Tensile Fracture Morphology/Properties Correlation of High-density/Linear Low-density Polyethylene Blends

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ABSTRACT

Morphologically distinct binary polymer blends have been prepared by melt mixing of high-density polyethylene (HDPE) and various linear low-density polyethylene (LLDPEs) for the entire range of blend composition under identical processing conditions. The morphology of the tensile fracture surfaces of the parent polymers and their blends are quite interesting and show good correlation with thermal and mechanical properties. The HDPE forms linear and interpenetrating fibrils with large interfibrillar separation, whereas, octene containing LLDPE (OLLDPE) with almost equal number of branching to that of HDPE shows nicely formed twisted fibrils. On the other hand, pentene containing LLDPE ( PLLDPE) manifests a straight fibrillar morphology with well defined boundary comprising many thin fibrils with alternative thick and thin regimes and perfection, whilst butene containing LLDPE (BLLDPE) shows thick comparatively smooth and well defined and imperfect boundary of tensile fracture. The blends morphology is quite distinct to that of parent polymers. The physical properties, melting and crystallization behaviour show good correlation to the fineness, twisting and discontinuity of the fibrils.

Key Words: HDPE/LLDPE blend, tensile fracture morphology, mechanical properties, thermal properties, crystallization and melting

INTRODUCTION

The morphology of polymers and their blends has strong influence on the various properties [1–4]. The morphology could be engineered by varying the processing conditions, post-processing treatment and the thermal history, however the easiest way to negotiate morphology is blending. This field of study of interaction morphology is very important specifically for the case of polyethylene (PE) due to the availability of number of grades with different types, content and distribution sequence of number

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and length of branching co-monomer. The linear low-density polyethylene (LLDPE) with the variation of types, content and distribution sequence of co-monomer is available with wide range of properties. Their good heat sealing properties and their film forming capabilities are of utmost importance characteristics, which can be incorporated to HDPE which lacks these properties but has superior mechanical properties.

In this paper, different tensile fractured morphology of blends of HDPE with three different types of LLDPE viz. octene containing LLDPE (OLLDPE), pentene containing LLDPE (PLLDPE) and butene containing LLDPE (BLLDPE), are presented. An effort is also made to correlate morphology to the thermal properties of these blends. These blends were characterized by differential scanning calorimetry (DSC), tensile testing and scanning electron microscopy (SEM). It is shown that the mechanical properties and thermal behaviour of HDPE/LLDPE blends, to great extent, depend upon the fineness of the fibrils of the tensile fractured surface.

EXPERIMENTAL

Materials
The HDPE (Hostalene GF 7745F) used for this study was a product of Polyolefine Industries Ltd., Bombay, India. The LLDPEs were procured from market and analyzed for their characteristic properties. The properties of the polymers are listed in Table 1. The number of methyl groups per 100 carbon atoms (i.e., CH₃/100 C) was calculated by IR spectroscopy, according to the method prescribed by Willbrum [5].

Blend Preparation
Premixed dry tumble blended HDPE and LLDPE chips were melt blended in a single screw extruder (Betol 1820) with L/D ratio of 17 and screw speed of 22 rpm. The temperature profile of the extruder was kept as 160 °C at the feed, 200 °C at the compression zone and 210 °C both at the melting zone and at the die end. The extruded strands, so obtained, were cooled in a water bath at 30 °C and granulated subsequently after allowing a maturation time of 8 h.

Injection Moulding
The granulated samples were injection moulded to test specimens on a Windsor injection moulding machine (SP-30) using a temperature profile similar to the one used in the blending extruder. The injection time and cooling period were kept at 3 and 5 s, respectively. The specimens ejected from the mould were allowed to cool down to room temperature under ambient condition.

Measurements
Tensile Testing
The tensile properties were measured at room temperature (23±2 °C) after allowing a maturation time of 24 h in accordance with ASTM D638 on Instron Universal Tensile Tester (4202). The gauge length was kept at 50 mm with a crosshead speed of 500 mm/min. The high crosshead speed facilitated the failure of LLDPE within stipulated time. From the stress-strain curves, following properties (based on

<table>
<thead>
<tr>
<th>Properties</th>
<th>OLLDPE</th>
<th>PLLDPE</th>
<th>BLLDPE</th>
<th>HDPE</th>
</tr>
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<tbody>
<tr>
<td>[n] at 115 °C in decalin (dL/g)</td>
<td>1.647</td>
<td>1.597</td>
<td>1.740</td>
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</tr>
<tr>
<td>Melt flow index (g/10 min)</td>
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<td>0.75</td>
<td>1.00</td>
<td>0.75</td>
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<tr>
<td>Density (g/mL)</td>
<td>0.920</td>
<td>0.920</td>
<td>0.918</td>
<td>0.952</td>
</tr>
<tr>
<td>CH₃/100 carbon</td>
<td>2.53</td>
<td>3.11</td>
<td>3.40</td>
<td>1.81</td>
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<td>123.1</td>
<td>131.0</td>
</tr>
<tr>
<td>Tₜ (°C)</td>
<td>102.2</td>
<td>107.8</td>
<td>103.8</td>
<td>114.2</td>
</tr>
<tr>
<td>ΔHc (J/g)</td>
<td>49.8</td>
<td>39.5</td>
<td>49.2</td>
<td>154.4</td>
</tr>
<tr>
<td>Tensile at yield (MPa)</td>
<td>15.0</td>
<td>10.0</td>
<td>10.4</td>
<td>28.0</td>
</tr>
</tbody>
</table>
average of 5 samples) were calculated: Young's modulus (from the initial slope of the stress-strain curve), yield strength and elongation-at-yield (from the first maxima of the curve), ultimate tensile strength and elongation-at-break (where samples physically failed).

**Scanning Electron Microscopy**
The micrographs of the tensile fractured surfaces were photographed by Cambridge scanning electron microscope (Stereoscan 360). The tips of the fracture surfaces were cut and mounted keeping the surface normal to the electron beam. The samples were silver-coated by vacuum deposition.

**Differential Scanning Calorimetry**
The crystallization exotherms and melting endotherms of samples were recorded on a Perkin-Elmer differential scanning calorimeter (DSC-7) both at cooling and heating rates of 10 °C/min in nitrogen atmosphere. Powdered samples of about 10 mg were used for this study. All the samples were first run through a heating cycle from ambient temperature to 145 °C and then through a cooling cycle keeping 2 min at 145 °C.

The percent crystallinity (X) is calculated by applying the relationship:

\[
X\% = \left( \frac{\Delta H_c}{\Delta H_{100}} \right) \times 100
\]

Where \(\Delta H\) is the heat of crystallization or melting of the sample and \(\Delta H_{100}\) is the heat of crystallization for 100% crystalline PE.

**RESULTS AND DISCUSSION**
The tensile fractographs of distinct fibril-like morphology of various constructions are apparent for LLDPEs (Figure 1a,b,c) and HDPE (Figure 1d). The fibrils of OLLDPE (Figure 1d) possess rod-like appearance with clearly describable imperfections. The average diameter of OLLDPE fibrils is around 5 µm. The interconnected and twisted fibrils with large interfibrillar separation are seen from tensile fracture surface morphology of HDPE (Figure 1d) with an average size of 3 µm, which are not as perfect as that of OLLDPE.

The development of morphology from melt is a complex process of organization of chains to form super-ordered structure. On large tensile deformation the crystallites deform to form fibrillar crystallites. In this process of transformation the amorphous phase also takes part. In earlier communications [6], it has been mentioned that the LLDPEs used in this study differ considerably depending upon the types of co-monomer present, their arrangement in the polyethylene sequence, the degree of branching and pendant groups (i.e., methyl groups). The OLLDPE contains shorter pendant groups among the three and that the PLLDPE a medium (i.e., propyl group) and that the OLLDPE a longest (i.e., hexyl group) pendant group. The degree of branching varies in the order as HDPE< OLLDPE< PLLDPE< BLLDPE with a numerical value of 1.81, 3.36, 3.94 and 4.23, respectively (Table I). These variations strongly determine both the crystalline and amorphous morphology [7–9] of these pure components, which has also been reflected in the tensile fracture micrographs of pure components.

The co-monomer and branches present in the backbone of polyethylene chain act as perturbances in the linear ethylene sequence. These perturbances may be so great (block types copolymer) that this part of chain together with some parts of linear ethylene sequences would become rejected from the crystallites. There is also probability of such rejected linear ethylene sequences to nucleate a separate crystallinity or entrapped in other growing crystallite and form branched interconnected and interpenetrating crystallites [10].

It is obvious that these sequences also limit the crystallites boundary. It may so happen that small perturbing sequence (small co-monomer sequence) or...
isolated co-monomer link in the polyethylene sequence entrapped in crystallites and cause the unit cell to expand [7].

The thermal properties (i.e., both exotherm and endotherm) of parent polymers and blends are shown in Table 2. From Tables 1 and 2, one may appreciate that the tensile strength at yield and heat of crystallization ($\Delta H_c$) of HDPE are 28 MPa and 154.4 J/g, whereas, the respective values of OLLDPE are 15 MPa and 50 J/g and that of PLLDPE are 10 MPa and 39.5 J/g, whilst the BLLDPE has maintained a tensile at yield of 10.4 MPa and heat of crystallization of 49.2 J/g.

The high $\Delta H_c$ of HDPE could be attributed to the higher crystallinity arising out of highly linear polyethylene chains with very little perturbation (i.e., low CH$_3$/1000 C ratio) in the chain, which allows crystallization to a high degree. This higher degree of crystallinity and interlinked crystallites show high tensile strength at yield. The OLLDPE which has relatively higher CH$_3$/100 C value (Table 1) shows lower crystallinity and lower tensile strength at yield [11—13] than HDPE. However, the discrepancy of heat of crystallization and tensile strength between BLLDPE ($\Delta H_c=49.2$ J/g) and PLLDPE ($\Delta H_c=39.5$ J/g) with approximately equal tensile strength (i.e., 10 MPa).

Figure 1. SEM micrographs of tensile fractured surface of unblended polymers: (a) OLLDPE, (b) PLLDPE, (c) BLLDPE, (d) HDPE.
may lie with the difference in interconnectivity of fibrils as is apparent from Figure 1b and c.

Figure 2a,b,c shows the tensile fracture surfaces of HDPE/LLDPE blend containing various LLDPE at 10% blend composition. The lamella-like structure [14] of HDPE/OLLDPE blend at this composition is apparent from Figure 2a. The HDPE/PLLDP shows fibrillar morphology, which is closer to the morphology of the minor component (i.e., as evident from fracture micrographs of LLDPE) to some extent. The morphology of HDPE/BLLDP matches the morphology of HDPE.

The $T_c$ and $\Delta H_c$ for all the blends are close to each other (Table 2) at 10% LLDPE content. The numerical value of $T_c$ and $\Delta H_c$ are around 115.4 °C and 150–154 J/g (about 55% crystallinity), irrespective of the types of LLDPE present in the blend. However, one may appreciate the differences in the heat of melting ($\Delta H_m$) and temperature of melting ($T_m$) of the blends. The HDPE/OLLDPE blend with regular fibrils manifests slightly higher $\Delta H_m$ values (around 142.5 J/g) than HDPE/PLLDP blend with not so clearly formed fibrils (which has 139 J/g).

Similar trend were observed for the values of tensile at yield for the blend containing 10% LLDPE (Table 3). The HDPE/OLLDP blend with less pronounced fibrillar morphology registered yield strength of 23.4 MPa, which is lowest among the blends. The tensile at yield of PLLDP blend at this composition is slightly higher than OLLDP (i.e., 24.7 MPa), whereas, the yield strength of BLLDP is the highest. Though both the blends of LLDPE (pentene and butene containing LLDPE) display similar morphology, however, the absence of fibrillar separation in the latter case may be the reason of displaying higher tensile at yield (25.6 MPa for 10% BLLDP blend).

Figure 3a,b,c presents the tensile fracture micrographs of blends at 25% LLDPE content. In general, all the blends show distinct fibrils of various constructions. The fibrils of blend of OLLLDP at this composition are not sufficiently distinct (Figure 3a). The approximate diameter of fibrils is 5–8 μm. The interlinking fibrils of HDPE/PLLDP at 25% LLDPE

### Table 2. Thermal properties of HDPE/LLDPE blends.

<table>
<thead>
<tr>
<th>Blend</th>
<th>Blending ratio</th>
<th>$T_c$ (°C)</th>
<th>$\Delta H_c$ (J/g)</th>
<th>$T_m$ (°C)</th>
<th>$\Delta H_m$ (J/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDPE/OLLDP</td>
<td>100/00</td>
<td>114.2</td>
<td>154.4</td>
<td>131.0</td>
<td>148.2</td>
</tr>
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<td></td>
<td>90/10</td>
<td>115.4</td>
<td>153.8</td>
<td>130.2</td>
<td>142.5</td>
</tr>
<tr>
<td></td>
<td>75/25</td>
<td>114.5</td>
<td>118.1</td>
<td>129.5</td>
<td>114.0</td>
</tr>
<tr>
<td></td>
<td>50/50</td>
<td>113.5</td>
<td>103.5</td>
<td>129.0</td>
<td>101.1</td>
</tr>
<tr>
<td></td>
<td>25/75</td>
<td>117.7</td>
<td>66.5</td>
<td>127.1</td>
<td>70.5</td>
</tr>
<tr>
<td></td>
<td>00/100</td>
<td>102.3</td>
<td>49.8</td>
<td>124.0</td>
<td>48.3</td>
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<tr>
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<td>154.4</td>
<td>131.0</td>
<td>148.2</td>
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<td>127.8</td>
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<td>39.5</td>
<td>123.0</td>
<td>52.6</td>
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<tr>
<td>HDPE/BLLDP</td>
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<td>114.2</td>
<td>154.4</td>
<td>131.0</td>
<td>148.2</td>
</tr>
<tr>
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<td>75/25</td>
<td>113.5</td>
<td>133.3</td>
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<td>25/75</td>
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<td>68.8</td>
<td>126.7</td>
<td>75.5</td>
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<td></td>
<td>00/100</td>
<td>103.8</td>
<td>49.2</td>
<td>123.1</td>
<td>47.6</td>
</tr>
</tbody>
</table>

$T_c$: Crystallization temperature; $\Delta H_c$: Heat of crystallization; $T_m$: Melting temperature; $\Delta H_m$: Heat of melting.

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Figure 2. SEM micrographs of tensile fractured surface of HDPE/LLDPE (90/10) blends: (a) HDPE/OLLDPE, (b) HDPE/PLLDP, (c) HDPE/BLLDPE.

Figure 3. SEM micrographs of tensile fractured surface of HDPE/LLDPE (75/25) blends: (a) HDPE/OLLDPE, (b) HDPE/PLLDP, (c) HDPE/BLLDPE.
content, are quite distinct (Figure 3b). The diameter of the fibrils is around 5-8 µm. The fibrils of BLLDPE (Figure 3c) are regular and the average diameter of these fibrils is around 3-5 µm.

The $T_m$ and $\Delta H_c$ of HDPE/OLLDPE and HDPE/PLLDP blends at 25% LLDPE content are 114.5 °C, 115 °C, and 118 J/g, 125.3 J/g (Table 2), respectively. Though their $T_m$s are comparable, the $\Delta H_c$ value is considerably higher for PLLDPE, which is also reflected by the prominence of fibrils in the fracture morphology. Similar, though less distinguishable differences are seen in the $\Delta H_m$ values (Table 2), the $T_m$ show noticeable differences. The $T_m$ of PLLDPE blend is higher than PLLDPE at this composition. The predominant branching in the fibrils, as reported by various authors [11-13], may pronounce loose folds in the lamellae and the nucleation of these pendant group/side chains. From these pendant group/side chains crystallites may grow and as a result the branching may take place. The predominant group of PLLDPE perhaps has undergone such nucleation and shows branching of the crystallites.

In this case of branching, as the part of chain stays in the amorphous zone it may bring down the melting temperature to a great extent (as is evident by low $T_m$ value). In this composition the blend, containing 25% BLLDPE, shows the lowest $T_m$ and the highest $\Delta H_c$ among the blends. The low value of $T_m$ indicates some sort of hindrance operative in the process of crystallization; however, broader spread of crystallization temperature provides high heat of crystallization (i.e., 133.3 J/g). The highest $T_m$ (i.e., 130 °C) may support the regular and prominent morphology of HDPE/BLLDPE blend at 25% LLDPE composition.

The tensile at yield manifest similar behaviour i.e., BLLDPE blend at 25% composition shows highest tensile at yield among the blends with a value of 23.3 MPa (Table 3). The tensile strength at yield of PLLDPE blend, with less prominent fibrils, is 22.5 MPa which is even less than BLLDPE and in turn

Table 3. Tensile properties of HDPE/LLDPE blends.

<table>
<thead>
<tr>
<th>Blend</th>
<th>Blending ratio</th>
<th>$\sigma_y$ (MPa)</th>
<th>$\sigma_b$ (MPa)</th>
<th>$\varepsilon_y$ (%)</th>
<th>$\varepsilon_b$ (%)</th>
<th>$E_M$ (Gpa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDPE/OLLDPE</td>
<td>100/00</td>
<td>28.0</td>
<td>18.0</td>
<td>13.0</td>
<td>130.0</td>
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<td></td>
<td>90/10</td>
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<td>16.9</td>
<td>102.7</td>
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<td>75/25</td>
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<td>20.0</td>
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<td>0.43</td>
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</table>

$\sigma_y$: Tensile at yield; $\sigma_b$: Tensile at break; $\varepsilon_y$: Elongation at yield; $\varepsilon_b$: Elongation at break; $E_M$: Young's modulus.
greater than the tensile at yield of OLLDPE (i.e., 21.8 MPa) with less distinct fibrils.

Figure 4a,b,c shows the fracture morphology of the HDPE/LLDPE blends at 50/50 composition. The fibrils of OLLDPE blend (3–5 µm in diameter) are branched and quite prominent. The fibrils of PLLDPE (3–5 µm in diameter) are regular and rather prominent whilst the fibrils of BLLDPE (2–5 µm diameter) blend show interconnected flake in appearance, and are branched and discontinuous.

The $T_{sp}$ and $AH_L$ for all the blends at this 50% composition seem to be compatible. The $AH_m$ values of the blends are also comparable (Table 2). The $T_m$ values of the blends of LLDPE containing pentene and butene as co-monomer at 50/50 composition are close at 127.7 °C; however, the $T_m$ value of OLLDPE blend is higher by 1 °C.

The tensile strength at yield of PLLDPE blend is the highest (Table 3) among this category of 50% blend composition. The yield strength of PLLDPE blend is 20.3 MPa followed by OLLDPE blend with 19.16 MPa and that of BLLDPE with 18.6 MPa, respectively. The values of tensile at yield are difficult to explain either by crystallinity or by the endothermic peak temperature, however, the apparent discrepancy in fracture morphology would provide the adequate detail for the interpretation of this tensile behaviour. The discontinuities of BLLDPE are quite apparent from the micrographs, which signify the non-participation of all the fibrils in the load bearing process. The regularity of such fibrils in the OLLDPE blend seems to ensure greater participation and hence higher tensile strength at yield than BLLDPE blend. The PLLDPE with regular and distinct fibrils display highest tensile at yield in this class of blend composition (i.e., 50/50).

Figure 5a,b,c shows the micrographs of tensile fracture surface at 75% LLDPE content. The fibrils of OLLDPE blend show quite distinct type of morphology which is not apparent in the pure components. These fibrils are finer (i.e., 1–2 µm diameter), well formed and display no interfibrillar separation.

The fibrillar construction of LLDPE containing pentene and butene are almost similar notwithstanding PLLDPE fibrils are thicker (8–10 µm dia)
Figure 5. SEM micrographs of tensile fractured surface of HDPE/LLDPE (25/75) blends: (a) HDPE/OLLDPE, (b) HDPE/PLLDP, (c) HDPE/BLLDPE.

than BLLDPE (3–5 μm dia).

The $T_m$ and $\Delta H_m$ values (Table 2) reveal that the $T_m$ of OLLDPE blend is considerably higher (i.e., 117.7 °C), which is even higher than pure components, followed by PLLDPE and BLLDPE. The $\Delta H_m$ values, however, show different trend. The $\Delta H_m$ of PLLDPE maintains higher value at 124.6 J/g followed by BLLDPE at 69.0 and 66.5 for OLLDPE at this 75% composition. The $\Delta H_m$ shows similar trend as that of $\Delta H_c$. The $T_m$ of PLLDPE is highest at 128.8 °C followed by OLLDPE at 127.1 °C and BLLDPE at 126.7 °C.

The tensile strength at yield of PLLDPE blend at 75% composition is 22.0 MPa which is considerably higher than the blends of octene and butene containing LLDPE with values of 15 and 14.5 MPa, respectively. The fibrils of PLLDPE have the highest coarseness among the 75% LLDPE containing blend and shows highest $\Delta H_m$ of about 118.4 J/g in comparison to the 70.5 and 75.5 for OLLDPE and BLLDPE, respectively. These coarse fibrils signify the deformation of the crystallites of PLLDPE blend at this composition group to considerably bigger crystallites as compared to the OLLDPE and BLLDPE blend thus indicates the higher crystallizability of PLLDPE blend. This observation is supported by the $\Delta H_c$, $T_m$ and $T_c$ values (Tables 2 and 3).

The observed non-linearity in the values of elongation at break (column $\varepsilon_b$ of Table 3) could be due to sensitivity of this material property to the test conditions. The overall variation indicates that the LLDPE rich blends have a higher elongation at break values than the HDPE rich blends. However the sudden drop of the elongation at break, by addition of only a 10 wt% LLDPEs indicates the high phase separation due to incompatible nature of the blends.

CONCLUSION

Variable morphology is formed on blending of HDPE with LLDPEs at different compositions and these differences amplify in the tensile fracture surfaces. The presence of second component plays catastrophic
role indicating the morphology of the blend. Though the morphology of 50/50 blends is difficult to predict, the morphology of one component rich blend remotely followed the morphology of rich-percent component. The HDPE forms thick distinct and separated fibrils, whereas octene LLDPE (OLLDE) shows thinner intertwined fibrils and that of pentene based LLDPE (PLLDE) shows uniform, untwisted not so distinct fibrils. The distinct rod-like non-uniform fibrils are seen in the fracture surface of butene-based LLDPE (BLLDE).

The physical parameters of fibrils such as fineness, imperfections, twisting etc., show close relationship to the crystallization characteristics, melting behaviour and mechanical properties of blends and pure polymers. The relatively thick and without flaw fibrils showed better mechanical properties, higher heat of transitions ($\Delta H_m$, $\Delta H_\infty$), and lower crystallization temperature.

REFERENCES