



This is to certify that the

dissertation entitled

Processing Discontinuous Fiber Polymer Composites:
Fiber Alignment using Electric Fields and
Microstructure-Property Relationships

presented by

Murty N. Vyakarnam

has been accepted towards fulfillment
of the requirements for

Ph.D. degree in Chemical Engineering

Laurence T. Dyal
Major professor

Date 12/2/96

LIBRARY
Michigan State
University

PLACE IN RETURN BOX to remove this checkout from your record.
TO AVOID FINES return on or before date due.

DATE DUE	DATE DUE	DATE DUE
JUN 01 6 2005 061605	_____	_____
DEC 06 2009 072510	_____	_____
_____	_____	_____
_____	_____	_____
_____	_____	_____
_____	_____	_____
_____	_____	_____

**PROCESSING DISCONTINUOUS FIBER POLYMER COMPOSITES:
FIBER ALIGNMENT USING ELECTRIC FIELDS AND
MICROSTRUCTURE-PROPERTY RELATIONSHIPS**

By

Murty Narayan Vyakarnam

A DISSERTATION

Submitted to
Michigan State University
in partial fulfillment of the requirements
for the degree of

DOCTOR OF PHILOSOPHY

Department of Chemical Engineering
Composite Materials and Structures Center

1996

Copyright by
Murty Narayan Vyakarnam
1996

e

H

c

a

a

c

m

a

m

a

D

A

ABSTRACT

PROCESSING DISCONTINUOUS FIBER POLYMER COMPOSITES: FIBER ALIGNMENT USING ELECTRIC FIELDS AND MICROSTRUCTURE-PROPERTY RELATIONSHIPS

By

Murty Narayan Vyakarnam

Discontinuous fiber polymer composites are finding widespread use because of their ease in processability and improvements in performance over un-reinforced polymers. However, their use is limited to non-structural applications due to the difficulties in controlling fiber orientation and fiber length reduction during melt processing of long fibers at high volume fractions. Micromechanics models indicate that the elastic behavior of an aligned discontinuous fiber composite should approach the behavior of a continuous fiber composite if the length of the reinforcement exceeds critical fiber length. This provided the motivation for research in the development of a process that has the potential to manufacture aligned discontinuous or short fiber polymer composites and subsequently investigate the microstructure-property relationships.

The unique combination of fiber alignment using electric fields in air and the recent advances in polymer powder processing resulted in a novel, high speed, solvent free Aligned Discontinuous Fiber (ADF) composite process. A semi-continuous laboratory prototype ADF process has been developed which has the following unit operations: alignment of

n

i

m

an

st

co

M

co

(t

conducting or insulating fibers in air using electric fields in an orientation chamber; polymer powder coating/impregnation of fibers, and; compression molding of the powder coated ADF preform into a composite. It has been found that alignment of dielectric E-glass fibers in air using electric fields is an extremely fast process (less than 1 second), but the non-buoyant nature of the fiber motion makes the alignment behavior very complicated and is a balance of polarization forces due to the electric fields; hydrodynamic forces due to fluid resistance, and; the rotational behavior of fibers during free fall. Chopped E-glass fibers of lengths ranging from 3 to 25 mm have been successfully aligned in air using A.C. electric fields of intensities ranging from 300 to 600 KV/m.

The ADF process has been demonstrated for chopped E-glass fiber and nylon12 matrix system and the properties of the composites fabricated using the process improve with an increase in fiber alignment and an increase in fiber length. Improvements in modulus and strength values of the ADF composites with fiber alignment ranged from 70 to 100 %, when compared to equivalent composites that were manufactured with random fiber orientation. Micromechanics analysis based on fundamental reinforcement theories, indicated that in the chopped fiber-thermoplastic systems, it is the effective aspect ratio of the fiber aggregate (bundle) that controls the elastic behavior of ADF composites.

To my mother
and
in loving memory of my father.

ACKNOWLEDGMENTS

I would like to express my sincere appreciation and gratitude to Professor Lawrence T. Drzal for the guidance, vision and support he provided throughout my graduate research at Michigan State University. I thank him for the many opportunities he provided that helped me grow as a better researcher and achieve the goals of my education.

I would like to thank all my committee members: Prof. C. Petty, Prof. K. Jayaraman, Prof. P. Duxbury and Prof. J. Asmussen for their valuable guidance and input from time to time during my doctoral research. In particular, I want to thank Prof. Petty and Prof. Jayaraman for their ardent desire to impart knowledge and enhancing my understanding in the areas of fluid flow and polymer rheology. My thanks are also to Prof. Duxbury and Prof. Asmussen for the many useful suggestions they provided on harnessing electro-magnetic fields for my research.

Many individuals helped me in developing the Aligned Discontinuous Fiber (ADF) composite process. In particular, I would like to thank the following individuals: John Brandon of MSU Cyclotron for introducing me to the practical aspects of high voltage engineering; Mike McLean of the DER machine shop for the timely fabrication of various

con

in

wa

pro

Min

dur

NS

Ma

State

to th

up a

prov

sacri

and e

Anan

a fell

suppo

components that helped me build the process; Richard Wagner for experimental assistance in summer '95; and Dr. Serban Petcu for helping me with the high speed video studies. I want to thank Bob Schweizer of Owens Corning and Philip Chu of Vetrotex CertainTeed for promptly supplying me with glass fibers of specific requirements. I would also like to thank Mike Rich and Brian Rook for their excellent cooperation and assistance provided to me during these last several years. Finally, I acknowledge the financial support provided by the NSF Center for Low-Cost High-Speed Polymer Composite Processing and Michigan Materials & Processing Institute.

I was fortunate to have many friends and colleagues, who made my stay at Michigan State and the Composite Center in particular, intellectually and socially stimulating. I want to thank Sanjay, Rik, Himanshu, Mark, Brent and Ed for sharing and bearing with me many up and downs in this phase of life and making it a very memorable one.

Finally, I want to thank my mother for the inspiration and encouragement she provided me to set higher goals in life. I dedicate this dissertation to her for the many sacrifices she made to offer me good education. I also want to thank my sisters for their love and encouragement throughout my education. I want to extend special thanks to my wife Anandita, for her love and friendship ever since we met in graduate school. With she being a fellow doctoral student, she understood the tribulations of research and provided excellent support in the hours of crisis.

I

I

C

Ch

Chapt

TABLE OF CONTENTS

List of Tables	xi
List of Figures	xii
Chapter 1. Introduction	1
1.1 Introduction	2
1.2 Discontinuous Fiber Composites	3
1.2.1 Fiber Length	5
1.2.2 Fiber Orientation	7
1.2.3 Fiber Volume Fraction	8
1.2.4 Polymer Matrix	9
1.3 Processing Vs Performance	11
1.4 Research Motivation	15
1.5 Novel Processing Approach	16
1.6 Dissertation Outline	18
References	19
Chapter 2. Review of Powder Processing and Fiber Alignment Techniques	20
PART A: Polymer Powder Processing of Composites	21
2.1 MSU Powder Process	22
2.2 Charging of Polymer Powders	26
PART B: Fiber Alignment Techniques	28
2.3 Flow Induced Alignment	28
2.3.1 Slurry / Hydraulic Based	28
2.3.2 Spray-up or Pneumatic	31
2.3.3 Molding and Extrusion	31
2.4 External Field Induced Alignment	33
2.4.1 Using Electric Fields	33
2.4.2 Using Magnetic Fields	37
2.5 Summary and Discussion	37
References	40
Chapter 3. Fiber Alignment Using Electric Fields	42
3.1 Introduction	43
3.1.1 Electric or Magnetic Fields?	44
3.1.2 Fiber Dimensions	46

3.2	Theory of Fiber Orientation	48
3.2.1	E-Field Polarization	49
3.2.2	E-Field Alignment Time	52
3.2.3	Fiber Hydrodynamics	55
3.2.3.1	Orientation Behavior of Fibers During Free Settling ...	55
3.2.3.2	Fiber Motion in Shear Flows	58
3.2.3.3	Settling Velocity of Fibers	59
3.3	Experimental Studies	61
3.3.1	Experimental Setup	62
3.3.2	Behavior in D.C. Fields	65
3.3.3	Behavior in A.C. Fields	69
3.3.3.1	Effect of E-field Intensity and Fiber Aspect Ratio	70
3.3.4	Fiber Settling Behavior	82
3.4	Summary of Results	86
	References	88
Chapter 4.	Aligned Discontinuous Fiber (ADF) Composite Process	90
4.1	Novel Concept	91
4.2	ADF Process Design	94
4.2.1	Unit Operations	94
4.2.2	Fiber Feeder	97
4.2.2.1	Fiber Pre-alignment	99
4.2.3	Electric Field Orientation	101
4.2.3.1	Orientation Chamber	101
4.2.3.2	Electrode Design	102
4.2.3.3	High Voltage Source	106
4.2.4	Deposition Platform	108
4.2.5	Powder Coating	110
4.3	Process Performance	112
4.3.1	Fibers	113
4.3.2	Polymer Characterization	114
4.3.3	Operating Conditions	114
4.3.4	Fiber Orientation Distribution (FOD) Measurement	117
4.3.5	Aligned Vs Random	121
4.3.6	Concentration Effects	123
4.4	Process Consolidation	128
4.4.1	Process Consolidation Cycle	130
4.4.2	Consolidation Pressure	130
4.5	Manufacturing	133
4.5.1	Processing Rates	133
4.5.2	High Speed Manufacturing Schemes	136
4.6	Summary	138
	References	141

Chapter 5.	Microstructure-Property Relationships	142
5.1	Introduction	142
5.1.1	Microstructure-Property Relationship Flowchart	145
5.2	Microstructure Descriptors	147
5.2.1	Fiber Orientation Distribution	150
5.2.2	Fiber Aspect Ratio	152
5.2.3	Fiber-Matrix Interaction	154
5.3	Elastic Property Predictions	156
5.4	Reinforcement Models	160
5.4.1	Shear Lag Theory	161
5.4.2	Eshelby's Inclusion Approach	164
5.4.3	Halpin-Tsai's Relationships	166
5.5	Experimental Results	169
5.5.1	Sample Preparation and Tensile Testing	169
5.5.2	Effect of Fiber Alignment	169
5.5.3	Effect of Fiber Length	170
5.6	Model Predictions and Discussion	170
	References	177
Chapter 6.	Conclusions and Future Work	179

T

T

T

T

T

T

T

T

T

T

T

LIST OF TABLES

Table 1.1	Typical Properties of Reinforcing Fibers	4
Table 1.2	Typical Properties of Polymer Matrices'	10
Table 2.1	Triboelectric Series of Polymers	27
Table 3.1	Pertinenet Fiber Properties'	45
Table 3.2	Behavior of Fibers in D.C. Fields	67
Table 4.1	Typical Operating Variables	116
Table 5.1	Constituent Properties	149
Table 5.2	Volume Fraction and Void Fraction of ADF Composites	149
Table 5.3	Actual Fiber Orientation Distributions of “Aligned” and “Random” ADF composites.	151
Table 5.4	Fiber Bundle Length and Aspect Ratios	154
Table 5.5	Model Predictions Compared with Experimental Modulus Results. . .	174

1

2

3

4

5

6

7

8

9

10

11

12

13

14

LIST OF FIGURES

Figure 1.1	Concept of critical fiber length based on Kelly-Tyson model.	6
Figure 1.2	Stress transferred to a fiber as a function of critical aspect ratio.	6
Figure 1.3	Performance versus processability trends of polymer composite material systems.	12
Figure 2.1	Prototype MSU Powder Prepreg Process	24
Figure 2.2	MSU High Speed Powder Prepreg Process	25
Figure 2.3	ERDE Glycerine process for fiber alignment	30
Figure 2.4	Budd slurry process for preform manufacture	30
Figure 2.5	Process to make oriented chopped fiber mats using electric fields	35
Figure 2.6	Process for alignment of fibers using electric fields in dielectric fluid ..	35
Figure 3.1	Forces acting on a fiber in the orientation chamber	50
Figure 3.2	Schematic representation of the orientation state in free falling fibers ..	57
Figure 3.3	Experimental setup to investigate fiber alignment in electric fields	63
Figure 3.4	High speed videographs of free falling E-glass fibers (0.5" long) in the oreintation chamber recorded at 1000 frames/sec under two conditions. Top: Fiber alignment in A.C. field of intensity 400 KV/m. Bottom: Random fiber orientation under no E-field.	71
Figure 3.5	High speed videograph of free falling E-glass fibers (0.25" long) aligning in the oreintation chamber in an A.C. field of intensity 400 KV/m (recorded at 1000 frames/sec)	72

P

Fi

Fig

Fig

Fig

Fig

Fig

Figure

Figure

Figure

Figure

Figure

Figure 3.6	Fiber orientation distribution of E-glass fibers settled at the bottom of the orientation chamber at A.C. field intensities of 300, 400 and 500 KV/m (Top FODs for 1" fibers; Bottom FODs for 0.25" fibers)	74
Figure 3.7	High speed videographs showing greater tendency of 0.5" fibers in attaining a stable horizontal fiber orientation state than 0.25" fibers.	76
Figure 3.8	High speed videographs taken by two cameras showing the natural tendency of 1" long fibers in attaining a stable horizontal orientation state. Each frame is a composite shot of the top view (right) and side view (left)..	77
Figure 3.9	Alignment time simulations based on Demetrides and Fuchs models.	81
Figure 3.10	Settling velocities of fibers at observation point B.	84
Figure 3.11	Settling velocities of fibers at observation point C	85
Figure 4.1	Novel concept of Aligned Discontinuous Fiber (ADF) composite processing	92
Figure 4.2	Schematic of the prototype Aligned Discontinuous Fiber (ADF) composite process.	95
Figure 4.3	Fiber feeder cum pre-aligner.	98
Figure 4.4	Mechanism of fiber pre-alignment.	100
Figure 4.5	Electrode edge effect on fiber orientation.	103
Figure 4.6	Electric field near the electrode edge.	105
Figure 4.7	Electrode design with a neutralizing field for continuous processing.	107
Figure 4.8	Circuit diagram for high voltage distribution.	109

Fig

Fig

Fig

Fig

Fig

Fig

Fig

Fig

Fig

Fig

Fig

Fig

Fig

Figure 4.9	SEM micrographs of E-glass fiber bundles coated (Top) and uncoated (Bottom) with nylon-12 powder..	111
Figure 4.10	Digital images of 1" long chopped E glass fibers settled on the deposition platform. Left: Aligned in E-field of 400 KV/m, Middle: Random with no E-field, Right: Manually aligned.	115
Figure 4.11	Fiber orientation distribution measurement.	118
Figure 4.12	FODs of different fiber length E-glass fibers that have aligned in E-field of intensity 400 KV/m. The control case of random orientation is also shown for comparison..	122
Figure 4.14	“Volume of influence” surrounding a fiber under two different orientation states: random and Pre-aligned.	126
Figure 4.15	Non-interacting fiber concentration as a function of aspect ratio.	127
Figure 4.16	Typical consolidation process cycle of a preform with a semi-crystalline polymer matrix..	129
Figure 4.17	Schematic of proposed continuous ADF process incorporating polymer powder coating; Fiber alignment using electric fields; Continuous ADF composite sheet manufacture..	137
Figure 5.1	Microstructure-property relationship flowchart.	146
Figure 5.2	“Bundle” reinforcement effect due to in-compatible fiber-matrix sizing.	155
Figure 5.3	“Filament” reinforcement effect due to compatible fiber-matrix sizing	155
Figure 5.4	Micro-laminate concept for elastic property determination of ADF composites.	158
Figure 5.5	Prediction of modulus for aligned discontinuous fiber composites as a function of the reinforcement aspect ratio.	168

Figure 5.6	Effect of fiber alignment and fiber length on the tensile modulus.	171
Figure 5.7	Effect of fiber alignment and fiber length on the tensile strength.	172

Introduction

1.1	Introduction	2
1.2	Discontinuous Fiber Composites	3
1.2.1	Fiber Length	5
1.2.2	Fiber Orientation	7
1.2.3	Fiber Volume Fraction	8
1.2.4	Polymer Matrix	9
1.3	Processing Vs Performance	11
1.4	Research Motivation	15
1.5	Novel Processing Approach	16
1.6	Dissertation Outline	18
	References	19

1.1 Introduction

Composite materials are a preferred combination of two or more different materials or identifiable phases which possess synergistic advantages over the performance of individual constituents. In the case of high performance composites, the reinforcing phase consists of high modulus fibers and the matrix phase consists of a polymer, metal or ceramic material. Composite materials are fundamentally different from monolithic materials in terms of the spatial variation of properties due to the variation in the microstructure of the constituents which provides tremendous opportunities to utilize these materials in a variety of situations. Polymer composites in particular, are increasingly replacing traditional materials in many areas ranging from aerospace, automotive, infrastructure to sporting goods and bio-medical applications due to their superior weight to stiffness and strength ratios; resistance to chemical degradation; and better toughness properties. Beyond the superior properties of fiber reinforced polymer composites, the reason for their success and bright future lies in the fact that unlike monolithic materials, the properties of these materials can be tailored by varying the microstructural features in particular the fiber orientation and fiber length.

The focus of this research is on the development of a process that can manufacture aligned discontinuous fiber composites, where the fibers can be aligned using electric fields. This chapter will give an overview of discontinuous fiber composites with an emphasis on factors that influence both processing and performance. Then the research motivation and objectives of this dissertation will be presented.

1.

dis

co

ori

a q

(we

rein

a g

dim

the

the

perf

disce

the n

On t

proce

there i

and th

those f

1.2 Discontinuous Fiber Composites

The reinforcing fibers of a composite material can be in a continuous or discontinuous form and offer a wide range of properties (Table 1.1). In the case of continuous fiber reinforcement the composite can be in the form of a laminate with the orientation state of the fibers ranging from unidirectional to a lay-up sequence that results in a quasi-isotropic laminate. Alternatively, continuous fibers can also be in a textile form (woven, knitted or braided) with a wide range of performance capabilities. Textile or fabric reinforced composites are popular because of the processing advantages they offer such as a greater degree of freedom in controlling precisely the fiber orientation state in a two dimensional state. However, the process of weaving and stitching is an expensive step and the inferior behavior of these composites in compression compared to tension are some of the drawbacks. When the fibers are discontinuous, there is a significant shift in the performance as well as the processing envelope of these composites. The physical discontinuity in the fiber length acts as a source for discontinuity in the stress-transfer from the matrix to the fiber which results in the reduction of composite mechanical properties. On the contrary, the discontinuous nature of the fibers offers tremendous advantages in processing of discontinuous fiber composites.

Discontinuous fiber composites are also referred to as short fiber composites, but there is no rigorous basis for the distinction between the words “discontinuous” and “short” and the two words are often used interchangeably¹. Certain classes of composites especially those from injection molding processes are generally referred to as short fiber composites,

while

fiber

comp

comp

and the

of proc

and the

discon

perfect

Table 1.1 Typical Properties of Reinforcing Fibers^{2,3}

Fiber	Diameter (μm)	Sp. Gravity	Modulus (GPa)	Strength (GPa)
PAN Carbon - AS4	8	1.8	248	4.07
PAN Carbon - IM7	5	1.78	301	5.31
Glass E	10 - 13	2.55	72.4	3.45
Glass S	10 - 13	2.49	86.9	4.3
Aramid*	11.9	1.45	131	3.62
Polyethylene**	27	0.97	172	3.0
Boron	140	2.7	393	3.1
Stainless Steel	10 - 50	7.9	190	1.4 - 2.1
β -SiC Whiskers	0.1 - 0.5	3.15	550 - 830	6.9 - 34.5
Carbon Whiskers	< 1.0	2.25	145	20.7

* Kevlar (DuPont); **Spectra 1000 (Allied Signal)

while whisker reinforced metal matrix composites are generally referred to as discontinuous fiber composites. When the reinforcing fibers are typically in aggregates or bundles the composites are referred to as chopped fiber composites or long discontinuous fiber composites when the length of fiber bundles exceed half inch.

Fiber lengths vary anywhere from 0.1 cm to 5 cm in discontinuous fiber composites and the orientation state can vary from completely random to perfectly aligned. Rheology of processing is a function of the fiber aspect ratio, orientation state, the fiber volume fraction and the viscosity of the suspension medium. Incidentally, the mechanical performance of the discontinuous fiber composite is also closely related to the above parameters. So, it makes perfect sense to keep the issues of processing and performance close together in

discontinuous fiber composites. The microstructural features of discontinuous fiber composites that are critical in processing and performance are introduced in the following sections.

1.2.1 Fiber Length

The length of the fibers that are used in discontinuous fiber composites can vary widely depending on the process and the type of fibers. Since, length of the fiber is an extremely important parameter in the performance of discontinuous fiber composites, a simple yet a very intuitive concept known as the *critical fiber length* will be introduced at this stage while detailed analysis will be presented in Chapter 5. According to reinforcement due to slip, in a representative volume element in a perfectly aligned discontinuous fiber composite, the tensile stress (σ_c) is transferred from the matrix to the fiber through interfacial shear stress (τ) near the fiber ends assuming a perfect bonding between the fiber and the matrix (Figure 1.1). This analysis is simple because it assumes the matrix to be ideally elastic-plastic and hence the interfacial shear stress is a constant value⁴. The minimum fiber length in which the fiber of diameter d_f can attain maximum allowable stress (σ_{fu}) is defined as the critical length, l_c for an average interfacial shear stress of τ_y .

$$\frac{l_c}{d_f} = \frac{\sigma_{fu}}{2\tau_y} \quad (1)$$

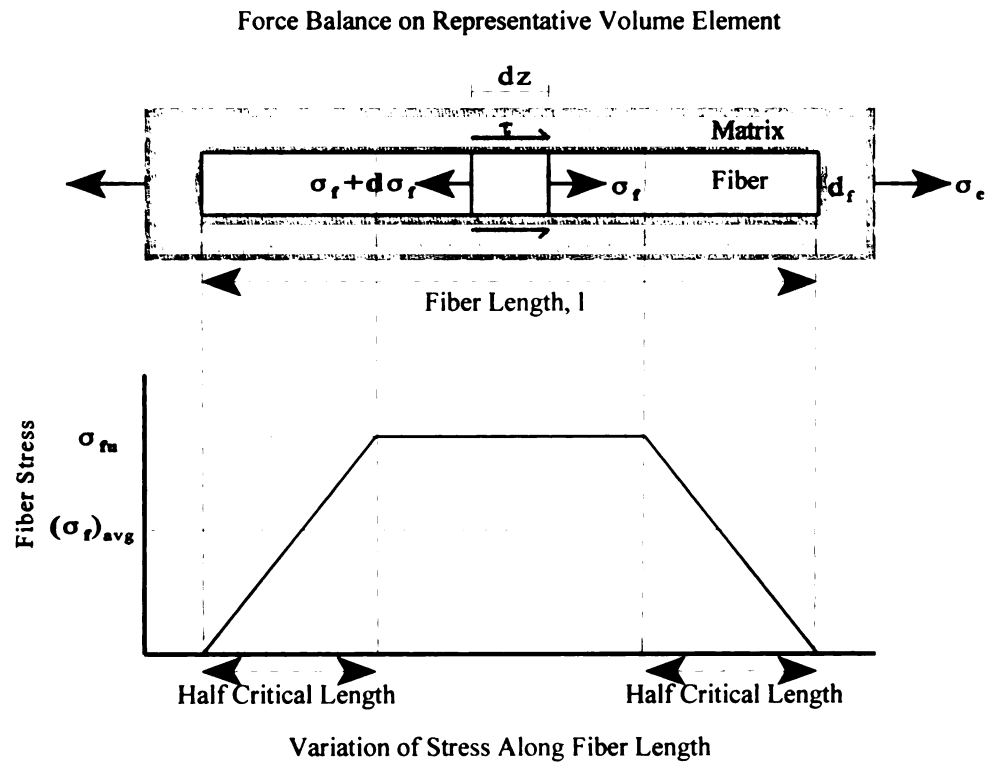


Figure 1.1 Concept of critical fiber length based on Kelly-Tyson model.

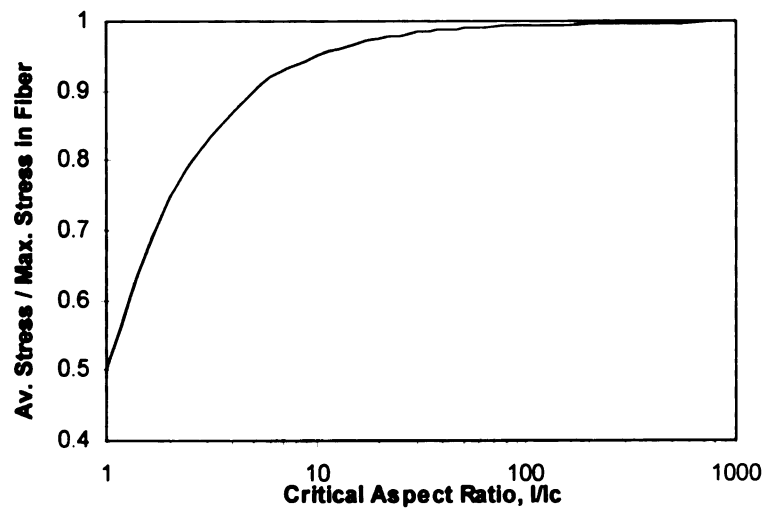


Figure 1.2 Stress transferred to fiber as a function of critical aspect ratio.

Below the critical length, the full reinforcement potential of the fiber is not realized but as the length (l) increases beyond l_c it can be easily shown that the average stress level in the fiber $(\sigma_f)_{av}$ approaches the maximum attainable in a continuous fiber, $(\sigma_f)_{max}$ (Figure 1.1). The average fiber stress $(\sigma_f)_{av}$ is obtained from the area under the curve in Figure 1.1 and is given by the following relation ⁵.

$$(\sigma_f)_{av} = (\sigma_f)_{max} \left[1 - (1 - q) \frac{l_c}{l} \right] ; \quad q = \frac{1}{2} \quad (2)$$

From Eq. 1.2, it can be seen that as the length of the fiber increases, $(\sigma_f)_{av}$ approaches $(\sigma_f)_{max}$ (Figure 1.2). From this stress analysis it can be concluded that the performance of discontinuous fiber composites approaches that of continuous fiber composites when fiber length exceeds 50 times the critical length and when the fibers are perfectly aligned.

1.2.2 Fiber Orientation

Fiber orientation becomes the dominant factor in influencing the composite performance once the fiber lengths exceed the critical length. To get an idea of the influence of fiber orientation, let us consider the more straight forward case of continuous fiber composite lamina at different fiber orientations. Piggott⁶ summarizes the off-axis properties of composites based on engineering constants, which can be used to predict the composite properties at an angle θ from the principal axis. Based on the experimentally determined engineering constants of longitudinal (E_{11}), transverse (E_{22}), and shear (G_{12}) moduli and the major Poisson ratio (ν_{12}), the off-axis tensile modulus, E_θ is given by:

$$\frac{1}{E_{\theta}} = \frac{\cos^4\theta}{E_{11}} + \left(\frac{1}{G_{12}} - \frac{2\nu_{12}}{E_{11}}\right)\sin^2\theta\cos^2\theta + \frac{\sin^4\theta}{E_{22}} \quad (3)$$

From this relationship it can be shown that there is a tremendous drop in the stiffness properties of composites as the fibers are mis-aligned from the direction of the applied stress. In the case of discontinuous fiber composites, it is important to note that the effect on stiffness and strength is expected to be similar and it will be a function of fiber aspect ratio also. One of the objectives of this research is to see the effect of fiber orientation distribution on the elastic behavior of discontinuous fiber composites.

1.2.3 Fiber Volume Fraction

The volume fraction of the short fibers in the composite affects both the final mechanical performance of the discontinuous fiber composite and the rheology of flow at the processing stage. Typically, the fiber volume fraction (V_f) varies between 50 and 75 % for continuous fiber composites, while in discontinuous fiber composites it can vary anywhere from 10 to 60 %. The maximum theoretical volume fraction (V_{fmax}) that can be obtained for aligned continuous fibers is:

$$V_{fmax} = \frac{\pi}{4} = 0.785 \quad \text{for square array}$$

$$V_{fmax} = \frac{\pi}{2\sqrt{3}} = 0.907 \quad \text{for hexagonal array}$$

Hi

pa

fib

of

alig

alig

large

prop

1.2.4

distin

crysta

resins

are ba

polym

compo

shelf li

thermo

viscosi

Higher volume fractions cannot be processed because of the theoretical limitations in packing fibers in a given volume. The maximum packing volume fraction for discontinuous fibers that are not perfectly aligned will always be lower than 78.5% and will be a function of the fiber length, fiber orientation and fiber cross sectional shape and area. As fiber alignment improves, the ability to pack a higher volume fraction of fibers increases. Fiber alignment thus has the dual effect of allowing higher volume fraction, of fibers as well as larger aspect ratios, which result in significant improvements in the stiffness and strength properties of the composite.

1.2.4 Polymer Matrix

Polymers are broadly classified as thermosets and thermoplastics with further distinction made between the various thermoplastic polymers based on their degree of crystallinity. In the field of high performance advanced polymer composites, thermosetting resins have been the predominant choice, whereas a majority of the short fiber composites are based on thermoplastic resins. In terms of global consumption trends, 80 % of the total polymer shipments are thermoplastic resins, however, only less than 20% of advanced composites are based on thermoplastic resins.

High strain-to-failure, increased toughness, shorter molding cycles, infinite prepreg shelf life, recyclability and repairability are some of the advantages of thermoplastics over thermosets which has caused an increase in their utilization. However, the high melt viscosities of thermoplastics is a problem when it comes to processing advanced fiber

Table 1.2 Typical Properties of Polymer Matrices^{7,8}

Polymer	Type†	Tg (°C)	Tm / HDT (°C)	Tensile Modulus (GPa)	Tensile Strength (MPa)
Nylon 66	TP-SC	78	260	1.6 - 3.5	82
Nylon 12	TP-SC	40	175	1.24	35
Polypropylene	TP-SC	-10	176	1.5	31
HDPE	TP-SC	-20	132	1.23	34
PEEK*	TP-SC	143	360 - 400	3.2	100
Polycarbonate	TP-A	150	130	2.4	65
Epoxy**	TS		80	3.38	55 - 130
Polyester	TS		60 - 205	2.1 - 3.5	35 - 104
Vinyl ester	TS		93 - 135	3 - 3.5	76

† TP - Thermoplastic; SC - Semi-Crystalline; A - Amorphous; TS - Thermoset

* ICI; **Shell Epon 1072

composites with continuous fibers at high volume fractions. The advantages with thermosetting resins has been low shrinkage, excellent fiber-matrix adhesion, good chemical resistance, high thermal resistance and easy processability. On the other hand, the low viscosity monomeric composition of thermosetting matrices, lends itself to infiltration and coating processes subsequently which is cured to its final cross linked state through a chemical reaction.

From Table 1.2, one can conclude that polymers do not differ drastically when it comes to their mechanical performance. However, there are substantial differences among the different types of polymers in terms of thermal and rheological properties that virtually

dictate the mode of composite processing. Thermoplastic polymers are highly viscous while thermosetting polymers are at a low viscosity when used in their monomeric form. When it comes to amorphous polymers there is a gradual drop in its mechanical performance above T_g , while the drop in properties are more sharp in the case of semi-crystalline thermoplastic polymers and does not occur till the temperature is near the melting temperature T_m .

1.3 Processing Vs Performance

If the complete polymer composite materials system is taken into consideration then performance and processing seem to be opposing trends as shown in Figure 1.3. The performance of composites improve drastically when one moves from the injection molded composites to the advanced continuous fiber composites. On the contrary, in general, ease in processing drops when one moves from discontinuous fiber composites to continuous fiber systems. Uniaxial orientation of continuous fiber reinforced polymer matrix composites offers the highest attainable stiffness and strength properties. Full advantage of these properties is taken by designing composite structures where the predominant stresses are coincident with the fiber direction. This leads to a high degree of anisotropy which must be overcome for practical structural applications by arranging the fibers in plies stacked at predetermined angles to each other, leading to expensive labor intensive lay-up methods⁹. To overcome this limitation textile preforms are being used but fiber crossover coupled with limitations on minimum radii of curvature in the finished part, limit the geometry of finished structures to simple shapes. Attempts to overcome this limitation by adopting textile

Figur

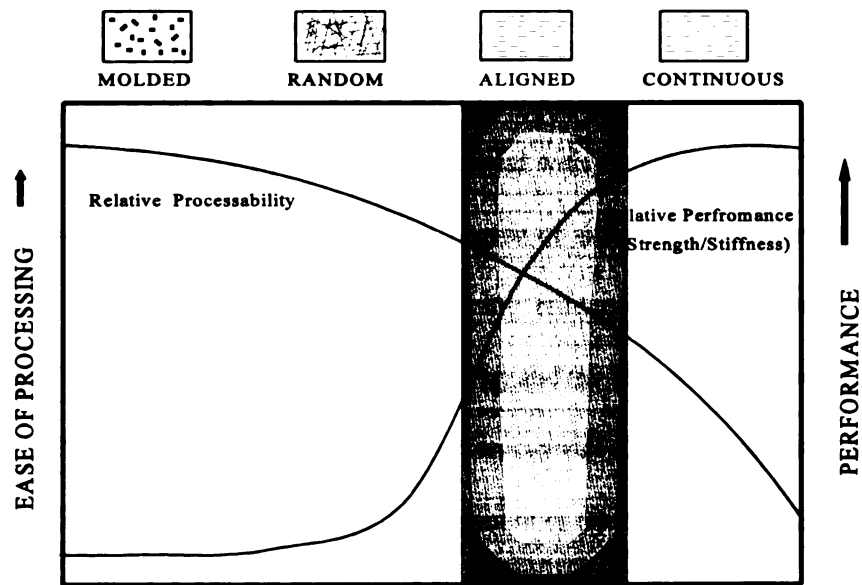


Figure 1.3 Performance versus processability trends of polymer composite material systems.

meth

dry f

form

will

unid

of da

mater

matr

shape

prop

distrib

that c

deper

unifor

canno

of the

comp

of cor

comp

methods to the handling of fibers, viz. braiding and weaving, to form complex shapes with dry fibers followed by liquid resin impregnation results in higher cost than other prepreg forms. In terms of performance, laminate theory suggests that a part made from woven fabric will exhibit reduced tensile and compressive strengths compared to a part made of unidirectional materials due to crimping of fibers to some degree in the weave. In the case of damage tolerance and fracture toughness, woven materials outperform unidirectional materials.

Fabrication of composites with a random array of short fibers dispersed in a polymer matrix results in a discontinuous composite material with isotropic properties, that can be shaped into complex geometries with a high degree of automation. However, structural properties of these composites can be drastically reduced because of the orientation distribution of fibers; discontinuity in reinforcement; and the lower volume fraction of fibers that can be packed into a random array. High performance of discontinuous fiber composites depends on the aspect ratio of the fiber (l/d), alignment of fibers, fiber-matrix adhesion, and uniform fiber packing. In the case of aligned discontinuous fiber composites the end effects cannot be neglected and the l/d ratio is taken into consideration in predicting the strengths of the composite but some of the disadvantages of woven fabric are eliminated. An optimum composite system which would provide superior mechanical performance approaching that of continuous fiber composites, and that which would provide ease in the fabrication of complex geometrical parts as in the case of short fiber composites, is an aligned

discon

1.3)

establis

compo

•

•

•

•

•

•

•

•

discontinuous fiber composite due to its high packing of axially oriented fibers (Figure 1.3)¹⁰.

There are two generalizations that should be borne in mind when it comes to establishing a qualitative relationship between the performance and processing of polymer composites.

- The reinforcing fibers are the dominant phase when it comes to mechanical performance. The stiffness and strength values of fibers vary a lot but are generally an order of magnitude higher than that of the typical polymers (Table 1.1 and 1.2). Hence, the microstructure of the reinforcing phase: the fiber orientation distribution, fiber aspect ratio and the fiber-matrix adhesion tends to be the controlling factor in the overall performance of the composite.
- On the contrary, the stiffness and strength values of the various classes of polymers are quite close and there is little variation between the two classes of polymers: thermoplastics and thermosets. However, they differ widely in the thermal and rheological behavior (Table 1.2). So, more often than not the selection of the composite processing technique is dictated by the polymer matrix. For example, low viscosity monomer impregnation is the choice of prepregging when the matrix is a thermoset while the choice narrows down to powder prepregging or hot melt impregnation when the matrix is thermoplastic. Another example is Resin Transfer Molding which is typically used for thermosetting resins while fiber reinforced injection molding invariably pertains to high melt viscosity thermoplastic resins.

Hence

poly

rheo

chara

ultim

1.4

a sub

the al

proce

a high

stems

manu

appli

increa

can b

distrib

Therefore, processing of a composite material is generally dictated by the behavior of the polymer.

Hence, the problem of optimizing performance and processing of discontinuous fiber polymer composites is a function of both the reinforcement microstructure as well as the rheological and thermal behavior of the polymer. The overall ease in processing is characterized by the speed of manufacturing, productivity, and scope for automation and ultimately the cost.

1.4 Research Motivation

Controlling the orientation state of discontinuous fiber polymer composites has been a subject of intense research in recent years. Although the significance of fiber alignment and the ability to control the orientation state of fibers has been a goal for some time a versatile processing methodology that can be operated at high speeds of operation and amenable to a high degree of automation, at the same time does not exist. The motivation of this research stems from the tremendous need for such a processing technique that can be used in the manufacturing of aligned discontinuous fiber composites that can be used for structural applications in the automotive, durable goods and infra-structure sectors. It will not only increase the envelope of utilization of discontinuous fiber composites but also provide what can be termed as microstructure controlled composites where the fiber orientation distribution is controlled based on the ultimate part performance.

It is hypothesized that if short discontinuous fibers of lengths exceeding critical fiber lengths can be aligned in electric fields in air and if these fibers can be suitably coated with fine polymer powder, then a high speed processing methodology can be established to manufacture aligned discontinuous fiber composites with stiffness and strength properties approaching those of continuous fiber composites. Both the fiber alignment in electric fields in air and the consolidation of powder coated fibers are rapid steps that will eventually lead to a high speed processing methodology. Further, such a processing technique will establish a method to manufacture microstructure-controlled discontinuous fiber composites. The objectives of this research are:

- Understand the alignment behavior of fibers in air using electric fields
- Develop a process that combines fiber alignment using electric fields with powder coating of fibers to establish a high speed process that can manufacture aligned discontinuous fiber composites
- Verify the microstructure-property relationships of discontinuous fiber composites, and establish the concept of processing microstructure controlled discontinuous fiber composites.

1.5 Novel Processing Approach

Recent advances in the area of powder coating of fibers at MSU^{11,12,13} in combination with the phenomenon of aligning fibers in electric fields, has paved the way to conceptualize and develop a novel high speed processing methodology that can manufacture aligned discontinuous fiber composites. Full realization of the stiffness to weight benefits of these

composites is possible due to effective fiber alignment combined with the ability to pack and process at higher volume fraction of fibers. Absence of solvents or liquids in the Aligned Discontinuous Fiber (ADF) process would improve the speed of processing many folds and make the process environmentally benign. The fiber alignment technique of the ADF process is simple in concept, with the scope for retrofitting this technique in an existing composite sheet or lamination processing unit. Unlike continuous fiber random mat reinforced composites which have poor drapeability and which have problems of delamination under compression, aligned discontinuous fiber composites can be flexible and can be molded or stamped into complex parts. It is envisaged that ADF composites, with unique performance and processability capabilities, will expand the range of the applicability of discontinuous fiber thermoplastic composite materials.

Numerous studies have been conducted over the past couple of decades to relate the microstructure-property relationships of discontinuous fiber composites. Unfortunately, in a majority of the studies there is a variation in the fiber aspect ratio or the fiber orientation distribution could not be determined accurately, resulting in inconclusive verification of the validity of the micromechanics models with experimental results. An important contribution from making composites using the ADF process is to verify the micromechanics models that are available for discontinuous fiber composites with a well characterized composite with a known fiber orientation distribution. This will further the understanding of the interaction between fiber orientation distribution, fiber aspect ratio and the fiber-matrix interaction including adhesion. Finally, the ability to control the microstructure goes beyond improving

pe

le

pr

1.0

mo

rev

tec

fie

in

inc

wi

usi

Fin

Ch

performance of the composite material in a particular direction. In fact it could potentially lead to the processing of materials where the thermal, electrical, optical or magnetic properties can be spatially controlled.

1.6 Dissertation Outline

The dissertation is divided into six Chapters. This chapter set the background and motivation for research and identified the problem. Chapters 2 follows with a detailed review of the literature on powder processing of composites and various fiber alignment techniques. In Chapter 3, the investigation into the behavior of fiber alignment in electric fields is presented including the theory of fiber orientation and the experimental setup used in the investigations. Chapter 4 outlines the ADF process development in its entirety including process design, process consolidation and manufacturing schemes. Chapter 5 deals with microstructure-performance relationships of aligned discontinuous fiber composites using both model predictions from fundamental micromechanics and experimental results. Finally, conclusions from this work and recommendations for future work are given in Chapter 6.

1.

2.

3.

4.

5.

6.

7.

8.

9.

10.

11.

12.

13.

References

1. Chou, T. W., *Microstructural Design of Fiber Composites*, Cambridge University Press, pp 169-230, 1992.
2. Milewski, J. V. and Katz, H. S., *Handbook of Reinforcements for Plastics*, Van Nostrand Reinhold Co., New York, 1987.
3. Mallick, P. K. "*Fiber Reinforced Composites - Materials, Manufacturing and Design*", Marcel Dekker, Inc., New York, pp 18-19, 54-65, 1990
4. Kelly, A. and Tyson, W. R., "Tensile Properties of Fiber-Reinforced Metals: Copper/Tungsten and Copper/Molybdenum", *J. Mech. Phys. Solids*, Vol. 13, p329-350, 1965.
5. Agarwal, B. D. and Broutman, L. J., "*Analysis and Performance of Fiber Composites*", John Wiley & Sons, 1990.
6. Piggott, M., "*Load Bearing Fibre Composites*", Pergamon Press, 1980.
7. *Modern Plastics Encyclopedia*, McGraw-Hill, New York, 1996.
8. Bigg, D. M., "Processing of Thermoplastic Composites" in *Polymer Rheology and Processing*, Eds. Collyer, A. A. and Utracki, L. A., Elsevier Applied Science, London, pp 381-406, 1990.
9. Foley, M. F., "Techno-Economics of Automated Composite Manufacturing Techniques", *SAMPE Quarterly*, January 1991.
10. Chang, I. Y. and Pratte, J. F., *J. Thermoplastic Composite Materials*, Vol 4, p.227-252, 1991.
11. Drzal, L. T., et. al., "Powder Impregnation of Advanced Composite Materials", in *Polymer Powder Technology*, Eds. Narkis, M. and Rosenzweig, N., John Wiley & Sons Ltd., pp 511-530, 1995.
12. Iyer, S. R., "*Continuous Processing of Unidirectional Prepreg*", Ph.D. Dissertation, Michigan State University, 1990.
13. Vyakarnam, M. N., "*Development of a High Speed Powder Process to Manufacture Composite Prepreg*", M.S. Thesis, Michigan State University, 1992.

Review of Powder Processing and Fiber Alignment Techniques

PART A: Polymer Powder Processing of Composites	21
2.1 MSU Powder Process	22
2.2 Charging of Polymer Powders	26
PART B: Fiber Alignment Techniques	28
2.3 Flow Induced Alignment	28
2.3.1 Slurry / Hydraulic Based	28
2.3.2 Spray-up or Pneumatic	31
2.3.3 Molding and Extrusion	31
2.4 External Field Induced Alignment	33
2.4.1 Using Electric Fields	33
2.4.2 Using Magnetic Fields	37
2.5 Summary and Discussion	37
References	40

The concept of combining polymer powder processing and fiber alignment using electric fields in air offers a rapid, solvent free, processing technique to manufacture polymer composites reinforced with discontinuous fibers with a preferred fiber orientation. The literature review will be presented in two parts: Part A will focus on polymer powder processing; and Part B will focus on fiber alignment techniques. The conclusions from this review will be summarized at the end of the chapter.

PART A: Polymer Powder Processing of Composites

Polymer composite materials are emerging as a potentially revolutionary new material form capable of vastly increasing the applications for polymers. These materials have an aerospace history which emphasized high structural performance with cost being a secondary consideration. Durable goods (transportation, appliance, construction, etc.), infrastructure (bridges, decking, etc.) and off-shore oil platform applications, promise to increase the use of polymer composite materials by several times if lower cost and environmentally safe processing methods can be found to manufacture these materials.

The main objective of any process for the manufacture of polymer composite materials is to completely surround each individual reinforcing fiber (typically having a diameter of 5 - 20 μ) with a polymer matrix in the final fully densified condition. Because of the reactivity or high viscosity of polymer matrices, low temperature methods using organic solvents have usually been used to manufacture these material forms. This keeps the

manufact

seriously

composit

free proce

No

manufactu

chapter on

processing

review her

charging c

impregnati

2.1 MS

Var

tow to pro

developed

deposited

A novel m

speaker or

principle ti

manufacturing costs high. In addition, even low levels of voids (1%) or retained solvent seriously reduce the mechanical properties of the resulting composite material. New composite processing employing dry polymer powders offers a low cost, high speed, solvent free processing technique.

Novel processing methods have been developed at MSU in the last several years to manufacture continuous fiber powder prepreg employing polymer powders. A recent book chapter on powder processing of composites by Drzal et. al.¹ describes the various powder processing methods and the principles of powder impregnation and consolidation. The review here will be focussed to a brief description of the MSU Powder process and the charging characteristics of polymer powders which is key to the powder coating and impregnation of fibers.

2.1 MSU Powder Process

Various modes of dispersing the polymer powder and impregnating on a spread fiber tow to produce what is a *prepreg* or *towpreg* in both wet and dry conditions have been developed. The powder impregnation processes differ mainly in the way the particles are deposited on the fibers and the particle-fiber forces responsible for holding them in place. A novel method for spreading a tow of fibers using low frequency acoustic energy from a speaker or other gas vibrating means was developed where the spreader operates on the principle that the pulsating velocity of the gaseous medium drives the spreading of the

collimated fiber tow into individual filaments. The spread width can be controlled by the amplitude of the acoustic energy and the tension in the fiber tow.

Creating a uniform and controllable dispersion of fine cohesive polymer powder using acoustic energy was studied by Iyer and Drzal². It was found that cohesive powders in the size ranges 5 to 30 μm could be aerosolized in a chamber where the powder bed is subjected to low frequency vibration from an acoustic source. A prototype MSU Powder prepreg process was developed by Iyer et al.³ where the size of the impregnated particles and the fibers were of the same orders of magnitude. In this process, spread fiber tow¹² is passed through the aerosol cloud that hovers at the top of the aerosolizer (Figure 2.1). Tribocharged polymer particles electrostatically adhere to the fibers which are later sintered in place (refer § 2.2). The rate of deposition is directly controlled by the amplitude of the acoustic energy. Subsequently, a scaled up version of this process which operates at high speeds was developed by Vyakarnam and Drzal⁴, in which the aerosol of polymer powder, generated acoustically, is entrained into a separate impregnation chamber (Figure 2.2). In this chamber, spread fibers come in contact with the settling aerosol particles in a counter current fashion. The size of particles is again critical as it affects both the charge-to-mass ratio acquired and the ratio of inertial to viscous forces on the particles in suspension. The rate of deposition can be controlled by the aerosol concentration entrained into the impregnation chamber, which can be indirectly controlled by the amplitude of the acoustic energy and the aerosol entrainment rate⁵.

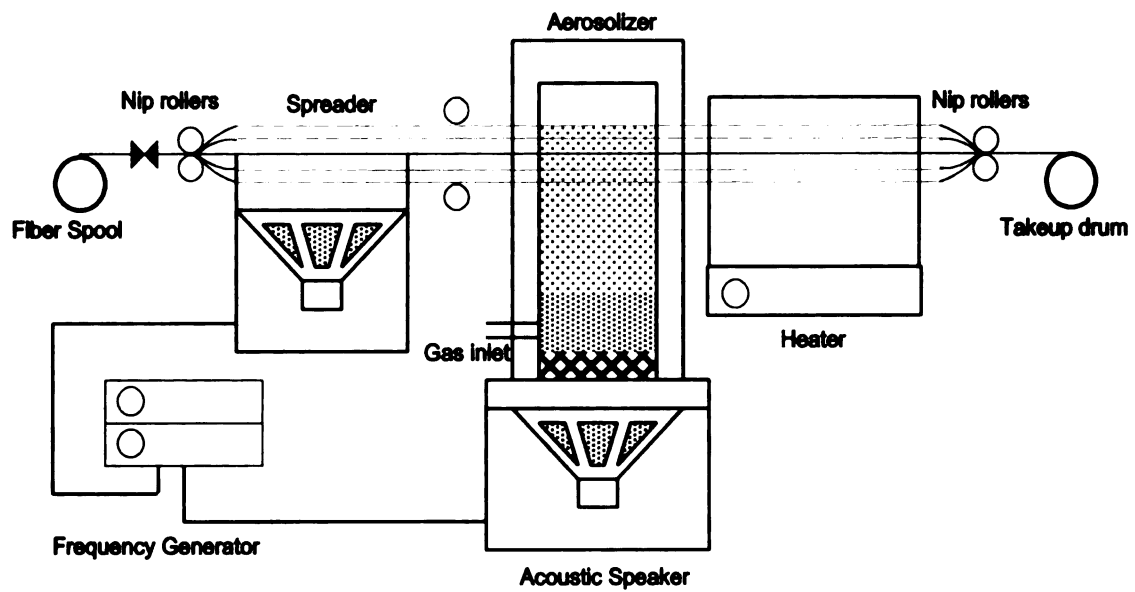


Figure 2.1 Prototype MSU Powder Prepreg Process.

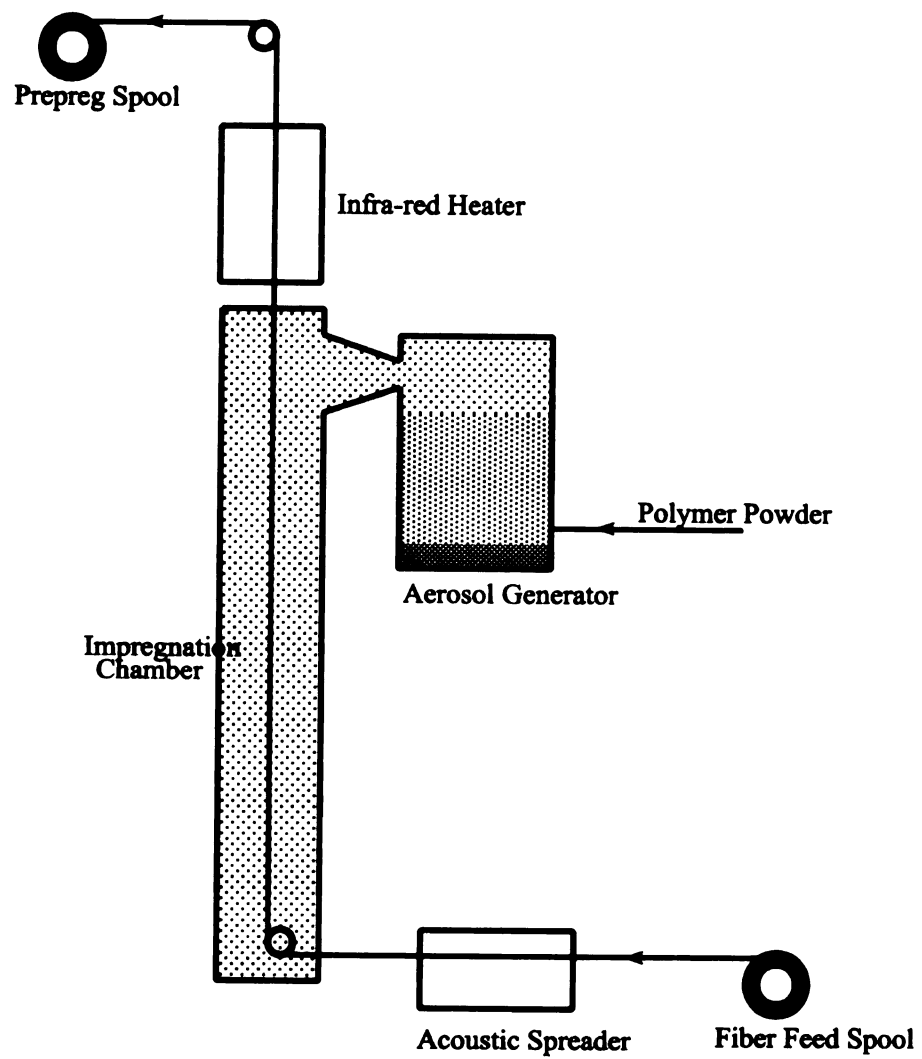


Figure 2.2 MSU High Speed Powder Prepreg Process.

2.2 Charging of Polymer Powders

In most powder processes, polymer particles are either charged by an electric field or acquire charge due to *tribocharging*. Tribocharging or triboelectrification is a phenomenon of surface charging of powder particles that occurs due to the frequent contact of the powder particles with other surfaces. In the case of electrostatic fluidized beds, field charging of particles takes place due to an imposed electric field.

When particles of different work functions are brought into sliding and/or frictional contact, tribocharging is invariably involved. Work function, is the energy required to knock off an electron from an atom and is a strong function of the materials's chemical composition, polar function groups if present and impurities. Materials can be arranged in a Triboelectric Series (Table 2.1) depending upon the sign and magnitude of charge acquired on the surface due to contact and or frictional electrification. Galo and Lama⁶ and others have separately arranged many polymers in a triboelectric series and found a strong correlation between the macroscopic property of relative permittivity and the sign and magnitude of charge of a material. The sign and magnitude of the charge becomes decisive as the distance between the two materials increases in the triboelectric series. Once a particle acquires surface charge, the rate at which the redistribution proceeds depends on the electrical relaxation time of the particle. Relaxation time is a product of particle resistivity and permittivity⁷. Most polymers being insulating materials with resistivity greater than 10^{13} ohm-cm have relaxation times in minutes or even hours which makes them more likely to

Table 2.1 Triboelectric Series of Polymers

Material	Permittivity	Sign of Charge
Polyamide	3 - 6	+ve
PPS	3.9	+ve
Epoxy resin	4	+ve
PEEK	3.2	+ve
Epoxy Polyester	3 - 4	+ve
Polycarbonate	2.9	-ve
Polystyrene	2.6	-ve
Polyethylene	2.2 - 2.7	-ve
PTFE	2.1	-ve

retain surface charges for long periods of time. It has also been reported that work function decreases with an increase in particle size which often results in large particles getting positively charged and small particles getting negatively charged⁷. This phenomenon often results in bi-polar charging of polymer powders that have a broad particle size distribution.

The size of the particle has a great effect on the magnitude of charge. The surface charge on a spherical particle increases with the square of the diameter while the mass increases with the cube of the diameter. Hence, the charge to mass ratio increases with decrease in particle size. In polymer powders, the effect of tribocharging has been found to be more pronounced in the case of size ranges under 50 microns.

PART B: Fiber Alignment Techniques

In composite materials reinforced with continuous or discontinuous fibers the spatial variation in properties can be potentially achieved by varying the fiber orientation in a controlled manner. Fiber orientation can be precisely controlled in the case of continuous fiber composites while controlling the orientation of discontinuous or short fibers is a challenge. The exponential growth in the use of discontinuous fiber reinforced plastics in the recent years has sparked intense activity in understanding and developing processes that can control fiber orientation and thereby make aligned discontinuous fiber composites. Growing demand has also been experienced for oriented fiber preforms that can be used in Resin Transfer Molding (RTM) or Structural Reaction Injection Molding (S-RIM) processes to make large structural parts in automotive and aerospace applications. Further, it is also envisioned that controlling fiber orientation in an attempt to manufacture microstructure controlled composite materials holds tremendous amount of potential for the advanced materials of the future. It is expected that material selection and design, part design and manufacturing will be highly automated and integrated into one task. A review based on published literature and patents on fiber alignment techniques that pertain to manufacturing of aligned/oriented discontinuous fiber composites is presented in this part.

2.3 Flow Induced Alignment**2.3.1 Slurry / Hydraulic Based**

ERDE Glycerine Process⁸: In this process developed by Explosives Research and Development Establishment (UK), a dilute dispersion of fibers in glycerine is passed via a

baffled reservoir through a tapered slit at a constant throughput onto a filter bed which is under vacuum (Figure 2.3). Fiber alignment is caused due to the velocity gradients that are setup between the accelerating streamlined flow and the stationary boundary of the tapered slit. The viscous drag on the fibers created by the velocity gradients orients the fibers. The fibers align in the direction of the motion of the liquid and are collected on the filter bed in a reciprocating fashion. The carrier liquid is rapidly removed without a loss in alignment using a filter surface with low resistance and a high capacity displacement pump. The major factors that affect the fiber orientation in this process are the viscosity of glycerine (which is a function of temperature); horizontal velocity of the of the orientation hopper; air pressure; distance between the slit to the filter bed; and the slit width. Very good fiber alignments were achieved using this process and the process was used by Kacir et al⁹. in the preparation of oriented short glass fiber mats in order to study the mechanical performance of aligned short fiber composites.

Budd Slurry Process: Another slurry process, popularly known as the Budd Slurry process (Figure 2.4) for preform manufacture has been developed in the recent years as a means to manufacture net shape preform for automotive applications. A composite is made from this preform using the conventional Resin Transfer Molding (RTM) processes. A well agitated liquid suspension of fibers is used to create a fiber mat by either draining the liquid or raising a filter bed through the suspension. Some degree of fiber orientation can be achieved by suitable placement of baffles in the reservoir, but the primary focus of this process is not for inducing fiber alignment but to achieve a random orientation distribution. Higher packing

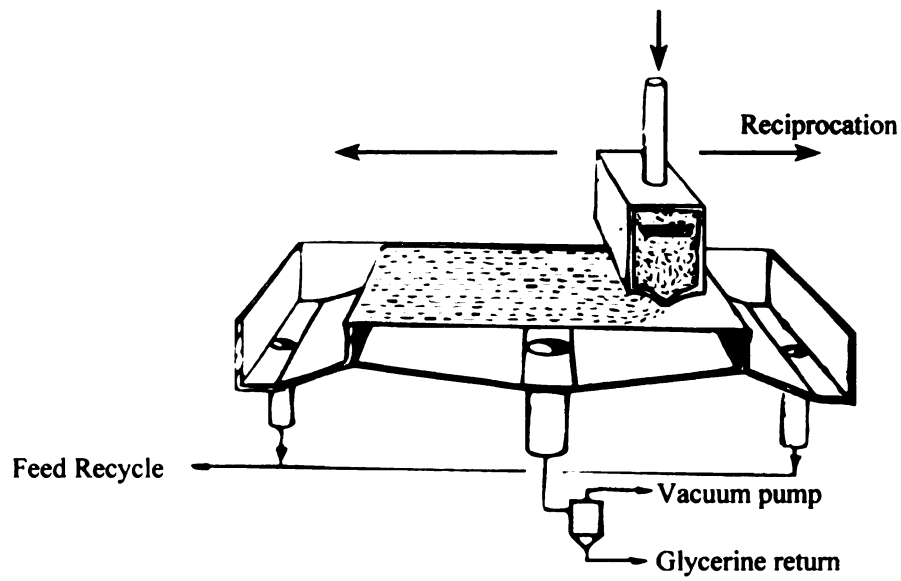


Figure 2.3 ERDE Glycerine process for fiber alignment ⁸.

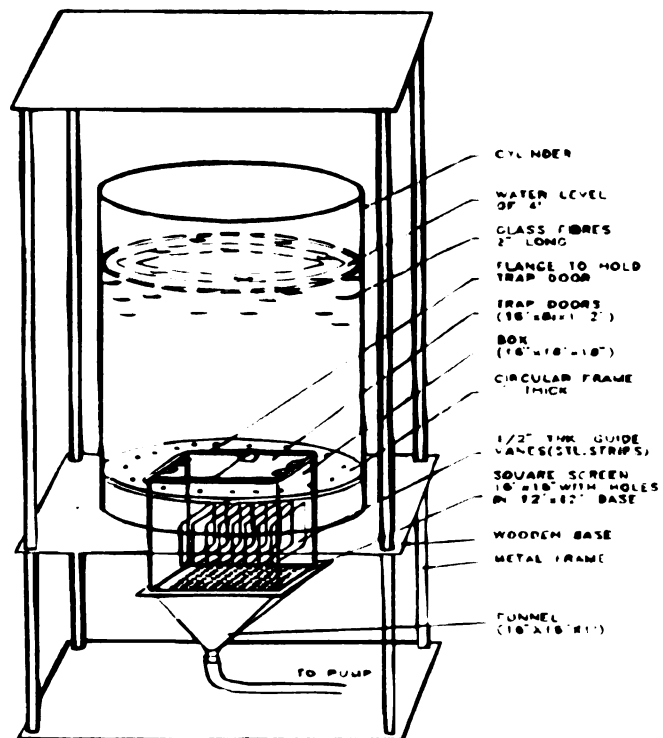


Figure 2.4 Budd slurry process for preform manufacture ¹⁰.

fraction of fibers is said to be achievable over typical spray-up techniques. Developmental efforts are underway to obtain a more uniform variation in the areal weight of the preform that is manufactured and also establish the cost benefit analysis over the competing spray-up techniques¹⁰.

2.3.2 Spray-up or Pneumatic

For industrial RTM and S-RIM processes, it is essential that the preforming step be very rapid to be cost effective. Owens-Corning¹¹ has developed a directed preforming process with robotic control for industrial RTM and S-RIM preform applications. This processing methodology is very fast and fiber orientation can be controlled by using some mechanical means like deflector plates. Fibers exiting from a hose impinge on a set of deflector plates which are at an angle to each other, and some control is achieved in the degree of orientation as the fibers build the preform. Typically stiff, thick and long (1 - 3 inches) fiber bundles can be effectively oriented by this technique which results in poor matrix impregnation and inflexible fiber preforms. Efforts have also been directed at improving the transport of the fibers, at high throughputs without clogging, using a swirl flow approach¹².

2.3.3 Molding and Extrusion

In the techniques described so far, fibers are oriented in a step separate from the flow of the resin step. In the molding and extrusion processes the fibers are in a suspension with volume fractions typically around 10 to 30 %, and aspect ratios around 10 to 30, in the

polymer melt that will ultimately form the matrix. In these processes, fiber orientation is flow induced hence the intent is to predict the orientation state of the fibers rather than controlling it, as the polymer melt flows into a mold cavity. Fiber orientation prediction and control during typical polymer melt processes like injection molding or extrusion is based on the same principles except for the difference in the flow regimes from a fluid mechanics point of view. In these processes, the viscosities of the polymer melts are so high that conditions of creeping flow are well satisfied and hence the problems can be tracked by analytical solutions to some extent. Advances made in the rheology of fiber suspensions is enabling prediction of fiber orientation distribution in molding operations. However, the effects of fiber concentration is still a complex problem and is an area of active research¹³.

Injection Molding: In injection molding, typically, the flow of molten polymer filled with short fibers into a thin cold walled mold can be treated as Hele-Shaw flow and the high shear rates established near the wall aligns the fibers along the direction of the flow while the extensional flow in the core aligns the fibers transverse to the flow direction. This phenomenon transforms into a skin and core micro-structure when the melt freezes with the fibers aligned in the flow direction in the skin and fiber transverse to the flow in the core. The most common flow pattern is from a converging or parallel runner channel through a narrow gate, which then diverges into a plate and gives a two dimensional flow path. Convergent and parallel flow induces orientation parallel to the streamlines, but divergent flows tend to introduce fiber orientation normal to the flow direction. The flow pattern is also influenced by friction and chilling of the charge at the mold surfaces, which affects the

fiber orientation distribution along the thickness of a plate type molding, and can result in a three or five zone fiber orientation distribution.

Within the limitations of molding, a certain degree of control over the orientation distribution of the fibers can be achieved by optimizing the effects of the following parameters: (a) design of the cavity; (b) position and form of the runners and gates; (c) injection rate; (d) temperature of the charge; and (e) mold temperature.

Extrusion: In the case of extrusion, elongational flow in the core region of the extruder die is a powerful mechanism for orienting fibers in any direction by changing the geometry of the die. Goettler et al.¹⁴ of Monsanto were probably the first to do extensive studies in developing practical die geometries to control fiber orientation in extrusion.

2.4 External Field Induced Alignment

2.4.1 Using Electric Fields

Studying the physics of fiber alignment in electric fields is referred to as “one of the longest known effects of electric field” by S. Whitehead in a letter in response to an article entitled “The Orientation of Fibers in an Electric Field” by J. O. Isard that appeared in 1954 in the British Journal of Applied Physics¹⁵. So, it can be said that the physics of fiber alignment is well established. Demetriades of Stanford University in 1958 established the theory for ellipsoidal alignment in electric fields for neutrally buoyant fibers with essentially zero Reynolds numbers. The early studies on fiber alignment were used as a way of

measuring the dielectric constants of fibers. Work in controlling the orientation of fibers for some end use has also been the subject of continuous investigation over the last seventy years, initially in the textiles industry and then more recently in the composites industry. In 1971, Monsanto obtained a patent for a process that aligned particles in a resin to produce a continuous filament, probably the first patent with composite material as the motivation. The wood fiber industry saw the introduction of commercial equipment from the Berol Corporation^{11,13} in 1972 for orienting wood fibers in an electric field for the continuous production of flake and fiber board. Recently, Knoblach¹⁶ has published a review of the problems and potential associated with electric field alignment of discontinuous fibers. The principle limitations in developing a high-speed, low-cost method using an electric field for orienting discontinuous fibers rests in (a) the time it takes to orient the fibers in the field and (b) the maximum rate at which the fibers can be deposited. Three processes developed in the composite materials area which use electric or magnetic fields are discussed below.

Alignment of Wood Fiber:²⁵ This process aligns wood fibers to produce boards for pencil stock. Wood fibers were dispersed in air and drawn by a vacuum down a chute in which parallel electric field is established. The aligned fibers are collected on a continuous mesh belt and later consolidated. The same group, working with Morrison-Knudson¹⁵ later extended the technology to manufacture oriented chopped glass fiber mats where a complicated array of electrodes are embedded at the bottom of the mat that is being formed to force orientation of the fibers as they descend (Figure 2.5). One of the primary reasons why they could not

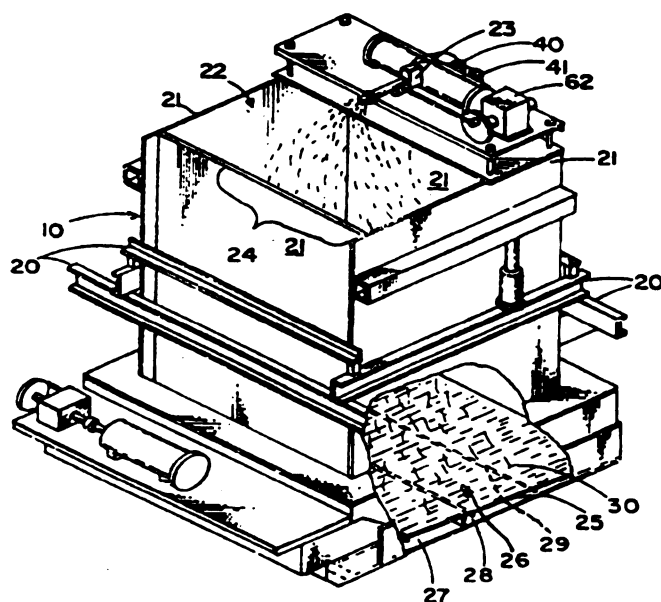


Figure 2.5 Process to make oriented chopped fiber mats using Electric fields²⁹.

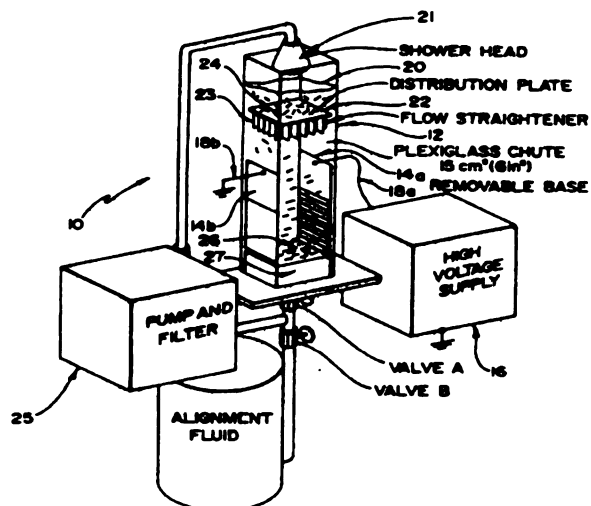


Figure 2.6 Process for alignment of fibers using electric fields in dielectric fluid¹⁷.

achieve the high levels of alignment without the elaborate set of electrodes is because they did not take advantage of the orientation state of the free settling fibers which were coming under the influence of the electric field. A binder resin is optionally sprayed on the oriented chopped fiber mat and heated to retain the integrity of the mat. But this mat is just a precursor to another process of resin transfer molding before becoming a final composite part, which reduces the overall speed of processing of a composite part.

Prepreg Bundle Alignment in a Dielectric Fluid: A recent effort in this area has been the development of a process by Knoblach¹⁷ at MIT. This process produced uniaxially aligned mats of discontinuous graphite fibers (Figure 2.6). Prepreg bundles are dispersed in a dielectric fluid settle through an electric field. The bundles are aligned with the field before settling on the bottom of the low viscosity fluid tank, to form a mat of prepreg bundles. After draining the fluid, the mat is consolidated to form aligned composite plate. The use of dielectric fluid creates additional steps like draining the liquid from the tank ensuring there is no loss of fiber orientation and the subsequent drying of the wet fiber mat.

Whisker orientation: Giacomel¹⁸ developed the concept of whisker alignment, in a transverse direction between fibers or lamina during the curing cycle of the matrix, using electromagnetic fields. The whiskers retain their orientation in the final composite form which improves the structural integrity of the laminate. This approach was more for batch processing than continuous processing.

2.4.2 Using Magnetic Fields

In order for magnetic fields to align fibers the fibers need to be ferro-magnetic. This puts a serious limitation on the types of fibers that can be aligned for typical composite manufacture, since majority of the fibers used in composite materials like glass, carbon and aramid fibers are not ferromagnetic. These fibers can be aligned if they are specially coated with ferro-magnetic materials like nickel etc. Due to this very reason, a continuously operated process that makes oriented discontinuous fiber composites does not exist. Experimental and analytical analysis were conducted by some groups like Shine and Armstrong¹⁹ of MIT and Hatta et al²⁰ of Japan to investigate the alignment behavior of fibers coated with magnetic materials. No attempts were made to develop any continuously operated process to make aligned discontinuous fiber composites using magnetic fields.

2.5 Summary and Discussion

The fiber alignment techniques that have been developed so far can be broadly classified as either wet/slurry methods or dry methods.

In the wet methods^{21,22,23} the fibers are usually in a well agitated liquid suspension and a fiber mat is created by either draining the liquid or raising a filter bed through the suspension. Control of fiber orientation is limited, but can be achieved to some degree by guiding vanes or other means like electric fields when the liquid is dielectric²⁴. The drawbacks in these processes is the introduction of an additional step of drying the wet mat which reduces the speed of manufacturing drastically, and secondly the fact that fiber mat

preform has to be further processed by reaction injection molding or sheet impregnation to result in a composite part. The mechanical performance of the final part is sometimes lowered due to the presence of voids entrapped during the drying of the wet fiber mat.

Dry methods^{25,26,27,28,29} usually rely on electric fields or pneumatic means³⁰ to control fiber orientation and are generally faster than the wet processes. The main drawback of the existing processes is the limited understanding of the dynamics of fiber motion which has resulted in elaborate schemes to create electric fields by the earlier inventors to orient fibers, thereby reducing process flexibility in changing fiber orientation during a run. Another major hurdle in the speed of fabrication is the fact that the fiber mat preform, formed by these processes, needs to be further processed by reaction injection molding or sheet impregnation before a composite part can be made.

All the processes described so far cannot be evaluated on a common platform because of the differences in terms of the fiber lengths that were considered and the flow regimes of individual situations depending upon the fluid medium. Therefore, caution should be exercised in comparing the degree of fiber orientation from one technique to the other. Often, the fiber alignment technique is treated independently and the benefits of processing ease is lost when the processing technique is evaluated in totality for manufacturing aligned discontinuous fiber composite. Some of the limitations that exist in terms of the desirables for composites is:

- ability to align long fibers at high volume fractions

- high speeds of fiber alignment
- high speeds of the final composite manufacture

Recent development of a high-speed MSU Powder Prepreg Process to make composite materials by Vyakarnam and Drzal³¹, using an entrained aerosol of fine tribo-charged polymer particles to coat the fibers, along with the known phenomenon of fiber orientation in electric fields has created a basis upon which an Aligned Discontinuous Fiber (ADF) composites process can be developed to potentially overcome all the above limitations. The details of the ADF process development will be taken up in Chapter 4.

References

1. Drzal, L. T., Padaki, S., Vyakarnam, M. N., Fernandes, J. H., "Powder Impregnation of Advanced Composite Materials", in *Polymer Powder Technology*, Eds. Narkis, M. and Rosenzweig, N., John Wiley & Sons, 1995.
2. S. R. Iyer and L. T. Drzal, *J. of Thermoplastic Composite Materials*, Vol 3, pp 325-355 (1990)
3. S. Iyer, L. T. Drzal, and K. Jayaraman, "*Method for Coating Fibers with Particles By Fluidization in a Gas*", US Patent Nos. 5,102,690; 5,123,373; 5,128,199 (1992)
4. M. N. Vyakarnam and L. T. Drzal, *Apparatus and High Speed Method for Coating Elongated Fibers*, US Patent no. 5,310,582 (1994)
5. M. N. Vyakarnam and L. T. Drzal, *Polymer Processing Society (Conf. Proc.)* pp 31-32 (1992)
6. C. F. Galo, and W. L. Lama., *J. of Electrostatics*, Vol. 2, pp 145-150 (1976)
7. A. G. Bailey, *Powder Technology*, Vol. 37, p 71-85, (1984)
8. Bagg, G. E. G., Evans, M. E. N., and Pryde, A. W. H., *Composites* Vol 1, 1969.
9. Kacir, L., Narkis, M., and Ishai, O., "Oriented Short Glass-Fiber Composites", *Polymer Engineering and Science*, Vol 15, p 525, 532, 1975.
10. Soh, S. K., "Slurry process for Preform Manufacture", Proc. 10th Annual ASM/ESD ACCE, Dearborn, 1994.
11. Jander, M., "Industrial RTM - new developments in molding and preforming technologies", Proc. 10th Annual ASM/ESD ACCE, Dearborn, 1991.
12. Serban, P. and Petty, C., "Flow control of chopped glass fiber bundles through flexible tubes", Presented at AIChE Annual Meeting, Miami, 1995.
13. Tucker, C. L. and Advani, S. G., "Processing of Short Fiber Systems" in *Flow and Rheology in Polymer Composites Manufacturing*, Ed. S. G. Advani, Elsevier Science, 1994.
14. Goettler, L. A., Leib, R. I., and Lambright, A. J., *Rubber Chem. Technol.*, 52, 838, 1979.

15. Isard, J. O., "The Orientation of Fibers in Electric Field", *British Journal of Applied Physics*, Vol 6, p 176-179, 1955.
16. Knoblach, G. M., "Processing, Electric Fields for Fiber Orientation", in *Encyclopedia of Composite Materials*, McGraw-Hill, p. 413-424, 1991.
17. Knoblach, G. M., "Using Electric Fields to Control Fiber Orientation During the Manufacturing of Composite Materials", *SAMPE J.*, vol. 25, No.6, p.9, 1989.
18. Giacomel, J. A., *US Patent No. 4,560,603*, 1983.
19. Shine, A. D. And Armstrong. R.C., "Experimental studies of suspended ferromagnetic fibers in a magnetic field", *Rheologica Acta*, Vol. 26, p 162-171, 1987.
20. Yamashita, S. et. al., "Fiber Orientation Control of Short Fiber Composites: Experiment", *J. Of Composite Materials*, Vol. 23, p 32-41, 1989.
21. Bagg, G. E. G., Evans, M. E. N., and Pryde, A. W. H., *Composites* Vol 1, 1969.
22. Kacir, L., Narkis, M., and Ishai, O., "Oriented Short Glass-Fiber Composites", *Polymer Engineering and Science*, Vol 15, p 525, 532, 1975; Vol 17, p 234, 1977; Vol 18, p 45, 1978.
23. Soh, S. K., "Slurry Process for Preform Manufacture", *Proc. 10th. Annual ASM/ESD Advanced Composites Conference & Exposition*, Dearbon, 1994.
24. Knoblach, G. M., "Using Electric Fields to Control Fiber Orientation During the Manufacturing of Composite Materials", *SAMPE J.*, vol. 25, No.6, p.9, 1989.
25. Talbot, J. W. and Logan, J. D., *U.S. Patent No. 4,113,812*, 1978.
26. Carpenter, C. T. and Stunkard, N. W., *U.S Patent No. 4,111,294*, 1978.
27. Logan, J. D., *U.S Patent No. 4,432,916*, 1984.
28. Talbot, J. W., Peters, T. E. and Logan, J. D., *U.S Patent No. 4,664,856*, 1987.
29. Peters, T. E. at al, *U. S. Patent No. 5,017,312*, 1991.
30. Ericson, M. L. and Berglund, L. A., *Composites Science and Technology*, 49 p 121-130, 1993.
31. Vyakarnam, M. N. and Drzal, L. T., "Apparatus and High Speed Method for Coating Elongated Fibers", *U.S. Patent No. 5,310,582*, 1994.

Fiber Alignment Using Electric Fields

3.1	Introduction	43
3.1.1	Electric or Magnetic Fields?	44
3.1.2	Fiber Dimensions	46
3.2	Theory of Fiber Orientation	48
3.2.1	E-Field Polarization	49
3.2.2	E-Field Alignment Time	52
3.2.3	Fiber Hydrodynamics	55
3.2.3.1	Orientation Behavior of Fibers During Free Settling	55
3.2.3.2	Fiber Motion in Shear Flows	58
3.2.3.3	Settling Velocity of Fibers	59
3.3	Experimental Studies	61
3.3.1	Experimental Setup	62
3.3.2	Behavior in D.C. Fields	65
3.3.3	Behavior in A.C. Fields	69
3.3.3.1	Effect of E-field Intensity and Fiber Aspect Ratio	70
3.3.4	Fiber Settling Behavior	82
3.4	Summary of Results	86
	References	88

3.1 Introduction

In the proposed Aligned Discontinuous Fiber (ADF) process, fibers are fed from the top of an orientation chamber which consists of an electrode configuration to align fibers as they settle under gravity. The fibers are aligned in the direction of the applied electric field. The physics of fiber orientation in electric fields has been previously investigated as pointed out in § 2.4. However, in almost all the previous studies the experiments were so designed that the dimensions of the fibers and/or the fluid medium in which they were immersed were so chosen that the fiber motion fell under viscous flow regimes, thereby amenable to analytical treatment and/or accurate experimental verification. The objective of this research is to align fibers using electric fields in a manner that would be useful in manufacturing discontinuous fiber polymer composites, whose properties approach those of continuous fiber composites. The phenomenon of fiber alignment in air, especially of fibers of dimensions that are useful in composite materials, offers tremendous technological significance by providing the scope for rapid solvent free processing in a variety of situations. Excepting some literature in the field of aerosol science for fibrous dispersions (sizes ranging from a micron to several hundred microns), there is little published literature on the orientation behavior of fibers in air for lengths ranging from a millimeter to 50 millimeters. The approach taken in characterizing and understanding the fiber alignment behavior in electric fields is by analyzing the theoretical situations that have been developed so far and making quantitative or qualitative extensions from them by careful experimental design.

E-glass fibers are chosen for developing the ADF process due to their extensive use in discontinuous fiber composites compared to carbon fibers (Table 3.1). Another important distinction between glass and carbon fibers is in the electrical conductivity which will play a role in their behavior in electric fields. Glass fibers are dielectric in nature which will make it longer to align in electric fields compared to carbon fibers which are conductive. However, once the processing principles for dielectric fibers in electric fields are established, it will be relatively easy to translate the principles to carbon fibers rather than going vice-versa. Before the theory of fiber orientation is presented, the reason for choosing electric fields is discussed. Also the limits to fiber dimensions and flow regimes that are required for the development of a meaningful high speed processing technique are outlined.

3.1.1 Electric or Magnetic Fields?

The first question that needed to be addressed in the quest for a rapid fiber alignment technique was which fields (electric or magnetic) would be most appropriate. Despite the similarities, the differences have a strong bearing on the utilization of one type of field over the other especially when it comes to polymer composites. Glass, carbon and aramid fibers which form the bulk of the reinforcements in composite applications cannot be aligned using magnetic fields unless they are coated with a ferro-magnetic material. Electric fields on the other hand can align any axisymmetric body due to polarization, as long as there is a dielectric mis-match between the body and the fluid medium.

Table 3.1 Pertinent Fiber Properties

Fiber	Sp. Gr.	Diameter (μm)	Critical Length*(μm)	Dielectric constant†
E-Glass	2.55	10 - 13	400 - 1000	5.9-6.6
AS4 Carbon	1.8	8.0	400 - 600	-
Aramid**	1.45	12.0	800 - 1200	3.6

* Approximate ranges with epoxy matrix systems¹ ; ** Kevlar-49 (DuPont); † at 1 MHz

Secondly, electric fields are inexpensive and easy to generate. All that is needed to generate an electric field is a high voltage source and an electrode made out of a conducting material. The geometry and the position of the electrodes can be easily designed to create a desired E-field configuration. In the case of magnetic fields, large coils are required and it is more difficult and cumbersome to configure a desired field geometry.

An important distinction between the two fields lies in the way they breakdown in a fluid medium. As voltage is increased beyond what is termed a “breakdown” voltage, electric fields ionize the molecules and create an avalanche of electrons, which tries to find the nearest positive electrode or a grounding point to discharge. This effect is commonly referred as the corona discharge. The theoretical breakdown in air occurs at a field intensity of 3000 KV/m over a distance of more than about 1 millimeter^{2,3}. This value of breakdown voltage is a strong function of the gas pressure; the gap width between electrodes and humidity. Under practical conditions in air, it is difficult to operate at greater than 1000

KV/m without frequent field breakdown. The breakdown voltage establishes an upper limit on the maximum torque that can be applied to align a body. In the case of a magnetic field, there is no such limitation due to a field breakdown. Torques which are several orders of magnitude higher than what can be obtained from electric fields in air can be easily obtained by magnetic fields in practical situations. Magnetic fields are the choice when the material is magnetic like nickel or steel fibers.

3.1.2 Fiber Dimensions

The length of the fibers that will be used in the investigations is determined using the critical fiber length concept that was introduced in §1.2.1. One of the objective of this research was to determine if the properties of aligned discontinuous fiber composites made by the ADF process will approach the properties of continuous fiber systems. This stipulates that the length of the fibers be at least 10 times the critical fiber length. The fiber dimensions that are of interest in discontinuous fiber polymer composites are typically larger than those encountered in electro-rheological fluids, whiskers reinforcements and fibrous aerosols. A priori it is difficult to determine precisely the critical fiber length for any given fiber-matrix system, because of the experimental difficulties involved and the variations in the interfacial adhesion⁴. However, since most polymers have mechanical properties of the same order of magnitude (see Table 1.2), a reasonable extrapolation from E-glass fiber-epoxy data can be made to estimate the fiber lengths that will be needed in this study (Table 3.1)

While the optimum length of the fibers for the ADF process should be long enough so that reinforcement properties approach the properties of continuous fiber composites, they should also be short enough so that shaping of the composite over small radii of curvature is possible. Based on this reasoning, fibers of nominal lengths ranging from 3 to 25 mm were selected for this study. These fibers are a few times greater than critical fiber length to greater than 10 times the critical fiber length (Table 3.1).

Discontinuous glass fibers are commercially available as chopped fibers. Chopped fibers are aggregates (bundles) of individual fibers (or filaments) held together by a sizing. Commercial glass fibers are always sized to protect the fibers from damage during handling and also to provide a good interface between the fiber and the matrix. The typical length of chopped fibers ranges from 0.125 to 3 inches and the number of filaments per bundle can vary from a few hundred to a few thousand. During manufacturing operations like extrusion and injection molding, chopped glass fibers are broken down to shorter lengths as well as get dispersed into individual filaments due to the shearing action encountered. The reason for widespread commercial availability of chopped fibers is because individual filaments in aggregates or bundles have integrity and are easy to handle in mechanical operations. It is almost impossible to handle short fibers if they are in filament form due to the small fiber diameters (10 - 13 microns). The fibers will tend to form fuzzy and entangled masses with little possibility to be separated and aligned into individual fibers.

Another aspect that needs to be considered is the effect of the aggregate nature of chopped fibers in the alignment behavior as well as the reinforcement behavior. The effective aspect ratio of the fiber bundle is a direct function of the aggregate nature of fibers i.e the size of the fiber bundle. From a micro-mechanics view point, only the length of an individual filament is important and not the size of the bundle as long as each filament is surrounded by matrix. From a processing viewpoint the bundle size affects the process in two significant ways. First, the number of filaments in the fiber bundle will affect both the E-field alignment times and the hydrodynamics of fiber motion since both these are functions of fiber aspect ratio. Secondly the bundle size will affect resin impregnation into the fiber bundle in the consolidation step. Ideally, the fiber bundles should have as small a number of filaments as possible for bundle integrity and mechanical handling, so that individual filaments do not easily separate and create a fuzz. Commercially available chopped fiber bundles typically have about 1000 filaments or more. Preliminary analysis indicated that fiber bundles of that size may be difficult to align in electric fields in air, therefore, bundles with filaments ranging from 100 to 400 were obtained from Owens-Corning and Vetrotex CertainTeed.

3.2 Theory of Fiber Orientation

In the fiber alignment process three things are taking place, all of which contribute in effective fiber alignment in air using electric fields.

- Fiber polarization and alignment in E-fields.
- Hydrodynamic resistance.

- Initial fiber orientation state, when the fibers are entering the orientation chamber and the final steady-state orientation behavior of free falling fibers.

Each of the above events is discussed separately first, but it will be shown later through experimental results and theoretical analysis that the fiber alignment process is in actuality a combination of all the three events. The purpose of this section is to provide a theoretical framework on the fiber alignment process based on prior studies and extensions from those results for the cases under study in the dissertation. Experiments that are conducted to verify some of these concepts and their validity for the fibers that are of interest in composites manufacture will be taken up in section §3.3.

3.2.1 E-Field Polarization

Conductive as well as dielectric fibers can be aligned in an electric field as long as there is polarization of the fiber, which can be achieved when there is a difference between the dielectric constant of the fiber and the medium surrounding it. Conductive fibers are polarized to a greater degree than non-conductive (dielectric) fibers due to the presence of free electrons which allow a rapid migration of free charges (of the order of 10^{-13} seconds) towards the end. In the case of a dielectric particle only molecular alignment can take place. The force diagram on a single fiber orienting in an electric field while settling due to the gravitational field in air is shown in Figure 3.1. Electric fields induce a dipole in the fiber which appears as surface charges at its ends. Columbic forces between the ends and the field causes an electric torque (T_E) which causes the fibers to align parallel to the field.

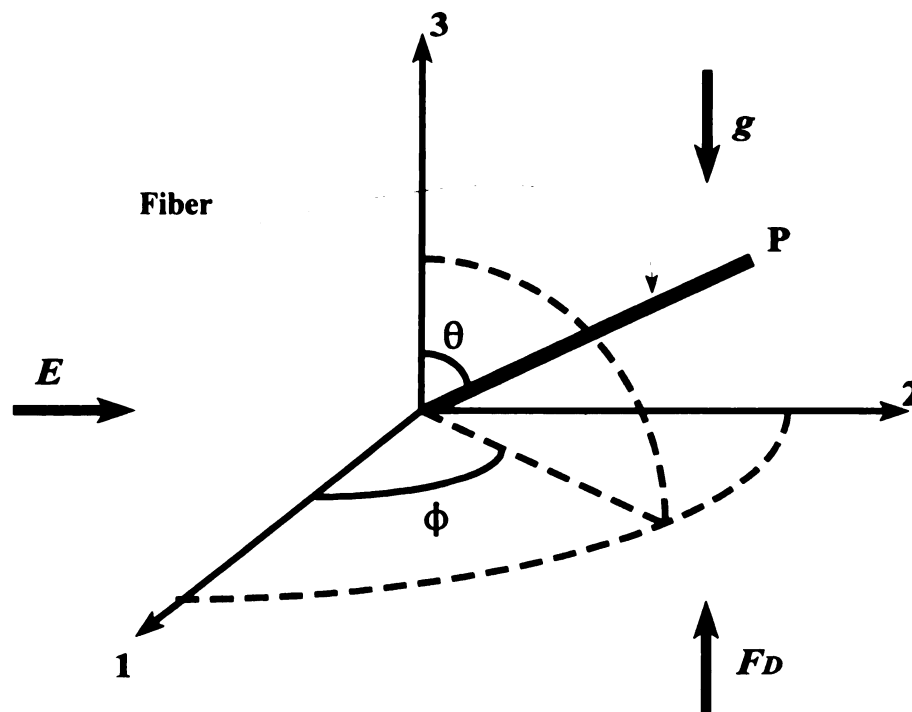


Figure 3.1 Forces acting on a fiber in the orientation chamber.

This torque is balanced by the hydrodynamic (viscous) drag on the rotating fiber (T_H). Consider a neutrally buoyant fiber in the fluid medium in the absence of inertial effects, then the two torques balance.

$$T_H + T_E = 0 \quad (1)$$

The torque due to an E-field of intensity, E_o on a dielectric prolate ellipsoid in a dielectric fluid is derived using energy arguments⁵ and the equation is a function of the volume (V) of the particle and the polarization parameter, W_E . When $\theta = 0$, T_E is given by (see Figure 3.1):

$$T_E = V W_E \sin 2\phi \quad (2)$$

$$W_E = \kappa_2 \epsilon_o E_o^2 (q - 1)^2 F \quad (3)$$

Polarization parameter, W_E depends on the dielectric constant of the fiber (κ_1) and the fluid medium (κ_2) and E_o . “F” is a factor related to the ratio of dielectric constants (q) of the fiber to the fluid medium ($q = \kappa_1 / \kappa_2$); “A” is an elliptical integral constant and “e” is a function of the aspect ratio ($r_e = a/b$) of the ellipsoid.

$$F = \frac{1.5A - 1}{2 [1 + (q - 1)(A - 1)] [1 + (q - 1)A/2]} \quad (4)$$

$$A = \frac{2e - \ln\left(\frac{1+e}{1-e}\right)}{2r_e^2 e^3} + 1 ; \quad e = \sqrt{1 - \frac{1}{r_e^2}} \quad (5)$$

The balancing between hydrodynamic forces and electric torques on an ellipsoid results in the rotation of the particle and eventual alignment with the E-field. The time taken to align a fiber based on its electrical nature and the electrical field intensity is presented the next section.

3.2.2 E-Field Alignment Time

Demetriades⁶ derived the equation for the rotation of a prolate ellipsoid, assuming inertial effects are negligible, particles are neutrally buoyant and Reynolds number is far less than 1. Mason and co-workers had done some extensive theoretical studies and experimental observations on the effect of electric fields on particle motion for a variety of flow situations and summarized the results in a review article⁷. It was experimentally found by them that the analytical model derived by Demetriades to determine the alignment time (t) for a dielectric elliptical body to orient from an initial angle ϕ_i to ϕ_f along the electric field lines of intensity E_o holds good under the given assumptions and is given by the following relationship.

$$t = \frac{\eta}{8\pi\epsilon_o\kappa_2 E_o^2 P(q, r_e)} [\ln(\tan\phi_f) - \ln(\tan\phi_i)] \quad (6)$$

The polarization function $P(q, r_e)$ is related to the electrical and geometrical nature of the ellipsoid. For dielectric ellipsoids it is:

$$P(q, r_e) = \frac{(3A - 2)(q - 1)^2 Q(r_e)}{8\pi[2 + (q - 1)A][(q - 1)A - q]} \quad (7)$$

$$q = \frac{\kappa_1}{\kappa_2} \quad r_e = \frac{a}{b}$$

For conductive ellipsoids, $q \rightarrow \infty$, $P(q, r_e)$ is given by:

$$P(q, r_e) = \frac{(3A - 2) Q(r_e)}{8\pi A(A - 1)} \quad (9)$$

The parameter $Q(r_e)$ is related to the elliptical integral function (A) and the aspect ratio of the ellipsoid (r_e).

$$Q(r_e) = \frac{2r_e^2 + (1 - 2r_e^2)A}{4(r_e^2 + 1)} \quad (10)$$

Alignment times are directly proportional to the viscosity of the fluid medium and inversely proportional to the square of the electric field strength. For example, a conductive carbon fiber has a dielectric constant of infinity while an insulating glass fiber has a value of 6.2. Dry air has a dielectric constant of 1. Both fibers would be capable of being oriented although the carbon fiber would respond much faster. This relationship (Eq. 6) shows that

the alignment time is a very strong function of E-field intensity and the polarization function P. P is in turn related to the ratio of dielectric constants of the fiber and the medium and the aspect ratio. Better fiber alignments are possible at higher E_o . However, it is to be noted that this equation can only serve as a first approximation to determine the alignment times of the fibers that can be used in the ADF process. This is because the equation is valid for fiber motions in the viscous flow regime (implying very small fibers or a very viscous medium), whereas the glass fibers that are being experimented in the setup are relatively large. Besides, the fluid medium being air results in the fiber motions to lie beyond the viscous flow regime. Mason and co workers⁷ have verified this theory for a number of different types of particles in the colloidal and non-colloidal ranges (largest size tested with this model was 900 μm) under shear flow conditions.

The theoretical principles underlying the alignment of air borne particles in an electric field have also been studied by Fuchs⁸, where the hydrodynamic resistance provided by air is neglected. Therefore, torque resulting from shear forces associated with laminar flows is neglected. Further, Fuchs considers most airborne particles to be conductive because of the moisture adsorbed on the surfaces when the particles are exposed to ambient conditions of relative humidities greater than 30 %. The equation for angular or rotational mobility of prolate ellipsoids was derived and the alignment time (after simplification for large aspect ratios) is given as:

$$t = \frac{\eta}{1.5\pi\epsilon_o E_o^2 \cos\phi \sin\phi} \quad (11)$$

In this equation, notice there is no dependence on the aspect ratio of the fiber. When a fiber is aligned from $\phi_i = 85^\circ$ to $\phi_f = 5^\circ$, the time to align any particle is 4×10^{-5} s and 1×10^{-5} s for an E field intensity of 250 and 500 KV/m respectively. If the conduction effect is neglected then the particle is treated as a dielectric and the alignment time increases several orders of magnitude to around a second. Lilienfeld⁹, verified the alignment of fibers that were up to 20 microns in length in air and found reasonable agreement with Fuchs's contention that air borne particles (with RH > 30%), behave like conductive particles.

3.2.3 Fiber Hydrodynamics

3.2.3.1 Orientation Behavior of Fibers During Free Settling

Does a fiber fall vertically or horizontally during settling in an unbounded fluid medium? In other words do fibers have any tendency to fall in a preferred orientation state? Answers to these questions will be ascertained by considering the case of a single fiber in a dilute situation, since it is quite difficult to predict the orientation state of a suspension of fibers with fiber-fiber interactions. An insight into these questions will be very useful in understanding the alignment behavior of fibers especially because of the non-buoyant nature of the fibers. Qualified arguments are made which are supported by both theoretical and experimental investigations^{10,11,12}. The orientation behavior of ellipsoidal particles is analyzed in the following three flow regimes based on Reynolds number (Re) of the particle.

$$Re = \frac{d_f V \rho}{\eta} \quad (12)$$

Where d_f is the fiber diameter (or effective bundle diameter), V is the fiber velocity, where ρ is the fluid density and η is the fluid viscosity. Figure 3.2 mimics the orientation behavior of fibers in two dimensions as they settle in the different flow regimes. This figure depicts initial orientation state (vertical, horizontal and oblique at 45°) of the fiber when it is released and the final orientation state of the fiber that is attained at steady state.

- **Viscous Regime ($Re < 1$):** When an ellipsoidal particle or fiber is falling very slowing through a resisting medium then the orientation state of the body remains unchanged and maintains the initial state of orientation in which it was released. This holds good for any initial state of orientation (see Figure 3.2). This is because the resultant torque due to viscous forces acts through the center of the ellipsoid which nullifies any coupling effect. Certainly, the ellipsoid should be perfectly symmetrical for this phenomenon to hold good. This is the regime where creeping flow conditions apply and most of the analytical modeling work pertains to this regime.
- **Intermediate Regime ($Re \sim 1$):** As Re gets greater than 0.1, the nature of the ellipsoidal motion begins to change. The particles start to orient themselves in such a way that the drag forces are balanced with the gravitational forces and they occupy an orientation state which has the maximum resistance to free fall. In other words a fiber will tend to orient itself with its long axis normal to the direction of free fall. The orienting force increases with increase in Re until a stable orientation is

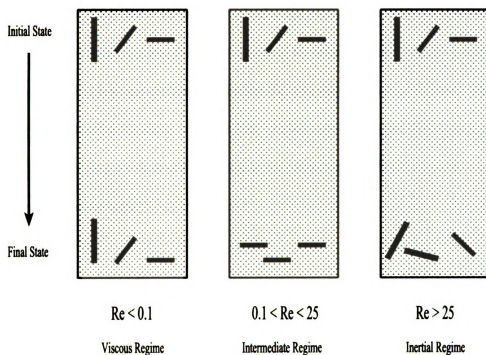


Figure 3.2 Schematic representation of the orientation state in free falling fibers.

achieved at some value of Re between 10 to 100. This kind of stability in the orientation state is also in agreement with the theoretical predictions one gets from potential flow conditions. Under these conditions, a couple acts on an ellipsoidal particle to orient normal to the flow direction.

- **Inertial Regime ($Re > 1$):** As the Re value increases above one and passes through the stable motion regime, there lies a critical Re where vortex shedding starts to take place and starts to get difficult to predict the orientation state of a fiber. This can be anywhere from a Re value of 25 to 100. At Re values beyond this region, turbulent effects also play a role and it once again becomes very difficult to predict accurately the orientation state.

3.2.3.2 Fiber Motion in Shear Flows

In a recent book chapter by Tucker and Advani¹³, a review of the current research that surround flow induced fiber orientation prediction in short fiber composite processing pertaining to molding operations has been presented. Although, the fiber alignment and motion in the ADF process is not in the viscous flow regimes, prior research in this area provides useful insights into the behavior of fibers in the process developed. Jeffery¹⁴ solved the problem of the motion of a single ellipsoidal particle in a shear field and found that ellipsoidal particles align in the direction of shear flow and transverse to flow direction in elongational flows. Further, Jeffery's equation predicts that a single particle in a simple

shear flow ($v_1 = Gx_2$; $v_2=v_3=0$) will undergo a periodic rotation, which is a strong function of aspect ratio of the particle.

$$T = \frac{2\pi}{G} \left(r_e + \frac{1}{r_e} \right) \quad (13)$$

Where T is the time period of rotation and r_e is an equivalent aspect ratio. Jeffery's equations have been experimentally verified¹⁵ and found that the predictions of this model to be accurate when all the assumptions are met. An important conclusion that one can draw from the above relationship is that as the aspect ratio (length) of the fiber increases, the period of rotation increases in a shear flow. Due to the large difference in the densities between glass fibers and air, the fiber motion is far from neutrally buoyant and shear flow situations arise, because of the translation and rotation of the fiber during free fall. If we apply the phenomenon of fiber motion in shear flow as predicted by Jeffery, then it may be concluded that the stability of the fiber motion increases as the length of the fiber increases. And stability of fiber motion improves fiber alignment for non-buoyant particles. Based on these arguments it will be shown in § 3.3.3.1 that 1" long fibers align better than 1/4" long fibers in the ADF process.

3.2.3.3 Settling Velocity of Fibers

The dimensions of the fibers under investigation (a few millimeters to 50 millimeters in length) are beyond the region where Brownian effects are encountered, hence these effects

can be safely neglected from the present analysis. Extensive analytical treatment and experimental work has been done to determine drag on non-spherical particles when they settle in the viscous regimes. For non spherical particles, orientation and the shape factor become critical in the determination of the drag coefficient. There are two limiting conditions for the state of fiber orientation during the settling of a cylinder: (a) cylinder axis parallel to the direction of fluid flow (Vertical case), and (b) cylinder axis normal to the direction of fluid flow (Horizontal case). Analytical expressions for drag force F , (with subscript V and H representing the vertical and horizontal cases respectively) have been obtained for ellipsoids by Oberbeck in 1876. Batchelor¹⁶ offers a model to predict the drag on cylinders based on slender body theory. The drag force of a cylinder settling under the extreme cases of horizontal and vertical orientations can be used to estimate the settling times of fibers of various lengths and diameters. Dynamic shape factors k are obtained using the simple relations which follow, where "a" is the diameter and "L" length of the cylinder respectively; " r_e " is the aspect ratio; $\epsilon = \ln(L/2 r_e)$; and the subscript "ve" stands for equivalent sphere terms.

$$k = \frac{u_{ve}}{u} = \frac{F}{F_{ve}}; \quad d_{ve} = ar_e^{1/3} \quad (14)$$

$$\begin{aligned} F_V &= 2\pi\eta Lu \left[\frac{\epsilon + 0.307\epsilon^2}{1 - \epsilon/2} + 0.426\epsilon^3 \right] \\ F_H &= 4\pi\eta Lu \left[\frac{\epsilon + 0.307\epsilon^2}{1 + \epsilon/2} + 0.119\epsilon^3 \right] \end{aligned} \quad (15)$$

Using the above equations with $d_{ve} = a(1.5 r_e)^{1/3}$, the shape factors for the cylinder become

$$\begin{aligned} k_V &= (2/3)^{4/3} r_e^{2/3} \left[\frac{\epsilon + 0.307\epsilon^2}{1 - \epsilon/2} + 0.426\epsilon^3 \right] \\ k_H &= 2(2/3)^{4/3} r_e^{2/3} \left[\frac{\epsilon + 0.307\epsilon^2}{1 + \epsilon/2} + 0.119\epsilon^3 \right] \end{aligned} \quad (16)$$

Fibers under investigation can be treated as slender bodies, and it would be sufficient for the present study to use the appropriate relations developed above to determine the drag on a fiber. The hydrodynamic drag on the fiber can be estimated if the settling velocity of the fiber is known. In section § 3.3.4 the settling velocities of the fibers which are experimentally determined are presented.

3.3 Experimental Studies

In the presentation on the theory of fiber orientation in electric fields, various aspects were discussed that affect the alignment process. However, it was realized that there were several limitations in the prior studies (as discussed below) which made it important to experimentally verify the alignment behavior of fibers in air before designing a process that can manufacture aligned discontinuous fiber composites.

- First, the fiber alignment time derivations were based on small ellipsoidal particles in creeping flows. The dimensions of the fibers that are going to be used in composite manufacture are at least an order of magnitude larger than the fibers that

were experimentally verified by the models discussed above by previous investigators.

- Secondly, the alignment behavior was characterized for neutrally buoyant particles. In the case where fibers like glass or carbon are to be aligned in air, the density mismatch between the fibers and the fluid medium (air) is extremely large ($\sim 10^3$). So, it is expected that rotational motions of the fibers in the three dimensions will play a role in the fiber alignment process.
- Thirdly, all the models were derived for the case of a single fiber aligning in a fluid medium. In real processing situations, fiber-fiber and fiber-wall interactions will play a major role in the effectiveness of the alignment process. This is an extremely complicated problem to be solved from first principles, hence experimental studies were conducted which shed some light on this aspect.

3.3.1 Experimental Setup

The critical aspect in the ADF process prototype development has been the ability to control fiber orientation and determine the various parameters that influence it. An experimental setup was built to study the orientation behavior of fibers as they settle due to gravitational force in electric fields with air as the fluid medium (Figure 3.3). A variety of glass fibers of lengths ranging from 0.125" to 1" have been investigated in this setup. The forces that are encountered by the fiber are shown in Figure 3.1. The key parameters

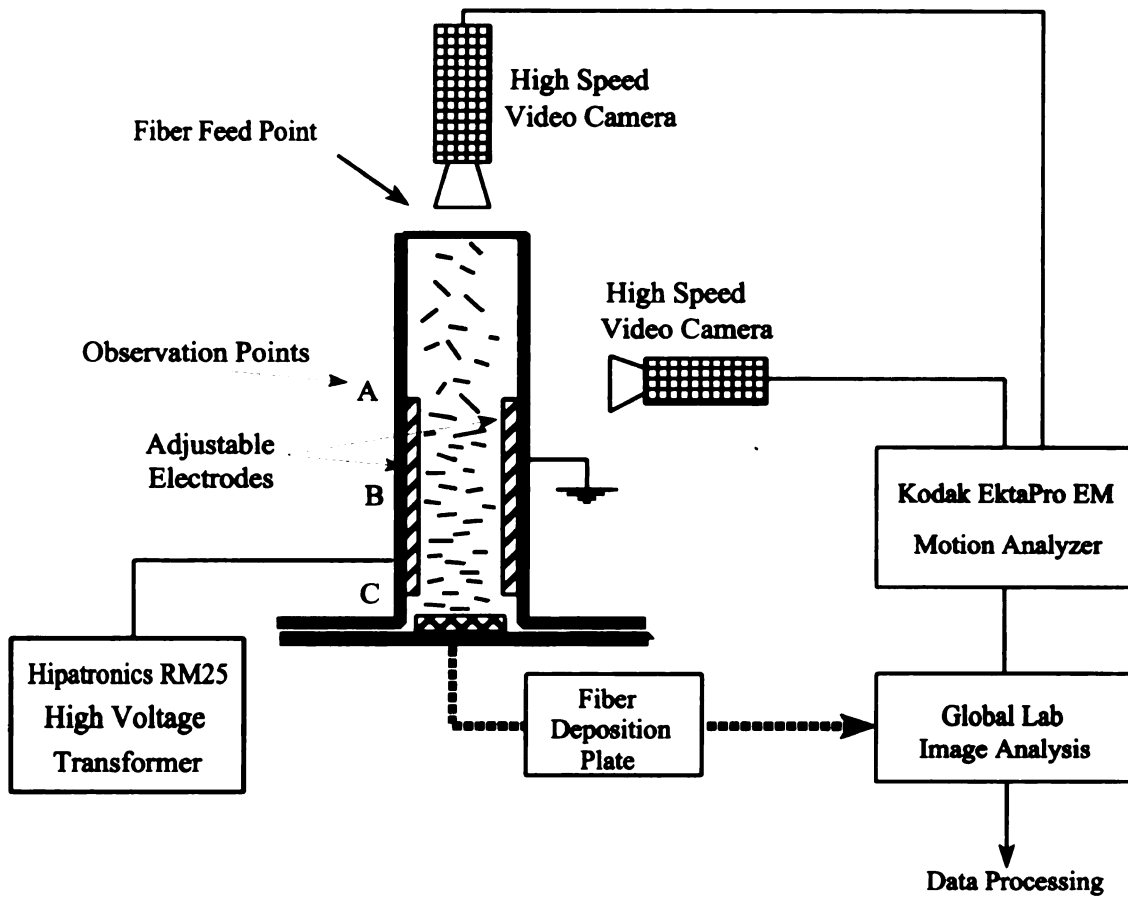


Figure 3.3 Experimental setup to investigate fiber alignment in electric fields.

investigated are the fiber alignment times, fiber settling behavior and fiber orientation distributions (FODs) as a function of E-field intensity and fiber geometric dimensions.

The experimental setup comprises of an open ended rectangular chamber with two vertical parallel copper electrode plates of dimensions 50 cm x 20 cm. These electrodes are supported by an insulating fiberglass framework. The electrodes are mechanically held in a groove provided in a set of insulating (phenolic) brackets. The insulating framework and other fixtures are fastened by plastic screws to avoid any distortion in the electric field due to the presence of conducting material.

The setup is so designed that the gap width between the electrodes can be easily varied from 3 to 10 cm in steps of 1 cm. The electrode gap width can be varied to account for large fibers and to control E-field intensity. The fibers are fed from the top in a controlled fashion and deposited on a bottom plate. The bottom plate on which the fibers are deposited can be located at two different heights (B and C), so that experiments can be performed at two different electrode heights of 25 and 50 cm.

A high speed video camera and Kodak EktaPro EM high speed motion analyzer is used to study the electro-dynamics of fiber motions. Recording up to 12,000 frames per second can be achieved by this system but a recording rate of 1000 frames per second was found to be adequate for this study. The recordings are stored in a video cassette recorder and printed on a thermal paper frame by frame. A provision was established to download

the images from the high speed motion analyzer into a computer through a frame grabber board. These individual digital images are further processed using Global Lab Image analysis software¹⁷ to determine fiber orientations or fiber translations.

The high voltage source was generated using a RM 25 Hipatronics transformer. In view of the high voltages that this transformer would generate and the potential for breakdown in air, it was not operated in air. The transformer is housed in a special tank that contains silicone oil, which is a dielectric fluid that has a greater resistance to voltage breakdown than air. A variac was used to control the electric field intensity (E) between the parallel plate electrodes. The electrode gap width was changed when it was felt necessary to maintain at least a 1:3 ratio between the fiber length and the gap width.

3.3.2 Behavior in D.C. Fields

The behavior of fibers in electric fields was observed for the first time in this study using D.C. fields. The observations were made in the experimental setup that was connected to a high voltage source generated from Hipatronics 60 KV D.C. power supply. The objective was to bracket the conditions of the electric fields under which the fibers would align. The gap width of the electrodes and the voltage was controlled to give a net variation in field intensity from 130 KV/m to 600 KV/m. The polarity of the high voltage electrode was tried with both -ve and +ve settings.

The behavior of four different types of glass fibers were tested by dropping them from the top of the orientation chamber in a random fashion. One was commercial grade chopped fibers (PPG 3830 of length 0.19 inches) which are meant to be used in molding operations and the other three types of fibers were hand cut to half inch length from continuous fiber rovings (see Table 3.2). General conclusions were arrived on the behavior of these glass fibers in D.C. fields by observations as noted in Table 3.2.

The general conclusion obtained from these scouting experiments was that glass fibers came under a strong electrophoretic force in D.C. fields, which disturbed any alignment achieved. The fibers got charged in the presence of high intensity fields and thereby got attracted to either the + ve or - ve electrode based on the sign of the charge the fibers acquired (Table 3.2). There seemed to be some relation between the type of sizing and the type of charge it acquired. This is apparent from the fact that the second and third type of fibers got attracted towards the -ve electrode while the fourth one moved towards the ground plate.

This first set of experiments, were encouraging in terms of the fact that E-fields were having an effect on the fibers of dimensions that are of interest for composite materials and there seemed to be a potential for fiber alignment. In other words electrical torques could be seen in action. The electrophoretic behavior of the fibers was however, complicated because it was not clear if the glass fibers were charged prior to their entry into the orientation chamber or if they got charged the moment they came under the influence of the

Table 3.2 Behavior of Fibers in D.C. Fields

Fiber Type	Observations
Chopped glass fiber strands (3/16")	Appeared too heavy and little effect of the field could be seen. Fell at the bottom of the chamber without any alignment.
Water sized glass fiber strands (1/2")	Came under electrophoretic effect and the fibers drifted to the high voltage plate (which was at - 45 KV). This drifting occurred near the top of the chamber.
Polyester sized glass fiber strands (1/2")	Effect of the field was observed. Drift was towards the -ve electrode.
Minimally sized glass fiber strands (1/2")	Strongly drifted towards the electrode but this time towards the ground (or away from the -ve electrode)

field. If it is assumed that the drift is due to the charging of the fibers as they come under the influence of the field, then this phenomenon can be explained by the nature of dielectric polarization.

Glass fibers being dielectric in nature and with the possibility of surface moisture and ionic moieties on the surface, it can be expected that the dielectric constant is a strong function of the frequency of the electric field. Depending on the frequency (f) of the field, polarization of dielectrics takes place in two stages. First, the moment the body comes under the influence of the field, instantaneous polarization due to electronic and atomic effects (electronic polarization) takes place. Later on the slower processes due to dipole orientation

and/or ionic migration (lossy polarization) takes place. With sufficient time the polarization of a dielectric material will reach saturation levels^{18,19}.

In the case of D.C. fields ($f = 0$), saturation polarization will be attained i.e both electronic as well as dipole polarizations will take place and this results in a strong electrical torque acting on the fiber. The strong electrophoretic effect observed in D.C. fields can be attributed to the strong electrical torque acting on the fiber combined with the negligible viscous resistance offered by air. If the time to attain lossy polarization is far less than $1/f$, then it is expected that saturation polarization will be attained. However, if the time to orient the dipoles is greater than $1/f$, then there will be no polarization due to the movement of permanent dipoles. Under A.C. fields, with the ability to control the frequency, f , it is possible that above a certain frequency the degree of polarization can be lesser than that obtained with D.C. fields. This can turn out to be more suitable for an effective fiber alignment process, especially when the fluid resistance is very low.

Some general conclusions were drawn from this set of experiments:

- Glass fibers of dimensions that are of interest in the composite materials have the potential to be aligned in electric fields in air as the medium.
- Fibers come under a strong electrophoretic force in the D.C. fields and thereby drift. So, it may be concluded that A.C. fields would provide a better choice to align fibers. The frequency of the A.C. field at which optimum alignment is possible can be

estimated from the polarization times of the fiber material and its surface characteristics. Initial calculations indicated a low frequency of 60 Hz should work for this application, since polarization times of these fibers are estimated to be of the order of 10^{-4} sec²⁰. Secondly, the strategy was to investigate with A.C. fields at a frequency of 60 Hz for the alignment purposes since A.C. fields of that frequency are the most easiest and inexpensive to generate.

- It was also noted that the mode of fiber feeding had a significant effect on the alignment process. For example, when fibers are in clumps and or entangled, they fall through the orientation chamber with hardly any effect of the field. Therefore, for effective fiber alignment, the fibers should be as free as possible.

3.3.3 Behavior in A.C. Fields

The majority of the experimental work was conducted with A.C. fields at 60 Hz. This frequency of the A.C. field is considered low enough to neglect magnetic field effects⁵ and the relationships presented in § 3.2.1 and § 3.2.2 for the electrostatic field cases can be applied here as well. The effectiveness of the electric field on the behavior of orientation was observed at three stages: A, B and C (Figure 3.3). It is important to note from Figures 3.1 and 3.3, that in the orientation chamber, the E-field is normal to the parallel plate electrodes i.e. the E-field lines are oriented along axis "2". In the case of a perfect E-field fiber alignment $\theta = 90^\circ$ and $\phi = 90^\circ$. The likelihood of this kind of a fiber alignment occurring under practical condition is remote and possible only when the fiber is neutrally

buoyant. Glass fiber alignment in air is a non-buoyant case and it is possible that a fiber gets aligned in the E-field with $\phi = 90^\circ$ yet θ may not equal 90° . It will become apparent in the following sections that the effectiveness of fiber alignment on the deposition plate improves when $\theta \rightarrow 90^\circ$.

A high speed video camera with Kodak's Ektapro motion analyzer was used to study the electrodynamics of the fibers. Glass fibers of lengths ranging from 0.125 to 1 inch and E-field intensities ranging from 0 to 600 KV/m were used in this study. It was observed that an electric field intensity of greater than 300 KV/m was capable of orienting these dielectric fibers. As expected, it was observed that with an increase in the field intensity the degree of alignment increased. High degree of fiber alignments were recorded at the bottom of the orientation chamber for all the fiber lengths investigated. One such set of videographs is shown in Figure 3.4, where 0.5 inch long glass fiber bundles are aligning in an E-field of intensity 400 KV/m. Note the random orientation state of fibers in the top videograph, where there is no E-field. Similarly, Figure 3.5 shows 0.25 inch long glass fibers aligning in an E-field of intensity 400 KV/m.

3.3.3.1 Effect of E-field Intensity and Fiber Aspect Ratio

Ultimately, the alignment of fibers that settle on the deposition platform is of primary concern. So, the angles of the fibers that settled on the deposition platform were measured with respect to the electric field direction and an orientation distribution was plotted. The

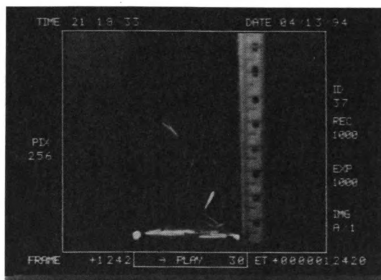
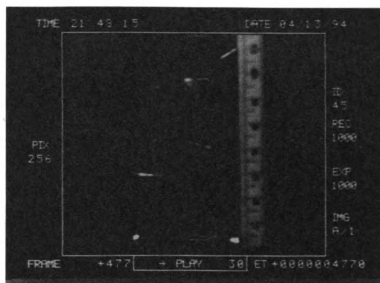


Figure 3.4 High speed videographs of free falling E-glass fibers (0.5" long) in the orientation chamber recorded at 1000 frames/sec under two conditions. Top: Fiber alignment in A.C. field of intensity 400 KV/m. Bottom: Random fiber orientation under no E-field.

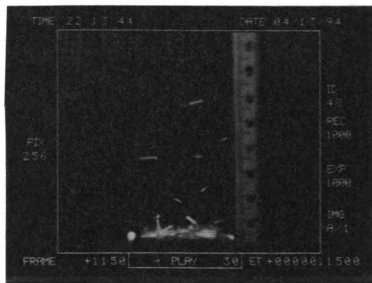


Figure 3.5 High speed videograph of free falling E-glass fibers (0.25" long) aligning in the orientation chamber in an A.C. field of intensity 400 KV/m (recorded at 1000 frames/sec).

details of the measuring technique for determining the fiber orientation distribution (FOD) are given in § 4.3.4. The fiber orientation distributions as a function of field intensity and aspect ratio is shown in Figure 3.6 for 0.25 and 1 inch long fiber cases. From the orientation distributions, it may be concluded that 1 inch long fibers align better than the 0.25 inch fibers at each of the E-field intensities 300, 400 and 500 KV/m. The FOD for 0.25 inch fibers improves (width of the distribution narrows) with an increase in the field intensity from 300 to 500 KV/m whereas the improvement of alignment in 1 inch long fibers is small. This data seems to contradict the relationships presented in § 3.2.2, where it was shown that the time required to align a fiber increased with fiber length (also see Figure 3.9). In other words for the same height of the orientation chamber and the field intensity, the 1 inch long fibers should align to a lesser degree than the 0.25" fibers and subsequently have a broader FOD. Secondly, the improvement in the degree of alignment as E_0 increased from 300 to 500 KV/m does not seem to follow a square relationship (see Eqs. 6 and 11). These results led to the investigation into the influence of hydrodynamics of fiber motion on the overall fiber alignment process in electric fields in air.

From a series of high speed experiments it was concluded that fiber alignment improves as θ tends to 0° during the fiber motion in the orientation chamber. This is due to two reasons. First, from the high speed videography it was observed that the impact energy absorption characteristics of the deposition plate was found to be a critical source for misalignment of fibers after they got aligned in the electric field. Moreover, the bouncing of the fibers on impact with the deposition plate decreases as the angle θ tends to 90° .

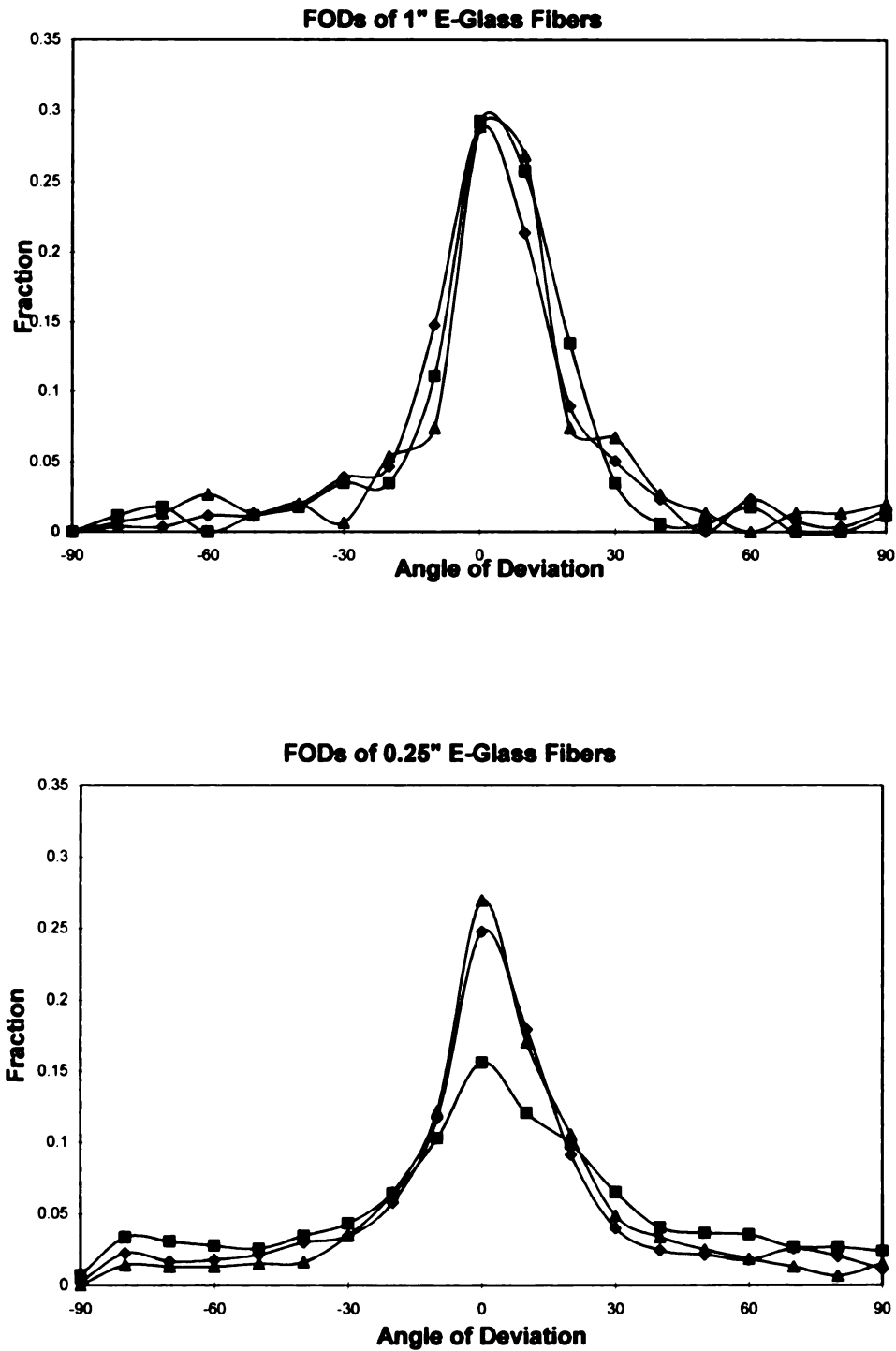


Figure 3.6 Fiber orientation distributions of 1" and 0.25" E-glass fibers settled at the bottom of the orientation chamber at A.C. field intensities of 300 (■), 400 (◆) and 500 (▲) KV/m.

Secondly, it was observed that the degree of alignment in the orientation chamber and on the deposition plate was dependent on the angle θ of the fiber at the entry into the electric field. This implies that as the fibers tend to fall horizontally, the effectiveness of the E-field to orient the fibers to the desired angle improves. Infact, the mis-alignment of fibers that seems to persist despite increasing the field intensity from 300 to 500 KV/m may be partly attributed to this phenomenon.

Under the same fiber feeding conditions for both the 0.25" and 1" long fibers at the top of the orientation chamber, it has been found that the 1" long fiber tends to attain a stable horizontal position by the time it is half way through the chamber whereas the 0.25" fiber does not attain a stable motion. This can be evidenced from the set of high speed videographs shown in Figures 3.7 and 3.8 taken in the absence of electric field. Figure 3.7 is a set of videographs taken at the bottom of the orientation chamber showing 0.25" fibers settling. It is clear that although there is some natural tendency by the fibers to occupy a horizontal plane a good number of them are also in a three dimensional random orientation state. Whereas, the tendency of the 1" fiber to occupy a horizontal plane is so predominant that it was recorded in a series of videographs as shown in Figure 3.8. Each frame is split into two to record the images from the two cameras that were used. One camera was placed on the top of the orientation chamber (right side of the frame) while the other was placed in front of the orientation chamber (left side of the frame). See Figure 3.3 for details. In the first frame, a 1" long fiber is released vertically. By following the motion of the fiber which

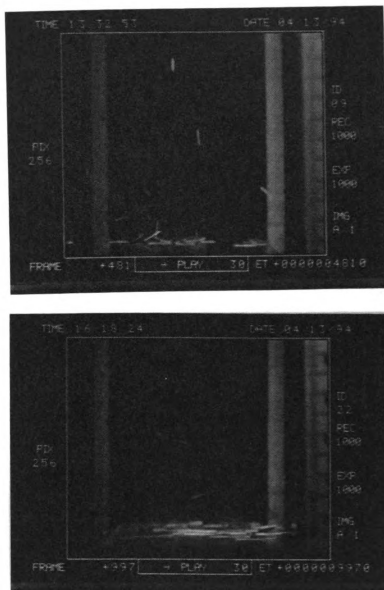


Figure 3.7 High speed videographs showing a greater tendency of 0.5" fibers in attaining a stable horizontal fiber orientation state than 0.25" fibers.

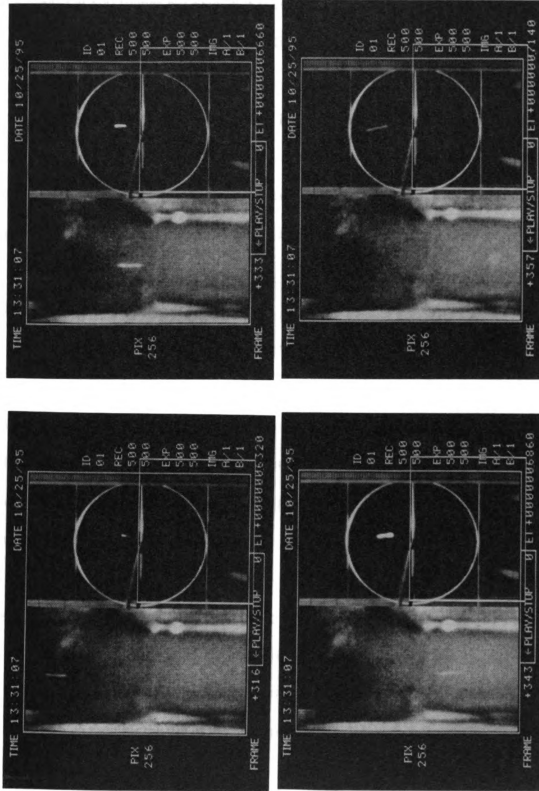


Figure 3.8 High speed videographs taken by two cameras showing the natural tendency of 1" long fibers in attaining a stable horizontal orientation state. Each frame is a composite shot of the top view (right) and side view (left).

is recorded at 500 frames per second, it is evident that within a short period of time, 0.082 seconds from frame 316 to frame 357, the fiber occupies a horizontal plane.

The reason for better alignment of 1 inch long fibers over 0.25 inch fibers can be partly explained by the above observations. And the reason for better stability in the fiber motion as the length increases can be directly explained using Jeffrey's model for fiber motion in shear flows (as discussed earlier in § 3.2.3.2).

3.3.3.2 Alignment Time

The fiber alignment time can be determined in the case of a neutrally buoyant particle situation by measuring the time it takes to rotate from an initial angle ϕ_i to a final angle ϕ_f with respect to the E-field lines. Although it seems very simple for experimental determination, in the fiber/air case that is under investigation, the fiber motion is not neutrally buoyant and therefore the fiber alignment in the "2" direction is coupled with fiber motion in the "3" direction (Figure 3.1).

- With the suspension medium being air, the stability of fiber motions and the effect of external drafts could not be avoided. Combined with this, the total settling and alignment times for the fibers under investigation used were less than 0.5 seconds, which requires the use of the high speed video camera and the cumbersome lighting arrangements to record the events.

- Ideally, in order to record both the alignment and drift of the fiber in the orientation chamber two high speed video cameras are needed. One camera can traverse down the orientation chamber at approximately the same speed at which the fiber is falling. A second camera at the top of the orientation chamber which has a dynamic focussing capability to track the fiber as it is falling. This camera will record the drift in the fiber translation. Although this kind of an arrangement was not available, a wealth of information was obtained by high speed videography using at least one and sometimes two cameras in fixed positions.
- Glass fibers are translucent and reflective in light at certain angles of incidence. This nature of the fibers combined with their small dimensions makes it very difficult to get good images of the fiber as it is translating and rotating. Optimum bright lighting arrangements had to be made from the front and the top of the orientation chamber to get the fiber in each frame, which made experimentation quite difficult especially when the electric fields also had to be used.
- The gap width of the electrodes could not be spaced wide apart so that there are no wall effects during fiber motion. For most of the experiments a minimum E value of 300 KV/m was needed to orient the fibers, this put a limitation on the maximum gap width that can be used because of the limitation in the maximum voltage that can be generated by the transformer.

Therefore, an alternative approach was used that combines experimental observations with model predictions for alignment times and arrive at an estimate for fiber alignment time

for the glass fibers. Both Fuchs and Demetriades models were used in conducting simulations for alignment times of glass fiber bundles that were used in the experimentation and subsequent development of the ADF process. Since the fibers were aggregates of filaments, an equivalent aspect ratio had to be determined before using the models for fiber alignment. Microscopic examination of the chopped fiber bundles indicated that they are elliptical in cross section and the individual filaments are packed in almost a square array instead of a hexagonal array. In order to translate the bundle size into an equivalent fiber diameter, it is considered that the elliptical cross section of the bundle can be transformed into a circular cross section with all the fibers close packed in a square array. Aspect ratios for all the E-field and hydrodynamic computations are based on this equivalent bundle diameter, d_{be} . For a given number of fibers per bundle (N_{fb}) the d_{be} is given by the following relationship.

$$\frac{d_{be}}{d_f} = \sqrt{\frac{4}{\pi} N_{bf}} \quad (17)$$

Both Demetriades and Fuchs models give similar trends in alignment times excepting the curves are slightly offset and the model by Fuchs predicts longer times (Figure 3.9).

The height of the electrodes was based on the alignment time computations which was estimated to be about 0.5 seconds for a fiber bundle of length 1 inch, the fiber bundle will get aligned in an electric field of intensity ranging from 250 to 500 KV/m. Considering that the settling velocities of glass fiber bundles was around 1 m/s (refer §3.3.5), the height

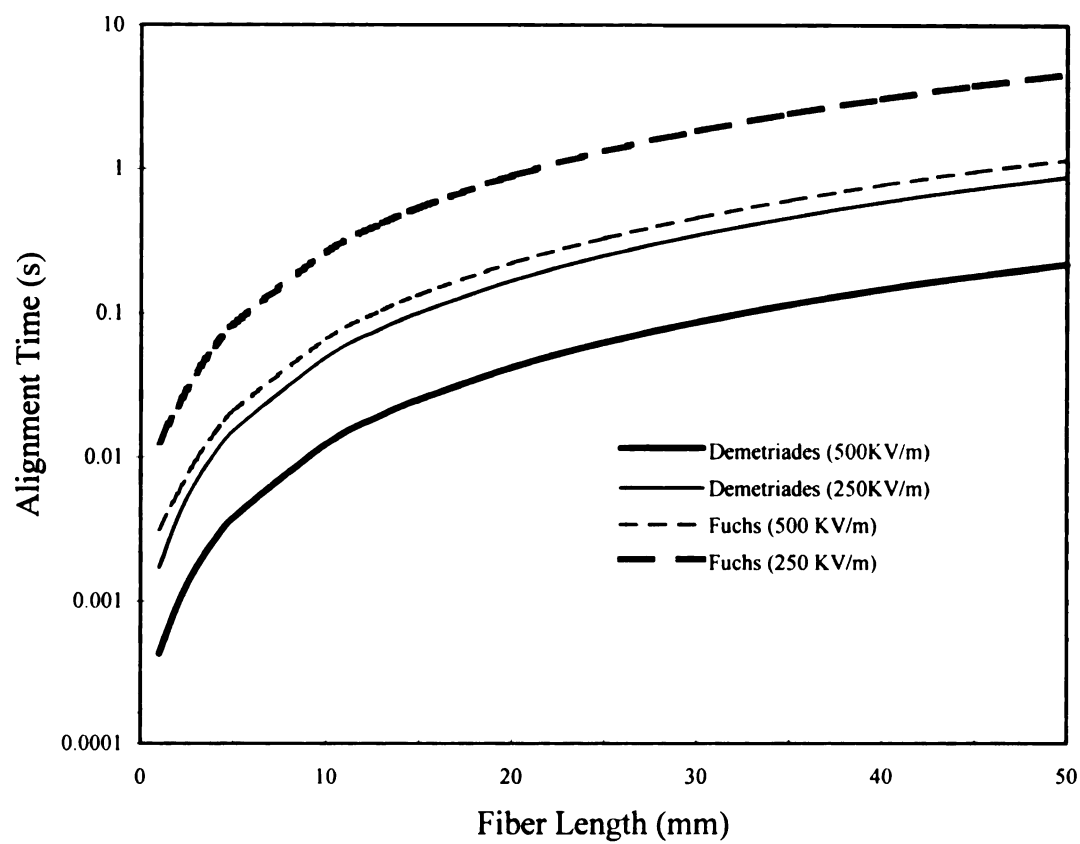


Figure 3.9 Alignment time simulations based on Demetriades and Fuchs models.

of the

orient

alignm

invest

3.3.4

of the

some

bundle

•

•

of the electrode is designed to be 50 cm. Observations were made at different heights of the orientation chamber using a series of high speed video recordings. High degree of fiber alignments were recorded at the bottom of the orientation chamber for all the fiber lengths investigated.

3.3.4 Fiber Settling Behavior

Slow motion analysis of the high speed video was used to assess the settling behavior of the fibers. The objective here being to compare and see if the settling behavior follows some of the theoretical predictions that were discussed in § 3.2.3. The motion of the fiber bundle is also qualitatively characterized (see Figure 3.1 for the coordinate system) as:

- Planar*** - bundles fall with a rotation in the horizontal plane (1-2);
- Horizontal*** - the bundles with almost no inclination in the 1-2 plane;
- Vertical*** - the bundles fall with no inclination in the 1-3 plane;
- Tumble*** - when the motion cannot be classified under any of the above.

- The fibers tend to fall *Horizontal* under terminal conditions. In the presence of an A.C. electric field (400KV/m), the fiber motion tends to be more *Horizontal* with almost no *Planar* motion. There was no significant difference in the terminal settling velocities of fiber bundles with and without the electric field.
- The impact energy absorption characteristics of the deposition plate has been identified as a critical source for misalignment of oriented fibers.

- Terminal settling velocities of 0.25, 0.5, and 1 inch long fiber bundles is about the same indicating *slender body* behavior.
- Reynolds Nos. indicate that the fiber bundles fall in the intermediate regime. Inertial effects have to be considered as the bundles are not settling under *creeping flow*.

Settling Velocity: From the high speed video recordings, the fiber motion was digitized frame by frame and fibers which had motions that were close to either “vertical” or “horizontal” in orientation were selected. The velocity of the translation of the centroid of the fiber with respect to a reference frame, was measured from a series of images and an average value is calculated. This kind of measurement was made for three different fiber lengths tested and the settling velocities are plotted as a function of the fiber length and the type of motion that is characterized. Figure 3.10 shows the spread of the data points when the fiber motion is recorded at observation point B, while Figure 3.11 shows the results at the point C (the bottom of the orientation chamber). It has been found that there is little or no increase in the settling velocity of the fibers as they move from region B to C, indicating terminal conditions have been attained. A careful analysis of the spread in the settling velocity data shows that at both points B and C the values range from 1.0 to 1.6 m/s. The orientation state of the fiber has a very strong influence on the settling velocity. The settling velocity of the fiber in a vertical orientation is the highest and the settling velocity in a planar or horizontal orientation is the lowest. From the relations given in § 3.2.3.3 for slender bodies it can be seen that as the aspect ratio of a fiber increases the settling velocity tends to level off. It may be concluded that the aspect ratios of the glass fiber bundles under

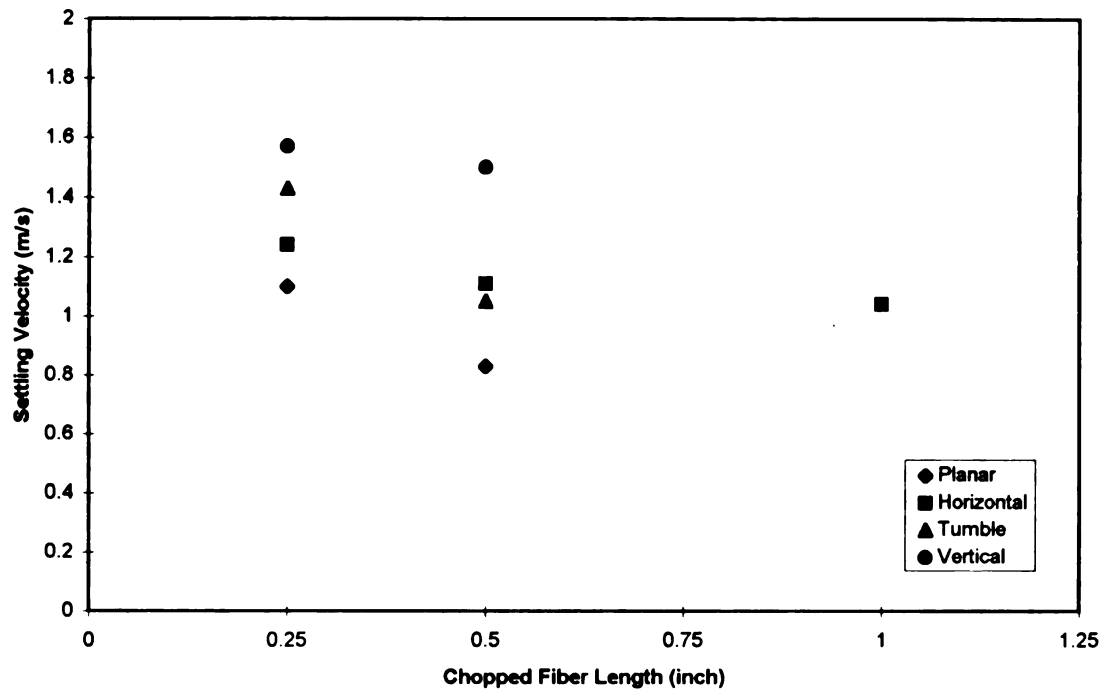


Figure 3.10 Settling velocity of fibers (Observation Point B).

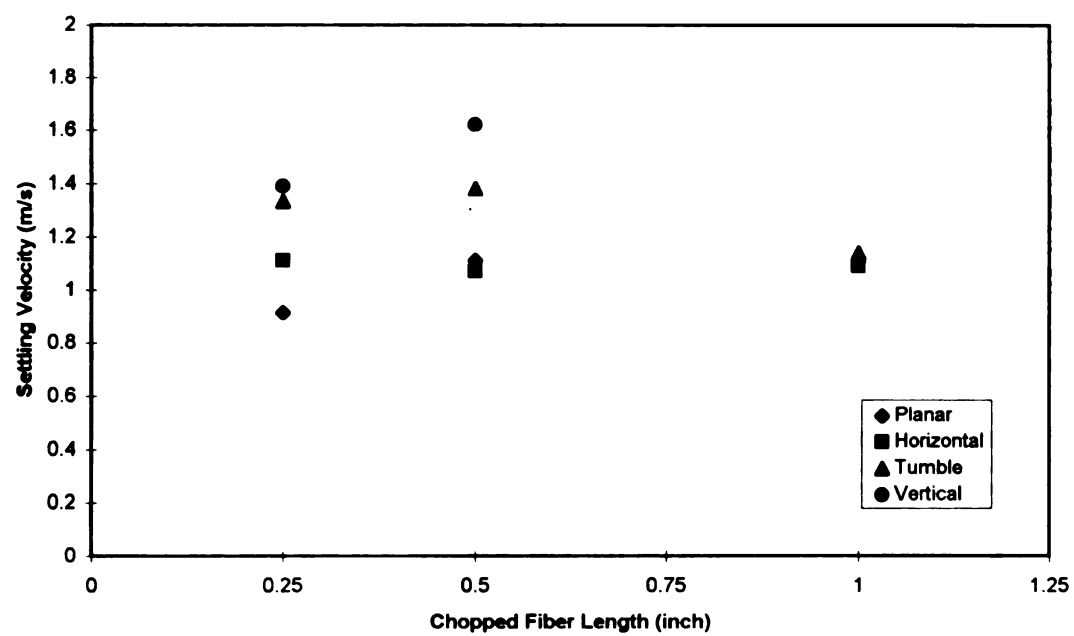


Figure 3.11 Settling velocity of fibers (Observation Point C).

investigation (lengths ranging from 0.25" to 1") were long enough to reach the asymptotic values.

3.4 Summary of Results

E-glass fibers of lengths ranging from 0.125 to 1 inch have been successfully aligned using alternating current (A.C.) electric fields with intensities ranging from 300 to 600 KV/m. A.C. fields were preferred over D.C. fields to avoid electrophoretic effects which disturbed the fiber alignment process. Based on the polarization characteristics of dielectric materials, A.C. fields with a frequency of 60 Hz was chosen and found to be adequate for effective fiber alignment in the ADF process.

The fiber alignment in air using electric fields is extremely fast and the estimates for E-glass range between 0.25 to 0.5 seconds for the fibers lengths investigated (ranging from 0.125 to 1 inch). It is very interesting to note that the theoretical predictions from Demetriades model for a 1 inch long fiber bundle predicts 0.25 s and 0.06 s at 250 and 500 KV/m respectively while Fuchs model predicts 1.3 s and 0.3 s for the same situation. So despite the fact that $Re < 1$ does not hold good for the fiber motion in this study, the models (especially Demetriades) serves to give a reasonably good estimate of alignment times for a first order approximation.

A very interesting settling behavior has been observed in this study. The orientation state of the fibers as they approach near the bottom of the orientation chamber tends to

become horizontal, the trend being more predominant as the fiber length increases from 0.25 to 1.0 inch. Based on the settling velocity and the Reynolds numbers (10 - 25)that were calculated this behavior made perfect sense. This tendency of fiber settling has some unique benefits to the fiber alignment phenomenon, since it brings stability in the fiber motion during free fall. The alignment behavior of the fibers in air is more complicated than it is in a more viscous fluid medium due to the non-buoyant nature of fiber motion. With air as the fluid medium, the viscous resistance is small and gravitational and the rotational forces become important. Fiber alignment becomes a strong function of not just the E-field strength but also on the stability in the fiber motion. Although it is predicted (in the case of buoyant situation) that the alignment time increases with fiber aspect ratio, it has been found that longer fibers have better alignments for the same electric field intensity due to their more stable motion during free fall.

References

1. Communication by Mike Rich based on unpublished data, 1995.
2. Glor, M., "Electrostatic Hazards in Powder Handling", John-Wiley & Sons, 1988.
3. Meek, J. M. And Craggs, J. D., "Electrical Breakdown of Gases", John Wiley & Sons, 1978.
4. Herrera-Franco, P. J. And Drzal, L. T., "Comparison of methods for the measurement of fiber/matrix adhesion in composites", *Composites*, Vol. 23, 1992.
5. Stratton, J. A., *Electromagnetic Theory*, McGraw-Hill, pp 215-217, 1941.
6. S. T. Demetriades, *J. Chem. Phys.*, Vol 29, p. 154, 1958.
7. Arp, P. A., Foister, R. T., and Mason, S. G., "Some electrohydrodynamic effects in fluid dispersions", *Advances in Colloid and Interface Science*, Vol 12, p 295-356, 1980.
8. Fuchs, N. A., *The Mechanics of Aerosols*, The Macmillan Co., 1964.
9. Lilienfeld, P., "Rotational Electrodynamics of Airborne Fibers", *J. of Aerosol Science*, Vol. 16, No. 4, p 315-322, 1985.
10. Jayaweera, K. O. L. F. and Mason, B. J., "The behavior of freely falling cylinders and cones in a viscous fluid," *J. Fluid Mech.*, 22, pp 709. 1965.
11. Pettijohn, E. and Christiansen, E., *Chem. Eng. Prog.*, 44, pp 157, 1948.
12. Newsom, R. K. and Bruce, C. W., "The dynamics of fibrous aerosols in a quiescent atmosphere", *Phys. Fluids*, 6 (2), pp 521, 1994.
13. Tucker, S. G. and Advani, S. G., Processing of short fiber systems, *Flow and Rheology in Polymer Composites Manufacturing*, Ed. Advani, S. G., Elsevier Science, 1994.
14. Jeffery, G. B., "The motion of ellipsoidal particles immersed in a viscous fluid, *Proc. R. Soc.*, London, A 102, pp161, 1922.
15. Jayaweera, K. O. L. F. and Mason, B. J., "The behavior of freely falling cylinders and cones in a viscous fluid," *J. Fluid Mech.*, 22, pp 709. 1965.
16. Batchelor, G. K., *J. Fluid Mech.*, Vol 44, p 419, 1970.

16. Global Lab Image Analysis Software Manual, 1995.
17. Bartnikas, R. and Eichhorn, R. M., "Engineering Dielectrics Volume IIA, Electrical Properties of Solid Insulating Materials: Molecular Structure and Electrical Behavior", ASTM, 1983.
18. Morton, W. E. and Hearle, J. W. S., "Physical Properties of Textile Fibers", The Textile Institute (UK), 1993.
19. P. Lilienfeld, "Rotational Electrodynamics of Airborne Fibers", J. of Aerosol Science, Vol. 16, No. 4, p 315-322, 1985.

Aligned Discontinuous Fiber (ADF) Composite Process*

4.1	Novel Concept	91
4.2	ADF Process Design	94
4.2.1	Unit Operations	94
4.2.2	Fiber Feeder	97
4.2.2.1	Fiber Pre-alignment	99
4.2.3	Electric Field Orientation	101
4.2.3.1	Orientation Chamber	101
4.2.3.2	Electrode Design	102
4.2.3.3	High Voltage Source	106
4.2.4	Deposition Platform	108
4.2.5	Powder Coating	110
4.3	Process Performance	112
4.3.1	Fibers	113
4.3.3	Operating Conditions	114
4.3.4	Fiber Orientation Distribution (FOD) Measurement	117
4.3.5	Aligned Vs Random	121
4.3.6	Concentration Effects	123
4.4	Process Consolidation	128
4.4.1	Process Consolidation Cycle	130
4.4.2	Consolidation Pressure	130
4.5	Manufacturing	133
4.5.1	Processing Rates	133
4.5.2	High Speed Manufacturing Schemes	136
4.6	Summary	138
	References	141

* Filed for US Patent (1996).

4.1 Novel Concept

Recent advances in the area of polymer powder coating of fibers at MSU^{1,2,3} in combination with the physics of aligning fibers in electric field, has paved the way to conceptualize and develop a novel high speed processing methodology that can manufacture aligned discontinuous fiber composites. Full realization of the stiffness to weight benefits of these composites is possible due to effective fiber alignment combined with the ability to consolidate at higher volume fraction of fibers. Absence of solvents or liquids in the Aligned Discontinuous Fiber (ADF) process improves the speed of processing many fold and makes the process environmentally friendly. The fiber alignment technique of the ADF process is rapid and simple in operation and design, and potentially adaptable to integration into existing composite sheet or lamination processing. Unlike continuous fiber random mat reinforced composites which have poor drapeability and which have problems of delamination under compression, aligned discontinuous fiber composites are flexible and can be molded or stamped into complex parts. In this chapter, the details of the process that was designed, built and operated are presented. The continuous manufacturing schemes that can be derived from this approach are also presented at the end of the chapter.

The concept of the ADF process is very versatile and can lead to three possible routes for manufacturing discontinuous fiber polymer composites (Figure 4.1).

- The first one (shown by a solid line) can be a semi-continuous process where the ADF preform impregnated with polymer powder (at volume fractions > 50 %) is stacked to a desired thickness and consolidated under a temperature and pressure

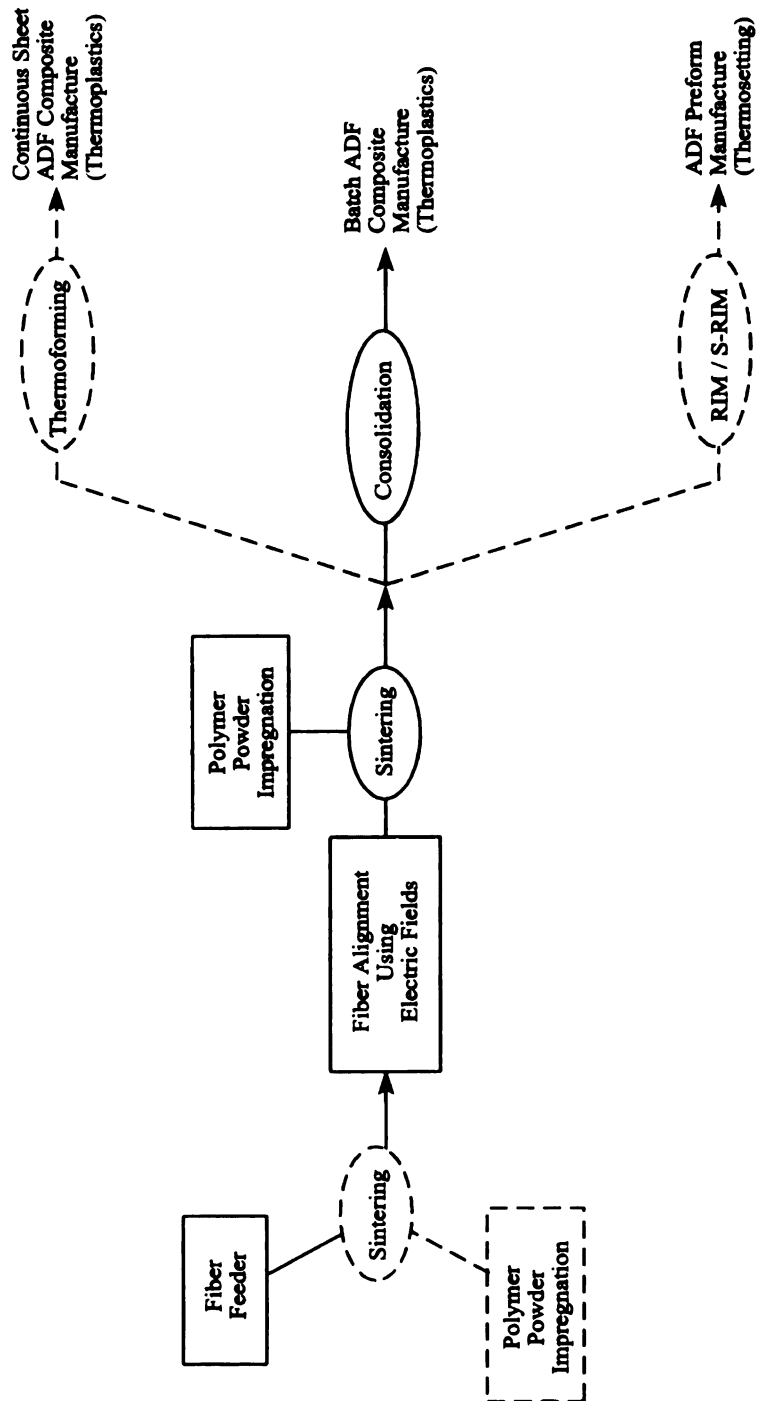


Figure 4.1 Novel concept of Aligned Discontinuous Fiber (ADF) composite process.

cycle to obtain a composite. This approach is well suited for the high melt viscosity thermoplastic polymers. It reduces the time and pressure requirements during the consolidation step since the polymer is already in place and has to flow only locally.

- The second approach (top dotted line), also well suited for thermoplastics, consists of the continuous manufacturing of ADF composite in sheet form. Blanks cut from this sheet can be thermoformed to a desired part, using high speed manufacturing techniques. It is envisioned that the continuous manufacturing of ADF composites will be a direct outcome of the principles established in the first approach.
- The third option (bottom dotted line) has potential for use with thermosetting resins. Here, the amount of powder coating is kept to a minimum (volume fractions $< 10\%$), so that sintered particles act as binders that hold the ADF preform together in a semi consolidated state. This preform is then injected with a low viscosity cross-linking resin via Reaction Injection Molding (RIM) or Structural Reaction Injection Molding (S-RIM) process to form an ADF composite. The resin gels and forms a solid network with time and temperature (depending on the cure kinetics). This resin injection approach is well suited for thermosetting polymers and fabricating large parts.

Having identified the potential features of this novel concept, the focus of this research is to design, fabricate and develop a prototype Aligned Discontinuous Fiber (ADF) composite process that achieves to establish the following objectives:

- High speed processing capability for manufacturing aligned discontinuous fiber polymer composites, using electric fields in air.
- Improvement in mechanical properties of discontinuous fiber composites with fiber alignment over random discontinuous fiber composites.

4.2 ADF Process Design

The prototype Aligned Discontinuous Fiber (ADF) composite process that has been developed is shown schematically in Figure 4.2. This prototype ADF process is designed with the dual capability of operating under batch mode when the deposition platform is stationary; or semi-continuous mode when the platform repetitively travels back and forth. Chopped fibers are fed from a specially designed fiber feeder in to the orientation chamber. Fibers are aligned in this chamber by the electric fields and settle on the moving deposition platform. The speed controllable linear platform mimics the motion of a moving belt.

4.2.1 Unit Operations

For the purposes of understanding and designing the ADF composite process it was necessary to first identify the unit operations and establish the design parameters focusing on the principles of processing rather than the rate of manufacturing (see Figure 4.2).

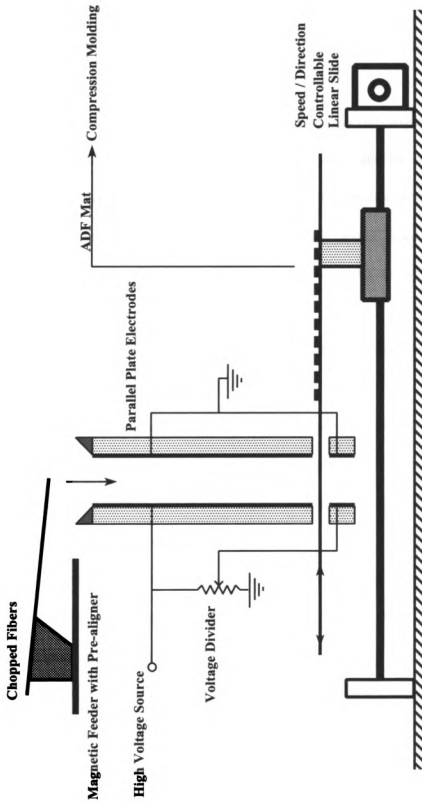


Figure 4.2 Schematic of the prototype Aligned Discontinuous Fiber (ADF) composite process.

The unit operations identified for the ADF process are:

- Fiber alignment using electric fields
- Polymer powder impregnation of fibers
- Consolidation of polymer powder coated aligned discontinuous fiber preform

As shown in Figure 4.1, the polymer powder impregnation and the subsequent sintering operation can take place either before or after the fiber alignment step. In order to keep the development of this process focussed to fiber alignment in electric fields, the powder coating step is taken up after the fiber alignment step. The following design features have been identified as critical in developing the ADF process.

- Method for pre-alignment of fibers using a suitable feeding system to control transfer into the orientation chamber.
- An orientation chamber with the necessary residence time for fiber alignment and the flexibility to provide the desired electric field intensity and configuration.
- Identification of the effect of fiber alignment during dynamic deposition conditions when the platform is set in motion.
- Method for controlled impregnation of fibers with tribo-charged polymer powder.
- Determination of the ADF production and processing rates as a function of the maximum fiber concentration in the orientation chamber.
- Determine sensitivity of the degree of fiber orientation in the ADF preform mat on the mechanical performance of the ADF composites.

Design criterion was established for each of these features.

4.2.2 Fiber Feeder

The role of fiber feeding was critically evaluated for effective fiber alignment in electric fields. It has been observed that fibers having a random orientation state before entering the orientation chamber cannot be easily aligned by the E-field due to the tumbling nature of the fiber motion. This tumbling motion and random orientation state is also detrimental to the final degree of alignment because once the fiber hits the moving platform (belt or veil) at the bottom of the orientation chamber, the fibers will rebound and get misaligned. Fiber feeding poses another problem due to potential entanglement and clogging of the feeder, which results in uneven and intermittent fiber delivery. A number of designs of short fiber feeders were considered which were simple and inexpensive; which had the potential to feed the fibers with minimum clogging problems; and which had the scope to provide some preferred orientation to the fibers as they entered the orientation chamber.

The final design consisted of a modified vibratory FMC Syntron Magnetic feeder (Model FTOC). This feeder was chosen to feed the short fibers, with or without powder impregnation, into the orientation chamber (Figure 4.3). The vibratory motion provided by the feeder coupled with the inclination of the feed chute to the horizontal plane provides forward motion to the fibers. The vibratory mechanism also reduces entanglement and clogging of fibers. The mass flowrate of the fibers can be controlled by regulating the amplitude of the vibratory feeder. The feeder has a V-grooved chute, through which material flows downward. This type of an arrangement could not feed the fibers into the orientation chamber with a preferred fiber “pre-alignment”.

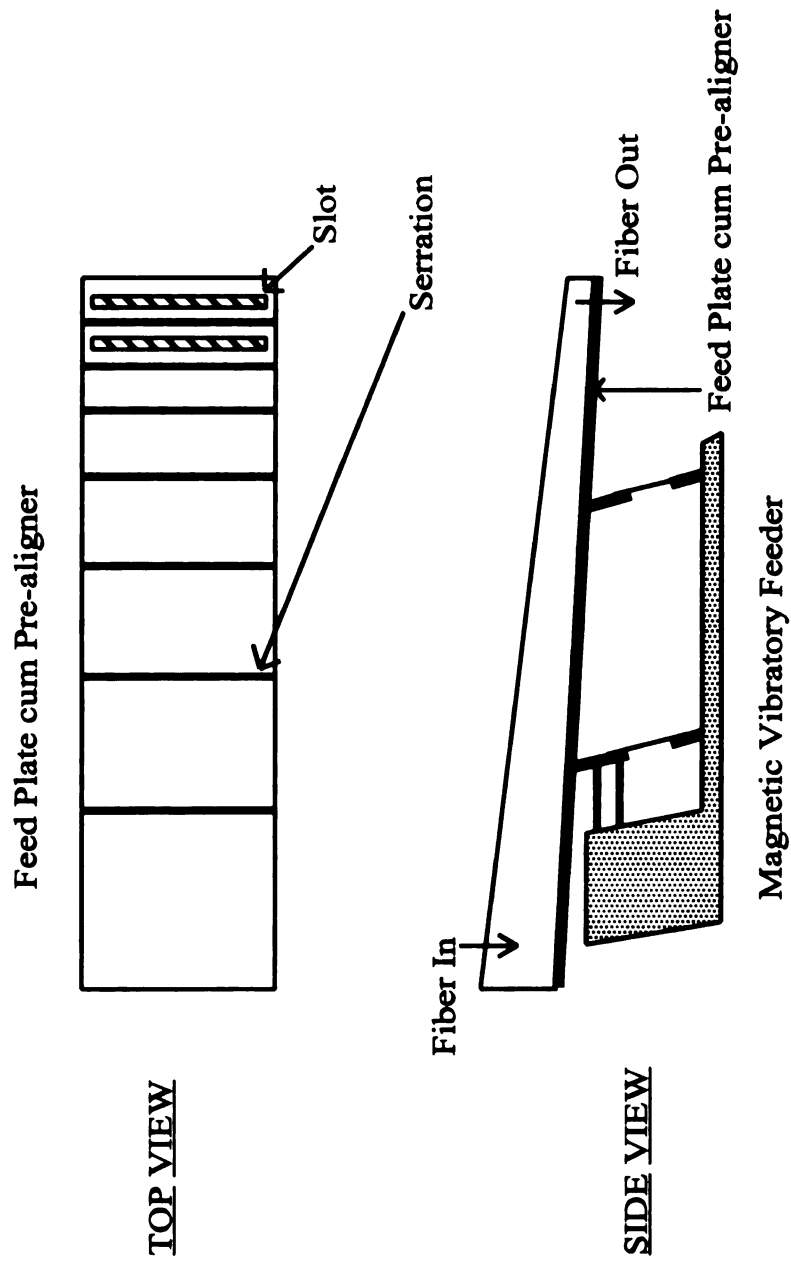


Figure 4.3 Fiber feeder cum pre-aligner.

As discussed earlier in § 3.4, the most desired configuration for “pre- alignment” is the fibers entering the orientation chamber in a horizontal plane ($\theta \rightarrow 0$). A unique pre-alignment system was developed and incorporated in the feeding system such that fibers fall with their long axis parallel to the deposition platform at the bottom of the orientation chamber.

4.2.2.1 Fiber Pre-alignment

The pre-aligner consists of a rectangular plate mounted on the vibratory mechanism which propels the fibers in one direction (Figure 4.4). The plate with raised corrugations at predetermined locations on the surface forces the fibers to align parallel to the corrugations as they are transported forward. When fibers are fed from one end of the plate in a random fashion, the pre-aligner changes the orientation of the fibers by the time they traverse the plate and then pass through a set of slots into the orientation chamber as shown in schematically in Figure 4.4. The corrugations were fabricated by embedding 0.0625 inch diameter stainless steel rods onto the plate. Corrugations on this plate are located at 2.0, 1.5, 1.0 and 0.5 inch spacings so that any fiber that is less than 2.0 inches long pre-aligned parallel to the corrugations as the fibers move towards the exit slots. If a fiber is greater than 2.0 inches, then it is possible that it can bridge over the corrugations without pre-alignment during its traverse. Because of the pre-alignment of the fibers as they exit through the slots in the plates and into the orientation chamber, a predominantly planar orientation state is maintained as depicted in Figure 4.4.

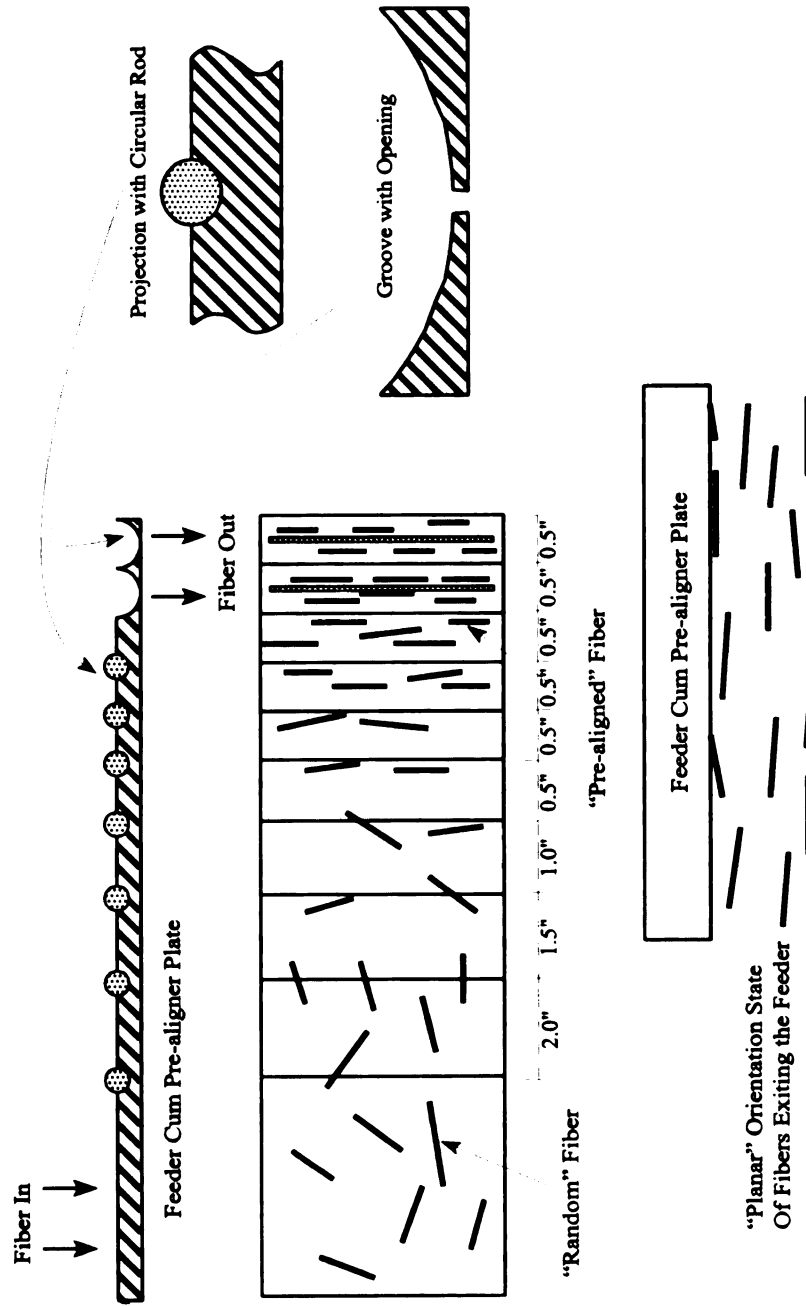


Figure 4.4 Mechanism of fiber pre-alignment.

4.2.3 Electric Field Orientation

Electric field orientation forms the key step in the ADF process. The orientation chamber and the electrodes were designed based on the principles of fiber alignment described in Chapter 3. The alignment of freely falling fibers in electric fields is further complicated when the fibers settle on a moving platform. The good alignment that is achieved when the fibers reach the bottom of the orientation chamber in a batch mode is totally mis-aligned when the linear platform is set in motion. During the development of the semi-continuous ADF process, it was found that the edge effects of the electrodes play a critical role in the fiber alignment process. Special features had to be incorporated in the electrode design to overcome edge effects.

4.2.3.1 Orientation Chamber

The fiber orientation chamber must have the necessary residence time for fiber alignment and the flexibility to provide the required electric field intensity and field configuration. The minimum electrode height can be calculated from the product of the two parameters: (a) fiber alignment time; and (b) fiber settling velocity.

$$\text{Min. Electrode Height} = \text{Alignment Time} \times \text{Settling Velocity} \quad (1)$$

These parameters have been estimated from the experiments on the prototype for glass fibers in lengths ranging from 0.125 to 1.0 inches. Based on this calculation a height of 50 cm was chosen for the electrode height as explained in Chapter 3. This orientation chamber is rectangular in cross-section with an open top and bottom, and fabricated of insulating

materials. The parallel plate electrodes are mounted on the inside faces. The height of the orientation chamber is 50 cm and the electrode gap width can be varied from 3 to 15 centimeters. The width of the ADF preform that is formed by the process was based on the size of the composite test panel that would be needed to determine composite mechanical properties. So the width of the orientation chamber was set at 25 cm, which produces an ADF preform of width 20 cm in this semi-continuous process.

4.2.3.2 Electrode Design

A parallel plate electrode configuration, which creates E-field lines that are parallel between the electrodes is used in the process for alignment of fibers in the direction of the moving platform. Depending on the electric field intensity and the fiber aspect ratios, the fibers tend to align along the field lines and deposit on the moving veil, located on the platform. In the batch mode when the deposition platform is stationary, the E field distortion at the edge of the electrode at the bottom of the orientation chamber did not affect the alignment of the fibers in any significant way (Figure 4.5). In the continuous mode of operation, the fibers that were already on the deposition platform would get mis-aligned as they passed under the high voltage electrode in the reverse direction. There was literally a “sweeping action” and the fiber alignment would be completely lost. Edge effects are created at the bottom of the electrodes because the edges have small radii of curvature and the tendency of the E-field to redirect itself toward the closest grounding point (Figure 4.6). The proximity of the deposition platform compared to the opposite ground electrode

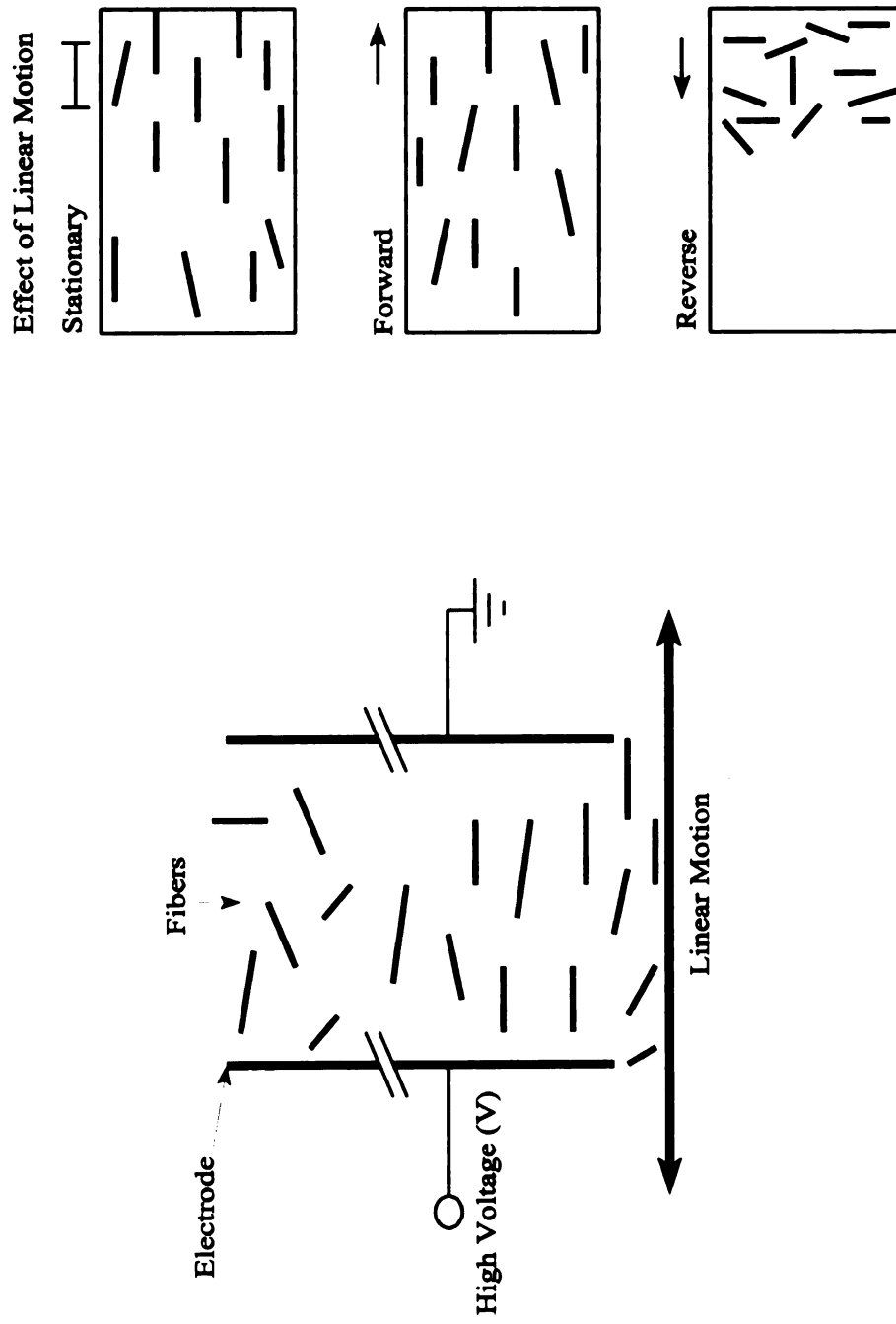


Figure 4.5 Electrode edge effect on fiber orientation.

provides a good source for grounding if the deposition platform is grounded. This will occur when the height between the electrode edge and the deposition platform, h is smaller than the electrode gap width, d . This edge effect is compounded because of the sharp radius of curvature at the bottom of the electrodes. A region of very high field intensity can be formed which can be many times the intensity in the regions far from the electrode edges. The E-field intensity (E) at the two different locations is given by the following relationships.

$$\begin{aligned}
 \text{Far Field: } E &= \frac{V}{d} \quad \text{where } d = \text{electrode gap width} \\
 \text{Near Edge: } E &\sim \frac{V}{2r} \quad \text{where } r = \text{radius of the edge} \\
 \text{Near Edge: } E &\sim \frac{V}{h} \quad \text{where } h = \text{electrode gap height}
 \end{aligned} \tag{2}$$

When a voltage of 25 KV is applied to the high voltage electrode with an electrode gap width of 5 cm, the E-field intensity (E) far from edge is 500 KV/m. At the same time, at the edge of the electrode which has a thickness of 0.0625 inch, the intensity will be about 15,000 KV/m. This is much higher than the far field intensity and any fiber near such a field is bound to get distorted. A corona discharge often takes place at this point because these intensities are higher than the breakdown voltages of 3000 KV/m in air.

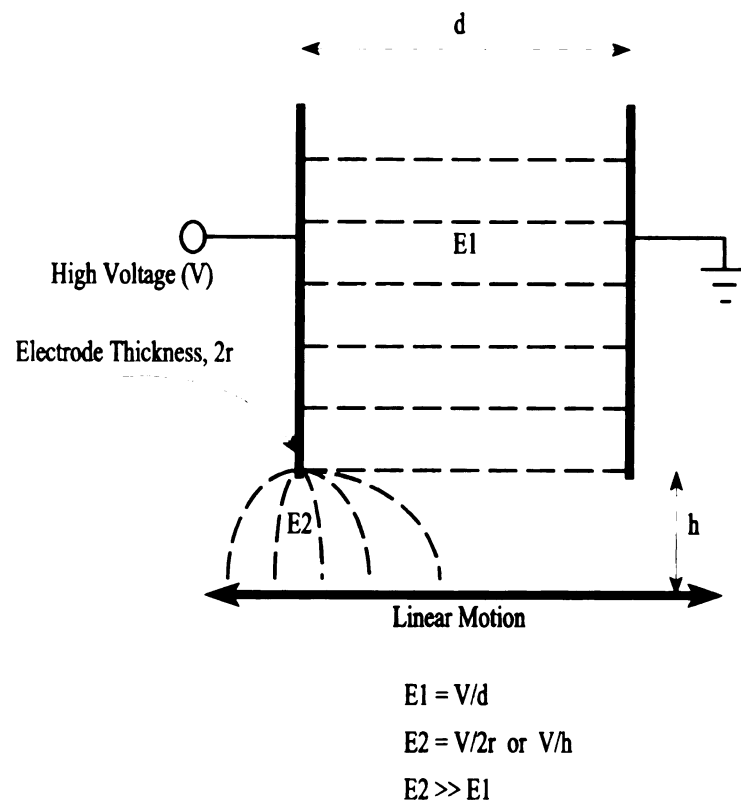


Figure 4.6 Electric field near the electrode edge.

To overcome this edge effect, the edge of the electrode plate was so designed that the radius of curvature is much greater, which will reduce the intensity. A conductive brass cylindrical rod was retrofitted to the edge of the plate which provided it with an effective radius of curvature of 0.25 inch (Figure 4.7). This decreased the degree of mis-alignment of fibers due to the reduction in the field intensity near the edge to 3900 KV/m, but mis-alignments could not be completely eliminated. A more effective solution to the problem of edge effects was found in a novel electrode design which created a neutralizing field just below the edge of the high voltage electrode (Figure 4.7). The neutralizing field is created by the set of electrodes that are placed directly below the deposition substrate. The high voltage of this neutralizing electrode was derived from a voltage divider. As a consequence, the electrode edge effects were completely eliminated from the process when the platform was set in motion both in the forward as well as the reverse directions. With this modification, the fibers that were once aligned on the mat did not get mis-aligned even at relatively high speeds of the deposition platform.

4.2.3.3 High Voltage Source

The high voltage needed for the ADF process is generated using a high voltage (HV) Hipatronics transformer (Model RM25). A.C. fields are created by linking the secondary output of the transformer to parallel plate electrodes made of copper. The electric field intensity can be controlled by two means: (a) by controlling the line voltage using a variac

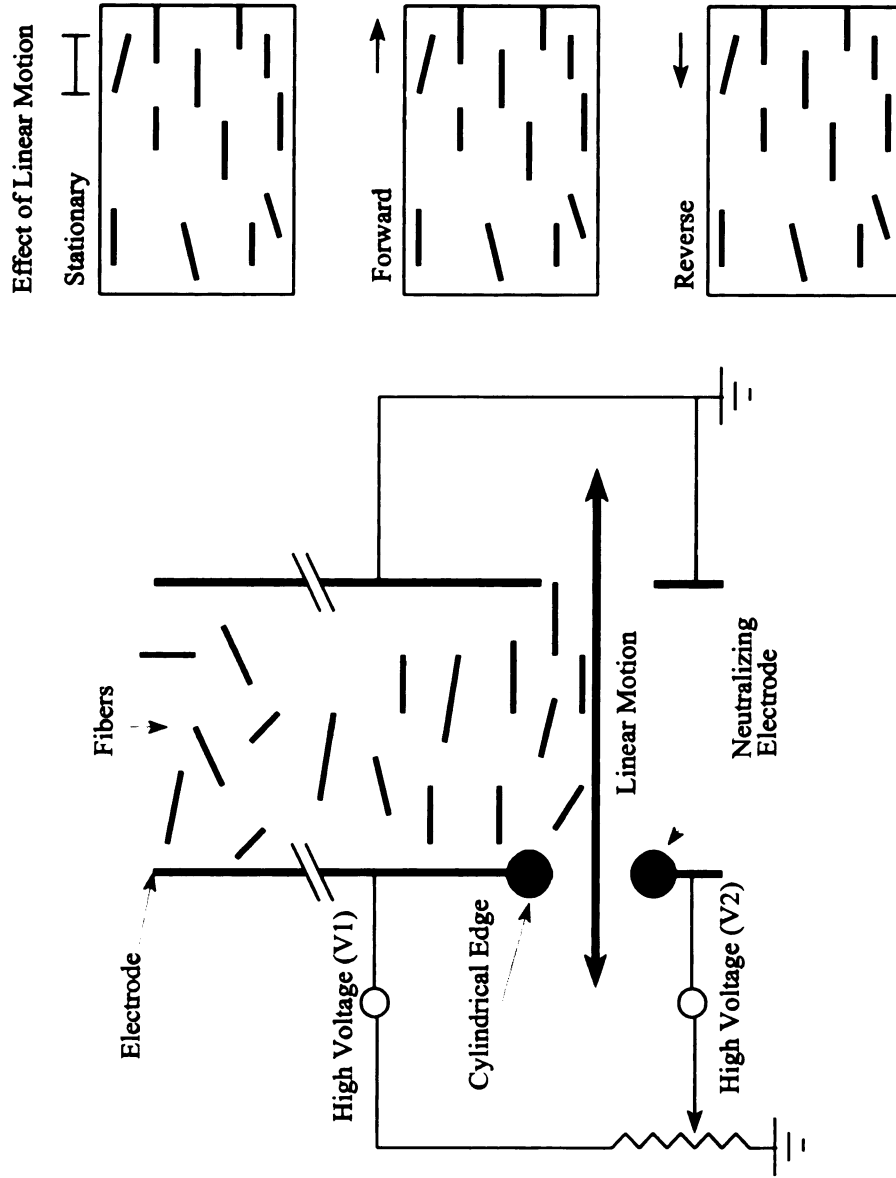


Figure 4.7 Electrode design with a neutralizing field for continuous processing.

thereby controlling the input voltage on the primary windings of the transformer; or (b) changing the gap width of the parallel plate electrodes. For a primary input voltage of 110V, the transformer has a rated output of 25 KV at 15 mA. The gap width of the electrodes can be varied from 3 to 15 cm, thereby creating a variation in electric field intensity from 833 KV/m to 167 KV/m. The voltage is monitored using a high voltage Fluke 80K-40 probe whose signal is sent to a digital Fluke 45 multi meter.

Figure 4.8 shows the circuit diagram of the voltage distribution to the various electrodes. The variac provides the first control on primary voltage of the HV transformer. The secondary from this HV transformer is sent to a voltage divider. The top electrode receives the full voltage of the secondary, while the bottom electrode receives a fraction of the voltage of the top electrode in four possible steps: 100, 75, 50 and 25 %. The settings are determined based on the processing conditions.

4.2.4 Deposition Platform

The aligned fibers with or without powder impregnation are deposited on a Teflon release film placed on a sliding deposition platform in the laboratory prototype. The forward and backward motion of the deposition platform on the linear slide simulates a continuous manufacturing operation. If fiber alignment is not disturbed during the forward and reverse motion, it can safely be assumed that the fibers will not be disturbed in the event of exposure of the aligned fibers to subsequent fields of the multiple orientation chambers. So the principles that are developed with this prototype can be directly translated to the

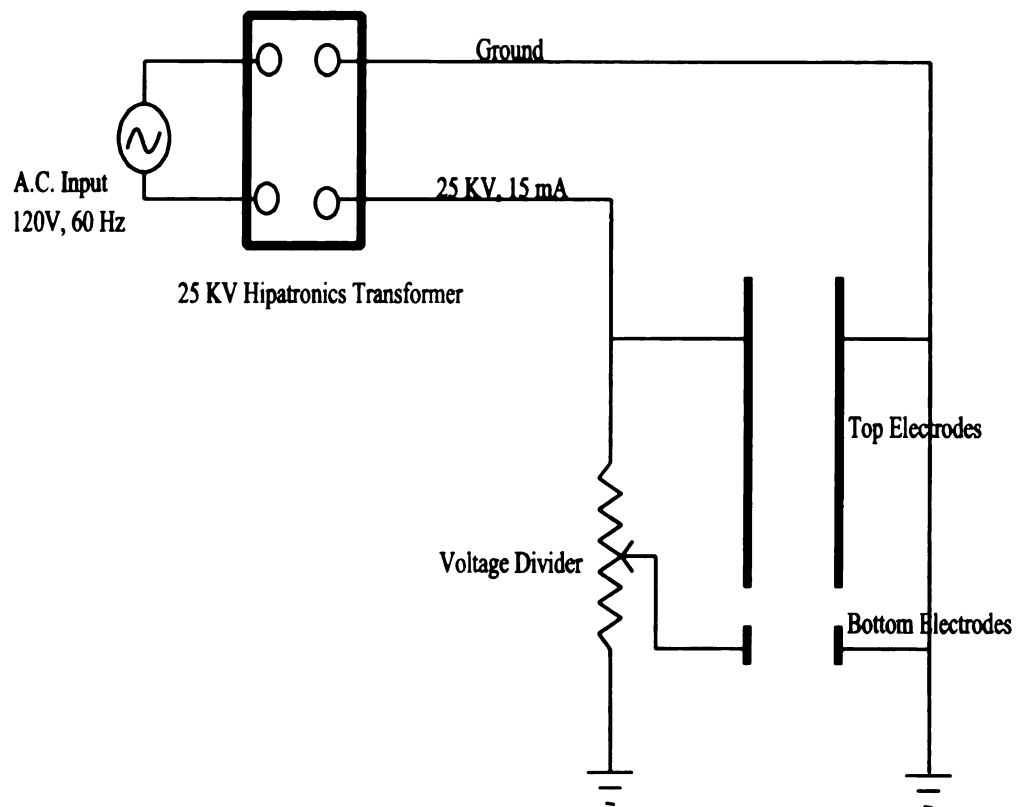


Figure 4.8 Circuit diagram for high voltage distribution.

manufacturing situation as discussed in § 4.5.4. The linear slide is driven by a D.C. gear motor connected to a speed regulator. Controllable forward and reverse motion results in the building up of an uniform aligned discontinuous fiber preform with the desired thickness. The largest uncut size of the ADF preform that can be formed is 20 x 35 cm, which allows physical property measurements to be conducted. The total traverse length of the platform is 30 inches. The forward and reverse motion is controlled by a speed controller and the speed of the platform can range from 10 cm/min to 180 cm/min. An operating speed of 50 cm/min was chosen for the prototype ADF process.

4.2.5 Powder Coating

Provision must be made to incorporate a step for controlled impregnation of fibers with tribo-charged polymer powder. The mode of impregnating polymer matrix on fibers is by bringing in contact an aerosol of fine tribo-charged polymer powder and fibers, to get a uniform coating of particles on the fiber. Acoustic aerosolization is an effective means to generate an aerosol of fine polymer powders of sizes less than 30 microns⁴. This aerosol can be entrained through a nozzle where the fibers are introduced, similar to the idea used in the high speed MSU Powder Prepreg Process developed by Vyakarnam and Drzal⁵. The amount of matrix or binder that gets deposited on the fibers is controlled by controlling the particle concentration flux in the aerosol, which is directly related to the matrix volume fraction in the composite. Figure 4.9 shows electron micrographs of E-glass fibers coated with 10 micron size nylon 12 particles. It is important to note that the uniform distribution of

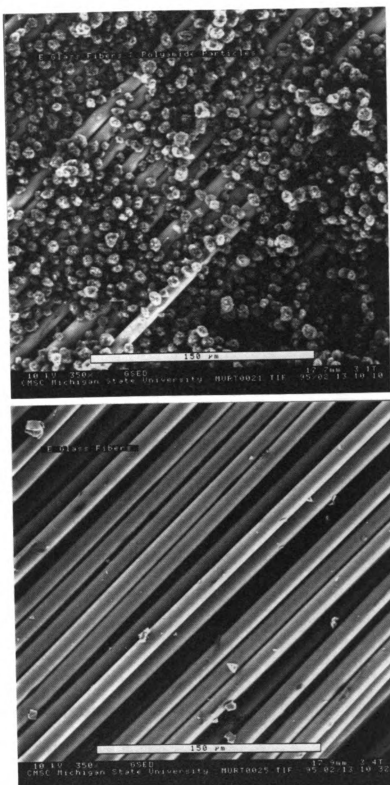


Figure 4.9 SEM micrographs of E-glass fiber bundles coated (Top) and uncoated (Bottom) with nylon-12 powder.

polymer particles around the fibers requires the polymer to only flow locally over small distances, thereby reducing the polymer melt flow times during the compression molding step⁶. This improves matrix impregnation, minimizes void formation during consolidation and improves the mechanical performance of the part. Powder impregnation makes compression molding of the ADF preform into a final composite part a rapid step.

Since the focus of the ADF process was more on the development of the fiber alignment technique, a continuous mode of impregnating discontinuous fibers in an aerosol of fine polymer powder was not developed specifically for the ADF process. Such a system can however, be easily developed based on the earlier work on powder processing. For the present investigations, polymer powder is introduced after the fiber alignment step by uniformly spraying a measured quantity of the powder on the ADF preform. The mass of powder that is required is calculated as per the volume fraction of the final composite. The ADF preform with the polymer particles is passed under an infra-red heater to sinter the particles in place. A conventional oven was also used to sinter the nylon 12 particles on the glass fibers making the ADF preform handleable.

4.3 Process Performance

First, the fibers and the polymer matrix used in the process development are characterized for their physical and thermal properties that are useful in determining the operating conditions of the process. Later the process operating principles and performance will be taken up.

4.3.1 Fibers

E glass chopped fibers of nominal lengths 0.125, 0.25, 0.5 and 1 inch were supplied by Owens-corning. The number of filaments per bundle was approximately 250. The length range and the number of filaments that were used in demonstrating the ADF process was based on the fiber alignment behavior in electric fields.

Length Distribution: The actual length of 100 fibers was measured that were picked randomly from the lot. The length distribution was plotted and it was found that chopping method used by Owens-Corning was extremely efficient and all the fibers were within 5 % of the nominal length. The lengths of the smaller length fibers were also measured at random and found to close to the nominal value. Based on these observations, the nominal length of the chopped fiber was used for all computation and felt that it was not necessary to include a length distribution.

Aspect Ratio Distribution: For each of the fiber length measured, its mass was also weighed. It was found that the mass distribution was not as sharp as the length distribution. The number of filaments for each of the fibers was also determined by a simple mass balance and found that there is variation in the number of filaments per bundle. The mean value was 227 instead of the nominal number of 250 that was reported (see Table 5.4). Since all the discontinuous fibers were obtained from the same rovings, this mean value for the number of filaments is used in all computations.

4.3.2 Polymer Characterization

The nylon-12 (polyamide) polymer powder was obtained from Atochem, Inc. The mean particle size of the powder was determined using Malvern Particle Size analyzer and the Image analysis techniques and was found to be 10 microns. The thermal transitions of this semi-crystalline polymer were evaluated using a Differential Scanning Calorimetry (DSC). The melting point of this polymer has a sharp onset starting from 171 °C and the glass transition temperature could not be detected clearly but is reported to be 40 °C.

4.3.3 Operating Conditions

The schematic of the ADF process given in Figure 4.2 can be referred to again for this section. Chopped fibers are fed from the specially designed fiber feeder in to the orientation chamber. Fibers are aligned in this chamber by the electric fields and settle on the moving deposition platform. The alignment of 1 inch long E glass fibers in electric field of intensity 400 KV/m, is shown by the digital images of the fibers that have settled on the deposition platform (Figure 4.10). The images of the fibers when there is no E-field and the best efforts to align manually using a pair of tweezers are also shown in the figure for comparison. The fibers are then sprayed uniformly with the polymer powder of a measured quantity. The speed controllable linear platform mimics the motion of a moving belt. The prototype ADF process is designed with the flexibility of operating under batch mode when the platform is stationary; or semi-continuous mode when the platform is set in linear motion. The aligned fibers deposit on a Teflon release film which is placed on the top of the linear platform.

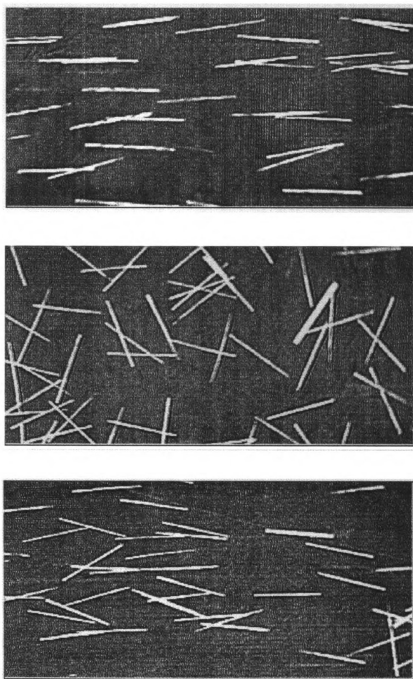


Figure 4.10 Digital images of 1" long chopped E-glass fibers settled on the deposition platform. **Left:** Aligned in E-field of 400 KV/m, **Middle:** Random with no E-field, **Right:** Manually aligned.

Table 4.1 Typical Operating Variables

Variable	Operating Range
Velocity of Deposition Platform	50 cm/min
E-Field Intensity (Top)	300 - 500 KV/m
Neutralizing E-Field Intensity (Bottom)	300 - 500 KV/m
Electrode Gap width (d)	4 - 8 cm
Electrode gap height (h)	1 - 2 cm
Fiber Feeder Amplitude	3 - 4 on dial
Oven Temperature for Sintering	200 °C
Processing Rate†	900 g/hr (2 lbs/hr)

† Estimated from semi-continuous operation data.

To demonstrate the operation of the prototype ADF process, a model composite system was chosen: chopped E-glass fiber and nylon-12 polymer matrix. Based on the fiber alignment behavior in the process and the polymer sintering temperature, the optimum operating conditions were obtained to operate the ADF process in a semi continuous mode to produce an ADF preform. The operating variables and their ranges in a typical processing situation are given in Table 4.1.

4.3.4 Fiber Orientation Distribution (FOD) Measurement

One of the objectives of developing the ADF process was to demonstrate the feasibility of making ADF composites with controllable fiber orientation distributions (FODs). The orientation distribution of the fibers is needed in the ADF process for three reasons:

- Serve as a feedback on the effectiveness of the fiber alignment process.
- Determine the effect of various parameters on the degree of fiber orientation.
- Predict the mechanical properties of the ADF composites using micromechanics models.

Typically, the fiber orientation distribution in a composite is obtained by a measurement of a statistically significant number of fiber orientations by sectioning the final composite and reconstructing the orientation of individual fibers by the aspect ratio of the ellipse that is formed. This is a very tedious technique, and numerous approximating techniques have to be used in these methods and often bias is introduced against those which are normal to the plane of observation⁷⁸. Because of the many shortcomings in the existing measuring techniques⁹, a protocol has been developed for evaluating the fiber orientation distribution in the ADF process, which can be used for both the above mentioned purposes.

The measurement step consists of three steps (Figure 4.11). Typically once the processing is done for the manufacture of an ADF composite, its FOD is obtained in an

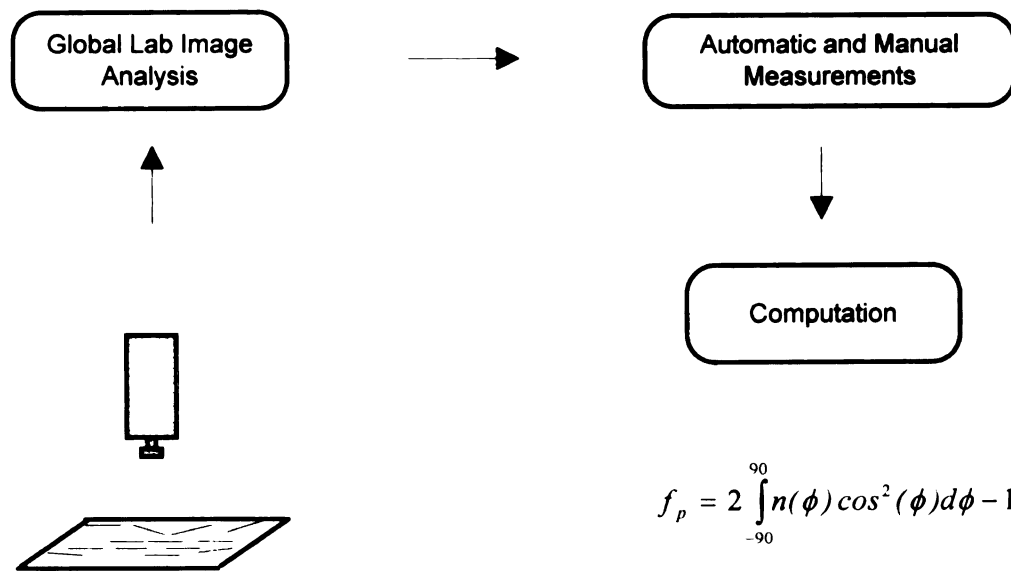


Figure 4.11 Measurement of fiber orientation distribution.

indirect way by image analysis. The method utilized in obtaining the FOD consists of running the ADF process once again under identical conditions but this time not with the intention of making a composite but recording a series of images of the fibers that are deposited on the Teflon release film using a Panasonic CCTV (Model WV 1410) camera which is connected to a computer with a frame grabber board. Recording a proper image was of utmost importance, because this is the step where bias could be introduced in this technique. For example, glass fibers shine and reflect light when incident from an angle. If the lighting is not uniform over the region of interest, it is very possible to record fibers that are oriented in a certain direction only which shine more prominently than the other fibers that are in the other directions. This kind of an artifact finally results in showing that fibers are aligned in a particular direction. The error in this kind of a biased image gets worse when the digital image is subjected to filtering operations. So before recording a series of images it was always checked to see if fibers in all the directions were being recorded.

A number of images of the fibers that are deposited on the Teflon release film are recorded to give a statistically significant FOD, which typically contains the orientations of approximately 1000 fibers. Typically, the longer the fiber length the more the number of images have to be recorded to get significant number of fibers. Global Lab image analysis software is used to treat the digital image to a few filtering operations which sharpen the images of the fibers for edge detection and also improve contrast of the fibers against the background. The “Particle Classify” routine in this software can be trained to identify fibers

and their orientations automatically. This feature reduces the time for data analysis drastically. However, in the case when there is excessive bridging or networking of fibers, this feature is not very effective and the fibers have to be individually identified to determine their orientation. Once the fiber orientations are logged, the pertinent data can be exported to any spread sheet environment like Microsoft Excel. All the statistical analysis is performed at this stage for the determination of the FOD.

The force field that the fibers encounter in the orientation chamber is depicted in Figure 3.1. If the fiber orientation at this stage needs to be described then it can be done by a three dimensional description (using ϕ and θ). So the unit vector p_i of the fiber describing the orientation in the three axes is given by:

$$\begin{aligned} p_1 &= \sin\theta\cos\phi \\ p_2 &= \sin\theta\sin\phi \\ p_3 &= \cos\theta \end{aligned} \tag{3}$$

Assuming the orientation of fibers is a even function and not biased in one direction, the fiber orientation distribution function, $N(\theta, \phi)$ may be described by the following trigonometric relationship.

$$N(\theta, \phi) = A \cos^m \theta \sin^n \phi \tag{4}$$

where A is chosen to satisfy the normalization condition of the distribution. Although, the formulation of FOD function will be similar to the one that will be used in the orientation chamber, there are certain differences in the assumptions. First, it can be safely assumed in

the case of ADF preform that the fibers are in a state of planar orientation (2D) and only ϕ is required to determine the FOD and θ is 90° . In the case of thin ADF samples (noting that length of fibers are far greater than the thickness of the sample) a digitized image of the fibers on the mat will suffice in determining the FOD. Therefore, in the case of a 2D (planar) orientation, the FOD function, $N(\phi)$ will simplify to

$$N(\phi) = A_p \cos^m \phi \quad (5)$$

The Planar Orientation Parameter f_p will be used to reflect the FOD in a given condition.

$$f_p = 2 \int_0^\pi N(\phi) \cos^m \phi \, d\phi - 1 \quad (6)$$

In the FOD measurement for composites, it has been carefully verified that the FOD of the ADF preform is not disturbed during the subsequent consolidation processing of the composite. This was verified using a few tracer fibers with known orientations before consolidation and checking their orientation state after the consolidation. Therefore, it may be concluded that the FOD obtained by this method is a true representation of the FOD in the ADF composite.

4.3.5 Aligned Vs Random

Using the above method a series of FODs were obtained for each of the chopped glass fiber mats that were produced under the two conditions: randomly oriented and aligned in an E-field of 400 KV/m cases (Figure 4.12) at the optimum operating conditions as specified in Table 4.1. As expected, the orientations of the fibers in the random case for all

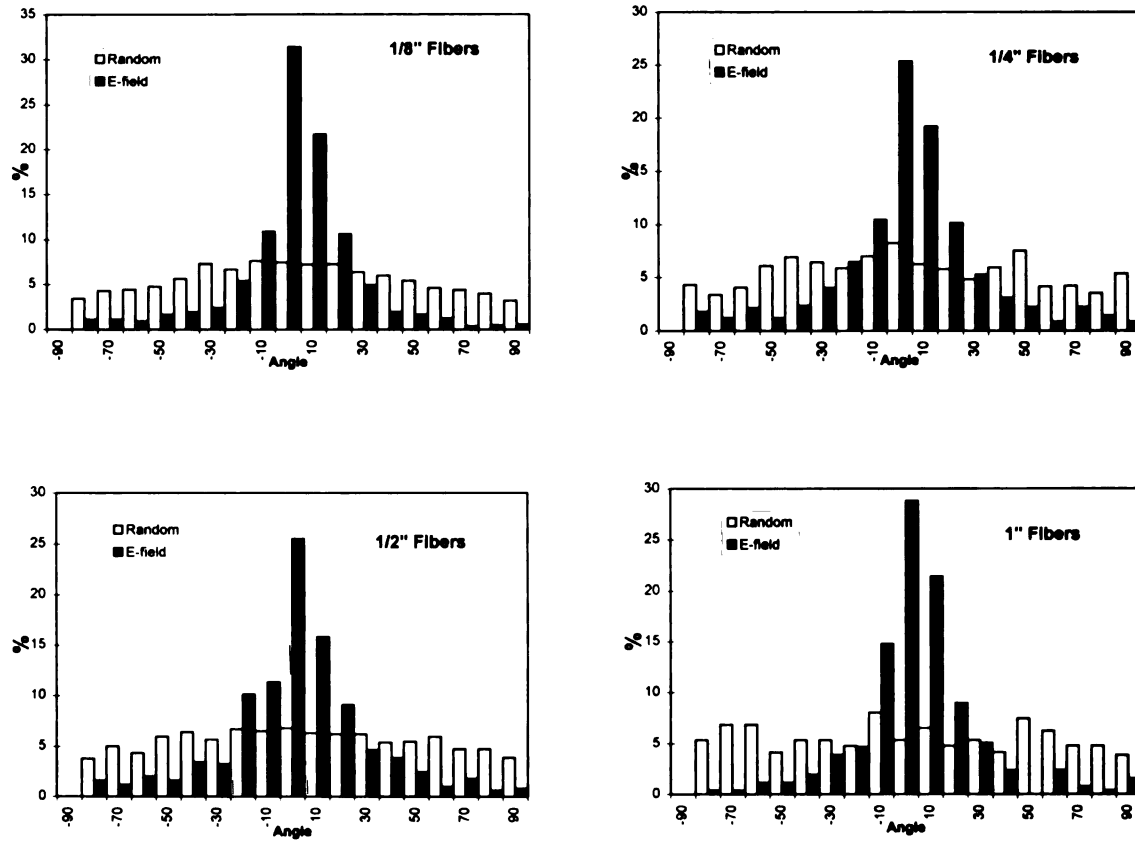


Figure 4.12 FODs of different fiber length E-glass fibers that have aligned in E-field of intensity 400 KV/m. The control case of random is also shown for comparison.

the four fiber lengths indicate a uniform spread of orientation distribution, without any significant bias in any direction. The unbiased fiber orientation distribution of the control random cases serves to establish that there is no extraneous induced alignment in the process other than that caused by the E-field. In the case of aligned fiber mats, for all the four fiber lengths, the orientations of about 70% of the fibers lie between $\pm 20^\circ$ and nearly 80 - 90% of the fibers between $\pm 30^\circ$, indicating a high degree of fiber alignment in the direction of the E-field.

4.3.6 Concentration Effects

The rate of processing is limited by the concentration effects in the orientation chamber. If the concentration of the fibers is increased from the very dilute case to a concentrated case, the probability of fibers touching each other starts to increase. As the probability of fiber interactions increase the probability of a bridging phenomenon, taking place increases in the orientation chamber. The bridging phenomenon not only causes a high degree of mis-alignment but also has the detrimental effect of shorting the fields in the case of conductive fibers. Infact, concentrated fiber suspension also reduces the effective permittivity of the suspension. An interacting concentration of fibers will result in a high probability of arcing or even subsequent breakdown in the electric field. This phenomenon of "fibration" has been looked into by Okagawa and Mason¹⁰ for electric fields, and by Fermigier and Gast¹¹ in the case of magnetic fields, for spherical and axisymmetric particles in both colloidal and non-colloidal systems. However, the size range of particles studied by these authors is much smaller compared to the dimensions of the fibers useful in composite

manufacture of this study. It is also known that beyond a certain concentration of fibers and particles, the electric field lines will be distorted, thereby lowering the efficiency of alignment.

If C_f is defined as the theoretical maximum of the non-interacting concentration of fibers, then it is related to the aspect ratio of the fiber and the fiber orientation state. The lowest C_f is obtained when the fiber orientation state is a 3-dimensional random state, implying a fiber will have freedom to rotate with respect to all the three axes. However, in the ADF process, the fibers are fed in a predominantly planar fashion using the pre-aligner (Figure 4.4). Figure 4.14 shows the “volume of influence” surrounding a fiber under the two extreme orientation states of the fibers under consideration. This intuitive analysis into the effect of orientation state on the maximum non-interacting concentration of fibers has a direct bearing on the way the fibers should be fed into the orientation chamber. It can be easily shown by the following relationships that the predominantly planar orientation state of the fibers increases C_f compared to the random state.

Random Orientation: In this case, the fibers are in a complete state of three dimensional randomness and have the freedom to assume any orientation state. A conservative estimate of the maximum concentration of the fibers that is non-interacting can be obtained by considering each fiber to have a “sphere of influence”. This sphere of influence has a diameter equal to that of the length of the fiber. Taking the volumetric ratio of the fiber to that of the “sphere of influence” of diameter equal to the length of the fiber (l) we get the relationship for C_f .

$$C_f = \frac{\frac{\pi}{4}d^2l}{\frac{1}{6}\pi l^3} = 1.5 \frac{d^2}{l^2} \quad (7)$$

Pre-aligned Orientation: The unique design of the fiber feeder plate ensures that the fibers are pre-aligned before they enter the orientation chamber. First, the fibers are in a predominantly, planar orientation state. Secondly, the fibers are also pre-aligned parallel to the slots (Figure 4.3) which makes them pack better. For the purposes of making a conservative estimate of the concentration of fibers in a given volume, it is assumed that the fiber can subtend up to an angle of 30° (see Figure 4.14). A “cylindrical volume of influence” can be visualized and calculated with a diameter equal to $l/2$ ($= 2 \times l/2 \sin 30^\circ$) and height equal to the fiber length, l .

$$C_f = \frac{\frac{\pi}{4}d^2l}{\frac{\pi}{4}\left(\frac{l}{2}\right)^2l} = 4 \frac{d^2}{l^2} \quad (8)$$

Figure 4.15 is a plot of C_f as a function of the fiber aspect ratio for the two orientation states. From these relationships, it may be seen that for a chopped fiber bundle of say an aspect ratio 100, the limiting non-interacting concentration is 0.00015 for the random case while it is 0.0004 for the pre-aligned case. Therefore it may be concluded that by the unique design of the fiber feeder, the theoretical maximum non-interacting concentration of fibers (C_f) in the orientation chamber has been increased by more than 2.5 times. This increase in the C_f value

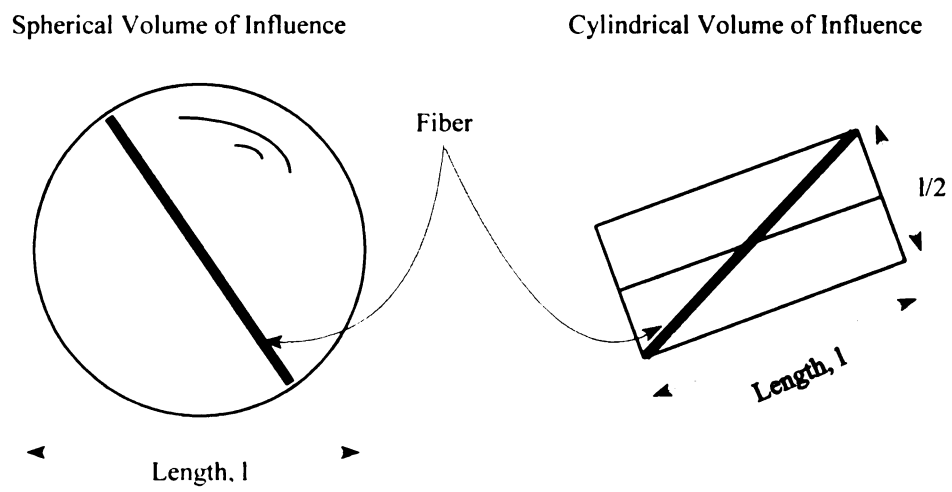


Figure 4.14 “Volume of influence” surrounding a fiber under two Different orientation states: Random and Pre-aligned.

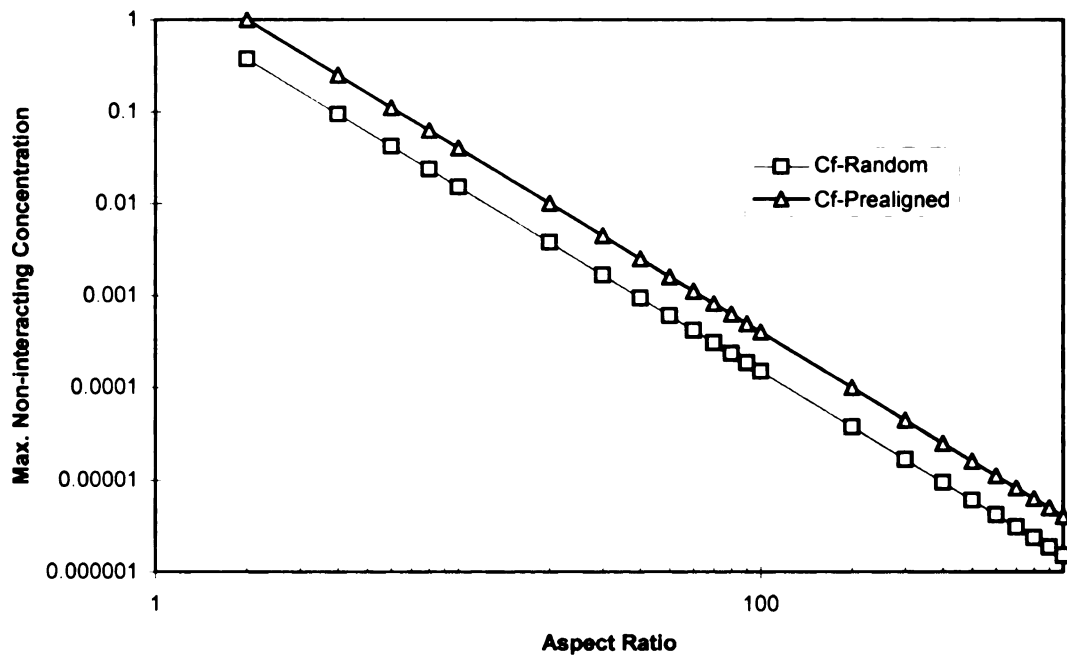


Figure 4.15 Non-interacting fiber concentration as a function of aspect ratio.

results in an increase in the production rate proportionally. However, it is also important to point out here, that this concentration effect on the electric field and its distortion is unknown. Although, production rate will be increased by this effect, it is unknown if the degree of fiber alignment will remain the same.

4.4 Process Consolidation

Consolidation is the processing step in which the ADF preform is compression molded under temperature and pressure to get a fully densified and essentially void free composite. In the thermoplastic powder coated chopped fiber preform, the flow of the polymer matrix is local and there is no bulk flow of the molten polymer because of the uniform distribution of the polymer particles on the surface of the fiber bundles (see Figure 4.9). This factor is a distinct advantage for the high melt viscosity thermoplastic polymers that are subjected to flow with low permeabilities in fibrous composite beds. This results in faster processing times and lower operating pressures in the consolidation process. A typical consolidation process cycle is shown in Figure 4.13. Consolidation primarily involves the heating of the preform from the ambient temperature (T_a); applying pressure after the temperature of the polymer reaches the processing temperature (T_p) which is typically above the melt temperature (T_m) for semi-crystalline polymers. The part is held at T_p under pressure till the flow of the polymer is complete and autohesive strength is developed. It is subsequently cooled under pressure till the temperature falls below the glass transition temperature of the polymer (T_g) to obtain a fully densified composite.

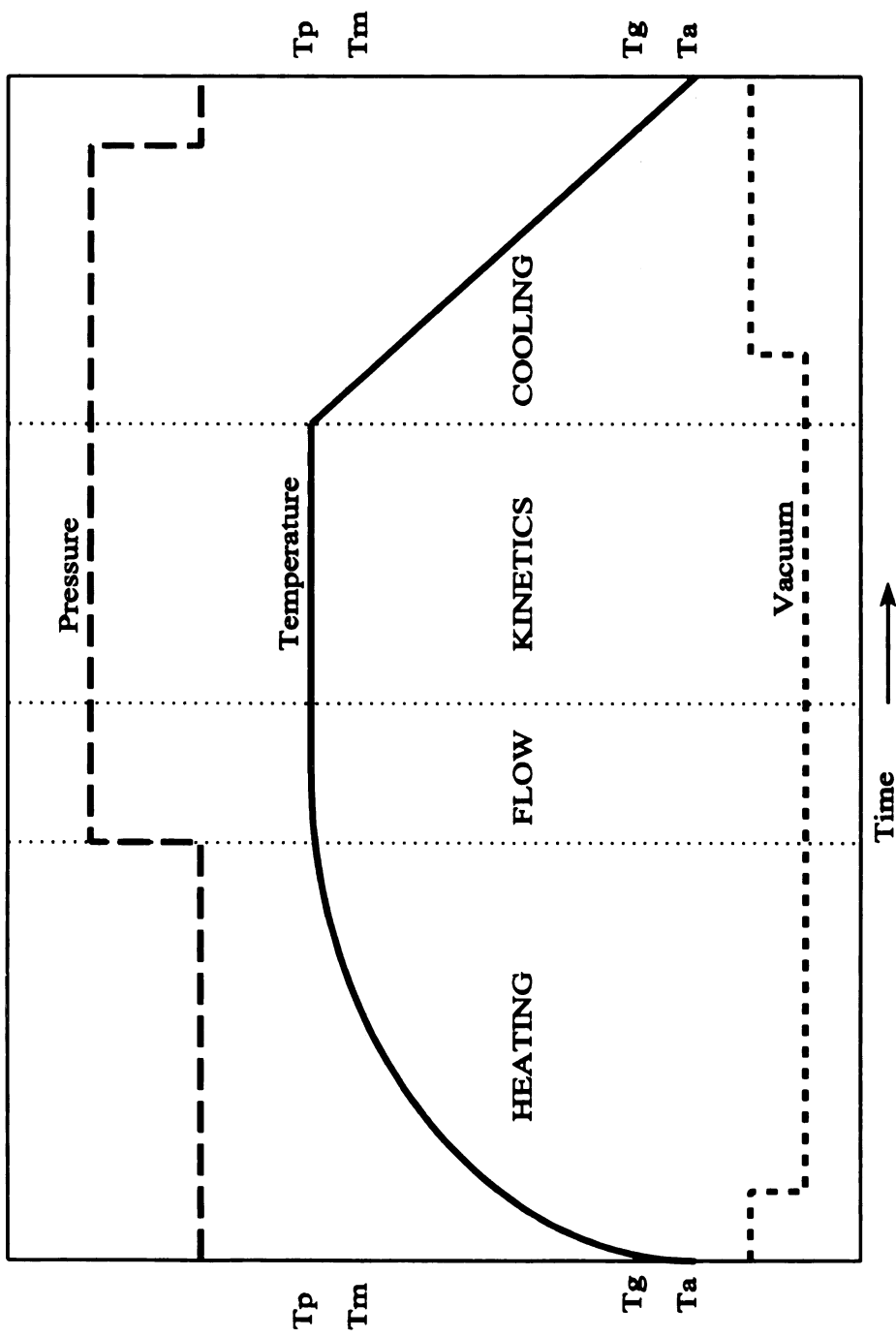


Figure 4.16 Typical consolidation process cycle of a preform with a semi-crystalline polymer matrix.

4.4.1 Process Consolidation Cycle

ADF mats that were produced in the process were carefully transferred to a compression molding caul plate, without disturbing the fiber orientation distribution. A known number of plies were stacked up depending upon the final thickness of the composite part desired. Vacuum assisted compression molding was performed in an instrumented Carver Press to make a part with minimum voids. A differential scanning calorimetry studies were conducted on the nylon-12 matrix to determine the thermal transitions and the crystallization behavior before developing the optimum consolidation cycle. It was also verified that the FOD of the ADF preform was not disturbed during the consolidation process.

Typically eight plies of the ADF preform of sizes 5" x 7" are stacked in an open mold with dams on all four sides to prevent resin flow out. The preform is heated in Carver press to 200 °C and held at that temperature for 10 minutes under 100 psi pressure. Vacuum was applied as soon as the preform temperature reached 100 °C, so that all the entrapped air as well as the moisture if any, will be removed from the system. Vacuum is pulled off during the cooling stage after the temperature drops below the melting point of the polymer, i.e 175 °C. The cooling is controlled by the water flow rate and a steady cooling rate is maintained. Pressure is maintained till the temperature of the resin drops below T_g.

4.4.2 Consolidation Pressure

Recently, a consolidation model has been developed for continuous fiber powder preregs by Padaki¹², incorporating the various phenomena that occur during the

consolidation process. The results from this study are used to arrive at some of the processing parameters for consolidating ADF preforms. Though several of the features of this model are applicable for powder coated ADF preforms, caution was exercised in adopting the results for ADF preforms in view of some differences in the material form.

The consolidation process is most commonly treated as a load distribution problem where the pressure applied to the composite preform (p) is balanced by the average resin pressure (P_r) and the effective stress in the fiber network (σ). The resin pressure is estimated by modeling a squeezing flow of a particle between two flat surfaces. The force, F that is needed to squeeze a particle from an initial height, h_o to a final height, h in time, t is given by:

$$F = \frac{8\eta a^4}{t} \left(\frac{1}{h^2} - \frac{1}{h_o^2} \right) \quad (9)$$

Where, a can be a characteristic dimension of the particle which is equal to h_o . The final height, h is determined to be the inter-fiber distance in a fully densified composite of a given volume fraction.

The average effective stress (σ) in the fiber network has been modeled by Gutoswski¹³, as a deformation of the fiber network that occurs during any consolidation process.

$$\sigma = A_s \frac{\sqrt{\frac{V_f}{V_o}} - 1}{\left(\sqrt{\frac{V_a}{V_f}} - 1 \right)^4} \quad (10)$$

V_f is the varying fiber volume fraction of the lay-up that is being consolidated from an initial volume fraction of V_o and V_a is the maximum possible volume fraction that is possible for the fiber architecture. A_s is called the “spring constant” which is a function of the fiber bending stiffness (E) and a geometric parameter, β . β is typically the span length to span height ratio for the fiber beam network.

$$A_s = 3\pi \frac{E}{\beta^4} \quad (11)$$

A_s has been determined for AS4 carbon fibers ($E = 34 \times 10^6$ psi) to be 0.125 resulting in $\beta = 225$. β value for chopped glass fibers is not known, but certainly will be much smaller than continuous fiber unidirectional preforms, and will also be dependent on the fiber orientation.

This model describes the phenomenon when an initial load is applied it is carried by the polymer matrix. As the polymer is squeezed the volume fraction of the fiber network increases and the stiffness of the fiber network increases. It is to be noted here that the

typical volume fraction of discontinuous fiber composites is less than 50 % while for the continuous fiber composites it is over 50%, indicating substantially lower pressures are needed in consolidating ADF preforms.

4.4.3 Void Fraction

Each panel that is consolidated is tested for volume fraction and void fraction using a burn off test. At two small pieces of the composite weighing approximately 1 gm. are cut from random locations of the panel and the actual volume is measured by water displacement. These samples are then burnt off in a muffle furnace at 600 °C for 3 hrs and the weight of the fibers remaining is measured. The volume fraction of fibers and the void content is then determined (see Table 5.2).

4.5 Manufacturing

The focus of this chapter so far has been on the development of the ADF process - its design, operation and performance. Now the issues involved in the making of ADF preforms is discussed. First, the processing rates for the prototype process are derived and later the high speed methodology of the ADF process and the continuous manufacturing schemes are taken up.

4.5.1 Processing Rates

The overall processing rate in terms of mass of ADF preform produced per time (P) is a function of the mass flux in the orientation chamber that is settling on the deposition

platform per unit time and the dimensions of the orientation chamber. If the fiber settling velocity is S_t , the critical concentration of the fibers in the orientation chamber is C_f (vol/vol), the area of cross section of the orientation chamber is A_{oc} , and the density of the fibers is ρ , then P is given by the following relationship.

$$P = \rho C_f S_t A_{oc} \quad (12)$$

If the fiber feed rate is precisely known then that value can be used in arriving at C_f by a simple mass balance. The rate at which the thickness (T) of the ADF preform grows under batch mode is given by:

$$\frac{dT}{dt} = \frac{C_f S_t}{A_{oc}} \quad (13)$$

The thickness of the ADF preform (T) that is formed when the deposition platform is set in motion at a velocity, V is given by:

$$T = \frac{C_f S_t A_{oc}}{V W_{oc}} \quad (14)$$

Equations 11 and 12 are useful in predicting the thickness of the ADF preform that is formed under the “batch” and “continuous” modes respectively.

The velocity of the deposition platform (V) plays a direct role in the rate of processing. However, the velocity of the platform cannot be increased without consideration to its effect on the fiber orientation distribution and the secondary operations like the sintering times for the polymer particles. With the electrode modifications that incorporated a neutralizing field, it was observed that at the velocities of interest (around 50 cm/min) the deposition platform velocity did not have any discernable effect on the fiber orientation distribution. It can be expected however, at much higher velocities, air currents would introduce mis-alignments. The velocity of the platform/belt will be a function of the downstream processing like polymer particle sintering and heat transfer times needed when continuous operation of the ADF composite sheet is the product.

The principle of ADF processing has been established for the case where there is only one orientation chamber. If high production rates are needed, it can be easily seen that it can be achieved by using multiple number of orientation chambers in series. So the scale up factors for ADF process are:

- area of cross section of the orientation chamber (A_{oc})
- number of orientation chambers (N_{oc}).

The production rate (P) in the case of a multiple orientation cell system will be given by the following relationship (see also § 4.5.1):

$$P = N_{oc} \rho C_f S_t A_{oc} \quad (15)$$

4.5.2 High Speed Manufacturing Schemes

The ADF processing methodology offers a solution to the problem of reducing the cycle time in the fabrication of an aligned discontinuous structural composite and lends itself for a highly automated process, due to the following three reasons:

- (a) a simple and rapid orientation technique using electric fields;
- (b) controlled powder impregnation of the matrix gives immense flexibility in the fiber volume fractions that can be obtained and eliminates the additional step of resin transfer molding as the case would have been if a preform is the final product of the process; and
- © by incorporating a technique to increase the concentration of fibers that can be processed in the orientation chamber, thereby increasing the production rate.

As pointed out in the beginning of this chapter, the concept of ADF processing can be applied in a variety of ways. Just the fiber alignment itself can be incorporated in any continuous preform or composite sheet manufacturing process. The incorporation of the polymer powder provides still greater benefits in terms of high speeds of solvent free processing. Figure 4.16 shows the schematic of a continuous ADF process that can be conceived based on the principles developed in this study. Alternatively, a moving veil with a layer of the polymer matrix film can be passed through the orientation chamber. The

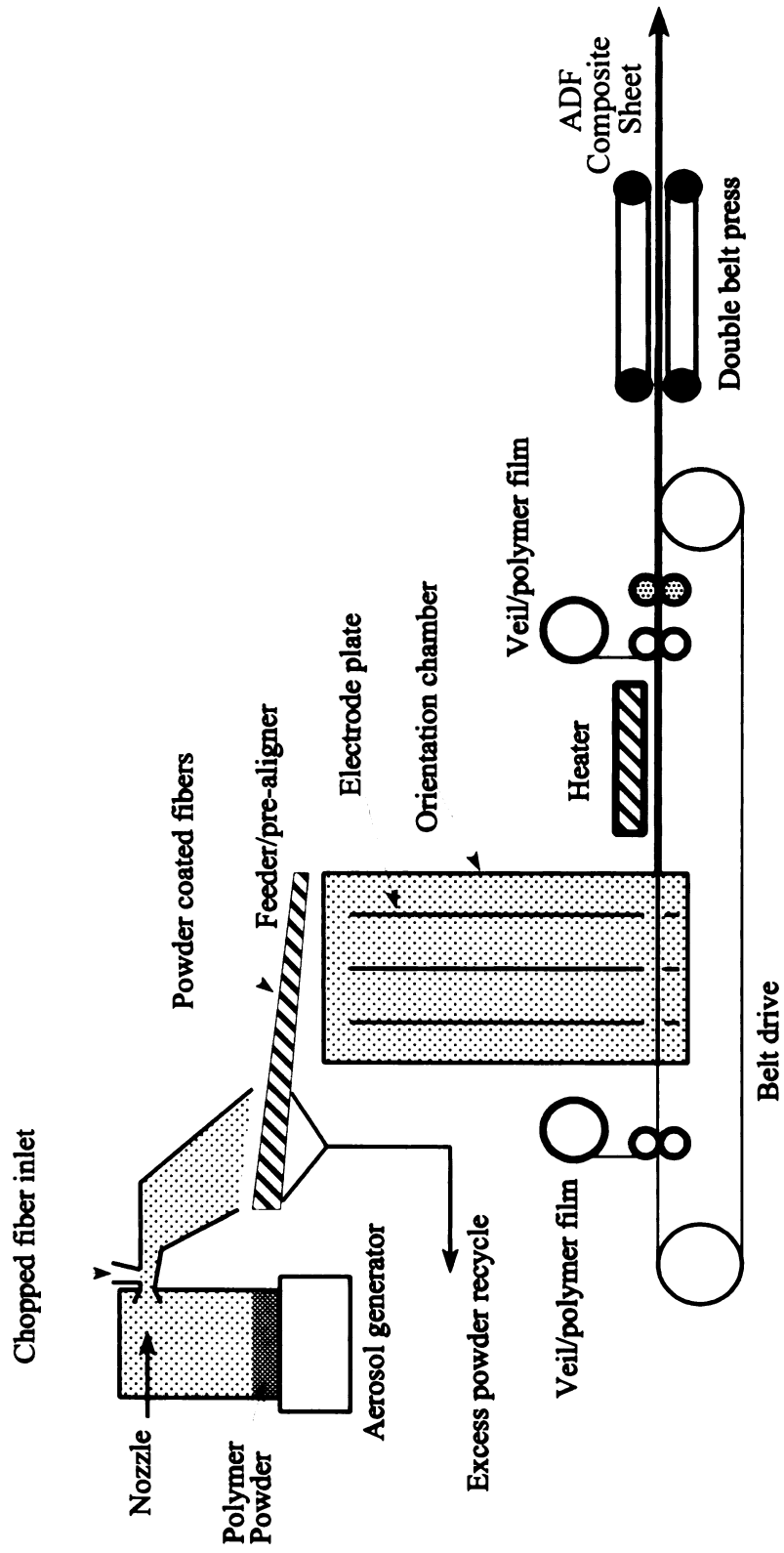


Figure 4.17 Schematic of proposed continuous ADF process incorporating polymer powder coating; Fiber alignment using electric fields; Continuous ADF composite sheet manufacture.

deposited fibers can then be covered with another layer of the polymer film and heat treated to form a handleable mat or laminate with the desired fiber orientation. The thickness of such a material can be built by a series of orientation chambers and later compression molded, typical of a sheet lamination process to form a composite sheet. The consolidation cycle needed to process these composites will be the same as described above for the same polymer matrix.

4.6 Summary

A semi-continuous prototype process has been developed that can manufacture aligned discontinuous fiber composites using electric fields. The key steps in the process are: fiber alignment in air using electric fields; powder coating/impregnation of fibers and; consolidation of the powder coated ADF preform into a composite. The ADF process has been demonstrated for E-glass fiber and nylon12 matrix system and can be easily extended to many other fiber-matrix combinations.

The design of the orientation chamber for controlling fiber orientation in air has two unique features that counter the edge effects of the electric fields when the deposition platform is set in motion during the continuous processing of ADF preforms.

- Create an equal and opposite field under the high voltage electrode by placing another electrode under the moving slide/mat/veil. This neutralizes the edge effects of the high voltage electrode, which otherwise disturbs the fiber

orientation of the fibers that are aligned by the E-field and that are deposited on the mat.

- Designed the edges of the electrodes which are closer to the moving mat with a conductive cap which has a cylindrical cross-section with a relatively large radius of curvature. The cylindrical edge reduced the effective E-field intensity at the edge, thereby reducing the edge effects and minimizing E-field breakdowns.

Developed a unique vibratory fiber feeder and pre-aligner. Fibers with or without powder impregnation can be fed into the orientation chamber without entanglement. Fibers falling from the feeder pre-aligner plate tend to fall with a planar orientation state instead of a three dimensional random state which makes them align faster in the direction of the electric field. This feeding mechanism also has the potential to increase the theoretical critical processing concentration of fibers that can be used in the orientation chamber.

The fiber orientation distribution in the ADF preform as well as the orientation chamber was obtained using digital image analysis techniques. Under the prototype ADF processing conditions, for all the E-glass fiber lengths nearly 70 % of the fibers of the fibers were aligned $\pm 20^\circ$ and nearly 80 - 90 % within $\pm 30^\circ$, indicating a high degree of finer alignments using electric fields. Further, improvements in alignments can be made based on the understanding gained on the alignment behavior of fibers in air.

Polymer powder coating of the fibers and thereby controlling the matrix volume fraction in the final composite is one of the biggest advantages of the ADF process. The combination of powder coating with electric field alignment makes the process a potentially high-speed, low-cost process to make aligned discontinuous fiber composites. This step eliminates the additional step of resin transfer. The technology of powder coating is well established in the group and extensively patented and hence can be easily incorporated in the ADF process developed.

The prototype process in its current configuration has a rated capacity of manufacturing 2 lbs/hr of ADF preform. Conventional scale up factors i.e. increasing the width of the orientation chamber from the existing 20 cm to the desired width and using multiple orientation chambers to build up ADF preform rapidly result in direct increases in production rates. These scale-up factors can be directly applied to the process without affecting the principle of operation in any way.

References

1. Drzal, L. T., Padaki, S., Vyakarnam, M. N., Fernandes, J. H., "Powder Impregnation of Advanced Composite Materials", in *Polymer Powder Technology*, Ed.. Narkis, M. and Rosenzweig, N., John Wiley & Sons Ltd., pp 511-530, 1995.
2. Iyer, S. R., "*Continuous Processing of Unidirectional Prepreg*", Ph.D. Dissertation, Michigan State University, 1990.
3. Vyakarnam, M. N., "*Development of a High Speed Powder Process to Manufacture Composite Prepreg*", M.S. Thesis, Michigan State University, 1992.
4. Iyer, S. R. and Drzal, L. T., *Powder Technology*, 57(2), p 127-133, 1989.
5. Vyakarnam and Drzal, US. Patent 5,310, 538, 1994.
6. Padaki, S. and Drzal, L. T., "Development of a Process and Consolidation Model for Powder Prepreg Composites", *Proc. 10th. Annual ASM/ESD Advanced Composites Conference & Exposition*, Dearborn, 1994.
7. Fisher, G. And Eyerer, P., "Measuring Spatial Orientation of Short Fiber reinforced Thermoplastics by Image Analysis", *Polymer Composites*, Vol. 9, No. 4, p 297-307, 1988.
8. O'Connell, P. A. And Duckett, R. A., *Comp. Sci. & Tech.*, Vol . 42, p 329-347, 1991.
9. Liang, E. W., Poslinski, A, J. and Stokes, V. K., "Fiber Orientation in Discontinuous Fiber Composites: An Overview, General Electric Report No. 93CRD136, 1993.
10. Okagawa, A. and Mason, S. G., *J. Colloid Interface Sci.*, Vol 47, p 568, 1974.
11. Fermigier, M. and Gast, A. P., "Structure Evolution in a Paramagnetic Latex Suspension", *Journal of Colloid and Interface Science*, Vol 154, No. 2, 1992.
12. Padaki, S., "A Process and Consolidation Model for Polymer Powder Impregnated Composite Tapes", Ph.D. Dissertation, Michigan State University, 1995.
13. Gutowski, T. G., Morigaki, T., and Cai, Z., "Consolidation of Laminate Composites", *Journal of Composite Materials*, Vol. 21, pp. 172-188, 1987.

Microstructure-Property Relationships

5.1	Introduction	143
5.1.1	Microstructure-Property Relationship Flowchart	145
5.2	Microstructure Descriptors	147
5.2.1	Fiber Orientation Distribution	150
5.2.2	Fiber Aspect Ratio	152
5.2.3	Fiber-Matrix Interaction	154
5.3	Elastic Property Predictions	156
5.4	Reinforcement Models	160
5.4.1	Shear Lag Theory	161
5.4.2	Eshelby's Inclusion Approach	164
5.4.3	Halpin-Tsai's Relationships	166
5.5	Experimental Results	169
5.5.1	Sample Preparation and Tensile Testing	169
5.5.2	Effect of Fiber Alignment	169
5.5.3	Effect of Fiber Length	170
5.6	Model Predictions and Discussion	170
	References	177

5.1 Introduction

Discontinuous or short fiber composites pose a rich array of microstructural combinations that often makes them very attractive for such a wide range of applications. However, it is also this variation in microstructural features that makes it difficult to describe the thermo-elastic behavior of these materials using simple models¹. These materials can be anything from fiber reinforced injection molded polymer composites to whisker reinforced polymer/metal matrix composites or chopped glass fiber Sheet Molding Compound (SMC) composites where the mechanical performance of these materials is enhanced by the presence of fibers. In a majority of the processes that are used to fabricate discontinuous fiber composites, there is generally little or no control on the fiber orientation distribution (FOD) during the processing stage, and more often than not the FOD of these composites is not known, which makes the prediction of properties unreliable. The development of the Aligned Discontinuous Fiber (ADF) composite process using electric fields, has the potential to control the orientation state of fibers during the processing stage of these composites. The ability to fabricate discontinuous fiber composites with microstructure that is known a priori provides a unique opportunity to both investigate the fundamental microstructure-property relationships and evaluate the ability of the ADF composites. This study has two objectives.

- First it serves to validate the guiding principle that motivated the development of the ADF processing method i.e. the elastic performance of discontinuous fiber composites approach that of continuous fiber composites when the length of the fibers are greater than critical fiber length and when they are aligned in the direction

of the stress. Some of the questions that are addressed in this study are: What reinforcement architecture is important in chopped fiber-thermoplastic systems? Is it the individual fiber aspect ratio or the bundle (aggregate) aspect ratio?.

- Secondly, this study will provide insight into the elastic behavior of the chopped fiber - thermoplastic systems. This class of composites is gaining popularity for semi-structural applications in the automotive and durable goods industry because of their improved energy absorption characteristics; environmental friendliness due to recyclability, repairability and solvent free processing; and finally due to high speed manufacturing. Most of the composite reinforcement theories and their experimental verification have focussed on either continuous or single fiber cases in thermosetting resins due to their extensive usage in the composites materials area so far. The effect of the aggregate (bundle) nature of the fibers on mechanical properties of discontinuous fiber reinforced composites is not well understood^{1,2,3}. Despite, the extensive use of discontinuous fiber composites and the extensive data on properties, very few studies exist, with the exception of Kacir et. al⁴ and Piggott et. al⁵. that combine theoretical property predictions with accurate experimental validation.

Interestingly, the aspect ratio of fibers which played such a significant role in the alignment behavior in electric fields also seems to be a crucial factor in the reinforcement behavior. So, it was all the more logical to investigate the microstructure-property

relationships of discontinuous fiber composites and arrive at conclusions that will have a direct bearing on both the processing and performance of discontinuous fiber composites.

5.1.1 Microstructure-Property Relationship Flowchart

In this chapter, composite reinforcement theories are analyzed and modeled for the discontinuous fiber composites, to predict the modulus of these composites as a function of the fiber orientation distribution, fiber aspect ratio and the fiber-matrix interaction. These predictions are then compared with experimentally determined tensile properties. The overall approach in modeling the microstructure-property relationship is depicted in the flowchart shown in Figure 5.1. Computations are performed with input from the fiber-matrix constituent properties and the fiber volume fraction. The microstructure descriptors: fiber orientation distribution (FOD) and fiber aspect ratio are individually evaluated and also become inputs to the reinforcement models. The fiber orientation distribution in the composite is represented by a statistical distribution function. Modified shear lag, modified Eshelby's elastic solution and Halpin-Tsai models are analyzed and evaluated for the effect of finite fiber aspect ratio on the modulus of perfectly aligned discontinuous fiber composites. The properties from the material direction can be transformed to the three principal directions by invoking the transformation matrix. Property transformation is integrated over the thickness of the composite using the fiber orientation distribution (FOD) function. The computations in the preceding steps leads to the prediction of the elastic properties of the ADF composites. The computed values are then compared with

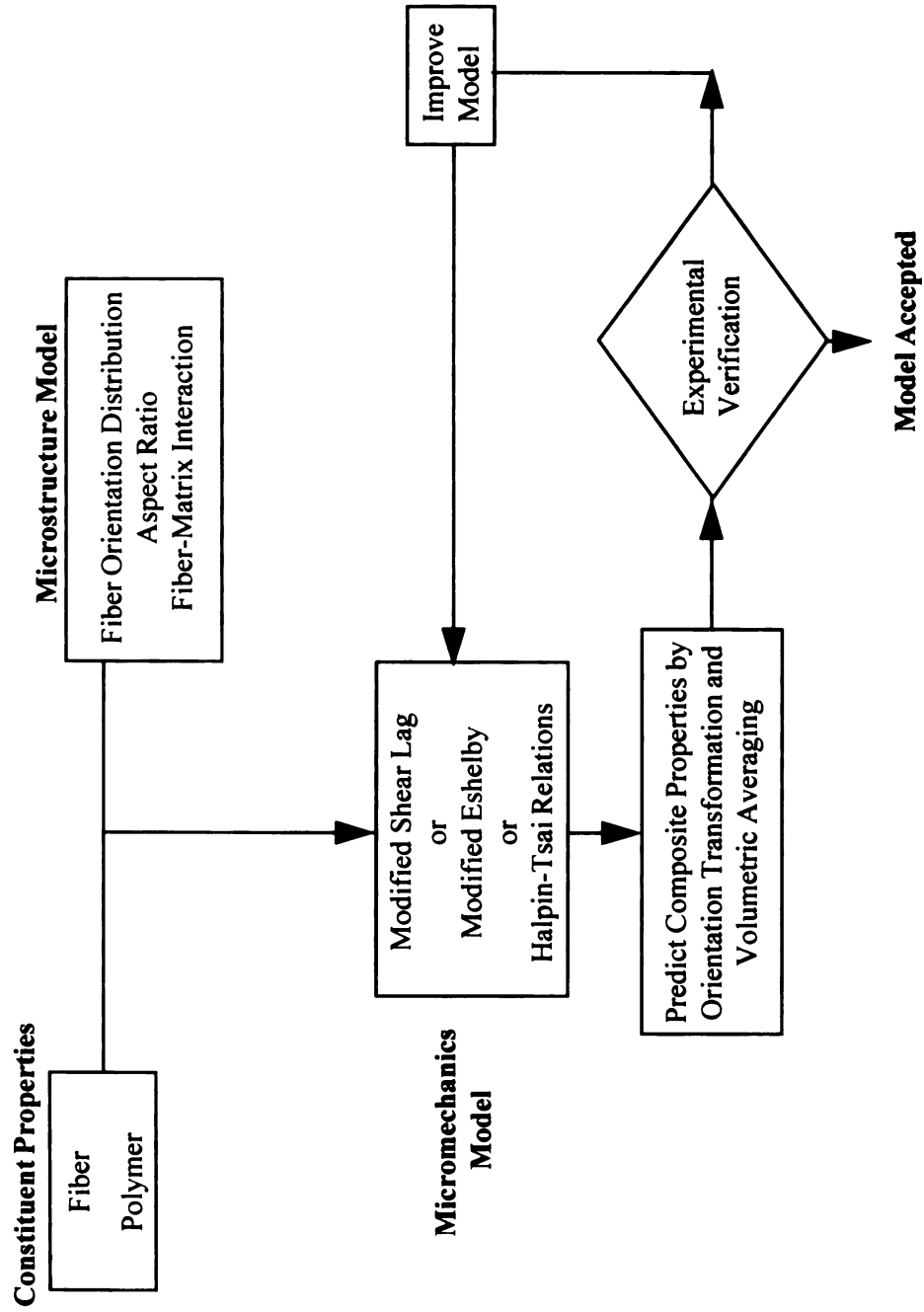


Figure 5.1 Microstructure-property relationship flowchart.

experimentally determined values. A comparison between the experimental results and the model predictions will be presented at the end of the chapter.

It should be noted that the term “ADF composite” used in this chapter refers to any discontinuous fiber composite that is made using the ADF process described in Chapter 4. For micromechanics purposes, these composites will be further qualified by the terms “Aligned” and “Random” representing the cases when ADF composites were made using the E-fields to align the fibers and without using the E-field respectively. Further, the Aligned ADF composites should not be considered as perfectly aligned discontinuous fiber composites. In fact they have some misalignment of fibers and the true fiber orientations are described by the FOD.

5.2 Microstructure Descriptors

Three principal features of the microstructure that dictate the final mechanical properties of discontinuous fiber-matrix composite system for a given fiber volume fraction, are: (a) fiber orientation distribution (b) fiber aspect ratio and © fiber-matrix interaction. The term fiber-matrix interaction includes the effect of fiber sizing and the interfacial adhesion and also the fiber wet-out or resin impregnation inside the fiber bundle in case of aggregate reinforcement. In fact, the first two factors play a major role in the modulus predictions compared to the third factor, which becomes critical in the case of strength and fracture behavior⁶. The effect of fiber volume fraction on discontinuous fiber composites can be

easily extrapolated once the effect of the microstructural features are well understood. Modulus and strength values tend to be linear functions of the fiber volume fraction.

To keep the experimental part of the study focussed to the influence of FOD and the aspect ratio, the fiber volume fraction is fixed at 40 % and the same constituents; E glass fibers and nylon-12 matrix, whose properties are shown in Table 5.1 are used throughout the study. Each of the test panels that were fabricated were verified for fiber volume fraction using a burn out test. At least two samples were tested from each panel and the fiber volume fraction was found to be around 40 % with a $\pm 2\%$ variation (Table 5.2). The burn out test also gave the void content in the composite sample. It was typically around 2 to 3 % or slightly higher in a few cases.

The void content seemed to be on the higher side when compared with good continuous fiber composites which are typically under 1 %, but this value seemed reasonable for compression molded chopped fiber thermoplastic composites because of the difficulties in matrix impregnation into the chopped fiber bundles. Studies^{7,8} relating the effect of void content on the reduction in mechanical properties, indicate that the void content does not have a significant effect on the modulus values as long as it is typically 3 % and under. However, strength values could be more influenced by the presence of voids. Optimization of processing conditions should further reduce the void content.

Table 5.1 Constituent Properties

Property	E-Glass	Nylon-12
Tensile modulus, E_{11} (Msi)	10.5	0.19
Shear Modulus, G_{12} (Msi)	4.3	0.07
Poisson Ratio, ν_{12}	0.22	0.33

Table 5.2 Volume Fraction and Void Fraction of ADF Composites

Fiber length (in)	Orientation	Fiber Volume Fraction (%)	Void fraction (%)
0.125	Random	41.3	2.6
0.25	Random	41.7	3.0
0.5	Random	42.5	2.9
1.0	Random	38.0	3.5
0.125	Aligned	41.9	3.2
0.25	Aligned	40.7	2.7
0.5	Aligned	44.6	2.7
1.0	Aligned	42.5	4.3

5.2.1 Fiber Orientation Distribution

One of the objectives of developing the ADF process was to demonstrate the feasibility of making ADF composites with controllable fiber orientation distributions (FODs). Typically once the processing is done for the manufacture of an ADF composite, its FOD is obtained in an indirect way by image analysis. The method adopted in obtaining the FOD consists of running the ADF process once again under identical conditions but this time not with the intention of making a composite but recording a statistically significant number of images using a Panasonic CCTV. A number of images of the fibers that are deposited on the Teflon release film are taken to give a statistically significant FOD. The Global Lab image analysis software is used to process the digital image, identify fibers and determine their orientations in each image. It has also been carefully verified that the FOD of the ADF mat is not disturbed during the subsequent processing of the composite. Therefore, it may be concluded that the FOD obtained by this method is a true representation of the FOD in the ADF composites (Table 5.3).

The general three dimensional orientation state of short fiber in a composite can be described by the angle θ and ϕ . The transformation matrix from the original coordinate system of the fiber to that of the three principal directions can be obtained by T (3D) as defined below.

$$T(3D) = \begin{bmatrix} \sin\theta\cos\phi & \sin\theta\sin\phi & \cos\theta \\ \cos\theta\cos\phi & \cos\theta\sin\phi & -\sin\theta \\ -\sin\phi & \cos\phi & 0 \end{bmatrix} \quad (1)$$

Table 5.3 Measured Fiber Orientation Distributions of "Aligned" and "Random" ADF Composites.

Fiber Length Angle (+/- 5)	1/8"		1/4"		1/2"		1"	
	Random (%)	Aligned (%)	Random (%)	Aligned (%)	Random (%)	Aligned (%)	Random (%)	Aligned (%)
-85	3.43	1.10	4.29	1.80	3.77	1.62	5.36	0.39
-75	4.29	1.10	3.37	1.16	5.00	1.21	6.85	0.39
-65	4.42	0.91	4.06	2.11	4.34	2.02	6.85	1.17
-55	4.75	1.65	6.13	1.16	5.94	1.62	4.17	1.17
-45	5.62	1.92	6.90	2.32	6.42	3.43	5.36	1.95
-35	7.31	2.38	6.44	4.01	5.66	3.23	5.36	3.89
-25	6.69	5.39	5.90	6.44	6.70	10.10	4.76	4.67
-15	7.60	10.88	6.97	10.45	6.51	11.31	8.04	14.79
-5	7.43	31.35	8.28	25.34	6.79	25.45	5.36	28.79
5	7.23	21.66	6.28	19.22	6.32	15.76	6.55	21.40
15	7.27	10.60	5.82	10.14	6.23	9.09	4.76	8.95
25	6.40	4.94	4.83	5.28	6.23	4.65	5.36	5.06
35	5.99	1.92	5.98	3.06	5.38	3.84	4.17	2.33
45	5.41	1.65	7.51	2.22	5.47	2.42	7.44	0.00
55	4.62	1.19	4.14	0.84	5.94	1.01	6.25	2.33
65	4.38	0.37	4.21	2.22	4.72	1.82	4.76	0.78
75	4.00	0.46	3.52	1.37	4.72	0.61	4.76	0.39
85	3.18	0.55	5.36	0.84	3.87	0.81	3.87	1.56
fp	0.18	0.74	0.12	0.63	0.10	0.63	0.02	0.73

For planar orientation, the elements of the rotational transformation T (2D) simplify to the following form.

$$T(2D) = \begin{bmatrix} \cos\phi & \sin\phi & 0 \\ -\sin\phi & \cos\phi & 0 \\ 0 & 0 & 1 \end{bmatrix} \quad (2)$$

It is pertinent to point out here that the fibers in the ADF composites lie in a planar state and hence the two dimensional transformation matrix (2D) will suffice. This assumption is valid when the length of the fibers is greater than the thickness of the composite. For a planar (2D) orientation of fibers, the orientation state can be characterized by a single parameter f_p , which is a trigonometric average of the distribution obtained from the above transformation, when the FOD is symmetrical.

$$f_p = 2\langle \cos^2\phi \rangle - 1 \quad (3)$$

Table 5.3 gives the FODs and the f_p values of the ADF composites that were fabricated.

5.2.2 Fiber Aspect Ratio

A statistically significant number of fiber lengths and their weights were measured for each type of fiber that was supplied and used for making ADF composites. The fiber length distribution was found to be very narrow in each case and hence the nominal length was used in all subsequent computations. The mean number of filaments per bundle were determined from the weight and length measurements of typically 100 fiber bundles of each

length and type. It was found that there was a variation in the number of filaments per bundle and was not as sharp a frequency distribution as was the case with fiber length. The fiber length and the mean number of filaments in each type of fiber bundle that was investigated in this study is given in Table 5.4. In the chopped glass fiber systems, the reinforcement aspect ratio is unknown. It is not clear if it is just the ratio of bundle length to the bundle diameter or if it is the ratio of the length of the bundle to the diameter of individual filament. It is postulated that it will depend on the degree of impregnation into the fiber bundle. Good impregnation is achieved when there is a good wet out between the fiber and the matrix and when an optimum time-temperature-pressure consolidation cycle is used. It is also felt that good thermoplastic resin impregnation into the fiber bundles can be achieved only when some shearing action. The compatibility of the sizing and the resin impregnation was investigated for the polyester sized and the nylon sized cases as shown in the micrographs (Figures 5.2 and 5.3). The fiber bundles distinctly remain as bundles in the ADF composite fabricated with polyester sized fibers due to poor wet out (Figure 5.2), while in the case of nylon sized fibers there is a good dispersion of the fibers and the nylon12 matrix has impregnated the individual fiber bundle (Figure 5.3).

As explained in § 3.3.3.1, the aspect ratio of the chopped fiber transformed into an equivalent fiber bundle aspect ratio.

$$a_e = \frac{d_{be}}{d_f} = \sqrt{\frac{4}{\pi} N_{bf}} \quad (4)$$

Table 5.4 Fiber Bundle Length and Aspect Ratios

Nominal Length (mm)	Filament Aspect ratio	Effective Bundle Ratio
3.2	246	14.5
6.3	485	28.5
12.7	977	57.5
25.4	1985	114.9

Note: Supplied by Owens Corning with polyester compatible sizing; average filament diameter - 13 μm and the average effective bundle diameter is 221 μm based on a square packing of an average number of 227 filaments per bundle.

In the event of an aspect ratio distribution, the root mean square of the effective aspect ratio (a_e) is to be taken:

$$a = \sqrt{\langle a_e^2 \rangle} \quad (5)$$

$$\langle a_e^2 \rangle = \int a_e^2 p(a) da$$

The chopped fibers used in the ADF process had a very tight distribution in lengths. The number of filaments that were present in the fiber bundle were also consistent. Therefore, it was assumed that there was no significant distribution in the fiber length and hence, mean fiber lengths are used in the determination of aspect ratios.

5.2.3 Fiber-Matrix Interaction

These fiber-matrix interactions are quite complicated to describe in simple quantitative terms and hence in the present study they will be dealt in qualitative terms. Two extremes cases are generated by choosing two sizing chemistries. The first one is a polyester

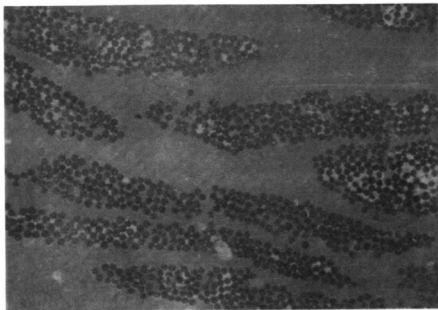


Figure 5.2 "Bundle" reinforcement effect due to incompatible fiber-matrix sizing.

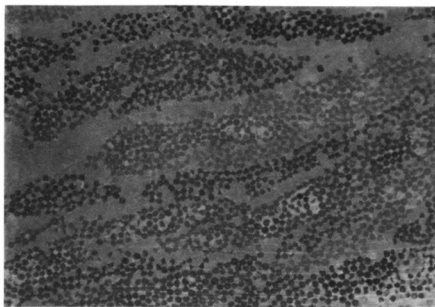


Figure 5.3 "Filament" reinforcement effect due to compatible fiber-matrix sizing.

compatible sizing which is certainly not the optimum one for nylon 12 matrix and the other one is a nylon compatible sizing. The intent is not to relate the performance of discontinuous fiber composites with different adhesion levels as was done in a very comprehensive study for continuous fiber composites by Madhukar and Drzal⁶, but to make observations on the effect of sizing chemistry on the modulus and strength values. Microscopic examination of the composites made from these different sizings indicated a more “aggregate” or “bundle” phenomenon in the polyester compatible sizing (Figure 5.2) and the fibers are more “dispersed” in the nylon compatible case (Figure 5.3).

5.3 Elastic Property Predictions

The generalized Hooke’s law for the stress (σ)-strain (ϵ) relationship for any material is given by:

$$\begin{aligned}\sigma_i &= C_{ij} \epsilon_j \\ \epsilon_i &= S_{ij} \sigma_j\end{aligned}\tag{6}$$

Where, C_{ij} is the stiffness matrix and S_{ij} is the compliance matrix and $[S] = [C]^{-1}$. Fiber reinforced composite materials are usually highly anisotropic in nature and may require up to 21 elastic constants instead of the 2 elastic constants that are required for isotropic materials to relate stress and strain in the body. The engineering properties like tensile modulus, shear modulus and poisson’s ratio in the three principal directions are obtained from the compliance matrix, $[C]$.

The problem of the elastic behavior of discontinuous fiber composites can be simplified by considering it to be equivalent to a series of orthotropic laminates of perfectly aligned discontinuous fiber composite plies stacked up in a series with different fiber orientations (Figure 5.4). The sequence of fiber orientation will be a weighted function of the fiber orientation distribution in the composite. By invoking the laminate approach, the implicit assumption is that plane stress conditions apply as defined below.

$$\sigma_3 = \tau_{23} = \tau_{31} = 0 \quad (7)$$

The assumption of planar orientation holds good as long as it is known that there is no tilting of the fibers “out of plane” and when the length of the fiber is sufficiently longer than the thickness of the composite. Both of these conditions are satisfied in the ADF composites that are fabricated in this process. This results in further reduction in the number of elastic constants that are needed to characterize the elastic behavior in the plane to five, as indicated in the $\epsilon - \sigma$ relationship below.

$$\begin{bmatrix} \epsilon_1 \\ \epsilon_2 \\ \gamma_{12} \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} & 0 \\ S_{12} & S_{22} & 0 \\ 0 & 0 & S_{66} \end{bmatrix} \begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \tau_{12} \end{bmatrix} \quad (8)$$

As shown in Figure 5.4, now the off axis properties have to be determined. A transformation matrix (T) is used to relate the stresses in 1-2 direction with the x-y direction.

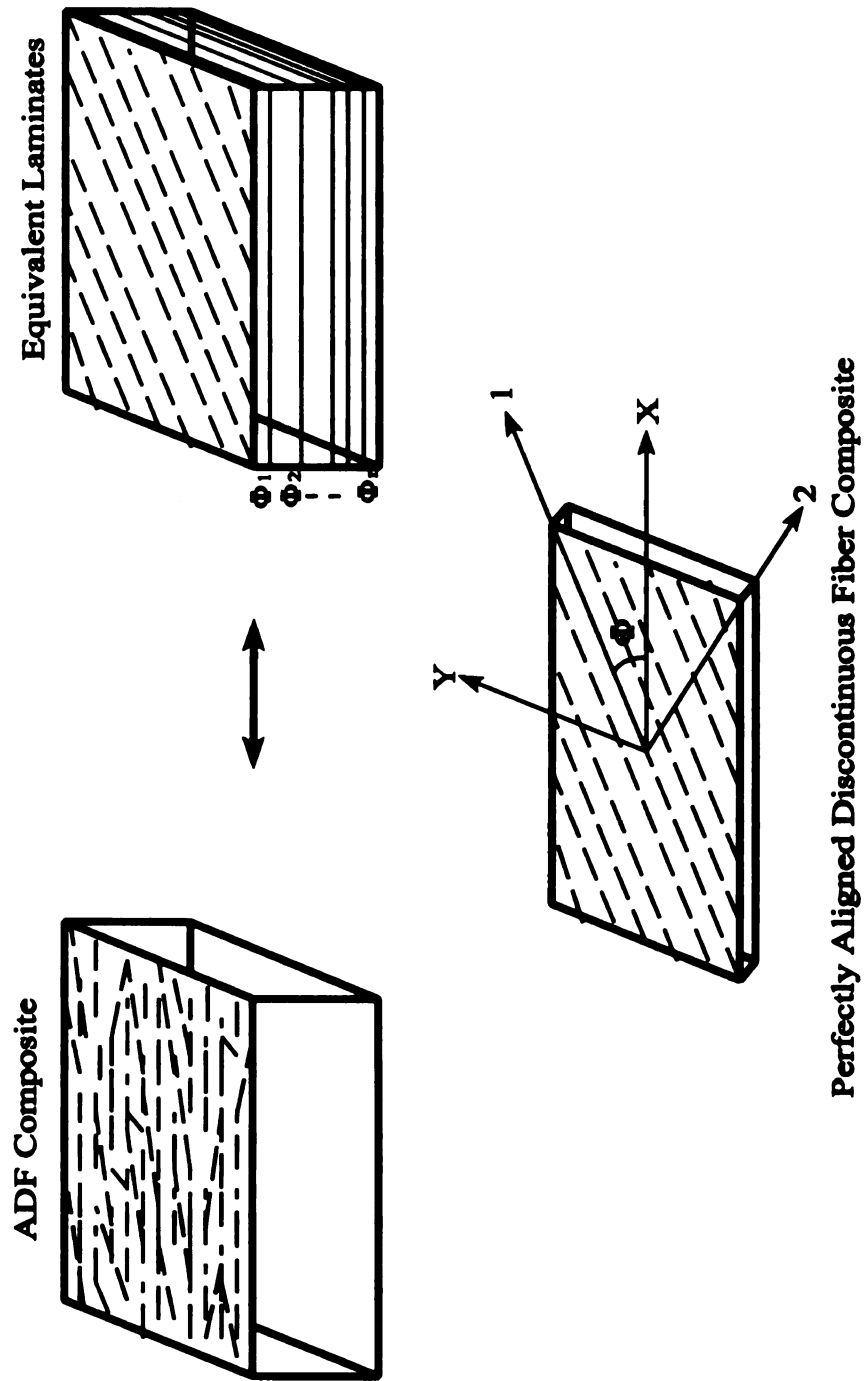


Figure 5.4 Micro-laminate concept for elastic property determination of ADF composites.

$$\begin{bmatrix} \sigma_x \\ \sigma_y \\ \tau_{xy} \end{bmatrix} = \begin{bmatrix} \cos^2\phi & \sin^2\phi & -2\sin\phi\cos\phi \\ \sin^2\phi & \cos^2\phi & 2\sin\phi\cos\phi \\ \sin\phi\cos\phi & -\sin\phi\cos\phi & \cos^2\phi - \sin^2\phi \end{bmatrix} \begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \tau_{12} \end{bmatrix} \quad (9)$$

The transformation matrix, T also transforms the strain in the 1-2 direction to x-y direction.

On further simplification of these matrix operations to determine the engineering properties in the 1-2 direction as a function of the x-y direction⁹, we get the expression for modulus (E_x) as function of the engineering properties (E_1 , E_2 and G_{12}) in the material direction.

$$\frac{1}{E_x} = \frac{\cos^4\phi}{E_1} + \left(\frac{1}{G_{12}} - \frac{2\nu_{12}}{E_1} \right) \sin^2\phi \cos^2\phi + \frac{\sin^4\phi}{E_2} \quad (10)$$

Now, the important thing is to determine the engineering properties E_1 , E_2 and G_{12} as a function of the reinforcement aspect ratio. Fundamental composite reinforcement models will be analyzed and used in the prediction of these engineering properties for the perfectly aligned discontinuous fiber composites and then predict the properties of ADF composites and later compare with experimental results. Since volumetric averaging over the thickness of the composite may be applied in the case of a uniform strain, the modulus of the composite E_c is finally obtained by integrating the expression for E_x with the fiber orientation distribution.

$$E_c = \int_0^\pi E_x(\phi) n(\phi) d\phi \quad (11)$$

The planar fiber orientation distribution function $n(\phi)$ gives the relative distribution of fibers with respect to the external coordinate axis, where the reference axis (x) can be the machine direction in the ADF process.

5.4 Reinforcement Models

In an effort to model the thermo-elastic properties of discontinuous fiber composite system, various composite reinforcement theories are evaluated for their applicability to composites reinforced with finite aspect ratio fibers and variable orientation state dispersed in a thermoplastic matrix. Shear lag, Eshelby's approach and Halpin-Tsai semi-analytical relationships are the three most used reinforcement models in composite mechanics. The applicability of one model over the other in different situations is still not well understood especially in the case of chopped fiber - thermoplastic matrix systems. Hence, all three models were considered in this analysis. For each of the models discussed below, E_c is computed as a function of the aspect ratio. The overall strategy is to obtain the microstructure-property relationships of discontinuous fiber composites as a function of the fiber aspect ratio, and fiber orientation distribution.

The Rule of Mixtures (ROM) models generally give good predictions in the case of composites with "perfect" bonding between the fiber and the matrix. *A priori* one cannot assume its applicability to short or discontinuous fiber composites because the bounds are too far apart for any practical utility and do not take into account the fiber aspect ratio or the fiber orientation distribution.

Voigt² bound (upper bound) is obtained by applying a constant strain to a composite consisting of uniformly distributed continuous fibers in a matrix. The composite property P is a simple weighted summation (parallel reaction) of the constituent properties.

$$\begin{aligned} P &= \sum P_i V_i \\ E_c &= E_f V_f + E_m V_m \end{aligned} \quad (12)$$

Reuss² bound (lower bound) is obtained by applying a constant stress to a composite consisting of uniformly distributed continuous fibers in a matrix. The reciprocal of the composite property P is a weighted summation of the reciprocal of constituent properties (series reaction).

$$\begin{aligned} P &= \sum \frac{V_i}{P_i} \\ \frac{1}{E_c} &= \frac{V_f}{E_f} + \frac{V_m}{E_m} \end{aligned} \quad (13)$$

or a fiber volume fraction of 40 %, E_c for the E-glass fiber and nylon-12 matrix system, the bounds will lie between 4.3 Msi (upper) and 0.3 Msi (lower). The futility of using the rule of mixtures for the prediction of modulus of discontinuous fiber systems can be immediately appreciated when these wide bounds are compared with experimental results given in § 5.5.

5.4.1 Shear Lag Theory

Shear lag theory was originally proposed by Cox¹⁰, where the tensile stress is transferred from the matrix to the fiber through shear stresses. Here both fibers and the

matrix behave elastically and the interface transfers the stress from the fibers to the matrix without yielding or slip. Perfect interfacial bonding is assumed and the axial strain of the composite is taken as that of the matrix in the far field. Using this approach the stiffness of the composite and the stress for the onset of matrix yielding or interfacial sliding can be calculated with the relationship given below.

$$E_c = E_f V_f \left[1 - \frac{\tanh(ns)}{ns} \right] + E_m V_m$$

$$\text{where } n^2 = \frac{2E_m}{E_f(1 + \nu_m)\ln(1/V_f)} \quad s = \frac{l_f}{d_f} \quad (14)$$

In the Kelly-Tyson¹¹ slip model, which is a special case of the shear-lag theory it is assumed that the matrix shear modulus is a constant which results in a constant interfacial shear stress. The stress transfer relationship gets simplified as explained in § 1.2.1 (see also Figure 1.1). Based on this slip model, the concept of critical fiber fragmentation length and its experimental determination has been developed by Drzal¹² and others. Matrix plasticity is applicable to well bonded reinforced metals and frictional sliding to reinforced polymers and ceramics. In the case of polymer matrices the slip-elastic formulation is presented by Piggott¹³ seems to explain the behavior better. Further modifications to the shear-lag model are introduced by taking into account the effect of the stress transfer across the fiber ends for discontinuous fiber composites. This factor is important when small aspect ratio are being dealt or small fiber/matrix stiffness ratios, for example in the case of injection molded composites or whisker reinforced metal matrix composites. This factor is incorporated in the present analysis.

One of the chief drawbacks of the original shear-lag model has been the source of error that is introduced due to neglecting the transfer of normal stresses across the end of the fiber. There have been several attempts to correct this situation. The recent solution presented by Clyne¹⁴ is not only simple but also intuitively logical and hence will be considered here along with the original Cox's model. The modification is brought about by introducing an expression for the fiber end stress (σ_e). Since the value of σ_e must lie between the peak stress in the fiber (σ_{fo}) and the far field stress in the matrix (σ_{mo}), the following expression is proposed.

$$\sigma_e = \frac{\sigma_{fo} + \sigma_{mo}}{2} \quad (15)$$

For the modified Shear Lag model, taking into account the fiber end stress transfer, we get

$$E_c = E_f V_f \left[1 - \frac{(E_f - E_m^*)}{E_f} \frac{\tanh(ns)}{ns} \right] + E_m V_m \quad (16)$$

$$\text{where } E_m^* = \frac{E_f [1 - \text{sech}(ns)] + E_m}{2}$$

The modified shear-lag model by Clyne has been tested for whisker reinforced metal matrix composites and found that the modulus values are closer to experimental values than those predicted by the original shear-lag model. This is primarily due to the small aspect ratios of the reinforcement (aspect ratio of whiskers is typically < 10) where the modification has the maximum effect.

5.4.2 Eshelby's Inclusion Approach

Eshelby¹⁵ solved for the elastic field in and around an ellipsoidal inclusion and also the associated strain energy of this system. The original model assumes an ellipsoidal inclusion with a uniform non-elastic strain, embedded in an elastic body. In the case of an ellipsoidal inclusion in a matrix of stiffness tensor, C_M , the stress (σ_I) within the inclusion can be obtained using Hooke's law in terms of the elastic strain, $(\epsilon^C - \epsilon^T)$.

$$\sigma_I = C_M (\epsilon^C - \epsilon^T) \quad (17)$$

For a shape change (ϵ^T) in the inclusion due to the imposed external stresses, all that is required is the knowledge of the final constrained strain, ϵ^C . Eshelby found that ϵ^C can be obtained from ϵ^T using a tensor which is known as the Eshelby "S" tensor. This tensor is a function of the inclusion aspect ratio and the Poisson's ratio and is given by

$$\epsilon^C = S \epsilon^T \quad (18)$$

With the constrained shape known, the inclusion stress can be evaluated as

$$\sigma_I = C_M (S - I) \epsilon^T \quad (19)$$

The S tensor allows for the calculation of the uniform stress and strain within the inclusion without having to look at the complicated stress field in the matrix itself. Numerous advances have been made to account for fiber-fiber interactions in composites by Mori-Tanaka¹⁶ and others^{17,18}.

Taya and Arsenault¹⁹ compared Shear-lag and Eshelby models for the prediction of modulus in short whisker reinforced metal matrix composites (MMC). The focus of their study was to compare the two models for aspect ratios less than 10. The rigorous solution of Eshelby's approach gives better predictions of the elastic properties in the event of small aspect ratio inclusions (<10) and when the modulus mis-match between the stiff fibers and the matrix is small ($E_f/E_m < 10$). This implies, this approach is more applicable in the case of whisker reinforced metal matrix composites than typical polymer matrix composites, where both these conditions are satisfied. In the case of short fiber reinforced polymer composites, say injection molded composites, the fiber aspect ratios are typically between 10 and 50 and $E_f/E_m > 20$. At these aspect ratios the modulus values start leveling off and often the computational effort of this approach does not merit usage of this model among the users of polymer composites.

Eshelby's approach was also considered by McGee and McCullough²⁰ to predict the elastic properties of Sheet Molding Compounds (SMC). The random nature of the fibers in SMCs creates a grainy micro-structure of ellipsoids (aspect ratios <5) where each ellipsoid is an aggregate of well aligned filaments. The problem is similar in some respects to that of the ADF composites, where the chopped fibers remain as fiber bundles even after consolidation. However, the aspect ratios are considerably higher in the case of ADF composites compared to the SMC composites (see Table 5.3). In SMC composites the fibers curve and get entangled creating grainy microstructure which can be modeled as ellipsoids of aspect ratios typically less than five. A computer program called SMC that was

developed at the University of Delaware is used in this study to predict the composite modulus at different fiber aspect and different fiber orientation parameters based on Eshelby's approach. The fiber orientation distribution is considered symmetrical and characterized by a convenient trigonometric average parameter “fp” defined in the following fashion:

$$f_p = 2\langle \cos^2 \phi \rangle - 1 \quad (20)$$

When $f_p = 1$, the fibers are perfectly aligned and when $f_p = 0$, they are perfectly random. An f_p value of 0.5 implies the fibers are reasonably aligned. The SMC program is used to determine the modulus E_c of the composite, using the f_p value and the aspect ratio of the reinforcing fiber.

5.4.3 Halpin-Tsai's Relationships

Halpin-Tsai²¹ equations are obtained by reducing rigorous elastic solutions to simpler analytical forms where the fiber geometries are taken care through the use of some empirical factors. The relations that are pertinent for the estimation of composite modulus or any property, are given below where a and b are geometrical dimensions of the reinforcement.

$$\frac{P}{P_m} = \frac{1 + \zeta \eta V_f}{1 - \eta V_f} \quad (21)$$

$$\eta = \frac{(P/P_m) - 1}{(P/P_m) + \zeta} \quad \zeta = 2 \frac{a}{b}$$

The relations have been verified for injection molded composites where the fiber aspect ratios are typically greater than 10 and it has been found that experimental results are close to predictions obtained from these relations. Halpin and Kardos²² have also predicted the properties of short fiber composites based on these relations and using micro-laminate analogy.

Figure 5.5, shows a simulation of the composite modulus (E_c) as a function of the aspect ratio, using the Shear lag models and the Halpin-Tsai relationships and the SMC program. Note these results are for the perfectly aligned cases. In all the models, the modulus values increase sharply in the beginning and reach an asymptotic value as soon as the aspect ratios reach around 100. Notice how the incorporation of the stress at fiber ends in the shear-lag model has shifted the curve upwards and reaches the asymptotic value sooner. In the simulations of perfectly aligned discontinuous fibers composites itself there is quite some variation in the prediction of each model.

In the real ADF composite, the mis-alignment of fibers has to be taken into account. It is needless to say how an accurate FOD is very important in predicting the properties of the actual composite. In the next section the elastic properties of the actual composites are predicted by incorporating the actual fiber orientation distributions of individual composites and compared with experimental results.

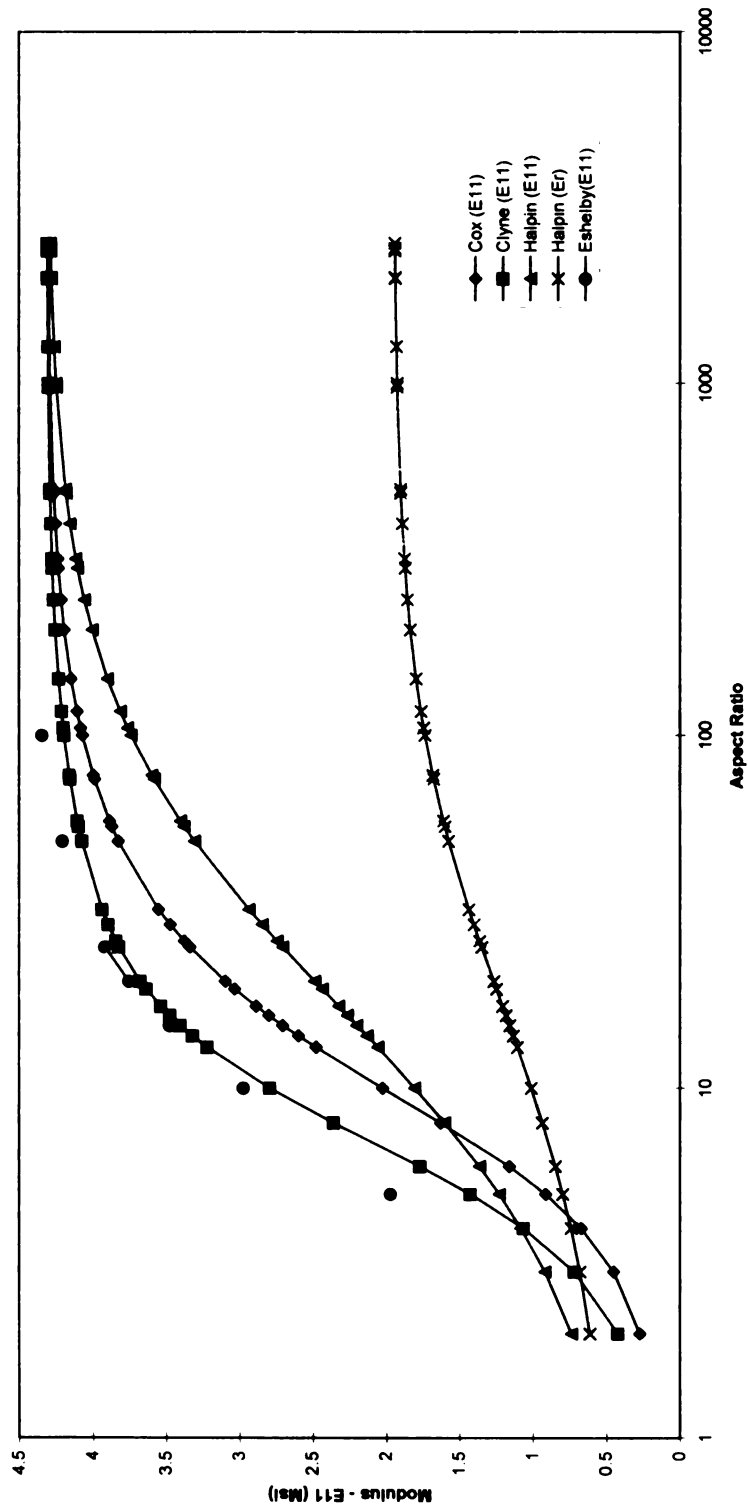


Figure 5.5 Prediction of modulus for aligned discontinuous fiber composites as a function of the reinforcement aspect ratio.

5.5 Experimental Results

5.5.1 Sample Preparation and Tensile Testing

The mechanical performance of ADF composites was characterized by determining the tensile properties using ASTM D638. A series of ADF composites were prepared using the ADF process using the fibers supplied by Owens-Corning (see Table 5.1) with lengths ranging from 0.125 to 1". For each fiber length, two panels were fabricated - one is the "Aligned" case operated at an E field intensity of 400 KV/m and the other is the "Random" case without any E field. FODs are determined for each of these cases and are given in Table 5.3. Chopped glass fiber-nylon12 ADF composite panels (4.75"x7"x0.0625") having a constant fiber volume fraction of approximately 40%, were cut into dog bone specimens using a CO₂ laser source of 360 Watts operated at a cutting speed of 30"/min. The specimens were then carefully cleaned from the edges to avoid any crack initiation during the test. Uniaxial load was applied on the dog bone specimens at a cross head speed of 0.2"/min in a standard UTS machine. This unit was also equipped with a laser extensimeter, which provided the strain on the sample. Tensile modulus and tensile strength values were obtained from each specimen tested and five specimens were tested in each panel.

5.5.2 Effect of Fiber Alignment

The tensile modulus and tensile strength of the ADF composites are plotted as a function of the fiber length in Figure 5.6 and Figure 5.7 respectively. As expected there was a significant improvement in the tensile properties of the ADF composites when the fibers were aligned, compared to the base line case (with the same fiber volume fraction) where the

fibers are randomly oriented. For each of the fiber lengths, the improvements in modulus values were nearly 70 to 100 %, while the improvements in the strength values ranged between 60 to 90 % over the non-aligned case and the un-reinforced nylon-12 matrix.

5.5.3 Effect of Fiber Length

The tensile properties of the ADF composites increases with fiber length as one would expect from the predictions of composite reinforcement theories. Both tensile modulus and strength values increase significantly with length when the fibers are aligned and the increasing trend seems to go beyond a fiber length of 1 inch. In the random fiber case, the modulus values seem to level off once the fiber length reaches 0.5", indicating that fibers longer than this length would not improve the stiffness of the composite in any significant way. A similar trend was observed for the strength curve but to a lesser extent.

5.6 Model Predictions and Discussion

Based on the critical fiber length postulation that was presented in Chapter 1, the properties of aligned discontinuous fiber composites should approach the properties of continuous fiber composites when the length of the fiber is about ten times the critical fiber length (say about 0.5 inches). Indeed, the experimental results show significant improvements in properties (Figures 5.6 and 5.7). However, the modulus and the strength values do not seem to level off once the fiber length exceeds 0.5 inches, indicating the performance levels of continuous fiber composites have not been completely achieved. So the question that remains unanswered is, what is the actual reinforcement length? Secondly,

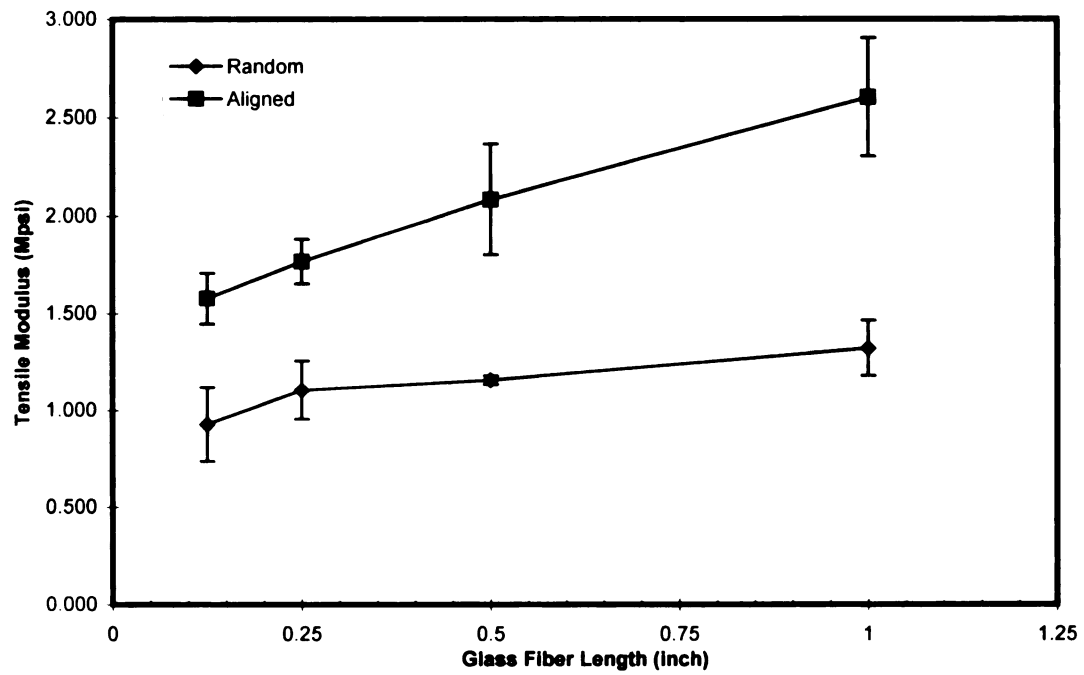


Figure 5.6 Effect of fiber alignment and fiber length on the tensile modulus.

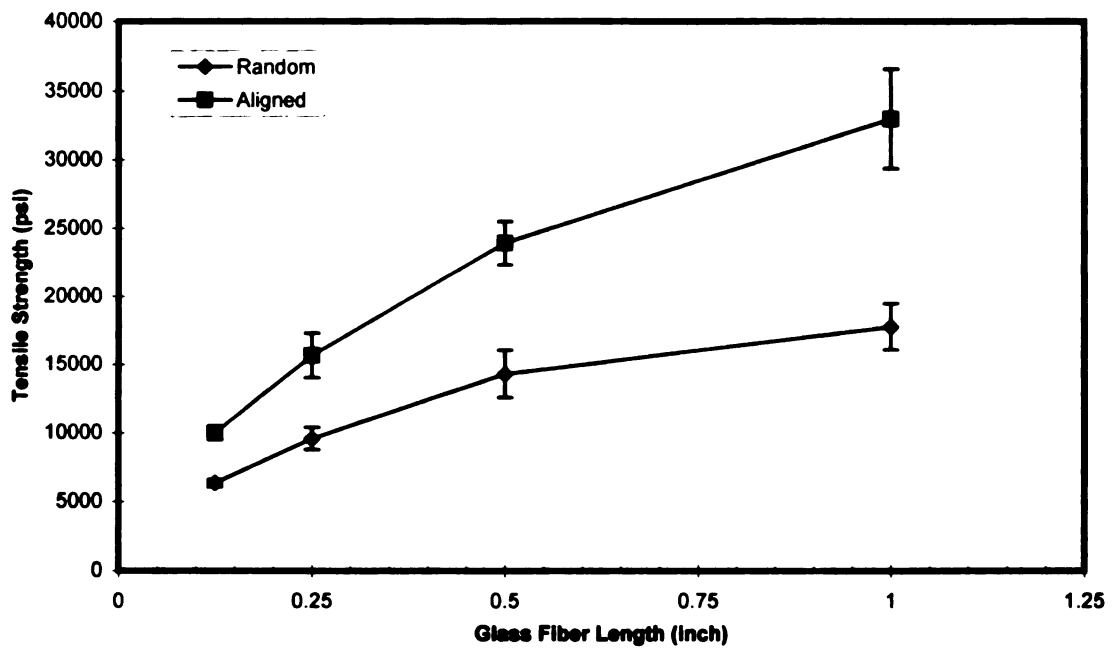


Figure 5.7 Effect of fiber alignment and fiber length on the tensile strength.

what is the effect of fiber mis-alignment on the properties in the aligned fiber cases? Thirdly, how good are the predictions from the reinforcement models that were elaborated in § 5.4.

These questions can be answered by looking at the predictions obtained from the fundamental composite reinforcement theories which also take into account the actual FOD of the composites. Based on the above formulations and the experimental data obtained computations were performed to determine the modulus of the composite, E_c at the two extremes of the reinforcement aspect ratio. These two extremes of the aspect ratios are the limits of interest based on the observations made from the micro-structural micrographs i.e “filament reinforcement “ and “bundle reinforcement” (Figures 5.2 and 5.3). Table 5.4 shows the details of the model predictions for the actual ADF composites that were fabricated and tested. Based on the chopped fiber length, the two different aspect ratios are considered: “filament” aspect ratio and “bundle” aspect ratio. Relationships presented in § 5.4 are then used to obtain E_c for the two different aspect ratios. These modulus values for the “perfectly aligned composites” are then used to obtain the modulus of the actual ADF composite by integrating the effect of fiber orientation using the FOD. Notice for each fiber length, two fiber orientation states were tested: aligned and random. The fiber orientation state was also characterized by the term “fp” to get the predictions from the SMC program.

Table 5.5 Model Predictions Compared with Experimental Modulus Results**"Aligned" ADF Composites**

Fiber Length	fp	Expt. Result	Halpin-Tsai (Msi)		Eshelby (Msi)	
(in)		Msi (Std. Dev.)	"Filament"	"Bundle"	"Filament"	"Bundle"
0.13	0.74	1.58 (0.13)	2.37	1.48	3.22	3.22
0.25	0.63	1.77 (0.11)	2.16	1.66	2.92	2.89
0.50	0.63	2.08 (0.28)	2.38	2.04	2.93	2.72
1.00	0.73	2.6 (0.30)	2.4	2.21	3.25	2.59

"Random" ADF Composites

Fiber Length	fp	Expt. Result	Halpin-Tsai (Msi)		Eshelby (Msi)	
(in)		Msi (Std. Dev.)	"Filament"	"Bundle"	"Filament"	"Bundle"
0.13	0.18	0.93 (0.19)	1.17	0.87	1.86	1.86
0.25	0.12	1.10 (0.15)	1.15	0.97	1.97	1.96
0.50	0.10	1.16 (0.02)	1.12	1.01	2.00	1.87
1.00	0.02	1.32 (0.14)	1.07	1.02	2.08	1.71

Halpin-Tsai relations under predict the modulus for small aspect ratio fibers and this has been evident in the earlier comparisons between Shear lag and Halpin-Tsai relations also (Figure 5.5). A closer look at the predictions made by the Eshelby technique indicates that it over predicts and secondly the effect of aspect ratio also seems to decrease much faster than any of the other techniques. When the fiber bundle effect is considered, the predictions made by the Halpin-Tsai method are closer to the experimental data (within the experimental scatter).

Based on the experimental results and the theoretical predictions it can be concluded that the chopped E-glass fiber - nylon12 composite system that was studied, the reinforcement microstructure is an aggregate of individual filaments and that the effect of reinforcement is a function of the bundle aspect ratio. Since, the critical fiber length concept of the shear-lag theory was based on the assumption of an individual filament surrounded by matrix on all sides, it cannot be directly applied to this situation when the reinforcement microstructure is an aggregate of individual filaments and hence was not computed for comparison with experimental values (Table 5.4). Further work is required in modifying the shear-lag concept for reinforcements that are aggregates of filaments. However, the reinforcement trend that was shown in Figure 1.2 as a function of fiber length as predicted by the critical fiber length concept is evidenced in the experimental results shown in Figure 5.6. This proves that the underlying concept of critical fiber length exists even in the case of “bundle reinforcement”.

Finally, the following conclusions can be summarized from this study of the microstructure-property relationships.

- The properties of the discontinuous fiber composites improve with increase in fiber alignment and an increase in fiber length.
- In the chopped fiber-thermoplastic systems, the effective reinforcement aspect ratio is a function of the bundle size and fiber-matrix interaction which includes resin impregnation in to the bundle and the sizing compatibility.
- The effective reinforcement aspect ratio approaches the filament aspect ratio only when the fiber-matrix interaction is optimized and the effective reinforcement aspect ratio approaches the bundle aspect ratio when there is no resin impregnation in to the fiber bundle.

References

1. Carman, G. P. and Reifsnider, K. L., "Micromechanics of short-fiber composites", *Composite Science and Technology*, 43, pp 137-146, 1992.
2. Eduljee, R. F. and McCullough, R. L., "Elastic properties of composites", in *Material Science and Technology - A Comprehensive Treatment*, Vol 13, Ed. Chou, T. W., VCH, 1993.
3. Ericson, M. L. And Berglund, L. A., "Processing and Mechanical Properties of Oriented Preformed Glass-Mat-Reinforced Thermoplastics", *Composites Science and Technology*, Vol. 49, p121-130, 1993.
4. Kacir, L., Ishai, O. And Narkis, M., "Oriented Short Glass-Fiber Composites. IV. Dependence of Mechanical Properties on the Distribution of Fiber Orientations", *Polymer Engineering and Science*, Vol 18, No. 1, p45-52, 1978.
5. Piggott, M. R., Ko., M. and Chuang, H. Y., "Aligned Short-Fiber Reinforced Thermosets: Experiments and Analysis Lend Little Support for Established Theory", *Composites Science and Technology*, Vol 48, p291-299, 1993.
6. Madhukar, M. S. and Drzal, L. T., "Fiber-Matrix Adhesion and its Effect on Composite Mechanical Properties: II. Longitudinal (0°) and Transverse (90°) Tensile and Flexure Behavior of Graphite/Epoxy Composites", *J. Of Composite Materials*, Vol. 25, p 958-991, 1991.
7. Tang, J. M., Lee, W. I., Springer, G. S., "Effects of Cure Pressure on Resin Flow, Voids and Mechanical Properties", *J. Of Composite Materials*, Vol. 21, pp 421-440, 1987.
8. Harper, B. D., Staab, G. H., Chen, R. S., "A Note on the Effects of Voids upon the Hygral and Mechanical Properties of AS4/3502 Graphite/Epoxy", *J. Of Composite Materials*, Vol. 21, pp 280-289, 1987.
9. Piggott, M. R., "Load Bearing Fiber Composites", Pergamon, 1980.
10. Cox, H. L., "The elasticity and strength of paper and other fibrous materials", *British J. of Applied Physics*, 3:72, 1952.
11. Kelly, A. And Tyson, W. R., "Tensile Properties of Fiber-Reinforced Metals: Copper/Tungsten and Copper/Molybdenum", *J. Mech. Phys. Solids*, Vol. 13, p329-350, 1965.
12. Herrera-Franco, P. J. And Drzal, L. T., "Comparison of Methods for the Measurement of Fiber/Matrix Adhesion in Composites", *Composites*, Vol. 23, No. 1, 1992.
13. Piggott, M. R., "Short Fiber Polymer Composites: A Fracture Based Theory of Fiber Reinforcement", *J. of Composite Materials*, Vol 28, No. 7, 1994.

14. Clyne, T. W., " A Simple Development of the Shear Lag Theory Appropriate for Composites with a Relatively Small Modulus Mismatch", *Materials Science & Engineering*, A122 (1989) 183-192.
15. Eshelby, J. D., "Proc. Roy. Soc. Lond., A241, 376, 1957.
16. Mori, T. and Tanaka, K., *Acta Metall.*, Vol 21, 571, 1973.
17. Christensen, R. M., "Mechanics of Composite Materials", John Wiley and Sons, New York, 1979.
18. Mura, T., " Micromechanics of Defects in Solids", Nijhoff, The Hague, 1987.
19. Taya, M. and Arsenault, R. J., "A Comparison Between a Shear Lag Type Model and an Eshelby Type Model in Predicting the Mechanical Properties of a Short Fiber Composite", *Scripta Metallurgica*, Vol. 21, p 349-354, 1987.
20. McCullough, R. L., " Micro-models for Composite Materials-Particulate and Discontinuous Fiber Composites" in *Micromechanical Materials Modeling*, 1989.
21. Halpin, J. C. And Kardos, J. L., "The Halpin-Tsai Equations: A review", *Polymer Engineering and Science*, Vol 16, No. 5, 1976.
22. Halpin, J. C., and Kardos, J. L., "Strength of Discontinuous Reinforced Composites: I Fiber Reinforced Composites", *Polymer Engineering and Science*, Vol 18, No 6, 1978.

Conclusions and Future Work

The problem of processing discontinuous fiber polymer composites with fiber alignment in a preferred direction was solved by a unique combination of fiber alignment using electric fields in air and polymer powder processing, which resulted in a rapid, solvent free processing technique. The development of the prototype Aligned Discontinuous Fiber (ADF) composite process was a result of the integration of the fundamental understanding and investigations into the following aspects:

- Behavior of fiber alignment in air using electric fields and
- Microstructure-property relationships of discontinuous fiber composites.

The conclusions from this study are summarized below.

1. A semi-continuous prototype process has been developed that can manufacture aligned discontinuous fiber composites using electric fields. The key steps in the process are: alignment of conducting or insulating fibers in air using electric fields; powder coating/impregnation of fibers, and; consolidation of the powder coated ADF preform into a composite. The ADF process has been demonstrated for E-glass fiber and nylon12 matrix system and can be extended to other fiber-matrix combinations

2. E-glass fibers of lengths ranging from 0.125 to 1 inch have been aligned using alternating current (A.C.) electric fields with intensities ranging from 300 to 600 KV/m. A.C. fields were preferred over D.C. fields to avoid electrophoretic effects which disturbed the fiber alignment process. Since the polarization times of the fibers are very rapid, A.C. fields with a frequency of 60 Hz was found to be adequate for effective fiber alignment.
3. Fiber alignment in air using electric fields is extremely fast. The alignment times for dielectric fibers like E-glass range between 0.1 to 0.5 seconds for the fibers lengths investigated (ranging from 0.125 to 1 inch). This rapid alignment makes the ADF processing technique very attractive for high speed processing.
4. The alignment behavior of the fibers in air is more complicated than it would be in a more viscous fluid medium due to the non-buoyant nature of fiber motion in air. A balance exists between the electrical torque due to the polarization of fibers; hydrodynamic forces due to fluid resistance and rotational torque due to resultant shearing action. With air as the fluid medium, the viscous resistance is small and gravitational and the rotational forces become important. Fiber alignment is a strong function of E-field strength, aspect ratio and the stability in the fiber motion. Although it is predicted (in the case of buoyant situation) that the alignment time increases with fiber aspect ratio, it has been found that longer fibers have better

alignments for the same electric field intensity due to their more stable motion during free fall.

5. The final design of the chamber for controlling fiber orientation in air has two unique features that counter the edge effects of the electric fields when the deposition platform is set in motion during the continuous processing of ADF preforms.
 - A field equal in intensity and opposite in direction has been added under the high voltage electrode by placing under the moving slide/mat/veil. This neutralizes the edge effects of the high voltage electrode, which otherwise disturbs the fiber orientation of the fibers that are aligned by the E-field and that are deposited on the mat.
 - A special design for the edges of the electrodes which are closer to the moving mat has been developed. A conductive cap which has a cylindrical cross-section with a relatively large radius of curvature has been used to reduce the effective E-field intensity at the edge, thereby reducing the edge effects and minimizing E-field breakdowns.
6. Developed a unique vibratory fiber feeder and pre-aligner. Fibers with or without powder impregnation can be fed into the orientation chamber without entanglement. Fibers falling from the feeder pre-aligner plate tend to fall with a planar orientation state instead of a three dimensional random state which makes them align faster in the direction of the electric field. This feeding mechanism has the added benefit of

increasing the theoretical critical processing concentration of fibers that can be used in the fiber orientation chamber.

7. The fiber orientation distribution in the ADF preform as well as the orientation chamber was obtained using digital image analysis techniques. Under the prototype ADF processing conditions, for all the E-glass fiber lengths nearly 70 % of the fibers of the fibers were aligned $\pm 20^\circ$ and nearly 80 - 90 % within $\pm 30^\circ$, indicating a high degree of finer alignments using electric fields. Further, improvements in alignments can be made based on the understanding gained on the alignment behavior of fibers in air.
8. Polymer powder coating of the fibers and controlling the matrix volume fraction in the final composite is one of the biggest advantages of the ADF process. The combination of powder coating with electric field alignment makes the process a potentially high speed-low cost process to make aligned discontinuous fiber composites. This step eliminates the additional step of resin transfer. The technology of powder coating is well established in the group and extensively patented and hence can be easily incorporated in the ADF process developed.
9. The properties of the discontinuous fiber composites improve with an increase in fiber alignment and an increase in fiber length. Improvements in modulus of the ADF composites ranged from 70 to 100 %, when compared to equivalent composites

that were manufactured without the electric field. Similar improvements in strength values have also been noted. The improvement in properties due to a combination of alignment and increasing the length from 0.125 to 1 inch increases the property by nearly 300 %.

10. Discontinuous fiber composites with known fiber orientation distribution and fiber aspect ratio had been fabricated using the ADF process. This provided a unique opportunity to apply fundamental micromechanics analysis to compare theoretical predictions with experimental results. Also, it provided the basis to verify one of the primary objective of the ADF process development - the properties of ADF composites should approach the properties of continuous fiber composites as the length of the fibers increase and as the fibers are aligned in the direction of the stress. This microstructure-property relationships also integrates processing and performance in the ADF process because the fiber aspect ratio is a critical parameter in the fiber alignment process itself.
11. Analysis from microstructure-property relationships indicated that in the chopped fiber-thermoplastic systems, the effective reinforcement aspect ratio is a function of the bundle size and fiber-matrix interaction which includes resin impregnation into the bundle and the sizing compatibility. Further, it may be concluded that the effective reinforcement aspect ratio approaches the filament aspect ratio only when the fiber-matrix interaction is optimized and the effective reinforcement aspect ratio

approaches the bundle aspect ratio when there is no resin impregnation in to the fiber bundle.

12. Fundamental fiber reinforcement theories were used to predict the properties of the ADF composites that were fabricated in the process with a known fiber orientation distribution and aspect ratio. The Halpin-Tsai model was found to predict the properties reasonably well, while the model based on Eshelby's approach over predicts the modulus. The shear-lag approach could not be directly applied because of the aggregate nature of the fibers. In all the models the predictions were closer when the reinforcement aspect ratio was taken to be the bundle aspect ratio rather than filament aspect ratio.

Future Research

The work presented in this dissertation establishes the concept and operating principles of a processing technique for manufacturing aligned discontinuous fiber composites. It is envisaged that this kind of approach will eventually lead to high speed, highly automated, solvent free, manufacture of microstructure controlled material forms, where material performance based on the microstructure is taken care right at the manufacturing step. High speeds of fabrication are possible and amenable to CAD/CAM robotic manufacturing techniques. This will especially prove to be very effective in the case of a complex lay-up sequence often needed in a large part. Based on the Computer-Aided Design, one can program the field directions in the orientation chamber and the fiber and

matrix powder feed rate to lay up powder impregnated fibers in the desired location with the desired fiber orientation with the orientation chamber being moved by a robotic arm.

In this project the focus was on controlling the fiber orientation for reinforcement, but the concept can be exploited for a variety of other functions properties like electrical conductivity and the thermal properties by a suitable selection of the fibers and the matrix. A polymer composite ADF material form with controlled electrical conductivity may find applications in electro-magnetic shielding. A polymer composite ADF material form with preferred fiber orientation may also become very critical in bio-materials especially in the area of body implants. This ADF material form also has great applications in the automotive, sports and recreation industries where light weight structural materials are needed.

Some remaining specific issues that need to be investigated for this processing approach are:

- A comprehensive model needs to be developed for characterizing the electrodynamics of fiber motions incorporating inertial effects for the fiber dimensions that are of interest to composites manufacture.
- The limitations on the processing concentrations of fibers need to be established based on the E-field distortion effects due to fiber-fiber interactions in the orientation chamber.
- Investigate if the processing rate can be increased by applying a vacuum, through the orientation chamber. If such a measure is adopted, how will the fiber alignment dynamics change?

- Investigate the effect of multiple orientation chambers in series and study the effect of fiber mis-alignment if any, as the ADF preform builds up in thickness.
- Investigate, hybrid fiber systems where a mixture of fibers of different dielectric and/or geometrical features are fed simultaneously through the orientation chamber to get different fiber orientation distributions in different directions.
- Engineer different type of electrodes to create field configurations to manufacture unique material forms and potentially 3D orientation structures.
- Study the stamping or thermoforming behavior of ADF composites and characterize stampability of these materials as a function of fiber length, fiber orientation and polymer rheology.