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**ALTERNATE ENERGY SOURCES
FOR
THERMOPLASTIC BINDING
AGENT CONSOLIDATION**

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Engineering Technology Division
Oak Ridge National Laboratory**

October 1998

**MANAGED AND OPERATED BY
LOCKHEED MARTIN ENERGY RESEARCH CORPORATION
FOR THE UNITED STATES
DEPARTMENT OF ENERGY**

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ABSTRACT

A study was conducted to investigate microwave and electron beam technologies as alternate energy sources to consolidate fiber coated with a thermoplastic binding agent into preforms for composite molding applications. Bench experiments showed that both microwave and electron beam energy can produce heat sufficient to melt and consolidate a thermoplastic binding agent applied to fiberglass mat, and several two- and three-dimensional fiberglass preforms were produced with each method. In both cases, it is postulated that the heating was accomplished by the effective interaction of the microwave or electron beam energy with the combination of the mat preform and the tooling used to shape the preform. Both methods contrast with conventional thermal energy applied via infrared heaters or from a heated tool in which the heat to melt the thermoplastic binding agent must diffuse over time from the outer surface of the preform toward its center under a thermal gradient. For these reasons, the microwave and electron beam energy techniques have the potential to rapidly consolidate thick fiber preforms more efficiently than the thermal process. With further development, both technologies have the potential to make preform production more cost effective by decreasing cycle time in the preform tool, reducing energy costs, and by enabling the use of less expensive tooling materials. Descriptions of the microwave and electron beam consolidation experiments and a summary of the results are presented in this report.

1. INTRODUCTION

A study was conducted to investigate microwave and electron beam technologies as alternate energy sources to consolidate fiber coated with a thermoplastic binding agent into preforms for composite molding applications. Bench experiments showed that both microwave and electron beam energy can produce sufficient heat to melt and consolidate the thermoplastic binding agent, and several two- and three-dimensional glass fiber preforms were produced with each method. Descriptions of the microwave and electron beam consolidation experiments and a summary of the results are presented in this report.

2. BACKGROUND

Thermoplastic binding agents are used in minute quantities to coat fiber reinforcements that are used in the manufacture of preforms. Typically, layers of the fiber reinforcement are placed in a tool and heated via ovens, infrared heaters, etc., to raise the temperature of thermoplastic binding agent above its softening point. A combination of heat and pressure applied to the fiber causes it to assume the shape of the tool. Removal of the heat source causes the binding agent coating the fibers to cool and solidify, yielding a fiber preform. The preforms are later placed in closed molds and infiltrated with resin to produce complex-shaped composite parts.

Although thermoset resins can also effectively bind (bond) fiber reinforcement plies together into a preform, this study focused on thermoplastic consolidation because of the greater processing flexibility and cost savings these materials confer to the composite manufacturer. Thermoplastic binding agents are tack-free ("dry") after application and therefore the coated fiber can be handled easily and cleanly in the manufacturing operation. Thermoset resins, on the other hand, can be wet or sticky, depending on the type and amount of coating applied to the fiber. Thermoplastic resins have an unlimited shelf life and can be stored indefinitely at room temperature while B-staged thermoset resins require refrigeration or freezer storage to prevent

cure advancement in the resin. Preforms fabricated with thermoplastic binding agents are still pliable (soft) for easier conformability to the mold cavity. Preforms fabricated with a thermoset binding agent can be rigid, depending on the amount and type of resin applied. Finally, because the thermoplastic bond can be reversed (de-bonded) with the application of heat, preforms consolidated with a thermoplastic binding agent can conceivably be reheated and repaired (adjusted) if necessary. Slight deformations or adjustments in preforms consolidated with thermoset resins may not be possible without the application of force and/or by breaking the preform.

The premise of this study is that melting of the thermoplastic polymer can be induced by the rapid deposition of electron beam and microwave energies into the fiber reinforcement. For example, many materials exposed to microwave energy undergo localized heating. This heating effect should cause the thermoplastic binding agent to melt and flow in a way that is comparable to the conventional thermal process. The heating levels achieved should depend largely on the degree to which the microwave energy successfully “couples” with the exposed material, and the power of the microwave unit. Microwave coupling efficiency with a material will depend on the frequency of the microwave energy as well as the material’s chemistry (composition).

Electron beam energy deposition into a material has also been demonstrated to produce localized heating of the substrate. The heating levels achieved depend on the energy and power level of the electron beam, the exposure time, and the material density, with high(er) density materials achieving higher temperature rises than low(er) density materials. Electron beam energy may also induce cross-linking in some polymers, increasing the material’s modulus, strength, and/or hardness.

3. EXPERIMENTAL

All experiments were conducted using a commercially available random fiberglass mat product obtained from Vetrotex Certaineed Fiberglass Reinforcements. The product description is Unifilo Continuous Strand Mat, designated as U750, and is produced from randomly dispersed

continuous strands of E-glass, held together with a thermoplastic binding agent. The mat is supplied in continuous 50 in. wide rolls and has an areal weight of 1.5 oz./sq. ft.

Technical literature obtained from Vetrotex Certaineed state that the mat should be formed during the viscous phase of the thermoplastic binding agent, which is at a temperature at or above 85°C (185°F). For a conventional thermal process, with heat applied from infrared heaters, the literature recommends that the mat layer temperature should be at least 120°C (248°F) at the inner core of layers and reach no more than 220°C (428°F) at the surface.

The process trials to consolidate the U750 mat with microwave and electron beam energy, and the results of these experiments are described in the following sections.

3.1 Microwave Energy Trials

Bench scale experiments were conducted to demonstrate the feasibility of consolidating the fiberglass mat into two- and three-dimensional shapes using microwave energy. Figure 1 is a photograph of some of the preforms that were prepared with these methods. It is postulated that the heating was accomplished in these experiments by the microwave energy coupling effectively with the combination of the fiberglass mat and tooling used to shape the preform. Based on available resources, the objective of the experiments was to demonstrate the viability of the technique (proof-of-principle) rather than optimization of the process for a production scenario.

The majority of the experiments were conducted using a T4000 Variable Frequency Microwave Oven that is maintained and operated by the Metals and Ceramics Division. Microwave exposures were conducted by sweeping across a range of microwave frequencies rather than exposing the sample to a single frequency. Advantages of the variable frequency method are that it can effect uniform heating over a large volume at a high energy coupling efficiency¹, thereby eliminating hot and cold temperature spots within the sample. Variable frequency processing can also provide precise frequency tuning to optimize the coupling efficiency with a specific material and make adjustments if the material properties (and coupling efficiency) change during prolonged microwave exposure.²

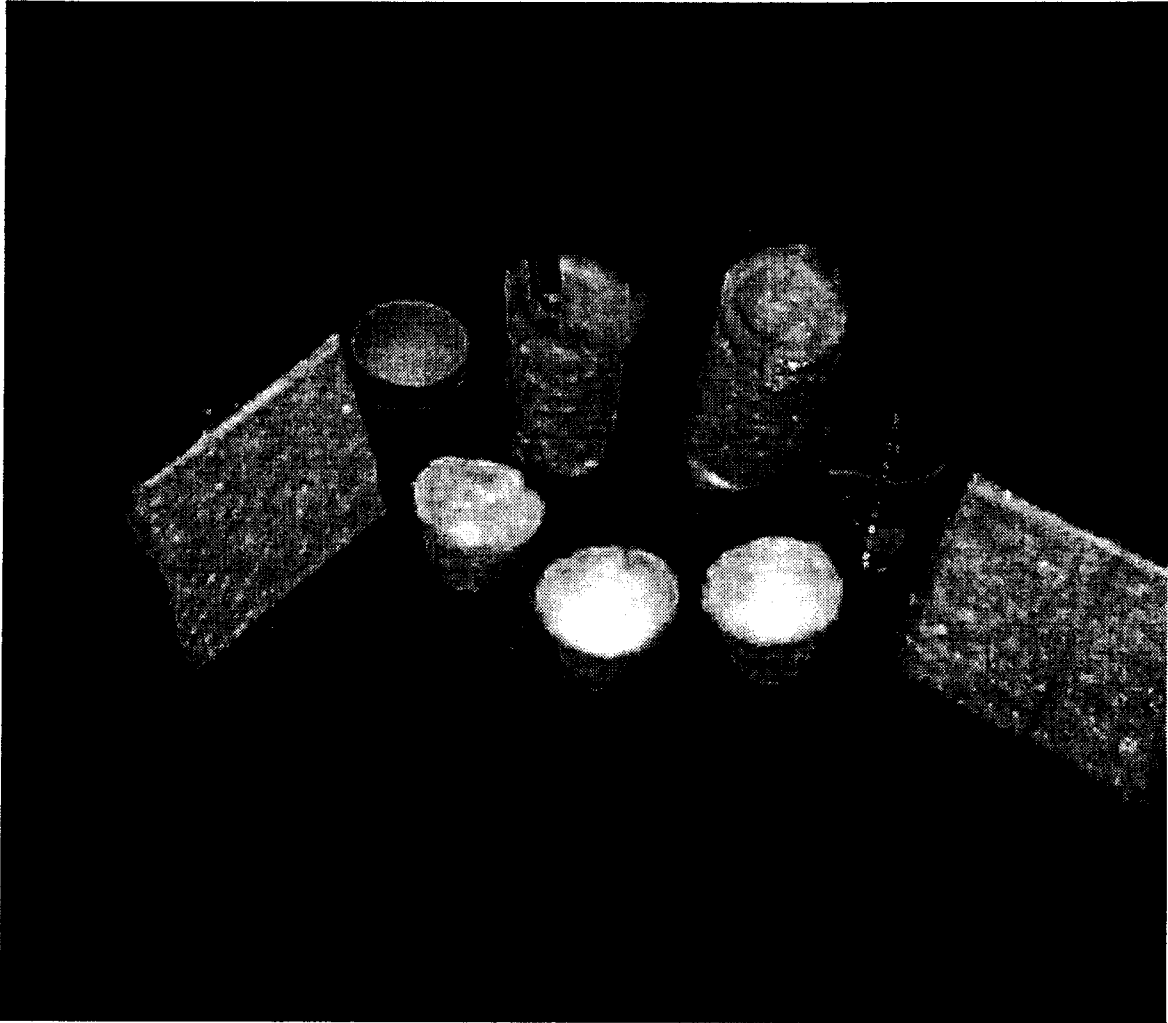


Figure 1. Fiberglass preforms consolidated with microwave energy

To consolidate the fiberglass mat into a preform, minimum pressure was required to ensure contact between the molten thermoplastic in adjacent fiber layers. In these experiments, pressure was applied to the samples during exposure via a piston actuated by approximately 23 to 30 psi air pressures. This was sufficient to ensure contact between the fiberglass layers as the thermoplastic-binding agent melted. After the microwave exposure was completed, the

sample cooled and the thermoplastic binding agent solidified, thereby bonding together the adjacent mat layers.

A limitation of microwave processing is that electrically conductive materials (such as metals) cannot be readily used as tooling materials because they will arc in the electric field. Depending on coupling efficiency, it is also possible to achieve very high temperatures rapidly during microwave exposure, and that would preclude the use of certain plastics and other low temperature materials for tools. With additional resources, characterization and process trials it is theoretically possible to identify tooling materials that are acceptable for this application, particularly if a microwave frequency can be identified that will couple selectively with the preform rather than the tool. To conserve resources, however, a decision was made to use only non-metallic materials capable of withstanding high temperatures ($>150^{\circ}\text{C}$). The tooling materials chosen for these experiments included quartz plates, Coors ceramic crucibles, Kimax glass beakers, and glass bottles that were all readily available from laboratory supplies.

The initial attempts to couple microwave energy with the fiberglass mat were conducted at the upper frequency range of the oven, in the vicinity of 12 to 15 GHz. Previous ORNL experience with fiberglass laminate samples suggested that these frequencies might also effectively couple with the mat. Fiberglass mat preforms were prepared by consolidating (fusing) layers of fiberglass mat with the following tooling and under the following conditions:

1. Nominal 5 in. x 5 in. area size fiberglass samples were prepared by placing 5 to 6 layers of mat between two 0.25 in. thick quartz plates and applying pressure via the microwave oven's air actuated cylinder. The samples were exposed to a microwave frequency of 13.78 GHz and 165 W, and with a frequency range centered about the median frequency of 1 GHz. (In other words, the sample was exposed to a frequency sweep from 13.28 GHz to 14.28 GHz). Exposure times were for 30 minutes, 1 hour, and 2 hours.

The sample exposed for 30 minutes did not consolidate, indicating that this microwave exposure was insufficient to raise the temperature of the thermoplastic binding agent above the melting point. However, the samples exposed for 1 and 2 hours did consolidate into nominal 0.16 to 0.18 in. thick preforms. The quartz plate surface temperature was nominally 75°C after the 1 hour exposure and 85°C after the 2 hour exposure, indicating some heating of the tooling material had occurred in these experiments.

2. Fiberglass mat (3 to 5 layers) was wrapped around the inside of a Coors porcelain crucible. The crucible size was nominally 2.5 in. diameter (top) x 2 in. high. A second crucible was nested within, with its OD pressed against the fiberglass layers. Pressure was applied to the two nested crucibles and the fiberglass lay-up via the microwave oven's air actuated cylinder. The samples were exposed for 3, 5, and 10 minutes to a microwave frequency range of 13.63 to 13.32 GHz and 130 W, with a frequency range centered about the median frequency of 0.25 GHz for the 3 minute exposure and 1 GHz for the 5 and 10 minute exposures. All of the samples fused together into 0.1 to 0.2 in. thick preforms, and with the shape and dimensions of the crucibles. In this set of experiments, the crucible material coupled significantly with the microwave frequency. The nominal crucible surface temperatures were 110°C, 126°C, and 133°C, respectively after the 3, 5, and 10 minute exposures.
3. Fiberglass mat (3 to 5 layers) was wrapped around the inside of a nominal 2.5 in. diameter x 3.5 in. high (250 ml) Kimax beaker. A 1.8 in. diameter x 4 in. high glass bottle was nested with its OD pressed against the fiberglass layers. Pressure was applied to the nested beaker, fiberglass, and bottle via the microwave oven's air actuated cylinder. The samples were exposed for 5 and 10 minutes to a microwave frequency of 13.63 GHz and 130 W, with a frequency range centered about the median frequency of 1 GHz. Both of the samples fused together into 0.1 to 0.2 in. thick preforms with the shape and dimensions of the beaker/bottle assembly. In this set of experiments, the glass bottle and beaker coupled significantly with the microwave frequency. The nominal bottle surface temperatures were 126°C and 161°C, respectively, after the 5 and 10 minute exposures.

Experiments were also conducted to assess the fiberglass mat's coupling efficiency with the microwave energy source, and **without** the benefit of additional heating arising from coupling of the microwave energy with the tooling material. A section of mat was jellyroll-wrapped into a nominal 1 in. ID x 2 in. OD x 5 in. long cylinder and exposed freestanding (no tool) for 10 minutes to a microwave frequency range of 13.25 to 13.6 GHz and 135 to 140 W, with a frequency range centered about the median frequency of 1 GHz. The mat's physical appearance did not change and the surface temperature climbed only slightly to ~35°C. It was concluded that the maximum power level of the T4000 oven (200 W) was insufficient to produce rapid heating of the fiberglass.

This experiment was repeated on freestanding fiberglass mat samples using the more powerful custom-built variable frequency microwave oven that is maintained and operated jointly by the ORNL Metals & Ceramics Division and Lockheed Martin Energy System's Development Division. A sample was exposed for various times up to 15 minutes to 4 to 8 GHz exposures at

250 W, 1000 W, and 2000 W, with a frequency range centered about the median frequency of 0.6 GHz.

These experiments were inconclusive because there was no reliable method to assess coupling of the fiberglass with the microwave energy source. Measurements made of the surface temperature of the mat after exposure were inaccurate because the open-weave fiberglass cooled too rapidly to get an accurate reading after exposure. Without applied pressure from a tool, it was impossible to get the mat layers to bond together and therefore it could not be determined if the thermoplastic binding agent had melted during exposure. It was obvious, however, that the fiberglass was getting hot during the 2000 W exposure because the thermoplastic binding agent turned brown at the ID of the jellyroll-wrapped cylinder due to thermal decomposition.

A HP8510A Network Analyzer was used to characterize the microwave energy coupling absorption spectrum for the studied fiberglass mat as a function of the microwave frequency. The purpose was to determine if a microwave frequency exists that couples directly with the fiberglass mat, and thereby can induce heating independently, or without the presence of an auxiliary coupling material (tool). The network analysis experiments indicated that the mat absorbs strongly at the frequency of 2.34 GHz. Unfortunately, the resources remaining for this study did not permit experimental verification of these results.

However, these results do indicate that the microwave consolidation process is economically viable for fiberglass preform production. That is because the indicated range of strong absorption indicated by the Network Analyzer is wide enough that the commonly used 2.45 GHz frequency is inside this region and also very close to the mat's maximum absorption peak of 2.34 GHz. Currently, 2.45 GHz is one of the least expensive frequencies in watts per dollar in the microwave market today. This fact makes the 2.45 GHz microwave processing frequency economically very acceptable for fiberglass preform production.

3.2 Electron Beam Energy Trials

Bench scale experiments were conducted to demonstrate the feasibility of consolidating fiberglass mat into two- and three-dimensional shapes using electron beam energy. Figure 2 is a photograph of some of the preforms that were prepared with these methods. Similar to the microwave energy process trials, it is postulated that the heating was accomplished in these experiments by the interaction of the electron beam energy with the combination of the fiberglass mat and the tooling used to shape the preform. As with the microwave process trials described in the preceding section, the objective of these experiments was to demonstrate the viability of this technique (proof-of-principle) rather than optimization of the process for a production scenario.

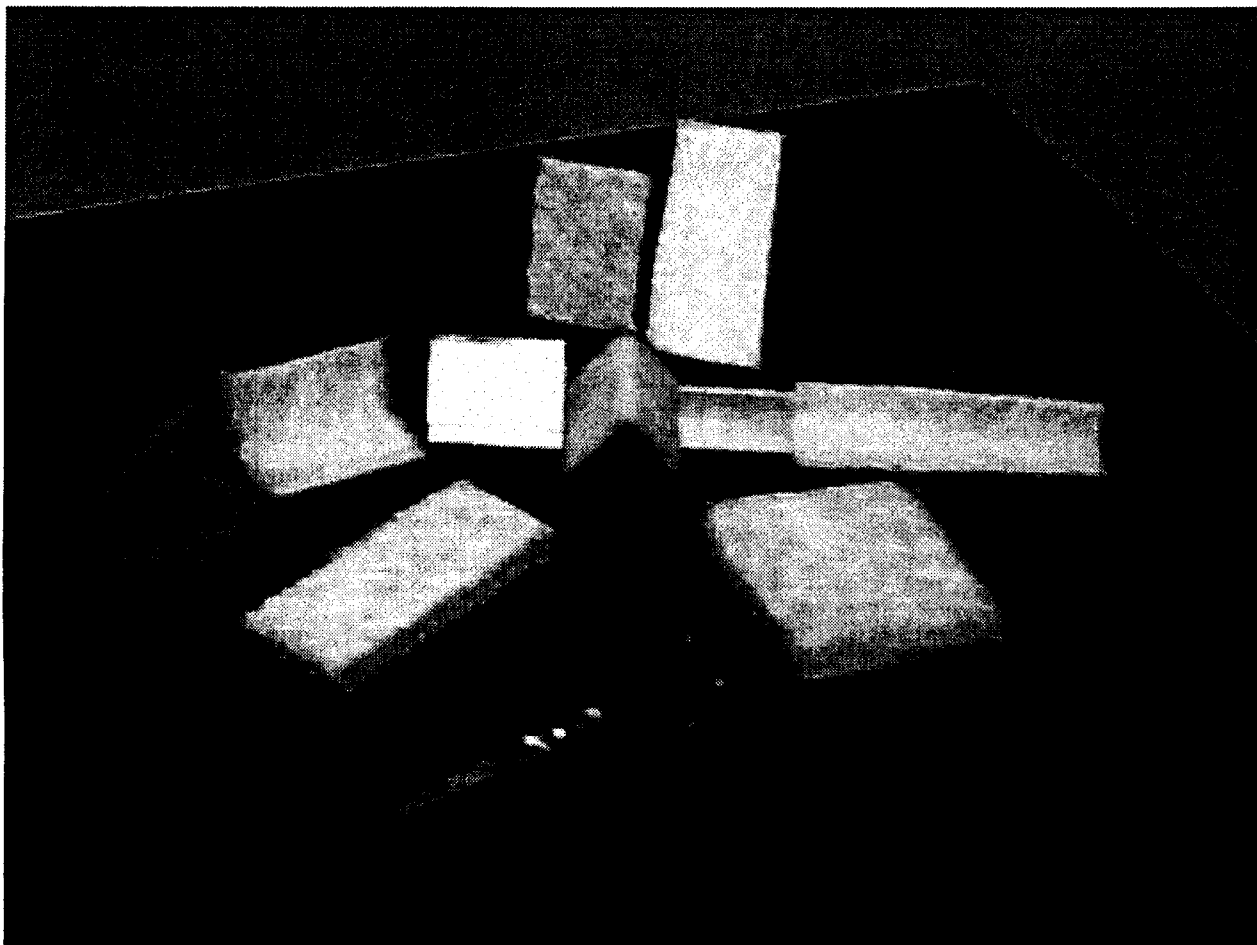


Figure 2. Fiberglass preforms consolidated with electron beam energy

All electron beam process trials were conducted using a 10 MeV, 1 kW Impella Accelerator which is owned and operated by Acision Industries, Inc. at the Atomic Energy Canadian Laboratory (AECL) facility, located in Pinawa, Canada. The irradiations were conducted by laying the samples on a tray, which was moved slowly back and forth under the electron beam by a conveyer belt. The samples were irradiated in multiple passes of 25, 50, or 75 kGy per pass until the desired total accumulated dose was achieved.

Electron energy is deposited in irradiated materials through a variety of interactions and mechanisms that depend on the material's composition. Materials with a high Z-number or density, such as metals, typically will show a greater temperature rise with electron irradiation than low-density materials. In this study, tooling materials with a range of densities and thickness were tested to assess their impact on fiberglass consolidation. The tooling materials included aluminum, ceramics, Styrofoam, glass, fiberglass composite and polyethylene plastic.

Preforms were prepared by pressing layers of fiberglass mat between two tooling surfaces and exposing the samples sequentially to two passes at 50 kGy per pass and two passes at 75 kGy per pass for a total dose of 250 kGy. The tools were pressed together with hand-tightened steel c-clamps in order to apply the minimum pressure necessary to ensure contact (and bonding) between the molten thermoplastic in adjacent mat layers. After irradiation, the samples were allowed to cool to ambient conditions prior to removal of the c-clamps.

In these experiments, it was observed that the fiberglass turned from a natural white to a light brown color as a result of the irradiation. The color change is attributed to a change in the light absorption spectrum brought about by the accumulation of electrons at electron traps in the fiberglass.

Examples of successful preform consolidation are presented below. In these experiments, electron beam energy was used to rapidly heat the tooling materials as well as the fiberglass mat, enabling the thermoplastic binding agent to melt and consolidate under pressure.

1. Preforms were prepared by sandwiching 3 to 4 layers of fiberglass mat between two sections of 2 in. x 2 in. x 3 in. long right angle aluminum channel, and with a wall thickness of nominally 0.25 in. Pressure was applied via c-clamps spaced evenly along the length of the channel. After irradiation, the 0.08 to 0.09 in. thick V shaped preform was well consolidated across its entire surface area.
2. Preforms were prepared with the same procedure described above except that the tooling consisted of two sections of 2 in. x 2 in. x 3 in. long right angle channel made from pultruded fiberglass composite. The channel wall thickness was nominally 0.25 in. After irradiation, the 0.08 to 0.09 in. thick V shaped preform was well consolidated across the entire surface area.
3. Preforms were prepared by sandwiching 3 to 4 layers of fiberglass mat between two 1 in. x 1 in. x 7.25 in. long right angle sections of aluminum channel, and with a nominal wall thickness of 0.125 in. Pressure was applied via c-clamps spaced along the length of the channel. After irradiation, the 0.08 to 0.09 in. thick V shaped preform was well consolidated across the entire surface area.
4. Nominal 3 in. x 6 in. area size fiberglass mat samples were prepared by placing 4 layers of mat between two 0.15 in. thick fiberglass composite sheets and applying pressure via c-clamps. This preform was nominally 0.11 in. thick and was well consolidated.
5. Fiberglass mat (4 to 5 layers) was wrapped around the inside of a Styrofoam coffee cup. A Coors porcelain crucible was nested inside the cup with its OD pressing against the fiberglass layers. The cup size was nominally 3 in. diameter (top) x 3.5 in. high and the crucible size was nominally 2.5 in. diameter (top) x 2 in. high. Pressure was applied to the nested cup and crucible via a c-clamp pressing together the bottom surfaces of the cup and crucible. Both the sides and the bottom of the cup-shaped 0.13 in. thick preform showed good consolidation after exposure. In this case, melting of the thermoplastic binding agent is attributed primarily to the heating of the porcelain crucible as a result of electron beam exposure, and transfer of the crucible's heat to the fiberglass mat via conduction.

Samples given the same 250 kGy dose but prepared with minimal mass and/or density tooling, and specimens of unsupported jellyroll-wrapped fiberglass mat were not successfully consolidated. Probable reasons include the inability of the electron beam to interact efficiently with the fiberglass mat to produce the temperatures necessary to melt the thermoplastic binding agent, and in some cases, insufficient pressure to effect a bond between the fiberglass plies. The following is a summary of these process trials.

6. Four layers of fiberglass mat were wrapped around the inside of an aluminum foil gel cup with nominal dimensions of 3 in. diameter (top) x 2 in. high x 0.006 in. wall thickness. A second gel cup was nested inside the first gel cup with its OD pressing against the fiberglass layers. Pressure was applied to the nested gel cups and fiberglass via a c-clamp pressing together the bottom surface of the cups.

After irradiation, the only fiberglass mat that was consolidated was from the location that was between the c-clamp pads between the bottom surfaces of the cups. The gel cup tooling was very flimsy, and it is speculated that the flexible (unsupported) sides of the cups did not provide sufficient pressure to effect a bond at the sides of the preform. There may have also been an additional positive heating contribution at the bottom of the cups due to the interaction of the electron beam with the high-density steel c-clamp pads.

7. Fiberglass mat (3 to 5 layers) was wrapped around the inside of a polyethylene cup that had the nominal dimensions of 3.25 in. diameter (top) x 3.75 in. high x 0.03 in. wall thickness. A second polyethylene cup was nested inside the first cup with its OD pressing against the fiberglass layers. Pressure was applied to the nested cups and fiberglass via a c-clamp pressing together the bottom surface of the cups.

After irradiation, the only fiberglass surface that was consolidated was from the location that was between the c-clamp pads during exposure between the bottom surface of the cups. It is speculated that there was no heating contribution from the low-density polyethylene cups' interaction with the electron beam. Insufficient contact pressure between the plies at the sides of the cup may have also been a problem with this experiment. Again, there may have also been an additional positive heating contribution at the bottom of the cups due to the interaction of the electron beam with the high-density steel c-clamp pads.

8. A section of mat was jellyroll-wrapped into a nominal 1 in. ID x 2 in. OD x 4 in. long cylinder and irradiated in a freestanding (no tool) position. The objective was to determine the effects of electron beam irradiation on the fiberglass mat, and without contributions from an auxiliary tooling material. Results of this experiment were inconclusive because of the lack of pressure to facilitate bonding between the layers. Other than the brown discoloration, the outside wraps of fiberglass were unchanged and were easily unwrapped (i.e. had not bonded together). The inside wraps were tacky and stuck together slightly during unwrapping of the jellyroll-wrapped sample. This process trial suggests that the fiberglass was heated during irradiation, and that the inner wraps had better heat retention (cooled less) due to insulation from the outer wraps.

Further experiments were conducted to determine whether the heating observed in the aforementioned preforms was a result of the interaction of the beam with the fiberglass mat or if

the tooling was the primary heating source. In these experiments, estimates were also made of the minimum electron beam dose required to melt the thermoplastic binding agent and consolidate the fiberglass plies into a preform.

Previous work had shown that without adequate pressure to bond the plies together, it was difficult to detect any evidence that the thermoplastic binding agent had melted during the electron beam exposures. That is because if the plies are not held closely in contact with one another, they separate once the sample cools back to ambient temperature. Therefore, fiberglass mat samples were prepared by sandwiching 4 to 5 layers of mat between 6 in. x 6 in. plates that were cut from 0.125 in. thick polyethylene sheet. Pressure was applied to the polyethylene plates and mat via c-clamps. The polyethylene plates, being made from a low-density material, could be expected to be relatively transparent to the electron beam while still being rigid enough to maintain pressure on the fiberglass mat during irradiation.

The samples were exposed to 25, 50, and 100 kGy in 25 kGy per pass steps to determine the minimum exposure threshold at which the mat would heat and consolidate into a preform. For comparison, samples pressed between 0.125 in. thick and 0.25 in. thick aluminum plates were also exposed in these experiments to assess the impact of the tooling on mat heating efficiency. A thermocouple was used to monitor the temperature of the fiberglass in the samples irradiated to 125 kGy. Figure 3 is a plot of the temperature with time and dose results for the three types of tooling.

The results showed that with only 25 kGy exposure, none of the fiberglass mat samples consolidated into preforms. With 50 kGy exposure, the samples pressed between 0.125 in. thick and 0.25 in. thick aluminum plates consolidated, but not the samples pressed between the polyethylene plates. After a dose of 125 kGy, there was partial consolidation in the sample that had been pressed between the polyethylene plates, indicating that some heating occurred in the fiberglass mat. The preforms prepared with the 0.125 in. thick and 0.25 in. thick aluminum plates were well consolidated and nominally 0.100 to 0.125 in. thick. The combination of results indicate that heating and consolidation of the mat is partly a function of the fiberglass's

interaction with the beam, but there is also a significant heating contribution from the electron beam's interaction with the high density aluminum plates.

Figure 3 shows that the mat temperature for the sample pressed between the polyethylene plates was only 80°C after the 125 kGy dose (5th pass) while the temperature of the mat sandwiched between the 0.125 in. thick and 0.25 in. thick aluminum plates were both greater than 120°C. Because the consolidated mat thickness was only 0.100 to 0.125 in., the contribution from heat diffusing from the aluminum plates into the mat is believed to be the significant factor in the consolidation of these preforms.

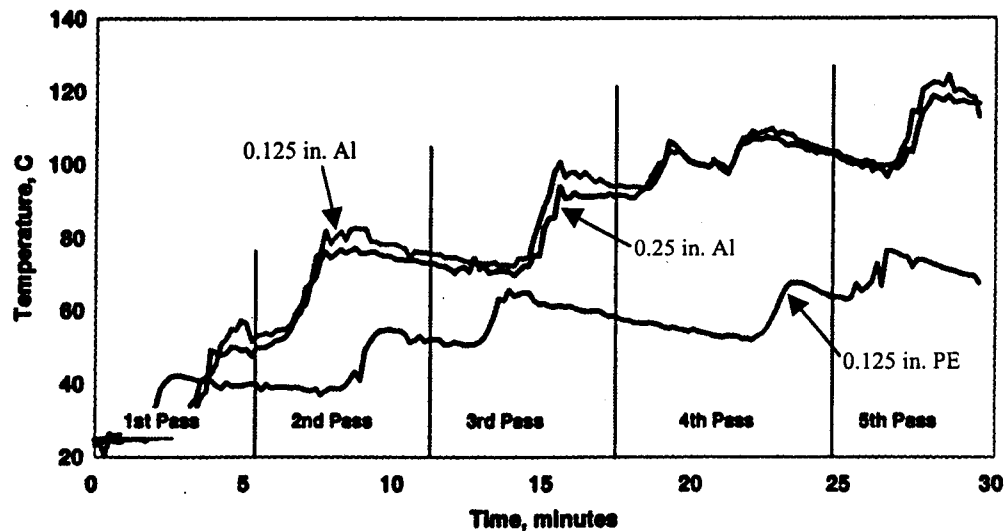


Figure and data provided by Acsion Industries Incorporated.

Figure 3: Fiberglass mat temperatures with time and dose (25 kGy per pass)

However, an intriguing result was obtained when this experiment was repeated with thicker preforms. Fiberglass mat samples that were 0.75 in. and 0.9 in. thick were clamped between 0.25 in. thick aluminum plates, and were successfully consolidated with an electron beam dose of three passes at 50 kGy per pass, or a total of 150 kGy. Due to the insulating characteristics of the fiberglass mat, it is believed that consolidation at the center of the preform wall thickness was also a result of the electron beam's interaction with the fiberglass, as opposed to only heat diffusion from the aluminum tool.

These results are exciting because they demonstrate that a thick volume of preform can be rapidly and simultaneously heated and consolidated with electron beam energy. For thick preforms, this process offers the potential to be more rapid, uniform, and use less energy than the conventional thermal processes whereby heat must diffuse from radiant heaters, a heated tool, etc., from the outside surface of the preform to its center. Risks associated with a conventional thermal process such as overheating (thermally degrading) the binding agent at the hottest points (the outside surface in contact with the tool) and/or insufficient consolidation/bonding of the inner plies due to poor thermal diffusion through the preform should also be minimized.

4. TECHNOLOGY ASSESSMENT

Experiments conducted during the course of this study have successfully demonstrated that both microwave and electron beam energy can produce heat sufficient to melt and consolidate a thermoplastic binding agent coating applied to fiberglass mat. For both technologies, it is postulated that the heating is accomplished by effective interaction of the microwave or electron beam energy with the combination of the mat preform and/or the tooling used to shape the preform.

Microwave and electron beam energy consolidation contrast with the conventional thermal process in which the energy to melt the binding agent is applied via infrared heater banks or a heated tool and must diffuse with time from the outer surface of the preform toward its center under a thermal gradient. Raising the internal temperature of a thick preform with an external heat source applied to the outside surface can be difficult because there is a time lag associated with the diffusion of heat from the outer to the inner portions of the preform. The time to heat the entire preform thickness depends on the temperature gradient between the inner and outer portions, and the thermal conductivity of the material. The task is made more difficult because a dry fiber preform conducts heat very poorly because of the large volume of air enveloped between the fibers.

Methods for improving (reducing) processing times with a conventional thermal process are limited. One method is to increase the magnitude of the thermal gradient to decrease the heat up (and production) times for thick preforms, but there are two drawbacks with this method. The first is that thermoplastic binding agents require careful temperature control in order to reach the minimum melt temperature of the center of the preform without exceeding the upper temperature limit of the polymer at the outer surface. Too high a temperature applied to the surface may degrade (char) the binding agent and/or damage the preform. The other drawback with increasing the outer surface temperature is that it requires more energy and possibly higher temperature tooling materials. Both factors can increase expenses in a high volume production process.

By contrast, microwave energy has the potential to penetrate and couple with the entire through thickness of the preform material immediately and instantaneously, thereby eliminating the thermal gradient and possible hot and cold spots through the preform. Similarly, electron beam energy is capable of instantaneous and deep penetration into a material substrate. Both processes have the potential to more rapidly and efficiently consolidate thick fiber preforms than the conventional thermal process. The risks associated with the thermal process such as overheating (thermally degrading) the binding agent at the hottest points (the outside surface in contact with the tool) and/or insufficient consolidation/bonding of the inner plies due to poor thermal diffusion through the preform should be reduced with these methods.

The information obtained as part of this study does not indicate which of the two consolidation technologies (microwave or electron beam energy) is better, or would be preferred from a manufacturing standpoint. The strengths and weaknesses of each method will require an assessment for each application. Some key differences between microwave and electron beam consolidation are listed below.

Facility and Capital Equipment Costs Electron beam facilities have relatively high facility and capital equipment costs associated with the cost of the accelerator as well as the need for shielding, radiation monitoring, etc. In some cases, these costs

may be shared by arrangements in which an electron beam facility is jointly owned and operated by various partners. Alternatively, a manufacturer may purchase electron beam time from an existing facility. Microwave ovens, by contrast, are relatively less expensive and have lower facility costs.

Tooling Options Microwave technology has more limitations in terms of the types of materials that can be exposed to microwave energy, primarily because electrically conductive materials will arc in the electric field. This narrows the options for using common tooling materials such as aluminum and steel, as well as for processing certain types of conductive fiber into preforms, including carbon fiber. With characterization and process trials, it is theoretically possible to identify tooling materials that are acceptable for this application, particularly if a microwave frequency can be identified that will couple selectively with the preform rather than the tool. Electron beam irradiation, on the other hand, can be easily conducted on a wide range of materials including electrically conductive materials such as metals, carbon fiber, etc. It is this versatility that might give electron beam processing an advantage over microwaves in certain applications.

5. CONCLUSIONS

Experiments conducted during the course of this study have successfully demonstrated that both microwave and electron beam energy can produce heat sufficient to melt and consolidate a thermoplastic binding agent coating applied to fiberglass mat. For both technologies, it is postulated that the heating is accomplished by effective interaction of the microwave or electron beam energy with the combination of the mat preform and/or the tooling used to shape the preform. Although feasibility has been demonstrated by this study, further research will be required to characterize these techniques, understand their limitations and refine them for a production scenario.

Experiments performed to characterize the microwave energy coupling absorption spectrum for the studied fiberglass mat as a function of the microwave frequency indicate that the mat absorbs strongly at the frequency of 2.34 GHz, which lies close to the range of the commonly used 2.45 GHz frequency. The 2.45 GHz frequency is one of the lower cost frequencies in watts per dollar in the microwave market today, which makes this processing frequency economically competitive for fiberglass preform production.

The electron beam irradiation experiments indicated that more effective consolidations are achieved by using a high density (such as aluminum) tool. Heating and consolidation in thinner samples is partly a function of the fiberglass interaction with the beam, but there is also a contribution from the heat produced by the interaction of the electron beam with the higher density aluminum plates. Fiberglass mat samples clamped between (low-density) polyethylene plates did not heat and consolidate as well as the samples clamped between aluminum plates for a given electron beam dose.

Thick preforms have been successfully consolidated in this study with electron beam energy. Due to the insulating characteristics of the fiberglass mat, heating at the center of the fiberglass volume and away from the tool surface is believed to be a result of both the electron beam's interaction with the fiberglass as well as the aluminum tool. The minimum dose required to consolidate 0.75 in. and 0.9 in. thick preforms clamped between 0.25 in. thick aluminum plates was on the order of 150 kGy.

Both electron beam and microwave technologies have the potential to make preform production more cost effective than conventional thermal processing by decreasing cycle time in the preform tool, reducing energy costs, and by enabling the use of less expensive tool materials. In particular, these consolidation technologies offer advantages in the production of thick preforms. Beneficiaries of these consolidation technologies are industries that require large quantities of mass-produced fiber preforms for composite molding. These include the automotive industry as well as other producers of molded fiberglass products.

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REFERENCES

1. F. L. Paulauskas, A. D. McMillan, & C. D. Warren, "Adhesive bonding Via Exposure to Variable Frequency Microwave Radiation," Proc. MRS Microwave Processing of Materials V Symposium, (Vol. 430), San Francisco, CA, 1996.
2. F. L. Paulauskas and T. T. Meek, "Processing of Thermoset Prepreg Laminate via Exposure to Microwave Radiation," MRS Microwave Processing of Materials IV Symposium, (Vol. 347), San Francisco, CA, 1994.
3. C. J. Janke, et. al., "Electron Beam Curing of Polymer Matrix Composites," ORNL/TM6115, Oak Ridge, TN, May 1997.

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