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CONF-940411--29

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**Spring Materials Research Society (MRS) - Meeting in San Francisco, California, April 4 - 8, 1994**

Prepared by the
*Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831
managed by
Martin Marietta Energy Systems, Inc.
for the
U.S. DEPARTMENT OF ENERGY
under Contract No. DE-AC05-84OR21400

# PROCESSING OF THERMOSET PREPREG LAMINATES VIA EXPOSURE TO MICROWAVE RADIATION

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## ABSTRACT

Studies of microwave-assisted curing of neat resins (epoxy) and unidirectional glass and carbon fiber laminates have shown that a substantial reduction in the curing time was obtained [1]. This may be explained by the penetration of microwave energy directly and throughout the laminate with enhancement of the kinetics of the chemical reaction.

Results of this work indicate that the microwave assisted curing of glass fiber laminates also shows a substantial reduction of the required curing time.

Microwave radiation of 2.45 GHz has been demonstrated to be an acceptable method to cure unidirectional carbon fiber laminates. Also, effective curing of crossply (0/90) laminates through this method was observed when proper rotation of the parts accompanied the curing process. This is in accordance with previous work [2,3].

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 [4]. Nevertheless, a moderate reduction in the curing time of these thin laminates was observed due to hybrid curing.

## 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 cure and consolidate 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, 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.

In the preliminary stages of this program, experimental work was undertaken with ERL 2258 neat resin epoxy-based samples. (See Table I.) These preliminary 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 2.45 GHz radiation. 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* by using microwave radiation in the processing of neat resin. The results were in accordance with the fundamental theoretical hypothesis, that microwave energy could accelerate the curing (crosslinking) in polymers. These results indicate the feasibility of curing polymers and encourage further studies in the composite area.

In these 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 behavior with these two types of reinforcing fibers, can be observed. The 6 in. x 6 in. (15 cm x 15 cm) composite laminates samples for the initial experiments were UNIDIRECTIONAL and made of 8 piles of prepreg material. The material was processed at different power levels in a 2.45 GHz microwave furnace. Properties of similar laminated composites processed conventionally and with microwave radiation (in the 2 ft. x 2 ft. x 2 ft. microwave oven) were compared.

These results, similar to the neat resin, also indicated a remarkable reduction in the curing time for both types of composite laminates. The curing time with microwave radiation was reduced by a factor of approximately 5 to 6. A comparison in the ultimate tensile strength between samples of microwave processed and conventionally cured laminates indicated equivalent results. The range of values (in a specific direction relative to the fibers) of the microwave processed samples, in general, overlaps the values obtained for the conventionally cured samples.

This composite program was extended to study the feasibility of curing/crosslinking MULTIDIRECTIONAL laminate composites. For the multidirectional laminate study, the same composite base materials were selected for equal comparison with previous work.

## SAMPLE-PREPARATION

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 this location. See Figure 1.

For the processing, these multidirectional samples are placed in the ceramic hot press inside the microwave furnace (Figure 2, microwave processed samples) or between the metal plates of a standard heating press for conventionally processed samples. For the processing of the unidirectional laminates, the large 2 ft. x 2 ft. x 2 ft. cavity was used with the isostatic press located in the middle of this cavity.

In the multidirectional work, the final laminate samples were fabricated using eight adjacent layers of prepreg material with their fibers oriented differently relative to each other. Two basic configurations of fiber orientations were selected for the final carbon and glass fiber laminate samples:

| | | |
|---|---|---|
| $[0/90]_{2S}$ | : | 0/90/0/90 / 90/0/90/0 |
| $[0/45/90/-45]_S$ | : | 0/45/90/-45 / -45/90/45/0 |

This indicates the fiber orientation of each single layer of prepreg in the laminate.

## PROCESSING AND TESTING EQUIPMENT

A small fixed frequency, multi-mode cavity, with enhanced high-power-density microwave oven was used. This high-power-density microwave oven was developed by installing a small cavity inside of the 2 ft. x 2 ft. x 2 ft. (61 cm. x 61 cm. x 61 cm.) standard industrial microwave oven. With this reduction in the volume in the processing furnace, a higher electric field (power density) is obtained. 2.45 GHz microwave radiation was fed into the small cavity from a 5.5 Kw Cober SF6 power supply. This is the same power supply used throughout this program. With this experimental approach a possible additional reduction in the curing time of the laminates may be possible. See Figure 2. The dimension of this small oven was $2\lambda \times 2\lambda \times 2\lambda$, with $\lambda$ being a wavelength for 2.45 GHz frequency. ($\lambda$ = 12.24 cm). Similar to the 2 ft. x 2 ft. x 2 ft. oven, this small furnace (24 cm x 24 cm x 24 cm) was configured to allow the installation at the top and bottom of a ceramic ($Al_2O_3$) push rod for the application of the required pressure on the laminate during the curing process. Glass plates (pyrex or quartz) were used to distribute the pressure uniformly over the entire sample while allowing transmission of the microwave energy through these glass plates to the laminates.

Prior to the start of the testing program, a calibration check of the clamping force of the ceramic press inside of the furnace was undertaken. Microwave and conventionally produced samples were processed under equal specific pressure (10 to 11 psi). A leakage check in the microwave cavities (small and large ovens) was conducted. Here a Narda-Probe calibrated for 2.45 GHz frequency was used. For the evaluation of the ultimate tensile strength properties (ASTM D3039) a standard Instron testing machine, model 1125 was used.

## SAMPLE PROCESSING

For conventional processing, standard manufacturing curing sequence and time recommended by the material manufacturer was followed. The required cure time exceeded FIVE HOURS. 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.

For microwave processing, in order to obtain knowledge of the power density inside the cavity it was necessary to preevaluate this power distribution inside the microwave cavity. To accomplish this, a heat sensitive paper was placed between glass plates and microwave power was applied for the curing time that the samples required for one discrete step in the total curing process. These tests were conducted using, as close as possible, the final experimental configuration. These tests were repeated throughout the experimental program to confirm any change in power density in the cavity. This resulted in the generation of power maps for the microwave cavity used, and for the processing parameters applied. This study revealed that the surface covered by the 6 in. x 6 in. laminate surface was exposed to three well-defined hot spots inside of the sample surface.

To proceed with the microwave sample curing, samples were stacked according to Figure 1 and subsequently placed in between the push rods of the ceramic press located inside the microwave cavity. The microwave oven does not possess the capability to rotate the sample while being exposed to microwave radiation. The cavity power density calibration process indicated the necessity to rotate the sample during the curing process. A curing sequence was established, at discrete intervals at which the parts were rotated through a given angle.

Previous experimental work with unidirectional laminates indicated excellent reduction in the curing time of laminate samples exposed to microwave radiation. However, a close examination of these laminates showed a sticky consistency just barely at the sample edge and corners which may be an indication of incomplete curing of the material in this area. Additionally, due to the inability of a bench-scale experiment to determine with accuracy the exact moment in which the curing process has been completed, it was decided to lengthen the curing time by an additional ten minutes beyond the point that a visual observation indicated an acceptably cured sample.

For the specific case of multidirectional laminates, a total of eight glass fiber laminates were cured via microwave radiation, four of each glass fiber configuration. A total of five carbon laminates were processed via microwave, three of the $[0/90]_{2S}$ configurations and two of the $[0/45/90/-45]_S$ configuration. Two of each single fiber orientation/configuration for each type of fiber were processed using the conventional approach. This resulted in a total of eight laminate samples processed conventionally. For the unidirectional laminates, six acceptable laminates for each type of fiber were produced.

## RESULTS

Based on visual observation, the glass fiber laminates processed via microwave showed a darker color (brown) in the central region of the sample. This may be explained by the stationary condition of the sample center during the rotation sequence. Characterization of the power density in the small cavity indicates a moderate field intensity in this central area. Carbon-fiber laminates do not exhibit this darker central area due to the natural black color of this laminate.

The results indicate a remarkable reduction in the curing time for all the laminates when cured using microwave radiation. The curing time when using microwave radiation was reduced by a factor of five. See Table II and III.

In contrast to the results obtained with glass-fiber laminates, the carbon-fiber laminates did not exhibit substantial curing when exposed only to direct incidence of the 2.45 GHz microwave radiation. Despite this observed characteristic of these multidirectional carbon-fiber laminates, a moderate reduction in the curing/crosslink time was achieved through hybrid heating. It was observed that the pyrex glass plates used as a press were heated by microwave radiation resulting in the partial microwave, partial thermal curing of the carbon samples. See Table III. Advantages of this nature also has been observed in the hybrid processing of ceramic materials.

### Mechanical Properties

A comparison of the ultimate tensile strength between samples of microwave processed and conventionally cured glass fiber laminates indicates similar or equivalent results. These data were obtained from twelve tensile strength samples. These twelve samples were taken from two laminates of equal fiber configuration, six samples from each laminate. This will make these strength samples more representative of a single curing condition of the laminates. Results are presented in Tables IV and V. Unidirectional laminate mechanical strength data is shown in Table IV. This data shows that microwave cured unidirectional laminates (parallel and perpendicular to the fibers) are comparable to conventionally cured unidirectional laminates. Table V shows that the strongest glass fiber laminates were microwave cured; however, the range exhibited by the microwave cured $[0/45/90/-45]_S$ laminates was wider than for those processed conventionally. The difference was within the error of measurement. The range for the $[0/90]_{2S}$ laminates cured with microwave radiation was less than for those conventionally processed; however, the difference was again within the error of measurement. During these tensile strength tests, no differences were noticed in the rupture behavior of the glass fiber specimens.

In the specific case of the $[0/90]_{2S}$ glass fiber tensile strength samples, it was noted that rupture never occurred in the area corresponding to the central slightly darker area of the laminate; (fixed area with respect to the sample rotation). Additional study is recommended to be able to explain this observation.

To study the affect of high electric field intensity burn spots (hot spots) on the laminates, two samples were taken with these burn spots and tested under tensile test conditions. These samples show rupture initiation exactly from inside these burn regions, resulting in tensile strength that is significantly reduced. For example, on two samples $[0/45/90/-45]_S$, the measured tensile strength was 23.64 ksi and 23.26 ksi, as compared with the average tensile strength of 36 ksi.

No tensile strength testing was performed on samples from carbon fiber laminates because of the uncertainty of complete curing from direct exposure to the microwave radiation.

## CONCLUSIONS

- Glass fiber reinforced epoxy matrix laminates/composites can be effectively cured through exposure to microwave radiation.
- Microwave heating can significantly reduce the curing/crosslinking of unidirectional and multidirectional glass fiber/epoxy laminates up to 1/5 to 1/6 of conventional cure time.
- Tensile strength tests show that unidirectional and multidirectional glass fiber laminates processed by either microwave, or conventional techniques exhibit equivalent values of the ultimate tensile strength.
- Unidirectional carbon fiber laminates demonstrated satisfactory coupling (curing) to 2.45 GHz radiation. Tensile strength tests of these samples show comparable values to the conventionally processed samples.
- Although multidirectional carbon fiber laminates could not be directly cured through exposure to 2.45 GHz radiation, reduction of approximately 1/3 of conventional cure time was achieved by hybrid curing. This alternate approach for curing of the laminate is only applicable to thin wall laminates.
- It is clear that microwave processing of these laminates offers, in general, a faster processing alternative over conventional curing; however, also clear is the necessity to optimize the processing parameters which control the microwave curing process.

## ACKNOWLEDGEMENTS

This work was supported by funding provided by the Laboratory Directed R & D (LDRD) program at Oak Ridge National Laboratory (ORNL).

## REFERENCES

1. J. Mijovic and J. Wijaya, Polymer Composites 11 (3), 184 (1990).

2. J. Wei and M. C. Hawley, Sampe J., 11 (1), 33 (1991).

3. Y. F. Chen and Y. C. Lee, Proceedings of the American Chemical Society Division of Polymer Materials: Science and Engineering. 60, 680 (1989).

4. W. I. Lee and G. S. Springer, J. Composite Materials 18, 357 (1984).

**TABLE I. Comparison Between Microwave - Processed and Oven - Cured Thermoset Samples.**

- Resin: ERL 2258 (epoxy).
- Samples thickness: about 1 in.
- Microwave energy used: 2.45 GHz, 100 to 200 watts.
- The $T_G$ was measured by differential scanning calorimetry.

| | Microwave - Cured Samples | | Oven - Cured Samples |
|---|---|---|---|
| $T_G$ (°C) | 169 | Equivalent | 168 - 172 |
| Hardness (Rockwell 15T) | 73.2 | | 70.2 |
| Cure Time (minutes) | 30 | 10 - 12 times reduction | 360[1] |

[1]Two hours at 185°F followed by 4 hours at 300°F, plus additional time for heating and cool-down ramps.

**FIGURE 1. Industrial Microwave / Consolidation Cure Press.**

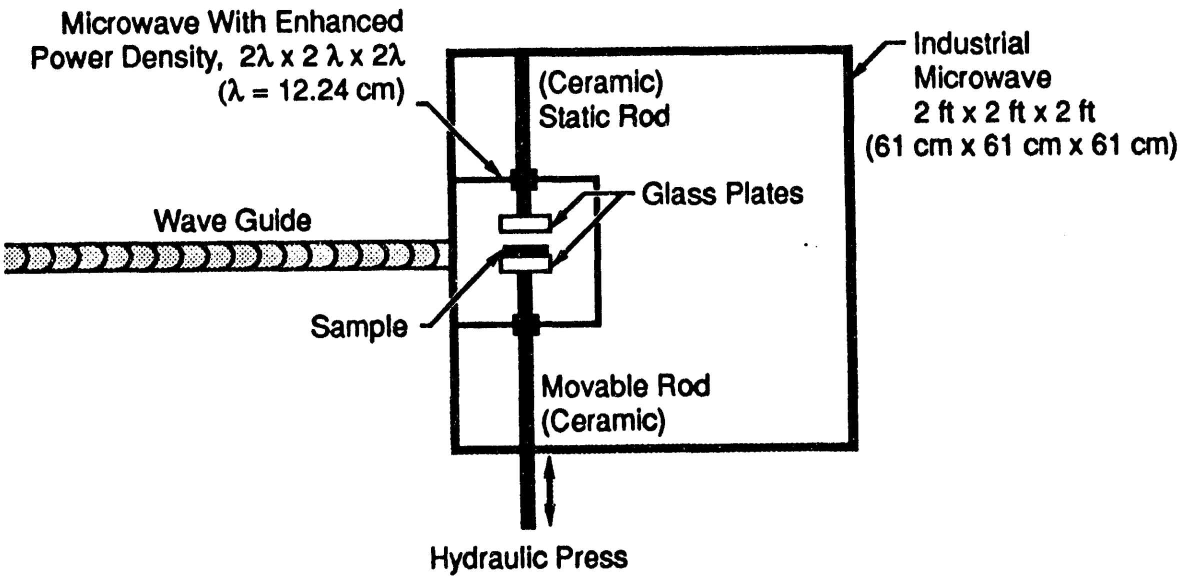

**FIGURE 2. Composite Laminate / Sample Configuration.**

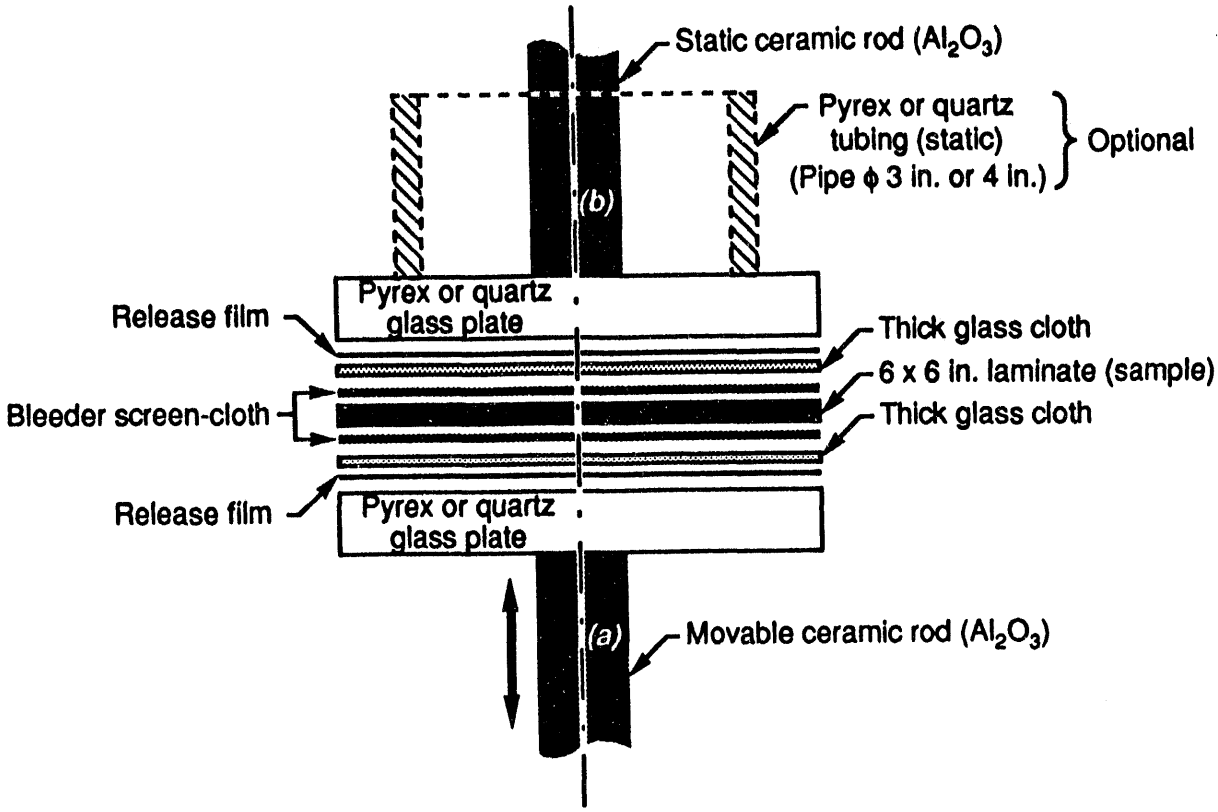


**TABLE II. Comparison Between Microwave - Processed and Oven - Cured Prepreg Laminate (Unidirectional).**

**CURING TIME, minutes**

| | Microwave - Cured Samples | Oven - Cured Samples |
|---|---|---|
| Carbon-fiber/Epoxy Laminate<br><br>(Hercules IM6G-3501-6)<br>Sample: 6 in. x 6 in., 8 plies | 40-80 min | > 330 min<br><br>(without post-cure) |
| Glass-fiber/Epoxy Laminate<br><br>(Scotchply 1003-UMI)<br>Sample: 6 in. x 6 in., 8 plies | 35 - 80 min<br>(200 min cured with very low power) | > 300 min<br><br>(without post-cure) |

**TABLE III. Comparison Between Microwave - Processed and Oven - Cured Multidirectional Prepreg Laminate.**

| CURING TIME, minutes | | |
|---|---|---|
| Multidirectional Laminates | Microwave - Cured Samples * | Oven - Cured Samples |
| Carbon-fiber/Epoxy Laminate<br><br>(Hercules IM6G-3501-6)<br>Sample: 6 in. x 6 in., 8 plies | $[0/90]_{2S}$: 95 - 105 min.<br><br>$[0/45/90/-45]_S$: 95 - 110 min. | > 330 min.<br><br>(without post-cure) |
| Glass-fiber/Epoxy Laminate<br><br>(Scotchply 1003-UMI)<br>Sample: 6 in. x 6 in., 8 plies | $[0/90]_{2S}$: 55 to 65 min.<br><br>$[0/45/90/-45]_S$: 55 to 65 min. | > 300 min.<br><br>(without post-cure) |

* 10 min. additional to assure total curing are included

**TABLE IV. Comparison Between Microwave - Processed and Oven - Cured Prepreg Laminate.**

- Microwave energy used: 2.45 GHz; 200 to 1000 watts
- Specific consolidation pressure: 10 - 11 psi.
- Number of layers in all samples: 8.
- Sample dimension: 6 in. x 6 in.
- Unidirectional laminates

| Laminates | | ULTIMATE TENSILE STRENGTH, psi<br>Microwave-Cured Samples | Oven-Cured Samples |
|---|---|---|---|
| Carbon-fiber/Epoxy Prepreg | 0° (parallel to the fibers) | $265 - 305 \times 10^3$ | $285 - 315 \times 10^3$ |
| (Hercules IM6G-3501-6)<br>Sample Thickness:<br>≈0.040 in., After Curing | 90° (perpendicular to the fibers) | 3400 - 3900 | 2800 - 4500 |
| Glass-fiber/Epoxy Prepreg | 0° | $125 - 133 \times 10^3$ | $120 - 130 \times 10^3$ |
| (Scotchply 1003-UMI)<br>Sample Thickness:<br>≈0.070 in., After Curing | 90° | 3400 - 4000 | 4400 - 4900 |

**TABLE V. Comparison between microwave-processed and oven-cured multidirectional prepreg laminate.**

- Microwave energy used: 2.45 GHz; 200 to 1000 watts.
- Specific consolidation pressure: 10-11 psi.
- Number of layers in all samples: 8.
- Sample dimension: 6 in. x 6 in.
- Multidirectional laminates
- Tensile strength samples taken at the 0°-direction of the fiber orientation

| Laminates | | | ULTIMATE TENSILE STRENGTH, ksi: Microwave-Cured Samples | ULTIMATE TENSILE STRENGTH, ksi: Oven-Cured Samples |
|---|---|---|---|---|
| Glass-fiber/Epoxy Prepreg (Scotchply 1003-UMI) Sample Thickness: ≈0.070 in., After curing | $[0/90]_{2S}$ | Average | 54.8 | 53.8 |
| | | Range | 52 - 58 | 50 - 58 |
| | $[0/45/90/-45]_S$ | Average | 36.5 | 36.2 |
| | | Range | 31 - 43 | 34 - 40 |

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