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Research Article

Bioabsorbable Characteristics of Poly (Lactic Acid) (PLA) for a Fundamental Solution to the Problem of Microplastics Tea Bag SOILON® Made from PLA Fibers

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Abstract

The aim of this research was to solve the problems of accidental ingestion of microplastics such as in case of tea bag filter, in addition to the solid wastes management of food packaging plastics after use. PLA was not only aerobic compostable but also anerobic digested to biogas by methane fermentation at 50°C after a month, similar to that in composting at 60°C. Also, PLA was hydrolytic degraded in physiological saline solution at 37°C, that is the simulated condition of human body. Due to the relative slow degradation rate of PLA, its medical applications have been mainly to bone-fixation, taking about three months. The degradation of PLA in the human body by hydrolytic scission of ester linkage yielded lactic acid which was a naturally occurring acid without ill effects. It was suggested that PLA would remain temporarily in the body and disappear upon biodegradation followed by bioabsorption.

Keywords

Biodgradable, Bioabsorbable, Microplastics, Tea Bag, Poly (Lactic Acid)

1. Introduction

For nearly 40 years, oil-based synthetic fibers such as nylon and polyester have been used for tea bag filter applications. The current crisis in the management of solid waste including conventional oil-based plastics has focused attention on the development of bio-based biodegradable plastic like poly (lactic acid) (PLA) [1-5]. PLA has been highlighted as a sustainable carbon-neutral biodegradable plastic without increase in atmospheric CO_2 levels because of its availability from agricultural renewable resources like corn. PLA is a biodegradable aliphatic polyester with thermoplastic processability and has favorable both mechanical properties and biodegradation characteristics [6, 7].

Figure 1. *Tea Bag SOILON® (YAMANAKA INDUSTRY).*

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In 1999, YAMANAKA INDUSTRY CO., LTD. has launched the world's first tea bag, SOILON®, mesh type of woven fabric composed of PLA fibers (Figure 1) [6, 7]. The SOILON® intends for the safety and security by using PLA from non-GMO (non-genetically modified) grains. The tea bag of SOILON® is compostable with other organic wastes after use and eco-friendly since it does not increase atmospheric CO_2 levels.

Whereas, over the past few years, new problems are arising. Humans, at the top of the food chain on earth, are inevitably exposed to danger of accidental ingestion of microplastics, because animals or fishes tend to ingest anything by mistake and we also have a risk of accidental ingestion of them from food packaging materials. In 2018, researchers from the Environment Agency Austria and the Medical University of Vienna studied eight people from different parts of the world and found microplastics (PP, PET etc.) in all the participants' bodies, in which the average number of microplastic was 20 pieces out of 10g stool sample [8].

In 2019, scientist from McGill University, Canada reported that plastic teabags released billions of micro/nanoplastics (MNPLs) and also called for more investigation into the health effects of MNPLs [9]. Furthermore, scientists from Universitat Autònoma de Barcelona, Spain investigated the release of MNPLs from three commercially available teabags and found many MNPLs in the leachates recently [10]. The leachate particles were then stained and used to expose human intestine-derived cell in order to assess their biointeractions. The results demonstrated that after 24 h of exposure to MNPLs, there was significant uptake of them in the cells. They are warming human health problems caused by teabag-derived MNPLs.

Recent reports highlight the generation of micro/nanoparticles even during the normal use of plastic food contact materials, such as water bottles, teabags, and food containers. The teabag filters were analyzed by SEM to detect surface irregularities that potentially induce breaks and release of particulate material. These structures suggest that some imperfections might appear due to the friction with hard guides of machines during fiber spinning followed by weaving process in which the fibers are running at high speed.

These problems have been focusing attention on the biodegradable followed by bioabsorbable plastics in human body without ill effects, even if you had them accidentally. It's notable that PLA is not only biodegradable but also bioabsorbable in human body without ill effects. In this paper, it will be described in respect of bioabsorbable performances of PLA in human body, in addition to biodegradation mechanism and characteristics in composting or in methane fermentation for bio-recycling after use.

2. Materials and Methods

2.1. Tea Bag SOILON®

Tea bag SOILON® is a mesh type of woven fabric made from PLA monofilaments by melt spinning. PLA resin used does not contain any additives and the oil used for spinning and weaving process was removed in the course of scouring process after weaving. Tea bag preparation was achieved by ultrasonic shealing in two shapes, tetrahedral bag like pyramid and conventional tetragonal bag, as shown in Figure 1.

2.2. Biodegradation Tests

2.2.1. Aerobic Composting

The compostability of PLA nonwovens was evaluated under simulated composting conditions. Evaluation of the ultimate aerobic biodegradation and disintegration was conducted by measurement of released carbon dioxide at 60±2°C after 45 days, according to ISO 14855 [11]. This test method is designed to yield a percentage and rate of conversion of carbon of the substance to released carbon dioxide. Also, Aerobic biodegradation of PLA fabric in compost at 58±2°C was carried out in comparison with those of cotton, wool, and PET [3]. Finally, Biodegradation behavior of SOILON® after ruse in compost at 60°C was evaluated.

2.2.2. Anerobic Digestion Such as Dry Methane Fermentation at High Temp

The ultimate anaerobic biodegradability and disintegration under high-solids, anaerobic-digestion based on carbon conversion of PLA to methane and carbon dioxide was evaluated according to ISO 15985 [11]. Recently, dry methane fermentation biogas power generation facility which decompose PLA products after use with other organic wastes at high temperature (50-55 ℃) by anaerobic microorganisms has been constructing [12, 13], in which biogas generation of SOILON® in the dry methane fermentation at 50 ℃ was evaluated.

2.2.3. Hydrolytic Degradation of PLA in Physiological Saline Solution at 37°C

The most common synthetic biodegradable polymers in medical applications are the poly (α-hydroxy acid) s, including poly (glycolic acid) (PGA) and poly (lactic acid) (PLA). The rate of hydrolysis of PGA and PLA fibers in physiological saline solution at 37°C, that is the simulated condition of human body, was evaluated, in order to presume the rate of degradation of microplastics in human body when you had them accidentally.

3. Results and Discussion

3.1. Composting or Methane Fermentation of PLA

Figure 2 shows aerobic composting test result of PLA fibers according to ISO 14855.

The cumulative biodegradation of PLA nonwovens compared with cellulose powder as a positive reference indicates that the biodegradability of PLA is nearly equal to cellulose after 45 days. [11]

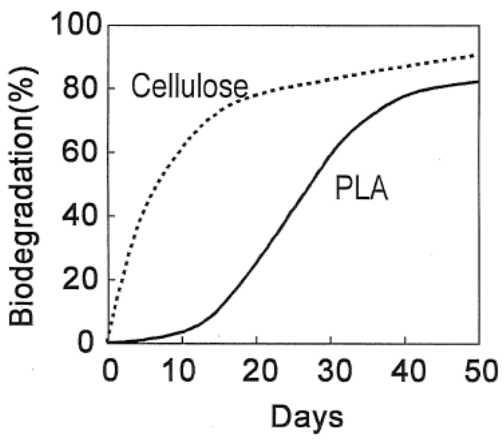

Figure 2. Aerobic composting test result. of PLA fibers according to ISO 14855.

SOILON® has been successfully composted in applications where a municipal or industrial composting infrastructure controlling high temperature and high humidity is in place, as shown in Figure 4. SOILON® tea bag filter with string and tag can be wholly compostable because both the paper tag and the string are also composed of PLA-based materials. In 2014, the tea bag SOILON® made from PLA fibers has been certified in accordance with the industrial composting standards, EN13432 by Din Certco and ASTM D6400 by BPI, in addition to OK Compost by Vincotte. It is the world's first tea bag filter as a compostable product in municipal and industrial composting facilities [7]. However, it is not recommended for use in typical backyard composting due to lack of controlling of temperature and humidity under inconsistent conditions.

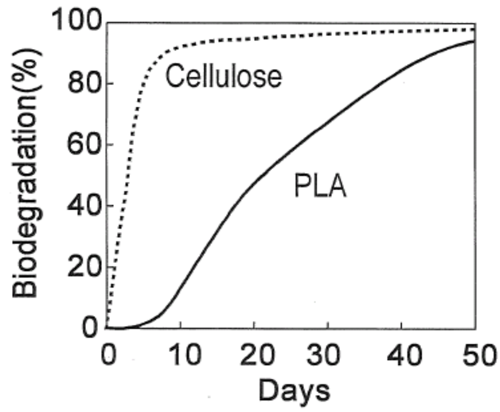

Figure 3. Anaerobic digestion test result of PLA fibers according to ISO 15985.

Before After 15 days After 30 days

Figure 4. Biodegradation behavior of SOILON® in compost at 60°C.

PLA is completely biodegradable when exposed in biologically active environments such as municipal compost facilities which regulate temperature, moisture, and aeration. In fact, PLA fibers are degraded more rapidly than other natural biodegradable fibers such as wool or cotton in compost. Figure 5 shows the result of actual aerobic fresh composting of PLA fabric at 58°C in comparison with wool, cotton and PET ones. The PLA fabric has been degraded more rapidly than other biodegradable natural fibers like wool or cotton. It means that PLA fibers have an excellent compostability characteristic for bio-recycling such as composting or methane fermentation after use [2, 3].

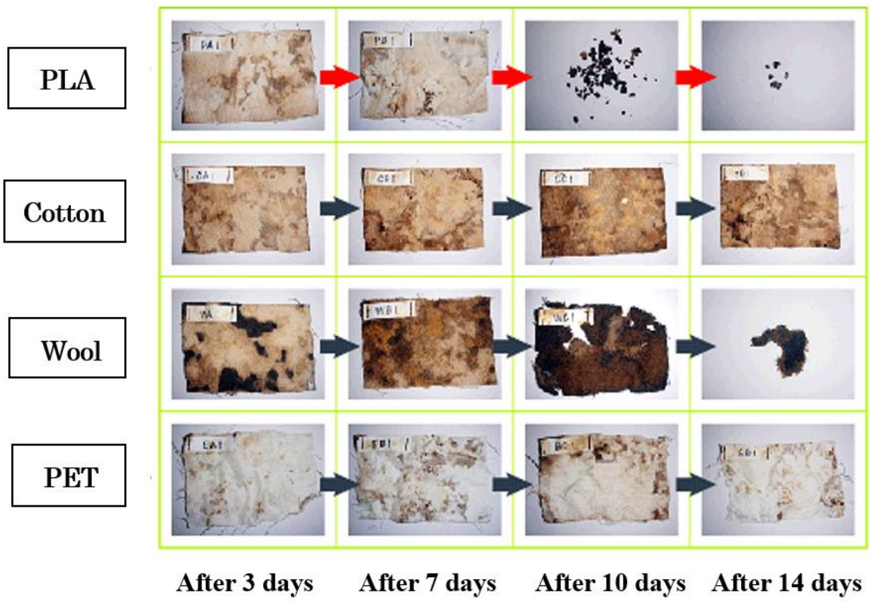

Figure 5. *Aerobic biodegradation of PLA fabric in compost at 58±2°C in comparison with those of cotton, wool, and PET.*

On the other hands, evaluation of the ultimate anaerobic biodegradability and disintegration under high-solids was based on carbon conversion of PLA to methane and carbon dioxide according to ISO 15985. The result represented in Figure 3 shows that anaerobic-digestion of PLA occurs at 52±2°C under high solids and static, non-mixed conditions, which is nearly equivalent to that of cellulose after 45 days [11].

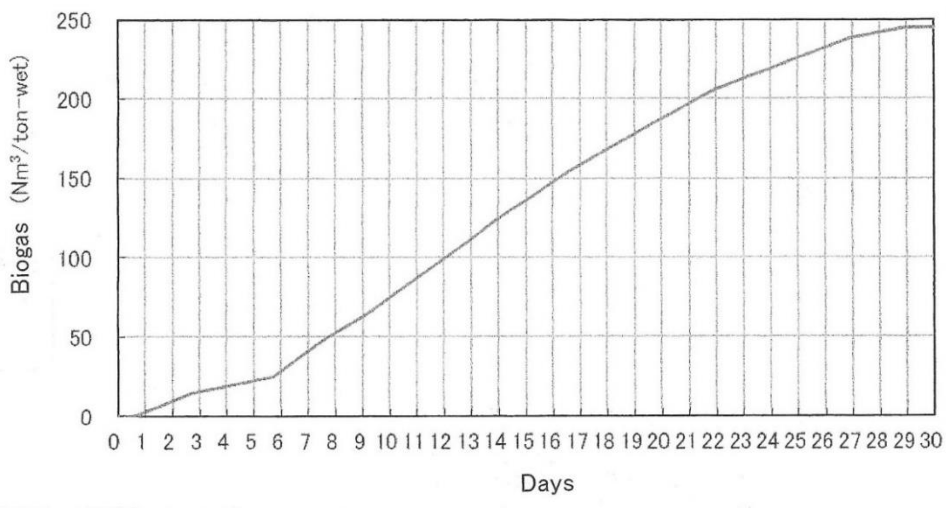

Figure 6. *Biogas generation of SOILON® in the dry methane fermentation at 50°C.*

Figure 6 shows the result of biogas generation of SOILON® in the dry methane fermentation at 50°C by ORIX Corporation. It can be seen that PLA could be readily anaerobic-digested to biogas, mainly composed of methane, after a month similar to that in composting at 60°C.

3.2. Biodegradation Mechanism of PLA

Under typical use and storage conditions like room temperature, PLA is stable. However, under very specific conditions of high temperature (55°C <) and high humidity (80% RH<), typified by a composting condition, PLA will start disintegration within around 2 weeks (10-20 days), followed by bacterial digestion of the fragmented residues having low molecular weight oligomers and monomer to give carbon dioxide and water after around 5 weeks (30-45 days), as shown in Figure 7 [14].

Figure 7 shows the typical two-step biodegradation curve of PLA under composting conditions at 60°C. In the primary degradation phase, PLA undergoes chemical hydrolysis which is both temperature- and humidity-dependent, and does not involve any enzymatic degradation by microorganisms. As the number-average molecular weight (M_n) reaches approximately 10 000-20 000 after around 2 weeks, microorganisms present in the soil begin to digest the lower-molecular weight oligomers and monomer, producing carbon dioxide and water.

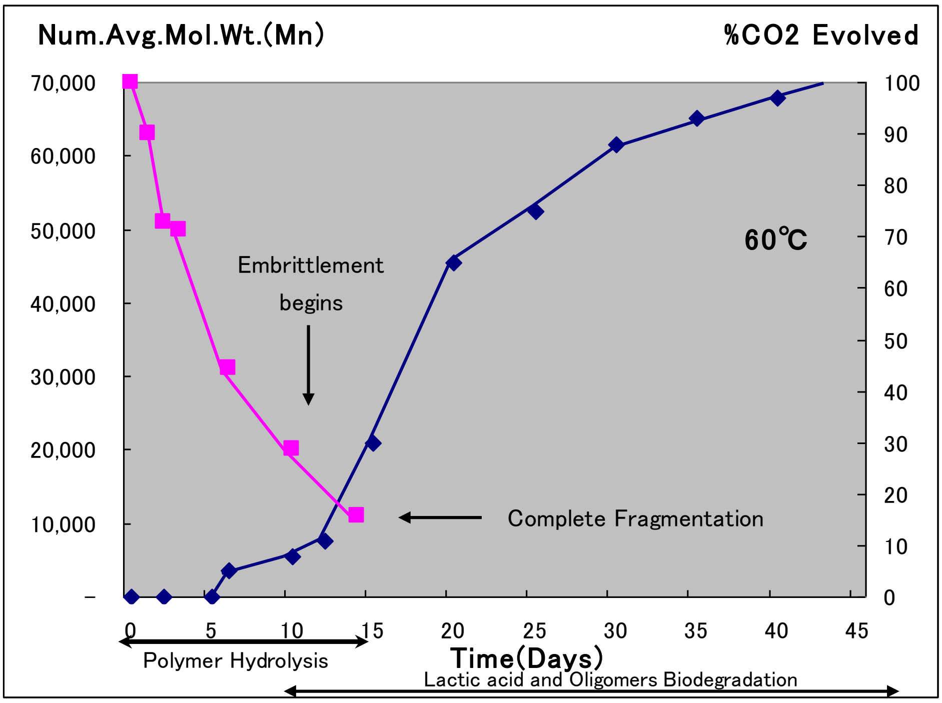

Figure 7*. Biodegradation of PLA in compost at 60°C.*

This two-step biodegradation mechanism differs distinctly from those of other biodegradable polymers which degrade by direct bacterial attack with enzymatic degradation on the polymer surface, that is a single-step surface erosion process [15]. Few reports exist the microbial enzymatic degradation of high-molecular weight PLA.

3.3. Hydrolytic Degradation of PLA in Physiological Saline Solution

Figure 8 shows the rate of hydrolysis of PGA and PLA fibers in physiological saline solution at 37°C, that is the simulated condition of human body. The longest and the largest medical application of biodegradable polymers is in suture. The synthetic polymer used for this purpose is mainly PGA and its copolymer with PLA (90:10). The faster rate of degradation of PGA just much the rate of tissue regeneration, only taking a week. PLA fibers are not suitable for sutures, because they degrade very slowly when compared with PGA, as shown in Figure 8. Due to the slow degradation rate of PLA, its medical applications have been used mainly to bone-fixation, taking about three months [16, 17]. Figure 8 shows that PLA maintained the strength more than 90% of its initial strength for three months required for bone fixation, then decreased slowly.

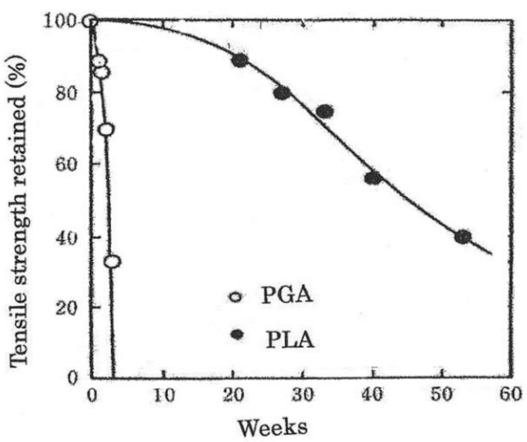

Figure 8*. Changes in tensile strength of PGA and PLA fibers in physiological saline solution at 37°C.*

3.4. Bioabsorbable Performances of PLA for Medical Applications

Biodegradable materials have been studied extensively for medical applications over the past five decades. Their advantages over nondegradable biomaterials include eliminating the need to remove implants and providing long-term biocompatibility. The most important requirement for medical applications is that the degradation rate of biodegradable implants must match the rate of tissue regeneration. Secondly, the material released from biodegradable implants never cause toxic effects in the body [16, 17].

PLA has shown to be the most promising due to its relatively strong mechanical properties for many medical implants such as bone fixation for the fractured bones to regenerate, and is approved by regulatory agencies in many countries. Recently, PLA has been replacing metallic ones for the fixation of fractured bones in the forms of plates, pins, screws, and wires, as shown in Figure [18].

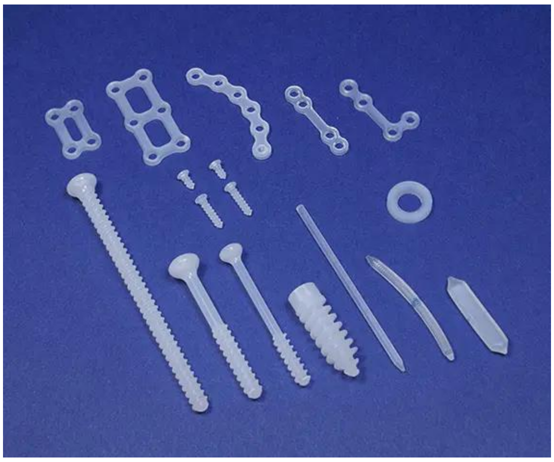

Figure 9. *PLA screws, rods, and miniplates for bone fixation (GUNZE MEDICAL LIMITED).*

4. Conclusion

PLA is an aliphatic polyester which is the synthetic polymer of lactic acid. The degradation of PLA in the human body by hydrolytic scission of ester linkage yields lactic acid. Lactic acid is a naturally occurring acid associated with muscular construction in animals and humans, which can be decomposed by the body's normal metabolic pathways. In the body, lactic acid is converted to pyrubic acid and enters the tricarboxylic acid (TCA) cycle to yield carbon dioxide and water. This conversion yields chemical energy in the form of ATP (adenosine triphosphate) for cell growth and maintenance.

Other compounds present in the PLA polymer are trace amounts of catalyst. Most effective and commonly used catalyst for ring opening polymerization of lactide (the cyclic dimer of lactic acid) are based on tin 2-ethylhexanoate. Due to its low toxicity, tin 2-ethylhexanoate has been approved by the FDA [17]. Other hazardous substances such as endocrine disrupters are not included.

PLA is basically harmless and no ill effects on the human body if you had a small amount of microchip or fragment of PLA accidentally, because of its biodegradable followed by bioabsorbable characteristics. PLA remains temporarily in the body and disappear upon biodegradation followed by bioabsorption. As you know well, almost all foods not only fermented foods like cheese or yogurt, but also natural foods like beef steak contain lactic acid. Also, there are many lactic acid bacteria like Lactobacillus to promote digestive health in our intestines.

Lactic acid is a naturally occurring acid with unique taste and flavor which is essential for all life on earth. Professional tasters told us that SOILON® tea bag filter made from PLA fibers offers a pure taste and flavor without offensive smells, when compared with those from oil-based synthetic fibers like polypropylene (PP), nylon or polyethylene terephthalate (PET) reeking a little bit offensive smell.

We are now in the midst of new frontier of material revolution contributing to new filter for both tea and coffee, aiming for harmonious coexisting with nature. We are convinced that PLA is the key material with hidden possibilities [19, 20].

Abbreviations

PLA	Poly (Lactic Acid)
PGA	Poly (Glycolic Acid)
MNPLs	Micro/Nanoplastics
SEM	Scanning Electron Microscope
FDA	Food and Drug Administration

Author Contributions

Masatsugu Mochizuki is the sole author. The author read and approved the final manuscript.

Conflicts of Interest

The authors declare no conflicts of interest.

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Biography

Masatsugu Mochizuki graduated from Kyoto University in 1986 with a degree in polymer chemistry. After graduation, he joined Unitika Ltd as a researcher in R&D and got his Doctor of Engineering from Kyoto Univ. in 1995. He has continued research on the development of biodegradable plastics and their applications multidisciplinary in industry and academia to solve environmental issues such as solid wastes management for 38 years since 1986 and published many books and papers as a pioneer in the world. After retirement from Unitika, he was invited to Research Professor at Kyoto Institute of Technology in 2007 and is currently a technical adviser of YAMANAKA INDUSTRY Co., LTD.

Research Field

Masatsugu Mochizuki: Polymer Chemistry, Biomaterials, Biodegradable Polymers, Bioabsorbable Polymers, Material Designing, Polymer Processing, Fibers and Nonwovens, Applications