Original Research

Degradation of Biodegradable/Degradable Plastics in Municipal Solid-Waste Landfill

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Abstract

This paper provides information about biodegradability of biodegradable/degradable materials advertised as 100%-degradable or certified as compostable in a municipal solid-waste landfill. It describes a 12-month experiment with five samples that took place in a landfill. The results, demonstrated that the polyethylene sample with the additive (samples 1) and sample labelled as 100% degradable (sample 2) had not decomposed, and no physical changes had occurred; however, their color had changed slightly. Samples 2 and 3 (certified as compostable) did not decompose. However, sample 3 exhibited minor disruptions. Sample 5 was a control reference sample to confirm that the conditions of decomposition were suitable during the experiment, which was the case. The cellulose filter paper (sample 5) degraded after 8 months, implying that it was fully biodegraded (100%) and that the conditions required for biodegradation to occur in a sampling environment were present.

Keywords: biodegradation, degradability, degradable/biodegradable plastics, waste, municipal solid waste landfill

Introduction

Tremendous amounts of generated waste have made acquiring sufficient areas for landfill sites more difficult. Lack of capacity of waste landfills has become a serious problem, especially in urban areas, and effective reduction or decomposition of waste is now necessary to secure adequate landfill capacity. Among several compounds in waste landfill sites, plastics are estimated to make up approximately 20-30% of the volume of municipal solid waste (MSW) landfill sites [1].

Because plastics are recalcitrant to microbial degradation, they would remain in landfill sites semi-permanently. 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 [1]. Thus, biodegradable polymers – as a potential partial solution of these problems – were developed during the last decade [2]. Biodegradable polymers can decompose into carbon dioxide, methane, water, inorganic compounds, or biomass via microbial activities within the natural environment [3]. Moreover, biodegradable plastics are designed to degrade under environmental conditions or in municipal and industrial biological waste treatment facilities. Aerobic composting as well as anaerobic biogasification of waste are currently in use, the latter process becoming more and more established because of the added benefit of energy conservation due to biomass recovery [2].

In response to this problem of plastic waste in landfills, 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. Commercialized biodegradable plastics include the following three groups, depending on their main

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components: bacterial polymers, plant-derived polymers, and chemically synthesized polymers [1].

Biodegradation of plastics depends on both the environment in which they are placed and the chemical nature of the polymer. Biodegradation is an enzymatic reaction; hence it is very specific to the chemical structures and bonds of the polymer. There are different mechanisms of polymer biodegradation. The process of polymer biodegradation is affected by many factors that can be roughly grouped as follows and can vary widely.

1) The properties of polymer: chemical composition, type of functional groups, molecular mass, internal structure, content of additives, contaminants, etc.
2) The environment in which the process takes place: temperature, illumination, humidity, mechanical and chemical impact (pH, available oxygen, the presence of other compounds, etc.) [4].

In real conditions the process of biodegradation takes place in the environment where a large variety of microorganisms is present. As far as municipal waste is concerned, the raw waste composition and the set of microorganisms present in each batch is unique, impossible to reproduce in laboratory. Because of this specificity, the terms 'biodegradation' and 'biodegradability' are not absolute attributes of a given material but are closely related to the specific conditions in the environment in which the process is to occur. Thus, in addition to studies using standardized laboratory methods, it is necessary to conduct studies of the biodegradation process in real conditions [4].

Studies of the degradation of biodegradable plastics in the composting process have been carried out [4-12] 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 biobased polymeric 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 compostability of biodegradable materials under real conditions [8].

Less attention has been paid, however, to biodegradable plastics degradation in waste landfills. Ishigaki [1] reported that certain degradation potentials for various types of biodegradable plastics existed in waste landfill sites, and suggested that the appropriate management of waste landfills might lead to the stimulation of polymer-degrading microorganisms. But investigations regarding biodegradable plastics degradation in waste landfill sites have not been executed, and the benefit of biodegradable plastics for saving the capacity of the landfill sites has remained unclear [1].

It is clearly important to study the impact of these biodegradable/degradable materials on waste management, thereby realizing the truth benefit and the need to establish adequate waste management systems and legislation. In the present study, degradability of commercial biodegradable/degradable materials advertised as 100%-degradable or certified as compostable within the MSW landfill Štěpánovice were investigated.

Aims and Objectives

The overall rate of degradation of organic materials occurring in MSW landfills is slow. Studies have shown that food waste and newspapers excavated from landfills are readily identifiable following 20 years of burial. Although the overall rate of degradation of organic materials in landfills may exist if favorable environment conditions are present. The rate at which degradation proceeds is a function of many environmental variables, including moisture content, pH, temperature, number and types of organisms present, and solid waste composition [13].

The degradation process for commercial biodegradable/degradable materials following burial in modern sanitary landfills is not well known. To realize the recovery and recycling via landfilling, it must be clarified whether and how these different types of biodegradable/degradable materials or certified as compostable degrade under the special conditions of a modern sanitary landfill. Actually, investigations exist almost exclusively concerning the aerobic degradation of biodegradable plastics under laboratory conditions or in technical composting facilities as the standards are fixed for that way of recycling.

The objective of this study was to measure the rate and extent of deterioration of biodegradable/degradable materials advertised as 100%-degradable or certified as compostable in an MSW landfill. The composition of the solid waste buried at the landfill was determined and environmental conditions at the burial site were measured to determine if favourable conditions existed for degradation to proceed. Tests were conducted to measure the extent of degradation of biodegradable/degradable plastic materials during 12 months of burial in an MSW landfill.

Materials and Methods

Landfill Site Description

The study site is located in the Czech Republic, Pilsen Region, 1 km north of Štěpánovice commune and 1 km south of Dehtín commune. GPS coordinates of the test point are 49º26'15.934"N, 13º16'55.352"E . In this area, the mean annual precipitation is 582 mm and the mean annual temperature is 8.0ºC. The landfill has been operating since summer 1996. It is situated in the northern part of a widely opened valley directed toward W-E. The bottom part of this area is restricted with a nameless stream being the right tributary of the Úhlava River. The upper part of the area is covered with woodland vegetation predominated by *Pinus sylvestris*. The southern slope is used for agriculture. The landfill is located on the northern slope from the valley axis. In the past, the landfill area was used as the meadow [14, 15].

In terms of maintenance, the landfill is classified in the S-category – other waste, sub-category S-OO3. The landfill has a total authorized volume of about 569,000 m^3; at the moment, it is being used to dispose of mixed municipal waste. The landfill (Fig. 1) is formed by three sub-landfills: A, B, C.

Table 1. Material composition of samples.

Sample	Type	Description
1	HDPE+TDPA	100% degradable
2	N/A	100% degradable
3	Natural material	Compostable 7P0073
4	Starch and Polycaprolactone	OK Compost AIB VINCOTTE
5	Cellulose (blank)	Filtering paper

The total amount [Mg] of each type of waste placed in the landfill MSW Štěpánovice in 2004-11 is shown in Fig. 2. The greatest amount of waste placed in the landfill in the reporting period represents mixed municipal waste (55.780 Mg), followed by bulky waste (32.831Mg), and bricks (30.014 Mg) [16].

Samples

As research samples we selected carrier bags that are commonly available in retail chains in Europe. They state that they are biodegradable, degradable, or other designation. In addition, 2 bags certified for composting were chosen. In total, the experiment contained 5 samples: 4 carrier bags and cellulose filtering paper as a control sample.

Description of the Samples

Types and descriptions of the samples are listed in Table 1. For easier orientation the examined samples were labelled with numbers 1-5. At the beginning of the experiment, all 5 samples were photographically documented. Pictures are shown in Fig. 3.

The experimental subjects were made of HDPE with the TDPA additive (sample 1) and polyethylene labelled as 100%-degradable (sample 2). The control sample (cellulose

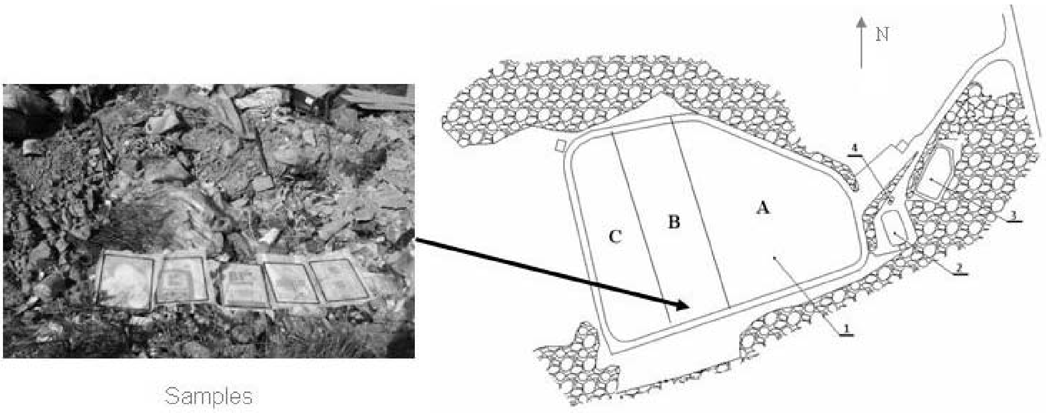


Fig. 1. Map of Štěpánovice MSW landfill.
1 – landfill, 2 – drained water tank, 3 – rainwater reservoir, 4 – entrance gate

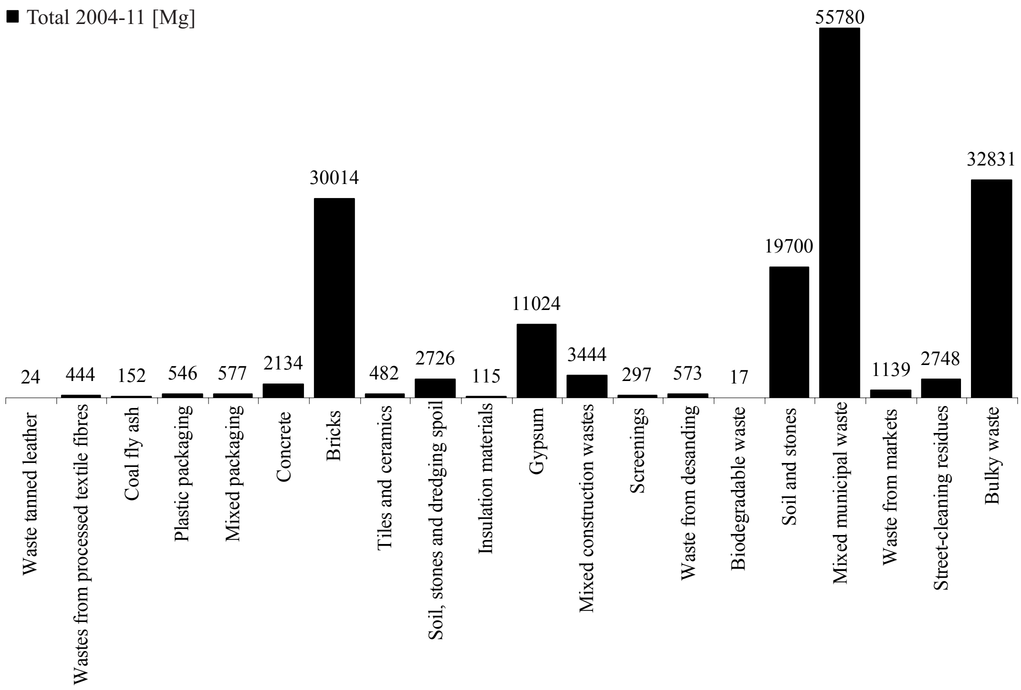


Fig. 2. Total quantity of individual waste in 2004-11 [16].

Fig. 3. Samples labelled with numbers 1-5.

filtering paper) was to check the potential of biological decomposition in the tested environment (sample 5).

Biodegradation Test: Procedure

Samples (1, 2, 3, 4, 5) were placed into frames that were designed and manufactured in 2012. Used frames are the authors' utility models registered in Czech Industrial Property Office. A 3D image of the metal frame is presented in Fig. 4.

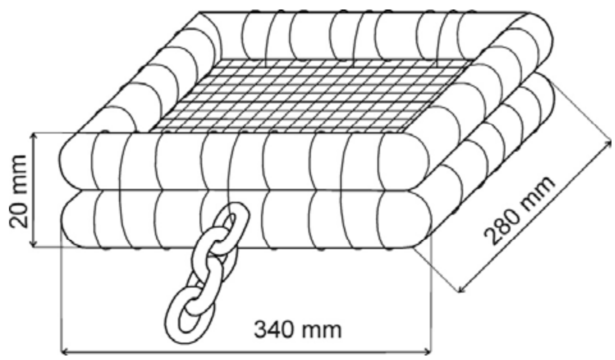


Fig. 4. Image of the metal frame.

The frames were made of metal rods as follows: width 280 mm, length 340 mm, and height 20 mm. A 1×1 mm polyethylene mesh was fixed onto the frames. The frames were designed so that they would facilitate the placement and identification of the samples in the landfill and at the same time the removal of the samples from the given environment. The experimental samples were inserted into the produced frames; in four cases, the experimental samples were complete plastic bags (samples 1, 2, 3, 4) and the fifth (control) sample was represented by cellulose filtering paper. The frames with the samples were properly marked and photographed to document future visual comparison.

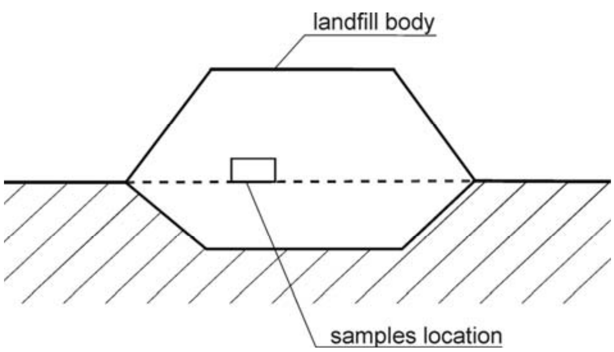


Fig. 5. Placement of samples in the in the landfill.

The experiment started in September 2012. The samples were brought to MSW landfill in Štěpánovice. All five samples were buried in one of the three sub-landfills (sub-landfill B) (Fig. 1) Samples were laid over the surface of the landfill then buried in normal municipal waste to a final depth of 1 m. Placement of samples in the landfill is presented in Fig. 5. In these conditions, the experimental period was estimated to be 12 months. Samples were inspected twice (May 2013, September 2013) when they were removed from the landfill body. A digital camera was used to take pictures. The materials were inspected for colour, texture, shape, and changes in dimensions.

In order to be able to analyze possible effects of weather on the landfill, biodegradation and decomposition of disposable plastic bags, we asked the landfill operator for the records of landfill temperature, average daily temperatures, and precipitation amounts in the concerned area for the monitored period (2012-13). The data on monthly precipitation amounts in millimeters and average monthly air temperatures in degrees Celsius (T) in the monitored experimental period are presented in the diagrams (Figs. 6 and 7).

The average monthly air temperatures in the vicinity of the landfill ranged during the reference period (September 2012-September 2013) from -1.85ºC (the lowest mean value was achieved in February 2013) to 13.86ºC (the highest mean value was achieved in July 2013). Average monthly temperature at the landfill body, measured at a depth of 1m, ranged in the monitored period from 3.4ºC (February 2013) to 20.58ºC (August 2013). The average monthly temperatures at landfill body measured at a depth of 1 m copy the course of monthly average temperatures in the surroundings of the landfill plus 5.33ºC higher values on average.

The average monthly precipitation amounts in the reference period ranged from 0.89 mm to 5.13 mm. The highest average monthly precipitation amounts were measured in October 2012 (3.7 mm) and in June 2013 (5.13 mm). The lowest average monthly precipitation amounts were measured in January 2013 at 0.89 mm.

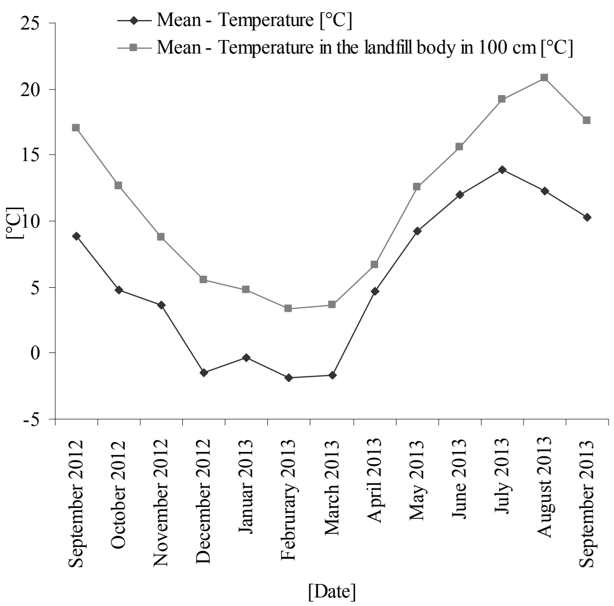


Fig. 6. Landfill temperature and average daily temperatures for the given period (2012-13).

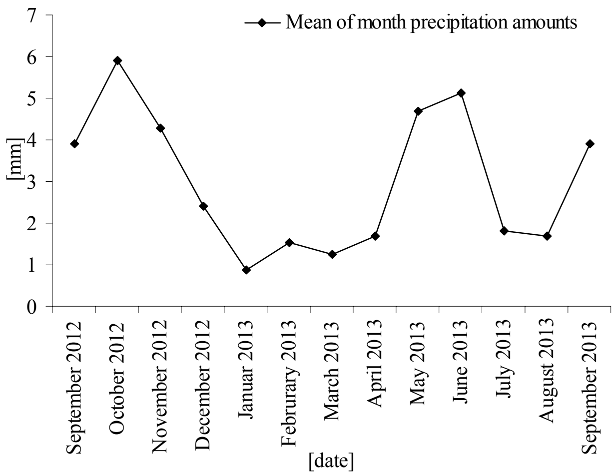


Fig. 7. Mean of monthly precipitation amounts (2012-13).

Results

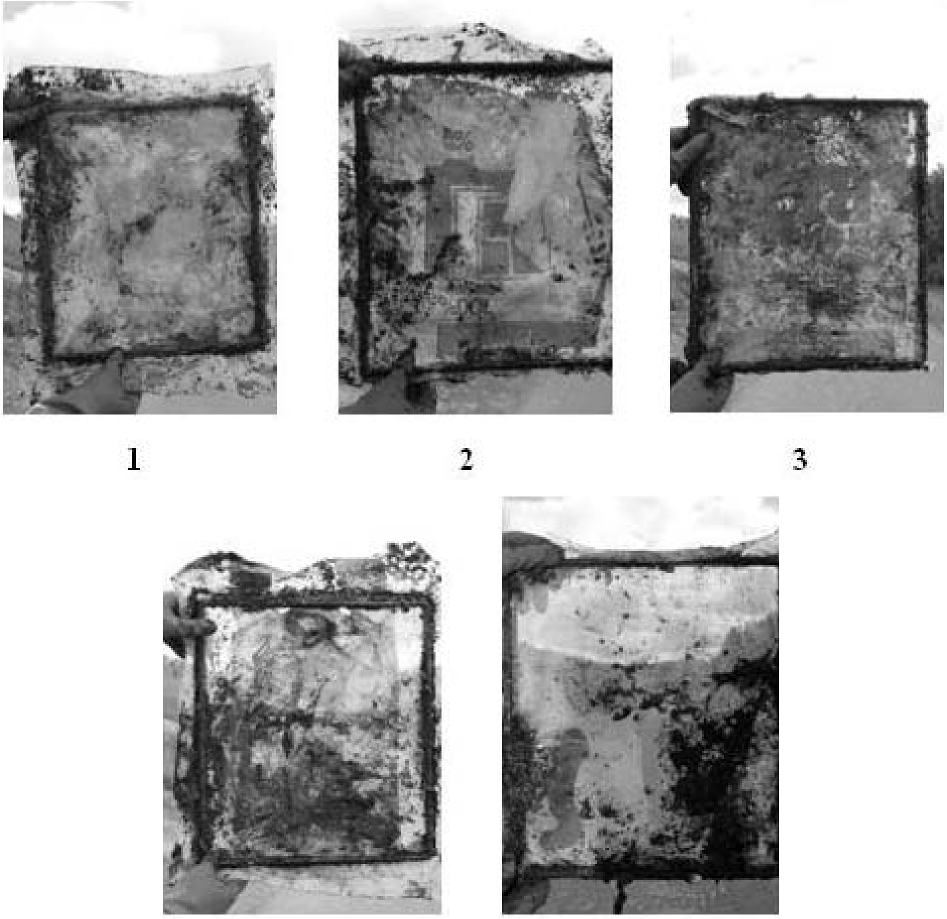
Eight months after the initiation of the experiment the first control of the examined samples stored at the landfill body took place. In May 2013 all five samples were removed from the landfill body using heavy equipment and workers from the landfill. After collection of the samples visual inspection and photo-documentation of the samples were carried out (Fig. 8).

Sample 1 (HDPE+TDPA) showed no modification of structure, no degree of disintegration or decay, nor any change of color (of the printing). Similar results were observed with regard to sample 2 (N/A). Samples 3 (natural material) and 4 (starch and polycaprolactone) did not experience any structural or color changes. Sample 5 (cellulose-blank) had 95% disintegrated, which confirmed that in the place where the samples were stored there were suitable conditions for degradation.

After visual inspection was conducted, all 5 samples were placed into the same marked place in the landfill body using heavy equipment, and kept therein until further inspection scheduled for September 2013.

The second visual inspection of the samples was carried out in September 2013, i.e. 4 months after the first inspection and 12 months after the initiation of the experiment. All five samples were removed from the landfill body using heavy equipment and workers. After their removal the samples were washed for more precise determination of the degree of decomposition, followed by visual inspection and photo-documentation. Photographs of the samples taken during the second inspection are shown in Fig. 9.

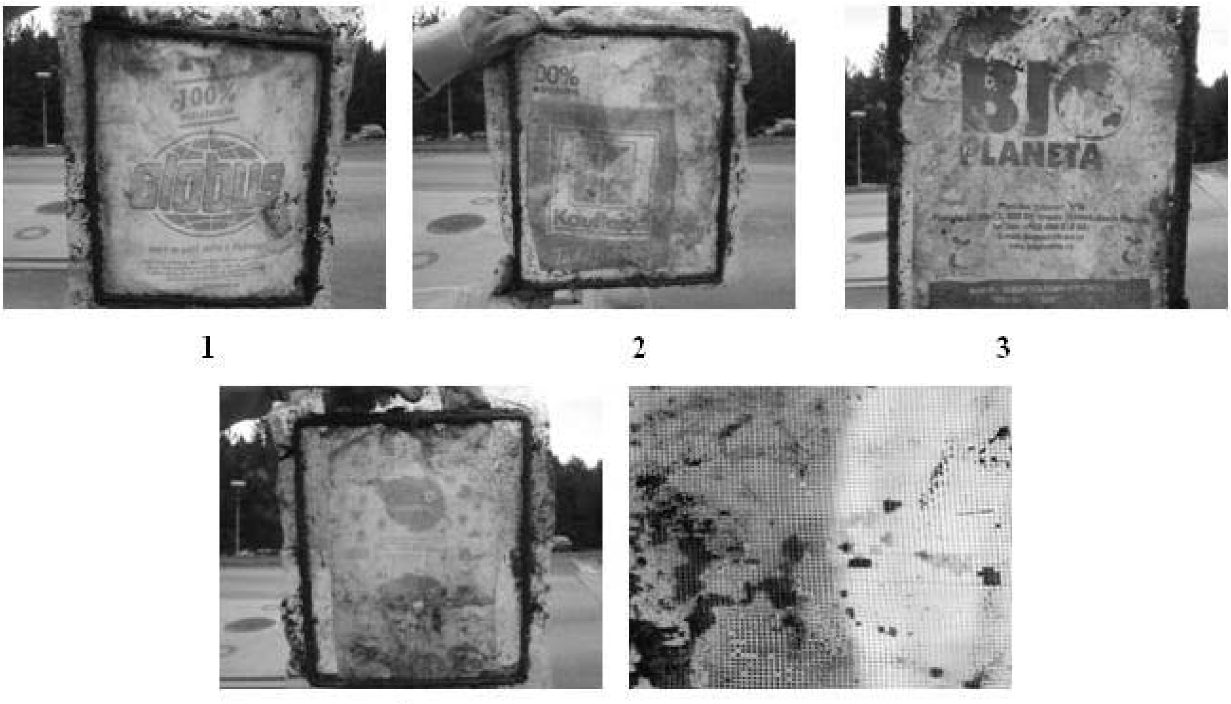
Sample 1 (HDPE+TDPA) showed no modification of structure and no degree of degradation and disintegration: however, the color of the printing changed. Similar results were observed for sample 2 (N/A) – no change of structure, no signs of decay or decomposition, yet the color of printing had changed. Sample 3 (natural material) experienced certain changes of structure – degradation (slight disruption of integrity). Sample 4 (starch and polycaprolactone) showed no signs of degradation and there were no color alterations. Sample 5 (cellulose-blank) exhibited a high degree of decomposition (99%), thus sample 5 is displayed



1 2 3

4 5

Fig. 8. First visual inspection of the samples – May 2013.



1 2 3

4 Detail of Sample No. 5

Fig. 9. Second inspection of the samples – September 2013.

in detail (Fig. 9). The results obtained by sample 5 confirm the presence of suitable conditions for decomposition at the landfill body.

Following visual inspection and photo-documentation, all 5 samples were placed into the same marked place at the landfill body using heavy equipment again, and kept therein until further inspection in May 2014.

Discussion

Although biodegradable plastics are gaining public attention as a prospective alternative to petroleum-derived plastic, the degradability of biodegradable plastics in waste landfills remains unclear [1]. However, Billingham [18] states that several studies of the behavior of the polyethylene sample with additive TDPA film in landfill have been reported [19]. All show clear evidence that microbially-generated heat in a landfill site (30-55ºC) is enough to lead to oxidative embrittlement of polyethylene containing prodegradant, within a few months.

A large-scale trial has been conducted at a commercial landfill site near Birmingham, UK, over the period January 2001-March 2002. Samples of LDPE film with and without prodegradant were laid over the surface of the landfill then buried in normal municipal waste to a final depth of 3 m. During the test period, the weather was generally poor and the temperature recorded by thermocouple probes close to the test films rarely exceeded 30ºC. The maximum temperature recorded was 38ºC in November 2001, due to the combination of the heat from summer sunshine and the microbial activity it promotes. Samples of test films were recovered from the landfill after 10 months of burial. Results clearly confirm the earlier conclusions that polyolefins can be made sufficiently unstable with appropriate additives that they degrade to embrittlement in an acceptable time, even in a cold landfill [18].

In our research the temperatures at the landfill body (at a depth of 1 m) were significantly lower (max 22ºC, 5-7 August 2013) than the temperatures described in the above experiments, yet the conditions suitable for biodegradation were present, which was confirmed by the control sample (Sample 5 – filter paper). At the end of the experiment the samples showed negligible changes (color, structure).

Conclusion

The corroboration of decomposition in laboratory conditions according to current standards answers only the question whether plastic material is biologically degradable. However, it does not answer the crucial question whether the materials are successfully degradable in the environment of a landfill. The goal of the experiment was to verify and test methodology of decomposition of the above-described samples in real conditions of MSW landfill. In the research conducted in 2012-13, experimental samples were placed in the MSW Štěpánovice landfill and were checked and visually assessed.

Research in real conditions is not supported by norms nor exist methodologies describing procedures for the research of the decomposition of these materials in real conditions [11].

After the expiration of the experimental period (12 months) it was found that the polyethylene sample with the additive (samples 1) and sample labelled as 100% degradable (sample 2) had not decomposed and no degradation nor physical changes had occurred; however, their color had slightly changed. For the time being, research has confirmed that the samples made of polyethylene with additives do not degrade or disintegrate in a landfill. Samples 2 and 3 (certified as compostable) have not decomposed. However, sample 3 exhibited minor disruptions. The cellulose filter paper (sample 5) degraded after 8 months, implying that it was fully biodegraded (100%) and that the conditions required for biodegradation to occur in a sampling environment were present. Termination of the experiment is scheduled for September 2015.

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