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MORPHOLOGICAL ASPECTS OF CRACK GROWTH IN CRYSTALLINE POLYPROPYLENE

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The microscopic processes of crack growth in amorphous polymers have been clarified to a large extent in recent years [1]. In addition it is known that crack growth is modified by increased crystallinity in such a way that fatigue cracks propagate much more slowly [2]. Usually the mechanical data are correlated with the amount of crystallinity, as obtained by density measurements [3]. The purpose of this work is to find out whether there is an additional effect of morphology. Therefore a crystalline polymer was used, in which it was relatively easy to change the distribution of the crystalline zones in a wide range, mainly by different isothermal crystallization conditions. An attempt was made to investigate the crack paths in these different microstructures in order to find a correlation with macroscopic data of fracture toughness and fatigue crack propagation. The results should give information on the microstructure which leads to the best resistance to crack growth of a given polymer.

For this investigation an isotactic Polypropylene (Novolen PP 1120 LX, BASF, $Mn = 2 \cdot 10^5$) with a portion of 5% atactic polymer was used. A few supplementary experiments were carried out on a material with a content of 20% atactic polymer (Novolen PP 1320 L, BASF, $Mn = 2 \cdot 10^5$). Isotropic sheets of these materials were subjected to five different heat treatments which are graphically represented in Figure 1. The resulting microstructures are shown in Figure 2 and characterized in Table 1.

Mechanical tests were conducted with compact tension specimens for all these microstructures at ambient temperature. In addition fatigue experiments were carried out, the results of which will not be reported here. It may be mentioned, however, that the mechanical behaviour at high amplitudes corresponds to that of the toughness testing. The K_c-values for the five microstructures are shown in Figure 3. The microstructures have been arranged in a sequence of decreasing average values. A comparison with Table 1 indicates that there is no correlation with degree of crystallinity. Therefore a careful investigation of the morphological aspects of crack growth seemed to be promising.

Figure 4 shows three micrographs taken at the crack tip of the microstructures during the onset of crack propagation. Microstructure I which contains the finest dispersion of crystalline aggregates as well as microstructure III with very small spherulites (see Table 1) show craze bundles similar to those which are known from glassy polymers. The crack propagation is controlled by crazing. Its direction is exclusively normal to the applied load. Microstructure V demonstrates the other extreme. The crack essentially follows the boundary zones of the original spherulites. The local direction of the crack therefore deviates from the normal of the applied load up to more than 45°. Microstructure II is characterized by spherulites which are embedded in a matrix, which consists principally

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of microstructure I. Polygonal interfaces are rare. The crack propagation occurs as in microstructure I by crazing. It is, however, highly modified by the interaction with the spherulites. The easy crack path of microstructure V is only seldom available. A radial or tangential trans-spherulitic path is observed most frequently. The inner zone of the spherulite is usually avoided by the crazes and the cracks.

The scanning micrograph (Figure 5a) shows all different fracture features which occur during unstable crack propagation. They are interpreted in the schematic drawing Figure 6b.

In Figure 5b the influence of the higher atactic portion on crack growth is demonstrated. In this case localized plastic deformation in spherulites can occur. When the crack tip meets a boundary of a spherulite, a sliding process along the radial direction of the deformed spherulite is observed, indicating an inhomogeneity of plastic deformation in the material. It is likely that this process is found in the PP with lower atactic content at slightly elevated temperature.

According to our microscopic investigations and to other works on the structure and deformation of PP-spherulites in thin films [4 - 7] as well as in bulk material [8 - 11] different microstructural features can be classified, leading to specific resistance to crack propagation. The resistance values increase in the following sequence:

the polygonal interfaces of the former spherulites,
→ interfaces of spherulites with the fine crystalline matrix,
→ radial or tangential trans-spherulitic paths,
→ paths through the central zone of spherulite,
→ fine crystalline matrix.

In the case of microstructure V the particular weakness of the polygonal zone is also demonstrated by a deviation of $\alpha = 80°$ from the main direction. (see Figure 4c). Using this value for α and the relation

$$\frac{G_1}{G_2} > \frac{1}{\cos\alpha}$$

a ratio of nearly 5:1 for the trans (1)- to the inter (2)- spherulitic crack extension force follows.

In order to explain measured macroscopic fracture behaviour it seems reasonable to consider a partial resistance to crack growth of the microstructure elements. Under the condition that the plastic zone size is larger than the microstructure features (for example the diameter of the spherulites), the fracture toughness or the fatigue crack velocity should follow from these partial values and the particular volume portions of the microstructure elements.

If there is a great difference between the individual crack resistances of the possible crack paths, the crack will deviate highly from the normal direction in order to stay in the softer component. In this case the crack propagation will be determined exclusively by the crack resistance in this soft component and by a geometrical factor. The same selectivity is found when the plastic zone size is smaller than the dimension of the microstructure feature.

Details about the molecular processes during crack propagation and the influence of the orientation of the individual crystal lamellae on the direction of the individual crack paths in the spherulites can only be obtained by transmission electron microscopy. Experiments using this method are being carried out at present.

The results of this work indicate, that the degree of crystallinity does not completely characterize the mechanical properties of a polymeric material. In spite of similar crystallinity two specimens show different fracture behaviour for different morphologies.

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Table 1 The Degree of Crystallinity and the Mean Size of the Spherulites as Obtained by Different Thermal Treatments. The Degree of Crystallinity was Determined by Density Measurements

Morphology No.	I	II	III	IV	V
Degree of crystallinity (%)	60	66	67	71	76
Mean spherulite diameter (μm)	10	200*)	25	100	400

*) This value belongs to the coarse spherulites

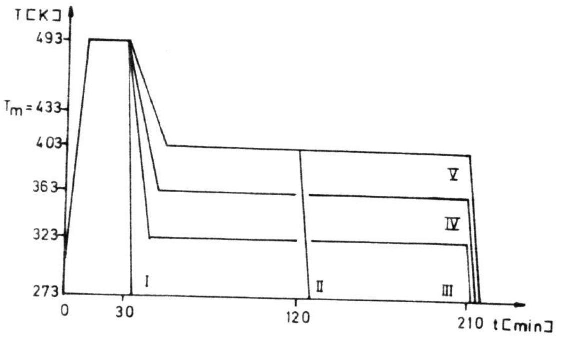


Figure 1 Thermal Treatment of Bulk Polypropylene Plates (50 x 50 x 3 mm^3). The Polymer Plates were Treated and Melted in a Closed Metal Box. After Melting the Material was Either Quenched to Room Temperature or Isothermally Crystallized at Different Temperatures for 90 or 180 Minutes

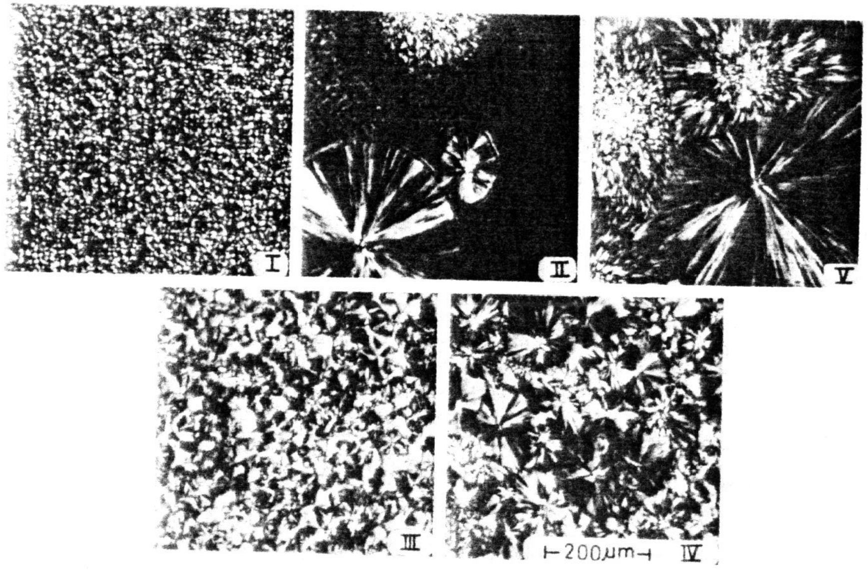


Figure 2 Transmission Light Micrographs of Five Microstructures, Resulting from Different Heat Treatments. Morphology I Consists of Many Spherulite Kernels Which Have Formed During Quenching from the Melt. In Morphology II Discrete, Coarse Spherulites (nearly 50 Volume %) are Embedded in a Matrix of the Type I. The Structures III - V Represent Complete Spherulitic Morphologies with Different Spherulite Diameters (See Table 1)

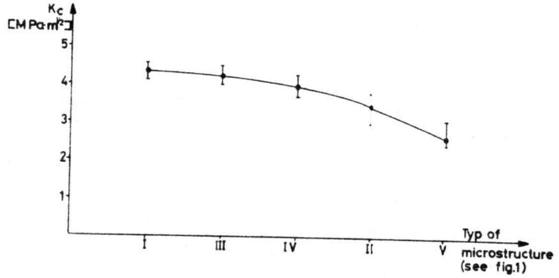


Figure 3 K_c - Values Obtained with Razor-Notched Compact Tension Specimens of the Five Different Morphologies and Arranged in a Sequence of Decreasing Values (Specimen Dimension 50 x 50 x 3 mm^3; Cross Head Speed v = 5 mm/min; Test Temperature T = 293 K)

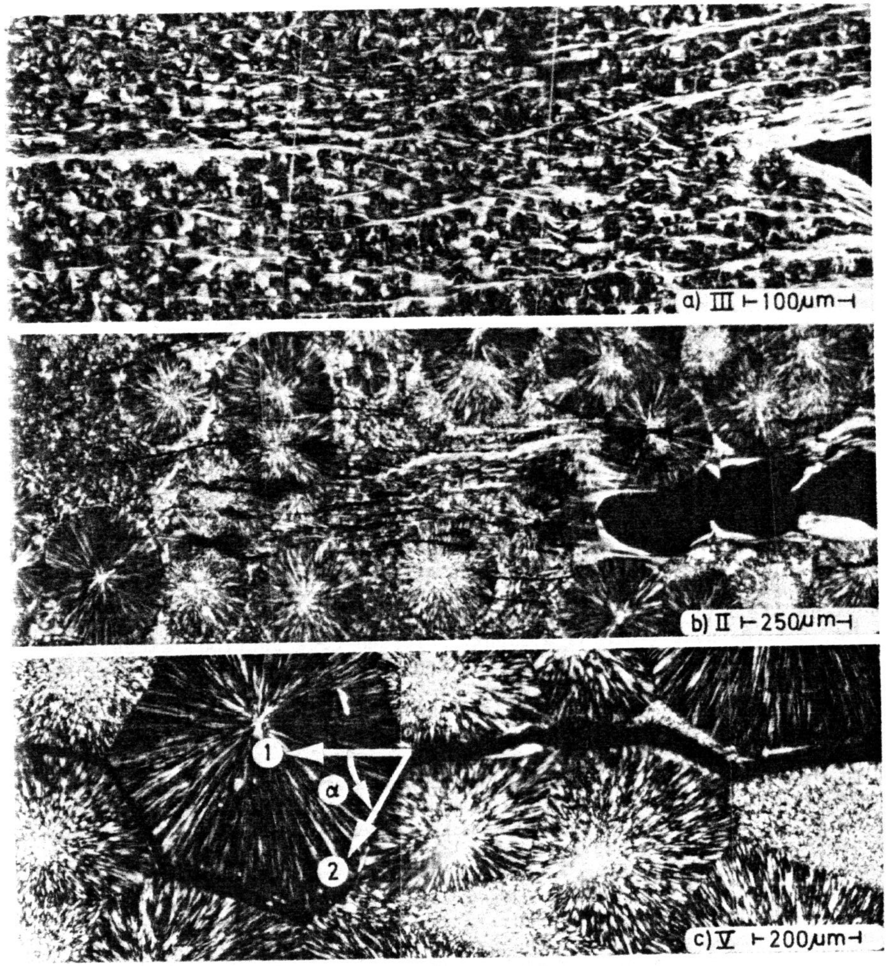
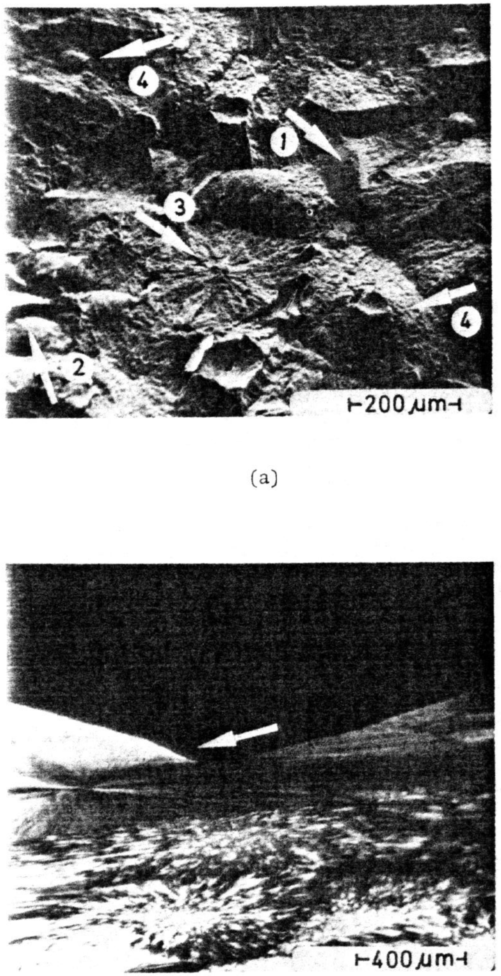


Figure 4 Microscopic Observation of the Crack Paths in the Morphologies III, II and V. In the Case of Microstructure V the Interspherulitic Crack Path Deviates up to α ≈ 80° from the Normal Crack Direction, Indicating that the Ratio of Trans- to Inter-Spherulitic Strength must be more than 5



(a)

(b)

Figure 5 (a) Scanning Micrograph of a Brittle Condition with Trans- and Inter-Spherulitic Fracture in PP 1120 LX (see Figure 6b)

(b) Radial Sliding Process in a Spherulite of a Polypropylene Film with a Portion of 20% Atactic Polymer. The Direction of the Applied Stress is Horizontal, the Arrow Indicates the Sliding Direction. In this case Separation Occurs by Localized Plastic Shear

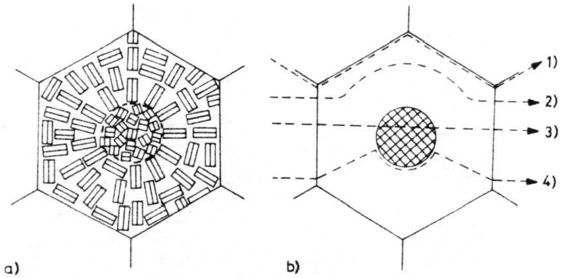


Figure 6 (a) Schematic Representation of Orientation of Crystal Lamellae in a PP-Spherulite. In the Central Region the Sets of Lamellae are Randomly Oriented (after [4]). In the Outer Zone, One Set of Lamellae is Radially Oriented, the Other is Approximately Tangentially Oriented (Crosshatched Structure after [5])

(b) Possible Crack Paths as Related to the Spherulite Structure. Path 1) Indicates the Inter-Spherulitic Crack Growth, Paths 2), 3) and 4) Show Tangential and Radial Crack Growth Through a Spherulite