

**DEVELOPMENT AND ANALYSIS OF LONG FIBER REINFORCED  
POLYPROPYLENE COMPOSITES**

by

**K. SENTHIL KUMAR**

**Centre for Polymer Science and Engineering**

**Submitted**

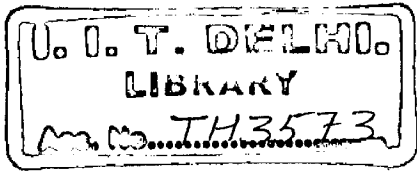
in fulfillment of the requirements of the degree of Doctor of Philosophy

to the



**INDIAN INSTITUTE OF TECHNOLOGY DELHI**

**December 2007**



1. Polypropylene



TH  
678.742  
SEN-D

## CERTIFICATE

This is to certify that the thesis entitled “**Development and Analysis of Long Fiber Reinforced Polypropylene Composites**” submitted by **Mr. K. Senthil Kumar** to the Indian Institute of Technology, Delhi, for the award of degree of **Doctor of Philosophy**, in Polymer Science and Technology, is a record of bonafide research work carried out by him. **Mr. K. Senthil Kumar** has worked under our guidance and supervision and has fulfilled the requirements for the submission of this thesis, which to our knowledge has reached the requisite standard.

The results contained in this thesis are original and have not been submitted, in part or full, to any other University or Institute for the award of any degree or diploma.

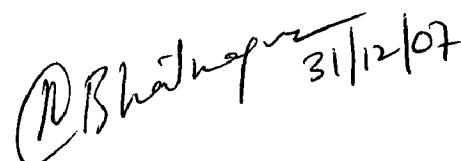


**(Dr. A. K. Ghosh)**

**Professor**

**Centre for Polymer Sci. and Engg.**

**IIT Delhi**



**(Dr. N. Bhatnagar)**

**Associate Professor**

**Department of Mechanical Engg.**

**IIT Delhi**

## *Acknowledgement*

*It is a pleasant aspect to express my deep sense of gratitude to all who have helped me along my way through the doctoral studies and a memorable stay at IIT Delhi. There are, of course, very few words to properly thank all of them.*

*My foremost thanks go to my supervisor Prof. Anup K Ghosh and Dr. Naresh Bhatnagar. I am deeply indebted to them for their mentoring, able guidance, stimulating suggestions and inspiring thoughts which have been of tremendous help for me throughout my doctoral studies. They have always been there, through every thick and thin and encouraged me to move forward in life. I truly appreciate their patience and tolerance during my numerous misadventures. The many hours of discussions we had in which they showed their enthusiasm and positive attitude towards this study and topics which strayed rather far from 'composites', kept me on the right track. I appreciate and acknowledge their help in developing my self confidence and sense of autonomy. Their overly enthusiasm and integral view on research and their mission for providing nothing short of high quality work has made a deep impression on me. I hereby very frankly confess that the present research endeavor would not have seen the day's light without the esteemed guidance and support of Prof. Anup K Ghosh and Dr. Naresh Bhatnagar.*

*I would also like to thank other members of my Ph.D committee Prof. B. L. Deopura, Prof. R. K. Mittal and Prof. S. N. Maiti who provided me with valuable suggestions and advice. I am also thankful to Prof. A. K. Gupta, Prof. I. K. Varma, Prof. Veena Choudhary, Prof. R. K. Pandey and Prof. A. R. Ray for their support.*

*A journey is easier when traveled together. Interdependence is certainly more valuable than independence. This thesis is the result of more than four years of work in CPSE, IIT Delhi, accompanied by the support from many people. My special thanks to Mr. Devander Singh, Mr. Surender Sharma, Mr. Ashok Kapoor, Mr. Shivkant and all official staffs for their immediate help*

whenever needed. Also I sincerely thank Mr. Tulsiram, Mr. Kustwaha, Mr. Ram Chander and Mr. Mukhesh who have always helped me in the best possible way they could. They were ready to tender all kind of help anytime and were constant source of encouragement. My sincere regards to them all.

My colleagues and friends, Joy, Ananthapadmanabha, Kavita, Sangita, Dilip, Prasath Balamurugan, Prakashan, Selvakumar, Parvin, Muthulakshmi, Dipti, Pooja, Neetu, Purnima, Nimisha, Rashmi, Satpal, Saikat, Upender, supported me in my research work. I wish to thank them for all their constant help, support, interest and valuable hints. Thanks to Pramila and Aditi for taking care of thesis illustrations and processing my official documents.

The principle, to see the good in everything and be a constructive part of the whole, made me feel responsible to participate in lot of activities other than research. I am grateful to various people who have not been directly involved in this project described in this thesis, but with whom I have been in touch during my Ph.D study. Thanks to Dr. Shashi Motilal, for the views and discussion that helped me to keep the universal spirit that is essential for scientific research.

I am deeply indebted to my beloved parents who are at the back of my each and every movement. I wish to record my sincere appreciation and thanks to them for their invaluable support, patience and encouragement throughout my study years. Without them there is no way in which I could possibly have accomplished this stupendous task. My father still provides a role model for his sincerity and hard working nature and my mother for her love, care, patience and persistent inspiration for my journey in this life. I am thankful to my brother, Saravana Kumar who in spite of being younger to me had always been a helping hand.

I would like to take this opportunity to thank one and all who have helped me during the completion of this project. In the end, I thank all those who do not figure in the list above and have helped me in one way or the other in the completion of this project.

  
K. Senthil Kumar

## ABSTRACT

Long fiber-reinforced thermoplastics (LFRT) have received much attention because of its possible processability by conventional technologies. The aim of this work is to develop long fiber reinforced polypropylene (LFPP) composites and to correlate the mechanical properties with its microstructure. The work has been carried out in four different phases. In the first phase, a radial impregnation die (RID) was designed and fabricated and retrofitted with an existing single screw extruder. The die was designed to have maximum possible impregnation of glass fiber roving with thermoplastic melt in the extruder. The quality of impregnation was assessed and the impregnated strands were chopped to 3 mm, 6 mm 9 mm and 12 mm length as discontinuous long fiber pellets. Processing these long fiber pellets in injection molding machine was the second phase of work. Fiber length distributions and fiber orientations were also analyzed. The third phase of the work concentrates on the static and dynamic mechanical properties of injection molded composites. Analyzing the LFPP composites for fracture toughness, fatigue and failure behaviour was the final phase of this research work. Structure-toughness relationship was correlated using microstructure efficiency concept. Fatigue life of LFPP composites were determined using tension-tension test method. Fatigue crack growth propagation was observed for different LFPP composites and correlated with microstructure. Failure behaviour was observed using SEM pictures. This work makes a useful experimental foundation in developing strategies with long fiber thermoplastic composites and to understand the relationship between its improved structure and properties.

## TABLE OF CONTENT

<i>Abstract</i>	<i>vi</i>
<i>List of figures</i>	<i>xiv</i>
<i>List of tables</i>	<i>xx</i>
<b>Chapter 1</b> <b>Introduction</b>	
1.1 <i>Introduction</i>	1
1.2 <i>Classification of composites</i>	2
1.3 <i>Thermoplastic and thermosetting composites</i>	5
1.4 <i>Strengthening a composite</i>	8
1.5 <i>Importance of fiber length</i>	8
1.6 <i>Long fiber reinforced thermoplastic composites</i>	12
1.6.1 <i>History</i>	12
1.6.2 <i>Market</i>	13
1.6.3 <i>Need for research</i>	15
1.6.4 <i>Motivation</i>	16
1.6.5 <i>Process performance</i>	16
1.6.6 <i>Property performance</i>	17
1.8 <i>Format of thesis</i>	19

## **Chapter 2    Literature Review**

<b>2.1</b>	<i>Development of long fiber reinforced thermoplastic composites</i>	21
2.1.1	<i>Impregnation</i>	24
2.1.2	<i>Injection molding</i>	27
2.1.3	<i>Fiber/ matrix adhesion</i>	28
<b>2.2</b>	<i>Analysis of long fiber reinforced thermoplastic composites</i>	31
2.2.1	<i>Fiber length and fiber orientation analysis</i>	32
2.2.2	<i>Mechanical property analysis</i>	34
2.2.3	<i>Fracture, fatigue and failure analysis</i>	39
<b>2.3</b>	<i>Problem definition</i>	42
<b>2.4</b>	<i>Objective and approach</i>	43
<b>2.5</b>	<i>Methodology</i>	44

## **Chapter 3    Preparation of Long Fiber Polypropylene Pellets**

<b>3.1</b>	<i>Materials</i>	48
3.1.1	<i>Glass fiber roving</i>	48
3.1.2	<i>Polypropylene</i>	48
3.1.3	<i>Characterization of polypropylene</i>	49
3.1.4	<i>Maleic anhydride grafted polypropylene</i>	52
<b>3.2</b>	<i>Impregnation</i>	54
3.2.1	<i>Requirements for impregnation</i>	55
3.2.2	<i>Impregnation methods</i>	56

3.3	<i>Hot melt impregnation in extruder</i>	58
3.3.1	<i>Features of extruder</i>	59
3.3.2	<i>Melt flow in an extruder</i>	61
3.3.3	<i>Viscosity control in an extruder</i>	62
3.3.4	<i>Melt flow differences in an extruder</i>	64
3.4	<i>Radial impregnation die</i>	65
3.4.1	<i>Design complexity</i>	65
3.4.2	<i>Design considerations</i>	65
3.4.3	<i>Die design</i>	66
3.4.4	<i>Design parameters</i>	69
3.5	<i>Preparation of impregnated strands</i>	70
3.5.1	<i>Process parameters</i>	71
3.6	<i>Extruder screw / Die characteristic curves</i>	72
3.7	<i>Evaluation of impregnation</i>	74
3.8	<i>Preparation of pellets</i>	78
3.9	<i>Summary</i>	79

#### **Chapter 4    *Processing of Long Fiber Polypropylene Pellets***

4.1	<i>Background</i>	82
4.2	<i>Injection molding</i>	83
4.2.1	<i>Machine specifications</i>	83
4.2.2	<i>Process parameters</i>	84
4.2.3	<i>Machine parameters</i>	85

4.2.4	<i>Mold parameters</i>	86
4.3	<i>Preparation of test specimens</i>	87
4.4	<i>Fiber length analysis</i>	88
4.4.1	<i>Procedure for fiber length determination</i>	88
4.4.2	<i>Assessment of fiber length distributions</i>	88
4.4.3	<i>Fiber length at nozzle</i>	90
4.4.4	<i>Fiber length at sprue</i>	91
4.4.5	<i>Fiber length at runner</i>	92
4.4.6	<i>Fiber length after gate</i>	93
4.4.7	<i>Fiber length in mold cavity</i>	94
4.5	<i>Fiber attrition in injection molding</i>	96
4.6	<i>Fiber orientation analysis</i>	97
4.6.1	<i>Forms of orientation</i>	97
4.6.2	<i>Orientation distribution function</i>	98
4.6.3	<i>Orientation tensor components</i>	100
4.6.4	<i>Orientation parameter</i>	102
4.7	<i>Fiber orientation in LFPP composites</i>	103
4.7.1	<i>Sample preparation by polishing method</i>	103
4.7.2	<i>Orientation in skin layer</i>	105
4.7.3	<i>Orientation in transition layer</i>	106
4.7.4	<i>Orientation in core layer</i>	108
4.7.5	<i>Fiber orientation factor in composites</i>	109
4.8	<i>Summary</i>	110

## **Chapter 5    *Mechanical Properties of Long Fiber Polypropylene Composites***

<b>5.1</b>	<b><i>Background</i></b>	<b>114</b>
<b>5.2</b>	<b><i>Fiber/matrix interface</i></b>	<b>115</b>
5.2.1	<i>Single fiber pull-out test</i>	116
5.2.2	<i>Optimization of compatibilizer</i>	117
5.2.3	<i>Critical fiber length</i>	119
<b>5.3</b>	<b><i>Mechanical properties of LFPP</i></b>	<b>120</b>
5.3.1	<i>Tensile strength</i>	121
5.3.2	<i>Theoretical model for tensile strength</i>	122
5.3.3	<i>Tensile modulus</i>	125
5.3.4	<i>Theoretical model for tensile modulus</i>	127
5.3.5	<i>Flexural properties</i>	130
5.3.6	<i>Impact strength</i>	132
<b>5.4</b>	<b><i>Dynamic mechanical properties of LFPP</i></b>	<b>134</b>
5.4.1	<i>Conservative modulus</i>	136
5.4.2	<i>Mechanical loss modulus</i>	137
5.4.3	<i>Mechanical loss tangent</i>	138
5.4.4	<i>Activation energy</i>	139
5.4.5	<i>Kinetics of glass transition</i>	140
5.4.6	<i>Storage modulus at <math>T_g</math></i>	146
5.4.7	<i>Activation energy at <math>T_g</math></i>	147
<b>5.5</b>	<b><i>Summary</i></b>	<b>149</b>

## **Chapter 6     *Fracture Toughness, Fatigue and Failure Analysis of LFPP composites***

<b>6.1</b>	<i>Background</i>	<b>151</b>
<b>6.2</b>	<i>Fracture Toughness</i>	<b>153</b>
6.2.1	<i>Crack growth resistance</i>	<b>153</b>
6.2.2	<i>Specimen preparation</i>	<b>155</b>
6.2.3	<i>Fracture toughness</i>	<b>156</b>
6.2.4	<i>Microstructural efficiency concept</i>	<b>157</b>
6.2.5	<i>Structure – Toughness relationship</i>	<b>160</b>
<b>6.3</b>	<i>Fatigue Analysis</i>	<b>164</b>
6.3.1	<i>Fatigue test methods</i>	<b>165</b>
6.3.2	<i>Sample preparation and testing condition</i>	<b>166</b>
6.3.3	<i>Fatigue life of composites</i>	<b>167</b>
6.3.4	<i>Fatigue crack propagation</i>	<b>171</b>
6.3.5	<i>Structure – Fatigue relationship</i>	<b>176</b>
<b>6.4</b>	<i>Failure Analysis</i>	<b>179</b>
6.4.1	<i>Microscopic failure mechanism</i>	<b>180</b>
6.4.2	<i>Influence of compatibilizer</i>	<b>180</b>
6.4.3	<i>Influence of fiber length</i>	<b>183</b>
6.4.4	<i>Failure behaviour of impact tested samples</i>	<b>183</b>
<b>6.5</b>	<i>Summary</i>	<b>185</b>

<b>Chapter 7</b>	<b><i>Summary of Results and Conclusions</i></b>	
7.1	<i>Summary of Results</i>	187
7.1.1	<i>Development of LFPP composites</i>	189
	a) <i>Preparation of pellets</i>	189
	b) <i>Processing of pellets</i>	190
7.1.2	<i>Analysis of LFPP composites</i>	191
	a) <i>Mechanical properties</i>	191
	b) <i>Fracture, fatigue and failure analysis</i>	192
7.2	<i>Conclusions</i>	193
7.3	<i>Scope for future work</i>	194
	<i>References</i>	197
	<i>Appendix 1</i>	207
	<i>List of Publications &amp; Biography</i>	221