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# Shear-induced epitaxial crystallization in injection-molded bars of high-density polyethylene/isotactic polypropylene blends

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## Abstract

As a continuation of our previous works on exploring shear-induced epitaxial crystallization of polyolefin blends during practical molding processing [Na et al. Polymer 2005; 46, 819 and 5258], the present study focused on the importance of molecular weight on the formation of epitaxial structure in injection-molded bars of high-density polyethylene (HDPE)/isotactic polypropylene (iPP) blends. By choosing two kinds of HDPE and two kinds of iPP with high molecular weight or low molecular weight, four blends with different molecular weight combinations can be designed. After making the blends via melt mixing, the injection-molded bars were prepared in a so-called dynamic packing injection molding equipment where repeated shearing was imposed on the melts during the solidification stage. Crystal structure and orientation were estimated mainly through 2D-WAXD. Our results indicated that an appropriate matching of low molecular weight HDPE and high molecular weight iPP was more favorable for epitaxial crystallization than other component matches. The effects of orientation and epitaxy on the re-crystallization behaviors of polyolefin blends have been elucidated in detail through PLM experiments. Moreover, epitaxy has been proved to play a positive effect in determining the ultimate mechanical properties of injection-molded bars.

*Keywords:* Epitaxial crystallization; Polyolefin blend; Injection molding

## 1. Introduction

As a complicated crystalline phenomenon, epitaxial crystallization has been intensively investigated in many polymer blends, such as polyamide/polypropylene(PP) [1], polyester/PP [2], polymethylpentene/polytetrafluoroethylene(PTFE) [3], PP/PTFE [4,5], polyethylene(PE)/PTFE [6,7] and PE/PP [8–13]. Many researches concerning epitaxial behavior have focused on the blends consisting of zigzag chain polymers and helix chain polymers, typical example is high-density polyethylene (HDPE)/isotactic polypropylene (iPP) blend.

The previous researches about the mechanism and the impact factors on epitaxial crystallization of HDPE/iPP blends were mostly conducted under ideal experimental conditions both for its scientific aspects and economic implications. In HDPE/iPP blends, the zigzag chains of PE are at ±50°apart from the *c*-axial direction of iPP lattice. This relationship has been explained in terms of the interaction of HDPE chains with rows of methyl groups that populate the (010) plane of iPP $\alpha$-crystals, since PE chains fit exactly into the valleys with a 0.5 nm intermolecular distance formed by the methyl groups of PP [14]. Geometric lattice matching for (100) plane of HDPE and (010) plane of iPP is considered to be

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indispensable, and only a 10–15% disregistry is the upper limit for the occurrence of epitaxial growth. Yan and Yang [9,16] proposed a critical temperature for epitaxial crystallization under the given experimental conditions, that is, a lower temperature is favorable for epitaxial growth. Since the epitaxy growth needs rigorous requirements, the investigations about epitaxial crystallization of polymer blends are usually limited to ideal experiment conditions and rarely extended into the practical processing states. In the past works of our group, the expitaxial structures of HDPE on the oriented iPP lamellae have been unambiguously demonstrated in the injection-molded bars [13] and the stretching films [12], in which chain orientation induced by shearing has been proved to pronouncedly facilitate epitaxial crystallization. Since various factors, such as composition, domain size and cooling rate, could affect either directed crystallization or epitaxy, therefore, a competition between bulk nucleation and interfacial nucleation may play an important role on determining the ultimate crystalline structures of HDPE/iPP blends in practical processing.

The present paper is a continuation of our previous work on exploring the possibility to achieve epitaxial crystallization in the injection-molded bars of HDPE/iPP blends. Since the molecular weight matching between HDPE and iPP may affect degree of orientation and sequence of crystallization of HDPE and iPP, so it can determine the occurrence and the degree of epitaxial crystallization. In this time, our major attention will be devoted to elucidate the effect of molecular weight (melt flow index) on epitaxial crystallization. Four blends with different molecular weight combination were prepared. To achieve the obvious epitaxial structures during molding processing, a so-called dynamic packing injection molding technology was adopted to prepare highly oriented blends, which relies on the application of repeated shear field to melt/solid interface during the solidification stage. The mutual-lamellar orientation and the epitaxial architecture between HDPE and iPP were mainly characterized by 2D wide-angle X-ray scattering, and the re-crystallization behaviors of as-prepared samples were measured by polarizing light microscopy. Finally, orientation and epitaxy that impacted the mechanical properties of injection-molded bars, were demonstrated.

## 2. Experimental

### 2.1. Materials

Two kinds of high-density polyethylene and two kinds of isotactic polypropylene used in this study were all commercially available. Melt flow index (MFI) was measured at 190 °C and 2.16 kg loading for HDPE and at 230 °C and 2.16 kg loading for iPP to quantitatively represent the magnitude of average molecular weight. A higher MFI means a lower average molecular weight and vice versa. The detailed information of polymers is listed in Table 1. HDPE F600 possesses a lower MFI while HDPE 2911 has a higher one; the MFI of PP T30s was lower than that of PP W75. All blends had a fixed PP content of 70 wt% and were labeled according to their molecular weight (MFI); they are represented as HMW-HDPE/HMW-iPP, HMW-HDPE/LMW-iPP, LMW-HDPE/HMW-iPP, and LMW-HDPE/LMW-iPP.

Table 1
Various parameters of polyolefin used in this work

| Material | Trademark | Supplier | $T_m$ (°C) | $T_c$ (°C) | MFI (g/10 min) |
|---|---|---|---|---|---|
| HDPE | F600 | Korea Petrol Co. | 130.4 | 117.0 | 0.05 |
| HDPE | 2911 | Lanzhou Petrol Co. | 129.8 | 113.8 | 20.5 |
| iPP | T30s | Dushanzi Petrol Co. | 165.2 | 112.7 | 2.8 |
| iPP | W75 | Wuhan Petrol Co. | 164.4 | 114.6 | 60.0 |

### 2.2. Sample preparation

HDPE and iPP were melt compounded together using a TSSJ-25 co-rotating twin-screw extruder with a barrel temperature of 160–190 °C. After pelleting and drying, HDPE/iPP blends were injected into a mold with the aid of a SZ100 g injection molding machine at a barrel temperature of 190 °C, and then dynamic packing injection molding (DPIM) technology was applied, whose main feature was to introduce repeated shearing to the cooling melt during the packing stage by two pistons that moved reversibly with the same frequency (0.3 Hz). Some parameters of injection molding are presented in Table 2 and the schematic representation of DIPM can be found in our previous works [17,18].

### 2.3. 2D wide-angel X-ray scattering

The 2D wide-angle X-ray scattering (2D WAXS) experiments were conducted on a SEIFERT (DX-Mo8*0.4s) diffractometer equipped with a 2D Mar345 CCD X-ray detector. The wavelength of the monochromatic X-ray from Mo radiation was 0.71 nm and the sample-to-detector distance was 400 mm. The samples were placed with the orientation (flow direction) perpendicular to the beams. All the 2D WAXS patterns given in this article have extracted the background thus allows a qualitative comparison between various samples. Azimuthal scans (0 ∼ 360°) of 2D WAXS were made for the (110) plane of HDPE and (040) plane of iPP at a step of 1°. The orientation level of various planes could be calculated by the orientation parameter $f$,

$$f = \frac{3\langle \cos^2 \varphi \rangle - 1}{2} \tag{1}$$

Table 2
Processing parameters of dynamic packing injection molding

| Parameters | Values |
|---|---|
| Injection pressures | 90 MPa |
| Oscillating packing pressures | 4 MPa |
| Oscillating frequency | 0.33 Hz |
| Holding time | 3 min |
| Melt temperature | 190 °C |
| Mold temperature | Room temperature |

$$\left\langle \cos^2 \varphi \right\rangle = \frac{\int_0^{\frac{\pi}{2}} I(\phi) \sin \phi \cos^2 \phi \, \mathrm{d}\phi}{\int_0^{\frac{\pi}{2}} I(\phi) \sin \phi \, \mathrm{d}\phi} \qquad (2)$$

where $\varphi$ is the angel between the normal of a given (*hkl*) crystal plane and shear flow direction, and $I$ is the intensity. Its limiting values of orientation parameter $f$, taking $\varphi = 0$ as the shear flow direction, are −0.5 for a perfectly perpendicular orientation and 1.0 for a perfectly parallel orientation. An un-oriented sample gives $f = 0$.

### 2.4. Polarizing light microscopy

The morphological observations of HDPE/iPP crystallization were investigated on a Leica DMIP polarizing light microscopy (PLM) equipped with a Linkam THMS 600 hot stage. Thin slices were cut from the intermediate zones of injection-molded bars, inserted between two microscope cover glasses, melted at 180 °C and squeezed to obtain thin films. Then, slices were heated to 180 °C and held for 2 min to achieve thermal equilibrium. Subsequently, samples were cooled to 100 °C at a constant rate of 6 °C/min. For comparison purpose, thin films were then heated to 190 °C, holding for 5 min to erase the previous thermal history and then cooled with the same cooling rate (6 °C/min). The proceedings of non-isothermal crystallization were recorded by taking photomicrographs at appropriate intervals, using a Canon PowerShot 550 digital camera.

## 3. Results and discussions

### 3.1. Epitaxial crystalline structures in HDPE/iPP injection-molded bars

Fig. 1 shows the 2D WAXS patterns, obtained at the intermediate zones of injection-molded bars of four blends. Corresponding azimuthal scans of 2D WAXS are also included for clarity. From inner to outward, the reflection circles or arcs are originated from (110), (040) and (130) planes of iPP α-modification and (110), (200) planes of orthorhombic crystallites of HDPE. The reflections of (111) and (−131) planes of iPP are overlapped with (110) plane of HDPE due to similar $2\theta$ angle. Note that the strong reflections of (*hk*0) plane for iPP component at the equator in these four samples indicate that the *c*-axes of iPP lamellae are preferentially oriented along the shearing direction. For the (110) plane of iPP, four reflection arcs symmetrically emerge around the meridian, which are considered to be an indication of lamellar branching through homoepitaxy between α-crystals themselves [1].

As to the reflection of HDPE (110) plane, four intensive arcs appear at about ±50° apart from the meridian, while being the shear direction. Combination with the permanent equatorial reflection of (200) plane, this special reflection can be ascribed to the epitaxial growth of HDPE lamella onto that of iPP, since HDPE chains are inclined about 50° to the iPP chain axis and the contact plane is established to be (100) HDPE and (010) iPP [14]. These results are consistent with that has been described by our previous works [12,13]. However, it should be noted that there are two modest peaks at 90°and 270°for HMW-HDPE/HMW-iPP and HMW-HDPE/LMW-iPP blends, whereas no features are found for LMW-HDPE/HMW-iPP and LMW-HDPE/LMW-iPP blends. The only explanation for the inexistence of the peaks at 90° and 270°in (110) plane for these two samples is the diffraction spots of (110) plane shift to the position where (111) plane of iPP occupies and strengthen them, as indicated by our previous work [12]. This can be ascribed to the epitaxial growth of HDPE lamellae onto that of iPP for LMW-HDPE/HMW-iPP and LMW-HDPE/LMW-iPP blends. As for the combination of HMW-HDPE/HMW-iPP and HMW-HDPE/LMW-iPP blends, although it is cannot be arbitrarily considered that there is no epitaxial grown in them, the extent should be lower than other two blends. For HDPE crystallites, it is usually observed that the reflection intensity of (110) plane on the meridian is much stronger than that of (020) plane if direct crystallization occurs in its bulk, otherwise the intensity of the latter will be stronger or equivalent to the former when there exists epitaxial crystallization. In addition, Fig. 1f gives another information that the diffraction intensity of (200) plane in LMW-HDPE/HMW-iPP blend is stronger than that of LMW-HDPE/LMW-iPP blend, which means that LMW-HDPE/HMW-iPP has a better orientation of PE. Therefore, it can be concluded that the level of epitaxy in LMW-HDPE/HMW-iPP blend is higher than that in LMW-HDPE/LMW-iPP blend. So it can be logically concluded that the appropriate molecular weight matching for facilitating epitaxial growth is iPP (high molecular weight)/HDPE (low molecular weight).

Recently, there are many reports on the crystallization kinetics and morphologies in polyolefin blends under shear [19,20]. Among these studies, the role of the long-chain species in flow or shear-induced crystallization has attracted the most attention. It was found that if the long chains were present, shear-induced crystallization was more sensitive to shear treatment and orientation of the sample was much higher [19]. In our work, one expects a highest degree of orientation in HMW-HDPE/HMW-iPP blends and lowest in LMW-HDPE/LMW-iPP blends. Table 3 shows the values of orientation parameters at the intermediate zones in various injection-molded bars, which can give a half-quantitative comparison between the degrees of lamellae orientation in different samples. In all samples, the orientation parameters of (110) plane of HDPE and (040) plane of iPP are high whatever the molecular weight is whether large or small, which implied that shearing has induced highly oriented structures in all of samples. However, the high degree of epitaxial crystallization is found only in LMW-HDPE/HMW-iPP blend and LMW-HDPE/LMW-iPP blend, and is absent (or weak) in HMW-HDPE/HMW-iPP blend and HMW-HDPE/LMW-iPP blend. That is, HDPE with high molecular weight is difficult to epitaxial grow on

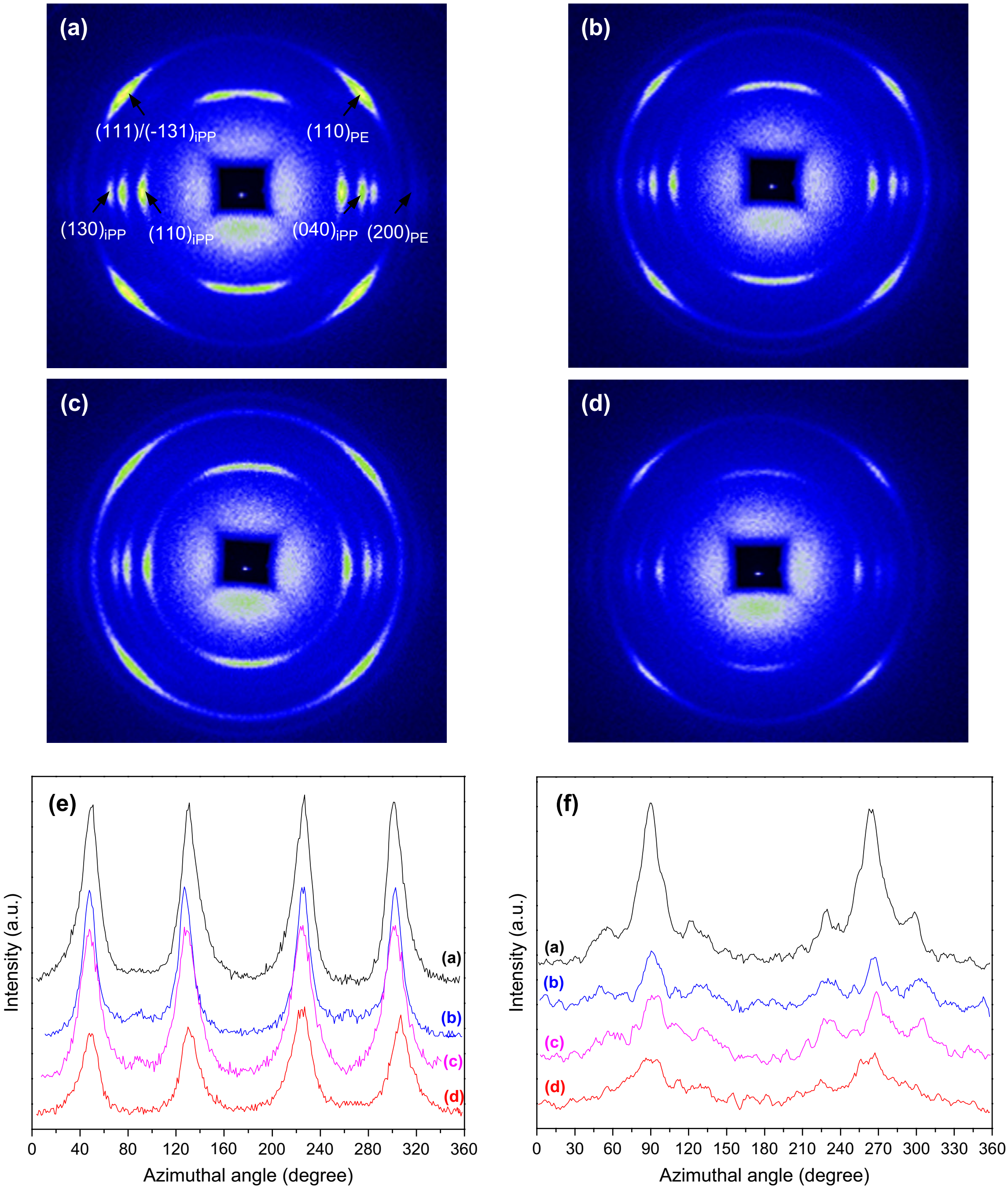

Fig. 1. 2D-WAXS patterns at the intermediate zones of injection-molded bars: (a) LMW-HDPE/HMW-iPP; (b) HMW-HDPE/HMW-iPP; (c) HMW-HDPE/LMW-iPP; (d) LMW-HDPE/LMW-iPP. Azimuthal scans of both; (e) (110) and (f) (200) planes of HDPEs for above four samples are also included. The shearing direction is vertical.

Table 3
Orientation parameters at the intermediate zones of various molded bars: (1) LMW-HDPE/HMW-iPP; (2) LMW-HDPE/LMW-iPP; (3) HMW-HDPE/HMW-iPP and (4) HMW-HDPE/LMW-iPP

| Sample | (110) Plane of HDPE | (040) Plane of iPP |
|---|---|---|
| (1) | 0.96 | 0.95 |
| (2) | 0.95 | 0.94 |
| (3) | 0.93 | 0.94 |
| (4) | 0.94 | 0.92 |

the oriented iPP. This is probably due to its higher density of entanglement and slower crystallization rate.

The hierarchical structure along the thickness of injection-molded bar can be ascertained through obtaining 2D-WAXS patterns at different regions. For an example, Fig. 2 shows 2D-WAXS patterns that represented different zones in the bar of LMW-HDPE/LMW-iPP blend. The azimuthal scans of HDPE (110) planes are carefully picked out and also included. It can be concluded that the epitaxy occurs mostly in the intermediate zone, less in the core, hardly in the skin

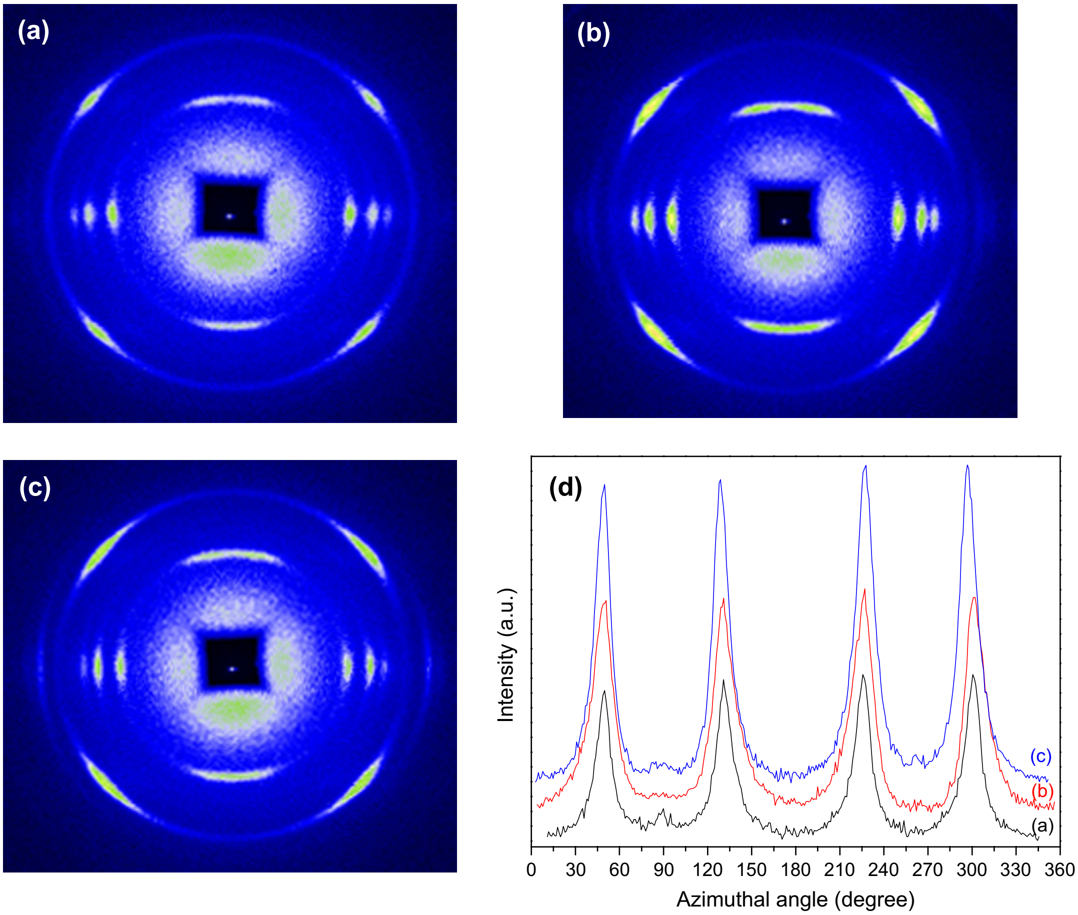

Fig. 2. 2D-WAXS patterns of LMW-HDPE/HMW-iPP at different zones of molded bar obtained via DPIM: (a) the skin; (b) the intermediate zone; (c) the core. (d) Azimuthal scans of (110) plane of HDPE for the above three samples are also included. The shearing direction is vertical.

of injection-molded bar. We know that there exists a temperature gradient in the sample with the highest temperature in the core and lowest in the skin. Thus the polymer chains are almost immediately frozen in the skin and no time to adjust them into epitaxial growth. While in the core the oriented molecular chain can be easily relaxed, leading to a relative low orientation. Therefore, less epitaxial growth is observed.

### 3.2. Re-crystallization behaviors

For a complementary understanding about the crystallographic features of HDPE/iPP blends with oriented and epitaxial structure induced by repeated shearing during injection-molded processing, the re-crystallization behaviors of as-prepared samples were elucidated through PLM experiments. One expects direct observation of crystal formation from an oriented melt via real-time optical experiments. Showing as an example, optical micrographs, taken during non-isothermal crystallization of the injection-molded bar of LMW-HDPE/LMW-iPP blend, cooled from 180 °C to room temperature, at a cooling rate of −6 °C/min, are shown in the Fig. 3. Below the melting point, there is a region of about 30 °C, where no crystallite can be observed. Decreasing the temperature down to 137 °C, some thread-like crystallites, formed via self-seeding of shish structure and attributed to iPP, appear in the view field (Fig. 3a). The size of these thread-like crystallites is not uniform, which is a indication of the existence of shish structure with different length caused by the different molecular weight. With decreasing of the temperature, a lateral growth of thread-like crystallites is observed (Fig. 3b and c). With further decrease in temperature a few new spherulites via heterogeneous nucleation appear (Fig. 3d). Clearly, there exist two kinds of crystal growth during cooling, thread-like crystallites at relatively high temperature followed by spherulites at lower temperature. The former can only grow laterally due to the large aspect ratio, whereas the latter can divergently grow. To identify the effect of epitaxial crystallization on the shish structure among the four samples, the same experiment was repeated with different blends. The similar threadlike crystallites are always observed, that is, the shape and size of these anisotropic crystals are similar. The only difference is that the temperatures at which shish structures appear are different from one another. At this moment one cannot find obvious relationship between shish and epitaxy from the information supplied by the PLM results.

It is expected that the crystallization will be retarded and crystal morphology will be changed after the relaxation of the oriented structure by melting at higher temperature or at a longer time. Fig. 4 shows the development of

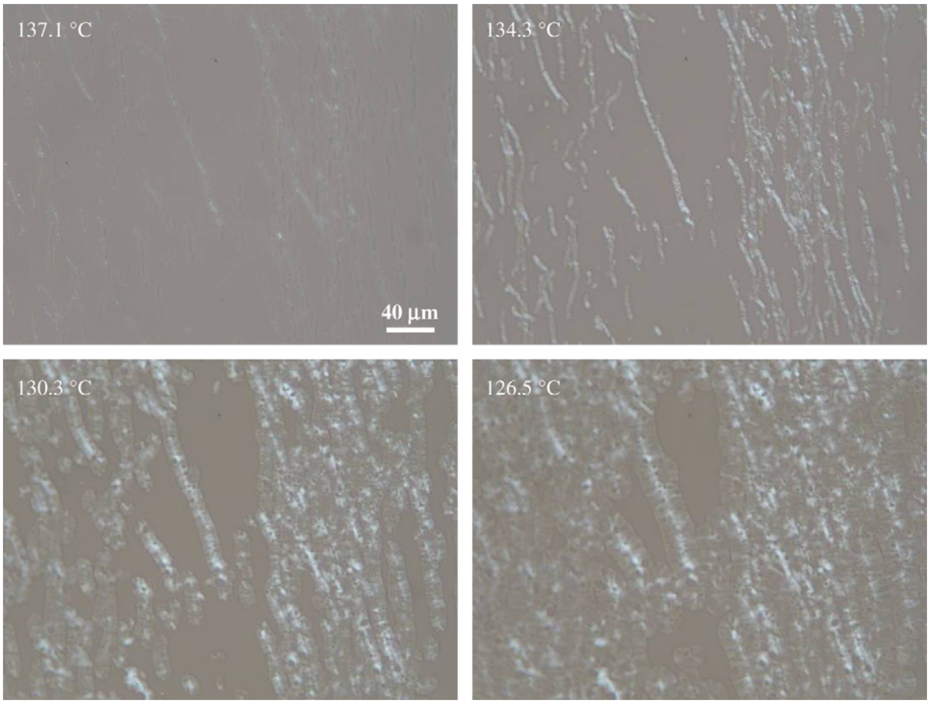

Fig. 3. Optical pictures of LMW-HDPE/LMW-iPP taken at interval for the sample melted at 180 °C and held for 2 min, then cooled at a rate of −6 °C/min.

spherulites of iPP with temperature, taken from the cooling stage, of the injection-molded bar of LMW-HDPE/LMW-iPP blend after annealing at 190 °C for 10 min. As can be seen, the crystallization can only proceed at larger supercooling, about 126 °C, and only spherulites emerge. The density of nucleation is much lower than that shown in Fig. 3. Clearly, shish structure has been relaxed to the random coil and the nucleation mechanism is changed, from self-nucleation on shish structure to heterogeneous nucleation on the impurities.

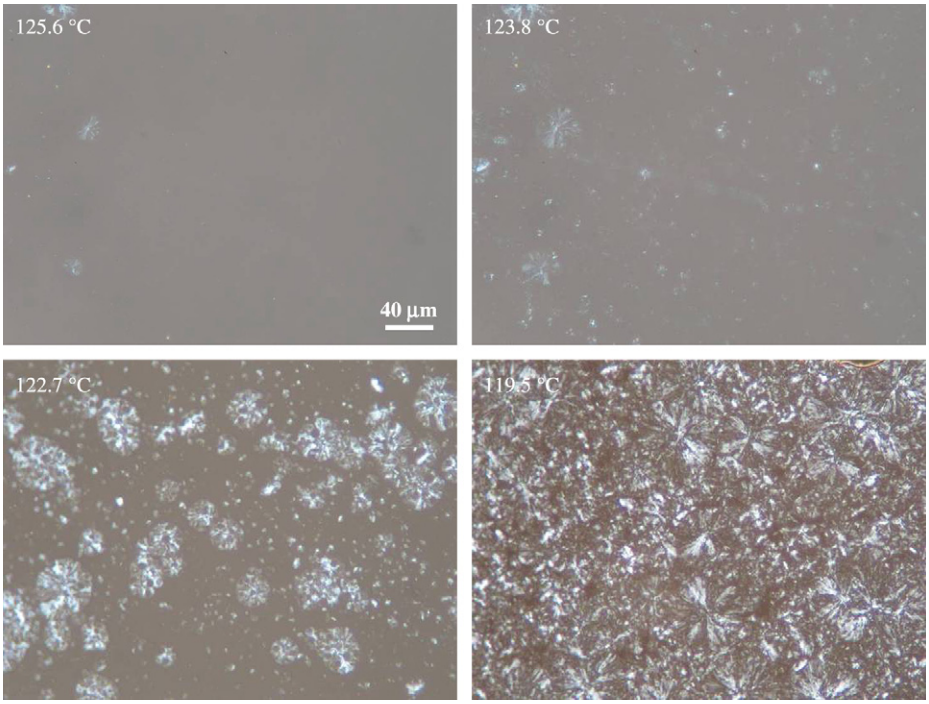

Fig. 4. Optical pictures of LMW-HDPE/LMW-iPP taken at interval for the sample melted at 190 °C and held for 10 min, then cooled at a rate of −6 °C/min.

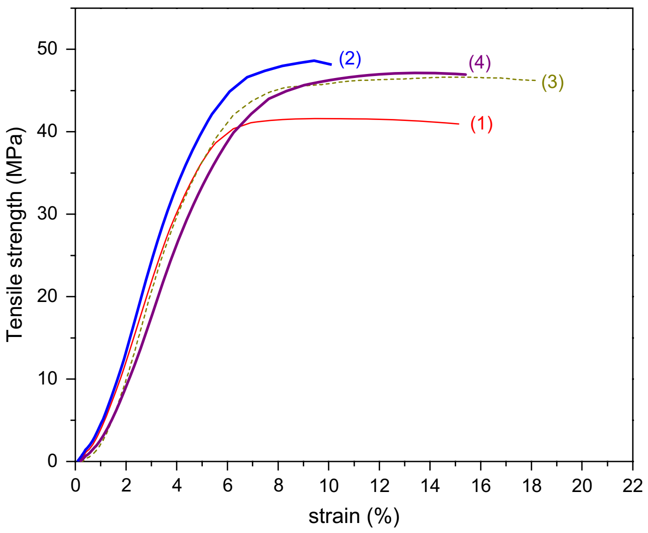

Fig. 5. Selective stress–strain curves of various samples obtained via DPIM: (1) HMW-HDPE/HMW-iPP; (2) LMW-HDPE/HMW-iPP; (3) HMW-HDPE/LMW-iPP and (4) LMW-HDPE/LMW-iPP.

### *3.3. Mechanical properties*

Stress–strain curves of various samples obtained via DPIM are shown in Fig. 5. Values of tensile strength and tensile modulus are listed in Tables 4 and 5. For the injection-molded bars prepared via DPIM, LMW-HDPE/HMW-iPP blend has the highest tensile strength with 48.8 MPa, followed by LMW-HDPE/LMW-iPP blend (46.6 MPa) and HMW-HDPE/LMW-iPP blend (46.5 MPa), meanwhile, HMW-HDPE/HMW-iPP blend has the lowest strength of 41.6 MPa. To our knowledge, LMW-HDPE/HMW-iPP blend and LMW-HDPE/LMW-iPP have higher tensile strength that could attribute to the epitaxial crystallization, for it can improve mechanical performances by the bridging of the amorphous interlamellar phase of one component by the crystalline lamellar component of the other phase. The longer molecular chain of HDPE in HMW-HDPE/HMW-iPP should have an higher density of entanglement, which won't favor the extending of the molecular chains. In this case, the interaction of HDPE chains with rows of methyl groups will be weakened as compared with molecular weight of HDPE, leading to a weak epitaxial crystallization. Thus a lower tensile strength is obtained through the overall lowered degree of orientation. It is valuable to note that LMW-HDPE/HMW-iPP blend prepared via DPIM has improved 30% more than that of HMW-HDPE/HMW-iPP in tensile strength, whereas the strengths for these two counterparts prepared via conventional injection molding are 37.4 MPa and 40.5 MPa, respectively. As to tensile modulus, similar result can be found: DPIM sample of LMW-HDPE/HMW-iPP blend has the highest modulus, improved by 30% compared to its counterpart prepared via conventional molding. Nevertheless, there seems no significant difference between either the basal polymers of the two HDPEs or those of the two iPPs, as depicted in Table 6. From the above discussion, the conclusion can be drawn that epitaxial crystallization favors an improvement of tensile properties of polyolefin blends.

As for impact strength is considered, LMW-HDPE/LMW-iPP blend has the best impact performance of 48.1 kJ/m$^2$, better than LMW-HDPE/HMW-iPP blend and HMW-HDPE/HMW-iPP blend, both are nearly 40 kJ/m$^2$; HMW-HDPE/LMW-iPP blend is the poorest, just as 28.3 kJ/m$^2$. It should be pointed out that the impact strengths of those DPIM samples have been improved by 541%, 1097%, 933%, and 732% compared with the corresponding samples prepared via conventional molding, respectively. Considering that the impact strength of the HMW-HDPE is higher than that of LMW-HDPE, a similar conclusion can be drawn that epitaxy plays a positive influence on impact strength.

## 4. Conclusions

The effect of molecular weight on epitaxial crystallization has been investigated in detail mainly with the aid of 2D WAXS. The result in this work suggests that the combination of high molecular weight iPP and low molecular weight HDPE is favorable for epitaxial growth. For a sample obtained via DPIM, epitaxy occurs mostly at shear layer, less in core, hardly in skin. Thread-like crystals, though have no evident relation to epitaxy, are found when the samples were cooled after held at 180 °C for 2 min but they cannot be seen when the holding temperature is higher (190 °C). It's also proved that epitaxial crystallization could improve the mechanical properties of injection-molded bars of HDPE/iPP blends.

Table 4
Mechanical properties of molded bars obtained via DPIM: (1) LMW-HDPE/HMW-iPP; (2) LMW-HDPE/LMW-iPP; (3) HMW-HDPE/HMW-iPP and (4) HMW-HDPE/LMW-iPP

| Properties | (1) | (2) | (3) | (4) |
|---|---|---|---|---|
| Tensile strength (MPa) | 48.8 | 46.6 | 41.6 | 46.5 |
| Tensile module (MPa) | 1124 | 925 | 1002 | 1051 |
| Impact strength (kJ/m$^2$) | 39.5 | 48.1 | 37.2 | 28.3 |

Table 5
Mechanical properties of molded bars obtained via conventional injection molding: (1) LMW-HDPE/HMW-iPP; (2) LMW-HDPE/LMW-iPP; (3) HMW-HDPE/HMW-iPP and (4) HMW-HDPE/LMW-iPP

| Properties | (1) | (2) | (3) | (4) |
|---|---|---|---|---|
| Tensile strength (MPa) | 37.4 | 36.7 | 40.5 | 38.1 |
| Tensile module (MPa) | 872 | 1015 | 956 | 994 |
| Impact strength (kJ/m$^2$) | 3.3 | 7.5 | 3.6 | 3.4 |

Table 6
Mechanical properties of molded bars of various pure polymers prepared via DPIM

| Properties | PP (T30s) | PP (W75) | PE (F600) | PE (2911) |
|---|---|---|---|---|
| Tensile strength (MPa) | 46.8 | 49.2 | 77.4 | 75.0 |
| Tensile module (MPa) | 1093 | 1093 | 1002 | 1171 |
| Impact strength (kJ/m$^2$) | 11.8 | 9.6 | 24.9 | 16.6 |

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