



	<b>Experiment title:</b> Formation of self-reinforced iPP composite from oriented tapes under elevated pressures	<b>Experiment number:</b> ME-638
<b>Beamline:</b> ID-11	<b>Date of experiment:</b> from: 17/06/2004 to: 21/06/2004	<b>Date of report:</b> 22/11/2004
<b>Shifts:</b> 12	<b>Local contact(s):</b> Dr. Silvia Capelli	<i>Received at ESRF:</i>
<b>Names and affiliations of applicants (* indicates experimentalists):</b> Liron Shavit-Hadar <sup>*1</sup> , Dmitry M. Rein <sup>*1</sup> , Rafail Khalfin <sup>1</sup> , Yachin Cohen <sup>*1</sup> , Ann E. Terry <sup>*2</sup> , Sanjay Rastogi <sup>2</sup> <sup>1</sup> Department of Chemical Engineering, Technion-Israel Institute of Technology, Haifa Israel 32000 <sup>2</sup> Dept. of Chemical Engineering, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands		

In this study we investigated the melting and recrystallization of oriented tapes of isotactic polypropylene (iPP) under elevated pressure. Here we report some of the preliminary results. Figure 1 shows two X-ray patterns obtained under pressure of 650 bar and in a temperature scan which is relevant for the compaction technology previously suggested by the group<sup>1</sup>. Figure 1a displays the reflections of the original state which is the  $\alpha$  crystalline form. It is possible to see that the original sample is highly oriented. From that point the sample was completely melted in two cycles. In each, the sample temperature reached the value of  $\sim 240^{\circ}\text{C}$ . Figure 1b displays the reflections of the final state. In that figure it is possible to see that there is a complete loss of orientation. In addition, there is a peak which corresponds to the  $\gamma$  crystalline form (hardly visible, but marked with a dashed line).

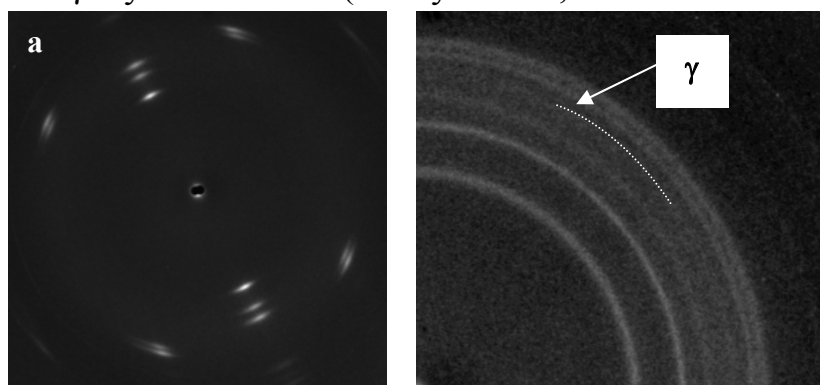


Figure 1: X-ray diffraction patterns at room temperature of unidirection iPP tapes. The sample had undergone two runs of complete melting until temperature of  $\sim 240^{\circ}\text{C}$  and recrystallization under pressure of 650 bar: (a) initial state of the sample. (b) final state of the sample. This pattern is one quadrant of the detector plane whereby (0 0 0) is located at the lower left corner of the pattern. The orientation direction is diagonal from lower left.

As was expected, the appearance of the  $\gamma$  form is getting more pronounced as the pressure under which the melting and recrystallization occurs gets higher. Figure 2 represents few X-ray diffraction patterns of a sample which undergone two runs of heating until complete melting and cooling, under pressure of 2000 bar. The finger print of the  $\gamma$  form is clearly visible. In addition, it is possible to see that the height of that peak from the second run is lower than that obtained from the first run (relatively to their base lines). That means that the content of the  $\gamma$  form obtained from the second heating-cooling run is smaller.

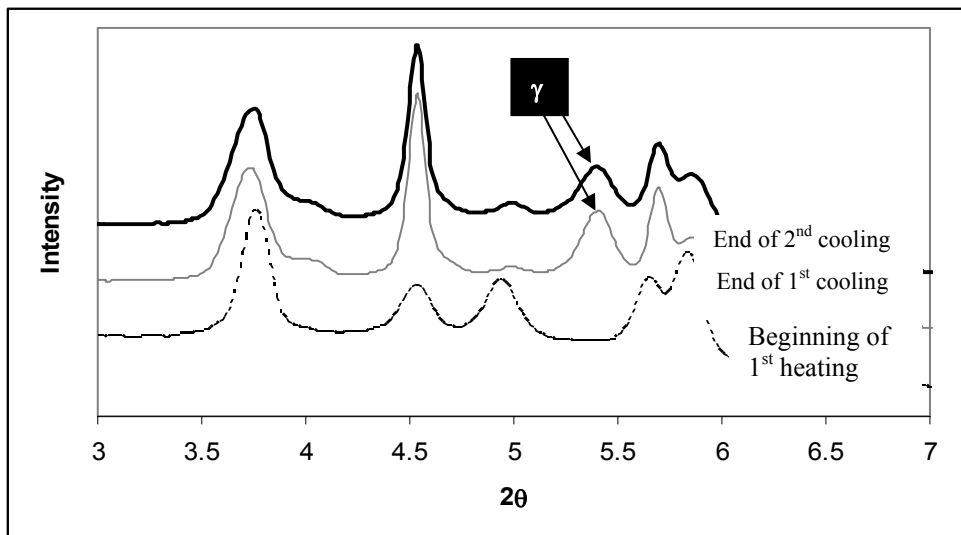


Figure 2: X-ray diffraction patterns of the unidirectional iPP tapes runs two runs of complete melting until temperature of  $\sim 300^{\circ}\text{C}$  and recrystallization under pressure of 2000 bar.

This phenomenon is clearer in Figure 3, which represents the relative content of the  $\gamma$  form compared to the  $\alpha$  form as a function of the compaction pressure. The content was calculated

from the empirical relation<sup>2</sup>: 
$$X_{\gamma} = \frac{h_{\gamma}}{h_{\gamma} + h_{\alpha}} \cdot 100$$

Where  $h_{\alpha}$ ,  $h_{\gamma}$  are the heights of the (130) peak of the  $\alpha$  form and the (117) peak of the  $\gamma$  form, respectively, measured from a base line. Surprisingly, the  $\gamma$  form content as a function of pressure in the first cycle exhibits a minimum, while we expected a constant increase of the content with the pressure<sup>3,4</sup>. Furthermore, a difference in the content between the first run and the second run is clearly observed. The highly oriented state of the original tapes can suggest the reason for that difference: the first run creates an unoriented state from a highly oriented one. Therefore, the second run starts and ends in unoriented state. Since orientation leads to preferred formation of the  $\gamma$  form, which until now was observed only in injection molding<sup>5</sup>, we expected that the first run will lead to a higher content of the  $\gamma$  form. Analysis of the melting and crystallization processes under pressure is currently in progress.

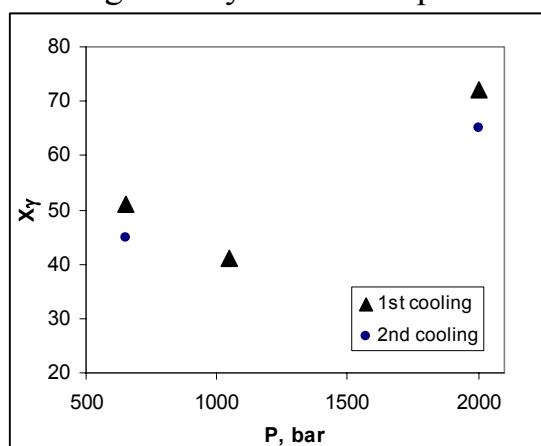


Figure 3: the relative content of the  $\gamma$  form compared to the  $\alpha$  form as a function of the compaction pressure.

1. Rein D.M.; Vaykhansky L.E.; Khalfin R.L.; Cohen Y., "Controlling the properties of single-polymer composites by surface melting of three reinforcing fibers", *Polymers for advanced technologies* 2002, **13**, 1046-1054.
2. Pae K.D., " $\gamma$ - $\alpha$  solid-solid transition of isotactic polypropylene", *Journal of Polymer Science A-2* 1968, **6**, 657-663.
3. Nakafuku C., "High pressure DTA study on the melting and crystallization of isotactic polypropylene", *Polymer* 1981, **22**, 1673-1676.
4. Campbell R.A.; Phillips P.J.; Lin J.S., "The gamma phase of high-molecular weight polypropylene: 1. Morphological aspects", *Polymer* 1993, **34**, 4809-4816.
5. Kalay G.; Bevis M.J., "Processing and physical property relationships in injection molded isotactic polypropylene. 2. Morphology and crystallinity", *Journal of Polymer Science B: Polymer Physics* 1997, **35**, 265-291.