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IMPROVED COMPRESSION MOLDING TECHNOLOGY FOR CONTINUOUS FIBER REINFORCED COMPOSITE LAMINATES - Part II: AS-4/Polyimidesulfone Prepreg System

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Table of Contents

FOREWORD

This work was conducted in Polymeric Materials Branch (PMB), NASA Langley Research Center (LaRC). This report represents Part II of a series of investigations on the use of an improved compression molding procedure for the fabrication of continuous fiber reinforced composite laminates. Part I was published as a NASA Contractor Report CR-187572 (May 1991), which dealt with the molding of AS-4/LaRC-TPI 1500 (HFG) thermoplastic prepreg system. This work (Part II) deals with the AS-4/Polyimidesulfone prepreg system which undergoes imidization reaction during lamination processing at elevated temperatures.

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IMPROVED COMPRESSION MOLDING TECHNOLOGY FOR CONTINUOUS FIBER REINFORCED COMPOSITE LAMINATES - PART II: AS-4/Polyimidesulfone Prepreg System

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ABSTRACT

AS-4/polyimidesulfone ($PISO_2$) composite prepreg was utilized for this improved compression molding technology investigation. This improved technique employed molding stops which advantageously facilitates the escape of volatile by-products during the B-stage curing step, and effectively minimizes the neutralization of the consolidating pressure by intimate interply fiber-fiber contact within the laminate in the subsequent molding cycle. Without modifying the resin matrix properties, composite panels with both unidirectional and angled plies with outstanding C-scans and mechanical properties were successfully molded using moderate molding conditions, i.e., 660°F and 500 psi, using this technique. The size of the panels molded were up to 6.00" x 6.00" x 0.07". A consolidation theory was proposed for the understanding and advancement of the processing science. Processing parameters such as vacuum, pressure cycle design, prepreg quality, etc. were explored.

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INTRODUCTION

Compression molding flat panels of fiber reinforced resin matrix composite laminates is the simplest form of a molding process which employes matched metal dies. In this process, individual prepreg plies are cut into the desired dimensions from a flat sheet of prepreg material. The number of prepreg plies is dictated by the desired final part thickness. These plies are stacked inside the cavity of the female mold and subjected to consolidation under heat and compression force. The prepreg plies can be oriented during the stacking sequence to yield orthotropic, quasi-isotropic or isotropic laminates as required for various applications.

Despite the simplicity in tooling design, the compression molding of composite laminates is by no means a trivial process [1-10]. Due to the extremely high viscosity of polyimide resins in their fully imidized state, the impregnation of resin into the fiber tows becomes very difficult. It is frequently easier to impregnate material by the technique of solution prepregging. Prepolymers with a low degree of imidization reaction are first dissolved in solvents and subsequently coated onto the fiber tows and drum wound to form the prepreg. During the molding cycle, the resin matrix is further imidized under elevated temperature and consolidated by pressure [11-13].

In order to achieve fully consolidated, void-free composite parts with good mechanical properties, various molding parameters need to be understood and controlled. For the class of prepreg system investigated here, the volatile by-product escape mechanism and the bulk consolidation behavior are the two molding parameters which require extra attention. From a processing point of view, these two parameters work against each other and some compromises have to be made. It is understood that full consolidation of the composite part requires pressure to squeeze out voids formed by the volatile by-products and to facilitate interply prepreg layer fusion of the resin matrix. However, pressure applied too early in the molding cycle will effectively block the volatile escape paths, which leads to a poor quality C-scan of the part. On the other hand, higher resin viscosity associated with an advanced degree of imidization tends to trap the volatiles within the viscous matrix and make the delayed pressure application ineffective in void-free consolidation of the part.

A consolidation theory is proposed which provides a guideline for a molding cycle (temperature and pressure profile) design that adjusts various molding parameters. This

technique was demonstrated before with AS-4/LaRC-TPI 1500 fully imidized thermoplastic prepreg [14]. In the present investigation we deal with a reactive prepreg system. It is demonstrated that well consolidated composite panels with exceptional C-scans and mechanical properties can be consistently molded by this method with a moderate pressure level, as opposed to conventional molding techniques requiring much higher pressure levels.

MOLDING EXPERIMENTS

Materials

Polyimidesulfone polymer was invented by NASA Langley Research Center in the early 1980s [15,16]. This material was subsequently licenced and manufactured by High Technology Services, Inc. (HTS), Troy, New York. The resin system, Techimer 2001, used in this work was used in the as-received condition from HTS with 30% $PISO_2$ solids in diglyme solvent solution. Unidirectional AS-4 unsized fibers with 12,000 filaments per tows were impregnated and drum wound with the resin solution to form carbon fiber prepreg. For various batches of prepreg material, the solids content of the solution used in the prepregging process ranged from 27 to 40%.

The drum wound prepreg was then cut into sheets, sealed in plastic bags, and stored in the freezer at 25°F (-4°C) until required for molding.

Effect of Vacuum on the Consolidation Process

Run #406 (with vacuum)

A 12 ply 3.00" x 3.00" composite laminate (40% resin content by wt.) was processed by a vacuum press according to the cure cycle shown in Table 1. The cure cycle consists of four cure steps with various combinations of temperature and pressure. Each step consists of 0.5 hour hold at temperature. A full vacuum of 29" Hg was used at each step in the cycle. The cycle was interrupted at the end of each cure step so that information regarding any geometrical change and laminate consolidation quality (C-scan) could be measured. The results are tabulated in Table 1. In the first two steps (400 and 500°F) of the cure, the molding process included the following: two stops measuring 3.00" long, by 0.125" height and 0.156" wide were inserted at the sides of the mold. Prepreg plies were cut to a size of 3.00" x 2.68" and stacked between the stops. The height of the stacked laminate was higher than the stops at room temperature. During the molding process at cure steps 1 and 2, pressure was applied to compress the laminate to the thickness of the stops. The laminate pressure was essentially zero when the male mold contacted the stops but the mold remained in contact with and continued to supply heat through conduction from the

platen to the laminate. This loosely compacted structure in the unconsolidated laminate provides the necessary paths for the volatiles. It is noted that at the end of the second 500°F cure step, about 89% (3.66 vs. 4.11g) of the total volatiles generated during the entire cure cycle were depleted from the specimen. The thickness of the laminate does not change much, as anticipated. The C-scans are poor, however, as shown in Figure 1, because of the volatile paths remaining in the laminate have not been collapsed. The stops were removed from the mold after the second (500°F/0psi/.5hr.) cure step was completed. At the 600°F cure step a pressure of 400 psi was applied to help consolidate the laminate. A large change in thickness was observed, and the C-scan shows a dramatic improvement in the quality of the laminate. The consolidation pressure was raised to 2,000 psi during the final 660°F cure step. As a result, the laminate thickness decreases slightly, while the C-scan shows that complete consolidation with excellent quality is reached.

Table 1. Geometrical changes of the 12 ply unidirectional3.00"x3.00" composite laminate in Run #406

	Press Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	24.90	---	---	---
1	400/0/.5	22.081	2.82	11.32	0.131±.001
2	500/0/.5	21.24	3.66	14.70	0.127±.001
3	600/400/.5	21.072	3.83	15.37	0.093±.002
4	660/2000/.5	20.79	4.11	16.51	0.089±.001

There was very little excess resin squeezed out of the laminate during the entire cure cycle. It was noted, however, that at the completion of 600°F/400psi/.5hr. cure step (after removal of the stops from the mold), a 0.32" in-plane lateral (perpendicular to the fiber direction) dimensional change occured, and a much smaller change of 0.034" measured in the thickness direction (see Table 1). The observed apparent unbalance (with a factor of 3.5) in the conservation of volume (0.32" x 3.00" x 0.093" = 0.087 in^3 increase from the dimensional change in the lateral direction vs. 0.034" x 3.00" x 3.00" = 0.306 in^3 decrease due to the dimensional change in the thickness direction) suggests that a considerable amount of voids had been filled by the resin due to the applied consolidation pressure

which, in turn, leads to an improved laminate C-scan as shown in cure step 3, Figure 1. These observations also indicate that the $PISO_2$ resin system was highly viscous when the consolidation was attempted at the third cure step (600°F/400psi/.5hr.). The resin had been advanced to a degree of imidization which offered a certain level of adhesive strength in the fiber-matrix interface. Because of the unevenness of the in-plane resin distribution, the resin was forced by the consolidation pressure to diffuse (instead of flowing) through the fibrous structure in the in-plane direction, and the filaments of the fiber tows were rearranged accordingly during the consolidation process - remaining essentially parallel.

Optical microscopic pictures of the edges of the laminate are shown in Figure 2. Both the end view (fiber direction) and the side view (in-plane lateral direction) of the laminate after cure step 1 (400°F/0psi/0.5hrs) are included for comparison. An uneven outflow of resin in the fiber direction between the plies is visible. It is clearly shown that the permeability of the fibrous structure in the fiber direction predominates over both the perpendicular and transverse in-plane lateral direction. Also included in Figure 2 is the end view of the laminate after cure step 3 (600°F/400psi/0.5hrs). At this step 400psi pressure was applied which caused a dimensional change in the transverse direction (see Table 1). A smaller amount of resin was squeezed to the edge. The laminate is consolidated and contains embeded wavy filament tows within the prepreg plies. This indicates unevenness in the resin distribution and/or the status of cure during the consolidation process.

Run #412 (without vacuum)

In order to compare the effect of vacuum on the consolidation process between conventional (nonvacuum) and vacuum compression molding of $PISO_2$ composite laminates (40% resin content by weight), 11 ply 3.00" x 3.00" panel was compression molded without vacuum using the same press cycle and stops used with Run #406 in Table 1. The results of dimensional changes and C-scans of the panel are shown in Table 2 and Figure 3. Although marginally fewer imperfections are noted for the panel processed without vacuum when comparing the series of C-scans in Figures 1 and 3, a similar pattern of consolidation behavior is clearly exhibited at each of the four cure steps. The molding process without vacuum shows a lower level (14.4 vs 16.51%, from Tables 2 and 1, respectively) of percentage weight loss, which may contribute to the observed marginally inferior C-scan (Figure 3) quality in the final cure step of Run #412.

Owing to the use of stops within the mold, the thickness of the laminate, as tabulated in Table 2, did not change in the first two cure steps. A similar unbalance in the

conservation of volume is observed at the completion of 400 psi pressure applied at 600°F (cure step 3) with the stops removed. The laminate is seen to reduce 0.036" in thickness and expand 0.32" in the in-plane lateral dimension to reach a full 3.00" in width. Optical photomicrographs of Run #412 panel edges are shown in Figure 4. Similar characteristics were exhibited at the four cure steps during processing with vacuum in Run #406 as shown in Figure 2. Such a similarity indicates that the uneven outflow of resin was caused by the pressure 100 psi used to compress the laminate to the height of the stops while the matrix resin was melting. Vacuum had little or no influence on the resin flow.

A direct comparison on the surface characteristics of prepreg sheets which were B-staged either with or without employing vacuum was also studied. Four B-stage temperatures, i.e., 400°, 500°, 600° and 660°F, with 0.5 hrs. duration each, were used and the optical photographs are shown in Figure 5. Similar foamy surface characteristics were noted for these two sets of prepreg.

It was therefore concluded that the application of vacuum in compression molding $PISO_2$ composite laminates is helpful, but not a critical factor in achieving a consolidated laminate with good C-scan quality.

Table 2. Geometrical changes of the 11 ply unidirectional 3.00"x3.00" composite laminate in Run #412

	Press Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	22.589	---	---	---
1	400/0/.5	19.843	2.75	12.2	0.124±.004
2	500/0/.5	19.426	3.16	14.0	0.123±.002
3	600/400/.5	19.388	3.20	14.2	0.087±.005
4	660/2000/.5	19.330	3.26	14.4	0.083±.005

Effect of Consolidation on the Pressure Cycle Design

Run #409

The pressure application time during a cure cycle (temperature profile) is the single most important parameter in the pressure cycle design for the molding of composite laminates. In order to design an optimal pressure cycle, a determination of the critical volatile depletion level is required. A set of experiments was designed and the results are tabulated in Table 3 and shown in Figure 6. The press cycle differs from that of Run #406 (Table 1) only in cure step 2, where, with the stops removed, 100 psi is applied for 0.5 hours. Cure step 1 (400/0/0.5) in both runs resulted in a comparable level of weight loss (11.32 vs. 11.6%, from Table 1 and 3, respectively). However, a 1.6% less weight loss (14.7 vs. 13.1%) is realized in the cure step 2 of Run #409. A C-scan shows that this extra pressure applied during cure step 2 in Run #409 does not benefit the laminate consolidation at all. Instead, it was detrimental to the consolidation process of later cure steps, by dramatically reducing the volatile escape channels within the laminate, as can be seen from the significant change of thickness (0.125" to 0.106"). C-scans of cure steps 3 and 4 (Figure 6) show poorer quality than that of Run #406 (Figure 1), despite the use of identical processing conditions.

Table 3. Geometrical changes of the 11 ply unidirectional 3.00"x3.00" composite laminate in Run #409

	Press Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	22.49	---	---	---
1	400/0/.5	19.88	2.61	11.6	0.125±.001
2	500/100/.5	19.54	2.95	13.1	0.106±.002
3	600/400/.5	19.46	3.03	13.5	0.089±.002
4	660/2000/.5	19.24	3.25	14.5	0.085±.001

Based on the results discussed above, it is concluded that timing of the pressure cycle application is extremely critical. Early application of pressure in the cure cycle can be harmful to the consolidation process by effectively blocking the volatiles escape paths

within the laminate. Preliminary results indicate that the consolidation pressure can be safely applied when the weight loss of the (40% resin content) $PISO_2$ laminate reaches about 14.5% (or equivalent to about 90% volatile depletion), but not at the 11.6% wt. loss level.

Run # 408

Due to the success of the cure cycle employed in Table 1 which produced a $PISO_2$ composite laminate panel with an excellent quality C-scan, a set of experiments was next designed to investigate the possibility of reducing the consolidation pressure. The resin content of the prepreg was kept at 40% by wt.. Experimental results and the C-scan of each cure step are shown in Table 4 and Figure 7. The employed press cycle produced a consolidated laminate with inadequate C-scans (see Figure 7, steps 3 and 4). When compared with the geometrical changes of the composite panel of Run #406 in Table 1, we note immediately that the reduced pressure in cure step 3 (600°F/200psi/0.5hr) resulted in a panel about 10% thicker (.104" vs. .93" from Tables 4 and 1, respectively). The cure steps using a reduced consolidation pressure (steps 3 and 4) also led to a reduced rate of percentage weight loss.

Table 4. Geometrical changes of the 12 ply unidirectional 3.00"x3.00" composite laminate in Run # 408

	Press Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	24.72	---	---	---
1	400/0/.5	21.78	2.94	11.91	0.132±.001
2	500/0/.5	21.13	3.59	14.50	0.129±.002
3	600/200/.5	21.05	3.67	14.86	0.104±.002
4	660/1000/1.5	20.72	4.00	16.20	0.091±.001
5	660/2000/.5	20.46	4.26	17.20	0.089±.001

Judging from the C-scans in Figure 7, it is conceivable that cure step 3, with the reduced consolidation pressure, leaves a considerable amount of unfilled micro-void pockets inside the laminate. Because of the continuous build up in the resin viscosity due to the imidization reaction subsequent to cure step 4 (660/1000/0.5) using a reduced pressure of 1000 psi is not large enough to achieve the same degree of consolidation as did the higher pressure (2000 psi) used in cure step 4 (660/2000/0.5) of Run #406 (Table 1). It is also noted that a longer consolidation time, 1.5 vs. 0.5 hrs, was used in cure step 4. Such a prolonged duration at a reduced pressure level did not result in a consolidated panel with good quality (comparing the C-scans of step 4, Figures 1 and 7).

This observation suggests that the level of pressure applied is more critical than the length of the cure time employed in the latter stages of the consolidation process for the $PISO_2$ composite laminate, where the resin has reached an appreciable level of rigidity under the prescribed processing temperatures. Finally, a cure step 5 (660/2000/0.5) is applied to the laminate. A C-scan of the resulting panel in Figure 7 again indicates that an improved consolidation quality can be achieved by an elevated pressure (2000 psi) within a short period of time (0.5 hrs.).

Run #411

The search for the minimum consolidation pressure in the compression molding of $PISO_2$ composite laminates led to another set of experiments, following the cure cycle shown in Table 5. This cure cycle differs from the one used in Run #408 (Table 4) only in cure step 4, where 1500 psi is employed. The C-scan results are shown in Figure 8. C-scans of cure steps 1 and 2, with the aid of stops, are identical to those reported before, and are not included in Figure 8. Comparing these C-scans with those shown in Figure 7 (Run #408), it is established that 1500 psi pressure applied for 0.5 hrs. in the final stage (cure step 4) of consolidation yields a consolidated panel with a much improved C-scan quality.

When comparing the results with those shown in Figure 1 (Run #406), it is noted, however, that a reduced pressure of 1500 psi for 0.5 hrs. in the final stage (cure step 4) of the consolidation process does not achieve the same level of C-scan quality (see cure step

4, Figure 1). This is attributable to the 200 psi pressure applied prematurely in the preceding curing step 3.

Table 5. Geometrical changes of the 11 ply unidirectional 3.00"x3.00" composite laminate in Run # 411

	Press Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	22.37	---	---	---
1	400/0/.5	19.67	2.70	12.08	0.128±.002
2	500/0/.5	19.33	3.04	13.61	0.127±.001
3	600/200/.5	19.27	3.10	13.87	0.099±.002
4	660/1500/.5	19.23	3.14	14.05	0.082±.002

The required consolidation pressure of 2,000 psi was considered unacceptably high in these molding experiments of the AS-4/$PISO_2$ composite system discussed above. However, such a high pressure level could never produce the same level of consolidation for panels with the fully cut pattern used in the conventional molding process without the aid of stops. In addition, the need of using a high consolidation pressure was attributed to the poor prepreg quality as will be discussed later. Further, an improved molding cycle using a moderate pressure of 500 psi had been developed for a new prepreg batch with better quality than the one used in the experiments discussed so far.

Effect of Prepreg Quality on the Cure Cycle Design

A new batch of drum wound $PISO_2$ prepreg designated by the letters CS was prepared for molding. Photographs of the CS prepreg and the prepreg used to date in the development study of the consolidation process as described above are shown in Figure 9

for comparison. It is noted that the CS prepreg exhibits not only a smoother surface but is also more uniform in resin distribution without apparent spots or strips of resin starvation. This batch of $PISO_2$ CS prepreg has a resin content of 46.64% and will be used in the following composite laminate consolidation studies.

Run CS270

A 10 ply unidirectional panel was compression molded under vacuum following the same cure cycle as Run #406 in Table 1. The geometrical changes of the composite panel and the evolution of C-scan quality between cure steps are shown in Table 6 and Figure 10, respectively. Measurements are absent for cure step 1 (400°F/0psi/0.5hrs) because the molding process from cure step 1 to 2 was not interrupted during the run. The C-scan of cure step 2 (500°F/0psi/0.5hrs), with the aid of stops, is identical to that shown in Figure 1, and is not included in the Figure.

Table 6. Geometrical changes of the 10 ply unidirectional 3.00"x3.00" composite laminate in Run CS270

	Press Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	18.92	---	---	---
1	400/0/.5	---	---	---	---
2	500/0/.5	15.39	3.53	18.65	0.122±.002
3	600/400/.5	15.35	3.57	18.88	0.071±.001
4	660/2000/.5	15.26	3.66	19.33	0.069±.001

It is noted that cure step 3 yields a panel with similar consolidation quality as that of Run #406 when comparing the corresponding C-scans in Figures 1 and 10. However, at the completion of cure step 4, the panel consolidation quality is worse as shown by the C-scan. A closed examination of the thickness changes between the molding steps of panels #406 and CS270 from Tables 1 and 6 shows that a comparable laminate thickness (.127" vs. .122") is initially achieved due to the use of molding stops in both runs. Subsequent thickness reduction in step 3 (with stops removed) using a consolidation pressure of 400 psi shows that the panel of Run CS270 was 9.2% thinner than that of Run #406 when

compared on a per ply basis (7.1 vs. 7.75 mils). Considering that identical cure steps were followed by both panels, such a difference was likely attributed to the variations of prepreg batches.

It is conceivable that this extra reduction in thickness of panel CS270 effectively blocked the volatiles escape paths within the laminate, so that the by-products of further reactions occuring at the elevated temperature of cure step 4 (660°F) were essentially trapped in, and contributed to the observed deterioration of the panel C-scan quality.

Run CS271

In order to verify the reasons postulated above, an experimental cure cycle was designed and is tabulated in Table 7. In this experiment, the molding process through cure steps 1 to 3 was not interrupted and the molding stops were used throughout the cycle. As a result, at the completion of cure step 3, the panel thickness (.124") is close to the thickness of the stops. The C-scan shown in Figure 11 reveals a completely unconsolidated laminate due to the absence of consolidation pressure. The lack of consolidation also provides the desired volatile escape paths within the laminate for the subsequent curing step at elevated temperature. During final cure step 4, a consolidation pressure of 2,000 psi was applied, and a void free composite panel was achieved as can be seen from Figure 11.

Table 7. Geometrical changes of the 10 ply unidirectional, 3.00"x3.00" composite laminate in Run CS271

	Press Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	18.85	---	---	---
1	400/0/.5	---	---	---	---
2	500/0/.5	---	---	---	---
3	600/0/.5	15.58	3.27	17.33	0.124±.001
4	660/2000/.5	15.54	3.31	17.56	0.069±.001
5	695/500/.5	---	---	---	---

The above discussion indicates that the prepreg quality is very critical to the development of the molding process for a composite laminate. For a given resin matrix composite system, different prepreg qualities cause the design of completely different consolidation cycles in order to yield laminate panels with good C-scans.

The consolidated composite panel was also subjected to high temperature aging at 600°F for 16 hrs. Bowed and blistered surface appearances resulted. This is expected because of the severe aging condition (about 210°F above the matrix glass transition temperature) employed. This aged panel was then compression molded at 695°F and 500 psi. A well consolidated laminate with an excellent C-scan resulted as can be seen in Figure 11.

Effect of Oven B-staging on the Cure Cycle Design

Run CS278

Due to the success of the molding cycle developed for the compression molding of $PISO_2$ composite laminates with superior consolidation quality (e.g., Run CS271, Figure 10), a natural extension of the molding cycle development is therefore to conduct cure steps 1 to 3 for the laminate in a conventional hot air circulating oven using molding stops, instead of the press.

A 10 ply unidirectional $PISO_2$ laminate was stacked in a 3.00" x 3.00" female mold. With a male mold piece weighing 6.3 lbs (equivalent to 0.7 psi) resting on top of the laminate stack, the whole assembly was placed in a conventional forced air circulation oven and cured according to the molding cycle outlined in Table 8. At the completion of cure step 3 in the oven, the mold was taken out of the oven and placed in the vacuum press which was used to complete the final cure step, as shown in Table 8 and Figure 12.

The resultant panel possessed comparable dimensions and consolidation quality to the other panels previously discussed. This experiment also demonstrated that with the aid

of molding stops a much lower pressure (500 psi) is adequate for the molding of $PISO_2$ panel with superior consolidation quality.

Table 8. Geometrical changes of the 10 ply unidirectional 3.00"x3.00" composite laminate in Run CS278

	Molding Cycle* (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	18.76	---	---	---
1	400/0/.5	---	---	---	---
2	500/0/.5	---	---	---	---
3	600/0/.5	15.35	3.42	18.19	0.164±0.016
4	660/500/.5	15.24	3.52	18.76	0.069±0.001

*Cure steps 1 to 3 were conducted in a forced air circulation oven without interruption. Cure step 4 was conducted in a vacuum press under the conditions indicated.

Run CS277

A run following the cure cycle identical to that of Run CS278 was conducted. The processing data are tabulated in Table 9. Once again the C-scan for the final composite panel exhibits excellent consolidation quality as shown in Figure 13.

Table 9. Geometrical changes of the 10 ply unidirectional 3.00"x3.00" composite laminate in Run CS277

	Molding Cycle* (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	19.39	---	---	---
1	400/0/.5	---	---	---	---
2	500/0/.5	---	---	---	---
3	600/0/.5	---	---	---	---
4	660/500/.5	15.00	4.39	22.64	0.068±.001

*Cure steps 1 to 3 were conducted in a forced air circulation oven without interruption. Cure step 4 was conducted in a vacuum press under the conditions indicated.

The successful extension of the process to include the use of an air circulating oven has greatly enhanced the usefulness of this molding cycle. The obvious benefits include the reduction of the processing cycle time and the use of mass production for the manufacture of $PISO_2$ composite laminates.

The reasons for the success in using the cure cycle designed for this investigation, which consistently yielded well consolidated polyimidesulfone composite laminates with superior C-scans, are attributed to the implementation of the two following critical processing techniques:

1) Use of molding stops inside the mold during the B-stage period of the cure cycle for the composite laminate. During this period, the composite laminate sees effectively zero consolidation pressure, which results in a loosely packed structure with abundant volatile escape paths for the reaction by-products.

2) For the final step of the cure cycle, the molding stops are removed from the mold before the external load is applied. A rearranged fiber/resin matrix composite structure resulted from the application of consolidation pressure, which filled the excess volume inside the mold created by the removal of the molding stops. Such an excess volume is critical in the final consolidation process because it offers a possible side way movement of the fiber/resin matrix in the lateral direction, and consequently the voids are suppressed by the consolidation pressure, which is otherwise absorbed by the intimate interply fiber-fiber contacts within the laminate.

Run CS279

A molding experiment was conducted with lower consolidation pressure than that used in runs CS277 and 278. The molding cycle and the geometrical changes of the composite panel are tabulated in Table 10. A 10 uni-ply $PISO_2$ laminate was stacked in a

3.00" x 3.00" female mold. With a male mold piece weighing 6.3 lbs (equivalent to 0.7 psi) mounted on the top of the laminate, the whole assembly was B-staged in a conventional forced air circulation oven at 400, 500 and 600°F for 0.5 hours at each step. At the completion of cure step 3 (600°F) in the oven, the mold was taken out of the oven and placed in the vacuum press which was used to complete the consolidation cure step at 660°F for 0.5 hours, with 250 psi applied throughout the step. As shown in Table 10, both the weight loss and the panel thickness are comparable to that of run CS278 (Table 8) at the completion of cure step 4. However, the C-scan shown in Figure 14 indicates a panel with only about 60% void free consolidation. Obviously, the pressure of 250 psi used in the consolidation step is inadequate.

Table 10. Geometrical changes of the 10 ply unidirectional, 3.00"x3.00" composite laminate in Run CS279

	Molding Cycle* (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	18.00	---	---	---
1	400/0/.5	---	---	---	---
2	500/0/.5	---	---	---	---
3	600/0/.5	14.92	3.10	17.19	0.156±.011
4	660/250/.5	14.84	3.16	17.55	0.069±.001
5	675/400/.5	14.81	3.20	17.75	0.067±.001

*Cure steps 1 to 3 were conducted in a forced air circulation oven without interruption. Cure steps 4 and 5 were conducted in a vacuum press under the conditions indicated.

An effort was made to salvage this panel. It was compression remolded at 675°F for 0.5 hours, with a consolidation pressure of 400 psi (step 5 in Table 10). Few geometrical changes resulted. The C-scan shown in Figure 14 indicates little or no improvement in the consolidation quality of the panel.

It is conceivable that a considerable degree of the volatile escape paths within the composite laminate were closed earlier by the consolidation pressure (250 psi) applied at

cure step 4. Meanwhile, such a pressure was unfortunately too low to complete either the squeeze out of the volatiles or to fill the microvoids within the laminate resin matrix. Subsequently, the resultant blockage of the volatile escape paths was seen to effectively prevent further consolidation of the panel under the higher temperature and pressure of cure step 5. Also, the intimate interply fiber-fiber contact now occuring in the laminate prevents further compaction without using excessive pressure.

The significance of the critical timing for the application of pressure, as discussed in runs 408, 409 and 411 above, with regard to the composite panel consolidation quality can also be envisioned by comparing the C-scans of runs CS278 and CS279 discussed above.

Comparisons of Consolidation Quality Between Conventional and Improved Molding Technologies

Run CS1052 (Conventional)

We used a new batch of $PISO_2$ prepreg to conduct further molding experiments. This batch of prepreg was drum wound at 41% resin weight fraction measured by acid digestion. The acid digestion was performed in concentrated sulfuric acid mixed with an equal weight of 30% hydrogen peroxide.

A composite panel was molded according to the conventional molding technique. 10 ply unidirectional prepreg pieces 3.00"x3.00" were cut and stacked in a female mold with a cavity measuring exactly 3.00 inches by 3.00 inches. Without internal or external stops, the whole assembly with a male mold in place was placed in a vacuum press and cured according to the cycle tabulated in Table 11. The cycle was interrupted at the completion of each cure step, and the panel dimensions measured and C-scans taken.

The C-scan shown in Figure 15 reveals a progressive improvement in the composite consolidation quality as the cure cycle progressed. At the completion of the final cure step, however, only about 80% overall void free consolidation was achieved, despite the use of a very high pressure (2,000 psi) under an elevated molding temperature (660°F). It is noted from Table 11 that there is a dramatic reduction in the panel thickness from the initial 0.091 in. to about 0.061 in. at the completion of cure step 2. Less than 10% change

in thickness (from 0.061 to 0.056 in.) is observed in the subsequent cure steps. The panel thickness at the end of cure step 2 (0.061 in) is thinner than those (0.067 - 0.069 in.) observed in runs CS277-279, in spite of the fact that lower molding temperature and pressure were employed. Such a large thickness reduction observed in panel CS1052 is due to a lower viscosity level, which results from a lower degree of imidization reaction completed at the end of cure step 2.

The consolidation quality of the panel is therefore attributable to the premature application of pressure during an early stage of the cure cycle. It is conceivable that a significant degree of volatile escape paths were blocked at the end of cure step 2, as indicated by the dramatic thickness reduction, which results in a composite laminate with compacted structure. Such a compacted structure effectively prevents further consolidation in subsequent cure steps. A higher percentage of final weight loss (27.3%) is also noted for this panel. This is attributed to the observed higher degree of fiber/resin wash-out in the in-plane lateral direction in response to the high consolidation pressure applied.

Table 11. Geometrical changes of the 10 ply unidirectional, 3.00"x3.00" composite laminate in Run CS1052

	Molding Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	17.57	---	---	0.091
1	400/200/.5	---	---	---	---
2	500/200/.5	13.14	4.43	25.23	0.061±.002
3	600/500/.5	13.00	4.57	26.00	0.058±.001
4	660/2000/.5	12.79	4.78	27.30	0.056±.001

Run CS1051 (Improved)

A 20 ply unidirectional 3.00"x3.00" composite panel was molded in this experiment. The $PISO_2$ prepreg was initially cut to a dimension measuring 3.00" by 2.75".

Two molding stops measuring 0.125" each in width were added to each side of a 3.00" x 3.00" female mold. The prepreg plies were then fitted exactly to this 3.00" by 2.75" configuration. The cure cycle listed in Table 12 was used in a vacuum press. The cycle was interrupted at the completion of cure step 3 so that the geometrical changes of the panel could be measured. With the molding stops removed, the panel was then molded at the final cure step of 660°F with 500 psi pressure applied from the start to complete the cure cycle. The C-scan in Figure 16 reveals a composite panel with outstanding consolidation quality.

The resultant panel possesses in-plane dimensions measuring 3.00" by 3.00". This in-plane dimensional change indicates a lateral deformation occuring in the laminate structure in response to the consolidation pressure applied at cure step 4. It also accounts for a significant portion of the observed thickness reduction from 0.218" to 0.123". Such an expanded deformation in the lateral direction allows interply fiber-fiber nestings to occur. Consequently, microvoids created inside the laminate during the B-stage period of the cure cycle were suppressed or filled by the matrix resin and the consolidation of the fiber-resin composite structure was achieved.

When compared to panel CS1052, it is noted that a composite panel with much better consolidation quality is achieved by a less involved cure cycle and a lower consolidation pressure. The advantages of using the improved molding technique in CS1051 over the conventional technique used in CS1052 are clearly demonstrated by these experiments.

Table 12. Geometrical changes of the 20 ply unidirectional, 3.00"x3.00" composite laminate in Run CS1051

	Molding Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	34.54	---	---	---
1	400/0/.5	---	---	---	---
2	500/0/.5	---	---	---	---
3	600/0/.5	27.37	7.16	20.74	0.218±.002
4	660/500/.5	27.36	7.18	20.80	0.123±.002

Cross-Ply Composite Panels

Run CS275 [(0)$_3$/(90)$_5$/(0)$_3$]

An attempt was also made to mold a cross-ply composite panel using the same molding cycle with vacuum, except that a reduced pressure of 500 psi was applied in the final cure step. The cross-ply laminate was laid up in the fashion of (0)$_3$/(90)$_5$/(0)$_3$. The geometrical changes of the panel during the molding cycle are tabulated in Table 13. Here again, the molding process from cure step 1 to 3 was not interrupted and the molding stops were used. At the completion of cure step 3, the panel thickness (0.128") is near to that of the stops. This indicates that the laminate experienced zero consolidation pressure, and the C-scan as shown in Figure 17 is poor, as expected. At the completion of cure step 4, the thickness of the panel was found to be comparable, on a per ply basis (7.3 vs. 7.0 mils), to that of the unidirectional panel. The C-scan reveals that a well consolidated laminate results from the application of a moderate consolidation pressure of 500 psi.

Table 13. Geometrical changes of the cross-ply* 3.00"x3.00" composite laminate in Run CS275

Press Cycle	(°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	19.51	---	---	---
1	400/0/.5	---	---	---	---
2	500/0/.5	---	---	---	---
3	600/0/.5	16.27	3.25	16.63	0.128±.005
4	660/500/.5	16.18	3.33	17.06	0.080±.002

*11 ply [(0)$_3$/(90)$_5$/(0)$_3$]

Run CS276 [(0)$_3$/(90)$_5$/(0)$_3$]

Another cross-ply laminate panel with identical lay-up scheme as that of run CS275 [(0)$_3$/(90)$_5$/(0)$_3$] was compression molded by a conventional press (without vacuum) following the same molding cycle shown in Table 14. The first three cure steps were not

interrupted and the molding stops were used, which account for the panel thickness of 0.123" at the completion of cure step 3. A 500 psi consolidation pressure was applied for 0.5 hrs at 660°F in the final cure step. The resultant laminate is thinner but still comparable, on a per ply basis (6.7 vs. 7.0 mil), to that of unidirectional (Table 18 below) and cross-ply (Table 13) panels processed under vacuum. Such a reduction in thickness is also manifested in the increasing weight loss of the panel. Nevertheless, a well consolidated panel was achieved as shown by the C-scan in Figure 18. When compared to C-scans for identical panels processed under vacuum (Table 13 and Figure 17), it can be seen that use of vacuum during compression molding only marginally benefits the quality of laminate consolidation.

Table 14. Geometrical changes of the cross-ply* 3.00"x3.00" composite laminate in Run CS276

	Press Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	19.52	---	---	---
1	400/0/.5	---	---	---	---
2	500/0/.5	---	---	---	---
3	600/0/.5	15.66	3.86	19.76	0.123±.002
4	660/500/.5	15.15	4.01	20.56	0.073±.002

*11 ply $[(0)_3/(90)_5/(0)_3]$

Run CS1053 $(0/90)_5$

In the following experiments, compression moldings of isotropic and orthotropic composite panels were investigated.

Ten uni-directional prepreg plies were initially cut to a dimension measuring 2.75" by 2.75". These plies were then stacked in a female mold in a fashion of $(0/90)_5$ to make an orthotropic composite panel. With the male mold inserted, the whole assembly was molded in a vacuum press following the cure cycle listed in Table 15. During the B-stage period (cure steps 1 and 2) two molding stops with a total width measuring 0.25" were used. The

cure cycle was interrupted at the completion of cure step 2, and the geometrical changes of the panel were measured. Because of the use of the stops, the panel experienced practically zero pressure and was loosely compacted with ample volatiles escape paths provided for within the laminate. The thickness of the panel (0.120") is approximately the same as the height (0.125") of the stops. With the molding stops removed, the panel was remolded according to the cure step 3 at 660°F with 1,000 psi pressure applied throughout the cure step to complete the cure cycle. C-scan of the finished panel shown in Figure 19 reveals a composite panel with numerous discret minivoids.

The final in-plane dimensions of the panel were measured at 2.8125 inches by 2.8125 inches. Because of the orthotropic lay-up, the observed slight (0.0625") increased dimension in the lateral direction is likely attributed to the pattern skidding, instead of expansion deformation of the fiber-resin matrix, in response to the applied consolidation pressure. In other words, the 0, 90 orientation pattern among plies in this laminate locked the pattern position and prevented resin matrix shear which occurs along with fiber nesting during the molding of unidirectional ply laminate discussed above. Therefore, the apparent absence of the interply fiber-fiber nesting capabilities of the composite laminate with an orthotropic lay-up is the main reason for the poorer consolidation quality obtained with this panel.

Table 15. Geometrical changes of the cross-ply* 2.75" x 2.75" composite laminate in Run CS1053

	Molding Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	14.71	---	---	---
1	400/0/.5	---	---	---	---
2	600/0/.25	12.27	2.44	16.58	0.120±.002
3	660/1000/.5	12.27	2.44	16.58	0.068±.001

*10 ply orthotropic panel $[0/90]_5$

Despite the apparent differences between the lay-up sequence, the final thickness of the orthotropic panel (0.068") is comparable in the thickness to the unidirectional panels

(CS277-279) molded with provision for the interply fiber-fiber nesting to occur during the consolidation step.

Run CS1056 $(0/90)_5$

In order to investigate the pressure effect on the consolidation quality of the $(0/90)_5$ orthotropic composite panel, the following experiment was conducted.

Ten uni-directional prepreg plies were initially cut to a dimension measuring 2.75" by 2.75". Except for the consolidation pressure, the cure cycle used was identical to that of run CS1053. The results are tabulated in Table 16. The final in-plane dimensions of the panel were measured at 2.78 inch by 2.78 inches. Because of the orthotropic lay-up, the observed slight (0.03125") increased dimension in the lateral direction is again likely attributed to the pattern skidding, instead of expansion deformation of the fiber-resin matrix, in response to the applied consolidation pressure. At the completion of cure step 3, the panel is noted to be 13% thicker than that of run CS1053 (.077 vs. .068 inches). The reason is unknown.

Table 16. Geometrical changes of the cross-ply* 2.75" x 2.75" composite laminate in Run CS1056

	Molding Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	15.872	---	---	---
1	400/0/.5	---	---	---	---
2	600/0/.25	12.978	2.90	18.27	0.120±.002
3	660/500/.5	12.968	2.91	18.33	0.076±.001
4	680/2000/.5	12.963	2.91	18.33	0.073±.001

*10 ply orthotropic panel $[0/90]_5$

C-scans of the panel taken at the completion of cure steps 3 and 4 are shown in Figure 20, together with that from the panel CS1053. Despite the use of higher pressure,

the C-scan of step 4 (2,000 psi at 680°F) is noted to be marginally worse than that of CS1053 (1,000 psi at 660°F). This result can be attributed to the premature blocking of the volatiles ventilation paths within the laminate by the preceding cure step (500 psi at 660°F). Nevertheless, it is noticeable that enhanced consolidation quality can be obtained with increasing consolidation pressure for the orthotropic panel investigated.

Run CS1054 $[(0)_2/(+45)_2/(-45)_2/(90)_2]_2$

Sixteen unidirectional prepreg plies were initially cut to a dimension measuring 2.75" by 2.75". These plies were then stacked in a female mold in a fashion of $[(0)_2/(+45)_2/(-45)_2/(90)_2]_2$ to make a quasi-isotropic composite panel. With the male mold inserted, the whole assembly was molded in a vacuum press following the cure cycle listed in Table 17. During the B-stage period (cure steps 1 and 2) two internal molding stops were used. The cure cycle was interrupted at the completion of cure step 2, and the geometrical changes of the panel were measured. Because of the use of stops, the panel experienced practically zero pressure and was loosely compacted with ample volatiles escape paths created within the laminate. The thickness of the panel (0.186") is approximately the same as the height of the internal stops. With the molding stops removed, the panel was remolded according to the cure step 3 at 660°F with 1,000 psi pressure applied throughout the cure step. A C-scan of the finished panel shown in Figure 21 reveals a consolidated composite panel exceeding 90% void free overall.

Table 17. Geometrical changes of the cross-ply* 2.75"x 2.75" composite laminate in Run CS1054

	Molding Cycle (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	25.60	---	---	---
1	400/0/.25	---	---	---	---
2	600/0/.25	21.02	4.58	17.90	0.186±.002
3	660/1000/.5	21.00	4.60	17.92	0.106±.001

*16 ply quasi-isotropic panel $[(0)_2/(+45)_2/(-45)_2/(90)_2]_2$

The double ply lay-ups of $[(0)_2/(+45)_2/(-45)_2/(90)_2]_2$ in each orientation was designed to provide the desired degree of interply fiber-fiber nesting capabilities for the composite laminate during the consolidation stage. Final in-plane dimensions of the panel were measured at 3.00" in the longitudinal fiber direction without stops by 2.78" in the in-plane lateral direction. The final lateral dimension is close to the original value of 2.75" indicating that there was only little side way movement during the final consolidation step. Considering the quasi-isotropic layup scheme, the observed increases in the longitudinal dimension are likely attributed to pattern skidding occuring during the B-stage period, when there were no stops present in that direction. When comparing to panel CS1053, it is clear that such a lay-up scheme *does offer* some degree of the desired nesting capabilities among interply prepreg layers, which translates into a panel with improved consolidation quality under comparable molding conditions.

The final thickness of the isotropic panel is noted to be thinner than the thickness of the orthotropic panel (CS1053), i.e., 0.066" vs. 0.068" on a per ply basis; another indication of the occurrence of interply fiber-fiber nesting during the consolidation step.

6" x 6" Unidirectional Composite Panel

Run CS272

Using this newly developed compression molding cycle for the CS batch of $PISO_2$ prepreg, an attempt was made to mold a 6.00" x 6.00" composite panel. The geometrical changes of the laminate during the cure cycle and the C-scans of the panel are shown in Table 18 and Figure 22, respectively. The weight loss and the thickness reduction scheme as the cure steps progressed are seen comparable to those of 3.00" x 3.00" panels. At the completion of cure step 4, a well consolidated composite panel with superior C-scan quality is again reached as shown in Figure 22.

Table 18. Geometrical changes of the 10 ply unidirectional 6.00"x6.00" composite laminate in Run CS272

Press Cycle	(°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	76.61	---	---	---
1	400/0/.5	---	---	---	---
2	500/0/.5	---	---	---	---
3	600/0/.5	62.20	14.41	18.81	0.122±.005
4	660/2000/.5	62.13	14.48	18.90	0.070±.002

CONSOLIDATION THEORY

A comparison between the conventional and the improved molding technologies resulting from this investigation is shown schematically in Figure 23.

During the B-stage period of the cure cycle, molding stops are used in the improved molding technique (Figure 23a). The composite prepreg layers are cut to fit in the remaining space of the mold cavity. The male mold is closed to the stops such that the stacked prepreg layers experience practically zero pressure. This arrangement results in a loosely packed laminate structure which offers abundant volatile escape paths for the reaction by-products generated during the B-stage curing step.

On the other hand, no molding stops are used in the conventional molding technique (Figure 23b). The composite prepreg layers are cut to fit the entire space of the mold cavity. The male mold is rested on top of the stacked prepreg layers. This arrangement results in a denser laminate structure, i. e., both of the intraply (lateral direction) and interply (between prepreg layers in the vertical or z direction) fiber-fiber intimate contact exists (see Figure 23b). Volatiles escape paths for the reaction by-products are therefore severely reduced, which makes the consolidation of a void free composite laminate more difficult.

It is clear that application of either partial or full consolidation pressure at this stage following the conventional molding techniques is not advisable. It has been observed that in some situations where lower B-stage temperatures are employed, the degree of imidization is low at this stage of the cure cycle. Any consolidating pressure applied will not only block the volatiles escape paths within the fibers, but also squeeze out too much resin. The significance of critical timing for the pressure application during a cure cycle can be envisioned from this illustration of the figure.

During the final consolidation stage of the cure cycle, molding stops are removed in the improved molding technique (Figure 23c). The removal of the molding stops creates an excess volume within the cavity of the mold, which allows possible side way movement (lateral direction) of the fiber/resin matrix in response to the applied consolidating pressure. Such a movement results in a rearrangement of the laminate structure. A lessor degree of applied pressure is absorbed by the otherwise intimate interply fiber-fiber contact.

Consequently, better consolidation quality is achieved, minimizing residual void content within the laminate.

On the other hand, the intimate interply fiber-fiber contact prevails in the conventional molding process (Figure 23d). The consolidation pressure is largely absorbed by such fiber-fiber contact, and a composite laminate with poor consolidation quality can be expected.

Composite laminates with the same resin content are expected from both molding techniques. However, it is conceivable that thicker laminates are likely by the conventional molding process when using the same numbers of prepreg plies and the same molding pressure.

The final consolidation quality of the composite laminate molded by the improved technique depends heavily on the resin matrix viscosity after B-stage. The B-stage conditions are critical for the following reasons: a B-stage at elevated temperatures will advance the resin reaction to an extent that the resin matrix becomes highly viscous. Under such a situation, suppression of voids becomes difficult and can not be achieved without unreasonably high consolidation pressure. On the other hand, an inadequate B-stage will result in a resin matrix with a low level of curing advancement. Although the low viscosity associated with the lower degree of resin cure advancement is beneficial for required resin flow properties, the volatile by-products generated by the excessive reactions occuring at the elevated temperature are detrimental to the consolidation quality, especially during the final consolidation step where intimate interply and intraply fiber-fiber contacts prevail.

COMPOSITE MECHANICAL PROPERTIES

The temperature and pressure cycles developed in this investigation have been shown to consistently produce composite panels from the AS-4/$PISO_2$ prepreg material with exceptional C-scan quality. However, C-scan is known only as a useful first line quality control tool. A composite laminate with good C-scan does not necessarily guarantee a acceptable level of mechanical properties. Because of the much lower consolidation pressure used in this work when compared to conventional molding processes, it is imperative to evaluate the mechanical properties of each of these consolidated composite panels.

Short Beam Shear (SBS) Strength

A 20 ply unidirectional 3.00" x 3.00" panel has been molded successfully in Run CS1051 (Table 12). The C-scan of the panel showed an outstanding consolidation quality (see Figure 16). Thickness of the consolidated panel was 0.123 inches. Twenty one short beam shear (SBS) samples were prepared and tested at room temperature. A mean value of 11.81 ksi was obtained. The standard deviation of 0.59 ksi accounts for approximately 5% of the mean value. SBS strength between 11 and 12 ksi at room temperature was reported earlier by St. Clair and Yamaki [15]. These two sets of data are compared in Figure 24 for the same unidirectional fiber-resin composite laminate. While the molding conditions were not reported in [15], it is seen that a comparable level of SBS strength was achieved in this investigation by panel (CS1051) molded under moderate conditions (660°F and 500 psi).

Run CS1055

In order to investigate the effect of post curing on the mechanical properties of the $PISO_2$ composite laminate, additional SBS specimens were prepared. A 20 ply unidirectional 3.00" x 3.00" composite panel was molded for the experiment. The initial lay-up of the prepreg plies in the mold and the subsequent cure cycle, except for the additional post curing of the panel, was identical to that used in run CS1051. The post curing schedule tabulated in Table 19 consists of annealing the panel for 2 hours each at 550 and 600°F. C-scans of the post cured panel shown in Figure 25 reveal a composite panel comparable to panel CS1051 (see Figure 16).

Similar to panel CS1051, panel CS1055 possesses in-plane dimensions measuring a full 3.00" by 3.00". This in-plane dimensional change indicates a lateral deformation occuring in the laminate structure in response to the consolidating pressure applied at cure step 3. Such an expanded deformation in the lateral direction allows interply fiber-fiber nesting to occur. It is noted, however, that in spite of the employment of an identical cure cycle, this panel is slightly thicker than the panel CS1051, i.e., 0.063" vs. 0.0615" on a per ply basis. The 2.4% difference in thickness indicates that a lesser degree of interply fiber-fiber nesting occured in panel CS1055. However, the effect on the mechanical properties is not expected to be significant.

Table 21. Geometrical changes of the 20 ply unidirectional 3.00"x3.00" composite laminate in Run CS1055

	Molding Cycle* (°F/psi/hr)	Wt. (g)	Wt. Loss (g)	Wt. Loss (%)	Thickness (in)
	Ambient	34.24	---	---	---
1	400/0/.5	---	---	---	---
2	600/0/.25	28.06	6.17	18.0	0.303±.002
3	660/500/.5	28.04	6.19	18.1	0.130±.002
4	550/500/2	28.04	6.19	18.1	0.127±.002
5	600/500/2	28.04	6.19	18.1	0.126±.001

*Identical to run CS1051 except for the additional post curing schedule at 550 and 600°F.

Twenty one SBS samples were prepared and tested at four temperatures, i.e., 27°, 93°, 150° and 177°C. Results of these tests are shown in Figure 25. Also shown in the Figure are data reported by St. Clair and Yamaki [15] on the same composite system. The multi-temperature test was conducted using Instron test equipment. It is noted from the Figure that, a comparable overall level of SBS strength between these two sets of test results was achieved at the elevated temperatures. A significant difference, i.e., 9.5 vs. 11.5 ksi, at room temperature is evident in the test results. A few samples from panel CS1055 were also prepared and tested at room temperature on United test equipment. An

SBS strength of 11.46 ksi is measured. This value is closer to that reported by St. Clair and Yamaki and is also included in Figure 25.

It is concluded from these studies that:

i) A difference of 20% in the SBS strength was observed between different testing machine.

ii) Unlike the AS-4/LaRC-TPI 1500 (HFG) composite system [14], post cure annealing at moderate temperatures (550 and 600°F for 2 hours), generated no appreciable effect on the SBS strength of the AS-4/$PISO_2$ composite system.

Flexural Strength

Flexural strength specimens were prepared from panel CS272. Measurements were performed at room temperature. The results are shown in Figure 26. Also included in the Figure are the data reported by St. Clair and Yamaki on the same composite system. As noted from the figure, a comparable level of flexural strength, approximately 195 Ksi (standard deviation σ = 23.8 Ksi) at room temperature, was recorded for each set of specimens.

CONCLUSIONS

The following conclusions can be drawn from this investigation:

1. An improved compression molding technique was developed for the AS-4/polyimidesulfone prepreg system. This prepreg system can be consistently molded into void-free unidirectional or cross-ply laminate panels with superior mechanical properties. The moderate molding conditions, i.e., 660°F and 500 psi, used in this improved technique produced superior C-scans when compared with conventionally molded composite panels.

2. Unidirectional composite panels up to 6.00" x 6.00" x 0.070" have been molded by this technique. Unmodified aromatic polyimide material such as the polyimidesulfone studied here have a reputation for poor processability. The procedures described in this report provide a repeatable method for producing acceptable $PISO_2$ composite panels. The key components of the improved technique are providing multiple volatile escape paths within the laminate initially and providing for lateral movement of the B-staged composite during the final consolidation step in the cure cycle. The lateral movement allows interply fiber-fiber nesting to occur during the final consolidation, effectively minimizing the absorption of the applied consolidation pressure by the otherwise intimate interply fiber-fiber contact within the laminate. Such a movement can not occur in the conventional molding process using fully sized prepreg patterns. The ply fibers contact each other and without lateral movement, compaction at a given pressure ceases and leaves interstice voids resulting in a poor quality composite panel.

3. Employment of vacuum during the molding process was found to expedite the escape of volatile by-products. However, only marginal improvement of the C-scans were realized for the consolidated panels.

4. Timing of the pressure cycle application is extremely critical. Early application of pressure during the cure cycle can be harmful to the final consolidation process by effectively blocking the volatile escape paths within the laminate, thus trapping voids.

5. Prepreg quality is very critical to the molding process and in the development of a viable molding cycle. Prepreg with large variations in resin distribution can cause the development of a totally erroneous cure cycle. Prepreg with uniformly distributed resin matrix and tightly controlled limits lends itself to quality molding with reproducible results.

6. The B-stage molding step of the improved molding technique can be accomplished in a press or in a conventional forced air circulating oven. The successful extension of the process to include the use of an oven has greatly enhanced the usefulness of this molding cycle. The obvious benefits include the reduction of the press processing cycle time and the rapid preparation of B-staged $PISO_2$ laminates, ready for the final press molding cycle.

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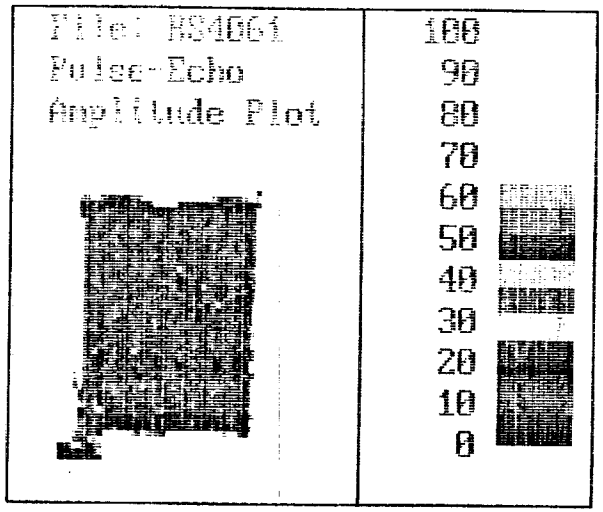
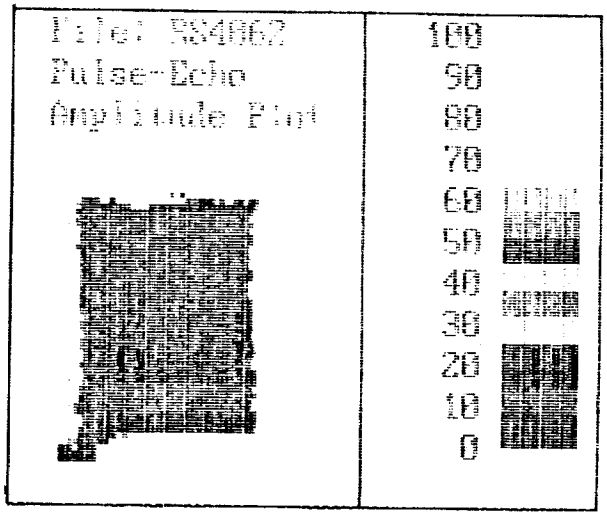
LIST OF FIGURES

Vacuum Press Molding of Polyimidesulfone Composite Laminate

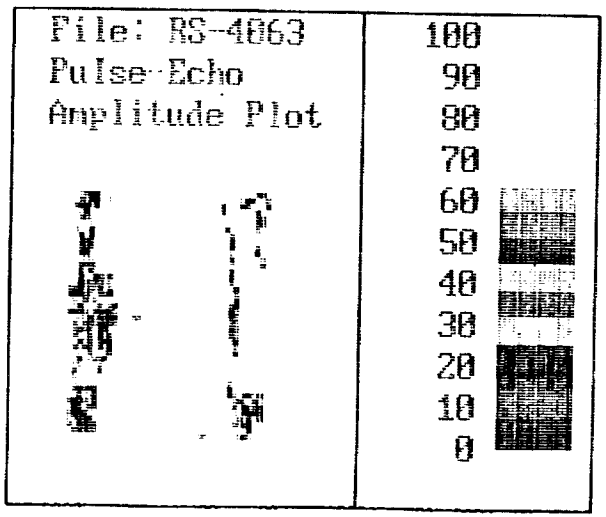
Curing Step 1 - 400°F/0psi/.5hrs



Curing Step 2 - 500°F/0psi/.5hrs



Curing Step 3 - 600°F/400psi/.5hrs



Curing Step 4 - 660°F/2000psi/.5hrs

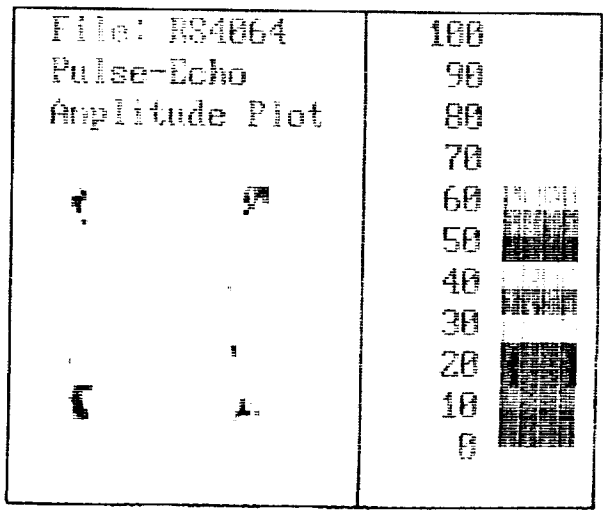


Figure 1. C-scans of panel 406 at various molding steps of the laminate consolidation cycle.

Vacuum Press Molding of Polyimidesulfone Composite Laminate

Run #406-1 8x
400°F/0psi/0.5hrs.
End View
(Fiber Direction)

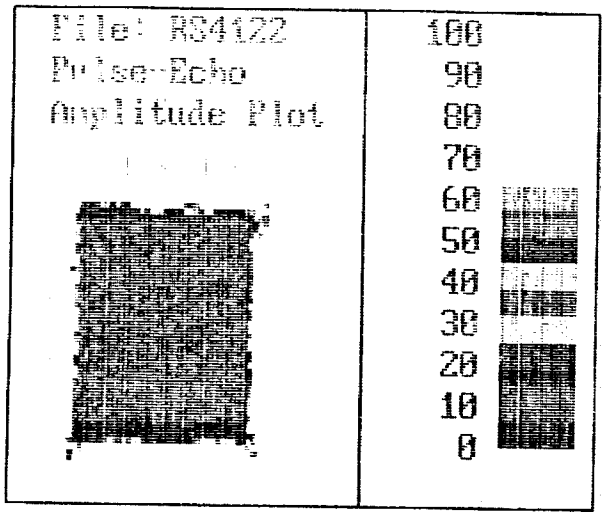
Run #406-1 8x
400°F/0psi/0.5hrs.
Side View
(Lateral Direction)

Run #406-3 8x
600°F/400psi/0.5hrs.
End View
(Fiber Direction)

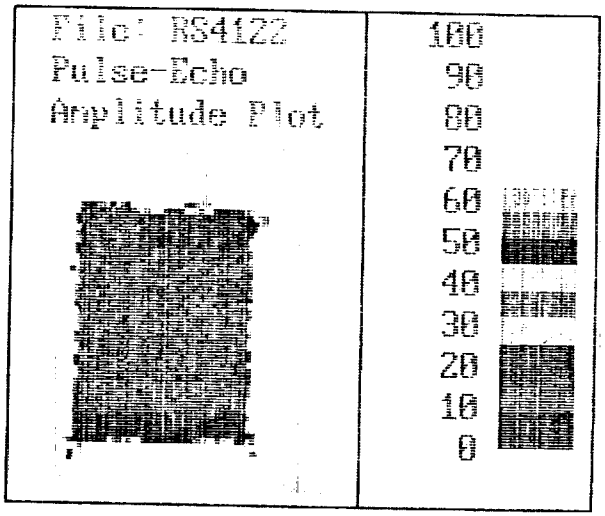
Figure 2. Optical micrographs for the edges of panel 406 at various molding steps.

Conventional Press Molding of Polyimidesulfone Composite Laminate

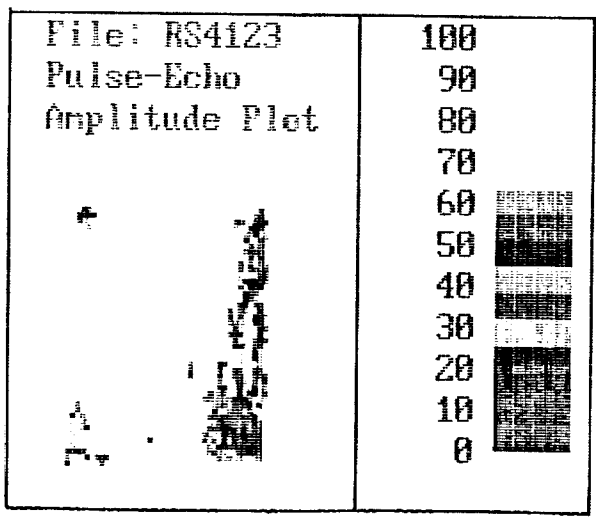
Cure Step 1 - 400°F/0psi/.5hrs



Cure Step 2 - 500°F/0psi/.5hrs



Cure Step 3 - 600°F/400psi/.5hrs



Cure Step 4 - 660°F/2000psi/.5hrs

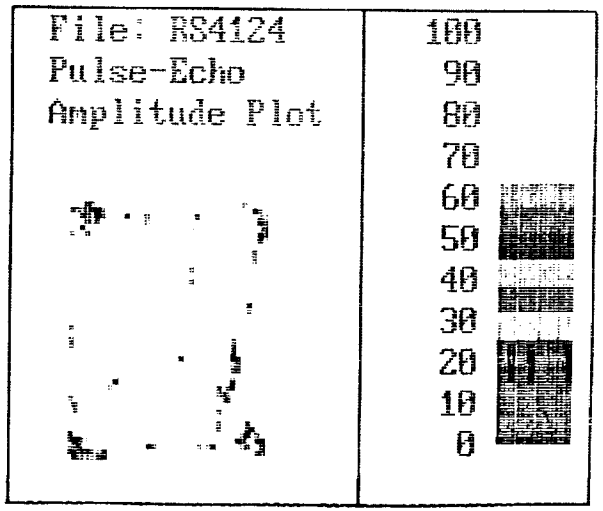
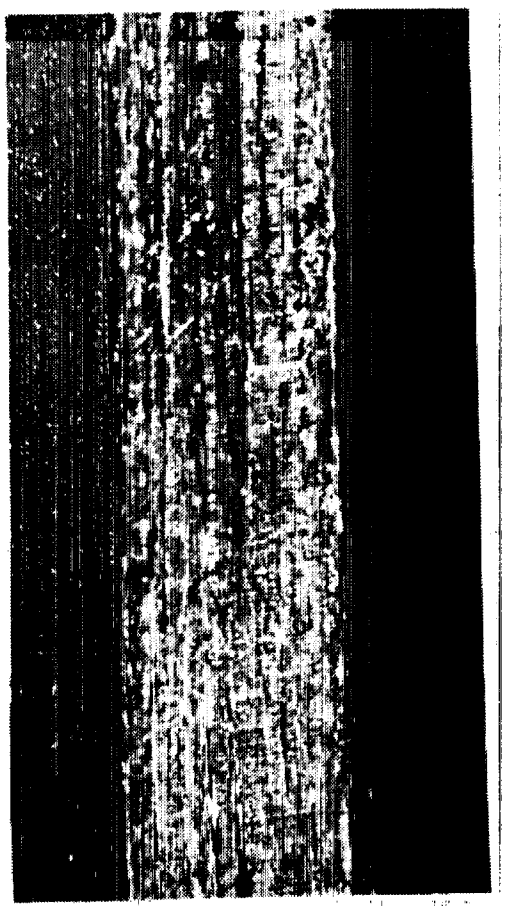


Figure 3. C-scans of panel 412 at various molding steps of the laminate consolidation cycle.

Run #412-1 8x
400°F/0psi/0.5hrs.
End View
(Fiber Direction)



Run #412-1 8x
400°F/0psi/0.5hrs.
Side View
(Lateral Direction)

Run #412-3 8x
600°F/400psi/0.5hrs.
End View
(Fiber Direction)

Figure 4. Optical micrographs for the edges of panel 412 at various molding steps.

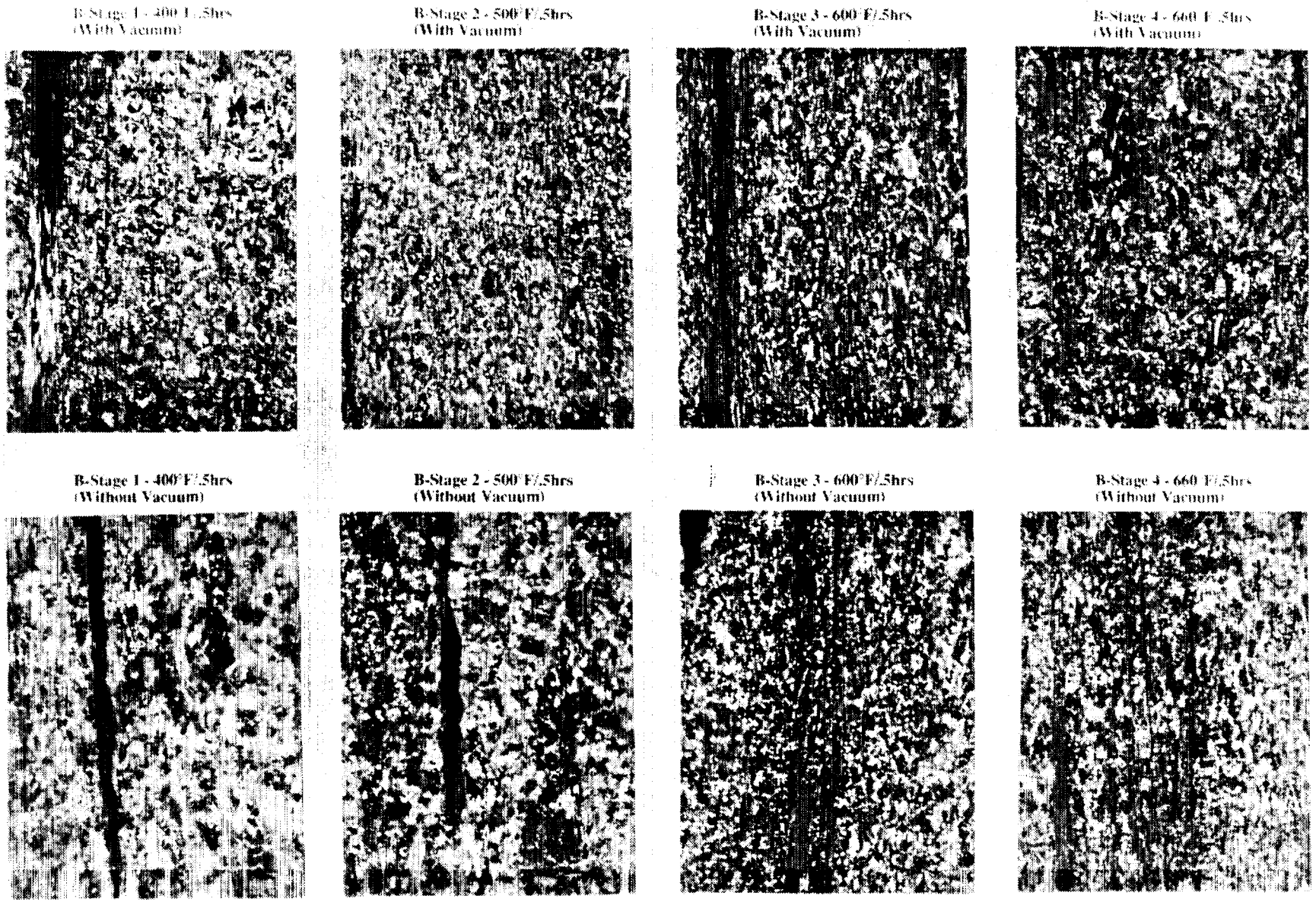
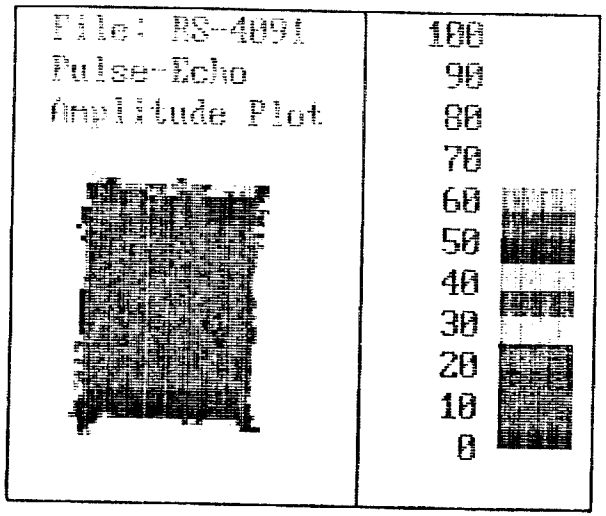


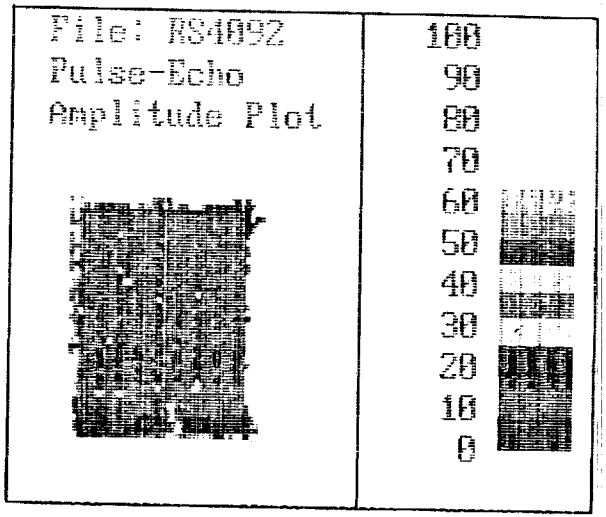
Figure 5. Comparisons of the surface characteristics of the B-staged prepreg sheets with and without employing the vacuum.

Vacuum Press Molding of Polyimidesulfone Composite Laminate

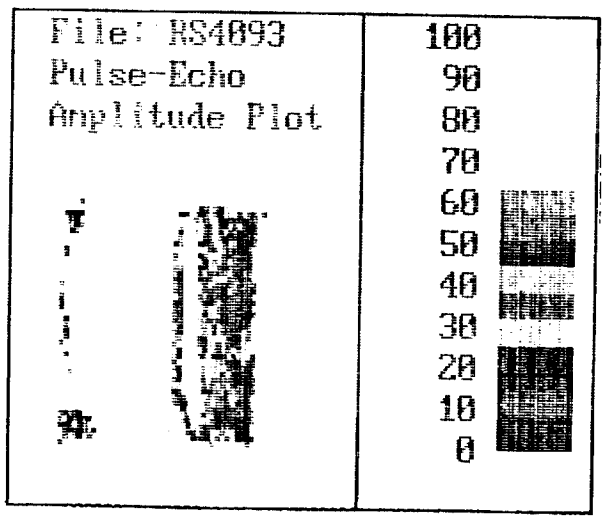
Curing Step 1 - 400°F/0psi/.5hrs



Curing Step 2 - 500°F/100psi/.5hrs



Curing Step 3 - 600°F/400psi/.5hrs



Curing Step 4 - 660°F/2000psi/.5hrs

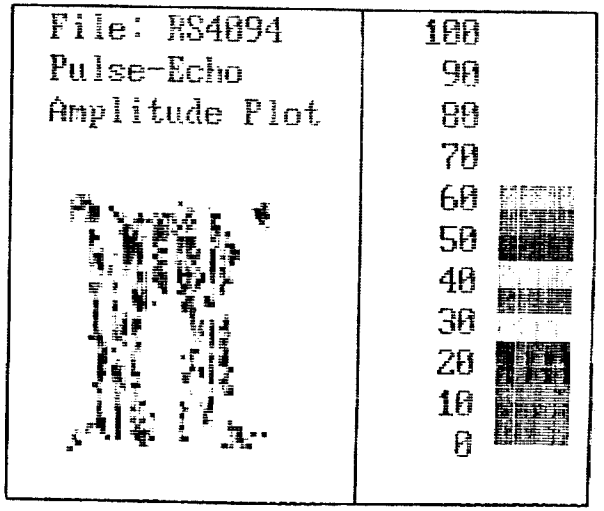
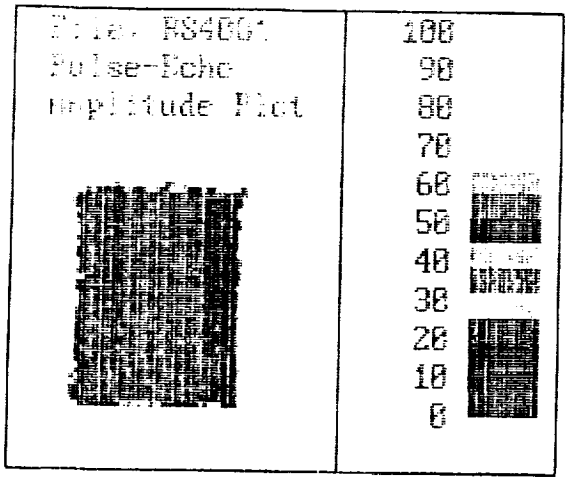


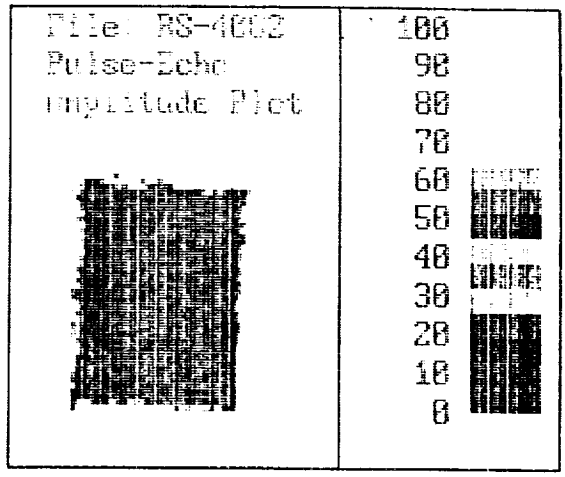
Figure 6. C-scans of panel 409 at various molding steps of the laminate consolidation cycle.

Vacuum Press Molding of Polyimidesulfone Composite Laminate

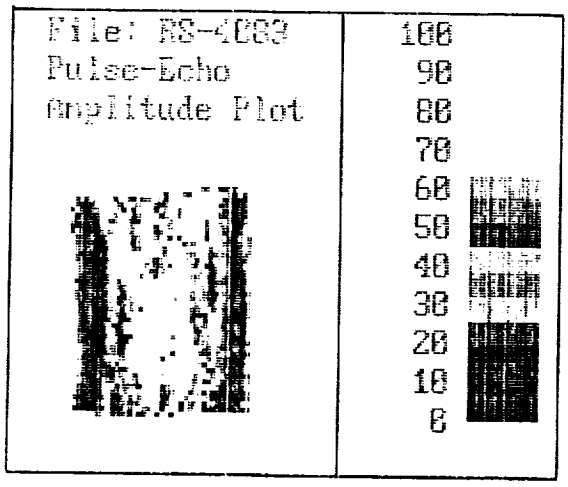
Curing Step 1 - 400°F/0psi/.5hrs



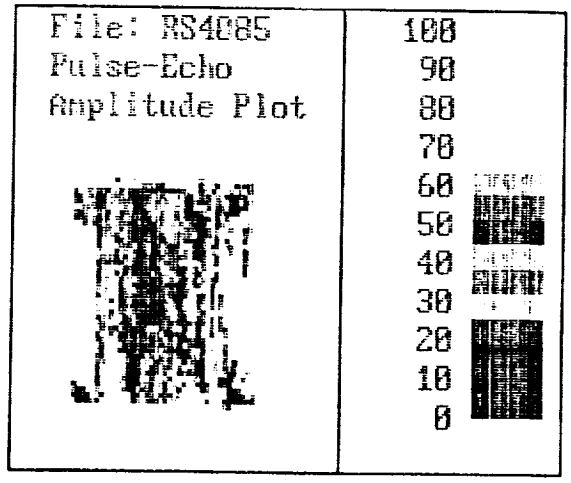
Curing Step 2 - 500°F/0psi/.5hrs



Curing Step 3 - 600°F/200psi/.5hrs



Curing Step 4 - 660°F/1000psi/1.5hrs



Curing Step 5 - 660°F/2000psi/.5hrs

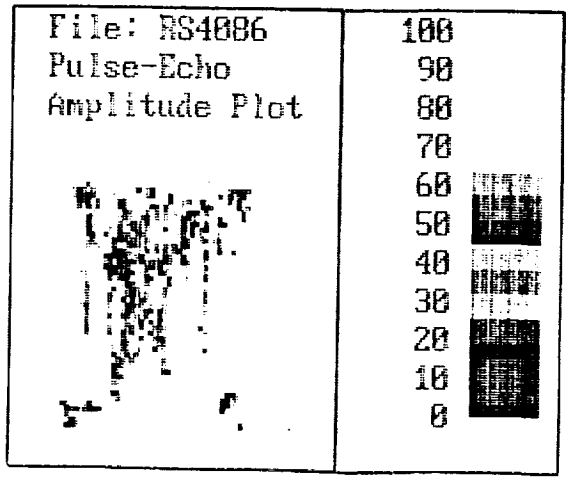
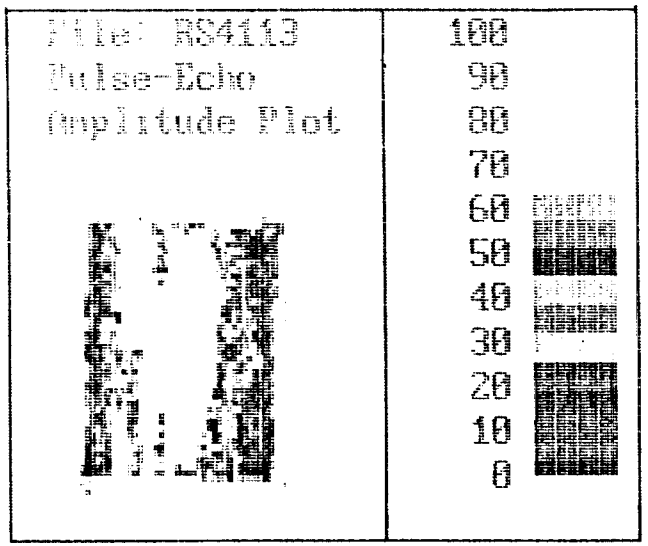


Figure 7. C-scans of panel 408 at various molding steps of the laminate consolidation cycle.

Vacuum Press Molding of Polyimidesulfone Composite Laminate

Cure Step 3 - 600°F/200psi/.5hrs



Cure Step 4 - 660°F/1500psi/.5hrs

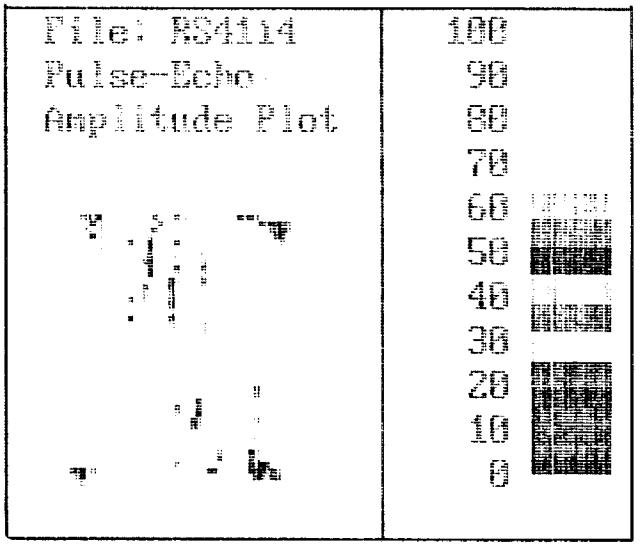


Figure 8. C-scans of panel 411 at various molding steps of the laminate consolidation cycle.

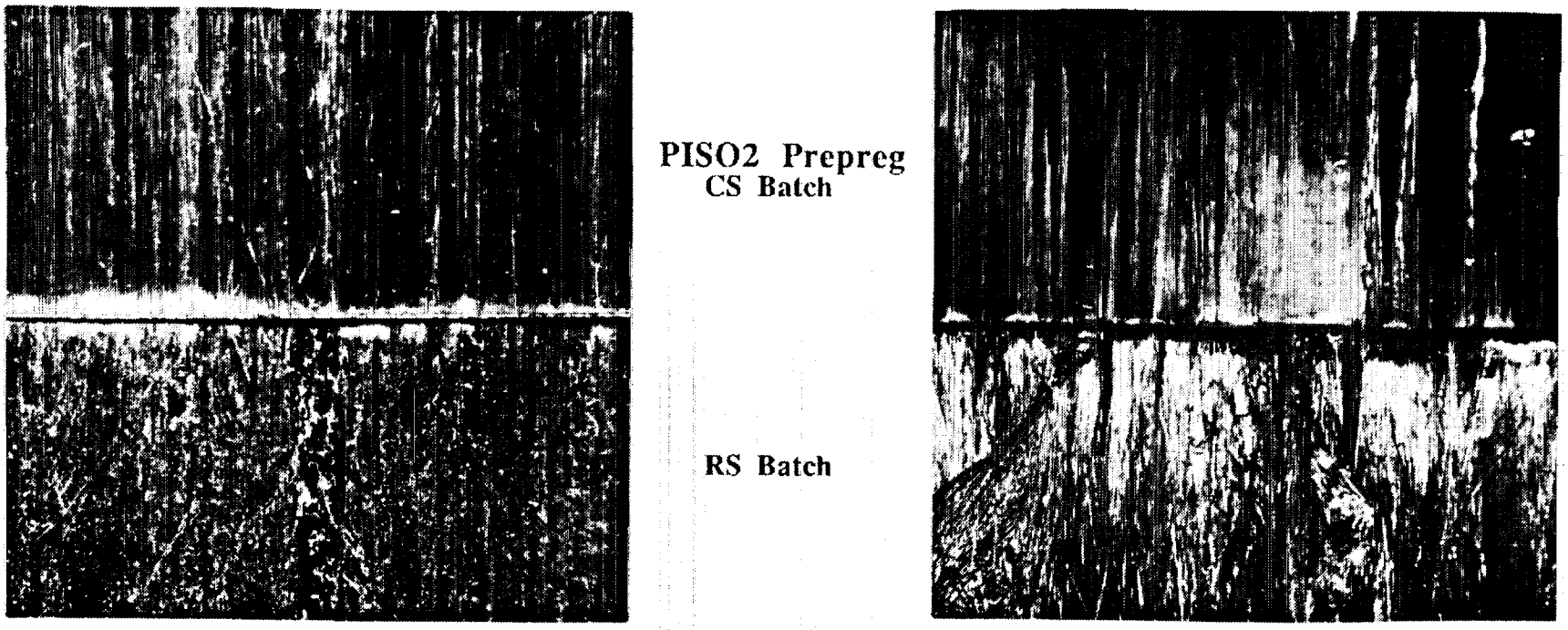
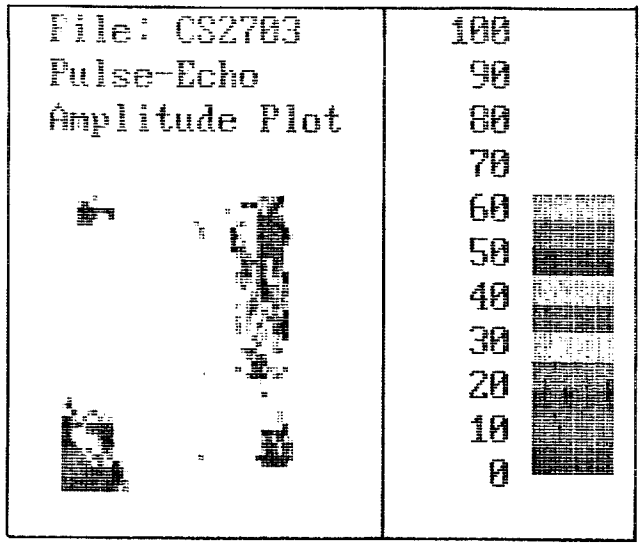


Figure 9. Comparisons of the surface characteristics of the two batches as-made AS-4/ polyimidesulfone prepreg.

Vacuum Press Molding of Polyimidesulfone Composite Laminate

Cure Step 3 - 600°F/400psi/.5hrs



Cure Step 4 - 660°F/2000psi/.5hrs

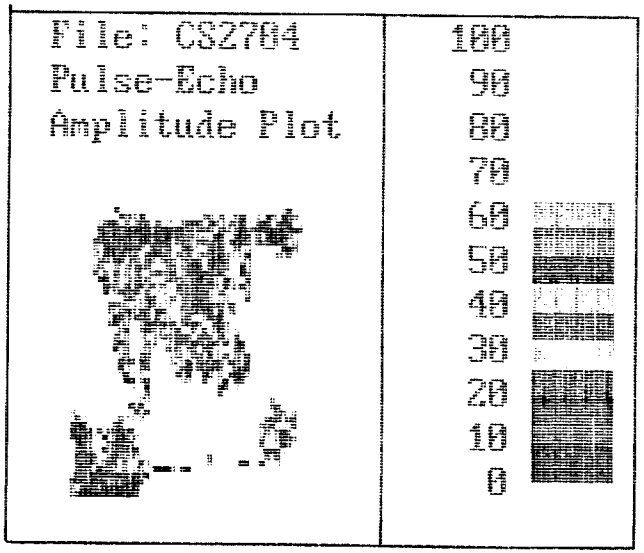
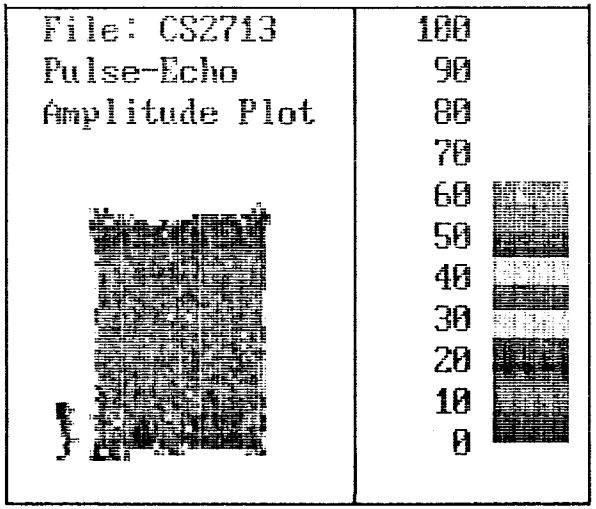


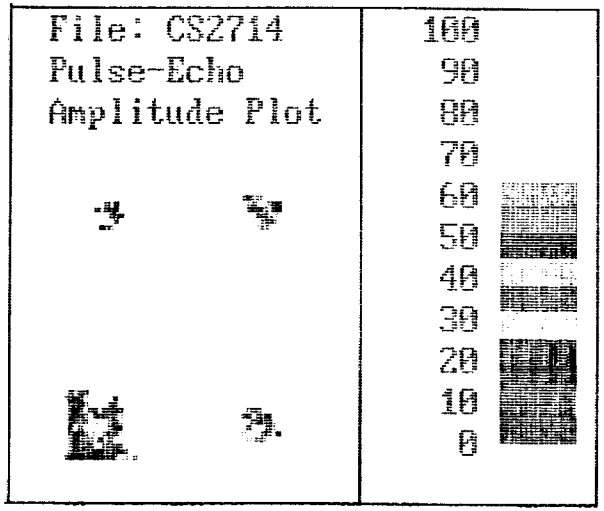
Figure 10. C-scans of panel CS270 at various molding steps of the laminate consolidation cycle.

Vacuum Press Molding of Polyimidesulfone Composite Laminate

Cure Step 3 - 600°F/0psi/.5hrs



Cure Step 4 - 660°F/2000psi/.5hrs



Cure Step 5 - 695°F/500psi/.5hrs

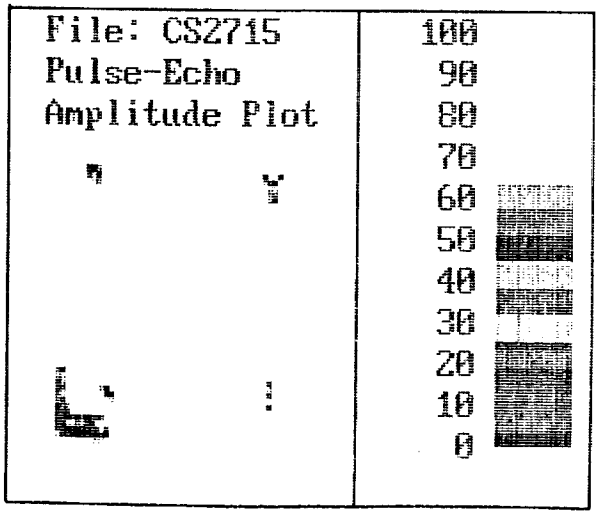


Figure 11. C-scans of panel CS271 at various molding steps of the laminate consolidation cycle.

Cure Step 4 - 660°F/500psi/.5hrs

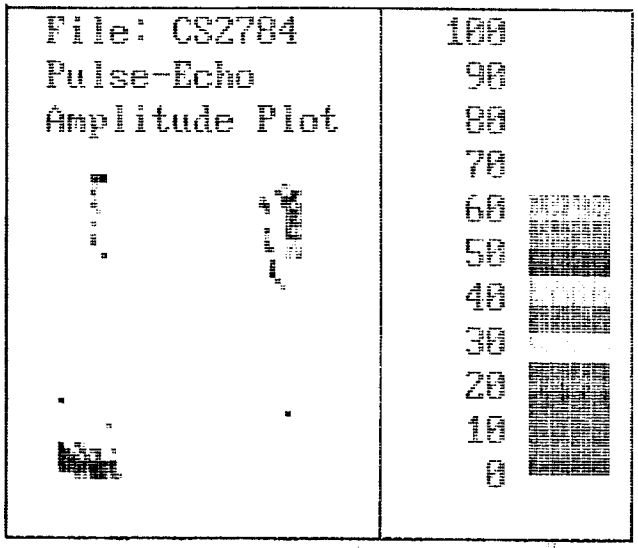


Figure 12. C-scan of the consolidated panel CS278.

Cure Step 4 - 660°F/500psi/.5hrs

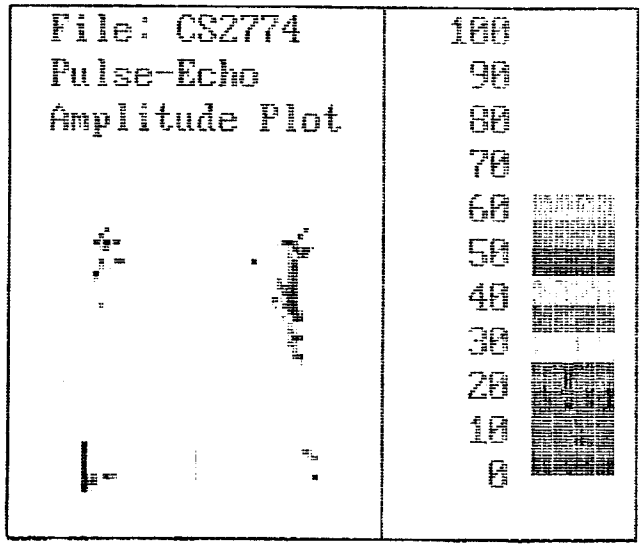
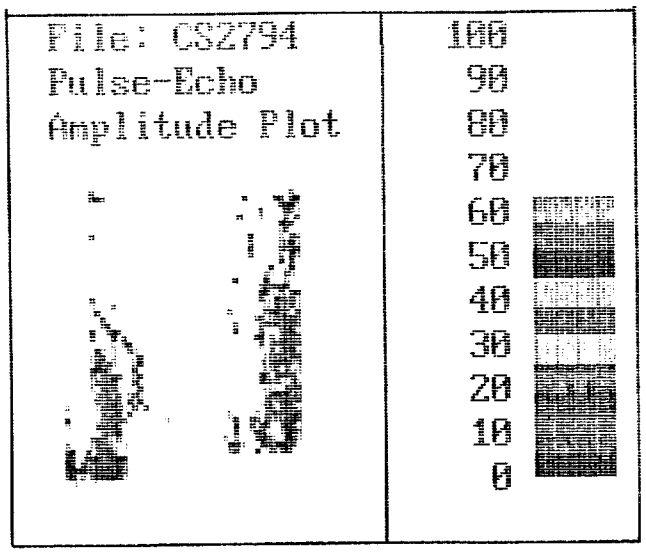


Figure 13. C-scan of the consolidated panel CS277.

Vacuum Press Molding of Polyimidesulfone Composite Laminate

Cure Step 4 - 660°F/250psi/.5hrs



Cure Step 5 - 675°F/400psi/.5hrs

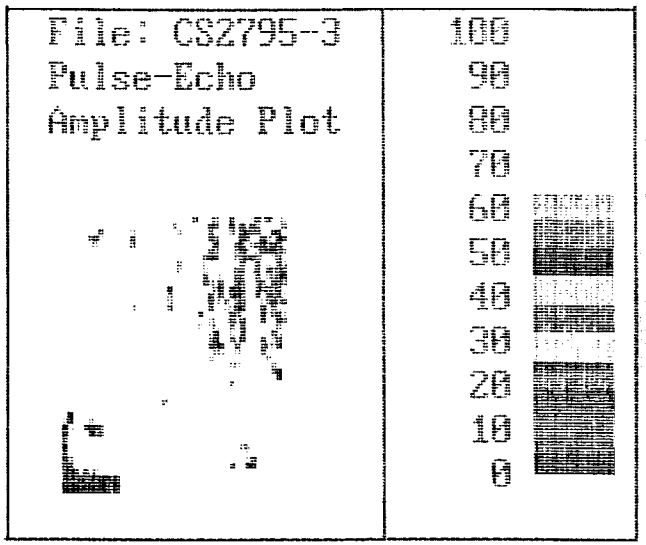
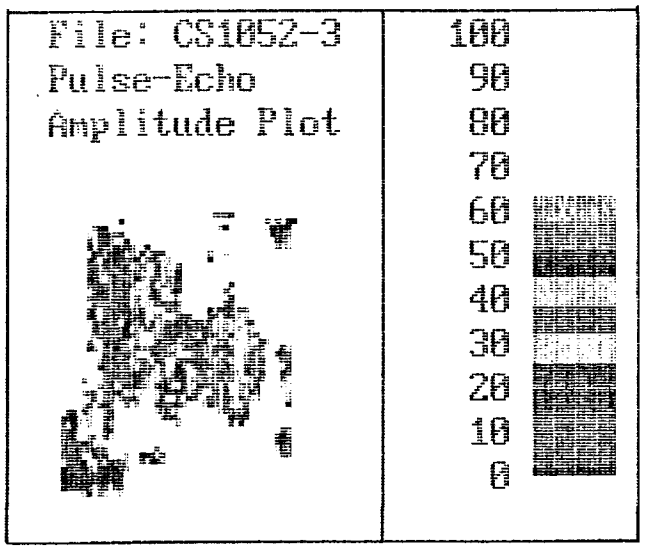


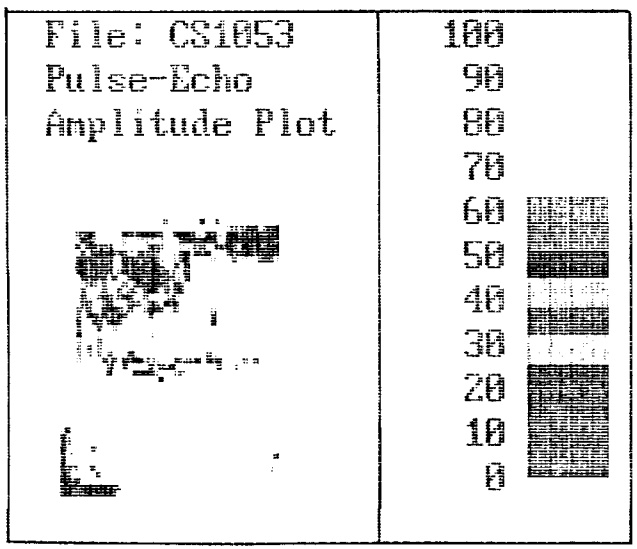
Figure 14. C-scans of panel CS279 at various molding steps of the laminate consolidation cycle.

Vacuum Press Molding of Polyimidesulfone Composite Laminate

Cure Step 1 - 400, 500°F/200psi/.5hrs



Cure Step 2 - 600°F/500psi/.5hrs



Cure Step 3 - 660°F/2000psi/.5hrs

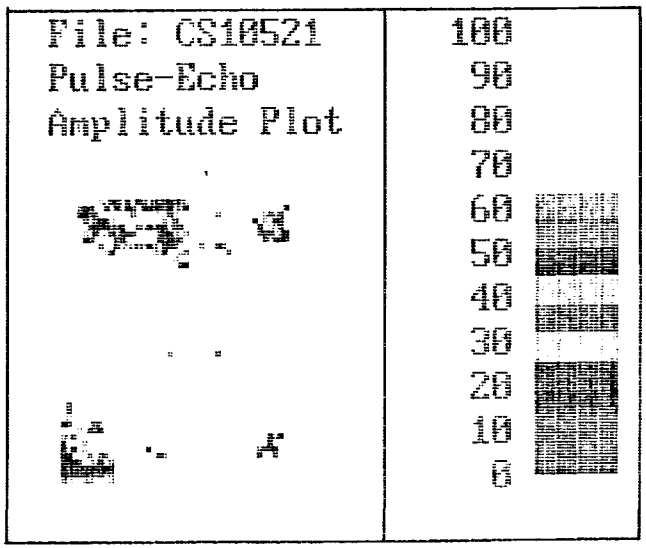


Figure 15. C-scans of panel CS1052 at various molding steps of the laminate consolidation cycle.

Cure Step 4 - 660°F/500psi/.5hrs

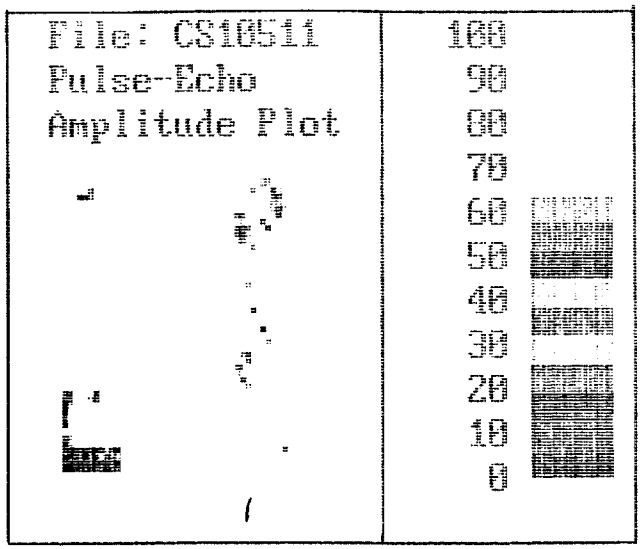


Figure 16. C-scan of the consolidated panel CS1051.

Cure Step 4 - 660°F/500psi/.5hrs
$[(0)_3\ (90)_5\ (0)_3]$

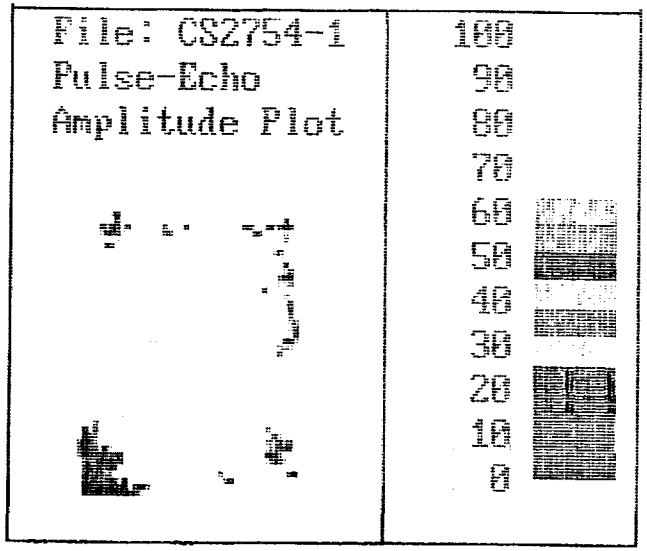


Figure 17. C-scan of the consolidated panel CS275.

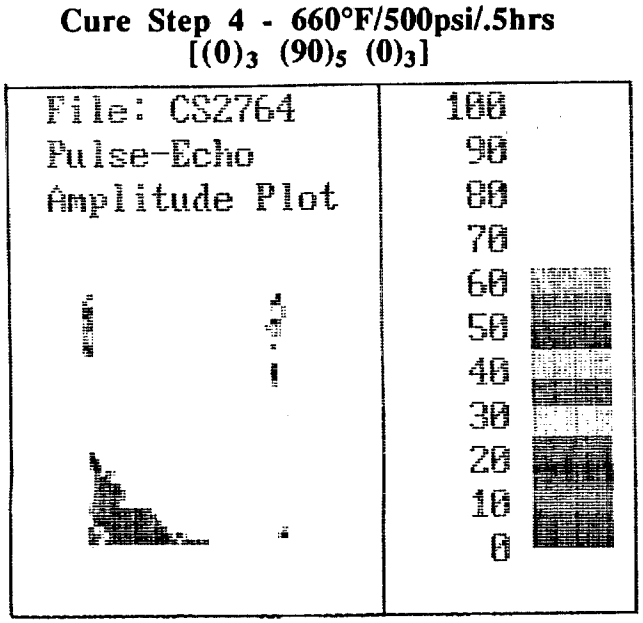


Figure 18. C-scan of the consolidated panel CS276.

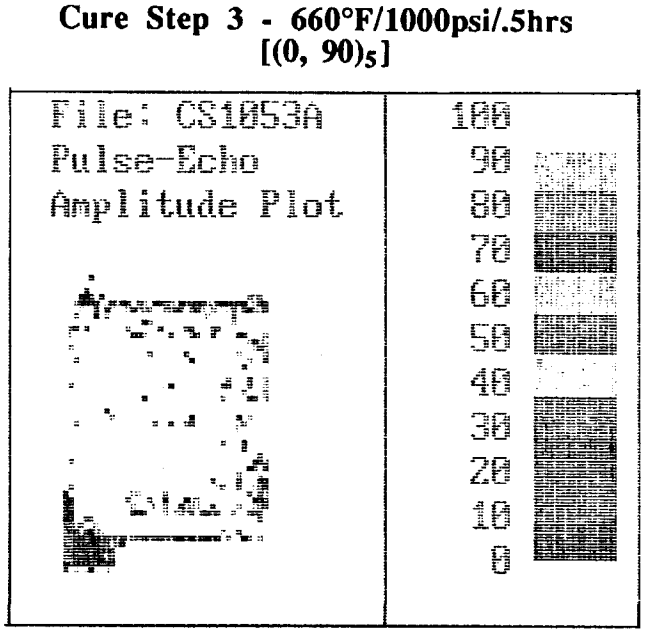
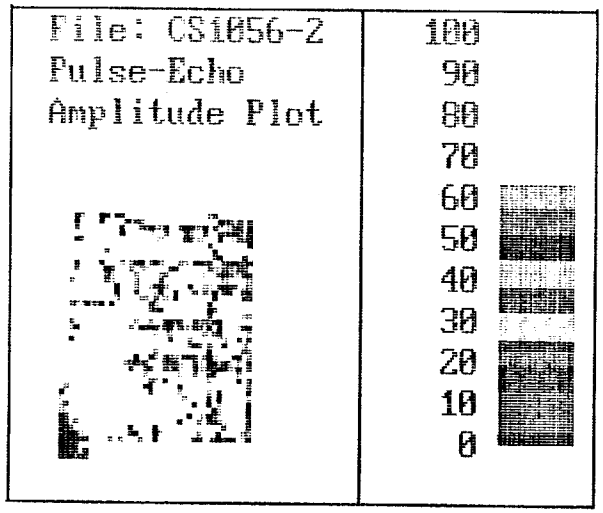


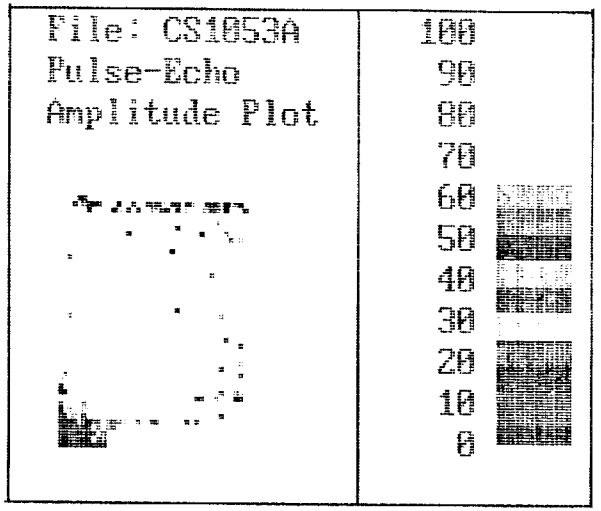
Figure 19. C-scan of the consolidated panel CS1053.

Vacuum Press Molding of Polyimidesulfone Composite Laminate $[0, 90]_5$

CS1056(3) - 660°F/500psi/.5hrs



CS1053(3) - 660°F/1000psi/.5hrs



CS1056(4) - 680°F/2000psi/.5hrs

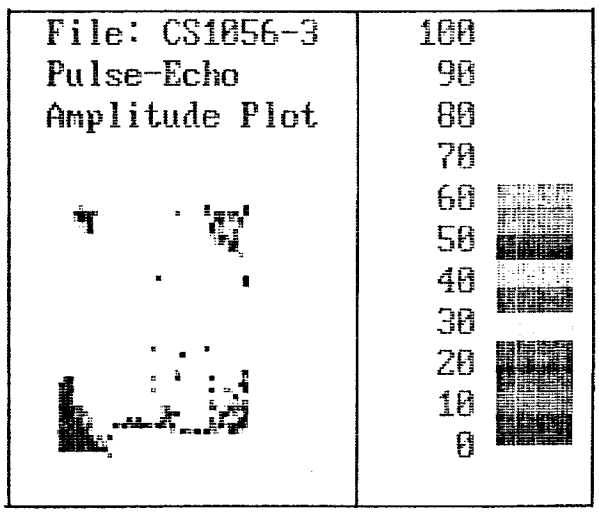


Figure 20. C-scans of panel CS1056 at various molding steps of the laminate consolidation cycle. (C-scan of panel CS1053 is also included for comparison).

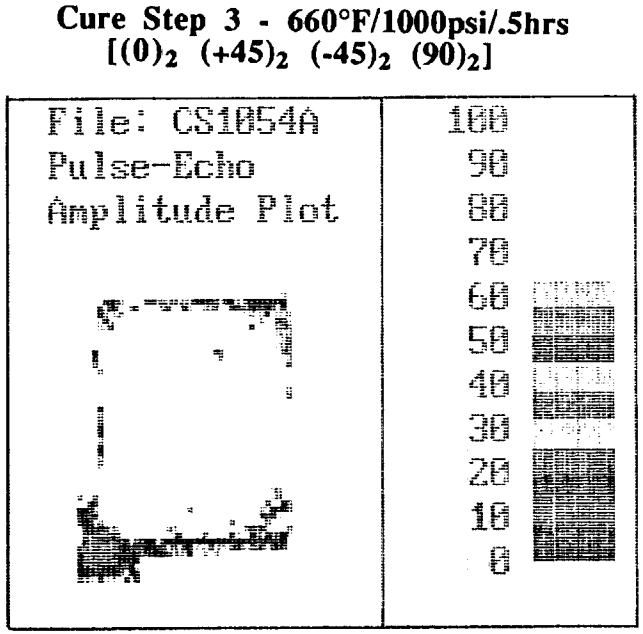


Figure 21. C-scan of the consolidated panel CS1054.

Cure Step 4 - 660°F/2000psi/.5hrs

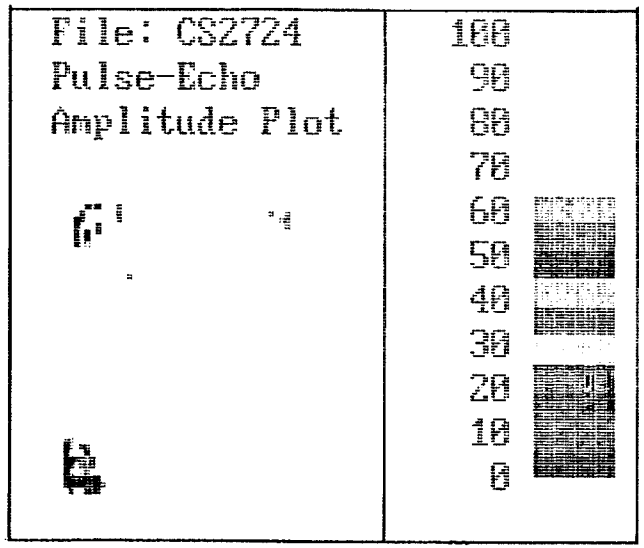


Figure 22. C-scan of the consolidated panel CS272.

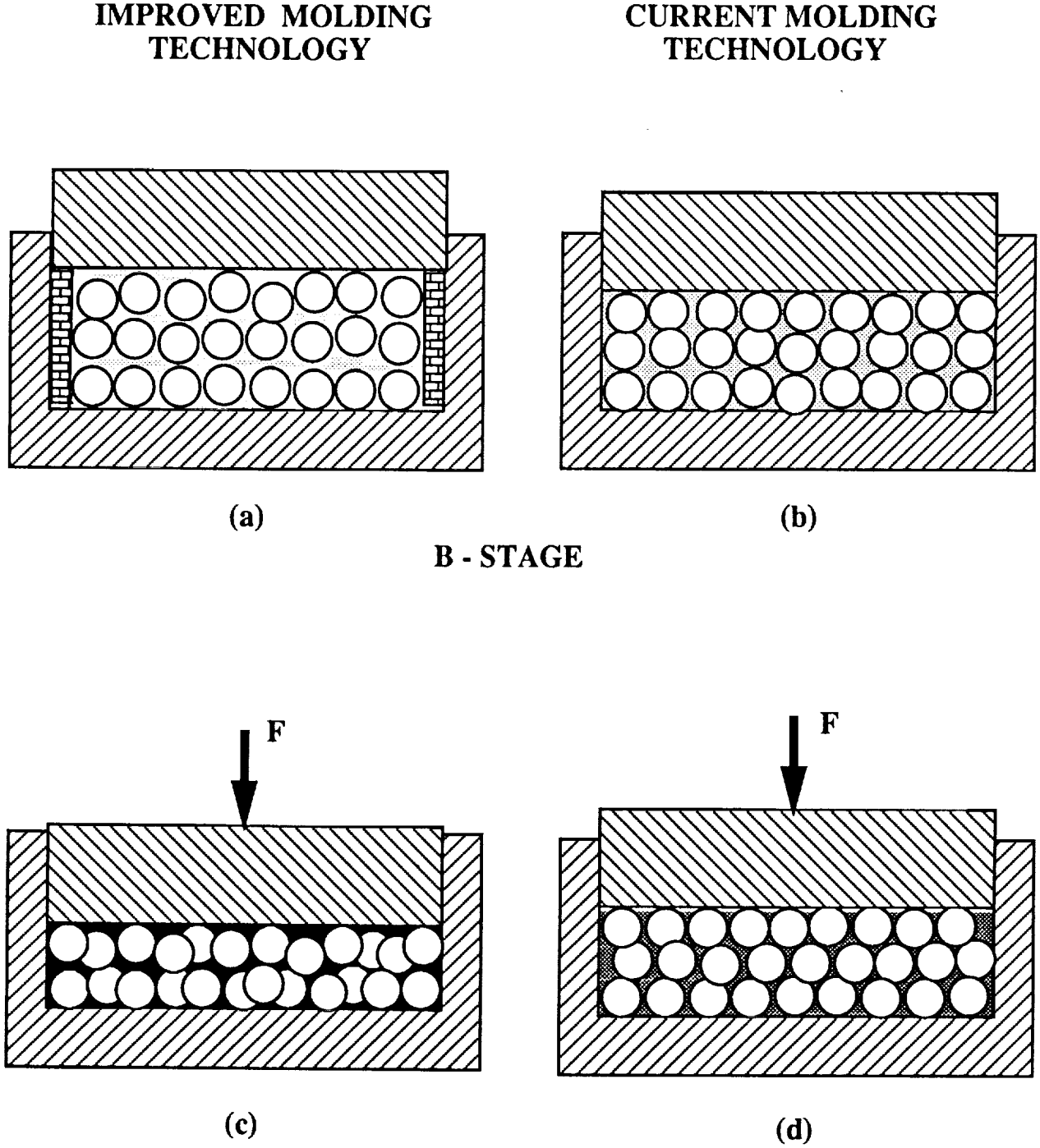


Figure 23. A schematic illustration of the improved molding procedure with the aid of internal molding stops.

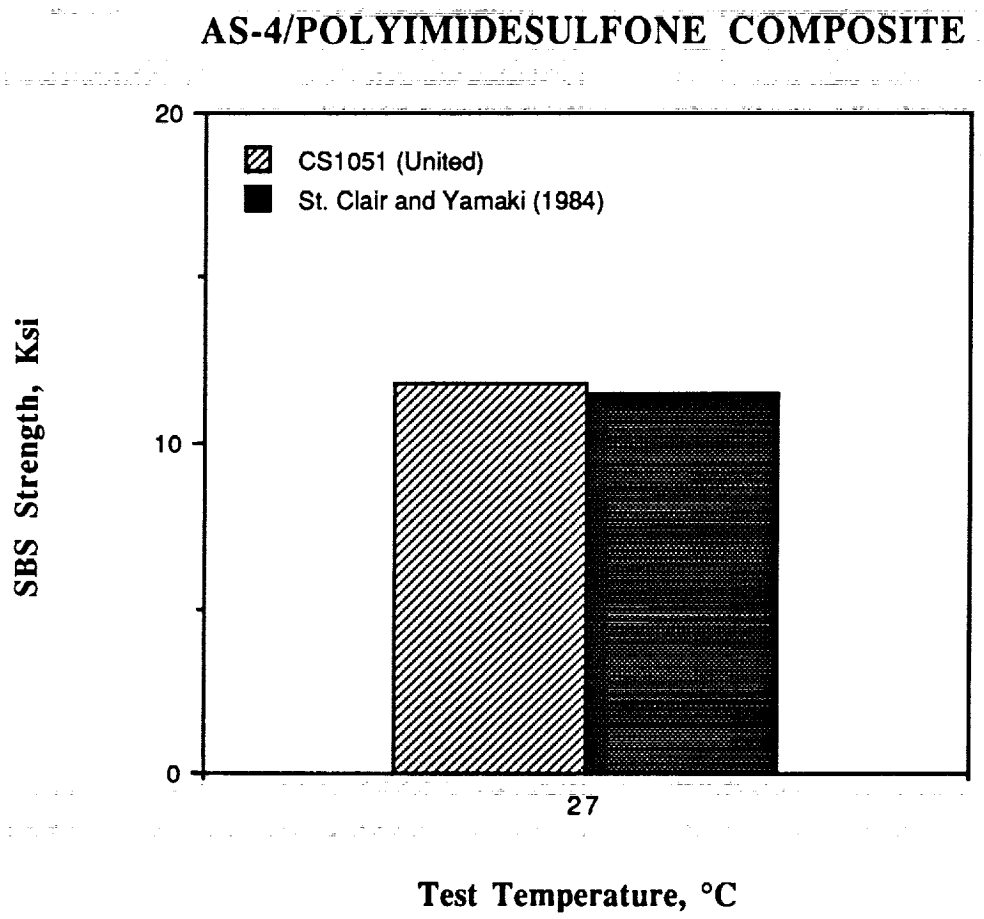


Figure 24. Short Beam Shear strength of the AS-4/polyimidesulfone composites at ambient temperature.

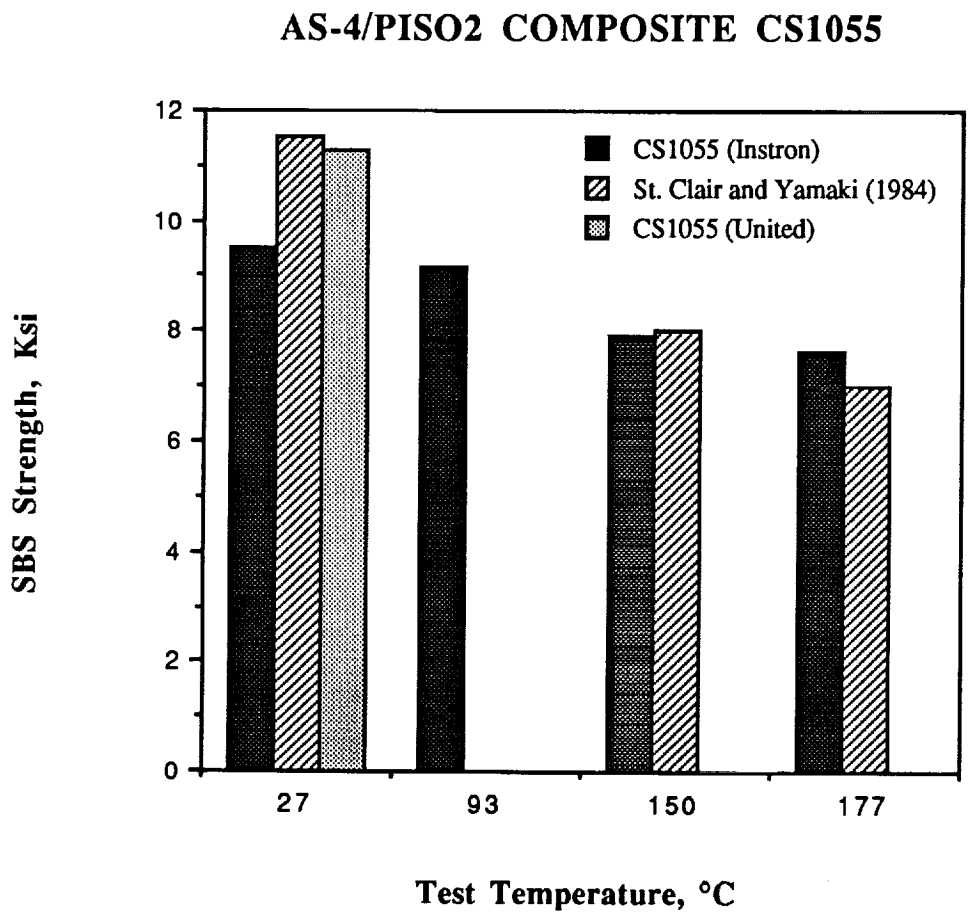


Figure 25. Short Beam Shear strength of the AS-4/polyimidesulfone composites at ambient and elevated temperatures.

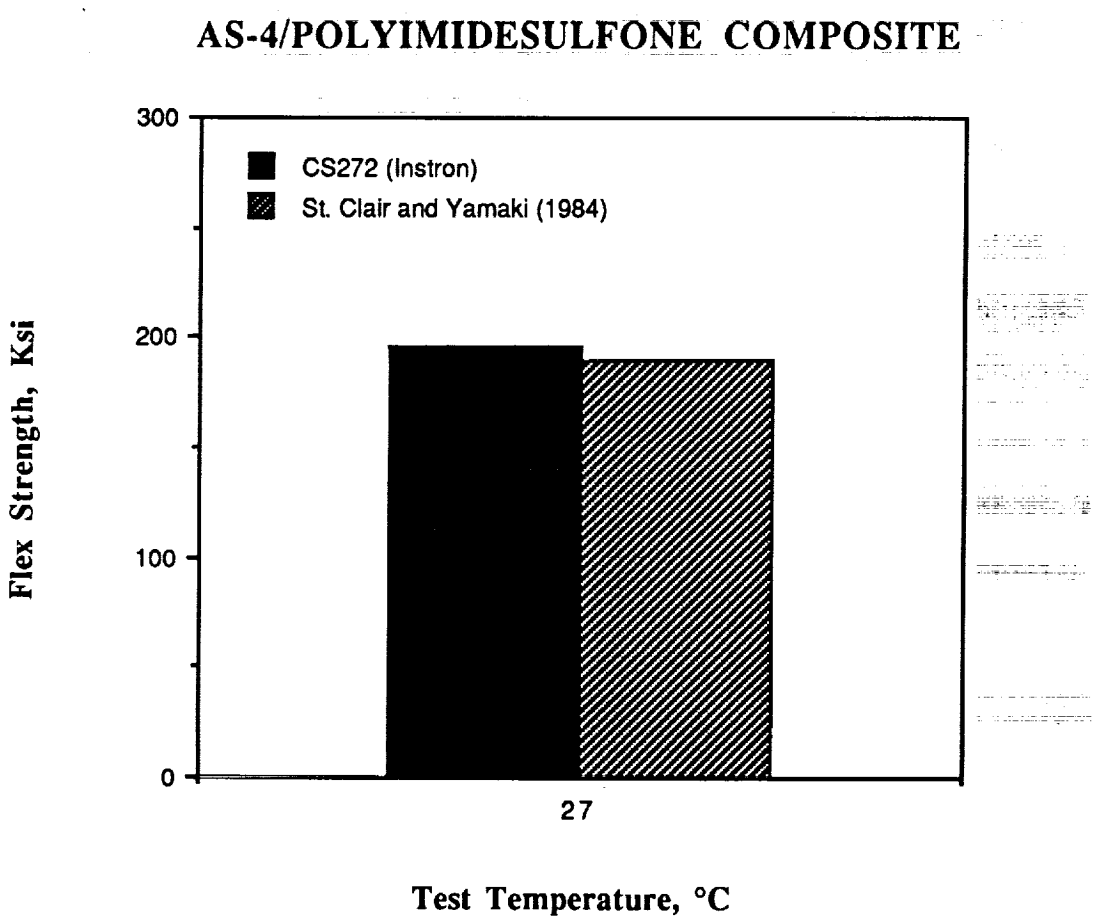


Figure 26. Flexural strength of the AS-4/polyimidesulfone composites at ambient temperature.

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16. Abstract

AS-4/polyimidesulfone ($PISO_2$) composite prepreg was utilized for this improved compression molding technology investigation. This improved technique employed molding stops which advantageously facilitates the escape of volatile by-products during the B-stage curing step, and effectively minimizes the neutralization of the consolidating pressure by intimate interply fiber-fiber contact within the laminate in the subsequent molding cycle. Without modifying the resin matrix properties, composite panels with both unidirectional and angled plies with outstanding C-scans and mechanical properties were successfully molded using moderate molding conditions, i.e., 660°F and 500 psi, using this technique. The size of the panels molded were up to 6.00" x 6.00" x 0.07". A consolidation theory was proposed for the understanding and advancement of the processing sciences. Processing parameters such as the effect vacuum, effect of pressure cycle design, effect of prepreg quality etc. were explored.

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