

# Advancing the Technology Development for Better Quality Wood Plastic Composites: Processability Study

by

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# **I. Certificate of Approval**

## II. Abstract

Wood Plastic Composites (WPC) have advantages over natural wood such as improved stiffness, recyclability, and waste minimization. However, issues such as the difficulty of processing WPC with conventional methods, volatile emission from the wood and the composites' lack of strength must be addressed. A system for continuous extrusion of rectangular profiles of WPC was developed and some critical processing strategies were identified. The use of a lubricant and a calibrator also improved the profile extrusion of WPC. In this work, glass was also added to improve WPC's mechanical strength. Generally, a glass content of 2.5% appears to improve the properties but further addition does not have a significant effect. Foaming of WPC, which can enhance their properties, was investigated through studying the effect of heating time and temperature on void fraction and cell density.

**Keywords:** composites, green materials, wood plastic composites, extrusion, biocomposites, coupling agent, sawdust, post consumer HDPE, profile extrusion, batch foaming, tensile strength, flexural strength, impact strength.

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## VII. Nomenclature

$\rho_f$ = density of the foamed samples

$\rho_u$ = density of the unfoamed samples

$\sigma_F$ = The flexural strength in MPa

$\sigma_T$ = tensile strength in MPa

A= Area of SEM micrograph

$A_s$ = in tensile tests is the cross sectional area of the gauge length in the sample, in  $m^2$

b= in flexural tests width of beam tested, in mm

d= in flexural tests depth of beam tested, in mm

f = in impact tests, factor for conversion to joules; f = 0.11299

$F_T$ = in tensile tests, the maximum load in Newton

h= in impact test, mean-failure height as applicable, in inches

K= correction factor for glass = 0.34 (between 2D and 3D arrangements)

L = in flexural tests, support span; in mm

M= Magnification factor of SEM micrograph

$M_a$ = weight of the foamed samples were measured in air

MFE= mean-failure energy in J

$M_s$ = mass of the rectangular samples

$M_w$ = weight of the foamed samples were measured in distilled water

n= Number of bubbles

No= the cell density

P= in flexural tests maximum load in N

$T_c$ = Theoretical tensile strength of composite

$T_g$ = Theoretical tensile strength of glass

$T_h$ = Theoretical tensile strength of HDPE

$T_w$ = Theoretical tensile strength of wood

$V_f$ = void fraction

$V_g$ = volume fraction of glass

$V_h$ = volume fraction of HDPE

$V_s$ = volume of the rectangular samples

$V_w$ = volume fraction of wood

$w$ = in impact test constant mass in lb;  $w = 5$  lb

## **Chapter 1 Introduction**

This chapter serves as an introduction to the topic of this study. The main topic is the advancement of the technology development for better quality Wood Plastic Composites (WPC) through processability studies and improvement of the mechanical properties of WPC through the addition of small amounts of glass fiber. A minor topic deals with batch foaming WPC. The introductory chapter begins by providing a context to the overall study. Changes and developments in society that make the study of WPC important are discussed. Such changes include the substantial increase in human population and the consequential strain on natural resources such as forests and the associated harmful results. The green mentality and the shift in attitude favouring environmentally-friendly products are also discussed to highlight the momentum products such as WPC are gaining these days. New methods and technologies in biocomposites that are worth examining along with theories worth testing are then mentioned. In the next section, the objective of the study is stated. Finally, the chapter is concluded with a look ahead to the rest of the dissertation.

## **1-1 Background**

### **1-1-1 Human population**

There has been an unprecedented increase in human population over the past few decades. The current world population of about 6.5 billion is forecasted to double to about 13 billion in the next six decades [1]. Such increase puts a heavy burden on the planet and its already dwindling resources. Wood has been a basic and invaluable commodity for humans as far back as history goes. It has been applied in basic human necessities such as shelter and fuel to more modern applications such as decoration and electricity distribution lines. The demand for wood will naturally only increase as populations increase, studies show that there is a positive and significant correlation between population growth and deforestation [2]. The insatiable quest for wood led to massive deforestation in the past, especially in the developing world where wood is more valuable to the simple lifestyle. Despite warnings and measures to curb this destructive behaviour, deforestation rates have increased in some developing countries [2]. The importance of forests extend beyond the supply of wood to benefits such as stabilization of the climate, providing biodiversity and purification of water [3, 4]. Losing forests has other harmful effects such as reduction of rainfalls, increase of atmospheric carbon and adverse effects on human health [1, 5].

### **1-1-2 Waste Disposal**

Another pressing environmental issue is the increasing global waste and difficulty in disposal. It is well-documented that the planet is running out of materials based on fossil

fuels (such as plastics) and other finite resources such as metals. Furthermore, the disposal of such material that are neither biodegradable nor recyclable is in itself harmful. Methods such as incineration pollute the environment and dumping in landfills harms the habitat. Governments and international organisations were thus forced to set legislations favouring materials that are recyclable or come from renewable resources. For instance, in the U.S. executive Order 13101 (1998) titled '*Greening the Government Through Waste Prevention, Recycling and Federal Acquisition*' set priorities and inclinations towards materials that have considerable amounts of biobased content [6]. Also, European Guideline 2000/53/EG, directed by the European Commission stipulates that 95% of the weight of an automobile has to be recyclable by 2015 [7].

Two major contributors to waste are plastic and wood. Every year, 100 million tonnes of plastics are produced around the world [8]. Most of this huge amount end up incinerated or dumped in landfills; for instance, in the U.S. plastics accounted for about 12% of the total municipal solid waste in 2005. Similarly, the majority of wood waste ends up in landfills. In 2002 about 63 million metric tonnes of wood waste was generated in the United States alone [9].

### **1-1-3 Environmental Mentality**

Such adverse affects of irresponsible human actions and the manifestation of global warming have led to an environmental paradigm shift around the world. Governments are drafting legislations and customers are demanding products that result in more environmentally friendly practices. This is more than a 'knee-jerk' reaction to the extreme weather phenomena that are frequent as of late around the world. This new paradigm shift

has forced manufacturers along with material providers to consider the environmental affect of their products throughout their life cycle (such as processing, disposal etc) [10]. This demand for environmentally friendly practices through green methods has been at the centre of global campaigns by social movements which aim to protect the environment through sustainable developments [10, 11].

As a consequence of the abovementioned and other reasons, there is a new interest in renewable resources. Science and industry turn to nature to solve the growing environmental problem. Furthermore, natural materials are readily available for the most part which encourages current research into them [12-14]. The focus of research work these days revolves around maximizing the efficiency of raw materials and minimizing the creation of waste [7, 15]. Using natural fibers in different applications is seen by some as a 'panacea' for the various challenges that the environment faces [10]. Plastics revolutionized the world of material in the past, and now the same world might be revolutionized by green composite material [16]. A specific area where some of the focus has shifted is the building sector. Currently, buildings are a major harm to the environment due in part to the waste they generate and the energy they consume. For instance, it is estimated that in Europe the service sector accounts for about 40% of energy use and green house emissions with the majority coming from buildings [17, 18]. In china, buildings consume 11.3% of the energy generated and come second only to the industry sector in energy consumption, and this percentage is forecasted to rise [19-21]. It is thus beneficial to find materials that are lighter in weight than traditional building materials (concrete, metals etc.) to reduce energy consumption.

When it comes to waste, the adverse effects of buildings (throughout their life cycle) are as alarming. It is estimated that construction account for about 50% of the waste in a landfill in the UK , 20-30% in Australia and about 33% in the US [22-24]. Such statistics call for an immediate search for better materials to be used in structural applications to replace conventional materials [6, 10].

The above factors along with others call for the development of materials that are environmentally friendly. WPC can play a role in this movement since it can reduce and reuse waste [25]. Furthermore, WPC have the ability to replace wood, which would help with deforestation issues [26-28]. The use of WPC is in line with the green movement and they are a promising sustainable green material that can be viable [7, 29]. All this enthusiasm will not be enough unless green materials become viable technically and commercially, something that needs to be addressed before they are mass marketed [30]. The question is whether such material will have the necessary mechanical strength and durability along with their positive environmental impact; a lot of work and research is necessary to answer this question [10].

#### **1-1-4 WPC**

WPC have benefited from an increasing number of research studies and developments in technology. Using wood in a thermoplastics matrix is a relatively recent phenomenon driven by three major current factors. Besides the already-discussed environmental regulation, factors giving WPC momentum include: improvements in processing technology and development of suitable chemical coupling agents (which bind the

chemically incompatible plastic and wood together) [29-31]. An indicator of this is the increasing number of patents in WPC [29]. WPC involve two different industries that are not very familiar with each other [32]. The plastic industry is used to high flowing, high temperature processing conditions, which is not common in the forest industry leading to difficulties in manufacturing WPC. In fact, most of the current WPC processing methods are based on those used by the plastic and composite industries [30]. Major concerns when it comes to processing wood and other natural fibers are associated with moisture release during processing which is different in plastic processing [30]. The typical high temperature used in plastic processing leads to moisture and volatile release in natural fibers which have negative side effects [33]. Furthermore, for natural fibers to be well-dispersed in the plastic matrix, high speed mixing (which typically requires high temperatures) is necessary. Modifications and adjustments have been made (and still need to be made) in order to combine both industries in one. These include special compounding technologies that balance between high temperature needs of plastics and low temperatures necessary for processing natural fibers [34]. Profile extrusion is the most economical process for producing shaped final products. The production rates are considerably faster than other methods. Thus it is the preferred industrial method when it comes to WPC. Currently, there are many unique processing technologies (mainly based on extrusion) being developed to meet the special needs of WPC [35].

The different nature of wood and plastic requires special binding agents. In the absence of these binding (or coupling) agents, both phases do not mix well and the stress transfer required to improve the mechanical properties of the composite does not occur [30]. Hence, the presence and optimization of coupling agents is a major requirement in WPC.

There has been much research focusing on the optimum coupling agent but more research is required in this integral area [36].

In order for WPC to be applied in more areas, improvements in their relatively low strength, heavier weight and density are necessary [33, 34, 37, 38]. The idea of using more than one filler in WPC (known as hybrid systems) has gained interest in recent years [39]. Having more than a single filler is useful since the end product will benefit from certain characteristics of each filler. These hybrid systems can benefit WPC by combining the high strength properties of glass with the low weight and sustainability of wood [38]. In particular, the addition of glass to WPC has proven to improve their mechanical properties [34, 39-41]. Improvements in mechanical strength can enable WPC to gain more ground in structural applications, which can dramatically alter the nature of materials used in such applications [6]. However, employing hybrid systems in WPC is still a new concept [42] and additional research is required; for instance in improving the adhesion of hybrid systems [6, 34, 42].

As previously mentioned, WPC suffer from heavier weight and higher density [33, 34, 37, 38, 43]. Foaming WPC can considerably improve their mechanical and physical properties. Reduction in weight and density will save costs at production, reduce the amount of resources necessary and save energy in applications such as buildings and automotive. However, more research work about foaming WPC is needed since the available literature is very sketchy and promising areas such microcellular foaming are not fully understood yet [43, 44].

## ***1-2 Objective***

Previous studies have shown the potential of WPC. These materials fit right in with the green movement and have the ability to cause substantial changes in some industries. However, WPC need better processing methods and suffer from drawbacks that prevent them from fulfilling this potential. Several studies conclude that more research work into WPC is necessary. The objective of this study was to contribute to and augment previous studies by:

1. Studying the processability of WPC using conventional methods (extrusion in general and profile extrusion in particular.)
2. Adding low amounts of glass fiber to improve the mechanical properties (tensile, flexural and impact) of WPC.
3. Investigating the concept of foaming in WPC by examining ways to optimize the parameters.

## ***1-3 Outline of Thesis***

Chapter 1 serves as an introductory chapter providing background information on WPC highlighting the importance of the study and setting its objectives. Chapter 2 is a survey of the literature providing more information on the topic illustrating more issues. Chapter 3 contains the methods used for shedding light on to the material and technologies employed in the study. Chapter 4 provides the results of the processability studies, those of the addition of glass fibers to WPC and the concept of foaming along with a discussion

of these results. Chapter 5 is a conclusion of the study summarizing its findings and providing recommendations for future work.

## **Chapter 2 Review of the Literature**

The chapter starts with an overview of composites in general and biocomposites in particular. Natural and inorganic fillers are then briefly discussed. More focus is then directed towards a subset of biocomposites which is Wood Plastic Composites (WPC). An overview is given followed by aspects such as the advantages of WPC, their application and market share, means of manufacturing and current challenges. A closer look is then taken at improving the processability of WPC and enhancing their mechanical properties through the addition of coupling agents and glass fibers. Finally, the concept of foaming is reviewed.

## **2-1 Composites**

Mathews and Rawlings define Composites as materials having two or more distinct constituents each present in reasonable proportion and with different properties [45].

Composites are not a new concept and have been used in ancient civilizations. There are natural composites such as wood and bone along with synthetic ones such as bricks and concrete. The motivation behind composites is the improvement in material performance, such as reduction of weight and cost and increasing strength by using the best properties of each of the constituent components. Composites generally consist of a reinforcement phase of strong, stiff materials that is frequently fibrous in nature, embedded in a continuous matrix phase. The matrix phase is often weaker and more compliant than the fibrous one. Two of the main functions of the matrix are to transmit externally applied loads, via shear stresses at the interface, to the reinforcement and to protect the latter from environmental and mechanical damage.

### **2-1-1 Biocomposites**

Fowler et al. [30] defined biocomposites as composite materials consisting of one or more phase(s) derived from a biological origin. The reinforcing phase may come from plant fibres (cotton, flax, hemp) or fibres from recycled wood or waste paper. The Matrix phase may be derived from renewable resources such as vegetable oils or starches. However, at the present time most matrices are synthetic, fossil-derived polymers thermoplastic such as polyethylene (PE), polypropylene (PP), polystyrene (PS) and polyvinyl chloride

(PVC), or thermosets such as unsaturated polyesters, phenol formaldehyde, isocyanates and epoxies. The properties of a composite are dictated by the intrinsic properties of the constituents. Materials from renewable resources are being sought to replace the matrix phase of composite materials along with the reinforcing elements to tackle some of the sustainability issues associated with using synthetics in composites [46, 47]. These days, the main markets for most biocomposites are the construction and automotive sectors [7]. However, with further developments and improvements, new applications and opportunities and applications will arise. A particular area that offers significant potential for growth is in the replacement of pressure treated wood. Tighter restrictions have been introduced on the use of certain preservatives [48, 49]. Additionally, improvements in the mechanical performance of existing biocomposites [50] through innovation, such as the introduction of new processing, additives and fibre types may result in more diverse applications.

Drzal L.T. et al [6] gave an overview of natural fiber reinforced polyester composites emphasizing the prospects of such composites in building applications. Plastics (either bio or petroleum based) cannot be used for load bearing application because of their lack of stiffness, strength and dimensional stability. On the other hand, fibers have the necessary stiffness and strength but are not fit for load bearing applications due to their fibrous structure. In fiber-reinforced plastic composites, the fibers act as a reinforcing agent held together by the plastic matrix resulting into a material form with sufficient strength and stiffness for structural applications. Composite materials based on renewable resources (biocomposites) have the potential to produce low-cost structural components which serve as more sustainable alternatives to conventional structural materials.

A motivation for these emerging products is the ongoing pressure to conserve forests for ecological and climatic conditions. The replacement of conventional building materials, such as pure petroleum-based plastics and additives such as glass fibers, are becoming priorities. Governmental regulations, such as Executive Order 13101 (1998) in the U.S. titled '*Greening the Government Through Waste Prevention, Recycling and Federal Acquisition*' set priorities and inclinations towards materials that have considerable amounts of biobased content. Besides environmental benefits, the use of Biocomposites has advantages such as lighter weight, good insulation and sound absorption properties and reduction in energy consumption. The authors conclude their paper by asserting the need to better understand biocomposites and carry out additional research into aspects such as: surface treatment of natural fibers, moisture and thermal durability and stability, economically viable manufacturing processes and new design methodologies involving hybridization at both the constituent and structural levels.

Adhikary et al. [25] explored the use of waste sawdust and post-consumer recycled HDPE for the production of the HDPE-wood flour composites. The global production of plastics, at about 100 million tonnes per annum, results in a significant percentage of municipal solid waste [8]. For instance, of the 246 million tonnes of MSW generated in United States in 2005 waste plastics account for 11.8% [51]. There have been several attempts to recycle the post-consumer plastics to reduce the consumption and environmental impact of virgin plastics. In Western Europe, plastic recovery in 2004 was about 8.25 million tonnes (39% of total amount of plastics consumed) [52]. In the United States the recycled amount was only 5.7% of the plastics generated in 2005 [53].

It is well established that the properties of the recycled high density polyethylene (rHDPE) obtained from the post-consumer milk bottles were not that different from those of virgin high density polyethylene (vHDPE). Hence, rHDPE could be used for various applications similar to vHDPE [10]. Besides lessening waste disposals, using recycled plastics reduce the product costs. rHDPE pellets and flakes are 31–34% cheaper than vHDPE [54]. Similarly, a large amount of wood waste is mainly destined for landfill. In 2002 about 63 million metric tonnes of wood waste was generated in the United States alone [9]. The use of waste wood (sawdust) in wood plastic composites (WPC) helps in decreasing these disposal costs. Past studies show that waste wood in the form of wood flour, fibres or pulp is suitable as a filler for polyolefins [55, 56].

## ***2-2 Use of Fillers in Polymeric Composites***

### **2-2-1 Natural Fillers**

Ashori [7] reviewed developments in using plant-based fillers in WPC and their applications in automotives industries. Fibers can be classified into two main groups: manmade and natural. Natural fibers are then subdivided based on their origins, coming from minerals, animals or plants. Plant fibers are mainly composed of cellulose while animal fibers consist of proteins. Plant fibers can be classified according to which part of the plant they are obtained from. Drzal et al. [6] gave a more detailed description of natural fibers, their subdivisions and some examples in Figure 2-1. In figure 2-2, pictures of common biofibers are provided.

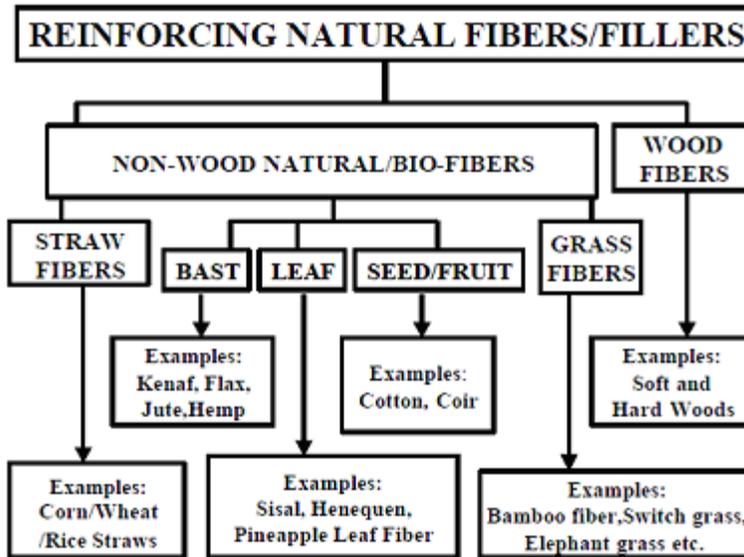


Figure 2-1 Natural Fillers in Biocomposites [6]



Figure 2-2 Types and Sources of Common Biofibers [6]

Some of the advantages of natural fibers over traditional reinforcing fibers (such as carbon and glass) the authors mention include their low density (1.2-1.5 vs. 2.55 g/cm<sup>3</sup> for E-glass), low cost, adequate specific properties, and biodegradability [6]. In reality, biocomposites derived from natural fibers and petroleum-based thermoplastics are not

fully environmentally friendly. That is because the matrix resins are non-biodegradable; however, the biobased content of the final composite material falls within the definition of biobased materials. It is noteworthy that these Biocomposites maintain a balance between economics and environment. Nonetheless, glass fibers will remain the leading reinforcement material owing to their excellent overall performance and their low cost. Still, the use of natural fibers in FRP (fiber reinforced plastics) to replace glass fiber is gaining momentum in a few applications. The idea of a composite containing both natural and synthetic fillers (hybrid systems) for improved structural performance is a well-known idea in engineering design. The authors mention a specific study that put the optimum glass amount that improve the tensile, flexural and impact properties of a hybrid of glass and sisal at 5.7% while that of glass and pineapple leaf fiber is 8.6% [42].

Mohanty et al. [57] investigated the use of biocomposites in the auto industry. Manufacturers are moving away from steel towards other materials such as aluminum, plastics and composites to reduce vehicle weight [58]. Experts estimate that polymer and polymer composites will comprise about 15% of total car weight in the near future [59]. Furthermore, the demand for natural fibers as fillers/reinforcements in plastics in North America is forecasted to increase annually by about 60% in the building industry and by about 30% in the automotive industry [60]. Figure 2-3 puts these numbers in perspective with those of other fillers; figure 2-4 illustrates the North American market for natural fiber composites.

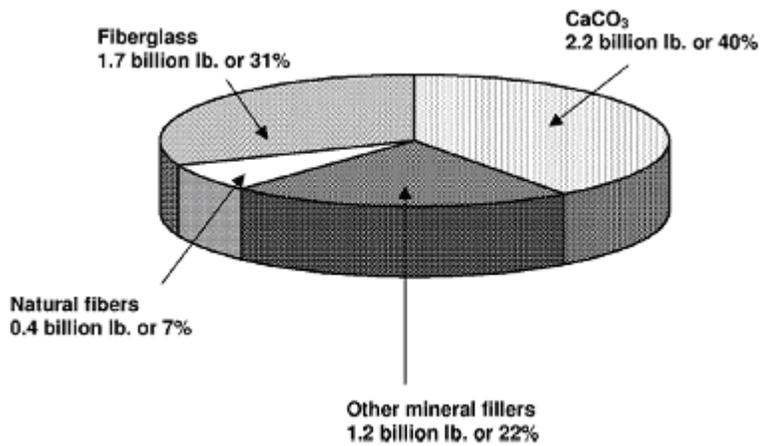


Figure 2-3 Demand for plastic fillers/reinforcements in North America in 2000 [57]

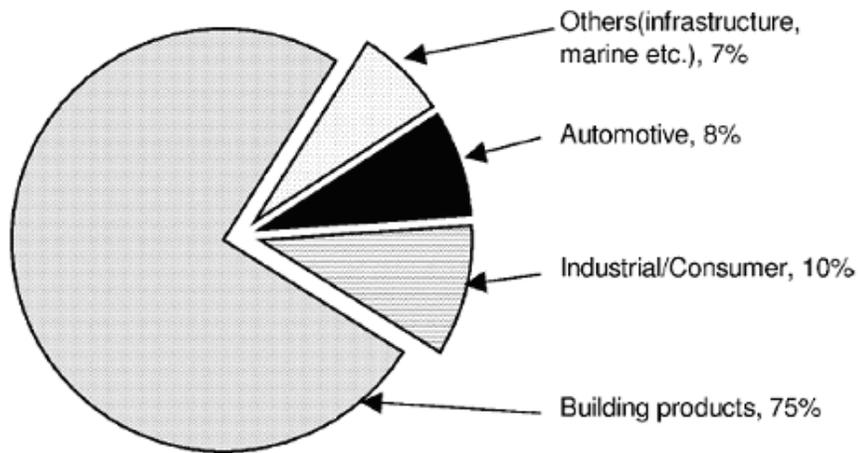


Figure 2-4 Markets for natural fibers composites in North America in 2000 [57]

A drawback of natural fibers when compared with synthetic fibers is that their behaviour is dependent on factors such as their source. For instance, their mechanical properties depend on whether the fibers are taken from plant stem or leaf, the age of the plant, and the geographic location of the plant.

Wood is a major natural fiber used as filler in different industries and its nature was investigated by Adhikary et al. [25]. Wood is a lignocellulosic material made up mainly of three constituents: 42–44% cellulose, 27–28% hemicelluloses, and 24–28% lignin with

along with some minor constituents [61]. The aligned fibril structure of cellulose (a major component of wood) along with strong hydrogen bond has high stiffness; hence the addition of the wood flour increases the stiffness of the thermoplastic based composites. Furthermore, Lignin is an amorphous polymer which does not greatly affect the mechanical properties of wood flour; yet it plays a central role in binding the cellulose fibrils that allow efficient stress transfer to the cellulose molecules.

### **2-2-2 Inorganic Fillers**

According to Biron [62], glass fibres are the most commonly used fibers for plastic reinforcements and represent 95% of all reinforcements used. Glass fibres are normally made from glass spun in the melt that is then assembled and protected by sizing with silane or organic materials. Some of the advantages of glass fibres include their versatility of the sizing, which leads to good compatibility with all polymers. Also, they have high specific mechanical properties, dimensional stability and chemical resistance. Other properties of glass include their high thermal resistance and density, brittleness under high stresses during processing and abrasiveness which is harmful to tools. There are several types of glass but E-glass represents about 90% of all fiber reinforcements, other types include S-glass and D-glass. When used as a reinforcement of polymers, glass fibers come in various forms. A common form is the chopped (or milled) glass fibers which are dispersed into the polymer. They are typically 0.1–20 mm in length and 5–25  $\mu\text{m}$  in diameter with aspect ratio from 4 to 4000. Other forms include mats which are chopped fibres 50 mm in length that are held together with a binder to form a sheet. The properties of chopped glass fiber reinforced polymer depend on factor such as the fibre

content, the aspect ratio, the sizing of the fibres to enhance adhesion to the matrix, the real length of the fibres in the final part, the quality of the fibre dispersion and the anisotropy in the final part.

Hetzer and De Kee [63] shed some light on another type of fillers: nanofillers. The main difference between nanofillers and conventional ones lies in the size of the filler. The size of conventional fillers ranges between 10  $\mu\text{m}$  and 1 cm; that of a nanofiller ranges between 1nm and 500 nm. Nanoclays, a type of nanofillers, have a high aspect ratio which allows lower loading of fillers to attain the desired mechanical results. About 3 to 5 % loading of nanoclay is used as opposed to the typical greater than 30% for conventional fillers. The high aspect ratio of the nanoclays is believed to allow for a greater interfacial area for adhesion which in turn enhances reinforcement properties. The use of nanoclays is hampered by their high cost and their use is still limited pending further research.

### ***2-3 Wood Plastic Composites***

According to Ashori [7] WPC include any composites containing plant fibers and thermosets or thermoplastics. He contends that one of the main goals of such composites, or 'green chemistry' in general, is to use raw natural products to a maximum efficiency and bring waste to a minimum. These goals are in-line with those of biocomposites and are further aided by regulations. For instance, it is reported in this paper that according to European Guideline 2000/53/EC, directed by the European Commission, 85% of the weight of a vehicle was to be recyclable by 2005. Furthermore, this percentage is to be increased to 95% by 2015. Moreover, 85% of the vehicle should be recoverable through

mechanical recycling or reuse and 10% through energy recovery or thermal recycling. This directive has not been met satisfactorily and a new report on its implementation of is due on June 2009 [64, 65]. Therefore, it is not surprising that almost all the major auto manufacturers in Germany (Volkswagen, Audi Group, BMW, and Daimler-Benz) now use WPC in a variety of applications. Such initiatives render plant fibers the fastest-growing type of polymer additives (as fillers and reinforcements) [10].

Faruk et al. [44] in their review of microcellular wood fiber reinforced polymer composites describe WPC as novel materials that combine the best attributes of wood and plastics. On one hand, the plastic industry sees in wood a relatively inexpensive filler that is readily available which can enhance mechanical properties such as stiffness and processing conditions such as increased profile extrusion rates [66]. On the other hand, the forest industry see plastics as means to make innovative construction materials with characteristics that wood does not have; for instance resistance to environmental adversities such as moisture and insect attacks. It is also mentioned that almost all WPC can be machined, sanded, stained, and fastened in the same manner as wood without needing any further technologies.

Hetzer and De Kee [63] cite yet another environmental regulation, the Environmental Protection Agency's (EPA) Chromated Copper Arsenate (CCA) 2007 which may soon eliminate the use chromate copper arsenic (CCA) [67]. CCA is typically used as a pesticide and a wood preservative in the pressure treatment process and can leach out of the wood to contaminate the water table. Clearly, the use of WPC will eliminate this problem further enhancing the profile of WPC. Owing in part to these emerging new

environmental regulations, the WPC market is going through a substantial growth, averaging over 20% annually since 1998.

Jiang and Kamdem [68] state that the commonly used thermoplastics in manufacturing WPC include high and low density polyethylene (HDPE and LDPE,) polypropylene, polyvinyl chloride, polystyrene and polymethylmethacrylate (PMMA.) These facts are further broken down, according to the North American market share in 2002, into an 83% market share for PE, 9% for PVC, and 7% for PP-based WPC as illustrated in figure 2-5 [69].

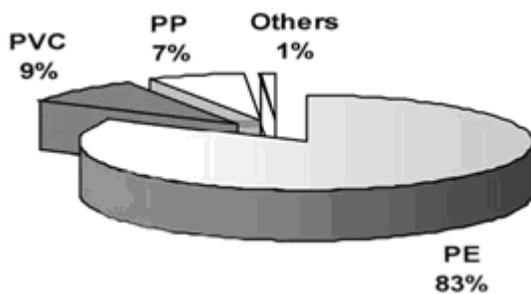
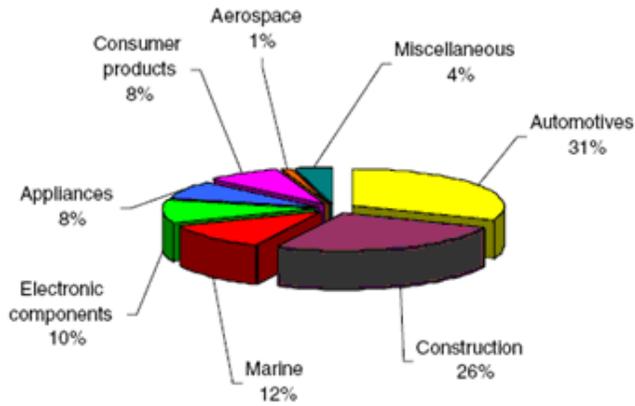


Figure 2-5 North American market share in 2003 of various thermoplastics in WPC [69]

### 2-3-1 Advantages, Applications and Manufacturing of WPC

The advantages of WPC are similar to those of biocomposites mentioned earlier. Ashori [7] mentions the obvious fact that industries are always on the look out for innovative materials and improved processes to produce better products to increase their profit margin and keep their technological edge. Therefore, it is not surprising to see WPC being used in a large number of applications from aerospace to electronics as evident in figure 2-6 [7].



**Figure 2-6 Areas of applications of WPC in 2000 in Europe [7]**

Some of the advantages of using WPC in the auto industry are: reduction in material weight and energy consumption, enhancement of acoustic performance and processing time, lowering production cost, improving safety and shatterproof performance under extreme temperature changes, and improving biodegradability of the auto interior parts [70]. WPC are used as trim parts in dashboards, parcel shelves, seat cushions, backrests, door panels and cabin linings.

Jiang and Kamdem [68] further explain the cost advantage of WPC by stating that wood fiber is one of the lower-cost fillers for plastics. Figure 2-7 illustrates this advantage; the average market prices of wood fiber is less than one-tenth that of glass fibers and is comparable to other low-cost inorganic fillers, such as talc and calcium carbonate.

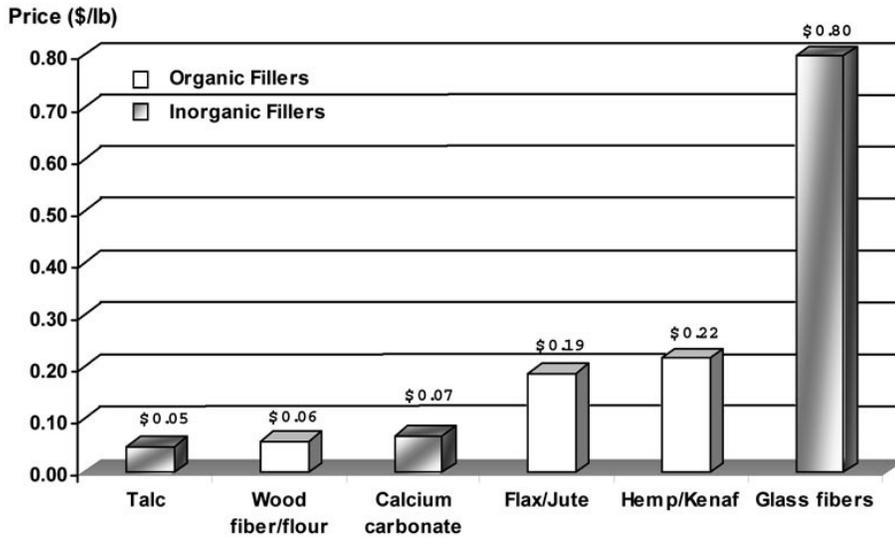


Figure 2-7 Average market price for plastic fillers in 2004 [68]

The use of WPC in construction applications is discussed by Fowler et al. [30]. Products formed from WPC are typically used in outdoor decking, window and door frames.

However, due to poor mechanical performance of WPC (as per the construction market) their uses are limited to non-structural applications. For example, the use of WPC in decking applications is limited to the deck and not the supporting structure.

The manufacturing techniques of WPC, Biocomposites in general, are mainly based on existing methods for processing plastics and other composite materials. Fowler et al [30] state the previous statement and explain some of these techniques. These techniques include press moulding, compression moulding, extrusion, and injection moulding. The majority of current Biocomposites that have thermoplastic matrices (such as WPC) mainly use compounding and extrusion in their manufacturing methods (especially when the fibers used are relatively short.) During compounding, the thermoplastic polymer is heated until it melts either by an external heat source or as a result of mechanical shearing

in the extruder. The wood flour, along with other additives, are then added and thoroughly mixed to obtain a homogeneous mixture. Once the ingredients have been thoroughly mixed, the compound can be extruded directly into the final form or pelletized and collected for further extrusion or injection moulding processes.

### **2-3-2 Challenges in WPC**

Rowell [32] concedes that there are some issues that need to be taken care of before WPC can reach its full potential. A major source for these issues is the fact that WPC involves two different types of industries. The plastic industry is accustomed to high flowing, high temperature processing conditions, something that the wood/agricultural industry is not really familiar with. Also, shrinkage/swelling in the plastic industry is normally due to temperature; while the biomass in the WPC shrinks and swells due to moisture.

Jiang and Kamdem [68] further discuss the challenges issues with WPC in their review on the research and technical development of WPC made of PVC and WF. The natural phase incompatibility between the non-polar, hydrophobic thermoplastic and the polar, hydrophilic wood causes a weak interface between the matrix and the filler. Furthermore, the dispersion of the fillers in the viscous matrix is hindered by strong wood-wood interactions resulting from the physical entanglement of the fibers, the polar nature of wood and the hydrogen bonding [71, 72]. The wood fibers usually contain 8 to 12% moisture, which under normal plastic processing temperatures will evaporate and swell the polymer melt, and result in inconsistent flow, irregular foaming and defects and voids inside the product [73]. Furthermore, wood also has both low thermal stability and bulk

density along with wide particle size range, causing problems with feeding and conveying while processing [74].

Bledzki and Gassan [73] argue that the main problem in processing WPC is the poor dispersion between the plastic matrix and the fiber which is caused by the tendency of untreated wood to form large aggregates, as discussed earlier. This in turn would considerably reduce the reinforcing ability of the fiber defeating one of the main purposes of using the fiber. A way to improve the dispersion of wood fibres is with the use of coupling agents or thermoplastic polymers. Another major problem in processing WPC is thermal degradation. This is due to the fact that the rate of thermal decomposition of lignocelluloses (a main component in wood) increases exponentially with an increase in temperature. Typical processing temperatures of WPC are in the range of 180–200 °C which is enough for decomposition to occur [75]. Such decomposition of the lignocellulose creates volatiles which generate voids in WPC, thereby diminishing their mechanical properties. Georgopoulos et al [76] echo the previous concerns by further pointing out that thermal degradation of the fibres in WPC leads to poor organoleptic properties (such as poor colours and odours) and generates gaseous products when processing takes place at temperatures above 200 °C. Such changes and emissions create low density, high porosity, and consequently reduced mechanical properties.

Furthermore, Rizvi et al. [33] conclude that as the wood fiber content increases, the effective viscosity of WPC increases significantly. Such increase causes the processing pressure to increase which create processing problems. Two possible solutions for this problem are increasing the processing temperature and/or using a resin with a high melt

index (MI.) However, increasing the processing temperature above 200 °C may lead to a degradation of WPC and volatile emissions, which in turn affects the processing conditions and the final properties of the product [43]. Furthermore, using a resin with a high MI is not recommended if the WPC is being foamed since a very high MI prevents the formation of a fine cellular structure. Moreover, typical WPC manufacturing techniques such as extrusion mixing, hot-press lamination and injection moulding are processes where melt-rheological properties are significant. Melt viscosity decreases as shear rate increases and increases as filler content increases (as mentioned above) [73].

### **2-3-3 Processing Methods and Strategies of WPC**

Caulfield et al. [35] maintain that extrusion is the most common processing method for WPC. It is capable of processing the highly viscous WPC and can also shape long, continuous profiles that are common in applications such as building materials. Such profiles range from a simple solid shape to highly engineered and hollow. Within the method of extrusion, there are many extruder types and processing strategies. For instance, some processors run compounded pellets through single-screw extruders to form the final shape. Other processors compound and extrude final shapes in one step using twin-screw extruders. Furthermore, there are others that use several extruders in tandem, one for compounding and the others for profiling.

Different equipments have been developed to handle the different processing requirements in WPC. For instance, it is advantageous to remove moisture from the wood component to improve processing; this can be achieved using several methods. It can be done during a separate compounding step in the first extruder in a tandem process, by

using the first part of an extruder as a dryer in some in-line process or by using other drying techniques. Besides drying methods, other equipment systems include those for materials handling, extruder design, die design, and downstream equipment. Downstream equipment are those necessary after the extrusion process, they include such as cooling tanks, pullers and cut-off saws. Equipment manufacturers are developing complete processing lines unique for WPC. Some of the new extrusion technologies being licensed use processes that are very different from conventional extrusion processing.

Other WPC processing technologies include injection molding and compression molding but their total production is much less than that using extrusion [35]. These alternative methods have advantages when processing of a complicated shape is necessary or when a continuous piece is not wanted. In such cases, the composite formulation must be modified to meet processing requirements. For instance, in injection molding low viscosity is needed which may limit wood content. There are different types of extrusion, such as filament extrusion, profile extrusion etc. However, the majority of WPC are manufactured by profile extrusion where the molten composite is forced through a die to make a continuous profile of the desired shape

Rowell [32] adds that more products can be made, with the same quantity of material, using profile extrusion. Furthermore, the products are lighter in weight and can be stronger. The ongoing development in extrusion equipment is progressing to produce more efficient mixing systems that produce faster processing throughput and minimize fiber damage along with new methods of product formations.

### 2-3-4 Use of Coupling Agent

Fowler et al. [30] studied some of the technological challenges in bringing biocomposites to a wider market. The interface between the matrix and the fiber in a composite is crucial in terms of performance. This is because the externally applied loads are transferred to the reinforcement via shear stresses over the interface. Therefore, for a proper reinforcing function, good bonding between the matrix and the fiber is necessary. Natural fibres are generally hydrophilic and do not bond well with the hydrophobic matrix polymer. Ways to promote adhesion between the two phases include introducing additives such as compatibilizers to the polymer/fibre mix during processing or chemical modification of the fibre prior to composite manufacture.

Rowell [32] explains the nature of the most common compatibilizers used today: maleic anhydride grafted polyethylene (MAPE) or maleic anhydride grafted polypropylene (MAPP). Figures 2-8 and 2-9 illustrate the mechanism of compatibilization.

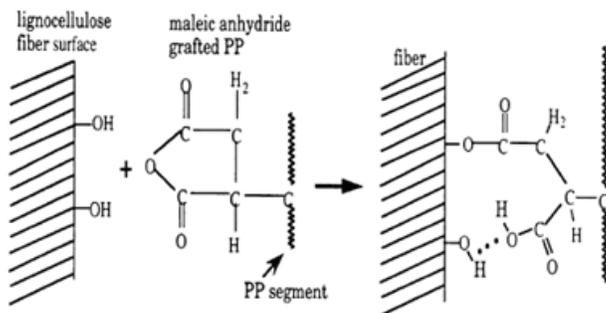
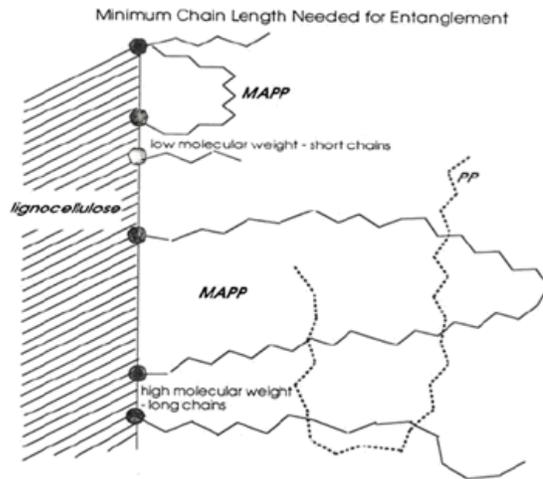


Figure 2-8 Mechanism of reaction between MAPP and a lignocellulose fiber [32]



**Figure 2-9 Entanglement of melted PP with the reacted MAPP-lignocellulose [32]**

First, the anhydride end of the MAPE/ MAPP reacts with a hydroxyl group on the surface of the biofiller forming an ester linkage. Then, the PE/PP tail on the grafted MAPE/MAPP gets entangled with the molten thermoplastic resulting in a mechanical link between the hydrophobic thermoplastic and the hydrophilic biofiber which also enhances the mechanical properties of the resulting composite [25].

Adhikary et al. [25] studied the effects of the fiber loading and addition of coupling agent on the mechanical properties of waste sawdust and post-consumer recycled HDPE. It was observed that the tensile strength of the composite along with the stiffness increased due to the addition of a coupling agent (maleated polypropylene). This increase is attributed to the improved interfacial bonding between the HDPE matrix and the reinforcing wood flour due to the formation of ester bonds as described earlier [77-80].

Lu et al. [36] investigated issues regarding the use of coupling agent in an HDPE-wood composite. The use of maleated polyethylene (MAPE) or maleic anhydride grafted-polyethylene (PE-g-MA) in HDPE/LDPE-starch composites has been extensively used as

a compatibilizer [81-82]. However, it is claimed that there have been few reports on the use of MAPE as a coupling agent in WPC [83] raising the need to assess the efficacy of MAPE in improving the interfacial bonding strength in WPC. Out of seven coupling agents investigated in this study, 100D and 226D (both maleated polyethylene produced by DuPont) improved the interfacial bonding strength between HDPE and wood the most. The reason is that the maleic anhydride groups of MAPE were easily hydrolyzed into double carboxylic groups. These double carboxylic groups were in turn more reactive for esterification under an initiator than the monocarboxylic groups of other coupling agents.

### **2-3-5 Mechanical Properties of WPC**

Improving the mechanical properties is a common theme in many WPC studies. A major stumbling block in WPC realizing their potential is their weak mechanical strength. It was observed by Adhikary et al. [25] that the tensile strength of the composite and its stiffness increase due to the addition of a coupling agent (maleated polypropylene). Also, the composite material becomes stiffer with the addition of wood flour; however, the strain at failure decreases (which means less ductility). Furthermore, in the absence of a coupling agent, numerous irregularly shaped microvoids occurred which deteriorated the transfer of stress from the matrix to the fibers which had an adverse effect on the mechanical properties. However, these composites had a higher maximum strain at failure than the MAPP-coupled ones. Flexural strength studies reveal a similar trend, as of tensile strength, although less variation is observed in the flexural strength with different formulations than in the case of tensile strength [25]. This is justified by the same coupling mechanism as in the previous section.

The load–displacement curves of pure HDPE and HDPE-wood composites were also studied to observe the effects of the coupling agent addition and filler loading [25]. These curves initially display relatively high slopes but as failure occurs, the loads drop off quickly. The pure HDPE specimen reach their ultimate elongation capacity after significant deformation and then the load displays a gradual decrease until the test's conclusion, which implies a ductile behaviour. Also, adding the wood flour loading in the composites increased the stiffness but reduced the elongation at break. This brittle behaviour is explained by poor interface bonding between the HDPE matrix and wood flour along with the stress concentration at the fiber ends. During loading, fractures occur at filler locations in the composite which are more brittle than the other parts. Therefore, during loading, these composites show little initial change in the specimen appearance until the maximum load was reached when the specimen failed suddenly with extensive breakage at the interface between the matrix and the filler. As in the case of modulus, improving the adhesion between fibers and HDPE did not enhance elongation at break.

Lu et al. [84] observed that the tensile strength increased as the wood fiber content increases (at low fiber loading, < 15%) and peaked at 15% fiber loading. The tensile strength then decreased as the fiber loading increased, especially at loadings above 20%. It is important to note that this is valid when no coupling agent was used. Different coupling agents at different levels were then added to a 50% HDPE – 50% wood composites and the resulting tensile strengths were compared. It was concluded that the two maleated polyethylenes (100D and 226D) showed the best results. Furthermore, the interfacial bonding strength was found to be related to the concentration of the coupling

agents. Amongst all the tested coupling agents, the coupling agent performance was better at low concentrations (around 3 %.) As the coupling agent loading was increased to about 5% the tensile strength decreased (with 226D) and did not show significant increase (100D). A possible explanation for this behaviour was stated to be that an excess of coupling agents generated several byproducts that interfered with the coupling agent resulting in low bonding strength at the interface [81-82]. Also, the existence of an excess coupling agent may enlarge the gap between the HDPE matrix and the wood flour and weaken their interface.

Pickering et al. [85] provided an explanation for the general reduction in the strength of wood fibres as the fibre weight percentage increases, even in the presence of coupling agents. This reduction can be attributed to fibre agglomeration which is more likely to happen at higher fibre contents. Also, fibre–fibre interactions from hydrogen bonding are considered to greatly hinder fibre dispersion [86]. In general, samples made with coupling agents show improved interfacial bonding, particularly at the low fiber loading where the matrix retains close proximity with the fibre. In this study; however, wood fibers appear to be well dispersed through all of the samples. Hence, the abovementioned reason does not seem to explain the reduction of strength at increased fiber loading. An alternative rationalization of this reduction may have to do with the increased occurrence of voids at higher fibre contents. Moisture and other volatiles emitted during fiber processing may create these voids.

Cui et al. [87] studied the affect of surface treatments (including coupling agents) on the mechanical properties of a waste saw dust-post consumer HDPE composite with the aim

of using the composite in structural applications. One of the properties investigated was the impact strength of the composite. The impact strength of this WPC decreased with the increase in fiber loading for all of the surface treatment methods. This might be due to the presence of wood fiber ends within the body of the WPC which can cause crack initiation leading to failure. These fiber ends act as notches which in turn generate sizeable stress concentrations when loaded which may lead to the initiation of cracks in the HDPE matrix. Also, the interactions between neighbouring wood fibers seem to restrict matrix flow causing the matrix to be more brittle [88].

John and Thomas [16] reviewed cellulosic fiber-reinforced polymeric composites. An interesting conclusion the authors mention is that natural fibers act more ductile if the microfibrils (millions of fibrous units [30]) have a spiral orientation to the fiber axis. Moreover, if the microfibrils are parallel to the fiber axis, the fibers will be inflexible, rigid and have a high tensile strength. It is also concluded that the tensile strength of a natural fiber increases with increasing cellulose content.

### **2-3-6 Use of Glass Fibers**

It maybe beneficial to add another filler (such as glass) besides coupling agents to improve the mechanical properties of WPC. Panthapulakkal and Sain [89] studied the properties of injection-moulded natural fiber (hemp) and glass fiber reinforced polypropylene composites intended for semi-structural or structural applications. The addition of 15 wt % glass fiber along with 25 wt % hemp and 5 wt% compatibilizer increased the tensile strength of the composite by 13%. This improvement was attributed

to the fact that glass fiber is stronger and stiffer than natural fiber. Nonetheless, the authors cite a previous study of theirs along with few others where the addition of glass fiber has little or no affect on the tensile properties of a natural fiber composite. A possible explanation for that is that glass fibers may suffer breakage during processing.

Increasing the glass fiber content from 0 to 15 wt % improved the impact strength of the composite by 35% [39]. Typically, the impact strength of a composite is a measure of the material's ability to resist fracture failure under stress applied at high speed and is directly related to the material's toughness. Fibers play a vital role in the impact resistance of fiber reinforced composites since they interact with the crack formation and act as stress-transferring medium. The increase in impact strength with glass fiber addition may then be attributed to the improved resistance offered by the glass fibers in the composites.

Kalaprasad et al. [40] investigated the tensile properties of surface treated sisal-glass reinforced LDPE. It was concluded that the tensile properties, except elongation at break, increase as the volume fractions of glass increases. The increase in tensile strength of this hybrid composite is attributed to the higher tensile strength of glass fibre relative to the sisal fibre and also to the higher degree of dispersion of sisal fiber in the presence of glass fibres [90, 41]. It is also noted that since glass fibres have a smaller diameter, they can pack well in the interstitial spaces between irregularly spaced sisal fibres, leading to a close-packed and denser composite structure. Consequently, agglomerates of fibres formed from a polydispersed (versus a homogenous monodispersed) system, such as this hybrid system, are supposed to carry a larger proportion of load. Therefore, the tensile properties of such systems are expected to be better. However, the agglomerates formed

between glass fibers only will behave differently and therefore the agglomeration hinders the uniform dispersion of fibres in the matrix. Two general conclusions reached in this paper are the fact that almost every modification improves the tensile properties of the composites and that the volume fraction of glass in the system increases the tensile properties slowly and until they either level off or decrease.

Jiang et al. [41] studied the impact strength of short glass fiber/wood reinforced PVC. They concede that there are some disadvantages associated with glass when it is added to WPC (increase in cost, equipment abrasion etc.) Nonetheless, glass fiber is potentially a good reinforcement agent for such composites especially in cases where high impact strength and modulus are required. It is also concluded that glass fiber lengths and content levels are vital to the mechanical properties of the final products [91].

## ***2-4 Foaming***

Foaming is another technology that aims to improve WPC properties. Saunders and Klemperer [92] explain some of the fundamentals of foam formation. Most foamed polymers (including thermoplastics) are produced by the dispersion of gas throughout a fluid polymer phase and stabilising the resultant foam. Generally, foam expansion is achieved by increasing the bubble size before stabilising the system. The dispersed gas can come from an array of sources: a gas (such as CO<sub>2</sub>) can be dissolved in the polymer (normally under pressure); a gas may also be generated by the decomposition of a chemical blowing agent - such as p,p'-Oxybis (Benzenesulfonylhydrazide) ;or a low boiling liquid (such as pentane) maybe dissolved in the polymer and then converted to a

gas either by reducing the pressure or heating. Upon the dispersion of gases, bubbles typically begin to form. They form due to pressure reduction or heat. The foam may be closed cell (if the cell membranes surrounding the bubble remain intact) or open cell (if the surrounding cells rupture.)

An important concept in the cellular structure of foamed polymers is that of microcells which was discussed by Park et al [93]. The concept was to create a large number of bubbles that are smaller than the existing flaws in a polymer. Suh developed this method as a response to an industrial demand to reduce material cost for some polymer products without compromising mechanical properties. Microcellular foams are characterized by cell densities in the  $10^9$ - $10^{15}$  cell/cm<sup>3</sup> range, cell sizes in the 0.1-10  $\mu$ m and specific density reductions in the 5-95% range. Microcellular plastics are produced by using a thermodynamic instability in a polymer and gas solution to promote a large bubble density in a polymer matrix. There are two basic steps involved in processing microcellular plastics. The first is polymer/gas solution formation which is accomplished by dissolving an inert gas (such as CO<sub>2</sub> and N<sub>2</sub>) in a polymer matrix under a high pressure. The solution contains a gas concentration of between 5-20% in the polymer matrix. The solution formation is governed by gas diffusion in the polymer, which is typically a slow process. Ways to increase this process includes increasing the gas pressure and the temperature. The second step the authors describe involves subjecting the solution to a thermodynamic instability to nucleate microcells. To achieve this, the solubility of gas in the solution is lowered by controlling the pressure, the temperature or both. Now, the system seeks a state of lower free energy which results in the clustering of gas molecules in the form of cell nuclei. This provides a relatively short free distance for

the gas molecules in the solution to diffuse through before reaching a cell nucleus. The free energy of the system is lowered as the gas diffuses into the cells. Nucleation can occur throughout the material (homogeneous) or at high energy regions such as phase boundaries (heterogeneous). The nucleation process is integral in producing microcellular plastics because it governs the cell morphology of the material and subsequently its properties.

Upon nucleation, cells continue to grow as the available gas diffuses into cells, provided that little resistance is faced [93]. The cells grow and reduce the polymer's density as more gas molecules diffuse from the polymer matrix into the nucleated cells. The rate of this growth is governed by the diffusion rate and the stiffness of the viscoelastic polymer/gas solution. Cells grow slowly if the stiffness of the solution is too high. In such cases, the solution's temperature can be increased to lower the stiffness. Generally, the cell growth process is mainly controlled by the temperature of the system, the time allowed for cells to grow, the viscoelastic properties of the polymer/gas solution, the state of supersaturation and the hydrostatic pressure applied to the polymer matrix.

Shutov and Visco [94] explained some of the foaming methods. The most commonly used method involves blowing agents which include chemical and physical blowing agents. Chemical blowing agents (CBA) liberate gases as a result of chemical reactions or due to thermal decomposition. CBAs represent the most common and widely used blowing agents; they include three types of compounds. The first type includes compounds that liberate gas as a result of a reversible equilibrium thermal decomposition. This reversible nature of the reaction may result in a decrease of the gas content in the

system which may lead to pressure drop in the foams' cell and an eventual shrinkage of the material. Examples include bicarbonates and alkaline-earth metals. The second type consists of compounds that liberate gas as a result of an irreversible thermal decomposition. The liberated gases are mainly  $N_2$ ,  $CO_2$  along with other gases. Examples of these compounds include aliphatic and diazo compounds. The last type includes compounds that liberate gases as a result of chemical interactions of the components. Examples of this kind include the interaction of acids with carbonates. Using CBAs has two advantages: they are easily processed with ordinary equipment and they are easy to introduce into the composition to be foamed. Another noteworthy classification of CBA depends on whether they are exothermic or endothermic during thermal decomposition. Exothermic CBAs dominate the market thanks to the large pressure they can develop during the foaming process. However, the advantages of endothermic CBAs (such as improved cell structure) have recently been publicised.

In physical blowing agents (PBA), gases are liberated as a result of a physical process (desorption, evaporation) at reduced pressures or elevated temperatures [94]. Most PBAs are liquids and they don't undergo chemical transformation during the process. PBAs also include gases that are directly introduced into polymer composition for the purpose of forming cellular structures. Compounds that can be classified as PBA include inert non-toxic gases such as  $CO_2$  and  $N_2$  that have low permeability in most polymers. Also, other compounds include low-boiling liquids where gases are liberated by reducing the pressure and/or increasing the temperature. Examples include aromatic hydrocarbons and alcohols. The other PBA compounds include solids (PBA-sorbents) where saturation of the sorbent

with gas is performed under pressure, gases are then liberated upon heating (thermal desorption). Examples of this kind include activated carbons and silica gels.

When compared to CBA, PBA-based foam processing is more environmentally friendly, has no decomposition temperature requirements, and in general produces better cell morphology [43]. However, PBA-based foaming is more technologically challenging and requires capital investment (system modifications and gas injection systems) [43].

Park [43] reviewed the concept of foamed WPC. Foaming WPC can significantly improve their physical and mechanical properties. Besides reducing the material required, the production of foamed WPC requires lower temperatures and faster speeds which lead to lower production cost. However, more literature about foaming WPC is needed since what is available is very sketchy. One of the major challenges in foaming WPC is the use of wood as a filler. Wood, like many other natural fibers, starts degrading upon heating and releases moisture and other volatiles especially if extrusion is the processing technique. The moisture is retained in the melt in a separate phase (liquid or gaseous) since it has a very low solubility in most plastics [95]. Once the melt exits the extruder, the moisture is vaporized because of the pressure drop while the extrudates' temperature is high. This can lead to a poor cell structure and a blistered surface resulting in a non-uniform cell distribution and a large average cell size. Furthermore, when the resulting foam cools, the water vapor condenses at around 100°C forming a vacuum within the cells of the foam resulting in shrinkage of the extrudate.

Faruk et al. [44] reviewed some of the issues and processing methods involved in microcellular wood fiber reinforced polymers. A major difference between WPC foams and

other polymeric foams is that wood fiber retains its solid phase and is not plasticized during processing. Hence, the dissolution is restricted to the polymer since the gas does not dissolve in the wood fiber [96]. Consequently, the amount of gas that can be dissolved in the mixture, and utilized for homogeneous nucleation, is restricted. Furthermore, interfacial regions such as those between the wood fiber and HDPE are not wetted and may provide channels for fast gas movement [97]. This leads to some undissolved gases to be retained in the micro-voids at these interfacial regions which may lead to a notion of higher solubility due to the increase in the apparent/effective diffusion [96]. In actuality, and instead of being dissolved, some of the blowing gases may remain in a separate phase.

The authors further illustrate that the presence of solid wood fibers, during the foaming process, increases the probability for heterogeneous nucleation to occur at the solid melt interfaces than for homogeneous nucleation [44]. Typically, heterogeneous nucleation occurs either because of the entrapped gas in the micro-voids at the interfacial regions or as a result of an increase in the free energy of the system caused by reduced surface tension at the interface of the fiber and the liquid polymer [96]. As previously mentioned, wood fiber releases volatiles when processed at high temperatures, which may affect the cell nucleation. Some of these volatiles are soluble in the polymer ( $\text{CO}_2$ ), some have low solubility ( $\text{H}_2\text{O}$ ) and the solubility of some volatiles is unknown.

The authors then discuss batch processing which is one of the earliest foaming processes [44]. In this process, a polymer sample is typically placed in a high-pressure chamber where it is saturated, under high pressure and at ambient temperature, with an inert gas (such as  $\text{CO}_2$  or  $\text{N}_2$ ). Then, the pressure is released and the sample is heated to create a

thermodynamic instability (this also rapidly lowers the solubility of the gas in the polymer). This drives the nucleation of a large number of microcells, which grow to produce the foam expansion. Due to the low rate of gas diffusion into the polymer at room temperature, batch foaming requires a very long time for the saturation of the polymer with gas, which is a major drawback for this process. The authors further discuss other processing technologies include extrusion (most widely used and cost effective) and injection molding. The authors argue that despite the promises of microcellular foamed composites, it still remains a poorly understood concept in its infancy.

Matuana et al. investigated the batch foaming of PVC-wood composites [96]. In this study, the relationships between foaming temperature and the cell size and cell density of the composites were established. It was concluded that the cell size increased with the increase of foaming temperature due to the increase of void fraction and cell coalescence. Also cell densities tend to decrease with the increase of foaming temperatures. In particular, the density decreased significantly after a foaming temperature of more than 90 °C. This was attributed to the activated cell coalescence by the lowered melt strength at high temperatures. As for the effect of foaming time, it was observed that the cell density decreases as foaming time increases. This was attributed to the coalescence of cells which occurred during the foaming process.

Rachtanapun et al discussed batch foaming of polyolefin blends-wood composites [97]. It is argued that research work on foaming of semi-crystalline polymers (such as HDPE and PP) is less than that of amorphous ones (such as PVC) due to the difficulties in foaming the former category of polymers. Consequently, there is less research on polyolefin-wood

composites when compared to composites of amorphous polymers and wood. The solubility of gas in the wood fibers and in the crystalline regions of the polymer blend (HDPE-PP) is negligible; therefore, increasing the wood fiber content in the blend reduces the volume of the amorphous portion of the polymer in the blend available for gas solution. This in turn decreases the solubility of gas. Also, it is mentioned that increasing the wood fiber content increased the diffusivity of CO<sub>2</sub> which is attributed to the poor interfacial adhesion between the fiber and the polymer blend. The void fraction of the foamed blends decreased as the wood fiber content was increased. The void fraction is governed by the nucleation of bubbles and their growth which in turn are heavily dependent on diffusivity (the rate of gas loss) and solubility (gas uptake in the material). As mentioned above, increasing the wood content decreased the solubility but increased the diffusivity. This signifies that solubility and diffusivity are not the only variable affecting the void fraction in this case. The viscoelastic properties of the matrix also play a role in determining the void fraction. High stiffness and high melt viscosity (both characteristics of wood fiber) provide high resistance for cell growth [96]. Therefore, increasing wood fiber content into the blend along with high diffusivity and lower gas solubility prevented bubble growth.

The authors conclude that the addition of wood fiber into the blend composite had a detrimental effect on its foamability [97]. For instance, the addition of a mere 5 per-hundred-resin (phr) wood fibers in the blend composites resulted in reduction of average cell size and cell-population density when compared to the pure HDPE-PP blend. The reduction in cell size may be due to the quick diffusion of gas to the environment during

the foaming process. Also, poor interfacial adhesion between the polymer matrix and wood fibers provide channels for the gas to escape quickly and easily to the environment.

## **Chapter 3 Methodology**

This chapter discusses the methodology used in this work. The materials used in this work are cited first. Next, the methodologies followed in preparing the materials are mentioned, these include compounding (by filament extrusion), compression molding and profile extrusion. This is followed by a review of the ASTM standards used for the tensile, flexural and impact tests. Finally, the batch foaming methodology is reviewed. Preparation of the samples, sorption experiments and characterization of foams are all cited.

### ***3-1 Materials***

#### **3-1-1 HDPE**

The plastic used in this study was HDPE (2710, MI=17, by Nova Chemical). A reason why HDPE was chosen is because it is the most widely used thermoplastic and it is relatively inexpensive; also, plastics account for 11.8% of the 246 million tones of municipal solid waste in the United States in 2005 [51, 98]. Only 5.7% of the plastics generated in the US in 2005 were recycled [53]. Also, recycled HDPE pellets and flakes are 31–34% cheaper than virgin HDPE [54]. Another reason why HDPE was chosen is that thermoplastics with low softening temperature and high melt flow index (such as the HDPE used here) are preferred in WPC [32] for higher production rates. So using HDPE as the matrix helps solves recycling/recyclability and processing issues. Figure 3-1 below is an image of the used HDPE pellets



**Figure 3-1 The used HDPE pellets**

### **3-1-2 Wood Fiber**

The wood fiber (WF) used was standard softwood (pine) grade 12020 supplied by American Wood Fibers. Pine is a popular type of wood in North America, this grade was selected due to its fine powdered form. Also, softwoods are longer than hardwood and their structure is less complex, so they are preferred in some applications [31]. The majority of wood waste is destined for landfill. For instance, in 2002 about 63 million metric tonnes of wood waste was generated in the United States alone [9]. The use of waste wood (sawdust) in WPC helps in decreasing these disposal costs. It has been shown that waste wood in the form of wood flour, fibres or pulp is suitable as a filler for polyolefins [55, 56].

### **3-1-3 Coupling Agent**

The coupling agent, used for improving the adhesion between the hydrophobic HDPE and the hydrophilic WF, was the maleic anhydride-g- HDPE (MAH-g-PE, Fusabond MB-100D, 0.8~1.1 wt% of MAH, MI 2.0 g/10min, from DuPont (Canada) and in all compositions it was 3 % in weight, or per hundred resin (phr), of mixed HDPE and WF (unless otherwise stated). This brand was shown to be more effective than other coupling

agents when the composite contains HDPE and wood [36]. Also, it was shown that 3% is the optimum coupling agent amount in the same composite [36].

### **3-1-4 Glass**

The glass-fiber (GF) used was E-glass by Owens Corning Milled Fiber grade 737-BD which is 1/32" in length and 16 micron in diameter. E-glass is the most widely used glass reinforcement in plastics, is preferred for general purposes and has a good cost to performance ratio [62]. Glass is used as a reinforcement due to its superior strength properties.

### **3-1-5 Lubricant**

In profile extrusion, a lubricant was used to reduce the viscosity and slip at wall. The lubricant used was TPW 709 (propriety blend of non metallic internal/external processing aids) provided by Sturktol Canada ltd.

## ***3-2 Preparation of Material***

### **3-2-1 Initial Extrusion**

The materials were compounded using a Leistritz co-rotating intermeshing twin-screw compounding extruder with a 27-mm diameter (model ZSE27) with side stuffer. The HDPE and coupling agent were dry blended together as were the WF and GF (whenever it was used). All the materials were fed through a Brabender Technologies feeder (model no DDW-MD5-FW40/3PLUS-50). The feeding operations were controlled through

Congrav RC 4A operating unit by Brabender. The feeding rate ranged between 2-4 kg/hr. A software, Macromatex Twin Screw Control System (TSCS) by Leistritz, was used to set and control the extrusion processing parameters. The mixture was then extruded, and cooled upon exiting the extruder die and subsequently pelletized in a Scheer Bay Co. pelletizer (model no BT-25). Table 3-1 below shows the processing parameters used in this extrusion.

<b>Parameter</b>	<b>Value</b>	<b>Comments</b>
Zone 1 Temperature	160 °C	Feeding and melting zones
Zone 2 Temperature	160 °C	
Zone 3 Temperature	160 °C	
Zone 4 Temperature	160 °C	
Zone 5 Temperature	160 °C	
Zone 6 Temperature	160 °C	Compression and mixing zones
Zone 7 Temperature	155 °C	
Zone 8 Temperature	150 °C	
Zone 9 Temperature	140 °C	
Zone 10 Temperature	140 °C	Die zones
Zone 11 Temperature	140 °C	
Screw Speed	250 RPM	

**Table 3-1 Processing parameters in initial extrusion**

### ***3-3 Compression molding and sample preparation***

The pellets were compression-molded into sheets using a Carver Hydraulic press. The mold, filled with pellets, was compressed at 160°C under 4.5 metric tons for one minute. The force was then reduced to zero for another minute under the same temperature to purge away volatiles. This procedure was repeated for five minutes and the mold was then compressed under 6.8 metric tons for ten straight minutes. After forming, the sheets were cooled to ambient temperature under 5 p.h.i using a Carver Hydraulic cooler press

and then cut into the desired shape..

### 3-3-1 Profile Extrusion

In the case of profile extrusion, the WPC pellets (80 wt % HDPE, 20 wt % wood, 2.5 wt % glass fibers, 3 wt % coupling agent and 4 wt % lubricant) were fed in a similar fashion to the initial extrusion. The downstream system (ONYXEDS-100C) by Onyx Extrusion Technology Inc. consisted of a calibrator, a cooling water bath, a puller and a cutter.

Table 3-2 below shows the processing parameters used in profile extrusion.

Parameter	Value	Comments
Zone 1 Temperature	135 °C	Feeding and melting zones
Zone 2 Temperature	140 °C	
Zone 3 Temperature	145 °C	
Zone 4 Temperature	150 °C	
Zone 5 Temperature	155 °C	
Zone 6 Temperature	160 °C	Compression and mixing zones
Zone 7 Temperature	155 °C	
Zone 8 Temperature	150 °C	
Zone 9 Temperature	145 °C	
Zone 10 Temperature	140 °C	Die zones
Zone 11 Temperature	135 °C	
Screw Speed	250 RPM	

**Table 3-2 Processing parameters for profile extrusion**

### **3-4 ASTM Methods**

#### **3-4-1 Tensile Tests**

The tensile tests were carried out using a LLOYD Instruments LS100 Universal Testing Machine (General Purpose with Yield Setup) according to ASTM D638, but with the cross-head speed of 1 mm/min. The results were obtained using NEXYGEN MT software and analyzed using the same software and Excel. For each composition five samples were tested and used for interpreting the results. The sheets were cut into dog-bone shaped samples conforming to ASTM D638 type –V with a gauge dimensions of about 14 mm x 2.6 mm x 3.2 mm.

#### **3-4-2 Flexural Tests**

The flexural tests were carried out using a LLOYD Instruments LS100 Universal Testing Machine (General Purpose Three Point Bend Setup) according to ASTM D790 with a preload of 1 N; the samples were tested until the appropriate deflection was achieved. The results were obtained using NEXYGEN MT software and analyzed using the same software and Excel. For each composition five samples were tested and used for interpreting the results. The sheets were cut according to ASTM D790, each sample had roughly the following dimensions: 124 mm x 12 mm x 3.2 mm.

#### **3-4-3 Impact Tests**

The impact tests were carried out on a Gardner heavy duty impact tester (model IG-1142) according to ASTM D5420. The results were analyzed using Excel; nine samples of each

composition were tested and used to interpret the results. The sheets were cut according to ASTM D5420 into 41 mm x 41 mm x 3.2 mm samples.

### ***3-5 Foaming - Batch Processing***

#### **3-5-1 Preparation of Samples**

The pellets were compression-molded into sheets using a Carver Hydraulic press. The mold, filled with pellets, was compressed at 160°C. After forming, the sheets were cooled, under pressure, to ambient temperature and cut into 6.2 x 1.2 x 0.32 cm samples, five samples were used each time.

#### **3-5-2 Sorption Experiments**

The samples were weighed and then placed in a high pressure (800 psi) chamber under ambient temperatures and saturated with an inert gas (CO<sub>2</sub>) for two different durations (1 day and 2 days). These long durations were necessary to allow enough gas to be diffused into the polymer since, around room temperature, the rate of gas diffusion into the composite is low. The CO<sub>2</sub> uptake was measured by weight gain immediately after pressure release. The solubility of the gas is rapidly lowered by releasing the pressure and heating the samples. After the samples were weighed, they were immediately immersed in a hot glycerin bath at three different temperatures levels (155°C, 165°C and 175°C). The samples were heated for three different time period (50, 60 and 70 seconds) and then immediately quenched in a cold water bath.

### **3-5-3 Characterization of foams**

#### **3-5-3-1 Void Fraction (Density)**

The samples' densities were measured by a water displacement technique according to ASTM D-792 and using a Mettler Toledo measurement device. The mass of a sample is measured first in air, then it is measured in distilled water to calculate the density. Density values were used in the results section to calculate parameters such as void fraction and cell density.

#### **3-5-3-2 Morphology**

The morphology of the samples was investigated using a scanning electron microscopy (SEM). The samples were first immersed in liquid nitrogen and fractured to ensure that the microstructure remained intact. Next, the samples were sputter coated by platinum for better viewing under the microscope. The SEM used was JEOL JSM 6060. SEM pictures are used to characterize parameters such as the number of cells/bubbles and to determine whether a cellular structure has been achieved.

## **Chapter 4 Results and Discussion**

The study reported here examines in details the first problem stated in Chapter 1. First, improving the filament extrusion processability of WPC is performed. Then, sample preparation techniques and the effect of the inclusion of small amounts of glass fibers on the tensile, flexural and impact properties are presented and discussed. The effect of the amount of coupling agent on the tensile strength is briefly discussed. Then, an optimum reinforcing combination of wood and glass is chosen to carry on profile extrusion processability studies. Finally, the same combination was batch foamed to investigate the foaming of WPC.

### ***4-1 Improving Processability of WPC: Compounding of WPC pellets***

The study reported here examines in details the first objective stated in Chapter 1. A major source of issues in WPC processing is the fact that it involves two different types of industries. The plastic industry is accustomed to high flowing, high temperature processing conditions, something that the wood/agricultural industry is not really familiar with. Also, shrinkage/swelling in the plastic industry is normally due to temperature; while the biomass in the WPC shrinks and swells due to moisture [32]. This chapter discusses the steps taken to improve the processability of WPC in filament extrusion.

#### **4-1-1 Description of Extrusion System**

The materials (HDPE, wood, coupling agent and were compounded using a Leistritz co-rotating intermeshing twin-screw compounding extruder with a 27-mm diameter (model

ZSE27) with side stuffer. The HDPE and coupling agent were dry blended together since both are pellets of relatively equal size. The same case was between the wood fiber and glass (whenever used). The materials were combined in a per hundred resin basis (phr). All the materials were fed through a Brabender Technologies feeder (model no DDW-MD5-FW40/3PLUS-50). The mixture was then extruded, and cooled upon exiting the filament die and subsequently pelletized in a Scheer Bay Co. pelletizer (model no BT-25). Figure 4-1 below is a picture of the WPC pellets at this stage.



**Figure 4-1 Extruded WPC pellets**

Initially, the wood and glass were fed from the side stuffer in order to have less residence time to minimize any fiber breakage during processing. However, there were several issues in feeding the mixture. The fibers tended to form aggregates and agglomerate at the hopper throat of the stuffer, which prevented them from being conveyed to the screws for further processing. This resulted into a composite that had a composition different than the intended one (more matrix than reinforcement.) The feeding difficulty occurred due to the polar nature of the WF and the resultant tendency to adhere to each other and onto the hopper body instead of sliding through the throat onto the screw. This build-up would ultimately clog the opening and the problem would be exacerbated due to wedge effect. The HDPE and coupling agent were mixed together first, the glass and wood were then mixed and finally all constituents were mixed together and fed from the main feeder.

## 4-1-2 Processing Parameters

The processing parameters play an integral part in determining the quality of the product and ultimately its properties. Table 4-1 below shows the parameter values that were found to be good for compounding.

Parameter	Value	Comments
Zone 1 Temperature	160 °C	Feeding and melting zones
Zone 2 Temperature	160 °C	
Zone 3 Temperature	160 °C	
Zone 4 Temperature	160 °C	
Zone 5 Temperature	160 °C	
Zone 6 Temperature	160 °C	Compression and mixing zones
Zone 7 Temperature	155 °C	
Zone 8 Temperature	150 °C	
Zone 9 Temperature	140 °C	
Zone 10 Temperature	140 °C	Die zones
Zone 11 Temperature	140 °C	
Screw Speed	250 RPM	

**Table 4-1 Processing parameters in initial extrusion**

The eleven heating zones can be broken down, according to their function, into three major sections. Zones 1 through 5 are feeding, melting and conveying zones; zones 6 through 9 are compression and mixing zones; and zones 10 and 11 are for the die system.

Since only one feeder was used here, the first five zones were responsible for allowing the raw materials to move more easily through. The temperatures here (and in other zones) should be high enough for the dominant material (HDPE) to melt since the wood and glass do not melt. Typically, the melting temperature of HDPE is around 140°C, so that was chosen as a minimum temperature initially. An important factor to keep in mind is that wood fibers begin to degrade starting around 180°C which may deteriorate the composite, so the maximum temperature in any zone should be less than that [73]. The temperature in the initial zones was kept at a value of 160°C so as to devolatilize the wood fibers [29]. The volatiles given out in these stages were purged out from the vent/opening for the side stuffer. The next four zones compress and disperse the material by shear mixing.. The devolatilization at the initial high value of 160°C was followed by decreasing temperatures of 155 and 150°C to prevent/reduce further volatile emissions. The last zones are associated with the die and their temperatures should be low so that the outer skin of the compound is solid enough. When temperatures around 150°C and slightly higher were chosen, the melt was too hot upon exiting the die and the surface quality was poor. Values around 130°C were then tried. However, such values were too low and the exiting extrudate swelled causing pelletizing problems. A value of 140°C for both zones was found to be the best. A screw speed of 250 rpm ensured proper mixing, good output rate and low melt temperature at the die.

## ***4-2 Evaluation of Mechanical Properties using Compression***

### ***molding: Effect of Inclusion of Glass Fiber in WPC***

#### **4-2-1 Compression Molding and Sample Preparation**

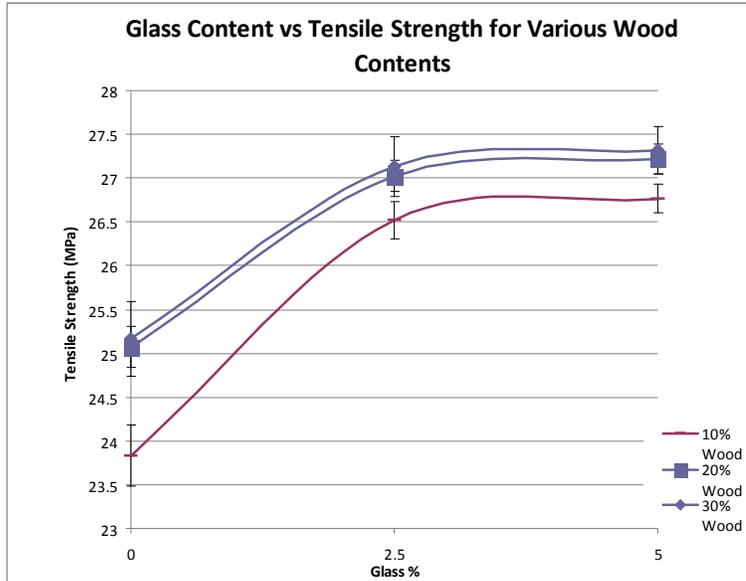
The pellets produced were compression-molded into sheets using a Carver Hydraulic press. The mold, filled with pellets, was compressed at 160°C under 4.5 metric tons for one minute. The force was then reduced to zero for another minute under the same temperature to purge away volatiles. This procedure was repeated for five minutes and the mold was then compressed under 6.8 metric tons for ten straight minutes. After forming, the sheets were cooled to ambient temperature under 5 p.h.i using a Carver Hydraulic cooler press and then cut into the desired shape. Initially, the pellets were compression-molded without being pre-dried. However, the finished plates had bubbles in them (even if the wood flour was dried before extrusion) which rendered them useless for testing since the presence of bubbles leads to wrong results. As the wood fiber content increased, the presence of bubbles increased. This is due to the fact that wood contains moistures and different volatiles that are released under high temperatures. Therefore, the pellets were dried for 48 hours in an 80°C oven, which was sufficient.

#### **4-2-2 Tensile Strength**

The tensile strength,  $\sigma_T$ , is calculated according to:

$$\sigma_T = \frac{F_T}{A_s} \quad (1)$$

Figure 4-2 summarizes the results illustrating the relationship between the amount of glass fiber and the tensile strength of the composite for different wood contents.



**Figure 4-2 Effect of glass on tensile strength of WPC for various wood contents**

Increasing the glass content from zero to 2.5% increases the tensile strength by 11% for 10 % wood; by 7.8% for 20 % and 30 % wood. However on increasing the glass content from 2.5 to 5% the increases are insignificant for all wood concentrations. Generally, it seems that the tensile strength increases initially as the glass content increase to 2.5% and further increasing it to 5% does not have a significant effect. Similarly, increasing the wood content from 10 to 20 % increases the tensile strength by 5.2% in the absence of glass, but this effect is insignificant when GF is used. Increasing the wood content from 20 to 30% did not significantly increase the tensile strength. There appears to be an optimum amount of fiber content (for both glass and wood) for the best tensile strength. Additional increases in fiber contents do not show a significant effect at this stage. Further work investigating issues such as dispersion to determine the reason behind this behaviour is necessary.

### 4-2-2-1 Theoretical Results

Figures 4-3 below shows the theoretical results for the tensile strength of an HDPE-glass composite. Figure 4-4 shows the theoretical and experimental results for the tensile strength of an HDPE-wood composite. Since there are three components here, two different equations were used to determine the theoretical tensile strength values. These values were obtained using the rule of mixture (ROM) principle. First, the tensile strength of the HDPE-glass composite was calculated using the upper bound formula [98]:

$$T_{c1} = T_h V_h + k T_g V_g \quad (2)$$

The tensile strength of the HDPE-wood composite was calculated according to the lower bound formula [98]:

$$T_{c2} = \frac{T_h T_w}{T_w V_h + T_h V_w} \quad (3)$$

Comparing these figures with that of figure 4-2 it appears that the tensile strength of the WPC is affected more by the wood than the glass. In figure 4-4, the theoretical values are slightly higher than the experimental ones, however, at 10% wood, the difference is bigger. Also, the addition of wood appears to slightly increase the tensile strength of the composite as it is increased from 10 to 20%. There is no significant difference in strength between 20 and 30% in experimental or theoretical results.

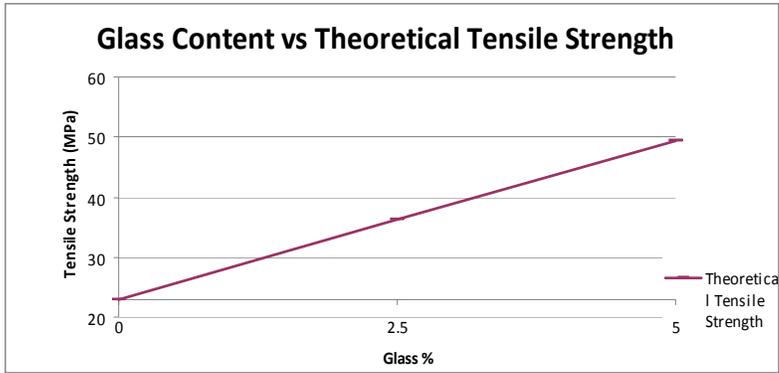


Figure 4-3 Theoretical results for the tensile strength of an HDPE-glass composite

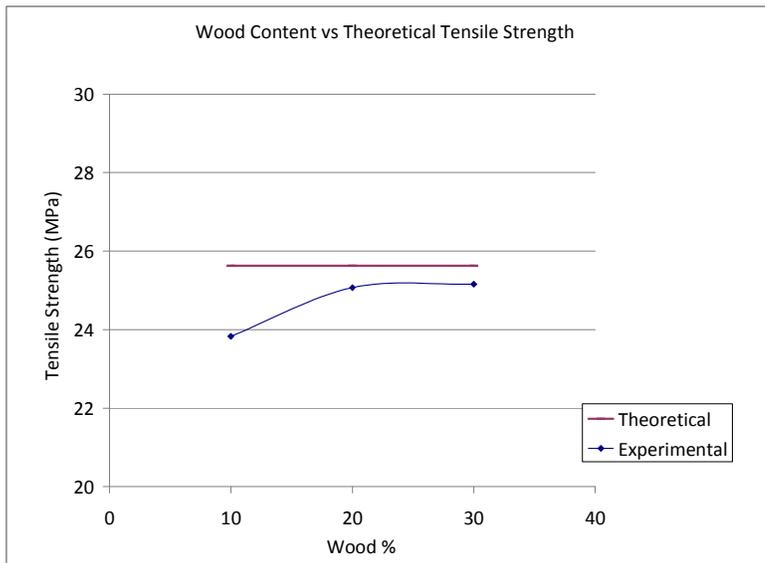


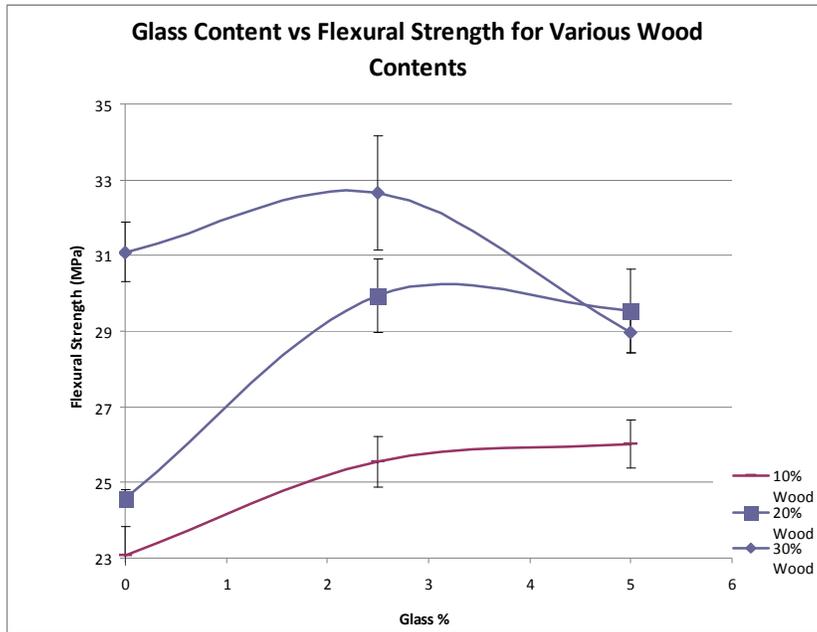
Figure 4-4 Theoretical and experimental results for the tensile strength of an HDPE-wood composite

### 4-2-3 Flexural Strength

The flexural strength,  $\sigma_F$ , was calculated according to:

$$\sigma_F = \frac{3PL}{2bd^2} \quad (4)$$

Figure 4-5 shows the relationship between the amount of glass fiber and the flexural strength of the composite for different wood contents.



**Figure 4-5 Effect of glass on flexural strength of WPC for various wood contents**

The flexural strength follows a somewhat similar trend to the tensile one. Increasing the glass content from zero to 2.5% increases the flexural strength by 11% for 10 % wood; by 22% for 20 % wood and by 5% for 30 % wood. Further increasing the glass content from 2.5 to 5% has no statistically significant effects for 10% and 20% wood but decreases it by 11% for 30 % wood. In general, it seems that the flexural strength increases initially as the glass content increase to 2.5%. Increasing this amount to 5% shows a significant decrease at 30% wood. In a similar fashion, increasing the wood content from 10 to 20 % increases the flexural strength by 6.6% in the absence of glass, by 17% with 2.5% glass and by 13% with 5% glass. Increasing the wood content from 20 to 30% increased the tensile strength by 26.43% in the absence of glass, by 9% with 2.5% glass but decreased it by 2% with 5% glass. Again, there seems to be an optimum amount of fiber content (for both glass and wood) for the best flexural strength. Further increases in fiber contents do

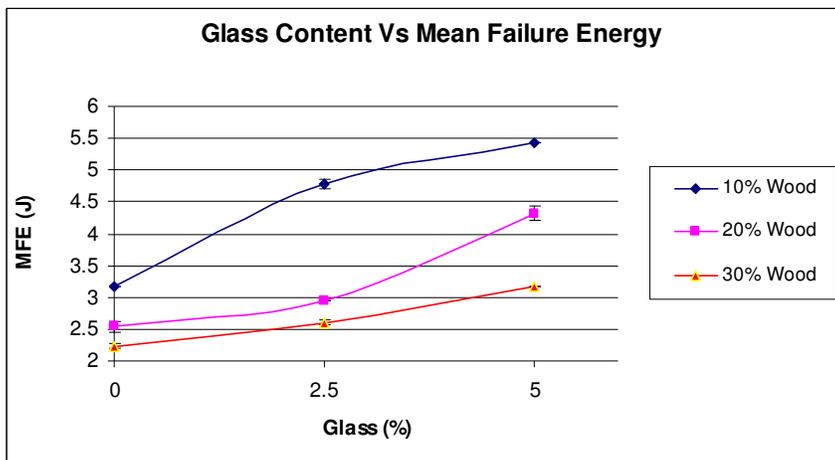
not show a significant effect at this stage and at times decreases the strength. A possible explanation for this is the lack of proper dispersion in high wood and glass contents.

#### 4-2-4 Impact Tests

The impact strength was calculated as the mean failure energy according to:

$$MFE = hwf \quad (5)$$

Figure 4-6 summarizes the results illustrating the relationship between the amount of glass fiber and the mean failure energy (a measure of impact strength) of the composite for different wood contents.



**Figure 4-6 Effect of glass on impact strength of WPC for various wood contents**

Increasing the glass content from zero to 2.5% increases the impact strength by 51% for 10 % wood; by 16% for 20 % wood and by 17% for 30 % wood. Further increasing the glass content from 2.5 to 5% increases the impact strength by 14% for 10 % wood, by 47% for 20 % wood and by 21% for 30 % wood. Generally, it seems that the impact strength increases as the glass content increase from zero to 2.5%, especially at 10% wood. Further increasing the glass content to 5% increases the impact strength, especially at 20% wood. However, increasing the wood content from 10 to 20 % decreases the

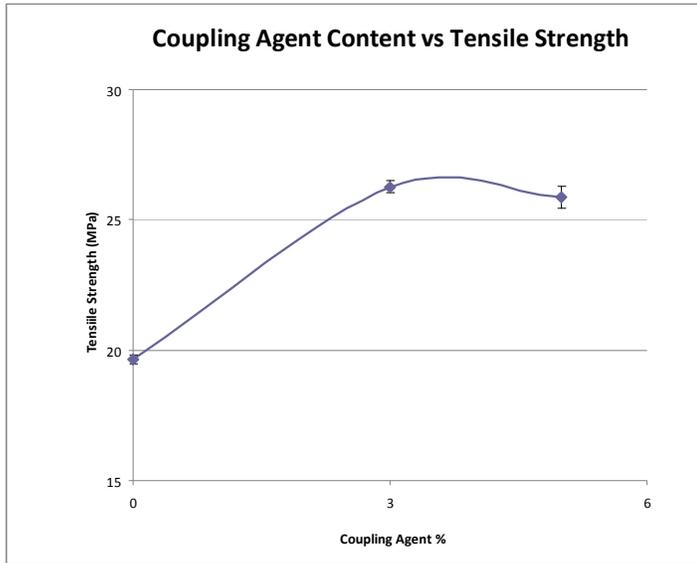
impact strength by 20% in the absence of glass, by 38% with 2.5% glass and by 20% with 5% glass. Increasing the wood content from 20 to 30% decreased the impact strength by 12% in the absence of glass, by 11% with 2.5% glass and by 27% with 5% glass. There appears to be an optimum amount of fiber content (for both glass and wood) for the best impact strength. Further increases in fiber contents do not show a significant effect at this stage. HDPE is less brittle than wood and glass and that is why the impact strength seems to increase as the wood content decreases (which means more HDPE). Although glass is brittle, fibreglass is not as brittle, and is smoother with little flaws which enhances its impact properties.

In general, it seems that increasing the glass content increases the impact strength of the composite. This is because glass has superior mechanical strength when compared to HDPE and wood. Wood does not appear to have a significant effect on the tensile and flexural strength but adversely affects the impact strength, possibly due to weaknesses in the interfacial adhesion between wood and plastic.

#### **4-2-5 Effect of Coupling Agent on Tensile Strength**

The amount of coupling agent was studied to investigate the fact that increasing glass content from 2.5% to 5% does not significantly increase the tensile and flexural strength. The weight percent of coupling agent used in all the studies so far was 3% as previously discussed. This amount was increased to 5% to see if it has a significant effect on the strength. Coupling agents are believed to improve the binding between the matrix and the reinforcement which in turn improves the stress transfer from the matrix to the

reinforcement and vice versa. Tensile testing was chosen because it most tests the binding strength of composites. Figure 4-7 shows the relationship between coupling agent content and tensile strength for a composite containing 20% wood and 2.5% glass reinforcement.



**Figure 4-7 Relationship between coupling agent content and tensile strength for a composite containing 20% wood and 2.5%**

Increasing the coupling agent content from zero to 3% increases the tensile strength by 34%. Further increasing the coupling agent amount to 5% has no significant effect on the tensile strength. It appears that further increasing the amount of coupling agent does not significantly increase the binding between the matrix and the reinforcement and hence will not have a significant effect on the strength properties of WPC.

### ***4-3 Improving Processability of WPC: Profile Extrusion***

Profile extrusion is the most economical process for producing net shaped final products which have constant cross-sections. The production rates are considerably faster than either compression molding, or injection molding. The aim here was to optimize the configuration of the profile extrusion equipment. According to the manufacturers, the TS extruder is designed to produce WPC in a single processing stage, in which the plastic is fed through the main hopper and the WF through the side stuffer. However, the extruded profiles were of very poor quality. In the industry, profile extrusion of WPC typically uses WPC pellets rather than raw material to ensure proper handling and mixing. Moreover two-step processing in which the WPC pellets are compounded before profile extrusion is better as it offers the opportunity of devolatilizing the WF during the compounding stage. This reduces the harmful effects of volatiles during the profile extrusion stage. Therefore, the profile extrusion was carried out in two stages. The materials were first compounded using TSE alone using the temperature profile give in Table 4-1 with higher temperatures in the initial zones to help in devolatilization. The compounded WPC pellets were then fed again into the profile extrusion system for better results. Only one WPC composition was used 80 wt % HDPE, 20 wt % wood, 2.5 wt % glass fibers, 3 wt % coupling agent. This formulation contains the appropriate amount of wood and glass that was found to give the best mechanical properties as discussed in the previous section. Since the ultimate aim of WPC studies is to replace wood in many applications, choosing 20% rather than 10% would ensure more wood-like properties without significantly compromising the strength. Also, a glass content of 2.5% is

sufficient for improved mechanical strength. The temperatures in the zones after the side stuffer opening were kept at low values to keep the volatiles emissions at a minimum. Upon exiting the profile die, the extrudate went through a downstream processing system (ONYXEDS-100C) by Onyx Extrusion Technology Inc. consisting of a cooling water bath to cool and shape the extrudate; a puller to draw the material giving it longitudinal shape; and finally a cutter to cut the extrudate into the desired length.

#### **4-3-1 Use of Lubricants**

Lubricants are used in WPC as processing aids to increase throughput rates, reduce friction between material and equipment, and improve viscosity during extrusion [99, 100]. The main argument for using a lubricant is that it helps in turning the flow of the mixture from a laminar one (which causes the extrudate's outer layers to stick to the wall and the ones in the middle to go through) to a more uniform plug flow [101]. It was sometimes observed that when the extrudate comes out, the outer shell is hard while the inner material is very soft. This can be due to the fact that the outer layers have longer residence time since they were stuck to the walls, which were at lower temperature, and were having a hard time coming out, unlike the flowing middle layers. A non-metallic lubricant was used, and the manufacturer recommended mixing 4 wt%; this amount was added to the material as mentioned in the last section during the initial filament extrusion. The resulting pellets were then used for profile-extrusion. Initially, the pellets were not dried, which caused them to agglomerate and not be processed well. A drying for 48 hours in an 80°C oven was sufficient for the smooth feeding of the pellets.

### 4-3-2 Cooling System

The processing parameters values used in the profile extrusion were initially the same as those used for the initial filament extrusion. However, even when using WPC pellets as raw material, the final product was not adequate. The major problem identified was the insufficient cooling of the extrudate upon exiting the die which results into improper shapes. Figure 4-8 below shows profile obtained at this stage.



**Figure 4-8 Initial profile obtained**

The material is hard from the outside as can be seen from the initial straight part; however, the inside is still very hot and soft and gushes out from any small gap creating the shapes similar to that shown in the figure. This is because the temperature of the die was decreased to cool the extrudate which resulted in a cooled shell but not a cooled inside.

A simple cooling system was then created. The profile die was fitted with tubes using appropriate connections; there were two tubes connected to each of the four sides of the die. One pair of those tubes supplied cold water while the other was used to drain the

cooling water (which would be warmer by then) out of the die. Small shower heads were also used for additional cooling by spraying the extrudate with cold water upon its exit.

Figure 4-9 below shows this cooling system.



**Figure 4-9 Initial cooling system**

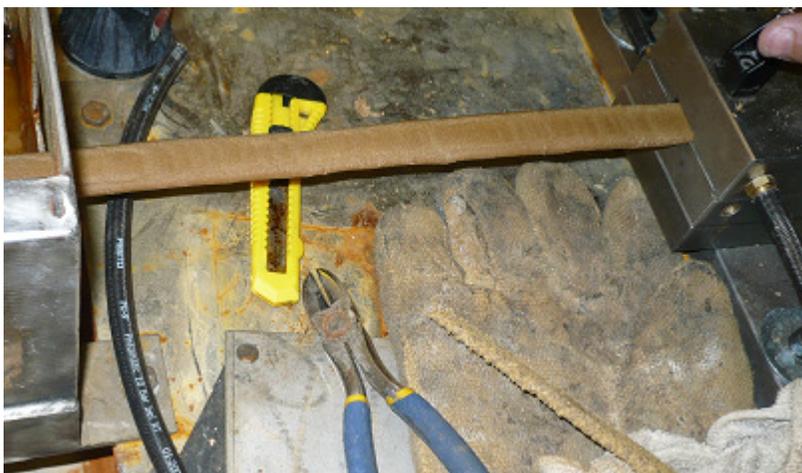
The extrudate improved slightly but not sufficiently. Less material gushed out and the straight part was longer than before; however, the material bent upon entering the water bath and it was hard to pull it and cut it. Such profile is not acceptable since the aim is to have straight and smooth profiles for testing purposes.

It was then decided to use a calibrator system between the die and the water bath to properly cool and shape the extrudate. An order was placed for custom built basic calibrator system containing two sections that shape the extrudate using a vacuum and water pump respectively. A distance of about 15 cm between the profile die and the calibrator was selected based upon recommendation of Onyx. If the distance was too short, the extrudate would go into the calibrator too hot, get stuck inside the calibrator causing build up. A distance that was too long caused the extrudate to be hard upon entering the calibrator and come out in an irregular shape. The water and vacuum pump

provided with the system were not effective. No major changes were initially observed after using the calibrator. Furthermore, neither pumps had the capability to provide sufficient cooling leading to several technical issues. It was also observed that the vacuum pump had the bigger effect. The suction provided by the vacuum pump ensures that the extrudate's outer skin would stick to the walls of the calibrator giving the extrudate a proper shape. Another vacuum pump that is more powerful was used, in the absence of any water cooling, and the extrudate had a good shape. Figure 4-10 below shows the set up of the die and calibration system while figure 4-11 shows a proper profile exiting the calibrator to the water bath.



**Figure 4-10 Set up of calibration system**



**Figure 4-11 Profile exiting calibrator to water bath**

### 4-3-3 Processing Parameters

As mentioned in the previous sections, the processing parameters for profile extrusion were initially identical to those used in the initial filament extrusion. However, the final product did not have as smooth a surface as desired, which can be seen in figure 4-12.



**Figure 4-12 Surface of profile obtained with initial processing parameters**

One of the problems repeatedly faced was the fact that the outer shell of the extrudate was hard and cold but the inner part was hot and soft which caused the hot material to gush out as mentioned earlier. Also, it was noted that the WPC pellets would get stuck in the screws in the first zone because of the high temperature. A way to solve both problems was to decrease the temperature in the affected zones. Moreover, keeping the processing temperature lower also helps in reducing the volatile emissions as previously explained. Therefore, the temperatures were decreased from 160 °C to values starting at 135 °C in the first zone and incrementing by about 5 °C to 155 °C in zone 5. In the middle zones, which are responsible for mixing and compression, the temperatures were slightly decreased by 5 °C starting at 160 °C, at zone 6 which is near the side stuffer vent, and ending at 145 °C.

The temperature at the last die was also slightly decreased to help cool the exiting profile so that it hardens and form into a proper shape. The screw speed was significantly decreased from 250 to 100 rpm. This is because the melt temperature was starting to increase to about 200 °C, which may cause degradation of WPC and a way to decrease the melt temperature is to lower the screw RPM. This increase was attributed to the high viscosity of the WPC pellets and the increased friction between the materials and equipment, even after adding a lubricant. The new processing parameters are listed in table 4-2 below.

<b>Parameter</b>	<b>Value</b>	<b>Comments</b>
Zone 1 Temperature	135 °C	Feeding and melting zones
Zone 2 Temperature	140 °C	
Zone 3 Temperature	145 °C	
Zone 4 Temperature	150 °C	
Zone 5 Temperature	155 °C	
Zone 6 Temperature	160 °C	Compression and mixing zones
Zone 7 Temperature	155 °C	
Zone 8 Temperature	150 °C	
Zone 9 Temperature	145 °C	
Zone 10 Temperature	140 °C	Die zones
Zone 11 Temperature	135 °C	
Screw Speed	100 RPM	

**Table 4-2 Processing parameters for profile extrusion**

The profile obtained with these parameters is shown in figure 4-13, the improvement in surface quality over that of figure 4-12 can be seen.



**Figure 4-13 Profile extruded with new processing parameters**

#### ***4-4 Foaming of WPC***

Foaming of wood plastic composites (WPC) can improve some of the properties of WPC such as reducing their weight and increasing their impact strength. Such improvements have the potential to help WPC find applications in areas where it has been previously overlooked due to factors such as its low strength and high density when compared with natural wood. The foaming process involves many parameters that interact to determine the quality of the final product. Some of these parameters are investigated here to better understand foaming.

##### **4-4-1 Foam Characterization Parameters and their Calculations**

The density of the foamed samples ( $\rho_f$ ) was determined according to (ASTM D792-00)

$$\rho_f = 0.9977 \left( \frac{M_a}{M_w} \right) \quad (6)$$

where 0.9977 is the density of distilled water at room temperature in g/cm<sup>3</sup>. The density of the unfoamed samples,  $\rho_u$ , was determined by:

$$\rho_u = \frac{V_s}{M_s} \quad (7)$$

The void fraction ( $V_f$ ) was determined according to:

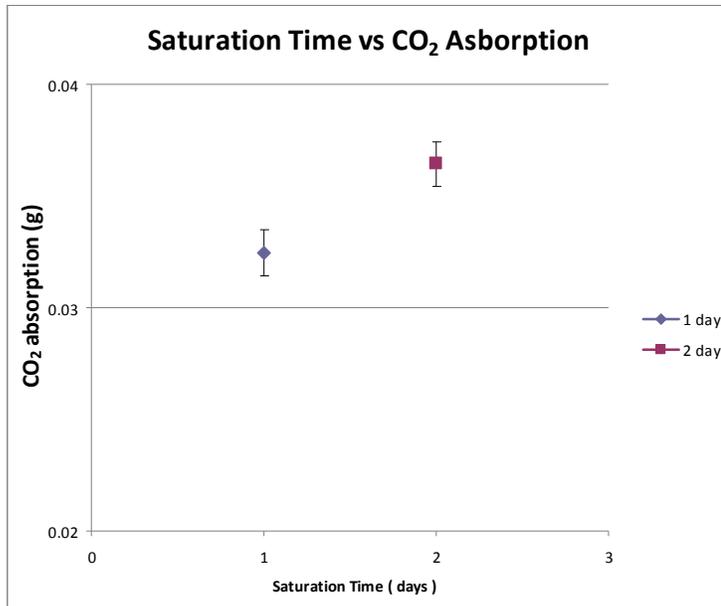
$$V_f = 1 - \left( \frac{\rho_f}{\rho_u} \right) \quad (8)$$

The cell density,  $N_0$ , is calculated as: [102]

$$N_0 = \left[ \frac{nM^2}{A} \right]^{3/2} \left[ \frac{1}{1 - V_f} \right] \quad (9)$$

#### 4-4-2 Effect of Saturation Time on the CO<sub>2</sub> Uptake

Figure 4-14 shows the relationship between the CO<sub>2</sub> uptake and saturation times of 1 and 2 days.

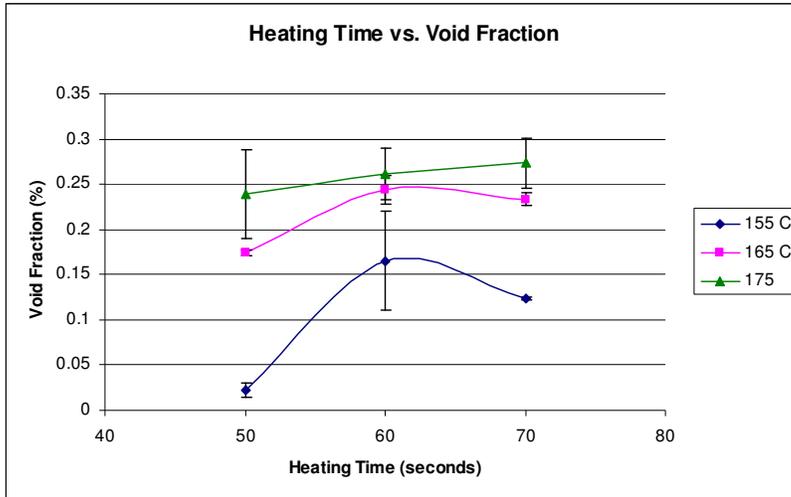


**Figure 4-14 Relationship between the CO<sub>2</sub> uptake and saturation times**

Doubling the time of gas saturation does not seem to have a very significant effect on the CO<sub>2</sub> uptake. The time cost of doubling the period does not merit the slight increase in CO<sub>2</sub> uptake and it seems that a saturation time of one day is enough. After these results, subsequent samples were saturated for one day only.

#### **4-4-3 Effect of Heating Time on the Void Fraction of the Foamed Samples**

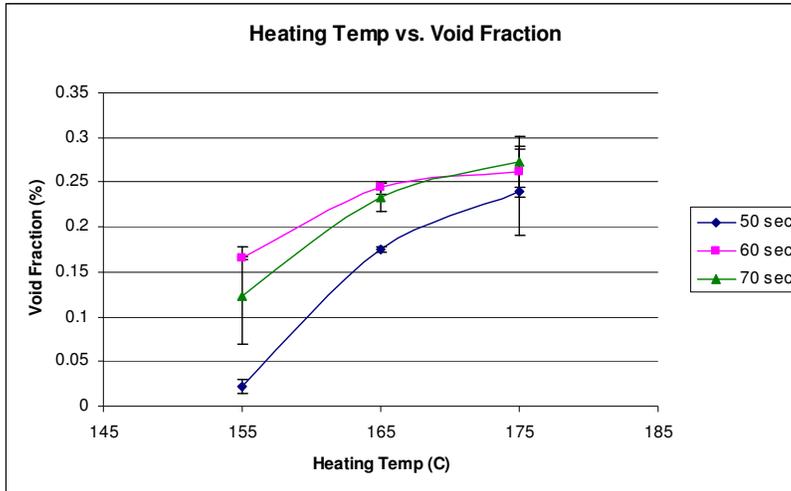
Figure 4-15 shows the relationship between the void fraction and the heating time at three different temperature levels for a saturation time of one day.



**Figure 4-15 Heating time versus void fraction at different heating temperatures**

The void fraction appears to increase as the heating temperature increases. Furthermore, increasing the heating time duration from 50 to 60 seconds increases the void fraction at all temperatures, but increasing the time further does not significantly increase the void fraction. It even has a negative effect on the void fraction when the heating temperature is 155°C. This is due to the fact that at this lower temperature the material is not soft enough to expand fully, and at longer times, the gas diffuses out of the polymer matrix, thereby, lowering the density

Figure 4-16 shows the relationship between the void fraction and the heating temperature at three different time durations for a saturation time of one day.



**Figure 4-16 Heating temperature versus void fraction at different heating time durations**

Increasing the heating temperature seems to increase the void fraction, especially as the temperature increases from 155 to 165°C. Further increasing it to 175°C does not significantly increase the void fraction. Also, increasing the heating time from 60 to 70 seconds does not have a significant effect on the void fraction. It was noted that as the heating temperature increases (especially to 175°C) that the shape of the sample deteriorates. The softening temperature of HDPE (the predominant material in the composite) is about 140°C. The specimen can sustain the higher temperature for short time duration before complete softening occurs, but at durations of 60 and 70 seconds, the sample showed some deterioration.

The highest void fraction (about 27%) was obtained at a heating temperature of 175°C with a heating time of 70 seconds. In general, the void fraction seems to increase as both the heating time and temperature increase but starts to taper off at 175°C and 70 seconds.

#### 4-4-4 Effect of Heating Temperature on Cell Density

Figure 4-17 shows the relationship between the cell population density and the heating temperature at different time durations.

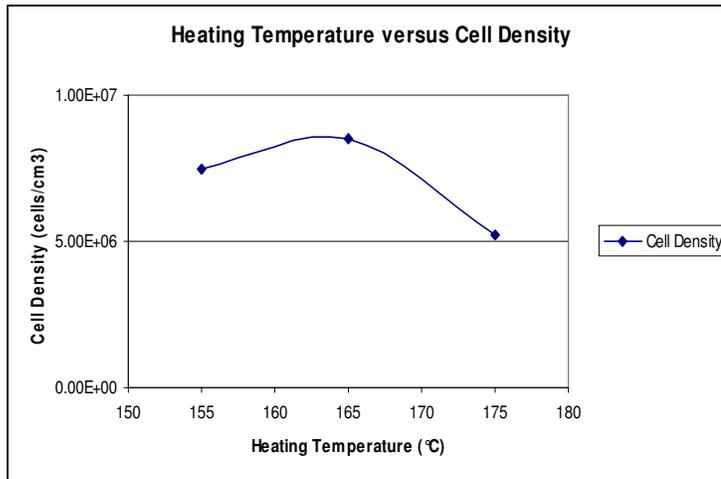
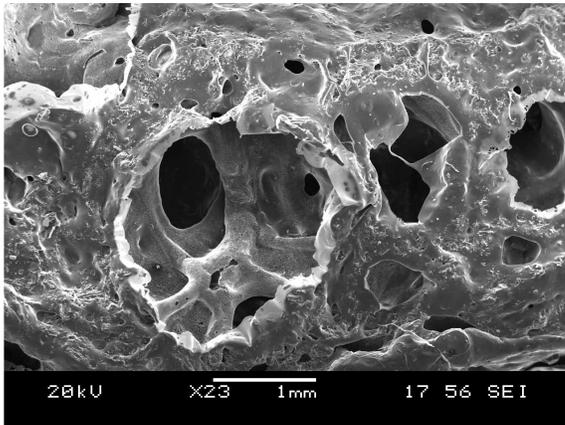


Figure 4-17 Heating Temperature versus Cell Density

Taking the order of magnitude into consideration, it does not appear that the foaming conditions significantly affect the cell density. However, it can be seen that the cell density is highest at the processing temperature of 165°C. This can be explained by the fact that at lower temperatures the polymer is quite stiff and the gas does not generate enough pressure to form all possible cells particularly in the core region which heats up at the last. As a result some gas diffuses out of the specimen on cooling without having contributed to foaming. At the higher temperature the matrix becomes too soft and as a result many of the formed bubbles coalesce with each other giving rise to larger cells and lower cell density [103]. The cell densities obtained were in the order of  $10^6$  cells/cm<sup>3</sup>.

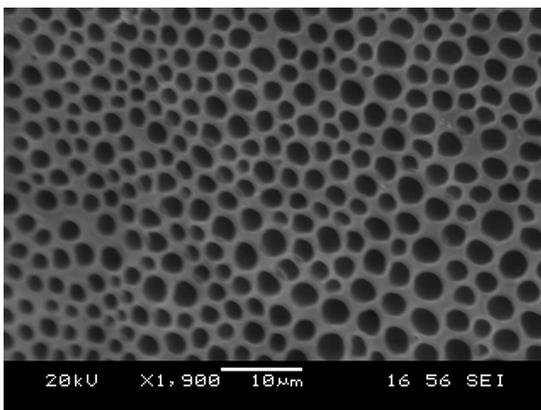
#### 4-4-5 Foam Morphology: SEM Pictures of Foamed Structures

Figure 4-18 shows the typical foam morphology of the foamed specimen. The largest cell size was of the order of 500 micrometer.

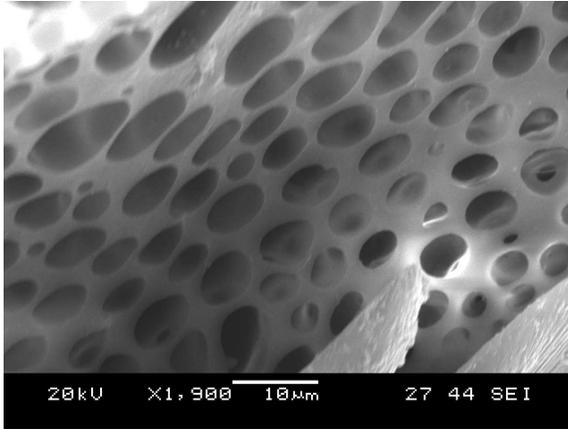


**Figure 4-18 SEM of sample in Figure 4-15 with less magnification**

The best structures, shown below on figures 4-19 and 4-20, were obtained with a saturation time of one day, a heating time of 175°C for 60 seconds and a heating time of 165°C for 70 seconds. But these were within larger cells. Most expansion occurred in the bubbles formed at the interface of the wood and the plastic, but as seen in the SEM, some microcellular foaming occurred within the plastic matrix. But these foam will not affect the properties in the presence of the larger foams



**Figure 4-19 SEM of a sample foamed at 175°C for 60 seconds**



**Figure 4-20 SEM of a sample foamed at 165°C for 70 seconds**

This agrees to some degree with the case of the void fraction where the best void fraction occurred at a one day saturation time with high heating temperatures and times. The structure in 4-19 appears more organized than that of 4-20. In fact, the highest void fraction was achieved at 175°C and 60 seconds, the same conditions as that in figure 4-19. However, such cellular structure occurred only in few locations over the sample. For instance, the structure in figure 4-19 occurred near the edge of the sample. Figure 4-18 is of the same area but with far less magnification, and it shows the presence of large bubbles and the sporadic dispersion of the cellular structure around them.

## Chapter 5 Conclusion and Recommendations

In this work, a number of technology development features for better quality Wood Plastic Composites were studied. Three topics were discussed: the processability of WPC using conventional methods (extrusion in general and profile extrusion in particular); adding low amounts of glass fiber to improve the mechanical properties (tensile, flexural and impact) of WPC; and investigation of the concept of foaming WPC by examining ways to optimize the parameters in batch foaming.

Processing of WPC, either with filament/profile extrusion or with compression molding requires adjustments in the methods used. The processing parameters need to be tailored according to the raw materials used. Extruding the constituent materials for filament extrusion calls for different conditions than extruding the pellets for profile extrusion. The different zone temperatures and the screw speed affect the surface quality and processability of WPC. Further modifications to the system such as cooling systems and inclusion of a lubricant as a processing aid are needed to ensure proper final product. In the case of compression molding, the pellets had to be dried to ensure that the samples were free from bubbles which adversely affect the mechanical properties. The drying stage may be bypassed at compounding but is required before compression molding.

The inclusion of small amounts of glass fiber increases the tensile, flexural and impact properties of WPC to a certain limit. Further increasing the glass content does not significantly increase these properties under the conditions used. The amount of coupling agent was increased to study its affect on the binding strength through the use of tensile

tests. Increasing the coupling agent content from 3 to 5% has no significant effect on the tensile strength. Therefore, it was concluded that increasing the amount of coupling agent at high fiber loading may not significantly increase the strength of WPC. Comparing the theoretical tensile strength calculations with the experimental one, it appears that the properties of WPC at the tested fiber contents are more affected by wood than by glass.

The batch foaming studies conducted show that it is possible to foam WPC with a low glass content. Doubling the time of gas saturation does not seem to have a very significant effect on the CO<sub>2</sub> uptake. In general, the void fraction seems to increase as both the heating time and temperature increase but starts to taper off at 175 °C and 70 seconds. It does not appear that the foaming conditions significantly affect the cell density; the cell density seemed to be highest at the processing temperature of 165°C and cell densities obtained were in the order of 10<sup>6</sup> cells/cm<sup>3</sup>. Morphological studies showed that the best structures were obtained with a saturation time of one day, a heating time of 175°C for 60 seconds and a heating time of 165°C for 70 seconds (these conditions also gave the best void fraction.) However, these good structures were localized and mostly occurred within the plastic matrix. Such foam will not affect the properties in the presence of the larger foams.

### ***5-1 Future Work***

Some of the recommendations for future work include:

- Parametric studies using Batch foaming to identify more factors involved in foaming of WPC

- Identifying the effects of other parameters such as other CA and lubricants for improving the properties GF in WPC
- Inline Extrusion foaming of WPC instead of batch foaming to save time and cost

Profile extrusion of WPC with higher wood content

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