The effect of rotor speed on EVA/PBAT blend using Twin screw extruder and foamability of EVA/PBAT blend

Chanchai Thongpin¹,*, Kanyapak Theanchai¹, Pattama Riangpha¹ and Weena Anantawut¹

Department of Materials Science and Engineering, Faculty of Engineering and Industrial Technology, Silpakorn University, Sanamchandra Palace Campus, Nakhon Pathom, 73000, Thailand.

*Corresponding author: THONGPIN_C@su.ac.th

Abstract. This research is aimed to study EVA/PBAT blends with various EVA and PBAT composition and screw speeds. At PBAT contents 40% EVA and PBAT present as co-continuous phase morphology. The morphology did not change due to screw speed. At PBAT 50% by weight, co-continuous phase occurred and the morphology becomes dispersed as screw speed was increased to 80 and 100 rpm. Extrudates obtained from twin screw extruder were tensioned under tensile force. The results show elastomeric behavior of the blends. Elongation at break was higher than 800 % strain and exhibited strain hardening characteristic. Secant modulus of EVA/PBAT blends was increased with PBAT contents due to the high modulus of semi-crystalline PBAT. Content of EVA controls elongation ability of the blends. EVA/PBAT blends with the content of 40/60, 50/50, 60/40, 70/30 and 80/20 were form for foam and found that all composition could be able to be foam. This could be further studied and carried on for shoes sole application.

1. Introduction

Dispersion of ethylene-vinyl acetate copolymer (EVA) are randomly disperse in main chain by ethylene copolymer, polar vinyl acetate (VAc) units. Different VAc content in copolymers changes EVA from plastics to elastomers in several applications. A good example of this is solar cell encapsulant[1], foot ware midsole and toy industry. Each application chosen are dependent upon the grades of EVA. Each type of the EVA is determined according to the type of curing agent, which can be of three types: standard cure (SC), fast cure (FC) and ultra-fast cure (UFC). In order to increase the yield of the rolling process, the fast or ultra-fast cure formulations are more common as fast cure materials can achieve higher yield due to a reduction of almost 40% of the lamination process. The EVA cure is needed to change the physical properties of an encapsulant by chemical reaction through polymerization, in the presence of heat and catalyst, alone or in combination [2]. In addition, the blends of EVA/PBAT possess high thermo shrinkage and high stability when subjected to aging, weathering, and aggressive media. Therefore, the EVA/PBAT blends are used in many applications including foam material, multilayer packaging films and sheets, automobile parts, agricultural films, medical tubes, and so on [3]. Nevertheless, an important shortcoming in blending different polymers is that the polymers often form immiscible and incompatible blends, displaying a coarse morphology with particles of dispersed phase badly adherent to the matrix and badly distributed. Consequently, a compatibilization process is necessary in order to modify the interfacial properties in immiscible
polymer blend, thus leading to a reduction of the interfacial tension coefficient and the formation and stabilization of the desired morphology [4].

Poly (butylene adipate-Co-terephthalate) (PBAT), also known under the trade name of Ecoflex, has attracted great interest as potential substitution of low density polyethylene (LDPE), especially for packaging materials such as lawn waste bags, food containers or film wraps, among others. Apart from its biodegradability, PBAT offers attractive physical properties, including high flexibility, high strength and good tear resistance, and it is approved for food contact applications [5]. In addition, it has short moulding cycles and excellent sealing performance, while it is easy to process at high extrusion speeds [6]. As a widely used PBAT blend which can blend with other polymers such as PLA/PBAT blends [7], PBAT/EVOH blends [8], ATC/PBAT blends [9] and PP/PBAT blends [10].

In this part of our research EVA/PBAT blends will be prepared and their morphology, mechanical properties will be evaluated. The chain extender such as joncryl4300 will be added in order to investigate its effect of EVA/PBAT blends. As EVA has been mainly applied for foam using as shoes sole application, this early work will preliminarily study the foamability of these blends.

2. Experiment
2.1. Materials
Polyethylene vinyl acetate, EVA, N8038 was purchased from TPI POLENE Co. Ltd., Thailand, of which the Tm, density and melt flow index are 86 °C, 0.941 g/cm³ and 2.8 g/10 min respectively. Polybutylene adipate-co-terephthalate, PBAT, Ecoflex F blend C12000 was obtained from POLYMAT Co. Ltd., Thailand, of which its Tm, density and melt flow index are 190 °C, 1.260 g/cm³ and 3.8 g/10 min respectively.

2.2. Melt Blending of EVA/PBAT
Before melt blending, EVA and PBAT were dried at 50°C under oven for 6 hours. EVA/PBAT with various composition ratios was fed into a twin screw extruder (SHJ-25 ENMACH, Thailand) with the temperature profile from zone 1-7 and die as follows: 120/120/120/130/130/140/150/120 and various screw speed at 60, 80 and 100 rpm. EVA/PBAT extrudate was collected and cooled to room temperature before being kept in a plastic bag until used. In the case of blend with joncryl4300, the blend was mixed with joncryl4300 before being fed into the extruder.

2.3 Characterization
Morphology: The morphology of the EVA/PBAT blend was investigated using scanning electron microscope, SEM (TM3030 Hitachi, Japan). The cryogenic fractured surface of the extrudates were platinum coated using sputtering before investigation under SEM using 15.0 kV accelerated of electron beam produced from tungsten filament.

Tensile test: Tensile properties were performed by using Instron universal testing machine (Instron 5900, USA) at room temperature, crosshead speed at 500 mm/min and under load 5 kN follow ASTM D 412 type C.

3. Results and Discussion
3.1 Morphology of EVA/PBAT
SEM micrographs of EVA/PBAT with various ratios and different rotor speed of mixing are shown in Figure 1-3. Figure 1 show EVA/PBAT at the composition ratio 10-40 % EVA of which EVA was found dispersed in PBAT phase seen as dark spherical particles except for as high as 40 % EVA where EVA particles appeared as sea-island morphology where EVA presents as island. It could be noticed that the sea-island morphology of EVA in PBAT could be overcome when the screw speed was increased to 100 rpm due to high shearing during melting. For blend systems containing low contents of PBAT shown in Figure2, phase inversion occurred. PBAT was found dispersed in EVA being as major phase and PBAT particles size was increased with PBAT content as shown in Figure2(b). Co-continuous and sea-island morphology were present in the blend with 60% PBAT content [11]. This is
caused by the higher MFI of PBAT (3.8 g/10 min) than that of EVA (2.8 g/10 min) resulting larger dispersed phase comparing between 60/40 and 40/60 of EVA/PBAT. In addition, morphology of the blend was changed to co-continuous phase with small PBAT phase size at higher screw speed. At EVA content 70-90 % by weight, PBAT become disperse in EVA. Also its particles were reduced with the high shear rate applied by high screw speed. In fact co-continuous phase morphology has also occurred at screw speed 60 rpm with the content of 50/50 as seen in Figure 3. The higher screw speed led to high shearing and hence sheared PBAT into smaller particles.

**Figure 1** SEM micrographs of high PBAT composition blends speed of mixing at 60, 80 and 100 rpm.
Figure 2 SEM micrographs of low PBAT composition blends speed of mixing at 60, 80 and 100 rpm.

Joncryl4300 is in fact epoxidized soybean oil (ESO) with molecular weight of 5500 [12], its structure is shown in Figure 6 (c). As shown in Figure 6 (b) that addition of joncryl4300 leads to larger EVA particles (dark domain) compared with unfilled joncryl4300 blends. This could be due to interaction between EVA molecule was increased by the interaction between ESO and EVA molecules as found by the MFI of EVA/ESO [13]. In our research, MFI of PBAT filled with joncryl4300 was decreased from 18.564 g/10min to 11.888 g/10 min where that of EVA was increased from 3.244 to 3.490 g/10 min. These results indicated that addition of joncryl4300 results in high viscosity of PBAT and resulting in shearing applied on EVA phase. As a result EVA phase become changed as shown for in Figure 3.

Figure 3 SEM micrographs of EVA/PBAT blend with composition of EVA and PBAT 50/50.

3.2 Tensile Properties

Effect of rotor speed of mixing

Tensioning of EVA/PBAT blends were performed at the crossed head speed of 500 mm/min using extrudate extruded from twin screw extruder. Their stress-strain of the representative blend composition of 30/70, 50/50 and 70/30 mixed at different screw speeds at 60, 80 and 100 rpm are exhibited in Figure 4. Tensile properties obtained from stress-strain curves obtained from the blends are also depicted in Figure 5. It could be noticed that stress-strain behavior of the blends shows strain hardening behavior due to their elastomeric character. As the screw speed was increased, dispersed particles become smaller and hence tensile strength was increased especially for the blends containing high EVA contents.

Figure 4 Stress strain curves of EVA/PBAT blends represented by the ratios 30/70, 50/50 and 70/30 rotor speeds of mixing at 60 (a), 80 (b) and 100 rpm (c).
Figure 5 Secant modulus (a), tensile strength at break (b) and elongation at break (c) of EVA/PBAT blend with various blend compositions and different screw speeds.

Effect of joncryl4300 on EVA/PBAT blend
As shown in mechanical behavior, it was found that the properties were controlled by EVA domain in the blend. Addition of joncryl4300 was added in order to improve compatibility of the blends. As explained earlier that addition of joncryl4300 led to decreased MFI of PBAT and slightly increased MFI of EVA this results in phase changed of EVA/PBAT as show in Figure 6. The more joncryl4300 added the lower secant modulus of EVA/PBAT blend was obtained, as elucidated in Figure 7. However, tensile strength and elongation at break were not significantly changed by the presence of joncryl4300. This should be due to the elastomeric behavior of EVA/PBAT blends, where strain hardening control strength and elongation at break of the blends.

Figure 6 SEM micrographs of EVA/PBAT blends without processed at 100 rpm. (a) and without (b) Joncryl4300) depicted in (c)

Figure 7 Secant modulus (a), tensile strength at break (b) and elongation at break (c) of EVA/PBAT blend, composition 50/50 with joncryl4300 content of 0, 1 and 3 phr.

Foam ability of EVA/PBAT blends
As mentioned earlier that these blends will be applied to foam formation thus some blends were preliminarily selected and kneaded on a two roll mill with dicumyl peroxide and blowing agent and
consecutively form foam under compression molding at 160 °C. Foam morphology of EVA/PBAT blends is shown in Figure 8. Morphology of foam formed show that blends of containing PBAT from 20-60 % could be processed. More information about the foam is going on and will be studied in detail and be reported in our further work.

**Conclusion**

EVA/PBAT blends with various EVA and PBAT composition were prepared using twin screw extruder and screw speeds were varied as 60, 80 and 100 rpm. At PBAT contents 40% EVA, PBAT present as co-continuous phase morphology. The morphology did not change due to screw speed. At PBAT as high as 50% by weight, co-continuous phase occurred and the morphology becomes dispersed as screw speed was increased to 80 and 100 rpm. Extrudates obtained from twin screw extruder were tensioned under tensile force. The results show elastomeric behavior of the blends. Elongation at break was higher than 800 % strain and exhibited strain hardening characteristic. Secant modulus of EVA/PBAT blends was increased with PBAT contents due to the high modulus of semi-crystalline PBAT. Content of EVA controls elongation ability of the blends. EVA/PBAT blends with the content of 40/60, 50/50, 60/40, 70/30 and 80/20 were form for foam and found that all composition could be able to be foam. This could be further studied and carried on for shoes sole application.

**Figure 8** Foam prepared from EVA/PBAT blends at the content 40/60 50/50 60/40 70/30 and 80/20 without joncryl4300 and with joncryl4300.

**References**

[1] Jin, J., Chen, S. and Zhang, J. 2010 *Polymer Degradation and Stability* **95** p 725-732.
[2] Cândida, M. Oliveira, D.C., Sonia, A., Diniz, A., Marcelo, C., Viana, A., Freitas, D.V. and Lins, C. 2018 *Renewable and Sustainable Energy Reviews* **81** p 2299-2317.
[3] Alothman, Y.O. 2012 *Advances in Materials Science and Engineering* p 1-10.
[4] Mantia, F.P. Morreale, M., Botta, L., Mistretta, C. M., Ceraulo, M. and Scaffaro, R. 2017 *Polymer Degradation and Stability* **145**(1) p 79-92.
[5] Ferreira, V.F., Cividanes, S.L., Gouveia, F.R. and Lona, L. 2019 *Polymer Engineering & Science* **59** p 7-15.
[6] Liu, W. , Liu, S., Wang, Z., Liu, J., Dai, B., Chen, Y. and Zeng, G. 2020 *European Polymer Journal* **122** p 1-8.
[7] Nofar, M., Salehiyan, R., Ciftci, U., Jalali, A. and Durmuş, A. 2020 *Composites Part B: Engineering* **182**(1) p 1-35.
[8] Elhamnia, M., Motlagh, H.G. and Jafari, H.S. 2020 *Polymer Testing* **82** p 1-31.
[9] Sellami, F., Kebiche, S.O., Marais, S., Couvrat, N. and Fatyeyeva, K. 2019 *Reactive and Functional Polymers* **139** p 120-132.
[10] Oliveira, A.T., Oliveira, R.R., Barbosa, R., Azevedo, B.J. and Alves, S.T. 2017 Carbohydrate Polymers 168 p52-60.

[11] Dil, J.E., Carreau, J.P. and Favis, D.B. 2015 Polymer 68 p 202-212

[12] Reddy, M. 2014 joncryl ADR (epoxy-functional polymer or oligomers) (online) Available: https://www.slideshare.net/MuniswamyReddy [2020, February 23].

[13] Khankrua, R., Pongpanit, T. Paneetjit, P. Boonmark, R. Seadan, M. and Suttiruengwong, S. 2019 Polymer MDPI 11 p 1-17.