Use of the Coextrusion Technique for Producing Flame-Retardant and Antistatic Fibers

INTRODUCTION

In recent years, considerable interest has been aroused in the fiber industry by the use of additives to develop flame-retardant fibers and antistatic fibers. When additives are used, fundamental questions arise both as to the role they play in the process of fiber formation and the mode of distribution, location, and shape of the additive particles that will maximize their effectiveness. Understandably, problems become quite complicated when the base fiber-forming polymer and the additives form two phases in the molten state.

The addition of flame-retardant additives to melt spinnable polymers prior to extrusion into fibers is an attractive approach for achieving improved flame retardancy. This is especially so in view of the technical and economic difficulties associated with the development of the inherently flame-retardant polymers. A very substantial amount of research and development has been carried out on the study of candidate flame-retardant additives for melt spinnable polymers and is reported in a number of patents.¹ Nevertheless, very little, if any, attention has been given to the distribution, location, and shape of such additives within the fiber structure in such a way as to maximize flame retardancy at *minimum* additive contents. Clearly, the preferred location of suitably distributed and shaped additives is the one that optimizes their flame-retardant effectiveness. The same is true in the situation where antistatic fibers are to be produced by using an antistatic agent as additive. Again, much effort has been spent on finding chemicals possessing antistatic properties for fiber spinning, and the results are reported in a number of patents.²

In order to control the preferential location of additives in the melt-spun fiber, the authors have very recently employed the coextrusion technique for producing flame-retardant fibers. As a matter of fact, the coextrusion technique has long been used in the fiber industry, dating back to the pioneering research of Sisson and Morehead³ and later researchers.^{4,5} The primary motivation of these studies was to produce crimped fibers, very similar to natural wool fibers. For this, Sisson and Morehead³ spun two viscose rayon solutions side by side through circular spinnerette holes and successfully produced crimped fibers which possessed a certain degree of bilateral structure. Similarly, Hicks et al.⁴ developed the bicomponent acrylic fiber which also produced crimps owing to bilateral structure.

More recently, some fundamental studies have been reported on the coextrusion of polymer melts in circular dies.⁶⁻¹⁰ These studies report how the interface shape in the extrudate might be affected either by the difference in the rheological properties of the two polymers coextruded and/or by the



Fig. 1. Schematic of the cross section of the fiber with an additive in the core, in which A denotes the fiber-forming polymer and B denotes polymer A with additives.

2913

© 1976 by John Wiley & Sons, Inc.



Fig. 2. Schematic of the experimental apparatus.



Fig. 3. Schematic of the feedblock design.

manner in which the two polymers meet each other at the die inlet (i.e., side by side, concentrically, or eccentrically).

In the present study, an additive was first dispersed in a fiber-forming polymer, and then two polymer streams, one with an additive and the other without an additive, were coextruded through spinnerette holes, the former forming the core and the latter the sheath, as shown schematically in



Fig. 4. Photographs of the cross section of the LDPE fiber at different stretch ratios: (a) V_L/V_0 = 0; (b) V_L/V_0 = 3.2; (c) V_L/V_0 = 8.6. Melt temperature was 200°C.

Figure 1. In this way, it was possible to stop the migration of the additives to the surface of the spun fiber, which is essential for flame-retardant fibers. Because of the toxicity of almost all flame-retardant components, their presence on the fiber's surface can give rise to skin irritation.

EXPERIMENTAL

Melt spinning experiments were carried out in the apparatus recently constructed in connection with an earlier research program of Han,^{8,10} with a slight modification. It consisted of two melt supply units (two extruders), a static mixer, a spinnerette, and temperature control devices. The heart of the experimental apparatus is the proper design of the system of feeding two polymers into the spinnerette, and the design of the spinnerette hole itself. As shown schematically in Figure 2, stream B of the fiber-forming polymer is melt blended with a flame-retardant component and fed to the inner side of the spinnerette hole; and stream A, the same fiber-forming polymer *without* a flame-retardant component, is fed to the spinnerette hole outside of stream B. Figure 3 shows a schematic of the die design.

The polymers melt spun were polypropylene (Exxon Chemical, Resin E115), high-density polyethylene (Union Carbide, DMDJ 4306), and low-density polyethylene (Union Carbide, DYNF 1). As flame-retardant component, we used the commercially available antimony silico oxide (NL Industries, ONCOR 23A). The purpose of our study was to determine whether this particular coextrusion technique has a potential usefulness for producing fibers that retain a flame-retardant component within the fiber core.

It should be emphasized that an additive must be very well dispersed in a molten polymer if its effectiveness is to be maximized. When additives are liquid (and, as a matter of fact, most antistatic



Fig. 5. Photographs of the cross section of the LDPE fiber with a very thin skin shielding the core: (a) $V_L/V_0 = 0$; (b) $V_L/V_0 = 3.8$. Melt temperature 200°C.

components and organic flame-retardant components are available in the liquid state), they are expected to form small droplets in the fiber-forming polymer while being spun. Therefore, one may surmise that the method of mixing strongly influences the state of dispersion, location, and even shape, of the droplets in the molten polymer. In our experiment, a flame-retardant component was first dry-blended with the polymer and then fed to the extruder wherein mixing occurred. In order to improve the state of dispersion of the flame-retardant component in the molten polymer, an additional mixing device, called "Static Mixer" (Kenics Corp.), was installed between the extruder and the die (see Fig. 2).

At each spinning condition (flow rate and temperature), extrudate samples were collected at different stretch rates. These samples were later cross-sectioned and examined under an optical microscope to determine the shape of the interface and the state of dispersion of the flame-retardant component.

RESULTS AND DISCUSSION

Figure 4 gives representative photographs of the low-density polyethylene (LDPE) fiber cross section at different stretch ratios. Note that the core (dark area) contains the flame-retardant component, shielded completely by the LDPE (white area) that contains no flame-retardant agent. Figure 5 is given to demonstrate that one can indeed obtain a very thin layer shielding the core completely by controlling the volumetric flow rates of two streams, one stream being fed to the inside of the spinnerette hole and another stream fed to the outside of the core stream.

Figure 6 gives photomicrographs of the fiber cross section. These photographs were taken with the aid of an optical microscope. It is seen that the flame-retardant component is distributed uniformly and that the additive particles stay within the core.

The coextrusion technique as demonstrated above can be used to distribute the additives at any preferred location in fibers of any cross section, e.g., trilobal, star-shape, rectangular, etc. This would not be readily realized by other extrusion techniques.

It should be pointed out that the coextrusion technique presented above can also be used to produce antistatic fibers. Since an additive such as an antistatic component can give rise to adverse effects on the physical/mechanical properties of the fibers melt spun, control of the distribution, location, and shape of additive particles is very important for meeting the fiber properties desired. The



Fig. 6. Microphotographs of the cross section of the HDPE fiber: (a) edge portion; (b) center portion.

coextrusion technique as demonstrated above can produce fibers with an additive (or additives) shielded completely by a *thin* skin of the fiber-forming polymer. Hence, one can eliminate the possibility of a health hazard in the case where such an additive (or additives) is toxic and can cause skin irritation when the fibers are used in garment fabrics.

This work was supported in part by American Enka Company, to which the authors are very grateful.

References

1. U.S. Pat. 3,629,365 (1971); Brit. Pat. 1,279,357 (1970); Jap. Pats. 7,360,142 (1973); 7,360,141 (1973); 7,244,981 (1972); 7,310,443 (1973); 7,395,445 (1973); 7,355,950 (1973); 7,227,137 (1972); 7,223,973 (1972); 7,227,138 (1972); 7,482,744 (1974); 7,427,617 (1974).

2. U.S. Pats. 3,560,419 (1971); 3,794,631 (1974); Fr. Pat. 2,012,439 (1970); Jap. Pats. 7,008,172 (1970); 6,932,309 (1969); 7,026,970 (1970); 7,026,974 (1970); 7,105,223 (1971); 7,107,213 (1971); 7,105,211 (1971); 7,110,495 (1971); 7,107,174 (1971); 7,116,319 (1971); 7,118,614 (1971); 7,126,894 (1971); 7,133,130 (1971); 7,133,131 (1971); 7,140,490 (1971); 7,142,333 (1971); 7,227,805 (1972); 7,236,459 (1972).

3. W. E. Sisson and F. F. Morehead, Text. Res. J., 23, 152 (1953); ibid., 30, 153 (1960).

4. E. M. Hicks, J. F. Ryan, R. B. Taylor, and R. L. Tichenor, Text. Res. J., 30, 675 (1960).

2918 JOURNAL OF APPLIED POLYMER SCIENCE VOL. 20 (1976)

5. E. M. Hicks, E. A. Tippets, J. V. Hewett, and R. H. Brand, in *Man-Made Fibers*, Vol. 1, H. Mark, S. A. Atlas, and E. Cernia, Eds., Interscience, New York, 1967.

6. J. H. Southern and R. L. Ballman, Appl. Polym. Symp., No. 20, 1234 (1973).

7. J. H. Southern and R. L. Ballman, J. Polym. Sci. (Polym. Phys. Ed.), 13, 863 (1975).

8. C. D. Han, J. Appl. Polym. Sci., 17, 1289 (1973).

9. B. L. Lee and J. L. White, Trans, Soc. Rheol., 18, 467 (1974).

10. C. D. Han, J. Appl. Polym. Sci., 19, 1875 (1975).

Chang Dae Han Young Woo Kim

Department of Chemical Engineering Polytechnic Institute of New York Brooklyn, New York 11201

Received January 20, 1976