A New Very Large Long-period in the X-ray Small-angle Diffraction of Polyethylene Filaments

By Takani Mochizuki

(Received March 25, 1959)

The meridional long-period X-ray diffraction in polyethylene was measured by several authors[1-4]. However the reported long-period spacings are divergent. For instance, a spacing given by Kiessig[2] is 98 Å, whereas that by Statton[3] is 240 Å. These discrepancies might be due to the difference in polyethylene specimens and also their preparations.

This communication is to report that a further large long-period spacing has been observed recently in the meridional small-angle scattering for low-pressure polyethylene filaments.

Polyethylene ("Fortiflex A" of Celanese Corp. of America; Melt index: 0.7) filament specimens were prepared with an extruder at 210°C (Sample A), and stretched at 140°C up to 100% (Sample B), 200% (Sample C) and 300% (Sample D). From the X-ray diffraction photographs, a fairly high orientation of the crystallites was seen for Sample A and a typical uniaxial orientation of the crystallites was observed for Sample D. The small-angle scatterings of X-rays from these samples were

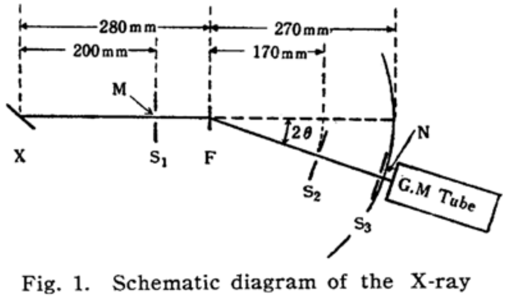

Fig. 1. Schematic diagram of the X-ray diffractometer.
X: X-ray focus
S_1: 1st slit 0.15 mm.
S_2: 2nd slit 0.5 mm.
S_3: 3rd slit 0.1 mm.
F: filament sample
M, N; Soller slit

1) E. P. H. Meibohm and A. F. Smith, *J. Polymer Sci.*, 7, 449 (1951).
2) K. Hess and H. Kiessig, *Kolloid-Z.*, 130, 10 (1953).
3) W. O. Statton, *J. Polymer Sci.*, 28, 423 (1958).
4) I. Sakurada, Y. Nukushina and Y. Tanaka, *Chem. of High Polymers*, 15, 771 (1958).

measured in the direction parallel to the fiber axis with an X-ray diffractometer having the following slit system, shown in Fig. 1, Cu $K\alpha$ radiation (38 KV. 15 mA) filtered with nickel foil being used.

Curves of the diffracted X-ray intensity are shown Fig. 2. All samples show a very strong and distinct peak of intensity. The center of the peak for Samples A and D is found using Bragg's law to correspond to a spacing of the order of 260 Å and 460 Å, respectively. Such a large long-period of about 460 Å has not been observed previously for synthetic polymers. From the intensity curves of Samples B and C, it seems that two kinds of long-period (260 Å and 460 Å) coexist in these samples.

Further, high-pressure polyethylene ("Tenite #864" of Eastman Chem. Products; Melt index: 0.3) was examined for comparision. Filaments were prepared in the same way as above, and stretched up to 270% at 100°C (Sample E). The intensity curve of the meridional small-angle interference of Sample E is given by the dotted line in Fig. 2, which shows a broad and low intensity peak. The Bragg distance of this peak is calculated to be 177 Å,

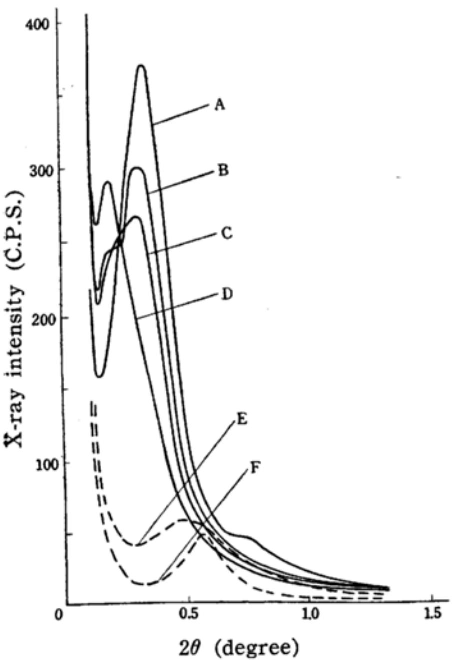

Fig. 2. Intensity curves of small-angle X-ray scattering.
A: original filament
B: 100% stretched filament
C: 200% stretched filament
D: 300% stretched filament
E: high-pressure polyethylene filament
F: polyvinyl alcohol fiber

which is in agreement with the value given by Meibohm and Smith[1] and Sakurada, Nukushina and Tanaka[4].

Some points of interest come to light in this experiment. In the low-pressure polyethylene, a very strong small-angle interference was observed as compared with the intensities obtained for high-pressure polyethylene and polyvinyl alcohol fibers[5]. It seems likely that this marked difference in the intensities might be partly due to the difference between the mean electron densities of the crystalline and amorphous parts, and has something to do with similar difference, shown in Fig. 3, in intensities of the meridional reflections (0 2 0) from both samples[5].

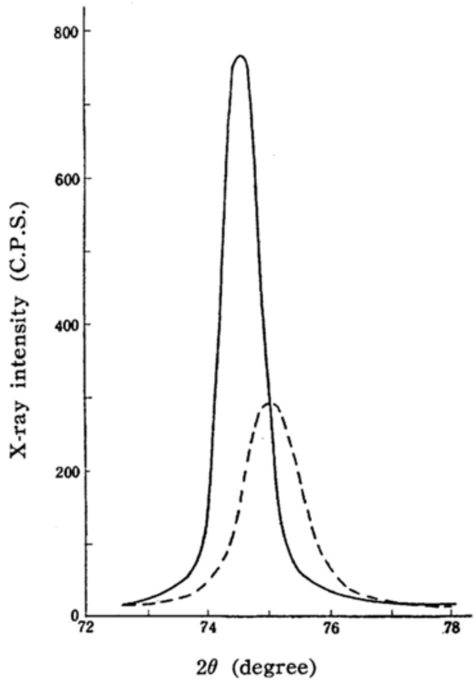

Fig. 3. Intensity curves of (0 2 0) reflection —— polyethylene ------ polyvinyl alcohol

Besides, it is to be noted that the curve A shows a second intensity hump. It corresponds to the second order reflection of the spacing of 260 Å. Such small intensity hump has not been observed in the curves B, C and D, and hence, it seems that the regular fine texture is destroyed by stretching process.

The author expresses his sincere thanks to Professor I. Nitta, Osaka Univercity, for his kind guidance and encouragement

5) The detailed results of polyvinyl alcohol fibers will be reported elsewhere.

throughout this experiments. He is also indebted to Mr. S. Miyazaki for supplying the samples.

Research Laboratory
Kurashiki Rayon Co., Ltd.
Kurashiki