DUBBLE – EXPERIMENT REPORT

We kindly request you to answer the questions (max 2 pages) and return the form to NWO **within 2 months of the completion of the experiment** to dubble@nwo.nl

Beam time number: 26-02-710		**File number:** 35714
Beamline: BM 26B	**Date(s) of experiment:** 11/12/2014 – 13/12/2014	**Date of report:** 1-2-2015
Shifts: 9	**Local contact(s):** G. Portale	

1. **Who took part in the experiments?** (Please indicate names and affiliations)
   M. van Drongelen, E.M. Troisi and H.J.M. Caelers, all affiliated to the department of Mechanical Engineering, Eindhoven University of Technology. And *Y. Zhou*, affiliated to the department of Chemical Engineering and Chemistry, Eindhoven University of Technology.

2. **Were you able to execute the planned experiments?**
   YES, the main goal of our experiment was to investigate the structure evolution during film blowing at multiple positions from the die, for 3 different materials at 4 different processing conditions each. The total set (12 conditions) was finished within the amount of shifts provided.

3. **Did you encounter experimental problems?**
   NO, everything was fine.

4. **Was the local support adequate?**
   YES, installing the setup went smoothly, software and hardware working accordingly.

5. **Are the obtained results at this stage in line with the expected results as mentioned on the project proposal?**
   YES, see the brief summary below. Pictures of the setup are provided below in Fig. 1.

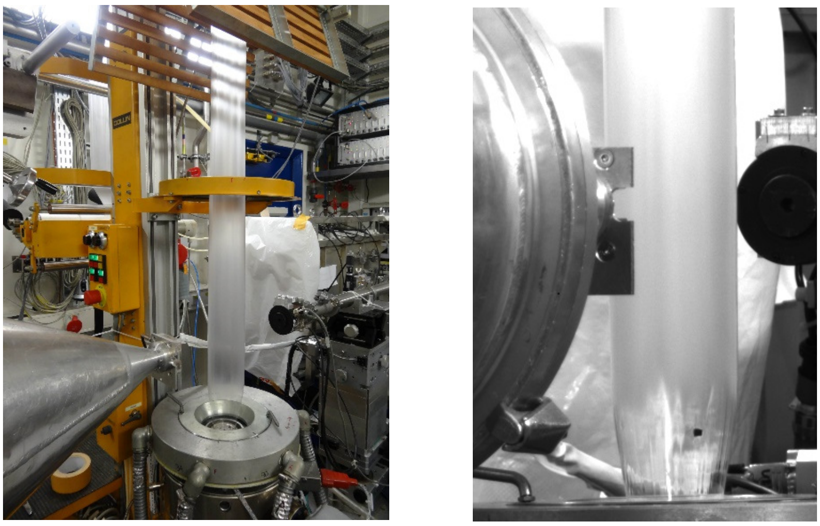

Figure 1: left: film blowing setup placed inside the experimental hutch of BM26B and right: bubble with particle for tracing the trajectory

Three materials (two LLDPE film blowing grades with different short side-chain branching content and one LLDPE blended with 10% of a long chain branched LDPE) were processed at 2x2 different conditions. Take-up ratio (speed of the rolls vs speed of the melt at the die) and blow-up ratio (final diameter of the bubble vs diameter of the die) were varied. The height of the die with respect to the beam was changed 9 times (4 -22 cm range). Time from the die was obtained from a particle tracing technique, a screenshot is plot in Figure 1 (right). This technique can also be used to determine the height of the frost-line, which is a measure for the melt-solid transition and is equivalent to the point where the speed of the film becomes stationary.

Using detailed analyses of the SAXS patterns obtained we can plot the long period, linear crystallinity and lamellar thickness (from auto-correlation function) versus distance and from the die, see Figure 2.
Ten patterns of ten seconds were acquired at each position, the average values are shown with standard deviation. These first results indicate that, as expected, the long period (Figure 2 left) decreases more rapidly when the applied stress is higher, i.e. higher BUR and/or TUR. Also the film thickness becomes thinner and higher cooling rates can be found, inducing faster crystallization kinetics (Figure 2 centre). The evolution of the lamellar thickness (Figure 2 right) is a strong function of the level of undercooling. With the maximum final film thickness, i.e. at minimum TUR and BUR, the cooling rate is low. Consequently, a slightly higher lamellar thickness values are obtained. The frostline is observed at approximately 50% of the final crystallinity (~30-40% from DSC), which indicates the crystallinity will further increase after the frostline by growth of spherulites. Moreover, secondary crystallization (local insertion of lamellae) may occur.

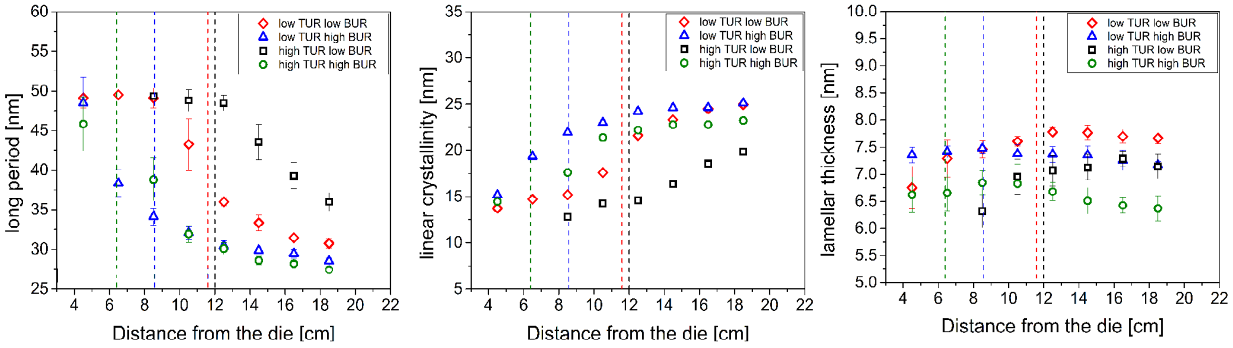

Figure 2: Results for LLDPE1 long period (a), linear crystallinity (b) and lamellar thickness (c) versus distance from the die, respectively

<u>Outlook</u>

- Data for the remaining two materials will be analysed. In addition, molecular orientation levels can be determined via Herman's orientation function.
- Samples produced during all experimental conditions are saved and will be tested in the near future using tensile tests. It is expected that the final lamellar thickness and orientation level will have a drastic effect on the stress-strain behaviour.
- The morphology of the final samples will be investigated using combined SAXS/WAXD.

From these experiments, which involve processing conditions relevant for industry, we can deduce in detail the effect of molecular architecture and conditions on structure evolution from the melt. The data is used as validation and/or input for film blowing models that are currently being developed in our group in Eindhoven. These models include material functions to describe the crystallization kinetics and compare this to our experimental results.

6. Are you planning follow-up experiments at DUBBLE for this project?
NO, not at this time. We obtained sufficient amounts of data to draw conclusions on this type of experiment. Changing the material is not an option at the moment.

7. Are you planning experiments at other synchrotrons in the near future?
NO

8. Do you expect any scientific output from this experimental session (publication, patent, ...)
YES, data analyses is still in progress for 2 of the materials. We expect to publish the outcome in a peer reviewed journal approximately Q3 2015.

9. Additional remarks
NO