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FROM A MICRO-POLYMERIC PIPE TO A MINI-POLYMERIC PULSATING HEAT PIPE

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ABSTRACT

In this work, a preliminary study related to the design and manufacturing of a micro polymeric pipe for micro heat exchangers was performed. Among possible polymeric materials, a thermoplastic copolyester elastomer composed of polybuthylene-terephthalate, as crystalline phase, and long glycol chains, as the amorphous one (Hytrel® 6356, Dupont) was selected, both unloaded and loaded with 7 % w/w carbon powder. A microextrusion process was set up to obtain microtubes and the thermo-mechanical characteristics of the produced pipes were studied. Thermal properties of extruded Hytrel® remained almost the same, in terms of melting temperature ($T_m \cong 208$ °C) and enthalpy change of fusion ($\Delta H \cong 45$ mJ/mg), although the material was C-loaded. The thermo-mechanical tests performed on unloaded and C-loaded Hytrel® microtubes at 25 and 70 °C (possible working temperature) detected a considerable increase in the mechanical properties of C-loaded Hytrel®, compared to the unloaded one. In particular, a relevant improvement of the elastic modulus at 70 °C for the C-loaded microtubes was observed, demonstrating a better thermal stability at high temperature. Moreover, the fabrication of a micro heat exchanger prototype and prteliminary tests with different cooling fluids confirmed the possibility of using Hytrel® for electronic applications, as a good thermal exchange was evidenced.

INTRODUCTION

Heat management is an important problem in almost every industrial field, particularly for electronic devices, where the small dimensions and the lightness of thermal equipments must show stricter and stricter qualifications. Due to the presence of high temperature gradients, which straightforwardly involves heating/cooling powers, these characteristics are sometimes difficult to obtain.

Metals, that are good thermal conductors, easy to process and sometimes relatively light, are traditionally used to produce heat pipes [1, 2, 3]. Alternatively, heat pipes are built with ceramic materials [4]. Recently, the possibility of using polymeric materials instead of traditional conducting materials is under investigation [5].

The main advantage of the use of polymers is represented by their flexibility, so that they can be applied in miniaturized electronic devices, adapting them to the device geometry. On the other hand, the main problem in the use of polymers for this application is the fact that usually these materials have a very low thermal conductivity (lower than 1 W/mK). This requisite is in total disagreement with one of the most important characteristic of thermal equipments (thermal conductivity of about 20 W/mK). To increase thermal conductivity of polymers, different approaches have been proposed in [6] and [7]. Another possible solution is the loading with fillers, such as carbon nanoparticles or nanotubes.

Nowadays, two of the most interesting systems of heat transfer under investigation are represented by micro heat exchangers and mini pulsating heat pipes. In micro heat exchangers, flow channels are represented by capillaries, whereas in mini polymeric pulsating heat pipes the heat excess is transferred from the condenser to the evaporator, and viceversa, by mini pipes. For these types of heat pipes, the low wettability of polymeric surfaces, together with the material low density, flexibility and mechanical strength, suggests the development of a new technology for heat exchangers and mini heat pipes, based on extruded polymeric micro and mini tubes.

In this work, a polymeric material has been newly proposed for a possible use in the fabrication of a micro-heat pipe. The extrusion process was set up and the physico-chemical, morphological and thermo-mechanical properties of the microtubes, unloaded and loaded with carbon nanoparticles were investigated.

MATERIALS AND METHODS

Among different polymeric materials with adequate properties for fabricating microtubes for heat pipes [8-14], Hytrel® 6356 (DuPont, [13]), a thermoplastic co-polyester elastomer composed by polybutylene-terephthalate, as crystalline phase, and long glycol chains, as the amorphous

one, was selected.

To increase the thermal conductivity of the polymer (approximately ≈ 0.2 W/mK), Hytrel® 6356 was loaded with carbon nanoparticles (Figure 1). Different quantities of carbon nanoparticles kindly provided by Gimac srl (Castronno, VA, Italy) were used to set up the extrusion process (25% w/w down to 7% w/w). Best extrusion results were obtained with the lower quantity of C-nanoparticles (i.e., 7% w/w).

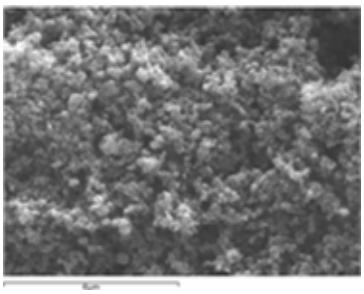

Figure 1 - SEM image of C-nanoparticle used as filler for Hytrel® 6356 microtubes.

Extrusion

In the extrusion process of polymers, raw thermoplastic materials in the form of small beads is gravity fed from the hopper into the barrel of the extruder (Figure 2). The material enters through the feed throat and comes in contact with the screw. The rotating screw forces the beads forward into the barrel which is heated to the desired melt temperature by independently controlled heaters that gradually increase the temperature of the barrel from the rear to the front. At the front of the barrel, the molten polymer enters into the die that gives to the final product the designed profile.

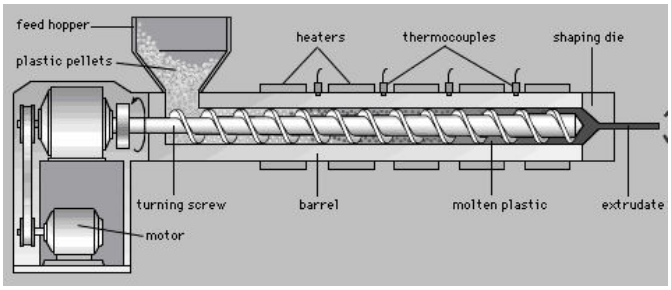

Figure 2 - Scheme of a polymer extruder.

Hytrel® 6356 microtubes were extruded by using a four-heating-zones horizontal Ø12/20D micro-extruder (Gimac srl, Castronno, VA, Italy) with a coaxial screw. The main advantages of micro-extrusion *vs* traditional extrusion systems [12] are the following ones:

- facility in controlling all the process system and industrial scaling up;
- smaller amount of material to be used;
- greater dimensional precision for the extruded products.

Different extrusion parameters have to be set up to obtain the optimal working conditions: temperatures related to the six thermoregulation zones (T_1-T_6), located among the hopper and the die, screw speed (S_s), and towing speed (S_t). After that, Hytrel® 6356 microtubes with different properties (Table 1) were extruded using the optimized parameters (Table 2). Hytrel® 6356 microtubes were cooled down by air exposure or in a water bath.

Table 1 - Extruded Hytrel® 6356 microtubes and related acronyms.

Hytrel® microtubes characteristics	Acronym
high wall thickness, cooled in air, unloaded	**HW-a**
high wall thickness, cooled in water, unloaded	**HW-w**
low wall thickness, cooled in water, unloaded	**LW-w**
low wall thickness, cooled in water, C-loaded	**LW-w-C**

Before the extrusion process, Hytrel® 6356 was dehydrated for at least 3 hours at 100°C, as recommended in the data sheet of the material, to avoid polymer hydrolysis and consequently extrusion problems. Moreover, once extruded, microtubes were heat-treated to remove possible residual stresses due to transformation process.

Table 2 –Extrusion parameters for the different Hytrel® 6356 microtubes.

Type of microtube	T_1 [°C]	T_2 [°C]	T_3 [°C]	T_4 [°C]	T_5 [°C]	T_6 [°C]	S_s [rpm]	S_t [m/min]
HW-a	205	215	220	225	230	220	7	27
HW-w	205	215	220	225	230	220	7	27
LW-w	210	215	220	225	225	220	1	9.9
LW-w-C	210	215	220	225	225	220	4	9.9

Morphological analysis

Samples of each type of microtubes were cut by using a microtome (Leica CM 1850), and observed with a stereo optical microscope (Nikon SMZ 800). Internal and external diameters were calculated by an image software.

Atomic Force Microscope (AFM, Autoprobe CP, Park Scientific Instruments) was used to investigate the microtube morphology and roughness. Three AFM analysis were performed to analyze in contact mode the internal surface of unloaded and loaded microtubes (50x50 μm^2 area).

Thermo-Gravimetric Analysis (TGA)

Thermo-gravimetric analysis (TGA, Mettler TA2000 system) was carried out to evaluate the amount of carbon nanoparticles in the microtube after the extrusion process. TGA was performed on 2 mg of LW-w-C in the temperature range 25 - 1000 °C in air flow.

Differential Scanning Calorimetry (DSC)

Thermal properties of pellets and extruded Hytrel® 6356 microtubes were investigated by differential scanning calorimeter (DSC, Seiko 6200) as reported in Table 3, with a heating rate of 20°C/min under nitrogen atmosphere. Melting temperature (T_m) and enthalpy heat (ΔH) were drawn from the DSC thermograms.

Table 3 – Parameters for DSC analysis.

		from [°C]	to [°C]
1st analysis		-150	350
2nd analysis	1st run	25	130
	cooling*		
	2nd run	-150	350

* cooling was performed at 10°C/min

Tensile Mechanical Tests

To evaluate the stress-strain/temperature behavior of the extruded microtubes, tensile mechanical tests were performed at a crosshead speed of 200 mm/min with a MTS 1/MH electromechanical instrument equipped with pneumatic grips and a thermostatic cell (CTD-200, MTS). Tests were carried out at r.t. and 70°C chosen as possible working temperature of electronic devices with tubular samples (n = 5) and a gage length of 50 mm. Grips pressure was fixed at 1 bar to avoid slippage of the sample. Tests were performed following the ASTM D 638-02 standard practice [18]. Tensile parameters drawn from the stress-strain curves were: tangent modulus (E), stress (σ_b) and strain (ε_b) at break.

Dynamic Mechanical Analysis (DMA)

Dynamic Mechanical Analysis (DMA) were performed with a DMA 2980 analyzer (TA Instrument) in tensile mode. Storage modulus (E'), loss modulus (E'') and tanδ (tanδ=E''/E') were recorded in the temperature range of 5 - 195°C (T ramp = 1°C/min), at 1 Hz and 15 μm strain application. Tests were performed in duplicate, according to the ASTM 1640-99 standard practice [19,20].

RESULTS AND DISCUSSION

Morphological characterization

By stereo optical microscopy (Figure 3) the values for the external ($Ø_{ext}$) and internal ($Ø_{int}$) diameters and, as a consequence, the value of wall thickness (W_t), were detected (Table 4).

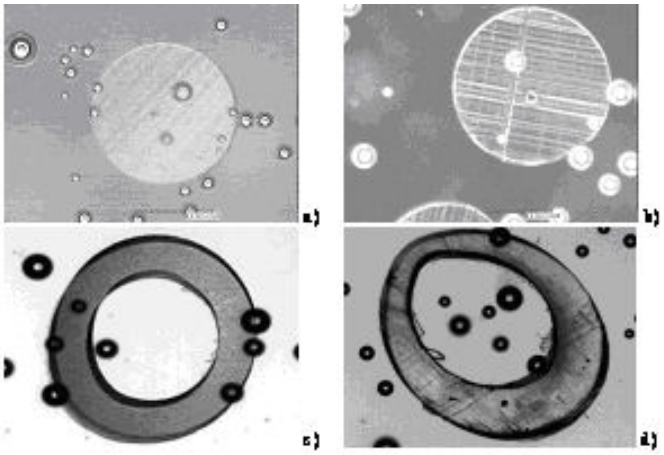

Figure 3 – Cross-section of Hytrel® 6356 microtubes: a) HW-a; b) HW-w; c) LW-w; d) LW-w-C.

Table 4 – Hytrel® 6356 microtubes dimensions.

Type of microtube	$Ø_{ext}$ [μm]	$Ø_{int}$ [μm]	W_t [μm]
HW-a	391	23.5	183.75
HW-w	400	30.0	185.00
LW-w	391	248	71.50
LW-w-C	392	250	71.00

AFM analyses showed a different material organization onto the surface. In particular, unloaded Hytrel® 6356 showed a globular morphology (Figure 4a), probably due to the phase separation between hard and soft domains created by the presence of differently organized segments (PBT and glycol chains). On C-loaded microtubes, the extrusion direction was evidenced (Figure 4b). Despite the different surface morphology (Figure 4), the values of average roughness (R_a) were approximately the same for LW-w and LW-w-C (respectively, 715 ± 54 Å and 727 ± 25 Å). The extrusion process allowed to obtain microtubes with good dimensional uniformity and adequate internal roughness values as demonstrated by the optical microscope observations and AFM analysis.

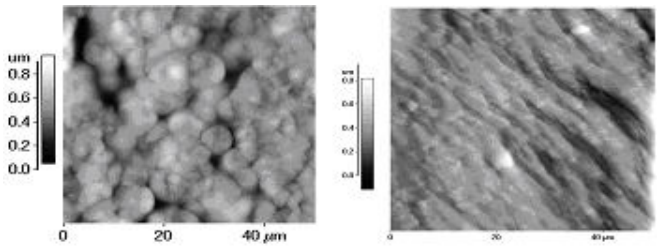

Figure 4 – Topography of unloaded (a) and C-loaded (b) Hytrel® 6356 microtubes by AFM.

TGA analysis demonstrated that at 369 °C almost all the polymer was burned off (≈ 92 %). The quantity of carbon powder found in the crucible was about 8% w/w (Figure 5), confirming that all the amount of C-nanoparticles loaded in the hopper was incorporated into the Hytrel® 6356 microtubes.

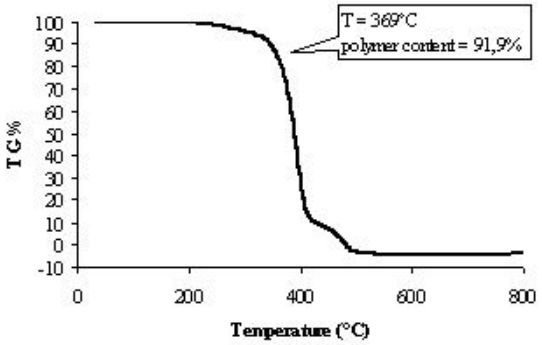

Figure 5 – TGA thermogram of LW-w-C

Differential Scanning Calorimetry (DSC) evidenced significant differences among pellets, and unloaded and C-loaded Hytrel® 6356 microtubes (Figure 6 and Table 5). The higher wall thickness produced a better crystals reorganization in unloaded Hytrel® 6356 microtubes. ΔH value for unloaded microtubes was lower than that detected for pellets and C-

loaded microtubes. This result can be explained by a different organization due to the extrusion process of LW-w microtubes and by an important role played by C-nanoparticles in the LW-w-C microtubes. These data confirmed the different surface organization detected by AFM analysis.

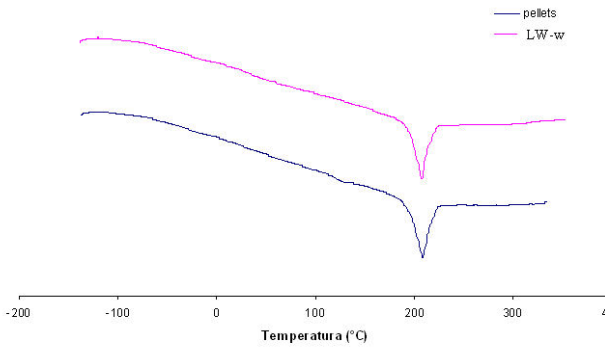

Figure 6 – Thermograms of Hytrel® 6356 pellets and LW-w microtube.

Table 5 – Thermal properties of Hytrel® 6356

	T_m [°C]	ΔH_m [mJ/mg]
pellets	208.8	48.70
HW-a	207.6	46.79
HW-w	207.9	50.61
LW-w	207.9	42.21
LW-w-C	207.0	47.32

The considered mechanical parameters of the extruded microtubes are reported in Table 6.

Table 6 – Mechanical properties of Hytrel® 6356 microtubes.

Type of microtube	T [°C]	E [MPa]	σ_b [MPa]	ε_b [%]
HW-a	25	146.6 ± 24.8	/*	/*
	70	56.3 ± 23.5	/*	/*
HW-w	25	101.8 ± 31.4	30.5 ± 2.8	505.4 ± 35.3
	70	47.3 ± 17.8	24.2 ± 4.3	530.2 ± 68.3
LW-w	25	56.7 ± 17.1	58.2 ± 10.3	487.8 ± 49.2
	70	6.6 ± 2.1	34.7 ± 3.2	463.2 ± 29.5
LW-w-C	25	93.7 ± 15.2	48.6 ± 3.9	397.0 ± 42.3
	70	48.0 ± 13.8	29.7 ± 11.9	373.9 ± 55.7

* the values were out of the crosshead run limit.

Tensile mechanical tests showed an elastomeric behavior for Hytrel® 6356 microtubes. It is possible to observe that decreasing the wall thickness, the stiffness of the microtubes changed. In particular, LW-w microtubes, compared with HW-w microtubes (same cooling condition, i.e. water), exhibited low E values and higher stress at break either at r.t. and 70°C. Loaded Hytrel showed a higher stiffness in comparison with unloaded microtubes, while maintaining a good tenacity (Figure 7).

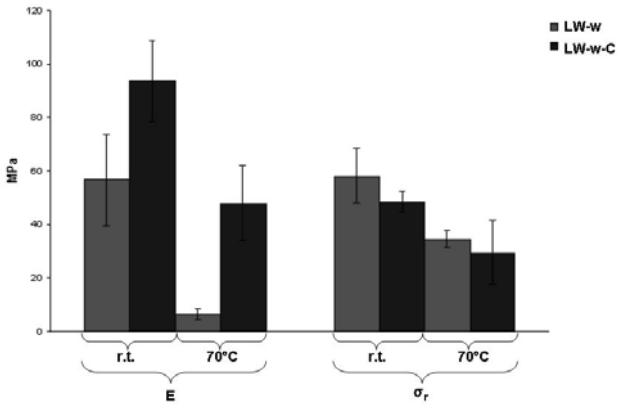

Figure 7 – Comparison between E and σ_b values at r.t. and 70°C for unloaded and C-loaded Hytrel 6356 microtubes.

From DMA analysis, it was possible to observe that the different cooling of the extruded microtubes did not influence the thermo-mechanical properties. DMA demonstrated that between 25 and 70 °C Hytrel® 6356 microtubes did not exhibit any transition, with a Tan δ vs T curve almost linear for all the temperature range of the analysis (Figure 8). Moreover, for unloaded and C-loaded Hytrel® 6356 microtubes, Loss Modulus, Storage Modulus and Tan δ did not change considerably, as reported in Figure 8. The E' curve showed a different trend in the 60 - 120°C temperature range, exhibiting a higher and constant value. These results could be significant for a change in thermo-mechanical behavior due to the presence of the inorganic filler.

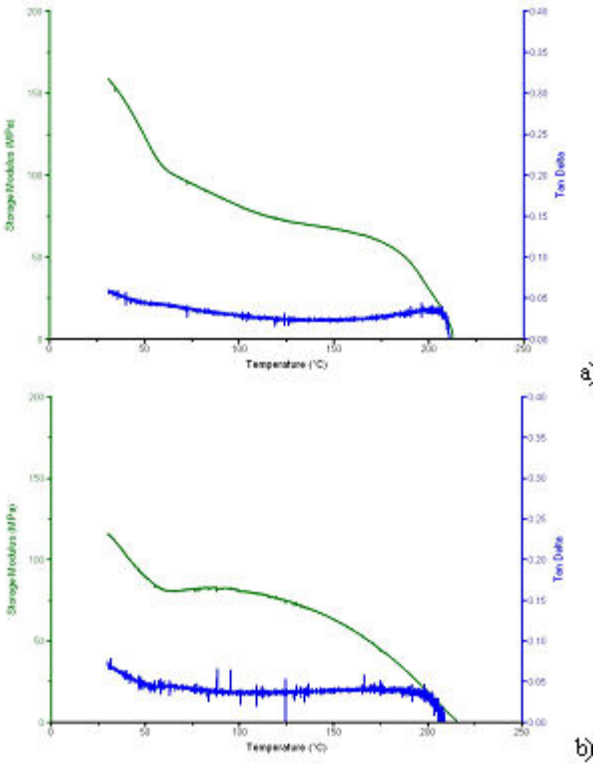

Figure 8 –Storage Modulus (green curve) and Tan δ (blue curve) for (a) LW-w and (b) LW-w-C Hytrel® 6356 microtubes.

Heat flow dissipation tests

A micro-heat-exchanger prototype (Figure 8) was designed and fabricated by M. Marengo et al. (Università di Bergamo, Italy), to test the dissipated heat flow related to the use of unloaded and C-loaded microtubes instead of more traditional materials. The exchanger was composed by LW-w or LW-w-C Hytrel® 6356 microtubes, nitrogen as working fluid was used to cool a central process unit (CPU). From this preliminary tests, it was established that Hytrel® 6356 microtubes caused a good maximum dissipated heat flow value (about 6.72 kW/m^2).

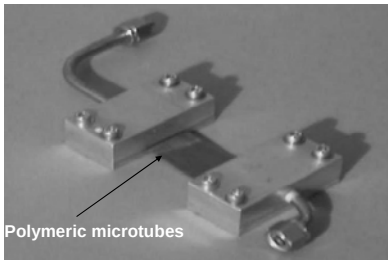

Figure 9 - Micro-heat exchanger prototype

CONCLUSIONS

From the obtained results it is possible to state that carbon powder addition stabilized the Hytrel® properties as well as increased its thermal conductivity. These findings will now help the design and fabrication of polymeric pipes of increased size, i.e. mini-polymeric pulsating heat pipes.

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