

strain at the ends of the crystal, whether this consists of reentrant folds or of endgroups. If the entire crystal is folded at the end the size might be limited because of this. If the molecules are folded at the ends and re-enter the same rod, one wonders why the lamellae have a different characteristic fracture than the type I lamellae in which they are presumably also folded.

It is not known just when during the sample preparation process the kinks formed in the lamellae. One suspects that they are due to some increase in pressure following growth; possibly they may have formed during the fracture process. Deliberate attempts to induce their formation may help explain the deformation process in polymers. As in the case of polytetrafluoroethylene,^{7,17} the presence of these large morphological structures should simplify study of polymer deformation.

At the present time we cannot interpret the small-angle x-ray diffraction measurements listed in Table I. The values for samples C and D are in satisfactory agreement with the relative melting points but not with the lamella thicknesses observed in the electron microscope. Also, one wonders that the diameter of the rod-like crystals does not contribute to the diffraction pattern. The measured spacings were near the maximum resolution of the presently available camera; it is hoped to remeasure the samples in the near future. The relative melting points of C and D are also difficult to explain in view of the fact that the thicker type III lamellae, as observed in the microscope, are in the sample with the lower melting point.

It should be evident from this discussion that there remains a considerable area of research in this field of the structure of polymers crystallized under high pressure. It is believed likely that polymers other than polyethylene can also be crystallized in the form of these extended chain lamellae. The structure of the bands in polytetrafluoroethylene, which was first explained in terms of a structure like that discussed in possibility (1) above⁸ and then in terms of folded chains,^{7,21} is probably related to these type III lamellae.

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Résumé

La morphologie du polyéthylène linéaire cristallisé sous des pressions allant jusqu'à 5300 atmosphères a été examinée. Des micrographies électroniques des surfaces de rupture obtenues à partir de ces échantillons montrent que la majorité du polymère, aux pressions les plus élevées, cristallise sous forme de lamelles de chaînes étirées, qui peuvent atteindre une épaisseur de 3 microns. Des spectres de diffraction électronique montrent que les molécules sont normales par rapport aux lamelles. A des pressions plus basses une partie du polymère cristallise sous forme de lamelles à chaînes repliées, dont la proportion augmente avec une diminution de la pression. Des bandes de superposition bien définies peuvent être observées dans des lamelles à chaînes étendues plus épaisses. On suppose que le fractionnement du poids moléculaire ou bien un alignement des molécules bout-à-bout, suivi par un repliement des chaînes, a lieu pendant la croissance des lamelles à chaîne étirée.

Zusammenfassung

Die Morphologie von unter Drucken bis zu 5300 Atmosphären kristallisiertem linearen Polyäthylen wurde untersucht. Aus elektronenmikroskopischen Aufnahmen von an diesen Proben hergestellten Bruchflächen geht hervor, dass im Falle der höchsten Drucke der Grossteil des Polymeren in Form von bis zu 3 Mikron dicken, aus entfalteten Ketten bestehenden Lamellen kristallisiert. Elektronenbeugungsdiagramme zeigen, dass die Moleküle senkrecht zu den Lamellen stehen. Bei niedrigeren Drucken kristallisiert ein mit sinkendem Druck zunehmender Anteil des Polymeren in Form von aus gefalteten Ketten bestehenden Lamellen. In den dickeren, aus entfalteten Ketten bestehenden Lamellen konnten deutliche Knickbanden festgestellt werden. Es wird angenommen, dass während des Wachstums der aus entfalteten Ketten bestehenden Lamellen entweder eine Molekulargewichtsfractionierung oder eine End-zu-End-Ausrichtung der Moleküle und anschliessende Faltung auftritt.

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