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Polyimide Matrix Composites: Polyhimidesulfone/LARC-TPI (1:1) Blend

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Abstract

Polyimide matrix composites were fabricated from unidirectional unsized AS-4 carbon fiber and a doped 1:1 blend of two polyimides: benzophenone dianhydride-3,3'-diaminodiphenylsulfone (PISO$_2$) and benzophenone dianhydride-3,3'-diaminobenzophenone (LaRC-TPI). To enhance melt flow properties, the molecular weight of the PISO$_2$ was controlled by end-capping with phthalic anhydride and addition of five percent by weight p-phenylenediamine-phthalic anhydride bisamic acid dopant. Prepreg was drum-wound using a diglyme slurry comprised of the soluble polyamideacid of PISO$_2$, the soluble bisamideacid of the dopant, and the insoluble imidized LARC-TPI powder. Melt flow studies with a rotary rheometer and parallel plate plastometer on neat resin and prepreg helped develop an optimum cure cycle. Composite mechanical properties at room and elevated temperatures; dry and moisture-saturated, were evaluated, including short beam shear strength and flexure, tensile, shear, and compression properties. Two 18" x 24" skin-stringer panels were fabricated, one of which was tested in compression to failure.

1. INTRODUCTION

Polymer research at NASA Langley Research Center has emphasized the development of high temperature thermoplastics such as polyimides and polyarylene ethers. They offer attractive mechanical properties at elevated temperatures for adhesive and composite matrix applications involving high speed aircraft. The purpose of this paper is to continue the evaluation study reported previously[1] on polyimide blends as matrices for carbon fiber reinforced composites.

In that study, a large number of compositions were investigated involving LARC-TPI, 1, polyimidesulfone. (PISO$_2$), 2, Figure 1, and blends of LaRC-TPI preimidized powder in polyamideacid diglyme slurries of 1 and 2. The study revealed that the slurry technique provided an excellent method for fabricating prepreg from otherwise insoluble polymer powders and afforded well-consolidated void-free composites with outstanding properties. The use of polyimide blends and bisamideacid additives to improve processability was also demonstrated.

Further research has shown that slurry blends with LaRC-TPI preimidized powder in PISO$_2$ polyamideacid solution demonstrated better melt flow characteristics than blends of the same powder in LaRC-TPI polyamideacid solution.[2] Therefore, this paper will deal only with the 1:1 slurry composition of PISO$_2$:LaRC-TPI preimidized powder in diglyme. This ratio proved to be the best compromise between melt flow, derived primarily from the preimidized LARC-TPI powder, and the ability to prepreg thick

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slurries.

2. EXPERIMENTAL*

2.1 Starting Materials

The PISO$_2$ polyamideacid solution, endcapped with phthalic anhydride, was obtained commercially from High Tech Services as a 30 percent solution in diglyme, inherent viscosity (0.5 percent in dimethylacetamide at 30°C) 0.57 dl/g. Its synthesis has been reported elsewhere.[1,3,4] The LARC-TPI 2000 polyimide powder was obtained in several lots from MTC, Inc. According to MTC assay, depending on the lots, imide content varied from 85 to 94 percent, polyamic acid from 5 to 15 percent, and isoimide from traces to 4 percent. The bisamideacid used to dope the slurries was prepared from one mole of p-phenylenediamine and two moles of phthalic anhydride as described elsewhere. (5)

Stable slurries were formulated by diluting the 30 percent PISO$_2$ solution to 15 percent and adding LARC-TPI powder to achieve a 1:1 ratio by weight of polymer solids. Concentrations ranging from 24-26 percent solids afforded slurries that were stable for several hours at room temperature. The bisamideacid dopant was added as a diglyme solution at a w/w concentration of 2.5 percent.

2.2 Prepreg/Composite Fabrication

Slurries were prepregged by passing a single tow through a dip tank and onto an 18-inch diameter multiple speed drum winder as described previously.[1] Fiber areal weights could be varied from 145 to 200 g/m^2 and resin contents from 38 to 50 percent, w/w. Unsized Hercules AS-4 12K tow carbon fiber was used exclusively in this study. Prepreg volatile content ranged from 5 to 15 percent before oven drying. Because the prepreg dried so easily at room temperature, it was immediately removed from the drum and cut as required while still tacky and drapeable. Oversize plies were further dried in a circulating air oven under restraint at 204°C and stored at room temperature. SEM photomicrographs

* Use of trade names or manufacturers does not constitute an official endorsement, either expressed or implied, by the National Aeronautics and Space Administration.

showed the 2-6 micrometer particles of LARC-TPI powder to be rather evenly distributed through the thickness of the tow and along the fiber axis.

The dried plies were trimmed to size and stacked in a closed mold between Kapton^R film spray-coated with Frekote^R 33. No bleeder cloths were employed since polymer flow was minimal. Moldings were done in a 25 ton four post upacting press containing 12 inch electrically heated platens. The press was outfitted with a linear voltage differential transformer (L.V.D.T.), cantilevered over the side, to measure platen movement during ply consolidation. Heating and cooling rates of about 7°C per minute were employed. From the L.V.D.T. response, the following cure cycle was developed and used exclusively: at 300 psi, ramp to the following temperatures, 260°, 276°, 300°, and 343°C, and hold at each temperature until flow as detected by the L.V.D.T. stops. Hold times varied with part thickness. However, the hold at 343°C was always one hour.

Panels were evaluated ultrasonically as previously described[1] at a preestablished sensitivity level adequate to detect microvoids in T300/5208 carbon fiber/epoxy composites. Composite fiber weight percentages and fiber volumes were calculated from experimentally determined values of fiber areal weight, panel dimensions and weight, and fiber density.

2.3 Characterization Equipment

DSC, TMA, mechanical test machine, and DCB test procedures and equipment were described previously.[1] Flexure tests were run at a 32:1 span:depth ratio. Scanning electron micrographs were taken with an Hitachi Model S-510 SEM. Procedures for longitudinal and transverse tension and iosipescu shear tests on unidirectional specimens have been reported.[6] Compression tests were made on O$_{48}$, 1.5 in. x 1.75 in. fully instrumented specimens as described by Shuart.[7] Neat resin GIC values were obtained on miniturized specimens using a procedure reported by Hinkley.[8]

The Rheometrics System 4 rheometer was used to measure isothermally the melt viscosity of LARC-TPI powders . The standard torsional mode at an oscillatory frequency of 10 rad/sec was employed after

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the sample pellet had melted, wetted the platens, and reached temperature. Similar measurements on LARC-TPI powders have been reported. [9] Melt flow properties were also studied with a Tinius Olsen parallel plate plastometer modified as described elsewhere.[10] All measurements were taken at 300 psi using a 3°C heating rate, the same conditions employed for composite fabrication. L.V.D.T. displacements were corrected for apparatus thermal expansion. Densified specimens used in the study were press molded at RT from powders which were either obtained commecially or precipitated from solution, washed and dried.

3. RESULTS AND DISCUSSION

3.1 Characterization of LARC-TPI 2000 Powder

Melt viscosities of the various lots of LARC-TPI 2000 powder were difficult to measure because, when heated, the reactive powders gave off volatiles and increased in viscosity. Volatiles apparently come from cyclodehydration of polyamideacid and reactive end group chain propagations. The optimum processing window for composite fabrication appears to be during the early stages of the melt before viscosity buildup reaches 10^5-10^7 poise and the volatiles cannot be removed. In later lots of the 2000 powder, lots 25-801 and 25-802, the melt viscosity during an isothermal measurement at 347°C increased from 200 poise to a maximum 40,000 poise over 40-50 minutes. These lots were the most extensively used in this study. Melt viscosity studies of some previous lots have been reported.[9]

A series of DSC curves of lot 25-801 powder is shown in Figure 2. The undried as-received material displayed a very strong crystalline melt endotherm centered at 282°C. On reheating, the Tm is not seen; only a Tg at 219°C is observed. Apparently, after a 350°C exposure the material remains amorphous. The composite cure cycle was adjusted upward to take advantage of this fact. DSC studies on similar LARC-TPI powders have been reported.[9]

Moldings were made by first preheating the powder in an open die in an oven at 300°/0.5 hour and 350°C/1 hour to remove all volatiles. The die was then transferred to a 350°C preheated press, the die closed, and 300 psi applied for 1 hour at 350°C followed by rapid cooling under pressure. X-Ray diffraction patterns of the moldings showed only the standard halo typical of amorphous materials. Void-free plates up to 6 in. x 6 in. x 0.3 in. were made by this procedure.

Table I lists tensile properties, fracture toughness, and Tg values for neat resin plates made from powder lots 92-712 and 25-801 and from consolidated commercial LARC-TPI film. Tensile properties from the two sources of LARC-TPI were in excellent agreement. Moduli were higher than those reported by Bell and coworkers[11] for 300°C air-cured LARC-TPI film (540 Ksi), an indication that some chemical/physical interaction may be occurring in the powder at the higher cure temperature. The Tg value of the molding was lower than that obtained from film,[11] probably because the molecular weight was somewhat lower. GIc values were extremely high (15.9 in-lb/in^2), reflecting the toughness observed in many similar thermoplastic materials.[12]

3.2 Characterization of Blended Powder

DSC curves of the blended powder from the doped polyimide formulation are shown in Figure 3. The powder was predried at 204°C to remove solvent and help imidize polyamideacid. The DSC of the predried powder displayed two very strong endotherms centered at 225° and 284°C. The former is attributed to the Tg of partially imidized PISO$_2$ and to imidization reactions,[3,10] the latter to the Tm of the LARC-TPI. When reheated, the powder afforded a DSC featured only by a broad complex endotherm from 225° to 260°C, assigned to overlapping Tgs of the PISO$_2$ and LARC-TPI polyimide structures. As expected, no evidence of a Tm was seen.

3.3 Parallel Plate Plastometer Studies on Neat Resins

3.3.1 Background

Repeated attempts were made to mold void-free PISO2:LARC-TPI composites at 300 psi utilizing flow information from the L.V.D.T. attachment to the press. Panels of highest consolidation were obtained when isothermal holds were placed at critical "flow transitions" during heat-up. These holds were maintained until platen movement subsided. The complex

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ramp/hold cure cycle described in Section 2.2 resulted. Such a procedure has been extremely helpful in determining molding cycles of low flow, difficult-to-process matrix materials.

However, to screen the flow properties of new materials and develop preliminary consolidation cycles under rheological conditions simulating press/autoclave conditions, a miniturized test was needed. Parallel plate plastometer procedures were developed to conduct preliminary flow/consolidation studies on small neat powder and prepreg samples at various heating rates and pressures. The technique helped correlate the melt flow behavior of neat powders of LARC-TPI, PISO$_2$, and their blends and prepregs with the successful complex ramp/hold cure cycle used to fabricate PISO$_2$:LARC-TPI (1:1) composites. This information was then used to optimize that cycle.

3.3.2 Melt Flow Studies

Plastographs of LARC-TPI (lot 25-801) and PISO$_2$ powders are shown in Figures 4 and 5. The LARC-TPI curves are characterized by a rapid rate of platen closure at 250°C, just below Tm. Total squeeze out of resin is observed. The PISO$_2$ powder dried at 204°C is characterized by rapid platen movement in the 225°-260°C range near Tg. Total loss of resin is observed. For the powder predried at 300°C, rapid platen movement occurs at 250°-260°C but does not afford total squeeze out. The curve levels out at 32 percent of the original gap indicating that some chemical/physical reaction which increases resin stiffness must occur when PISO$_2$ is exposed at 300°C in air for an hour. Obviously, predrying to 300°C is not desired for either material. Further, the data indicates that holds should be built into the molding cycle to take advantage of the rapid flow exhibited by each material at specific temperatures.

This data was utilized to interpret the plastographs in Figure 6 of the doped polyimide blend. The ramp/hold cycle shown in Figure 6 is the one optimized for composite fabrication, except the duration of each isothermal hold was reduced for experimental convenience. No flow is seen at 204°C, the predry condition. Flow begins at about 225°C, coincident with the Tg of partially imidized PISO$_2$. Although LARC-TPI has a Tg in this region, the effect on

flow would be insignificant due to its semi-crystalline nature. Flow associated with the Tg of imidized PISO$_2$ is expended during the hold at 260°C. A temperature rise to 276°C initiates melting of LARC-TPI crystallites and the hold there expends the majority of the flow produced by Tm. Above 276°C, some flow is incurred from additional melting and thermoplastic flow under 300 psi pressure. The holds at 300° and 343°C, while producing no further flow, prevent recrystallization during cool-down and probably initiate some chain extension and chemical/physical interactions which increase stiffness.

During the entire cycle, polyamideacid is converting to polyimide and chain extension is taking place, both of which occur with concomitant evolution of water of cyclodehydration. The latter, if not properly removed, can be the source of microvoids in the finished part. The holds permit dramatic drops in viscosity which, in turn, allow volatiles to be evolved at a controlled rate and consolidation to occur before chemical/physical reactions build the viscosity to undesired levels.

3.4 Composite Studies

3.4.1 Prepreg Melt Flow Study

Figure 7 shows a series of 5 plastographs, each made with a billet of stacked (0/90/0/90/0) prepreg, AS-4/PISO$_2$:LARC-TPI (1:1)/2.5% dopant, each billet predried at a different temperature for one hour. Runs were made with the optimized ramp/hold cure cycle. The initial platen movement in the 3 prepregs dried below 204°C was attributed to loss of solvent and imidization. In the temperature range from 220° to 260°C, platen movement was similar in all plastographs to that observed for the neat resin blend in Figure 6, except the rate of movement increased as the drying temperature was increased. Platen movement at Tm was relatively small compared to that of the neat resin in Figure 6. Apparently, fiber loading increases with decreased platen separation until most of the applied load is carried by the fibers and no further movement is observed. This study clearly demonstrates that the prepreg must be predried to 204°C to remove volatiles and prevent premature flow associated with imidization. Such flow at an early stage in the cycle is undesirable; it prematurely loads the fibers such that flow due to Tg and

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Tm cannot be fully exploited. The latter is critical to help remove air and water of cyclodehydration and wet the fibers.

3.4.2 Additional Composite Cure Studies

Figure 8 contains C-scans of four 3 in. x 3 in. AS-4/PISO$_2$:LARC-TPI(1:1) O$_{20}$ panels. All scans were run at the same signal amplitude and gain. The panel in scan 8a contained 2.5 percent dopant and was processed by the optimized ramp/hold cure cycle. The scan is virtually transparent except for a few voids at the edge and 3 triangularly positioned black spots caused by bolts supporting the panel in the water bath. The panel in scan 8b contained the same polyimide-doped matrix but was processed by a straight-up cure cycle using a 5°C/min. heating rate. The poor quality of the panel shows that this cure condition is unsatisfactory.

The scan in Figure 8c was made from a panel containing the undoped polyimide blend and processed by the optimized ramp/hold cure cycle. The poor quality of the laminate demonstrates the effect a 2.5 percent (w/w) addition of bisamideacid dopant has on polyimide resin melt flow properties[5] and ability to consolidate plies and fabricate void-free parts. The panel in scan 8d contained the same undoped polyimide and was processed by a straight-up cure cycle. As expected, the panel was of poor quality. Additional studies with 3 in. x 3 in. O$_{20}$ laminates where the optimum holds were either eliminated or altered generally afforded panels whose C-scans indicated unacceptable levels of voids. The effect of these on mechanical properties, especially at elevated temperature, will be determined.

3.4.3 Composite Properties

A description of each of the composites whose mechanical and toughness properties are reported herein is given in Table II. Short beam shear strengths and flexure properties at 4 temperatures, dry and moisture saturated, are listed in Table III. Tensile and shear properties at two temperatures, dry and moisture saturated, are given in Table IV and compression properties at room temperature dry are given in Table V.

The flexure properties were generally higher than those observed for undoped AS-4/PISO$_2$:LARC-TPI (2:1) composites in the earlier study.[1] Retention of both dry and wet strength at elevated temperatures was outstanding. About 80 percent retention of room temperature dry flexure strength was observed at 177°C/wet. Short beam shear strengths were relatively low compared to those for epoxy composites and for undoped polyimides in the earlier study.[1] This is in contrast to the iosipescu shear strengths, Table IV, which were outstanding. Both shear strength and modulus values at the 3 conditions shown in Table IV were higher than those of many selected thermoplastic and thermoset composites, including APC-2, 8551-7, and 2220-1 and -3.[6]

Longitudinal tensile values were standard for AS-4 fiber-reinforced laminates. Transverse properties were reasonably low, probably because of relatively poor fiber/resin interfacial adhesion. Compression tests were run at room temperature on thick specimens using a newly developed technique, so very little comparative data is available except that shown in Table V.[7]

The compression strengths for epoxy appear to be very low when compared to values obtained from other widely-used compression test methods. However, they appear to be reasonable for a test where interlaminar shearing is the dominate failure mode for unidirectional specimens. Both the epoxy and the polyimide specimens failed by this mechanism. Interestingly, the 118 Ksi value for the polyimide is approximately 87 percent of the epoxy value, whereas in other compression tests, thermoplastic compression strengths are only about 70 percent of those observed for thermosets.[12] Considering the quality of the drum-wound prepreg (e.g., fiber alignment, resin distribution, etc.), the polyimide value is very good. It is also notable that compression strength drops about 10 percent (118 to 106 Ksi) when specimens contain microvoids.

The mode I fracture toughness of a O$_{24}$ composite was measured using the double cantilever beam test. Values were 6.2 in-lb/in^2(initiation) and 8.8 in-lb/in^2 (propagation). The initiation value is close to that predicted from the relationship of neat resin Glc versus composite Glc.[13]

3.4.4 Skin-Stringer Panel

Two 18 in. x 24 in. T-bar structural test panels were fabricated according to the schematic in Figure 9. Conventional drum-

wound AS-4/PISO$_2$:LARC-TPI (1:1)/2.5% doped prepreg was used having a fiber areal weight of about 135 g/m^2, a thickness of 6 mils, and solvent (diglyme) content of approximately 5-15 percent to maintain tack and drape. Steel tooling was sprayed with a fluorocarbon release agent and baked at 232°C/1 hour. Prepreg was applied in a (0/45) layup to tooling bars arranged in opposed "C-channels" comprising the web and flanges of the stiffener. Additional prepreg in a (0/90/45) layup was applied to the cap of the stiffener and the stiffener element was placed on the preplied skin, a (90/45/0) layup. The entire assembly was placed on a steel caul sheet, vacuum bagged with 3 mil Kapton^R film and high temperature strip sealant, and autoclaved at 300 psi according to the optimum ramp/hold cure cycle discussed above. Photos of the planform and edge of a trimmed panel are shown in Figures 10 and 11, respectively. One panel was instrumented for a compression test and failed at 30.7 Ksi compression load. An identical AS-4/3502 epoxy panel failed at 41.9 Ksi.

4. CONCLUDING REMARKS

Well-consolidated high quality unidirectional and angle-plied carbon fiber reinforced composites were fabricated from a 1:1 blend of LARC-TPI preimidized powder slurried in PISO$_2$ polyamideacid diglyme solution doped with 2.5 percent (w/w) bisamideacid of p-phenylenediamine/2 phthalic anhydride. A complex optimized cure cycle was developed for this blend using information from parallel plate plastometry, melt viscosity, and DSC studies. Considerable effort was made to perfect characterization tools and methodology to aid in the fabrication of low flow matrices. The flexural, shear, and tensile properties at room and elevated temperatures, dry and wet, demonstrate the potential of this blend as a matrix material. Further research is required to improve melt flow characteristics and reduce the thermally induced volatiles of these low flow, thermally stable systems.

5. ACKNOWLEDGMENTS

5. REFERENCES

1. N. J. Johnston and T. L. St. Clair, Intl. SAMPE Tech. Conf. Series, 18, 53 (1986); SAMPE J., 23(1), 12 (1987).
2. N. J. Johnston, unpublished results, NASA-Langley Research Center.
3. T. L. St. Clair and D. A. Yamaki in Polyimides, K. L. Mittal, Ed., Plenum Pub. Co., NY, 1984, Vol. 1, P. 99; NASA TM-84574, 1982.
4. J. F. Dezern amd P. R. Young. Int. J. Adhesion and Adhesives, 5(4), 1985, 183.
5. J. R. Pratt, T. L. St. Clair, H. D. Burks, and D. M. Stoakley, Sci. Adv. Matl. Proc. Eng. Series, 32, 1036 (1987).
6. S. L. Coguill and D. F. Adams, Final Technical Report for NASA Grant NAG1-277, Dec. 1988. U. of Wyoming Report UW-CMRG-R-88-114, Dec. 1988; NASA CR-xxxxx, February, 1989.
7. M. J. Shuart, AIAA Paper No. 88-2293, AIAA/ASME/ASCE/AHS 29th Structures, Structural Dynamics and Materials Conf., Williamsburg, VA, April 18-20, 1988.
8. J. A. Hinkley, J. Appl. Polym. Sci., 32, 5653 (1986).
9. H. D. Burks, T. H. Hou, and T. L St. Clair, SAMPE Quart., 18(1), 1 (1986).
10. T. H. Hou and T. L. St. Clair, SAE Technical Paper Series, SP-748, 31 (1988).
11. V. L. Bell, B. L. Stump, and H. Gager, J. Polym. Sci.:Polym. Chem., 14, 2275 (1976).
12. N. J. Johnston and P. M. Hergenrother, NASA TM-89104, Feb. 1987; Sci. Adv. Matl. Proc. Eng. Series, 32, 1400 (1987).
13. D. L. Hunston, R. J. Moulton, N. J. Johnston, and W. D. Bascom, ASTM STP-937, 1987; D. L. Hunston, Composites Tech. Rev., 6, 176 (1984).
14. J. N. Dickson, S. B. Biggers, and J. H. Starnes, Jr., Seventh DoD/NASA Conf. on Fibrous Composites in Structural Design, Denver. CO, June 17-20, 1985.
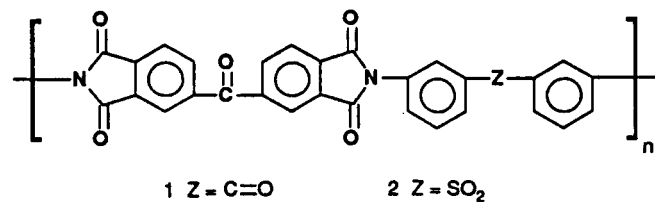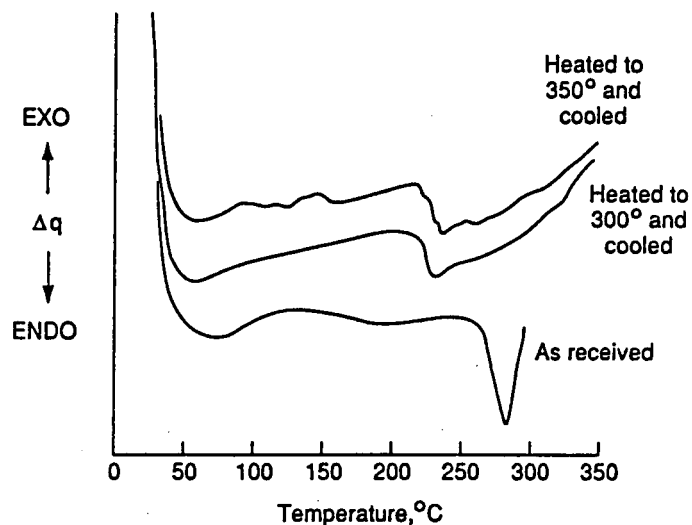
FIGURE 1. MOLECULAR STRUCTURE OF LARC-TPI, 1, AND PISO$_2$, 2.

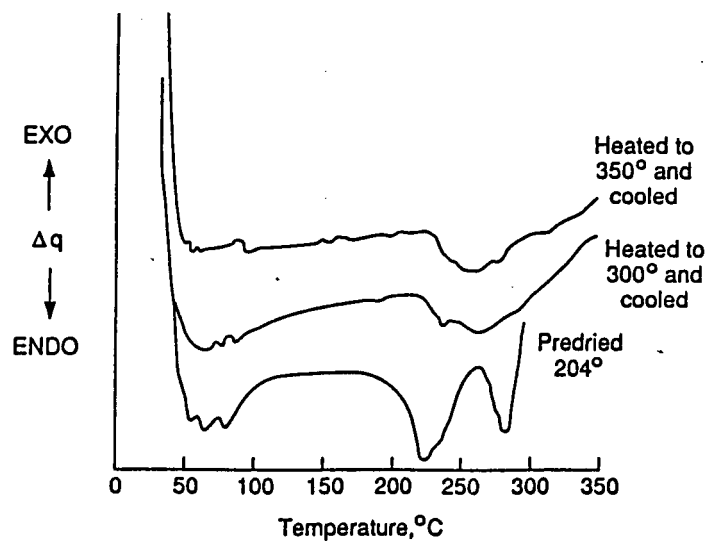FIGURE 2. DSC OF LARC-TPI PREIMIDIZED POWDER (MTC LOT 25-801)

FIGURE 3. DSC OF POWDER PRECIPITATED FROM DIGLYME BLEND OF
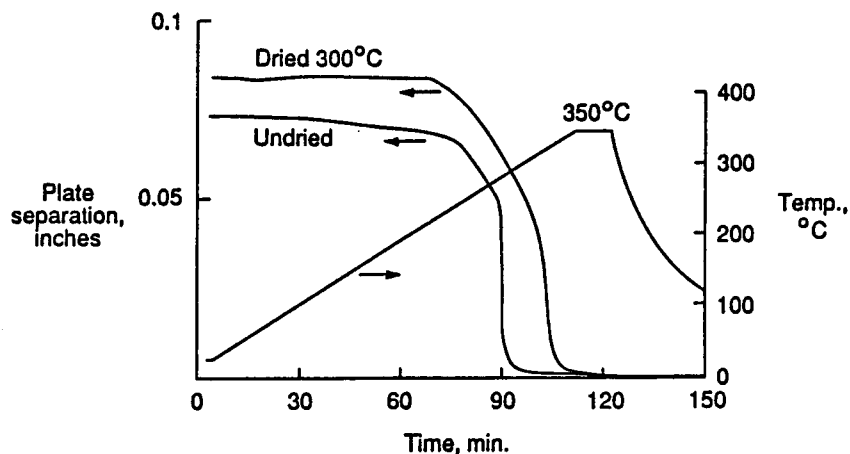PISO$_2$:LARC-TPI(1:1)/2.5% DOPANT

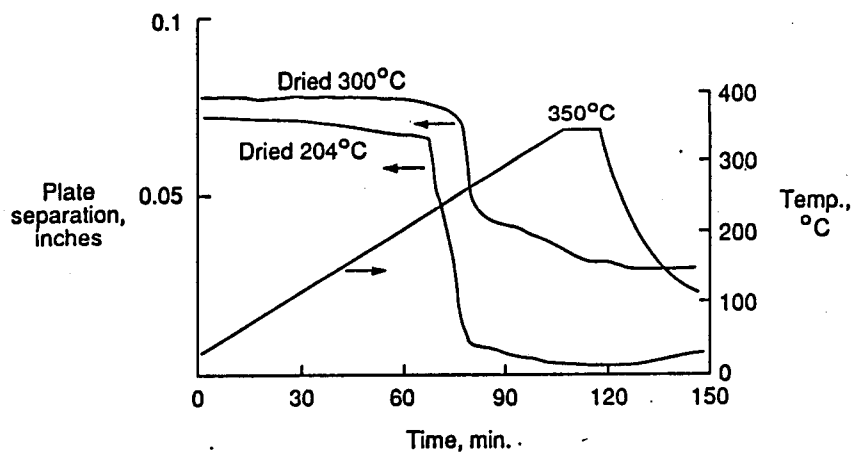FIGURE 4. PLASTOGRAPHS OF LARC-TPI PREIMIDIZED POWDER (MTC
LOT 25-801).

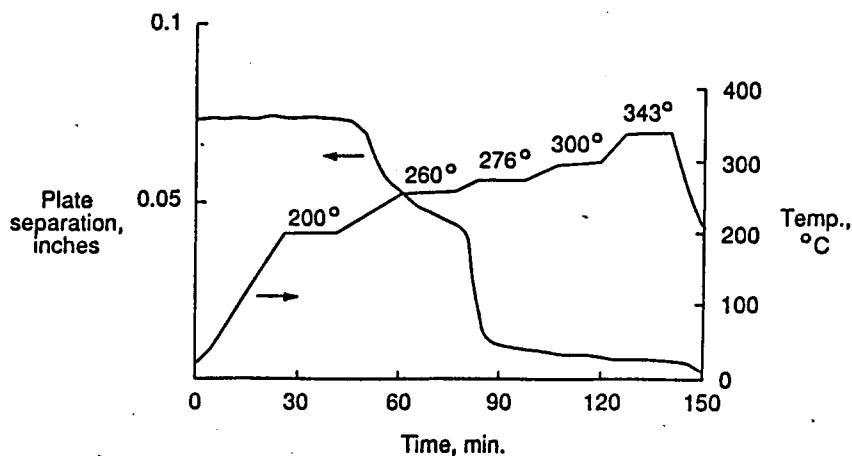FIGURE 5. PLASTOGRAPHS OF PISO$_2$ POWDER PRECIPITATED FROM
DIGLYME SOLUTION.

FIGURE 6. PLASTOGRAPH OF POWDER PRECIPITATED FROM DIGLYME
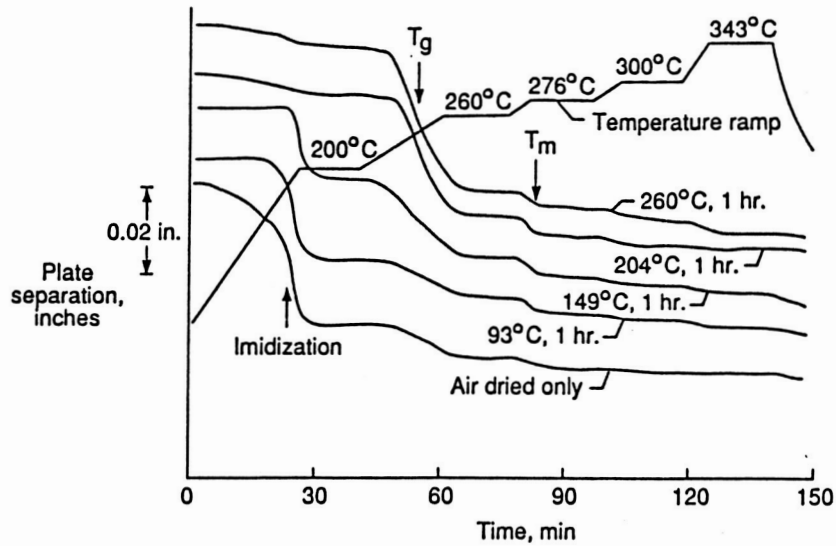BLEND OF PISO$_2$:LARC-TPI(1:1)/2.5% DOPANT.

FIGURE 7. PLASTOGRAPHS OF PREPREG MADE WITH AS-4 FIBER AND
PISO$_2$:LARC-TPI(1:1)/2.5% DOPANT BLEND. CURVES
OFFSET ALONG THE ORDINATE FOR DISPLAY PURPOSES.

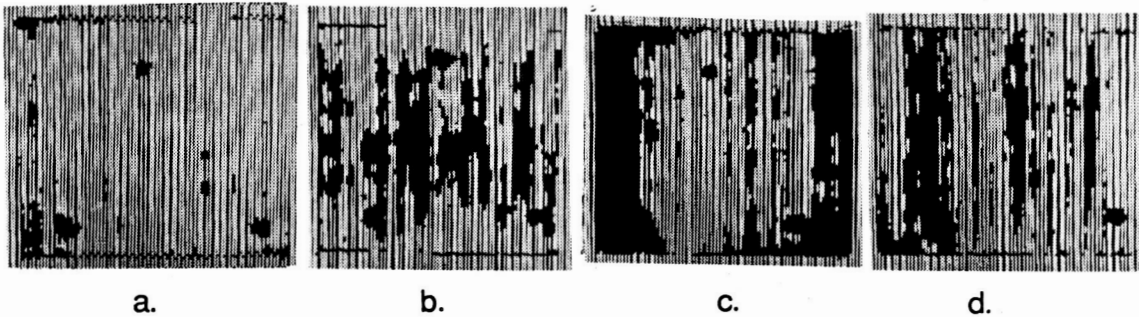a.   b.   c.   d.

FIGURE 8. C-SCANS OF 3"X3" $(0)_{20}$ AS-4/PISO$_2$:LARC-TPI(1:1)
COMPOSITES.
a. 2.5% DOPED FORMULATION, OPTIMIZED CURE CYCLE.
b. 2.5% DOPED FORMULATION, STRAIGHT-UP CURE CYCLE.
c. UNDOPED FORMULATION, OPTIMIZED CURE CYCLE.
d. UNDOPED FORMULATION, STRAIGHT-UP CURE CYCLE.

1.20″

1.25″

[±45/∓45/0₂/ ±45/0₆/90/0₂]ₛ

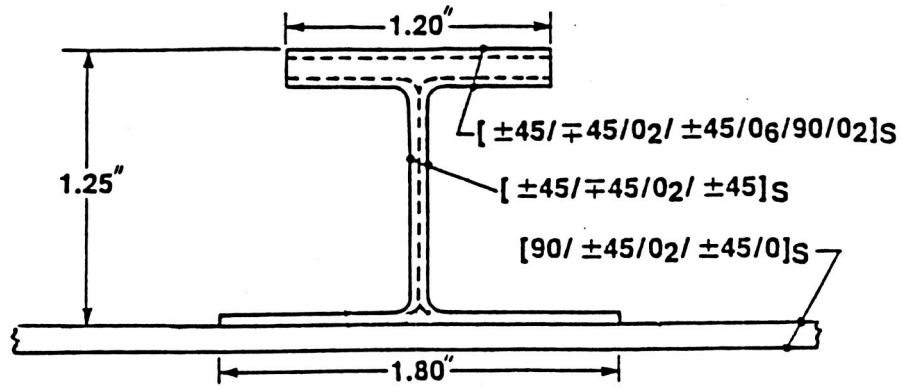[±45/∓45/0₂/ ±45]ₛ

[90/ ±45/0₂/ ±45/0]ₛ

1.80″

FIGURE 9.   END VIEW SCHEMATIC OF THE STRINGER PORTION OF THE 18"X24" POLYIMIDE SKIN-STRINGER PANEL.
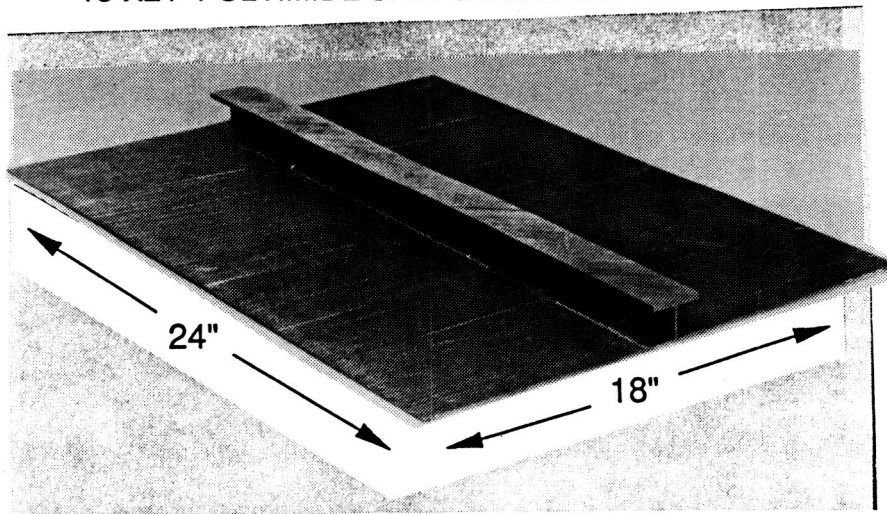

24″

18″

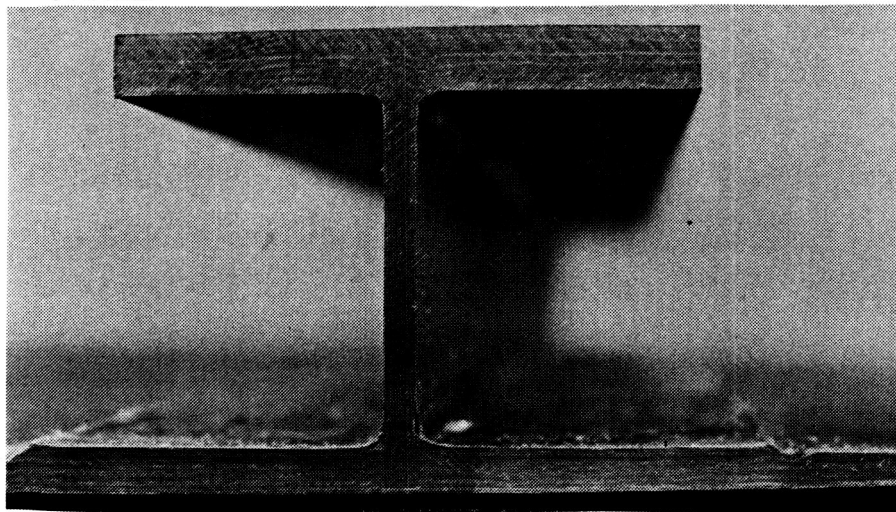FIGURE 10. PHOTOGRAPH OF THE18"X24" POLYIMIDE SKIN STRINGER-PANEL.

FIGURE 11. END VIEW PHOTOGRAPH OF THE STRINGER PORTION OF THE 18"X24" POLYIMIDE SKIN-STRINGER PANEL.

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Table I

Neat Resin Properties of Molded LARC-TPI[1]

Property	Molded Powder	Molded Film
Tensile Strength, Ksi	16.5[2]	16.9[5]
Tensile Modulus, Ksi	659	630
Tensile Strain-to-Failure, %	3.1	3.2
G_{Ic}, in-lb/in^2	15.9[3,4]	-
T_g, °C (DSC)	255[4]	-

[1] Molded tensile bars 4 in. long, gauge length 1 in.; average of 3 specimens; tensile data courtesy of J. T. Hartness, BASF Structural Materials, Inc.
[2] LARC-TPI powder lot 92-712
[3] Average of 4 specimens; Std. Dev. 3.3 in-lb/in^2
[4] LARC-TPI powder lot 25-801
[5] Film from MTC, Inc.

Table II

Description of AS-4/PISO$_2$:LARC-TPI (1:1)/2.5% Doped Composites[1]

Panel No.	Layup	Test	Thick., in.	Fiber Vol., %	Fiber Wt., %	T_g, °C dry/wet	C-Scan
GD-747	0_{10}	Flex.	0.059	57	65	247/231[2]	good
GD-793	0_{20}	SBS	0.113	52	60	237/220[2]	good
GD-837	0_{48}	Compr.	0.315	47	56	-	bad
GD-840	0_{48}	Compr.	0.267	51	59	-	fair
GD-841	0_{48}	Compr.	0.268	54	61	-	good
JS-325	0_{24}	DCB	0.182	58	66	-	good

[1] All panels were 6 in. x 6 in.
[2] Estimated from TMA, expansion mode

Table III

Short Beam Shear and Flexure Properties of
AS-4/PISO$_2$:LARC-TPI (1:1)/2.5% Doped Composites

Test Temp., °C	Specimen Condition	SBS[1] St., Ksi	Std. Dev., Ksi	Flex.[2] St., Ksi	Std. Dev., Ksi	Flex. Mod., Msi	Std. Dev., Msi	Defl., %
RT	dry	10.3	0.8	307	13	16.2	0.65	2.1
RT	wet	10.1	0.5	303	5	16.1	0.56	2.0
93	dry	10.3	0.9	283`	5	16.9	0.53	1.8
93	wet	8.6	1.1	261	9	15.3	0.21	1.7
150	dry	7.4	0.9	247	18	16.6	0.17	1.6
150	wet	6.9	0.5	256	5	16.7	0.81	1.6
177	dry	7.4	0.6	249	4	16.5	0.27	1.6
177	wet	5.4	0.3	244	14	16.4	0.54	1.5

[1] Panel GD-793
[2] Panel GD-747; moisture wt. gain 0.30%

Table IV

Tensile and Shear Properties for Dry Unidirectional
AS-4/PISO$_2$:LARC-TPI (1:1)/2.5% Doped Composites[1]

Test	Test Temp., °C	Specimen Condition	Strength, Ksi	Modulus, Msi	Strain-to-Failure, %
Axial tension[2]	23	dry	256	17.4	1.35
	100	dry	239	19.6	1.13
	100	wet	227	17.3	1.30
Transverse Tension[3]	23	dry	4.8	1.19	0.48
	100	dry	6.4	1.33	0.47
	100	wet	3.9	1.40	0.30
Iosipescu Shear	23	dry	19.1	0.95	>7
	100	dry	16.0	0.78	>14
	100	wet	14.7	0.82	>18

[1] Data from U. of Wyoming, Grant NAG1-277, Dr. D. F. Adams, PI
[2] Poisson's Ratio: 23°C dry, 0.31; 100°C dry 0.30; 100°C wet, 0.44
[3] CTE, 10^{-6}/°C: 23°C and 100°C dry, 22.3

Table V

Composite Compression Properties

Material	Strength, Ksi	Std. Dev., Ksi	Modulus, Msi	Std. Dev., Msi	Number Specimens	C-Scan
PI[1]	118	3	16.0	0.1	4	good
PI[1]	106	-	14.6	-	2	bad
Epoxy[2]	135	-	19.5	0	2	good

[1] AS-4/PISO$_2$:LARC-TPI(1:1)/2.5% Doped Composite
[2] AS-4/3502

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15. Supplementary Notes

16. Abstract

Polyimide matrix composites were fabricated from unidirectional unsized AS-4 carbon fiber and a doped 1:1 blend of two polyimides: benzophenone dianhydride-3,3'-diaminodiphenylsulfone ($PISO_2$) and benzophenone dianhydride-3,3'-diaminobenzophenone (LARC-TPI). To enhance melt flow properties, the molecular weight of the $PISO_2$ was controlled by end-capping with phthalic anhydride and addition of 5 percent by weight p-phenylenediamine-phthalic anhydride bisamic acid dopant. Prepreg was drum-wound using a diglyme slurry comprised of the soluble polyamideacid of $PISO_2$, the soluble bisamideacid of the dopant, and the insoluble imidized LARC-TPI powder. Melt flow studies with a rotary rheometer and parallel plate plastometer on neat resin and prepreg helped develop an optimum cure cycle. Composite mechanical properties at room and elevated temperatures, dry and moisture-saturated, were evaluated, including short beam shear strength and flexure, tensile, shear, and compression properties. Two 18" x 24" skin-stringer panels were fabricated, one of which was tested in compression to failure.

17. Key Words (Suggested by Author(s))		18. Distribution Statement
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