Joining of Thirty Three Percent by Weight Random Glass Fibre Reinforced Polystyrene Using Variable Frequency Microwave

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ABSTRACT: This paper extends the range of applications for Variable Frequency Microwave (VFM) (2 – 18 GHz) facilities to joining thirty three percent by weight glass fibre reinforced polystyrene composite [PS/GF (33%)]. With a given power level, the composite was exposed to various exposure times to microwave irradiation. The primer or coupling agent used was 5-minute two-part adhesive containing 100% liquid epoxy and 8% amine, i.e. Araldite, which was more readily microwave reactive than the composite itself. Bond strengths of the joints were lap shear tested and results were compared with those obtained using fixed frequency (2.45 GHz) microwave processing. The VFMF was operated under software control, which provided automatic data logging facilities. The maximum lap shear bond strength of joint was 430 N/cm^2 using variable frequency microwave facility while that obtained by fixed frequency microwave configuration was only 331 N/cm^2. The former is nearly 30% stronger than the latter.

Keywords: variable frequency microwaves (VFM), glass fibre-reinforced polystyrene, lap shear bond strength, microwave irradiation and Araldite

1. INTRODUCTION

Industrial applications of microwaves are relatively new technology. Factors that hinder the use of microwaves in materials processing are declining, so the prospects for the development of this technology seem to be very promising [1]. The mechanisms that govern the energy distribution process during microwave joining of materials include dipole friction, current loss and ion jump relaxation. This results in a relatively uniform heat distribution throughout the entire exposure to microwave irradiation, immediately in front of rectangular or circular waveguides [2-5]. The fast heating rate encountered using microwave energy can thus lead to reduced processing time and consequent energy efficiency. These advantages have encouraged the development of facilities for joining a range of thermoplastic composites autogenously and heterogeneously. In the heterogeneous mode, at room temperature, transparent materials, including a range of thermoplastic and thermosetting resins can be bonded using two part adhesives cured at fast rates when exposed to focused microwave irradiation [3,5,6].

In conventional microwave processing, microwave energy is launched at a fixed frequency of either 915 MHz or 2.45 GHz or 5.8 GHz or 24.125 GHz into a waveguide or cavity and it brought with it the inherent heating uniformity problems like hot spots and thermal runaway [7,8]. Thermal runaway is the uncontrolled rise in temperature in some hotter parts of a material subject to microwave heating. This is because the hotter parts will absorb more microwave energy than any other part of the material and convert it into heat [2]. A US based company developed a new technique for microwave processing, known as variable frequency microwave (VFM) technique, to solve the problems brought about by fixed frequency microwave processing. The technique was geared towards advanced materials processing and chemical synthesis. It offered rapid, uniform and selective heating over a large volume at a high energy coupling efficiency. This was accomplished using preselected bandwidth sweeping around a central frequency employing tunable sources such as travelling wave tubes as the microwave power amplifier. Selective heating of complex samples and industrial scale-up were now viable [8,9]. Successful applications have been reported in the areas of curing advanced polymeric encapsulants, rapid processing of flip-chip underfills, materials characterisation, curing profiles for various adhesives, structural bonding of glass to plastic housing [9,10].

When microwave energy of a fixed frequency, eg 2.45 GHz was launched into a waveguide e.g. WR340, as depicted in Figure 1(a), containing a piece of material, some areas of the material would experience higher electric field strength than the others; the situation would even be more serious if the microwave energy was launched into a multimode cavity because many resonant modes will be established. Figure 1(b) shows the fixed electric field pattern across any cross section of the joint of the test pieces during fixed frequency heating. Those areas with higher electric field strength would be heated more, creating hot spots, which could even lead to thermal runaway. With variable frequency microwave heating [9], as shown in Figure 2(a), more than one thousand frequencies were launched into the cavity sequentially. Each incident frequency set up its own electric field pattern across any cross section of the joint of the test pieces, and therefore resulted in hot spots at different locations at different time, as shown in Figure 2 (b). Different areas were heated under different frequencies at different times. When a sufficient bandwidth was used, every element of the test piece would experience hot spots at one or more frequencies during sweeping. Therefore, time-averaged uniform heating could be achieved with proper adjustment of the frequency sweep rate and sweep range. Another advantage of the VFM heating is the capability of providing precise frequency tuning to optimise the coupling efficiency.

2. VFM PROCESSING OF MATERIALS

The variable frequency microwave facilities (VFMF) are located at Industrial Research Institute Swinburne (IRIS), Swinburne University of Technology, Australia and consist of a Microcure 2100 Model 250 with a frequency sweep range of 2 - 8 GHz operating at a nominal power of 250W, and of a Wari-Wave VW1500 with a frequency range of 6.5 – 18 GHz at a nominal power of 125 W. The VFM facilities consist of a curing cavity and an oven control system, which is linked to a PC for programme input. The dimensions of the cavity for Microcure 2100 Model 2500 are 300 mm x 275 mm x 375 mm. The cavity dimensions of Wari-Wave VW 1500 are 250 mm x 250 mm x 300 mm. In these experiments, two halves of lap shear test piece of the sample, as shown in Figure 3 were joined together using the VFM energy with Araldite as primer. The lapped area for the joint was 1.0 cm x 2.0 cm. The bond surfaces were first roughened with coarse, grade 80 emery paper. The roughened surfaces were then cleaned and degreased by immersing them in methanol. After drying, five-minute two-part Araldite of around 1.5 to 2 cubic centimetres was applied to the two roughened surfaces to increase the mechanical keying or interlocking [11]. The two test pieces were then brought together and the total pressure applied was about 4 N. Programme with the required parameters was then written and input to control the VFMF via the PC. For Microcure 2100 Model 250, the input power level could be varied in steps of 10 W, starting from 50 W to 250W. During cavity characterisation, the actual amount of power that passed through the test pieces with respect to time was measured using fibre optic; in addition, the power reflected back from the material could also be detected. By this way, the best frequency range to process a material by microwaves can be found. Ku et al. found that the best frequency ranges to process PS/GF (33%) were 8.5 – 9 GHz or 10 – 12 GHz [12].

3. PROGRAMME FOR PS/GF (33%)

In order to have enough power to process PS/GF (33%) at 200 W, Microcure 2100 Model 250 VFM oven has to be used. The frequency range for this equipment is from 2 to 8 GHz but the best frequency to process PS/GF (33%) is 8.5 – 9 GHz. It is therefore necessary to make a compromise and a central frequency of 7.45 GHz has been selected in accordance with the 'compromised' best frequency to process PS/GF (33%). Since the bandwidth of the sweep should be greater or equal to 1.1 GHz, the selected bandwidth was 1.1 GHz [13]. The actual start and stop frequencies would be centre frequency $\pm \frac{bandwidth}{2}$, i.e. the sweep would be from 6.9 GHz to 8 GHz. Because the sweep time could range from 0.1 second to 100 seconds, the chosen sweep time was 0.1 second. Since the material loss tangent was relatively low, a power level of 200 W was selected

[14-16]. The processing temperature was set at 95^{o}C with a deadband (precision) of 1^{o}C and the total processing time was set at 8 minutes. The maximum permitted temperature was set at 100^{o}C, above that the machine was switched off automatically. The programme for joining PS/GF (33%) were as follows: central frequency = 7.45 GHz; bandwidth = 1.1 GHz; sweep time = 0.1 secs; power output = 200 Watts; set temperature = 95 ^{o}C; deadband = 1 ^{o}C; duration = 8 minutes; maximum temperature = 100 ^{o}C.

The centre sweep frequency, 7.45 GHz, and its sweep bandwidth, 1.1 GHz, were found to be very close to that required for processing the epoxy resin primer, Araldite at optimum conditions [17].

A maximum temperature of 100 ^{o}C was selected because it was very near to the melting point of one of the main constituents of the composite, the polystyrene (PS). The reason for setting this maximum temperature was to avoid excessive temperature rise, which forms hot spots and thermal runaway. Programs for other exposure times are also written. Results of the process will be given in the result section later on.

4. FIXED FREQUENCY MICROWAVE PROCESSING OF MATERIALS

In the fixed frequency microwave processing of PS/GF (33%), two microwave configurations are used. One is a microwave focus, high energy rate, fixed frequency equipment with slotted waveguide, as shown in Figure 4 and the other is the variable frequency microwave (VFM) oven but set at a fixed frequency of 2.5 GHz.

A. FIXED FREQUENCY EQUIPMENT

This involves the use of a TE_{10} mode rectangular waveguide operating in a standing wave configuration. Slots were machined in the waveguide allowing the adhesive layer on the specimens to pass through the microwave region. PS/GF (33%) specimens with the same lap area and surface treatment were placed in a standard rectangular waveguide as depicted in Figure 5 [6,18]. To avoid microwave radiation leakage, the slotted waveguide was enclosed in a modified commercial microwave oven case (Figure 4). One to one and a half millilitre of Araldite were smeared on both surfaces of the lapped area. A short circuit was adjusted to ensure that the maximum of the standing wave coincided with the lapped area of the specimen [4,6]. The input power to the system was in a step function and could only be 240W, 400W, 640W and 800W. The power was changed by altering the power of the source. The duration of exposure could be increased in steps of 1 second. The change of temperature during the joining process was not measured. The samples were exposed to 400 W and 800 W of power at different exposure times. The magnetron was operating at 2.45 GHz. The bonds formed were lap shear tested and the results are outlined later.

B. PROGRAMME FOR PS/GF (33%) USING VFM FACILITY BUT AT A FIXED FREQUENCY OF 2.5 GHz.

The program for joining PS/GF (33%) were as follows: Central frequency = 2.5 GHz; bandwidth = 0.0 GHz; power output = 200 Watts; set temperature = 95 ^{o}C; deadband = 1 ^{o}C; duration = 9 minutes; maximum temperature = 100 ^{o}C.

5. RESULTS

A. PS/GF (33%) JOINING USING VFM

During processing, it was found that the temperature rose steadily with no sign of hot spots or thermal runaway as shown in Figure 6. The maximum temperature reached was 95^{o}C at time equalled to 480 seconds. For obtaining lap shear test results, several sets of test pieces were joined at different duration. A Shimadzu tensile testing machine was used for the lap shear test. The load range of 2000 N and a load rate of 600 N per minute were used for the test [19]. The results are summarised in Figure 7. The lap shear bond strength increases from 380 N/cm^2 at an exposure time of 180 seconds to 430 N/cm^2 (peak value) at an exposure time of 420 seconds. Up to these times of microwave exposure, the failures were at bond line. At exposure times of 450 and 480 seconds, the lap shear strengths were 405 N/cm^2 and 370 N/cm^2 respectively. The quality of the bonds was not good. The failures are at the parent material. This means that the parent material [PS/GF (33%)] is weakened by the excessive exposure to microwave irradiation.

This behaviour is similar to that of LDPE/GF (33%) [Thirty three percent by weight random glass fibre reinforced low-density polyethylene] when it is exposure to excessive microwave energy [20]. At an exposure time of 420 seconds, the peak lap shear strength is 430 N/cm^2, which is 55% higher than the average lap shear bond strength obtained by curing the Araldite under ambient conditions. For all intervals of exposure to VFM, the lap shear bond strengths are stronger than the average lap shear bond strength procured under ambient environment.

B. PS/GF (33%) JOINING USING FIXED FREQUENCY SLOTTED RECTANGULAR WAVEGUIDE

With reference to Figure 8, it was found that with 400 W power level, peak lap shear bond strength was achieved by exposing the test pieces to microwaves for 2 minutes; the lap shear bond strength (326 N/cm^2) at this exposure duration exceeded that obtained by ambient conditions (conventional) curing by 17% but the time required was a mere of 0.2 % of its counterpart. For exposure times of one and a half to four and a half minutes, the lap shear strength obtained at microwave cured filler were higher than those obtained by allowing the adhesive to set under ambient conditions. With a power level of 800 W, maximum lap shear strength (331 N/cm^2) was achieved when the exposure time was 45 seconds and it exceeded the ambient conditions cured lap shear strength by 19 % but the time required was only 0.08% of its counterpart. The lower lap shear strength obtained, for test pieces exposed to microwaves for over 2 minutes and 45 seconds for power levels of 400 W and 800 W respectively, may be due to over-curing of the adhesive [4]. With some exposure durations, the bond strengths might be higher than those cured conventionally because the parent material may have melted and diffused into the primer or interface and this was reflected in the softening of the lapped area after it was just removed from the applicator and examined using low power microscopy [4]. Lap shear tests revealed that sixty percent of the failures were due to failure of the adhesive and took place at the joint interface. The remaining failures took place at the original materials [PS/GF (33%)]. With a power level of 400 W and exposure time of 2 minutes, the highest lap shear strength achieved was 326 N/cm^2; while with a power level of 800 W and exposure time of 45 seconds, the peak lap shear strength obtained was 331 N/cm^2. It can therefore be argued that the higher the power level used the shorter the exposure time required to achieve higher bond strength. These were up to 29 % and 62 % respectively stronger than the conventionally cured test pieces.

C. A. PS/GF (33%) JOINING USING VFM BUT WITH A FREQUENCY FIXED AT 2.5 GHz

With a VFMF a fixed frequency of 2.5 GHz was chosen. The same primer was also used and the power level of 200 W was chosen. While the processing temperature was set at 95^{o}C and the processing time was set at 540 seconds. The maximum temperature was set at 100^{o}C to prevent overheating. As a temperature of 100^{o}C (precision set was 1^{o}C) was attained at time equalled to 290 seconds, the machine was automatically stopped. It was found that temperature rose steadily and slowly in the first one hundred seconds as shown in Figure 9. The steep rise in temperature was observed when time equalled to 101 seconds. Heat conductivity was still very good at the beginning of the process and the heat was conducted to the surrounding area quickly as the temperature reached below 80^oC. Above this temperature and in time duration of 210 seconds, hot spot(s) developed into thermal runaway and the machine was shut down when the temperature of 100^oC was reached at time equalled to 290 seconds. Since the power level of 200 W caused hot spots and possibly thermal runaway, the power level was reduced to 150 W and several sets of test pieces were processed at different time intervals. Figure 10 shows the temperature versus time diagram for PS/GF (33%) joined at 150W and for a period of 570 seconds. The maximum temperature of 95^oC was recorded in running this experiment and the optimum lap shear test results are detailed in later paragraphs.

Figure 11 shows the 'apparent' bond strength of PS/GF (33%) bonded with two-part five-minute Araldite as primer, using VFMF. The word apparent was used because all test pieces failed at the parent material and not at the bondline. This meant that the bondline was stronger than the parent material. It was found that the apparent peak bond strength of $266N/cm^2$ was observed at an exposure time of 480 seconds. Values for other exposure times were just above 250 N/cm^2.

6. DISCUSSION

Referring to Figure 7, it is found that the maximum lap shear strength obtained by joining PS/GF (33%) using variable frequency microwave (VFM) facility with Araldite as primer is 430 N/cm^2, which is 62% higher than that (266 N/cm^2) (Figure 11) obtained using

VFMF but at a fixed frequency of 2.5 GHz. It takes only 420 seconds to employ the former parameters to arrive at the peak lap shear bond strength. While, it takes 480 seconds (15% longer) to use the latter parameters to achieve the maximum lap shear strength. In general, the lap shear bond strengths obtained at various exposure times by using the former parameters are higher than those procured by the latter. In addition, no hot spot (Figure 12) can be identified if the joining process is carried out using VFM configuration. On the other hand, hot spots, as indicated by a white arrow in Figure 13, can be found if the joining is carried out using VFMF but at a fixed frequency of 2.5 GHz.

Referring to Figures 8 and 11, it is found that the lap shear strengths, including the peak one, obtained using VFMF but at a fixed frequency of 2.5 GHz, were lower than those obtained using the fixed frequency microwave configuration (Figure 4). The apparent peak bond strength was only 82 % of that obtained with a power level of 400 W, using the fixed frequency facilities. It was only 80.5% of that obtained with a power level of 800 W and was marginally higher (3.5%) than its ambient cured rival [6,12]. In addition, the exposure times in VFM were much longer than its counterpart. At peak bond strength, the exposure time using VFMF was 4 times longer than that of its counterpart with a power level of 400 W. With the power level of 800 W, the value increased to 10.7 times. This was mainly due to the fact that the output power used in the VFM facility was only 150 W. Even if the maximum power output of 250 W was employed, the results did not improve significantly. Since the low power output made the processing time long, hence in microwave processing the power output played a significant role. Since the failure of the test pieces were at the parent material, it could be argued that the bondline strength should be more than 266 N/cm^2. In fact, it can be more than 430 N/cm^2 as in the case of joining the material using VFM facility.

7. CONCLUSIONS

PS/GF (33%) has been successfully joined by Araldite using VFMF with excellent bond quality (Figure 12). The outstanding average lap shear bond strength obtained was 803 N/cm^2, which was 189% higher than the average lap shear bond strength procured under ambient conditions. These outstanding results have to be contributed to the success of the VFMF as the latter can exactly find out the best frequency to process a material. It can therefore be argued that finding the optimum cavity conditions (characterisation) of a material using VFMF is the first step in microwave processing of the material. When variable microwave frequency is used, characterisation of a material is a 'must' because it identifies the exact frequency range within which the maximum coupling of the microwave energy into the load can be obtained. In this research, the characterisation of the material, PS/GF (33%) and the primer, Araldite was carried out. It is a much-preferred way of using microwave energy because without it time, effort and money would be spent unwisely. The best frequency range to process both the adherend and the primer is very near to each other and there is significant overlapping area between them [11,15]. There would be no problem as to which frequency range is chosen in processing the materials. If the best frequency range to process the two materials are quite far apart, then it is necessary to find out which material is more microwave reactive. The frequency range of the more lossy material, ie with higher loss tangent will then be selected for processing the materials.

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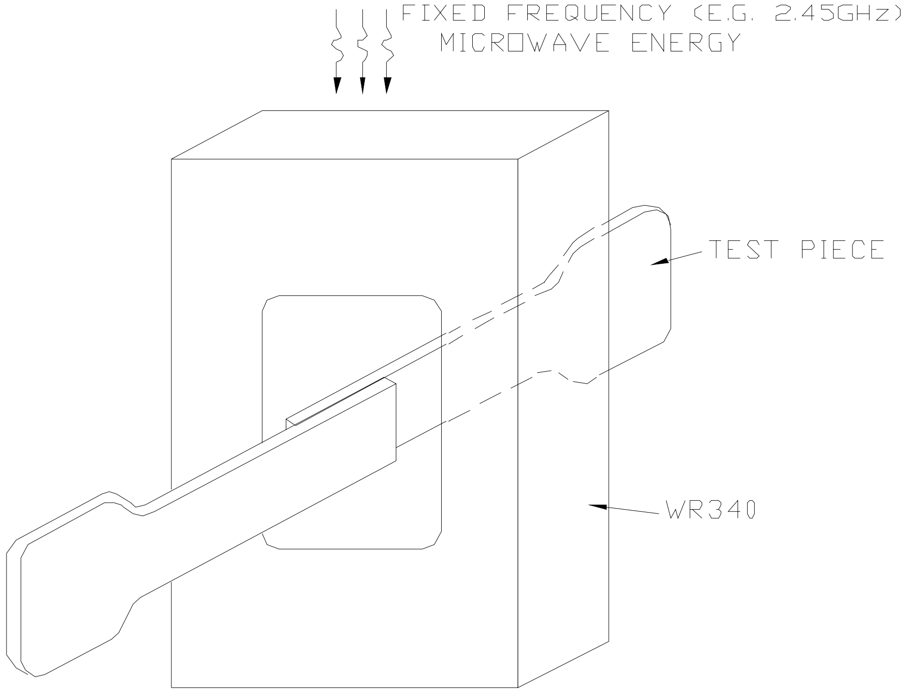
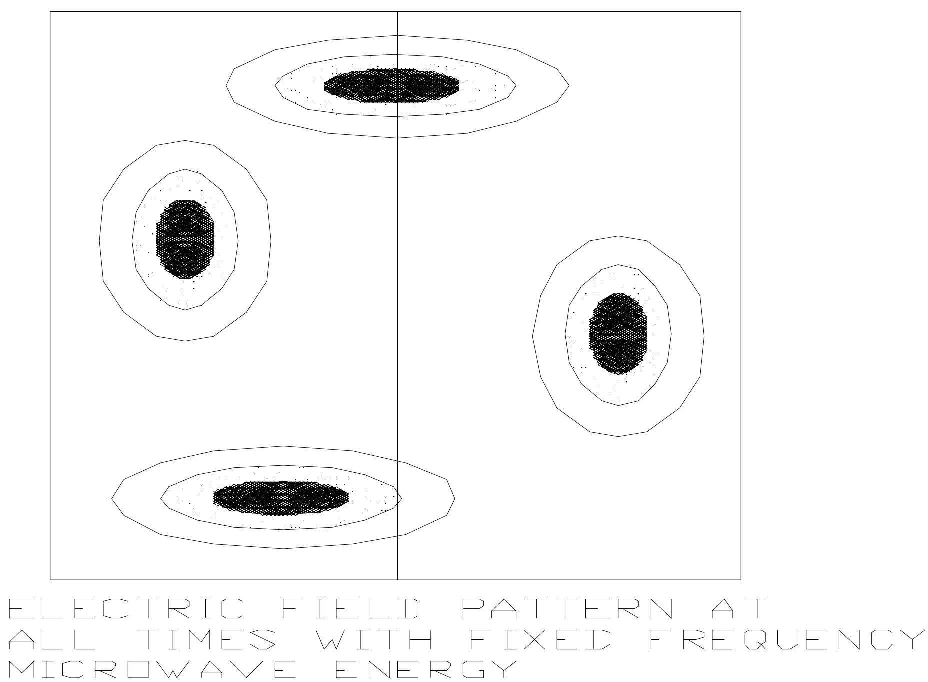
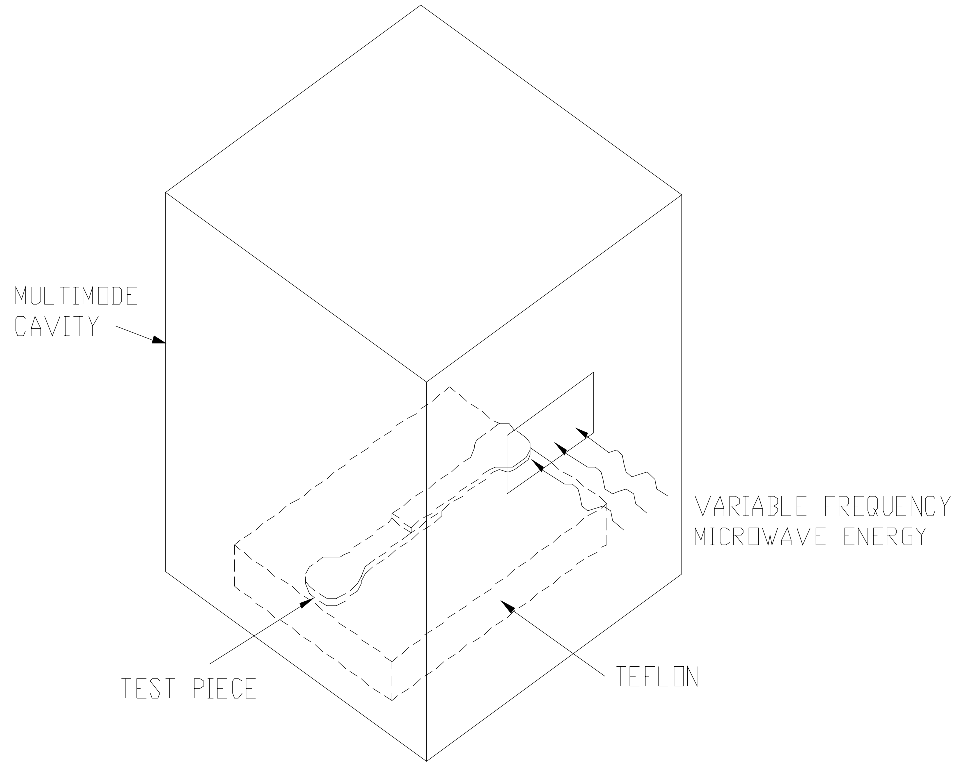


a) 2.45 GHz Microwave Energy Launched into a Single Mode Applicator

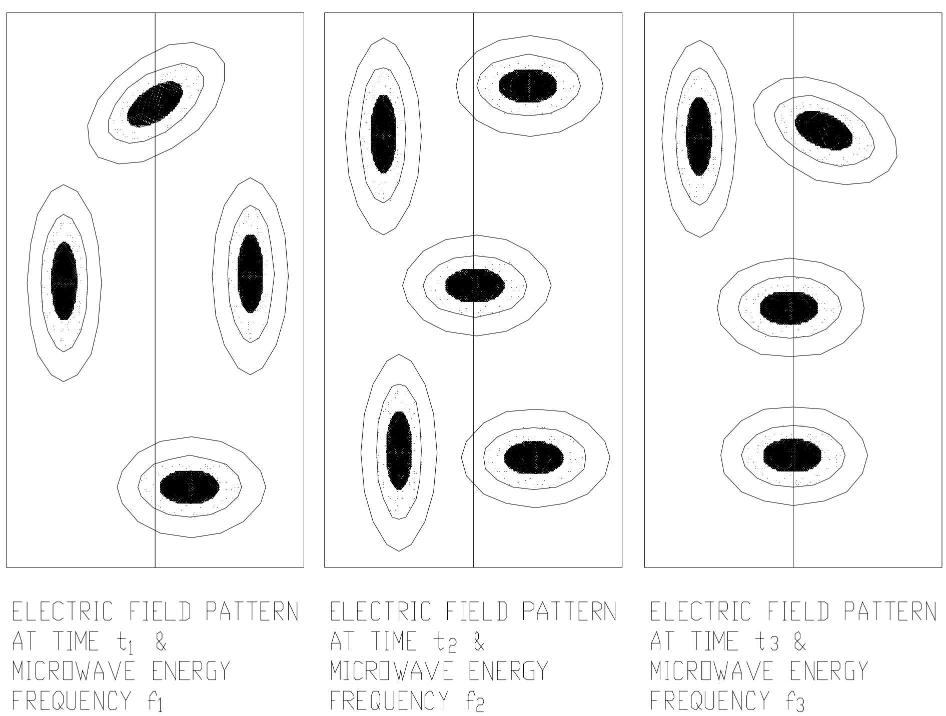


b) Electric Field Pattern for (a)

Figure 1: Fixed Frequency Microwave Heating – Nonuniform Heating



a) Variable Frequency Microwave Energy Launched into Multi Mode Cavity



b) Electric Field Pattern at Different Times in (a)

Figure 2: Variable Frequency Microwave Heating – Time-Averaged Uniform Heating

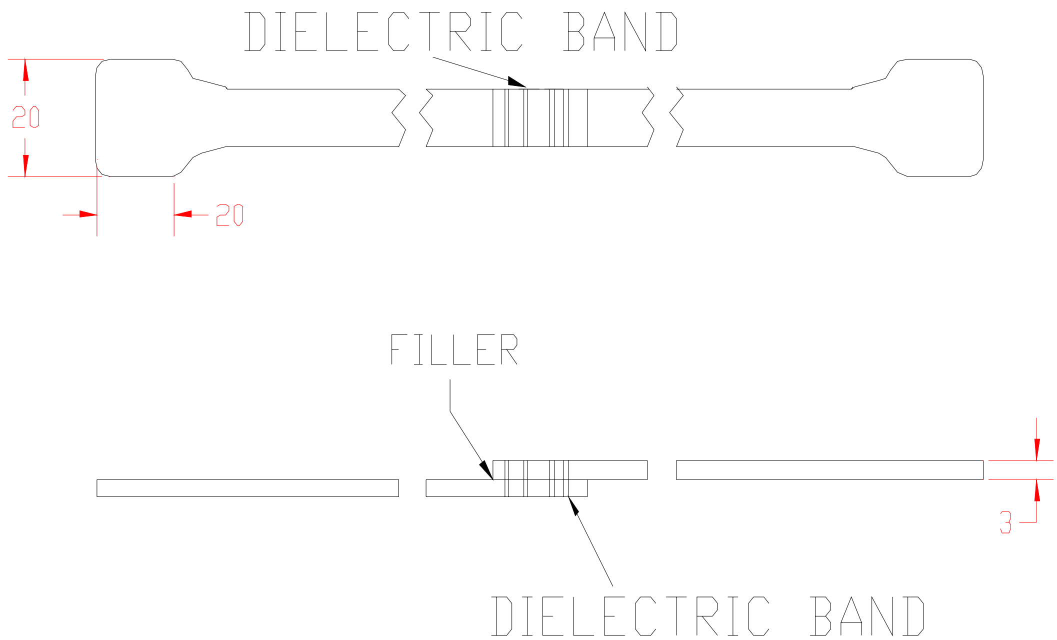


Figure 3: Two Mirror Image Test Pieces of PS/GF (33%)

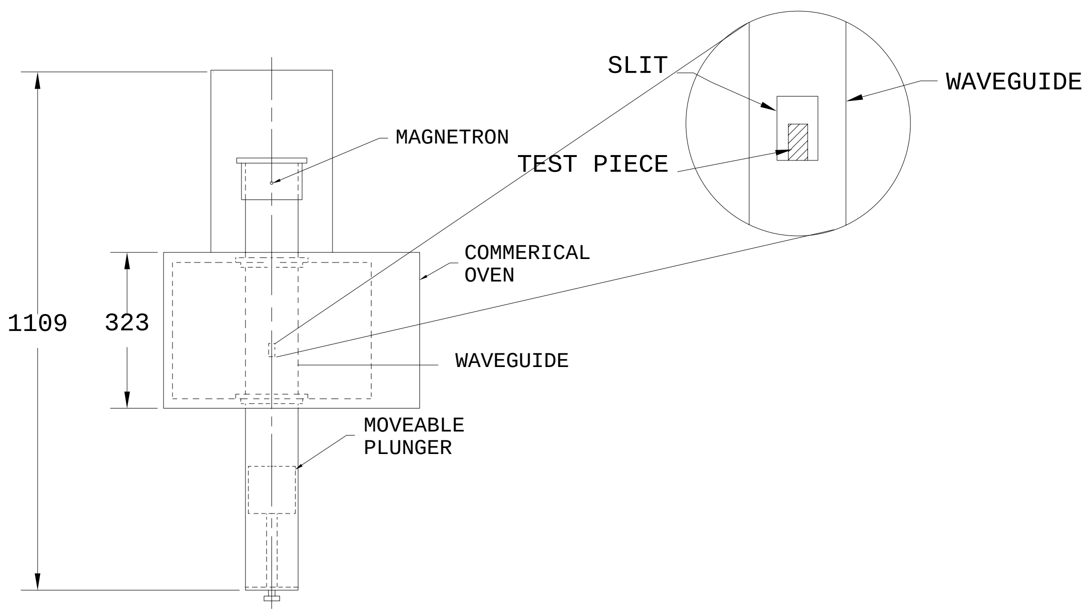


Figure 4: Slotted Rectangular Waveguide Microwave Configuration with All dimensions in mm

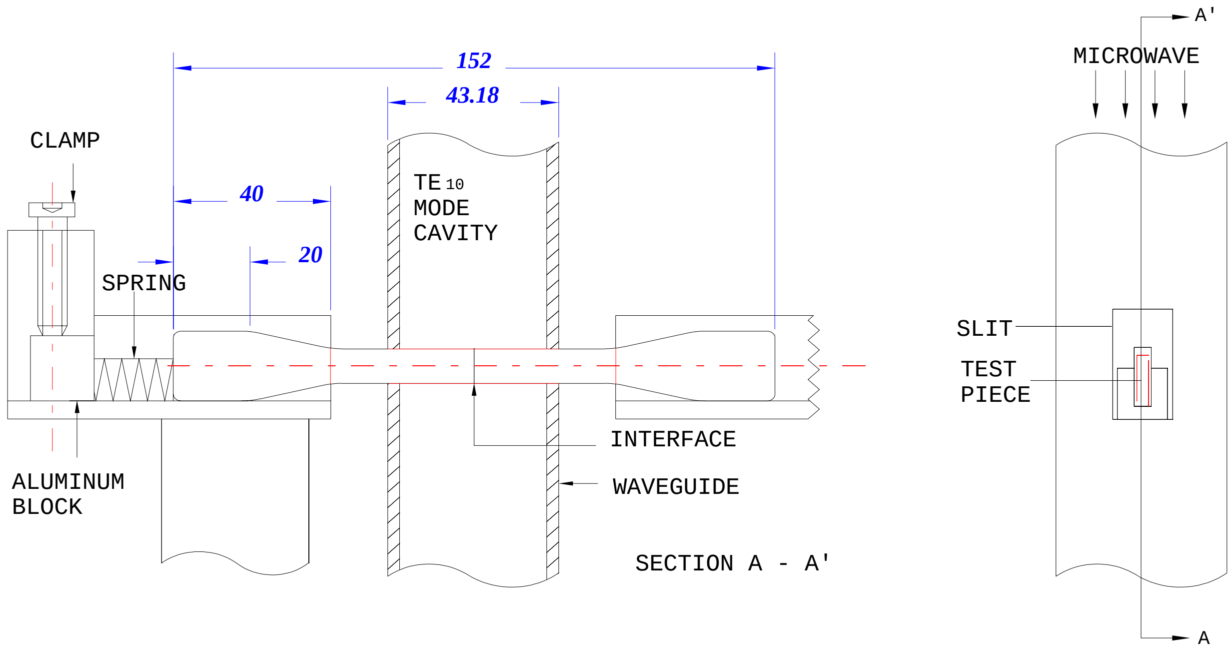


Figure 5: Slotted Rectangular Waveguide Used for Joining PS/GF (33%) with Araldite Using Fixed Frequency Equipment

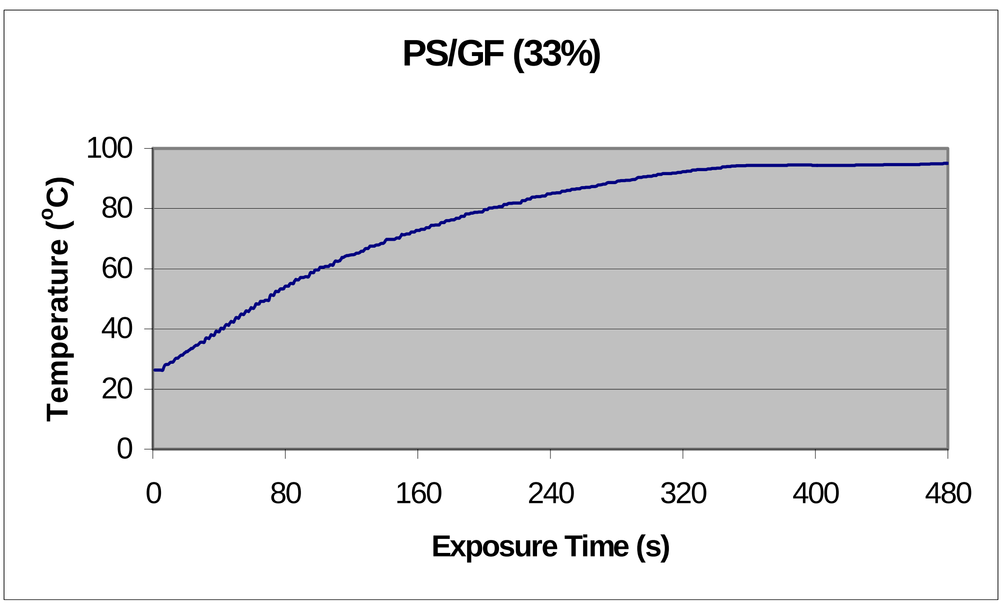


Figure 6: Time vs Temperature for PS/GF (33%) for 480 seconds at 200 W using VFM

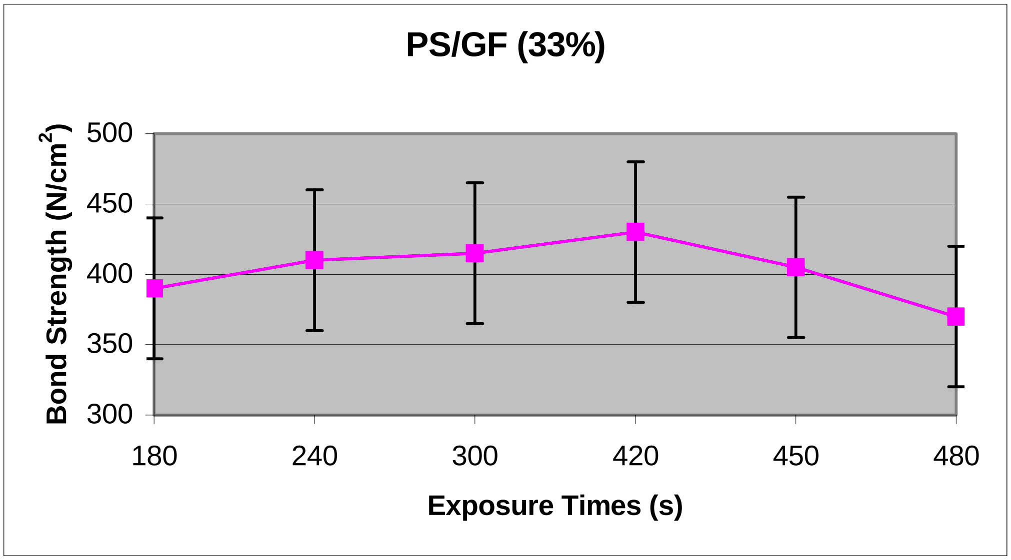


Figure 7: Lap Shear Strength of PS/GF (33%) Bonds Joined by VFM

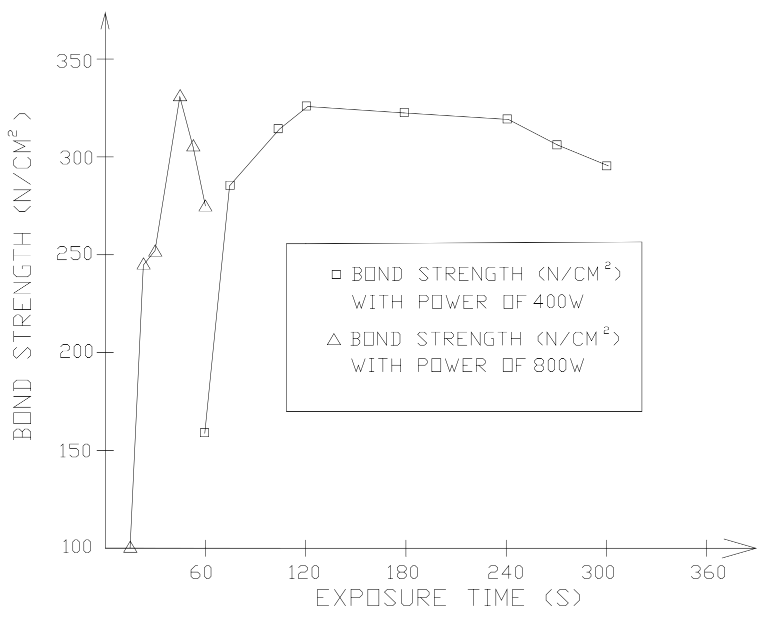


Figure 8: Bond Strengths of PS/GF (33%) and Five-Minute Two-Part Adhesive

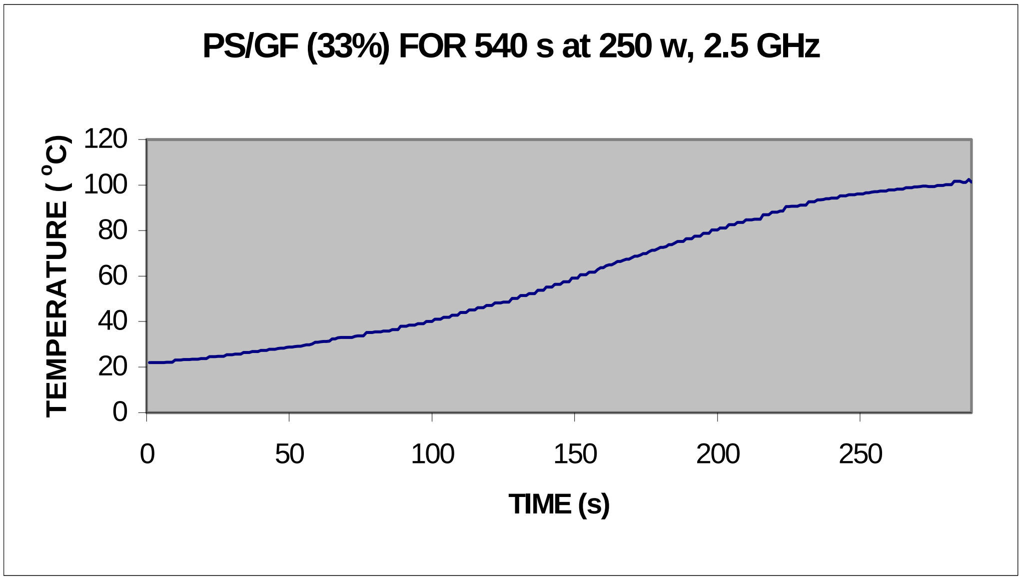


Figure 9: Temperature versus Time for PS/GF (33%) at 250 W

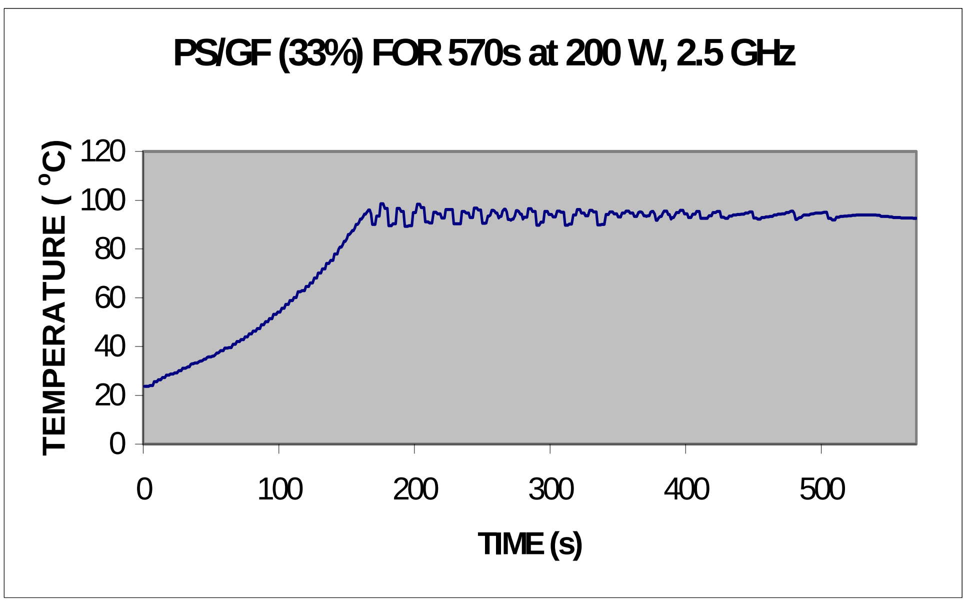


Figure 10: Temperature versus Time for PS/GF (33%) at 200 W using VFMF but at a Fixed Microwave Frequency of 2.5 GHz

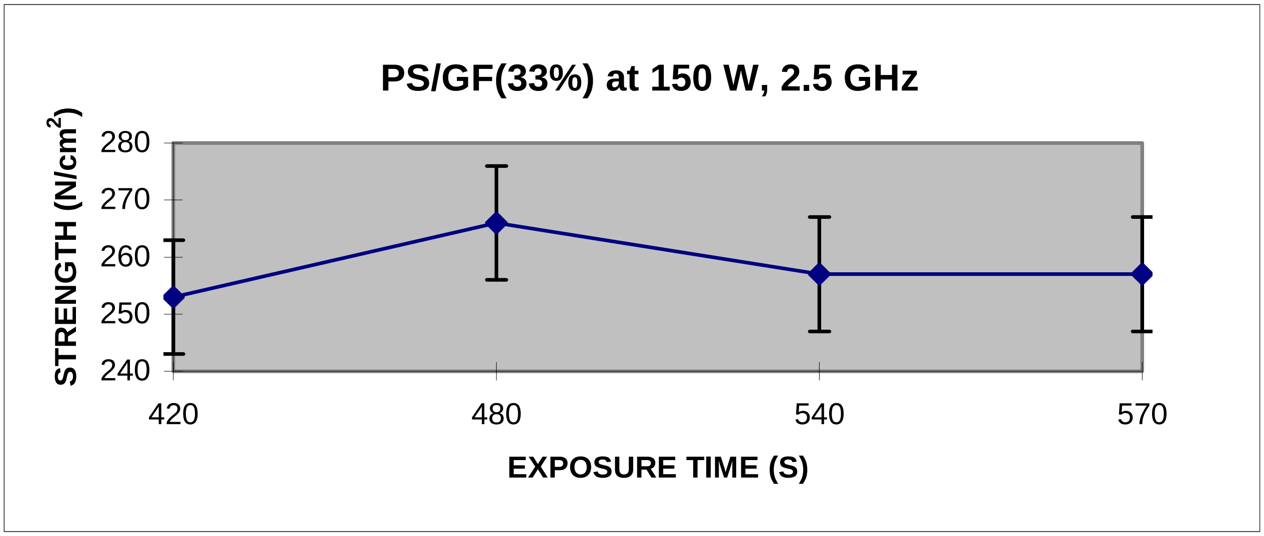


Figure 11: Bond Strength of PS/GF (33%) with Araldite using VFMF but at a Fixed Microwave Frequency of 2. 5GHz