A comparison study on mechanical properties of virgin and recycled polylactic acid (PLA)

S Budin*, N C Maideen, M H Koay, D Ibrahim and H Yusoff
Faculty of Mechanical Engineering, Universiti Teknologi MARA, Cawangan Pulau Pinang, 13500 Permatang Pauh, Penang, Malaysia

*salinabudin@uitm.edu.my

Abstract. Huge usage of Polylactic Acid (PLA) has contributed to the high wastes which mainly come from the worn parts. Recycling of the wastes is seen to be a permissible solution to reduce the quantity of wastes in the landfills. This work aims to compare the mechanical properties of virgin and recycled PLA. The recycled material is collected from the waste produces from the injection molding process. The mechanical testing samples for both virgin and recycled materials are prepared using an injection molding process. The results show that the mechanical properties of recycled PLA lower than the virgin PLA. The tensile strength of recycled PLA decreases by 11% as compared to virgin PLA. On the other hand, TRS is also observed drop by 5% for recycled PLA compared to virgin PLA. A similar trend is found in the impact strength and hardness of properties where its impact strength drops by 50% of recycled PLA. For hardness property, it is noted that the hardness decreases by 4% of the recycled PLA. The decreasing value of the mechanical properties is due to thermal degradation of the PLA molecular chain during the injection molding process which has consequently affected the mechanical properties of the recycled PLA.

1. Introduction
The polymer waste has become a major concern in most of the developed countries. The demand for polymer parts and components has increased immensely during the past 50 years [1]. It has been reported that polymer waste contributes approximately 58% from overall solid waste at the UK, which mainly derives from commercial and household packaging [2]. In India, approximately 133760 tons of plastic waste is discharged every day [3]. In Rio de Janeiro City, in the year 2003, it has reported that 554,800 tons of plastic waste are deposited every year [4]. In other reviews by Madi [5], it has reported that 2000 tons of plastic waste is generated in Qatar every year. A similar trend was also observed in other countries which demonstrated that polymer waste is one of the major global issues.

For such reason, numbers of research and developments have been focused on the production on eco-sustainable and “green” polymer, by replacing non-biodegradable polymer matrices with a biodegradable polymer. Several biodegradable, natural-derived polymers exist, such as polysaccharides, proteins, polyesters, lignin, lipids, natural rubber, some polyamides, polyvinyl alcohols, polyvinyl acetates, and polycaprolactone. These polymers are degraded through enzymatic reactions in suitable environments. Among those biodegradable polymers, polyester group, especially polylactic acid (PLA) is the most attractive biodegradable polymer due to its better processing ability and properties as compared to other biodegradable plastic [6]. PLA has good mechanical properties which are higher than polystyrene (PS), PP, and polyethylene (PE).
However, PLA is very brittle with less than 10\% elongation at break and low toughness which limits its use in applications that need plastic deformation under a high stressed procedure. The shortcoming of PLA can be easily resolved by reinforced the PLA matrix with natural or synthetic fibers or associate it with selected additives which allow it to obtain specific end use characteristics.

Extensive researches on PLA especially, PLA blend has resulted in high demand in PLA parts for various applications such as medical and packaging. For example, the PLA/ PHA (Polyhydroxyalkanoates) blend is a beneficial material for food packaging applications. On the other hand, PLA/PMMA blend has been suggested as an initial substrate for cell growth in the medical applications [7]. With such versatile applications, huge usage of PLA has contributed to the high waste which mainly comes from the worn parts. A slow degradation process as compared to the amount of the worn parts has again created another issue in solid waste management. Previous works have reported that the degradation of PLA in the soil is slow and take a long time for degradation to start due to the slow rate of hydrolysis [8].

The recycling of PLA wastes is seen to be a permissible solution to reduce the quantity of PLA wastes in the landfills and those indiscriminately discarded in the environment. It leads to a reduction of problems such as the spread of diseases, contamination of soil, air and water bodies [4]. The recycling polymer could reduce 80\% of greenhouse gas emission besides acting as a renewable source of material which supports the social-ecological sustainability [9]. However, one of the most important conditioning factors when substituting a virgin PLA for a recycled PLA is whether the original characteristics are preserved in the recycled PLA [10]. When comparing with virgin material, recycled material tends to have less desirable properties including mechanical and rheological properties which caused by permanent chemical and physical changes of the molecular structures or continuous change in the thermodynamic equilibrium of the molecular structure [9]. The reduction in the molecular weight after successive recycling cycles degrades the mechanical properties of recycled material. The mechanical recycling at elevated temperatures is reported to induce degradation of the macromolecular structure, resulting in chain scission. Many works have proven the properties of recycled plastics are degraded. For example, studies conducted by Rahimi et al. [11] on recycled acrylonitrile-butadiene-styrene (ABS) have proved that a reduction in the shrinkage and impact properties on recycled ABS as compared to the virgin material. The authors also observed that no significant effect on tensile stress and flexural strength.

A research work by Wang et al. [12] investigated the difference of properties between recycled ABS and virgin ABS. The results showed that the mechanical properties of the recycled ABS dropped significantly when the impact strength decreased over 80\%. However, a contradicted finding was reported by Gracia et al. [13] that recycling of ABS for one and twice times have better results of strength in the tensile test. In work conducted by Hirayama and Saron [14] revealed that the tensile strength, impact strength and velocity of recycled ABS decreased when compared to the virgin ABS. However, recycled high impact polystyrene (HIPS) showed negligible differences in tensile strength, Young Modulus, melt flow index and impact resistance as compared to the virgin HIPS.

Janajreh et al. [15] reported that the reprocessing of PVC is tougher but slightly higher on the stiffness level as compared to the virgin sample. In other work, Raj et al. [16] have studied on mechanical properties of recycled PP. The result indicated that the tensile strength is decreased for the 1st recycling PP as compared to the virgin PP. They also reported that the TRS and impact strength increased for the 1st recycling PP as compared to the virgin PP. In other works, Ronkay [17] examined the effect of recycling on the rheological, mechanical and optical properties of Polycarbonate (PC). The author reported that the average molecular mass of PC decreases by approximately 8\% during the first injection molding and causes about 25\% of the increase in the melt flow rate. The author has also concluded that a recycled material content above 10wt\% of the total weight causes a detectable difference in the violet absorption and 20wt\% of recycled material in the composition may significantly deteriorate the mechanical and optical properties of PC.

Although many works related to recycling plastic material have been reported, limited works have been done on recycled PLA. Thus, exploration and investigation of recycled PLA could promote the usage of recycled PLA and at the same time help to reduce the PLA waste and its
impact on the environment. In this work, the mechanical properties of virgin and recycled PLA are studied and compared. The tensile strength, transverse rupture strength (TRS), impact strength and hardness properties were investigated.

2. Methodology

2.1 Material

The virgin material was purchased from the local supplier in an injection molding grade. The pallet is in a cylindrical shape, with an average length and diameter of 3 mm. The virgin pellets are shown in Figure 1. Prior to sample processing, the pallet is required to undergo drying in an oven at a temperature of 50°C for 24 hours to avoid flaws or void occurs in the samples.

![Figure 1. Virgin pallet](image)

2.2 Sample preparation

Samples for tensile test, transverse rupture test (TRS), impact test and hardness test for 100% virgin composition were first prepared using reciprocation injection of the molding machine. The barrel zone of the injection molding machine consists of nozzle, rear, middle and front. The temperatures set up at the nozzle, rear, middle and front were 185°C, 195°C, 165°C, and 60°C respectively. The packing and holding pressures were at 80 MPa and 70 MPa respectively. The injection process was continuously running for 100 cycles to produce 100 of the samples. Five samples were randomly selected for each test; tensile test, TRS, impact test and hardness test.

The wastes; runners and gates, from the processing of virgin material samples are collected and crushed using a grinding machine. The recycled ground material is an angular shape with the size of less than 4 mm. The recycled ground material is shown in Figure 2.

The recycled ground material was then injected thru injection molding machine under the same processing conditions to produce samples with a composition of 100% recycled PLA. Similarly, the injection process was continuously running for 100 cycles to produce 100 of the samples. Five samples were randomly selected for each test; tensile test, TRS, impact test and hardness test.
2.3 Sample testing

The tensile strength was obtained using Universal Testing Machine based on ASTM D638 at room temperature. The gauge distance is 103.2 mm with a moving speed of 1 mm min\(^{-1}\). A sampling rate is 20 pts s\(^{-1}\) and full-scale load range of 50 kN.

The TRS was obtained using Universal Testing Machine based on ASTM D790 at room temperature. The span is set at 60 mm with a crosshead moving speed of 1 mm min\(^{-1}\). A sampling rate is 20 pts s\(^{-1}\) and full-scale load range of 50 kN.

The impact data were gained using Izod/Charpy Impact Tester according to the ASTM D256 at room temperature. The hammer impact is set at 5.42 J. The mass of the pendulum is 0.906 kg.

Finally, the hardness measurement was done at room temperature using Vickers Hardness tester at a load of 500g using diamond pyramid indentation. The Hv hardness value (MPa) was derived from the residual projected area of indentation according to Equation 1.

\[ Hv = 1.854 \left( \frac{F}{d^2} \right) \]  \hspace{1cm} (1)

where \(d\) is the length of the impression diagonal (mm) and \(F\) the applied contact load (N). As to ensure the consistency of the experimental results, each test is repeated for five times. The final results are calculated based on the average of five data obtained in the process.

3. Result and Discussion

3.1 Tensile test

Figure 3 shows the tensile strength for virgin and recycled PLA. The average tensile strength for a virgin sample is 42.87 N/mm\(^2\) and the average tensile strength for a recycled sample is 32.01 N/mm\(^2\). From the result, it is clearly indicated that the tensile strength of recycled PLA is decreased by approximately 11%. The decreasing in tensile strength is due to thermal degradation of the PLA molecular chain during the injection molding process. The breaking of molecular chain occurs when the heat is exposed during the processing stage.
3.2 Transverse rupture strength (TRS)

The average TRS results for the virgin sample and the recycled sample is shown in Figure 4. Similarly, TRS has also dropped by 5% for the recycled PLA when compared to virgin PLA. The TRS values for virgin and recycled PLA are 61 N/mm² and 58 N/mm² respectively. This indicated that recycled PLA will deflect faster as compared to virgin PLA under the same applied load. Due to shear forces and thermal impact encountered during the injection molding, it has caused a breaking of the molecular chain and reduction in molecular weight. Thus, increases the brittleness of the material [16].

3.3 Impact test

Figure 5 shows the impact strength of virgin and recycled PLA. The average impact strength for virgin PLA and recycled PLA is 0.178 J/m and 0.082 J/m respectively. It is noted that there is a tremendous decrease in the impact test which is approximately 50%. The finding is consistent with Rahimi et al. [11] findings, which observed drastically a decrease in the impact strength for recycled ABS. The drastic decrease in the impact strength is also observed by Hirayama and Saron [8] in their investigation of recycled ABS. They reported that the impact strength for virgin ABS is 17.8 kJ/m² which drops to 4.4 kJ/m² after the first recycling. This effect may be caused by the deterioration of the molecular chains which has consequently resulted in the stiffness of the material [17].
3.4 Hardness test

Similar trends are recorded on hardness properties where the hardness decreases by 4% for the recycled PLA as illustrated in Figure 6. The results show the hardness for virgin PLA is 21.39 HV whereas the hardness for recycled PLA is 20.57 HV. The finding is consistent with tensile strength which also decreases by 5% for the recycled PLA as compared to virgin PLA. The decreasing in tensile strength reduces the ability of the polymer to withstand an applied load without failure and consequently reduces the ability to resist the deformation.

4. Conclusions

The diversity of recycled PLA on mechanical properties has been studied. The main conclusions reached based on the current study are summarized as follows:
1. The mechanical properties of recycled PLA decrease as compared to the virgin PLA.
2. The average tensile strength of the recycled PLA has dropped by 11% with respect to the virgin PLA.
3. A similar trend is observed on the TRS of recycled PLA decreases by 5% as compared to virgin PLA.
4. The hardness properties for the recycled PLA decreases by 4% when comparing to virgin PLA.
5. The most significant decrease in percentage is observed on the impact of energy where it’s decreased by 50% for recycled PLA as compared to virgin PLA.
6. The decreasing value of the mechanical properties is mainly due to the shear forces and thermal degradation on the PLA molecular chain occurs during the injection molding process. The reduction in the mechanical properties by 100% of recycled PLA limits the application of recycled PLA, especially when it is used in producing similar products.
Although recycled PLA has exhibited lesser desirable properties, the weakness can be overcome by blending recycled PLA with suitable additive or filler.

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References
[1] Singh N Hui D Singh R Ahuja I P S Feo L and Fraternali F 2017 Recycling of plastic solid waste: A state of art review and future applications Compos. Part B Eng. 115, p. 409-422.
[2] Turku I Keskisari A Karki T Puurtinen A and Marttila P 2017 Characterization of wood plastic composites manufactured from recycled plastic blends Compos. Struct. 161, p. 469-476.
[3] Sheriff K M M Subramanian N Rahman S and Jayaram J 2017 Integrated optimization model and methodology for plastics recycling: Indian empirical evidence J. Clean. Prod. 153, p. 707-717.
[4] Pacheco E B A V Ronchetti L M and Masanet E 2012 An overview of plastic recycling in Rio de Janeiro Resour. Recov. Conserv. 60, p. 140-146.
[5] Madi N K 2013 Thermal and mechanical properties of injection molded recycled high-density polyethylene blends with virgin isotactic polypropylene Mater. Design 46, p. 435-441.
[6] Murariu M and Dubois P 2016 PLA composites: From production to properties Adv. Drug Deliv. Rev. 107, p. 17-46.
[7] Hamad K Kaseem M Ayoyoob M Joo J and Deri F 2018 Polyacrylic acid blends: the future of green, light and tough Prog. Polym. Sci. 85, p. 83-127.
[8] Vasile C Pamfil D Rapa M Darie-Nita R N Mitelut A C Popa E E Popescu P A Draghi M C and Popa M E 2018 Study of the soil burial degradation of some PLA/CS biocomposites Compos. Part B Eng. 142, p. 251-262.
[9] Gu F Hall P Miles N J Ding Q and Wu T 2014 Improvement of mechanical properties of recycled plastic blends via optimizing processing parameters using the Taguchi method and principal component analysis Mater. Design 62, p. 189-198.
[10] Mantia F P L and Morreale M 2011 Green composites: A brief review Compos. Part A Appl. Sci. Manuf. 42, p. 579-588.
[11] Rahimi M Esfahanian M and Moradi M 2014 Effect of reprocessing on shrinkage and mechanical properties of ABS and investigating the proper blend of virgin and recycled ABS in injection molding J. Mater. Process. Tech. 214, 11 p. 2359- 2365.
[12] Wang J Li Y Song J He M Song J and Xia K 2015 Recycling of acrylonitrile-butadiene-styrene (ABS) copolymers from waste electrical and electronic equipment (WEEE), through using an epoxy-based chain extender Polym. Degrad. Stab. 112, p. 167-174.
[13] Gracia M G T Schlatter M Cabrera F M Manzanares J T and Hanafi I 2016 Recycling of Acrylonitrile-Butadiene-Styrene using injection molding machine Proc. Technol. 22, p. 399-406.
[14] Hirayama D and Saron C 2018 Morphologic and mechanical properties of blends from recycled acrylonitrile-butadiene-styrene and high-impact polystyrene Polymer 135, p. 271-278.
[15] Janajreh I Alshrah M and Zamzam S Mechanical recycling of PVC plastic waste stream from cable industry: A case study Sustain. Cities Soc. 18, p. 13-20.
[16] Raj M M Patel H V Raj L M and Patel N K 2013 Studies on mechanical properties of recycled polypropylene blended with virgin polypropylene Int. J. Sci. Inv. Today 2, 3 p. 194-203.
[17] Ronkay F 2013 Effect of recycling on the rheological, mechanical and optical properties of polycarbonate Acta Polytech. Hung. 10, 1 p. 209-220.