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**Preparation of different types of carbon fiber reinforced
polymer by using electrodeposition resin molding method**

(電着樹脂成形法を用いた各種炭素繊維強化ポリマーの製造)



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Preparation of different types of carbon fiber reinforced polymer by using electrodeposition resin molding method



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*A thesis submitted in partial fulfillment of the requirements for the degree of Doctor of
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Abstract

Composite material is one of the key components for the enormous development in aircraft and automobile industries. The amazing features of composite materials made it a suitable alternative of steels. Carbon fiber reinforced polymer (CFRP) is one of the most widely used composite materials because of having excellent properties such as good corrosion resistance, high strength and stiffness, low density, and long fatigue life in the fiber direction. The growing interest of CFRP has led many researchers to improve the manufacturing methods of CFRP. This thesis presents a comprehensive study on the production and characterization of advanced carbon fiber reinforced polymer (CFRP) composites using the innovative and scalable Electrodeposition Resin Molding (EDRM) technology. In this technique, an electro-activated deposition solution served as the anode and the carbon fiber served as the cathode. Epoxy was chosen as the electro-activated deposition solution. The primary objective was to explore the potential of EDRM not only as an alternative to conventional composite manufacturing methods, but also as a platform for developing different types of CFRPs with enhanced mechanical and dynamic properties.

The first phase of the research focused on optimizing key EDRM processing parameters, including current intensity, voltage, and thermal curing conditions on the static and dynamic properties of composites. As for the dynamic properties, vibration test of the specimen had been carried out to obtain the natural frequencies and damping of the composites. In regard of the static properties, four-point bending test and tensile test were applied on the specimen to derive the bending strength and young modulus of the composites. Additionally, the effect of heat curing temperature and period on the composite properties is also investigated. Two different fiber orientations were used to prepare the preform. One was plain-weave [PW0°]₈ and another was [TFP/TFP/PW45°/PW0°]₈ which was prepared by using a sewing machine called Tailored Fiber Placement (TFP) machine. Three different voltage conditions (100V, 150V and 200V) and for each voltage condition, five different current conditions (0.50A, 0.75A, 1.00A, 1.25A and 1.50A) were applied. The effect of the voltage and current condition of EDRM was not that significant on the vibrational characteristics of composites, while it had some effect on the bending strength. However, the heat-curing temperature as well as the heat curing period had very noticeable effect on the bending strength of composites. Among the various conditions tested, composites cured at 230°C and processed under 1.5 A current

exhibited the highest flexural strength, highlighting the influence of heat and current on resin infiltration and interfacial bonding.

Three novel applications beyond conventional CFRP laminates were examined. Initially, cellulose nanofiber (CNF) sheets were used as dielectric layers inside the laminate to create CFRP-based structural capacitors. These composites' mechanical strength and detectable capacitance suggested that they would make good lightweight structural elements that store energy. Although the Young's modulus was somewhat compromised, it was discovered that the capacitance and bending characteristics of composites were significantly improved by the use of CNF as a dielectric. The bending strength of CFRP with CNF is around 6% more than that of CFRP without CNF. Additionally, using CNF into CFRP resulted in a maximum capacitance of 813 nF/m².

Next, the impact of curvilinear fiber architecture more especially, sine and parabola wave patterns on CFRP vibrational behavior was explored. Curved fiber routes considerably changed the dynamic response, according to modal analysis, and sine-shaped fibers offered the best damping and energy dissipation properties. The potential of fiber steering in adjusting composite vibration performance for dynamic applications is highlighted by these findings.

Finally, Nylon 6 hybrid composites were developed by embedding thermoplastic layers within the epoxy matrix. Toughness, bending strength, and impact resistance were all enhanced by this arrangement. The successful incorporation of Nylon 6 into the composite system and improved fiber-matrix adhesion were validated by SEM and Raman spectroscopy. All the results from the experimental testing showed that the carbon fiber/nylon 6/epoxy composite outperformed carbon fiber/epoxy composites in all mechanical tests conducted. The bending strength of CFRP increased almost double by adding nylon 6. Additionally, the CFRP including nylon 6 consumed 38% higher impact energy than normal CFRP.

Overall, this study shows that EDRM provides a flexible platform for creating high-performance, tailored composite materials in addition to being a practical production method for CFRPs. In aerospace, automotive, and multifunctional engineering applications where weight reduction, structural efficiency, and functional integration are crucial, the results provide insightful information for future CFRP component design.

DEDICATION

This dissertation is dedicated to my beloved mother, my siblings, and my teachers, who built me up to be capable of pursuing a doctoral degree.

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Table of Contents

<i>Abstract.....</i>	<i>ii</i>
<i>Acknowledgements.....</i>	<i>v</i>
<i>Table of Contents</i>	<i>vii</i>
<i>List of Figures.....</i>	<i>x</i>
<i>List of Tables</i>	<i>xiv</i>
1. Introduction.....	2
1.1 Background	2
1.2 Different types of composite manufacturing technique	7
1.2.1 Auto-clave molding method.....	8
1.2.2 De-autoclave molding method	9
1.2.3 Liquid resin molding method	12
1.2.3.1 Electrodeposition resin molding method	13
1.3 Objectives of this research.....	17
1.4 Significant of this research	18
1.5 Research Questions	19
1.6 Structure of thesis	19
2. Preparation of CFRP by altering manufacture variables of EDRM.....	23
2.1 Introduction	23
2.2 Methodology	24
2.2.1 Materials	24
2.2.2 Preparing the Preforms.....	24
2.2.3 Electrodeposition resin molding method.....	26
2.2.3 Heat Curing.....	29
2.2.4 Specimen Preparation	30
2.2.5 Vibration Test	31
2.2.6 Four-point bending test	32
2.2.7 Tensile test	34

2.3 Result and Discussion.....	38
2.3.1 Electrodeposition resin molding	38
2.3.2 Natural frequency and damping	39
2.3.3 Bending strength and bending modulus	45
2.3.4 Young modulus	49
2.3 Conclusion	50
2.4 Future Studies.....	51
3. Preparation Curvilinear CFRP and evaluate its vibrational response.....	55
3.1 Introduction	55
3.2 Methodology	57
3.2.1 Preparation of curvilinear preform	57
3.2.3 Vibration Test	64
3.2.4 FEM	65
3.3 Result and Discussion.....	66
3.3.1 Vibration Test	66
3.4 Conclusion	71
3.5 Future Scope	72
4. Preparation of CFRP capacitor by incorporating CNF	75
4.1 Introduction	75
4.2 Methodology	78
4.2.1 Material	78
4.2.2 Preparation of preform and dielectrics	79
4.2.3 Sample preparation	82
4.2.4 Capacitance Test	84
4.2.5 Four-point bending test	85
4.2.6 Tensile test	86
4.2.7 Scanning Electron Microscope (SEM).....	86
4.3 Result and Discussion.....	87
4.3.1 Capacitance test.....	87
4.2.2 Four-point bending test	90
4.2.3 Tensile test	92
4.2.5 SEM Morphology	93

4.4 Conclusion	95
4.5 Future Scope	96
5. Preparation of Nylon 6 based Carbon fiber/epoxy composite and evaluate their compatibility	99
5.1 Introduction	99
5.2 Methodology	102
5.2.1 Materials and method.....	102
5.2.2 Sample Preparation	102
5.2.3 Vibration Test	105
5.2.4 Charpy impact Test	106
5.2.5 Four-point bending test	107
5.2.6 Roughness test	108
5.2.7 Raman Spectroscopy	109
5.3 Result and Discussion.....	111
5.3.1 Vibration Test	111
5.3.2 Charpy Impact test	112
5.3.3 Four-point bending test	114
5.3.4 Roughness test	116
5.3.5 Raman Spectroscopy	119
5.3 Conclusion	124
5.5 Future Studies.....	124
6 Conclusion.....	127
6.1 Conclusion	127
6.2 Suggestions of Future research.....	131

List of Figures

<i>Fig. 1.1: Different types of composite manufacturing method</i>	<i>8</i>
<i>Fig. 1.2: Autoclave molding method [75].....</i>	<i>9</i>
<i>Fig. 1.3: Main manufacturing technologies for continuous fiber-reinforced composite structures: (a) Hand layup and automated layup with dry fibers or prepreg; (b) RTM technology including resin injection and preform curing process; (c) A preform waiting for curing; (d) Autoclave and OoA curing process; and (e) Additive manufacturing with in situ consolidation [73].....</i>	<i>11</i>
<i>Fig. 1.4: Autoclave and OoA molding technology. (a) Autoclave and the temperature and pressure monitoring/controlling system, (b) The largest autoclave in the world from Boeing Company in Chicago, USA [74]. 2012, ACS Process Systems), and the forward fuselage of Boeing 787 [74], (c) Schematic diagram of OoA; (d) Schematic diagram of the cross sections of composites molded by autoclave or OoA, (e) Curing deformation schematic diagram in autoclave molding process, and (f) The wing of MC-21 [75] and the fuselage skin of X-55, an advanced composite cargo aircraft, molding by OoA process [76]</i>	<i>11</i>
<i>Fig. 1.5: Different steps during liquid resin molding method [77]</i>	<i>13</i>
<i>Fig. 1.6: Schematic diagram of EDRM</i>	<i>15</i>
<i>Fig. 1.7: Actual set-up of EDRM</i>	<i>15</i>
<i>Fig. 1.8: Mechanism of EDRM where it shows the different stages of resin impregnation onto the fiber surface in the EDRM process.</i>	<i>16</i>
<i>Fig. 2.2: The left figure is the graphical representation of EDRM where the right figure is the actual set-up for the experiment. Carbon fiber acts as a cathode here and the steel mesh acts as an anode.</i>	<i>26</i>
<i>Fig. 2.3: Mechanism of electro-activated resin molding method [27]</i>	<i>27</i>
<i>Fig. 2.6: Cross-section of the specimen observed by SEM.....</i>	<i>30</i>
<i>Fig. 2.7: Specimen of (a) Vibration test, (b) Four-point bending test, (c) Tensile test</i>	<i>31</i>
<i>Fig. 2.8: Configuration of equipment for vibration test.</i>	<i>32</i>
<i>Fig. 2.9: Experimental setup of vibration test.</i>	<i>32</i>
<i>Fig. 2.19. Relationship of Voltage and current with time during the electrodeposition process</i>	<i>39</i>
<i>Fig. 2.20. Modal visualization of the composites</i>	<i>40</i>

<i>Fig. 2.21. Natural Frequencies of CFRP at different EDRM conditions</i>	40
<i>Fig. 2.22. Modal damping of CFRP at different EDRM conditions</i>	41
<i>Fig. 2.23: Natural Frequencies of CFRP at a) 100V 1.5A b) 150V 1.5A c) 200V 1.5A EDRM Condition (Different heat curing conditions)</i>	43
<i>Fig. 2.24: Modal Damping of CFRP at a) 100V 1.5A b) 150V 1.5A c) 200V 1.5A EDRM Condition (Different heat curing condition)</i>	44
<i>Figure 2.25: Natural frequencies of composites prepared by EDRM and VaRTM</i>	45
<i>Figure 2.26: Modal damping of composites prepared by EDRM and VaRTM</i>	45
<i>Fig. 2.27: Bending strength of CFRP on different EDRM conditions</i>	46
<i>Fig. 2.28: Bending modulus of CFRP on different EDRM conditions</i>	47
<i>Fig. 2.29: Bending strength of CFRP on different heat-curing conditions, (a) using temperature profile A, (b) using temperature profile B</i>	48
<i>Fig. 2.30: Bending modulus of CFRP on different heat curing conditions, (a) using temperature profile A, (b) using temperature profile B</i>	49
<i>Fig. 2.31: Young modulus of CFRP at different heat curing conditions</i>	50
<i>Fig. 3.1: (a) Plain-weave sheet, (b) Carbon fiber bundle.</i>	58
<i>Fig. 3.9: Sine shaped preform fabricated by TFP.</i>	62
<i>3.2.2 Preparation of the sample</i>	62
<i>Fig. 3.13: Modal visualization of mode 1 of parabola model a) from experiment b) from FEM</i>	67
<i>Fig. 3.15: Modal visualization of mode 3 of parabola model a) from experiment b) from FEM</i>	68
<i>Fig. 3.16: Modal visualization of mode 1 of sine model a) from experiment b) from FEM</i>	68
<i>Fig. 3.17: Modal visualization of mode 2 of sine model a) from experiment b) from FEM</i>	68
<i>Fig. 3.18: Modal visualization of mode 3 of sine model a) from experiment b) from FEM</i>	69
<i>Fig. 3.19: Natural Frequencies of a) unidirectional model b) parabola model c) sine model</i>	71
<i>Fig. 4.1: The left figure is a basic structure of a capacitor, and the right figure employed the basic structure for CFRP where the carbon fiber is the electrode and dielectrics are the</i>	

<i>insulator [110]. Two preforms containing four plain weave (PW) carbon fiber layers are on both sides of the dielectrics to make the structural capacitor.</i>	<i>78</i>
<i>Fig. 4.2: Different types of dielectrics that had been inserted into the CFRP for capacitor preparation</i>	<i>80</i>
<i>Fig. 4.3: Preparation of the mixture of Epoxy and CNF by adopting doctor blading method to have a uniform thickness throughout.</i>	<i>81</i>
<i>Fig. 4.4: Preparation of CNF film from the epoxy-CNF mixture by drying and curing</i>	<i>81</i>
<i>Fig. 4.5: The relationship of electrical current, resistance and voltage with time. The left axis shows the voltage values, whereas both the resistance and current were displayed by right axis. The green, orange and yellow lines represent the voltage, resistance and current respectively.</i>	<i>83</i>
<i>Fig. 4.6: The schematic diagram of the capacitance test that shows the specimen is held in between two copper plates, and the values are shown in a display connected by the electrodes.</i>	<i>84</i>
<i>Fig. 4.7: The actual diagram of the capacitance test that shows the specimen is held in between two copper plates, and the values are shown in a display connected by the electrodes.</i>	<i>85</i>
<i>Fig. 4.8: Impedance and capacitance of CFRP including CNF. The data of Sample A to sample B are shown here. The blue bar shows the impedance values whereas the brown bar portrays capacitance value.</i>	<i>88</i>
<i>Fig. 4.9: Impedance and capacitance of CFRP with different dielectrics. The yellow bar shows the capacitance value whereas the green bar portrays impedance values.</i>	<i>89</i>
<i>Fig. 4.10: Bending strength and Bending Modulus of CFRP including CNF. Samples C, E and F showed the capacitance; hence the bending data of these samples are presented.</i>	<i>91</i>
<i>Fig. 4.11: Bending strength and Bending modulus of CFRP including different dielectrics. The bending data of CFRP without any dielectrics are also presented here for comparison with CFRP with dielectrics.</i>	<i>91</i>
<i>Fig. 4.12: Image of CFRP cross-section including CNF film as a dielectric. The right figure the magnified version of the left figure that used x500 magnification.</i>	<i>94</i>
<i>Fig. 4.13: Image of CFRP cross-section CFRP including plastic as a dielectric. The magnified version of the left figure is given in the right figure that uses x100 magnification and x500 magnification.</i>	<i>95</i>

<i>Fig. 4.14: Image of CFRP cross-section CFRP taken by SEM; a) SEM cross-sectional image (x1500), b) SEM image after image processing with Image J.....</i>	<i>95</i>
<i>Fig. 5.1: Interaction of epoxy resin with polyamide [199].....</i>	<i>102</i>
<i>Fig. 5.2: Orientation of the preforms.....</i>	<i>104</i>
<i>Fig. 5.3: Hot-press machine</i>	<i>105</i>
<i>Fig. 5.4: Hot-pressed nylon 6</i>	<i>105</i>
<i>Fig. 5.5: Charpy Impact test machine</i>	<i>107</i>
<i>Fig. 5.6: Roughness testing machine</i>	<i>108</i>
<i>Fig. 5.7: Roughness testing machine</i>	<i>109</i>
<i>Fig. 5.8: Laser isotope machine</i>	<i>109</i>
<i>Fig. 5.9: (a) Raman Spectroscopy machine; (b) specimen under lens in Raman Spectroscopy machine.....</i>	<i>110</i>
<i>a) mode 1 of CFRP with Nylon 6 b) Mode 2 of CFRP with Nylon 6.....</i>	<i>111</i>
<i>a) mode 1 of CFRP without Nylon 6 b) Mode 2 of CFRP without Nylon 6.....</i>	<i>111</i>
<i>Fig. 5.10: Modal Visualization of CFRP.....</i>	<i>111</i>
<i>(a).....</i>	<i>112</i>
<i>Fig. 5.11: The vibration data of composites obtained from vibration test. (a) natural frequency; (b) modal damping.....</i>	<i>112</i>
<i>Fig. 5.12: Impact energy of specimens including and excluding nylon 6.....</i>	<i>114</i>
<i>Fig. 5.13: Bending strength of specimens including and excluding nylon 6</i>	<i>115</i>
<i>Fig. 5.14: Bending modulus of specimens including and excluding nylon 6.....</i>	<i>116</i>
<i>Fig. 5.15: Bending strength and modulus of specimen of specimens including and excluding of hot-pressed nylon 6.....</i>	<i>116</i>
<i>Fig. 5.16: Surface topography of specimen including and excluding nylon 6</i>	<i>117</i>
<i>Fig. 5.17: Roughness parameter Ra of specimens including and excluding nylon 6</i>	<i>118</i>
<i>Fig. 5.18: Roughness parameter Rz of specimens including and excluding nylon 6.....</i>	<i>119</i>
<i>Fig. 5.19: Raman spectroscopy of nylon 6</i>	<i>120</i>
<i>Fig. 5.20: Raman spectroscopy of specimen without nylon 6 and comparison with pure nylon 6.....</i>	<i>121</i>
<i>Fig. 5.21: Raman spectroscopy of specimen with nylon 6 and comparison with pure nylon 6</i>	<i>121</i>
<i>Fig. 5.22: Surface of specimen (a)without nylon 6 by Raman spectroscopy; (b) with nylon 6 by Raman spectroscopy.....</i>	<i>121</i>

<i>Fig. 5.23: SEM image of the cross-section of sample 1 after bending test.....</i>	<i>123</i>
<i>Fig. 5.24: SEM image of the cross-section of specimen with hot-pressed nylon 6 after bending test.....</i>	<i>123</i>

List of Tables

<i>Table 2.1. Code names for different heat curing condition</i>	<i>29</i>
<i>Table 2.2 Density of the specimen</i>	<i>38</i>
<i>Table 4.1. CNF application methods and sample descriptions.</i>	<i>81</i>
<i>Table 4.2. Dielectric properties of Different dielectrics.....</i>	<i>89</i>
<i>Table 4.3. Types of CNF application as dielectrics.....</i>	<i>92</i>
<i>Table 5.1. Experimental condition.....</i>	<i>104</i>

Chapter One

General Introduction

1. Introduction

1.1 Background

It is inevitable that the automotive and aviation industries have undergone a transformation in the coming ten years. One of the best aspects of this revolution was its structural growth. This occurred shortly after these industries began using composite materials primarily. In airplanes, the use of composites changed from secondary to load-bearing primary structures, and during the past few decades, there have been notable advancements in the study, design, and production of composites. The Boeing firm was the first to adopt composite materials as main components in its 787 model, which accounted for about half of the aircraft's weight. After Boeing, Airbus also used composite materials in their A350 XWB model [1,2]. The concept behind the creation of composite materials is to combine the desirable properties of the constituents in a way that is not possible with conventional materials. A composite material is a combination of two or more materials that are both physically and chemically different. Two basic components of composite material are the matrix and reinforcement [3]. Materials with different properties can be created by carefully choosing the type and amount of reinforcement, as well as its alignment and orientation in a polymer matrix. Because of these enhanced material properties and the capacity to adjust structural parameters, composite materials are an amazing substitute for conventional materials, which have been used in engineering for a very long time. Composites come in a wide variety of forms, from nanocomposites to bio-composites. These days, carbon fiber reinforced polymer, or CFRP, is the most widely used composite. Carbon fiber reinforced plastics (CFRP) are lightweight and have high specific strength and specific stiffness, which makes them widely used in a wide range of fields such as aerospace, automobiles, and sporting goods [4]. In the field of application such as the body structure of aircraft, spacecraft, and automobiles, as well as blades for wind power generation, they contribute greatly to the reduction of environmental impact by improving fuel efficiency and reducing carbon dioxide emissions, and demand for them is increasing year by year. These characteristics of CFRP are becoming increasingly important as material contributes to solving problems in today's sustainable society. It is projected that CFRP consumption will amount to \$25.2 billion worldwide [5]. Although CFRP was first primarily used in the aircraft industry [6], it is currently utilized in a wide range of industries. However, research was concentrated on enhancing CFRP's impact performance and delamination resistance, particularly in the

development of multifunctional and environmentally friendly composites [7–15]. CFRP is a good substitute for steel, but it also has drawbacks. It has a low tolerance for damage, is highly vulnerable to moisture, and requires a significant investment. Researchers made a number of attempts to enhance CFRP's qualities or reduce defects and deformation. Khalili et al. [16] and Soden et al. [17] evaluated the primary mechanical properties of a number of laminates under various loading conditions. Furthermore, some researchers evaluated CFRP and GFRP composites' impact and static indentation characteristics [18–21]. To account for thermo-viscoelastic effects on residual stress and deformation during the curing of flat composite laminates, Liu et al. [22] introduced an enhanced analytical model. Based on this model, the generation of residual stresses and deformation in unsymmetrical composite laminates was also analyzed. There has been extensive research focused on modeling the mechanical behavior of composite materials. For instance, Zhang et al. [23] developed a three-dimensional explicit finite element model to analyze the dynamic response of CFRPs exposed to X-ray radiation and to predict their performance under real service conditions in spacecraft applications. Moreover, Zhou et al. [24] employed a high-intensity ultrasonic liquid processor to uniformly disperse carbon nanofibers (CNFs) in SC-15 epoxy resin. Their study found that adding 2.0 wt% of CNFs led to the highest improvement in tensile strength. The newly formulated CFRP composites showed increases of 22.3% in flexural strength and 11% in tensile strength. To fabricate hybrid composites incorporating both micro and nano components, Shariatnia et al. [25] introduced a new processing approach. They utilized cellulose nanocrystals to assist in dispersing pristine carbon nanotubes (CNTs) into CFRP composites, without using surfactants or chemical functionalization. Their results indicated that the addition of 0.2 wt% CNTs and 0.2 wt% cellulose nanocrystals improved the flexural strength and interlaminar shear strength by 33% and 35%, respectively, when compared to standard CFRP specimens. In the aerospace industry, aluminum alloys are widely used for structural applications [26,27]. However, CFRP composites offer damping capabilities that are up to 100 times greater and a stiffness-to-weight ratio up to five times higher than aluminum alloys, depending on the type of carbon fiber used [28]. Hence, it is not surprising that CFRPs have gradually started to replace traditional thick metallic materials, particularly in applications prone to vibration. The vibrational properties of hybrid CFRP composites are largely influenced by the sequence of laminate stacking and the orientation of the fibers [29–31].

Numerous studies have been conducted to investigate the vibration behavior of CFRPs. Selim et al. [32] examined the vibration characteristics of CFRP tubes filled with shear thickening

fluids, while Fischer et al. [33] explored the dynamic response of sandwich structures filled with similar fluids. Badie et al. [34] investigated the influence of fiber orientation and stacking sequence on the natural frequency of composite laminates. Honda and Narita [35] developed an analytical method to calculate the natural frequencies and vibration modes of laminated plates with curvilinear fibers. Lin et al. [36] applied different finite element methods (FEM) to analyze the natural frequencies and damping behavior of CFRP and GFRP plates. Their findings revealed substantial twisting behavior, which was more prominent when most of the strain energy was stored in the fiber through tension/compression, rather than in the matrix through shear deformation. These results confirmed the suitability of the FEM-based damp component model as a robust analytical tool for composite material structures.

As the popularity of CFRP is skyrocketing day by day, the researchers are focusing on the manufacturing methods of preparing CFRP by which it can be prepared more easily and efficiently. A perfect manufacturing technology, according to Potter [37], would have qualities such as minimal material costs, flexibility, good shape, excellent properties and high quality. Even though all the major sectors naturally want a production technology that simultaneously satisfies these needs, none currently exist. Currently, the autoclave molding method is widely adopted as a method for producing CFRP. This method achieves excellent product quality by curing the prepreg under high temperature and pressure, but it is also a challenge for expensive equipment and a time-consuming process. In this method, an autoclave is used which is mainly a mold whose upper side is flexible and lower side is rigid. Carbon fiber is impregnated with resin, in a semi-cured state the prepregs are formed after which the resin is cured at high temperature and high pressure. The autoclave is basically a pressure vessel which provides high pressure for molding. Stefaniak et al. [38] fabricated CFRP by using autoclave method for aerospace structure with good dimensional accuracy. A very ancient manufacturing method for preparing composite is Dry hand lay-up and spray lay-up technique. These methods are one of the simplest methods for composite fabrication. In these methods, a rolling stick or spray is used to apply resins uniformly into the fibers respectively. Hand lay-up technique is suitable for producing prototypes [39] whereas spray lay-up is ideal for moderate or small production [40,41]. Though high quality CFRP can be produced by this method, this is quite expensive and not suitable for mass production [42]. For this reason, in fields such as the automotive industry, which require mass production, improving production efficiency and reducing manufacturing costs have become important issues. Against this background, de-autoclave molding technology has been actively researched and developed in recent years [43]. As a

result, the focus on de-autoclave molding method became huge to reduce the cost and productive enhancement which highlights the massive usage of additive manufacturing in preparation of CFRP [44,45]. Particular attention has been paid to new molding technologies using CFRTP (Carbon Fiber Reinforced Thermoplastics). Ueda et al. [46] report that attaching rolling rollers to 3D printers reduces void rates and greatly improves the mechanical properties of CFRP. On the other hand, a study by Blythe et al. [47] found that the ATL (Automated Tape Laying) method was used to suppress fiber orientation variations, resulting in a 16.08% increase in flexural strength. By using a 3D printer or automated tape laying, a three-dimensional object can be obtained by additive manufacturing method. In this method, a tape like preregs is laminated and layered on top of each layer to form composites directly by heating and pressurizing [48]. Munoz-Guijosa et al. [49] beautifully explained the benefit of using 3D printing for composite manufacturing method which allows flexible lamination and good compatibility with autoclave. Another type of de-autoclave molding method is Filament winding method which is very suitable for producing hollow composites with high tensile strength [50]. Leong et al [51] used both compression and injection molding to fabricate the composites. It was concluded in their study that injection molding has some dimensional tolerances limitations whereas compression molding can be used for large composite panels. Though the application of injection molding is very wide in aerospace or automobile industries, a surface treatment is usually necessary for improving the fiber-matrix compatibility [52,53]. Silicon rubber molding method is another composite manufacturing method to duplicate any necessary parts easily. With a help from 3D modeling, the same design can be manufactured by using this method [54,55]. Another method called Resin film infusion is also used for composite fabrication in which a resin film is melted by heat onto the preform and the resin is impregnated by pressurization [56]. In addition, Garschke et al. [57] have precisely analyzed the influence of heating conditions in the RFI (Resin Film Infusion) method on the fluidity and final molding quality of the resin and obtained important insights for optimizing the molding conditions. There is another composite fabrication method which is gaining popularity nowadays because of its easy handling and procedure. This method is called liquid resin molding method where the resin is precipitated onto the carbon fiber to obtain CFRP. Among various methods, RTM (Resin Transfer Molding), VaRTM (Vacuum Resin Transfer Molding), PCM (Prepreg Compression Molding) methods are mostly used. In RTM method, a thermosetting resin is used as a core material whereas the carbon fiber is set in a molding beforehand. Then the resin is injected onto the fiber and cured. On the other hand, in VaRTM

method, the resin is poured and impregnated by using a vacuum. VaRTM is a very popular composite fabrication method that had been used for the past two decades [58]. It is basically a modification of resin transfer molding which uses a vacuum for assisting continuous flow of the resin from one side of the mold to another. Leclerc et al. [59] investigated the effect of resin injection velocity on void rate in the RTM method and reported that capillary number plays an important role in injection conditions. In addition, Abusrea et al. [60] worked to improve the performance of composite joints using the VaRTM method and improved the strength of joints by suturing and superposition techniques of multi-layer covers. One of challenging tasks of VaRTM is to reduce the change in part thickness over the distance [61,62]. Different endeavors were taken to modify the process parameters. A different orientation of the lay-up was used by Yuexen et al. [63] whereas a new configuration was introduced by Song [64] to locate the pressure variation. In PCM method, the prepreg is heated with a mold first and then pressed at high pressure. Additionally, a study by Lee et al. [65] confirms a significant reduction in void rate by using vacuum pressure in the PCM method, which indicates that the mechanical properties of the final CFRP are improved.

In this study, in addition to these existing technologies, we propose electro-deposition resin molding (EDRM: Electro-Deposition Resin Molding) as a new liquid resin molding method [66-69]. EDRM is a method of electrochemically depositing resin on the surface of the fiber by applying electrodeposition technology, which is an automobile coating technology, by energizing carbon fibers. This method is easier and more efficient compared to other molding methods which was first introduced by Katagiri et al [70], aiming to present an easy and cost-effective composite fabrication method. A study by Takisawa et al. [71] suggests that CFRP produced by EDRM has an advantage in attenuation properties compared to similar materials made by the VaRTM method, suggesting that this technology has great potential for low-cost manufacturing and mass production of CFRP. Some work had been done using this method. The effect of adding cellulose nanofiber (CNF) into CFRP prepared by EDRM had been investigated [72]. The mechanical properties were substantially increased by adding CNF. The maximum elastic modulus and tensile strength were 2.3 times and 1.7 times of the CFRP specimen without the CNF sheet, respectively. In another study, heat storable CFRP had been prepared by adopting EDRM as a fabrication method [73]. Both thermal conductivity and heat storage capacity were measured and found to be 2.2 W/m K and 2.8 kJ/m² respectively. Additionally, a comparative study between the CFRP with hole and without hole had been carried out to check the reinforcement [74]. It had been found that the tensile strength of the

specimen with the hole decreased to 68% of the tensile strength of the specimen without the hole. There are many experimental parameters in the EDRM manufacturing process, and it is necessary to evaluate their influence on the mechanical properties of the final CFRP. The investigation of manufacture parameters such as the thermosetting temperature and time optimization on flexural strength in the production of CFRP by EDRM and the impact of voltage and current conditions on the flexural properties of composites are yet to explore. Moreover, the evaluation of other experimental parameters is also insufficient. The motivation of this thesis is to use this easy and efficient method for manufacturing different types of CFRP and investigate various conditions of this method for optimization. Therefore, the purpose of this study is to evaluate the influence of energization conditions in EDRM on the static and dynamic properties of CFRP, and to investigate the relationship between the electrical voltage and current at the end of energization and the mechanical properties by measuring the flexural modulus and flexural strength using a four-point flexure test, and dynamic properties using vibration test. As this is a new method, exploring this method to prepare various types of CFRP is also very crucial. As of that, this study prepared curvilinear CFRP, structural capacitor, nylon 6 based carbon fiber/epoxy composites by using this new method EDRM. These results are expected to provide important basic data for the practical application of EDRM and contribute to the further development of the technology.

1.2 Different types of composite manufacturing technique

Composite materials can be fabricated through a range of techniques, each suited to specific material systems, structural requirements, and production volumes. Understanding these methods is essential for selecting the most appropriate approach for a given application. Below are the most widely used composite manufacturing techniques:

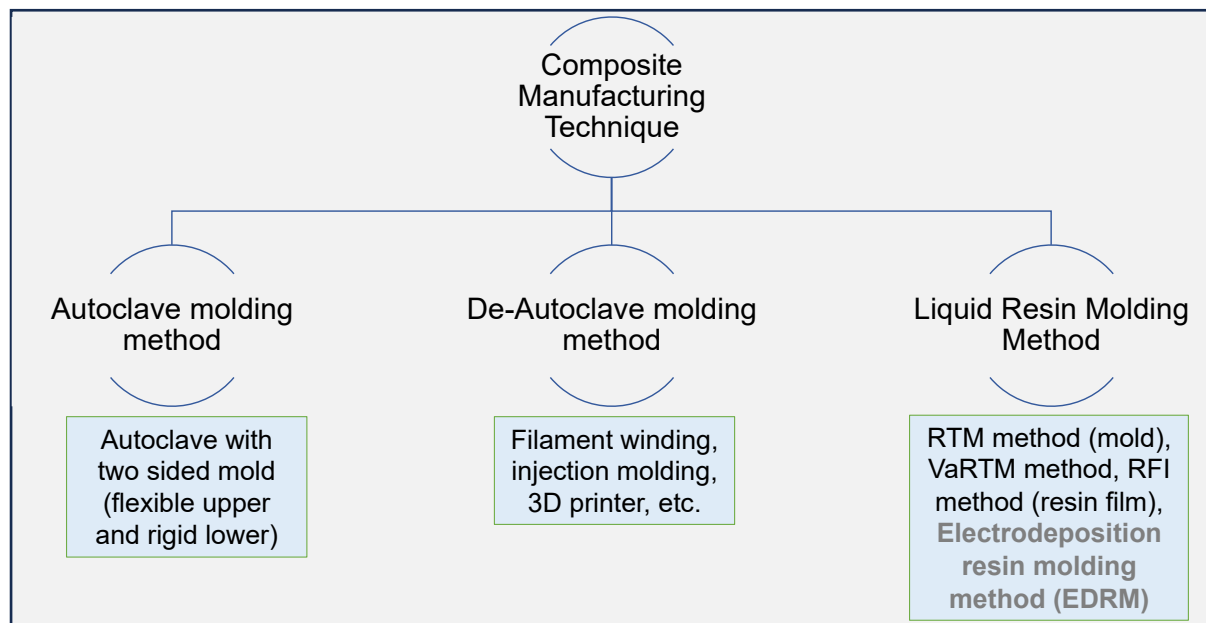


Fig. 1.1: Different types of composite manufacturing method

1.2.1 Auto-clave molding method

The aerospace, automotive, and high-performance sporting goods sectors are the main users of autoclave molding (shown in Fig. 1.2), a very sophisticated composite production process. Pre-impregnated (prepreg) fiber reinforcements are used in this process; they are placed into a mold and then heated and pressured within an autoclave chamber. Usually, the procedure starts with the meticulous manual or automated lay-up of prepreg components into a male or female mold. After that, the lay-up is sealed with a vacuum bag and covered with breather fabrics and release films. In an autoclave, the entire assembly is subjected to controlled curing cycles at temperatures ranging from 120°C to 180°C, depending on the resin system, and under vacuum and high-pressure conditions (typically 6–10 bar). The capacity to create composites with higher mechanical properties, low void content, and efficient fiber consolidation is the main benefit of autoclave processing. Vacuum and external pressure work together to remove air entrapment and encourage even resin dispersion. Furthermore, ideal curing is ensured by careful control of the temperature cycle, producing components with precise dimensions and strong structural integrity. Autoclave molding has several benefits, but it is time-consuming and capital-intensive, which makes it less appropriate for high-volume or budget-conscious applications. Its accessibility in smaller-scale industrial settings is further restricted by the need for specialized equipment and controlled surroundings. Nonetheless, autoclave molding

continues to be the gold standard in composite manufacturing for crucial applications where fatigue resistance, strength-to-weight ratio, and perfect surface quality are crucial, such as satellite structures, fuselage parts, and airplane wings.

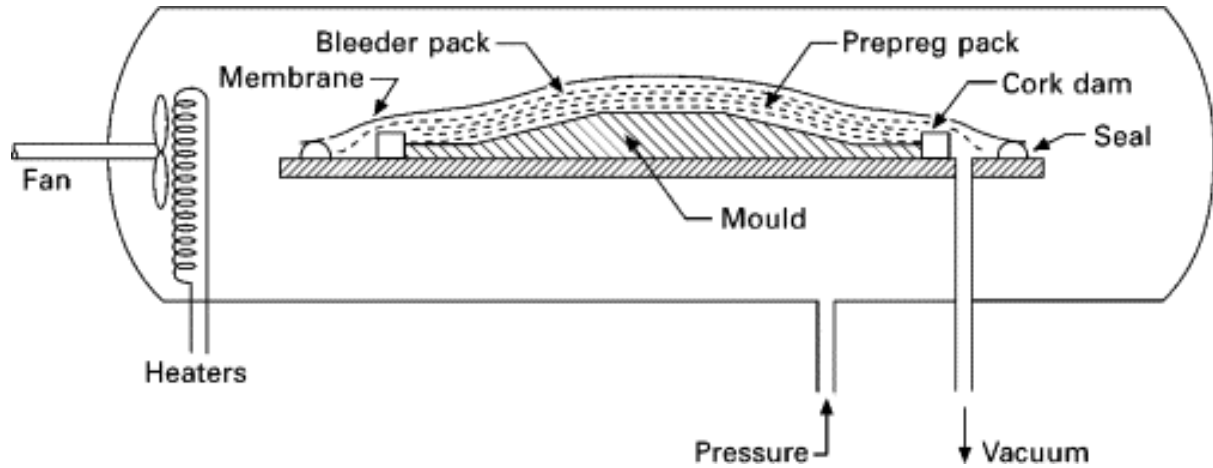


Fig. 1.2: Autoclave molding method [75]

1.2.2 De-autoclave molding method

A series of sophisticated composite manufacturing techniques known as the "De-Autoclave molding method" was created to do away with the necessity of conventional autoclave curing. The high cost, energy consumption, and infrastructure requirements of autoclave systems, especially for big or complicated structures, have led to the development of these methods. De-autoclave techniques seek to drastically cut cycle time and manufacturing costs while maintaining comparable mechanical performance and structural quality. Out-of-autoclave (OoA) prepreg materials, which are designed to cure efficiently under vacuum pressure alone, often with the aid of a heated oven or hot press, are usually used in this method. Out-of-autoclave (shown in Fig. 1.3) prepreg materials, which are designed to cure efficiently under vacuum pressure alone often with the aid of a heated oven or hot press are usually used in this method. Vacuum bagging is used to compact the laminate after lay-up, which is done similarly to autoclave techniques. As directed by the resin system, the assembly is cured in a controlled environment oven at a high temperature and vacuum (usually about 1 bar) rather than in an autoclave. Recent developments in fiber wet-out and resin composition have made it possible for De-Autoclave methods to achieve autoclave-quality void contents (typically less than 1%), particularly when moisture management and carefully regulated cure cycles are used. De-

autoclave processing is a broad category that includes methods including filament winding, resin infusion under flexible tooling (RIFT), injection molding, 3D printing, and oven-cured OoA preregs.

The benefits of De-autoclave methods include:

- Reduced capital and operational cost
- Lower energy consumption
- Scalability for large components
- Simplified infrastructure
- However, certain limitations remain, such as:
- Slightly lower fiber volume fraction
- Greater sensitivity to ambient conditions (e.g., humidity)
- More careful process control needed to ensure low void content

As a result, De-autoclave molding techniques are becoming increasingly popular in the wind energy, automotive, marine, and aerospace sectors, particularly for big parts or secondary structures where autoclave utilization is impracticable or prohibitively expensive.

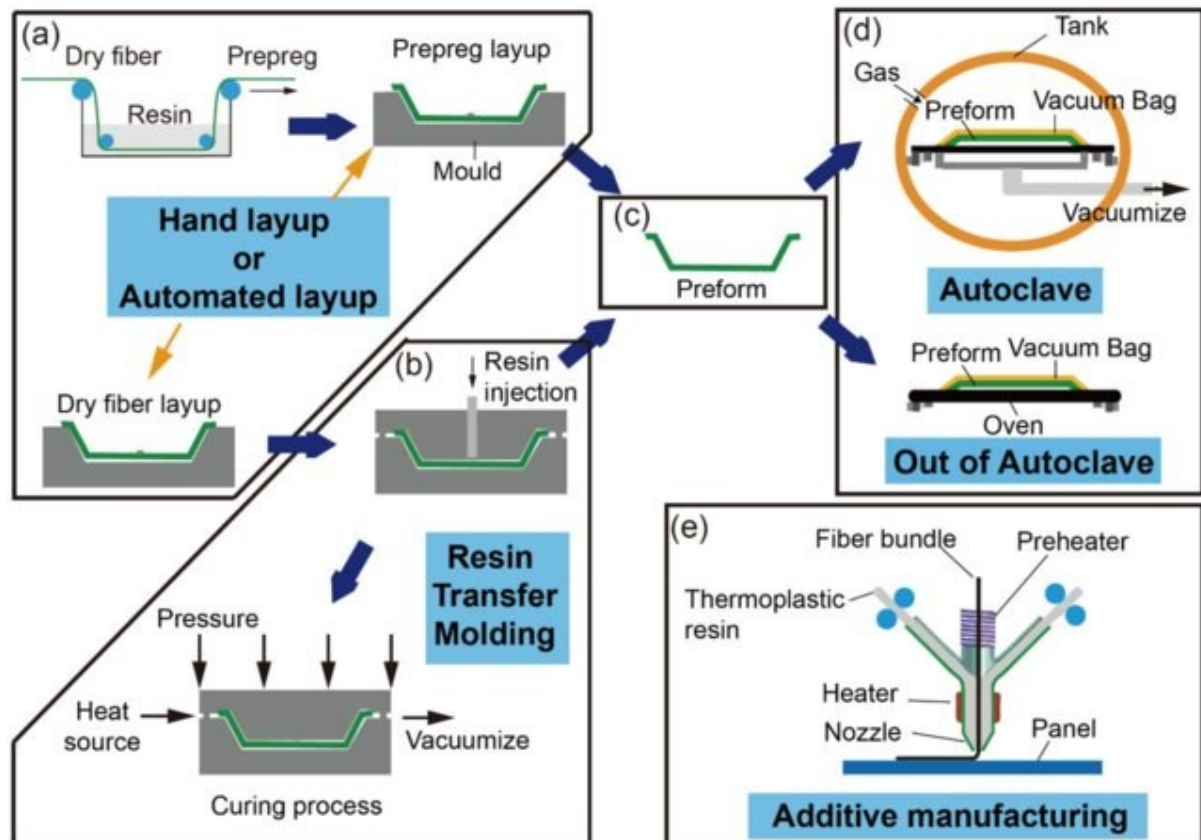


Fig. 1.3: Main manufacturing technologies for continuous fiber-reinforced composite structures: (a) Hand layup and automated layup with dry fibers or prepreg; (b) RTM technology including resin injection and preform curing process; (c) A preform waiting for curing; (d) Autoclave and OoA curing process; and (e) Additive manufacturing with in situ consolidation [73]

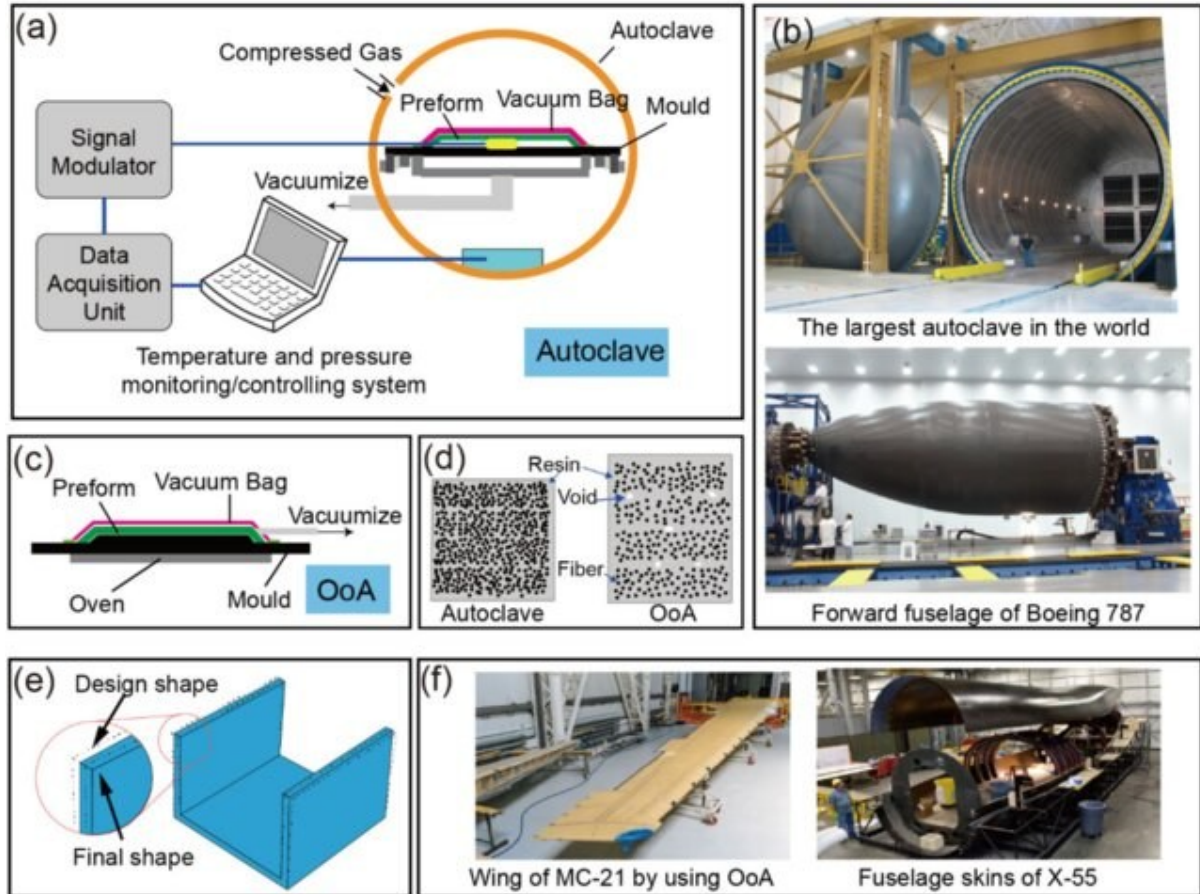


Fig. 1.4: Autoclave and OoA molding technology. (a) Autoclave and the temperature and pressure monitoring/controlling system, (b) The largest autoclave in the world from Boeing Company in Chicago, USA [74]. 2012, ACS Process Systems), and the forward fuselage of Boeing 787 [74], (c) Schematic diagram of OoA; (d) Schematic diagram of the cross sections of composites molded by autoclave or OoA, (e) Curing deformation schematic diagram in autoclave molding process, and (f) The wing of MC-21 [75] and the fuselage skin of X-55, an advanced composite cargo aircraft, molding by OoA process [76]

1.2.3 Liquid resin molding method

The Liquid Resin Molding (LRM) method is a popular composite manufacturing technique that involves shaping dry fiber reinforcements inside a mold and then impregnating them with liquid thermosetting resin. This class of methods offers versatility, cost-effectiveness, and scalability for producing complex structural parts, particularly in the automotive, aerospace, marine, and wind energy industries. The LRM process (shown in Fig. 1.5) generally consists of three main steps:

Preform Preparation: Dry fabric or fiber reinforcements (such as glass, carbon, or aramid) are laid into a mold, frequently using tailored stacks, stitched mats, or 3D reinforcements.

Resin Injection or Infusion: A catalyzed liquid resin is introduced into the mold under pressure, vacuum, or both, and the resin flows through the fiber architecture, thoroughly wetting the reinforcements.

Curing and Demolding: Depending on the resin's chemistry, the resin is either cured at room temperature or at a higher temperature following full wet-out. After curing, the part is taken out of the mold and may be post-cured or trimmed. Several widely used LRM strategies include RTM, or resin transfer molding uses a closed mold and moderate pressure to inject resin. A less expensive variation is called vacuum-assisted resin transfer molding (VARTM), in which resin is drawn into the fiber preform by vacuum.

The Seemann Composite Resin Infusion Molding Process (SCRIMP): It is a patented method that uses specific distribution media to improve flow and decrease voids. Resin films are positioned next to dry fibers and solidified using heat and pressure in a process known as resin film infusion (RFI).

Advantages:

- Good control over fiber alignment and resin content
- Capable of producing large and complex shapes
- Lower tooling cost compared to prepreg/autoclave processes
- Compatible with automation and scalable for medium to high production

Limitations:

- Resin viscosity and flow must be carefully controlled
- Risk of incomplete wet-out or voids if not properly managed
- Typically, lower fiber volume fractions than autoclave/prepreg methods

Liquid Resin Molding is especially valuable for parts needing excellent strength-to-weight ratios, economic effectiveness, and design flexibility. As resin chemistry and infusion methods continue to advance, LRM's use in structural and semi-structural composite applications keeps growing.

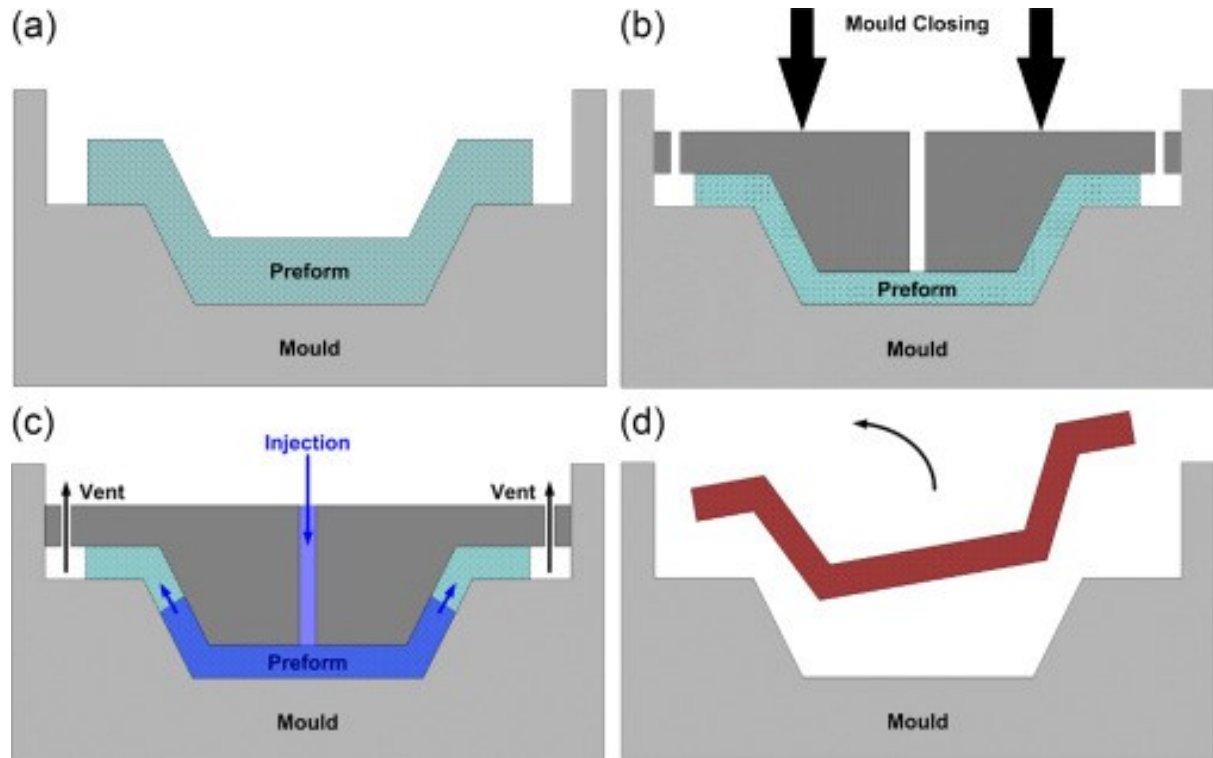


Fig. 1.5: Different steps during liquid resin molding method [77]

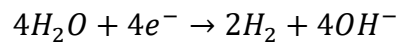
1.2.3.1 Electrodeposition resin molding method

EDRM is a molding technology in which a carbon fiber laminate is energized in an electrolyte containing an epoxy group and epoxy resin is deposited on the surface of the carbon fiber located on the cathode side. In this process, as shown in Fig. 1.8, the epoxy resin gradually precipitates from the carbon fiber surface, and the resin is impregnated between the fibers. After resin impregnation by electrodeposition is completed, the excess electrolyte remaining inside the preform is pressed off by a press machine. After that, the preform is heated and cured in a forced circulation thermostat with a metal plate and a vise evenly pressurized, and finally CFRP (carbon fiber reinforced plastic) is molded. Compared to conventional liquid resin molding technology, EDRM has the potential to reduce equipment costs and improve productivity. In addition, by adjusting the electrodeposition conditions, it is possible to finely

control the precipitation rate and impregnation amount of the resin. In this method, a conductive mold or substrate is immersed in a resin-containing electrolyte solution, where electric potential is applied to drive the deposition of resin onto the reinforcement or mold surface. The deposition occurs through electrophoretic or electrochemical mechanisms, depending on the resin system and additives used. Once the resin has formed a uniform coating or film around the reinforcement, curing is initiated thermally or chemically, leading to final part formation. The typical EDRM process involves substrate and mold preparation where conductive molds or electrodes are cleaned and pretreated for adhesion; electrodeposition of resin under applied voltage where resin particles or molecules migrate and deposit on the fiber preform; resin infiltration and consolidation where the deposited resin wets and penetrates the fiber network, supported by capillary and electrostatic forces; curing and demolding that heat or UV curing finalizes the part; the mold is then removed. A schematic diagram is shown in Fig. 1.6 and the actual setup of the EDRM is shown in Fig. 1.7. The chemical reaction of epoxy resin precipitation in the electrolyte is as follows [78,79].

Cathode reaction:

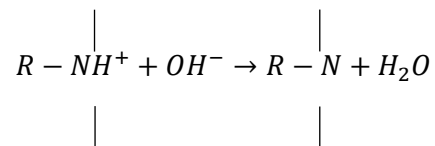
When electrons are supplied to the carbon fiber preform, the reduction reaction of water represented by the following equation proceeds.



The hydroxide ions (OH^-) produced react with the epoxy resin particles dispersed in the electrolyte.

Resin precipitation:

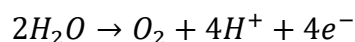
Epoxy resin particles have a cationic functional group, which reacts with OH^- to neutralize and become insoluble. Due to this reaction, resin particles precipitate on the surface of the carbon fiber. This process is shown by the following reaction equation.



Here, R indicates a molecular structure comprising an epoxy resin.

Anode reaction:

At the anode, the oxidation reaction of water proceeds, which is represented by the following formula.



The generated cations (H^+) play a role in maintaining the charge balance of cationic resin particles in the electrolyte.

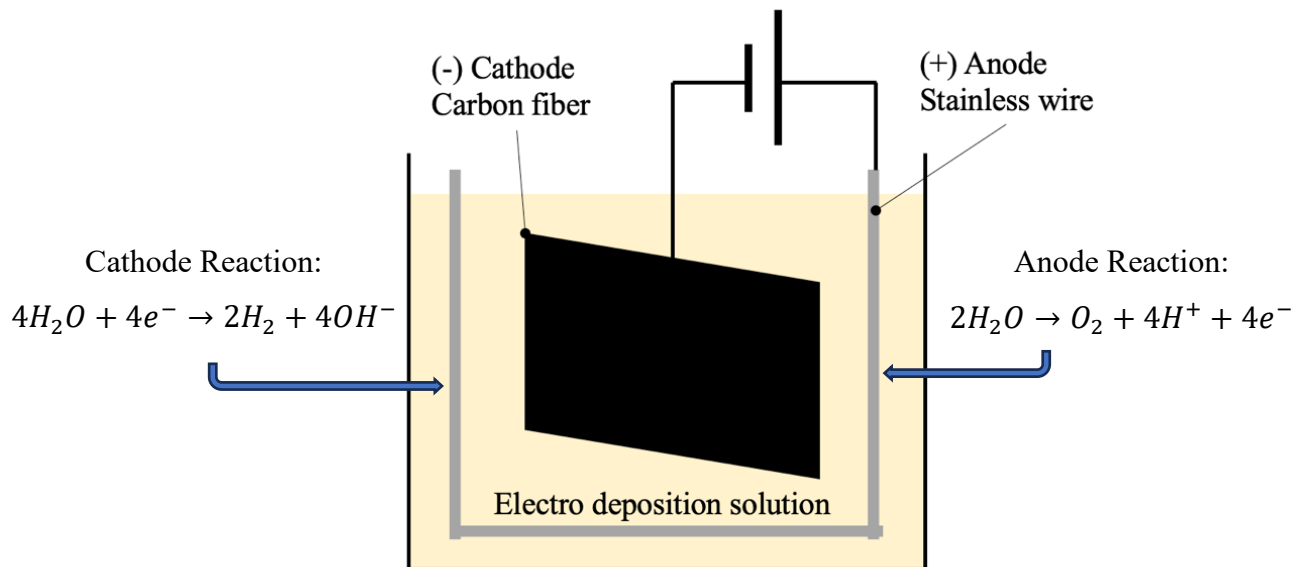


Fig. 1.6: Schematic diagram of EDRM



Fig. 1.7: Actual set-up of EDRM

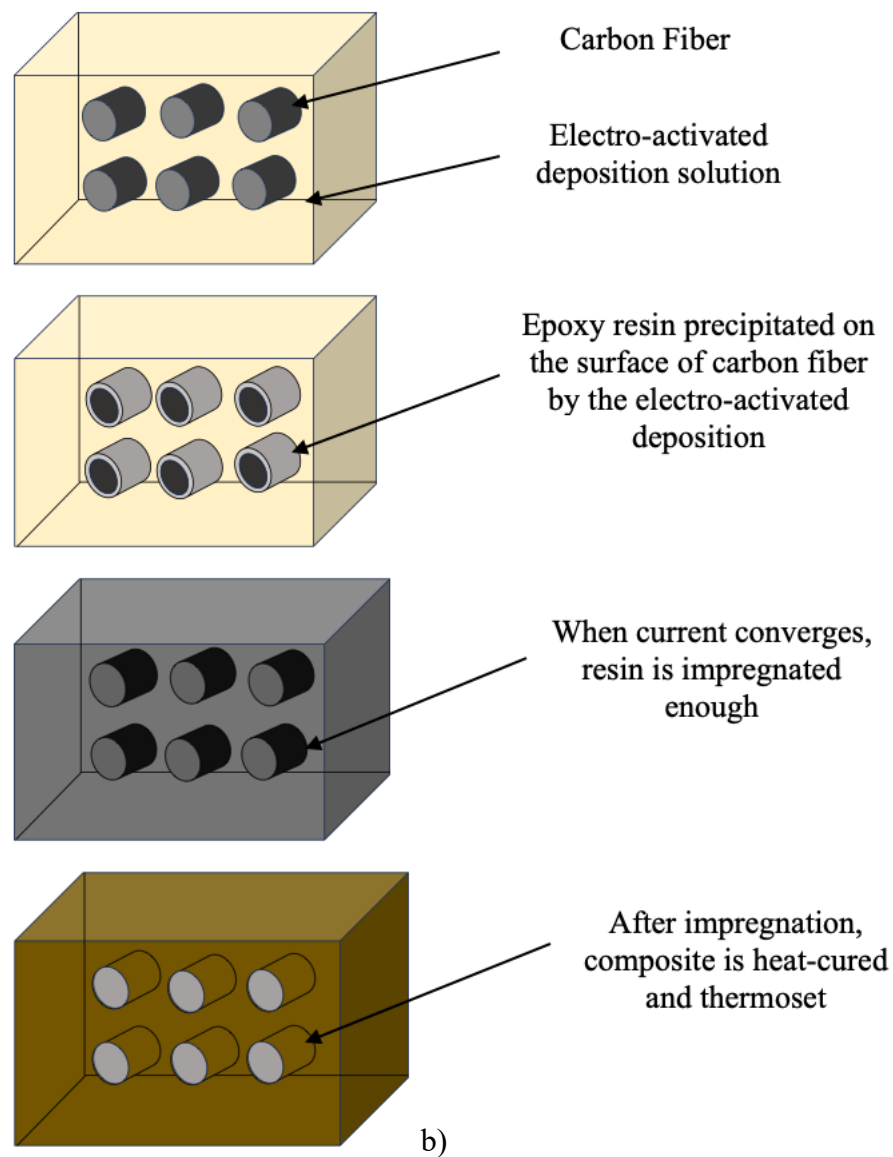


Fig. 1.8: Mechanism of EDRM where it shows the different stages of resin impregnation onto the fiber surface in the EDRM process.

EDRM offers the following benefits:

- Superior resin coverage and uniformity, especially on complex geometries.
- Low void content and improved fiber-matrix interface strength.
- Less waste of resin and eco-friendly processing
- Capable of resin precipitation on complex shaped preforms
- Good adhesion makes 3D structure preparation easier

- Capacity to add functional particles (such as graphene, MXenes, or carbon nanotubes) for additional characteristics like conductivity or barrier protection during electrodeposition

But EDRM also must deal with issues like:

- Limited uniformity and availability on a commercial scale
- Needs electrically conductive molds or substrates
- Complicated resin bath chemistry management for reliable performance.
- Difficulty in reproducibility

As a cutting-edge substitute for creating high-performance polymer matrix composites, EDRM is attracting more attention from researchers, particularly in situations requiring precision deposition, electrical conductivity, or multifunctionality. It is a potential method for both structural and functional applications in advanced energy devices, electronics, and aircraft because of its incorporation into next-generation composite systems.

1.3 Objectives of this research

Goals and objectives are important to strategic planning because they turn the mission and vision into specific measurable targets. Goals and objectives are concrete and help translate the mission and vision into reality. Every research paper must have some objectives. It's like if there is no objective of a paper, there is nothing to be gone through. As being the fact, the objectives of this research are given below:

1. To prepare CFRP by an easy and efficient method EDRM
2. To optimize the manufacture parameters of EDRM and check its effect on the static and dynamic properties of CFRP.
3. To use EDRM for fabricating different types of CFRP.
4. To prepare lightweight structural capacitors by using CNF
5. To check the effect of CNF inclusion into CFRP properties.
6. To prepare Curvilinear preform by using TFP machine.
7. To assess the vibration properties of curvilinear CFRP
8. To investigate the effect of adding nylon 6 into carbon fiber/epoxy composites.

1.4 Significant of this research

The advancement of lightweight, robust, and multipurpose materials in industries including aerospace, automotive, defense, and energy depends on the fabrication of high-performance Carbon Fiber Reinforced Polymer (CFRP) composites. Despite their effectiveness, traditional manufacturing techniques like resin transfer molding and autoclave curing are frequently constrained by their high energy requirements, lengthy processing periods, and substantial resin waste. This study investigates EDRM technology as a new, scalable, and energy-efficient method for creating different kinds of CFRP composites. EDRM provides more control over resin distribution and interface characteristics by directly depositing resin onto carbon fiber substrates using electrochemical principles. The following crucial elements are what make this work significant:

Innovative Processing: By combining electrochemical deposition and resin molding, EDRM offers a special method for fiber impregnation and precision coating that does not require high-pressure systems.

Improved Interfacial Bonding: The resin and carbon fibers make close contact due to the electrostatic attraction during deposition, which can greatly increase the composite's interfacial adhesion and mechanical strength.

Functional Tailoring: In order to create multifunctional CFRPs with additional electrical, thermal, or barrier properties, functional tailoring allows nanoparticles (such as CNFs) to be directly incorporated into the resin matrix during electrodeposition.

Sustainability and Efficiency: By doing away with the autoclave and avoiding the use of extra resin, EDRM lowers processing waste and energy consumption. This fits in nicely with the current needs for making composites in a sustainable way.

Design Versatility: The method enables the creation of specialized CFRP structures with regulated resin concentrations and layering techniques, providing versatility for a range of applications, including energy storage components and lightweight panels.

All things considered; this study adds to the technological toolkit for sophisticated composite manufacturing. Additionally, it provides fresh perspectives on the structure-property-processing relationships of CFRP systems made with electro-assisted techniques, which could pave the way for the development of intelligent, scalable, and reasonably priced composites.

1.5 Research Questions

The possible questions that can pop us from this research could be as follows:

1. How does the electrodeposition process influence the resin distribution and fiber-matrix adhesion in CFRP composites?
2. Can electrodeposition parameters (e.g., voltage, time, resin concentration) be optimized to improve the mechanical and electrical performance of CFRP composites?
3. How does the performance of CFRP fabricated via EDRM compare to that of composites made by conventional methods such as autoclave curing or vacuum-assisted resin transfer molding (VaRTM)?
4. What is the effect of incorporating functional nanomaterials (e.g., CNFs) into the electrodeposited resin on the multifunctional properties of CFRPs?
5. How does microstructural morphology (fiber wetting, void content, interfacial characteristics) vary across different resin formulations used in EDRM?
6. How does EDRM influence the dielectric, static, or dynamic behavior of CFRPs in harsh operating environments?
7. What is the effect of adding extra nylon 6 thermoplastic polymer into the carbon fiber-based epoxy composites?
8. Does this EDRM method can be used in mass production?
9. How does the EDRM process vary for producing bigger size of the preform?
10. Is EDRM a scalable and environmentally sustainable alternative to traditional molding techniques for industrial-level CFRP fabrication?

1.6 Structure of thesis

This thesis is divided into six chapters. The contents of each chapter are briefly given below

- ◆ Chapter 1: This chapter introduces the background of the study, research problem, research aims and specific objectives, the significance of the proposed research, the scopes and limitations, the research question and the structure of the dissertation.
- ◆ Chapter 2: The second chapter will include the details procedure of preparation of CFRP by using EDRM and the effect of manufacture variables on the static and dynamic properties of CFRP. It will discuss the production parameters of EDRM such as voltage, current, curing time and temperature on the bending, tensile and vibrational properties of CFRP.

- ◆ Chapter 3: The whole experimental procedure of CFRP capacitor preparation and the effect of adding CNF into CFRP to increase its multifunctionality will be presented in this chapter. It will also discuss the comparative analysis of CNF with other dielectrics such as paper, tissue, jute fiber and plastics on the multifunctional performance of CFRP.
- ◆ Chapter 4: The next chapter will emphasize the preparation procedure of curvilinear CFRP and its vibrational characteristics that were measured both experimentally and numerically. It will also discuss the preparation of curvilinear preforms such as sine, parabola by using Tailored fiber placement (TFP) machine and later resin-impregnated by EDRM
- ◆ Chapter 5: The fifth chapter will provide a detailed process of preparation of carbon fiber/nylon 6/epoxy composites and effect of inserting additional nylon 6 thermoplastics into CFRP composites on their mechanical properties. The roughness parameter and Raman spectroscopy of the CGRP will also be discussed in this chapter.
- ◆ Chapter 6: This chapter will summarize and critically discuss the main findings of the research, practical implications of the work, and how it advances the field and set out suggestions for future work.

Chapter Two

Preparation of CFRP by altering manufacture variables of EDRM

Abstract (Chapter two)

This study used the electrodeposition resin molding method (EDRM), an easy and effective technique, to create composite materials. Because it's a new technique, consideration of the impact of various production parameters on composite qualities was necessary. The effects of altering these EDRM voltage and electrical current conditions on the static and dynamic properties of composite materials were examined and tuned to fit the objective. Three distinct voltage values (100, 150, and 200V) and five different current conditions (0.5, 0.75, 1.00, 1.25, and 1.50A) were used for the manufacturing variables. In accordance with that, the effectiveness of heat treatment was also evaluated using two distinct temperature profiles. The bending strength of composites was somewhat influenced by the voltage and current conditions of the EDRM, but not in a particularly significant way. However, the bending strength of composites was significantly impacted by the heat-curing temperature as well as the heat-curing time.

2. Preparation of CFRP by altering manufacture variables of EDRM

2.1 Introduction

Electrodeposition Resin Molding (EDRM) is an innovative liquid resin molding process that adapts electrophoretic deposition principles to deposit resin directly onto conductive carbon fiber reinforcements using electrical energization [80,81]. The main benefits of EDRM are its potential for scalability, cost effectiveness, and ease of use. Carbon fiber substrates, which serve as the cathode, are submerged in an electrolytic bath made of resin, an electric field is applied to cause resin deposition, and the material is then thermally cured [82]. Previous research has demonstrated that CFRPs made using EDRM can outperform CFRPs made using conventional VaRTM techniques in terms of mechanical performance and damping qualities [83]. However, the influence of various manufacturing parameters on the final properties of CFRPs produced by EDRM remains underexplored. As discussed in section 1.2.3.1 Electrodeposition resin molding method, this method depends on various factors. As the electrodeposition is the main way of resin impregnation in this method, electrodeposition also depends on the voltage and current of the system to start the precipitation of resin. Additionally, resin viscosity, fiber impregnation, matrix crosslinking, and fiber-resin interfacial strength are all significantly influenced by critical variables such voltage, current density, deposition duration, resin composition, and curing conditions. The final flexural strength, elastic modulus, damping behavior, and void content of the composite may all be significantly changed by adjusting these parameters [84-86]. To fully realize its potential for high-performance CFRP manufacturing, a methodical investigation into the optimization of EDRM process parameters is necessary. By examining the effects of variations in electrical energization conditions (voltage and current), curing temperature and time, and resin properties on the mechanical and dynamic behavior of CFRPs, this study seeks to close that gap.

The research also explores the versatility of EDRM in fabricating different types of CFRP structures, including flat laminates, curvilinear composites, structural capacitors, and nylon 6-reinforced carbon fiber/epoxy composites which will be discussed in the next few chapters. Moreover, this study will help to establish EDRM as a viable and scalable method for producing next-generation composites by assessing the structure–property relationships under various manufacturing settings. In addition to advancing scientific knowledge of the EDRM

process, the results should offer fundamental information that may be used to manufacture CFRP lightweight structural and functional components on an industrial scale.

2.2 Methodology

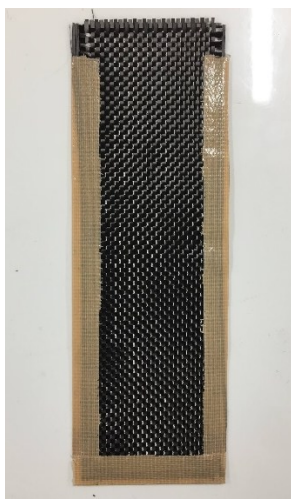
2.2.1 Materials

As for the fabrication, two different carbon fibers were used. TR3110MS (Mitsubishi Chemical Corporation) is a carbon fiber plain woven material and TR50S12L (Mitsubishi Chemical Corporation) used carbon fiber bundle. For the electrodeposition solution, Elecoat AR cosmetics (Shimizu Co., Ltd.) is used which contains epoxy groups. The resin is deposited on the cathode size while the electricity is being delivered in this cationic electrodeposition solution. The solution needs to be diluted because of the high concentration. The equal amount of distilled water is combined with the solution to dilute it (epoxy: water = 1:1). In order to avoid liquid degradation, direct sunshine, and hot surroundings, the solution is kept in a sealed container.

2.2.2 Preparing the Preforms

The plain weave carbon fiber was used for preparing the preform, each preform containing four layers of it. Two preforms were attached together forming eight layers of plain weave 0° lamination, forming $[PW0^\circ]_8$. The carbon tows were used only for tensile test specimen preparation. The carbon tows were sown on top of the base layers forming $[TFP/TFP/PW45^\circ/PW0^\circ]_s$. The orientation plain woven $[PW0^\circ]_8$ consisting of eight layers of carbon fiber, and all layers are in the same direction as the axis, in other words, one of the fiber bundles forming the plain weave is parallel to the longitudinal direction. First, with the help of a cutter, layers are cut into a size of 200 mm $\times$ 60 mm shown in Fig. 2.1(a). Eight equal size carbon fibers were cut. Then four layers of carbon fiber were attached together with a tape to make a preform. Then each preform was gone through the electro deposition and attached together to form $[PW0^\circ]_8$. The orientation $[TFP/TFP/PW45^\circ/PW0^\circ]_s$ also consists of eight layers, but different lamination. The base material is plain-woven carbon fiber while the bundle of carbon fiber tows was sown on top of the base material. For the base material, one layer is cut into a size of 260 mm $\times$ 100 mm. The direction of the fiber is in the same direction as the axis. Then another layer of the same size is cut but the direction of this layer is in 45° angles to

the direction of the axis, in other words, the fiber bundles are 45° angle to the longitudinal direction. Then these two layers are attached with a tape. Then this preform is placed on a sewing machine for further preparation. In this case, a Tailored Fiber Placement (TFP) machine, TCWM-101 (Tajima Kogyo Co., Ltd.) was used for sewing a layer of carbon fiber in one direction. On top of the base material, two layers of carbon fiber were sown, while the base material was attached with the base frame of the structure. The other two layers were attached forming four layers preform by using the TFP. The TFP layers are also parallel to the longitudinal direction. In similar way, the two preforms containing four layers each were attached together to form $[TFP/TFP/PW45^\circ/PW0^\circ]_s$ lamination.



(a)



(b)



(c)



(d)

Fig. 2.1: Appearance of preform (a) before EDRM, (b) after EDRM, (c) after heat curing, (d) preform for VaRTM.

2.2.3 Electrodeposition resin molding method

The graphical representation of the electrodeposition resin molding method is shown in Fig. 2.2, and the mechanism of this process is shown in Fig. 2.3. To control the voltage and current accurately, a stabilized DC power supply unit (Kikusui Electronics. PAT650-12.3T) was used. The preparation of the preform is the initial step in this procedure. The procedure outlined in section 2.2.2 was implemented to prepare the preform. The preform was then submerged in the electrodeposition solution while maintaining an open top portion of its perimeter. Typically, the electrode is attached to that upper section. So that no carbon fiber could seep into the solution, the remaining three sides of the preform were taped up. The preform was dipped into the solution to a depth of more than two thirds. The solution, which was kept in a beaker constructed of borosilicate glass, was used. A stainless-steel mesh covers the inside surface of the beaker and essentially acts as an anode. The preform was hung from a support, and a crocodile clip was used for connecting the preform.

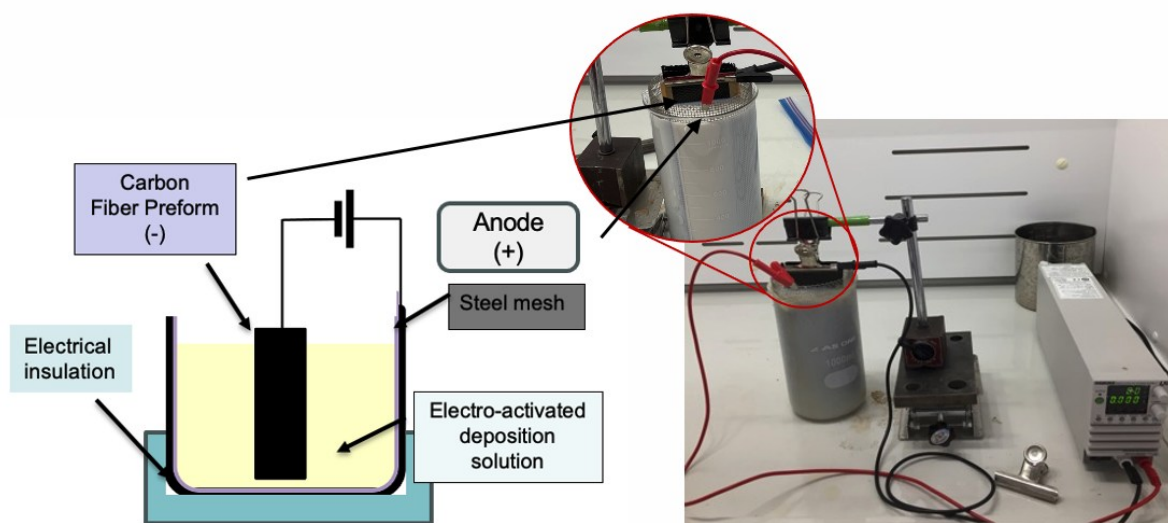


Fig. 2.2: The left figure is the graphical representation of EDRM where the right figure is the actual set-up for the experiment. Carbon fiber acts as a cathode here and the steel mesh acts as an anode.

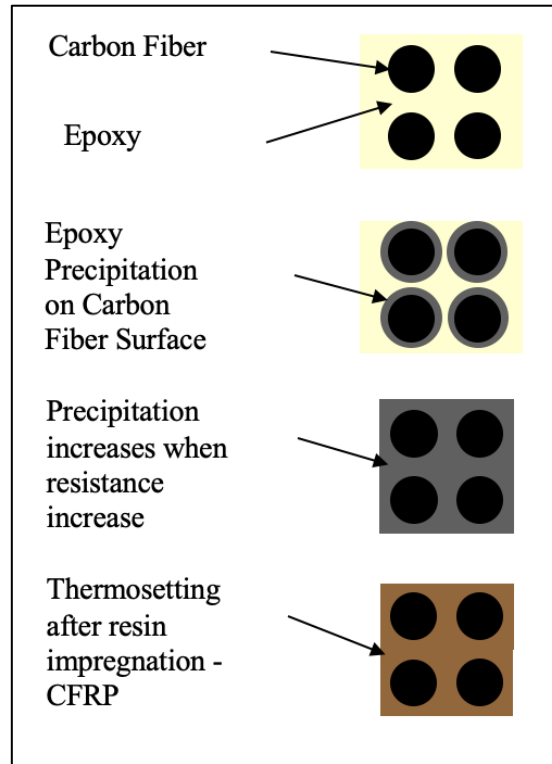


Fig. 2.3: Mechanism of electro-activated resin molding method [27]

To start the electrodeposition, the first thing that is required is to select a voltage and current condition. For initial electrical current condition, 0.5A had been chosen. This condition was changed five times as 0.5A, 0.75A, 1.00A, 1.25A and 1.50A to check the differences in electrical current on the mechanical characteristics of CFRP. Yet, once the resin precipitates onto the fiber, the resistance begins to rise. Since the initial current of 0.5A was maintained constant, the voltage rises according to Ohm's law. The voltage was kept constant at 200V for the first condition. Three different voltages 200V, 150V, and 100V were used in this study to examine how varying voltages affected the mechanical properties of CFRP. By setting the voltage limit currently, it switches to voltage control and shifts to a voltage constant energizing condition. The power source was shut off when the current started to decrease to a saturation level, signaling the end of the precipitation. The amount of electrodeposition solutions used will determine how long it takes to attain the saturation level. The preform was removed from the resin once all wires were unplugged and the power source was switched off. The appearance of the preform after electrodeposition is shown in Fig. 2.1(b).

2.4 Vacuum resin impregnation method

In the vacuum resin impregnation (VaRTM) method, the preform is impregnated with resin along the molding mold using the vacuum pump's negative pressure for the production of composite materials. The VaRTM method's outline is shown in Fig. 2.4, and the state of resin impregnation using the VaRTM method is shown in Fig. 2.5. Epoxy resin 801N for the base material (Mitsubishi Chemical Corporation) and epoxy resin curing agent 3080 (Mitsubishi Chemical Corporation) was used for VaRTM. Because the resin was quite viscous, peel plies were positioned between the resin diffusion media and the preform's upper and bottom sides to prevent the preform and resin diffusion media from molding together. Furthermore, after resin impregnation, heat curing was carried out using a forced circulation type thermostat VTR-115 (Isuzu Manufacturing Co., Ltd.). For three hours, 215°C was raised and maintained.

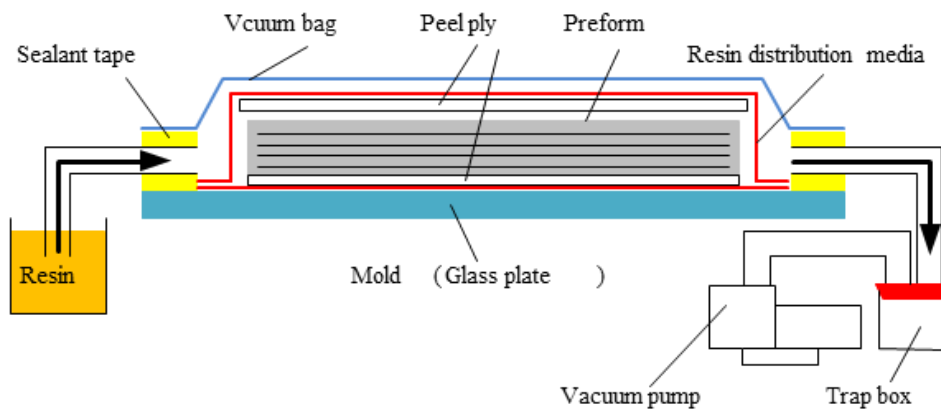


Fig. 2.4: Schematic diagram of VaRTM.

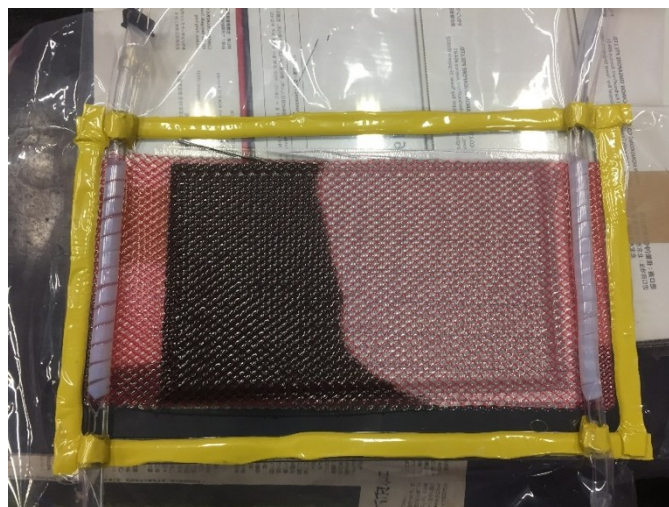


Fig. 2.5: Process of VaRTM.

2.2.3 Heat Curing

After the EDRM, the two preforms were then attached as a single preform containing eight layers of carbon fiber. Then this preform was placed between two fluoropolymer film which together was put between two metal plates. The metal plates were tightened and pressurized with the help of a vise. The appearance of preform after heat curing is shown in Fig. 2.1(c). For heat curing two different temperature profiles had been followed in this study. For the first profile, the final temperature was chosen to be 215°C while for the second profile it was 230°C. Both profiles had two stages. For the first stage, the temperature reaches from room temperature to 215°C or 230°C according to profile. The duration of the first stage was two hours. In the second stage, the same temperature was kept constant for different hours such as 2 hours, 4 hours and 6 hours to investigate the effect of heat curing period on the mechanical properties. Table 1 provides different codenames for different heat-curing conditions. Then from this heat-cured preform, specimens for various tests were prepared. Fig. 2.6 shows an example of a cross-sectional surface of a specimen produced by the electrodeposition resin impregnation method which was captured by a digital microscope.

Table 2.1. Code names for different heat curing condition

Temp Profile-A (2 hours)	TP A1
Temp Profile-A (4 hours)	TP A2
Temp Profile-A (6 hours)	TP A3
Temp Profile-B (2 hours)	TP B1
Temp Profile-B (4 hours)	TP B2
Temp Profile-B (6 hours)	TP B3

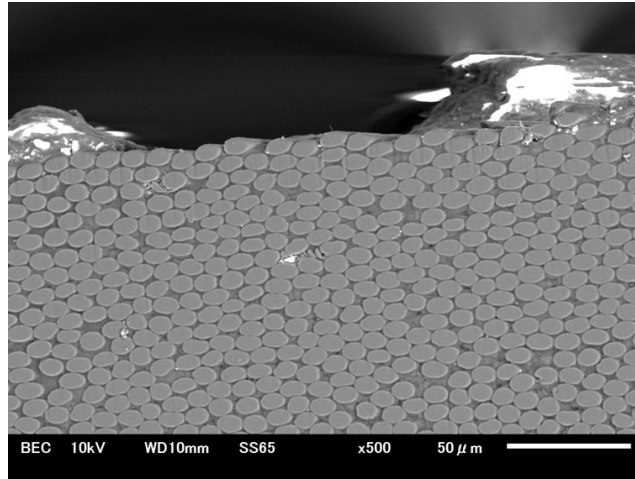
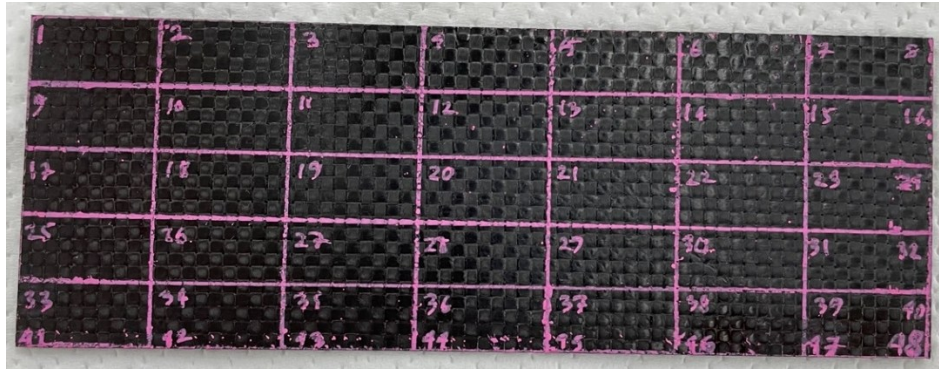


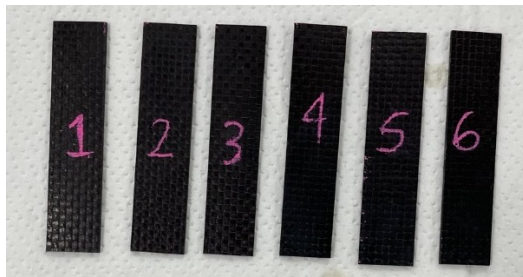
Fig. 2.6: Cross-section of the specimen observed by SEM

2.2.4 Specimen Preparation

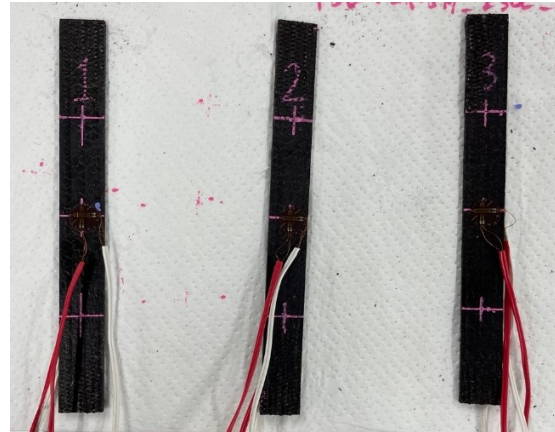
As one of the main objectives of this research is to assess the static as well as dynamic properties of CFRP, different types of specimens were prepared. The specimen for the vibration test was prepared initially. After the heat curing, by using the composite material cutting machine AC-200CF (Maruto Manufacturing Co., Ltd.), the preform was cut according to the specifications of different tests. For vibration test, a rectangular shape of specimen is cut by this machine. The size of the specimen was 140 mm $\times$ 50 mm. After finishing the vibration test, the specimen was used for the bending test. The same specimen was used for both vibration and bending tests to check the relationship under the same condition. But the bending test specimen size is quite different than that of vibration test. Six equal size of specimen was cut from the specimen that was used for vibration test. The size of each specimen was 70 mm $\times$ 15 mm. However, the tensile test specimen was cut directly after finishing the heat treatment. Three equal pieces of specimen were cut from the preform prepared before. The size of each specimen was 150 mm $\times$ 15 mm. All specimen pictures are shown on Fig. 2.7. After preparing all specimens, the density of the specimens made from five different conditions were measured. The values are given in Table 2.2. The average density of these CFRP was found to be around 1415 kg/m³.



(a)



(b)



(c)

Fig. 2.7: Specimen of (a) Vibration test, (b) Four-point bending test, (c) Tensile test

2.2.5 Vibration Test

A vibration test had been performed in this study to assess the dynamic properties of the fabricated carbon fiber composites where the natural frequencies and damping had been evaluated. To conduct this vibration test, DS0320 software (Ono Sokki) has been used. The sample was prepared by the procedure mentioned in section 2.3. There were 48 nodes in the specimen. Then the specimen was hung from a standby using a rubber band, and the acceleration sensor was attached at node no. 30 of the specimen. Each node was impacted by a hammer five times, and only the average value of these five struck was recorded. After recording all values, the natural frequencies and damping were obtained by using a software called MEscape. (Vibrant Technology). The configuration and the experimental setup of vibration tests are shown in Fig. 2.8 and 2.9 respectively.

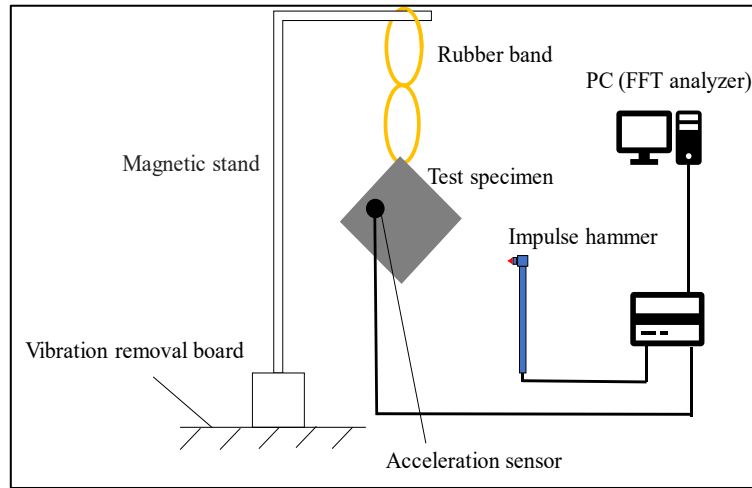


Fig. 2.8: Configuration of equipment for vibration test.

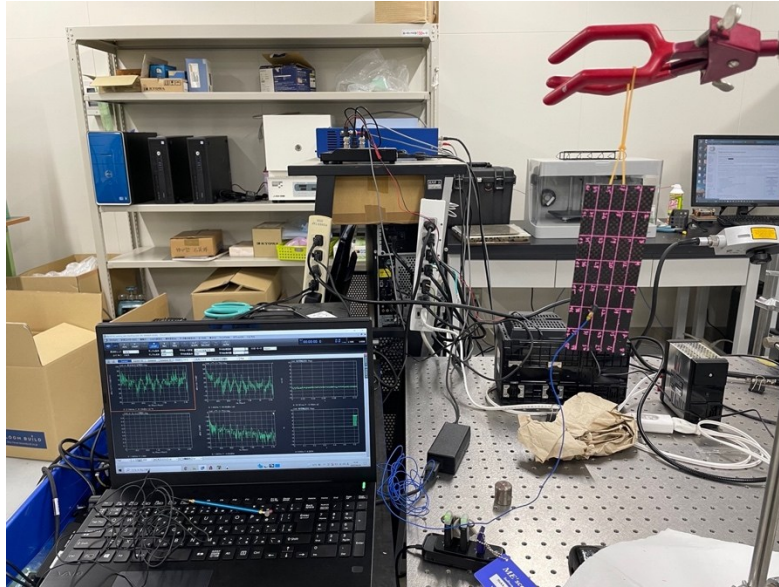


Fig. 2.9: Experimental setup of vibration test.

2.2.6 Four-point bending test

As the method of bending test, four-point bending test had been adopted instead of three-point bending test in this study. As a testing machine, the Ez Graph (Shimadzu Corporation) was used. By attaching a jig for a four-point bending test and moving the crosshead downward, force in the bending direction can be applied to the specimen. The distance between the upper

indenters was set to 10 mm and the distance between the lower indenters was set to 50 mm. The test speed was determined by the following equation described in JIS K 7074 [25].

$$V = \frac{0.21S_r L^2}{h} \quad (1)$$

V : Test speed (mm/min)

S_r : Strain rate (m^{-1}), $S_r = 0.01$

L : Fulcrum distance (mm)

h : Thickness of specimen (mm)

By putting the value of $L = 50$ mm and $h = 1.5$ mm into the equation, we get $V = 3.5$. the average thickness of the specimen was considered in this case. Therefore, the test speed was set to 3 mm/min. The specimen for the four-point bending test had been prepared according to the method mentioned in section 2.4. The specimen was loaded under specific conditions by operating a PC software TRAPEZIUM (shown in Fig. 2.11). When the displacement reached 9 mm, the tester was closed.

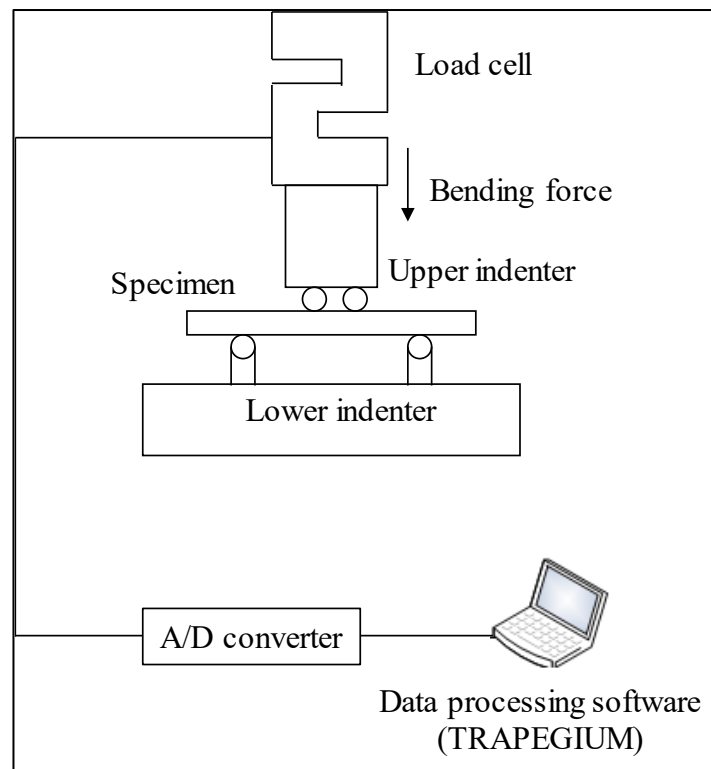


Fig. 2.10: Configuration of equipment for 4-point bending test [67].

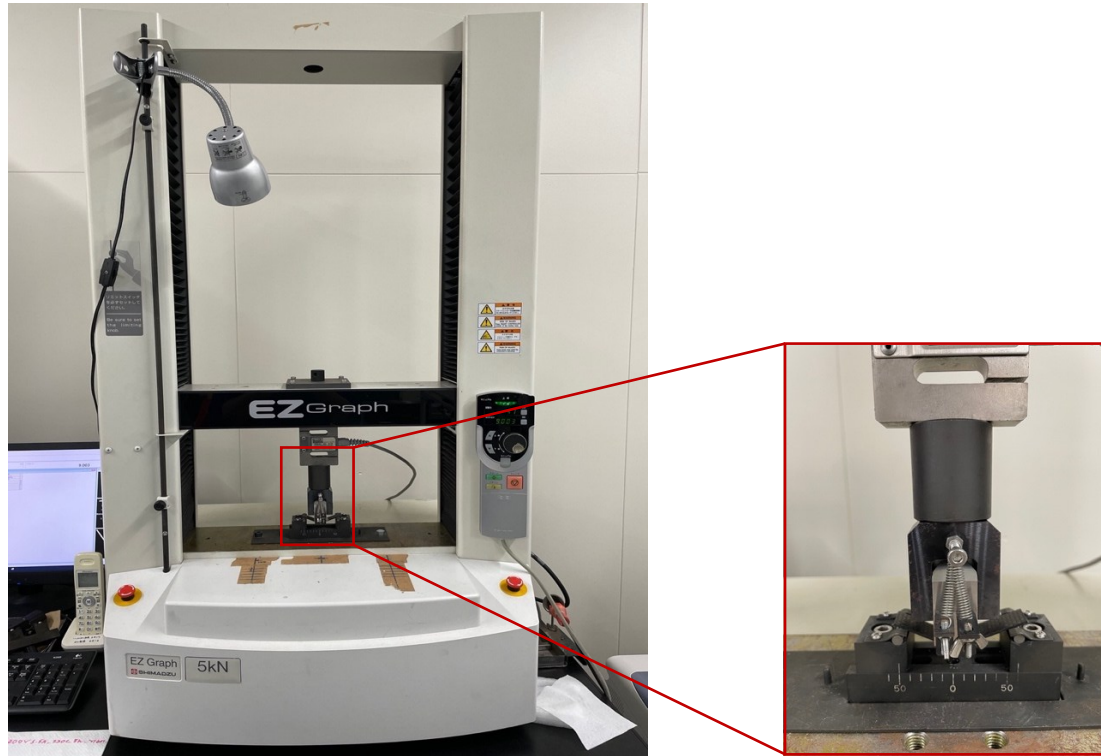


Fig. 2.11: (a) Four-point bending testing machine EZ Graph; (b) Specimen in contact with both upper and lower indenter

2.2.7 Tensile test

Tensile test was done to evaluate the young modulus of the material. For the testing machine, the autograph AGS-10kNX (Shimadzu Corporation), a tabletop precision universal testing machine had been used. The distance between the grippers is 75 mm, and the test speed is 1 mm/min with reference to JIS K7164 [25]. For strain measurement, the strain gauge FLAB-2-11-1LJC-F (Tokyo Sokki Co., Ltd.) was used. In addition, for adhesion of the strain gauge to the specimen, the instantaneous adhesive for the strain gauge (Kyowa Electric Co., Ltd.) was used. In order to amplify the voltage-change of the strain-gauge, the dynamic strain meter (shown in Fig. 2.14) DPM-711B (Kyowa Electric Co., Ltd.) was used to simultaneously measure the vertical strain and the transverse strain, respectively. The configuration and the testing machine are shown in Fig. 2.12 and 2.13 respectively.

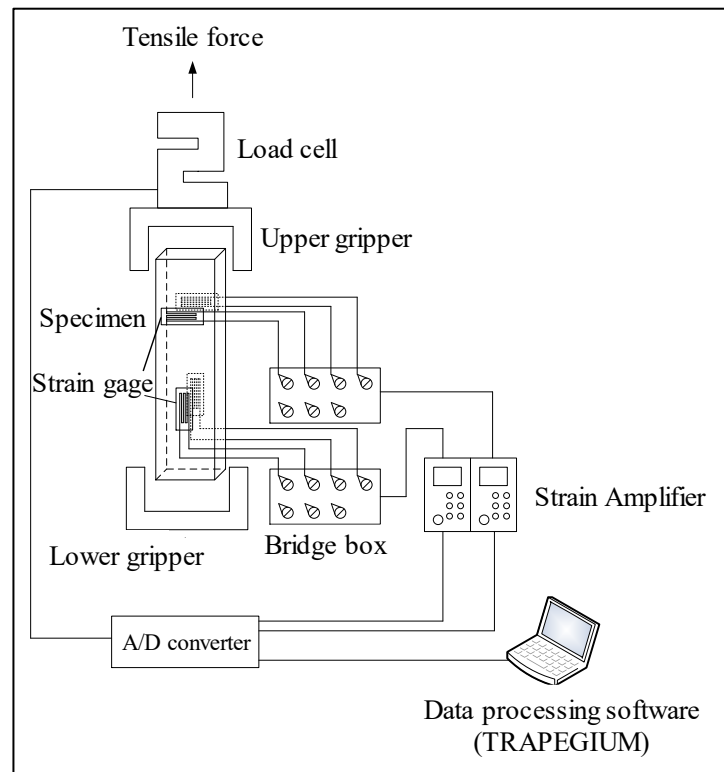


Fig. 2.12: Configuration of equipment for tensile test [67]

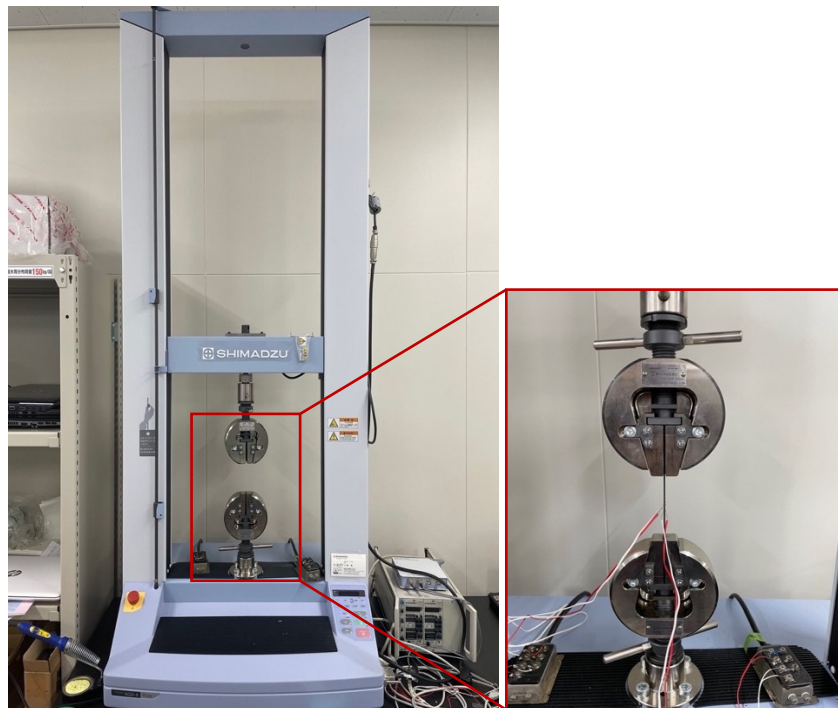


Fig. 2.13: (a) Tensile testing machine AGS-10kNX; (b) Gripped specimen.

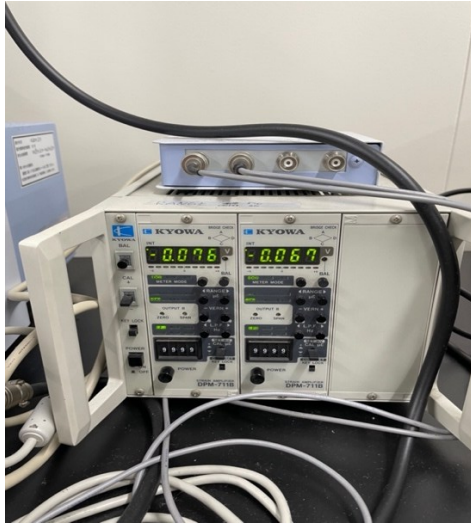
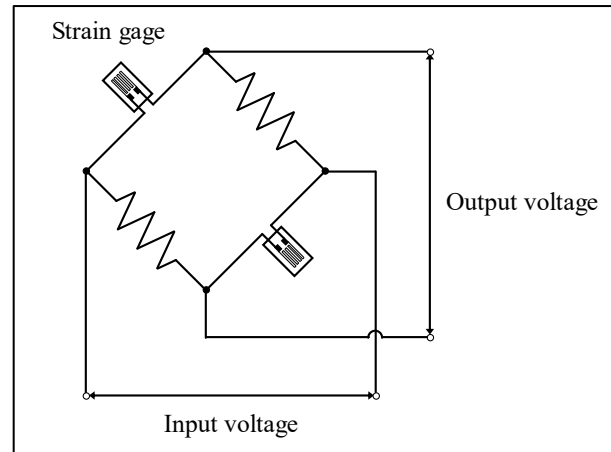


Fig. 2.14: (a) Strain Amplifier DPM-711B; (b) Electric circuit for detection of strain.

2.5.4 Cross-sectional observation

For observation, the microscope VW-600 was shown on Fig. 2.15 (Keyence Co., Ltd.). The microscope can record images, measure length, and perform data processing, and the magnification of the zoom lens is 20, 30, 50, 100, 150. For observing the cross section, 100 times and 150 times were used depending on the thickness. Using sandpaper (800, 1000, and 2000) and acetone, the observation surface was washed using Kim Wipe. Fig. 2.16 shows an example of a cross-sectional surface of a specimen produced by the electrodeposition resin impregnation method.



Fig. 2.15: Microscope

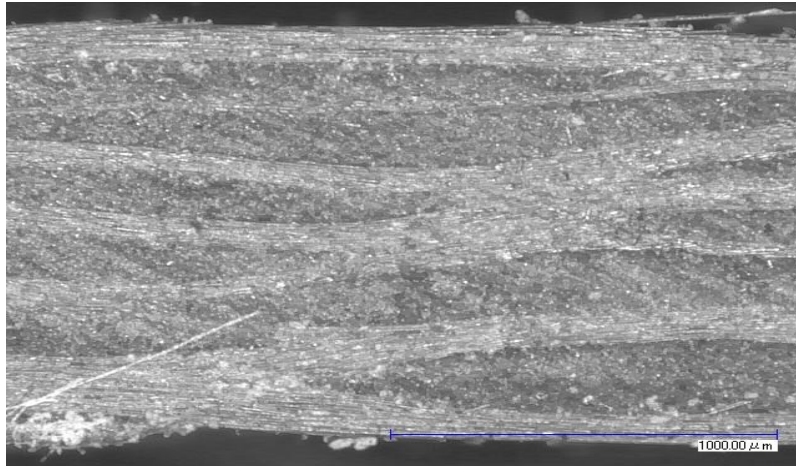


Fig. 2.16: Cross-sectional observation of CFRP [PW0°]₈ laminates made by EDRM.

2.5.5 Density Measurement

In the density measurement of fabricated composite, the manual process, such as measuring the dimensions and mass, can be little deviated. Some unevenness in the surface makes it difficult to measure actual thickness. In this study, an electronic balances GR-200 was used to measure the density which is shown in Fig. 2.17. Here, the density was measured using Archimedes' principle from mass in air and mass in water. The sample for density measuring from the actual CFRP produced, and each of the samples had a size of 10 mm × 10 mm which is shown in Fig 2-45. The density of the specimen is shown in Table 2.2.



Fig. 2.17: Analytical Balance

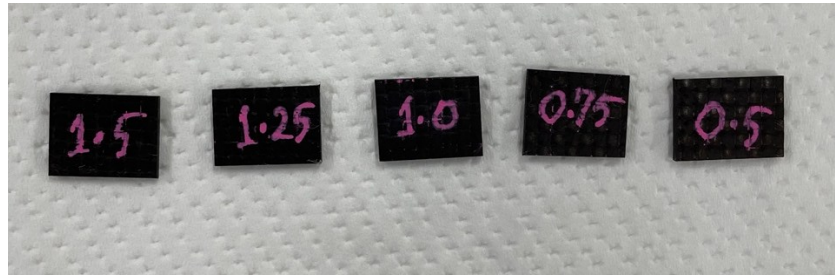


Fig. 2.18: Specimen for density measurement

Table 2.2 Density of the specimen

Voltage [V]		Density [kg/m ³]			
Current [A]		1	2	3	Average
200V	0.50A	1433.2	1455.3	1419.2	1435.9
	0.75A	1473.4	1410.7	1374.2	1419.4
	1.00A	1451.9	1454.4	1355.9	1420.7
	1.25A	1392.0	1392.6	1396.8	1393.8
	1.50A	1414.4	1419.1	1414.6	1416.0

2.3 Result and Discussion

2.3.1 Electrodeposition resin molding

As mentioned in section 2.3, different voltage and electrical current conditions were applied to the specimen. As an example, the relationships of both voltage and current with time are shown in Fig. 2.19 where 150V and 1.5A conditions were applied. The left scale and yellow line portray the relationship of voltage with time, while the right scale and blue line show the relationship of current with time. As can be seen, the voltage reached the maximum value of 150V within around 220 sec and was constant throughout. The current was fixed at 1.5A until the voltage reached the maximum and dropped to 0.1A due to a sudden increase of resistance. The diffusivity of the polymer in the electro-activated deposition solution affects the precipitation rates [20]. After around 17 mins, the current converges which indicates the convergence of resistance. The very small change of the current exhibits that the precipitation

on the fiber surface is complete. The preform after going through the electro-deposition is shown in Fig. 2.1(b). The precipitation of the resin onto the fiber surface is visible.

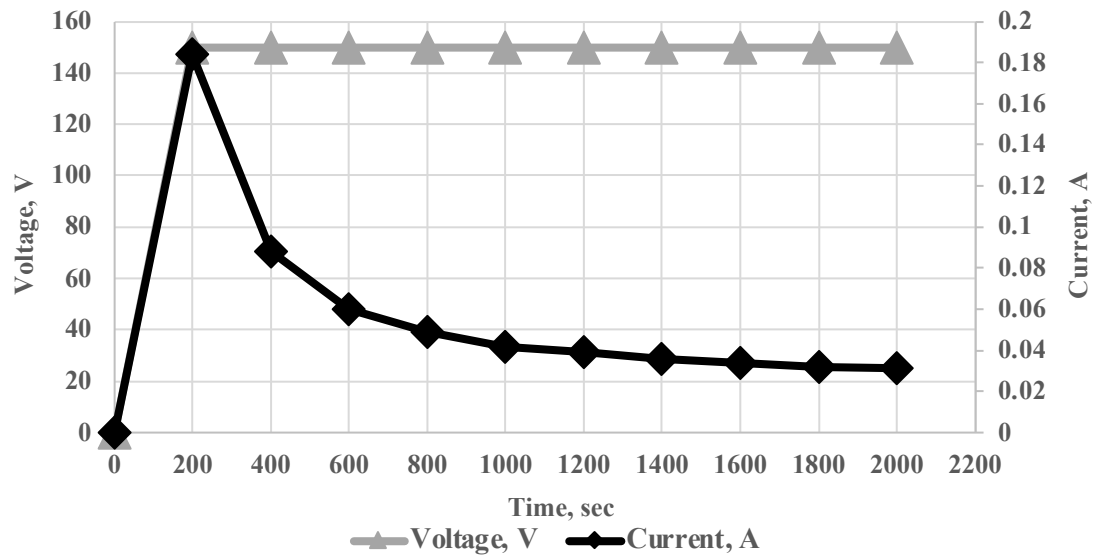


Fig. 2.19. Relationship of Voltage and current with time during the electrodeposition process

2.3.2 Natural frequency and damping

After preparing the specimen following section 2.2 and performing the test following section 2.5, the data of natural frequencies and damping of the fabricated composites had been found. The data of both natural frequencies and damping was found up to five modes. Additionally, the sample's modal visualization was attained. Fig. 2.20 depicts a modal visualization example. For investigating the composites' vibrational properties, only $[PW0^0]_8$ was used as preform. The modal visualizations showed the typical scenario for mode 1 to mode 5. In the contour diagram representing the mode shape, the absolute amount of out-of-plane displacement is visualized by color, and the displacement increases as it approaches red. Nevertheless, a comparative study for the natural frequencies and damping for different EDRM conditions were shown in Fig. 2.21 and Fig. 2.22 respectively. It is evident from Fig. 2.21 that there is not a significant shift in the frequencies. The range was incredibly small, so it wasn't important. For mode 1, the natural frequencies range from 355 Hz to 370 Hz. The maximum and minimum natural frequencies for mode 2 were 509 Hz and 491 Hz, respectively. The natural frequencies for modes 3 and 4 were 880 Hz to 930 Hz and 1320 Hz to 1370 Hz, respectively. The highest and lowest frequencies for mode 5 were 1820 Hz and 1700 Hz, respectively. As can be seen,

there is relatively little difference in the natural frequencies under varied voltage and current conditions. The modal damping situation in Fig. 2.22 illustrates the same scenario. Despite having a zigzag appearance, the range was insufficient and insignificant. The largest difference for various conditions was 0.6, while the smallest difference was 0.1, which was quite minor.

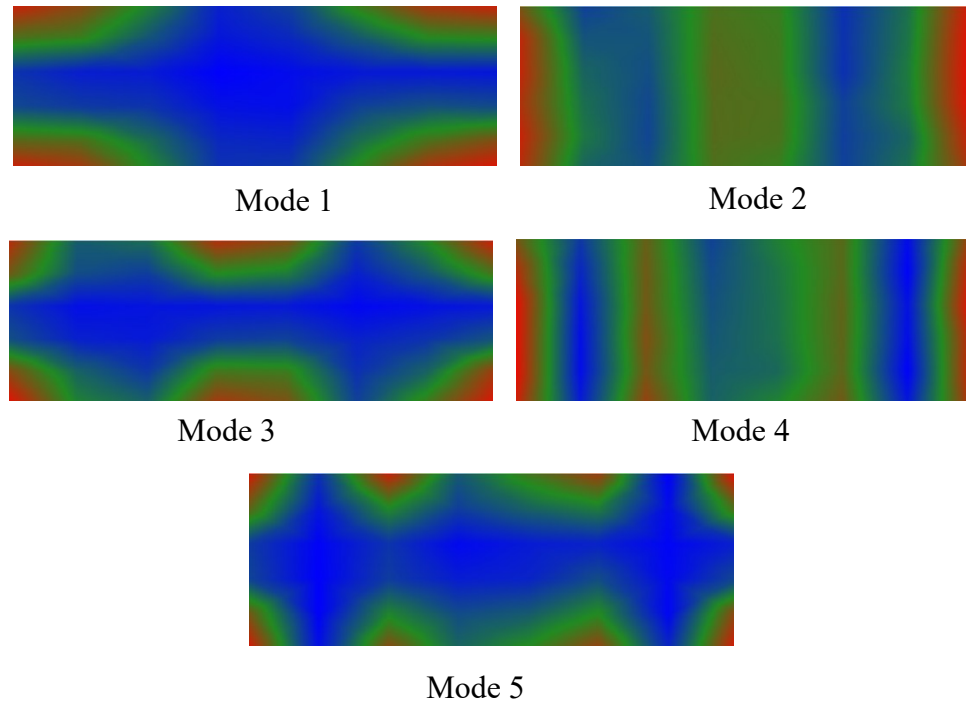


Fig. 2.20. Modal visualization of the composites

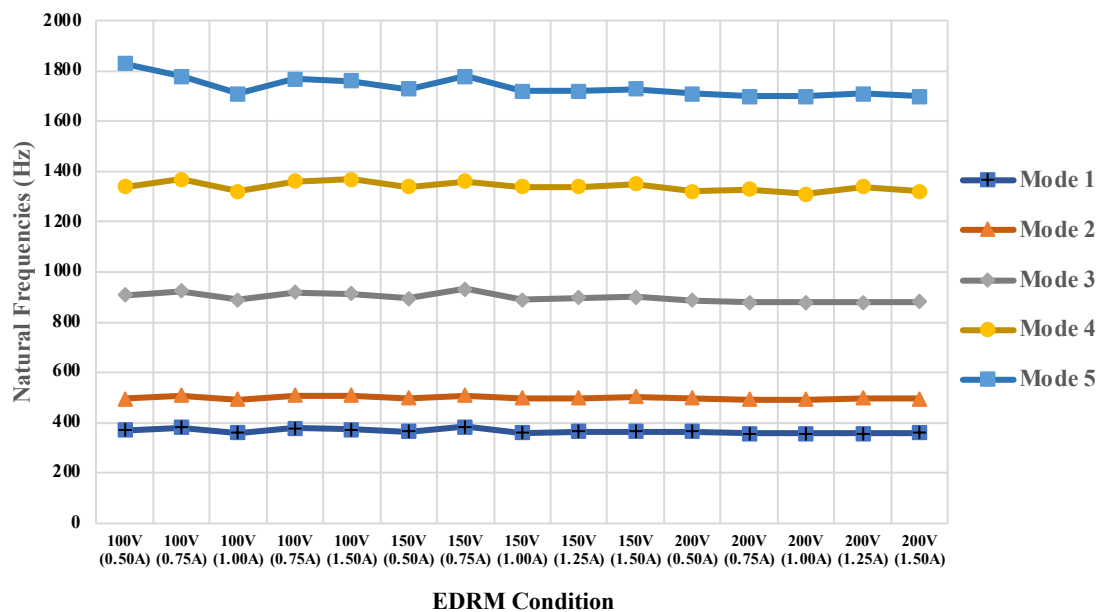


Fig. 2.21. Natural Frequencies of CFRP at different EDRM conditions

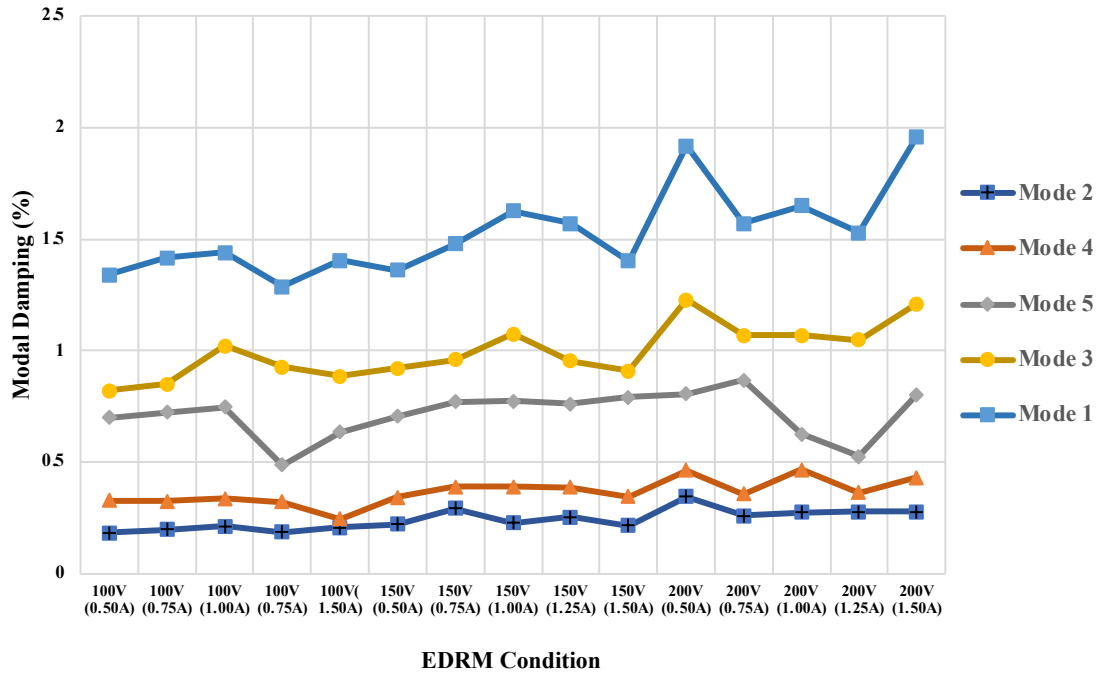
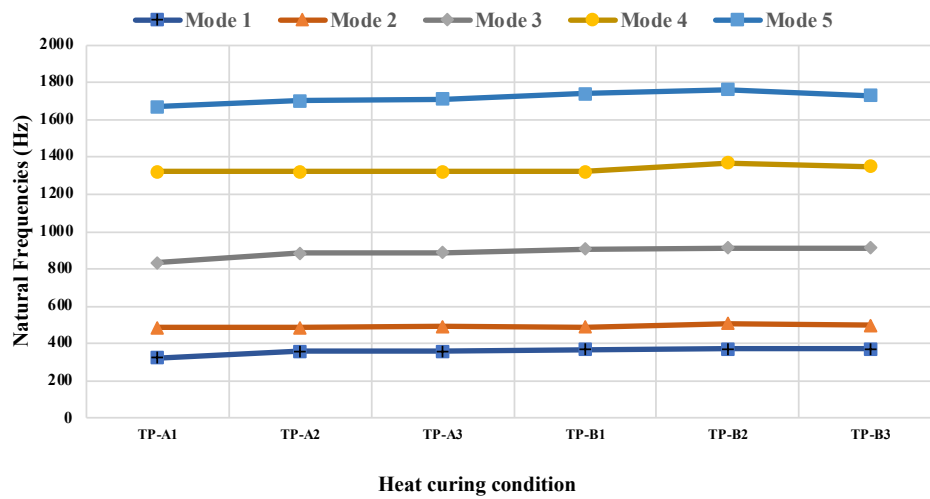


Fig. 2.22. Modal damping of CFRP at different EDM conditions

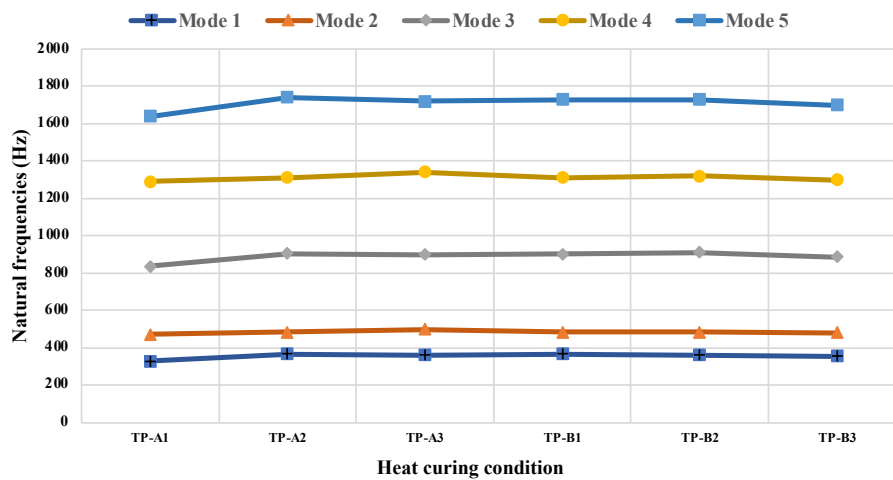
Fig. 2.23 (a-c) depicts the composite's natural frequencies under various heat curing conditions, and the EDM condition for these figures were 100V (1.50A), 150V (1.50A), and 200V (1.50A) respectively. Each image compares natural frequencies with a constant EDM condition but a variable heat curing temperature and duration. The three graphs all displayed the same pattern. The line is practically straight because the variation in values was so insignificant, clearly showing how the frequencies changed slightly when the heat curing conditions changed. These three plots revealed that the heating time and temperature had minimal impact on the modal frequencies.

The value of modal damping when the heat curing period was changed is shown in Fig. 2.24 (a-c). The graph's characteristics and the circumstances are the same as those for the natural frequencies that were previously explained. However, the situation was very dissimilar from that of natural frequencies. The adjustment in heating time or temperature has no impact on the natural frequencies. However, the figure for damping indicated a different value than the other conditions when heated for 4h at 215°C. Every value from mode 1 to mode 5 was distinct from other circumstances. For instance, when using mode 1, the damping for 4h heating session was 5.39 while it was 1.4 during an 8h period. Similarly, the 4h condition for mode 3 was 226%

greater than the 8h condition. However, there weren't significant variations between modes 2 and 4. When the heating temperature was 230°C, the damping of the 4 hours heating period was higher than that of the 6h and 8h heating times despite the modest variances. It clearly holds forth that shorter heating time has higher damping values than longer period.



(a)



(b)

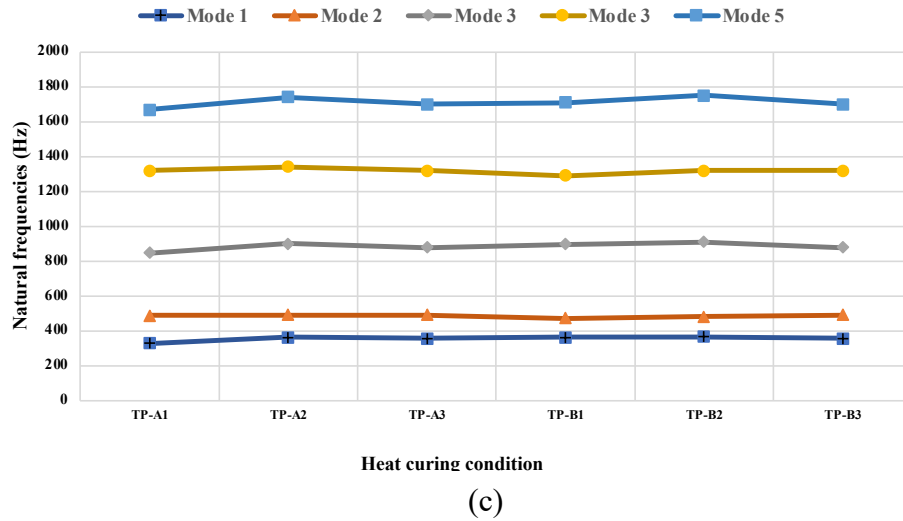
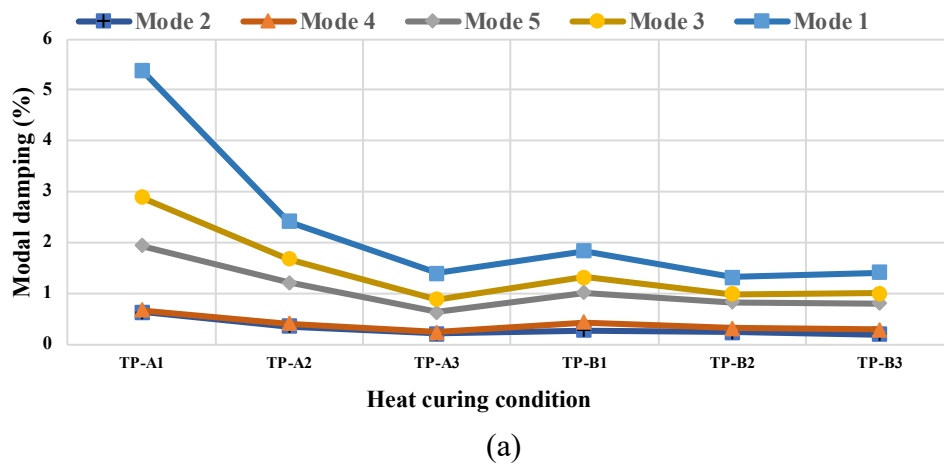
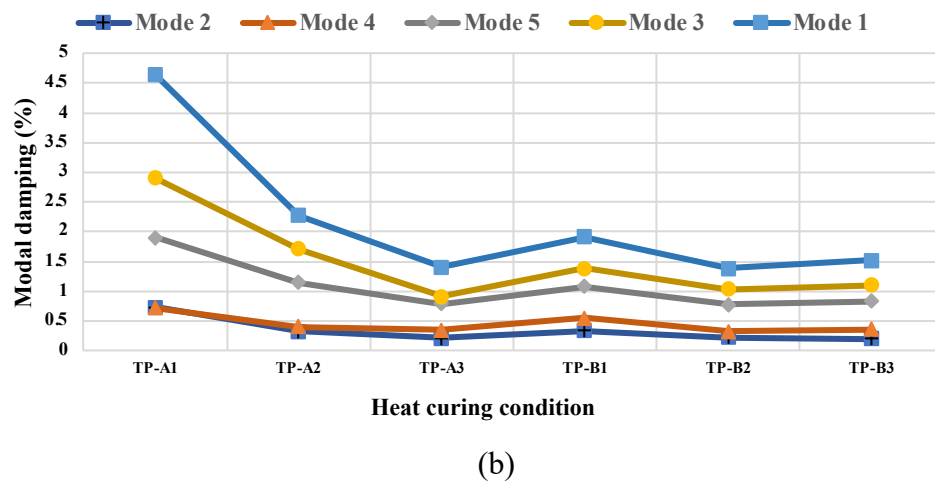


Fig. 2.23: Natural Frequencies of CFRP at a)100V 1.5A b) 150V 1.5A c) 200V 1.5A EDRM Condition (Different heat curing conditions)



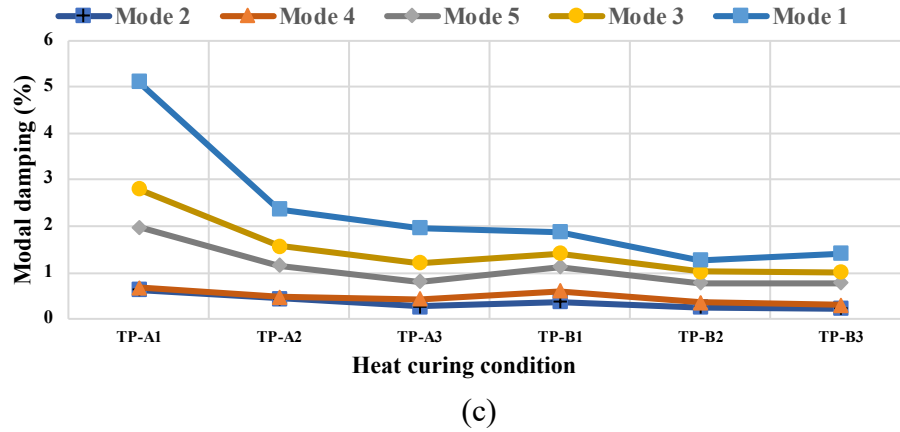


Fig. 2.24: Modal Damping of CFRP at a) 100V 1.5A b) 150V 1.5A c) 200V 1.5A EDM Condition (Different heat curing condition)

Figure 2.25 and 2.26 are showing a comparative analysis of the vibration properties of CFRP prepared by EDM and VaRTM. Figure 2.25 portrays the comparison of natural frequencies between the CFRP prepared by EDM and VaRTM. It can be seen from the figure that the natural frequencies of the specimen made by EDM is lower than the specimens made by VaRTM. The natural frequencies of specimens made from VaRTM is around 19%, 6%, 35%, 15% and 31% higher than the specimens from EDM for mode 1 through 5 respectively. In case of modal damping, completely different scenario was noticed from figure 2.25. Specimens made by EDM has high damping than those made from VaRTM. From mode 1 through 5, the EDM based specimen showed 164%, 77%, 183%, 95% and 166% higher damping characteristics than VaRTM based specimen respectively. In other words, CFRP prepared by EDM has lower natural frequency and higher modal damping compared to CFRP made by VaRTM. In spite of the fact that, both EDM and VaRTM are epoxy based, but the resin are not same. Consequently, a perfect comparison can not be possible. It is not possible to evaluate the resin directly since it is difficult to make a sample of the electrodeposition solution alone.

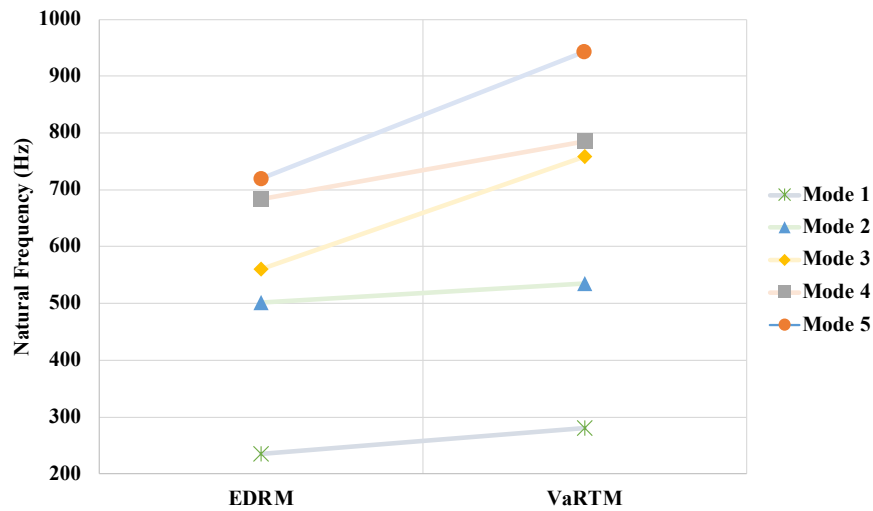


Figure 2.25: Natural frequencies of composites prepared by EDRM and VaRTM

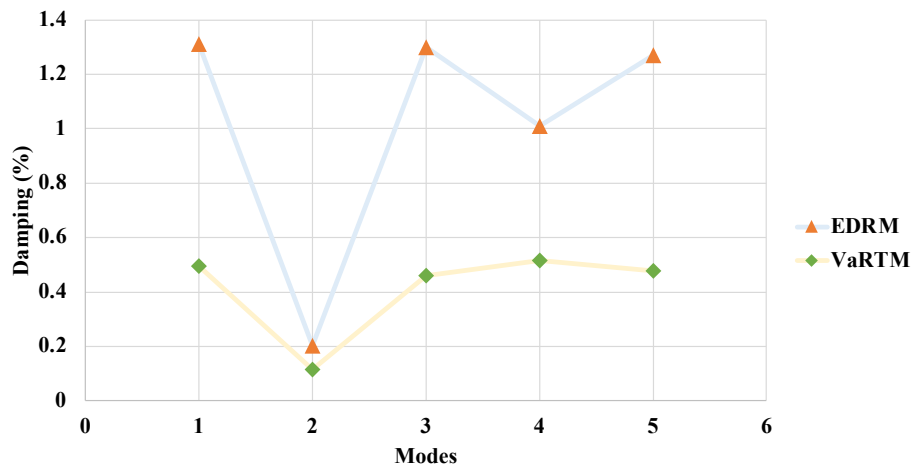


Figure 2.26: Modal damping of composites prepared by EDRM and VaRTM

2.3.3 Bending strength and bending modulus

By doing the four-point bending test, the bending strength and bending modulus of the material was evaluated. The number of specimens that were used for four-point bending test were 6. As the concentration varies depending on how often the electrodeposition solution is used, it is challenging to compare the time needed for electrodeposition by voltage. Fig. 2.27 shows the comparative value of bending strength with an error bar shown when the electrical current and voltage condition of EDRM was changed. As mentioned earlier, five different current conditions (0.50A, 0.75A, 1.00A, 1.25A and 1.50A) and three different voltage conditions

(100V, 150V and 200V) were used for the bending test. For the preform, only [PW0°]₈ was used. As can be seen from the figures, the changes in the bending strength for different voltages are not significant. On average, the values of 200V and 100V were higher than that of 150V. The maximum strength was achieved when 200V was applied and when the current condition was 1.50A. While observing the changes of values due to the changes of voltage and current condition, the heating condition was kept fixed. The heating time was 8 hours and the temperature was 230°C. Additionally, it can be noticed that the higher current condition had pretty higher value than the lower current condition. 1.00A, 1.25A and 1.50A current conditions had larger values than 0.50A and 0.75A of current conditions. It is because while using the higher current condition, the resistance becomes lower which makes the precipitation onto the fiber perfectly. From the foregoing, it was discovered that the voltage had little effect on how difficult it was for the material to deform in relation to the force in the bending direction but current have some effect on that regard. In addition, Fig. 2.28 shows the comparative value of bending modulus when the electrical current and voltage condition of EDRM was changed with error bars shown. It was noticeable from the figures that bending modulus was hardly affected by the change of voltage and the current condition of EDRM. The differences in the values were so small that it is inevitably insignificant.

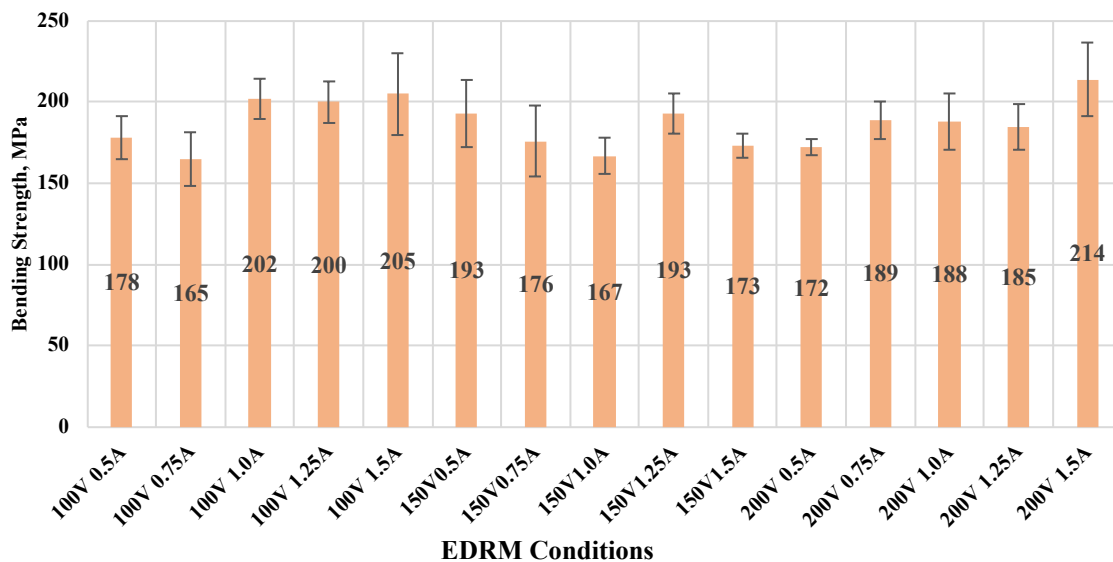


Fig. 2.27: Bending strength of CFRP on different EDRM conditions

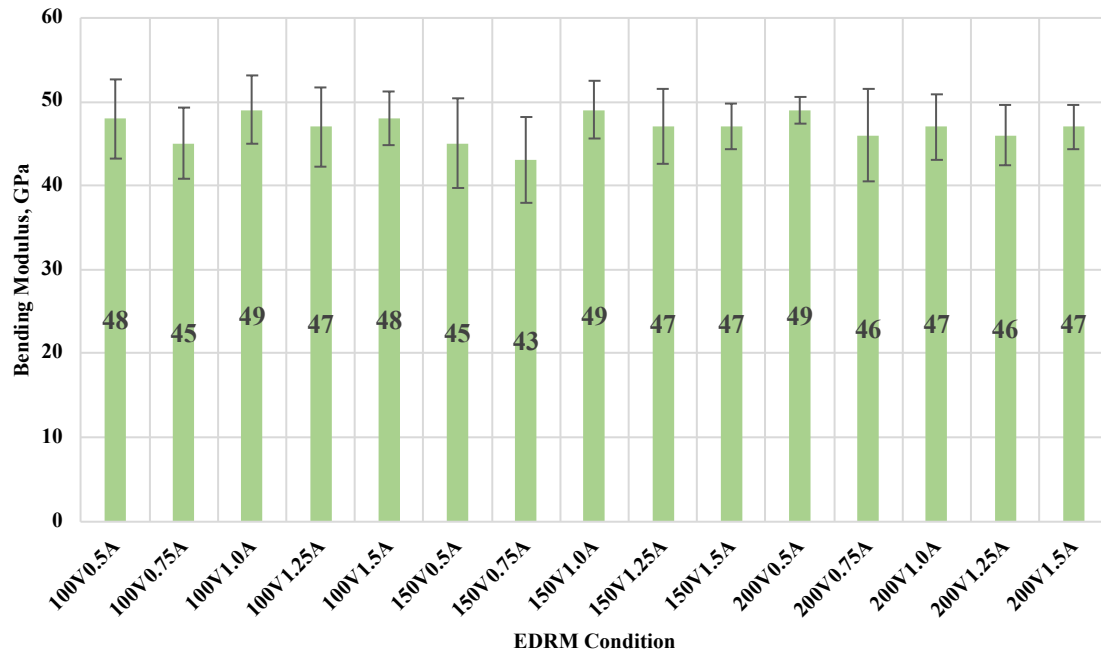


Fig. 2.28: Bending modulus of CFRP on different EDM conditions

A comparison was shown in Fig. 2.29 where the average values and error range of the bending strength of composites prepared from different heating time were compared. Fig 2.29(a) and 2.29(b) shows the value when temperature profile A and B were used respectively. It can be seen that the values were greatly affected by the change of heat curing period as well as temperature. The condition 215°C and 4 hours showed the lowest value while 230°C and 6 hours showed the highest value. In case of 215°C heating temperature condition, the value of bending strength was increasing when the specimens were kept in the incubator for longer period. For example, the average data of bending strength for 4 hours (215°C), 6 hours (215°C) and 8 hours (215°C) are 84 MPa, 131 MPa and 205MPa respectively when the EDM condition was 100V and 1.5A. However, in case of 230°C heating temperature condition, the maximum value was achieved when the specimen was kept 6 hours in the incubator for heat curing. For EDM condition 200V and 1.5A, the value for 6 hours (230°C) was 209 MPa while for 4 hours (230°C) and 8 hours (230°C) were 145MPa and 193 MPa respectively. Again, the heating temperature also has an impact on the flexural strength. As can be seen, the value became larger when the heating temperature was changed from 215°C to 230°C. Consequently, it is noteworthy that the high temperature of the heat curing can substantially increase the bending strength of the specimen but if it is kept for a longer period, it may decrease the strength. It is probable that high temperatures for longer time may weaken the fiber matrix

bond which results in poorer strength. The optimum time for heat curing is definitely 6 hours, which can be observed from the figures provided. Considering the position of the specimen, the lower part of the specimen showed good value compared to the upper part. It can be a fact that the lower part that is immersed deeply in the figure can achieve a good amount of precipitation. On the other hand, in case of bending modulus, the effect of the heat curing didn't seem very effective. The comparison was shown in Fig. 2.30. On average, the higher temperature as well as high curing period had little higher value, but the difference is negligible.

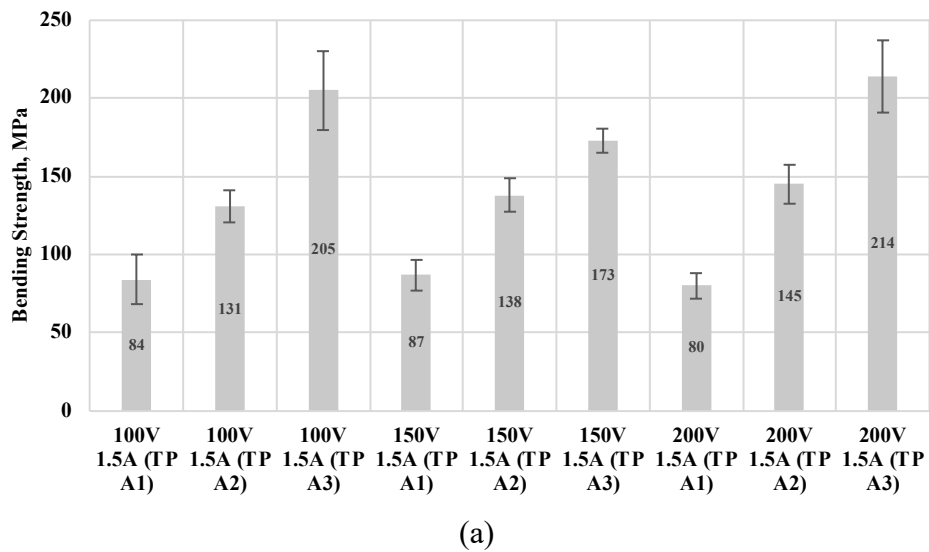
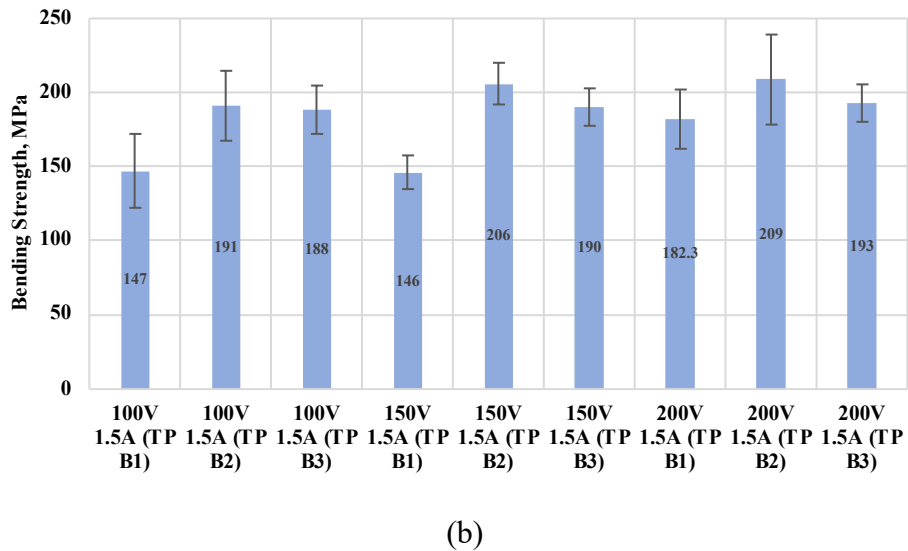
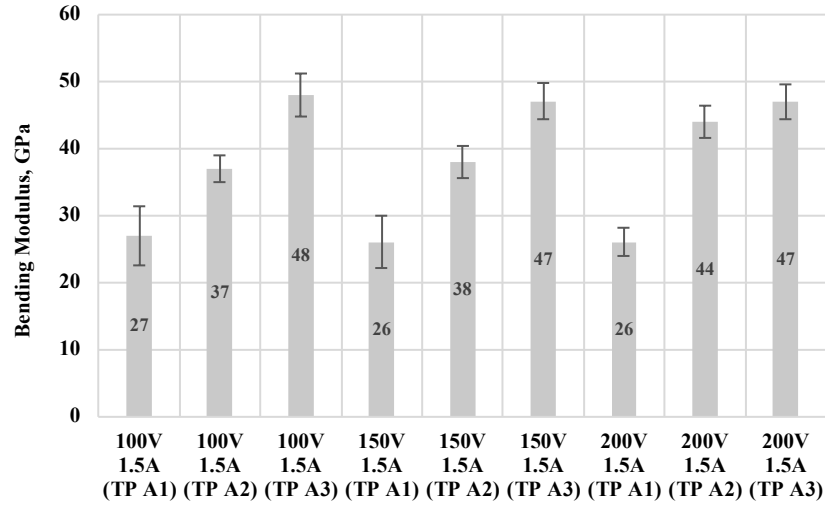
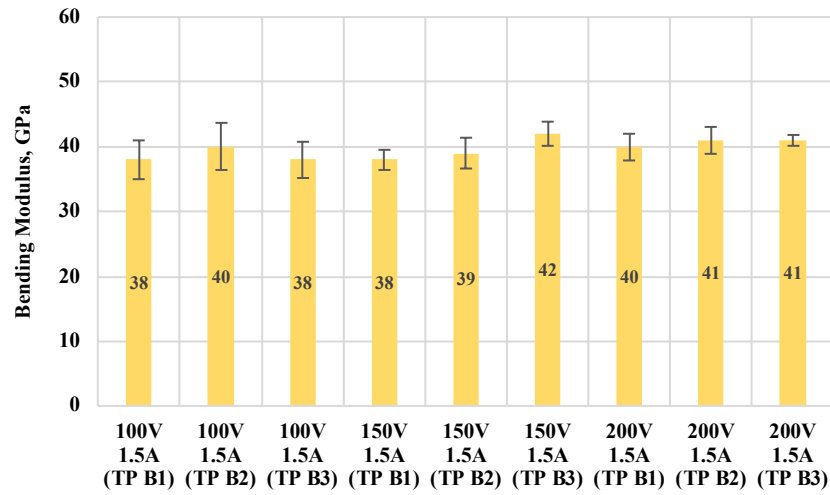


Fig. 2.29: Bending strength of CFRP on different heat-curing conditions, (a) using temperature profile A, (b) using temperature profile B



(a)



(b)

Fig. 2.30: Bending modulus of CFRP on different heat curing conditions, (a) using temperature profile A, (b) using temperature profile B

2.3.4 Young modulus

The young modulus of the carbon fiber composite was obtained by doing tensile test. For tensile tests, the number of specimens is 3, and Fig. 2.31 shows a graph in which the average value of the Young's modulus under each heating condition. A value twice the standard error was used in the positive and negative directions from the mean value. Furthermore, when comparing under heating conditions, the current was fixed at 1.50A, but three voltages were used. For heat curing, only 230°C was used but three different durations were used. In case of

tensile test, two different kinds of preform $[PW0]_8$ and $[TFP/TFP/PW45/PW0]_s$ were used. Fig. 16 shows a scattered values of the Young's modulus with different heating time. The error range is very small, and the values varied from 56 GPa to 69 GPa. The highest values were achieved for 6-hour heating conditions and 100V (1.5A) EDRM conditions while the highest values were achieved for 4-hour heating conditions and 200V (1.5A) EDRM conditions. The values didn't change that much for voltage change but changed little for heating duration. Nevertheless, tensile deformation is more likely to be caused by fibers carrying a larger proportion of the load than bending deformation. However, the young modulus of $[TFP/TFP/PW45/PW0]_s$ was much higher than that of $[PW0]_8$. The average Young modulus of $[TFP/TFP/PW45/PW0]_s$ was 95 GPa which was almost 37% higher than that of $[PW0]_8$. Albeit it can be argued that this technique is sufficiently tailored for real-world applications. If manufacturing process optimization and manufacturing parameter optimization are carried out in the future, it is believed that fabrication of CFRP with outstanding mechanical characteristics due to high efficiency is achievable.

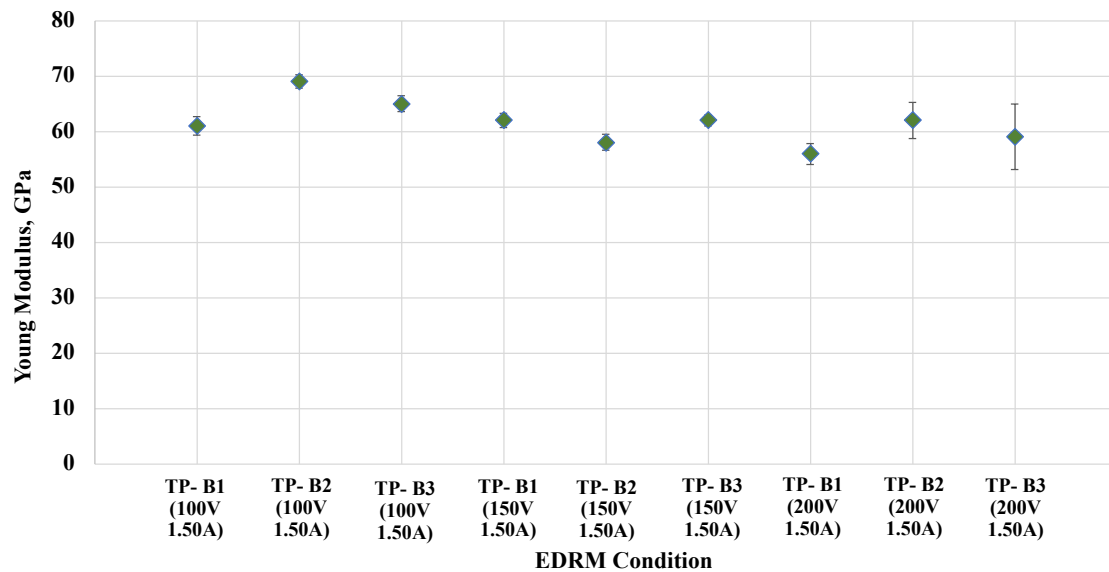


Fig. 2.31: Young modulus of CFRP at different heat curing conditions

2.3 Conclusion

This research systematically examined the effects of heating conditions and electrical energization parameters, specifically voltage and current on the static (flexural) and dynamic

(vibrational) properties of Carbon Fiber Reinforced Polymer (CFRP) composites fabricated via the Electrodeposition Resin Molding (EDRM) method. The overarching objective was to identify and optimize favorable EDRM process parameters that yield high-performance CFRP composites in an efficient and scalable manner. The results show that heat treatment had a significant impact on the final composite qualities among the several fabrication parameters evaluated, but many of the other EDRM variables showed just a slight sensitivity. According to this, thermal curing conditions are crucial in determining mechanical performance results, even if the EDRM process is resilient and tolerant to a variety of operating environments. It's interesting to note that CFRP composites made with various voltage-current combinations performed similarly, with no change in stiffness and mechanical strength. Nonetheless, the 1.5A current condition consistently produced the best bending strength, suggesting that this is the ideal electrical configuration for optimizing structural performance. Regarding thermal curing, it was found that composite properties acquired after 8 hours of curing under Temperature Profile A were almost identical to those obtained after 6 hours of curing under Temperature Profile B. From the perspective of productivity, this discovery is especially significant. In order to balance mechanical integrity and time efficiency, Temperature Profile B with a 6-hour heat treatment period is advised as the best thermal curing technique. Overall, this study demonstrates that the EDRM approach is a versatile, efficient, and energy-efficient way to create CFRPs and offers a clear understanding of how processing parameters affect the functional properties of the composite.

2.4 Future Studies

Although this study investigates the effect of the manufacturing variables of the EDRM on composite properties, further research is required to improve the effectiveness of this process. In order to evaluate their impact on microstructure, the quality of the fiber-matrix interface, and property uniformity, future studies should investigate a larger range of current and voltage values, including pulsed or ramping current modes. Thermomechanical analysis (TMA) or dynamic mechanical analysis (DMA) to evaluate how the composites behave under fluctuating temperature and load conditions which are crucial for aerospace and automotive applications can be conducted. The effect of the manufacturing parameters can also be investigated on the thermal or electrical properties of CFRP. By implementing all these optimum conditions, this method can significantly contribute to the simple and effective preparation of composites at an industry level.

Chapter Three

Preparation Curvilinear CFRP and evaluate its
vibrational response

Abstract (Chapter three)

This study investigates the influence of curvilinear fiber trajectories on the vibrational behavior of carbon fiber reinforced polymer (CFRP) composites. Using tailored fiber placement, three distinct fiber alignment patterns such as unidirectional, parabola, and sine wave were designed and fabricated to assess how fiber orientation affects dynamic performance. The composites were prepared using the Electrodeposition Resin Molding (EDRM) method, which allowed precise resin impregnation and uniform lamination across all models. Modal analysis was conducted through both experimental testing and finite element simulations, focusing on the first three natural vibration modes. The results revealed that while unidirectional laminates exhibited higher natural frequencies due to their stiffness along a single axis, the parabola and sine wave designs provided a more distributed deformation pattern and improved energy dissipation. Among the curvilinear models, the sine pattern demonstrated the most favorable vibrational response, with enhanced damping behavior and reduced stress concentration.

3. Preparation Curvilinear CFRP and evaluate its vibrational response

3.1 Introduction

The demand for lightweight and high-performance materials has significantly increased in recent decades, especially in sectors such as aerospace, automotive, wind energy, and robotics. Among the materials that have attracted considerable attention, Carbon Fiber Reinforced Polymers (CFRPs) stand out due to their exceptional specific strength, stiffness, fatigue resistance, and corrosion resistance [87-89]. Their adaptability to complex geometries and high load-bearing capacity makes CFRPs indispensable in designing both structural and dynamic components. However, with evolving performance requirements, particularly in dynamic loading and vibration-sensitive environments, traditional CFRP configurations especially those with straight (unidirectional) fiber paths are reaching their limitations [90].

In conventional laminated composites, fibers are typically aligned in straight lines to maximize strength and stiffness along specific directions. While effective in static applications, such unidirectional fiber configurations can suffer from poor performance in complex loading scenarios and are susceptible to interlaminar stresses, delamination, and vibration-induced failures [91]. This has led to the exploration of curvilinear fiber paths, where fibers are intentionally steered along non-linear trajectories such as parabolic or sinusoidal paths. These variable-stiffness laminate composites (VSLCs) have shown promising improvements in stress distribution, vibration damping, and structural stability under dynamic loads [92,93]. Pedro et al [94] looked on the impact of curvilinear path angles on both linear and nonlinear flutter behavior.

Curvilinear fiber placement enables the tailoring of mechanical properties across the laminate by controlling local stiffness and deformation behavior. The use of non-straight fiber paths such as parabola-shaped and sine wave inspired trajectories provides designers with an additional degree of freedom to influence vibrational characteristics, including natural frequencies, damping behavior, and mode shapes. These attributes are crucial in applications where vibration suppression, noise control, and structural integrity under cyclic loading are of paramount importance.

Advances in composite manufacturing technologies, particularly automated fiber placement (AFP) and tailored fiber placement (TFP) techniques, have enabled the precise implementation

of these curvilinear patterns. The AFP technique merges key advantages of two established methods, the differential tow payout characteristic of filament winding and the compaction and cut-restart abilities typical of automated tape laying (ATL) systems. During the fiber placement process, individual prepreg tows are compacted and bonded to the lay-up surface using a heated roller compaction system [95].

Recent studies have demonstrated that CFRPs with curvilinear fiber alignment offer improved vibrational response compared to traditional straight-fiber configurations [96,97]. The mechanical behavior of composite laminates with curvilinear fiber trajectories is strongly influenced by the geometry of the fiber paths, resulting in materials with non-uniform stiffness and directional anisotropy [98]. Martin and Leissa [99] proposed locally varying fiber volume fractions to enhance buckling resistance, applying the Ritz method for analysis. Hyer and Lee [100] utilized finite element analysis (FEA) to evaluate the strength and buckling behavior of plates with variable fiber orientations and demonstrated that such configurations can carry higher loads than plates with unidirectional fibers. Expanding on this, Gürdal et al. [101-103] proposed techniques to define arbitrary fiber paths by linearly varying the fiber orientation between two points, supported by both experimental and computational studies. Their findings confirmed that composites with curvilinear fibers possess enhanced mechanical performance compared to traditional laminates. Further, Honda and Narita [104] investigated the vibrational behavior of plates reinforced with quadratic fiber trajectories using the Ritz method and verified the superiority of spatially tailored laminates in dynamic applications. Honda et al. [104-108] had estimated damping characteristics of optimized curvilinear paths and assessed the vibration characteristics of CFRP that was prepared by using EDRM. Their studies found that by taking into account the strain energy ratios in each direction, specific damping capacity of a TFP layer can be determined. The damping characteristics of CFRPs with any fiber shapes can be designed with their determined damping parameters.

The ability to reduce vibration amplitude and shift resonance frequencies through fiber steering opens up new avenues for composite-based dynamic structures. Despite the growing interest in this domain, there remains a lack of comprehensive research on the comparative vibrational performance of CFRPs with different curvilinear fiber paths specifically unidirectional, parabolic, and sine wave geometries fabricated under similar processing conditions. While simulation-based studies have offered theoretical insights, there is a need for experimental validation through fabrication and dynamic testing of such laminates.

This present work basically focuses on fabricating nonlinear carbon fiber-based composite by using the EDRM method. An optimum electrical condition of EDRM was applied for fabricating the composites. The natural frequency as well as the modal shape of the composites of different curvilinear pattern had been obtained and compared with those acquired by finite element analysis. Understanding the influence of fiber path geometry on the vibrational response of CFRPs can lead to better design methodologies for lightweight, high-stiffness, and vibration-resistant components. This work contributes to the growing field of smart and adaptive composite materials, paving the way for customizable structural dynamics through geometric tailoring. The findings can be applied in aerospace structures (e.g., wings, panels), automotive body parts, sports equipment, and robotic arms, where dynamic performance is as critical as static strength. Moreover, this research aims to fabricate CFRP laminates with three distinct fiber alignment configurations unidirectional, parabola, and sine pattern using a Tailored Fiber Placement (TFP) machine. The goal is to evaluate and compare the vibrational characteristics of each laminate, focusing on natural frequency, modal damping and modal shapes. The broader objective is to determine how fiber trajectory influences dynamic performance, thereby guiding design strategies for vibration-critical applications in next-generation composite structures.

3.2 Methodology

In this study, a curvilinear CFRP had been prepared by using the electrodeposition resin molding (EDRM) method and its vibration properties were evaluated. Initially, the curvilinear preform was prepared by using the TFP machine, later the epoxy resin was impregnated through EDRM, heat cured and the sample for vibration test was prepared. The experimental output from the vibration test was later compared with the output of finite element analysis conducted by using Ansys APDL. The details of these steps are described in depth below.

3.2.1 Preparation of curvilinear preform

The model used in this study has a CFRP laminate with dimensions of 200 mm wide $\times$ 100 mm in height and Fig. 3.1 shows the Carbon fiber plain weave TR3110MS (Mitsubishi Chemical Corporation) that was used as base material as shown in 0° , 45° . The fiber shape described later is sewn in two layers by a fiber sewing machine to laminate two layers in the direction, and the base material side of the product is similarly produced. were made into 8

symmetrical stacks facing each other. As for the angle, the right direction of the paper is 0° and the counterclockwise direction is the positive direction. Three types of fiber shapes as shown in Fig. 3.5 were adopted, and each model was fabricated by electrodeposition resin impregnation. Since it has been reported that the thickness of curved fibers changes depending on the magnitude of the overlap ratio, which indicates the degree of overlap between the fibers, and affects the natural frequency of the fibers.

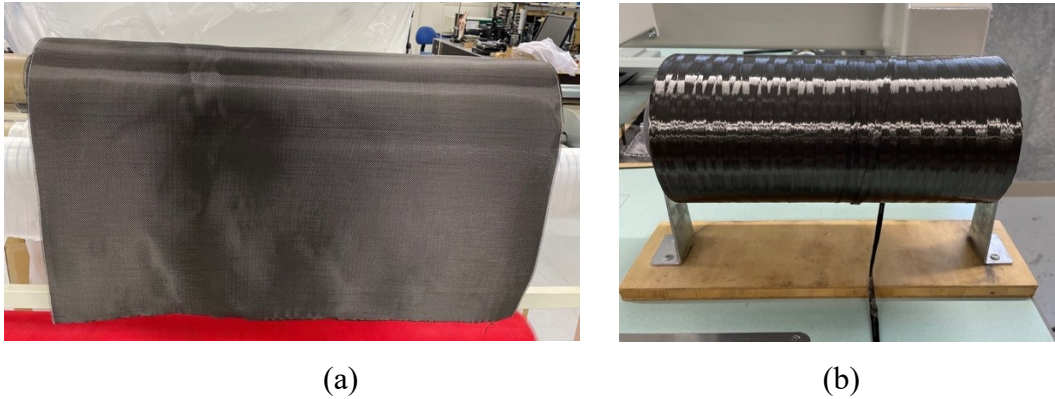


Fig. 3.1: (a) Plain-weave sheet, (b) Carbon fiber bundle.

The fiber shape was determined so that the overlap rate was small. Fig. 3.5(a) has a unidirectional, linear fiber shape and is called the unidirectional model. Fig. 3.5(b) has a fiber shape that replicates the parabolic shape passing through the three points of the center point of the plate and the corner of the plate in a short direction and is called the Parabola model. Fig. 3.5(c) is 25 with a period of 200 mm of the width of the board and an amplitude of one-fourth of the height of the board. It has a fiber shape that replicates the sin curve shape of mm in the short direction and is called the Sin model. The toe spacing of the model was 2.5 mm, which did not cause any gap or overlap with the adjacent tow. Just as the Parabola model has a tow spacing of 2.5 mm on the parabolic axis, the Sin model has a tow spacing of 2.5 mm at the peak of the waveform. The tow was arranged so that it became. In this study, laminated fiber material (preform), which is the stage before impregnation with resin, is prepared by a fiber sewing machine (Tailored Fiber Placement Machine). A fiber sewing machine is a machine that can sew carbon fiber bundles (CF tows) to base material such as non-woven fabric or carbon fiber plain weave material in a multilayered and local manner. It was developed by IPF Dresden and Tajima Kogyo Co., Ltd. in the 1990s. Figure 3.2 shows the fiber sewing machine TCWM-101 (Tajima Industries, Ltd.) used in this study. The CF tow used TR50S12L (Mitsubishi Chemical Corporation). As shown in Fig. 3.3, the CF tow is fixed by sewing the

upper and lower threads to the substrate in a zigzag pattern surrounding the tow. When sewing the tow, the fiber sewing machine can continuously change the fiber orientation angle due to the movement of the stage and the rotating head, so that the preforms with curved reinforced fibers can be produced. It is possible. The preform prepared in this study is shown in Fig. 3.7, Fig. 3.8 and 3.9.

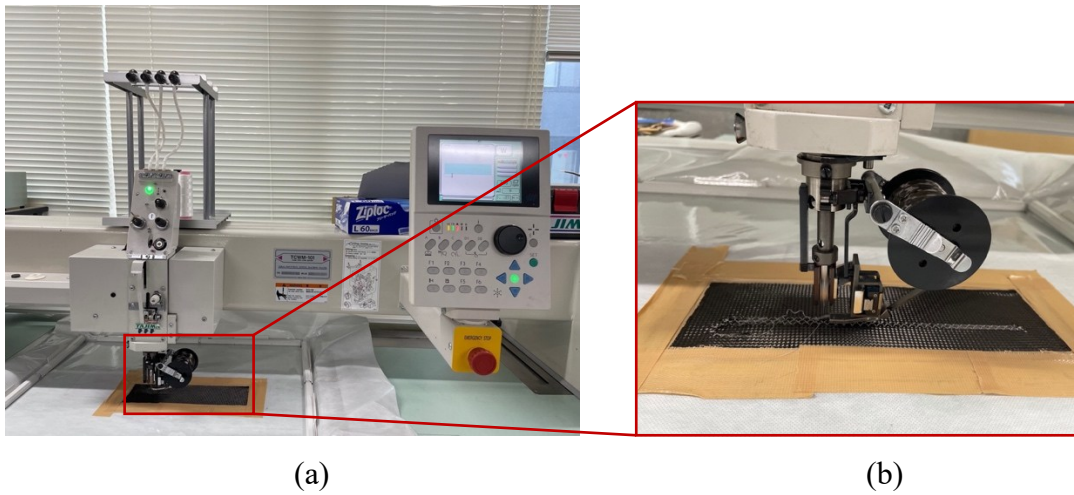


Fig. 3.2: (a) Tailored fiber placement (TFP) machine; (b) Head of TCWM-101

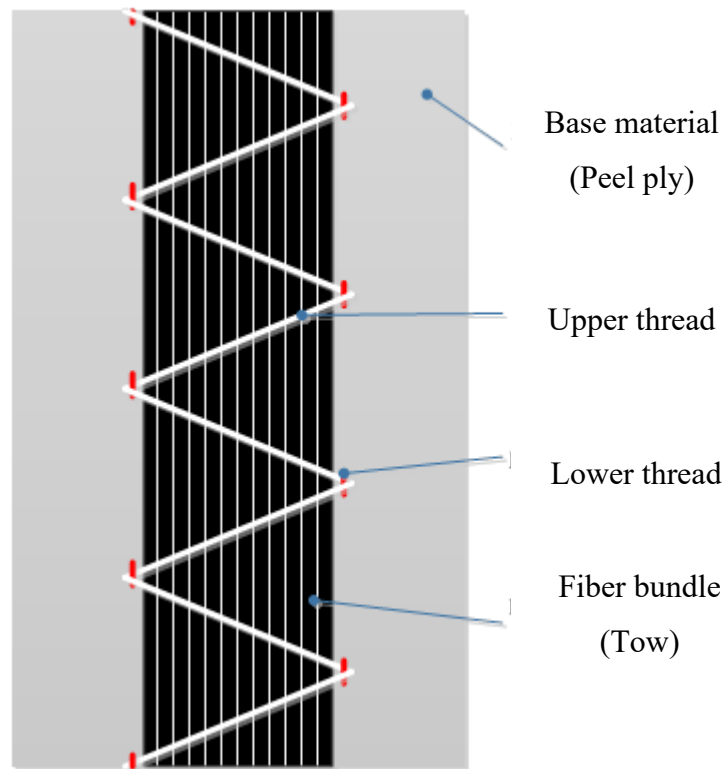


Fig. 3.3: Schematic diagram of fiber placement.

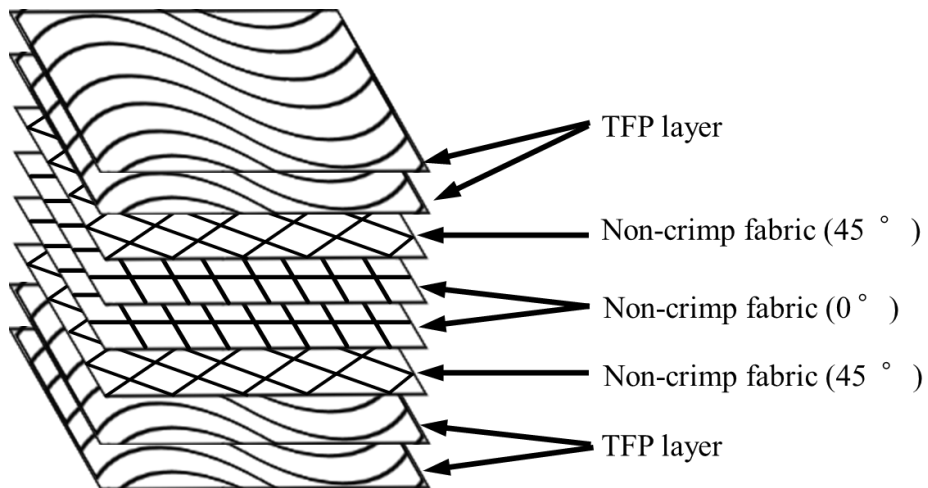


Fig. 3.4: Internal structure of 8 layers.

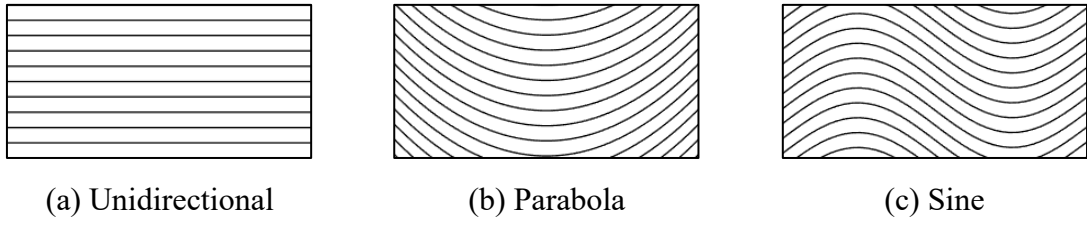


Fig. 3.5: Typical fiber shapes employed for the comparison.

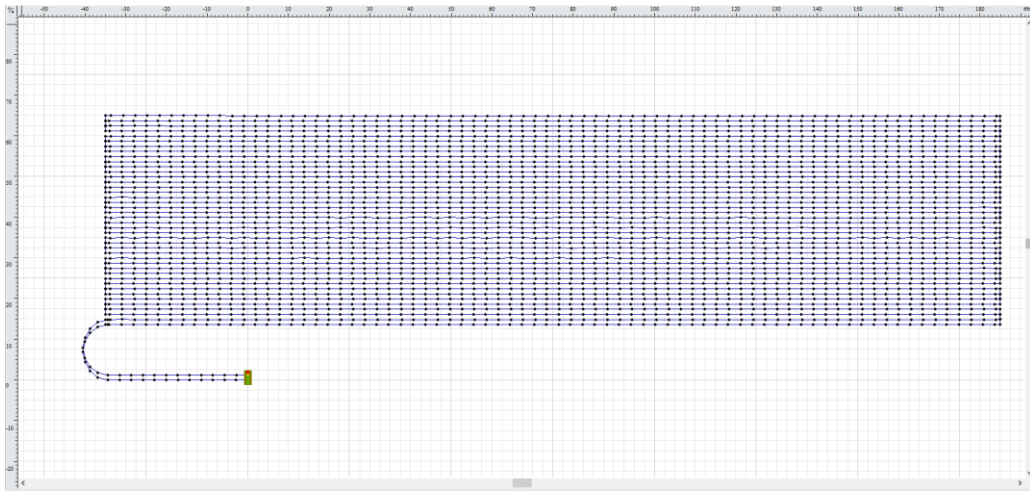


Fig. 3.6: Stitching data

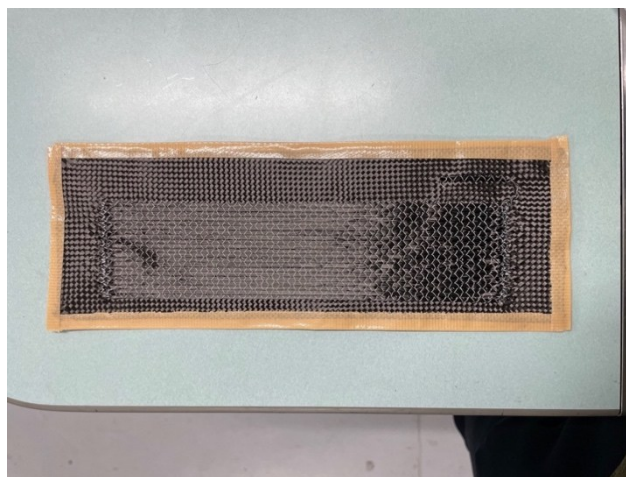


Fig. 3.7: Plain weaves preform fabricated by TFP.

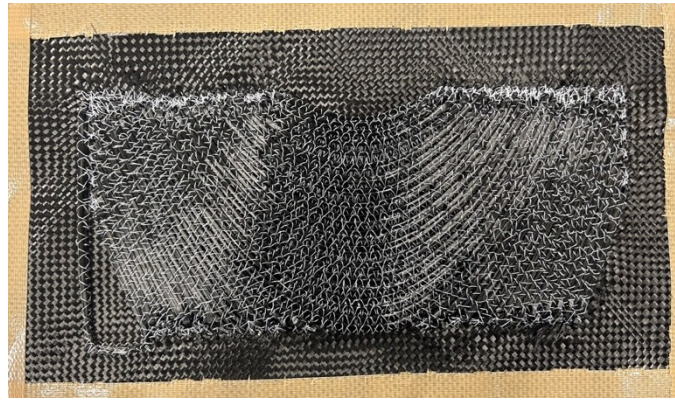


Fig. 3.8: Parabola shaped preform fabricated by TFP

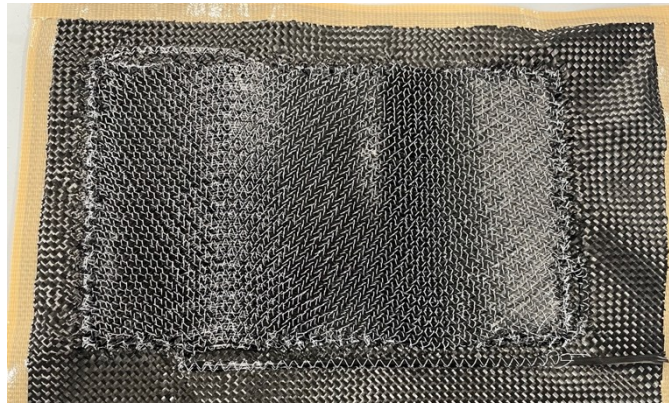


Fig. 3.9: Sine shaped preform fabricated by TFP.

3.2.2 Preparation of the sample

The electrodeposition method had been adopted to impregnate the resin onto the carbon fiber in this method. The carbon fiber was used as a cathode, and while the resin was contained in a steel meshed glass beaker. The steel mesh was connected to the positive side of the battery to make it as anode. Two different curve patterns had been chosen for this experiment: sine and parabola pattern. The first step was to prepare a curvilinear preform. The curvilinear preform was prepared by using a Tailored Fiber Placement (TFP) sewing machine. The fiber orientation that had been used in this experiment was a four layer of carbon fiber where the base two layers are plain-weave 0° and 45° respectively, and on top of the base layers, there were two layers of TFP ([PW0/PW45/TFP/TFP]). The two TFP layers were based on the pattern, for example sine

and parabola. After the preparation of the curvilinear preform, EDRM of the fiber was carried out. The EDRM methods were carried out following the procedure described in [article 1.2.3.1 Electrodeposition resin molding method](#), however, the beaker size was bigger in this case since the size of the preform is bigger. The experimental setup is shown in Fig 3.10, and the preform after the EDRM is shown in Fig. 3.11 (a-c).

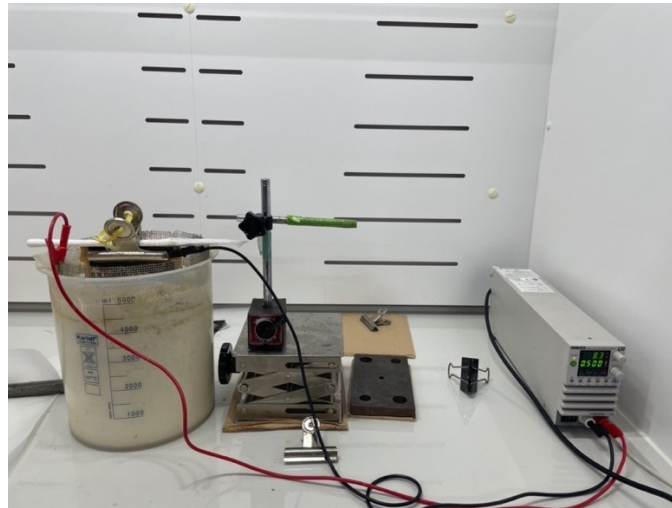
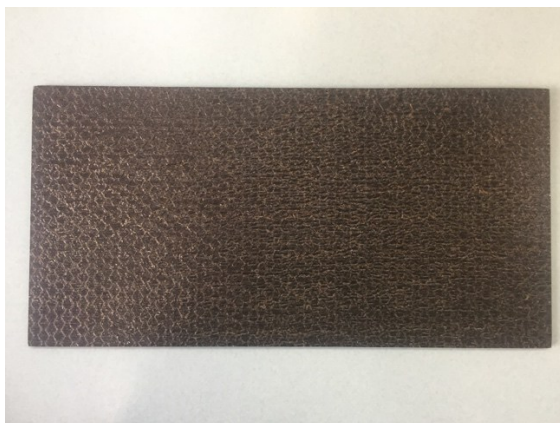


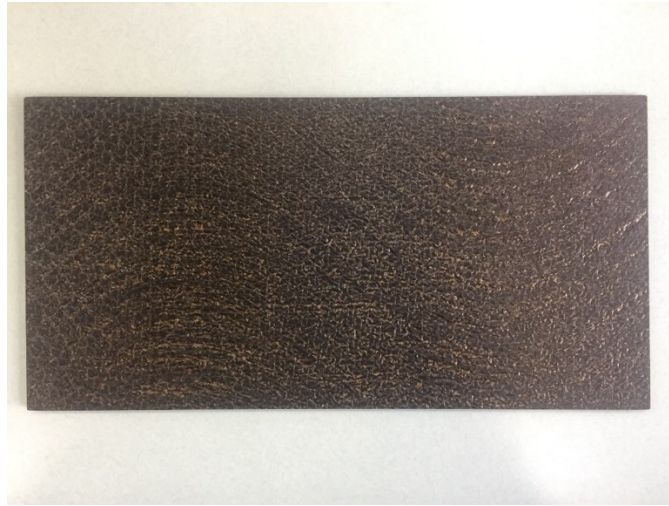
Fig. 3.10: Curvilinear CFRP resin impregnation during Electro-Activated Deposition Resin Molding.



(a) Unidirectional



(b) Parabola



(c) Sine

Fig. 3.11: CFRP made by Electro-Activated Deposition Resin Molding.

3.2.3 Vibration Test

The natural frequency and mode shape of the free vibration of the prepared specimen were determined by experimental mode analysis. The vibration test was carried out by the same as 2.2.5 Vibration Test. In order to fix a magnet-stand on the seismic isolation table and simulate the boundary conditions of free all around, the accelerometer 352C22 (PCB Piezotoronics) is attached to the specimen by hanging it from the stand using two rubber bands connected in series. Excitation is performed with a small impulse hammer 086E80 (PCB Piezotoronics). The accelerometer's response acceleration signal and the excitation force signal of the force sensor in the impulse hammer are sent to the spectral analyzer DS2000 (Ono Sokki Co., Ltd.). The hammering point and measuring points are shown in Fig. 4.12. D-conversion was performed, and high-speed Fourier transform, and data acquisition were performed using the general-purpose FFT analysis software DS-0221 (Ono Sokki Co., Ltd.). In the above way, the transfer function with each response can be obtained and modal parameters can be obtained. In this study, we adopted the roving hammer test, which is one of the single-point reference tests, as a measurement method, in which the position of the acceleration sensor is fixed at a node and the vibration by an impulse hammer is performed at multiple nodes. Although it is possible to fix the excitation point at one point and move the acceleration sensor, the principle of obtaining the response between the nodes is the same, so a simpler roving hammer test was used. To visualize the mode shape, we used the mode analysis software ME'scope VES (Vibrant Technology).

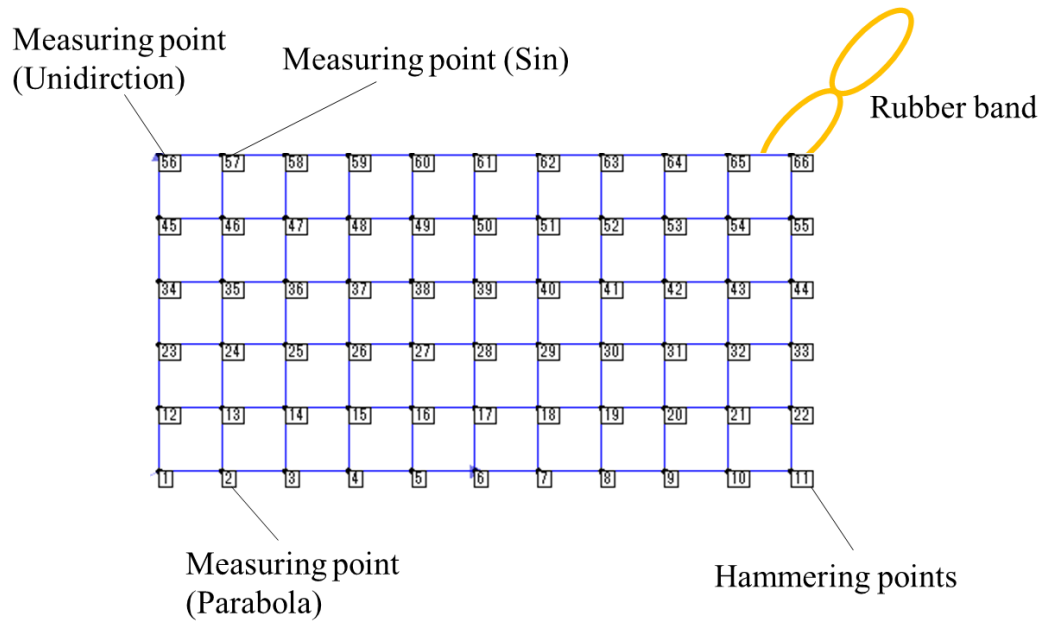


Fig. 3.12: Hammering points and measure points

3.2.4 FEM

In this study, ANSYS Mechanical APDL 18.1, a general-purpose finite element analysis software, was used for numerical analysis. Since the target of the analysis was the vibration characteristics of the thin plate, the element SHELL281 of the 8-node quadrilateral element was applied. The elements were analyzed with 200 elements divided into 20 sections in width and 10 in height, in a square with a side length of 10 mm. The base layer was expressed by specifying the orthotropic direction at 0° and 45° . For the representation of curved fibers of the sewing layer on FEM, by differentiating the mathematical formula representing each fiber shape and obtaining the derivative coefficient at the center coordinates of each element, the orientation angle of continuously changing fibers is expressed. The boundary conditions were free for all circumference, and the mode shape at the natural frequency and each natural frequency was obtained from the first order to the sixth order. The thickness of each layer was the result of cross-sectional observation. For the sewing layer, the thickness of the upper and lower two layers was averaged. For the density ρ , test pieces were prepared from the actual specimens, and the density obtained by the density measuring instrument was used.

3.3 Result and Discussion

3.3.1 Vibration Test

Figures 3.14 to 3.18 illustrate the first three vibrational mode shapes of carbon fiber-reinforced polymer (CFRP) laminates with parabola and sine-shaped fiber configurations. Each mode is shown side by side (a) representing the experimental observations and (b) showing the results from finite element analysis (FEM). These visual comparisons allow us to evaluate the dynamic behavior and verify the accuracy of the simulation model. The natural frequency and modal shape of the free vibration of the prepared specimen were determined by experimental mode analysis. In this experiment, two different sewing patterns had been chosen to make the curvilinear preform. The average thickness of the sine and parabola pattern were 1.85 mm and 1.62 mm respectively. Six modes had been extracted from both experiment and analysis. The absolute amount of out-of-plane displacement is color-coded in the contour diagram used to show the mode shape, and the displacement rises as it gets closer to red. As blue gets closer, the displacement gets smaller. Though all other modal shapes obtained from the experiment matched quite well with those obtained from the analysis, the third mode of the sine model had little difference. However, it was possible to confirm that both fiber models have a curved mode shape. In case of mode 1 of parabola model, both the experimental and FEM results show a symmetric bending shape, which is characteristic of a fundamental flexural mode. The deformation is centered and even spread, indicating that the parabola pattern allows good distribution of stiffness along the length. The close match between the experimental and FEM mode shapes also validates the reliability of the simulation. For mode 2, the structure begins to show more complex vibration behavior. There is a visible peak in the center and opposing movements on either side. This anti-symmetric deformation suggests a higher-order mode. The FEM analysis captures this transition very well, reflecting the influence of the curved fibers on how the laminate responds dynamically. In mode 3, the deformation pattern becomes more intricate, with multiple nodal lines across the surface. The fibers in the parabola layout appear to guide the vibration in a more dispersed way, reducing sharp gradients. Again, the experimental and simulation results are in good agreement, reinforcing the idea that the parabolic fiber path contributes to more stable and evenly distributed vibration modes. Additionally, for sine patterns, in the first mode of the sine-patterned laminate, a similar symmetric bending pattern is observed. However, compared to the parabola model, the vibration appears more diffused, likely due to the periodic nature of the sine fiber layout. The

FEM output mirrors this behavior well. On the one hand, in Mode 2 introduces asymmetry, where one half of the panel moves in opposition to the other. The sine wave alignment causes the stiffness to vary more noticeably along the surface, resulting in slightly more localized vibration in some areas. The FEM visualization clearly reflects these localized responses. On the other hand, for mode 3, the sine laminate shows multiple zones of movement, with the wave-like fiber pattern influencing the flow of vibration across the plate. The simulation again replicates this pattern closely. Overall, the sine configuration encourages a more flexible and energy-absorbing response than the parabola or unidirectional layouts.

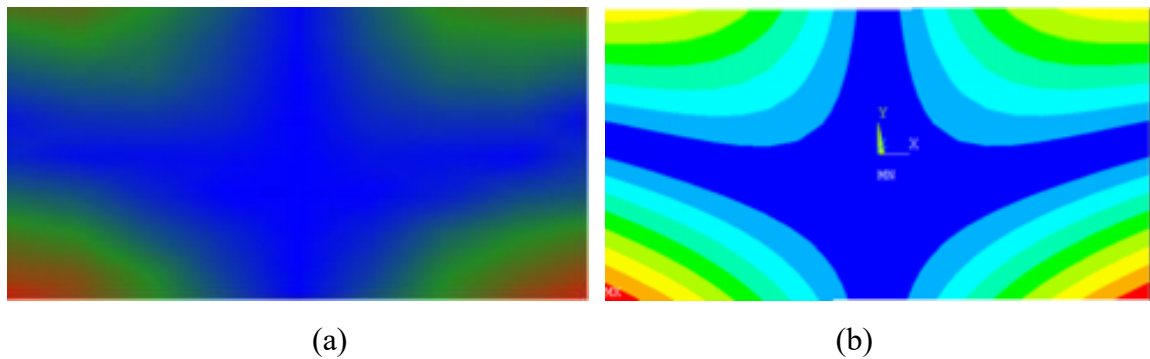


Fig. 3.13: Modal visualization of mode 1 of parabola model a) from experiment b) from FEM

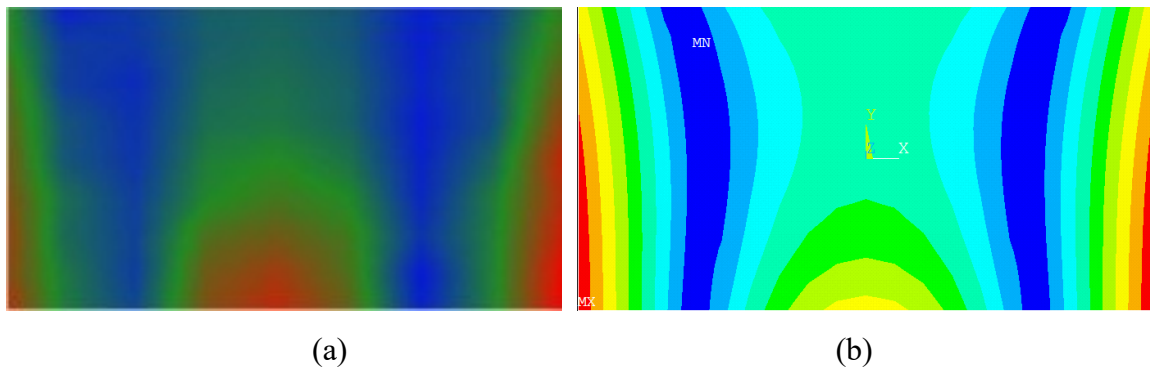


Fig. .14: Modal visualization of mode 2 of parabola model a) from experiment b) from FEM

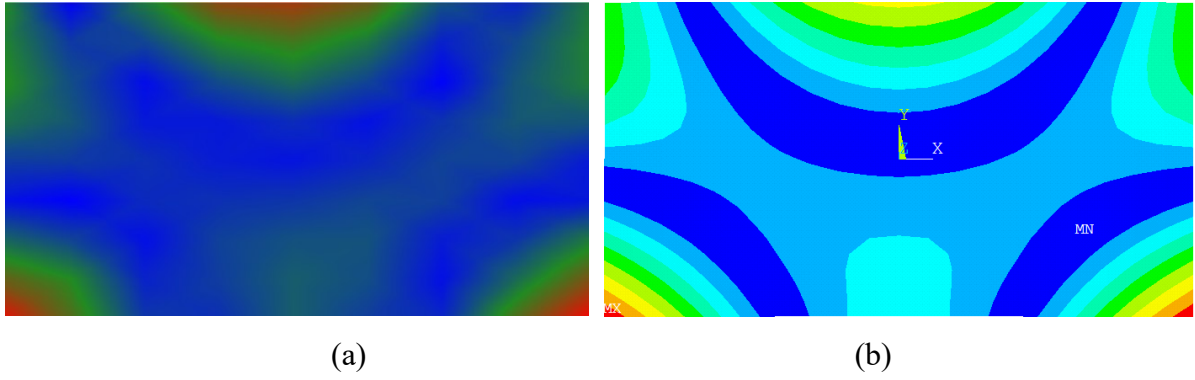


Fig. 3.15: Modal visualization of mode 3 of parabola model a) from experiment b) from FEM

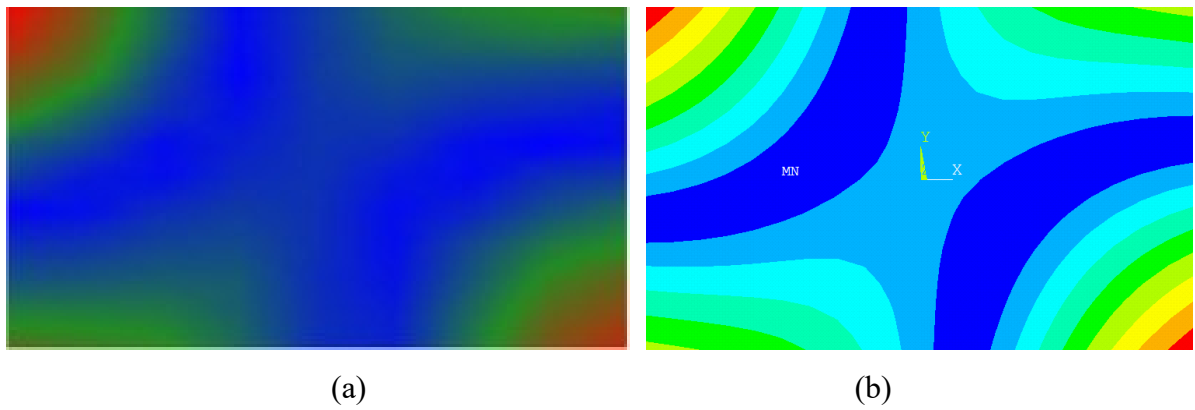


Fig. 3.16: Modal visualization of mode 1 of sine model a) from experiment b) from FEM

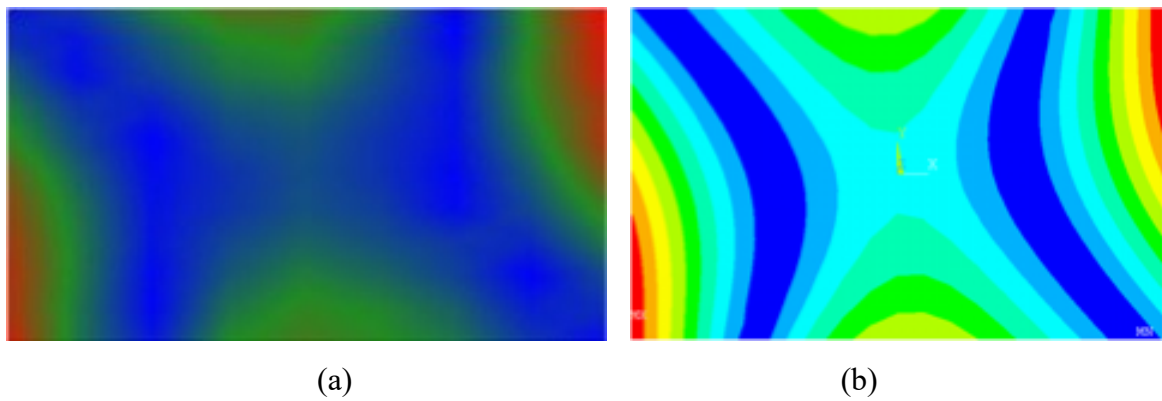


Fig. 3.17: Modal visualization of mode 2 of sine model a) from experiment b) from FEM

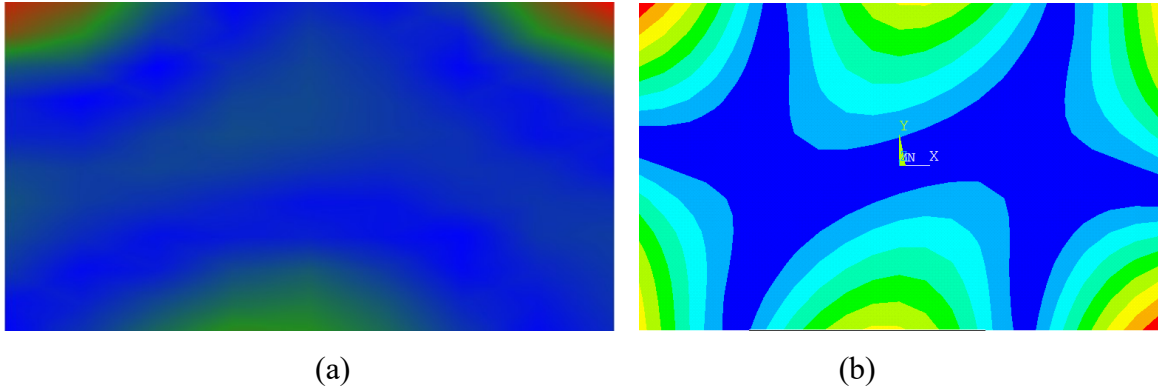
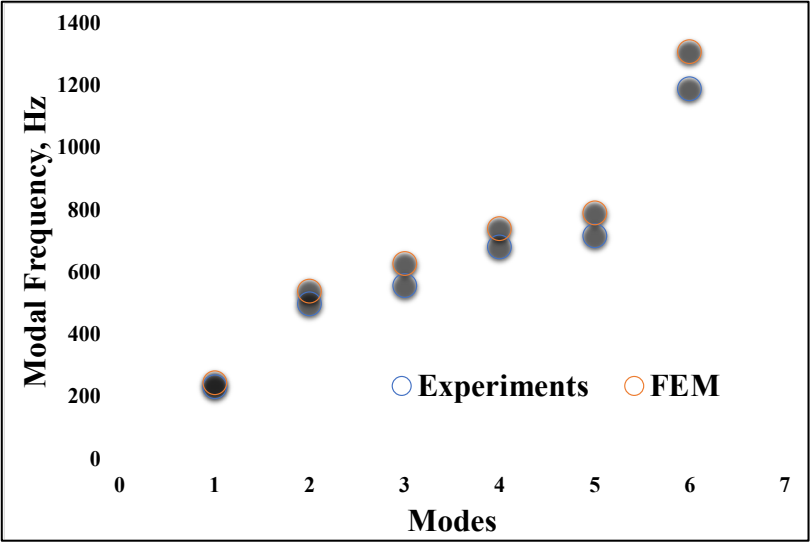
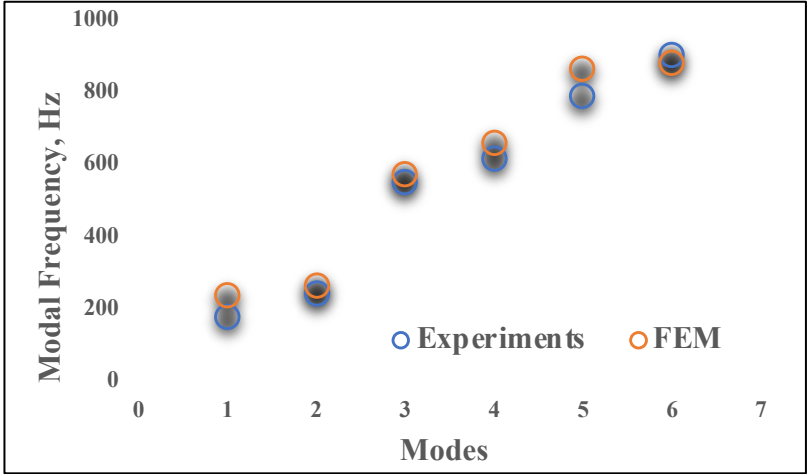


Fig. 3.18: Modal visualization of mode 3 of sine model a) from experiment b) from FEM

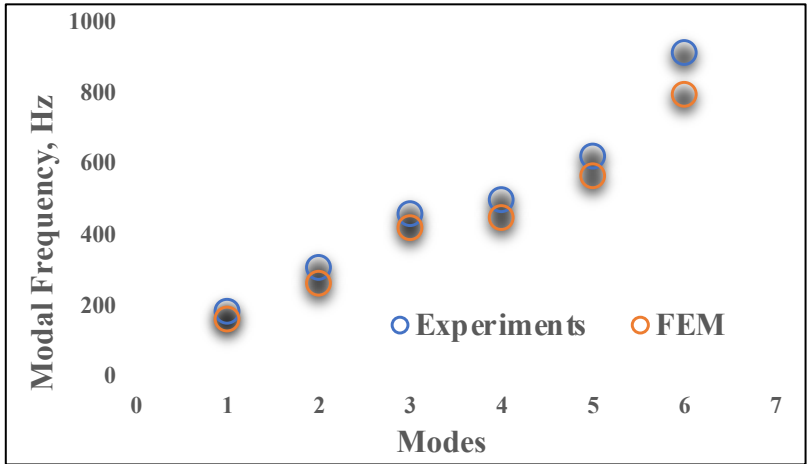
The natural frequencies of both the models are portrayed in Fig. 3.19 (a-c). The unidirectional laminate shows the highest natural frequencies, which is expected because the straight fibers offer the greatest resistance to bending along their alignment. This configuration is optimal for stiffness but may not perform well in terms of vibration damping. In addition, the parabola model falls in the middle, with slightly lower frequencies than the unidirectional version. The curved fibers cause the stiffness to vary gradually along the surface, which helps reduce vibration concentrations and spread energy more uniformly during dynamic loading. Moreover, the sine model exhibits the lowest natural frequencies among the three. This is likely due to its highly non-linear fiber path, which introduces more flexibility and makes the structure better at absorbing and dissipating vibrational energy. Such behavior could be advantageous in situations where vibration isolation is more important than maximum stiffness. The largest difference in the parabola model was as much as -24.6% and the lowest difference was -2.4% when comparing the natural frequencies of the experiment and analysis. For the sine model, it was 18.2% and 9.3% respectively.



(a)



(b)



(c)

Fig. 3.19: Natural Frequencies of a) unidirectional model b) parabola model c) sine model

3.4 Conclusion

This study aimed to investigate how fiber trajectory influences the vibrational behavior of carbon fiber-reinforced polymer (CFRP) laminates by comparing three different configurations: unidirectional, parabola, and sine. Through both experimental modal testing and finite element analysis (FEM), we evaluated the first three natural modes for each design, along with their corresponding natural frequencies.

The results clearly demonstrate that curvilinear fiber paths have a significant impact on how CFRP laminates respond to vibrational excitation. In the unidirectional model, fibers aligned in a single direction led to the highest natural frequencies, reflecting greater stiffness along that axis. However, the vibration energy in this case was more localized, suggesting that while stiffness is maximized, energy dissipation is limited, potentially making such laminates more prone to fatigue or vibration-related failure in dynamic environments. In contrast, the parabola and sine models, both with curvilinear fiber paths, displayed lower natural frequencies, indicative of greater flexibility. The parabola-shaped fibers provided a smoother transition of stiffness across the laminate, allowing for more even distribution of vibrational energy. This translated to well-defined, stable mode shapes that still maintained good structural performance while slightly reducing peak frequencies. The sine-patterned laminate, on the other hand, showed the lowest natural frequencies and the most flexible behavior among the three. The periodic nature of the sine wave fiber orientation promoted vibration damping by spreading deformation across the entire structure. This configuration also exhibited enhanced energy dissipation and reduced stress concentration zones, which are advantageous characteristics in structures subjected to cyclic or impact loading. Importantly, the FEM results aligned closely with experimental findings in all cases, validating the accuracy of the simulation model and supporting its usefulness for future predictive design.

Overall, these findings highlight the critical role of fiber orientation in tailoring the dynamic response of CFRP structures. While unidirectional fiber alignment remains ideal for applications where high stiffness is a priority, curvilinear designs particularly the sine pattern offer significant advantages in scenarios where vibration damping, energy distribution, and dynamic stability are more important. This research underscores the potential of fiber path

engineering as a valuable tool in composite design and opens the door for advanced, application-specific CFRP components, especially in fields like aerospace, automotive, robotics, and precision instruments where vibration control is crucial. Future work may further explore hybrid fiber orientations, optimization techniques, and material combinations to fine-tune these effects for even more specialized applications.

3.5 Future Scope

Future prospects may include fabricating three-dimensional structures and investigating vibration characteristics using EDRM and investigating the vibration characteristics of composite materials. To gain a more thorough knowledge of dynamic behavior, the study can be expanded to incorporate various matrix systems, lay-up sequences, and environmental factors (such as humidity or temperature). Lastly, the design of next-generation composite structures may be greatly improved by using machine learning and optimization algorithms such as particle swarm optimization or sequential approximation optimization techniques to forecast the optimal fiber pathways for loading scenarios.

Chapter Four

Preparation of CFRP capacitor by incorporating CNF

Abstract (Chapter four)

In this study, carbon fiber reinforced polymer, or CFRP is presented as a multifunctional material that combines capacitance, lightweight, and strength. In order to investigate the possibility of CFRP functioning as a structural capacitor, its electrical and mechanical performance were evaluated. The development of CFRP as a multifunctional composite is the main goal of this effort for lowering overall fuel and energy consumption. The dielectric materials employed in this investigation included cellulose nanofiber (CNF), paper, tissue, plastic, and jute fiber. Electrodeposition resin molding method (EDRM), a novel composite manufacturing technique, was employed in the composite's fabrication. Electrodepositions were used for applying current and deposit resin between carbon fibers. Following EDRM, the dielectric material was sandwiched between two carbon fiber preforms and heated to cure the material, creating the structural capacitor. To examine the capacitance characteristics, capacitance tests were carried out, while for the mechanical properties, tensile and four-point bending were performed. It was found that the use of CNF as dielectric had substantially enhanced the capacitance and bending properties of composites, however, the Young's modulus was decreased. CFRP that includes CNF has around 6% higher bending strength than CFRP that excludes CNF. Additionally, a maximum capacitance of 813 nF/m² was achieved by employing CNF into CFRP.

4. Preparation of CFRP capacitor by incorporating CNF

4.1 Introduction

One of the notable advantages of using CFRP is its multifunctionality. Multifunctional materials have innumerable benefits. For instance, wires can be eliminated by electrically conductive material, actuators may be eliminated by shape-morphing material, severe fire protection mechanisms are eliminated by a flame-resistant material. To put it another way, renewable material reduces the necessity for ongoing raw material extraction. Of course, a combination of these materials eliminates the need for all of these and possibly more, since this material combination frequently creates different functions that are not possible with just one of the components [110-112]. Francesco et al. [113] sandwiched prepreg of glass fiber reinforced plastic (GFRP) plies with piezoelectric nanofibers to create a self-sensing composite laminate. Because copper wires and carbon are inherently connected, their method eliminated the necessity for extra electrical cables. Sensor networks that use piezoelectric materials integrated into the composite materials can be used for building a self-sensing system. In order to create multifunctional structures (MFS), it involved a network of piezo-ceramic actuators/sensors inserted in a layer that was placed between carbon fiber-based epoxy composite laminates. Moreover, numerous thermal management systems with varying needs could make use of the multifunctional nano-fibrillated cellulose (NFC) based composite films, especially for uses like electrical insulation and electronic packaging that call for both thermal conductivity and electrical insulation [114]. Sukanta et al. [115] created structural composites based on conductive layers, using both CFRP and GFRP to guard against electromagnetic interference. Both structural composites were able to block electromagnetic interference radiation when a conductive layer based on doped polyaniline was applied. However, CFRP also has the potentiality of acting as a capacitor. Although capacitors are bulkier than other components like resistors, diodes, and transistors, they are essential energy-storing devices used in electrical circuits. Capacitors based on dielectrics are typically used in electronics because those based electric double layers are unable to function at elevated frequencies. The technique for preparing the CFRP as a capacitor, a multifunctional composite that combines strength, light weight, and capacitance is presented in this paper. The notion of composite structural power storage materials was initially presented by Chung and Wang [116], which led to the emergence of a new area of study for composites. Because carbon fiber is electrically conductive and has a high strength, CFRP can function as a capacitor following the basic

concept of a capacitor shown in Fig. 4.1. They made use of conductive carbon fibers for the electrodes that stored electric charge as well as for structural reinforcement. An electric capacitor of the parallel-plate type was constructed by inserting separator or dielectric films between neighboring carbon fiber laminas [117]. Under mechanical loading, they are vulnerable to interfacial damage [118]. Numerous research projects have been undertaken to improve mechanical and electrical performance of CFRP in order to get around the constraints and use them for multipurpose but as a single material [119].

Although the goal of this research is to make multifunctional and high-performing polymer composite capacitor material, the main purpose was to use cellulose nano fiber (CNF) for the enhancement of the electrical and mechanical characteristics of CFRPs. CNF will be used as a dielectric, whereas the carbon fiber will be used as electrodes. CNFs are environmentally friendly materials that can be made from raw wood. Along with being a good insulator, CNF also has great strength to weight ratio [120]. Few research has employed CNF to improve the mechanical characteristics of various resins [121, 122]. CNFs have been shown to improve the resin qualities. It was found in our previous studies that both the tensile strength and elastic modulus were greater than those of the CFRP specimen excluding the CNF sheet when the weight fraction of CNF sheet was 16%. As the weight percent of CNF sheet grew, the mechanical characteristics of CFRP specimens including CNF sheet improved as well, reaching 16%. Nevertheless, the mechanical characteristics of CFRP that has CNF sheet included declined when the weight percentage of CNF sheet reached 29% [123]. Gaurav et al. [124] looked into the impact of reinforcing polyvinyl acetate (PVAc) adhesive with CNF and cellulose nanocrystals (CNC). Elastic stiffness as well as the shear strength of PVA joints were greatly increased by the addition of nanocellulose to PVAc adhesive. Furthermore, Carlson et al. [125] also looked into how the type and thickness of the dielectric material affected the composite properties. As dielectrics, they employed paper with varying surface weights, polyamide (PA), polyester (PET), and polycarbonate (PC) films. Their studies found that the interlaminar shear strength increases with paper thickness. Furthermore, compared to the thickest paper, the interlaminar shear strengths of PA, PET, and PC films were approximately 22 MPa, 20 MPa, and 10 MPa, respectively. Conversely, adding CNF to CFRP can enhance not only the bending and tensile qualities, but also its thermal and dielectric properties and dimensional stabilities [126-128]. While there are many benefits to utilizing CNF, there are also certain drawbacks that should not be disregarded. Because CNF is hydrophilic, moisture is absorbed by the CNF-reinforced polymer, which could lead to poor mechanical performance.

Moreover, handling CNF dispersion is more expensive than handling other fillers. Again, it's critical to apply the optimum amount of CNF to the composites because too much of it can degrade the composite's qualities [125] and make production more challenging [129]. Therefore, for overall performance, the optimal amount of CNF is necessary. Notwithstanding certain drawbacks, CNF's excellent mechanical qualities, good insulation qualities, improved thermal stability, and high strength-to-weight ratio make it suitable for use in lightweight structural capacitor applications.

Although many researchers had prepared structural capacitors by employing different dielectrics, the inclusion of bio-based nanomaterials such as CNF into structural capacitors to increase its multifunctionality is yet to be explored. While delivering both structural integration and electrical energy, CFRP capacitors do not require additional space to fulfill their functions, unlike typical capacitors. This structural capacitor's use of CNF makes it feasible for biodegradable robots or green aircraft where eco-friendly design is required. As a result, there is a great need for structural energy storage systems based on nanofibers. Additionally, different manufacturing techniques or prepreps had been used for fabricating the structural capacitor till now, however, this study adopted a new manufacturing technique called electrodeposition resin molding (EDRM) technique that had been developed and established in earlier studies [130-133]. Epoxy resin is used as polymer in this method, but it may require hydrophobic treatment since CNF is hydrophilic [134, 135]. However, hydrophobic treatment was not required in this case to apply CNF to the CFRP surface because the electro-activated deposition solution (epoxy) was water-based. This was a major reason to choose CNF as dielectrics in this investigation, and the dispersed CNF was applied repeatedly to the surface of the carbon fiber preform that had been resin impregnated. Since each fiber is coated with epoxy, EDRM should theoretically have capacitance without insulators, but sadly, this did not occur due to the failure of epoxy to be a separator and led us to use dielectrics. Manufacturing is the primary obstacle. As previously discussed, an optimum weight percentage of CNF dispersion is required to have a good composite quality. In addition, a good dispersion on the matrix is also necessary for the correct homogeneity of CNFs yet van der Waals forces cause them to tend to stay together in bundles [136]. Furthermore, as CNF acts as an insulator in this research, a precise control of the dielectric thickness for separating the electrode was quite challenging. In order to contemplate the performance of CNF as insulator in CFRP capacitor, this study compared CNF with other dielectric materials, including plastic, jute fiber, tissue paper, and bond paper, as dielectric separator layers. The mechanical and electrical attributions

of each dielectric material was described, and the comprehensive multifunctional performance of the composite was assessed.

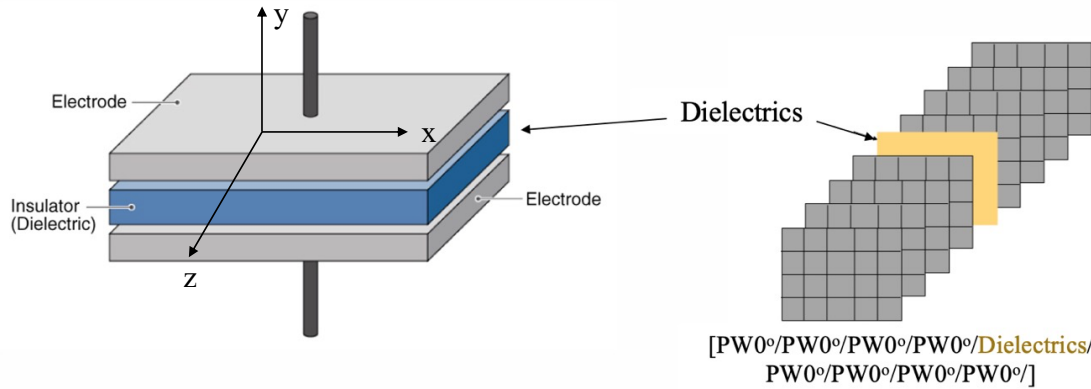


Fig. 4.1: The left figure is a basic structure of a capacitor, and the right figure employed the basic structure for CFRP where the carbon fiber is the electrode and dielectrics are the insulator [110]. Two preforms containing four plain weave (PW) carbon fiber layers are on both sides of the dielectrics to make the structural capacitor.

4.2 Methodology

4.2.1 Material

In order to prepare structural capacitors, carbon fiber reinforced polymer had been used as an electrode and different dielectrics materials had been used as an insulator. As for electrodes, carbon fiber plain-woven (PW) material (TR3110MS - Mitsubishi Chemical Corporation) and epoxy (Elecoat AR cosmetics - Shimizu Co., Ltd.) were used for fiber and matrix respectfully. As for insulation, tissue paper (kitchen towel), bond paper (A4 printing paper), plastic (Securlon® L-1000, Airtech), jute fiber (woven) and cellulose nano fiber (Daio Papers Co. Ltd., Japan) were used. The CNF (2wt%) used in this research was obtained from broad-leaf wood, had a diameter of about 20 to 80 nm. Different ways of CNF application to the CFRP were followed to find the suitable one that meets the objective. Tissue paper, bond paper, plastic, jute fiber, and CNF have dielectric constants of about 2~3, 3~5, 2~4, 3~6 [137-140]

and 4~10 [116]. These dielectric materials were selected in order to compare the performance of CNF dielectrics because they are readily available, reasonably priced, and exhibit strong electrical insulation qualities, all of which align with our goals.

4.2.2 Preparation of preform and dielectrics

The experiment was started with the preparation of the preform. Four layers of carbon fiber, each measuring 200 mm by 60 mm, were cut and attached together with a tape. Two similar preforms were used for preparing each sample, and each preform was gone through EDRM separately. However, dielectrics preparation was a crucial step in this research as capacitance greatly depends on this. For dielectrics such as tissue paper, bond paper, jute fiber and plastic (shown in Fig. 4.2) were cut as the same size of the preform and placed in between two preforms after EDRM. In the case of CNF, six different ways were followed which are given in Table 4.1 with description. For the preparation of sample A to sample D, the same procedures were followed except the CNF application method. Initially, the preform with four layers of carbon fiber was run through EDRM. Then, epoxy was applied to another preform that had the same carbon fiber layers through EDRM. CNF was applied on the first preform and the second preform was put on top of it. Sample A was prepared in this way. While pressing the second preform on top of the first preform, it was noticed that some CNF was coming out of the preforms. In order to keep the CNF into the preform, the CNF was dried for 1 hour (Sample B) and 1 day (Sample C) before putting the second preform on it at room temperature (25°C). In Sample D, CNF and epoxy were mixed together to make a mixture (see Fig. 4.3) and the mixture was inserted into the specimen following the same procedure followed in Sample A. The method for creating the CNF film used in Sample E and F is shown in Fig. 4.4. The mixture of CNF and epoxy that were prepared earlier was poured between two plates and applying the doctor's blading method, the mixture was handled uniformly. After that, the mixture was left for a full day at room temperature (25°C). That is how the CNF film was prepared. To examine the potential effects of moisture content and interface quality on the overall performance of the composite, a different method for applying CNF into CFRP was used. While fresh and wet CNF was put in sample A, samples B and C had dried CNF at separate times for better handling and moisture removal. This is because wet CNF tends to slide up when compressed. Consequently, sample A had the highest moisture content whereas sample C had the lowest. To create a distributed dielectric, epoxy and CNF were combined in sample D. Additionally,

sample E had a distinct dielectric layer for insulation in the form of a dry CNF film. Additional plastic films were placed within sample F to shield the CNF film from extreme heat and pressure. The dielectric layer was placed in between two preforms to form $[PW0^\circ/PW0^\circ/PW0^\circ/PW0^\circ/DIELECTRIC/PW0^\circ/PW0^\circ/PW0^\circ/PW0^\circ]$ orientation. Dielectric layers such as paper, plastic, tissue, jute fiber and CNF film had respective thickness of 0.087 mm, 0.068 mm, 0.220 mm, 0.994 mm and 0.056 mm. Additionally, normal CFRP without any dielectrics layer having an orientation of $[PW0^\circ]_8$ was prepared as well for comparison with CNF-induced CFRP.

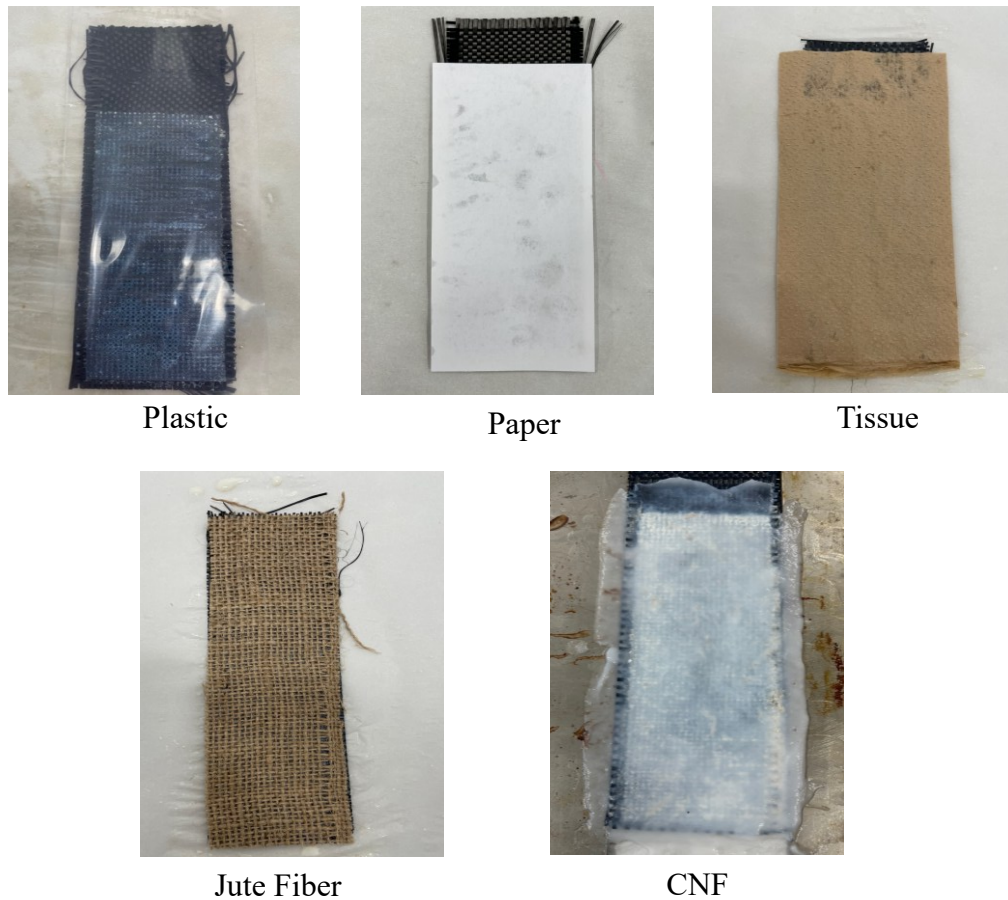


Fig. 4.2: Different types of dielectrics that had been inserted into the CFRP for capacitor preparation

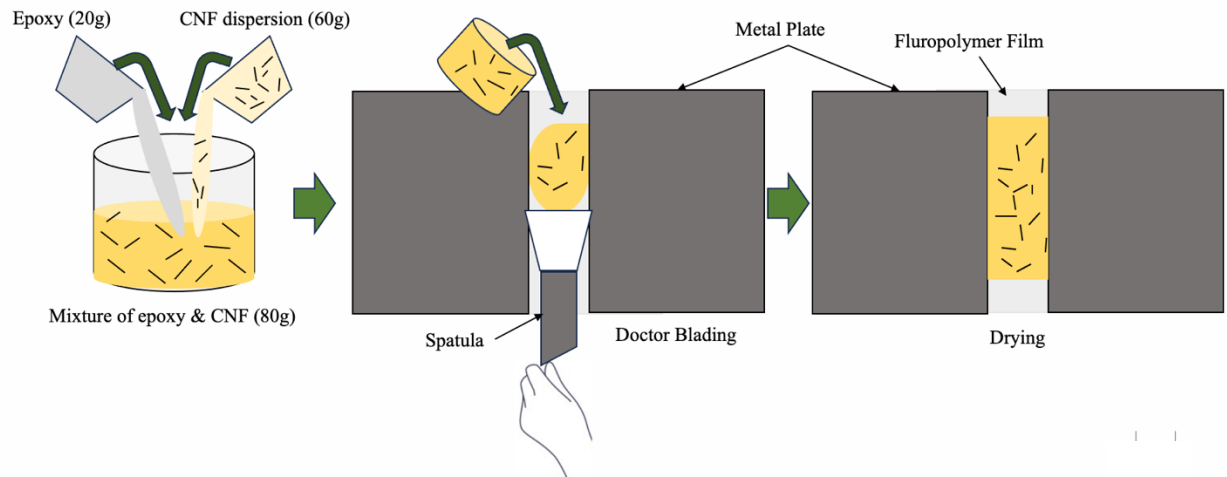


Fig. 4.3: Preparation of the mixture of Epoxy and CNF by adopting doctor blading method to have a uniform thickness throughout.

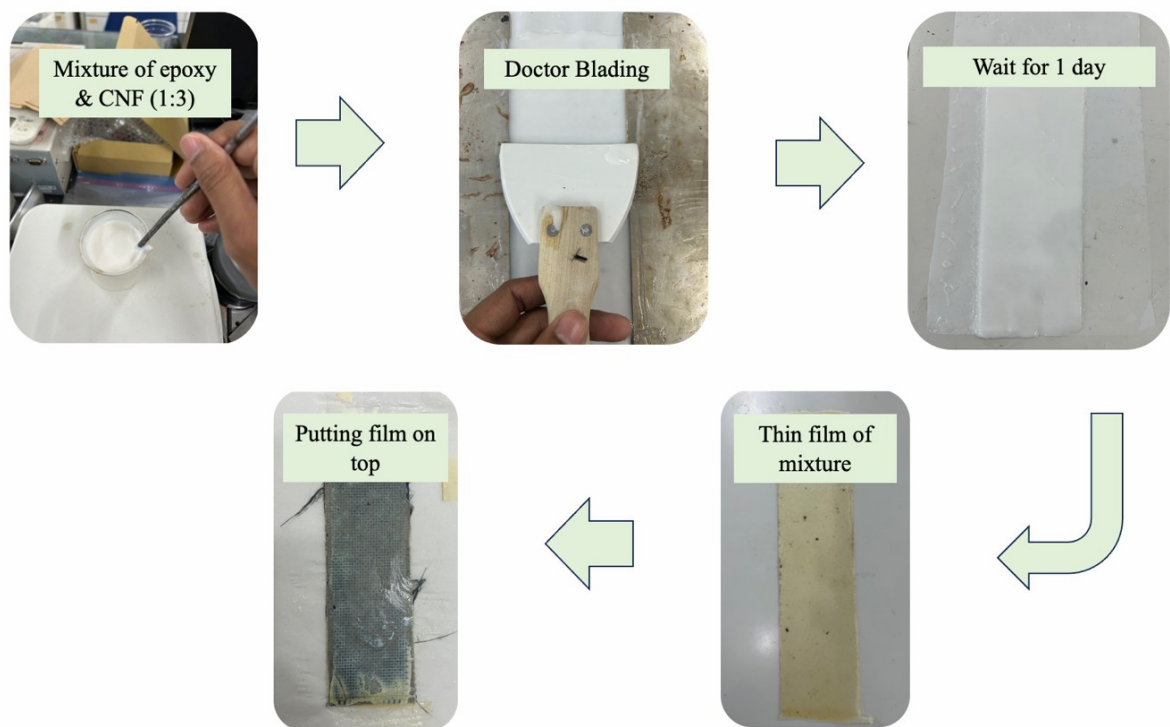


Fig. 4.4: Preparation of CNF film from the epoxy-CNF mixture by drying and curing

Table 4.1. CNF application methods and sample descriptions.

Samples	Dielectric preparation types
Sample A	CNF is directly applied on the top of preform after EDRM

Sample B	CNF is directly applied on the top of preform and waited for 1 hour at 25°C
Sample C	CNF is directly applied on the top of preform and waited for 1 day at 25°C
Sample D	A mixture of CNF and epoxy is applied on the top of preform after EDRM
Sample E	A film made by mixing CNF and epoxy (3:1) is placed between two preforms
Sample F	Two plastic sheets are placed between two preforms and CNF film is placed between the plastics

4.2.3 Sample preparation

After the preparation of preform and dielectrics, EDRM of the preform was carried out. The graphical representation of EDRM along with an actual set-up was portrayed in Fig. 1.6 and 1.7. Here, in the process of EDRM, the fiber acts as a cathode and is immersed into epoxy (electrodeposition solution). So, when the power supply is turned on, the resin starts to precipitate on carbon fiber surface with an increase of resistance. After a while, the electrical current converges with the increase of resistance as the voltage is kept constant when it reaches maximum value. Convergence of the electrical current signifies that the precipitation is complete. Then it is heat-cured and thermoset. That is how CFRP is made by using this method. The mechanism of EDRM is shown in Fig. 1.8. The details of the EDRM process is provided in article 1.2.3.1 Electrodeposition resin molding method. 165V and 0.5A were used as the voltage and electrical current conditions for the fabrication process. Fig. 4.5 illustrates the relationships among voltage, current and resistance with time under conditions of 165V and 0.50A. The left scale shows the correlation between voltage and time, while the other side illustrates the relationship between current and time as well as resistance and time. It can be observed that after approximately 400 seconds, the voltage reached its maximum of 165V and remained constant thereafter. Up until the voltage hit its maximum, the current was fixed at 0.50A and dropped to 0.03A with the increase of resistance. The resistance increased to around 6500 ohms. The current converges after around thirty-one minutes, signaling the completion

of precipitation. The minute change in the electrical current indicates that the fiber surface precipitation has reached its peak.

After the EDRM of both of the preforms are finished, the dielectric was inserted between two preforms as a sandwich pattern. Other ways of inserting insulators were also adopted as refer to Table 3.1. However, after completion of both EDRM and dielectric insertion, it was put inside the incubator for heat treatment. Before the heat treatment process, it was pressed by using a press machine, and the whole specimen was kept in between two fluoropolymer films following that. All of these were then put inside of two heavy metal plates. The temperature for heat curing was followed by author's previous study [132-135]. The maximum temperature for the heat treatment was 230°C. Initially, in the first two hours, the temperature had risen to 230°C from room temperature (25°C). For the next four hours, the temperature was maintained at constant temperature. After the heating was finished, it was kept as it was in the oven to cool down to room temperature.

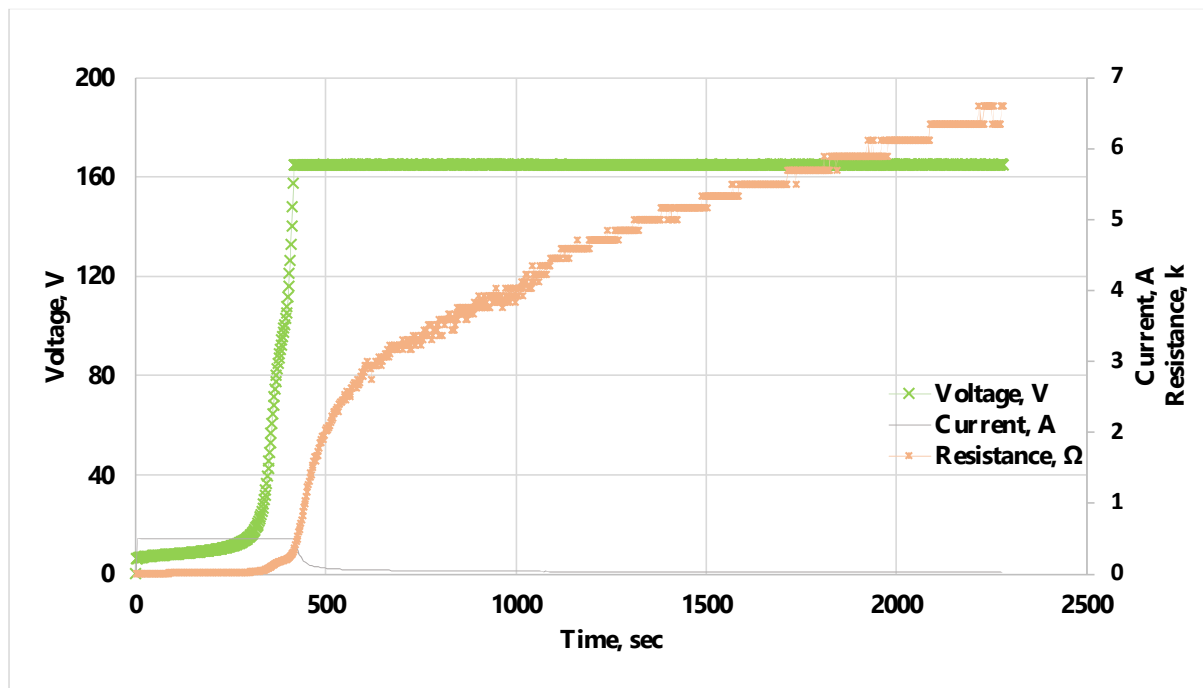


Fig. 4.5: The relationship of electrical current, resistance and voltage with time. The left axis shows the voltage values, whereas both the resistance and current were displayed by right axis. The green, orange and yellow lines represent the voltage, resistance and current respectively.

4.2.4 Capacitance Test

After the preparation of the sample, specimen for capacitance were cut by using a rotary cutter. Five specimens ($n=5$) had been cut from each sample, and each specimen was of same size ($8\text{ mm} \times 8\text{ mm}$). Every specimen was scratched by using sandpaper and then swiped by using acetone. Following that, the impedance and capacitance of the specimen had been measured by using an impedance tester (Hioki Impedance Analyzer IM3570 as shown in Fig. 4.7). The frequency at which the test was carried out was 2 kHz [116]. Despite measuring the capacitance and impedance over a broad frequency range, a constant frequency was selected so that the many samples could be readily compared under the same circumstances. Before measuring the capacitance of the specimen, adjustments were required. Adjustments were made by using a plastic insulator (same thickness as specimen) for open condition, and by touching the two electrodes for short condition. After the adjustments, the frequency was set at 2 kHz. Following that, the specimen was sandwiched between two round copper plates, and the two electrodes were put on both sides of the copper plates. Fig. 3.6 displays the test procedure's schematic diagram. The data of capacitance were collected to check the effect of CNF as a dielectric.

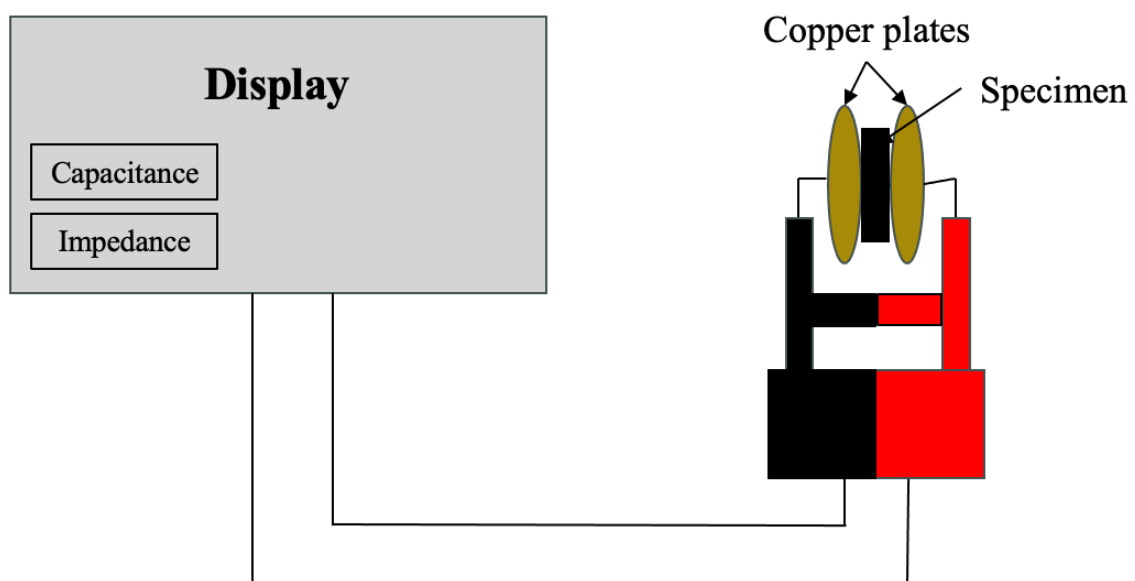


Fig. 4.6: The schematic diagram of the capacitance test that shows the specimen is held in between two copper plates, and the values are shown in a display connected by the electrodes.

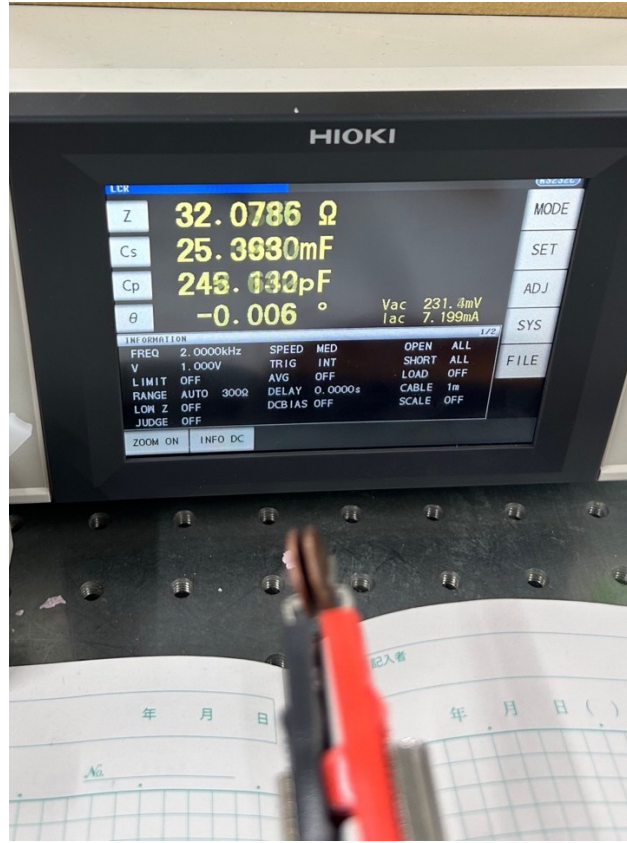


Fig. 4.7: The actual diagram of the capacitance test that shows the specimen is held in between two copper plates, and the values are shown in a display connected by the electrodes.

4.2.5 Four-point bending test

Four-point bending tests had been chosen for examining how the use of dielectrics affected CFRP's bending strength and modulus. The test procedure was followed the same way as described in article 2.2.6 Four-point bending test. From the same sample used for capacitance test, three specimens were cut out in order to verify the multifunctionality of the samples. The sample was used for both capacitance and bending test to investigate the effect of dielectric inclusion to CFRP. The head speed of 3.5 mm/min was determined. The lower indenters were spaced 50 mm apart and the upper indenters were spaced 10 mm apart. Eq. (1) was used for calculating the specimen's bending stress (σ_b),

$$\sigma_b(MPa) = \frac{6P_b(kN)L(mm)}{5b(mm)h^2(mm^2)} \quad \text{eq. (1)}$$

$$E_b(GPa) = \frac{59P_b(kN)L^3(mm^3)}{250\delta(mm)b(mm)h^3(mm^3)} \quad \text{eq. (2)}$$

where P_b is the test power (kN), L is the span length, δ is the deflection, h is the thickness, and b is the specimen's width. Loading span and support span had a length of 40 mm and 80 mm respectively maintain a 1:2 ratio as per the standard. When the maximum load was applied, the bending strength of the specimen was measured. The slope obtained from the elastic area of the load-deflection curve had been used to calculate the flexural modulus (E_b) using eq. (2) by following JIS K 7074 [141, 142]. The specimens were loaded under particular conditions by operating a PC software named Trapezium (Shimazu Corporation).

4.2.6 Tensile test

The Tensile strength and Young's modulus of the material were evaluated using a tensile test. Additionally, the correlation between stress and strain was evaluated, and the engineering relationship between these two were confirmed. The tensile test was followed the same procedure described in article 2.2.7 Tensile test. Tensile test was applied to Sample B, Sample C, Sample E and CFRP excluding any CNF or dielectrics for comparison. Samples A and D were not put through tensile testing since they showed no capacitance, while Sample F was omitted since it included plastic in addition to CNF. Sample B was chosen in order to compare drying time and film structure with Samples C and E, even though it lacked capacitance. The load cell was strain gauge type load cell which had a maximum capacity oof 10kN. A dynamic strain meter and a strain gauge from Kyowa Electrics Co. Limited. was used for measuring the vertical strain and lateral strain. The test speed was set to be 1 mm per minute and the grippers were spaced 75 mm apart as per JIS K7164 [142].

4.2.7 Scanning Electron Microscope (SEM)

A scanning electron microscope (SEM JSM- 6510LA) was employed to look into the cross section of the specimen that had been fabricated by EDRM. With a typical probe current and an accelerating voltage of 10 kV, the SEM was operated in High Vacuum, Secondary Electrons mode. The sample size of the specimen for SEM was cut into a rectangle of 8 mm × 8 mm. The side of the specimen for cross-section observation was sanded multiple times by using

sandpapers. In addition, in order to determine the fiber volume content (V_f) based on the 1,500-magnified image observation findings obtained by the SEM, the image processing program ImageJ (Ver. 1.54f) was used. The Gaussian Blur filter was used as a pre-processing step to minimize image noise, followed by binarization processing to extract the carbon fiber region and opening treatment as a post-processing step.

4.3 Result and Discussion

4.3.1 Capacitance test

Impedance and capacitance of the specimen with different dielectrics were obtained from the test. Fig. 4.8 and 4.9 shows the value of obtained capacitance data. Additionally, Table 4.2 summarizes the electrical performance of the structural capacitors of various dielectric material used. The left axis and blue bar show the value of impedance, whereas the right axis and orange bar portray the value of capacitance in the case of Fig. 4.8. Considering CNF, it can be noticeable from Fig. 4.8 that sample C showed the maximum capacitance of 813 nF/m^2 while sample E showed 469 nF/m^2 of capacitance. Sample A, B and D didn't show any capacitance characteristics. It is because the CNF displaced due to the pressure from metal plate and failed to separate the electrode. In the case of Fig. 4.9, the right axis and yellow bar portray the value of capacitance, whereas the left axis and green bar show the value of impedance. From this figure, it is visible that the specimens with dielectrics such as paper, tissue, jute fiber showed capacitance except plastic. The low thermal stability of plastic is the cause of its lack of capacitance. As a result, it failed to function as an insulator since it melted down when the sample was heated during the heat curing process under elevated temperature and pressure. The melt down of the plastic was later confirmed by observing the cross-sectional image which indicated the deformation of plastic that led to loss of dielectric function. Paper and tissue showed almost the same capacitance around 703 nF/m^2 and 672 nF/m^2 respectively. Paper has moderate capacitance and can produce stable, flat dielectric layers, but its weak fiber-matrix adhesion can make it fragile. Additionally, tissue paper, which can offer a thin and porous layer, is easily compressed, which could result in non-homogeneous dielectric layers. Jute fiber showed a very negligible amount of capacitance compared to other dielectrics because of its natural conductivity. Since both CNF and the electrodeposition solution (epoxy) used in this study were dielectrics, the overall impedance increased as resistivity increased. From Table 4.2,

it is noticeable that the tissue paper had the highest dielectric constant while the CNF film had the lowest. In spite of that, the compressibility and irregular thickness had reduced the dielectric effectiveness of tissue paper, whereas the ultra-thin layer of CNF film resulted in a high capacitance per dielectric thickness as same as bond paper (625pF/mm). Jute fiber showed a very poor capacitance performance because of its thick layer and low permittivity. However, compared to other dielectrics, the capacitance of employing CNF as a dielectric exhibited greater value. In addition to its capacitance and other dielectric characteristics, CNF is more environmentally friendly, more sustainable, and better biodegradable than other dielectrics.

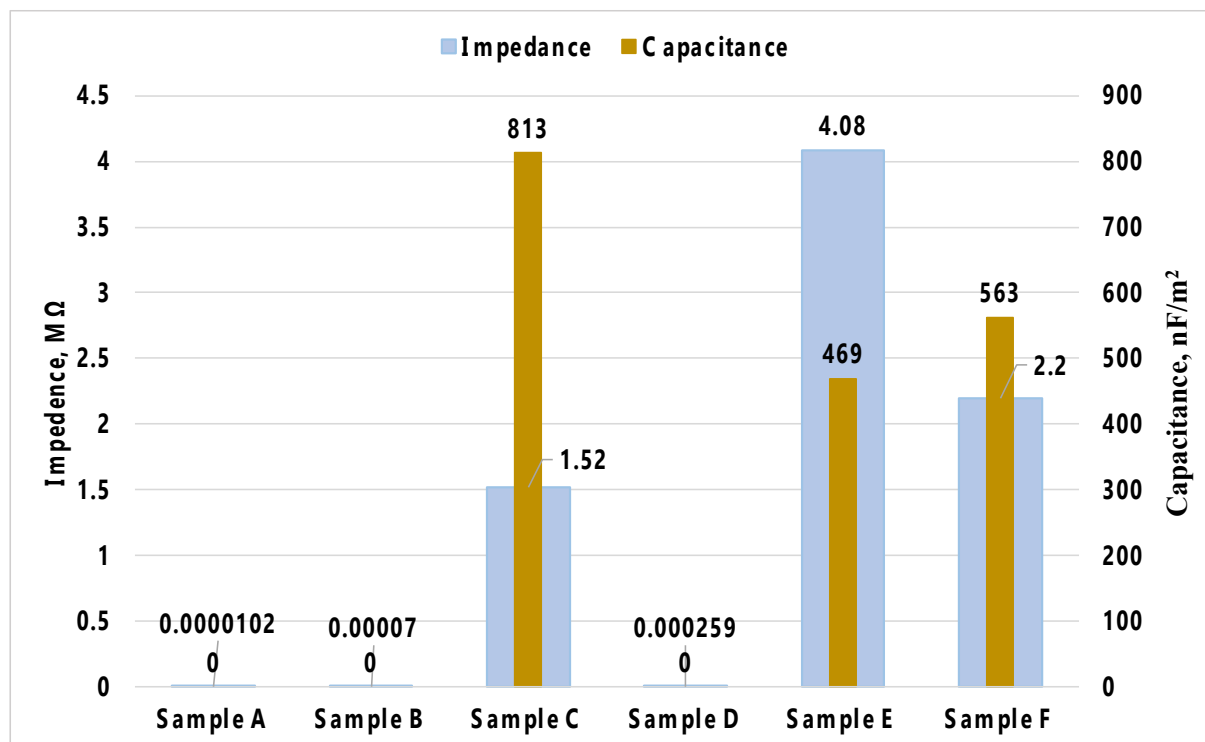


Fig. 4.8: Impedance and capacitance of CFRP including CNF. The data of Sample A to sample B are shown here. The blue bar shows the impedance values whereas the brown bar portrays capacitance value.

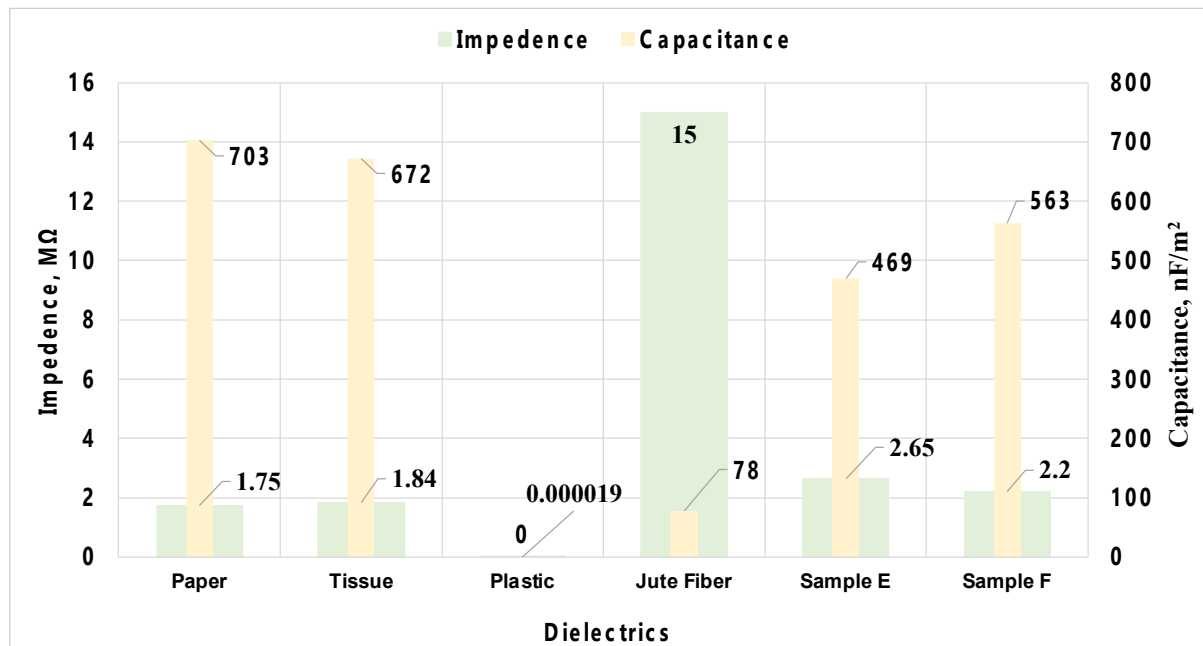


Fig. 4.9: Impedance and capacitance of CFRP with different dielectrics. The yellow bar shows the capacitance value whereas the green bar portrays impedance values.

Table 4.2. Dielectric properties of Different dielectrics

Dielectrics	Dielectric thickness, mm	Total thickness, mm	Electrode area, mm ²	Dielectric constant, ϵ	Capacitance/dielectric thickness, (pF/mm)
Bond-Paper	0.072	1.543	68	5.38	625
Tissue-Paper	0.117	1.590	70	8.11	367
Jute-Fiber	0.406	1.894	72	3.47	12
Plastic	----	1.451	66	----	----
CNF-Film	0.048	1.477	67	2.46	625

4.2.2 Four-point bending test

The results obtained from the bending test are shown in Fig. 4.10 and 4.11. Both bending strength and bending modulus are depicted in a single graph. The left axis shows the value of bending strength, while the right axis portrays the values of flexural modulus in both figures. On the one hand, Fig. 4.10 shows the bending properties of CFRP that had been incorporated with different approaches of CNF addition. As the Sample A, B and D did not show any capacitance, only the bending test results of Sample C, E and F are shown in the figure. From the figure, it can be seen that the bending strength of Sample E and Sample F is higher than Sample C. On the other hand, Fig. 4.11 depicts the bending properties of CFRP with different dielectrics. It is noticeable that the bending strength of CFRP with dielectrics such paper, tissue and jute fiber had low bending strength compared to those with CNF. CNF as dielectrics actually has a higher bending strength and the differences between them are very little. Though the film had maximum bending strength, nevertheless, CNF 1 day which has maximum capacitance, had a bending strength of 200 MPa which is almost same as normal CFRP. The strength got weaker because waiting longer after EDRM reduced the moisture resulting in a poor mixture of fiber and matrix. However, incorporating plastic with CNF film increased the bending strength by 38% because plastic acted as a matrix along with epoxy to increase the bending properties and protected the CNF film from damage due to pressure and high heat. Albeit, CNF has the potential to enhance the adhesion between the fiber and matrix which helps in increasing the load transfer efficiency and resisting the interlaminar shear during bending. As for why, adding CNF can delay the crack initiation that results in an increase of bending strength of the CFRP. Additionally, in case of flexural modulus, the variation among the samples is not significant, but the CFRP combining CNF film has the maximum bending modulus of 54 GPa. Following that, Sample F, which is CFRP that has both CNF film and plastics has the bending modulus of 52 GPa. Moreover, the bending modulus of CFRP with dielectrics such as paper, tissue, plastic and jute fiber showed the same scenario of bending strength. Inserting CNF film into the CFRP produced the maximum bending modulus of 54 GPa.

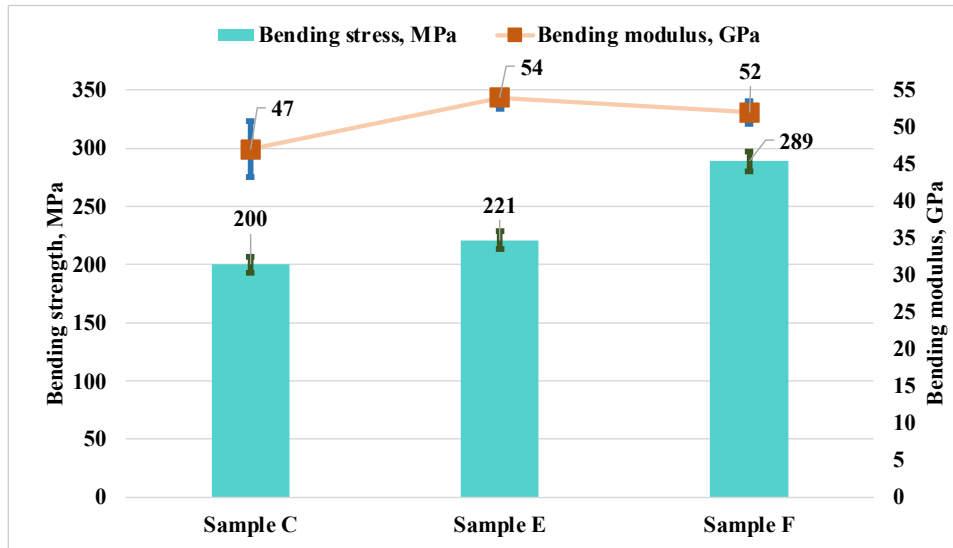


Fig. 4.10: Bending strength and Bending Modulus of CFRP including CNF. Samples C, E and F showed the capacitance; hence the bending data of these samples are presented.

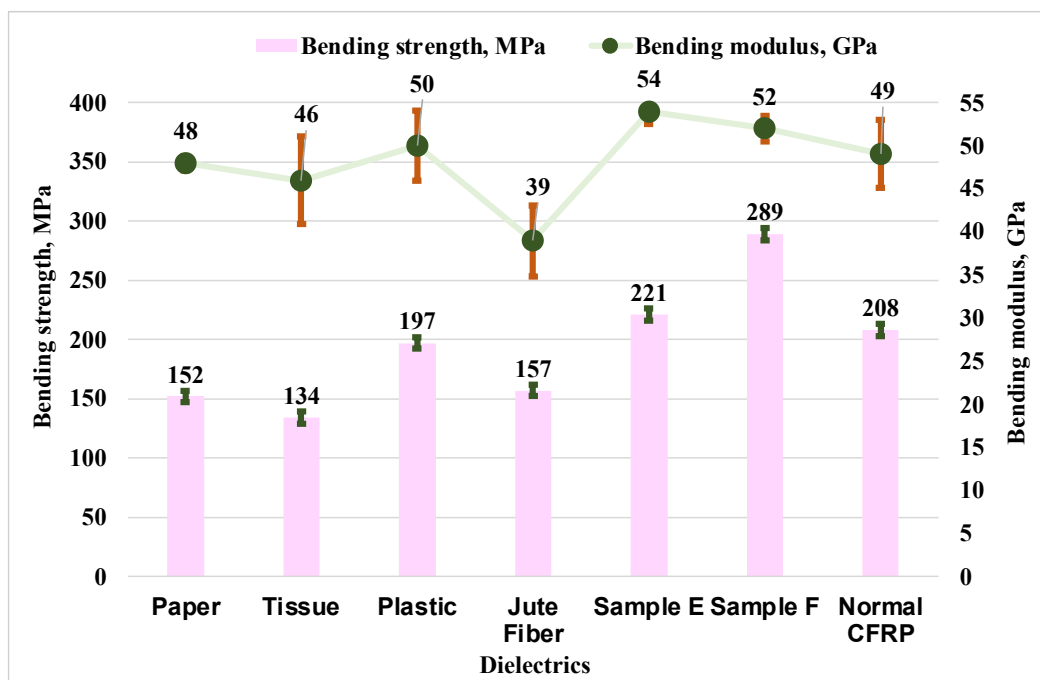


Fig. 4.11: Bending strength and Bending modulus of CFRP including different dielectrics. The bending data of CFRP without any dielectrics are also presented here for comparison with CFRP with dielectrics.

4.2.3 Tensile test

The value of tensile modulus and tensile strength obtained from the tensile test are displayed in Table 4.3. Tensile test was conducted on Sample B, C and E. These data were compared to the data of CFRP that has no CNF inclusion. The CFRP with the maximum tensile strength and tensile modulus was the one without CNF. The CFRP's tensile strength and modulus were both reduced by the addition of CNF. When the CNF layer is not dry, the Young's modulus drops by 20% and after 24 hours of drying, it decreases by 25%. When the CNF layer is still wet, its tensile strength decreases by 8.9%, and after a 24-hour drying time, it drops by 17%. The estimated value of the tensile modulus of CNF can be obtained by simply applying the rule of mixture [143] to Sample B. The Young's modulus of normal CFRP (E_{CFRP}) is 49 GPa, and the Young's modulus of CFRP with CNF inclusion (E) is 47 GPa, according to data given in Table 2. Sample B (t) had an average thickness of 1.12 mm, while normal CFRP (t_{CFRP}) had an average thickness of 1.02 mm. From the equation of rules of mixture, we get,

$$E = v_{CFRP}E_{CFRP} + v_{CNF}E_{CNF} \quad \text{eq. (3)}$$

$$v_{CFRP} = \frac{t_{CFRP}}{t} = 0.91 \quad \text{eq. (4)}$$

$$v_{CNF} = 1 - v_{CFRP} = 0.09 \quad \text{eq. (5)}$$

If we put the values of eq. (4) and eq. (5) in eq. (3), we get,

$$E_{CNF} = 26.8 \text{ GPa}$$

The Young's modulus of CNF was estimated to be 26.8 GPa based on the simple calculation above. Since CNF has a very low modulus, adding it to CFRP in the neutral axis may dilute the overall stiffness which resulted in a slight decrease in the elastic modulus of CFRP.

Table 4.3. Types of CNF application as dielectrics

Sample	Description	Young Modulus GPa	Tensile Strength MPa

1	CFRP	without	49.0	347
	CNF			
2	Sample B		47.0	351
3	Sample C		36.6	286
4	Sample E		28.8	224

4.2.5 SEM Morphology

The cross-sectional surface of Sample F that uses CNF film as dielectrics is depicted in Fig. 4.12. The SEM image shows the alignment of the fiber and matrix as well as a continuous dielectric layer throughout the specimen separating two preforms that are actually working as electrodes. The uniform layer of the dielectric ensures the separation of electrodes and acts as a capacitor. There are no visible voids at the CNF and CFRP interface, indicating good adhesion. Furthermore, even with CNF insertion, the alignment of the carbon fiber appears to remain unaltered and well-aligned. Though the CNF film was able to separate the electrodes, its thickness varied throughout the specimen. On the contrary, a cross-sectional surface of a specimen which uses plastic film is displayed in Fig. 4.13. It is evident that the plastic does not function as a separator of the electrodes as it melts, and as a result, it does not exhibit capacitance properties. Nonetheless, the deformation of plastic did not interrupt the fiber architecture or fiber-matrix bond indicating a preserved mechanical strength. Moreover, Fig. 3.14(a) shows a 1500x magnified image obtained from SEM and Fig. 4.14(b) portrays a binarized version of Fig. 15(a) to obtain the fiber volume fraction data. It was verified that a dark gray resin was evenly precipitated around the light gray carbon fiber in the 1,500x observation result from Fig. 3.14(a). The fiber volume content (V_f) was calculated around 70% from the processed image from Fig. 3.14(b).

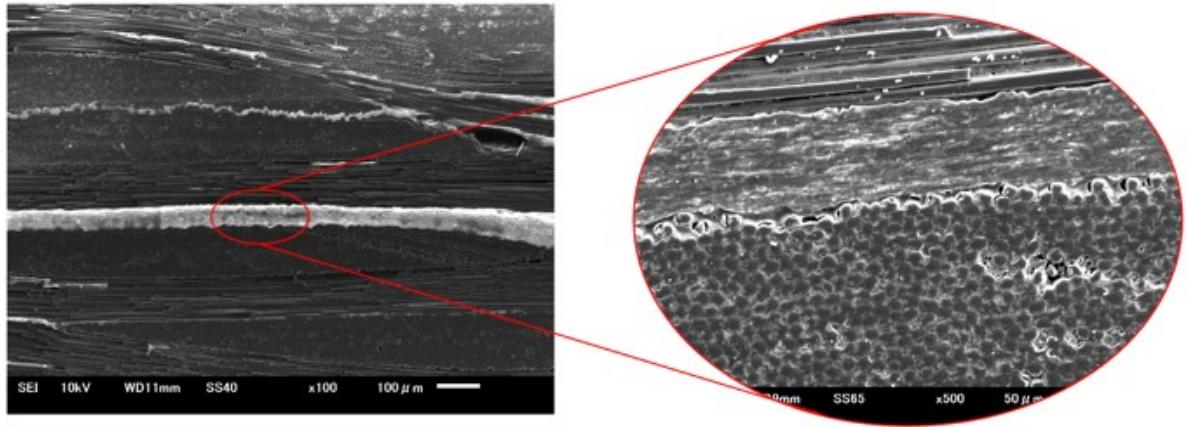


Fig. 4.12: Image of CFRP cross-section including CNF film as a dielectric. The right figure the magnified version of the left figure that used x500 magnification.

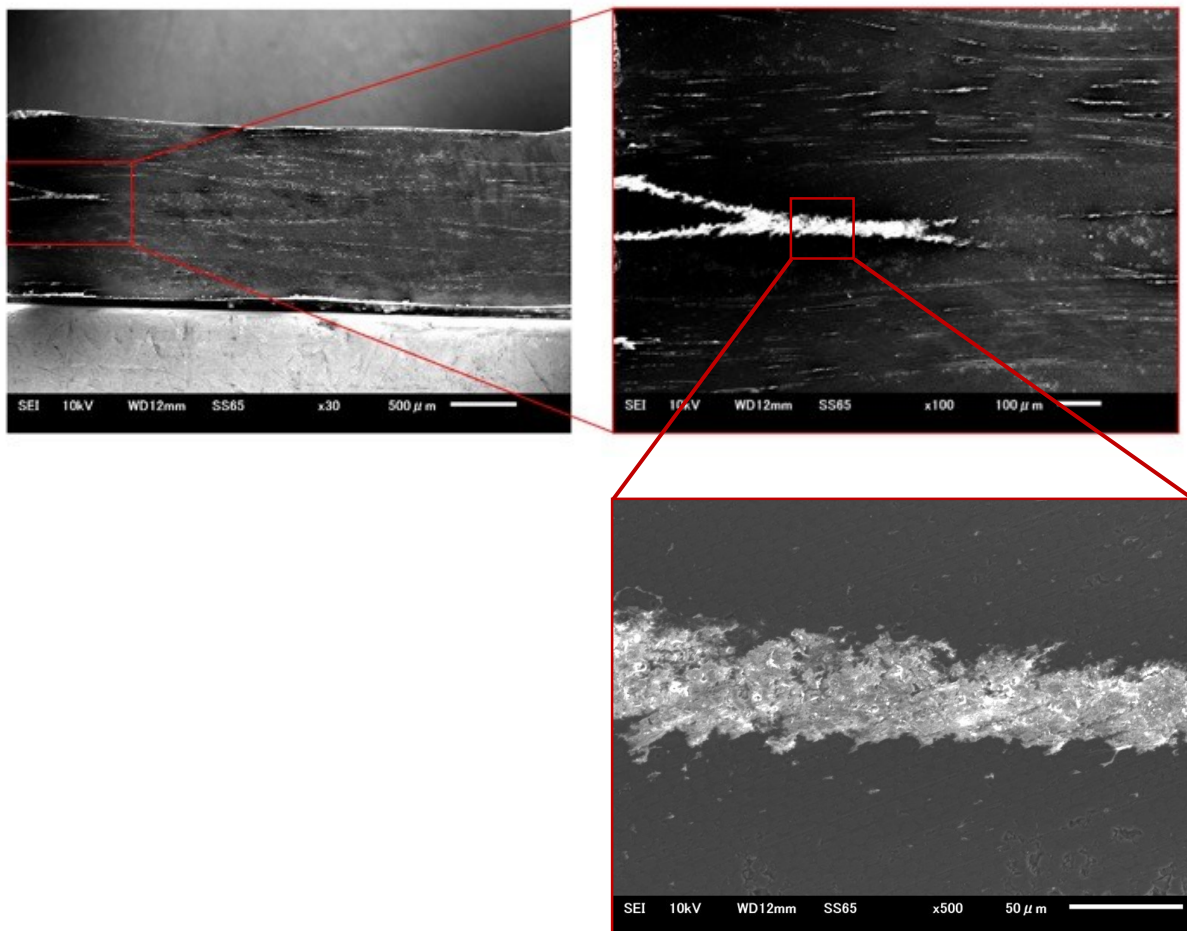


Fig. 4.13: Image of CFRP cross-section CFRP including plastic as a dielectric. The magnified version of the left figure is given in the right figure that uses x100 magnification and x500 magnification.

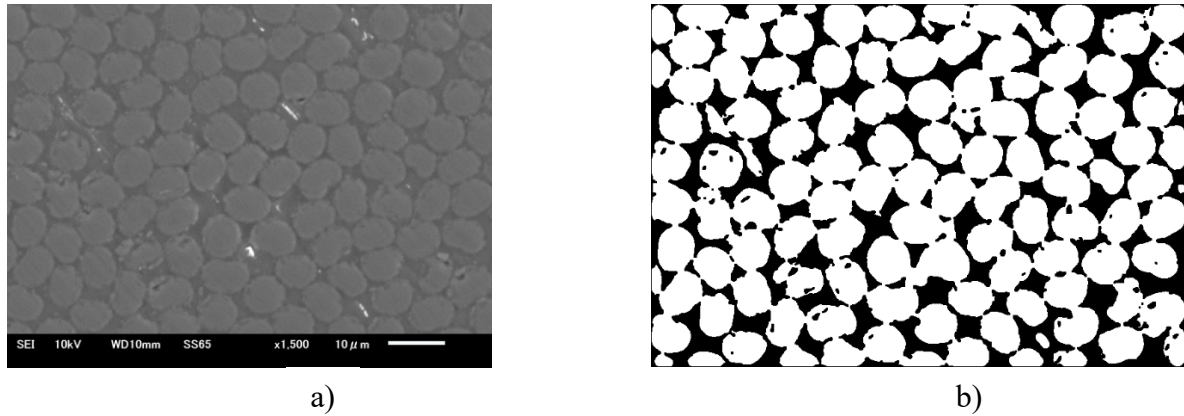


Fig. 4.14: Image of CFRP cross-section CFRP taken by SEM; a) SEM cross-sectional image (x1500), b) SEM image after image processing with Image J

4.4 Conclusion

This work studied the mechanical and electrical features of CFRP made by using EDM. CNFs were used for enhancing both capacitance and bending properties of CFRP. For a comparative analysis, various dielectric types were also employed into CFRP in addition to CNF. Moreover, EDM is a new composite manufacturing method that has been applied to check the possibility of fabricating structural capacitors. By employing CNF, CFRP can be acted as a capacitor with having maximum of 813 nF/m² which is even higher than other dielectrics such as paper, tissues and jute fiber. In addition, adding CNF into CFRP can improve the bending characteristics. Bending strength of CFRP was increased by around 6 percent. Therefore, by serving as a reinforcing dielectric, CNF exhibits a synergistic effect that enhances both electrical insulation and load bearing capability. The addition of CNF, however, decreased the elastic modulus of the CFRP. Consequently, it can be said from the above discussion that CNF can be inserted into CFRP for preparing capacitors without compromising the mechanical properties significantly. A major issue in structural energy storage systems is striking a compromise between mechanical strength (6% increase in bending strength) and electrical performance (813 nF/m² capacitance), which is accomplished in this study. In addition to

improving the structural capacitor's sustainability, these energy-storing devices have a wide range of other uses. It is possible to use this structural capacitor within electrical vehicles where a small amount of power is needed. Additionally, they can also be used in place of the lightweight structural component of unmanned aerial vehicles (UAVs) or aerospace the lightweight structural part of aerospace or unmanned automatic vehicles (UAV) to store energy along with providing a stiff framework, consequently reducing the space and weight. Even though the cost of materials may be higher during fabrication than for existing capacitors, their long-term sustainability and mechanical robustness end up reducing the lifetime cost. To conclude, it can be mentioned that multifunctional CFRP including strength and capacitance can be fabricated by using EDRM with minimal impact to the tensile properties.

4.5 Future Scope

Although this study investigates the possibility of preparing structural capacitors by EDRM, the capacitance found in this research is quite pleasant. As a result, further research is required to implement this to make capacitors with enhanced electrical and mechanical properties to meet the increased demand of lightweight structural capacitors. If the capacitance can be enhanced more, this will contribute greatly to the field of lightweight energy storage applications.

Chapter Five

Preparation of Nylon 6 based Carbon fiber/epoxy
composite and evaluate their compatibility

Abstract (Chapter five)

Carbon fiber reinforced polymer (CFRP) has been widely researched for their extraordinary properties and huge applications in aircraft and automobile sectors. This study explores incorporating additional nylon 6 thermoplastics into carbon fiber reinforced epoxy thermoset composites. The objective of this work is to check the compatibility of thermoplastic nylon 6 and thermoset epoxy by evaluating the performance of resultant composite material. Charpy impact test and four-point bending test had been carried out to characterize mechanical properties of Carbon fiber/nylon 6/epoxy composite. A roughness test was performed to check the roughness of CFRP with and without nylon 6 inclusion. In order to ensure the presence of nylon 6 on the surface of the composites, Raman spectroscopy was conducted. All the results from the experimental testing showed that the carbon fiber/nylon 6/epoxy composite outperformed carbon fiber/epoxy composites in all mechanical tests conducted. The bending strength of CFRP increased almost double by adding nylon 6. Additionally, the CFRP, including nylon 6 consumed 38% higher impact energy than normal CFRP.

5. Preparation of Nylon 6 based Carbon fiber/epoxy composite and evaluate their compatibility

5.1 Introduction

Carbon fiber-reinforced polymers (CFRPs) composites are widely recognized as advanced materials due to their excellent combination of properties such as low density, high specific strength, stiffness, lightweight nature, and resistance to chemical and environmental degradation [144-146]. These composites are formed by combining two or more distinct materials typically fibers and a polymer matrix and can be further modified by the inclusion of fillers. This multi-phase structure enables CFRPs to achieve properties significantly different from those of their individual constituents, such as improved mechanical strength, thermal and electrical conductivity, electromagnetic shielding, optical transparency, and more [147-152]. These attributes make CFRPs highly desirable across industries such as automotive, marine, construction, and sports equipment [153-156].

The polymer matrix plays a crucial role in the overall performance of CFRPs. It not only holds the carbon fibers together but also ensures effective stress transfer between them, contributing to structural integrity [157]. Additionally, the use of polymers allows for the creation of lightweight yet durable materials, which directly enhances fuel efficiency and reduces emissions in transportation sectors, aligning with sustainability goals [158]. Based on the nature of the polymer matrix, CFRPs are broadly categorized into thermosetting and thermoplastic composites. Thermosetting matrices, such as epoxies, have been widely researched and industrially adopted due to their excellent thermal stability and dimensional precision. However, a major drawback of thermoset-based composites is their non-recyclability, which poses significant environmental and cost-related concerns [159-162]. In contrast, thermoplastic-based CFRPs have emerged as a promising alternative due to their toughness, recyclability, damage tolerance, and fast processing capabilities. Their potential for in-situ consolidation, indefinite shelf life, and ease of welding and repair make them ideal for future aerospace applications [163-168]. While thermosets offer superior structural accuracy and thermal resistance, thermoplastics are favored for their impact resistance, toughness, and environmental advantages [169-172].

Despite their individual benefits, studies comparing the two have highlighted nuanced differences. For example, Yan et al. [173] found that carbon fiber/epoxy (CF/EP) laminates showed more uniform fiber distribution, while carbon fiber/polyamide 6 (CF/PA6) composites achieved higher interfacial shear strength due to better fiber impregnation. Similarly, Liu et al. [174] observed that delamination in thermosetting composites is more concentrated, whereas thermoplastic composites exhibit more uniform delamination across the thickness, which can be advantageous in terms of damage tolerance. Furthermore, Meshram et al. [175] reported that incorporating nylon into epoxy systems improved tensile strength and thrust force, while Islam et al. [176] demonstrated that adding nylon mesh enhanced mechanical properties but decreased Young's modulus and electrical conductivity, indicating improved insulation. In another approach, Malek et al. [177] utilized nylon powder to reduce voids in epoxy composites by promoting better fusion.

The inclusion of nylon 6 (PA6) in epoxy matrices is primarily aimed at toughening the brittle epoxy resin and enhancing the overall mechanical performance. Although PA6 has favorable characteristics for epoxy toughening, it has not been extensively studied in CFRP systems. Traditionally, materials such as rubber particles [178-181] and high-performance thermoplastics like polysulfone, polyetherimide, polyphthalazone ether ketone (PPEK), polyphenylene oxide (PPO), polyimide (PI), and polyether ether ketone (PEEK) have been used for this purpose [182-188]. These materials act as ductile inclusions, absorbing impact energy and hindering crack propagation. An additional advantage of using PA6 is its ability to form strong adhesive bonds with epoxy when processed at temperatures above 200°C. Various adhesion theories have been proposed to explain this behavior, including mechanical interlocking, chemical bonding, diffusion, physical adsorption, electrostatic interaction, and weak boundary layer theory [189-198]. According to Gul et al. [199], nylon 6 forms a hydrogen bond with epoxy to create an entangled structure that toughens the epoxy, and the chemical reaction is shown in Fig. 5.1.

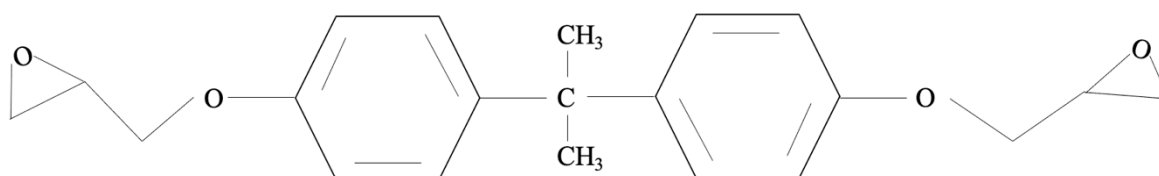
As previously noted, numerous efforts have been made to enhance the toughness of epoxy resins using various thermoplastic materials. Among these, nylon 6 presents promising potential due to its unique mechanical and thermal characteristics. Reinforcing epoxy which is one of the most widely used polymer matrices in composite systems with nylon 6 sheets may lead to the development of novel materials with significantly improved performance. This study specifically explores how the incorporation of nylon 6 layers into epoxy-based CFRP composites influences their mechanical and thermal behavior. The experimental framework

was carefully designed to assess the impact of nylon 6 integration on key properties such as flexural strength, tensile strength, and impact resistance. In addition, a novel fabrication technique was adopted for producing these composites. In this method, epoxy resin was first electrochemically impregnated into carbon fiber layers, followed by the strategic placement of nylon 6 sheets within the laminate structure. The composite was then heat cured at approximately 230°C to promote strong interfacial bonding between the epoxy matrix and the nylon 6 layers. This processing approach, known as Electrodeposition Resin Molding (EDRM), was originally introduced by the authors in prior work [200-204]. The current study aims to investigate the effect of incorporating nylon 6 into carbon fiber/epoxy composites fabricated via this novel EDRM method. The research focuses on evaluating the mechanical properties such as bending strength, impact resistance, and surface morphology of the modified composites compared to conventional CF/epoxy systems. The main goal is to assess how nylon 6 influences the structural performance of CFRPs and whether it contributes to improved toughness and durability, potentially expanding their use in high-performance and damage-tolerant applications.



Aromatic Polyamide

+



Epoxy Resin

Ring opening reaction



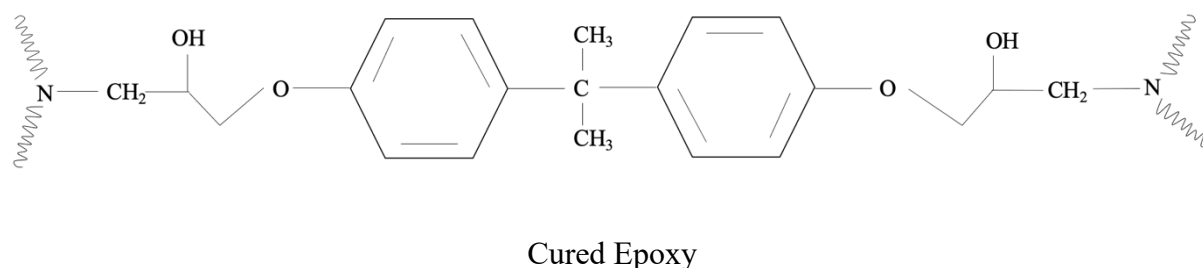


Fig. 5.1: Interaction of epoxy resin with polyamide [199]

5.2 Methodology

5.2.1 Materials and method

In this study, plain weave material was adopted in consideration of the retention of a stable shape. Unidirectional carbon fiber plain weave (PW) TR3110MS (Mitsubishi Chemical Corporation) was used the reinforcement. Thermoset epoxy resin from Elecoat AR (Shimizu Co., Ltd.) was chosen as polymer material in this work. Additionally, nylon 6 sheet (Kokugo Co., Ltd.) having thickness of 0.3 mm was chosen as a thermoplastic polymer to check the compatibility of nylon 6 and epoxy during the composite preparation. The fabrication method was followed by the steps mentioned in the article [1.2.3.1 Electrodeposition resin molding method](#).

5.2.2 Sample Preparation

Initially, the electrodeposition solution was injected into a beaker in which a stainless-steel mesh is placed along the inside wall of the beaker. The preform was prepared as shown in Fig. 5.1 (a) by stacking single sheet of carbon fiber plain weave with a width of 65 mm $\times$ a length of 200 mm and fixing the edges with duct tape to prevent the weaving from collapsing. In addition, the sizing material has not been removed. The upper part of the preform was fixed with a metal clip, immersed in the electrodeposition solution, and then held with a stand, and the power supply PWH401H (Kikusui Electronics Industry Co., Ltd.), stainless steel mesh, and eyeball clip were connected using an alligator clip. When the power supply was started, energizing based on the energization conditions set by the sequence creation and control

software SD027-PWR-01 (Kikusui Denshi Kogyo Co., Ltd.) started. The initial current was set to 0.5 A and the voltage limit was set to 165 V. The switching from current control to voltage control, resin precipitation has progressed sufficiently by decreasing the current, and energization was terminated. The electrical resistance value at the end of energization was calculated, and this was used as an index of the electrodeposition progression. This action was repeated four times for four preforms. Then nylon 6 was inserted into the preform in four different ways. 1 layer of nylon 6 was sandwiched in between the preforms in a way that both sides of the nylon 6 layer had two layers of carbon fiber layers as shown in Fig. 5.2 (c) (Sample - 1). Two layers of Nylon 6 layers was inserted in a way that the middle layer of the sample has two carbon fiber layers, and top and bottom also have carbon fiber layers as shown in Fig. 5.2 (d) (Sample - 2). Again, three layers of nylon 6 layers were put in the preform alternatively with carbon fiber layers as shown in Fig. 5.2 (e) (Sample - 3) and all three layers are in the middle sandwiched with carbon fiber layers as shown in Fig. 5.2 (f) (Sample - 4). In this study, we investigated the effect of adding additional polymer into the CFRP composites and their layouts on the mechanical properties of CFRP. After inserting the nylon 6 layers, the preform was sandwiched with a paper towel and a metal rod was used for pressing out excess moisture. A tabletop test press SA-302 (manufactured by Tester Sangyo Co., Ltd.) was used to apply a pressure of 50 MPa and glue the preforms together. The preform was then sandwiched between PTFE film and metal plate and pressurized using a vise. The pressurized preform was placed in a forced circulation type incubator VTRF-150 (manufactured by Isuzu Corporation) and heat-cured at 230°C for 6 hours. After the heat curing was completed, the sample was returned to room temperature (25°C) by natural cooling before being removed. CFRP was cut using a composite material cutting machine AC-200CF (Maruto Manufacturing Co., Ltd.) to prepare a specimen for various mechanical tests. When cutting, except for the end, only the central part of the plate that was sufficiently impregnated with resin was used. The average thickness of the specimen without any nylon 6 layer was about 1 mm. Additionally, to facilitate the melting of nylon 6, the layer of nylon 6 was hot-pressed before inserting into the epoxy impregnated carbon fiber layers by using a hot press machine (Fig. 5.3). The nylon 6 layers after the hot-pressed is shown in Fig. 5.4.

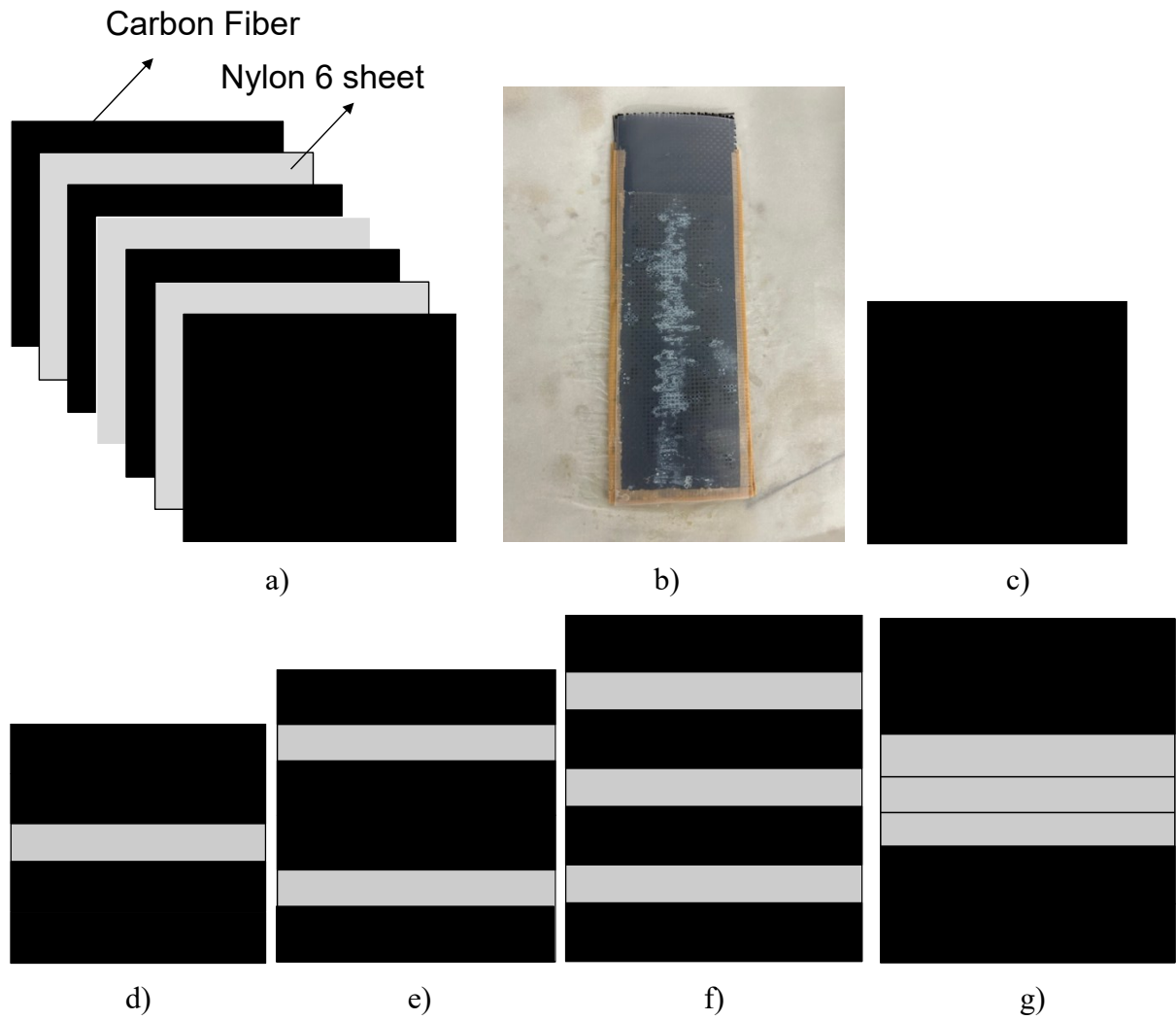


Fig. 5.2: Orientation of the preforms

Table 5.1. Experimental condition

Sample	Stacking sequence
1	Four Carbon fiber layers [PW0°] ₄
2	Four Carbon fiber layers and one nylon 6 layer [PW0°/PW0°/ Nylon6 /PW0°/PW0°]
3	Four Carbon fiber layers and two nylon 6 layers [PW0°/ Nylon6 /PW0°/PW0°/ Nylon6 /PW0°]
4	Four Carbon fiber layers and three nylon 6 layers [PW0°/ Nylon6 /PW0°/ Nylon6 /PW0°/ Nylon6 /PW0°]
5	Four Carbon fiber layers and three nylon 6 layers



Fig. 5.3: Hot-press machine

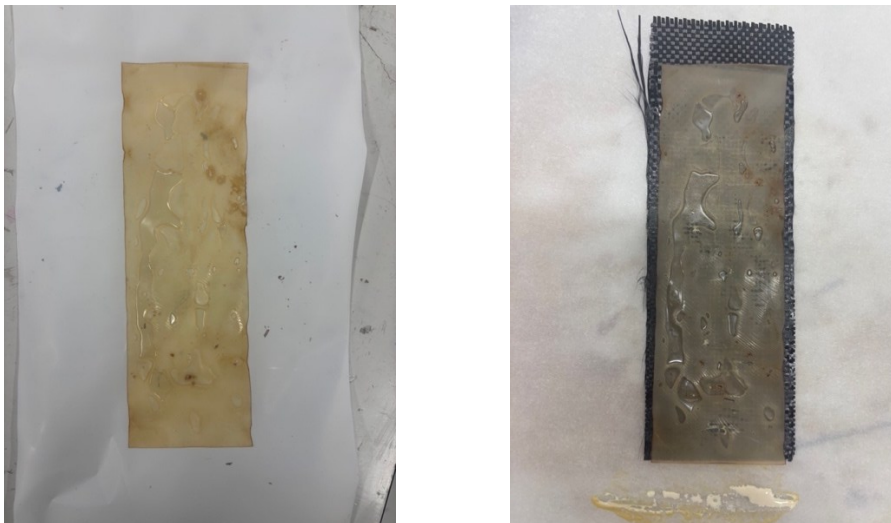


Fig. 5.4: Hot-pressed nylon 6

5.2.3 Vibration Test

As soon as the samples were ready, a vibration test was carried out. The specimens had been divided into 48 nodes. The test was carried out by following the same procedure mentioned in

5.2.4 Charpy impact Test

In order to assess the impact characteristics of CFRP with and without nylon six layers, the instrumented Charpy impact test was first conducted in accordance with ISO 179-1 [205]. Test specimens of three types ($n = 3$) were made. The specimen's dimensions were trimmed to 16 mm by 90 mm. According to ASTM guidelines (ASTM D6110), these measurements have been established [206]. A pendulum impact testing machine shown in Fig. 5.5 (Model no. 258, Yasuda Seiki Seisakusho, Ltd., Tokyo, Japan) was used to conduct the instrumented Charpy impact test. A single swing of the pendulum with the impact line halfway between the supports in a flatwise orientation broke the specimens, which were fastened to the machine anvils at a span length of 22.95 cm. The pendulum's striking nose was marked by its 150° angle. The specimens were struck by the striking hammer with a kinetic energy of 2 J and a speed of 3.8 m/s. By using the equation below, the impact energy was calculated.

$$E = WR[(\cos \beta - \cos \alpha) - (\cos \alpha' - \cos \alpha)(\frac{\alpha + \beta}{\alpha + \alpha'})]$$

In the above equation, WR is the moment around the hammer's rotation axis (N.m), α is lifting angle of hammer, α' is the swing angle when the hammer is swung from the lifting angle and β is the hammer swing angle after the specimen breaks.

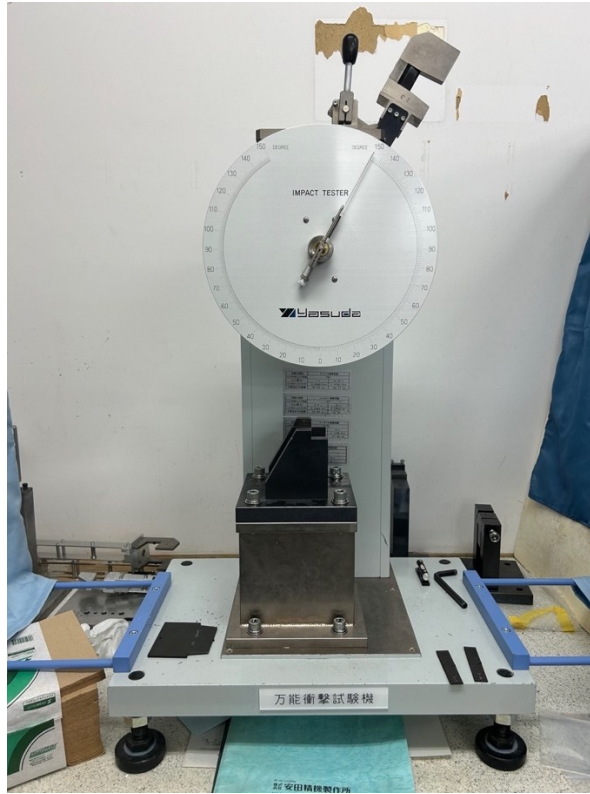


Fig. 5.5: Charpy Impact test machine

5.2.5 Four-point bending test

Following the impact test, a four-point bending test was used to confirm the CFRP with nylon 6 layer's static properties in accordance with ASTM D6272 [207]. Using a rotary cutter, the CFRP specimen, both with and without nylon 6, was carefully sliced into rectangles that were 70 mm long and 15 mm wide. The carbon fibers were aligned with the specimen's longitudinal direction. The same procedure described in article [2.2.6 Four-point bending test](#) was followed to carry out this test. For each condition, as depicted in Table 5.1, six specimens were prepared and tested ($n = 6$). A load was applied at the mid-span position of a four-point bending test using a universal testing machine (Ez Graph, Shimadzu Co., Ltd., Tokyo, Japan). A head speed of 3.5 mm/min was used. The distance between the upper and bottom indenters was 10 mm and 50 mm, respectively. The stiffness (P/δ) was computed by measuring the applied load (P) and deflection (δ) at the mid-span point. Furthermore, the maximum load was used to determine the bending strength, and the stress-strain relationship curve was used for determining the bending modulus.

5.2.6 Roughness test

In order to check the surface roughness of the specimen with and without the nylon 6 layers, roughness test was carried out by using a roughness tester machine from Mitsuyoto Corporation (model no. SV-648 shown in Fig. 5.6), and a 3D laser microscope from Keyence (model no. VK-X200 shown in Fig. 5.7 and 5.8). Same specimen size (40 mm x 40 mm) was prepared for both of the tests following JIS B 0601:2001. The measuring force at average position of the stylus was 0.75mN and the measuring force change rate is 0 N/m. In order to evaluate the roughness of the surface, the roughness parameter Ra (average amplitude) and Rz (maximum amplitude) was measured from the roughness curve obtained. The laser microscope measured the value at a spot diameter of 1 mm, while the roughness tester measured the values for three different distances of the specimen- 8mm, 2.5 mm and 0.8 mm.

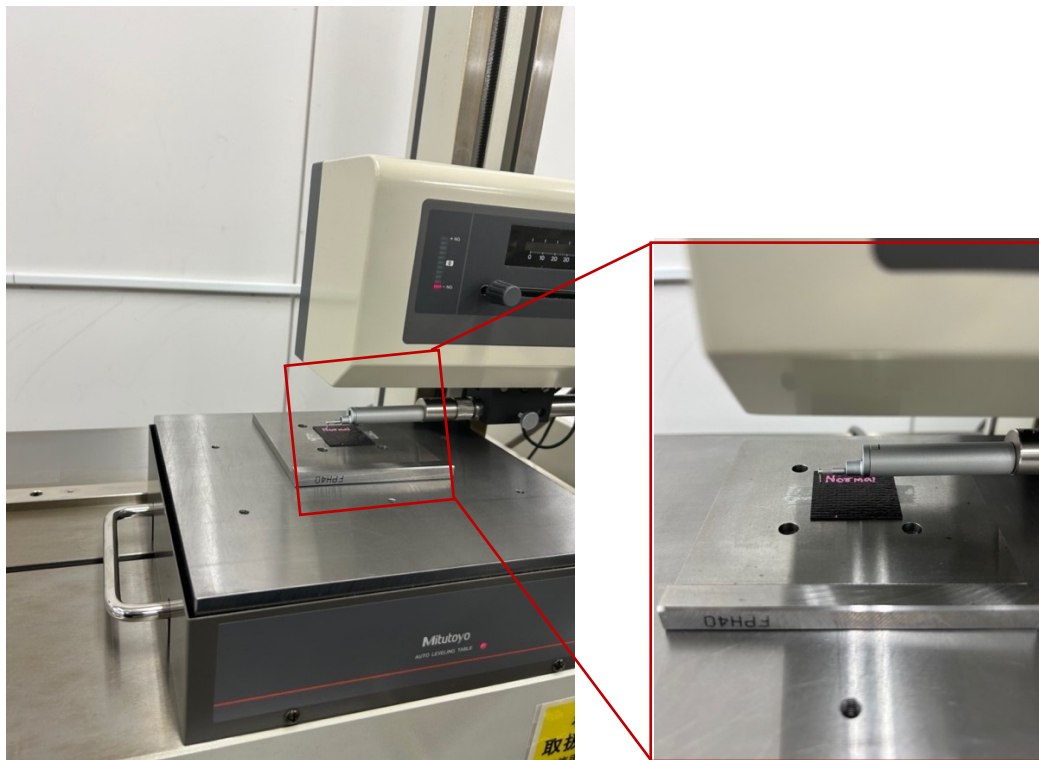


Fig. 5.6: Roughness testing machine



Fig. 5.7: Roughness testing machine

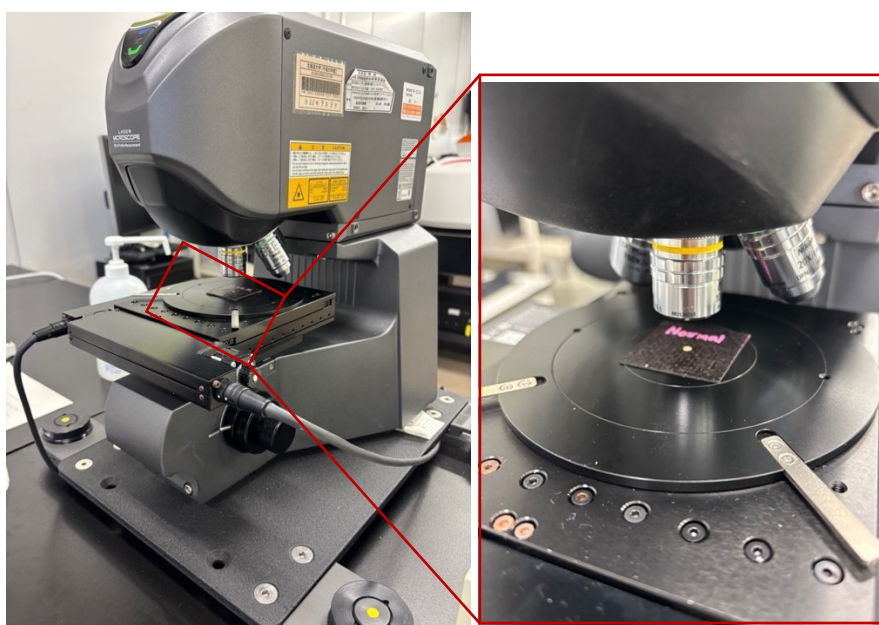


Fig. 5.8: Laser isotope machine

5.2.7 Raman Spectroscopy

As Raman spectrometer system a Renishaw Raman microscope (shown in Fig. 5.9) equipped with an Olympus BX41 confocal microscope was used, the system being operated by the

WiRE3.2 software. All the Raman spectra of this polymer in the spectral range from 100 to 3500 cm^{-1} and with spectral resolution (FWHM) of 4.0 cm^{-1} were obtained using a laser excitation wavelength of 785 nm. The laser power on the sample was 2.0 mW; the 50x objective (NA = 0.25) used yields a laser spot diameter of 1 mm, resulting in an irradiance of 1.1 $\text{mW } \mu\text{m}^{-2}$. All spectra were collected with fifteen accumulations with an exposure time of 10.0 s for each accumulation; afterwards we applied an automatic baseline correction to all spectra.

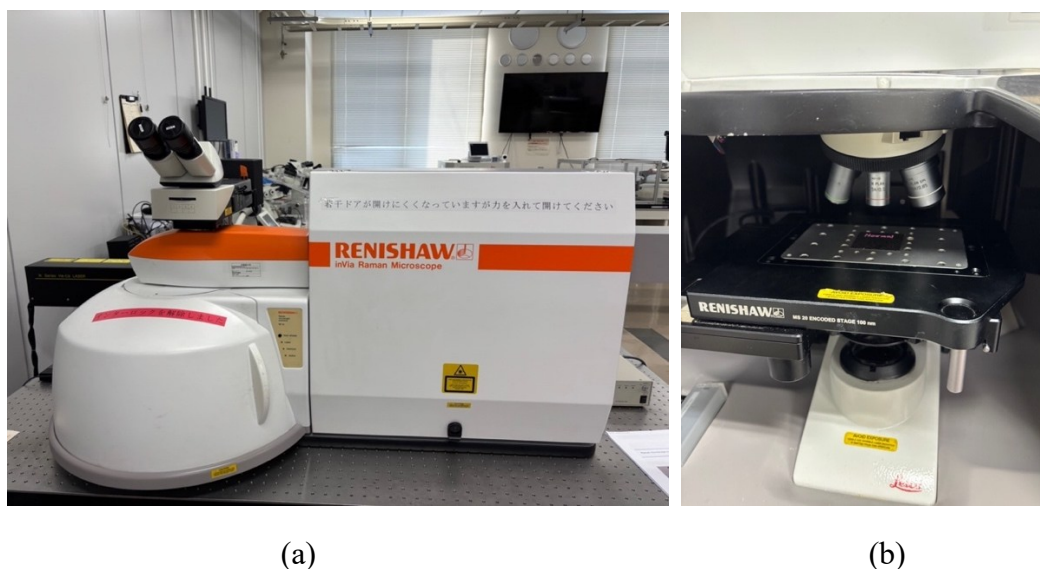


Fig. 5.9: (a) Raman Spectroscopy machine; (b) specimen under lens in Raman Spectroscopy machine

5.2.8 SEM

The specimen's cross section and the impact of nylon 6 were examined using a scanning electron microscope (SEM JSM-6510LA). The SEM was run in High Vacuum, Secondary Electrons mode with a standard probe current and an accelerating voltage of 10 kV. The SEM specimen's sample size was trimmed into an 8 mm by 8 mm rectangle. The specimen's side for cross-sectional inspection was repeatedly scraped using multiple sandpapers of 800 mm, 1500 mm, 2000 mm and 4000 mm sequentially. Following the impact and bending tests, the specimen which had one layer of Nylon 6 was examined. A specimen with a layer of preheated nylon 6 and one without the preheated nylon 6 but normal nylon 6 were both observed.

5.3 Result and Discussion

5.3.1 Vibration Test

Results were obtained from the test and are shown in Fig. 5.10 and 5.11. Figure 5.10 shows the modal visualization of sample A and B respectively. The mode shape's contour diagram uses color to depict the absolute amount of out-of-plane displacement, with the displacement increasing towards red. Nevertheless, Figure 5.11 (a) and (b) show the data of natural frequency and modal damping respectively. It can be seen from the figures that natural frequency of Sample A is higher than Sample B in case of mode 1, 3 and 5, on the contrary, it is lower than Sample B in case of mode 2, 4 and 6. Furthermore, when modal damping is considered, it showed the opposite trends. In other words, where natural frequencies were lower, damping was higher and where natural frequencies were higher, damping was lower. As mode 1 indicates the shear modulus of the materials, which further indicates that the shear modulus of Sample B is higher than the Sample A.

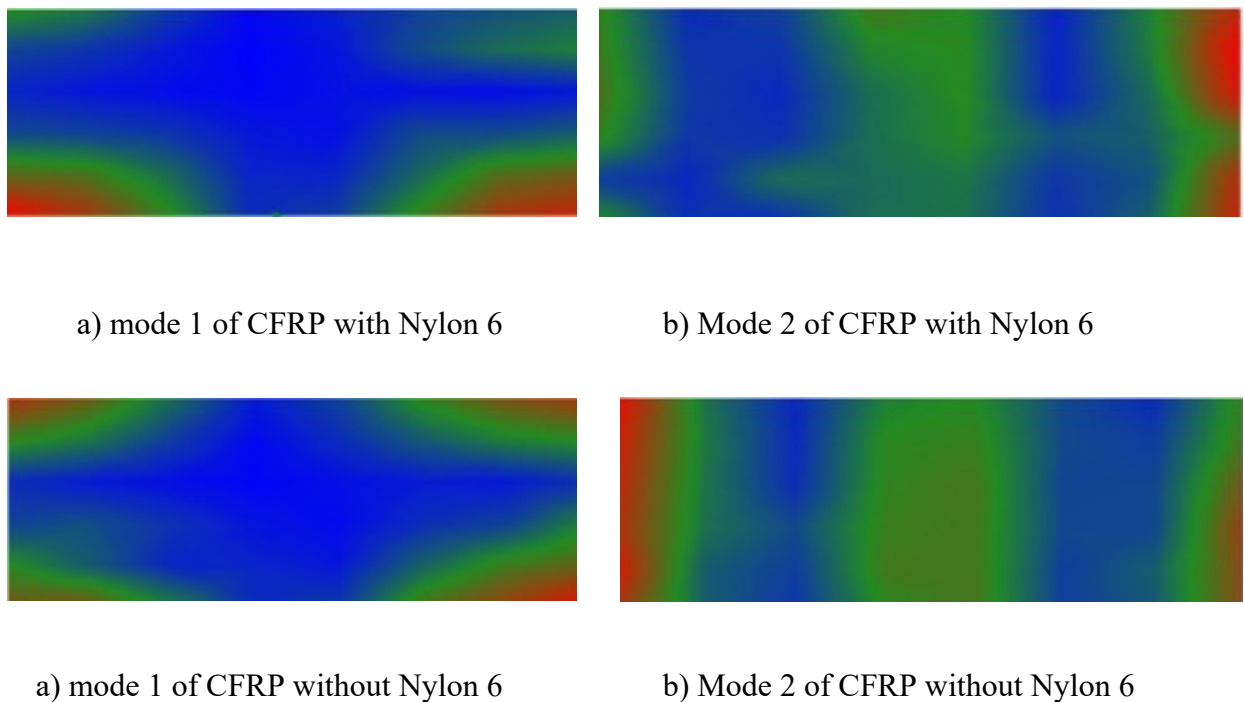


Fig. 5.10: Modal Visualization of CFRP

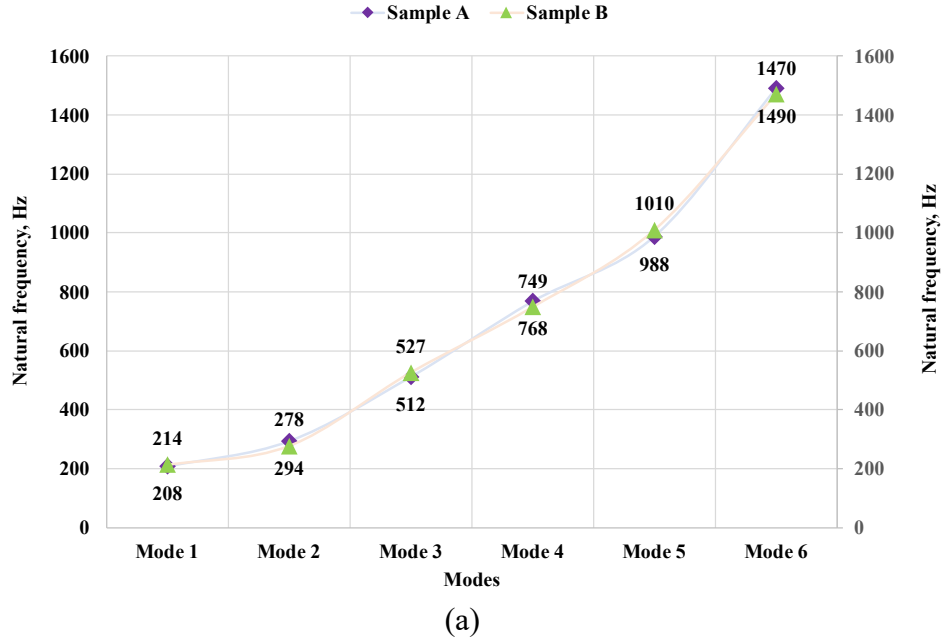
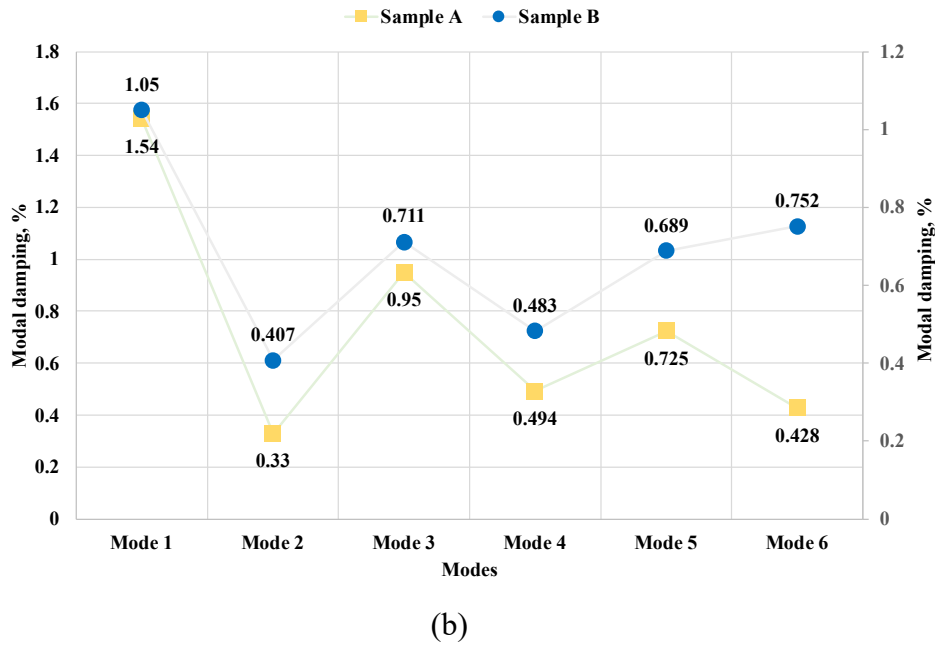


Fig. 5.11: The vibration data of composites obtained from vibration test. (a) natural frequency; (b) modal damping

5.3.2 Charpy Impact test

The charpy impact test was conducted to evaluate the impact energy required to break the specimen. The impact energy that required to break the specimen 1 to 5 are depicted in Fig. 5.12. The left scale shows the impact energy of the different specimens, and the impact energy

per average thickness of each specimen is also provided on top of each bar. From the figure, it is evident that adding nylon 6 into carbon fiber/epoxy increases the impact of energy. The higher the layer of nylon 6, the impact energy is also higher. As observed, Sample 5, which is the CFRP with three nylon 6 layers in the middle, had the highest impact energy value at 993 mJ, while Sample 1, which has no nylon 6 layer in the CFRP had the lowest at 561 mJ. With comparable impact energy levels of 806 mJ and 831 mJ, respectively, Samples 3 and 4 demonstrated a comparatively greater capability for energy absorption than Samples 1 and 2. An intermediate value of 598 MJ was shown in Sample 2 that has one layer of nylon 6 into CFRP. The standard deviation among repeated tests is reflected in the error bars, which are shown by green lines and show the variability within the data for each sample. Adding the nylon 6 layers alternatively with carbon fiber layers do not have significant difference in energy absorption, however, adding three layers into the middle of the specimen has a great value of impact energy which is around 77% higher than CFRP with no nylon 6 layers. However, adding extra nylon 6 layers also contributed to the extra thickness of the specimen. The average thickness of sample 1 to 5 was around 0.956 mm, 0.974 mm, 1.01 mm, 1.062 mm and 1.084 mm.

Because of its higher impact energy and comparatively thinner shape, Sample 5 once again shows the highest value when impact energy per thickness is taken into account, coming in at about 850 mJ/mm. A significant performance is also shown by Sample 4, which has an impact energy per thickness of about 750 mJ/mm. On the other hand, Sample 1 exhibits a small impact energy per thickness of roughly 600 mJ/mm, indicating a lower sample thickness in relation to the energy absorbed, even if it has the lowest impact energy.

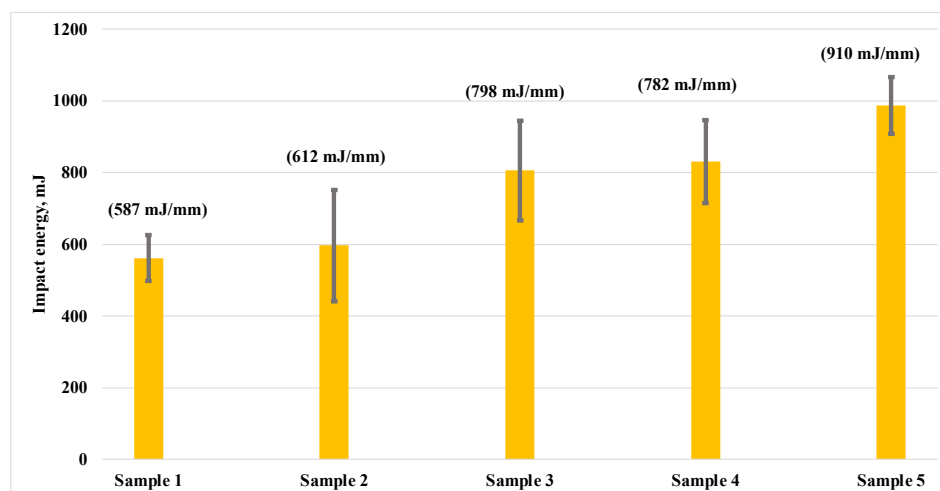


Fig. 5.12: Impact energy of specimens including and excluding nylon 6

5.3.3 Four-point bending test

The bending strength and bending modulus of the five samples are given in Fig. 5.13 and 5.14. The dark grey bars in Fig. 5.13 show bending strength of the samples in MPa, while the red bars in Fig. 5.14 portray the bending modulus in GPa. Each graph also shows the bending strength and bending modulus, expressed in MPa/mm and GPa/mm, respectively, divided by their corresponding thickness. With the bending strength of around 314 MPa, sample 2 (1 layer of nylon 6) showed the maximum bending strength, followed by sample 4 (3 layers of nylon 6), which had the bending strength of almost 258 MPa. Although samples 1, which is the thinnest sample and sample 5 which have bending strengths of 205 MPa and 237 MPa, respectively, their bending strengths per thickness are closer, at 214 and 218 MPa. The standard deviation from the repeat tests is shown in the error bars, which are brown lines that show the variability within the measurements. Overall, the trend indicates that the nylon 6 inclusion definitely increase the bending strength of carbon fiber/epoxy composites, and sample 2 which has one layer of nylon 6 performs better than others in terms of bending strength and bending strength per thickness. As a four-point bending test applied a gradual and controlled load to the material, sample 1 has the maximum resistance to deformation than other samples and it can operate better at low strain rate and under gradual loading. It is easier for nylon 6 to melt and react with epoxy if it is applied to the specimen in a single layer. The more nylon 6 is combined with epoxy, the more resilient the specimen will be. Nylon 6 added in many layers is less likely to mix with epoxy and become weaker over time than when added in a single layer under gradual loading and low strain rates. From Fig. 5.14, it is also noticeable that sample 2 has the highest bending modulus of around 45 GPa, whereas sample 1 has a bending modulus of roughly 35 GPa. The bending moduli of 40 GPa and 39 GPa, respectively, are comparable in Samples 4 and 3. At 34 GPa, Sample 5 has the lowest bending modulus. With a bending modulus per thickness of roughly 45 GPa/mm, Sample 2 continues to hold the top spot, followed by Sample 4 at 39 GPa/mm. Because of its smaller geometry relative to the overall modulus, Sample 5 exhibits the lowest bending modulus per thickness. Overall, Sample 2 demonstrates the best performance in both bending modulus and modulus per thickness, suggesting its superior material properties in comparison to the other samples. However, the variations in bending modulus between the samples are not so significant.

Additionally, Fig. 5.15 shows the bending strength and modulus data under various hot-

pressing settings. In this case, the nylon 6 layers was first hot pressed at three different temperatures (150°C, 200°C and 220°C) and inserted in between the preforms. Only a single layer of nylon 6 was used in this case for comparison. The purple bars show the bending strength (left axis), and the green dotted line shows the bending modulus (right axis). The samples' bending strength rose as the hot press temperature rose, peaking at about 450 MPa at 200°C. The bending strength of the sample without hot pressing was approximately 250 MPa, and at 150°C, it was marginally greater than at 220°C, measuring about 380 MPa and 330 MPa, respectively. This indicates that 200°C is the ideal temperature for increasing bending strength. The trend indicates a consistent rise in bending modulus with an increase in hot press temperature. At roughly 35 GPa, the sample without hot pressing has the lowest bending modulus. The modulus rose to around 38 GPa at 150°C, 45 GPa at 200°C, and 50 GPa at 220°C. This suggests that increased hot press temperatures result in a noticeable improvement in stiffness. The standard deviation across replicate tests is reflected in the error bars, which are blue lines that show measurement variability. These findings imply that hot pressing improves both bending strength and modulus, with 200°C for bending strength and 220°C for bending modulus being the ideal temperatures for obtaining the greatest material performance.

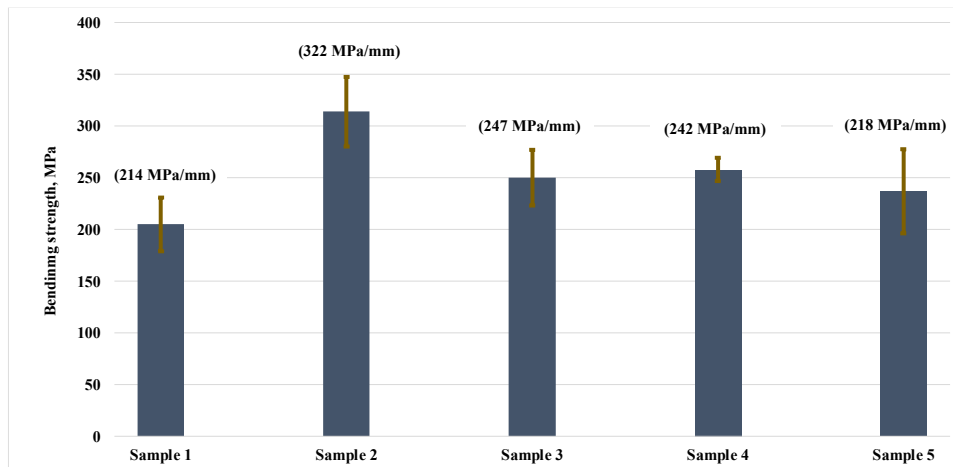


Fig. 5.13: Bending strength of specimens including and excluding nylon 6

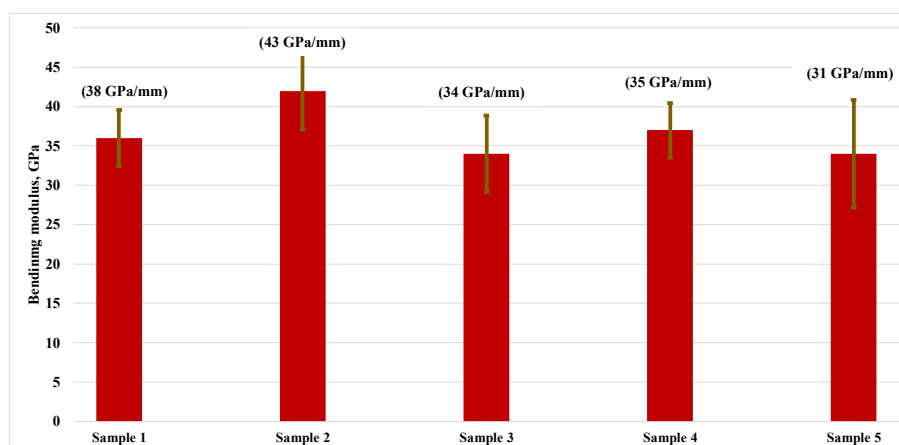


Fig. 5.14: Bending modulus of specimens including and excluding nylon 6

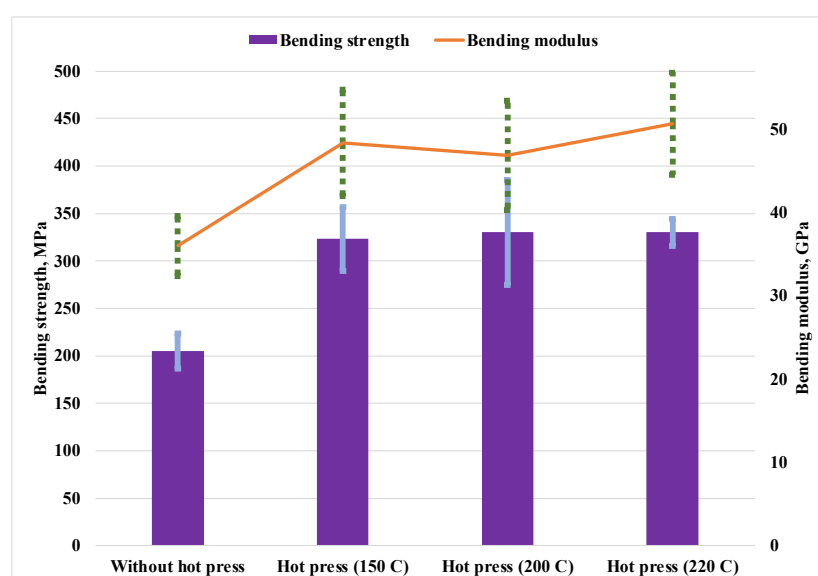


Fig. 5.15: Bending strength and modulus of specimen of specimens including and excluding of hot-pressed nylon 6

5.3.4 Roughness test

The images in Fig. 5.16 show the surface topography of the material as observed under laser isotope microscopy. The 3D surface plots display the height variation across the scanned area, with color gradients indicating different elevation levels. From Fig. 5.16 (a), it can be observed that the morphology of the surface is comparatively smooth, with a small amount of roughness. Localized areas exhibit mild elevations, indicating a generally level surface with minor undulations. The height variation ranges from 0.0 to 73.6 μm . With red denoting the highest

spots and blue denoting lesser elevations, the color scale shows a progressive change in height. The roughness parameter achieved for this specimen was $R_a = 5.5 \mu\text{m}$ and $R_z = 46.8 \mu\text{m}$. On the other hand, from Fig. 5.16 (b), it is noticeable that the surface in this picture exhibits more noticeable characteristics, such as sharper peaks and deeper grooves. The surface exhibits higher roughness and substantial structural heterogeneity, and the height variation ranges from 0.0 to $156.6 \mu\text{m}$. This points to a more uneven surface, which was probably caused by the particular procedure or circumstances employed when preparing the sample. These images provide detailed insight into the surface morphology, which can significantly impact the material's mechanical properties, such as adhesion, friction, and wear resistance. The roughness parameter achieved for this specimen was $R_a = 17.6 \mu\text{m}$ and $R_z = 112.7 \mu\text{m}$.

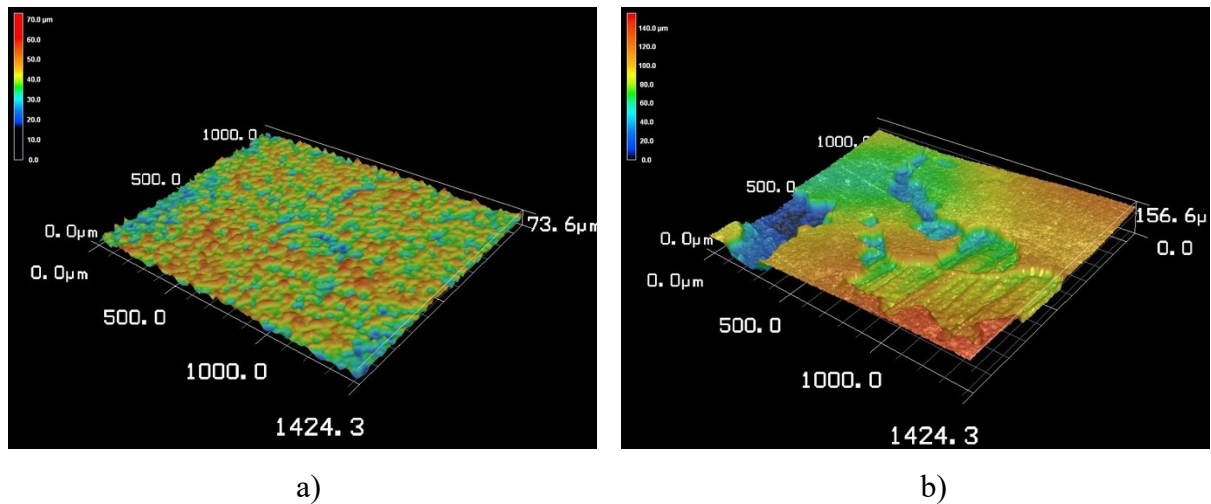


Fig. 5.16: Surface topography of specimen including and excluding nylon 6

The surface roughness parameters R_a (mean roughness) and R_z (maximum height of the profile) for samples with and without Nylon 6 inclusion are shown in Fig. 5.17 and Fig. 5.18. The measurements made at three distinct sample sizes 8 mm, 2.5 mm, and 0.5 mm are represented by the data points. Fig. 5.17 shows the mean roughness (R_a) for samples with and without Nylon 6. Triangles show measures that include Nylon 6, whereas diamonds show measurements that do not. The sample without Nylon 6 has a rougher surface than the sample with Nylon 6 ($\sim 12 \mu\text{m}$) at the 8 mm size, as shown by its higher R_a value ($\sim 16 \mu\text{m}$). With the sample containing Nylon 6 at about $8 \mu\text{m}$ and the one excluding it at about $10 \mu\text{m}$, the 2.5 mm samples exhibit a smaller variation in R_a . The tendency continues at the 0.5 mm size, where the excluding Nylon 6 sample has a higher R_a of around $6 \mu\text{m}$ than the including Nylon 6

sample, which is about $4\text{ }\mu\text{m}$. Fig. 5.18 presents the maximum height of the profile (R_z) for the same samples. The sample without Nylon 6 has a higher R_z value ($\sim 100\text{ }\mu\text{m}$) at the 8 mm size than the sample with Nylon 6 ($\sim 60\text{ }\mu\text{m}$). The excluding Nylon 6 sample has a R_z value of roughly $45\text{ }\mu\text{m}$ at 2.5 mm, whereas the containing Nylon 6 sample has a lower R_z value of roughly $30\text{ }\mu\text{m}$. The tendency holds true at 0.5 mm, with the Nylon 6-excluded sample having a higher R_z value ($\sim 20\text{ }\mu\text{m}$) than the Nylon 6-included sample ($\sim 15\text{ }\mu\text{m}$).

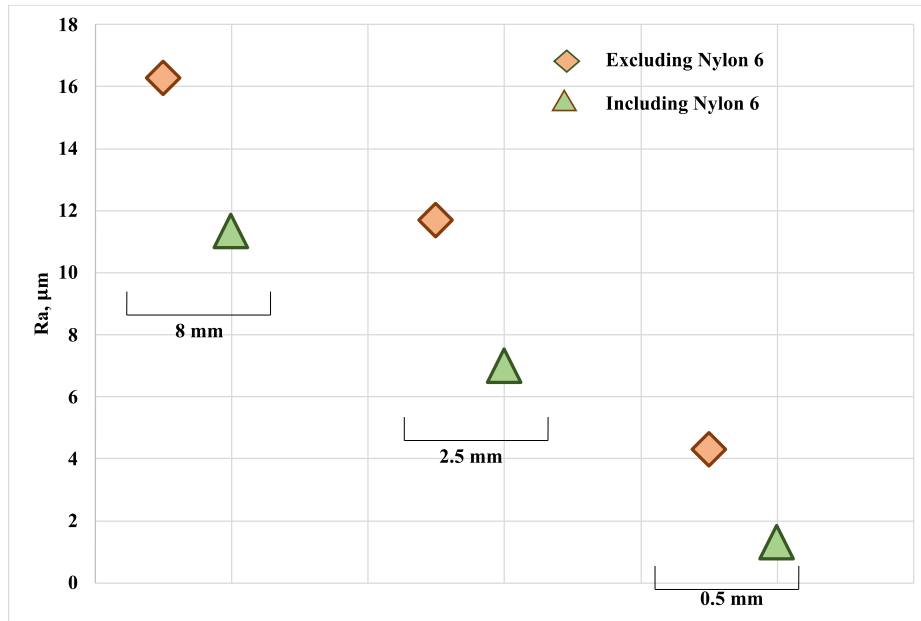


Fig. 5.17: Roughness parameter R_a of specimens including and excluding nylon 6

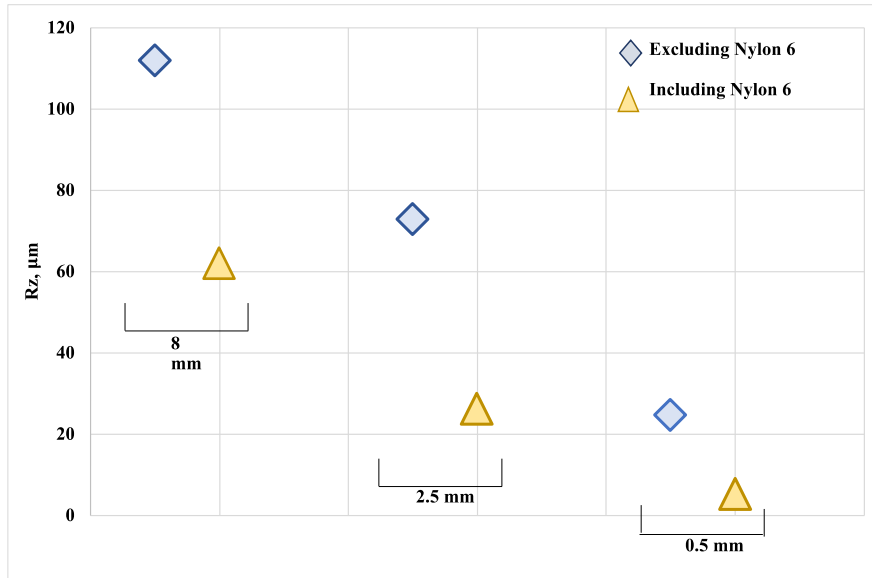


Fig. 5.18: Roughness parameter R_z of specimens including and excluding nylon 6

5.3.5 Raman Spectroscopy

A thorough comparison of the composite material and Nylon 6 is made possible by this Raman spectral analysis, which reveals both parallels and divergences in their vibrational modes, which are connected to their molecular structures and material characteristics. Fig. 5.19 presents the Raman spectrum of Nylon 6, showcasing its characteristic peaks. The x-axis represents the wavenumber (cm^{-1}), and the y-axis represents the intensity (arbitrary units). The corrected Raman spectra of Nylon 6 is shown by the blue line. Because of its molecular vibrational modes, it exhibits the wide, pronounced peaks that are typical of Nylon 6. Notable peaks that are typical of polymeric materials like Nylon 6 are seen at around 450 cm^{-1} , 1350 cm^{-1} , and 1600 cm^{-1} . The red circles show the particular peaks in the spectrum that have been found by previous researchers [208] and are utilized for additional comparison and analysis. The vibrational modes of several functional groups within the Nylon 6 structure are represented by these peaks. The red dashed vertical lines highlight the key peaks associated with Nylon 6. These are used for comparison with the composite material's spectrum to distinguish the specific contributions from Nylon 6 and the composite components. The distinctive peaks of Nylon 6, located at comparable wavenumbers in the composite spectrum, are shown by the vertical dashed red lines. These peaks serve as benchmarks for contrast. Raman spectra of the composites with and without Nylon 6 layers are shown in Fig. 5.20 and 5.21 respectively. In particular, the composite and Nylon 6 spectra both exhibit the peak at about 1350 cm^{-1} , which

is frequently linked to the D-band of carbon materials. This suggests that the two materials share structural characteristics or links. The composite and Nylon 6 spectra both share the G-band peak, which is located at about 1600 cm^{-1} . This suggests that at these particular wavenumbers, the composite has nylon 6 material embedded into its surface. Additional peaks or intensity shifts in the composite spectrum indicate the presence of additional components or materials. Due to the presence of additives or additional components, the composite material has characteristics that are different from those of Nylon 6, as evidenced by these variations in peak intensities and locations. Additionally, the surface of the specimen with and without nylon 6 that were observed by Raman spectroscopy is shown in Fig. 5.22.

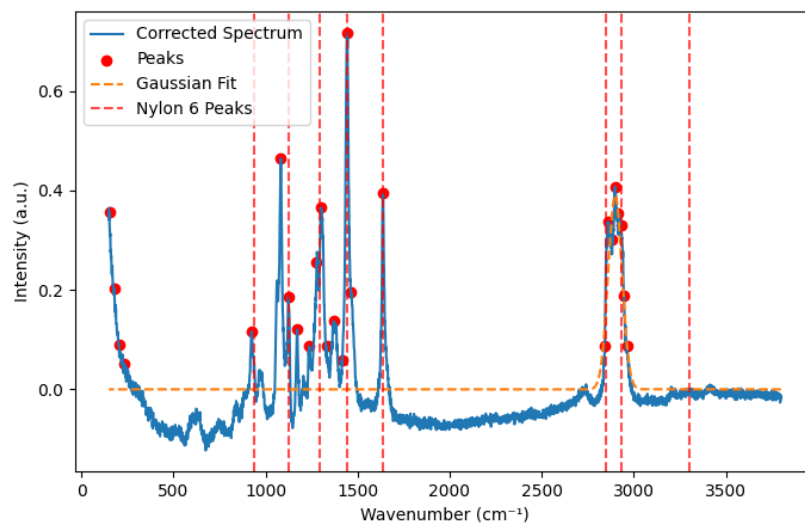


Fig. 5.19: Raman spectroscopy of nylon 6

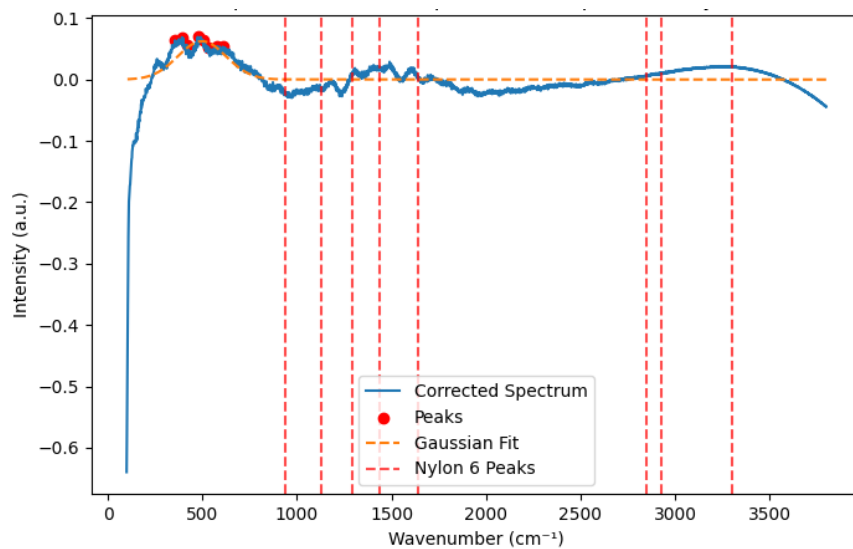


Fig. 5.20: Raman spectroscopy of specimen without nylon 6 and comparison with pure nylon 6

6

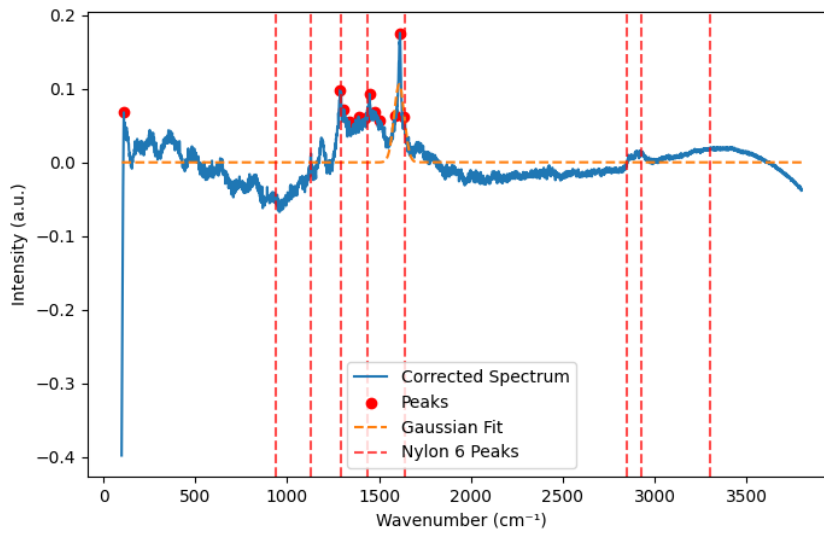


Fig. 5.21: Raman spectroscopy of specimen with nylon 6 and comparison with pure nylon 6

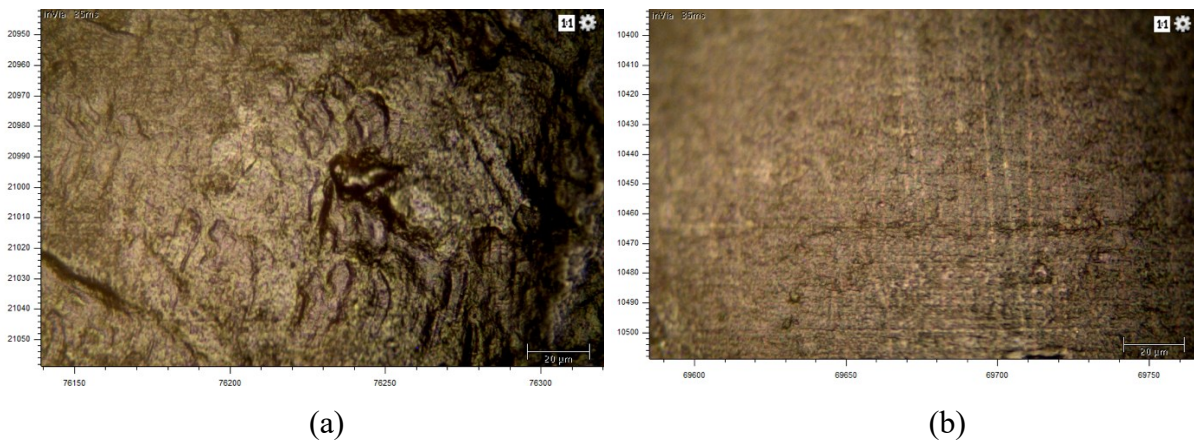


Fig. 5.22: Surface of specimen (a)without nylon 6 by Raman spectroscopy; (b) with nylon 6 by Raman spectroscopy

5.3.6 SEM

The material's microstructure and fracture surface are visible at different magnifications in the scanning electron microscope (SEM) pictures provided in Fig. 5.23. The interior morphology and the existence of important microstructural elements that influence the material's

mechanical performance are highlighted in the pictures. A wider perspective of the fragmented surface is shown by these pictures. The fracture lines, which show the locations of material failure, are apparent. The pictures show clear shear features along the fracture paths, indicating that the material has experienced severe deformation. Here in the bottom left image, the broken surface is viewed in greater detail, making the fracture morphology easier to see. The brittle character of the failure under the imposed stress is indicated by the visible broken fibers and the pull-out and delamination in some areas of the structure. This zoomed-in section emphasizes the presence of specific microstructural features, such as the fiber-matrix interaction and the fracture of individual fibers. The inset further highlights these features in more detail, revealing the intricate fiber pull-out and microcrack propagation. These SEM pictures provide important information about the material's behavior under stress and the fracture mechanisms. The material appears to experience a mixed failure mode, comprising both brittle fracture and fiber-matrix debonding, based on the observed fractures, which include fiber breaking and delamination.

The delaminated layers and the fiber-matrix interaction are highlighted in Fig. 5.24, which give a general view of the fractured surface. Clear fracture lines and zones of separation are seen throughout the sample, indicating the impact of interfacial weakness and shear stress. A closer look at the interior fracture characteristics is provided by the zoomed-in image. It displays intricate details like the multilayer structure and fiber pull-out evidence, which point to the mechanisms of failure. The material may have brittle failure as a result of stress buildup at the interfaces, as indicated by the clear separation of the layers and the existence of voids between the fibers.

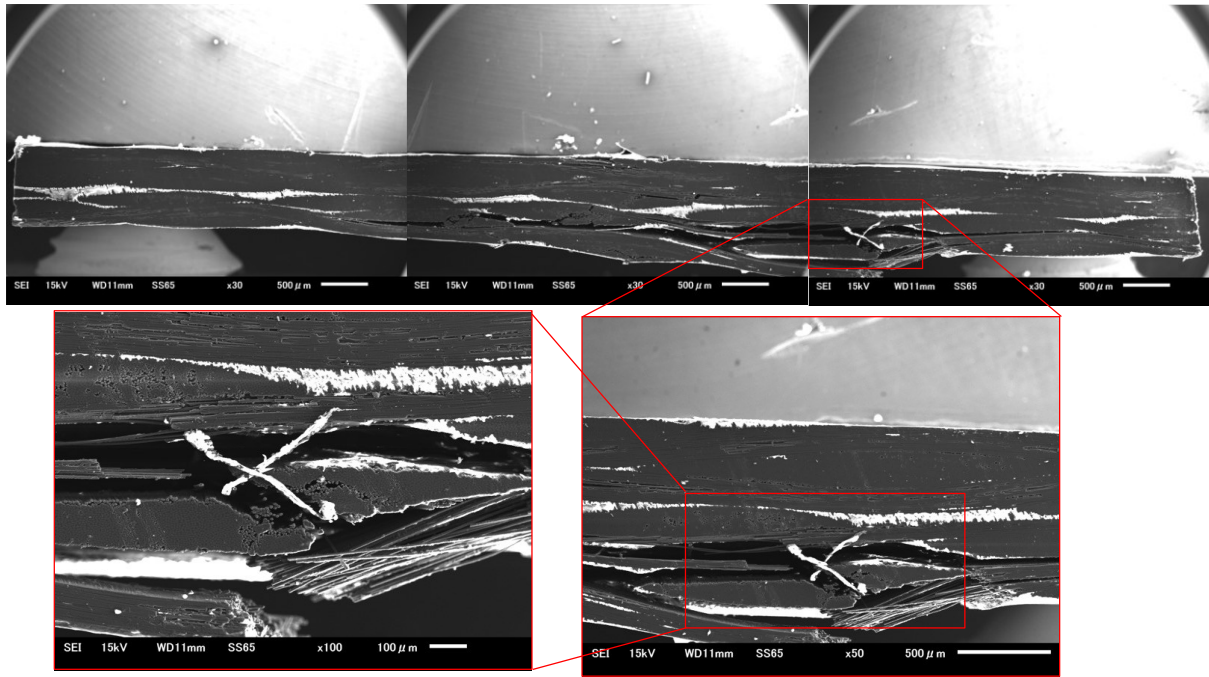


Fig. 5.23: SEM image of the cross-section of sample 1 after bending test

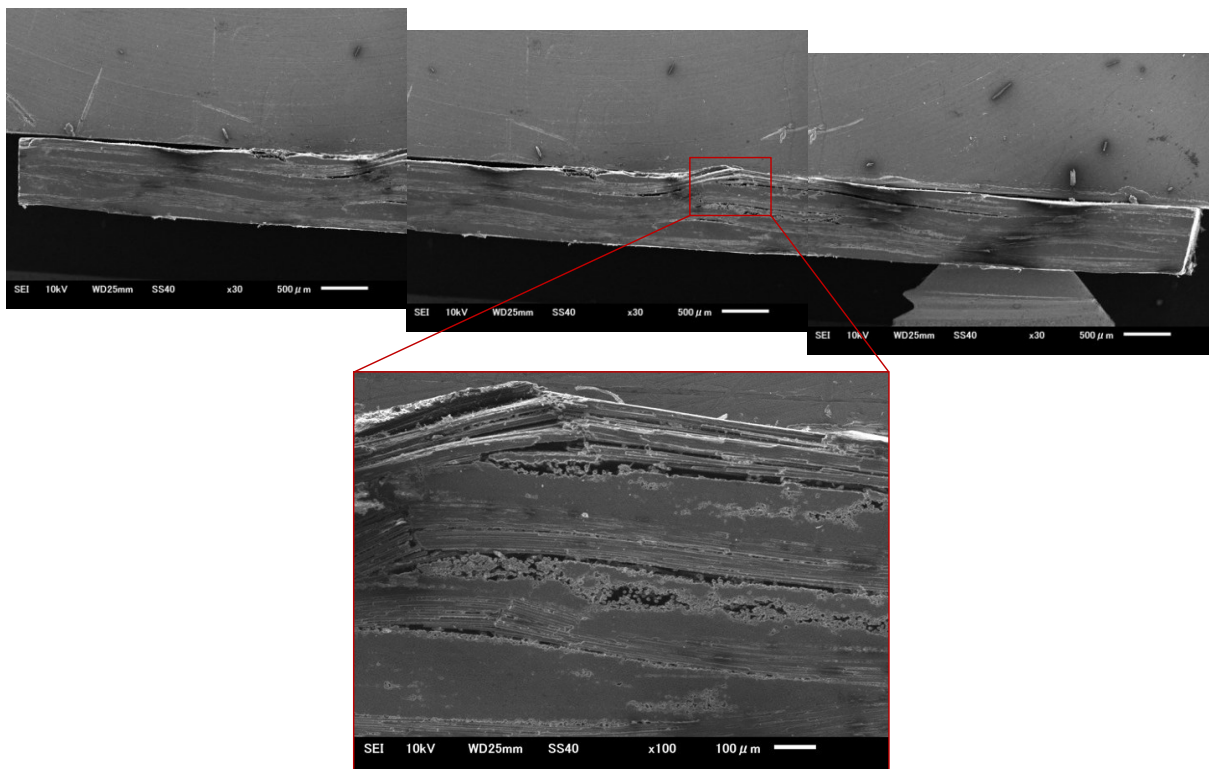


Fig. 5.24: SEM image of the cross-section of specimen with hot-pressed nylon 6 after bending test

5.3 Conclusion

This research focused on evaluating how the incorporation of Nylon 6 influences the performance of carbon fiber/epoxy composites produced through the electrodeposition resin molding (EDRM) technique. The experimental findings highlighted that introducing Nylon 6 into the composite matrix led to noticeable improvements in mechanical performance. Specifically, composites reinforced with Nylon 6 demonstrated higher bending strength and modulus than their counterparts made solely with carbon fiber and epoxy. This suggests that Nylon 6 contributes to improved resistance to deformation under applied loads, likely due to a synergistic interaction between the carbon fibers and the thermoplastic, which in turn boosts the composite's stiffness and load-bearing efficiency. Moreover, the presence of Nylon 6 had a positive effect on the impact resistance of the material. Composites containing this thermoplastic absorbed more impact energy, indicating enhanced toughness. These improvements align with the known properties of Nylon 6, which is often recognized for its ability to dissipate energy effectively during sudden or high-stress impacts. Microscopic analysis using scanning electron microscopy (SEM) further supported these findings. The images showed more uniform resin dispersion and better fiber-matrix adhesion in the Nylon 6-modified samples. Fracture surfaces exhibited reduced fiber pull-out and delamination, signaling stronger interfacial bonding between the carbon fibers and the surrounding matrix attributes typically associated with improved mechanical integrity. Additional insight was gained through Raman spectroscopy, which confirmed the successful incorporation of Nylon 6 by identifying distinct vibrational signatures corresponding to both the carbon fibers and the thermoplastic. Comparison with spectra of pure Nylon 6 revealed clear evidence of molecular integration within the composite, validating the structural presence of Nylon 6 in the upper surface of final material. In summary, the addition of Nylon 6 to carbon fiber/epoxy systems via EDRM not only enhanced key mechanical properties such as flexural strength, stiffness, and impact absorption but also contributed to improved interfacial bonding and overall durability. These results position Nylon 6 as an effective toughening agent capable of improving the structural resilience and reliability of CFRPs, making the material more suitable for demanding applications where both high strength and impact resistance are critical.

5.5 Future Studies

Although the mechanical and interfacial enhancements brought about by adding Nylon 6 layers

to carbon fiber/epoxy composites were effectively established in this study, a number of promising directions still need to be investigated. The compatibility of epoxy with different thermoplastics, including polyethylene, polyester, polypropylene, polyvinyl chloride, and others, can be compared in this study. For a more thorough investigation, additional techniques like transmission electron microscopy, thermogravimetric analysis, Fourier transform infrared spectroscopy, etc., could be used. Besides, a chemical analysis of the compatibility of nylon 6 and epoxy can be in interesting directions as well.

Chapter Six

Concluding Remarks and Future studies

6 Conclusion

6.1 Conclusion

This research has explored a novel and scalable approach to prepare different types of CFRP composites with high performance by using electrodeposition resin molding technique. Unlike other conventional composite manufacturing methods, EDRM not only just offers a promising route for easier resin impregnation technique, but also an efficient technique to produce high quality composites. The advantages of EDRM such as low void formation, good fiber-matrix adhesion, easy handling etc. make it suitable for mass production of CFRP which is a weighty need nowadays in several structural industries. The flexibility of preparing complex shaped composites, 3D structures and layers adhesion has made EDRM stand out from other conventional techniques. The necessity of using the EDRM perfectly with its best condition, the study about its process parameter was essential. On the one hand, the evaluation of changing the voltage and current conditions of EDRM was necessary as these electrical conditions play the vital role of resin precipitation on the fiber surface. On the other hand, as EDRM handles the thermoset resin for the matrix part of the composites, heat curing plays a very significant role in the composite qualities. Consequently, the evaluation of these heat curing temperatures and heat curing period was also required. Hence, this study initially investigated the effect of these manufacture variables of EDRM on the composite properties especially the static and dynamic characteristics of composites. Vibration test was performed to check the dynamic properties of the composite by which the natural frequencies and damping of the composites were obtained. In addition, Young's modulus, bending modulus, and bending strength were compared using tensile tests and four-point bending tests. The initial voltage and current condition of EDRM was 100V and 0.50A respectively. Then the voltage and current were changed to observe the influence. The vibration test and four-point bending test were done in accordance with this. When studying the impact of the heating conditions, 1.5A current conditions with three different voltage conditions were chosen from the four-point bending test. In terms of heating settings, the first heat curing temperature was 215°C which was changed to 230°C later. For each heating condition, three different periods: 4 hours, 6 hours and 8 hours were applied to inspect the effect of heat curing period on the composite properties. After that, the tensile tests were carried out with the best heating condition and best current condition.

The systematic evaluation of these EDRM parameters reveals that the electrical conditions such as voltage and current have negligible effect on the vibration and mechanical properties of the composites, however, both the heat curing temperature and heat curing period had significant impact on the bending properties of composites. According to the information above, it is possible to create CFRP using the electrodeposition resin impregnation process with mechanical properties that are on pace with or better than those of other autoclave and de-autoclave molding techniques.

As discussed earlier, this new EDRM technique needed to be explored more before establishing this method at an industrial level. Crucially, the research did not limit itself to standard composite laminates but extended EDRM's versatility to fabricate complex and multifunctional structures such as CFRP capacitors, curvilinear fiber-reinforced composites, and Nylon 6 hybrid composites. As required, after evaluating the process parameters of this method, preparation of a structural capacitor was conducted by using EDRM. Cellulose nanofiber was incorporated with CFRP to prepare this energy storing device which is demanding research in renewable energy industries currently. The incorporation of cellulose nanofibers (CNF) as a dielectric material demonstrated that EDRM can effectively produce lightweight structural capacitors with promising mechanical strength and energy storage capabilities. Along with CNF, other dielectric materials such as paper, tissue, jute fiber and plastics were also used for comparison purposes with CNF. Different applying methods of CNF were carried out to find the best way to apply CNF so that it does not affect the bending and tensile behavior as well. The CNF inclusion into the CFRP not only increased the electrical performance of the composites but also enhanced the mechanical properties as well which make it suitable as an energy storing device. The ability of EDRM to fabricate multifunctional composite structures is validated by this dual functionality. Good interfacial bonding and homogeneous resin distribution both essential for structural integrity and dielectric performance were made possible by the technique. These findings create new opportunities for lightweight parts in aerospace and automotive systems, where it's critical to combine functionality and weight reduction. However, the capacitance found in this research was quite deficient, further research for increasing the conductivity of the electrode and dielectric properties of the insulators should be conducted for using this device for energy storage. Nevertheless, this device can be used where small power is required and the structure can itself serve as an electronic device. In addition, they can be used to store energy and provide a rigid framework, which will reduce weight and space. They can also be used in place of the

lightweight structural component of aerospace or unmanned aerial vehicles (UAVs). Their long-term sustainability and mechanical resilience ultimately result in a lower lifetime cost, despite the fact that the materials may cost more to fabricate than for conventional capacitors. In conclusion, it can be said that EDRM can be used to produce multifunctional CFRP, which includes strength and capacitance, with no effect on the tensile properties.

Furthermore, EDRM was also employed for preparing a curvilinear fiber reinforced polymer composite. Different curvilinear pattern was sewn by using tailored fiber placement (TFP) machine. Sine and parabola pattern were created to check the effect of fiber paths with unidirectional carbon fiber. The vibration characteristics of these curvilinear fiber-based composites were evaluated by both experimentally and numerically. The results revealed that the vibrational behavior of CFRP laminates was greatly affected by the addition of curved fiber pathways, particularly sine and parabola wave patterns. The curvilinear designs showed better energy dissipation properties, lower natural frequencies, and more distributed deformation than conventional unidirectional laminates. Good agreement between finite element simulations and experimental modal analysis supported the validity of the findings. The most flexible and vibration-dampening patterns were sine wave designs, which suggested that they are appropriate for dynamic and vibration-sensitive applications such precision equipment, robotic arms, and aeronautical panels. This work demonstrates that fiber path tailoring can be an effective technique for adjusting composite materials' dynamic response. All things considered, these results demonstrate how important fiber orientation is in adjusting CFRP structures' dynamic responsiveness. In situations where vibration dampening, energy distribution, and dynamic stability are more crucial, curvilinear designs particularly the sine pattern offer substantial benefits, even though unidirectional fiber alignment is still best for applications requiring high rigidity. This study highlights fiber path engineering's potential as a useful tool in composite design and paves the way for sophisticated, application-specific CFRP components.

Additionally, this EDRM technique was also employed to prepare CFRP containing both thermoset and thermoplastic polymers. EDRM basically handles epoxy resin which is a thermosetting resin, however, this study has prepared a thermoplastic composite reinforced by carbon fiber. The effect of adding extra thermoplastic into carbon fiber reinforced composites was studied in this conduct. Additional Nylon 6 layers were inserted into carbon fiber/epoxy composites and their mechanical properties such as impact resistance, flexural strength,

young's modulus and surface morphology were assessed. Nylon 6 was chosen to add with carbon fiber/epoxy composites because nylon 6 can react with epoxy if the reaction is handled over 200°C. As the heat curing of the EDRM takes place in the incubator at around 230°C, the effect of the nylon 6 addition into the epoxy was examined. When Nylon 6 sheets were included using the EDRM approach into epoxy-based CFRP laminates, the impact resistance, bending strength, and general toughness were all noticeably improved. The hybrid structure leveraged the ductile and energy-absorbing nature of thermoplastic Nylon 6 while maintaining the stiffness and strength provided by carbon fibers and epoxy resin. The results show that adding the nylon 6 layer actually increased both the impact resistance and bending strength of the composite. However, if the nylon 6 layer is hot-pressed before adding the nylon 6 layer into the epoxy impregnated fiber, the bending strength improves more. Generally, the bending strength of carbon fiber/epoxy composite increased by more than 60% by adding hot-pressed nylon 6 layers. The nylon 6 gets melted due to high heat and pressure and gets mixed with the epoxy. Additionally, Raman spectroscopy confirmed the molecular incorporation of Nylon 6 into the composite matrix. From the Raman spectroscopy, the presence of nylon 6 was also found on the top of the surface of the composites. This basically indicates the mobility of nylon 6 as it gets melted due to heating. The melting of nylon 6 was further confirmed by the cross-sectional analysis carried out using scanning electron microscope (SEM). SEM analysis revealed improved fiber-matrix adhesion and reduced delamination, which contributed to better mechanical performance under both static and dynamic loading. Due to the melting of nylon 6, the surface becomes really smoother compared to the CFRP excluding nylon 6 layers. From the SEM images, the CFRP including hot-pressed nylon 6 layer showed the complete meltdown of nylon 6 layers while the nylon 6 layer without hot-pressing got partially melted. This result reveals that the melting of nylon 6 can actually toughen the epoxy more which further results in a more strength in bending and impact properties of composites. This method showed a practical way to create toughened CFRP structures that can be used in applications that need strength and longevity, like automobile parts, protective panels, and structural frames in challenging conditions.

Collectively, these results show that EDRM is flexible enough to create sophisticated, application-specific composites in addition to being appropriate for creating conventional CFRP laminates. The method has a lot of potential for developing lightweight structural materials for robotics, smart infrastructure, automotive, and aerospace applications.

6.2 Suggestions of Future research

Though the research that had been conducted in this study makes EDRM a promising composite manufacturing technique, further research is required to make this efficient in the industry level. Moreover, based on the promising results of this study, several future directions are evident:

As this method investigates the parameters such as electrical current, voltage and heat curing conditions on the composite properties, however, this method also depends on other parameters such as electrolyte density, temperature of the resin, position of the cathode, electrode geometry, electrolyte composition, electrode distance etc. A systematic study of the effect of these parameters can be conducted. Again, the effect of wider ranges of voltage and current would be an important conduct as well. Moreover, this study only compared the vibrational properties of composites prepared by EDRM and VaRTM. More detailed comparison with other composite preparation techniques and the assessment of mechanical, electrical and thermal properties can be carried out in future.

EDRM was also used to prepare a multifunctional composite by adding the CNF into CFRP composites. Although the mechanical and vibrational properties were the main emphasis of this study, adding thermal conductivity, electrical conductivity, and dielectric breakdown strength will give a more comprehensive understanding of the behavior of the composite for multipurpose applications. The electrical properties assessment was only done by capacitance testing, other methods for electrical property assessment can be carried out along with impedance spectroscopy over a broader frequency range. Measurement of multifunctional qualities can include thermal, morphological and biodegradable properties as well. The enhancement of conductivity of the electrode by adding other nanofillers can also be an interesting approach for this multifunctionality. Moreover, long-term performance evaluation under humidity, UV radiation, thermal cycling, and mechanical fatigue would be essential for deploying EDRM composites in real-world applications as well.

This approach also investigated the incorporation of curvilinear fiber paths specifically parabola and sine wave patterns into CFRP laminates and compared with unidirectional carbon fibers, nevertheless, other patterns such as circular, quadrant etc. should also be investigated and compared for a broad perspective for the curvilinear paths. Moreover, combining

curvilinear patterns with embedded electronics could serve as a new frontier in structural health monitoring systems.

Another phase of this research was to include nylon 6 into epoxy-based composite. This study can compare the compatibility of epoxy with other thermoplastics such as polyethylene, polyester, polypropylene, polyvinyl chloride etc. Additionally, other characteristics technique such as Fourier transforms infrared spectroscopy, transmission electron microscopy, thermogravimetric analysis etc. could be performed for more detailed analysis. The vibration characteristics and impact testing can be compared with numerical analysis to have a validation numerically as well. Moreover, pilot-scale or continuous EDRM systems can be developed for mass production of CFRP parts, potentially replacing more expensive autoclave-based processes in commercial sectors. Last but not the least, merging EDRM with additive manufacturing or hybrid layering could create complex, geometry-optimized composite structures for aerospace and defense industries.

In summary, this study not only demonstrates that the EDRM methodology is a viable substitute for conventional CFRP manufacturing processes, but it also creates a wide range of opportunities for further study and industrial innovation.

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Chapter 1

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