

Characterization of Structural Knot Distributions in UHMWPE Fibers^{*}

Hao-jun Xu^a, Min-fang An^a, You Lv^a, Zong-bao Wang^{a**} and Qun Gu^b

^a Ningbo Key Laboratory of Specialty Polymers, Faculty of Materials Science and Chemical Engineering, Ningbo University, Ningbo 315211, China

^b College of Material Engineering, Ningbo University of Technology, Ningbo 315211, China

Abstract Microbeam wide-angle X-ray diffraction (WAXD) experiments were carried out at different structural knot positions of SIOC and M4 fibers of ultra-high molecular weight polyethylene (UHMWPE). The optical microscope images revealed that SIOC fiber had bamboo-like structural knots, and M4 fiber had chaotic distribution of structural knots. WAXD patterns showed the monoclinic unit cell in the whole M4 fiber, but different lamellar orientations in the bamboo joint of SIOC fiber. In addition, small-angle X-ray scattering (SAXS) patterns confirmed that the SIOC fiber contained uniform distribution of shish structures, and differential scanning calorimetry (DSC) measurements showed that its less branched and short chains benefited the orthorhombic-hexagonal phase transformation.

Keywords: Microbeam X-ray diffraction; UHMWPE fiber; Branched chain; Structural knot; Orientation distribution.

INTRODUCTION

UHMWPE fibers produced by gel-spinning method have excellent performance such as high strength, high modulus, impact and cutting resistance^[1]. UHMWPE fibers have semicrystalline structure and are composed of crystallites together with more or less structural knot because of elastic flow instabilities^[2] in the production processing and the existence of UHMWPE slightly branched long chains. Several studies were performed to investigate the effect of defect on the properties of specific fibers^[3, 4]. Strawhecker *et al.*^[5] found the defect appeared to be an amorphous region forming an inclusion on UHMWPE surface using AFM. There is, therefore, a strong need to ascertain the connection between the structural defect and the quantity of branched chains in the same technological parameter.

Microbeam X-ray diffraction experiments (beam size is down to several microns) at synchrotron sources can be used feasibly for structural characterization at different area of single fiber^[6–9]. In contrast to laboratory microbeam WAXD experiments on single fiber requiring a few hours per image, synchrotron sources diffraction experiments only require a few seconds per image^[10–12]. The microbeam diffraction using beam diameter of approximately 2 μm was employed to yield position-resolved maps in UHMWPE and viscose fibers. Riekel *et al.*^[13] found that preferred orientation was observed for an UHMWPE fiber where orthorhombic plus monoclinic phases were found to coexist in the outer parts of the fiber. Müller *et al.*^[14] revealed that the cellulose molecules were far better aligned in the fiber skin than in their core and there was no significant difference between skin and core concerning both mean crystallite size and crystallinity. Schulte *et al.*^[15] provided detailed

^{*} This work was financially supported by the National Natural Science Foundation of China (No. 51273210), the Open Research Fund of State Key Laboratory of Polymer Physics and Chemistry, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences and the K.C.Wong Magna Fund in Ningbo University.

^{**} Corresponding author: Zong-bao Wang (王宗宝), E-mail: wangzongbao@nbu.edu.cn

Received November 14, 2015; Revised December 15, 2015; Accepted December 22, 2015

doi: 10.1007/s10118-016-1774-8

structural information of the transcrystalline interphase developing at the boundary between the UHMWPE fiber and the HDPE matrix using microbeam X-ray diffraction measurements.

Melt processing of polyethylene produces partially crystalline structure with folded-chain crystallites imbedded in less ordered polymer matrix^[16]. Gel-spinning UHMWPE fibers were reported to have a highly oriented and extended chain crystalline structure with limited disordered domains in the interfibrillar and intercrystalline regions^[17, 18]. In the microfibrils, the long crystal blocks were connected by short disorder domains^[19]. These disordered domains were thought to contain physical entanglements, taut tie molecules (TTM), chain ends, and other imperfections.

In previous research, the influences of branched chains on UHMWPE chains in the melt processing and the structural knot distribution in single UHMWPE fiber have not been involved. And the existence of branched chains is directly related to the mechanical properties of UHMWPE fibers. In the present work, we investigated the structures of two different UHMWPE fibers with different quantities of branched chains using X-ray microbeam diffraction, differential scanning calorimetry and small angle X-ray scattering.

EXPERIMENTAL

Materials

UHMWPE resins of M4 and SIOC were provided by Sinopec Beijing Yanshan Company and Shanghai Institute of Organic Chemistry, respectively. The viscosity-average molecular weights were determined for polyethylene as $M_v = 3.5 \times 10^6$ and 2.1×10^6 . The SIOC resin contained less than 1 branched chain per 100000 carbon atoms (specific data can not be collected due to the instrument measure limit) and the M4 resin had 7 branched chains. The two different UHMWPE resins were used to produce fibers with the same gel-spinning method. 8 wt% UHMWPE solutions in paraffin oil were prepared at 150 °C. Upon cooling this solution a gel formed and was fed to the spinning apparatus. The gel was extruded into filament at 245 °C with an extrusion rate of 8 m/min, using a conical die with an exit of 1 mm. The paraffin oil was extracted from filaments with dichloromethane. Afterwards, three-step hot drawing to 20 draw ratios was carried out at the temperature of 110–130 °C.

Analytical Methods

Tensile test of M4 and SIOC fibers with no twisting was performed on an Instron 5567 tension testing machine at a tensile rate of 250 mm/min, and the fiber initial gauge length was 500 mm.

A single fiber isolated from a fiber bundle was selected under microscope and fixed across a metal ring for X-ray microbeam diffraction experiment. After the experiment, the fiber was glued to a metal sample stage for scanning electron microscope (SEM) after shadowing with gold. The morphology of single UHMWPE fiber was observed by a Hitachi S-4800 field emission scanning electron microscope operated at 4.0 kV.

The X-ray microbeam diffraction experiment was carried out on beamline BL15U at Shanghai Synchrotron Radiation Facility (SSRF). The photon source of bending magnet of SSRF delivers X-rays of 5 keV to 20 keV. The experiment system uses a pair of Kirkpatrick-Baez (K-B) mirrors (Xradia Inc.) as high resolution X-ray focusing optics exhibiting a high sensitivity for structure detection and allowing image exposure with a reasonable dwell time. A monochromatic beam of wavelength $\lambda = 0.068$ nm was obtained by a monochromator. The beam diameter at the sample position was estimated to be about $3.5 \mu\text{m} \times 3 \mu\text{m}$ (horizontal $\times$ vertical), which was determined by the knife-edge technique using a thin tantalum blade. The intensities of the incident and transmitted X-ray beam were monitored by two ionization chambers positioned upstream (I_0) and downstream (I_1) of the sample, respectively^[20]. Fiber samples were placed on a high precision 7-axis stage and can be X-ray scanned in three translation directions (x , y and z). An optical microscope allowed positioning of the sample in the X-ray microbeam. An image-intensified MarCCD detector with video-readout was used (2048×2048 pixels of 319×319 DPI). The sample to detector distance was calibrated with 204 mm. The exposure time was 4 s per scan point.

Differential scanning calorimetry (DSC) measurements were performed with a Mettler Toledo DSC instrument under nitrogen atmosphere with a flow rate of 10 mL/min. The melting of UHMWPE fibers was

measured with 3 to 5 mg fragment fibers and a heating rate of 10 K/min in aluminum pans.

The small angle X-ray scattering (SAXS) measurement was carried out on beamline BL16B at SSRF. A Si(III) flat double crystal monochromator (DCM) and a double focusing toroidal mirror were used in the beamline optics. The X-ray wavelength was 0.124 nm. Two dimensional SAXS patterns were collected by using a MAR CCD X-ray detector (2048×2048 pixels of 322×322 DPI). The beam intensity monitor before sample was a N_2 gas ionization chamber, and the monitor after sample adsorption was a photodiode in the beam stop. Two scatter-less slits (Xenocs) were used to depress parasitic scattering. The sample holder was mounted onto an optical table. The distance of sample to SAXS detector was adjusted to 5.08 m, scattering from a sample transmits in vacuum, passing Kapton window to hit the detector^[21].

The X-ray measurement data analysis (distance calibration, background correction, radial and azimuthal integration) was carried out using the Fit2d software package^[22].

RESULTS AND DISCUSSION

Mechanical Performance

The tensile properties of M4 and SIOC fibers are shown in Table 1. The tensile strength and Young's modulus of SIOC fiber can reach 26.25 and 1124 cN/dtex, respectively, which are 3.8% and 9.0% higher than those of M4 fiber. The less branched and short chain fiber shows better mechanical performance.

Table 1. The tensile properties of M4 and SIOC fibers

Sample	Tensile strength (cN/dtex)	Young's modulus (cN/dtex)
M4 fiber	25.29 ± 1.01	1031 ± 24
SIOC fiber	26.25 ± 1.15	1124 ± 35

Morphology

In Fig. 1, which is a typical SEM image, the fiber has a diameter of 38 μm . The surface was characteristic for UHMWPE fiber possessing a smooth skin layer. The fiber was placed on a high precision x/z (as shown in Fig. 1, the y-axis was just for X-ray focusing) translation stage. Here, the fiber was positioned in the center of a high resolution optical microscope with precisely known location to the X-ray beam. With this calibration, it was possible to scan reproducibly from the center to edge. Perpendicular to the fiber axis (along the x-axis), the step size was 5 μm from fiber center to edge. In order to ensure the results accuracy, it was repeatedly scanned with an interval 20 μm along the z-axis. Each row of data points was marked as row a, b, c, d, e. All scanning X-ray diffraction data points were identified with specific positions shown in Fig. 1.

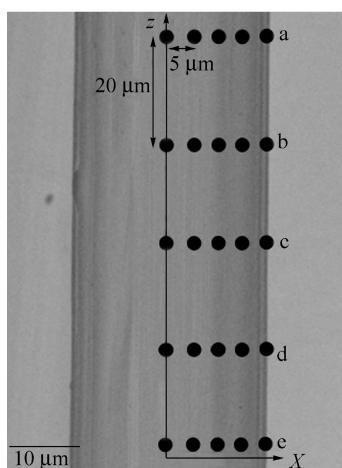


Fig. 1 SEM image of a single UHMWPE fiber (The black points were identified for the specific scanning X-ray diffraction data points.)

Figure 2 shows the optical microscope images of M4 and SIOC fibers. The inset images are magnified views of the black rectangles. The cross-shaped mark is used for locating the position of X-ray microbeam in the view of the optical microscope. The M4 fiber has chaotic structural knot distribution and SIOC fiber has more like a bamboo-like structural knot distribution. M4 fiber is filling with structural knots in the whole fiber, but SIOC fiber contains smaller percentage structural knots just in the bamboo-like joints (highlight section). It is generally believed that the formation of structural defects is correlated with the structure of gel fibers and the structural evolution of dried fibers during the hot drawing process. For the structural knot in UHMWPE fibers, disentanglement of long chains has the major effect. The disentanglement is difficult during the hot drawing process since the molecular weight of sample is too large and the drawing temperature can not be too high, therefore the disentanglement generally occurs during the dissolve process. the SIOC resin containing less branched and short chains is more easily disentangled than M4 resin with the same dissolve condition. Therefore SIOC fiber contains less structure knot than M4 fiber. In the inset image, the white points are the X-ray diffraction positions corresponding to Fig. 1 (just mark the fiber's core and skin).

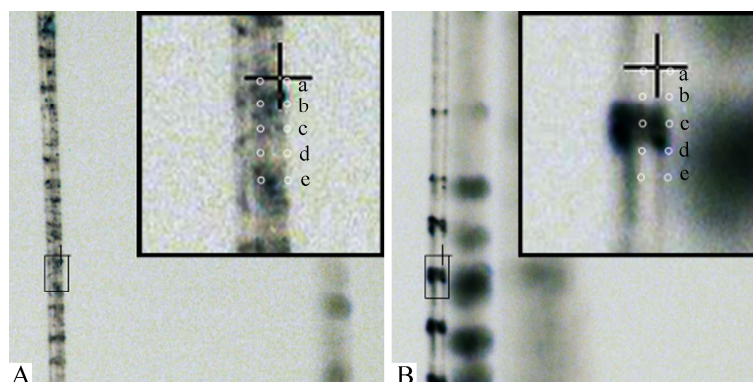


Fig. 2 Optical microscope images of M4 (A) and SIOC (B) fibers (The inset images are magnified views of the black rectangles.)

Crystal Structure

Figure 3 illustrates the schematics of the microbeam WAXD experiments together obtained from core to skin positions of the two kinds of fibers. As mentioned in the previous section, the diffraction patterns of M4 and SIOC fibers (left patterns a1, b1, c1 for fiber core, right a5, b5, c5 for skin as shown in Fig. 2) give evidence for significant structural differences. The two-dimensional patterns show the variation of diffraction intensity between the core and skin because the core has a much thicker thickness. Also there are different lamellar orientations at the structural knot part (see Fig. 3c1) comparing with the normal part (see Fig. 3b1) in the SIOC fiber.

Figure 4 shows the intensity distribution function versus 2θ values obtained from the cake integration (-2° — 2°) of the two-dimensional WAXD patterns of Figs. 3(a) and 3(c). It is noteworthy that the angular positions of the diffraction peaks do not vary with the location of the X-ray beam. Hence, the lattice cell parameters in the core or in the skin remain constant. All integrated intensity distributions indicate the core intensity is stronger than the skin one. The reason for this is that the diffraction pattern from the fiber center always represents superposition of the signals from core and skin. All patterns show the orthorhombic unit cell, as $(110)^{\text{Orth}}$ and $(200)^{\text{Orth}}$ reflections are detected, and the monoclinic unit cell, as $(010)^{\text{Mono}}$ reflection appears, with the whole M4 fiber. This may be due to a band of monoclinic phase extending around the circumference of the fiber. The amount of monoclinic crystals is much smaller than the amount of orthorhombic crystals. The monoclinic structure is, however, not distributed as a thin layer on or between the surfaces of orthorhombic crystallites^[23]. Since the monoclinic signal in microbeam WAXD does not show an observable increase from M4 fiber's core to skin, the monoclinic phase seems not to melt. It is usually assumed that the monoclinic domains

transform to the orthorhombic phase by rotation of the chains^[24]. Furthermore, comparing with SIOC, the M4 fiber has more branched chains and exhibits chaotic structural knot distribution (see Fig. 2A). As to the SIOC fiber, no monoclinic reflection is found both in the core and in the skin. This result is in disagreement with previous microdiffraction studies on a single UHMWPE fiber, which suggests that the monoclinic phase mainly appears at the fiber skin^[13]. The explanation is that the appearance of monoclinic phase is mainly due to the existence of branched chains at the main UHMWPE chains during hot drawing processing, which mainly has an influence on the transformation of orthorhombic into monoclinic polyethylene^[25].

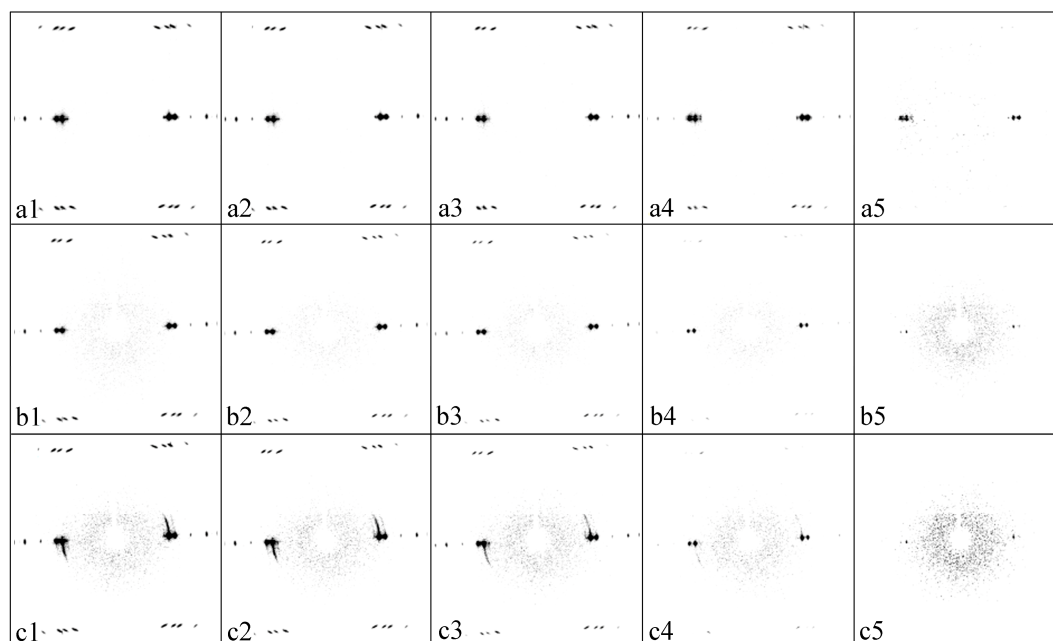


Fig. 3 Two-dimensional microbeam WAXD patterns of UHMWPE fibers: (a1–a5) M4 fiber, row a, (b1–b5) SIOC fiber, row a, normal part, (c1–c5) SIOC fiber, row c, bamboo-like joint (The fiber axis is vertical.)

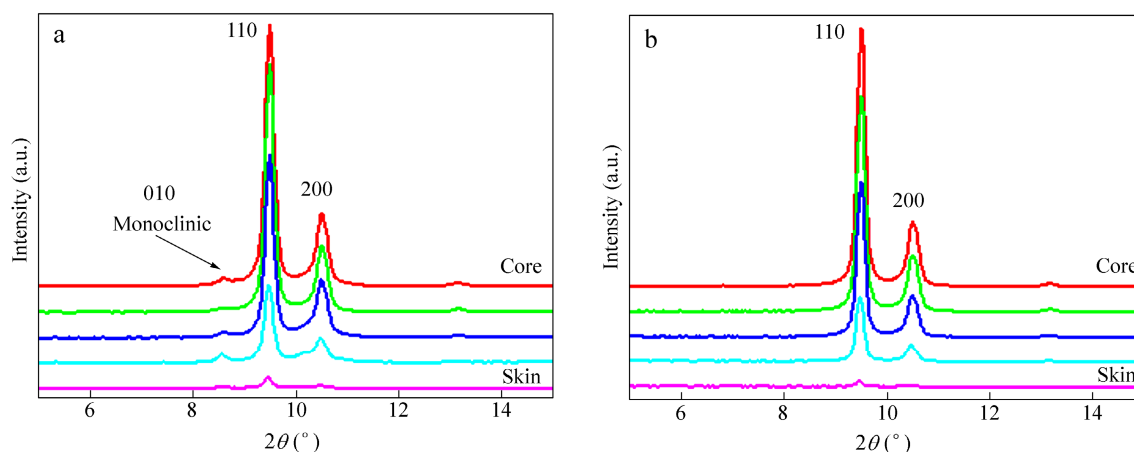


Fig. 4 Intensity distribution functions versus 2θ values corresponding to the M4 (a) and SIOC (b) fibers. The integrated intensity distributions are from two-dimensional diffraction patterns Fig. 3(a) and 3(c).

Crystallite Orientation

For a quantitative analysis of the orientation observed in the two-dimensional microbeam WAXD patterns of Figs. 3(a) and 3(c), azimuthal scanning of the $(110)^{\text{Orth}}$ reflection was obtained. Figure 5 shows the azimuthal

intensity profiles of the $(110)^{\text{Orth}}$ reflection corresponding to the diffraction patterns from fiber core to skin. It can be clearly seen that the diffraction patterns have a protuberance near 180° (corresponding to the two-dimensional pattern, Fig. 3c1). As to the less branched and short chain (SIOC) fibers, the structural knot caused by specific production processing just leads to the different lamellar orientations but no transformation of orthorhombic into monoclinic polyethylene.

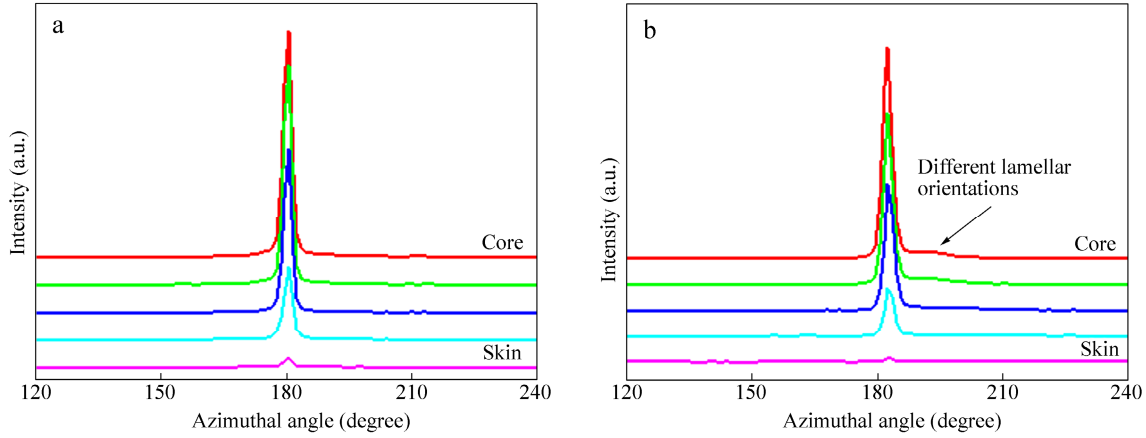


Fig. 5 Azimuthal intensity profiles of the $(110)^{\text{Orth}}$ reflection corresponding to the M4 (a) and SIOC (b) fibers. The integrated intensity distributions are from two-dimensional diffraction patterns Fig. 3(a) and 3(c).

The values of the degree of orientation associated with the lattice planes hkl can be derived from the azimuthal intensity distribution functions (Fig. 5) by using Polanyi equation:

$$\cos \phi_{hkl} = \cos \theta_{hkl} \cos \psi \quad (1)$$

where θ_{hkl} denotes the Bragg diffraction angle. The equation correlates the azimuthal angle ψ along the Debye circle with the angle θ_{hkl} between the fiber axial direction and the normal vector of the lattice plane. And the orientational parameter $\cos^2 \phi_{hkl}$ can be obtained by:

$$\cos^2 \phi_{hkl} = \frac{\int_0^{\pi/2} I_{hkl}(\phi) \cos^2 \phi_{hkl} \sin \phi_{hkl} d\phi_{hkl}}{\int_0^{\pi/2} I_{hkl}(\phi) \sin \phi_{hkl} d\phi_{hkl}} \quad (2)$$

So a measure for the degree of orientation in uniaxially oriented samples is the orientational parameter proposed by Hermans^[26]. It is expressed as follows:

$$S_{hkl} = \frac{3 \langle \cos^2 \phi_{hkl} \rangle - 1}{2} \quad (3)$$

Note that the degree of orientation in the present case assumes values in the range $0 < S_{hkl} < 1$. For a perfect orientation of the lattice planes with their normal parallel to the fiber axial direction, the degree of orientation would be $S_{hkl} = 1$, while for an isotropic sample in the randomly oriented state, the degree of orientation would be $S_{hkl} = 0$. Usually, the degree of orientation lies between the above two limiting values.

The degree of orientation distributions is given in Fig. 6 at the five positions (5 μm lateral separation from core to skin) and five rows on the fiber schematically depicted in Fig. 1 and Fig. 2. As to the M4 fiber (Fig. 6A), rows c, d, e have reasonable orientation distributions increased consequently from 0.40 (core) to 0.72 (skin) but rows a, b have superior orientation distributions at the region of fiber core, which can be assumed that they may have alignment structure around the knot structure regions in the branched chain fiber. As to the SIOC fiber (Fig. 6B), row c which is in the bamboo-like structural joint (see Fig. 2B) shows a slight lower orientation

distribution than rows a, b, c, d. The whole fiber has orientation distributions increasing from about 0.70 (core) to 0.95 (skin) because of the less branched and short chains. In principle, the orientation distributions of SIOC fiber are better and more regular than those of the M4. And the skin/core structure of SIOC fiber is in agreement with previous microdiffraction research on single viscose fiber, which suggests that the skin has much higher orientation of the crystalline regions and higher alignment of the polymer chains^[14].

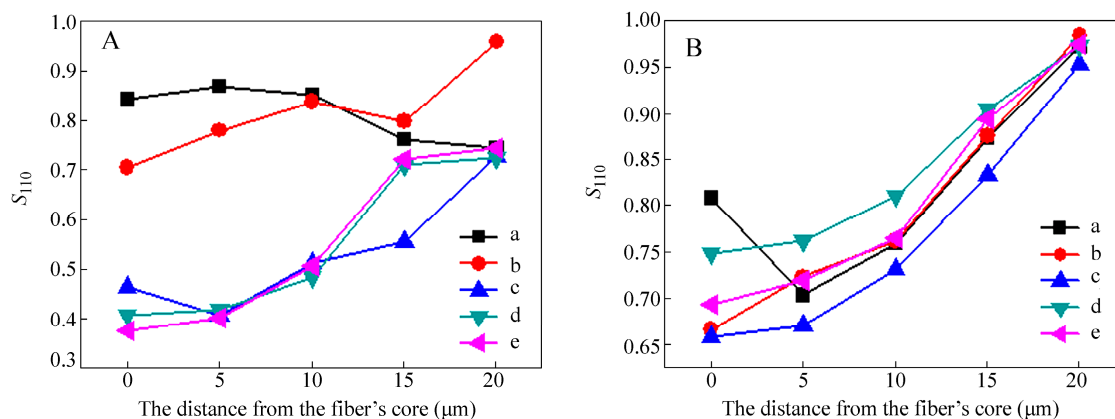


Fig. 6 Position dependent orientational parameters obtained from azimuthal intensity profiles of the (110)^{Orth} reflection corresponding to the M4 fiber (A) and SIOC fiber (B)

Melt Processing

Melting of UHMWPE fibers begins at around 138 °C and ends at around 164 °C (Fig. 7). Stanislav *et al.*^[27] founded that melting of the monoclinic phase was completed by 135 °C. As to the SIOC fiber, the temperature endotherm observed at about 147 °C is the main melting peak, which is little lower than that of the M4. It is followed by a small peak at about 159 °C. Absence of these melting endotherms from DSC of the SIOC fiber suggests fibrillar structure with interfibrillar stress and constraint of the highly crystalline matrix containing large and extended chain crystals. The much elevated temperature (159 °C) endotherm is from melting of the hexagonal crystalline structure and possibly from randomization of chains to the gauche state in the melt^[28]. The reported solid-state orthorhombic-hexagonal phase transformation occurs following partial melting at the temperature range. The M4 fiber DSC shows a single melting endotherm without the orthorhombic-hexagonal phase transformation which indicates the existence of branched chains significantly reduces or eliminates the crystal transformation from orthorhombic phase to hexagonal phase. If the heat of fusion value of 293 J/g is used for the equilibrium crystals of polyethylene^[29], the crystallinities of M4 and SIOC fiber are 79.6% and 88.0%, respectively.

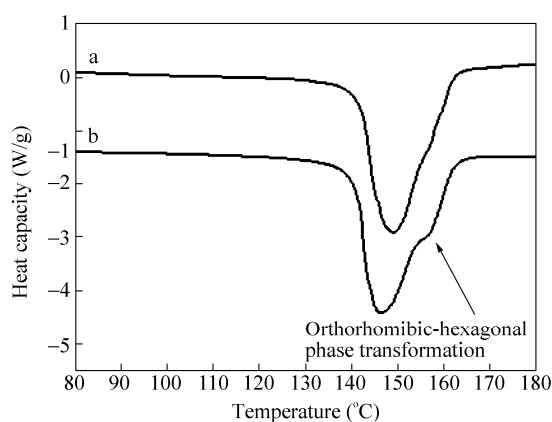


Fig. 7 DSC melting of two UHMWPE fiber samples: (a) M4 fiber and (b) SIOC fiber

Shish Structure

Selected two-dimensional SAXS patterns are illustrated for M4 and SIOC fibers. Strong equatorial streaks appear for both samples, as indicated by the arrow in Fig. 8(a). The appearance of the equatorial streak is consistent with the formation of shishes (In fact this shish structure is the extended chain crystal which has similar structure with shish) having strong electron density contrast with the surrounding matrix which may be related to the multiple shish in the UHMWPE fiber^[30]. It can be clearly seen that the SIOC fiber's equatorial streak is stronger than the M4s, which may imply the SIOC fiber has better shish structure.

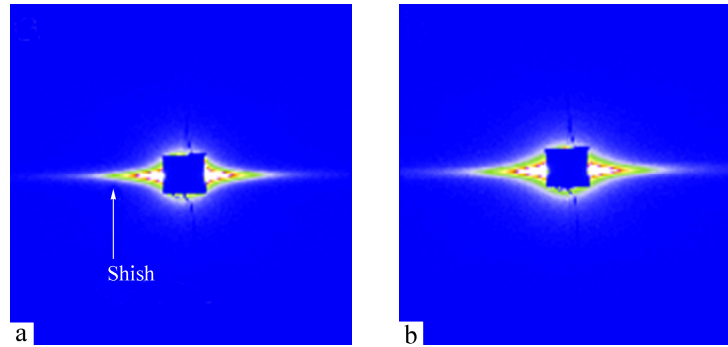


Fig. 8 Two-dimensional SAXS patterns of a UHMWPE fiber: (a) M4 fiber and (b) SIOC fiber

In order to investigate the difference of the shish structure between M4 and SIOC fibers, the average shish length, L_{shish} , and the misorientation of shishes, B_{ϕ} , are calculated in the following section. These parameters were obtained by using the Ruland streak method to analyze the equatorial streak feature in SAXS^[31–33]. Ruland demonstrated that the size and orientation distributions of longitudinal voids in polymer and carbon fibers in real space could be estimated from the equatorial streak of SAXS in reciprocal space. Since the method is principally based on the separation of experimentally measured azimuthal breadth from contributions of scatterer length and misorientation, the method can also be applied to calculate the average length of shish and its average misorientation (as shown in Fig. 9).

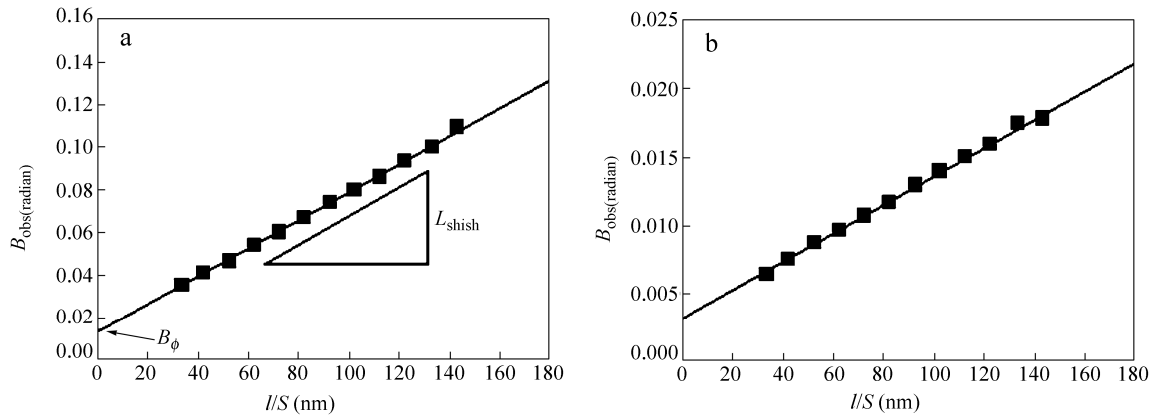


Fig. 9 Plots of azimuthal integral width B_{obs} versus the value of $1/S$, which is used to determine the average shish length and the shish misorientation for (a) M4 fiber and (b) SIOC fiber

It is assumed that all azimuthal distributions can be modeled by Lorentz functions, the observed azimuthal width, B_{obs} , can be related to the length of shish, L_{shish} , and the azimuthal width, B_{ϕ} , due to misorientation of shish by the following equation:

$$B_{\text{obs}} = \frac{1}{L_{\text{shish}} S} + B_{\phi} \quad (\text{Cauchy-Cauchy}) \quad (4)$$

where B_{obs} represents the integral width of the azimuthal profile from the equatorial streak at S , and S is the scattering vector, $S = 2(\sin\theta)/\lambda$, where θ being the scattering angle and λ being the X-ray wavelength. On the basis of Cauchy-Cauchy equation, L_{shish} can be obtained from the slope, and the misorientation width, B_{ϕ} can be obtained from the intercept of the plots.

The M4 and SIOC fiber structural parameters of shish, L_{shish} and B_{ϕ} , obtained by the Ruland's streak analysis using Figs. 9(a) and 9(b) are 663 nm, 0.013 and 1061 nm, 0.006. The increase in length of shish and decrease in shish misorientation can be explained that the coiled UHMWPE chains network is easily disentangled and developed to a uniform distribution shish structure in SIOC fiber with less branched short chains.

CONCLUSIONS

Microbeam X-ray diffraction measurements provided detail structural differences between two kinds of UHMWPE fibers. The two-dimensional WAXD patterns showed the monoclinic unit cell appearing in the whole M4 fiber. And there were just the different lamellar orientations existing in the bamboo joint of the SIOC fiber. The transformation of orthorhombic into monoclinic in M4 fiber was mainly influenced by the less branched and short chains on the main polyethylene chains. As a result, the orientation distributions of SIOC fiber were better and more regular than those of the M4 fiber. The DSC result of M4 fiber indicated the existence of branched chains significantly reduced or eliminated the orthorhombic-hexagonal phase transformation. The shish structures confirmed the uniform distribution shish structure with less branched and short chains. Finally the transformation of the structural knot in hot drawing processing and the elimination of the structural knot may be useful for enhancing the mechanical properties of UHMWPE fiber in the future.

ACKNOWLEDGEMENTS We thank Shanghai Synchrotron Radiation Facility (SSRF) for supporting the WAXD and SAXS tests.

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