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Enhancing the surface finish of long fiber thermoplastic extrusion-compression molded parts using in-mold film technology

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ENHANCING THE SURFACE FINISH OF LONG FIBER THERMOPLASTIC
EXTRUSION-COMPRESSION MOLDED PARTS USING IN-MOLD FILM
TECHNOLOGY

Thesis

Submitted to

The School of Engineering of the
UNIVERSITY OF DAYTON

in Partial Fulfillment of the Requirements for

The Degree

Master of Science in Materials Engineering

by

Jessica Lynn Ravine


UNIVERSITY OF DAYTON

Dayton, Ohio

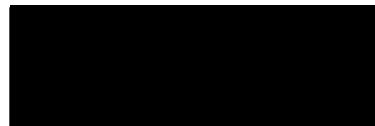
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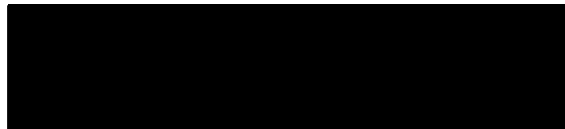
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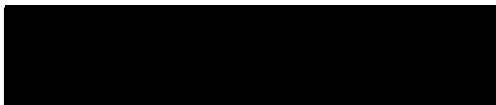
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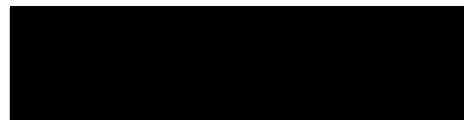
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ABSTRACT

ENHANCING THE SURFACE FINISH OF LONG FIBER THERMOPLASTIC EXTRUSION-COMPRESSION MOLDED PARTS USING IN-MOLD FILM TECHNOLOGY

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A limiting factor for the use of long fiber thermoplastics (LFT) in semi-structural applications is the material's less than ideal surface finish. Painting LFT material is complex, costly and harmful to the environment. For LFT extrusion-compression molded materials, specifically glass-reinforced polypropylene, in-mold film technology is explored as an alternative approach for addressing this limitation. Processing variables anticipated to influence the success of incorporating a surface film in the compression molding process are identified. A design of experiments is crafted to evaluate the effect of these variables and to establish optimum operating parameters. Gloss, adhesion of surface film to substrate panel, and chip resistance testing are performed on panels from the experimental runs. Statistical analysis identifies film preheat temperature and film thickness to be significant variables, while residence time in mold and glass content of the LFT substrate panel are found to be statistically insignificant on the properties considered. Optimum operating parameters for

product from two film suppliers are established. For Film Supplier I, whose product line currently includes only one film thickness of 1.70 millimeters, freezing the film at zero degrees Celsius prior to processing yields the best gloss rating for the final product. For Film Supplier II, using the thinnest film available, 0.43 millimeters, and preheating the film to 50 degrees Celsius prior to processing are recommended to maximize gloss adhesion of the film to the LFT substrate panel, while using the thickest film available, 1.52 millimeters, and freezing the film at zero degrees Celsius prior to processing is favorable for maximizing gloss. Therefore, Film Supplier II operating parameters must be compromised based on the requirements for individual applications. In-mold film technology is identified as a promising approach for enhancing the surface finish of LFT extrusion-compression molded parts. Future work is necessary to address the heavy marring resulting from chip resistance testing on all panels, as well as to enhance gloss properties.

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TABLE OF CONTENTS

APPROVAL PAGE.....	ii
ABSTRACT.....	iii
ACKNOWLEDGEMENTS.....	v
LIST OF FIGURES.....	viii
LIST OF TABLES.....	x
CHAPTER	
I. INTRODUCTION.....	1
Long Fiber Thermoplastics.....	1
LFT Processing Methods.....	3
Advantages to using LFT Materials.....	5
Applications.....	6
Limitations due to Surface Finish.....	8
II. REVIEW OF RELATED RESEARCH AND LITERATURE.....	10
In-Mold Film Products.....	10
In-Mold Films with Thermoset Substrates.....	12
In-Mold Film Direct Thermoforming.....	13
In-Mold Films with Injection Molded Thermoplastics.....	14
In-Mold Films with Glass-Mat Thermoplastics.....	16
In-Mold Films with LFT-ECM.....	16
III. EXTRUSION-COMPRESSION MOLDING PROCESS.....	19
IV. MATERIALS.....	24
Substrate Panel.....	24
Surface Films.....	25

V. DESIGN OF EXPERIMENTS.....	27
Measurables.....	27
Variables.....	28
Levels and Matrix Design.....	29
Processing Runs.....	33
VI. TESTING.....	36
Adhesion of Surface Film to LFT Substrate Panel.....	36
Gloss.....	38
Chipping Resistance.....	39
VII. DATA.....	41
Adhesion of Surface Film to LFT Substrate Panel.....	41
Gloss.....	44
Chipping Resistance.....	45
VIII. ANALYSIS.....	48
Film Supplier I.....	48
Film Supplier II.....	52
IX. SUMMARY AND CONCLUSIONS.....	57
X. FUTURE WORK AND RECOMMENDATIONS.....	60
BIBLIOGRAPHY.....	62
APPENDICES	
A. Pull-off Testing Failure Modes.....	66
B. Pull-off Testing Raw Data.....	69
C. Gloss Testing Raw Data.....	71

LIST OF FIGURES

1. LFT Definition Relative to Other Composite Materials.....	2
2. Extrusion-Compression Molding Cell.....	21
3. Schematic of Film Insertion during Molding Process.....	21
4. LFT-ECM Panel with Supplier I Film.....	22
5. LFT-ECM Panel with Supplier II Film.....	22
6. Helmet Rib Geometry with Film.....	23
7. Complex Geometry with Film.....	23
8. Layered Surface Film Structure.....	26
9. Portable Adhesion Tester.....	37
10. Milled Ring with Pull-off Stud; Abraded Test Area.....	38
11. Gloss Meter.....	38
12. Gravelometer.....	39
13. Adhesion Testing Failure Modes.....	41
14. Micrograph of Interfacial Melt Bond.....	44
15. Close-up View of Representative Melt Bond Images.....	44
16. Representative Marring on Supplier I Films.....	46
17. Representative Marring on Supplier II Films.....	46
18. Main Effects Plots for Film Supplier I Gloss Response.....	50
19. Significant Main Effects Plots for Supplier II Adhesion Response.....	52

20. Main Effects Plots for Film Supplier II Gloss Response.....	54
21. Variable Interactions for Film Supplier II Gloss Response.....	54
A1. Representative Failure Mode 1 for Film Supplier I.....	66
A2. Representative Failure Mode 1 for Film Supplier II.....	66
A3. Representative Failure Mode 2 for Film Supplier II.....	67
A4. Representative Failure Mode 3 for Film Supplier II.....	67
A5. Representative Failure Mode 4 for Film Supplier I.....	68
A6. Representative Failure Mode 4 for Film Supplier II.....	68

LIST OF TABLES

1. Design of Experiment Goals, Responses and Variables.....	29
2. Design of Experiment Matrix for Film Supplier I.....	31
3. Design of Experiment Matrix for Film Supplier II.....	32
4. Variable and Level Definitions.....	32
5. Processing Matrix.....	34
6. Uncoded Processing Matrix.....	35
7. Number of Chips Rating System.....	40
8. Chip Size Rating System.....	40
9. Adhesion Testing Failure Modes.....	41
10. Summary of Tensile Pull-off Test Results.....	43
11. Summary of Gloss Test Results.....	45
12. Marring Rating Data.....	47
B1. Pull-off Tensile Strength Data for Panels 1 – 20.....	69
B2. Pull-off Tensile Strength Data for Panels 21 – 40.....	70
C1. Gloss Data for Panels 1 – 15.....	71
C2. Gloss Data for Panels 16 – 40.....	72

CHAPTER 1

INTRODUCTION

Long fiber thermoplastics extrusion-compression molding is a promising material and process solution for semi-structural applications; however, its use in such applications with surface finish requirements has been limited due to unsatisfactory color, gloss and other cosmetic characteristics of parts produced by this process. Incorporating a surface film in the compression molding process is explored as a candidate technology for addressing this limitation.

Long Fiber Thermoplastics

Composite materials are composed of two or more distinctly different materials that when combined form a product exhibiting superior properties to those possessed by the individual constituents¹. Composites typically consist of reinforcement such as fiber, and a resin or matrix material surrounding the reinforcement and acting as a binder. Resins can be categorized as thermosets or thermoplastics. Thermosets start as low molecular weight materials, and during "cure" undergo chemical reactions to form high molecular weight cross-linked polymers. These densely cross-linked three dimensional networks are characterized by strong covalent bonds², and once the polymerization reaction takes place it cannot be reversed. Thermoplastics, on the other hand, are

already polymerized to a high molecular weight material, and can be melted and reformed due to their uncrosslinked, linear structure.

Composite reinforcement can include particulate and continuous or discontinuous fibers¹. Discontinuous fibers are formed by chopping continuous fiber, and can further be classified as short or long. Long fiber thermoplastics refer to a composite consisting of thermoplastic resin and discontinuous fiber having a minimum length of approximately two centimeters, although the length classifying a fiber as “long” is a topic of debate and also varies with the process being considered³. The position of long fiber thermoplastics within the family of composite materials is illustrated in Figure 1.

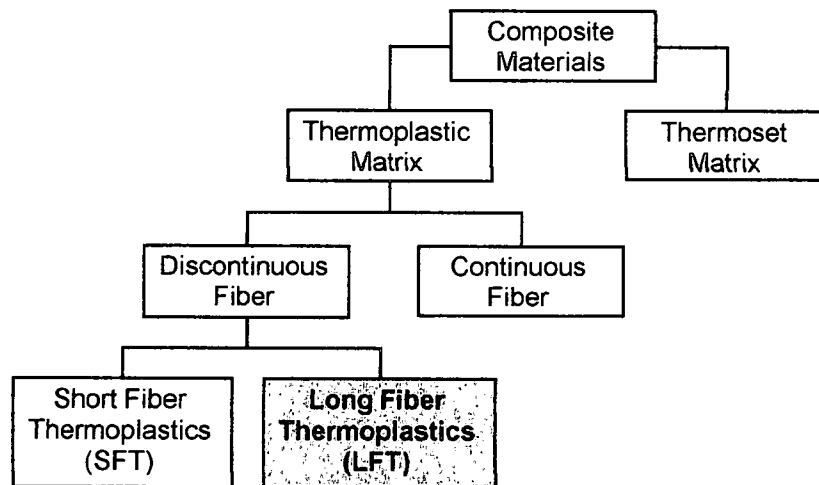


Figure 1: LFT Definition Relative to Other Composite Materials

LFT materials can be reinforced with glass, carbon or aramid fibers and are typically made of low cost thermoplastic polymers such as polypropylene, polyamide, polyethylene and polycarbonate⁴.

LFT Processing Methods

Long fiber thermoplastic processes often begin with a pultrusion process in which continuous fiber is pulled through a resin bath to form a continuous spool of fiber pre-impregnated with a thermoplastic matrix⁵. These pre-consolidated tapes can then be chopped into "granules" or "pellets" of desired length for further processing. Alternatively, LFT granules or pellets can be prepared by wire coating or cross-head extrusion⁶. LFT processes including this precursor technology are referred to as LFT-G or LFG, where "G" represents granules. The granules or pellets can then be incorporated into compression molding or injection molding processes⁷.

LFT compression molding processes can include both glass mat-reinforced compression molding and extrusion-compression molding. Glass mat-reinforced thermoplastics, or GMT, are semi-finished sheets in which chopped or continuous glass fibers are randomly oriented in-plane in a thermoplastic matrix. GMT was developed in the 1960's, and represented the driving force for thermoplastic composite developments for many years⁷. The GMT sheets can be "dry processed" via melt impregnation, where polymer films and glass fiber mats are layered and consolidated via compression molding. Alternatively, GMT can be "wet processed" by dispersing chopped fibers with polymer powder in a fluid, followed by straining, drying and consolidation⁸. Chopped fibers used in either of these processing techniques can be of a length classifying the resulting GMT as LFT material (LFT-GMT)³.

In extrusion-compression molding (ECM), pre-consolidated tapes (or comingled tape and fiber or LFT granules or pellets) are fed into a plasticator, where they are melted and mixed at low shear rates to minimize fiber degradation. The molten, dispersed extrudate of LFT is discharged from the extruder and subsequently compression molded to form the desired geometry. When LFT granules or pellets are extruded and compression molded, the process can be referred to as LFG-ECM⁷.

Similarly, when LFT granules or pellets are a precursor for injection molding, the process can be referred to as LFG-IM. Recent developments in LFT processing have largely focused on direct processing techniques that avoid the step of semi-finished products, referred to as D-LFT. D-LFT processes can also be applied to both extrusion-compression molding and injection molding (D-LFT-ECM and D-LFT-IM, respectively). D-LFT processes in use include three basic technological approaches⁹. The first technology uses two extruders to compound chopped fibers and polymer melt. The second approach uses a single twin-screw-extruder to compound polymer and continuous roving. The third technology also compounds polymer and continuous roving, but uses two separate twin-screw-extruders. There are also variations of these as well as the rest of the above processes, distinguished by various equipment design and operating parameters.

Another area of recent technical interest is referred to as tailored or hybrid LFT, where oriented fiber, fabrics or unidirectional materials are incorporated into the above processes as local reinforcements³.

Advantages to using LFT Materials

Primary advantages to choosing LFT materials over traditional metal components for applications such as automotive include significant cost and weight savings. One study evaluating LFT's potential for use in a bus battery access door reported weight savings of 60% and a cost reduction of 44% compared to the existing steel door, while meeting or exceeding all design requirements¹⁰.

Maintaining considerable fiber length during processing allows LFT composites to retain a higher percentage of mechanical properties exhibited by continuous fiber composites. For example, for glass fiber-reinforced polypropylene, increasing fiber length from 1.2 to 4.2 millimeters is experimentally found to yield a six percent increase in flexural modulus, 33 percent increase in flexural strength, three percent increase in tensile modulus, 51 percent increase in tensile strength and 122 percent increase in notched Izod impact strength¹¹.

Another important advantage of LFT materials is that unlike continuous fiber-reinforced composites, they can be processed using traditional plastics molding equipment¹⁰. The ability to build upon established processing techniques makes higher production capacity and increased part consistency less formidable challenges than often faced by new materials and techniques. In addition, the thermoplastic matrix facilitates mixing of additives to impart application-specific properties such as fire retardancy, thermostability, ultraviolet stability and color as well as to improve fiber/matrix adhesion characteristics³.

Other beneficial properties of LFT materials include corrosion resistance, toughness, durability, noise absorption, good crash features due to high energy absorption and ductile fracture failure⁷, low moisture sensitivity (except polyamide), low emissions, amenability to thermal post-processing such as welding and reforming, unlimited shelf life without the need for cooling⁷, short cycle times and amenability to functional integration, as well as intrinsic recyclability.

Applications

Low cost, low weight, prominent physical and mechanical properties and the added benefit of processability make LFT composites an attractive alternative to traditional materials such as steel in structural and semi-structural applications. The automotive industry often pioneers the investigation of promising light weight technologies, and such is the case with long fiber thermoplastic materials. Approximately half of the current United States market for thermoplastics composites represents consumption by the automotive industry¹². Specific automotive applications include front-ends, bumper beams, dashboards, under body shields, seat structures, body structures, hoods, spare wheel well and cover, door modules and floor panels¹². The ability to produce parts at medium to high capacities makes LFT materials promising for mass-transit applications in buses, trucks and the railroad industry.

The average growth of long fiber polypropylene applications in the automotive industry was 17 percent over the ten years between 1992 and 2002¹³. GMT is becoming a mature market, while LFT-G and D-LFT have

realized rapid growth since 1995 and 1999, respectively. The LFT-G market growth between 2002 and 2005 was over 18 percent, while D-LFT market growth was in excess of 20 percent¹³. Europe appears to be playing a leading role in LFT development, based in part on the European Union's ratified end-of-life vehicle (ELV) directive, for which LFT's recycling potential is an important factor. The objectives of Directive 2000/53/EC of the European Parliament and Council, issued in September of 2000, include the minimization of the impact of end-of life vehicles on the environment by prevention of waste from vehicles and the reuse, recycling and recovery of end-of life vehicles and their components. The directive targets to increase the reuse and recycling to a minimum of 85% by average weight per vehicle and year no later than January, 2015¹⁴. Europe's LFT market is anticipated to grow to over 110 kilotons per year by 2009⁷.

In 2002, thermoplastic composites market distribution in the United States was 75 percent GMT, 12.5 percent LFT-G and 12.5 percent D-LFT, totaling 15 kilotons¹³. Asia's thermoplastic composites market was three times smaller, and included 75 percent GMT and 25 percent LFT-G. United States LFT market growth is projected to be greater than ten percent per year in coming years, while market growth in Asia is anticipated to be between ten to 20 percent per year. The United States and Asia do have a higher market share of non-automotive and non-polypropylene applications than Europe¹³. Non-automotive applications for LFT materials have included tailcones for army tank guns⁴, steam valve covers, protective helmets and lawn mower chain housings.

Limitations Due to Surface Finish

The extensive list of benefits for selecting LFT materials for semi-structural and structural applications includes low cost, low weight, impressive mechanical properties, superior impact properties and processability. The ability to manufacture components with large areas and thin walls at high production capacities makes LFT the first viable alternative to steel for exterior automotive body panels. However, this market potential remains untapped as a result of the inability to achieve the surface quality characteristics or paintability required for exterior body panels¹⁵.

Car exterior body panels include horizontal panels, vertical panels and bumpers and trim parts¹⁵. LFT's stiffness, strength and impact resistance can meet the stringent requirements of horizontal body panels as well as tailgates, while vertical panels have less strict structural requirements. The primary requirement of bumpers and trim parts is impact resistance. LFT has the ability to satisfy these property requirements, but is sometimes incompatible with traditional painting techniques due to the low surface energy of some thermoplastics. In addition, paint lines are capital intensive and often introduce harmful volatile organic compounds (VOC's) into the environment. It is estimated that painting and associated operations can constitute 30 to 50 percent of part cost¹⁶, often representing the largest investment in an auto assembly plant. Painting often requires multi-step processes including acid washing and subsequent rinsing prior to paint application, and paint lines can occupy nearly

half of plant floor space. Such operations can generate over 1,500 tons per year of VOC emissions¹⁷.

The incentive to develop surface film technology as an alternative to painting include reduction in capital costs and volatile emissions, as well as additional benefits such as ultraviolet light protection, weatherability and scratch resistance. Non-automotive applications for which in-mold film technology could be used are also widespread, including products such as helmets, agricultural equipment¹⁶, hand-held electronics and control panels for home appliances, as well as lawn and garden, construction and marine applications¹⁷.

CHAPTER II

REVIEW OF RELATED RESEARCH AND LITERATURE

Although the concept of using in-mold film technology is in the early stages of development for the LFT extrusion-compression molding process, a little more work has been done with in-mold film technology for thermosets, thermoplastic neat and fiber-reinforced injection molding and glass-mat compression molding processes. Lessons learned from these studies and existing applications may in some ways accelerate the development of the technology for LFT extrusion-compression molding.

In-Mold Film Products

A diverse number of in-mold film products are currently offered by a variety of suppliers. Use of the films in thermoplastic injection molding processes is common for household appliance control panels and portable electronic devices. The films are often processed by polymer extrusion, and color layers and clear, protective topcoats can be imparted by additional pigmented plastic layers or via dry paint layers that are sprayed on and subsequently cured. Despite the variety of film products available, the multilayered film structure approach is common and appears to be recognized within the industry as the most viable method for attaining multiple product specification requirements. A

plastic layer facilitates thermal processing, followed by a carrier layer for color and a clear topcoat for gloss and durability.

Bayer MaterialScience, LLC has been a main player in the development of in-mold film technology, an effort reportedly undertaken with exterior auto body components in mind from the start¹⁸. Bayer's films include a thermally stable plastic carrier film such as polycarbonate, topped with a dual-cure, thermoset polyurethane coating. The base coat with the desired color and/or metallic effect and clear coat are applied to the carrier film, and each is dried with heat. The partial curing imparts flexibility that allows the film to be stretched through thermoforming without cracking. In this step, the top coat builds long polyurethane chains that are still formable because they are not yet crosslinked. The films are then sold to processors who thermoform them into the desired shape, after which a high-energy ultraviolet light exposure step is required to complete crosslinking of the clear coat. The thermoplastic carrier imparts thermoformability while the polyurethane top coat is ultraviolet resistant, chemical resistant, high-gloss and designed to be durable enough to withstand weather, road grit and mechanical car washes. The films can be used in thermoplastic injection molding or in polyurethane reaction injection molding. Bayer's thermoset topcoat provides durability achieved with high crosslinking, a result not attainable with typical thermoplastic films.

BASF is another frontrunner in in-mold film development. BASF's line of "paintless film molding (PFM) skins" includes colors, textures and grained effects¹⁹. BASF has chosen styrenic materials for both the film and backing

composite as a result of the material's lower coefficient of linear thermal expansion compared to many other thermoplastics. This material choice is designed to minimize potential issues presented by combining composite and steel parts in assembled automobiles. Typical molding times with PFM skins are 30 to 40 seconds, and the skins are typically backmolded with 20 to 30 percent glass-reinforced acrylonitrile-butadiene-styrene (ABS) or styrene-acrylonitrile (SAN).

Similar products are available from Avery Dennison, Soliant, Mayco Plastics, Senoplast USA and GE Plastics, among others¹⁷.

In-Mold Films with Thermoset Substrates

One study by Florida State University²⁰ incorporating Avery Dennison Avloy film with glass fibers and vinyl ester resin in the "RIDFT" Resin Infusion between Double Flexible Tooling process identifies one of the primary drawbacks for using thermoset materials versus thermoplastic for in-mold film processing. The heat required to soften the paint film adequately to attain proper adhesion instigated accelerated cure of the vinyl ester resin. This reduction in the time available for forming impaired the processability of the composite system.

One example of in-mold film technology that has successfully transitioned to market is evidenced by the optional roof module of the Opel Zafira²¹. The Class-A surface roof module is produced by back-filling polyurethane glass fiber-reinforced foam with an un-named high-gloss thermoplastic film. Success of the approach is reportedly largely attributed to the coefficient of linear thermal expansion (CTE) of the polyurethane foam being similar to that of aluminum,

imparting outstanding heat resistance to the roof module. The trendsetting design of the “panorama” roof module provides overhead storage and incorporates four glass panels, making the vehicle brighter and seemingly roomier while expanding the view. Although the fiber-reinforced polyurethane foam adds mechanical and workability properties, the load-bearing element of the roof module is a 2.1 by 1.1 meter frame without film finish.

The roof module of the 2003 Smart two-seat roadster is described by GE Plastics as the first paintless horizontal body panel with a “Class A” finish. The component is a long glass-reinforced polyurethane composite processed with in-mold film technology¹⁷.

In-Mold Film Direct Thermoforming

A dry paint film from Soliant, LLC has been thermoformed to produce a relatively large paint-film application – the rocker molding for the 2003 General Motors Chevrolet Trailblazer North Face Edition²². This part is 6.5 inches long and has high-gloss and color matching the body of the trailblazer. The Soliant film is called Fluorex and consists of a thermoformable masking film that is stripped off after forming, a fluoropolymer clear coat, a color coat and a tie/coat primer. The substrate is a 0.150 inch thermoplastic olefin (TPO) sheet extruded and laminated to the paint film. The rocker panel is thermoformed directly, not backfilled via injection molding.

In-Mold Films with Injection Molded Thermoplastics

A study to evaluate the effect of injection molding parameters on mechanical and morphological properties of polypropylene film and matrix specimens found that increasing injection speed, barrel temperature and holding pressure lead to increased bonding between the film and substrate surfaces, possibly attributed to molecular interdiffusion²³. Bending properties with and without film attached are found to be similar, supporting theory that static properties are less effected by film-substrate bonding than dynamic properties such as impact strength. Impact strength is found to decrease in the samples with film. A subsequent study by the same group confirms this result can be attributed to higher crystallinity caused by obstructed cooling during the injection molding process for film-insert specimens²⁴. In fact, higher crystallinity is observed not only near the film-substrate interface, but within the total substrate structure. Higher crystallinity is also found to correlate with increase in barrel temperature.

Another study considers interfacial characteristics of polycarbonate films injection molded with polycarbonate-ABS substrate²⁵. Increasing molecular weight of the substrate by blending polycarbonate with ABS enhances both impact resistance and interfacial adhesion properties of the film-insert molded parts. Film thickness is found to play a critical role in film-substrate adhesion, likely as a result of several factors. First, increasing film thickness results in a decrease in cavity volume, creating a higher pressure against the film (facilitating melt bonding) when processing parameters such as injected material amount are

not adjusted. Thicker films can also enhance impact resistance of the bulk part by transitioning energy from the substrate to the film (despite increasing substrate brittleness by impairing cooling during molding). A thin film can also improve impact resistance by resisting crack propagation by delaminating at the film-substrate interface. Oligomers were found to hinder mechanical interlocking and intermolecular interaction at the film-substrate interface, an initially counter-intuitive result as the oligomers were anticipated to improve interfacial adhesion based on surface migratory and high tacticity characteristics. It is theorized that the oligomers migrating to the surface of the moldings are preventing potential interaction between resin molecular chains with those at the film surface.

Finally, an in-mold film injection molding study with film and substrate consisting of polyethylene terephthalate investigates the effect of localized molecular orientation on interfacial properties²⁶. An ultra-high-speed injection molder is used and two injection speeds are considered to create a high shear rate between melt resin and film surface, potentially creating localized molecular orientation. The moldings are subjected to isothermal annealing at different temperatures and time intervals to induce slow crystallization, allowing the observation of interfacial molecular reorganization and crystallinity. Increased crystallization is confirmed in all specimens via differential scanning calorimetry (DSC) and Fourier transform infrared spectroscopy (FTIR), although more substantial crystallization is observed in specimens subjected to a higher injection speed. Lower injection speed is found to promote random orientation of substrate molecules, allowing them to navigate more freely across the interface

to interact with film molecules, leading to stronger interfacial bonding. The higher injection speed appears to have oriented molecules anisotropically toward the flow direction, minimizing substrate-film molecular interaction and reducing interfacial adhesion.

In-Mold Films with Glass-Mat Thermoplastics

Several trials have been conducted in recent years to explore GMT as a candidate process for exterior body panel applications²⁷. Waviness of the surface as a result of matrix shrinkage, fiber print-through and flow lines are among the difficulties encountered during such trials. Lessons learned include the need for a two-layer film concept to satisfy both surface appearance and structural requirements, CTE match between film and substrate materials to minimize warpage and a dry molding process to achieve a homogeneous appearance on the final part. In addition, the substrate material must be able to be processed at temperatures low enough to avoid damage to the surface film and the interfacial adhesion attained at that process temperature must be characterized.

In-Mold Films with LFT-ECM

Dieffenbacher is reportedly working with BASF and the Fraunhofer Institute to apply in-mold film technology to D-LFT extrusion-compression molding¹⁹. The compression molded parts exhibit 50 percent higher impact resistance than those processed via injection molding. Prototype components employ styrenic thermoplastics and reportedly have passed a first successful

“feasibility study.” The compression molding trials have exhibited less fiber orientation and less warpage than injection molded components. The BASF skins are thermoformed then backmolded with 20 to 30 percent glass-reinforced ABS or SAN, with cycle times of 30 to 40 seconds, whereas 60 seconds are required for comparable sheet molding compound (SMC) processing. BASF reports that roof modules will be the first step.

The first D-LFT exterior part consisted of glass-reinforced polypropylene and was introduced at the end of 2005 on the lower section of a rear hatch of a European car¹⁹. However, the part does not employ in-mold film technology and does not have a Class A finish, but rather a stippled grain surface. Latest developments on D-LFT-ECM in-mold film processing include chopped fiber feeding for enhanced fiber dispersion to avoid print-through at the surface of Class A exterior body panel parts, as well as efforts to incorporate additional local reinforcements.

CHAPTER III

EXTRUSION-COMPRESSION MOLDING PROCESS

The process utilized in this study is LFG-ECM, extrusion-compression molding with pellets of pre-impregnated fiber. Direct LFT (LFT-D) technology was not employed only due to lack of access to associated equipment.

The utilized extrusion-compression molding cell, housed at the National Composite Center, consists of a 150 millimeter barrel diameter C. A. Lawton plasticator and 400 ton down-acting compression molding press. The plasticator has the ability to accommodate pellets, tapes and comingled fiber and tape. Pellets used are typically in the size range of 12 to 50 millimeter length. The low shear reciprocating screw is driven by a hydraulic motor. The low-wear cylinder is electrically heated; temperature is controlled using an Allen Bradley Programmable Controller and can range up to 800°F. The screw is designed to minimize oxidation during processing, as well as to maintain a consistent material backpressure to reduce potential process variation. The length to diameter ratio of the cylinder is 34:1, and stroke is greater than or equal to 500 millimeters. The screw rotational speed can range from ten to 45 revolutions per minute, promoting low shear to minimize fiber degradation during processing. The maximum shot size is 14 pounds, which can be discharged in one, two or three increments. A hydraulic knife cuts the extrudate prior to release from the

plasticator. Using this system, material can be processed at a rate of 9.2 pounds per minute.

The extrudate must be manually transferred from the plasticator to the compression mold. The press has a maximum clamp tonnage of 400 tons and stands 20 feet tall with a maximum clamp stroke of 72 inches. The bed area available for molding is 32 by 72 inches. Clamp speed can range from one to 800 inches per minute.

The mold employed during this study is a 9.5 inch by 23.5 inch flat panel mold. The thickness of the panels can be varied from 0.15 to 0.50 inches. The mold features hydraulic ejectors at six different points to minimize local stresses. The mold is designed with the ejectors on the bottom half; therefore, during processing the LFT extrudate is placed in the mold, then the surface film is centered atop the extrudate prior to closing the press, allowing the surface film to be on the flat panel surface. It is worth noting that in the majority of in-mold film studies considered, the film is placed in a mold first then backfilled with LFT material. The available mold design and the nature of the extrusion-compression molding process versus the injection molding processes focused on in related research to date facilitated this approach.

A diagram of the extrusion-compression molding facility is illustrated in Figure 2. A schematic of the film insertion process is provided in Figure 3. Examples of LFT extrusion-compression molded panels with films from Supplier I and II are exhibited in Figures 4 and 5, respectively. Examples of other parts processed with LFG-ECM in-mold film technology are shown in Figures 6 – 7,

demonstrating the ability to apply the technology to parts with more complex geometries.

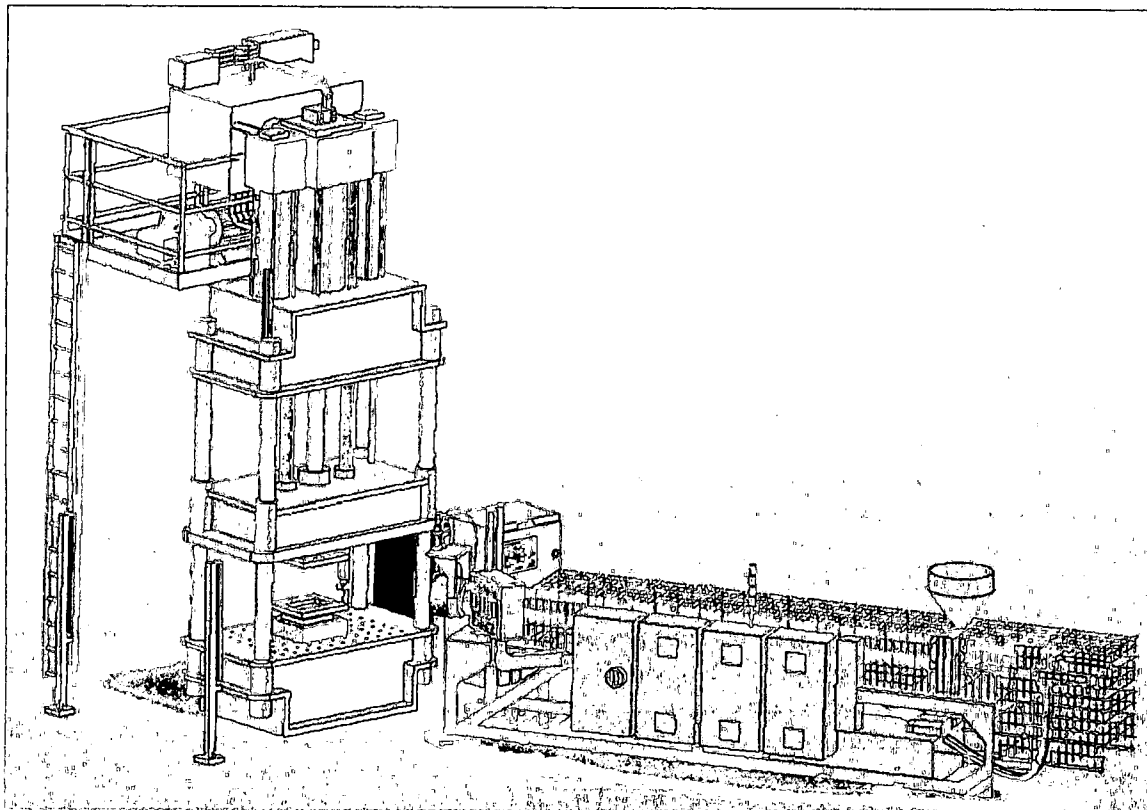


Figure 2: Extrusion-Compression Molding Cell. Illustration by Pritam Das, National Composite Center. Used with permission.

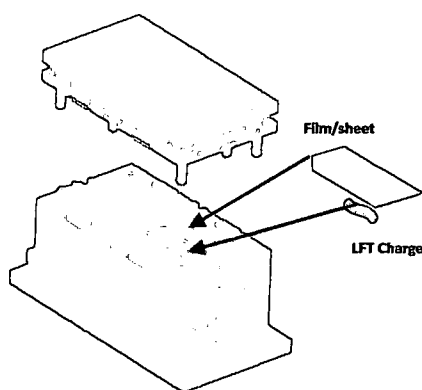


Figure 3: Schematic of Film Insertion during Molding Process. Illustration by Pritam Das, National Composite Center. Used with permission.



Figure 4: LFT-ECM Panel with Supplier I Film



Figure 5: LFT-ECM Panel with Supplier II Film



Figure 6: Helmet Rib Geometry with Film

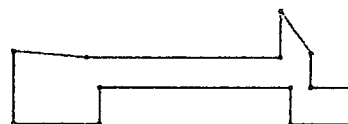


Figure 7: Complex Geometry with Film

CHAPTER IV

MATERIALS

Materials used in this study are chosen in part due to availability to mitigate cost. All materials, including LFT pellets and both varieties of surface films, are donated to the National Composite Center and used with permission.

Substrate Panel

Polypropylene-glass LFT pellets are selected for use in the study as polypropylene is the most common thermoplastic currently in use in the automotive industry¹³. Beneficial properties include short cycle time, toughness and high damage tolerance, low emissions, low sensitivity to moisture, recyclability, relatively low cost, unlimited shelf life and amenability to thermal post-processing³. Fiberglass is the only reinforcement economical enough for widespread commercial use at this time, and the material combination is known to have the ability to satisfy a large number of structural and semi-structural application requirements. Polypropylene-glass LFT materials are theoretically possible of attaining up to 90 percent of continuous polypropylene-glass strength¹², and mechanical properties as a function of fiber length and content are well-established compared to data available for other material systems^{5,6,11,28}.

In addition, glass fiber sizing in polypropylene matrix is further developed compared to many other resin systems.

Pellet lengths of 12.5 and 25 millimeters are both commercially available. Critical fiber lengths for many polypropylene-glass mechanical properties are estimated to be in the nine to 12 millimeter range^{5,11,28}. Anticipating some fiber degradation even with the low-shear extrusion process, a pellet length of 25 millimeters is selected to ensure that critical fiber lengths are maintained upon processing.

Surface Films

Surface films from two suppliers are considered in this study. Both film sources offer a thermoplastic olefin (TPO) backing which is expected to melt bond with the polypropylene substrate panel. In each case, the TPO layer is topped with an adhesive-backed carrier layer, which appears to be an extruded pigmented thermoplastic having a clear topcoat. The film structure is illustrated in Figure 8. The exact constituents of the film layers are confidential to the manufacturer.

The film from the first supplier is available in one thickness of 1.70 millimeters, having a blue color. Films from the second supplier are provided in three thicknesses, 0.43, 0.76 and 1.52 millimeters, and are black in color.

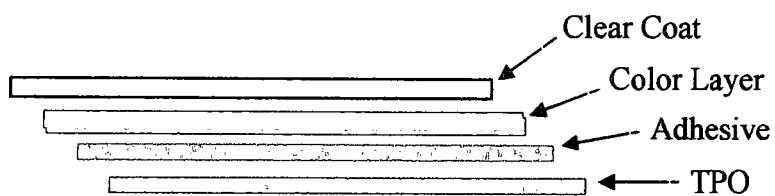


Figure 8: Layered Surface Film Structure

CHAPTER V

DESIGN OF EXPERIMENTS

Design of experiments is utilized to establish a reasonable number of processing runs while still considering several variables and levels. Availability of materials, schedule constraints and costs of equipment use and personnel for experimentation as well as subsequent testing and analysis are all limiting factors that often make consideration of every potentially significant variable infeasible. Design of experiments minimizes the number of runs required to conduct an investigation without significantly compromising thoroughness, or can be used as a screening method to focus a study on variables that are identified as important.

Measurables

First, measurables to be considered are defined. Although surface film technology could potentially be used in many applications, automotive exterior body panels are the application of primary interest. Long glass fiber-reinforced polypropylene has been successfully implemented in many structural and semi-structural automotive applications¹³, and the static and dynamic mechanical properties for this material system with surface film have been studied in the literature^{23,24}. The primary interest of this study is to evaluate the technology as a candidate for enhancing the surface finish of LFT extrusion-compression

molded parts. Adhesion of surface film to LFT substrate, gloss and chipping resistance are identified as properties that are critical to deeming the technology viable for automotive exterior applications. These properties are defined as the measurables for the design of experiment.

Variables

Next, process variables potentially affecting the properties of interest are brainstormed. The list of potentially contributing factors is extensive, and the versatility of the extrusion-compression molding equipment allows the operator to vary many parameters such as temperature of the plasticator cylinder and compression mold, rotation speed of the cylinder, size of plasticator extrudate, placement location of the LFT extrudate within the mold, closing and opening speed of the mold, clamp tonnage and time the mold is closed.

Glass content of the LFT material is expected to contribute to adhesion of the film to the LFT substrate. Increasing glass content could limit the matrix available for melt bonding with the film, thereby decreasing interfacial strength. Temperature of the film prior to insertion in the mold is anticipated to affect interfacial adhesion and gloss. Preheating the film could promote melt bonding by softening the substrate layer of the film, but could potentially inhibit gloss by softening the protective top layer of the film. Time within the mold is expected to be significant to all of the variables considered, for reasons similar to those outlined for the film preheat temperature variable. Film thickness is likewise anticipated to be an important variable for all of the measurables established, an assertion supported by studies on in-mold film injection molding²³⁻²⁵. These four

variables, residence time in mold, glass content of LFT substrate, film preheat temperature and film thickness are selected for consideration in the design of experiment. The affect of these variables on the properties of interest will be characterized. In addition, it is evident that operating parameters must be optimized for each material system and film product individually. The design of experiment will serve the secondary purpose of establishing optimum operating parameters for the extrusion-compression molded glass-polypropylene and film systems considered with respect to interfacial adhesion, gloss and chipping resistance. The goals of the design of experiment in terms of responses and variables are summarized in Table 1.

Table 1: Design of Experiment Goals, Responses and Variables

Goals	Responses	Variables
Characterize effect of variables on responses Establish optimum operating parameters	Interfacial adhesion Gloss Chip resistance	Residence time in mold Glass content of LFT substrate Film preheat temperature Film thickness

Levels and Matrix Design

It was desired to consider three levels for each variable to thoroughly assess the significance of each on the measurables and to confidently establish optimum operating parameters. However, a full factorial experiment having four variables and three levels would comprise $3^4 = 81$ processing runs, hardly a reasonable amount of processing to conduct due to material, time and cost constraints.

Two film suppliers are considered in the study; one of the suppliers has a film of only one thickness, 1.70 millimeters, while the other supplier offers their

film in three thicknesses, 0.43, 0.76 and 1.52 millimeters. Therefore, the film thickness levels are limited to these options, although typically it is desired to have equal increments between levels. The films from the respective suppliers both have a thermoplastic olefin (TPO) backbone; however, the film constituents and structure are not the same and considering both in the same design of experiment could introduce additional variables and hinder the establishment of optimum operating parameters for each product. Therefore, separate designs of experiment are created for the two film suppliers.

Since the first film supplier has only one film thickness available, a design of experiment with only three variables is established for this product. A full factorial experiment with three variables and three levels still requires 27 processing runs. Therefore, the number of levels for each variable is reduced to two. However, three centerpoint runs are added to facilitate detection of any curvilinear effects²⁹, resulting in a design of experiment matrix with $2^3 + 3 = 11$ runs. The three variable, two level full factorial plus centerpoint design of experiment matrix for the first film supplier is outlined in standard design of experiment notation in Table 2.

Table 2: Design of Experiment Matrix for Film Supplier I

Standard Run Order	Variable A	Variable B	Variable C
1	-1	-1	-1
2	-1	-1	1
3	0	0	0
4	1	-1	-1
5	0	0	0
6	-1	1	-1
7	-1	1	1
8	0	0	0
9	1	1	1
10	1	-1	1
11	1	1	-1

A Box-Behnken design is selected for the second film supplier offering multiple film thicknesses. The Box-Behnken approach allows a manageable number of processing runs while still considering four variables and three levels each. This response surface method is efficient yet still allows potential quadratic effects to be considered³⁰. The design of experiment matrix established for the second film supplier is outlined in standard design of experiment notation in Table 3.

Table 3: Design of Experiment Matrix for Film Supplier II

Standard Run Order	Variable A	Variable B	Variable C	Variable D
1	0	0	0	0
2	-1	-1	0	0
3	1	-1	0	0
4	-1	1	0	0
5	1	1	0	0
6	0	0	-1	-1
7	0	0	1	-1
8	0	0	-1	1
9	0	0	1	1
10	-1	0	-1	0
11	1	0	-1	0
12	0	0	0	0
13	0	0	0	0
14	0	0	0	0
15	-1	0	1	0
16	1	0	1	0
17	0	-1	0	-1
18	0	1	0	-1
19	0	-1	0	1
20	0	1	0	1
21	-1	0	0	-1
22	1	0	0	-1
23	-1	0	0	1
24	1	0	0	1
25	0	-1	-1	0
26	0	1	-1	0
27	0	-1	1	0
28	0	1	1	0
29	0	0	0	0

The levels chosen for each of the variables are defined in Table 4. Note that the design of experiment for the first film supplier does not include variable D as only one film thickness of 1.70 mm is available.

Table 4: Variable and Level Definitions

Variable	Variable Description	Level -1	Level 0	Level 1
A	Residence Time in Mold [sec]	60	180	300
B	Glass Content [wt %]	10	30	50
C	Film Preheat Temperature [°C]	0	25	50
D	Film Thickness [mm]	0.43	0.76	1.52

Processing Runs

Typically, randomizing runs within the experimental matrix is recommended to minimize potential bias introduced during processing³¹. However, such randomization during processing is not entirely feasible for every variable. For instance, changing glass content during the extrusion-compression molding process is a time-consuming task. Further, the extruder must be completely cleaned out before introducing a new material cycle; therefore, repeatedly switching glass content would result in excessive material waste. It would also facilitate efficient processing to be able to conduct runs with the same film preheat temperature in concurrent fashion. Therefore, the two design of experiment matrices are combined and then batched in terms of glass content and film preheat temperature. The runs within batches are then randomized to minimize bias. The combined, batched and subsequently randomized design of experiment matrix is presented in standard design of experiment notation in Table 5 and in uncoded format in Table 6. When combined, the two design of experiment matrices comprise 40 runs. The original standard run order is included for reference, with "A" representing the runs originating from the Supplier I design of experiment and "B" representing the runs originating from the Supplier II design of experiment. The film thickness variable is left blank for the Film Supplier I runs as only one thickness is available and is not considered a variable in that design of experiment. During processing, runs are not duplicated due to material and equipment operation costs.

Table 5: Processing Matrix

Processing Run Order	Standardized Run Order	A	B	C	D
1	B1	0	0	0	0
2	B12	0	0	0	0
3	B13	0	0	0	0
4	B6	0	0	-1	-1
5	B11	1	0	-1	0
6	B8	0	0	-1	1
7	B10	-1	0	-1	0
8	A1	-1	-1	-1	
9	A4	1	-1	-1	
10	B25	0	-1	-1	0
11	A10	1	-1	1	
12	B27	0	-1	1	0
13	A2	-1	-1	1	
14	B17	0	-1	0	-1
15	B19	0	-1	0	1
16	B3	1	-1	0	0
17	B2	-1	-1	0	0
18	B24	1	0	0	1
19	B29	0	0	0	0
20	B14	0	0	0	0
21	A3	0	0	0	
22	B21	-1	0	0	-1
23	B23	-1	0	0	1
24	B5	1	1	0	0
25	B20	0	1	0	1
26	B4	-1	1	0	0
27	B18	0	1	0	-1
28	A11	1	1	-1	
29	B26	0	1	-1	0
30	A6	-1	1	-1	
31	A7	-1	1	1	
32	B28	0	1	1	0
33	A9	1	1	1	
34	B16	1	0	1	0
35	B9	0	0	1	1
36	B15	-1	0	1	0
37	B7	0	0	1	-1
38	B22	1	0	0	-1
39	A5	0	0	0	
40	A8	0	0	0	

Table 6: Uncoded Processing Matrix

Processing Run Order	Standardized Run Order	Residence Time in Mold [sec]	Glass Content [wt %]	Film Preheat Temperature [°C]	Film Thickness [mm]
1	B1	180	30	25	0.76
2	B12	180	30	25	0.76
3	B13	180	30	25	0.76
4	B6	180	30	0	0.43
5	B11	300	30	0	0.76
6	B8	180	30	0	1.52
7	B10	60	30	0	0.76
8	A1	60	10	0	
9	A4	300	10	0	
10	B25	180	10	0	0.76
11	A10	300	10	50	
12	B27	180	10	50	0.76
13	A2	60	10	50	
14	B17	180	10	25	0.43
15	B19	180	10	25	1.52
16	B3	300	10	25	0.76
17	B2	60	10	25	0.76
18	B24	300	30	25	1.52
19	B29	180	30	25	0.76
20	B14	180	30	25	0.76
21	A3	180	30	25	
22	B21	60	30	25	0.43
23	B23	60	30	25	1.52
24	B5	300	50	25	0.76
25	B20	180	50	25	1.52
26	B4	60	50	25	0.76
27	B18	180	50	25	0.43
28	A11	300	50	0	
29	B26	180	50	0	0.76
30	A6	60	50	0	
31	A7	60	50	50	
32	B28	180	50	50	0.76
33	A9	300	50	50	
34	B16	300	30	50	0.76
35	B9	180	30	50	1.52
36	B15	60	30	50	0.76
37	B7	180	30	50	0.43
38	B22	300	30	25	0.43
39	A5	180	30	25	
40	A8	180	30	25	

CHAPTER VI

TESTING

ASTM test methods are employed to evaluate adhesion of the surface film to the LFT substrate panel, gloss and chipping resistance. Testing was performed by the University of Dayton Research Institute (UDRI)'s Coatings Laboratory.

Adhesion of Surface Film to LFT Substrate Panel

ASTM D4541: Standard Test Method for Pull-off Strength of Coatings using Portable Adhesion Testers is selected for characterizing the interfacial adhesion of the experimental panels. Test specifications can vary amongst industries and applications; however, the test method and conditions employed are representative of approaches referenced within the automotive industry literature³². The adhesion tester used is a SEMicro model PATTI-110, exhibited in Figure 9.

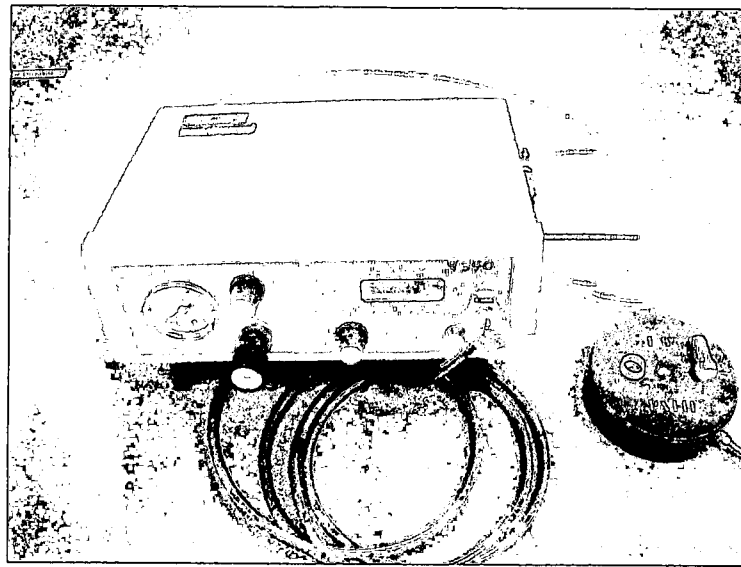


Figure 9: Portable Adhesion Tester

The specimens tested were approximately four-inch by four-inch flat panel samples. Five such samples for each of the experimental runs were considered to establish an average tensile pull-off strength for each of the 40 panels.

The specimens were prepared by milling a ring down to the substrate with the inside diameter of the ring being equal to the outside diameter of the stud. After milling the ring, the surface of the test area was lightly abraded with 240 grit sand paper to enhance adhesion of the stud to the specimen surface, as illustrated in Figure 10. Representative specimens were provided to UDRI in advance to facilitate proper adhesive selection.

The test apparatus uses compressed air to apply a perpendicular load to the pull stud. Failure occurs in the weakest layer of the test specimen.

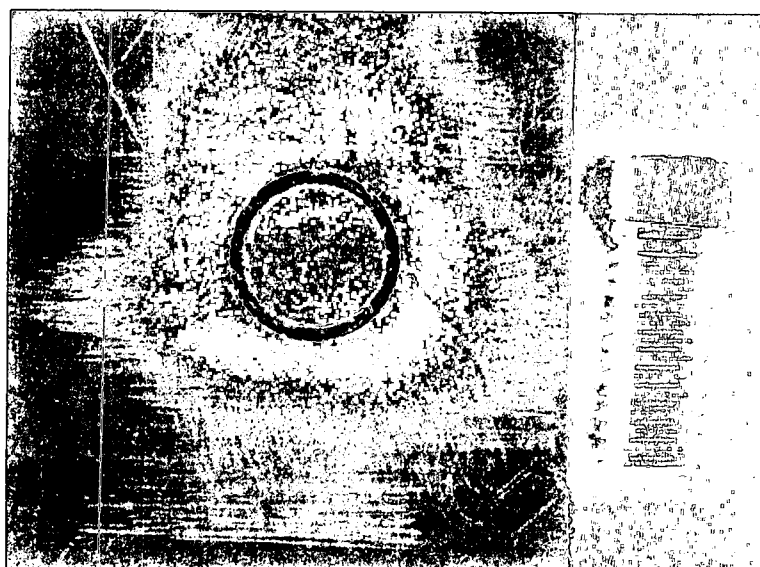


Figure 10: Milled Ring with Pull-off Stud; Abraded Test Area

Gloss

Gloss was measured at 60° in accordance with ASTM D523: Standard Test Method for Specular Gloss using a BYK Gardner model 4528 micro-TRI-gloss meter, exhibited in Figure 11.

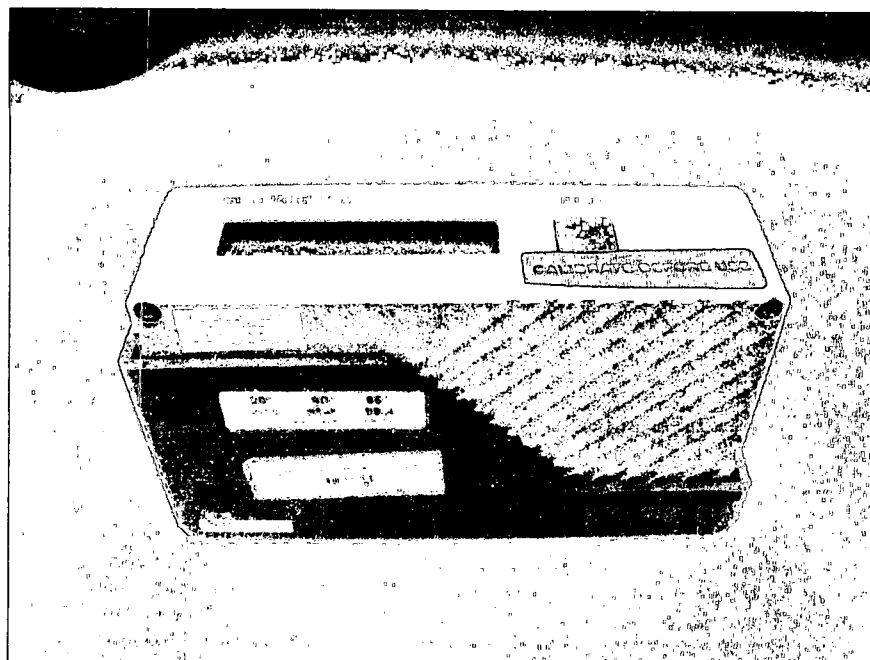


Figure 11: Gloss Meter

Gloss is measured on a scale of 0 to 100 with 100 being a surface with 100 percent gloss and 0 being a gloss free surface. Each specimen is measured for gloss by taking four measurements with a 90° rotation of the gloss meter after each measurement, and then averaging the four measurements. The gloss of each panel is calculated as the average gloss of the eight specimens from that panel.

Chipping Resistance

Chipping resistance is measured in accordance with ASTM D3170: Standard Test Method for Chipping Resistance of Coatings using a Q-Panel model MTG gravelometer, exhibited in Figure 12.

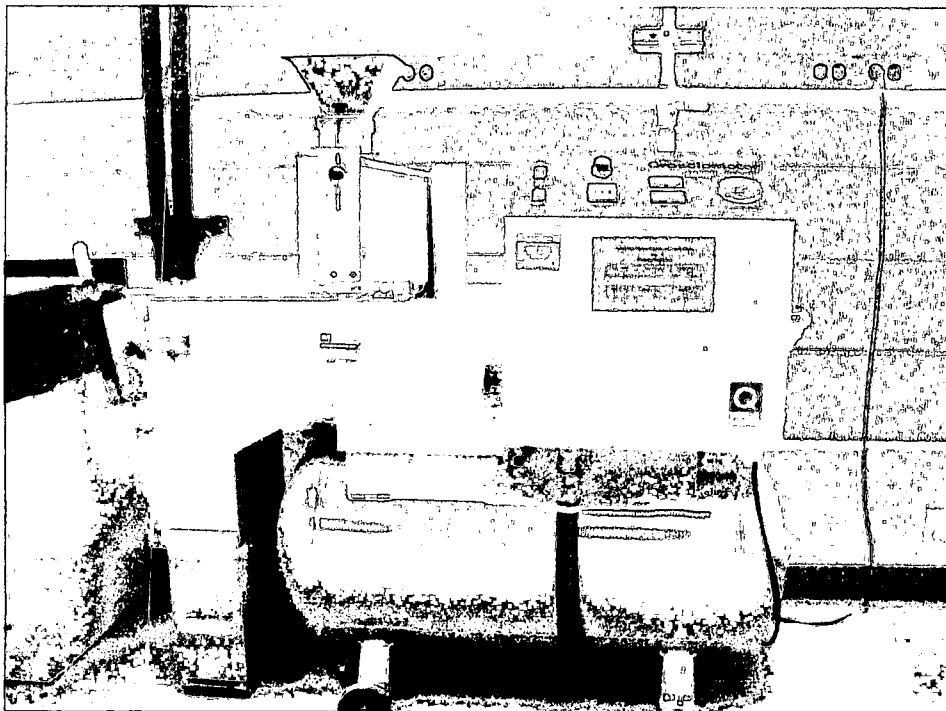


Figure 12: Gravelometer

Three four-inch by six-inch specimens from each panel are provided for chipping resistance testing. The chipping resistance is reported as a three-part

visual rating in terms of the number of chips, the size of the chips, and observed failure mode. The rating system for the number of chips is outlined in Table 7, and the rating system for chip size is provided in Table 8.

Table 7: Number of Chips Rating System

Rating	Number of Chips
10	0
9	1
8	2 – 4
7	5 – 9
6	10 – 24
5	25 – 49
4	50 – 74
3	75 – 99
2	100 – 149
1	150 – 250
0	> 250

Table 8: Chip Size Rating System

Rating	Chip Size
A	< 1 mm
B	1 – 3 mm
C	3 – 6 mm
D	> 6 mm

CHAPTER VII

DATA

Adhesion of Surface Film to LFT Substrate Panel

Failure mode is defined as the place within the specimen structure where separation of stud from the specimen occurred. The failure modes observed during testing are listed in Table 9 and illustrated in Figure 13. Examples of all four failure modes for specimens with films from both suppliers are exhibited in Appendix A. Failure Modes 3 and 4 are not observed in panels with films from the first supplier.

Table 9: Adhesion Testing Failure Modes

Failure Mode	Description
1	Adhesive failure of the stud adhesive to the surface of the specimen
2	Adhesive failure of the top colored layer to the TPO layer
3	Adhesive failure of the TPO layer to the LFT substrate panel
4	Cohesive failure within the LFT substrate panel

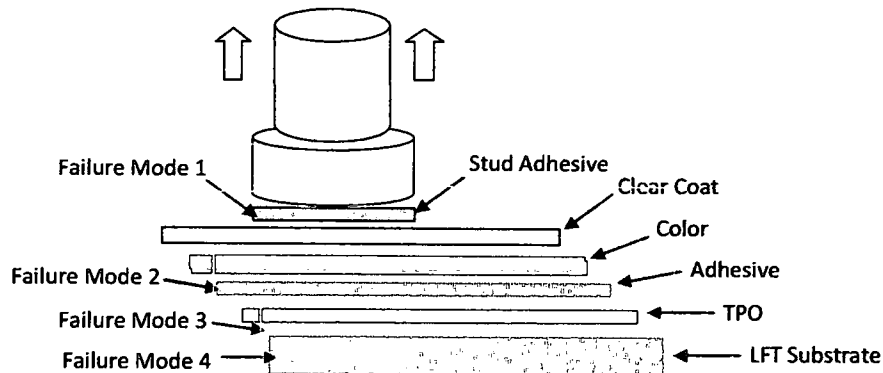


Figure 13: Adhesion Testing Failure Modes

The pull-off testing data is tabulated in Appendix B. A summary of results is outlined in Table 10. In some cases, different specimens from the same panel exhibited different failure modes, and where this occurred the data is reported in terms of the predominate failure mode and the secondary failure mode. When the only failure mode was failure of the stud adhesive, the tensile strength of the composite system is unknown, and the data is reported as being greater than the tensile strength of the largest value for the failed stud adhesive.

Table 10: Summary of Tensile Pull-off Test Results

Panel ID	Predominate Failure Mode	Pull-off Tensile Strength [psi]	Std Dev [psi]	Secondary Failure Mode	Pull-off Tensile Strength [psi]	Std Dev [psi]
1	2	618	133	None		
2	2	830	24.7	None		
3	2	754	75.4	None		
4	2	744	58.9	None		
5	2	760	71.7	None		
6	2	665	89.5	4	491	SDP
7	2	665	89.5	4	418	SDP
8	1	> 557		None		
9	1	> 589		None		
10	2	731	54.4	None		
11	1	> 614		None		
12	1	> 671		None		
13	1	> 540		None		
14	2	764	55.8	None		
15	3	479	128	2	753	34.6
16	2	721	53.8	None		
17	2	746	82.1	3	601	
18	2	740	53.9	None		
19	2	739		None		
20	2	755	77.5	None		
21	1	> 499		None		
22	2	737	84.3	None		
23	2	857	72.6	3	650	75
24	2	780	14.3	4	424	228
25	4	385	15.3	None		
26	2	779	24.6	4	414	SDP
27	1	> 426		None		
28	1	> 585		None		
29	4	601	144	3	732	SDP
30	1	> 520		4	442	SDP
31	1	> 569		None		
32	2	728	98.1	4	524	208
33	1	> 520		4	422	SDP
34	1	> 785		2	850	SDP
35	2	773	96.2	4	716	SDP
36	2	819	76.7	None		
37	2	823	57.0	None		
38	2	765	86.6	3	748	SDP
39	1	> 520		None		
40	1	> 483		None		

Optical analysis is performed on selected panels to confirm melt bonding between the LFT substrate and TPO film layer. A representative micrograph is shown in Figure 14, with a close-up view of the melt bond layer in Figure 15.

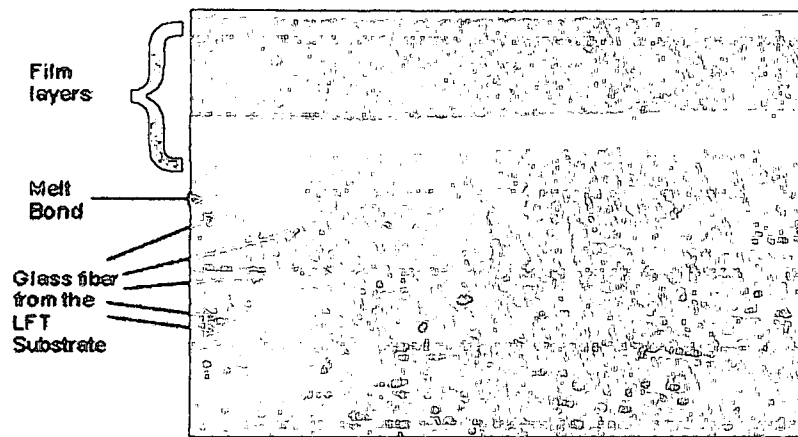


Figure 14: Micrograph of Interfacial Melt Bond



Figure 15: Close-up View of Representative Melt Bond Images

Gloss

All raw data from the gloss testing can be found in Appendix B, where each measurement represents an average of four readings. A summary of gloss results is provided in Table 11.

Table 11: Summary of Gloss Test Results

Panel ID	Average 60° Gloss	Standard Deviation	Panel ID	Average 60° Gloss	Standard Deviation
1	80.1	0.9	21	87.2	0.4
2	83.6	0.7	22	80.6	0.7
3	80.6	1.0	23	86.1	1.0
4	32.2	6.8	24	75.9	1.4
5	79.4	0.9	25	84.0	1.3
6	83.6	1.3	26	83.2	1.1
7	62.0	1.7	27	70.3	1.6
8	87.5	0.6	28	87.5	0.9
9	86.7	0.5	29	79.1	1.5
10	48.6	1.0	30	85.4	0.4
11	83.2	1.2	31	83.6	1.0
12	82.2	0.3	32	84.4	0.4
13	85.5	0.7	33	83.6	0.8
14	85.1	0.7	34	83.9	0.7
15	86.3	0.6	35	84.8	0.8
16	78.9	0.8	36	85.6	0.6
17	84.6	0.7	37	80.6	1.8
18	83.1	0.6	38	59.2	7.9
19	77.1	1.1	39	86.2	0.5
20	74.9	2.0	40	85.5	0.6

Chipping Resistance

No chipping is observed on any of the specimens; however, there is severe marring on the surface of each specimen. While marring is not a criterion for this test, an attempt is made to quantify the marring using the chipping rating scales defined in Tables 7 and 8. The marring observed is essentially the same for each panel tested with only minor differences in the number of mars detected. In all cases the observed marring occurs within the clear topcoat layer of the test specimen. Figures 16 and 17 exhibit representative images of typical marring for each of the two film suppliers. The marring ratings for each of the three four-inch by six-inch specimens from all 40 panels are presented in Table 12.

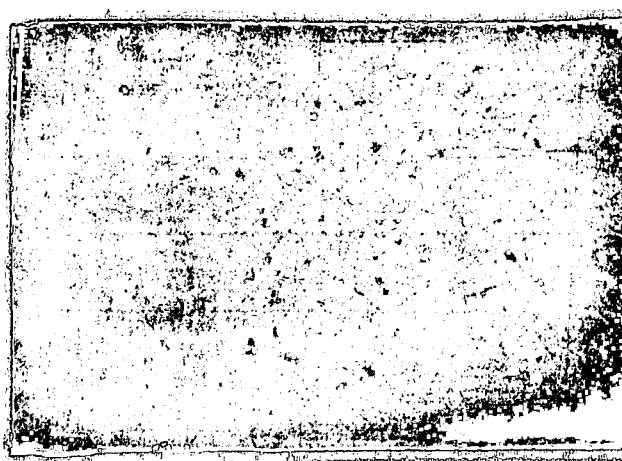


Figure 16: Representative Marring on Supplier I Films



Figure 17: Representative Marring on Supplier II Films

Table 12: Marring Rating Data

Specimen ID	Number Rating	Size Rating	Failure Mode Rating	Specimen ID	Number Rating	Size Rating	Failure Mode Rating
1F	2	B	T	21F	1	B	T
1G	2	B	T	21G	2	B	T
1H	2	B	T	21H	2	B	T
2F	1	B	T	22F	3	B	T
2G	1	B	T	22G	1	B	T
2H	1	B	T	22H	2	B	T
3F	1	B	T	23F	2	B	T
3G	1	B	T	23G	2	B	T
3H	1	B	T	23H	2	B	T
4F	1	B	T	24F	1	B	T
4G	2	B	T	24G	2	B	T
4H	1	B	T	24H	2	B	T
5F	2	B	T	25F	2	B	T
5G	2	B	T	25G	1	B	T
5H	1	B	T	25H	1	B	T
6F	1	B	T	26F	1	B	T
6G	1	B	T	26G	1	B	T
6H	1	B	T	26H	3	B	T
7F	1	B	T	27F	2	B	T
7G	2	B	T	27G	2	B	T
7H	1	B	T	27H	1	B	T
8F	1	B	T	28F	2	B	T
8G	1	B	T	28G	2	B	T
8H	1	B	T	28H	2	B	T
9F	1	B	T	29F	3	B	T
9G	2	B	T	29G	3	B	T
9H	1	B	T	29H	1	B	T
10F	3	B	T	30F	1	B	T
10G	2	B	T	30G	2	B	T
10H	1	B	T	30H	1	B	T
11F	1	B	T	31F	1	B	T
11G	1	B	T	31G	1	B	T
11H	1	B	T	31H	1	B	T
12F	1	B	T	32F	2	B	T
12G	1	B	T	32G	1	B	T
12H	2	B	T	32H	2	B	T
13F	1	B	T	33F	1	B	T
13G	1	B	T	33G	1	B	T
13H	2	B	T	33H	1	B	T
14F	1	B	T	34F	1	B	T
14G	2	B	T	34G	2	B	T
14H	2	B	T	34H	2	B	T
15F	2	B	T	35F	1	B	T
15G	2	B	T	35G	1	B	T
15H	1	B	T	35H	2	B	T
16F	1	B	T	36F	1	B	T
16G	2	B	T	36G	2	B	T
16H	1	B	T	36H	2	B	T
17F	2	B	T	37F	1	B	T
17G	1	B	T	37G	1	B	T
17H	1	B	T	37H	1	B	T
18F	1	B	T	38F	3	B	T
18G	1	B	T	38G	1	B	T
18H	2	B	T	38H	1	B	T
19F	2	B	T	39F	1	B	T
19G	2	B	T	39G	1	B	T
19H	1	B	T	39H	1	B	T
20F	2	B	T	40F	2	B	T
20G	3	B	T	40G	2	B	T
20H	2	B	T	40H	2	B	T

CHAPTER VIII

ANALYSIS

Statistical analysis is performed on the adhesion, gloss and chipping resistance testing to characterize the significance of each of the variables on these properties. Results are further used to establish optimum operating parameters for film products from both suppliers.

Film Supplier I

All pull-off tests on films from the first supplier failed in the adhesive between the stud of the test apparatus and the surface film. This indicates that unfortunately the stud adhesive chosen is weaker than the bonds within all layers of the test specimen, disallowing characterization of the test specimen pull-off tensile strengths. Only two specimens did not fail in the stud adhesive, one specimen from run 30 and one from run 33. These two specimens exhibited Failure Mode 4, cohesive failure within the LFT substrate panel, at 442 and 422 pounds per square inch (psi), respectively. The average value of stud adhesive failure indicates that the pull-off tensile strength of the panels with films from Supplier I is typically at least greater than 545 psi. However, characterization of the affect of the design of experiment variables on film adhesion properties and

optimization of operating parameters to maximize adhesion of film to substrate cannot be established due to the limited success of the testing.

In general, surface films for automotive exterior bumper fascia are preferred to hold to at least 15 psi in a 90° pull-off test on a one inch sample at 70°F³². Under similar test conditions, the film Supplier I specimens burst at an average of 11.2 psi, with individual values ranging from 6.8 to 15.1 psi. Due to the fact that observed failure was within the stud adhesive, it is reasonable to assert that optimized operating parameters and proper adhesive selection for this film product will easily meet or exceed the industry standard for this property requirement.

The gloss data from the panels with films from Supplier I is considered in an analysis of variance (ANOVA) which included main effects, two and three variable interactions as well as a curvature term. Using a 95% level of significance, only film preheat temperature (variable C) is statistically significant. Figure 18 shows the main effects plots for the three variables with respect to the gloss response. Variables A and B, residence time in mold and glass content of LFT substrate, have negligible effect. Also exhibited in Figure 18 is a plot of the mean gloss for each of the 11 runs from the Film Supplier I design of experiment, presented in standard run order as defined in Table 1. No variable interactions are found to be significant.

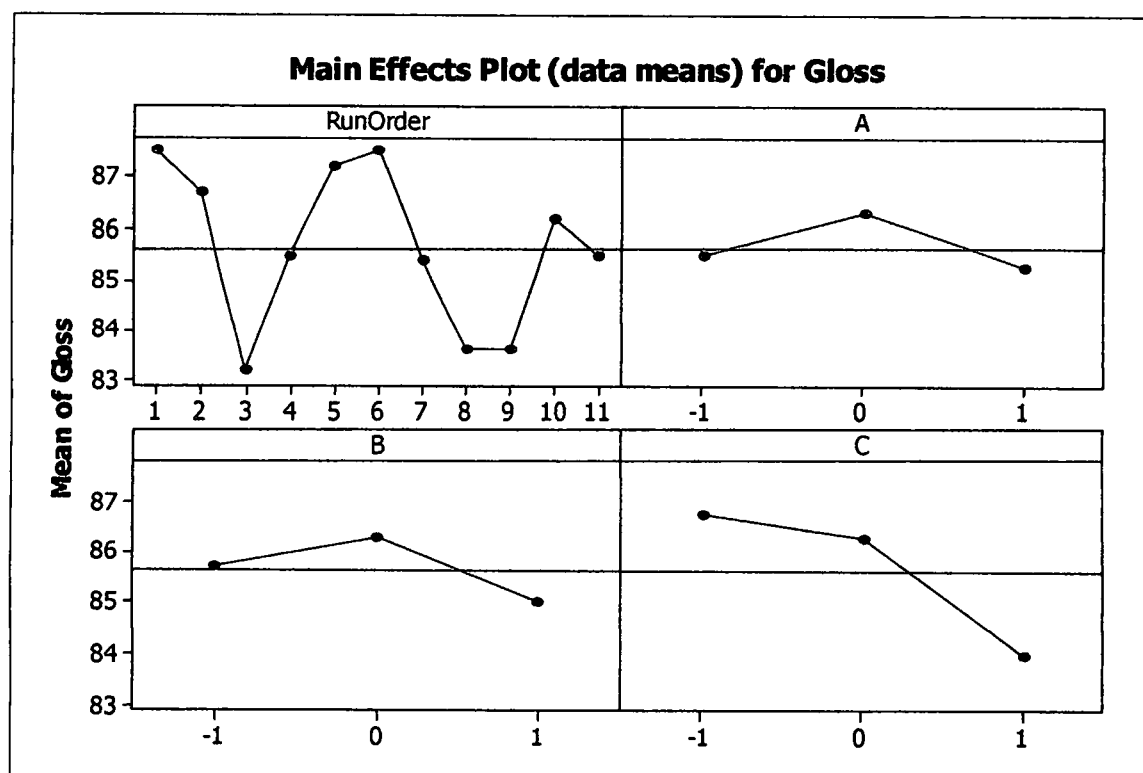


Figure 18: Main Effects Plots for Film Supplier I Gloss Response

Freezing the film to 0°C prior to molding is observed to yield the best gloss, although feasibility of implementing this pre-process step on the industrial scale has yet to be determined. The gloss rating is found to decrease with increasing film preheat temperature. Intuitively, it makes sense that the gloss of the protective topcoat of the film could be degraded with increasing temperature, although the constituents of the multiple film layers are not disclosed. However, it may not be prudent to state that freezing the film prior to molding would be optimum in a more comprehensive sense when other properties are considered. Although the Film Supplier I adhesion data is not meaningful for statistical analysis, it is likely that film preheat temperature may affect gloss and adhesion of the film to the substrate panel in conflicting manners. There is likely a compromising film preheat temperature that would be optimum for the

requirement of attaining both property specifications, promoting interfacial adhesion via melt bonding while also minimizing degradation in gloss performance.

The highest gloss values observed for Film Supplier I are in the high 80's range of the zero to 100 gloss rating scale. Requirements for interior in-mold film thermoplastic components can be as low as the five to 15 gloss unit range³³; however, exterior automotive and aerospace applications often have a minimum requirement of 90 gloss units while the majority have more stringent specifications in the mid 90's range³⁴. A select few manufacturers have attempted to divert consumers from high-gloss finish expectations, as demonstrated by the textured thermoplastic exterior of the Th!nk City Electric Car³⁵. However, while some end-users may be willing to compromise cosmetic properties for environmentally friendly products, wide acceptance and use of alternative body panel technologies will almost certainly rely on the ability of these technologies to attain existing gloss specifications.

Although no chipping is observed on any specimens according to the ASTM definition, severe marring is observed on all of the specimens upon chip resistance testing with the gravelometer. The marring is consistent amongst all panels, with no significant variations in the number of marring incidences, size of such defects or their location within the multilayered structure. All marring is observed in the topcoat of the film structure for each specimen.

Film Supplier II

An ANOVA considering main effects, two variable interactions and quadratic effects is performed for the four variable design of experiment for Film Supplier II. A 95% level of significance is used for variable discrimination. Results for the adhesion data indicate that film preheat temperature (variable C) and film thickness (variable D) are significant to this response. Figure 19 is a plot of these significant main effects. No interactions are statistically significant.

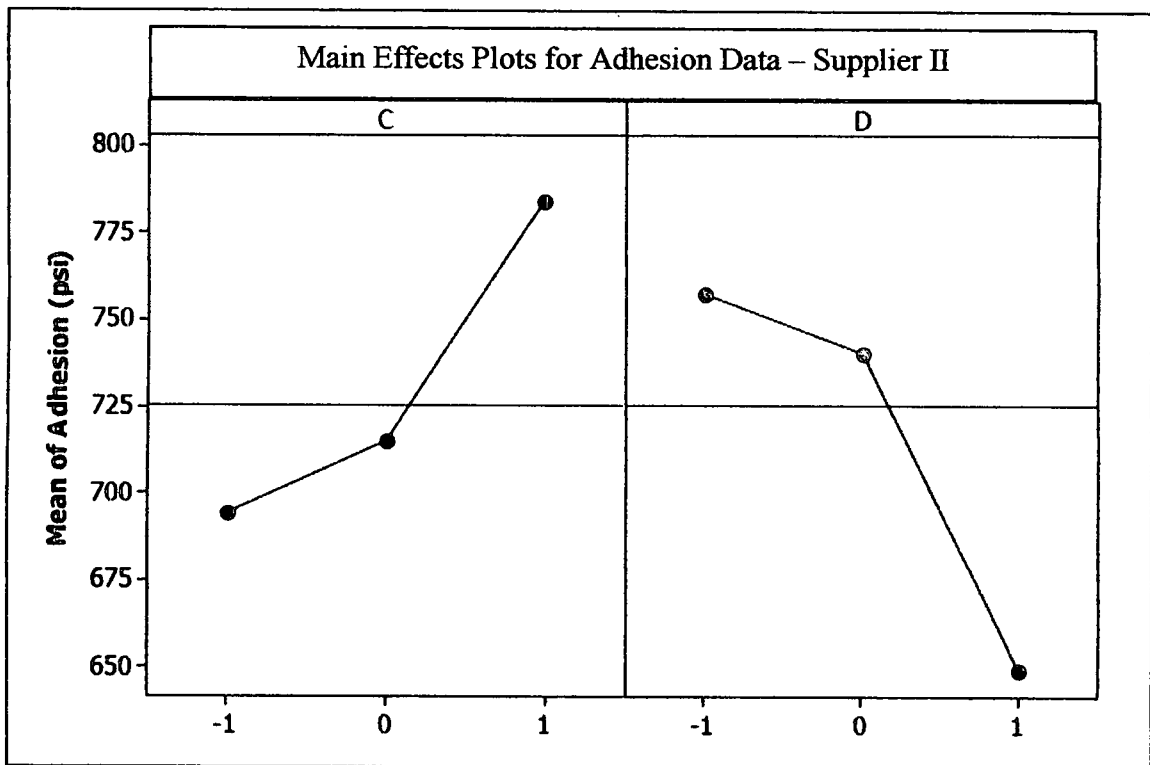


Figure 19: Significant Main Effects Plots for Supplier II Adhesion Response

Highest film preheat temperature and lowest film thickness yield optimum pull-off tensile strength for Film Supplier II. Both results are intuitively consistent with anticipated results. Higher film preheat temperature facilitates melt bonding between the TPO film backing and LFT substrate. Thinner films further promote melt bonding by maximizing heat transfer from the mold to the inner

TPO/substrate interface by decreasing the distance the heat must be transferred. On the other hand, it is also anticipated that thicker films could potentially increase interfacial adhesion by increasing pressure of the substrate against the film due to smaller volume for the extrudate to expand within; however, this result appears to be insignificant or less dominate than that introduced by the enhanced heat transfer. The average burst strength of the panels with the thinnest film and highest preheat temperature is over 20 psi, exceeding the industry standard for exterior bumper fascia³².

For the gloss response, film preheat temperature (variable C), film thickness (variable D) and their interaction (C*D) are found to be significant. Using a least squares fit for these coded variables, the gloss level as a function of film preheat temperature and film thickness is established in Equation 1.

$$\text{Gloss Level} = 77.2 + 9.72 (C) + 8.33 (D) - 11.8 (C*D) \quad (1)$$

The main effects plots and interactions for the gloss response are shown in Figures 20 and 21, respectively.

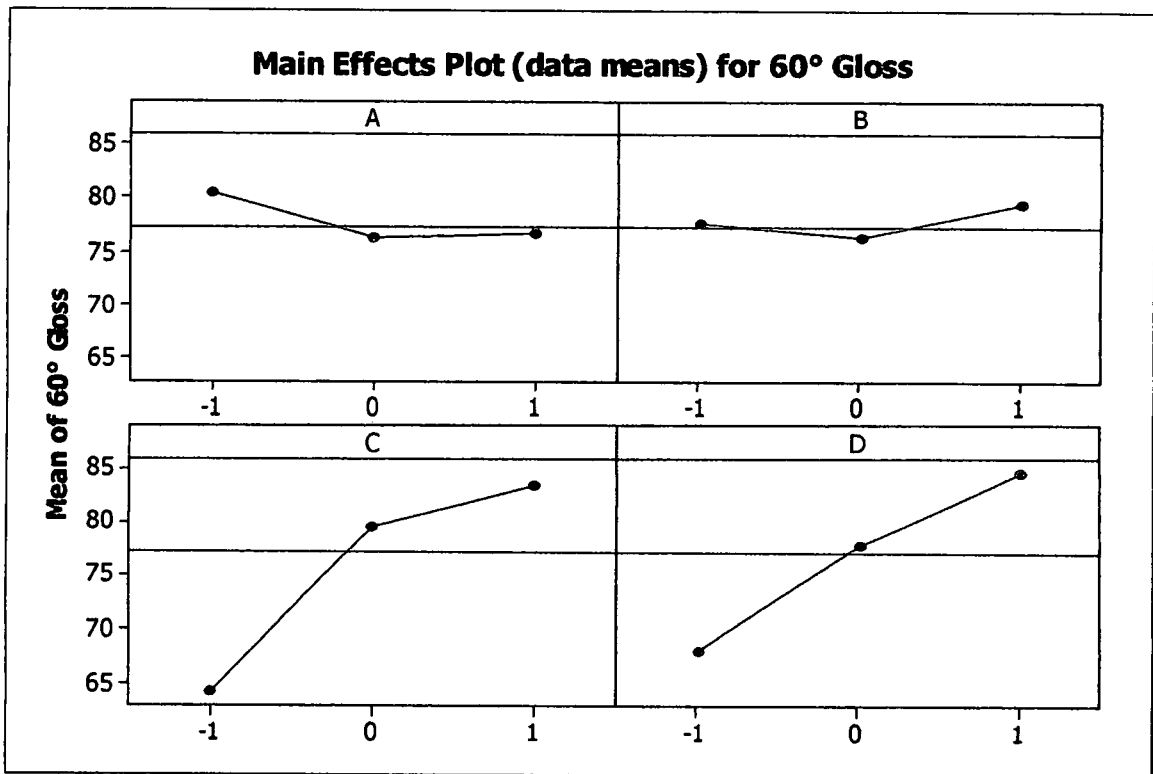


Figure 20: Main Effects Plots for Film Supplier II Gloss Response

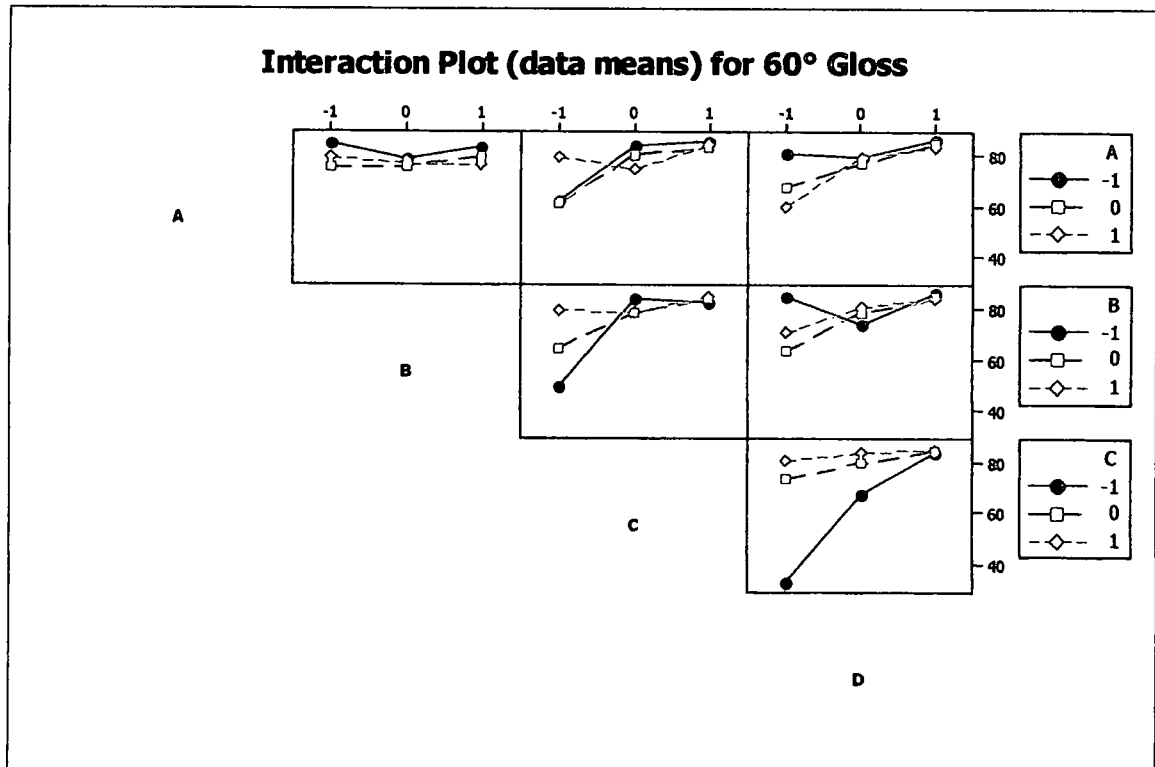


Figure 21: Variable Interactions for Film Supplier II Gloss Response

As can be seen in Figure 20, the main effect plot for variable C indicates that the highest film preheat temperature yields superior gloss rating, an opposite relationship from that observed for Film Supplier I. The thickest film, 1.52 millimeters, is recommended for maximizing Supplier II gloss. It should be noted that the thickest Supplier II film is still thinner than the 1.72 millimeter Supplier I film.

The fact that the highest film preheat temperature yields the best gloss is somewhat counter-intuitive. A possible explanation could be that the topcoat is a thermoset material which cures with increasing temperature, therefore making the higher film preheat temperature favorable for gloss rating. However, the interaction of this variable with the thickness variable could lead to a different overall conclusion. The gloss response as a function of film preheat temperature and thickness established in Equation 1 is maximized within the range of the levels considered for each variable using Microsoft Excel Solver. Here, the lowest film preheat temperature and the thickest film are found to yield the maximum gloss rating. The interaction between the film preheat temperature and thickness variables leads to the ultimate identification of lowest film preheat temperature in conjunction with the thickest film as optimum for gloss rating, and indicates that the interaction of the temperature and thickness has a stronger effect than the temperature alone. Using the Solver analysis, it is determined that if Supplier II did offer a 1.70 millimeter thick film, this would be the optimum thickness and the optimum film preheat temperature would still be the lowest level. These results corroborate the conclusions drawn from the experimental

data from Supplier I. The best Supplier II gloss results are also in the high 80's range of the gloss of the gloss scale, indicating that development work is required to improve gloss characteristics for some applications.

As with Film Supplier I, no chipping is observed on the Film Supplier II panels upon gravelometer testing; however, severe marring is apparent. Again, the amount, size and locations of the marring are consistent amongst the Film Supplier II panels. Obviously, a much more robust topcoat will need to be demonstrated before use in automotive exterior body panels can be considered viable.

CHAPTER IX

SUMMARY AND CONCLUSIONS

Interfacial adhesion, gloss and chip resistance results are encouraging for consideration of long fiber thermoplastic extrusion-compression molding with in-mold film technology as viable for semi-structural and structural applications requiring enhanced surface finish characteristics. Although adhesion results from Film Supplier I are inconclusive due to weak stud adhesive selection, pull-off testing data from Film Supplier II demonstrates the ability of extrusion-compression molded LFT panels with surface films to attain industry pull-off testing requirements. Gloss properties exhibited here indicate that a gloss unit increase of ten to 15 percent will be required for insertion for automotive exterior applications. Although extensive marring was observed on all panels as a result of chip resistance testing, the gravelometer exposure represents more damaging conditions than may be considered criteria for many applications. Improvements in topcoat formulation are expected to improve gloss and marring resistance properties to meet product specification requirements.

The effects of the material and process variables on the properties considered are characterized. Residence time in mold and glass content of the LFT material do not have a statistically significant effect on any of the three responses with the levels investigated. However, glass content will affect the

structural properties of the LFT components, as characterized in literature. Film preheat temperature and film thickness are both found to significantly affect gloss and adhesion properties. Lower film preheat temperature and thicker films are found to enhance gloss, while lower film preheat temperature and thinner films promote interfacial adhesion.

Optimum operating parameters for each film supplier are also established. For Film Supplier I with only one film thickness, using the lowest film temperature level, 0°C, is best for the gloss rating of the levels considered. This indicates the top layer of the film consists of a material that experiences reduction in gloss value with increasing temperature. However, the feasibility of implementing such a processing requirement on a production capacity will have to be considered. Adhesion data for Film Supplier I is inconclusive due to weak adhesive employed during testing, although it is suspected that lower film preheat temperatures may not also be the best parameter for promoting interfacial adhesion.

For Film Supplier II, gloss readings ultimately indicate that selecting the lowest film preheat temperature, 0°C, and thickest film, 1.52 millimeters, yield the most desirable gloss results when the interaction between the two variables is considered. Film thickness is the dominant of the two significant main effects, likely due to the fact that the increased thickness insulates the glossy surface of the film from the molten LFT extrudate. The adhesion main effects for Film Supplier II support using the highest level for film preheat temperature, 50°C, in conjunction with the lowest level for film thickness, 0.43 millimeters. As expected, the operating parameters recommended for adhesion are the opposite

of those recommended with respect to gloss. It has been demonstrated that the Supplier II films have the ability to exceed industry pull-off testing standards, while the gloss will have to be improved at least ten to fifteen percent. In an effort to achieve gloss requirements, adhesion may have to be compromised to a value that may not be the maximum attainable yet still meets industry standards. This compromise will have to be established based on the specific requirements for each component.

In-mold film technology can be applied to LFT extrusion-compression molded materials to enhance their surface finish. Developing the technology for this process will lead to the use of extrusion-compression molded LFT materials for semi-structural and structural applications requiring large parts to be produced at high volume. The extensive benefits of this technology over alternatives such as steel, painted LFT, unreinforced plastics and injection molded thermoplastics with surface films will drive the development of in-mold film LFT extrusion-compression molding at a rapid pace. The films considered here have exhibited limitations that can be and already are being addressed by industry to facilitate application development³⁶.

CHAPTER X

FUTURE WORK AND RECOMMENDATIONS

Material optimization or engineering of a new top coat is one of the most important needs realized here for the film products considered. Marring resistance and high gloss, as well as scratch and chemical resistance, will need to be significantly improved and demonstrated before in-mold film technology will be viable for applications such as exterior automotive body panels where exposure to unfavorable weather conditions as well as gravel and debris are almost an everyday occurrence.

In addition, repeating this design of experiment with duplicate runs to verify results is recommended. Performing a narrowed design of experiment to further optimize operating levels for significant variables could also be beneficial. Mechanical testing to confirm structural properties for each material system considered will be required.

Applying the technology to complex geometries will be necessary for automotive parts such as bumpers and other commercial opportunities such as helmets. Designing a compression mold where the film can be placed in the mold prior to and underneath the LFT extrudate, as opposed to centering the mold on top of the extrudate as required with existing equipment considered here, will be desirable even for flat parts. Thermoforming the film to the

application shape prior to insertion in the mold has been demonstrated as a successful approach to applying in-mold film technology to complex geometries for other processing methods, and can be adapted to extrusion-compression molding.

The multi-layered film approach appears to be widely accepted as a viable method to address the varied property requirements of many applications. However, although some general trends have been established, it seems apparent that specific processing operating parameters will need to be optimized individually for each material system, process and film product considered.

Despite the material and process optimization needs outlined here, in-mold film technology coupled with LFT extrusion-compression molding is certainly a promising technology for semi-structural and structural applications, small and large, requiring high volume production capacity and enhanced surface finish properties. Implementing the technology for mass-scale automotive applications will reduce emissions by eliminating VOC-intensive paint lines as well as reducing fuel consumption by minimizing weight. Further, recyclability of the associated materials will drastically reduce the waste introduced every year by retired vehicles. These goals have been demonstrated tangible and will continue to drive LFT-ECM in-mold film technology development efforts.

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APPENDIX A: PULL-OFF TESTING FAILURE MODES

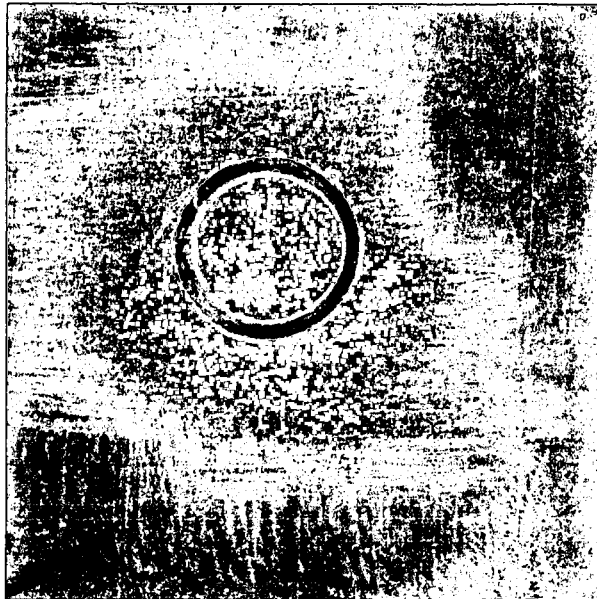


Figure A1: Representative Failure Mode 1 for Film Supplier I

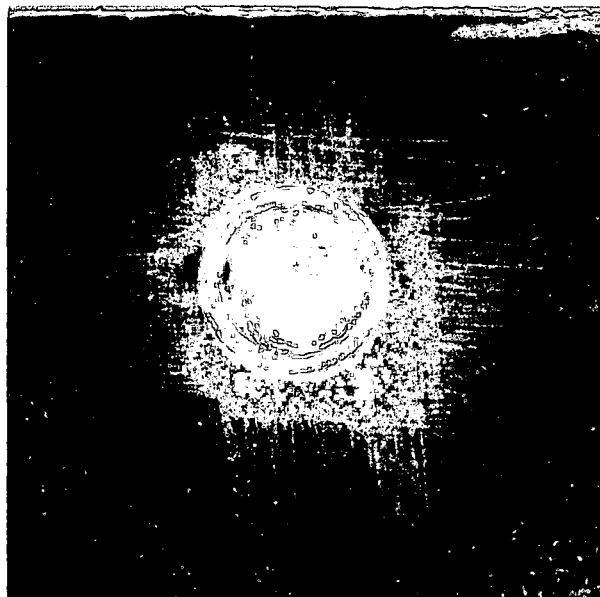


Figure A2: Representative Failure Mode 1 for Film Supplier II

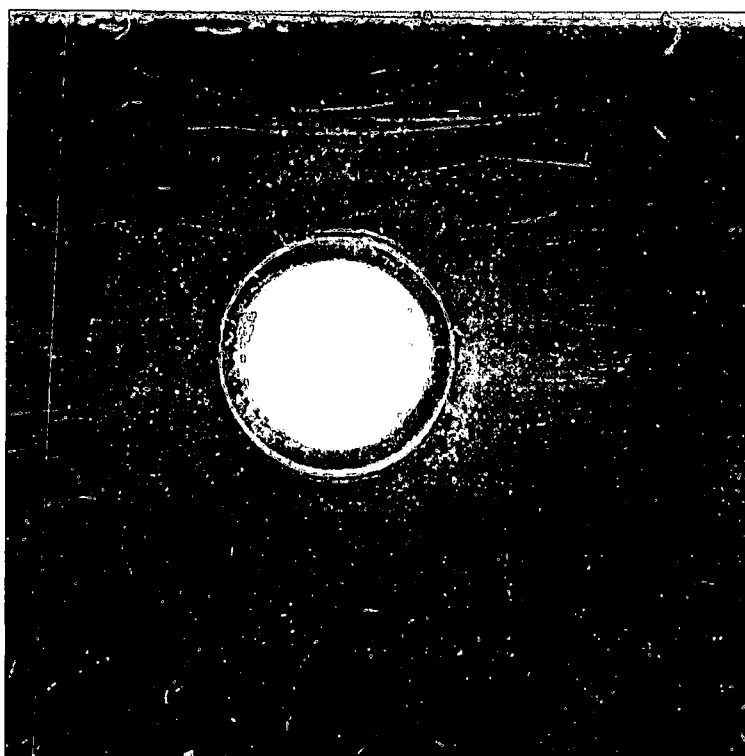


Figure A3: Representative Failure Mode 2 for Film Supplier II



Figure A4: Representative Failure Mode 3 for Film Supplier II

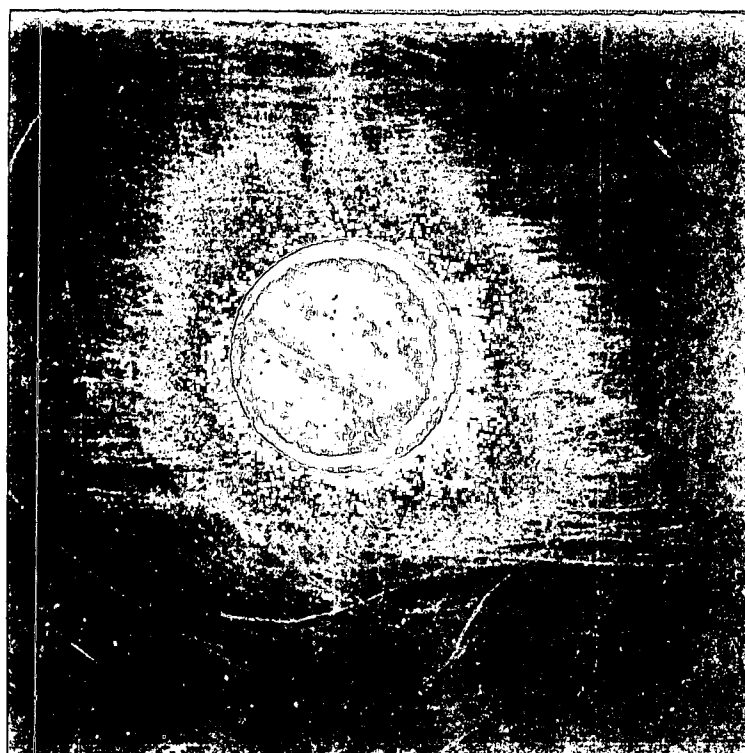


Figure A5: Representative Failure Mode 4 for Film Supplier I

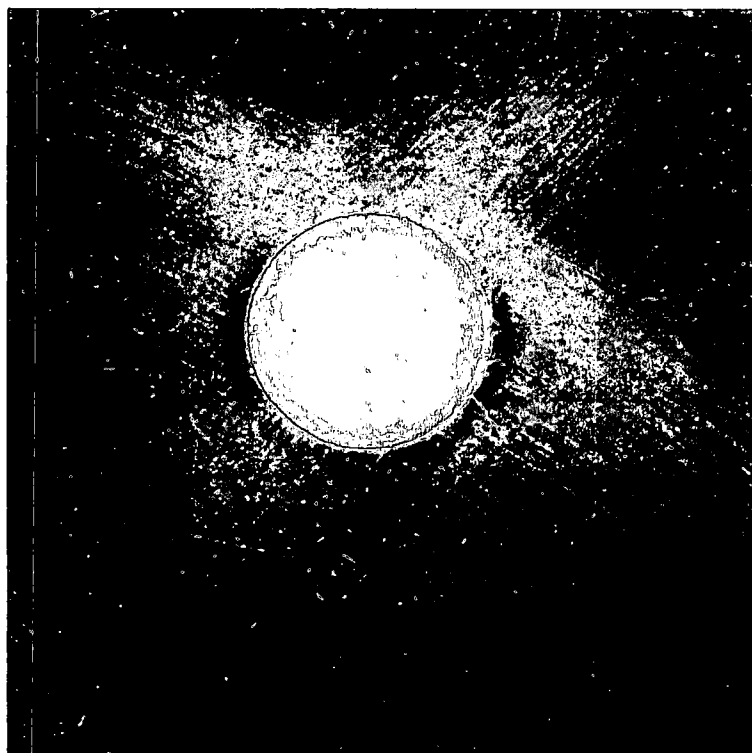


Figure A6: Representative Failure Mode 4 for Film Supplier II

APPENDIX B: PULL-OFF TENSILE STRENGTH DATA

Table B1: Pull-off Tensile Strength Data for Panels 1 – 20

Specimen ID	Burst Pressure [psi]	Failure Mode	Pull-off Tensile Strength [psi]	Specimen ID	Burst Pressure [psi]	Failure Mode	Pull-off Tensile Strength [psi]
1A	12.5	1	508	11A	11.7	1	475
1B	12.9	2	524	11B	15.1	1	614
1C	17.5	2	712	11C	11.8	1	479
1D	4.2	1	169	11D	13.9	1	565
1E	5.6	1	226	11E	13.3	1	540
2A	20.9	2	850	12A	12.3	1	499
2B	20.6	2	838	12B	10.4	1	422
2C	20.3	2	826	12C	12.6	1	512
2D	20.8	2	846	12D	16.5	1	671
2E	19.4	2	789	12E	9.1	1	369
3A	20.4	2	830	13A	12.3	1	499
3B	16.7	2	679	13B	8.9	1	361
3C	19.8	2	806	13C	7.1	1	287
3D	17.2	2	699	13D	10.0	1	406
3E	14.2	1	577	13E	13.3	1	540
4A	19.9	2	810	14A	18.4	2	748
4B	11.4	1	463	14B	18.9	2	769
4C	19.4	1	789	14C	16.7	2	679
4D	17.9	2	728	14D	19.6	2	797
4E	17.1	2	695	14E	20.3	2	826
5A	16.1	2	655	15A	10.2	3	414
5B	19.0	2	773	15B	15.4	3	626
5C	19.8	2	806	15C	17.9	2	728
5D	14.3	1	581	15D	19.1	2	777
5E	19.8	2	806	15E	9.8	3	397
6A	12.1	4	491	16A	12.7	1	516
6B	10.4	1	422	16B	17.0	2	691
6C	14.8	2	601	16C	16.5	2	671
6D	17.9	2	728	16D	19.5	2	793
6E	18.0	1	732	16E	17.9	2	728
7A	10.3	4	418	17A	11.5	1	467
7B	14.8	2	601	17B	16.1	2	655
7C	9.1	2	369	17C	20.0	2	814
7D	NA	1	No Data	17D	14.8	3	601
7E	17.9	1	728	17E	18.9	2	769
8A	10.3	1	418	18A	18.0	2	732
8B	13.7	1	557	18B	19.4	2	789
8C	12.5	1	508	18C	16.7	2	679
8D	12.7	1	516	18D	19.7	2	801
8E	13.0	1	528	18E	17.2	2	699
9A	10.0	1	406	19A	19.1	2	777
9B	9.9	1	401	19B	7.9	1	320
9C	10.0	1	406	19C	18.7	2	761
9D	10.1	1	410	19D	17.7	2	720
9E	14.5	1	589	19E	17.2	2	699
10A	16.5	2	671	20A	19.7	2	801
10B	18.3	2	744	20B	17.8	2	724
10C	19.1	2	777	20C	20.6	2	838
10D	15.6	1	634	20D	19.0	2	773
10E	18.3	1	744	20E	15.7	2	638

Table B2: Pull-off Tensile Strength Data for Panels 21 – 40

Specimen ID	Burst Pressure [psi]	Failure Mode	Pull-off Tensile Strength [psi]	Specimen ID	Burst Pressure [psi]	Failure Mode	Pull-off Tensile Strength [psi]
21A	6.2	1	250	31A	12.8	1	520
21B	12.3	1	499	31B	11.6	1	471
21C	10.4	1	422	31C	14.0	1	569
21D	9.3	1	377	31D	9.5	1	385
21E	9.0	1	365	31E	8.9	1	361
22A	20.1	2	818	32A	9.3	4	377
22B	16.6	2	675	32B	12.6	1	512
22C	16.1	2	655	32C	16.2	2	659
22D	13.8	1	561	32D	16.5	4	671
22E	19.7	2	801	32E	19.6	2	797
23A	17.3	3	704	33A	11.0	1	446
23B	20.7	2	842	33B	10.5	1	426
23C	14.7	3	597	33C	11.7	1	475
23D	19.5	2	793	33D	12.8	1	520
23E	23.0	2	936	33E	10.4	4	422
24A	18.8	2	765	34A	16.4	1	667
24B	19.2	2	781	34B	19.3	1	785
24C	19.5	2	793	34C	16.7	1	679
24D	14.4	4	585	34D	15.7	1	638
24E	6.5	4	263	34E	20.9	2	850
25A	14.4	1	585	35A	17.8	1	724
25B	9.5	4	385	35B	16.4	2	667
25C	9.4	4	381	35C	21.0	2	855
25D	9.1	4	369	35D	19.6	2	797
25E	10.0	4	406	35E	17.6	4	716
26A	18.5	2	753	36A	21.0	2	855
26B	19.5	2	793	36B	20.8	2	846
26C	18.8	2	765	36C	20.8	2	846
26D	10.2	4	414	36D	16.8	2	683
26E	19.8	2	806	36E	21.3	2	867
27A	6.4	1	259	37A	21.3	2	867
27B	4.8	1	193	37B	20.6	2	838
27C	10.5	1	426	37C	20.4	2	830
27D	2.4	1	95	37D	17.8	2	724
27E	5.8	1	234	37E	21.0	2	855
28A	9.8	1	397	38A	20.3	2	826
28B	14.4	1	585	38B	18.4	3	748
28C	12.7	1	516	38C	16.2	1	659
28D	12.7	1	516	38D	14.2	1	577
28E	8.4	1	340	38E	17.3	2	704
29A	18.0	3	732	39A	10.9	1	442
29B	12.3	4	499	39B	12.8	1	520
29C	17.3	4	704	39C	11.7	1	475
29D	19.5	1	793	39D	12.1	1	491
29E	15.2	1	618	39E	11.6	1	471
30A	12.8	1	520	40A	9.1	1	369
30B	11.2	1	455	40B	11.9	1	483
30C	10.9	4	442	40C	10.1	1	410
30D	9.8	1	397	40D	9.4	1	381
30E	8.4	1	340	40E	9.8	1	397

APPENDIX C: GLOSS TESTING RAW DATA

Table C1: Gloss Data for Panels 1 – 15

Specimen ID	60° Measurement	Specimen ID	60° Measurement	Specimen ID	60° Measurement
1A	75.0	6A	82.6	11A	83.6
1B	82.6	6B	82.5	11B	83.0
1C	81.9	6C	79.6	11C	80.9
1D	83.1	6D	86.0	11D	83.6
1E	80.9	6E	87.5	11E	83.8
1F	77.0	6F	83.2	11F	84.4
1G	76.2	6G	82.5	11G	82.0
1H	84.2	6H	84.7	11H	84.6
2A	68.6	7A	49.0	12A	82.4
2B	82.9	7B	70.5	12B	84.4
2C	80.9	7C	53.1	12C	81.4
2D	83.3	7D	61.0	12D	80.9
2E	82.2	7E	61.5	12E	83.4
2F	84.6	7F	71.3	12F	81.3
2G	85.0	7G	61.2	12G	80.5
2H	86.4	7H	68.7	12H	83.0
3A	31.6	8A	88.1	13A	84.2
3B	82.1	8B	87.8	13B	85.1
3C	74.1	8C	86.7	13C	84.6
3D	79.1	8D	87.6	13D	86.5
3E	80.2	8E	87.2	13E	86.1
3F	80.6	8F	86.7	13F	84.9
3G	83.0	8G	88.2	13G	85.7
3H	84.8	8H	87.4	13H	86.6
4A	25.5	9A	85.9	14A	86.3
4B	31.9	9B	87.5	14B	87.3
4C	25.1	9C	88.0	14C	86.5
4D	32.3	9D	87.3	14D	85.8
4E	34.1	9E	87.4	14E	84.5
4F	24.3	9F	85.2	14F	80.3
4G	34.0	9G	84.9	14G	85.7
4H	50.1	9H	87.1	14H	84.1
5A	73.0	10A	64.4	15A	85.8
5B	80.6	10B	40.6	15B	87.4
5C	76.2	10C	20.4	15C	87.5
5D	77.2	10D	21.7	15D	84.9
5E	79.7	10E	54.3	15E	87.3
5F	80.0	10F	69.2	15F	86.6
5G	83.5	10G	52.6	15G	85.0
5H	85.1	10H	65.5	15H	85.5

Table C2: Gloss Data for Panels 16 – 40

Specimen ID	60° Measurement	Specimen ID	60° Measurement	Specimen ID	60° Measurement
16A	83.1	25A	80.6	34A	84.3
16B	77.7	25B	87.7	34B	86.2
16C	80.1	25C	84.3	34C	84.7
16D	78.4	25D	84.9	34D	82.8
16E	80.1	25E	81.2	34E	85.2
16F	79.6	25F	85.2	34F	79.0
16G	75.8	25G	83.1	34G	83.6
16H	76.7	25H	85.3	34H	85.4
17A	85.6	26A	84.7	35A	85.8
17B	84.7	26B	84.6	35B	84.1
17C	83.6	26C	82.7	35C	83.8
17D	86.1	26D	80.8	35D	86.0
17E	84.7	26E	80.5	35E	87.4
17F	83.8	26F	85.2	35F	83.4
17G	83.9	26G	82.3	35G	86.0
17H	84.7	26H	84.8	35H	81.5
18A	84.8	27A	72.3	36A	86.6
18B	84.7	27B	75.7	36B	87.3
18C	83.7	27C	69.7	36C	85.8
18D	82.4	27D	71.2	36D	85.9
18E	81.8	27E	61.3	36E	86.3
18F	83.9	27F	75.4	36F	81.8
18G	79.2	27G	67.5	36G	85.1
18H	84.1	27H	69.3	36H	86.1
19A	77.6	28A	88.1	37A	83.7
19B	76.3	28B	87.7	37B	80.6
19C	76.2	28C	87.6	37C	74.8
19D	76.4	28D	89.0	37D	80.4
19E	76.4	28E	88.5	37E	80.8
19F	79.5	28F	85.8	37F	77.6
19G	74.7	28G	87.6	37G	83.3
19H	79.7	28H	85.8	37H	83.3
20A	74.5	29A	75.6	38A	50.5
20B	74.3	29B	84.0	38B	57.0
20C	76.1	29C	79.4	38C	48.4
20D	77.1	29D	81.2	38D	53.4
20E	73.2	29E	75.1	38E	66.9
20F	79.5	29F	83.7	38F	61.9
20G	71.2	29G	74.1	38G	64.0
20H	73.4	29H	79.9	38H	71.1
21A	86.7	30A	86.6	39A	86.1
21B	89.1	30B	86.6	39B	85.7
21C	85.6	30C	86.5	39C	86.6
21D	87.7	30C	86.7	39D	87.0
21E	86.9	30E	86.2	39E	86.0
21F	88.6	30F	82.7	39F	86.3
21G	85.9	30G	83.7	39G	84.8
21H	87.1	30H	84.3	39H	86.9
22A	77.9	31A	83.9	40A	85.7
22B	84.6	31B	83.5	40B	86.1
22C	77.9	31C	84.8	40C	85.1
22D	83.5	31D	84.2	40D	85.1
22E	80.6	31E	84.0	40E	86.2
22F	78.8	31F	83.1	40F	85.2
22G	77.7	31G	82.6	40G	84.7
22H	83.6	31H	82.5	40H	86.1
23A	88.0	32A	83.1		
23B	86.0	32B	84.0		
23C	84.2	32C	84.4		
23D	86.7	32D	83.6		
23E	85.0	32E	83.5		
23F	86.5	32F	85.1		
23G	84.3	32G	86.3		
23H	87.9	32H	85.3		
24A	62.8	33A	85.4		
24B	78.6	33B	81.6		
24C	77.1	33C	84.3		
24D	70.2	33D	83.9		
24E	71.5	33E	83.8		
24F	81.1	33F	85.5		
24G	82.4	33G	81.2		
24H	83.3	33H	83.0		

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