	International Biodegradable Polymers Association and Working Groups
ISO	International Standards Organization
JBPA	Japanese BioPlastics Association
JIS	Japanese Institute for Standardization
LCA	Life Cycle Analysis
LDPE	Low Density Polyethylene
LLDPE	Linear Low Density Polyethylene
Mn	Manganese
Mo	Molybdenum
MODA	Microbial Oxidative Degradation Analyzer
MoEV	Ministry of Environment and Water
NERDC	Northeast Recycling Council
NF	*Norme Française*, French Norm
Ni	Nickel
NTSQP	China National Centre for Quality Supervision and Testing of Plastic Products
O_2	Oxygen
OECD	Organization of Economic Co-operation and Development
OPA	Oxo-biodegradable Plastics Association
PAH	Polycyclic Aromatic Hydrocarbon
Pb	Lead
PBAST	Polybutylene adipate co-succinate co-terephthalate
PBAT	Polybutylene adipate terephthalate
PBS	Polybutylene succinate
PBSA	Polybutylene succinate adipate

O.W.S.

Code: DSL-1

Page: 10 / 118

Date: Aug-09-2013

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

PCB	Polychlorinated biphenyl
PCL	Polycaprolactone
PE	Polyethylene
PET	Polyethyllene terephthalate
PHA	Polyhydroxyalkanoate
PHB	Polyhydroxybutyrate
PHBH	Poly-3-hydroxybutyrate-co-3-hydroxyhexanoate
PHBV	Polyhydroxybutyrate valerate
PLA	Polylactic acid
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinylchloride
PVOH	Polyvinyl alcohol
REA	Renewable Energy Association (Organics Recycling Group)
SBC	Sustainable Biomaterials Collaborative
Se	Selenium
SERDC	Southeast Recycling Development Council
SGLS	Singapore Green Labeling Scheme
SPI	Society of the Plastics Industry
TDPA	Totally Degradable Plastic Additives
TPS	Thermoplastic starch
UAE	United Arab Emirates
UNI	*Ente Nazionale Italiano di Unificazione*, National Normalization Institute of Italy

US(A)   United States (of America)

USCC   US Composting Council

UV   Ultraviolet

Zn   Zinc

4. EXECUTIVE SUMMARY

The unmanaged disposal of plastic waste is a mounting environmental issue. Conventional non-(bio)degradable plastics, when unmanaged, are accumulating in nature, leaving behind an undesirable visual footprint.

It is against this background that (bio)degradable plastics started to appear on the market and can, taken into account their end-of-life options, reduce both visual pollution and accumulation in nature.

Currently, two major groups of (bio)degradable plastics exist. "Biodegradable plastics" cover polymers like polyesters from fossil and renewable raw materials, potentially also in combination with starch and cellulose, polyhydroxyalkanoates and others like PLA which degrade in one or more environments, depending on the conditions. The second group uses non-biodegradable conventional polymers and blends in one or more additives which would make the polymer biodegradable if exposed to oxygen, heat and/or light. These additives can be of inorganic nature ("oxo-degradable") or of organic nature ("enzyme-mediated degradable").

The purpose of this study is to evaluate the differences, advantages and disadvantages of "oxo-degradable plastics", at this moment the most widely spread type of additivated plastics amongst this family, versus "biodegradable plastics" for the purpose of creating a uniform and scientifically correct communication.

For reasons of good readability, the terms "biodegradable plastics" and "oxo-degradable plastics" are being used to represent respectively plastics which are ultimately biodegradable, mineralizing into carbon dioxide, water and new biomass, as defined by the harmonized EU norm EN 13432, and plastics that follow the degradation mechanism of "oxo-degradation", characterized by oxidative cleavage of macromolecules, as defined by CEN/TR 15351. However, it must be noted that the term "oxo-degradation" is not standardized, only the process is.

The first chapter of the study deals with **biodegradable plastics**, or plastics for which degradation is caused by biological activity, more in particular enzymatic, microbial and/or fungal activity. The first biodegradable plastics entered the market in the late 80s, and since then, the market share increased relatively fast, although still representing only a tiny share of the total plastic market.

Based on the raw materials used, biodegradable plastics can be divided into 5 different categories: plastics based on starch, cellulose based plastics, biodegradable plastics obtained via chemical synthesis, biodegradable plastics produced by bacteria and biodegradable plastics of petrochemical origin. Each of these categories has their benefits and challenges which are explained more in detail in this study.

The majority of the biodegradable plastics are compostable, a definition which has been laid down in several standards and norms from which in Europe EN 13432 can be considered as the most important norm due to its harmonized and binding character. The counterparts of EN 13432 are ASTM D 6400 and ASTM D 6868 (US), AS 4736 (Australia) and ISO 17088 and ISO 18606 (worldwide).

In order for a plastic to be categorized as compostable, four criteria must be fulfilled:

1. <u>Chemical characteristics</u>: The product must contain at least 50% organic matter and may not exceed several heavy metal limits.

2. <u>Biodegradation</u>: The products should biodegrade for at least 90% within 6 months under controlled composting conditions. Biodegradation, or mineralization, is defined as the conversion of the organic C to CO_2.

3. <u>Disintegration</u>: The product, under the form which enters the market, should, within a timeframe of 12 weeks, fragment sufficiently to visually undetectable components (< 2 mm) under controlled composting conditions.

4. <u>Ecotoxicity</u>: The compost obtained at the end of the composting trial, eventually containing undegraded residuals from the product, should not pose any negative effects to the germination and growth of plants (and also earthworms in case of AS 4736).

Following the above, it can be noted that compostability comprises (much) more than just biodegradability. A product that is compostable is always biodegradable, but a product that is biodegradable is not per se compostable.

EN 13432, and its counterparts, are, however, only applicable for industrial composting, leaving an open space with regard to standardization for home compostability and biodegradation in other environments, like soil, fresh water, marine water and anaerobic digestion.

Objective proof of (bio)degradation (and compostability) of biodegradable plastics is available in different forms. The most robust evidence are the many certificates. Certified materials have been tested according to well defined and recognized test methods and fulfil the requirements of internationally accepted standards and norms like EN 13432. Furthermore, both the laboratory, responsible for the testing, as well as the certification bureau, responsible for the review of the test results, need to be accredited and/or independent.

Besides certification, further proof of (bio)degradation of biodegradable plastics can be found in round robin testing and of course the many publically available scientific articles. In general, it can be concluded that all biodegradable plastics biodegrade completely under industrial composting conditions, a smaller group also biodegrades under home composting conditions and in soil and an even smaller group also in fresh and marine water or even under anaerobic conditions.

Conventional plastics enriched with additives are discussed in detail in the second chapter of this study. The majority of these additivated plastics are **oxo-degradable plastics**. These conventional plastics are enriched with inorganic metal salts that should cause the plastic to degrade by a process initiated by oxygen and accelerated by light and/or heat. A smaller share however uses organic additives which are claimed to be consumed by the micro-organisms during which these excrete acids and enzymes that should break down the plastic into materials that are easily consumed by microbes. This latter group of additivated plastics is called "enzyme-mediated degradable plastics" and is only discussed briefly in this study as the focus lays on the oxo-degradable plastics.

For many years, the US guideline ASTM D6954 was the only guide available for testing oxo-degradable plastics. However, since 2009, several other guides and standards were developed in Europe and the Middle East: XP T54 980 and AC T51-808 (France), UAE.S 5009 (United Arab Emirates), BS 8472 (UK), SPCR 141 (Sweden) and JS 2004 (Jordan).

The majority of these guides and standards are composed of three so called 'Tiers':

1. Abiotic degradation (Tier 1): Using either accelerated or real-time conditions, samples are subjected to a combination of oxygen, heat and/or light to reduce the molecular weight and/or mechanical properties.

2. Biotic degradation (Tier 2): The residues from Tier 1 are retrieved for biodegradation testing using the environment in which the material is intended to end up after disposal (e.g. compost, soil, water, landfill,…). In most cases the amount and rate of CO_2 production, in case of aerobic biodegradation, and additionally CH_4 production, in case of anaerobic biodegradation, is measured.

3. Ecotoxicity (Tier 3): By using a variety of living organisms, including plants, earthworms and aquatic organisms, the effect of the residues from Tier 2 on the growth, survival and/or immobilization of fauna and flora can be determined.

However, a distinction needs to be made between guidelines and standards. Guidelines like ASTM D 6954 and BS 8472 are comparable with test methods and merely prescribe how the different tests need to be performed. Standards however, like UAE.S 5009 and SPCR 141, also contain specific pass or fail criteria and accompanying timescales in which these criteria need to be met. Consequently, it is not possible to claim conformity with guidelines, only with standards.

Since the introduction of the oxo-degradable plastics however, a lot of questions arose on whether these biodegradable variations of the well-known conventional plastics are indeed really biodegradable. On exposure to oxygen, heat and/or light, these polymers are claimed to disintegrate into small fragments, thereby reducing the visual footprint, however, these fragments can accumulate in the environment if biodegradation happens only partially or not at all.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

A significant part of this study therefore focusses on the available data on the (bio)degradation of oxo-degradable plastics.

According to the oxo-degradable industry, the most likely disposal routes for oxo-degradable plastics are recycling, soil surface exposure (through littering and the use of mulching films) and landfilling. Water exposure seems to be a possible disposal route as a result of littering.

The Oxo-biodegradable Plastics Association (OPA) furthermore states that oxo-degradable plastics are not marketed for composting. Unlike biodegradable plastics, despite what is sometimes claimed, oxo-degradable plastics are not compostable, as agreed upon by an important share of the industry. Oxo-degradable plastics do not meet the requirements of (industrial and/or home) compostability set out in the different standards, which means that they cannot be used for e.g. compostable food service applications.

However, according to the oxo-degradable industry, the oxo-degradable plastics do biodegrade completely, but not in the foreseen timeframe as set forth in the different standards on (industrial and home) composting. Numerous scientific articles are available on the biodegradation of oxo-degradable plastics, from which two articles showed a significant level of biodegradation (see Table 1).

Table 1. Most promising biodegradation results for oxo-degradable plastics found in literature

Author(s)	Abiotic degradation	Biotic degradation	
		Compost	Soil
Jackubowicz et al.	10 days at 65°C	43% after 607 days	91% after 733 days
Chiellini & Corti	44 days at 55°C	49-63% after 600 days	28% after 430 days

It must however be noted that the above two articles are the only articles with a considerable percentage of biodegradation. All other articles reported no or only a (very) low level of biodegradation. It can therefore be concluded that the rate and level of biodegradation of oxo-degradable plastics is at least questionable and irreproducible.

The oxo-degradable plastics industry however questions the reliability of the internationally recognized test methods for the determination of the biodegradation. More specifically, it is claimed that the CO_2 production is not the correct parameter for the determination of the mineralization level of oxo-degradable products as oxo-degradable plastics biodegrade at a (much) lower rate. Yet, many natural lignin containing materials are also characterized by a slow mineralization rate and have been tested successfully for biodegradation using these internationally recognized test methods. On other words, these test methods do result in reliable data on biodegradation, even on (very) long term.

In addition to the lower CO_2 production rate, oxo-degradable plastics are also claimed to have a lower tendency to get converted to CO_2 (and a higher tendency to get converted into biomass). However, evidence of such a higher degree of bio-assimilation has not yet been generated.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 16 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

As the conversion of organic carbon to CO_2 is not recognized by the oxo-degradable plastics industry as the optimal test method, many authors used other parameters to quantify the biodegradation, like microbial colonization, biofilm formation and ADP/ATP ratio. Also the change in physical properties and especially the decrease in molecular weight is often used as proof of biodegradation

Chiellini et al. tested the biodegradation of a thermally treated additivated LDPE and obtained a plateau at 42-48% after 100 days in fresh water for the low molecular weight extracts with a molecular weight of about 1,000 Dalton. The complete material, including the low molecular weight extracts, showed a mean molecular weight of approximately 4,500 - 5,000 Dalton and only reached a biodegradation level of 12% after 100 days. Molecular weight extracts between 7,500 and 10,000 Dalton showed no significant biodegradation. In other words, the molecular weight of a plastic decreases over time when exposed to oxygen, heat and light, but there is no proof that this continues to levels which result in complete biodegradation.

Even if molecular weight would continue to decrease over time, this only happens under very specific conditions. The majority of the authors used temperatures ranging from 55°C to 70°C. In addition, these (very) high temperatures were also maintained for relatively long periods, ranging from 44 days (at 55°C) to 80 days (at 70°C). Time-temperature superposition principles have been established in the last years as a methodology to translate these accelerated conditions to real-life conditions and the most referenced principle is the Arrhenius principle. However, Celina et al. proved that there is no guarantee that the overall behaviour is of an Arrhenius form. Furthermore, it must be noted this principle is based on the assumption that there is a linear correlation between molecular weight and temperature over a wide range of temperature and not only over a smaller partial range, which cannot be guaranteed. Evidence of the decrease of molecular weight of oxo-degradable plastics at ambient temperature is not available.

The third and last chapter deals with **certification**. An overview of the different certification opportunities for both oxo- and biodegradable plastics are shown in Table 2.

Compostability can be certified by several certification bodies worldwide. Within Europe, the seedling logo from European Bioplastics and the OK Compost logo from the Belgian certification body Vinçotte are the most important certificates and logos. On a business-to-business level they are both equally well known, whereas on a business-to-consumer level each has geographical preferences. There do exist some other (national) certificates and logos in Europe, but these are less dominant.

Certification for home compostability, and certainly for biodegradation in other environments, is less developed. The OK Compost Home logo from Vinçotte, representing home compostability, is today the most dominant logo on a worldwide scale, although similar other certification opportunities exist as well. With regard to biodegradation in other environments, only Vinçotte and the Swedish Technical Research Institute SP have created some certification systems.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Table 2. Overview of the available certification opportunities for biodegradable plastics

Environment	Certification body
Industrial compostability	European Bioplastics (Europe) Vinçotte (Belgium) DIN CERTCO (Germany) Biodegradable Products Institute (USA) Cedar Grove (USA) Japanese BioPlastics Association (Japan) Australasian BioPlastics Association (Australia & New Zealand) Consorzio Italiano Compostatori (Italy) SP Technical Research Institute (Sweden) Catalonian government (Catalonia, Spain)
Home compostability	Vinçotte (Belgium) DIN CERTCO (Germany) Australasian BioPlastics Association (Australia & New Zealand) Organics Recycling Group – Renewable Energy Association (UK)
Biodegradability in other environments	Vinçotte (Belgium) SP Technical Research Institute (Sweden)
Oxo-degradation	Emirates Authority for Standardization and Metrology (UAE) Oxo-biodegradable Plastics Association (UK) SP Technical Research Institute (Sweden) Biosystems America (USA) Singapore Environment Council (Singapore)

Many biodegradable plastics are certified for industrial and/or home compostability, while only a few are certified for biodegradability in soil and/or fresh water. Oxo-degradable plastics are not certified for industrial or home compostability, nor for biodegradability in soil or fresh water.

Nevertheless, several associations and institutes have created certification systems and accompanying logos for oxo-degradable plastics based on the above discussed guidelines and standards. In this context, several oxo-degradable plastics are certified by the Emirates Authority for Standardization and metrology (ESMA) conform UAE.S 5009. In other words, this means that these products were tested by an independent and accredited laboratory and fulfilled the criteria of UAE.S 5009 (molecular weight level of 5,000 Dalton or lower within 4 weeks and a biodegradation value of at least 60% within 6 months). Yet, the most promising results found in literature, i.e. those obtained by Jackubowicz showing 91% biodegradation in soil after 2 years, only reported a molecular weight value of 8,800 and a biodegradation percentage of only 5% after 6 months. Also Chiellini & Corti, who reached biodegradation levels of > 60% in 1.6 years, only reached 4-7% biodegradation within 6 months.

The final conclusion of this study can be summarized as follows:

1. Biodegradable plastics:
   - The majority do meet the requirements of industrial composting standards, while others biodegrade in other environment as well;
   - Solid proof of biodegradation is available through certification by accredited laboratories;
   - Based on raw materials used, 5 categories can be distinguished: starch based, cellulose based, chemically synthesized, produced by bacteria and fossil based;
   - Standards on specifications are well established for industrial composting, but less developed for home composting and biodegradation in other environments.
   - Biodegradation takes place through biological activity, more in particular enzymatic, microbial and/or fungal activity;

2. Oxo-degradable plastics:
   - Oxo-degradable plastics do not meet the requirements of industrial and/or home compostability set out in different standards (Oxo-biodegradable Plastics Association states that they are not marketed for composting);
   - Conventional plastics additivated with transition metal salts;
   - Since 2009 strong increase in number of standards and guides, although no consistency in content and pass levels (if available);
   - (Bio)degradation claimed to be initiated by oxygen (but inhibited by moisture) and accelerated by UV light and/or heat;
   - Very few positive biodegradation results obtained (those could not be repeated under the same conditions, not by the same author, not by other authors);
   - No proof of Arrhenius' time-temperature superposition principle at wide range of temperature, which makes extrapolation from abiotic degradation at elevated temperature to real-life conditions scientifically incorrect;
   - Alternative methods (carbonyl index, molecular weight, microbial growth, ADP/ATP,…) no proof of complete biodegradation, only proof of biological activity;
   - Not compostable: better term 'thermo- or photo-fragmentable plastics'?
   - Different certification institutes, but not always independent or transparent.
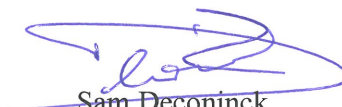
3. Enzyme-mediated degradable plastics:
   - Very few data available on biodegradation
   - Conventional plastics additivated with organic additives;
   - (Bio)degradation claimed to take place through enzymes excreted by micro-organisms during the consumption of the additive;

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5. BIODEGRADABLE PLASTICS

5.1. Introduction

Biodegradable plastics can, based on the raw materials used during production, be divided into 5 different categories:
- Starch based biodegradable plastics
- Cellulose based biodegradable plastics
- Biodegradable plastics obtained via chemical synthesis
- Biodegradable plastics produced by bacteria
- Biodegradable plastics of petrochemical origin

It must be noted that biodegradable plastics which are made from renewable raw materials are not automatically biodegradable, while on the other hand biodegradable plastics are not necessarily made from renewable raw materials.

The majority of the certified biodegradable plastics are currently produced from renewable raw materials. However, there are also fossil based biodegradable plastics that do fulfil the requirements as set forth by the different norms on compostability. Finally, also a combination of both renewable and non-renewable raw materials can eventually lead to a biodegradable plastic.

5.2. Degradation mechanism

Several factors can lead to the degradation of biodegradable plastics. These include light, heat, biological activity, mechanical impact, etc. This chapter deals with biodegradation only, which is the degradation caused by biological activity, more in particular enzymatic, microbial and/or fungal activity. For most biodegradable plastics, biodegradation is a single step process, requiring only biological activity. Only a few biodegradable plastics follow a 2-step degradation profile, where for instance heat is responsible for initiating the degradation process. Whatever the degradation mechanism which applies, biodegradation continues once initiated and is claimed to result in complete biodegradation.

A major distinction is made between biodegradation taking place under conditions where oxygen (O_2) is available, so-called aerobic biodegradation, and conditions where oxygen is not available, so-called anaerobic biodegradation. In the case of aerobic biodegradation, the process is a basic biological process in which organic, carbon-based matter is oxidized leading to conversion of carbon (C) to carbon dioxide (CO_2). This is equivalent to respiration and can considered to be the opposite of photosynthesis. Oxygen is consumed through which carbon of the sample is converted into carbon dioxide and water (H_2O). Some carbon can remain as residual sample or in metabolites, representing the total residual carbon. Some carbon is used to produce new biomass, representing the biomass yield.

$$C_{sample} + O_2 \quad \rightarrow \quad CO_2 + H_2O + C_{residual} + C_{biomass}$$

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

In anaerobic biodegradation, no oxygen is consumed. In this case, the sample is converted into methane (CH_4) and carbon dioxide, residual sample or metabolites and biomass. Anaerobic conditions are created when oxygen is not present or when oxygen is consumed or depleted more rapidly than it is replaced (mostly by diffusion). Natural examples are bottoms of rivers, swamps and lakes and the rumen of herbivores. Man-made examples are landfills, septic tanks, anaerobic digesters for treatment of wastewater, sludge or solid waste.

$$C_{sample} \quad\quad \rightarrow \quad\quad CH_4 + CO_2 + C_{residual} + C_{biomass}$$

From these equations it can be seen that true biodegradation is based on CO_2 (and eventually CH_4) production or O_2 consumption. Other parameters such as visual disappearance, weight loss, decrease in molecular weight, disappearance of parent compound, decrease in mechanical characteristics such as tensile strength, tear resistance, etc. are only secondary effects and at best only demonstrate partial and incomplete biodegradation. They cannot be used to proof (full) biodegradability.

A second important aspect with regard to biodegradation, is the importance of the environment in which it is supposed to take place. Rate and degree of biodegradation are determined by various factors which can be different from one environment to the other. These factors include moisture content (going from aquatic to dry), oxygen availability (aerobic or anaerobic), temperature (high in industrial composting, ambient to low in soil and water), type of microbiology (bacteria and/or fungi and/or actinomycetes), density of microbiology (high in composting and in wastewater treatment, low in soil and marine), salt concentration, etc. For biodegradable plastics, mainly the temperature and the presence or absence of fungi have a big impact on the degree of biodegradation.

The other factors mainly have an influence on the rate of biodegradation. As a consequence, biodegradable plastics can be biodegradable in one environment, but not in the other. Therefore it can be concluded that a biodegradability claim without specifying an environment in which it is supposed to take place is an incomplete or even useless claim.

A ranking of major environmental niches with regard to aggressiveness of biodegradation is given in Figure 1.
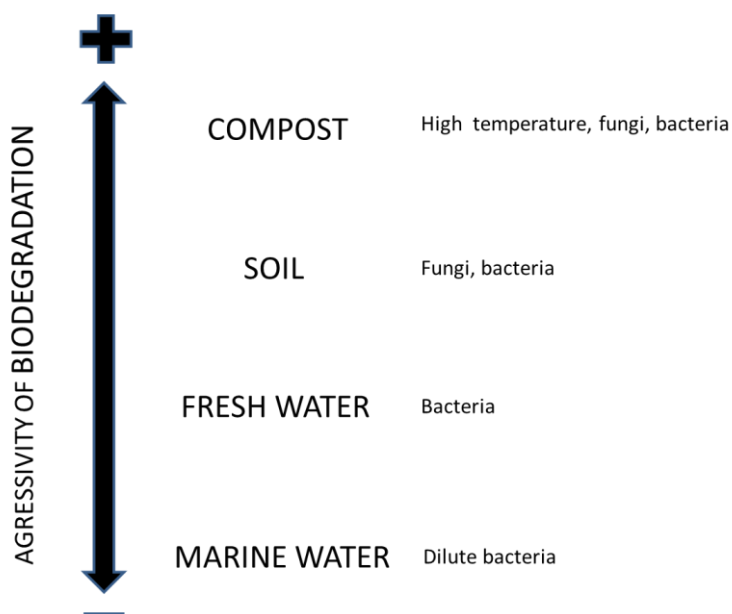
BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Figure 1. Relation between environment and aggressivity of biodegradation

5.3. Standards and legislation

It must be noted that the (bio)degradability of a product can be very different from one environment to another (compost, soil, water, landfill, etc.). For this reason specific biodegradation tests are needed and have been designed for each environmental compartment.

5.3.1. Industrial Compostability

The definition of (industrial) compostability is laid down in several standards and norms, which were developed by the International Standards Organization (ISO), the European Committee on Normalization (CEN), the American Society for Testing and Materials (ASTM), the German Institute on Normalisation (DIN), the Japanese Institute for Standardization (JIS) and Standards Australia (AS).

Currently, Europe has two key norms on industrial compostability: EN 13432 (2000): "*Packaging - Requirements for packaging recoverable through composting and Biodegradation - Test scheme and evaluation criteria for the final acceptance of packaging*" and EN 14995 (2007) "*Plastics - Evaluation of compostability - Test scheme and specifications*". Since EN 14995 can only be used for plastic materials and EN 13432 is applicable for packaging in general, EN 13432 is considered as the most important norm on industrial compostability in Europe. Secondly, and more importantly, EN 13432 has been published under mandate M 200 rev.3 given to CEN by the European Commission and the European Free Trade Association to support the European Council and Parliament Directive on Packaging and Packaging Waste [94/62/EC] and has since consequently been adopted by all European Member States. It has also been accepted by the European Commission, giving it the status of a harmonized EU norm (Official Journal of the European Communities L 190, July 12th, 2001[1]).

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 22 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

The main counterparts of EN 13432 and EN 14995 are

- ASTM D 6400-12 *"Standard specification for labeling of plastics designed to be aerobically composted in municipal or industrial facilities"*
- ASTM D 6868-11 *"Standard specification for labeling of end items that incorporate plastics and polymers as coatings or additives with paper and other substrates designed to be aerobically composted in municipal or industrial facilities"*
- AS 4736-2006 *"Biodegradable plastics – Biodegradable plastics suitable for composting and other microbial treatment"*
- ISO 17088:2012 *"Specification for compostable plastics"*
- ISO 18606:2013 *"Packaging and the environment – Organic recycling"*

The most important difference between these norms is the geographical value. While EN 13432 is mainly referred to in Europe, ASTM D 6400 and ASTM D 6868 mainly in the United States and AS 4736 in Australia, ISO 17088 and ISO 18606 have a global character.

In spite of some minor differences between these norms, the essence is identical and consists of four criteria which must be fulfilled in order to call a product compostable.

1. Chemical characteristics: The product must contain at least 50% organic matter and may not exceed the heavy metal limits as specified in Table 3.

2. Biodegradation: The products should biodegrade for at least 90% within 6 months under controlled composting conditions. Biodegradation, or mineralization, is defined as the conversion of the organic C to CO_2 and can considered to be the degradation on a chemical level. Consequently, this characteristic is linked to the chemical composition of the sample.

3. Disintegration: Unlike the other criteria, the third requirement refers to the physical form of the product instead of to the chemical composition. The product, under the form which enters the market, should, within a timeframe of 12 weeks, fragment sufficiently to visually undetectable components (< 2 mm) under controlled composting conditions. Consequently, this characteristic is linked to the thickness and the physical construction (e.g. laminate, coating, etc.) of the sample.

4. Ecotoxicity: Finally, the compost obtained at the end of the composting trial, eventually containing undegraded residuals from the product, should not pose any negative effects to the germination and growth of plants (and also earthworms in case of AS 4736).

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

From these criteria it becomes obvious that compostability comprises more than just biodegradability. A packaging which is compostable is always biodegradable, while a packaging which is biodegradable is not always compostable (as it can be too thick for disintegration or can contain toxic components). Furthermore, while composting is an intentional end-of-life scenario, managed by the local or regional waste management associations via the separate collection of organic waste, biodegradation is an unintentional and uncontrolled process and can indirectly be linked to littering. Because of this reason, and in order to prevent littering, several authorities prefer (or even mandate) the use of the term "compostable" instead of "biodegradable".

Table 3. Heavy metal limits as prescribed by the different norms on compostability

Metal	Limit values (ppm on total solids)		
	Europe* EN 13432 (2000)	USA[**] ASTM D 6400-04	Canada BNQ P 9011-911-5
Zn	150	1400	463
Cu	50	750	189
Ni	25	210	45
Cd	0.5	19.5	5
Pb	50	150	125
Hg	0.5	8.5	1
Cr	50	-	265
Mo	1	-	5
Se	0.75	50	4
As	5	20.5	19
F	100	-	-
Co	-	-	38

* The heavy metals as prescribed by EN13432 are identical to the ones prescribed by AS 4736 applicable in Australia. ISO 17088 and ISO 18606 both refer to the national and/or regional regulations dealing with metals, other elements and toxic substances in the environment.

** Maximum levels for USA (according to ASTM D 6400 heavy metal content must be less than 50% of those prescribed for sludges or composts in the country where the product is sold).

Apart from AS 4736, which requires an additional earthworm toxicity test, but which, as a standard, is geographically limited to Australia only, it can be stated that EN 13432, ISO 17088 and ISO 18606 are more stringent when compared to ASTM D 6400 as the limit values for heavy metals are lower.

Looking specifically at biodegradation, EN 13432 is less strict when compared to ISO 17088 and ISO 18606 as organic constituents present at levels between 1% and 10% (on dry weight basis) do not need to be tested separately. In other words, EN 13432 leaves the choice between testing the individual components or the final product, while ISO 17088 and ISO 18606 require separate (biodegradation) testing for each component between 1% and 10%.

When compared to ASTM D 6868 however, ISO 17088 and ISO 18606 still allow more flexibility. While in ISO 17088 and ISO 18606 the determination of components between 1% and 10% is based on the final product, ASTM D 6868 requires this to be calculated on the coating alone.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 24 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

The requirements of ISO 17088 and ISO 18606 are completely identical. However, as ISO 17088 is only applicable for plastics and ISO 18606 for packaging in general, ISO 18606 can be considered as the most important norm on industrial compostability on a worldwide scale.

However, whereas ASTM and ISO norms have been developed by the industry and therefore have a voluntary character, EN 13432 has been endorsed by the European Commission and is therefore a harmonized EU standard with a higher juridical value and a more binding character when compared to its equivalents.

5.3.2. Home compostability

Unlike for industrial compostability, there are only two (inter)national norms in place which define the criteria for home compostability:

In April 2010, the National Normalization Institute of Italy (UNI, Ente Nazionale Italiano di Unificazione) published UNI 11355 "*Manufatti plastici biodegradabili in compostaggio domestic*" (Plastic items biodegradable in home composting). This norm is partially based on UNI 11183 "*Materie plastiche biodegradabili a temperature ambiente*" (Plastic materials biodegradable at room temperature) and prescribes criteria for heavy metals (similar to EN 13432), biodegradation (to be performed in compost, soil or water), disintegration (conform ISO 20200) and ecotoxicity (with both terrestrial and aquatic organisms).

Also in 2010, Standards Australia published AS 5810 "*Biodegradable plastics – Biodegradable plastics suitable for home composting*". Similar to UNI 11355, this standard also prescribes criteria for heavy metals (similar to AS 4736), biodegradation (to be performed in compost), disintegration (conform ISO 16929 or ISO 20200) and ecotoxicity (only with terrestrial organisms).

Despite the fact that the first norms on home compostability were only published in 2010, some certification bodies already started to use EN 13432 and some additional requirements several years earlier to determine whether a product can be categorized as home compostable or not.

In general, next to the difference in temperature, ambient or 28°C for home composting instead of elevated or 58°C for industrial composting, also the timeframe in which the criteria need to be met are different (longer) for all of the above mentioned norms and schemes, and this both for biodegradation and disintegration.

Furthermore, in July 2009, a Royal Decree became effective in Belgium that defines three properties of a product depending on its end-of-life management option, including, besides 'home compostable', also 'industrial compostable' and 'biodegradable' (the latter only for products with intended disposal in soil, such as agricultural mulching film). This decree determines the requirements and standards that have to be fulfilled to claim any of these terms and also prohibits the use of the term 'biodegradable' on packaging as this could lead to littering[2].

5.3.3. Biodegradation

5.3.3.1. *Soil biodegradability*

Up to today, only two (inter)national standards exist defining specifications for soil biodegradability.

In 2005, the French Normalization Association AFNOR (Association Française de Normalisation), published NF U 52-001 *"Biodegradable materials for use in agriculture and horticulture - Mulching products".* This standard prescribes criteria for heavy metals (similar to EN 13432), organic substances (including, but not limited to, carbamates, organic amines and phthalates), biodegradation (in compost, soil and water) and ecotoxicity (with both terrestrial and aquatic organisms). It is however unclear in how far this French standard is widely used.

More recently, in October 2012, the National Normalization Institute of Italy (UNI, Ente Nazionale Italiano di Unificazione) published UNI 11462 "*Materiali plastici biodegradabili in suolo*". This standard prescribes criteria for heavy metals (similar to UNI 11183 and EN 13432), biodegradation (to be performed in soil conform ISO 17556) and ecotoxicity (with both terrestrial and aquatic organisms).

Furthermore, during several years CEN has made a big effort to develop a European standard but a consensus could not be reached. The main reason was a disagreement on the possibility for a weathering pre-treatment step in which polymers would first be exposed to light, temperature and moisture.

In general, for soil, it can be noted that disintegration is largely determined by the intended use. For some applications, the plastic product should preferably disintegrate after a few months only while for other applications the product should much longer remain intact. Consequently, many experts share the opinion that disintegration requirements should not be included in a soil standard.

5.3.3.2. *Aquatic biodegradability*

Historically, the first standards with specifications on biodegradability were published by OECD. Chemicals can claim ready biodegradability when 60%/70% biodegradation (% determined by the parameter being followed) is reached within 28 days and also a 10% to 60% increase is seen within a period of maximum 10 days. Pre-exposure of inoculum is not allowed.

Inherent biodegradability does not include a maximum test duration. Primary inherent biodegradability is defined as reaching more than 20% biodegradation. Ultimate inherent biodegradability is defined as reaching 70% biodegradation. Pre-exposure and pre-adaptation are allowed.

At CEN level, EN 14987 *'Plastics - Evaluation of disposability in waste water treatment plants - Test scheme for final acceptance and specifications'* was published in 2006. Pass level for biodegradation is 90% which must be reached within a maximum of 56 days. Aquatic biodegradation tests must be used, with only sludge as inoculum source, test temperature must be ambient. Specifications for water solubility and dispersability are included as well. On the other hand, requirements for chemical analyses or ecotoxicity are not defined.

A higher need for additional specifications on water degradability can be expected when more biodegradable flushable products are developed and put onto the market.

5.3.3.3. *Marine biodegradability*

Although biodegradable plastics could have many applications for marine conditions, developments on standards have been limited to ASTM level with US Navy as a major driver. In 2005 ASTM D 7081 *'Standard Specification for Non-Floating Biodegradable Plastics in the Marine Environment'* was published. Besides full biodegradation in a composting test (ASTM D 5338), 30% biodegradation in a marine test must be achieved within a period of 6 months, disintegration in a marine environment must be at least 70% (smaller than 2 mm) within a period of 3 months while also an aquatic toxicity test is required.

5.3.3.4. *Anaerobic digestion*

Criteria for acceptance in anaerobic digestion are given as an option only in EN 13432 and described rather vague. Biodegradation under anaerobic conditions must be at least 50% within 2 months while disintegration must be at least 90% smaller than 2 mm within a period of 5 weeks combined anaerobic digestion and aerobic stabilization.

In other standard organizations, no norms exist with specifications for anaerobic digestion. Nevertheless, by several producers of biodegradable polymers this is felt as a need and it can be expected that in the next years (2013-2015) serious efforts will be undertaken to develop more specific and valuable standards on specifications for anaerobic digestion.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 27 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

5.4. Types of biodegradable plastics

As discussed under 5.1, based on the raw materials used, biodegradable plastics can be divided into 5 different categories:
- Starch based biodegradable plastics
- Cellulose based biodegradable plastics
- Biodegradable plastics obtained via chemical synthesis
- Biodegradable plastics produced by bacteria
- Biodegradable plastics of petrochemical origin

5.4.1. Starch based biodegradable plastics

Different biopolymers can be extracted directly from biomass. Among them the most important ones are the polysaccharides. Starch is a polysaccharide that can be extracted from various agricultural products or waste of food plants and can be modified in such a way that biodegradable plastics can be produced. Furthermore, these biodegradable plastics can be processed using the same equipment as for the conventional non-degradable plastics. The most important sources of starch are corn, grain (wheat), potato, tapioca and rice[3].

Starch can also be processed by extrusion into a half fabricate, known as thermoplastic starch (TPS).

Some producers: Biotec (Bioplast, from corn and potatoes), Limagrain (Biolice, from corn and flour), Novamont (Mater-Bi ®, from corn), Plantic (Plantic®, from corn), Rodenburg (Solanyl ®, from potato products)

Applications: Packaging (trays, nets), films (shopping bags, mulching film), catering (plates, cutlery, cups), foam (loose-fill protection), plant pots, etc.

End-of-life options: Industrial and home composting (the latter only for certain types), biodegradation in soil, physical recycling (theoretical) and anaerobic digestion (certainly possible for starch, but for the entire product it will depend on the used copolymer). It must be noted that for specific applications like mulching film, organic waste collection bags and body bags, the biodegradability is a significant added value.

Mixtures or blends of thermoplastic starch and biodegradable plastics from petrochemical origin are also often used as these blends reduce the water sensitivity and improve the flexibility of the products, making it possible to customize the material properties of the final product.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Table 4. Benefits and challenges of starch based biodegradable plastics

Benefits	Challenges
Potentially widely biodegradable (industrial and home composting, soil)	Possible competition with food crops
Renewable resources	Sensitivity to water (in case of TPS)
The same conversion processes as for conventional non-degradable plastics can be used	Elevated cost (2 to 3 times more expensive than conventional polymers)
Heat resistant (up to 110 ° C)	
Permeable	

5.4.2. Cellulose based biodegradable plastics

Cellulose is a polysaccharide which is the main component of the cell walls of plants. Consequently, the availability of cellulose is very high. Basically, three different types of biodegradable plastics can be made from cellulose: biodegradable plastics from natural cellulose fibres, biodegradable plastics from regenerated cellulose and biodegradable plastics from modified cellulose[4].

Regenerated cellulose, or cellophane, is a thin, transparent film obtained by temporarily modifying the cellulose during processing. The good properties of cellulose, high temperature stability and high strength, are maintained during the production of cellophane. In recent years, the cellophane production has decreased significantly due to the availability of alternative packaging materials and the use of carbon disulfide required for the production of cellophane[5].

Cellulose can also be chemically modified to thermoplastic processable cellulose. Known variants include cellulose acetate, propionate and butyrate. Yet, these variants do not always comply with the norms on compostability. Cellulose diacetate is however biodegradable and compostable[4].

Some producers: Daicel (cellulose acetate), Clarifoil (cellulose acetate), FKuR Kunststoff GmbH (Biograde®, cellulose in combination with petrochemical based biodegradable plastics), Innovia (Natureflex, cellophane).

Applications: Packaging (films, twist wraps), tape, envelope windows

End of Life options: Industrial and home composting and anaerobic digestion. Cellophane is also biodegradable in soil, water and marine conditions. For cellulose acetate recycling with paper and cardboard is also a possibility.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Table 5. Benefits and challenges of cellulose based biodegradable plastics

Benefits	Challenges
Widely biodegradable (industrial and home composting, biodegradable in soil, water and marine conditions in case of cellophane)	Use of carbon disulfide for the production of cellophane
Renewable resources (at least for 95%)	High energy and water consumption in the production of cellophane, resulting in a negative contribution to LCA
Heat resistant	

5.4.3. Biodegradable plastics from chemical synthesis

Biodegradable plastics can also be produced via chemical synthesis or the polymerization of (biobased) monomers. These types of biodegradable plastics are also called biopolyesters as they have many characteristics in common with conventional non-degradable polyesters. The most important biopolyester is polylactic acid (PLA), which is produced via the chemical synthesis of lactic acid. The lactic acid itself is derived from starch based sugar (dextrose) through bacterial fermentation. The starch needed to produce PLA can be extracted from agricultural products like potatoes, corn, wheat, etc., but also from waste products from the food industry such as molasse, whey, etc.

PLA can be processed to fibres and film, like most thermoplastics. PLA is often also blended with renewable and/or non-renewable materials to improve certain properties[6]. Currently, PLA comprises about 40% of the biodegradable plastics market, making it the most important and most widely used biodegradable plastic at this moment[7].

Some producers: Pyramid Bioplastics (Uhde Inventa-Fischer), Boehringer Ingelheim (RESOMER®), Futerro, PURAC (lactides), Musashino Chemical Laboratory (Biofront), NatureWorks (Ingeo™), Mitsui, Novamont.

Applications: Packaging (trays, bottles, films, shrink films), catering (cups for cold drinks), textiles (fibres), medicine (implants, screws, sewing equipment,…), electronics (cell phone, computer,…)

End of Life options: Industrial composting and anaerobic digestion if operating under thermophilic conditions (temperature >50-55°C), mechanical recycling (PLA- and PET-bottles can be removed separately from a mixed plastic waste stream) and chemical recycling.

Currently, the majority of the produced PLA is used for packaging. By 2020 however, it is expected that there will be a shift towards fibers[5].

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 30 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Table 6. Benefits and challenges of biodegradable plastics from chemical synthesis

Benefits	Challenges
Biodegradable (industrial composting and anaerobic digestion if operating under thermophilic conditions)	Not all types degrade at low temperature (< 50 - 55°C).
Renewable resources	Low heat resistance
Similar conversion processes as for conventional non-degradable plastics can be used	Gas barrier properties (e.g. difficult to hold carbonated drinks)

5.4.4. Biodegradable plastics produced by bacteria

Different types of bacteria (Alcaligenes spp., Pseudomonas spp., etc.) are able to naturally produce polyhydroxyalkanoate (PHA) if unfavourable environmental conditions occur. Unfavourable conditions mean the limited availability of essential elements (nutrients) like nitrogen, phosphorus, sulphur, oxygen and/or magnesium and the excess of carbon. In such conditions PHA is accumulated in the cell body as intracellular granules[8].

Since PHA possesses valuable properties such as thermoplastic workability and a high resistance to water, these bacteria can be used for the production of biodegradable plastics. Furthermore, PHA is also biodegradable under both aerobic and anaerobic conditions and can be produced from renewable raw materials.

The properties of PHA are determined by the PHA monomer composition or more specifically by the diet of the bacteria. It is thus possible to produce a wide range of biodegradable plastics with different material properties, from flexible to rigid to rubbery. Important examples of copolymers are polyhydroxybutyrate (PHB), polyhydroxybutyrate valerate (PHBV) and poly-3-hydroxybutyrate-co-3-hydroxyhexanoate (PHBH).

Besides the microbial production of PHA, different plants and yeasts are also capable of producing PHA. Unlike bacteria, genetic manipulation is in this case required to ensure that plants and yeasts produce PHA[4].

Polyhydroxybutyrate (PHB) is a biopolymer present in all living organisms. Different bacteria produce PHB in large quantities as a storage material replacing fat, oil or starch. PHB is with regard to molecular weight, stiffness, melting point and glass transition temperature characteristics comparable with conventional non-degradable plastics like polypropylene. Furthermore, PHB is also non-toxic and completely biodegradable.

New developments are aimed at introducing the PHA producing gen into plants so that PHA is produced by plants also besides by bacteria.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 31 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Some producers: Biologic Material Co. (PHBV), Biomer (PHB), Metabolix (Mirel[TM]), PHB Industrial S.A. (Biocycle®), Tianan Meridian (PHA).

Applications: Implants, electronics, ball pens, catering, fibres, films

End of Life options: Industrial and home composting, biodegradation in soil, water and the marine environment, anaerobic digestion and chemical recycling.

It must be noted that hybrids of PHA and PLA are also promising as in that case the disadvantages of both PHA and PLA are eliminated, resulting in a biodegradable plastic with better material characteristics. By using PLA, which is available in larger quantities and at a lower cost, also a lower price of the final product can be obtained[5].

Table 7. Benefits and challenges of biodegradable plastics produced by bacteria

Benefits	Challenges
Widely biodegradable (industrial and home composting, soil, water, marine conditions and anaerobic digestion)	Production capacity is today still relatively small although this might change in the near future when the new plant of Mirel becomes operational
Renewable resources	High energy demand due to the fermentation process, resulting in a negative contribution on LCA
Heat resistant	
	High cost
High efficiency in production possible through metabolic engineering	
Similar conversion processes as for conventional plastics can be used	

5.4.5. Biodegradable plastics of petrochemical origin

A large number of biodegradable plastics are based on petroleum resources obtained chemically from synthetic monomers. The most used petrochemical based biodegradable plastics are polyvinyl alcohol (PVOH), polybutylene succinate (PBS), polycaprolactone (PCL), polybutylene adipate terephthalate (PBAT) and polybutylene adipate co-succinate co-terephthalate (PBAST).

PVOH is a water soluble synthetic polymer which is produced by partial or complete hydrolysis of polyvinyl acetate. The properties of PVOH dependent on the level of humidity. Under conditions of higher humidity, more water is absorbed which will act as a plasticizer. However, when coming in contact with compost or soil, PVOH will be absorbed by the clay particles present in the compost or soil, leaving the PVOH no longer available for degradation.

PBS is an aliphatic copolyester obtained by the combination of diols such as 1,2-ethanediol, 1,3-propanediol or 1,4-butadenediol, and dicarboxylic acids such as adipic, sebacic or succinic acid. The biodegradability of PBS depends on the structure. In this context, the addition of adipic acid, which decreases the cristallinity, tends to increase the overall biodegradation of the product.

PCL is obtained by ring opening polymerization caprolactone in the presence of aluminium isopropoxide. PCL is widely used as a PVC solid plasticizer or for polyurethane applications, but it finds also some applications based on its biodegradable character in domains such as controlled release of drugs and soft packaging. Due to its relatively low melting point, PCL is mostly blended with other biodegradable plastics (mostly starch based) to improve the overall product characteristics.

Aromatic copolyesters like PBAT and PBAST are often based on terephtalic diacid and are flexible and tough.

Some producers: BASF (PBAT, Ecoflex®), DuPont (PBAT, Biomax®), IRE Chemical (PBS, EnPol®), Showa Denko (PBS, Bionolle®), Solvay (PCL, CAPA®).

Applications: Packaging, films, bags

End of Life options: Industrial composting

Table 8. Benefits and challenges of biodegradable plastics of petrochemical origin

Benefits	Challenges
Biodegradable (industrial composting)	Biodegradability in other environments besides industrial composting unclear
Technical characteristics	Petrochemical based

5.5. Industrial associations

There are several industrial associations specifically lobbying for biodegradable plastics active in Europe. The most important one is European Bioplastics[9], representing industrial manufacturers, converters and users of biodegradable and/or biobased plastics and their derivative products. European Bioplastics supports and promotes the market introduction of the biodegradable and/or biobased plastics innovation. In this context, it seeks dialogue with all interest groups and governmental bodies involved and provides expert advice and assistance to its members.

In addition to European Bioplastics, which is active on a European level, there also exist several national organizations such as the Italian Bioplastic and Biodegradable and Compostable material Association (Assobioplastiche), the Belgian BioPackaging (BBP), the Dutch compostable products association (*Belangenvereniging Composteerbare Producten Nederland*, BCPN) and the French compostable products association (*Club Bio-plastiques*).

5.6. Data on degradation of biodegradable plastics

Objective proof of (bio)degradation (and compostability) of biodegradable plastics is available under the form of certificates (see chapters 7.1 and 7.2). The very first certificate was granted already at the end of the nineties, by Vinçotte, when standards and criteria were in place. Since, the number of certificates at Vinçotte exponentially increased and by mid-2012 Vinçotte had certified more than 600 products, inks, additives, intermediates and basic materials.

In parallel with Vinçotte, also DIN CERTCO assigned their first (seedling) certificate in 1997 and has grown exponentially since. Mid-2012, DIN CERTCO had certified more than 450 products, additives, intermediates and basic materials. Also the other certification bureaus worldwide (BPI, ABA, JBPA) noticed a significant increase in the number of certificates in the last years.

Although on a B-2-C level each compostability logo has its geographical value, certificates are, on a B-2-B level considered to be equal as they are all based on the same set of standards. In this context, European certification bureaus have also granted certificates to companies outside Europe, including the US, Brazil, Asia, Australia and the Middle-East. Similarly, also certification bodies outside Europe have several European companies amongst their clients.

All certification bureaus only work with recognized laboratories which have been audited and approved by the certification bureau for biodegradability and compostability testing. Only if the below criteria are being met, laboratories can be recognized and reports can be accepted for certification.

- The technical competence of the laboratory personnel is kept up-to-date by training;
- The personnel is free from any commercial, financial and other pressures which might influence their technical judgment and that any influence of outside persons or organizations on the results of inspection and testing is precluded;
- The testing laboratory does not engage in any activities that may endanger the trust in its independence of judgment and integrity in relations to its testing or calibration activities;
- An ISO 17025 quality assurance management system is in place;
- The testing laboratory is not in any way connected to the certification bureau, as such fulfilling the need of having three independent parties involved in the testing and certification process (product or material producer/supplier, testing laboratory and certification bureau).

In addition to the fact that reports are always coming from independent, recognized and reliable laboratories, these are always also reviewed in detail by the certification body in order to evaluate the validity of the results and to judge whether the product or material fulfils the different criteria as set forth by the applicable standard.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 34 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

In most cases, reports are being reviewed by one member of the certification committee after which the dossier is presented to and discussed at the certification committee prior to granting the certificate. In the US however, reports need to be reviewed by three members of the certification committee in case the material or product has not yet obtained a certificate in Europe. In case the material or product is already certified in Europe, review is done by only one member. Also within Europe there is no back-to-back agreement between the different certification bureaus.

Similar to the set of criteria which recognized laboratories need to fulfil in order to get recognized and approved by certification bureaus, also the certification bureaus themselves need to comply with several rules and regulations in order to become accredited for the certification of materials and products. Currently these requirements are laid down in the European norm EN 45011 (1998) '*General requirements for bodies operating product certification systems*' and the international standard ISO 17065 (2012) '*Conformity assessment – Requirements for bodies certifying products, processes and services*'.

ISO 17065 includes obligatory requirements on, amongst others:

- Management of impartiality, disallowing any commercial, financial or other pressures that can compromise its impartiality. Therefore, the certification bureau has to identify risks on an on-going basis arising from its activities, relationships or personnel;
- Non-discrimination, making its services accessible to all applicants whose activities fall within its scope of operation;
- Personnel, ensuring complete independence of all employees from any commercial and other interest;
- Confidentiality, covering all information obtained or created during the performance of certification activities, except for information that the client makes publically available;
- Management systems, proving the consistent fulfilment of the requirements of ISO 17065.

In other words, in case a material or product is certified in accordance with one of the above discussed standards or schemes, test results were first obtained through an independent, accredited and recognized laboratory and afterwards reviewed and approved by an independent and accredited certification bureau. These results can therefore be considered as highly reliable.

Besides the several hundred certified materials and products, from which the underlying reports and data are confidential, also a lot of publically available data is available on the biodegradation and compostability of biodegradable plastics. The most relevant results are shown in Table 9 and are discussed in detail below. In order to allow a direct comparison of the different results, each article was granted with a value.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Even though this is a subjective evaluation from the authors, the value given is based on five different parameters. Depending on the situation per parameter, 1 point or no points were granted to the article.

1. Testing facility:
   Test results are ideally obtained through independent testing performed at an accredited laboratory. In this context, no points were granted in case results were obtained at universities or non-accredited laboratories or research centres while 1 point was granted in case testing was performed by an independent and accredited laboratory.

2. Review:
   Results gain value in case these have been reviewed by independent specialists which were not involved in the work. Therefore, in case the article has been peer reviewed and published in a scientific magazine, 1 point was allocated. In case results were not peer reviewed, no points were granted.

3. (Bio)degradation method:
   Numerous test methods exist on a worldwide scale and many of these were reviewed, accepted and published by the (inter)national standards associations (ISO, CEN, ASTM,…). Hence, studies which used these (inter)nationally accepted test methods to quantify the biodegradation were granted 1 point, while articles which referred to self-developed test methods were granted zero points.

4. Quality control:
   As discussed above, reference materials always need to be included to validate the test method and conditions. All referenced articles did include a reference material, but not all showed a sufficient biodegradation level within the foreseen time frame. In this context, no points were assigned to the articles which showed insufficient biodegradation rates for the reference material, while 1 point was granted to articles which did pass the validation criteria.

5. Number of replicates:
   As biodegradation is a biological process, variation cannot be avoided. Nevertheless, this can be quantified by using several replicates. Therefore, in case 2 or more replicates were used, 1 point was granted. In case only one replicate was used, the articles received no points.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Table 9. Overview of the most relevant biodegradation results for biodegradable plastics

Source	Test material	Result	Value
Biodegradation in compost (elevated temperature)			
Certification	Many different types	At least 90% within 180 days	5
Round robin testing[10]	PCL	100% after 67 days	4
Round robin testing[10]	PBAT (Ecoflex)	81% (100% relative) after 33 days	4
Round robin testing[10]	PLA	98% after 150 days	3
Round robin testing[10]	PCL	100% after 25-30 days	3
Biopack[11]	PLA & PLA/PLC blend	100% after 76 days (for both polymers)	4
Forbioplast[12]	PLA	100% after 80 days	4
Forbioplast[12]	PHB	90% after 80 days	4
Hydrus[13]	PBAT/PLA blend (Ecovio)	100% after 81 days	4
Hydrus[13]	PLA (Bioflex)	90% after 84 days	4
Jayasekara[16]	PBSA/starch blend	> 90% after 45 days	2
Feuilloley[17]	PCL/starch blend (Mater-Bi)	> 90% after 50 days	4
Feuilloley[17]	PBAT (Ecoflex)	60% after 50 days	4
CRU[18]	PLA	61% (85% relative) after 45 days and increasing	3
CRU[18]	TPS	60% (84% relative) after 45 days and increasing	3
CRU[18]	PHA	64% (89% relative) after 45 days and increasing	3
CRU[18]	PBAT (Ecoflex)	60% (84% relative) after 45 days and increasing	3
Kale[19]	PLA	78-84% after 58 days	4
Pradhan[23]	PLA	85-90% after 100 days	4
Kijchavengkul[26]	PLA	65% after 63 days and increasing	2
Rudeekit[22]	PLA	100% (relative) after 120 days	5
Weng[27]	PHB & PHBV	81% after 60 days (for both polymers)	2
Kijchavengkul[28]	PBAT (Ecoflex)	Approx. 70% after 25 days	3

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 37 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Table 9 (Continued). Overview of the most relevant biodegradation results for biodegradable plastics

Source	Test material	Result	Value
Biodegradation in soil (ambient temperature)			
Fair[14]	PBS (Bionolle)	100% after 110 days	3
Ratto[15]	PBSA	70-75% after 368 days	3
Biodegradation in soil			
Feuilloley[17]	PCL/starch blend (Mater-Bi)	Approx. 90% after 84 days	4
Biodegradation in fresh water (ambient temperature)			
Feuilloley[17]	PCL/starch blend (Mater-Bi)	Approx. 80% after 117 days	4
Biodegradation under anaerobic digestion conditions			
Yagi[23]	PLA	70-90% after 80-110 days at 55°C	3
	PCL	90% after 60 days	4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

A first source of data is round robin testing[10]. In 2005, the Japan Bio-industry Association (JBA) organized a round robin test to validate the MODA test method for the determination of biodegradation under controlled composting conditions. Laboratories from China, Italy, India, the United States, Sweden and Belgium participated to this round robin testing. Two laboratories directly compared the MODA test method with the ISO 14855 test method. The first laboratory tested three plastic polymers: PCL, PLA and PBAT (Ecoflex). PCL and Ecoflex were tested in duplicate and showed a biodegradation level of respectively 100% and 81% (100% relative to cellulose) after respectively 67 and 33 days, while PLA was tested in singular and showed 98% biodegradation after 150 days. Similarly, the second laboratory tested PCL in triplicate and obtained complete biodegradation after approximately 30 days.

Besides round robin testing, many biodegradation tests have also been performed as a part of European projects. Biopack[11], a project focusing on proactive biobased cheese packaging, included biodegradation testing in accordance with ISO 14855 on pure PLA and a mixture of PLA and PCL. Both materials obtained 100% relative biodegradation within 90 days. In Forbioplast[12], a project on forest resource sustainability through biobased composite development, PLA reached complete biodegradation within approximately 80 days. Also PHB was tested in Forbioplast conform ISO 14855 and showed 90% biodegradation after 80 days.

Finally, a PBAT/PLA blend (Ecovio) and a PLA based copolyester blend (Bioflex) were tested as a part of the European project Hydrus[13], a project with the main objective to develop cross-linked flexible biobased and biodegradable pipe and drippers for micro-irrigation applications. Ecovio reached complete biodegradation within approximately 80 days, while Bioflex showed a biodegradation level of 90% after 90 days.

Also soil biodegradation tests were performed in several European projects. PBS (Bionolle) was tested in a mixture of different types of soil as a part of the European project FAIR[14] and reached a biodegradation level of 100% after 110 days.

A third and last source of data on the biodegradation and compostability of biodegradable plastics are scientific articles. As mentioned previously (see 5.4.3), PLA is currently the most widely used biodegradable plastic worldwide. Because of this, the majority of the scientific articles on the biodegradability and compostability of biodegradable plastics deal with PLA and only a few deal with other biodegradable plastics like PHA, TPS, PBAT, PBS(A) and PCL.

In 1999, Ratto et al.[15] investigated the processability, mechanical and thermal properties and biodegradability (in soil) of polybutylene succinate adipate (PBSA) and its blends with granular corn starch. The authors noted that the presence of starch significantly increased the biodegradation rate (see Figure 2). While PBSA alone reached a biodegradation level of 70-75%, and increasing, after 368 days, the addition of 5% to 15% starch resulted in a higher biodegradation rate with biodegradation levels of 65-70% after already 126-168 days.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

A polymer based on a blend of starch and Bionolle[TM] (PBS(A)) has been tested for its biodegradation under industrial composting conditions by Jayasekara et al.[16] in 2003. The polymer obtained a biodegradation level of more than 90% after 45 days (see Figure 3). In addition to the biodegradation, also the disintegration was measured. Similar to the biodegradation, complete fragmentation was obtained within 5-6 weeks (see Figure 4).

In 2005, Feuilloley et al.[17] monitored the biodegradation of two biodegradable plastics: Mater-Bi (40% PCL/60% starch) from Novamont and Ecoflex (PBAT) from BASF (see Figure 5). Testing was performed in ten different environments, covering compost, soil, fresh water and anaerobic conditions.

Mater-Bi showed biodegradation levels of 75-88%, depending on the environment in which it was tested. Only in the closed bottle tests a lower biodegradation level was measured. Ecoflex only showed positive biodegradation results under controlled composting conditions, which was in line with the expectations.

Similar to the work done by Feuilloley, the Chico Research Foundation (CRU)[18] tested in 2007 a series of commercially available biodegradable plastics, including a PLA lid/straw, a TPS based BioBag trash bag, a PHA bag and an Ecoflex (PBAT) bag. Testing was done both on lab-scale, in triplicate, as well as on industrial scale using windrow and in-vessel composting systems.

All biodegradable plastics disintegrated completely in the commercial composting plants and also the laboratory tests showed positive results. Within 45 days, all samples reached a relative biodegradation level of at least 80% and net biodegradation was still increasing rapidly.

Further work on PLA was done by Kale et al.[19], who investigated the biodegradation performance of PLA bottles under simulated industrial composting conditions. Testing was done using a cumulative measurement respirometric system based on ISO 14855 and after 58 days an absolute biodegradation level of 84% was obtained, or 98% on a relative basis compared to the positive reference (see Figure 6).

In addition, Kale et al.[20] also compared the compostability of two commercially available PLA packages, a bottle and a deli container, under industrial composting conditions. The disintegration profile of both packages is shown in Figure 7 and Figure 8, from which it can be concluded that already after 30 days complete disintegration was obtained for both the bottle and deli container.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS
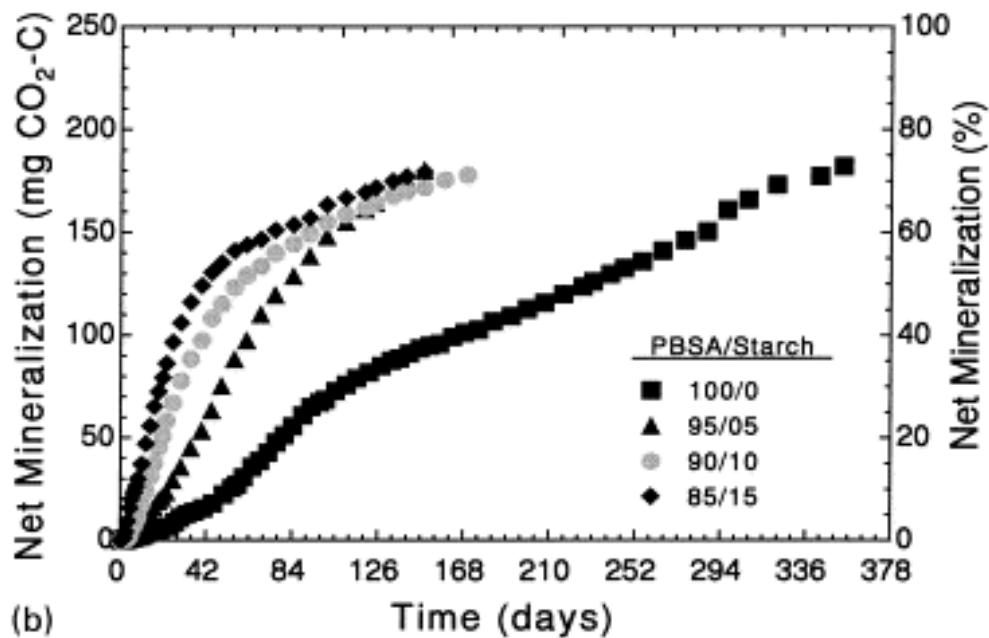

Figure 2. Biodegradation profile of PBSA and PBSA/starch systems in soil at 30°C, as reported by Ratto et al.
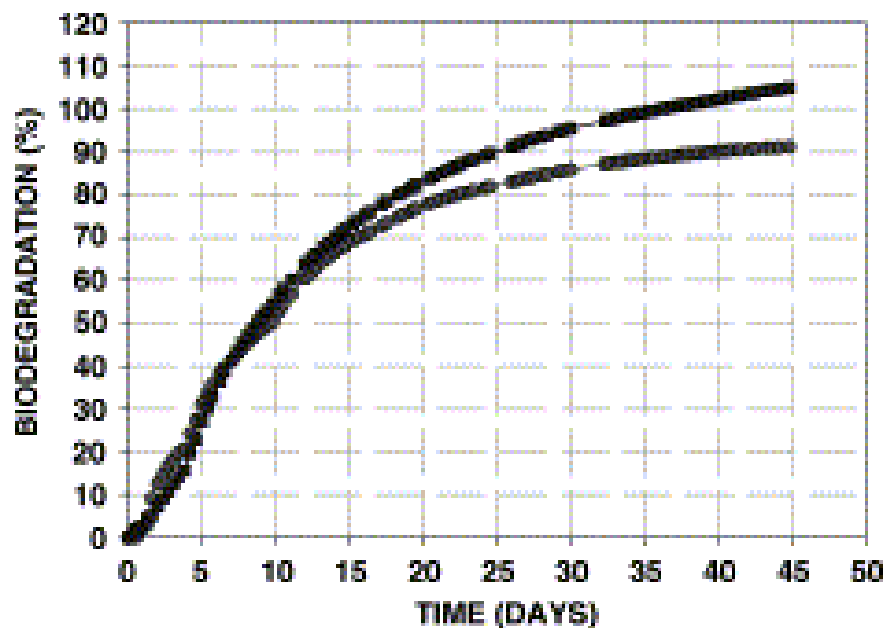

Figure 3. Biodegradation profile of Bionolle[TM] starch blend in compost at 58°C, as reported by Jayasekara et al. The upper curve represents cellulose, the lower curve the polymer.

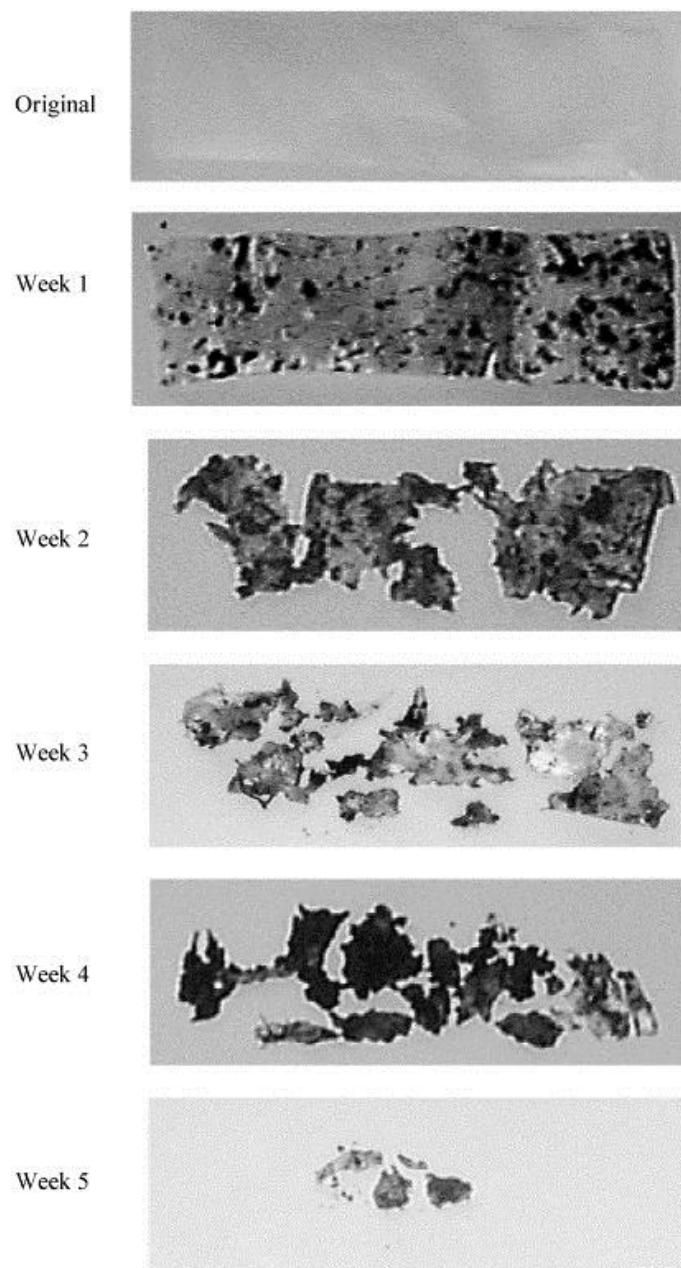**BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS**

Figure 4. Disintegration profile of Bionolle^{TM} starch blend in compost at 58°C, as reported by Jayasekara et al.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS
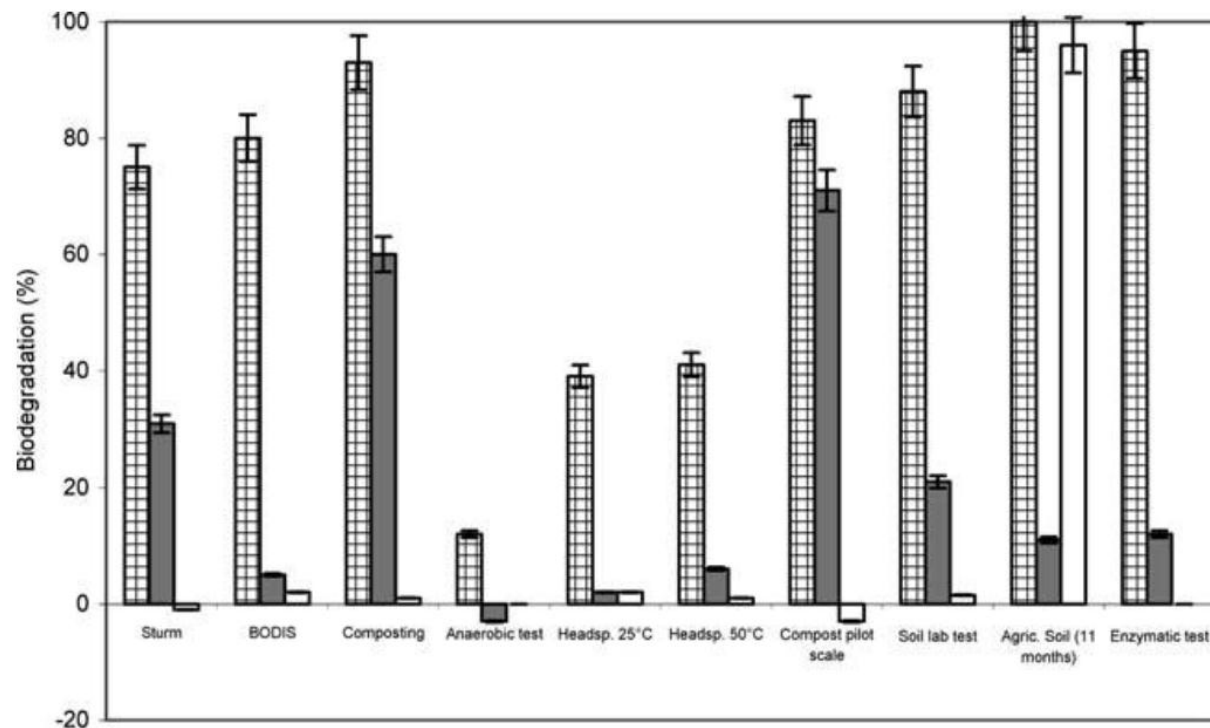
Figure 5. Overview of the biodegradation results obtained in different environments as reported by Feuilloley (2005). Left column: Mater-Bi. Middle column: Ecoflex.
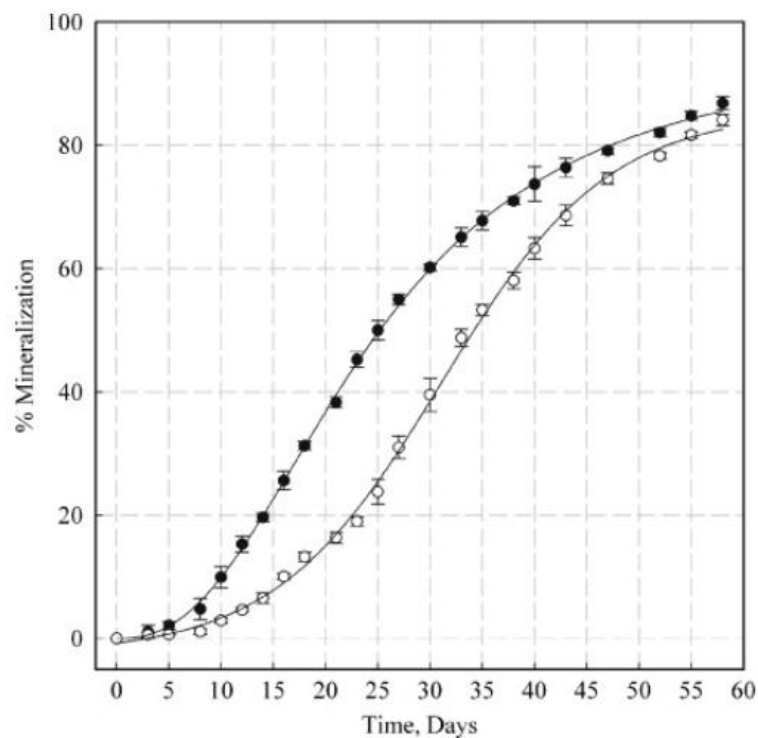
Figure 6. Biodegradation profile of PLA in compost at 58°C using the cumulative respirometric measurement system, as reported by Kale et al

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS
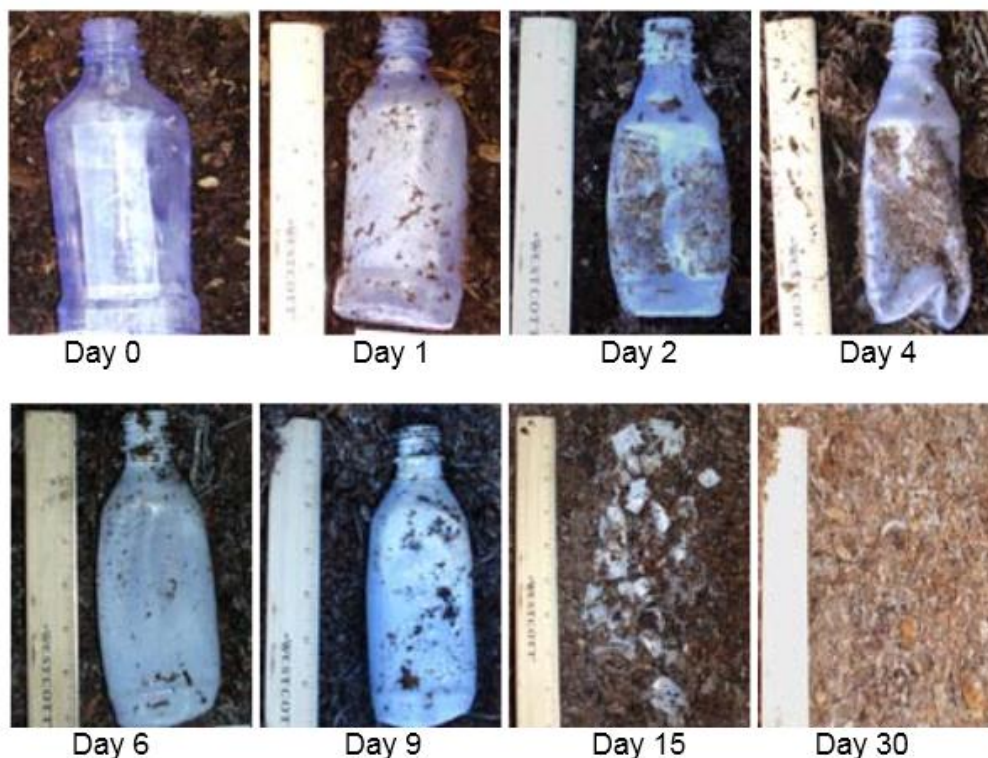
Figure 7. Disintegration profile of a PLA bottle under industrial composting conditions, as reported by Kale et al.
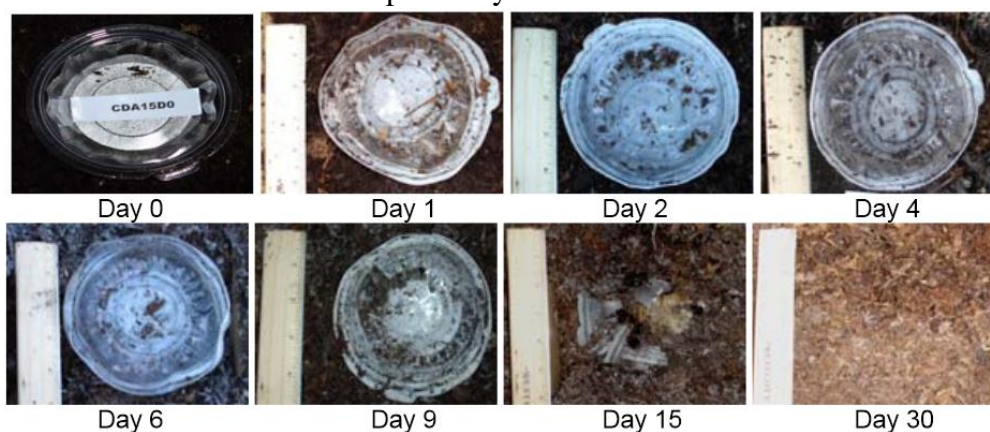
Figure 8. Disintegration profile of a PLA deli container under industrial composting conditions, as reported by Kale et al.

A similar industrial composting trial was performed by Carver County Environmental Services in 2008, commissioned by Water Billboards[21]. Water Billboards' PLA bottles were tested in a commercial composting plant for 12 weeks. The bottles were retrieved on a regular time interval and after approximately 2 weeks the majority of the bottles had already disintegrated completely. At the end of the composting trial, only one single piece of the mouth of the bottle (thickest part of the bottle) was retrieved. All other material was completely disintegrated.

The biodegradation and disintegration of PLA under industrial composting conditions was also studied by Rudeekit et al[22]. Using a lab-scale composting pile, complete disintegration was obtained within 34 days. The biodegradation test, based on two replicates per test series, was performed in accordance with ISO 14855 at 58°C, also simulating industrial composting conditions. After 120 days, both the positive reference cellulose and the PLA showed a biodegradation level of approximately 85% while net biodegradation activity was still being measured (see Figure 9).

More recently, Pradhan et al.[23] determined the extent of biodegradation of PLA and composites of PLA with wheat and soy straw in a lab scale simulated composting facility. Also this study concluded that PLA rapidly biodegrades under industrial composting conditions, reaching a biodegradation level of almost 90% within 100 days (see Figure 10).

Finally, PLA was also tested under anaerobic conditions. Yagi et al.[24] performed in 2009 biodegradation tests on PLA (and PCL) at thermophilic conditions (55°C) under aqueous conditions and obtained for PLA biodegradation levels of 70-90% within 80-110 days (depending on the dilution factor used in the inoculum). PCL showed a similar degradation rate and reached a level of 80-90% within 25-65 days, depending on the particle size (see Figure 11).

More data on the biodegradation of PLA was reported by Rudnik[25] (> 95% biodegradation in compost at 58°C after 55 days) and Kijchavengkul et al.[26] (approximately 65% biodegradation, and increasing, in compost after 63 days).
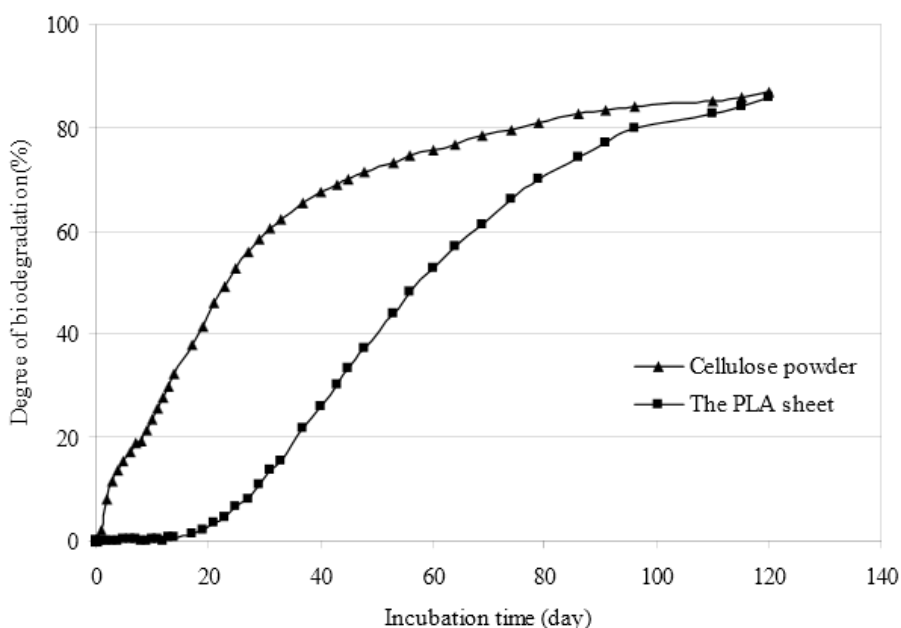

Figure 9. Biodegradation profile of PLA in compost at 58°C, as reported by Rudeekit et al.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 45 / 118

Revision: 4

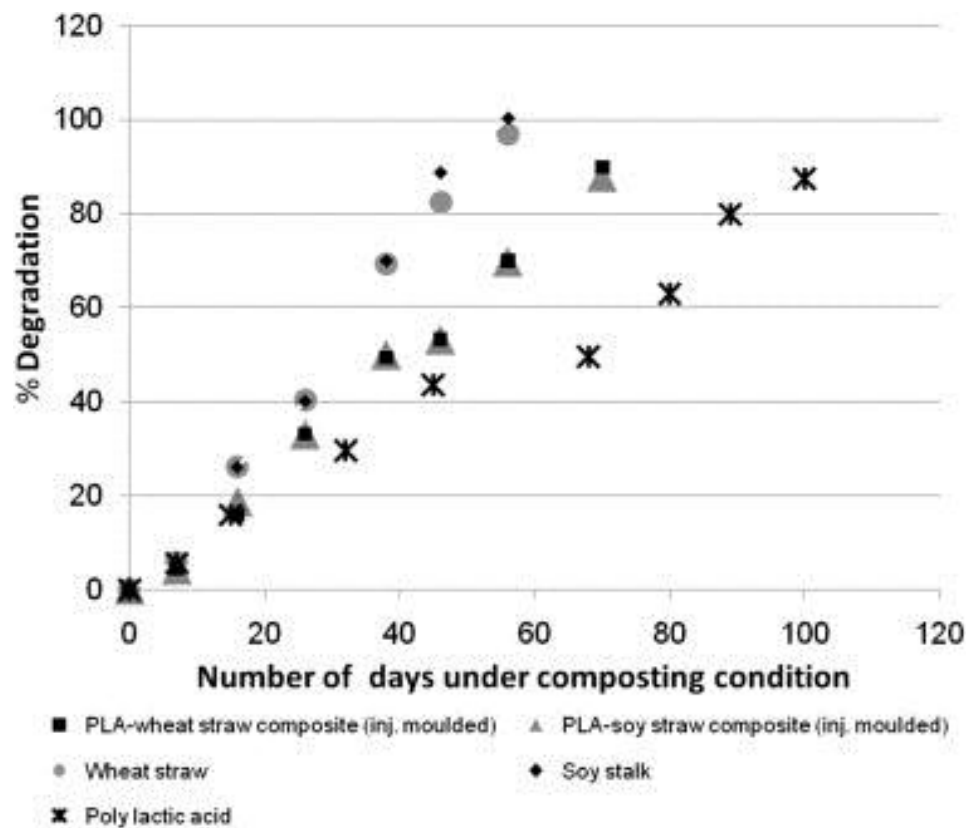**BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS**

Figure 10. Biodegradation profile of PLA and PLA composites in compost at 58°C, as reported by Pradhan et al.
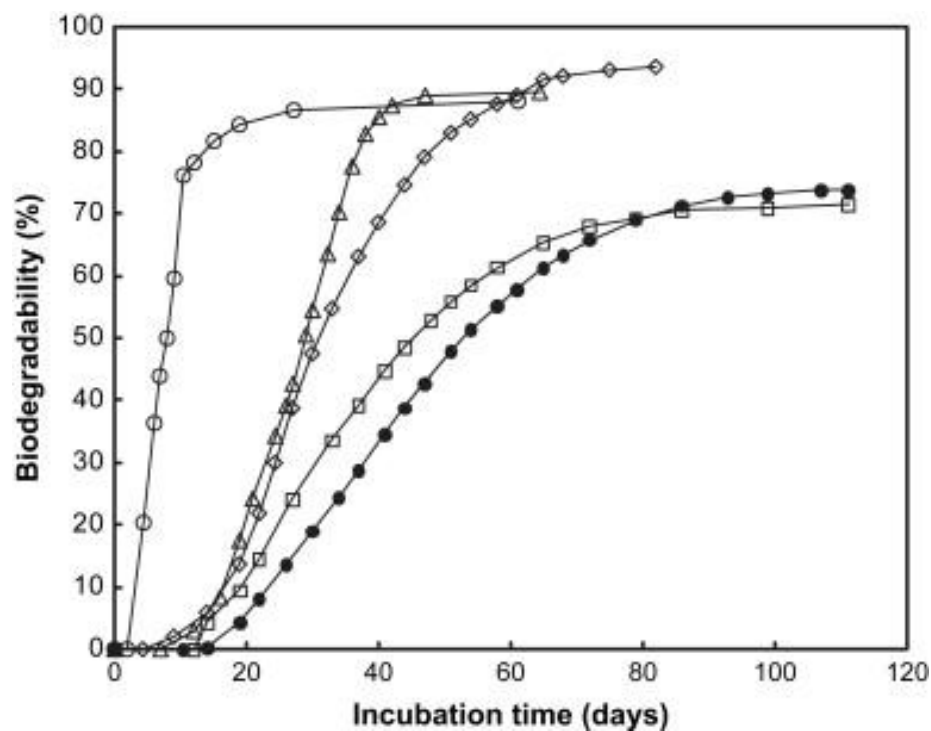

Figure 11. Biodegradation profile of PCL (△) and PLA (●) under anaerobic conditions at 55°C, as reported by Yagi et al.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Weng et al.[27] tested in 2010 a PHB and a PHBV sample under industrial composting conditions on both a pilot and a laboratory scale. In the lab scale biodegradation test, both samples obtained a biodegradation percentage of 81% after 60 days of testing (see Figure 12). In the pilot scale composting test, complete disintegration was measured within 39-42 days. Figure 13 shows the results up to 39 days. During the inspection at the 42 days point, no single piece of PHBV material could be retrieved.
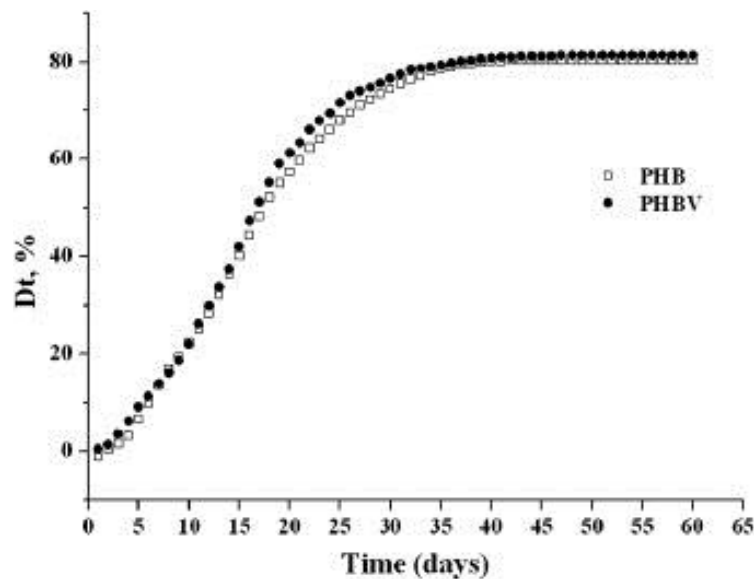

Figure 12. Biodegradation profile of PHB and PHBV in compost at 58°C, as reported by Weng et al.
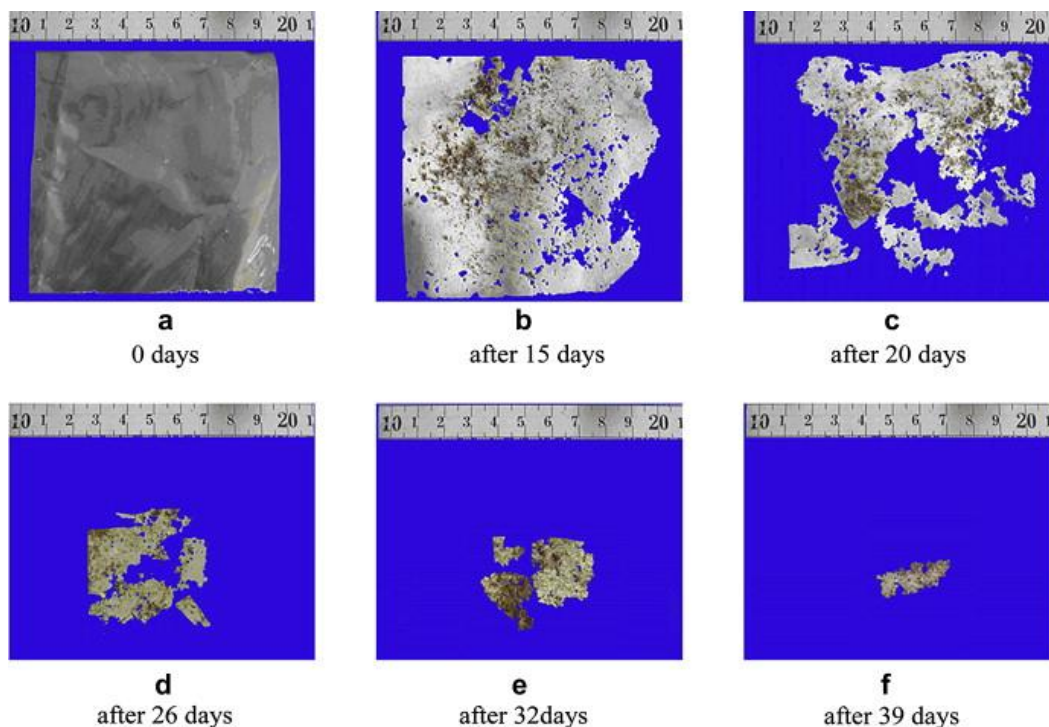

Figure 13. Disintegration profile of PHBV under industrial composting conditions, as reported by Weng et al.

Also PBAT has been tested many times. In 2010, Kijchavengkul et al.[28] investigated the effect of different types of compost on the biodegradation and hydrolysis rate of PBAT (Figure 14). When using mature compost, PBAT showed a rapid biodegradation and reached a level of approximately 70% within 20 days (90% relative compared to the reference material). When using compost derived solely from food waste or yard waste, lower biodegradation rates were obtained. After 45 days, biodegradation levels of approximately 45% and 35% were obtained. However, it must however be noted that biodegradation was still increasing after the 45 days point.

In 2010, BASF commissioned OWS with a pilot-scale composting test for the qualitative evaluation of the disintegration of four different products, including two EN13432 certified biodegradable plastic bags (see Figure 15)[29]. Already after 4 weeks of composting, complete disintegration was obtained for the biodegradable plastic bags (Figure 16 and Figure 17).
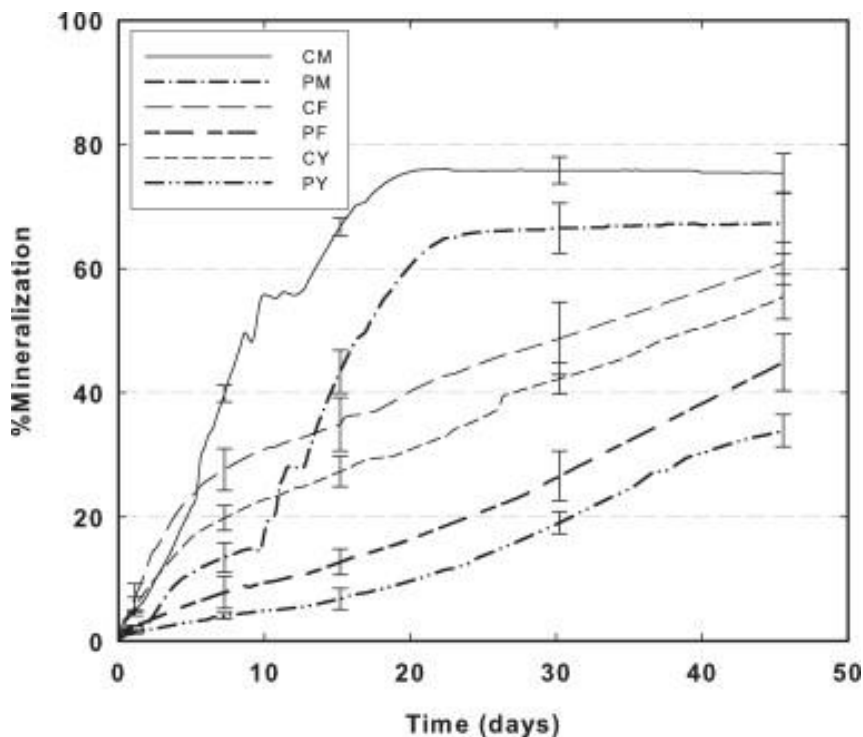

Figure 14. Biodegradation profile of PBAT in manure compost (PM), food compost (PF) and yard compost (PY) at 58°C, as reported by Kijchavengkul et al.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 48 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Figure 15. Visual presentation of the EN13432 certified biodegradable plastic bags as tested by OWS in 2010

Figure 16. Visual presentation of the retrieved pieces of the first EN13432 certified biodegradable plastic bag after 3 weeks of composting, as reported by OWS in 2010

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 49 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Figure 17. Visual presentation of the retrieved pieces of the second EN13432 certified biodegradable plastic bag after 2 weeks of composting, as reported by OWS in 2010

6. OXO-DEGRADABLE PLASTICS

6.1. Introduction

Oxo-degradable plastics are based on conventional plastics, like polyethylene (PE), polypropylene (PP), polystyrene (PS) and polyethylene terephthalate (PET), to which additives that should cause the plastic to degrade by a process initiated by oxygen and accelerated by light and/or heat have been added ("oxo-degradable" additives). The additives are typically metal salts of carboxylic acids or dithiocarbamates based on cobalt (Co), iron (Fe), manganese (Mn) or nickel (Ni), with Co being used more for packaging and Fe and Ni more for mulch film. Other transition metals like Cerium (Ce) have also been reported to exhibit strong pro-oxidative effects.
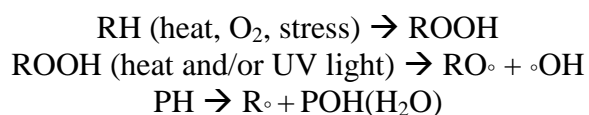
The actual content of metal in a salt is typically less than 10% with the addition level of the active ingredients being approximately 0.1% of the finished film[30].

Next to oxo-degradable plastics, plastics can also be additivated with organic additives that are claimed to be consumed by micro-organisms during which these excrete acids and enzymes that should break down the plastic (so called "enzyme-mediated degradable plastics"). It must be noted that this study, and this chapter in particularly, is focusing only on oxo-degradable plastics. However, for sake of completeness, a (short) introduction to "enzyme-mediated degradable plastics" is given in chapter 6.4.2., including information on producers, applications, end-of-life options and available data on (bio)degradation of "enzyme-mediated degradable plastics".
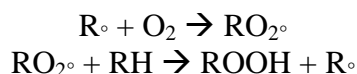
6.2. Degradation mechanism

According to Scott[31-35] and others[36], the general degradation process of oxo-degradable plastics consists of three stages (with RH = plastics molecule):
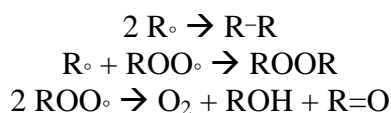
1. The initiation stage

$$RH \text{ (heat, } O_2, \text{ stress)} \rightarrow ROOH$$
$$ROOH \text{ (heat and/or UV light)} \rightarrow RO\circ + \circ OH$$
$$PH \rightarrow R\circ + POH(H_2O)$$

2. The propagation stage

$$R\circ + O_2 \rightarrow RO_2\circ$$
$$RO_2\circ + RH \rightarrow ROOH + R\circ$$

3. The termination stage

$$2\ R\circ \rightarrow R\text{–}R$$
$$R\circ + ROO\circ \rightarrow ROOR$$
$$2\ ROO\circ \rightarrow O_2 + ROH + R{=}O$$

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

In the initiation stage, the polymer chains are claimed to be cleaved by heat, atmospheric oxygen or mechanical stress, resulting in hydroperoxide groups [$ROH(H_2O)$]. In the propagation and termination stage, the degradation process continues and biodegradable intermediates are expected to be produced via the following reaction:

$$RO\circ + RH \rightarrow \text{alcohols, acids, esters, ketones,}\ldots$$
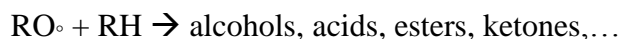
In all this, the function of the transition metals is to catalyse the decomposition of the hydroperoxide groups, not the initial cleavage of the polymer chain. In other words, it is claimed that heat, O_2, UV light and/or stress need to 'trigger' the first step of degradation (initiation stage), after which the transition metal should act as a catalysator to degrade the polyolefin further down into biodegradable intermediates like alcohols, acids, esters and ketones.

Once alcohols, acids, esters and ketones are formed, the oxo-degradable plastic should have lost strength and should have been degraded to a material of lower molecular weight compounds which can, accordingly, now be consumed by bacteria and fungi and converted to CO_2 and water[37]. Following the Oxo-degradable Plastics Association (OPA), and research done by Arnaud et al.[38], the molecular weight level at which biodegradation commences is 40,000 Dalton[39], although the majority of the guidelines and standards on oxo-degradation refer to 10,000 or even 5,000 (see Table 10-).

OPA states further that exposure to sunlight accelerates the degradation, but the process of oxo-degradation, once initiated, continues even in the absence of light, so long as air is present. It is therefore claimed that oxo-degradable plastics will degrade much more quickly outdoors at (sub)tropical temperatures than indoors at room temperature.

However, Day et al.[40] also showed that the degradation process is only achieved under dry conditions as moisture inhibits or at least significantly decreases the oxo-degradation process. As a consequence, best effects are obtained in air ventilated dry ovens.

The inhibitory effect of temperature and moisture was also revealed by the Advanced Materials Center (AMC), an independent and accredited testing laboratory based in the US who studied the performance of oxo-degradable films at different temperatures and under both high and low humidity conditions[41].

Accordingly, the time needed for complete biodegradation can be 'programmed' by varying the amount of additive and/or its formulation. Therefore they state that oxo-degradable plastics can be customized in such a way that degradation starts as soon as their programmed useful life is ended, varying from less than one year to several years.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 52 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

6.3. Standards and legislation

For many years, ASTM D 6954 (2004) was the only guide available for testing oxo-degradable plastics. The French Standardization Association (AFNOR) did publish in 2007 the experimental standard XP T54-980, but as the latter only covered mulching film and ASTM D 6954 covers all plastic products, ASTM D 6954 remained the sole reference until a few years ago.

Since 2009, several other standards were developed in Europe and the Middle East, leaving the industry with, for the time being, the following 7 guides and standards for oxo-degradable plastics:

United States: ASTM D6954 (2004) - Standard Guide for Exposing and Testing Plastics that Degrade in the Environment by a Combination of Oxidation & Biodegradation;

France: XP T54 980 (2007) – Plastics – Mulching Films made from Additivated Polyolefin with Life Time Controlled in the Environment for use in Agriculture and Horticulture;

United Arab Emirates: UAE.S 5009 (2009) – Standard and Specification for Oxo-biodegradation of Plastic bags and other disposable Plastic objects;

United Kingdom: BS 8472 (2011) - Methods for the assessment of the oxo-biodegradation of plastics and of the phyto-toxicity of the residues in controlled laboratory conditions;

Sweden: SPCR 141 (2010) – Certification rules for Classification for treatment of polymeric waste;

France: AC T51-808 (2012) – Plastics – Assessment of Oxobiodegradability of Polyolefinic Materials in the form of Films;

Jordan: JS 2004 (2012) – Packaging – Specification for Oxo-biodegradable Plastics and Packaging.

The majority of the above mentioned guides and standards is composed out of three so called 'Tiers':

Tier 1: Abiotic oxidative degradation

Using either accelerated or real-time conditions, samples are subjected to a regime of heat or light exposure in order to determine the time needed to reduce the molecular weight and/or the mechanical properties (e.g. elongation at break).

Tier 2: Biodegradation

The residues from the Tier 1 tests are subject to biodegradation testing using the environment in which the material is intended to end up after disposal (e.g. compost, soil, water, landfill,…). In all cases, except for the French standard AC T51-808, the residues are mixed with the inoculum and the amount and rate of CO_2 production, in case of aerobic biodegradation, and additionally CH_4 production, in case of anaerobic biodegradation, is measured.

Tier 3: Ecotoxicity

In order to show that the residues from the abiotic and biodegradation processes are not harmful to the environment, a measurement of the effect of the residues from Tier 2 on a variety of living organisms should be performed. These tests can include the measurement of germination rates of seeds such as cress, the growth and survival rate of earthworms (OECD Guideline 207), the growth of a variety of plants (OECD Guideline 208) and also survival, growth and/or immobilization of aquatic organisms like Rotifers, Daphnia or algae.

Table 10 gives an overview of the requirements of the different standards and guides for oxo-degradable plastics. More details are given in the following chapters.

Table 10. Overview of the requirements of the different guides and standards for oxo-degradable plastics

	Oxidative degradation	**Degradation**	**Environmental safety**
ASTM D 6954	Material characteristics: - 5% or less elongation at break - Average molecular weight of 5,000 or less	Biodegradation: - 60% for homopolymers - 90% for heteropolymers Disintegration: Not included	Heavy metals: The untreated material may not exceed the heavy metal limits as prescribed by the EPA Toxicity: No requirements
XP T54-980	Material characteristics: Increase in absorbance at 1713 cm^{-1} varying between 2/10,000 to 2/100, depending on the type of film (Class A or B) and type of lamps used (Xenon or Mercury)	Biodegradation: Not included Disintegration: Not included	Heavy metals: The untreated material may not exceed the heavy metal limits as prescribed by EN 13432, while also limit levels for PCBs and certain HAPs are prescribed. Toxicity: No significant difference in growth and/or germination with plants, earthworms and algae, compared to the control

UAE.S 5009	Material characteristics:
	- 5% or less elongation at break
	- Average molecular weight of < 5,000
	- Gel fraction of < 5%
	  • within 4 weeks
		Biodegradation:
		- 60% biodegradation
		  • within 6 months
		
		Disintegration:
		Not included
			Heavy metals:
			The untreated material may not exceed the heavy metal limits as prescribed by EN 13432 and BNQ P 9011-911-5 (for Co only)
			
			Toxicity:
			Not included

Table 8 (Extended). Overview of the requirements of the different guides and standards for oxo-degradable plastics

BS 8472	Material characteristics: Material must break or fragment after embrittlement testing	Biodegradation: No requirements Disintegration: Not included	Heavy metals: Not included Toxicity: No requirements

SPCR 141	Material characteristics:
	 - 5% or less elongation at break
	 - Average molecular weight of < 10,000
	 - Gel fraction of < 10%
	   • within 4 weeks in case of thermal peroxidation at 70°C
		Biodegradation:
		 - 60% for homopolymers
		 - 90% for heteropolymers
		   • within 24 months
		
		 Disintegration:
		 - 90% disintegration
		   • within 24 months
			Heavy metals:
			 The untreated material may not exceed the heavy metal limits as prescribed by EN 13432
			
			 Toxicity:
			 90% germination and biomass yield with plants compared to the control

AC T51-808	Material characteristics: Increase in absorbance at 1714 cm^{-1}, depending on the test performed and type of film tested (Class A to D)	Biodegradation: - ADP/ATP $\geq$ 3 after 180 days - ATP$_{test}$ $\geq$ 3 x ATP$_{blank}$ during first 6 months - Positive viability test with bacteria Disintegration: Not included	Heavy metals: Not included Toxicity: Not included

O.W.S.

Code: DSL-1

Page: 56 / 118

Date: Aug-09-2013

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

6.3.1. ASTM D6954-04

For Tier 1, ASTM D 6954-04, the US 'Standard Guide for Exposing and Testing Plastics that Degrade in the Environment by a Combination of Oxidation and Biodegradation', refers to ASTM D 5510 and ASTM D 5208, two test procedures which simulate the first oxidation process of the oxo-degradable plastic through respectively thermal degradation and UV degradation. As soon as the oxo-degradable plastic has reached 5% or less elongation at break and the fragmented parts show an average molecular weight of 5,000 or less, Tier 2 can be initiated.

Depending on the polymer type and the conditions of the Tier 1 testing, there is the possibility that, rather than causing a reduction in the average molecular weight of the polymer, gel formation or cross-linking will occur. Cross-linked polymers are generally not biodegradable, so the extent of any cross-linking needs to be measured. In this context, ASTM D 6954 prescribes a maximum limit of 10%.

The residues obtained after Tier 1 should be subjected to biodegradation testing using the environment in which the plastic is expected to end up after being disposed of. ASTM D 6954 therefore refers to ASTM D 5988, ASTM D 5338 and ASTM D 5526, representing respectively biodegradation in soil, under controlled composting conditions and under landfill conditions. No reference is made towards the number of replicates for testing.

For plastics consisting of a single polymer (homopolymers), 60% of the organic C needs to be converted to CO_2 before ending the test. For products consisting of more than one polymer (heteropolymers), 90% of the organic C must be converted to CO_2 before ending the test.

Tier 3 is endpoint specific in that it is focused on post degradation analysis after completion of Tier 2 testing. The effects of residual plastics on toxicity need to be measured as well as the impact on the soil, compost and/or water quality to evaluate the safety of biological aerobic and/or anaerobic degradation. ASTM D 6954-04 suggests verifying the effect on plant germination, plant growth, earthworm survival and Rotifer mobilization. However, the guide does not prescribe at which concentration the plastic residues need to be tested, while concentration is still the most important parameter for toxicity testing.

Besides toxicity testing, ASTM D 6954 also states that heavy metals must be determined directly on the material before degradation in order to verify that their concentrations lay within the acceptable limits as prescribed by the Environmental Protection Agency (EPA).

It must be noted that ASTM D 6954-04 is not a standard, but a guide. It does not contain any specific timescales as the time taken to commence and complete the processes of degradation and biodegradation is product specific as oxo-degradable plastics are claimed to be 'programmable' during the production process.

6.3.2. XP T54 980

In 2007, AFNOR, the French Association for Standardization, published a first (experimental) norm on oxo-degradable products, and more specifically on oxo-degradable mulching films: XP T54 980 'Plastics – Mulching Films made from Additivated Polyolefin with Life Time Controlled in the Environment for use in Agriculture and Horticulture'. Compared to other guidelines and standards, XP T54 980 allows a classification of the mulching films in two classes, depending on their lifetime:

Class A: oxo-degradable mulching film with an expected lifetime of 1-4 months
Class B: oxo-degradable mulching with an expected lifetime of 3-6 months

Unlike the other guidelines and standards on oxo-degradable products, XP T54 980 does not use a tiered approach, although it does require proof of conformity through the same degradation and environmental fate processes. Besides the heavy metal limits of NF U 52-001:2005, which are identical to those prescribed by EN 13432, XP T54 980 also requires analysing some organic substances (see Table 11).

Table 11. Limits for organic substances as prescribed by XP T54 980

Organic substance	Limit values (ppm on dry solids)
PCBs	0.8
PAH – Fluoranthene	4
PAH – Benzo(b)fluoranthene	2.5
PAH – Benzo(a)pyrene	1.5

The possible toxic effects of (the residues of) the oxo-degradable mulching film on plants, earthworms and algae need to be verified after a thermal ageing process at 60°C for 250 hours for Class A films and 950 hours for Class B films. The following criteria are included in XP T54 980 to evaluate the toxicity:

- Plants: no significant difference in growth and germination between test and blank soil;
- Earthworms: not more than 10% difference in survival rate between test and blank soil + no significant negative effect on the weight of the earthworms;
- Algae: no significant difference in growth between test and blank soil

Finally, XP T54 980 asks to quantify the degradability of the oxo-degradable mulching film via the change in mechanical characteristics: (1) via the increase in absorption at 1713 cm^{-1}, which represents accumulation of carbonyl groups, or (2) via the determination of the elongation at break. In both cases, three situations need to be analysed:

1. Thermal ageing at 60°C for 250 hours, allowing to verify whether degradation occurs in the pre-usage phase;
2. Second thermal ageing at 60°C for 250 hours for Class A films and 950 hours for Class B films, allowing to verify whether degradation is sufficient in soil;
3. Photo ageing using Xenon or Mercury lamps for 40-300 hours for Class A films and 100-700 hours for Class B films, allowing to verify the degradation behaviour of the mulching film during use.

In each of the three above situations, the variation in absorbance at 1713 cm^{-1} needs to be below a certain level, varying from 2/10,000 to 2/100 to be in line with XP T54 980.

6.3.3. UAE.S 5009:2009

The Emirates Authority for Standardization and Metrology (ESMA), who is charged with the responsibility for standardization activities in the United Arab Emirates (UAE), published in 2009 UAE.S 5009: 'Standard and Specification for Oxo-biodegradation of Plastic bags and other disposable Plastic objects'.

Unlike ASTM D 6954 and others, UAE.S 5009 is built up of only 2 Tiers, covering abiotic oxidation (Tier 1) and biodegradation (Tier 2).

To simulate the oxidative degradation processes likely to occur in the UAE, abiotic degradation need to be simulated. UAE.S 5009 does not prescribe any specific test methods for the Tier 1 testing, but refers to ASTM D 6954 and BS 8472, with the latter still under construction at the time of publishing. Tier 1 testing can therefore be performed according to ASTM D 5510 and ASTM D 5208.

The extent of oxidation needs to be evaluated by measuring the loss in mechanical properties, decrease in molecular weight and determination of the gel content. Tier 1 testing can be considered as successful if all of the below mentioned requirements are met within 4 weeks of abiotic degradation testing:

- Average molecular weight of less than 5,000 Dalton;
- Gel fraction of less than 5%;
- Elongation at break of 5% or less.

The residual material obtained from Tier 1 testing is then tested for its ultimate aerobic biodegradability under controlled conditions in Tier 2. As in Tier 1, UAE.S 5009 does not prescribe any specific test method, but refers to ASTM D 6954 and BS 8472, allowing as such biodegradation testing in soil (ISO 17556 and ASTM D 5988), under controlled composting conditions (ASTM D 5338) and under landfill conditions (ASTM D 5526). Testing needs to be performed in triplicate and a biodegradation level of at least 60% needs to be obtained within 6 months.

UAE.S 5009 does not include any toxicity testing, but does require that the original, untreated plastic material remains within the heavy metal limits of EN 13432 and BNQ P 9011-911-5 (for Co only, see Table 3).

Finally, UAE.S 5009 also clearly states that all tests need to be performed at accredited laboratories.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 59 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

6.3.4. BS 8472

Similar to ASTM D 6954, the UK standard for oxo-degradable plastics BS 8472 'Methods for the assessment of the oxo-degradation of plastics and of the phyto-toxicity of the residues in controlled laboratory conditions' is also composed of 3 Tiers.

The abiotic oxidation step can be realized using photo-oxidation and/or thermal oxidation. Photo-oxidation should be mimicked using the exposure cycle as prescribed by ISO 4892-3, while thermo-oxidation should be performed conform ASTM D 5510, which is also one of the options in ASTM D 6954. The degree of oxidation is measured by two simple embrittlement tests: bending the material so that its opposite edges touch and rubbing the sample between thumb and first finger. If the material break or fragments under one of both embrittlement tests, Tier 1 can be terminated. BS 8472 does not require any material characterization analysis to quantify the degree of oxidation.

The second Tier, covering biodegradation in soil in accordance with ISO 17556, uses the residues obtained after Tier 1 testing. In parallel with the biodegradation test, which requires triplicate testing, residues of Tier 1 testing are added at a 0.2% concentration to soil for the subsequent plant toxicity tests. As soon as 50% biodegradation is reached, plant toxicity testing, conform OECD 208, can be started (Tier 3).

Similar to ASTM D 6954, BS 8472 is not a specification, merely a test method. The document does not contain pass or fail criteria and contains only guidelines for performing (some of the) laboratory tests. Consequently, it is not possible to claim conformity with BS 8472 is not possible.

6.3.5. SPCR 141

SPCR 141, which was published in 2010 by SP, the Technical Research Institute of Sweden, sets forth a set of certification rules for the classification for treatment of polymeric waste. The document covers several types of polymeric waste: industrially compostable polymeric waste (Appendix 1), polymeric waste compostable in small scale (home) composts (Appendix 2), polymeric waste for digestion (Appendix 3), polymeric waste degradable by abiotic and subsequent biological degradation (A+B degradable, Appendix 4), Mechanical recycling of polymeric waste (Appendix 5), Controlled incineration of polymeric waste (Appendix 6) and Treatment of particular polymeric waste (Appendix 7). Each appendix contains specific technical requirements and test methods.

The abiotic degradation test of Tier 1 simulates or accelerates the degradation processes likely to occur in a chosen application and disposal environment. Like UAE.S 5009, SPCR 141 also refers to ASTM D 6954 and BS 8472 for the test methods applicable for photo- and thermo-oxidation and states that the abiotic degradation must be sufficiently rapid. The period of thermal peroxidation at 70°C can therefore under no circumstance exceed 4 weeks.

The extent of degradation after Tier 1 testing is evaluated by measuring the loss in mechanical properties, decrease in molecular weight and determination of gel content. Only if the below requirements are met, Tier 1 testing can be stopped:

- Average molecular weight of less than 10,000 Dalton;
- Gel fraction of less than 10%;
- Elongation at break of 5% or less.

After the abiotic degradation process, the residue material is tested for its ultimate biodegradability in Tier 2. Testing can be performed in any defined biological active environment, but SPCR 141 suggests aerobic aqueous biodegradation testing in fresh water according to ISO 14852 or ultimate aerobic biodegradability testing in soil according to ISO 17556. Despite the chosen environment, homopolymers need to reach 60% and heteropolymers 90% within 24 months in order to comply with SPCR 141.

While the majority of the guides and standards on oxo-degradable plastics only discuss biodegradation in Tier 2, SPCR 141 also included disintegration testing. If the oxo-degradable product has a thickness exceeding 0.2 mm, the residue material obtained after Tier 1 also needs to be subjected to a disintegration test on a laboratory scale under conditions simulating a specific, biologically active disposal environment (soil, fresh water or any other defined biological active environment). If nothing else is specified for a particular application, no more than 10% of the original mass shall, within a time frame of 24 months, and after sieving over 2 mm, remain on the sieve.

As a side remark it must be noted that all standards on industrial and home compostability, refer to 3 respectively 6 months in which the product should disintegrate sufficiently (i.e. for at least 90%). The 24 months period can therefore be considered as rather long and not in line with real-life processes taking place in industrial and home composting.

Finally, Tier 3 of SPCR 141 covers toxicity testing with plants. After biodegradation or disintegration, the soil or water obtained shall be analyzed to guarantee that no negative effects are observed on plants through plant germination and plant growth testing in accordance with SP method 4149, which is based on OECD 208 and annex E of EN 13432. The pass level as prescribed by EN 13432, 90% germination and biomass yield compared to the control, applies, although SPCR 141 does not prescribe at which concentration the residues need to be tested.

In addition to the plant toxicity testing, and as also required by UAE.S 5009, SPCR 141 also requires that the oxo-degradable material does not exceed the heavy metal limits as foreseen by EN 13432 (see Table 3).

6.3.6. AC T51-808

Following XP T54 980, which was published in 2007, AFNOR published a second standard on oxo-degradable films in 2012: AC T51-808 'Plastics – Assessment of Oxo-biodegradability of Polyolefinic Materials in the form of Films'. Where XP T54 980 only covered mulching films for use in agriculture and horticulture, AC T51-808 covers plastic films in general. However, it must be noted that only films up to a thickness of 250 μm are being covered. AS T51-808 does not apply to thicker films.

AC T51-808 distinguishes two types of films:

- Type I: Oxo-degradable films containing stabilizers only to cover storage, requiring as such only a short oxidation period to start the degradation process;
- Type II: Oxo-degradable films containing stabilizers to cover both storage and use, requiring as such a long(er) oxidation period to start the degradation process.

Following the above distinction between Type I and Type II films, AC T51-808 further classifies films in 4 classes (see Table 12).

Table 12. Classes as prescribed by AC T51-808 for oxo-degradable films

Class	Time of storage and use	Temperature of storage and use
A	12	20
B	24	20
C	12	30
D	24	30

The first part of AC T51-808 covers the abiotic degradation. Similar to XP T54 980, AC T51-808 uses the increase in absorbance at $1714cm^{-1}$ or the change in elongation at break to quantify the oxidation process. Depending on the Type/Class of film, three tests are prescribed:

Test 1: Thermo-oxidation, covering storage of the film prior to use;
Test 2: Photo-oxidation, covering the actual use of the film;
Test 3: Combination of thermo- and photo-oxidation, covering the end of life of the film.

Depending on the test and type of film, conformity is reached if the increase in absorbance at $1714cm^{-1}$ or the elongation at break does or does not pass a certain value, related to the thickness of the film or not.

Once all abiotic degradation requirements are met, the residues obtained after photo-oxidation (Test 2) need to be tested for their biodegradability. Unlike other standards and guidelines, where CO_2 production is used as a parameter to quantify biodegradability, AC T51-808 uses the ratio of adenosine di-phosphate (ADP) over adenosine tri-phosphate (ATP) measured in the cells of bacteria.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

ATP, present in all living cells, consists of adenosine and three phosphate groups from which the last two bonds contain a high level of stored energy. To take advantage of this high energy bonds, ATPase cuts off the last phosphate group, turning ATP into ADP. During this process, the stored energy is released and used. On the other hand, when carbohydrates and other food sources are consumed and degraded within the cells, the energy released is used to reattach the phosphate group to ADP, turning it back into ATP.

According to AC T51-808, testing needs to be performed in a synthetic aqueous inoculum spiked with a single bacteria, *Rhodococcus rhodochrous*, and ATP levels need to be determined at several intervals: at start, after 4, 8, 12, 30, 60, 90 and 120 days and at end (after 180 days). The ADP level is only measured at the end of the test.

Finally, AC T51-808 prescribes the following three pass levels for biodegradation:

  - During the first 6 months, the ATP level in the test reactor needs to be at least the threefold of the ATP level in the blank reactor;
  - At the end of the test, after 180 days, the ratio ADP over ATP needs to be smaller than or equal to 3; and
  - At the end of the test, the *Rhodococcus rhodochrous* needs to show a sufficient growth rate in the presence of another, more preferably nutrient source.

Given that living cells maintain a ratio of ATP over ADP at a point ten orders of magnitude from equilibrium, with ATP concentrations up to a thousand fold higher than the concentration of ADP[42], the above prescribed pass ratio of ADP over ATP of maximum 3 cannot be considered as solid proof of biodegradation and only represents minimal activity within the cell.

This was also confirmed by Fontanella et al.[43]. The authors measured only moderate to low biodegradation levels in compost and soil for oxo-degradable LDPE and LLDPE samples even though ADP/ATP ratios far below 3 were obtained after 180 days (see Table 13). Fontanella et al. also tested a third oxo-degradable samples based on HDPE. This sample did not show an ADP/ATP ratio below 3, but showed a similar biodegradation level of the LDPE and LLDPE samples who did had a ADP/ATP ratio (far) below 3.

Table 13. Biodegradation percentage in compost and soil and accompanying ADP/ATP ratio of oxo-degradable HDPE, LDPE and LLDPE samples

Sample	ADP/ATP ratio	% biodegradation after 325 days in compost at 58°C	% biodegradation after 352 days in soil at 25°C
HDPE	5.8	6	5
LDPE	0.3	17	9
LLDPE	0.8	24	11

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 63 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

6.3.7. JS 2004

In 2004, the Jordan Standards Organization published the JS 2004 (2012) 'Packaging – Specification for Oxo-biodegradable Plastics and Packaging'. Unfortunately no information could be found on the required test methods and pass levels.

6.3.8. National legislation

As of January 1st, 2012, the government of the United Arab Emirates (UAE) legislated to require oxo-degradability of plastic carrier bags. At the beginning of 2013, the UAE extended the range of products to include now almost all disposable plastic products made of polyethylene (PE), polypropylene (PP) or polystyrene (PS) (see also 7.3.1).

Following the decision of the UAE's government, also the government of Pakistan decided that, as of April 2013, plastic bags (and all other plastic products) made of PE, PP or PS must be made with oxo-degradable technology. Similar to the situation in the UAE, the 'Prohibition of Non-degradable Plastic Products (Manufacture, Sale and Usage) Regulations, 2013' prohibits not only the manufacturing of conventional and bio-based disposable PE, PP or PS products in the capital city Islamabad, but also prevents them being imported into Islamabad.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 64 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

6.4. Producers of (oxo-)degradable additives

6.4.1. Oxo-degradable additives

Oxo-degradable plastics are currently used in Europe (France, Italy, Sweden, Switzerland, the Balkan states…), the Gulf states (Bahrain, Kuwait, Oman, Qatar, Saudi Arabia and the United Arab Emirates), India, China, South Africa, Mexico, Iran, Yemen and several other third world countries. In the US oxo-degradable plastics are not yet as widespread as in other countries and parts of the world.

Some producers:	Symphony Environmental Technologies ($d_2w^®$ additive), EPI Environmental Technologies (TDPA$^®$), Wells Plastics (Reverte$^®$), Willow Ridge Plastics (PDQ-M, PDQ-H, BDA and OxoTerra^{TM}), Add-X Biotech (Addiflex$^®$), EcoPoly Solutions (EcoPoly$^®$)
Applications:	Carrier and refuse bags, bin liners, gloves, mulching film, wrapping, bread bags, shrink film, bubble wrap, bottles, cups,…
End of life options:	Claims to be making the plastic biodegradable in all aerobic environments, including compost, soil and water

6.4.2. Enzyme-mediated degradable additives

Similar to oxo-degradable plastics, conventional plastics can also be enriched with organic additives, resulting in so called "enzyme-mediated degradable" plastics. The degradation process is claimed not to be initiated by heat, UV light, mechanical stress or oxygen, but by the micro-organisms themselves. According to the producers of "enzyme-mediated degradable" additives, the organic additive, together with its carrier material (in most cases this is ethylene vinyl acetate), is consumed by the micro-organisms, during which these excrete acids and enzymes that should break down the plastic into materials that are easily consumed by microbes.

The technology can be applied to both common as well as uncommon (conventional) plastics. The minimum loading rate is 1%. It is said that lower percentages do not initiate biodegradation, while higher percentages do not (significantly) accelerate biodegradation. The plastics are claimed to be recyclable, should have the same properties as conventional plastics and would be less expensive when compared to biodegradable plastics.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 65 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Some producers: Earth Nurture (ENA® additive), ECM BioFilms, Bio-Tec Environmental (Ecopure®), Enso Plastics

Applications: Same as conventional plastics

End of life options: Claims to be making the plastic biodegradable in industrial composting facilities, home composting bins, landfills, anaerobic digesters and water

Claims of Earth Nurture are based on two biodegradation tests. The first biodegradation test, performed by Biosystems Atlanta in accordance with ASTM D 5338, shows only results of the CO_2 production (65 ml/g after 14 weeks) but no data on the percentage of biodegradation. A second test, covering biodegradation under anaerobic conditions in line with ASTM D 5511, shows 1% biodegradation after approximately 55 days. From this, it is calculated that, using a straight line extrapolation, which is scientifically incorrect, a 3 mm thick sample would biodegrade completely within 12 years and a 12.5 − 15 μm thick sample within 1 month.

ECM BioFilms states that it has tested several plastic products enriched with ECM's additives at renowned and independent testing laboratories, all showing results from which it can be concluded that the products are biodegradable under both aerobic and anaerobic conditions. Test results or reports are however not available, but they claim that biodegradation will occur in more than one year (assuming a minimum loading of 1%).

Bio-Tec Environmental also states on their website that several ASTM D 5511 tests have been performed showing that EcoPure® additivated plastics are biodegradable under anaerobic conditions. Results or reports are however not available.

Enso Plastics finally, reports 24.7% biodegradation within 160 days, measured by an independent third party laboratory. It is however unclear under which conditions these results were obtained (although it is assumed it was under anaerobic conditions). Detailed results or reports are not available.

6.5. Industrial associations

The UK based Oxo-biodegradable Plastics Association (OPA) is the trade organization representing the oxo-degradable additive industry.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 66 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

6.6. Position papers

Since the introduction of the oxo-degradable plastics, several national and international organizations have either come out with a strong position against the use of oxo-degradable additives, or urge caution in weighing environmental and recycling claims as they relate to the use of these additives.

Just recently, the Bioplastics Council of the Society of the Plastics Industry (SPI) updated their 2010 position paper as new studies have become available and new terminology is now being used in the marketplace[44]. According to the council, claims on oxo-(bio)degradation are still invalid and misleading as these are not supported by scientific evidence or proof of meeting the standards of accepted, third-party vetted specifications. Since no peer reviewed data has been released publicly relating to mineralization rates that support the claims of complete biodegradation, the term 'oxo-(bio)degradable', and more specifically biodegradation in general, lacks meaning and is not supported by any recognized industry certifications or third-party peer reviewed scientific data.

Already in 2009, European Bioplastics published their position paper on oxo-degradable plastics and stated that the use of the term 'oxo-(bio)degradable' without reference to existing standards is considered as misleading and as such not reproducible and verifiable[45]. Under these conditions, the term 'oxo-(bio)degradable' is, according to European Bioplastics, free of substance.

SPI and European Bioplastics also refer to the *Guides for the Use of Environmental Marketing Claims* of the US Federal Trade Commission (FTC), revised in October 2012[46]. The section on (bio)degradable claims which the FTC notes is applicable to oxo-degradable, oxo-biodegradable and similar claims, states that (a) marketers may make an unqualified degradable claim only if they can provide that the entire product or package will completely break down and return to nature within a reasonably short period of time (defined as within one year) after customary disposal and (b) unqualified degradable claims for items that are customarily disposed in landfills, incinerators and recycling facilities are deceptive because these locations do not present conditions in which complete decomposition will occur within one year. Following this, the FTC has now already taken action against several companies using the oxo-degradable technology and using the word 'biodegradable' for marketing purposes for making false and unsubstantiated claims.

Besides SPI and European Bioplastics, several other associations have also advised against the use of oxo-degradable plastics. These include for instance the Association of Postconsumer Plastic Recyclers (APR)[47], the Environmental Protection Agency (EPA)[48], the Southeast Recycling Development Council (SERDC)[49], the Association of Oregon Recyclers[50], the European Plastics Recyclers, the Northeast Recycling Council (NERDC)[51] and the New Mexico Recycling Coalition[52].

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 67 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

6.7. Data on degradation of oxo-degradable plastics

6.7.1. Data on abiotic degradation

6.7.1.1. Molecular weight

The abiotic degradation of oxo-degradable plastics has been studied for many years[53-56]. In most peer reviewed papers, reference is being made to the molecular weight decrease as proof of abiotic degradation. Fontanella et al.[43] reported in 2010 that a heat treatment of additivated HDPE, LDPE and LLDPE films at 60°C for 400 hours reduced the molecular weight to values typically around 4,000 Dalton. Similarly, Husarova et al.[54] observed that oxidation of additivated LDPE film at 70°C for 40 days reduced the molecular weight of 260,000 to 6,400 Dalton. Chiellini et al.[55] monitored the decrease in molecular weight of thermo-oxidized additivated LDPE samples (70°C) and measured a reduction of 148,000 respectively 158,000 Dalton to values just below 5,000 Dalton (depending on the relative humidity). Also Jackubowicz et al.[57] measured a significant molecular weight decrease from 131,500 to 8,800 in 10 days at 65°C.

Furthermore, independent testing performed by RAPRA[58,59] also showed that both additivated LDPE and additivated PP samples showed a molecular weight reduction to values below 5,000 after 15 - 40 days of thermo- (40-50°C) and/or photo-oxidation.

Although in almost all cases a significant decrease in molecular weight has been measured, only few papers obtained a molecular weight value below 5,000 Dalton, the pass level for sufficient abiotic degradation as stipulated by both ASTM D6954 and UAE.S 5009:2009.

Some authors also used the carbonyl index to quantify the abiotic degradation. In most cases a shift in the absorption spectrum towards the carbonyl region (1750 – 1700 cm^{-1}) was registered, which is assumed to be an indication of the formation of different oxidized products.

Despite the fact that the above discussed papers all measured a significant decrease in molecular weight, it must be noted that this decrease was only measured after an oxidation step at elevated temperature, varying from 40°C to 75°C. These temperatures are however not representative for real-life conditions, requiring the authors to make use of time-temperature superposition principles to calculate the time needed to obtain similar molecular weight reduction but at ambient temperature. The most commonly used technique is a linear extrapolation following the Arrhenius equation. However, as degradation is caused by a number of different mechanisms, there is no guarantee that the overall behaviour is of an Arrhenius form. Celina et al.[60] clearly demonstrated in 2005 the non-Arrhenius behaviour in accelerated aging.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 68 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Nevertheless, in 2003, Jackubowicz[61] demonstrated a significant decrease in molecular weight for additivated PE samples which were thermo-oxidized for 2, respectively 8 weeks at 70°C, respectively 60°C. When applying the Arrhenius equation, Jackubowicz calculated that it would take 2.5-4.5 years before a molecular weight of 10,000 Dalton would be reached at ambient temperature (25°C). However, these figures are based on the assumption that there is a linear correlation between molecular weight and temperature over a wide range of temperature and not only over a smaller partial range, which cannot be guaranteed.

Following this, Ojeda et al.[62] studied the abiotic degradation of an additivated HDPE/LDPE blend under real-life outdoor conditions in Brazil and measured a molecular weight decrease of 183,000 to 8,300 Dalton in 280 days. As mentioned earlier, and as also confirmed by different oxo-degradable additive producers, the rate at which the molecular weight reduces over time largely depends on the oxo-degradable additive concentration in the polymer. Data on the loading rate of the additivated HDPE/LDPE blend used by Ojeda et al. was however not available.

Based on the above, a reduction in molecular weight of oxo-degradable plastics cannot be questioned if these are oxidized under the influence of light, heat and/or oxygen. The question is how far this reduction continues and whether a sufficiently low level is reached at which the plastic does indeed becomes biodegradable.

6.7.1.2. Fragmentation

As oxo-degradable plastics require heat and oxygen to degrade, some producers claim to be compatible with industrial compostability plants (note that oxo-degradable plastics which require light are not compatible as composting is always done in the absence of light). According to the producers, the oxo-degradable plastics should fragment during composting and should as such not (visually) contaminate the end product (compost).

Raninger & Steiner[63] monitored in 2000 the degradation of an oxo-degradable polyethylene sample in a full-scale composting test. After 26 weeks of in-vessel and windrow composting, it was concluded that the presence of the oxo-degradable polyethylene sample did not interfere with the composting process. However, even though the study only claims compatibility of oxo-degradable bags in industrial composting plants, results can be misleading and need to be clarified further.

Prior to the actual composting process, the oxo-degradable bags were shredded, reducing the oxo-degradable bags in size down to 5 to 25 cm² (even though DIN V 54900, the German equivalent of EN 13432 at that time, prescribed the use of 10 x 10 cm pieces). Following this, the shreds were mixed with the (inert) rejects of the composting process and stored outside for 2 weeks where temperatures up to 80° were measured (which is very high for Austria and can therefore be questioned). Afterwards the oxidized residues were mixed with organic waste and composted for 6 months. At the end of the process, the authors reported a mass loss of approximately 60% on wet weight basis (35% on dry weight basis) for the organic waste including the oxidized residues. In other words, these results show a considerable reduction in weight, but can most probably be allocated only to the organic waste itself. It cannot be considered as proof of (bio)degradation of the oxo-degradable plastic shreds.

Finally, disintegration was only quantified visually as sieving was not performed at the end of the composting process. The authors stated that the oxo-degradable plastic pieces were reduced in size throughout the composting process, resulting in pieces ranging from 1 cm to 4 cm. Taking into account that the input material was already reduced in size to 5 to 25 cm² (corresponding to a width of 2-5 cm), the fragmentation cannot be considered as significant.

Furthermore, a 2-year pilot study, reported by Davis et al.[64] in 2004 , assessed the suitability of using oxo-degradable plastic bags for the collection and open windrow composting of organic waste. The bags and content were shredded prior to the 12-weeks composting trial. At the end of the test it could be concluded that the oxo-degradable plastic bags did not degrade at the same rate as the organic matter, resulting in a finished compost with a highly visible proportion of (oxo-degradable) plastic. Due to the high volume of plastic in the screened rejects, internal recycling of the rejects was no longer possible and rejects had to be landfilled.

Also in 2007, the Chico Research Foundation (CRU)[18] tested several commercially available oxo- (and bio-)degradable plastics in three composting environments including traditional windrow, in-vessel manure, in-vessel food waste and in-vessel municipal solid waste. While the biodegradable plastics composted completely, the oxo-degradable plastic remained entirely intact in all composting plants and did not show any sign of disintegration (see Figure 18).

Figure 18. The oxo-degradable plastic bags at the end of the 180 days in-vessel composting process, as reported by the Chico Research Foundation in 2007

Finally, in 2010, BASF commissioned OWS with a pilot-scale composting test for the qualitative evaluation of the disintegration of four different products, including one oxo-degradable plastic bag from Goody[29]. Unlike the biodegradable plastics, which completely disintegrated within 4 weeks, the oxo-degradable plastic bag remained completely intact throughout the 12 weeks composting trial (see Figure 19).

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Commissioned by the Australian Competition and Consumer Commission (ACCC), OWS retested the oxo-degradable plastic bags from Goody Bags and obtained the same results. Based on this data, the ACCC took Goody to court as they falsely claimed compliance with AS 4736, the Australian standard on industrial compostability. As such, Goody was not in line with the Australian Trade Practices Act 1974, which requires that businesses provide consumers with accurate information about goods and services and states that manufacturers and/or retailers that make false or misleading representations on their packaging (including their plastic bags) or on any other product - even on imported items - will find themselves in breach of the Act. Finally, the Federal Court of Australia ordered Goody to publish corrective notices on its website and in newspapers, implement a Trade Practices Compliance and Education Program and contribute to ACCC's costs of the proceedings.

Already in 2004 and 2006 similar lawsuits were held. In Italy, against Coop Italia, and in Australia, against Earthstrength Bags.

Figure 19. Visual presentation of the oxo-degradable plastic bag at start (left) and after 12 weeks of composting (right), as reported by OWS in 2010.

O.W.S.

Code: DSL-1

Page: 71 / 118

Date: Aug-09-2013

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

6.7.2. Data on biotic degradation

A significant amount of data is available on the biodegradation of oxo-degradable plastics. However, before going into the details of the available data, an overview is given of the most important parameters which need to be taken into account when evaluating (the validity of) biodegradation results:

1. Several methods exist to quantify the biodegradation of a plastic product. In the majority of the publications biodegradation was determined by a change in physical properties, like molecular weight, biomass weight, elongation at break and relative viscosity. Others referred to microbial growth on the surface of the polymer or the change in adenosine diphosphate (ADP) concentration in the micro-organisms[65]. However, only a few authors referenced the conversion of organic C to CO_2, while this latter method is the most direct and only correct measurement to quantify biodegradation.

2. Besides the test sample, also a reference sample needs to be taken along. This reference material should reach a minimum level of biodegradation during the first days/weeks of testing in order to assure optimal testing conditions. Table 14 gives an overview of these validity criteria for the three most referenced biodegradation tests in the oxo-degradable plastics industry;

Table 14. Validity criteria for the reference material for biodegradation testing

Test method	Environment	Criteria
ISO 14855	Industrial/Home composting	70% within 45 days
ISO 17556	Biodegradation in soil	60% at the end of the test
ASTM D5511	Biodegradation in AD/landfill	70% within 15 days

3. As biodegradation testing is still performed in biological systems, variation cannot be avoided. Therefore, testing should be done using at least two, preferably three replicates for all test series. Furthermore, the variation in CO_2 production between the different replicates of the reference material should be less than 20% at the plateau phase or at the end of the test (cfr. ISO 14855, ISO 17556 and ASTM D5511);

4. Finally, also the inoculum used should be evaluated for its quality. In case of ISO 14855 testing, the compost inoculum in the blank control should have produced more than 50 and less than 150 mg of CO_2 per g of volatile solids (mean values) after 10 days.

Numerous scientific articles are available on the biodegradation of oxo-degradable plastics. An overview of the most relevant results are shown in Table 15 and are discussed in detail below.

Similar to the publications on biodegradable plastics (see Chapter 5.6), each article was granted with a value, allowing as such a comparison between the many results discussed below.

O.W.S.				
Code:	DSL-1		Page:	72 / 118
Date:	Aug-09-2013		Revision:	4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Compared to the biodegradable plastics, were the evaluation was based on 5 different parameters, one extra parameter was included for the evaluation of the results of the oxo-degradable plastics:

> Abiotic degradation method:
> As oxo-degradable plastics require oxygen and light and/or heat to (bio)degrade, an preceding abiotic degradation phase is required. However, several methods are available, and some are much more severe than others. Consequently, experiments which were based on non-realistic pretreatment methods, i.e. at temperatures of 70°C or higher, did not receive any points. Experiments which used more realistic pretreatment methods, i.e. at temperatures below 55°C for longer periods and up to 65°C but for shorter periods (less than 10 days), were granted 1 point.

The 5 parameters which were also used for the evaluation of the results on the biodegradable plastics are briefly shown below:

1. Testing facility:
   No points were granted in case results were obtained at universities or non-accredited laboratories or research centres while 1 point was granted in case testing was performed by an independent and accredited laboratory.

2. Review:
   In case the article has been peer reviewed and published in a scientific magazine, 1 point was allocated. In case results were not peer reviewed, no points were granted.

3. Biotic degradation method:
   Studies which used these (inter)nationally accepted test methods to quantify the biodegradation were granted 1 point, while articles which referred to self-developed test methods were granted zero points.

4. Quality control:
   No points were assigned to the articles which showed insufficient biodegradation rates for the reference material, while 1 point was granted to articles which did pass the validation criteria.

5. Number of replicates:
   In case 2 or more replicates were used, 1 point was granted. In case only one replicate was used, the articles received no points.

Table 15. Overview of the most relevant biodegradation results for oxo-degradable plastics

Author	Abiotic treatment	Result	Status at end of test	Value
Biodegradation in compost (elevated temperature)				
Chiellini & Corti[67,68]	44 days (55°C)	28% for the < 0.6 mm parts (430 days)	Still increasing	4
Feuilloley[17]	None	No significant biodegradation (50 days)	-	5
CRU[18]	None	2% (45 days)	Plateau was reached	4
Ojeda[71]	Real life exposure (1 year)	5% (3 months)	Still increasing slowly	4
Husarova[54]	40-80 days (70°C)	23% (460 days)	Plateau was reached	3
Husarova[54]	40-80 days (70°C)	19% (250 days)	Plateau was reached	3
Husarova[54]	40-80 days (70°C)	7% (400 days)	Plateau was reached	3
Fontanella[43]	Corresponding to 3 years outdoor	5-25% (325 days)	Plateau was reached	4
Jackubowicz[57]	10 days (65°C)	43% (607 days)	Plateau was reached	6
Biodegradation in compost (ambient temperature)				
Ojeda[71]	Real life exposure (1 year)	12% (3 months)	Plateau was reached	3
Biodegradation in soil (ambient temperature)				
Chiellini & Corti[67,68]	44 days (55°C)	49-63% for the < 0.6 mm parts (600 days)	Still increasing slowly	4
Chiellini & Corti[67,68]	44 days (55°C)	46% for the < 0.6 mm parts (520 days)	Still increasing slowly	4
Jackubowicz[61]	28 days (70°C)	60-65% (200 days <u>at 60°C</u>)	Still increasing slowly	4
Feuilloley[17]	None	No significant biodegradation (84 days)	-	5
Husarova[54]	40-80 days (70°C)	13-16% (460 days)	Still increasing slowly	3
Fontanella[43]	Corresponding to 3 years outdoor	4-12% (352 days)	Plateau was reached	3
Jackubowicz[57]	10 days (65°C)	91% (733 days)	Plateau was almost reached	6

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 74 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Table 15 (Extended). Overview of the most relevant biodegradation results for oxo-degradable plastics

Author	Abiotic treatment	Result	Status at end of test	Value
Biodegradation in fresh water (ambient temperature)				
Feuilloley[17]	None	No significant biodegradation (180 days)	-	5
Feuilloley[17]	None	No significant biodegradation (117 days)	-	5
Chiellini[69]	25 days (70°C)	42-48% for the low Mw extracts (100 days)	Plateau was reached	3
Chiellini[69]	25 days (70°C)	12% for the complete sample (100 days)	Plateau was reached	3
Biodegradation in marine water (ambient temperature)				
CRU[18]	None	No disintegration (60 days)	-	4
Biodegradation under anaerobic digestion conditions				
Feuilloley[17]	None	No significant biodegradation (58 days)	-	5
CRU[18]	None	No significant biodegradation (15 days)	Plateau was reached	4
NSF International[70]	None	No significant biodegradation (60 days)	Plateau was reached	4
NSF International[72]	None	4% (60 days)	Plateau was reached	4
OWS[73]	None	No significant biodegradation (60 days)	Plateau was reached	5
Biodegradation in landfill				
MJ Carter Associates[66]	Real life exposure (14 months)	Increase and decrease in melt flow index	-	n.a.
Biodegradation in pure cultures				
Volke-Sepulveda[76]	15 days (80°C)	0.37-0.57% (31 months)	Plateau was reached	n.a.
Bonhomme[77]	300 hours (60°C)	Colonization	-	n.a.
Lee[81]	4-20 days (70°C)	Decrease in molecular weight	-	n.a.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

In 2002, EPI commissioned M. J. Carter Associates to demonstrate that polyethylene additivated with their TDPA additive would mechanically and chemically break down in a landfill environment[66]. Both the oxo-degradable plastic as well as a conventional polyethylene sample were disposed of in a landfill site in the UK and monitored for 14 months. As conversion to CO_2 could not be measured, the melt flow index was used to evaluate the degradation (it is assumed that the melt flow index increases as the polymer degrades and the molecular weight drops). The study showed that, below a temperature of 30°C, there was no significant difference between the oxo-degradable and conventional plastic. Once passed 30°C, after 11 months, the melt flow index of the oxo-degradable plastic samples increased, although only for less than 50% of the replicates. Data on the other replicates was not reported, assuming that those replicates showed melt flow indexes equal or lower to the initial value. At the end of the trial, after 14 months, the melt flow index of the oxo-degradable plastic samples decreased again, below the initial value. The authors concluded that 30°C should be considered as a critical temperature which needs to be obtained before degradation starts. However, given that nor an increase in the melt flow index, nor a decrease in molecular weight can be considered as proof of biodegradation, the above conclusion cannot be considered as scientifically correct.

In 2003, after the sample had undergone a thermal-oxidative degradation treatment of 44 days at 55°C, mimicking the thermophilic phase of a full scale composting process, Chiellini & Corti[67,68] tested the biodegradability of oxidized LDPE in soil and compost. Information on the additive loading level was not available. After a lag phase of approximately 150 days, during which no significant biodegradation was observed, the CO_2 production increased, resulting in a biodegradation level of 49-63% after approximately 600 days, and proceeding (see Figure 20). Testing was performed in triplicate and this for both polymer over soil ratios (70 mg/g soil versus 35 mg/g soil or 'Q1' and 'Q2' in Figure 20).

In a second soil burial test a lag phase of approximately 180 days was observed after which the biodegradation increased to 46% after approximately 520 days. The biodegradation test in compost also showed a lag phase of approximately 180 days, with a final biodegradation level of 28%, and proceeding, after approximately 430 days. Testing was performed in triplicate.

Also in 2003, Jakubowicz[61] reported 60-65% biodegradation after 200 days for two additivated polyethylene samples which were reduced in molecular weight to 5,000 Dalton and lower (exact figures were not available) after they were thermally oxidized for 4 weeks at 70°C (see Figure 21). The biodegradation test was performed in soil, although at 60°C instead of at ambient temperature. No data was available on the number of replicates used, from which it is assumed that only one replicate per sample was tested.
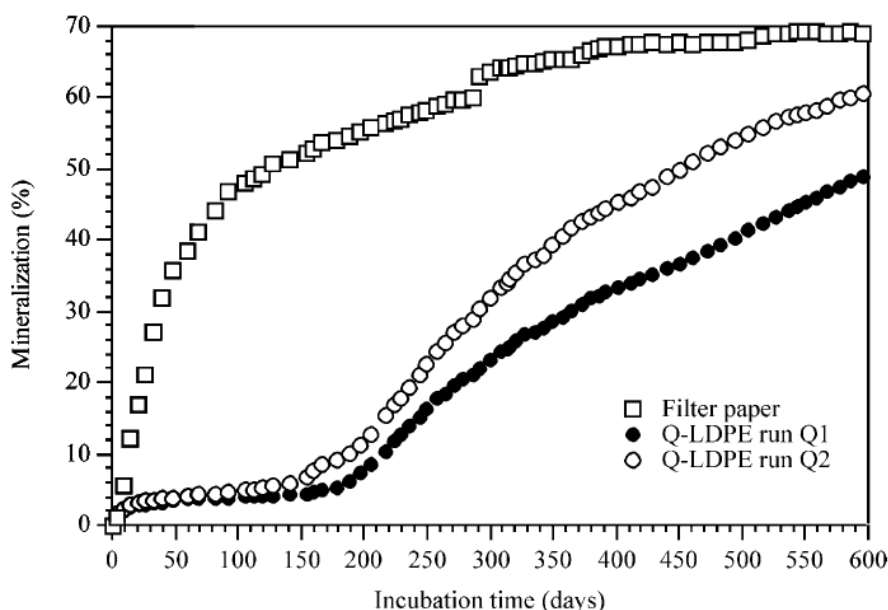
Figure 20. Biodegradation profile in soil of thermally degraded LDPE samples as reported by Chiellini et al. (2003)

Additional research on the additive loading rate showed that the biodegradation rate of the sample containing twice as much additive (AF20) was not significantly higher, although one could conclude from Figure 21 that the biodegradation of sample AF10 is levelling off and is tending towards a plateau.

Feuilloley et al.[17] tested in 2005 a number of plastics and polymers, including also one oxo-degradable plastic, in ten different biodegradation tests, covering as such all possible environments (compost, soil, fresh water and even anaerobic conditions). In nine out of ten tests, the biodegradation level obtained for the oxo-degradable plastic ranged from zero to maximum 1.8%. Only in the soil burial test positive degradation results were obtained after 11 months, although it must be noted that this conclusion was based on the visual disappearance of the plastic and not on the conversion of C to CO_2 (see Figure 22).

Following the biodegradation tests in soil and compost performed in 2003, Chiellini et al.[69] also performed biodegradation tests on thermally treated additivated LDPE samples in fresh water in 2007. After an accelerated ageing of 25 days at 70°C, biodegradation levels of 42-48% were observed after 100 days of testing for samples containing up to 15% of EPI's oxo-degradable masterbatch (see Figure 23). It should however be noted that these levels of biodegradation need to be assigned only to the extracts of the oxidized polymer, containing the fragments with the lowest molecular weight (1,080-1,270 Dalton). The complete film, with a molecular weight ranging from 4,400-5,100 Dalton, only reached a maximum biodegradation level of 12% after 100 days, while no significant degradation was observed for molecular weights between 7,600 and 10,100 Dalton. Similar to Chiellini's previous publications, testing was done in triplicate.
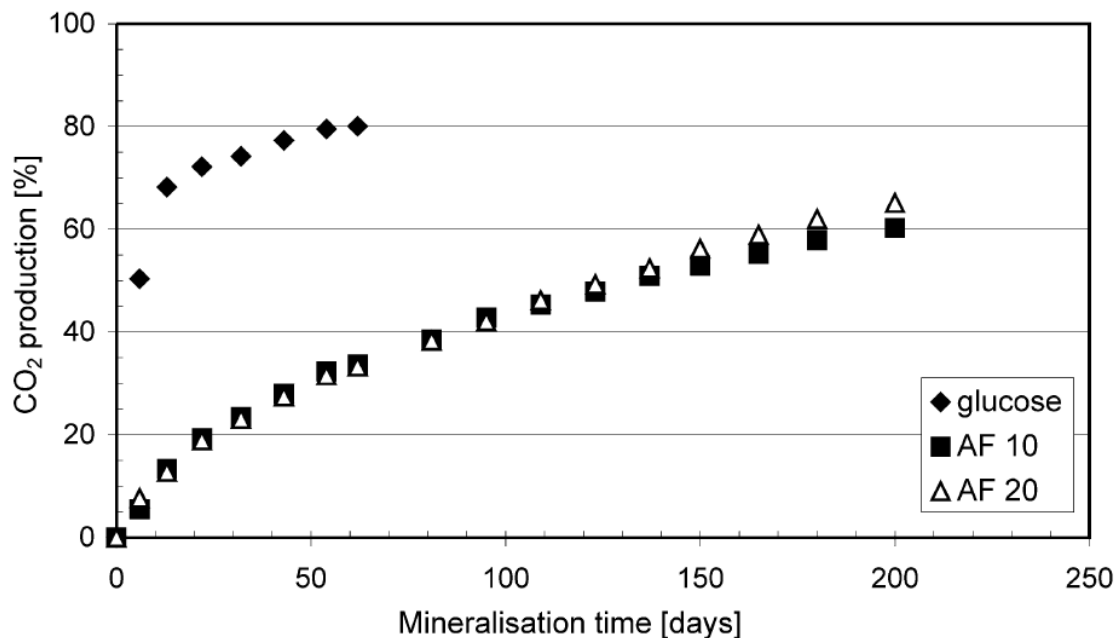
Figure 21. Biodegradation profile of thermally oxidized PE samples in soil at 60°C as reported by Jackubowicz (2003)
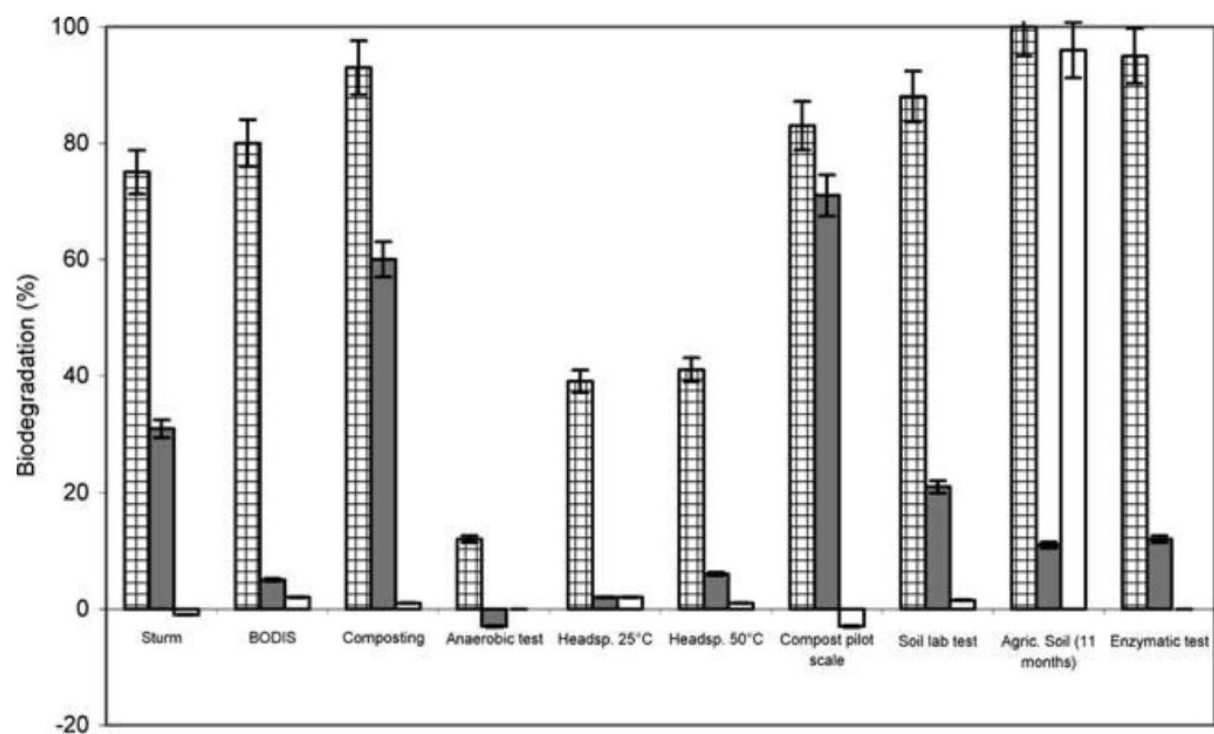
Figure 22. Overview of the biodegradation results obtained in different environments as reported by Feuilloley (2005). The results of the oxo-degradable plastic are shown in the third (white) column

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS
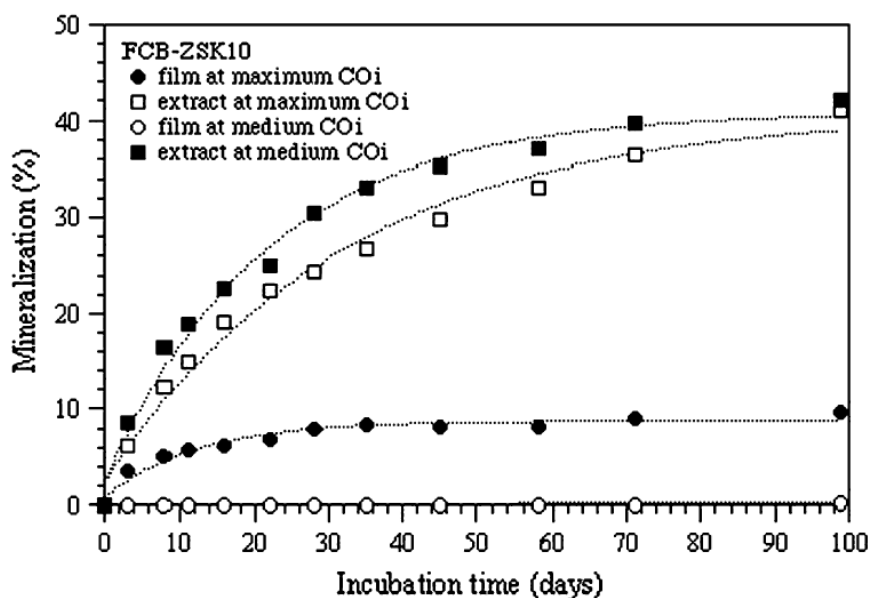
Figure 23. Biodegradation profile of thermally treated LDPE sample and its acetone extracts as reported by Chiellini et al. (2007)

In 2007, the Chico Research Foundation (CRU)[18] performed an evaluation of the compostability of environmentally degradable plastic packaging and disposable food service ware. The researchers tested several commercially available degradable plastics, including one oxo-degradable plastic bag containing EPI's oxo-degradable additive (data on the additive level was not available). The laboratory testing, performed in triplicate, showed that after 45 days of biodegradation testing in compost, a biodegradation level of 2% was obtained for the oxo-degradable plastic, which was not significantly different from the biodegradation level obtained for the conventional LDPE sample.

Next to the composting trials, CRU also tested the oxo-degradable plastic bags for biodegradation in marine water and under anaerobic conditions. After 60 days in seawater, the oxo-degradable plastic bag remained floating on the surface, showing no disintegration at all. With regard to the biodegradation testing under anaerobic conditions, also no biodegradation for the oxo-degradable plastic was measured during the 15 days test.

In 2009, the Biodegradable Products Institute (BPI) commissioned NSF International to conduct an ASTM D 5511 test at 52°C, representing biodegradation under high-solids anaerobic digestion conditions, on an oxo-degradable polyethylene bag sold by Green Genius[70]. After 60 days of testing in triplicate, no significant biodegradation was measured.

Also in 2009, Ojeda et al.[71] performed biodegradation testing on naturally weathered oxo-degradable HDPE and LLDPE samples. After 1 year of weathering, biodegradation testing was performed at 25°C and 58°C in compost and results showed approximately 5%, respectively 12% biodegradation after 3 months (based on three replicates, see Figure 24). Information on the oxo-degradable additive level used was not available.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS
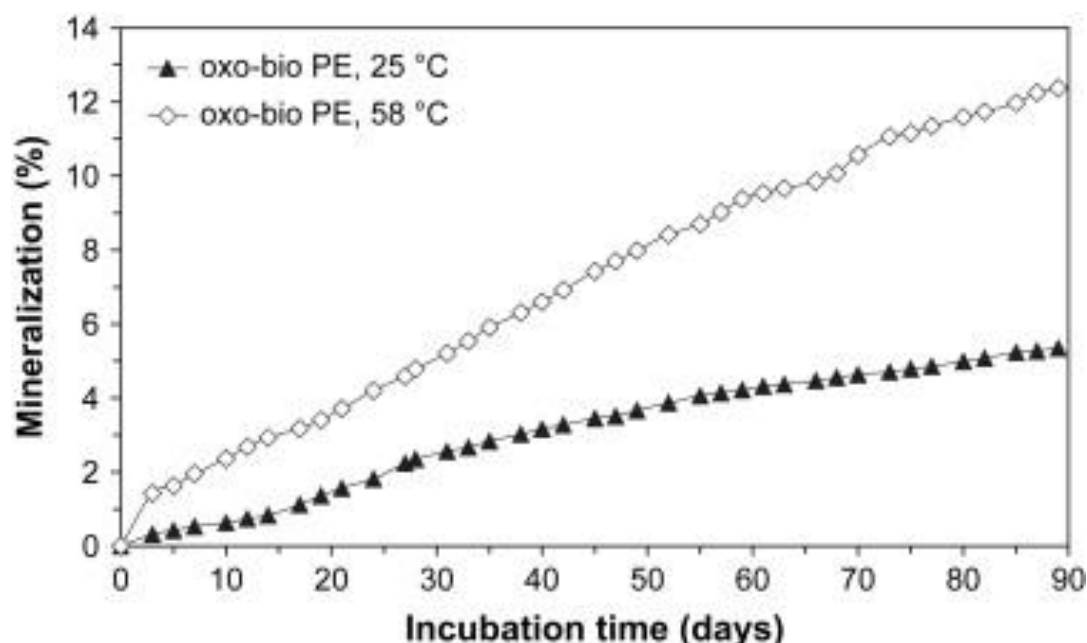
Figure 24. Biodegradation profile of a naturally weathered PE sample in compost at 28°C and 58°C, as reported by Ojeda (2009)

Further research on the biodegradation of oxo-degradable plastics in soil was performed in 2010 by Husarova[54], obtaining no more than 13-16% biodegradation, depending on the duration of (thermal) oxidation (40 or 80 days at 70°C), after more than 460 days for oxo-degradable plastics containing $CaCO_3$ as a filler (see Figure 25). The oxo-degradable plastic without filler only reached 7% of biodegradation after almost 400 days. Biodegradation testing was also performed in compost, reaching no more than 23%, respectively 19% for the oxo-degradable plastic with and without $CaCO_3$ (see Figure 26). Both the testing in compost and soil were performed using three replicates.

Similar to Husarova, also Fontanella et al.[43] measured only moderate to low biodegradation levels for oxo-degradable HDPE, LDPE and LLDPE sample which have been oxidized for a period corresponding to three years of outdoor conditions. Figure 27 shows that after 352 days in compost respectively soil biodegradation levels of 5-25% respectively 4-12% were obtained. Furthermore, it must be noted that a plateau was reached for the biodegradation test in compost, while also in soil biodegradation was starting to level off. Information on the oxo-degradable additive level was not available. Testing was done in triplicate.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS
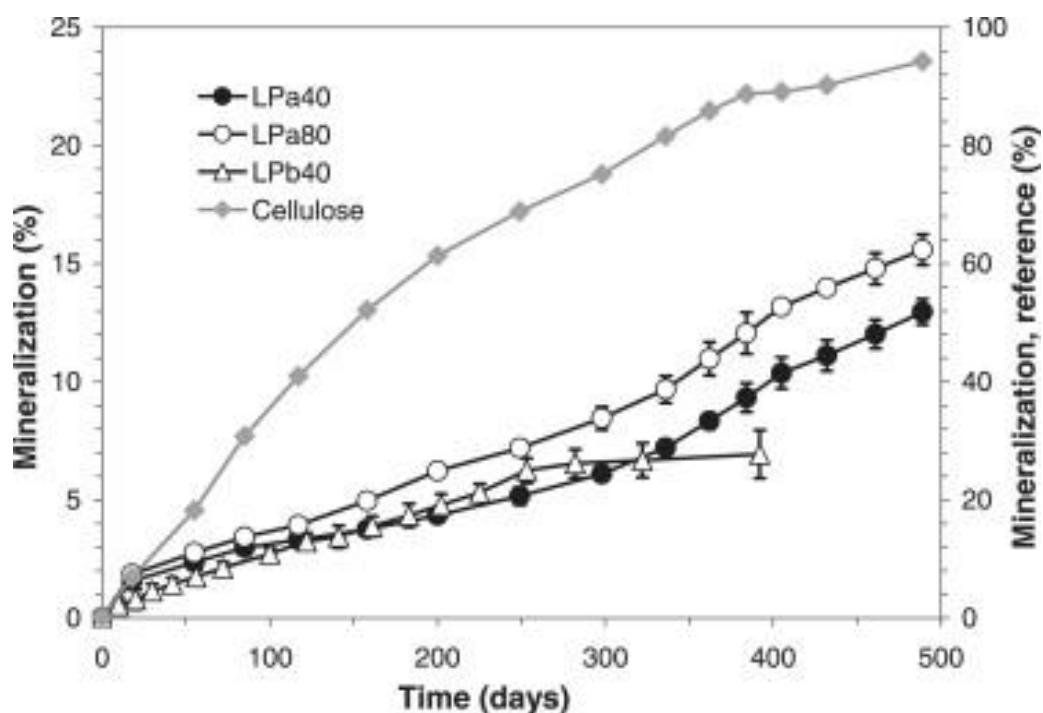
Figure 25. Biodegradation profile of thermally oxidized LDPE samples in soil at 25°C as reported by Husarova (2010). Mineralization of the cellulose is shown on the right axis.
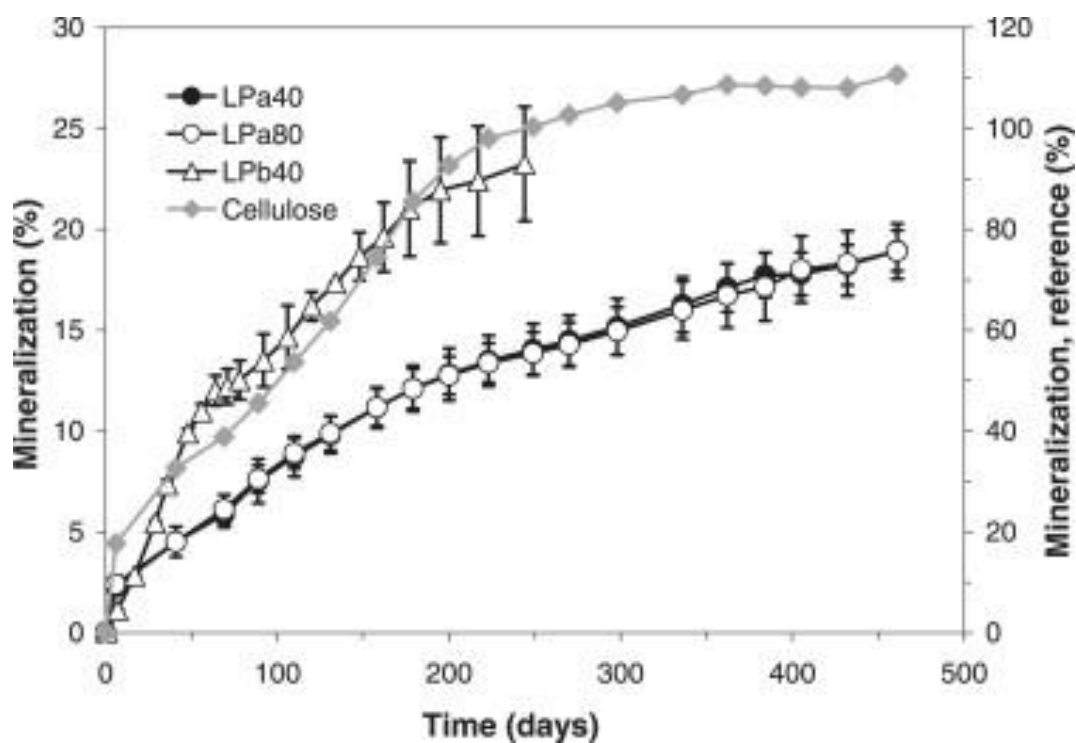
Figure 26. Biodegradation profile of thermally oxidized LDPE samples in compost at 58°C as reported by Husarova (2010). Mineralization of the cellulose is shown on the right axis.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS
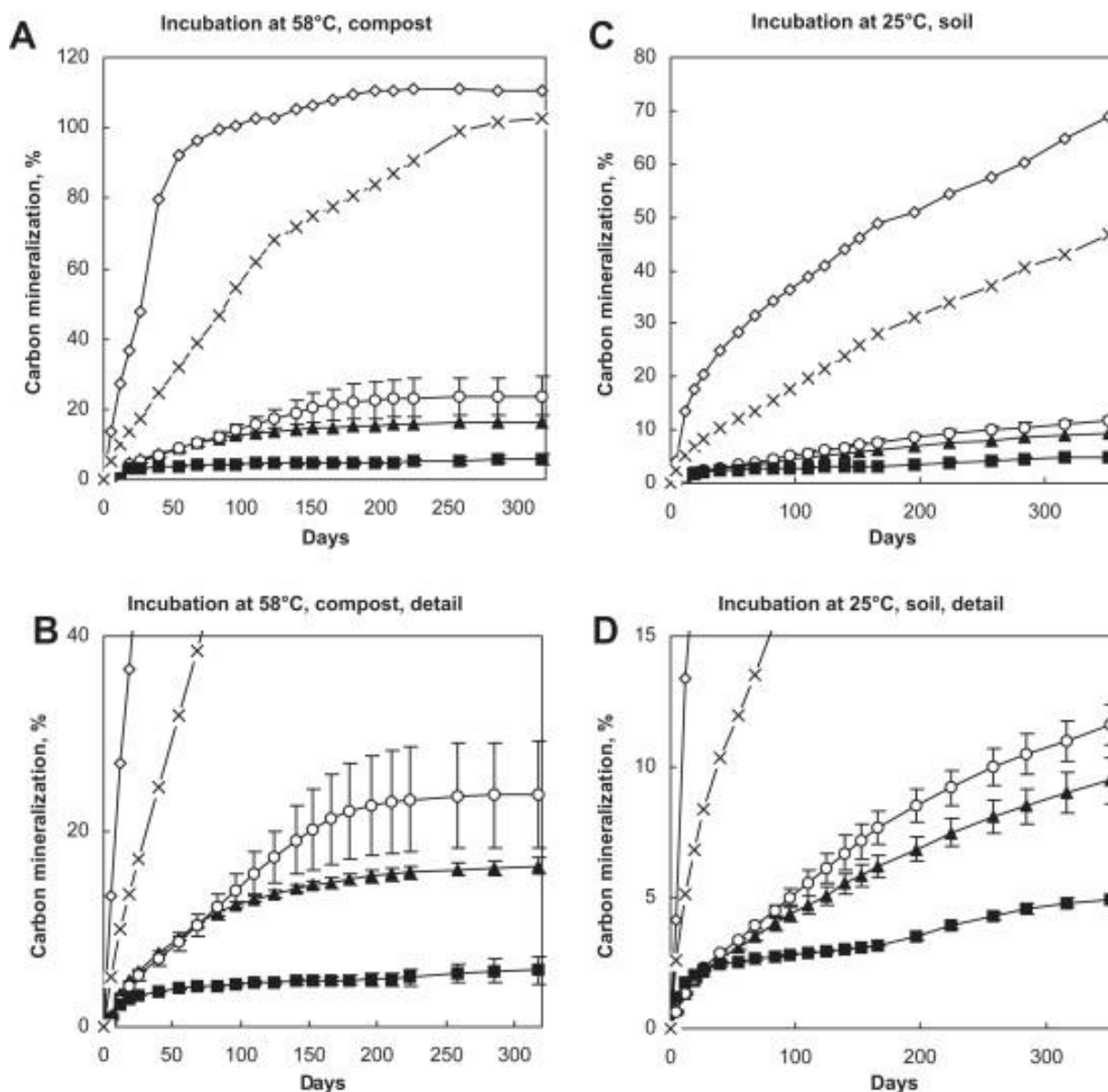
Figure 27. Biodegradation profiles of photo- and thermo-oxidized LDPE, HDPE and LLDPE samples in compost and soil, as reported by Fontanella (2010)

Also in 2010, the BPI commissioned NSF International and OWS to analyse the Aquamantra bottle produced from oxo-degradable PET[72,73]. Both test laboratories performed testing in triplicate conform ASTM D5511 for 60 days under thermophilic conditions (52°C). At the end of the test, NSF measured an overall biodegradation level of 4% (see Figure 28). OWS obtained no significant biodegradation after 60 days (see Figure 29).

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS
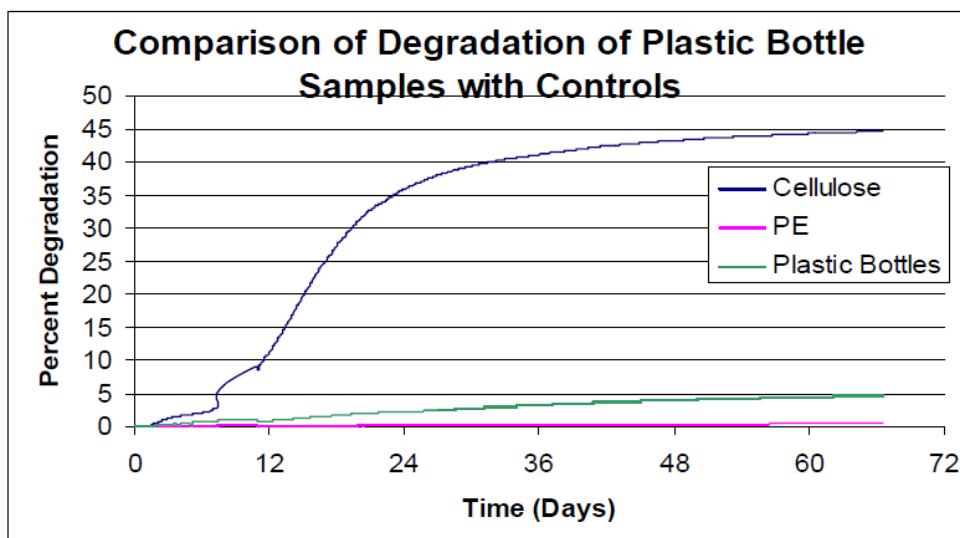
Figure 28. Biodegradation profile of an oxo-degradable PET bottle under high-solids anaerobic digestion conditions (ASTM D5511), measured by NSF in 2009
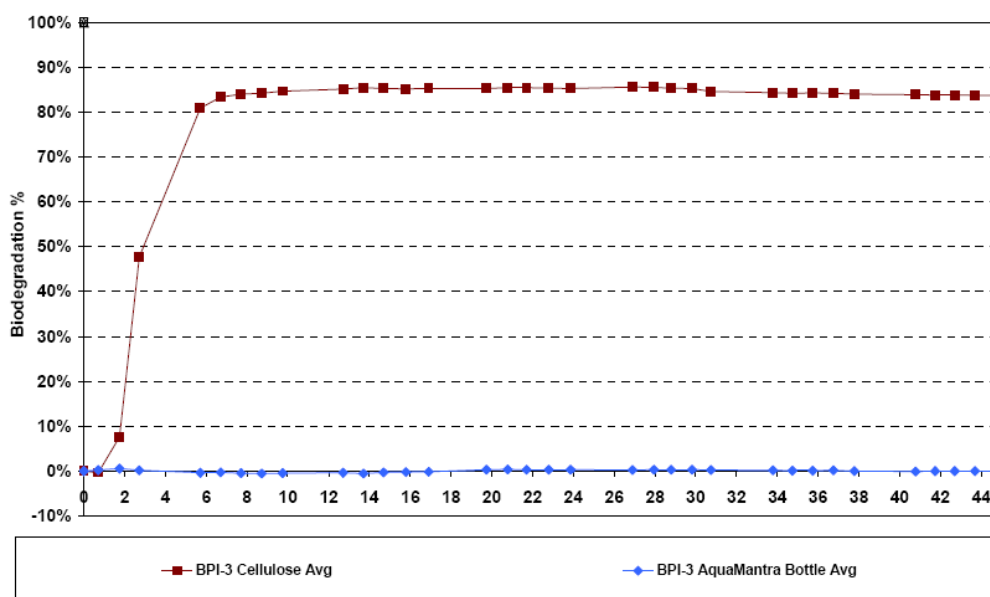
Figure 29. Biodegradation profile of an oxo-degradable PET bottle under high-solids anaerobic digestion conditions (HSAD), measured by OWS in 2009

Further research by Jakubowicz et al.[57] was performed in 2011. Thermal oxidation of an oxo-degradable polyethylene sample of P-Life Japan (10 days at 65°C) resulted in a molecular weight drop to 8,800 after which biodegradation was measured in both compost (testing in duplicate) and soil (testing in triplicate). Information on the additive loading level was not available.

After 607 days of incubation, and after a lag phase of approximately 180 days, a mean biodegradation level of 43% was obtained in compost (see Figure 30). In addition, it must be noted that biodegradation was slowly reaching a plateau at the end of the test. Furthermore,

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

the test was also characterized by a (very) large standard deviation. One replicate showed a biodegradation level of approximately 30% after 607 days, while the other replicate showed a value of approximately 50%.

Within the same time frame, 79% biodegradation was measured in soil, increasing further to 91% after 733 days, and continuing, albeit at a (much) reduced rate (see Figure 31). Compared to the biodegradation test in compost, standard deviation remained within acceptable limits.
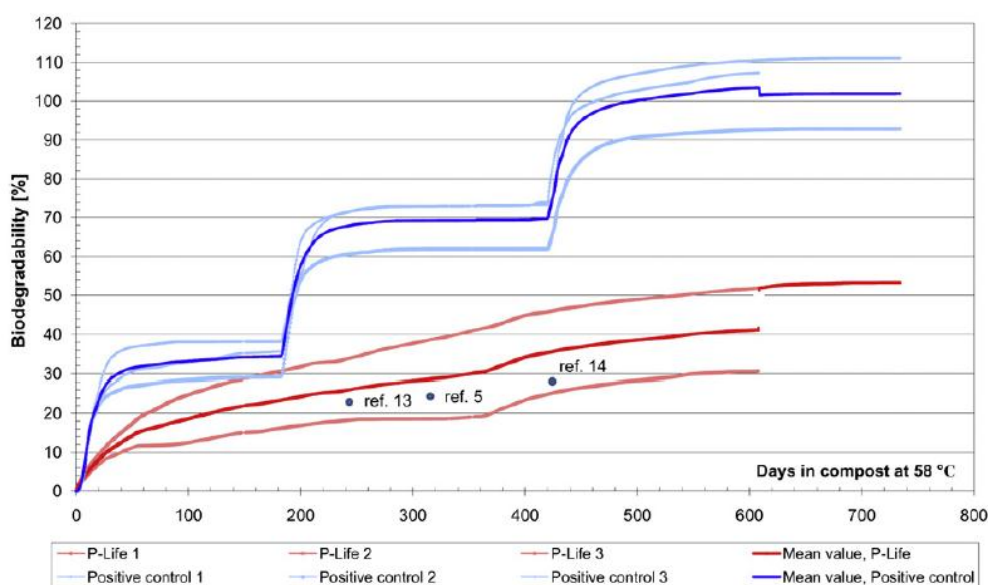
Figure 30. Biodegradation profile in compost of a thermally oxidized PE sample as reported by Jackubowicz in 2011
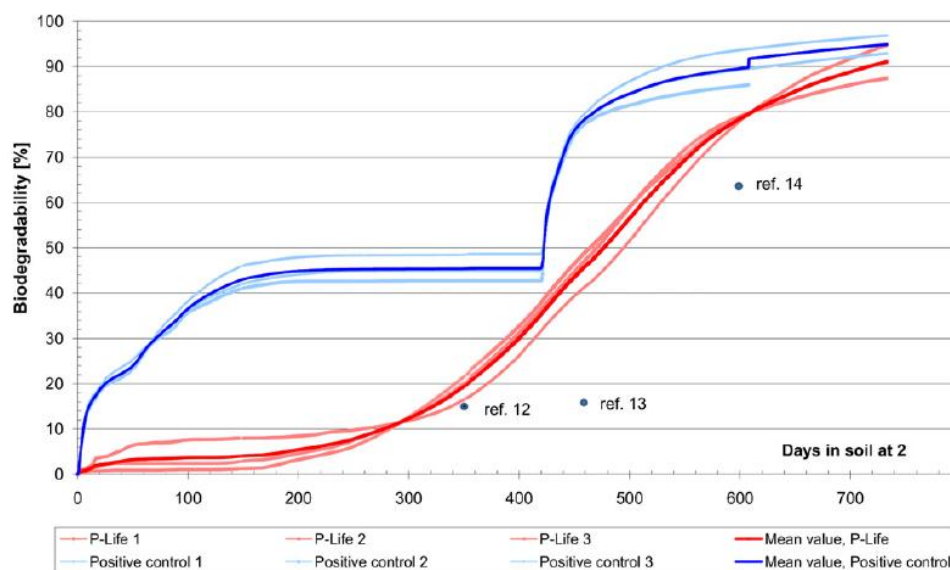
Figure 31. Biodegradation profile in soil of a thermally oxidized PE sample as reported by Jackubowicz in 2011

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

From the above results obtained by Jackubowicz, but also based on the results obtained by Chiellini in the past years, one could assume that biodegradation of oxo-degradable plastics in soil happens at a (much) higher rate when compared to compost. However, other publications, including those of Fontanella and Husarova, show the opposite. Furthermore, as compost, certainly in case of industrial composting, which is performed at elevated temperature, can considered to be more aggressive when compared soil, one would expect higher biodegradation rates in compost instead of in soil (just as with biodegradable plastics). In other words, there is an inconsistency in the biodegradation results obtained for oxo-degradable plastics.

Finally, more data can be found on the websites and in position papers of oxo-degradable additive producers, although in most cases only a short summary of the results is shown with no further information on the calculations. A few examples: Commissioned by Wells Plastics, the Chemical Industry Institute of Shihezi University performed a biodegradation test in line with ISO 14855 on a PE mulching film recovered from a field after one growing season and obtained 77% biodegradation in 45 days. A second sample from Wells Plastics, an oxo-degradable lunch box, was tested by the China National Centre for Quality Supervision and Testing of Plastic Products (NTSQP) in accordance with ISO 14855, showing a biodegradation level of 33% after 126 days. In Spain, Condis Supermercats, a local supermarket chain, has commissioned LGAI Technological Center to perform a biodegradation test conform ISO 14855 on their T-shirt bags produced by Symphony Environmental Technologies. After 67 days of testing, 82% biodegradation was measured.

For the latter testing laboratory however, other biodegradation reports and results have been found to be incorrect. More specifically, the percentage of biodegradation was not calculated correctly, resulting in a biodegradation of > 50% while it should have been zero. Therefore, without any further information on the test method used and results obtained, these results cannot be considered as valid nor reliable.

The majority of the above discussed biodegradation tests have been carried out in complex media like compost, soil, fresh and marine water, and anaerobic digestion, sampled directly from nature. However, albeit to a lesser extent, few studies have been using selected bacterial strains in (mostly) liquid cultures[74-80] to proof biodegradation of oxo-degradable plastics.

Already in 1991, Lee et al.[81] monitored the degradation of thermo-oxidized oxo-degradable polyethylene samples (up to 20 days at 70°C) in the presence of *Streptomyces* bacteria and *Phanerochaeta* fungus. While a decrease in molecular weight was recorded in the presence of bacteria, no effect was seen in the presence of the fungus. The actual level of biodegradation was not measured, as the authors stated that a decrease in molecular weight is sufficient proof of biodegradation. In 2002, Volke-Sepulveda et al.[76] incubated thermally oxidized (15 days at 80°C) additivated low-density polyethylene (LDPE) with *Aspergillus niger* and *Penicillium pinophilum*. Even though significant morphological and structural changes were observed for the LDPE after 31 months, mineralization levels of only 0.37 - 0.57% were measured. Furthermore, in 2003, Bonhomme et al.[77] also observed colonization of *Rhodococcus rhodochrous* and *Clado-sporium cladosporoides* on thermo-oxidized polyethylene, which was sufficient for the author to claim positive biodegradation.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 85 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

7. CERTIFICATION SYSTEMS

There are several certification options and accompanying logos available in the market displaying the industrial/home compostability, biodegradability or oxo-degradability of a product. The main logos and norms, if applicable, on which these certifications are based, are listed below.

7.1. Compostability

Within Europe there are several certification logos available. This is also the situation European Bioplastics is lobbying for as it eliminates a monopoly and forces the different certification bodies to continuously update their system and follow the market. On the other hand, this is not the ideal situation for producers as now different bodies need to be contacted in order to certify their products throughout Europe. Also towards communication a multitude of logos provides problems.

7.1.1. Industrial compostability

The seedling logo and the OK Compost logo are the two dominant logos in Europe. On a business-to-business level they are both equally well known, whereas on a business-to-consumer level each has geographical preferences as explained further.

7.1.1.1. Seedling logo

Organization: European Bioplastics
Norm(s):   EN 13432, ASTM D 6400, EN 14995 and ISO 17088
                       (+ more detailed certification scheme with additional rules)
Geographical value: Germany, The Netherlands, Switzerland, Poland and the UK

The property rights on the logo belong to European Bioplastics, formerly the International Biodegradable Polymers Association and Working Groups (IBAW). For many years, the management of the seedling logo was contracted to DIN CERTCO, a German certification bureau based in Berlin. At the beginning of 2012, however, European Bioplastics also assigned Vinçotte, a Belgian certification bureau based in Brussels, to manage the seedling logo together with DIN CERTCO. Products that are certified can carry the seedling logo (see Figure 32).

Figure 32. Seedling logo for industrially compostable products

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 86 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

7.1.1.2. OK Compost logo

Organization: Vinçotte (formerly AIB Vinçotte)

Norm: EN 13432 (+ more detailed certification scheme with additional rules)

Geographical value: Belgium, France, Italy, Spain and the UK
(the latter mainly for the OK Compost Home logo, see further)

The second well-known (industrial) compostability certification system in Europe is OK Compost, managed by Vinçotte based in Brussels, Belgium. Historically, the system was already launched in 1994, at the request of local governments who wanted to use compostable biowaste collection bags. Approved products can carry the OK Compost logo (see Figure 33), with a product-specific S-number.

Figure 33. OK Compost logo for industrially compostable products

7.1.1.3. DIN-Geprüft Compostable logo

Organization: DIN CERTCO

Norm: EN 13432, ASTM D 6400, EN 14995, ISO 17088 and AS 4736

Geographical value: Not yet well defined, but most probably in line with the geographical value of the seedling logo of European Bioplastics

At the end of 2012, DIN CERTCO launched a new certification system, owned by TÜV Rheinland and managed by DIN CERTCO. Approved products can carry the DIN-Geprüft Compostable logo (see Figure 34).

Figure 34. DIN-Geprüft Compostable logo for industrially compostable products

O.W.S.

Code: DSL-1

Page: 87 / 118

Date: Aug-09-2013

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

7.1.1.4. *Compostable logo*

Organization: Biodegradable Products Institute (BPI)
Norm: ASTM D 6400 and ASTM D 6868
Geographical value: USA and Canada

In the USA, a compostability certification program was started in 2000 by a joint effort of BPI, an industry organization of bioplastic producers, and the US Composting Council (USCC), representing the interests of the composting industry. The certification program is based on ASTM D 6400, covering plastic product, and ASTM D 6868, covering coated paper and natural products. Approved products can carry the compostable logo (see Figure 35).

Since January 2012, BPI has contracted the management of the logo to NSF International, a US based organization that provides standards development and product certification, but also product testing.

Figure 35. BPI-USCC logo for industrially compostable products

7.1.1.5. *Cedar Grove Composting logo*

Organization: Cedar Grove
Norm: based on ASTM D6400 and ASTM D6868 with additionally mandatory full-scale test
Geographical value: USA and Canada

In the USA, another compostability acceptance system is being operated by Cedar Grove, a composting company in Seattle, Washington, with several side activities including compostability testing and sales of compost, soil amendments as well as compostable products. On top of conformity with international standards, products need to pass full-scale testing in the Cedar Grove system. Approved products can carry the Cedar Grove Composting logo (see Figure 36).

Figure 36. Cedar Grove Composting logo for industrially compostable products

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 88 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

7.1.1.6. GreenPla logo

Organization: Japan Bioplastics Association (JBPA)
Norm: Green PLA certification scheme
Geographical value: Japan

In Japan, a certification system is being operated by the Japan BioPlastics Association (JBPA), formerly known as the Biodegradable Plastics Society, Japan (BPS). Approved products can carry the GreenPla logo (see Figure 37).

Figure 37. GreenPla logo for industrially compostable products

7.1.1.7. Australian seedling logo

Organization: Australasian Bioplastics Association (ABA)
Norm: AS 4736
Geographical impact: Australia and New Zealand

The Australasian Bioplastics Association (ABA) represents members in Australia and New Zealand. ABA runs a certification scheme based on Australian standards and consequently, industrial compostability certification is based on AS 4736, including also earthworm toxicity testing. Under license of European Bioplastics, ABA uses the seedling logo for approved products (see Figure 38).

Figure 38. Seedling logo of ABA for industrially compostable products

7.1.1.8. Others

In some other, mainly European, countries more initiatives have been taken to launch industrial compostability logos. Most of them can however mainly be considered as local initiatives.

Country: Italy
Organization: Consorzio Italiano Compostatori (CIC)
Norm: based on EN13432 with additionally mandatory full-scale test

Figure 39. CIC logo for industrially compostable products

Country: Sweden
Organization: SP Technical Research Institute of Sweden
Norm: SPCR 141 (based on EN 13432)

Figure 40. SP's logo for industrially compostable products

Country: Catalonia, Spain
Organization: *Departament de Medi Ambient i Habitatge*
                               (Department of Environment and Housing)
Norm: Unclear, but presumable based on EN13432

Figure 41. Logo of the Catalonian Department of Environment and Housing for industrially compostable products

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 90 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

7.1.2. Home compostability

The OK Compost Home logo is today the most dominant logo for home compostable products worldwide.

7.1.2.1. OK Compost Home

Organization: Vinçotte (formerly AIB Vinçotte)
Norm: Own developed certification scheme based on EN 13432
Geographical value: Europe & USA

In 2003, the Belgian certification agency Vinçotte launched the OK Compost Home certification scheme for approval of home compostable products and packaging. Since, at that time, no international standards on home composting existed, it is based on a scheme developed by Vinçotte itself. The scheme is largely based on the requirements as set forth by EN 13432, but has been adapted to ambient temperature. Similar to the OK Compost logo, approved products can carry the OK Compost Home logo (see Figure 42), with a product-specific S-number.

Figure 42. OK Compost Home logo for home compostable products

The OK Compost Home certification scheme has in the meantime been adopted in Californian legislation as well. Senate Bill No. 567, published on October 8, 2011, states that, as of January 1st, 2013, plastics products can only be labelled 'home compostable' if they meet the requirements of the OK Compost Home scheme as set forth by Vinçotte[82].

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 91 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

7.1.2.2. DIN-Geprüft Home Compostable logo

Organization: DIN CERTCO
Norm: AS 5810
Geographical value: Not yet well defined, but most probably in line with the geographical value of the seedling logo of European Bioplastics

In parallel with their certification system for industrially compostable products, launched at the end of 2012, DIN CERTCO also construed a similar certification system for home compostable products. Approved products can carry the DIN-Geprüft Home Compostable logo (see Figure 43).

Figure 43. DIN-Geprüft Home Compostable logo for home compostable products

7.1.2.3. Australian Home Compostable logo

Organization: Australasian Bioplastics Association (ABA)
Norm: AS 5810
Geographical impact: Australia and New Zealand

In 2010, the first standard on home compostability was published in Australia. AS 5810 is based on AS 4736, the Australian standard for industrial composting, but has been adapted to ambient temperature. Shortly after, the ABA also created an accompanying logo for approved products (see Figure 44).

Figure 44. Australian Home Compostable logo for home compostable products

7.1.2.4. Others

Similar to the certification systems and logos created for industrially compostable products, some other countries have also been taken initiatives to launch home compostability logos.

Country: UK
Organization: Organics Recycling Group – Renewable Energy Association (REA)
Norm: OK Compost Home certification scheme, adopted in 2011

Figure 45. REA's logo for home compostable products

Also in the USA, the OK Compost home scheme has been taken over by the Sustainable Biomaterials Collaborative (SBC) for their specifications on home compostability. Furthermore, in 2011, OK Compost Home was referred to in Californian state legislation.

7.2. Biodegradation in other environments

Certification systems for other environments besides composting are much less developed. Only the Belgian certification bureau Vinçotte and the Swedish Technical Research Institute SP have created some systems.

Vinçotte has created two certification systems, both under the header of OK Biodegradable but with additionally the environment specified, e.g. OK Biodegradable Soil and OK Biodegradable Water (see Figure 46).

Figure 46. OK Biodegradable Soil and OK Biodegradable Water logo

OK Biodegradable Soil is based on a scheme developed by Vinçotte itself and includes requirements with regard to biodegradation, chemical characteristics and ecotoxicity. Disintegration is not included as for applications in soil it supposed to be a product requirement instead of an environmental requirement. OK Biodegradable Water is mainly based on EN 14987.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 93 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

SP Technical Research Institute developed, as a part of their certification system for industrially compostable products, also a system for biodegradable products. The system includes requirements with regard to biodegradation and chemical characteristics which is identical to those for industrially compostable products. Approved products can carry SP's logo for biodegradable products (see Figure 47).

Figure 47. SP's logo for biodegradable products

7.3. Oxo-degradation

7.3.1. Emirates Authority for Standardization and Metrology (ESMA)

At the beginning of 2012, the United Arab Emirates' Ministry of Environment and Water (MoEV) made it mandatory to use oxo-degradable technology for plastic carrier bags. At the beginning of 2013, the MoEV extended the range of plastics products to include almost all disposable plastic products made of PE, PP or PS, covering, but not limited to, flexible shopping bags and semi-rigid plastic packaging for food, magazines, consumer-durables, garbage bags, bin-liners for household use, shrink wrap, pallet wrap, cling film and other plastic articles normally used over short periods and subsequently discarded. Plastic products which are locally produced and those that are imported into the UAE do now need an ECAS (Emirates Conformity Assessment Scheme) Registration Certificate issued by the Emirates Authority for Standardization and Metrology (ESMA).

The plastic products must contain oxo-degradable additives from suppliers which have been audited by ESMA (see Figure 48) and need to comply with UAE.S 5009:2009. End products that receive a certificate can bear a mark of conformity as shown in Figure 48.

Figure 48. ESMA's mark of conformity for oxo-degradable plastics

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

The few certified products, which are based on the additives of Symphony Environmental Technologies and Wells Plastic, are also listed on the website of ESMA and for each product reference is being made to a test report and test facility. The test facility is in most cases the additive producer itself or one if its affiliates. Conform UAE.S 5009:2009, testing needs to be performed in an accredited laboratory, but the standard does not state anything on the need for independent third party testing.

For a few cases, also the test reports are available, but these only cover the abiotic degradation part, not the biotic degradation. Furthermore, while weathering was performed at 50°C, extrapolation was applied in order to calculate the rate of degradation (fragmentation) at 30°C using the Arrhenius equation.

In other words, even though several products are certified, data to support these certificates is not available on the website.

Table 16. List of ESMA certified oxo-degradable additive producers

Company	Logo/Brand
Symphony Environmental Technologies Plc	
Wells Plastic Ltd	
Willow Ridge Plastics	
EPI Environmental Products Inc	
EnerPlastics LLC	

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 95 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

7.3.2. Oxo-biodegradable Plastics Association (OPA)

The UK based Oxo-biodegradable Plastics Association (OPA) is an industrial association active in the field of oxo-degradable plastics (see also 6.5). In this context, OPA offers certification to and permits its logo to be used in respect of plastic products which are shown by reports of testing institutions approved by the OPA, to be oxo-degradable, oxo-biodegradable and non ecotoxic (see Figure 49).

For the definition of oxo-degradation and oxo-biodegradation, OPA refers to TC 249/WG 9 of CEN which defines oxo-degradation as 'degradation identified as resulting from oxidative cleavage of macromolecules' and oxo-biodegradation as 'degradation identified as resulting from oxidative and cell-mediated phenomena, either simultaneously or successively'. OPA does not make any reference to one or more guidelines and/or standards on which certification is based and does not seem to have a certification scheme with specifications and rules.

Figure 49. OPA's logo for oxo-degradable plastics

7.3.3. SP Technical Research Institute of Sweden

At the end of 2009, the SP Technical Research Institute of Sweden published SPCR 141, a set of rules for the classification for treatment of polymeric waste (see also 6.3.5). Based on SPCR 141, Appendix 4, which focusses on polymeric waste degradable by abiotic and subsequent biological degradation, SP also developed an accompanying certification scheme.

Certification can be obtained for prodegradant systems and masterbatches, materials and final products. Approved items can be marked with SP's logo for oxo-degradable plastics (see Figure 50) along with a clarifying text.
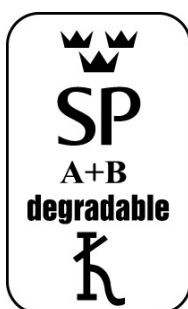
Figure 50. SP's logo for oxo-degradable plastics

Currently 1 prodegradant additive is certified by SP: SMC 2522 from the Japanese company P-Life Japan Inc.

7.3.4. Biosystems America

Biosystems America is a US based independent testing facility with activities centred around research and development on biological systems. The division based in Atlanta performs, amongst others, testing conform ASTM D 6954 and has, in parallel, also set up a certification of product performance for materials and products which pass the different criteria of ASTM D 6954 in their laboratory. An accompanying logo was not designed.

Research learns that today, only a couple of products have been certified by Biosystems Atlanta and should therefore, according to Biosystems Atlanta, comply with ASTM D 6954. Certified products are three products from Intertape Polymer Group (stretch films Genesys®, ProLite^{TM} and Handwrap IV – no information on the used additive) and one product from Wells Plastics (LDPE film with Reverte – reference SR3340).

7.3.5. Singapore Green Labeling Scheme

The Singapore Green Labeling Scheme (SGLS), administered by the Singapore Environment Council and member of the international Global Ecolabeling Network (GEN), was launched in 1992 to endorse consumer products and services that have less undesirable effects on the environment. SGLS has a list of product categories covering a wide variety of applications. Products than comply with SGLS can carry the Singapore Green Label (see Figure 51).

Figure 51. Singapore Green Label

Currently, SGLS is working on a new product category for oxo-degradable polypropylene and polyethylene products. The certification will not be based on a standard, but sets forth its own set of criteria:

- Abiotic degradation (conform ASTM D 5208 and ASTM D 5510):
    o Molecular weight < 10,000 Dalton
    o Elongation at break of 5% or less (for at least 75% of the test samples)
    o Gel fraction < 5%

- Biotic degradation:
    o The scheme assumes that if the material has shown a molecular weight < 10,000 Dalton, the material is no longer a plastic and has become inherently biodegradable (even though there is no scientific proof for this). Biodegradation testing is therefore not needed (as it is time-consuming and expensive, dixit the scheme)

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 97 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

- Heavy metal content:
  - The limit values as set forth by EN 13432 and as shown in Table 3 may not be exceeded

- Ecotoxicity:
  - Testing shall be carried out according to OECD 207 (earthworm toxicity testing) and OECD 208 (plant toxicity testing)

Timescales are not prescribed as the composition of the oxo-degradable plastics is designed to be variable and conditions in the open environment are also variable.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 98 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

8. DISCUSSION & CONCLUSIONS

Known end-of-life treatments of plastics include: re-use, recycling, composting (biodegradation), energy recovery and landfill.

Biodegradable plastics are not a solution for littering. In order to benefit from the advantages of biodegradability, these plastics should be properly disposed of in the appropriate (composting) facilities.

According to the oxo-degradable industry, the most likely disposal routes for oxo-degradable plastics are recycling, soil surface exposure (through littering and the use of mulching films) and landfilling, although water exposure seems to be a possible disposal route as a result of littering.

Professor Gerald Scott, chairman of the Scientific Advisory Board of the Oxo-biodegradable Plastics Association (OPA), clearly states that oxo-degradable plastics are not designed for degradation deep in **landfill**[83-84]. If oxo-degradable plastics were to end up in landfill, they would, according to OPA, disintegrate in the surface layers of the landfill as long as oxygen is present. Oxo-degradable plastics therefore only degrade in the upper layer of landfills and emit CO_2 at a low rate, but remain inert in the absence of oxygen and therefore do not exert CH_4, which is more harmful for global warming[85].

OPA also claims that, biodegradable plastics on the other hand biodegrade both in the surface layers of a landfill, emitting CO_2 at a high rate, if there is enough microbial activity, and in the depths of a landfill, in the absence of air, generating CH_4. However, as discussed above (see 5.4), it must be noted that the family of biodegradable plastics comprises a broad range of different materials from which some have been tested for their biodegradation behaviour in landfill. An accelerated landfill test (ASTM D5526) conducted on a starch based film showed that biodegradation in landfill is only triggered at a 65% moisture content, while landfills are typically being operated at a humidity level of 25%. Furthermore, for the reference material cellulose no biodegradation was observed at a 40% moisture level[86]. A similar study, also based on ASTM D5526 testing, was performed on PLA and showed that PLA remains as inert in landfill as conventional plastics[87]. In other words, while ASTM D5526 testing is performed under optimal conditions, simulating 'highly managed' landfills with biogas recuperation and leachate recirculation, the majority of landfills is unmanaged having conditions for (bio)degradation far from optimal with a low moisture content that does not allow biodegradation.

OPA's extrapolation of positive biodegradation results on biodegradable plastics obtained under aerobic conditions to anaerobic conditions is incorrect. As explained earlier, the microbial population in anaerobic conditions differs significantly from that of aerobic conditions, as such strongly affecting the biodegradation behaviour of materials.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

OPA also states that oxo-degradable plastics are not designed for anaerobic digestion and are normally not marketed for **composting** as they are not meant to be collected together with the organic waste fraction[84]. However, composting is especially interesting for mixed waste streams in which plastics are co-mingled with wet organic waste and according to Professor Telmo Ojeda, member of the Scientific Advisory Board of OPA, oxo-degradable additives are mainly applied in thin PE and PP packaging and are therefore often contaminated with food and organic materials[88].

As explained in 7.1.1 and 7.1.2, several certification systems exist for industrial and home composting and a large number of biodegradable plastics have been tested by independent third parties and are certified industrial and/or home compostable. Despite what is sometimes claimed, oxo-degradable plastics on the other hand are not compostable, as agreed upon by an important share of the industry, including its representing association OPA, as they do not meet the requirements of industrial and home compostability as set forth by the different standards.

According to OPA, oxo-degradable plastics do indeed not degrade (quickly) in low temperature windrow composting, but developments would be on-going for in-vessel composting which is claimed to be run at higher temperatures[89]. It is correct that there is a higher temperature ingredient from outside to inside in windrow systems, but this is partially avoided via frequent turning. While in-vessel systems reach the optimum temperature immediately, windrow systems have to cope with a lag phase of 1-2 days first. Nevertheless, the highest temperatures are still reached in uncontrolled windrow systems (70°C and higher), while in-vessel systems are usually operated at 60-65°C, be it for a longer period. In other words, and assuming temperature plays a key role in the composting of oxo-degradable plastics, one would assume compostability to be better in windrow composting instead of in in-vessel composting.

It seems the industry contradicts itself when it comes to the behaviour of oxo-degradable plastics in industrial composting conditions. While some are claiming compostability, others openly state that oxo-degradable plastics are not compostable. Based on the present literature study, it can unequivocally be concluded that oxo-degradable plastics do not comply with any of the available standards on industrial compostability and are therefore not compostable. This means that oxo-degradable plastics cannot be used for compostable food service applications.

As oxo-degradable plastics are often, misleadingly, promoted as compostable, chances are relatively high that they will end up in industrial composting plants after all. Being non-compostable, (pieces of) oxo-degradable plastics will be sieved out at the end of the composting process, together with the other rejects. Rejects are usually composed out of branches, twigs and other thick organic materials which are internally recycled to the beginning of the process. However, if these rejects would become contaminated with oxo-degradable plastics, they can no longer be used for internal recycling and would need to be landfilled (which is much more expensive).

A similar discussion is taking place for biodegradable plastics as well. Whereas the European standard EN 13432 on industrial compostability prescribes that at least 90% of the biodegradable plastic should pass through a sieve of 2 mm after 12 weeks, the more advanced 'high-tech' composting installations use (much) shorter composting periods, claiming that this is too short to treat compostable products. Biodegradable plastics, but also other natural products like corn cobs, twigs and hedge trimmings will not be composted after only a few weeks and will require a second turn in the industrial composting facility to complete their composting process. Furthermore, whereas biodegradable plastics are certified at their maximum thickness, just disintegrating sufficiently within the prescribed 12 weeks period, in reality, the vast majority of the biodegradable plastics appearing on the market have a (much) lower thickness, as such disintegrating (much) faster and in a (much) shorter timeframe.

From a testing point of view, the oxo-degradable plastics industry questions the reliability of the currently available test methods for the determination of the **biodegradation**, including ISO 14855 (for compost), ISO 17556 (for soil) and ISO 14851 (for water). More specifically, it is claimed that the CO_2 production is not the correct parameter for the determination of the mineralization level of oxo-degradable products as biodegradable plastics emit CO_2 at a fairly high rate, while oxo-degradable plastics, and many natural lignin containing materials according to the oxo-degradable plastics industry, biodegrade at a (much) lower rate. However, (ultimate) biodegradation is still defined as the breakdown of an organic compound by micro-organisms to CO_2, water, mineral salts and new biomass. In other words, CO_2 production is the only (correct) parameter for the quantification of biodegradation, even if biodegradation takes several months or years.

Yet, many natural lignin containing materials have been tested successfully for biodegradation using either ISO 14855 or ISO 17556 (see Table 17 and Table 18). The majority of these tests were characterized by a long testing period, up to 2.5 years, during which CO_2 production was continuously measured. Based on these results it can be concluded that natural lignin containing materials do indeed degrade at a (much) lower rate when compared to biodegradable plastics and that, despite the low CO_2 production rate, these test methods do result in reliable data on biodegradation, even on long term.

Table 17. Biodegradation results for natural lignin containing materials obtained after testing conform ISO 14855 in compost

Source	Sample	Result
EU project contract no. 212239[12]	Wood fibres	71.1% in 728 days
EU project BIOPAL-CT-2002-02431[90]	Oak leaves	65.1% in 124 days
EU project FAIR-CT98-3919[14]	Flax fibres Flax fibres	63.6% in 225 days 91.3% in 60 days
EU project AIR2-CT93-1099[91]	Birch leaves Oak leaves Pine needles	75.8% in 365 days 58.4% in 365 days 51.8% in 365 days

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 101 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Table 18. Biodegradation results for natural lignin containing materials obtained after testing conform ISO 17556 in soil

Source	Sample	Result
EU project Contract no. 212239[12]	Wood fibres	50.2% in 134 days
	Wood fibres	69.7% in 664 days
EU project BIOPACK-CT-2000-00799[11]	Oak leaves	91.1% in 902 days
	Oak leaves	50.0% in 220 days
EU project FAIR-CT98-3919[14]	Straw	66.4% in 270 days
	Flax fibres	94.8% in 365 days
	Broom fibres	91.8% in 365 days
EU project AIR2-CT93-1099[91]	Birch leaves	56.2% in 365 days
	Oak leaves	55.8% in 365 days
	Pine needles	61.9% in 365 days

In addition to the lower CO_2 production rate, oxo-degradable plastics are also claimed to have a lower tendency to get converted to CO_2 (and a higher tendency to get converted into biomass). However, evidence of such a higher degree of bio-assimilation has not yet been generated.

Besides the conversion of solid carbon to gaseous carbon dioxide, also microbial colonization or biofilm formation is used to quantify biodegradation. Based on results obtained by Volke-Sepulveda et al.[76], it must however be concluded that microbial growth is no proof of mineralization. Test methods based on microbial colonization or biofilm formation were initially developed to determine bio-resistance of materials and products, not to determine biodegradation or mineralization. A blend of 95% conventional polyethylene and 5% starch will also show significant microbial growth, but the conventional polyethylene part will not biodegrade. Furthermore, as a set of specific microbial species does not represent real-life conditions, these results cannot be considered as representative for environmentally relevant conditions.

Finally, also the change in physical properties and especially the decrease in molecular weight is often used as proof of biodegradation. As discussed under 6.3 and as shown in Table 10, several standards and guidelines on oxo-degradable plastics prescribe a molecular weight level equal to or lower than 10,000 Dalton (SPCR 141) or 5,000 Dalton (ASTM D6954 and UAE.S 5009) for the **abiotic degradation** phase. It is assumed that levels below 10,000 Dalton, respectively 5,000 Dalton, would 'trigger' biodegradation, while for values above 10,000 Dalton, respectively 5,000 Dalton no biodegradation would/could occur (although, conform OPA, this minimum value would be 40,000 Dalton).

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Chiellini et al.[69] did some further research in 2007 on the effect of molecular weight on the biodegradation behaviour and obtained biodegradation levels of 42-48% after 100 days in fresh water for the low molecular weight extracts of thermally treated additivated LDPE (70°C for 25 days). These extracts had a molecular weight of about 1,000 Dalton, or a factor 5 to 10 below the limit level as prescribed by SPCR 141, ASTM D6954 and UAE.S 5009. The complete material, including the low molecular weight extracts, showed a mean molecular weight of approximately 4,500 - 5,000 Dalton and only reached a biodegradation level of 12% after 100 days. Molecular weight extracts between 7,500 and 10,000 Dalton showed no significant biodegradation.

It can indeed be concluded that the biodegradation rate increases with a decreasing molecular weight. However, it can also be concluded that a decrease to a mean value of 5,000 or even 10,000 Dalton cannot be considered as proof of biodegradation. After 25 days at 70°C, Chiellini et al.[69] only reached a mean molecular weight value of just below 5,000 Dalton while only partial biodegradation was measured (which can most probably be linked to the low molecular weight extracts). Further research was done by Jackubowicz[61], who stated that the temperature is the most important factor. The average molecular weight value of additivated PE dropped below 5,000 after 2 weeks at 70°C and after 8 weeks at 60°C. Oxidation beyond respectively 2 weeks at 70°C and 8 weeks at 60°C did nonetheless not yield in a further decrease in molecular weight. In other words, the molecular weight of a plastic decreases over time when exposed to (very) high temperatures, but there is no proof that this continues below 5,000 Dalton.

Even if molecular weight would continue to decrease over time, this only happens under very specific conditions. As can be seen from Table 15, the majority of the authors used temperatures ranging from 55°C to 70°C. In addition, these (very) high temperatures were also maintained for relatively long periods, ranging from 44 days (at 55°C) to 80 days (at 70°C). Time-temperature superposition principles have been established in the last years as a methodology to translate these accelerated conditions to real-life conditions and the most referenced principle is the Arrhenius principle, although Celina et al.[60] proved that there is no guarantee that the overall behaviour is of an Arrhenius form. Nevertheless, using this equation, Jackubowicz[61] calculated that a thermally oxidized PE sample which obtained a molecular weight of 10,000 Dalton within approximately 3 days, 8 days and 44 days at respectively 70°C, 60°C and 50°C, would reach the same value but at 25°C after only 2.5 years (during which the small oxo-degradable plastic fragments can be ingested by invertebrates, birds, animals or fish[30,92,93,94]). It must furthermore be noted these figures are based on the assumption that there is a linear correlation between molecular weight and temperature over a wide range of temperature and not only over a smaller partial range, which cannot be guaranteed. Evidence of the decrease of molecular weight of oxo-degradable plastics at ambient temperature is not available.

Besides the (very) high temperatures, abiotic oxidation is often also performed at dry conditions, even though Day et al.[40] and independent testing at AMC[41] showed that moisture inhibits, or at least significantly decreases, the oxo-degradation process. The conditions at which abiotic oxidation is mimicked are therefore not representative for real-life conditions, especially when taking into account humid conditions like compost and fresh and marine water.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

In addition to the temperature and moisture content, also the loading rate at which the oxo-degradable additive is used in the plastic plays an important role in the rate of biodegradation. Only few articles reported on the additive loading rate, which varied from 1-10%. It is therefore unclear whether the loading rate of samples used for laboratory testing equals the loading rate of samples used for the market.

It must be concluded that the rate and level of (bio)degradation of oxo-degradable plastics is at least questionable. The abiotic degradation is mostly mimicked using conditions which are not representative for real-life conditions, while for the biotic degradation some positive results were obtained in literature, while others reported no or only a (very) low level of (bio)degradation. Furthermore, the positive biodegradation results obtained could not be repeated under the same conditions, not by the same author, not by other authors.

Nevertheless, despite the irreproducibility, several oxo-degradable plastics are certified by ESMA conform UAE.S 5009, which means that these products were tested by an independent and accredited laboratory and reached a molecular weight level of 5,000 Dalton or lower within 4 weeks and a biodegradation value of at least 60% within 6 months. Yet, the most promising results found in literature, showing 91% biodegradation in soil after 2 years, only reported a molecular weight value of 8,800 and a biodegradation percentage of only 5% after 6 months[57]. Also Chiellini & Corti[67,68], who reached biodegradation levels of > 60% in 1.6 years, only reached 4-7% biodegradation within 6 months.

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

In general, following conclusions can be drawn:

1. Biodegradable plastics:
   - The majority do meet the requirements of industrial composting standards, while others biodegrade in other environment as well;
   - Solid proof of biodegradation is available through certification by accredited laboratories;
   - Based on raw materials used, 5 categories can be distinguished: starch based, cellulose based, chemically synthesized, produced by bacteria and fossil based;
   - Standards on specifications are well established for industrial composting, but less developed for home composting and biodegradation in other environments.
   - Biodegradation takes place through biological activity, more in particular enzymatic, microbial and/or fungal activity;

2. Oxo-degradable plastics:
   - Oxo-degradable plastics do not meet the requirements of industrial and/or home compostability set out in different standards (Oxo-biodegradable Plastics Association states that they are not marketed for composting);
   - Conventional plastics additivated with transition metal salts;
   - Since 2009 strong increase in number of standards and guides, although no consistency in content and pass levels (if available);
   - (Bio)degradation claimed to be initiated by oxygen (but inhibited by moisture) and accelerated by UV light and/or heat;
   - Very few positive biodegradation results obtained (those could not be repeated under the same conditions, not by the same author, not by other authors);
   - No proof of Arrhenius' time-temperature superposition principle at wide range of temperature, which makes extrapolation from abiotic degradation at elevated temperature to real-life conditions scientifically incorrect;
   - Alternative methods (carbonyl index, molecular weight, microbial growth, ADP/ATP,…) no proof of complete biodegradation, only proof of biological activity;
   - Not compostable: better term 'thermo- or photo-fragmentable plastics'?
   - Different certification institutes, but not always independent or transparent.

3. Enzyme-mediated degradable plastics:
   - Very few data available on biodegradation;
   - Conventional plastics additivated with organic additives;
   - (Bio)degradation claimed to take place through enzymes excreted by micro-organisms during the consumption of the additive;

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 105 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

9. SUGGESTIONS FOR FURTHER RESEARCH

As discussed in chapter 6.7, numerous results are already available on the biodegradability of oxo-degradable plastics. Nevertheless, results are not always consistent as both positive and negative results were obtained. Furthermore, in case of a positive outcome, results could not be confirmed by other authors or laboratories. In this context, the following laboratory testing could be suggested for further research:

9.1. Effect of molecular weight on biodegradation

As already extensively discussed above, oxo-degradable plastics are assumed to become biodegradable once the molecular weight level drops below a certain threshold value. According to the US guideline ASTM D6954 and the United Arab Emirates standard UAE.S 5009, this threshold limit should be 5,000 Dalton, although the Swedish standard SPCR 141 refers to 10,000 Dalton.

Figure 52 shows a common profile of the molecular weight decrease over 90 days. The blue solid line is characterized by a fast decrease during the first days and is levelling off towards the end. The majority of the above discussed articles stopped the abiotic degradation phase after a period varying from 10 to 80 days, but only a few obtained molecular weight values below 10,000 Dalton (see red line in Figure 52), respectively 5,000 Dalton. Nevertheless, according to the oxo-degradable plastics industry, the molecular weight will decrease further over time and eventually will reach a value (far) below 5,000 Dalton and lower (see blue dotted line in Figure 52).
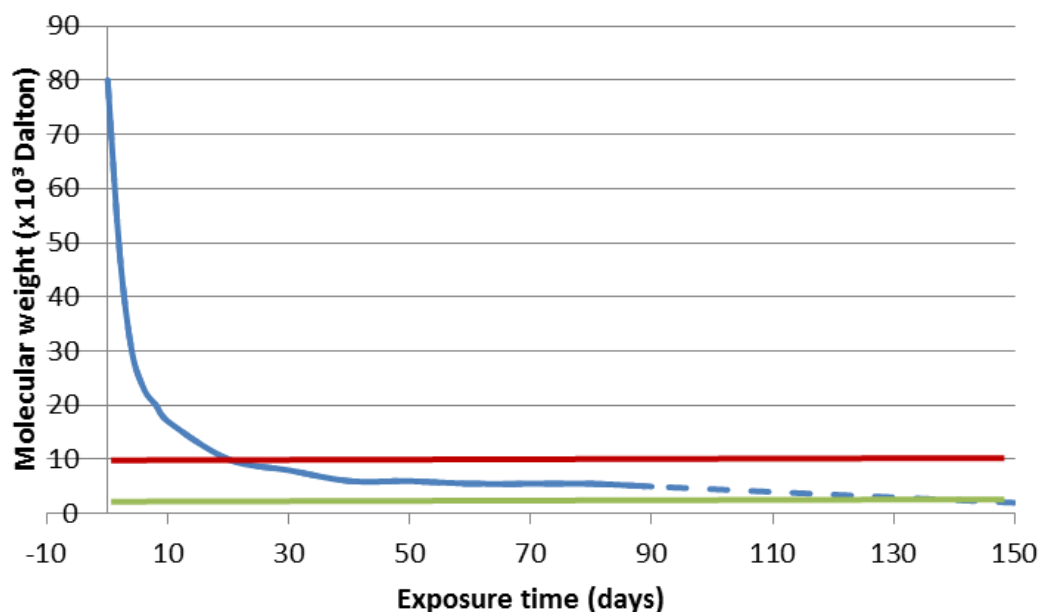
Figure 52. Molecular weight decrease of oxo-degradable plastics during abiotic pretreatment

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 106 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

Based on the data shown in chapter 6.7, it can however not be concluded that the material becomes completely biodegradable once a molecular weight value of 10,000 Dalton, respectively 5,000 Dalton or lower is reached. Therefore, it seems that this threshold value is product dependent or has a value (much) lower than 5,000 Dalton (see green line in Figure 52).

Secondly, it can also not be guaranteed that the molecular weight will continue to decrease, as shown by the blue dotted line in Figure 52, but if so, it can be questioned whether it reaches a value below the threshold value or not.

In other words, two main questions remain:
- Does the threshold value exist and if so, what is this value?
- Does the molecular weight decreases below the threshold value?

In order to provide an answer to the above two questions, the following biodegradation testing program could be performed:

(1) Phase 1:
The abiotic degradation phase could be extended until no further decrease in molecular weight is monitored. During the oxidation phase, samples can be retrieved from the weathering chamber and used for biodegradation testing. As such, abiotically degraded oxo-degradable materials of different molecular weights, starting from 5,000 Dalton and lower, can be tested in parallel for biodegradation.

(2) Phase 2:
The disadvantage of the testing proposed in Section 1 is that the molecular weight will, after a certain period of oxidation, reach its lowest value possible and we cannot be sure whether this value lays above or below the threshold value. Therefore, oxo-degradable plastic material with a varying molecular weight value, starting from 500 Dalton up to the lowest molecular weight value measured in Phase 1, could be produced and tested for biodegradation.

9.2. Biodegradation based on $^{14}CO_2$ measurements

As discussed in chapter 6.7.2, several methods exist to quantify the biodegradation of a plastic product. Some authors refer to the change in physical properties, others to the decrease in molecular/biomass weight and also the ADP concentration in the micro-organisms is nowadays used to measure biodegradation. Nevertheless, the conversion of organic C to CO_2 is the most direct and therefore only correct measurement.

However, the oxo-degradable plastics industry questions whether this latter method is indeed the correct method. Furthermore, they also state that the 90% pass level is too high as they claim that, when compared to biodegradable plastics, a (much) larger share of the plastic is converted into biomass instead of in CO_2.

In order to verify this, an oxo-degradable plastic could be produced from ^{14}C and aged until a constant molecular weight is obtained. Afterwards, the residual material can be collected for subsequent biodegradation testing during which the amount of emitted $^{14}CO_2$ can be calculated, and as such also the percentage of biodegradation.

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 108 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

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O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 109 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

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O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 110 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

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O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 113 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

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O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 115 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

11. LIST OF TABLES

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 116 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

12. LIST OF FIGURES

O.W.S.

Code: DSL-1

Page: 117 / 118

Date: Aug-09-2013

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS

O.W.S.

Code: DSL-1

Date: Aug-09-2013

Page: 118 / 118

Revision: 4

BENEFITS AND CHALLENGES OF BIO- AND OXO-DEGRADABLE PLASTICS