

In the supplement, we discuss the melt instability problem on the precursor formation of isotactic polystyrene (iPS) in Linkam CSS-450. The present shear experiments were mainly done in the shear rate range of  $7\text{--}90\text{ s}^{-1}$  at a temperature of  $250\text{ }^\circ\text{C}$ . Unfortunately we could not measure the viscosity in Linkam cell, but we have roughly estimated the viscosity of the blend sample which was  $\sim 10^3\text{ Pa s}$ , resulting in a wall shear stress of  $\sim 0.1\text{ MPa}$  at  $90\text{ s}^{-1}$ . This value is close to the onset of instability. However, as shown in the previous paper [*Polymer* **2009**, *50*, 2095] we observed the similar oriented precursor of iPS in micrometer scale at lower shear rate ( $4\text{ s}^{-1}$ ), at higher temperature ( $270\text{ }^\circ\text{C}$ ) and at lower molecular weight ( $M_w = 400\,000$ ). This suggests that the instability is not essential for the oriented precursor formation. In addition, we show other examples to say the melt instability is not essential for the oriented precursor. In the shear-induced crystallization experiments on polyethylene (PE) [*Polymer* **2005**, *46*, 1878] and isotactic polypropylene (iPP) [*Macromolecules* **2006**, *39*, 7617] we also observed similar oriented structure in micrometer scale at rather low shear rate ( $\sim 7\text{ s}^{-1}$ ) at low molecular weight (low viscosity). The results were cited from the papers in Figures A and B. It is believed that that formation of the oriented precursor is not due to the instability effect.

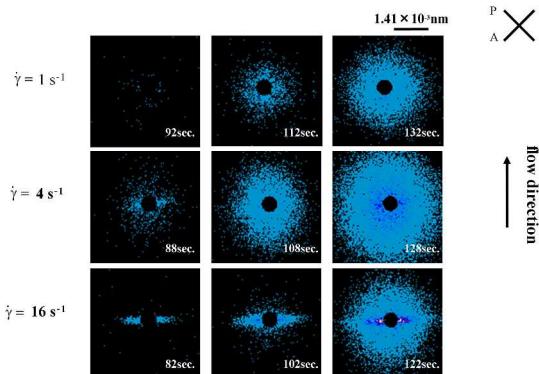


Fig.4  
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Figure A. Time evolution of DPLS (depolarized light scattering) pattern form PE during the crystallization process at  $129\text{ }^\circ\text{C}$  after pulse shear with shear rates =  $\dot{\gamma}1, 4$  and  $16\text{ s}^{-1}$  from top to bottom at a given shear strain  $\varepsilon = 3200\text{ \%}$  [*Polymer* **2005**, *46*, 1878].

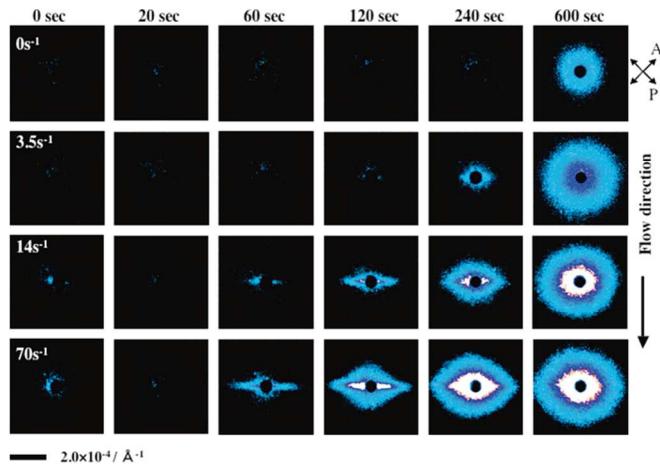


Figure B. Time evolution of 2D DPLS patterns during crystallization process of iPP at 132 °C for various shear rates,  $\dot{\gamma} = 0, 3.5, 14$  and  $70 \text{ s}^{-1}$  [*Macromolecules* **2006**, *39*, 7617].

Another example is shown below to suggest that melt instability is not essential for the oriented precursor in micrometer scale. In shear-induced crystallization experiment on iPP under an experimental condition [shear rate =  $70 \text{ s}^{-1}$ ,  $T = 132 \text{ }^{\circ}\text{C}$ ,  $M_w = 238\ 000$ , for details see *Macromolecules* **2006**, *39*, 7617], we observed very homogenous orientation of melt before crystallization by POM which is shown in Figure C.

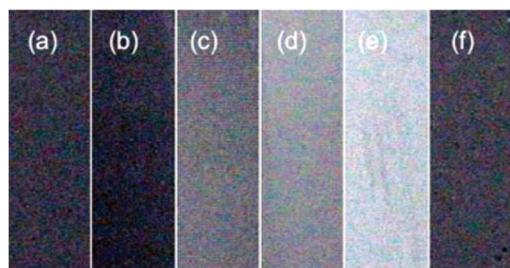


Figure C. POM images before (a), during (b-e), and after (f) shearing for 1 s. (a) 1 s before application of the shear; (b) 0.25, (c) 0.5, (d) 0.75, and (e) 1.0 s after starting the shear; (f) 2 s after cessation of the shear. Shear rate is  $70 \text{ s}^{-1}$  and shear direction is vertical [*Macromolecules* **2006**, *39*, 7617].

This result means that melt instability does not occur or, at least is not dominant. In case of iPS, the crystallization occurs very fast so that it is hard to take a POM picture before formation of the oriented precursor (or before crystallization), but we observed homogeneous POM pictures

above  $\sim 280$  °C. This temperature is a critical temperature for the oriented precursor formation. This suggests that the instability is not a dominant factor for the oriented precursor formation in the present experiments.

As mentioned in the main text, “small objects were occasionally observed and then disappeared”, which seems to indicate the melt instability. However, it is not the case as shown below. Such small objects were also observed at low shear rate and at high temperature ( $\sim 280$  °C) [*Polymer* **2009**, 50, 2095; *J. Polym. Sci., Part B: Polym. Phys.* **2011**, 49, 214], implying that the melt instability is not a dominant factor for the formation. We think that the small oriented precursor (but not so small because we can see by POM) which does not include crystallites with higher melting temperature than the nominal melting temperature is occasionally formed. Such crystallites may be often formed even in the large oriented precursor, but the large oriented structure also includes crystallites with higher melting temperature than the nominal melting temperature, and hence they can survive even above the nominal melting temperature. This was confirmed in a two-step decay of the size in the oriented structure as shown in our previous paper [*J. Polym. Sci., Part B: Polym. Phys.* **2011**, 49, 214]. Therefore we believe that the small objects are not due to the melt instability.

We also comment on the bubble formation. In low shear experiments below  $\sim 90$  s<sup>-1</sup> we hardly observed bubbles, but at a high shear rate of 90 s<sup>-1</sup>. Even at the high shear rate, the area occupied by bubbles was limited in a small area. In the vicinity of these bubbles, the localized deformation could result in the orientation of iPS and aPS. However, we consider that the orientation of iPS and aPS was dominated by the applied shear, not the local flow fields in the vicinity of these bubbles because the oriented precursor was formed when bubbles were not formed. Therefore we consider that the instability effect is not a dominant factor resulting in orientation of molecular chains. Based on the above discussions it is considered that the instability is not dominant for the formation of oriented precursor in micrometer scale but the instability effect was not negligible at a high shear rate of 90 s<sup>-1</sup>.