Inorganic-Organic Hybrid Monofilaments: A Preliminary Study

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Introduction: Traditional melt-extruded, small diameter filaments having a cross-sectional area of less than 2 mm^2, such as monofilaments and multifilament yarns used for manufacturing different knitted and woven textile constructs, monofilament sutures, and multifilament braided sutures, are known to be based on thermoplastic crystalline polymers comprising linear chains. An exception to the traditional practice was disclosed earlier by one of the present investigators, wherein polyaxial polymers (with a monocentric branching point) were prepared and converted to strong monofilaments useful for the production of surgical sutures and allied medical products[1,2]. It is also traditional to incorporate less than 2 weight percent of solid inorganic additives in textile fibers as delustering agents (e.g., TiO_2) and to a lesser extent, colorants and heat stabilizers. And frequently, these additives tend to aggregate in the polymer melt and interfere with extrusion of fibers having small cross-sectional areas during melt-spinning. In spite of the availability of a great number of inorganic additives that can conceptually impart unique and useful properties to extruded filaments if used in quantities exceeding 2 weight percent, earlier investigators have not explored this option to avoid known or perceived complications in the melt-spinning of such inorganic-organic hybrid systems. These facts and contemporary needs for unique hybrid microcomposites in filament form provided a strong incentive to pursue the present study, which is directed toward a new family of inorganic-organic hybrid filaments. These are intended to contain at least 30 weight percent of an inorganic component uniformly dispersed as microparticles in an organic polymeric matrix to impart one or more useful properties to medical devices. More specifically, this communication deals with hybrid monofilaments of microparticulate barium sulfate in absorbable segmented copolyesters. The hybrid monofilaments are expected to be used as radiopaque components of endourological devices with tailored absorption/disintegration and strength retention profiles[4].

Materials and Methods: Composite Polymer Preparation—Inorganic-organic composites of polyaxial segmented copolyesters, HC-1, were prepared by end-grafting a polyaxial poly(trimethylene carbonate) in glycolide and ε-caprolactone in presence of microparticulate anhydrous barium sulfate. Similarly, inorganic-organic composites of linear copolyesters, HC-2, based on *l*-lactide and ε-caprolactone were prepared in presence of barium sulfate. The composites were ground and traces of monomer were removed by heating under reduced pressure. The polymers were then evaluated for their inherent viscosity (I.V.) and thermal properties using a differential scanning calorimeter (DSC). **Hybrid Monofilament Preparation—**Copolyester composites HC-1 and HC-2 were melt extruded into hybrid monofilaments, HF-1 and HF-2, respectively, as described in an earlier disclosure[3] using a single screw ¾" extruder equipped with a melt pump and a 60-mil die. For monofilament orientation, a series of godets and hot shoes were used to achieve a desired diameter. The hybrid monofilaments were evaluated for key mechanical properties using an MTS Synergie 200 Universal Tester.

Results and Discussion: The properties of typical polyaxial and linear composite copolyesters with greater than 30 weight percent microparticulate barium sulfate are summarized in Table I and demonstrate the ability to produce composites with over 30 weight percent inorganic dispersed phase in an absorbable matrix. More specifically, the data demonstrate that composites of high molecular weight (measured in terms of I.V.) polyaxial and linear crystalline copolyesters can be prepared. The properties of typical hybrid monofilaments are given in Table II. The data demonstrate the ability to prepare such hybrid monofilaments with moderate strength and low modulus in spite of their high inorganic content. The data in Table II are also indicative of the high uniformity of the barium sulfate microparticulate and absence of aggregate that could have led to brittle fracture and unacceptable monofilament properties.

Table I. Properties of Typical Polyaxial and Linear Copolyester Composites

Polymer	DSC Data		I.V. (dl/g)
	Δ H (J/g)	Melt Temp (ºC)	
HC-1	86.9	217.2	0.88
HC-2	25.9	142.1	1.17

Table II. Mechanical Properties of Typical Hybrid Monofilaments

Material	Diameter (mm)	Max Load (N)	Strength (ksi)	Modulus (ksi)	Elongation (%)
HF-1	0.32±0.01	12.1±0.28	21.9±0.9	147±30	34.7±2.8
HF-1	0.35±0.01	13.1±0.46	20.4±1.0	119±14	36.7±2.3
HF-2	0.34±0.01	22.8±0.69	37.3±1.3	265±37	42.1±1.1
HF-2	0.43±0.01	29.2±0.46	29.2±1.9	215±12	59.5±1.1

Conclusions: Available results demonstrate the feasibility of preparing inorganic-organic composites of high molecular weight absorbable copolyesters and barium sulfate as uniformly dispersed particles. Results also show that those composites can be converted to moderately strong hybrid monofilaments with unexpectedly low modulus. Thus, further exploration of variable compositions with different loadings of inorganic microparticulates as components of unique absorbable radiopaque endourological devices is warranted.

References:

1. Shalaby S.W., U.S. Patent 6,462,169 (2002)
2. Shalaby S.W., U.S. Patent 6,794,485 (2004)
3. Shalaby S.W., U.S. Patent Appl. 60/737,021 (2005)
4. Shalaby S.W. & Clinkscales K. W., U.S. Patent Appl. 11/346,117 (2006)