

Properties of Bioplastic Sheets Made from Different Types of Starch Incorporated With Recycled Newspaper Pulp

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Abstract

The use of biodegradable material based on natural polysaccharides, particularly starch helps to reduce the usage of non-degradable materials. In this study, three types of starch were used to produce the bioplastic sheets (cassava, corn and potato). The sheets were produced with the mixture of bioplastic (B) incorporated with recycled newspaper pulp fibre (N) at four different ratios (newspaper pulp fibres:bioplastic) N50%:B50%, N30%:B70%, N10%:B90% and N0%:B100%. Water absorption and tensile properties were investigated for these bioplastic sheets which were done in room temperature. Cassava-based bioplastic sheet had the worst water repellent while corn starch-based bioplastic sheets had the lowest water absorption percentage. Based on the ratios, bioplastic sheet N30%:B70% shows the lowest percentage of water absorption. Result also showed that as the amount of bioplastic ratio increase, the tensile strength decrease. The optimum mixture of fibres/bioplastic was N50%:B50% which obtained highest percentage of tensile strength. Elongation at break was increased as the bioplastic increased.

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Introduction

Plastics made from petroleum-based have many drawbacks. It needs a large amount of energy in the production process, besides it took years to degrade and at the same time caused serious hazards to the environment (Azahari *et al.*, 2011; Averous & Pollet, 2012; Jones *et al.*, 2013). In order to shift to sustainable pathways, the development of biodegradable products has increased years ago and it continues to be the area that attracts scientists to involve with new green materials and improvement ideas (Jones *et al.*, 2013; Satyanarayana *et al.*, 2009; Md Enamul *et al.*, 2013). Renewable natural polymer resources such as starches were one of the most attractive materials because of its' inherent biodegradability, ready availability and low cost (Azahari *et al.*, 2011; Patel *et al.*, 2011; Tang *et al.*, 2007). Biodegradation of bioplastic can be characterized with the loss of weight, change in tensile strength, change in dimensions, change in chemical and physical properties, carbon dioxide production, bacterial activity in soil and change in molecular weight distribution (Singh & Sharma, 2008). Nowadays, starch is widely used in the fields of food technology, engineering, pharmaceutical, packaging and agriculture (Guimaraes *et al.*, 2010; Liew & Khor, 2015; Stasiak *et al.*, 2013; Funke *et al.*, 1998). Different approaches have been made to increase the usage of starch as natural biopolymer for the production of biodegradable products which led to various other applications (Funke *et al.*,

1998; Guimaraes *et al.*, 2010; Yun *et al.*, 2008). Starch contains primarily of branched and linear chains molecules mainly composed of amylose and amylopectin (Aranda Gracia *et al.*, 2015; Mali *et al.*, 2006). Amylose considered being an essentially linear polymer composed almost entirely of α -1, 4 bonds while amylopectin is a polymer with highly branch points linked by α -1, 6 bonds (Sasaki *et al.*, 2000; Thomas & Atwell, 1999). Both of these chains show very different physical and chemical properties; great number of amylose in starches influenced the properties such as the strength of film, the gelatinization, glass transition temperature, granules size and presence of plasticizers (Alves *et al.*, 2007; Aranda Gracia *et al.*, 2015; Dufresne & Vignon, 1998; Mali *et al.*, 2006; Talja *et al.*, 2007).

It is well known that the main drawbacks of using starch are high solubility in water and low mechanical strength. Previous attempts have been made to improve the properties of starch; blending the starch with degradable synthetic polymers such as polyvinyl alcohol and polylactic acid, addition of lignocelulosic fibres as reinforcement and utilize the agro waste (Azahari *et al.*, 2011; Dufresne & Vignon, 1998; Satyanarayana *et al.*, 2009). This study focused on the effect of bioplastic starch towards the properties of the sheets, therefore the ratio of newspaper pulp and bioplastic begun at equal amount of newspaper pulp fibres and bioplastic starch (N50%:B50%) and then carried out by increasing the bioplastic content (B). The bioplastic sheets incorporated with fibres was believed could enhance the performance of the sheets. In order to investigate the properties of bioplastic sheets which were made from different starch based incorporated with recycled newspaper pulp, the sheets were evaluated for its weight changes in water absorption testing and the strength of each sheet after underwent tensile strength testing.

Materials and Methods

Preparation of material

Materials used to produce the bioplastic sheets were newspaper pulp fibres, starches (cassava, corn and potato), glycerol (99.5%), water and vinegar. Newspaper pulp underwent fibrillation (20 minutes) and screened. Sheets made from 100% (w/w) bioplastic acted as Control.

Fabrication of bioplastic sheets

Bioplastic was prepared by mixing starch, glycerol, water and vinegar together based on the ratio of newspaper pulp and bioplastic (in weight). The calculation was made based on Liew and Khor (2015) with slight modification. These materials were mixed well before the addition of pulp fibres and heated on hot plate at 70⁰C for 40 minutes with frequent stirring. The mixture was then spread evenly in a mold (15cm x 15 cm x 2 mm) into bioplastic sheets before underwent semi drying in the oven about 30- 45 minutes. The aim of semi drying was to assist the removal of the sheets. For final drying, the sheets were dried in the oven for 24 hours at 70⁰C.

Testing

Water absorption was measured based on ASTM D570-98. Dried sheets were weighed for the initial weight and then placed in a bath of distilled water at room temperature. The sheets were removed

after 24 hours where water on the surface of the sheets was wiped off and weighed the sheets again. Results were obtained from the average of 5 replicates based on the equation:

$$\text{Water absorption percentage (\%)} = (W_i - W_f) / W_f \times 100\% \quad (1)$$

where W_i is initial weight (g) before immersion in water and W_f is final weight (g) after immersion in water. The tensile strength was measured by using universal testing machine according to the ASTM D882-02 with slight modification. The sheets were cut into 100mm x 25mm, with the gauge 50mm, and conducted with the speed of 2 mm/min.

Results and discussion

Water sensitivity is one of the important criterions in many applications and it was verified that the mechanical properties also dependent on the moisture content (Funke *et al.*, 1998; Dufresne & Vignon, 1998). Figure 1 illustrated the percentage of water absorption against the ratio of newspaper pulp fibres/bioplastic. From the graph, it can be seen that water absorption increased by increasing of bioplastic percentage. Results showed that cassava-based bioplastic sheets ($R^2 = 0.5633$) had the highest water absorption percentage followed by potato-based bioplastic ($R^2 = 0.5315$) sheet and corn-based ($R^2 = 0.55$) bioplastic sheet. For each starch-based sheets, bioplastic sheets N0%:B100% (Control) had the highest water absorption percentage due to pure bioplastic content followed by sheets N10%:B90%; cassava-based bioplastic had the highest water absorption among other bioplastic sheets with 81.09%. Control had higher water absorption percentage compared to other sheets due to high content of starch. Starch itself is hydrophilic and vulnerable to moisture. Besides, the starch granule busted during the gelatinization which eased water diffusion (Azahari *et al.*, 2011).

The presence of fibres in the sheets decreased the water absorption. However, it was not enough to enhance the repellency of water absorption. High amount of bioplastic was the main factor of high percentage of water absorption. At this point, the sheets become more sensitive towards water because of the hydrophilic nature of starch. The presence of hydroxyl groups in starch interacted with the water molecules, besides the rupture of the starch granules during gelatinization of starch that eased the water absorption (Azahari *et al.*, 2011). It also was proved by the previous studies which found that water absorption increased at high amount of starch (Azahari *et al.*, 2011; Aranda Garcia *et al.*, 2015; Dufresne & Vignon, 1998). Bioplastic sheets N30%:B70% for each starch-based sheets of all starches showed the lowest percentage of water absorption. Corn-based bioplastic sheets had the lowest percentage 22.69% followed by potato-based bioplastic sheets 32.35% and cassava-based bioplastic sheets 39.48%.

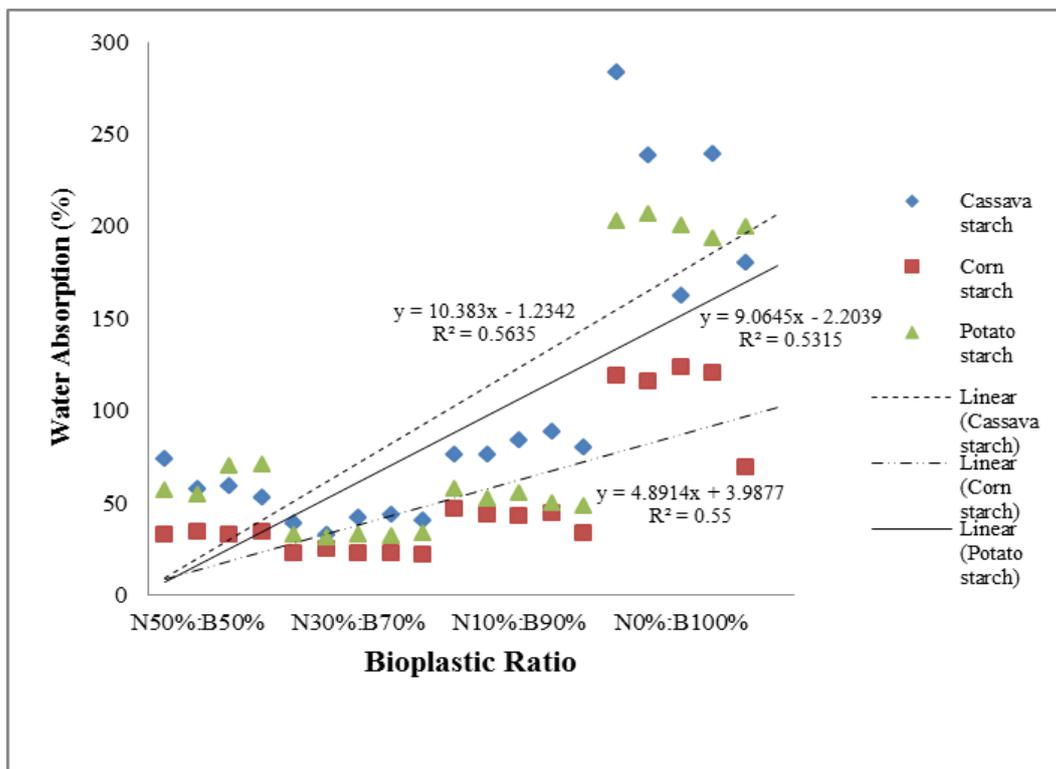


Figure 1. Trend graph for water absorption of bioplastic sheets with different ratios of bioplastic and newspaper pulp fibers.

Funke *et al.*, (1998) mentioned that improvements of water absorption can be enhanced by adding small amount of fibres. It can be seen in Figure 1 that the optimum content of fibres in bioplastic sheets was 30% which showed the lowest water absorption. Bioplastic sheets with 50% of fibers content of all starches were slightly higher than bioplastic sheets incorporated with 30% of fibers. It may be due to high amount of fibres caused the bioplastic sheets to have less widespread availability and poor interfacial bonding between fibres, resulted in the presence of micro voids within the fibres and bioplastic which provides more spaces for water to penetrate and also high amount of porous tubular structures in fibres that accelerates the penetration of water (Ashori & Sheshmani, 2010; Azahari *et al.*, 2011; Dufresne & Vignon, 1998; Shakeri & Ghasemian, 2009).

Bioplastic sheets was the foundation of bioplastic pot making that may be subjected to various kinds of stress during use, which means the determination of mechanical properties includes not only the scientific but also technology and practical aspects (Azahari *et al.*, 2011; Tang *et al.*, 2008). Figure 2 shows the effect of different ratios of bioplastic sheet towards the tensile strength. The trend graph showed that potato-based bioplastic sheets ($R^2 = 0.9413$) has the highest tensile strength followed by cassava-based bioplastic ($R^2 = 0.904$) sheet and corn-based ($R^2 = 0.882$) bioplastic sheet. The highest tensile strength obtained from bioplastic sheets N50%:B50% for each starch-based. The potato-based bioplastic sheet obtained the highest tensile strength percentage (4.87 N/mm^2) followed by cassava-based bioplastic sheet (4.5 N/mm^2) and corn-based bioplastic sheet (3.59 N/mm^2). Control sheets for all starch based showed the lowest tensile strength compared to others sheets. Without the presence of fibres as reinforcement in the Control sheets, it became weak towards tensile force. Curvelo *et al.*,

(2001) stated that the presence of fibres as the reinforcements would enhance the mechanical properties. When fibres incorporated at ratio of 50%, the bioplastic sheets gave the highest result compare to other bioplastic. Moreover, it shows that the tensile strength increases as the ratio of the two materials (newspaper pulp fibers: bioplastic) simultaneously increase. However, as the amount of starch increased, the tensile strength decreased due to less fibres-matrix interactions and filler-filler interaction become more prominent (Azahari *et al.*, 2011; Liew & Khor, 2015).

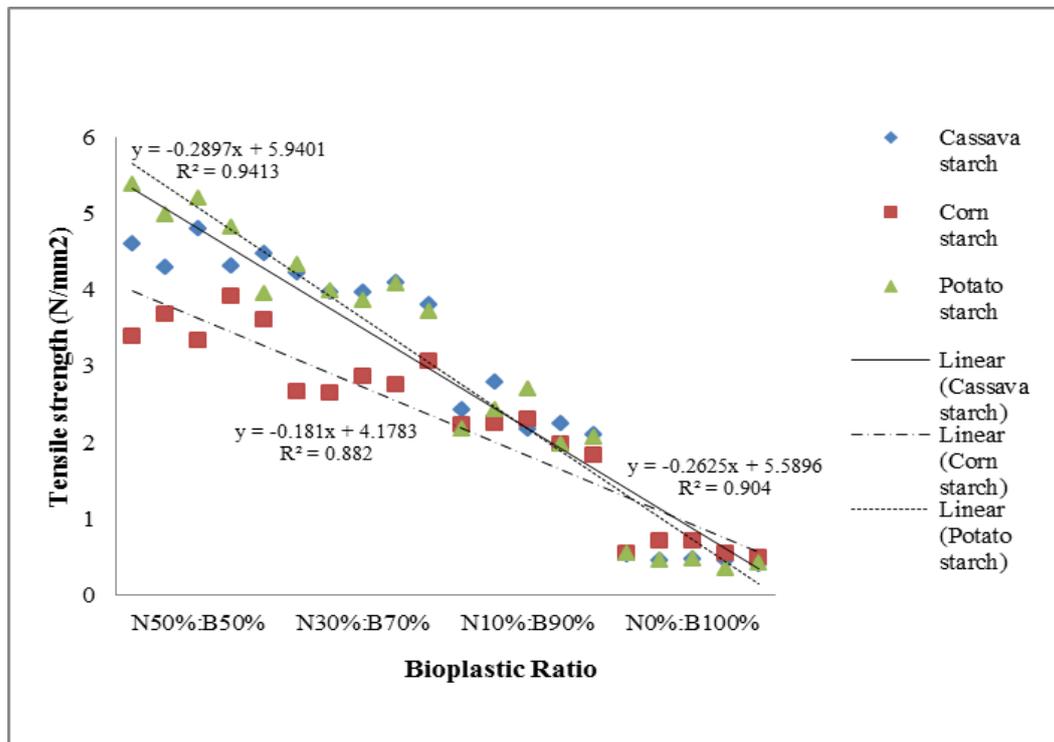


Figure 2. Trend graph for tensile strength of bioplastic sheets with different ratios of bioplastic and newspaper pulp fibres.

The elongation at break was shown in Figure 3. The trend graph showed different patterns from the tensile graph. The percentage of elongation at break continuously increased as the amount of bioplastic increased. This is because of the flexibility of bioplastic which contained high amount of starch and glycerol (Azahari *et al.*, 2011). For each starch-based, Control showed the highest percentage of elongation. The trend graph showed that potato-based bioplastic sheets ($R^2 = 0.72$) has the highest percentage of elongation followed by corn-based bioplastic ($R^2 = 0.5928$) sheet and cassava-based ($R^2 = 0.2518$) bioplastic sheet. Bioplastic sheets N50%:B50% for each starch-based has the lowest percentage of elongation at break. High fibres content caused the sheets to become less flexible but has better tensile strength due to the presence of fibres-matrix bonding (Azahari *et al.*, 2011). Bioplastic sheets N10%:B90% has the highest percentage of elongation at break after Control. High presence of bioplastic in the sheets caused the sheets more flexible and can hold before it broke. There were many types of biodegradable plastics that available in the market which applied in various purposes such as food wrapping, engineering and agriculture and even for medical treatment. Gaspar

et al., (2005) had used biodegradable polymers to produced biodegradable starch-based plastics. In comparison, in terms of water absorption and tensile strength, sheets produced in this study had higher water absorption percentage and lower tensile strength due to different types of ingredients that influenced the properties of the sheets. Besides, the process of making the biodegradable plastics also contributes to different results. However, the bioplastic sheets that have been produced in this study can be applied in the fieldwork as done by Liew & Khor (2015).

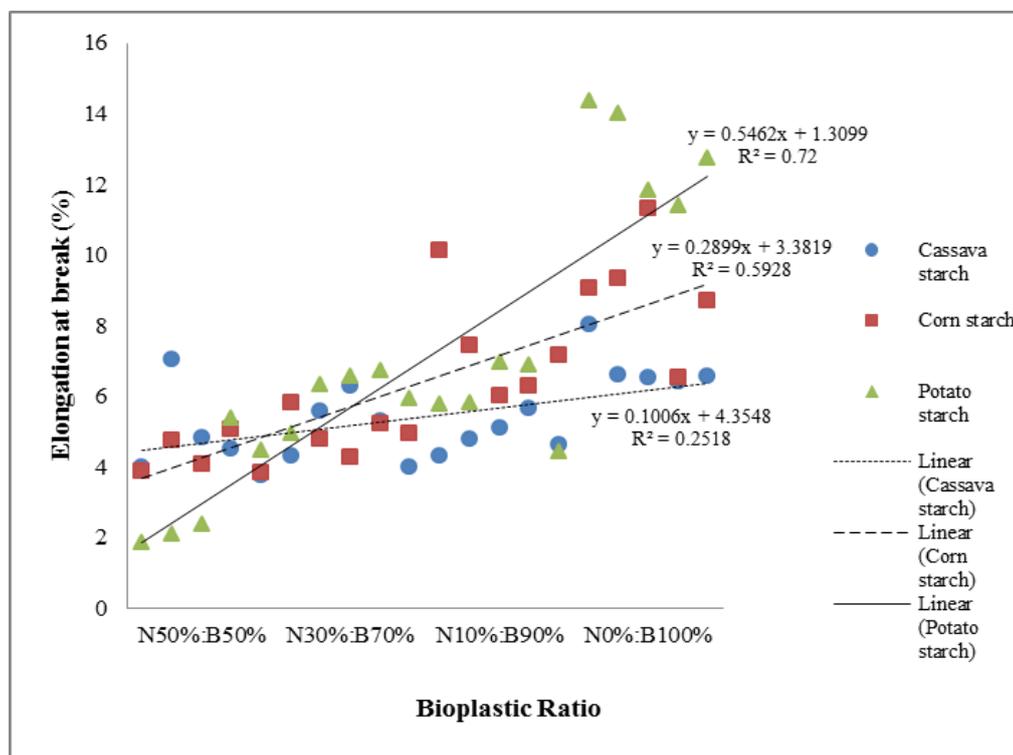


Figure 3. Trend graph for elongation at break of bioplastic sheets with different ratios of bioplastic and newspaper pulp fibres.

Conclusion

This study showed that different types of starch and ratios of fibres/bioplastic sheet will affected the water absorption and tensile strength properties. The results obtained showed that the cassava-based bioplastic sheets had the highest percentage in water absorption and corn-based bioplastic sheets had the lowest water absorption. Highest value of water absorption resulted in bioplastic sheets N10%:B90% while lowest water absorption was reached at the ratio of N30%:B70% for all starch-based bioplastic sheets. Tensile test showed that for each starch-based, the highest percentage of tensile was obtained in bioplastic sheets N50%:B50%. Elongation at break was increased as the amount of bioplastic increased. Bioplastic sheets N10%:B90% has the highest elongation at break after Control. Further studies are therefore necessary to improve the properties of starch-based bioplastic.

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References

- [1] Alves, V. D., Mali, S., Beleia, A. & Grossmann, M. V. E. (2007). Effect of glycerol and amylose enrichment on cassava starch film properties. *Journal of Food Engineering*, **78**, 941-946.
- [2] Aranda Garcia, F. J., Gonzalez Nunez, R., Jasso Gastinel, C. F., & Mendizabal, E. (2015). Water absorption and thermomechanical characterization of extruded starch/poly (lactic acid)/agave bagasse fiber bioplastic composites. *International Journal of Polymer Science*, **2015**, Article ID 343294. DOI: 10.1155/2015/343294.
- [3] ASTM D8570-98. Standard Test Methods for Water Absorption.
- [4] ASTM D882-02. Standard Test Methods for Tensile Properties of Thin Plastic Sheeting.
- [5] Ashori, A. & Sheshmani, S. (2010). Hybrid composites made from recycled materials: moisture absorption and thickness swelling behavior. *Bioresource Technology*, **101**, 4717-4720.
- [6] Averous, L. & Pollet, E. (2012). Biodegradable Polymers. In: Averous, L. & Pollet, E. (eds.). *Environmental Silicate Nano-Biocomposites, Green Energy and Technology*. Springer-Verlag, London.
- [7] Azahari, N. A., Othman, N. & Ismail, H. (2011). Biodegradation studies of polyvinyl alcohol/corn starch blend films in solid and solution media. *Journal of Physical Science*, **22**(2), 15-31.
- [8] Curvelo, A. A. S., Carvalho, A. J. F. & Agnelli, J. A. M. (2001). Thermoplastic starch-cellulosic fibres composites: Preliminary results. *Carbohydrates Polymers*, **45**, 183-188.
- [9] Dufresne, A. & Vignon, M. R. (1998). Improvement of starch film performances using cellulose microfibrils. *Macromolecules*, **31**, 2693-2696.
- [10] Funke, U., Bergthaller, W. & Lindhauer, M. G. (1998). Processing and characterization of biodegradable products based on starch. *Polymer Degradation and Stability*, **59**, 293-296.
- [11] Gaspar, M., Benko, Zs., Dogossy, G., Reczey, K. & Czigany, T. (2005). Reducing water absorption in compostable starch-based plastics. *Polymer Degradation and Stability*, **90**, 563-569.
- [12] Guimaraes, J. L., Wypych, F., Saul C. K., Ramos, L. P. & Satyanarayana, K. G. (2010). Studies of the processing and characterization of corn starch and its composites with banana and sugarcane fibers from Brazil. *Carbohydrate Polymers*, **80**, 130-138.
- [13] Jones, A., Zeller, M. A. & Sharma, S. (2013). Thermal, mechanical, and moisture absorption properties of egg white protein bioplastics with natural rubber and glycerol. *Progress in Biomaterials*, **2**, 12.
- [14] Liew, K. C. & Khor, L. K. (2015). Effect of different ratios of bioplastic to newspaper pulp fibres on the weight loss of bioplastic pot. *Journal of King Saud University-Engineering Sciences*, **27**, 137-141.
- [15] Mali, S., Grossmann, M. V. E., Garcia, M. A., Martino, M. N. & Zaritzky, N. E. (2006). Effect of controlled storage on thermal, mechanical and barrier properties of plasticized films from different starch sources. *Journal of Food Engineering*, **75**, 453-460.
- [16] Md Enamul, H., Tan, J. Y. & Leng, C. Y. (2013). Sago starch-mixed low density polyethylene biodegradable polymer: Synthesis and characterization. *Journal of Materials*, **2013**, Article ID 365380. DOI: 10.1155/2013/365380.
- [17] Patel, P. N., Garmer, K. G., Patel, M. N., Patel, P. R. & Sen, D. J. (2011). Biodegradable polymers: An ecofriendly approach in newer millenium. *Asian Journal of Biomedical and Pharmaceutical Sciences*, **1**(3), 23-39.
- [18] Sasaki, T., Yasui, T., and Matsuki, J. (2000). Effect of amylose content on gelatinization, retrogradation, and pasting properties of starches from waxy and nonwaxy and their F1 seeds. *Cereal Chemistry*, **77**(1), 58-63.
- [19] Satyanarayana, K. G., Arizaga, G. G. A. & Wypych, F. (2009). Biodegradable composites based on lignocellulosic fibers - An overview. *Progress in Polymer Science*, **34**, 982-1021.
- [20] Shakeri, A. & Ghasemian, A. (2009). Water absorption and thickness swelling behavior of polypropylene reinforced with hybrid recycled newspaper and glass fiber. *Applied Composite Material*, **17**, 183-193.
- [21] Singh, B. & Sharma, N. (2008). Mechanistic implications of plastic degradation. *Polymer Degradation and Stability*, **93**, 561-568.
- [22] Stasiak, M., Molenda, M., Oplakinski, I. & Blaszcak, W. (2013). Mechanical properties of native maize, wheat, and potato starches. *Czech Journal Food Science*, **31**(4), 347-354.
- [23] Tang, S., Zou, P., Xiong, H. & Tang, H. (2007). Effect of nano-SiO₂ on the performance of starch/polyvinyl alcohol blend films. *Carbohydrate Polymers*, **72**, 521-526.

- [24] Talja, R. A., Harry. H., Roos, Y. H. & Jouppila, K. (2007). Effect of various polyols and polyol contents on physical and mechanical properties of potato starch-based films. *Carbohydrate Polymers*, **67**, 288-295.
- [25] Thomas, D. J. & Attwell, W. A. (1999). *Starch Structure*. American Association of Cereal Chemist, Inc., Minnesota.
- [26] Yun, Y. H., Wee, Y. J., Byun, H. S. & Yoon, S. D. (2008). Biodegradability of chemically modified starch (RS4)/PVA blend film: Part 2. *Journal Polymer Environment*, **16**, 12-18.