Development and Characterization of Poly(trimethylene terephthalate)-Based Bicomponent Meltblown Nonwovens

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Received 27 February 2001; accepted 28 March 2001

ABSTRACT: Poly(trimethylene terephthalate) (PTT)-based mono and bico meltblown webs have been produced by using a Reicofil® Bi-Component Meltblown Line at TANDEC, located at the University of Tennessee, Knoxville, TN. Thermal and flow properties of PTT were first examined by DSC (differential scanning calorimetry) and with a Melt Indexer for an effective experimental design through the Surface Response Methodology (SRM). The processability of meltblowing in a wide range of operating windows was extensively investigated. Melt temperature, melt throughput, air temperature, airflow rate, and DCD (distance of collector to die) were considered as primary process control variables. The produced webs were characterized for fiber diameter, bulk density, air permeability, hydrostatic head, tensile properties, and heat shrinkage. Non-round and curly or twisted fibers were observed in the bico PP/PTT webs by SEM (scanning electrical microscope). The PTT grade studied is quite suitable for the meltblown process. The PTT/PP-based bico webs showed enhanced barrier properties and heat resistance. © 2002 John Wiley & Sons, Inc. J Appl Polym Sci 83: 1280–1287, 2002

Key words: bicomponent fibers; meltblowing; meltblown nonwovens; PTT (polytrimethylene terephthalate)

INTRODUCTION

Thermoplastic resins have been extruded to form fibers and webs for a number of years. The most common thermoplastics for this application are polyolefins and polyesters. Other materials, such as polyetheresters, polyamides, and polyurethanes, are also used for this purpose. Each material has its characteristic advantages and disadvantages vis-a-vis the properties desired in the final product to be made from such fibers. The term "bicomponent" usually refers to fibers that have been formed from at least two polymers extruded from separate extruders but spun together to form one fiber. The configuration of such a bicomponent (bico) fiber may be a sheath/core arrangement, wherein one polymer is surrounded by another, or a side-by-side arrangement. It is often desirable that the fabrics have the combination of the advantages of different polymers in one spun fiber. In nonwovens industries, bico fibers have been developed in recent years for meltblown and spunbond processes.

Meltblowing is a one-step process to make microfiber nonwovens directly from thermoplastic polymers with the aid of high-velocity air to attenuate the melt filaments. It has become an important industrial technique in nonwovens because of its ability to produce fabrics of microfiber structure suitable for filtration media, thermal

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Journal of Applied Polymer Science, Vol. 83, 1280–1287 (2002)

DOI 10.1002/app.2295

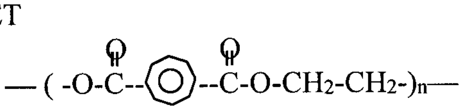
Table I Physical Properties of PET, PTT, and PBT

	Polymer		
Properties	PET (Wellman)	PTT (VFR50009)	PBT (Ticona Celanex 1300A)
Melting Point (°C)	262	230	228
Glass Transition (°C)	80	46	29
Density (g/cm^3)	1.40	1.33	1.41
Moisture Content Before Drying (%)	0.49	0.20	0.35
Moisture Content After Drying (%)	0.00	0.00	0.02
MFR* (g/10 min.)	375 @ 300°C	800 @ 300°C	216 @ 280°C

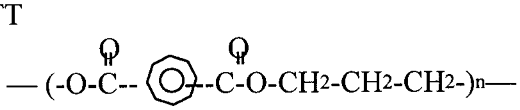
insulators, battery separators, oil absorbents, and many laminate applications. Polypropylene (PP) is the most widely used polymer for this process. Others, such as polyethylene (PE), poly(ethylene terephthalate) (PET), poly(butylene terephthalate) (PBT), and polyamide (PA) can also be used to produce meltblown webs. A lot of efforts have been made in the past 30 years on process study,[1] new resin and product development, and process improvement.[2,3]

PTT, a member of the polyester family, is based on a three-carbon diol. It bears many similarities to PET and PBT, which are based on a two-carbon diol and a four-carbon diol, respectively. The physical properties of these polymers are listed in Table I, and their chemical structures are shown below:

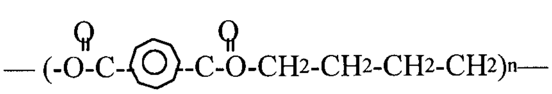
PET



PTT



PBT



EXPERIMENTAL

Trial Design and Web Preparation

Because meltblowing is a highly complex and multi-variable process for which knowledge of the mechanistic model is lacking, surface response methodology (SRM) was applied in this research to develop the mono PTT meltblown webs and study the processability of PTT.[3] Melt temperature, melt throughput, air temperature, airflow rate, and DCD (distance of collector to die) were considered as primary control variables in the process. DSC scanning and melt flow rate were measured to determine the proper experimental range of temperature. The heating rate of DSC was 10°C/min. The temperature was set from 50 to 350°C. DSC was applied to determine the melting temperature and thermal behavior or stability of the resin. Melt flow rate (MFR) or melt flow index (MFI or MI) was also measured to help determine the processing temperature. MFR is widely used in plastic industry to describe the fluidity of a polymer melt. It is a simple flow value of the amount of material extruded at a standardized temperature through a die under pressure from a set mass over a period of 10 min. The melt at the higher temperature flows more easily and corresponds to higher MFI. The resin usually is not recommended for the meltblowing process if the MFR value is too low (<100) at the processing temperature, or increasing temperature should be considered for the production as long as no oxidation occurs.

Like PET and PBT, PTT absorbs moisture, which causes hydrolytic degradation of PTT at melt processing temperatures. Drying of the polymer is required before meltblowing and the MFR measurement. The drying condition was 120°C for 3 h, which reduced moisture content from 0.22% before drying to 0.003% (30 ppm) after drying. The MFR values of PTT were 385 at 270°C and 844 at 300°C, indicating that a melt temperature of 270–300°C is suitable for the meltblowing process.

The designed processing conditions are shown in Table II: The melt throughput was 0.3–1.5 g/hole/min, the melt temperature was 270–305°C (520–580°F), air temperature was from 230–310°C (450–530°F), air flow rate was 300–700 SCFM (standard

Table II Processing Conditions for PPT (Shell VFR 50009)[a]

Sample No.	Melt Temp (°F)	Melt Temp (°C)	Throughput (g/hole/min)	Air Temp (°F)	Air Temp (°C)	Air Flow Rate (SCFM)	DCD (in.)
1	520	271	0.9	490	254	500	15
2	535	279	0.6	470	243	400	17
3	535	279	0.6	510	266	600	13
4	535	279	0.6	470	243	400	13
5	535	279	0.6	510	266	600	17
6	535	279	1.2	470	243	400	13
7	535	279	1.2	510	266	600	17
8	535	279	1.2	470	243	400	17
9	535	279	1.2	510	266	600	13
10	550	288	0.3	490	254	500	15
11	550	288	0.9	490	254	500	15
12	550	288	0.9	490	254	300	15
13	550	288	0.9	490	254	700	15
14	550	288	0.9	450	232	500	11
15	550	288	0.9	530	277	500	19
16	550	288	0.9	490	254	500	15
17	550	288	0.9	490	254	500	15
18	550	288	1.5	490	254	500	15
19	565	296	0.6	470	243	400	13
20	565	296	0.6	510	266	600	17
21	565	296	0.6	470	243	400	17
22	565	296	0.6	510	266	600	13
23	565	296	1.2	470	243	400	17
24	565	296	1.2	510	266	600	13
25	565	296	1.2	470	243	400	13
26	565	296	1.2	510	266	600	17
27	580	304	0.9	490	254	500	15

[a] Air gap/setback = 0.8/1.0 mm.

cubic/foot per minute), and DCD was 11–19 inches. Based on preparation of the mono PTT webs, PTT/PP bicomponent meltblown webs were made at the ratios 25/75, 50/50, and 75/50 (Table III). The web basis weight was controlled to reach the same target weight at 1 oz/yd^2 (39.1 g/m^2). The grades of PTT and PP were Shell VFR 50009 and Exxon 3546G, respectively.

Tests and Characterization

Testing of these mono- and bicomponent webs included basis weight, bulk density, fiber diameter, air permeability (ASTM D 737), tensile properties (ASTM D 1117), and hydrostatic head (IST 80.4-92). The fiber diameter was measured by optical microscopy with the software of Image Pro. Scanning electron microscopy (SEM) was applied to examine the fiber structure of mono- and bicomponent webs. Heat resistance was evaluated by heat shrinkage of the web, which was determined by the geometric average of heat shrinkage in the machine direction and heat shrinkage in the cross-machine direction as shown in the following equation:

$$\text{average heat shrinkage} = (\text{MD shrinkage} \times \text{CD shrinkage})^{1/2} \quad (1)$$

RESULT AND DISCUSSION

The measured web properties of all the mono PTT webs are shown in Table IV. Based on all the processing conditions and corresponding web properties, a dynamic relationship of web properties to the processing conditions was built by SRM.[2] Our previous research showed that it is an effective and efficient statistical method for systematically studying and modeling of the mono

Table III Processing Conditions for Bico PPT/PP Webs (Shell PTT 50009/Exxon PP 3546G)[a]

Sample No.	PTT/PP Ratio	Melt Temp (°F)		Air Temp (°F)	Throughput (g/hole/min)	DCD (in.)	Air Flow Rate (SCFM)
		PTT	PP				
bico1a	75/25	550	520	500	0.6	12	400
bico1b	75/25	550	520	500	0.6	14	500
bico1c	75/25	550	520	500	0.6	12	600
bico1d	75/25	550	520	500	0.6	19	500
bico2a	50/50	550	520	500	0.6	8	300
bico2b	50/50	550	520	500	0.6	10	400
bico2c	50/50	550	520	500	0.6	12	500
bico3a	25/75	550	520	500	0.6	10	400
bico3b	25/75	550	520	500	0.6	10	300
bico3c	25/75	550	520	500	0.6	8	300

[a] Air gap/setback = 0.8/1.0 mm.

meltblown process. The relationship of the response characteristics to the processing conditions at melt temperature and throughput of 288°C (550°F) and 0.8 g/hole/min, air temperature of 249°C (480°F), flow rate of 630 SCFM, and DCD of 13 inches is shown in Figure 1. As seen in

Table IV Characterization of PTT Mono MB Webs

Sample ID	Bulk Den. (g/cm^3)	Fiber Diameter		Break Ext. (%)	Tenacity (mN/tex)	Air Perm. ($ft^3/ft^2/min$)	Hydro-head (cm)
		Average (μm)	CV (%)				
1	0.115	4.76	27.4	58.54	1.08	314.6	12.03
2	0.085	3.84	18.1	88.10	0.71	318.8	13.10
3	0.117	2.86	25.6	30.95	2.15	128.6	19.20
4	0.103	2.88	30.8	49.04	0.59	161.6	15.93
5	0.112	2.87	22.8	33.18	1.61	132.8	17.07
6	0.098	3.77	28.2	52.98	1.41	321.0	9.43
7	0.101	2.87	26.6	40.28	1.78	266.0	10.23
8	0.083	3.83	25.4	68.06	1.12	411.6	9.13
9	0.112	2.41	23.6	34.58	2.50	212.2	9.60
10	0.110	2.56	40.9	29.80	2.00	85.8	13.77
11	0.101	2.98	22.3	41.64	1.20	186.2	9.97
12	0.083	3.99	27.5	81.60	1.26	346.6	9.13
13	0.116	2.39	21.6	26.50	3.23	132.4	11.23
14	0.111	2.99	28.8	27.06	2.39	149.0	11.53
15	0.088	2.67	31.0	50.88	1.09	201.2	9.90
16	0.105	3.24	19.7	38.18	1.22	170.0	10.00
17	0.104	3.18	20.0	33.22	1.56	168.8	9.93
18	0.101	3.22	29.6	34.60	1.73	239.4	9.67
19	0.110	3.04	22.4	20.36	1.34	158.4	9.50
20	0.113	2.02	25.2	26.98	2.07	110.5	11.40
21	0.106	3.36	19.0	25.14	0.92	112.8	9.90
22	0.118	2.21	22.1	15.52	3.65	71.4	11.77
23	0.087	3.00	23.2	32.02	0.90	192.8	9.13
24	0.112	2.22	22.4	24.70	3.33	133.4	11.03
25	0.099	2.83	20.5	32.72	1.55	203.4	9.53
26	0.101	2.51	27.4	30.26	2.15	150.8	10.30
27	0.099	3.01	20.0	30.18	2.62	128.2	10.50

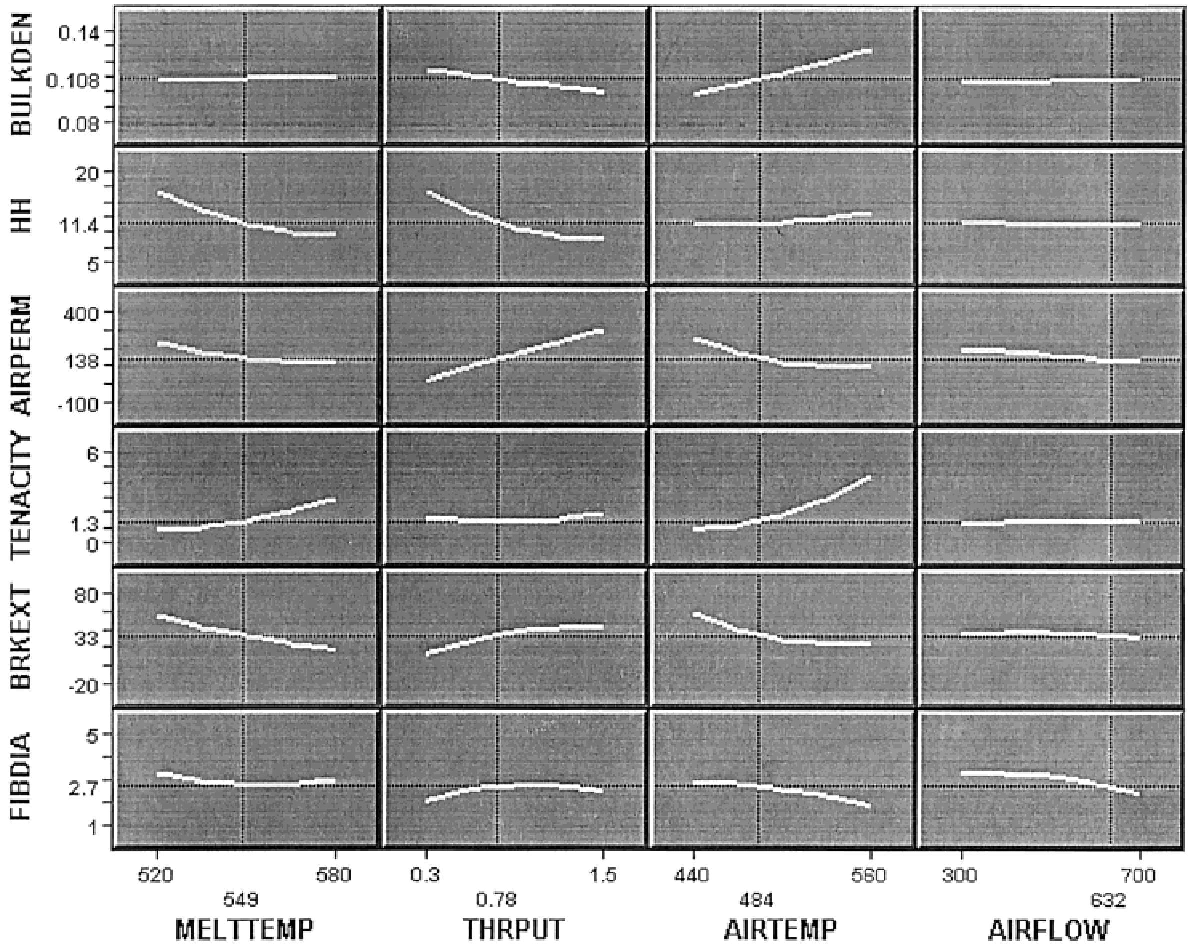


Figure 1 Dynamic Relationships of Properties to Processing Conditions of the PTT Webs.

the first column in Figure 1, an increase in melt temperature for this case results in decreases in fiber diameter, air permeability, breaking elongation, and hydrostatic head. An increase of tenacity was observed as melt temperature increased. The second column shows the effect of melt

Table V Properties of Bicomponent PTT/PP Webs

Sample ID	Bulk Den. (g/cm^3)	Fiber Diameter Average (μm)	Fiber Diameter CV (%)	Break Ext. (%)	Tenacity (mN/tex)	Air Perm. (ft^3/ft^2/min)	Hydro-head (cm)
bico1a	0.110	3.00	22.5	12.10	2.33	110.2	38.77
bico1b	0.109	2.47	26.4	10.42	2.87	90.2	40.37
bico1c	0.111	1.71	21.4	11.82	3.39	80.3	42.00
bico1d	0.092	2.27	25.6	20.32	1.45	82.3	31.77
bico2a	0.112	2.22	24.9	37.02	5.97	95.6	41.30
bico2b	0.103	2.12	23.9	15.74	3.81	82.2	45.87
bico2c	0.098	1.92	24.1	14.04	2.90	85.2	45.17
bico3a	0.107	1.98	26.1	10.96	3.58	53.1	59.10
bico3b	0.111	2.34	30.7	15.10	4.60	73.5	52.10
bico3c	0.116	2.20	23.4	12.32	12.84	62.5	59.17

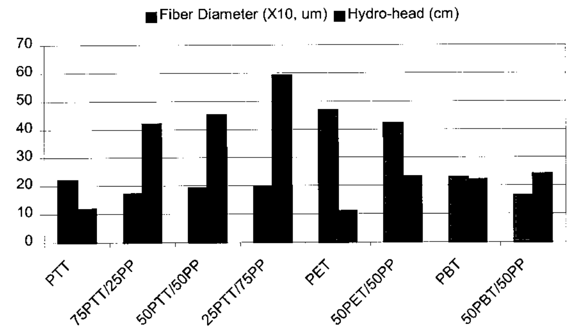


Figure 2 Hydrohead and Fiber Diameter for Bico Meltblown Webs (Melt throughput = 0.6 g/hole/min).

throughput. As the melt throughput increases, fiber diameter increases, and bulk density and hydrohead decrease. The third and fourth columns illustrate that as air temperature and flow rate increase, fiber diameter decreases and tenacity increases.

The properties of bico PTT/PP webs are shown in Table V. Fine meltblown fabrics of fiber diameter from 1.71 to 2.41 μm were produced at melt throughput of 0.6 g/hole/min for mono PTT and bico PP/PTT, as shown in Figure 2. The bico PTT/PP showed better barrier properties than mono PTT meltblown webs in the fiber diameter range of 1.71 to 2.41 μm. The hydrohead of bico PTT/PP webs were about four times higher than PTT mono webs. Compared with PET mono and bico webs, the hydrohead of PTT/PP bico webs was also higher than that of mono PET and bico PET/PP webs. The reason for higher barrier properties of PTT/PP bico webs may result from structure and morphology of the bico fiber.

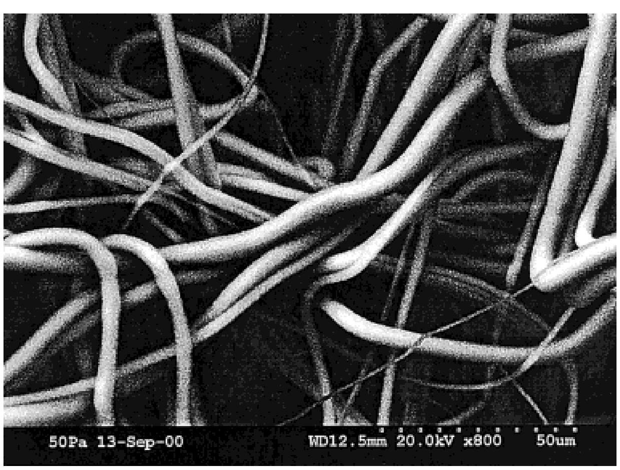


Figure 3 SEM Photograph for PTT Mono Meltblown Web.

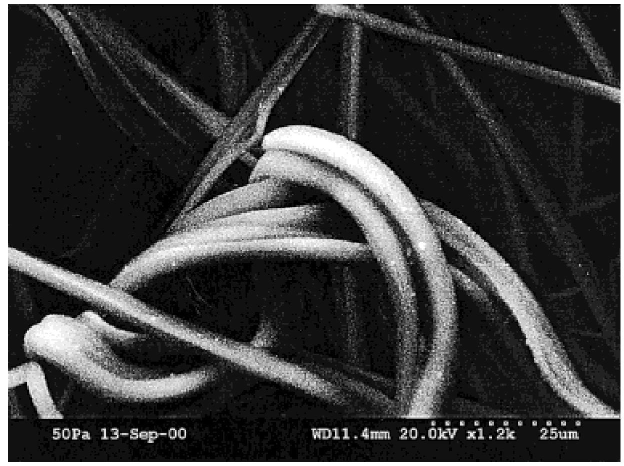


Figure 4 Bico Structure of 25PTT/75PP Meltblown Web.

SEM photographs of PTT mono meltblown webs are shown in Figure 3. It can be seen that the PTT mono meltblown fibers have a round and smooth morphology as do other mono meltblown fibers. SEM pictures of PTT/PP bico meltblown webs are shown in Figure 4. Nonround cross-sections and more crimped or twisted fibers were observed, which might be due to thermal properties and rheological gradients of the melts on each side of the bico fiber in the web-forming process. Also, from Figure 5, some curly fibers were observed for bico PTT/PP webs, which may be the cause of better barrier properties of the webs. The curly fibers (crimped or twisted) would result in a longer tortuous path through which gas would pass.

As shown in Figure 6, the bico 75PP/25PTT web resulted in negligible shrinkage when sub-

Figure 5 Curly Filaments in 50PTT/50PP Bico Meltblown Web.

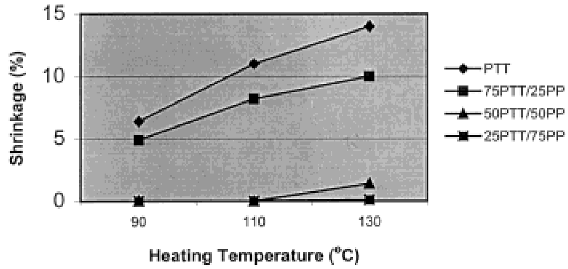


Figure 6 Heat Shrinkage of PTT Mono and PTT/PP Bico Meltblown Webs at 90°C, 110°C, 130°C for 7 Minutes.

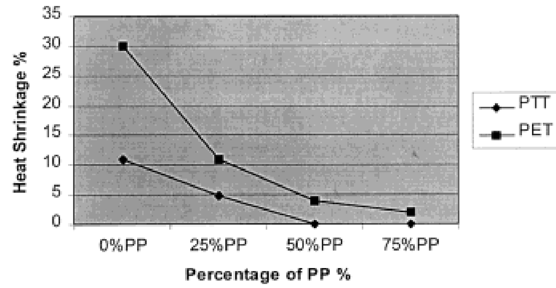


Figure 8 Comparison of Heat Shrinkage for PTT and PET Bico Meltblown Webs at 90°C for 7 Minutes.

jected to heat, without any tension at temperatures of 90, 110, and 130°C for 7 min. The 50PP/50PTT web resulted in only slight shrinkage over the temperature/exposure conditions of 90–130°C and <2% shrinkage at 130°C. On the other hand, the 25PP/75PTT web also showed enhanced heat resistance and the heat shrinkage was notably reduced compared to mono PTT. PP did not show the shrinkage at 90 and 130°C.

The heat shrinkage of mono PET and PTT meltblown webs is shown in Figure 7. The PET web was also subjected to heat without any tension at temperatures of 90, 110, and 130°C for 7 min and showed higher shrinkage (16–28%) than that of PTT (6.4–14%). The high shrinkage of 100% PET meltblown webs may be readily explained by theory. Because PET crystallizes relatively slowly in the melt blowing process, solidification occurs before the small amount of stress-induced orientation can result in significant crystallization, not as occurs in conventional high-speed melt spinning and in some spunbond processes. The lower shrinkage for PTT resulted from the fast crystallization of PTT. The heat shrinkage for three different ratios of PTT/PP and PET/PP bico meltblown webs conditioned at 90°C

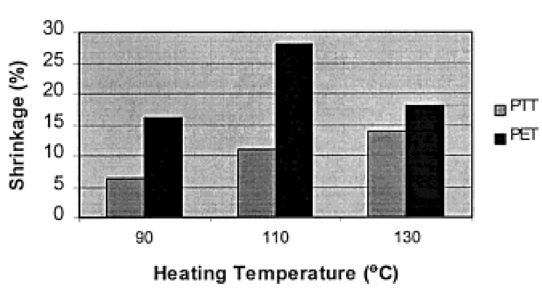


Figure 7 Comparison of Heat Shrinkage for PTT and PET Mono Meltblown Webs at 90°C, 110°C, 130°C for 7 Minutes.

for 7 min is compared in Figure 8. PTT/PP bico webs show lower heat shrinkage than PET/PP bico webs. As shown in Figure 8, 50% PP may result in shrinkage-free PTT/PP bico meltblown webs, which may expand the application of PTT in some areas requiring dimensional stability.

SUMMARY

Mono- and bi-component PTT meltblown fiber webs were produced on the Reicofil® side-by-side bicomponent meltblown line. SRM was applied for the web development with melt throughput from 0.3 to 1.5 g/hole/min, melt temperature from 270 to 305°C (520 to 580°F), air temperature from 230 to 310°C (450 to 530°F), air flow rate from 12.5 to 29.2 SCFM/in, and die DCD from 11 to 19 inches. The fiber diameter obtained was in the range of 1.71 to 4.76 μm. PTT and bico PTT/PP exhibited excellent meltblown processability and web quality. Compared with conventional (mono) round and smooth meltblown fibers, the bico PTT/PP webs showed the structure of nonround cross-section and twisted fibers. The barrier properties and heat shrinkage resistance of the bico webs were notably improved.

The authors thank Shell Chemical Company for financial support for this project and TANDEC operators for web processing.

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