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PLA/PBAT/ENR TERNARY BLENDS: HEAT DISTORTION TEMPERATURE, OXYGEN TRANSMISSION RATE AND WATER VAPOR TRANSMISSION RATE

MISTURAS TERNAIRAS DE PLA/PBAT/ENR: TEMPERATURA DE DISTORÇÃO TÉRMICA, ÍNDICE DE TRANSMISSÃO DE OXIGÊNIO E ÍNDICE DE TRANSMISSÃO DE VAPOR DE ÁGUA

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Abstract
The aim of this research was to study the heat distortion temperature, oxygen transmission rate and water vapor transmission rate of PLA/PBAT/ENR ternary blends. Climate change, pollution, and depletion of natural resources are pressing global environmental issues. As the urgency of these issues increases, the need for sustainable alternatives becomes more apparent. Hence, the biodegradable plastics still have a limitation, i.e. high cost. This research studied methods of lowering the cost of biodegradable plastic and of improving its properties by using epoxidized natural rubber as another polymer of polymer blends. Then, ternary blends of poly (lactic acid), epoxidized natural rubber and poly (butylene adipate-co-terephthalate) were investigated and prepared by using twin-screw extruder, followed by use of a blown film extruder to fabricate biodegradable films. The proportion of ENR in the ternary blends was fixed at 10 wt%, with the remainder being PLA and PBAT. Moreover, the heat distortion temperature, oxygen gas transmission rate, and water vapor transmission rate were investigated.

Keywords: Biodegradable Polymers. Polymer Blends. Poly (Lactic Acid). Epoxidized Natural Rubber. Poly (Butylene Adipate-Co-Terephthalate).

Resumo
O objetivo desta pesquisa foi estudar a temperatura de distorção térmica, a taxa de transmissão de oxigênio e a taxa de transmissão de vapor de água de misturas ternárias PLA/PBAT/ENR. As mudanças climáticas, a poluição e o esgotamento dos recursos naturais são questões ambientais globais urgentes. À medida que a urgência dessas questões aumenta, a necessidade de alternativas sustentáveis se torna mais evidente. Portanto, os plásticos biodegradáveis ainda têm uma limitação, ou seja, o alto custo. Esta pesquisa estudou métodos para reduzir o custo do plástico biodegradável e melhorar suas propriedades usando borracha natural epoxidada como outro polímero de misturas poliméricas. Em seguida, misturas ternárias de poli(ácido lático), borracha natural epoxidada e poli(butileno adipato-co-tereftalato) foram investigadas e preparadas usando uma extrusora de dupla rosca, seguida pelo uso de uma extrusora de filme soprado para fabricar filmes biodegradáveis. A proporção de ENR nas misturas ternárias foi fixada em 10% em peso, sendo o restante PLA e PBAT. Além disso, foram investigadas a temperatura de distorção térmica, a taxa de transmissão de gás oxigênio e a taxa de transmissão de vapor de água.

Palavras-chave: *Polímeros Biodegradáveis. Misturas de Polímeros. Poli(ácido láctico). Borracha Natural Epoxidada. Poli(adipato-co-tereftalato de butileno).*

1 INTRODUCTION

The creation of compostable films to slightly replace petrochemical-based materials is affected by concerns about the environment. High waste generation is one of the major environmental concerns the world faces today, primarily driven by the consumption of non-biodegradable plastics and the agricultural methods (Noelia *et al*., 2025; Shakeel, 2023). On the other hand, the costs of biomaterials and synthetic plastics have a substantial difference when the weight basis is considered (Mohd & Shafat, 2022). In this research, ternary blends of poly (lactic acid), epoxidized natural rubber and poly (butylene adipate-co-terephthalate) were investigated and prepared by using twin-screw extruder, followed by use of a blown film extruder to fabricate biodegradable films. To obtain lower cost, attain biodegradation and improve some of the properties of the films, ENR derived from the partial epoxidation of natural rubber molecules was selected to be incorporated into the biodegradable films. Polymer blending is a vital method to obtain new materials that can meet different requirements. The incorporation of ENR with other polymers illustrates an important route to overcome the limitations of biodegradable materials. PLA is eco-plastics that are both bio-based and decomposable (Mohd & Shafat, 2022). Whereas, polylactic acid (PLA) is a type of thermoplastic aliphatic polyester produced by microbes through fermentation of plant starch like corn, sugarcane cassava. Its good physical properties and commercial availability make it very attractive, not only as a substitute for non-biodegradable polymers for commercial applications, but also for specific applications in medicine and agriculture. The main limitations of PLA are its high price and brittleness, which restricts its use. Blending PLA with ENR have been shown to improve its toughness and is a good to address the issue of cost-effectiveness (Yew et al., 2012; Yuan & Ruckenstein, 1998).

This research work studied ternary blends with PLA, PBAT and ENR; it was found that the incorporation of PBAT greatly increased the ductility of PLA/ENR blends. Then, blending a PLA/ENR binary blend with another flexible polymer could be a useful way to attain a new kind of material with excellent integrated performance. PBAT is a biodegradable random copolymer, specifically a copolyester of adipic acid, 1,4-butanediol and terephthalic acid. PLA/PBAT blends were investigated by (Jiang *et al*., 2006). The blend showed decreased tensile strength and modulus; however, elongation

and toughness dramatically increased. Whereas, the failure mode changed from brittle fracture of neat PLA to ductile fracture of the blends. In this respect, PBAT could be considered as a good candidate for toughening of more rigid polymers such as an ENR/PLA binary blend. Then, ENR/PLA/PBAT ternary blends were fabricated via a twin-screw extruder, followed by using a blown film extruder. This kept the ENR contentment at 10% by weight; the remainder consisted of other biodegradable polymers, PLA and PBAT, which varied in different ratios.

2 MATERIALS AND METHODS

PLA (grade 4032D) was purchased from NatureWorks (Blair NE, USA). The PLA was dried in a hot oven at 60 ^{o}C for 24 h. PBAT was purchased from BASF(Brazil) under trade name Ecoflex ®. ENR was purchased from Muang Mai Guthrie Public Co. Ltd. (Thailand). The ENR had an average molecular weight of $6.4x10^{5}$.

2.1 Processing

PLA was blended with ENR and PBAT at various compositions as shown in Table 1. Blends were fabricated in a twin-screw extruder (LTE 20-40) with diameter 26 mm. A process length to diameter ratio (L/D) is 40/1. The conditions of processing were 160-180 ^{o}C. Finally, pellets are cut by pelletizer to pellet sizes of 2.5 mm and pellets obtained at 5.5 m/min. The films were developed via blown film extrusion type LE20-30/C Single screw) with diameter 100 mm, produced by Lab Tech Engineering Company.

2.2 Water vapor permeability (WVP)

Water vapor permeability (WVP) was measured using water vapor permeation tester model Lyssy L80-4000 and determined according to ISO 15106-I:2003 (E).

2.3 Oxygen gas transmission rate (OTR)

Oxygen gas transmission rate was measured using oxygen permeation tester; Illinois model 8000 and determined according to ASTM D3985-05.

2.4 Heat distortion temperature (HDT)

The *heat distortion temperature* (*HDT*) is a measure of the polymer's resistance to distortion under a given load at an elevated temperature. The *heat distortion temperature* (*HDT*) was measured using heat distortion tester model HDT VICAL AUTO and determined according to ASTM D-648. The *temperature* at which a bar of material is deformed is recorded as the *HDT* at *264 psi.*

Table 1

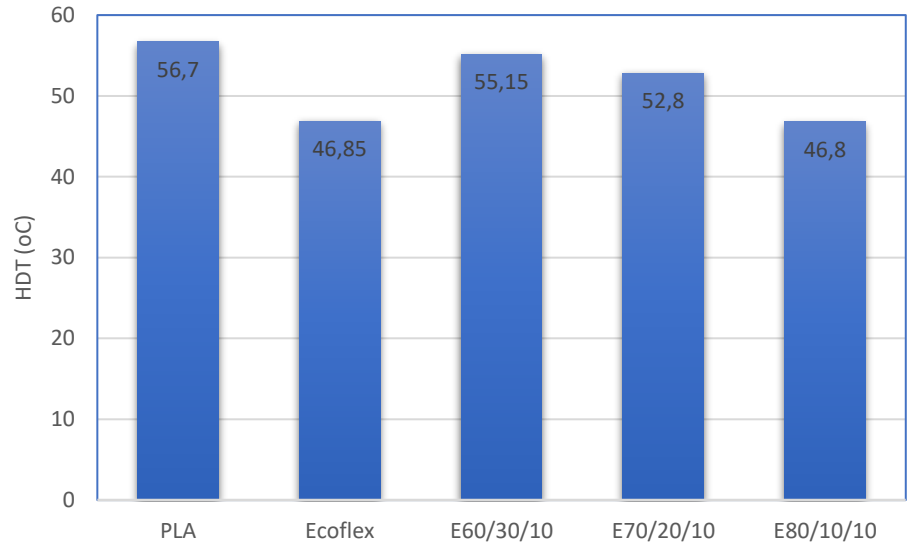
Sample designation and formulation

Sample code	Composition	Part (wt.%)
E60/30/10	Ecoflex/PLA/ENR	60/30/10
E70/20/10	Ecoflex/PLA/ENR	70/20/10
E80/10/10	Ecoflex/PLA/ENR	80/10/10

3 RESULTS AND DISCUSSION

Figure 1

Heat distortion temperature of biodegradable films



The heat distortion temperature is shown in Table 1. and Figure 1. The heat distortion temperature of biodegradable films increased with an increase in PLA content because the addition of PLA in blends lead to the increase of thermal stability. Whereas, the heat distortion temperature of E60/30/10 exhibited the maximum value (55.15 ^{o}C).

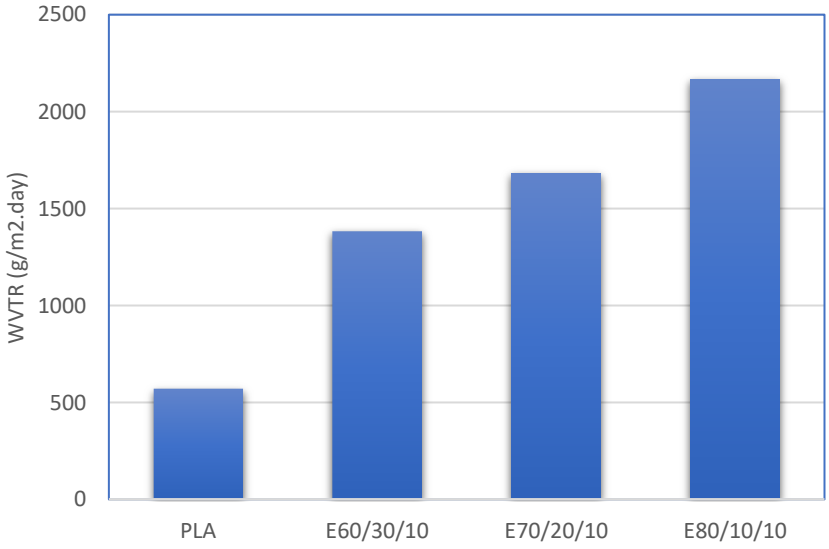
Table 2

Water vapor permeability (WVP) of biodegradable films

Sample code	Water vapor transmission rate (at 35^{o}C, 90%RH) g/m^2.day
Pure PLA	571
E60/30/10	1,384
E70/20/10	1,683
E80/10/10	2,167

Figure 2

Water vapor transmission rate of biodegradable films



The film water vapor permeability (WVP) data is shown in Table 2 and Figure 2. The water vapor transmission rate (WVTR) increased with an increase in PBAT content in order to the water vapor permeability (WVP) of pure PBAT is higher when compared with the values of other materials such as PLA.

Figure 3

Oxygen transmission rate of biodegradable films

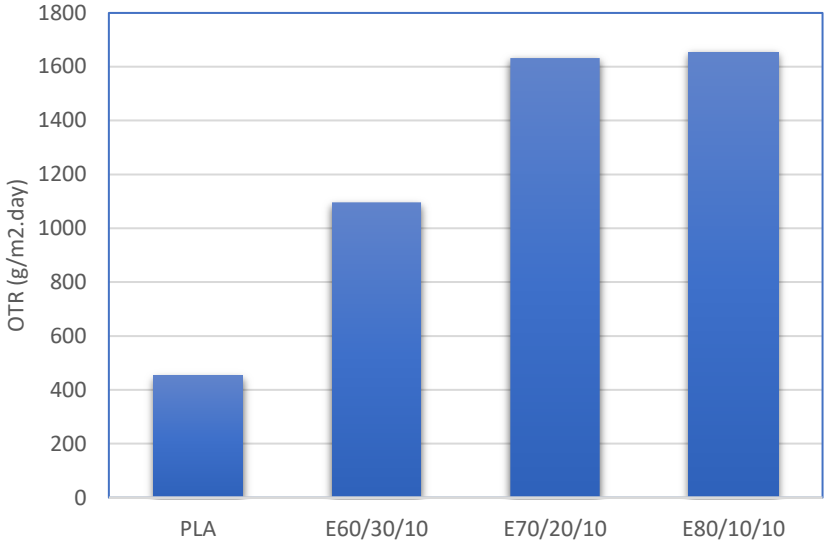


Table 3

Oxygen transmission rate of biodegradable films

Sample code	Oxygen gas transmission rate (at 38ºC, 90%RH) $g/m^2.day$
Pure PLA	454
E60/30/10	1,096
E70/20/10	1,632
E80/10/10	1,653

Table 3 and Figure 3 exhibit the oxygen transmission rate of biodegradable films. The results revealed that there is an increase in oxygen transmission rate, whereas it decreases in polylactic acid (PLA) containing in blends. In addition, oxygen transmission rate (OTR) is a measure of the volume of oxygen that diffuses through a barrier over a given period of time, the experimental results revealed that the oxygen transmission rate of the biofilm (E80/10/10) was higher than that of others because the oxygen transmission rate of polylactic acid (PLA) is dramatically low.

4 CONCLUSION

The heat distortion temperature, oxygen transmission rate and water vapor transmission rate of PLA/PBAT/ENR ternary blends were studied. For the properties of ternary blends, it was found that the heat distortion temperature of blends produced using a blown film extruder decreased with an increase in PBAT content. Whereas, it was found that the water vapor transmission rate (WVTR) increased with an increase in PBAT content due to the higher of PBAT when compared with the value of other materials such as PLA. Furthermore, the increase of PBAT containing in blends lead to the increase of oxygen transmission rate of biodegradable films.

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Authors' Contribution

All authors contributed equally to the development of this article.

Data availability

All datasets relevant to this study's findings are fully available within the article.

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