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Engineering Technology Division

VARIABLE FREQUENCY MICROWAVE (VFM) CURING, PROCESSING OF THERMOSET PREPREG LAMINATES

FINAL REPORT

Felix L. Paulauskas
Oak Ridge National Laboratory

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ABSTRACT

The objective of this work was to investigate the beneficial effect of the **variable frequency microwave (VFM)** technology to cure thermosetting prepreg laminates. Further, it was to investigate the interrelationship and effect on the curing process of frequency, band width, and curing time with different types of laminates.

Previous studies of microwave-assisted curing of neat resins (epoxy) and unidirectional glass and carbon fiber laminates with a fixed frequency of 2.45 GHz, have shown that a substantial reduction in the curing time was obtained [1]. Results of this earlier work indicate that the microwave-assisted curing of multidirectional glass fiber laminates also show a substantial reduction of the required curing time. This may be explained by the penetration of microwave energy directly and throughout the laminate with enhancement of the kinetics of the chemical reaction.

The fixed frequency microwave radiation of 2.45 GHz has been demonstrated to be a partially acceptable method to cure unidirectional carbon fiber laminates. Multidirectional carbon fiber/epoxy laminates demonstrate a lack of coupling during the curing process. A direct curing of these laminates was not possible by microwave radiation with the experimental approach used in agreement with previous work [1,2]. In addition to this short coming, the unidirectional laminate samples cured with the fixed frequency are visually nonuniform. Localized areas of darker colors (burn, hot spots, overheating) are attributed to the formation of standing waves within the microwave cavity. For this reason, the laminates are subject to proper rotation while curing through fixed frequency.

The present research indicates that **variable frequency microwave** technology is a sound and acceptable processing method to effectively cure uni-, bi- or multi-directional thermosetting glass fiber laminates. Also, this methodology will effectively cure unidirectional thermosetting carbon fiber laminates. For all these cases, this technology yielded a substantial reduction in the required cure time of these laminates. Multidirectional carbon fiber laminates demonstrated a lack of coupling of VFM energy during the curing process.

1. INTRODUCTION

Advanced polymer matrix composites have a combination of physical attributes that make them potentially attractive for many applications, particularly where high specific strength and stiffness are needed. One barrier to their widespread use is the long cycle time typically required to consolidate and cure a finished component. Composites are often hand-assembled as a lay-up of prepreg tape, vacuum bagged, then cured in an autoclave under heat and pressure for 2 to 12 hours. Similar metal parts can usually be made in seconds to minutes by stamping or machining. As a result, some composite materials are competitive only in applications where specific performance is required or the cost savings due to lower weight can be shown to overshadow the increase in original manufacturing cost.

If processing cycle times could be reduced significantly, composites would be used in a much broader range of applications. Microwave heating may potentially speed the curing process because the volumetric deposition of microwave energy is more efficient than conduction from the surface. Furthermore, microwave heating seems to enhance polymerization kinetics in some systems, although the exact mechanism by which this occurs is still not understood.

Preliminary work was undertaken with ERL 2258 neat epoxy-based resin samples. These initial results were very encouraging. Samples ranging from 1/16 in. to 1 in. (1.6 mm to 25 mm) thick were cured at times varying from 2 minutes to 30 minutes using a fixed frequency of 2.45 GHz. The microwave power required for this experiment was astonishingly small, on the order of 100 to 200 watts. Similar samples of neat resin cured by conventional oven required a time of 360 minutes. A comparison between microwave processed and oven cured resin shows equivalent mechanical and physical properties. The curing time *was reduced significantly* (factor of 10 to 12) by using microwave radiation in the processing of neat resin [1]. The results were in accordance with the fundamental theoretical hypothesis, that microwave energy would accelerate the curing (crosslinking) in polymers. These results indicate the feasibility of curing polymers and encourage further studies in the composite area.

This microwave study has been extended to the area of polymer matrix composites. The results in these works also indicate a reduction in the curing time with an overlap in the mechanical property values in the microwaved processed samples compared with the conventionally processed samples [1,3].

Most studies reported in recently published literature have used fixed frequency microwave systems at 2.45 GHz. The reason for this is the ready availability of magnetrons at this frequency. However, fixed frequency systems have unpredictable power non-uniformity distribution within the microwave cavity due to the formation of standing waves, resulting in an extremely heterogeneous energy distribution. The negative effect will be the generation of very high power density areas (hot-spots), over heating, etc. of the part being processed. In the processing of thermosetting polymer substances, this problem will be aggravated by the low thermal conductivity and the fact that the crosslinking (polymerization) process is highly exothermic. The processing of thermoset laminates

via fixed frequency microwave radiation often show visible areas of over curing (black, charred areas) that are attributed to these poor non-uniformities inside the microwave cavity.

This investigation pertains to the beneficial effect of variable frequency microwave (VFM) radiation on the processing of thermosetting laminates. Prior work with this type of technology in the processing of neat resins indicates an improvement in this area. It has been shown that frequency sweeping greatly improves heating uniformity in the slabs or disks of neat resin reducing the required curing time [4].

VFM technology uses a traveling wave tube to sweep a range of frequencies. This sweeping of frequency range will excite many different microwave modes within the microwave cavity, thus enhancing the power uniformity. Continuous sweeping through several modes within a very short time interval (milliseconds) results in an averaged, more uniform energy distribution in the processing cavity. The result of this effect is the elimination of non-uniform heating (power distribution) in the workpiece. This technology also provides a better consistency and reproducibility of the heating characteristic in the workpiece, independent of its location inside the processing cavity [5,6]. Figure 1 schematically compares the power distributions in a fixed frequency and in a variable frequency microwave cavity. Note the homogenization effect in the power distribution within the variable frequency cavity. This technology also has the advantage of selection of a frequency (center frequency), which can maximize the power absorption in the workpiece. Subsequent sweeps in a range of frequencies on both sides of this center frequency will optimize the energy absorption in the workpiece. In some cases, a variable frequency microwave system can be made to function as a single frequency source by choosing a center frequency and making the sweeping frequency range equal to zero. In this case, the center frequency becomes the fixed frequency.

Conventional curing processes involve ovens, presses and autoclaves that are large and expensive capital investments. These devices are very energy inefficient. Only a small portion of the total energy consumed by these conventional curing systems goes into the chemical kinetics to produce crosslinking in the polymeric material. VFM technology is expected to be more efficient by providing volumetric energy deliverance into the polymeric material or workpiece, directly at the intergranular or molecular level.

The application of this technology to thermoset laminates will permit the advancement toward the needed manufacturing breakthrough in this area. Furthermore, it is imperative to explore the interrelationship between center frequency, band width, applied microwave power, resin and cure times, and different types of fibers and their orientation in the composite. Special attention should be given to the study of the effect of higher frequencies in the processing of these laminates. It is expected that these higher frequencies (10 - 20 GHz) will have a more positive impact in the processing of these materials due to their potential for a higher density of energy deposition per time unit.

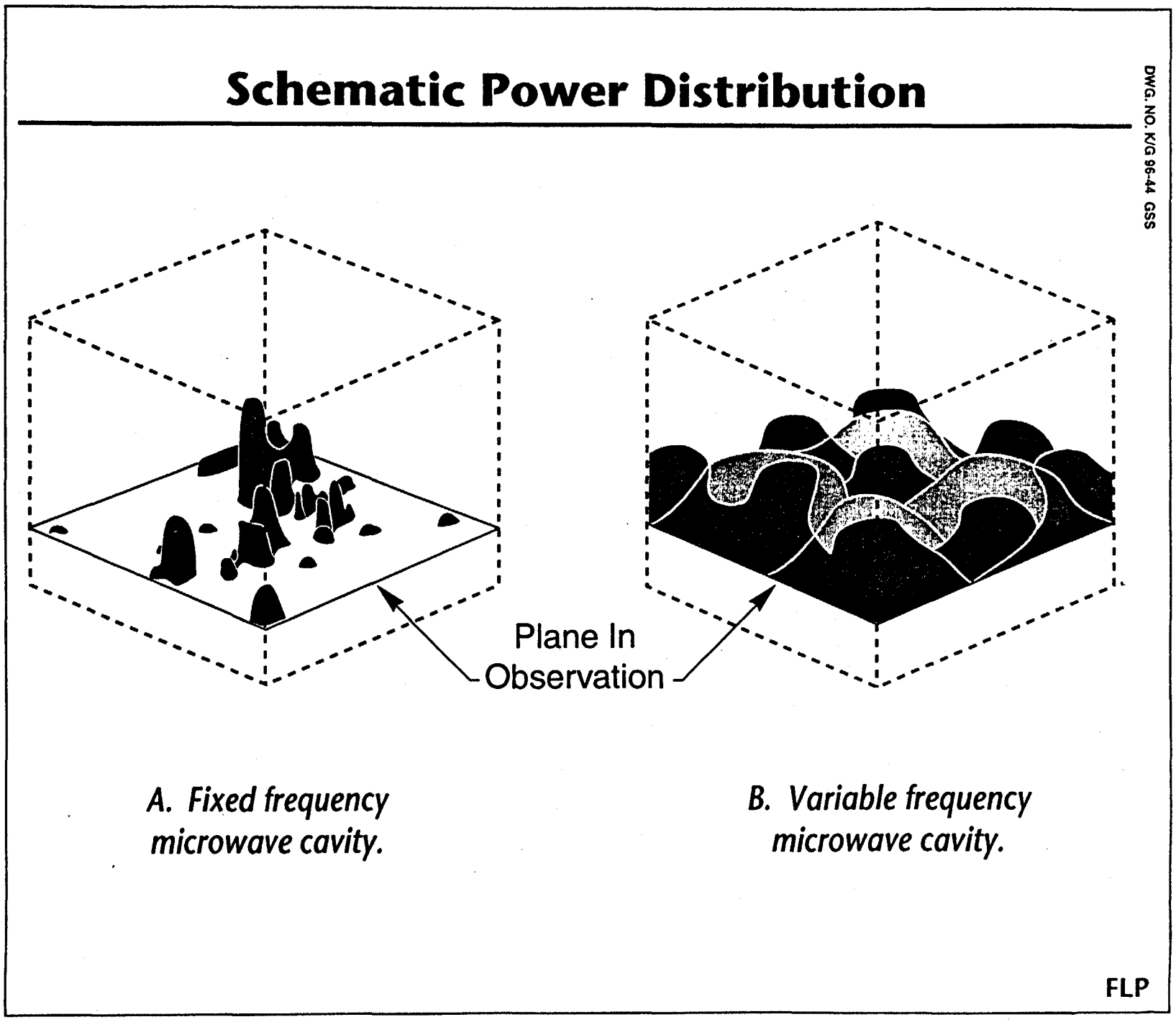
Schematic Power Distribution
DWG. NO. K/G 96-44 GSS
Plane In
Observation
A. Fixed frequency microwave cavity.
B. Variable frequency microwave cavity.
FLP

Figure 1

2. EXPERIMENT

2.1 Experimental Materials

In these VFM assisted composite studies, two different kinds of composite laminates were selected for evaluation:

a) Carbon fiber/epoxy (Hercules IM6G-3501-6)
b) Glass fiber/epoxy (Scotchply 1003-UMI)

With this selection, the different microwave coupling behaviors with these two types of reinforcing fibers, can be observed. The 6 in. x 6 in. (15 cm x 15 cm) composite laminate samples for the initial experiments were manufactured using eight superposed layers of each basic thermoset prepreg material, where the fibers in each layer were oriented in various directions to obtain the desired fiber configuration. Three basic (laminate) configurations of fiber orientation were selected:

$[0]_8$	:	0/0/0/0 / 0/0/0/0 (unidirectional)
$[0/90]_{2S}$	:	0/90/0/90 / 90/0/90/0 (bidirectional)
$[0/45/90/-45]_S$	:	0/45/90/-45 / -45/90/45/0 (multidirectional)

These numbers are degrees and indicate the fiber orientation of each single layer of prepreg in the laminate relative to a 0 degree reference.

2.2 Sample Preparation

The preparation of these laminate specimens is a time consuming, and intricate process. After each new superposed layer is positioned, the partially manufactured specimen is placed under a vacuum to assure an air-free interlayer condition.

The laminate sample is placed between two thick glass cloths with the objective to collect the excess resin which is squeezed out of the laminates during the curing process. To avoid adhesion between the laminate sample and thick glass-cloth, a teflonated, fine-screen film (bleeder screen-cloth) is used. The bleeder cloth allows the resin to percolate towards the thick glass-cloth. Similarly, to avoid bonding at the quartz or pyrex glass plates, a release film is placed at these location. See Figure 2.

For the processing, these laminate samples are placed in the consolidation press inside the microwave furnace (Figure 3) or between the metal plates of the standard heating press for conventionally processed samples.

Composite Laminate / Sample Configuration

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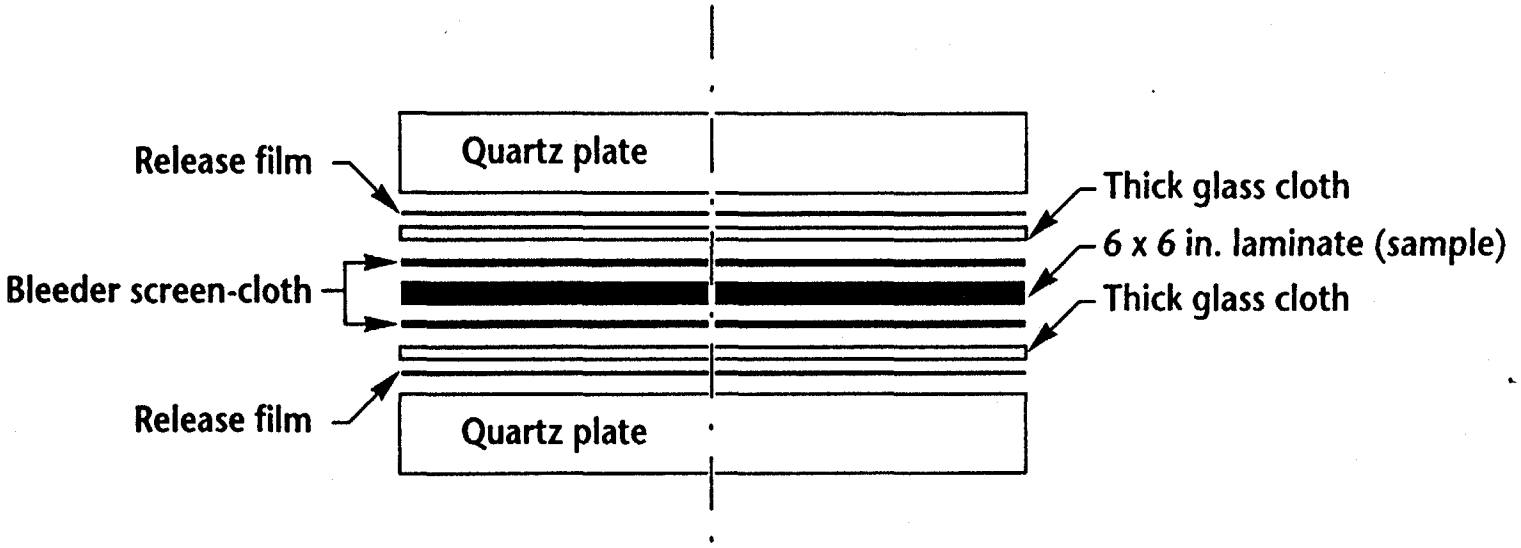

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Figure 2

Variable Frequency Microwave Processing: Configuration Of The Laminate Curing System Inside The Cavity

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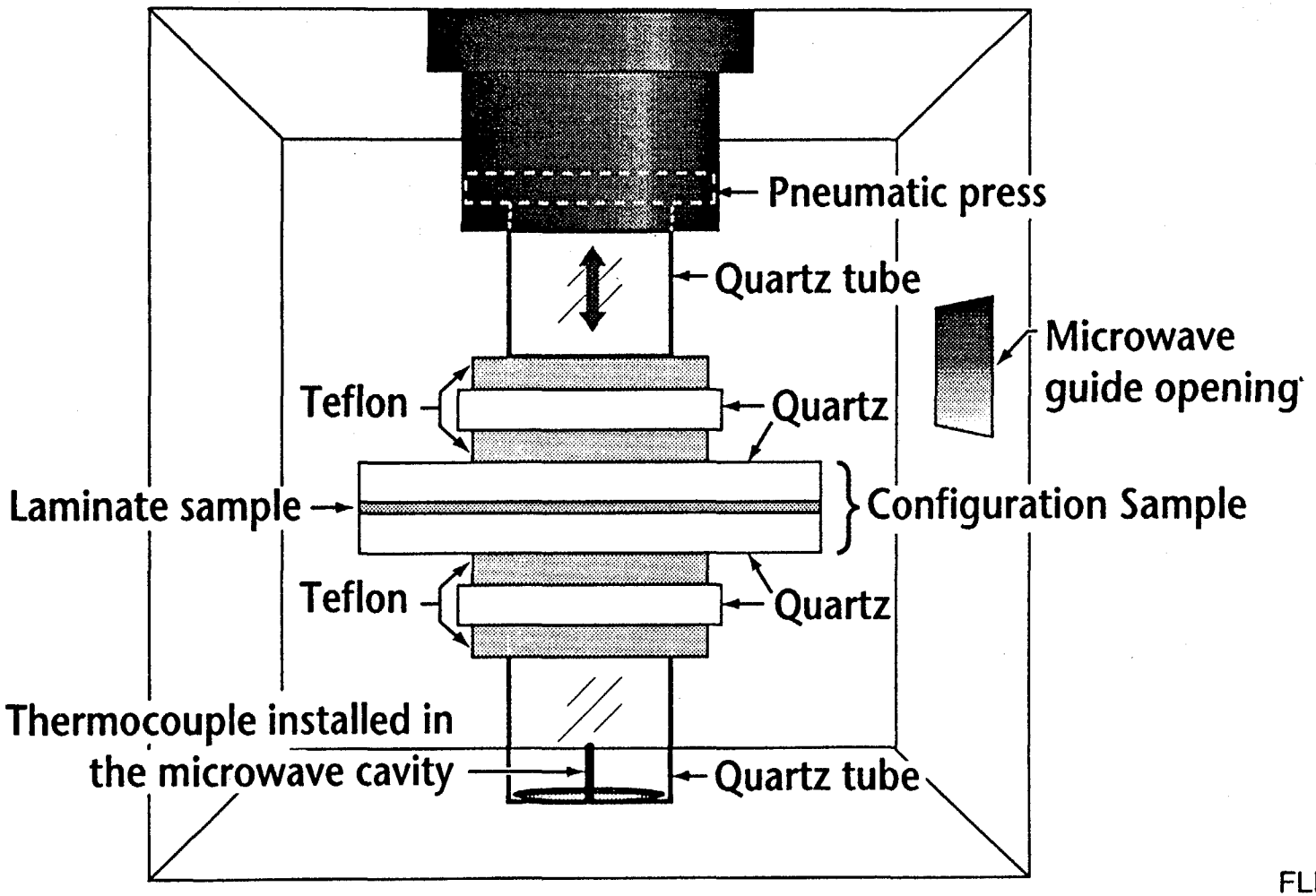


Figure 3

2.3 Processing Equipment

A multimode, variable frequency microwave furnace, Model T-4001E (prototype furnace) was utilized in this project. This furnace is manufactured by Lambda Technologies, Inc. This equipment is capable of delivering a maximal nominal (forward) power of 200 watts over two selected frequency ranges of 2.5 - 7.5 GHz and 7.5 - 17.5 GHz. This VFM oven may be operated at a fixed frequency, or the entire bandwidth or any segment therein may be swept at a discrete interval rate of 100 milliseconds to 2 minutes. The dimensions in this VFM oven are: 12.0 x 12.0 x 10.0 inches.

To prevent the possible migration of any volatiles or generated gases into the waveguide and subsequently contaminating the power sensors, a thin sheet of polyimide film was placed over the opening of the waveguide to the cavity.

This microwave oven features a built-in pneumatic driven consolidation press, capable of delivering the required specific compression load on the laminate with curing.

Prior to the start of the testing program, a calibration check of the clamping force of the pneumatic built-in press was undertaken. A leakage check in the VFM cavity was also conducted.

2.4 Microwave Processing

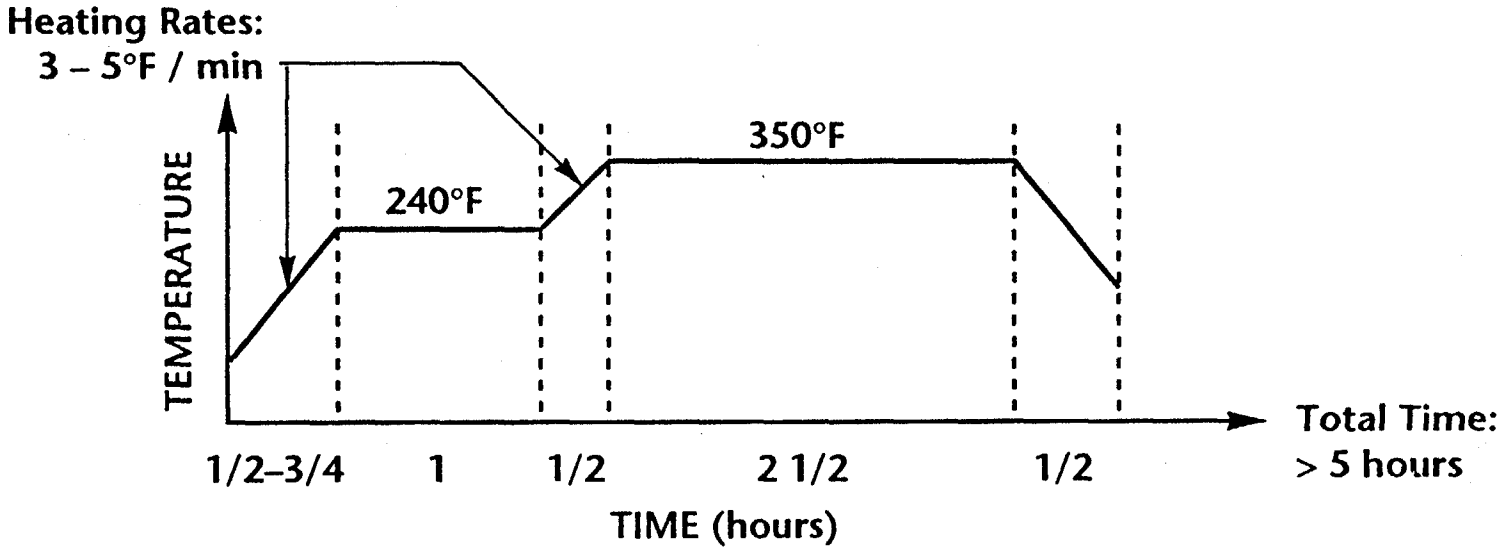
For conventional processing, the standard curing sequence and time recommended by the material manufacturer was followed. The required time exceeded five hours for the standard cure cycle. If an additional postcure cycle is required, then the overall curing cycle can be as long as eight to ten hours. The conventional cure cycle for the carbon laminate is depicted in Figure 4. In the conventionally cured samples, due to the dynamics of the heat transfer during the curing, the resin is heated very slowly, inwardly over a relatively long period of time.

In the preliminary stages of this program, start-up and pre-run experimental processing of the laminates with the VFM system was undertaken. The intent of these pre-runs was to evaluate the microwave oven for its stability, uniformity and repeatability of the process parameters, such as temperature inside the cavity, and forward and reflected power as a function of the selected center frequency and bandwidth. All these pre-runs were undertaken with the laminate samples in the cavity and were carried out with the low band frequency (2.5 to 7.5 GHz).

Evaluative trials were also undertaken to gain knowledge about the microwave energy deposition on the glass plates of the sample configuration. The principle of microwave processing is the deposition of energy directly and volumetrically quasi-equally into the workpiece. A substantial energy deposition on the system containing or surrounding the part to be processed will cause an "indirect" curing, called hybrid heating. In this case, the container or mold is heated and then the heat is transmitted by conduction, convection, or radiation to the workpiece. In this project, it is very important to reduce this hybrid heating to a minimum. Here, an evaluation of the temperature increases of the glass plates in the sample configuration, (Figure 2) without a laminate specimen in

Typical Conventional Cure Cycle
Carbon-Fiber-Prepreg Laminate: IM6G-3501-6

DWG. NO. K/G 96-435 GSS



For high-temperature applications, a *post-cure* cycle is recommended to raise the laminate glass transition temperature (Tg).

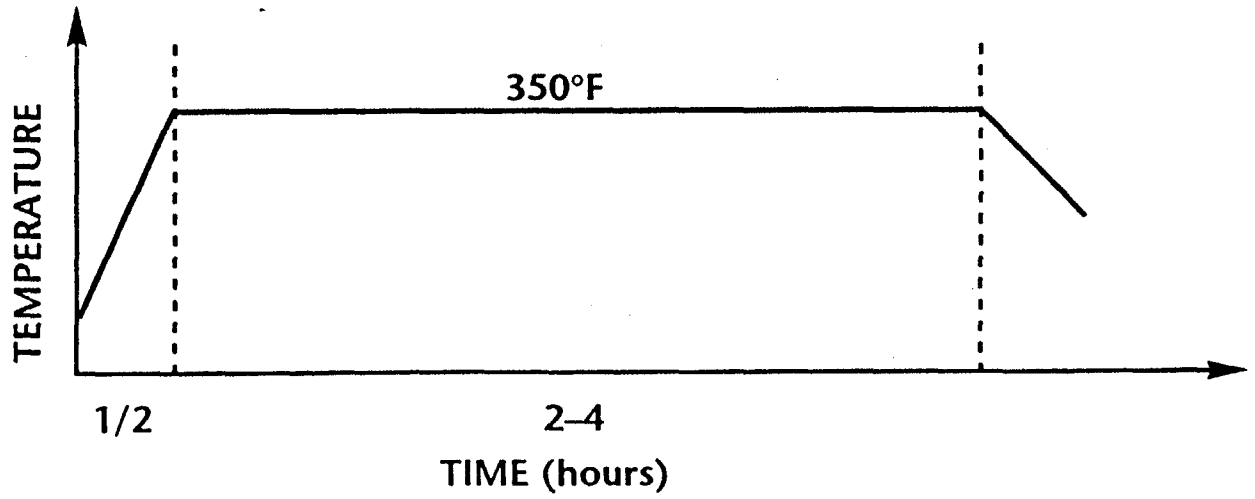


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Figure 4

between plates, was undertaken under similar conditions to when laminate samples are processed. The results are shown in Figures 5 and 6. The pyrex plates suffer a remarkable temperature increase compared to the quartz plates where both types of plates were processed for 15 minutes under equal conditions (Figure 5). For the contrary, the quartz plates demonstrated a minute temperature increase when subjected to an extended processing of 60 to 90 minutes (Figure 6). For this reason, only quartz plates were used in this VFM project.

For the processing of the laminate samples with the VFM system, the sample configuration indicated in Figure 2 is placed in the pneumatic press inside the oven and subsequently subjected to a specific compressive load of 11 to 12 psi. (See Figure 3).

For the low frequency band (2.5 to 7.5 GHz) a center frequency of 5.00 GHz was selected. This frequency was selected in the pre-run trials based in a high forward power deliverance and good coupling characteristics of the laminates at this center frequency. The frequency was swept at ±0.50, ±1.00 and ±2.00 GHz on both sides of the center frequency (see Figure 7). The frequency range (Δf) will be defined as the entire swept region; i.e., 1.00 GHz, 2.00 GHz and 4.00 GHz. For the high frequency band (7.5 - 17.5 GHz) a center frequency of 13.80 GHz was selected. Here the frequency was swept ±0.50 GHz on both sides of the center frequency only (see Figure 8).

The selected cure times for the entire program were 45, 60, 90, and 120 minutes. In some specific cases a 75 minute cure time was selected in order to complement the experimental data with an additional point.

3. RESULTS

Based on visual observation, the glass fiber laminates processed via VFM technology did not exhibit any "brown spot" areas of overheating or heat damaged areas. This result was similar for the low and high frequency ranges. Due to the natural light color of these glass laminates, any heat damaged areas or burn spots on these samples will be revealed conclusively. Contrary to the VFM experience, similar samples cured with fixed frequency microwave technology showed darker areas or regions of overheating [1]. These localized areas of overheating are attributed to the formation of standing waves within the fixed frequency microwave cavity. To avoid this common phenomenon in fixed frequency processing, the samples are rotated, or moved to another location inside the cavity, at discrete intervals throughout the complete curing process.

Similar qualitative visual observations can not be made with the carbon fiber laminates due to the natural dark (black) color of these laminates. Unidirectional carbon fiber laminates demonstrated very satisfactory curing with variable frequency technology. Frequency sweeping is undoubtedly an excellent technology for the curing of all the above indicated composite laminates.

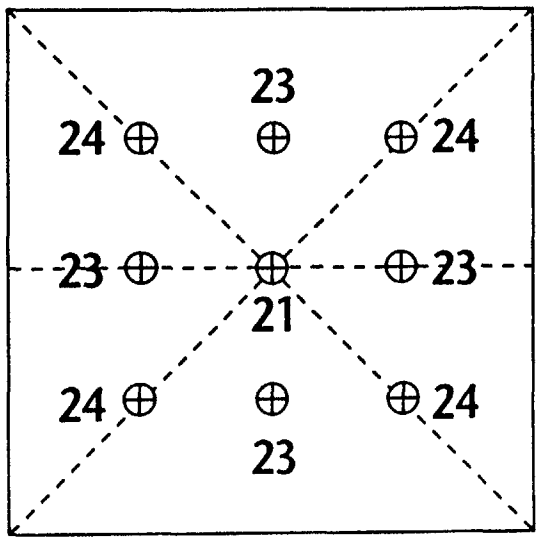
Variable Frequency Microwave Processing : Surface Temperature Increase On the Glass Plates Without Laminate Samples In Between Plates

DWG. NO. K/G 95-646 GSS

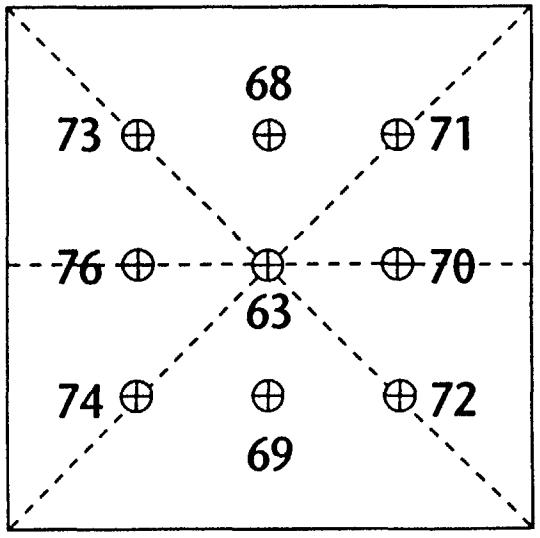
Center frequency (CF) :	5.00 GHz
Frequency range (Δf) :	1.00 GHz
Sweeping rate :	100 ms
Microwaved for :	15 min at 190 / 195 Watts
Initial plates temperature :	19°C
Plate dimension :	3/8 in. , 6 in. x 6 in.

Temperatures In °C Measured With A Rapid Response Surface Probe

QUARTZ PLATES



PYREX PLATES



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Variable Frequency Microwave Processing : Surface Temperature Increase On the Quartz Plates Without Laminate Samples In Between Plates

DWG. NO. K/G 95-657 GSS

Center frequency (CF) :	5.00 GHz
Frequency range (Δf) :	1.00 GHz
Sweeping rate :	100 ms
Microwaved for :	60, 90 min at 190 / 195 watts
Initial plates temperature :	20°C
Plate dimension :	3/8 in. , 6 in. x 6 in.

Temperatures In °C Measured With A Rapid Response Surface Probe

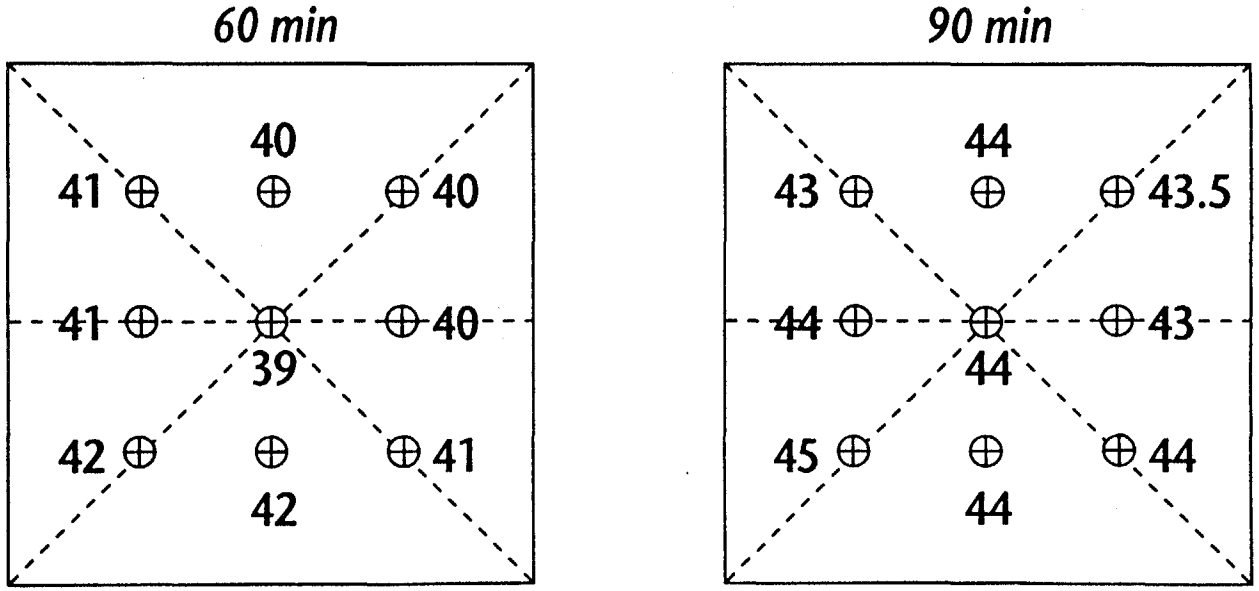

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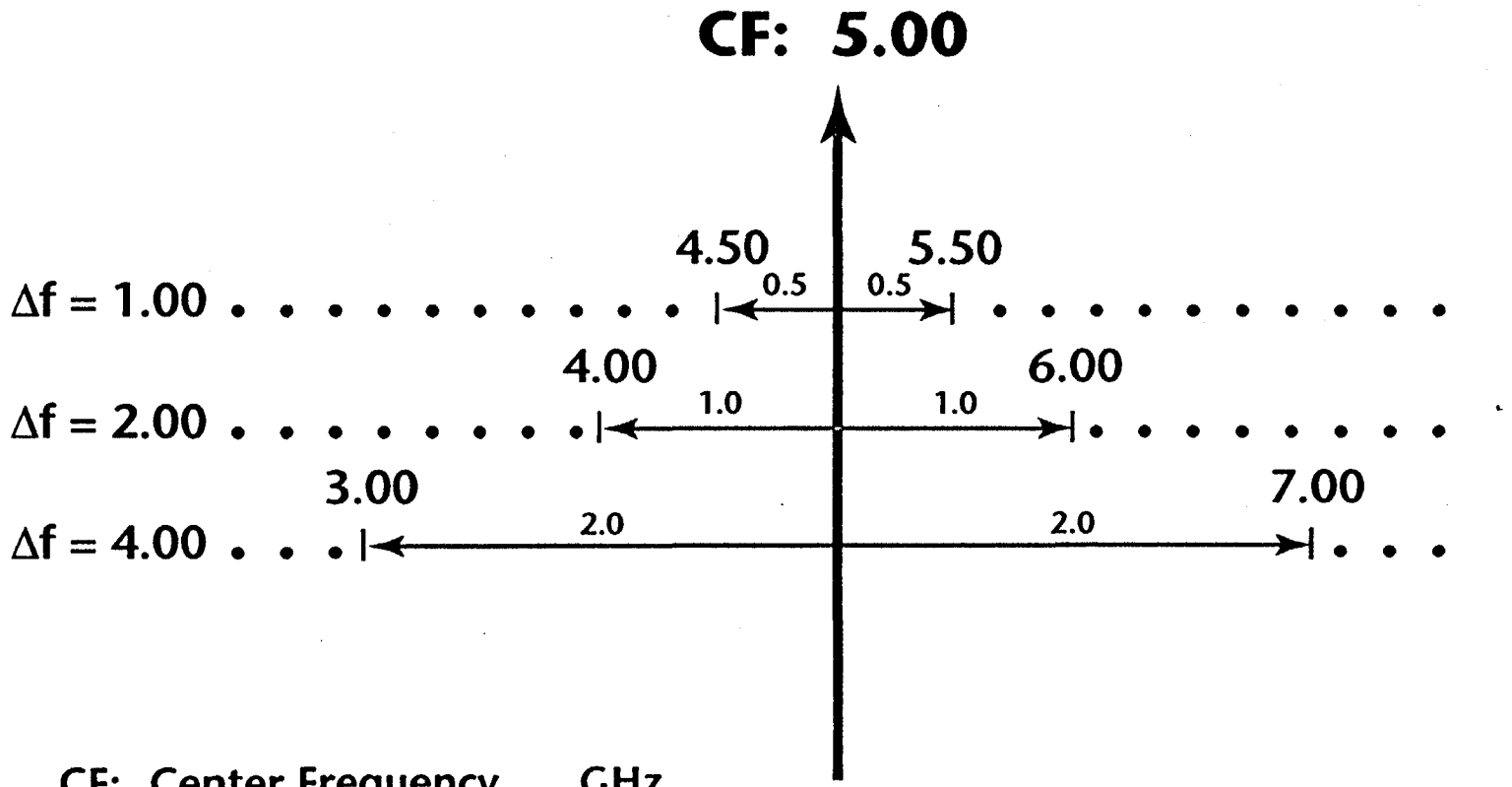
Figure 6

Variable Frequency Microwave Processing Thermosetting Prepreg Laminates. Processing Spectrum

DWG. NO. K/G 96-43R GSS



CF: Center Frequency GHz
Δf: Frequency Range GHz
Sweeping Rate 100 ms
Low Frequency Range *(2.5 to 7.5 GHz)*

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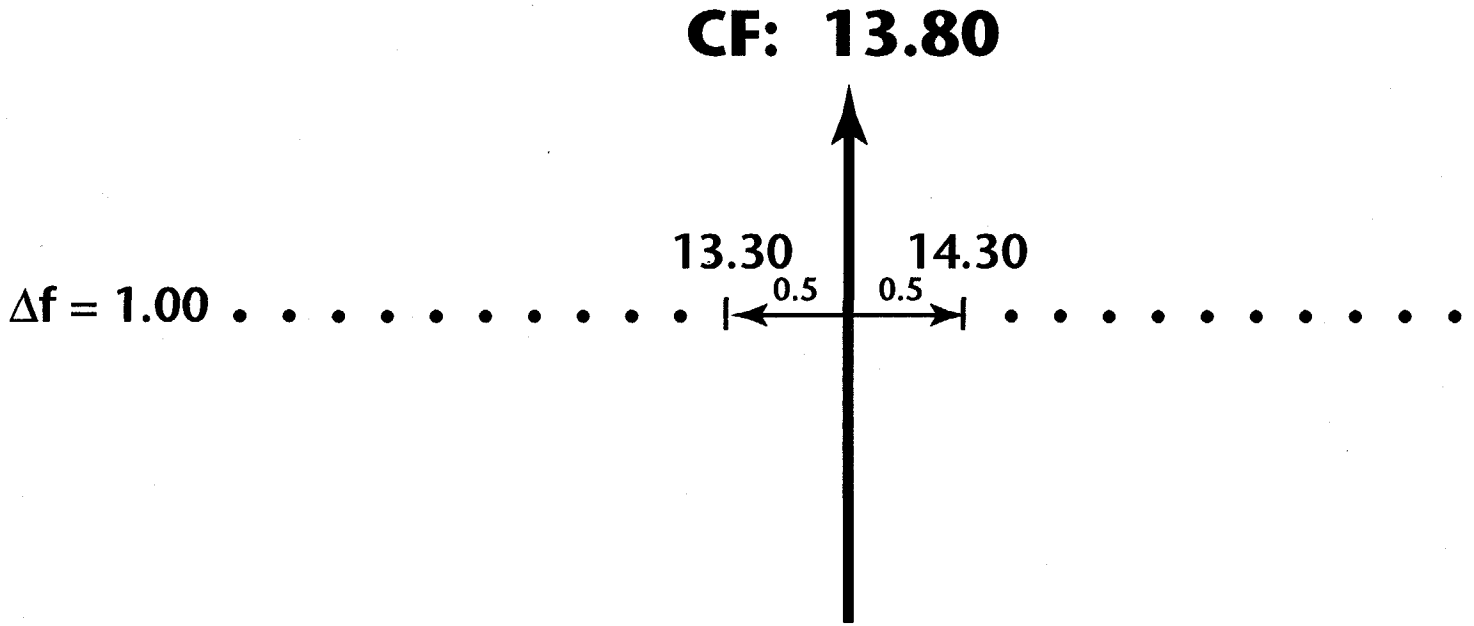
Figure 7

Variable Frequency Microwave Processing Thermosetting Prepreg Laminates. Processing Spectrum

DWG. NO. K/G 96-434 GSS



CF:	Center Frequency	GHz
Δf:	Frequency Range	GHz
	Sweeping Rate	100 ms
High Frequency Range		*(7.5 to 17.5 GHz)*

FLP

Figure 8

Bidirectional and multidirectional carbon fiber laminates exhibited a lack of coupling with variable frequency microwave energy. These two types of laminates were unable to be cured directly through exposure to any of the frequencies used in the processing spectrums. However, some degree of curing was observed in the bidirectional carbon fiber laminates. This degree of curing appeared to be acceptable and homogeneous throughout the samples. Similar results were achieved in the processing of these laminates with fixed frequency microwave radiation [1]. Although proper processing of these bi- and multidirectional carbon fiber laminates can not be obtained through direct microwave radiation, curing is still achievable by hybrid curing. Here the quartz plates of the sample configuration (see Figure 2) should be exchanged with another plate/material (e.g. specifically pyrex glass) with high "lossy" characteristics. This alternate approach for curing of laminates is recommended only for thin wall parts.

3.1 Differential Scanning Calorimetry (DSC)

Differential scanning calorimetry was performed on representative samples from each processing time to measure the degree of cure. Additionally, the homogeneity of the cure was examined by taking several DSC samples from various locations on each sample. It was found that there is a direct correlation between cure time and degree of cure. Glass fiber laminates that were exposed to VFM radiation for 45 minutes exhibited an average glass transition temperature (T_g) of only 155 °C while those samples that were processed for 120 minutes had an average T_g of 178 °C (see Figure 9). With two hours processing time, the T_g is equivalent to those samples conventionally processed for approximately 7 to 10 hours. Unidirectional carbon samples exhibited somewhat lower glass transition temperatures (approximately 140 °C) as compared to the conventionally processed samples (approximately 175 °C). Bidirectional and multidirectional samples were unable to be cured using 5.0 GHz as the center frequency. However, the degree of cure appeared visually to be homogeneous throughout all samples for the entire range of processing times.

3.2 Mechanical Properties

During the microwave processing of the laminates it is interesting to observe the profiles of power levels (forward and reflected) and temperature inside the cavity for the entire curing process. The microwave equipment features only digital monitoring or display of these processing variables. No real time data acquisition was available. Time related data of these process variables was taken (read-out) at discreet intervals of time. These profiles of power and temperature in the microwave cavity can be observed in Figures 10, 11, and 12. These figures indicate that the processing time for each sample was 120 minutes at a frequency of 5.00 GHz (low frequency range). The temperature profile in the microwave cavity can only be used as a reference value for comparison between laminate samples. Figure 3 displays the location of this thermocouple inside the cavity. This thermocouple is enclosed inside the lower quartz tube and sealed with a teflon plate at the top. In addition to this fact, the thermocouple is located at a considerable distance from the sample being processed. Under this condition, this thermocouple will provide temperature values of the air inside of an enclosed system, quasi-independent of the microwave cavity temperature or sample temperature.

Variable Frequency Microwave Processing Thermosetting Prepreg Laminates Unidirectional Glass-Fibers Laminates, 8 plies Tg vs. Curing Time

DWG. NO. K/G 96-458R GSS

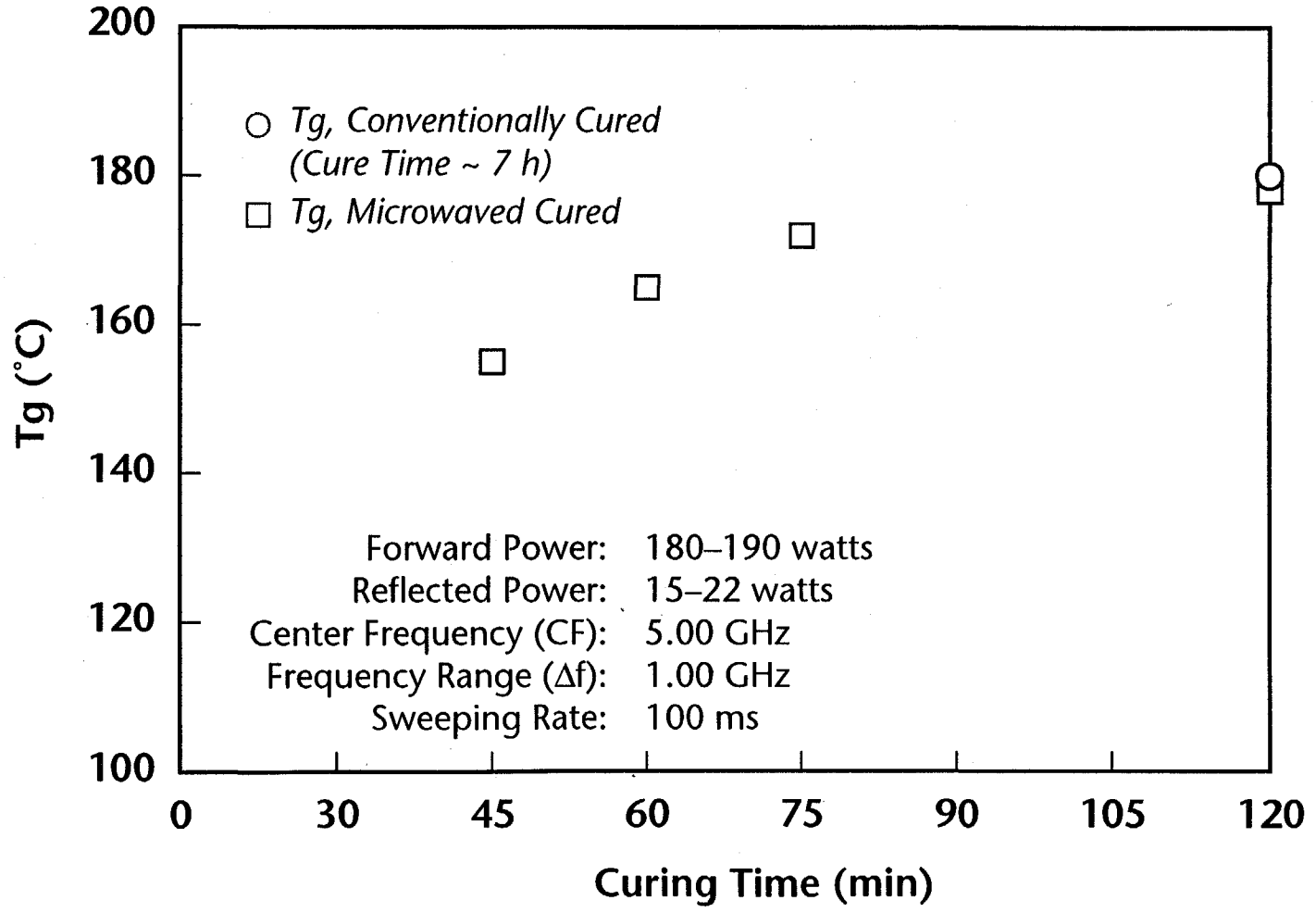


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Figure 9

Variable Frequency Microwave Processing: Profiles Of Power Level And Thermocouple Readout For Temperature In Cavity During The Processing Of Thermosetting Prepreg Laminates

DWG. NO. K/G 95-645R2 GSS

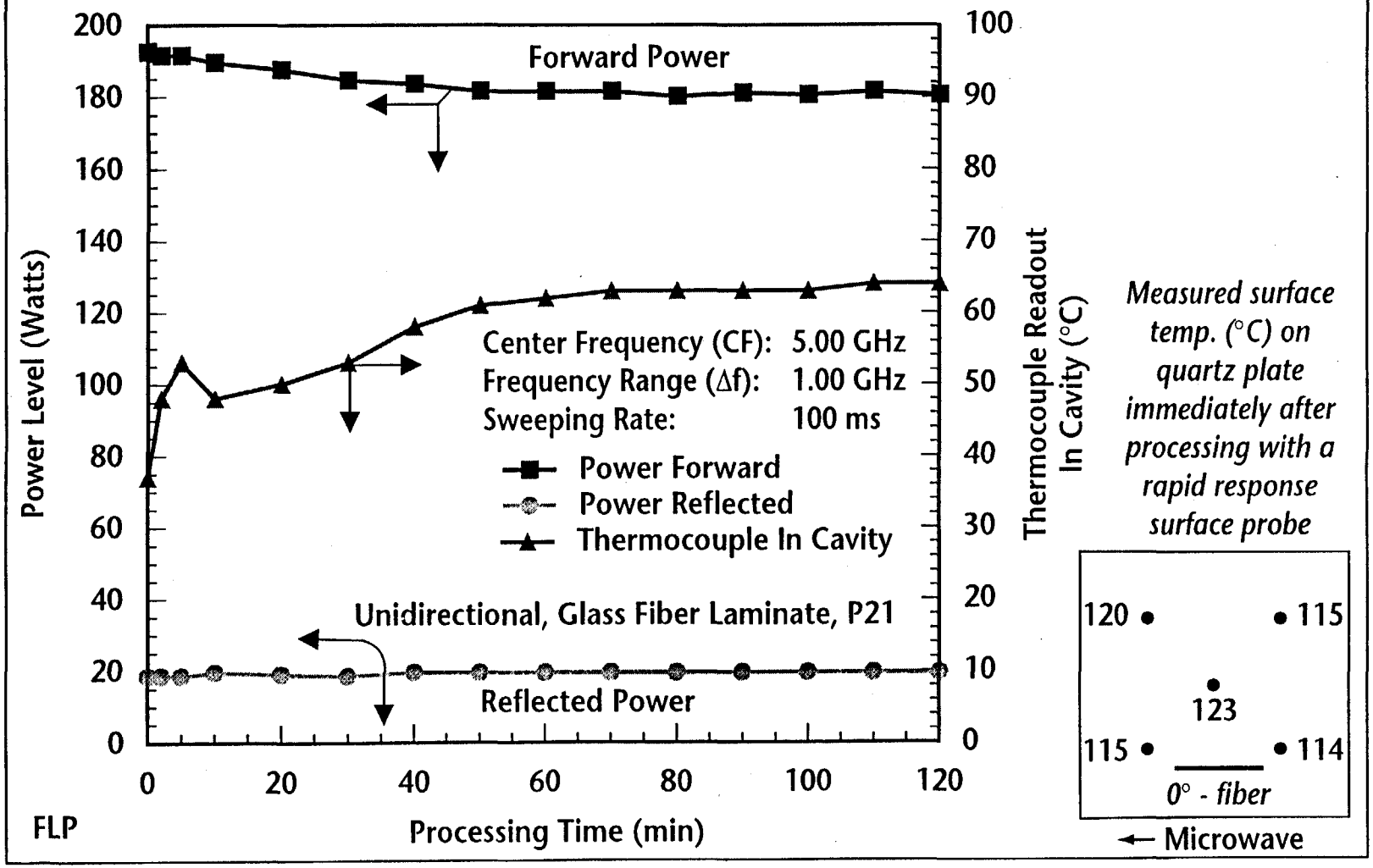

Figure 10

Variable Frequency Microwave Processing: Profiles Of Power Level And Thermocouple Readout For Temperature In Cavity During The Processing Of Thermosetting Prepreg Laminates

DWG. NO. K/G 95-647R2 GSS

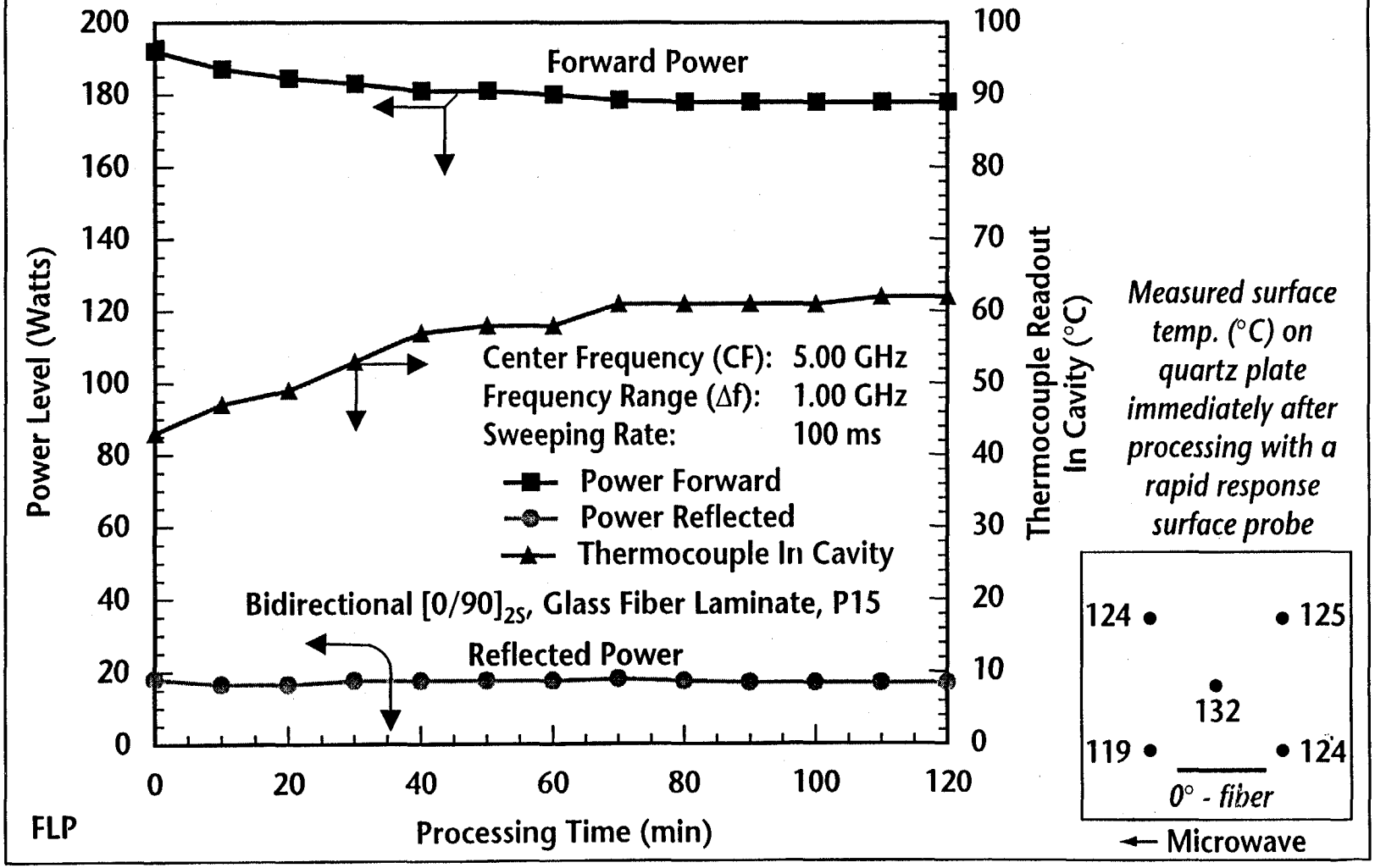

Figure 11

Variable Frequency Microwave Processing: Profiles Of Power Level And Thermocouple Readout For Temperature In Cavity During The Processing Of Thermosetting Prepreg Laminates

DWG. NO. K/G 95-681R2 GSS

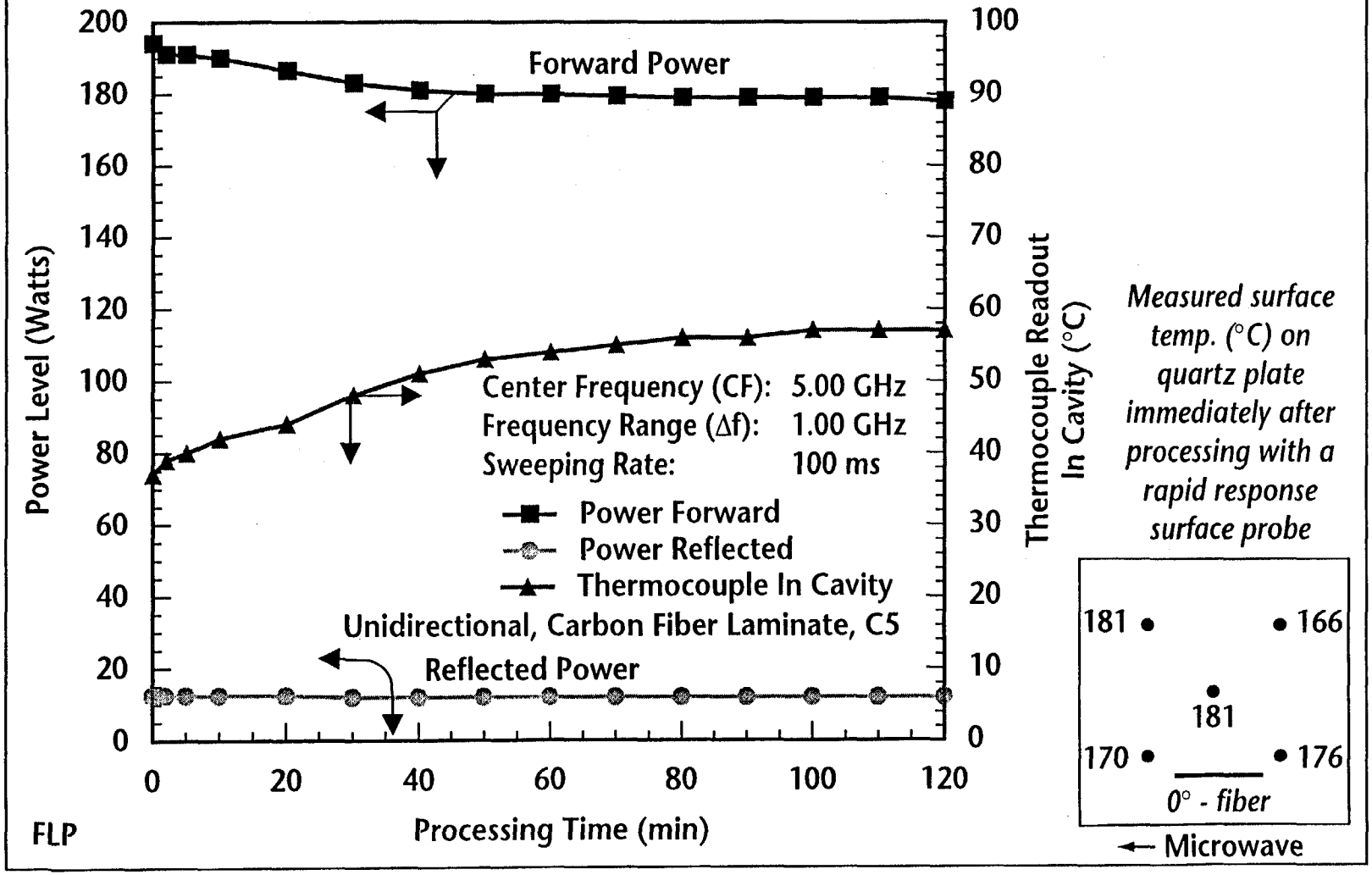

Figure 12

The forward power and the thermocouple read-out both changed steadily and gradually with the processing time; forward power declined and the thermocouple signal increased. The reflected power changed negligibly. The temperature differences measured on the quartz plate, immediately after the completion of the curing process, for the carbon fiber laminates are recognizably higher than for the glass fiber laminates. Possible reasons for these temperature differences include: i) different types of resin, ii) different exotherm characteristics, iii) different types of fiber with different reflective properties, iv) different coupling/lossy characteristics of both types of laminates.

Figure 13 depicts the data of Tables I and III, which are for unidirectional glass fiber laminates processed via VFM with a center frequency of 5.00 GHz. This data compares the ultimate tensile strength (σ_B) for VFM cured samples with conventionally processed samples. The microwave processing data were collected over various cure times. Both, carbon fiber laminates and glass fiber laminates exhibited a smooth increase in the ultimate tensile strength as a function of sample cure time. It is important to denote that different resin systems were used for the two different types of studied laminates. The limit of a two hour microwave processing time was selected based on the premise that any longer cure time would not represent a significant improvement between microwave curing and conventional curing techniques. In spite of the fact that σ_B varies in a steadily increasing manner, the samples that were microwave cured for 2 hours did not achieve strength values equivalent to those for conventionally cured samples.

The difference in the ultimate tensile strengths observed between the conventionally cured fiber laminates, subjected to a standard conventional cure cycle, and the carbon fiber laminate samples cured via microwave process was of the order of ten percent. A larger difference in σ_B was observed for the glass fiber processed with the different methodologies. The glass-fiber laminates cured conventionally were subjected to an additional post cure cycle with a subsequent increase in σ_B. As indicated in Figure 13 less than maximum nominal input power was utilized in the curing process (forward power minus reflected power is approximately equal to 175 watts). A microwave applicator with superior forward power capability would very likely produce an increase in σ_B observed values for identical processing times.

Table II represents data similar to Table I, but only the configuration in the ply orientation is different. Ply orientation in these glass fiber laminates were $[0/90]_{2S}$ and $[0/45/90/-45]_S$. In both studied ply configurations the fibers were the main contributor to the observed σ_B values and not the resin. Very little variation in σ_B was observed as a function of microwave curing time and also between conventional and microwave processed samples. This is in accordance with the above statement that the fibers are the main contributor for the σ_B values. These results also point out that no evident damage occurs to the fibers through exposure to microwave radiation.

Tables IV to VI represent similar results obtained using a center frequency of 13.80 GHz (higher frequency band) over a frequency range of 1.00 GHz. (In tables V and VI, there is not data for samples cured at 13.80 GHz for 45 minutes due to physical damage incurred during sample preparation of the tensile strength specimens cutting the 6 x 6 inches laminates). Again the results are a comparison of σ_B for microwave processed and conventionally processed samples. As before,

Table I

Variable Frequency Microwave Processed And Conventionally Cured Unidirectional Glass Fiber Thermosetting Laminates, 8 Plies

Ultimate Tensile Strength (σ_B) vs Curing Time (CT)

DWG. NO. K/G 96-58R2 GSS

Center Frequency: 5.00 GHz
Frequency Range: 1.00 GHz
Forward Power: 180 – 190 watts
Reflected Power: 15 – 22 watts
Sweeping Rate: 100 ms

Ultimate Tensile Strength, σ_B (psi)

Variable Frequency Microwaved						Oven-cured samples with post cure, cure time: > 7 h, $[0]_8$ - laminates	
		90° (Perpendicular to the fibers)		0° (Parallel to the fibers)		90° (Perpendicular to the fibers)	0° (Parallel to the fibers)
Ply Orientation	Cure Time CT (min)	Average	Range	Average ($\times 10^3$)	Range ($\times 10^3$)	Average	Average ($\times 10^3$)
$[0]_8$	45	2985	2900 – 3025	N/A	N/A	6920	121.1
$[0]_8$	60	4150	3623 – 4580	116.5	114.1 – 121.5		
$[0]_8$	75	4887	4430 – 5300	N/A	N/A	Range	Range ($\times 10^3$)
$[0]_8$	90	5178	4910 – 5414	121.6	117.2 – 128.5	6625 – 7090	114.5 – 126.5
$[0]_8$	120	5392	5050 – 5520	120.9	113.4 – 128.3		

FLP

Table II

Variable Frequency Microwave Processed And Conventionally Cured Glass Fiber Thermosetting Prepreg Laminates, 8 Plies

Ultimate Tensile Strength (σ_B) vs Curing Time (CT)

DWG. NO. K/G 96-59R2 GSS

Center Frequency: 5.00 GHz
Frequency Range: 1.00 GHz

Forward Power: 180 – 190 watts
Reflected Power: 15 – 22 watts
Sweeping Rate: 100 ms

Ultimate Tensile Strength, σ_B (psi)

		Variable Frequency Microwaved		Oven-cured samples with post cure, cure time: >7 h, $[0/90]_{2S}$ - laminates	
Ply Orientation	Cure Time CT (min)	Average ($\times 10^3$)	Range ($\times 10^3$)	Average ($\times 10^3$)	Range ($\times 10^3$)
$[0/90]_{2S}$	60	58.6	54.5 – 65.4	64.0	60.3 – 69.3
$[0/90]_{2S}$	90	70.7	69.4 – 75.8		
$[0/90]_{2S}$	120	61.2	56.6 – 68.0		

Ultimate Tensile Strength, σ_B (psi)

		Ultimate Tensile Strength (in 0° direction), σ_B (psi)		Oven-cured samples (post cured) $[0/45/90/-45]_{2S}$ - laminates	
Ply Orientation	Cure Time CT (min)	Average ($\times 10^3$)	Range ($\times 10^3$)	Average ($\times 10^3$)	Range ($\times 10^3$)
$[0/45/90/-45]_S$	60	47.4	44.4 – 51.5	48.0	44.3 – 51.5
$[0/45/90/-45]_S$	90	45.1	42.4 – 48.7		
$[0/45/90/-45]_S$	120	47.0	44.5 – 49.9		

FLP

Table III

Variable Frequency Microwave Processed And Conventionally Cured Unidirectional Carbon Fiber Thermosetting Laminates, 8 Plies

Ultimate Tensile Strength (σ_B) vs Curing Time (CT)

Center Frequency: 5.00 GHz
Frequency Range: 1.00 GHz
Forward Power: 180 – 190 watts
Reflected Power: 9 – 15 watts
Sweeping Rate: 100 ms

DWG. NO. K/G 96-60R2 GSS

Ultimate Tensile Strength, σ_B (psi)

Variable Frequency Microwaved						Oven-cured samples (non-post cured) cure time: ≈ 4.5 h, $[0]_8$ - laminates	
		90° (Perpendicular to the fibers)		0° (Parallel to the fibers)		90° (Perpendicular to the fibers)	0° (Parallel to the fibers)
Ply Orientation	Cure Time CT (min)	Average	Range	Average ($\times 10^3$)	Range ($\times 10^3$)	Average	Average ($\times 10^3$)
$[0]_8$	45	3390	2890 – 4090	—	—	6567	304.3
$[0]_8$	60	4636	4470 – 4862	—	—	Range	Range ($\times 10^3$)
$[0]_8$	90	5441	5350 – 5490	—	—	6400 – 7520	295.3 – 312.3
$[0]_8$	120	5952	5578 – 6180	276.1	261 – 292		

$[0 / 90]_{2S}$ - Laminate	
Average ($\times 10^3$)	Range ($\times 10^3$)
166.7	157.0 – 176.0

$[0 / 45 / 90 / -45]_S$ - Laminate	
Average ($\times 10^3$)	Range ($\times 10^3$)
87.1	82.8 – 91.6

FLP

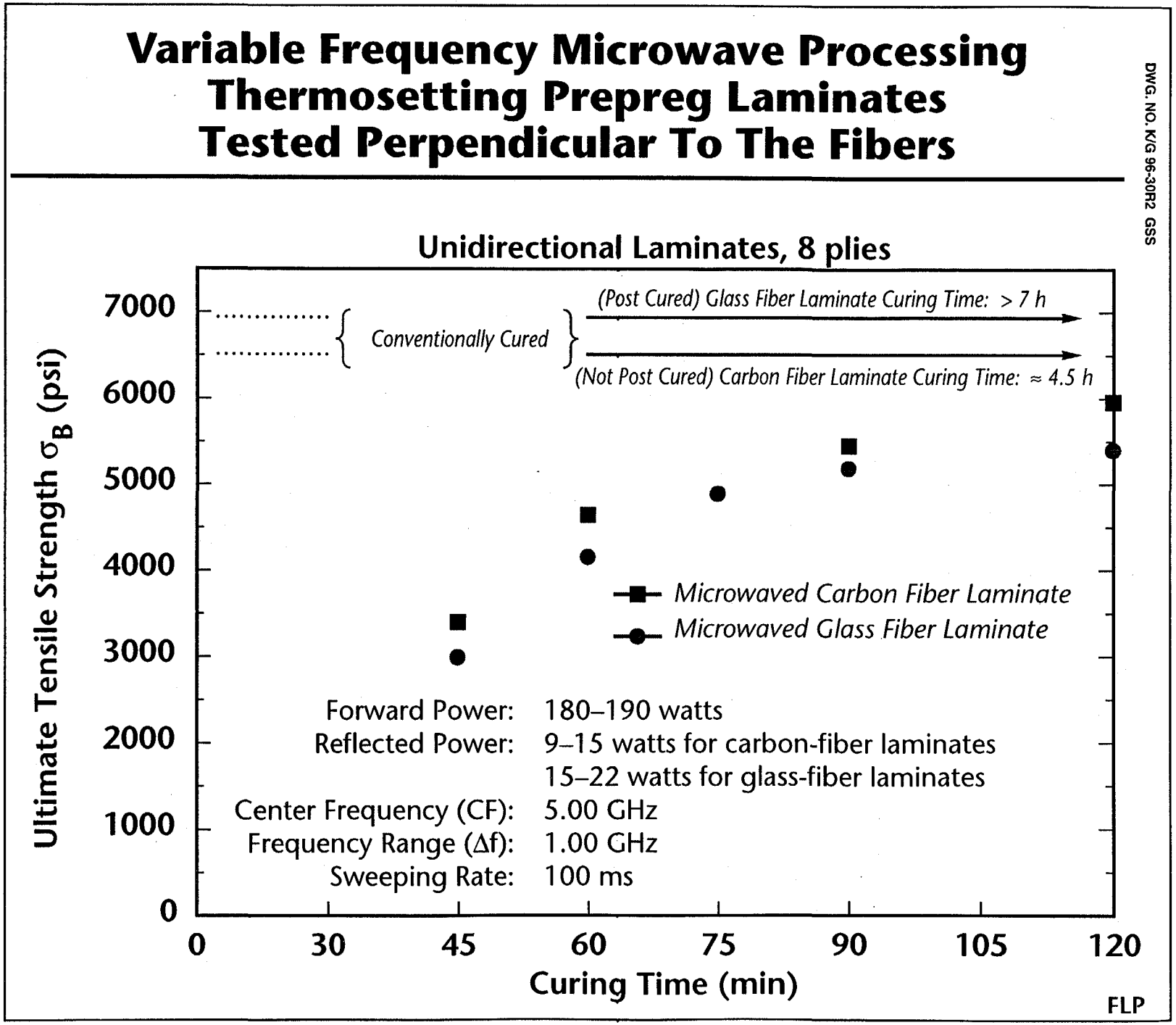
Variable Frequency Microwave Processing
Thermosetting Prepreg Laminates
Tested Perpendicular To The Fibers
DWG. NO. K/G 96-30R2 GSS
Unidirectional Laminates, 8 plies
Ultimate Tensile Strength σ_B (psi)
7000
6000
5000
4000
3000
2000
1000
0
Conventionally Cured
(Post Cured) Glass Fiber Laminate Curing Time: > 7 h
(Not Post Cured) Carbon Fiber Laminate Curing Time: ≈ 4.5 h
Microwaved Carbon Fiber Laminate
Microwaved Glass Fiber Laminate
Forward Power: 180–190 watts
Reflected Power: 9–15 watts for carbon-fiber laminates
15–22 watts for glass-fiber laminates
Center Frequency (CF): 5.00 GHz
Frequency Range (Δf): 1.00 GHz
Sweeping Rate: 100 ms
0
30
45
60
75
90
105
120
Curing Time (min)
FLP

Figure 13

Table IV

Variable Frequency Microwave Processed And Conventionally Cured Unidirectional Glass Fiber Thermosetting Laminates, 8 Plies

Ultimate Tensile Strength (σ_B) vs Curing Time (CT)

DWG. NO. K/G 96-461R GSS

Center Frequency: 13.80 GHz
Frequency Range: 1.00 GHz
Forward Power: 162 – 174 watts
Reflected Power: 1 – 2 watts
Sweeping Rate: 100 ms

Ultimate Tensile Strength, σ_B (psi)

		Variable Frequency Microwaved				Oven-cured samples with post cure, cure time: > 7 h, $[0]_8$ - laminates	
		90° (Perpendicular to the fibers		0° (Parallel to the fibers)		90° (Perpendicular to the fibers	0° (Parallel to the fibers)
Ply Orientation	Cure Time CT (min)	Average	Range	Average ($\times 10^3$)	Range ($\times 10^3$)	Average	Average ($\times 10^3$)
$[0]_8$	45	3160	3100 – 3358	N/A	N/A	6920	121.1
$[0]_8$	60	5357	5088 – 5506	N/A	N/A		
$[0]_8$	75	N/A	N/A	N/A	N/A	Range	Range ($\times 10^3$)
$[0]_8$	90	5595	5141 – 5882	N/A	N/A	6625 – 7090	114.5 – 126.5
$[0]_8$	120	5850	5401 – 6277	N/A	N/A		

FLP

Table V

Variable Frequency Microwave Processed And Conventionally Cured Glass Fiber Thermosetting Prepreg Laminates, 8 Plies

Ultimate Tensile Strength (σ_B) vs Curing Time (CT)

DWG. NO. K/G 96-460R GSS

Center Frequency: 13.80 GHz
Frequency Range: 1.00 GHz
Forward Power: 162 – 174 watts
Reflected Power: 1 – 2 watts
Sweeping Rate: 100 ms

Ultimate Tensile Strength, σ_B (psi)

Ply Orientation	Cure Time CT (min)	Ultimate Tensile Strength (in 0° direction), σ_B (psi) Average ($\times 10^3$)	Ultimate Tensile Strength (in 0° direction), σ_B (psi) Range ($\times 10^3$)	Oven-cured samples (post cured) $[0/45/90/-45]_{2S}$ - laminates Average ($\times 10^3$)	Oven-cured samples (post cured) $[0/45/90/-45]_{2S}$ - laminates Range ($\times 10^3$)
$[0/45/90/-45]_S$	60	46.3	45.1 – 47.5	48.0	44.3 – 51.5
$[0/45/90/-45]_S$	90	46.7	44.9 – 49.2		
$[0/45/90/-45]_S$	120	45.9	44.7 – 47.1		

FLP

Table VI

Variable Frequency Microwave Processed And Conventionally Cured Unidirectional Carbon Fiber Thermosetting Laminates, 8 Plies

Ultimate Tensile Strength (σ_B) vs Curing Time (CT)

Center Frequency: 13.80 GHz
Frequency Range: 1.00 GHz
Forward Power: 162 – 174 watts
Reflected Power: 1 – 2 watts
Sweeping Rate: 100 ms

Ultimate Tensile Strength, σ_B (psi)

Variable Frequency Microwaved						Oven-cured samples (non-post cured) cure time: ≈ 4.5 h, $[0]_8$ - laminates	
		90° (Perpendicular to the fibers)		0° (Parallel to the fibers)		90° (Perpendicular to the fibers)	0° (Parallel to the fibers)
Ply Orientation	Cure Time CT (min)	Average	Range	Average ($\times 10^3$)	Range ($\times 10^3$)	Average	Average ($\times 10^3$)
$[0]_8$	45	N/A	N/A	—	—	6567	304.3
$[0]_8$	60	4492	4298 – 4586	—	—	Range	Range ($\times 10^3$)
$[0]_8$	90	5804	5404 – 6265	—	—	6400 – 7520	295.3 – 312.3
$[0]_8$	120	6080	5961 – 6219	—	—		

$[0 / 90]_{2S}$ - Laminate	
Average ($\times 10^3$)	Range ($\times 10^3$)
166.7	157.0 – 176.0

$[0 / 45 / 90 / -45]_S$ - Laminate	
Average ($\times 10^3$)	Range ($\times 10^3$)
87.1	82.8 – 91.6

DWG. NO. K/G 96-459R GSS

FLP

in Figure 14, σ_B increased steadily up to a value below those obtained for conventionally cured samples. Once more, the carbon fiber laminates cured at 13.80 GHz and two hours yielded values of σ_B less than ten percent different than those samples cured conventionally. Evident is the fact that σ_B values obtained from microwaved glass fiber laminates improved by using this higher center frequency.

In Figures 15 and 16 are illustrated the effects of varying the center frequencies on the mechanical properties measured from the different types of laminates. Clearly, with the higher center frequency, the mechanical properties experienced in both types of laminates can increase. The reason for this is the increase in energy deposition associated to the higher energy photons incident on the laminates.

Figure 17 illustrates the effect of a variation in the frequency range (Δf) and forward power level on the mechanical properties of the unidirectional glass fiber laminates. Here, only resin properties were evaluated. To be able to explain the overlapping data in Figure 17, many possible factors should be taken into consideration. When the frequency is swept over a wider range or band, more instantaneous modes are occurring and consequently the accumulative effect of these modes will result in a better power homogenization or distribution in the workpiece. A wider frequency band increases also the probability that a better coupling frequency for this specific material will reside inside the new chosen frequency band. Nevertheless, the time-averaged real power in the cavity is slightly reduced with the utilization of a wider frequency band. The selected property for the evaluation of the advancement of the crosslinking process in this case was the ultimate tensile strength. This test is not sensitive enough to detect tiny or minuscule changes in material characteristics. The sum of all these factors may explain the overlapping of the data in Figure 17.

The data obtained with a frequency range of 1.00 GHz and a forward power of 115 to 120 watts depict a minimal variation in the values of the ultimate tensile strength over an extended range of the processing time. This can be explained based in the low forward power level applied to the sample. The applied power was unable to deposit or couple an adequate quantity of energy into the material to carry out the chemical reaction further to reach the required speed and level.

Figure 18 illustrates the T_g data from Figure 9 but at this time, this T_g data is associated to the corresponding ultimate tensile strength data of identical samples. As indicated in Figure 18, both curves follow each other in a steady and smooth fashion and show an increase as a function of the microwave processing time.

Similar to the results obtained with a fixed frequency (2.45 GHz) multimode cavity [1], VFM processing was ineffective for the proper curing of multidirectional carbon fibers. It is interesting to evaluate the processing of these laminates via hybrid curing. Through proper selection of the clamping plates (sample configuration, Figure 2) with suitable dielectric loss characteristics, this type of laminate may be processed when its thickness is limited such that a quasi-uniform thermal gradient can be achieved throughout the thickness of the sample.

Variable Frequency Microwave Processing Thermosetting Prepreg Laminates Tested Perpendicular To The Fibers

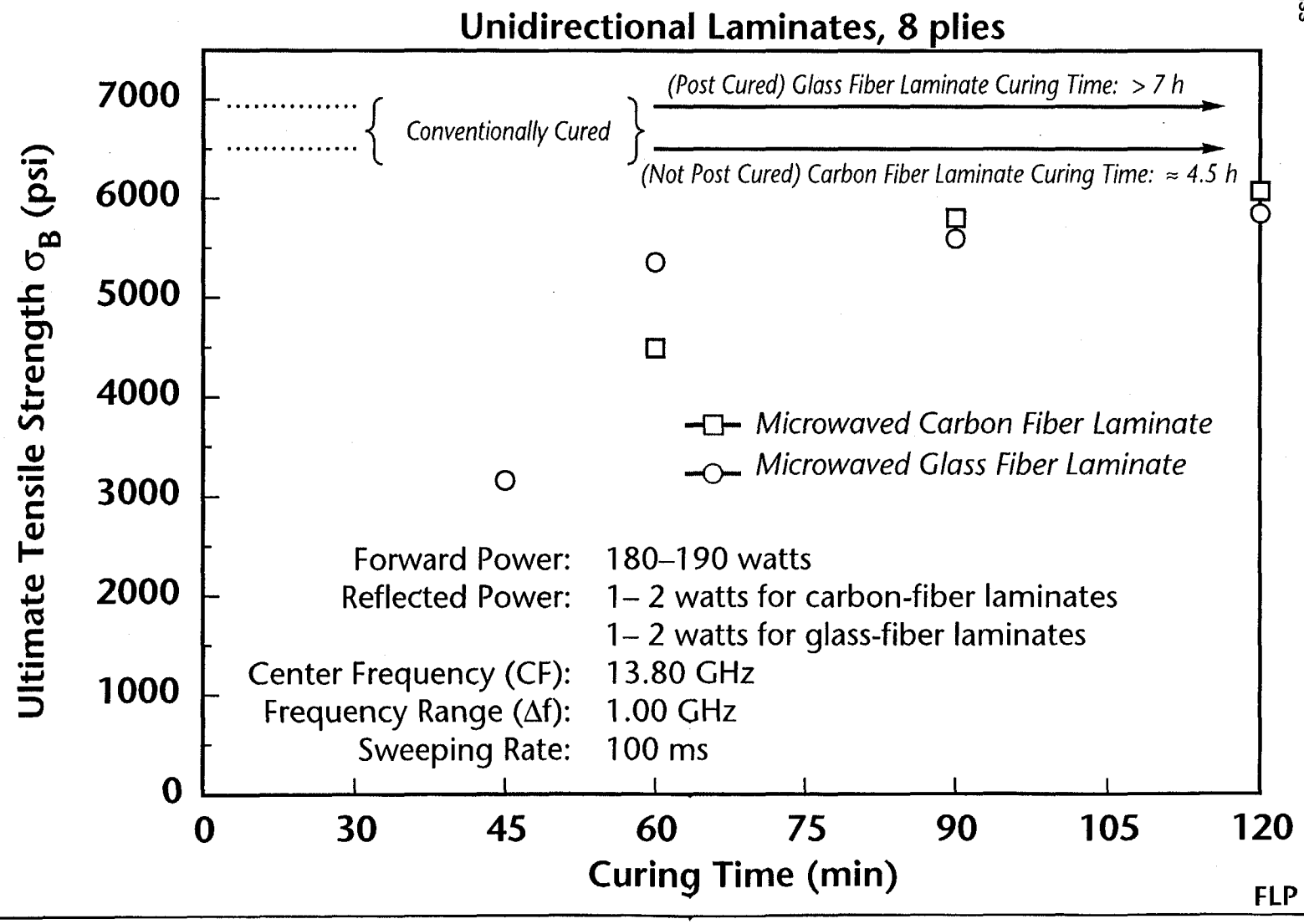

Figure 14

Variable Frequency Microwave Processing Glass Fiber Thermosetting Prepreg Laminates Tested Perpendicular To The Fibers

DWG. NO. K/G 96-466R GSS

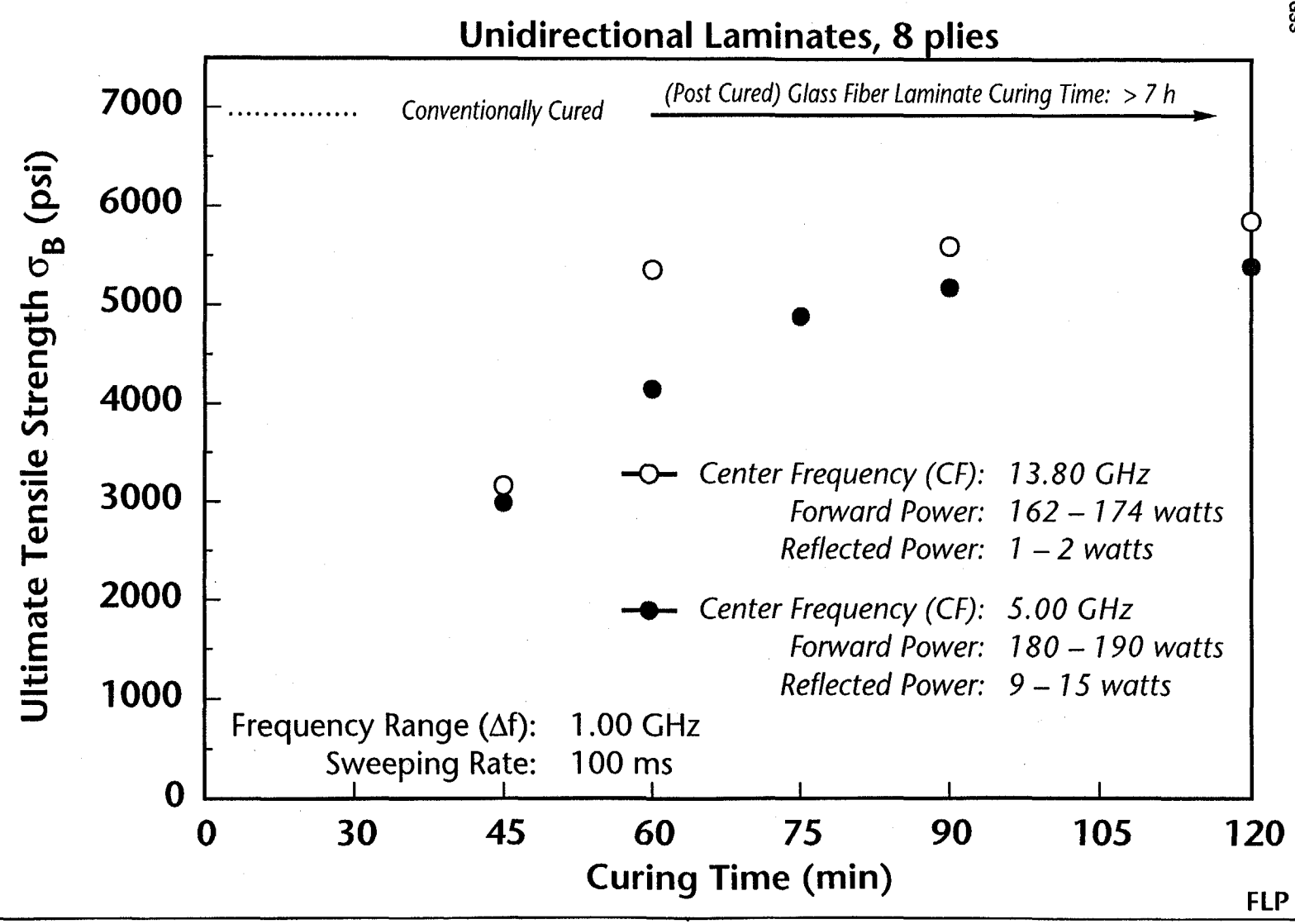

Figure 15

Variable Frequency Microwave Processing Carbon Fiber Thermosetting Prepreg Laminates Tested Perpendicular To The Fibers

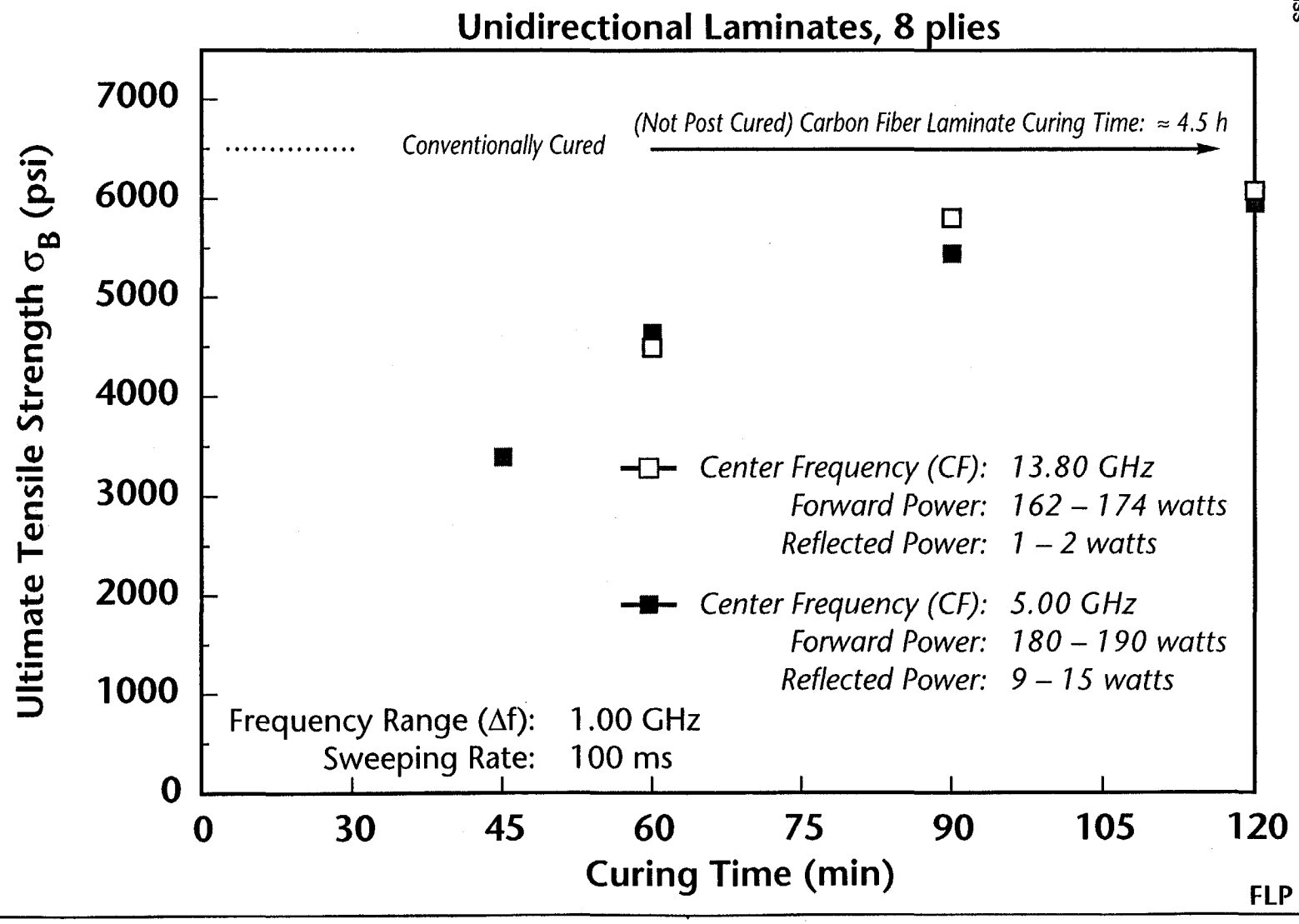

Figure 16

Variable Frequency Microwave Processing Thermosetting Prepreg Laminates Unidirectional Glass-Fiber Laminate, 8 Plies

DWG. NO. K/G 96-31R3 GSS

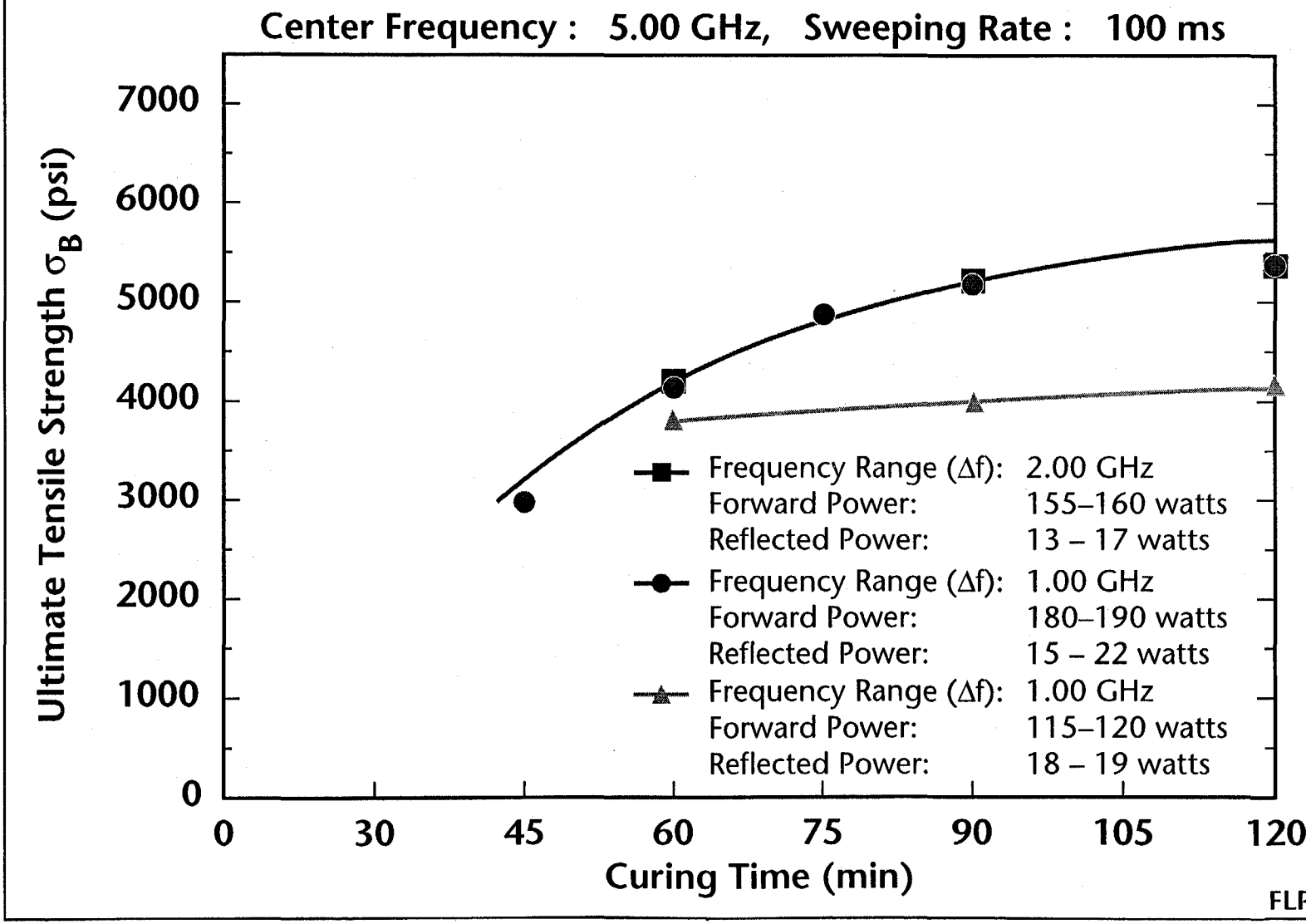

Figure 17

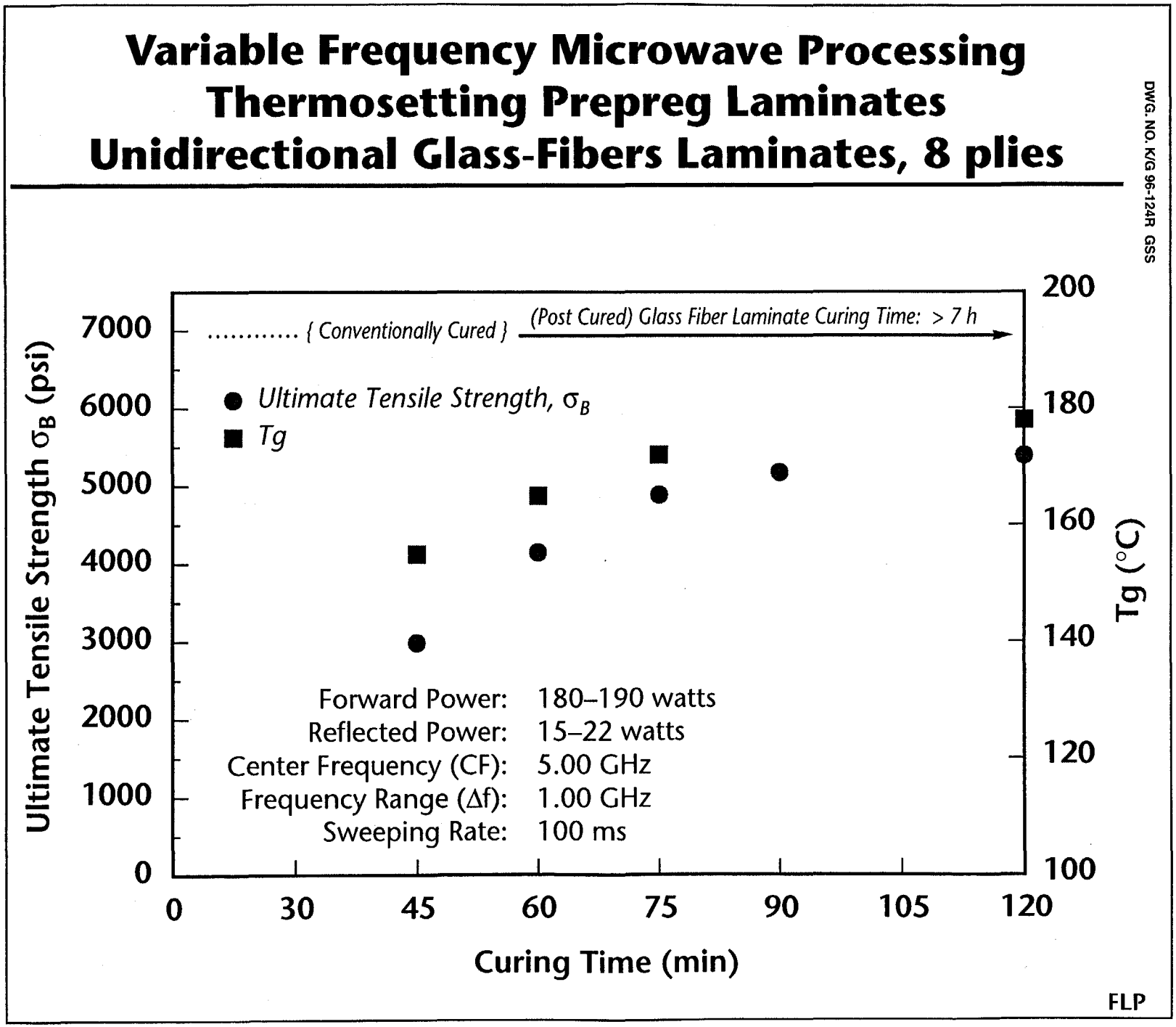
Variable Frequency Microwave Processing
Thermosetting Prepreg Laminates
Unidirectional Glass-Fibers Laminates, 8 plies
DWG. NO. K/G 96-124R GSS
..... { Conventionally Cured }
(Post Cured) Glass Fiber Laminate Curing Time: > 7 h
Ultimate Tensile Strength, σ_B
Tg
Forward Power: 180–190 watts
Reflected Power: 15–22 watts
Center Frequency (CF): 5.00 GHz
Frequency Range (Δf): 1.00 GHz
Sweeping Rate: 100 ms
Ultimate Tensile Strength σ_B (psi)
7000
6000
5000
4000
3000
2000
1000
0
Tg (°C)
200
180
160
140
120
100
0
30
45
60
75
90
105
120
Curing Time (min)
FLP

Figure 18

Table VII indicates the effect of an additional conventional postcure cycle on already microwave cured laminate samples. The ultimate tensile strength consistently experienced an increase with this additional postcuring cycle. In spite of the long microwave process time of 120 minutes, these results disclose a noncompleted microwave crosslinking process. The existing gap in the ultimate tensile strength required an additional treatment of the microwaved laminate.

This gap in the mechanical properties can be closed through: i) a longer process time with actual microwave equipment, resulting in a too long and unacceptable processing; ii) utilization of more powerful microwave equipment, capable of delivering higher forward power; iii) superimposing conventional heat at a desired curing temperature while at the same time exposing the sample to microwave radiation may accelerate the curing process and yield superior mechanical properties. The presence of the conventional thermal field should enhance the coupling of the sample to microwave radiation.

From the results obtained, the use of a variable frequency microwave applicator with higher power output at the higher possible frequency is strongly recommended for the processing of these laminates.

It is apparent that a direct comparison or correlation between variable frequency microwave and fixed frequency microwave technologies is difficult to establish. This is based on the fact that variable frequency microwave eliminates such weaknesses as hot-spots and heterogenous energy deposition into the workpiece. However, fixed frequency units are much less expensive and are available with much greater output power ratings. It is clear that an area of improvement in fixed frequency systems is the applicator design to be able to remove or reduce the effect of nonuniform field distribution (hot-spots).

4. CONCLUSIONS

Thermoset glass fiber reinforced laminates/composites can be effectively cured through exposure to variable frequency microwave radiation.

Variable frequency microwave heating can significantly reduce the curing/crosslinking time of unidirectional and multidirectional glass fiber/epoxy laminate up to 1/4 to 1/5 of conventional cure time.

Results indicate that variable frequency microwave curing caused no burning or hot-spots to appear on the processed laminates. This fact makes the technology very attractive for large or extended parts with complex geometries, e.g. shells. This is an improvement compared to fixed frequency microwave technology.

Tensile strength tests indicate that unidirectional and multidirectional glass fiber laminates processed by either variable frequency microwave, or conventional techniques exhibit values of the ultimate tensile strength approaching those of conventional curing techniques.

Table VII
Comparison Of The Ultimate Tensile Strength Of Unidirectional Laminates Tested Perpendicular To The Fibers On Microwave Processed Laminates And Subsequently With An Additional Conventional Postcuring Cycle Of 4 Hours At 350°F

Microwave Processing Conditions:

Frequency:	13.80 GHz	Frequency Range:	1.00 GHz
Forward Power:	162 – 174 watts	Reflected Power:	1 – 2 watts

Sweeping Rate: 100 ms

Glass Fiber Laminates:

a) Sample Number: G2 Microwave Cure Time: 90 min
   Ultimate Tensile Strength: 5595 psi
   – After an additional, conventional postcuring cycle,
   Ultimate Tensile Strength: 6922 psi

b) Sample Number: G5 Microwave Cure Time: 120 min
   Ultimate Tensile Strength: 5850 psi
   – After an additional, conventional postcuring cycle,
   Ultimate Tensile Strength: 7218 psi

Carbon Fiber Laminate:

a) Sample Number: C22 Microwave Cure Time: 120 min
   Ultimate Tensile Strength: 6080 psi
   – After an additional, conventional postcuring cycle,
   Ultimate Tensile Strength: 6313 psi

Straight Conventionally Cured Laminates:

a) Glass Fiber Laminate (with an extra postcuring cycle, total cure time: > 7h)
   Ultimate Tensile Strength: 6920 psi

b) Carbon Fiber Laminate (no postcuring cycle, standard cure time ≃ 4.5 h)
   – Ultimate Tensile Strength: 6567 psi

FLP

Unidirectional carbon fiber laminates demonstrated satisfactory coupling (curing) to 5.00 GHz and 13.80 GHz microwave radiation. Tensile strength tests of these samples also exhibit values approaching the conventionally cured samples with an observed difference of less than ten percent.

Although multidirectional carbon fiber laminates could not be directly processed through exposure to 5.00 GHz and 13.80 GHz variable frequency microwave radiation, a reduction from the conventional cure time may be achieved by hybrid curing. This alternate approach for curing laminates is only applicable to thin wall laminates.

The highly uniform variable frequency microwave energy distribution, evaluated on a time averaged basis, allowed these results to be obtained with a low real net power of approximately 160-170 watts, as compared to an industrial fixed frequency, multimode configuration of 1 to 2 kwatts. The power requirement is also low when compared with the nominal power required in conventional processing equipment, for example, rubber curing presses.

Variable frequency microwave processing of composite laminates with a higher center frequency will result in an increase of the ultimate tensile strength of the laminate resins. This is valid under the condition that all the remaining process parameters are kept unchanged. A variable frequency microwave applicator with higher output power at higher frequencies should reduce the curing time for these types of samples.

5. ACKNOWLEDGMENTS

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