BLEND OF BIODEGRADABLE THERMOPLASTICS WITH LIGNIN ESTERS

by

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Thesis submitted to the Graduate Faculty of the Virginia Polytechnic Institute and State University

in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

in

Wood Science & Forest Products

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22nd April, 1998
Blacksburg, VA, U.S.A.
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ABSTRACT

Thermoplastic blends of several biodegradable polymers with lignin (L) and lignin esters were prepared by solvent casting and melt processing. Among the biodegradable thermoplastics were cellulose acetate butyrate (CAB), poly-hydroxybutyrate (PHB), poly-hydroxybutyrate-co-valerate (PHBV), and a starch-caprolactone blend (SCL). Lignin esters included acetate (LA), butyrate (LB), hexanoate (LH), and laurate (LL). Blend characteristics were analyzed in terms of thermal and mechanical properties. The results indicate widely different levels of interaction between two polymer constituents. Melt blended samples of CAB/LA and CAB/LB were compatible on a 15-30 nm scale when probed by dynamic mechanical thermal analysis, and the glass transition temperatures of the blends followed Fox equation, whereas those of CAB/LH and CAB/LL showed distinct broad transitions on the same scale. Melt blending produced well dispersed phases whereas large phase separation evolved out of solvent casting. Crystallinity and melting points of PHB and PHBV were affected by the incorporation of lignin component, revealing some interaction between the blend constituents. Blends of SCL with L and LB revealed significant effect on crystallinity and melting temperatures of poly-caprolactone component, revealing polymer-polymer interaction between SCL and lignin components. An increased degree of crystallinity was observed in the case of higher-$T_g$ L compared to lower $T_g$ LB. Improvement in modulus (and in some cases strength also) was observed in almost all blends types due to the glassy reinforcing behavior of lignin.
Learning is such a process that it is not possible to tell exactly how one acquired a given body of knowledge. This thesis is dedicated to all those who taught me and helped me to acquire that knowledge over the years.
ACKNOWLEDGEMENTS

I acknowledge my foremost indebtedness to Prof. Wolfgang G. Glasser for his invaluable guidance and support, and for providing me the opportunity to pursue my graduate studies at Virginia Tech. I would also like to extend my gratitude to Prof. Herve' Marand and my committee members, Dr. Charles E. Frazier and Dr. Richey M. Davis, for their comments and suggestions. I like to acknowledge the help from Prof. D. G. Baird and Dr. Mike McLeod for the viscosity measurements.

I am thankful to Eastman Chemical Company (Kingsport TN, U.S.A.) and Prof. Ramani Narayan (Michigan State University) for the supply of materials for the research.

I express many thanks to all the members in our group for their technical, scientific and administrative help and the pleasant working atmosphere. In particular, I would like to thank Dr. Rajesh K. Jain for his help at each step of my research work. I am also thankful to Ulli Becker and Jason Todd for many valuable discussions and helpful suggestions. Without their help, it would have been difficult for me to complete my work.

I also like to acknowledge the assistance provided by Jody Jervis with the molecular weight determinations and Kathy with the electron microscope.

Last, but not the least, I wish to thank my parents, my brother, and sister-in-law for all their motivation, inspiration and cooperation all through my life.
PREFACE

Recently, there is great interest and awareness in ‘environmentally friendly’ polymers for various applications ranging from packaging materials to automobile interiors. This research is aimed particularly towards developing materials for biodegradable composites and related applications by modifying existing biodegradable polymers. This study deals with: a) an investigation of the possible factors affecting miscibility/compatibility of polymer blends of biodegradable polymers such as cellulose ester (CAB), polyhydroxy-alkanoates (PHA’s), and a starch-caprolactone blend (SCL, tradename “Envar”) with lignin esters having varying lengths of side groups; b) the influence of the lignin component in the blend; c) effect of lignin ester component on crystallization of PHA’s and SCL.

Concepts of biodegradation and compatibility are introduced along with other relevant theories in chapter-I. Chapter-II deals with the synthetic and analytical details of lignin esters used as blend components with the biodegradable polymers. Chapters-III, IV and V are concerned with the detailed studies of blends of CAB, PHA’s and SCL respectively. Each chapter of the thesis (except chapter-I) has its own abstract, experimental sections, discussions and references followed by illustrations. Due to similarity in the topics covered in each chapter, some repetition of material could not be avoided. Apologies are made for these inconveniences and other mistakes that might have avoided my sight.
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<td>$\text{DP}_n$</td>
<td>Degree of polymerization</td>
</tr>
<tr>
<td>DS</td>
<td>Degree of substitution</td>
</tr>
<tr>
<td>$\Delta H_m$</td>
<td>Enthalpy of fusion (J/gm)</td>
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<td>$\Delta H_c$</td>
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</tr>
<tr>
<td>$M_n$</td>
<td>Number average molecular weight (Daltons)</td>
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<tr>
<td>$M_w$</td>
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