Control of Microstructure in the Spinning of Mesophase Fibers

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Introduction

Micrographic studies of mesophase carbon fiber [1,2,3,4] have defined a number of characteristic features; the present work seeks to understand their origin by applying polarized-light micrography to selected sections through spinnerets that are quenched while the mesophase is flowing under spinning conditions. The spinnerets are not designed for fiber-spinning per se, but for ease of quenching, sectioning, and observation of the formation and relaxation of flow-induced microstructures.

Experimental

This abstract summarizes observations for a single flow condition applied to an alkylbenzene-based 100%-mesophase pitch (from Mitsubishi Oil) with a Mettler softening point of 285°C. The flow temperature was 300°C where the viscosity is about 96 Pa-s [5]. The spinneret configuration is shown in Fig. 1; the average velocity in the flow conditioning tube was 6.6 mm/min, giving a shear rate of 1482 s^{-1} in the 460-µm capillary. A single 325-mesh screen is placed above the spinneret in order to produce a well-defined microstructure in mesophase moving downstream to encounter the 80-mesh screen rotated 45° from the first screen. The mesophase above the spinneret is stirred to maintain homogeneity, but slowly to avoid bubble incorporation.

Micrographic Observations

The letters in Fig. 1 specify location for the crossed-polar micrographs of Fig 2. Transverse section A shows the circumferential microstructure (with concentric preferred orientation) induced by stirring the mesophase flowing to the first screen. Section B, a NE quadrant just below this screen, shows the grid cells imposed by flow around each wire of the screen. Traces of short-lived ripples within each cell and oriented cell walls are still present [5]. The mesophase within each cell is coarse, often dominated by a single $+2\pi$ disclination; the sensitive-tint response within cells is largely yellow, and the concentric orientation, maintained in large part after transit through the screen, is accommodated by the tendency of this disclination to locate in the SW corner of each cell. C shows the NE quadrant just prior to transit of the 80-mesh screen. The cell boundaries coarsen by disclination reactions but some concentric orientation is retained within the cells. D shows the grid texture after transit of the 80-mesh screen; rippled structures superimpose on the regular pattern of 325-mesh cells. The square arrays of $+2\pi$ disclinations lie at 45° to the coarse screen, and the fine cell walls, also at 45°, can be seen to define a "grid-within-a-grid". E shows the effect of convergent flow within the cone leading to the capillary; sensitive-tint response demonstrates that convergent flow causes a change from concentric to radial orientation. This preferred orientation strengthens at larger distance from the center. Shear at the wall introduces some zig-zag banding.

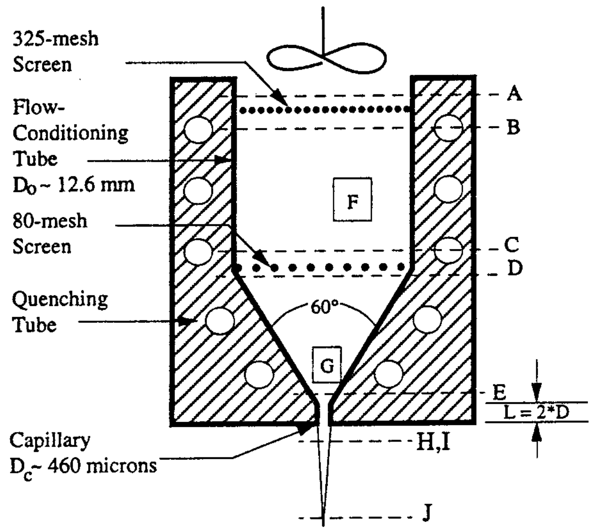

Fig. 1 Schematic diagram of a spinneret design for quenching during the spinning of mesophase pitch.

Longitudinal sections F, G show that convergent flow also imposes strong axial orientation on the misoriented layers flowing from the straight section of the spinneret. The bright streaks in G appear to be $+2\pi$ disclinations with relaxed cores; by sensitive-tint illumination, most of these are yellow on the left side and blue on the right, so the disclination structure indicates the direction of mesophase flow.

Sections H and I show the grid-within-a-grid texture in an extruded rod that also retains the radial preferred orientation. The interior grid, with the regular pattern of the $+2\pi$ disclinations, confirms the microstructure Taylor and Cross [4] observed in finished fibers; the distance between the extinction contours changes by square root of two upon rotation of the stage by 45° [4]. The grid-within-a-grid is still identifiable in as-spun filaments (section J); the preferred orientation is strong enough to induce the characteristic radial crack.

Conclusions

Mesophase pitch spun at viscosities near 100 Pa-s has a strong memory for disclination arrays whose geometry (but not disclination number) can be retained at least to 50-µm filaments.

Convergent flow is potent in orienting mesophase to the flow direction and in imposing radial orientation upon a filament.

If a specific microstructural feature is desired, e.g., strongly oriented cell walls, the spinneret must be designed to capture this structure before it decays during flow to the capillary; this point has been made by Hara [6].

Higher resolution micrography should be applied to evaluate how far the microstructural mechanisms observed here extend to the scale of 10-µm filaments.

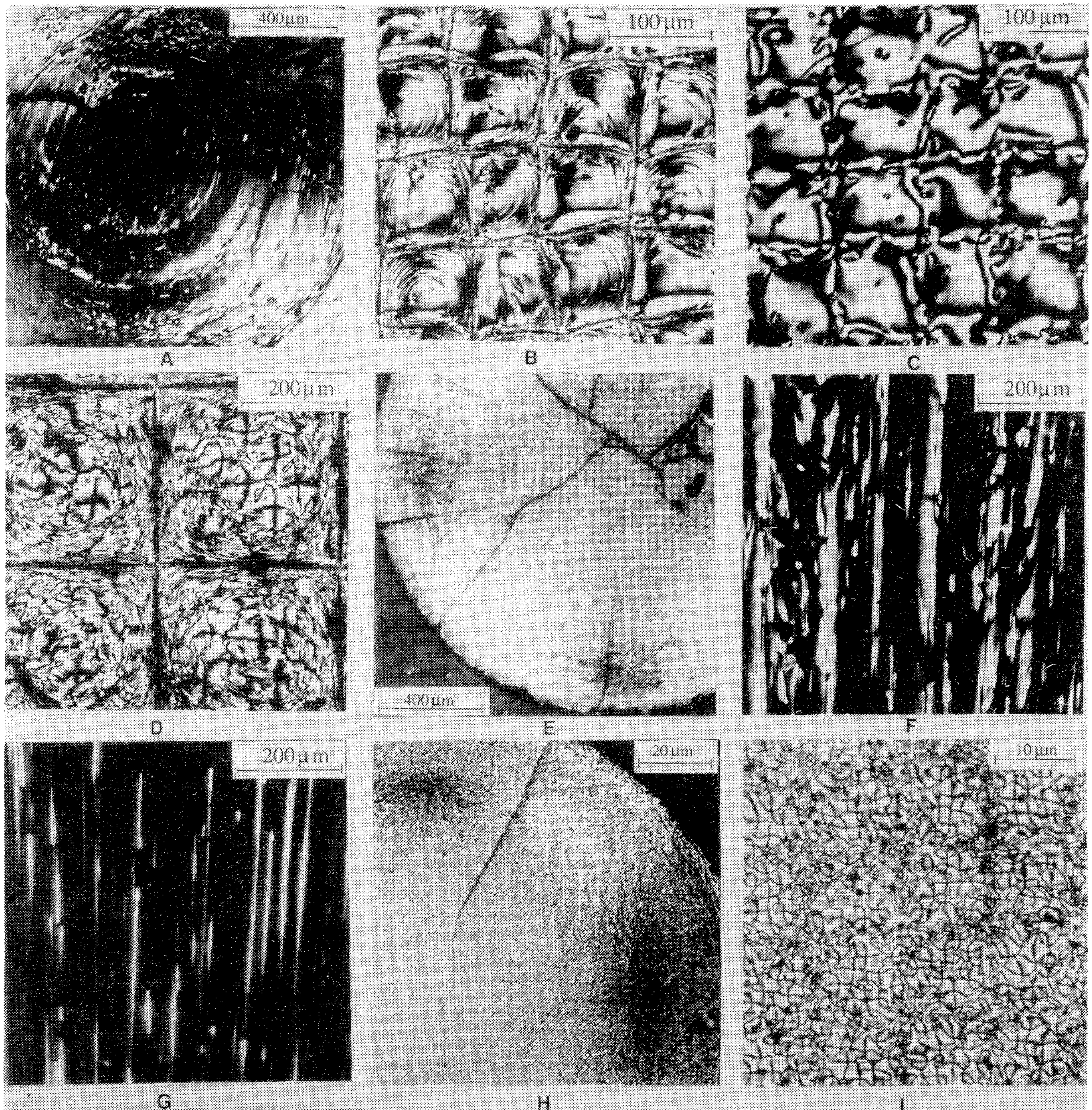

Fig. 2 Transverse and longitudinal views of the microstructure of a mesophase pitch flowing in the spinneret shown in Fig. 1.

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