

1998

Effect of Recycled Fiber, Compatibilizer and Pre-Formed Fiber Handsheet on the Performance of Wood-Polyolefin Composites.

Chin-yin S. Hwang

Louisiana State University and Agricultural & Mechanical College

Follow this and additional works at: https://digitalcommons.lsu.edu/gradschool_disstheses

Recommended Citation

Hwang, Chin-yin S., "Effect of Recycled Fiber, Compatibilizer and Pre-Formed Fiber Handsheet on the Performance of Wood-Polyolefin Composites." (1998). *LSU Historical Dissertations and Theses*. 6679. https://digitalcommons.lsu.edu/gradschool_disstheses/6679

This Dissertation is brought to you for free and open access by the Graduate School at LSU Digital Commons. It has been accepted for inclusion in LSU Historical Dissertations and Theses by an authorized administrator of LSU Digital Commons. For more information, please contact gradetd@lsu.edu.

INFORMATION TO USERS

This manuscript has been reproduced from the microfilm master. UMI films the text directly from the original or copy submitted. Thus, some thesis and dissertation copies are in typewriter face, while others may be from any type of computer printer.

The quality of this reproduction is dependent upon the quality of the copy submitted. Broken or indistinct print, colored or poor quality illustrations and photographs, print bleedthrough, substandard margins, and improper alignment can adversely affect reproduction.

In the unlikely event that the author did not send UMI a complete manuscript and there are missing pages, these will be noted. Also, if unauthorized copyright material had to be removed, a note will indicate the deletion.

Oversize materials (e.g., maps, drawings, charts) are reproduced by sectioning the original, beginning at the upper left-hand corner and continuing from left to right in equal sections with small overlaps. Each original is also photographed in one exposure and is included in reduced form at the back of the book.

Photographs included in the original manuscript have been reproduced xerographically in this copy. Higher quality 6" x 9" black and white photographic prints are available for any photographs or illustrations appearing in this copy for an additional charge. Contact UMI directly to order.

UMI

A Bell & Howell Information Company
300 North Zeeb Road, Ann Arbor MI 48106-1346 USA
313/761-4700 800/521-0600

**EFFECT OF RECYCLED FIBER, COMPATIBILIZER AND
PRE-FORMED FIBER HANDSHEET ON THE PERFORMANCE OF
WOOD-POLYOLEFIN COMPOSITES**

A Dissertation

**Submitted to the Graduate Faculty of the
Louisiana State University and
Agricultural and Mechanical College
in partial fulfillment of the
requirement for the degree of
Doctor of Philosophy**

in

The School of Forestry, Wildlife, and Fisheries

by

Chin-Yin Hwang

B.S., National Chung-Hsing University, Taiwan, ROC, 1976

M.S., National Taiwan University, Taiwan, ROC, 1986

May 1998

UMI Number: 9836879

UMI Microform 9836879
Copyright 1998, by UMI Company. All rights reserved.

**This microform edition is protected against unauthorized
copying under Title 17, United States Code.**

UMI
300 North Zeeb Road
Ann Arbor, MI 48103

ACKNOWLEDGEMENTS

I am greatly indebted to Dr. Elvin T. Choong and Dr. Chung-Yun Hse for their support, advice, and guidance through this project. Special appreciation is also due to Dr. W. Ramsay Smith, Dr. William Daly, and Dr. Julia Denslow for their critical review and valuable suggestions. I am grateful to Dr. Vernon Wright for offering invaluable statistical consultation.

I also wish to express my gratitude to Dale Huntsberry at the USDA Forest Service, Southern Research Station for his technical assistance; Dr. Richard Vlosky, JoAnn Doucet and Pat LeFeaux at the Louisiana Forest Products Laboratory for their logistical assistance. I also acknowledge Dr. Neils DeHoop, Belinda Fuller, Robert Davidge, and Bernard Wright for their proofreading the manuscript.

Special thanks are extended to the Louisiana Forest Products Laboratory for offering logistical support, and to the National Science Council and Taiwan Forestry Research Institute, Taiwan, ROC for providing an opportunity for my doctoral study.

For donation of test materials, sincere thanks are due to Exxon Chemical Co., Eastman Chemical Co., Dow Plastics, and Bordon Chemical Inc. Without the help from industry, this project could not have been completed.

Finally, I would like to acknowledge my husband Chau-Chin Lin and son Monroe Lin, who have been secondary to my research for five years. Their encouragement, support, patience, and love give me strength to accomplish this

project. This dissertation is dedicated to my mother and my late father who have always had confidence in my potential and encouraged me to become a person of broad humanity.

TABLE OF CONTENTS

ACKNOWLEDGMENTS	ii
LIST OF TABLES	vii
LIST OF FIGURES	x
LIST OF ABBREVIATIONS.....	xiv
ABSTRACT	xvi
CHAPTER 1	
INTRODUCTION	1
1.1. LITERATURE CITED	4
CHAPTER 2	
EFFECTS OF CONFIGURATION AND PROCESSING VARIABLES ON THE PROPERTIES OF WOOD FIBER-POLYETHYLENE COMPOSITES	
2.1. INTRODUCTION	5
2.2. MATERIALS AND METHODS	10
2.2.1. PART I. Characterization of Wood Fibers from Four Sources	10
2.2.1.1. Materials	10
2.2.1.2. Methods	10
2.2.2. PART II. Effects of Recycled Materials on the Properties of Wood Fiber-Polyethylene Composites	11
2.2.2.1. Materials	11
2.2.2.2. Methods	12
2.3. RESULTS AND DISCUSSION	18
2.3.1. Part I. Characterization of Wood Fibers from Four Sources	18
2.3.1.1. Chemical Analysis	18
2.3.1.2. Fiber Length Measurement	20
2.3.2. Part II. Effects of Recycled Materials on the Properties of Wood Fiber-Polyethylene Composites	28
2.3.2.1. Effect of Recycled Fiber on the Properties of Hardboards	28
2.3.2.2. Effect of Configuration on the Properties of Wood Fiber-Polyethylene Composites	44
2.3.2.3. Effects of Some Processing Factors on the Properties of Wood Fiber-Polyethylene Composites	53
2.4. CONCLUSIONS	70
2.5. LITERATURE CITED	72

CHAPTER 3	
EFFECT OF COMPATIBILIZER ON THE WETTABILITY AND INTERFACE ADHESION OF WOOD-PLASTIC COMPOSITES	78
3.1. INTRODUCTION	78
3.2. LITERATURE REVIEW	81
3.2.1. Effect of Maleated Polypropylene	81
3.2.2. Wettability of Wood	83
3.2.3. Interfacial Shear Strength Between Wood and Plastics	85
3.3. MATERIALS AND METHODS	88
3.3.1. Part I. Wettability of MAPP-Treated Birch Plywood and Polyolefin Polymers	88
3.3.1.1. Materials	88
3.3.1.2. Methods	89
3.3.2. Part II. Bulk Interfacial Shear Strength Between Birch Wood and Polyolefin Polymers	92
3.3.2.1. Materials	92
3.3.2.2. Methods	93
3.4. RESULTS AND DISCUSSION	96
3.4.1. Part I. Wettability of MAPP-Treated Birch Plywood and Polyolefin Polymers	96
3.4.1.1. Contact angle in MAPP-Treated Birch Plywood ...	96
3.4.1.2. Contact angle Measurement for Polyolefin Polymers	108
3.4.2. Part II. Bulk Interfacial Shear Strength Between Birch Wood and Polyolefin Polymers	119
3.4.2.1. Interfacial Shear Strength	119
3.4.2.2. Failure Mode and Air Bubble Occurrence	136
3.5. CONCLUSIONS	141
3.6. LITERATURE CITED	143
CHAPTER 4	
DEVELOPMENT OF WOOD FIBER-POLYPROPYLENE LAMINATES WITH DIFFERENT CONFIGURATIONS	149
4.1. INTRODUCTION	149
4.2. MATERIALS AND METHODS	153
4.2.1. Materials	154
4.2.1.1. Wood Fiber	154
4.2.1.2. Plastic Films	154
4.2.2. Methods	154
4.2.2.1. Hand Sheet Preparation	154
4.2.2.2. Wood Fiber-Polypropylene Laminate Manufacture	157
4.2.2.3. Measurement of Handsheet Porosity	157
4.2.2.4. Tensile Strength Measurement	158
4.2.2.5. Dimensional Stability	158
4.2.2.6. Scanning Electronic Microscopy (SEM)	159

4.3. RESULTS AND DISCUSSION	159
4.3.1. Basic Handsheet Properties	159
4.3.2. Mechanical and Physical Properties of Wood Fiber- Polypropylene Laminates	166
4.3.2.1. Tensile Properties	167
4.3.2.2. Dimensional Stability	175
4.3.2.3. Factors Affecting Laminate Properties	181
4.3.3. SEM Observation of Wood Fiber-Polypropylene Laminates	186
4.4. CONCLUSIONS	187
4.5. LITERATURE CITED	188
CHAPTER 5	
CONCLUSIONS	191
VITA	194

LIST OF TABLES

Table	Page
2.1. Size distribution of polyethylene powder	13
2.2. Hot pressing schedules for manufacturing of wood fiber-plastic composites	13
2.3. Processing variables in investigating properties of wood fiber-plastic composites	18
2.4. Chemical compositions of four wood fiber types	20
2.5. Average weighted fiber lengths of four wood fiber types at five disintegrating revolutions	22
2.6. Two-way ANOVA with contrasts for effects of fiber type (FT) and disintegrating revolution (DR) on weighted fiber length	22
2.7. Physical and mechanical properties of fiberboards made from various fiber compositions	28
2.8. Contrast analysis of physical and mechanical properties of fiberboards with different fiber compositions	30
2.9. Statistics and estimated parameters for linear regression analysis of fiberboard properties on virgin fiber ratio as denoted by model $Y = \sum b_i X_i$	31
2.10. Contact angle of three wetting liquids for fiberboards made from different virgin fiber and recycled fiber ratios	40
2.11. ANOVA of contact angle for fiberboards with various fiber compositions	40
2.12. Physical and mechanical properties of wood fiber-polyethylene composites with different configurations	45
2.13. Physical and mechanical properties of wood fiber-polyethylene composites with VIR:OCC1:PE=4:4:2 at various processing conditions	55
2.14. Three-way ANOVA of some processing factors on properties of wood fiber-polyethylene composites with VIR:OCC1:PE=4:4:2	57
2.15. Contact angle of wood fiber-polyethylene composites with VIR:OCC1:PE= 4:4:2 at different processing conditions	65

2.16	ANOVA of some processing factors on contact angle of wood fiber-polyethylene composites with VIR:OCC1:PE=4:4:2	66
3.1.	Basic physical properties for plastics	90
3.2.	Experiment design for measuring contact angle of birch plywood	91
3.3.	Experiment design for testing bulk interfacial shear strength of birch dowel and polyolefin polymers	91
3.4.	Contact angle for birch plywood treated with various E-43 concentrations (CON).....	95
3.5.	ANOVA results on effects of drying method (DRY), coupling agent concentration (CON), and liquid type (LIQUID) on contact angle measurement of birch plywood	99
3.6.	Statistics and coefficients for polynomial regression analysis of E-43 solid content (X) and contact angle (Y) with different liquid types and drying conditions as denoted by mode $Y=b_0+b_1X+b_2X^2$..	106
3.7.	Contact angle of E-43 with various concentrations on polyolefin surfaces	109
3.8.	Contact angle of five wetting liquids on polyolefin surfaces	109
3.9.	ANOVA results of effects of plastic type (PT) and E-43 concentration (CON) on contact angle of plastics	110
3.10.	ANOVA results of effects of plastic type (PT) and wetting liquid (LIQUID) on the contact angle of plastics	110
3.11.	Interfacial shear strength between plastic matrices and birch wood dowels treated with various E-43 concentration and adhesives	121
3.12.	ANOVA based on effects of plastic type (PT) , adhesive type (AT), and E-43 concentration (CON) on interfacial shear strength of birch dowels and plastic matrices	122
3.13.	Solid E-43 content (SCC) and solid resin content (SRC) for wood dowels and plastic matrices with plastic type combined	131
3.14.	Statistics and coefficients for polynomial regression analysis of SCC (X) and ISS (Y) with different plastic and adhesive types as denoted by model $Y=b_0+b_1X+b_2X^2$	134
3.15.	Frequencies of failure mode on the interface of wood-plastic model composites	135
4.1.	Basic handsheet properties for each treatment	156
4.2.	Mechanical and physical properties of different fiber content (NOMFC) and layer (LAYER) combinations for wood fiber-polypropylene laminates with a target specific gravity of 1.0	165

4.3. Analysis of variance of fiber content (NOMFC) and layer (LAYER) effects on mechanical and physical properties of wood fiber-plastic laminates	171
4.4. Results of regression analysis of physical and mechanical properties (Y) with respect to independent variables of specific gravity (SPGR) and actual fiber content (ACTFC) using model $\text{Ln}(Y)=b_0+b_1\text{Ln}(X)$	182

LIST OF FIGURES

Figure	Page
2.1. Schematic presentation of laboratory-scaled propeller glue spreader	14
2.2. Schematic presentation of laboratory-scaled automatic air-forming system	15
2.3. Experimental procedures in manufacturing wood-polyolefin composites	16
2.4. Fraction of each mesh class for OCC1 fiber type at various disintegrating revolutions	23
2.5. Fraction of each mesh class for OCC2 fiber type at various disintegrating revolutions	24
2.6. Fraction of each mesh class for ROP fiber type at various disintegrating revolutions	25
2.7. Fraction of each mesh class for VIR fiber type at various disintegrating revolutions	26
2.8. Average fiber length of four fiber types with combined disintegrating revolutions	27
2.9. Correlation between virgin fiber ratio and panel density of fiberboard	33
2.10. Relationship between virgin fiber ratio and internal bond strength of fiberboard	34
2.11. Correlation between virgin fiber ratio and bending modulus of rupture of fiberboard	35
2.12. Correlation between virgin fiber ratio and bending modulus of elasticity of fiberboard	36
2.13. Correlation between virgin fiber ratio and water absorption of fiberboard	37
2.14. Correlation between virgin fiber ratio and thickness swelling of fiberboard	38
2.15. SEM micrograph of fiberboard made from 100% virgin fiber (A) and 100% recycled old corrugated cardboard (B)	41
2.16. Contact angle of fiberboard measured by three wetting liquids	42
2.17. Effect of wetting liquid and virgin fiber ratio on contact angle of fiberboard	43

2.18.	Internal bond strength of wood fiber-polyethylene composite with various configurations	46
2.19.	Modulus of rupture for wood fiber-polyethylene composites with various configurations	47
2.20.	Modulus of elasticity for wood fiber-polyethylene composites with various configurations	48
2.21.	Water absorption for wood fiber-polyethylene composites with various configurations	49
2.22.	Thickness swelling for wood fiber-polyethylene composites with various configurations	50
2.23.	Effect of hot press temperature on internal bond strength of wood fiber-polyethylene composites	59
2.24.	Effect of panel density on internal bond strength of wood fiber-polyethylene composites	60
2.25.	Modulus of rupture for wood fiber-polyethylene composites with virgin fiber : recycled fiber : PE= 4:4:2 at two panel density levels	61
2.26.	Modulus of elasticity for wood fiber-polyethylene composites with virgin fiber : recycled fiber : PE=4:2:2 at two panel density levels	62
2.27.	Effect of panel density on water absorption of wood fiber-polyethylene composites	63
2.28.	Effect of hot press temperature on contact angle of wood fiber-polyethylene composites	67
2.29.	Effect of hot press time on contact angle of wood fiber-polyethylene composites	68
2.30.	Effect of wetting liquid on contact angle of wood fiber-polyethylene composites	69
3.1.	Schematic diagram of the interfacial shear strength test procedure (A) test specimen, (B) specimen and fixture layout	94
3.2.	Contact angle of E-43 treated birch plywood by drying method	100
3.3.	Contact angle of E-43 treated birch plywood by E-43 concentration level	101
3.4.	Contact angle of four wetting liquids on E-43 treated birch plywood	102
3.5.	Contact angle of four wetting liquids on air-dried birch plywood surface treated with various E-43 concentrations	103
3.6.	Contact angle of four wetting liquids on oven-dried birch plywood surface treated various E-43 concentrations	104

3.7.	Contact angle of E-43 on birch plywood coated with four adhesives	107
3.8.	Contact angle of E-43 on three polyolefin surfaces	111
3.9.	Dependence of contact angle of E-43 on E-43 concentration for three polyolefins	112
3.10.	Average contact angle of four wetting liquids on three polyolefin surfaces	114
3.11.	Average contact angle of three polyolefin polymers measured by four wetting liquids	115
3.12.	Contact angle of four wetting liquids on three polyolefin surfaces ..	116
3.13.	Contact angle of four adhesives on E-43 treated polypropylene surface	117
3.14.	Interfacial shear strength of wood-polyolefin model composites by plastic type	123
3.15.	Interfacial shear strength of wood-polyolefin model composites by adhesive type	124
3.16.	Interfacial shear strength of wood-polyolefin model composites by E-43 concentration	125
3.17.	Dependence of interfacial shear strength on E-43 concentration and adhesive type for wood-LDPE model composites	126
3.18.	Dependence of interfacial shear strength on E-43 concentration and adhesive type for wood-LLDPE model composites	129
3.19.	Dependence of interfacial shear strength on E-43 concentration and adhesive type for wood-PP model composites	130
3.20.	Percentage of adhesive failure in three wood-polyolefin model composites	138
3.21.	Percentage of adhesive failure in wood-polyolefin model composites by adhesive type	139
3.22.	Percentage of adhesive failure in wood-polyolefin model composites by E-43 concentration	140
4.1.	Typical hot pressing spectra in fabricating fiber-polypropylene laminates	160
4.2.	Dependence of handsheet thickness on grammage	161
4.3.	Micrograph of handsheet porosity for treatment with 50% fiber content	162

4.4.	Micrograph of handsheet porosity for treatment with 60% fiber content	163
4.5.	Micrograph of handsheet porosity for treatment with 70% fiber content	164
4.6.	Effect of plastic layer on tensile strength of fiber-polypropylene laminates	172
4.7.	Interaction of fiber content and plastic layer on elongation of fiber-polypropylene laminates	173
4.8.	Interaction of fiber content and plastic layer on tensile modulus of fiber-polypropylene laminates	174
4.9.	Effect of fiber content on dimensional stability of fiber-polypropylene laminates	176
4.10.	Effect of plastic layer on dimensional stability of fiber-polypropylene laminates	177
4.11.	Interaction of fiber content and plastic layer on water absorption of fiber-polypropylene laminates	178
4.12.	SEM micrograph of tensile fracture in fiber-polypropylene laminate	183
4.13.	SEM micrograph of inner (A) and outer (B) surfaces for treatment with NOMFC=50 and LAYER=1	184
4.14.	SEM micrograph of inner (A) and outer (B) surfaces for treatment with NOMFC=70 and LAYER=3	185

LIST OF ABBREVIATIONS

ACTFC	Actual fiber content
ANOVA	Analysis of variance
AT	Adhesive type
CON	E-43 concentration
DR	Disintegrating revolution
DRY	Drying method
E-43	Epolene E-43
ELONG	Elongation at break
FT	Fiber type
IB	Internal bond strength
ISO	Isocyanate
ISS	Interfacial shear strength
LAYER	Layer of PP films between two handsheets
LE	Linear expansion
LDPE	Low density polyethylene
LIQUID	Wetting liquid
LLDPE	Linear low density polyethylene
MAPP	Maleated polypropylene
MOE _b	Bending modulus of elasticity
MOE _t	Tensile modulus of elasticity
MOR _b	Bending modulus of rupture
MOR _t	Tensile modulus of rupture
NO	No adhesive (control)
NOMFC	Nominal fiber content
OCC	Old corrugated cardboard
Panel A	Panel with 100% virgin fiber 0% recycled fiber
Panel B	Panel with 80% virgin fiber 20% recycled fiber
Panel C	Panel with 60% virgin fiber 40% recycled fiber
Panel D	Panel with 40% virgin fiber 60% recycled fiber
Panel E	Panel with 20% virgin fiber 80% recycled fiber
Panel F	Panel with 0% virgin fiber 100% recycled fiber
Panel G	Panel with homogeneous structure and VIR:OCC1:PE=80:0:20
Panel H	Panel with homogeneous structure and VIR:OCC1:PE=0:80:20
Panel I	Panel with homogeneous structure and VIR:OCC1:PE=40:40:20
Panel J	Panel with layered structure and VIR:OCC1:PE=60:40:0

Panel K	Panel with layered structure and VIR:OCC1:PE=40:40:20
Panel L	Panel fabricated at 350°F for 5 min with density 50 pcf
Panel M	Panel fabricated at 350°F for 5 min with density 60 pcf
Panel N	Panel fabricated at 350°F for 7.5 min with density 50 pcf
Panel O	Panel fabricated at 350°F for 7.5 min with density 60 pcf
Panel P	Panel fabricated at 400°F for 5 min with density 50 pcf
Panel Q	Panel fabricated at 400°F for 5 min with density 60 pcf
Panel R	Panel fabricated at 400°F for 7.5 min with density 50 pcf
Panel S	Panel fabricated at 400°F for 7.5 min with density 60 pcf
Panel T	Panel fabricated at 450°F for 5 min with density 50 pcf
Panel U	Panel fabricated at 450°F for 5 min with density 60 pcf
Panel V	Panel fabricated at 450°F for 7.5 min with density 50 pcf
Panel W	Panel fabricated at 450°F for 7.5 min with density 60 pcf
PCF	Panel density in pound per cubic foot
PE	Polyethylene
PF	Phenol formaldehyde
PP	Polypropylene
PT	Plastic type
ROP	Recycled office paper
SCC	Solid E-43 content
SPGR	Specific gravity
SRC	Solid resin content
TEMP	Hot pressing temperature
TIME	Hot pressing time
TS	Thickness swelling
UF	Urea formaldehyde resin
VIR	Virgin fiber
VFR	Virgin fiber ratio

ABSTRACT

Effects of recycled materials on the properties of hardboards, of compatibilizer on the wettability and interfacial shear strength of wood and plastics, and of handsheet configuration on the properties of wood fiber-polyolefin laminates were studied. Recycled fibers from three sources showed considerably higher holo- and alpha-cellulose contents, lower extractive and lignin contents, and shorter fiber length than virgin southern pine fiber. Replacing virgin fiber with recycled fiber adversely affected mechanical properties, dimensional stability and wettability of fiberboards. Using layered structures while substituting 20% virgin fiber with polyethylene appreciably increased dimensional stability. Internal bond strength of hardboards made from virgin fiber, recycled fiber and polyethylene was affected by both hot press temperature and panel density, whereas bending properties and water absorption were affected only by panel density. Contact angles on birch plywood surfaces increased upon addition of maleated propylene (E-43) and decreased with increasing E-43 concentration. For E-43-treated wood surfaces, contact angles among the four wetting liquids were in the order of phenol resin > urea resin > isocyanate > water. Contact angles on polyolefin polymer surfaces followed the sequences of polypropylene > linear low-density polyethylene > low-density polyethylene. Interfacial shear strength was highest in polypropylene and lowest in low density polyethylene. The effect of E-43 was most pronounced in polypropylene, and less so in low-density polyethylene and linear low-density polyethylene.

Increasing E-43 concentration beyond a certain extent decreased interfacial shear strength. Laminates made by interleaving Kraft pulp handsheets and polypropylene films exhibited comparable ultimate tensile strength with natural fiber-reinforced plastics. Effects of fiber content and layer (number of plastic films between two consecutive handsheets) were significant on the tensile properties of wood fiber-polypropylene laminates. Laminates made from one layer showed better ultimate tensile strength, but the interaction between fiber content and layer offset one another on the modulus of elasticity. Increasing fiber content and number of layer decreased dimensional stability. With proper control of plastic layer between two handsheets, wood fiber-polypropylene laminates with as high as 70% fiber loading can be fabricated with favorable mechanical properties.

CHAPTER 1

INTRODUCTION

The rapidly changing economic and environmental needs of today's society are putting ever-increasing pressures on the forest products industry to increase the efficient use of the wood fiber resource as well as to use environmentally friendly processes and technologies. Meanwhile, the disposal of solid waste has become a worldwide problem which also puts pressure on the need to recycle valuable resources from wastes. McKeever and coworkers (McKeever 1995, McKeever *et al.* 1995) identified three major sources of wood waste in the United States: (1) municipal solid waste (MSW), (2) new construction and demolition wastes, and (3) primary timber processing mill residues. In 1993, the total amounts of wood wastes and paper products generated under these three categories summed up to 83.0, 29.2, and 74.5 million tons, respectively. These figures indicated that wood wastes, so abundant in the waste stream, should be retrieved and reused.

One way to reuse wood waste is to combine it with plastic materials, which comprise 8% of the MSW (EPA 1990). Wood fibers, strong, lightweight, abundant, nonabrasive, and nonhazardous, are excellent fillers and reinforcing materials for the plastics industry; therefore, wood fibers can be combined with plastics to make wood-plastic composites.

Melt-blending and air-forming methods are commonly used to fabricate composites including wood-plastic composites. Conventional fiberboards are

made largely by the air-forming method. Since the interface of fiberboards is discontinuous, poor stress transfer between the interfaces limits the use of fiberboard as load-bearing materials. However, if recycled wood waste and plastics can be incorporated into fiberboard without significant reduction in strength properties, the resulting wood fiber-plastic composites can serve as low-cost substitutes for conventional wood composites.

In melt-blended composites, where reinforcing fillers and matrix materials form continuous interfaces, stress transfer is critical to the mechanical properties of the final product. Problems associated with these types of composites are primarily due to poor dispersion of wood fibers in the plastic matrices and poor compatibility between the hydrophilic wood fibers and the hydrophobic plastics. Several methods have been developed to enhance interfacial bonding, such as the use of compatibilizer as a processing aid. Therefore, it is important to understand the interfacial adhesion between wood and plastic.

On the other hand, the deterioration of fiber induced by repeated kneading and blending of two components at elevated temperatures and the agglomeration of wood fibers caused by interfiber hydrogen bonds should also be taken into account. A wood fiber-plastic laminate, made by interleaving treated or untreated paper sheets with plastic films, can eliminate these problems. This type of composite can be fabricated to preserve the interfiber hydrogen bonding, avoid reduction in fiber aspect ratio, enhance interface adhesion by means of interpenetration, and eventually to serve load-bearing purposes.

This research is composed of three interrelated projects to address the aforementioned issues. The major purposes of this research are presented as follows.

Chapter 2 examines the feasibility of introducing recycled fibers and plastic powder for the manufacture of conventional fiberboard. The chemical composition and fiber length distribution of recycled and virgin fibers from four sources were characterized. Furthermore, a series of air-laid composites were fabricated to investigate the effects of virgin fiber ratio, panel configuration, and some processing factors on the strength properties of wood-polyethylene composites.

Chapter 3 investigates the interfacial adhesion between wood and plastics. The effects of compatibilizer and adhesive type on the wettability of birch plywood and polyolefin polymers were studied. In addition, compatibilizer and adhesive effects on the bulk interfacial shear strength of wood and plastics were investigated, using a modified single-fiber pullout method.

Chapter 4 describes the development of wood fiber-plastic laminates with various schemes. Feasibility of such composites as load-bearing material was assessed by evaluating the effects of fiber loading on strength properties of the final products.

From air-laid composites to laminates, wood fiber-plastic composites provide a broad spectrum of end-use applications. Potential uses range from commodity products of large quantity, with minimum performance requirements,

to high value-added specialty products with rigorous performance requirements (Rowell *et al.* 1993).

1.1. LITERATURE CITED

Environment Protection Agency. 1990. Characterization of municipal solid waste in the United States: 1990 Update. Environmental Protection Agency, EPA/530-SW-90-042. Washington DC. 103p.

Rowell, R.M., T. Rials, H. Spleter, D. Luneke, R.A. Arola, R. Narayan, P. Davis, J. Simonson, T. Friberg, D. White, and R. W. Hemingway. 1993. Opportunities for composites form recycled wastewood-based resources: a problem analysis and research plan. *For. Prod. J.* 43(1):55-63.

CHAPTER 2

EFFECTS OF CONFIGURATION AND PROCESSING VARIABLES ON THE PROPERTIES OF WOOD FIBER-POLYETHYLENE COMPOSITES

2.1. INTRODUCTION

Paper and paper products account for as much as 40% of the municipal solid waste (MSW) generated annually in the United States (EPA 1990). Corrugated boxes, newspapers, and office papers are three major waste sources, the total weight generated summed up to approximately 23.1, 13.3, and 7.3 million tons respectively in 1988. The Environmental Protection Agency estimated that in 1988 nearly 45% of all corrugated boxes were recovered, while newspaper and office papers were recycled at a rate of approximately 33 and 22 percents, respectively.

In addition, vast quantities of wood waste are generated in the United States each year. McKeever and coworkers (McKeever 1995, McKeever *et al.* 1995) identified three major sources of wood waste in the United States: (1) municipal solid waste (MSW), (2) new construction and demolition wastes, and (3) primary timber processing mill residues. In 1993, the amount of wood waste, paper and paperboard, and yard trimmings constituted 7%, 38%, and 16%, respectively, of all MSW. New construction and demolition wastes totaled of 28.8 million tons. Primary timber processing mill residues, in the form of sawdust, planer shavings, and chips (Youngquist *et al.* 1990), amounted to 74.5 million tons.

These figures imply that fiber sources are abundant in the waste stream. The pulp and paper industry has been the largest user of recycled paper. The forest products industry has also endeavored to reuse these recyclable fiber resources. Some attempts have been made to reuse these materials alone to make panel products. The strength properties of the resulting products, however, either failed to meet commercial standards (Laundrie and McNatt 1975, Walters *et al.* 1977, Rowell and Harrison 1992, Krzysik *et al.* 1992, Rowell and Lange 1994) or marginally passed the standards (Krzysik *et al.* 1992). Therefore, virgin fibers were later blended with recycled fibers to improve panel strengths (Deppe 1985, Krzysik *et al.* 1992, Krzysik *et al.* 1993).

Another alternative to the reutilization of cellulosic materials from the waste stream is to combine them with nonwood materials to make composite products (Hamilton and Youngquist 1991, Rowell *et al.* 1991, Youngquist *et al.* 1992, Wenger *et al.* 1992, Rowell *et al.* 1993, Youngquist *et al.* 1993). Wood flour has been used as filler and reinforcing material in the plastic industry. Therefore, it is technically possible to use these two components to manufacture wood-plastic composites. In addition, since plastics comprise about 8 percent of the MSW (EPA 1988, EPA 1990), it is economically and environmentally desirable to use recycled plastics.

Not all thermoplastics are suitable for fabricating wood-plastic composites. The selection of plastic matrices is critical, because of the thermal degradation of hemicellulose, lignin and sometimes cellulose at elevated temperatures.

Among all thermoplastics, polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), and polystyrene (PS) are most frequently used, due to their compatibility in processing temperature with wood (Sanadi *et al.* 1997).

Depending on manufacturing method, wood-plastic composites can be divided into melt-blended composites and air-laid composites; the latter also includes nonwoven mat composites (Hamilton and Youngquist 1991, Youngquist *et al.* 1993). The major differences between these two composite products are their fiber loadings. In melt-blend composites, the maximum fiber loading is generally less than 50% of the total volume; whereas in nonwoven mat composites, fiber loading can be as high as 90%.

Numerous researchers have concentrated on developing and improving the properties of composites. The Center for Research in Pulp and Paper at the University of Quebec-Trois-Rivières in Canada has conducted studies on a number of factors affecting the performance of wood fiber-polystyrene composites. The factors investigated include wood species (Maldas *et al.* 1989a), processing variables (Sean *et al.* 1991), coupling agents (Maldas *et al.* 1988, Maldas *et al.* 1989b, Maldas and Kokta 1990a), aging and recycling conditions (Maldas and Kokta 1989, Maldas and Kokta 1990b, Maldas *et al.* 1990) and hybrid fibers (Maldas and Kokta 1990c, Maldas and Kokta 1991).

Voluminous research projects have also been conducted to study the effects of several factors affecting wood fiber-polyolefin composites. The factors studied are filler types (Klason *et al.* 1984, Woodhams *et al.* 1984, Clemons

1995, Hon 1993, Sanadi *et al.* 1994), processing variables (Takase and Shiraishi, 1989, Yam *et al.* 1990, Myers *et al.* 1993), coupling and/or dispersing agents (Dalvåg *et al.* 1985, Kishi *et al.* 1988, Raj *et al.* 1989, Raj *et al.* 1990, Raj and Kokta 1989, Gatenholm *et al.* 1993, Caulfield *et al.* 1993), and aging conditions (Raj and Kokta, 1995).

All of the above-mentioned wood-plastic composites were melt-blended composites. The overall results of these studies showed that with appropriate surface modification and proper processing conditions, wood fibers can serve as satisfactory fillers in thermoplastic materials, resulting in enhanced stiffness, comparable strength properties and, most of all, lower cost.

However, studies on nonwoven mat composites are less abundant. Research projects conducted by the USDA Forest Products Laboratory reported that combining western hemlock fibers with polyester as well as phenolic resin showed better overall mechanical and physical properties than using wood fibers and thermoplastics alone (Youngquist *et al.* 1992). Using this technique, second-generation panels made from recycled materials showed favorable strength properties (Youngquist *et al.* 1995).

Even though wood fiber-filled thermoplastics have promising features which can be tailored to a broad spectrum of applications (Youngquist *et al.* 1992), wood fibers have been used mainly as low-cost fillers, i.e., additives, to the plastic industry. A nonwoven technique is an alternative option of using wood fiber as the major component. However, it requires special machinery to

fabricate the product. Wu *et al.* (1994) fabricated an air-laid panel product to serve as a substitute for conventional particleboard which resulted in improved dimensional stability without using special machinery. They devised a series of two-component systems containing wood particles and thermoplastic additives to examine the effect of various fiber compositions on the properties of wood-plastic composites. Their results showed a reduction in bending properties (MOE and MOR) and bonding strength (IB) with increase in thermoplastic content. In comparison between the two plastic types, the reduction was less in polystyrene than in polyethylene; in comparison among the three adhesive types, the reduction was less in isocyanate than in urea-formaldehyde and phenol-formaldehyde.

Rowel *et al.* (1993) stated that more information is needed to understand the basic properties of recycled materials. Therefore, this study was undertaken to optimize the processing factors of wood fiber-plastic composites with structured experimental designs and to investigate in detail more complicated formulations. This research was divided into two parts. Part I was designed to determine chemical and physical properties of recycled wood fibers from three sources and one type of virgin fiber. Part II involved three experiments: (1) investigation on the effect of recycled fiber on properties of hardboards, (2) enhancement of properties of wood fiber-polyethylene composites by various panel configurations, and (3) determination of the effect of some processing variables on properties of wood fiber-polyethylene composites.

2.2. MATERIALS AND METHODS

2.2.1. Part I. Characterization of Wood Fibers from Four Sources

2.2.1.1. Materials

In this study, four types of fibers were used: (1) OCC1 – Old corrugated cardboard from local recycled industry, (2) OCC2 – Old corrugated cardboard, (3) ROP – Recycled office paper (mainly waste Xerox paper), and (4) VIR – Southern pine virgin fibers from fiberboard manufacturing. OCC2 and ROP were reclaimed from residue office waste at the LSU campus and were shredded to manageable sizes without deinking.

2.2.1.2. Methods

2.2.1.2.1. Chemical Analysis

Characterization of chemical composition of the four fiber sources was conducted in accordance with ASTM standards, including alcohol-benzene extractive content (ASTM D 1105-84), holocellulose content (ASTM D 1104-56), alpha-cellulose content (ASTM D 1103-60), and Klason lignin content (D 1106-84) (ASTM 1994). These tests contained replications of 24, 12, 6, and 6, respectively, in a completely randomized design. An analysis of variance was performed to compare differences in chemical components among the four fiber types.

2.2.1.2.2. Fiber Length Measurement

From each of the four fiber sources, fifteen dry fiber sample were collected. Each sample weighed 70 g, and was saturated in 2500 ml water

overnight. These samples were then further divided into three groups, each containing five samples. These five samples within one group were placed separately in a disintegrator (Hermann Manufacturing Co., rpm \approx 120) to randomly disintegrate at 100, 200, 300, 400, and 500 revolutions. From each sample, six subsamples weighing 20 g, were randomly selected. Each subsample was placed in a water classifier (The Bauer Bros. Co.), which divided the fibers into four fractions determined by mesh size of four screens, i.e., 14(A), 28(B), 48(C), and 80(D). The six subsamples, each containing four fractions, were then divided into two subgroups. Each of the fractions of one subgroup was oven-dried and weighed to estimate its weight proportion. Fractions from the other subgroup were stained by 1% safranin aq. solution, and their fiber lengths were measured using a microscopic projector (Haas Instrument Co.). A total of 9,000 fibers was measured. Based on the weight proportion and the average length of each fraction, weighted fiber lengths were calculated and compared in accordance with the principle of two-way factorial treatment arrangement.

2.2.2. Part II. Effects of Recycled Materials on the Properties of Wood Fiber-Polyethylene Composites

2.2.2.1. Material

Virgin southern pine fibers (VIR) and recycled fibers (OCC1) were obtained as described in Part I. Urea formaldehyde (Chembond UF), with 65% solid content, pH 8.0, was supplied by Neste Co. The polyethylene (PE) plastic was obtained from commercial garbage bags (about 8 mil). These bags were shredded into squares approximately 1x1 -inch with a paper cutter. The squares

were then ground to powder using a conventional disk refiner. The size distribution of PE powder is listed in Table 2.1.

2.2.2.2 Methods

2.2.2.2.1. Effect of Recycled Fiber on the Properties of Hardboards

A completely randomized design was employed to investigate the effect of adding recycled fibers to the southern pine hardboard. The amounts of recycled fibers added were 0%(A), 20%(B), 40%(C), 60%(D), 80%(E), and 100%(F) by weight.

For panels containing both virgin and recycled fibers, the two types of fibers were first hand-mixed to achieve better fiber distribution, then blended with UF resin (10% by weight) in a laboratory-scaled propeller glue spreader (Figure 2.1). The mixture was then air-laid in a specially designed automatic air-forming system (Figure 2.2). In panels containing only one type of fibers, no hand-mixing was performed.

The furnish was compressed in a hot press at 350°F for 5 minutes, according to the schedules shown in the second column of Table 2.2. A total of 18 panels with nominal dimensions of 12x12x1/4 -inch were made with three replicates for each virgin fiber level. After hot pressing, the panels were stored in the laboratory at room temperature until equilibrium, then cut into various test samples to determine their physical and mechanical properties.

Properties investigated include (1) Internal bond (IB), (2) 3-point bending modulus of rupture (MOR_b) and bending modulus of elasticity (MOE_b), (3) water

absorption after 2 (WA2) and 24 hours (WA24), (4) thickness swelling after 2 (TS2) and 24 hours (TS24), and (5) contact angle measurement. All the strength property test procedures were done in accordance with ASTM D-1037-94 (ASTM 1994). Two specimens were extracted from each panel for bending test, 3 for IB, and 1 for both WA and TS. Contact angles were measured by the direct optical method. Three wetting liquids (UF, PF, and water) were selected to determine the wettability of hardboards with various virgin fiber contents. An analysis of variance was performed to examine the influence of recycled fibers on the properties of hardboard.

Table 2.1. Size distribution of polyethylene powder.

Mesh	Average %
< 18	1.62 (0.30) ¹
18 -- 45	30.65 (2.16)
45 -- 60	21.24 (1.59)
60 -- 80	18.16 (1.43)
> 80	28.32 (2.82)

¹. Numbers in the parentheses represent standard deviations.

Table 2.2. Hot pressing schedules for manufacturing of wood fiber-plastic composites.

Pressure (pound)	----- Hot pressing time (min.) -----	
	5 min. schedule	7.5 min. schedule
250	1	2.5
150	2	2
75	1	2
25	1	1

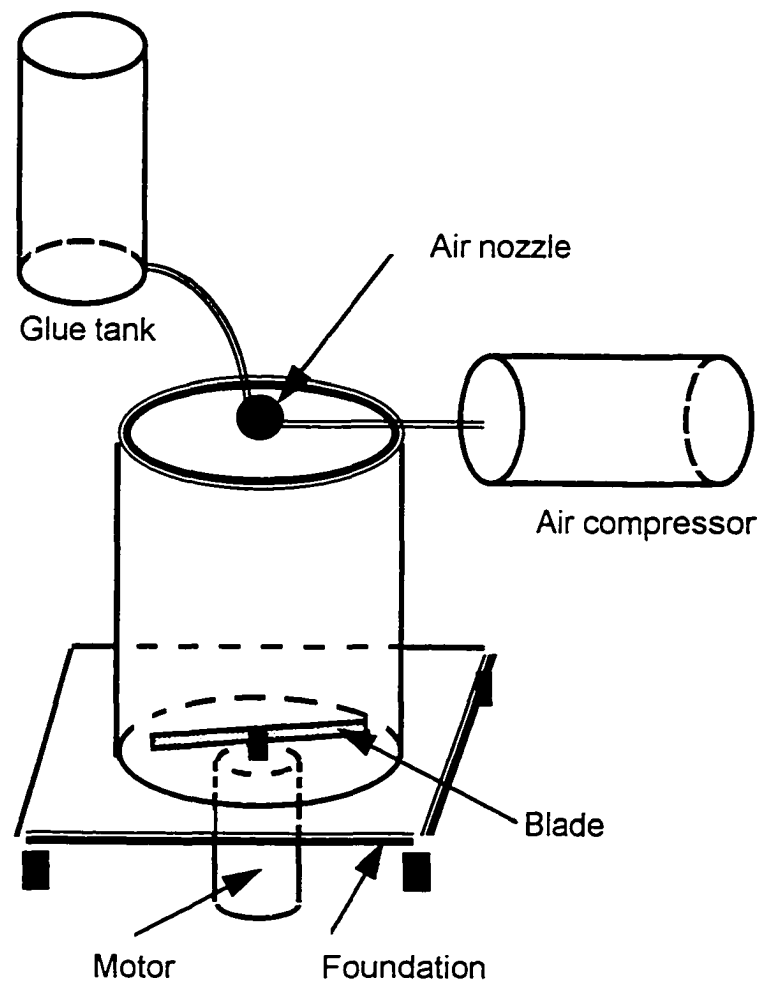


Figure 2.1. Schematic presentation of laboratory-scaled propeller glue spreader.

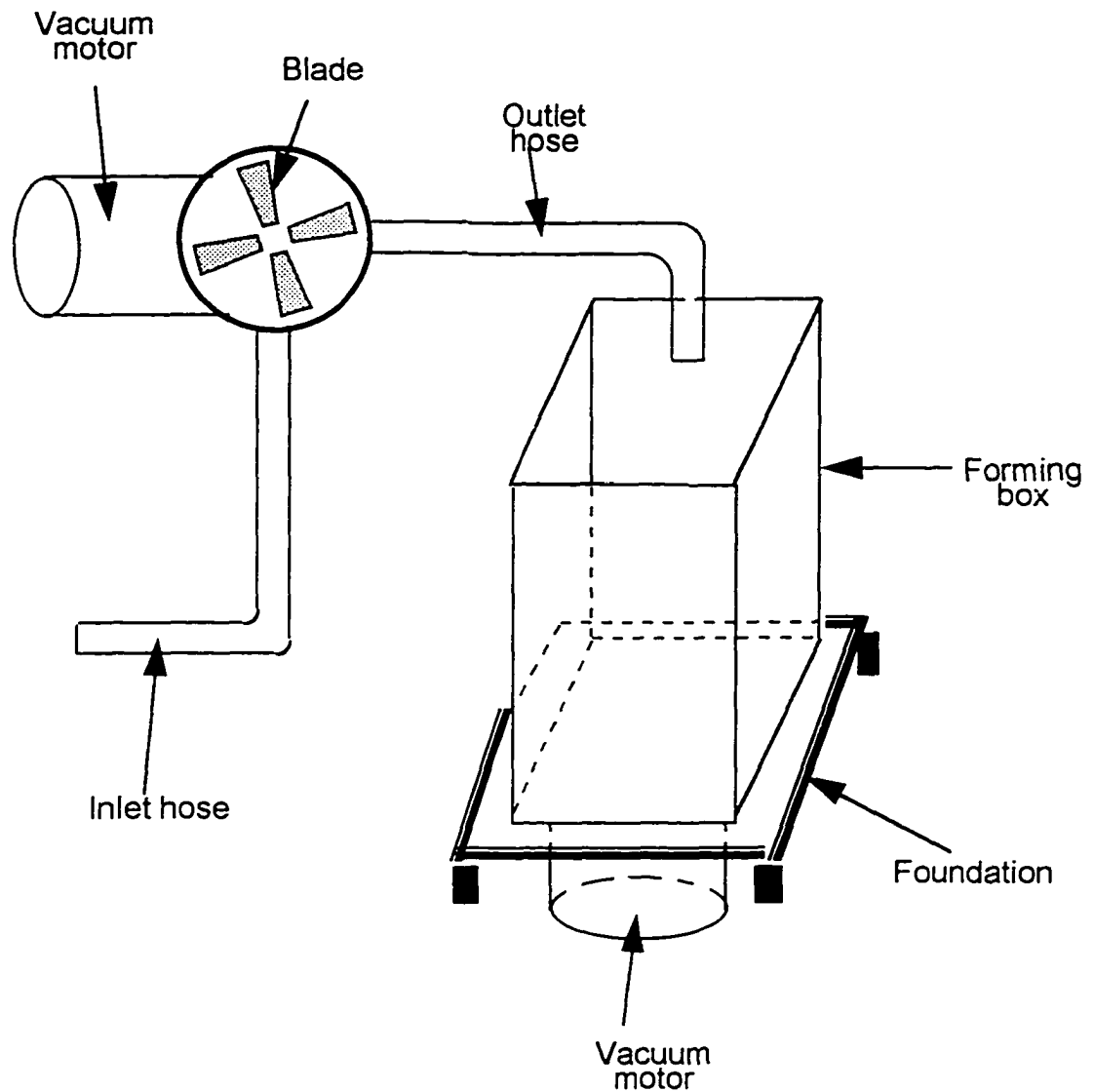


Figure 2.2. Schematic presentation of laboratory-scaled automatic air-forming system.

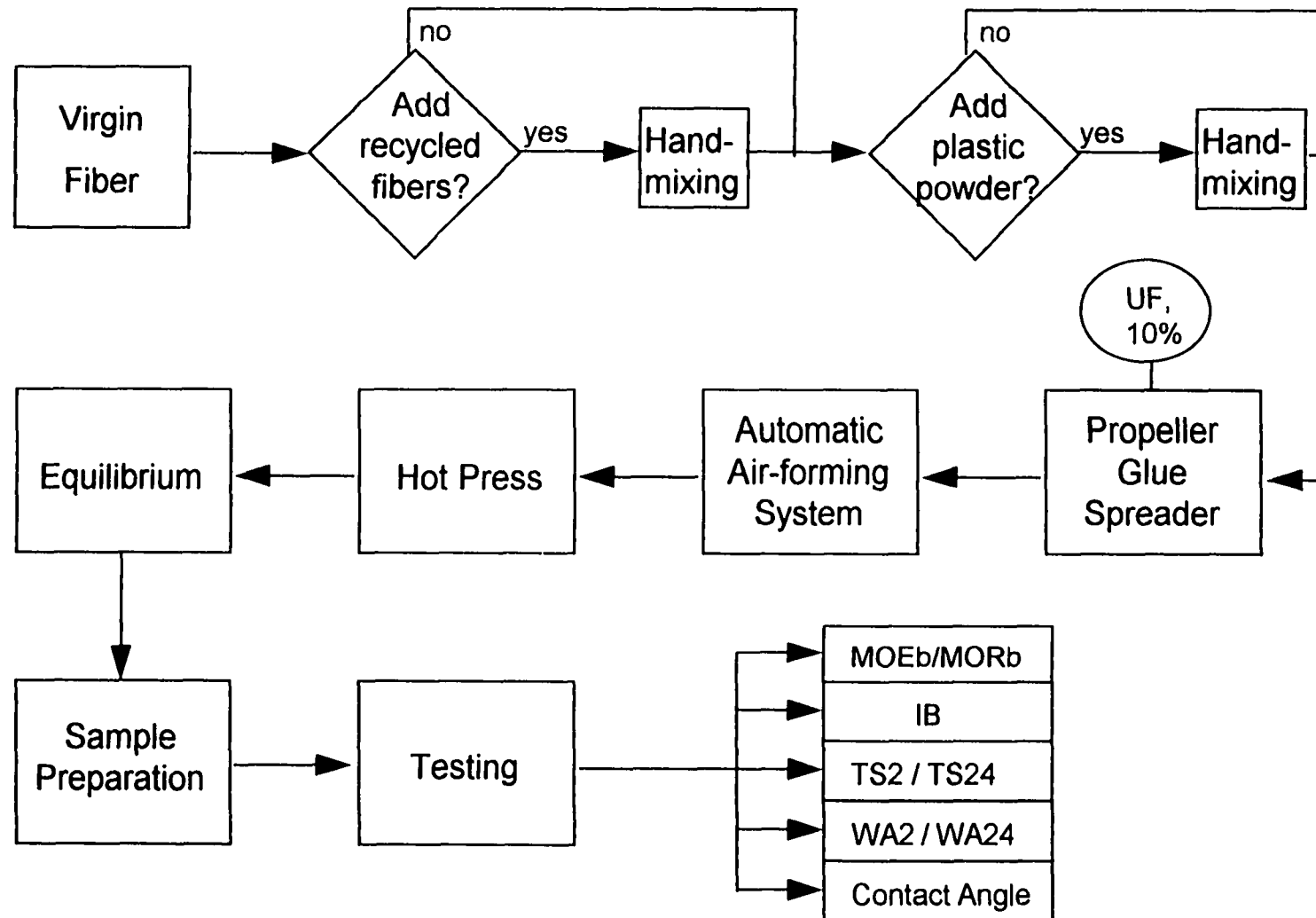


Figure 2.3. Experimental procedures in manufacturing wood-polyolefin composites.

2.2.2.2.2. Effect of Configuration on the Properties of Wood Fiber-Polyethylene Composites

Five configuration schemes of wood fiber-polyethylene composites containing virgin fiber, recycled fiber and recycled polyethylene were fabricated. Among five schemes, three were homogeneous and two were layered structures (Table 2.12). To obtain satisfactory distribution, in treatments which contained PE, fibers were hand-mixed with PE before spreading with UF resin in a propeller glue spreader. In treatments which contained both virgin and recycled fibers, the two fibers were first hand-mixed prior to adding PE.

In the fabrication of homogeneous hardboard, analogous forming plots described in the previous section were used. In the manufacturing of layered panels, the face layer was air-formed on the bottom of the forming box and then topped with the core and another face layer. The layered structures contained 10% virgin fibers on each face layer. Furnishes were compressed and consolidated in a similar manner as shown in Figure 2.3. Each panel configuration contains four 12(W)x12(L)x1/4(T)-inch panels in a completely randomized design. Previously described analysis methods were conducted to evaluate the effects of layer structure and addition of PE on the physical properties of hardboards.

2.2.2.2.3. Effects of Some Processing Factors on the Properties of Wood Fiber-Polyethylene Composites

Panels with compositions consisting of virgin fiber (VIR) : recycled fiber (OCC1): polyethylene (PE)=4:4:2 were fabricated in a manner conformed to a

previously mentioned manufacturing process. The dimensions of the panels were 12(W)x12(L)x1/4(T)-inch, and each treatment combination contained three replicates. The testing for strength properties and contact angles of wood fiber-polyethylene composites was done in the same manner as described previously.

A completely randomized design with factorial treatment arrangement was adopted. The hot pressing schedules and levels of factors investigated are presented in Tables 2.2 and 2.3, respectively. A three-way ANOVA was performed to investigate the effects of hot press temperature (TEMP), nominal panel density (PCF), and hot press time (TIME) on the properties of wood fiber-polyethylene composites. The effect of wetting liquid (LIQUID) was also included in the contact angle measurements, therefore a four-way ANOVA was performed.

Table 2.3. Processing variables in investigating properties of wood fiber-plastic composites.

Variables	Level	Description
Hot press temperature (TEMP)	3	350, 400, 450 °F
Density (PCF)	2	50 and 60 pounds per cubic foot
Hot press time (TIME)	2	5 and 7.5 min.

2.3 RESULTS AND DISCUSSION

2.3.1. Part I. Characterization of Wood Fibers from Four Sources

2.3.1.1. Chemical Analysis

The contents of alcohol-benzene extractive, holo-cellulose, alpha-cellulose, and lignin in wood fibers from the four different sources are

summarized in Table 2.4. Results of Tukey's test among the four types of fibers for each chemical component are also presented in Table 2.4. Alcohol-benzene extractive and lignin contents showed the same ranking with respect to the four fiber types. Virgin fibers (VIR) had the highest contents of alcohol-benzene extractive and lignin, followed by two old corrugated cardboards (OCC1 and OCC2), and then recycled office paper (ROP). The ranking for holo- and alpha-cellulose contents were $ROP \approx \text{or} > OCC1 > OCC2 > VIR$.

The average contents of alcohol-benzene extractive, holo-cellulose, and lignin in VIR are comparable to the results obtained by Shupe (1996). The lowest holo- and alpha-cellulose contents and highest extractive and lignin contents in virgin fiber is due to VIR is from thermomechanical pulp. Whereas the opposite trends found in ROP is because Xerox paper is mostly made of bleached softwood kraft pulp. Corrugated cardboard, on the other hand, is generally made of unbleached kraft pulp as the liner board, and mixed hardwood semichemical pulp as the corrugating medium (Koning 1983).

The discrepancy between OCC1 and OCC2 may be due to the complexity of cardboard composition, variation among different products, addition of recycled fibers, and/or the method of recycling. Old corrugated cardboard can be a good recyclable fibrous material, since it comprised roughly 13% of the MSW, or 23.1 of 179.6 million tons in 1988. However, using old corrugated cardboard as a source of low-cost fillers of thermoplastics may result in lack of uniformity during processing and in the final products.

Table 2.4. Chemical compositions of four wood fiber types.

Fiber type	Alcohol-benzene extractive content		Holo-cellulose content		Alpha-cellulose content		Lignin content	
	----- % -----							
OCC1	3.69	B	81.82	A	75.47	B	16.01	B
	(0.94) ¹		(0.57)		(0.51)		(0.42)	
OCC2	2.04	C	74.52	B	73.75	C	11.17	C
	(0.60)		(0.96)		(1.01)		(0.98)	
ROP	0.41	D	82.78	A	80.72	A	0.73	D
	(0.34)		(0.37)		(1.05)		(0.13)	
VIR	5.85	A	71.30	C	61.46	D	27.56	A
	(1.06)		(1.35)		(1.05)		(0.37)	

¹. Numbers in the parentheses are standard deviations.

2.3.1.2. Fiber Length Measurement

The average weighted fiber lengths of the four fiber types at different disintegrating revolutions are presented in Table 2.5. In VIR, the average fiber length was about 3 mm, whereas, in OCC1, OCC2, and ROP, the fiber lengths were about 2mm. The results of two-way ANOVA (Table 2.6) indicate that fiber type (FT) and disintegrating revolution (DR) main effects were significant at $\alpha=0.05$ level. The interaction (FT*DR), however, was not significant, indicating that the changes in average fiber length with respect to changes in DR among the four fiber types were consistent.

The dependence of fraction on number of disintegrating revolutions for the four fiber types is shown in Figures 2.4 to 2.7. These figures show different patterns. In VIR (Figure 2.7) and OCC2 (Figure 2.5), portion A remained the

dominating fraction for all disintegrating revolutions. However, in VIR each of the four portions was the same with respect to number of revolutions. In contrast, in OCC2, the portion A trended downward and the portions B, C and D upward with an increase in disintegrating revolution. Fractions of VIR were unaffected by disintegration. In both OCC1 (Figure 2.4) and ROP (Figure 2.6), portion A was the major fraction at lower disintegrating revolutions. As the number of revolutions increased, portions B and C became more prominent. These patterns indicate that OCC1 and OCC2 behaved differently to disintegration. OCC2 was more resistant to disintegration than OCC1.

Tukey and contrast tests were performed to clarify the influences of FT and DR, respectively. Results of the Tukey's test show that by combining all revolution levels, the average fiber length of VIR was significantly higher than those of OCC1, OCC2, and ROP (Figure 2.8). This suggests that, in this study, recycled fibers had shorter lengths than virgin fibers, and the fiber lengths of all the three types of recycled fibers were the same. On the other hand, results of contrasts reveal that there was a linear relationship between fiber length and number of disintegrating revolutions.

From these results, we can conclude that recycled fibers from different origins tend to have different chemical compositions even though their fiber lengths may not be significantly different. The mechanical properties of individual fibers, however, are also important factors in the utilization of recycled fibers, but our understanding of these properties is still limited.

Table 2.5. Average weighted fiber lengths of four wood fiber types at five disintegrating revolutions.

Fiber type	Disintegrating revolution				
	100	200	300	400	500
	mm				
OCC1	2.28 (0.12) ¹	2.11 (0.14)	1.88 (0.11)	1.90 (0.14)	1.85 (0.22)
OCC2	2.06 (0.29)	2.05 (0.33)	1.98 (0.16)	2.03 (0.23)	2.03 (0.15)
ROP	1.90 (0.36)	2.04 (0.09)	1.83 (0.16)	1.81 (0.13)	1.86 (0.12)
VIR	3.21 (0.07)	2.91 (0.22)	2.86 (0.09)	2.78 (0.20)	2.78 (0.22)

¹. Numbers in the parentheses are standard deviations.

Table 2.6. Two-way ANOVA with contrasts for effects of fiber type (FT) and disintegrating revolution (DR) on weighted fiber length.

Source	DF	SS	MS	F	P value
FT	3	9.9510	3.3170	88.50	0.0001
DR	4	0.5516	0.1379	3.68	0.0121
--- Linear	1	0.4539	0.4539	12.11	0.0012
--- Quadratic	1	0.0763	0.0763	2.04	0.1614
--- Cubic	1	0.0048	0.0048	0.13	0.7220
--- Quartic	1	0.0167	0.0167	0.44	0.5089
FT*DR	12	0.3476	0.0290	0.77	0.6738
ERROR	59	1.4993	0.0375	---	---

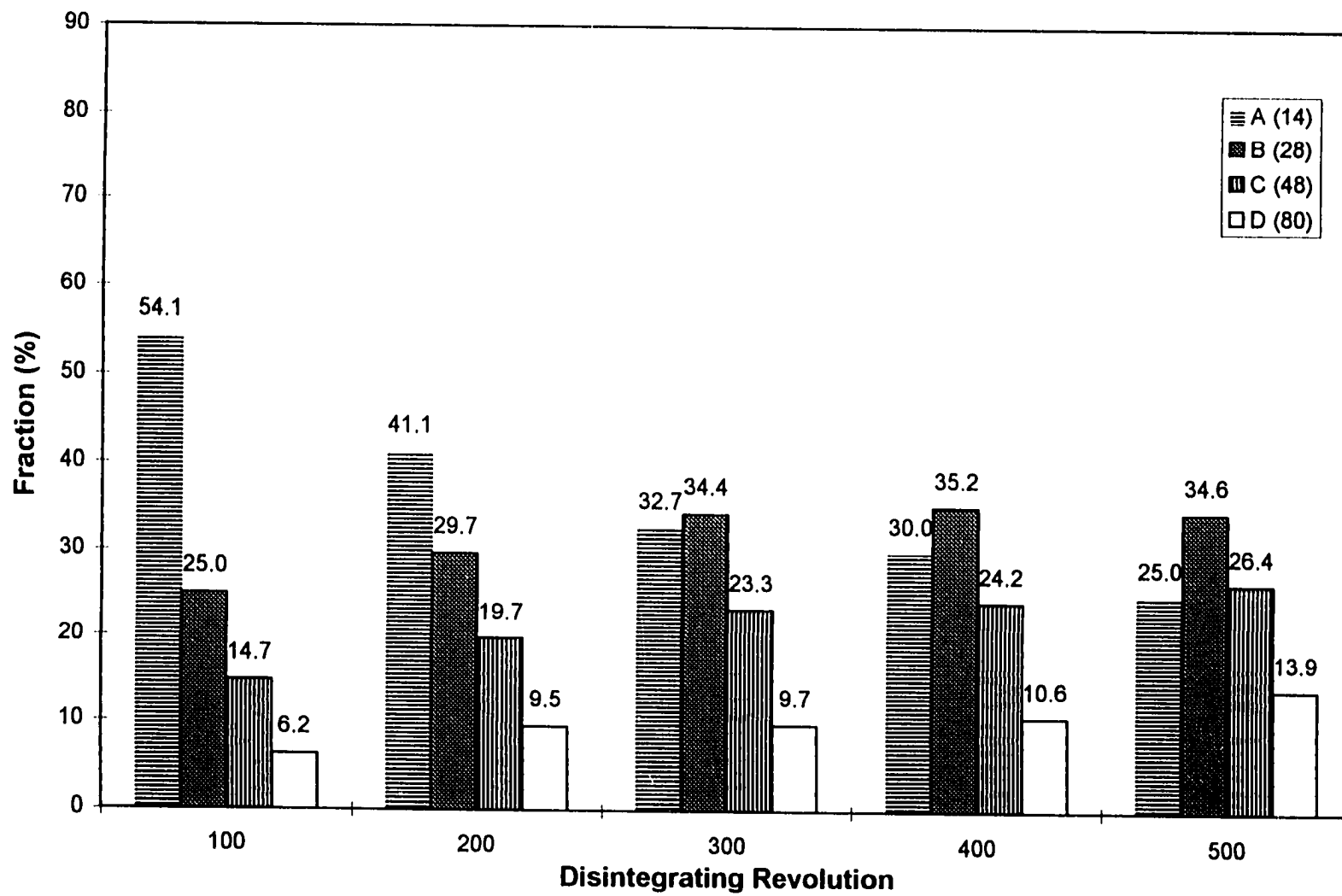


Figure 2.4. Fraction of each mesh class for OCC1 fiber type at various disintegrating revolutions.

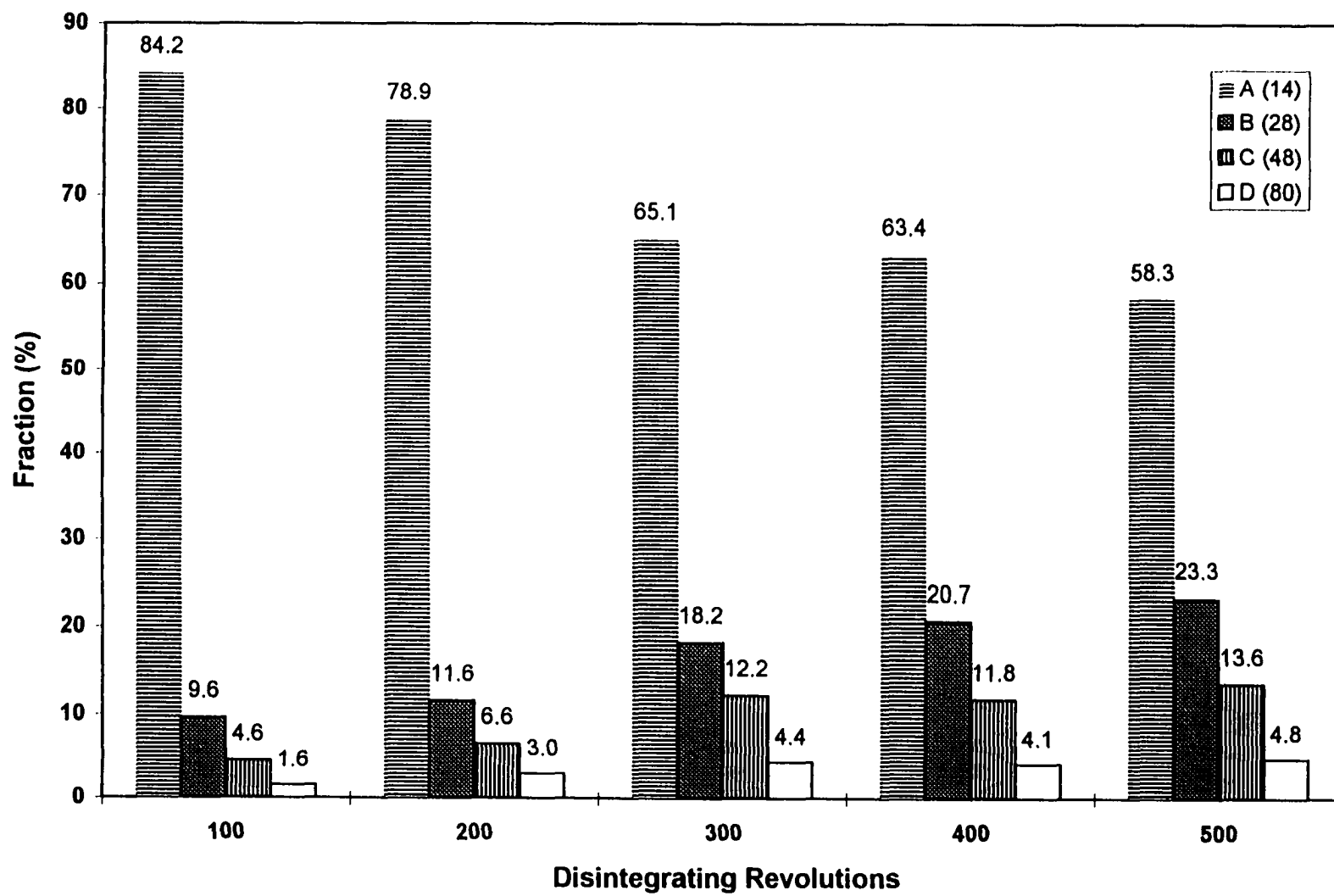


Figure 2.5. Fraction of each mesh class for OCC2 fiber type at various disintegrating revolutions.

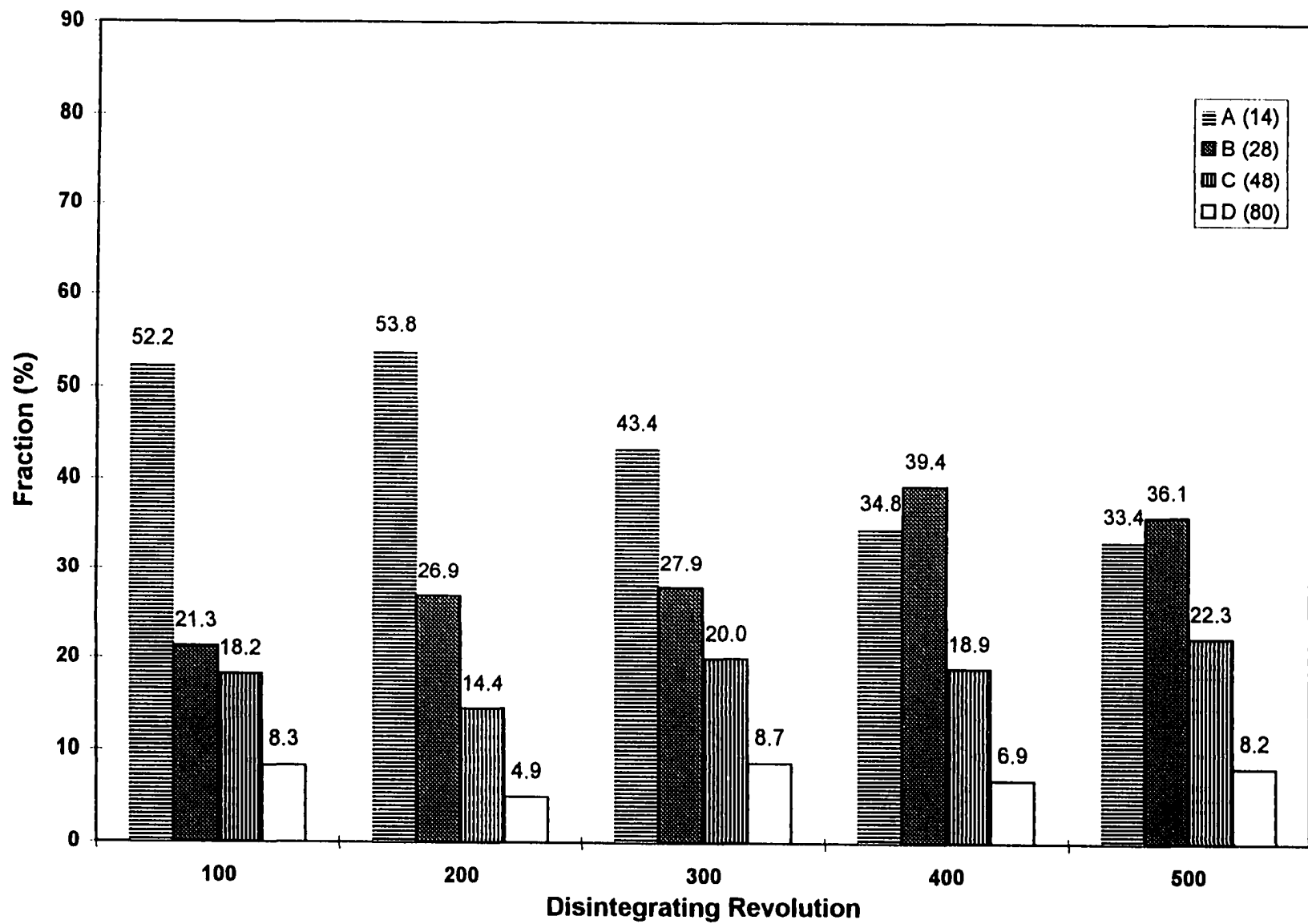


Figure 2.6. Fraction of each mesh class for ROP fiber type at various disintegrating revolutions.

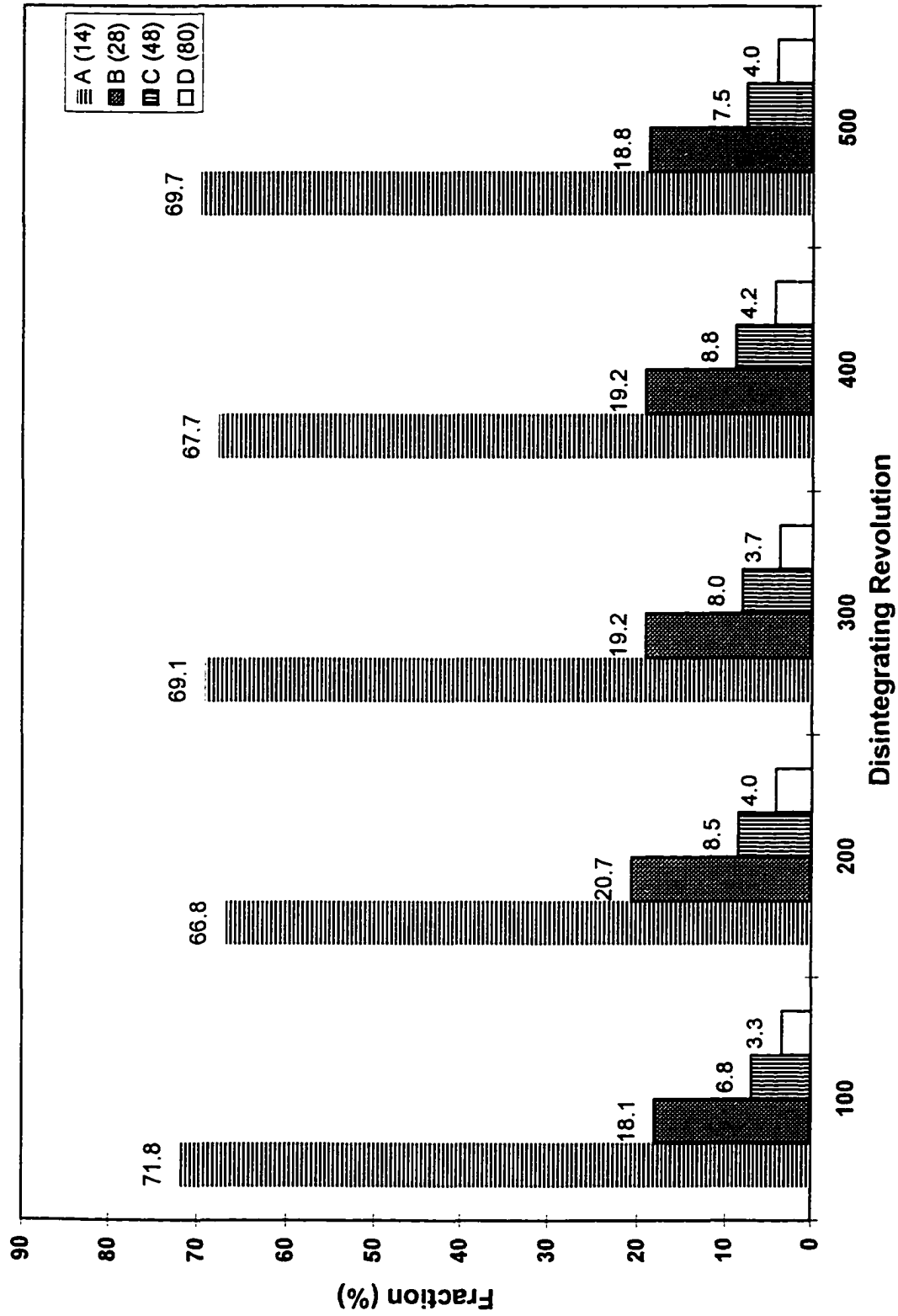


Figure 2.7. Fraction of each mesh class for VIR fiber type at various disintegrating revolutions.

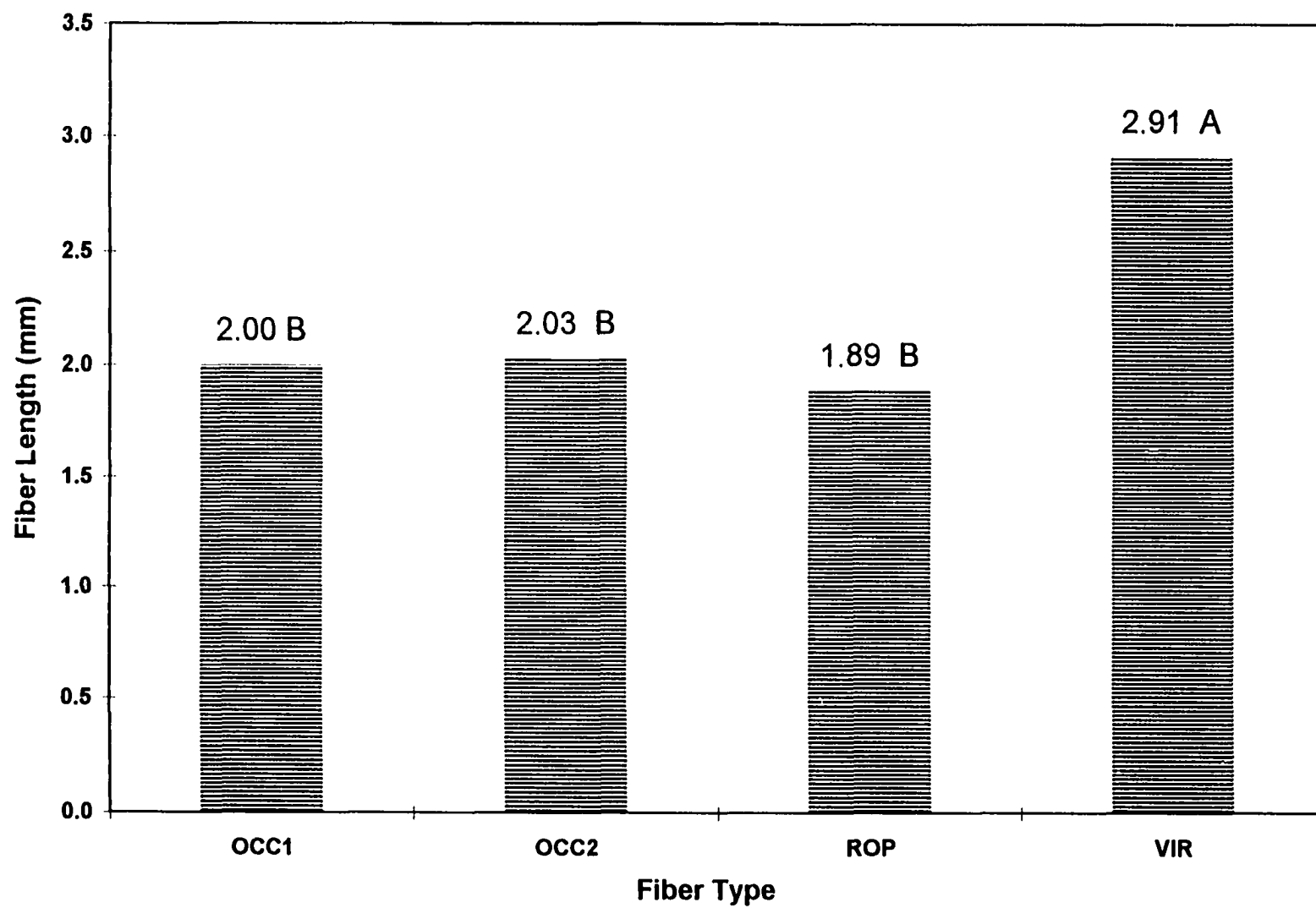


Figure 2.8. Average fiber length of four fiber types with combined disintegrating revolution.

Table 2.7. Physical and mechanical properties of fiberboards made from various fiber compositions.

Property	A ¹	B	C	D	E	F
MC (%)	4.57 (0.36) ²	4.68 (0.11)	5.27 (0.17)	5.44 (0.24)	5.41 (0.05)	5.40 (0.07)
Density (pcf)	48.27 A ³ (1.42)	46.31 AB (1.58)	46.91 AB (1.42)	45.77 AB (1.21)	43.79 B (2.36)	43.09 B (1.42)
IB (psi)	130.69 A (7.38)	83.49 B (15.04)	71.31 B (8.60)	44.04 C (4.70)	35.43 CD (1.88)	21.78 D (2.12)
MOR _b (psi)	4538 A (1612)	3029 AB (547)	2734 AB (301)	1969 B (349)	1392 B (386)	1051 B (127)
MOE _b (10 ³ psi)	410 A (131)	311 AB (53)	301 AB (40)	207 B (57)	177 B (38)	135 B (19)
WA2 (%)	75.03 D (9.20)	89.44 CD (9.30)	95.51 CD (6.72)	109.17 BC (5.59)	127.87 AB (13.04)	139.56 A (6.51)
WA24 (%)	87.59 D (5.77)	102.29 CD (6.50)	105.66 CD (5.99)	117.00 BC (4.86)	135.72 AB (13.02)	146.16 A (6.36)
TS2 (%)	21.71 E (2.82)	24.86 DE (1.13)	26.87 CD (0.56)	29.76 BC (0.94)	32.73 AB (1.03)	35.64 A (0.59)
TS24 (%)	25.00 D (2.03)	27.59 CD (0.68)	29.33 C (0.48)	32.64 B (0.86)	35.94 A (1.62)	38.75 A (0.24)

¹. The compositions of virgin fiber (by weight) in each treatment were: A:100, B:80, C:60, D:40, E:20, and F:0%, respectively.

². Numbers in parentheses are standard deviations.

³. Numbers followed by same character denote not significant at alpha=0.05 level.

2.3.2. Part II. Effects of Recycled Materials on the Properties of Wood Fiber-Polyethylene Composites

2.3.2.1. Effect of Recycled Fiber on the Properties of Hardboards

Table 2.7 shows the physical and mechanical properties of hardboard made from different compositions of virgin fibers and recycled fibers. The average moisture content for all panels ranged from 4.5 to 5.5%. The values in

the table indicate that adding recycled fiber (OCC1) adversely affected physical and mechanical properties of hardboard.

Due to large variations in properties, Hartly F-max tests were performed to detect the violation of the assumption of homogeneity, and the results supported the legitimacy of further analyses. The impact of recycled OCC fiber on the physical and mechanical properties of hardboard was studied using contrast analysis (Table 2.8). All physical and mechanical properties of hardboard, except IB, were found to be linearly dependent on the virgin fiber ratio. In IB, the relationship with fiber ratio were quadratic. Regression analyses were subsequently performed to estimate the coefficients, as shown in Table 2.9 and Figures 2.9 to 2.14.

2.3.2.1.1. Panel Density (PCF)

The graph in Figure 2.9 shows a linear relationship between panel density and virgin fiber ratio, and that 58% of the variations in panel density was attributed to virgin fiber ratio. The reduction in panel density with increasing recycled fibers may be caused by the automatic air-forming system. Since recycled fibers are lighter in weight and smaller in size, the unadjustable motor speed tends to blow off more recycled fibers than virgin fibers, leading to a slight reduction in panel density. Moreover, panels made from recycled fibers are more difficult to compress but easier to spring back, thus contributing further to the variations in panel density.

Table 2.8. Contrast analysis of physical and mechanical properties of fiberboards with different fiber compositions.

Property	Contrast	DF	MS	F	P
Density	Linear	1	51.3365	19.77	0.0008
	Quadratic	1	0.5752	0.22	0.6463
	Cubic	1	0.2315	0.09	0.7704
	Error	12	2.5961		
IB	Linear	1	21972.4035	342.64	0.0001
	Quadratic	1	1183.6067	18.46	0.0010
	Cubic	1	163.9225	2.56	0.1358
	Error	12	64.1277		
MOR _b	Linear	1	22887483.73	41.89	0.0001
	Quadratic	1	793074.48	1.45	0.2515
	Cubic	1	140908.76	0.26	0.6208
	Error	12	546373.44		
MOE _b	Linear	1	148944.5733	33.58	0.0001
	Quadratic	1	1438.4121	0.32	0.5795
	Cubic	1	51.2641	0.01	0.9162
	Error	12	4435.0344		
WA2	Linear	1	8740.3954	114.02	0.0001
	Quadratic	1	48.6289	0.63	0.4412
	Cubic	1	0.0145	0.00	0.9892
	Error	12	76.6559		
WA24	Linear	1	7011.2563	121.91	0.0001
	Quadratic	1	57.3526	1.00	0.3377
	Cubic	1	3.0165	0.05	0.8227
	Error	12	57.5117		
TS2	Linear	1	396.0419	200.63	0.0001
	Quadratic	1	0.2433	0.12	0.7316
	Cubic	1	0.1497	0.08	0.7877
	Error	12	1.9740		
TS24	Linear	1	404.2689	295.21	0.0001
	Quadratic	1	1.9119	1.40	0.2603
	Cubic	1	0.1411	0.10	0.7537
	Error	12	1.3694		

Table 2.9. Statistics and estimated parameters for linear regression analysis of fiberboard properties on virgin fiber ratio as denoted by model $Y = \sum b_i X_i$.

Panel property	Statistics			Estimated parameter		
	F	P	R ²	b ₀	b ₁	b ₂
Density	22.44	0.0002	0.5838	43.2184	0.0494	---
IB	139.40	0.0001	0.9489	24.1488	0.2101	0.008127
MOR _b	46.26	0.0001	0.7430	801.4444	33.0133	---
MOE _b	40.16	0.0001	0.7151	123.6282	2.6632	---
WA2	132.75	0.0001	0.8924	138.3538	-0.6451	---
WA24	127.24	0.0001	0.8883	144.6268	-0.5778	---
TS2	256.34	0.0001	0.9412	35.4614	-0.1373	---
TS24	328.70	0.0001	0.9536	38.4779	-0.1387	---

2.3.2.1.2. Mechanical Property (IB, MOR_b and MOE_b)

Virgin fiber ratio contributed approximately 95% of the variations in the IB of fiberboard. The relationship, which was parabolic (Figure 2.10), was due to variations in panel density and inferiority in individual strength properties of recycled fiber. In addition, the finer recycled fibers may increase the surface area which results in lower resin content per unit surface area. The more hydrophilic recycled fibers may also induce overpenetration of adhesive. The IB, however, can be improved by adding more adhesives (Rowell and Harrison 1992, Krzysik *et al.* 1993).

The dependency of bending properties on virgin fiber ratio is shown in Figure 2.11 for MOR_b and Figure 2.12 for MOE_b. At least 70% of the variations

in both MOR_b and MOE_b can be explained by virgin fiber ratio. The reasons for these adverse effects in recycled fibers are the same as those in IB. However, the impact of strength was more drastic in IB than in MOR_b and MOE_b . In comparison with panel A (100% VIR), panel F (100% OCC1) showed 83% lower IB, and 77% and 67% lower MOR_b and MOE_b , respectively.

2.3.2.1.3. Dimensional Stability (WA and TS)

The effect of virgin fiber ratio on water absorption is shown in Figure 2.13 for 2-hour (WA2) and 24-hour (WA24) water absorption. In both absorptions, 89% of the variations were attributed to virgin fiber ratio. The slopes of these two lines were slightly different, showing that the effect of fiber composition was slightly stronger on WA 2 than on WA24. In this study, fiberboards made from recycled fiber show enormous water uptake, which was far above the 40% commercial standard for 1/4-inch service class hardboard (Suchsland and Woodson 1986). It is because all of these panels were fabricated in the laboratory and they neither received heat treatment nor were subjected to oil tempering.

The dependencies of 2-hour (TS2) and 24-hour (TS24) thickness swelling on virgin fiber ratio are shown in Figure 2.14. The slopes of the two straight lines were negative and almost identical (-0.1387 vs -0.1373), indicating that recycled fiber affected the thickness swelling adversely. Both equations have relatively high coefficients of determination (0.954 vs 0.941). This phenomenon can be

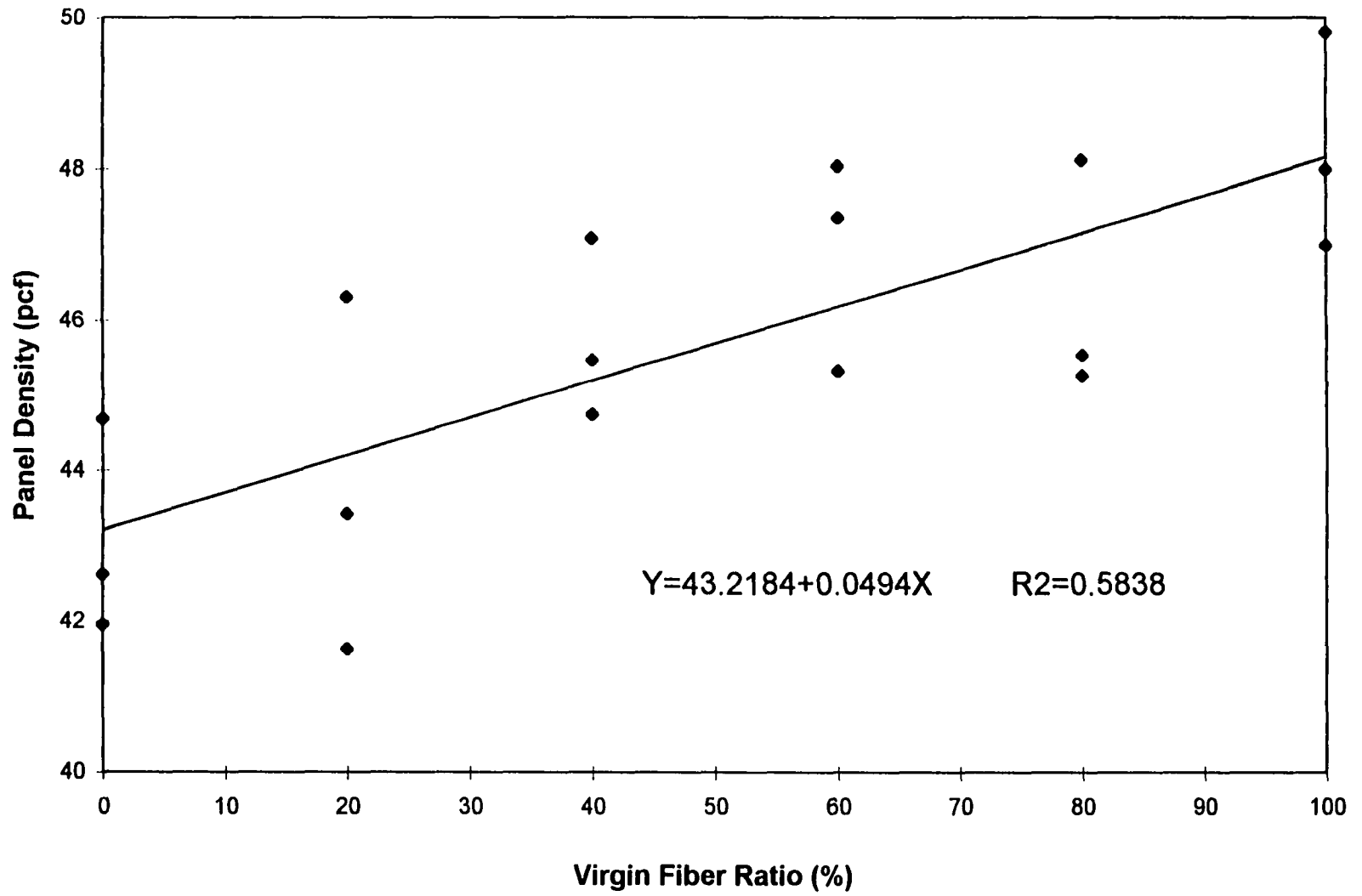


Figure 2.9. Correlation between virgin fiber ratio and panel density of fiberboard.

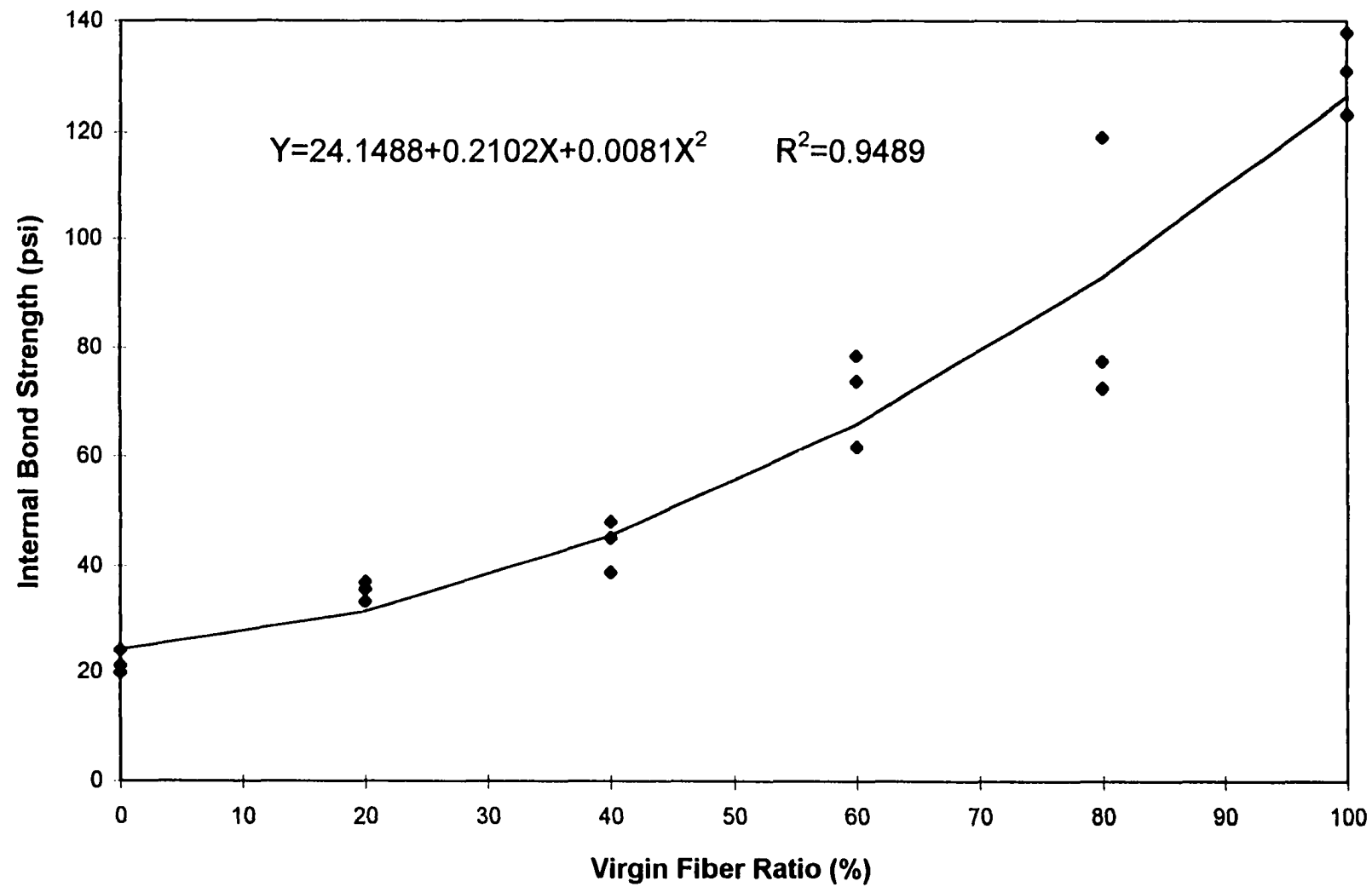


Figure 2.10. Relationship between virgin fiber ratio and internal bond strength of fiberboard.

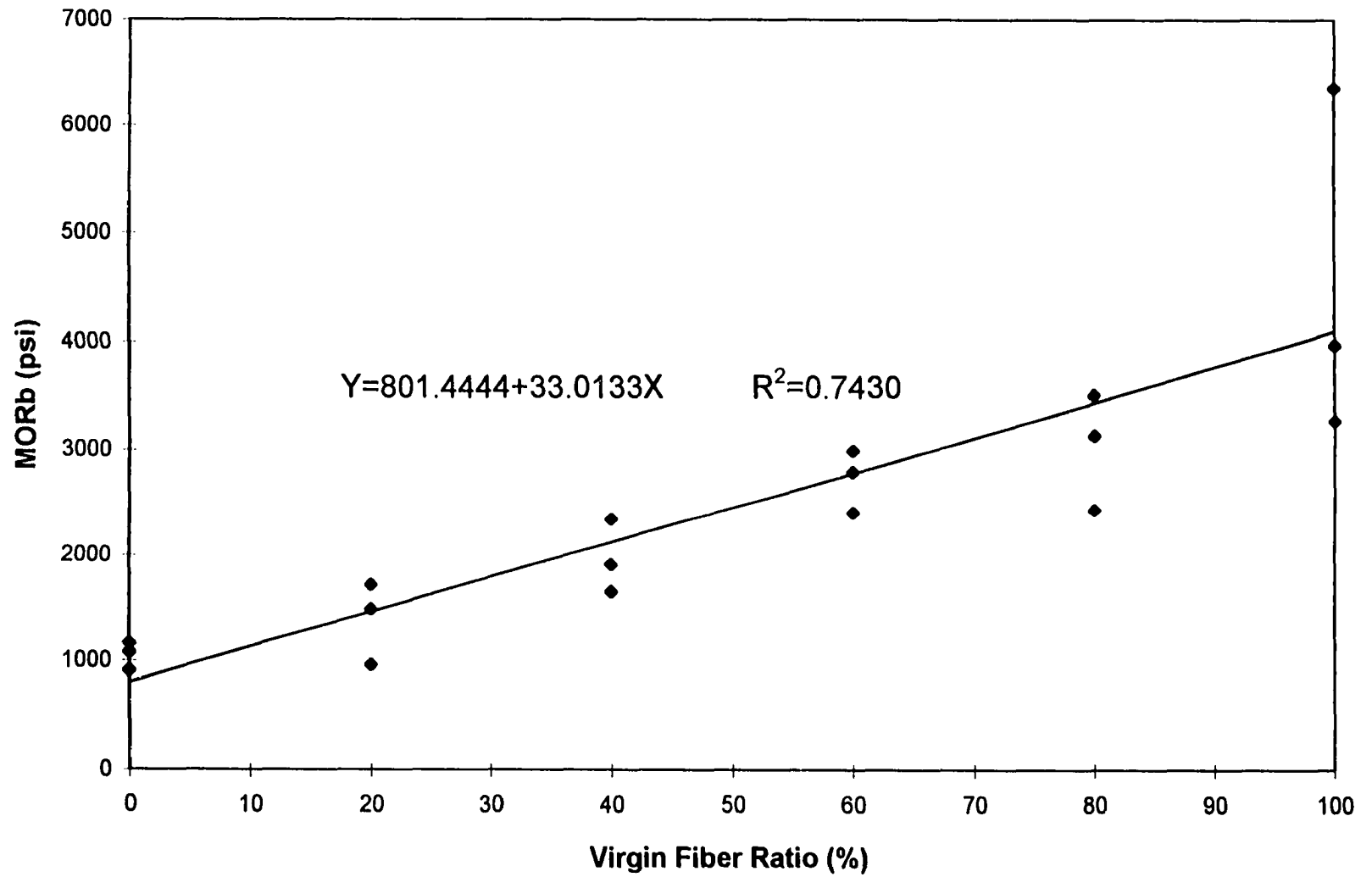


Figure 2.11. Correlation between virgin fiber ratio and bending modulus of rupture of fiberboard.

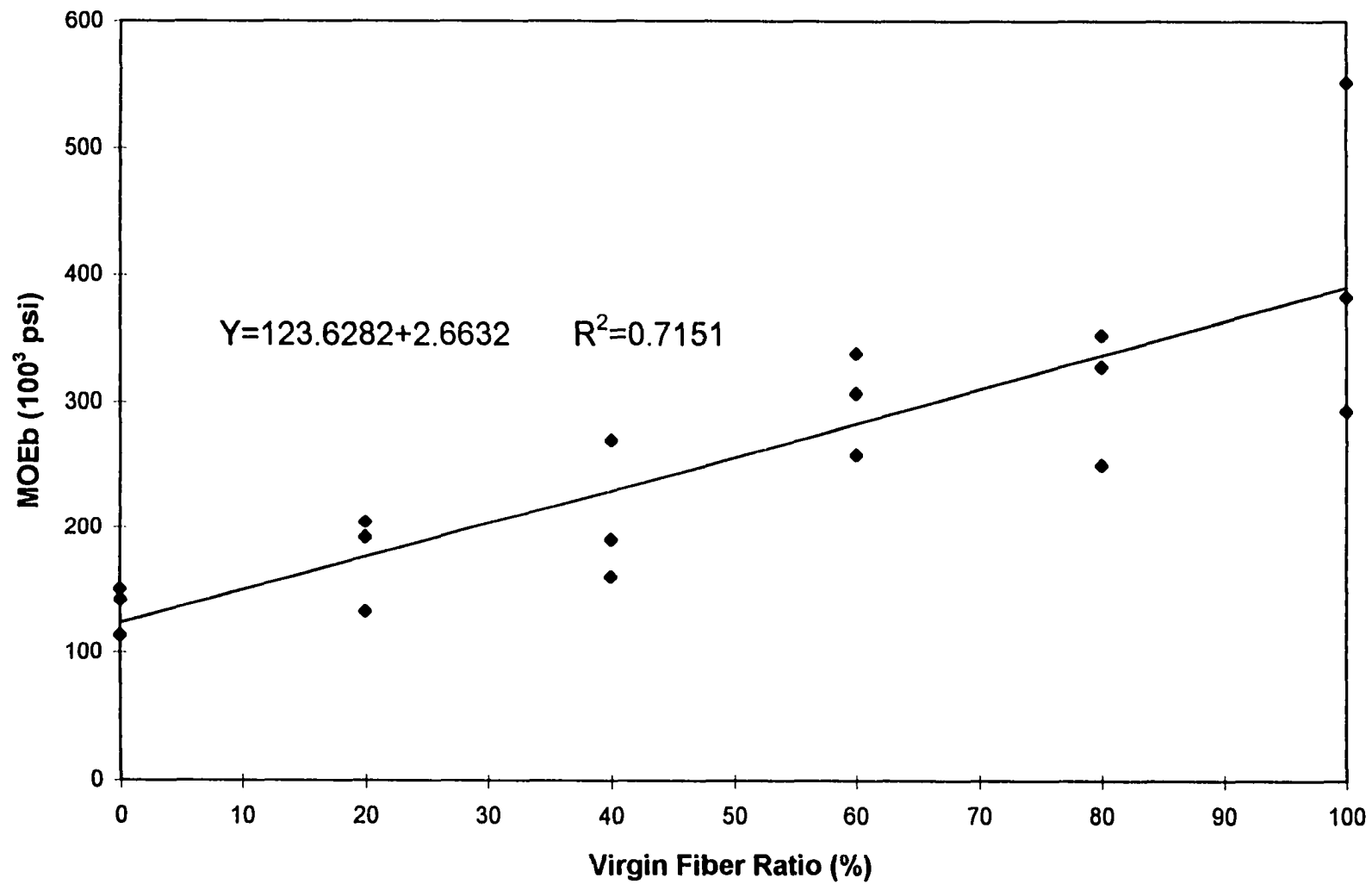


Figure 2.12. Correlation between virgin fiber ratio and bending modulus of elasticity of fiberboard.

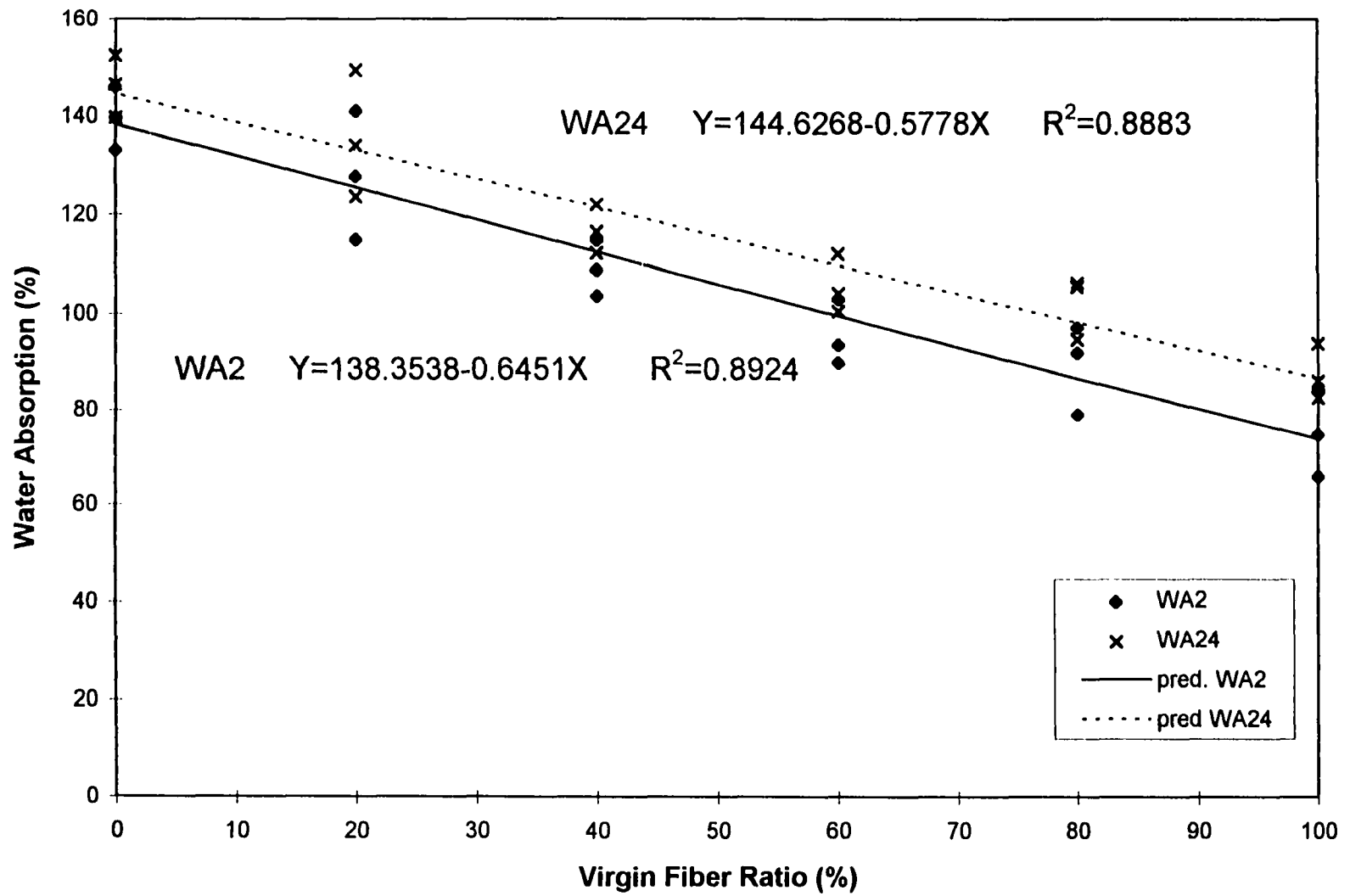


Figure 2.13. Correlation between virgin fiber ratio and water absorption of fiberboard.

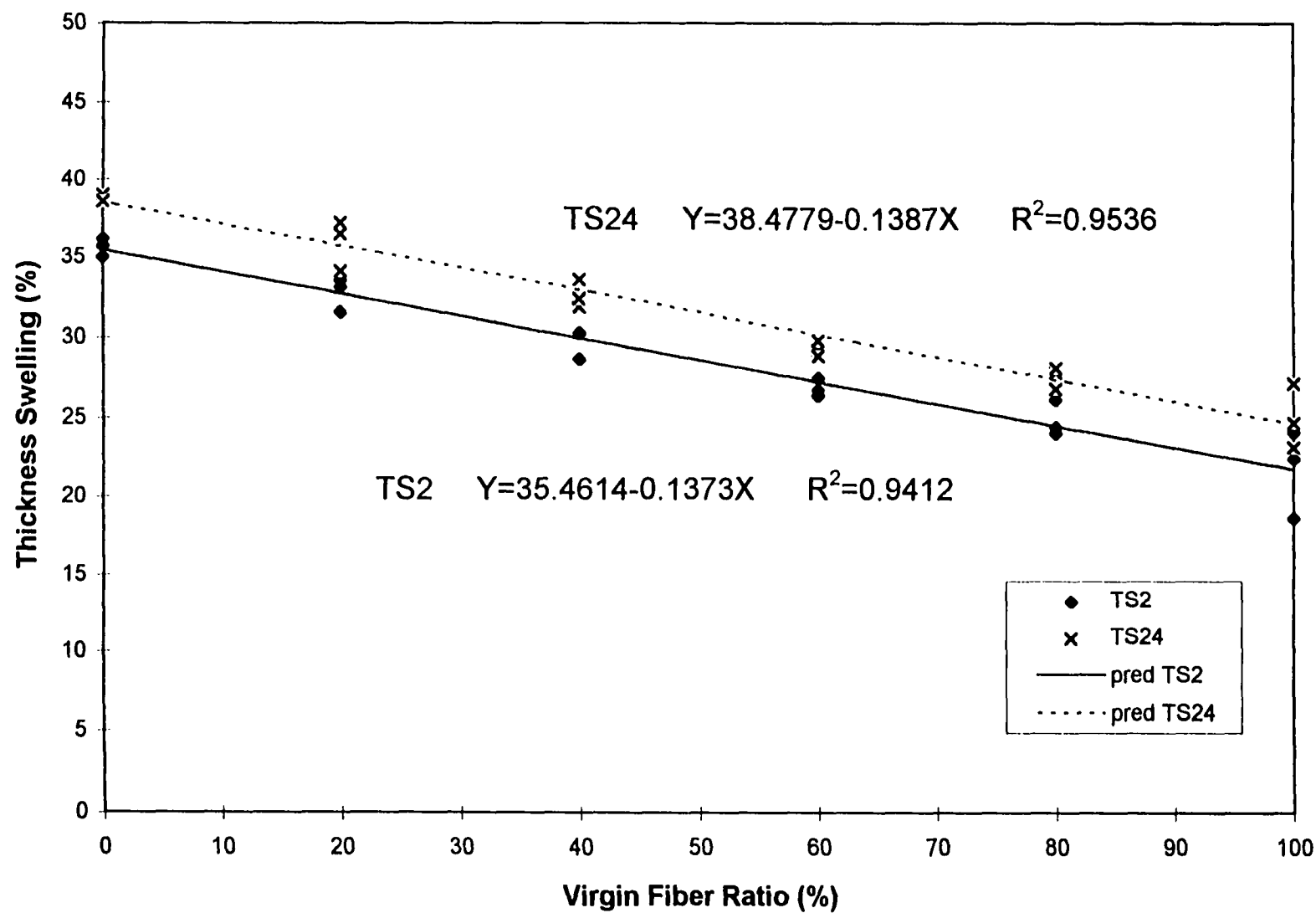


Figure 2.14. Correlation between virgin fiber ratio and thickness swelling of fiberboard.

explained from physical, chemical, and anatomical aspects. Physically, the average fiber length of OCC1 is considerably shorter than that of VIR; chemically, OCC1 has higher holocellulose content and lower lignin content than VIR; and anatomically, (Figure 2.15) OCC1 is “fuzzier” when compared with VIR.

The uptake of water in hardboard from recycled fibers was far from commercial standards, but the deviations of thickness swelling from standard value (30%) were less dramatic. If WA and TS are not considered, panel A qualifies for commercial class 2-standard hardboard, panel B for class 4-service hardboard, and panel C for class 5-industrialite hardboard. The addition of 60% or more recycled fibers failed to meet the requirements of any commercial hardboard.

2.3.2.1.4. Contact Angle

Contact angle measurements with different wetting liquids on hardboard of various fiber compositions are listed in Table 2.10. This is a split plot design with factorial treatment arrangement. No trend could be detected from the data; however, results of ANOVA (Table 2.11) indicate highly significant ($p < 0.01$) differences in virgin fiber ratio (VFR) main effect which was placed on the main plots, and in wetting liquid (LIQUID) main effect on the subplots. The two-way interaction between virgin fiber ratio and wetting liquid was also highly significant. Since the F value of wetting liquid main effect was very large (347.5), Tukey's tests were performed despite significant interaction. The effect of wetting liquid on panel contact angle (Figure 2.16) indicates that when different virgin fiber

ratios were combined, wetting liquid UF had the highest contact angle, followed by water, and then PF. The effect of virgin fiber ratio on the contact angle (Figure 2.17) was not the same with different wetting liquids. For UF and water, the contact angle did not change with virgin fiber ratio. On the contrary, for PF, the contact angle increased sharply as the recycled fibers were first introduced into the system, then leveled off with increased recycled fiber ratio, indicating that the PF resin was more sensitive to the addition of recycled fibers.

Table 2.10. Contact angle of three wetting liquids for fiberboards made from different virgin fiber and recycled fiber ratios.

Liquid	A ¹	B	C	D	E	F
	----- degree -----					
PF	76.28 (3.97) ²	95.64 (3.27)	92.31 (2.43)	94.73 (2.93)	98.22 (3.19)	98.09 (2.37)
UF	102.66 (3.31)	104.00 (2.50)	104.23 (3.70)	104.33 (2.59)	104.09 (3.39)	102.33 (4.12)
WATER	99.28 (2.90)	100.48 (3.10)	98.13 (3.98)	97.13 (2.32)	98.34 (2.13)	96.45 (3.23)

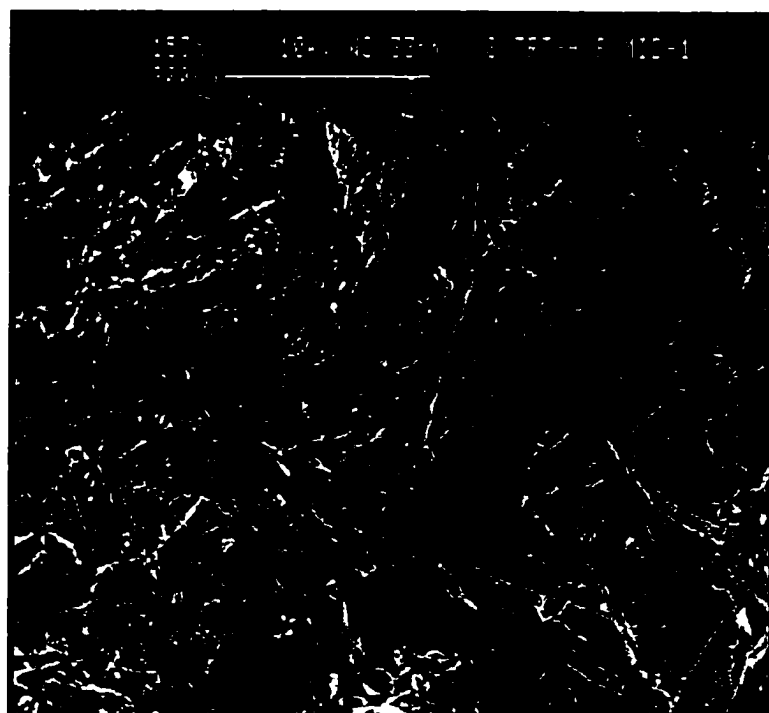
¹. The compositions of virgin fiber (by weight) in each treatment were: A:100, B:80, C:60, D:40, E:20, and F:0%, respectively.

². Numbers in parentheses are standard deviations.

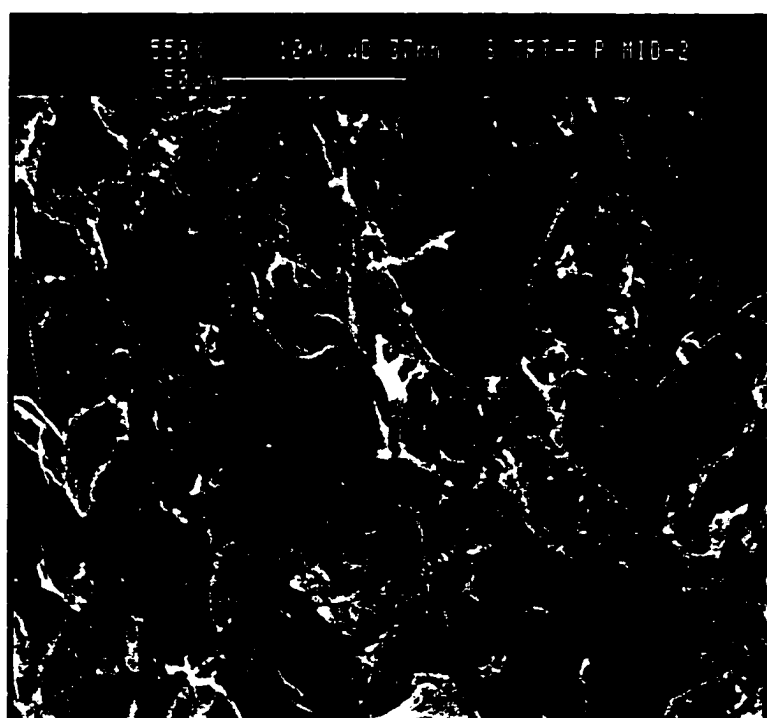
Table 2.11. ANOVA of contact angle for fiberboards with various fiber compositions.

Source ¹	DF	MS	F	P
VFR	5	250.6450	29.59	0.0001
REP(VFR)	12	30.9279	3.65	0.0001
LIQUID	2	2943.4167	347.50	0.0001
VFR*LIQUID	10	328.0405	38.73	0.0001
ERROR	186	8.4704	-----	-----

¹. VFR: Virgin fiber ration, LIQUID: Wetting liquid.



(A)



(B)

Figure 2.15. SEM micrograph of fiberboard made from 100% virgin fiber (A) and 100% old corrugated cardboard (B).

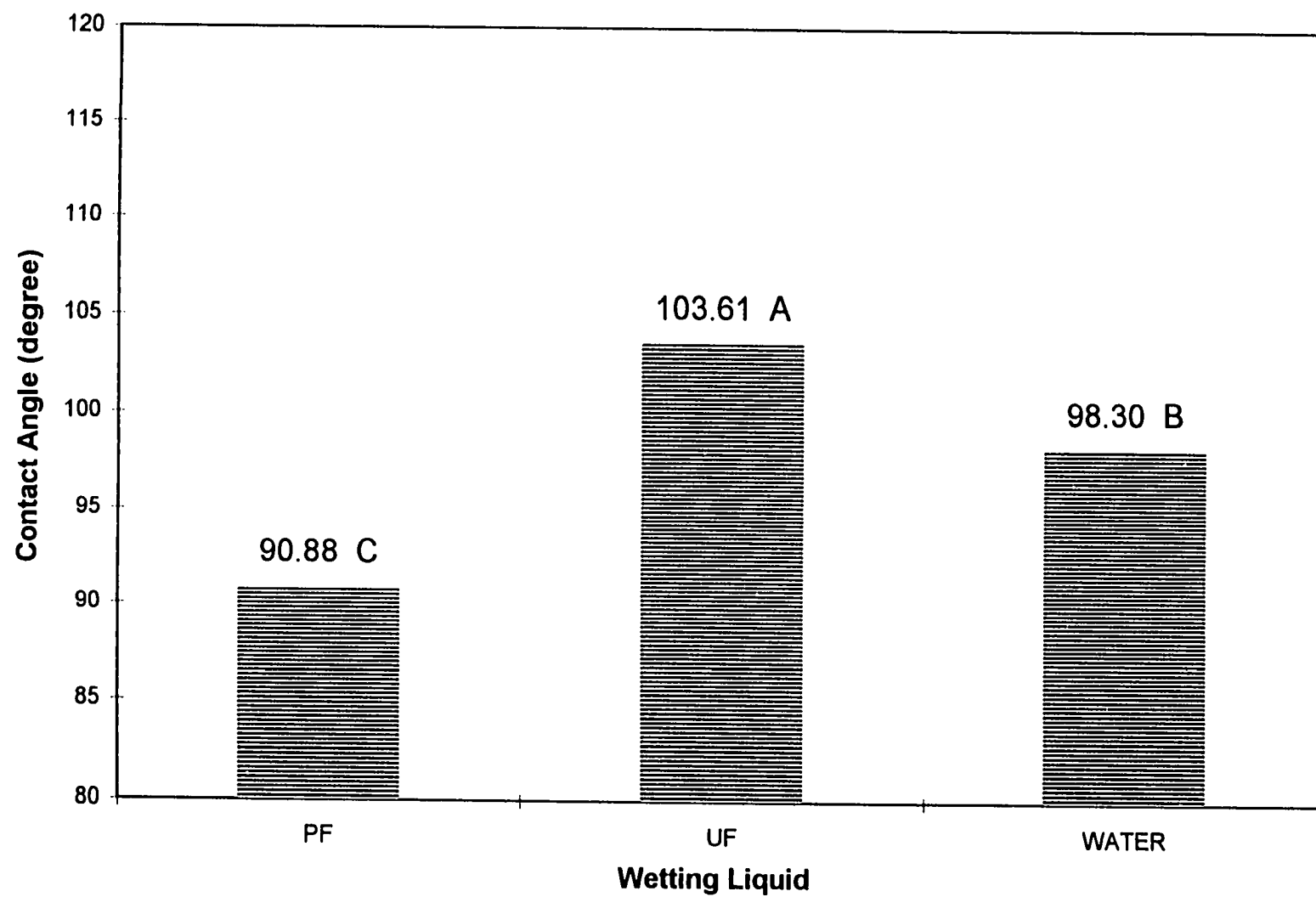


Figure 2.16. Contact angle of fiberboards measured by three wetting liquids.

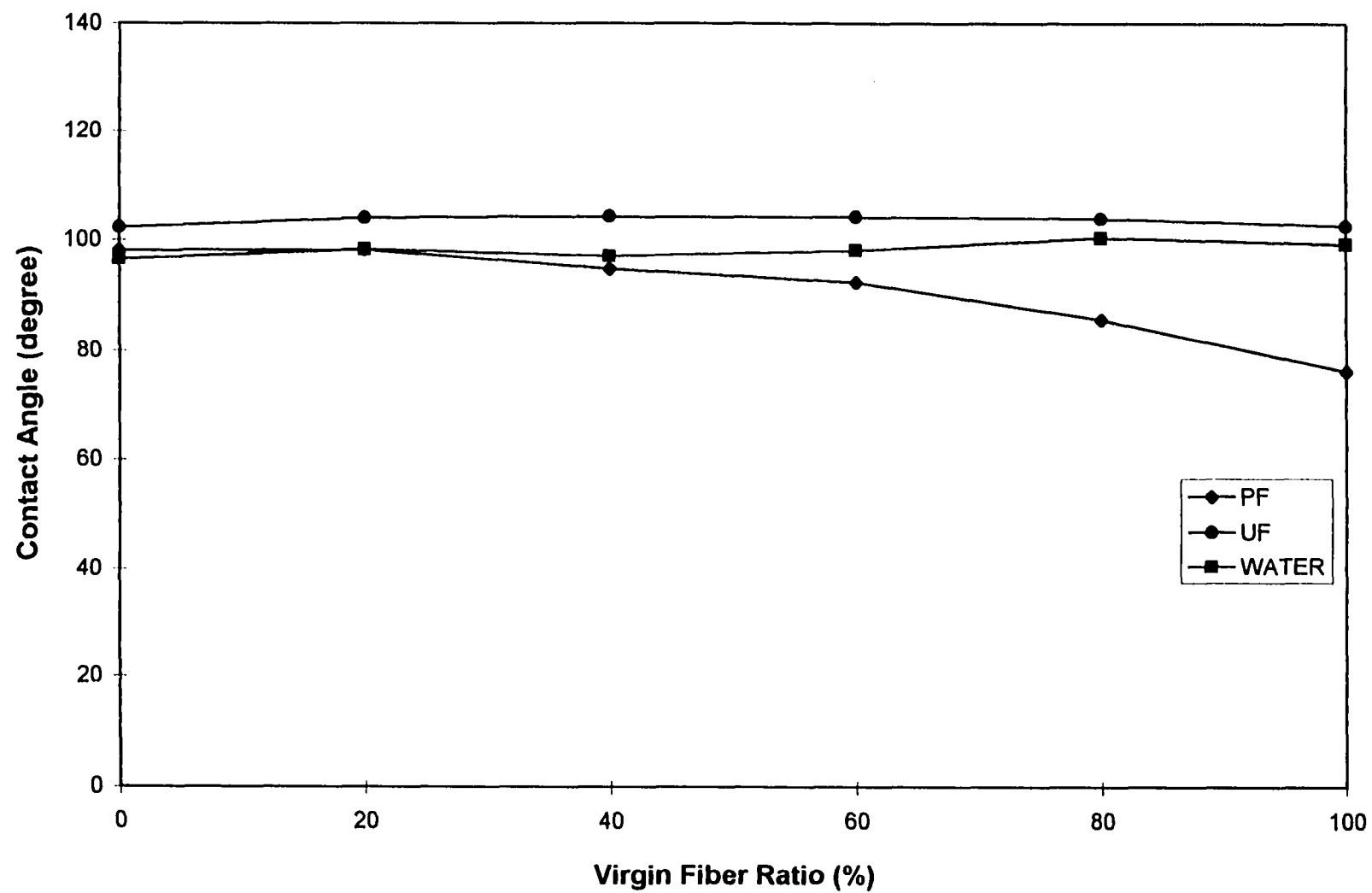


Figure 2.17. Effect of wetting liquid and virgin fiber ratio on contact angle of fiberboard.

2.3.2.2. Effect of Configuration on the Properties of Wood Fiber-Polyethylene Composites

In this study, recycled fibers and recycled polyethylene powder were introduced simultaneously in the making of fiberboard. Five schemes were used to enhance the properties of hardboard. Three homogeneous configurations were designed to investigate the impact of recycled polyethylene on virgin fiber (VIR), old corrugated cardboard (OCC1), and the mixture of both (VIR+OCC1). The amount of PE added was 20% of the total weight. At the same time, two layered structures were developed to enhance the bending properties of fiberboard. Both schemes contained 10% virgin fibers on two faces; one had PE, and the other did not. Average moisture contents for the five configurations ranged from 3.9 to 4.5%, and panel densities ranged from 41 to 45 pcf.

2.3.2.2.1. Mechanical Property (IB, MOR_b and MOE_b)

Physical and mechanical properties of fiberboards with various structures are summarized in Table 2.12. This is a completely randomized design. Hartley F-max tests of homogeneity were performed before one-way ANOVA was employed. The difference in strength properties (i.e., IB, MOR_b and MOE_b) among the five configurations were highly significant ($P < 0.01$). Tukey's test, as shown in Figures 2.18 - 2.20, revealed a general trend for all strength properties in the 5 schemes: homogeneous configurations followed $G > I > H$ pattern. The layered structures were undistinguishable between I and H while significantly lower than G. The implications are as follows:

(1) at given PE content and homogeneous configuration (G vs H vs I), IB and bending properties were related to virgin fiber ratio,

(2) for the same material composition (I vs K), homogeneous or layered structure did not affect IB and bending properties,

(3) within the same layered structure (J vs K), replacing 20% virgin fiber with PE did not significantly decrease IB, MOR_b and MOE_b of fiberboard.

Table 2.12. Physical and mechanical properties of wood fiber-polyethylene composites with different configurations.

Denotation	G ¹	H	I	J	K
Configuration	Homo	Homo	Homo	Layered	Layered
VIR:OCC1:PE	80:0:20	0:80:20	40:40:20	60:40:0	40:40:20
MC (%)	4.04 (0.38) ²	4.55 (0.19)	3.98 (0.32)	4.21 (0.41)	3.89 (0.36)
Density (pcf)	44.89 (1.75)	41.01 (0.97)	43.10 (1.95)	44.50 (2.81)	44.39 (2.48)
IB (psi)	108.52 (19.27)	30.38 (9.60)	66.23 (10.09)	41.25 (9.86)	68.50 (12.82)
MOR _b (psi)	3317 (178)	1339 (454)	2339 (369)	1933 (461)	2032 (189)
MOE _b (10 ³ psi)	284 (13)	138 (43)	231 (39)	204 (41)	201 (16)
WA2 (%)	70.28 (5.21)	109.05 (14.58)	91.67 (8.49)	117.00 (15.64)	91.69 (8.73)
WA24 (%)	80.99 (5.86)	113.98 (13.94)	96.93 (8.10)	123.87 (15.77)	95.41 (8.59)
TS2 (%)	14.85 (1.11)	22.48 (2.32)	19.24 (2.15)	35.71 (3.37)	21.76 (0.94)
TS24 (%)	16.66 (0.54)	24.97 (2.18)	19.85 (0.91)	38.11 (3.52)	23.29 (0.92)

¹ VIR:OCC1:PE=virgin fiber : recycled fiber : polyethylene.

² Numbers in parentheses are standard deviations.

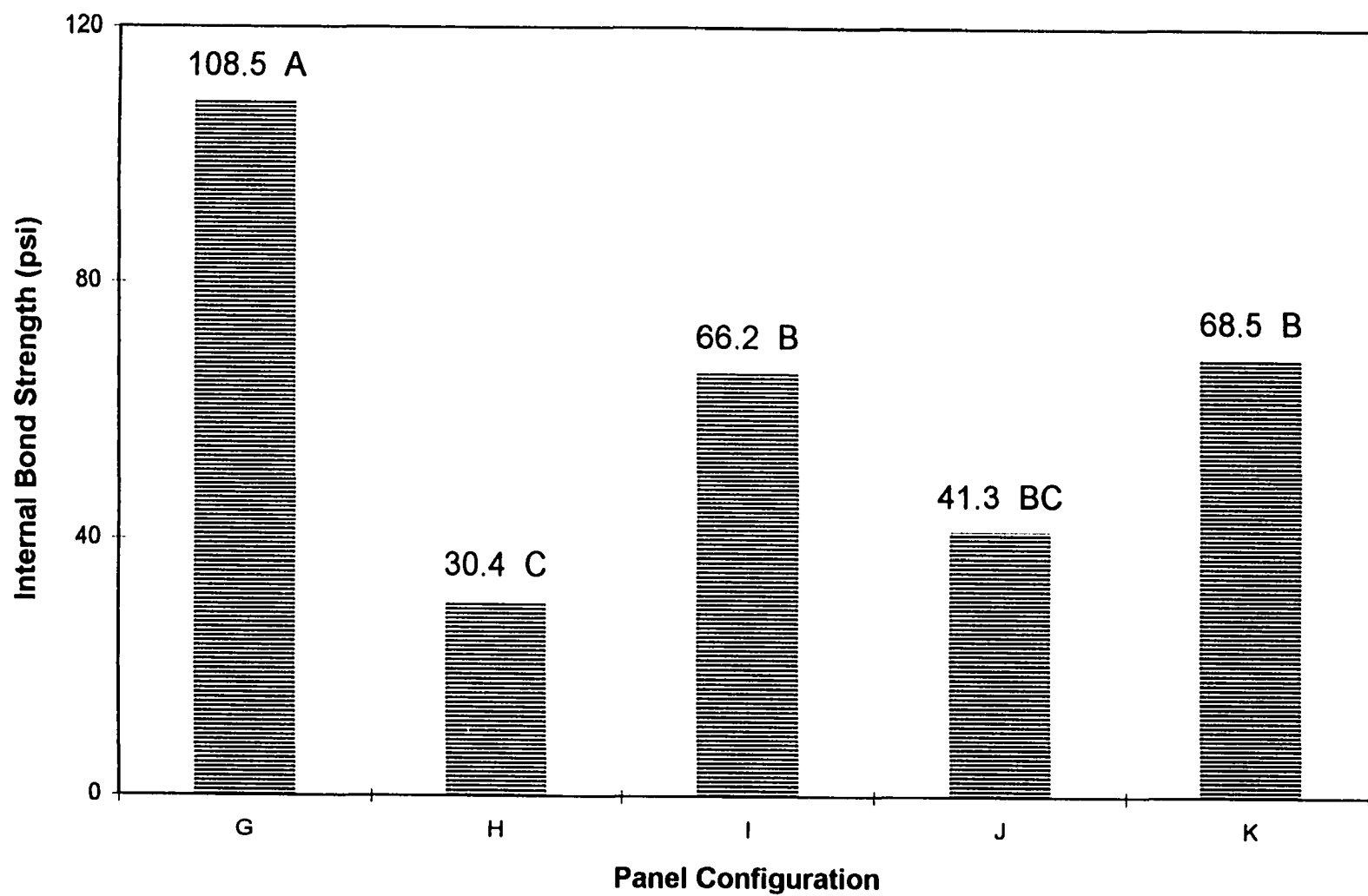


Figure 2.18. Internal bond strength of wood fiber-polyethylene composite with various configurations.

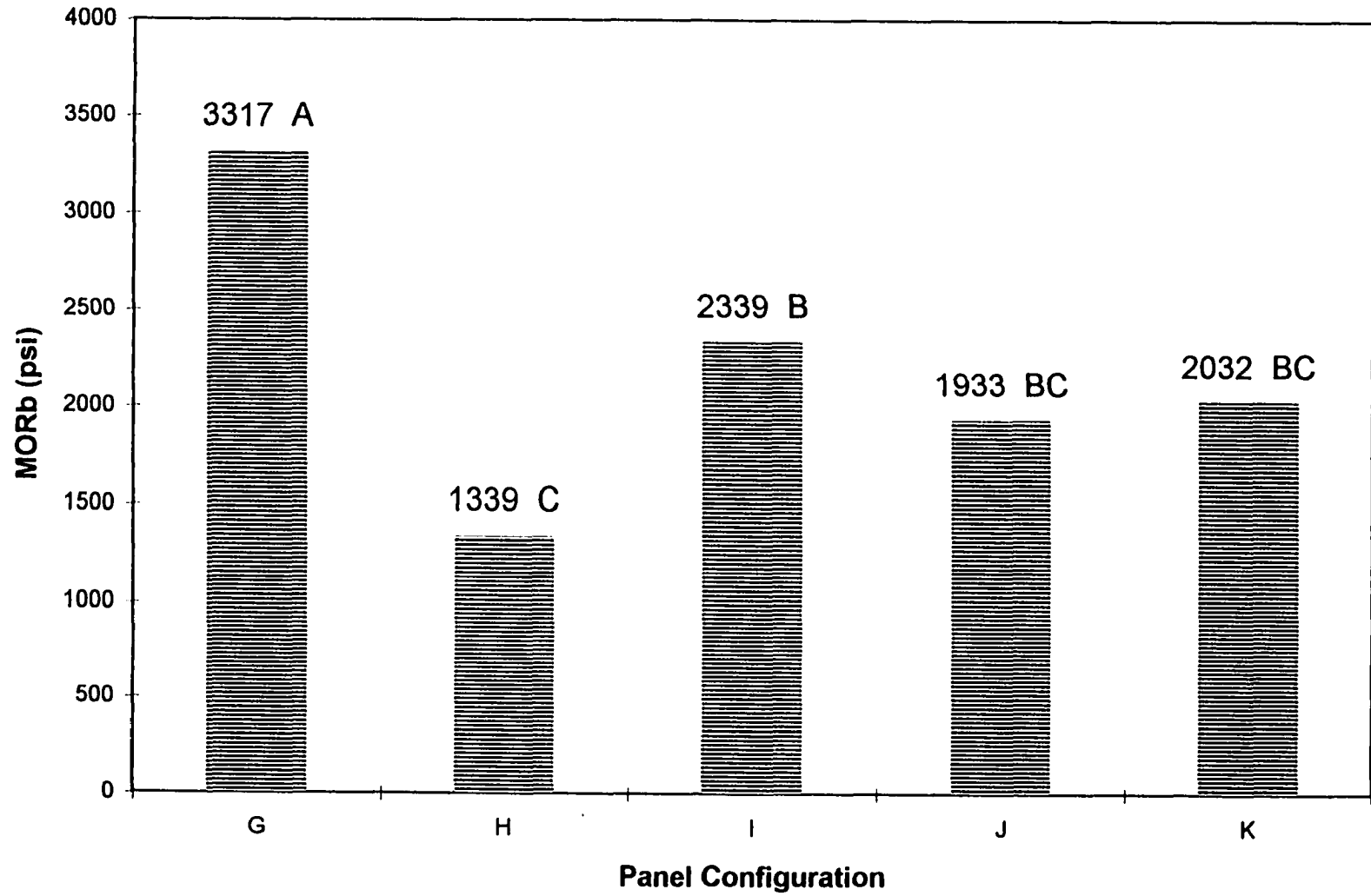


Figure 2.19. Modulus of rupture for wood fiber-polyethylene composites with various configurations.

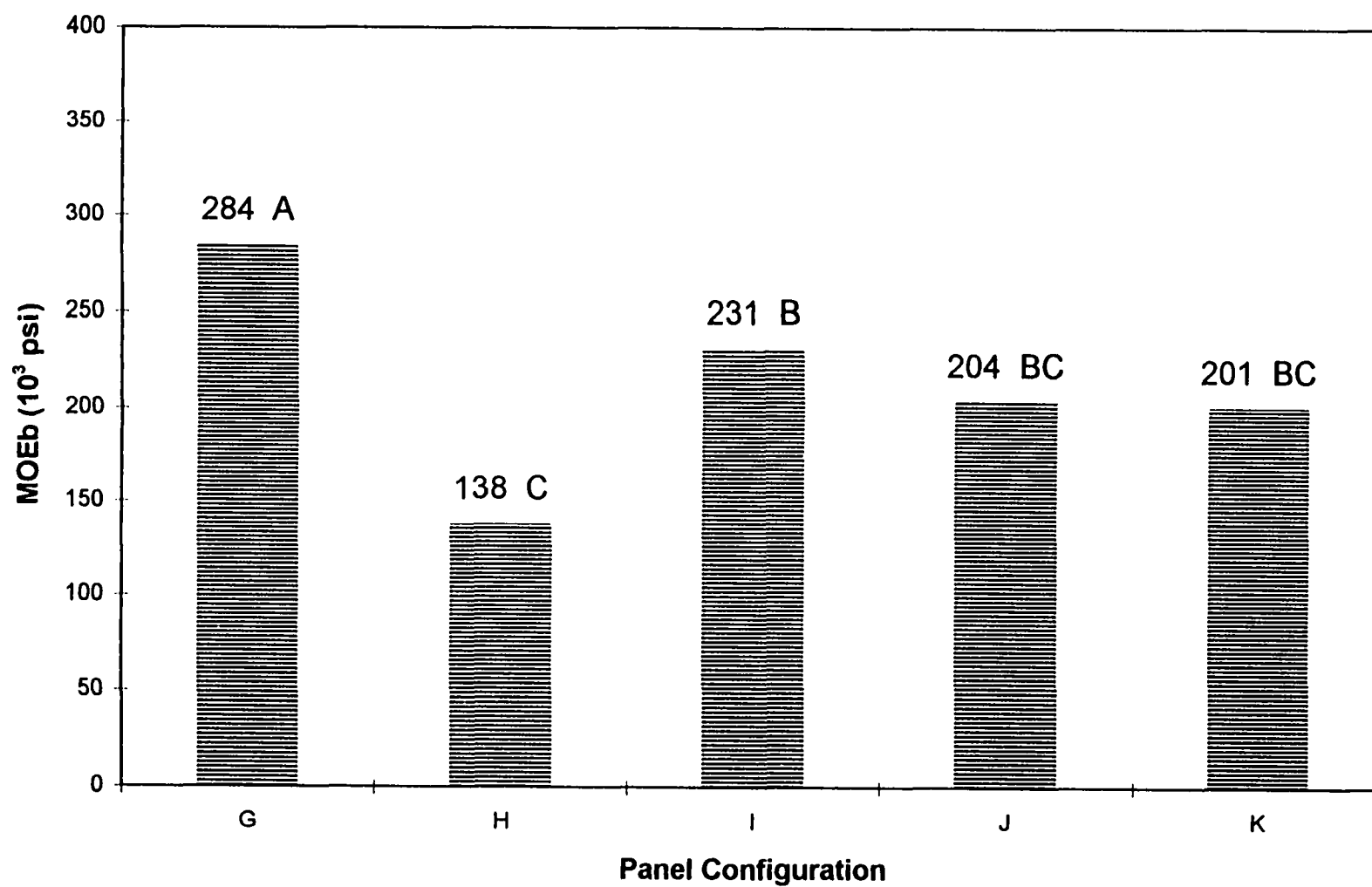


Figure 2.20. Modulus of elasticity for wood fiber-polyethylene composites with various configurations.

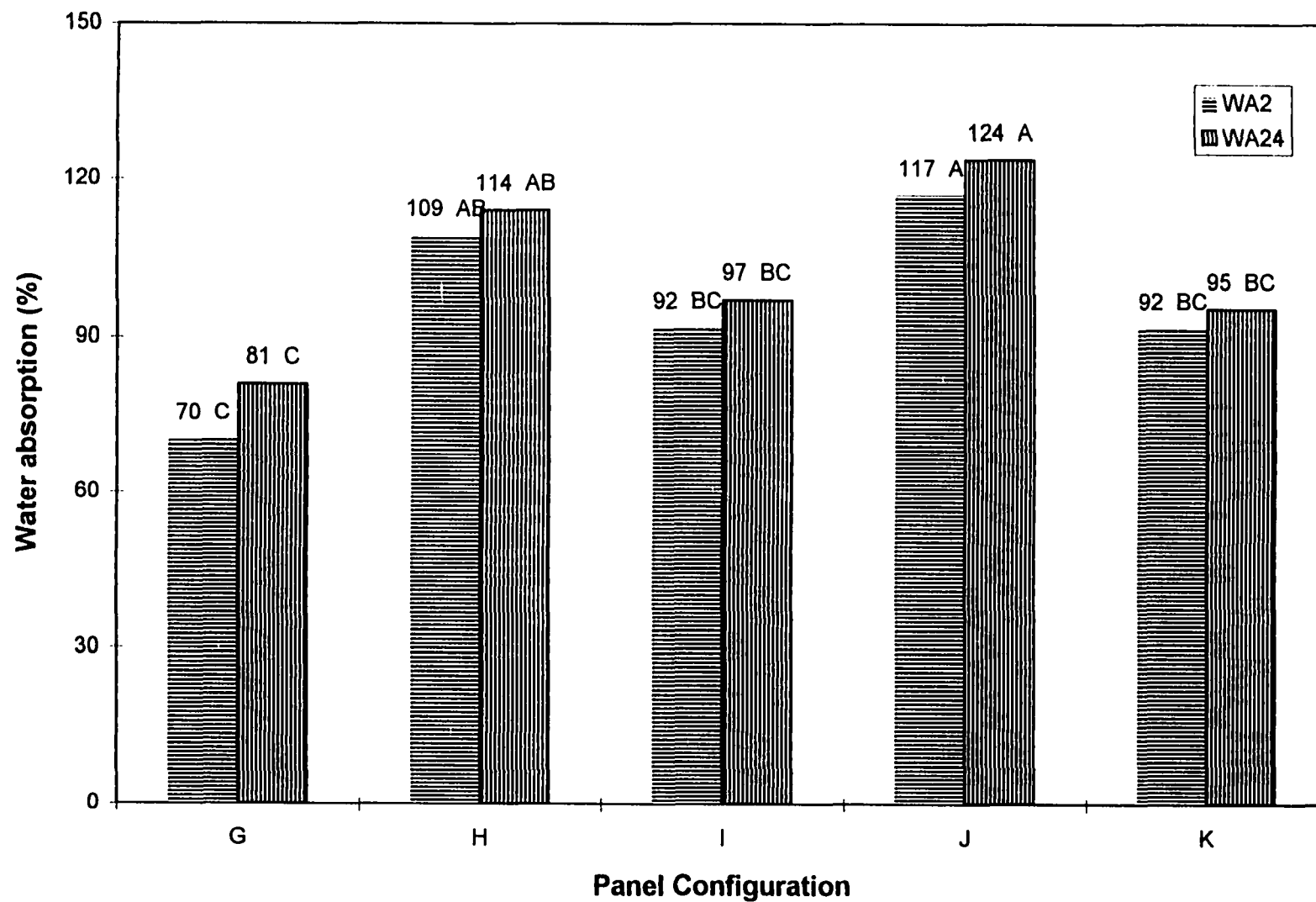


Figure 2.21. Water absorption for wood fiber-polyethylene composites with various configurations.

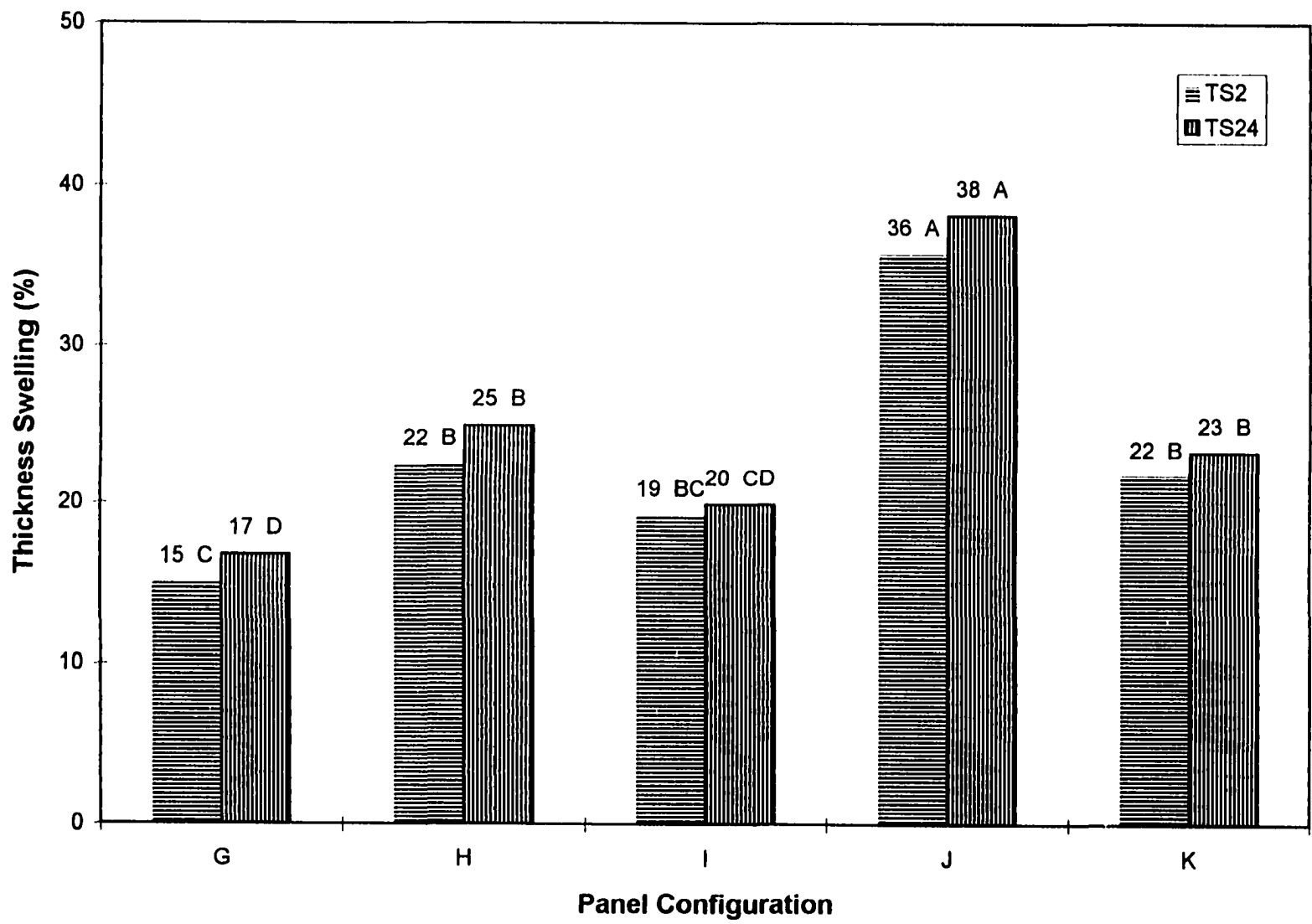


Figure 2.22. Thickness swelling for wood fiber-polyethylene composites with various configurations.

The effects of virgin fiber ratio on panel strength properties have been described previously in Section 2.3.2.1. The influence of adding recycled PE can be evaluated by comparing scheme G with treatments A and B. The IB, MOR_b and MOE_b of G were lower than those in A but slightly higher than those in B (except MOE_b). These values also indicate that the impact of adding PE was more serious in MOR_b and MOE_b than in IB. Scheme G passed the commercial standard for class 4 -service hardboard and scheme I conformed with class 5 - industrialite hardboard. Replacing 20% of recycled fiber with PE (scheme H vs. treatment F) showed an improvement in IB (41 vs. 22 psi) and MOR_b (1339 vs. 1051 psi); but these hardboards still failed to meet commercial standards.

The purpose of setting up layered structures was to improve the bending properties of the resultant panels. However, the effect was not significant within the three-component system (i.e., VIR, OCC1, and PE). A comparison within the two-component system (i.e., VIR and OCC1) shows that the layered structure (J) is inferior to the homogeneous structure (C). The cause of this phenomenon was probably due to layered structure that altered the densification of furnish during hot pressing.

In contrast to homogeneous configuration, the layered structure with 10% virgin fiber on each face was less sensitive to the addition of PE. When 20% virgin fiber was replaced with PE, scheme K had comparable strength properties as scheme J. Both K and J passed the standards of class 5-industrialite hardboard.

2.3.2.2.2. Dimensional Stability (WA and TS)

In dimensional stability, WA2, WA24, TS2, and TS24 also shared a similar trend from high to low: J - H - (K or I) - G (Figures 2.21 and 2.22). The phenomenon indicates the following:

(1) at given PE content and homogeneous configuration (G vs H vs I), WA and TS were related to virgin fiber ratio,

(2) for the same material composition (I vs K), homogeneous or layered structure did not affect dimensional stability,

(3) within the same layered structure (J vs K), replacing 20% virgin fiber with PE significantly improved the dimensional stability of fiberboard.

As mentioned in the previous section, increasing the amount of recycled fiber leads to inferior dimensional stability. In the three-component system, the analogous trend still held. Within the two-component system, replacing hydrophilic fibrous material with hydrophobic PE (A vs G and F vs H) improved dimensional stability. The results are in agreement with Wu *et al.* (1994). The reasons for that may be due to the contribution of hydrophobic PE which blocks the voids inside panel, resulting in reduction of water uptake and restraint in thickness swelling. Moreover, poor wettability of PE leads to excess adhesive available for wood fibers, which enhances the bonding property and hence the dimensional stability of the resulting panels.

The difference between dimensional stability of homogeneous and layered configurations may not be considerable. The addition of PE into a layered

structure improved the WA and TS of fiberboard. Panels without PE (scheme J) showed the lowest dimensional stability. Despite the lack of oil-tempering and heat treatments, all panels with PE showed satisfactory enhancement in thickness swelling which conforms to commercial requirements of Standard hardboard.

2.3.2.3. Effects of Some Processing Factors on the Properties of Wood Fiber-Polyethylene Composites

A completely randomized design with factorial treatment arrangement was devised to investigate the effect of hot press temperature (TEMP), time (TIME) and nominal panel density (PCF) on the physical and mechanical properties of wood fiber-polyethylene composites. The target panels belonged to a three-component system with formulation VIR:OCC1:PE=40:40:20. Table 2.13 reports the mean values of all properties for each treatment combination. The table shows that average moisture contents for all treatment combinations were between 2.3 and 5.2%, and the actual panel densities were somewhat lower than the nominal density. The results of two-way ANOVA are summarized in Table 2.14.

2.3.2.3.1. Mechanical Property (IB, MOR_b and MOE_b)

A three-way ANOVA (Table 2.14) detected TEMP and PCF main effects, but did not reveal any interaction. Figure 2.23 shows that the IB was highest at 350°F, but was the same at 400 and 450° F.

Thermal decomposition of wood constituents at elevated temperature has been reported by literature (Stamm 1964, Suchsland and Woodson 1986, Fengel

and Wegener 1984), hence it may be responsible for the drop in IB. In addition, under the influence of fiber geometry, the fibers tend to lay horizontally during mat-forming, resulting in parallel-to-panel-surface fiber orientation in the panels. Consequently, IB not only indicates bonding strength between fiber and resin, but also reflects tensile strength perpendicular to fibers and fiber bundles. The reduction of IB may also be due to: (1) the presence of microfissures, which initiated wood failure during tensile tests, (2) the degradation of adhesive at elevated temperatures, and (3) the precure of adhesive before density profile develops.

The effect of panel density is shown in Figure 2.24. The increase in IB with density is reasonable, because at a given wood density, increasing panel density results in higher compaction ratio, which in turn enhances the strength properties of the panel.

Table 2.14 and Figures 2.25 and 2.26 showed that both MOR_b and MOE_b were only affected by panel density, and not by temperature and time. Generally, the strength properties of wood are reduced as a result of interactions of temperature-time-moisture content in the air (Stamm 1964, Shimizu *et al.* 1968, Hills 1984, Mitchell 1987). However, Stamm (1964) reported that the lost of MOR_b during the 5-day period at 200°F under atmospheric condition was merely about 0.5% for softwoods, and even less for hardwoods. Moreover, due to the viscoelastic nature of wood, deformation in tensile perpendicular to panel surface is less than those of bending and compression. Therefore, at elevated

Table 2.13. Physical and mechanical properties of wood fiber-polyethylene composites with VIR:OCC1:PE=4:4:2 at various processing conditions.

Treatment	L	M	N	O	P	Q
TEMP ¹ (°F)	350	350	350	350	400	400
PCF ¹	50	60	50	60	50	60
TIME ¹	5.0	5.0	7.5	7.5	5.0	5.0
MC (%)	5.22 (0.38) ²	4.06 (0.27)	3.92 (0.11)	3.36 (0.07)	3.79 (0.27)	3.34 (0.21)
Density (pcf)	48.81 (2.33)	53.27 (2.04)	47.47 (1.94)	54.72 (0.42)	48.74 (2.35)	55.96 (2.88)
IB (psi)	67.80 (0.36)	102.53 (7.68)	85.13 (20.21)	92.33 (21.38)	56.63 (10.91)	75.43 (6.44)
MOR _b (psi)	2221 (418)	3019 (834)	1619 (298)	3860 (1280)	1922 (272)	3597 (904)
MOE _b (10 ³ psi)	228 (50)	300 (76)	181 (30)	367 (135)	199 (20)	341 (79)
WA2 (%)	68.59 (8.38)	55.01 (5.33)	70.56 (8.40)	45.15 (5.21)	67.40 (9.91)	43.16 (9.32)
WA24 (%)	75.78 (7.99)	60.39 (4.85)	76.11 (7.51)	54.31 (4.06)	72.11 (9.48)	52.06 (7.26)
TS2 (%)	18.21 (0.87)	17.56 (0.99)	16.98 (0.57)	16.60 (1.40)	16.90 (0.98)	15.23 (1.57)
TS24 (%)	18.66 (0.52)	18.58 (0.65)	17.79 (0.41)	18.05 (0.99)	17.67 (1.41)	17.30 (0.38)

----- To be continued -----

----- Table 2.13. continued -----

Treatment	R	S	T	U	V	W
TEMP (°F)	400	400	450	450	450	450
PCF	50	60	50	60	50	60
TIME (min)	7.5	7.5	5.0	5.0	7.5	7.5
MC	3.11	2.38	3.28	2.91	3.24	3.35
(%)	(0.38)	(0.20)	(0.21)	(0.10)	(0.09)	(0.52)
Density	50.51	57.19	46.46	54.51	49.64	57.28
(pcf)	(4.15)	(2.08)	(3.23)	(0.95)	(1.74)	(1.08)
IB	50.20	76.83	56.03	77.60	48.10	82.70
(psi)	(5.60)	(1.18)	(4.01)	(11.00)	(3.86)	(5.78)
MOR _b	2100	4182	2662	3760	2121	3382
(psi)	(419)	(779)	(432)	(64)	(109)	(146)
MOE _b	214	438	278	403	239	403
(10 ³ psi)	(51)	(75)	(46)	(19)	(11)	(25)
WA2	59.85	40.76	73.72	53.34	60.22	41.41
(%)	(16.29)	(8.94)	(14.37)	(2.22)	(5.66)	(8.65)
WA24	67.42	49.89	78.49	57.51	66.87	50.47
(%)	(14.07)	(5.26)	(13.68)	(2.20)	(3.43)	(3.49)
TS2	16.57	16.41	16.54	16.72	15.46	15.93
(%)	(0.45)	(1.05)	(1.83)	(1.06)	(0.28)	(2.31)
TS24	18.39	18.98	17.80	18.19	16.91	18.76
(%)	(0.10)	(0.24)	(2.01)	(0.96)	(0.46)	(1.54)

¹. TEMP, PCF, TIME denote hot press temperature (°F), nominal panel density (pcf), and hot press time (min), respectively

². Numbers in parentheses are standard deviations.

Table 2.14. Three-way ANOVA of some processing factors on properties of wood fiber-polyethylene composites with VIR:OCC1:PE=4:4:2.

Property/Source	DF	MS	F	P
IB				
TEMP	2	1855.77	17.08	0.0001
PCF	1	5150.45	47.41	0.0001
TIME	1	0.13	0.00	0.9722
TEMP*PCF	2	41.26	0.38	0.6881
TEMP*TIME	2	31.53	0.29	0.7507
PCF*TIME	1	11.11	0.10	0.7519
TEMP*PCF*TIME	2	365.44	3.36	0.0515
ERROR	24	108.64	---	---
MOR _b				
TEMP	2	330146.86	0.88	0.4272
PCF	1	20955032.11	55.94	0.0001
TIME	1	1820.44	0.00	0.9450
TEMP*PCF	2	366717.36	0.98	0.3902
TEMP*TIME	2	556346.19	1.49	0.2466
PCF*TIME	1	1011365.44	2.70	0.1134
TEMP*PCF*TIME	2	346978.36	0.93	0.4098
ERROR	24	374630.81	---	---
MOE _b				
TEMP	2	11508.02	3.01	0.0666
PCF	1	208689.84	55.11	0.0001
TIME	1	2132.73	0.56	0.4602
TEMP*PCF	2	2290.57	0.60	0.5542
TEMP*TIME	2	4375.74	1.16	0.3318
PCF*TIME	1	13816.32	3.65	0.0681
TEMP*PCF*TIME	2	1091.74	0.29	0.7521
ERROR	24	3786.46	---	---

----- To be continued -----

----- Table 2.14. continued -----

Property / Source	DF	MS	F	P
WA2				
TEMP	2	151.48	1.74	0.1971
PCF	1	3691.98	42.39	0.0001
TIME	1	468.51	5.38	0.0292
TEMP*PCF	2	4.50	0.05	0.9498
TEMP*TIME	2	68.81	0.79	0.4653
PCF*TIME	1	6.53	0.07	0.7866
TEMP*PCF*TIME	2	60.10	0.69	0.5113
ERROR	24	87.10	---	---
WA24				
TEMP	2	118.31	1.91	0.1703
PCF	1	3144.41	50.69	0.0001
TIME	1	244.56	3.94	0.0586
TEMP*PCF	2	0.03	0.00	0.9995
TEMP*TIME	2	38.41	0.62	0.5468
PCF*TIME	1	0.12	0.00	0.9654
TEMP*PCF*TIME	2	25.57	0.41	0.6668
ERROR	24	62.03	---	---
TS2				
TEMP	2	5.0467	3.26	0.0561
PCF	1	1.2027	0.78	0.3871
TIME	1	2.5921	1.67	0.2083
TEMP*PCF	2	1.2043	0.78	0.4710
TEMP*TIME	2	2.0800	1.34	0.2803
PCF*TIME	1	1.0747	0.69	0.4133
TEMP*PCF*TIME	2	0.3742	0.24	0.7874
ERROR	24	1.5501	---	---
TS24				
TEMP	2	0.3819	0.40	0.6778
PCF	1	1.7380	1.80	0.1924
TIME	1	0.1145	0.12	0.7337
TEMP*PCF	2	1.0477	1.08	0.3541
TEMP*TIME	2	2.9001	3.00	0.0686
PCF*TIME	1	1.8998	1.97	0.1737
TEMP*PCF*TIME	2	0.2363	0.24	0.7850
ERROR	24	0.9663	---	---

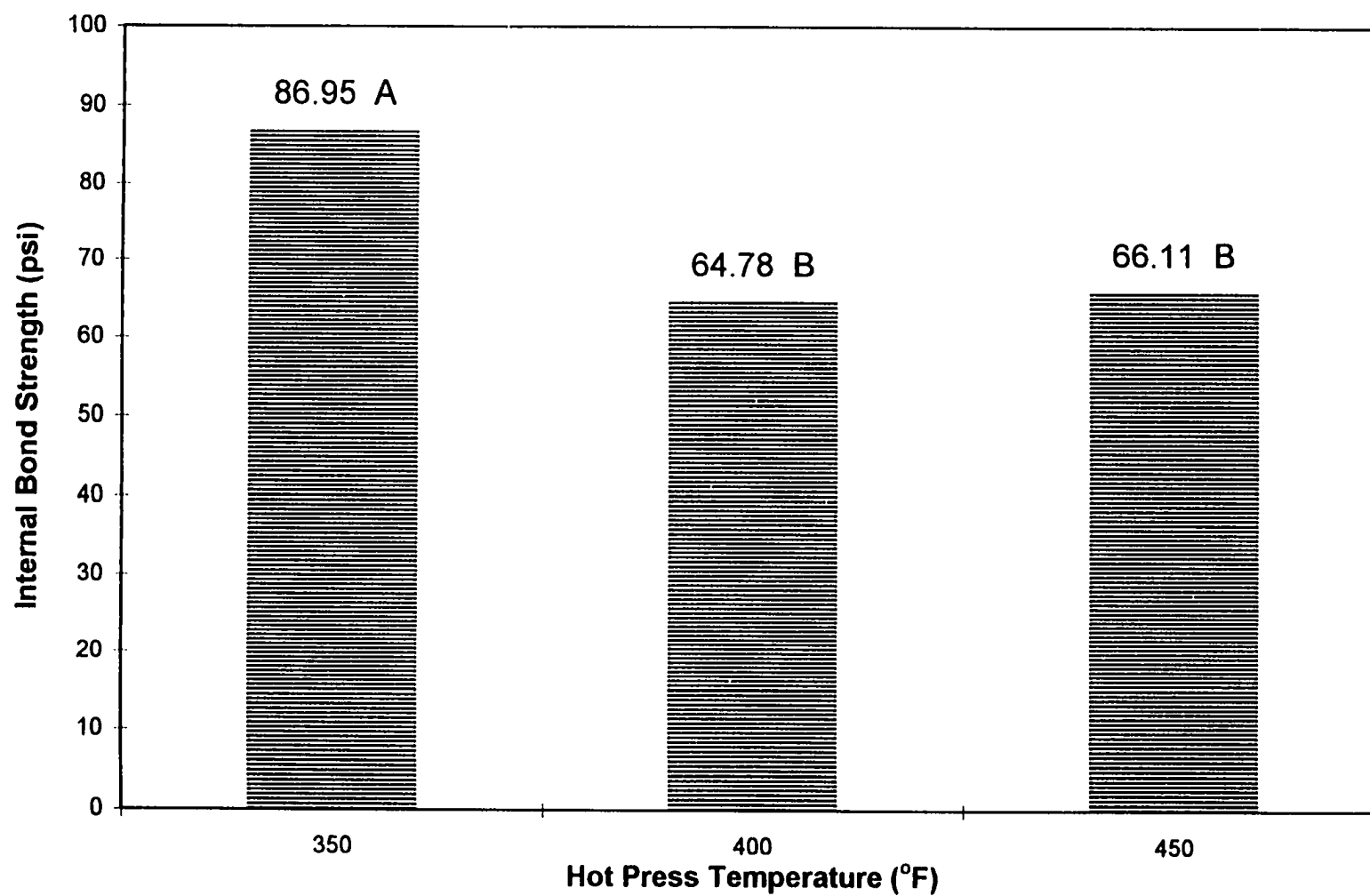


Figure 2.23. Effect of hot press temperature on internal bond strength of wood fiber-polyethylene composites.

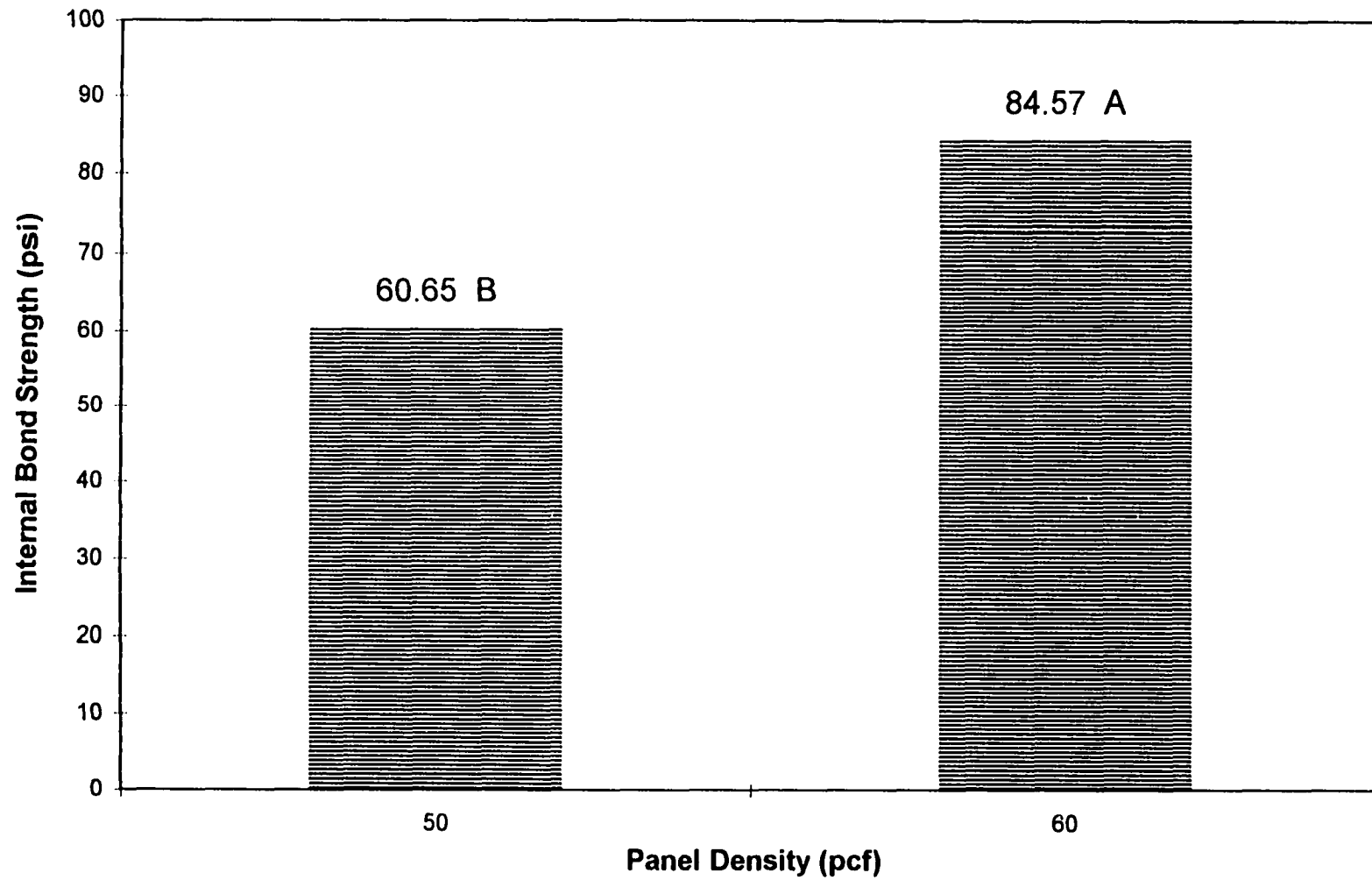


Figure 2.24. Effect of panel density on internal bond strength of wood fiber-polyethylene composites.

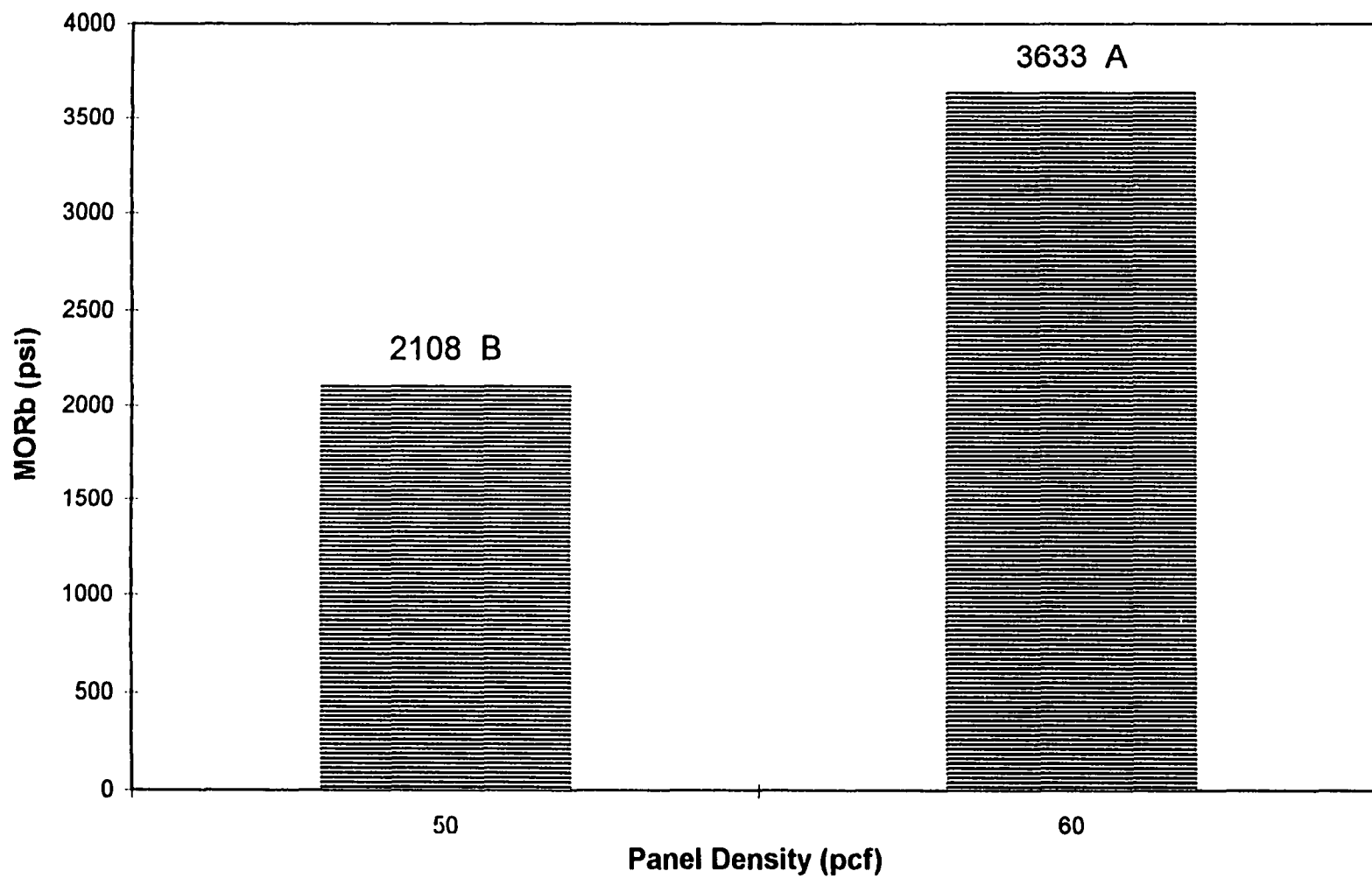


Figure 2.25. Modulus of rupture for wood fiber-polyethylene composites with virgin fiber : recycled fiber : PE=4:4:2 at two panel density levels.

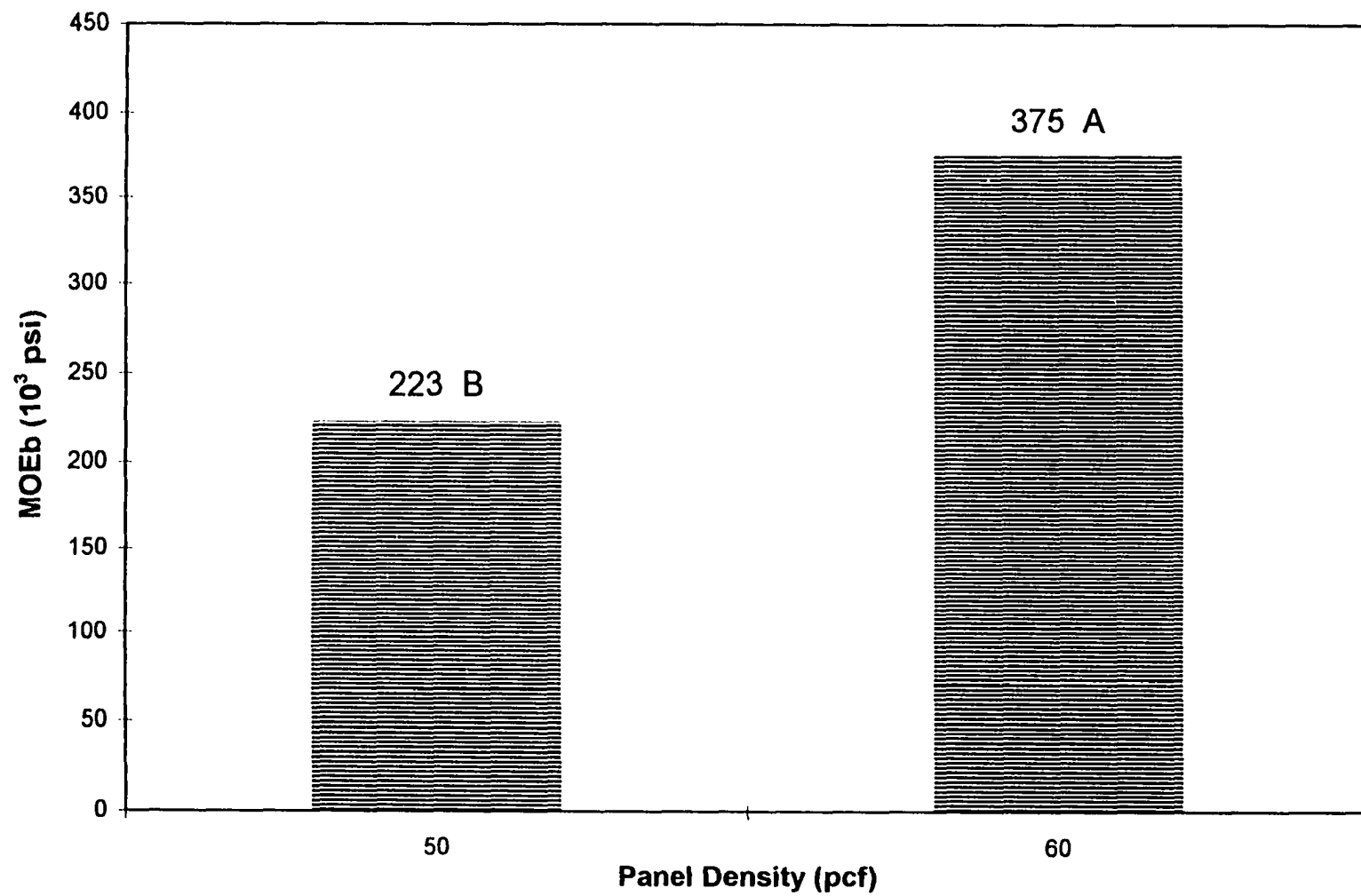


Figure 2.26. Modulus of elasticity for wood fiber-polyethylene composites with virgin fiber: recycled fiber : PE=4:4:2 at two panel density levels.

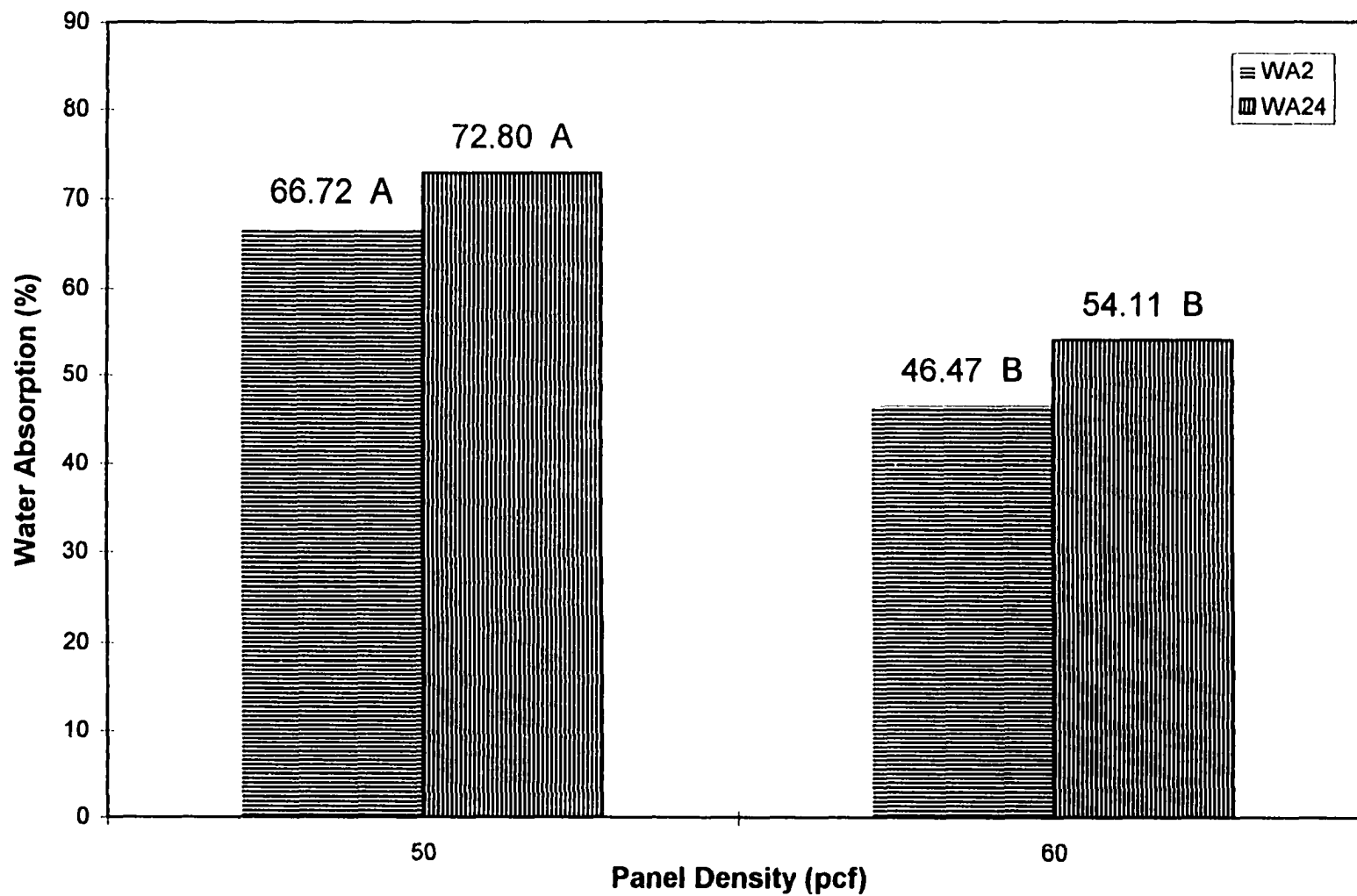


Figure 2.27. Effect of panel density on water absorption of wood fiber-polyethylene composites.

temperatures for as long as 7.5 min, the influence of temperature and time is more pronounced on IB than on the bending properties of hardboard.

2.3.2.3.2. Dimensional Stability (WA and TS)

In dimensional stability, both WA2 and WA24 were affected by PCF. The influence of TEMP was more pronounced in WA2 ($p=0.0292$) than in WA24 ($p=0.0586$). On the other hand, TS was not affected by TEMP, PCF, and TIME (Table 2.14). The effects of panel density on WA2 and WA24 are shown in Figure 2.27. Low density wood fiber-polyethylene composites tended to retain more water in immersion test. This phenomenon is understandable since low density panels have more voids which provide extra space to hold the absorbed water. However, it is not unusual for high density conventional particleboards to exhibit high thickness swelling (Maloney 1977). The effect of panel density on the TS of wood fiber-polyethylene composites was not significant. The reasons may be due to: (1) the volumetric ratio of hydrophilic wood fiber is much smaller than in conventional hardboard, (2) PE blocks the sorption sites of wood fiber, and (3) hydrophobic PE facilitates greater restraining force to offset the hygroscopic expansion of wood fiber. Even without oil-tempering and heat treatment, wood fiber-polyethylene composites showed satisfactory TS when compared with commercial standards.

2.3.2.3.3. Contact Angle

A split plot design with factorial treatment arrangement was applied to analyze the effect of hot press temperature (TEMP), hot press time (TIME),

nominal panel density (PCF), and wetting liquid type (LIQUID). The average contact angle values for all treatment combinations are shown in Table 2.15. The table reveals that contact angles measured by PF, UF and water fell between 89-105°, 96 -112°, and 95-108°, respectively. Results of ANOVA for split plot design (Table 2.16) indicates that the main effects of TEMP, TIME, and LIQUID were highly significant ($p < 0.01$) with no detectable interactions. Results of Tukey's tests are illustrated in Figures 2.28 - 2.30.

Table 2.15. Contact angle of wood fiber-polyethylene composites with VIR:OCC1:PE= 4:4:2 at different processing conditions.

Treat- ment	----- Wetting Liquid -----			Treat- ment	----- Wetting Liquid -----		
	PF	UF	WATER		PF	UF	WATER
	----- degree -----				----- degree -----		
L	89.21 (6.21)	98.89 (5.18)	95.92 (5.19)	R	98.59 (5.22)	105.87 (4.52)	103.80 (3.56)
M	89.18 (2.38)	96.88 (4.88)	95.34 (2.66)	S	100.69 (3.22)	109.46 (2.39)	104.88 (2.15)
N	90.76 (2.12)	100.03 (3.83)	98.66 (2.99)	T	102.36 (2.89)	108.73 (1.65)	108.19 (2.35)
O	92.94 (4.04)	100.69 (5.52)	100.03 (3.33)	U	98.10 (5.65)	106.91 (4.33)	104.85 (3.47)
P	96.76 (4.97)	104.92 (5.79)	103.44 (3.20)	V	105.36 (2.53)	111.98 (5.10)	107.78 (3.52)
Q	92.28 (2.94)	101.93 (2.68)	101.55 (2.47)	W	102.54 (4.22)	111.28 (6.14)	106.83 (4.01)

Contact angle of wood fiber-polyethylene composites was positively related to hot press temperature, i.e., wettability of panel decreases as temperature increases (Figure 2.28). On the other hand, hot press time also

negatively affected the wettability of wood fiber-polyethylene composites (Figure 2.29). The reduction of wettability after high temperature drying was attributed to surface inactivation (Christiansen 1990), hence the extent of wettability reduction can be related to heating temperature and time. Contact angles can be ranked from high to low as UF, WATER, and PF (Figure 2.30). Comparisons among three wetting liquids show similar trends with the aforementioned two-component system (Figure 2.16). However, contact angles in the three-component system were slightly higher than those in the two-component system because of the addition of PE.

Table 2.16. ANOVA of some processing factors on the contact angle of wood fiber-polyethylene composites with VIR:OCC1:PE=4:4:2.

Source	DF	MS	F	P
TEMP	2	4044.8542	35.15	0.0001
PCF	1	73.6726	0.64	0.4315
TIME	1	1072.8904	9.32	0.0055
TEMP*PCF	2	64.2831	0.56	0.5793
TEMP*TIME	2	9.5011	0.08	0.9210
PCF*TIME	1	260.0904	2.26	0.1458
TEMP*PCF*TIME	2	35.7938	0.31	0.7356
REP(TEMP*PCF*TIME)	24	115.0897	10.63	0.0001
LIQUID	2	2618.0109	241.73	0.0001
TEMP*LIQUID	4	14.6924	1.36	0.2485
PCF*LIQUID	2	4.3979	0.41	0.6666
TIME*LIQUID	2	29.9699	2.77	0.0641
TEMP*PCF*LIQUID	4	13.6882	1.26	0.2837
TEMP*TIME*LIQUID	4	22.7481	2.10	0.0802
PCF*TIME*LIQUID	2	3.0146	0.28	0.7572
TEMP*PCF*TIME*LIQUID	4	5.8255	0.54	0.7080
ERROR	372	10.8305		

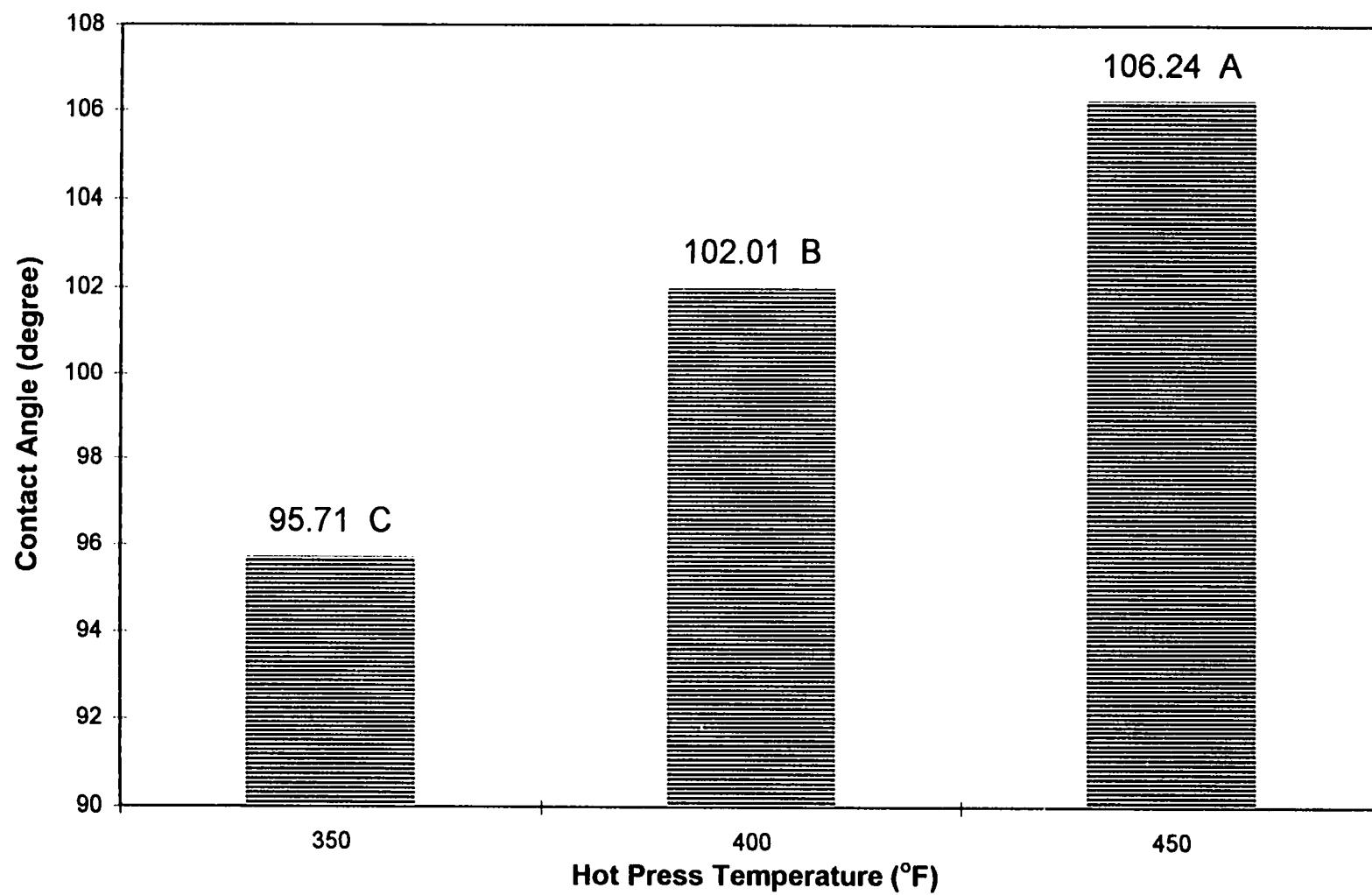


Figure 2.28. Effect of hot press temperature on contact angle of wood fiber-polyethylene composites.

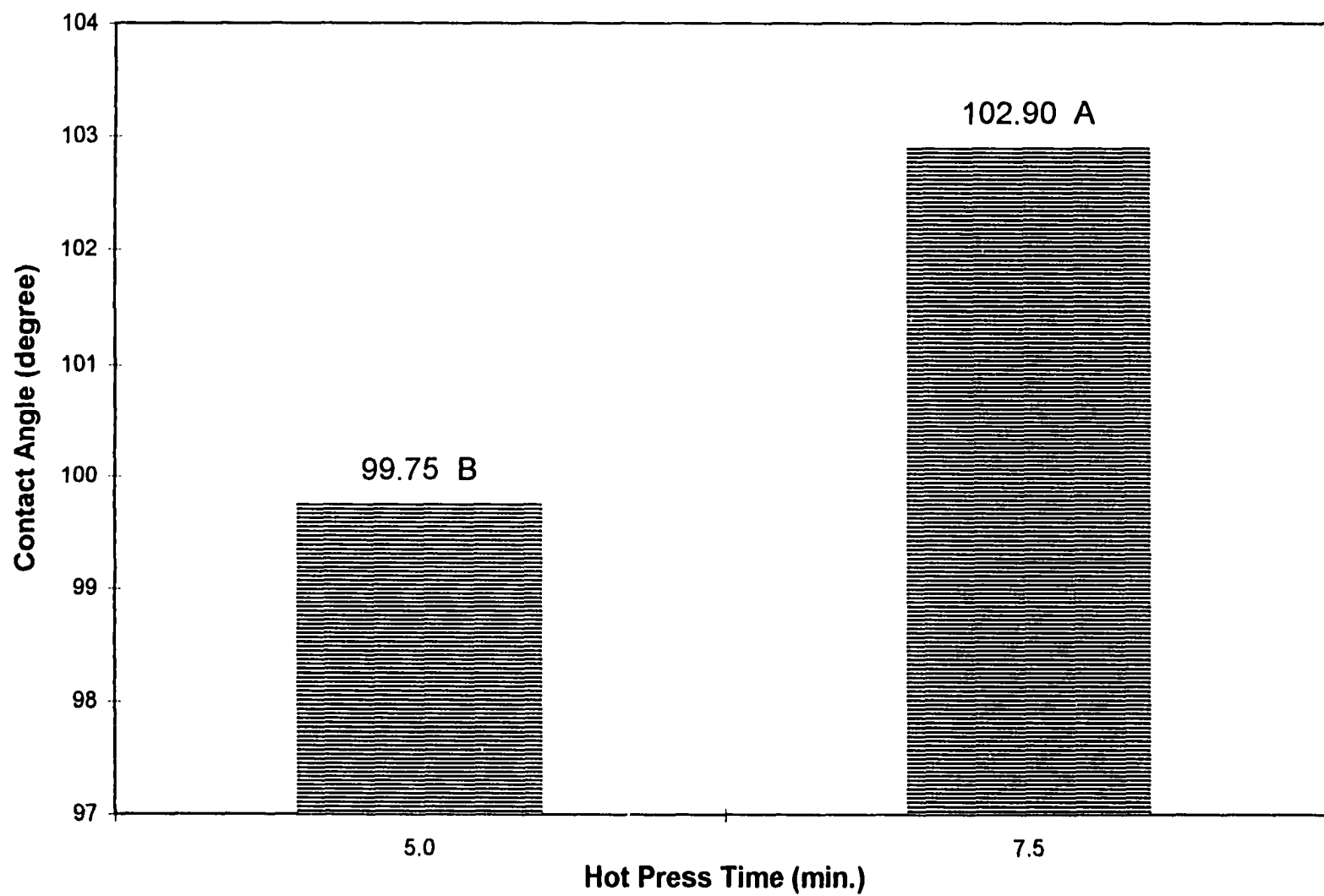


Figure 2.29. Effect of hot press time on contact angle of wood fiber-polyethylene composites.

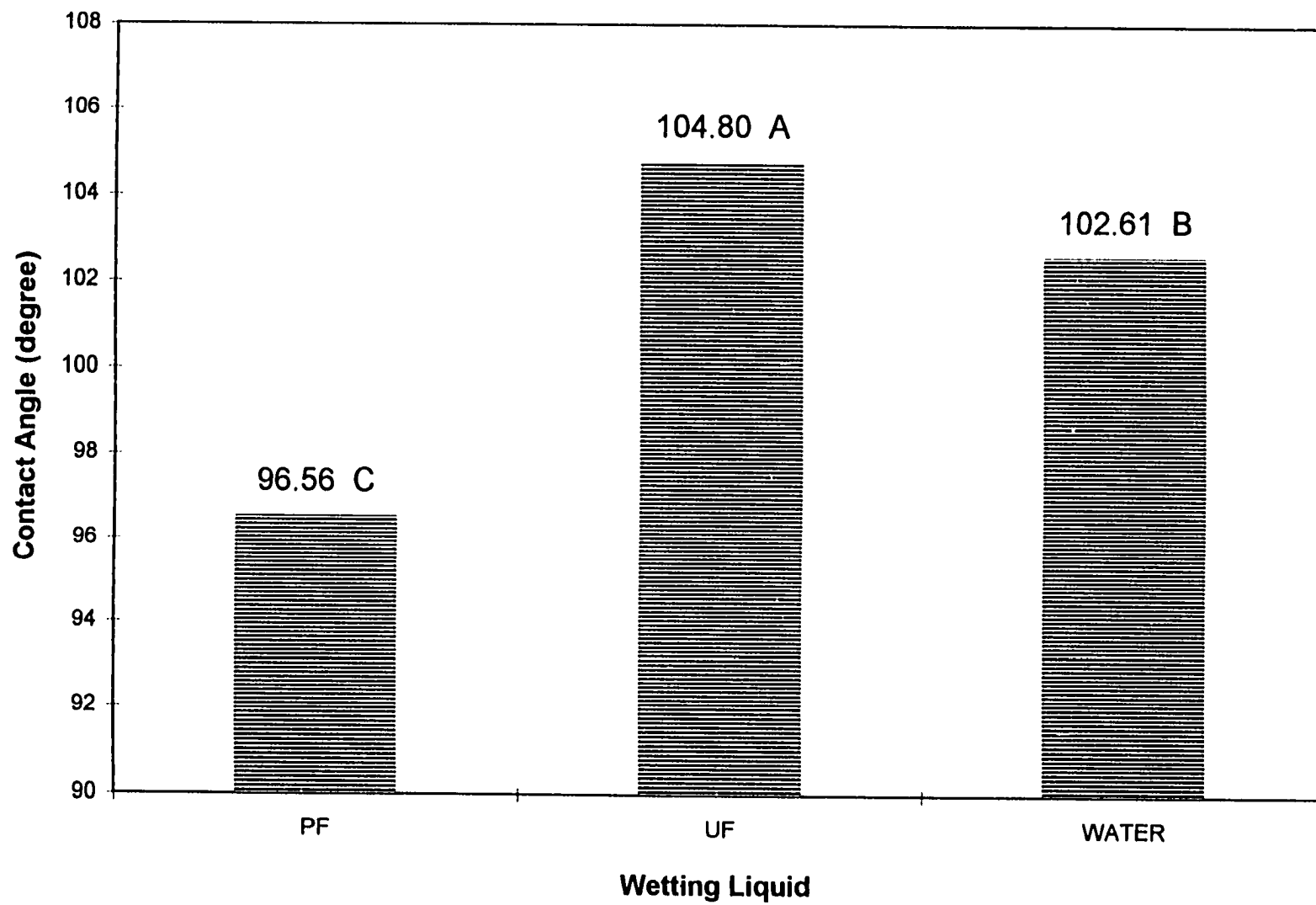


Figure 2.30. Effect of wetting liquid on contact angle of wood fiber-polyethylene composites.

2.4. CONCLUSIONS

Studies on chemical compositions and fiber measurement of virgin and recycled fibers from four sources showed significant differences among fiber types. Virgin southern pine fibers showed substantially higher alcohol-benzene extractive content and lignin content than the three types of recycled fiber. Recycled fibers from waste office paper and old corrugated cardboard had considerably higher holo- and alpha-cellulose contents than virgin fibers. Differences in chemical composition existed between fibers recycled from the two sources of old corrugated cardboard. Among the four fiber types, recycled office paper exhibited the lowest extractive and lignin contents and highest holo- and alpha-cellulose contents. Average fiber lengths of the three recycled fiber types were statistically the same, but appreciably shorter than virgin fiber. Average fiber length was reduced by disintegrating revolution.

Replacing virgin fiber with recycled fiber adversely affected physical and mechanical properties of fiberboard. Bending properties and dimensional stability were linearly dependent on virgin fiber ratios. At least 70% of the variations in these properties was attributed to fiber composition. Internal bond strength showed polynomial relationship with virgin fiber ratio. As high as 95% of the variations in fiberboard IB can be explained by fiber composition. Wettability of fiberboard, as measured by contact angle, decreased as recycled fiber increased. Based on strength properties, panels with 20% and 40% recycled fiber contents conformed to standards for class 4-service and class

5-industrialite hardboard, respectively. All panels with recycled fiber content greater than 40% failed to meet any commercial requirement.

Five schemes were devised to investigate the effects of panel configuration and polyethylene filler on the properties of wood fiber-polyethylene composites. With fixed polyethylene content (20%) and homogeneous configuration, IB, MOR_b, MOE_b, and dimensional stability were directly related to virgin fiber ratio. Layered structure showed similar strength properties and dimensional stability with homogeneous configuration. However, using layered structure with 40% recycled fibers, reducing the virgin fiber ratio from 60 to 40% by substituting 20% polyethylene, did not affect panel strength properties, but appreciably increased dimensional stability.

Three levels of hot press temperature, two levels of panel density, and two levels of hot press time were formulated to examine the responses in strength properties and dimensional stability of wood fiber-polyethylene composites. Experimental results indicated that, at a given composition, i.e., virgin fiber : recycled fiber : polyethylene=4:4:2, internal bond strength was affected by both hot press temperature and panel density; whereas bending properties and water absorption were altered by panel density. Thickness swelling, however, was unaffected. Hot press temperature and hot press time noticeably affected wettability of wood fiber-polyethylene composites. Taking into account all the properties tested, the most appropriate processing conditions for wood fiber-polyethylene composites with 4:4:2 composition was hot pressing at 350°F for

5 min. with panel density of 60 pcf. Panels fabricated in such conditions passed specifications for commercial class 4-service hardboard.

2.5. LITERATURE CITED

American Society for Testing and Materials. 1994. Standard testing methods of evaluating the properties of wood-based fiber and particle panel materials. ASTM D 1037-94. Philadelphia, PA.

Brown, R.P. 1988. Handbook of Plastic Test Methods. pp. 303-304. John Wiley & Sons, New York.

Caulfield, D.F., J.A. Koutsky, and D.T. Quillen. 1993. Cellulose/polypropylene composites: the use of AKD and ASA sizes as compatibilizers. In: M.P. Wolcott (ed.), Wood-fiber/polymer composites: Fundamental concepts, processes, and material options: Proc. of 1st Wood Fiber-Plastic Composite Conference, Madison, WI. Forest Products Society, 1993: 128-134.

Christiansen, A.W. 1990. How overdrying wood reduces its bonding to phenol-formaldehyde adhesives: A critical review of the literature. Part I: Physical responses. Wood Fiber Sci. 22(4): 441-459.

Clemons, C. 1995. Exploratory microscopic investigation of impacted paper fiber-reinforced polypropylene composites. In: D.F. Caulfield, R.M. Rowell, and J. A. Youngquist (eds.), Wood fiber-Plastic Composites- Virgin and recycled wood fiber and polymers for composites: Proc. of the Woodfiber-Plastic Composites Conference, May 1-3, 1995, Madison, WI. Forest Product Society; 1995:173-179.

Dalvåg, H., C. Klason, and H.-E. Strömvall. 1985. The efficiency of cellulosic fillers in common thermoplastics. Part II. Filling with processing aids and coupling agents. Intern'l J. Polymeric Mater. 11:9-38.

Deppe, H-J. 1985. The utilization of wastepaper and refuse fiber material for particleboard and MDF. In: Proc. 19th International Particleboard Symposium, T.M. Maloney (Ed.), Washington State University. Pullman, WA, 19:127-143.

Environment Protection Agency. 1988. Characterization of municipal solid waste in the United States: 1960 to 2000 Update. Environmental Protection Agency, Washington DC.

Environment Protection Agency. 1990. Characterization of municipal solid waste in the United States: 1990 Update. Environmental Protection Agency, EPA/530-SW-90-042. Washington DC. 103p.

Fengel, D. and G. Wengener. 1984. Wood chemistry, ultrastructure, reactions. Walter de Gruyter, New York. p. 319-344.

Gatenholm, P., J. Felix, C. Klason, and J. Kubát. 1993. Methods for improvement of properties cellulose-polymer composites. In: M.P. Wolcott (ed.), Wood-fiber/polymer composites: Fundamental concepts, processes, and material options: Proc. 1st Wood Fiber-Plastic Composite Conference, Madison, WI. Forest Products Society, 1993: 20-24.

Hamilton, T.E., J.A. Youngquist. 1991. Alternative uses of recovered fibers. In: Focus '95+ Landmark paper recycling symposium proceedings; 1991 March 19-21; Atlanta, GA. TAPPI, 1991: 333-341.

Hills, W.E. 1984. High temperature and chemical effects on wood stability. Part I: General considerations. Wood Sci. Technol. 18:281-293.

Hon, D.N. -S. and S.T. Sean. 1993. Composites from newsprint fiber and polyolefin: their processability and properties. In: M.P. Wolcott (ed.), Wood-fiber/polymer composites: Fundamental concepts, processes, and material options: Proc. of 1st Wood Fiber-Plastic Composite Conference; Madison, WI. Forest Products Society, 1993: 63-67.

Kishi, H., M. Yoshioka, A. Yamanoi, and N. Shiraishi. 1988. Composites of wood polypropylenes I. Makuzai Gakkaishi 34(2): 133-139.

Klason, C., J. Kubát and H.-E. Strömvall. 1984. The efficiency of cellulosic fillers in common thermoplastics. Part 1. Filling without processing aids or coupling agents. Intern. J. Polymeric Mater. 10:159-187.

Koning, J.W.Jr. 1983. Corrugated fiberboard. In: R.E. Mark (ed.), Handbook of Physical and Mechanical Testing of Paper and Paperboard. Marcel Dekker, Inc. New York. Vol 1: 385-408.

Krzsik, A.M., J. A. Youngquist, J.H. Muehl, R.M. Rowell, P. Chow, and S.R. Shook. 1992. Dry-process hardboards from recycled newsprint paper fibers. In: R.M. Rowell, T.L. Laufenberg, and J.K. Rowell (eds.), Materials interactions relevant to recycling of wood-based materials: Proc. of Materials Research Society Symposium; 1992 April 27-29; San Francisco, CA. Materials Research Society, 1992: 73-79. Vol. 266.

Krzysik, A.M., J. A. Youngquist, R.M. Rowell, J.H. Muehl, P. Chow, and S.R. Shook. 1993. Feasibility of using recycled newspapers as a fiber source for dry-process hardboards. *For. Prod. J.* 43(7/8):53-58.

Laundrie, J.F. and J.D. McNatt. 1975. Dry-formed medium-density hardboards from urban forest materials. USDA Forest Ser. Res. Paper FPL 254.

Maldas, D. and B.V. Kokta. 1989. The effect of aging conditions on the mechanical properties of wood fiber-polystyrene composites: Chemithermomechanical pulp as a reinforcing filler. *Composites Sci. Tech.* 36:167-182.

Maldas, D. and B.V. Kokta. 1990a. Influence of phthalic anhydride as a coupling agent on the mechanical behavior of wood fiber-polystyrene composites. *J. Appl. Polym. Sci.* 41:185-194.

Maldas, D. and B.V. Kokta. 1990b. Effect of recycling on the mechanical properties of wood fiber-polystyrene composites. Part I: Chemithermomechanical pulp as a reinforcing filler. *Polymer Composites.* 11(2):77-83.

Maldas, D. and B.V. Kokta. 1990c. Effect of fiber treatment on the mechanical properties of hybrid fiber reinforced polystyrene composites. Part II: use of glass fiber and wood pulp as hybrid fiber. *J. Adhesion Sci. Technol.* 4(2): 89-97.

Maldas, D. and B.V. Kokta. 1991. Performance of treated hybrid fiber-reinforced thermoplastic composites under extreme conditions: Part I: Use of mica and wood pulp as hybrid fiber. *Polymer Degradation Stability* 31:9-21.

Maldas, D., B.V. Kokta, and C. Daneault. 1989a. Thermoplastic composites of polystyrene: Effect of different wood species on mechanical properties. *J. Appl. Polym. Sci.* 38:413-439.

Maldas, D., B.V. Kokta, and C. Daneault. 1989b. Influence of coupling agents and treatments on the mechanical properties of cellulose fiber-polystyrene composites. *J. Appl. Polym. Sci.* 37:751-775.

Maldas, D., B.V. Kokta, C. Daneault, and Z. Koran. 1990. Effect of extreme conditions on the mechanical properties of wood fiber-polystyrene composites I. Chemithermomechanical pulp as reinforcing filler. *DREVÁRSKY VÝSKUM* Zväzok 125 pp.1-37.

Maldas, D., B.V. Kokta, R.G. Raj, and C. Daneault. 1988. Improvement of the mechanical properties of sawdust wood fibre-polystyrene composites by chemical treatment. *Polymer* 29:1255-1265.

Maloney, T.M. 1977. Modern Particleboard and Dry-Process Fiberboard Manufacturing. p. 175-176. Miller Freeman Publications, San Francisco, LA.

McKeever, D.B. 1995. Resource potential of wood-based wastes in the United States. In: Proc. of 2nd Biomass Conference of the Americas: Energy, Environment, Agriculture, and Industry, August 21-24, 1995, Portland, OR. National Renewable Energy Laboratory, 1995: 77-87.

McKeever, D.B., J.A. Youngquist, and B.W. English. 1995. Sources and availability of recovered wood and fiber for composite products. In: Proc. 29th International Particleboard/Composite Materials Symposium, April 4-6, 1995, Pullman, WA. Washington State University, 1995:207-214.

Myers, G.E., I.S. Chahyadi, C. Gonzalez, and C.A. Coberly. 1993. Wood flour and polypropylene or high-density polyethylene composites: Influence of maleated polypropylene concentration and extrusion temperature on properties. In: M.P. Wolcott (ed.), Wood-fiber/polymer composites: Fundamental concepts, processes, and material options. Proc. 1st Wood Fiber-Plastic Composite Conference, Madison, WI. Forest Products Society, 1993: 49-56.

Raj. R.G. and B.V. Kokta. 1989. Compounding of cellulose fibers with polypropylene: Effect of fiber treatment on dispersion in the polymer matrix. J. Appl. Polym. Sci. 38: 1987-1996.

Raj. R.G, B.V. Kokta, and C. Daneault. 1990. Wood flour as a low-cost reinforcing filler for polyethylene: studies on mechanical properties. J. Mater. Sci.25: 1851-1855.

Raj. R.G. and B.V. Kokta. 1995. Effect of aging cycle on mechanical properties of HDPE-pretreated wood fiber composites. In: D.F. Caulfield, R.M. Rowell, and J. A. Youngquist (eds.), Woodfiber-Plastic Composites- Virgin and recycled wood fiber and polymers for composites. Proc. Wood Fiber-Plastic Composites Conference, May 1-3, 1995, Madison, WI. Forest Product Society, 1995:235-239.

Raj. R.G, B.V. Kokta, D. Maldas, and C. Daneault. 1989. Use of wood fibers in thermoplastics. VII. The effect of coupling agents in polyethylene-wood fiber composites. J. Appl. Polym. Sci. 37: 1089-1103.

Rowell, R.M. and S. Harrison. 1992. Fiber based composites from recycled mixed paper and magazine stock. In: R.M. Rowell, T.L. Laufenberg, and J.K. Rowell (eds.), Materials interactions relevant to recycling of wood-based materials. Proc. Materials Research Society Symposium, April 27-29, 1992, San Francisco, CA. Materials Research Society, 1992: 65-72. Vol. 266.

Rowell, R.M. and S.E. Lange. 1994. Effects of recycling on the properties of fiberboards made from recycled magazine paper. In: P.R. Steiner (ed.), Proc. 2nd Pacific Rim Bio-based Composites Symposium, November 6-9, 1994, Vancouver, Canada. University of British Columbia, 1994: 270-276.

Rowell, R.M., J.A. Youngquist, and D. McNatt. 1991. Composite from recycled materials. In: T.M. Maloney (ed.). Proc. 25th International Particleboard/Composite Materials Symposium, April 9-11, 1991, Pullman, WA. Washington State University, 1991; pp. 301-314.

Rowell, R.M., T. Rials, H. Spleter, D. Luneke, R.A. Arola, R. Narayan, P. Davis, J. Simonson, T. Friberg, D. White, and R. W. Hemingway. 1993. Opportunities for composites from recycled wastewood-based resources: a problem analysis and research plan. *For. Prod. J.* 43(1):55-63.

Sanadi, A.R., D.F. Caulfield, and R.E. Jacobson. 1997. Agro-fiber thermoplastic composites. In: R. M. Rowell, R. A. Young, and J.K. Rowell (eds.), *Paper and composite from agro-based resources*. Lewis Publishers, New York.

Sanadi, A.R., R.A. Young, C. Clemons, and R.M. Rowell. 1994. Recycled newspaper fibers as reinforcing fillers in thermoplastics: Part 1 - Analysis of tensile and impact properties in polypropylene. *J. Reinforced Plastics Composites*. 13:54-67.

Sean, S.T., B. Sanschagrin, B.V. Kokta, and D. Maldas. 1991. Effect of processing variables on the mechanical properties of compression molded polystyrene-wood fiber composites. *Angew Makromol. Chemie* 184:157-166.

Shimizu, K., F. Teratani, and K. Miyazuki. 1968. Effect of the thermal treatment on wood hemicellulose II. The change of xylan by heating. *Mokuzai Gakkaishi* 14(7):376-381.

Shupe, T.F. 1996. The effects of silvicultural treatments on certain mechanical and physical properties of Loblolly pine wood composites. Ph. D. dissertation, Louisiana State University, Baton Rouge, LA.

Stamm, A.J. 1964. Thermal degradation of wood and cellulose. *Indust. Eng. Chem.* 48(3):413-417.

Suchsland, O. and G.E. Woodson. 1986. Fiberboard manufacturing practices in the United States. USDA Forest Service, Agric. handbook 640. 263 p.

Takase, S. and N. Shiraishi. 1989. Studies on composites from wood and polypropylenes. II. *J. Appl. Polym. Sci.* 37:645-659.

Walters, C.S., J.T. Pfeffer, and P. Chow. 1977. Production of panelboard from residue left from the fermentation of organic waste for methane. *For. Prod. J.* 27(9):12-20.

Wenger, T.H., J.A. Youngquist, and R.M. Rowell. 1992. Opportunities for composites from recycled wood based resources. In: R.M. Rowell, T.L. Laufenberg, and J.K. Rowell (eds.), *Materials interactions relevant to recycling of wood-based materials: Proc. Materials Research Society symposium*, April 27-29, 1992, San Francisco, CA. Materials Research Society; 1992: 3-15. Vol. 266.

Woodhams, R.T., G. Thomas, and D.K. Rodgers. 1984. Wood fibers as reinforcing fillers for polyolefins. *Polymer Eng. Sci.* 24(15):1166-1171.

Wu, Y., C.Y. Hse, E.T. Choong, and C. Y. Hwang. 1994. Effect of resin variables and plastic components on wood-plastic composites. In: P.R. Steiner (ed.), *Proc. 2nd Pacific Rim Bio-based Composites Symposium*, November 6-9, 1994, Vancouver, Canada. University of British Columbia, 1994: 64-71.

Yam, K.L., B.K. Gogoi, C.C. Lai, and S.E. Selke. 1990. Composites from compounding wood fibers with recycled high density polyethylene. *Polymer Eng. Sci.* 30(11):693-699.

Youngquist, J.A., B.W. English, R.M. Rowell. 1993. Alternative uses for waste paper in wood-based composite products. In: *Proc. 1993 Recycling symposium*; February 28-March 4, 1994, New Orleans, LA. TAPPI Press, 1993: 359-365.

Youngquist, J.A., A.M. Krzysik, J.H. Muehl, and C. Carll. 1992. Mechanical and physical properties of air-formed wood-fiber/polymer-fiber composites. *For. Prod. J.* 42(6):42-48.

Youngquist, J.A., G.E. Myers, and T.M. Harten. 1992. Lignocellulosic-plastic composites from recycled materials. In: *Emerging technologies for materials and chemicals from biomass. Proc. of symposium*, August 26-31, 1992, Washington, DC. American Chemical Society; 1992. Chap. 4. ACS symposium series 476.

Youngquist, J.A., G.E. Myers, J.H. Muehl, A.M. Krzysik, and C.M. Clemons. 1995. EPA project summary: Composites from recycled wood and plastics. EPA/600/SR-95/003. 9 p.

CHAPTER 3

EFFECT OF COMPATIBILIZER ON THE WETTABILITY AND INTERFACE ADHESION OF WOOD-PLASTIC COMPOSITES

3.1. INTRODUCTION

Wood flour and fibers are excellent fillers for thermoplastics because of their low density, low cost, high strength and stiffness, desirable fiber aspect ratio, flexibility during processing, and biodegradability (Collier *et al.* 1995, Felix and Gatenholm 1991). However, satisfactory dispersion of wood fillers in the matrices of thermoplastics has always been a problem caused by the hydrophilic nature of wood and the hydrophobic nature of plastic (Felix *et al.* 1994). The surface characteristics of cellulosic fibers prohibit the formation of a durable interface in the plastic composites, causing failure in stress transfer from one phase to another. Therefore, to enhance the affinity between these two components, the surface properties of one or another must be modified. The modification mechanisms include: reconstructing physical structure, changing surface topography, altering chemical nature of the surface, and removing weak boundary layer (Gent and Hamed 1990).

The compatibility of wood and plastic can be improved by introducing a compatibilizer into the system. Bifunctionality of compatibilizers are advocated to increase the adhesion between the interface of wood and plastic, and thus enhance stress transfer. Maleated polypropylene (MAPP) is the most commonly used compatibilizer for polypropylene and polyethylene polymers. The mechanisms of this compatibilizer have been proposed by Felix and Gatenholm

(1991), Kishi *et al.* (1988), Woodhams *et al.* (1984), and Sanandi *et al.* (1997).

The carboxylic groups in MAPP provide covalent and hydrogen bonding to the hydroxyl groups of cellulosic fiber surface. The long chains of MAPP, on the other hand, entangled the matrix of polypropylene, permitting segmental crystallization.

In the manufacture of natural fiber-thermoplastic composites, it is necessary to understand the improvement of interfacial bonding between wood and plastic after surface treatment in order to provide guidelines in designing strength properties of the final products. Most of the research has focused on indirectly investigating the mechanical performance of the composite materials. However, enhancement of interfacial properties can be evaluated directly by studying the adhesion of interphase using a model composite.

The basic requirements of adhesion between a solid substrate and a liquid adhesive rely on three major characteristics of adhesive: wettability, solidification, and sufficient deformability to reduce the buildup of elastic stress during the formation of joint (Zisman 1976). Therefore, to understand the effect of compatibilizer on the adhesion between wood and plastic, it is necessary to characterize the interrelationship in terms of wettability of liquid ingredients in wood-plastic composites as well as to examine the improvement of bonding strength advocated by using a compatibilizer.

Recently, dynamic contact angle has been used to evaluate the wettability of heat-treated cellulose fibers and acetylated cellulosic fibers, and to correlate

wettability with interfacial shear strength (Liu 1994, Liu *et al.* 1994). This technique provides insights to the knowledge of interphase properties of wood and thermoplastics. However, the data is inconclusive due to the small sample size. Interfacial shear strength between MAPP and wood has also been studied by Sanadi and co-workers (Sanadi *et al.* 1993). They modified the traditional single-fiber pull-out technique to minimize difficulties caused by using fragile and deformable natural fibers and to correctly assess interfacial bond strength. The results obtained have provided valuable information pertaining to the development of interfacial bond strength assessment methodology.

Research on interfacial adhesion needs to incorporate experiment designs that eliminate the influence of biological variation that is inherent in natural cellulose fibers. This is the major reason that this study was undertaken.

Accordingly, the purpose of this study was to examine the effect of maleated polypropylene compatibilizer on the interfacial properties of wood and polyolefins. This study is divided into two parts. In the first part, the effect of MAPP treatment on wettability of birch plywood was investigated. The wettability of polyolefin polymers by several wetting liquids and MAPP was also included. In the second part, a split plot design was employed and the modified single-fiber pull-out tests were performed to investigate the interrelationships between a four-component system of wood-adhesive-compatibilizer-plastic, simulating the interphase of a wood-thermoplastic composite.

3.2. LITERATURE REVIEW

3.2.1. Effect of Maleated Polypropylene

Maleated polypropylene (MAPP) can be used as a low molecular-weight additive (compatibilizer) in a polymer blend system, as well as a high molecular weight substitute for polypropylene. Numerous studies have investigated the effects of MAPP on the performances of wood-polypropylene composites. Most research showed positive effects in adding MAPP to the multiphase system.

Using MAPP as a low molecular weight additive, Gatenholm *et al.* (1993) confirmed the effects of compatibilizer from qualitative micrographs of fracture surface and crystallization for treated fibers, and also from quantitative improvement of mechanical properties of the composites. Myers *et al.* (1993) investigated the effects of MAPP concentrations on the performances of wood flour-polypropylene (WF-PP). They found that adding MAPP greatly improved bending strengths and moduli, tensile strength, and notched impact energy at all extrusion temperature levels. Czvikovszky *et al.* (1993) reported increased modulus, but not ultimate strength, of the MAPP treated wood fiber-reinforced plastic composites. However, little or no advantage was gained by using more than 1 to 2 percent MAPP. Similar results were also reported with other wood fiber-thermoplastic composites (Woodhams *et al.* 1993, Selke *et al.* 1993) and with agricultural fiber-reinforced thermoplastic composites (Jacobson *et al.* 1995, Karmaker and Youngquist 1995, Sanadi *et al.* 1995).

Contradictory to these reports, experiments conducted by Kolosick (1993) showed no improvement of peeling force, and no microscopic trace of nucleation in MAPP treated wood-polypropylene composites. Tai and coworkers (Tai *et al.* 1992) reported that adding 4% MAPP to Douglas-fir bark fibers led to a positive effect, but higher concentration (20%) of MAPP resulted in a poor interface, which they speculated was due to weak boundary layers created by the low molecular weight MAPP.

Using MAPP as a high molecular weight substitute for polypropylene, Mohanakrishnan *et al.* (1993) grafted maleic anhydride functional groups to polypropylene, and then blended it with three different types of low-density hardwood pulps using the reactive extrusion processing technique. The composites showed evidence of improved interface in SEM micrographs and satisfactory mechanical improvements with an increase of 108% in tensile modulus for treated polymer. Collier *et al.* (1995), using simultaneous and sequential compounding of wood fiber with MAPP, found improvements of tensile strengths over neat polypropylene. Both of them found that the maleation process which resulted in chain scission and molecular weight reduction suffered the decrease in strengths of modified polymers over unmodified ones.

The results of aforementioned studies imply that chemical properties, application techniques, and processing methods of MAPP may affect the effectiveness of MAPP in a thermoplastic matrix.

3.2.2. Wettability of Wood

Contact angle is an adverse measure of wettability (Zisman 1964, Zisman 1976). It is thermodynamically determined by the balance between liquid-solid adhesive forces and cohesive forces within the liquid (Johnson and Dettre 1993). In other words, contact angle is a quantitative measure of the physical interaction between liquid and solid (Hodgson and Berg, 1988). It is defined by the Young-Dupré equation, which relates the surface tension of a liquid (γ_{lv}) to the surface free energy of solid-vapor (γ_{sv}) and solid-liquid (γ_{sl}) interfaces (Zisman, 1964, Zisman 1976, Johnson and Dettre, 1993). That is:

(Eq. 3.1)

$$\cos\theta = \frac{\gamma_{sv} - \gamma_{sl}}{\gamma_{lv}}$$

A direct optical method has been used to measure the contact angle of liquids against wood surfaces. Freeman (1959) determined the relationships of physical and chemical properties of wood with bonding quality. He found significant linear dependence of shear ratio, shear strength and wood failure with $\cos\theta$. In another study, he found a positive correlation between average slip load and $\cos\theta$ of species with various wettabilities (Freeman and Wangaard 1960). Gray (1962) estimated the surface tensions for different wood species and found the linear relationship between critical surface tension and $\cos\theta$, derived by Zisman (1964), was valid in all wood species tested.

Hse (1972) first measured the contact angle of heterogeneous liquid adhesives on southern pine veneer surface. He found a linear relationship between contact angle of phenolic adhesives and glue bond shear strength, wood failure, and delamination. However, Jordan and Wellons (1977) attempted to relate the wettability and gluability for four Southeastern Asian hardwood veneer species but found inconsistent responses.

Wood is a composite of cellulose, hemicellulose and lignin, therefore, its wettability is affected by the individual ingredients. Regenerated polymer films have been synthesized to measure the contact angle of each wood ingredient. Kikata and Masuda (1969) measured the contact angle of a lignin-carbohydrate complex film and found a linear relationship between liquid surface tension and $\cos\theta$. Similar results were reported by Luner and Sandell (1969), who found that both cellulose and hemicellulose behaved as low-energy surfaces similar to polyethylene and nylon. Lee and Luner (1972) studied the wettability of different lignin films, and reported that the interaction between lignin and water was not only governed by the hydrogen bonding but also by specific interactions such as dipole-dipole interaction.

Wilhelmy's method was introduced by Miller and Young (1975) to measure contact angles of filaments with inherent anisotropy in nature. This dynamic contact angle analysis was adopted by Young (1976) who examined the contact angle of wood pulp fibers. Klunness (1981) used a much simpler and less expensive method to calculate the $\cos\theta$ of wood fibers without measuring their

perimeters. In a recent study, Liu (1994) correlated the contact angle of treated fibers and their interfacial shear strength with plastics. This method was also applied to solid wood by other investigators (Casilla *et al.* 1981, Casilla *et al.* 1984, Kalnins and Katzenberger 1987), and was claimed to give more reproducible results to overcome the absorption and surface chemical heterogeneity of wood (Gardner *et al.* 1991).

Like other polymeric materials, surface characteristics of wood are time, temperature, and environment dependent (Andrade 1988, Gunnells *et al.* 1994). Contact angle increases with aging (Herczeg 1965, Nguyen and Johns 1979), and decreases with weathering (Kalnins and Feist 1993). Contact angle also decreases with increasing temperature (Gunnells *et al.* 1994), but drying, which results in surface inactivation, decreases wood wettability (Troughton and Chow 1971). Removal of extractives may or may not affect wettability of wood, depending on species (Nguyen and Johns 1979).

3.2.3. Interfacial Shear Strength Between Wood and Plastics

The adhesion property is determined by characterizing wettability, which evaluates the affinity of two phases upon initial contact. However, bonding strength upon solidification of the adhesive is also important. In fiber-reinforced plastics, there is no satisfactory method of measuring the bond strength between fiber and matrix (Hull 1981). Tensile, flexural and impact strengths of the composites are assessments of overall performance of the composite, not interfacial adhesion.

Several methods have been developed to directly characterize the interface of reinforced composites, namely, the single-fiber fragmentation test, the single-fiber pull-out test, and the microbond test (Sanadi *et al.* 1992, Sanadi *et al.* 1993, Tao *et al.* 1993, Liu 1994, Liu *et al.* 1995). These three techniques have been widely used to approximate the interfacial shear strength of fiber-reinforced thermoplastic composites using a model composite.

In the single-fiber fragmentation test, a single fiber is embedded in the center of a dog-bone polymer matrix with the longitudinal axis parallel to the load direction. The fiber breaks into fragments on the extension of matrix, and the fragments approach to the fiber critical length. The fiber fragment lengths are determined with a microscope, and the interfacial strength (τ) are calculated as follows:

$$\tau = \frac{\sigma_{fu} d}{2l_c} \quad (\text{Eq. 3.2})$$

where σ_{fu} is the fiber tensile strength, d is the fiber diameter, and l_c is the fiber critical length. This method has recently been applied to natural fiber-plastic composites. (Tai *et al.* 1992, Shaler 1993, Felix and Gatenholm 1994). There are some limitations in applying this method: (1) ultimate strain and strength of the matrix should be much greater than those of fiber (Sanadi *et al.* 1992, Tao *et al.* 1993), (2) modulus of the matrix should be significantly lower than its counterpart, and (3) fiber diameter should be constant (Liu *et al.* 1995).

In the single-fiber pull-out test, fiber is embedded in a cylindrical matrix button with the fiber axis perpendicular to the surface of the button, and tensile load, parallel to the fiber axis, is applied to the specimen. The continuous tension causes a crack to develop in the fiber-matrix interface at the top of the embedded length. When the crack has propagated to the full length of the interface, the fiber debonds from the matrix (Penn and Lee 1989), the interfacial shear strength (τ) can then be determined as follows:

$$\tau = \frac{F}{\pi d l} \quad (\text{Eq. 3.3})$$

where F is the debonding force, d is the fiber diameter, and l is the fiber embedded length. Sanadi and coworkers (1992) examined the interfacial shear strengths between wood and two types of thermoplastics. Westerlind *et al.* (1984) studied the interfacial properties and the failure modes in the polymer-bonded dry-formed network of rayon fibers, using the single-fiber pull-out technique. The method, however, has several restrictions. That is, (1) the relationship between shear stress and embedded fiber length could be nonlinear (Sanadi *et al.* 1993, Penn and Lee 1989), (2) the meniscus effect requires extra effort to measure the embedded length (Miller *et al.* 1987, Liu 1994), and (3) the small embedded length makes sample preparation a very difficult step (Tao *et al.* 1993, Liu *et al.* 1995).

Microbond test involves placing microdroplet(s) of resin, which becomes ellipsoid after curing, onto the fiber surface (Miller *et al.* 1987). The interfacial

shear strength can be calculated using the same equation as the single-fiber pull-out test. Liu's group has successfully applied this method in natural fiber-thermoplastic composites (Gardner *et al.* 1994, Liu 1994, Liu *et al.* 1995). The microbond technique was developed to solve the meniscus problems associated with the single-fiber pull-out test (Miller *et al.* 1987, Liu 1994). It is, however, difficult to control the shape and size of the microdroplets (Sanadi *et al.* 1992), as well as to eliminate the stress concentration at the top of the droplet caused by nonuniform shear stress along the interface (Miller *et al.* 1987, Tao *et al.* 1993).

These three methods were designed to measure the bonding exactly on the interphase; however, using model composites may not represent the real world situation. The morphology of natural fibers and their susceptibility to deformation worsen the situation. Therefore, in selecting a method to evaluate interfacial shear strength between wood and plastic, wood dowels instead of wood fibers could be used in a modified single-fiber pull-out test to assess the "bulk" interfacial shear strength.

3.3. MATERIALS AND METHODS

3.3.1. Part I. Wettability of MAPP-Treated Birch Plywood and Polyolefin Polymers

3.3.1.1. Materials

A piece of nominal 1/4-inch thick 4- by 8-foot birch plywood was obtained from a local mill. The compatibilizer was a low molecular weight emulsion type maleated polypropylene (MAPP), Epolene E-43 from the Eastman Chemical Co.

Four wetting liquids, namely, water, urea formaldehyde (UF), phenol formaldehyde (PF), and isocyanate (ISO), were used to measure the contact angle. Both urea formaldehyde (Casco resin TD-33C) and phenol formaldehyde (Cascophen 1770-3) resins were supplied by the Borden Chemical Inc. The typical viscosity at 25°C is 175 cps for UF and 130 cps for PF, and the solid content are 65% and 53%, respectively. The isocyanate used in this study was ISOBOND, a product of Dow Plastics, which has 30.9% NCO content and 185cps viscosity (25°C).

The three types of polyolefin polymers used in this study were supplied by the Exxon Chemical Co. They were polypropylene (PP), linear low-density polyethylene (LLDPE), and low-density polyethylene (LDPE). Specifications for these plastics are tabulated in Table 3.1.

3.3.1.2. Methods

3.3.1.2.1. Contact Angle Measurement for Birch Plywood

The birch plywood was cut into 0.5(W) x 2.0(L) -inch specimens which were polished with #600 sand paper and vacuum cleaned to remove debris before subjecting to different drying conditions. Half of the specimens were air-dried in desiccators over anhydrous CaSO_4 for 2 days, and the other half were oven-dried at $103 \pm 2^\circ\text{C}$ for 24 hours. All specimens were brush-coated with E-43 emulsion of various concentrations onto the surface, and then conditioned in desiccators over anhydrous CaSO_4 for 48 hours.

The apparent contact angles of various wetting liquids on the surface of treated birch plywood specimens were measured by the direct optical method using a Kernco (model G-1) contact angle meter. All measurements were done within 5 seconds after deposition of droplets. To control the biological variation of the birch plywood, contact angles of the four wetting liquids were measured in every specimen.

The variables investigated included drying condition, concentration of E-43, and liquid type. Replications for concentration level 0, 10, 20, 30, and 40% were 25, and for concentration level 1.25, 2.5, and 5% were 10. A split plot design (Table 3.2) with 2x8x4 factorial treatment arrangement was employed to analyze the factors of interest.

Table 3.1. Basic physical properties for plastics.

Plastic Type	Melt index ¹	Density ²	Characteristic point ³
	---- g/10 min ----	----g/cm ³ ----	---- °C ----
PD7292N E7 (PP)	4.0	0.900	95
LL-3001 (LLDPE)	1.0	0.917	125
LD-200 (LDPE)	7.5	0.917	104

¹. Test methods for melt index are based on ASTM D1238 for PP and LLDPE, Exxon method for LDPE.

². Test methods for density are based on ASTM D792 for PP and Exxon method for LDPE and LLDPE.

³. Temperatures listed are deflection temperature at 66 psi for PP using ASTM D 638 method and melting points for LLDPE and LDPE using Exxon method.

Table 3.2. Experiment design for measuring contact angle of birch plywood.

Variable	Level	Description
Drying method (DRY)	2	Oven-dry (OD) and air-dry (AD)
Coupling agent concentration (CON)	8	0 (control), 1.25, 2.5, 5, 10, 20, 30, 40 %.
Liquid type (LIQUID)	4	Distilled water (WATER), urea-formaldehyde (UF), phenol-formaldehyde (PF), and isocyanate (ISO).
Replication	10, 25	

Table 3.3. Experiment design for testing bulk interfacial shear strength of birch dowel and polyolefin polymers

Variable	Level	Description
Plastic type (PT)	3	low density polyethylene (LDPE) linear low density polyethylene (LLDPE) polypropylene (PP)
Adhesive type (AT)	4	control (NO) urea formaldehyde (UF) phenol formaldehyde (PF) isocyanate (ISO)
E-43 concentration (CON)	5	0 (control), 10, 20, 30, 40 %.
Replication	6	

Birch plywood specimens were also coated with UF, PF, ISO and NO (no adhesive coated) to investigate the interfacial relationship in wood-adhesive-compatibilizer. The solid contents coated were approximately 10 and 4 lbs per 1000 ft² for UF/PF and ISO, respectively. Coated specimens were dried for two

days, before contact angles of E-43 emulsion were measured. The experiment was a completely randomized design with 10 replicates in each treatment.

3.3.1.2.2. Contact Angle Measurement for Plastics

Plastic pellets were placed in TEFLON® containers and melted in an oven for two hours. The temperature settings for LDPE, LLDPE, and PP were 140°, 160°, and 200°C, respectively. After the molten plastics were set and conditioned in room temperature for 24 hours, contact angles of the four wetting liquids were measured using the same methodology described in the previous section. A split plot design with 3x4 factorial treatment arrangement was also used to investigate the effect of wetting liquid on the plastic type. The effect of compatibilizer (E-43) concentrations on the plastic type were also evaluated in an analogous manner with 3x4 factorial levels. Each treatment combination contained 24 replicates. Contact angles of four wetting liquids, UF, PF, ISO, and water, on polypropylene surfaces treated with 40% E-43 emulsion were also investigated in a separate study of completely randomized design. Each treatment had 10 replicates.

3.3.2. Part II. Bulk Interfacial Shear Strength Between Birch Wood and Polyolefin Polymers

3.3.2.1. Materials

Nominal 3/8-inch birch wood dowels were obtained from a local mill. The polyolefin polymers, compatibilizer, and adhesive resins used in this experiment were the same as described in previous section.

3.3.2.2. Methods

Birch dowels were cut into 2 $\frac{3}{8}$ -inch long samples, and then oven dried at $103\pm 3^{\circ}\text{C}$ until constant weight. These oven-dried dowels were then transferred to desiccators over anhydrous CaSO_4 to cool down to room temperature. About 30 g of plastic pellets were melted in TEFLON® containers at various temperature for two hours. The temperature settings for LDPE, LLDPE, and PP were 140° , 160° , and 200°C , respectively.

Wood dowels were embedded in the molten plastic matrices using specially designed gigs, and remained in the oven for 30 minutes. Two hours before embedding, the adhesive was applied onto the surface of birch dowels at the interfacial area, and then set to dry in a desiccator. In a similar manner, the compatibilizer was applied one hour before the embedding procedure.

To minimize shrinkage of the plastics, especially PP, test specimens were cooled gradually by turning off the oven and letting them remained in the oven until room temperature was reached. Interfacial shear strength tests were carried out in an Instron Universal Testing Machine at a load cell speed of 0.2 in/min, using the tension fixture on the dowel end and the internal bond fixture on the plastic matrix end. The schematic diagram for the testing procedure is shown in Figure 3.1. Bulk interfacial shear strength (ISS) was determined using Equation 3.3. This experiment was a split plot design with $3\times 4\times 5$ factorial treatment arrangement, as shown in Table 3.3.

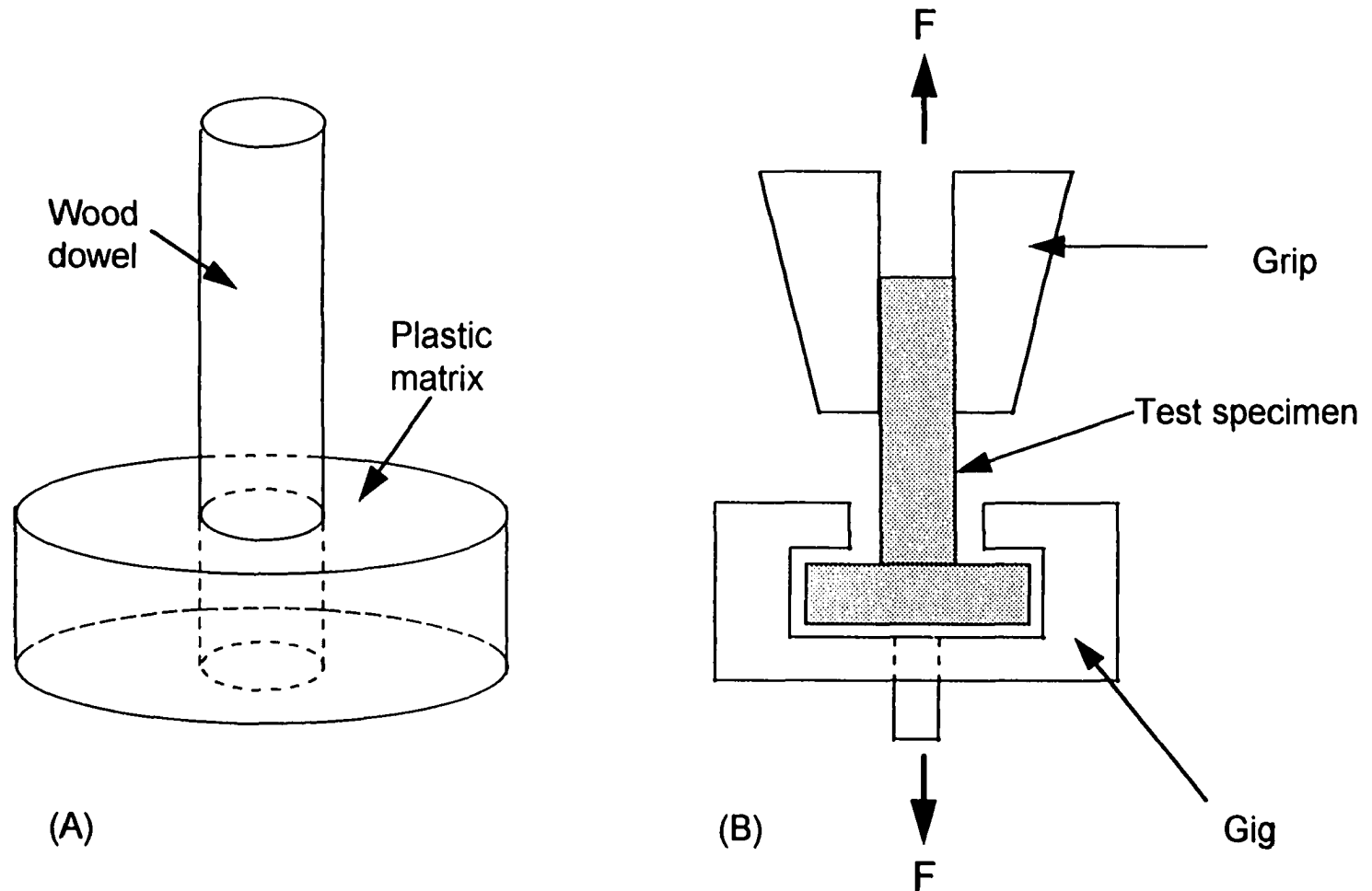


Figure 3.1. Schematic diagram of the interfacial shear strength test procedure (A) test specimen, (B) specimen and fixture layout.

Table 3.4. Contact angle for birch plywood treated with various E-43 concentrations (CON).

Drying method/ CON(%)	Liquid type ¹			
	ISO	PF	UF	water
<i>Air-dry</i>				
0	28.3 (3.1) ²	75.2 (6.4)	78.8 (6.6)	51.7 (9.3)
1.25	43.5 (2.0)	78.3 (1.4)	90.6 (2.8)	39.6 (4.0)
2.5	47.5 (3.7)	79.2 (4.4)	91.0 (3.5)	29.0 (11.2)
5	48.0 (3.9)	82.5 (3.2)	90.6 (3.0)	6.0 (4.1)
10	45.3 (4.8)	85.9 (7.3)	91.2 (8.0)	0.0 (0.0)
20	39.5 (3.4)	54.4 (9.1)	36.2 (8.4)	0.0 (0.0)
30	32.7 (4.9)	38.5 (7.5)	24.8 (4.4)	0.7 (2.2)
40	27.6 (3.1)	33.2 (2.9)	22.0 (2.7)	1.1 (2.7)
<i>Oven-dry</i>				
0	27.3 (4.0)	84.3 (6.1)	89.2 (6.3)	67.7 (8.7)
1.25	40.9 (3.9)	85.2 (1.0)	91.4 (0.9)	46.8 (6.5)
2.5	48.8 (3.2)	86.1 (1.4)	91.7 (1.4)	35.2 (7.4)
5	47.0 (1.9)	86.6 (2.5)	92.0 (1.6)	6.0 (4.0)
10	44.0 (3.5)	88.5 (8.8)	92.3 (11.8)	0.0 (0.0)
20	42.6 (3.3)	66.3 (7.6)	46.7 (10.0)	0.0 (0.0)
30	38.4 (8.6)	46.7 (12.1)	32.3 (11.1)	0.9 (3.1)
40	30.1 (3.4)	32.8 (3.7)	22.5 (2.5)	2.6 (5.1)

¹. ISO, PF, UF and WATER denote polyisocyanate, phenol formaldehyde, urea formaldehyde, and water, respectively.

². Numbers in parentheses are standard deviations.

3.4. RESULTS AND DISCUSSION

3.4.1. Part I. Wettability of MAPP-Treated Birch Plywood and Polyolefin Polymers

3.4.1.1. Contact Angle in MAPP-Treated Birch Plywood

The average contact angles of the four wetting liquids against birch plywood treated with various concentrations of E-43 compatibilizer are shown in Table 3.4. In both air-dry and oven-dry treatments, contact angles of isocyanate (ISO), phenolic resin (PF), and urea resin (UF) increased at lower E-43 concentration levels, then dropped progressively as E-43 concentration increased. On the contrary, contact angle of water dropped continuously as E-43 concentration increased. Water was most sensitive to E-43 treatment; and, in concentration level above 5%, there was complete wetting. The control groups (0% E-43) in Table 3.4 also indicate that contact angles of water was greatly affected by drying method, whereas ISO was least affected.

ANOVA was performed using SAS General Linear Model on square root transformed data, since data sets were unbalanced and did not pass Hartley F-max tests. However, results of ANOVA based on the original data (Table 3.5) coincided with the transformed data; therefore, for simplicity, the original data was used for further discussion. All of the interaction terms were highly significant ($p < 0.01$). However, because the F values in drying method (DRY), E-43 concentration (CON) and wetting liquid type (LIQUID) were very large, as compared with the three-way interaction (DRY*CON*LIQUID), the main effects still provided useful information.

The effect of drying method was highly significant ($p=0.0001$), as shown in Figure 3.2. Oven-dried birch plywood specimens had higher contact angle than air-dried ones when all other factors were combined, implying that samples which were subjected to different drying methods before application of E-43 affected their wettabilities. Surface inactivation caused by drying maybe responsible for the loss of wettability in oven-dried wood (Christiansen 1990).

The effect of E-43 concentration level on contact angles is illustrated in Figure 3.3. When all wetting liquids and drying methods were combined, initial increase in E-43 concentration resulted in an increase in contact angle, however, as concentration further increased the contact angle decreased appreciably. This is reasonable since MAPP can form hydrogen bonding with the hydroxyl groups on the surface of cellulosic fiber, and also with the hydroxyl group-rich wetting liquids. ISO, however, cannot form hydrogen bonds. Its NCO functional groups are reactive with the hydroxyl groups of wood and the carboxylic groups of MAPP. Therefore, at lower concentrations of E-43, the carboxylic groups of MAPP tend to orient toward wood fiber, leaving long-chain PP back bones facing outward. Low surface free energy of PP induces higher contact angle. However, when E-43 concentration is too high the orientation is interrupted, the carboxylic groups and back bones of MAPP become randomly oriented. This results in greater possibility for PF, UF and ISO to interact with carboxylic groups of E-43, yielding lower contact angles.

The four wetting liquids (Figure 3.4) had dramatically different wettabilities on E-43 treated surface. In decreasing order of contact angle, were PF, UF, ISO, and water. Contact angle between a liquid and a solid substrate is affected by many factors, such as surface tension and viscosity of the liquid, surface molecular packing, chemical constitution and critical surface tension of the solid, and the interaction between liquid and solid. In comparing wettability of various wetting liquids, the solid substrates which were subjected to the same treatment should exhibit the same surface properties. Thus, any difference in wettability is due to liquid type and the interaction between liquid and solid. The viscosity and surface tension of these four wetting liquids are different. Since they have different chemical constitutions, their interactions with the solid substrate are also different. These differences resulted in the different average values of contact angle for the four wetting liquids. The polarity of water and reactivity of ISO may be responsible for their higher wettabilities on E-43-treated wood surface.

The dependence of contact angle on E-43 concentration for various wetting liquids is shown in Figure 3.5 for air-dried plywood, and in Figure 3.6 for oven-dried plywood. Both figures show that contact angles of UF and PF increased slightly at lower levels of E-43 concentration, and decreased drastically with changes in E-43 concentration from 10% to 20%, then decreased slightly with further increase in E-43 concentration. The reduction in contact angle from low concentration to high concentration was most pronounced in UF. This phenomenon implied that the effect of MAPP treatment in improving wettability

is more effective in UF for both air-dry and oven-dry conditions. The decrease in contact angle from 10 to 40% E-43 concentration was 75% for UF and 62% for PF.

Table 3.5. ANOVA results on effects of drying method (DRY), coupling agent concentration (CON), and liquid type (LIQUID) on contact angle measurement of birch plywood.

Source	DF	MS	F	P value
DRY	1	3724.2911	54.5	0.0001
CON	7	50093.5894	733.03	0.0001
DRY*CON	7	378.2107	5.53	0.0001
REP(DRY*CON)	294	68.3378	2.74	0.0001
LIQUID	3	153946.1049	6165.48	0.0001
DRY*LIQUID	3	305.5519	12.24	0.0001
CON*LIQUID	21	11328.1299	453.69	0.0001
DRY*CON*LIQUID	21	158.6145	6.35	0.0001
ERROR	882	24.9690	---	---

The initial contact angles of ISO were lower than UF, PF, and water for both air-dry and oven-dry methods, but they increased profoundly at lower levels of E-43; as E-43 concentration further increased, contact angles decreased gradually. Among the three adhesives, ISO was most sensitive to the addition of E-43, the increase in contact angle from 0 to 1.25% E-43 concentration was 54 and 50% for air-dry and oven-dry, respectively. On the other hand, the reduction of contact angle at higher concentration levels was less dramatic in

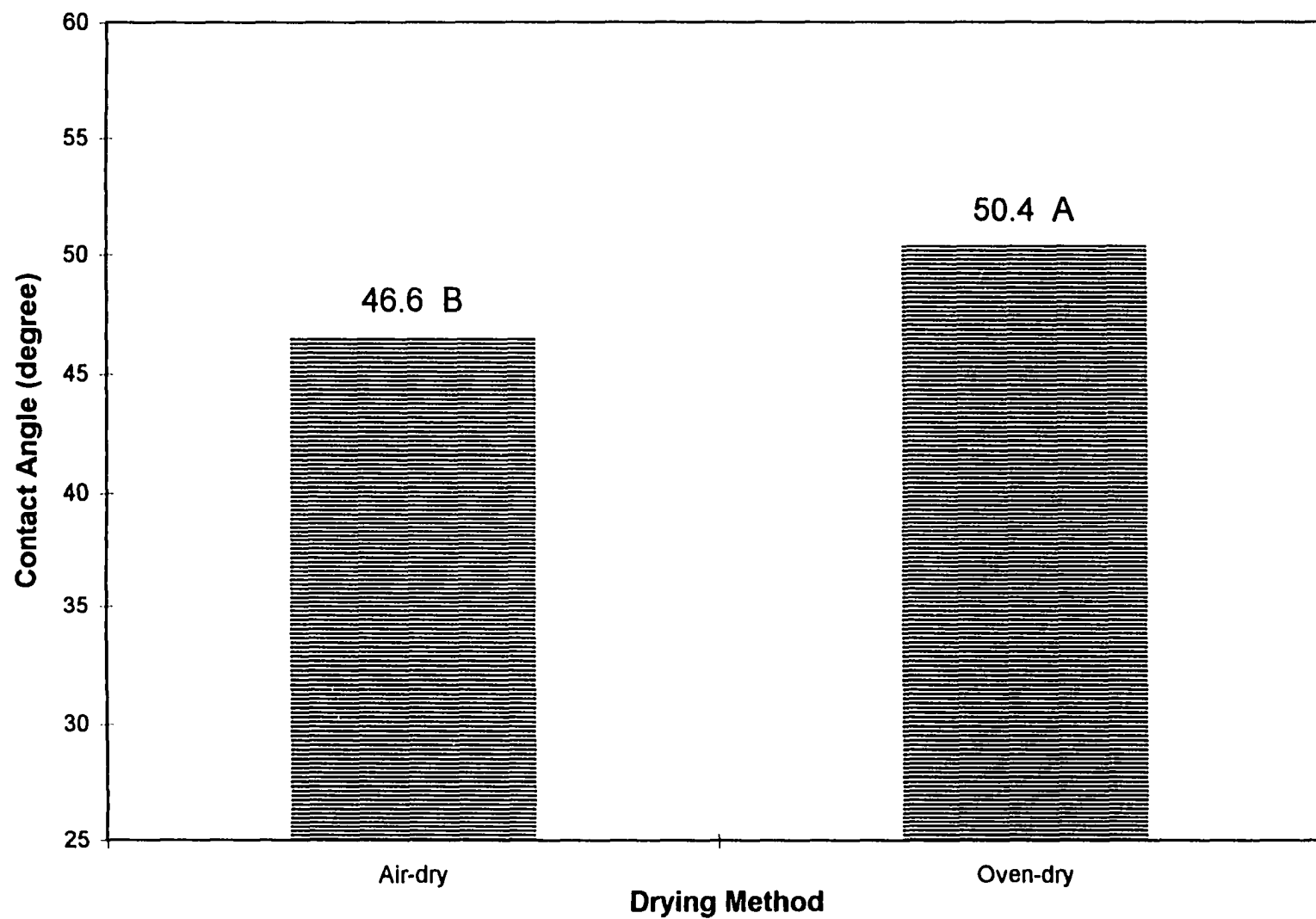


Figure 3.2. Contact angle of E-43 treated birch plywood by drying method.

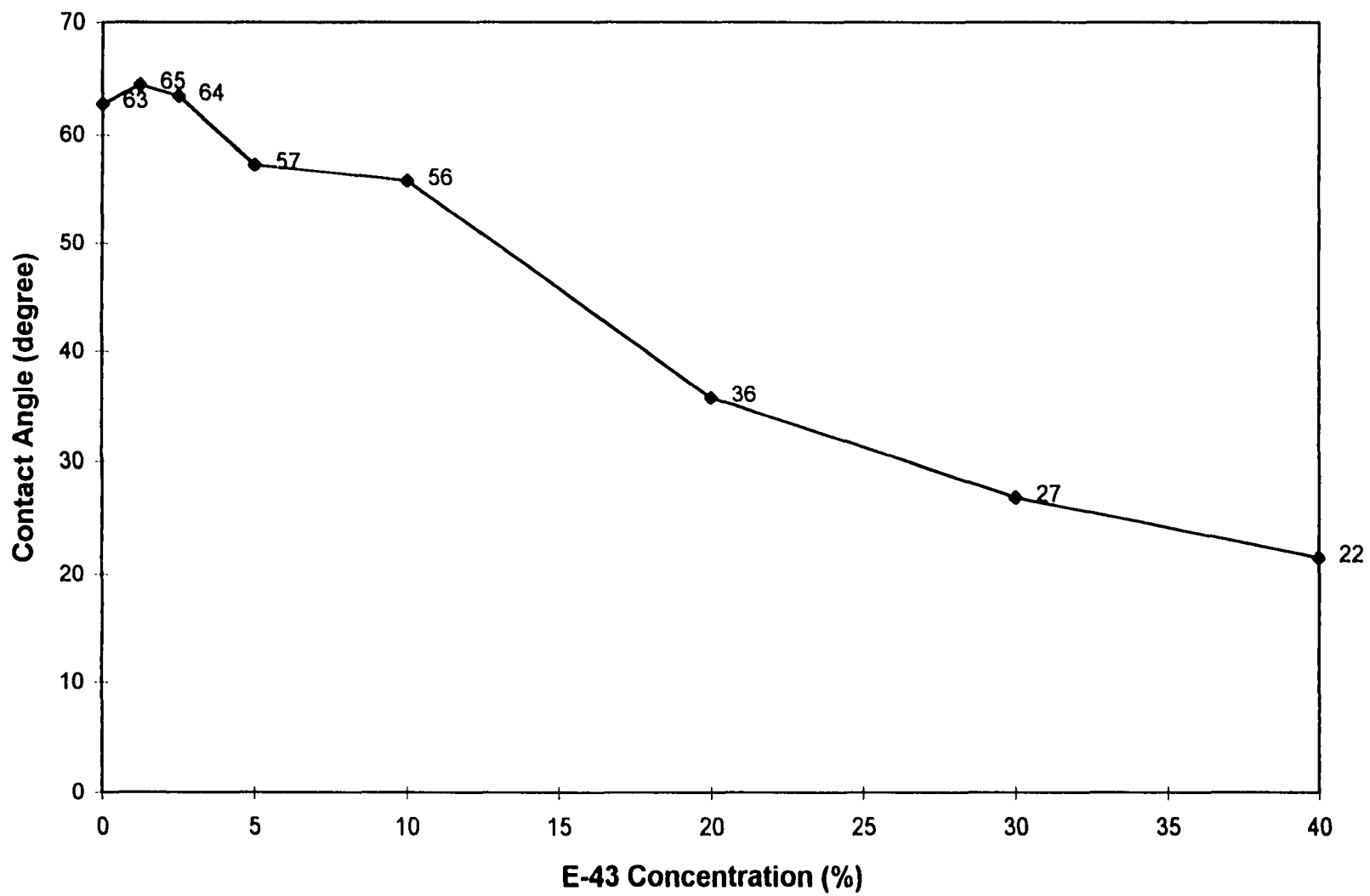


Figure 3.3. Contact angle of E-43 treated birch plywood by E-43 concentration level.

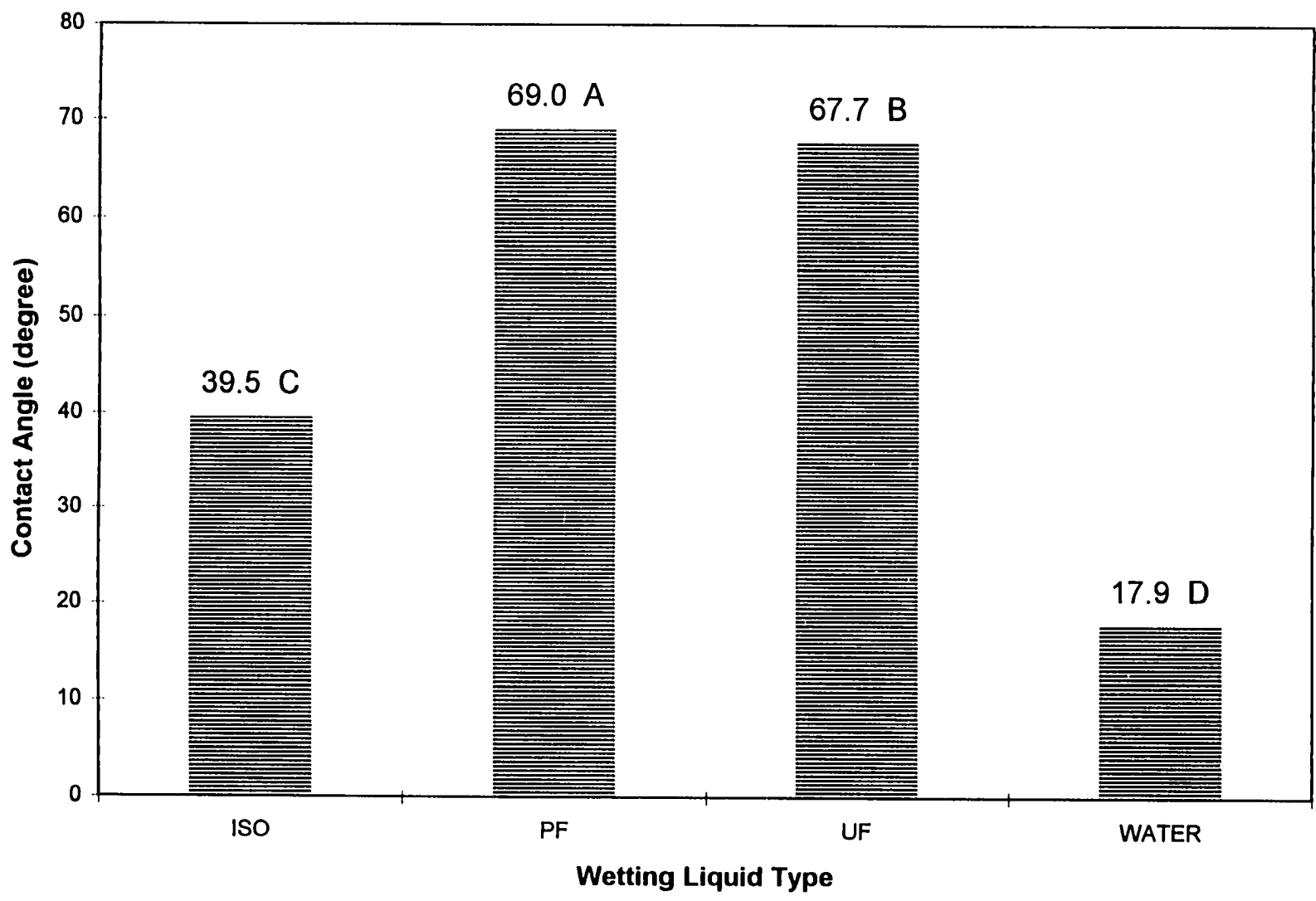


Figure 3.4. Contact angle of four wetting liquids on E-43 treated birch plywood .

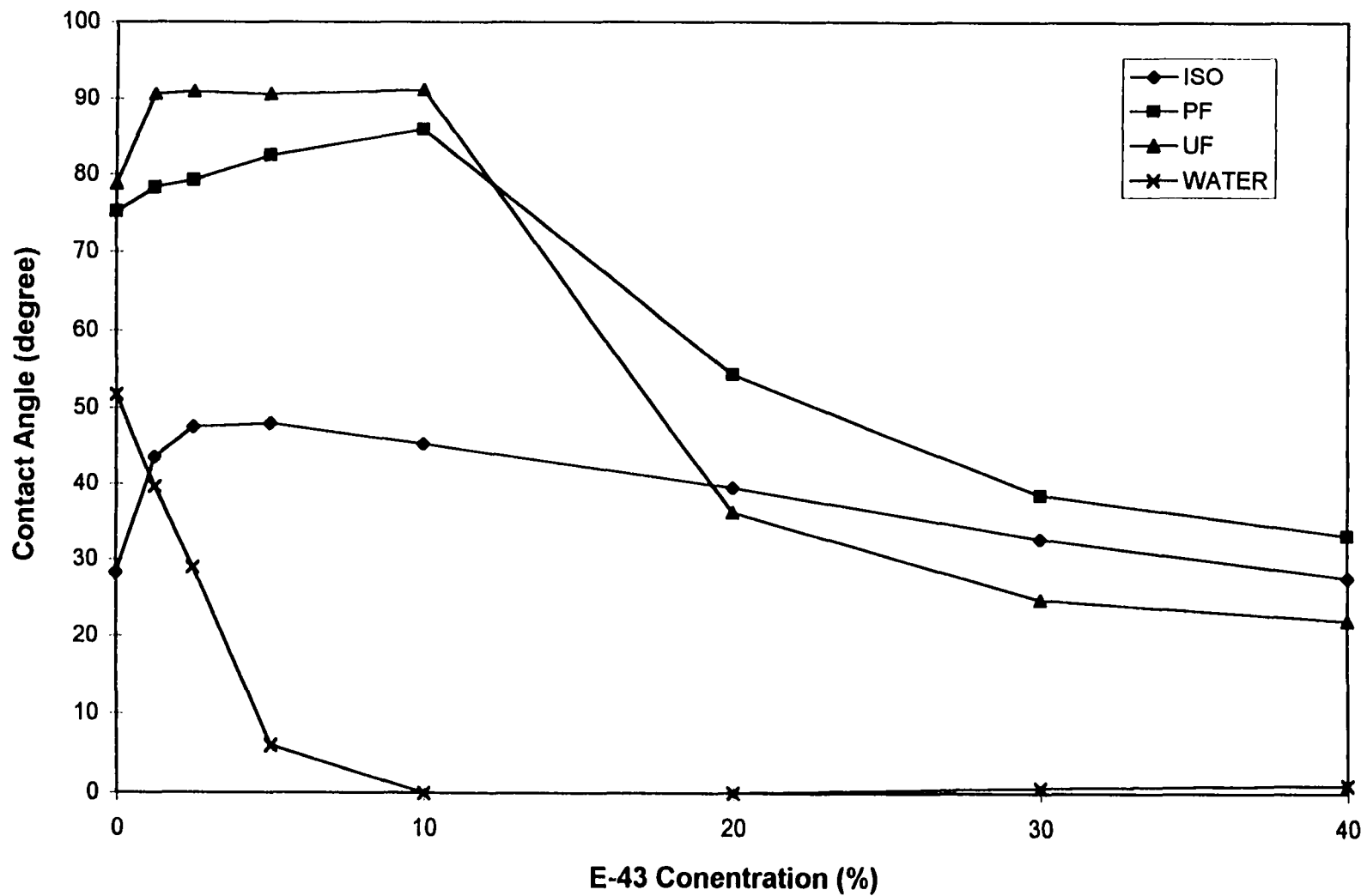


Figure 3.5. Contact angle of four wetting liquids on air-dried birch plywood surface treated with various E-43 concentrations.

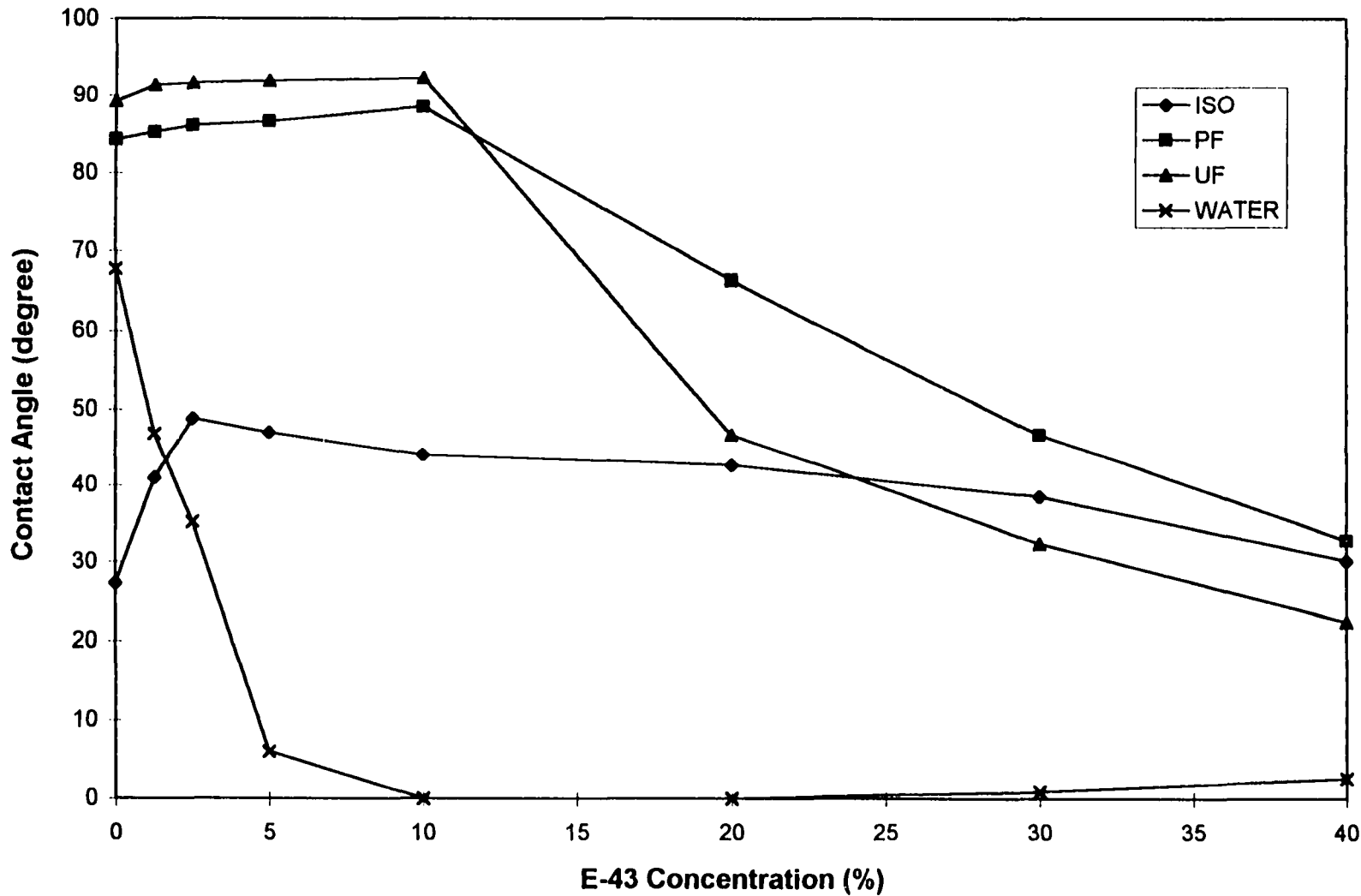


Figure 3.6. Contact angle of four wetting liquids on oven-dried birch plywood surface treated with various E-43 concentrations.

ISO as compared to UF and PF, since the total reduction in contact angle of ISO from 10 % to 40% E-43 concentration was approximately 39% and 31% for air-dry and oven-dry, respectively.

Water reacted totally different from the other wetting liquids, and was the major reason for the three-way interaction. Contact angle of water decreased drastically upon the initial addition of E-43, and then reached zero, i.e., complete wetting, and remained zero as E-43 concentration exceeded 10%. This phenomenon indicates that water can change the equilibrium of MAPP from anhydride form to carboxylic form. As explained previously, UF, PF and ISO showed upward trends between 0 to 5% E-43 due to the outward orientation of MAPP backbone which is less wettable. However, water, being a polar and non-polymeric material, is able to react with MAPP to form carboxylic groups, regardless the orientation of MAPP. Hence its contact angle decreased progressively until the point (10%) when wood surface was totally covered by E-43 particles. Therefore contact angle of water was more sensitive to the application of low concentration E-43 than the other wetting liquids.

The relationship of contact angle with actual E-43 solid content on birch plywood surface was also investigated. Results of polynomial regression analyses, as shown in Table 3.6, indicate that 69-80%, and 76-85% of the variations in contact angles of PF and UF could be explained by E-43 solid content.

Table 3.6. Statistics and coefficients for polynomial regression analysis of E-43 solid content (X) and contact angle (Y) with different liquid types and drying conditions as denoted by mode $Y=b_0+b_1X+b_2X^2$.

Drying method/ Liquid type ¹	Statistics			Partial coefficient		
	F	P	R ²	b ₀	b ₁	b ₂
Air-dry						
PF	169.88	0.0001	0.6909	83.250	-7619	232539
UF	239.33	0.0001	0.7590	92.521	-13446	572558
Oven-dry						
PF ²	635.57	0.0001	0.8060	89.663	-3677	---
UF	447.10	0.0001	0.8547	96.054	-6899	123830

¹. PF and UF denote phenol and urea formaldehyde, respectively.

². Linear model fit better than quadratic model.

Figure 3.7 shows the average contact angles of E-43 on birch plywood coated with the four types of adhesive, that is, ISO, PF, UF, and NO (no adhesive, control). Results of Tukey's test also revealed the ranking of E-43 contact angle as NO>PF>ISO>UF. For 40% E-43, UF-coated surface showed the highest wettability, while the NO-coated (clean plywood surface) showed the lowest wettability. For the three adhesive types, this result coincided with the ranking of contact angles of adhesive on 40% E-43 treated surface (Table 3.4), implying that the interface wettabilities of wood-compatibilizer-adhesive and wood-adhesive-compatibilizer were of the same rank. The magnitude of contact angle, however, was lower in the former and higher in the later.

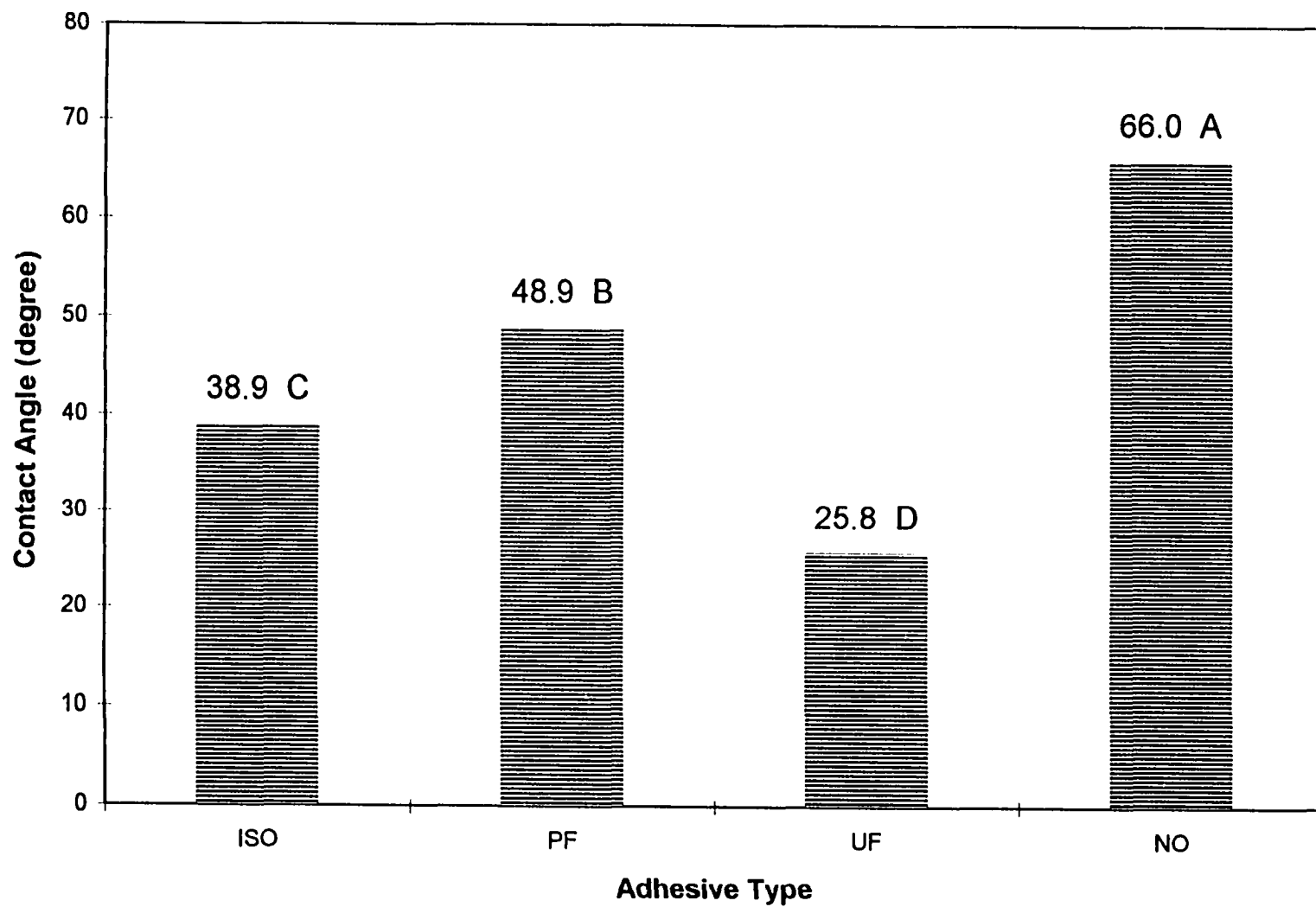


Figure 3.7. Contact angle of E-43 on birch plywood coated with four adhesives.

3.4.1.2. Contact Angle Measurement for Polyolefin Polymers

The wettabilities of polyolefin polymers were measured in two ways: (1) with E-43 compatibilizer and (2) with four wetting liquids. These were split plot designs with plastic type (PT) placed in the main plots, E-43 concentration (CON) or wetting liquid (LIQUID) in the subplots. The average contact angles for various E-43 concentrations (Tables 3.7) ranged from 45 to 58° for LDPE, 52 to 63° for LLDPE, and 66 to 68° for PP. The average contact angles of four wetting liquids (Tables 3.8) ranged from 51 to 92° for LDPE, 56 to 95° for LLDPE, and 65 to 101° for PP.

The ANOVA in Table 3.9 reveals that both plastic type (PT) (and E-43 concentration (CON) main effects as well as their interaction (PT*CON) were highly significant ($p=0.0001$). The F value for plastic type was 18 times higher than those of PT*CON interaction, therefore the relationship of plastic type with contact angle is also presented (Figure 3.8). It shows that among the three polyolefin polymers, PP had the highest mean value, whereas LDPE the lowest. Since the surface of close-packed $-CH_3$ groups are less wettable than $-CH_2$ groups (Zisman 1964, Zisman 1976), the wettability of PP, LDPE, and LLDPE are expectedly to be different, due to differences in chemical composition, conformation, and degree of crystallinity. These differences in contact angle may also be the result of critical surface tension, which is lower in PP than in polyethylene (Wu, 1982).

Table 3.7. Contact angle of E-43 with various concentrations (CON) on polyolefin surfaces.

CON	Plastic type		
	LDPE	LLDPE	PP
--%--	Degree		
10	45.4 (5.5) ¹	52.7 (6.4)	66.5 (3.4)
20	49.1 (6.4)	56.8 (3.9)	65.9 (3.3)
30	49.8 (5.4)	56.8 (5.2)	66.7 (3.8)
40	58.9 (1.8)	62.5 (3.4)	67.6 (2.7)

¹. Numbers in parentheses are standard deviations. Sample size=24.

Table 3.8. Contact angle of five wetting liquids on polyolefin surfaces.

LIQUID ¹	Plastic type		
	LDPE	LLDPE	PP
	Degree		
ISO	49.3 (4.3) ²	56.1 (5.3)	65.1 (4.1)
PF	76.7 (4.4)	80.6 (3.1)	90.2 (4.2)
UF	83.9 (3.4)	87.1 (4.2)	97.1 (4.4)
WATER	91.9 (1.5)	94.4 (1.6)	100.9 (2.5)

¹. ISO, PF, UF and WATER denote polyisocyanate, phenol formaldehyde, urea formaldehyde, and water, respectively.

². Numbers in parentheses are standard deviations. Sample size=24.

The relationships of contact angle with plastic type and E-43 concentration (Figure 3.9) indicate that wettability of PP was constant regardless of E-43 concentration level. For LDPE and LLDPE, the responses were nearly parallel,

i.e., the contact angles increased slightly with increases in E-43 concentration. For a nonpolar low-energy surface like polyolefin polymer, its interactions with liquids are mostly by dispersion force. MAPP is configurationally more similar to PP than to LDPE and LLDPE, thus E-43 concentration may affect their wettabilities differently.

Table 3.9. ANOVA results on effects of plastic type (PT) and E-43 concentration (CON) on contact angle of plastics.

Source	DF	MS	F	P value
PT	2	6127.5757	188.65	0.0001
REP(PT)	69	32.4819	2.04	0.0001
CON	3	847.2699	53.13	0.0001
PT*CON	6	170.4166	10.69	0.0001
ERROR	207	15.9482	---	---

Table 3.10. ANOVA results on effects of plastic type (PT) and wetting liquid (LIQUID) on contact angle of plastics.

Source	DF	MS	F	P value
PT	2	4155.3301	185.93	0.0001
REP(PT)	69	22.3495	1.96	0.0001
LIQUID	3	21006.5258	1844.96	0.0001
PT*LIQUID	6	57.3246	5.03	0.0001
ERROR	207	11.3859	---	---

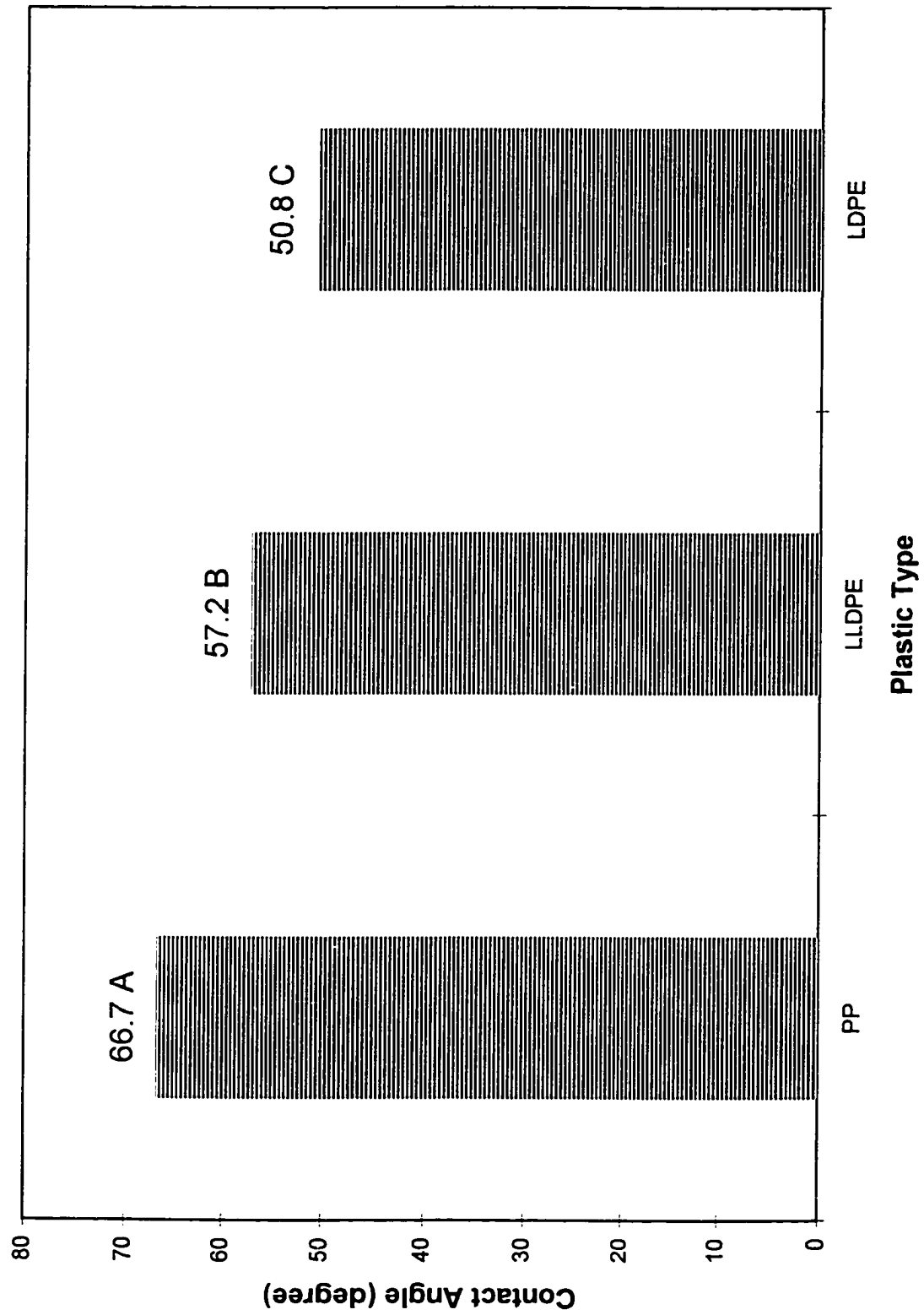


Figure 3.8. Contact angle of E-43 on three polyolefin surfaces.

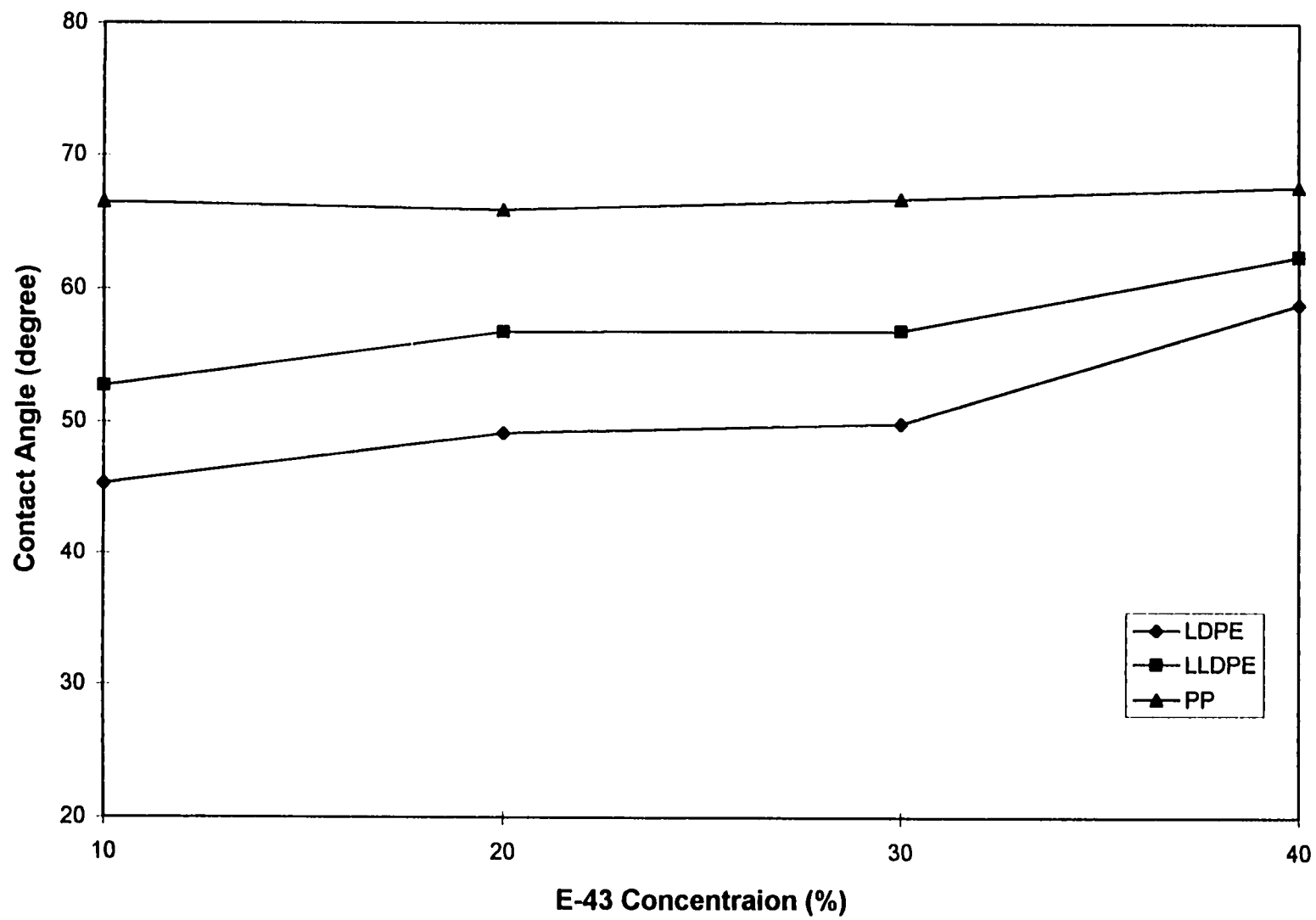


Figure 3.9. Dependence of contact angle of E-43 on E-43 concentration for three polyolefins.

The ANOVA in Table 3.10 reveals that plastic type and wetting liquid main effects (PT and LIQUID) and two-way interaction (PT*LIQUID) were highly significant ($p < 0.01$). The relationship of contact angles with plastic types by averaging the four wetting liquids (Figure 3.10), showed the same trend of contact angle as $PP > LLDPE > LDPE$, indicating that PP may have lower surface free energy than LDPE and LLDPE and hence is less wettable to most liquids. The effect of wetting liquid type (Figure 3.11) shows that ISO was most capable of wetting polyolefin polymers, and water was least. PF resin showed more affinity to polyolefin polymers than did UF resin. Compared with previous results (Figure 3.4 and Table 3.4), water generally had the strongest affinity to MAPP-treated wood ($\theta = 0 \sim 40^\circ$), and the least affinity to polyolefin polymers ($\theta = 92 \sim 101^\circ$). The affinity of water to untreated wood was intermediate ($\theta = 51 \sim 66^\circ$).

Among the three adhesives, ISO exhibited relatively strong wettability to both wood ($\theta = 27 \sim 43^\circ$) and plastics ($\theta = 49 \sim 65^\circ$). ISO is a reactive wood adhesive because it comprises mostly of low molecular weight dimers, diphenylmethane diisocyanate (MDI), which react actively with the hydroxyl groups present in water and cell wall components. On the other hand, the dipole-dipole interaction is weaker in ISO, and its nonaqueous carrier has higher affinity to nonpolar polyolefin polymers. Furthermore, ISO is said to possess an outstanding adherence capability to virtually everything (Johns 1982). This may explain the strong affinity of ISO to both wood and plastics. Compared with PF,

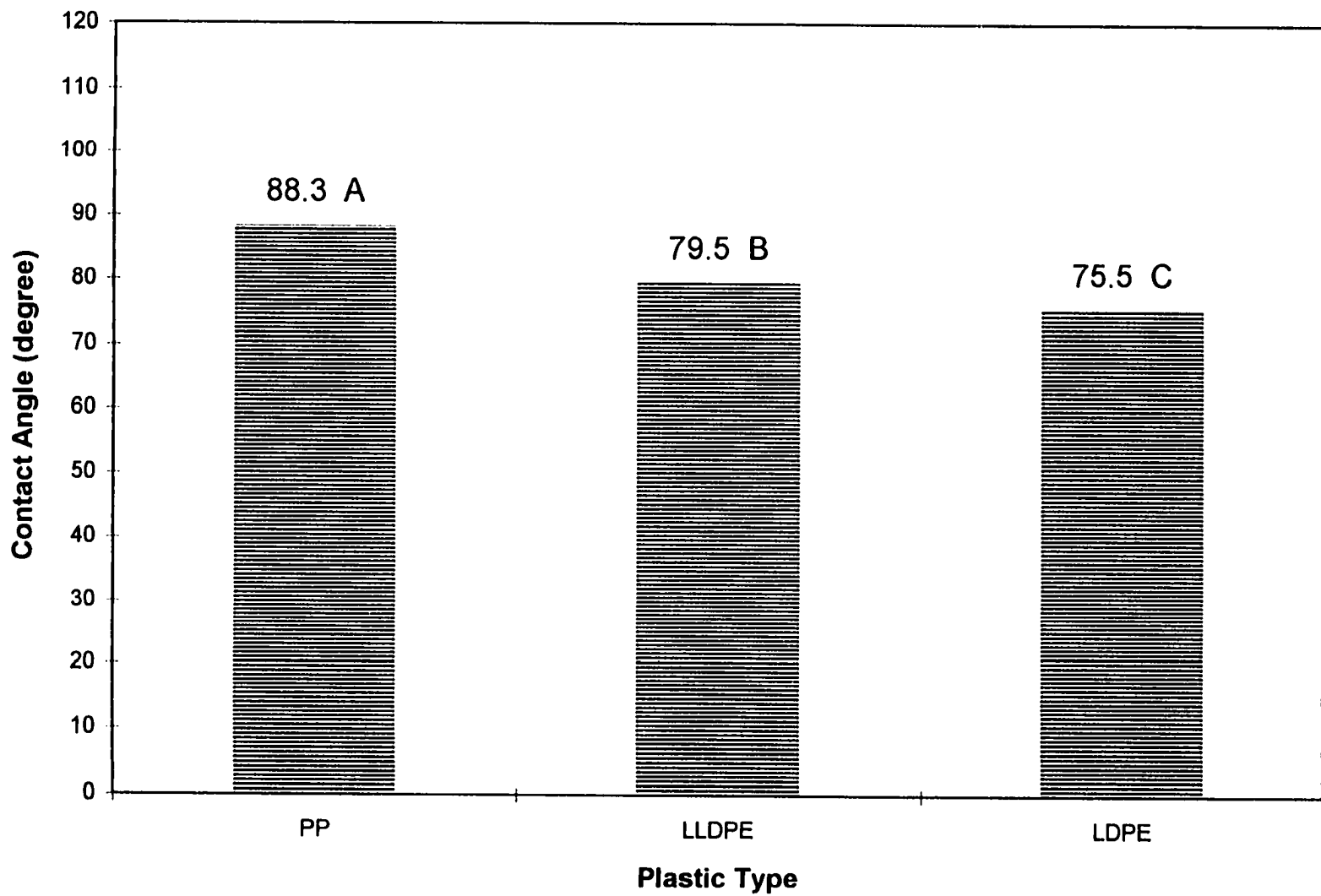


Figure 3.10. Average contact angle of four wetting liquids on three polyolefin surfaces.

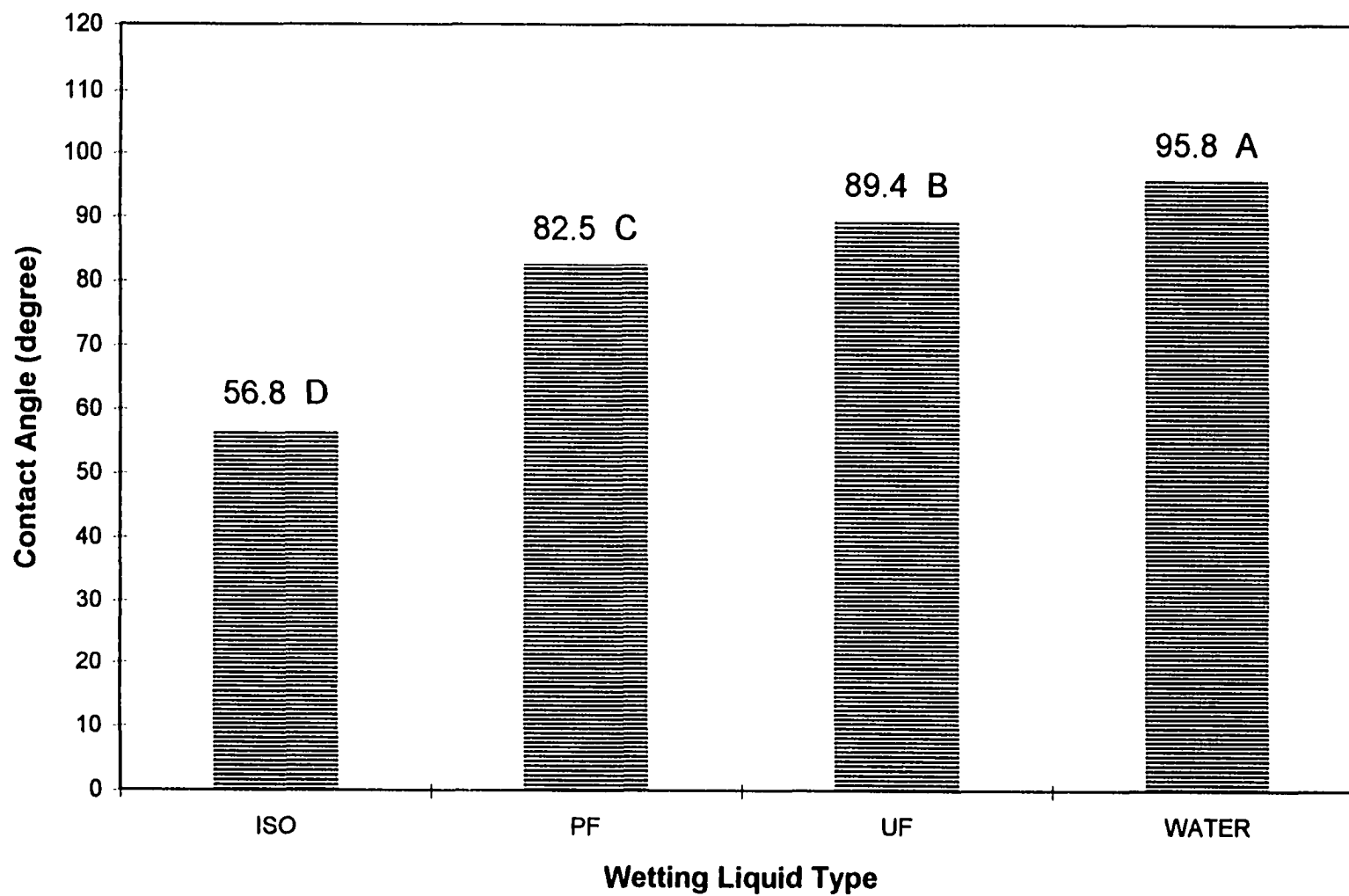


Figure 3.11. Average contact angle of three polyolefin polymers measured by four wetting liquids.

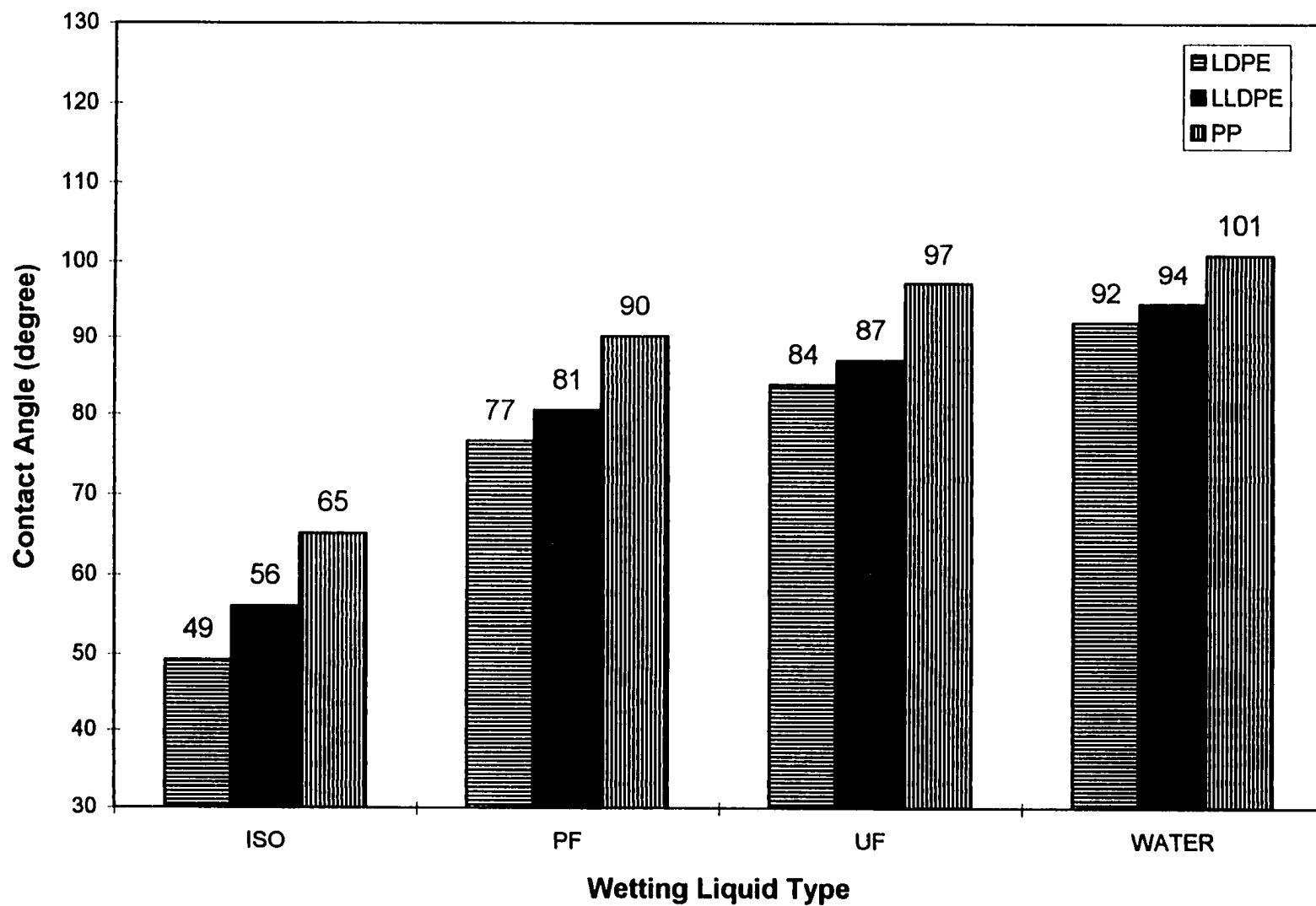


Figure 3.12. Contact angle of four wetting liquids on three polyolefin surfaces.

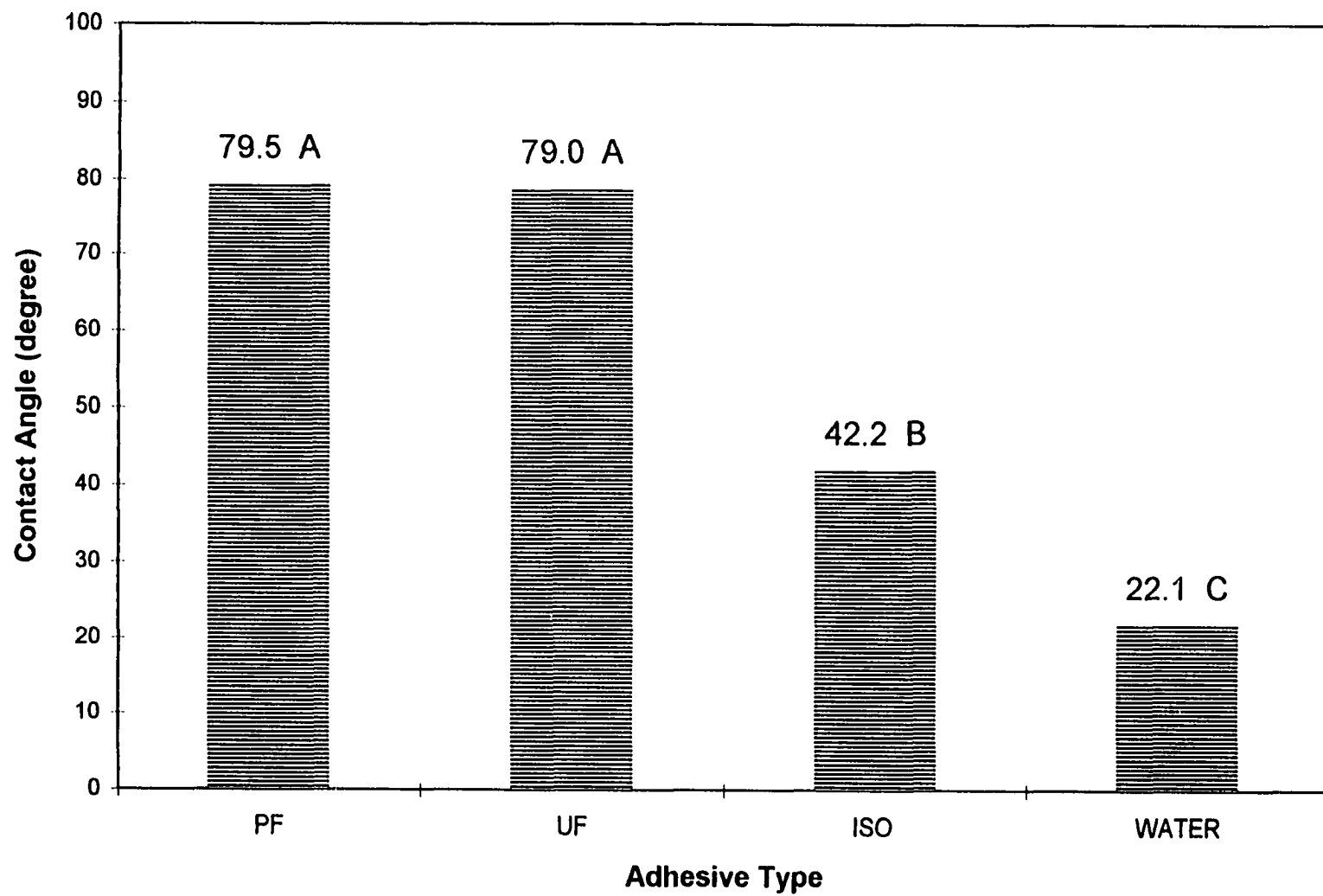


Figure 3.13. Contact angle of four adhesives on E-43 treated polypropylene surface.

UF had higher affinity to higher concentration E-43 treated wood ($\theta=22\sim46^\circ$ vs $\theta=32\sim66^\circ$), and lower affinity to untreated wood ($\theta=79\sim89^\circ$ vs $\theta=75\sim84^\circ$), lower concentration E-43 treated wood ($\theta=90\sim92^\circ$ vs $\theta=78\sim88^\circ$), and polyolefin surfaces ($\theta=84\sim97^\circ$ vs $\theta=77\sim90^\circ$). This may be due to less hydroxyl groups in UF than in PF. However, the difference in wettability between PF and UF was less dramatic. The interactions between liquid type and plastic type are illustrated in Figure 3.12. The responses of wetting liquid (LIQUID) to different plastic types (PT) were consistent in the order of PP>LLDPE>LDPE and water>Uf>PF>ISO. The LIQUID*PT interaction is presumably due to the inconsistency in the magnitude of difference.

The contact angles of the four wetting liquids on E-43 coated polypropylene surface are presented in Figure 3.13. Results of Tukey's test indicate that PF had the highest average contact angle, followed by UF, ISO, and water. This trend was similar to that of E-43 treated birch plywood (Figure 3.4), but was notably different from that of untreated polyolefin polymers (Figure 3.11). This demonstrates that E-43 can dramatically change substrate surface properties, conforming to the theory that a contact angle is affected only by the outermost constitution of the surface (Zisman 1964, 1976).

The wettabilities of wood and plastics by different wetting liquids suggest that if wettability is the major consideration of adhesive selection, ISO may be better than PF and UF, assuming that other factors such as cost, processibility, bond strength etc. are not considered. No MAPP is needed, with ISO serving

as wood and plastic binder. If UF and PF are used, addition of MAPP may enhance the surface properties. However, in the design and manufacture of composites, bond strength is always the prime concern. The judgement should thus take into account the interfacial bond adhesion, as presented in the next section.

3.4.2. Part II. Bulk Interfacial Shear Strength Between Birch Wood and Polyolefin Polymers

3.4.2.1. Interfacial Shear Strength

The mean interfacial shear strengths (ISS) for all treatment combinations (Table 3.11) show that, interfacial shear strengths between birch dowels and polyolefin polymers increased drastically with the addition of E-43, except for ISO-bonded LDPE and LLDPE which tended to decrease. In the control groups (0% E-43 concentration) within each of the three plastic types, ISO had the highest interfacial shear strength value among all other adhesives in both LDPE (410 vs 112~186 psi) and LLDPE (400 vs 140~253 psi). Moreover, in LDPE, ISO-0 (ISO-bonded with 0% E-43) even had the highest interfacial shear strength (410 psi) among all other treatment combinations (112~342 psi). The addition of E-43 inhibited the interfacial shear strengths of ISO-bonded LDPE and LLDPE composites, and enhanced the interfacial shear strengths for composites bonded with the other three adhesives. However, the most favorable treatment combinations for LLDPE were E-43 treated composites without applying any adhesive, i.e., NO-20 (562 psi) and NO-30 (531 psi). In PP, the no adhesive added composite (NO) showed the highest mean value of interfacial shear

strength among all control groups (416 vs 256~316 psi). With the addition of E-43 to PP, all the four adhesives had a two to three fold increment in interfacial shear strengths.

In a modified single fiber pull-out test, Sanadi *et al.* (1993) reported that the interfacial shear strength between low-molecular-weight polyethylene and 2 mm birch dowel was 290 psi for untreated and 450 psi for MAPP-coated model composites. These values are of the same order of magnitude as the LLDPE matrices in this study. However, Felix and Gatenholm (1994) showed appreciably higher value of 885 psi (6.1 Mpa), as compared to 416 psi in the control group in this study (PP-NO-0, i.e., without E-43 and adhesive). This difference may be due to the presence of air bubbles in the interphase of wood and matrix interfering with stress transfer between the two bulk phases. Although birch dowels were carefully oven-dried, applying adhesive and E-43 on the contact area before embedding in the matrix polymer could cause the increase moisture content on the interface. Even though the coated specimens were stored in desiccators, they might not be dry enough to eliminate excess moisture. Moreover, condensation water upon curing of UF and PF resins might also be responsible for the presence of air bubbles.

The ANOVA in Table 3.12 reveals the significance of the highest order interaction (PT*AT*CON) ($F=4.0$), however, the dominating plastic type (PT) ($F=537.8$), adhesive type (AT) ($F=54.4$) and E-43 concentration (CON) ($F=190.7$) main effects should also be presented. The effect of plastic type on interfacial

Table 3.11. Interfacial shear strength between plastic matrices and birch wood dowels treated with various E-43 concentration (CON) and adhesives.

Plastic type / CON	Adhesive type			
	NO	UF	ISO	PF
- % -	psi			
LDPE				
0	176.9 (14.6) ¹	186.5 (35.3)	410.3 (33.3)	111.9 (12.4)
10	279.4 (34.4)	235.0 (49.4)	341.9 (52.6)	287.5 (60.1)
20	259.2 (30.0)	228.0 (36.8)	316.7 (53.2)	240.2 (82.3)
30	219.6 (38.3)	211.4 (33.0)	335.9 (57.0)	170.1 (33.1)
40	178.3 (29.5)	151.9 (29.4)	262.8 (15.7)	153.9 (51.9)
LLDPE				
0	253.1 (15.8)	163.2 (16.9)	400.7 (69.1)	140.5 (13.5)
10	492.9 (29.8)	430.8 (58.4)	313.4 (39.2)	513.0 (47.8)
20	561.7 (83.3)	430.9 (82.2)	286.6 (40.7)	408.2 (62.3)
30	531.0 (40.7)	424.0 (96.4)	321.7 (25.3)	272.9 (34.1)
40	377.8 (78.6)	339.0 (69.8)	259.4 (27.0)	278.1 (38.8)
PP				
0	416.1 (62.4)	316.2 (40.7)	311.1 (41.2)	255.5 (31.3)
10	906.8 (106.8)	1001.3 (126.1)	904.8 (123.7)	767.9 (149.3)
20	967.5 (138.6)	895.9 (92.4)	861.3 (138.4)	744.1 (83.5)
30	918.1 (74.9)	895.9 (160.7)	663.9 (94.3)	566.3 (97.6)
40	818.0 (101.1)	769.7 (110.1)	649.8 (90.0)	524.5 (72.6)

¹. Numbers in parentheses are standard deviations. Sample size=6.

Table 3.12. ANOVA based on original data on effects of plastic type (PT), adhesive type (AT), and E-43 concentration (CON) on interfacial shear strength of birch dowels and plastic matrices.

Source	DF	MS	F	P value
PT	2	7137437.69	537.78	0.0001
REP(PT)	15	13272.04	2.82	0.0004
AT	3	255766.70	54.44	0.0001
CON	4	895722.40	190.66	0.0001
AT*CON	12	44440.78	9.46	0.0001
PT*AT	6	154844.51	32.96	0.0001
PT*CON	8	283293.55	60.30	0.0001
PT*AT*CON	24	18815.65	4.00	0.0001
ERROR	285	4698.11	---	----

shear strength (Figure 3.14) indicates an order of PP>LLDPE>LDPE, which was the same as contact angle measurement, i.e., the opposite of wettability (Figures 3.8 and 3.10). The average interfacial shear strength of PP (708 psi) is 2-3 times stronger than those of LDPE (238 psi) and LLDPE (360 psi). Though MAPP may react identically with wood at the interface of wood-polyolefin composites, its reaction to plastic matrices may be different. The attraction between MAPP and polyolefin polymer is mostly Van der Waals physical interaction. Since MAPP is configurationally more like PP, the attraction between MAPP and PP may be stronger than its counterparts, therefore exhibiting higher interfacial shear strength. Moreover, the differences among the three plastic types also coincided with their degrees of crystallinity.

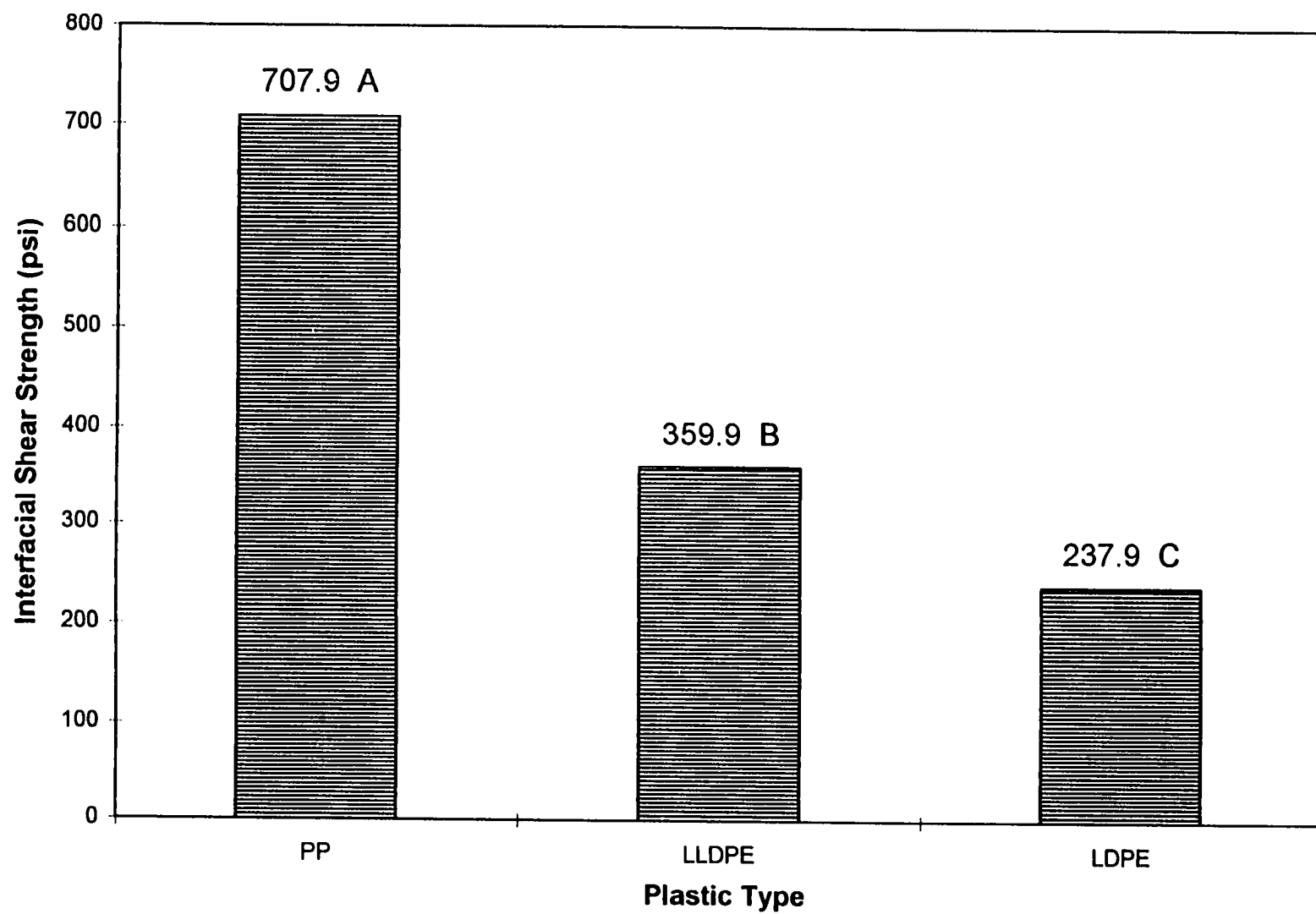


Figure 3.14. Interfacial shear strength of wood-polyolefin model composites by plastic type.

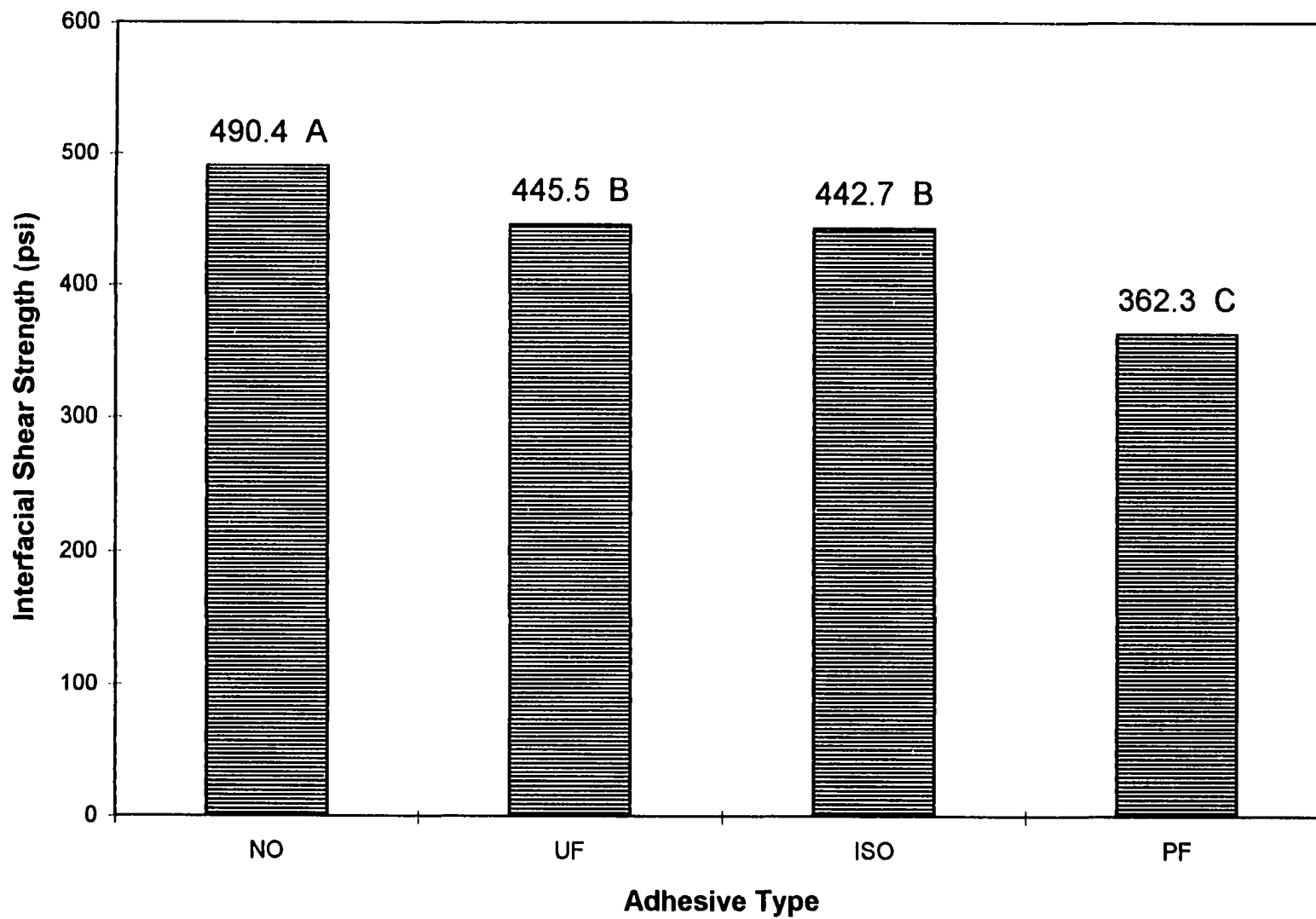


Figure 3.15. Interfacial shear strength of wood-polyolefin model composites by adhesive type.

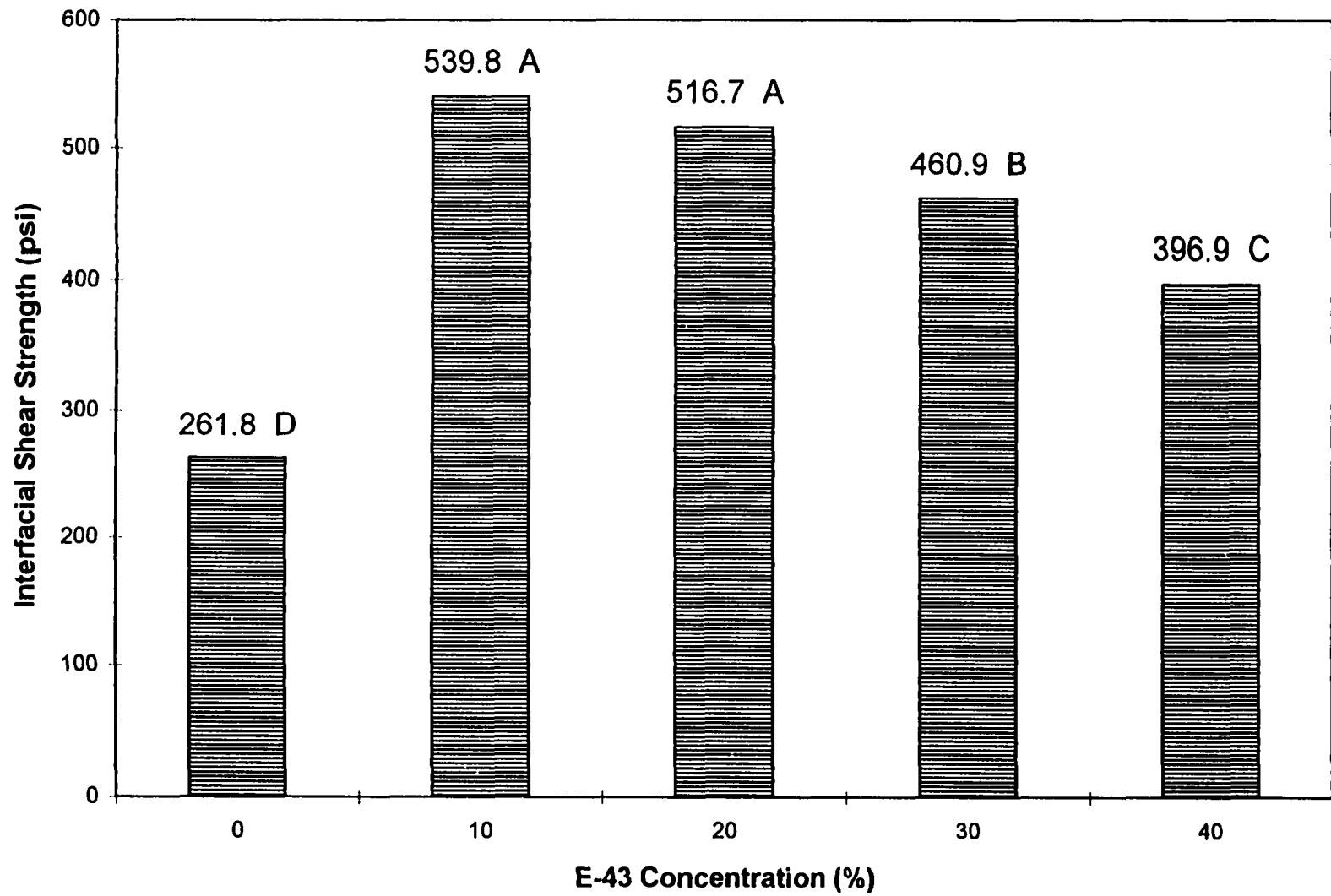


Figure 3.16. Interfacial shear strength of wood-polyolefin model composites by E-43 concentration.

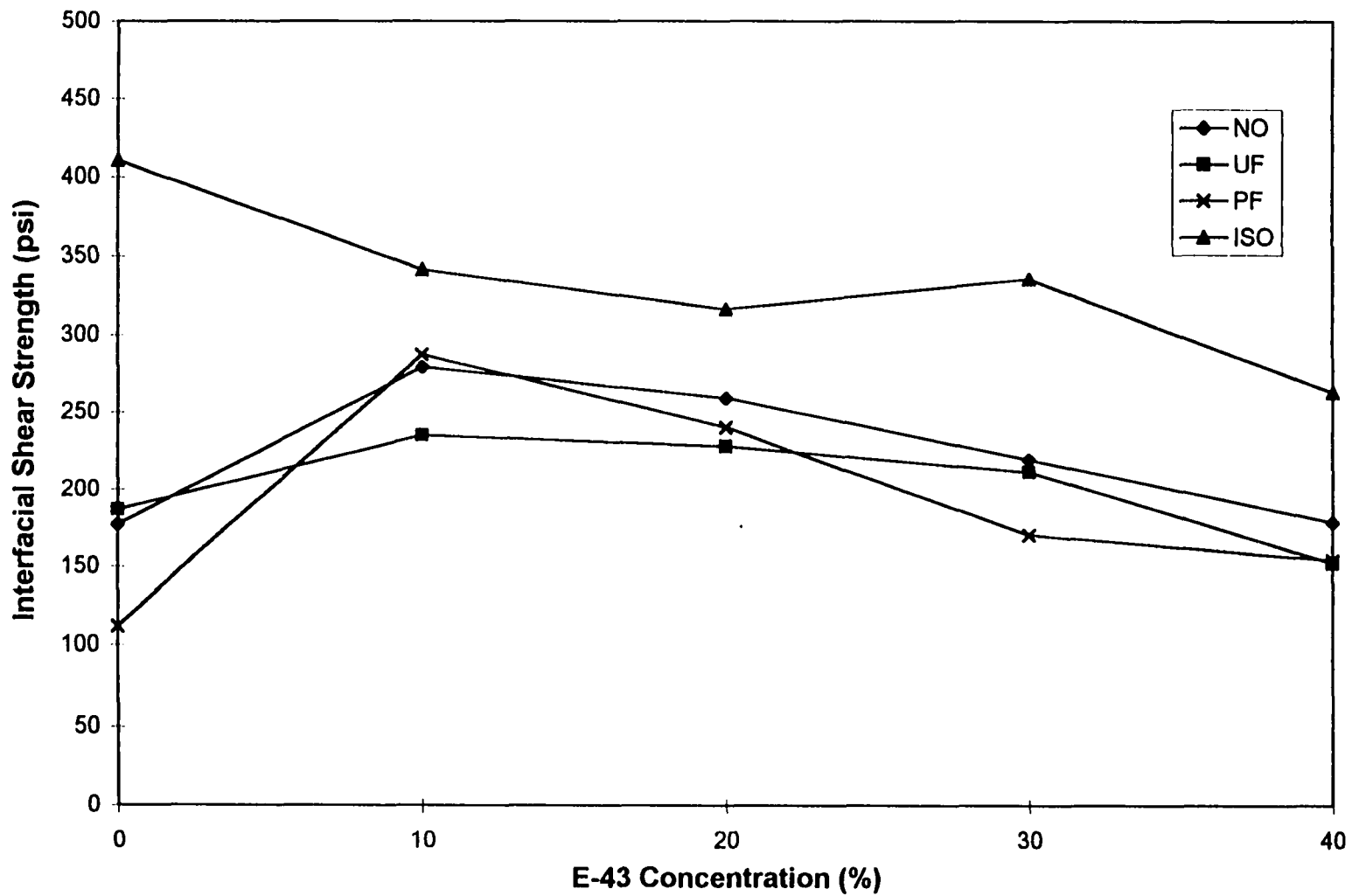


Figure 3.17. Dependence of interfacial shear strength on E-43 concentration and adhesive type for wood-LDPE model composites.

The effect of the adhesive type on interfacial shear strength (Figure 3.15) indicates that if other factors were combined, wood-polyolefin composites without adhesive as binder (NO) showed superior interfacial shear strength, whereas PF performed poorly in the interphase. These do not conform to the results of contact angle measurements in wood or plastic (Figures 3.7 and 3.11). Conversely, the effect of E-43 concentration on interfacial shear strength (Figure 3.16) somewhat coincided to the wettability of plastics (Figure 3.9), since the interfacial shear strength decreased with increasing contact angle (i.e., decreasing wettability).

The three-way interaction (PT*AT*CON) was analyzed individually by plastic type. For LDPE (Figure 3.17), the performance of ISO was superior to the other three adhesives at all E-43 concentration levels. Unlike the other three adhesives which trended upward and then downward with increasing E-43 concentration, ISO showed a downward trend with increasing E-43 concentration despite some minor fluctuations. UF-bonded composites exhibited comparable mean interfacial shear strength with that of no-adhesive-bonded composites at 0% E-43 concentration, and had slightly lower values at all other concentration levels. The increment in interfacial shear strength was most pronounced in PF from 0% to 10% E-43 concentration. However, the reduction in interfacial shear strength from 10% to 40% E-43 concentration level was most drastic in PF. Interfacial shear strength of PF was comparable with no-adhesive (NO) at 10% E-43 concentration, and comparable with UF at 40% E-43 concentration.

Similar to LDPE, interfacial shear strength of LLDPE bonded with ISO trended downward with increasing E-43 concentration (Figure 3.18). That is, the addition of E-43 showed a negative effect in the interfacial shear strength of ISO-bonded LDPE and LLDPE. For the other three adhesives in LLDPE, the trends were also similar to those of the three adhesives in LDPE. The improvement upon addition of E-43 was most dramatic in PF-bonded LLDPE, with interfacial shear strength increased by approximately 3.5 times (140 psi for PF-0 vs 513 psi for PF-10). However, it was most sensitive to increase in E-43 concentration. PF had equivalently high interfacial shear strength with no-adhesive bonded LLDPE at 10% concentration level, and it had equally low interfacial shear strength with ISO at 40% E-43 concentration level. The most favorable E-43 concentration for NO, UF, and PF was in the range between 0% to 10%.

In PP, the responses of interfacial shear strength to different adhesive types and concentration levels are shown in Figure 3.19. All four types of adhesive showed similar trends in which interfacial shear strength trended upward and then downward with increasing E-43 concentration. UF showed superior interfacial shear strength to PF at all concentration levels, and superior to ISO upon the addition of E-43. The interfacial shear strengths for UF, PF and ISO showed a negative response when E-43 concentration level exceeded 10%. Conversely, the interfacial shear strength of the composites without adhesive (NO) increased slightly with an increase in concentration from 0 to 20%, then fell beyond 20% concentration.

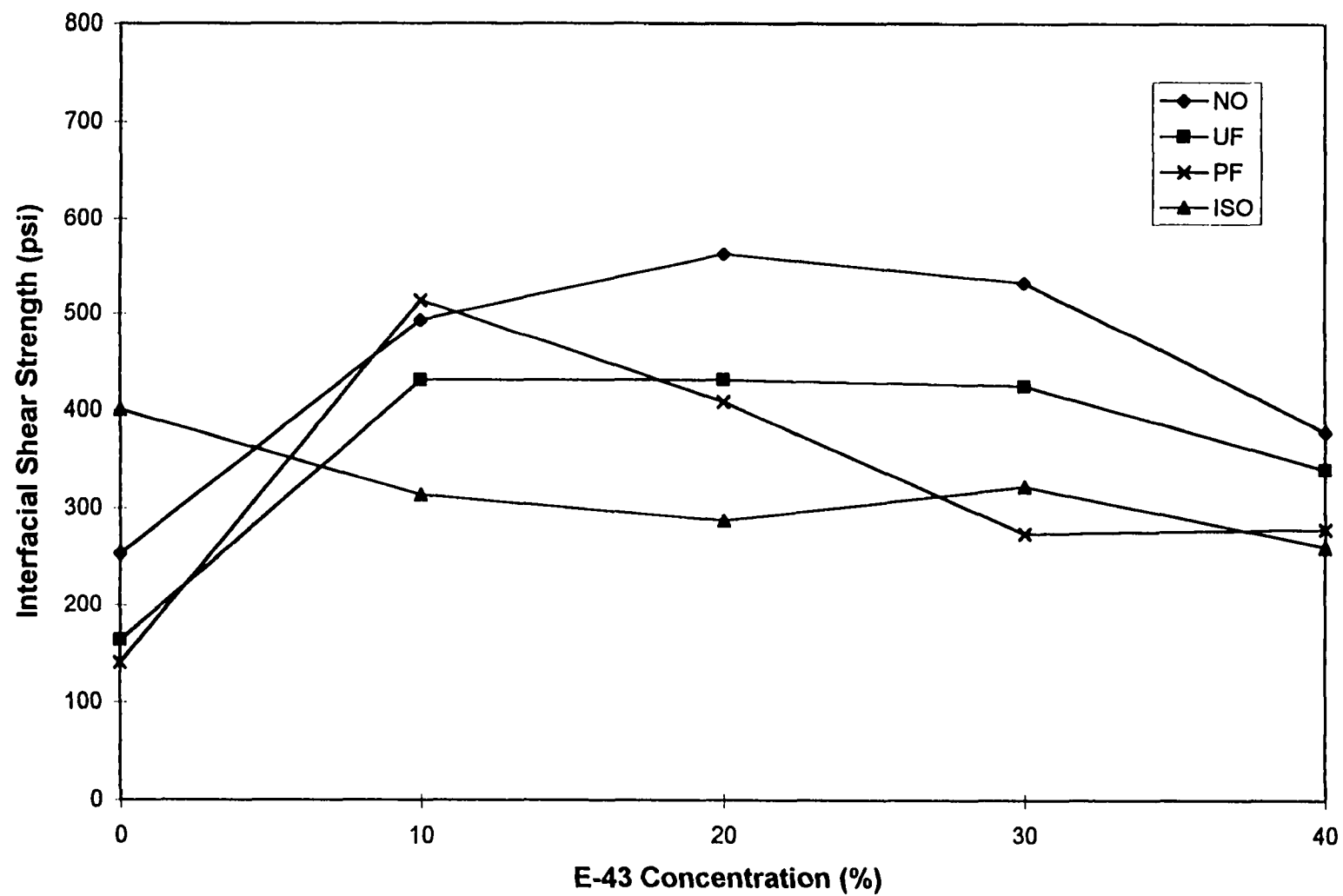


Figure 3.18. Dependence of interfacial shear strength on E-43 concentration and adhesive type for wood-LLDPE model composites.

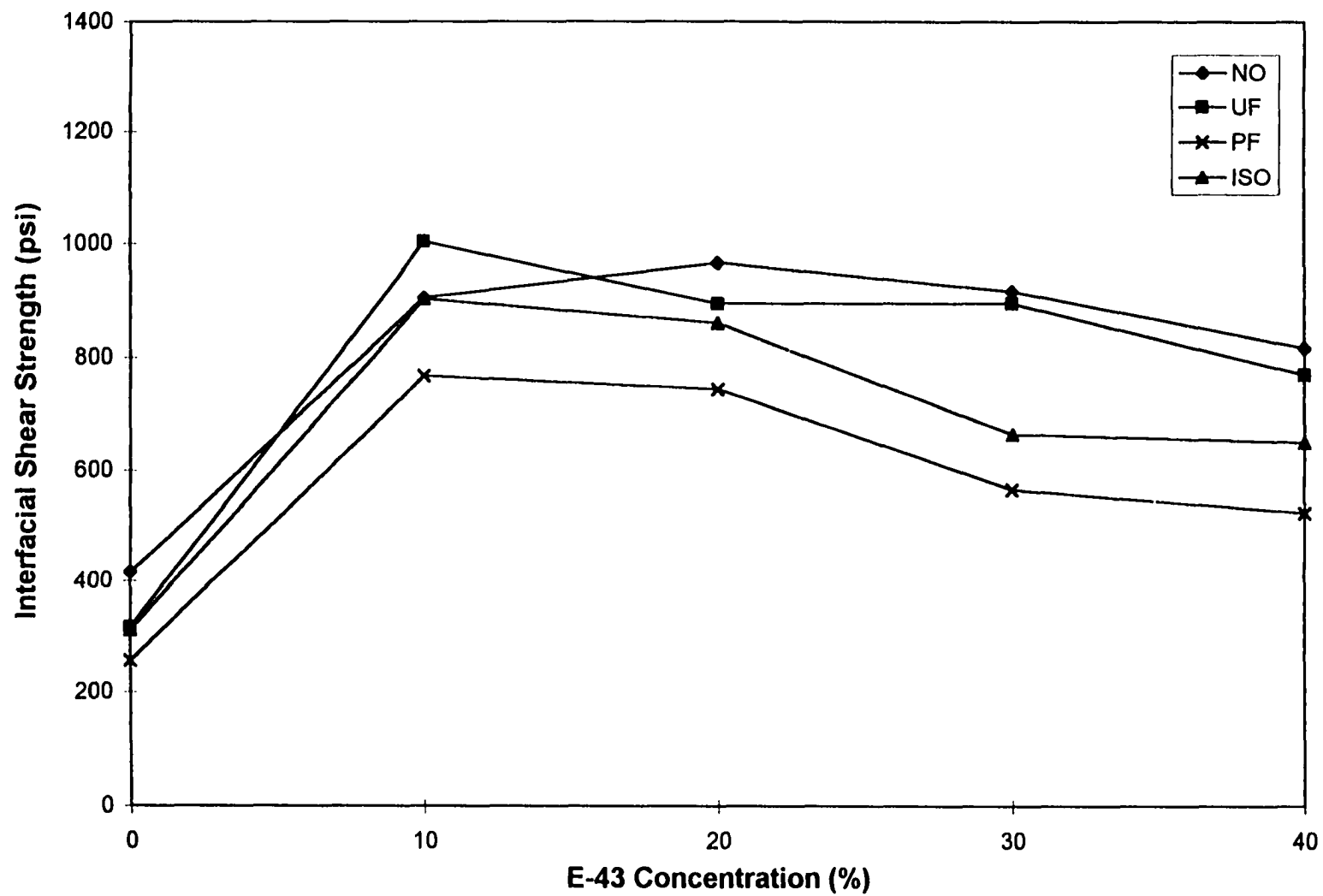


Figure 3.19. Dependence of interfacial shear strength on E-43 concentration and adhesive type for wood-PP model composites.

In terms of interfacial shear strength, as indicated in Table 3.11, the most appropriate treatment combinations were ISO-0 (ISO-bonded with 0% E-43 concentration) for LDPE, NO-20 (no-adhesive-bonded with 20% E-43 concentration) for LLDPE, and UF-10 (UF-bonded with 10% E-43 concentration) for PP. Except for ISO-bonded LDPE and LLDPE, addition of E-43 showed a positive effect on interfacial shear strength, but, lower concentrations were preferred. E-43 had the highest efficiency in PP.

Table 3.13. Solid E-43 content (SCC) and solid resin content (SRC) for wood dowels and plastic matrices with plastic type combined.

CON	Adhesive type ¹			
	NO	UF	ISO	PF
- % -	lb per 1000 sq ft			
SCC				
0	---	---	---	---
10	2.16 (0.55) ²	0.58 (0.23)	0.84 (0.21)	1.13 (0.31)
20	4.93 (1.64)	1.37 (0.84)	1.96 (0.44)	2.19 (0.63)
30	5.72 (1.33)	1.85 (0.55)	3.03 (0.67)	3.37 (1.31)
40	8.73 (1.47)	3.64 (1.24)	4.90 (0.71)	5.21 (0.98)
SRC				
0	---	7.84 (1.84)	3.95 (0.68)	5.66 (1.24)
10	---	7.51 (1.69)	3.77 (0.46)	5.42 (1.22)
20	---	8.22 (1.49)	4.15 (0.95)	5.39 (0.83)
30	---	8.18 (1.19)	3.80 (0.89)	5.43 (0.59)
40	---	8.11 (1.48)	3.92 (0.99)	5.64 (1.20)

¹. ISO, PF, UF and NO denote polyisocyanate, phenol formaldehyde, urea formaldehyde, and no adhesive, respectively.

². Numbers in parentheses are standard deviations.

The relationship between contact angle and interfacial shear strength was not consistent in this study. Liu and co-workers (Liu *et al.* 1994) tried to correlate the total surface energy and polar component of the surface energy with interfacial shear strength as measured by the microbond method. The results were inconclusive since there was no statistical support. The positive relationship between fiber surface characteristics and interfacial shear strength was reported to be valid only for heat-treated and acetylated fibers. However, wetting process is merely the first step toward good adhesion. The solidification of adhesive and the formation of a deformable joint also play important roles in interface adhesion. The correlation between wettability (contact angle) and interfacial shear strength is still not clear at this point; however, this study shows that good wetting is not a guarantee of good adhesion.

Solid E-43 content (SCC) and solid resin content (SRC) were also calculated for each specimen. Results of solid E-43 content and solid resin content with plastic type combined are listed in Table 3.13. Due to manual application of E-43, solid E-43 content showed some variations with different adhesives. Specimens which have no adhesive (NO) received and absorbed more E-43 than those which have adhesive. The application of E-43 after an adhesive was brushed on the interface area around the dowel may be the reason for higher variations in solid E-43 content. The overall solid E-43 contents for all concentration levels were 1.18, 2.61, 3.49, and 5.62 lb/1000 ft² from 10% through 40% level, respectively. In solid resin content, there were minimal variations

among all E-43 concentration levels within each adhesive type; but greater variations among adhesive types. UF had the highest average solid resin content, whereas ISO had the lowest. With all other factors pooled, solid resin contents were 3.92, 5.51, 7.97 lb/1000 ft² for ISO, PF, and UF, respectively. All plastic types were assumed to obtain same loading of solid E-43 content and solid resin content.

From regression analyses, interfacial shear strength was found to be independent of solid resin content but dependent on solid E-43 content. Therefore, solid resin content was excluded from the model fitting. Regression analyses for solid E-43 content for all treatment combinations are shown in Table 3.14. Polynomial regression models best fit most of the treatment combinations, only interfacial shear strength of ISO-bonded LDPE showed linear dependence on solid E-43 content. The partial coefficients also indicate that, except for the ISO-bonded LDPE and LLDPE, interfacial shear strength increases with initial addition of E-43 and decreases as solid E-43 content further increases. Coefficient of determinations (R^2) of all models show that solid E-43 content accounted for approximately 20 to 74% of the variabilities of interfacial shear strengths in wood dowel-polyolefin composites, indicating that some other factors may also contribute to the variation in interfacial shear strengths. The interaction between adhesive and E-43 may be responsible for this phenomenon.

Table 3.14. Statistics and coefficients for polynomial regression analysis of SCC (X) and ISS (Y) with different plastic and adhesive types as denoted by model $Y=b_0+b_1X+b_2X^2$.

Plastic / Adhesive type ¹	Statistics			Partial coefficient		
	F	P	R ²	b ₀	b ₁	b ₂
LDPE						
ISO	19.734	0.0001	0.4134	383.478	-22.221	---
NO	8.466	0.0014	0.3854	199.435	23.798	-2.739
UF	4.882	0.0155	0.2656	197.675	25.631	-7.601
LLDPE						
ISO	11.533	0.0002	0.4607	382.965	-55.477	6.669
NO	15.659	0.0001	0.5370	290.320	87.697	-7.679
PF	3.646	0.0397	0.2126	246.591	90.820	-
UF	11.548	0.0002	0.4610	229.218	191.694	-39.239
PP						
ISO	4.361	0.0228	0.2442	515.730	202.645	-34.933
NO	38.567	0.0001	0.7407	480.765	189.800	-17.229
PF	5.711	0.0085	0.2973	400.041	184.755	-29.105
UF	7.200	0.0031	0.3478	582.903	353.379	-70.582

¹. ISO, PF, UF and NO denote polyisocyanate, phenol formaldehyde, urea formaldehyde, and no adhesive, respectively.

². Linear model $Y=b_0+b_1X$ is better than polynomial model.

Table 3.15. Frequencies of failure mode on the interface of wood-plastic model composites.

Adhesive type ¹		NO			UF			ISO			PF		
Failure mode ²		0	1	2	3	0	1	2	3	0	1	2	3
Plastic type/ CON ³													
LDPE													
-- % --													
0		6	0	0	0	6	0	0	0	1	1	4	0
10		5	0	1	0	6	0	0	0	2	1	3	0
20		5	0	1	0	6	0	0	0	2	1	3	0
30		6	0	0	0	6	0	0	0	2	1	3	0
40		6	0	0	0	6	0	0	0	2	1	3	0
LLDPE													
0		6	0	0	0	6	0	0	0	5	0	1	0
10		6	0	0	0	6	0	0	0	5	0	1	0
20		6	0	0	0	6	0	0	0	6	0	0	0
30		6	0	0	0	6	0	0	0	6	0	0	0
40		6	0	0	0	6	0	0	0	6	0	0	0
PP													
0		6	0	0	0	4	0	2	0	3	0	3	0
10		0	1	1	4	0	0	2	4	0	0	1	5
20		0	0	0	6	0	0	2	4	0	0	2	4
30		0	0	0	6	0	0	3	3	2	0	1	3
40		1	0	1	4	1	1	3	1	1	0	5	0

¹. ISO, PF, UF and NO denote polyisocyanate, phenol formaldehyde, urea formaldehyde, and no adhesive, respectively.

². 0: clean interface; 1: wood failure; 2: plastic failure; 3: undetermined due to broken dowel.

³. CON: E-43 concentration.

3.4.2.2. Failure Mode and Air Bubble Occurrence

Failure mode on the interface was recorded and presence of air bubbles was examined following the fiber pull-out test. The frequencies of failure mode are presented in Table 3.15. E-43 showed extremely low affinities for LDPE and LLDPE, as their failure modes were exclusively “clean interface” due to weak adhesive force. Plastic and wood failures occurred in more than two thirds of ISO-bonded LDPE model composites, indicating better interface adhesion with stronger adhesion forces than cohesion forces. These results are in concordance with average interfacial shear strength magnitudes in Table 3.11. The addition of E-43 did not show any distinguishable differences among the four adhesive types in LDPE and LLDPE, but the addition of E-43 caused a much higher percentage of “undetermined” failure mode in NO-, UF-, and ISO-bonded PP model composites. Undetermined failure mode is caused by dowel failure at the grip end while the interface remains intact. Dowel failure was due to loss of strength from thermal degradation when wood dowels were subjected to 200°C for 30 minutes during the manufacture of PP model composites. When the adhesive bonding between wood dowel and PP is not very strong, as was the case in 0% E-43 concentration, the wood dowel can be pulled out before it breaks. On the other hand, if the adhesion force on the interface exceeds dowel strength, the dowel breaks. Thus the intrinsic interfacial shear strength values should be higher in those PP model composites.

The relationship between the percentage of adhesive failure and plastic type (Figure 3.20) shows that over 80% of LDPE, 98% of LLDPE, and 17% of PP model composites had adhesive failure. The relative ranking of adhesive failure in LDPE and LLDPE was consistent with that of interfacial shear strength. The relationship of interfacial shear strength with percentage adhesive failure, however, was not consistent with respect to PP due to the high frequency of undetermined mode failure. The effect of adhesive type (Figure 3.21) reveals that ISO exhibited the lowest percentage of adhesive failure, and the frequencies of adhesive failure for NO (no adhesive), PF, and UF were similar to one another. The effect of E-43 concentration on failure mode (Figure 3.22) indicates that adhesive failure was highest at 0% E-43 concentration, decreased appreciably when E-43 was added to the interface, and slightly increased with an increase in concentration level. This trend coincided with the results of interfacial shear strength (Figure 3.16).

The presence of air bubbles was recorded for every specimen. The occurrence of air bubbles was most abundant in LDPE (91%), followed by LLDPE (72%), and then PP (53%). The lower percentage of air bubbles in PP was due to the undetermined mode resulted from dowel failure. For adhesive types, air bubble frequency was higher in adhesive bonded composites (80-87%) than in composites without adhesive (37%). This is because UF and PF adhesives add moisture on the interface. As the moisture evaporates at elevated temperatures, it is trapped by molten plastic matrices and hence creates air

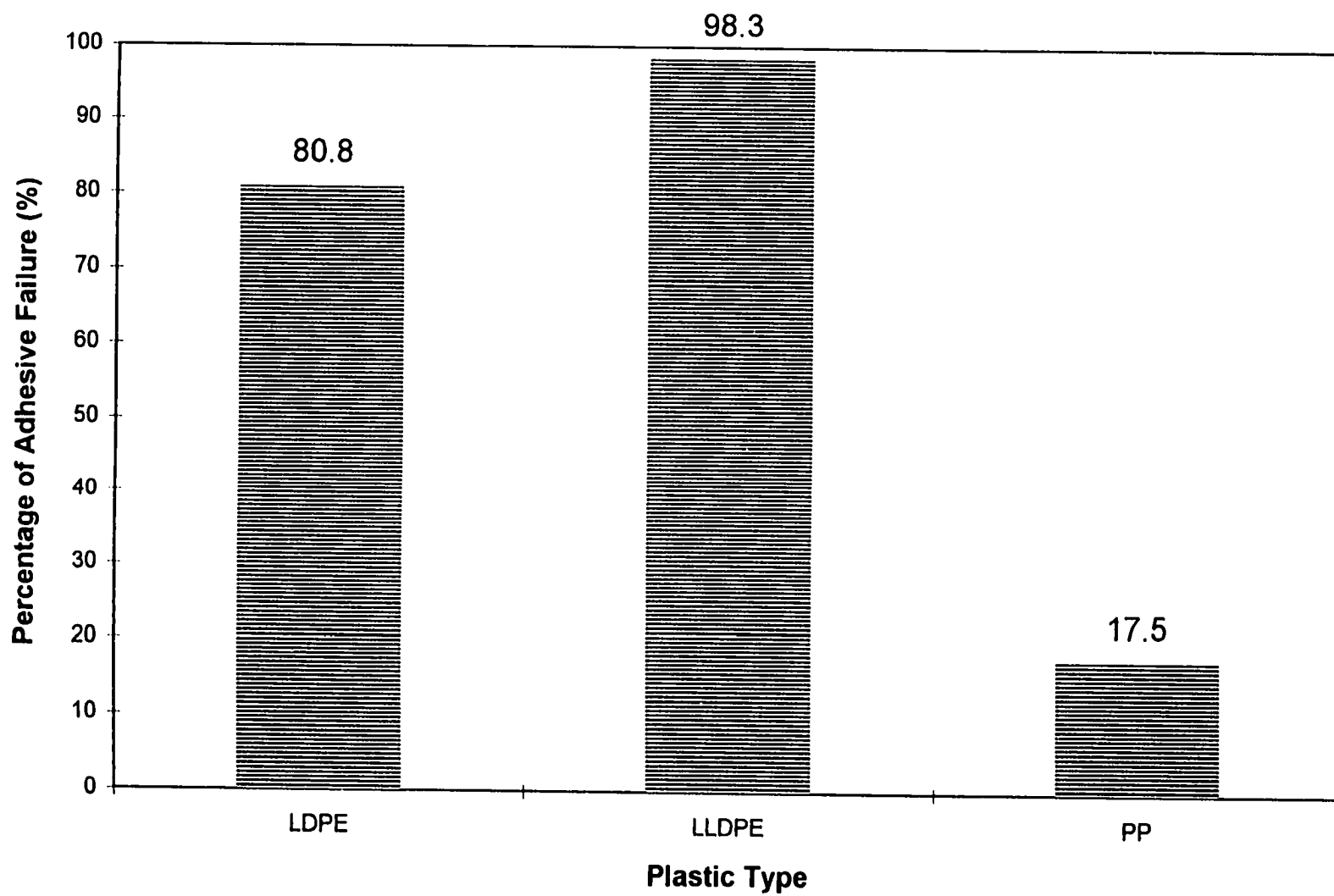


Figure 3.20. Percentage of adhesive failure in three wood-polyolefin model composites.

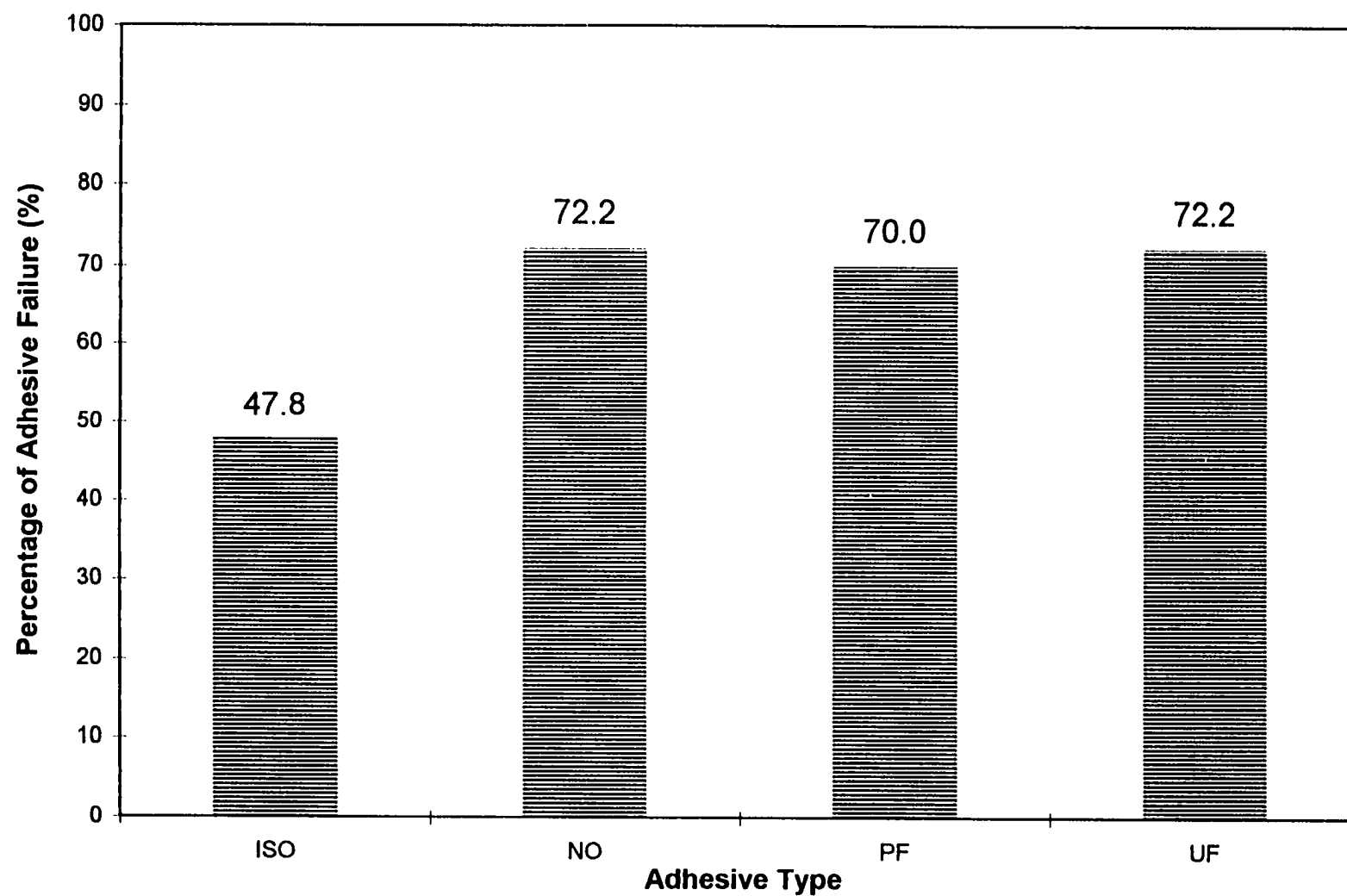


Figure 3.21. Percentage of adhesive failure in wood-polyolefin model composites by adhesive type.

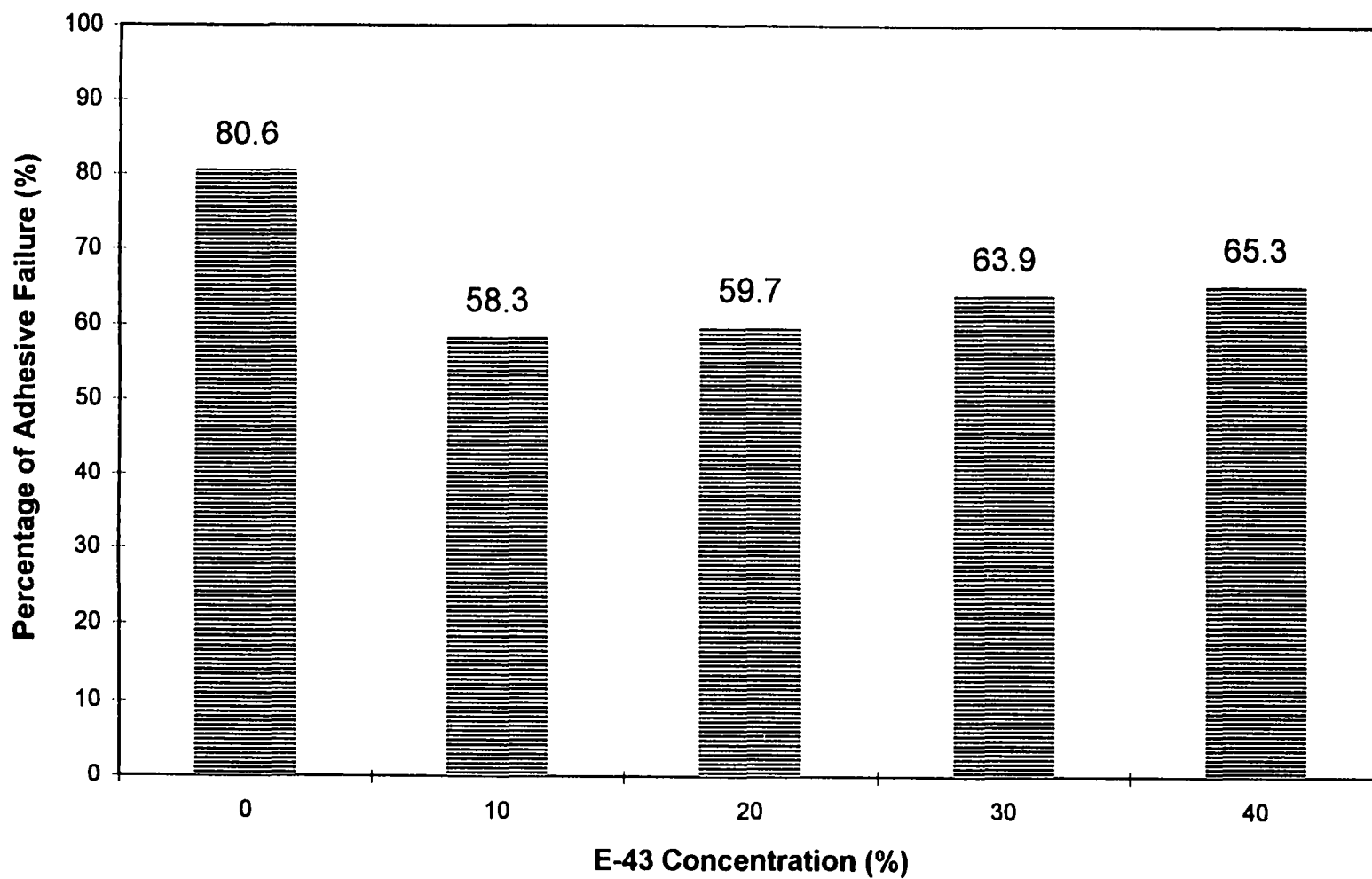


Figure 3.22. Percentage of adhesive failure in wood-polyolefin model composites by E-43 concentration.

bubbles on the interface. Air bubble frequencies in relation to E-43 concentration shows that 25% of the untreated composites were free of air bubbles. The presence of air bubble appeared to decrease as E-43 was added to the interface, and slightly increased with an increase in E-43 concentration level.

The presence of air bubbles may inhibit stress transfer, thus deteriorates the interface adhesion. The results presented here do not conform with interfacial shear strength values. This is predominately because the recording of air bubble occurrence was qualitative, not quantitative. The actual volume occupied by air bubbles is difficult to calculate. The amount of interface discontinuity interface caused by air bubbles is therefore undeterminable.

3.5. CONCLUSIONS

For E-43-treated wood surfaces, contact angles among four wetting liquids were in the order of PF>UF>ISO>WATER. Contact angles increased upon addition of E-43, then decreased as concentrations further increased for UF, PF, and ISO. Experiment results also show that the E-43-treated surface could be completely wetted by water, and partially wetted by three other wetting liquids. The reduction in contact angle was most abrupt between 0% and 10% E-43 concentration for water, and between 10% and 20% for UF and PF. ISO was most sensitive to the addition of E-43, and least sensitive to the increase in E-43 concentration.

Among the three polyolefin polymers, the contact angles of PP, either wetted with various concentration of E-43 or with various wetting liquids, showed

the highest value but was less sensitive to changes in E-43 concentration. However, contact angles of LDPE and LLDPE gradually increased with an increase in E-43 concentration. For all four wetting liquids, their contact angle ratings for different plastic types all followed the sequences of PP>LLDPE>LDPE and water>UF>PF>ISO.

Interfacial shear strengths measured by a modified single-fiber pull-out test indicate that the overall average interfacial shear strength was highest in PP and lowest in LDPE. ISO-bonded LDPE without E-43 treatment showed the highest interfacial shear strength among all other LDPE model composites. On the other hand, E-43-treated wood surface without addition of adhesive displayed overall superior performance than all other adhesive-bonded LLDPE. Addition of E-43 greatly enhanced the interfacial shear strengths for all plastic types, except for ISO-bonded LDPE and LLDPE. The effect of E-43 was most pronounced in PP, and less in LDPE and LLDPE. However, increasing E-43 concentration from 10% to 40% exhibited negative effects toward interfacial shear strength. Regression analyses for the interfacial shear strength on E-43 solid content for all adhesive and plastic type combinations show that interfacial shear strength could be partly explained (20-75%) by solid E-43 content, except in PF-bonded LDPE. There was no correlation between interfacial shear strength and solid resin content for all adhesive types. The most efficient amount of solid E-43 content to improve interfacial shear strength did not coincide with

the amount to achieve best wettability. Furthermore, better wetting did not indicate better interfacial bond strength.

Adhesive failure prevailed in LDPE and LLDPE model composites while PP showed predominately cohesive failures either in wood dowel or in plastic matrix. About one third of PP model composites exhibited dowel failure at the grip end, resulting in undetermined failure mode; therefore, their true interfacial shear strengths are believed to be higher than the apparent interfacial shear strengths. Results of failure mode were in good agreement with interfacial shear strengths. This modified single-fiber pull-out test showed great feasibility for measuring interfacial shear strength. However, air bubbles resided at the interfaces of approximately 80% of the model composites which might impair stress transfer. Further improvement in this method would increase its applicability.

3.6. LITERATURE CITED

Andrade, J.A. 1988. Polymer surface and interface dynamics: an introduction. In: J.D. Andrade (ed.). Polymer Surface Dynamics. Plenum Press, New York. pp. 1-8.

Casilla, R.C., S. Chow, and P.R. Steiner. 1981. An immersion technique for studying wood wettability. Wood Sci. Technol. 15:31-43.

Casilla, R.C., S. Chow, P.R. Steiner, and S.R. Warren. 1984. Wettability of four Asian meranti species. Wood Sci. Technol. 18:87-96.

Collier, J.R., M. Lu, M. Fahrurrozi, and B. J. Collier. 1995. Reactive composite systems. In: D. F. Caulfield, R. M. Rowell, and J. A. Youngquist (eds.), Woodfiber-Plastic Composites: virgin and recycled wood fiber and polymers for composites. Forest Product Society, 1995: 67-73.

Czvikovszky, T., V. Lopata, G. Boyer, W. Kremers, C. Saunders, and A. Singh. 1993. Electron-beam processing of wood flour-reinforced polypropylene. In: M.P. Wolcott (ed.), *Wood-Fiber/Polymer Composites: fundamental concepts, processes, and material options*: Proc. 1st Wood Fiber-Plastic Composite Conference, Madison, WI. Forest Products Society, 1993: 68-74.

Felix, J.M. and P. Gatenholm. 1991. The nature of adhesion in composites of modified cellulose fibers and polypropylene. *J. Appl. Poly. Sci.* (42):609-620.

Felix, J.M. and P. Gatenholm. 1994. Effect of transcrystalline morphology on interfacial adhesion in cellulose/polypropylene composites. *J. Mater. Sci.* 1994:3043-3049.

Felix, J.M., P. Gatenholm, and H.P. Schreiber. 1994. Plasma modification of cellulose fibers: Effect of some polymer composite properties. *J. Appl. Poly. Sci.* (51):285-295.

Freeman, H.A. 1959. Relation between physical and chemical properties of wood and adhesion. *For. Prod. J.* 9(12): 451-458.

Freeman, H.A. and F.F. Wangaard. 1960. Effect of wettability of wood on glue-line behavior of two urea resins. *For. Prod. J.* 10(6): 311-315.

Gardner, D.J., N.C. Generalla, D.W. Gunnells, and M.P. Wolcott. 1991. Dynamic wettability of wood. *Langmuir* 7(11): 2498-2501.

Gardner, D.J., F.P. Liu, M.P. Wolcott, and T.G. Rials. 1994. Improving interfacial adhesion between wood fibers and thermoplastics: A case study examining chemically modified wood and polystyrene. In: P.R. Steiner (ed.), *Proc. 2nd Pacific Rim Bio-Based Composites Symposium*, Vancouver, Canada. University of British Columbia, 1994:55-63.

Gatenholm, P., J. Felix, C. Klason, and J. Kubát. 1993. Methods for improvement of properties of cellulose-polymer composites. In: M.P. Wolcott (ed.), *Wood-Fiber/Polymer Composites: fundamental concepts, processes, and material options*. Proc. 1st Wood Fiber-Plastic Composite Conference, Madison, WI. Forest Products Society; 1993: 20-24.

Gent, A.N. and G.R. Hamed. 1990. Fundamentals of adhesion. In: I. Skeist (ed.), *Handbook of Adhesives*. Van Nostrand Reinhold Inc. New York. 3rd ed. pp.39-73.

Gray, V.R. 1962. The wettability of wood. *For. Prod. J.* 12(9):452-461.

Gunnells, D.W., D.J. Gardner, and M.P. Wolcott. 1994. Temperature dependence of wood surface energy. *Wood Fiber Sci.* 26(4):447-455.

Herczeg, A. 1965. Wettability of wood. *For Prod. J.* 15(11):499-506.

Hodgson, K.T. and J.C. Berg. 1988. Dynamic wettability properties of single wood pulp fibers and their relationship to absorbency. *Wood Fiber Sci.* 20(1): 3-17.

Hse, C.Y. 1972. Wettability of southern pine veneer by phenol formaldehyde wood adhesives. *For. Prod. J.* 22(1):51-57.

Hull, D. 1981. An introduction to composite materials. Chapter 3. 1st ed. Cambridge University Press, New York.

Jacobson, R.E., R.M. Rowell, D.F. Caulfield, and A.R. Sanadi. 1995. Property improvement effects of agricultural fibers and wastes as reinforcing fillers in polypropylene-based composites. In: D.F. Caulfield, R. M. Rowell, and J. A. Youngquist (eds.), *Woodfiber-Plastic Composites: virgin and recycled wood fiber and polymers for composites*. Forest Product Society, 1995: 211-219.

Johns, W.E. 1982. Isocyanates as wood binders – a review. *J. Adhesion.* 15: 59-67.

Johnson, R.E. and R.H. Dettre. 1993. Wetting of low-energy surfaces. In: J.C. Berg (ed.), *Wettability*. Marcel Dekker, Inc. New York. pp. 1-74.

Jordan, D.L. and J.D. Wellons. 1977. Wettability of Dipterocarp veneers. *Wood Sci.* 10(1):22-27.

Kalnins, M.A. and W.C. Feist. 1993. Increase in wettability of wood with weathering. *For. Prod. J.* 43(2):55-57.

Kalnins, M.A. and C. Katzenberger. 1987. Wettability and water repellency of wood: a faster, more convenient method. In: J.F. Kennedy, G.O. Phillips, and P.A. Williams (eds.), *Wood and Cellulosics: industrial utilization, biotechnology, structure and properties*. Ellis Horwood Ltd., West Sussex, England. Chapter 45. pp. 409-416.

Karmaker, A.C. and J.A. Youngquist. 1995. Fiber attrition during injection molding and its effect on the mechanical performance of natural fiber-reinforced polypropylene composite. In: D. F. Caulfield, R. M. Rowell, and J. A. Youngquist (eds.), *Woodfiber-Plastic Composites: virgin and recycled wood fiber and polymers for composites*. Forest Product Society; 1995: 96-98.

Kikata, Y. and K. Masuda. 1969. Wettability of wood. *Mokuzai Gakkaishi* 15(5):237-240.

Kishi, H., M. Yoshioka, A. Yamanoi, and N. Shiraishi. 1988. Composites of wood polypropylenes I. *Makuzai Gakkaishi* 34(2): 133-139.

Klungness, J.H. 1981. Measuring the wetting angle and perimeter of single wood pulp fibers: a modified method. *TAPPI* 64(12):65-66.

Kolosick, P.C., G.E. Myers, and J.A. Koutsky. 1993. Bonding mechanisms between polypropylene and wood: coupling agent and crystallinity effects. In: M.P. Wolcott (ed.), *Wood-Fiber/Polymer Composites: fundamental concepts, processes, and material options: Proc. 1st Wood Fiber-Plastic Composite Conference*, Madison, WI. Forest Products Society, 1993: 15-19.

Lee, S.B. and P. Luner. 1972. The wetting and interfacial properties of lignin. *Tappi*. 55(1):116-121.

Liu, F.P. 1994. Charactering interfacial adhesion between wood fibers and a thermoplastic matrix. Ph.D. dissertation, West Virginia University. pp. 5-8, pp.56-79.

Liu, F.P., T.G. Rials, M.P. Wolcott, and D.J. Gardner. 1995. Interactions between wood fibers and amorphous polymers. In: D. F. Caulfield, R. M. Rowell, and J. A. Youngquist (eds.), *Woodfiber-Plastic Composites: virgin and recycled wood fiber and polymers for composites*. Forest Product Society, 1995: 74-81.

Liu, F.P., M.P. Wolcott, P.J. Gardner, and T.G. Rials. 1994. Characterization of interface between cellulosic fibers and a thermoplastic matrix. *Comp. Interface* 2(6):419-432.

Luner, P. and M. Sandell. 1969. The wetting of cellulose and wood hemicelluloses. *J. Polymer Sci. Part C. No. 28*. pp. 115-142.

Miller, B., P. Muri, and L. Rebenfeld. 1987. A microbond method for determination of the shear strength of a fiber/resin interface. *Comp. Sci. Technol.* 28: 17-32.

Miller, B. and R.A. Young. 1975. Methodology for studying the wettability of filaments. *Textile Res. J.* 1975 May, 359-365.

Mohanakrishnan, C.K., R. Narayan, and J.D. Nizio. 1993. Reactive extrusion processing of polypropylene-lignocellulosic blend materials. In: M.P. Wolcott (ed.), *Wood-Fiber/Polymer Composites: fundamental concepts, processes, and*

material options: Proc. 1st Wood Fiber-Plastic Composite Conference, Madison, WI. Forest Products Society, 1993: 57-62.

Myers, G.E., I.S. Chahyadi, C. Gonzalez, and C.A. Coberly. 1993. Wood flour and polypropylene or high-density polyethylene composites: influence of maleated polypropylene concentration and extrusion temperature on properties. In: M.P. Wolcott (ed.), *Wood-Fiber/Polymer Composites: fundamental concepts, processes, and material options*. Proc. 1st Wood Fiber-Plastic Composite Conference, Madison, WI. Forest Products Society, 1993: 49-56.

Nguyen, T. and W.E. Johns. 1979. The effects of aging and extraction on the surface free energy of Douglas fir and redwood. *Wood Sci. Technol.* 13: 29-40.

Penn, L.S. S.M. Lee. 1989. Interpretation of experimental results in the single pull-out filament test. *J. Comp. Technol. Res.* 11(1):23-30.

Sanadi, A.R., R. M. Rowell, and R.A. Young. 1992. Estimation of fiber-matrix interfacial shear strengths in lignocellulosic-thermoplastic composites. In: R.M. Rowell, T.L. Laufenberg, and J.K. Rowell (eds.), *Materials Interactions Relevant to Recycling of Wood-Based Materials: Proc. Materials Research Society Symposium*; April 27-29, 1992, San Francisco, CA. Materials Research Society. Vol. 266:81-92.

Sanadi, A.R., R. M. Rowell, and R.A. Young. 1993. Evaluation of wood-thermoplastic-interphase shear strengths. *J. Mater. Sci.* 28:6347-6352.

Sanadi, A.R., K. Walz, L. Wieloch, R.E. Jacobson, D.F. Caulfield, and R. M. Rowell. 1995. Effect of matrix modification on lignocellulosic composite properties. In: D. F. Caulfield, R. M. Rowell, and J. A. Youngquist (eds.), *Woodfiber-Plastic Composites: virgin and recycled wood fiber and polymers for composites*. Forest Products Society, 1995: 166-172.

Sanadi, A.R., D.F. Caulfield, and R.E. Jacobson. 1997. Agro-fiber thermoplastic composites. In: R.M. Rowell, R. A. Young, and J.K. Rowell (eds.), *Paper and Composites from Agro-Based Resources*. Lewis Publishers, New York. pp.377-402.

Selke, S.E. and J. Childress. 1993. Wood fiber/high-density polyethylene composites: ability of additives to enhance mechanical properties. In: M.P. Wolcott (ed.), *Wood-Fiber/Polymer Composites: fundamental concepts, processes, and material options*. Proc. 1st Wood Fiber-Plastic Composite Conference; Madison, WI. Forest Products Society, 1993: 109-111.

Shaler, S.M. 1993. Mechanics of the interface in discontinuous wood fiber composites. In: M.P. Wolcott (ed.), *Wood-Fiber/Polymer Composites: fundamental concepts, processes, and material options*. Proc. 1st Wood Fiber-Plastic Composite Conference; Madison, WI. Forest Products Society, 1993: 9-14.

Tai, W.C., S.L. Quarles, and T.G. Rials. 1992. The effect of compatibilizers on interfacial bonding in lignocellulosic fiber/polyethylene composites. In: J.F. Kenndey, G.O. Phillips, and P.A. Williams (eds.), *Cellulosics: Chemical, Biochemical and Material Aspects*. Ellis Horwood, New York, 1992. pp 507-512.

Tao, W., J.R. Collier, and B.J. Collier. 1993. Evaluation of interfacial adhesion in sheath/core composite fibers. *J. Appl. Polym. Sci.* 47:1115-1122.

Troughton, G.E. and S.Z. Chow. 1971. Migration of fatty acids to white spruce veneer surface during drying: relevance to theories of inactivation. *Wood Sci.* 3(3):129-133.

Westerlind, B., M. Rigdahl, H. Hollmark, and A. De Ruvo. 1984. Interfacial properties of regenerated cellulose fiber and thermoplastic system. *J. Appl. Polym. Sci.* 29:175-185.

Woodhams, R.T., G. Thomas, and D.K. Rodgers. 1984. Wood fibers as reinforcing fillers for polyolefins. *Polymer Eng. Sci.* 24(15):1166-1171.

Woodhams, R.T., S. Law, and J.J. Balatinecz. 1993. Intensive mixing of wood fibers with thermoplastics for injection-molded composites. In: M.P. Wolcott (ed.), *Wood-fiber/polymer composites: Fundamental concepts, processes, and material options*. Proc. 1st Wood Fiber-Plastic Composite Conference, Madison, WI. Forest Products Society, 1993: 75-78.

Wu, S. 1982. *Polymer interface and adhesion*. Dekker, New York, p.184.

Young, R.A. 1976. Wettability of wood pulp fibers--applicability of methodology. *Wood Fiber* 8(2):120-128.

Zisman, W.A. 1964. Relation of the equilibrium contact angle to liquid and solid constitution. In: *Contact angle, wettability and adhesion*. Advanced in Chemistry Series No. 43. American Chemical Society. pp 1-51.

Zisman, W.A. 1976. Influence of constitution on adhesion. In: I. Skeist (ed.), *Handbook of Adhesives*, 2nd ed. Van Nostrand Reinhold Co. New York. pp 33-71.

CHAPTER 4

DEVELOPMENT OF WOOD FIBER-POLYPROPYLENE LAMINATES WITH DIFFERENT CONFIGURATIONS

4.1. INTRODUCTION

In lignocellulosic fiber-reinforced plastics, the dispersion of fibers in the matrices of plastics facilitates stress transfer within the composites, if there is adequate fiber-plastic interface adhesion. To improve interfacial adhesion, several surface treatments on either hydrophilic wood fibers or hydrophobic plastics have been studied. However, there are two main problems that affect the stress transfer between the two components: (1) the agglomeration of wood fibers, caused by interfiber hydrogen bond, which inhibits homogenous dispersion of fibers within matrix materials, and (2) the reduction of fiber aspect ratio and strength, caused by vigorous mixing during high-temperature processing, which deteriorate the composite strengths. On the other hand, a composite, in the form of fiber-plastic laminate, can be fabricated to retain interfiber hydrogen bonding, to avoid reduction in fiber aspect ratio, to enhance the interface adhesion by means of interpenetration, and eventually to serve load-bearing purposes.

The development of paper-plastic laminates dates back more than five decades when Cox and Pepper (1944) prepared paper-phenolic resin laminates.

Since then, the paper, plastic, and textile industries have developed a broad spectrum of fiber-thermoplastic composites, ranging from fiber-plastic laminates to nonwovens.

The forest products industry favors utilization of wood fibers as a major component. Theoretically, the maximum volume of compactly aligned cylindrical fibers packed into a composite is about 90%. However, when the fiber volume is 80%, composite properties decline due to the inability of plastics to infiltrate and wet the fibers. Generally, 70% fiber volume is the limit (Krock and Broutman 1967, Michell and Willis 1978). Laminates made of paper and plastic films are preferred because they allow a higher proportion of cellulosic fiber.

A paper-thermoplastic laminate can be made by several methods. One method is to interleave oriented or unoriented paper with plastic films, followed by hot pressing the stack to form a sandwich structure (Prud'homme 1977, Michell *et al.* 1978). Another method is to hot press a stack of plastic-impregnated or coated paper to form a layer structure (Zadorecki and Flodin 1985, Zadorecki and Flodin 1986, Michell *et al.* 1976). Plastics in the form of solid (Caulfield *et al.* 1993) or liquid (solution/dispersion) (Engman *et al.* 1976) can also be mixed with or added to cellulosic fibers to fabricate the composites. In the composites made from the latter methods, interfiber hydrogen bond is lacking; instead, thermoplasticity (Marchessault *et al.* 1975) or thermobond (Goldstein 1984) accounts for the interface adhesion.

A few studies have focused on the development of fiber-plastic laminates. For instance, Fisa and co-workers (Fisa and Marchessault 1974, Fisa *et al.* 1976) used encapsulation techniques to control the locus of polymerization and crystallization of polyethylene to the fiber surface. The resulting composites

maintain continuous a microporous structure of fibrous network while possessing the hydrophobic surface nature of plastic polymer. The microporosity provides channels for liquid and gas to pass through, creating the possibility of rubbing water into the sheet.

Using composite theories to oriented paper sheets (Prud'homme and Robertson 1976), Prud'homme (1977) compared the theoretical and experimental mechanical strengths of polymer-paper laminates made by sandwiching poly(methyl methacrylate) and low density polyethylene respectively to oriented and unoriented black spruce hollocellulose paper. He concluded that if the original paper is strong, the variation in modulus for both oriented and unoriented laminates and the breaking strength for unoriented laminates can be predicted by rule of mixture. On the other hand, if the original paper is weak, the polymer increased fiber-to-fiber bond strength of the paper, hence the predicted strength properties were lower than observed values.

Michell *et al.* (1976) investigated the strength properties of paper-polyethylene laminates made by hot-pressing kraft paper pre-coated with polyethylene. Mechanical properties of the resulting laminates were comparable with those of glass-filled high density polyethylene and paper-phenolic laminates. However, the composites were sensitive to relative humidity change, as evidenced in the reduction of strength properties. Later, Michell *et al.* (1978) tried to solve this problem by using acetylation of individual fibers and paper, and by crosslinking the paper with formaldehyde. The resultant wet strengths of

composites were greatly enhanced, but, at the expense of initial strength properties. Their work was summarized in Michell and Wallis (1978) and Campbell *et al.* (1980).

To enhance the adhesion between cellulose and plastics, Zadorecki and Flodin (1985, 1986) investigated the performance of fiber-polymer composites treated with coupling agents. The treated composites showed enhanced mechanical properties over untreated counterparts, both in dry and wet conditions, and the dimensional stabilities were also markedly improved. The improvement in environmental aging behavior for treated composites is due to the covalent bonds between treated fibers and polymers which allow the matrix to correspond with fiber upon shrinkage. The same goal was achieved by Westerlind *et al.* (1987) using the plasma treatment. The improved mechanical properties, as measured by double cantilever beam test, were attributed to the incorporation of polar groups in fiber surfaces which increased the surface energy of fiber. This promoted penetration of polyethylene as well as primary and secondary interfacial attractive forces of plasma treated polymer surfaces.

Engman *et al.* (1976) found that the mechanical properties of the plastics play a minor role in the composites made from conventional paper with high elastic and strength properties. However, the polymer can lower the shrinkage forces of the sheets by producing internal restraint, hence reducing the internal stress level in the material.

Although fiber-plastic laminates have not received great attention, their capability of incorporating a higher percentage of fiber as load-bearing material should not be ignored. A preliminary study was conducted to screen some factors that affect the performance of fiber-polymer laminates. Polypropylene, instead of polyethylene, was selected. The preliminary results indicated that, for a specific polymer film, fiber loading and the number of plastic layer might be two major reasons affecting the interpenetration of polymer melts into the fiber network.

This study was undertaken to further assess the effects of fiber loading and configuration on the mechanical properties and dimensional stability of untreated fiber-polypropylene laminates. In this laminate structure, fiber loading can be controlled by adjusting weight of handsheets while the number of plastic layers between two handsheets can also be manipulated by using commercially available thin films of polypropylene.

4.2. MATERIALS AND METHODS

A completely randomized design with factorial treatment arrangement was employed to investigate the effect of configurations on the tensile properties of wood fiber-plastic laminates. It included 3 levels of plastic layer (1, 2, and 3 ply) and 3 levels of fiber content (50, 60, and 70%). Twenty seven samples were produced with each treatment containing 3 replicates.

4.2.1. Materials

4.2.1.1. Wood Fiber

Southern pine kraft pulp was obtained from a local pulp mill with an average fiber length of 3.0 mm (standard deviation of 0.2 mm) and aspect ratio of 58.2 (standard deviation of 5.7).

4.2.1.2. Plastic Films

Polypropylene (PP) films, obtained from Plastic Suppliers Company, consisted of two-side corona treated OPP (oriented polypropylene) with nominal gauge 0.75 mil. The films were chosen as thin as possible so that the layers between the two consecutive handsheets could be manipulated.

4.2.2. Methods

4.2.2.1. Handsheet Preparation

In order to achieve more precise grammage (weight per unit area) control of the handsheets, southern pine Kraft pulp from the mill was first hand-squeezed to uniform dryness. For each 500 g of this squeezed pulp, 1 gallon water was added to make a slurry of similar consistency. The slurry was subjected to a vacuum process of 22 torr in a deckle box to remove excess water. The resulting mat consisted of a water to oven-dried pulp ratio of approximately 4:1.

Samples of calculated weight were taken from these mats to make the handsheets using a deckle box. However, unlike the regular handsheet forming, these handsheets remained on the screen until they were dried at room temperature and ready to peel off. Air-dried handsheets were cut and oven-

dried for later use. All plastic films and handsheets were cut into squares to make the 5 x 5- inch wood fiber-plastic laminates.

The total handsheet weight per laminate in each treatment was calculated from the nominal fiber content (NOMFC) of its respective target laminate. Since PP films of fixed thickness and area should have the same unit weight, this weight determined the number of PP sheets per laminate at given fiber content. The unit weight per handsheet and the number of handsheets for a given treatment combination could thus be decided depending on the number of plastic layers (i.e., 1, 2 or 3) required to comprise this treatment combination. Table 4.1 summarizes the basic properties of handsheets fabricated in such a manner.

Twenty-one to 97 handsheets (a total of 40 to 193 layers) were used to fabricate a laminate. The actual fiber content (ACTFC) of laminates for all treatments were approximately 2 to 4% lower than their nominal fiber contents, because the control over the consistency of pulp slurry during paper making was underestimated. This was due to the fixed-thickness nature of PP film which led to slight deviations from the exact nominal fiber content. However, if the target laminate is not too thin, the underestimation can be adjusted and the fixed-thickness becomes a minor problem. All of the actual fiber content for the three fiber content levels retained their difference level around 10%; therefore, the analyses based upon actual fiber content are considered valid.

Table 4.1. Basic handsheet properties for each treatment.

----- Treatment -----		Laminate	Hand	-----			Handsheet	-----		
NOMFC ¹	LAYER	ACTFC ¹ (% by wt)	sheet per laminate	Thickness	Grammage	Apparent density	Porosity	Pore size	Pore perimeter	Pore number
				---- mil ----	-- lb/10 ³ ft ² -	-- pcf --	---- % ----	10 ⁻⁸ inch ²	10 ⁻² inch	
50	1	46.11 (0.20)	97	3.5 (0.0)	3.22 (0.18)	11.07 (0.59)	19.23 (1.46)	3.18 (0.19)	0.70 (0.02)	865 (19)
50	2	46.79 (0.06)	49	5.9 (0.2)	6.89 (0.36)	14.04 (0.78)	10.10 (1.54)	1.04 (0.17)	0.38 (0.03)	1291 (229)
50	3	47.05 (0.09)	33	8.3 (0.2)	10.16 (0.47)	14.62 (0.72)	2.30 (0.51)	1.11 (0.25)	0.38 (0.03)	277 (102)
60	1	56.81 (0.16)	79	4.5 (0.1)	4.91 (0.16)	13.12 (0.32)	16.19 (0.80)	1.93 (0.23)	0.48 (0.03)	1080 (95)
60	2	58.31 (0.07)	40	8.4 (0.3)	10.59 (0.66)	15.07 (0.54)	3.15 (0.83)	0.85 (0.08)	0.32 (0.01)	456 (85)
60	3	57.63 (0.12)	27	12.1 (0.9)	14.44 (0.79)	14.39 (1.50)	0.45 (0.15)	0.60 (0.04)	0.27 (0.01)	95 (28)
70	1	66.85 (0.06)	61	7.0 (0.7)	7.76 (0.91)	13.27 (0.72)	5.91 (0.78)	1.17 (0.17)	0.37 (0.03)	647 (65)
70	2	68.81 (0.09)	31	13.7 (0.5)	16.38 (0.71)	14.30 (0.40)	0.30 (0.25)	0.58 (0.09)	0.27 (0.02)	61 (43)
70	3	67.25 (0.17)	21	19.7 (1.1)	23.30 (1.17)	14.22 (0.57)	0.02 (0.01)	0.48 (0.11)	0.24 (0.04)	7 (3)

¹ NOMFC=Nominal fiber content, ACTFC=Actual fiber content, SPGR=Specific gravity.² Numbers in the parentheses are standard deviations and sample size for every treatment is 5.

4.2.2.2. Wood Fiber-Polypropylene Laminate Manufacture

Handsheets were laid up in such a manner that they alternated with plastic films to make a laminate with two exterior surfaces placed with handsheets. In this study, the term plastic layer (LAYER) represents the number of PP films between two consecutive handsheets.

The preassembled wood fiber-polypropylene laminate was conditioned in an oven of $100\pm3^{\circ}\text{C}$. After conditioning, the mats were transferred immediately to a 5x5 -inch hot press with the platen temperature preheated to 400°F . Sufficient pressure (about 9 tons) was applied so that the platens closed to 1/8 -inch stops in approximately 15 seconds. However, on initial contact with the lay-up, the caul temperature dropped abruptly. When the caul temperature returned to 375°F (in approximately 2 minutes), the heater was turned off and the lay-up remained compressed until completion of hot-pressing. The total hot press time was 10 min. after closure. The molten stacks were solidified by turning on the water cooling system to bring the hot press to room temperature. The average hot pressing spectra for consolidating the laminates is shown in Figure 4.1. It shows that even though the caul temperature could not be automatically controlled, it remained between $350\text{--}400^{\circ}\text{F}$ during the entire hot pressing operation.

4.2.2.3. Measurement of Handsheet Porosity

To investigate the relationships between porosity of handsheets and strengths of resulting wood fiber-plastic laminates, an image analysis system

(Optimas 6.0) was employed. This system was interfaced with a camera which was attached to an optical microscope. For each treatment, 5 handsheets were sampled and cut into a dimension of 0.4(W)x2(L) - inch, dyed with 10% fast blue, mounted on slides, and examined microscopically. Five sampling points of 1.3 inch in width and 1 inch in height were measured within each sample handsheets. The threshold for microscopic resolution was 124 pixel. Data for pore area, perimeter, and number were obtained. Simple linear regression analysis was performed to determine the relationships between handsheet characteristics and laminate properties.

4.2.2.4. Tensile Strength Measurement

Two dog-bone tensile specimens of 5(L)x1.25(W)x0.125(T) -inch with neck 0.5 -inch were cut from each laminate replicate for tensile testing in accordance with ASTM D638-91(ASTM 1991). Modulus of rupture (ultimate strength, MOR), percentage elongation at break (ELONG), and modulus of elasticity (MOE) were analyzed using ANOVA and Tukey's test.

4.2.2.5. Dimensional Stability

One specimen of 5(L)x0.5(W)x0.125(T) -inch was sampled from each laminate and immersed in boiling distilled water for two hours. Water absorption (WA), linear expansion (LE) and thickness swelling (TS) were measured in accordance with ASTM D-1037-94 (ASTM 1994). ANOVA and Tukey's test were also performed to evaluate the effect of fiber content and layer.

4.2.2.6. Scanning Electronic Microscopy (SEM)

SEM samples were taken from the fracture surface of the tensile specimens. Samples were vacuum-dried at $60\pm 3^{\circ}\text{C}$ before coating, then mounted and analyzed on a scanning electronic microscope. The inner, outer, and fracture surfaces were investigated. The inner surface of a sample was obtained by peeling along the thickness (transverse) direction perpendicular to the axis of the tensile specimen to examine the exposed surface.

4.3. RESULTS AND DISCUSSION

4.3.1. Basic Handsheet Properties

The data on thickness and weight per unit area (grammage) of handsheets (Table 4.1) indicate that the greater the fiber content and layer, the greater the thickness and grammage. The relationship between thickness and grammage of handsheets (Figure 4.2) was linear. As high as 99% of the variability of handsheet thickness could be explained by grammage. On the other hand, the apparent density of a handsheet is related to its apparent thickness and grammage, since the value was calculated by dividing grammage with apparent thickness.

The fibers created a network of pores which is an important part of paper structure. In this study, porosity is the percentage of void and pore area per unit area, as measured with an image analysis system (Figures 4.3–4.5). These figures show the transition of porosity with changing NOMFC and LAYER. Theoretically, porosity can be calculated from apparent density of handsheet and

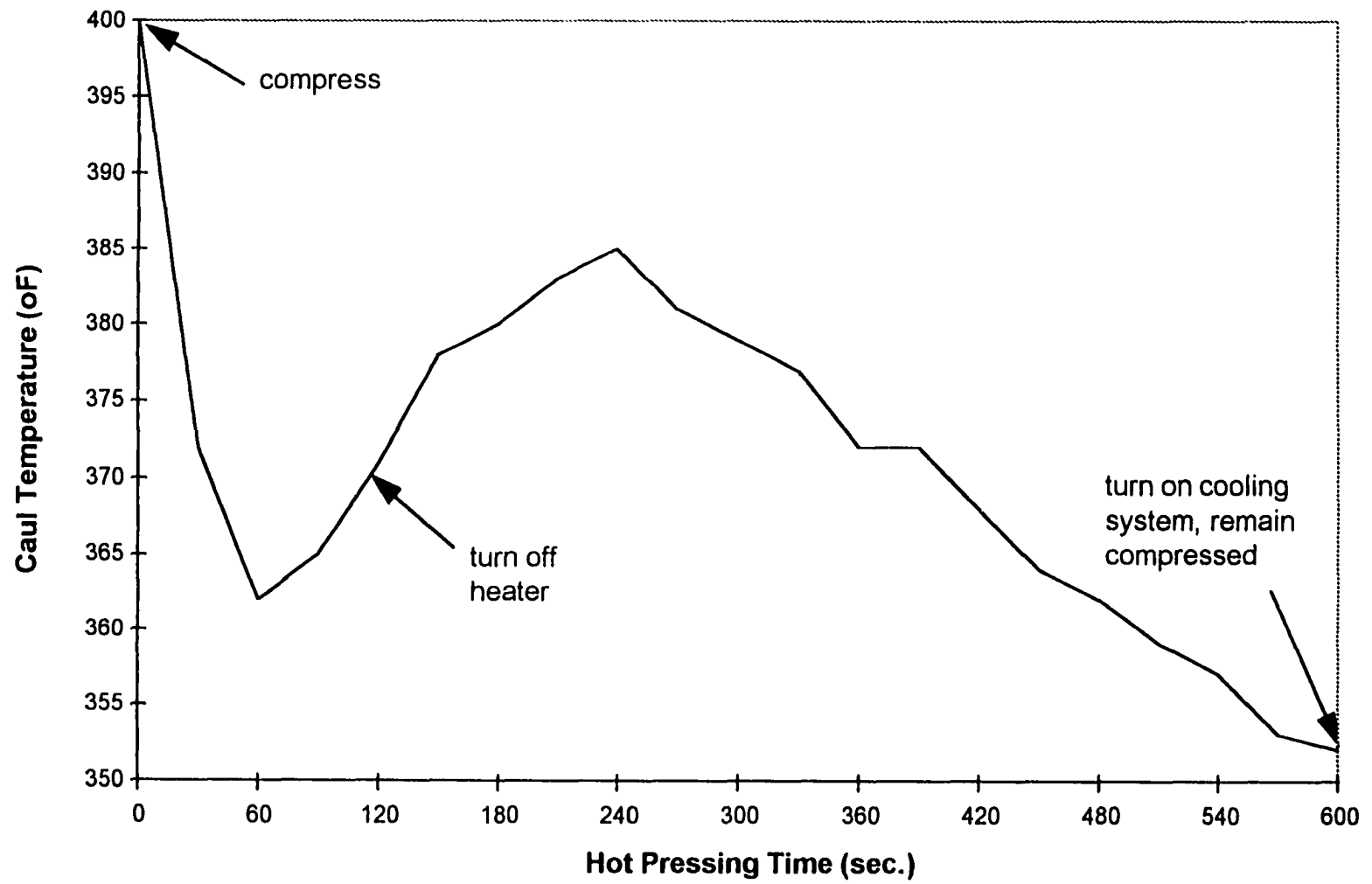


Figure 4.1. Typical hot pressing spectra in fabricating fiber-polypropylene laminates.

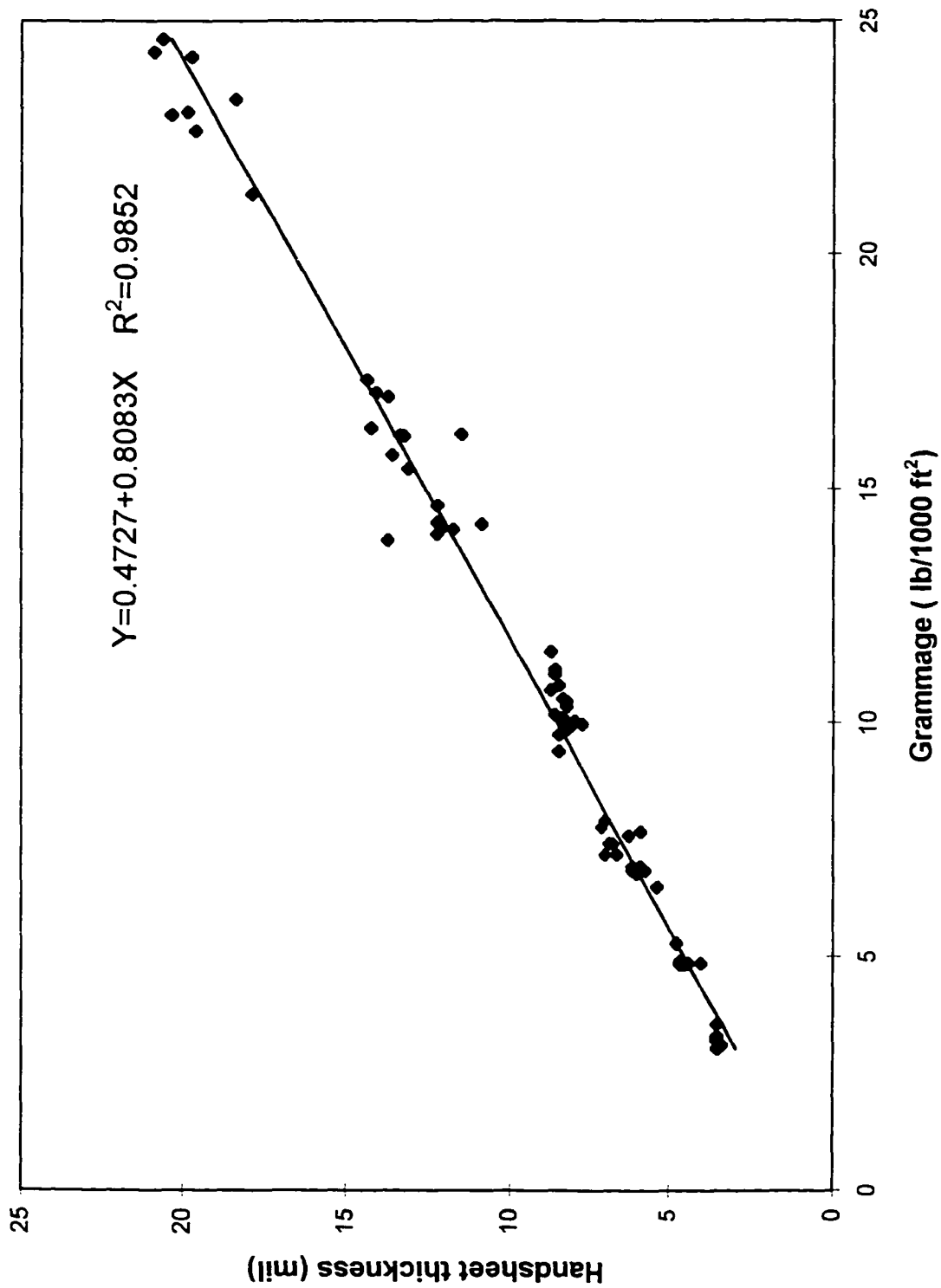
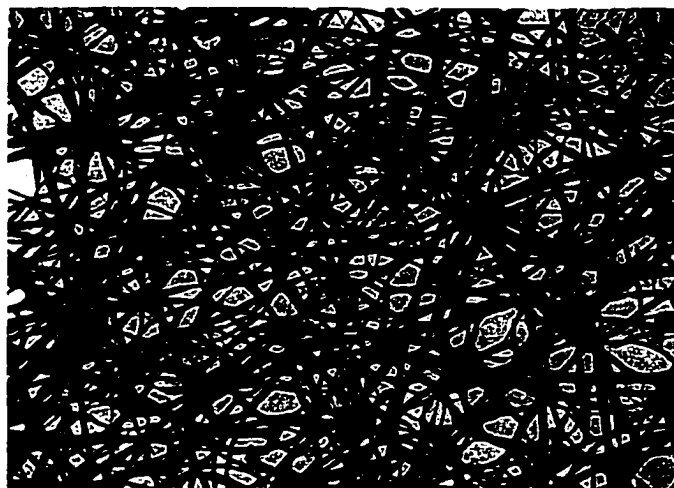
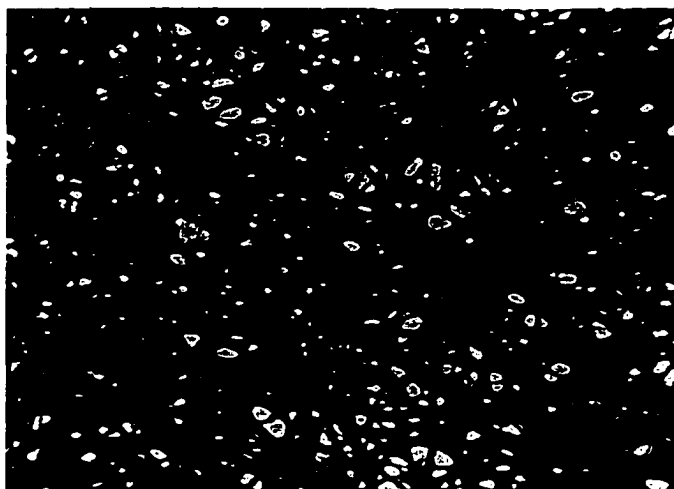


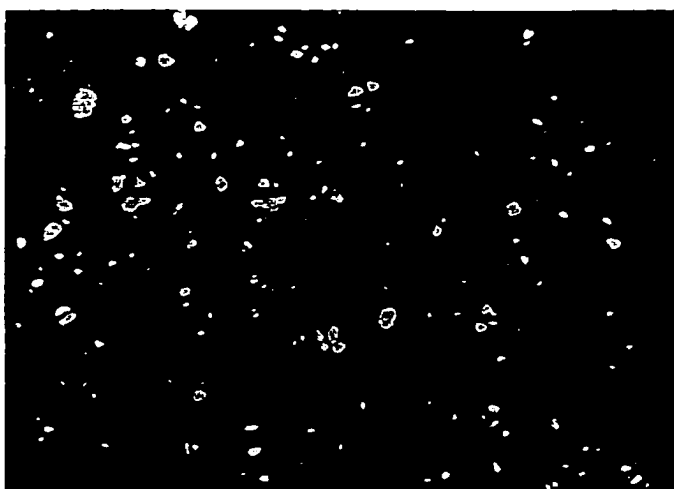
Figure 4.2. Dependence of handsheet thickness on grammage.



(A)

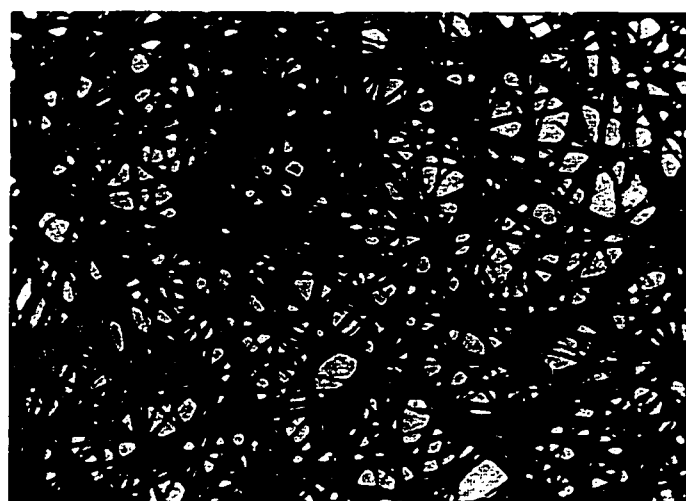


(B)

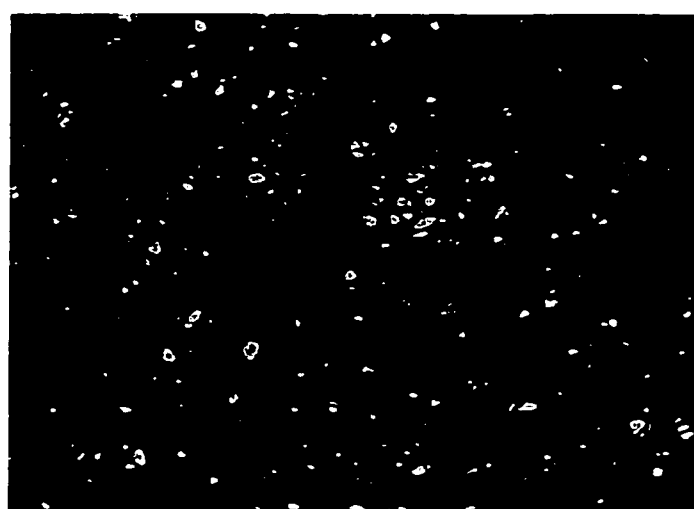


(C)

Figure 4.3. Micrograph of handsheet porosity for treatment with 50% fiber content. A: LAYER=1, B: LAYER=2, C: LAYER=3.



(A)



(B)



(C)

Figure 4.4. Micrograph of handsheet porosity for treatment with 60% fiber content. A: LAYER=1, B: LAYER=2, C: LAYER=3.

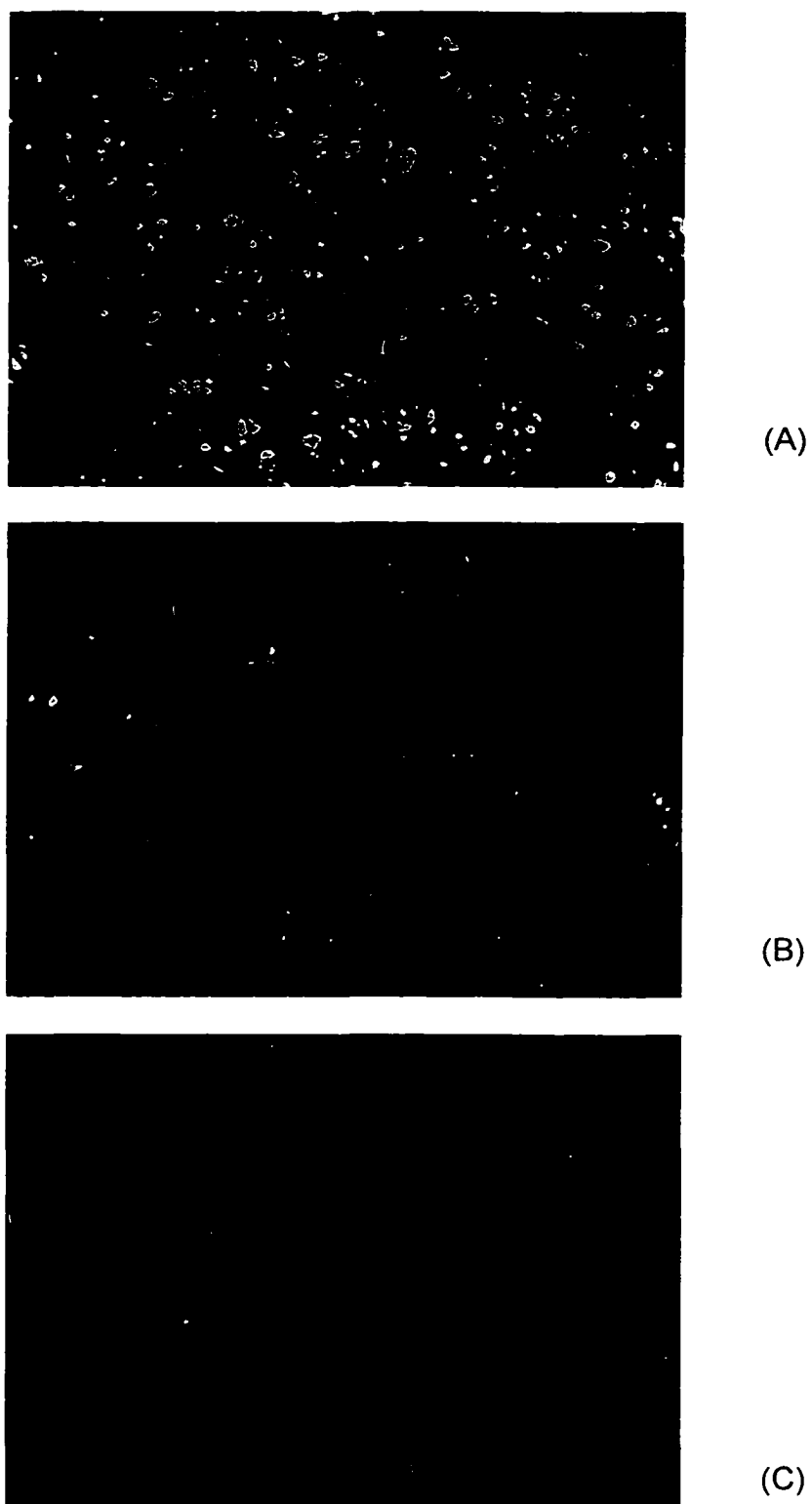


Figure 4.5. Micrograph of handsheet porosity for treatment with 70% fiber content. A: LAYER=1, B: LAYER=2, C: LAYER=3.

Table 4.2. Mechanical and physical properties of different fiber content (NOMFC) and layer (LAYER) combinations for wood fiber-polypropylene laminates with a target specific gravity of 1.00.

NOMFC	LAYER	MOR _t	ELONG	MOE _t	WA	LE	TS	SPGR
		- psi -	--%--	10 ³ psi		----- % -----		
50	1	6350 (400)	6.65 (0.44)	160 (53)	5.08 (0.54)	0.16 (0.07)	5.02 (0.44)	1.037 (0.015)
50	2	6064 (168)	6.62 (0.20)	188 (46)	6.81 (2.26)	0.31 (0.14)	6.32 (1.89)	1.028 (0.025)
50	3	6209 (58)	6.29 (0.72)	210 (23)	7.48 (0.88)	0.34 (0.16)	7.08 (0.70)	1.028 (0.007)
60	1	6559 (129)	10.15 (0.46)	216 (24)	11.60 (0.86)	0.95 (0.05)	10.96 (0.60)	1.002 (0.007)
60	2	6441 (93)	11.49 (0.82)	207 (15)	14.57 (0.55)	1.20 (0.16)	12.70 (0.39)	1.019 (0.010)
60	3	6129 (445)	10.60 (1.52)	198 (33)	14.85 (0.65)	1.24 (0.06)	12.89 (0.38)	1.009 (0.010)
70	1	6351 (66)	7.97 (0.53)	259 (29)	18.78 (0.60)	1.04 (0.05)	18.42 (0.39)	0.995 (0.003)
70	2	6548 (102)	11.61 (0.20)	177 (55)	26.30 (0.82)	1.27 (0.18)	21.12 (0.34)	0.997 (0.002)
70	3	6200 (278)	11.97 (0.90)	154 (39)	24.47 (1.86)	1.39 (0.09)	19.66 (0.12)	0.995 (0.010)

1. NOMFC = nominal fiber content. LAYER = Plastic layers per laminae.
MOR_t = Tensile modulus of rupture. ELONG = Elongation at break.
MOE_t = Tensile modulus of elasticity. WA = Water absorption.
TS = Thickness swelling. LE = Linear expansion.
SPGR = Specific gravity.
2. Numbers in the parentheses are standard deviations
3. Sample size for every treatment is 3.

density of the solid phase material (Bristow 1986); therefore, porosity should be closely related to apparent density of handsheet. However, as shown in Table 4.1, such a relationship did not exist.

A loglinear model $\ln(Y)=b_0+b_1\ln(X)$ was employed to estimate the relationship between porosity and grammage. Such a model could explain 65% of the variabilities of porosity. It fit well at the higher grammage region but not lower grammage region. This, presumably, is due to the lack of observations in the low-density region. Table 4.1 also shows great variability in pore sizes, pore perimeters and pore numbers for handsheets in all treatment combinations. The variability was largely due to insufficient sample size, the grammage-localization characteristics of handsheets, and limitation of 2-D image analysis system.

4.3.2. Mechanical and Physical Properties of Wood Fiber-Polypropylene Laminates

The average mechanical and physical properties of wood fiber-polypropylene laminates with a target specific gravity of 1.0 are shown in Table 4.2. The specific gravity for all treatment combinations slightly deviated from the target; however, treatments with low NOMFC seemed to overestimate specific gravity, while treatments with high NOMFC seemed to underestimate this parameter. The average specific gravities for 50, 60, and 70% fiber content were 1.03, 1.01, and 1.00 respectively. With regard to tensile properties, the overall range for average MOR_t was between 6,000 to 6,600 psi (41.4 to 45.5 Mpa), and the coefficients of variation (CV) were between 1 to 8%, indicating that manipulating NOMFC and LAYER may have some effects on MOR_t . However,

the average MOE_t values for all treatment combinations were between 150,000 to 260,000 psi (1,034 to 1,793 Mpa), with CV level as high as 33%. This implies that high variability may hinder the effect of NOMFC and LAYER on MOE_t . On the other hand, the values estimated for elongation at break (ELONG) for all treatment combinations were in a broad range of 6 to 12 %. Treatments with NOMFC=50 had lower elongation at the break, suggesting that the effect of NOMFC and LAYER might have a significant effect on elongation. The thickness has a profound restriction on composites made from paper. This study shows that up to 1/8 -inch thick wood fiber-polypropylene laminates could be made to achieve comparable (Takase and Shirishi 1989) or better (Myers *et al.* 1993) ultimate tensile strength (MOR_t) than those of wood fiber-reinforced polypropylene composites reported previously.

In dimensional stability, Table 4.2 shows that there was an increasing trend in the average WA, LE, TS values with increasing NOMFC and LAYER. Further statistical analysis (Table 4.3) clarified these trends.

4.3.2.1. Tensile Properties

4.3.2.1.1. Modulus of Rupture (MOR_t)

Table 4.3 shows that the main effect of LAYER on MOR_t was significant. The relationship of LAYER main effect on the MOR_t ($p=0.036$) indicates that MOR_t declined as LAYER increased (Figure 4.6). That is, with NOMFC effect pooled, laminates made from interleaving one layer of PP film with one layer of handsheet exhibited the best mean MOR_t , which was about 4% higher than

LAYER=3. At a given NOMFC, the fewer the LAYER the thinner the constitutive handsheet. As a result of thinner handsheets, the apparent contact area between PP and fibrous network increased, which was a good indication of better interlocking. Moreover, the thinner the handsheet, the more easily the molten plastic could penetrate through the voids between interconnecting fibers and fuse together with the adjacent plastic films, forming a continuous matrix with lesser voids. This matrix, upon consolidation, exhibited a strong mechanical anchor, thus enhancing the contact between wood fiber and polypropylene matrix. LAYER 1 had better MOR_t than LAYER 2 and 3, due to the improvement in specific contact area and mechanical anchor.

The NOMFC*LAYER interaction ($p=0.0914$) and NOMFC main effect ($p=0.1245$), shown in Table 4.3, were not significant at $\alpha=0.05$ level for MOR_t 's of all treatment combinations. Research on wood fiber-reinforced plastic (Maldas *et al.* 1988, Raj *et al.* 1989, Raj *et al.* 1990) had indicated a decreasing trend of MOR_t with increasing fiber loading when fiber loading was less than 50%. In this study, the main effect of NOMFC was not significant, probably due to decrease in MOR_t may level off when fiber loading exceeds a certain extent.

4.3.2.1.2. Elongation at Break (ELONG)

The ANOVA results on ELONG indicate that NOMFC*LAYER interaction was highly significant ($p=0.0006$). The interaction between NOMFC and LAYER on the ELONG of wood fiber-PP laminates is shown in Figure 4.7. The LAYER effect was less profound at two lower NOMFC levels (50 and 60), and

NOMFC=70 was most sensitive to changes in LAYER. ELONGs of NOMFC=60 were greater than those of NOMFC=50 for all LAYER levels. Therefore, the NOMFC*LAYER interaction was primarily derived from the inconsistent trend in NOMFC=70. In wood fiber-plastic laminate, the interfiber hydrogen bond remains intact; but without additives, the adhesion between fiber and plastic is mostly by mechanical anchor. The effect of mechanical anchor decreases as fiber content increases, resulting in higher viscoelastic deformation. These might be the reasons that NOMFC=60 showed overall higher ELONG than NOMFC=50. Compared to those of NOMFC=60, NOMFC=70 had lower ELONG at lower LAYER level, approximately the same ELONG at intermediate LAYER level, and higher ELONG at higher LAYER level. The improvement of ELONG at lower LAYER level for NOMFC=70 was probably due to the moderate pore size and pore distribution (Figure 4.5 (A)) which enhanced the interpenetration of fiber network and polypropylene, and the appropriate handsheet thickness which facilitated paper strength.

4.3.2.1.3. Modulus of Elasticity (MOE_t)

MOE_ts for all treatment combinations exhibited more complicated relationships. Table 4.3 shows that only NOMFC*LAYER interaction was highly significant ($p=0.0014$), whereas NOMFC and LAYER main effects were not significant at $\alpha=0.05$. As shown in Figure 4.9, MOE_t of NOMFC=60 remained relatively constant at all LAYER levels, but MOE_t of NOMFC=50 and =70 showed reverse trends toward LAYER level.

Fiber content had positive effect on MOE_t in wood fiber-polypropylene laminates, if the interfacial adhesion was adequate. At LAYER=1, the handsheet thickness was minimal; therefore, PP films and handsheets could achieve good interfacial contact. However, in NOMFC=50, handsheets were so thin that more air was trapped in the laminate, thus inhibiting the stress transfer in the matrix. Moreover, due to insufficient thickness and large pores (Figure 4.3(A)), the handsheets failed to facilitate appropriated enforcement to the laminate. Hence at LAYER=1, MOE_t increased with fiber content. The positive effects of fiber content on MOE_t of fiber-reinforced thermoplastics have also been shown in polystyrene (Maldas *et al.* 1988), high density polyethylene (Raj *et al.* 1990) and linear low density polyethylene (Raj *et al.* 1989).

On the other hand, at LAYER=3 where handsheet thickness was maximal and pores were rare (Figure 4.5(C)), interfacial adhesion became a problem for higher NOMFC level. Therefore, NOMFC=70 displayed the lowest MOE_t , whereas NOMFC=50 displayed the highest value. As a result, completely different trends of MOE_t s coexist at two extreme levels of LAYER and subsequently offset the NOMFC and LAYER main effects.

The above results imply that, with appropriate manipulation of LAYER, wood fiber-polypropylene laminates with as high as 70% NOMFC displayed high MOR_t , high MOE_t , and moderate ELONG. Since these laminates were made from untreated, uncalendered and unoriented handsheets, their performances can be improved with optimized processing.

Table 4.3. Analysis of variance of fiber content (NOMFC) and layer (LAYER) effects on mechanical and physical properties of wood fiber-plastic laminates.

Source	DF	MS	F	P
----- MOR _t -----				
NOMFC	2	80486.37	2.34	0.1245
LAYER	2	138088.93	4.02	0.0360
NOMFC*LAYER	4	81289.04	2.37	0.0914
ERROR	18	34336.30	----	----
----- ELONG -----				
NOMFC	2	50.7542	89.91	0.0001
LAYER	2	6.9922	12.39	0.0004
NOMFC*LAYER	4	4.6079	8.16	0.0006
ERROR	18	0.5645	----	----
----- MOE _t -----				
NOMFC	2	976667753	1.43	0.2659
LAYER	2	1604765210	2.34	0.1245
NOMFC*LAYER	4	4805000286	7.02	0.0014
ERROR	18	684471165	----	----
----- WA -----				
NOMFC	2	633.44	470.16	0.0001
LAYER	2	46.38	34.42	0.0001
NOMFC*LAYER	4	7.00	5.20	0.0058
ERROR	18	1.35	----	----
----- LE -----				
NOMFC	2	2.5068	185.94	0.0001
LAYER	2	0.1863	13.82	0.0002
NOMFC*LAYER	4	0.0064	0.47	0.7549
ERROR	18	0.0135	----	----
----- TS -----				
NOMFC	2	417.33	722.96	0.0001
LAYER	2	10.12	17.53	0.0001
NOMFC*LAYER	4	1.02	1.77	0.1793
ERROR	18	0.58	----	----

NOMFC = nominal fiber content.

MOR_t = Tensile modulus of rupture.

MOE_t = Tensile modulus of elasticity.

TS = Thickness swelling.

SPGR = Specific gravity.

LAYER = Plastic layers per laminae.

ELONG = Elongation at break.

WA = Water absorption.

LE = Linear expansion.

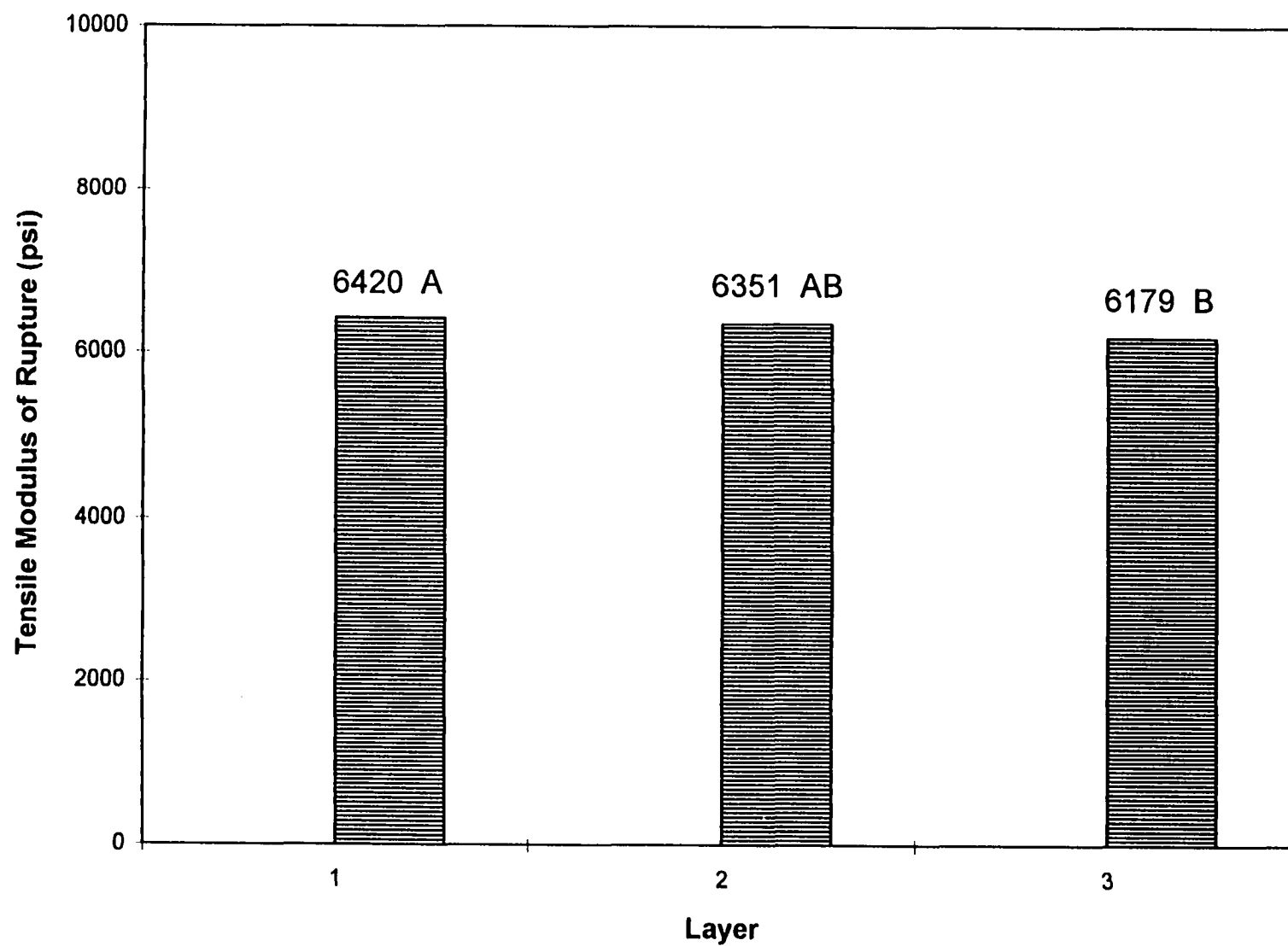


Figure 4.6. Effect of plastic layer on tensile strength of fiber-polypropylene laminates.

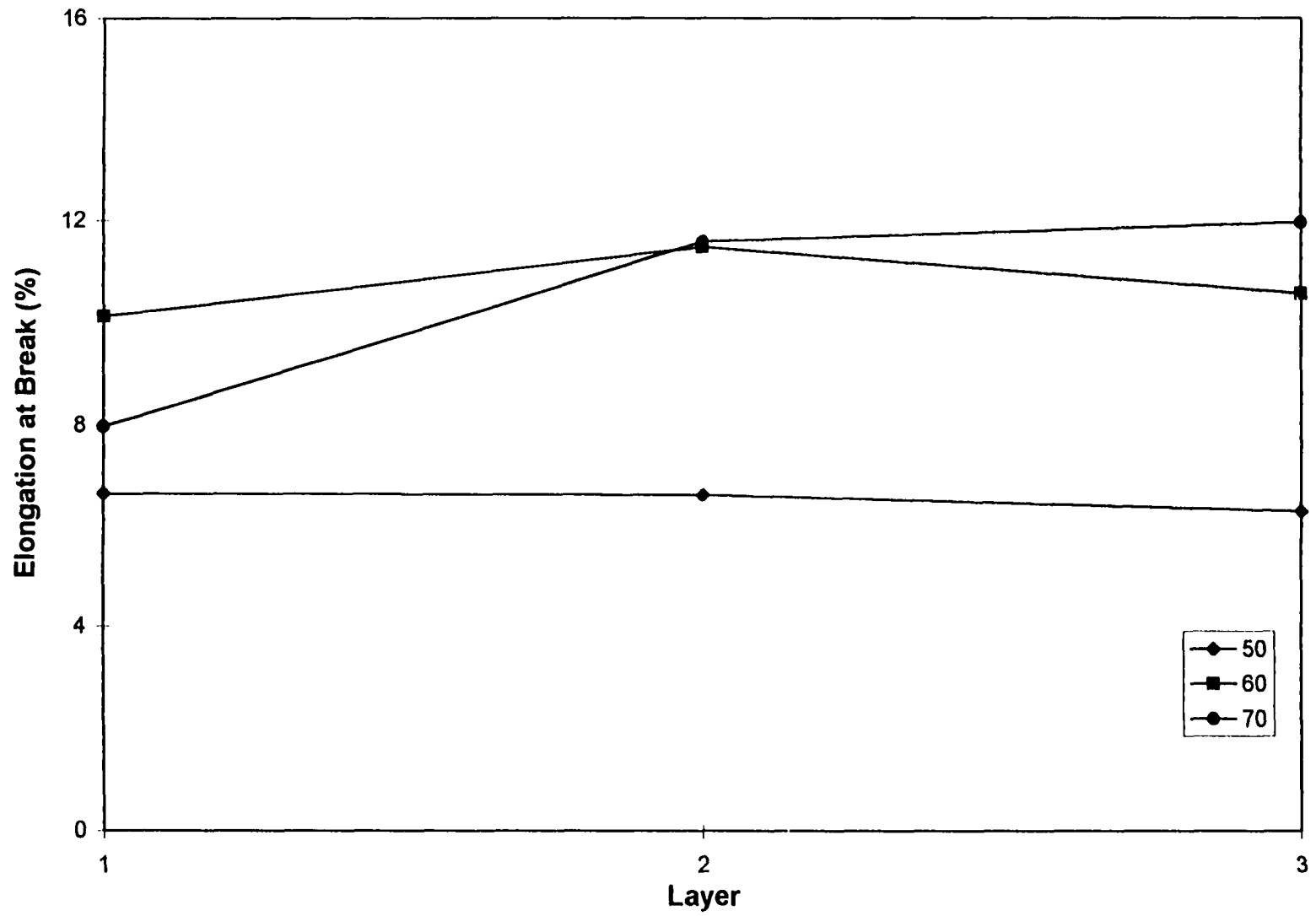


Figure 4.7. Interaction of fiber content and plastic layer on elongation of fiber-pp laminates.

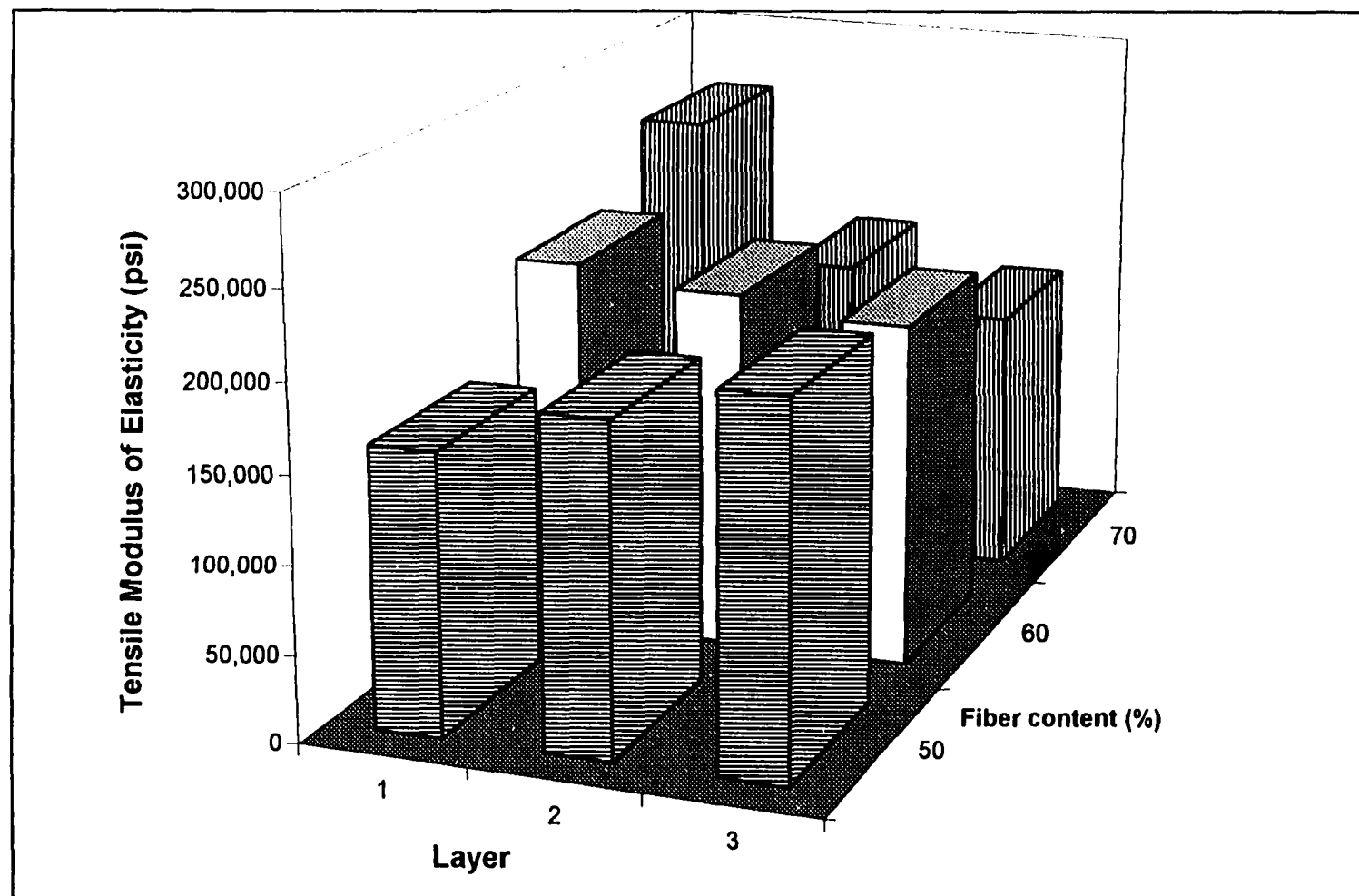


Figure 4.8. Interaction of fiber content and plastic layer on tensile modulus of fiber-polypropylene laminates.

4.3.2.2. Dimensional Stability

4.3.2.2.1. Water Absorption (WA)

The ANOVA results (Table 4.3) show the significant effects of NOMFC ($p=0.0001$) and LAYER ($p=0.0001$). The NOMFC*LAYER interaction were also highly significant ($p=0.0058$) for WA. When all LAYER levels were pooled, WA increased with increase in NOMFC (Figure 4.9). The WAs in NOMFC=70 and NOMFC=60 were 3.5 and 2.1 times, respectively, those of NOMFC=50, indicating that more hydrophilic sorption sites were contained in wood fiber-polypropylene laminate surface for the higher NOMFC levels.

The LAYER main effect (Figure 4.10) shows that, at pooled NOMFC level, LAYER=1 revealed the lowest WA; LAYER 2 and 3 had WA about 1.3 times that of LAYER 1. This is presumably due to the increased contact area and lower grammage of handsheets. These allowed PP films to penetrate into the voids and pores of handsheets, such that the hydrophobic PP effectively blocked the sorption sites on the fiber surfaces, resulting in a lower WA.

The NOMFC*LAYER interaction (Figure 4.11) shows that as LAYER increased, WA of wood fiber-PP laminates gradually increased both at NOMFC=50 and NOMFC=60 levels. The trends were very consistent for these two NOMFC levels since they were parallel to each other. However, at NOMFC=70, WA firstly increased from LAYER=1 to LAYER=2, then slightly declined at LAYER=3. The inconsistency that caused significant interaction might be due to the random chance effect.

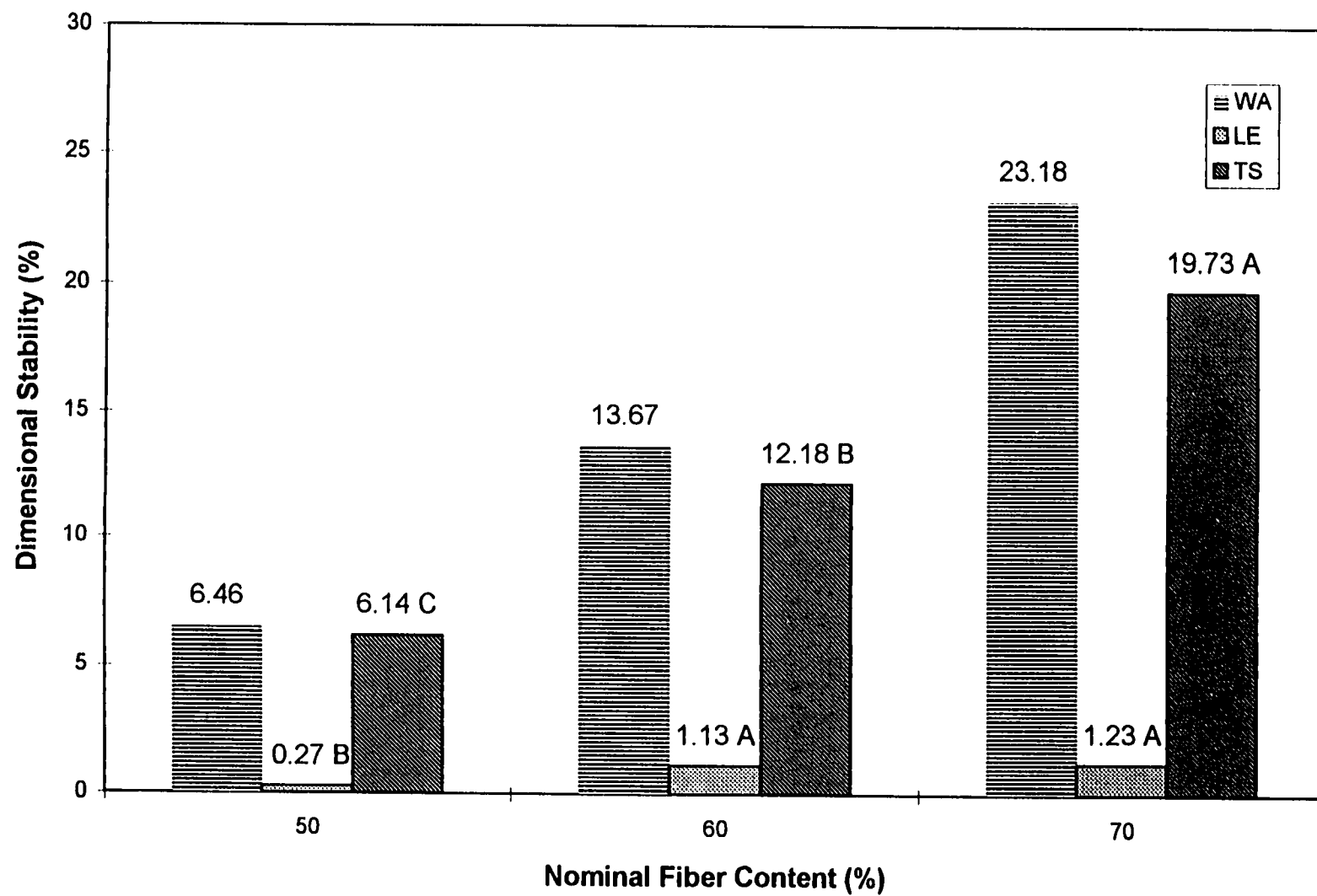


Figure 4.9. Effect of fiber content on dimensional stability of fiber-polypropylene laminates.

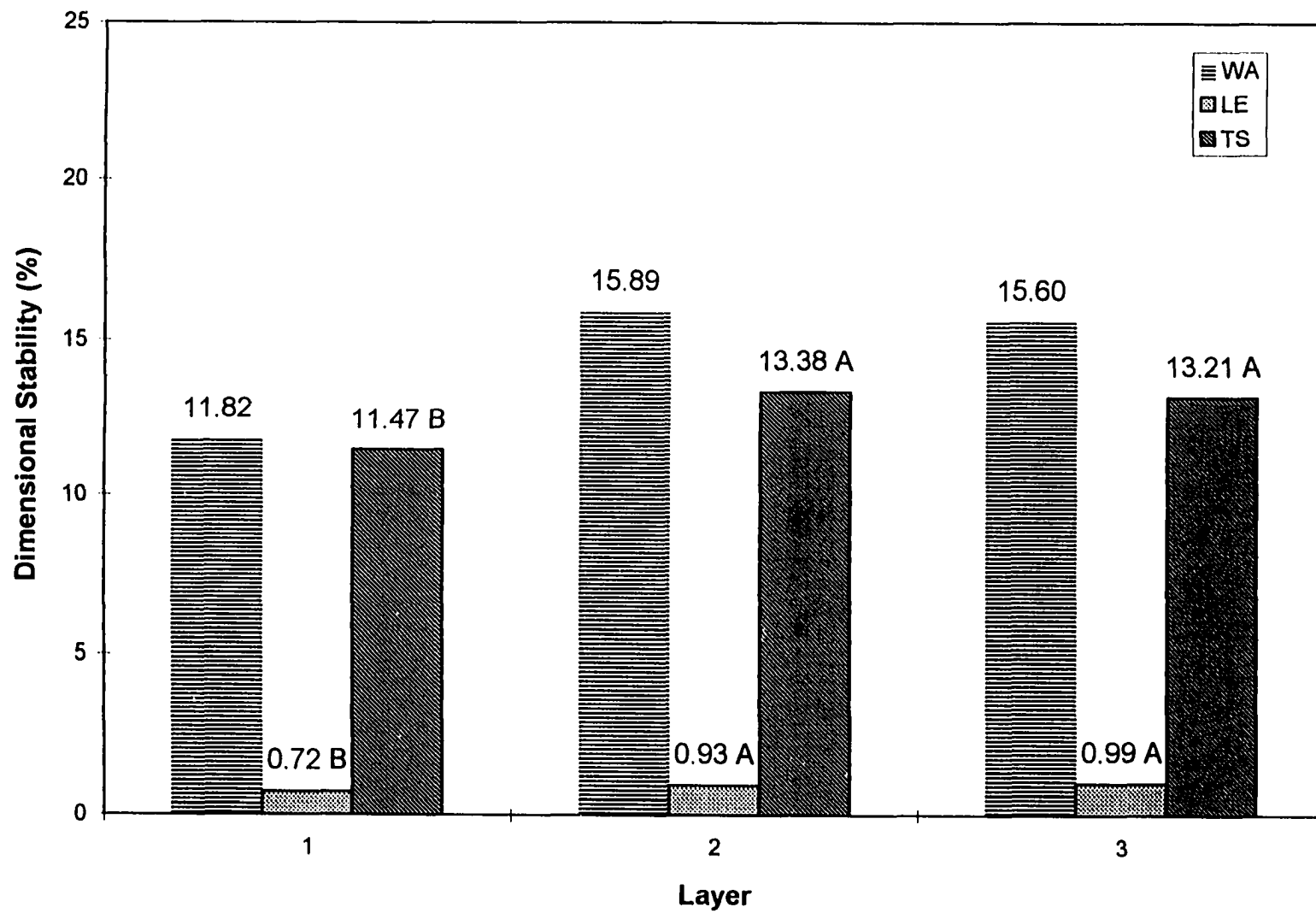


Figure 4.10. Effect of plastic layer on dimensional stability of fiber-polypropylene laminates.

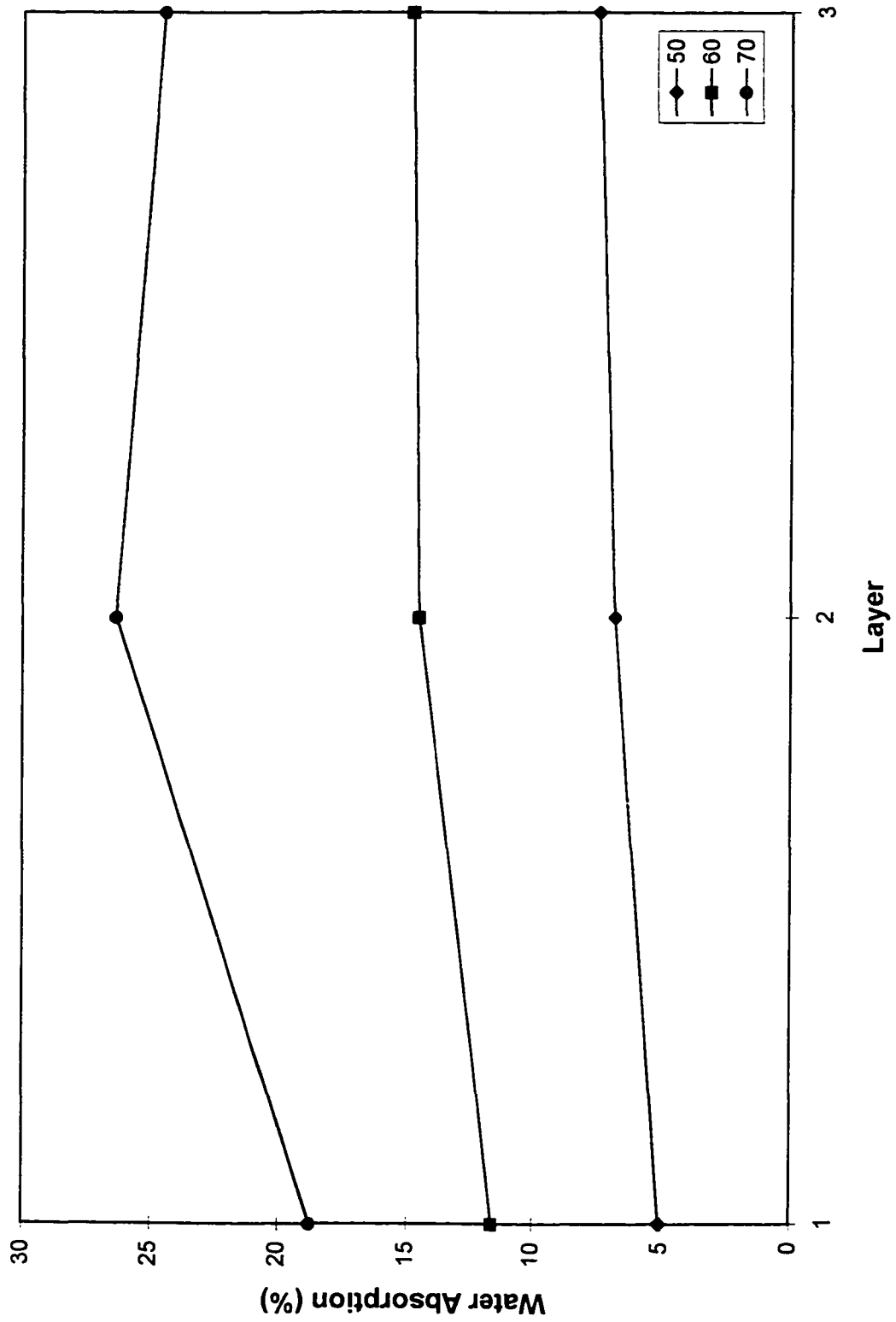


Figure 4.11. Interaction of fiber content and plastic layer on water absorption of fiber-pp laminates.

According to Michell *et al.* (1978) and Campbell *et al.* (1980), the rates of water absorption were lower for wood fiber-polyethylene laminates with plastic films placed on the two outer surfaces than those laminates with handsheets on the two external faces. However, the mechanical properties fell eventually to the same level for those two types of geometry. The WA test in this study was performed two hours after immersion in boiling water. Since all the fiber-polypropylene laminates had handsheets at external faces, the WA rate would be higher and the total water uptake would be close to saturation.

4.3.2.2.2. Linear Expansion (LE)

In LE, the main effects of NOMFC ($P=0.0001$) and LAYER ($p=0.0002$) were highly significant, whereas the NOMFC*LAYER interaction was not significant ($p=0.7549$) (Table 4.3). As shown in Figure 4.9, NOMFC=50 had significantly lower LE than the other two levels of NOMFC, as a result of Tukey's test. Although NOMFC=70 had substantially higher water uptake, the LE appeared to be less sensitive to the increase in NOMFC as fiber content reached a certain level. For wood fiber-polypropylene laminates with NOMFC=50, the high PP content and low grammage accounted for the better coverage of fiber surfaces by plastic, resulting in lower longitudinal hygroexpansion upon immersion in boiling water. As NOMFC increases, such hygroexpansion of fibers also increases. However, further increment in NOMFC would not significantly affect LE, since the hygroexpansion was restricted by the adjacent hydrophobic plastic films. The same phenomenon in LAYER main effect are presented in

Figure 4.10. From Tukey's test, LAYER showed a similar trend as NOMFC, due to similar mechanism as described previously. At lower LAYER level, PP could easily interpenetrate into handsheets, thus blocking their sorption sites. As LAYER increased, the handsheet grammage correspondingly increased, causing less effective interpenetration of PP films to handsheets, which subsequently led to higher longitudinal hygroexpansion. However, further increase in LAYER inhibited hygroexpansion by the bulk of PP films, hence LE remained stable.

2.3.2.2.3. Thickness Swelling (TS)

In TS, the ANOVA (Table 4.3) reveals significant NOMFC ($p=0.0001$) and LAYER ($p=0.0001$) main effects. As shown in Figure 4.9, the trend was analogous to that of WA, i.e., NOMFC=70 and NOMFC=60 had TS 3.2 and 2.0 times, respectively, that of NOMFC=50. This indicates that the transverse, but not the longitudinal, hygroexpansion of handsheet could freely develop without restriction from adjacent PP films. Again, the effect of LAYER on TS was also analogous to that on WA, i.e., LAYER 1 was lower than LAYER 2 and 3; LAYER 2 and 3 had TS about 1.2 times that of LAYER 1. Therefore TS responded more readily than LE to WA, and water uptake had less effect on LE than on TS.

Since the measurements of WA, TS and LE were all obtained from the same specimens, the values of WA, TS and LE were highly correlated. Because TS and LE all had clear-cut NOMFC and LAYER main effects without the interference of NOMFC*LAYER interaction, it was speculated that the NOMFC*LAYER on WA was due to random chance effect.

The hydrophilic nature of wood fiber has always been a problem in dimensional stability. The handsheets fabricated were of a random network, that is, an isotropic structure. However, from the results of dimensional stability, wood fiber-polypropylene laminates were anisotropic, which displayed highly dissimilar TS and LE responses toward water uptake. Since these handsheets received no surface treatment, the dimensional stability could be improved.

4.3.2.3. Factors Affecting Laminate Properties

The effect of porosity of handsheet on mechanical properties (MOR_t , $ELONG$, MOE_t) of wood fiber-polypropylene laminate was investigated. There was no significant correlation between porosity and MOR_t , nor between porosity and MOE_t . However, correlation between $ELONG$ and porosity was significant ($p=0.01$). The relationships between hygroscopic properties of wood fiber-polypropylene laminate and their respective specific gravity ($SPGR$) and actual fiber content ($ACTFC$) were also investigated. A loglinear model denoted by $\ln(Y)=b_0+b_1\ln(X)$ was used and the estimated statistics are presented in Table 4.4.

Even though MOR_t and MOE_t did not show significant relationship with specific gravity and $ACTFC$ when all plastic layers were pooled, $ELONG$ exhibited declining trends with specific gravity and increasing trends with $ACTFC$. Approximately 34% and 59% of the variation in $ELONG$ could be explained by specific gravity and $ACTFC$. The dimensional stability of the laminate also showed a similar trend; however, WA , LE and TS had higher dependency than

ELONG on specific gravity and ACTFC. About 67 to 73% of the variation in dimensional stability could be explained by specific gravity, whereas ACTFC accounted for much more variability in WA ($R^2=0.92$) and TS ($R^2=0.94$) than in LE ($R^2=0.73$). This indicates that dimensional stability was predominately affected by fiber content, while some other factors besides specific gravity and ACTFC may also have influence on mechanical properties of wood fiber-polypropylene laminates.

Table 4.4. Results of nonlinear regression analysis of physical and mechanical properties (Y) with respect to independent variables of specific gravity (SPGR) and actual fiber content (ACTFC) using model $\ln(Y)=b_0+b_1\ln(X)$.

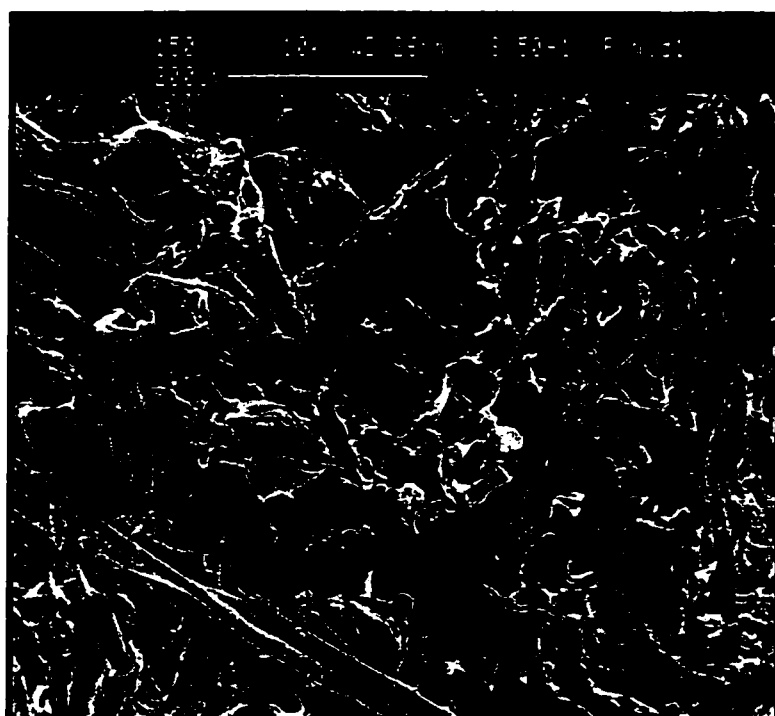
Dependent variable (Y)	Statistics			Partial coefficient	
	F	P	R^2	b_0	b_1
	X = SPGR				
ELONG	13.077	0.0013	0.3434	2.2968	-8.6734
WA	63.065	0.0001	0.7161	2.8487	-26.9674
LE	52.035	0.0001	0.6755	0.0853	-38.3009
TS	69.585	0.0001	0.7357	2.7188	-24.6607
	X = ACTFC				
ELONG	36.604	0.0001	0.5949	-3.1393	1.3209
WA	285.488	0.0001	0.9195	-11.7569	3.5382
LE	65.969	0.0001	0.7252	-18.9224	4.5950
TS	390.859	0.0001	0.9399	-10.6048	3.2274
SPGR	43.376	0.0001	0.6344	0.3843	-0.0922

¹. ELONG = Elongation at break.
TS = Thickness swelling.

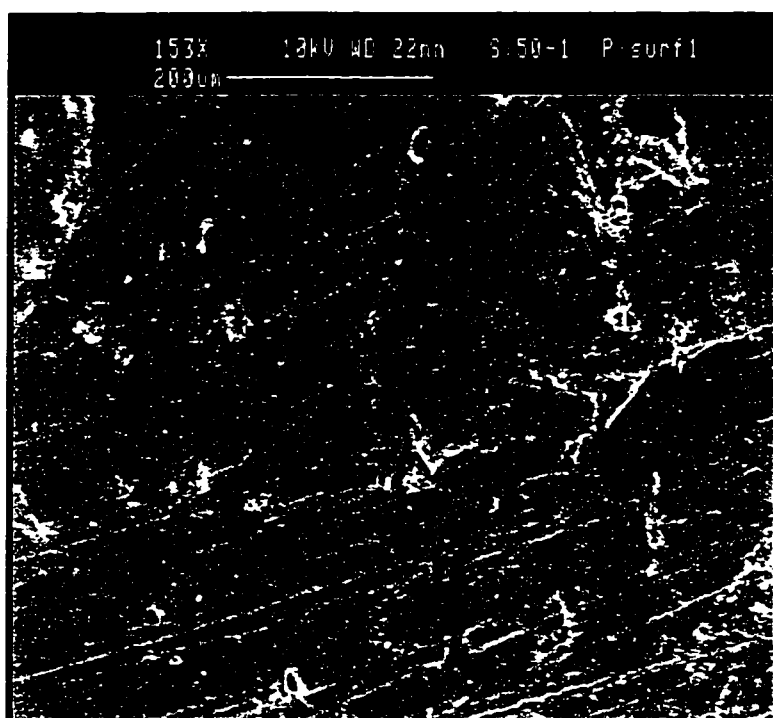
WA = Water absorption.
LE = Linear expansion.



Figure 4.12. SEM micrograph of tensile fracture for fiber-polypropylene laminate.

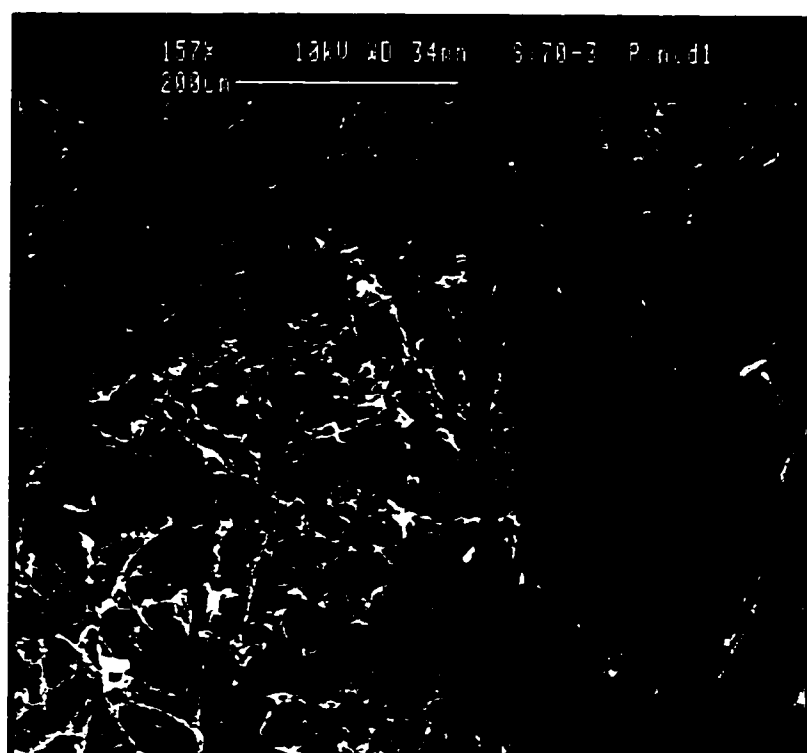


(A)

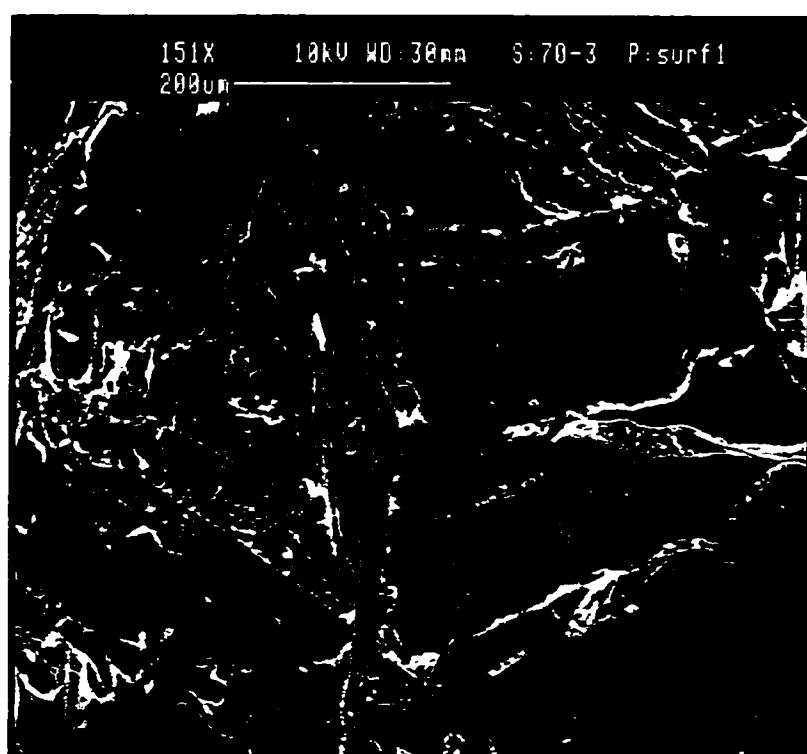


(B)

Figure 4.13. SEM micrograph of inner (A) and outer (B) surfaces for treatment with NOMFC=50 and LAYER=1.



(A)



(B)

Figure 4.14. SEM micrograph of inner (A) and outer (B) surfaces for treatment with NOMFC=70 and LAYER=3.

4.3.3. SEM Observation of Wood Fiber-Polypropylene Laminates

The failure modes of a composite material can be divided into four categories: fiber failure, microcracking of the matrix, separation of fibers from the matrix (debonding) and delamination; and these failure modes can happen separately or jointly (Agarwal and Broutman 1990). The typical fracture surface of wood fiber-polypropylene laminate under longitudinal tensile load was predominately fiber breakage with minor fiber pullout (Figure 4.12). This was similar to the failure mode of unidirectional composites with an intermediate fiber volume fraction (Agarwal and Broutman 1990). SEM micrographs (Figures 4.13 - 4.14) demonstrated the contrast between treatments 50-1 (NOMFC=50 and LAYER=1) and 70-3 (NOMFC=70 and LAYER=3) in the inner layer and surface. These two treatments were the most extreme cases in this study. Treatment 50-1 had the lowest fiber content and handsheet grammage, while treatment 70-3 was the opposite. Figure 4.13(A) shows the interpenetration of PP films to the fiber network, showing the effect of the mechanical anchor. The constitutive handsheets were so thin in treatment 50-1 that, during hot pressing, the molten PP could migrate to the outer surfaces and cover the fiber network (Figure 4.13(B)). For treatment 70-3, interpenetration also took place in the middle layer, but to a lesser extent, presumably due to less plastic content and difficulty of penetration through the thicker handsheets (Figure 4.14(A)). Treatment 70-3 showed a lesser coverage of PP on the surface of fiber network (Figure 4.14(B)). The voids appeared outside and inside the laminates, indicating a potential

problem in stacking numerous handsheets and PP films, which might inhibit stress transfer and cause lower mechanical properties of the resulting laminate. A vacuum procedure is suggested in the future study to remove air bubbles inside the films and fiber network before hot pressing.

4.4. CONCLUSIONS

Wet-formed Kraft pulp handsheets were interleaved with polypropylene films to fabricate laminates of 1/8-inch thick, with comparable ultimate tensile strengths to those of natural fiber-reinforced plastics. Effects of NOMFC and LAYER on tensile properties and dimensional stability of laminates were also investigated.

Experimental results show that laminates of LAYER 1 had better MOR_t than those of LAYER 3, due to better interpenetration of plastic, as evidenced in SEM observation. The effects of NOMFC and LAYER on ELONG were not consistent at two higher NOMFC levels. However, NOMFC=50 showed lowest ELONG at all LAYER levels. Interaction between NOMFC and LAYER compensated the main effects on MOE_t . At LAYER=1, more permeable but weaker and thinner handsheets led to the increase of MOE_t with NOMFC. At LAYER=3, stronger and thicker but less permeable handsheets resulted in the reverse relationship in MOE_t with NOMFC. With proper control of LAYER, wood fiber-polypropylene laminates with fiber loading as high as 70% could be fabricated with favorable mechanical properties.

Dimensional stability of wood fiber-polypropylene laminates were appreciably affected by both NOMFC and LAYER. An increase in NOMFC and LAYER led to an increase in WA, LE and TS. WA and TS showed a similar trend, but LE was less sensitive to changes in NOMFC and LAYER. The difference in magnitude between TS and LE indicated that the anisotropic characteristic of wood fiber-polypropylene laminate should be taken into consideration. Without surface treatment, lower NOMFC and LAYER showed best dimensional stability.

SEM micrographs showed that the major failure mode of wood fiber-polypropylene laminates was fiber breakage. Voids existed inside and on the surface of the laminates; therefore, precautions should be taken to avoid the enclosure of air bubbles and the excess shrinkage of PP in order to improve the strength properties.

Untreated wood fiber-polypropylene laminates showed promising features. With improved processing, wood fiber-plastic laminates can serve as industrial load-bearing materials.

4.5. LITERATURE CITED

Agarwal, B.D. and L.J. Broutman. 1990. Analysis and performance of fiber composites. 2nd ed. John Wiley and Sons, Inc., New York. pp. 87-92.

American Society for Testing and Materials. 1991. Standard test method for tensile properties of plastics. ASTM D 638-91. Philadelphia, PA.

American Society for Testing and Materials. 1994. Standard testing methods of evaluating the properties of wood-based fiber and particle panel materials. ASTM D 1037-94. Philadelphia, PA.

Bristow, J.A. 1986. The paper surface in relation to the network. In: J.A. Bristow and P. Kolseth (ed.), *Paper structure and properties*. Marcel Dekker, Inc. New York. pp. 169-182.

Campbell, M.D., R.S.P. Coutts, A.J. Michell, and D. Wallis. 1980. Composites of cellulosic fibers with polyolefins or cement. A short review. *Ind. Eng. Chem.* 19:596-601.

Caulfield, D.F., J.A. Koutsky, and D.T. Quillen. 1993. Cellulose/polypropylene composites: the use of AKD and ASA sizes as compatibilizers. In: M.P. Wolcott (ed.), *Wood-fiber/polymer composites: Fundamental concepts, processes, and material options*. Proc. 1st Wood Fiber-Plastic Composite Conference, Madison, WI. Madison, WI. Forest Products Society, 1993: 128-134.

Cox, H.L. and K.W. Pepper. 1944. Paper-based plastics. Part 1, the preparation of phenolic laminated boards. *J. Soc. Chem. Ind.* May 1944:150-154.

Engman, C., E. Alfthan, and E. de Ruvo. 1976. Mechanical properties of paper-polymer composites. *Tappi* 59(5):117-121.

Fisa, B. and R.H. Marchessault. 1974. Preparation of a microporous composites: Encapsulation of paper by polyethylene. *J. Appl. Polym. Sci.* 18:2025-2037.

Fisa, B., J.F. Revol, and R.H. Marchessault. 1976. Physical and mechanical properties of polyethylene-encapsulated paper. *Tappi* 59(8):88-91.

Goldstein, G. 1984. Future markets and competitive threats to nonwovens. *Tappi* 67(10):44-48.

Krock, R.H. and L.J. Broutman. 1967. Principles of composites and composites reinforcement. In: L.J. Broutman and R. H. Krock (eds.), *Modern composite materials*. pp. 3-26. Addison-Wesley Pub. Co., Reading, Mass.

Maldas, D., B.V. Kokta, R.G. Raj, and C. Daneault. 1988. Improvement of the mechanical properties of sawdust wood fibre-polystyrene composites by chemical treatment. *Polymer* 29:1255-1265.

Marchessault, R.H., B. Fisa, and J.F. Revol. 1975. Nascent polyethylene-cellulose composite. *ACS Symposium Series No 10: Cellulose Technology Research* pp. 147-159.

Michell, A.J., J. Vaughan, and D. Willis. 1976. Wood fiber-synthetic polymer composites. I. Laminates of paper and polyethylene. *J. Polym. Sci. Symposium No. 55*: 143-155.

Michell, A.J., J. Vaughan, and D. Willis. 1978. Wood fiber-synthetic polymer composites. II. Laminates of treated fibers and polyolefins. *J. Appl. Polym. Sci.* 22:2047-2061.

Michell, A.J. and D. Willis. 1978. Cellulosic fibers for reinforcement. *Appita*. March 1978:347-354.

Prud'homme, R.E. 1977. Mechanical properties of polymer-paper laminates. *J. Appl. Polym. Sci.* 21:947-957.

Prud'homme, R.E. and A.A. Robertson. 1976. Composites theories applied to oriented paper sheets. *Tappi* 59(1):145-148.

Raj. R.G., B.V. Kokta, and C. Daneault. 1990. Wood flour as a low-cost reinforcing filler for polyethylene: studies on mechanical properties. *J. Mater. Sci.* 25: 1851-1855.

Raj. R.G, B.V. Kokta, D. Maldas, and C. Daneault. 1989. Use of wood fibers in thermoplastics. VII. The effect of coupling agents in polyethylene-wood fiber composites. *J. Appl. Polym. Sci.* 37: 1089-1103.

Westerlind, B., A. Larsson and M. Rigdahl. 1987. Determination of the degree of adhesion in plasma-treated polyethylene/paper laminates. *Int'l. J. Adhesion Adhesives* 7(3):141-146.

Zadorecki, P. and P. Flodin. 1985. Surface modification of cellulose fibers. II. The effect of cellulose fiber treatment on the performance of cellulose-polyester composites. *J. Appl. Polym. Sci.* 30: 3971-3983.

Zadorecki, P. and P. Flodin. 1986. Surface modification of cellulose fibers. III. Durability of cellulose-polyester composites under environmental aging. *J. Appl. Polym. Sci.* 31:1699-1707.

CHAPTER 5

CONCLUSIONS

Chemical compositions of virgin and recycled fibers from four sources showed significant differences among fiber types. Virgin southern pine fibers showed highest alcohol-benzene extractive content and lignin content, while recycled fibers from waste office paper and old corrugated cardboard had considerably higher holo- and alpha-cellulose contents. Average fiber lengths of the three recycled fiber types were statistically the same, but appreciably shorter than virgin fiber. Disintegrating revolution negatively affected average fiber length.

Replacing virgin fiber with recycled fiber adversely affected physical and mechanical properties of fiberboard. Bending properties and dimensional stability were linearly dependent on virgin fiber ratio. Internal bond strength had a parabolic relationship with virgin fiber ratio. Wettability of fiberboard, as measured by contact angle, decreased as recycled fiber increased. All panels with recycled fiber content greater than 40% failed to meet any commercial requirement.

With fixed polyethylene content (20%), IB, MOR_b, MOE_b, and dimensional stability were directly related to virgin fiber ratio. Layered structures showed similar strength properties and dimensional stability with homogeneous configuration. However, using layered structures with 40% recycled fiber, reduction virgin fiber from 60% to 40% by replacing with polyethylene did not affect panel strength properties, but appreciably increased dimensional stability.

At a given composition, i.e., virgin fiber:recycled fiber:polyethylene=4:4:2, internal bond strength was affected by both hot press temperature and panel density, whereas bending properties and water absorption were altered by panel density. However, thickness swelling was unaffected. Hot press temperature and hot press time noticeably affected wettability of wood fiber-polyethylene composites. Panels fabricated at 350°F for 5 min. with target panel density of 60 pcf resulted in the best panel properties which passed specifications for commercial class 4-service hardboard.

Contact angles on birch plywood surfaces increased upon addition of E-43 and decreased with an increase in E-43 concentration. For E-43-treated wood surfaces, contact angles among the four wetting liquids were in the order of PF>UF>ISO>water. Contact angles on polyolefin polymer surfaces, on the other hand, followed the sequences of PP>LLDPE>LDPE and water>UF>PF>ISO.

Interfacial shear strengths measured by a modified single-fiber pull-out test indicate that the overall average interfacial shear strength was highest in PP and lowest in LDPE. For LDPE, wood surface treated with ISO and without E-43 showed the highest interfacial shear strength. In LLDPE, E-43-treated wood surfaces without the addition of adhesive displayed a superior performance. With few exceptions, the addition of E-43 greatly enhanced the interfacial shear strengths for all plastic types. The effect of E-43 was most pronounced in PP, and less in LDPE and LLDPE. However, increasing E-43 concentration exhibited a negative effect toward interfacial shear strength. The most efficient

amount of solid E-43 content needed to improve interfacial shear strength was different from the amount needed to achieve best wettability.

Adhesive failure prevailed in LDPE and LLDPE model composites while PP showed predominately cohesive failures either in wood dowel or in plastic matrix. About one third of the PP model composites exhibited dowel failure at the grip end, resulting in an undetermined failure mode. Results of failure mode were in good agreement with interfacial shear strengths.

Wet-formed Kraft pulp handsheets were interleaved with polypropylene films to fabricate laminates of 1/8-inch thickness which had comparable ultimate tensile strengths with natural fiber-reinforced plastics. Experimental results showed that laminates of LAYER=1 had better MOR_t than those of LAYER=3, due to better interpenetration of plastic. Effects of fiber content and LAYER on elongation at break were not consistent at two higher fiber content levels; however, 50% fiber content showed lowest elongation at break at all LAYER levels. Interaction between fiber content and LAYER compensated the main effects on MOE_t . An increase in fiber content and LAYER led to an increase in WA, LE and TS. WA and TS showed a similar trend, but LE was less sensitive to changes in fiber content and LAYER. With proper control of LAYER, wood fiber-polypropylene laminates with fiber loading as high as 70% can be fabricated with favorable mechanical properties. However, without any surface treatment, lower fiber content and LAYER had the best dimensional stability.

VITA

Chin-Yin Hwang was born in Taipei, Taiwan. She received her bachelor of science degree in Forestry (Wood Science) from National Chung-Hsing University in 1976. Upon graduation, she worked as a temporary research associate at the Taiwan Forestry Research Institute where she found that research would be her lifelong career and started to work in that direction. She was twice the recipient of the Fellowship of National Science Council. Ten years after her bachelor degree, she received her master of science degree in Forestry (Wood Science) from National Taiwan University in 1986. In 1993, she took a study leave from her assignment as an assistant scientist with the Taiwan Forestry Research Institute, and started pursuing her doctor of philosophy degree in the School of Forestry, Wildlife, and Fisheries, Louisiana State University. She plans to complete her doctoral studies in May of 1998 and continue her career in research in Taiwan, Republic of China.

DOCTORAL EXAMINATION AND DISSERTATION REPORT

Candidate: Chin-Yin Hwang

Major Field: Forestry


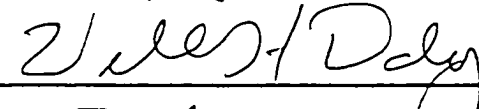
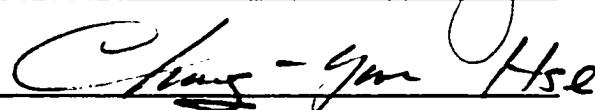
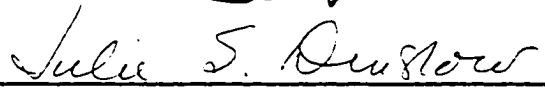
Title of Dissertation: Effect of Recycled Fiber, Compatibilizer and Pre-formed Fiber Handsheet on the Performance of Wood-Polyolefin Composites

Approved:


Major Professor and Chairman


Dean of the Graduate School

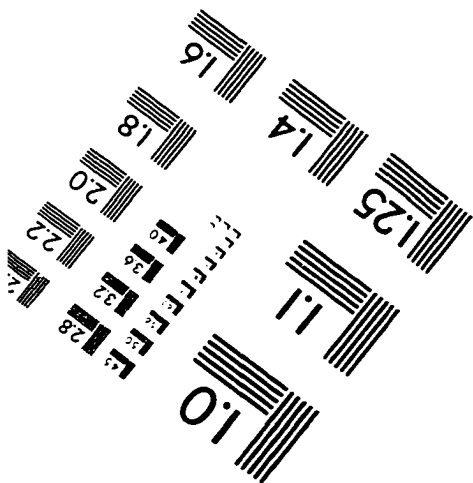
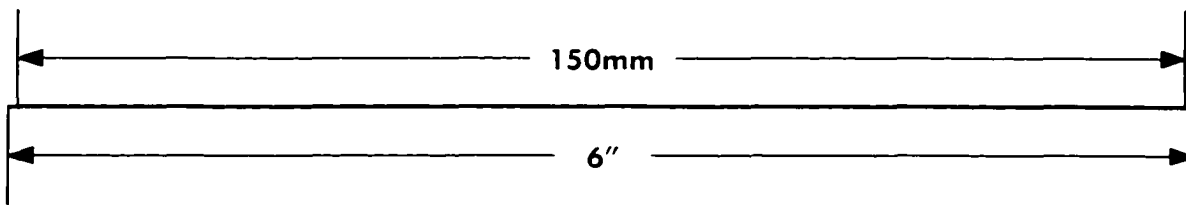
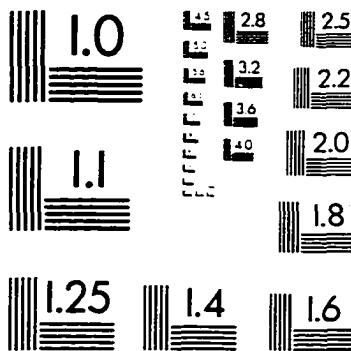
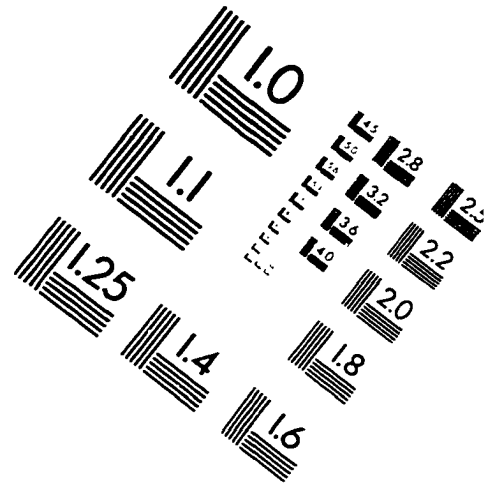
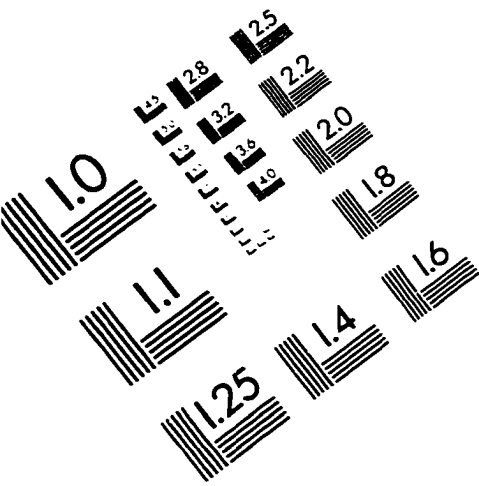
EXAMINING COMMITTEE:

Date of Examination:

March 5, 1998

IMAGE EVALUATION TEST TARGET (QA-3)



APPLIED IMAGE, Inc
1653 East Main Street
Rochester, NY 14609 USA
Phone: 716/482-0300
Fax: 716/288-5989

© 1993, Applied Image, Inc.. All Rights Reserved

