Effects of Shear Rate and Capillary Entry Angle on Fiber Properties

W. D. Nothwang, T. R. McCormick, S. Sheikh, B. Starks, C. C. Fain and D. D. Edie,
Center for Advanced Engineering Fibers and Films
Clemson University, Clemson, SC 29634

Introduction

In many high-performance electronic applications, excessive heat generation limits circuit life to as little as 20 hours. Although pitch-based carbon fibers, with thermal conductivities from three to five times that of copper at room temperature, would appear to be ideal for these and other applications where heat transfer is critical, their high cost (approximately $1000/lb) makes them impractical for high-volume consumer applications. Much of the cost of producing these fibers is the direct result of the extremely high heat treatment temperatures (~3000 °C) needed to develop and perfect the crystalline structure needed for high fiber thermal conductivity (1,2).

Several researchers noted that changing the melt spinning conditions altered the fiber microstructure (2,3,4,5). If the fiber's lattice structure could be optimized during melt spinning, it might be possible to lower final heat treatment temperatures and, thus, production costs.

Experimental

A designed set of experiments was employed to investigate the effect of shear rate on fiber properties. Mitsubishi Gas Chemical Co. supplied a large sample of AR-mesophase (AR 24-15X1) for this study(6). Two sets of experiments were conducted, during which ribbon-shaped fibers (3) were spun on a bench-scale extrusion apparatus at a constant extension ratio of 140 and a temperature of 302 °C, and throughput was varied. The first set of experiments employed a spinneret containing profiled-entry capillaries(7), while in the second set of experiments a spinneret containing flat-entry capillaries was used. The utilization of a profiled-entry capillary and a flat-entry capillary allowed the effects of the capillary entry angle to be investigated.

A constant oxidation schedule was used for all fiber sets, and they were all treated to 2400 °C under similar conditions.

The electrical resistivities were measured using a monofilament four-point resistance test (1). Utilizing Wide Angle X-Ray Diffraction (WAXRD) the lattice parameters and crystal misorientation (Z value) of the fiber sets were measured and compared. A regression analysis was applied to quantify the influence of shear rate and capillary entry angle on these critical fiber properties.

Results and Discussion

The crystal misorientation angle (Z value) was used as a initial indicator of fiber properties. Figure 1 shows an increase in as-spun Z value as the shear rate increased for fibers spun through a flat-entry capillary.

The Z value was measured after heat treatment to quantify the improvement in lattice parameters with heat treatment for fibers spun using a flat-entry spinneret. Again, an increase in Z value is noted in Figure 2 as shear rate increased, but there is a noticeable drop in Z value above 4500 s^{-1}. The electrical resistivities closely follow the graphitized Z values. Tests are currently being conducted to discern if the transition at 4500 s^{-1} is an experimental artifact or a true transition.

The uniform strain (slope of the Nelson-Riley function) in the c-direction is shown to increase for fibers spun using a flat-entry spinneret with increasing shear in Figure 3. The negative sign indicates that the strain is compressive in nature. The increased compressive strains correlate closely with the as-spun misorientation angle.

The high compressive strain in the c-direction also tends to favor crystal growth in the a-direction in fibers extruded through a flat-entry capillary. Figure 4 shows a slight increase in $L_a(100)$ as shear rate increases. The L_c was approximately constant over the shear range.

Conclusions

Higher shear rates within flat-entry capillaries tend to increase the c-direction compressive strain, increase the crystal misorientation up to ~4500 s^{-1}, and favor larger L_a crystallite lengths within the fibers. It appears that, highly ordered fibers may be produced at low shear rates and at high shear rates. However, these experiments indicate that a transition region exists, and should be avoided during fiber production.

Tests show that profiling the capillary entrance affects the fiber properties. Results from these experiments will be presented at the conference.

Acknowledgments

The authors would like to thank the Office of Naval Research for funding this research, and Amoco Performance Products Inc. for their assistance, cooperation and advice.

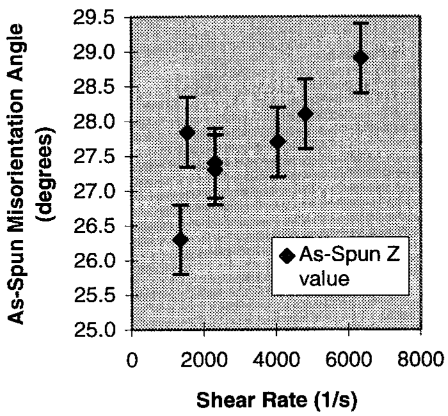

Figure 1. As-spun misorientation angle as a function of shear rate for fibers spun using flat-entry capillaries.

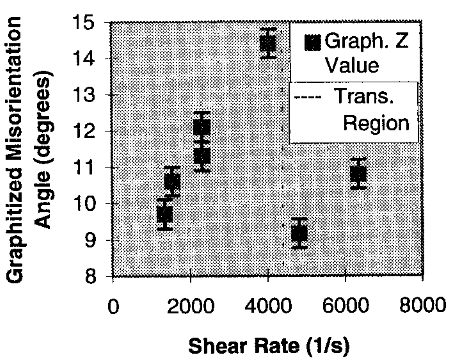

Figure 2. Graphitized misorientation angle vs. shear rate for ribbon-shaped AR-mesophase pitch-based carbon fibers spun using a flat-entry spinneret.

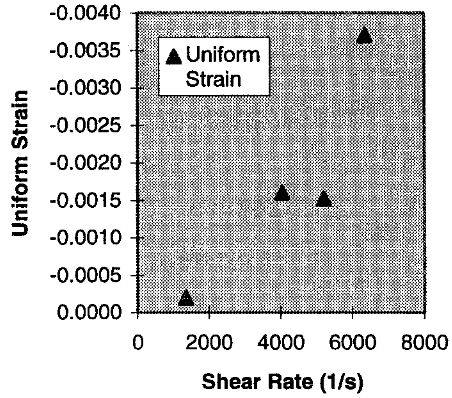

Figure 3. Uniform strain dependance on shear rate for fibers spun using a flat-entry spinneret.

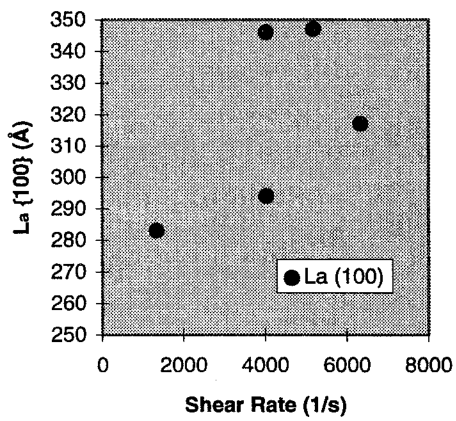

Figure 4. Crystal size, based on the 100 peak, as related to shear rate within a flat-entry capillary.

References

1. Bright, A. A. and Singer, L. S., *Carbon*, vol. 17, 1979, 59-69.
2. Robinson, K. E. and Edie, D. D., *Carbon*, 1996, 34(1), 13-36.
3. Edie, D. D., Robinson, K. E., Fleurot, O., Jones, S. P. and Fain, C. C., *Carbon*, 1996, 32(6), 1045-1054.
4. McHugh, J. J., Liu, G. Z. and Edie, D. D., *TANSO*, 1992, no. 155, 417-425.
5. Edie, D. D., Fain, C. C., Robinson, K. E., Harper, A. M. and Rogers D. K., *Carbon*, 1993, 31(6), 941-949.
6. Yoon, S. H., Korai, Y., Mochida, I. and Kato I., *Carbon*, 1994, 32(2), 273.280.
7. McHugh, J. J. and Edie, D. D., *Liquid Crystals*, 1995, 18(2), 327-335.