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PROCESSING STUDY OF A POLYIMIDESULFONE ADHESIVE

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INTRODUCTION

A polyimidesulfone which shows promise as a structural adhesive and matrix resin[1] was recently prepared at NASA-Langley Research Center. This material is a high temperature thermoplastic with a reported glass transition temperature of 273°C (522°F). Even though this polymer contains the sulfone unit in its backbone, it is not susceptible to attack by common solvents as are the commercial sulfones. The starting materials for this polymer are also readily available at reasonable costs ~$40/kg (~$10/lb).

The initial study of the polyimidesulfone[1] indicated the system should perform well as a structural adhesive at elevated temperature. However no in-depth bonding process cycle development was performed. This report details the results of a study to better understand the parameters that effect the adhesive properties of the polymer for titanium alloy adherends. The study included tape preparation, use of primers, and determination of effective press and simulated autoclave bonding conditions. The polymer was characterized at various stages using Fourier Transform Infrared Spectrophotometry (FTIR), glass transition temperature determination, flow characterization, and weight loss measurements. Finally the adhesive strengths of the fabricated lap shear specimens were mechanically determined.

EXPERIMENTAL

Preparation of the Polymer. The monomers used to prepare the polyimidesulfone (PISO2) were 3,3',4,4'-benzophenonetetracarboxylic dianhydride (BTDA) and 3,3'-diaminodiphenylsulfone (3,3'DDS). The polymer grade BTDA, m.p. 215°C (419°F), was used as-received from Gulf Chemicals.

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The 3,3'DDS was used as-received from FIC Corporation, m.p. 165-167°C (329-333°F). The polymer, prepared as a 25 percent solids solution in diglyme,[1] was used to prepare the adhesive tape for this study.

Characterization. Lap shear strength was obtained according to ASTM D-1002 using a Model TT Instron Universal Testing Machine. The average lap shear strengths reported represent at least four lap shear specimens tested for any one condition. Specimens were soaked at temperature in a clam-shell, quartz-lamp oven and were held at temperature for ten (10) minutes prior to testing. Temperatures were controlled to within ±3°C (±5°F) for all tests. Glass transition temperatures (T_g) for the adhesive of fractured lap shear specimens were determined by thermomechanical analysis (TMA) on a DuPont 943 Analyzer in static air at a heating rate of 5°C/min (9°F/min) using a hemispherical probe with a 15g mass. Infrared spectra were obtained using a Nicolet Model 3600 Fourier Transform Infrared Spectrophotometer (FTIR) using a diffuse reflectance (DR) technique.

The percent weight loss, often referred to as volatile content, was determined for the adhesive tapes by recording the weight loss after heating a sample in a forced-air oven at 343°C (650°F) for 0.5 hr. Measured weight included the glass cloth and polymer. The tapes were previously conditioned by heating the sample in a forced-air oven at 100°C (212°F) for one hour.

The adhesive's flow characteristics were determined by rapidly heating a 1.6 cm (0.625 in.) diameter adhesive tape in a press and holding for 2 min at temperature under 160 psi pressure. The percent flow was obtained by measuring the average diameter of the pressed tape relative to the tape's initial diameter and multiplying by 100.

Preparation of Adhesive Tape. The PISO2 adhesive tape was prepared by brush-coating the 25% solids solution in diglyme on to 112 E-glass cloth with A-1100 finish (γ-aminopropylsilane). Prior to coating, the glass cloth (tightly mounted in a metal frame) was initially oven-dried for 1 hr at 100°C (212°F) then heated overnight at ~60°C (140°F). The 0.01 cm (0.004 in) thick glass cloth served as a carrier for the adhesive as well as for bondline thickness control and an escape channel for solvent and reaction by-products. Coatings of the polymer solution were applied to the glass cloth until a thickness of 0.020-0.025 cm (0.008-0.010 in) was obtained. After each application, the tape was air-dried ~0.5 hr, placed in a forced-air oven, and exposed to the following schedule:

(1) Room temperature (RT) → 60°C (140°F), hold 5 min

(2) 60°C (140°F) → 100°C (212°F), hold 1 hr

(3) 100°C (212°F) → 160°C (320°F), hold 1 hr

After the above treatment, the adhesive tape contained 0.02 g/cm^2 of polymer plus remaining solvent. Some of the tape was subsequently heat treated further. Portions of this adhesive tape were subsequently removed after each successive heat treatment consisting of 15°C (27°F) steps above the 160°C (320°C) and held for 0.5 hr up to a temperature of 235°C (455°F). The last tape prepared was further held for a total of 17 hrs at 235°C (455°F).

Adhesive Bonding. The prepared adhesive tapes were used to bond titanium adherends (Ti 6Al-4V, per Mil-T-9046E, Type III Comp. C) with a nominal thickness of 0.13 cm (0.50 in). The four-fingered Ti(6Al-4V) panels were surface treated with a Pasa-Jell 107* treatment to form a stable oxide

*Trade name for a titanium surface treatment available from Semco, Glendale, CA.

on the surface. The treated adherends were primed within one hour of the surface treatment by applying a thin coat, 0.005 cm (0.0002 in) of the 25% solids PISO2 solution on the surface to be bonded and heating in a forced-air oven for 5 min at 60°C (140°F), 1 hr at 100°C (212°F), 1 hr at 160°C (320°F), and 0.5 hr at 175°C (347°F). The primed adherends were stored in a polyethylene bag and then placed in a desiccator until needed. Lap shear specimens were prepared by inserting the adhesive tape between the primed adherends using a 1.27 cm (0.50 in) overlap (ASTM D-1002).

Several bonding cycles were used during this study. The initial cycle selected (Cycle 1) was based on applying the pressure at the temperature where the PISO2 starts to soften, i.e., ~250°C (482°F) (ref. 1, figs. 4 and 5) and thermally exposing to a maximum cure temperature of 343°C (650°F). The following processing cycles were used:

Cycle 1

(1) Contact pressure, heating rate 4°C/min (7°F/min), RT → 343°C (650°F), apply 1.38 MPa (200 psi) at 250°C (482°F)

(2) Hold 5 min at 343°C (650°F)

(3) Cool under pressure to ~150°C (302°F) and remove from bonding press

Preheated Press Cycle

(1) Preheat press to 343°C (650°F)

(2) Place assembly to be bonded in press

(3) When assembly reaches 343°C (650°F), apply 1.38 MPa (200 psi) and hold for 5 min

(4) Cool under pressure to ~150°C (302°F) and remove from bonding press

Modified Cycle 1

(1) Contact pressure, heating rate of 4°C/min (7°F/min), RT → 343°C (650°F), apply 0.34 MPa (50 psi) at 250°C (482°F)

(2) After 5 min at 343°C (650°F), apply 1.38 MPa (200 psi)

(3) Hold another 5 min at 343°C (650°F)

(4) Cool under pressure to ~150°C (302°F) and remove from press

Simulated Autoclave Cycle (Mod. Cycle 1 with Full Vacuum)

Cycle 2

(1) Contact pressure, heating rate of 7°C/min (13°F/min), RT → 325°C (617°F), apply 1.38 (200 psi) at 280°C (536°F)

(2) Hold 15 min at 325°C (617°F)

(3) Cool under pressure to ~150°C (302°F) and remove from press

Cycle 3

Same as Cycle 2 except a heating rate of 4°C/min (7°F/min) was used

The above processing cycles together with (w) or without (w/o) an adhesive primer were used to determine the effects on the lap shear strength (LSS) of various heat treated adhesive tapes, of a slow processing cycle and rapid processing (preheated press) cycle with and without primer, of a vacuum (simulated autoclave) processing cycle, and a processing cycle previously reported.[1]

RESULTS AND DISCUSSION

Resin Chemistry and Properties. The proposed reaction for the formation of the subject PISO2 is shown in Figure 1. The reaction to form

the polyamide acid was performed in diglyme because this solvent has been reported to produce polymers with good adhesive strengths[2] and to allow facile solvent removal during processing. The meta-oriented diamine was chosen because the meta-diamines have exhibited improved processability and adhesive strength compared to para-orient diamines.[3] During bonding, the thermal imidization of the polyamide acid results in linear, high molecular weight polyimides which possess adequate flow to allow for thermoplastic processing. Part of the thermoplasticity relates to the flexibility of the polymer chain due to the meta linkages in the diphenylsulfone portion.[1] The thermooxidative stability, the T_g of a PISO2 film, the effects of solvent exposure on films of PISO2, and the polymer softening as determined by parallel plate plastometer techniques are given in reference 1.

The T_g's determined on the adhesive from the center portion of the fractured lap shear specimens ranged from 215°C (419°F) to 256°C (493°F). These values are lower than the 273°C (523°F) T_g reported for the PISO2 film which had been treated to 300°C (572°F) for 1 hr.[1] These data are shown in Tables I-IV. This type of behavior was noted previously for the NR-150B2 adhesive, a thermoplastic system, where the T_g measured for the adhesive in the bondline (ref. 4, Table XV, p. 49) ranged from 273°C (523°F) to 300°C (572°F) compared to that measured for a moulding, 332°C (630°F) (ref. 4, p. 3) of the same composition.

For the tests conducted with the 175°C (347°F), 0.5 hr polyimidesulfone tape, the T_g for the fractured lap shear specimens tested at RT and 232°C (450°F) were about the same, ~218°C (424°F) (Table 1). Also there was no difference in the measured T_g of those tested at RT or 232°C (450°F) with or without primer. There appears to be a general increase in T_g with

increased staging of the adhesive tapes as noted in Tables I-IV. Comparing T_g results for those bonded with the three tapes using Cycle 1, with primer, and tested at RT (Table 1) to those bonded using Mod. Cycle 1, with primer, tested at RT (Table III) shows the T_g's to be approximately the same. There was an exception for the 175°C (347°F), 0.5 hr tape results which shows a T_g of 24°C (43°F) higher for the Mod. Cycle 1. Bonding with vacuum (simulated autoclave), Table IV, had little affect on the measured T_g's of the adhesive from the fractured lap shear specimens for the three adhesive tapes, Table III.

Diffuse Reflectance FTIR (DR-FTIR) spectra of PISO2 adhesive tapes with various thermal treatments are shown in Figure 2. Of particular interest are the bands due to the amide, imide, and anhydride portions of the adhesive resin as well as those bands due to the diglyme solvent. The amide bands are located around 3320 cm^{-1} and 1540 cm^{-1}. The bands due to the imide are around 1780, 1380, and 730 cm^{-1}. Some distortions obtained by use of diffuse reflectance when analysing polyimides has been previously discussed.[5] The decrease in the size of the amide band at 3320 cm^{-1} and the disappearance of the band at 1540 cm^{-1} together with the increase in size of the imide bands (1780, 1380, and 730 cm^{-1}) suggest imidization is proceeding with increased time and/or temperature of the thermal treatment. The removal of the diglyme solvent with thermal treatment is evident by the decrease in size of the band slightly below 3000 cm^{-1} which is indicative of aliphatic carbon-hydrogen absorption. The band around 1850 cm^{-1} suggests that a small amount of anhydride is present in all three samples. Aromatic protons, previously hidden, become apparent as the solvent is lost as shown by the absorption slightly above 3000 cm^{-1}. Also notable is the

disappearance of the acid absorption at 2800-2550 cm^{-1}. Good agreement was obtained with previously reported DR-FTIR spectra for this PISO2 prepared in a similar manner.[5]

The percent weight loss (percent volatiles) determined for the series of PISO2 adhesive tapes is given in Figure 3. The colors of the original tapes change from glossy bright yellow for those treated at 160°C (320°F),1 hr and 175°C (347°F), 0.5 hr to a matt yellow to a burnt orange for that treated at 235°C (455°F) for 17 hrs. The weight loss decreases with each successive thermal treatment from 7.8 percent for the 160°C (320°F), 1 hr treatment to 1.2 percent for that treated at 235°C (455°F) for 17 hrs. The adhesive weight loss was due primarily to residual solvent, imidization by-products (water), and chain-extension by-products (water). The extent of foaming also decreases in the same order as the weight loss. Color changes from yellow-beige, to a light brown, to medium brown, and to dark brown after the heat treatments, with increasing temperature/time.

A certain amount of flow is necessary in order for an adhesive to wet the mating surfaces as well as to produce a void-free, controlled bondline. Too much flow results in voids in an adhesive which tends to weaken the bond joint. Flow measurements were made for the series of PISO2 adhesive tapes to determine which tape or tapes held the most promise for producing good adhesive bonds. Pieces of 1.6 cm (0.063 in.) diameter tapes were held under 160 psi pressure for 2 min at the temperatures of 316°C (600°F), 343°C (650°F), and 371°C (700°F). The appearance of the samples after flow determination is shown in Figure 4. Results of the flow characterization tests indicate reasonable flow (14.3 to 19.0 percent) can be obtained for those tapes treated to 175°C (347°F) for 0.5 hr. However because a primer

would most likely be used (a study to determine the effects on LSS with and without primer was performed), the tapes treated at 235°C (455°F) for 0.5 hr and 235°C (455°F) for 17 hrs. and which exhibited less flow were also further investigated as adhesive tapes in the processing study for bonding titanium adherends.

Cycle 1-Slow Process Bonding Cycle. In order to evaluate the effect of a primer on bond strength for the selected PISO2 adhesive tapes, the slow processing cycle (Cycle 1) was used to bond both primed and unprimed Ti(6Al-4V) adherends. Lap shear tests were conducted at RT and 232°C (450°F). The data are presented in Figure 5 and Table I. Also included in Table I is the bondline thickness and the type of failure, cohesive (Co) or adhesive (Ad). Bondline thicknesses obtained ranged from 0.20 cm (0.0079 in.) to 0.24 cm (0.0093 in.). No correlations or trends based on the type of failures are evident except that most failures were cohesive. The range of LSS for each condition is indicated by the dashed lines for each condition as shown in Figure 5. The use of a primer appears to result in increased LSS for those tested at RT for all three tapes. However, those tested at 232°C (450°F) produce the same strengths whether a primer is or is not used. At this point in the study, the best compromise as indicated by the LSS at RT and 232°C (450°F) would be to use a primer and the tape treated at 235°C (455°F) for 17 hrs.

Preheated Press Bonding Cycle. Figure 6 and Table II present the lap shear test results of bonding primed and unprimed Ti(6Al-4V), using a preheated press,to determine the effect of a rapid heat-up bonding process. Reasonable success was obtained with the 175°C (347°F), 0.5 hr tape as indicated by the thin bondline and high RT LSS. However, poor results were

obtained for the 235°C (455°F), 0.5 hr tape and the 235°C (455°F), 17 hr tape as indicated by the thick bondlines and low lap shear strengths at both RT and 232°C (450°F). Again, failures were primarily cohesive and, in most cases, there were voids in the bondline. Comparing lap shear strengths for the 175°C (347°F), 0.5 hr tape for Cycle 1 and the preheat cycle indicates higher strengths for the preheat cycle for both RT and 232°C (450°F), with and without primer. A problem was encountered when attempting to bond with a preheated press. By the time full pressure could be applied, the assembly had already reached a temperature high enough for the adhesive bonding operation to result in a bondline that was thick and full of voids. Applying the pressure instantaneously would most likely have prevented the above from occurring.

Modified Cycle 1. Cycle 1 was modified by applying less pressure at 250°C (482°F), 50 psi instead of 200 psi, and then applying the 200 psi after a 5 min hold at 343°C (650°F). The reason for the modification was to improve the chances for escape of the volatiles that are still in the system or produced during further processing to 343°C (650°F). Table III presents the results for this process for tests at RT and 232°C (450°F). Failures were primarily cohesive. The primed adherend lap shear strengths for Mod. Cycle 1 are compared to Cycle 1 in Figure 7. Use of Mod. Cycle 1 results in improvement in LSS for the RT tests except for the 175°C (347°F), 0.5 hr tape. Higher strengths were also evident for the Mod. Cycle 1 at 232°C (450°F) for all three tapes. This modification of Cycle 1 resulted in improved strengths.

Simulated Autoclave Process Using Mod. Cycle 1. Because most processing of large bonded structures is accomplished in large autoclaves, a

simulated autoclave processing procedure was investigated and the results compared to the same process without the use of vacuum. This process, as a result of the vacuum, should remove the volatiles generated during the bonding procedure more easily. Lap shear specimens to be bonded were vacuum bagged and bonded in a hydraulic press. A full vacuum (~29-30" gage) was held throughout the bonding cycle. Table IV presents the data generated for this vacuum process for the same three tapes previously investigated with Mod. Cycle 1. The type of failure for all tests was primarily cohesive. Figure 8 shows a comparison of the LSS at RT and 232°C (450°F) for specimens bonded with and without vacuum using Mod. Cycle 1. No significant differences were evident except for the 175°C (347°F) 0.5 hr tape treatment specimens bonded using vacuum and tested at RT which had a higher strength, 4090 psi, than that bonded without vacuum, 3020 psi.

A processing cycle (Cycle 2) reported in ref. 1 was used to bond Ti(6Al-4V) with the 175°C (347°F), 0.5 hr tape. The same cycle, except for the heating rate (Cycle 3), was also used for a comparison of the effects of heating rate. Results are given in Table V for tests at RT and 232°C (450°F) and are also shown in Figure 9 which includes LSS for Cycle 1 and Mod. Cycle 1. No significant differences in strengths were obtained for either Cycle 2 or Cycle 3. The data for Cycle 2 obtained in this study and that reported in ref. 1 do not compare favorably, i.e., 2970 psi compared to 4150 psi at RT and 1110 psi compared to 2620 psi at 232°C (450°F). Possible reasons responsible for this disagreement are subtle differences in adhesive resin preparation, differences in tape preparation differences in adherend preparation, and differences in bonding techniques. As presented in Figure 9, Mod. Cycle 1 produced the highest 232°C (450°F) strength of the four

cycles shown, [1640 psi at 232°C (450°F) using the 175°C (347°F), 0.5 hr tape].

Although the effect of a postcure on the LSS had not been determined in this study, the 232°C (450°F) thermal aging data presented in reference 1, Table II, indicates the possibility of improving the high temperature LSS using a high temperature postcure. The 232°C (450°F) LSS increased from 2920 psi for 1000 hrs of thermal aging at 232°C (450°F) to 3560 psi for 5000 hrs aging.

SUMMARY

A thermoplastic polyimidesulfone (PISO2) recently prepared at NASA-Langley Research Center has shown promise as a structural adhesive. The high molecular weight, linear aromatic system has been shown to be flexible, tough, solvent resistant, and thermooxidatively stable. Because of these properties and the indicated initial potential of the polymer as a structural adhesive, a bonding process cycle development was performed to better understand the parameters that affect the adhesive properties of the polymer.

Flow characteristics were determined for a series of thermally treated PISO2 adhesive tapes which indicated adequate flow was obtainable for producing good adhesive bonds.

FTIR spectra obtained for these adhesive tapes showed good agreement with data previously reported for this PISO2 adhesive for similarly prepared tapes. The spectra indicated the loss of solvent and imide formation as a result of thermally treating the adhesive tape.

Measured glass transition temperatures (T_g) of the adhesive taken from the center portion of fractured lap shear specimens ranged from 215°C (419°F) to 256°C (493°F) and were lower than the T_g reported for a film treated to 300°C (582°F) for one hour. The T_g's measured for the adhesive of the fracture lap shear specimens generally increased with the increased temperature and/or time of thermal pretreatment given to the adhesive tape.

The use of a PISO2 primer when bonding titanium with Cycle 1 resulted in increased lap shear strength at room temperature (RT) compared to that without a primer; however, the results at 232°C (450°F) were the same with or without a primer.

In general, higher lap shear strengths were obtained at RT and 232°C (450°F) for the Modified Cycle 1 as compared to Cycle 1, indicating the importance of applying sufficient pressure at the right moment in a processing cycle.

Using the same processing cycle and pretreated adhesive tapes for bonding titanium with vacuum (simulated autoclave bonding) or without vacuum, resulted in little, if any, difference in the lap shear strengths at RT and 232°C (450°F).

The best compromise of lap shear strength values at RT and 232°C (450°F) were obtained with Mod. Cycle 1 and the tape pretreated at 235°C (455°F) for 17 hrs [3650 psi at RT and 2870 psi at 232°C (450°F)]. However, as indicated by the beneficial effects of thermal aging reported in the literature, higher strengths could possibly be obtained with use of a high temperature postcure.

The processability of PISO2 as an adhesive for potential structural applications has been shown. The availability and low cost of the starting

materials for the preparation of PISO2 make it attractive for commercial use.

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TABLE I. - TEST RESULTS FOR PISO2 BONDED TITANIUM USING CYCLE 1[a] WITH AND WITHOUT PRIMER

Tape treatment	w or w/o primer	Test Temperature		Avg. LSS psi	Failure type,[b] percent Co/Ad	Avg. bondline thickness,		Glass transition temperature, T_g,	
		°C	°F			cm	in.	°C	°F
175°C(347°F),0.5 hr	w/o	RT	RT	2730	70/30	0.21	0.0083	218	424
		232	450	1170	59/41	0.20	0.0081	219	426
	w	RT	RT	3210	94/6	0.21	0.0082	217	423
		232	450	1020	70/30	0.20	0.0080	216	421
235°C(455°F),0.5hr	w/o	RT	RT	2000	28/72	0.23	0.0090	--	--
		232	450	1360	35/65	0.22	0.0087	--	--
	w	RT	RT	2720	55/45	0.23	0.0090	252	486
		232	450	1480	92/8	0.24	0.0093	--	--
235°C(455°F),17 hr	w/o	RT	RT	1820	32/68	0.20	0.0080	--	--
		232	450	2260	89/11	0.20	0.0079	--	--
	w	RT	RT	3040	79/21	0.20	0.0079	250	482
		232	450	2380	100/0	0.21	0.0083	--	--

[a] Cycle 1: Contact pressure, heating rate 4°C/min (7°F/min), RT→343°C (650°F), apply 1.38 MPa (200 psi) at 250°C (482°F); hold 5 min at 343°C (650°F); cool under pressure to ~150°C (302°F) and remove.

[b] Cohesive failure-Co, adhesive failure-Ad.

TABLE II. - TEST RESULTS FOR PISO2 BONDED TITANIUM USING PREHEATED PRESS CYCLE[a] WITH AND WITHOUT PRIMER

Tape treatment	w or w/o primer	Test Temperature		Avg. LSS psi	Failure type,[b] percent Co/Ad	Avg. bondline thickness,		Glass transition temperature, T_g,	
		°C	°F			cm	in.	°C	°F
175°C(347°F),0.5 hr	w/o	RT	RT	4220	94/6	0.09	0.0036	--	--
		232	450	2160	70/30	0.09	0.0035	--	--
	w	RT	RT	4070	100/0	0.10	0.0038	244	471
		232	450	1770	78/22	0.08	0.0033	--	--
235°C(455°F),0.5hr	w/o	RT	RT	1530	40/60	0.36	0.0144	--	--
		232	450	830	38/62	0.38	0.0150	--	--
	w	RT	RT	1340	75/25	0.46	0.0183	239	462
		232	450	620	78/22	0.58	0.0227	--	--
235°C(455°F),17 hr	w/o	RT	RT	1870	90/10	0.35	0.0138	--	--
		232	450	940	99/1	0.39	0.0152	--	--
	w	RT	RT	1410	100/0	0.38	0.0149	251	484
		232	450	1030	100/0	0.34	0.0135	--	--

[a] Preheated Press Cycle: Preheat press to 343°C (650°F), place assembly to be bonded in press, when 343°C (650°F) again reached, apply 1.38 MPa (200 psi) and hold for 5 min, cool under pressure to ~150°C (302°F) and remove.

[b] Cohesive failure-Co, adhesive failure-Ad.

TABLE III. - TEST RESULTS FOR PISO2 BONDED TITANIUM USING MODIFIED CYCLE 1[a] WITH PRIMER

Tape treatment	Test Temperature		Avg. LSS psi	Failure type,[b] percent Co/Ad	Avg. bondline thickness,		Glass transition temperature, T_g,	
	°C	°F			cm	in.	°C	°F
175°C(347°F),0.5 hr	RT	RT	3020	81/19	0.28	0.0090	241	466
	232	450	1640	99/1	0.22	0.0088	--	--
235°C(455°F),0.5hr	RT	RT	3260	57/43	0.19	0.0075	243	469
	232	450	2430	92/8	0.20	0.0077	--	--
235°C(455°F),17 hr	RT	RT	3650	87/13	0.18	0.0072	256	493
	232	450	2870	100/0	0.20	0.0078	--	--

[a] Mod. Cycle 1: Contact pressure, heating rate of 4°C/min (7°F/min), RT→343°C (650°F), apply 0.34 MPa (50 psi) at 250°C (482°F), after 5 min at 343°C (650°F), apply 1.38 MPa (200 psi), hold another 5 min, cool under pressure to ~150°C (302°F) and remove.

[b] Cohesive failure-Co, adhesive failure-Ad.

TABLE IV. - TEST RESULTS FOR PISO2 BONDED TITANIUM USING MODIFIED CYCLE 1 WITH VACUUM[a]

Tape treatment	Test Temperature		Avg. LSS psi	Failure type,[b] percent Co/Ad	Avg. bondline thickness,		Glass transition temperature, T_g,	
	°C	°F			cm	in.	°C	°F
175°C(347°F),0.5 hr	RT	RT	4090	99/1	0.22	0.0086	227	441
	232	450	1720	96/4	0.23	0.0089	--	--
235°C(455°F),0.5hr	RT	RT	3230	64/36	0.21	0.0084	241	466
	232	450	2420	96/4	0.22	0.0085	--	--
235°C(455°F),17 hr	RT	RT	3080	61/39	0.20	0.0081	249	480
	232	450	2620	95/5	0.21	0.0084	--	--

[a] Modified Cycle 1 with full vacuum (29-30" gage) throughout cycle.

[b] Cohesive failure-Co, adhesive failure-Ad.

TABLE V. - TEST RESULTS FOR PISO2 BONDED TITANIUM FOR CYCLE 2[a] AND CYCLE 3[b] USING THE 175°C (347°F) TAPE

Processing Cycle	Test Temperature		Avg. LSS psi	Failure type,[c] percent Co/Ad	Avg. bondline thickness,		Glass transition temperature, T_g,	
	°C	°F			cm	in.	°C	°F
2	RT	RT	2970	95/5	0.22	0.0086	215	419
	232	450	1110	92/8	0.21	0.0084	--	--
3	RT	RT	3010	94/6	0.22	0.0086	230	446
	232	450	840	80/20	0.22	0.0086	--	--

[a] Cycle 2: Contact pressure, heating rate of 7°C/min (13°F/min), RT→325°C (617°F), apply 1.38 MPa (200 psi) at 280°C (536°F), hold 5 min at 325°C (617°F), cool under pressure to ~150°C (302°F) and remove.

[b] Cycle 3: Same as Cycle 2 except a heating rate of 4°C/min (7°F/min) was used.

[c] Cohesive failure-Co, adhesive failure-Ad.

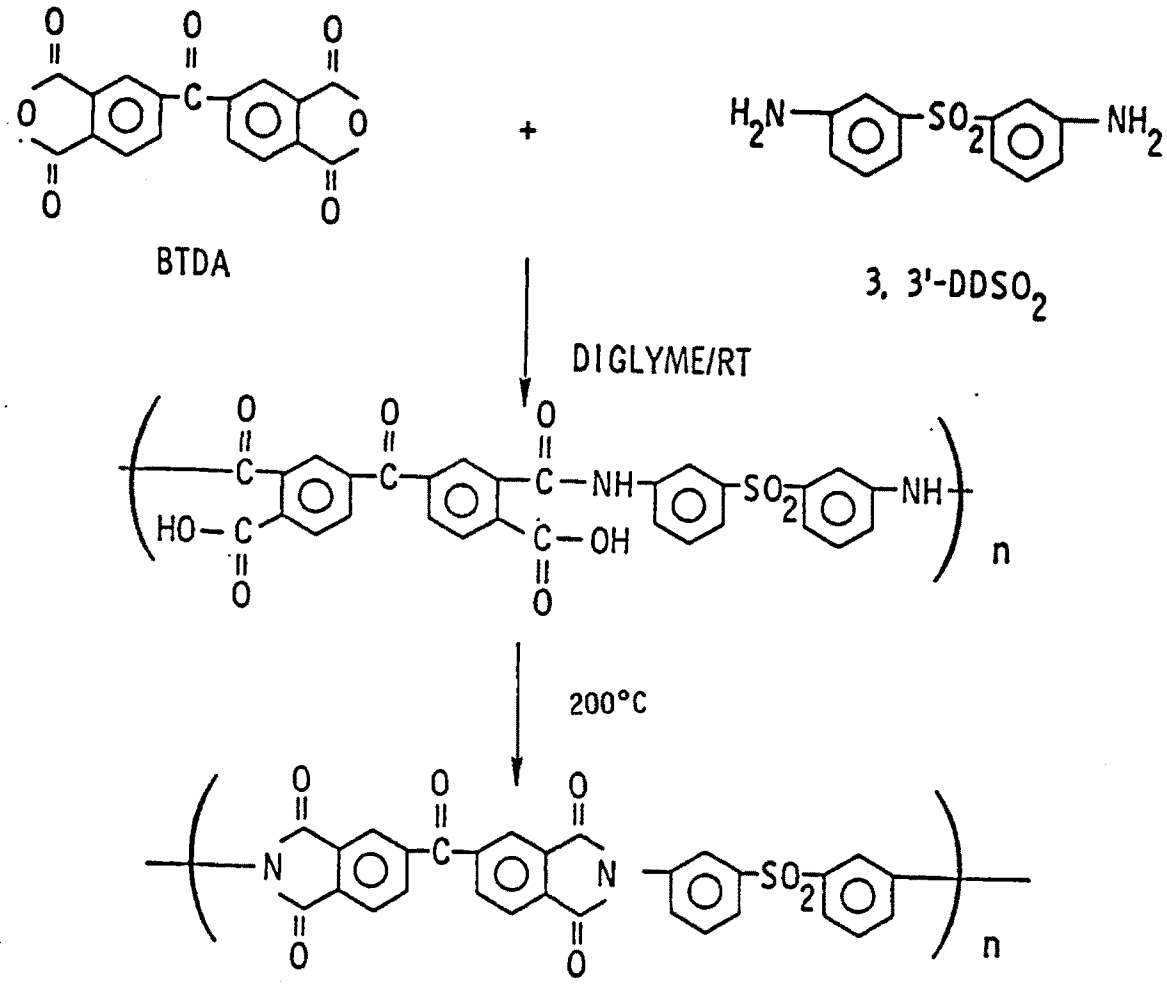


Figure 1. Polyimidesulfone preparation

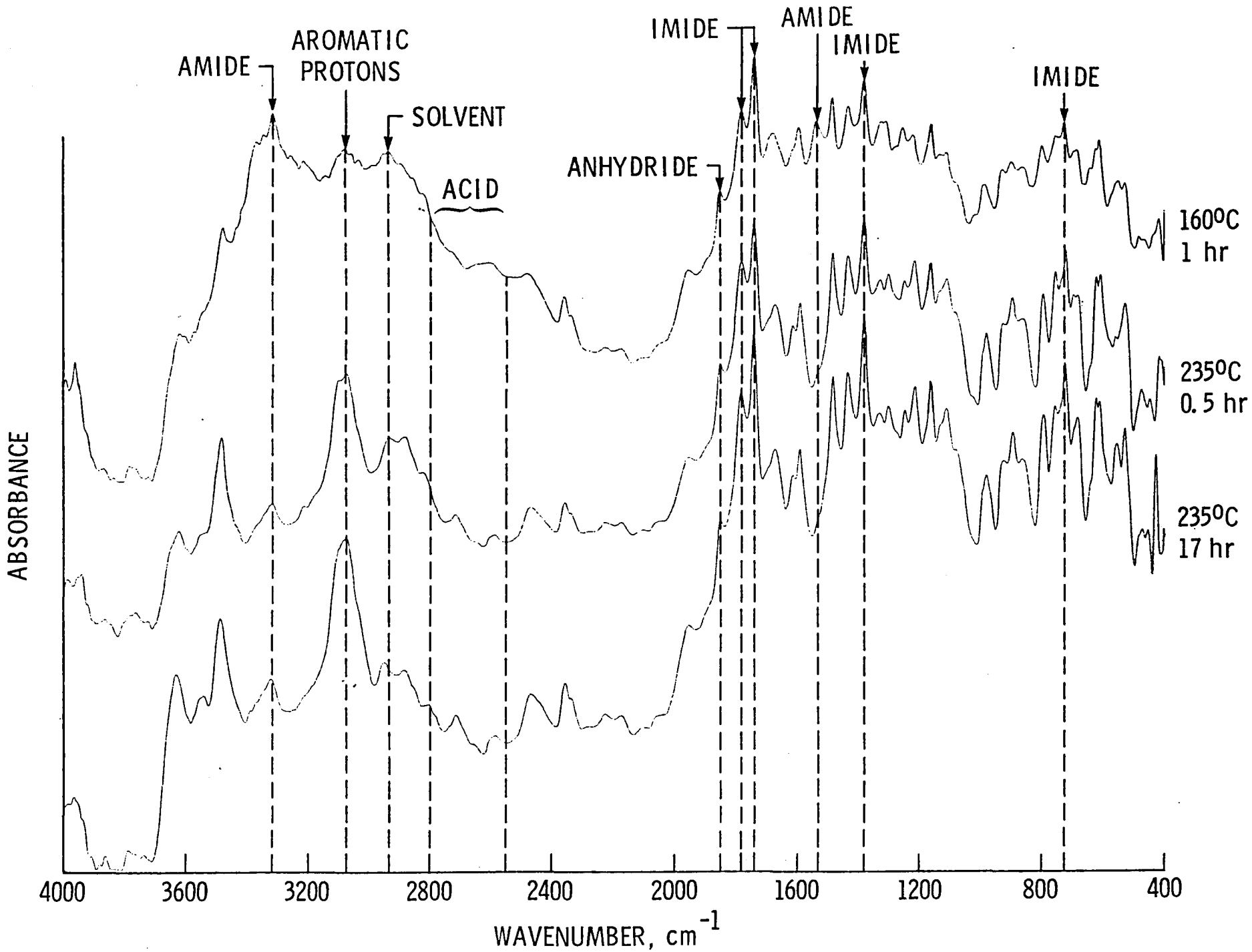
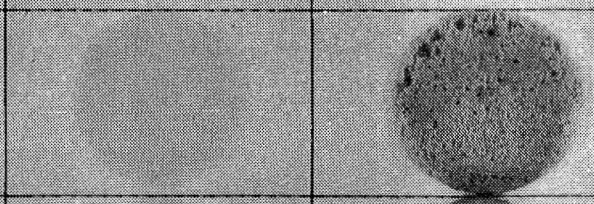
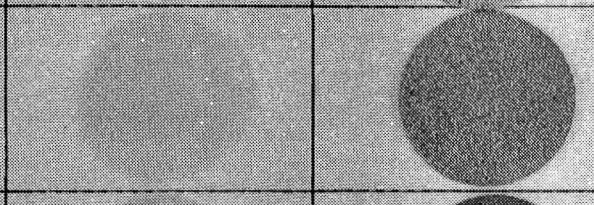
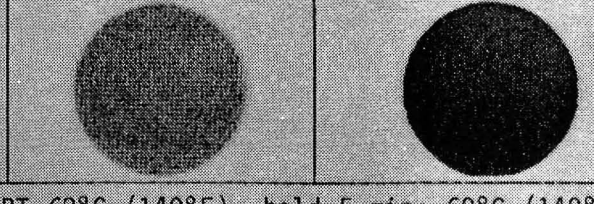
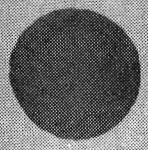
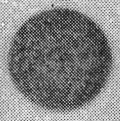
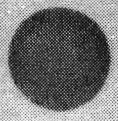
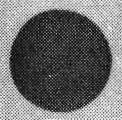


Figure 2. DR-FTIR spectra of polyimidesulfone adhesive tapes with various thermal treatments.

TAPE TREATMENT	APPEARANCE		PERCENT WT. LOSS
	INITIAL	AFTER 0.5 hr AT 343°C (650°F)	
+160°C (320°F)[a] 1 hr			7.8
+175°C (347°F) 0.5 hr			7.0
+190°C (374°F) 0.5 hr			5.5
+205°C (401°F) 0.5 hr			4.3
+220°C (428°F) 0.5 hr			3.5
+235°C (455°F) 0.5 hr			2.6
235°C (455°F) 17 hr			1.2

[a] Prior treatment: RT→60°C (140°F), hold 5 min; 60°C (140°F)→100°C (212°F), hold 1 hr.

Figure 3. Adhesive tape appearance before and after 0.5 hr at 343°C (650°F) and the percent weight loss.

TAPE TREATMENT	EXPOSURE[a]					
	316°C (600°F)		343°C (650°F)		371°C (700°F)	
	Appearance	Percent flow	Appearance	Percent flow	Appearance	Percent flow
plus 160°C (320°F) 1 hr[b]		25.4		28.6		--
Same as above plus 175°C (347°F) 0.5 hr		14.3		14.3		19.0
Same as above plus 190°C (374°F) 0.5 hr		0		0		1.6
Same as above plus 205°C (401°F) 0.5 hr		0		0		--
Same as above plus 220°C (428°F) 0.5 hr		0		0		--
Same as above plus 235°C (455°F) 0.5 hr		0		0		--
Same as above plus 235°C (455°F) 17 hr		0		0		0

[a] Exposure at temperature for 2 min and 160 psi.

[b] Prior treatment: RT→60°C (140°F), holds 5 min; 60°C (140°F)→100°C (212°F), hold 1 hr.

Figure 4. Appearance and percent flow of PISO2 adhesive tapes exposed to 316°C (600°F), 343°C (650°F), or 371°C (700°F) for 2 min with 160 psi pressure.

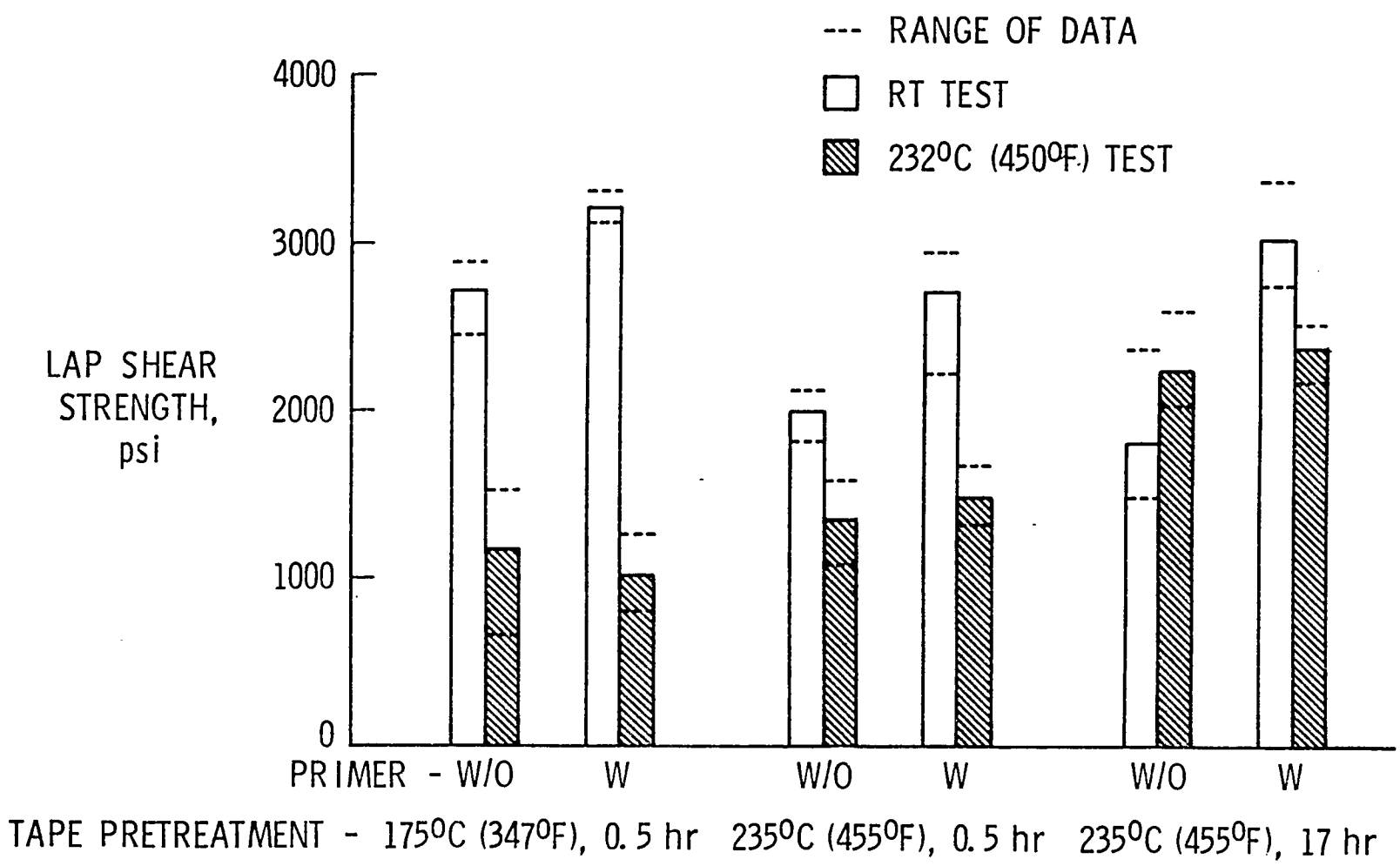


Figure 5. Effect of primer and adhesive tape pretreatment on bond strength for Cycle 1.

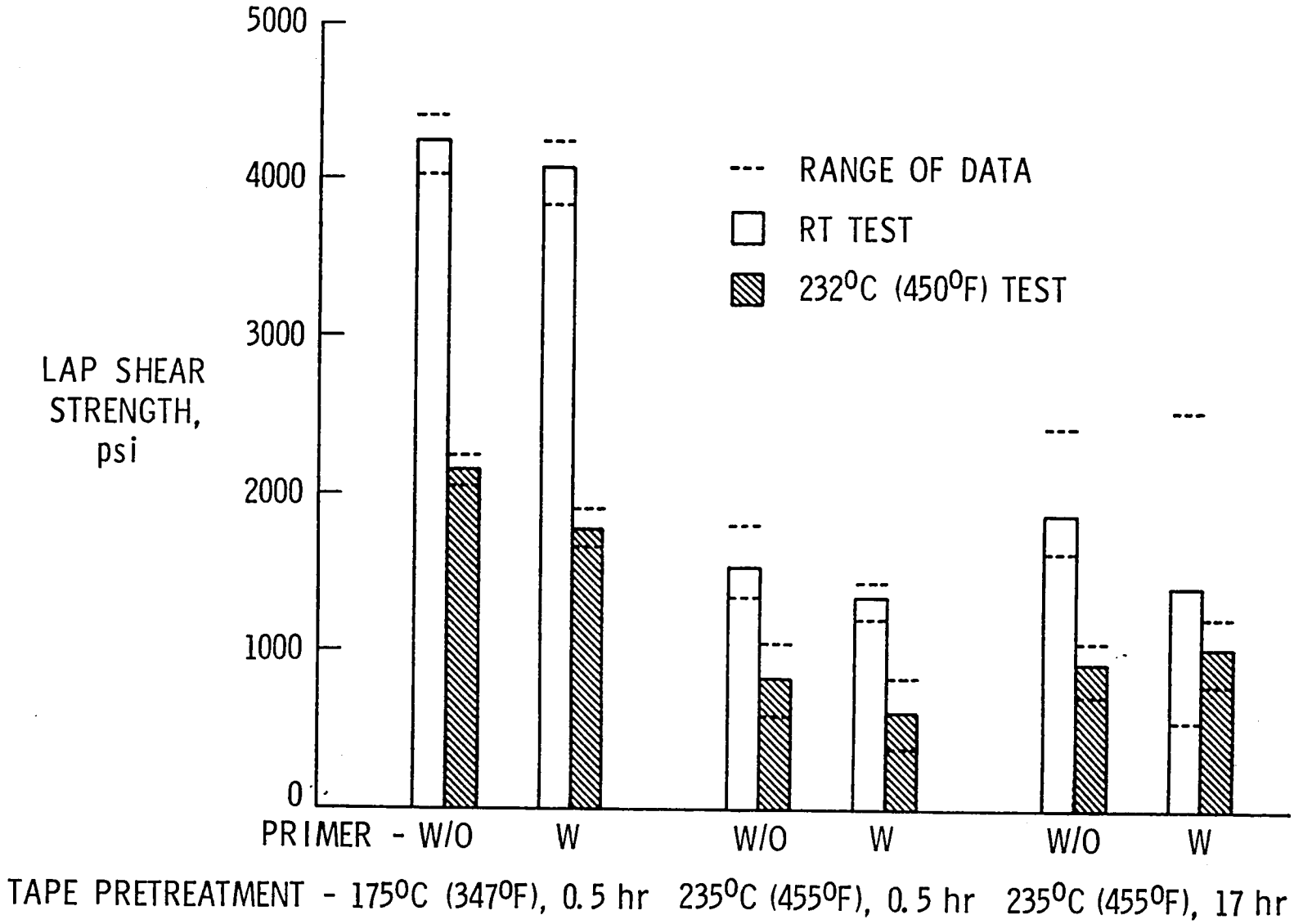


Figure 6. Effect of primer and adhesive tape pretreatment on bond strength for Preheated Press Cycle.

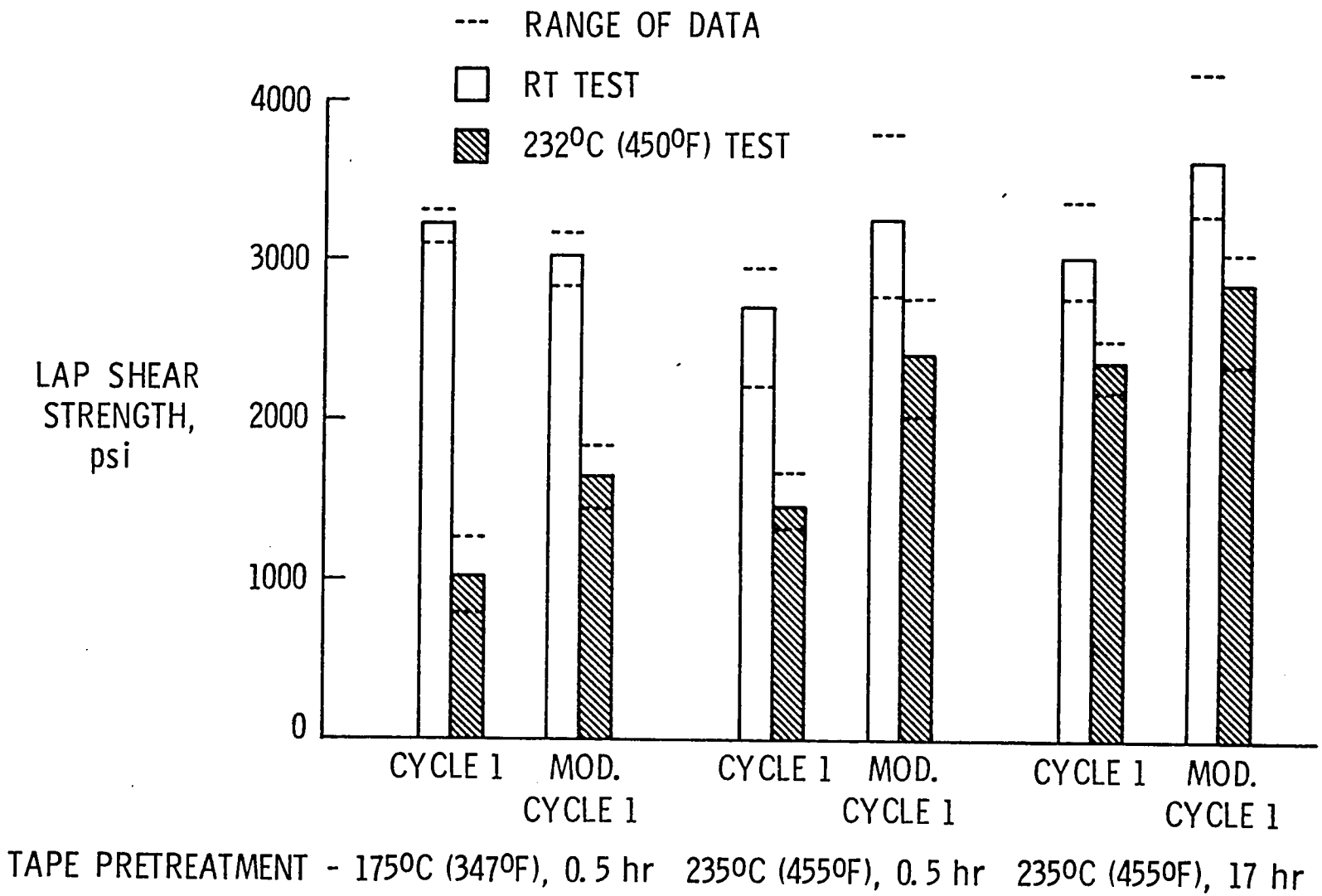


Figure 7. Comparison of lap shear strengths for Cycle 1 and Mod. Cycle 1.

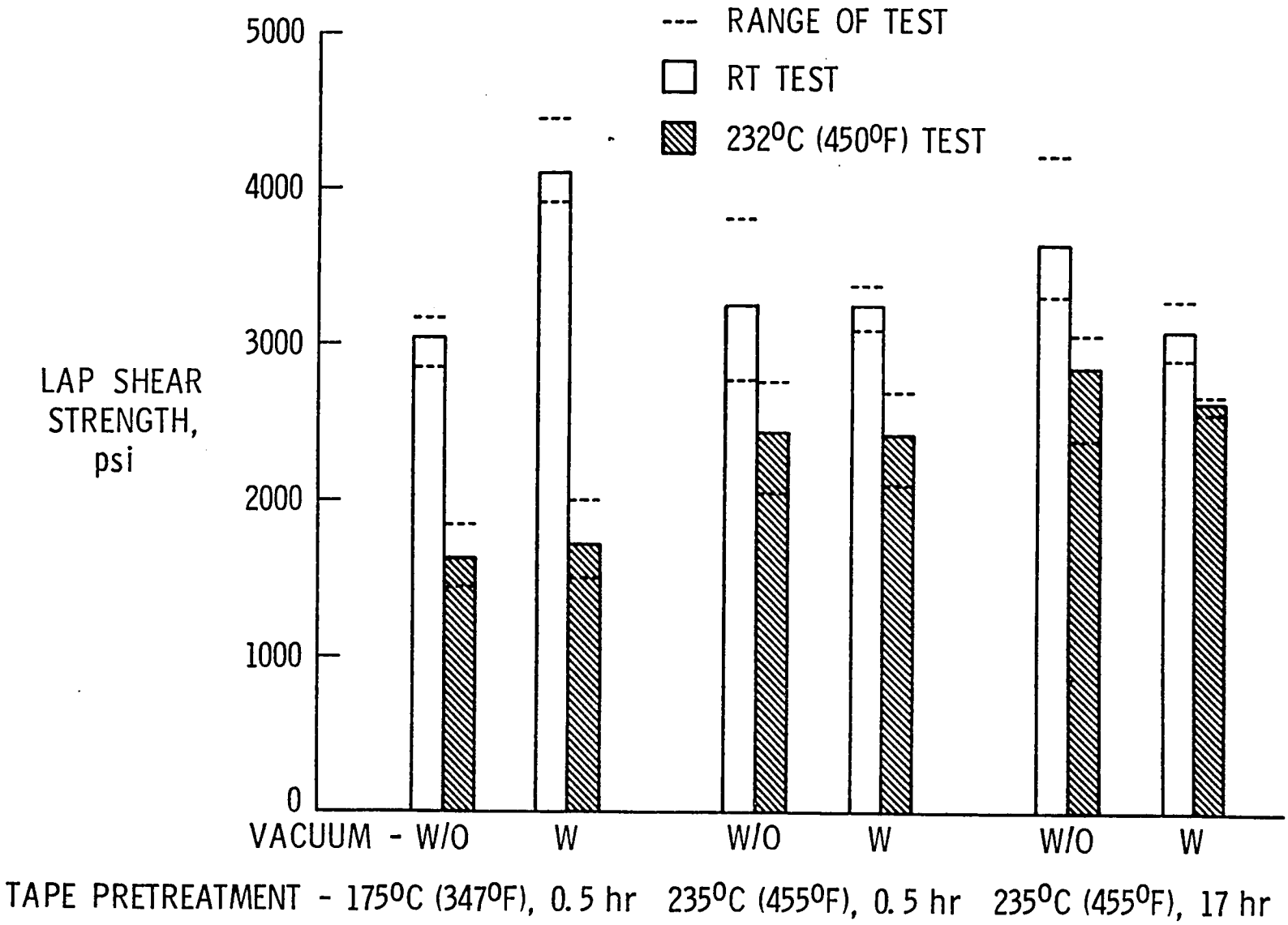


Figure 8. Effect of the use of vacuum during the bonding cycle.

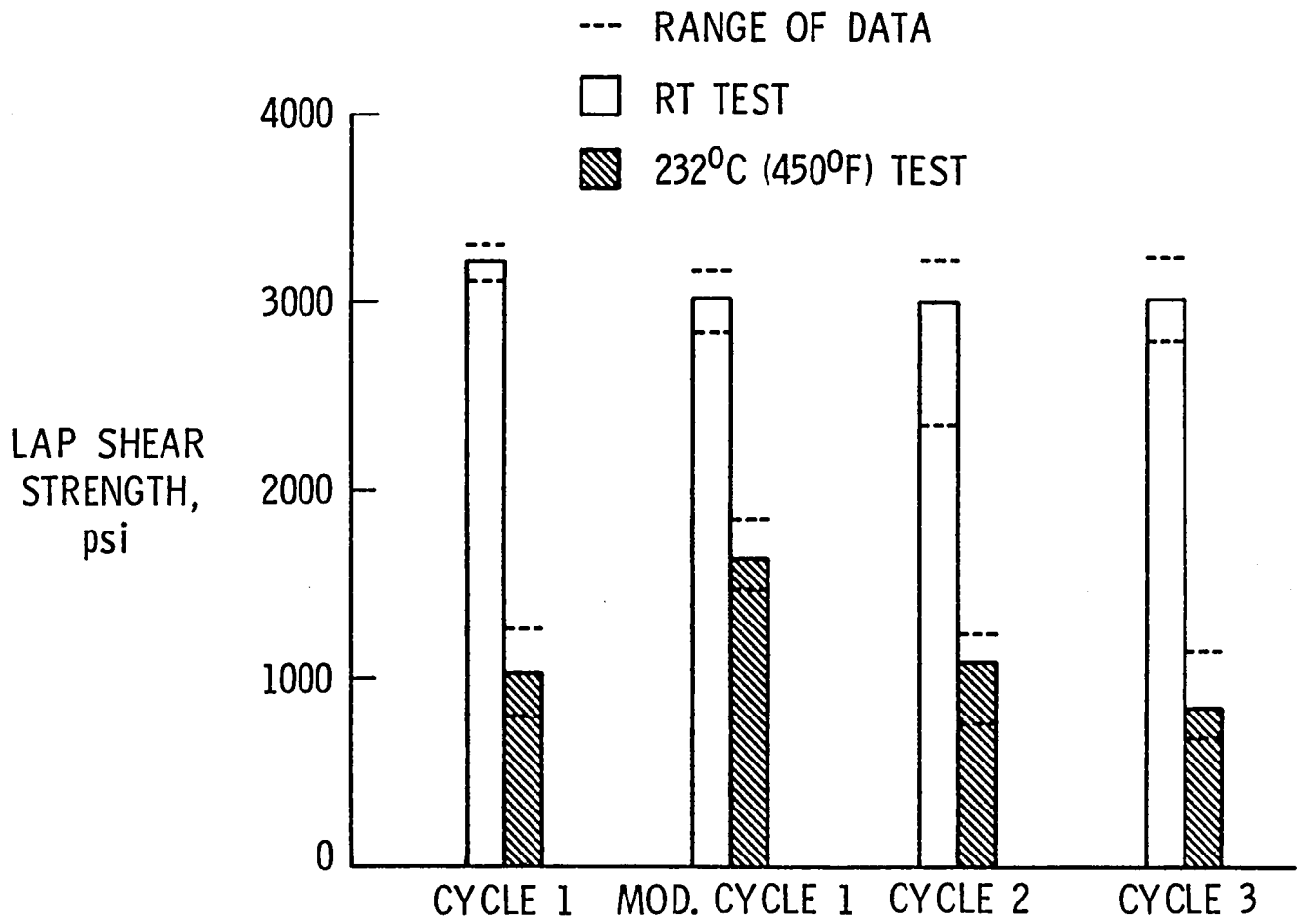


Figure 9. Lap shear strength results for PISO2 adhesive bonded titanium using four processing cycles [175°C (347°F), 0.5 hr TAPE].

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16. Abstract

An adhesive bonding process cycle study was performed for a polyimidesulfone (PISO2) recently developed at NASA-Langley Research Center as a promising structural adhesive. The high molecular weight, linear aromatic system possesses properties which make it attractive as a processable, low-cost material for elevated temperature applications. The report details the results of a study to better understand the parameters that affect the adhesive properties of the polymer for titanium alloy adherends. These include the tape preparation, the use of a primer, and press and simulated autoclave processing conditions. The polymer was characterized using Fourier Transform Infrared Spectroscopy, glass transition temperature determination, flow measurements, and weight loss measurements. The adhesive's strength determined by lap shear strength tests was used to evaluate the effects of the bonding process variations.

Higher lap shear strengths were obtained at room temperature when a primer (PISO2) was used; however, the results were the same at 232°C (450°F) with or without a primer. The use of a vacuum (simulated autoclave cycle) during titanium bonding produced similar lap shear strengths to those specimens bonded without a vacuum.

A bonding process cycle was developed which produces a good compromise of lap shear strength at room temperature (3650 psi) and at 232°C (450°F) (2870 psi). Further improvement could possibly be obtained by use of a high temperature postcure.

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