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H.S.KU

High Energy Rate of Joining Thermoplastic Composites Using Microwaves

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Industrial Research Institute, Swinburne (IRIS)
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ABSTRACT

One of the most important areas of the application of industrial microwave technology for materials processing is curing polymers and polymer based composites, and this is currently in a state of considerable flux. Curing of thermosetting composites in multi-mode applicators and joining of a range of thermoplastics in ridged waveguides have been some of the major achievements in the field. This research extends the applications horizon of microwaves in the area of random fibre reinforced thermoplastic composites joining and places emphasis on the development of facilities and methodologies aiming at maximising bond quality. It discusses the fixed frequency microwave equipment used, including a 0.8 kW variable control power generator operating at 2.45 GHz, waveguide and a tuning piston designed for obtaining a standing wave at the seam of the butted and lapped test pieces. The effect of power input and cycle time is detailed together with the underlying principles of test piece material interactions with the electromagnetic field. The process of heterogenous joining, with filler material, or primer, of 33% by weight of random glass fibre reinforced nylon 66, polystyrene (PS) and low density polyethylene (LDPE), thermoplastic composites is mentioned together with the process of autogenous joining of 33% by weight of random carbon fibre reinforced polystyrene (PS) and low density polyethylene (LDPE). The weldability dependence on the dielectric constant, dielectric loss, and hence loss tangent of these materials is also described and investigated. Measurements of the dielectric properties were carried out using the waveguide transmission, probe kit and resonant cavity techniques. X-ray photoelectron spectroscopy for surface analysis was carried out to assess if the filler material had diffused into the parent thermoplastic composite materials. Bond strength measurements of the welded thermoplastic composites were obtained using tensile shear tests. A variable frequency microwave (VFM) facility was also used to join the thermoplastic composites and bond strength results obtained were compared with those procured by the fixed frequency microwave configuration.
From the measured data of LDPE/GF (33%), PS/GF (33%) and nylon 66/GF (33%), it was found that at relatively high temperatures and frequencies, these materials became less reflective and permitted more microwave energy to be absorbed. At the same time, their dielectric loss factor increased with temperature and frequency and enabled more of the absorbed energy to be converted to heat. Therefore, within limits, higher temperatures and higher frequencies were more suitable for microwave-assisted joining of the three materials tested. As to the primer/filler, only liquid rapid Araldite was found to be the most favourable in microwave-assisted joining of fibre reinforced thermoplastic (FRTP) composites because of its considerably high value of loss tangent. Another important finding was that the dielectric probe method was found to be the recommended technique to use for measurements of $\varepsilon'$ and $\tan \delta$ in microwave joining of materials out of all the methods applied. However, for low loss materials, the values of $\tan \delta$ need to be acquired through the resonance cavity method.
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To myself for the courage to stick to it through thick and thin.

Harry S KU
Swinburne University of Technology
August, 2000
DECLARATION BY CANDIDATE

I, Harry Siu-lung KU, of 10 Gerrard Street, Toowoomba, Queensland, 4350, Australia, hereby declare that the thesis:

- contains no material which has been accepted for the award to the candidate of any other degree or diploma, except where due reference is made in the text of the thesis;

- to the best of the candidate’s knowledge contains no material previously published or written by another person except where due reference is made in the text of the thesis; and

- where the work is based on joint research or publications discloses the relative contributions of the respective workers or authors.

______________________                                              ____________________
(Signature)                                                                             (Date)
1. INTRODUCTION

1.1 Brief Outline of Industrial Applications of Microwaves  

1.2 Overview of Composite Materials  

1.3 Aims and Objectives of the Research and Development Work  

1.4 Contribution to New Knowledge  

1.5 Summary of Chapters  

1.6 Publications

2. LITERATURE REVIEW

2.1 Introduction  

2.2 Introduction to Composite Materials  

2.3 Fibres  

2.3.1 Types of Fibres  

2.3.2 Fibre Volume Fraction  

2.3.3 Fibre Orientation  

2.3.4 Fibre Architecture  

2.4 Matrix  

2.5 Interface
3. COMPOSITE MATERIALS BONDING AND MECHANISMS OF ADHESION

3.1 Introduction .................................................................19
3.2 Butt Joint versus Lap Joint .................................................20
3.3 Adhesion ......................................................................22
3.4 Primers .........................................................................22
3.5 Mechanisms of Adhesion ..................................................23
   3.5.1 Mechanical interlocking .............................................24
   3.5.2 Electronic Theory of Adhesion ....................................24
   3.5.3 Diffusion Theory of Adhesion ....................................25
   3.5.4 Adsorption Theory of Adhesion .................................25
3.6 Concluding Remarks.........................................................26

4. MICROWAVE ENERGY AND ITS INTERACTION WITH MATTER

4.1 Overview of Microwaves ...................................................27
4.2 Microwave Technology for Joining .................................27
4.3 Microwave Propagation ...................................................29
   4.3.1 Wave Equations and Solutions .................................29
   4.3.2 Standing Waves ..........................................................31
   4.3.3 Reflection and Transmission Coefficients ..................32
   4.3.4 Waveguide Modes .......................................................33
4.4 Propagation of Microwaves via a Rectangular Waveguide .......34
4.5 Microwave/Matter Interactions .........................................37
# 5. EXPERIMENTAL EQUIPMENT - MICROWAVE FACILITIES CONFIGURATION

5.1 Overview .................................................................41  
5.2 Short-circuit Plunger and Its Dimensions ...........................41  
5.3 Duty Cycle of the Magnetron .......................................46  
5.4 Risk Assessment ......................................................47  
5.5 Concluding Remarks ................................................51  

# 6. PERMITIVITY MEASUREMENT AT VARIOUS FREQUENCY AND ELEVATED TEMPERATURES

6.1 Introduction .........................................................52  
6.2 Different Measurement Methods for Permittivity .................54  
6.3 Reflection Coefficient and Depth of Penetration ..................54  
6.4 Calibration of Logic Network Analyser .............................58  
6.5 Theory of Waveguide Transmission Technique ...................59  
6.6 Waveguide Sizes and Sample Lengths .............................63  
6.7 Dielectric Probe Method .............................................64  

# 7. RESONANT CAVITY METHOD FOR MEASUREMENT OF DIELECTRIC LOSS FOR LOW LOSS THERMOPLASTIC COMPOSITES

7.1 Introduction .............................................................67  
7.2 Cavity Length ..........................................................70  
7.3 Calibration of Logic Network Analyser ..............................71  
7.4 Formulae for Calculating Q Factor ..................................75
7.4.1 Phase Turning Points ..............................................76
7.4.2 Magnitude Only Method ........................................77
7.4.3 Phase Slope Method .............................................78
7.4.4 $\pm 90^\circ$ Method ..............................................80

8. VARIABLE MICROWAVE FREQUENCY FACILITIES

8.1 Introduction .........................................................81
8.2 Interaction of VFM with Matter .................................82
8.3 Hardware of Microcure 2100 .....................................86
  8.3.1 Curing Cavity ................................................86
  8.3.2 Oven Control System .........................................87
  8.3.3 Signal Generator and High Power Amplifier .............87
  8.3.4 VFM Safety Printed Circuit Board ..........................87
     .................................................................87
  8.3.5 Transmission System ........................................88
  8.3.6 Temperature Monitoring System ............................88

8.4 Software System ...................................................88
  8.4.1 Introduction ................................................88
  8.4.2 Utilities Menu ..............................................89

8.5 Characterisation of Thermoplastic Matrix Composite (TMC)
    Materials ........................................................90
  8.5.1 Characterisation of TMC Materials from
       6.5 GHz to 18 GHz ...........................................90
  8.5.2 Characterisation of TMC Materials from
       2 GHz to 8 GHz ..............................................92

8.6 Programming Menu ..................................................94
  8.6.1 LDPE/GF (33%) Program (2-8 GHz) .......................94
  8.6.2 PS/GF (33%) Program (2-8 GHz) .........................96
8.6.3 Other Programs (2-8GHz) ..............................................97

8.7 Concluding Remarks ....................................................99

9. RESULTS AND DISCUSSION

9.1 Introduction ........................................................................100
9.2 Summary of All Findings ....................................................101
9.3 Results of Surface Analysis

.................................................................102

9.3.1 Low Power Microscopy .................................................102
9.3.2 X-ray Photoelectron Spectroscopy .................................103
9.3.3 Diffusion Theory Explanation ........................................109

9.4 Results of Permittivity Measurements ...............................109

9.4.1 Results of Waveguide Transmission Technique ............110
9.4.2 Results from Dielectric Probe Method ............................115
9.4.3 Waveguide Transmission Technique versus

Dielectric Probe ..............................................................120

9.4.4 Concluding Remarks ....................................................122

9.5 Results of Loss Tangent Using A Resonant Cavity Method ..........123

9.6 Best Frequency Range to Process Materials Using VFMF ..............129

9.7 Results of Bond Strength Tests .........................................130

9.7.1 Glass Fibre Reinforced Polystyrene

[PS/GF (33%)] ..............................................................130

9.7.2 Glass Fibre (33%) Reinforced Nylon 66

[Nylon 66/GF (33%)] ..................................................135

9.7.3 Glass Fibre (33%) Reinforced Low Density Polyethylene

[LDPE/GF (33%)] ......................................................137

9.7.4 Carbon Fibre (33%) Reinforced Polystyrene

[PS/CF (33%)] ..........................................................139

9.7.5 Carbon Fibre (33%) Reinforced Low Density Polyethylene
9.7.6 Effects of Different Processes on Bond Strengths ..........144
9.7.7 Cross Comparison of Results ..................................144

9.7.7.1 LDPE/GF (33%), PS/GF (33%) and Nylon 66/GF (33%)
   at 400 W .................................................................144

9.7.7.2 LDPE/GF (33%) and PS/GF (33%) at 800 W
   .......................146

9.7.7.3 LDPE/CF (33%) and PS/CF (33%) .........................147

9.7.7.4 Peak Bond Strengths and Fixed Frequency Joining ..........147

9.8 Concluding Discussion...........................................150

9.8.1 Relationship between the Microwaves and the Matrices ......151

9.8.2 Relationship between the Microwaves and the Fibres ......157

9.8.3 Energy to Break Bonds
   .................................................................165

9.8.4 Weakened Parent materials by Excessive Microwave
   Irradiation ...........................................................166

10. CONCLUSIONS AND RECOMMENDATIONS FOR
    FURTHER WORK

10.1 Conclusions .......................................................169

10.2 Recommendations for Further Work
   .................................................................170

REFERENCES ...............................................................172

Appendix A

Publications ..............................................................185
NOMENCLATURE

a  waveguide broad-face dimension
α  attenuation coefficient
α₁  attenuation coefficient at point 1
b  waveguide narrow-face dimension
β  phase coefficient or phase shift per unit length
β₁  phase coefficient at point 1
c  velocity of light, 3 x 10⁸ m/s
°C  degrees Celsius
D  larger dimension of the antenna
d  sample thickness
dB  decibel
dBm  decibel relative to an absolute power level of 1 mW
δ  loss tangent, skin depth
δᵉ  depth of penetration
Δf  change in frequency
Δt  change in time
ΔQ  change in Q factor
ΔV  sample volume
E  electric field (general)
E₀  electric field in an empty cavity
Eₛ  electric field in a cavity with sample
Eₓ  electric field in x direction
Eᵧ  electric field in y direction
Eᶻ  electric field in z direction
ε  complex relative permittivity
ε₀  permittivity in free space, 8.854 x 10⁻¹² F/m
ε'  dielectric constant
ε''  dielectric loss factor
f  frequency (general)
f_a  frequency at point a
f_b  frequency at point b
f_o  resonant frequency with empty cavity
f_s  resonant frequency with sample
\phi_m  phase difference between two extreme points
\gamma  complex propagation coefficient
GHz  Gigahertz
\Gamma_0  propagation coefficient in air filled waveguide
\Gamma_1  propagation coefficient in sample filled waveguide
H  magnetic field (general)
H_0  magnetic field in free space
H_x  magnetic field in x direction
H_y  magnetic field in y direction
H_z  magnetic field in z direction
L  sample length
\lambda_c  cut-off wavelength within the guide
\lambda_g  wavelength within the guide
\lambda_0  free space wavelength
m  metre (length)
mm  millimetre (length)
\mu  permeability
\mu_0  permeability in free space, 1.256 \times 10^{-6} \text{ H/m}
n  number of half wavelengths
Q  Q factor
Q_o  Q factor with empty cavity
Q_s  Q factor with sample
\rho  reflection coefficient
|\rho|  magnitude of reflection coefficient (general)
|\rho_{minimum}|  magnitude of minimum reflection coefficient
$|\rho_a|$ magnitude of reflection coefficient at point a

$|\rho_b|$ magnitude of reflection coefficient at point b

$S_{11}$ reflection coefficient from port 1 to port 2

$S_{22}$ reflection coefficient from port 2 to port 2

$S_{12}$ transmission coefficient from port 1 to port 2

$S_{21}$ transmission coefficient from port 2 to port 1

$\sigma$ conductivity

$\sigma_e$ electronic conductivity

$\sigma_i$ ionic conductivity

TE transverse electric field

TM transverse magnetic field

$U_{total}$ total averaged stored energy in cavity

$v_p$ velocity of propagation

$\omega$ resonant frequency (general), angular velocity ($2\pi f$)

$\omega_0$ resonant frequency with empty cavity

$\omega_s$ resonant frequency with sample

$Z_0$ wave impedance

$Z_{\omega}$ wave impedance

$Z_{os}$ intrinsic impedance of a material
# LIST OF ILLUSTRATIONS

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.1</td>
<td>Test Pieces in Position</td>
<td>19</td>
</tr>
<tr>
<td>3.2</td>
<td>Butt and Simple Lap Joints</td>
<td>21</td>
</tr>
<tr>
<td>3.3</td>
<td>Stepped Lap and Scarf Joints</td>
<td>22</td>
</tr>
<tr>
<td>4.1</td>
<td>Schematic Diagram of Cross-section of a Magnetron</td>
<td>28</td>
</tr>
<tr>
<td>4.2</td>
<td>Reflection and Transmission of a Normal Incidence Electromagnetic Wave</td>
<td>33</td>
</tr>
<tr>
<td>4.3</td>
<td>Synthesis of a Guided Wave Between Conducting Planes by Two Coherent Plane Waves</td>
<td>35</td>
</tr>
<tr>
<td>4.4</td>
<td>Lines Representation (2-D) of the Fields of TE$_{10}$ Mode in a Rectangular Waveguide</td>
<td>36</td>
</tr>
<tr>
<td>4.5</td>
<td>Lines Representation (3-D) of the Fields of TE$_{10}$ Mode in a Rectangular Waveguide</td>
<td>37</td>
</tr>
<tr>
<td>4.6</td>
<td>Interaction of Microwave with Materials</td>
<td>38</td>
</tr>
<tr>
<td>4.7</td>
<td>Dielectric Relaxation of Typical Polar Dielectric</td>
<td>40</td>
</tr>
<tr>
<td>5.1</td>
<td>Fixed Frequency Microwave Facilities Configuration</td>
<td>42</td>
</tr>
<tr>
<td>5.2</td>
<td>Stationary Wave in Waveguide</td>
<td>43</td>
</tr>
<tr>
<td>5.3</td>
<td>The Moveable Plunger</td>
<td>44</td>
</tr>
<tr>
<td>5.4</td>
<td>Non-contact Type Adjustable Short Circuit in Waveguide</td>
<td>45</td>
</tr>
<tr>
<td>5.5</td>
<td>Dimensions for the Moveable Plunger</td>
<td>45</td>
</tr>
<tr>
<td>5.6</td>
<td>Set-up for Leakage Measurement</td>
<td>48</td>
</tr>
<tr>
<td>5.7</td>
<td>Antenna Orientation in Leakage Measurement</td>
<td>49</td>
</tr>
<tr>
<td>6.1</td>
<td>Equipment Set-up for Waveguide Transmission Technique</td>
<td>59</td>
</tr>
<tr>
<td>6.2</td>
<td>Magnitude of $S_{21}$ against Frequency</td>
<td>62</td>
</tr>
<tr>
<td>6.3</td>
<td>Phase of $S_{21}$ against Frequency</td>
<td>62</td>
</tr>
<tr>
<td>6.4</td>
<td>Schematic Diagram of HP 85070 Dielectric Probe</td>
<td>66</td>
</tr>
<tr>
<td>7.1</td>
<td>Set-up for Cavity Method</td>
<td>68</td>
</tr>
<tr>
<td>7.2</td>
<td>A Resonant Cavity with End Plates Removed</td>
<td>68</td>
</tr>
<tr>
<td>7.3</td>
<td>Network Analyser Smith Chart Display after Calibration</td>
<td>72</td>
</tr>
</tbody>
</table>
7.4 Smith Chart for WR229 Cavity with LDPE/GF(33%) Sample in the Vicinity of Resonance ......................................................... 73
7.5 Reflection Coefficient Magnitude for WR229 Cavity containing LDPE/GF (33%) Sample ......................................................... 73
7.6 Reflection Coefficient Phase for WR229 Cavity with
LDPE/GF (33%) Sample ......................................................... 74
7.7 Simplified Diagram of Reflection Coefficient Magnitude .......... 74
7.8 Simplified Diagram of Reflection Coefficient Phase ................. 75
7.9 Shifted Phase View .......................................................... 77
7.10 Phase Angle against Frequency ............................................. 79
8.1 Cavity Size of VW 1500 ....................................................... 83
8.2 Cavity Size of Microcure 2100 Model 250 .............................. 83
8.3 Fixed Frequency Microwave Heating - Non-uniform Heating .......... 84
8.4 Variable Frequency Microwave Heating - Time-Averaged
Uniform Heating ............................................................... 85
8.5 A Block Diagram of the System of Microcure 2100 .................. 86
8.6 Percentage of Reflectance versus Frequency for LDPE/GF(33%) .... 91
8.7 Percentage of Reflectance versus Frequency for LDPE/CF(33%) .... 93
8.8 Percentage of Reflectance against Frequency for Nylon 66/GF (33%).. 93
8.9 Percentage of Reflectance against Frequency for Liquid Rapid
Araldite ................................................................. 94
8.10 Temperature versus Time LDPE/GF(33%) ................................ 96
8.11 Temperature versus Time for PS/GF(33%) at 200 W .................. 97
8.12 Temperature versus Time for PS/GF(33%) at 150 W .................. 98
9.1 The O and C Atom Intensities against Binding Energy of
LDPE/GF (33%) Plus Araldite .............................................. 104
9.2 The –C-C- and -C-O Bond Intensities against Binding Energy of
LDPE/GF (33%) ................................................................. 105
9.3 The O and C Atom Intensities against Binding Energy of
Araldite on Glass ............................................................... 106
9.4 The –C-C-, –C-O and C=O Bond Intensities against
Binding Energy of Araldite .................................................... 106
9.5 The C Atoms Intensity against Binding Energy of LDPE ............. 107
9.6 The –C-C- Bond Intensity against Binding Energy of LDPE ......... 108
9.7 Dielectric Constants of Air, Teflon, LDPE/GF (33%), Nylon 66, Nylon 66/GF (33%) at Room Temperature over Certain Frequencies ........................................ 110
9.8 Values of Dielectric Constant of LDPE/GF (33%) against Frequencies (2 – 12.5 GHz) ................................................. 112
9.9 Values of Dielectric Constant of PS/GF (33%) against Frequencies (2- 12.5 GHz) ......................................................... 113
9.10 Dielectric Constants of Nylon 66/GF (33%) at Elevated Temperatures over a Certain Frequencies .............................. 114
9.11 Dielectric Constants of Nylon 66/GF (33%) at Different Frequencies over a Range of Temperatures .............................. 114
9.12 Dielectric Loss Factors of Nylon 66/GF (33%) at Elevated Temperatures over Certain Frequencies ......................... 115
9.13 Dielectric Loss Factors of Nylon 66/GF (33%) at Different Frequencies over a Range of Temperatures ....................... 115
9.14 Dielectric Constants of LDPE/GF (33%) by Two Methods over a Frequency Range of 2 – 12.5 GHz ......................... 121
9.15 Dielectric Constants of PS/GF (33%) by Two Methods over a Frequency Range of 2 – 12.5 GHz ......................... 121
9.16 A Resonant Cavity with Sample at Bottom ........................................ 126
9.17 Bond Strengths of PS/GF (33%) and Five Minute Two Part Araldite .............................................................. 132
9.18 Bond Strengths of PS/GF (33%) with Araldite and at 2.5 GHz .... 134
9.19 Bond Strengths of Nylon 66/GF (33%) and Five Minute Two Part Adhesive ............................................................. 135
9.20 Bond Strengths of Nylon 66/GF (33%) with Araldite and at Variable Frequency ....................................................... 137
9.21 Bond Strengths of LDPE/GF (33%) and Five Minute Two Part Adhesive ............................................................. 138
9.22 Bond Strengths of LDPE/GF (33%) with Araldite and at Variable Frequency ....................................................... 139
9.23 Bond Strengths of PS/CF (33%) with No Filler ........................................ 140
9.24 Bond Strengths of PS/CF (33%) with No Primer and at Variable Frequency .......................................................... 141
9.25 Bond Strengths of PS/CF (33%) at Variable Frequency and Different Power Levels .................................................. 142
9.26 Bond Strengths of LDPE/CF (33%) with No Primer ............... 143
9.27 Bond Strengths of LDPE/CF(33%) with
No Primer and at 2.5 GHz .................................................. 143
9.28 Bond Strengths of Three Types of Materials at 400 W .............. 145
9.29 Bond Strengths of Two Types of Materials at 800 W ............. 146
9.30 Three Types of Basic Polymeric Materials ............................. 151
9.31 Locations of Carbon Filaments on test Piece ......................... 161
9.32 Locations of Black Marks ................................................. 161
9.33 The Intensity of Elements against Binding Energy in
LDPE/ GF (33%) Sample with Black Patches ......................... 163
9.34 The Intensity of Elements against Binding Energy in
Control Sample ................................................................... 163
9.35 The Two Peaks of Intensity for Carbon Atoms in the Sample
with Black Patches ............................................................... 164
9.36 The Peak of Intensity for Carbon Atoms in Control Sample ......... 164
10.1 Modified Fixed Frequency Microwave Facilities Configuration .... 171

<table>
<thead>
<tr>
<th>Plate</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plate 1</td>
<td>Menu-driven Graphical Interface ................................. 89</td>
</tr>
</tbody>
</table>
| Plate 2 | 30 Times Microscopy Photo of Diffusion of Araldite into
           Thermoplastic Composite ........................................ 103 |
| Plate 3 | 300 Times Microscopy Photo of Diffusion of Araldite into
           Thermoplastic Composite ........................................ 103 |
| Plate 4 | 1500 Times Microscopy Photo of Diffusion of Araldite into
           Thermoplastic Composite ........................................ 104 |
| Plate 5 | LDPE/CF(33%): Burnt Mark in Test Piece as a Result of
           Fixed Frequency Joining ........................................... 149 |
| Plate 6 | PS/CF(33%): No Burnt mark in Test Piece as a Result of Variable
           Frequency Joining .................................................. 149 |
Plate 7 100 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 70 seconds ........................................154
Plate 8 400 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 70 seconds ........................................154
Plate 9 100 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 80 seconds .................................155
Plate 10 400 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 80 seconds .................................155
Plate 11 1000 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 80 seconds ...............................156
Plate 12 100 Times Magnification of Bondline with Joining Parameters of VFM, 200 W and 420 seconds .................................158
Plate 13 400 Times Magnification of Bondline with Joining Parameters of VFM, 200 W and 420 seconds .................................158
Plate 14 100 Times Magnification of Bondline with Joining Parameters of VFM, 200 W and 450 seconds .................................159
Plate 15 400 Times Magnification of Bondline with Joining Parameters of VFM, 200 W and 450 seconds .................................159
<table>
<thead>
<tr>
<th>Table</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.1</td>
<td>$S_21$ Marker Values for LDPE/GF(33%) at 18°C</td>
<td>62</td>
</tr>
<tr>
<td>6.2</td>
<td>Waveguide Type, Frequency Range and Sample Length</td>
<td>64</td>
</tr>
<tr>
<td>7.1</td>
<td>Relationship between Initial and Targeted Frequencies</td>
<td>71</td>
</tr>
<tr>
<td>9.1</td>
<td>Percentage of Atoms of Microwaved LDPE/GF(33%) plus Araldite</td>
<td>105</td>
</tr>
<tr>
<td>9.2</td>
<td>Percentage of Atoms of Araldite on Glass</td>
<td>107</td>
</tr>
<tr>
<td>9.3</td>
<td>Summary of percentage of atoms for Three Different Materials Analysed</td>
<td>109</td>
</tr>
<tr>
<td>9.4</td>
<td>Dielectric Constant and Loss Values of Pure Water at 25°C</td>
<td>116</td>
</tr>
<tr>
<td>9.5</td>
<td>Dielectric Constant Values of LDPE/GF(33%), LRA, CRA over a Wide Band of Frequencies at 25°C</td>
<td>117</td>
</tr>
<tr>
<td>9.6</td>
<td>Loss Tangent Values of LDPE/GF(33%), PS/GF(33%), LRA and CRA over a Wide Band of Frequency at 25°C</td>
<td>117</td>
</tr>
<tr>
<td>9.7</td>
<td>Values of $\varepsilon'$ of LTRA, CTRA, LHSA and CHSA over a Range of Frequency</td>
<td>119</td>
</tr>
<tr>
<td>9.8</td>
<td>Values of $\tan\delta$ of LTRA, CTRA, LHSA and CHSA over a Range of Frequency</td>
<td>120</td>
</tr>
<tr>
<td>9.9</td>
<td>Values of $\varepsilon'$ and $\varepsilon''$ for WR340 with Sample [LDPE/GF(33%)] Standing Upright</td>
<td>123</td>
</tr>
<tr>
<td>9.10</td>
<td>Values of $\varepsilon'$ and $\varepsilon''$ for WR229 with Sample [LDPE/GF(33%)] Standing Upright</td>
<td>123</td>
</tr>
<tr>
<td>9.11</td>
<td>Values of $\varepsilon'$ and $\varepsilon''$ for WR159 with Sample [LDPE/GF(33%)] Standing Upright</td>
<td>125</td>
</tr>
<tr>
<td>9.12</td>
<td>Values of $\varepsilon'$ and $\varepsilon''$ for WR90 with Sample [LDPE/GF(33%)] Standing Upright</td>
<td>125</td>
</tr>
<tr>
<td>9.13</td>
<td>Values of $\varepsilon'$ and $\varepsilon''$ for WR159 with Sample [LDPE/GF(33%)] Laying at the Bottom</td>
<td>126</td>
</tr>
<tr>
<td>9.14</td>
<td>Values of Q factor for Air-filled Wr159 at 24°C</td>
<td>127</td>
</tr>
<tr>
<td>9.15</td>
<td>Values of Q factor for LDPE/GF (33%)-filled Wr159 at 24°C</td>
<td>128</td>
</tr>
</tbody>
</table>
9.16  Best Frequency Range to Process the 5 Types of Materials in the Frequency Range of 2 GHz to 18 GHz ..........................129
NOTE

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1. Introduction

1.1 Brief Outline of Industrial Applications of Microwaves

Industrial applications of microwaves are relatively new technology. Factors that have hindered the use of microwaves in materials processing have been declining so that prospects for the development of this technology seems to be very promising (Sutton, 1989). The two mechanisms of orientation polarisation and interfacial space charge polarisation, together with dc conductivity, have formed the basis of high frequency heating (Metaxas and Meredith, 1983; Siores, 1994). The general mechanisms that govern the energy dissipation process and the microwave/material interaction included dipole friction, current loss and ion jump relaxation (Siores, 1994). The material properties of greatest importance (NRC, 1994; Ku et al, 1998; 1999a) to microwave processing of a dielectric are the complex relative permittivity \( \varepsilon = \varepsilon' - j\varepsilon'' \) and the loss tangent, \( \tan \delta = \varepsilon'' / \varepsilon' \). The real part of the permittivity, \( \varepsilon' \), sometimes called the dielectric constant, mostly determines how much of the incident energy is reflected at the air-sample interface, and how much is absorbed. The most important property in microwave processing is the loss tangent, \( \tan \delta \), which predicts the ability of the material to convert the absorbed energy into heat. For optimum microwave energy coupling, a moderate value of \( \varepsilon' \) to enable adequate penetration, should be combined with high values of \( \varepsilon'' \) and \( \tan \delta \), to convert microwave energy into thermal energy. In a material with a very high loss tangent, the microwave energy density is being reduced with distance of penetration into the material. This phenomenon is known as the penetration depth.

1.2 Overview of Composite Materials

Composites are essentially a combination of more than one type of material. The final product obtains its good properties from each component. Sometimes a composite is the only material available to obtain the properties needed. Composites can be combinations of metal-metal, metal-ceramic, metal-plastic,
ceramic-ceramic, ceramic-plastic, or plastic-plastic. There are three groups of composite materials: particulates, fibres and laminates. The length of the fibres is critical in fibre reinforced composites.

The composite materials used in this research include 33 percent by weight carbon fibre reinforced low density polyethylene [LDPE/CF (33%)], 33 percent by weight glass fibre reinforced low density polyethylene [LDPE/GF (33%)], 33 percent by weight carbon fibre reinforced polystyrene [PS/CF (33%)], 33 percent by weight glass fibre reinforced polystyrene [PS/GF (33%)] and 33 percent by weight glass fibre reinforced nylon 66 [Nylon 66/GF (33%)].

1.3 Aims and Objectives of the Research and Development Work

The aims and objectives of the research and development work include:

- performing research and development work on the weldability of composite materials, using high energy microwaves;
- studying the heat transfer phenomena involved;
- studying the bonding mechanisms involved;
- enhancing the universality of the process to facilitate joining a range of composite materials;

1.4 Contributions to New Knowledge

The contributions to new knowledge presented in this thesis are itemised below for easy reference:

i) Method of measurement and analysis of dielectric constant, $\varepsilon'$, of 3 types of glass fibre reinforced thermoplastic composites, and of the dielectric loss, $\varepsilon''$, of Nylon 66/ GF (33%) using the waveguide transmission and dielectric probe techniques. It was also found that the liquid form of two-part five minute rapid Araldite (part A consisted of 100% liquid epoxy resin and part B comprised of 8% amine) had the highest dielectric loss
factor and is therefore best suited to microwave processing amongst the three types of Araldite tested;

ii) Method of measurement and analysis of the dielectric loss of low loss thermoplastic composites, eg LDPE/GF (33%) over a range of frequencies and at elevated temperatures, using the resonant cavity technique. It was found that in calculating the dielectric loss of the material, the averaged Q factors should be taken into consideration;

iii) The best frequency ranges for microwave processing of the 5 thermoplastic composite materials were obtained and subsequently it was found that variable frequency microwave (VFM) processing of thermoplastic composites was more advantageous over its fixed frequency counterpart;

iv) Determining, in terms of bond strength, the best combination of power level and time of exposure to fixed and variable frequency microwave irradiation for joining the 5 types of thermoplastic composite materials. The power level was found to be vital in joining thermoplastic composites irrespective of whether fixed or variable frequency microwave irradiation was used. Prolonged exposure of the carbon fibre reinforced thermoplastic composite materials to microwave energy was harmful and for LDPE/GF (33%) the bond strength would not improve much with increased duration of microwave irradiation, irrespective of whether fixed or variable frequency microwave energy was used. However, variable frequency microwaves (VFM) gave better bond quality both in terms of bond strength and appearance;

1.5 Summary of Chapters
Chapter 2 of this thesis consists of a literature review; the work performed by previous researchers is discussed. The chapter starts with giving the definition of composite materials. One of the constituents of a composite material is the matrix, the continuous phase; the other is present in the form of fibres or particulates, which act as reinforcement and this reinforcement forms a discontinuous phase that is uniformly dispersed throughout the matrix. Types, volume fraction, orientation angle and architecture of fibres are mentioned together with the matrix and interface outlined. Polymer matrix composites, the composites to be researched in this project, are then discussed. The reasons for using the thermosetting composites exclusively, until the last decade, are also mentioned. The fact that thermoplastic composites can be re-cycled an infinite number of times and the improvement in processing and joining technology of the materials fuelled the rapid acceptance of the materials as a substitute for their counterparts, the thermosetting composite materials. Finally, the reasons for choosing specific materials for this research are also discussed.

Chapter 3 describes the bonding and adhesion mechanisms of composite materials. The key issue in joining composite materials is bond formation with or without primer and in order to test the strength of this bond, a suitable testing method selected is outlined. The advantages and limitations of the commonly used, butt joint and lap joint are investigated. The terms, adhesion, adhesives and adhesion system are also detailed. Factors that are considered in using adhesives are also studied. The four theories of mechanisms of adhesion, namely mechanical interlocking, electronic theory, diffusion theory and adsorption theory are also briefly discussed.

Chapter 4 starts with an overview of microwaves. Typical microwave frequencies reserved for industrial applications are listed and the application of microwaves to joining of materials is also mentioned. The chapter then describes the propagation of microwaves in space. Wave equations and solutions are then outlined. Standing waves, reflection and transmission coefficients and
waveguide modes are interrelated and discussed. Because this project utilises rectangular waveguides for microwave propagation, the propagation of microwaves via those waveguides is detailed. In microwave and matter interactions, the terms absorber, transparent and opaque are used and explained. The term penetration depth and its importance in material processing using microwave energy is also mentioned.

Experimental equipment for joining the thermoplastic matrix composite material and procedures are detailed in Chapter 5. The chapter starts with an overview of the applicator design and the design theories behind it. This is followed by the design of the short-circuit plunger and its dimensions. The reasons for choosing the dimensions used are also described. The duty cycle of the magnetron used is detailed next and examples are used to illustrate the working principles. The chapter is summed up by risk assessments, which includes the risk assessment of the entire microwave facility configuration, the short-circuit plunger and the thermoplastic composite materials employed. Microwave leakage measurements were also taken to ensure that the risk of radiation leakage was below the acceptable level.

Chapter 6 explains the permittivity measurement of fibre reinforced thermoplastic (FRTP) matrix composites at various frequencies and elevated temperatures. The chapter starts by introducing several method of measuring permittivity. This is followed by the derivations of the reflection coefficient and the depth of penetration. Calibration of the logic network analyser for the waveguide transmission technique is then described. The reasons for using the waveguide transmission technique and the theories behind it are then mentioned. The need to use different waveguide sizes and sample lengths are also detailed. Finally, the grounds for using another method, the dielectric probe method, for measuring permittivity are given.

A resonant cavity method for the measurement of dielectric loss for low loss thermoplastic composites, eg LDPE/GF (33%), is discussed in chapter 7. The
reasons for using the method and theories behind it are first discussed. The relationship between the specific cavity length and the targeted frequency is then described. Calibration of the logic network analyser for the resonant cavity method is then outlined. Four methods are introduced and used for calculating the Q factors. These make use of the reflection coefficient phase turning points, magnitude, phase slope and ±90° frequencies respectively.

The variable microwave frequency (VFM) facility is described in chapter 8. The chapter starts with listing the drawbacks of conventional fixed frequency microwave processing. The meaning of VFM processing is then explained together with the two different types of machine located in Industrial Research Institute, Swinburne (IRIS). The interaction of VFM fields with materials follows. The system and subsystems of the more powerful of the two VFM machines, Microcure 2100 model 250, are mentioned briefly. Characterisation of thermoplastic matrix composite (TPC) materials is then explained. The materials characterised include LDPE/CF (33%), LDPE/GF (33%), PS/CF (33%), PS/GF (33%) and nylon 66/GF (33%). Programming of the machines for joining the FRTP composite materials together with their processing characteristics are detailed.

In chapter 9, results of surface analysis in bonding and adhesion, of measurements of complex permittivity using waveguide transmission technique, dielectric probe kit and a resonant cavity are described. The best frequency range for processing the five thermoplastic matrix composite materials and bond strengths obtained from the tests are detailed in the same chapter. A low power microscopy study and tests by x-ray photoelectron spectroscopy carried out on the joined surfaces of the composite material to prove, or otherwise the diffusion of the araldite into the thermoplastic and vice versa are outlined. The relationship between the matrices of the composites, the thermoplastics and their structures, and between the reinforcements, the fibres and the time of exposure to microwave irradiation, are thoroughly discussed. Finally, conclusions and recommendations for further work are detailed in chapter 10.
1.6 Publications

Publications arising from this research and development work include six refereed conference and twelve journal papers. Five of the conference papers have been published; of the eight journal papers, three have been published, three have been accepted for publication and the rest are in-press. For details, please see Appendix A at the end of the thesis.
2. Literature Review

2.1 Introduction

Published work in the areas of composite materials were searched and carefully studied. This enables the impartial assessment of other learned work and paves the way for independent research.

2.2 Introduction to Composite Materials

There is no universally accepted definition of composite materials. Definitions in literature vary considerably (Schwartz, 1995) and they consider different structural levels of matter. At the macrostructural level, gross structural forms of constituents, e.g., matrices, particles, and fibres are dealt with. A composite material (Mallick, 1997) is, therefore, a combination of two or more chemically different materials with a distinct interface between them. Microscopically, separate identities of the constituent materials are maintained, but the combined materials produce different characteristics and properties from those of the parent materials. One of the constituents forms a continuous phase, the matrix; the other is present in the form of fibres or particulates which act as reinforcement and this reinforcement forms a discontinuous phase that is uniformly dispersed throughout the matrix. The matrix material in a composite can be a polymer, metal, or ceramic. Depending on the matrix used, composite materials are classified as polymer matrix composites (PMCs), metal matrix composites (MMCs), or ceramic matrix composites (CMCs). Most of the commercially used composites have polymers as their matrix. Various chemical combinations, compositions, and microstructural arrangements are possible in each matrix category. Both MMCs and CMCs which find high temperature applications are attracting more users. The primary effect of the reinforcement relies somewhat on the matrix used. In polymer based composites, the fibre increases the modulus and strength of the composite materials many times because polymers have very low modulus and strength as compared to the reinforcing fibres. On the other hand, the fibre reinforcement in ceramic matrix composites increases the fracture toughness of the matrix and makes the materials less susceptible to
brittle failure. Ceramic matrix composites have low fracture toughness but their modulus is more or less the same as that of the reinforcing fibres. The addition of fibres with controlled interfacial properties to the ceramic matrix also increases its crack resistance significantly. Fibres can either be used in continuous or discontinuous lengths but are more effective when used in the former case. For discontinuous fibres, the fibre aspect ratio can be varied but the length is usually less than 6 mm (Strong, 1989). The most common form in which fibre-reinforced composites are used in structural applications is called a laminate. Laminates are obtained by stacking a number of thin layers of fibres and matrix and consolidating them into the desired thickness. Fibre orientation in each layer, as well as the stacking sequence of various layers can be controlled to generate a wide range of physical and mechanical properties for the composite laminates. Traditionally, fibres have been used in the reinforcement of polymer, metal and ceramic matrices but particulates are now finding wider applications because they can be incorporated into those matrices with more ease. Particulates, however, are generally less effective than fibres in reinforcing the matrix.

The term advanced composites is being used to differentiate composite materials with high-performance characteristics, generally strength and stiffness, from the simpler forms like reinforced plastics (Schwartz, 1997). Most advanced composites are used in structural applications. The fibres are long with length-to-diameter ratio of over 100. The strength and stiffness of the fibre is much greater, often a multiple of those of the matrix material.

2.3 Fibres

In a fibre reinforced composite material, fibres are the principal load-carrying component. The type, length, volume fraction and orientation of the fibres in the matrix can contribute the effectiveness of the fibre reinforcement. Appropriate choice of the above parameters is vital because it affects one or more of the following characteristics of a fibre reinforced composite: cost, density, tensile
strength and modulus, compressive strength and modulus, response to impact loads, electrical and thermal properties, and fracture and fatigue performance.

2.3.1 Types of Fibre

A large variety of fibres is commercially available with a wide range of densities, properties and cost. Most of these fibres are manufactured in the form of long continuous filaments and then combined in various fashions to produce strands, tows, rovings, yarns, fabrics, mats and so on. Short or discontinuous fibres are procured by chopping long discontinuous fibres into lengths ranging from 3 – 50 mm. Nearly all commercial fibres have round or nearly round cross sections. Most filament diameters are in the range of 5 – 40 µm except that of boron, which has a typical diameter of 140 µm, which gives a high resistance to buckling. These fibres are mostly brittle, have a low elongation to failure and exhibit a linear tensile stress-strain response.

In the fibre reinforced plastic industry, glass fibres have been the major reinforcement constituents, but the desire for lighter, stronger, stiffer structure had seen the introduction of carbon and polyamid fibres. Carbon fibres have exceptionally high strength and stiffness but their cost is also exceptionally high and their applications have been limited to areas where cost is unimportant; for example in aircraft components and sporting goods. Polyamid fibres are not as strong as carbon fibres but their performance is above that of glass fibres. They are less expensive than carbon fibres and exhibit exceptional impact resistance. Industrial applications include in the areas of pressure gas construction, aerospace and marine applications, and sporting goods (Schwartz, 1997). Despite the high average tensile strength associated with fibres, a large variation in their strength is observed. Some fibres like carbon and aramid fibres are anisotropic; while others, like glass and boron fibres are isotropic. Both carbon and aramid fibres had negative coefficients of thermal expansion in the longitudinal direction and can be wisely used to design composites with zero or near-zero thermal expansion. Aramid and polyethylene fibres, on the other hand,
are organic fibres and have low operating temperatures as compared to other fibres.

2.3.2 Fibre Volume Fraction

Volume fraction is an important parameter in controlling the properties of a fibre reinforced composite. In general, the higher the fibre volume fraction, the higher the modulus, strength, and many other physical/mechanical properties of the composite. If the fibres are arranged in a regular and repeating array throughout the matrix, the total volume can be divided into unit cells and a theoretical volume fraction can be calculated using the fibre diameter and the unit cell dimensions. However, in practice, the fibres are randomly distributed and therefore theoretical volume fractions have very little use in estimating the properties of the composite.

2.3.3 Fibre Orientation

Fibre orientation angle is defined as the orientation of the fibres with respect to the loading directions. It is an important factor in determining the properties of the composite material. In pure shear loading, fibres are most effective when the fibre orientations are at $+45^\circ$ and $-45^\circ$ angles with respect to the loading axis. On the other hand, in uniaxial tensile loading, fibres are most effective when the fibre angle is $0^\circ$.

2.3.4 Fibre Architecture

Fibre architecture refers to the arrangement of fibres, which can be linear, two-dimensional or three-dimensional. In the linear form, fibres can be either continuous or discontinuous. Continuous fibres are used in filament wound, pultruded or laminated structures in which the fibre orientation can be precisely controlled. Discontinuous fibres are either directly mixed with the matrix or combined with a binder to form a planar mat. In both cases, there is very little
control over the fibre orientation. Two- and three-dimensional architectures can be produced by means of textile processes like intertwining, interlacing, and interlooping continuous fibres in textile machines. These processes provide good control over the fibre orientation as well as fibre placement and can be used to produce a variety of complex shapes or preforms in a relatively short time. Two-dimensional architectures are used in laminated structures. A three-dimensional arrangement of continuous fibres is used when interlaminar failure or delamination becomes a problem. They can also be used to build composites with equal properties in x, y and z directions.

2.4 Matrix

Matrix is the continuous phase in a composite material, and it plays several important roles in the composite. It holds the reinforcement in place, acts as a path for stress transfer between fibres, and protects the reinforcement from an adverse environment. The selection of a matrix affects several matrix-dominated mechanical properties of the composite. The matrix also has a minor role in the longitudinal tensile properties of unidirectional continuous fibre composites.

In addition to influencing the mechanical properties of a composite, the matrix has a major influence on its processing characteristics. For instance, the moulding time of a thermosetting polymer matrix composite depends on the time required to complete the curing that transforms the liquid thermosetting polymer into a solid polymer. The viscosity of the thermosetting polymer is also important, as it determines the flow characteristics of the polymer in the mould, which in turn influences the wetting of fibres by the matrix, the void content in the cured part, and the extent of polymer flow through the fibre network in some processes.
2.5 Interface

The matrix/reinforcement interface also plays a vital role in the performance of a composite. The reinforcement strengthens the matrix only if a strong interfacial bond exists between them. The interfacial properties also influence the crack propagation in a composite and therefore its fracture toughness. The two most important energy-absorbing failure mechanisms in a fibre reinforce composite are fibre pullout and debonding at the fibre/matrix interface (Mallick, 1997). In most polymeric matrix composites, increased interfacial bond strength is achieved by fibre surface treatment, which helps in forming a chemical linkage between the fibres and the matrix across the interface. Generally, a mechanical bond is formed due to differential shrinkage as the polymer matrix cools down from the processing temperature. Higher shrinkage of the matrix around the fibre creates residual stresses in both the fibre and the matrix. At the interface, the residual stresses are compressive in the radial direction but tensile in the hoop direction. At times, microcracks can be initiated in the matrix by the residual tensile stress in the hoop direction. Furthermore, if the mechanical bonding is disrupted at low stresses and the fibre/matrix interface debonded, the matrix may be unable to transfer the stresses efficiently to the fibres and the strength of the polymer matrix would be low. Some research workers (Mallick, 1997) have suggested that there exists a thin but distinct interface between the fibre and the matrix in a polymeric matrix composite. The interface properties are different from those of the bulk matrix due to the fibre’s influence on the microstructure of the matrix in the immediate region surrounding the fibre.

2.6 Polymer Matrix Composites (PMCs)

Polymer matrix composites (PMCs) are fibre reinforced polymers in which either a thermoset or a thermoplastic polymer is used as the matrix. The development of PMCs for structural applications started in the 1950’s, and they are by far the most common fibre reinforced composite materials in use today. One reason for their wide acceptance is that their processing is simple and does not involve high
pressure or temperature. The equipment used for processing is also relatively cheap as compared with that for other types of composite materials. Processing of PMCs can also be highly automated. The most significant advantage though of PMCs is that they are lightweight materials with very high strength and modulus values. Their lightweight is entirely due to the low densities of their constituents. The relative density of the polymers used varies from 0.9 to 1.5, while that of the fibres ranges from 1.4 to 2.6 (Mallick, 1997). The specific gravity of PMCs ranges from 1.2 to 2.0 depending on the types of fibre and polymer used as well as their relative volume fractions. On account of their low densities, the strength-to-weight and strength-to-modulus ratios of PMCs are comparatively higher than that of many metals and their composites. Although the cost of PMCs can be higher than metals, particularly if boron or carbon fibres are used as reinforcements, their cost, on a unit volume basis, can be competitive with that of high performance alloys used in aerospace industry. Design flexibility of PMCs is its another advantage. Fibres in PMCs can be selectively oriented to resist load in any direction, thus producing directional strength or moduli. Likewise, the fibres in PMC can be controlled to provide a variety of thermal properties like the coefficient of thermal expansion. Other advantages of PMCs that have made them desirable in many applications include their high damping factors and resistance to corrosion. The composite strength is almost directly proportional to the basic fibre strength and can be improved at the expense of stiffness. High modulus organic fibres have been made with simple polymers by arranging the molecules during processing which results in a straightened molecular structure. Optimisation of stiffness and fibre strength remains a fundamental objective of fibre manufacture (Schwartz, 1997). The stress-strain diagram of composite materials can be nonlinear. The nonlinearity is not due to plastic deformation but due to a number of types of microscopic defects such as fibre/matrix interfacial bonding. These defects can grow in size as well as in number in various locations of the PMCs as the stresses are increased. There may not be any immediate failure but the stiffness of the material would progressively decrease.
2. Literature Review

2.7 Thermoplastic Composites

Thermoplastic composites are composite materials in which the matrix is thermoplastic i.e. it melts when heated and solidifies when cooled and the cycle can be repeated an infinite number of times provided that the heat does not cause any damage (Harris, 1979; Meng et al, 1995). In the last 11 – 12 years, great efforts have been made to develop thermoplastic composite materials. A large number of thermoplastic resins including polypropylene (PP), nylon (PA), polyetherimide (PEI), and polyether ether ketone (PEEK) and so on, have been used for the development (Meng et al, 1995). Despite its high cost, PEEK has been accepted for high-performance applications because of its lightweight, temperature resistance, and high mechanical properties. The main attractive features of thermoplastics over thermosets include better toughness/damage tolerance, unlimited shelf life, recyclability/low scrap, and rapid fabrication cycles (Lee, 1990; Meng et al, 1995). Their processing methods are generally simple and include solution process, melt impregnation, film stacking, fibre co-mingling and dry powder impregnation for thermoplastic preforms, and roll forming, filament winding, pultrusion process, thermoforming procedures and so on for the manufacture of thermoplastic composite parts (Meng et al, 1995; Okine, 1997). Thermoplastics can provide the potential for process automation and lower cost (Okine, 1997). In addition, post-processing or reprocessing of thermoplastic composites is feasible. The processing of thermoplastic composites is in many ways similar to those of their counterparts and yet different in many others. The differences in flow characteristics of the two kinds of plastic resins require the development of new and different processing techniques for thermoplastics (Okine, 1997). Most of the composite aerospace parts have been made from thermosets, which require extensive manual processing operations, and this explains the limited applications of composites in commercial planes. However, in recent years, process automation and lower cost of thermoplastic composites have made these materials attractive in structural applications in the automotive industry (Meng et al, 1995). The above factors make the growth in using thermoplastic composites in structural materials very
strong and welding technology development has fuelled that growth (Schwartz, 1992). The advantages of using thermoplastic composites over the frequently used thermosetting composites include their capability to be formed into complex shapes at lower costs and high productivity rates (Partridge, 1989; Varandan, 1991). Most thermoplastic composites are joined by fusion bonding and the processes employed consist of resistance welding, ultrasonic bonding, vibrational bonding, high frequency welding, traditional infrared heated air, hot plate, hot melt and room-cure adhesives. The advantages and limitations of the above processes (Schwartz, 1992) are well documented and the frequency range (Schwartz, 1995) used in high frequency welding was 3 to 40 MHz which is in the radio frequency range. In addition, a few papers on microwave joining of thermoplastic composites (Varandan, 1991; Strokes, 1989; Siores, 1993; Wu, 1992; Paulauskas et al, 1996) pave the way for microwave technology developments in polymer processing. The merits (Schwartz, 1995) of employing microwaves in joining thermoplastic composites have included a clean and reliable interface at the joints and fast joining time with minimum destruction of the properties of the bulk materials. Limitations encountered in other processes were minimised.

2.8 Materials Selection

Random GF (glass fibre) reinforced (33%) nylon 66 was chosen for this study because the loss tangent of nylon 66 can be high compared with other commonly used thermoplastic materials at room temperature (Michaeli, 1995).

Random GF reinforced (33%) LDPE was selected because there was a successful case (Wu and Benatar, 1992) of welding the composite with HDPE as a matrix using microwave energy and it was believed that LDPE would couple better to microwaves (National Research Centre, 1994) as its crystallinity is lower than that of HPDE. However, this composite is not readily available in the Australian market and it was manufactured specially for the project in the Plastic and Rubber Training and Education Centre (PARTEC) in Brisbane, Australia.
A polystyrene (PS) matrix was chosen firstly because it is a common thermoplastic polymer matrix (Shackelford, 1994) and secondly because its loss tangent (Michaeli, 1995) is very near to that of LDPE and thus a comparison could be made later on. The random carbon fibre (CF) (23.3%) reinforced PS and the random glass fibre (GF) (33%) reinforced PS were also manufactured by PARTEC.

Random CF (carbon fibre) reinforced (33%) LDPE was selected for comparison with random GF reinforced (33%) LDPE, since the difference in microwave absorption by the different fibres was a matter of concern in the investigation.

Finally, CF reinforced (33%) PS was selected for reasons similar to those mentioned above.

In all cases, the length of the reinforcing fibre was chosen to be 6 mm or less since other investigations (Lubin, 1982) have showed that optimum mechanical properties are attained when 6 mm fibres are used and there were only marginal differences in mechanical properties if fibre lengths of between 3 mm and 6 mm are utilised. The test pieces were injection-moulded to shape. It is worth noting here though that typical lengths of fibres used in reinforced injection moulding materials range from 0.8 to 25 mm (Strong, 1989).

2.9 Concluding Remarks

Experts in the field have predicted that composites will likely be the materials of the future and their applications have spread from aerospace to medicine, sports, construction, automotive, and beyond. An important term in composite materials is advanced composites which are defined as composites that contain a fibre-to-resin ratio of greater than 50% fibre by weight, with the fibres having a modulus of elasticity greater than 110 GPa (Luce, 1988). A breakthrough in the development of advanced composites occurred when an English organisation produced carbon fibres of high strength and stiffness in the mid 1960s. By the mid of the 1970s, the manufacture and use of carbon based composite materials
had become common, in the U.S.A., Europe and Japan. Composites have undercut the cost of some aluminium parts in the aerospace industry. The savings come from eliminating the labour needed to form a complex aluminium structure of varying thickness and to rivet many small aluminium pieces together. On the other hand, composites can be formed in one piece with extra layers of the epoxy-impregnated graphite cloth added wherever required to increase strength.

Thousands of fasteners and time-consuming operations have been eliminated through the use of advanced composites, and large parts have been successfully produced. Limitations to widespread use of composites include scarcity and high cost of equipment for automating the process, high material prices, few volume applications and lack of shared experience in advanced composites manufacturing (Ellen, 1985).
3. Composite Materials Bonding and Mechanisms of Adhesion

3.1 Introduction

The key issue in the high energy rate welding of thermoplastic composites by microwave energy is bond formation (Ku et al, 1997a). In order to have a bond to form, it is necessary to heat the thermoplastic of the composite material to its glassy or even melting point and then apply pressure to coalesce the two pieces. To enable the test pieces to be heated up to the required temperature in this study, they have to be located in a position that favours the absorption of the microwave energy applied. The test pieces are best located across the waveguide through which the microwave energy travels. This is done by cutting slits across the waveguide and then positioning the test pieces as shown in Figure 3.1.

Figure 3.1: Test Pieces in Position

In order to measure the bond strengths obtained after joining the composites with and without fillers, a tensile test or lap shear test has to be carried out, depending on the joint selected. The test is simple but it is necessary to be careful in cutting the test piece because tool marks left behind can cause the source of failure. Moreover, in the case of very stiff and strong composites like those with carbon or aramid unidirectional fibres, it is necessary to add end reinforcements in order to avoid crushing the test piece where it is clamped at both ends. If this is not done, failure may initiate in the crushed region (Geier, 1994). As a precaution
when conducting a quality control test, the test piece must always be fitted with reinforcements at both ends. These have to be glued on with a strong epoxy adhesive or riveted. Two tensile test standards, ASTM D 3039 – 76 (ATMS, 1990) and ISO 3268 (Geier, 1994) for composite materials were considered for use but owing to the nature of fibre reinforcement and size of the microwave facilities configuration, an ISO tensile test standard for plastic materials was modified and adopted (Ku et al, 1997a; 1997b). The two mirror image test pieces are cut using a band saw from a tensile test piece for the material. Since the composites have discontinuous fibres, the gauge length is made at least twice the longest fibre length and four times the average fibre length (ATMS, 1990). A Shimadzu tensile testing machine was used for the tensile test. A load range of 2000N and a maximum load rate of 600 N per minute were selected for the test (Bolton, 1996). The results of the tests are described and discussed in chapter 10.6. The terms: adhesion, adhesives and adhesive system will also be discussed in detail later. Primers or fillers were used to enhance the absorption of microwave energy by the composite, or to be cured so as to form a bonding agent between the two pieces of material. From the pilot study (Liu et al, 1996; Ku et al, 1997a; 1999a) it seemed that graphite powder was not a very suitable filler because it flashed too easily, but araldite was suitable. Surface preparation was also an important consideration for the process to take place smoothly because it improves contact with the adhesive. The interfaces of the two test pieces were therefore roughened by rubbing them against a piece of coarse, grade 80, emery paper.

### 3.2 Butt Joint versus Lap Joint

The method of applying pressure to the two samples depended on the type of joint selected to bring the two parts together. If a butt joint were selected, the test pieces would be spring loaded and clamped together with a force of approximately 10N. On the other hand, if a lap joint was used then the two test pieces were brought together by a rubber band, which encircled on four times and provided a clamping force estimated to be 4 N. The strength performance of
joints depended largely on the type of loading they were subjected to. The butt joint, shown in Figure 3.2(a) was poor if it was subjected to tensile load (Everett, 1995); while the simple lap joint, shown in Figure 3.2(b) was good if it was under shear load. In this research, the butt joint was first selected for the joining process but the bond strength obtained was unsatisfactory and it was therefore abandoned (Ku et al, 1997a; 1997b). Waters (1996) recommended that butt joints should be avoided whenever adhesives are used as fillers in joining materials. The two fundamental rules that apply in adhesive joint design are to maximise the joint area and to make sure that the joint line is as parallel to the direction of the load as possible. This implies that lap joint has to be chosen to further improve the shear strengths of the bonding connection. Therefore, for most experimental work, the lap joint configuration was chosen. A stepped lap joint being regarded as very good is depicted in Figure 3.3(a), and a scarf joint (Waters, 1996) regarded as excellent is shown in Figure 3.3(b). They were also considered for use in ensuring research work.

![Figure 3.2: Butt and Simple Lap Joints](image-url)
3. Composite Materials Bonding Mechanisms and Adhesion

3.3 Adhesion

Adhesion refers to the attraction between substances. Adherends in this research were joined with or without primers. In both cases, pressure was applied to the substrates to promote coalescence. If a primer was used, an adhesive, liquid two-part rapid araldite was selected (Ku et al, 1997a; 1997b; 2000a). An adhesive is defined as a material which when applied to surfaces of substances can join them together and resist separation. In the forthcoming sections, the importance of considering the adhesive joint from the point of view of being an adhesive system is emphasised.

3.4 Primers

The first three factors (Everett, 1995) to be considered in using adhesives as primers are their shelf life, pot life and closed assembly time. Shelf life is the period of time for which an adhesive may be stored without deteriorating to the point that performance will be unsatisfactory. Pot life refers to the period of time after mixing a two- or three-component adhesive during which the mixture is still workable; whereas closed assembly time means the period of time after which the parts coated with adhesive, assembled and held together can no longer
be easily manipulated to improve the alignment or orientation of one part to the other (Schneberger, 1983). The shelf life of the two-part adhesive used, ie rapid araldite containing 100% liquid epoxy and 8 % amine (Ku et al, 2000a) was infinite. Provided that the two parts did not come into contact with one another (Selleys, undated); its pot life was only five minutes. Its closed assembly time has not been fully investigated yet it appeared that it was not much longer than its port life. The pros and cons (Everett, 1995; Waters, 1996; Black, 1996; Kinloch, 1995; Bolger, 1983.) of using adhesives, as fillers are numerous but only a few are discussed here. One of the advantages of adhesive bonding is that the loads imposed on the joint are spread evenly over the whole joint area. Another important advantage is that almost all engineering materials can be joined to one another by adhesives. For adhesives to function properly, they have to be able to spread over the solid surface, to displace air and contaminants that could be present on the surface, and to provide an intimate molecular contact with the substrates.

One of the shortcomings when applying an adhesive is the requirement for surface preparation. Surfaces have to be roughened to provide an improved adhesive key (Waters, 1996) and to be cleaned (Black, 1996) for consistent and good joint performance. Another drawback is the relatively low operating temperature for the final products. The maximum operating temperature for the adhesive used was only 160°C (Selleys, undated) but some other adhesives may have operating temperatures of up to 300°C (Black, 1996).

3.5 Mechanisms of Adhesion

Adhesion mechanisms refer to the various types of intrinsic forces, which can operate across the adhesive/substrate interface. There are mainly four possible mechanisms of adhesion, namely (Kinloch, 1995): mechanical interlocking, electronic theory adhesion, diffusion theory of adhesion and adsorption theory of adhesion. Whilst the adsorption adhesion has the widest applicability, each of the others could be appropriate in certain circumstances and often make a
contribution to the intrinsic adhesion forces which are acting across the interface. All of them are therefore be briefly discussed in the following sub-sections.

### 3.5.1 Mechanical Interlocking

The theory of mechanical interlocking proposes that mechanical keying or interlocking of the adhesive to the irregularities of the adherend surface is the major source of intrinsic adhesion (Kinloch, 1995). However, the attainment of good adhesion between two smooth surfaces has caused the mechanical interlocking theory to lose its wide applicability (Tabor et al, 1969; Israelachvili et al, 1972; Johnson et al, 1971). On the other hand, in the plating of plastics, mechanical interlocking has been proved to contribute significantly to the intrinsic adhesion of metal to polymer (Kinloch and Andrews, 1973; Andrews and Kinloch, 1973; Gent and Schultz, 1972; Wake, 1982). The roughening of the substrate surface has been wrongly regarded as providing cavities suitable for establishing mechanical interlocking with an adhesive. In fact the effect of mechanical abrasion in an industrial environment is to remove dirt and agents such as machine oil, grease and scale from the surface of the adherend, which obviously helped subsequent wetting. A boundary layer is removed which otherwise would fail prematurely when the joint is loaded. To summarise, in certain cases, mechanical interlocking can contribute to intrinsic adhesion. However, the surface of the substrates has to be specially pretreated to provide the geometry necessary for this to occur and this happens in only a few instances (Kinloch, 1995).

### 3.5.2 Electronic Theory of Adhesion

If the adhesive and substrate have different electronic band structures there is likely to be some electron transfer on contact to balance Fermi levels, which can result in the formation of a double layer of electrical charge at the interface. Deryaguin and co-workers (Deryaguin et al, 1957; 1969) have proposed the electronic theory of adhesion and suggested that electrostatic forces arising from
such contact could contribute significantly to the intrinsic adhesion. In certain specialised instances, the influence of an electrostatic double layer on intrinsic adhesion has been clearly demonstrated but research workers have proved that in most of the cases, the electrical double layer generated as a result of a typical adhesive/substrate interaction does not contribute much to intrinsic adhesion (Kilnoch, 1995).

3.5.3 Diffusion Theory of Adhesion

The diffusion theory (Voyutskii, 1963; 1962; 1963; 1965) states that the intrinsic adhesion of polymers to themselves (autohesion), and to each other, is due to mutual diffusion of polymer molecules across the interface. This requires that the macromolecules, or chain segments of the polymers (adhesive and substrate) possess sufficient mobility and are mutually soluble. The above conditions are usually met in the solvent welding of compatible, amorphous plastics, and in the autohesion of elastomers. However, where the solubility parameters of the materials are not similar, or one polymer is highly crosslinked, crystalline or below its glass transition temperature, the interdiffusion is an unlikely mechanism of adhesion (Kilnoch, 1995).

3.5.4 Adsorption Theory of Adhesion

This is the most widely applicable theory of adhesion and proposes that, provided sufficiently intimate molecular contact is achieved at the interface, the materials will adhere because of the interatomic and intermolecular forces which are established between the atoms and molecules in the surfaces of adhesive and adherend. Van der Waals forces are the most common of such forces and are referred to as secondary bonds. In addition, chemical bonds can be formed across the interface and these bonds are called primary bonds.
3.6 Concluding Remarks

From the discussion in the above section, it is not difficult to deduce that in certain circumstances any of the four mechanisms of adhesion can be responsible for the intrinsic adhesion that enables an interface to resist separation under the application of applied stress. Nevertheless, evidence clearly indicates that in most situations, the adsorption theory prevails. Surface analysis on the joined surfaces of the test pieces was carried out to verify one or more of the mentioned theories and details are given in section 9.3.
NOTE

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4. Microwave Energy and Its Interaction with Matter

4.1 Overview of Microwaves

Microwaves are electromagnetic waves with frequencies ranging from 300 MHz to 300 GHz, and typical frequencies used for industrial applications are 915 MHz, 2.45 GHz, 5.8 GHz, 24.124 GHz (NRC, 1994). Microwaves for industrial applications such as materials processing are generated by a variety of microwave sources, the commonest being magnetrons, power grid tubes, klystrons, klystrodes, crossed-field amplifiers, travelling wave tubes, and gyrotrons.

At the customary microwave frequencies, the magnetrons are the workhorse for industrial applications. These are tubes used in conventional microwave ovens found almost in every domestic application with varying power levels of the order of a kilowatt at a nominal frequency of 2.45 GHz, and industrial ovens with output up to a megawatt from a single unit. Magnetrons are termed crossed field devices because the basic interaction depends upon electron motion in electric and magnetic fields that are perpendicular to one another and thus crossed (NRC, 1994). In its most familiar embodiment, shown schematically in Figure 4.1, a cylindrical electron emitter, or cathode, is surrounded by a cylindrical structure, or anode, at a high potential and containing cavities capable of supporting microwave fields. Magnets are arranged to supply a magnetic filed parallel to the axis and hence perpendicular to the anode-cathode electric field. The interaction between electrons travelling in these crossed fields and microwave fields supplied by the anode, causes a net energy transfer from the applied DC voltage source to the microwave field. The interaction occurs continuously as the electrons traverse the cathode-anode region. The magnetron is the most efficient of the microwave tubes with efficiencies up to 90 percent having been achieved.

4.2 Microwave Technology for Joining

There are several mechanisms by which electromagnetic energy can be converted into heat. The two most common are current losses and dipole friction (Siores, 1994). The former will occur in any material which permits conduction current to flow, ie which has a dc conductivity. The latter occurs in substances which have a polar molecule with a permanent electric dipole, eg water. The orientation of these will oscillate under the influence of an applied alternating electric field. Energy is absorbed during this process, and this is referred to as polarisation loss or dipole
friction loss. Maximum absorption occurs at the relaxation frequency of the molecules, e.g., in the region of 17 GHz for water at room temperature. These mechanisms result in a relatively uniform heat distribution throughout the entire area exposed to microwave radiation, e.g., immediately in front of the rectangular or circular waveguide employed. The fast heating rates provide by microwave energy result in shorter processing time and hence greater energy efficiency. These advantages have led to the development of microwave configurations for joining a range of ceramic and polymer materials heterogeneously and autogenously. In the heterogeneous mode, various types of thermoplastic and thermosetting resins could be bonded with the aid of primers or fillers. For some polymeric materials, the peak bond strength could be up to 90% of the parent material. For some ceramic materials, primers or fillers need to be used in order to get bond strengths as high as 85% of the parent ceramic materials (Siores, 1994).

The following sections present a brief theoretical overview of the interaction of electromagnetic waves and hence microwaves with matter. Consideration was given to the transmission of microwaves in free space, along waveguides and within matter. Interaction at the fibre reinforced thermoplastic composite/air interface is also considered. Maxwell's Equations are used to analyse the interaction of microwaves with materials.
4. Microwave and Its Interaction with Matter

4.3 Microwave Propagation

Electromagnetic waves are propagated through empty space at the velocity of light. In such a wave, the time varying magnetic field can be regarded as generating a time changing electric field, which in turn generates a magnetic field and the process is repeated. Microwaves are electromagnetic waves that have a frequency range of 0.3 to 300 GHz with corresponding wavelengths ranging from 1 m to 1 mm respectively. Microwave propagation in air or through materials is dependent on the dielectric and the magnetic properties of the medium. The electromagnetic properties of a medium are characterised by complex permittivity ($\varepsilon$) and complex permeability ($\mu$)

$$\varepsilon = \varepsilon' - j\varepsilon''$$
$$\mu = \mu' - j\mu''$$

where $\varepsilon = \varepsilon'$ and $\mu = \mu'$, since the real part of $\varepsilon$ or $\mu$ is usually much larger than the imaginary part. Once the wave properties were calculated by this approximation, the values were improved by taking the imaginary part of $\varepsilon$ and $\mu$ as a perturbation.

4.3.1 Wave Equations and Solutions

In wave propagation, if $z$ is the direction of wave propagation and $t$ is the time, the amplitude of the electric field and that of the magnetic field varies sinusoidally in both $z$ and $t$. The frequency and the wavelength specify how a wave behaves in time and in distance. A plane wave front is a plane normal to $z$ with a uniform field in $x$ and $y$. The wave has an electric field $E_x$ in the $x$ direction and a magnetic field $H_y$ in the $y$ direction. For simplification, in this study, it is assumed that $\varepsilon = \varepsilon'$ and $\mu = \mu'$, since the real part of $\varepsilon$ or $\mu$ is usually much larger than the imaginary part. Once the wave properties were calculated by this approximation, the values were improved by taking the imaginary part of $\varepsilon$ and $\mu$ as a perturbation.

The wave equations for the electric and magnetic fields derived from Maxwell’s equations and utilised in this thesis are:
4. Microwave and Its Interaction with Matter

\[
\frac{\partial^2 E_x}{\partial z^2} = -\omega^2 \varepsilon_{o} \mu_{o} \varepsilon' \mu' E_x \quad (4.1a)
\]

\[
\frac{\partial^2 H_y}{\partial z^2} = -\omega^2 \varepsilon_{o} \mu_{o} \varepsilon' \mu' H_y \quad (4.1b)
\]

where \(\varepsilon_{o}\) and \(\mu_{o}\) are respectively the free space permittivity \((8.854 \times 10^{-12} \text{ F/m})\) and free space permeability \((1.256 \times 10^{-6} \text{ H/m})\) (Kraus, 1992).

Time dependence as \(\exp(\text{i} \omega t)\) has been assumed, where \(\omega = 2\pi f\), and \(f\) is the frequency in Hz.

The solutions to Eqn 4.1a and Eqn 4.1b for a wave travelling forward in the +z direction are respectively:

\[
E_x = E_0 e^{\text{i} \omega t} e^{-\text{j} 2\pi z / \lambda} \quad (4.2a)
\]

\[
H_y = H_0 e^{\text{i} \omega t} e^{-\text{j} 2\pi z / \lambda} \quad (4.2b)
\]

where the wavelength is

\[
\lambda = \frac{\lambda}{f} \quad (4.2c)
\]

and the velocity is

\[
v_p = \frac{1}{\sqrt{\varepsilon_o \varepsilon' \mu_o \mu'}} \quad (4.2d)
\]

and for a wave travelling backward (-z) are respectively:

\[
E_x = E_0 e^{\text{i} \omega t} e^{\text{j} 2\pi z / \lambda} \quad (4.3a)
\]

\[
H_y = H_0 e^{\text{i} \omega t} e^{\text{j} 2\pi z / \lambda} \quad (4.3b)
\]

The wave has a time-dependence governed by \(\omega\), where \(\omega\) is the angular frequency. a. It also has a z variation dependent on \(2\pi/\lambda\), which is called the wave number. If the
4. Microwave and Its Interaction with Matter

dielectric medium is lossy, a complex $\varepsilon$ must be used. Taking $\varepsilon = \varepsilon' - j\varepsilon''$ instead of $\varepsilon'$ in Eqn 4.1a, the wave solution for the forward wave is then:

$$E_x = E_0 e^{-\alpha x} e^{j\omega t} e^{-j2\pi z / \lambda}$$

(4.4)

where the attenuation constant is: $\alpha = \frac{1}{2}(2\pi / \lambda)\varepsilon'' / \varepsilon'$

Eqn 4.4 shows that the amplitude of the wave decreases exponentially as it propagates, ie wave energy is dissipated during the propagation.

For the isotropic medium considered here, one remarkable property of the wave is that it carries an equal amount of energy in electric and magnetic fields. The electric energy density equals to $\frac{1}{2}\varepsilon_0\varepsilon' E_x^2$. The magnetic energy density is $\frac{1}{2}\mu_0\mu'H_y^2$. Since they are equal, $\frac{1}{2}\varepsilon_0\varepsilon' E_x^2 = \frac{1}{2}\mu_0\mu'H_y^2$. Therefore,

$$\frac{E_x}{H_y} = Z_0 = \left(\frac{\mu_0\mu'}{\varepsilon_0\varepsilon'}\right)^{\frac{1}{2}}$$

(4.5)

where $Z_0$ is the wave impedance. This relationship characterises the transverse electric and magnetic field profile of the wave and is thus also known as the characteristic impedance. In air or vacuum, $Z_0$ equals $377 \Omega$. Another property these waves is that at any plane normal to $z$, the electric and magnetic field are in phase. The wave impedance is thus a real quantity and has the same value at any point along $z$ (Metaxas and Meredith, 1983).

4.3.2 Standing Waves

The two properties of wave propagation stated earlier are true as long as the forward and backward travelling waves are separate. However, these conditions no longer hold when both the forward and backward travelling waves exist simultaneously. Consider the case that a conducting plane is placed at $z = z_L$, causing reflection of a backward travelling wave. Using Eqn 4.2a and Eqn 4.3a as well as Eqn 4.2b and Eqn
4.3b, and considering the fact that the electric field has to be zero and the magnetic field is maximum at the conducting plane:

\[ |E_x| = 2E_0 \left| \sin \left[ 2\pi (z - z_L) / \lambda \right] \right| \]

\[ |H_y| = 2H_0 \left| \cos \left[ 2\pi (z - z_L) / \lambda \right] \right| \]

\[ Z = \frac{E_x}{H_y} = iZ_0 \tan \left[ 2\pi (z - z_L) / \lambda \right] \]

Both the electric and the magnetic fields are in the form of a standing wave in the sense that all the energy carried by the wave forward is reflected back and that the waves are neither moving the energy forward nor backward. In this case, the impedance, \( Z \), is no longer a constant but varies as a tangent function in \( z \) (Metaxas and Meredith, 1983).

### 4.3.3 Reflection and Transmission Coefficients

A reflection also occurs when there is a discontinuity in the dielectric or magnetic property of the medium. In this case only a portion of the wave is reflected. Consider the case in which a boundary separates two media with \( \varepsilon_1, \mu_1 \), and \( \varepsilon_2, \mu_2 \) and wave impedance \( Z_1 \) and \( Z_2 \), respectively. Let an incident wave with electrical field amplitude of 1V/m travel forward in medium 1 normal to the boundary (Figure 4.2). Both \( Z_1 \) and \( Z_2 \) are real quantities; the reflected wave in medium 1 has an amplitude \( \frac{K - 1}{K + 1} \) volt/m and the wave transmitted to medium 2 has amplitude of \( \frac{2K}{K + 1} \) volt/m, where \( K = \frac{Z_1}{Z_2} \). The ratio \( \frac{K - 1}{K + 1} \) is called the reflection coefficient and the ratio \( \frac{2K}{K + 1} \) is called the transmission coefficient. At the boundary the sum of the tangential electric or magnetic field of the incident wave and that of the reflected wave are equal to the tangential electric or magnetic field of the transmitted wave (Kraus, 1992).
4. Microwave and Its Interaction with Matter

4.3.4 Waveguide Modes

There are three types of electromagnetic waves: traverse electromagnetic (TEM) waves, traverse electric (TE or H) wave, and traverse magnetic (TM or E) waves. In a TEM wave, all fields are traverse, i.e., at right angles to the direction of travel and this provides an approximation of the radiation wave in space. Both TE and TM waves are waves in the waveguides. In a TE wave, the z component of the electric field is missing and in TM wave the z component of the magnetic field does not exist. Each TE and TM wave in a waveguide can have different field configurations. Indexes m and n identify each field configuration. $TE_{mn}$ and $TM_{mn}$ modes can exist in rectangular waveguides and $TE_{nl}$ and $TM_{nl}$ modes in cylindrical waveguides, where m, n and l were the order of the modes. The physical size of the waveguides limits the values of m, n and l to lower ones and thus the number of the modes that can propagate in a waveguide is limited. For the rectangular waveguides, the TM wave has the field components $E_x$, $E_y$, $E_z$, $H_x$, and $H_y$, and the TE wave has the field components $E_x$, $E_y$, $E_z$, $H_x$, and $H_y$. The diagram illustrates the reflection and transmission of a normal incidence electromagnetic wave.
components $E_x$, $E_y$, $H_x$, $H_y$, and $H_z$. The rectangular waveguide has a height ‘b’ in the y direction and a width ‘a’ in the x direction.

4.4 Propagation of Microwaves via a Rectangular Waveguide

Now let us consider the propagation of electromagnetic waves, including microwaves through a rectangular waveguide. Consider a pair of plane waves, as depicted in Figure 4.3. These waves can be used to demonstrate the transmission of microwaves through a rectangular waveguide, as used in the experimental work. In the figure, a pair of plane waves of equal frequency and amplitude is shown propagating along paths which converge with angle $2\theta$. Their electric fields are polarised normal to the figure. The solid lines show the position at a time of maximum positive electric field; broken lines represent the location at a time of maximum negative electric field. The resultant electric field at points of intersection between the two types of lines is zero. Figure 4.3 also suggests that if the two waves are considered at some later time, their maximum electric field would have travelled forward along their respective axes of propagation but the points of resultant zero field would move along a direction bisecting the angle of convergence of the two waves. There are several loci of zero resultant fields and two are shown spaced apart but parallel to each other. Since the electric field intensity is zero along these loci, it is feasible to insert conducting planes vertical to the plane of the figure without affecting the field.

By eliminating the fields outside the planes, a single wave travelling between a pair of parallel conducting plates can now synthesised. The waveguide wavelength $\lambda_g$ is longer than that of the plane waves forming it; by geometry $\lambda_g = \lambda_0 / \cos \theta$. If the two planes are set at this fixed distance apart and the wavelength is increased, the zero electric field condition would remain satisfied only if the angle of convergence $2\theta$ is increased, and as a result $\lambda_g$ increases more rapidly than $\lambda_0$. At $\theta = 90^\circ$, for example, it becomes infinite. This condition happens when $\lambda_0$ is equal to twice the separation distance between the planes, and this critical wavelength is the cutoff wavelength $\lambda_c$. At wavelengths greater than $\lambda_c$ the wave cannot propagate between the planes, and the fields attenuate rapidly with distance.
A second pair of planes at right angles to the first set is introduced to form a closed tube with axis coincident with the direction of propagation. These planes are now at right angles to the electric field vector (E) and satisfy the condition that E can have a finite value normal to the conducting plane. The tube formed is a rectangular waveguide and the field inside it is the simplest mode of propagation, which has the lowest frequency that can propagate freely in a rectangular waveguide. Its characteristic includes an electric field vector (E) with one component only, linearly polarised parallel to the narrow faces of the waveguide, with a half sinusoidal variation between them, and with a maximum value at the midpoint.

In the direction between the broad faces the electric field is of constant value. The magnetic field had two components forming closed loops. One component, $H_z$, is along the axis of the waveguide and the other, $H_x$, is parallel to the broad faces. All components have sinusoidal variations with time, and also with respect to their position along the waveguide. $E_y$ and $H_x$ have their maximum values at the same
position along the axis of the waveguide and are in phase in time, whereas component $H_z$ has its maximum value displaced a quarter wavelength away along the axis.

Other field patterns can also exist as the wavelength is reduced which have more than one half sinusoid of variation of electric field between the narrow faces. Other modes have a sinusoidal variation of electric field in a direction normal to the broad faces. All these modes are designated $H_{mn}$ or $TE_{mn}$ where the suffixes $m$ and $n$ indicate the number of half sinusoids of variation of electric field along the $x$- and $y$-axis respectively. The lowest mode is attained when $m = 1$ and $n = 0$ giving the $H_{10}$ or $TE_{10}$ dominant mode. Traverse electric and magnetic fields are related by the wave impedance:

$$Z_{so} = \frac{|E_z|}{|H_y|} = f\mu_0\lambda_g$$

(4.7)

The propagation of an $H_{10}$ wave in a rectangular guide is not be discussed here in detail but the field pattern of the $H_{10}$ wave in rectangular waveguide is shown in Figures 4.4 (2-D) and 4.5 (3-D) respectively (Glazier and Lamnont, 1958).
4. Microwave and Its Interaction with Matter

4.5. Microwave/Matter Interactions

The nature of the above characteristics change (Sabburg, 1993) as the microwaves interact with matter. As far as electromagnetic/matter interactions are concerned, materials can be classified as absorber, transparent and opaque (Sutton, 1989). The results of microwaves/materials interactions are shown in Figure 4.6. High energy rate joining of thermoplastic composites using microwave is studied here because it is believed that the microwave/materials interactions of some of thermoplastic composites with and without fillers would favour the process.

Materials that are amendable to microwave heating are polarisable and have dipoles that reorient rapidly in response to changing electric field strength. If these materials possess low thermal conductivity combined with dielectric loss that increases significantly as the temperature increases, hot spots and thermal runaway could be experienced. Three processes could represent the interactions between microwaves and materials: charges movement due to electronic conduction, ionic conduction and rotation of electric dipoles. The processes can be illustrated in combination by considering materials that have an electronic conductivity $\sigma_e$, an ionic conductivity $\sigma_i$. 

![Figure 4.5: Line Representation (3-D) of the Fields of the TE$_{10}$ Mode in a Rectangular Waveguide](image-url)
and complex permittivity, $\varepsilon' - j\varepsilon''$. In the presence of an electric field, $E$, a current has to flow.

According to the Maxwell’s equations, the current density $j$ is:

$$J = [\sigma_e + \sigma_i + j\omega\varepsilon_o (\varepsilon' - j\varepsilon'')]E$$

$$= j\omega\varepsilon_o \varepsilon'E + \omega\varepsilon_o \varepsilon'\tan\delta E$$  \hspace{1cm} (4.8)

where

$$\tan\delta = \frac{\sigma_e + \sigma_i + \varepsilon''}{\omega\varepsilon_o \varepsilon'}$$  \hspace{1cm} (4.9)

The phase angle, $\delta$, relates to the time lag involved in a polarising material. Again it is pointed out that the loss tangent is the most important parameter in processing materials. The first term at the right hand side of Eqn 4.8 is the component of the current 90 degrees out of phase with the electric field. It is the displacement current that stores electric energy in the material. The average electric energy stored per unit volume is

$$W = \frac{1}{2}\varepsilon_o \varepsilon'E^2 \ (J/m^3)$$  \hspace{1cm} (4.10)
The second term at the right hand side of Eqn 4.8 is the component of the current that is in phase with the electric energy. Through this term the microwave energy is converted into heat energy for material processing. The average power per unit volume converted into heat is,

\[ P = \frac{1}{2} \omega \varepsilon'_0 \varepsilon' \tan \delta \ E^2 \ (\text{W/m}^3) \]  \hspace{1cm} (4.11)

Hence, the loss tangent characterises the ability of the material to convert absorbed microwave energy into heat with the degree of absorption being dependent on electric-field intensity, frequency, loss factor, and permittivity. The material properties of greatest importance in microwave processing of a dielectric are the complex relative permittivity \( \varepsilon = \varepsilon' - j \varepsilon'' \) and the loss tangent, \( \tan \delta = \varepsilon'' / \varepsilon' \) (NRC, 1994). The real part of the permittivity, \( \varepsilon' \), sometimes called the dielectric constant, mostly determines how much of the incident energy is reflected at the air-sample interface, and how much enters the sample. The loss tangent, \( \tan \delta \), predicts the ability of the material to convert the absorbed energy into heat. For optimum microwave energy coupling, a moderate value of \( \varepsilon' \), is needed to enable adequate penetration, and it should be combined with high values of \( \varepsilon'' \) and \( \tan \delta \), to convert microwave energy into thermal energy.

When the conductivity of a material is very large, the fields attenuate rapidly toward the interior of the sample due to the skin effect. The skin effect is related to the magnetic properties of the material. When a large current flows inside the sample due to high conductivity, a combination of the magnetic field with the current produces a force that pushes conducting electrons outward into a narrow area adjacent to the boundary. The effective extent of this flow is called the skin depth, \( \delta \). By definition the skin depth is the distance below the conductor surface at which the electric-field strength is reduced to \( \frac{1}{e} \) (Risman, 1991). \[ \delta = \frac{1}{\frac{1}{2} \omega \mu \mu'_0 \sigma} \ (\text{m}) \]
The penetration depth is defined as the distance from the surface of the material at which the power drops to $e^{-1}$ from its value at the surface (Metaxas and Meredith, 1983), ie

$$\delta_p = \frac{1}{2\alpha}$$  \hspace{1cm} (4.12)

where $\alpha$ is the attenuation factor.

At 2.45 GHz, the skin depth for a conductor eg brass is 2.6 $\mu$m, and that of an insulator, eg alumina is 187 m. When the skin depth is larger than the dimension of a sample, the effect can be neglected. Conversely, penetration of microwave energy is very limited and uniform heating is impossible. For a material having a polar molecule, eg water, the real and imaginary parts of the permittivity varies with frequency as shown in Figure 4.7. Because limited depth of penetration, it is not be possible to work at the relaxation frequency. Hence the choice of 2.45 GHz for domestic microwave heating.

**Figure 4.7: Dielectric Relaxation of Typical Polar Dielectric**
5. Experimental Equipment - Microwave Facilities Configuration

5.1 Overview

This chapter includes the design of a special microwave, single mode applicator for joining thermoplastic composites; the creation of a standing wave inside and along the applicator by the incident and reflected power of the system. The maximisation of electric field at the seam of the lapped area of the test pieces and the minimisation of microwave radiation leakage which was sought by replacing a physical short-circuit with a virtual one in the plunger are also the main aims of this chapter. With reference to Figure 5.1, the incident waves were generated by the magnetron. They travelled downward through three sections of WR340 waveguide and interacted with the test pieces located in the second section before being reflected back by the top face of the adjustable plunger. The plunger was designed and manufactured to have a sliding fit contact with the waveguide. The interaction between the incident and the reflected waves set up a standing wave (Glazier and Lamont, 1958), which could be arranged so that for the maximum electric field occurred at the seam of the butted joint or the effective area of the lapped test pieces. This was achieved by adjusting the moveable piston so that its top face was an odd multiple of \(\frac{\lambda_g}{4}\) from the centre of the slit; where \(\lambda_g\) was the wavelength within the waveguide.

5.2 Short-Circuit Plunger and Its Dimensions

The relationship between the wavelength within the guide, \(\lambda_g\) and the free space wavelength, \(\lambda_o\) applicable to the above mentioned experimental set up is as follows (Glazier and Lamont, 1958):

\[
(1/ \lambda_g^2) = (1/ \lambda_o^2) - (1/2a)^2
\]  

(5.1)

where ‘a’ is the larger of the internal dimension of the waveguide and was in mm. Using, the relationship

\[
c = f \lambda_o
\]  

(5.2)
where \( c \) was the velocity of light \( \approx 3 \times 10^8 \) ms\(^{-1}\)

and \( f \) was the frequency used in this project \( = 2.45 \) GHz,

therefore, \( \lambda_o = \frac{3 \times 10^{11}}{2.54 \times 10^9} = 122.45 \) mm.

For the waveguide used, WR340, \( a = 86.36 \) mm and \( b = 43.18 \) mm,
therefore, \( \lambda_g = 173.63 \) mm and \( \lambda_{g/4} = 43.41 \) mm.

Referring to Figure 5.2, the distance between the centre of the slit and the top face of the plunger, \( l \), was initially 286 mm. To create a maximum electric field, \( l \) had to be varied so that \( l = n \times \lambda_{g/4} \) and \( n \) was an odd integer. If \( n = 7 \) was chosen then \( l = 7 \times \lambda_{g/4} = 7 \times 43.408 \) mm = 303.9 mm. The distance, \( l \), could be varied by adjusting the plunger up and down by rotating the knob at the bottom of the plunger. Hence, in order to have a maximum of electric field at the specimen, the plunger was adjusted by 303.9 mm - 286 mm = 17.9 mm in the downward direction. This was the position of the electric field maximum when there was no test piece at room temperature and formed part of the initial set-up.

Inserting a dielectric in the waveguide alters the electric field configuration.
Leakage of power past the plunger was to be expected because of the sliding fit, and measures had to be taken to prevent it. It was minimised by using a non-contact method to produce an apparent short circuit at the front face of the plunger. The actual point of contact was arranged to be at a point where the current was effectively zero. This was accomplished as shown in Figure 5.3. At a distance of $\lambda/4$ from a short circuit (Glazier and Lamont, 1958), the traverse electric field, $E$ had a maximum value and the traverse magnetic field $H$ was zero. Thus the ratio of traverse components $E/H$ was infinite at this point and hence the input wave impedance of a $\lambda/4$ length of waveguide closed at its far end was infinite. Similarly, the input impedance of a $\lambda/2$ length was zero. In practice, however, neither the infinite nor the zero values can be attained.

In Figure 5.3, the part ABC was regarded as a narrow waveguide of length $\lambda/2$ short-circuited at C and folded at its centre, B. Thus its input impedance at A was zero, and there was an equivalent short-circuit at the front face of the piston, which appears as a continuous short-circuit across the waveguide. The sliding contact occurred at B, which was $\lambda/4$ from the end point C. At point B, the impedance was high with a small magnetic field applied (Glazier and Lamont, 1958) and the current was therefore small. As a result, radiation leakage through
the sliding contact was extremely small. Variations in contact resistance with movement of piston were of little importance. Here, actual physical short-circuits were replaced by virtual short-circuits and physical contact was made at a place where poor contact was not critical.

An example of a moveable plunger used are shown in Figure 5.4, where, $b = 20$ mm, $b_1 = 0.6$ mm $b_2 = 4.0$ mm (Rizzi, 1988).

Therefore, for present application in WR340 waveguide, when $b = 43.18$ mm, then

\[ b_1 \text{ by proportion should be } \frac{0.6}{20} \times 43.18 \text{ mm} = 1.30 \text{ mm and similarly} \]

\[ b_2 = 8.636 \text{ mm}. \]

Since the plunger should have a sliding contact with the waveguide, the clearance between the two was made to be 0.5 mm. The angle of $45^\circ$ was chosen for convenience of manufacture. The dimensions of the plunger are as shown in Figure 5.5.
Figure 5.4: An Example of Non-contact Type Adjustable Short Circuit in Waveguide

Figure 5.5: Dimensions of the Moveable Plunger
5.3 Duty Cycle of the Magnetron

There were 4 power level settings in the microwave controller system used: 100%, 80%, 50% and 30%, which were achieved by modifying the duty cycle during a 12 seconds period. The magnetron operation involved two (2) phases. The first was a transient phase and consisted of the cathode warming up, which took around one (1) second after which electrons were emitted. The next phase was a steady state phase when microwaves were generated. The duty cycle comprised two phases: “ON” phase and “OFF” phase. The “ON” phase of a duty cycle involved the transient phase (TP), plus the steady state operation (SS), plus the shut down period (SP) of the magnetron, the latter of which was very rapid and approached zero. The “OFF” phase meant power was not supplied to the magnetron.

At the 30% power setting, and for 15 seconds duration, the operation was as follows:
“ON” phase: TP + SS + SP; TP = one (1) second; SS = 30% x 12 seconds = 3.6 seconds; SP = 0 second.

Therefore, the heating cycle composed of five (5) seconds “ON” followed by seven (7) seconds “OFF” and followed by three (3) seconds “ON”. The total time was fifteen (15) seconds and the last three (3) seconds were merely a fragment of the next duty cycle. The 30% setting therefore referred to a percentage of the duty cycle and NOT the continuous microwave output power. It was 100% power delivered 30% of the time, but the average power was 30% of the full power. Over longer duration, the time average power output would approach 30%, but for short programme times, the researcher has to be aware of the difference between continuous wave output power and the duty cycle approach. The time weighted average power output between the two approaches could differ by as much as 70%.
5.4 Risk Assessment

Lately, significant changes have been made to the Australian Workplace Health and Safety Act with regard to hazardous substances. The changes have included more stringent requirements for registration of substances, risk assessment, record keeping and, in some instances, the need for health surveillance. Of the risks involved in this research, radiation leakage from the equipment could be by far the most worrying to people nearby. Careful measures were taken to cope with the problem. First, the magnetron was enclosed in a metal case on top of the microwave oven cavity enclosure so that microwaves could only travel downwards to and through the three sections of the waveguide to the top face (reflector) of the plunger. Virtually, no radiation could leak through the case to the surroundings. Power leakage was suspected and would be most serious through the slits into which the test pieces were inserted, but it did not propagate to the environment because it was contained inside the enclosure of the microwave oven cavity. Another possible leakage point was the sliding fit between the moveable plunger and its sleeve as shown in Figure 5.3, but the problem was minimised by the non-contacting short circuit technique described previously. The end of the plunger waveguide was also covered up by a metal plate and radiation leakage to the surrounding was virtually zero. However, in order to check that the equipment complied with the recommended exposure limit of 10mW/cm² (Jacques, 1992; NRC, 1994), measurements of radiation leakage were made using a power meter, power sensor and an antenna. A coaxial to waveguide (WR 340) adaptor was used as the antenna. The set-up is shown in Figure 5.6.

Measurements had to be taken more than a certain distance away from the microwave source and this was called far-field distance (Balanis, 1992).

\[
\text{Far-field distance} \approx \frac{2D^2}{\lambda_o} \quad (5.3)
\]

where D is the larger dimension of the antenna and in this case it was

3.4 x 25.4 mm = 86.36 mm
and from Eqn 5.1 and Eqn 5.2, \( \lambda_0 = 122.45 \) mm.

The far-field distance was therefore
\[
\frac{2(86.36)^2}{122.45} = 121.8 \text{ mm.}
\]

Measurements were taken at a distance of one (1) metre radius around the equipment, where an operator is most likely to stand. That distance satisfied the far-field distance requirement of the antenna of 121.8 mm. Measurements were taken at 30% and 50% power levels. The average range of the meter reading at the 30% power level was 10 - 57 \( \mu \text{W} \). That at the 50% power level was 22 - 73 \( \mu \text{W} \). Since power received (Balanis, 1997) from a plane wave is given by:

\[
\text{Power received} = (\text{Effective aperture}) \times (\text{Power density})
= (\text{Aperture efficiency}) \times (\text{Physical aperture}) \times (\text{Power density})
\]

(5.4)

For a rectangular waveguide aperture, the aperture efficiency is approximately 0.81 or 812% (Balanis, 1997). Hence:

\[
\text{Power density (W/cm}^2) = \frac{\text{Power received (W)}}{0.81 \times \text{ab}}
\]

(5.5)

where \( a = 8.636 \) cm and \( b = 4.318 \) cm.
Therefore, at 30% power level, and if the maximum value of power in the range was taken for calculation:

\[
\text{Maximum power density} = \frac{57 \times 10^{-6}}{0.81 \times 8.636 \times 4.318} = 1.819 \times 10^{-3} \text{ mW/cm}^2.
\]

Similarly, at 50% power level, the maximum power density is \(2.42 \times 10^{-3}\) mW/cm\(^2\).

In addition, the power received by the antenna was at its maximum if the electric field was parallel with the aperture of the antenna as shown in Figure 5.7. Measurements were also taken by varying the angles of the antenna so that the antenna had its maximum sensitivity and hence the maximum electric field could be recorded. All measurements showed that the power leakage was much less than 10 mW/cm\(^2\), ie, the safety limit. The leakage from the sliding fit of the plunger is suspected to be the highest contribution. When the antenna was located at 300 mm from the end of the sleeve of the piston, a range of readings of 0.53 mW/cm\(^2\) to 0.86 mW/cm\(^2\) were recorded, which were well below the risk level.
Figure 5.7: Antenna Orientation in Leakage Measurement

On top of all of the above safety measures, it was satisfying to know that the nonionising bands of the electromagnetic radiation, ie from DC to visible light did not carry enough energy to break any chemical bonds (Redhead, 1992; NRC, 1994). The only effects of nonionising radiation in the microwave region on human tissue are those derived from the energy – matter coupling mechanisms, particularly dielectric coupling. At present, the only confirmed effect is warming, from the conversion of electromagnetic energy to heat.

The glass fibre (GF) and carbon fibre (CF) filaments were embedded in the polymers and hence all the possible inhalation leakage risks (Luce, 1988; ACI, 1987; Gougeon, 1996) associated with them did not apply in this case.

The next item to be considered was the graphite powder. Graphite powder could emit some dust when sprayed onto the test pieces. The powder could cause irritation to the eyes and when inhaled it could lead to respiratory irritation. Goggles, disposable protective gloves and masks had therefore to be worn when spraying was carried out. As the quantity used was very small, the problem was not that serious. It could, however, flash when exposed to microwave energy of 240 W for a period of 6 seconds or more and this in turn could set some types of polymer alight. A compressed air system was therefore on standby so that if a fire was ignited, it could be put out immediately. A powder foam fire extinguisher was therefore installed in the nearby area for use in an outbreak of fire incident.

The next item to be discussed is the GF reinforced (33%) nylon 66; no adverse health effects should occur if the product is handled in accordance with the product label. The test pieces were injection-moulded products and were not granules, so they carried none of the risks (ICI Plastics, undated) associated with granules. However, exposure to the molten material could cause burns and it could adhere strongly to the skin. The material was combustible but had certain self-extinguishing properties (ICI Plastics, undated). The decomposed products included ammonia and carbon monoxide, so exposure to it had to be in
accordance with the National Occupational Health and Safety Commission (ICI Plastics, undated). Tests were therefore carried out in a ventilated area. The next material to be mentioned is polystyrene (PS). Again, the test pieces were moulded products and the inherited risks associated with the granules did not apply here (Dow Chemical, 1994). The polymer would burn when supplied with sufficient heat and oxygen and would emit toxic fumes when being with insufficient oxygen burned (Dow Chemical, 1994). The test area was therefore well ventilated. Personal protection measures were also followed while joining the material. The last material to be discussed here is low-density polyethylene (LDPE), which would burn in the presence of extreme heat and oxygen (Kemcor, 1995). The same treatment as above had to be carried out for skin contact and adherence by the molten material. Personal protection rules were therefore obeyed (Kemcor, 1995).

5.5 Concluding Remarks

This chapter has discussed the design of a special microwave, single mode applicator for joining thermoplastic composites. A standing wave was created inside and along the applicator by the incident and reflected power of the system. The maximum of the electric field was designed to occur at the seam of the lapped area of the test pieces. Any radiation leakage was avoided and a virtual short-circuit was designed to replace the physical short-circuit used in order to minimise microwave leakage and bring it down to below the acceptable levels.
6. Permittivity Measurement at Various Frequency and Elevated Temperatures

6.1 Introduction

Since $\varepsilon'$, $\varepsilon''$ and $\tan \delta = \frac{\varepsilon''}{\varepsilon'}$ are vital in microwave processing of materials and those electrical properties vary significantly with temperature and frequency, they were measured with the aid of a network analyser at elevated temperatures and in the frequency range 2.2 GHz to 12.5 GHz. The real part of the permittivity, $\varepsilon'$, sometimes called the dielectric constant, mostly determines how much of the incident energy is reflected at the air-sample interface, and how much enters the sample. This chapter shows the derivation of the reflection coefficient of a material, $\rho \approx \frac{(\sqrt{\varepsilon'} - 1)}{(\sqrt{\varepsilon'} + 1)}$ and the depth of penetration of a dielectric, $\delta_p \approx \frac{2\sqrt{\varepsilon'}}{\omega\varepsilon_0^* \mu_0 \varepsilon_0}$. Therefore, the larger the value of the real part of the complex permittivity, the more incident energy will be reflected by a dielectric, but the energy that enters the material will penetrate further than in a dielectric with the same $\varepsilon''$ but lower $\varepsilon'$. In addition, the loss tangent, $\tan \delta$, which predicts the ability of the material to convert the penetrating energy into heat needs to be taken into consideration. Measurements of $\varepsilon'$ and $\tan \delta$ are therefore critical in the microwave processing of materials with or without primers. The dielectric constant, $\varepsilon'$, dielectric loss, $\varepsilon''$, and hence complex relative permittivity, $\varepsilon$ and loss tangent, $\tan \delta$, of some commonly used thermoplastics have been measured (Osswald, 1995; Lynch, 1992; Shackelford, 1992) at various temperatures and frequencies. Such studies have provided information regarding the suitability for microwave processing of certain fibre-reinforced thermoplastic (FRTP) composites. Few measurements, if any, of the complex relative permittivity or the loss tangent of FRTP composites have been reported in the literature. This chapter describes a convenient and inexpensive laboratory method to obtain $\varepsilon'$, $\varepsilon''$ and hence $\tan \delta$. The method employs a network analyser and the technique is called the waveguide transmission technique, chosen because it
provides the widest possible frequency range with high accuracy. The hardware and software needed for the method was available at the University of Southern Queensland. The required data were collected at a range of elevated temperatures and over a band of frequencies. Results were also compared with those obtained by HP 85070B High-Temperature Dielectric Probe Kit (HP85070B Technical data Sheets, 1993; HP85070A Technical data Sheets, 1990). The relationship between \( \varepsilon' \) and \( \varepsilon'' \) or \( \tan \delta \) of the composites and their weldability by microwave energy were also studied, and are discussed in this chapter.

The use of microwave high-energy rate joining of FRTP composites has shown promising preliminary results (Liu et al, 1996; Ku et al, 1997a; 1997b; 1998; 1999a; 1999b; 1999c; 2000a) but more research work is needed so that the technology can find applications in manufacturing industry. Values of \( \varepsilon \) and \( \tan \delta \) of FRTP composites have rarely been reported in the literature and further more the recorded figures were at room temperature only. As these electrical properties varied significantly with rising temperature and frequency, values attained at room temperature cannot be appropriately used to predict the microwave-reactiveness of thermoplastic composites at elevated temperatures. This chapter describes the measurement of \( \varepsilon \) and \( \tan \delta \) of three types of random, 33% by weight, glass-fibre reinforced thermoplastic composites, and of three kinds of Araldite used as priming agents in the joining of the above three types of fibre-glass reinforced materials using microwave energy. The three thermoplastic matrices chosen for the study were the three most commonly used thermoplastics, namely, low density polyethylene (LDPE), polystyrene (PS) and nylon 66. The limited heat tolerance of the network analyser prevented measurements of the complex relative permittivity being taken at temperatures higher than 100°C in the frequency range 2.2 GHz to 12.5 GHz, which covers most industrial applications of microwaves.
6.2 Different Measurement Methods for Permittivity

Extensive methods (Ness, 1983; Sabburg et al, 1992; Senko and Tran, 1997) have been reported in the literature for the measurement of \( \varepsilon \) of different materials. A dielectric probe kit was available for use in conjunction with a network analyser (HP85070A Dielectric Probe Kit Data Sheet, 1990). The probe was placed against the material under test and \( \varepsilon \) was calculated from the reflection coefficient. Sequeria (1991) employed a one-port network analyser reflection measurement on multiple length samples to determine both \( \varepsilon \) and \( \mu \). Pham (1991) developed an in-situ monopole antenna probe and a numerical technique which extended the measurement frequency range for the measurement of \( \varepsilon \). Ness (1983; 1985) used two-port transmission measurements made by a semi-automatic network analyser via a waveguide and the same method (Sabburg et al, 1992) was used to measure the \( \varepsilon \) of different types of Australian soil materials. This formed the basis for the measurement system used in this thesis. A waveguide transmission technique is a convenient and inexpensive laboratory method that employs a network analyser to obtain \( \varepsilon \) for a length of sample filled waveguide. The complex permittivity is calculated off-line from transmission coefficient (S\(_{21}\)) data from the network analyser. From this measurement the sample dielectric constant, dielectric loss and loss tangent are calculated.

6.3 Reflection Coefficient and Depth of Penetration

To start with consider the propagation coefficient for a TEM wave in a lossy material (Glazier and Lamont, 1958):

\[
\gamma = \alpha + j\beta
\]

(6.1)

Then

\[
\gamma^2 = j\omega\mu_0\sigma - \omega^2\mu_0\varepsilon_0\varepsilon
\]

(6.2)
Permittivity Measurement at Various Frequency and Elevated Temperatures

The penetration depth is the depth at which the field strength has fallen to \( \frac{1}{e} \) times the surface value:

\[
\delta_p = \frac{1}{\alpha}
\]  

(6.3)

where \( \gamma \) is the complex propagation coefficient;
- \( \alpha \) is the attenuation coefficient;
- \( \beta \) is the phase coefficient or phase shift per unit length;
- \( \omega \) is the angular frequency;
- \( \mu \) is the permeability;
- \( \sigma \) is the conductivity;
- \( \varepsilon \) is the complex relative permittivity, \( \varepsilon = \varepsilon' - j\varepsilon'' \)

\( \delta_p \) is the penetration depth.

Equating Eqn 6.1 and Eqn 6.2 gives

\[
\gamma^2 = (\alpha + j\beta)^2 = j\omega\mu_0\sigma - \omega^2\mu_0\varepsilon_0\varepsilon
\]

(6.4)

Assuming no conduction loss in the material, ie \( \sigma = 0 \), Eqn 6.4 then becomes

\[
\alpha^2 + 2j\alpha\beta - \beta^2 = -\omega^2\mu_0\varepsilon_0(\varepsilon' - j\varepsilon'')
\]

or

\[
\alpha^2 + 2j\alpha\beta - \beta^2 = -\omega^2\mu_0\varepsilon_0\varepsilon' + j\omega^2\mu_0\varepsilon_0\varepsilon''
\]

(6.5)

The losses in glass fibre reinforced materials are small, so that to a first approximation loss may be neglected.

For a lossless material, \( \alpha = 0 \) and \( \varepsilon'' = 0 \) and by equating the real parts, Eqn 6.5 becomes

\[
-\beta^2 = -\omega^2\mu_0\varepsilon_0\varepsilon'
\]

or

\[
\beta = \omega\sqrt{\mu_0\varepsilon_0\varepsilon'}
\]

(6.6)

The reflection coefficient at normal incidence is given by (Kraus, 1992):
where $Z_{OS}$ is the intrinsic impedance of the material, equals to $j\omega\mu_0/\gamma$,
and for a lossless material $Z_{OS} = j\omega\mu_0/j\beta = \omega\mu_0/\beta$ (6.8)

Substituting Eqn 6.6 into Eqn 6.8 gives $Z_{OS} = \frac{\mu_0}{\sqrt{\mu_0\varepsilon_0\varepsilon'}} = \frac{1}{\sqrt{\varepsilon'}}$ (6.9)

Since, the intrinsic impedance of air is $Z_0 = \frac{\sqrt{\mu_0}}{\sqrt{\varepsilon_0}}$, (6.10)

By substituting Eqn 6.9 and Eqn 6.10 into Eqn 6.7 it can be deduced that

$$
\rho = \frac{1}{\sqrt{\varepsilon'}} - \frac{1}{\sqrt{\varepsilon'}} \approx \frac{1}{\sqrt{\varepsilon'}} - \frac{1}{\sqrt{\varepsilon'}}
$$

Therefore, reflection coefficient increases with $\varepsilon'$ and when $\varepsilon'$ is very large, $|\rho| \rightarrow 1$ ie total reflection.

The matrix materials are all good insulators at DC. If there is no conduction loss, ie $\sigma = 0$ then all losses will be polarisation losses. By equating the real and imaginary parts of Eqn 6.5:

$$
\beta^2 - \alpha^2 = \omega^2\mu_0\varepsilon_0\varepsilon' \text{ and } 2\alpha\beta = \omega^2\mu_0\varepsilon_0\varepsilon''.
$$

After re-arranging, the above,

$$
\frac{\beta^2}{(\omega^2\mu_0\varepsilon_0)} - \frac{\alpha^2}{(\omega^2\mu_0\varepsilon_0)} = \varepsilon'
$$

and

$$
\frac{2\alpha}{\omega\sqrt{\mu_0\varepsilon_0}} = \frac{\beta}{\omega\sqrt{\mu_0\varepsilon_0}} = \varepsilon''
$$

Therefore, Eqn 6.13 gives

$$
\frac{\beta}{\omega\sqrt{\mu_0\varepsilon_0}} = \frac{\varepsilon''}{\frac{2\alpha}{\omega\sqrt{\mu_0\varepsilon_0}}}
$$

Let

$$
\frac{\alpha}{\omega\sqrt{\mu_0\varepsilon_0}} = x
$$

and

$$
\frac{\beta}{\omega\sqrt{\mu_0\varepsilon_0}} = y
$$
Substituting Eqn 6.14 and Eqn 6.15 into Eqn 6.12 and Eqn 6.13 respectively the following can be obtained

\[ y^2 - x^2 = \varepsilon' \] \hspace{1cm} (6.16)

\[ y = \frac{\varepsilon''}{2x} \] \hspace{1cm} (6.17)

Substituting Eqn 6.17 into Eqn 6.16 gives

\[ \left(\frac{\varepsilon''}{2x}\right)^2 - x^2 = \varepsilon' \] \hspace{1cm} (6.18)

After re-arranging, Eqn 6.18 becomes \( \frac{\varepsilon''}{2} - x^4 = \varepsilon'x^2 \)

and by further re-arranging, it gives \( x^4 + \varepsilon'x^2 - \left(\frac{\varepsilon''}{2}\right)^2 = 0 \).

Therefore,

\[ x^2 = \frac{-\varepsilon' \pm \sqrt{(\varepsilon')^2 + 4(\varepsilon'')}}{2} \]

Since \( x^2 \) has to be positive, only the positive sign needs to be chosen.

Therefore,

\[ x^2 = \frac{-\varepsilon' + \sqrt{(\varepsilon')^2 + 4(\varepsilon'')}}{2} \] \hspace{1cm} (6.19)

Since \( \tan \delta = \frac{\varepsilon''}{\varepsilon'} \), Eqn 6.14 becomes

\[ \alpha^2 = \frac{1}{2} \omega^2 \mu_0 \varepsilon_0 \varepsilon \left[ \sqrt{1 + (\tan \delta)^2} - 1 \right] \] \hspace{1cm} (6.20)

For small loss tangent and by using the Binomial Theorem to expand the square root, an approximate solution can be derived to bring Eqn 6.20 to

\[ \alpha^2 \approx \frac{1}{2} \omega^2 \mu_0 \varepsilon_0 \varepsilon' \left[ 1 + \frac{1}{2}(\tan \delta)^2 + \ldots \right] \] or

\[ \alpha^2 \approx \frac{\omega^2 \mu_0 \varepsilon_0 \varepsilon'}{4} (\tan \delta)^2 \] and

\[ \alpha \approx \omega \sqrt{\mu_0 \varepsilon_0 \varepsilon} \frac{\tan \delta}{2} \] or \( \alpha \approx \omega \sqrt{\mu_0 \varepsilon_0 \varepsilon} \left( \frac{\varepsilon''}{2 \varepsilon'} \right) \).

Therefore penetration depth or skin depth, \( \frac{1}{\alpha} = \delta_p = \frac{2 \sqrt{\varepsilon'}}{\omega \varepsilon' \sqrt{\mu_0 \varepsilon_0}} \) for \( \tan \delta << 1 \).
Hence for good penetration, $\varepsilon'$ should be large and $\omega$ and $\varepsilon''$ should be small, i.e. small loss tangent. In short, the reflection coefficient increases with $\varepsilon'$ and the penetration depth of the material increases with $\varepsilon'$. So, a sample with high $\varepsilon'$ reflects more of the incident energy, but the energy which enters the sample will penetrate further (than in a sample having the same $\varepsilon''$ but lower $\varepsilon'$).

### 6.4 Calibration of the Network Analyser

The object of a network analyser is to approximate a perfect measurement system having an indefinite dynamic range, isolation, and directivity characteristics, no impedance mismatches in any part of the test set-up, and with a flat frequency response. The ‘perfect’ network analyser set-up was achieved by measuring the magnitude and phase response of known standard devices. Using this data in conjunction with a model of the measurement system to determine error contributions and using vector mathematics to compute the actual test device response by removing the error terms, accurate and reproducible measurements are obtained. The first step in using the equipment is to input the range of frequencies over which $\varepsilon$ is to be measured and to calibrate the equipment accordingly (HP8410B Hewlett Packard Product Information, 1980). Two ports, port 1 and port 2, of the equipment were calibrated using three types of standards, namely, short circuit, offset short circuit and matched termination. The short circuit was first connected to the first port for calibration. During the calibration process, the operator observed the progress of the calibration on the analyser screen. After that the short circuit was linked to the second port for the same purpose. The next standard used was the offset short circuit and acceptable calibration was similarly observed as in the previous case. The matched termination was used to carry out the final calibration. The network analyser was then instructed to calculate the reflection coefficients, $S_{11}$ or $S_{22}$ from the calibration data. The next stage was to couple the two waveguide to coaxial adaptors together so that the transmission coefficients, both forward and backward ($S_{21}$ and $S_{12}$) could be calculated. Equipment then computed the transmission coefficients, which were then saved in a register for later retrieval.
6.5 Theory of Waveguide Transmission Technique

The waveguide transmission technique was a convenient laboratory based method which employed a network analyser to obtain the ε of a length of sample filled waveguide. The ε value was calculated at a spot frequency off-line from manual readout of limited transmission coefficient, S21 or S12, data from the network analyser. Since the transcendental equation used in the off-line calculation had multiple solutions, the broadband S21 data was used as a check on the calculated ε values. A schematic diagram of the measurement set-up and the sample filled test cell is shown in Figure 6.1. From this measurement the sample dielectric constant, dielectric loss and loss tangent were calculated.

![Figure 6.1: Equipment Set-up for Waveguide Transmission Technique](image)

The idea of calibration was to obtain accurate and repeatable values of reflection and transmitted coefficients with an air filled waveguide as standard and then compared these values with the sample filled ones. Calibration data were temperature dependent and hence a fresh calibration was required at every temperature. During measurements, fixed lengths of waveguide were filled with the FRTP and a range of waveguide sizes was required to provide an almost continuous spectrum of frequencies from 2.2 - 12.5 GHz. The transmission coefficient between the reference planes is given as (Sabburg et al, 1992):
6. Permittivity Measurement at Various Frequency and Elevated Temperatures

\[ S_{12} = \frac{4\Gamma_1\Gamma_0}{(\Gamma + \Gamma_0)^2 \exp^{i\beta_2L} - (\Gamma_1 - \Gamma_0)^2 \exp^{-i\beta_2L}} \]  

(6.21)

where \( \Gamma_0 = j\beta_0 \), the propagation coefficient in air filled waveguide

\[ \Gamma_1 = \alpha_1 + j\beta_1 \], the propagation coefficient in sample filled waveguide

L is the sample length in mm.

The reasons for using a range of waveguide sizes and different values of L for each waveguide size is discussed later. The relative permittivity of the FRTP (Sabburg et al, 1992) obtained as:

\[ \varepsilon = \varepsilon' - j\varepsilon'' = \varepsilon'(1 - j\tan \delta) \]  

(6.22)

For non-magnetic materials the elements of the complex permittivity were obtained from the propagation coefficient as follows (Sabburg et al, 1992):

\[ \varepsilon = [1 + (\beta_1^2 - \alpha_1^2)(a/\pi)^2](c/2af)^2 \]  

(6.23)

\[ \tan \delta = \alpha_1\beta_1c^2/(2\pi^2f^2\varepsilon') \]  

(6.24)

where ‘a’ is the broad dimension of the waveguide and ‘c’ is the velocity of electromagnetic waves. Since \( S_{12} \) was known from measurements, in principle Eqn 6.21 can be solved for \( \Gamma_1 \), from which the complex permittivity of the sample could be calculated. Ness (1983), Sabburg (1992) and Ball (1997) detailed an iterative technique for getting very accurate estimates of \( \varepsilon' \). Very accurate estimates of \( \varepsilon' \) were obtained by noting consecutive frequencies at which the transmission coefficient was a multiple of 90°. At these frequencies \( \beta_1L \approx n\pi \), where \( n \) was the number of half wavelengths in the sample, \( n = \frac{1}{2}, 1, 1\frac{1}{2} \) etc.

Substitution of these values into Eqn 6.23 gave Ness’s initial estimate (Ness, 1983):

\[ \varepsilon' = [1 + (an/L)^2](150/af)^2 \]  

(6.25)
where \(a\) and \(L\) are in \(\text{mm}\) and \(f\) in \(\text{GHz}\). As frequency increased, \(n\) had to increase by one half for each \(90^\circ\) change in transmission coefficient phase so that it was an even integer at \(0^\circ\) and an odd integer at \(180^\circ\) phase angle, and it would take on consistent values at consecutive frequencies. If two such frequencies were available within the measurement frequency range and assuming that \(\varepsilon'\) was not changing rapidly with frequency Eqn 6.25 yields:

\[
[1 + (\frac{an_1}{L})^2]f_2^2 = [1 + (\frac{an_2}{L})^2]f_1^2
\]

(6.26)

Imposing an appropriate relationship between \(n_1\) and \(n_2\), the result was a quadratic equation, which gave an estimate of either one of these numbers. Rounding the solutions to the nearest odd or even integer or integer plus one half as appropriate, a reliable value of \(n\) was obtained. With the aid of which \(\varepsilon'\) was estimated from Eqn 6.25; the initial estimate of \(\tan\delta\) was much less critical and Ness’s (1983) suggestion was adequate. The initial estimates of \(\varepsilon'\) and \(\tan\delta\) were used to calculate \(\Gamma_1\) and \(S_{21}\). Figures 6.2 and 6.3 show the behaviour of the reflection coefficient, \(S_{21}\), for LDPE/GF (33%) which is a low loss material. The calculated values of \(S_{21}\) were compared with the measured one and \(\varepsilon'\) was adjusted until the phase angles agreed. After that \(\tan\delta\) was adjusted until the magnitudes matched. The cycle was repeated until the error was minimum. The process was cumbersome and tedious and computer software was therefore developed to speed up the procedure. The software requested the input of values of transmission coefficients and of those of three markers (Ku et al, 1998) (one particular set is summarised in Table 6.1 for ease of reference) as well as the broad waveguide dimension and the sample length, \(L\) for the initial estimate of the \(\varepsilon'\) of the sample. An accurate initial estimate of \(\varepsilon'\) was important because it enabled the iteration process to converge to the required result from the infinite number of solutions. Sometimes, the software failed to estimate the initial value of \(\varepsilon'\) and the program did not converge. In this situation, a manual estimate was required and thus obtained. The software then calculated the values of \(\varepsilon'\) at a range of frequencies dedicated to that particular type of waveguide.
6. Permittivity Measurement at Various Frequency and Elevated Temperatures

Figure 6.2: Magnitude of $S_{21}$ against Frequency

Figure 6.3: Phase of $S_{21}$ against Frequency

Table 6.1: $S_{21}$ Marker Values for LDPE/GF (33%) at 18°C

<table>
<thead>
<tr>
<th>Marker</th>
<th>Frequency (GHz)</th>
<th>Phase(degrees)</th>
<th>Magnitude (dB)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>8.528</td>
<td>0.78</td>
<td>-0.326</td>
</tr>
<tr>
<td>2</td>
<td>10.148</td>
<td>-90</td>
<td>-1.96</td>
</tr>
<tr>
<td>3</td>
<td>11.82</td>
<td>-179.69</td>
<td>-0.228</td>
</tr>
</tbody>
</table>
6.6 Waveguide Sizes and Sample Lengths

In order to cover the frequency range from 2.2 to 12.5 GHz, four different sizes of waveguides were used, namely: WR90, WR159, WR229 and WR340. Taking waveguide WR340 as an example, the recommended range of frequencies (HP Coaxial and Waveguide Catalogue, 1993) for single (TM$_{10}$) mode was only from 2.2 to 3.3 GHz. To obtain satisfactory results from the waveguide transmission technique, the sample length had to be at least three-quarters of a guide wavelength of the highest measurement frequency. The wavelength in the sample depended on the sample’s relative permittivity or dielectric constant, $\varepsilon'$ as (Kraus, 1992):

$$\lambda_g = \frac{\lambda_o}{\sqrt{\varepsilon'}} \left[ 1 - \left( \frac{\lambda_o}{2a\sqrt{\varepsilon'}} \right)^2 \right] \quad (6.27)$$

where $\lambda_o$ is the free space wavelength and ‘a’ is the broad side waveguide dimension.

Assuming the largest possible frequency, ie frequency = 3.3 GHz, $\lambda_o = 30/3.3 = 9.091$ cm and $a = 3.4 \times 2.54$ cm = 8.636 cm.

The dielectric constant of the sample is expected to be in the range between 2 to 5.

Assuming $\varepsilon' = 2$ then Eqn 6.27 yields $\lambda_g = 6.926$ cm and the sample length $L_2$ with $\varepsilon' = 2$ would be 5.194 cm or 51.94 mm. Other relevant values are included in Table 6.2. A program was written to calculate the maximum sample length required for each waveguide size.

For each waveguide size, the network analyser was calibrated using a short circuit, an offset short and a matched load. The cutoff frequency was then easily calculated from the fact that the cut-off wavelength $\lambda_c$ was equal to 2 ‘a’ where ‘a’ was the larger cross-sectional dimension of the rectangular waveguide (Metaxas and Meredith, 1983).
6. Permittivity Measurement at Various Frequency and Elevated Temperatures

Table 6.2: Waveguide Type, Frequency Range and Sample Length

<table>
<thead>
<tr>
<th>WR 340</th>
<th>WR 229</th>
<th>WR159</th>
<th>WR 90</th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency range</td>
<td>Permittivity Sample Length</td>
<td>Permittivity Sample Length</td>
<td>Permittivity Sample Length</td>
</tr>
<tr>
<td>2.2 - 3.3 GHz</td>
<td>2</td>
<td>35</td>
<td>2</td>
</tr>
<tr>
<td>3.3 - 4.9 GHz</td>
<td>3</td>
<td>28</td>
<td>3</td>
</tr>
<tr>
<td>4.9 - 7.05 GHz</td>
<td>4</td>
<td>25</td>
<td>4</td>
</tr>
<tr>
<td>8.2 - 12.4 GHz</td>
<td>5</td>
<td>22</td>
<td>5</td>
</tr>
</tbody>
</table>

Therefore, \( f_c = \frac{3 \times 10^8}{\lambda_c} = \frac{3 \times 10^8}{2 \times 2.54 \times 3.4 \times 10^{-3}} = 1.7369 \text{ GHz} \). The absolute maximum frequency of operation was that at which the first higher order mode \( H_{20} \) began to propagate in an air filled waveguide. This was \( f_{\text{maximum}} = \frac{3 \times 10^8}{a} = \frac{3 \times 10^8}{2.54 \times 3.4 \times 10^{-3}} = 3.4378 \text{ GHz} \). It should be noticed that the values of \( f_c \) and \( f_{\text{maximum}} \) encompassed the recommended frequency range of 1.8 - 3.3 GHz for the waveguide of WR340.

In addition to keying in the cutoff (minimum) and maximum frequency to the programme of calibration for a particular waveguide size, eg WR340, it was also necessary to input the delay time for the offset short circuit. Delay time for offset short circuit = length of offset/velocity of light. Since the length of offset was 35.5 mm, the delay time = \( \frac{35.5 \times 10^{-3}}{3 \times 10^8} = 0.11833 \text{ ns} \). Similarly, the values of \( f_c \), \( f_{\text{maximum}} \) and delay time for the waveguides WR229, WR159 and WR90 were calculated.

6.7 Dielectric Probe Method

A dielectric probe for use in conjunction with a network analyser is located at the University of Queensland, Department of Electrical Engineering. It was used as an alternative method of measuring sample permittivity. The connection set-up for the HP 85070 dielectric probe kit (HP85070B Technical Data Sheets, 1993; HP85070A Technical Data Sheets, 1990) is schematically illustrated in Figure
6.4. The probe kit method employed an open-ended coaxial line to measure the complex permittivity of materials. The open-ended coaxial probe consisted of a truncated section of a coaxial line (Blackham et al, 1991) and an optional extension of a ground plane to improve the contact with the sample under test (NRC, 1994). The input port of the sensor was connected to the measurement equipment through a coaxial cable. The parameter to be measured was the reflection coefficient at the interface between sensor and sample. The use of the automatic analyser as the measuring instrument significantly simplified and enhanced the accuracy of the measurement procedure. The system allowed measurements of the complex permittivity for a wide range of semi-solid, pliable solid and liquid materials and performed all of the necessary network analyser control, calculation, and data presentation functions. Equipment was convenient to use and the operator needed only to press the probe against (or immerse it in) the MUT (material under test) to obtain a measurement. The software controlled the network analyser and measured the complex reflection coefficient of the MUT. It subsequently converted the reflection coefficient into the complex permittivity of the MUT and results were displayed on the screen. The probe had to be first calibrated (Blackham et al, 1991; NRC, 1994) against air, metallic shorting block and pure water before it could be used to measure the ε of any material. Care was taken to avoid the presence of bubbles beneath the probe while calibrating the kit against pure water. A re-calibration facility was available to speed up the measurements while measuring ε over a range of temperatures, varying from -40°C to 200°C. The frequency range was from 200 MHz to 20 GHz. The diameter of the sample under test had to be greater than 20 mm and its thickness and flatness had to be over \( \frac{20}{(\varepsilon')^{\frac{1}{2}}} \) mm and less than 25µm respectively. The accuracy for the ε’ and tan δ were ±5% and ±0.05 respectively. The maximum recommended ε’ was less than 100 and the minimum recommended tan δ was 0.05. For high temperature measurements, the probe was made from materials of very low thermal expansion coefficient.
6. Permittivity Measurement at Various Frequency and Elevated Temperatures

Figure 6.4: Schematic Diagram of HP 85070 Dielectric Probe Kit

Results from the measurements using the waveguide transmission technique and dielectric probe kit will be detailed in section 9.4.
7. A Resonant Cavity for Measurement of Complex Permittivity of Low Loss Thermoplastic Composites

7.1 Introduction

Methods such as the dielectric probe and waveguide transmission had been successfully used to measure the dielectric constant ($\varepsilon'$) of low loss thermoplastic composites but they were found to give unreliable results when used to measure the dielectric loss, $\varepsilon''$ (Ku et al, 1998; 1999a; 1999b; 1999c; 2000b). This chapter describes a convenient and inexpensive laboratory method designed to obtain $\varepsilon'$, $\varepsilon''$ and hence loss tangent ($\tan \delta$) for low loss thermoplastic composite materials like glass fibre reinforced (33%) low-density polyethylene [LDPE/GF (33%)]. The method is called the resonant cavity method and schematic diagrams of the equipment and the cavity are depicted in Figures 7.1 and 7.2 respectively. It utilised a microwave network analyser to measure the two dielectric properties using the reflection method. The basis for this method was to measure the shift in quality (Q) factor and resonant frequency when the composite, eg LDPE/GF (33%) was inserted to the initially empty cavity. The cavity was made from standard waveguide, eg WR340 with short circuit plates screwed to flanges at each end (not shown in Figure 7.2). This chapter includes measurements of the dielectric loss of low loss thermoplastic composites, eg LDPE/GF (33%) over a range of frequencies and at elevated temperatures using the resonant cavity method. In calculating the dielectric loss of the material, the average of several different Q factor estimates should be used. Cavity perturbation measurements are highly accurate and found to be particularly advantageous in the determination of low loss tangent. The Q factor (Kraus, 1992) is usually defined as:

$$Q = 2\pi \times \frac{\text{energy stored}}{\text{energy lost per cycle}}$$  \hfill (7.1)

Because the Q factor was described in terms of energy storage inside the cavity, with the composite inside the cavity, the energy storage was perturbed. Altman
Figure 7.1. : Set-up for Cavity Method

Figure 7.2. : Resonant Cavity with End Plates Removed

(1964) has described in detail the application of the perturbation method to cavity resonators. Consider an initially unperturbed, air-filled cavity, which has a
resonant frequency, $\omega_o$, dielectric constant, $\varepsilon_o$, and volume, $V$. The fields inside the cavity can be represented by the electric field, $E_o$, and magnetic field, $H_o$. In the presence of a small sample of material such as LDPE/GF (33%) with a volume of $\Delta V$ and a dielectric constant, $\varepsilon'$, the resonant frequency in the cavity shifted to $\omega_s$ (Altman, 1964; Chan, 1998). The perturbation of the cavity resulted in the shift in the resonant frequency and also a change in the $Q$ factor. The basic assumptions behind this method is that the sample has to be very small compared to the cavity itself, so that a frequency shift that is small compared with the resonant frequency of the empty cavity is produced by the insertion of the sample (NRC, 1994). The following equations have been derived using the perturbation method (Altman, 1964; Meredith, 1998):

$$\left(\frac{\omega_s - \omega_o}{\omega_o}\right) = -\frac{\varepsilon_o (\varepsilon' - 1) \left( \int_{\Delta V} E_o^* E_s \, dv \right)}{4U_{total}} \quad (7.2)$$

$$\frac{1}{Q_s} - \frac{1}{Q_o} = \frac{\varepsilon_o \varepsilon'' \left( \int_{\Delta V} E_o^* E_s \, dv \right)}{2U_{total}} \quad (7.3)$$

where

- $\omega_s$: resonant frequency with sample;
- $\omega_o$: resonant frequency with empty cavity;
- $Q_s$: $Q$ factor for cavity with sample;
- $Q_o$: $Q$ factor for empty cavity;
- $\varepsilon_o$: permittivity of free space;
- $\varepsilon'$: real part of complex relative permittivity for sample;
- $\varepsilon''$: imaginary part of complex relative permittivity for sample;
- $E_o$: electric field in an empty cavity;
- $E_s$: electric field in a cavity with sample;
- $U_{total}$: total average stored energy in the cavity.
- $\Delta V$: sample volume

Suppose that the sample took the form of a thin slab of thickness, $t$, placed upright in the centre of the cavity and extending across the entire waveguide.
cross-section, as shown in Figure 7.2. In the empty cavity the only component of the electric field would be in the vertical direction. The introduction of a thin sample would cause very little alteration, since the tangential electric field is continuous across a material interface. Using this assumption, Eqn 7.2 and Eqn 7.3 were evaluated based on the configuration shown. If the sample was located at an electric field maximum, then after manipulation Eqn 7.2 becomes (Ku et al, 1999b; 1999c; 2000b):

\[
\left( \frac{\omega_s - \omega_0}{\omega_0} \right) \approx \frac{-(\varepsilon' - 1)t}{L} \tag{7.4}
\]

and Eqn 7.3 becomes:

\[
\frac{1}{Q_s} - \frac{1}{Q_0} \approx \varepsilon'' \left[ \frac{2t}{L} \right] \tag{7.5}
\]

where \( t \): thickness of the sample material in cm;

\( L \): length of the cavity in cm.

With the aid of Eqn 7.4 and Eqn 7.5, the complex relative permittivity of the sample and subsequently the loss tangent were evaluated.

### 7.2 Cavity Length

In order to make the cavity containing a sample resonant at a particular frequency, the empty cavity should be made resonant at a higher frequency, by an amount that can be estimated in advance. Taking the operating frequency of 2.45 GHz as an example, the empty cavity was made resonant at approximately 2.65 GHz when the waveguide selected was WR340. At the fundamental resonance, the cavity length was equal to half of the waveguide wavelength. Using Glazier and Lamont (1958)

\[
\left( \frac{1}{2L} \right)^2 + \left( \frac{1}{2a} \right)^2 = \left( \frac{1}{\lambda_0} \right)^2 \tag{7.6}
\]

where \( L \) is the length of the cavity in cm;

\( a \) is the broader side of the waveguide in cm;
A Resonant Cavity for Low Loss Thermoplastic Composites

$\lambda_0$ is the wavelength of electromagnetic field in free space in cm.

$\lambda_0 = \text{velocity of light} / f_0 = \frac{30 \times 10^6}{2.65 \times 10^9} = 11.32 \text{ cm.}$

$a = 2.54 \times 3.4 \text{ cm} = 8.836 \text{ cm}$ and substituting both values of $\lambda_0$ and $a$ into Eqn 7.6 gives $L = 7.5 \text{ cm.}$

Using Eqn 7.4, and assuming the values of $t = 0.4$, a typical dielectric constant of $\varepsilon' = 2.6$ and $f_0 = 2.65 \text{ GHz.}$

Eqn 7.4 becomes $f_s - f_0 \approx (2.6 - 1) \times \frac{0.4}{7.5}$ and $f_s$ becomes $2.44 \text{ GHz}$ which was not far from the operating frequency and thus the initial estimate of $f_0 = 2.65$ was close enough. Similarly, other targeted frequencies are included in Table 7.1.

<table>
<thead>
<tr>
<th>Targeted Frequency ($f$)</th>
<th>Initial Frequency ($f_0$)</th>
<th>Waveguide Type</th>
<th>Cavity Length (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>With Sample (GHz)</td>
<td>Without Sample (GHz)</td>
<td>Type</td>
<td></td>
</tr>
<tr>
<td>2.45</td>
<td>2.65</td>
<td>WR340</td>
<td>7.5</td>
</tr>
<tr>
<td>3.5</td>
<td>3.84</td>
<td>WR229</td>
<td>5.13</td>
</tr>
<tr>
<td>6</td>
<td>6.4</td>
<td>WR159</td>
<td>8.6</td>
</tr>
<tr>
<td>9</td>
<td>9.85</td>
<td>WR90</td>
<td>6.1</td>
</tr>
</tbody>
</table>

### 7.3 Calibration of Network Analyser

The network analyser had to be 1-port calibrated with open, short and broadband load standards in order to measure the reflection coefficient. One of the display modes available on the analyser is a Smith Chart, which is a polar plot of reflection coefficient. After making the correct calibration and before removing the broadband load, the analyser should display a point ‘P’ at the centre of the Smith Chart as depicted in Figure 7.3. The probe-coupled cavity was then
connected to the network analyser. Consider a WR229 containing a sample of LDPE/GF (33%) cavity, both ends of closed by two short-circuit plates. The plates were joined to the cavity with screws tightened to the same amount of torque force. The sample was made to stand upright in the middle of the cavity by placing it between two slabs of polystyrene foam that had a low value of permittivity, approximately equal to unity. When the cavity was resonant with the sample [LDPE/GF (33%)] at 24 °C, the select and view in the display menu of the network analyser showed that the loop of the reflection coefficient ‘C’ was repeatedly traced out, as depicted in Figure 7.4. The cavity was undercoupled because the loop did not enclose the centre of the Smith Chart. The reflection coefficient magnitude and phase displays provided by equipment are separately shown in Figures 7.5 and 7.6 respectively. Figures 7.7 and 7.8 are the simplified versions of the above two views respectively (Ku et al, 1999b; 2000c). After
this, the resonant frequency of the cavity without the sample was then measured. The

![Smith Chart for WR229 Cavity with LDPE/GF (33%) Sample in the Vicinity of Resonance](image)

**Figure 7.4.** Smith Chart for WR229 Cavity with LDPE/GF (33%) Sample in the Vicinity of Resonance

![Reflection Coefficient Magnitude for WR229 Cavity Containing](image)

**Figure 7.5:** Reflection Coefficient Magnitude for WR229 Cavity Containing
7. A Resonant Cavity for Low Loss Thermoplastic Composites

LDPE/GF (33%) Sample

Figure 7.6: Reflection Coefficient Phase for WR229 Cavity with LDPE/GF (33%) Sample

Figure 7.7: Simplified Diagram of Reflection Coefficient Magnitude
two slabs of polystyrene foam were also in place when the cavity was resonant with no sample. This time the reflection coefficient loop enclosed the centre of the Smith chart meaning that the cavity was overcoupled. The Q factors of the cavity with and without sample were then calculated from one of the four methods available which are described in the next section. The imaginary part of the permittivity of the sample [LDPE/GF (33%)] was found by the shift of the ‘Q’ factor.

### 7.4 Formulae for Calculating Q Factor

By modelling a cavity as a tuned circuit at the end of a transmission line, various relationships can be deduced between Q factor and properties of the reflection coefficient (Kwok and Liang, 1999; Ku et al, 1999b; 2000b). The four methods considered were phase turning points, magnitude only, ±90° method and phase slope method (Ku et al, 1999b). The phase turning point method was applicable only to the undercoupled case and the ± 90° method to the overcoupled
situations only; while the other two methods could be used for both undercoupled and overcoupled cases but the equations used were not totally the same.

7.4.1 Phase Turning Points

Consider the phase turning point method first; because of the inability of the network analyser to display something less than -180°, or more than 180°, the phase curve tended to return to 180° if it crossed the -180° barrier and vice-versa. In Figure 7.8 points P1 and P2 differ by 360°. This is called the ‘wrap around’ effect. The phase trace in Figure 7.8 had therefore to be shifted to the one shown in Figure 7.9 by removing the 360° discontinuity. The phase trace was now from O1, P2 and O2; the resonant frequency, f₀, could be found from Figure 7.5 and f₁ and f₂ were the frequencies of the two turning points employed in the calculation of Q factor. When the WR229 cavity was filled with the sample, the resonant frequency, f₀ at 24°C was found to be 3.40135 GHz and the frequencies and phases of the minimum and maximum points were respectively 3.3992 (f₁) GHz and -198.40° (θ₁), and 3.4035 (f₂) GHz and -108.45° (θ₂). If the phase difference between the two extreme points was 2φm, then

\[
\frac{f_b - f_a}{f_o} = \left(\frac{1}{Q}\right) \cos \phi_m
\]

(7.7)

where f₁, f₂ and f₀ are the frequencies of the extreme points and the resonant point respectively. Substituting the values of f₁, f₂, f₀ and cosφm into Eqn 7.7 gave

\[
Q_s = 0.7074 \times \frac{3.40135}{3.4035 - 3.3992} = 0.7074 \times \frac{3.40135}{0.0043} = 559.57,
\]

where Q_s is the Q factor with the sample (Chan, 1998; Ku et al, 1999b; 2000c).
7.4.2 Magnitude Only Method

The process was repeated with no sample in the cavity and at the same temperature and in this case the cavity was overcoupled. The magnitude only method was therefore used to calculate the Q factor with no sample. Referring to Figure 7.7, the magnitudes and frequencies of P3 and P4 found were -1.92889 dB ($\rho_a = 0.80086$) and 3.8272 GHz ($f_a$), and -1.91638 dB ($\rho_b = 0.8020$) and 3.8362 GHz ($f_b$) respectively. The average reflection coefficient at these points was therefore $|\rho| = 0.801435$. The magnitude and frequency of the dip, D, were -3.900 dB and 3.8319 GHz ($f_o$) respectively and the minimum reflection coefficient, $|\rho_{\min}| = 10^{-3.9} = 0.638263$.

Using Ku et al, (1999b; 2000c)

$$Q_o = \frac{f_o}{f_b - f_a} \frac{2}{(1 - |\rho_{\min}|)} \sqrt{\frac{|\rho_a|^2 - |\rho_{\min}|^2}{1 - |\rho|^2}}$$

(7.8)

where $Q_o$ is the Q factor with no sample.
The Q factor without sample was then found to be:

\[ Q_0 = \frac{38319}{0.009} \times \frac{2}{1 - (0.638263)^2} \times \sqrt{(0.801435)^2 - (0.638263)^2} = 1907.68 \]

Again, using Eqn 7.5 and substituting the values of the variables into this equation,

\[ 1 \frac{560}{513} - 1 \frac{1908}{5.13} = 2 \varepsilon'' \times 0.4 \]. This gives \( \varepsilon'' = 0.0809 \).

Modifying Eqn 7.4 so that it becomes

\[ \frac{f_s - f_o}{f_o} \cong -\frac{(\varepsilon' - 1)t}{L} \]  

(7.9)

and substituting the values of \( f_s = 3.40135 \), the shifted frequency and \( f_o = 3.8319 \), original frequency into Eqn 7.9 and it was found that \( \varepsilon' = 2.44 \).

If the cavity had been undercoupled a slightly different equation should have been used (Garner et al, 1990) and this is

\[ Q_o = \frac{f_o}{f_s - f_a} = \frac{2}{(1 + |\rho_{\text{min}}|)} \sqrt{|\rho|^2 - |\rho_{\text{min}}|^2} \]  

(7.10)

### 7.4.3 Phase Slope Method

The two remaining methods used were the phase slope and the ±90°. Consider the phase slope method first and the data for the WR229 with a sample of LDPE/GF (33%) at 24°C. The point O in Figure 7.10 represents the resonant frequency; its frequency and phase angle were 3.4013 GHz and -153.219° respectively. In order to calculate the rate of change of phase angle with respect to frequency, it was necessary to locate some points near the resonant frequency.
Points P and Q, which were adjacent to the resonant frequency point, O, were identified and shown in Figure 7.10. The coordinates of points P and Q were 3.4012 GHz and -158.898 °, and 3.4014 GHz and -147.859 ° respectively. Therefore, \( \Delta \phi = 11.309^\circ \) and \( \Delta f = 0.0002 \text{ GHz} \) and \( \frac{d\phi}{df} = \frac{11.039}{0.0002} \text{ degrees/GHz} = 963.334 \text{ rad/GHz} \). It should be noted that the slope was positive and the cavity was undercoupled. The magnitude at the resonant frequency was found to be -15.4224 dB and therefore \( |\rho_{\text{min}}| = 0.169387 \).

Using Ku et al (1999b; 2000c)

\[
Q_s = f_o \left( \frac{\frac{d\phi}{df}}{f_o} \right) \frac{|\rho_{\text{min}}|}{1 - |\rho_{\text{min}}|^2} \tag{7.11}
\]

\[
Q_s = 3.4013 \times 963.334 \times \frac{0.169387}{1 - (0.169387)^2} = 571.406,
\]

which was very close to 559.57 obtained by the phase turning point method.

![Figure 7.10: Phase Angle against Frequency](image)
NOTE

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8. Variable Frequency Microwave Facilities (VFMF)

8.1 Introduction

In conventional microwave processing, microwave energy is launched at a fixed frequency of either 915 MHz, 2.45 GHz, 5.8 GHz or 24.125 GHz (Thuery, 1992) into a waveguide or cavity and these fixed frequencies bring with them inherent non-uniformity heating problems like hot spots and thermal runaway, if the continuous microwave mode is applied. A new technique for microwave processing, known as the variable frequency microwave (VFM) technique has been developed to solve the problems brought about by the fixed frequency continuous microwave processing (Lambda Technologies, undated). The technique is geared towards advanced materials processing and chemical synthesis. It offers rapid, uniform and selective heating over a large volume at a high energy coupling efficiency. This is accomplished using a preselected bandwidth sweeping around a central frequency, by employing frequency agile sources such as travelling wave tubes as the microwave power amplifier. Selective heating of complex samples such as thermoplastic matrix composites and industrial scale-up opportunities are now viable (Wei et al, 1998). Successful applications include curing advanced polymeric encapsulants, rapid processing of flip-chip underfills, materials characterisation, curing profiles for various adhesives, structural bonding of glass to plastic housing (Fathi et al, undated; Anderson, undated; Clemons et al, undated; Wei, 1998). The manufacturer had so far produced three series of variable frequency microwave facilities namely, VW1500, Microcure 2100 and Microcure 5100 and Microcure 2100 which is further classified into three models: 250, 700 and 2000 (Lambda Technologies Inc., 1998). The maximum power level for the model 2000 is 1.8 kW. In the Industrial Research Institute, Swinburne, Swinburne University of Technology, there are two sets of VFM facilities; they are VW1500 and Microcure 2100 model 250. The VW1500 has a bandwidth range of 6 - 18 GHz at a nominal power of 125 W; its cavity dimensions are 250 mm x 250 mm x 300 mm. The Microcure 2100 model 250 has cavity size of 300 mm x 275 mm x 375
mm; the frequency band for this machine was 2 - 7 GHz operating at a nominal power of 250W. The cavity sizes for the two VFM facilities are shown in Figures 8.1 and 8.2. Most of the experimental results in this chapter were obtained utilising the Microcure 2100 model 250.

### 8.2 Interaction of VFM Fields with Materials

When the microwave energy at a fixed frequency, eg 2.45 GHz, is launched into a waveguide, eg WR340, as depicted in Figure 8.3(a), containing a piece of material, some areas of the material would have higher electric field strength than the others. This situation would even be more serious if the microwave energy was launched into a multimode cavity because many resonant modes are established. Figure 8.3(b) shows qualitatively the fixed electric field pattern at the joint of the test pieces during fixed frequency heating. Areas with higher electric filed strength are heated more, creating hot spots, which can eventually lead to thermal runaway. However, these hot spots can be reduced by using a mode stirrer or by rotating the sample. With variable frequency microwave heating (Wei, 1998), as shown in Figure 8.4(a), more than one thousand frequencies are launched into the cavity sequentially. Each incident frequency sets up its own electric field pattern and therefore results in hot spots generated at different locations and at different time intervals, as shown in Figure 8.4 (b). Simply, different areas are heated by different frequencies at different times. However, when a sufficient broad bandwidth is used, every element of the test piece experiences hot spots at one or more frequencies during sweeping. Therefore, time-averaged uniform heating can 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 at any short time interval to optimise the coupling efficiency.
Figure 8.1: Cavity Size of VW 1500

Figure 8.2: Cavity Size of Microcure 2100 Model 250
8. Variable Frequency Microwave Facilities

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

b) Electric Field Pattern for (a)

Figure 8.3: Fixed Frequency Microwave Heating – Nonuniform Heating
8. Variable Frequency Microwave Facilities

a) Variable Frequency Microwave Energy Launched into Multi Mode Cavity

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

Figure 8.4 Variable Frequency Microwave Heating – Time-Averaged Uniform Heating
8.3 Hardware of Microcure 2100

The Microcure 2100 consists of several separate subsystems. The subsystems (Lambda Technologies Inc, 1998) comprise curing cavity, oven control system, signal generator and high power amplifier (HPA) system, transmission system, and fibre optic base temperature monitoring system. A block diagram of the system is shown in Figure 8.5.

![A Block Diagram of the System of Microcure 2100](image)

Figure 8.5: A Block Diagram of the System of Microcure 2100

8.3.1 Curing Cavity

The cavity used in the experimental work was the metal enclosure where microwave processing or curing process took place. The cavity featured a manual hinged door with microwave seals around the perimeter of the facing door. The door seals provided good electrical contact and prevented microwave leakage from the cavity enclosure. The cavity contained four pass through ports for fibre optic temperature probes, and a choke and view port for the infrared temperature sensor when this option was selected. The samples being processed were placed directly on the bottom of the cavity. They were supported off the
cavity floor at a minimum distance of 20 mm by a suitable microwave transparent fixture such as a Teflon block (Lambda Technologies Inc., 1998).

8.3.2 Oven Control System

The oven control system was software driven. It supervised and controlled the Microcure operations. An industrial personal computer served as the central data collection and control system. It interfaced with, and supervised, all of the subsystems of the facilities. It was also responsible for operating the graphical user interface as well as maintaining the input and output functions of the VFM system.

8.3.3 Signal Generator and High Power Amplifier

Unlike most home microwave ovens or the microwave facilities configuration in the University of Southern Queensland, the microwave source of the Microcure 2100, was not a magnetron but a signal generation system comprising of a Yig oscillator, a voltage controlled attenuator, PC data acquisition and the VFM interface board (Lambda Technologies Inc., 1998). It generated all of the system’s low-level microwave energy and controlled all of the system’s high-level energy. The high power amplifier (HPA) amplified the low-level microwave energy generated from the signal generator section to levels high enough to provide adequate molecular excitation in the curing cavity. The HPA consisted of a travelling wave tube, a high voltage power supply, and a solid state amplifier that served as an intermediate power amplifier, and control logic. It communicated with the PC via an RS-232-C serial communication link.

8.3.4 VFMF Safety Printed Circuit Board (PCB)

This stand-alone device sensed the high-level microwave energy in the transmission system and at the same time monitored the cavity door safety interlocks and any other high level radio frequency (RF) safety interlocks thus
determining that they were in a safe position. If either the system detected a dangerous level of energy in the transmission system while an interlock was tripped or if the cavity was overheated, the VFMF safety PCB severed the safety interlock line in the HPA. This stopped the high-level microwave energy from being produced until a safe condition was selected (Lambda Technologies Inc., 1998).

8.3.5 Transmission System

The transmission system was for routing the high power microwave energy from the HPA to the cavity. The system consisted of a high power isolator, a dual directional coupler (combined with crystal detectors to provide power sampling for the system), waveguide, and an iris-syle launcher.

8.3.6 Temperature Monitoring System

Two types of temperature monitoring systems were available for the Microcure 2100. As standard, a single channel fibre optic measurement system was provided. This included a fibre optic probe within the cavity that had to be placed in contact with the surface at the location, which was monitored. An additional 3 (4 total) fibre optic channels were also installed. A second temperature device consisted of an infrared non-contact temperature measurement unit that provided specific surface temperature reading without making physical contacts with the parts being processed. The optical beam was targeted on parts being processed thus providing a temperature reading to the control system.

8.4 Software System

8.4.1 Introduction
The Micro 2100 system software automatically controlled all the functions of the Microcure 2100. However, most of the functions were controlled by a combination of manual and software controls. Plate 1 shows the main screen (Lambda Technologies Inc., 1998) which was grouped into three basic categories: status displays, quantitative displays, and controls. Reading from the top second line of the main screen, five sets of pull-down menus were found. The system used standard MS Windows, file-naming protocols. The utilities and event menus were the two most important ones and are described in more detail below.

Plate 1: Menu-driven Graphical Interface (Lambda Technologies, 1999)

8.4.2 Utilities Menu

Two important sub-menus under the utilities menu included characterisation and graphics. The characterisation menu was used to measure the characteristics of the cavity when the sample was loaded. The procedures were a sequence of operations whereby the machine shows graphically how the cavity, with the material loaded, performed over the operating frequency range. The input power
selected depended on the estimated loss tangent of the material. The higher the estimated loss, the lower was the power level selected. During characterisation of the loaded cavity, the machine operator read the variation in temperature and incident power as well as the reflected power via the monitor. The incident and reflected power levels versus frequency spectrum, as well as the percentage of reflectance against frequencies, were saved and viewed later but this was not the case for the variation in temperature. From this data, the operator made an educated estimation of bandwidths most suitable for VFM processing.

8.5 Characterisation of Thermoplastic Matrix Composite (TPC) Materials

The input power was selected on the basis of the estimated loss tangent of the material. The higher the estimated loss, the lower the power level selected. In the characterisation of LDPE/CF (33%) from a frequency range of 6.5 GHz to 18 GHz, the power selected was 50 W and the amount of energy reflected varied from 0 W to 30 W, ie 0% to 60%. In the characterisation of LDPE/GF (33%), using the same frequency range, the input power was 125 W and the reflected energy ranged from 0W to 100W, ie 0% to 80%. This indicated that LDPE/CF (33%) is lossier than LDPE/GF (33%). During characterisation of the loaded cavity, temperature variations were obtained via the monitor as well as incident power and reflected power levels from the cavity containing the sample. The incident and reflected power levels versus frequencies together with the percentage of reflectance against frequencies were monitored and recorded. From this data, the bandwidths most suitable for microwave processing were chosen.

8.5.1 Characterisation of TPC Materials from 6.5 GHz to 18 GHz.

The total operation bandwidth for VW1500 was from 6.5 GHz to 18 GHz. For the sake of keeping the temperature low, the characterisation bandwidth was broken into four equal sections. For each quarter of the operational bandwidth of the machine, three tests were performed. This was to ensure that the tests were repeatable irrespective of the position of the specimen in the cavity. From the
data, it was found that the effect of position was not large and the spectra for each test were very similar.
The output power for characterisation for PS/GF (33%) and LDPE/GF (33%) was 125 W and the maximum temperature reached was 32°C and 33°C respectively. On the other hand, the output power for the remaining three materials were 50 W and the maximum temperature reached was 65°C. This machine was not power levelled; the output power was frequency dependent. The output power for nylon 66/GF (33%), PE/CF (33%) and PS/CF (33%) in the frequency range of 9.375 GHz to 12.25 GHz was around 50 W. However, the power output was above 50 W at the lower frequency range and below 50 W at the higher frequency range. For the relatively lower loss materials, though the output power was set at 125 W, the actual power applied on the load was less than 100 W. While analysing the characterisation results, the four quarters of the data were grouped back into one.

Considering that the reflectance is the ratio of the reflected power to the incident power, the lowest percentage reflectance for LDPE/GF (33%) was found to be between 8.5 GHz to 9 GHz and 10.7 GHz to 12 GHz, as depicted in Figure 8.6. The percentage reflectance ranged from 0 to 15%. The best frequency ranges to process the material were therefore from 8.5 GHz to 9 GHz and from 10.7 GHz to 12 GHz out of the total frequency range from 6.5 GHz to 18 GHz (Ku et al, 2000d).

Figure 8.6: Percentage of Reflectance against Frequency for LDPE/GF (33%)
The procedures were repeated with PS/CF (33%), PS/GF (33%), LDPE/GF (33%) and Nylon 66/GF (33%) and the best frequency band to process these five materials in the frequency range of 6.5 GHz to 18 GHz was from 8 – 12 GHz (Ku et al, 2000d).

8.5.2 Characterisation of Materials from 2 GHz to 8 GHz.

This time the Microcure 2100 was used with the frequency range of the machine being from 2 – 8 GHz. LDPE/CF (33%) was expected to have a relatively high loss tangent because of the carbon fibre contained in the matrix and thus, a power level of 50 W was chosen. This was to ensure that the interaction of microwave energy and the sample was not too vigorous and that the facility could provide a complete sweep of frequency from 2 GHz to 8 GHz in a certain short period of time, eg 0.1 second, without making the temperature in the cavity dangerously high. The temperature adjacent to the sample was monitored during the cavity characterisation process and the machine was switched off once the temperature was over 105°C, which was not too far from the melting point of the material. Figure 8.7 illustrates the percentage of reflectance against frequencies. It was found that the percentage reflectance was lowest in the frequency range of 6.5 GHz to 8 GHz (Ku et al, 2000d). The percentage reflectance ranged from 0 to 35%. LDPE/CF (33%) was therefore best processed in this frequency range because it absorbed a greater proportion of the incident power.

When Nylon 66/GF (33%) was characterised, the input power was 100 W. In the frequency range of 2 GHz to 8 GHz, the best frequency range to process it was found to be from 6.5 GHz to 8 GHz as shown in Figure 8.8 (Ku et al, 2000d). The percentage reflectance ranged from 0 to 40%.
Figure 8.7: Percentage of Reflectance versus Frequency for LDPE/CF (33%)

Figure 8.8: Percentage of Reflectance against Frequency for Nylon 66/GF (33%)

Figure 8.9 shows that the best frequency ranges to process liquid rapid araldite (LRA) is again from 6.5 GHz to 8 GHz (Ku et al, 2000d). The percentage of reflectance ranged from 0 to 35. Similarly, the best frequency range to process PS/CF (33%), LDPE/GF (33%) and PS/GF (33%) were also found to be from 6.5 GHz to 8 GHz (Ku et al, 2000d). The percentage of reflectance of the three materials ranged from 0 to 40.

The best frequency range to process the five thermoplastic matrix composite materials in the frequency range of 2 GHz to 18 GHz is summarised and concluded in section 9.6.
8.6 Programming Menu

The two important sub-menus under the above menu were define event menu and monitor event. The user could write programs to control the machine by selecting one or more of the 15 standard instructions: fixed frequency, variable frequency sweep, max temperature, max reflected power, power output, RF on, RF off, wait for temperature, set temperature, set time, start module, stop module, number of parts-begin, number of parts-end, and loop.

8.6.1 LDPE/GF (33%) Program (2 – 8 GHz)

The first experiment was conducted using LDPE/GF (33%). From section 9.6, it would be found that the best frequency to process the above material using Microcure 2100 (Ku et al, 2000d). Since the material was processed with variable frequency sweep, it was necessary to identify the centre frequency for the sweep, which was found to be \( \frac{6.5\text{GHz} + 8\text{GHz}}{2} = 7.25 \text{GHz} \). Since the bandwidth of the sweep should be greater than 1.0 GHz (Lambda Technologies Inc., 1998; Bows, J.R., 1999), the selected bandwidth was 1.1 GHz. The actual start and stop frequencies were centre frequency \( \pm \frac{\text{bandwidth}}{2} \), ie the sweep was
from 6.7 GHz to 7.8 GHz. Because the sweep time could range from 0.1 second to 100 seconds (Lambda Technologies Inc., 1998), the chosen sweep time was 0.1 second. Since the material loss tangent was relatively low as measured, and discussed in chapter 7.4 (Ku et al, 1999b: 2000c; 2000e), a power level of 200 W was selected. The processing temperature was set at 95°C with a deadband of 1°C and the total processing time was set at 6 minutes. The maximum permitted temperature was set at 100°C, above, which the machine was switched off automatically. This temperature was very near to the melting point of the matrix of the composite. The reason for setting this maximum temperature was to avoid excessive temperature rise, which forms hot spots and gives rise to thermal runaway. The lapped area for the joint was 10 mm x 20 mm. The bond surfaces were roughened with coarse, grade 80 emery paper. The roughened surfaces were then cleaned and degreased by immersing them in methanol. After drying, two part 5-minute araldite of around 1.5 to 2 cubic centimetres was applied to the two roughened surfaces to increase the mechanical keying or interlocking as mentioned in section 3.5.1 (Ku et al, 2000e). The two test pieces were then brought together and the total pressure was about 4 N. The program parameters were as follows:

Variable Frequency = 7.25 GHz, Bandwidth = 1.1 GHz, Sweep time = 0.1 secs
Power Output = 200 Watts, Active
Set Temp = 95 Degs C, Variation = 1 Degs C, Processing time = 6 minutes
Max Temperature = 100 Degs C, Active

After processing, the graphics sub-menu was selected and viewed. The outcome was as shown in Figure 8.10. It was found that the temperature rose steadily with no sign of hot spots or thermal runaway. The maximum temperature reached was 95°C at time equal to 360 seconds (Ku et al, 2000e). For obtaining tensile shear tests results, several sets of test pieces were joined using different durations and details are given in section 9.7.3.
8. Variable Frequency Microwave Facilities

8.6.2 PS/GF (33%) Program (2 – 8 GHz)

For this material, the fixed frequency of 2.45 GHz, but rounded to 2.5 GHz by the machine, was launched into the cavity to enable a comparison to be made between LDPE/GF (33%) and PS/GF (33%). The same primer was also used and the power level of 200 W was also chosen while the processing temperature was set at 95°C and the processing time was set at 540 seconds.

The parameters were as follows:

Fixed Frequency = 2.5 GHz
Power Output = 200 W, Active
Set Temp = 95 Degs C, Variation = 1 Degs C, Processing time = 9 minutes
Max Temperature = 100 Degs C, Active

As a temperature of 110°C was attained at time equalled to 290 seconds, the machine was automatically stopped. The graphics sub-menu was then selected and viewed. The outcome is shown in Figure 8.11. It was found that temperature...
rose slowly and steadily in the first one hundred seconds. A steeper rise in temperature was observed after 100 seconds (Ku et al, 2000e).

Above this temperature, and at time duration of 210 seconds, hot spot(s) developed into thermal runaway and the machine was shut down when the temperature of 101°C was reached at a time equal 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 piece were processed at different time intervals. Figure 8.12 shows the temperature versus time diagram for PS/GF (33%) joined at 150W and for a period of 570 second. The maximum temperature recorded in running this program was 95°C (Ku et al, 2000e). Tensile shear test results can be found in section 9.7.1.

8.6.3. Other Programs (2 – 8 GHz)

Three sets of other experiments were performed to process the remaining three materials, Nylon 66/GF (33%), LDPE/CF (33%) and PS/CF (33%).
Again, for Nylon 66/GF (33%) the same filler material, liquid rapid Araldite, was used. With this material, the centre frequency was 7.25 GHz but the bandwidth of sweep was increased to 1.5 GHz to improve processing performance (Lambda Technologies, 1998). Since the Vicat temperature (Higgins, 1991) of Nylon 66 is higher than those of polystyrene and low density polyethylene, the maximum temperature was set at 100°C to prevent overheating of the araldite. Details of tensile shear test results and other findings are discussed in chapter 9.7.2.

With PS/CF (33%), no Araldite was used as a primer because the reinforcing carbon fibre would bring about larger loss to the material. Details of tensile shear test results are investigated in chapter 9.7.4.

The last material to be considered was LDPE/CF (33%). In this case, the fixed frequency of 2.45 GHz was used, and because of the anticipated arcing of the carbon fibre reinforcement, the duration of exposure to microwave energy were significantly reduced. Tensile shear test results are detailed in section 9.7.5.
8. Variable Frequency Microwave Facilities

8.7 Concluding Remarks

After analysing the diagrams and data collected in the experiments, it was found that the optimum frequency band for microwave processing of these five materials was in the range of 8 – 12 GHz. This data enables one to join the above five materials using microwave energy at their most favourable conditions.

Looking back at the two materials, LDPE/GF (33%) and PS/GF (33%), it was found that the dielectric properties of both of them were more or less the same (von Hipple, 1954; Metaxas and Meredith, 1983; Ku et al, 1997a; 1997b; 1998; 1999a; 1999b). It was therefore argued that if the former material was processed and joined under the same conditions as the latter, its results, including the bond strength and bond quality would be more or less the same as those of the latter and vice-versa. If LDPE/GF (33%) was joined under the same conditions as that of PS/GF (33%), hot spots or even thermal runaway were to be expected. Under the given circumstances, including a power output of 200 W and fixed frequency of 2.5 GHZ, processing resulted in thermal runaway and the machine was programmed to stop automatically (see Figure 8.11). In order to avoid thermal runaway, the power level had to be reduced to 150 W and this lengthened the joining time by 40% (see section 9.7) as compared to that of LDPE/GF (33%). The disparity between the two was mainly due to the fact that variable frequency microwave processing was not employed in the processing of PS/GF (33%). From the information discussed in this chapter, it appeared that variable frequency microwave processing was more versatile and advantageous than the fixed frequency bonding. Bond strength, tensile shear strength of each material, and the quality of the bonds are discussed in detail in section 9.7.
NOTE

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9. Results and Discussion

9.1 Introduction

In addition to the introductory section, this chapter has five main sections of results, sections 9.3 to 9.7, which are followed by a section of concluding discussion, section 9.8. Section 9.2 summarises all the findings of the research project. Section 9.3 discusses the results of the surface analysis on the joined surfaces of the thermoplastic matrix composites. The results of permittivity measurements of the thermoplastic composites at room and elevated temperatures using the waveguide transmission and probe kit techniques are described in section 9.4. Section 9.5 details the results for the loss tangent of the low loss thermoplastic matrix composite materials using the resonant cavity method. Section 9.6 gives details of the best frequency range to process the five thermoplastic composite materials. The results of the bond strength tests of the composites joined by fixed and variable frequency microwave irradiation are discussed in section 9.7.

From the measured data of LDPE/GF (33%), PS/GF (33%) and nylon 66/GF (33%), it was found that at higher temperatures and higher frequencies, these materials became less reflective and permitted more microwave energy to enter them. At the same time, their dielectric loss factor increased with temperature and frequency and enabled more of the absorbed energy to be converted to heat. Therefore, within limits, higher temperatures and higher frequencies were more suitable for microwave-assisted joining of the three materials. As to the primer/filler, only liquid rapid araldite was found to be the favourable in microwave-assisted joining of FRTP composites because of its higher values of loss tangent. Finally, the dielectric probe method was found to be the recommended technique to use for measurements of $\varepsilon'$ and $\tan \delta$ in microwave joining of materials.
9. Results and Discussion

9.2 Summary of All Findings

- discovering that dilution of Araldite into the thermoplastic composite and vice-versa had taken place during the joining of the latter by microwave irradiation (sections 9.3 9.8.1 and 9.8.2); this also explains the higher bond strength obtained;
- measuring the values of dielectric constant of three types of fibre glass reinforced thermoplastic matrix composite materials using the waveguide transmission technique (section 9.4.1);
- measuring the values of the dielectric constant and loss of two types of fibre glass reinforced thermoplastic matrix composite materials, and of three types of Araldite using the dielectric probe method (section 9.4.2);
- measuring the loss tangent of low loss fibre glass reinforced thermoplastic matrix composite materials, eg LDPE/GF (33%), using the resonant cavity method (section 9.5);
- discovering the best frequency range to process or join five types of fibre reinforced thermoplastic matrix composite materials using VFM (section 9.6);
- discovering, in terms of bond strength, the best combination of power level and time of exposure to fixed frequency microwave irradiation for joining the 5 types of thermoplastic matrix composite materials; also that the power level was vital in joining thermoplastic composites irrespective of whether fixed or variable frequency microwave irradiation was used; and furthermore that prolonged exposure of the carbon fibre reinforced thermoplastic composite materials to microwave energy is harmful (section 9.8.4);
- discovering that for LDPE/GF(33%) the bond strength does not improve much with increased duration of microwave irradiation, irrespective of whether fixed or variable frequency microwave energy is used (section 9.7.3);
- discovering that VFM gave better bond quality (section 9.7.7.3);
- discovering that fixed frequency microwave energy was not suitable for joining carbon fibre reinforced thermoplastic composites (section 9.7.7.3).
discovering that the microwave energy absorbed and converted to heat by the carbon fibres in the joining of carbon fibre reinforced thermoplastic composite materials is more than enough to break the C-C and C-H bonds of the matrix of the composites; the thermoplastic composite materials were charred and hence weakened (sections 9.8.2, 9.8.3 and 9.8.4)

**9.3 Results of Surface Analysis**

In this research project, some of the bond shear strengths of test pieces obtained, after exposure to a high power level of microwave energy, were higher than those cured conventionally because the primer may have diffused into the parent material and vice versa at the interface (Ku et al, 2000e). A low power microscopy study and tests by x-ray photoelectron spectroscopy were carried out on the joined surfaces of the composite to prove, or otherwise, the diffusion of the Araldite into the thermoplastic and vice versa.

**9.3.1 Low Power Microscopy**

First, two of the test pieces of glass fibre reinforced (33% by weight) low density polyethylene were joined by the two-part five-minute liquid Araldite without exposure to microwave energy, ie cured in ambient conditions. After crosscutting the lap joint, the joined area was viewed under a low power microscope. At sixty (60) times magnification, a white pale strip of bondline was found present on a grey background. With 100 and 400 times magnification, similar observations were found and there were a lot of glass fibre filaments around the area adjacent to the interface. Similarly, two test pieces were lap-joined with the aid of the same primer and were exposed to microwave energy at a power level of 800W for 70 seconds. Again, after crosscutting the lap joint, the joined area was investigated under a low power microscope. At 30 times magnification, the interface was easily identified as depicted in Plate 2 but there was no trace of glass fibre filaments. At higher magnifications of 300 and 1500 times, a considerable amount of glass fibre filaments were found on the bondline
as illustrated in Plates 3 and 4 and this implied that diffusion of the thermoplastic had taken place because glass fibre filaments were part of the composite.

Plate 2: 30 Times Microscopy Photo of Diffusion of Araldite into Thermoplastic Composite

Plate 3: 300 Times Microscopy Photo of Diffusion of Araldite into Thermoplastic Composite

9.3.2 X-ray Photoelectron Spectroscopy
The phenomenon of diffusion was further confirmed by the x-ray photoelectron spectroscopy surface analysis. Figure 9.1 shows the O and C atom intensities against the binding energy of LDPE/GF (33%) plus Araldite after the material had

Plate 4: 1500 Times Microscopy Photo of Diffusion of Araldite into Thermoplastic Composite

Figure 9.1: The O and C Atom Intensities against Binding Energy of LDPE/GF (33%) Plus Araldite.
been exposed to 800 W microwave energy for 70 seconds. Figure 9.2 shows the C-C and -C-O bond intensities against the binding energy of LDPE/GF (33%) plus Araldite. From the two figures, the percentage of different atoms present in LDPE/GF (33%) plus Araldite was worked out and is shown in Table 9.1.

![Figure 9.2: The -C-C- and -C-O Bond Intensities against the Binding Energy of LDPE/GF (33%)](image)

<table>
<thead>
<tr>
<th>Element</th>
<th>Area</th>
<th>ASF</th>
<th>Area/ASF</th>
<th>Atom (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>95,810</td>
<td>1</td>
<td>95,810</td>
<td>90.5</td>
</tr>
<tr>
<td>O</td>
<td>25,091</td>
<td>3</td>
<td>8,364</td>
<td>7.9</td>
</tr>
<tr>
<td>Si</td>
<td>680</td>
<td>1.02</td>
<td>667</td>
<td>0.6</td>
</tr>
<tr>
<td>N</td>
<td>1,177</td>
<td>1.84</td>
<td>640</td>
<td>0.6</td>
</tr>
<tr>
<td>Ca</td>
<td>2,654</td>
<td>6.3</td>
<td>421</td>
<td>0.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>105,901</td>
<td>100</td>
</tr>
</tbody>
</table>
Figure 9.3 illustrates the O and C atom intensities against the binding energy of Araldite on glass. Figure 9.4 depicts the –C-C-, -C-O and –C=O bond intensities against the binding energy of Araldite on glass. From the two figures, the percentage of different atoms in Araldite was worked out and is shown in Table 9.2. The C atom intensity against the binding energy of low density polyethylene
9. Results and Discussion

Figure 9.4: The -C-C-, -C-O and –C=O Bond Intensities against the Binding Energy of Araldite

Table 9.2: Percentage of Atoms of Araldite on Glass.

<table>
<thead>
<tr>
<th>Element</th>
<th>Area</th>
<th>ASF</th>
<th>Area/ASF</th>
<th>Atom (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>96,949</td>
<td>1</td>
<td>96,949</td>
<td>83.6</td>
</tr>
<tr>
<td>O</td>
<td>47,819</td>
<td>3</td>
<td>15,940</td>
<td>13.8</td>
</tr>
<tr>
<td>N</td>
<td>2,836</td>
<td>1.84</td>
<td>1,514</td>
<td>1.3</td>
</tr>
<tr>
<td>S</td>
<td>3,209</td>
<td>2.09</td>
<td>1,535</td>
<td>1.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>115,965</td>
<td>100</td>
</tr>
</tbody>
</table>

(LDPE) is shown in Figure 9.5. The C-C bond intensity against binding energy of LDPE is shown in Figure 9.6.
9. Results and Discussion

Figure 9.5: The C Atom Intensity against the Binding Energy of LDPE.

Figure 9.6: The -C-C- Bond Intensity against the Binding Energy of LDPE.
The percentage of atoms of the three different materials was studied and summarised in Table 9.3. Composite 1 was formed by having some rapid Araldite mixed into LDPE/GF (33%). Composite 2 was formed by having some LDPE/GF (33%) melted into the rapid Araldite. In fact Composite 1 is exactly equal to Composite 2 and only differs by the way by which they are expressed. The relative percentage of oxygen in Composite 2 (7.9 %) was lower than that in araldite (13.8%) and this was due to melting of LDPE/GF (33%) and subsequently transferring into the Araldite. The relative amount of oxygen was diluted. The relative percentage of oxygen in Composite 1 (7.9%) was higher than that in pure LDPE (0%) and this was because some Araldite had entered this composite. It could be argued that oxygen came from the reinforcing glass fibre and in this case the oxygen content of Composite 1 should only be four times the percentage of silicon, ie. 2.4 percent. However, this was not the case since oxygen was also present in ‘C-O’ form which indeed was present in Araldite. So it was deduced that the oxygen content in Composite 1 was mainly due to the araldite and as a conclusion, LDPE/GF (33%) was transferred into the Araldite.

Table 9.3. : Summary of the Percentage of Atoms for Three Different Materials Analysed.

<table>
<thead>
<tr>
<th>Atom Percentage Materials</th>
<th>Oxygen</th>
<th>Carbon</th>
<th>Silicon</th>
<th>Calcium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pure LDPE</td>
<td>0</td>
<td>A lot</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Rapid Araldite</td>
<td>13.8</td>
<td>83.6</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>[LDPE/GF(33%) + Rapid Araldite] = Composite 1</td>
<td>7.9</td>
<td>90.5</td>
<td>0.6</td>
<td>0.4</td>
</tr>
<tr>
<td>[Rapid Araldite + LDPE/GF(33%)] = Composite 2</td>
<td>7.9</td>
<td>90.5</td>
<td>0.6</td>
<td>0.4</td>
</tr>
</tbody>
</table>

9.3.3 Diffusion Theory Explanation

From the study and experiments in sections 9.3.1 and 9.3.2, it was concluded that the molecules of the Araldite had diffused into the LDPE/GF (33%) and vice-versa. Diffusion theory prevailed in this circumstance because the application of pressure and microwave heat had facilitated the intrinsic adhesion of polymers to
themselves (autohesion), and to each other. The macromolecules or chain segments of the polymers (adhesive and substrate) therefore possessed sufficient mobility and became mutually soluble. The original contribution of this section that was achieved was to discover that diffusion of Araldite into the thermoplastic composite and vice-versa had taken place during joining using microwave irradiation. This also explained the higher bond strength obtained.

9.4 Results of Permittivity Measurements.

This section describes the results of permittivity measurements in two parts. The first part of this section includes results of the permittivity measurements using the waveguide transmission technique. The next part describes the permittivity measurement results using the dielectric probe kit.

9.4.1 Results of the Waveguide Transmission Technique

In order to verify that the transmission line setup used measures what it ought to measure, materials with known dielectric constant values were assessed. Air and Teflon were chosen for this purpose and their dielectric constants were measured at room temperature. Figure 9.7 shows the dielectric constants for air and Teflon is 1.0 and 2.0 respectively at room temperature. These coincided very well with data in the literature (Metaxas and Meredith, 1983; von Hippel, 1995) and it was concluded that the equipment measured what it was supposed to. However, the dielectric loss for the two materials was not reliably measured as negative values were recorded; their losses were too low. The loss through the waveguide wall was of the same order as the loss of the materials and some compensation would need to be included into the software program to generate reasonable results. Figure 9.7 also illustrates the dielectric constant of nylon 66, low-density polyethylene (LDPE), and glass fibre-reinforced LDPE and glass fibre-reinforced nylon 66. The dielectric constant and loss of acrylic at room temperature (25°C)
and at the frequency of 3 GHz, as measured by the set-up, matched the published data (Metaxas and Meredith, 1983; von Hippel, 1995) of 2.6 and 0.015 respectively.

![Dielectric Constant Graph](image)

Figure 9.7: Dielectric Constants of Air, Teflon, LDPE/GF (33%), nylon 66/GF (33%), Nylon 66 at Room Temperature over Certain Frequencies.

The first material under test was 33% glass-fibre reinforced low-density polyethylene [LDPE/GF (33%)]. Figure 9.8 shows the relationship between the values of dielectric constant of LDPE/GF (33%) over a range of frequencies. It indicated that the values of dielectric constant did not change much with frequency and temperature changes but a tendency was established that the higher the temperature, the lower would be the dielectric constant, particularly at the higher frequency end.

As there was no published data for the values of $\varepsilon'$ of LDPE/GF (33%), simulated data was used to verify the authenticity of the measured data. The values (Metaxas and Meredith, 1983; von Hippel, 1995) of $\varepsilon'$ of LDPE (2.25) and glass fibre (3.78) at 25°C and at 3 GHz were employed to generate the $\varepsilon'$ of LDPE/GF (33%) at the same physical conditions. It was assumed that the mixture was homogeneous and the $\varepsilon'$ of the material was calculated by proportion as follows:

$$2.25 \times \frac{67}{100} + 3.78 \times \frac{33}{100} = 2.755$$
The dielectric constant measured by the waveguide transmission technique at the same physical condition was 2.6. This agreement showed that the measured data was rational. By a similar argument, $\varepsilon''$ could also be simulated and it was found to be 0.0018. This suggested that LDPE/GF (33%) should therefore be a low loss material and it was not appropriate to measure the loss tangent by this method.

The next material to be studied was PS/GF (33%). Figure 9.9 shows the values of dielectric constant of PS/GF (33%) over a range of frequencies. It depicts that the values of dielectric constant did not change much with frequency and temperature, but the dielectric constant tended to be lower at higher temperatures, especially towards the higher frequency end. Values of $\varepsilon'$ and $\varepsilon''$ of PS/GF (33%) were similarly simulated from published data (Blackham et al, 1991) and they were found to be 2.92 and 0.0021 respectively at 25°C and at 3 GHz. On the other hand, the measured values at the same conditions were 2.90 and 0.0136 respectively. The measured value of $\varepsilon'$ was very near to the simulated one and seemed to be reasonably accurate; while, the measured value of $\varepsilon''$ was much higher than the simulated one (6.5 times) and it was 7.6 times higher than that of the simulated LDPE/GF (33%). Since the $\varepsilon''$ of PS was low and was very near to
that of LDPE, it was therefore thought that the values obtained may be unreliable and further investigations were needed.

The last material to be discussed was nylon 66/GF (33%) where the higher the values of the temperature and frequency, the lower the values of the dielectric constant as depicted in figures 9.10 and 9.11 respectively. The simulated value of $\varepsilon'$ for nylon 66/GF (33%) at 10 GHz and at 25°C was 3.28 while that procured by measurement was 3.66; the measured value was 10% higher. The trends of the values of the measured dielectric constant for the three materials were the same and they followed the trend for most materials.

Figures 9.12 and 9.13 illustrate the change of loss tangent values for nylon 66/GF (33%) with frequency and temperature changes respectively. The simulated value of $\tan \delta$ for nylon 66/GF (33%) at the same ambient conditions was 0.0072 and the measured one was 0.0232; the measured value was 69% larger. This showed that the higher the temperature, the higher the value of the dielectric loss factor, but no such a trend with frequency existed. Since nylon 66 is a relatively higher loss material, it was reckoned that the values of the loss tangents obtained were reliable. Referring to Figure 9.13, at a frequency of 7 GHz and at a temperature of 90°C, the dielectric loss factor was at maximum. At this point, the maximum
amount of microwave energy penetrated was converted to heat to facilitate the joining process but on the other hand, the dielectric constant was relatively high and microwave energy penetration to the material was not adequate. A compromise value of $\tan \delta$ and $\varepsilon'$ was therefore chosen to obtain the greatest microwave energy penetration and maximum energy conversion to heat. Similar arguments were be made for LDPE/GF (33%) and PS/GF (33%) and joining the material by microwave energy at 12.5 GHz and at 95 degrees Celsius did not provide the optimum result.
9. Results and Discussion

Figure 9.12: Dielectric Loss Factors of Nylon 66/ GF (33%) at Elevated Temperatures over Certain Frequencies.

Figure 9.13: Dielectric Loss Factors of Nylon66/ GF (33%) at Different Frequencies over a Range of Temperatures.

9.4.2 Results from the Dielectric Probe Method

In order to verify that the equipment measured correctly, materials of known dielectric constant and loss values were first tested; $\varepsilon'$ and $\varepsilon''$ of pure water were therefore measured at the frequency range of 2 - 12.5 GHz and at 25°C. The measured values of $\varepsilon'$ and $\varepsilon''$ of pure water at 25°C and at 2 GHz and at 12.5 GHz are included with those of published data in Table 9.4 (Senko and Tran, 1996;
1997; Metaxas and Meredith, 1983; von Hippel, 1995). From this table it was found that there was some discrepancy in values between the measured and the published data (Senko and Tran, 1996; 1997; Metaxas and Meredith, 1983; von Hippel, 1995), but the variation did not suggest that results were erroneous.

Table 9.4: Dielectric Constant and Loss Values of Pure Water at 25°C

<table>
<thead>
<tr>
<th></th>
<th>2 GHz Values</th>
<th>12.5 GHz Values</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Measured</td>
<td>Literature</td>
</tr>
<tr>
<td>Dielectric Constant</td>
<td>77.6</td>
<td>80</td>
</tr>
<tr>
<td>Dielectric Loss</td>
<td>7.82</td>
<td>6.1</td>
</tr>
</tbody>
</table>

LDPE/GF (33%) was tested over a wide band of 2 - 12.5 GHz and a narrow band of 2.2 - 2.5 GHz, at room temperature. The material was cut into a diameter larger than 20 mm and a thickness \((\varepsilon' \cdot 20 \text{mm})\) as the \(\varepsilon'\) of LDPE/GF (33%) was 2.6, the thickness was made larger than 12.54 mm. Results of \(\varepsilon'\) and \(\tan \delta\) are included in Tables 9.5 and 9.6 respectively.

Over the frequency range of 2- 12.5 GHz, the probe kit measurements gave values of \(\varepsilon'\) from 2.69 in the lower frequency range to 2.61 at the higher frequency end. Values obtained were not too far away from those obtained by the waveguide transmission technique, but tended to be comparatively lower at higher frequencies. As to the \(\tan \delta\), the values increased progressively with frequencies. At or above 5 GHz the values were over 0.05 which was considered valid (HP85070B Technical Data Sheets, 1993; HP 85070A Technical Data Sheets, 1990) as far as the probe kit measurements were concerned, but the values were higher than those obtained using the simulated value of 0.0007 and were regarded as unreliable since it is well known that LDPE/GF (33%) is a low loss material. Over the narrow frequency range of 2.2 - 2.5 GHz, the \(\varepsilon'\) values were almost constant at 2.74. The values of \(\tan \delta\) were considered not valid within the said narrow frequency range because they were too large in magnitude.
Table 9.5: Dielectric Constant Values of LDPE/GF (33%), PS/GF (33%), LRA and CRA over a Wide Band of Frequencies at 25°C

<table>
<thead>
<tr>
<th>Materials</th>
<th>LDPE/GF(33%)</th>
<th>PS/GF(33%)</th>
<th>LRA</th>
<th>CRA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency (GHz)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>2.65</td>
<td>3.05</td>
<td>2.81</td>
<td>2.41</td>
</tr>
<tr>
<td>3</td>
<td>2.69</td>
<td>3.14</td>
<td>2.65</td>
<td>2.39</td>
</tr>
<tr>
<td>4</td>
<td>2.69</td>
<td>3.13</td>
<td>2.80</td>
<td>2.58</td>
</tr>
<tr>
<td>5</td>
<td>2.68</td>
<td>3.11</td>
<td>2.92</td>
<td>2.45</td>
</tr>
<tr>
<td>6</td>
<td>2.69</td>
<td>3.14</td>
<td>2.51</td>
<td>2.40</td>
</tr>
<tr>
<td>6.5</td>
<td>2.69</td>
<td>3.10</td>
<td>2.80</td>
<td>2.39</td>
</tr>
<tr>
<td>7.5</td>
<td>2.7</td>
<td>3.09</td>
<td>2.58</td>
<td>2.49</td>
</tr>
<tr>
<td>8</td>
<td>2.68</td>
<td>3.08</td>
<td>2.59</td>
<td>2.36</td>
</tr>
<tr>
<td>9.5</td>
<td>2.64</td>
<td>3.02</td>
<td>2.42</td>
<td>2.33</td>
</tr>
<tr>
<td>11</td>
<td>2.61</td>
<td>2.98</td>
<td>2.39</td>
<td>2.33</td>
</tr>
<tr>
<td>12.5</td>
<td>2.64</td>
<td>2.95</td>
<td>2.39</td>
<td>2.31</td>
</tr>
</tbody>
</table>

Table 9.6: Loss Tangent Values of LDPE/GF (33%), PS/GF (33%), LRA and CRA over a Wide Band of Frequencies at 25°C

<table>
<thead>
<tr>
<th>Materials</th>
<th>LDPE/GF(33%)</th>
<th>PS/GF(33%)</th>
<th>LRA</th>
<th>CRA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency (GHz)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.014</td>
<td>0.006</td>
<td>0.244</td>
<td>0.060</td>
</tr>
<tr>
<td>3</td>
<td>0.012</td>
<td>0.011</td>
<td>0.259</td>
<td>0.064</td>
</tr>
<tr>
<td>4</td>
<td>0.031</td>
<td>0.021</td>
<td>0.263</td>
<td>0.070</td>
</tr>
<tr>
<td>5</td>
<td>0.049</td>
<td>0.033</td>
<td>0.395</td>
<td>0.082</td>
</tr>
<tr>
<td>6</td>
<td>0.052</td>
<td>0.045</td>
<td>0.400</td>
<td>0.086</td>
</tr>
<tr>
<td>6.5</td>
<td>0.056</td>
<td>0.050</td>
<td>0.414</td>
<td>0.097</td>
</tr>
<tr>
<td>7.5</td>
<td>0.062</td>
<td>0.062</td>
<td>0.413</td>
<td>0.102</td>
</tr>
<tr>
<td>8</td>
<td>0.077</td>
<td>0.070</td>
<td>0.413</td>
<td>0.117</td>
</tr>
<tr>
<td>9.5</td>
<td>0.081</td>
<td>0.075</td>
<td>0.417</td>
<td>0.106</td>
</tr>
<tr>
<td>11</td>
<td>0.099</td>
<td>0.076</td>
<td>0.420</td>
<td>0.142</td>
</tr>
</tbody>
</table>

Another material under test was PS/GF (33%) and the results for ε' and tan δ over a frequency range of 2 - 12.5 GHz are given in Tables 9.5 and 9.6 respectively. The ε’ values varied from 3.1 at lower frequencies to 2.95 at higher frequencies.
and found to be slightly higher than the simulated values and those obtained by the waveguide transmission technique. The $\varepsilon''$ values above 6 GHz were found to be acceptable (HP85070A Technical Data Sheets, 1990; HP85070B Technical Data Sheets, 1993) as far as the probe kit method was concerned but since they were relatively higher in reality, they were not accepted.

The next material to be discussed was the two-part five-minute rapid Araldite, a primer used in the process. Part A of the Araldite was 100% liquid epoxy resin; part B of the Araldite was 8% amine. It was considered in two forms: liquid form rapid Araldite (LRA) and cured form rapid Araldite (CRA). In both cases, the two parts of Araldite were mixed and then poured into cylindrical aluminium tubes with an internal diameter of $22 \text{ mm}$ and a thickness larger than $\frac{20}{(\varepsilon')^2} \text{ mm}$ (HP85070A Technical Data Sheets, 1990; HP85070B Technical Data Sheets, 1993), where $\varepsilon'$ was the dielectric constant of the cured rapid araldite from literature (Metaxas and Meredith, 1983). The cured ones were prepared days before the measurements but they were still in semi-solid form during the test and cling wrapped. A thin film of LDPE (cling wrap) was placed on top of the cylindrical aluminium tubes to prevent the probe making direct contact with the material. They were tested in semi-solid conditions and when the probe was applied on the specimens, the surfaces were deformed and became much smoother so that the required parameters were measured correctly (HP85070A Technical Data Sheets, 1990; HP85070B Technical Data Sheets, 1993). The liquid ones were poured into the aluminium cylinders immediately prior to commencing measurements and again cling wrap was used to cover the top end of the cylinders. The values of $\varepsilon'$ of LRA decreased with increasing frequency while those of tan $\delta$ increased with increasing frequency. Results are provided in Tables 9.5 and 9.6. This implied that processing at higher frequencies favoured the process. It was found that the $\varepsilon'$ values of CRA were lower than those of LRA and this meant that microwaves entering the material penetrated further into
the material when the araldite was in liquid form. Furthermore, tan δ values of LRA were found to be much higher than in the fully cured specimens, thus suggesting that more of the energy that penetrated into the material be converted into heat. On account of the above behaviour, it was logical to conclude that the microwave processing time should be short to ensure that the araldite is not cured before the end of the processing cycle, since its liquid form favoured the process.

Another material tested was the rapid tertiary araldite. Part A of this resin was 100% epoxy resin and part B of it was 8% tertiary amine. It was in the liquid as well as the cured form. The two parts of the Araldite were mixed and then poured into the aluminium cylinders. Tables 9.7 and 9.8 show values of $\varepsilon'$ and tan δ the liquid rapid tertiary Araldite (LRTA) and cured rapid tertiary rapid Araldite (CRTA). The values of $\varepsilon'$ of LRTA were found to be similar to those of LRA but the values of tan δ of LRTA were up to 50% lower than its counterpart. However, the trend obtained was the same; values became larger as frequency increased. As to CRTA, the values of $\varepsilon'$ and tan δ were found to be similar to those of its counterpart.

Table 9.7: Values of $\varepsilon'$ of LRTA, CRTA, LHSA and CHSA over a Range of Frequencies

<table>
<thead>
<tr>
<th>Materials</th>
<th>Frequency (GHz)</th>
<th>LRTA</th>
<th>CRTA</th>
<th>LHSA</th>
<th>CHSA</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2</td>
<td>2.62</td>
<td>2.77</td>
<td>3.12</td>
<td>3.46</td>
</tr>
<tr>
<td></td>
<td>3.5</td>
<td>2.61</td>
<td>2.67</td>
<td>3.14</td>
<td>3.34</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>2.61</td>
<td>2.74</td>
<td>3.35</td>
<td>3.38</td>
</tr>
<tr>
<td></td>
<td>6.5</td>
<td>2.37</td>
<td>2.67</td>
<td>3.18</td>
<td>3.29</td>
</tr>
<tr>
<td></td>
<td>8</td>
<td>2.58</td>
<td>2.6</td>
<td>3.17</td>
<td>3.17</td>
</tr>
<tr>
<td></td>
<td>9.5</td>
<td>2.44</td>
<td>2.63</td>
<td>3.15</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>11</td>
<td>2.53</td>
<td>2.52</td>
<td>3.16</td>
<td>2.97</td>
</tr>
</tbody>
</table>

The last filler material to be discussed was the high strength Araldite. Part A of this Araldite was 100% liquid epoxy resin and part B was 100% polyamide. It was in two forms; liquid and cured. The values of $\varepsilon'$ and tan δ of liquid high
### Table 9.8: Values of tan δ of LRTA, CRTA, LHSA and CHSA over a Range of Frequencies

<table>
<thead>
<tr>
<th>Materials</th>
<th>LRTA</th>
<th>CRTA</th>
<th>LHSA</th>
<th>CHSA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency (GHz)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.117</td>
<td>0.086</td>
<td>0.2</td>
<td>0.073</td>
</tr>
<tr>
<td>3.5</td>
<td>0.116</td>
<td>0.089</td>
<td>0.167</td>
<td>0.079</td>
</tr>
<tr>
<td>5</td>
<td>0.129</td>
<td>0.112</td>
<td>0.249</td>
<td>0.112</td>
</tr>
<tr>
<td>6.5</td>
<td>0.163</td>
<td>0.123</td>
<td>0.277</td>
<td>0.142</td>
</tr>
<tr>
<td>8</td>
<td>0.196</td>
<td>0.133</td>
<td>0.278</td>
<td>0.181</td>
</tr>
<tr>
<td>9.5</td>
<td>0.222</td>
<td>0.134</td>
<td>0.280</td>
<td>0.201</td>
</tr>
<tr>
<td>11</td>
<td>0.275</td>
<td>0.141</td>
<td>0.285</td>
<td>0.201</td>
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<tr>
<td>12.5</td>
<td>0.287</td>
<td>0.140</td>
<td>0.286</td>
<td>0.203</td>
</tr>
</tbody>
</table>

Strength Araldite (LHSA) and cured high-strength Araldite (CHSA) are included in Tables 9.7 and 9.8. The values of $\varepsilon'$ and tan δ of LHSA and CHSA were found to be more or less the same. This may be due to the fact that even after days of curing, the CHSA was not fully cured and retained its liquid form dielectric properties. However, their tan δ values were much lower than those of LRA, and CHSA was considered to be not as suitable as a primer in microwave processing.

### 9.4.3 Waveguide Transmission Technique versus Dielectric Probe

Figure 9.14 shows the values of $\varepsilon'$ of LDPE/GF (33%) of the two methods plotted together over the frequency range of 2 - 12.5 GHz and at room temperature. Results matched very well but were lower than the simulated value of 2.755.

Figure 9.15 shows the values of $\varepsilon'$ of PS/GF (33%) of the two methods plotted together over the frequency range of 2 - 12.5 GHz and at room temperature. The probe kit results were higher than those obtained from the waveguide transmission technique, particularly in the lower frequency range, but still much lower than the simulated values.
9. Results and Discussion

Figure 9.14: The Dielectric Constants of LDPE/GF (33%) By Two Methods Over a Frequency Range of 2 - 12.5 GHz.

Figure 9.15: The Dielectric Constants of PS/GF (33%) By Two Methods over a Frequency Range of 2 - 12.5 GHz.

The waveguide transmission technique accurately measured the ε’ and ε” values of several commonly used materials like Teflon, air, LDPE/GF (33%) and PS/GF (33%). The situation was the same for the dielectric probe method except in the case of pure water, where some discrepancies occurred. Even though it seemed that the waveguide transmission technique (Blackham et al, 1991) produced more accurate values of ε’ of PS/GF (33%), there was no solid evidence to prove that it
was better than its counterpart in an industrial environment. In fact it proved to be more complicated and needed many different sizes of waveguide to measure values of the $\varepsilon'$ over a range of frequencies, eg 2 - 20 GHz. The dielectric probe method was found to be easy to use; sample preparation was also easier and the re-calibration facility made the re-calibration at each temperature of measurement affordable. If the accuracy required for the values of $\varepsilon'$ and $\tan \delta$ was not too significant, like in the case of microwave processing, then the dielectric probe method should be chosen. The HP probe kit was designed for measuring the $\varepsilon'$ and $\tan \delta$ values at elevated temperatures up to 200°C and over a range of frequencies from 200MHz to 20 GHz.

9.4.4 Concluding Remarks

From the measured data of LDPE/GF (33%), PS/GF (33%) and nylon 66/GF (33%), it was found that at higher temperatures and higher frequencies, these materials became less reflective and permitted more microwave energy to enter them because, their dielectric loss factor increased with temperature and frequency thus enabling more of the absorbed energy to be converted into heat. Therefore, within limits, higher temperatures and higher frequencies were more suitable for microwave-assisted joining of the three materials. As to the primer/filler, only liquid rapid Araldite was found to be the most favourable in microwave-assisted joining of FRTP composites because of its higher value of loss tangent. The dielectric probe method was the recommended technique to use for measurements of $\varepsilon'$ and $\tan \delta$ in microwave joining of materials, where accuracy is not important.
9.5 Results of Loss Tangent Using A Resonant Cavity Method

The loss tangents of low loss thermoplastic composite materials could not be measured using the methods mentioned in chapter 6. A resonant cavity method (chapter 7) was therefore used and the values of the dielectric loss, dielectric constant and Q factors of LDPE/GF (33%) using WR340, WR229 are given in Tables 9.9 - 9.10.

Table 9.9: Values of $\varepsilon'$ and $\varepsilon"$ for WR340 with Sample [LDPE/GF (33%)] Standing Upright.

<table>
<thead>
<tr>
<th>Temp</th>
<th>Dielectric Loss</th>
<th>Dielectric Constant</th>
<th>$Q_o$</th>
<th>$Q_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>24°C</td>
<td>0.00606</td>
<td>2.338</td>
<td>4795</td>
<td>1169</td>
</tr>
<tr>
<td>42°C</td>
<td>0.00678</td>
<td>2.325</td>
<td>4873</td>
<td>1077</td>
</tr>
<tr>
<td>60°C</td>
<td>0.00684</td>
<td>2.316</td>
<td>5005</td>
<td>1076</td>
</tr>
<tr>
<td>80°C</td>
<td>0.00711</td>
<td>2.315</td>
<td>4810</td>
<td>1035</td>
</tr>
</tbody>
</table>

Table 9:10: Values of $\varepsilon'$ and $\varepsilon"$ for WR229 with Sample [LDPE/GF (33%)]

<table>
<thead>
<tr>
<th>Temp.</th>
<th>Dielectric Loss</th>
<th>Dielectric Constant</th>
<th>$Q_o$</th>
<th>$Q_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>24°C</td>
<td>0.08090</td>
<td>2.440</td>
<td>1908</td>
<td>559</td>
</tr>
<tr>
<td>40°C</td>
<td>0.08169</td>
<td>2.540</td>
<td>1834</td>
<td>550</td>
</tr>
<tr>
<td>60°C</td>
<td>0.09099</td>
<td>2.534</td>
<td>1896</td>
<td>514</td>
</tr>
<tr>
<td>70°C</td>
<td>0.09444</td>
<td>2.368</td>
<td>1923</td>
<td>502</td>
</tr>
<tr>
<td>80°C</td>
<td>0.09644</td>
<td>2.527</td>
<td>2017</td>
<td>500</td>
</tr>
<tr>
<td>90°C</td>
<td>0.00983</td>
<td>2.525</td>
<td>2017</td>
<td>497</td>
</tr>
</tbody>
</table>

With both WR340 and WR229, the values of the dielectric loss of the two sets of data were quite close to each other. In both cases, the values increased with the rise in temperature. Although the increase was insignificant, a trend was established. With WR340, ie at a lower frequency, the values of the dielectric loss ranged from 0.00606 at room temperature to 0.0071 at 80°C; while those with WR229, ie at a higher frequency ranged from 0.00809 at 24°C to 0.00983 at 90°C. The values were, however, three to five times higher than the simulated
value of 0.0018 obtained at 3GHz and 25°C (Ku et al, 2000a). It could be argued that the experimental results were more reliable and accurate than the simulated one because they had been measured separately and independently and in different frequency ranges.

Now looking at the values of the dielectric constant of the material under test; it was found that both the results of WR340, decreasing from 2.338 at 24°C to 2.315 at 80°C, and of WR229, ranging down from 2.44 at 24°C, to 2.525 at 90°C, were lower than the measured data of 2.6 at room temperature (Ku et al, 1999b; 2000a; 1998), and simulated data of 2.755 (Ku et al, 2000a) respectively. The value of 2.6 was obtained from two different measurement techniques, waveguide transmission and dielectric probe methods (Ku et al, 2000a) and was therefore regarded as accurate and reliable. The values of dielectric constant from Tables 10.9 and 10.10, however, had the same trend as those measured from the other two techniques (Ku et al, 2000a), ie the values decreased with increasing temperature.

By applying the results in Table 9.11 and considering the results of WR159; it was found that the values of the dielectric loss of the MUT increased from 0.0209 at 24°C to 0.0217 at 80°C. Again, although the increase was insignificant, a trend was established. The values were much higher than those found in WR340 and WR229. The values were approximately three times higher than those found in WR340 and about twice than those found in WR229. It was difficult, at this point in time to conclude whether the results of WR159 were correct or not. But, first consider the results of WR90 at room temperature as tabulated in Table 9.12. It was found that the dielectric loss of the composite was 0.0234 that was very near to the results obtained from WR159. It was, however, inappropriate to make a conclusion on the correctness of the values of the dielectric loss of the composite at this moment. Considering the values of the dielectric constant of the MUT from WR159 and WR90; in both cases, the values ranged from 1.002 to

<table>
<thead>
<tr>
<th>WR159</th>
<th>Dielectric</th>
<th>Dielectric</th>
<th>Qo</th>
<th>Qs</th>
</tr>
</thead>
</table>

Table 9.11: Values of $\varepsilon'$ and $\varepsilon''$ for WR159 with Sample [LDPE/GF (33%)] Standing Upright.
9. Results and Discussion

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Loss</th>
<th>Constant</th>
</tr>
</thead>
<tbody>
<tr>
<td>24°C</td>
<td>0.0209</td>
<td>1.054</td>
</tr>
<tr>
<td>42°C</td>
<td>0.0217</td>
<td>1.054</td>
</tr>
<tr>
<td>60°C</td>
<td>0.0239</td>
<td>1.120</td>
</tr>
<tr>
<td>80°C</td>
<td>0.0217</td>
<td>1.100</td>
</tr>
</tbody>
</table>

Table 9.12: Values of $\epsilon'$ and $\epsilon''$ for WR90 with Sample [LDPE/GF (33%)] Standing Upright.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>WR90 Dielectric Loss</th>
<th>WR90 Dielectric Constant</th>
<th>$Q_o$</th>
<th>$Q_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>24°C</td>
<td>0.0234</td>
<td>1.0025</td>
<td>1830</td>
<td>277</td>
</tr>
</tbody>
</table>

1.112 which were very far away from the value of 2.6 obtained from other measuring techniques (Ku et al, 1999b; 2000a). The values of dielectric constant obtained from WR159 and WR 90 were therefore incorrect and were discarded. On the other hand, in order to make a conclusion on the values of the dielectric loss of the composite, a slight modification was made to the positioning of the composite material inside the WR159. The sample was laid down at the bottom of the cavity as depicted in Figure 9.16 and the slab was also trimmed to a thickness of only 2 mm to ensure its volume was not too large as compared to the volume of the waveguide. To calculate the $\epsilon'$ and $\epsilon''$ with the sample laying at the bottom of the cavity, Eqn 7.2 and Eqn 7.3 had to be manipulated in a different way. Eqn 7.2 has now become (Ku et al, 2000a)

$$
\epsilon' = \frac{1}{1 - \frac{2H}{t} \left( \frac{f_o - f_s}{f_o} \right)}
$$

and Eqn 7.3 (Ku et al, 2000a)

$$
\frac{\epsilon''}{\epsilon'} = \tan \delta = \left( \frac{1}{Q_s} - \frac{1}{Q_o} \right) \left[ 1 + \epsilon' \left( \frac{H}{t} - 1 \right) \right]
$$

\(9.2\)
9. Results and Discussion

Results of the measurements are included in Table 9.13, which shows that the values of the dielectric loss of the material ranged from 0.0196 at 24°C to 0.0262 at 80°C. They were very close to those obtained in Tables 9.11 and 9.12. At the same time, the values of the dielectric constant obtained were also very close to those achieved by the other measuring techniques. It could therefore be argued that both the values of the dielectric loss and dielectric constant obtained from WR159 with the sample lying at the bottom of the cavity were correct and accurate.

Table 9.13: Values of $\varepsilon'$ and $\varepsilon''$ for WR159 with Sample [LDPE/GF (33%)] Laying at the Bottom.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>WR159 Dielectric Loss</th>
<th>WR159 Dielectric Constant</th>
<th>WNQ</th>
<th>WNQ</th>
</tr>
</thead>
<tbody>
<tr>
<td>24°C</td>
<td>0.0196</td>
<td>2.61</td>
<td>5191</td>
<td>2013</td>
</tr>
<tr>
<td>42°C</td>
<td>0.0200</td>
<td>2.57</td>
<td>5082</td>
<td>1945</td>
</tr>
<tr>
<td>60°C</td>
<td>0.0213</td>
<td>2.60</td>
<td>4596</td>
<td>1822</td>
</tr>
<tr>
<td>80°C</td>
<td>0.0262</td>
<td>2.64</td>
<td>5015</td>
<td>1758</td>
</tr>
</tbody>
</table>

In addition to relocating the position of the sample in the resonant cavity, in order to acquire more accurate and reliable values of $\varepsilon'$ and $\varepsilon''$, the Q factor had to be manipulated. The value of the Q factor evaluated from any one of the four
methods mentioned should not be taken for granted and used as the only value in the calculation of the dielectric constant and dielectric loss. Instead, an averaged Q factor value was used.

Taking an air-filled WR159 at 24°C as an example, the values of Q factor evaluated through different methods are given in Table 9.14. It was found that the Q factor obtained by the $\pm 90^\circ$ method deviated much from the rest and should be discarded, and the averaged Q factor, $Q_o = \frac{(5008 + 5307 + 5258)}{3} = 5191$. Similarly, a LDPE/GF (33%)-filled WR159 at 24°C was again taken as an example and the sample laid on the floor of the cavity. The values of the Q factor are given in Table 9.15. In this case, the phase slope method gave the most unsatisfactory Q factor values and was discarded; the averaged Q factor $Q_s = 2013$.

Experience gained through the above mentioned analysis and synthesis of results showed that in many cases neither the phase slope method nor the $\pm 90^\circ$ method could give satisfactory results and they are therefore not recommended for use in the calculation of the Q factor (Ku et al, 1999b; 2000c). On the other hand, the magnitude method always generated reasonable Q factor values and was preferred.

<table>
<thead>
<tr>
<th>Method</th>
<th>Qo factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phase Slope</td>
<td>5008</td>
</tr>
<tr>
<td>Magnitude (-10 dB)</td>
<td>5307</td>
</tr>
<tr>
<td>Magnitude (-7 dB)</td>
<td>5258</td>
</tr>
<tr>
<td>$\pm 90^\circ$</td>
<td>4580</td>
</tr>
</tbody>
</table>

Looking at Tables 9.9 through 9.12, it can be deduced that the values of dielectric loss of LDPE/GF (33%) had two important characteristics. First, the values increased with the rise in temperature, but this was not very significant. Second, the values at lower frequencies, i.e. between 2.2 GHz to 4.9 GHz increased.
9. Results and Discussion

<table>
<thead>
<tr>
<th>Method</th>
<th>Qs factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phase Turning Points</td>
<td>2009</td>
</tr>
<tr>
<td>Magnitude (-4.3 dB)</td>
<td>2022</td>
</tr>
<tr>
<td>Magnitude (-2.6 dB)</td>
<td>2009</td>
</tr>
<tr>
<td>Phase Slope</td>
<td>1884</td>
</tr>
</tbody>
</table>

from 0.00606 to 0.00809 at room temperature but they rose to 0.0209 to 0.0234 at higher frequencies, ie 4.9 GHz to 12.5 GHz at 24°C. This implied that the composite was better processed by microwaves at higher frequencies as the value of the dielectric loss at 12.5 GHz was three times as much as that at 2.2 GHz. At the same time, it was also possible to confirm that the values of the dielectric constant of LDPE/GF (33%) ranging from 2.61 at 24°C to 2.64 at 80°C were correct and accurate. From the abnormal results of dielectric constant in Tables 9.11-9.12, it could be argued that the thickness of the sample, 4 mm, used was too large as it occupied too large a volume in the cavity (Ku et al, 1999b; 2000c; NRC, 1994). On top of it, laying the sample at the bottom of the cavity would not only give accurate results on the dielectric loss but also those on dielectric constant. In conclusion, the values of the dielectric loss of low loss thermoplastic composites, eg LDPE/GF (33%) and glass fibre reinforced (33%) polystyrene [PS/GF (33%)] were easily measured using the resonant cavity method provided the volume of the sample was not too large as compared with the volume of the cavity, and the sample was preferably laid at the bottom of the cavity. In addition, averaged Q factor values should be used in the calculation of $\varepsilon'$ and $\varepsilon''$. In summary, the original contributions of this section included measurement of the dielectric loss of low loss thermoplastic composites, eg LDPE/GF (33%) over a range of frequencies and at elevated temperatures, using the resonant cavity method, and discovering that in calculating the dielectric loss of a material, the averaged Q factors should be used.

9.6 Best Frequency Range to Process Materials Using VFM
After comparing and combining the results of characterisation in the frequency range from 2 GHz to 8 GHz using VW 1500, and of 6.5 GHz to 18 GHz using Microcure 2100, the best frequency range to process the five thermoplastic matrix composite materials was found to be in the frequency range from 2 GHz to 18 GHz. They are given in Table 9.16.

Table 9.16: Best Frequency Ranges to Process the 5 Materials in the Frequency Range of 2 GHz to 18 GHz.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Best processed Frequency Range (GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PS/CF(33%)</td>
<td>8.0 - 9.3 and 10.8 - 12.8</td>
</tr>
<tr>
<td>PS/GF(33%)</td>
<td>8.5 - 9.0 and 10.0 - 12.0</td>
</tr>
<tr>
<td>LDPE/GF(33%)</td>
<td>9.0 - 12.5</td>
</tr>
<tr>
<td>Nylon 66/GF(33%)</td>
<td>8.3 - 9.0 and 10.8 - 12.0</td>
</tr>
<tr>
<td>LDPE/CF(33%)</td>
<td>8.5 - 9.0 and 10.7 - 12.0</td>
</tr>
</tbody>
</table>
9. Results and Discussion

9.7 Results from Bond Strength Tests

This section describes the results obtained from bond strength tests from each material mentioned in section 2.8. Each section contains two parts of results; the first part consists of results obtained from the fixed frequency microwave facilities configuration and the second part comprises results obtained from variable frequency microwave (VFM) facilities. In the fixed frequency microwave facilities configuration, as described in sections 5.1 and 5.2, the input power to the system was a step function and could only be 240W, 400W, 640W and 800W. The duration of exposure could be increased in steps of 1 second. The change of temperature during the welding process was not measured. In the VFM, the input power level could be varied in steps of 10 W, starting from 50 W to 250W. The sweep time was very fast, eg 0.1s. During cavity characterisation, the actual amount of power that passed through the test pieces with respect to time was measured. In addition, the power reflected back from the material was also detected. However, the above capabilities were not available during normal bonding procedures. The change of temperature during the welding process was also recorded. The following sub-sections were then set aside for comparison, comments and discussion as well as chapter concluding remarks.

9.7.1 Glass Fibre Reinforced Polystyrene, PS/GF (33%)

The first part of this section describes the results obtained from bond strength tests of the above material joined using the fixed frequency microwave facilities configuration. The primer used was a five-minute two-part adhesive containing 100% liquid epoxy and 8% amine, which was microwave reactive (National Research Centre, 1994). Simple lap joints were selected for the connection of the two half test pieces. The lapped area was 20 mm x 10 mm. The maximum linear overlap of the test pieces was 20 mm with which the increase in overlap length would bring about a linear increase (Everett, 1995; Bolger, 1983) in the shear strength. The lapped areas were first roughened by rubbing them against coarse, grade 80, emery paper. They were then cleaned by immersing them in
methanol and allowed to dry in air before applying the primer onto them. After applying the filler, the two pieces were tightened by a rubber band which encircled the lapped areas four times as was depicted in Figure 3.1 in section 3.1. This was to fix the relative position of the two test pieces and to apply pressure onto the lap joint. The pressure on the lap joint was critical as the bond strength of the test pieces cured by leaving them in ambient conditions for 16 hours (Selleys, undated) with and without the rubber band were 611 N and 335 N respectively (Ku et al, 1997a; 1997b). In other words, the shear stresses were 3.055 N/mm² and 1.675 N/mm² respectively. Even with the applied pressure, the bond strength obtained for GF reinforced thermoplastic/GF reinforced thermoplastic was low as compared with that procured by the best metal/metal bond (Everett, 1995; Ku et al, 1997a; 1997b). The former was only 21.8% of the latter.

After tightening, the two halves of the test piece were positioned in the slot across the waveguide similar to the situation illustrated in Figure 3.1 in section 3.1, except from the fact that they stood there with no clamping. The test pieces were then exposed to two different power levels of either 400W or 800W with varying microwave exposure time. In all cases, only the parts smeared with filler were warmed or heated depending on the power level used and the time of microwave exposure. The test pieces were allowed to cool to room temperature before being shear tested to obtain maximum bond strength measurements. Results are summarised in Figure 9.17.

With reference to Figure 9.17, it was found that with 400 W power level, the peak bond strength was achieved by exposing the test pieces to microwaves for 2 minutes. The bond strength (651N) and hence the shear stress (3.255N/mm²) at this exposure duration exceeded that obtained by ambient conditions curing by 17%, but the time required was a mere of 0.2% of its counterpart (Ku et al, 1997a; 1997b; 1999a). For exposure times of one and a half to four and a half minutes, the shear stresses obtained using the microwave-cured filler were higher than those obtained by allowing the adhesive to set under ambient conditions.
With the power level of 800 W, the maximum bond strength (661N) and hence the maximum shear stress (3.305 N/mm²) were achieved when the exposure time was 45 seconds and it exceeded the ambient conditions cured bond strength by 19 %, but the time required was only 0.08% of its rival (Ku et al, 1997a; 1997b). The lower bond 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, could be due to over-curing of the adhesive. A PS/GF (33%) test piece was selected as an example and was exposed to 800 W power level for a period of 45 seconds. Viewing the sides of the test piece with the naked eye showed that outgassing had taken place and white ‘bubbles’ resulting from the release of dissolved air at elevated temperature were found. The white bubbles contained the primer and its colour implied that the filler was cured normally. The release of dissolved gases meant that the contact angle was decreased (Chawla, 1987) and the wettability of the filler on the composite was improved. This resulted in improved bonding quality and hence bond strength. Another test piece of PS/GF (33%) was exposed to 800 W power level for a period of 75 seconds and the sides of the test piece were viewed with the naked eye. It was found that the adhesive was charred, puffed up, leaving behind a yellowish resin mass. The
NOTE

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yellowish colour implied that the primer was over-cured (Ku et al, 1997a). A similar phenomenon has been described in the literature (National Research Council, 1994) with a one-component epoxy adhesive, dicyandiamide.

At some duration of microwave exposure, the bond strengths could be higher than those cured conventionally because the parent material could have melted and diffused into the primer or interface. This was reflected in the softening of the lapped area after it was just removed from the applicator (Ku et al, 2000b). The Heat affected zone (HAZ) seemed to be confined to the lapped area, which could be bent by hand easily, and outside of which the warming and heating effect of microwave energy could not be felt. Shear tests revealed that sixty percent of the failures were due to failure of the adhesive at the joint interface. The remaining failures took place at the original materials, PS/GF (33%). This meant that the adhesive bond strength exceeded the ultimate strength of the adherend and this case was more common in thin test pieces (Bolger, 1983). On the other hand, the thickness of test pieces used in this research was not too thin so it could be argued that the microwave energy played some role in increasing the shear strength of the material. With a power level of 400 W and exposure time of 2 minutes, the highest bond strength achieved was 720 N; while with a power level of 800 W and exposure time of 45 seconds, the peak bond strength obtained was 905 N. It could 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.

Figure 9.18 shows the apparent bond strength of PS/GF (33%) bonded with two-part five-minute Araldite as primer, using VFM (Ku et al, 2000b). The word apparent is used because all test pieces failed at the parent material and not at the bondline. This meant that the bondline was stronger than the weakened parent material. With the use of VFM, the machine was intentionally selected to operate at a fixed frequency of 2.5 GHz to enable comparison with other materials with ease. Other parameters for the processing were as mentioned in section 8.6.2. It
was found that the apparent peak bond strength, 532 N, appeared at an exposure time of 480 seconds. Values for other exposure times were just above 500 N. These values including the peak were lower than those obtained by the fixed frequency facilities. 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 procured with a power level of 800 W. It was marginally higher (3.5%) than its ambient cured rival (Ku et al., 1997b). In addition, the exposure times in VFM were much longer than its counterpart. At peak bond strength, the exposure time using VFM 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 was limited to 200 W. Even if the maximum power output of 250 W were employed, the results would not improve significantly. The low power output also 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 532 N. The weakness in the parent material might be due to its long exposure to microwave irradiation. In addition to this, in VFM the whole test pieces were exposed to microwave energy and not the joint and its adjacent areas only.
9.7.2 Glass Fibre Reinforced Nylon 66, Nylon 66/GF (33%)

With glass fibre reinforced nylon 66, the peak bond strengths obtained at exposure times of 35 and 55 seconds for the power levels of 400 W and 240 W respectively are depicted in Figure 9.19. They were 32% and 28% respectively higher than those obtained by curing the adhesive at room temperature conditions but the times required were only 0.06% and 0.1% of their counterparts. This material together with the adhesive seemed to couple with microwaves better than PS/GF (33%). Owing to the above fact, any excess filler that spilled over the sides and opposite faces of the interfaces had to be totally removed as the primer facing the microwave energy directly could bring about thermal runaway and the parent material could burn, depending on the degree of spill-over of the adhesive (Ku et al, 1997a; 1997b).

![Figure 9.19: Bond Strengths of Nylon 66/GF (33%) and Five Minute Two Part Adhesive](image)

With reference to Figure 9.19 and at a power level of 400 W, an exposure time to microwaves of over 42 seconds burned the test pieces even without spilling of the filler over the sides and the lapped area could also be easily bent with an
exposure time of 30 seconds or over. For a power level of 240 W, burning of test pieces occurred at an exposure time of over 62 seconds and the lapped area was also bent with ease when exposed to microwave energy of 55 seconds or over. When exposed to 65 seconds, the test pieces burned mildly and diffusion of parent material into the filler became more prominent (Ku et al, 1997a; 1997b; 1999e). This brought about higher bond strength than the 62 seconds of exposure. The bond strengths at an exposure time of 70 seconds were even higher than those exposed to 65 seconds. However, the test pieces were more seriously burnt which weakened the parent material and the bond quality was bad.

Figure 9.20 illustrates the bond strength of nylon 66/GF (33%) against exposure time. A variable frequency sweep was selected for the material with a centre sweep frequency of 7.25 GHz, and a sweep bandwidth of 1.5 GHz. The reason for the choice was explained in details in section 8.6.1. This centre sweep frequency and its sweep bandwidth were also best to process the primer, rapid araldite. An output power of 200 W was selected. Other parameters were unchanged and as mentioned in section 8.6.3. At most of the exposure duration, the bond strengths of the test pieces were above 1000N or shear strength of over 5 N/mm². All test pieces failed at the bondline (Ku et al, 2000b). At an exposure time of 35 seconds, the bond strength of the test piece bonded by VFM was around 1005 N, which was 1.5% higher than that obtained using the fixed frequency facilities and a power level of 400 W. Similarly, at an exposure time of 55 seconds, the bond strength obtained from VFM was 1050 N, which was 9% higher than that procured from its rival operating at 240 W. Figure 9.22 shows that, within limits, the longer the time of exposure to microwave energy, the higher would be the bondline strength of the material. At an exposure time of 100 seconds, the bond strength was 1305 N. Since the tensile strength of nylon 66/GF (33%) was 172.17 N/mm² (Ku et al, 1997b), there was plenty of room for improving its tensile shear strength without weakening the parent material.
9. Results and Discussion

Figure 9.20: Bond Strengths of Nylon 66/GF (33%) with Araldite and at Variable Frequency

9.7.3 Glass Fibre Reinforced Low Density Polyethylene, LDPE/GF (33%)

Referring to Figure 9.21 and at a power level of 800 W, the cluster of bond strengths at microwave exposure times ranging from 25 to 40 seconds, were best represented by the line 800PE1, ie their average value; while those resulting from microwave energy exposure were in the range of 45 to 65 seconds using the average line 800PE2. In both cases, the results obtained were similar to the work of other researcher (Siores and Groombridge, 1998) where high-density polyethylene (HDPE) was joined by microwave energy using similar facilities. A step function was formed using a shorter exposure times and the average bond strength obtained was only 97% of that cured in ambient conditions. It was observed that no diffusion of parent material to the primer had taken place. When longer exposure times were used, the average bond strength became 41% higher. The processing times were also merely 0.06% and 0.1% respectively of the conventional ones. At the power level of 400 W, the cluster of bond strengths obtained by exposing to microwaves from 135 to 240 seconds are best represented by the line 400PE1, their average value, was 18% higher than that cured in ambient conditions and the processing time was only 0.33% of its rival. The average bond strength of test pieces cured conventionally was relatively low,
9. Results and Discussion

Figure 9.21: Bond Strengths of LDPE/GF (33%) and Five Minute Two Part Adhesive

311 N, as stated by the manufacturer’s instructions for the adhesive (Selleys, undated).

LDPE/GF (33%) test pieces were also joined using VFM. The centre sweep frequency was the same as for nylon 66/GF (33%) because of the same reasons explained in section 8.6.1, but the sweep bandwidth was arbitrarily chosen as 1.1 GHz (Lambda Technologies Inc., 1998). Other parameters were as mentioned in section 8.6.1. With VFM, no bond was formed when the processing time was less than 150 seconds. Bonds started to form at an exposure time of 180 seconds. At an exposure time of 450 seconds, or over, the parent material was weakened because when it was subjected to tensile shear stress test the failure occurred at the parent material. Figure 9.22 shows that the bond strengths had the same characteristics as in the case of the fixed frequency facilities, ie the bond strength would not improve much with increasing duration of microwave irradiation (Ku et al, 2000d) with the average bond strength being only 378 N. This meant that its strength was only 21.5% higher than those cured under ambient conditions. Most of the failures took place at the bondline. Its strength was very near to that represented by line 400PE1 (367N) in Figure 9.21 but was 16 % lower than that
9. Results and Discussion

Figure 9.22: Bond Strength of LDPE/GF (33%) with Araldite and at Variable Frequency.

represented by line 800PE2. In addition, VFM needed longer time to weld the material. This was again due to the low power output of 200W.

9.7.4 Carbon Fibre Reinforced Polystyrene PS/CF(33%)

Test pieces of PS/CF (33%) were joined (Leong, 1998) using the fixed frequency facilities and without using primer. Pressure was still applied to test pieces as in the previous cases. Figure 9.23 illustrates the bond strength of PS/CF (33%) against time of exposure. Considering the graph at the power level of 240 W, the peak bond strength was 342 N and its time of exposure to microwave irradiation was 15 seconds (Lee, 1998). The one exposed to 20 seconds was discarded because arcing of carbon fibre took place and the test pieces were deformed thus giving poor weld quality. All test pieces broke at the bond line except the one exposed to 20 seconds of microwave energy, which failed at the parent material. At the power level of 400 W, the first four samples failed at the bondline and the remaining three test pieces were broken at the parent material. The joint quality of the last sample was poor because the material deformed as a result of graphite arcing. The peak bond strength was 444 N. With a power level of 640 W, the carbon fibre arced in a very short time, 7 seconds. Bond strengths with shorter
Results and Discussion

Figure 9.23: Bond Strengths of PS/CF (33%) with No Filler

time of joining were lower than those obtained from the 400 W power level. From the observations of the two graphs, it could be argued that a power level of 400 W best joined the material and higher bond strengths were achieved (Lee, 1998).

The time taken to obtain those bond strengths was also shorter. The addition of Araldite to the joined surfaces most definitely improved the bond strengths.

The second half of this section described the joining of PS/CF (33%) using VFM. The centre sweep frequency for it was 7.25 GHz and its sweep bandwidth was 1.5 GHz. The power output was 100 W. Other parameters are found in section 8.6.3. Figure 9.24 shows the bond strengths obtained for PS/CF (33%) versus time of exposure. In tensile shear tests, all samples failed at the parent materials but their values were low as compared to the tensile strength of the parent material (Ku et al, 2000d).
The load required to break the parent material was 1108 N. Since the strength of the processed material was significantly reduced, it appeared that the main reason for its weakness was due to excessive exposure to microwave irradiation. It could therefore be argued that within limits, a better strength of the joined material was achieved by reducing the time of its exposure to microwave energy.

The bond strengths of PS/CF (33%) versus power levels at 100 seconds of exposure time using VFM are shown in Figure 9.25. Under tensile shear tests, all failures took place at parent material. Their values were also very low as compared to the strength of the parent material. It seemed that the high level and long duration of microwave irradiation damaged the parent material. Since 50 W was the minimum power that VFM could launch into the cavity, a better strength of the joined material could only be achieved by reducing the duration of exposure time. However, there was also a limit to this, because a too short duration of exposure would result in incomplete joining and weak joint strength.

Figure 9.24: Bond Strengths of PS/CF (33%) with No Primer and at Variable Frequency
9. Results and Discussion

9.7.5 Carbon Fibre Reinforced Low Density Polyethylene (LDPE/CF)

With reference to Figure 9.26 (Liu, 1999) and the power level of 240 W, it was found that the peak bond strength of LDPE/CF (33%) was 299 N and was obtained at 10 seconds of exposure. Failure occurred at the parent material. The strength was very low as compared with the strength of parent material, 878 N. Considering the graph with the power level of 400 W, it was again found that the peak bond strength was 257 N and the time of exposure was 7 seconds. Again, the strength of the parent material was weakened by the microwave energy.

Now consider joining of LDPE/CF (33%) using VFM. A fixed frequency of 2.5 GHz was intentionally chosen to make comparison with similar material easier. Looking at Figure 9.27, it can be deduced that the peak bond strength is 432 N at an exposure time of 18 seconds. Failures for the first two points were at the bondline and failures for the last two points were at the parent material. In the first two cases, incomplete bonding gave rise to weak bonds (Liu, 1999; Ku et al, 2000d). The parent material was weakened by excessive exposure of microwave energy. When the exposure times were made above 20 seconds, arcing of graphite took place and the test pieces were deformed.
9. Results and Discussion

Figure 9.26: Bond Strengths of LDPE/CF (33%) with No Primer

Figure 9.27: Bond Strengths of LDPE/CF (33%) with No Primer and at 2.5 GHz
9.7.6 Effects of Different Processes on Bond Strengths

To enable readers to grasp the main points of the effects of different processes and parameters on bond strengths from sections 9.6.1 to 9.6.5, all the relevant information is summarised and tabulated in Table 9.17.

Table 9.17: Effects on Bond Strength by Different Processes and Parameters

<table>
<thead>
<tr>
<th>Materials</th>
<th>Facilities</th>
<th>Frequency (GHz)</th>
<th>Power (W)</th>
<th>Exposure Time (s)</th>
<th>Maximum Bond Strength (N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PS/GF (33%)</td>
<td>FF*</td>
<td>2.45</td>
<td>400</td>
<td>60</td>
<td>651</td>
</tr>
<tr>
<td></td>
<td>FF</td>
<td>2.45</td>
<td>800</td>
<td>45</td>
<td>661</td>
</tr>
<tr>
<td></td>
<td>VF#</td>
<td>2.50</td>
<td>150</td>
<td>480</td>
<td>532</td>
</tr>
<tr>
<td>PS/GF (33%)</td>
<td>VF</td>
<td>2.50</td>
<td>150</td>
<td>480</td>
<td>532</td>
</tr>
<tr>
<td>Nylon 66/GF (33%)</td>
<td>FF</td>
<td>2.45</td>
<td>240</td>
<td>55</td>
<td>964</td>
</tr>
<tr>
<td></td>
<td>FF</td>
<td>2.45</td>
<td>400</td>
<td>35</td>
<td>989</td>
</tr>
<tr>
<td></td>
<td>VF</td>
<td>7.25 ± 1.50</td>
<td>200</td>
<td>100</td>
<td>1305</td>
</tr>
<tr>
<td>LDPE/GF (33%)</td>
<td>FF</td>
<td>2.45</td>
<td>240</td>
<td>55</td>
<td>964</td>
</tr>
<tr>
<td></td>
<td>FF</td>
<td>2.45</td>
<td>400</td>
<td>35</td>
<td>989</td>
</tr>
<tr>
<td></td>
<td>VF</td>
<td>7.25 ± 1.10</td>
<td>200</td>
<td>100</td>
<td>1305</td>
</tr>
<tr>
<td>PS/CF (33%)</td>
<td>FF</td>
<td>2.45</td>
<td>240</td>
<td>55</td>
<td>964</td>
</tr>
<tr>
<td></td>
<td>FF</td>
<td>2.45</td>
<td>400</td>
<td>35</td>
<td>989</td>
</tr>
<tr>
<td></td>
<td>VF</td>
<td>7.25 ± 1.50</td>
<td>60</td>
<td>100</td>
<td>512</td>
</tr>
<tr>
<td>LDPE/CF (33%)</td>
<td>FF</td>
<td>2.45</td>
<td>240</td>
<td>55</td>
<td>964</td>
</tr>
<tr>
<td></td>
<td>FF</td>
<td>2.45</td>
<td>400</td>
<td>35</td>
<td>989</td>
</tr>
<tr>
<td></td>
<td>VF</td>
<td>7.25 ± 1.50</td>
<td>60</td>
<td>100</td>
<td>512</td>
</tr>
</tbody>
</table>

* High-power energy-focused fixed frequency (FF) microwave facility
# Variable frequency (VF) microwave facility

9.7.7 Cross Comparison of Results

9.7.7.1 LDPE/GF (33%), PS/GF (33%) and Nylon 66/GF (33%) at 400 W

Consider the results of the above three materials obtained using the fixed frequency facilities. The bond strengths of the three types of materials, procured by exposing the test pieces to a power level of 400 W, for different times are depicted in Figure 9.28. It was found that the peak or average bond strengths
obtained for Nylon 66/ GF (33%), LDPE/GF (33%) and PS/GF (33%) were 32%, 18% and 17% respectively and higher than those cured in ambient conditions.

Assuming that the effects of the 33% glass-fibre reinforcement and primer on the loss tangent, \( \tan \delta \), and the complex permittivity, \( \varepsilon \), of the materials were the same for the three types of thermoplastics, the loss tangent and the complex permittivity of the thermoplastics could be used to explain the characteristics of the bond strengths of the materials as was depicted in Figure 9.28. The loss tangents (Michaeli, 1995) of nylon and polyethylene were 0.0119 and 0.00116 respectively, subject to variations due to the presence of impurities and their respective dielectric constants (Metaxas and Meredith, 1983) were 3.02 and 2.25 respectively. The loss tangent of polyethylene was also slightly higher than that of polystyrene. Nylon 66 having the highest values of loss tangent and dielectric constant was best coupled with microwaves and the shortest joining times giving rise to the highest increase in bond strength (Ku et al, 1997b; 2000d).
9.7.7.2 LDPE/GF (33%) and PS/GF (33%) at 800 W

Again, the bond strengths of the above two types of materials, obtained by exposing the test samples to a power level of 800 W, at fixed frequency, and at different times are shown in Figure 9.29. It was found that the results matched with the loss tangent values of the two thermoplastic composite materials. The adhesive was unsuitable for gluing polyethylene under ambient conditions (Selleys, undated). The relatively low bond strength obtained in ambient-curing condition with primer for joining LDPE/GF (33%) rendered that procured by microwave processing looked promising (Ku et al, 1997a; 2000d). The application of microwave energy in this particular case was very successful because it caused significant improvement in bond strength as a result of diffusion of the filler to the parent material (Ku et al, 1999e).

![Figure 9.29: Bond Strengths of Two Types Materials at 800 W](image-url)
9.7.7.3 LDPE/CF (33%) and PS/CF (33%)

The dielectric properties of PS and LDPE were not much different (von Hipple, 1954; Metaxas and Meredith, 1983; Ku et al, 1997a; 1997b). Since both materials had the same percentage of weight of carbon fibres as their reinforcements, it could be argued that PS/CF (33%) and LDPE/CF (33%) had the same dielectric properties as well. However, neither the bond strength of PS/CF (33%) processed at variable frequency, nor that of LDPE/CF (33%) processed at fixed frequency in VFM seemed to be satisfactory as compared to the strengths of their parent materials respectively. But improvement were made by adopting suitable processing parameters for joining the materials as outlined in the second half of section 9.7.4. Now consider bond quality brought about by processing using different facilities. Plate 5 shows a burn mark on the joint of LDPE/CF (33%) joined using VFM at the fixed frequency of 2.5 GHz. The burn mark must have resulted from the hot spots in the material. The joint of PS/CF (33%) processed using VFM was perfect as depicted in Plate 6. It could therefore be argued that by following the recommendations made in section 9.7.4, VFM could produce stronger bonds for the two materials in concern with excellent joint quality.

Another noteworthy point associated with processing of carbon fibre reinforced thermoplastic materials was that fixed frequency joining was not recommended as the carbon fibre arced quickly and could cause thermal runaway thus resulting in deformed or even burnt samples. Even when VFM is employed, the operator should note that the power used should be low and the time of exposure should not to be too long because microwave energy would harm the parent material.

9.7.7.4 Peak Bond Strengths and Fixed Frequency Joining

Analysing at Figures 9.17, 9.19, 9.21, 9.23, 9.25 and 9.28, it can be deduced that in each figure the peak bond strength could be clearly identified at a particular
9. Results and Discussion

processing power level and it seemed that this was a characteristics of fixed frequency joining. On the other hand, the VFM joined test pieces did not show such a characteristic.
NOTE

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9. Results and Discussion

Plate 5: LDPE/CF (33%) : Burn Mark in Test Piece as a Result of Fixed Frequency Joining

Plate 6: PS/CF (33%) : No Burn Mark in Test Piece as a Result of Variable Frequency Joining
9.8 Concluding Discussion

The research and development work in this thesis started with the concept of joining fibre reinforced thermoplastic matrix composite materials using microwave irradiation with and without primers. The microwave configuration facility was then designed and manufactured to give maximum microwave irradiation at the joints of the test pieces to be bonded. The joints, with Araldite as primer, were then tensile shear tested and it was found that joints with Araldite as a primer cured by microwave irradiation were stronger than those obtained by curing the primer at ambient conditions. This led to the idea that the Araldite might have diffused into the thermoplastic matrix. In order to prove this, two surface analysis procedures were carried out. One was microscopy inspection and the other was x-ray photoelectron spectroscopy. Both analyses confirmed that diffusion had taken place.

It was also found that the relative complex permittivity and loss tangent of the dielectrics used were important in determining whether they were suitable for microwave processing or not. This led to the measurements of the dielectric constant and dielectric loss of the five fibre reinforced thermoplastic matrix composite materials using the waveguide transmission and dielectric probe techniques. It was proved that the dielectric loss of the low loss thermoplastic matrix composite materials could not be accurately measured by the two methods and a resonant cavity method was then employed. This was very successful.

Variable Frequency Microwave (VFM) processing was then used to find the best frequency range to process the five materials in the frequency range of 2 – 18 GHz. VFM was also used to join the five materials with and without primers. In most cases, VFM facility proved to be superior in joining fibre reinforced thermoplastic matrix composite materials. However, the main problem with VFM facility was its low power output, which resulted in higher exposure time intervals needed to produce high quality bonds.
9. Results and Discussion

9.8.1 Relationship between the Microwaves and the Matrices

The next topic to be discussed is the relationship between the microwave energy joining processes of the fibre reinforced thermoplastic matrix composite materials and the structure of the matrices, the polymers. The basic unit of polymer is the molecule or, more accurately, the macromolecule, which is composed of thousands of atoms. The spine of these molecules is the carbon chain of atoms and there are three basic types of polymeric materials. These are linear, space network and elastomeric and are shown in Figure 9.30 polymers (Flinn and Trojan, 1990; Morgan, and Gallagher, 1992).

![Figure 9.30: Three Types of Basic Polymeric Materials](image)

The *linear structure* is a chain that is not a straight line but more like spaghetti (Flinn and Trojan, 1990). These molecules slide by each other upon heating and form the thermoplastic polymers. The *space network structure* is rigid and makes up the thermosetting polymers. The coiled structures exhibit elastic extension of as much as 1000% and are aptly termed elastomers.
The molecular chains of a polymer may be completely tangled up or can be arranged in an orderly manner. If the chains are tangled up, the polymeric materials are said to be amorphous, otherwise they are crystallised (Flinn and Trojan, 1990). Regular molecules without bulky side groups or branches show a strong tendency to crystallise. It should be emphasised that partial rather than complete crystallisation is obtained at best. One of the thermoplastic matrix used in this research is LDPE which has a degree of crystallinity of 60% (Bolton, 1988) and therefore couples better with microwave energy than HDPE (NRC, 1994).

In this research, thermoplastic materials, eg LDPE, PS and nylon 66, were used as the matrices of the FRTP composites because they are generally flexible and relatively soft and when heated they become softer and more flexible. Their behaviour is totally due to their linear structure, in which the chain of each linear polymer slides over each other. In the joining of LDPE/GF (33%) using microwave energy with araldite as primer, it was expected that the Araldite will absorb more microwave energy than the LDPE/GF (33%) and convert it into heat (Ku et al, 2000a). The heat initially heated up the primer and the heated primer then conducted the heat into the composite. The matrix of the composite is a thermoplastic resin, LDPE with linear structure and low glass transition temperature, \( T_g \), -120°C. The percentage of free volume is therefore high (Flinn and Trojan, 1990) and the chains of each linear polymer slide over each other easily with the limited supply of conducted heat. This was proved by the microscopy study (Ku et al, 1999e).

Thirty three percent glass fibre reinforced low density polyethylene was joined using microwave energy with araldite as primer at a power level of 800W, frequency of 2.45 GHz and exposure times of 70 and 80 seconds. First, the sample with an exposure of 70 seconds to microwave irradiation was studied under Scanning Electron Microscopy (SEM). At 100 times magnification, it was found that the bondline was observed to have black colour, while the parent material in the matrix was grey. This is shown in Plate 7. There were shiny dots of glass fibre filaments that appeared as bright spots on the bondline. Such dots
were found to be numerous, as expected, in the parent material matrix. At 400 times magnification, it was found that the bondline occupied most of the view of the microscope. The glass fibre filaments appeared as bright patches in the bondline. The structure of the parent material adjacent to the bondline was the same as that which was observed away from the bondline. This is shown in Plate 8. No change of phase was inferred. However, since glass fibre filaments were present in the bondline, it could be argued that they had been “flushed” due to flowing of the melting parent material, ie LDPE/GF (33%) into the araldite, primer. Therefore, a change in phase, from solid to liquid, of the parent materials just adjacent to the bondline was observed (Plate 8). X-ray Photoelectron Spectroscopy also showed the diffusion of the parent material into the araldite and vice-versa (Ku et al, 1999e), which in turn confirmed the argument that there was flow of parent material towards the bondline and also a change of phase in the parent material.

Secondly, the sample with an exposure time of 80 seconds was studied. At 100 times magnification, similar microscopic characteristics were observed and are shown in Plate 9. At 400 times magnification, in addition to the microscopic view as in the 70-second exposure time, it was found that the colour and structure of the parent material just adjacent to the bondline were different to that observed away from the bondline. This implied that a phase change of parent material had taken place. This is shown in Plate 10 and the change in phase and the rise in temperature of the parent material had changed the loss tangent of the material (Ku et al, 2000f). It was experimentally found that the higher the temperature the higher the value of the dielectric loss (Ku et al, 1998; 1999a; 2000a). As a result of larger amounts of energy absorbed and converted to heat, the whole test piece became warm and pliable at the end of the joining process. In conclusion, microwave irradiation of the primer and the parent material brought about hybrid heating by the combination of microwave energy induced and thermal conduction that took place along the interface. This hybrid heating subsequently caused a phase change in the matrix, which became more susceptible to microwaves and thus hot spots occurred at macrolevel. At 1000 times magnification, the glass fibre filaments on the bondline can be identified easier (Plate 11).
NOTE

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9. Results and Discussion

Plate 7: 100 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 70 seconds

Plate 8: 400 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 70 seconds
9. Results and Discussion

Plate 9: 100 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 80 seconds

Plate 10: 400 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 80 seconds
Plate 11: 1000 Times Magnification of Bondline with Joining Parameters of 2.45 GHz, 800 W and 80 seconds
Thirty three percent glass fibre reinforced low density polyethylene was then joined using variable frequency microwave (VFM) energy with araldite as primer at a power level of 200W, central frequency of 7.25 GHz and bandwidth of 1.1 GHz (Ku et al, 2000d), and exposure times of 420 and 450 seconds. First, the sample with an exposure of 420 seconds to VFM irradiation was studied. At 100 times magnification, it was found that the bondline was observed to have black colour, while the parent material in the matrix was grey. This is shown in Plate 12. Only very few glass fibre filaments appeared on the bondline. At 400 times magnification, some glass fibre filaments were found as bright patches on the bondline as depicted in Plate 13 (Ku et al, 2000f). The microscopic observations found were similar to those found in Plates 5 and 6 respectively except there were less glass fibre filaments on the bondline. From this it was inferred that diffusion of the parent material to the araldite and vice-versa was not so obvious as in the fixed frequency case. Four hundred and twenty seconds was the maximum exposure time of the composite to VFM before the parent material was damaged (Ku et al, 2000d). Next, the sample with an exposure time of 450 seconds to VFM energy was investigated. At, 100 and 400 times of magnifications, similar microscopic characteristics were observed as in the case of an exposure time of 420 seconds and are shown in Plates 14 and 15 respectively (Ku et al, 2000f). In both cases, no change of phase was inferred. However, since more glass fibre filaments were present in the bondline, it could be argued that they had been “flushed” due to flowing of the melting parent material, ie LDPE/GF (33%) into the araldite, primer. Therefore, a change in phase, from solid to liquid, of the parent materials just adjacent to the bondline was observed.

9.8.2 Relationship between the Microwaves and the Fibres

The last but one topic to be discussed is the relationship between the joining process of the FRTP composites by microwave energy and the reinforcing filaments, glass or carbon fibre filaments. The loss tangent of glass fibre filaments was found to be very low, 0.0002, (Metaxas and Meredith, 1983; von Hippel, 1995; Ku et al, 1999a; 1999b; 2000b) and it did not increase much with rising temperature. They will not be discussed in detail because energy absorbed
9. Results and Discussion

Plate 12: 100 Times Magnification of Bondline with Joining Parameters of VFM, 200 W and 420 seconds

Plate 13: 400 Times Magnification of Bondline with Joining Parameters of VFM, 200 W and 420 seconds
NOTE

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9. Results and Discussion

Plate 14: 100 Times Magnification of Bondline with Joining Parameters of VFM, 200 W and 450 seconds

Plate 15: 400 Times Magnification of Bondline with Joining Parameters of VFM, 200 W and 450 seconds
by it will not contribute much to the heating the composite. On the other hand, the dielectric loss of carbon fibre filaments was expected to be much higher than that of glass fibre filaments and its importance in the joining process was therefore investigated in detail. Two carbon fibre filaments of 6-mm length used in the manufacture of the FRTP composites were laid down on the test piece [of LDPE/GF (33%)] used in the research and positioned side by side as shown in Figure 9.31. They were held in position by smearing an extremely small amounts of glue of low loss tangent to one end of the filament. The separation distance of the two filaments, d mm, was varied from 1mm to 5 mm in steps of 1 mm. They were then placed in the waveguide front of the fixed frequency (2.45 GHz) microwave facility.

It was found that, irrespective of the distance of separation of the filaments, d mm, the carbon filaments started to arc after exposure to 800 W power level microwave irradiation for 15 seconds. When the exposure time was increased to 18 seconds or more, conduction of heat from the carbon filaments to the surrounding area took place quickly and the surface of the test piece was burnt and became black. Heat was also conducted to the other end of the test piece. When the exposure time was 16 or 17 seconds, the test piece was slightly blackened around the tips of the carbon fibre filaments and the radius of the blackened part was found to be from 0.5 to 1 mm. This is shown in Figure 9.32.

Referring to Figure 9.32 and considering the black patch. It can be concluded that the carbon filament heated the matrix of the composite, LDPE, to its melting point, 115°C and continued to supply heat until it became charred. This means that the bonds in the LDPE were broken down leaving carbon particles behind. The black patch proved by x-ray photoelectron spectroscopy to be carbon formed from the breaking of the C-C and C-H bonds of LDPE (Ku et al, 2000f).
Figure 9.31: Locations of Carbon Fibre Filaments on Test Piece

Figure 9.32: Locations of Black Marks
In the surface analysis using X-ray Photoelectron Spectroscopy, two samples of LDPE/GF (33%) were used. One is a sample of LDPE/GF (33%) with black patches on it and the other is a clean LDPE/GF (33%) sample used as control. Referring to Figure 9.33, it is found that the elements present in the sample of LDPE/GF (33%) with black patches are oxygen, carbon and silicon. This is similar to the elements found in the control sample as illustrated in Figure 9.34. The intensity of carbon in Figures 9.33 and 9.34 is fitted into the curves of Figures 9.35 and 9.36 respectively. Referring to Figure 9.36, it is found that there is only one peak for the intensity of carbon atoms, which represents carbon atoms in C-H and C-C bonds in LDPE/GF (33%). Referring to Figure 9.35, it is found that there are two peaks for the intensity of carbon atoms; one represents carbon atoms in C-H and C-C bonds in LDPE/GF (33%) and the other represents independent carbon atoms (graphite). This concluded that carbon atoms exist independently in the black patches (Ku et al, 2000f).
**9. Results and Discussion**

**HDPE TREATED**

![Graph showing the intensity of elements against binding energy in an HDPE treated sample.](image)

Figure 9.33: The Intensity of Elements against Binding Energy in LDPE/GF (33%) Sample with Black Patches.

**HDPE CONTROL**

![Graph showing the intensity of elements against binding energy in a control sample.](image)

Figure 9.34: The Intensity of Elements against Binding Energy in Control Sample.
9. Results and Discussion

Figure 9.35: The Two Peaks of Intensity for Carbon Atoms in the Sample with Black Patches.

Figure 9.36: The Peak of Intensity for Carbon Atoms in Control Sample.
9. Results and Discussion

9.8.3 Energy to Break Bonds

Referring to Figure 9.32, the diameter of the carbon fibre is 0.5 mm and its length is 6 mm. Its volume = \( \pi/4 \times (0.5)^2 \times 6 \ mm^3 = 1.178 \ mm^3 \) or \( 1.178 \times 10^{-3} \ cm^3 \).

Since the density of the carbon fibre is 1.86 g/cm\(^3\) (Matthews and Rawlings, 1994), the mass of the carbon fibre = volume \times\ density

\[ = 1.178 \times 10^{-3} \ cm^3 \times 1.86 \ g/cm^3 = 2.191 \times 10^{-3} \ g. \]

The specific heat capacity of the carbon fibre is 0.61 J/gK (Borgnakka and Wylen, 1998), and the energy absorbed by the carbon fibre by bringing it from room temperature (25°C) to 115°C

\[ = \text{mass} \times \text{specific heat} \times \text{change in temperature} \]

\[ = 2.191 \times 10^{-3} \times 0.61 \times (115-25) = 0.12 \ (J), \text{ which was the energy transferred to} \]

the thermoplastic matrix, assuming there is no loss to the surrounding.

Referring to Figure 9.32, the diameter of the patch is 0.5 mm and its depth is 0.005 mm. Its volume = \( \pi/4 \times (0.5)^2 \times 0.005 = 0.982 \times 10^{-3} \ mm^3 = 0.982 \times 10^{-6} \ cm^3 \). Its mass = density \times\ volume = 0.92 \times 0.982 \times 10^{-6} = 0.9 \times 10^{-6} \ g.

In a mer of LDPE, there are one C-C bond and 4 C-H bonds. The energy required to break one-gram mole of the C-C bond is 334.88 kJ and of the 4 C-H bond is \( 4 \times 443.716 \) kJ (Flinn and Trojan, 1990).

The energy required to break the C-C bond (black patch) in Figure 9.32

\[ = \frac{0.9 \times 10^{-6}}{28} \times 334.88 kJ = 0.011 \ J. \]

Similarly, the energy required to break the C-H bond (black patch)

\[ = 4 \times \frac{0.9 \times 10^{-6}}{28} \times 443.716 kJ = 0.057 \ J. \]

Total energy required to break all the bonds of the LDPE (charred area)

\[ = 0.011 \ J + 0.57 \ J = 0.068 \ J. \]
The energy transferred from the carbon fibre to the matrix of the composite was found to be 0.12 J which was enough to break the bonds of the LDPE and char it (Ku et al, 2000f).

In joining LDPE/CF (33%) with a fixed frequency of 2.45 GHz and at the power level of 400W, it was found that the material started to arc after being exposed to microwave irradiation for 8 seconds (Liu, 1998). Again, in joining PS/CF (33%) with a fixed frequency of 2.45 GHz and at the power level of 400 W, the composite started to arc after being exposed to microwave energy for 11 seconds (Lee, 1999). In short, the composite materials started to arc in less than 11 seconds when exposed to the power of 400 W. Comparing these results with those of the independent carbon fibre filaments’ study in this research, it was found that the composite materials coupled better with microwave irradiation. The reason behind this is that the filament-to-filament distance in the FRTP composites is much less than 1 mm. The filaments, once heated, conduct heat in their respective neighbourhood easily and in addition, in the closely packed FRTP composites (33% by weight of fibre filaments), some of the filaments may have touched one another which may have led to d.c. conduction, both of which will heat the composites up and cause arcing at a much lower power level and time of exposure. On account of the above reasons and when the strength of the composite can be compromised, it can be argued that the amount of carbon fibre filaments in the composites can be reduced to cut the cost of the materials, which at the same time can couple with microwave irradiation more efficiently.

9.8.4 Weakened Parent Materials by Excessive Microwave Irradiation

The last topic to be discussed concerns the weakened parent material mentioned in sections 9.7.4 and 9.7.5 in which two of the five FRTP composite materials studied, PS/CF (33%) and LDPE/CF (33%), were weakened by excessive exposure to VFM irradiation. The discussion is split into two parts; one part deals with the effect of VFM energy on the reinforcement of the composite, ie the carbon filaments, while the other is concerned with that in the matrix of the composite, ie the thermoplastic.
Considering the joining of PS/CF (33%) using VFM with no primer, the power used was 100 W, the maximum exposure time was 120 seconds and the maximum temperature reached was 95°C, which was higher than the heat-distortion temperature (82°C) of PS, (Flinn and Trojan, 1990) and the temperature limit in air for carbon filaments impregnated in polyethylene or polystyrene (93°C) (Budinski, 1989). The matrix and the reinforcement of the composite were damaged. The bonds of the matrix, PS, were broken, leaving behind black carbon as mentioned in the previous paragraphs. The tensile strength values obtained for PS/CF (33%) were lower than those procured for PS/GF (33%) by around 100 N (Ku et al, 2000d) and this provided an extra proof that the carbon fibre filaments were damaged by prolonged VFM irradiation because the carbon fibre filament (3100 MPa) is stronger than the glass fibre filament (2200 MPa) (Matthews, and Rawlings, 1994). During the joining process, the carbon fibre filament coupled very well with microwave energy and was heated directly which caused damage to occur. On the other hand, the glass fibre filament did not absorb microwave irradiation directly but it was heated by the Araldite through thermal conduction. In addition, the oxidation temperature of the carbon fibre filament (505°C) is also closer to the maximum processing temperature (95°C). As a result, the carbon fibre filament became brittle and was damaged (Ku et al, 2000f).

Considering the joining of LDPE/CF (33%) using VFM at fixed frequency of 2.5 GHz with no primer, the power used was 100 W, the maximum exposure time was 20 seconds and the maximum temperature reached was 95°C, which was higher than the heat-distortion temperature (49°C) of LDPE, (Flinn and Trojan, 1990) and the temperature limit in air for carbon filaments impregnated in polyethylene or polystyrene (93°C) (Budinski, 1989). The matrix and the reinforcement of the composite were damaged. The bonds of the matrix, LDPE, were broken, leaving behind black carbon as mentioned in the previous paragraphs. Since fixed frequency had been used in the processing, hot spots had also contributed to the damage of the matrix in spite of its short exposure to microwave irradiation. The peak tensile strength value obtained for LDPE/CF (33%) were lower than those procured for LDPE/GF (33%) by around 32 N (Ku
et al, 2000d) and this provided an extra proof that the carbon fibre filaments were damaged by prolonged VFM irradiation because the carbon fibre filament is stronger than the glass fibre filament as mentioned previously (Ku et al, 2000f). Other arguments are similar to the case of PS/CF (33%).
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10. Conclusions and Recommendations for Further Work

10.1 Conclusions

The research and development work has confirmed that high energy rate joining of thermoplastic composites using microwave energy is feasible (Ku et al, 1997a; 1997b; 1999e; 2000d). LDPE/GF (33%), PS/GF (33%) and Nylon 66/GF (33%) were successfully bonded with primer using fixed as well as variable frequency microwave facilities. At the same time, LDPE/CF (33%) and PS/CF (33%) had also been successfully joined without filler and using both fixed and variable frequency microwave irradiation. The bond strengths obtained by joining thermoplastic composites like PS/GF (33%) using microwave energy were found to be higher than those obtained by curing the primer under ambient conditions. X-ray photoelectron spectroscopy surface analysis proved that the Araldite used had diffused into the thermoplastic composite matrices (Ku et al, 1999e).

The values of $\varepsilon'$ and $\varepsilon''$ as well as those of $\tan \delta$ played an important role in the successful joining thermoplastic composites by microwave irradiation and laboratory techniques were developed to measure them. The waveguide transmission technique and dielectric probe methods were used to measure the $\varepsilon'$ and $\varepsilon''$ values of the thermoplastic composites (Ku et al, 1998; 1999a). For low loss thermoplastic composites like LDPE/GF (33%), a resonant cavity method had to be employed to measure its $\varepsilon''$, and hence $\tan \delta$ (Ku et al, 1999b; 1999d). Transmission-line method was useful because it showed relaxation behaviour and transition in permittivity with changes in frequency for the MUT (NRC, 1994). However, certain drawbacks were associated with this method. First, precise sample fit is critical, with air gaps causing significant errors. Also materials that are brittle or difficult to machine are especially troublesome. Second, elevated temperature measurements using waveguide transmission technique are very difficult (Tinga, 1992). The entire section of the transmission line containing the test specimen must be contained in an oven (Ku et al, 1998; 1998a). Differences in thermal expansion between the sample holder and the sample under test made it difficult to maintain precise sample fit as the temperature increased. Also, changes in electrical properties and dimensions of the transmission
lines with increasing temperature must be accounted for to maintain calibration reference (Batt et al, 1992). At low temperatures, calibration of the dielectric probe involved the use of short and open circuits and of deionised water as standards. A modified calibration procedure, which uses short and open circuits and an alumina sample of known properties as standard, was developed to carry out calibration at temperatures as high as 1200°C. Otherwise, the measurement procedure and the calculation approach remain the same (NRC, 1994). The design of an open-ended coaxial probe suitable for high temperature measurements requires the selection of a probe material that has a very low thermal coefficient of expansion. A resonant cavity method can be highly accurate and particularly advantageous in the determination of relatively small loss tangents. The basic assumption behind this method is that the sample must be very small compared with the cavity itself, so that a frequency shift that is small compared with the resonant frequency of the empty cavity is produced by the insertion of the sample (Ku et al, 1999d; 1999f). For the purposes of measuring dielectric properties at high temperatures, up to 1200°C, the experimental set-up required the sample to be held in an adequate holder that was heated in a conventional furnace and transferred into the cavity for test.

During this research project it was also discovered that there are best frequency ranges existing for microwave processing for the five thermoplastic composites and that VFM processing of composites is more advantageous over its fixed frequency counterpart. The quality of the bonds obtained was also more superior. It was discovered that the power level was vital in microwave processing of thermoplastic composite materials. The best combinations of input parameters, ie time of exposure and power level, for obtaining quality bond and high strength using microwave irradiation were also obtained.

10.2 Recommendations for Further Work

In the area of the fixed frequency facilities, some modifications to the initial test rig were essential in order to explore more processing information, eg temperature change, electric field strength around the sample, actual power entered and reflected from the sample. The modifications included the addition of two directional couplers,
an E-field connector and a thermopile to measure the incident and reflected power, the electric field strength and the temperature during joining. The modified microwave facilities configuration is shown in Figure 10.1.

![Figure 10.1: Modified Fixed Frequency Microwave Facilities Configuration](image)

As far as VFM is concerned, it is desirable to carry out more experiments with diversified input parameters. The main drawback for the current equipment range is the low power input. Modifications and improvements are therefore suggested to increase the maximum power level to 2kW and Microcure 2100 model 2000 is suitable for this. The productivity for the VFM facilities need to improve significantly in order to reduce the cycle time.
NOTE

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References:


Altman, J.L. (1964), Perturbation Formulas For Cavities and Waveguides, Microwave Circuits, Van Nostrand, pp. 409-16.


References


HP8410B Automating the HP 8410B Microwave Network Analyser (1980), Hewlett Packard Product Information, June, pp. 5 -10.
HP85070A Dielectric Probe Kit, 200 MHz to 20 GHz (1990), Hewlett Packard Product Technical Data Sheets, August.


HP Coaxial and Waveguide Catalogue and Microwave Measurement Handbook (1993.).

ICI Plastics, MSDS for GF reinforced (33%) nylon 66 (undated), ICI Plastics, pp. 1-5.


Kemcor Australia (undated), MSDS for LDPE, pp. 1–4.


Ku, H.S., Siores, E., Taube, A and Ball, J.A.R. (1999d), Productivity Improvements Through the Use of Industrial Microwave Technologies, Proceedings of the 26th International Conference on Computer and Industrial


Ku, H.S., MacRobert, M., Siores, E. and Ball, J.A.R. (2000c), Characterisation of Thermoplastic Matrix Composites (TPC) Using Variable Frequency Microwave (VFM), Plastics, Rubber and Composites (accepted for publication).

Ku, H.S., MacRobert, M., Siores, E. and Ball, J.A.R. (2000d), Variable Frequency Microwave Processing of Thermoplastic Composites, Plastics, Rubber and Composites (accepted for publication).


Ku, H S, Siores, E and Ball, J A R (2000f), Relationship between Microwave Irradiation and Constituents of Composites During Joining Process, Transactions, Group 2, The Hong Kong Institution of Engineers, pp. (submitted for publication)


Metaxas, A.C. and Meredith, R.J. (1983), Industrial Microwave Heating, Peter Peregrinus Ltd., pp. 5-6, 28-31, 43, 211, 217, 278, 284-5.


References


Selleys, (undated), Araldite five minute epoxy adhesive user instructions, p. 1, 1 Gow Street, Padstow, NSW 2211, Australia.


Siores, E. (1993), Applications of Industrial Microwave Technology in Materials Welding and Joining.


PUBLICATIONS


