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TESTING THE BIODEGRADABILITY AND BIODEGRADATION RATES OF DEGRADABLE/BIODEGRADABLE PLASTICS WITHIN SIMULATED ENVIRONMENT

Summary

The objective of this study was to evaluate and compare the biodegradability and biodegradation rates of 'single-use' plastic bags available on the market and labeled as degradable/biodegradable. The test was carried out under both aerobic and anaerobic conditions. The project length was 20 months. The biodegradation results in the laboratory conditions demonstrate that none of the degradable/biodegradable bags showed visual changes and/or were broken into pieces and none of them experienced any disintegration or degradation. The cellulose filter paper (CFP) completely degraded after 10 days in the aerobic conditions and after 5 month in the anaerobic conditions, implying that the conditions required for biodegradation to occur in a sampling environment were present.

Key words: degradable, biodegradable, plastic bags, aerobic conditions, anaerobic conditions

INTRODUCTION

Plastic bags are a common means of carrying merchandise. In the Czech Republic, retailers, markets, and shops distribute these bags aimed to be used once, predominantly free of charge. After being used to carry goods from retailers to homes, most of the bags are disposed of or stored for reuse; in either case they eventually reach the landfills. Many chain stores in the Czech Republic, Poland and other eastern European countries have introduced degradable/biodegradable plastics and suggested consumers to avoid conventional plastic shopping bags. Plastic bags are appearing on the market with the claim of being "environmentally-friendly", "degradable", or "-bio-," "green-", and sometimes even "compostable", promising solution to littering "simply disappearing". For an average consumer these claims can be quite "misleading". Many doubts have been expressed as to whether these products can provide what they promise.

The objective of this study was to evaluate and compare the biodegradability and biodegradation rates of 'single-use' plastic bags available on the market labelled as degradable/biodegradable. The test was carried out under in both aerobic and anaerobic conditions. This assessment of biodegradable/degradable plastics bags is very important if these materials are going to be biologically treated in the future [Mohee et al., 2008]. The original project length was 12 months, but inevitably ran for 20 month.

LITERATURE REVIEW

In the past, plastic polymeric materials have been designed to resist degradation. However, with mounting environmental and legislative pressure to reduce plastic and packaging wastes, there has been an increased demand for biodegradable polymers that are compatible with the environment [Mohee et al., 2008]. A variety of biodegradable plastics (BDPs) is available today. They may be produced from renewable and fossil resources or the resources may be mixed. In spite of the variety, the content of biodegradable plastics (BDPs) on the market is still low. However, a future increase is expected. In the EU, the consumption of biodegradable plastics (BDPs) was estimated to be 0.02–0.03 Million Mg for 2001. For 2010 an increase up to 1.1 Million Mg is foreseen. A significant limitation for application of biodegradable plastics is the price which is approximately 2–3 times higher compared to conventional plastics at present [Körner et al., 2005]. The current price of such materials is their major drawback for using them in large-scale applications [Di Francoa et al., 2004].

The basic idea behind biodegradable plastics is taken from nature's cycle. Most of the plant material generated on earth is converted back into the original materials – carbon dioxide and water – by microorganisms. This cycle is considered a model for biodegradable plastics, which are produced from agricultural renewable resources [Körner et al., 2005]. However, biodegradable polymers are only beneficial when they can actually biodegrade. Consequently, biodegradable plastics have been the topic of many researches. Detailed review and characteristics of biodegradable materials and their degradation were described in Biodegradable Polymers Review [Vorman et al., 2009].

Several studies have been carried out to evaluate the degradation and biodegradation of degradable/biodegradable plastics. Most of the experiments had positive results. The aerobic biodegradation behaviour of plastics is well studied and many polymer degrading microorganisms have been isolated and identified. The evaluation of the anaerobic breakdown of plastics, however, is still in a developing stage and only few reliable investigations are available [Abou-Zeid et al., 2001].

COMMERCIALLY AVAILABLE PRODEGRADANTS

The use of prodegradants is an old technology that has only been commercially important in recent years. Patents surrounding the science of prodegradant additives and degradable polyolefins have dated as far back as the early 1940s. The following sections cover the popular commercial prodegradant and degradable polymers found in literature to date [Ammala et al., 2011].

A large percentage of commercially available prodegradants have been based on transition metals in the forms carboxylates and dithiodicarbamates for mostly polyolefins. Degradable finished products are widely available through many compounders, who have obtained license rights to use pre-packaged prodegradants from the manufacturers. Some popular prodegradant manufacturers are listed in Table 1 [Ammala et al., 2011].

Table 1. General information on transition metal based prodegradants commercially available

Tradename	Manufacturer	Active components	Loadings (wt%)	Degrading conditions		Polymer types
				No light	Anaerobic	
TDPA	EPI	Metal stearates (Fe, Ce, Co) and citric acid (typically Co)	2.III	Yes	Yes	PP, PE, PS
Renatura	Nor-X Industries	Iron stearate and combination of stabilisers/antioxidants	2	Yes	Possibly no	PP, PE
Reverte	Wells Plastics Limited	Undisclosed photo-inhibiting package, metal ion prodegradant package and biodegradation promotors (micronised cellulose)	1.V	Yes	Possibly no	PP, PE, PS, PET, ABS
AddiFlex	Add-X Biotech	Metal carboxylate (Fe, Mn, Cu, Co, Ni), starch, $CaCO_3$; manganese stearate has been identified for AddiFlex HE [61]	10–20 (AddiFlex A) 3–6 (AddiFlex HE) 1.5–6 (AddiFlex HES)	Yes	Possibly no	PP, PE, PS
d_2W	Symphony Environmental	Metal stearates and stabilisers (typically Mn)	1.III	NA	No	PP, PE
Scott–Gilead technology		Metal dithiodicarbamate		No	No	PP, PE

Source: [Ammala et al., 2011].

EPI is one of the key players in the current prodegradant market. EPI manufactures, distributes, sells and licence out their Total Degradable Plastics Additive (TDPA™) to other plastic fabricators to produce oxo-degradable polyolefinic products. Their prodegradants are used in a vast variety of finished products such as carrier and disposable waste bags, food packaging and food service items, "bubble wraps", personal care products (diapers), and agricultural mulch films [Ammala et al., 2011]. There are many licensees that use TDPA in many polyolefinic products (Symphony Environmental Ltd., EPI Environmental Products, Sancell, Copol International). Some of them are available on the Czech and Polish market.

MATERIAL AND METHODS
EXPERIMENTAL CONDITIONS AND MATERIALS

The biodegradability of plastic materials was determined under aerobic and anaerobic conditions. In both tests the test material was exposed under laboratory conditions. Several kinds of degradable/biodegradable bags (commercially available) were used in this study. One of them was a carrier bag or a "shopper-bag" made of HDPE and mixed with totally degradable plastic additive (TDPA additive). Another was a carrier bag or a "shopper-bag" made of polyethylene (PE) with the addition of pro-oxidant additive (d_2w additive). Further ones were bags labelled as 100% degradable/biodegradable within various periods of time, from three months up to three years. Before the samples were placed into the testing environments, they were photographed. This was to document any change in physical appearance of the samples, along with any rips or physical changes. Cellulose filter paper (CFP) was purchased from stationery and used as a positive control material. The chain stores who have introduced the degradable/biodegradable bags are not named in this article as their identification is irrelevant with regard to the aim this experiment. The emphasis has been put on discovering whether the bags are degradable/biodegradable or not. Nevertheless, the documentation connected to the chain stores is part of the research.

DETERMINATION OF BIODEGRADABILITY
UNDER AEROBIC CONDITIONS

The test started in October 2010 and was designed to investigate the degradation characteristics of degradable/biodegradable bags under aerobic conditions. Bag samples were obtained in chain stores in the Czech Republic and Poland. The elemental composition of each degradable/biodegradable bag was not analyzed. Four bag samples were cut laterally into approximate 70 mm by

70 mm square pieces and into 70 mm by 10 mm stripe pieces and placed into transparent polypropylene boxes covered by compound of soil and compost. The used compost corresponded to a three-month-old mature compost which was provided by a full-scale aerobic composting plant located in Brno - Černovice (Czech Republic). The characteristics of the used compost are shown in Table 2.

Table 2. Characteristics of the compost

Parameters	Value	Unit
Moisture	30 – 65	%
Combustibles	min. 20	%
Total Nitrogen	min. 0.6	% DM
pH	6.0 – 8.5	
Indecomposable ingredients	max. 2.0	%
C:N	max. 30	
Cd	2	$mg.kg^{-1}$
Pb	100	$mg.kg^{-1}$
Hg	1	$mg.kg^{-1}$
As	20	$mg.kg^{-1}$
Cr	100	$mg.kg^{-1}$
Mo	20	$mg.kg^{-1}$
Ni	50	$mg.kg^{-1}$
Cu	150	$mg.kg^{-1}$
Zn	600	$mg.kg^{-1}$

Source: composting plant Brno – Černovice.

The soil was collected from a rural area with a native vegetation of grasses. Boxes were perforated so that the water could flow off, and controlled weekly for aeration and eventual humidity replacement. The samples were exposed to conditions of saturated humidity, the latter being obtained through regular spray application of water. The samples were kept at 23°C±2°C. Cellulose filter paper (CFP) as a positive control was tested under the same conditions as the plastic bags. This additional test was to prove the bioactivity of the soil/compost used in the experiment. At intervals, the exposed samples were analyzed for visual inspection of fragmentation, documented by photography.

DETERMINATION OF BIODEGRADABILITY UNDER ANAEROBIC CONDITIONS

The degradable/biodegradable plastic bags in this research were placed in an anaerobic digester to assess the degradation in an environment without oxygen. The test started in November 2009 and was aimed to simulate and investigate the realistic degradation of degradable/biodegradable bags under anaerobic field conditions, such as landfills. There ware five samples and cellulose filter paper (CFP) as a positive control. Each degradable/biodegradable bag or reference were cut laterally into approximate 70 mm by 70 mm squares and added to transparent 1-liter glass bottle containing water and anaerobic sludge taken from the anaerobic digester from Brno (the municipal sewage treatment plant in the Czech Republic). All bottles were tested under controlled laboratory settings where permanent temperature of 40°C±2°C was maintained.

RESULTS AND DISCUSSIONS VISUAL OBSERVATIONS

The evaluation of visible changes in plastics can be performed in almost all tests. Effects used to describe degradation include roughening of the surface, formation of holes or cracks, de-fragmentation, changes in color, or formation of bio-films in the surface. These changes do not prove the presence of a biodegradation process in terms of metabolism, but the parameters of visual changes can be used as a first indication of any microbial attack. To obtain information about the degradation mechanism, more sophisticated observations can be made using either scanning electron microscopy (SEM) or atomic force microscopy (AFM). After an initial degradation, crystalline spherolites appear on the surface; this can be explained by a preferential degradation of the amorphous polymer fraction, etching the slower-degrading crystalline parts out of the material. In another investigation, Kikkawa et al. [2002] used AFM micrographs of enzymatically degraded PHB films to investigate the mechanism of surface erosion. A number of other techniques can also be used to assess the biodegradability of polymeric material. These include: Fourier transform infrared spectroscopy (FTIR), differential scanning colorimetry (DSC), nuclear magnetic resonance spectroscopy (NMR), X-ray photoelectron spectroscopy (XPS), X-ray Diffraction (XRD), contact angle measurements and water uptake [Ali Shah et al., 2008]. The application of these techniques in general goes beyond the scope of this review.

DEGRADATION OF BIODEGRADABLE BAGS UNDER AEROBIC AND ANAEROBIC CONDITIONS

The biodegrability of several types of plastic bags, namely HDPE mixed with totally degradable plastic additive (TDPA additive) and polyethylene (PE) with the addition of a pro-oxidant additive (d_2w additive), as well as cellulose filter paper (CFP) as a positive control and the others bags which were labelled by chain stores as 100% degradable/biodegradable, was tested under both aerobic and anaerobic environments. With regards to the aerobic environment, parameters (temperature, moisture) were also assessed to ensure that the test samples were under an appropriate environment. The temperature was monitored from day 1 when setting up, to day 122. The average temperature of the samples was 23°C±2°C. At no time during the period did the temperature go beyond 25°C and below 20°C. There was no distinguishable cellulose filter paper left after 10 days of experiment, implying that it was fully biodegraded (100%) and that the conditions required for biodegradation to occur in a sampling environment were present.

Using successively improved test methods, the anaerobic degradation was assessed for plastics being of commercial interest as degradable/biodegradable plastics. The anaerobic degradation was monitored for more than 365 days. The temperature was monitored from day 1 when setting up, to day 365 on a weekly basis. The average temperature of the samples was 40°C±2°C. The degradable/biodegradable bags did not show neither significant biodegradation nor visual changes and were not broken into pieces or easily crumbled when touched. The surface of four samples was smooth and there were no pinhole size observed on the surface of the biodegradable bags after the test. Four samples showed no decrease in color intensity, the sample pigment was still rich. Only one sample showed a visible change in pigmentation. There was distinguishable cellulose filter paper (CFP) left after 3 month, which was used as a control material under simulated landfill conditions, but degraded quite well within 5 months.

The use of plastics, especially polyolefins has increased significantly in recent decades largely due to their low cost, good mechanical properties and light weight. However, this increase in usage has also created disposal problems. Traditional disposal methods include recycling, incineration and burying in landfill. Since polyolefins do not easily degrade in the natural environment, the need for degradable polyolefins has become a major topic of research in order to manage such environmental problems [Ammala et al., 2011].

In most cases, plastic materials should manage a predetermined service life before physical degradation commences. There should be no significant changes in the physical and mechanical properties of the material during its service life. However, after the material has served its primary purpose, rapid biodegradation and disintegration should occur [Jakubowicz, 2003]. The environmental degradability of plastics is a complex process that is influenced by the nature of the plastics and the conditions to which they are exposed [Mohee et al., 2008]. Biodegradable plastics generally decompose into carbon dioxide and water, and consume oxygen under aerobic conditions, while they degrade into methane and carbon dioxide under anaerobic conditions [Choa et al., 2011].

This study was carried out in order to assess the biodegradability of several types of plastics bags under aerobic and anaerobic conditions and to find out whether there were any physical changes when exposed to this environment. The biodegradation results in the laboratory conditions demonstrate that none of the degradable/biodegradable bags showed visual changes and/or were broken into pieces and none of them experienced any disintegration or degradation. However, the cellulose filter paper (CFP) completely degraded after 10 days in the aerobic conditions and after 5 month in the anaerobic conditions, implying that it was fully biodegraded (100%) and that the conditions required for biodegradation to occur in a sampling environment were present.

CONCLUSION

1. To conclude, the outcome of this experiment must be evaluated as negative. The experiment did not bring along the anticipated results despite many studies have confirmed the biodegradability of the plastics [Bonhomme et al., 2003; Chiellini et al., 2003; Chiellini et al., 2006; Ammala et al., 2011]. The fact that the degradable/biodegradable bags have not degraded may be caused by various factors such as (i) inappropriately selected methodics, (ii) improper conditions for biodegradation, (iii) unduly short period for degradation.

2. Considering all the above mentioned facts, it can be concluded that many chain stores in the Czech Republic, Poland and other eastern European countries seem to have introduced standard plastic bags claiming these as degradable/biodegradable plastics or bags labelled incorrectly. The results of research haven't proved the feature of degradability/biodegradability of these bags in any way so far. The research will therefore continue in the following two years.

3. Large quantities of plastics are disposed in the landfills in Czech Republic every year. It can be presumed that the consumption of these bags will increase and the amount of plastic waste will increase proportionally. It is necessary to address this issue furthermore and search for the most suitable manner in which to treat this kind of waste. The same types of bags shall be subject to further testing, namely the process of composting in both laboratory conditions and real conditions.

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