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Research of the biodegradability of degradable/biodegradable plastic material in various types of environments*

Key words: compostable bag, degradable/biodegradable bag, controlled composting environment – laboratory-scale, domestic compost bin, landfill conditions

Introduction

Throughout the world the pollution of natural environment by hazardous substances is one of the most crucial environmental problems (Bespalov et al., 2016; Wang and Yang, 2016). Industrial activity, fermentation chambers, oil spills, harbour, urban stormwater as well as municipal waste landfills may cause pollution of soil (Radziemska and Fronczyk, 2015; Fronczyk et al., 2016), groundwater (Fronczyk and Radziemska, 2016) and air (Rozbicka and Rozbicki, 2014). One of the waste deposited in landfill are plastic bags that involve adverse environmental impacts. Plastic bags are made of non-renewable resources (i.e. petroleum), it takes hundreds of years to degrade, and usually contain substances that pollute the environment (Jakovcevic et al., 2014).

The most consumed synthetic polymer is polyethylene (PE), with a current global production of ca. 140 million tons per year (Sivan, 2011). Plastics production exceeds 180.10^9 kg per year, with a yearly increase in supply and demand. These plastics turn to solid waste after their end of life and will accumulate in the environment. Hence, from an environmentally friendly point of view, the production of biodegradable plastics (BP) is important to reduce the accumu-

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lation of plastic waste in the environment (Iovino et al., 2008). Plastic products are characterized as not easily degradable because of their relatively high stability and hydrophobic characteristics (Iovino et al., 2008). Plastics are inert, durable, hygienic, lightweight, cheap, and malleable. However, the main environmental disadvantage of plastics materials is that they do not readily break down in the environment and therefore can litter the natural environment (Mohee and Unmar, 2007). Plastic waste is recognized as one of the most troublesome categories of waste, and disposal of plastic waste has been blamed for shortening the life of landfill sites (Ishigaki et al., 2004). Consequently, considerable attention has been given to the development of BP materials derived from agricultural resources or, alternatively, to petroleum-based plastics modified with degradable additives. Biodegradable plastics can decompose into carbon dioxide, methane, water, inorganic compounds or biomass via microbial activities within the natural environment (Cho et al., 2010). Moreover BP are designed to degrade under environmental conditions or in municipal and industrial biological waste treatment facilities.

Many plastics that are labeled as "degradable" do not decompose very readily, and it is not clear that litter will be diminished to any great degree through their use. In addition, because not all plastics are or will be degradable, user confusion is and will be common. Multiple formulations mean not all degradable plastics address compost contamination, and most degradable plastics do not address other problems associated with plastics waste management (WM). Therefore it is not clear that degradable plastics constitute a major technological advance; in fact, overall they may be more harmful than helpful (Tonjes and Greene, 2013). Just because something has the prefix bio- (e.g. biodiesel, bioplastics and more) does not mean it is more environmentally friendly (Harding et al., 2016).

Single-use plastic bags

Plastic bags are a common means of carrying merchandise. In the European countries retailers, markets, and shops distribute these bags – intended to be used once. Many chain stores have introduced BP plastics and have suggested that consumers avoid conventional plastic shopping bags. An increasing number of products labeled with the terms environmentally friendly, degradable, bio-, green-, bio-based, and biodegradable are being developed as promising solutions to litter "simply disappearing" (Vaverková et al., 2012).

Compostable plastics

"Single-use" bags certified as compostable have been appearing on the market in recent years. Compostable polymers are being promoted as environmentally beneficial, especially if they can be derived from renewable resources and recovered through organic recycling (Vaverková et al., 2012). Biodegradable plastics, which have been designed to be easily degraded by microorganisms and to be absorbed by the natural environment or by waste landfills, are gaining public endorsement as a possible alternative to petroleum-derived plastic (Ishigaki et al., 2004). It is important to note that all compostable plastics are biodegradable, but not all BP are compostable (Balaguer et al., 2015).

Aims and objectives

Studies of the degradation of BP in various types of environments have been carried out (Ishigaki et al., 2004; Kale et al., 2006; Mohee and Unmar, 2007; Adamcová at al., 2013; Vaverková et al., 2014; Harding et al., 2016) and international standards for the compostable polymers have been developed by the American Society for Testing and Materials (ASTM), the International Standards Organization (ISO) and the European Committee for Standardization (ECN) for evaluation of the compostability of BP materials. ASTM standards, ISO standards and ECN standards allow evaluation of materials under laboratory conditions. As such and until now, no standard has focused on the degradability of degradable/biodegradable materials under real conditions (Kale et al., 2006).

The relatively high number of reports describing the biodegradability of a wide range of BP may lead to the inaccurate conclusion that most plastic polymers can be readily biodegraded. In fact, in terms of amounts, the production of the PE and polystyrene (PS) is, by far, greater than that of the rest of the other plastic compounds that are considered biodegradable. Furthermore, not all types of BP plastics are destroyed completely in natural environments, raising the question of the definition of biodegradable (Sivan, 2011). It is clearly important to study the impact of these materials on WM so to realize the truth benefit and the need to establish adequate waste management system (WMS) and legislation. In the present study, biodegradability of commercial degradable/biodegradable materials made of HDPE and mixed with totally degradable plastic additive (TDPA additive) or made of PE with the addition of pro-oxidant additive (d_2w additive), advertised as 100% degradable or certified as compostable within various types of environments were investigated.

Material and methods

Samples

The investigated materials in all the experiments: (i) controlled composting environment – laboratory-scale, (ii) real composting conditions – domestic compost bin, (iii) real composting conditions – industrial composting plant and (iv) landfill conditions, were obtained from chain stores in Europe. Commercially available bags were used in all studies and cellulose filter paper – CFP) (with dimensions 0.3 mm thickness) as a positive control (reference). One of them was a carrier bag or a "shopper-bag" made of HDPE and mixed with TDPA additive. Another was a carrier bag or a "shopper--bag" made of PE with the addition of d_2w additive. One was labeled as 100% degradable within various periods of time, from three months up to three years, and bags certified as compostable. The investigated materials are listed in Table 1.

TABLE 1. Degradable/biodegradable materials used in experiments

Sample	Type	Description
1	N/A	BIO-D Plast
2	HDPE+TDPA	100% degradable
3	N/A	100% degradable
4	starch	Compostable 7P0147
5	starch and polycaprolactone	OK Compost AIB VINCOTTE
6	N/A	Compostable 7P0202
7	natural material	Compostable 7P0073
8	cellulose (blank)	–

Controlled composting environment – laboratory-scale

The first test was carried out in a controlled composting environment. The biodegradation degree of the samples was evaluated following a modified version of Czech National Standard ČSN EN 14806 "Packaging – Preliminary evaluation of the disintegration of the packaging materials under simulated composting conditions in a laboratory scale test" and a modified version of Czech National Standard ČSN EN ISO 20200 "Plastics – Determination of the degree of disintegration of plastic materials under simulated composting conditions in laboratory-scale test" (ISO 20200:2004). The emphasis was put on discovering whether the bags are degradable/biodegradable or not (Vaverková et al., 2012).

Real composting conditions – domestic compost bin

The second test was carried out in composting conditions – domestic compost bin. This study was carried out in order to assess biodegradability of the samples under real conditions of home composting, and to find out whether there were any physical changes when exposed to natural composting environment (Fig. 1). The experimental samples were

FIGURE 1. Initiation of the experiment in compost bin

placed in home compost bins and were checked and visually assessed during the experiment (Vaverková et al., 2014a).

Real composting conditions – industrial composting plant

The third test was carried out in real composting conditions in 2011 and 2012. In both cases samples were placed into frames and inserted into one clamp within the compost pile to investigate biodegradation (Fig. 2). Frames have been designed and manufactured in 2011. The research of biodegradability was carried out in real conditions in the Central Composting Plant in Brno (Adamcová et al. 2013; Vaverková et al. 2014b).

FIGURE 2. Placement of samples in the compost pile

Landfill conditions

The fourth test was carried out in municipal solid waste (MSW) landfill conditions. Samples were placed into frames. All samples were buried into landfill (Fig. 3). Samples were laid over the surface of the landfill and then buried MW to a final depth of 1 m (Adamcová and Vaverková, 2014).

Results

Controlled composting environment – laboratory-scale

The CFP completely degraded after 10 days, implying that it was fully biodegraded and that the conditions required for biodegradation to occur in sampling environment were present. No breakthrough in disintegration was observed for samples made of HDPE with TDPA additive or made of PE with d_2w additive or sample labeled as 100% degradable (Samples 1–3). After composting period

FIGURE 3. Placement of the samples buried into landfill

in the laboratory-scale test, the test material still remained completely intact. The samples did not show any significant biodegradation or visual changes and were not broken into smaller pieces or easily crumbled when touched. The surface was smooth, and there were no pinholes observed on the surface after the test. The biodegradation of the certified compostable plastic bags proceeded very well (Samples 4–7). After composting period the different test materials seemed to completely disappear. This was confirmed at the end of the test. Table 2 presents the amounts of plastic pieces before (M_i) and after composting (M_r) from each reactor, as well as their corresponding disintegration degree (D).

Real composting conditions – domestic compost bin

From the results obtained during the biodegradation test in domestic compost bin, it can be concluded that the samples made of HDPE with TDPA additive and made of PE with d_2w additive or sample labeled as 100% degradable (Samples 1–3) have not decomposed, their color has not changed and that no degradation neither physical changes have occurred. Thus, the samples cannot be claimed to be biodegradable. Samples certified as compostable have not decomposed (Samples 4–7).

In contrast to the laboratory conditions, the real conditions (including home composting) are affected by a number of factors that cannot be influenced such as air temperature, pH of the environment, water content of the compost pile, precipitation etc. It is necessary to emphasize that the nature of the compost raw material plays an important role in polymers' degradation since different compost systems (i.e. manure, yard, and food waste) produce different microbiological activity. All these factors can significantly affect the rate and degree of degradation.

TABLE 2. Amounts of samples before (M_i) and after composting (M_r) and disintegration degree (D)

Sample	M_i [g]	M_r [g] *	D [%]*
1	3.45	3.7	0
2	7.02	7.5	0
3	7.03	7.2	0
4	7.01	0.03	99.6
5	7.02	0	100
6	7.01	0.009	99.9
7	7.02	0	100
8	7.02	0	100

*Mean value.

Based on this test it can be concluded that certified compostable plastic bags (Samples 4–7) showed complete level of biodegradation during the composting test. In contrast, test material made of HDPE with TDPA additive and the bag made of PE with d_2w additive or sample labeled as 100% degradable (Samples 1–3) remained completely intact at the end of the test.

Real composting conditions – industrial composting plant

The experimental samples were placed in the compost pile and were checked and visually assessed. Research in real conditions is not supported by norms, neither exist methodologies describing procedures for the research of decomposition of these materials in real conditions. Up to now, no laboratory tests were capable of copying the conditions of industrial composting plants. After the expiration of the experimental period it was found out that the samples made of HDPE with TDPA additive and made of PE with d_2w additive or sample labeled as 100% degradable have not decomposed, their color has not changed and that no degradation neither physical changes have occurred (Fig. 4). Samples certified as compostable were decomposed. Control reference sample confirmed that the conditions of decomposition were suitable during the experiment (Fig. 5).

The experiment was carried out in real conditions for the first time in 2011 and its repetition was carried out again in 2012 in order to verify the achieved experimental results. The results confirmed the findings from 2011.

Landfill conditions

In the research conducted in 2012––2014 (still ongoing) experimental samples were placed in the MSW landfill and

FIGURE 4. Not decomposed samples (Samples 1–3)

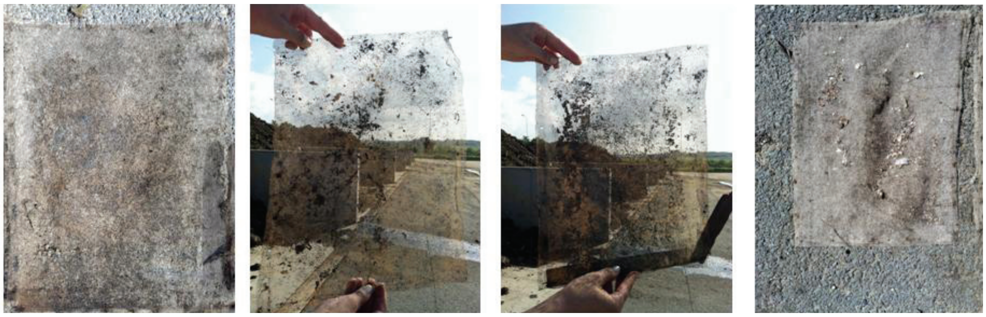

FIGURE 5. Decomposed samples (Samples 4–7)

were checked and visually assessed during the experiment. After the expiration of the experimental period (48 months) it was found out that the samples made of HDPE with TDPA additive and made of PE with d_2w additive or sample labeled as 100% degradable (Samples 1–3) have not decomposed and no degradation neither physical change have occurred (Fig. 6); however, their color has slightly changed. For the time being, the research has confirmed that the samples do not biodegrade or disintegrate in landfill. Samples certified as compostable have decomposed (Samples 4–7). Control reference sample confirmed that the conditions of decomposition were suitable during the experiment. The CFP biodegrade after 8 months, implying that it was fully biodegrade and that the conditions required for biodegradation to occur in a sampling environment were present.

mental conditions, and the acceleration of the degradation processes by various manipulations in order for experiments to be concluded timely means that determinations of when reactions occur under ambient conditions are often inexactly estimated (Tonjes and Greene, 2013). The closer experiments mimic environmental conditions, the better the likelihood of timing degradation effects correctly (Searle, 2003), although careful bench-scale tests in at least one instance considerably over predicted degradation measured in field experiments (Tonjes and Greene, 2013). Laboratory results can lead to incorrect descriptions of degradation potentials, or widely varying estimates of environmental persistence. Thus, many manufacturers claim their products undergo reactions faster or more completely than they actually do. Contrarily, those observing products

FIGURE 6. An example of not decomposed samples in landfill conditions

Discussion

Degradation potential of polymers is usually tested in laboratory experiments that simulate long exposure times. These processes do not exactly match environ-

which remain more intact in the environment than product specifications outline then fear that these products will remain undegraded for thousands of years or more (Tonjes and Greene, 2013).

Biodegradable plastics are primarily intended to address composting contamination (and litter issues). Compostable plastics require specific levels of moisture and oxygen for initial reactions to occur to make the polymers consumable by bacteria (Song et al., 2009). These conditions are usually only found in larger, industrial-commercial facilities, where materials are regularly turned, and usually have been pre-processed often shredded (Kale et al., 2007a).

Compostable plastics under standard, large-scale composting practices have been found to degrade well, with different kinds of substrates, such as yard waste, manure, and food waste (Kale et al., 2007b), or using different technologies, such as turned windrow or in-vessel (CSU Chico, 2007). These results have led to endorsement of their wider use (Tonjes and Greene, 2013).

However, reports of failure to perform by compostable-labeled plastics in at-home composting environments are common. Inadequate temperatures in these smaller piles, so that the key reaction for degradable/biodegradable plastics is not initiated, are thought to be the reason for much of the poor results (Farrington et al., 2005; Song et al., 2009). This has reignited controversies associated with earlier BP products, due to the mismatch between producer claims and consumer experiences (Tonjes and Greene, 2013).

Replacing recalcitrant plastics with plastics that have greater potential to degrade may result in greater degradation of the plastics themselves – if the degradable plastics encounter conditions that result in depolymerization. Burial of UV-sensitive plastics is not likely to result in any early plastics decay. Plastics where degradation is initiated by higher temperatures are more likely to start decomposing in most landfills. For instance, landfill cover film made of PE and TDPA lost integrity in one three month trial, and average molecular weight was reduced to less than 5 kDa after 14 months at another site (Swift and Wiles, 2004). Most BP that are "compostable" generally require moisture and oxygen for the process to proceed very far, however. Moisture may or may not be available in particular landfills or areas in landfills, but landfills generally are known to be lacking in oxygen. No studies of compostable plastics in landfill environments were located (ExcelPlas Australia, 2004), although some starch-based plastics have degraded in simulated anaerobic digesters (CSU Chico, 2007).

Although all polymers will degrade under certain conditions, plastics that are specifically designated as degradable/biodegradable have been manufactured to do so in an enhanced way. Biodegradable plastics are supposed to lose important materials properties within days-weeks--months after intended usage has been completed (Tonjes and Greene, 2013).

Conclusion

This long-term research was carried out in order to assess biodegradability of commercial materials made of HDPE and mixed with TDPA additive or made of PE with the addition of d_2w additive, advertised as 100% degradable or certified as compostable within various types of environments. They were investigated under different conditions in order to find out whether there were any physical changes when exposed to different environment. The results demonstrate that the materials made of HDPE and mixed with TDPA additive or made of PE with the addition of d_2w additive or advertised as 100% degradable did not biodegrade in any of the above-described conditions and remained completely intact at the end of the tests. The experiments did not yield the anticipated results. Biodegradation of the certified compostable plastic bags proceeded very well in laboratory-scale conditions and in real composting conditions – industrial composting plant, however, these materials did not biodegrade in real composting conditions – domestic compost bin and landfill conditions. The main conclusion that can be derived from these studies is that degradable/biodegradable plastics or plastics certified as compostable are not always suitable for home composting and that in home compost bin they do not degrade. As far as landfill environment is concerned, the research is still in progress. All test specimens will be analyzed after the end of the experiments using conventional Scanning Electron Microscope (SEM) techniques.

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Summary

Research of the biodegradability of degradable/biodegradable plastic material in various types of environments. Research was carried out in order to assess biodegradability of degradable/biodegradable materials made of HDPE and mixed with totally degradable plastic additive (TDPA additive) or made of polyethylene (PE) with the addition of pro-oxidant additive (d_2w additive), advertised as 100% degradable or certified as compostable within various types of environments. Research conditions were: (i) controlled composting environment – laboratory-scale, (ii) real composting conditions – domestic compost bin, (iii) real composting conditions – industrial composting plant and (iv) landfill conditions. The results demonstrate that the materials made of HDPE and mixed with totally degradable plastic additive (TDPA additive) or made of polyethylene (PE) with the addition of pro-oxidant additive (d_2w additive) or advertised as 100% degradable did not biodegrade in any of the above-described conditions and remained completely intact at the end of the tests. Biodegradation of the certified compostable plastic bags proceeded very well in laboratory-scale conditions and in real composting conditions – industrial composting plant, however, these materials did not biodegrade in real composting conditions – domestic compost bin and landfill conditions.

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