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Biodegradation of PLA and PLA miscanthus fibre composites

Literature review by Jan E.G. van Dam - Senior Scientist at Wageningen University & Research

Poly lactic acid (PLA) thermoplastic packaging materials are biocompatible in contrast to synthetic polymers such as PE, PP and PS, that do not biodegrade when disposed in the environment or in landfill conditions. However, complete disappearance of PLA in the environment may take several years[1]. The PLA is known to be biodegradable under specific composting conditions[2]. The anaerobic decomposition of PLA is known to be temperature dependent and therefore advised to be performed in controlled industrial composting[3]. Controlled composting in soil at ca 60°C (above glass transition temperature Tg of PLA) is influenced by the PLA crystallinity, pH and UV light[4]. Amorphous PLA is better accessible to microbial enzymes[5].

With addition of lignocellulosic fibres the mechanical properties of PLA result in higher stiffness and crystallization of the PLA polymer. The effects on biodegradation process is accelerating with higher fibre contents, as the fibres are affected themselves[6]. The biodegradation rates of lignocellulose filled PLA were found to be higher than that of pure PLA. A lag phase of ca 15 days is observed and the degree of degradability is ca 50% at 80 days[7].

Biodegradation of PLA composites with 25% cotton linter or 50% maple wood fibre were investigated. The composting experiments show that the onset time of degradation is increased[8]. The presence of lignocellulose fillers in PLA influence the degradation process at lower temperatures. Mass reduction of the composites during composting at 35°C is observed[9].

In soil burial conditions of starch and wood flour PLA composites, the addition of starch is shown to promote water diffusion with increased microbial growth and increased rate of PLA degradation[10].

[1] Qi, Xiang, Yiwei Ren, and Xingzu Wang. "New advances in the biodegradation of Poly (lactic) acid." International Biodeterioration & Biodegradation 117 (2017): 215-223.
[2] AS ISO 14855
[3] Shi, Bo, and Doris Palfery. "Temperature-dependent polylactic acid (PLA) anaerobic biodegradability." International Journal of Environment and Waste Management 10.2-3 (2012): 297-306.
[4] Karamanlioglu, Mehlika, Richard Preziosi, and Geoffrey D. Robson. "Abiotic and biotic environmental degradation of the bioplastic polymer poly (lactic acid): a review." Polymer Degradation and stability 137 (2017): 122-130.
[5] Pantani, Roberto, and Andrea Sorrentino. "Influence of crystallinity on the biodegradation rate of injection-moulded poly (lactic acid) samples in controlled composting conditions." Polymer degradation and stability 98.5 (2013): 1089-1096.
[6] Fortunati, E., et al. "Okra (Abelmoschus esculentus) fibre based PLA composites: mechanical behaviour and biodegradation." Journal of Polymers and the Environment 21.3 (2013): 726-737.
[7] Petinakis, Eustathios, et al. "Biodegradation and thermal decomposition of poly (lactic acid)-based materials reinforced by hydrophilic fillers. "Polymer Degradation and Stability 95.9 (2010): 1704-1707.
[8] Way, Cameron, et al. "Processing stability and biodegradation of polylactic acid (PLA) composites reinforced with cotton linters or maple hardwood fibres." Journal of Polymers and the Environment 21.1 (2013): 54-70.
[9] Quitadamo, Alessia, et al. "Cellulosic Waste Deriving Filler Content Effect on Biodegradation Behavior and Thermal Properties for HDPE and PLA Composites." Preprints (2019).
[10] Lv, Shanshan, et al. "Biodegradation behavior and modelling of soil burial effect on degradation rate of PLA blended with starch and wood flour." Colloids and Surfaces B: Biointerfaces 159 (2017): 800-808.

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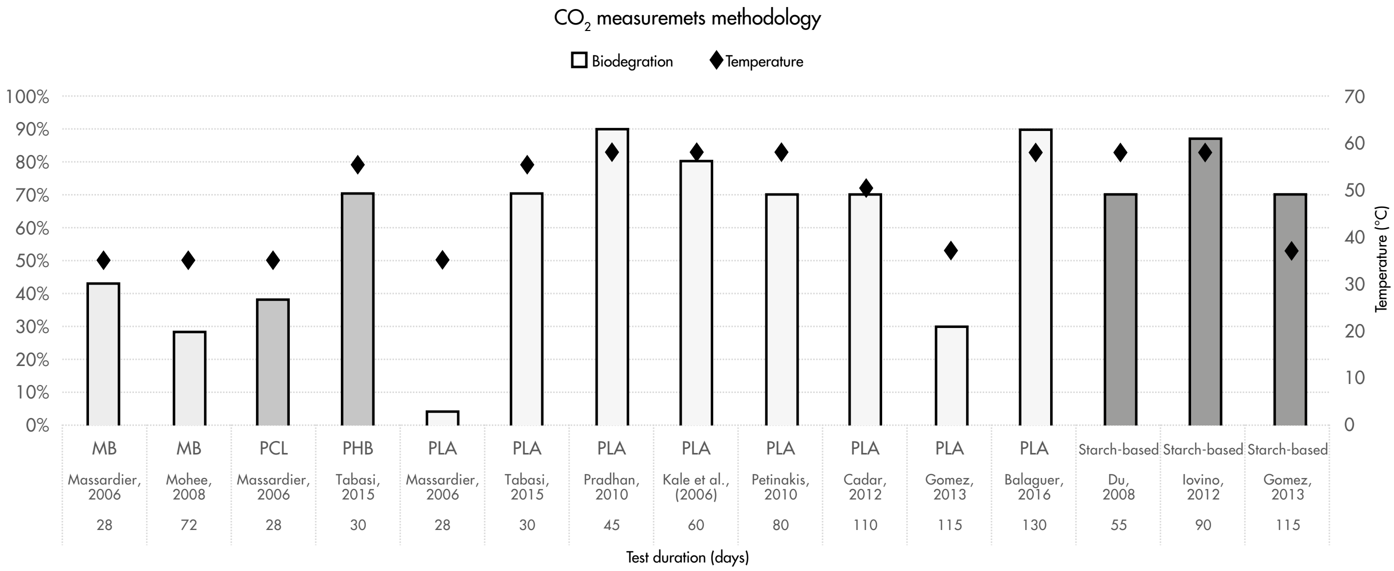
To determine the biodegradation of materials many different test have been developed[11], from visual inspection and measuring material strength loss, to weight loss and CO2 and CH4 liberation. Each method is using standardized conditions (aerobic, anaerobic, aquatic, with added enzymes, etc) that will vary in environment, test duration and severity[12]. See for detailed information : (aerobic ISO 20200; ISO 16929; ISO 14855; EN ISO 14851; EN 14806; EN 14045; EN13432; ASTM D5338; anaerobic ISO 1483; ISO 13975; EN 13432; ASM D5526-94d; ASTM D5511-02)[13].



Graph: example of biodegradation comparison of biodegradable polymers at various conditions[13]

[11] Eubeler, Jan P., et al. "Environmental biodegradation of synthetic polymers I. Test methodologies and procedures." TrAC Trends in Analytical Chemistry 28.9 (2009): 1057-1072.

[12] Calmon, Anne, et al. "Evaluation of material biodegradability in real conditions–development of a burial test and an analysis methodology based on numerical vision." Journal of environmental polymer degradation 7.3 (1999): 157-166.

[13] Ruggero, Federica, Riccardo Gori, and Claudio Lubello. "Methodologies to assess biodegradation of bioplastics during aerobic composting and anaerobic digestion: A review." Waste Management & Research 37.10 (2019): 959-975.

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It is very difficult to compare the data because of these varying methodologies, but the overall conclusions from all these studies is here summarized in below table and commented. It must be noted that exact and quantitative data are hard to give.

Table: Degradation / biodegradation rates of miscanthus and compounds compared to PP and HDPE[14], [15]

		Air-water	Water-soil	Composting	Sea-water	Comment	Mass loss
Miscanthus							
	Stem	+/-	+	++	+	Fully biodegradable	
	Chips	+	++	+++	++		
	Fibre	+	++	+++	++		
PLA-misc compounds	25 wt%	-	+	+	+	Fully biodegradable	
PLA		-	+/-	+	+	Fully biodegradable	84%, 700d
PP-misc compounds	25 wt%	-	-	-/+	-	Disintegrating/Partly biodegradable	25%, 180d
PP		-	-	-	-	Non-degradable	3%, 84d
Starch Polyester						Disintegrating/Partly biodegradable	20%, 130d
Oxo-bio		+	-	-	-	Decomposing in air	
HDPE		-	-	-	-	Non-degradable	
PET		-	-	-	-	Non-degradable	

The non-degradable polymers will persist in the environment for hundreds of years. They will be found back as such or in the form of particles or micro-particles (micro-plastics). The oxo bio-degradables will degrade to microplastic particles when exposed in the air.

[14] Calmon, Anne, et al. "Modelling easily biodegradability of materials in liquid medium-relationship between structure and biodegradability." Journal of environmental polymer degradation 7.3 (1999): 135-144.

[15] Degli Innocenti, Francesco. "Biodegradation behaviour of polymers in the soil." Handbook of biodegradable polymers'.(Ed. C Bastioli) pp (2005): 57-102.

Review of PLA- Miscanthus compounds in different end-of-life scenarios

- Case 1 : Pack ended in industrially managed compost

Industrial composting conditions of humidity, aeration and a temperature range between 50 and 60 °C, which will eliminate pathogenic organisms and decompose the organic polymers into CO_2 and H_2O. For organic recycling of bioplastics, the conditions should be following the EN 13432 norm on industrial compostability. PLA and PLA compounds with cellulosic fibres such as Miscanthus, will fully degrade within 6-12 weeks under aerobic composting conditions. The polylactic acid and polysaccharides (Cellulose – hemicellulose) present in the product will be fully digested.[1]

- Case 2 : incineration with energy recovery

In case of an end of life scenarios for PLA from post-consumer waste or post industrial waste by thermal recovery[2] in a MSWI (Municipal solid waste incineration) will yield heat (396 CO_2 eq/FU)[3] and electricity (365 CO_2 eq/FU) and release CO_2 and H_2O. In case of PLA compounds with Miscanthus the lignocellulose will contribute proportionally to the net energy recovery.

- Case 3 : incineration without energy recovery

The total PLA and Miscanthus compound will be fully converted to CO_2 and H_2O, without significant emissions of PAHs[4] or NOx.

- Case 4 : land fill / open air environment

Under common anaerobic landfill conditions the PLA does not degrade or produce biogas, due to its (semi)crystalline nature, which is difficult for micro-organisms to attack[5]. By the addition to PLA of Miscanthus particles[6], that will take up water from the environment and slowly decompose under these conditions, the compound may disintegrate and decompose faster[7]. There is no supporting studies identified yet, except for under aerobic composting conditions[8].

[1] https://www.european-bioplastics.org/bioplastics/waste-management/composting/

[2] https://www.natureworksllc.com/What-is-Ingeo/Where-it-Goes/Incineration

[3] Maga et al 2019 - https://www.sciencedirect.com/science/article/pii/S0921344919302319

[4] Chien, Yi-Chi, et al. "Combustion kinetics and emission characteristics of polycyclic aromatic hydrocarbons from polylactic acid combustion." Journal of the Air & Waste Management Association 60.7 (2010): 849-855.

[5] Kolstad, Jeffrey J., et al. "Assessment of anaerobic degradation of Ingeo™ polylactides under accelerated landfill conditions." Polymer Degradation and Stability 97.7 (2012): 1131-1141.

[6] Van den Oever, M. J. A., B. Beck, and J. Müssig. "Agrofibre reinforced poly (lactic acid) composites: Effect of moisture on degradation and mechanical properties." Composites Part A: Applied Science and Manufacturing 41.11 (2010): 1628-1635.

[7] Yussuf, A. A., I. Massoumi, and A. Hassan. "Comparison of polylactic acid/kenaf and polylactic acid/rise husk composites: the influence of the natural fibers on the mechanical, thermal and biodegradability properties." Journal of Polymers and the Environment 18.3 (2010): 422-429.

[8] Fortunati, E., et al. "Okra (Abelmoschus esculentus) fibre based PLA composites: mechanical behaviour and biodegradation." Journal of Polymers and the Environment 21.3 (2013): 726-737.

In open air the effects of UV light will cause oxidation and decomposition of the PLA polymer[9] and lignocellulose particles. The compound will become brittle and slowly degrade into its elements.

- Case 5 : marine environment

In marine environments the ageing of flax/PLA biocomposites was studied. The moisture uptake by the fibres results in faster decrease of mechanical properties[10]. The ASTM D-7081 standard test measurement of CO_2 evolution (ASTM D-6691) shows over 30% degradation after 180 days for PLA in marine environmental conditions[11]. Similar data are found for cellulose (paper).

[9] Copinet, Alain, et al. "Effects of ultraviolet light (315 nm), temperature and relative humidity on the degradation of polylactic acid plastic films." Chemosphere 55.5 (2004): 763-773.
[10] Le Duigou, Antoine, et al. "Long term immersion in natural seawater of Flax/PLA biocomposite." Ocean Engineering 90 (2014): 140-148.
[11] Greene, Joseph. "Marine Biodegradation of PLA, PHA, and Bio-additive Polyethylene Based on ASTM D7081." (2012).

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