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APPENDICES

Chapter III: Supporting information S1

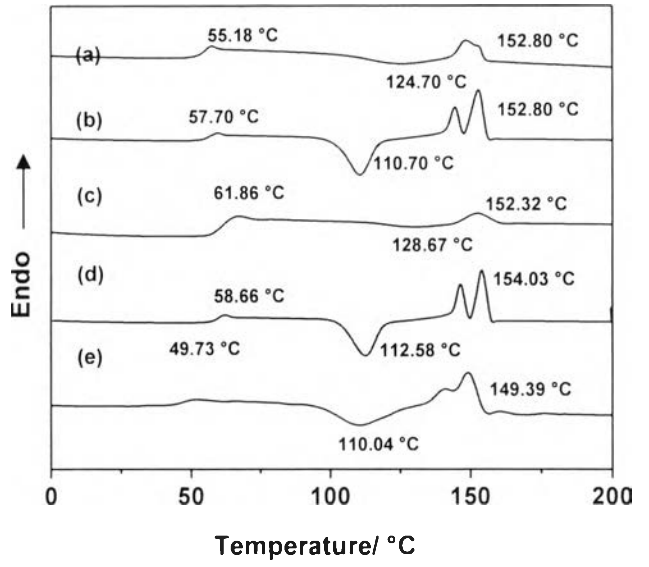


Figure S1 DSC thermograms of neat PLA (a) and PLA/PLA or silane-starch 10wt%: starch (b), GP-starch (c), AP-starch (d), and CP-starch (e).

Chapter III: Supporting information S2

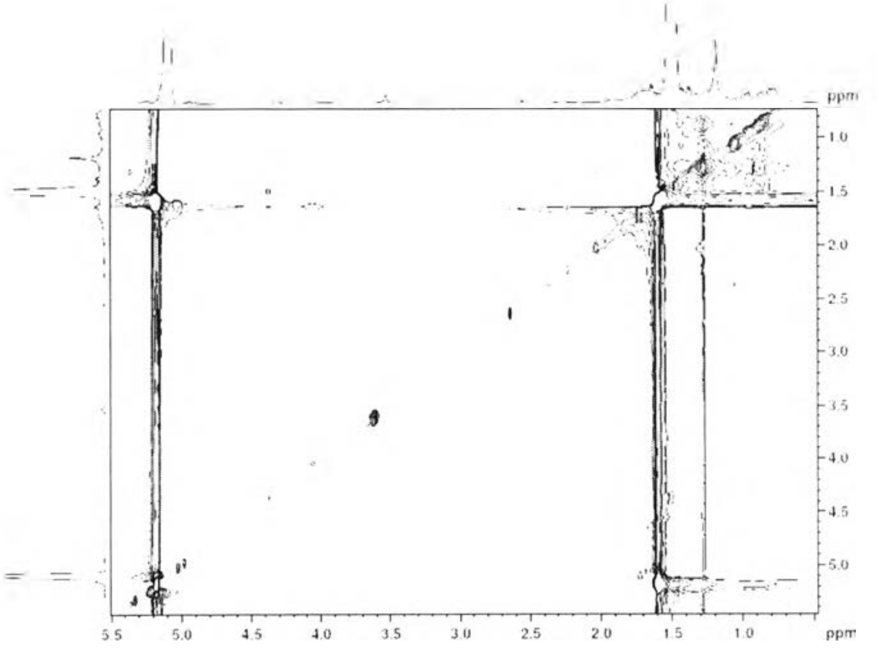


Figure S2 [^{1}H,^{1}H]-TOCSY-NMR spectra of PLA/starch in $CDCl_3$.

Chapter III: Supporting information S3

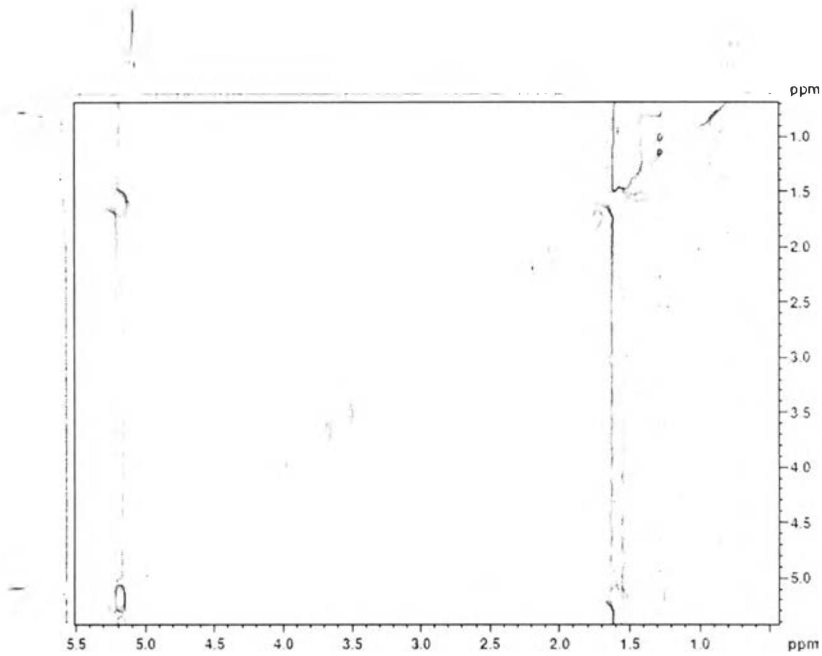


Figure S3 [^{1}H,^{1}H]-TOCSY-NMR spectra of PLA/GP-starch in $CDCl_3$.

Chapter III: Supporting information S4

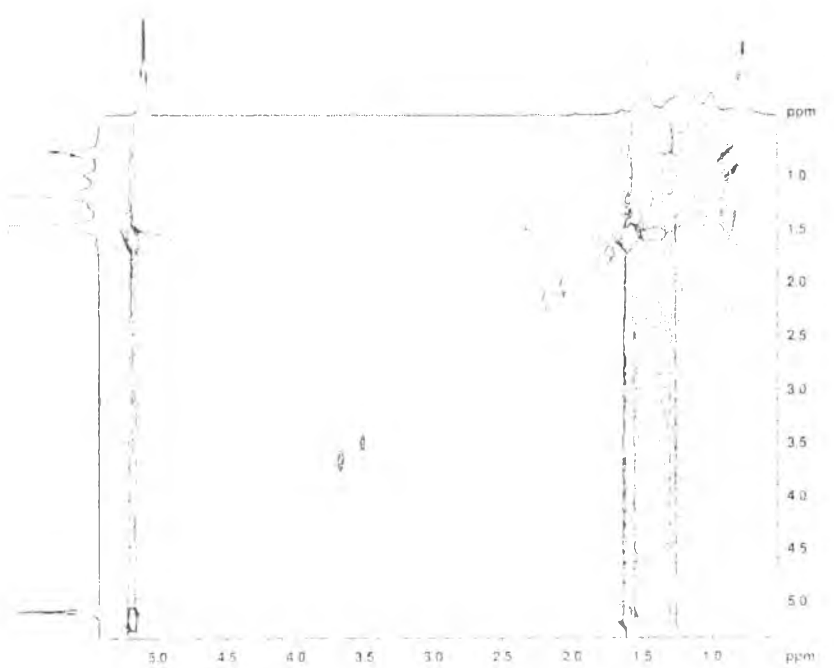
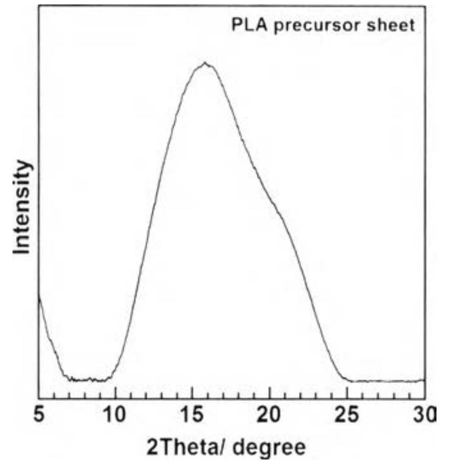


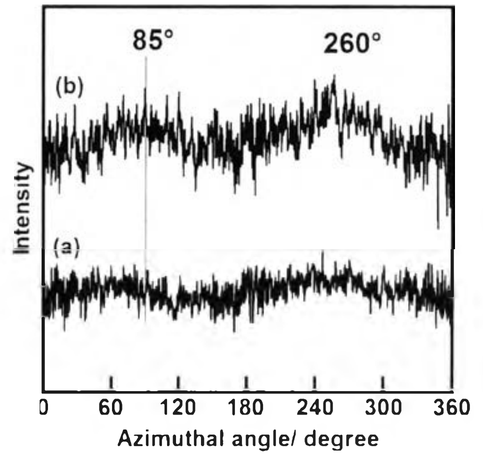
Figure S4 [^{1}H,^{1}H]-TOCSY-NMR spectra of PLA/AP-starch in $CDCl_3$.

Chapter IV: Supporting information S1



Supporting information S1 WAXD intensity profile of PLA precursor sheet at room temperature obtained from azimuthal integration of 2D-WAXD pattern in Fig. 5(a).

Chapter IV: Supporting information S2



Supporting information S2 Azimuthal scans of (200/110) planes converted from 2D-WAXD patterns (through view) of PLA precursor sheet (a) at 25 °C and (b) after annealing at 140 °C for 10 h.

The preferential MD orientation pattern of 3×3-BOPLA could be the chain orientation history initiated from the step of the cast-film extrusion of PLA precursor sheet as evidenced in Supporting information S2. Ideally, the perfect in-plane orientation along MD is found at azimuthal angles of 90° and 270° from 2D-WAXD pattern of through view. In this study, the azimuthal scan of PLA precursor sheet presents the partial orientation of (200/110) diffraction at azimuthal angle of ~85° and 260°. Their intensities became significant by annealing at 140°C for 10 h, insisting the initial MD orientation in PLA precursor sheet.

Chapter IV: Supporting information S3

Supporting information S3 Herman's orientation function (*f*) estimated from (200/110) and (203) diffraction planes for PLA precursor sheet and BOPLA films at various draw ratios and stretching rates.

Sample	**Herman's orientation function (*f*)**		
	200/110 planes		203 plane
	f for φ = 0°, (parallel to MD)	*f* for φ = 90° (perpendicular to MD)	*f* for φ = 60° to MD
PLA precursor sheet	0.08 ± 0.04	N/A	N/A
3x3-BOPLA, 3 mm/s	0.74 ± 0.01	N/A	0.33 ± 0.03
3x3-BOPLA, 16 mm/s	0.65 ± 0.01	N/A	0.50 ± 0.02
3x3-BOPLA, 37 mm/s	0.67 ± 0.02	N/A	0.71 ± 0.02
3x3-BOPLA, 75 mm/s	0.66 ± 0.02	N/A	0.76 ± 0.02
5x5-BOPLA, 3 mm/s	0.83 ± 0.01	0.75 ± 0.02	0.70 ± 0.03
5x5-BOPLA, 16 mm/s	0.72 ± 0.01	0.73 ± 0.01	0.57 ± 0.04
5x5-BOPLA, 37 mm/s	0.54 ± 0.03	0.57 ± 0.01	0.34 ± 0.04
5x5-BOPLA, 75 mm/s	0.31 ± 0.02	0.33 ± 0.02	0.11 ± 0.03

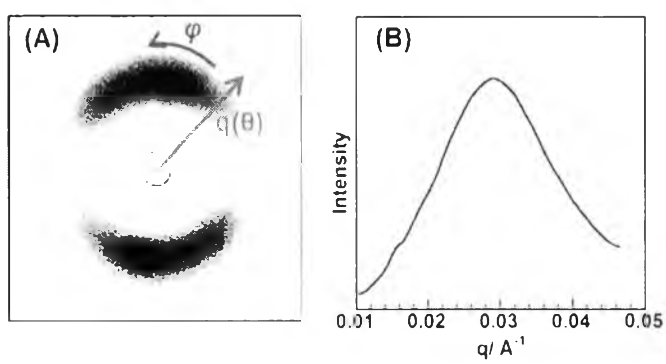
Herman's orientation function (*f*) was applied to describe the degree of orientation of the chain axis relative to the other axis of interest (*i.e.* MD and TD) as defined: $f = 3\langle\cos^2\phi - 1\rangle/2$ where $\cos^2\phi$ represents a square averaged cosine of angle ϕ between the crystallographic axis and fiber-axis. In this study, the calculation of *f* was simplified by simply calculating the misorientation appearing in the azimuthal scan (φ). The value of *f* can then be calculated by

$f = (180\,° - \Delta\varphi_{1/2})/180°$ where $\Delta\varphi_{1/2}$ represents the half width of the azimuthally scanned profiles for (200/110) and (203) diffraction planes. The f value is 1 for polymer chains oriented parallel to MD and/or TD whereas the f value is close to 0 for isotropic orientation.

Chapter IV: Supporting information S4

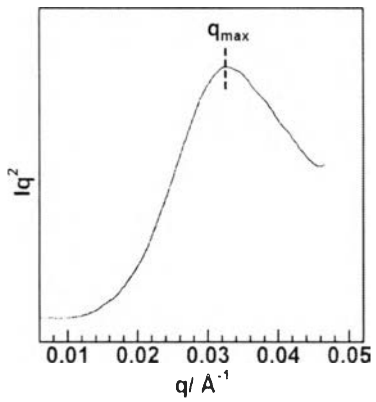
Calculation of long period, size and thickness of stacked lamellae for BOPLA films

The 2D-SAXS patterns in Figs. 5 and 6 were azimuthally integrated to collect the 1D-SAXS data (azimuthal integrated SAXS intensity vs scattering vector (q)) as demonstrated in Supporting information S4.1 (B).



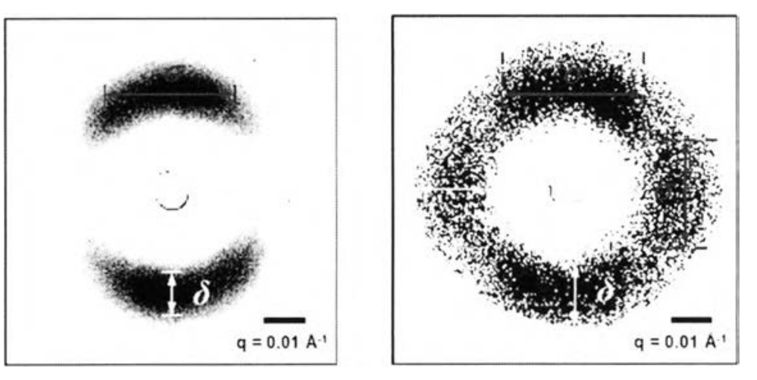
Supporting information S4.1 (A) 2D-SAXS of 3×3-BOPLA by 3 mm/s stretching rate with azimuthal integration directions and (B) plot of its SAXS intensity as a function of the magnitude of the q, calculated from scattering angle (2θ): $q = 4\pi(\sin\theta)/\lambda$.

The azimuthal integrated SAXS intensity (I) was multiplied by q^2 (Iq^2) to fit Lorentzian correction for determining long period of the stacked lamellae. The long period (Fig. 7a) was obtained from the maximum q value (q_{max}) by using $2\pi/q_{max}$ (Supporting information S4.2).



Supporting information S4.2 Plot of Iq^2 vs q of 3×3-BOPLA by 3 mm/s stretching rate.

The size (δ^{-1}) and thickness (σ^{-1}) of stacked lamellae for BOPLA films were calculated directly from 2D-SAXS patterns. The inversed size (δ) and thickness (σ) of the stacked lamellae were measured in q-value scale as examples in Supporting information S4.3. The δ and σ values were converted to length scale as follows: $\delta^{-1} = 2\pi/\delta$ and $\sigma^{-1} = 2\pi/\sigma$ for size and thickness of stacked lamellae, respectively, as presented in Fig. 7b-c.



Supporting information S4.3 Inversed values of size and thickness of stacked lamellae measured directly from 2D-SAXS pattern of (A) 3×3-BOPLA, 3 mm/s and (B) 5×5-BOPLA, 75 mm/s.

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Publications:

1. Jariyasakoolroj, P.; and Chirachanchai, S. Silane Modified Starch for Compatible Reactive Blend with Poly(Lactic Acid). Carbohydrate Polymers 2014, 106, 255–263.
2. Jariyasakoolroj, P.; Tashiro, K.; Hai, W.; Yamamoto, H.; Chinsirikul, W.; Kerddonfag, N.; and Chirachanchai, S. Isotropically Small Crystalline Lamellae Induced by High Biaxial-stretching Rate as a Key Microstructure for Super-tough Poly(L-lactic Acid) Film. Polymer, in press.
3. Jariyasakoolroj, P.; Supthanyakul, R.; Laobuthee, A.; Lertworasirikul, A.; Yoksan, R.; Phongtamrug, S.; and Chirachanchai, S. Potential Biodegradable Mulch Film from Poly(Lactic Acid)/Silane-modified Thermoplastic Starch Blend. Polymer Degradation and Stability, to be submitted.
4. Jariyasakoolroj, P.; Tashiro, K.; Chinsirikul, W.; Kerddonfag, N.; and Chirachanchai, S. Strengthening Hydrogen Bonds Between Poly(Lactic Acid) and Thermoplastic Starch *via* Biaxial-stretching for Retardant Retrogradation. (In Preparation)

5. Jariyasakoolroj, P.; Tashiro, K.; and Chirachanchai, S. Poly(L-lactic Acid)-Poly(Ethylene Glycol)-Poly(L-lactic Acid) Triblock Copolymer and its Performance in PLA-based Film. (In Preparation)

Proceedings:

1. Jariyasakoolroj, P.; and Chirachanchai, S. (2010, October 7-8) Compatibility Enhancement of Modified-thermoplastic Starch for Blending with Poly(Lactic Acid). Proceeding of the First Polymer Conference of Thailand (PCT-1), Bangkok, Thailand.
2. Jariyasakoolroj, P.; and Chirachanchai, S. (2012, August 28-31) Poly(Lactic Acid)-b-Polyethylene Glycol Block Copolymer (PLA-b-PEG) and its Performance in PLA Film. Proceeding of Asian Workshop on Polymer Processing 2012 (AWPP 2012), Kyoto, Japan.

Presentations:

1. Jariyasakoolroj, P.; and Chirachanchai, S. (2009, September 2) Starch Modification for Nucleation with Compatibility of Poly(lactic acid)/Starch Blends. Paper presented at the First Thai-Japan Bioplastics and Biobased Materials Symposium (AIST-NIA Joint Symposium), International Conference and Exhibition InnoBioplast 2009, Bangkok, Thailand. (Poster presentation)
2. Jariyasakoolroj, P.; and Chirachanchai, S. (2010, March 21-25) Role of Silane Coupling Agents on Nucleation and Compatibilization of Poly(Lactic Acid)/Starch Blends. Paper presented at the 239th ACS National Meeting, San Francisco, California. (Oral Presentation)
3. Jariyasakoolroj, P.; and Chirachanchai, S. (2010, September 9-11) Enhancement of Miscibility between Poly(Lactic Acid) and Starch Surface Modified with Silane Coupling Agents. Paper presented at InnoBioPlast Conference and Exhibition 2010, Bangkok, Thailand. (Poster presentation, Professional Award)
4. Jariyasakoolroj, P.; and Chirachanchai, S. (2010, October 7-8) Compatibility Enhancement of Modified-thermoplastic Starch for Blending with

Poly(Lactic Acid). Paper presented at The First Polymer Conference of Thailand (PCT-1), Bangkok, Thailand. (Poster presentation)

5. Jariyasakoolroj, P.; and Chirachanchai, S. (2011, April 1-3) PLA-Starch Blend: A Focus on Compatibility via Coupling Reaction. Paper presented at RGJ-Ph.D. Congress XII, Pattaya, Chonburi, Thailand. (Both Oral and Poster presentations)
6. Jariyasakoolroj, P.; and Chirachanchai, S. (2011, May 29 - June 3) Compatibilization Improvement of Poly(Lactic Acid)/Starch Blends via Coupling Reaction. Paper presented at Europolymer conference (EUPOC 2011) "Biobased Polymers and Related Biomaterials", Gargnano, Italy. (Poster presentation, The Best Poster Award)
7. Jariyasakoolroj, P.; and Chirachanchai, S. (2012, August 28-31) Poly(Lactic Acid)-b-Polyethylene Glycol Block Copolymer (PLA-b-PEG) and its Performance in PLA Film. Paper presented at Asian Workshop on Polymer Processing (AWPP 2012), Kyoto, Japan. (Oral presentation)
8. Jariyasakoolroj, P.; Tashiro, K.; Chinsirikul, W.; Kerddonfag, N.; and Chirachanchai, S. (2013, January 24-26) Study on Crystallization Behavior of Biaxially Stretched Poly(Lactic Acid)/Modified Thermoplastic Starch Films and their Consequent Mechanical Properties. Paper presented at The 4th International Conference and Exhibition on Bioplastics and Bio-based Materials, InnoBioPlast 2013: Advances in Bioplastics Industry and Opportunities in Asia (InnoBioPlast, 2013), Bangkok, Thailand. (Poster presentation)
9. Jariyasakoolroj, P.; Tashiro, K.; Chinsirikul, W.; Kerddonfag, N.; and Chirachanchai, S. (2014, March 20-21) Relationship of Microstructure Orientation and Toughness Improvement of Poly(L-lactic Acid) Film through Equal-biaxial Stretching. Paper presented at The 4th Polymer Conference of Thailand (PCT-4), Bangkok, Thailand. (Poster presentation)
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Viewpoint. Paper presented at IUPAC World Polymer Congress (MACRO 2014), Chaingmai, Thailand. (Oral presentation)

11. Jariyasakoolroj, P.; Tashiro, K.; and Chirachanchai, S. (2014, December 1) Controlling Microstructure of Poly(lactic acid) for Toughness Enhancement by Biaxial-Stretching as a Model Case Study. Paper presented at TRF Seminar Series 107 (Thailand-Malaysia): Green Polymers for Environment and Polymers for Quality of Life, Bangkok, Thailand. (Oral presentation)