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POLYMER CRYSTALS

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This Laboratory was established in October 1965, after the appointment of Dr. Kobayashi as a professor of this institute. The research in this laboratory is concentrated mainly in the field of structural and morphological studies of polymers in their solid state. The study of the relationship between the microstructure of a polymer and its physical properties is also within our field of research.

The main installations are various kinds of X-ray diffraction apparatus and electron microscopes. After the development of two ultra-high voltage electron microscopes (300 KV and 500 KV), an ultra-high resolution electron microscope with an extremely low temperature specimen chamber (1.4 Å, 4°K, 500 KV) was constructed in May 1974.

Some recent research projects carried out in this laboratory are as in the following.

I. Observation of Molecular Images by "HAREM"

We have built a "HAREM" (High Atomic Resolution Electron Microscope) with 1.4 Å resolution in May 1974, in cooperation with Prof. Uyeda. With this instrument, we have obtained an electron micrograph of chlorinated Cu-phthalocyanine molecule, which clearly showed a clover leaf composed of benzene rings with chlorine atoms attached to them.

II. Influence of Lattice Defects on the Structure and Properties of Polymers

The melting point of electron irradiated polyetylene crystal decreases and the lattice constants (both a and b) and lattice distortion increase, but the macroscopic volume of the specimen does not change with electron irradiation dose. Calculations indicate that the anisotropy of a polymer crystal (difference between the elastic moduli along and perpendicular to the molecular axis) increases the range of the distortion in a crystal caused by a defect. Very recently, the lattice disorder of ethylene-propylene random copolymer crystals which are considered to contain methyl groups as positive defects (in contrast to the negative defects induced by electron irradiation) was investigated. It is shown that the positive defects cause considerable amount of distortion of the first kind whereas the negative defects cause only the distortion of the second kind.

III. Study of the Entity of So-called "Amorphous Region"

The intensities of X-ray small angle scatterings from hot drawn high-density polyethylene specimens under the strains within elastic limit are measured. The

changes in the intensities and the long periods with the strains lead to the conclusion that the density of interlamellar region is considerably lower (about 30%) than the density of the melt extraporated to room temperature.

IV. Determination of the Setting Angle of Molecular Chain in Polyethylene Crystals

The setting angle of molecular chain in various polyethylene specimens were determined by means of electron diffraction and optical transform. The setting angle decreases from about 49° to 41° with an increase in the fold content of the specimen.

V. Mechanism of the Increase in the Crystallization Rate under Shear Stress

The crystallization kinetics of molten polyethylene subjected to a constant shear stress was investigated theoretically and experimentally. The rate of crystallization depends on the difference of entropy (Δs) between the crystalline and molten state. The constant shear stress in the melt decreases Δs causing an abrupt increase in the rate of crystallization and a decrease of the thickness of the folded-chain lamellar crystals. The process of structural formation during oriented crystallization is being followed by X-ray and light scattering. An X-ray television system for the direct observation of the crystallization process is now in use.

VI. Nonisothermal Crystallization of Polymers

The phenomenon of nonisothermal crystallization of polymers was investigated, both theoretically and experimentally. The fundamental equations were established for predicting the changes in temperature and crystallinity during the nonisothermal crystallization, and applied to the analysis of the melt spinning process.

VII. Prediction of Molecular Orientation during Elongational Melt Flow

The crystallization during melt spinning is considered to be a nonisothermal crystallization under constant tension. Molecular orientation just before the onset of crystallization determines the microstructure of spun fibers. For a thorough prediction of the process of melt spinning, an analysis of melt flow is required from both the rheological and structural aspects. Isothermal melt spinning experiments were carried out and the birefringence and flow properties were analyzed by using a Jeffreys model.

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