MODIFIED EPOXY RESIN-GLASS FIBER REINFORCED LAMINATES

By

R. G. RAJ

A thesis submitted to the
Indian Institute of Technology, Delhi
for the award of the degree of
DOCTOR OF PHILOSOPHY

Department of Textile Technology
INDIAN INSTITUTE OF TECHNOLOGY, DELHI

JUNE, 1983

CERTIFICATE

This is to certify that the thesis entitled "Modified Epoxy resin-Glassfiber Reinforced Laminates" being submitted by Mr. R.G. Raj to the Indian Institute of Technology, Delhi, for the award of the degree of Doctor of Philosophy in Textile Technology, is a record of bonafide research work carried out by him. Mr. R.G. Raj has worked under our guidance and supervision and has fulfilled the requirement for the submission of this thesis which to our knowledge has reached the requisite standard.

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

(A.K. Gupta)
Assistant Professor,
Centre for Materials Science and Technology,
Indian Institute of Technology,
New Delhi-110016, India.

(D.S. Varma)
Professor of Fibre Science &
Dean of Students,
Department of Textile Technology,
Indian Institute of Technology, Delhi,
New Delhi-110016, India.

ACKNOWLEDGEMENTS

I am greatly indebted to Professor D.S. Varma of Textile Technology Department, Indian Institute of Technology, Delhi, for his inspiring, painstaking guidance and constant encouragement throughout the course of this work.

I am specially thankful to Prof. (Mrs.) I.K. Varma, Head of Centre for Materials Science and Technology, IIT Delhi, for her keen interest and valuable suggestions throughout the course of this work.

I express my thanks to Dr. A.K. Gupta, Assistant Professor, Centre for Materials Science & Technology, for helping me during the absence of my guide, Prof. D.S. Varma.

My sincere thanks are to Head of Textile Technology Department, IIT Delhi, for providing me the necessary facilities. The help rendered by Dr. O.P. Behl and Dr. L.M. Manocha of National Physical Laboratory, Delhi during the initial phase of this work is gratefully acknowledged. The fracture toughness studies were carried out in Applied Mechanics Department, IIT Delhi, and I am thankful to Dr. R.K. Pandey and Dr. A.N. Kumar for providing me the laboratory facilities for carrying out this work.

I greatly appreciate my friends and colleagues for their benevolent cooperation. My inestimable thanks to Dr. V.K. Dhar, Mr. A.C. Vashista and Mr. Vijay Harwalkar for helping me during the compilation of the thesis. I am greatly indebted to my family members, especially to my father for his patience and understanding during the course of this study.

Last, but not least I express my sincere thanks to Mr. K.G. Padam for tracing the figures and Mr. Rajesh K. Arora for typing this thesis.

(R.G. RAJ)

ABSTRACT

The present work was undertaken to develop high temperature epoxy resin formulations based on diglycidyl ether of bisphenol-A (DGEBA) and aromatic diamines. Eight amines having different molecular weight and structure were used for curing DGEBA. Triethylene tetraamine (TETA) 4,4'-diaminodiphenyl sulfone (DDS), 4,4'-diaminodiphenyl methane were the commercially available materials. 9,9-bis(p-aminophenyl) fluorene (BAF) and four imide-amine hardeners were prepared in the laboratory. For this purpose one mole of pyromellitic dianhydride was reacted with two moles of p-phenylene diamine or 4,4'-diaminodiphenyl methane or 4,4'-diaminodiphenyl sulfone in dimethylformamide. The imide-amines obtained by cyclisation with acetic anhydride and sodium acetate have been designated as I-P, I-D, I-E and I-S respectively. Thermal cyclisation of amic acid was also investigated by using a low molecular weight model compound (N-phenyl maleamic acid). However this method was not used for preparing imide-amine hardeners because of high temperatures involved.

The imide-amines were characterised by ir spectroscopy and by determination of amine equivalent weight using acetylation method.

Curing studies of the DGEBA (Araldite LY 556, Ciba-Geigy, India Ltd.) were carried out with various amines using stoichiometric proportions of amines and 25, 50 and 75% of the stoichiometric amount of amines. Differential thermal analysis (DTA) and differential scanning calorimetry (DSC) were used to study the curing behaviour. From the exotherm, heat of polymerization (Δ_H) was determined. It was noticed that the position of exotherm as well as Δ_H depended on the structure of amines. An increase in the molecular weight of amines resulted in a decrease in Δ_H values.

Relative thermal stability of epoxy resin with different hardeners was studied by thermogravimetric analysis (TGA). The char yield (Y_c) at 500°C (in nitrogen atmosphere) was found to be higher in I-D, I-E and I-S cured resins. From the knowledge of various procedural decomposition temperatures and char yields it could be concluded that the thermal stability of cured epoxy resins improved by using imide-amines as hardeners.

Laminates were fabricated with glassfiber (E-glass woven rovings, plain weave, 280 g/m^2, Pilkington fibreglass India, Ltd.,) using compression moulding technique. Glass-fiber content of laminates was in the range of 62-66% by weight. Effect of curing and post-curing studies of laminates (fabricated using epoxy with different hardeners) on mechanical properties (tensile, flexural, short-beam shear and Izod-impact strength) was investigated. The maximum tensile strength (474 MN/m^2) was obtained in I-S cured laminates. The flexural strength of I-E and I-S cured laminates were 589 MN/m^2 and 616 MN/m^2 respectively. Fracture toughness of the laminates using single-edge notch type was carried out. The critical stress-intensity factor (K_{ic}), and critical strain energy release rate (G_{ic}) were determined. The DADPM and I-S cured laminates produced tougher composites. Fracture surface of the laminates was examined by scanning electron microscope (SEM). Failure by fiber pullout, and debonding was observed.

Laminates were aged at 100 and 200°C for various times and the mechanical properties were studied at room temperature. I-S cured laminates retained 75% of original tensile strength compared to 51% retention in DDS cured laminates after aging at 200°C for 200 hours. Effect of boiling water treatment and photodegradation on the mechanical properties of the laminates were investigated. Significant loss in mechanical properties was observed on boiling water treatment of laminates.

Unidirectional glassfiber-reinforced composites were fabricated with LY 556 and E-glass CF rovings. Commercial hardeners were used to study the effect of heat-up time and curing and post-curing studies on mechanical properties of the composites. Failure modes of the laminates were studied by SEM.

The post-cured (TETA) HY 951 laminates after impact test showed more of tensile failure with less fiber pullout. With higher curing times the fracture mode of flexural loaded (DADPM) HT 972 laminates changed to mixed tensile cum shear type failure.

CONTENTS