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Processing and Properties of Fiber Reinforced Polymeric Matrix Composites: II. Processing Robustness of IM7/PETI Polyimide Composites

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STATEMENT OF WORK

PETI is a class of new polyimides developed in the Composites and Polymers Branch, NASA Langley Research Center. The objective of this work was to investigate processing robustness of IM7/PETI composites. The following tasks were included in this investigation:

i) Determining the effect of B-staging conditions on the processability of IM7/PETI composite.

ii) Determining shelf life of IM7/PETI prepregs.

iii) Optimizing the molding conditions for IM7/PETI composite.

Previously documented report in this series includes:

- Processing and Properties of Fiber Reinforced Polymeric Matrix Composites: I. IM7/LARC™-PETI-7 Polyimide Composite, NASA CR-198254, December, 1995

PROCESSING ROBUSTNESS OF IM7/PETI POLYIMIDE COMPOSITES

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ABSTRACT

The processability of a phenylethynyl terminated imide (PETI) resin matrix composite was investigated. Unidirectional prepregs were made by coating an N-methylpyrrolidone solution of the amide acid oligomer onto unsized IM7. Two batches of prepregs were used: one was made by NASA in-house, and the other was from an industrial source. The composite processing robustness was investigated with respect to the effect of B-staging conditions, the prepreg shelf life, and the optimal processing window. Rheological measurements indicated that PETI's processability was only slightly affected over a wide range of B-staging temperatures (from 250°C to 300°C). The open hole compression (OHC) strength values were statistically indistinguishable among specimens consolidated using various B-staging conditions. Prepreg rheology and OHC strengths were also found not to be affected by prolonged (i.e., up to 60 days) ambient storage. An optimal processing window was established using response surface methodology. It was found that IM7/PETI composite is more sensitive to the consolidation temperature than to the consolidation pressure. A good consolidation was achievable at 371°C/100 Psi, which yielded an OHC strength of 62 Ksi at room temperature. However, processability declined dramatically at temperatures below 350°C.

1. EXPERIMENTAL*

1.1 IM7/PETI Prepregs

The synthesis of PETI oligomer was reported in an earlier paper [1]. The selected stoichiometric imbalance was ~ 9%, which resulted in a theoretical number average molecular weight (M_n) of ~ 5,000 g/mol. Two batches of IM7/PETI prepreg were used in this study. The sources and characteristics of these prepregs are summarized in Table 1. Unless otherwise stated, results discussed were obtained from prepregs made at NASA LaRC.

Table 1. IM7/PETI prepregs

Source	Lot	Width, in	FAW, g/m^2	Volatile cont., % w/w	Resin cont., % w/w	Handlability
[a]NASA LaRC	TM - 103	10.5	135	15.5	39.1	Moderately tacky and draperable
[b]ICI Fiberite	31740Q	24	143	22.9	36	Very tacky and draperable

[a]NASA Langley Research Center, Hampton, Virginia

[b]ICI Fiberite, Tempe, Arizona

1.2 Characterization Methods

Differential scanning calorimetry (DSC) was performed on a Shimadzu DSC-50 calorimeter. The glass transition temperature (Tg) was taken at the inflection point in the heat flow vs. temperature curve. Thermogravimetric analysis was performed on a Seiko TG/DTA 220 at 2.5°C/min in flowing (40mL/min) air or nitrogen.

Rheological measurements were conducted on a Rheometrics System 4 rheometer. Both neat resin and composite were characterized. Neat resin measurements were performed using parallel-plate geometry, while torsional rectangular geometry was employed for composite prepreg measurements. For neat resin measurement, sample specimen disks, 2.54 cm in diameter and ~ 1.5 mm thick, were prepared by press molding PETI powder at RT. The compacted resin disk was then loaded between parallel plates in the rheometer. For composite measurement, four to six plies of IM7/PETI prepreg

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

measuring 2.5" x 0.5" were stacked and gripped on both ends by the rheometer fixture, affording an effective gauge length of approximately 1.5". Dynamic mode was used in both cases. During measurement, a controlled strain of 5% and angular frequency of 10 rad/sec were maintained for specimen deformation. The resultant torque was recorded by a transducer. These torque values were decomposed into in-phase and out-of-phase components from which material properties such as storage modulus (G'), loss modulus (G") and complex viscosity (η^*) were readily derived by the principle of linear viscoelasticity.

Ultrasonic C-scans were performed at pulse echo mode using a 5 MHz Paramatrics transducer with 0.5" diameter and 4" focal distance, and a Paramatrics 5055 pulse receiver. Gate was set over an auxiliary glass reflector. All measurements were performed at attenuation value 10 dB, step and index 0.040 in., pulse repetition rate 0.03 MHz , and receiver gain -20 dB.

Open hole compression (OHC) strength values using Northrop specification [2] were measured at room temperature (RT) and 177°C (wet). Laminate lay-up was [42/50/8] or $[\pm45/90/0/0/\pm45/0/0/\pm45/0]_S$. Values for resin and void contents were determined by either image analysis or acid digestion, in which 1:1 w/w concentrated sulfuric acid and 30 percent hydrogen peroxide were used. The calculations were based on a density of 1.77 g/cc for the IM7 fiber and 1.37 g/cc for the cured PETI resin.

2. RESULTS AND DISCUSSION

2.1 Effect of Prepreg B-stage Conditions

A previously designed [1] composite molding cycle, designated as "Standard Cycle", consistently yielded well consolidated, void-free IM7/PETI laminates under 371°C and 200 Psi. This cycle, shown in Figure 1, consisted of the following steps:

1) Apply vacuum. B-stage prepreg 250°C for 1 hr in press without pressure.
2) Apply 1.38 MPa (200 Psi) at the end of 250°C/1 hr hold while increasing temperature at ~3°C/min to 371°C and hold for 1 hr.
3) Cool in press. Release pressure and vacuum below 100°C.

In order to obtain a void-free composite, step 1) in this cycle was a necessity for the removal of volatiles, i.e., carrier solvent and by-product (water) generated by the imidization reaction, from the wet prepreg before consolidation. TGA results indicated that less than 0.2% residual volatiles remained in the prepreg after 1 hr at 250°C [1].

From a processing viewpoint, a reasonably wide B-stage window is desired for the PETI prepreg materials to accommodate processing variables (for example, variations of time-temperature-pressure profile over a large part) incurred during composite fabrication. Lowering the B-stage temperature (Figure 1) will likely enable prepregs to retain excessive residual volatiles in the consolidating laminate, resulting in a voidy part. On the other hand, raising the B-stage temperature will potentially trigger the crosslinking reactions of the 4-phenylethynylphthalic anhydride (PEPA) endcaps, which will inevitably decrease the fluidity of the (curing) matrix resin; consolidation may no longer be achievable under the moderate 200 Psi. Consequently, it is desired to explore the B-stage window for this composite.

Rheological measurements were conducted on neat PETI resin for three temperature profiles, as shown in Figures 2 and 3. These profiles simulated temperature cycles used in the molding of IM7/PETI composites. The lower temperature hold (i.e., B-stage) steps were chosen at 250°, 300° (in Figure 2) and 350°C (in Figure 3), respectively. A higher temperature hold (i.e., final consolidation) step at 371°C was used in all cases. η^*, tan δ and T versus time were plotted.

For the two lower B-staging temperature profiles, values of η^* increased during the B-stage hold steps, indicating a slight softening of the specimen, which still, however, remained semi-solid (i.e., tan $\delta < 1$). In this period, tan δ (t) was lower for the higher temperature B-staged specimen, indicating an increased degree of viscoelastic solid-like characteristics of the PETI resin during the 300°C/1 hr hold period. A sharp drop in η^* occurred at approximately 300°C during the second ramp to 371°C. After melting, η^* continued to decrease until a minimum was reached at 371°C. Minimum $\eta^* = 10^3$ Pa sec and 3 x 10^3 Pa sec were measured for the 250°C/1 hr and 300°C/1 hr B-staged specimens, respectively. The subsequent increases in viscosities were due to the crosslinking reactions of the reactive PEPA end groups. These reactions were also manifested by formation of a gelation point (tan δ = 1, Figure 2). The specimens were transformed quickly to viscoelastic solid-like form with decreasing tan δ (i.e., decreasing resin fluidity). The slightly higher minimum viscosity exhibited by the 300°C/1 hr B-staged specimen indicated that processability of the matrix resin was slightly compromised due to the crosslinking reactions.

For specimen after a 350°C/0.5 hr B-staging, however, a minimum $\eta^* = 5 \times 10^4$ Pa sec was measured at 371°C (Figure 3). At the minimum viscosity, tan δ was less than 1 which was opposite to that measured for 250°C and 300°C B-staged specimens (see Figure

2). This behavior suggests that the 350°C/1 hr B-staging condition was too severe for the PETI resin. At 350°C, an appreciable degree of crosslinking was triggered along with volatile depletion, resulting in a severe loss of processability for this composite. The gelation point was not found during 371°C/0.5 hr hold; moreover, η^* values did not decrease when temperatures were raised from 371°C to 390°C in Figure 3.

The rheological behavior discussed above indicates that processability of the PETI matrix resin was only slightly affected by changing B-stage temperature from 250°C to 300°C. Residual volatiles in the matrix resin should be << 0.2% after B-staging at 300°C/1 hr. PETI melt fluidity increases rapidly as the temperature approaches 371°C. Pressure for laminate consolidation should be applied shortly after the B-stage period to obtain maximum benefit of the melt fluidity.

OHC strengths were measured for IM7/PETI composite panels consolidated at these B-stage conditions. Final consolidation at 371°C/ 1 hr and 200 Psi was employed in all cases. The results were tabulated in Table 2.

Table 2. Effect of B-stage conditions on the OHC strength values of IM7/PETI composites

Spec. ID	B-stage	Consolidation		OHC Strength (Ksi)	
		Temperature	Pressure (Psi)	RT	177°C (wet)
1	250°C/ 0.5 hr	371°C/1 hr	200	57.8±2.8	39.7±3.7
2	250°C/ 1 hr	371°C/1 hr	200	59.9	44.2
3	300°C/ 0.5 hr	371°C/1 hr	200	55.0±0.7	44.3±3.3
4	300°C/ 1 hr	371°C/1 hr	200	54.5±2.0	42.1±0.2
5*	Ramp from RT to 371°C	371°C/1 hr	200	55.4±2.3	39.6±1.8

*In this case, pressure was applied at 300°C during the temperature ramp. In other cases, pressure was applied at the end of B-stage period.

These results confirmed the conclusions drawn from the rheological measurements discussed above. When the standard deviations of measurement were considered, both RT and 177°C OHC strength values were indistinguishable among specimens. Averaged strength values were 56.5 Ksi at RT and 42.0 Ksi at 177°C (wet). The fact that the same strength level was achievable in specimen 5 suggests that the rate of volatile depletion from

PETI matrix resin is more sensitive to the B-stage temperature than to the duration of B-stage period.

2.2 Shelf Life of IM7/PETI Prepregs

Prepregs with polymeric resin matrix require long shelf life to withstand the severe fabrication environment where, over a period of time, trimming, laying, bagging and curing into structural parts occur. To evaluate shelf life, IM7/PETI prepregs were sealed in polyethylene bags and stored at RT for 30 and 60 days, respectively. Residual volatiles and rheological properties of these ambient-stored prepregs were monitored. OHC strengths were also measured for the specimens made from these prepregs.

Rheological measurements shown in Figure 4 include those measured for the as-fabricated, 30 days and 60 days ambient-stored prepregs, as well as for neat PETI resin. All measurements were performed under the same temperature profile, i.e., the standard temperature cycle with 250°C/1 hr B-stage and 371°C/1 hr consolidation. Values of η^* as a function of time were recorded. Variations among the $\eta^*(t)$ curves for different prepregs were attributable to the difference in number of prepreg plies used in each measurement. Higher η^* values measured for the prepreg specimens, when compared with those measured for the neat resin, were attributed to the reinforcing fibers in the prepregs. The results shown in Figure 4 suggest that IM7/PETI prepreg rheology was not affected by the prolonged ambient storage. Critical prepreg characteristics such as the initial softening temperature of matrix resin (i.e., ~ 260°C) and the temperature of minimum viscosity (i.e.,

Table 3. Effect of ambient storage for IM7/PETI prepregs

Duration of Ambient Storage	*Residual Volatiles (% w/w)	OHC Strength (Ksi)	
		RT	177°C (wet)
As-fabricated	18.5	59.9	44.2
†30 days	18	65.5 ± 1.0	46.3 ± 1.3
60 days	17	57.3 ± 2.3	43.1 ± 2.4

*Residual volatiles were measured by TGA on resin flash from the aged prepregs. % w/w values did not include fiber weight.

†ICI Fiberite prepreg

371°C) were all retained by the RT-aged prepregs. The delay in the initial softening temperature (at ~ 300°C, Figure 4) observed for the neat resin was attributed to specimen size and fixture geometry used in the measurement.

OHC strengths for the specimens made from these ambient-stored prepregs are tabulated in Table 3. Among the specimens made from NASA LaRC prepregs, no statistically significant differences exist in strength values. Specimens made from ICI Fiberite prepregs consistently yielded higher strength values, as observed in other cases (see Table 7 below) as well. IM7/PETI prepreg was shown to possess long shelf life.

2.3 Molding Cycle Optimization

2.3.1 Statement of Problem

Refering to the "Standard Cycle" in Figure 1, the critical molding variables and their functionalities during IM7/PETI composite consolidation are summarized in Table 4.

Table 4. Critical molding variables and functionalties for IM7/PETI laminate fabrication

Molding Variables		Functionalities
1	Heating rate	• Higher heating rate leads to shorter molding cycle and more economic fabrication process, however, the processing window may be narrowed for a reactive resin matrix. This is particularly true during the second ramp to 371°C.
2	B-stage temperature	• A necessary step for volatile depletion before commencing consolidation. • Selected temperature should be high enough for imidization reaction, but low enough to avoid crosslinking reaction by the endgroups in the molecules.
3	B-stage duration	• Selected duration time should be long enough to adequately deplete volatiles.
4	Consolidation temperature	• Selected temperature must fully exploit fluidity afforded by the reactive resin matrix, while at the same time, conforming to the capacity of molding equipment. • For PETI matrix, this temperature must be higher than T_m (~ 350°C) of crystallinity generated during 250°C/1 hr B-stage period [1].
5	Duration of consolidation	• Selected duration time should be long enough to achieve full consolidation of the laminate.
6	Consolidation pressure	• Selected pressure should be adequate for resin impregnation within the consolidating laminate without causing excessive resin loss.
7	Pressure application point (PAP)	• In order to fabricate void-free, well consolidated laminate, the pressure application point should be selected based upon a balance between two opposing factors: residual volatile contents in the prepreg and remaining fluidity of the matrix resin. Achieving such a balance could be difficult when dealing with reactive resin systems [1, 3-5].

Because of processing constraints such as slow heating rates (typically 1 - 4°F/min) and restricted pressure and temperature, associated with large commercial autoclaves, the molding conditions were restricted to 200 Psi or less and 371°C (700°F) or below. These constraints essentially fixed molding variables 1. The effect of prepreg B-stage conditions (temperature - time profile) had been investigated in section 2.1 above. It was concluded that processability of the PETI matrix.resin was only slightly affected in a wide range of B-stage temperatures from 250°C to 300°C. Therefore, for the sake of simplicity in this optimization study, variables 2 and 3 in Table 4 were fixed at 250°C and 1 hr, respectively. The pressure application point (PAP) was also fixed at the end of the B-stage period. At this PAP, it had been established that residual volatiles in prepregs were < 0.2% and residual matrix fluidity was fully exploited by holding through the entire consolidation period [1]. This left only two variables, i.e., consolidation temperature (CT) and consolidation pressure (CP), in the molding cycle to be optimized.

2.3.2 Experimental Design

The experimental arrangement under discussion was based on fractional factorial design. The problem was approached from the standpoint of response surface methodology [6, 7]. A set of experiments was organized which explored the regions of interest of the CT and CP (independent) processing variables. The experimental results were analized using a regression equation to estimate laminate consolidation quality (a dependent variable) anywhere in the regions of interest. This equation was also used to identify optimal combinations of CT and CP in the molding cycle which should produce maximum quality of consolidation.

Thirteen experiments with coded molding variables were organized as shown in Table 5 according to the Box-Wilson Central Composite (BWCC) design. Regions of interest for the two processing variables were selected as 325°C ≤ CT ≤ 371°C and 100 Psi ≤ CP ≤ 200 Psi. The scaling factors were $CT_{scaling\ factor}$ = 16.26°C and $CP_{scaling\ factor}$ = 35.36 Psi. Values of CT and CP for each experiment were obtained by the following formula and included in Table 5:

$$CT = CT_{coded} \times CT_{scaling\ factor} + 348$$
$$CP = CP_{coded} \times CP_{scaling\ factor} + 150$$

A unidirectional 3" x 3" - $[0]_{24}$ composite panel was fabricated at each experimental condition. Consolidation quality of each panel was measured by ultrasonic C-scans. Percentage of the area of each panel which possessed greater than 90% threshold consolidation, as measured by the digitized C-scan images, was also tabulated in Table 5. The 90% threshold cut-off was chosen based on the previous experiences which correlated C-scan to the composite quality of consolidation.

Table 5. Box-Wilson Central Composite Design

Run	CT_{coded}	CP_{coded}	CT (°C)	CP (Psi)	C-Scan Quality (%)
1	-1	-1	332	115	0.05
2	1	-1	364	115	5.05
3	-1	1	332	185	35.54
4	1	1	364	185	96.26
5	-1.414	0	325	150	0.07
6	1.414	0	371	150	83.8
7	0	-1.414	348	100	2.1
8	0	1.414	348	200	81.22
9	0	0	348	150	50.44
10	0	0	348	150	53.93
11	0	0	348	150	45.46
12	0	0	348	150	71.04
13	0	0	348	150	55.05

2.3.3 Regressional Analysis

Results in Table 5 were used to construct a non-linear regression equation of following form:

$$CSQ = b_0 + b_1\, CT + b_2\, CP + b_{11}\, (CT)^2 + b_{22}\, (CP)^2 + b_{12}\, (CT)\, (CP) \qquad (1)$$

where CSQ denoted C-Scan Quality of the panels in %. Procedure for analysis and the obtained values of coefficients of Eq. (1) were tabulated in Table 6.

An estimate of variance, S = 9.62, was obtained. The significance of each term in the regression equation was also examined by the standard error and the t test. These values were also included in Table 6. The results indicated a significance with at least 95% confidence level for each regression coefficient. Eq. (2) represented the final regression equation:

$$CSQ = 55.18 + 23.01\ CT + 29.82\ CP - 8.52\ (CT)^2 - 8.66\ (CP)^2 + 13.93\ (CT)\ (CP) \quad (2)$$

Table 6. Regression Analysis for the Box-Wilson Central Composite Design

Expt. Run	C-Scan Quality (%)	CT	CP	$(CT)^2$	$(CP)^2$	(CP) (CT)
1	.05	-0.05	-0.05	0.05	0.05	0.05
2	5.05	5.05	-5.05	5.05	5.05	-5.05
3	35.54	-35.54	35.54	35.54	35.54	-35.54
4	96.26	96.26	96.26	96.26	96.26	96.26
5	.07	-0.10	0	.14	0	0
6	83.80	118.49	0	167.60	0	0
7	2.10	0	-2.97	0	4.20	0
8	81.22	0	114.85	0	162.44	0
9	50.44	0	0	0	0	0
10	53.93	0	0	0	0	0
11	45.46	0	0	0	0	0
12	71.04	0	0	0	0	0
13	55.05	0	0	0	0	0
Total	580.01	184.11	238.58	304.64	303.54	55.72

Regression Parameters	b_0	b_1	b_2	b_{11}	b_{22}	b_{12}
	55.18	23.01	29.82	-8.52	-8.66	13.93
Strd. Error	3.41	3.41	3.41	3.65	3.65	4.81
t test	16.18	6.75	8.75	2.33	2.37	2.90
% Confidence	99+	99+	99+	95+	95+	97.5+

2.3.4 Optimal Molding Conditions

A CSQ contour surface mapping based on CT and CP molding variables was generated by means of Eq. (2) and shown in Figure 5. Contour curves with equal CSQ values were labeled in the Figure. A processing window (defined by CT and CP) which would yield a CSQ > 90% threshold consolidation over the entire (i.e., 100% label in Figure 4) 3" x 3" - $[0]_{24}$ panel was clearly identified. This optimal window was bordered by CT = 371°C, CP = 200 Psi and the 100% contour curve in the Figure. At the maximum allowable molding pressure, CP = 200 Psi, the optimal molding window in Figure 4 indicated that a CSQ > 90% threshold consolidation over 100% panel area was not achievable at CT < 355°C. It was reported that double crystalline melting peaks at 290°C and 353°C were induced during the 250°C/1 hr annealing in PETI resin [1]. Composite processability is likely to suffer at CT < 355°C due to the reduced fluidity with a semi-crystalline matrix.

Also included in Figure 5 were discrete RT and 177°C (wet) OHC strength values measured at several molding conditions and tabulated in Table 7. A few observations are worth noting:

i) At the maximum allowable processing temperature, CT = 371°C, good panel consolidation was achieved at CP = 200, 160 and 100 psi. Good consolidation at 371°C/100 Psi was only achieved by ICI Fiberite prepregs (see Table 1). The laminate quality of consolidation was clearly affected by the prepreg quality.

ii) When the CTs were lowered 11-16°C, even while maintaining good CPs, the elevated temperature OHC data dropped appreciably, e.g., Panels #10, 12 and 14 in Table 7.

iii) When the CTs were lowered 30°C, the OHC strength values were significantly affected, e.g., Panels #21 and #22 in Table 7.

These results strongly suggest that IM7/PETI composite molding is more sensitive to the consolidation temperature than to the consolidation pressure. They also show that the response surface methodology employed for the molding cycle optimization is a useful tool. The regression model accurately predicted that consolidation was not achievable at CT < 350°C. Predictions of the model were less accurate for lower CP molding conditions. However, failure of the model for the moldings at 371°C/100 Psi (e.g., Panels #7 and 8 in Table 7) is attributable to the poorer quality prepregs used.

Table 7. OHC strength values at various molding conditions

Panel ID	B-staging	CT/duration	CP, Psi	Test temp., °C	OHC strength, Ksi	Stad Devi. Ksi
1	250°C/1.0 hr	371°C/1.0 hr	200	RT	59.2	- - -
2	250°C/1.0 hr	371°C/1.0 hr	200	177 (wet)	44.2	- - -
3	250°C/1.0 hr	371°C/1.0 hr	175	RT	55.1	1.4
4	250°C/1.0 hr	371°C/1.0 hr	175	177 (wet)	43.9	2.4
5	250°C/1.0 hr	371°C/1.0 hr	160	RT	56.3	.5
6	250°C/1.0 hr	371°C/1.0 hr	160	177 (wet)	48.2	4.6
7*	250°C/1.0 hr	371°C/1.0 hr	100	RT	61.8	2.2
8*	250°C/1.0 hr	371°C/1.0 hr	100	177 (wet)	43.4	1.0
9	250°C/1.0 hr	360°C/1.0 hr	185	RT	54.6	1.8
10	250°C/1.0 hr	360°C/1.0 hr	185	177 (wet)	42.9	2
11	250°C/1.0 hr	355°C/1.0 hr	200	RT	52.9	.8
12	250°C/1.0 hr	355°C/1.0 hr	200	177 (wet)	37.1	1.8
13	250°C/1.0 hr	355°C/1.0 hr	175	RT	54.1	2.7
14	250°C/1.0 hr	355°C/1.0 hr	175	177 (wet)	32.9	4.1
15*	250°C/1.0 hr	350°C/1.0 hr	200	RT	58.4	0.1
16*	250°C/1.0 hr	350°C/1.0 hr	200	177 (wet)	36.5	1.8
17*	250°C/1.0 hr	350°C/1.0 hr	175	RT	57.5	1.1
18*	250°C/1.0 hr	350°C/1.0 hr	175	177 (wet)	34.3	2.2
19*	250°C/1.0 hr	350°C/1.0 hr	150	RT	52.4	8.0
20*	250°C/1.0 hr	350°C/1.0 hr	150	177 (wet)	35.3	1.7
21*	250°C/1.0 hr	340°C/1.0 hr	200	RT	44.7	2.7
22*	250°C/1.0 hr	340°C/1.0 hr	200	177 (wet)	31.9	1.8

*ICI Fiberite prepregs

3. CONCLUSIONS

Processing robustness of IM7/PETI composite was investigated with respect to the effect of B-staging conditions, the prepreg shelf life, and the optimal processing window. Rheological measurements suggest that processability of the PETI matrix resin is only slightly affected over a wide range of B-stage temperatures from 250°C to 300°C. Both RT and 177°C (wet) OHC strength values were statistically indistinguishable among composite specimens B-staged at 250°C/0.5 hr, 250°C/1 hr, 300°C/0.5 hr and 300°C/1 hr, respectively and consolidated at 371°C for 1 hr. Such a wide B-stage window allows IM7/PETI prepreg to accommodate processing variations associated with different sizes of fabrication tools and composite parts.

Rheological measurements were also found useful in the processability evaluation of ambient stored prepregs. Prepreg rheology was not affected by prolonged (i.e., up to 60 days) ambient storage. Critical prepreg characteristics such as the initial softening temperature of matrix resin (i.e., ~ 260°C) and the temperature of minimum viscosity (i.e., 371°C) were all retained by the aged prepregs. Both RT and 177°C (wet) OHC strength values were found to be unaffected as well.

Using Box-Wilson Central Composite design for the optimization of molding conditions, an optimal processing window was identified. This processing window as derived by the regressional model was also investigated using the composite OHC strength measurements. It was found that the response surface methodology employed was a useful tool in process optimization. The IM7/PETI composite is more sensitive to the consolidation temperature than to the consolidation pressure. At maximum allowable temperature, i.e., 371°C, good consolidation was achievable at 100 Psi. However, processability dropped dramatically at consolidation below 350°C. At 340°C/200 Psi, for example, the RT OHC strength was reduced from a nominal 56 Ksi to 45 Ksi. This was attributed to the reduced fluidity associated with the semi-crystalline matrix, which was generated during the 250°C/1 hr B-staging used throughout this investigation.

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Standard Cycle For IM7/LARC-PETI-5 Composites

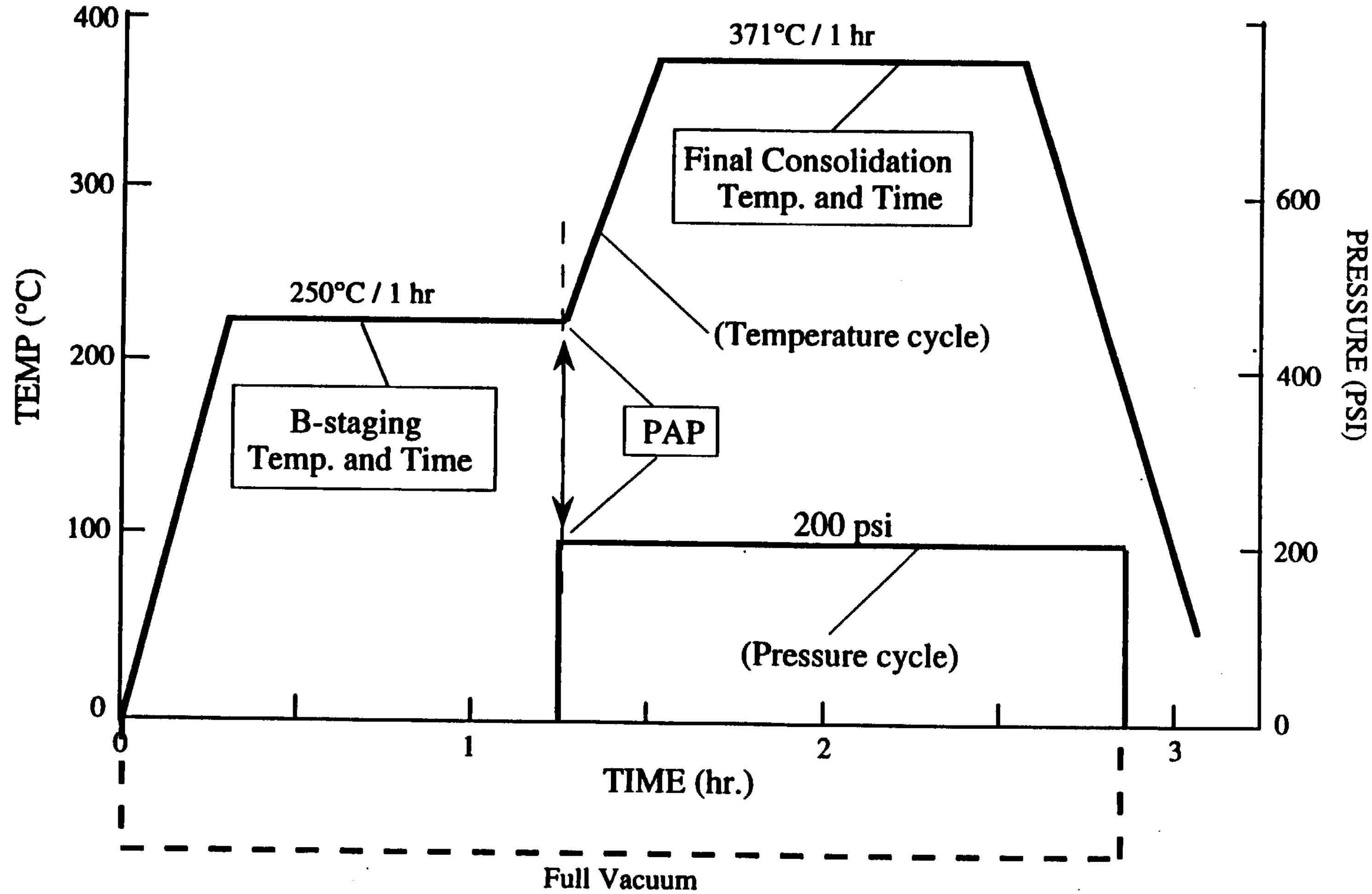

Figure 1 "Standard Cycle" for IM7/PETI composite molding.

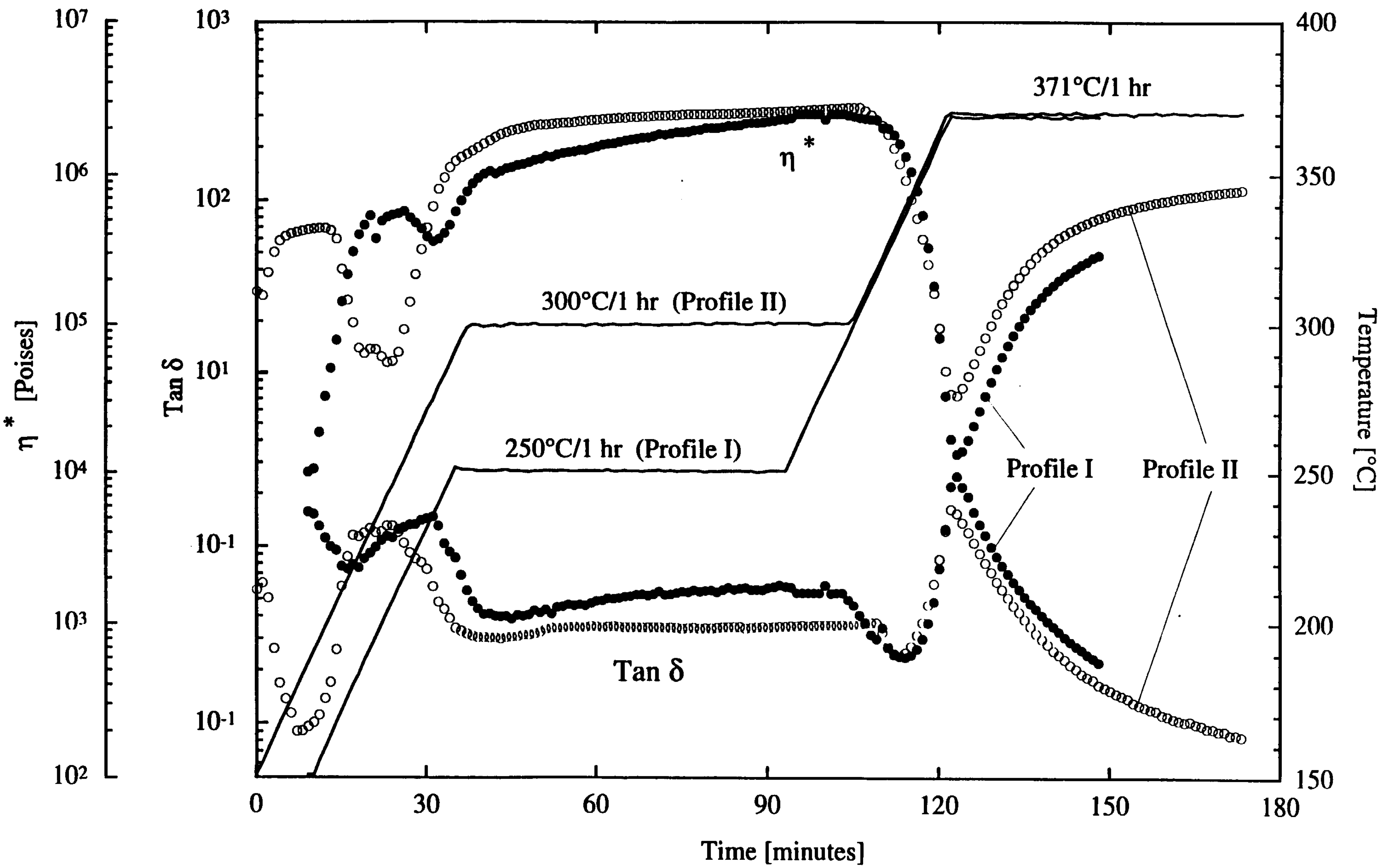

Figure 2. Rheological properties of PETI resins cured under two temperature profiles with 250°C/1 hr and 300°C/1 hr B-stage steps, respectively.

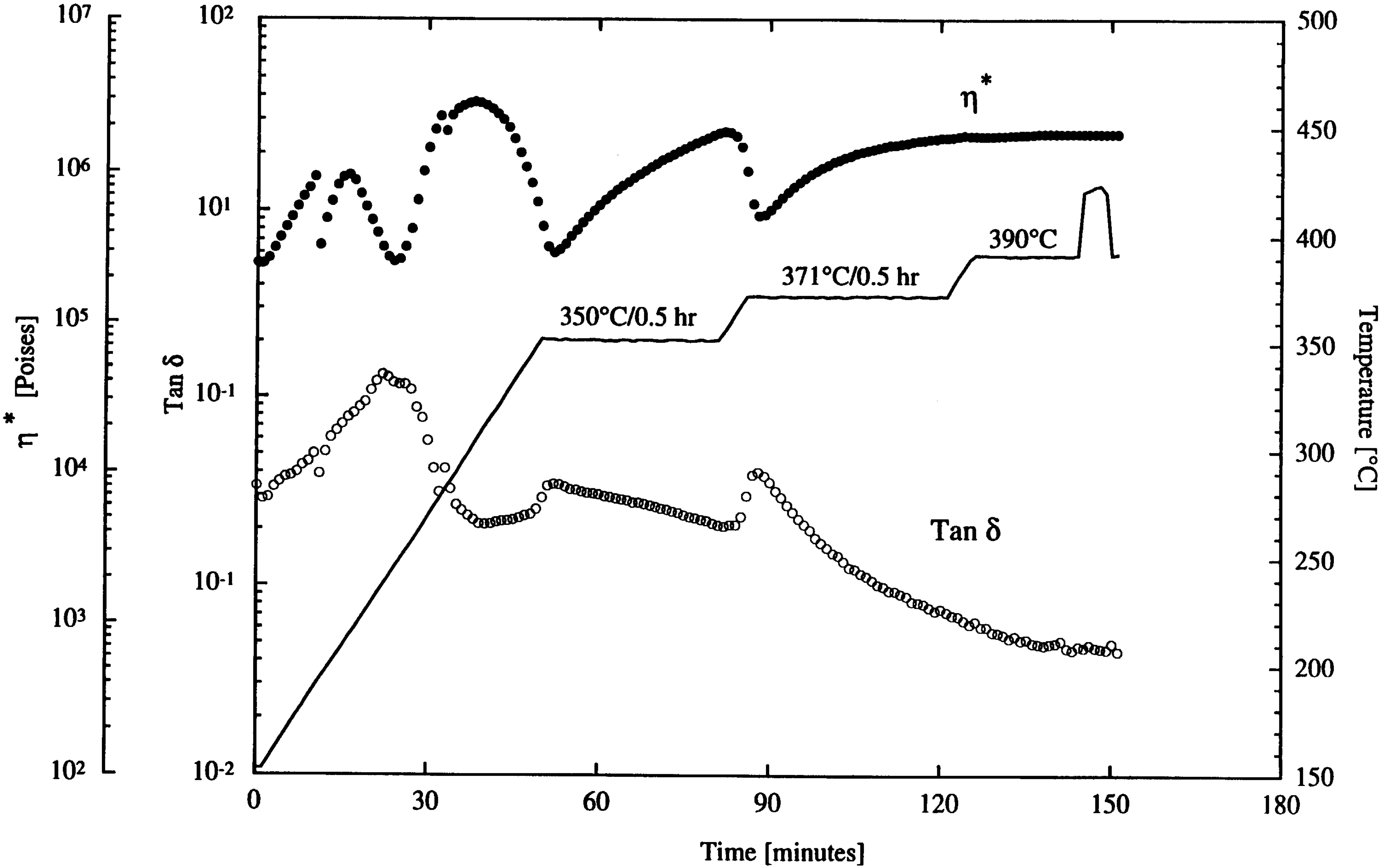

Figure 3. Rheological properties of PETI resins cured under a temperature profile with a 350°C/1 hr B-stage step.

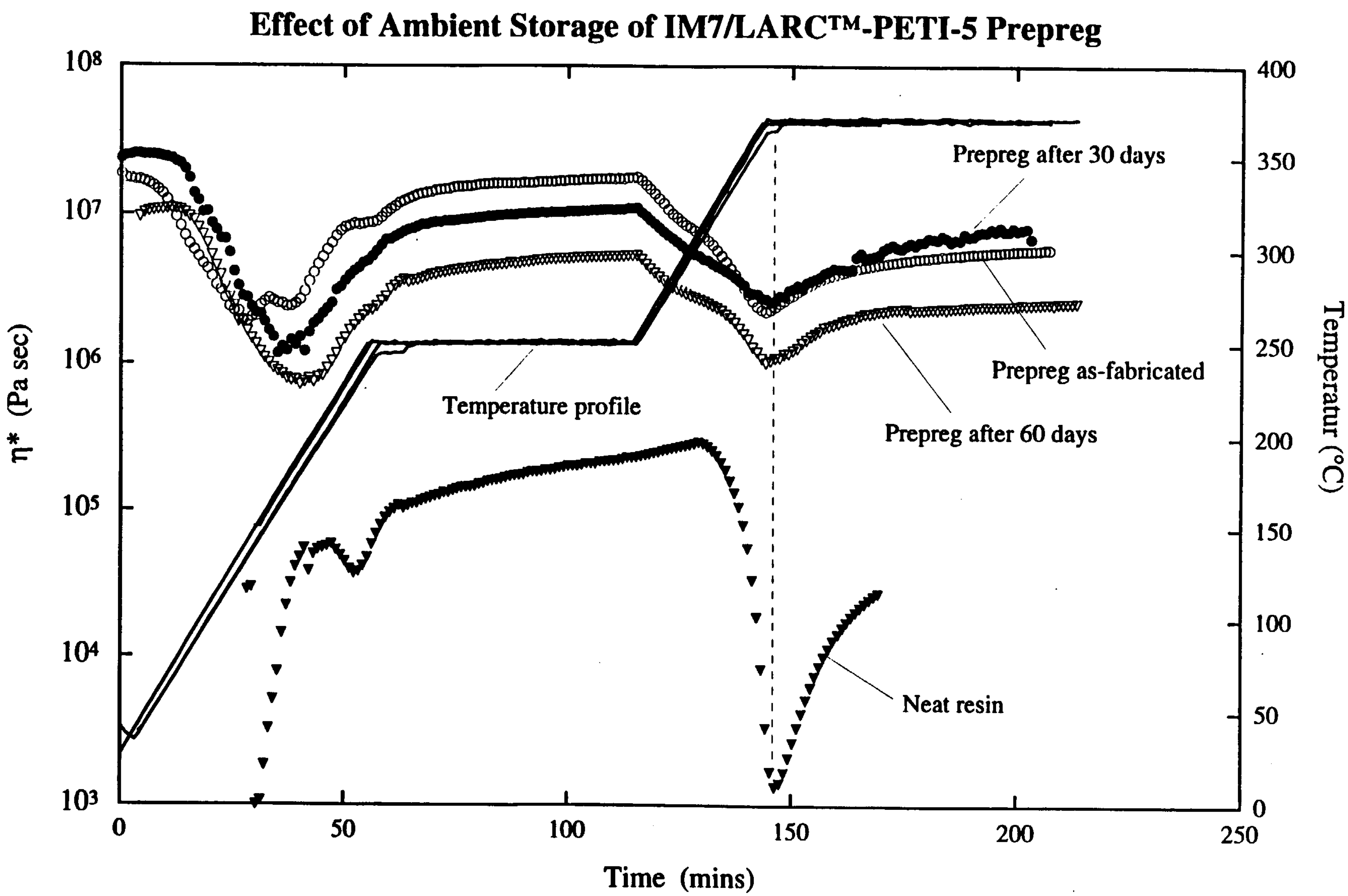

Figure 4. Rheological properties of IM7/PETI prepregs: as-fabricated, 30-day and 60-day ambient stored, respectively, cured under the "Standard Cycle". Rheological properties of neat PETI resin cured by the same cycle was also included.

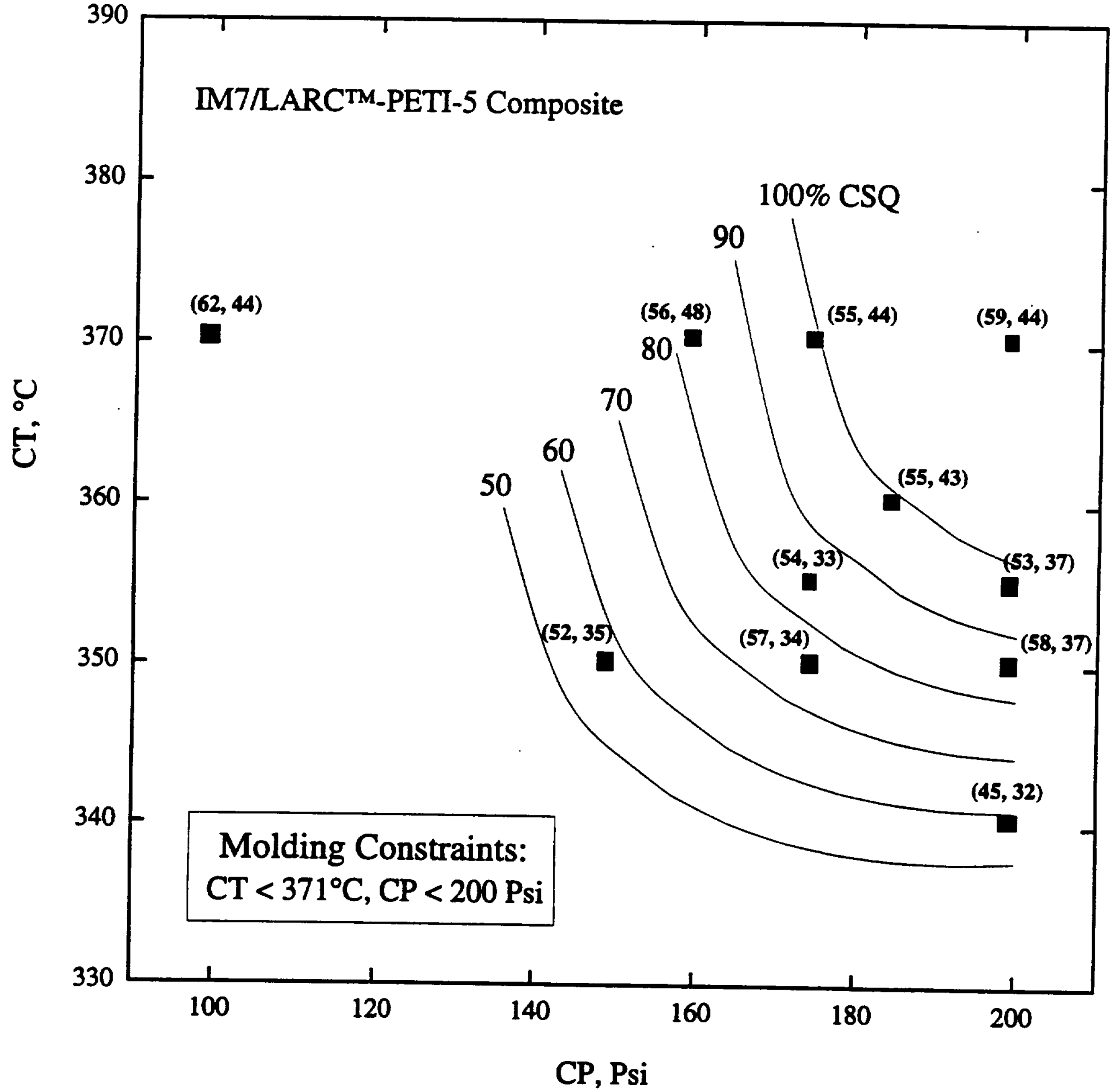

Figure 5. Response surface mapping constructed by the regression model derived for the optimal IM7/PETI molding cycle. RT and 177°C (wet) OHC strength values were also measured at discrete molding conditions.

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13. ABSTRACT (Maximum 200 words)

The processability of a phenylethynyl terminated imide (PETI) resin matrix composite was investigated. Unidirectional prepregs were made by coating an N-methylpyrrolidone solution of the amide acid oligomer onto unsized IM7. Two batches of prepregs were used: one was made by NASA in-house, and the other was from an industrial source. The composite processing robustness was investigated with respect to the effect of B-staging conditions, the prepreg shelf life, and the optimal processing window. Rheological measurements indicated that PETI's processability was only slightly affected over a wide range of B-staging temperatures (from 250°C to 300°C). The open hole compression (OHC) strength values were statistically indistinguishable among specimens consolidated using various B-staging conditions. Prepreg rheology and OHC strengths were also found not to be affected by prolonged (i.e., up to 60 days) ambient storage. An optimal processing window was established using response surface methodology. It was found that IM7/PETI composite is more sensitive to the consolidation temperature than to the consolidation pressure. A good consolidation was achievable at 371°C/100 Psi, which yielded an OHC strength of 62 Ksi at room temperature. However, processability declined dramatically at temperatures below 350°C.

14. SUBJECT TERMS B-stage, Composite Molding Cycle, Composite Processability, Composite Shelf Life, Response Surface Methodology, Carbon Fiber Composite, Mechanical Properties	15. NUMBER OF PAGES 21 16. PRICE CODE A03

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