MODELING OF STRUCTURE DEVELOPMENT DURING FLOW OF MESOPHASE PITCH THROUGH CAPILLARIES

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INTRODUCTION

Numerous studies have shown that the mechanical and thermal properties of melt spun mesophase pitch-based fibers are highly dependent on the microstructure generated during flow through the spinnerettes [1-2]. In a recent work Fathollahi [3] qualitatively analyzed the flow-induced microstructure of mesophase samples quenched during flow through capillaries. He reported that a fine microstructure readily developed during the flow and that increasing the throughput resulted in a finer microstructure. Also, for a given flow situation, he observed a finer structure near the wall of the capillary than in the core region. This is the direct result of the polydomain structure of mesophase pitch [4]. During flow the shear field applied causes the network of disclinations (the polydomain network) to deform and stretch resulting in a reduction in the size of the structure.

In the present study we used Computational Fluid Dynamics (a computer-based solution of the fundamental equations governing the flow of a particular fluid) to predict the microstructure that is generated during flow of mesophase pitch through round capillaries. The modeling work focused on the effect of capillary entry-angle on the development of microstructure. The flow-induced microstructure predicted by CFD matched that observed on mesophase samples extruded through round capillaries over a range of processing conditions.

MATERIALS AND METHODS

The mesophase pitch studied (provided by Mitsubishi Gas Chemical Company) was produced by catalytic polymerization of naphthalene (AR mesophase).

In order to experimentally analyze the flow-induced microstructure generated during flow of AR mesophase through round capillaries, a quenching apparatus (similar to that designed by McHugh [5] and illustrated in Figure 1) was used. This apparatus attached to the bottom of an Instron capillary rheometer model 2512-202. During a typical experiment, the molten pitch was forced through the capillary at a constant selected speed. The mesophase was allowed to flow until steady state was reached. Then, the flow of water was initiated in the quenching apparatus, instantaneously freezing the mesophase. After the entire system had cooled to room temperature, the capillary was removed from the quenching chamber. Finally, the capillary was cut transversely and/or longitudinally, and the flow-induced microstructure was analyzed under a polarized light microscope. In order to determine the effect of capillary entry-angle on the development of microstructure, four round capillaries with entry angles ranging from 30 ° to 90 ° were utilized in these experiments.

Polyflow, a Computational Fluid Dynamics package, was used to model the flow of AR mesophase through the capillaries. The viscoelastic nature of the mesophase pitch was taken into account by fitting its rheological behavior to a White-Metzner viscoelastic model. Also, following Marrucci's work on liquid crystalline polymers [6], the size of the flow-induced structure of the mesophase pitch, a, was assumed to depend on the rate of shear, $\dot{\gamma}$, as follows:

$$(1-a/a_0) \cdot ([a_0/a]^2-1) = a_0^2 . \mu_{II} . \dot{\gamma}/K , \qquad (1)$$

where a_0 is the size of structure at rest, μ_{II} is the viscosity of the mesophase in region II flow [6], and K is an average elastic constant of the mesophase. The numerical values of the parameters in equation (1) were determined during an earlier rheological study of AR mesophase [4].

RESULTS AND DISCUSSION

Optical analysis of the longitudinal section of the quenched capillaries showed that their entry-angle significantly affected the development of microstructure. For example, a vortex developed at the base of the entry counterbore for a flat-entry capillary, as illustrated in Figure 2(a). Note that during spinning a vortex is undesirable because it creates a region where pitch off-gassing and degradation are likely to occur. We observed that the vortex could be eliminated by machining an entry-angle at the base of the counterbore.

As Figure 2(b) illustrates, the model predicted the vortex for AR mesophase flow through the flat-entry capillary. The magnitude of this vortex agreed with optical analysis of quenched capillaries. The modeling of flow through capillaries with converging entrance regions also matched experimental observations (no vortex predicted).

flow through capillaries with converging entrance regions also matched experimental observations (no vortex predicted). The modeling also showed that under spinning conditions (fast flow) a die with an L/D of 3 was sufficient to allow the full development of structure, as observed experimentally.

Optical analysis of transverse sections of quenched capillaries showed that the texture observed in the upper channel matched our predictions, as illustrated in Figure 3. However, the small texture that developed during flow through the capillary die made only qualitative agreement possible in this region. In both cases, the size of the texture was smaller near the wall of the capillary than in the core region.

CONCLUSIONS

During this study, the flow of AR mesophase through round capillaries was successfully modeled. The results suggested that during spinning of AR mesophase into fiber form, a converging flow prior to extrusion through the capillary could eliminate vortex formation thereby reducing pitch degradation and/or off-gassing. Also, spinning dies with L/Ds about 3 (widely used in melt spinning processes) allow the full development of microstructure.

Finally, Marrucci's model for polydomain shrinkage during shear flow appears to predict the transverse texture that develops during flow of AR mesophase through capillaries.

ACKNOWLEDGMENTS

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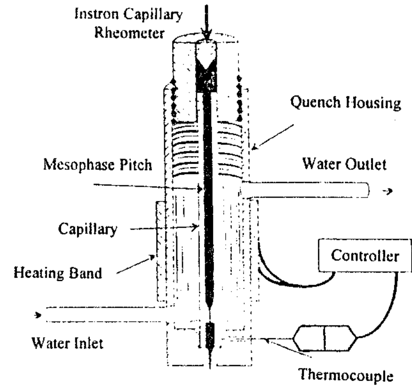
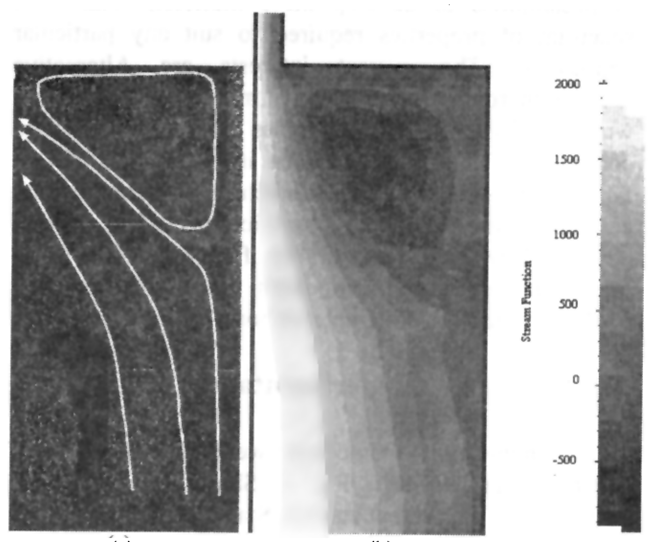


Figure 1. Design of mesophase pitch quenching apparatus.



(a) (b)

Figure 2. Comparison between an experimental picture of circular nine to one contraction flow of AR mesophase (a), and numerical calculations based on White-Metzner model (b). Note that color pictures clearly showing the flow pattern will be presented at the Conference.

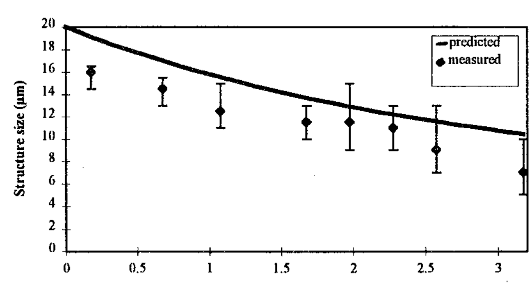


Figure 3. Comparison between predicted and measured size of the transverse texture in the upper channel.