



Chiang Mai J. Sci. 2018; 45(5) : 2079-2091

<http://epg.science.cmu.ac.th/ejournal/>

Contributed Paper

## Disintegration Testing of Biodegradable Poly(L-lactide) / Thermoplastic Polyurethane Melt Blended Films

Kanyarat Suthapakti\* [a], Robert Molloy [b,c] and Thanawadee Leejarkpai [d]

[a] Division of Product Development Technology, Faculty of Agro-Industry, Chiang Mai University, Chiang Mai 50100, Thailand.

[b] Polymer Research Group, Department of Chemistry, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand.

[c] Materials Science Research Center, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand.

[d] National Metal and Materials Technology Center, National Science and Technology Development Agency, Thailand Science Park, Pathum Thani 12120, Thailand.

\* Author for correspondence; e-mail: [kanyarat.cmu@gmail.com](mailto:kanyarat.cmu@gmail.com)

Received: 1 November 2017

Accepted: 30 April 2018

### ABSTRACT

Poly(L-lactide) (PLL) has been blended with a polycaprolactone-based thermoplastic polyurethane (TPU) elastomer as a toughening agent for potential use as a biodegradable film packaging material. The effects of adding a synthesized poly(L-lactide-*co*-caprolactone) (PLLCL) copolymer as a compatibilizer was also studied. Both 2-component (PLL/TPU) and 3-component (PLL/TPU/PLLCL) blends were prepared by melt mixing, hot-pressed into thin films and their compostability characterized in a standard ISO disintegration test under controlled laboratory-scale conditions. The results showed that both the 2- and 3-component blended film samples (25 × 25 mm) disintegrated completely (100%) to fragments less than 2 mm in size within 30 days. These results were similar to those for PLL alone which indicated that neither the TPU nor the PLLCL had retarding effects on the PLL's biodegradability. It is concluded that both the PLL/TPU and PLL/TPU/PLLCL blends meet the criteria for disintegration and compostability under simulated composting conditions.

**Keywords:** poly(L-lactide), thermoplastic polyurethane elastomer, poly(L-lactide-*co*-caprolactone, immiscible blend, disintegration test, compostability, biodegradable film

### 1. INTRODUCTION

Poly(L-lactide) (PLL), or poly(lactic acid) (PLA) as it is commonly referred to in the bioplastics industry, is a synthetic aliphatic poly- $\alpha$ -ester which has now become the most important commercial bioplastic currently

available. Its rise to prominence in recent years has been well documented and there is now an extensive library of information available on PLL in both books and journals [1-8]. Apart from being biodegradable and

compostable, the main advantages of PLL are its rigidity, good transparency as film, cups and bottles, and its processability using conventional thermoplastic processing equipment. However, PLL also has some notable disadvantages such as its low heat deflection temperature of around 60 °C, brittleness in certain applications, slow rate of crystallization, and inferior water vapor and gas barrier properties. Consequently, in order to diversify PLL's range of applications, there is increasing interest nowadays in how its properties can be modified by, for example, (1) copolymerization of L-lactide with other cyclic ester monomers, (2) the use of additives such as nucleating agents, plasticizers and impact modifiers, (3) blending with other polymers, and (4) nanocomposites with inorganic materials such as clay, zinc oxide, titanium dioxide and carbon nanotubes. Of these four approaches, blending with other polymers has received the most attention and it is this approach which has been employed in the work described here.

PLL has been blended with a wide range of different polymers, mainly aliphatic polyesters or polymers containing substituent ester groups in the expectation that the polar interactions between the ester groups would increase the blend's compatibility. Examples include blends with polycaprolactone (PCL), polyhydroxybutyrate (PHB), poly(butylene succinate) (PBS), poly(vinyl acetate) (PVAc), and cellulose acetate butyrate (CAB). Invariably however, these blends are largely immiscible over a wide range of composition. Despite the ester group interactions, there is only very limited compatibility in the amorphous part of the matrix. Consequently, these blends have so far found very limited application.

Following on from work recently published [9], this paper now describes the disintegration testing of thin films of melt

blends of PLL toughened with a PCL-based thermoplastic polyurethane (TPU) elastomer. Since previous studies [10-17] had shown that PLL-TPU blends were also largely immiscible, a novel aspect of this present work has been to incorporate a purpose-designed third component which can act as a compatibilizer by improving the interfacial adhesion between the PLL matrix and the dispersed TPU domains. This third component is a poly(L-lactide-*co*-caprolactone), PLLCL, statistical copolymer which has structural LL and CL repeat units in common with both the main PLL and PCL-based TPU components respectively. The target application for these PLL/TPU blends is biodegradable film packaging although other applications based on injection molded parts and extruded fibers are also possible.

## 2. MATERIALS AND METHODS

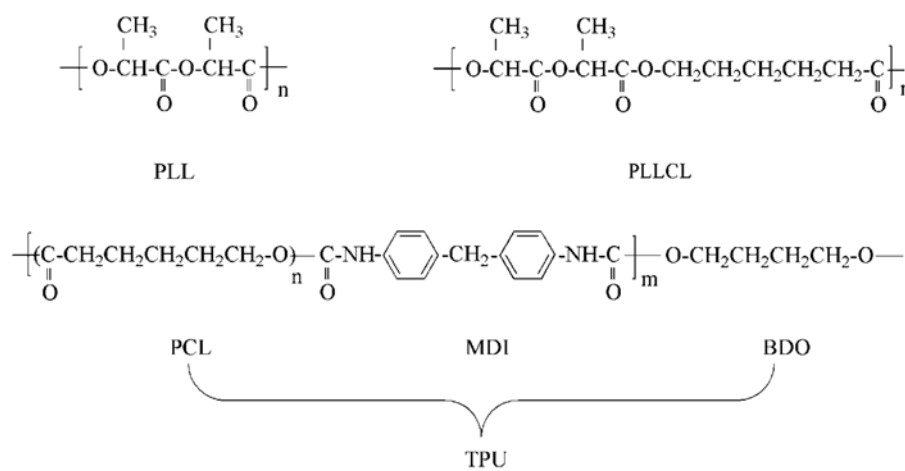
### 2.1 Materials

The PLL used in this research was a commercial product, Ingeo™ 4042D Film Grade PLA (NatureWorks®), in pellet form with number-average and weight-average molecular weights,  $\bar{M}_n$  and  $\bar{M}_w$ , of  $1.52 \times 10^5$  and  $2.55 \times 10^5$  g/mol respectively, as determined by gel permeation chromatography (GPC). The polycaprolactone-based thermoplastic polyurethane (TPU) elastomer used was also a commercial product, Pellethane™ 2102-75A (Lubrizol Corporation), in pellet form. Its block-type structure consisted of alternating hard and soft segments based on 4,4'-methylenediphenyl-1,1'-diisocyanate (MDI) and polycaprolactone diol respectively, with 1,4-butanediol (BDO) as a chain extender. The TPU elastomer had  $\bar{M}_n$  and  $\bar{M}_w$  values of  $1.28 \times 10^5$  and  $2.62 \times 10^5$  g/mol respectively from GPC.

The poly(L-lactide-*co*-caprolactone), PLLCL, copolymer was synthesized via the

ring-opening copolymerization in bulk of equimolar amounts of L-lactide (LL) and  $\epsilon$ -caprolactone (CL) at 120 °C for 72 hrs using 0.1 mol % stannous octoate ( $\text{SnOct}_2$ ) as the initiator. The copolymer was purified by cutting into small pieces and heating under vacuum at 40 °C for 24 hrs to constant

weight in order to remove any residual monomer. The PLLCL had  $\overline{M}_n$  and  $\overline{M}_w$  values of  $2.43 \times 10^4$  and  $4.40 \times 10^4$  from GPC and a composition of LL:CL = 51:49 (mol %) from  $^1\text{H-NMR}$ . The chemical structures of the PLL, PLLCL copolymer and TPU are shown in Figure 1.

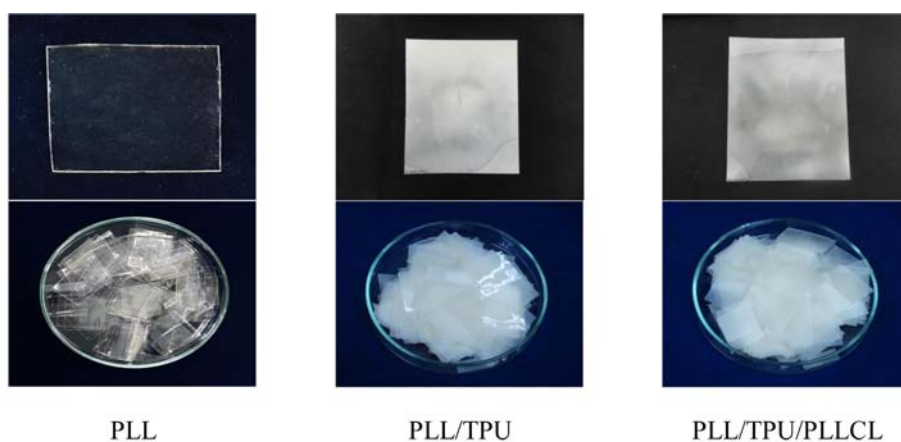


**Figure 1.** Chemical structures of the PLL, PLLCL and TPU blend components.

## 2.2 Polymer Blending and Film Preparation

Both 2- and 3-component blends were prepared in parts by weight (pbw) ratios of PLL/TPU 90/10 and PLL/TPU/PLLCL 90/10/10, as reported previously [9]. Blending was carried out by means of melt mixing using an internal mixer (Haake PolyLab) at temperatures of 195 °C for PLL/TPU and 190 °C for PLL/TPU/PLLCL for 20 mins. These were the lowest temperatures and shortest times necessary to ensure complete melting and homogeneous mixing while at the same time minimizing the possibility of thermal degradation.

After the melt blends had cooled and been ground into small pieces, thin films of thickness  $150 \pm 10 \mu\text{m}$  were prepared using a LabTech Engineering LP-S-20 Laboratory Press at a temperature of 180 °C for 5 min under a pressure of 20 MPa. The films, including those of PLL alone, were then cut into square pieces of dimensions  $25 \times 25 \text{ mm}$  and stored in a desiccator prior to testing. The films both before and after cutting are shown in Figure 2. Whereas the PLL film was transparent, the PLL/TPU and PLL/TPU/PLLCL blended films were translucent due to phase separation of the PLL and TPU.



**Figure 2.** The PLL, PLL/TPU and PLL/TPU/PLLCL films before and after cutting.

### 2.3 Instrumental Methods

pH was determined according to the ISO 10390:1994 standard method [18] using a Metrohm 785 DMP Titrino pH Meter.

Moisture content and total dry solids were determined according to the ISO 11465:1933 standard method [19] by drying at 105 °C to constant mass in a Binder drying oven series ED-240 D #05-89652.

Volatile-solids content was determined from the amount of residue remaining from a known mass of test material or compost after incineration in a Lenton furnace at 550 °C.

Total organic carbon contents of test materials were determined using a Shimadzu TOC-VCPN Total Organic Carbon Analyzer.

For microscopic analysis of film fracture surfaces, specimens were mounted on stainless steel stubs with a conductive carbon tape, gold-coated and then imaged using a JEOL JSM 5910KV Scanning Electron Microscope (SEM) operating at an accelerating voltage of 15 kV at 23 °C.

## 3. DISINTEGRATION TESTING

### 3.1 Standard Testing Procedure

Disintegration testing was performed according to the ISO 20200-2012 standard method [20]. The solid matrix used consisted

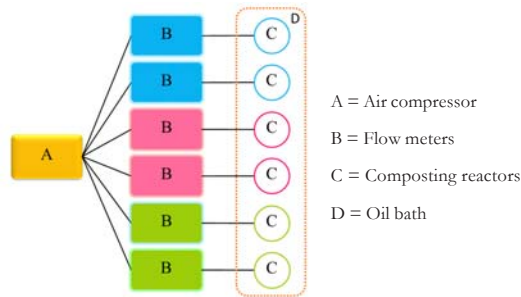
of a synthetic solid waste with mature compost taken from a commercial composting plant. The composition of the synthetic waste was as designated in the standard method. Square-shaped pieces of the film test materials (25 × 25 mm) were composted with this solid matrix in a temperature, aeration and humidity-controlled environment. The degree of disintegration was determined after the film samples had been composted for 84 days by sieving the final matrix through a vibrating sieve column comprising 10, 5 and 2 mm sieves in order to recover the non-disintegrated (size ≥ 2 mm) residues. The reduction in mass of the test sample was considered as disintegrated material for calculation of the degree of disintegration.

### 3.2 Sample Preparation and Composting System

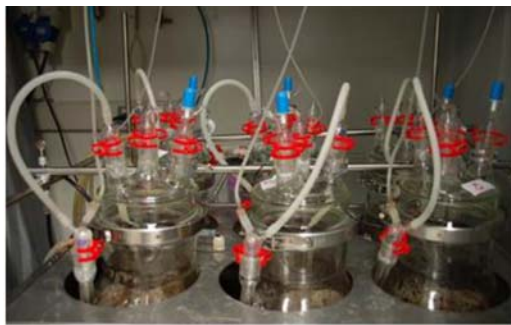
For each test, 3 composting reactors of volume 6 liters were each charged with a mixture of approximately 3000 grams of the synthetic waste and approximately 60 grams of the film sample cut into 25 × 25 mm squares. Each test was conducted in duplicate under controlled conditions in an oil bath maintained at a constant temperature of 58 ± 2 °C. The moisture content was maintained at 50-55% while the oxygen

level of the reactor was not less than 6% throughout the testing period.

During the incubation period, the synthetic waste and the film samples were transformed into compost by a composting process. The mixture in the composting reactor was regularly turned by hand and the moisture content was maintained by adding deionized water as required. A schematic diagram of the disintegration test system and a photograph of the composting reactors are shown in Figures 3 and 4 respectively.



**Figure 3.** Schematic diagram of the disintegration test system.



**Figure 4.** Photograph showing the composting reactors in the oil bath.

**3.3 Decrease in Volatile-Solids Content**

In each reactor, the percentage decrease *R* in the total volatile-solids content between the initial synthetic waste and the compost obtained at the end of the test was calculated from equation (1). According to the ISO 20200:2012 standard test method, the test

results are only valid for values of *R* ≥ 30%.

$$R = \frac{[m_i \times (DM)_i \times (VS)_i] - [m_f \times (DM)_f \times (VS)_f]}{[m_i \times (DM)_i \times (VS)_i]} \times 100 \quad (1)$$

where:

*m<sub>i</sub>* is the initial mass of the wet synthetic waste introduced into the reactor

(*DM*)<sub>*i*</sub> is the initial dry mass of the synthetic waste, expressed as a percentage divided by 100

(*VS*)<sub>*i*</sub> is the initial volatile-solids content of the synthetic waste, expressed as a percentage divided by 100

*m<sub>f</sub>* is the final mass of the compost  
 (*DM*)<sub>*f*</sub> is the final dry mass of the compost, expressed as a percentage divided by 100

(*VS*)<sub>*f*</sub> is the final volatile-solids content of the compost, expressed as a percentage divided by 100

**3.4 Degree of Disintegration**

The degree of disintegration *D* of each film sample, expressed as a percentage, was calculated from equation (2). Any material recovered from the sieving procedure which was ≥ 2 mm in size was considered to be non-disintegrated material. Conversely, any material which passed through the 2 mm sieve was considered to have disintegrated. The degree of disintegration was calculated separately for each of the 3 reactors and the results averaged. According to the ISO 20200:2012 standard test method, the test results are only valid if the values of *D* for the duplicate tests differ by not more than 10%.

$$D = \frac{m_i - m_r}{m_i} \times 100 \quad (2)$$

where:

*D* is the degree of disintegration

*m<sub>i</sub>* is the initial dry mass of the tested material

$m_r$  is the dry mass of the residual tested material recovered by sieving

## 4. RESULTS AND DISCUSSION

### 4.1 Synthetic Solid Waste and Test Material Properties

The various properties of the synthetic waste, both before and after testing, and the initial properties of the film test materials were analyzed according to standard test methods. The results are shown in Tables 1 and 2 respectively.

**Table 1.** Properties of the synthetic waste before and after sample testing.

Property	Synthetic Waste			
	Before testing	After testing		
		PLL	PLL/TPU	PLL/TPU/PLLCL
pH	5.63	7.14	7.16	7.27
Moisture content; MC (%)	55.71	53.50	43.15	48.85
Total dry solid; TS (%)	44.29	46.50	56.85	51.15
Volatile solid; VS (% on TS)	92.90	88.12	87.18	87.75

**Table 2.** Properties of the film test materials.

Property	PLL	PLL/TPU	PLL/TPU/PLLCL
Moisture content; MC (%)	0.54	1.14	1.52
Total dry solid; TS (%)	99.46	98.86	98.48
Volatile solid; VS (% on TS)	99.98	99.97	99.95
Total organic carbon (% on TS)	46.46	52.68	52.20

During the composting process, it was possible to detect a succession of specific odours. Within the first two or three days, the synthetic waste had an acidic odour which gradually decayed into an ammoniacal odour from day 5 to day 10 and lasted about 10 days. From Table 1, the pH of the synthetic waste, which was initially slightly acidic, increased after testing until it became approximately neutral. After composting for 84 days, the volatile solid content (%) decreased slightly for all test samples.

### 4.2 Disintegration Testing

The disintegration of the PLL, PLL/TPU and PLL/TPU/PLLCL test films was monitored at regular intervals throughout the 84 days of the test period. During the test, the synthetic waste became transformed into compost which could be observed when

turning over the composting matter and adding water as required. The visual appearance of the composting matter changed during testing. Initially, the synthetic waste was light yellow in colour due to the high sawdust concentration but gradually turned brown due to mycelium growth on the composting matter. This colour change occurred during the first week for the synthetic waste containing the PLL film sample but took longer, about 4 weeks, for that containing the PLL/TPU and PLL/TPU/PLLCL film samples.

The appearances of the film samples at various times during the testing period are shown in Figures 5-7 for the PLL, PLL/TPU and PLL/TPU/PLLCL films respectively. When comparing these results, the main features to note are:

- From visual inspection, the rate of disintegration into fragments smaller than 2 mm appeared to be in the order: PLL/TPU/PLLCL > PLL > PLL/TPU.

- For all samples, there appeared to be an induction period at the start of the test during which little change occurred. This induction period varied between 7 days for the PLL (Figure 5), 14 days for the PLL/TPU (Figure 6) and 14 days for the PLL/TPU/PLLCL (Figure 7).

- After the induction period, disintegration was fast and appeared to be complete within 30 days in all cases.

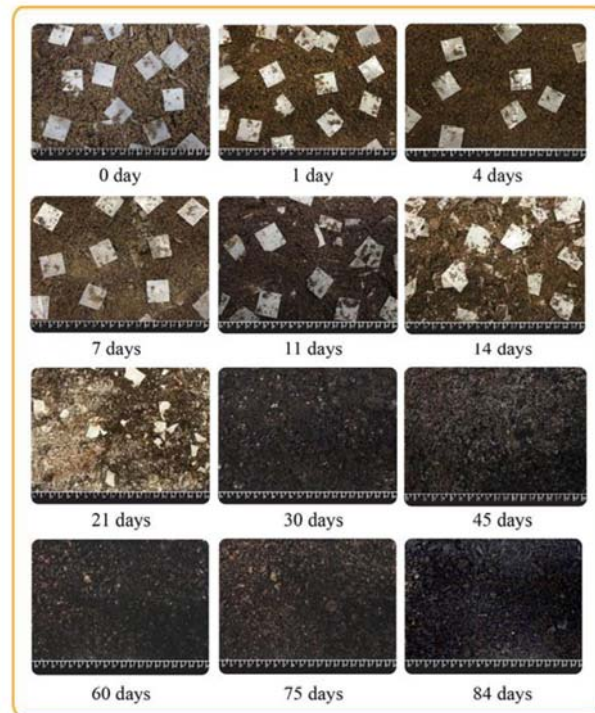
- In the final stage of disintegration, some small white particles of the films were still visible up to about 30 days, as shown

in Figures 5-7. However, after 45 days, no white particles could be detected through visual inspection.

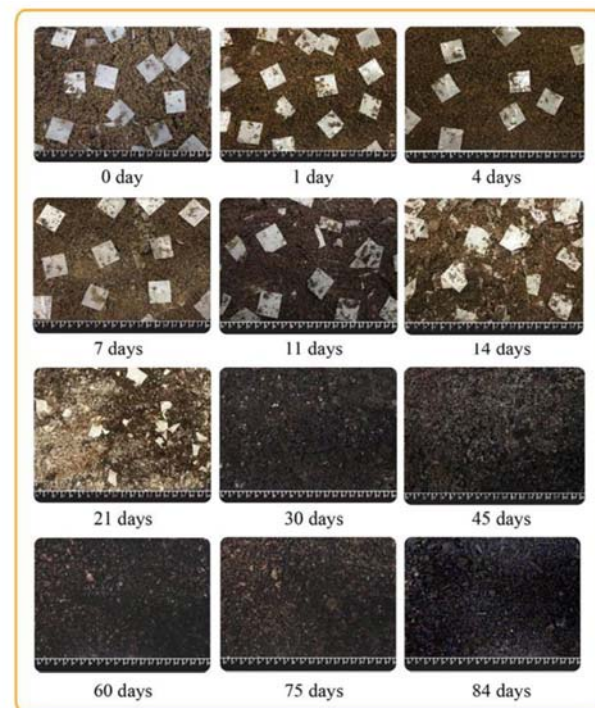
These results are therefore seen to be consistent with the well-documented two-stage bulk erosion process for the environmental biodegradation of PLL involving (1) simple hydrolysis to form low molecular weight oligomers and lactic acid followed by (2) microbial degradation of the fragmented residues by the microorganisms present [1-3, 21-23]. Whereas the first stage is a slow process during which hydration, hydrolysis and pore formation occur, the second stage microbial digestion of the hydrolysis products occurs much faster.



**Figure 5.** Appearance of the PLL film during the disintegration test. (Scale: 1 large division = 1 mm, 1 small division = 0.5 mm).



**Figure 6.** Appearance of the PLL/TPU film during the disintegration test. (Scale: 1 large division = 1 mm, 1 small division = 0.5 mm).



**Figure 7.** Appearance of the PLL/TPU/PLLCL film during the disintegration test. (Scale: 1 large division = 1 mm, 1 small division = 0.5 mm).

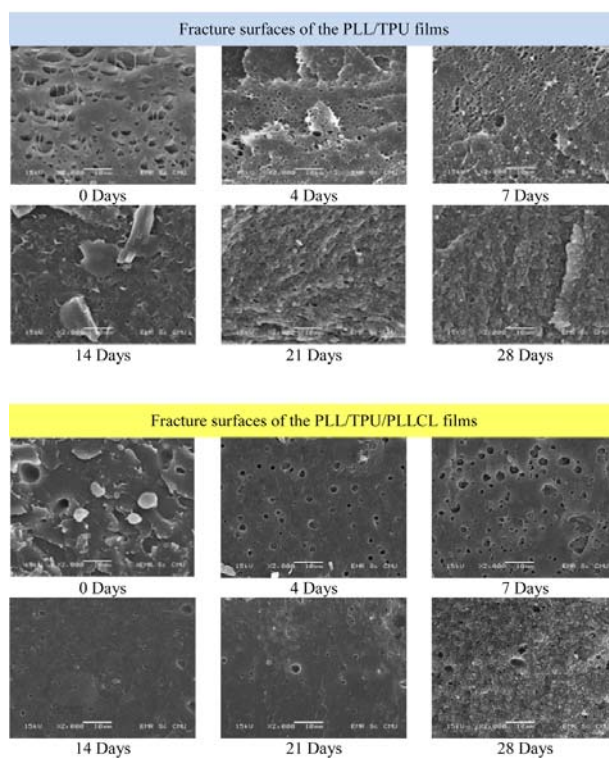


### 4.3 Matrix Morphology

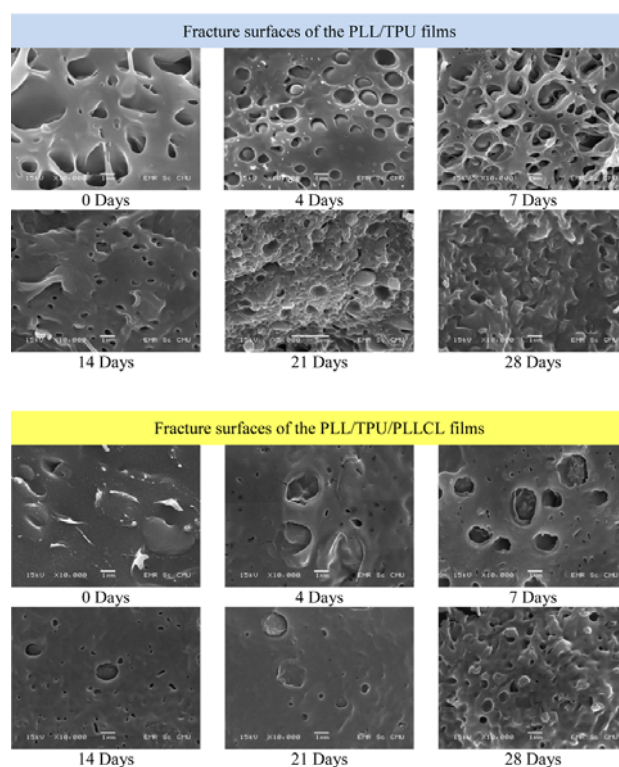
Changes in the matrix morphology of the PLL/TPU and PLL/TPU/PLLCL blended films during disintegration testing are shown in Figures 8 and 9 from SEM images taken of cross-sectional fracture surfaces at magnifications of  $\times 2,000$  and  $\times 10,000$  respectively. The phase separation of the main PLL matrix and the dispersed TPU domains can be clearly seen, especially at the higher magnification of  $\times 10,000$  in Figure 9. As reported previously [9], the outlines of the TPU domains are more diffuse in the case of the PLL/TPU/PLLCL blend due to the compatibilizing effect of the PLLCL at the PLL-TPU interfaces. In addition to the embedded TPU domains, there also appears to be a certain amount of internal pore structure.

Correlating these SEM images with the

previous optical images in Figures 6 and 7 is complicated by the fact that (a) the levels of magnification are much different and (b) the thin film samples have a high surface area-to-bulk ratio which means that surface erosion is likely to be just as important if not more so than bulk erosion. Consequently, disintegration may occur as a result of surface erosion, as shown in Figures 6 and 7, without the SEM images in Figures 8 and 9 showing similarly clear signs of bulk erosion. However, what the SEM images do show is that (1) changes in the fracture surfaces tend to occur at longer times than film fragmentation and (2) the addition of the PLLCL compatibilizer in the PLL/TPU/PLLCL 3-component blend improves the interfacial adhesion between the TPU domains and the surrounding PLL matrix and decreases porosity.



**Figure 8.** SEM images of the cross-sectional fracture surfaces of the PLL/TPU and PLL/TPU/PLLCL films at a magnification of  $\times 2,000$ . (Scale bar =  $10\ \mu\text{m}$ ).



**Figure 9.** SEM images of the cross-sectional fracture surfaces of the PLL/TPU and PLL/TPU/PLLCL films at a magnification of  $\times 10,000$ . (Scale bar = 1  $\mu\text{m}$ ).

#### 4.4 Decrease in Volatile-Solids Content

The weight changes of the synthetic waste mixtures containing the PLL, PLL/TPU and PLL/TPU/PLLCL film samples were as shown in Table 3. For each reactor, the percentage decrease in volatile-solids content ( $R$ ) from the initial synthetic waste to the compost obtained at the end was

calculated using the previous equation (1). The main purpose of this determination was not so much to compare the film samples as to validate the test in accordance with the requirements of the ISO 20200:2012 standard test method. Since the  $R$  values in Table 3 are all  $\geq 30\%$ , the test results can be considered to be valid.

**Table 3.** Decreases in the volatile-solids contents,  $R$ , of the synthetic waste mixtures containing the PLL, PLL/TPU and PLL/TPU/PLLCL film samples.

Film Sample	Characteristics of the synthetic waste						$R^*$ (%)
	Before testing			After testing			
	$m_i$ (g)	$(DM)_i$ (%)	$(VS)_i$ (%)	$m_f$ (g)	$(DM)_f$ (%)	$(VS)_f$ (%)	
PLL	3000.55	44.26	92.90	1570.92	46.50	88.12	48.11
PLL/TPU	3000.51	44.29	92.90	1398.73	56.85	87.18	43.85
PLL/TPU/PLLCL	3000.46	44.29	92.90	1547.72	51.15	87.75	43.73

\* Average values from 3 determinations; range of variation between the duplicate tests  $< 5\%$

#### 4.5 Degree of Disintegration

The degree of disintegration,  $D$ , was calculated using the previous equation (2) from the weight of the film fragments (size  $\geq 2$  mm) which remained after sieving at the end of the 84-day test period. Any fragments of size  $< 2$  mm which passed through the 2 mm sieve were considered to be disintegrated material. As the results in Table 4 show, no residual material was

recovered for any of the film samples after 84 days and so the value of  $D$  in each case was 100%. Moreover, no residual material, either in the form of fragments or particles, could be detected by visual inspection from 45 days onwards. Since  $D$  for the duplicate determinations for each sample did not vary by more than 10%, these results can be considered to be valid in accordance with ISO 20200:2012.

**Table 4.** Degree of disintegration,  $D$ , of the PLL, PLL/TPU and PLL/TPU/PLLCL film samples after 84 days of composting.

Film Sample	Mass of film sample (g)		$D^*$ (%)
	Before testing	After testing	
	$m_i$	$m_r$	
PLL	59.81	n/d	100.00
PLL/TPU	59.89	n/d	100.00
PLL/TPU/PLLCL	59.98	n/d	100.00

\* 100% for all 3 determinations for each sample and for both duplicate tests

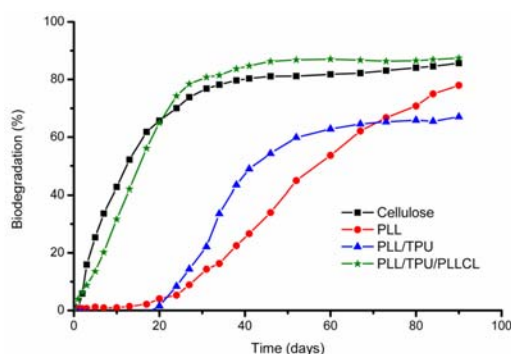
n/d = no residual material detected

#### 5. CONCLUSIONS

On the basis of these results, it can be concluded that, in common with PLL alone, both the PLL/TPU and PLL/TPU/PLLCL blended films underwent complete 100% disintegration in accordance with the ISO 20200:2012 standard test conditions. Therefore, the additions of the more hydrophobic TPU and PLLCL components did not appear to detract from the biodegradability of PLL. Furthermore, according to ISO 17088:2008 (Specifications for compostable plastics), the film samples can also be considered to be compostable materials since  $< 10\%$  of the films added at the start remained present in the  $\geq 2$  mm fraction after 84 days of the disintegration test [24].

It is interesting to compare these disintegration test results with the biodegradability test results previously

reported for the same PLL/TPU 90/10 and PLL/TPU/PLLCL 90/10/10 blends [9]. In contrast, the biodegradation-time profiles shown in Figure 10 were obtained from powdered samples of particle size  $< 500 \mu\text{m}$  and tested according to the ISO 14855-1:2005 standard test procedure for determining biodegradability by measuring the amount of  $\text{CO}_2$  gas evolved [25]. Since the final % biodegradations in Figure 10 for the PLL, PLL/TPU and PLL/TPU/PLLCL samples were all  $> 70\%$  relative to cellulose as a reference, they could be considered as "biodegradable plastics". The faster rate of biodegradation of the PLL/TPU/PLLCL was attributed to the effects of the PLLCL in lowering the glass transition temperature  $T_g$  and % crystallinity of the blend, thereby facilitating the diffusion of water through the matrix and accelerating the rate of hydrolysis.



**Figure 10.** Biodegradation-time profiles obtained from powdered samples under controlled composting conditions at  $58 \pm 2$  °C [9].

Finally, even though the disintegration and biodegradation tests are quite different in the ways that the data is obtained, combining the results can be used to conclude whether or not the tested materials will be sufficiently compostable under known conditions of biological waste treatment. From the laboratory-scale tests carried out here, it can be concluded that the PLL/TPU and PLL/TPU/PLLCL blended films meet the criteria for disintegration and compostability and therefore have potential for use as biodegradable packaging materials.

#### ACKNOWLEDGEMENTS

The authors wish to thank the Lubrizol Corporation for provision of the thermoplastic polyurethane and the National Research University Project under Thailand's Office of the Higher Education Commission for financial support.

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