PROFILE EXTRUSION OF WOOD PLASTIC CELLULAR COMPOSITES AND FORMULATION EVALUATION USING COMPRESSION MOLDING

By

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“Only mothers can think of the future-because they give birth to it in their children.”

-Maxim Gorky

Abstract

Wood Plastic Composites (WPCs) have experienced a healthy growth during the last decade. However, improvement in properties is necessary to increase their utility for structural applications. The toughness of WPCs can be improved by creating a fine cellular structure while reducing the density. Extrusion processing is one of the most economical methods for profile formation. For our study, rectangular profiles were extruded using a twin-screw extrusion system with different grades of HDPE and with varying wood fibre and lubricant contents together with maleated polyethylene (MAPE) coupling agent to investigate their effects on WPC processing and mechanical properties. Work has been done to redesign the extrusion system setup to achieve smoother and stronger profiles. A guiding shaper, submerged in the water, has been designed to guide the material directly through water immediately after exiting the die; instead of passing it through a water cooled vacuum calibrator and then through water. In this way a skin was formed quickly that facilitated the production of smoother profiles. Later on chemical blowing agent (CBA) was used to generate cellular structure in the profile by the same extrusion system. CBA contents die temperatures, drawdown ratios (DDR) and wood fibre contents (WF) were varied for optimization of mechanical properties and morphology. Cell morphology and fibre alignment was characterized by a scanning electron microscope (SEM).

A new compression molding system was developed to help in quick evaluation of different material formulations. This system forces the materials to flow in one direction to achieve higher net alignment of fibres during sample preparation, which is the case during profile extrusion. Operation parameters were optimized and improvements in WPC properties were observed compared to samples prepared by conventional hot press and profile extrusion.

Acknowledgement

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I thank my mother for her encouragement.

Above all thank to my God.

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List of Abbreviations and Symbols

μA	=	Micro amp
Φ_v	=	void fraction
δ	=	wall thickness
σ_T	=	Tensile strength
σ_F	=	Flexural strength
ρ_f	=	Density of foam in WPC
ρ_{nf}	=	Density of non-foamed WPC
ρ_p	=	Density of the polymer phase
ρ_p	=	density of the polymer phase
Ψ	=	Expansion factor of foamed sample
A	=	Area of the micrograph (cm^2) and
A_s	=	Cross-section area
b	=	Width of sample
d	=	Thickness of sample
Ecl	=	Tensile modulus of WPC
Ep	=	Tensile modulus of polymer
E_w	=	Tensile modulus of wood fiber
F_T	=	Applied tensile force
L	=	Distance between support points
m_c	=	weight of polymer per cell
M	=	Magnification factor
N	=	Cell density (cells/cm^3)
n	=	Number of cells
P	=	Applied bending force
T_c	=	Theoretical tensile strength of composite
T_p	=	Theoretical tensile strength of HDPE
T_w	=	Theoretical tensile strength of wood
V	=	Volume fraction
V_o	=	Net volume
V_s	=	Volume of the substance (solid phase)
V_g	=	Total volume of all voids
DDR	=	Draw Down Ratio
GPa	=	Giga Pascal
HDPE	=	High density poly ethylene
KV	=	Kilo Volt
MFI	=	Melt flow index
MPa	=	Mega Pascal
PVC	=	Poly Vinyl Chloride
PP	=	Poly Propylene
psi	=	Pound per square inch
ROM	=	Rule of Mixture
TSE	=	Twin Screw Extruder
WPC	=	Wood Plastic Composite

Nomenclature

PBA- Physical Blowing Agent

CBA- Chemical Blowing Agent

Thermosets- polymer that irreversibly cures

DDR- the ratio of die opening thickness to product thickness

Viscosity- measurement of resistance of a fluid deformed by stress

Capillary Rheometer- the most common device for measuring viscosity

Polyethylene- a thermoplastic polymer of long chains of monomer ethylene

Viscoelasticity- property that exhibit both viscous and elastic characteristics

Polyolefins- a polymer produced from a simple olefin (C_nH_{2n}) as a monomer

Hygroscopicity- ability to attract water molecules by absorption or adsorption

Thermal degradation of polymer- molecular deterioration due to overheating

PID- Proportional–integral–derivative controller is a generic control loop feedback mechanism

ASTM = American Society for Testing and Materials, is an international standards organization

Coupling agents- chemical substance reacts with both reinforcement and resin matrix of a composite material

Thermoplastics- polymer that turns to a liquid when heated and freezes to a very glassy state when cooled sufficiently

Rheometry- techniques to determine the rheological properties of material, that are the quantitative and qualitative relationships between deformations and stresses and their derivatives.

Melt flow index (**MFI)** - measurement of ease of flow of melt of a thermoplastic polymer. Defined as the mass of polymer, in grams, flowing in ten minutes through a capillary of a specific diameter and length by a pressure applied via prescribed alternative gravimetric weights for alternative prescribed temperatures

Chapter 1 Introduction

1-1 Background

Composite materials are defined as engineered materials made from two or more constituent materials with significantly different physical or chemical properties that remain separate and distinct on a macroscopic level within the finished structure. Mathews and Rawlings [1] defined composites as materials having two or more distinct constituents each present in reasonable proportion and with different properties. The most primitive man-made composite materials were made of straw and mud to form bricks for building construction. Wood is an example of natural composite made of cellulose fibers in the matrix of lignin [2]. Nowadays composites are popular in building, aerospace, automotive and in many other industries. Composite materials have gained enormous popularity due to their light weight to strength ratio and high performance characteristics.

1-1-1 History of Wood Plastic Composites (WPCs)

The advantages of wood fiber over inorganic fillers and reinforcements in thermoplastics include light weight, reduced abrasion of processing equipment, and renewability [3]. Use of wood fiber, as filler helps reduce raw material costs, and improve stiffness and dimensional stability.

Historical use of wood fiber (WF) in thermoplastics was very limited, perhaps, due to the fact that the plastic processors were unfamiliar with it [4]. Plastic industries and wood industries knew little about each other. They had few material and equipment suppliers in common and often processing of materials was very different and on different scales [5]. Many thermoplastic and thermoplastic composite manufacturers, who tried using wood or other natural fibers, were unaware of or did not address the issues of natural fiber's hygroscopicity and thermal degradation. However, WF's advantages of low cost, low weight relative to inorganic fillers and their availability have attracted increasing number of manufacturers. The following are some of the milestones in the history of WPC:

- 1983: American Woodstock (Sheboygan, Wisconsin) began producing automotive interior materials using Italian extrusion technology.

- 1991: The first international conference on wood fiber Plastic Composites was held in Madison, Wisconsin.

- 1990s (Early): Advanced Environmental Recycling Technologies (AERT, Junction, Texas) and a division of Mobil Chemical Company (Winchester, Virginia) produced solid wood-plastic composites with approximately 50% wood fiber in polyethylene. Strandex Corporation (Madison, Wisconsin) patented technology for extrusion of high wood fiber content (approximately 70% by weight) composites.

- 1993: Andersen Corporation (Bayport, Minnesota) produced wood fiber-reinforced polyvinyl chloride (PVC) sub sills for French doors.

- 1996: A few U.S. companies began producing a pelletized feedstock for the wood-plastic composites industry [6]

The use of other natural fibers is also increasing, particularly in automotive applications, but they are twice as expensive as wood fiber and availability of natural fiber is regional, and as a result, high transportation costs and storage issue are limiting their growth [7].

The decking, window and door profile, and automotive markets account for the majority of the wood-plastics manufactured in the United States. Among them, decking market is one of the largest in US. In 1987, home owners built approximately 3.6 million decks and spent an estimated US$1.9 billion on lumber for these decks [6]. Other

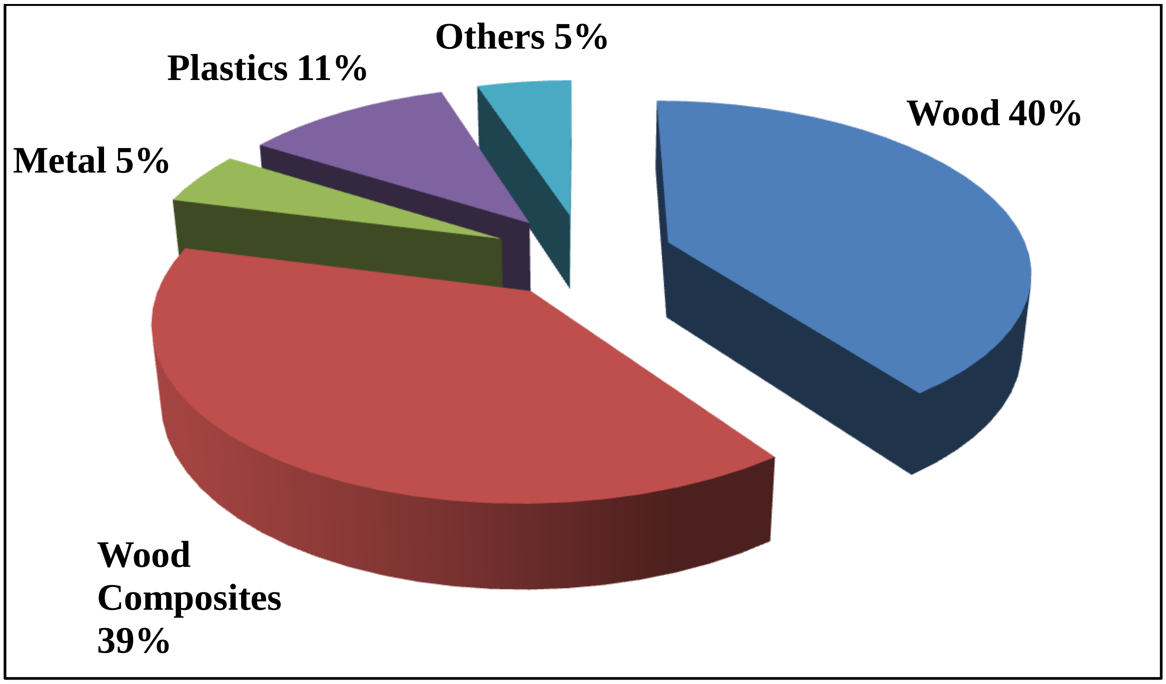


Figure 1-1 Materials used in light-frame residential construction (8)

products made of WPCs are wood shingles, pallets, flower pots, tool handles, hot tub siding, office accessories and others [4]. Figure 1-1 shows the percentages of materials used in light-frame residential construction industry [8].

1-1-2 Profile Extrusion

Extrusion is a process where polymer melt is forced to go through a metal die that continuously shapes the melt into a desired form [9]. In extrusion process molten polymer is propelled along a screw and forced through a die (slit) to form profile. Profile Extrusion is the process used to produce objects with fixed cross-sectional profiles. In extrusion process, material is pushed or drawn through a die with a desired cross-section. A basic extrusion system is shown in Figure 1-2 and a typical screw in Figure 1-3. Two major advantages of extrusion over other manufacturing processes are:

(i) Its ability to create very complex cross-sections and

(ii) Flexibility to work with materials that are brittle, since the material only encounters compressive and shear stresses.

It also forms finished parts with an excellent surface finish [10]. Extrusion may be continuous (theoretically producing indefinitely long material) or semi-continuous (producing many pieces). The first extrusion process was patented by Joseph Bramah in 1797 to extrude lead pipe [11]. In this process metals were preheated and then forced through a die via a hand driven plunger. In 1820, Thomas Burr, constructed the first hydraulic powered press [11]. At this time, the process was called squirting. Continuous extrusion is a popular and cost effective method to manufacture WPC profile [12, 13]**.** The production rates are also considerably faster than other methods. Commercial utility is expected to be elevated further, if manufacturing process of WPC through continuous extrusion can be improved further [14]. Extrusion lines are the major processing equipment for manufacture of linear profiles of WPC. During extrusion, resin, generally in the form of small pellets, is gravity fed through a top mounted hopper into the barrel of the extruder. Additives (in either liquid or pellet form) are often added and can be mixed into the resin prior to arriving at the hopper or fed through a separate hopper. The rotating screw forces the material forward into the barrel which is heated to the desired melt

Figure 1-2 *Examples of WPC lumber profiles [15]*

temperature. The molten materials come out of the die orifice and form profile of desired shape after solidification. Figure 1-3 shows some typical WPC profiles produced commercially.

1-1-3 Foaming

Structural applications of WPC are still limited due to their low impact strength and high density compared to natural wood and certain plastics. These limitations can be improved by microcellular or fine celled foaming. Foaming of WPC improves it's utility as a substitute of wood and plastic products, by reducing density, improving strength, recyclability, improving ductility, lowering material cost, and lowering weight [16]. Foaming also improves nail ability, screw ability, imparts better surface definition, sharper contours and results in reduction of material requirement. Polymers (including thermoplastics) are mostly foamed by the dispersion of gas throughout a molten polymer phase and stabilizing the resultant foam. Foaming can be achieved using blowing agents. There are mainly two types of blowing agents: (i) chemical blowing agents (CBAs) and (ii) physical blowing agents (PBAs). However, CBAs are most popular and their processing is also easier than PBAs.

1-1-4 Compression Molding

Compression molding provides a convenient method for quick evaluation of material formulations instead of labor and material intensive extrusion processing. During extrusion, wood fibers get aligned in the direction of the flow, whereas, in conventional compression molding these are generally randomly oriented in two dimensions. So, the test results obtained from samples prepared by conventional compression molding system might not represent the actual property of WPC after extrusion. This behavior can be corrected by designing a compression molding system that ensures better-alignment of fiber in one direction through unidirectional material flow [17]. We have designed a new compression molding system that facilitates directional flow during the molding process.

1-2 Objectives of the Thesis

Importance and growth of WPCs markets have been well established. However, more research needs to be done to overcome existing drawbacks in the properties of WPCs to widen their practical use. This will further increase the market share of these materials. The objective of this study is to contribute to and augment previous studies by:

i. **Profile extrusion:** Studying process ability of WPC in terms of profile extrusion, process parameters, and material grades & contents

ii. **Foaming:** Optimizing process parameters for foaming during profile extrusion e.g. Draw Down Ratio (DDR), die temperature, blowing agent, WF contents, etc.

iii. **Compression molding:** Building a new compression molding system, to be used to quickly evaluate the material formulation. Comparing sample results obtained from custom built compression molding, conventional compression molding and profile extrusion. Optimizing the process parameters (compression speed and die-press temperature) during compression molding and studying the effect of variation in GF and WF length and HDPE grades on the mechanical properties of WPC.

1-3 Motivations Behind the Thesis

The motivation behind this thesis was the necessities of synthesizing WPC profiles of lighter weight, higher strength to weight ratio and reduced cost. Since the increasing consumptions of wood is reducing forestry resources, it is imperative that research should be carried out to develop more viable alternatives to natural wood. The use of WPC also helps in eliminating the use of pesticides and wood preservatives to help preventing environment pollution. The advantages of WPCs over plastics are their higher mechanical properties, thermal stability, and resistance to ultraviolet degradation. In addition, wood itself is susceptible to fungal attack, insect attack, dimensional instability, shrinkage and distortion when it loses moisture [18]. Most species have density between 0.32-0.72 g/cc, depending on structure and moisture content and have poor decay resistance to moisture and temperature [18]. WPCs are free of these limitations, have improved dimensional stability, low maintenance, improved resistance to moisture and biological degradation, low cost, and are renewable and recyclable.

1-4 Outline of the Thesis

Chapter 1 contains introductory background information on WPC, profile extrusion, foaming and compression molding. Chapter 2 provides detailed literature review on above. Chapter 3 discusses the definitions and experimental methodologies and description of the experiments carried out. Experimental research approaches were mainly carried out for investigations whereas where as desktop research and quotation of references were also carried out to design experiments and to validate findings. Chapter 4 provides the results on the process ability studies for profile extrusion, application of CBAs for foaming and custom build compression molding. Chapter 5 presents the conclusion of the study, summary of findings and recommendations for future work. Below are the concentrations on which research was carried out:

i. **WPC Profile Extrusion:** At this stage optimization of process set up, extrusion parameters and optimization of HDPE grades, lubricant content and WF content were carried out to form WPC profiles.

ii. **Foaming Process:** Chemical blowing agent (CBA) was introduced to foam profiles during extrusion. Extrusion processing conditions, e.g DDR and die temperature; CBA content and WF contents were optimized.

iii. **Compression molding:** As discussed earlier, a compression molding system was developed for quick evaluation of samples. The process parameters (die-press temperature and press speed) of this new system were optimized. Samples prepared by the new system, the conventional system and profile extrusion were compared. Research was also carried out to study the effect of varying polymer MFI grades, wood fiber (WF) length and glass fiber (GF) length on WPC prepared by the custom built system to determine the direction of future research design for stronger and smoother profiles.

Chapter 2 Literature Review

2-1 Theoretical Background

Theoretical equations were used to characterize tensile and flexural strength, void fraction and cell density. Rule of mixture (ROM) was employed to generate theoretical result to compare with experimental results.

2-1-1 Terminologies

Terminology is the study of terms and their use. Below are the terminologies used for characterization of WPC:

Draw Down Ration (DDR) was calculated as the ratio of downstream puller linear speed to the extrusion linear speed during the extrusion runs.

Tensile Strength is the maximum stress that can be sustained by a structure in tension. Tensile strength, σ_T, is calculated according to equation 3 [19]:

$$\sigma_T = F_T / A_s \qquad (3)$$

Where,
F_T = *Applied tensile force*
A_s = *Cross-section area*

Flexural Strength is the strength at fracture. Flexural strength, σ_F, was calculated according to equation 4 [19]:

$$\sigma_F = 3PL / 2bd^2 \qquad (4)$$

Where,
P = Applied bending force
L = Distance between support points
b = Width of sample
d = Thickness of sample

The Total Void Fraction represents the total volume occupied by the gas within the foamed sample which can be determined from the foamed density and non-foamed density of WPC by using equation 5:

Void Fraction (%),

$$\Phi_v = (1 - (\rho_f / \rho_{nf})) \times 100 \quad \text{.......} \quad (5)$$

Where, ρ_f = Density of foam in WPC
ρ_{nf} =Density of non-foamed WPC

Cell Density, defined as the number of cells per unit volume with respect to the non-foamed composite was determined by following Equation 6 [20]:

$$N = [nM^2/A]^{3/2} \times \psi \quad \text{.......} \quad (6)$$

Where, N = Cell density (cells/cm^3)
n = Number of cells
M = Magnification factor
A = Area of the micrograph (cm^2) and
Ψ = Expansion factor of foamed sample

Rule of Mixture (ROM) principle was followed to generate theoretical results and was compared with experimental results. According to the ROM, the properties of a multiphase alloy or composite material are a weighted average (usually on the basis of volume) of the properties of the individual constituents [21]. Two mathematical expressions have been formulated for the dependence of the tensile modulus on the volume fraction of the constituent phase for the two-phase composite. These equations predict that the tensile modulus should fall between an [21]:

upper bound,

$$E_{cl} = E_pV_p + E_wV_w \quad \text{.........} (7)$$

and lower bound,

$$E_{cl} = E_PE_W/(E_PV_w + E_wV_h) \quad \text{.........} (8)$$

V= Volume fraction
E_{cl} = *Tensile modulus of WPC*
E_p = *Tensile modulus of polymer*
E_w = *Tensile modulus of wood fiber*

A JEOL JSM 6060 **Scanning Electron Microscope** (SEM), operated at an accelerating voltage of 20 kV and emission current of 47 μA, was used to investigate the morphology of samples.

2-2 Wood Plastic Composites (WPCs)

Polymers are long chains, also referred to as backbone, comprised of many atoms connected together. The atoms in the backbone are usually carbon (C), oxygen (O), nitrogen (N), or sulfur (S) and atoms attached to the long polymer backbone are normally combination of hydrogen (H), carbon, oxygen, chlorine (Cl), fluorine (F), and/or bromine (Br) [22]. Plastics are generally polymeric molecules and are a generic term for an extremely large and complex family of materials. Plastics are comprised of relatively simple monomer species that build up into a linear, branched, or 3-dimensional network macromolecule [23].

The advantages of WPC over plastics are:

i. Higher mechanical properties

ii. Thermal stability and

iii. Resistance to ultraviolet degradation

Since the increasing consumption of wood is reducing forestry resources, it is imperative that research should be carried out to develop more viable alternatives to natural wood. In addition, wood itself is susceptible to fungal attack, insect attack, dimensional instability, shrinkage and distortion when it loses moisture [18]. Most species have density between 0.32-0.72 g/cc, depending on structure and moisture content and have poor decay resistance to moisture and temperature [18]. WPCs are free of these limitations, have improved dimensional stability, low maintenance, improved resistance to moisture and biological degradation, low cost, and are renewable and recyclable. WPCs have emerged as a dynamic growth material in building construction and automotive applications. Almost all major auto manufacturers in Germany (Volkswagen, Audi Group, BMW, and Daimler-Benz) now use WPC in a variety of applications, which makes plant fibers the fastest growing type of polymer additives (as fillers and reinforcements) [24]. The use of WPC also helps in eliminating the use of pesticide and wood preservatives to help prevent environment pollution. WPCs account for an average annual growth rate of approximately 18% in Northern America and 14% in Europe (Gupta et al., 2007) and are one of the fastest growing sectors in plastic industry [25].

Research is ongoing on the WPCs to improve their physical and mechanical properties [26, 27], morphology [28], rheological properties [29-31] and durability. The wood fiber (WF) contents could be as high as ~70 weight percent in WPC products, and it is found that its tensile and flexural modulus increases with WF loadings [32]. However, higher WF contents lead to difficulty in industrial processing such as extrusion and injection molding due to high melt viscosity of wood-plastic (WP) blends [29-31]. This also influences the mechanical properties of WPC products [32]. Li *et al.* [30] investigated the melt behavior of WP blends using torque rheometry, and found that blend viscosity increases with increase in WF contents. Hristov *et al.* [31] studied the effect of matrix melt flow index (MFI) on melt behavior and extrudate distortions of wood filled metallocene polyethylene (PE) by rotational and capillary rheometers. It was found that high MFI PE composites exhibit less pronounced surface tearing than low MFI PE composites. During extrusion processing, the melt viscosity of the WP blends can be reduced by increasing the process temperatures and lubricants. However, higher processing temperatures cause degradation of WF (~200 ^{o}C), which results in overall decrease in mechanical properties of WPC [32]. Since, high molecular weight (low MFI) polymers have stronger mechanical properties; most WPCs were made of low MFI polymer [27, 33]. It was also found that addition of small quantity of coupling agents and lubricants improves extrusion processing and mechanical properties of WPCs [29-32]. Low molecular weight MAPE coupling agent tends to decrease the overall WP melt viscosity at lower shear rates thereby acts as an internal lubricant [34]. The coupling agent also improves the compatibility between WF and polymer, whereas lubricants increases the wet ability and produces wall-slippage in the polymer compounds.

The material characteristics such as WF size, contents, coupling agent, lubricant types and contents influence the mechanical and rheological properties of WPC [34-37]. Li and Wolcott [34] studied the rheology of HDPE/Maple flour/maleated polyethylene (MAPE)/lubricant composite with varied WF contents and particle sizes by capillary rheometry. The melt viscosity was changed significantly with WF contents rather than by WF particle sizes. Lubricant caused an excellent external lubrication. Zhang *et al.* [36] reported that due to use of lubricant in twin-screw extruder during extruding HDPE/WF composites, uniform distribution of WF in polymer matrix was observed with decrease in complex viscosity. In terms of processing, the extrusion processing conditions such as screw configuration, extrusion temperatures, through put and drawdown ratio have effect on mechanical property of WPC profile [38]. Charlton *et al.* [38] extruded recycled HDPE filled with 60 wt. % rice hulls composites through profile die, and experienced extrudate tearing at all throughputs and the magnitude of the tearing increased with increasing throughput and decreasing die land temperature. Hristov *et al.* [39] studied the effect on WP melt flow behavior and extrudate distortions of wood filler filled metallocene polyethylene (MPE) composites by capillary rheometer and reported that high MFI PE composite showed less surface tearing than low MFI PE composite. Carneiro *et al.* [40] reported that profiles distortion can be minimized through the use of a low draw down ratio (DDR).

WPCs suffer from some disadvantages compared with natural wood which includes the fact that they have low toughness and are lower in strength. This limits their applications in some areas [41-43]. These disadvantages can be offset with the addition of small amounts of glass fibers [44-46].

2-3 Profile Extrusion

Wood filled polymers have been manufactured industrially almost since the last 80 years [47]. At the early stage, thermosetting resins e.g. phenol-formaldehyde resins, were being used as polymer matrix. Lignocellulosic filled thermoplastics were produced in the late 1960s (Rowell *et al*. 2002) [48]. A number of scientific publications began to appear on this topic during the 1980s and the 1990s (Lu *et al*. 2000) and this trend is continuing [49]. Foaming is also being practiced industrially to improve properties of WPC e.g. (i) reduction of density, (ii) improvement of mechanical strength,(iii) nail ability, (iv) insulation and others.

The extruder is the heart of the WPC processing system. The primary purpose of the extruder is to melt and mix the polymer with wood and additives (compounding). The extruder then passes the compounded and molten WPC through the die to form profile. Cost of an extruder can vary from $150,000 to over $1 million depending on complexity of extrusion system [47]. There are four main types of extrusion systems used to manufacture WPC.

i. Single Screw Extruder: This is the simplest equipment and consists of a single screw inside the barrel. A typical single screw extruder for lumber production will have a barrel length to diameter (L/D) ratio of 34:1[47]. It has melting and metering sections, and a venting section to remove volatiles. Pre-compounded fiber filled polymer pellets are generally used during extrusion. Melting/mixing is done by barrel heating at start up and screw shear. Friction or shear forces melt most of the material during production Advantages are that it is a proven technology and has the lowest capital cost. Disadvantages are: lower output rates, requirement of a drying system, high screw rpm with greater risk of burning at the screw tip, and inability of keeping melt temperature low with higher head pressure [47].

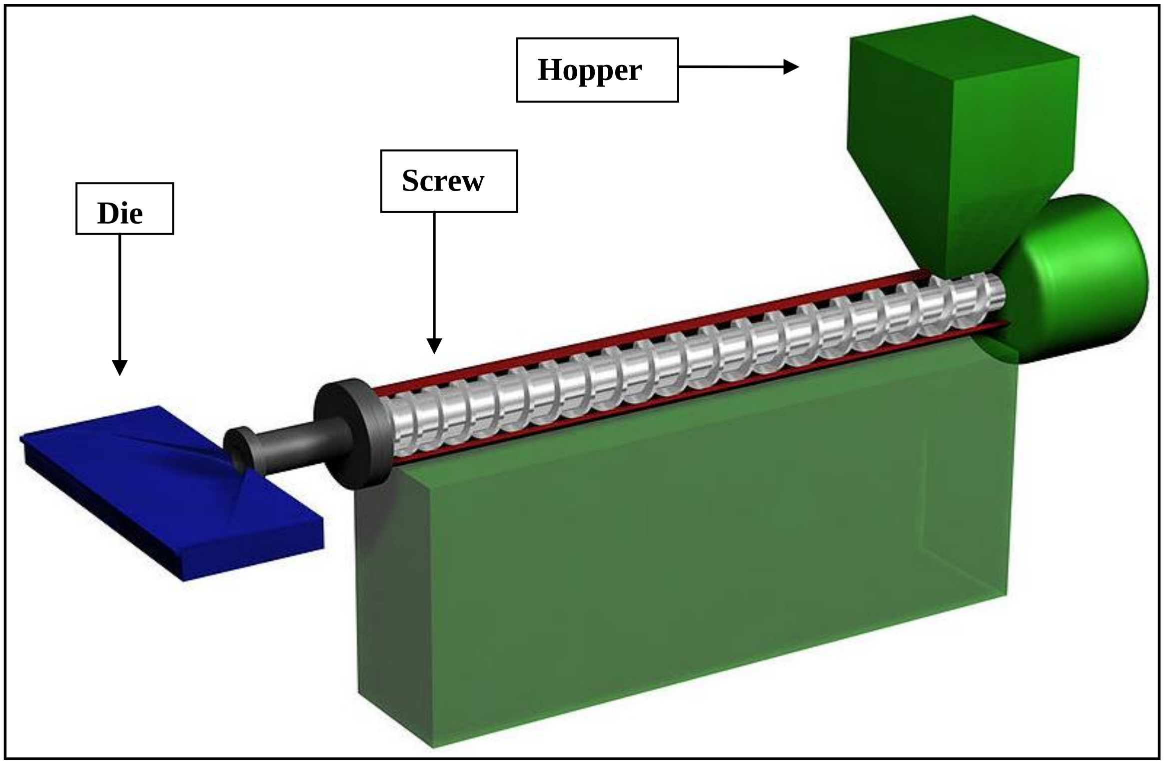


Figure 2-1 *A typical plastic single screw extruder [50]*

ii) Counter-rotating twin-screw extruder: Suitable for processing of heat sensitive polymers like rigid PVC, low temperature extrusion for fibers and foams, non-compounded materials like powder blends, materials that are difficult to feed, and materials, that requires degassing. They have either parallel or conical screw configurations. Melting/mixing mechanism is barrel heat and screw mixing through screw flight cut-outs and gear mixers. Advantages include low screw rpm and low shear mixing. Disadvantages include requirement of drying system, a pre-blending system is required, material transportation can impact the mix feed ratios [47].

iii) Co-Rotating Twin Screw & Hot Melt Single Screw Tandem System: In this case, a parallel co-rotating twin screw extruder is coupled with a "hot melt", single screw extruder. The material for this system is wood flour at ambient moisture content (5 to 8%) and the polymer and additives can be in their natural states [47]. Feed systems are gravimetric feeders and twin-screw side feeders. Melting/mixing is done by barrel heat, screw rpm and screw mixing. This system allows wood to be processed at ambient moisture content since the extruder is used to dry the fiber. Disadvantages include the need for peripheral feeding systems, high screw rpm and no screw cooling (greater risk of burning).

4) Woodtruder is another system which includes a parallel counter-rotating twin - and single-screw extruder, a die tooling system, a spray cooling tank with driven rollers, a traveling cut-off saw, and a run-off table. Wood fiber with ambient moisture content is placed into the unit's fiber feeder and dried within the twin screw. Plastics are melted separately and this prevents wood fibers from burning burned during plastic melting. Both WF and plastics are then mixed and any remaining moisture or volatiles are removed by vacuum venting. Major advantages of this process include no requirement of material preparation, and the ability to process fiber at ambient moisture content (5 to 8%) [47].

Figure 2-2 *WoodtruderÔ extrusion system: material feed, extruders & cooling tank [47]*

Extrusion processing conditions such as screw configuration, extrusion temperatures, throughput, drawdown ratio and others are carefully considered during extrusion process since they have strong relation with the mechanical properties of WPC [51].

2-4 Foaming

While studying the cellular structure of foamed polymers, graphical approach, which is deductions about the macroscopic properties from morphological parameters, is most widely followed [52]. According to widely accepted opinion, only by statistical method we may obtain correct characterization of porous media. The Gas Structural Element (GSE), is a statistically averaged model of a spatial structure consisting of a gas cavity (a cell) and its walls & ribs. The relationship between gas-filling ratio *G (*net GSEs) and the total volume of the system is given by equation (1) [52]:

$$G = V_g/(V_g+V_s) = V_g/V_o \quad \ldots\ldots\ldots \quad (1)$$

Here,
V_o = *net volume*
V_s = *volume of the substance (solid phase)*
V_g = *total volume of all voids*

During foaming, two types of foam cells are observed: open- and closed- cell. Closed-cell structure may entrap hydrogen, carbon dioxide and volatile liquids. The entrapped fluid (e.g. air) plays a major role in increasing in compressive strength and energy absorption capability for closed- cell foams[53]. On the other hand, open-cell foamed plastics have a higher absorptive capacity for water and moisture, a higher permeability to gas and vapor, less insulation capabilities for heat or electricity and a better ability to absorb and damp sound. The relationship between density and mean cell diameter of foam is deduced from equation (2) [52]:

$$m_c = \rho_c\,[4\pi/3((d+\delta)/2)^3 - 4\pi/3((d-\delta)/2)^3] = \pi/3(3d^3\delta+\delta^3)\rho_p \quad \ldots\ldots\ldots (2)$$

δ= wall thickness
d= spherical cell diameter
ρ_p= density of the polymer phase
m_c= weight of polymer per cell

The microcellular structures in the WPC can be achieved using i) Chemical blowing agents (CBAs) [54, 55]; ii) Physical blowing agents (PBAs) [56, 57] and different additives such as coupling and nucleating agents. CBAs liberate gas because of chemical reactions of CBAs or interaction of CBAs with other component, including thermal decomposition. The PBAs liberate gases as a result of physical processes (evaporation, desorption) at elevated temperatures or reduced pressures. However, since high-pressure equipment is required for PBA foam production, the equipment investment is higher in case of PBA than that for the CBA. Currently, PBAs are used for low-density foams and CBAs are used for high-density foams. Chemical blowing agents (CBAs) are easy to introduce, can be processed with ordinary equipment and production costs are lower. CBAs have different varieties including Inorganic Blowing Agents e.g. ammonium carbonate and Organic Blowing Agents e.g. azo and diazo compounds. A simple classification of CBA is endothermic CBAs which absorb heat and usually generate CO_2 and exothermic CBAs which release heat and usually generate N_2 during their chemical decomposition. Heat absorption by endothermic CBAs cool the polymer, increase the viscosity of the melt, stabilize the cellular structure, and reduce cell coalescence where as heat generated by exothermic CBAs may soften the polymer matrix and help cell coalescence during the foaming that results in poor cellular structure with large bubbles [58]. Exothermic CBAs have most of the market share although recently it was found that endothermic CBAs improve cell structure and workability. Foaming with CBAs need cross-linking[59]. Cross-linking stabilizes bubbles during expansion and enhances resistance to thermal collapse. Commercially three groups of CBAs are available: organic nitrogen compounds, sodium bicarbonates and its mixture with citric acid; and sodium borohydride. Foam density is inversely proportional to blowing agent level and cell size largely depends on the manufacturing process. Some important structural variables in foaming are cell density, cell size, cell geometry and open-cell content. While doing foaming of PP/WF composites through extrusion, Shuwen Zhang *et al.* [59] found that addition of a MAPP coupling agent increases the blowing agent efficiency and reduces the density and average cell size lower than 100 micron. Li *et al.* [60] found that the CBA types (endothermic vs. exothermic) and form (pure or master batch) did not affect the density and average cell size achieved for WPC foams, however maleated PE coupled formulation exhibited high void fraction. Rizvi *et al.* [55] examined effects of different endothermic CBAs on foam structure of extruded HDPE/WF composites, and found similar foaming behaviors with uniform and reasonably fine-celled structure (smaller than 100 μm) for all CBAs. Bledzki *et al.* [57] reported that exothermic foaming agent shows best performance considering cell size and diameter. The mechanical properties of MAPP coupled foamed WF-PP composites improved up to 80 %. On the other hand, the endothermic foaming agent reduced surface roughness by nearly 70 % compared to non-foamed composites.

Mechanical properties of the composites are influenced by the type of base polymer and processing conditions. Most polyolefin foams result in a closed-cell structure. Compressive strength increases rapidly as the foam density increases where as open cells results in lower compressive strength and in decrease of initial modulus. Incase cell orientation is parallel to the direction of load application, the compressive strength is found to be higher than the compressive strength perpendicular to the direction of cell orientations. If foam is compressed, the gas phase is compressed and the cell walls deform and buckle. Commercial foam production processes is done through Direct Extrusion, Accumulating Extrusion, Flash Extrusion, Foam Shaping, and Coalesced Strand Foam. Important factors to be considered during foam production are nucleation, bubble formation, surface tension, bubble growth, and others. Recently, the batch [61, 62], injection molding [55] and continuous extrusion [59, 60] processes have been used for microcellular foaming of WPCs. Matuana *et al.* reported batch process which is similar to the one invented at MIT for microcellular foaming where compression molded WPC was saturated with CO2 gas under high pressure, the pressure was released suddenly and the sample was immediately submerged in to hot oil bath. The pressure drop resulted in nucleation of large number of micro bubbles [52]. Bubble growth was facilitated by increasing the temperature. A tenfold expansion was reported. Torres *et al.*

studied bubble formation during extrusion in a single screw extruder and concluded that bubble formation heavily depends on feeding behavior of polymer-fiber blend [52]. Use of wood fiber contents might be as high as ~70 wt% and its tensile and flexural modulus could increase with the WF loadings [63].

Major phases involved in foaming of WPC are [52]:

i. Polymer/Gas Solution Formation
ii. Cell Nucleation
iii. Cell Growth

Figure 2-3 shows changes in polymer structure during foaming process. The commonly used exothermic or endothermic CBAs decompose at 185 to 220 C. This necessitates that foaming of WPC be done at higher temperature [52].The amount of moisture present in wood fiber has effect on foaming behavior. Composites with oven-dried wood flour exhibited a small temperature window in which foam density can be reduced to a desired density (0.6-0.8 g/cm^3). Since the amount of moisture was minimum in case of dried fiber the amount of open cell content would not be high. On the contrary non-dried wood flour composite foams do not exhibit sharp density reduction because of the presence of large amount of moisture and presence of many non dissolved moisture pockets formed due to low solubility of moisture. Gou *et al.* found that foam density of composite

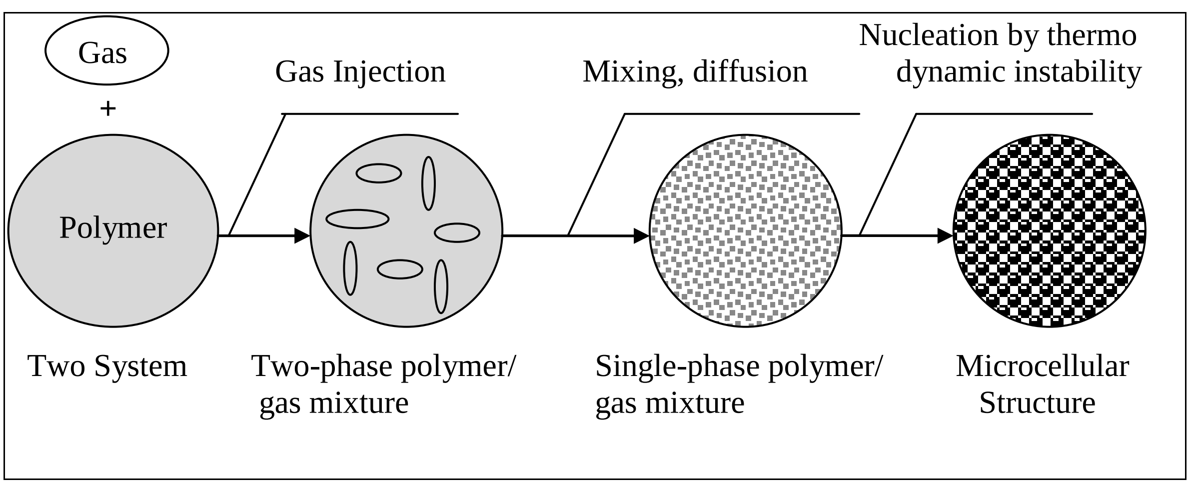


***Figure 2-3** Phase changes in polymeric foaming process [52]*

increased as devolatilizing temperature increased since more moisture and other volatiles were removed when the devolatilizing temperature was higher. However, over dried (e.g. at 150C) wood fiber showed the presence of bigger cells with low cell density which means drying at higher temperature increases the insoluble volatiles by degrading the wood flour and decreasing the soluble volatiles[52]. Guo *et al.* [64] reported that the highest processing temperature should be below 170 °C to minimize the adverse foaming effects of the volatiles generated from WF during extrusion. Finding optimum processing temperature is important to get acceptable cellular structure.

When the plastic matrix is filled with wood fiber, the viscoelastic properties of the matrix is different than the polymer [53, 63] and the effect of CBA types and contents on the cell morphology of WPC is also different.

2-5 Compression molding

In compression molding system compression force is applied among molds to get the desirable shape of molten materials through solidification. In this process good optimization could be obtained between the flow of material and the mechanical properties [65]. Meij [66] investigated non-isothermal effects on the flow of Glass Mat reinforced Thermoplastics (GMT) during moulding and suggested process control tools. Commercial moulding prediction tools issued from work on non -isothermal moulding are available [67, 68] and are commonly used for mould filling studies for new applications [69]. It was found that the fiber content and the polymer viscosity both have a large influence on the work of compression [66]. However, not much is known about the influence of each of the constituent materials on processing and about the evolution of the temperature and deformation fields during non-isothermal flow [70]. During industrial flow molding processing, composites are preheated and then transported to the mould, which is maintained at a temperature below the solidification temperature of the matrix. The mould is then closed at a high speed. When the flow is almost complete, the press is switched from displacement control to pressure control and a pressure is maintained until the mould is full and the material inside has cooled sufficiently. The material in the inlay zone cools first and its rheology and solidification behavior determines the extent of flow in the rest of the mould. The void content during dwell and initial compression is high due to lofting of the preforms at preheating. When the hot preform is compressed between the platens, the macro-voids are removed. During the initial phase of flow, some voids, still remain in the material are rapidly suppressed. The material then flows out from between the platens and flow ends until the material between the platens is solidified throughout thickness. However, surface void formation is still a major problem, especially for application in automotive industry [71]. Such voids require costly after treatment to enable good appearance after painting.

In compression molding, compounding step can be eliminated and mixing and molding can be done in one step [72, 73]. This will allow minimizing length reduction of the fibers and the time the fibers spend at high temperature. In this method the cellulose fibers get distributed evenly between polymer films and then the ‘‘sandwich’’ is created by pressing in a compression molder at adequate time and temperature. Bullions et al. [74, 75] prepared composites of kenaf bast, wood pulp, and poultry feather fiber by mixing with polypropylene and then passing them through a hot oven at a fast rate to melt the PP fibers but not degrade the natural fibers and a formed a prepreg. The prepreg is then compression-molded into laminate plates under pressure (4.44 MPa) and temperature (180 °C) [76].

However, still unsolved and often studied problem during compression molding is the reorientation of the fibers during processing [71]. It is very important to know the mechanism of reorientation since the final fiber orientation and distribution has strong influence on the strength properties. A properly designed mold can ensure greater uni-directional alignment avoiding void formations.

Chapter 3 Experimental Methods

3-1 Materials Used

3-1-1 HDPE

Among all plastics Polyethylene(PE) family has superior impact strength, excellent stretching behaviour, high chemical resistance, and is lower in cost. It also performs well at low temperatures. It is used more than any other polymer. Among the PE family High Density Poly Ethylene (HDPE) is the most widely used thermoplastic. The additional benefits of HDPE are their mechanical properties, like stiffness and tensile strength[77]. The global HDPE market in 2007 had a volume of more than 30 million tons [78]. In this thesis the plastics used were mainly HDPE. A grade of LDPE was also used for comparison. Four different types of HDPE used were:

Table 3-1 *List of HDPE and LDPE polymers used*

Polymer grade/suppliers	Code used	MFI (g/10min, 2.16 kg/190°C)	Density (g/cm^3)
HDPE 2710/ Nova Chemicals	HDPE-1	17	0.951
DMDA-8007/ DOW Chemicals	HDPE-2	8.25	0.965
HB-W355-A/ Nova Chemicals	HDPE-3	3	0.955
HB-L354-A/ Nova Chemicals	HDPE-4	0.3	0.955
LDPE (609-A)/ DOW Chemicals	LDPE-1	0.9	0.924

Another reason of choosing HDPE is their low softening temperature which is preferred in production of WPC [79].

In 2005, in the United States, plastics accounted for 11.8% of the 246 million tones of municipal solid waste [19, 80] and HDPE had a major share. However, recycled HDPE are 31–34% cheaper than virgin one [81]. Using this to form WPC is an effective solution of plastic waste disposal.

3-1-2 Wood Fiber

The yield of main product at saw mill may be as low as 20% of the log [82] and the rest are wastage. In 2002, in United States alone, about 63 million metric tons of wood waste was generated, majority of which was destined for landfill [83]. Waste wood is suitable as filler for polyolefin [84, 85] and use of waste wood also reduces disposal cost.

The wood fiber (WF-1) used was standard softwood (pine) grade 12020, particle length-75-125µm and 120-200 mesh, provided by American Wood Fibers. Pine wood is popular in North America and the above mentioned grade is found in fine powdered form. Softwoods are longer than hardwood and structure is less complex, so they are preferred in some applications [86]. However, to study the effect of fiber length on final properties of WPC, wood fiber (WF-2) with a different fiber length was used that was standard softwood (pine) grade 2020, particle length-250-850µm and 20-60 mesh supplied by American Wood Fibers.

3-1-3 Coupling Agent (CA)

Addition of coupling agents showed improvement in extrusion processing and mechanical properties of WPC products [87-91]. CA improves adhesion between hydrophobic HDPE and hydrophilic WF. CA also improves adhesion between plastic and GF [92]. Commercial grade maleated polyethylene (MAPE grade-NE-542-013 MFI: 13g/10min and density: 0.943 g/cm^3) was used as coupling agent (CA-1) and was provided by Struktol America. Since this CA was polyethylene based, it was considered to be more compatible with HDPE polymer and also with the Lubricant which was procured from Struktol as well. The low molecular weight MAPE coupling agent also helps decreasing the overall Wood plastic melt viscosity at lower shear rates and thereby acts as an internal lubricant [89].

Another type of coupling agent (CA-2), Maleic anhydride-g- HDPE (MAH- g-PE, Fusabond MB-100D, 0.8~1.1 wt% of MAH, MFI 2.0 g/10min, DuPont Canada), was also used to make samples using custom build compression molding to study improvement in mechanical properties due to variation in coupling agent.

3-1-4 Glass Fiber (GF)

Glass-fiber (GF) has superior strength than wood fiber and E-glass is the most widely used GF to reinforce plastics and exhibits performance rating at lower cost [36]. Two types of E-glass fibers were used, both provided by Owens Corning, to study the effect of fiber length on mechanical properties of WPC. GF-1 (Milled Fiber grade 737-BD which is 1/32" (0.8 mm) in length and 16 micron in diameter), GF-2 (Milled Fiber 144A-14C, 4mm in length and 14 micron diameter).

3-1-5 Lubricant

The difficulty in extrusion processing due to high melt viscosity because of high WF contents can be reduced using lubricants [89, 93-95]. Addition of lubricants already showed improvement in extrusion processing and mechanical properties of WPC products [87-91]. Lubricant also improves the melt homogenization, viscosity, surface quality and extruder output during processing and facilitates wet ability and wall slip between the WP melts and extruder. The lubricant used was TPW 709 (blend of non metallic internal/external processing aids) provided by Sturktol Canada ltd.

3-1-6 Chemical Blowing Agent (CBA)

Generally CBAs are used for high-density foams. Among them Endothermic CBAs have desirable property as during foaming they absorb heat at decomposition, which cools the polymer, increases the viscosity of the melt, stabilizes the cellular structure, and reduces cell coalescence [93, 95]. Rizvi *et al.* [52] examined the effects of different endothermic CBAs on foam structure of extruded HDPE/WF composites, and found similar foaming behaviors with uniform and reasonably fine-celled structure (smaller than 100 μm) for all CBAs, which were used. Safoam PE-20 (endothermic, 20 % active ingredient, gas evolved: 15-25 ml/g, main gas: CO_2, other gas: H_2O, decomposition temperature: 176 ^oC) was used as CBA, and was provided by Reedy International, USA.

3-2 Formulations of Pellets

At first WF was dried at 80 ^oC for 24 hrs prior to compounding. HDPE was also dried at 65°C for 12 hrs. Composite formulations were designed based on both wt% and parts per hundred parts of resin (phr) by mass proportion. The HDPE, WF, GF (whenever used), coupling agent, and lubricant (whenever used) were first dry mixed and then compounded using a co-rotating intermeshing twin-screw extruder (ZSE-27 Leistritz, screw diameter: 19 mm, L/D ratio: 40) with side stuffer. All the materials were fed through a Brabender Technologies feeder (model no DDW-MD5-FW40/3PLUS-50). Feeding operations were controlled through Congrav RC 4A operating unit by Brabender. Feeding rate was between 1-2 kg/hr. A software, Macromatex Twin Screw Control System (TSCS) by Leistritz, was used to set and control processing parameters. Screw speed was 125 rpm and melt pressure varied from 320 to 350 psi depending on the WP melts. Extruded strands were passed through water bath and were subsequently palletized by Scheer Bay Co. pelletizer (model no BT-25). Table 3-2 shows the list of different barrel temperatures of the extruder during formation of pellets. Figure 3-1 shows the schematic diagram of WPC compounding.

Table 3-2 TSE barrel temperatures for palletizing

Temp (°C)/zones	1	2	3	4	5	6	7	8	9	Die
Palletizing Temp.	160	160	160	160	160	150	150	150	150	140

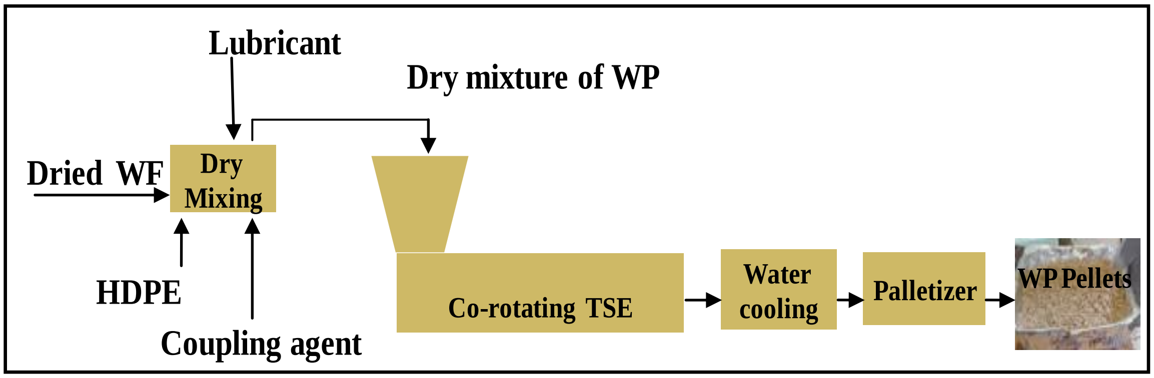


Figure 3-1 *Schematic diagram of WAPC compounding*

3-3 Formulating Profiles With and Without CBA

Rectangular profiles of 1 inch x ¼ inch cross-section were extruded with and without foaming agent. Initially, optimization was carried out on HDPE grade, lubricant content, WF content, and process set up, to obtain a proper profile without using CBA. In following steps foamed profiles were formed using CBA. Experiments were carried out to optimize CBA content, die temperature, profile draw down ratio (DDR) and WF content. Pellets were formed using a twin-screw extruder as mentioned earlier (see section 3-2 Formation of pellets) and were dried at 80 °C for 48 hrs. Dried pellets were fed in to the same extruder but different die arrangement through a feeder for profile extrusion.

3-3-1 Improving the Process Set Up

At first profile extrusion was carried out in an extrusion line of a twin-screw extruder (TSE). In addition to TSE, it included rectangular extrusion die/die land (19×6.4 mm), shaper (with water circulation to reduce temperature of WPC profile) and a downstream system (ONYXEDS-100C) by Onyx Extrusion Technology Inc. consisting of a vacuum calibrator (with air suction system to cool the WPC profile), a cooling water bath, a puller and a cutter. The WPC extruded through the die and die land was passed through the shaper followed by vacuum calibrator to get the shape and surface quality. Figure 3-2 shows die and die land. Profile was passed through the cold water bath before entering the puller. The calibrator is shown in Figure 3-3.

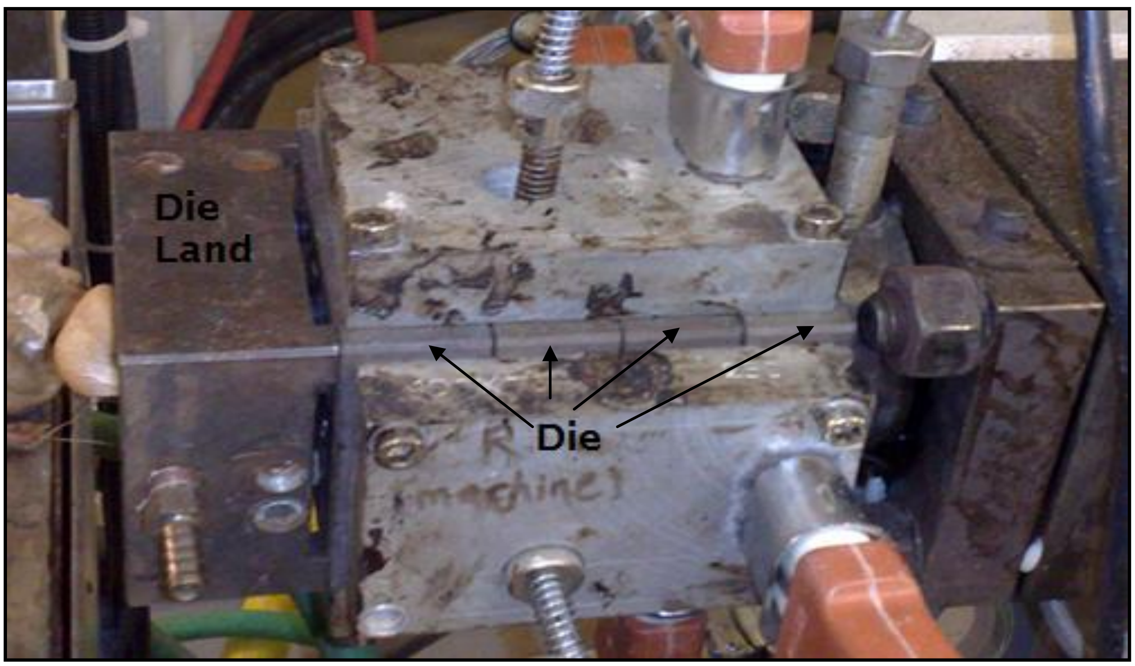


Figure 3-2 *Die and Die Land*

As we can see in Figure 3-3, the profile we got using existing system, has rough and non uniform surface quality, extrusion line was redesigned. In the new system, immediately after the shaper, WPC profile was immersed in to water rather than passing it through the calibrator. Also, instead of the calibrator, we have custom designed a guiding shaper, which we submerged in the water bath. The guiding shaper guided the material after the die, prohibited swelling and helped getting shape during solidification.

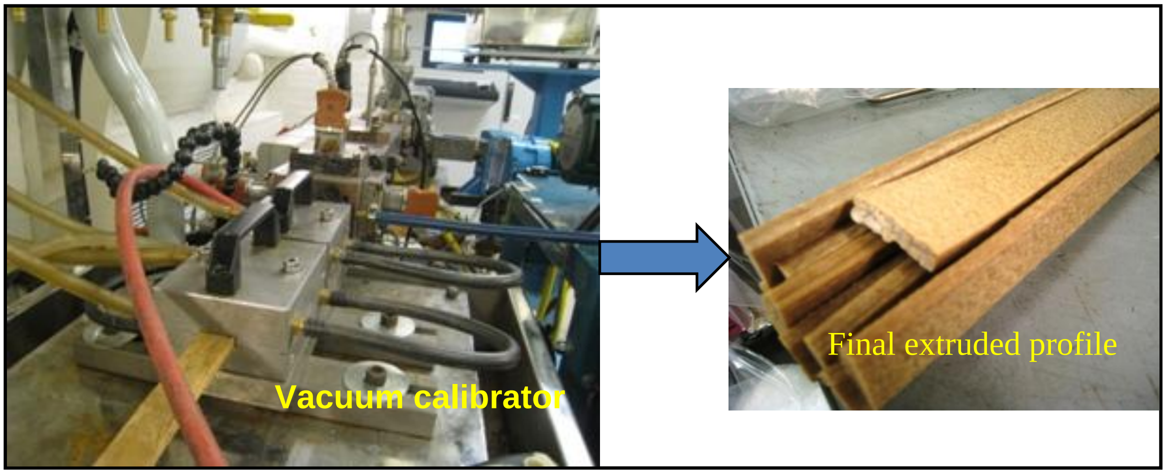
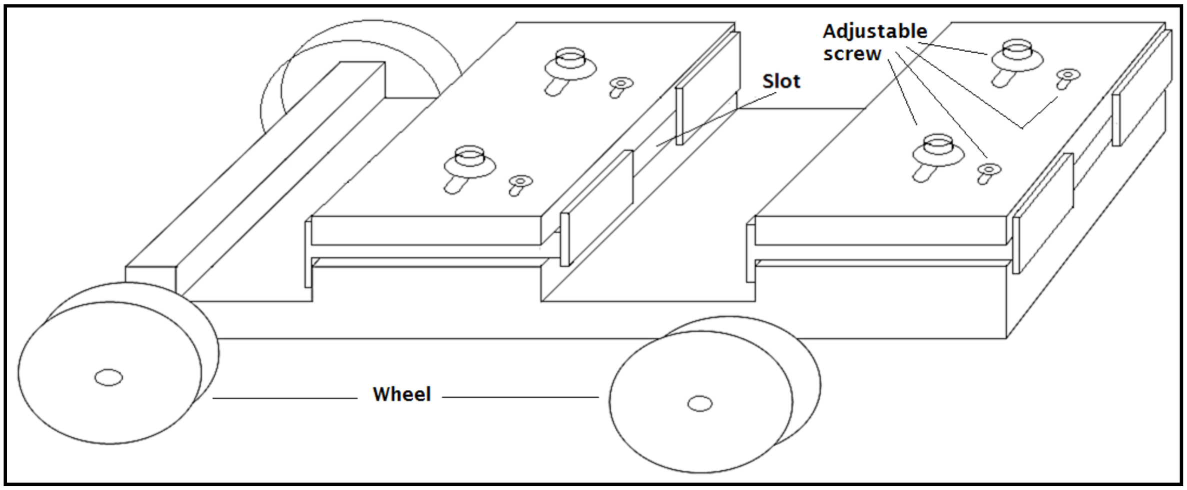
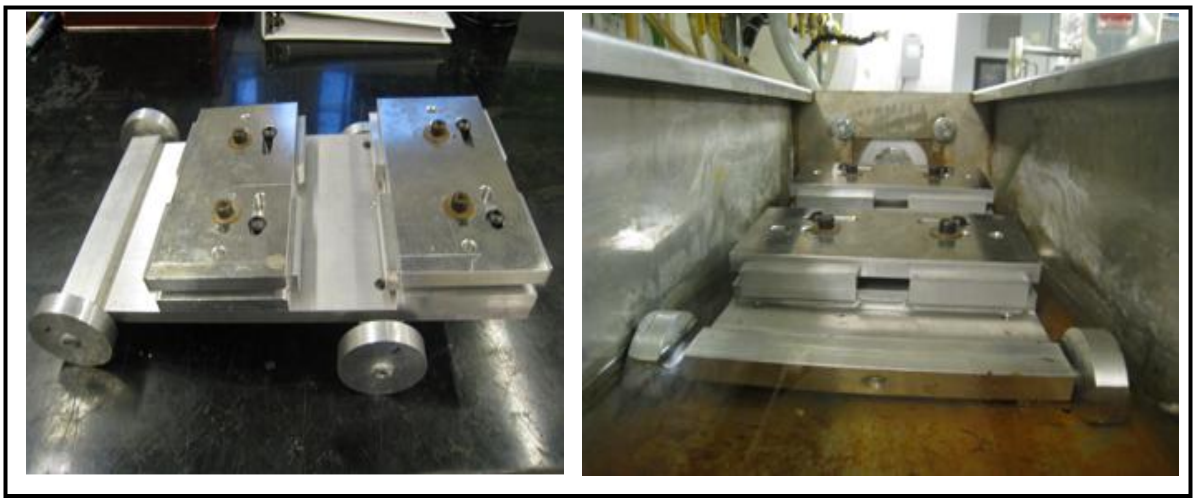


Figure 3-3 *Experimental conditions during profile extrusion (with Calibrator)*

So, the new profile extrusion line consisted of a twin-screw extruder, rectangular extrusion die/die land (19×6.4 mm), shaper (without air circulation), a cooling water bath, a guiding shaper, a puller and a cutter. The idea behind this was to improve surface quality and shape of the profile of WPC by dropping the temperature immediately after the shaper by directly immersing it in to water, instead of using the calibrator and suction cooling system and thus forming a surface skin quickly for better shape. Figure 3-4 shows the guiding shaper which was submerged in water. The guiding shaper is made of aluminum. There are two guiding zones with slots to facilitate better shape, (see figure 3-4(b) through which the materials are passed. Widths of the slots are adjustable with the aid of adjustable screw. During processing, as soon as the materials come out of the die through put, it passes through the guiding slot which is submerged in to the water. Figure 3-5 shows the modifications made in the extrusion system and the profiles obtained. Figure 3-6 is the schematic diagram of the ultimate profile extrusion line. Here, immediate after the die through put material is passed through water for quick solidification and then through the shaper guide to get proper shape. Then the profile passed through puller and cut by the cutting saw.



(a)



(b) *(c)*

Figure 3-4 *Schematic (a) & original diagram (b) and (c) of Custom built guiding shaper*

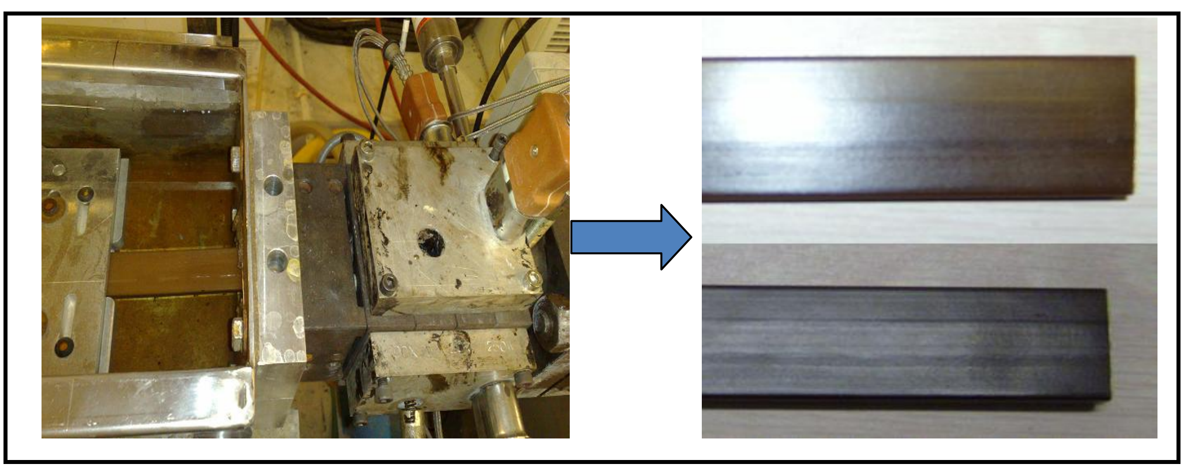


Figure 3-5 *Modifications of the extrusion system using the custom built guiding shaper*

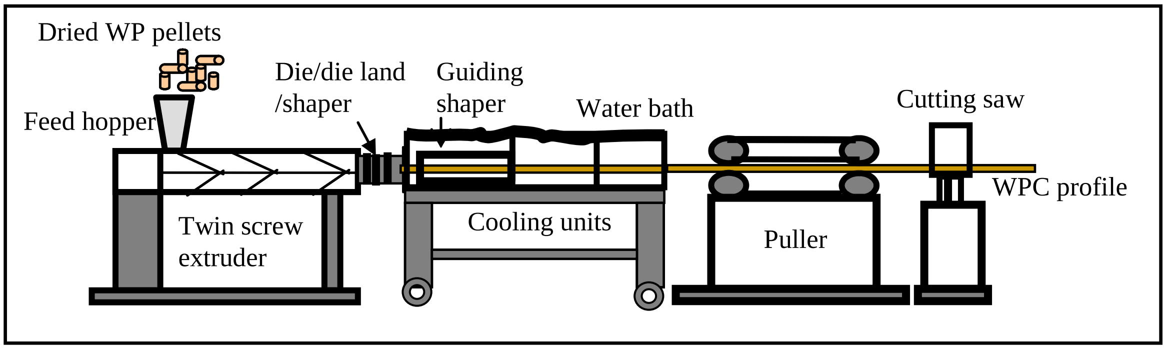


Figure 3-6 Schematic of WPC profile extrusion line

3-3-2 Compositions for Optimizing HDPE Grade

The WPC profiles were formulated using LDPE and HDPEs with different melt flow index (MFI) as shown in Table 3-3. WF-1 content was varied between 30% to 40 wt%, polymer contents between 50% to 63% and lubricant content (TPW 709) between 4-10 wt%. Amount of CA-1 content was fixed at 3 wt %. Table 3-4 shows the composition breakdown used to prepare WPC.

Table 3-3 *List of HDPE and LDPE polymers used*

Polymer grade/suppliers	Code used	MFI (g/10min, 2.16 kg/190°C)	Density (g/cm^3)
HDPE 2710/ Nova Chemicals	HDPE-1	17	0.951
DMDA-8007/ DOW Chemicals	HDPE-2	8.25	0.965
HB-W355-A/ Nova Chemicals	HDPE-3	3	0.955
HB-L354-A/ Nova Chemicals	HDPE-4	0.3	0.955
LDPE (609-A)/ DOW Chemicals	LDPE-1	0.9	0.924

Table 3-4 *WPC formulations to optimize HDPE grade (wt %)*

WPC sample code	Polymer grade	Wood flour content (%)	Polymer content (%)	Lubricant content (%)
HDPE1W40L7	HDPE-1	40	50	7
HDPE2W40L7	HDPE-2	40	50	7
HDPE3W30L4	HDPE-3	30	63	4
HDPE3W30L7	HDPE-3	30	60	7
HDPE3W30L10	HDPE-3	30	57	10
HDPE3W40L7	HDPE-3	40	50	7
HDPE4W40L7	HDPE-4	40	50	7
LDPE1W40L7	LDPE-1	40	50	7

3-3-3 Compositions for Optimizing Lubricant Content and WF Content

One of our objectives is to form WPC with maximum amount of WF. With increasing amount of WF higher lubrication is required. So with the variation in WF content, required amount of lubricant content may vary too. On the other hand, since presence of excessive amount of low molecular weight lubricant content may have impact on mechanical properties of WPC, so there could be a compromising maximum amount of lubricant content that we can use. WPC profiles were extruded varying the amount of lubricant and wood fiber content to find equilibriums. WF-1 content varied from 20-60 wt% and lubricant content (TPW 709) from 2-10 wt. %. Amount of CA-1 was fixed at 3 wt. %. The balance was HDPE-3. Table 3-5 contain the detail composition for WPC formulations.

Table 3-5 *WPC formulation for optimizing lubricant content and WF content (wt%)*

WPC sample code	**Wood flour content (%)**	**HDPE content (%)**	**Lubricant content (%)**
PE70W20LA7	20	70	7
PE60W30LA7	30	60	7
PE50W40LA7	40	50	7
PE40W50LA7	50	40	7
PE30W60LA7	60	30	7
PE65W30LA2	30	65	2
PE64W30LA3	30	64	3
PE63W30LA4	30	63	4
PE62W30LA5	30	62	5
PE61W30LA6	30	61	6
PE57W30LA10	30	57	10

3-3-4 Compositions for Optimizing CBA Content and Process Parameters (DDR and Die Temp)

Foamed Profiles were extruded using chemical blowing agent (CBA). WPCs were synthesized with various amounts to CBA content to optimize micro cellular foam structure with higher mechanical properties and surface qualities. Various extrusion parameters were studied which could have effect on foam cell formations during extrusion. Among them two of the most important parameters, DDR and die temperature, were optimized. The composite formulations were designed based on the parts per hundred parts of resin (phr) in mass proportions (Table 3-6). The amount of HDPE-3, WF-1, lubricant and CA-1 contents was fixed at 100, 50, 10 and 5 phr, respectively. CBA contents were varied from 0 to 6.5 phr as shown in Table 3-6.

Table 3-6 *Foamed wood-plastic composite formulation (phr by weight)*

Specimen Code	WF-1	HDPE-3	Lubricant	CA-1	CBA (Safoam PE-20)
F-1	50	100	10	5	0
F-2	50	100	10	5	1 (0.5%)
F-3	50	100	10	5	1.7 (1%)
F-4	50	100	10	5	2.5 (1.5%)
F-5	50	100	10	5	3.5 (2.0%)
F-6	50	100	10	5	5.0 (3.0%)
F-7	50	100	10	5	6.5 (4.0%)

3-3-5 Composition for Optimizing WF Content in Foaming

Profiles were extruded varying the WF and CBA contents to satisfy the objectives of producing WPC profiles with uniform foam cell structures using maximum amount of WF content and thus obtain light weight WPC with better surface quality and mechanical properties. Formulations were based on the parts per hundred parts of resin (phr) in mass proportion. The amount of HDPE-3, lubricant and CA-1 were fixed at 100, 10 and 5 phr, respectively. WF-1 content varied as 30, 50, 75 and 100 phr and CBA contents varied as 0, 1.7, 2.5, 3.5, 5.0 and 6.5 phr.

3-4 Compression Molding

In this study 2 types of compression molds were used: (i) Conventional hot press mold and (ii) Custom built die-press compression mold. At first pellets were prepared by dry blending all the materials which were extruded using the twin screw extruder as mentioned earlier (3-2 Formation of Pellets) and were dried at 80 ^{o}C for 24 hrs.

3-4-1 Formulation by Conventional Press

Pellets were compression-molded into sheets using a Carver Hydraulic press. Initial compression conditions were:

Temperature	: 160ºC
Pressure	: 4.5 metric tons
Time	: 1 minute

The compression pressure was then reduced to zero for another minute under the same temperature to purge away volatiles. This procedure was repeated for five minutes and materials were then compressed for ten straight minutes. After forming, the sheets where cooled to ambient temperature at less than 5 psi pressure using a Carver Hydraulic cold press and then cut into sample shape following the ASTM (American Society for Testing and Materials) standard for tensile and flexural tests.

3-4-1-1 Composition of Materials Used

Pallets composed of 74.5 wt% HDPE-1, 20wt% WF-1, 3 wt% CA-2 and 2.5wt% GF-1 was used to prepare compressed sheets.

3-4-2 Description of Custom Built Die-Press Compression Mold

A custom built compression molding system was developed that imparts directional flow of fibers in the polymer matrix and was used to make samples that would be more representative of the property of WPC achieved from profile extrusion. There are also potentials of preparing end use product with this equipment. The system comprises of a hydraulically-operated press, a block-die system and a pneumatically operated melting/mixing system (Figure 3-7 and 3-8).

Press

The press has a usable stroke of eight inches with a maximum loading capacity of 14 tons. Heating and cooling is PID controlled. Closing speed of the upper platen can be varied to a wide range. The die comprises of an upper platen and a bottom platen. The top and bottom platens each contain 8 cartridge heaters. During the experiments the heating rate as approximately 10°C/min. Cooling was achieved by blowing compressed-air which results in a cooling rate of about 3°C/min.

There are five 1/2 x 1/8 x 14 inch channels on the bottom platen in which the material flows in one direction which leads to better alignment of the fibers (Fig.3-9). At the feed side (right side in Figure 3-10) of the press there is a heated chamber where pellets were molten, agitated and then loaded into the die through a slot at the top side of the melting chamber. The material is then pressed by the top block of the mold causing it to flow through the channels towards left. Due to the melt flow under the applied pressure, the fibers in the matrix gained some alignment in the longitudinal direction of the specimen [50]. Before pressing, both the die parts are sprayed with silicon to help ease of material removal after cooling.

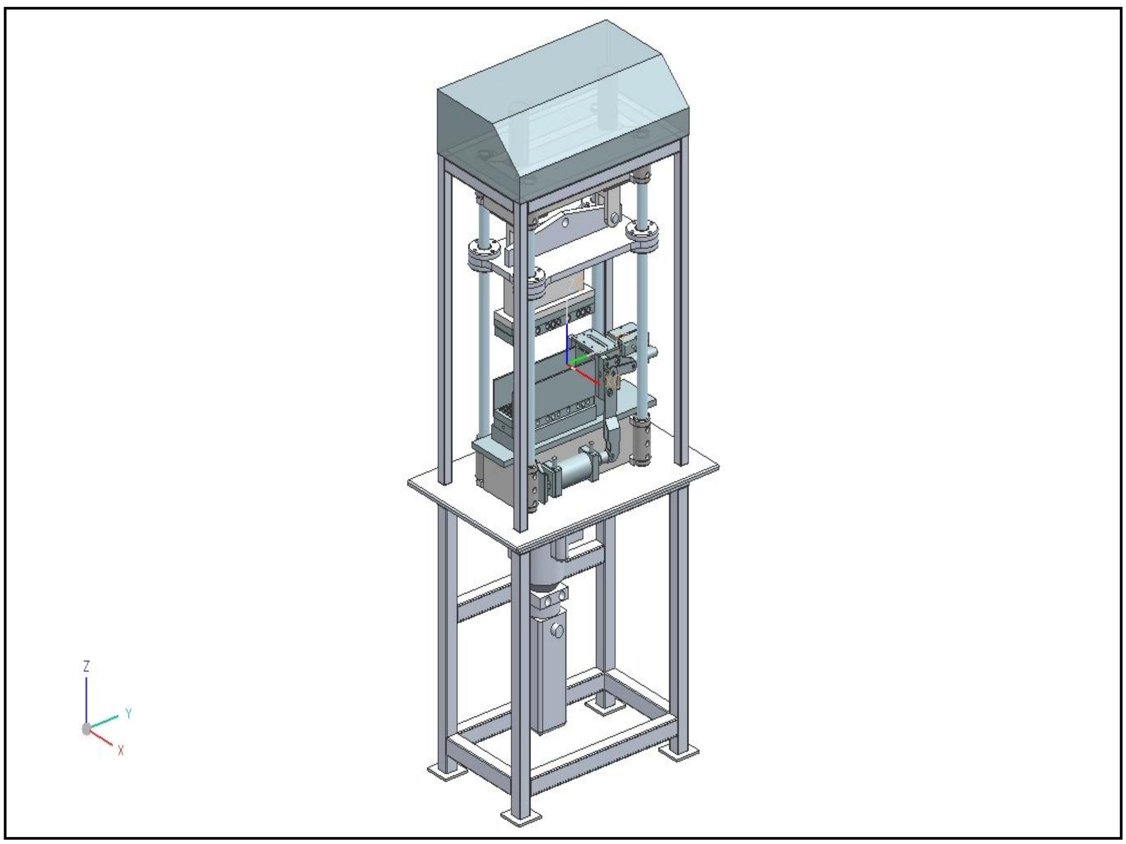


Figure 3-7*: Schematic Diagram of Press and block-die system in custom built compression molding*

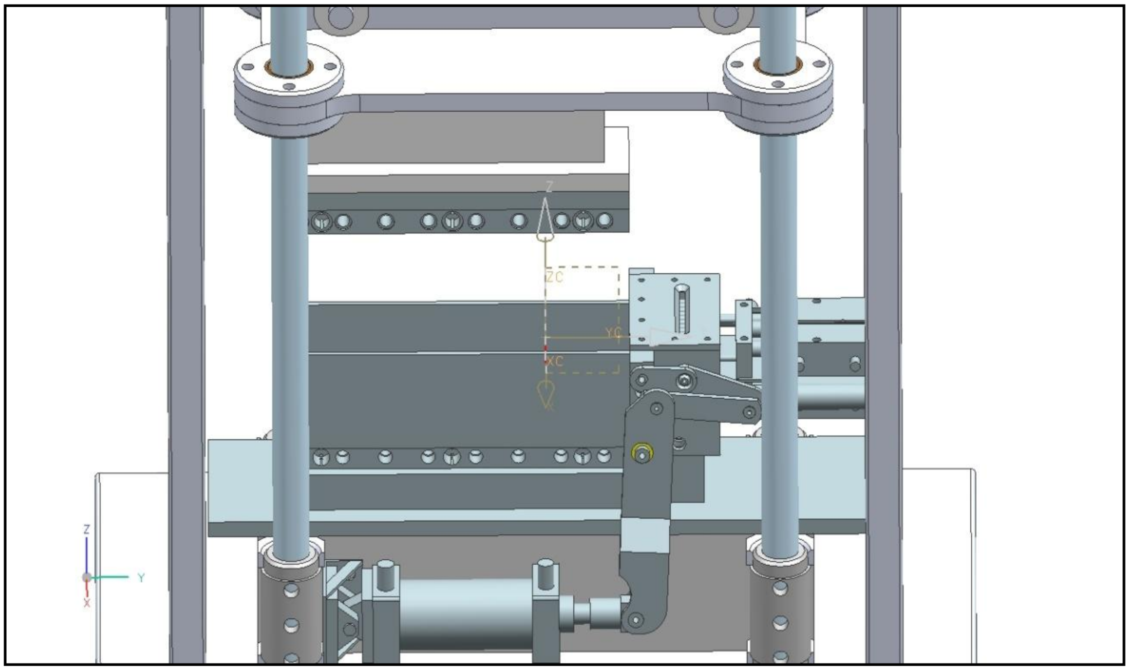


Figure 3-8- *Upper and bottom platen of the die*

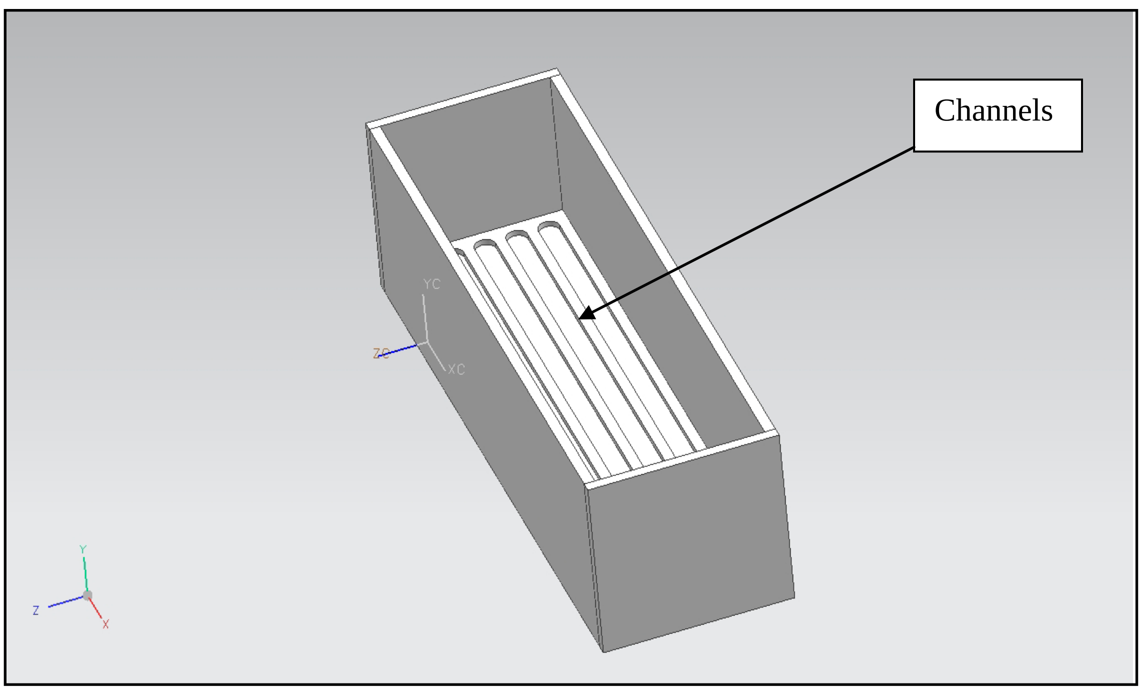


Figure 3-9: *Die used in heating-cooling press (Die –press)*

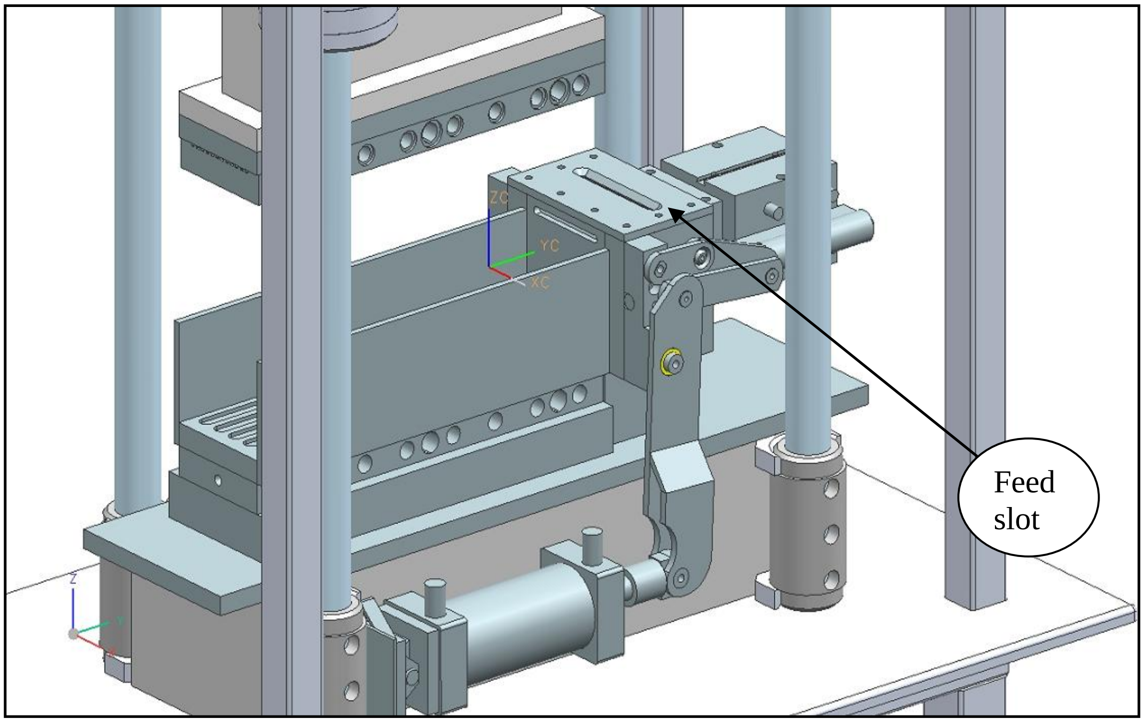
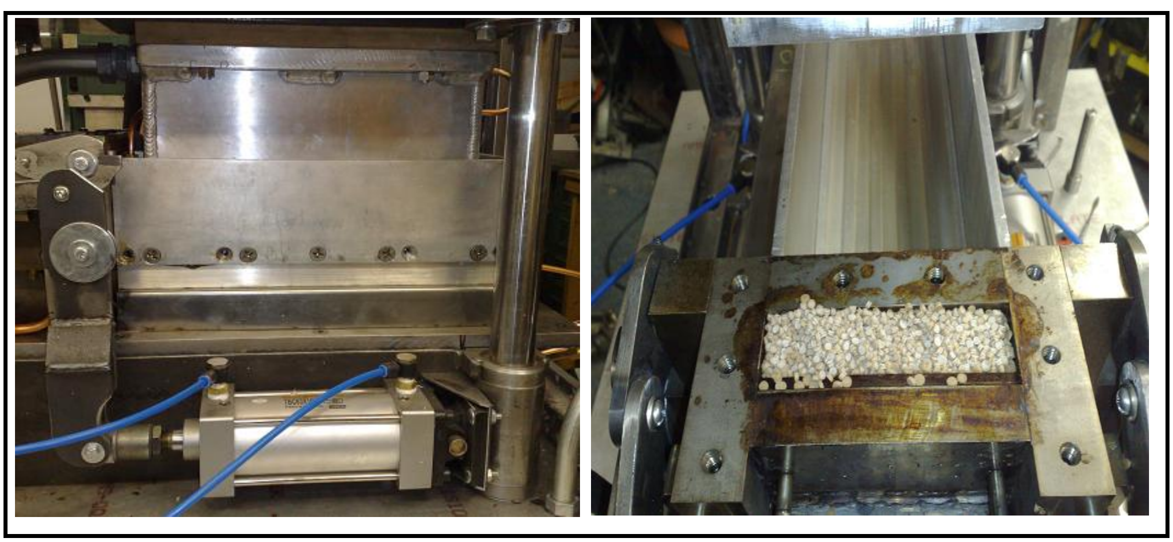


Figure 3-10 *Feed slot from the heating chamber*

3-4-2-1 Sample preparation

Samples were prepared using the custom built compression molding system and by following the process mentioned above. Upper and lower die temperatures were always kept the same. The actual die is shown in figure 3-11.



(a) *(b)*

Figure 3-11 *Custom Built Compression mold (a) and the heating chamber (b)*

3-4-2-1-1 Formulations to determine the Effect of Variation in WF, GF and HDPE Grades

Effects of different WF & GF length and MFI of HDPE on WPC were studied. Samples were prepared varying HDPE, WF and GF grades. Coupling agent (CA-2) grade was kept the same. 74.5 wt% HDPE, 20 wt% WF, 2.5 wt% GF and 3 wt% CA-2 (PE74.5W20G2.5CA3) were used. Table 3-7 contains detail composition of WPC.

Table 3-7 *Composition with different material grade*

	Sample Code	Polymer	WF	GF	CA
Group-1 (Variation in HDPE)	**S-1**	**HDPE-1**	**WF-1**	**GF-1**	**CA-2**
	S-2	**HDPE-3**	**WF-1**	**GF-1**	**CA-2**
Group-2 (Variation in WF)	**S3**	**HDPE-1**	**WF-1**	**GF-1**	**CA-2**
	S4	**HDPE-1**	**WF-2**	**GF-1**	**CA-2**
Group-3 (Variation in GF)	**S5**	**HDPE-1**	**WF-1**	**GF-1**	**CA-2**
	S6	**HDPE-1**	**WF-1**	**GF-2**	**CA-2**

3-4-2-1-2 Formulations for Optimizing Process Parameters (Speed and Die Temperature)

Experiments were carried out to optimize die closing speed and die platen temperature of the custom built compression mold. 74.5 wt% HDPE-1, 20 wt% WF-1, 3 wt% CA-2 and 2.5 wt% GF-1 were used.

3-4-2-1-3 Formulations for comparison between Custom built Compression Mold and Profile Extrusion

Experiments were carried out to compare mechanical properties among WPCs prepared by profile extrusion and custom built compression molding varying WF content.

HDPE-3, WF-1, lubricant and CA-1 were used with following compositions. The compositions for comparison have been mentioned in Table 3-8.

Table 3-8 *WPC Compositions for comparison*

WPC sample code	Wood flour content (%)	HDPE content (%)	Lubricant content (%)	CA (MAPE) content (%)
PE70W20LA7	20	70	7	3
PE60W30LA7	30	60	7	3
PE50W40LA7	40	50	7	3
PE40W50LA7	50	40	7	3
PE30W60LA7	60	30	7	3

3-5 Characterizing Methods

3-5-1 ASTM Methods

3-5-1-1 Tensile Tests

Tensile tests were carried out using a LLOYD Instruments LS100 (1with 100 KN load cell) Universal Testing Machine (General Purpose with Yield Setup) according to ASTM D638, with the cross-head speed of 5 mm/min. In case of both conventional and custom built compression molding Samples were cut into dog bone shape conforming to ASTM D638:

Type –IV For profile extrusion samples

Gauge Dimension: 115mm × 19mm × 6.4 mm (thickness)

Type – V For conventional and custom built compression molding samples

Gauge Dimension: 14 mm x 2.6 mm x 3.2 mm (thickness)

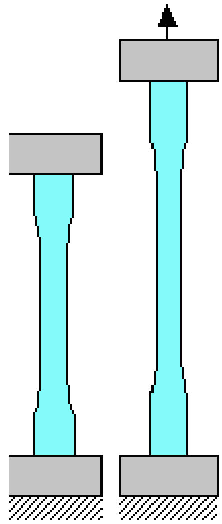


Figure 3-12 *Dog bone shaped samples for tensile testing ASTM D638*

3-5-1-2 Flexural Tests

The flexural tests were carried out using the same instrument, a LLOYD Instruments LS100 Universal Testing Machine with 100 KN load cell by changing the set up (General Purpose Three Point Bend Setup) according to ASTM D790; samples were then tested until the appropriate deflection was achieved. In case of compression (both conventional and custom build process), sheets were cut according to ASTM D790, each sample had roughly the following dimensions: 124 mm x 12 mm x 3.2 mm. In case of profile extrusion, sample size was 120 mm × 12.7 mm × 6.4 mm(thickness). Both the tensile and flexural testing results were obtained using NEXYGEN MT software and analyzed using the same software and Excel. 5 samples were tested for each composition. Measurements were performed at ambient conditions (23±2°C and 50% RH).

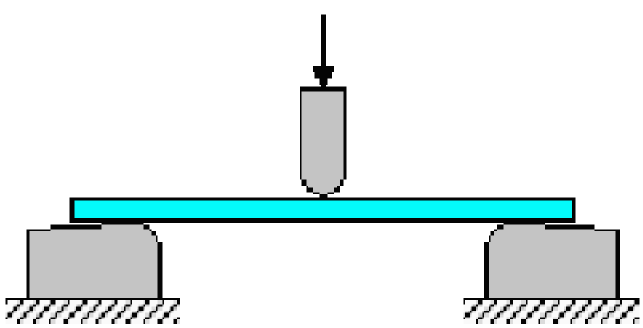


Figure 3-13 *Flexural testing ASTM D790*

3-5-2 Morphology

Samples were immersed in liquid nitrogen and fractured to ensure that the microstructure remained intact. Then samples were sputter coated by platinum using a sputter-coater. SEM pictures were used to investigate morphology, study direction of fiber alignment, number and shape of foamed cells/bubbles/voids and whether a cellular structure has been achieved or not. SEM pictures were taken using JEOL JSM 6060 **Scanning Electron Microscope** (SEM), operated at an accelerating voltage of 20 kV and emission current of 47 μA

3-5-3 Void Fraction and Cell Density

Density of samples was measured by a water displacement technique according to ASTM D-792 and using a Mettler Toledo balance. Sample mass was measured in air and then in distilled water to calculate the density. Using density value void fraction and cell density was calculated.

Chapter 4 Experimentations and Results

In this chapter results of the experiments and characterization tests, as described in chapter 3, have been analyzed. Improvement in profile extrusion set up was made to obtain profiles with smooth and even surface and uniform cross section. The matrix polymer grades were evaluated for optimum profile extrusion. Lubricant contents and WF content were also optimized. Once profile with smooth and even surface and uniform cross section was achieved, foaming with CBA was carried out. CBA content, process parameters (e.g. die temperature, draw down ratio (DDR)) and WF content was optimized. Results of samples formed by custom built die-press compression molding system were also analyzed and compared with samples from conventional hot press and profile extrusion. Optimization of process parameters in custom built die-press compression mold was carried out. SEM pictures were taken to study foam cell structure, alignment of fiber in composite matrix and others.

4-1 Process Ability of WPC: Profile Extrusion

4-1-1 Optimizing MFI of HDPE

Melt Flow Index (MFI) is an important parameter for optimizing profile extrusion process. WPC with HDPEs of different MFI ranging from 0.3 to 17 and with LDPE of 0.9 MFI were tested (Table 3-10). Tensile tests were carried out using a standard Lloyd Instrument following ASTM D638 [96] and flexural tests were carried out following ASTM D790.

The extruder barrel and die temperatures, screw speed and feeding rate (Table 4-1) were adjusted to guarantee the fixed level of extrudate throughput with different composite formulations. The velocity of the haul-off unit was adjusted in order to achieve the drawdown ratio (DDR) of 1.0.

Table 4-1 *Processing parameters for different composite formulations*

WPC sample code	Extruder zones/ Barrel temperature (°C)										Melt Pr. (psi)	Feed rate (kg/h)	Screw speed (rpm)
	1	2	3	4	5	6	7	8	9	Die/die land			
HDPE1W40L7	165	165	170	175	175	170	160	160	160	160/160	320	1.5	70
HDPE2W40L7	175	175	175	175	170	165	160	160	160	160/160	220	1.0	50
HDPE3W40L7	175	175	175	175	170	170	170	170	170	175/175	475	0.8	30
HDPE3W30L4	165	165	165	165	160	160	160	165	170	175/175	663	0.8	40
HDPE3W30L7	180	180	175	175	175	175	170	170	175	175/175	430	1.0	70
HDPE3W30L10	175	175	175	170	170	170	165	160	170	170/170	450	0.8	80
HDPE4W40L7	175	175	175	165	165	165	165	160	170	170/170	320	0.8	30
LDPE1W40L7	165	165	170	170	165	165	165	160	160	170/170	330	0.8	30

The mechanical test results of WPC with LDPE & HDPEs of various MFI indicated that MFI of polymer has significant influence on tensile and flexural properties of the composites (Table 4-2).

Table 4-2: *Flexural and Tensile test results by varying HDPE grades and using LDPE*

WPC sample	Tensile properties		Flexural properties	
	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)
HDPE1W40L7	13.6 (1.7)	1.18 (0.05)	21.4 (1.2)	1.19 (0.23)
HDPE2W40L7	16.0 (2.5)	1.67 (0.21)	29.4 (3.5)	1.18 (0.04)
HDPE3W30L4	13.5 (1.4)	1.13 (0.11)	24.1 (1.2)	1.13 (0.16)
HDPE3W30L7	15.1 (1.5)	1.05 (0.03)	21.0 (1.8)	1.10 (0.06)
HDPE3W30L10	10.0 (0.4)	1.32 (0.10)	17.9 (2.5)	1.13 (0.14)
HDPE3W40L7	19.2 (1.8)	1.33 (0.12)	35.4 (1.3)	1.45 (0.08)
HDPE4W40L7	21.9 (1.5)	1.39 (0.10)	36.7 (1.9)	1.30 (0.02)
LDPE1W40L7	10.9 (0.9)	0.99 (0.11)	16.5 (1.4)	0.85 (0.06)

It was observed that composite with low MFI HDPEs show comparatively higher tensile and flexural strength than composites with high MFI HDPEs. It is also apparent that WPCs with all the HDPEs show significantly higher strength than WPC with LDPE (Figure 4-1 and 4-2).

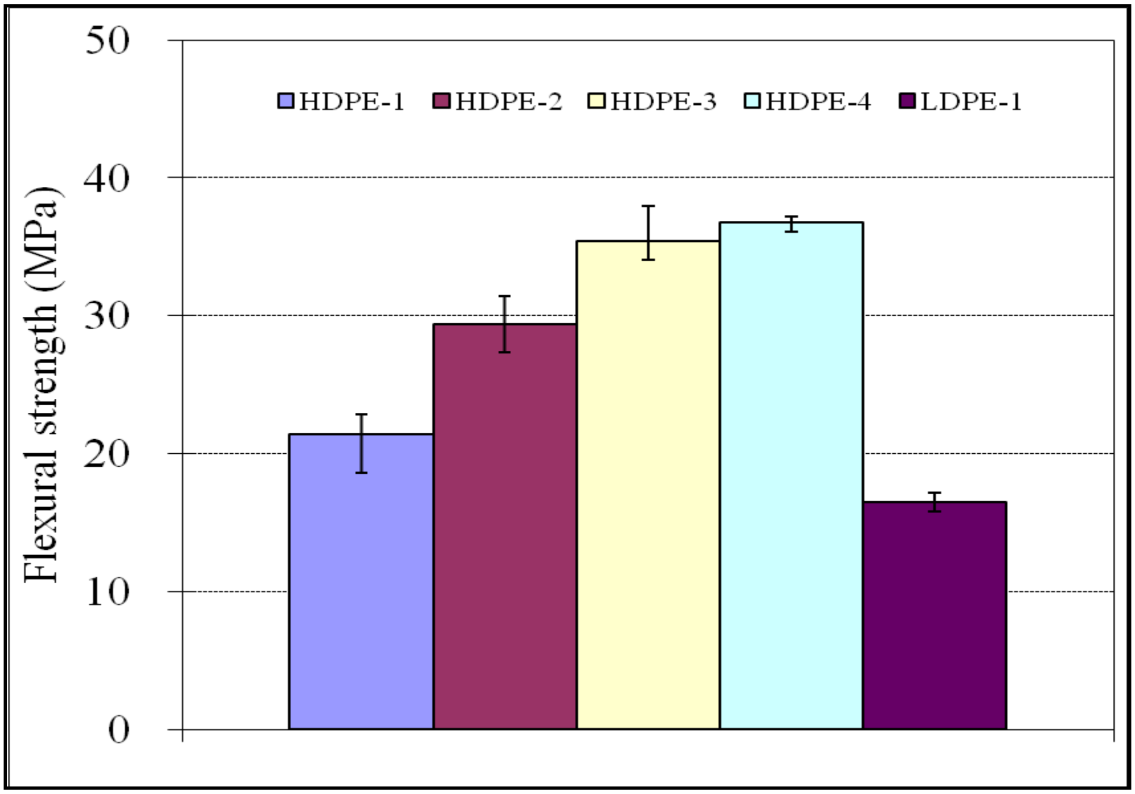


Figure 4-1 *Effect of polymer MFI on flexural strength of WPC at 40% WF content*

Samples with HDPE-4 showed almost 72% increase in flexural strength than samples with HDPE-1. WPCs with HDPE-3 exhibit 65% higher strength than WPCs with HDPE1. Tensile strength followed the similar trend. The possible reason could be that low MFI polymer had higher molecular weight and polymer chain entanglement, which results in the higher mechanical properties of composites if there is good WF distribution [97]. However, we have chosen to work with HDPE-3; though WPC with HDPE-4 showed higher flexural and tensile strength than the others. The reason behind this was to achieve our target to formulate WPC with increasing WF content. But while processing with HDPE-4, we have found it difficult to form WPC even with 40 wt% WF due to the high apparent viscosity. Although, lubricant was used to ease processing; but use of

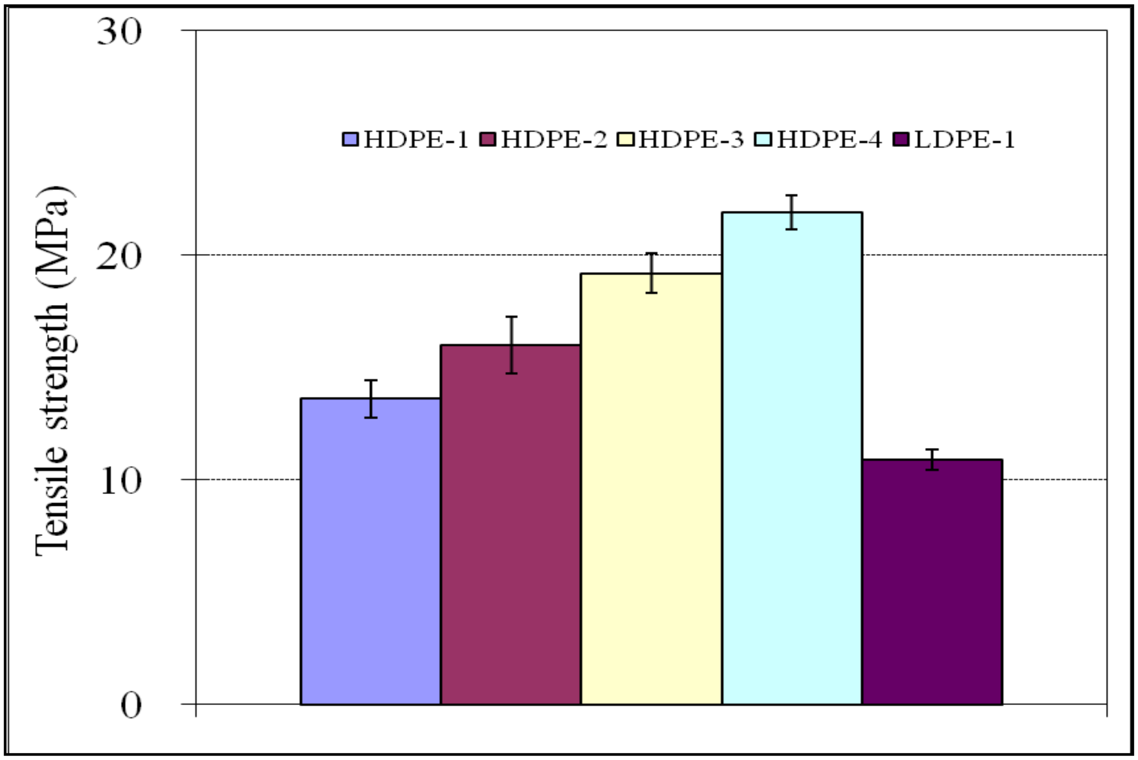


Figure 4-2 *Effect of polymer MFI on tensile strength of WPC at 40% WF content*

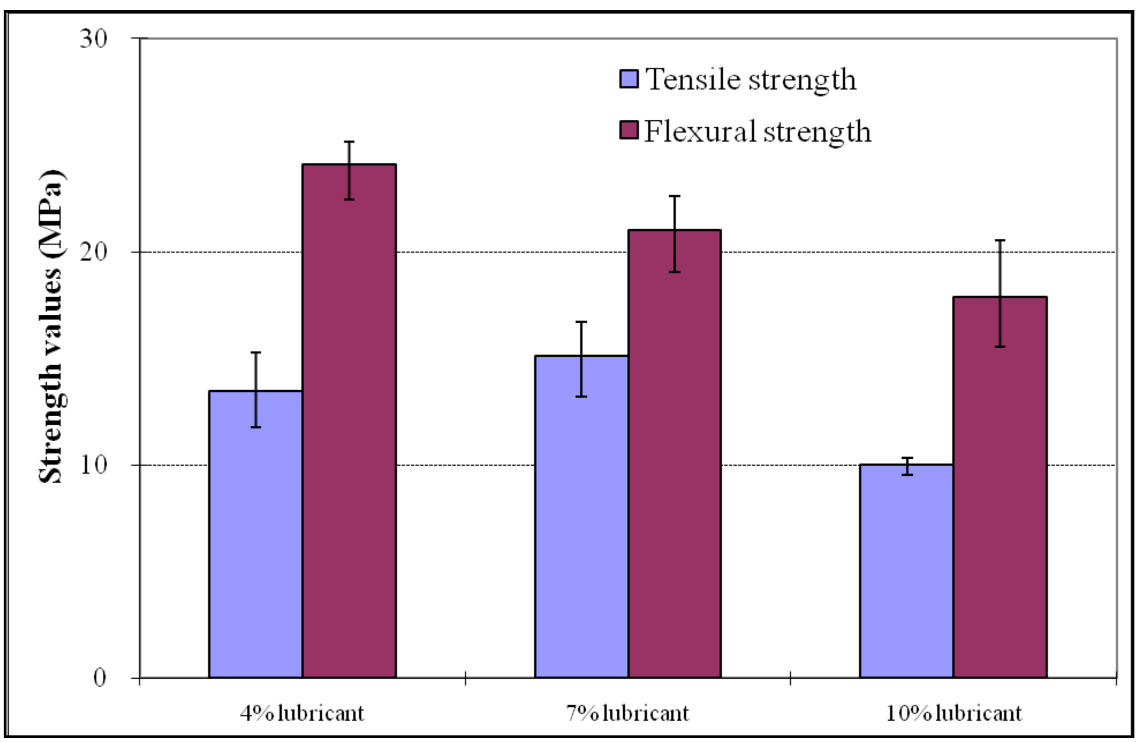


Figure 4-3 *Effect of lubricant contents on tensile and flexural strengths of WPC (30 wt. % WF in HDPE-3, MFI: 3.0)*

higher amount of lubricant reduced the strength performance of the composite (Figure 4-3). This could be attributed due to reduction of molecular weight due to presence of low molecular weight lubricant than those of the base resins. We had to find a balance between lubricant content mechanical performance and MFI of polymer. We worked further to optimize the amount of lubricant.

In addition, flexural modulus was found to be higher in WPC with HDPE-3 (1.45 GPa) than with HDPE-4 (1.30 GPa) (Figure 4-4).

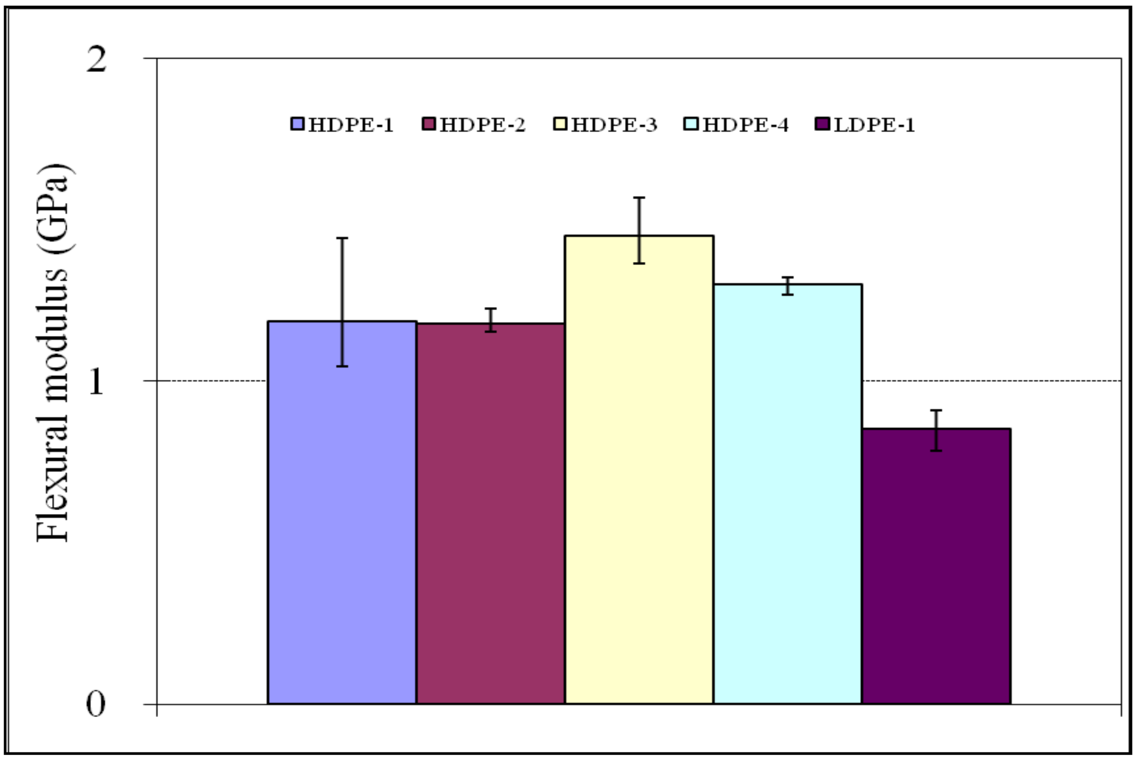


Figure 4-4 *Effect of polymer MFI on flexural modulus of WPC at 40% WF content*

4-1-2 Optimizing Lubricant Content

Effects of lubricant contents on the processing and mechanical properties of WPCs were investigated. The contents were varied from 2-10 wt. % with 30 wt% WF-1 and 3 wt. % CA-1 in HDPE matrix. Tensile strength was found to decrease (Figure 4-6) though flexural strength of the composite was increased (Figure 4-5) when lubricant content was increased from 2 to 7 wt. %.

Extrusion process conditions:

Extruder Temperature:
Zone 1-4 : 165 ^{o}C
Zone 5-7 : 160-165 ^{o}C
Zone 8 : 150-160 ^{o}C
Zone 9 : 150 ^{o}C
Die/Die Land: 145-160 ^{o}C

Screw speed (rpm) : 75
Draw down ratio (DDR) : 1.0
Melt Pressure : 394-871 psi

Both tensile and flexural strengths were decreased at 10 wt. % of lubricant contents. However, no clear trend was observed for tensile and flexural modulus with the increase in lubricant contents. The tensile modulus was decreased slightly up to 5 wt. % lubricants, and then increased slightly with increasing lubricant contents; however, variation remained within range of experimental errors. The flexural modulus was also increased slightly with the increasing lubricant contents up to 7 wt. %, but within the range of experimental errors. The slight variation in strength and modulus values probably resulted from the poor wetting and fibers dispersion. The poor bonding between HDPE matrix and WF due to insufficient wetting might have caused the lower tensile strength values at low lubricant contents. The drop in molecular weight of base resin with the presence of low molecular weight lubricant might have also aided the reduction. Further, a small amount of the lubricant is retained at the interface, which reduces the interfacial bonding between the WF and polymer matrix [98]. On the other hand, increase in flexural strength and both tensile and flexural modulus could be possible due to the synergistic effects between the lubricant and the coupling agent when added together that increased the bonding between HDPE matrix and WF due to better wetting and WF dispersion.

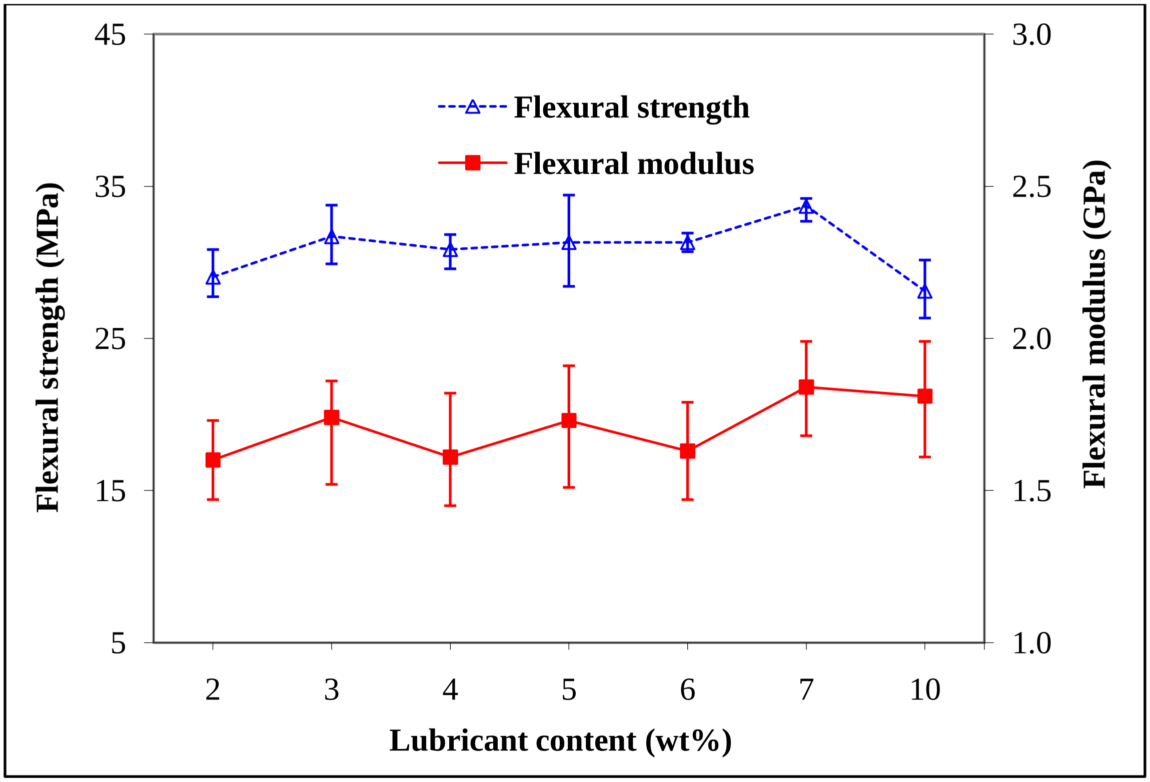


***Figure 4-5** Effect of lubricant contents on flexural properties of WPC*

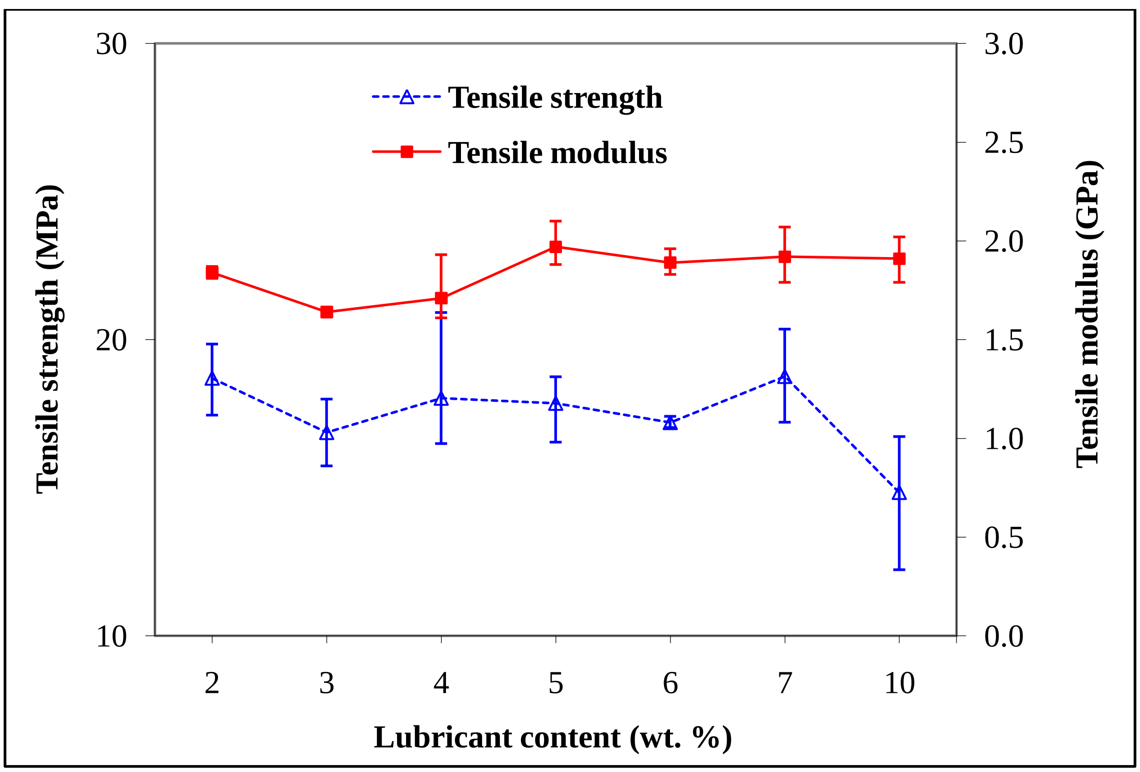


***Figure 4-6** Effect of lubricant contents on tensile properties of WPC*

The melt pressure decreased with the increase in lubricant content which can be explained by the inherent contribution of lubricant in increasing the wall slip which likely lowered the melt pressure, while the internal lubrication effect of MAPE also aided in this reduction [99].

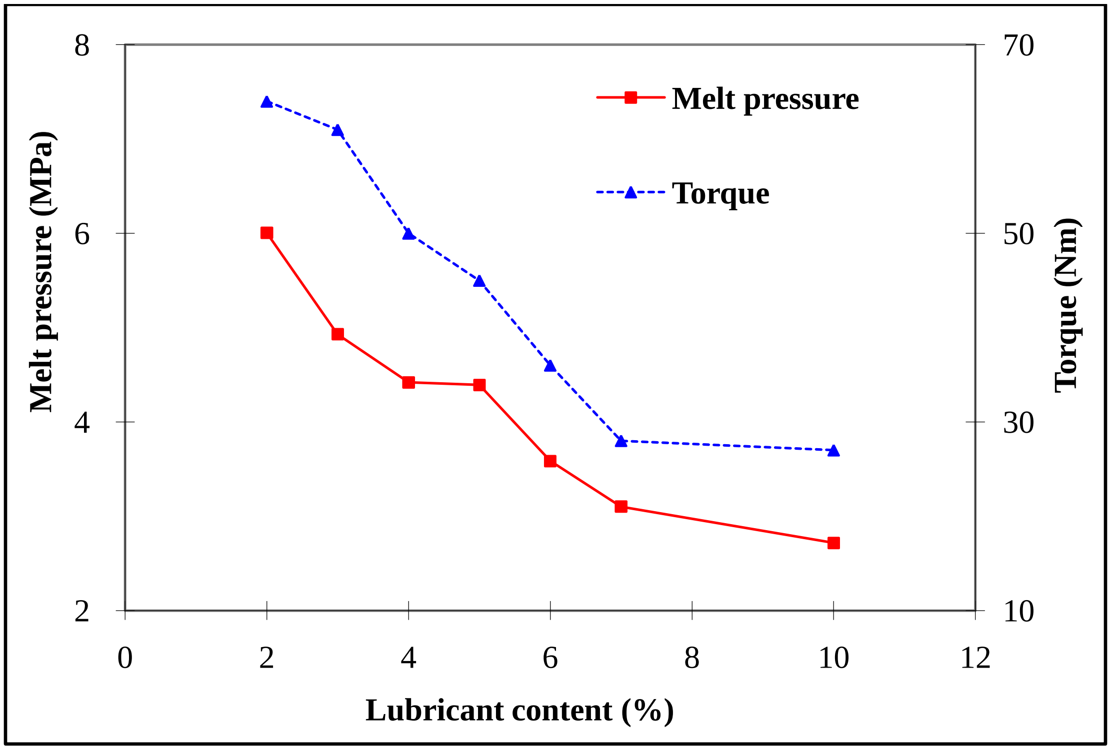


Figure 4-7 *Effect of lubricant contents on melt pressure and torque of WPC*

4-1-3 Optimizing WF Content

The tensile and flexural properties of WPC profiles were studied by varying WF-1 contents from 20-60 wt. % in HDPE-3 matrix with fixed CA-1 and lubricant contents. The variation in tensile and flexural strengths with different WF contents is shown in Figure 4-8. Tensile strength remained the same with increase in WF-1 contents from 20 to 50 wt. % but reduced by when increased from 50 to 60 wt. %. On the other hand, flexural strength was increased gradually up to 50 wt. % WF content but reduced by 41% when WF increased to 60 wt %. Further, the tensile and flexural modulus increased with

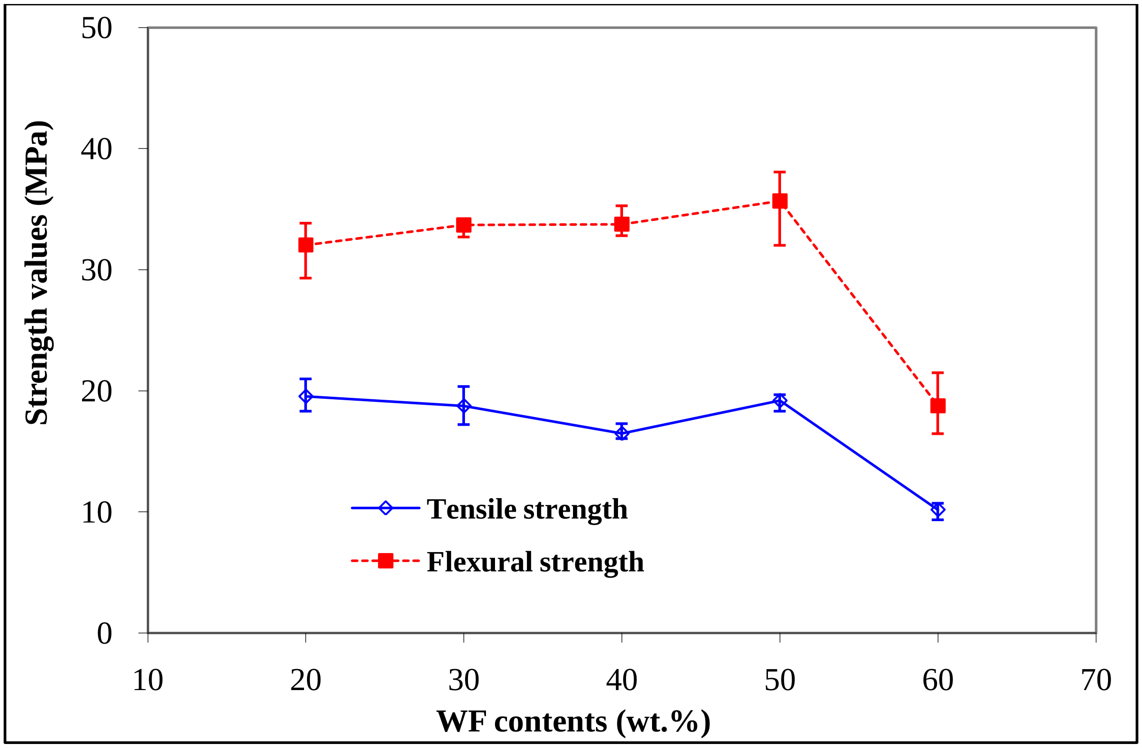


Figure 4-8 *Effect of WF contents on tensile and flexural strengths of WPC*

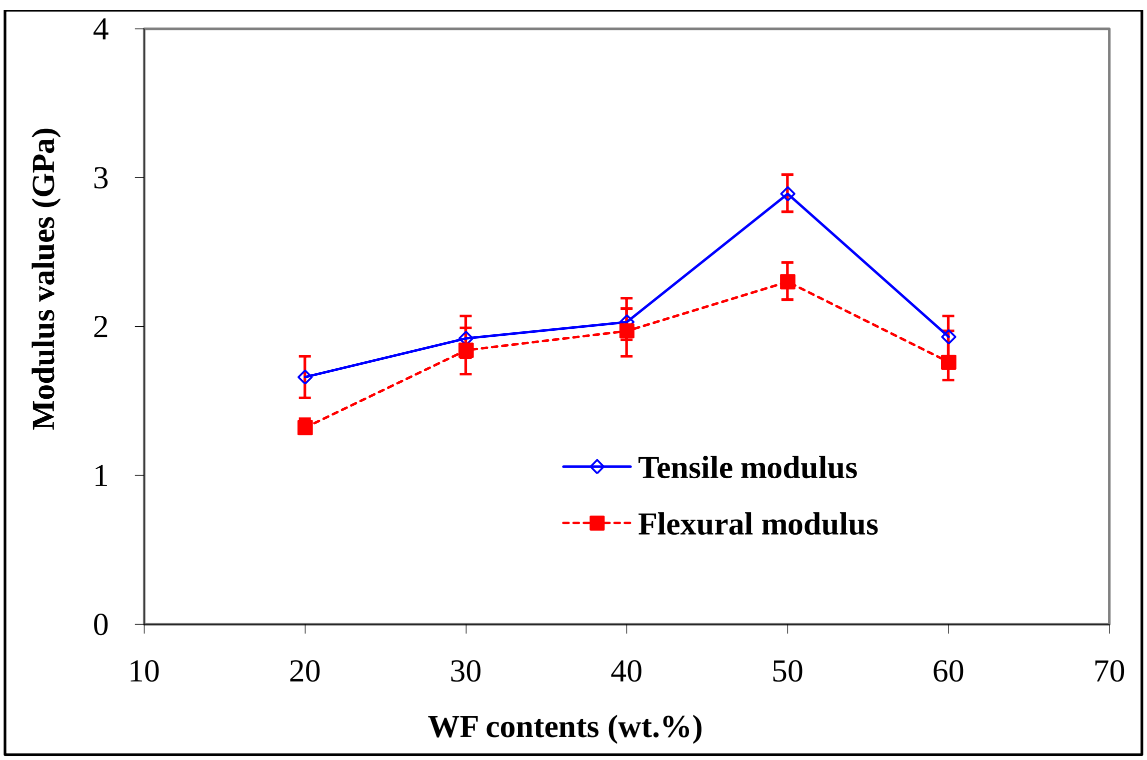


Figure 4-9 *Effect of WF contents on tensile and flexural modulus of WPC*

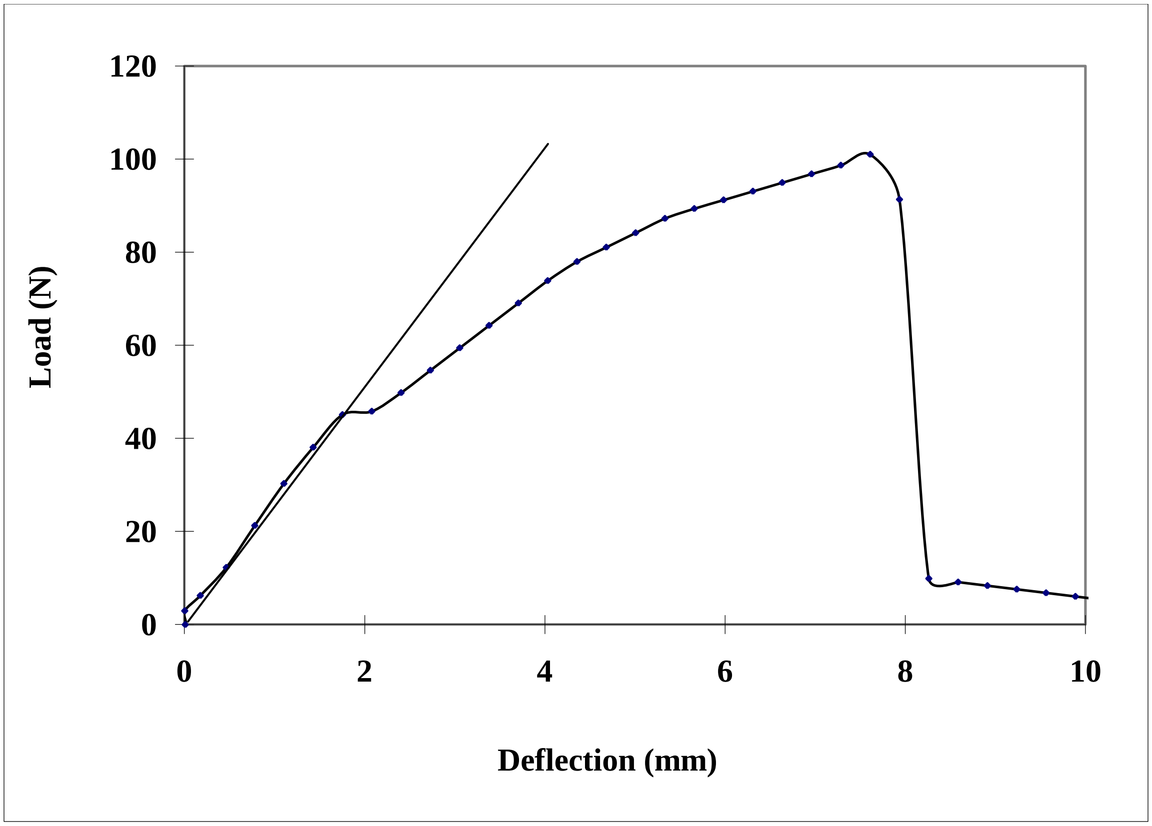


Figure 4-9 *Load vs deflection curve to calculate modulus value of WPC with 50% WF*

the increase of WF contents from 20 to 50 wt. % (Figure 4-9). However, both tensile and flexural modulus decreased by 33% and 23.5%, when WF contents increased from 50 to 60 wt. %. The optimal tensile and flexural properties were obtained with 50 wt. % WF at 7 and 3 wt. % lubricant and CA contents. The improvement in mechanical properties could be because of the stiffness of WF and composite homogeneity (WF distribution and wetting). But high WF content (60 wt. %) led to an increase in WPC melt viscosity and difficulty of wetting and fibers dispersion in polymer matrix. In such case, bonding strength between WF and HDPE could be weak due to the presence of many dry fibers, which could lead to property reduction. Moreover high WF contents increased the possibility of voids [100]. These could reduce the strength of WPC with high (60 wt %) WF content.

4-1-3-1 Comparison between Theoretical and Experimental Results of Tensile Modulus of WPC

Figure 4-10 shows the comparison between theoretical results and experimental results of tensile modulus. Theoretical results have been obtained following Rule of Mixture (ROM) principle (as described in Chapter 2 Literature Review, 2-2-1).

In all cases the experimental strength value was found to be in between upper and lower bounds except when WF content exceeded 60% WF, where the modulus value was found to be below the lower bound. This could be due to additional voids, generated from the high wood fiber content, which results a decrease in tensile modulus of WPC.

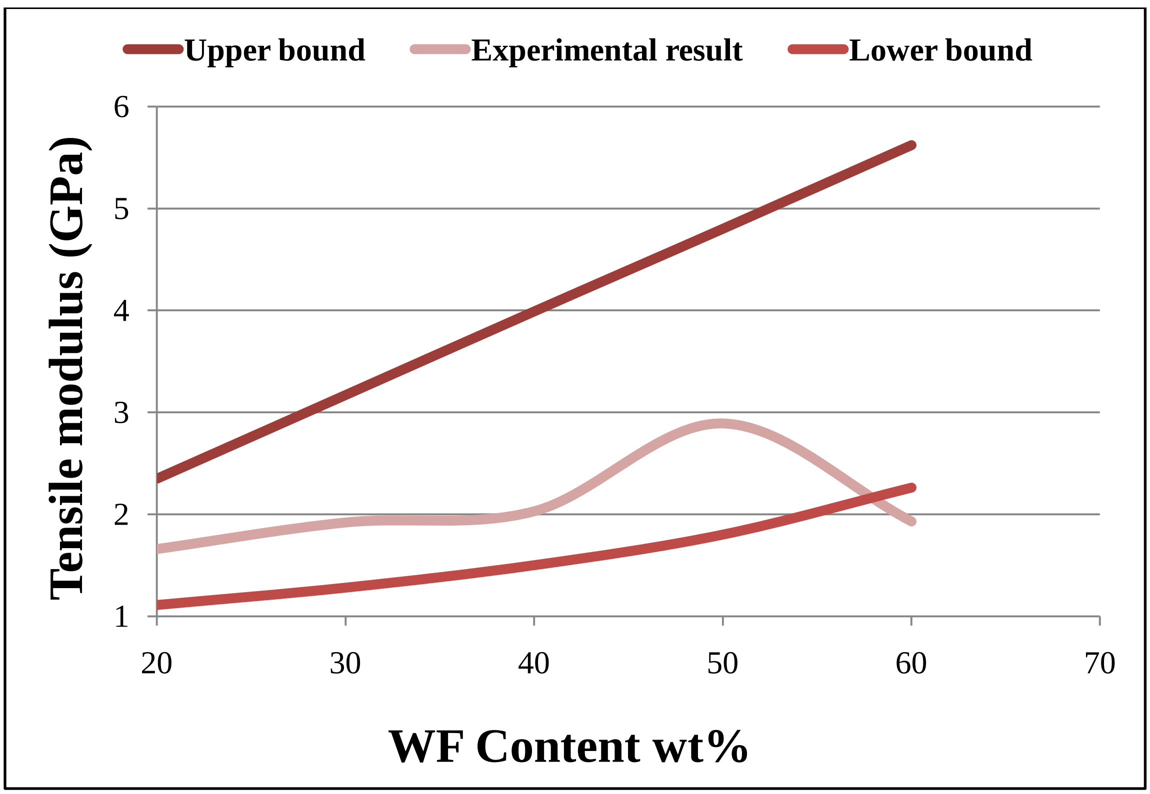


Figure 4-10 *Comparison between theoretical modulus and experimental modulus*

4-1-4 Formulations for Optimizing Process Parameters (Die Temp. & DDR) & CBA Contents

WPC dried pellets were mixed with CBA and foam extrusions runs were carried on the new profile extrusion set up with water submerged shaper. The processing conditions were kept almost same in all cases as listed in Table 7.

Table 4-3 *Process conditions for different foamed profile extrusion run*

Extruder zones/ Barrel temperature (^oC)									Screw speed (rpm)
1	2	3	4	5	6	7	8	9	
160	160	160	160	160	175	170	170	160	70

Extrusion runs were conducted by varying the die temperature from 150 to170 ^oC and DDR from 1.0- 1.5 for different CBA contents.

4-1-4-1 Optimizing Die Temperature and CBA Contents

Torque was monitored at 3 different die temperatures, 150 ^oC, 160 ^oC and 170 ^oC with and without CBA content (1.7 phr and 2.5 phr). It was observed that as the die temperature increased from 150 ^oC to 170 ^oC, torque decreased for all CBA contents studied (Figure 4-11). Higher die temperature reduced the melt viscosity, as a result the WPC melt flow smoothly through the die without any increase in torque during processing. Flexural properties were also investigated by varying the die temperatures and CBA contents (0 -6.5phr) at DDR of 1.0 (Fig.4-12). But no clear trend was observed.

However, in case of WPC with CBA content 6.5, flexural strength did not change noticeably beyond 160 ^oC where as with CBA content 3.5 strength decreased as die temperature increased.

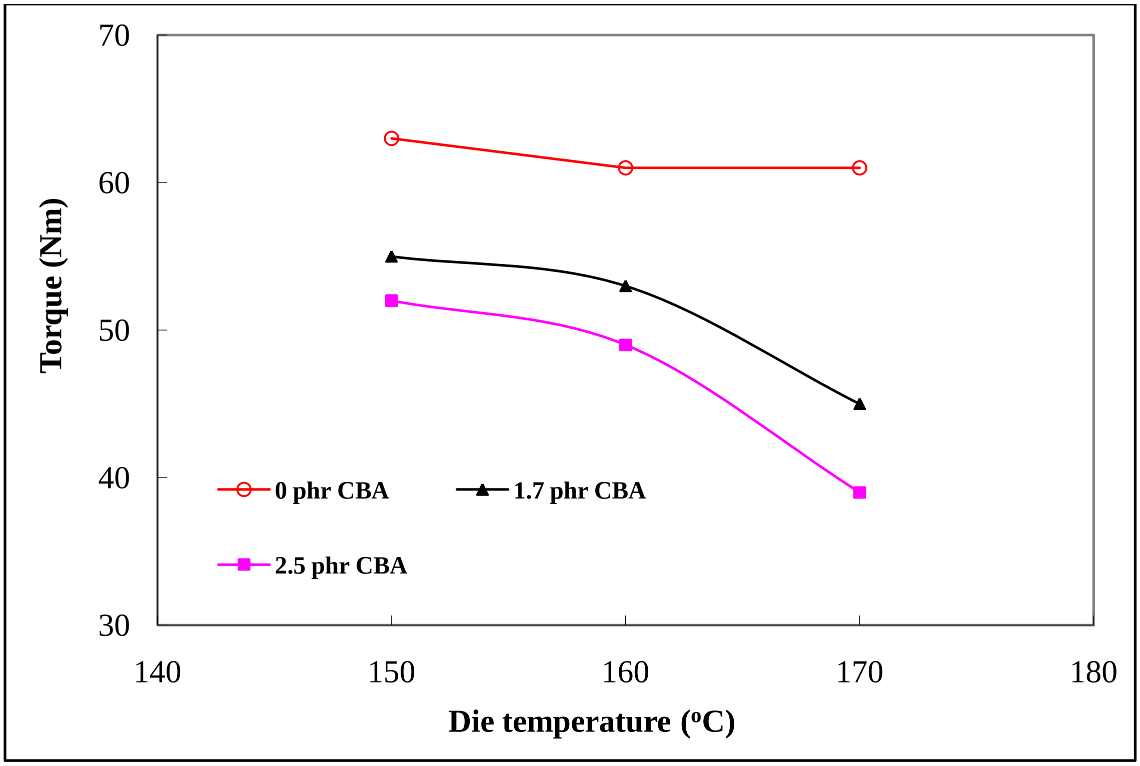


Figure 4-11 *Torque versus Die temperature for WPC foamed with different CBA content*

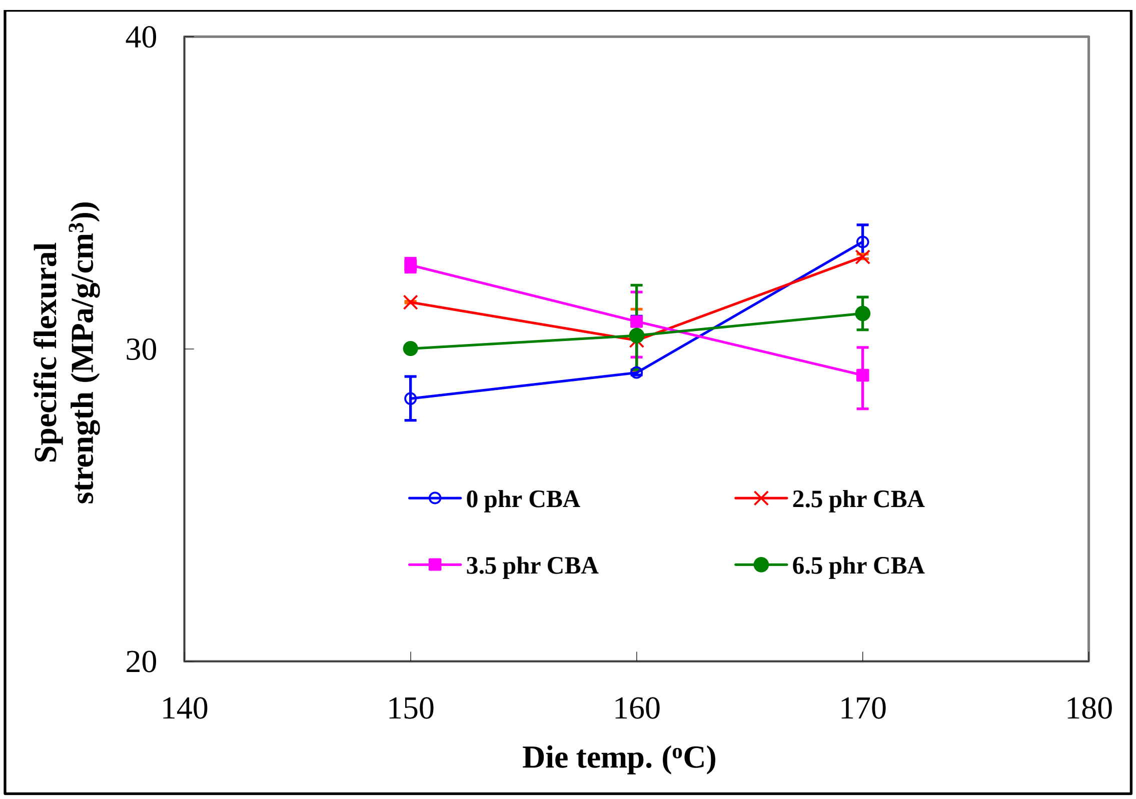


Figure 4-12 *Flexural strength vs. die temperature for WPC with different CBA content*

4-1-4-2 Optimizing DDR & CBA content

Effect of Draw Down Ratio (DDR) on density, surface quality and mechanical strength was studied on WPC prepared with 50phr (~ 30 wt%) WF content and with various CBA content (1-6.5 phr) and DDR. Density reduced with the increase in CBA contents and DDR values (Figure 4-13) ranging from 1.5 to 21 %(~ 0.820 g/cm^3 with 6.5 phr at 1.2 DDR, figure 4-16). Density was reduced slightly up to 2.5 phr CBA, then significantly as CBA contents increased. During foaming, at first cells are nucleated and then continue to grow by diffusion of gas into the nucleated cells leading to a reduction in density. However, at higher CBA content there is an increase in density which could be due to the coalescence and collapse of already formed cells. Change of density was found to be nearly independent of DDR though gradual reduction of density was observed with an increase in DDR at high CBA contents which could be due to collapse of foam cell. Figure 4-17 shows the collapse of foam cells at DDR 1.4 at CBA content 6.5 phr. The variation in density was insignificant (within 2.5 %) up to 1.7 phr CBA content with the increase of DDR from 1.0 to 1.3, whereas it was up to 4.5-13 % beyond 2.5 phr of CBA content. The density reduction with increasing DDR could be possible due to stretching [93]. Also micro-pores could be formed in the HDPE-3 polymer by crystalline orientation, alignment and lamellae separation by hot stretching [101]. At higher CBA content there is a slight increase in density which could be due to the coalescence and collapse of already formed cells.

The specific flexural strength was altered with the increase in DDR and CBA content. Specific flexural strength increased slightly with the increase in CBA content at 1.0 DDR. Our findings supports the postulate of Bledzki *et al.* [102] , where he concluded that the flexural properties of injection molded foamed PP-WF-MAPP composites slightly increased at low WF content (30 wt. %) with the increasing CBA contents. For non- foamed composites, both strength and modulus were decreased with increasing DDR values. However, foamed composites showed different trend. Generally, specific flexural strength was increased with the incorporation of CBA as compared to non-foamed WPC at 1.0 DDR. Maximum specific flexural strength was obtained with 3.5

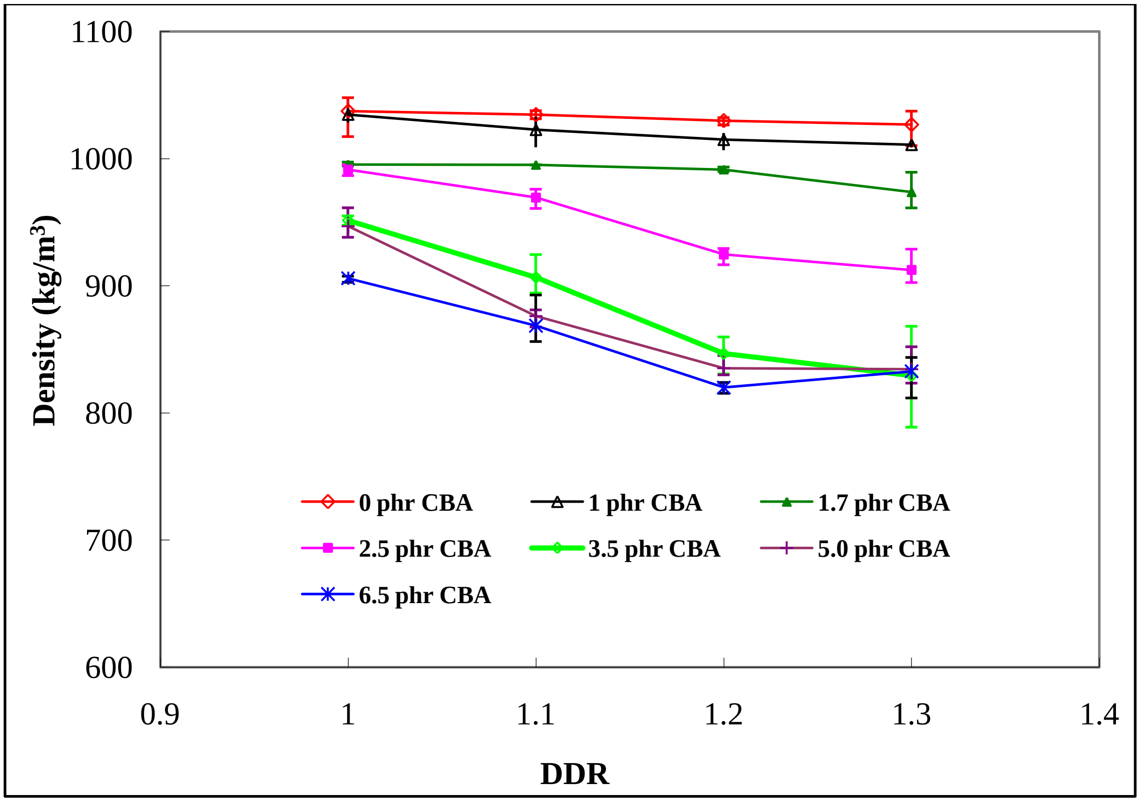


Figure 4-13 *Density versus DDR for WPC foamed with different CBA content*

phr CBA at 1.0 DDR and decreased with further increase in CBA contents. No clear trend was observed for the specific flexural modulus with the CBA contents at 1.0 DDR (Fig. 4-15).The rate of increase or decrease in these values largely depended on the CBA contents. This says that there should be an optimal DDR value for different CBA contents. The optimal flexural properties were obtained at DDR 1.1-1.2 depending on the CBA contents. The reason of this increase in flexural strength could be the presence of smaller and uniform cellular structure. However, with CBA content of 6.5 phr, the strength of WPC seemed to drop. This could be due to collapse of cell structures, resulting degraded morphology.

Void fraction was estimated from the density data in respect to variation in DDR and CBA content (Figure 4-13). Void fraction was found in a range of up to 25 % depending on CBA content.

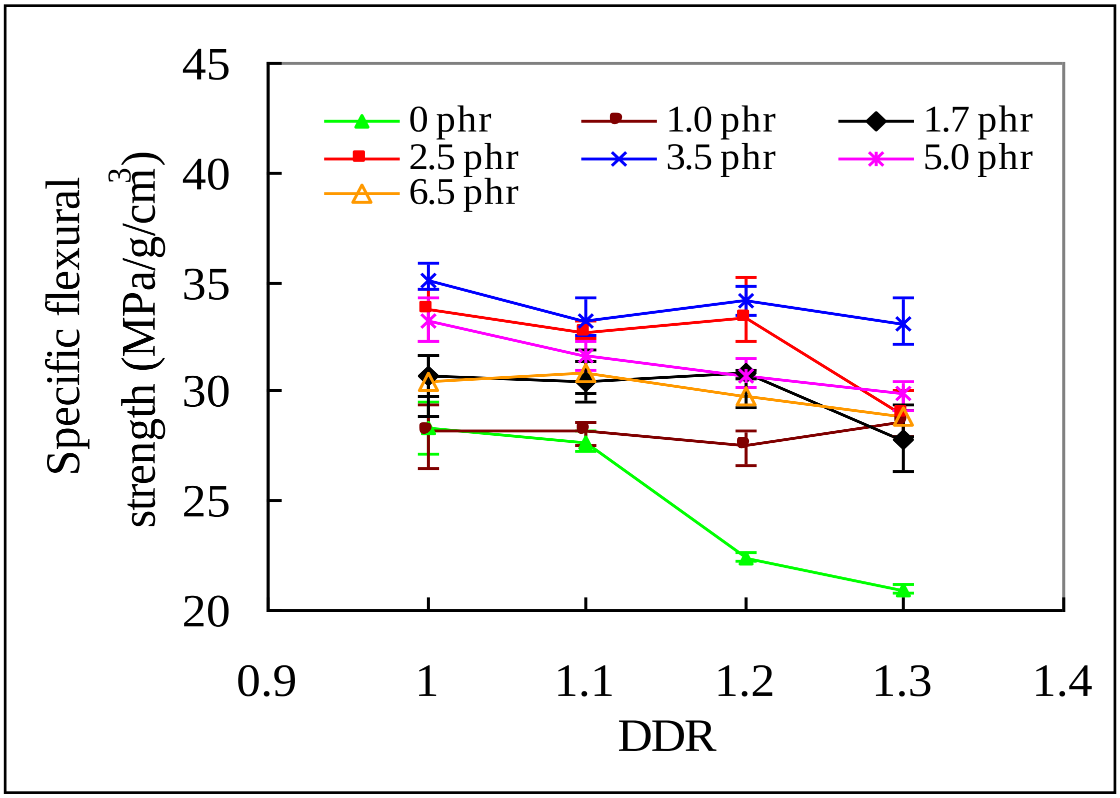


Figure 4-14 *Effect of DDR on specific flexural strength*

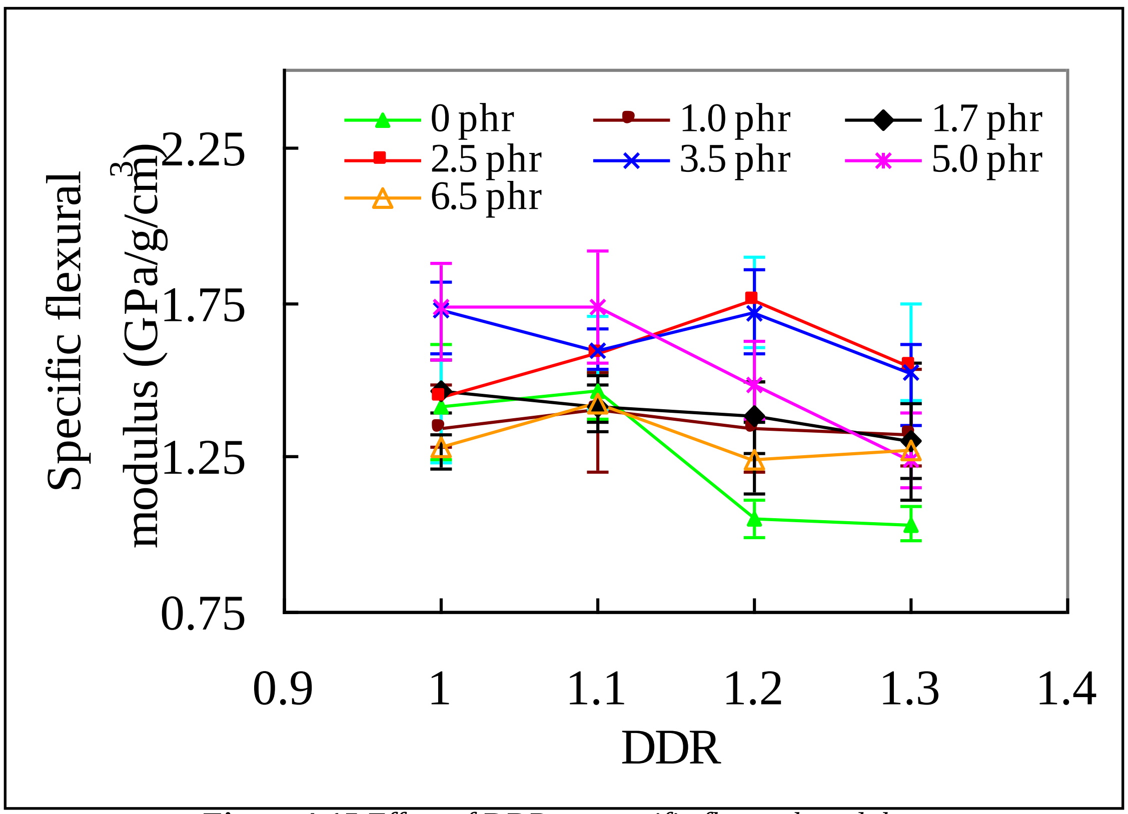


Figure 4-15 *Effect of DDR on specific flexural modulus*

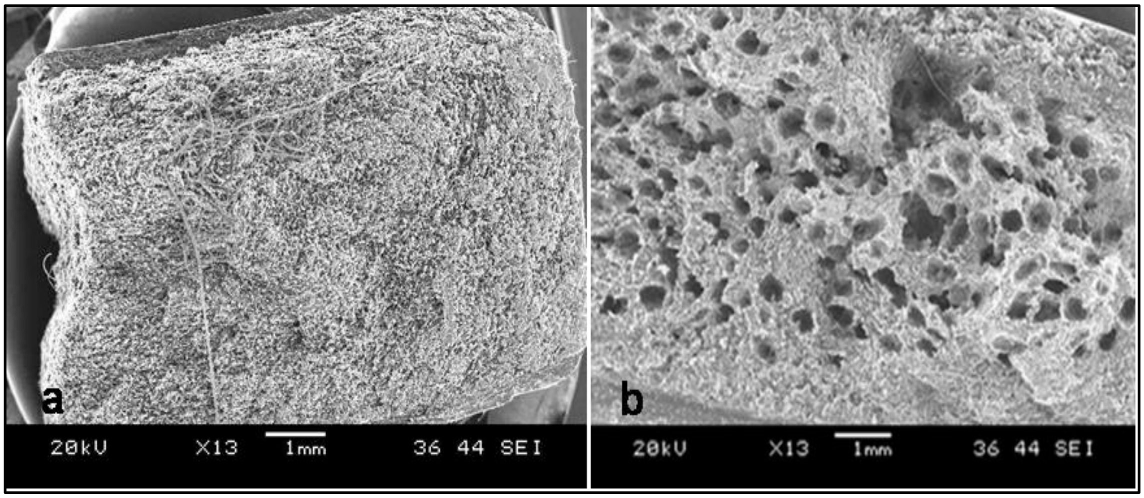


Figure 4-16. *SEM images of foamed WPCs at 1.2 DDR for (a) 0 and* (b) 6.5 phr CBA

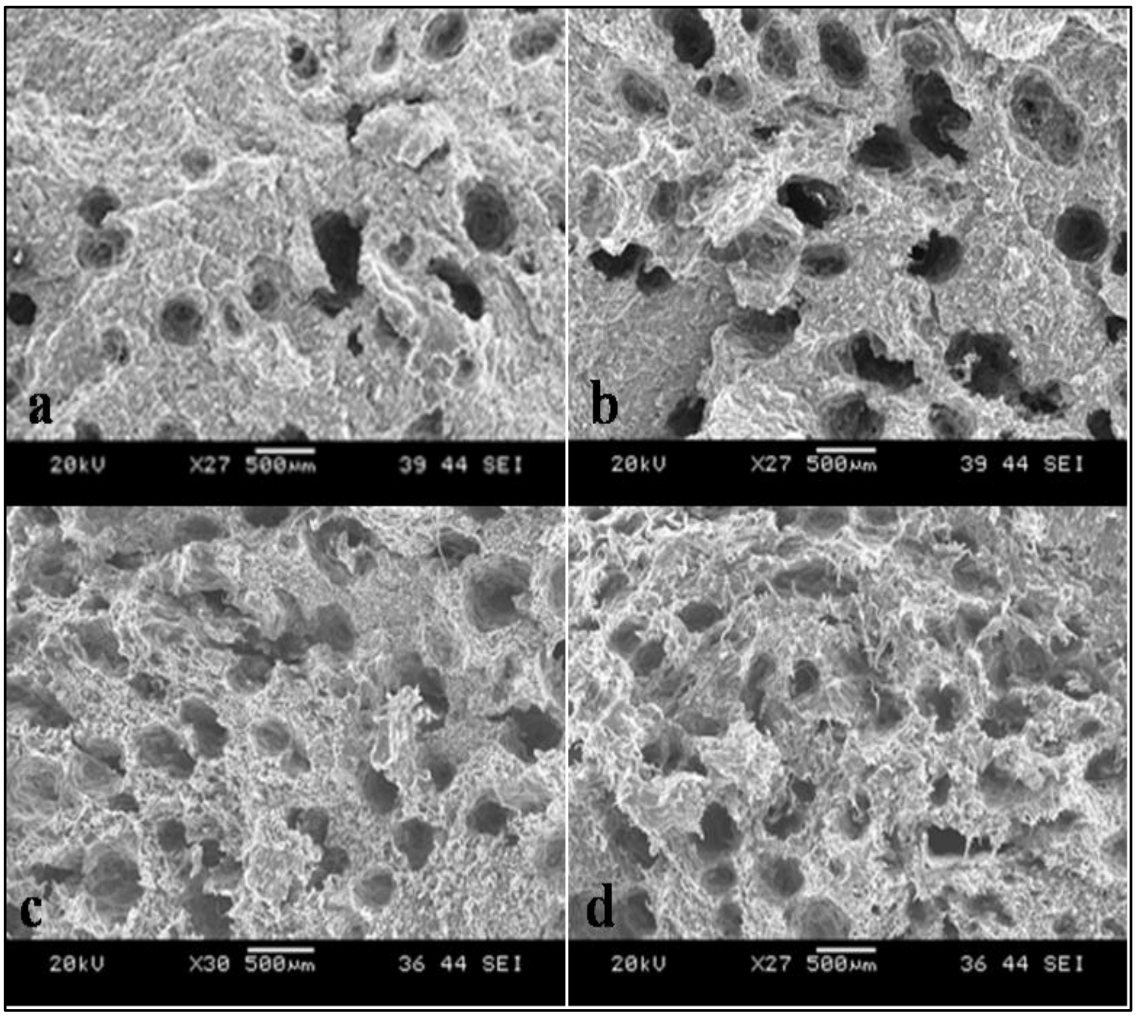


Figure 4-17 *Representative SEM images of fractured foamed at DDR of (a) 1.0 (b) 1.1 (c) 1.2 and (d) 1.4 for 6.5 phr CBA content composites*

The higher cell density and uniform cell distribution was observed at optimal DDR (Fig. 15) due to better melt properties, which ultimately increased the flexural properties of the foamed composites, the CBA contents and DDR employed. As CBA content increased and DDR increased, void fraction also increased (Figure 4-18). However, a decrease in void fraction was noticed as the CBA content increased beyond 3.5 phr and 1.2 DDR

For a given quantity of CBA, cell density has strong relationship with the average cell sizes since, the larger the cell size the fewer they are. The cell density of the foamed composites varied with the CBA contents and DDR used (Fig. 4-19). In general, the cell density was increased with the increase in CBA contents for all foamed composites which again depended on the DDR. For example, cell density of 6.5 phr CBA content composite increased from 2.1×10^3 to 1.6×10^4 cells/cm^3 when DDR was increased from 1.0 to 1.2 beyond which cell density decreased gradually. This shows that the cell densities of foamed composites have optimal values at some DDR value

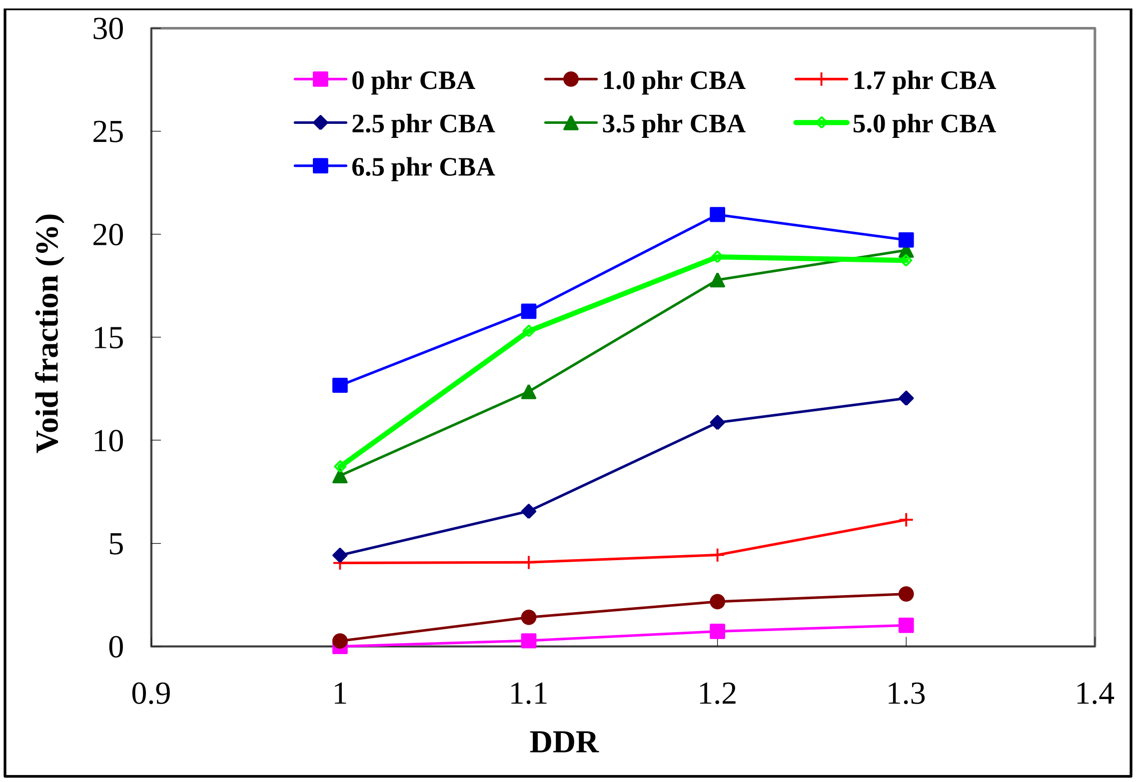


Figure 4-18 *Effect of DDR on void fraction*

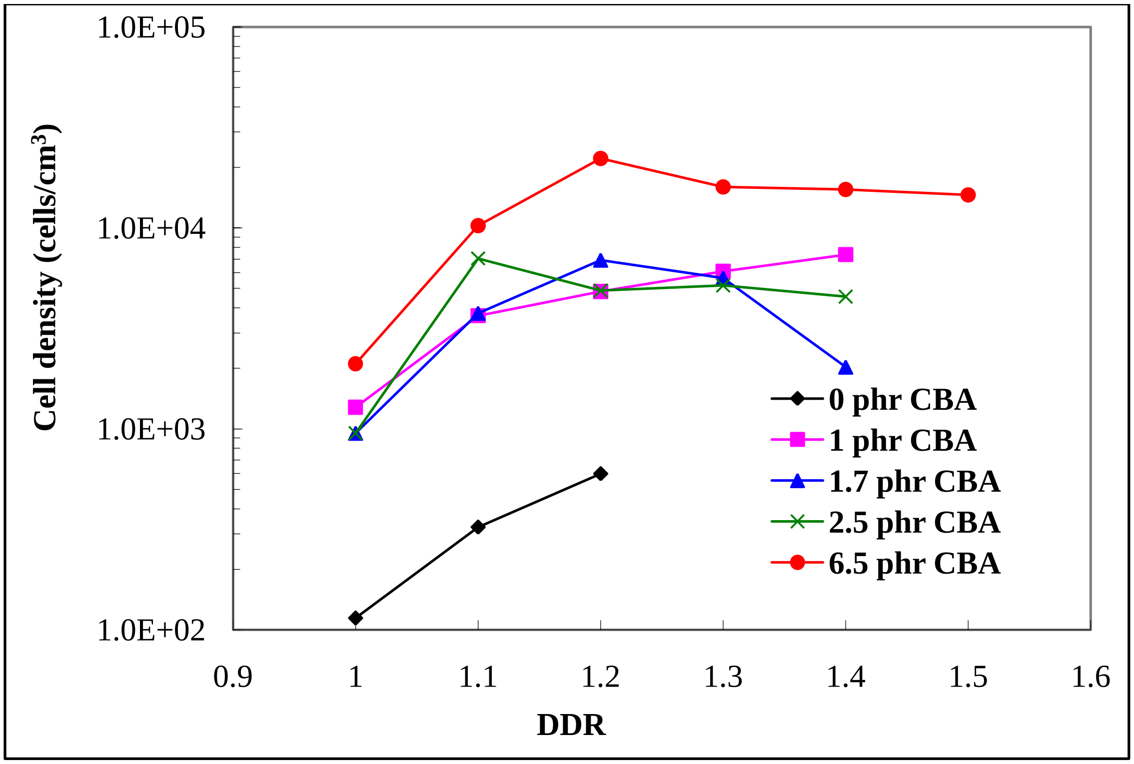


Figure 4-19 *Effect of DDR on cell density*

depending on CBA contents. The reduction in cell density at higher DDR could be due to formation of larger cells because of the coalescence/or collapse of already formed

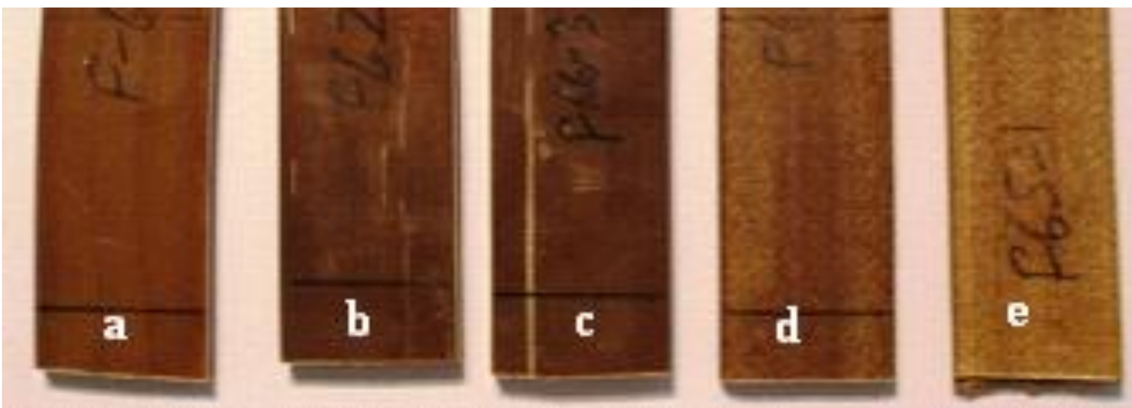


Figure 4-20 *Optical photographs of foamed WPC profile at (a) 1.0 (b) 1.1 (c) 1.2 (d) 1.3 and (e) 1.4 DDR, with 6.4 phr CBA content.*

bubbles [60]. However, surface quality did not alter noticeably at DDR up to 1.2 (20%) [Figure 4-20]. Further increase in DDR beyond 20 %, the dimension of profile gradually reduced and melt fracture occurred at 50 %.

4-1-4 -3 Optimizing WF Content in Foaming

Experiments were carried out to study the effect of variation in WF-1 content on density and flexural properties of WPC with varying CBA contents but with constant amount of lubricant and coupling agent. Palletizing and profile extrusion were carried out as mentioned before except DDR were kept as 1.2 and Die temperature was kept at 160 ºC. Process condition was same as Table 7.

As the CBA content increased density decreased. The lowest density was observed with CBA content 6.5 phr. No specific trend was found in alteration of flexural strength of WPC with varying CBA content and WF content. However, an overall increase in speific flexural modulus was found as WF content increased which could be explained by the reinforcemnet effects of the WF filler . But again, no trend was observed with change in CBA content.

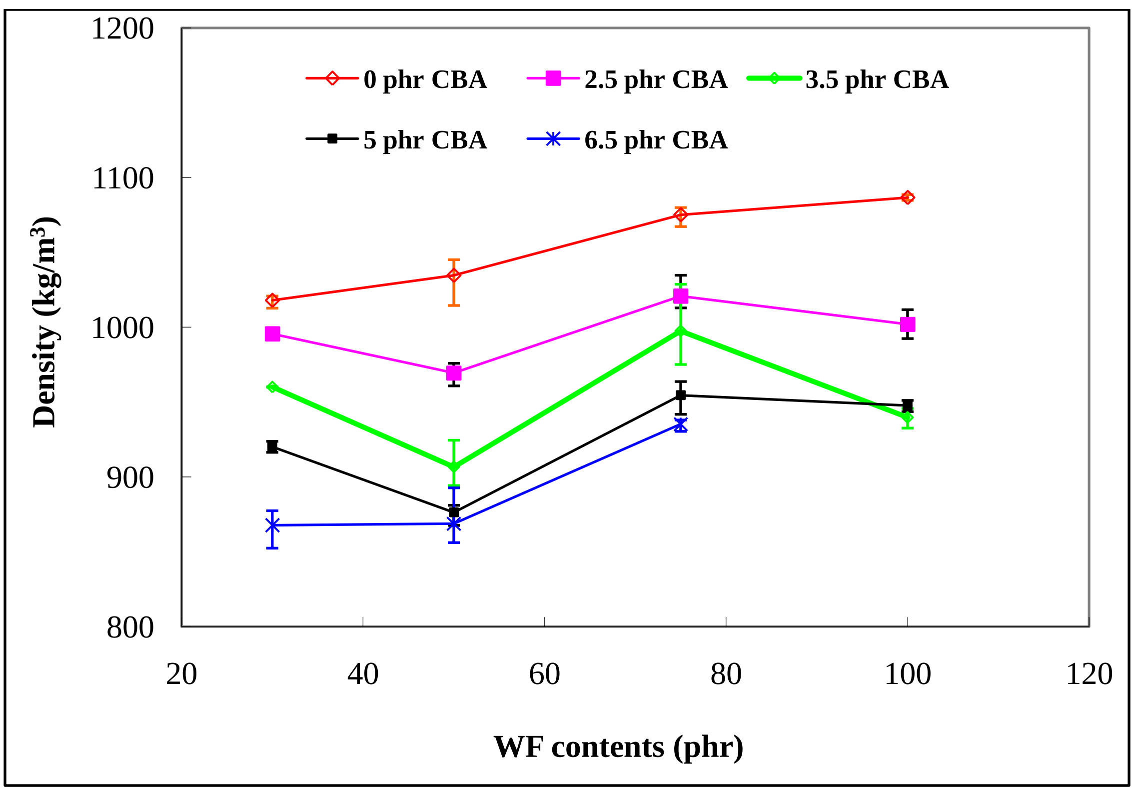


Figure 4-21 *Relationship between WF, CBA and density*

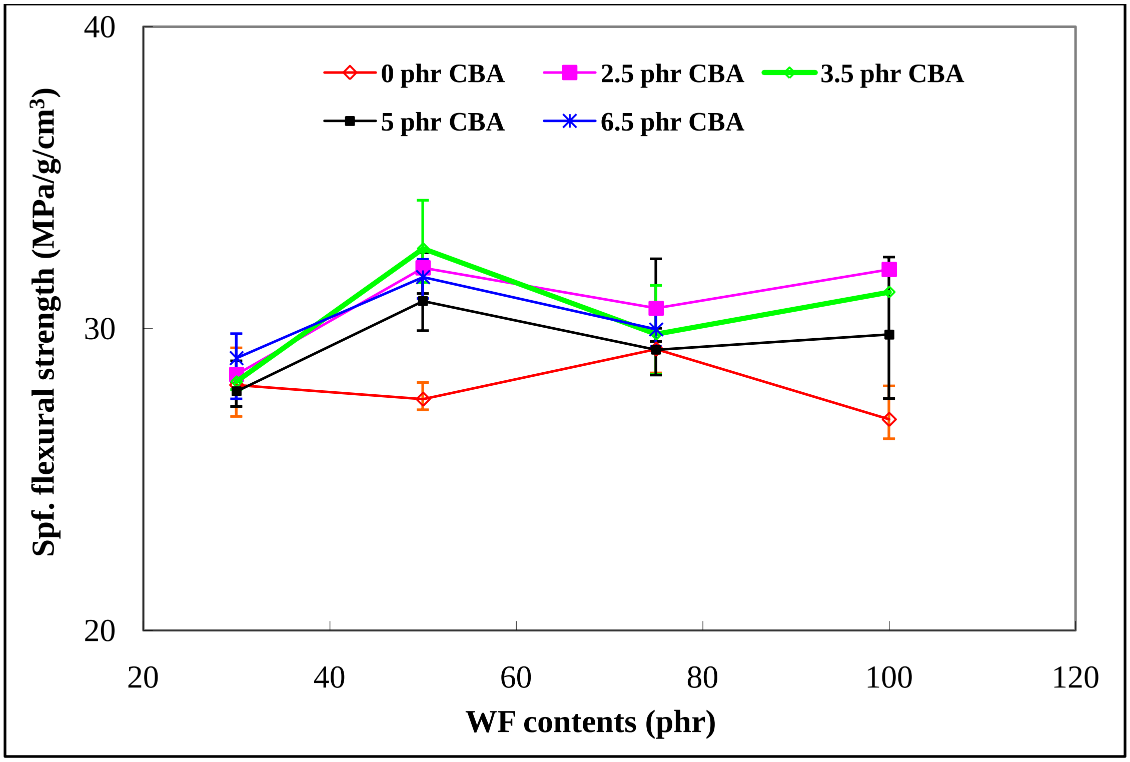


Figure 4-22 *Relationship between WF, CBA content and specific flexural strength of WPC*

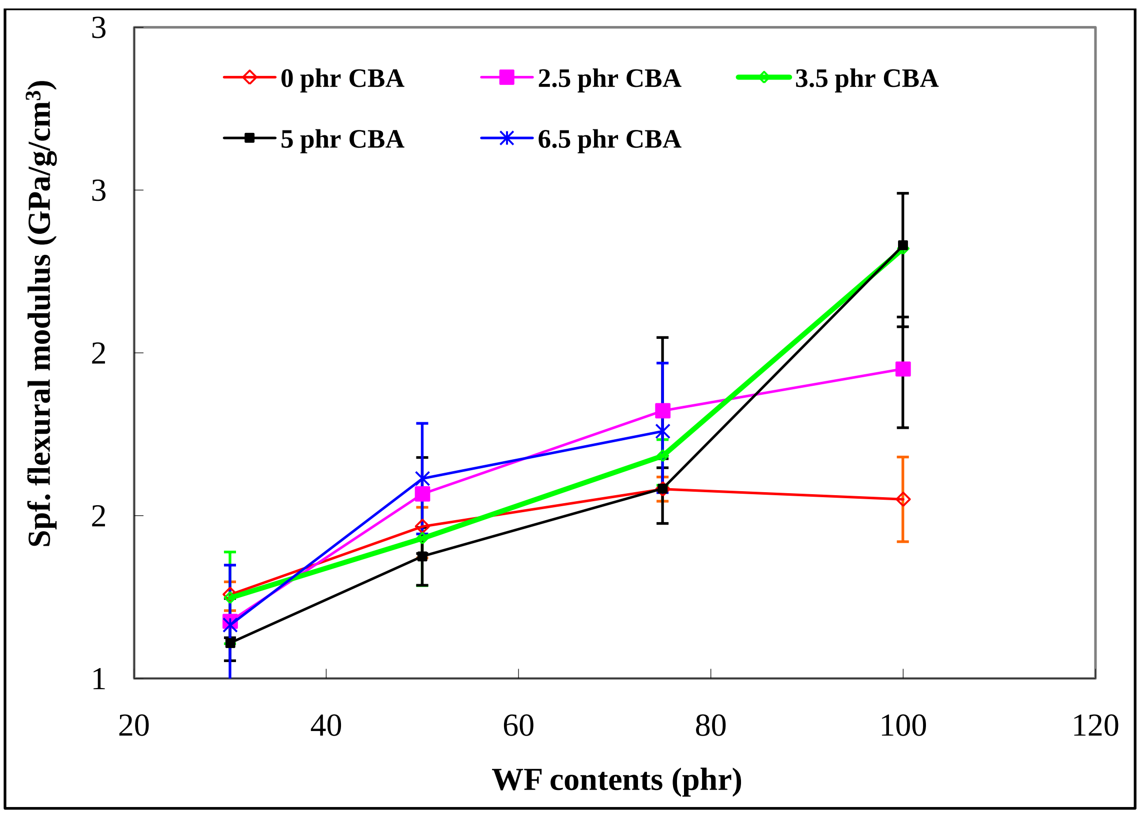


Figure 4-23 *Relationship between WF, CBA content andspecific flexural modulus of WPC*

4-2 Conventional Vs New Custom Built Compression Molding

WPC samples were prepared through conventional hot press and new custom built die-press with HDPE-1(74.5 wt%), WF-1(20 wt%), GF-1(2.5 wt%) and CA-2 (3 wt%). Process conditions were:

Die Temperature	: 160 ºC
Die Closing Speed	: 40mm/sec
Pressure	: 4.5 metric tons
Time	: 1 minute

The fiber alignment directions of the samples were investigated using a SEM (Figure 26(a) 26(b) 26(c)). Significant changes were noticed in the direction of GF fiber

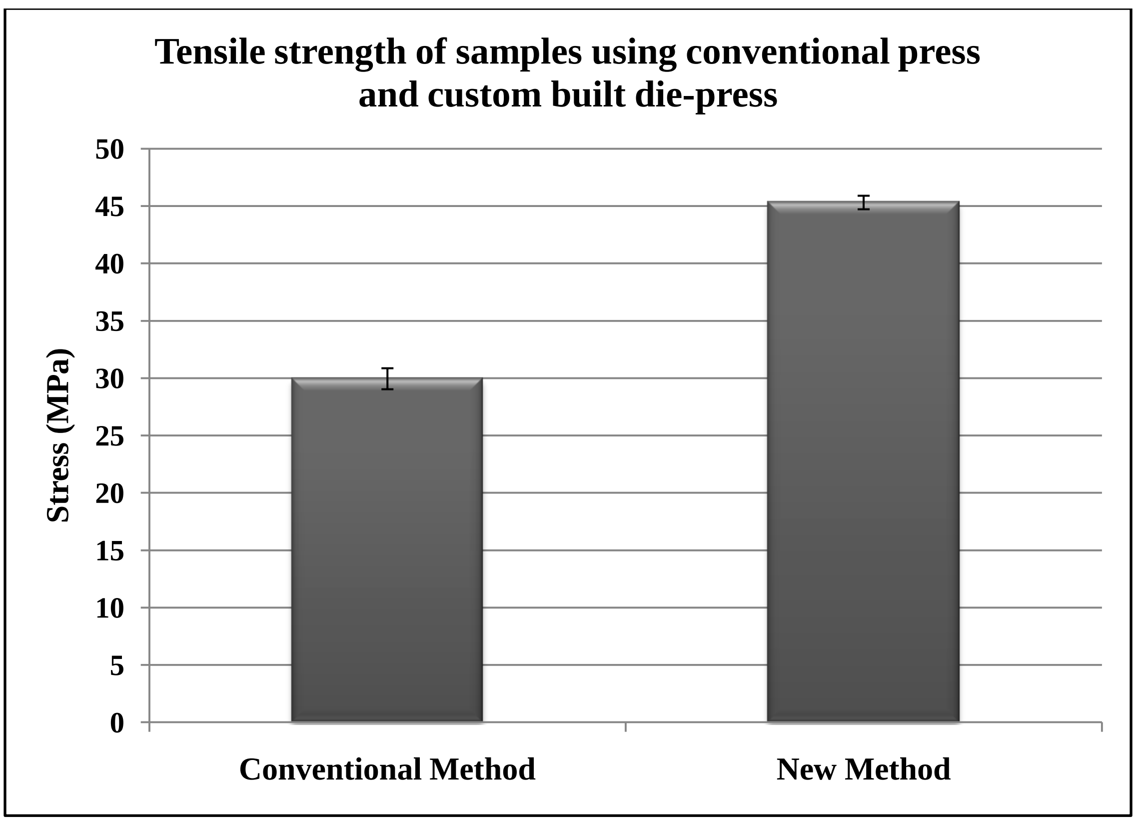


Figure 4-24 *Flexural strength of samples made by conventional press and custom-built die-press*

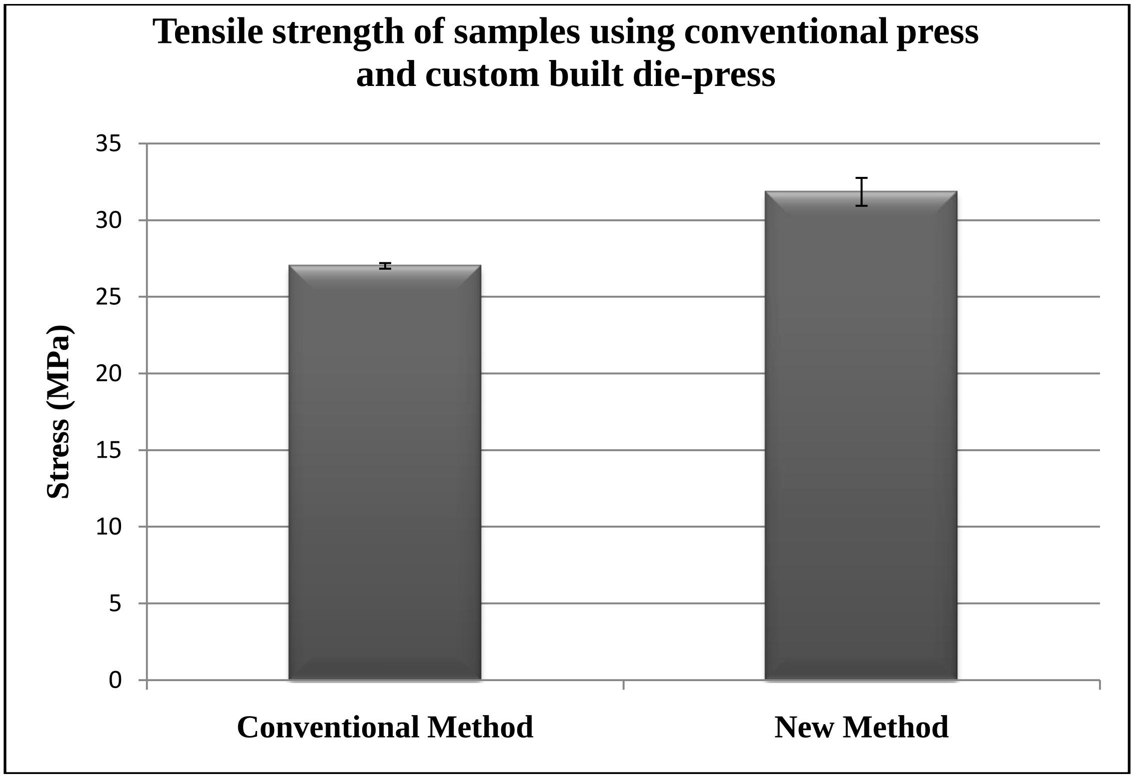
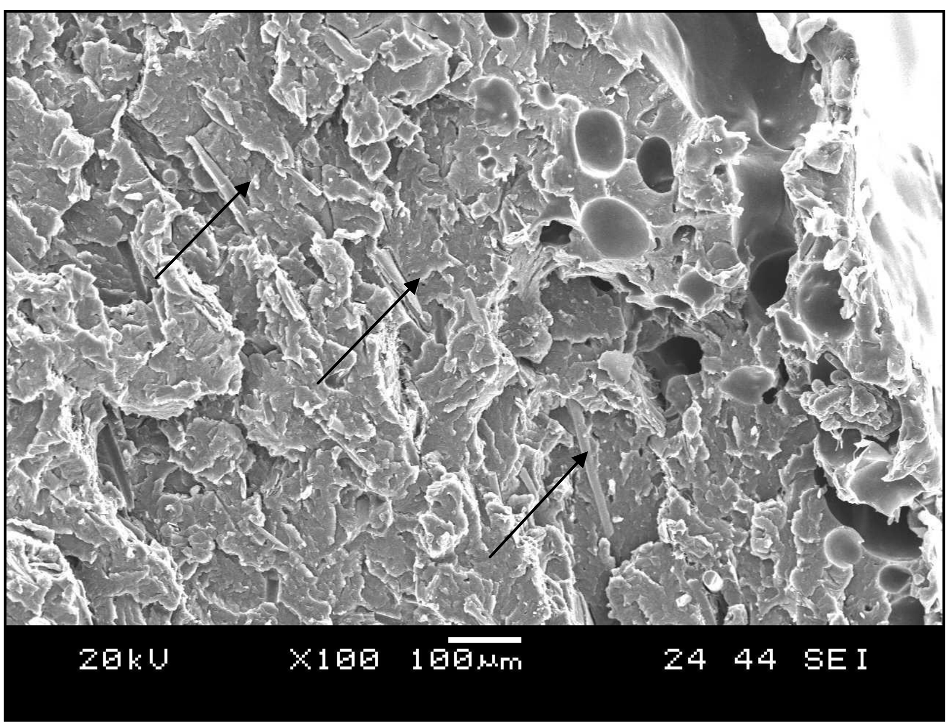
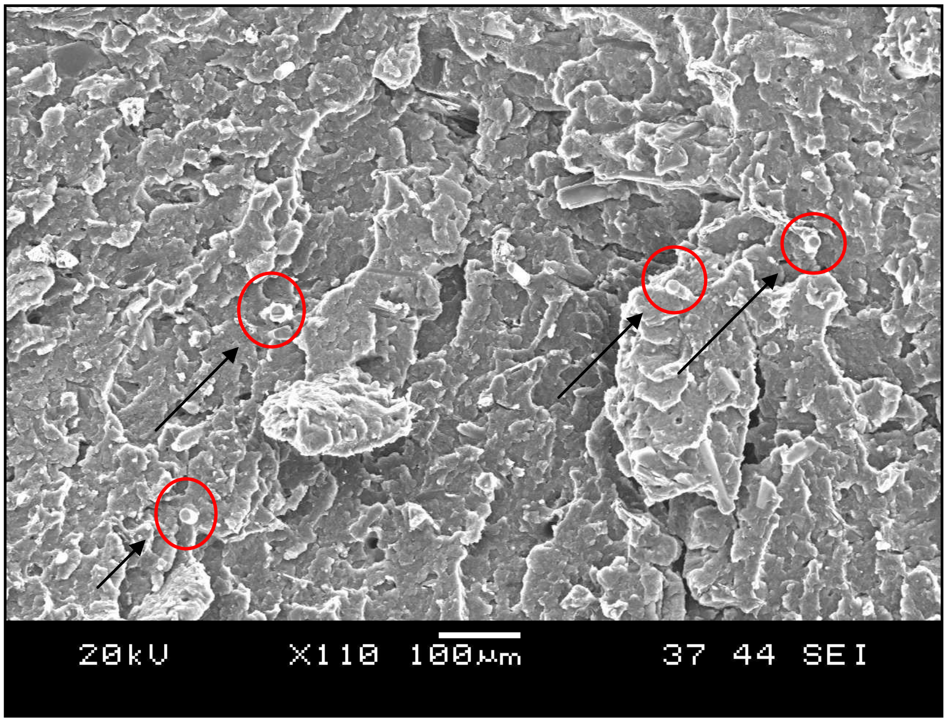


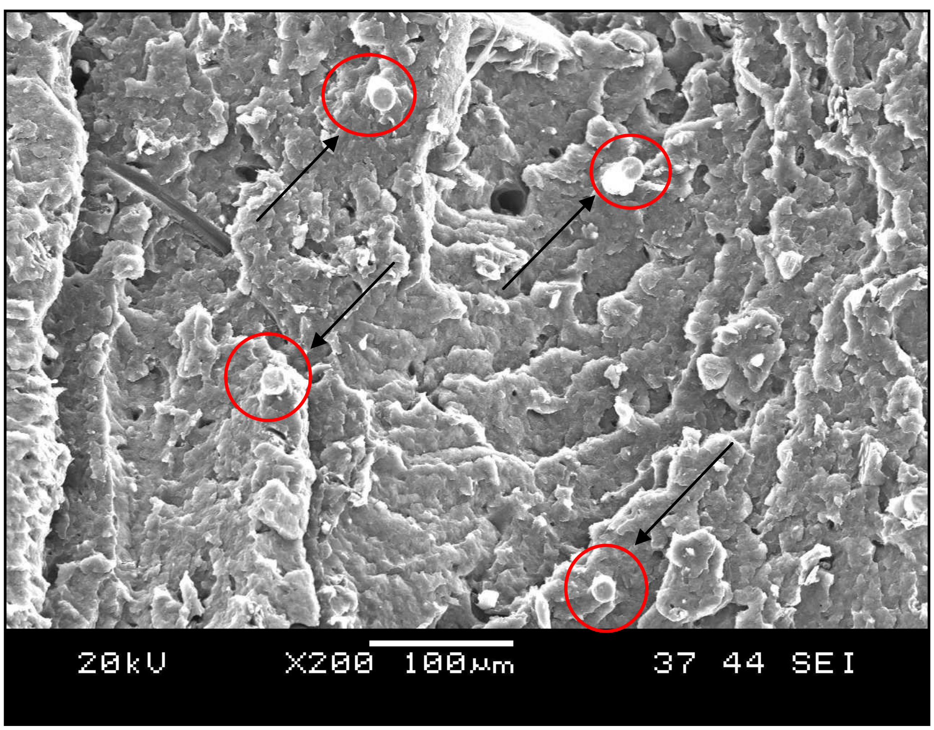
Figure 4-25 *Tensile strength of samples prepared by Conventional press*



(a)



(b)



(c)

Figure 4-26 *SEM of fiber alignment in the longitudinal direction both in conventional press (a) and new custom built press (b) & (c)* alignment. The new system made the fiber more aligned towards the longitudinal direction of the specimen as shown by the arrows. Again both the tensile and flexural strengths were noticed to be increased among the samples prepared by new custom build compression molding system, comparing to the conventional mold.

Using the new Die-press system flexural strength increased from 30 MPa to 45 MPa which is approximately 51% increase (Figure 4-24) and tensile strength increased from 27 MPa to 32 MPa, approximately 19% increase (Figure 4-25). These improvements are most likely due to better alignment of fiber throughout the specimen in the longitudinal direction, which was caused by flow of material in the channels during the compression process in the mold. However, the increase in tensile strength was not as large as it was in case of flexural strength. This could be due to edge effects during sample cutting, causing loss and/or pull out of fiber from the matrix structure that subsequently reduced tensile strength. Also it could be due to presence of bubbles in the of the dog-bone span (2.5mm X 3.2mm) for tensile test, which is very narrow compared to the cross-section of the sample (12.7mm X 3.2mm) for flexural test. Even presence of smaller bubbles in the narrow cross section could have caused bigger effect in reducing tensile strength.

4-3 Optimizing Process Parameters for the New Compression Molding

Two process parameters were studied; die temperature and die closing speed.

4-3-1 Optimizing Die Temperature

Samples were prepared with eight different die temperatures HDPE-1, WF-1, CA-2 and GF-1.

Process Conditions:

Die Closing Speed : 40mm/sec

Pressure : 4.5 metric tons

Time : 1 minute

Table 4-4 *Variation in Die temperature*

Sample	**T1**	**T2**	**T3**	**T4**	**T5**	**T6**	**T7**	**T8**
Temp °C	**80**	**100**	**107**	**112**	**117**	**122**	**127**	**132**

Highest flexural strength was observed among samples prepared at 117 °C which is 54 MPa (Figure 4-27). With gradual increase in temperature from 80 °C, the flexural strength increased gradually. But after 117°C, strength decreases as the temperature increases.

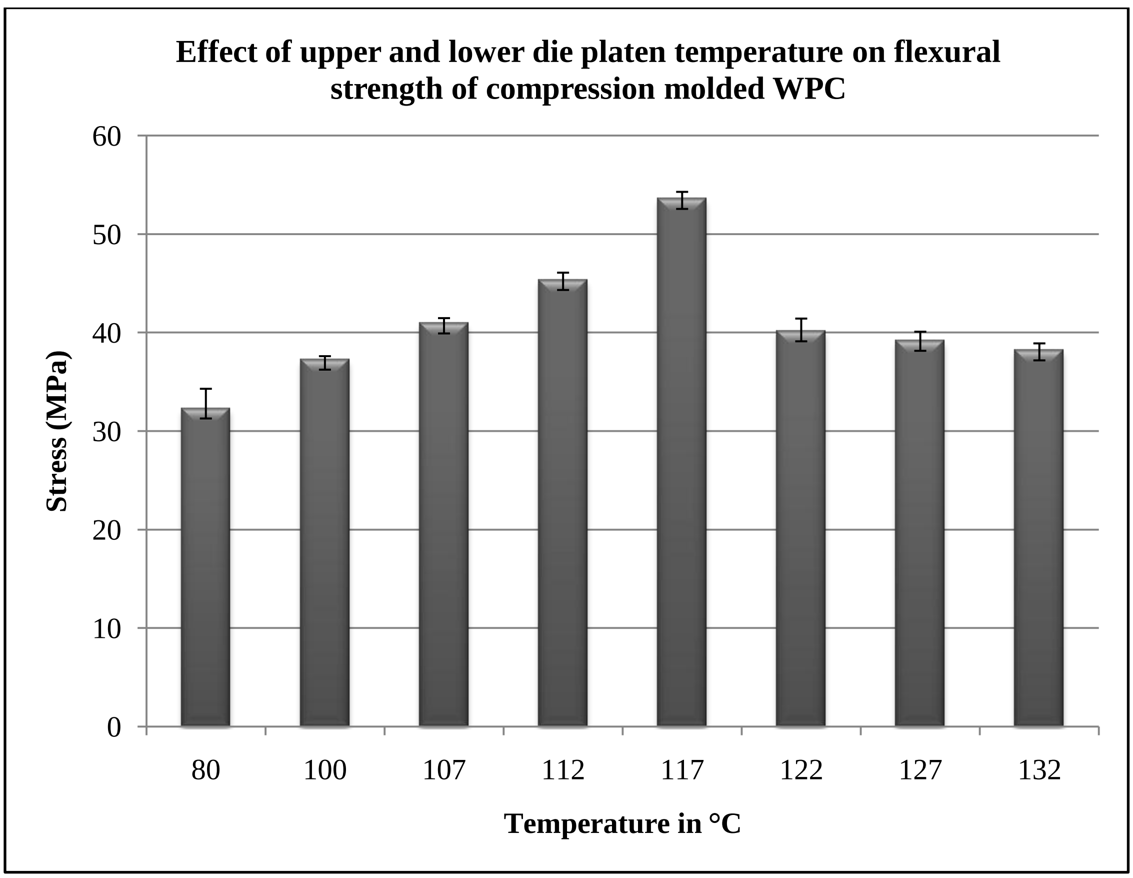


Figure 4-27 *– Flexural strength varying die platen temperature*

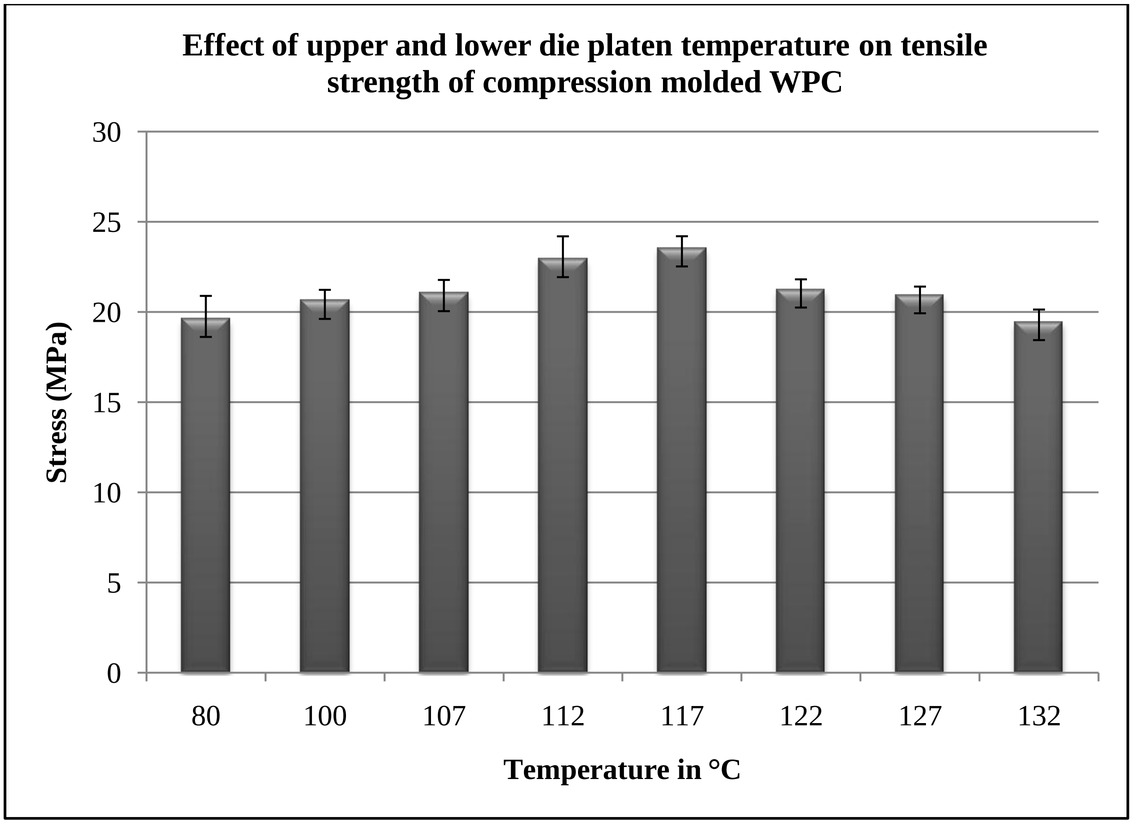
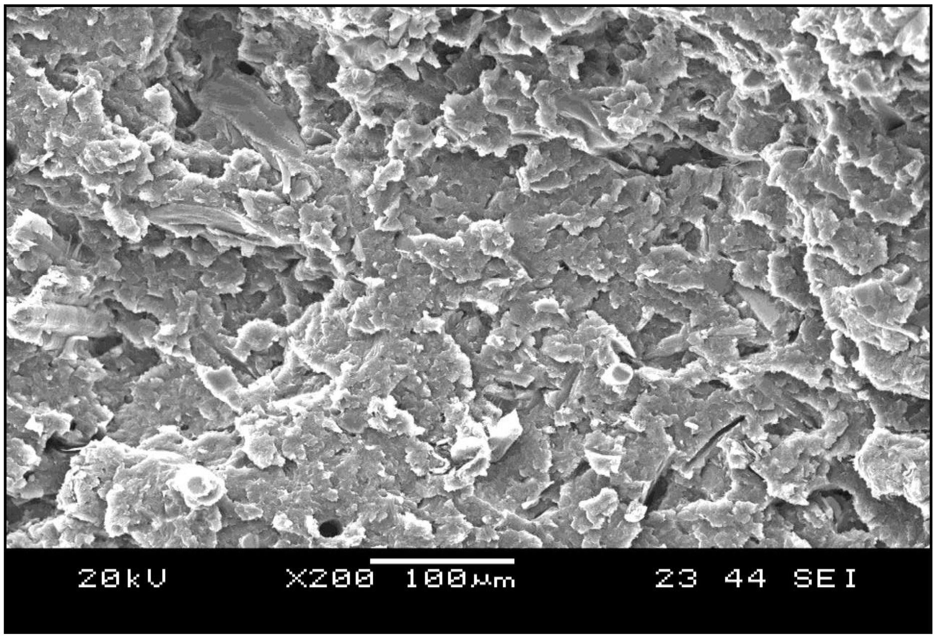
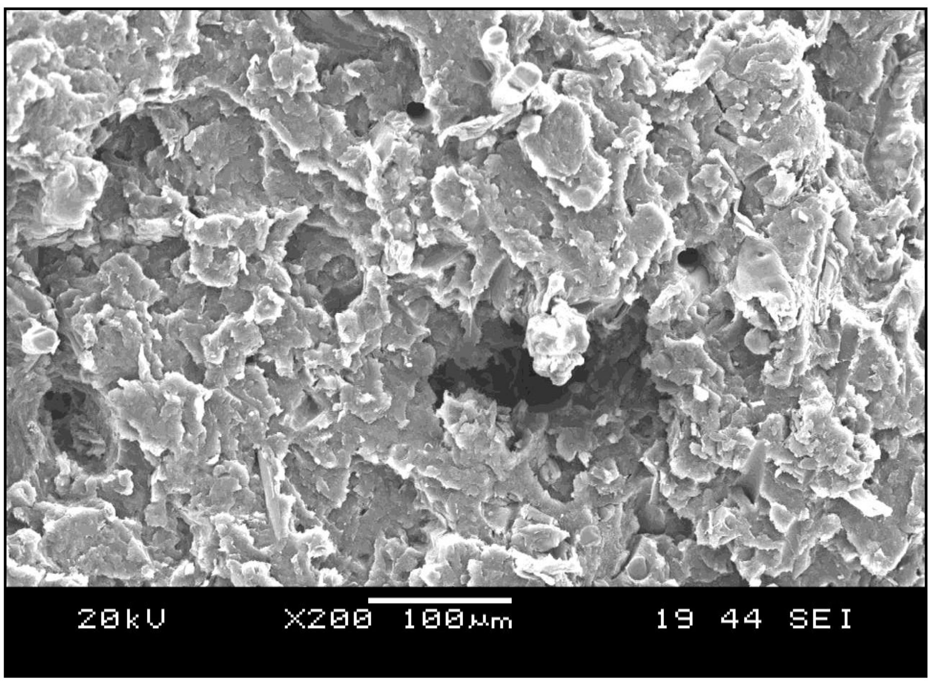


Figure 4-28- *Tensile strength varying die platen temperature*

The same pattern was exhibited in case of tensile strength. With gradual increase in temperature, strength increased, which was noticed to be highest at 117 °C, valued at 24 MPa (appx). But at beyond 117 °C, tensile strength dropped. Possible reason could be, at lower die temperatures the polymeric material does not get properly fused or welded during the compression process. Therefore voids and weld lines remain which make the structure weak. But at 117 °C the materials must have properly fused into the new shape. In addition, bubble generation is found to be minimal at 117 °C. Therefore, at temperatures beyond 117°C, due to volatilization from wood fibre, bubbles are generated and remain trapped in the structure leading to void formation(Figure 4-29(a) and 4-29(b)) and subsequently to lowering of strength.



(a)



(b)

Figure 4-29 *SEM images of X-section of samples prepared by varying die platen temperature (a)117°C (b) 80°C*

4-3-2 Optimizing Die Closing Speed

3 different die closing speeds were studied.

Process conditions:

Die temperature: 117 ºC

Pressure : 4.5 metric tons

Time : 1 minute

Table 4-5 *Variation in die closing speed*

Sample	S1	S2	S3
Speed(mm/sec)	10	25	40

Maximum flexural strength was observed at closing speed 40mm/sec which was 23% (appx) higher than samples prepared at 10 mm/sec. Very slight increase (1.7 %

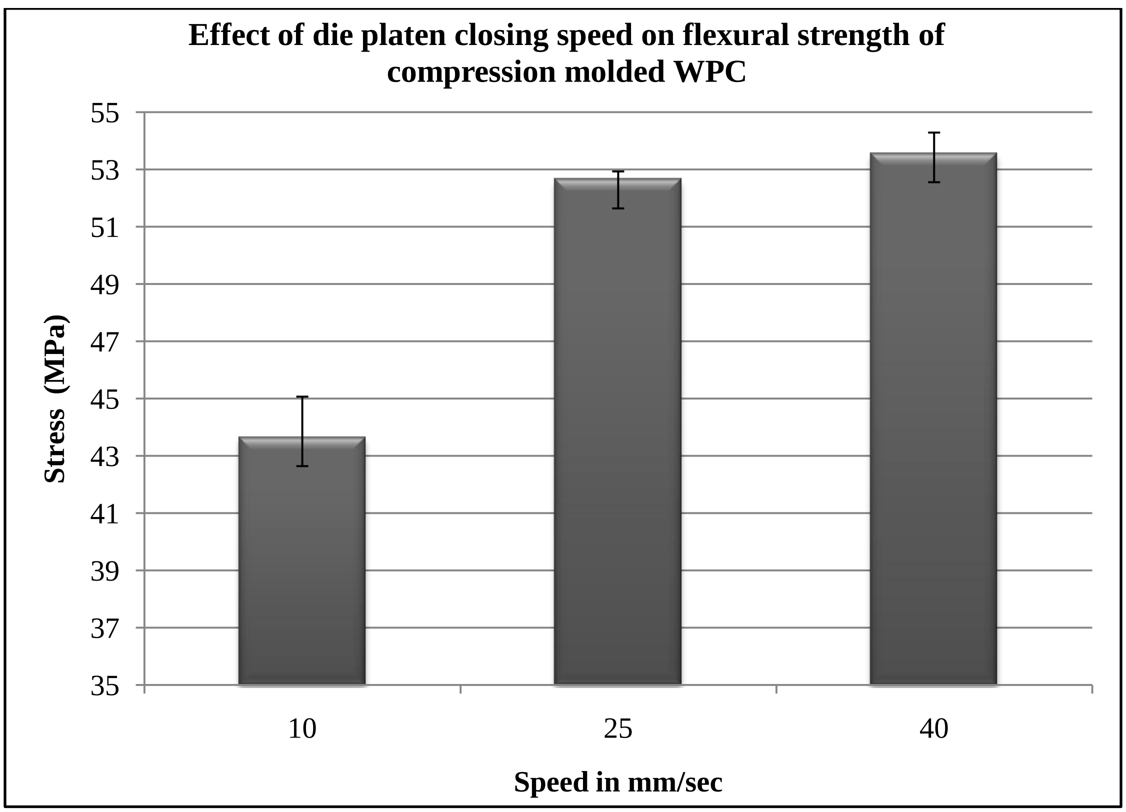


Figure 4-30 *Flexural strength of samples varying die platen closing speed*

appx) of flexural strength was sighted at 40mm/sec closing speed compared to the strength at 25mm/sec. Same trend was seen in case of tensile strength. Samples prepared at 40 mm/sec showed highest strength, which was 7.1% higher than the samples prepared at 10mm/sec.

Increase in strength with increase in speed is most likely due to the effect of pressure, generated by higher die press speed. This pressure possibly compressed the bubbles and reduced cavity formation. Increased pressure can also possibly create better adhesion between fiber and polymer which increases strength.

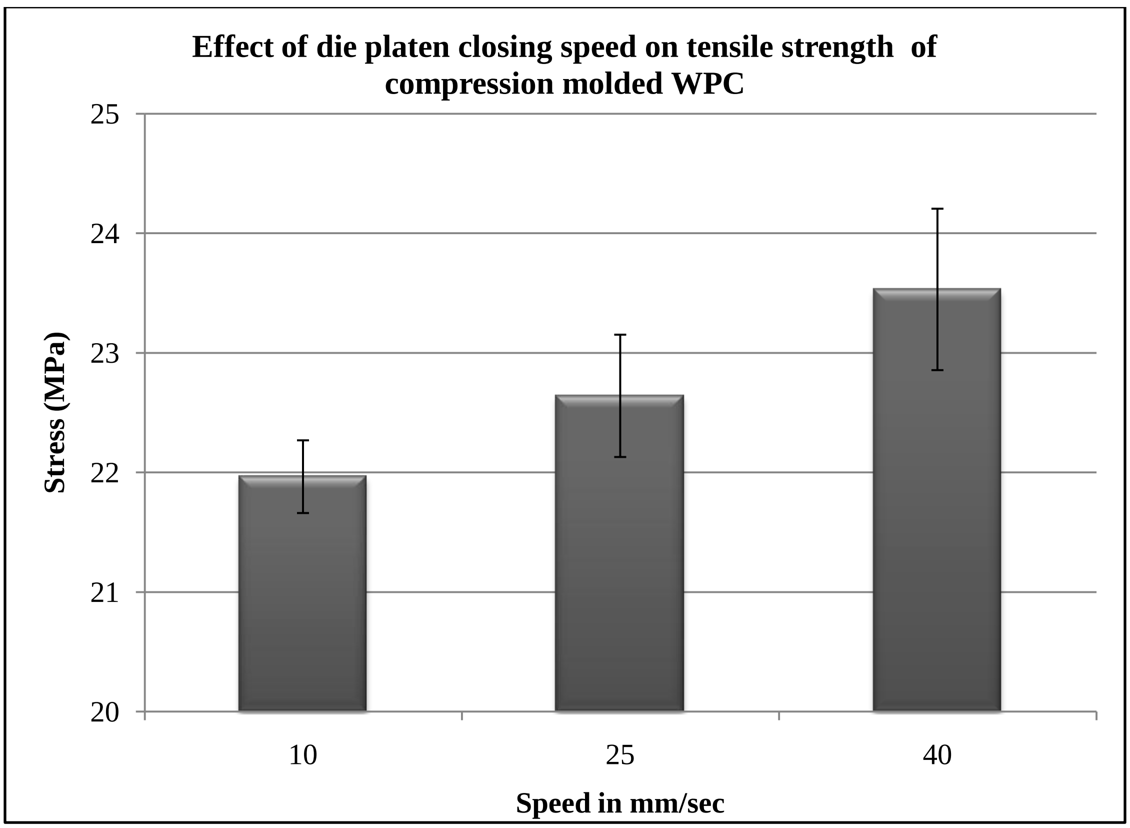


Figure 4-31 *– Tensile strength of samples varying die platen closing speed*

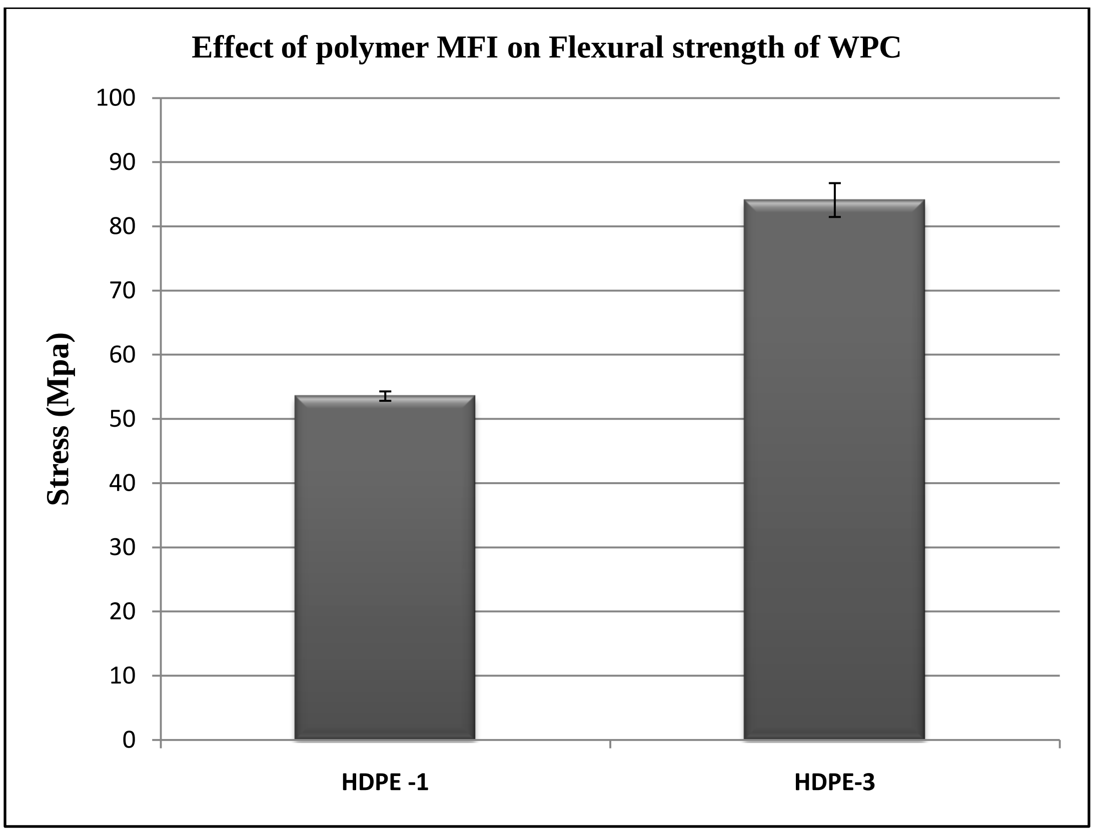
4-4 Effect of Variation in HDPE, WF and GF Grades

Effects of variation in grades of WF, GF and HDPE on mechanical strength of WPC were studied using the samples prepared by compression molding. Process conditions:

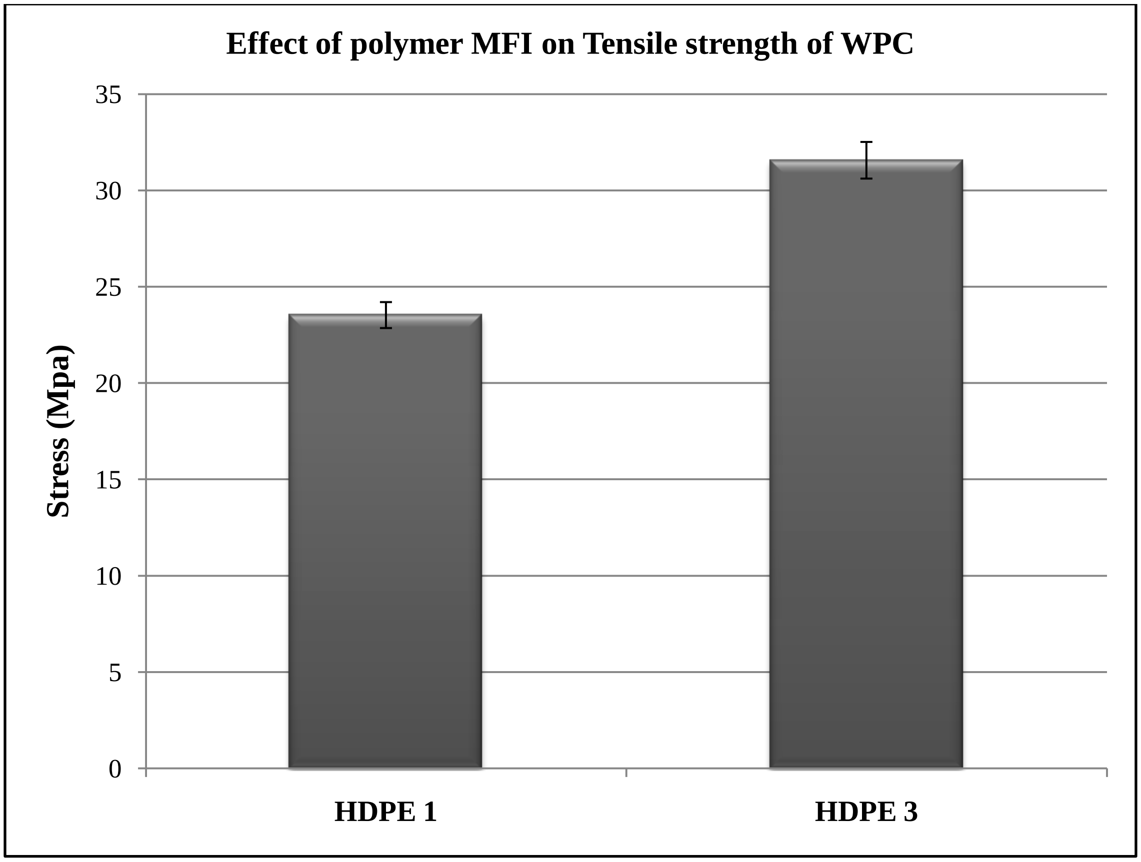
Die Temperature	: 132 ºC (HDPE-1) and 155 ºC (HDPE-3)
Die Closing Speed	: 40mm/sec
Pressure	: 4.5 metric tons
Time	: 1 minute

4-4-1 Effect of Variation in HDPE Grades

HDPE-1 and HDPE-3 was used to prepare samples with same type of WF, GF and lubricating agent. Maximum flexural strength was observed in WPC with HDPE-3 which was about 57% higher than the WPC with HDPE-1. Tensile strength also increased by 35% when the lower MFI HDPE- 3 was used. This is most likely due to higher molecular weight and greater polymer chain entanglement in polymer with lower MFI that increases mechanical properties of WPC when there is a good distribution of filler [103-105].



(a)

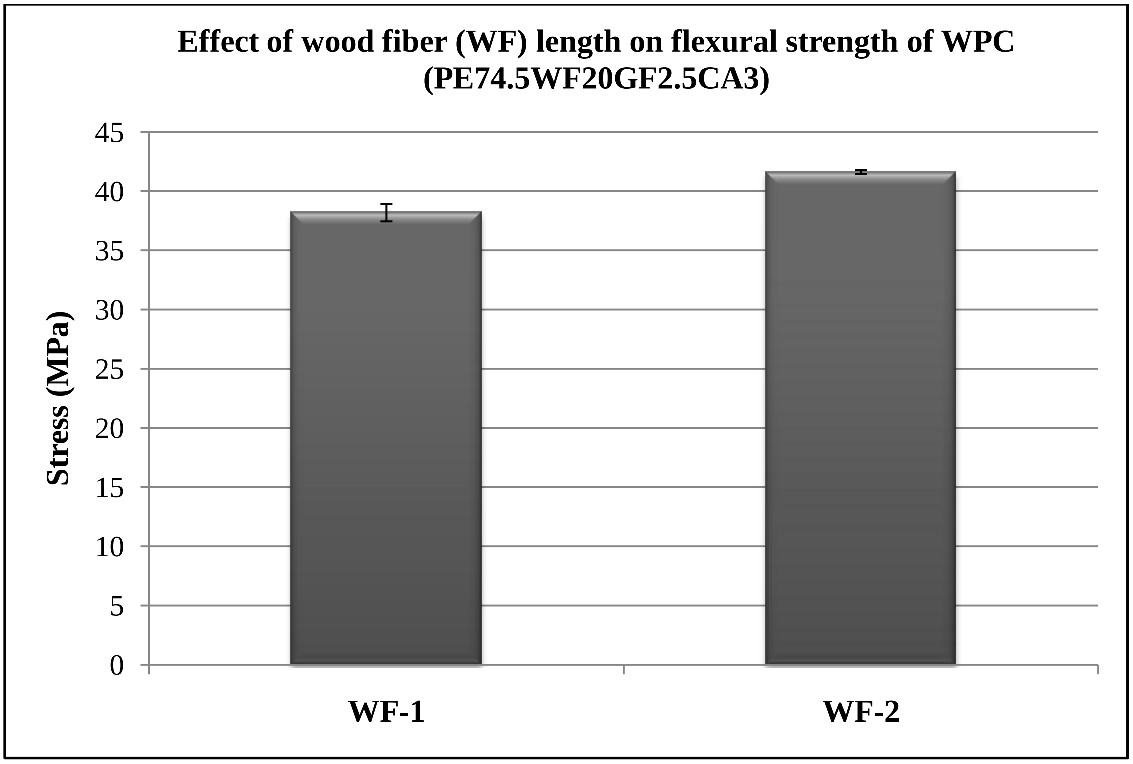


(b)

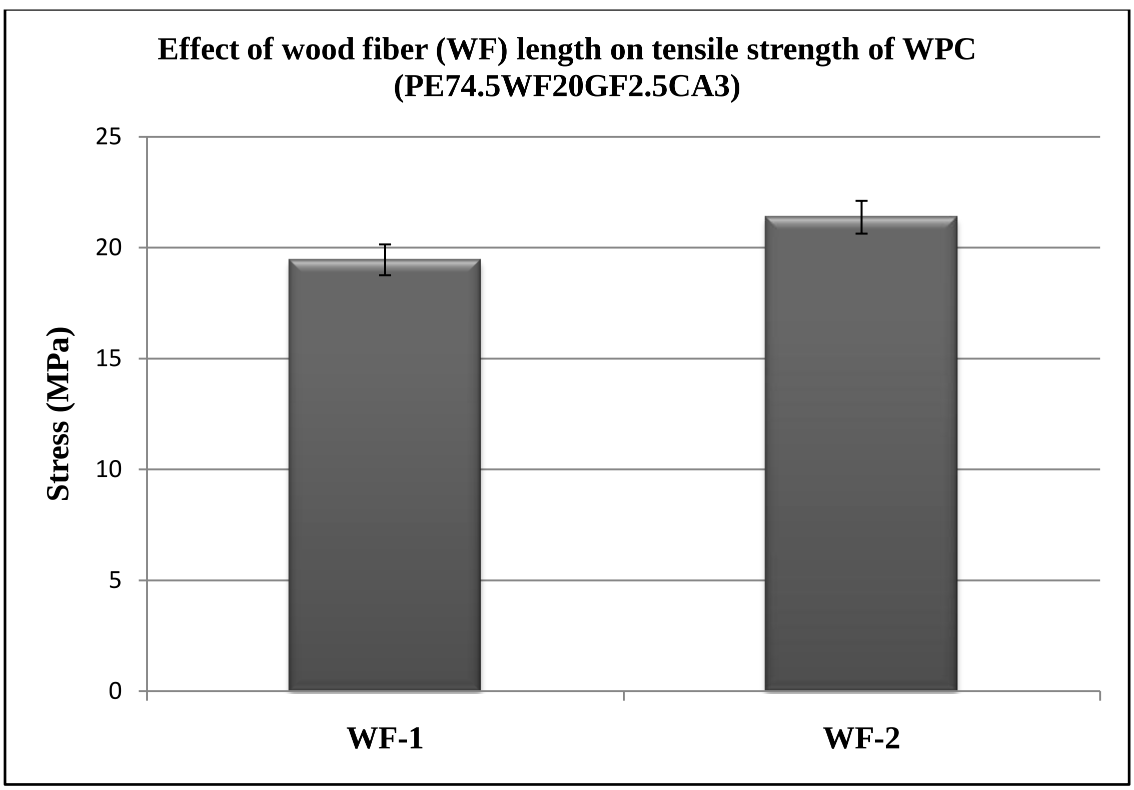
Figure 4-32 *Flexural (a) and Tensile (b) strength of WPC varying MFI of HDPE*

4-4-2 Effect of Variation in WF Length

Study was carried out on samples prepared with WF-1 and WF-2. WPC with longer WF showed higher flexural strength of 41.60 MPa; which was about 9% higher than WPC with shorter WF (38.20 MPa).Tensile strength increased by appx. 10% with WF 2 than WF 1.



(a)

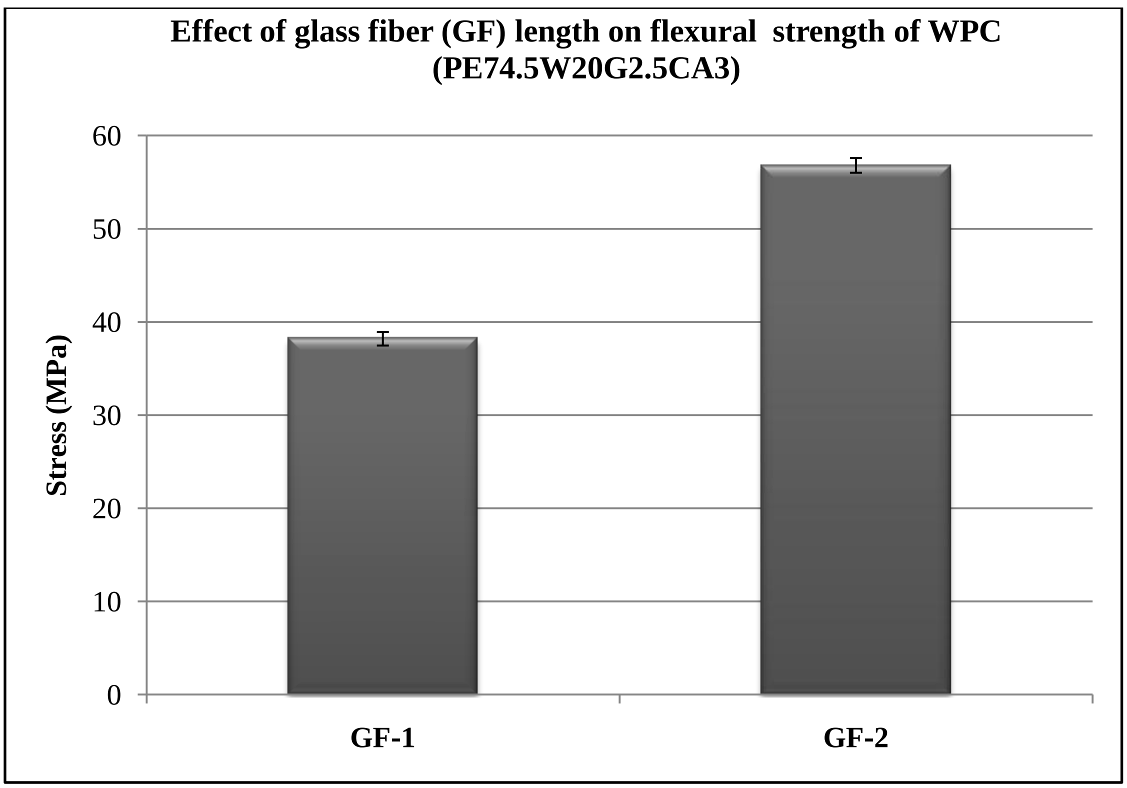


(b)

Figure 4-33 *Flexural (a) and Tensile (b) strength of WPC varying WF length*

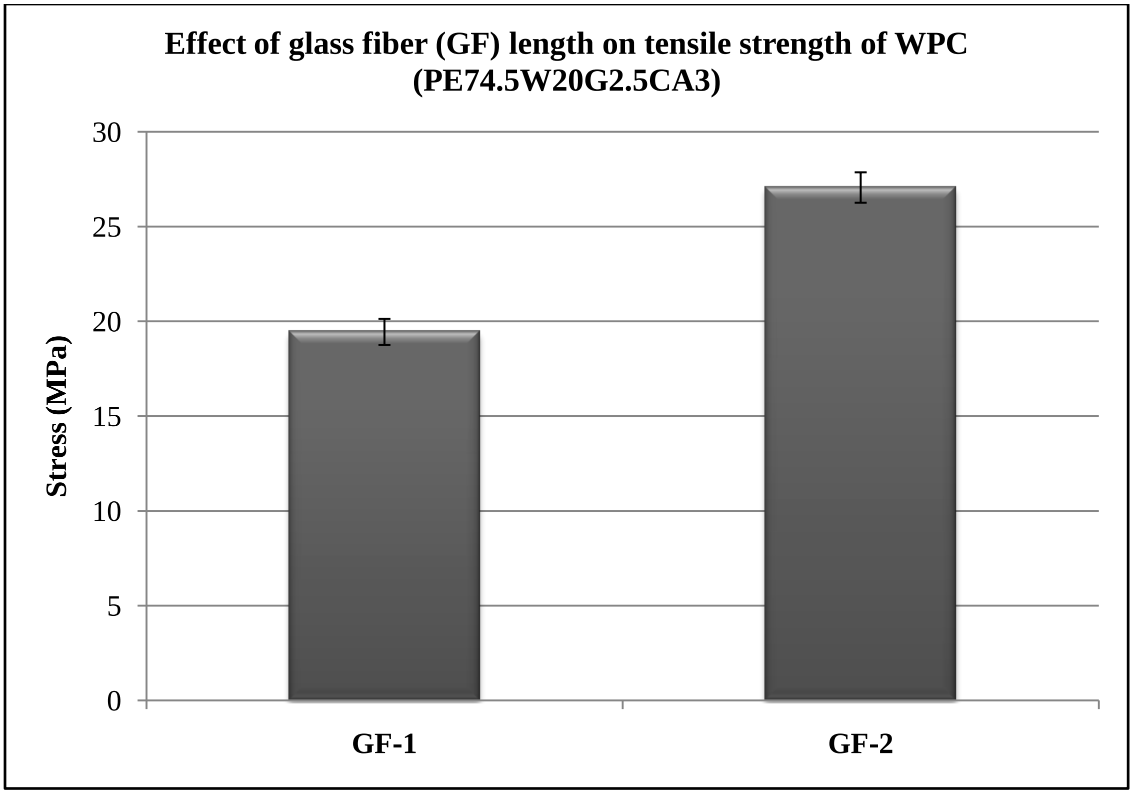
4-4-3 Effect of Variation in GF Length

Effects of two different GFs on mechanical properties of WPC were studied. An increase in 49% of the flexural strength was recorded in WPC with GF-2 than WPC with GF-1. Improvement in tensile strength was 40% with longer GF2. Figure 54(a, b) shows the SEM pictures and it can be seen that the longer glass fibers survive the processing as the GF strands are clearly visible.Increase in strength was also observed in WPC with longer WF probably because of better adhesion between wood fibres and the matrix polymer, uniform dispersion of WF in the polymer matrix and better orientation of the WF [106, 107]. Again, it is already established that the custom build directional



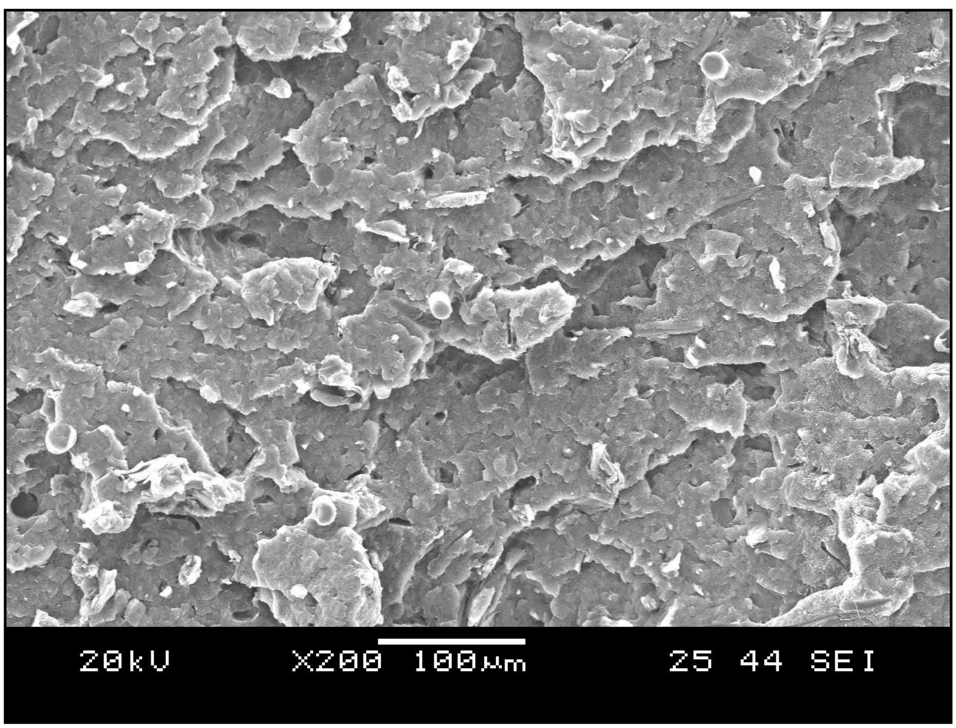
(a)

compression molding imparts better orientation of fibres in the longitudinal direction [108]. And longer fibres provide better reinforcing affect when they are better aligned.

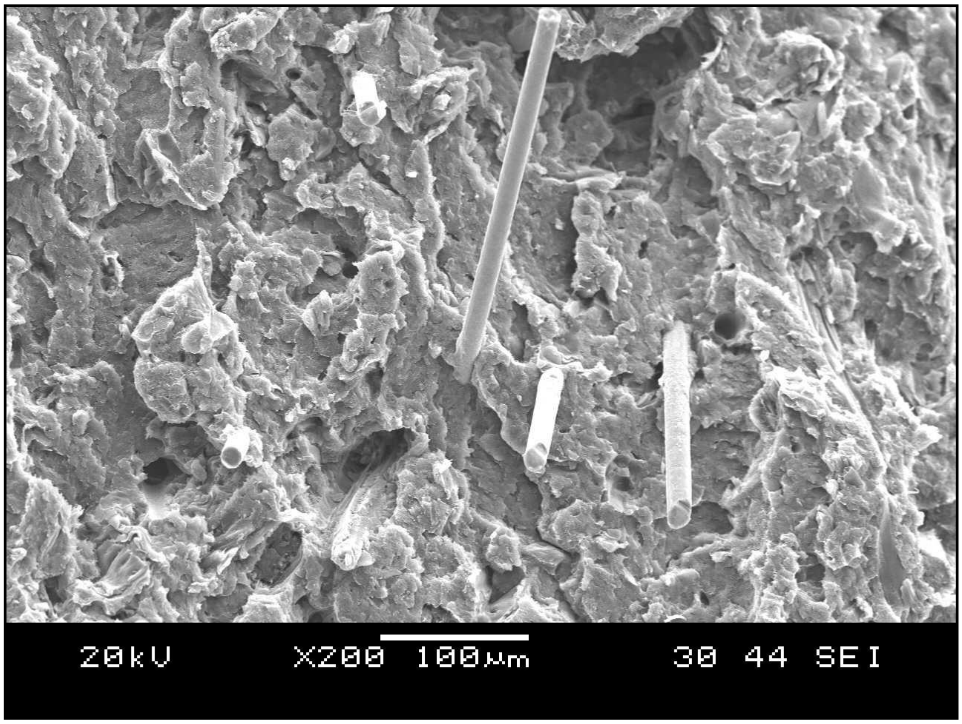


(b)

Figure 4-34 *Flexural(a) and Tensile(b) strength of WPC varying GF length*



(a)



(b)

Figure 4-35 *SEM images of X-section of samples prepared varying GF length (a) 0.8mm (1/32”) (b) 4mm*

4-5 Custom Built Compression Molding Vs Profile Extrusion

Study was carried out to compare mechanical properties of WPC, achieved by profile extrusion and custom built compression molding. WF-1 varied between 20 wt% to 60 wt%. Lubricant and CA-1 contents were kept constant and the balance was HDPE-3.

Process conditions:

Die temperature	: 117 ºC
Die Closing Speed	: 40mm/sec
Pressure	: 4.5 metric tons
Time	: 1 minute

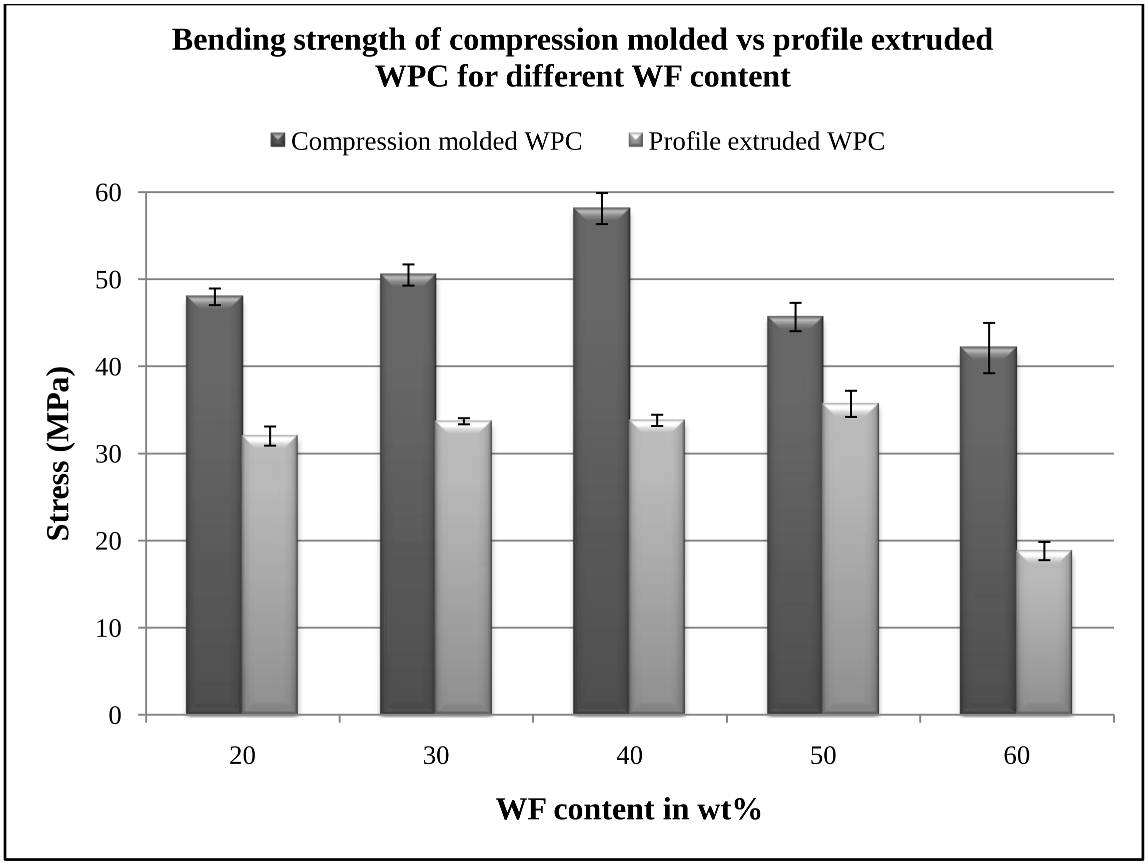


Figure 4-36 *Bending Strength of Compression Molded vs Profile Extruded WPC with Different WF Content*

For all compositions, flexural strength was found to be very high among samples prepared by compression mold, comparing to samples prepared by profile extrusion. For composition with 40% WF, the flexural strength of compression molded samples was found to be about 72% higher than profile extruded samples. However, among compression molded samples, composition with 40% WF showed the highest strength (58.10 MPa) where as among profile extruded samples, composition with 50% WF showed thehighest strength (35.70 MPa). Though, in case of profile extrusion the strength difference between WPC with 40% (33.80 MPa) WF and 50% WF is not so high (appx 5.60% increase) and with 50% WF the range of error bar is high comparing to WPC with 40% WF. This improvement among profile extruded samples could be due to better dispersion of WF in the polymer matrix because of high shear mixing in extruder. The reduction in flexural strength of 50% WF content could be due to insufficient adhesion between pellets when WF content is high.

Chapter 5 Summary and Recommendations

5-1 Summary

In this research a number of issues for improving the quality of WPC have been studied. The WPC profiles were produced from HDPE of varying MFI and WF, with different extrusion conditions. Varied material formulations were used in a lab scale twin-screw extruder. The MFI (melt viscosity) of polymer had a large influence on both tensile and flexural properties of the composites. Both tensile and flexural properties (strength and stiffness) were decreased as MFI increased. The tensile and flexural properties were increased with the increase in WF contents; however, rate of increment depended on polymer MFI. High WF content increased the melt viscosity of the composite and decreased bonding between WF and polymer due to poor wetting or lack of sufficient CA, which decreased the tensile and flexural strength. Both tensile and flexural properties (strength and modulus) of WPCs increased with the increasing WF contents up to 50 wt. % but decreased when the contents were increased to 60 wt%. Tensile strength was decreased but flexural strength increased with increasing lubricant contents up to 7 wt. %. Both properties decreased at lubricant content 10 wt. %. For practical application, a combination of proper HDPE grade, lubricant and MAPE can improve the mechanical properties and surface smoothness of the profile. The optimal tensile and flexural properties were observed with the 50 wt. % WF at 7 and 3 wt. % lubricant and MAPE contents.

WPC foamed profiles were also produced with different CBA contents and varying extrusion conditions (DDR and die temperatures). The effects of endothermic CBA content, WF content, Die temperature and DDR on WPC processing, on foam cell morphology and on flexural properties were investigated. The extruded dimensions, surface texture and color did not change noticeably up to 1.2 DDR, however was deteriorated significantly for DDR greater than 1.2. Optimized die temperature was found to be 160 ºC and DDR values 1.1-1.2 depending on the CBA contents. Strength value also varied depending on WF content.

Samples made by custom built die-press compression mold were evaluated and compared with samples prepared by conventional hot press and profile extrusion. The custom build die-press seemed to produce longitudinal fiber alignment and subsequently had positive effects on flexural and tensile strengths. Study was also carried out to optimize process parameters (Speed and platen temperature) during compression molding. Fibers in custom built compression molded samples were found to be better aligned than in samples prepared by conventional hot press mold, in the longitudinal direction at all temperatures and at all speeds. Variation in the amount of bubbles was observed at different temperature and speed. Optimum closing speed was found to be 40mm/sec at 117 °C of die temperature. Study was also carried out to learn the effect of MFI of HDPE and fibers length during compression molding. Lower MFI of HDPE produced stronger materials. As the fiber length increased (both in case of WF and GF) both tensile and flexural strength increased. Flexural strength of the samples prepared by compression molding was found to be higher than samples prepared by profile extrusion.

5-2 Recommendations

Some of the future work directions could be:

i. Optimization of the parameters e.g. temperature, feed rate, screw speed, others and material composition to produce foamed WPCs with improved properties.
ii. Incorporating GF to further improve strength of foamed WPC.
iii. Improving foam morphology to get enhanced properties.

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