

THE INVESTIGATION OF POLYLACTIC ACID BASED NATURAL FIBER REINFORCED BIOCOMPOSITES FOR AUTOMOTIVE APPLICATIONS

Süleyman Sinan ÖZTÜRK*¹

¹*R&D Department of TOFAŞ TÜRK OTOMOBİL FABRİKASI AŞ, 16110 Bursa, TURKEY*

Abstract

An automotive application with renewable sources as environmentally sustainable solution has been studied for weight reduction. Hemp as non-woven natural fiber (NNF) and Polylactic acid (PLA) non-woven fibers has been used to form continuous mat by needle-punch method. This biobased mat structure was then hot formed to achieve good mechanical properties. The composition of the fiber was 50:50 / NNF:PLA and the density of the final sheet was 1300 gr/m², the final thickness of the biocomposite after themoforming was 2.2 mm with 2.1 GPa Flexural Modulus and 17 MPa Tensile Strength and 2.3% elongation at break. In this study, the mechanical performance of this biocomposite structure under water immersion, 40°C and 80°C was studied. The charpy impact strength was found 21kJ/m². The major outcome of this study is replacing conventional reinforcement materials with renewable sources which offers lower weight and tolerable mechanical strengths.

Keywords: PLA polymer, Hemp natural fiber, biocomposite, automotive application

DOĞAL ELYAF TAKVİYELİ POLİLAKTİK ASİT BİYOKOMPOZİT YAPILARIN ARAÇ UYGULAMALARI İÇİN İNCELENMESİ

Özet

Araç hafifletme projesine yönelik çevresel sürdürülebilirlik çözümleri kapsamında doğal kaynakların kullanılması çalışma yapılmıştır. Kenevir doğal fiberleri ve Polilaktik Asit (PLA) fiberleri, iğneleme yöntemi ile sürekli matlar haline getirilmiştir. Bu kompozit yapıya ısı işlem ile form edilerek iyi mekanik özellikler kazandırılmıştır. Kompozisyondaki fiber oranı 50:50 / Doğal Elyaf:PLA ve yoğunluk 1300 gr/m², nihai kalınlık ise 2.2 mm olarak hazırlanmıştır. Elde edilen kompozit yapı 2.1 GPa Eğilme Modülü ve 17 MPa Kopma Dayanımı ve 2.3% kopma uzaması değerlerine ulaşmıştır. Bu çalışmada, biyokompozit yapının su emdirildikten sonra, 40°C ve 80°C yaşlandırma sonrası mekanik özelliklerinin değişimi incelenmiştir. Charpy Darbe Dayanımı 21kJ/m² olduğu bulunmuştur. Bu çalışmada, konvansiyonel olarak üretilebilecek fiber katkılarla güçlendirilmiş, sürdürülebilir kaynaklarla üretilmiş hafifletilmiş kompozit yapı çalışılmıştır ve araç projelerinde kullanılabilir mekanik özellikleri sağlanmıştır.

Anahtar Kelimeler: PLA polimer, Kenevir doğal elyaf, biokompozit, otomotiv uygulamaları

* Corresponding author: suleymansinan.ozturk@tofas.com.tr, <https://orcid.org/0000-0002-6011-5673>

1. INTRODUCTION

As the new regulations are decreasing the emission limits of the greenhouse gases year by year, automotive industry focuses on weight reduction as the weight of the car is one of the parameter for the energy consumption. Most of the automotive companies keep working on composite structures among with lower weight high strength metal solutions. Due to potential low density of carbon fiber and natural fiber, such fiber reinforced composite solutions result into weight reduction [1-2]. Natural fibers have been vastly studied since they are abundant, sustainable, environmental friendly and cheaper than carbon fibers [3] The bast fibers such as kenaf, hemp and flax can be used as reinforcement to polymeric matrix. The randomly oriented fibers are more easily formed and eliminate the distortion when compared with the continuous fibers [4]. The natural fibers are hydrophilic and adhesion to the hydrophobic polymer matrix like polypropylene can be enhanced by using high melt flow polymers and by heating during process. Heating to high temperature can degrade natural fibers and lose the elasticity of the structure [5]. The adhesion problem of cellulose based fibers with hydrophobic polymers was improved by coupling agents like maleic anhydride or surface modification of the fiber surface [6-7].

A greener solution to form biocomposite is to use biodegradable polyester polymer matrix such as PLA. Natural fiber: PLA composites have been studied with different manufacturing techniques like sheet forming, extrusion and compression to understand the mechanical properties [8,9] The drawback of such biocomposites is the sensitivity of the fiber to humidity and heat which limits the use in automotive components like door panels, parcel shelves and trim parts of dashboard. The automotive application of natural fiber composites goes back to 1940s when Henry Ford first used hemp composite solution. Current composite solutions are around 1500gr/m² and 1000gr/m² can be achieved with thermoset process.

The aim of this study is to understand the mechanical properties of 50%:50% hemp fiber reinforced PLA fibers needle-punched to obtain mat which is then heated and compression molded to form a sheet. The effect of temperature on mechanical behaviour of the sheet was studied. The effect of humidity and change of the mechanical properties when exposed to water was studied. A further characterization technique such as TGA and DSC was performed to observe thermal behaviour of the composite structure. The adhesion of PLA to natural fiber was controlled by stereomicroscopy to understand the structure of the composite.

2. MATERIAL AND METHODS

Hemp used in this study was from industrial straw of *Cannabis sativa* defibering followed by mechanical refining. Average length of the fibers were $117 \pm 8 \mu\text{m}$ with average diameter of $141 \pm 47 \mu\text{m}$. The average length of the PLA fibers were between 33mm to 165mm, the tenacity of the fibers were $1.8 \text{ cN/dtex} \pm 25\%$.

The natural fibers and PLA fibers were needle-punched as 50:50 to produce 1300gr/m² of mat. The mat is then hot-pressed with roller at 195°C for 45 seconds. The thermoformed sheet is then machine cut to produce specimen for mechanical tests.

The visual control was performed with Leica DMI8 stereomicroscope. The DSC curve was observed with Netzsch 204 F1 and TGA graph was collected with Mettler Toledo TGA/DSC 1 Star. The mechanical tests were done with Zwick 10kN Universal Testing Machine. The Charpy impact strength test was performed with Instron Charpy Impact Analyzer.

3. RESULTS AND DISCUSSION

3.1 Density of Composite sheets

The weight/area of the sheet was calculated by measuring the weight of a 100mmx100mmx2.2mm sample and dividing by the total area. The weight/volume of the sheet was calculated by measuring the weight and volume of the sheet.

Table 1. The density of the sheet

Sheet Sample	Weight/Area gr/m ²	Weight/Volume gr/cm ³
1	1392	0.54
2	1369	0.58
3	1324	0.56
Average	1362	0.56

3.2 DSC of Composite sheets

The DSC curve was collected first heating from 30°C to 220°C with 10K/min heating rate and kept isothermal for 5 minutes (Figure 1). The cooling rate was 10K/min with 3 minutes isothermal and the second heating was to 220°C with 10K/min heating rate. The nitrogen atmosphere with 70mL/min purge flow and 20mL/min gas flow was used. The melting peak of

PLA was calculated from second heating curve as 166.8°C with 22.3 j/g entalpy of fusion. With the equation 1, the crystallinity of the polymer $X_c(\%)$ can be calculated where ΔH_m is the entalpy of fusion (J/g), and ΔH_m^0 is the standard enthalpy of PLA for 100% crystallinity, equal to 93.7 J/g. The crystallinity of the PLA polymer is 6.7%.

$$X_c(\%) = \frac{\Delta H_m}{\Delta H_m^0} \quad (1)$$

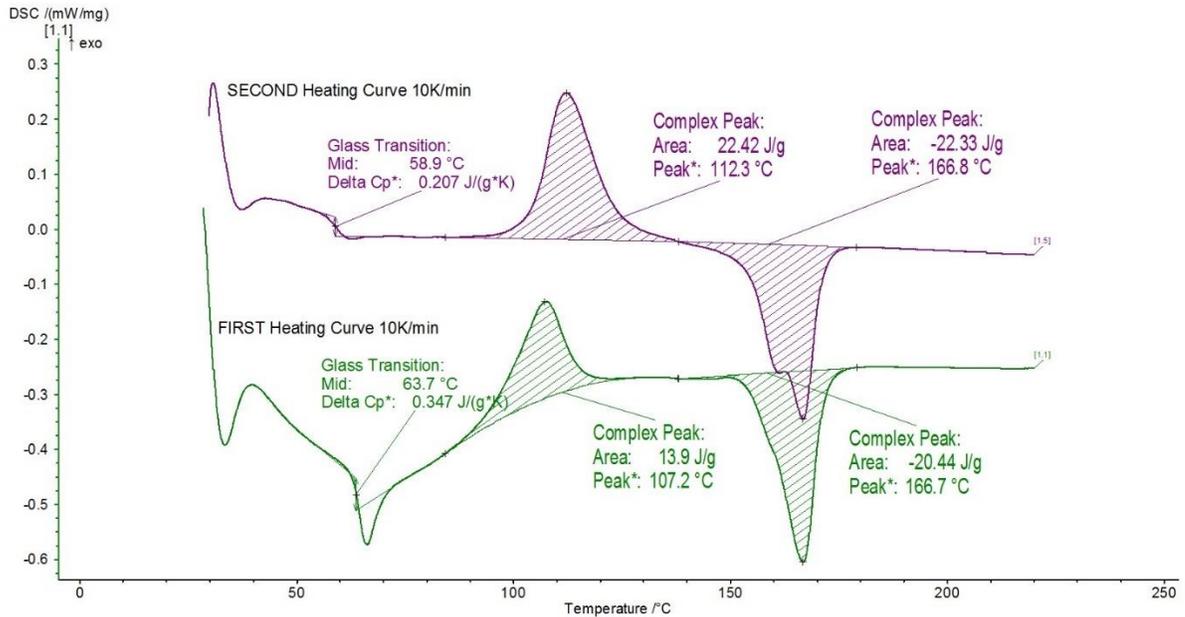


Figure 1. DSC curves of the composite structure, first heating curve (green), second heating curve (purple)

The glass transition of the PLA from first heating curve is 63.7 °C. The glass transition temperature of the polymer is important for defining the working temperature of the composite. In this case, the composite sheet will be further thermoformed to finished product where the composite will be glassy before 58.9 °C and softening will be affecting the mechanical properties.

3.3 TGA of Composite sheets

The TGA was measured to characterize the thermal stability of the composite structure (Figure 2). The heating curve was 20K/min from 30 °C to 600 °C under nitrogen atmosphere and isothermal for 5 minutes. Then heating to 800 °C with 20K/min under oxygen atmosphere. The flow of the gas was constant at 60 mL/min.

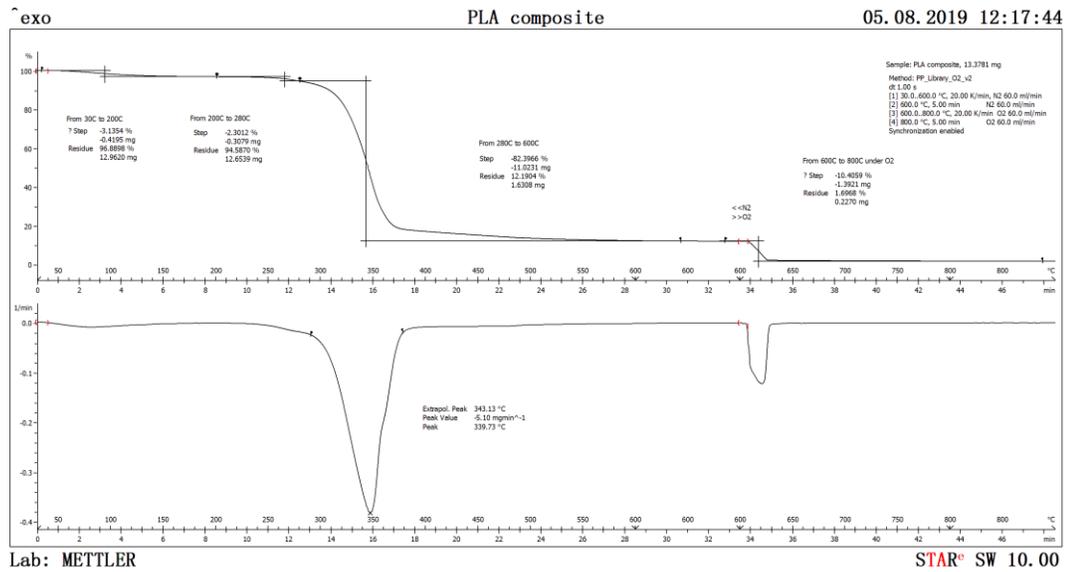


Figure 2. TGA graph of the composite structure (above), DTGA of the composite (below)

In the TGA graph, the weight loss of the composite was 82 % where from DTGA the decomposition peak was observed at 339.7 °C. The residual weight of the composite was 1.7 % which is mainly due to flame retardant used in the PLA fiber. The weight loss in the first step is due to water loss which was 3.1 %.

3.4 FT-IR spectrum of Composite sheet

The FT-IR spectrum was collected with Bruker Tensor II between 400-4000 cm^{-1} with 24 scans with 4 cm^{-1} resolution (Figure 3).

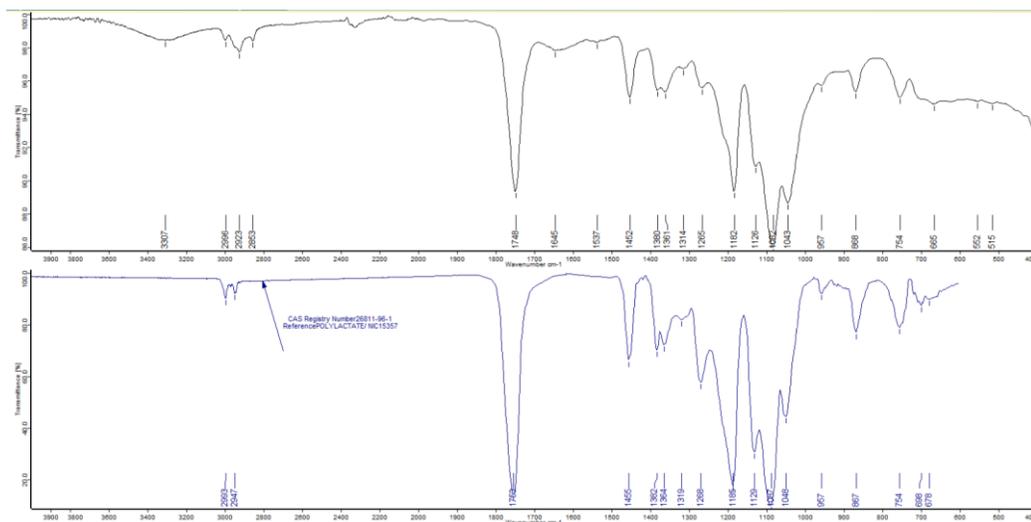


Figure 3. FT-IR spectrum of the composite structure (above), PLA from reference library CAS 26811-96-1 (below)

The -OH stretching band was observed between 3200-3500 cm^{-1} , the C=O stretching was at 1748 cm^{-1} , and C-O-C stretching was observed at 1182 cm^{-1} and 1082 cm^{-1} . The typical PLA spectrum was confirmed with reference library CAS 26811-96-1.

3.5 Mechanical testing of Composite sheet

3.5.1 Tensile Strength testing of Composite sheets

The composite sheets are mechanically cut according to ISO 527-4 Type2 where the testing speed was 50mm/min. Tensile stress was determined by dividing the force by the average specimen area. The tensile strength is the maximum tensile stress achieved before failure. The test is performed at 23 °C and another set of 3 samples were immersed in water for 4 hours and then measured. The results are listed in Figure 4.

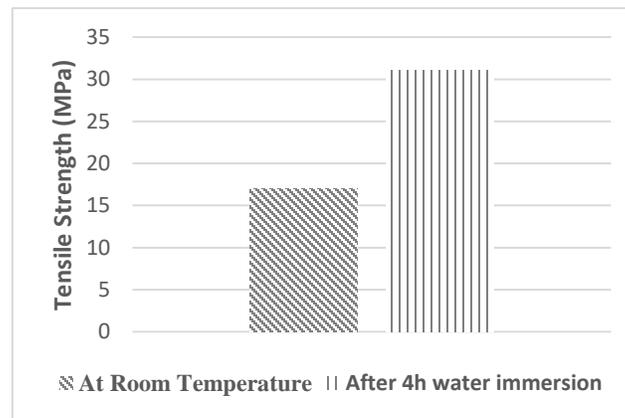


Figure 4. Tensile Strength of 50%:50% NF:PLA composite sheet at 23°C and after water immersion for 4 hours

The Tensile strength of composite at 23 °C was 17 MPa where after water immersion was increased to 31 MPa. The elongation at break at 23 °C was 2.3% where after water immersion the elongation increased to 5.2 %. Using water as treatment was acted as plasticizers to natural fibers. The cellulosic fibers are likely to swell after wetting which increases the elongation [10].

3.5.2 Flexural Strength testing of Composite sheets

The Flexural strength test was performed according to ISO 14125 Method A with three point set-up. The testing speed was chosen as 2 mm/min. Flexural strength as the maximum fiber stress before it cracks was measured. Flexural modulus also refers to modulus in bending is the ratio of maximum stress to maximum strain within elastic limit of stress-strain diagram.

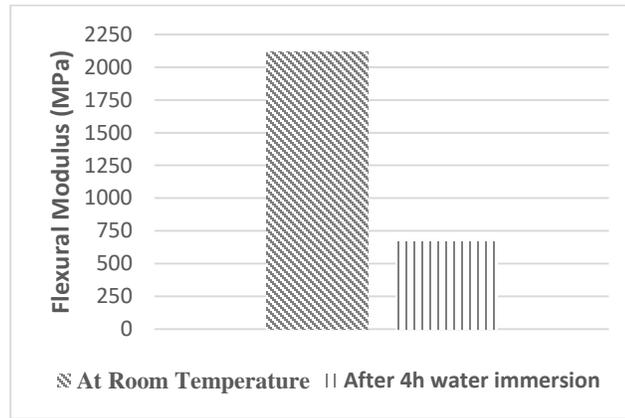


Figure 5. Flexural Modulus of 50%:50% NF:PLA composite sheet at 23°C and after water immersion for 4 hours

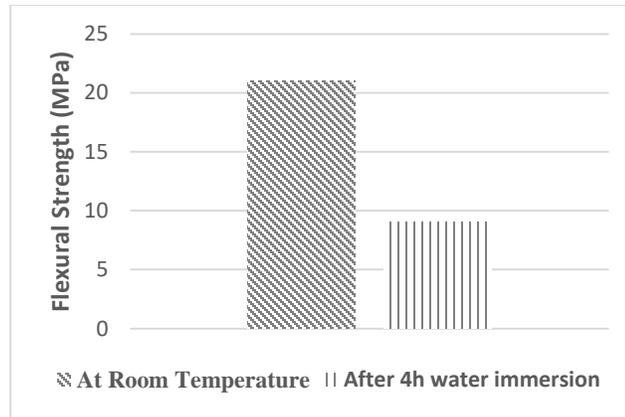


Figure 6. Flexural Strength of 50%:50% NF:PLA composite sheet at 23°C and after water immersion for 4 hours

The flexural modulus (Figure 5) and flexural strength (Figure 6) of the composite at 23 °C was 2113 MPa and 21 MPa, respectively. The immersion of composite to water for 4 hours resulted into a decrease in the flexural modulus and flexural strength to 667 MPa and 9 MPa, respectively.

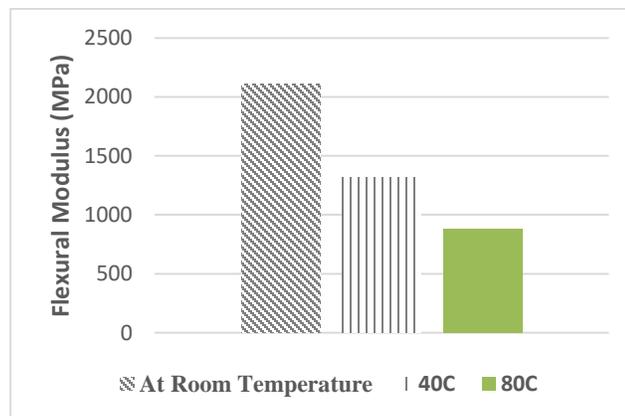


Figure 7. Flexural Modulus of 50%:50% NF:PLA composite sheet at 23°C, 40°C and 80°C

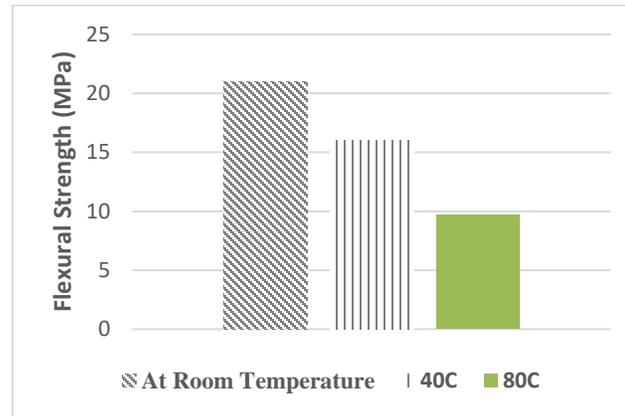


Figure 8. Flexural Strength of 50%:50% NF:PLA composite sheet at 23°C, 40°C and 80°C

The specimen are conditioned for 2 hours in the climatic chamber unit of the universal tensile machine at 23 °C, 40 °C and 80 °C. The flexural modulus decreases from 2113 MPa to 1314 MPa and 886 MPa when conditioned to 40 °C and 80 °C, respectively (Figure 7). The flexural strength decreased from 21 MPa to 16 MPa and 9.7 MPa after conditioning at 40 °C and 80 °C, respectively (Figure 8). The mechanical properties have decreased after heating which can be explained by the glass transition point observed around 63.7 °C in DSC first heating curve.

3.5.3 Charpy Impact testing of Composite sheets

The charpy impact test was performed according to ISO-179-1 (Table 2). The average of 21 kJ/m² was calculated from 5 specimen.

Table 2. The Charpy impact strength results at room temperature

Sample	Charpy impact kJ/m ²
1	21
2	22
3	21
4	21
5	21
Average	21.3

The charpy impact test above 18kJ/m² is like LyondellBasell Hostacom CR 1171 G, which is general polypropylene copolymer grade used for automotive interior trim parts.

3.6 Stereomicroscope image of Composite Sheet

The stereomicroscope image was taken at 5X magnification (Figure 8). The wetting of the natural fiber with the PLA polymer is observed where the voids are visible. No PLA fibers was observed in the composite structure where all the polymer fiber was melted.



Figure 8. Stereomicroscope image of NF: PLA composite

4. CONCLUSION

In the study, the natural fiber and PLA fiber were homogeneously mixed by needle-punch method. The low density of the composite resulted into a weight reduction solution which can be processed at industrial applications. The water immersion of composite sheet results into higher tensile strength which is mainly due to swelling of the fibers which acts as surface modification through the direction of the applied force. On the other hand, the bending test is perpendicular to fiber orientations which decreases flexural strength after water immersion. The mechanical strength has dropped by conditioning the specimen at high temperature due low glass transition of PLA. The wetting of the PLA to natural fiber was observed at stereomicroscope image where air gaps were observed.

Natural fiber composites are good candidates for interior automotive applications as sustainable and eco-friendly solution with low density and high strength with treatment. The major drawback for exterior automotive application is the water absorption and low glass transition



temperature. The study showed that PLA fiber can be alternatively used like PP fiber in compression moulding.

ACKNOWLEDGMENTS

This study was supported by TUBITAK project number: 3151221; we would like to thank Doç. Dr. Güralp Özkoç from Kocaeli University, Department of Chemical Engineering, for his support in this project.

REFERENCES

- [1] Bhattacharyya, D., Bowis, M., Jayaraman, K., Thermoforming woodfibre–polypropylene composite sheets, *Composites Science and Technology*, Volume 63, Issue 3-4, 353-365, 2003
- [2] Davey, S., Das, R., Cantwell, W.J., Kalyanasundaram, S., Forming of studies of carbon fiber composite sheets in dome forming processes, *Composite Structures*, 97, 310-316, 2013
- [3] Zampaloni, M., et al., Kenaf natural reinforced polypropylene composites: a discussion on manufacturing problems and solutions, *Composites: Part A*, 38, 1569-580, 2007
- [4] Hsiao, S.W., Kikuchi, N., Numerical analysis and optimal design of composite thermoforming process. *Computational Methods in Applied Mechanics and Engineering*, 177, 1-34, 1999
- [5] Xu, J., Widyorini, R., Yamauchi, H., Kawai, S., Development of Binderless Fibreboard from Kenaf Core, *Journal of Wood Science*, 52, 236-243, 2006
- [6] Feng, D., Caulfield, D.F., Sanadi, A.D., Effect of compatibilizer on the structure property relationships of kenaf-fiber/polypropylene composites, *Polymer Composites*, 22, 506-517, 2001
- [7] Mohanty, A.K., Drzal, L.T., Misra, M., Engineered reinforced polypropylene composites: influence of surface modifications and novel powder impregnation processing, *Journal of Adhesion Science and Technology*, 16, 999-1015, 2002
- [8] Auran Efendy, M.G., Pickering, K.L., Fibre orientation of novel dynamically sheet formed discontinuous natulra fibre PLA composites, *Composites: Part A*, 90, 82-89, 2016
- [9] Oksman, K., Skrifvars, M., Selin, J.F., Natural fibres as reinforcement in polylactic acid (PLA) composites, *Composites Science and Technology*, 63, 1317-1324, 2003



[10] Wang, W., Lowe, A., Kalyanasundaram, S., Effect of Chemical Treatments on Flax Fibre Reinforced Polypropylene Composites on Tensile and Dome Forming Behaviour, International Journal of Molecular Science, 16, 6202-6216, 2015