Biodegradable plastics from linier low-density polyethylene and polysaccharide: The influence of polysaccharide and acetic acid

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ABSTRACT

Global problems associated with conventional, non-biodegradable plastics have urged the society to use more eco-friendly biodegradable plastics. In this study, linear low-density polyethylene (LLDPE) was co-compounded with cassava-based thermoplastic starch (TPS) to prepare biodegradable plastics (i.e. plastics that can be degraded by microbes), in which three different LLDPE/TPS ratios were studied. Acetic acid was used to hydrolyze the polysaccharides by breaking the branched amylopectin that causes the TPS-containing composites brittle and stiff. The biodegradation properties of the LLDPE/TPS composites were determined by observing the level of microbial growth on the sample surface after incubation with potato dextrose agar medium that was inoculated with *Penicillium* sp. and *Aspergillus niger*. Burial test in a humid composting medium was also performed to validate the biodegradation properties. The mass change (%) was calculated in relative to the initial mass before burial test. The physical properties (tensile strength and elongation at break) of the bioplastics were determined using universal testing machine before and after burial treatment. The morphology of the sample surface was evaluated using scanning electron microscopy. The results showed that the microbial growth increases with increasing TPS content. Negative mass changes were observed on all samples that contain TPS, with increase in the magnitude with increasing TPS content. The tensile strength tends to increase in the first 28 days of burial period in a composting medium then decreases and plateaus, while the elongation at break decreases with increasing burial period. Moreover, samples that contain acetic acid showed less microbial attachment and less biodegradation compared to samples that does not contain acetic acid.

Keywords: LLDPE, cassava starch, acetic acid, biodegradability, compost burial.

INTRODUCTION

Environmental pollutions caused by plastics wastes is one of global problems that needs serious concerns. The plastics wastes found in ground water and even inside the digestive system of sea animals in recent days, in the form of either macro- or micro-plastics (Frias & Nash, 2019), is a problem not only in Indonesia but also around the globe (Botterell et al., 2019; Cho et al., 2019; Guzzetti et al., 2018; Karthik et al., 2018; Li et al., 2019; Rainieri & Barranco, 2018; Sloodmaekers et al., 2019; Sun et al., 2019; Teng et al., 2019; Wang et al., 2019; Windsor et al., 2019). Apart from development of recycling technologies for non-biodegradable plastics (Horodytska et al., 2018; Mwanza et al., 2018; Ragaert et al., 2017; Silveira et al., 2018; Thakur et al., 2018), development of biodegradable plastics (Chinaglia et al., 2018) is one of important solutions in coping with plastics wastes. According to ASTM standard (D6813), biodegradable plastics refer to polymeric materials that are “capable of undergoing decomposition into carbon dioxide, methane, water, inorganic compounds, or biomass in which the predominant mechanism is the enzymatic action of microorganisms, that can be measured by standardized tests, in a specified period of time, reflecting available

disposal condition” (Song et al., 2009).

The technology used for developing biodegradable plastics can be categorized into biotechnology and compounding. Biotechnology refers to the technology that uses enzymes and microorganisms to produce the polymer materials for the plastics products, such as poly(lactic acid) (PLA) and poly(hydroxyalkanoates) (PHA). The drawbacks of biotechnology are for example the possibility of occurring genetically modified organism (GMO) and transgenic plants with uncertain safety for humans. The investment funds needed for this type of technology is also high, and not yet applicable in Indonesia. On the other hands, compounding technology refers to the technology that blends and finds the optimum formulations between (bio)polymers in order to achieve the aimed biodegradation properties. There have been several company start-ups in Indonesia for this type of technology, namely Evoware, Enviplast, and Avani.

Polysaccharides and polyamides are among biopolymers that are often used to prepare biodegradable plastics (Ashter, 2016). While, linear low-density polyethylene (LLDPE) is one of synthetic polymers that have been widely used in the preparation of plastics that should meet properties like highly flexible, possess high

elongation at break, able to form thinner films compared to other types of polyethylenes, resistant to chemical attacks, transparent, and inexpensive. However, plastics made of solely LLDPE are, in general, non-biodegradable when disposed to the environment. Blending LLDPE with a polysaccharide- (starch-) based polymer has been reported to increase the biodegradation level of LLDPE (Nguyen et al., 2016). Cassava starch consists of amylose and amylopectin, in which the latter is significantly more brittle than the former due to its highly branched chemical structure. Thus, bioplastics with a high amylopectin content present low mechanical properties (Tanetrungroj & Prachayawarakorn, 2018). In order to reduce this problem, the starch should be hydrolyzed prior to blending and compounding with other polymer(s). The hydrolysis should lead to chain scission of the highly branched amylopectin and decrease in the brittleness of the material.

Acidic hydrolysis of starch can be performed using, for example, acetic acid. The acid attacks the amorphous (amylopectin) part that covers the crystalline (amylose) part, and then it attacks both amylopectin and amylose leading to the formation of shorter polymer chains (smaller molecules). It also facilitates the formation of linear polymer chains and further formation of bioplastics (Wang & Copeland, 2015). It has been previously reported that processing bioplastics with acetic acid increases the solubility of starch (Prabha & Ranganathan, 2017). Apart from its role as hydrolyzing agent, it is known that acetic acid possess antimicrobial properties (Bjarnsholt et al., 2015; Halstead et al., 2015). Hence, the presence of acetic acid should influence the biodegradability of bioplastics.

In this study, bioplastics were fabricated from LLDPE and thermoplastic starch (TPS) from cassava. Three different ratios of LLDPE/TPS were evaluated, i.e. 70/30, 50/50, and 30/70. Furthermore, the influence of acetic acid to the performance of bioplastics was also investigated. The biodegradability of LLDPE/TPS composites was determined using burial method in a composting medium according to previously reported protocol (Dilfi et al., 2017; Nguyen et al., 2016; Oragwu, 2016). Nguyen *et al.* (2016) reported that LDPE/starch-based bioplastics showed more biodegradation in a composting medium compared to in normal soil. The mass change, morphology, and tensile strength of the bioplastics were measured before and after burial tests.

MATERIALS AND METHODS

Materials

LLDPE resin Asrene UF 1810T (extrusion grade, density 0.921 g/cm³, and melt index 1 g/10 min (190°C/2.16 kg)) was purchased from PT Chandra Asri. Orang Tani® starch powder was used for preparing TPS formulations. Compatibilizer Lotrader 4210 and antioksidan Songnox 1010 were purchased from PT Bunga Permata and PT. Intera Lestari Polimer, respectively. Aquadest, stearic acid, and glycerol were purchased from Bratachem. Furthermore, TPS1 and TPS2 were prepared, following the

formulations as described in the “composite preparation” section.

Equipment

Internal mixer Haake Rheomix OS, hydraulic press, universal testing machine (UTM) Tinius Olsen-H25K, and Scanning Electrone Microscope (SEM) JSM06510LA were used during the compounding, pressing, physical testing, and morphological testing, respectively.

Methods

Composite preparation

LLDPE/TPS composites were prepared in an internal mixer at 130 °C and 40 rpm for 15 min. Based on experimental results in a rheomix at 40 rpm, the change of the process torsion showed a stable process at 130 °C. Too high and too low temperature may cause starch degradation and unmelted polymers, respectively. Three different LLDPE/TPS ratios were used, *i.e.* 70/30; 50/50 and 30/70. Prior to mixing with LLDPE, the starch powder was hydrolyzed using acetic acid solution. As a control, un-hydrolyzed starch powder was included in the study. Thus, two TPS formulations were prepared, *i.e.* TPS1 and TPS2. The former (TPS1) consists of starch 70%, water 9%, acetic acid 1%, and glycerol 20%, while the latter (TPS2) consists of starch 71%, water 9%, and glycerol 20% (TPS2 does not contain acetic acid). The starch, water, and acetic acid were mixed until a homogenous solution was obtained. The glycerol was then added and the solution was stirred for 15 min. Prior to addition of LLDPE, the solution was stored for 2 days to allow further mixing and hydrolysis of the starch. During compounding, other additives such as compatibilizer, antioxidant, and stearic acid were added. After the compounding process, a 1 mm-thick composite film was prepared using hydraulic press machine. Dumbbell specimens type 5 (Figure 1) were than prepared from the film using a punching machine.

Biodegradability test in potato dextrose agar (PDA)

The test in a PDA medium was performed following ASTM G-21 To this end, a 3 x 3 cm sample was placed on the PDA medium that was inoculated with either *Penicillium* sp. or *Aspergillus niger*. The sample was incubated at 40°C. After 2 and 6 weeks, the microbial growth on the sample surface was observed and quantified as follows: 0 refers to samples with no microbial growth; 1 refers to samples with ≤10% microbial surface coverage; 2 refers to samples with 10-30% microbial surface coverage; 3 refers to samples with 30-60% microbial surface coverage; and 4 refers to samples with 60-100% microbial surface coverage.

Biodegradability test in composting medium

The burial test using composting medium was performed at a laboratory scale using modified Amer and Saeed (2015) method. The composting medium consists of manure, husk charcoal, sand, and cocopeat, making the medium suitable for growing microbes such as fungi and

bacteria. Thus, a humid composting medium was added in a plastic container. Dumbbell specimens with standardized dimensions were prepared for each sample. The specimens were then buried inside the composting medium at a 10-cm depth from the air-composting medium interface. After an outdoor burial test for 56 days, the specimens were taken out and cleaned carefully to remove attached soil particles. Conditioning of specimens was performed at 23 ± 2 °C for 24 h. The biodegradability was observed based on the changes in mass, tensile strength, elongation at break, and morphology.

Mass change

The mass change of the composite after burial tests was calculated according to equation 1.

$$\text{Mass change} = [(B_0 - B_1)/B_0] \times 100\% \quad (1)$$

where B_0 and B_1 are the mass of the sample before and after burial test, respectively.

Tensile strength and elongation at break

The tensile strength and elongation at break of the samples were performed using a UTM following a standard procedure ASTM D 882 (type 5 Dumbell specimen was used).

Morphology

The morphology of the samples was observed before and after 14, 42, and 56 days of burial test, using SEM.

RESULTS AND DISCUSSION

Biodegradability Tests in PDA Medium

The LLDPE/TPS composites were contacted with PDA medium and incubated at 40 °C. Organoleptic observation was then performed, *i.e.* the visible microbial growth on the surface of the samples was monitored from time to time using bare eyes (Figure 1) and quantified based on the microbial surface coverage (Table 1). Figure 1 and Table 1 show that the microbial surface coverage of the samples increases with increasing TPS (starch) content on the LLDPE/TPS composite samples, both on LLDPE/TPS1 and LLDPE/TPS2. Although the microbial surface coverage on a sample does not directly mean that the sample is biodegradable, it does indicate that the sample attracts microbes, and this may lead to biodegradation of the sample by the microbes. Thus, Figure 1 and Table 1 indicate that higher content of starch leads to higher biodegradability of LLDPE/TPS composite samples. Furthermore, higher microbial surface coverage on the TPS-containing samples were observed after 42 days of incubation time compared to after 14 days of incubation time. This phenomenon validates the fact that starch is a suitable substrate for microbial growth and, expectedly, enzymatic actions that leads to biodegradation of the samples. No microbial surface coverage was observed on LLDPE samples. This indicates that LLDPE is not a

suitable substrate for microbial growth, possibly due to the fact that LLDPE cannot be hydrolyzed and digested by the microbes. Interestingly, despite the role of acetic acid as a hydrolysis agent, LLDPE/TPS1 samples that contain acetic acid show, in general, lower levels of microbial growth compared to LLDPE/TPS2 samples that contain no acetic acid. This observed phenomenon might be due to the antimicrobial activity of acetic acid, thus it inhibits the microbial growth (Al-Rousan et al., 2018; Juniawati et al., 2017).

Mass change

The changes in mass of LLDPE/TPS composite samples after burial treatment in composting medium were measured as one of indicators for biodegradation. The mass change values as a function of both burial period and TPS content for LLDPE/TPS1 and LLDPE/TPS2 samples are presented in Figure 2a and 2b, respectively. In both figures it is seen that while there are no mass changes observed for LLDPE with no TPS content (0%), there are negative changes of masses for all samples that contain TPS, with increasing burial period. The negative changes are more prominent at samples with higher TPS content, indicating the role of TPS in the biodegradability of LLDPE/TPS composites. Furthermore, it is seen that the negative mass changes in Figure 2b (LLDPE/TPS2) are relatively more prominent in comparison to those in Figure 2a (LLDPE/TPS1). Since TPS1 contains acetic acid while TPS2 does not, this observed phenomenon further indicates the role of acetic acid in inhibiting biodegradation process of the samples, due to its antimicrobial properties. These results are in agreement with the publication of Tawalkatu *et al.* (2015) that reported reduced biodegradability of acetic acid-and-acetic anhydride-modified starch compared to the unmodified one.

Morphology

The morphology of the samples before and after burial treatment in composting medium were studied using SEM method, with 5000× magnification. The SEM images obtained are presented in Figure 3.

It is seen in Figure 3 that the heterogeneity (roughness) of the sample surface, both before and after burial treatment, tends to increase with increasing TPS content. Moreover, for all LLDPE/TPS samples, more heterogeneous (rougher) samples are observed after 56 days burial treatment, compared to the initial sample (*i.e.* before burial treatment). Comparing LLDPE/TPS1 and LLDPE/TPS2 samples, the latter present more heterogeneity after the burial treatment. In line with the results presented in Figure 1 (qualitative organoleptic analysis after incubation treatment in PDA medium) and Figure 2 (quantitative mass change analysis after incubation treatment in PDA medium), the more heterogeneous morphology after burial treatment indicates more biodegradation due to the absence of acetic acid. Similar results were reported by Roy *et al.* (2015) that presented the SEM micrographs of polypropylene/potato starch. The authors reported more

Tabel 1. The quantitative level of microbial growth based on the microbial surface coverage.

Composite materials	Level of microbial growth		Level of microbial growth	
	<i>Penicillium sp.</i> after incubation time		<i>Aspergillus niger</i> after incubation time	
	14 days	42 days	14 days	42 days
LLDPE/TPS1 (w/w)				
70/30	1	3	1	3
50/50	1	4	2	4
30/70	1	4	3	4
LLDPE/TPS2 (w/w)				
70/30	1	3	1	3
50/50	3	4	3	4
30/70	4	4	2	4
LLDPE				
	0	0	0	0

Note:

0 : no microbial growth

1 : $\leq 10\%$ microbial surface coverage ($\leq 10\%$ of the sample surface is covered by the microbes)

2 : 10-30% microbial surface coverage

3 : 30-60% microbial surface coverage

4 : 60-100% microbial surface coverage

fractures, holes, and degradation on the morphology images of the samples after biodegradation process.

Tensile Strength and Elongation at Break

The tensile strength (TS) and elongation at break

(EB) of the samples were measured using a universal testing machine to study the physical properties before and after burial test. The TS values of LLDPE/TPS samples as a function of burial period and TPS content are presented in Figure 4a and 4b for LLDPE/TPS1 and LLDPE/TPS2,

LLDPE/TPS	<i>Penicillium sp.</i>				<i>Aspergiles niger</i>			
	Incubation time (days)				Incubation time (days)			
	14		42		14		42	
LLDPE/TPS1 (w/w)								
70/30
50/50
30/70
LLDPE/TPS2 (w/w)								
70/30
50/50
30/70
LLDPE

Figure 1. Macroscopic (camera) images of the sample surfaces after biodegradability test in PDA medium inoculated with *Penicillium sp.* and *Aspergillus niger* for 14 and 42 days.

respectively. The EB values of LLDPE/TPS samples as a function of burial period and TPS content are presented

in Figure 5a and 5b for LLDPE/TPS1 and LLDPE/TPS2, respectively.

Figure 2. The mass change of samples as a function of burial period and TPS content for a) LLDPE/TPS1 (contains acetic acid) and b) LLDPE/TPS2 (does not contain acetic acid).

Materials	Burial period (days)	
	0	56
LLDPE/TPS1		
70/30		
50/50		
30/70		
LLDPE/TPS2		
70/30		
50/50		
30/70		
LLDPE		

Figure 3. Scanning electron micrographs of samples at different LLDPE/TPS ratios before (0 day) and after (56 days) burial treatment.

It is seen in Figure 5 that the TS tends to increase with increasing burial period up to approximately 28 days. According to Vargha *et al.* (2016) the TS increase after several days of burial treatment might be due to an initial crosslinking between the polymer chains. Longer burial period (> 28 days) results in decrease of TS values.

It is worthwhile to note that the absorption of humidity from the composting medium into the samples promote two separated effects, *i.e.* increase in plasticity (due to the insertion of H₂O molecules between the polymer chains) and increase crystallinity (due to the chain scission of the long and bulky polymer chains), known as retrogradation. An increase in plasticity results in increase of TS values, while an increase in crystallinity

results in decrease of TS values.

It is seen in Figure 5 that the EB values decreases with increasing burial period, as well as increasing TPS content. The decrease of EB values with increasing TPS content demonstrates the more rigid properties of starch compared to LLDPE. Furthermore, the decrease of EB values with increasing burial period demonstrates the biodegradation process of the samples by the microbes. Biodegradation promotes chain scission that reduces the polymer chain length and branches, leading to increase in crystallinity and decrease in EB values. The decrease in TS and EB values after burial treatment of plastics samples have also been reported in literature (Ali *et al.*, 2013; Dilfi *et al.*, 2017).

Figure 4. The tensile strength (TS) values as a function of burial period and TPS content for a) LLDPE/TPS1 (contains acetic acid) and b) LLDPE/TPS2 (does not contain acetic acid).

Figure 5. The elongation at break (EB) values as a function of burial period and TPS content for a) LLDPE/TPS1 (contains acetic acid) and b) LLDPE/TPS2 (does not contain acetic acid).

CONCLUSIONS

The presented work aimed at studying the physical, morphology, and biodegradation properties of bioplastics prepared from linear low density polyethylene (LLDPE) and thermoplastic starch (TPS) composites. It was found that the TPS enhances microbial growth and thus biodegradation of LLDPE/TPS composites, as seen in the microbial surface coverage data that increase with increasing TPS content in the composite after incubation test in agar medium. This finding was further validated by data from burial tests in a composting medium that showed: 1) increasing negative mass change, 2) decreasing tensile strength, 3) decreasing elongation at break, and 4) increasing heterogeneity (roughness) in surface morphology, with increasing both TPS content and burial period. Interestingly, despite the role of acetic acid as a hydrolyzing agent for polysaccharide chains, the samples that contain acetic acid showed higher stability against biodegradation process. According to some previously published reports, this phenomenon might be due to the antimicrobial activity of the acetic acid. This study thus suggests formulations for bioplastics prepared from LLDPE/TPS composites and provides physical as well as biodegradation properties of such composites. More general, this study contributes in supporting the global efforts in the substitution of non-biodegradable with biodegradable plastics, which should lead to reduction of the global problems associated with plastics wastes.

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REFERENCES

- Al-Rousan, W. M., Olaimat, A. N., Osaili, T. M., Al-Nabulsi, A. A., Ajo, R. Y., & Holley, R. A. (2018). Use of acetic and citric acids to inhibit *Escherichia coli* O157:H7, *Salmonella* Typhimurium and *Staphylococcus aureus* in tabbouleh salad. *Food Microbiology*, 73, 61-66. <https://doi.org/10.1016/j.fm.2018.01.001>
- Ali, R. R., Rahman, W. A. W. A., Kasmani, R. M., Ibrahim, N., Mustapha, S. N. H., & Hasbullah, H. (2013). Tapioca starch biocomposite for disposable packaging ware. *Chemical Engineering Transactions*, 32, 1711-1716.
- Amer, Z. J. A., & Saeed, A. Q. (2015). Soil burial degradation of polypropylene/starch blend. *International Journal of Technical Research and Applications*, 3(1), 91-96.
- Ashter, S. A. (2016). 2 - Overview of Biodegradable Polymers. In S. A. Ashter (Ed.), *Introduction to Bioplastics Engineering* (pp. 19-30). Oxford: William Andrew Publishing.
- ASTM. (2009). *ASTM G-21-70: Standard practice for determining resistance of synthetic polymeric materials to fungi*. America: American Society for Testing and Materials.
- ASTM. (2013). *ASTM D6813: Standard guide for performance evaluation of hydraulic fluids for piston pumps*. America: American Society for Testing and Materials.
- ASTM. (2018). *ASTM D882: Standar test methode for tensile properties of thin plastic sheeting*. America: American Society for Testing and Materials.
- Bjarnsholt, T., Alhede, M., Jensen, P. Ø., Nielsen, A. K., Johansen, H. K., Homøe, P., & Kirketerp-Møller, K. (2015). Antibiofilm properties of acetic acid. *Advances in Wound Care*, 4(7), 363-372. <https://doi.org/10.1089/wound.2014.0554>
- Botterell, Z. L. R., Beaumont, N., Dorrington, T., Steinke, M., Thompson, R. C., & Lindeque, P. K. (2019). Bioavailability and effects of microplastics on marine zooplankton: A review. *Environmental Pollution*, 245, 98-110. <https://doi.org/10.1016/j.envpol.2018.10.065>
- British Standard. (1996). *BS EN ISO 527-3: BS 2782-3: Method 326 E: 1996 - Plastics - Determination of tensile properties Part 3: Test conditions for films and sheets*.
- Chinaglia, S., Tosin, M., & Degli-Innocenti, F. (2018). Biodegradation rate of biodegradable plastics at molecular level. *Polymer Degradation and Stability*, 147, 237-244. <https://doi.org/10.1016/j.polymdegradstab.2017.12.011>
- Cho, Y., Shim, W. J., Jang, M., Han, G. M., & Hong, S. H. (2019). Abundance and characteristics of microplastics in market bivalves from South Korea. *Environmental Pollution*, 245, 1107-1116. <https://doi.org/10.1016/j.envpol.2018.11.091>
- Dilfi, K. F. A., Kurian, T. S., R. S., & Bhat, S. G. (2017). Effect of maleation on the mechanical properties and biodegradation of linear low density polyethylene-starch blends. *International Journal of Engineering and Information Systems*, 1(2), 101-110.
- Frias, J. P. G. L., & Nash, R. (2019). Microplastics: Finding a consensus on the definition. *Marine Pollution Bulletin*, 138, 145-147. <https://doi.org/10.1016/j.marpolbul.2018.11.022>
- Guzzetti, E., Sureda, A., Tejada, S., & Faggio, C. (2018). Microplastic in marine organism: Environmental and toxicological effects. *Environmental Toxicology and Pharmacology*, 64, 164-171. <https://doi.org/10.1016/j.etap.2018.10.009>
- Halstead, F. D., Rauf, M., Moimen, N. S., Bamford, A., Wearn, C. M., Fraise, A. P., & Webber, M. A. (2015). The antibacterial activity of acetic acid against biofilm-producing pathogens of relevance to burns patients. *Plos One*, 10(9), e0136190-e0136190. <https://doi.org/10.1371/journal.pone.0136190>
- Horodytska, O., Valdés, F. J., & Fullana, A. (2018). Centrifugal dewatering performance in plastic films recycling. *Waste Management*, 80, 211-217.
- Juniawati, Miskiyah, & Widaningrum. (2017). Aplikasi vinegar sebagai biopreservative untuk menghambat pertumbuhan *Salmonella typhimurium* pada daging ayam segar. *Buletin Peternakan*, 41(2), 187-196.
- Karthik, R., Robin, R. S., Purvaja, R., Ganguly, D., Anandavelu, I., Raghuraman, R., & Ramesh, R. (2018). Microplastics along the beaches of southeast coast of India. *Science of The Total Environment*, 645, 1388-1399. <https://doi.org/10.1016/j.scitotenv.2018.07.242>
- Li, J., Lusher, A. L., Rotchell, J. M., Deudero, S., Turra, A., Bråte, I. L. N., & Shi, H. (2019). Using mussel as a global bioindicator of coastal microplastic pollution. *Environmental Pollution*, 244, 522-533. <https://doi.org/10.1016/j.envpol.2018.10.032>
- Mwanza, B. G., Mbohwa, C., & Telukdarie, A. (2018). Strategies for the recovery and recycling of plastic

- solid waste (PSW): A focus on plastic manufacturing companies. *Procedia Manufacturing*, 21, 686-693. <https://doi.org/10.1016/j.promfg.2018.02.172>
- Nguyen, D. M., Do, T. V. V., Grillet, A.-C., Ha Thuc, H., & Ha Thuc, C. N. (2016). Biodegradability of polymer film based on low density polyethylene and cassava starch. *International Biodeterioration & Biodegradation*, 115, 257-265. <https://doi.org/10.1016/j.ibiod.2016.09.004>
- Oragwu, I. P. (2016). Investigations on the corn-starch modified low density polyethylene blends. *Matter: International Journal of Science and Technology*, 2(1), 1-11. <https://doi.org/10.20319/mijst.2016.21.0111>
- Prabha, P. H., & Ranganathan, T. V. (2017). Mdelling and optimization of cassava starch based film for food pacakaging applications. *Pakistan Journal of Biotechnology*, 14 (4), 571-586.
- Ragaert, K., Delva, L., & Van Geem, K. (2017). Mechanical and chemical recycling of solid plastic waste. *Waste Management*, 69, 24-58. <https://doi.org/10.1016/j.wasman.2017.07.044>
- Rainieri, S., & Barranco, A. (2018). Microplastics, a food safety issue? *Trends in Food Science & Technology*, 84, 55-57. <https://doi.org/10.1016/j.tifs.2018.12.009>
- Roy, S. B., Shit, S. C., Sengupta, R. A., & Shukla, P. R. (2015). Studies on biodegradability, morphology, physicomechanical and thermal properties of pp/potato starch bio-composite. *International Journal of Current Engineering and Technology*, 5(2), 1074-1084.
- Silveira, A. V. M., Cella, M., Tanabe, E. H., & Bertuol, D. A. (2018). Application of tribo-electrostatic separation in the recycling of plastic wastes. *Process Safety and Environmental Protection*, 114, 219-228. <https://doi.org/10.1016/j.psep.2017.12.019>
- Slootmaekers, B., Catarci Carteny, C., Belpaire, C., Saverwyns, S., Fremout, W., Blust, R., & Bervoets, L. (2019). Microplastic contamination in gudgeons (*Gobio gobio*) from Flemish rivers (Belgium). *Environmental Pollution*, 244, 675-684. <https://doi.org/10.1016/j.envpol.2018.09.136>
- Song, J. H., Murphy, R. J., Narayan, R., & Davies, G. B. H. (2009). Biodegradable and compostable alternatives to conventional plastics. *Philosophical Transactions of the Royal Society B, Biological Sciences*, 364(1526), 2127-2139. <https://doi.org/10.1098/rstb.2008.0289>
- Sun, J., Dai, X., Wang, Q., van Loosdrecht, M. C. M., & Ni, B. J. (2019). Microplastics in wastewater treatment plants: Detection, occurrence and removal. *Water Research*, 152, 21-37. <https://doi.org/10.1016/j.watres.2018.12.050>
- Tanetrungroj, Y., & Prachayawarakorn, J. (2018). Effect of dual modification on properties of biodegradable crosslinked-oxidized starch and oxidized-crosslinked starch films. *International Journal of Biological Macromolecules*, 120 Part A, 1240-1246. <https://doi.org/10.1016/j.ijbiomac.2018.08.137>
- Tawakaltu, A.-A., Egwim, E. C., Ochigbo, S. S., & Ossai, P. C. (2015). Effect of acetic acid and citric acid modification on biodegradability of cassava starch nanocomposite films. *Journal of Materials Science and Engineering B*, 5(9-10), 372-379.
- Teng, J., Wang, Q., Ran, W., Wu, D., Liu, Y., Sun, S., & Zhao, J. (2019). Microplastic in cultured oysters from different coastal areas of China. *Science of The Total Environment*, 653, 1282-1292. <https://doi.org/10.1016/j.scitotenv.2018.11.057>
- Thakur, S., Verma, A., Sharma, B., Chaudhary, J., Tamulevicius, S., & Thakur, V. K. (2018). Recent developments in recycling of polystyrene based plastics. *Current Opinion in Green and Sustainable Chemistry*, 13, 32-38. <https://doi.org/10.1016/j.cogsc.2018.03.011>
- Vargha, V., Rétháti, G., Heffner, T., Pogácsás, K., Korecz, L., László, Z., & Kelemen, O. (2016). Behavior of polyethylene films in soil. *Periodica Polytechnica Chemical Engineering*, 60(1), 60-68. <https://doi.org/10.3311/PPch.8281>
- Wang, S., & Copeland, L. (2015). Effect of acid hydrolysis on starch structure and functionality: A review. *Critical Reviews in Food Science and Nutrition*, 55(8), 1081-1097. <https://doi.org/10.1080/10408398.2012.684551>
- Wang, T., Zou, X., Li, B., Yao, Y., Zang, Z., Li, Y., & Wang, W. (2019). Preliminary study of the source apportionment and diversity of microplastics: Taking floating microplastics in the South China Sea as an example. *Environmental Pollution*, 245, 965-974. <https://doi.org/10.1016/j.envpol.2018.10.110>
- Windsor, F. M., Tilley, R. M., Tyler, C. R., & Ormerod, S. J. (2019). Microplastic ingestion by riverine macroinvertebrates. *Science of The Total Environment*, 646, 68-74. <https://doi.org/10.1016/j.scitotenv.2018.07.271>