Supporting Information

Achieving High Ductile Polylactide Sheets with Inherent Strength via a Compact and Uniform Stress Conduction Network

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Figure S1. General structure of the styrene-acrylic multi-functional oligomeric chain extenders (CE). Where $R_1$-$R_5$ are H, CH$_3$, a higher alkyl group, or combinations of them; $R_6$ is an alkyl group, and $x$, $y$ and $z$ are each between 1
and 20.

Table S1. Composition, Extrusion Protocol, and Codification of the Prepared Formulations for different PLA.

<table>
<thead>
<tr>
<th>specimen code</th>
<th>matrix</th>
<th>extrusion condition</th>
<th>Number of LMEs</th>
<th>Draw ratio</th>
<th>ice-water cooling</th>
</tr>
</thead>
<tbody>
<tr>
<td>L-9-1</td>
<td>Linear PLA</td>
<td></td>
<td>9</td>
<td>1</td>
<td>-</td>
</tr>
<tr>
<td>L-9-2.16</td>
<td>Linear PLA</td>
<td></td>
<td>9</td>
<td>2.16</td>
<td>-</td>
</tr>
<tr>
<td>L-9-2.16-i</td>
<td>Linear PLA</td>
<td></td>
<td>9</td>
<td>2.16</td>
<td>√</td>
</tr>
<tr>
<td>B-0-1</td>
<td>LCBPLA</td>
<td></td>
<td>0</td>
<td>1</td>
<td>-</td>
</tr>
<tr>
<td>B-0-2.16</td>
<td>LCBPLA</td>
<td></td>
<td>0</td>
<td>2.16</td>
<td>-</td>
</tr>
<tr>
<td>B-0-2.16-i</td>
<td>LCBPLA</td>
<td></td>
<td>0</td>
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<td>√</td>
</tr>
</tbody>
</table>
Hermans’ orientation factor \(f_H\) can estimate the orientation of PLA crystals. For a set of \(hkl\) planes, the orientation function of Herman is defined as follows:\(^1\)

\[
f_H = \frac{3\langle \cos^2 \varphi \rangle_{kh} - 1}{2}
\]

(S1)

and orientation factor \(\cos^2 \varphi\) is defined as follows:

\[
\langle \cos^2 \varphi \rangle_{lkh} = \frac{\int_0^{\pi/2} I(\varphi) \cos^2 \varphi \, \sin \varphi \, d\varphi}{\int_0^{\pi/2} I(\varphi) \, \sin \varphi \, d\varphi}
\]

(S2)

The above \(\varphi\) is the azimuthal angle and \(I(\varphi)\) is the scattered intensity along the angle \(\varphi\). The azimuthal intensity distribution \(I(\varphi)\) was analyzed at \(2\theta = 16.7^\circ\), in which the peak represents the \((200, 110)\), \(\alpha\) reflections in different PLA.

*Figure S2.* DSC first heating curve for LCBPLA prepared by internal mixing. \(T_c\) increases as the amount of CE increases, implying the limited chain mobility in LCBPLA. The final \(X_c\) of LCBPLA shows a slight increase than linear PLA owing to the nucleation sites provided by LCBPLA backbone.
Figure S3. SEM images showing the network-like precursor of extruded PLA samples indirectly from the morphology of crystal network (etched at 25 °C for 12 h). Severe collapse appeared due to low crystallization degree of PLA samples.

Figure S4. Illustration of the location of skin layers and core layers of different PLA samples. (a) is one inset part of (A).
**Figure S5.** SEM images showing the network precursor of MSE LCBPLA without post-drawing indirectly from the morphology of crystal network by high-temperature etching (50 °C for 1 h).
**Figure S6.** SEM images showing the network-like precursor of MSE LCBPLA with post-drawing and ice-water cooling indirectly from the morphology of crystal network by high-temperature etching (30 °C for 4 h) was taken. Lower etching temperature was chosen due to the catastrophic deformation of the network-like precursor during tensile tests.

![SEM images showing the network-like precursor of MSE LCBPLA](image1)

**Figure S7.** Typical stress-strain curves of B-9-2.16-i.

![Typical stress-strain curves of B-9-2.16-i](image2)

**Figure S8.** Comparation of mechanical performance of PLA with different fabrication methods. (1) PLA-MIECO10,2 (2) P-S40T20,3 (3) 5% eSEPS51-PLA,4 (4) 10
BF/15TA-ESO/PLA, (5) PLA+ATBC+10T-AV, (6) PLA/TPU (70/30), (7) PLA-20A/ax8900MD, (8) 3.5 MPa CO2 treatment, (9) PLA/GPR/HDI (95/5/0.8), (10) PLLA/PDLA/15E-MA-GMA, (11) PLA/HDAPA40, (12) cPLA80PBSA20, (13) TPLA, (14) PLA/PCL (90 wt%:10 wt%)/8 phr EMA-GMA, (15) PLA/GNR, (16) 10.0% CNC-rD-PDLA, (17) PLA/20% NR-GMA, (18) 10% PHB-di-rub, (19) D80/10/10.
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