



DESIGNING HIGH WATER RESISTANCE WOOD-
PLASTIC COMPOSITES FROM STEAM EXPLoded
FIBER AND HIGH-DENSITY POLYETHYLENE

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**Laporan Akhir Projek Penyelidikan
Jangka Pendek**

**Designing High Water Resistance Wood-
Plastic Composites from Steam Exploded
Fiber and High-Density Polyethylene**

by

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Prof. Rozman Hj. Din



PEJABAT PENGURUSAN & KREATIVITI PENYELIDIKAN
RESEARCH CREATIVITY AND MANAGEMENT OFFICE [RCMO]

LAPORAN AKHIR PROJEK PENYELIDIKAN JANGKA PENDEK
FINAL REPORT OF SHORT TERM RESEARCH PROJECTS

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Composites of high-density polyethylene and acetylated steam-exploded *Acacia mangium* fibers (AAEF) were prepared using a heated 2 roll-mill, compression molded and tested for tensile properties. AAEF did not adhere well with HDPE and had resulted in a decrease in the tensile properties of the composites. Addition of maleated polyethylene, did not help in improving the tensile strength particularly for composites filled with AAEF of higher weight percent gain (WPG). However, a significant improvement in the property was observed

when AEF was used instead of AAEF in the compatibilized composite systems. HDPE-AAEF composites were immersed in water at room temperature for 30 days and tested for tensile properties after 2, 4, 6 and 30 days of immersion. All composites absorbed water but the water uptake decreased with increasing weight percent gain (WPG) of AAEF. Immersion time had negative effect on the tensile properties and the impact was lesser for composites filled with AAEF of higher WPG. For compatibilized HDPE-AAEF composites (filler content at 20 wt.%), the best is HDPE-AAEF 4.9 WPG-MAPE composite. The composite showed the lowest uptake of water and the highest tensile properties after 30 days of water immersion.

Komposit polietilina berketumpatan tinggi (HDPE) terisi 'acetylated' gentian kayu Acacia mangium terledak stim terekstrak alkali (AAEF) disediakan menggunakan penggiling bergulung dua panas. Komposit dimampatkan menggunakan pengacuanan mampatan dan diuji bagi sifat-sifat tensil. AAEF tidak melekat dengan sempurna dengan HDPE. Ini menyumbang kepada penurunan pada sifat-sifat tensil. Penambahan 'maleated polyethylene, tidak membantu meningkatkan sifat kekuatan terutamanya kepada komposit yang terisi AAEF pada 'weight percent gain' (WPG) yang tinggi. Walau bagaimanapun peningkatan yang ketara sifat-sifat tensil diperolehi apabila AEF digunakan didalam sistem komposit terserasi berbanding AAEF. Komposit HDPE-AAEF direndam dalam air pada suhu bilik selama 30 hari dan diuji bagi sifat-sifat tensil selepas 2, 4, 6 dan 30 hari rendaman. Semua komposit menyerap air tetapi kandungan serapan air berkurang dengan peningkatan WPG bagi AAEF. Tempoh rendaman memberi kesan negatif kepada sifat-sifat tensil. Kesan ini lebih nyata bagi komposit terisi AAEF pada WPG yang lebih tinggi. Untuk sistem komposit terserasi (kandungan pengisi 20 wt.%), komposit HDPE-AAEF 4.9 WPG-MAPE adalah yang terbaik kerana ia mempamerkan keserapan air yang terendah dan sifat-sifat tensil yang paling tinggi selepas 30 hari rendaman.

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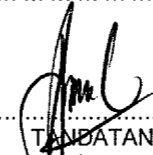
<u>Bahasa Malaysia</u>	<u>Bahasa Inggeris</u>
gentian kayu terledak stim	steam-exploded wood fiber
komposit polietilena	polyethylene composite
penserasi t	compatibilizer
serapan air	water absorption
sifat-sifat tensil	tensile properties

- 5) **Output Dan Faedah Projek**
Output and Benefits of Project

(a) * Penerbitan (termasuk laporan/kertas seminar)
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(Kindly state each type, title, author/editor, publication year and journal/s containing publication)

1. R. M. Taib, Z. A. M. Ishak, H. D. Rozman, and W. G. Glasser (2005). **Characterization of acetylated steam-exploded Acacia mangium wood fiber** .14th Scientific Conference and 15th annual General Meeting of the Electron Microscopy Society of Malaysia (EMSM). 5 – 7 Dis, Penang, Malaysia.
2. R. Mat Taib, S. Ramarad, Z. A. Mohd Ishak, and H. D. Rozman. **Effects of immersion time in water on the tensile properties of acetylated steam-exploded Acacia-mangium fibers filled polyethylene composites**. In

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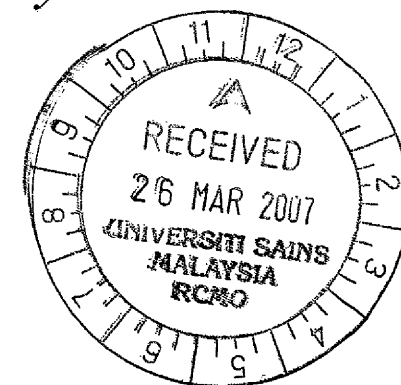
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Universiti Sains Malaysia

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FINAL RESEARCH REPORT FOR USM SHORT TERM GRANT

*Designing High Water Resistance Wood-
Plastic Composites from Steam-Exploded
Fiber and High-Density Polyethylene*

prepared by



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Engineering

Mac 2007

1.0 INTRODUCTION

Wood-plastic composites (WPCs) are candidates for use in building products that have limited structural requirements like decking, fencing, industrial flooring, landscape timbers, and railings because of low maintenance and high durability [1]. It is estimated that building products account for 70% of the total end market used of WPCs [2]. For such applications, WPCs are subjected to various environments like moisture, oil, temperature, and radiation during their service life. In order to utilize the full potential of WPCs, their behavior under these environments has to be determined. In particular, the effect of water must always be taken into account because moisture is always present due to the humidity of the atmosphere and the wood fiber itself is highly hygroscopic. Wood fiber is hygroscopic because the cell wall polymers, i. e., cellulose, hemicelluloses, and lignin, contain many hydroxyl groups as well as other oxygen containing groups such as esters, carbonyl and carboxyl, that attract water molecules through hydrogen bonding [3]. Water absorption in WPCs can lead to a build-up of moisture in the fiber cell wall that would result in fiber swelling and adversely affect dimensional stability [4]. Fiber swelling could also cause the matrix polymer to crack leading to more penetration of water molecules within the composite structures [5]. In addition, moisture build-up can also occur at the fiber-matrix interphase region [4]. This would degrade the interphase and results in inferior composite properties. The hygroscopic nature of wood fiber can be minimized by modifying the basic chemistry of the fiber cell wall polymers [6]. This is achieved by reacting some of the hydroxyl groups with chemicals such as acetic anhydride (acetylation) [6]. Another possible approach is to remove to some extent the cell wall polymers that are responsible for the property. Hemicelluloses and lignin are

hygroscopic and upon removal of these two components, an improved resistance to water absorption has been observed [7-9]. Techniques that can be used for this purpose include alkali treatment and steam explosion coupled with fiber fractionation. In addition, the absorption of water by WPCs can also be minimized by ensuring good wetting of the wood fiber by the hydrophobic matrix as well as sufficient fiber-matrix interaction [4]. Both of which can be achieved with the use of appropriate coupling agent [4] such as maleated polypropylene (MAPP) in wood fiber/polypropylene composite system. Although, many works have been reported on the detrimental effects of water absorption on dimensional stability and mechanical properties of WPCs [10-15], limited information is available on the change in composite properties at different length of water exposure. Such information is important particularly when designing WPCs specifically for applications that involve continuous exposure to high humidity environment.

In this research, an attempt was made to produce wood-plastic composites of high resistance to water absorptions. Both fiber pre-treatments via steam-explosion followed with fiber fractionation and acetylation were tried. In order to promote fiber-matrix interaction, a compatibilizer, maleated polypropylene, was used. The composites were immersed in water for 30 days and tested for tensile properties after 2, 4, 6 and 30 days of water absorption to characterize degradation in the properties. In order to aid in explaining the tensile properties results, fracture surfaces of the composites were analyzed using SEM.

2.0 LITERATURE REVIEW

2.1 Lignocellulosics as fillers and reinforcements for thermoplastics

The use of lignocellulosics as fillers and reinforcements in thermoplastics, notably polyolefins, has become more accepted by the plastic industry over the past few years. In the automotive industry for example, lignocellulosic/thermoplastic composites have been considered for most of the interior components such as head, door, and trunk liners. Such composites too have been considered and marketed as building materials like decking, fencing and flooring. The use of lignocellulosics as fillers and reinforcement in thermoplastic can reduce the product cost as the materials are cheap as well as promote degradation of the composite products after use. This is crucial as now more people are concerned on the impact of materials on the environment [16].

2.1.1 Lignocellulosic sources

Any substance that contains both cellulose and lignin is a lignocellulosic [6]. Lignocellulosics include wood; agricultural crops, like jute or kenaf; agricultural residues, such as bagasse or corn stalk; grasses; and other plant substances [6]. In many countries, wood is favored as a source of lignocellulosic fillers and fibers. For countries deficient in forests, agricultural crops (non-wood) have been favored [17]. Fibers from agricultural crops are classified into several categories depending on the part of the plant from which they are taken. This is clear from Figure 2.1 [18]. Other large sources of lignocellulosic fillers and fibers can come from recycling fiber-based products, such as paper and paper board; waste wood, such as demolition wood and old wooden pallets; and point source agricultural residues, such as rice hulls from a rice processing plant, sun

flower seed hulls from an oil processing units and bagasses from a sugar mill [19]. The two basic techniques to produce fillers are grinding and sieving. Fillers from lignocellulosics are differentiated by species and particle size distributions. Fibers, although are difficult to extract can lead to superior composite properties and act more as a reinforcement than as a filler [20]. Fibers from lignocellulosics can be extracted via several techniques as listed in Table 2.1. Another promising technique that can be used to extract fibers is steam explosion.

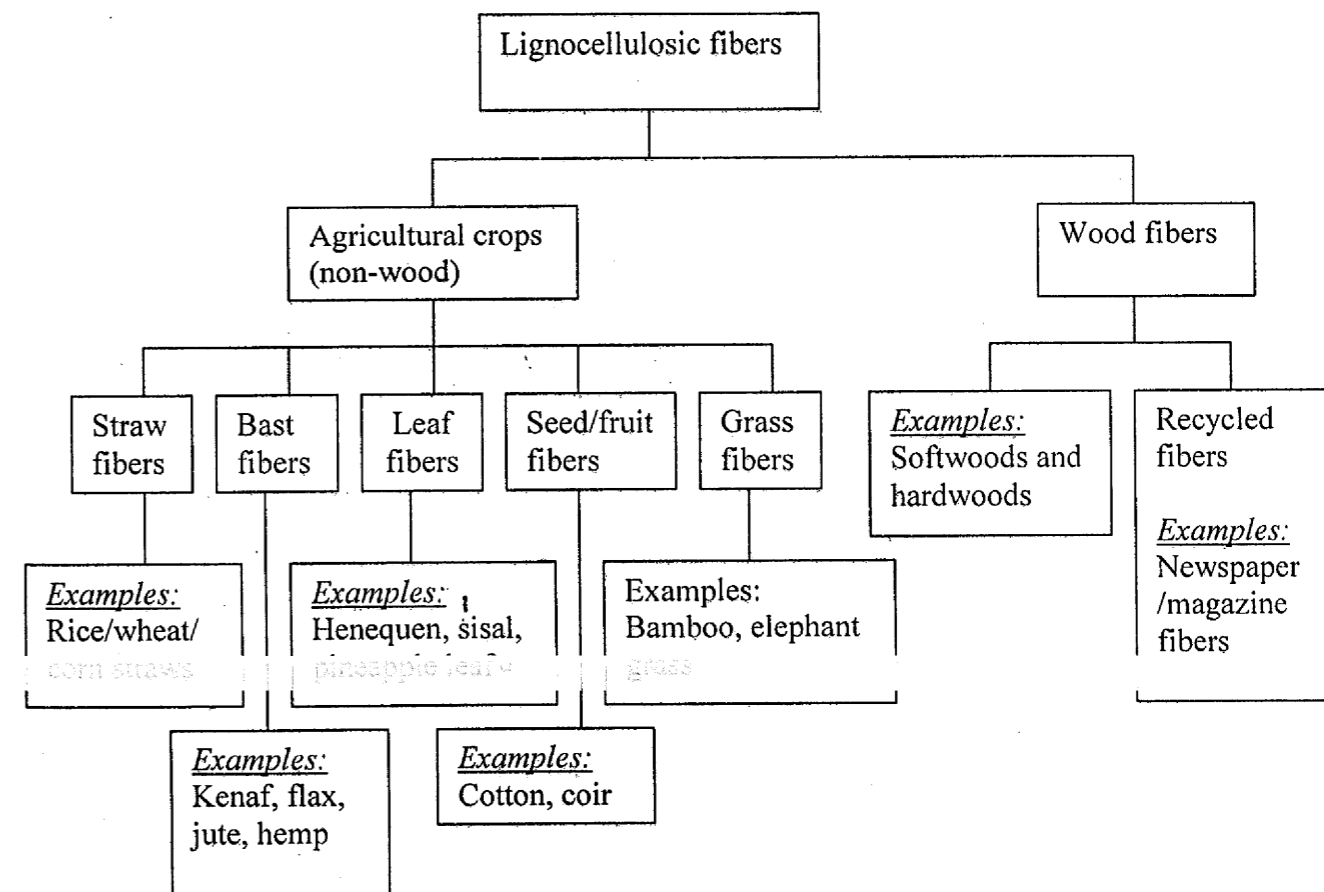


Figure 2.1 Schematic representation of lignocellulosics classification [18].

Table 2.1 Extraction methods of various lignocellulosic fibers [21].

Fiber	Method	Amount
Banana	Manual / raspador	1.5 wt% of stem
Coir	Retting / mechanical	8% of nut (this weighs 1.1 kg)
Jute	Retting and beating / chemical	3 – 4% of stem
Linseed	Retting / dry scratching	20 – 25% of dry straw
Mesta	Retting and beating / chemical	same as jute
Palmyrah	By hand (by beating)	0.5 kg per stalk
Pineapple	By hand / decorticator	2.5 – 3.5% of green leaves
Ramie	Decorticator	2.5 – 3.5% of bark
Sisal	Manual (beating) / microbial retting / decorticator	3 – 4% of green leaves

2.1.2 Chemical compositions, microstructure and mechanical properties

Lignocellulosics have three primary chemical constituents, cellulose, hemicelluloses, and lignin. Other minor include pectin, waxes, and water-soluble substances [22]. Table 2.2 lists the chemical composition of several lignocellulosics [23]. The chemical composition varies from plant to plant and within different parts of the same plant. It also varies within plants from different geographic locations, ages, climate, and soil conditions [24]. Information on the chemical composition of lignocellulosics is important because it determines the fillers' and fibers' properties and thus their applications [23]. For example those with higher cellulose contents are likely to exhibit greater tensile strength and this may result in better composite performance [16].

Table 2.2 Chemical compositions of several lignocellulosics (% of dry matter) [23].

Fiber	Cellulose	Hemicelluloses	Lignin	Pectin
Flax	81	14	3	2
Jute	72	13	13	< 1
Hemp	74	18	4	1
Sisal	73	13	11	1
Ramie	76	15	1	2
Coir	43	< 1	45	4
Straw	40	28	17	8
Wood	45	23	27	

A single lignocellulosic fiber consists of several cell walls. These cell walls are formed out of crystalline microfibrils based on cellulose, which are embedded in hemicellulose-lignin matrix [22]. The cell walls are divided into two sections: the primary wall, containing a loose irregular network of cellulose microfibrils and the secondary wall. In the secondary wall, the microfibrils are closely packed. The secondary wall is composed of three separate and distinct layers, S₁ (outer layer), S₂ (middle layer), and S₃ (inner layer). S₂ layer is the thickest and the most important in determining mechanical properties [25]. These fiber cell walls differ in their composition, i.e., ratio between cellulose and lignin/hemicellulose and in the orientation or spiral angle of the cellulose microfibrils [22]. The spiral angle is the angle that the helical spirals of cellulose microfibrils formed with the fiber axis [26]. Schematic representation of the fine structure of a lignocellulosic fiber is presented in Figure 2.2 [27].

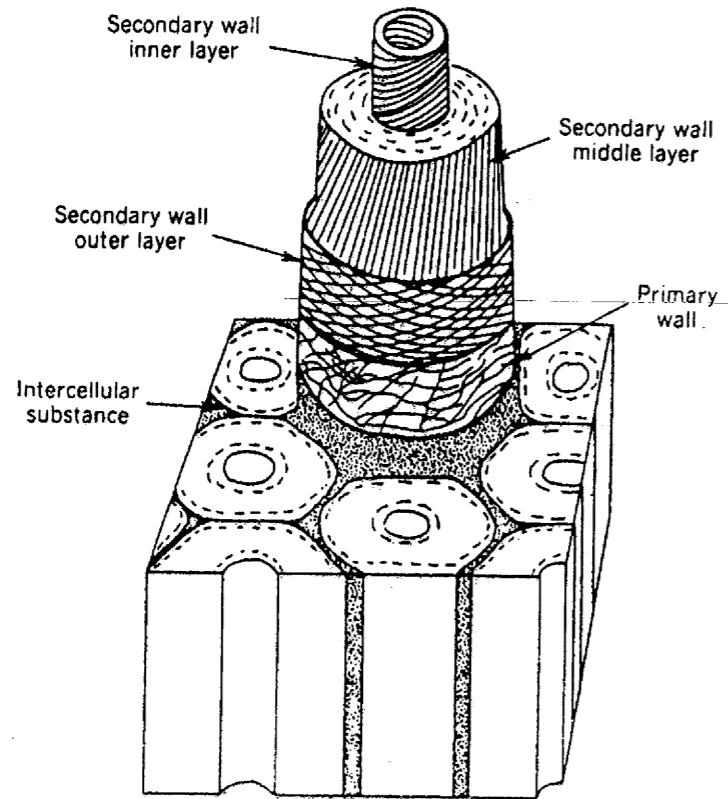


Figure 2.2 Schematic representation of the fine structure of a lignocellulosic fiber [27].

Mechanical properties of lignocellulosic fibers are strongly influenced by chemical composition and internal fiber structure [28]. The most efficient lignocellulosic fibers are those that have high cellulose content coupled with a low microfibril angle or spiral angle [16]. Figure 2.3 shows the relationship between the strength of lignocellulosic fibers and their cellulose contents and microfibril angles [27]. Other factors that affect mechanical properties of lignocellulosic fibers include degree of cellulose polymerization, crystallinity and orientation [29], fiber source, maturity, extraction techniques and the history of the fiber [30]. Agro-based fibers perform better

than wood-based fibers as reinforcing materials for thermoplastics. This is particularly due to their higher aspect ratio as well as mechanical properties [23].

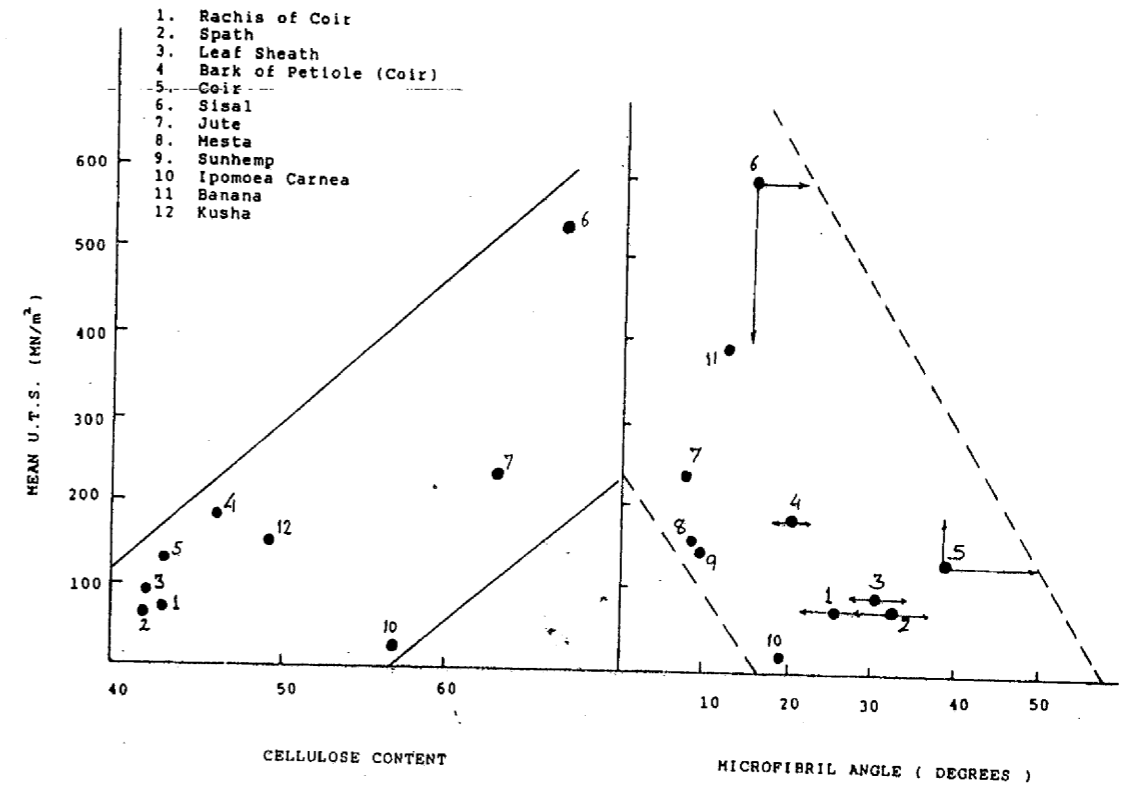


Figure 2.3 Relationship between the strength of lignocellulosic fibers and their cellulose contents and microfibril angles [27].

2.2 Advantages and disadvantages of lignocellulosic fillers/fibers

Lignocellulosic fillers/fibers compared to their inorganic counterparts such as talc and glass fiber, offer many advantages that make them more attractive as fillers and reinforcements for thermoplastics. Such advantages include lower density, lower volumetric cost, and higher specific properties particularly strength and stiffness. Their

non-abrasive nature permits high levels of filling resulting in composite with high stiffness properties without the usual machine wear problems associated with glass fiber and ceramic [16]. Material cost savings due to incorporation of the relatively low cost fillers/fibers and higher filling levels possible, coupled with the advantage of being non-abrasive to the mixing and molding equipment are benefits that are not likely to be ignored by the plastics industry for use in the automotive, building, appliance and other applications [16]. Lignocellulosic fillers/fibers are also non-toxic, easy to handle, and present no health problems like glass fiber that can cause skin irritations during processing [31] and respiratory diseases when the fibrous dust is inhaled [32]. Moreover, as reinforcement, lignocellulosic fibers can stiffen and strengthen the thermoplastic and can improve thermal stability compared with that of unfilled material. There are also some environmental and socioeconomic advantages that cannot be ignored. These include biodegradability, the wide variety of fibers available around the world, the generation of rural jobs, generation of nonfood agricultural/farm-based economy, low energy consumption, and low energy utilization [16]. Lignocellulosic/thermoplastic composites can be processed by traditional plastic processing technologies such as extrusion, injection-molding, thermoforming, pressing, and calendaring [33]. This enables mass production of lignocellulosic/thermoplastic based products. Lignocellulosic/thermoplastic composites also may eventually be recycled or burned to recover heat, without production of residue or toxic by-products [34].

Despite of the advantages mentioned above, lignocellulosic fillers/fibers are used only to a limited extent in industrial practice. The fillers/fibers exhibit some undesirable characteristics which result in inferior composite properties. Among the undesirable

characteristics include: 1) thermal instability at temperatures above 200° C [35], 2) high moisture absorption that can affect both dimensional stability and mechanical properties when the composites are exposed continuously to high humidity environments [16], 3) their hydrophilic nature that responsible for insufficient filler/fiber dispersion, poor fiber/matrix compatibility as well as interfacial adhesion [36], and 4) biodegradability, which limits the service live of lignocellulosic/thermoplastic composites particularly for outdoor applications. [37]

2.3 Modification of lignocellulosic filler/fiber properties

Lignocellulosic fillers/fibers are biopolymer composites made up primarily of cellulose, hemicelluloses, and lignin [6, 37]. These polymers make up the fiber cell wall and are responsible for most of physical, and chemical properties exhibited by lignocellulosic fillers/fibers [6, 37]. The performance of any lignocellulosic/thermoplastic composites is restricted by several undesirable properties of the filler/fiber itself. These undesirable properties can be minimized by changing the basic chemistry of the cell wall polymers [38]. This may lead to the production of high performance lignocellulosic/thermoplastic composites.

2.3.1 Thermal stability

Thermodegradation of lignocellulosics begins at a round 200°C [39]. Thus the processing temperatures of lignocellulosic/thermoplastic composites need to be ensured below 200°C to avoid the possibility of the fillers/fibers to degrade and/or to emit volatile products that could affect the composite properties [16]. This limits the use of

lignocellulosic fillers/fibers to lower melting polymers, such as polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), and polystyrene (PS) [16]. The cell wall polymers that responsible for the poor thermal stability of lignocellulosic fillers/fibers are hemicelluloses and lignin [40]. These polymers particularly hemicelluloses can be modified with several chemicals to enhance the thermal stability of lignocellulosic fillers/fibers. Such chemicals include acetic anhydride [41], benzyl methacrylate [42] and 2,4-dinitrochlorobenzene [43]. The improved thermal stability is attributed to the greater stability of the groups attached to the fiber cell wall polymers [41].

2.3.2 Moisture absorption

Lignocellulosic fillers/fibers are hydrophilic in nature and absorb moisture readily. This is because the cell wall polymers contain hydroxyl and other oxygen-containing groups (esters, carboxyl and carbonyl) that attract moisture through hydrogen bonding [6, 37]. The hemicelluloses are mainly responsible for moisture absorption, but the accessible cellulose, non-crystalline cellulose, lignin, and surface of crystalline cellulose also play major roles. Moisture absorption can result in swelling of lignocellulosic fillers and fibers [16]. This will lead to dimensional instability of lignocellulosic/thermoplastic composites when exposed continuously to high humidity environments. The composites may also suffer loss of mechanical properties as a result of moisture absorption. This is due to the deterioration of filler/fiber mechanical properties as well as the degradation of fiber/matrix interaction. The fiber modulus can decrease by nearly two-thirds or even more due to moisture absorption [44]. The tensile strength, on the other hand, is less affected. The poor fiber mechanical properties under wet

conditions are probably due to the softening of fiber cell wall polymers particularly the disordered zones of the cellulose microfibrils [44].

The extent of moisture absorption by lignocellulosic/thermoplastic composites is largely dependent on the availability of free hydroxyl groups on the surface of lignocellulosic fillers/fibers [45]. In order to reduce the amount of moisture absorbed, the number of these free hydroxyl groups need to be reduced. This can be achieved by replacing some of the hydroxyl groups with bonded chemical groups. Examples of chemicals that can be used include acetic anhydride, maleic anhydride, formaldehyde, propylene oxide, and butylenes oxide [37]. Moisture absorption of lignocellulosic fillers/fibers also can be reduced with good interfacial bonding between the filler/fiber and the polymer matrix. The improved interfacial bonding restricts free movement of water molecules within the composite structures [45]. This reduces the chances of the hydroxyl groups to come in contact with the water molecules. Chemicals that can be used to enhance fiber/matrix interfacial bonding include various types of coupling and compatibilizing agents such as maleic anhydride-grafted polypropylene (MAPP) and poly(methylene (polyphenyl isocyanate)) (PMPPIC). In addition to the improvement of interfacial adhesion, these chemicals also impart some hydrophobicity on lignocellulosic fillers/fibers. This reduces the hydrophilic nature of lignocellulosic fillers/fibers leading to better moisture resistance [46]. Prolonged exposure to moisture, however, can deteriorate fiber/matrix interaction leading to poor composite performance. The trapped water may act as a plasticizer and interrupts the van der Waal's bonds between the treated fiber and the matrix polymer at the interface. This leads to degradation of the fiber/matrix interaction and a decrease in composite properties [15].

The degradation of fiber/matrix interaction could only be avoided through the development of covalent bonds between fiber and matrix. This can be achieved with chemicals such as triazine-based coupling agents [44].

2.3.3 Biodegradability

Lignocellulosic fillers/fibers are degraded biologically because organisms such as fungi, bacteria, insects, and termites, recognize the carbohydrate polymers (mainly the hemicelluloses) in the cell wall and have specific enzyme systems to hydrolyze the polymers to digestible units [6, 37]. Biodegradation of the high molecular weight cellulose weakens the fiber cell wall because crystalline cellulose is primarily responsible for the strength of the cell wall. Strength is lost as the cellulose polymer undergoes degradation through oxidation, hydrolysis, and dehydration reactions [6, 37]. This results in a decrease in the filler/fiber properties and their life span leading to poor performance of lignocellulosic/thermoplastic composites. In order to produce composites with a long service life particularly for outdoor applications, it is necessary to interfere with the biodegradation process for as long as possible [37]. Biological resistance of lignocellulosic fillers/fibers can be improved by bonding chemicals to the cell wall polymers [37]. Among chemicals that can be used to enhance biological resistance include acetic anhydride, propylene oxide, and butylenes oxide. The mechanisms for protection of lignocellulosic fillers/fibers against microbiological degradation through chemical modification, is believed to be due to: 1) the lowering of the equilibrium moisture content below that needed for organisms attack [37], 2) the changes in the conformation and configuration requirements for the enzyme-substrate reactions [37],

and 3) the blocking of the hydroxyl groups of the cell wall polymers in such a way that the enzymes of the degrading organisms cannot recognize them as attackable substrates [47].

2.4 Improvement of filler/fiber–matrix interaction

Lignocellulosic fillers/fibers are polar and hydrophilic, whereas thermoplastic polymers such as polypropylene (PP) and polyethylene (PE) are nonpolar and hydrophobic. Because of this difference in polarity, lignocellulosic fillers/fibers are not chemically compatible with thermoplastic polymers. This contributes to difficulty in dispersing lignocellulosic filler/fiber in the thermoplastic melt and a poor interfacial adhesion that can lead to inferior composite properties [48]. Various chemicals have been tried to enhance the interfacial adhesion between lignocellulosic fillers/fibers and thermoplastic matrices. These chemicals are normally called coupling agents. Examples of coupling agents include isocyanates, anhydrides, silanes, and chlorotriazines.

For polypropylene-based composites, maleated polypropylene (MAPP), an hydride-modified copolymer, has been claimed as the most effective coupling agent [49]. There various types of MAPP depending on acid number and molecular weight. The effectiveness of MAPP in increasing the strength of lignocellulosic fiber/PP composites may be explained by enhanced interfacial adhesion (via the formation of an adhesive bridge) between the fiber and the matrix [50]. The role of MAPP in improving the interfacial adhesion can be explained in the following way. MAPP has anhydride carbonyl groups, which are susceptible to react with the hydroxyl groups of lignocellulosic fibers. The reaction between these groups leads to the formation of ester

bonds. In addition to the ester bonds, MAPP can also form hydrogen bonds with the hydroxyl groups. The long PP chains of MAPP can diffuse and form entanglements with the PP matrix that leads to the formation of an adhesive bridge between lignocellulosic fibers and PP. The formation of such bridge facilitates stress transfer from the polymer matrix to the fiber resulting in enhanced composite properties. In addition to enhanced fiber/matrix interfacial adhesion, MAPP may also improve the mechanical properties of lignocellulosic fiber/PP composites through compatibilization effect [36]. The compatibilization effect is achieved by reducing the total lignocellulosic fiber surface free energy, improving the polymer matrix impregnation, improving fiber dispersion, improving fiber orientation, and enhancing the interfacial adhesion through mechanical interlocking.

For PE-based composite systems, one can use maleated polyethylene (MAPE). There are also various types. In addition to acid number and molecular weight, variations are also based on the polymer used to graft with maleic anhydride such as high-density polyethylene (HDPE) or linear low-density polyethylene (LLDPE). How MAPE improve fiber-matrix interaction are similar to those described above for PP-based composite systems.

2.5 Steam-explosion pre-treatment and fiber fractionation

Steam explosion is an extensively studied and well-described method to defibrate and fractionate lignocellulosics into individual components, i.e., cellulose, hemicelluloses, and lignin [40]. In this process a sample is placed in a pressure vessel that is a digester, and vaporized using saturated steam for 20 sec. to 20 min. at high

temperature (200 – 270°C) and high pressure (14 – 60 kg/cm²). The pressure in the digester is then dropped quickly by opening the steam valve and the material is exposed to normal atmospheric pressure to cause explosion, which disintegrates the lignocellulosic material into individual fibers and/or fiber bundles [51]. Solid residue produced following the steam explosion treatment is called steam-exploded fiber (SEF) [40]. A severity factor, R_0 that groups the treatment temperature and time into a single variable, was used to characterize the severity of the treatment. The factor is, defined as [52]:

$$R_0 = \int_0^t \exp\left[\frac{(T_1 - T_0)}{14.75}\right] dt$$

where T_1 = reaction temperature, °C

T_0 = base temperature, °C

t = retention time, minute

Steam explosion can be conducted batch wise or continuously. A batch type device is available from IOTECH Corporation, while the continuous type device from STAKE Technology. Both companies reside in Canada. Figure 2.4 shows a continuous-type device for steam explosion [53].

Under the drastic pressure and temperature conditions of steam explosion, the resultant steam ionization and acetic acid formation catalyse the degradation of hemicelluloses and lignin, while only a partial degradation of cellulose occurs [54]. Hemicelluloses and lignin can be removed from the pretreated solid residue by successive extractions (fiber fractionation), first with water and then with organic solvents and/or

alkaline aqueous media [54]. Standard fractionation protocol to isolate the three components of a lignocellulosic material is shown in Figure 2.5 [55].

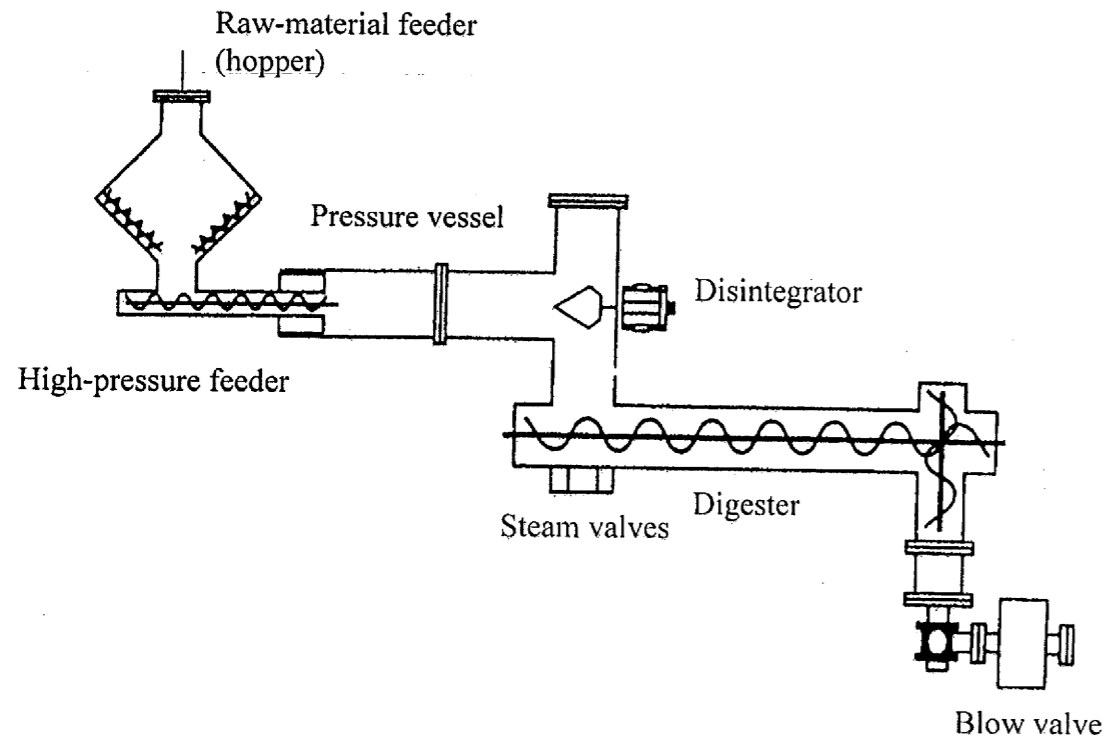


Figure 2.4 A continuous-type device for steam explosion [53].

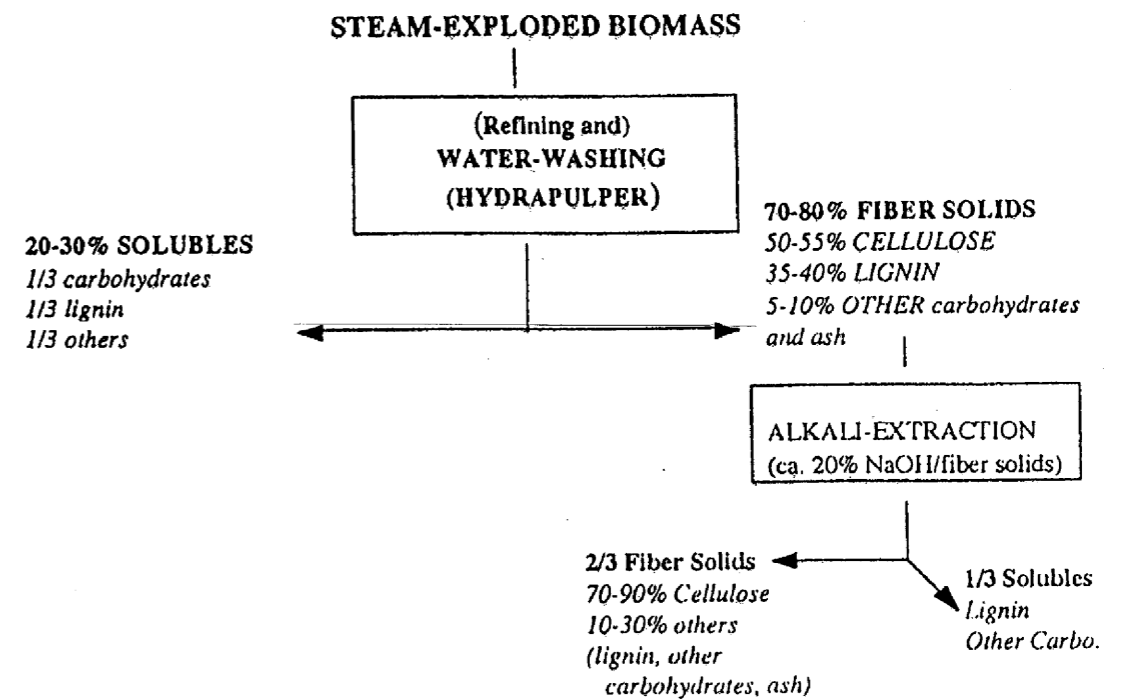


Figure 2.5 Standard fractionation protocol for steam-exploded lignocellulosic material [55].

2.5.1. Steam-exploded fiber

The use of steam-exploded fiber particularly AEF has received much attention in Australia, France, and Canada for the production of paper and paperboards [55]. Recently, several researchers have shown some interest in the use of AEF in thermoplastics. High-density polyethylene (HDPE) had been filled with steam-exploded bagasse fiber. The effects of compression molding temperature, fiber content and treatment with polymethylene polyphenol isocyanate (PMPPIC) on the composite properties were reported [56]. There was also a study on PP filled with steam-exploded hemp bast fiber. MAPP was used to enhance fiber/matrix interfacial adhesion [57]. Another study used steam-exploded broom fiber instead of hemp. In addition to thermal

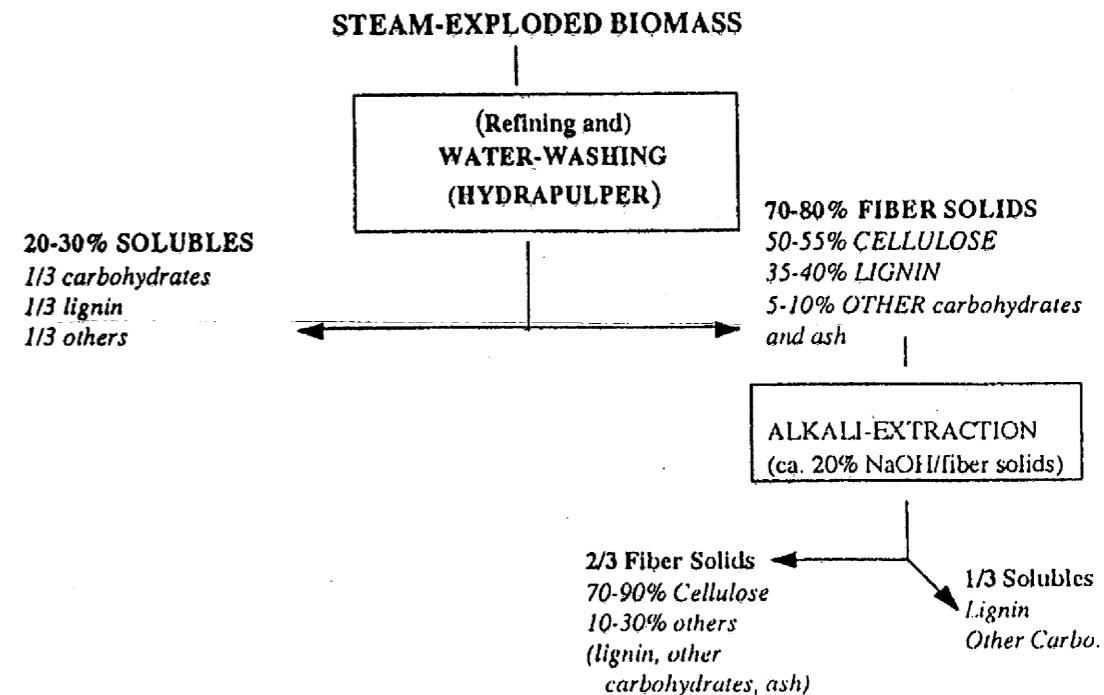


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and tensile properties, water absorption study too was carried out. The composite systems were compatibilized with the use of MAPP [58]. Positive results had been reported by these researchers, which suggest that AEF can be used as effective reinforcing filler for thermoplastics. In addition, AEF also has been claimed to exhibit better characteristics than fibers derived from other conventional fiber extraction methods like retting. Such characteristics include lower moisture uptake, higher thermal stability, better surface roughness, greater reactivity towards chemical and biochemical reagents, and higher cellulose crystallinity [40, 54, 55, 58] These desirable characteristics may help to minimize several existing problems of lignocellulosic/thermoplastic composites such as poor dimensional stability as well as mechanical properties under high humidity environments.

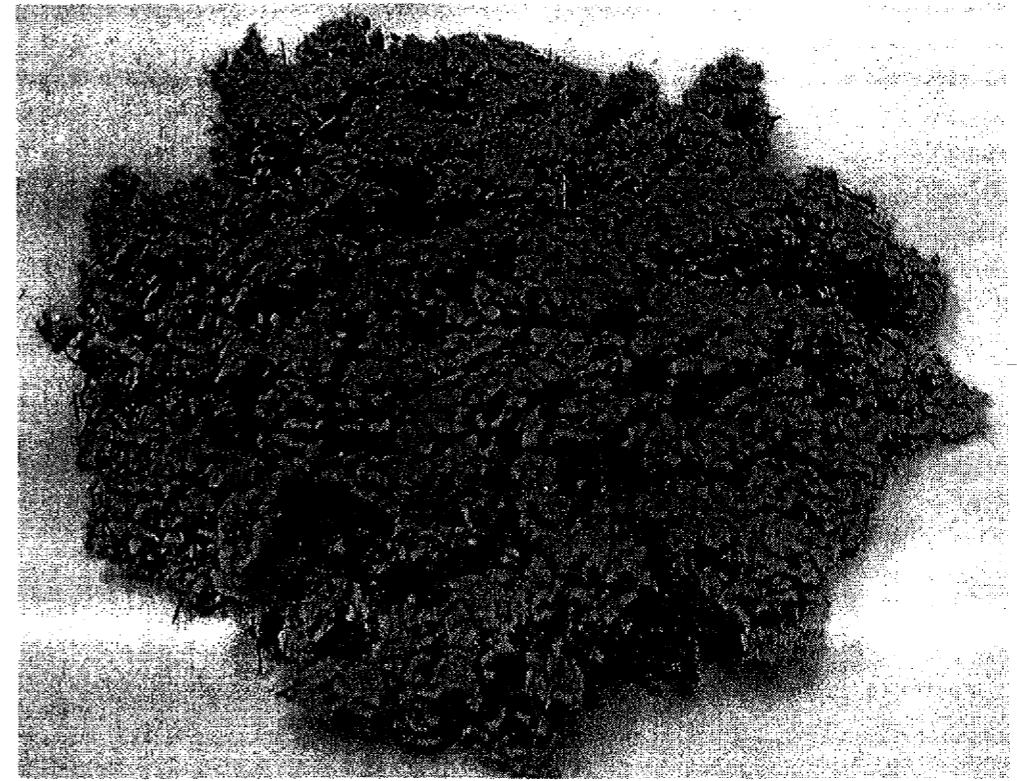


Figure 3.1. A photo of AEF.

A commercially available maleated polyethylene (MAPE), Fusabond[®]E MB226D (DuPont), was used as coupling agent to improve the compatibility and adhesion between AAEF and the PE matrix. Product data sheet for the materials can be found in Appendix B.

3.2 Compounding and molding

AAEF was initially dried for 3 hours at 105°C. PE and AAEF were melt-blended with or without MAPP using 2-roll mill at 160°C. The proportion of AEF was varied from 10 to 40 weight percent while the compatibilizer weight (if used) was at 5 wt. % (based on fiber dry weight). Following compounding, composite sheets were prepared by

melt pressing chips of the compounded material on a hot press at 160°C and 14.3 MPa for 10 minutes to form 1 mm thick plates. In addition, composites of HDPE and AEF were also prepared for comparison purposes.

3.3 Characterization

3.3.1 Chemical compositions

Chemical composition of AEF was carried out at FRIM according to TAPPI-T204 om-88 (ethanol-toulene) and TAPPI-T222 om-88 (Lignin).

3.3.2 Scanning electron microscopy (SEM)

Qualitative observations of AEF and AAEF surface morphology were carried out with a Ziess Supra 35 VP scanning electron microscope at an accelerating voltage of 10kV. Fiber samples were mounted on aluminum stubs and then gold-coated to minimize electron charging effects during examination. Examinations of the tensile fracture surface of the composites were also carried out with SEM to investigate the quality of filler / matrix interface.

3.3.3 Fourier transmission infrared spectroscopy (FTIR)

FTIR spectra of AEF and AAEF were obtained with the KBr technique, using a Perkin Elmer System 2000 spectrophotometer. All the FTIR spectra were recorded in absorbance units within the wavenumber range of 4000 – 400 cm^{-1} , with a resolution of 4 cm^{-1} . There were 32 scans for each spectrum.

3.3.4 'Wetting' analysis

AEF and AAEF samples were tested for their qualitative relative ability to be wetted by water and diethyl ether [59]. The fillers were immersed in test tubes containing water/ether two-phase mixture. The mixtures were shaken briefly and then allowed to stand at room temperature overnight. The mixtures were then examined to determine whether the fillers had fallen to the bottom of the water layer or remained at the water-ether interphase.

3.3.5 Thermogravimetric analysis (TGA)

Thermogravimetric analysis of AEF and AAEF was performed with a Perkin Elmer TG Analyzer TGA7. The analysis was carried out with 5 – 10 mg of sample at a heating rate of 10°C/min. under a constant flow of nitrogen. The loss in material weight was registered in the range from 30 to 600°C.

3.3.6 Melt flow index (MFI)

Melt flow index measurements were performed using a Dynisco polymer melt flow indexer in accordance with ASTM D 1238. The measurements were taken at 190°C using 2.16 kg weight.

3.3.7 Tensile test

Tensile test specimens were obtained from the compression-molded sheets. The specimens were cut from the molded sheets using a dumbbell cutter. Tensile measurements were conducted at room temperature according to ASTM D 638 using a

computerized laboratory tensile tester (M500 Tensometric AZ). The crosshead speed was 5 mm/min. All tensile properties, i.e. tensile strength, elongation at break and Young's modulus, were automatically calculated by the computing software provided. The properties were reported after taking the statistical average values of at least six measurements for each composite system. Tensile tests were also conducted on specimens after 2, 4, 6, and 30 days of water immersion.

3.3.8 Water absorption

Tensile specimens were used for the study of water absorption. After being vacuum dried at 70°C to a constant weight, the specimens were immersed in an immersion tank filled with distilled water at room temperature for 30 days. The weights of the specimens were measured at regular intervals using an analytical balance and the water uptake at any time (W_t) was calculated according to the following equation:

$$\text{Water uptake, } W_t(\%) = \frac{W_t - W_2}{W_2} \times 100$$

where W_t and W_2 are the weight after the exposure time t and the dry weight, respectively.

4.0 RESULTS AND DISCUSSION

4.1 Fiber Characterization

Treated and untreated AEF were characterized by means of SEM, FTIR, and TGA. The ability of the fiber to be wetted by water was also determined. The purpose was to investigate the change in surface morphology, surface chemistry and thermal stability of AEF as the results of the acetylation.

4.1.1 Chemical Composition

The average chemical composition of the Acacia mangium wood flour (WF) and AEF are given in Table 4.1. AEF as shown in Table 4.1 has lower percentage of lignin and hemicelluloses (this is based on the pentosan content) but high in α -cellulose than WF. This is expected since most of lignin and hemicelluloses have been removed to some extent during the fiber production which involved steam-explosion and fiber fractionation.

Table 4.1 Chemical composition of Acacia mangium wood flour (WF) and AEF.

Type of tests	WF	AEF
Ash content (%)	0.84	1.19
Ethanol-toluene content(%) - extractives	9.40	1.15
Holocellulose content (%)	76.48	85.67
α - cellulose content (%)	31.32	46.17
Lignin content (%)	38.95	10.77
Pentosan content (%)	20.78	2.48
(represent 85-90% of hemicelluloses)		

4.1.2 SEM

Figure 4.1 shows SEM micrograph of AEF. AEF as shown is composed of well-separated long fine ribbon-like fibers with diameter less than 20 μm . The average aspect ratio of AEF was not determined due to curved and twisted nature of AEF and fiber entanglements.

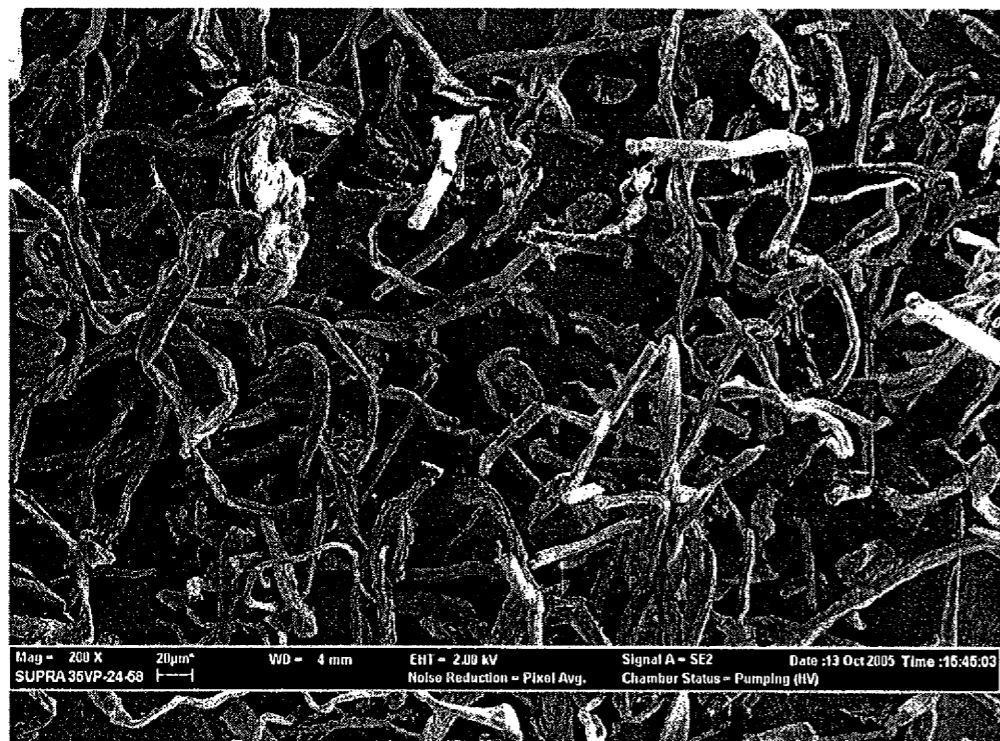


Figure 4.1. SEM micrograph of AEF at a magnification of 200 X.

Figure 4.2 compares the SEM micrographs of acetylated AEF (AAEF) at different weight percent gain (WPG) with untreated AEF. It can be seen that AAEF 4.9 WPG, AAEF 13.7 WPG as well as AAEF 18.3 WPG show kind of similar surface morphology to that of untreated AEF. The fibers appear clean and rough with little surface debris and

overgrowths. It is, therefore, reasonable to suggest that acetylation does not affect surface morphology of AEF.

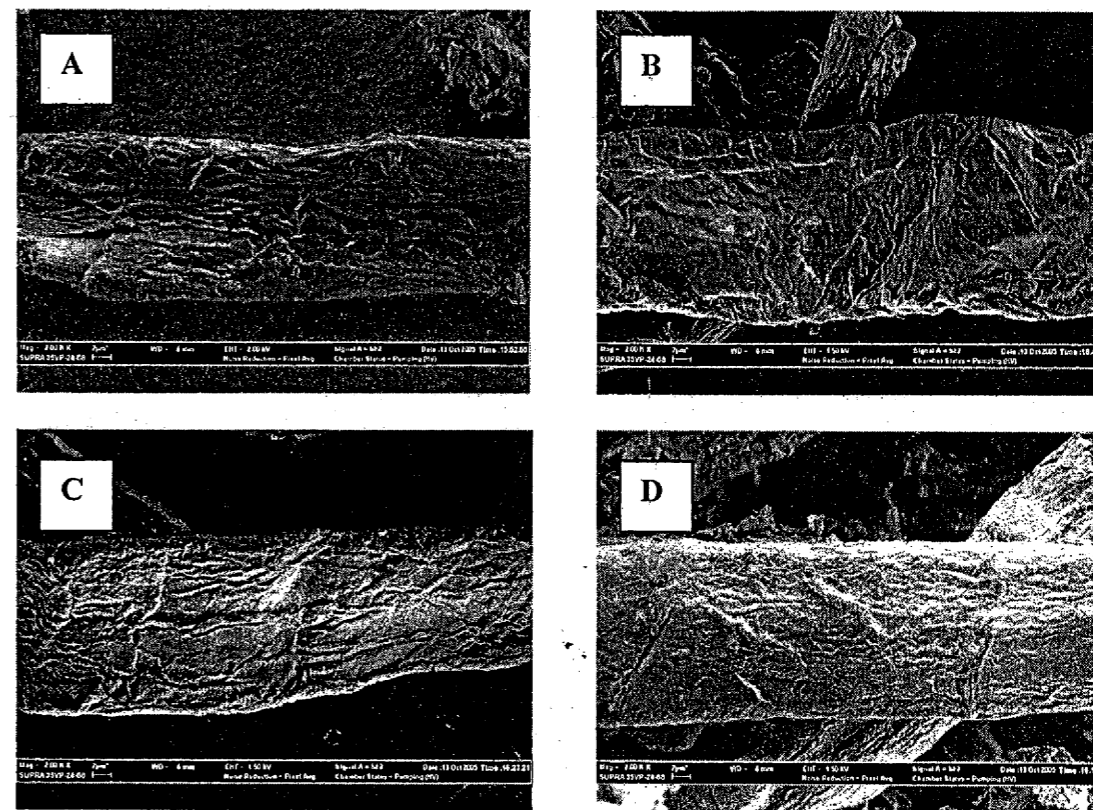


Figure 4.2 SEM micrographs of AEF and acetylated AEF (AAEF) at a magnification of 2000 X. (A) AEF, (B) AAEF 4.9 WPG, (C) AAEF 13.7 WPG, and (D) AAEF 18.3 WPG.

4.1.3 FTIR

Successful reaction of AEF with acetic anhydride will lead to the formation of ester bonds. To confirm the formation ester bonds, FTIR study was performed on both AEF and AAEF samples. Since the carbonyl ($\text{C}=\text{O}$) stretching vibration of various carbonyl-containing groups including esters absorb in the $1750\text{-}1720\text{ cm}^{-1}$ region, the discussion will be focused on this region.

The FTIR spectra of AEF and AAEF having different WPG are shown in Figure 4.3. The FTIR spectrum AEF does not show any absorption bands within the 1750-1700 cm^{-1} region. This is in contrast with typical wood samples which show absorption bands

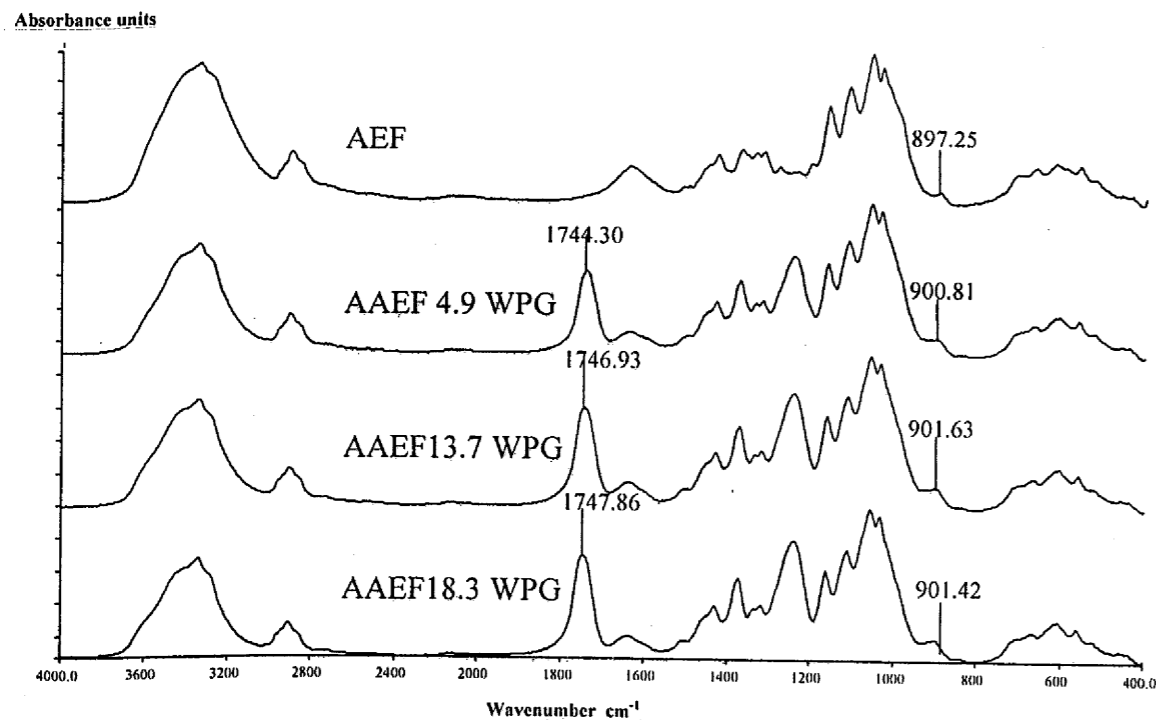


Figure 4.3 FTIR spectrum of AEF and AAEF.

within the region due to various carbonyl-containing groups such as acetyl groups in hemicelluloses, carbonyl aldehyde and esters in lignin and extractives and carboxyl groups in hemicelluloses, lignin and extractives [37]. Such spectrum of AEF is expected as most of these carbonyl-containing groups that are associated with lignin, hemicelluloses, and extractives have been removed during the extractions following the steam explosion treatment. Removal to some extent of these components from AEF is evident from Table 4.1.

A comparison of the FTIR spectra of AEF and AAEF provides clear evidence that the treatment of AEF with acetic anhydride has resulted in a change in absorption band intensities within the 1750-1700 cm^{-1} region. AAEF 4.9 WPG, AAEF 13.7 WPG, and AAEF 18.3 WPG distinctly show strong, sharp, and well-defined adsorption bands respectively at 1744, 1746, and 1748 cm^{-1} . The presence of these strong bands suggests the presence of significant amount of carbonyl-containing groups in the AAEF samples. Carbonyl-containing groups that most are probable responsible for these absorption bands are the ester bonds formed from the reaction between acetic anhydride and the hydroxyl groups of AEF. The FTIR results therefore confirmed successful reaction of AEF and acetic anhydride.

4.1.4 'Wetting' analysis

In this experiment, the wettability of AEF and AAEF samples was examined. The samples were separately shaken in ethyl ether-water mixture with a shaker and then allowed to stand at room temperature overnight. Figure 4.4 shows a photograph taken from the experiment. As shown in the Figure, AEF precipitated completely at the bottom of the lower layer (water layer), while all AAEF samples, i.e. AAEF4.9, AAEF13.7, and AAEF18.3 gathered together above the interface between water and ethyl-ether. These observations imply that AEF is hydrophilic, while AAEF samples are hydrophobic. Most of thermoplastic matrices are hydrophobic. Having hydrophobic wood fibers might improve compatibility with thermoplastic matrices to promote fiber-matrix interfacial adhesion and also facilitate fiber dispersion in the composite system. Another advantage is high resistance to water absorption as because of some of the hydroxyl groups of AEF

have reacted with acetic anhydride reducing the amount of accessible hydroxyl groups to interact with water molecules.

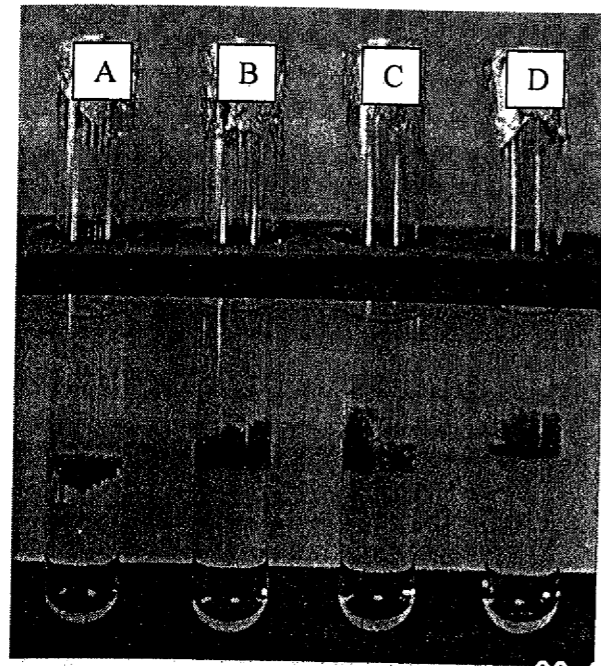


Figure 4.4 A photograph from the 'wettability' test. The existence of (A) AEF, (B) AAEF 4.9 WPG, (C) AAEF 13.7 WPG, and (D) AAEF 18.3 WPG within the ethyl-ether/water two layer systems.

4.1.5 TGA

The thermal behavior of AEF and AAEF in N_2 atmosphere was examined through TGA curves within the temperature range from 30 to 600°C. The purpose was to determine the effect of fiber surface treatment on the thermal stability of AEF. As shown in Figure 4.5, AEF exhibits rapid thermal decomposition between 200 and 380°C. AAEF samples, on the other hand show thermal degradation between 300 and 400°C.

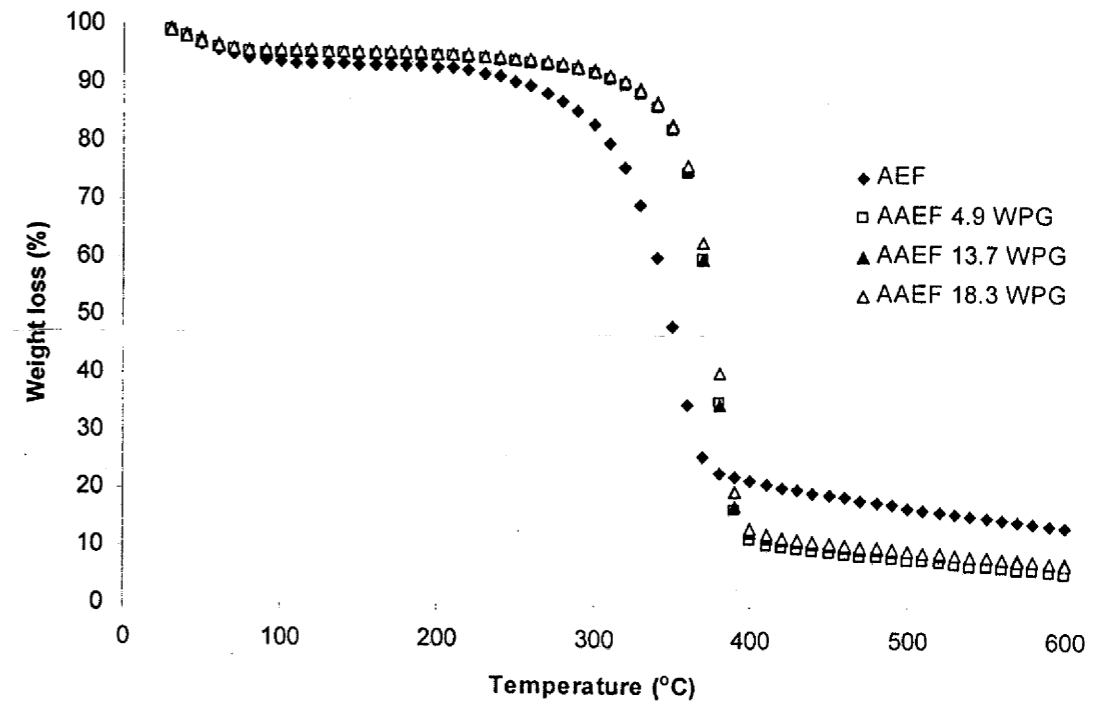


Figure 4.5 TGA curves for AEF and AAEF samples.

In order to characterize thermal stability, temperature corresponding 5, 10, and 15% loss in fiber weight are listed in Table 4.2. It is evident from the table that the treatment with acetic anhydride has resulted in a significant increase in the thermal stability of AEF. For the same amount of weight loss, it is observed that the temperature that corresponds to the loss is higher for AAEF of increasing WPG. The high thermal stability of AAEF samples is probably due to the greater thermal stability of acetyl groups attached to the fiber cell wall polymers.

Table 4.2 Thermal stability of AEF and AAEF.

Sample	Temperature corresponding to below weight losses (°C)		
	5 wt. %	10 wt. %	15 wt. %
AEF	72	259	294
AAEF 4.9 WPG	149	322	345
AAEF 13.7 WPG	222	328	347
AAEF 18.3 WPG	228	326	346

4.2 Composite Characterization

4.2.1 Tensile properties

4.2.1.1 Effect of filler content

Figure 4.5 shows the results of tensile strength for HDPE and AEF filled- HDPE (HDPE-AEF) composites. It is clear from the Figure that the tensile strength decreases with the addition of AEF. Further decreases in the property is observed with increasing filler content from 10 to 40 wt. %. To further understand the adverse effect of filler content on tensile strength, percent decrease of the property was plotted over filler content (Figure 4.6). A positive linear relationship is observed with R^2 of about 0.9107 suggesting strong influence of filler content on the tensile strength of HDPE-AEF composites. The decrease in tensile strength with the addition of AEF is expected due to inherent incompatibility between the polar AEF and the non-polar HDPE matrix. This is evident from the SEM micrograph in Figure 4.7 which shows long smooth pull-out of AEF fiber. Void too is obvious around the fiber as indicated by the arrow suggesting poor interaction between AEF and HDPE. In composites, poor fiber-matrix interaction will result in a decrease in composite strength due to lack of stress-transfer from the matrix to the fiber.

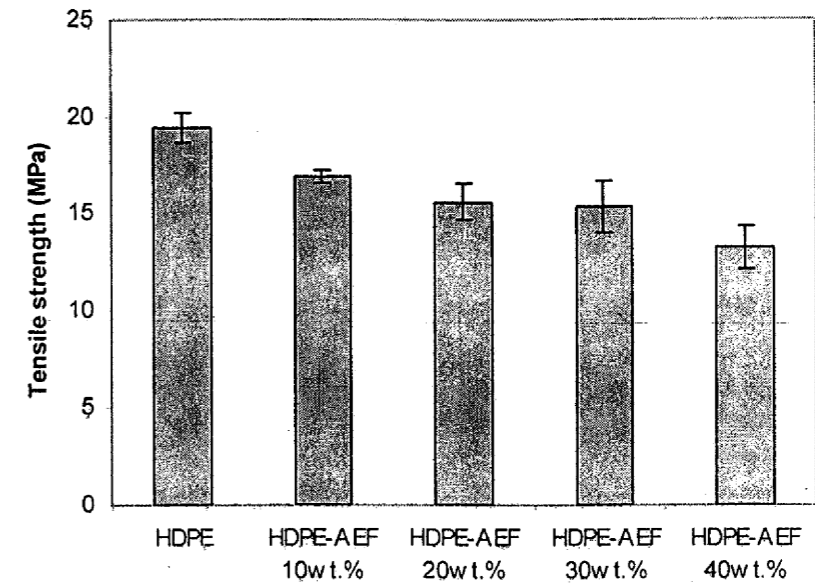


Figure 4.5 Effect of filler content on the tensile strength of HDPE and HDPE-AEF composites.

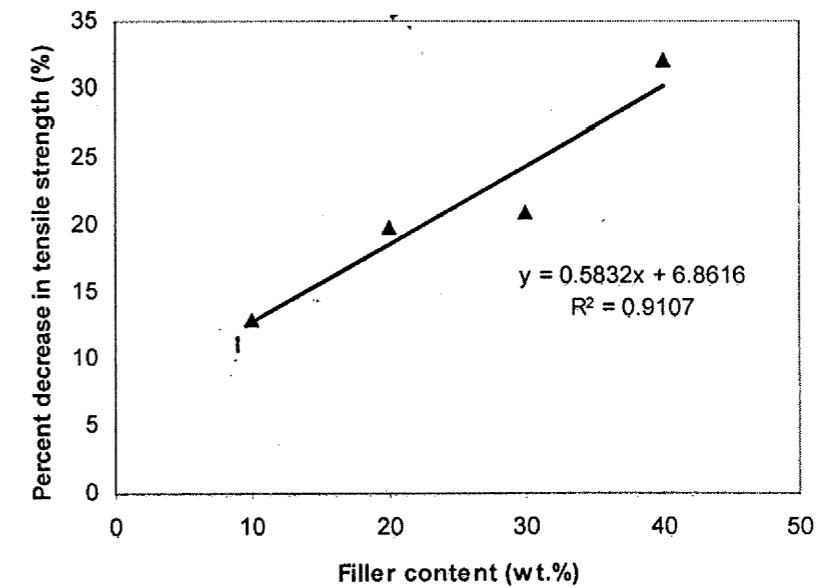


Figure 4.6 Percent decrease in tensile strength versus filler content for HDPE-AEF composites.

positive effect on the property except when HDPE is filled with AAEF 4.9 WPG.

Acetylation as has been shown earlier to change AEF behavior from hydrophilic to

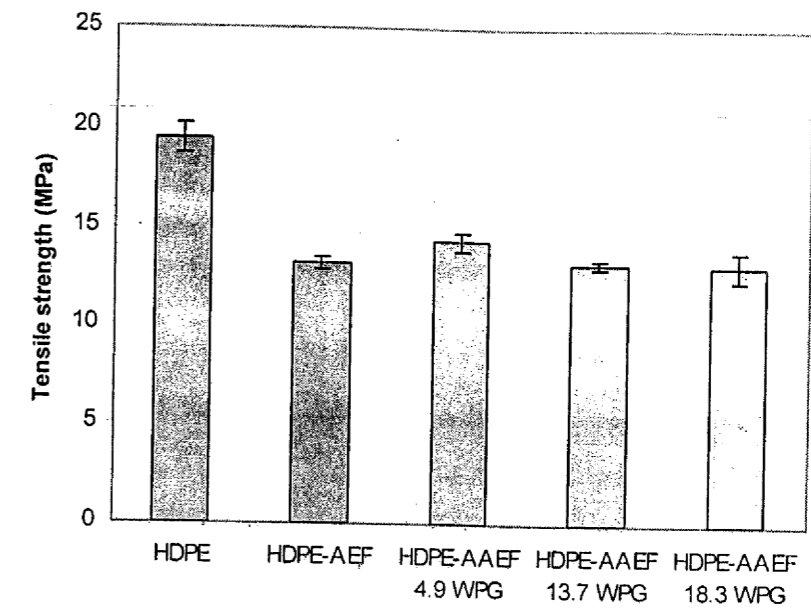


Figure 4.12 Tensile strength results for HDPE, HDPE-AEF and HDPE-AAEF composites.

hydrophobic. Since now both AAEF and HDPE are hydrophobic, it is predicted that AAEF might exhibit better compatibility with HDPE than AEF does. This would result in an increase in tensile strength of the composites as fiber-matrix interaction is now possible to facilitate stress transfer from HDPE to AAEF. This, however, is not observed. Although AAEF might be compatible with HDPE, AAEF needs to interact with HDPE either via secondary, primary, mechanical inter-locking or etc and form an acceptable interface for stress transfer to occur. Examination of the tensile fracture surfaces of HDPE-AAEF composites (Figure 4.13) reveals many long pull out of fibers. The pull-out

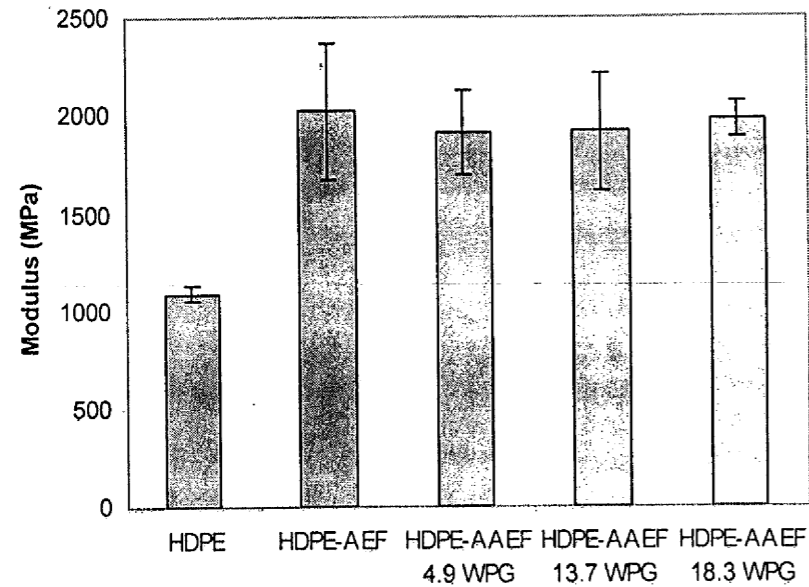


Figure 4.15 Modulus results for HDPE, HDPE-AEF and HDPE-AAEF composites.

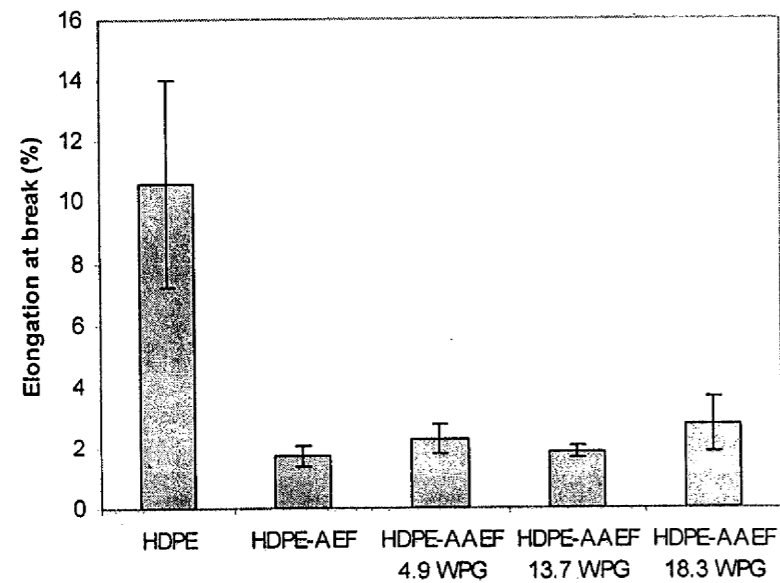


Figure 4.16 Elongation at break results for HDPE, HDPE-AEF and HDPE-AAEF composites.

Table 4.4 Tensile properties of HDPE and HDPE-AEF composites.

Sample	Tensile properties		
	Tensile strength (MPa)	Modulus (MPa)	Elongation at break (%)
HDPE	19.44 ± 0.76	1096.20 ± 41.40	10.59 ± 0.38
HDPE-AEF	13.21 ± 0.33	2018.40 ± 346.02	1.73 ± 0.33
HDPE-AAEF 4.9 WPG	14.32 ± 0.45	1910.00 ± 209.47	2.28 ± 0.49
HDPE-AAEF 13.7 WPG	13.18 ± 0.20	1914.80 ± 294.96	1.86 ± 0.20
HDPE-AAEF 18.3 WPG	13.10 ± 0.73	1979.00 ± 91.00	2.27 ± 0.91

4.2.1.3 Effect of acetylation and compatibilizer

Since AAEF did not show acceptable interaction with HDPE for improvement in tensile strength, MAPE was added to HDPE-AAEF composites during compounding. Filler content was fixed at 20 wt. % while MAPE at 5 wt. % (based on fiber dry weight). MAPE was also added to HDPE-AEF composite for comparison purposes.

Addition of MAPE has resulted in an increase in tensile strength of HDPE-AEF and HDPE-AAEF 13.7 WPG composites (Figure 4.17). This is evident by comparing tensile strength results of HDPE-AEF with HDPE-AEF-MAPE and HDPE-AAEF 13.7 WPG with HDPE-AAEF 13.7 WPG-MAPE composites. This increase in tensile strength suggests the existence of some kind interaction between the fibers, i.e. AEF and AAEF 13.7 WPG, and HDPE matrix. Fracture surface of both composites are shown in Figure 4.18. Smaller amount of pull-out of fibers and shorter as well are observed as compared to the composites with MAPE (Figure 4.13). Examination at a higher magnification of

1000 X reveals embedded AEF and AAEF 13.7 WPG fibers in HDPE matrix (Figure 4.19).

As shown in Figure 4.17, addition of MAPE to HDPE-AAEF composites does not show a positive effect for tensile strength. The property does not seem to increase but slightly decrease when MAPE is added to HDPE-composites filled with AAEF of increasing WPG. HDPE-AAEF 18.7 WPG-MAPE composite for example exhibits 12% reduction in the property as compared to HDPE-AAEF 4.9 WPG-MAPE composite. From the SEM micrographs (Figure 4.18), composites filled with AAEF of increasing WPG show longer pull-out of fibers and increasing in number. This can be understood by comparing Figure 4.18 (C) either with Figure 4.18 (D) or Figure 4.18 (E). The morphologies too are some what similar to that of HDPE-AEF composite (Figure 4.18 (A)). At 1000 X magnification (Figure 4.19), AAEF 4.9 WPG fiber is seen well embedded in HDPE matrix. AAEF 13.7 WPG fiber is also embedded in HDPE matrix although not as good as AAEF 4.9 WPG fiber. As for AAEF 18.3 WPG fiber, void (marked with an arrow) is observed around the fiber suggesting inferior fiber-matrix interaction. Due to acetylation between the anhydride group of acetic anhydride and the hydroxyl groups of AEF, some of these hydroxyl groups were acetylated reducing the amount of free hydroxyl groups. With increasing WPG, higher amount of hydroxyl groups were reacted leaving fewer free hydroxyl groups. When MAPE was added to HDPE-AAEF composites, these free hydroxyl groups left on AAEF would react with the anhydride groups of MAPE to form ester bonds. The fewer the free hydroxyl groups, the lesser the probability of the reaction to occur. As a result positive effect of adding MAPE to promote fiber-matrix interaction in HDPE-AAEF composites of increasing WPG is not

obvious as compared to HDPE-AEF-MAPE composite. Above argument is supported by the fact that all HDPE-AAEF-MAPE composites show significantly lower value than that of HDPE-AEF-MAPE composite. Percent decrease in tensile strength values of HDPE-AAEF-MAPE composites as compared to HDPE-AEF-MAPE composite are given in Table 4.5. The effect of adding MAPE to HDPE-AEF composite is discussed further in section 4.2.1.4.

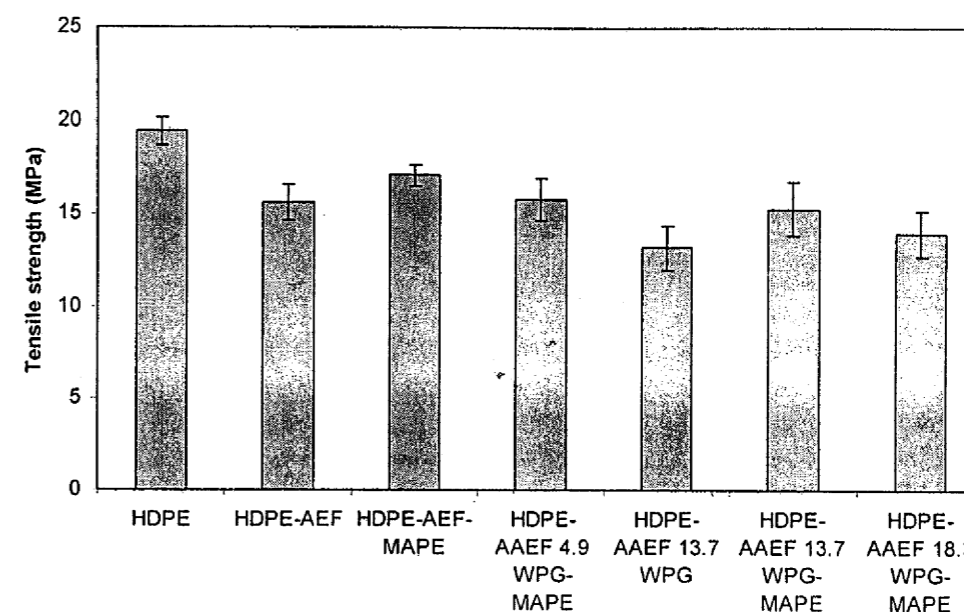


Figure 4.17 Tensile strength results for HDPE, HDPE-AEF, HDPE-AEF-MAPE, HDPE-AAEF, and HDPE-AAEF-MAPE composites.

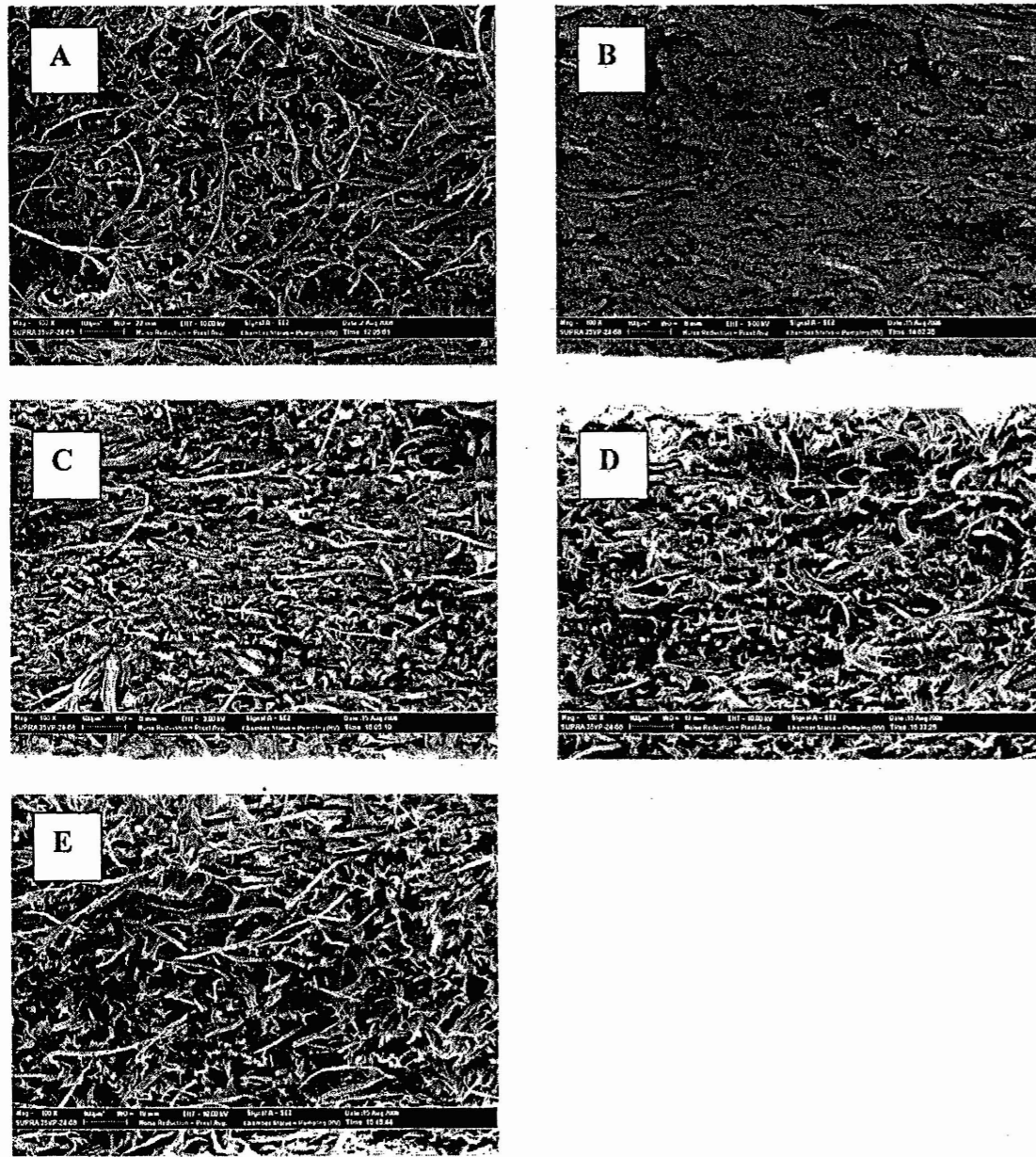


Figure 4.18 SEM micrographs of (A) HDPE-AAEF, (B) HDPE-AAEF-MAPE, (C) HDPE-AAEF 4.9 WPG-MAPE, (D) HDPE-AAEF 13.7 WPG-MAPE, and (E) HDPE-AAEF 18.3 WPG-MAPE composites at a magnification of 100 X.

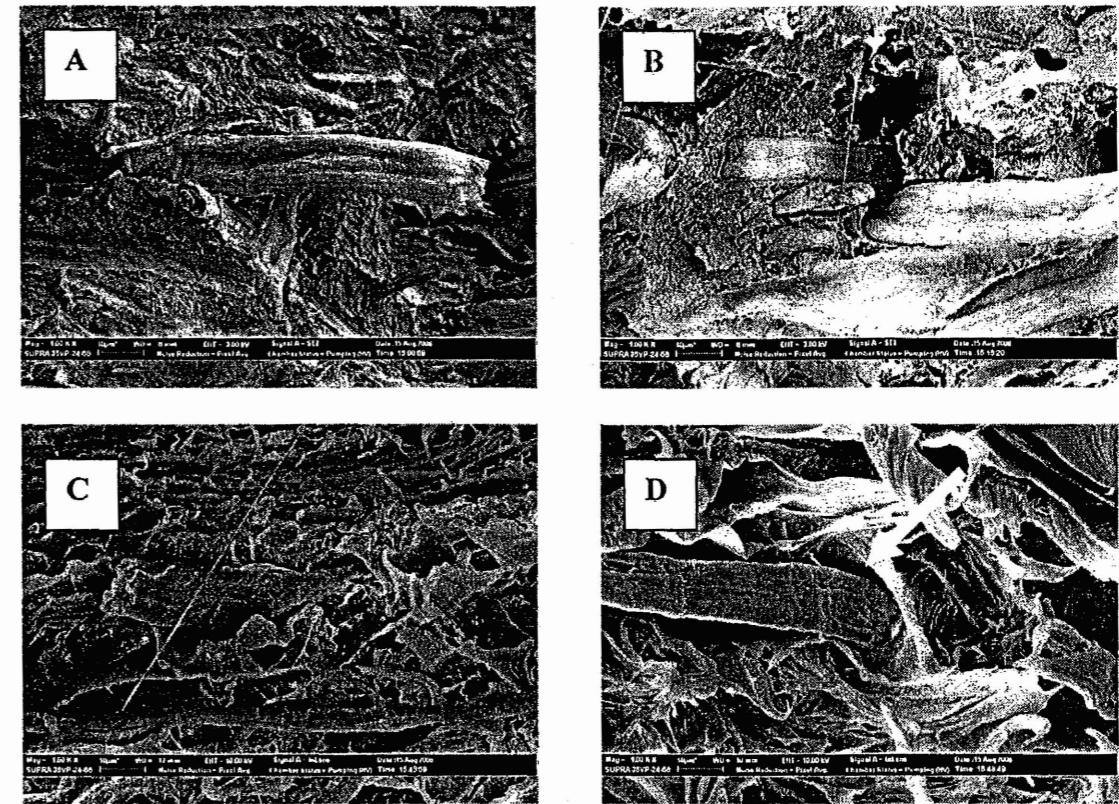


Figure 4.19 SEM micrographs of (A) HDPE-AAEF-MAPE, (B) HDPE-AAEF 4.9 WPG-MAPE, (C) HDPE-AAEF 13.7 WPG-MAPE, and (D) HDPE-AAEF 18.3 WPG-MAPE composites at a magnification of 1000 X.

Addition of MAPE to HDPE-AAEF composite had resulted in a slight decrease in modulus (Figure 4.20). HDPE-AAEF composites too experienced a decrease in the property. This adverse effect is obvious for HDPE-AAEF 18.7 WPG-MAPE composites. The free unreacted MAPE (maleic anhydride grafted polyethylene; the based polymer has lower molecular weight than HDPE) might act as a plasticizer for HDPE matrix.

As for elongation at break (Figure 4.21), an increase in the property is clear when MAPE is added to HDPE-AAEF composites of increasing WPG of the fiber. Similar to modulus, the free unreacted MAPE might act a plasticizer to improve the property.

Table 4.5 Tensile properties of HDPE, HDPE-AEF, HDPE-AEF-MAPE, HDPE-AAEF, and HDPE-AAEF-MAPE composites*.

Sample	Tensile properties		
	Tensile	Modulus	Elongation
	strength (MPa)	(MPa)	at break (%)
HDPE	19.44 ± 0.76	1096.20 ± 41.40	10.59 ± 3.38
HDPE-AEF	15.58 ± 0.97	1524.40 ± 215.52	3.37 ± 1.07
HDPE-AEF-AAEF	17.07 ± 0.57	1308.80 ± 112.25	2.96 ± 0.36
HDPE-AAEF 4.9 WPG-MAPE	15.81 ± 1.15	1205.78 ± 102.04	2.58 ± 0.27
	(-7%)	(-8%)	(-13%)
HDPE-AAEF 13.7 WPG	13.21 ± 1.16	1211.60 ± 136.70	4.05 ± 0.75
HDPE-AAEF 13.7 WPG-MAPE	15.29 ± 1.44	1134.55 ± 150.19	4.45 ± 0.86
	(-10%)	(-13%)	(+50%)
HDPE-AAEF 18.3 WPG-MAPE	13.95 ± 1.22	1085.91 ± 86.72	4.46 ± 1.25
	(-18%)	(-17%)	(+51%)

* Numbers in the parentheses are percent increase (+) or decrease (-) as compared to HDPE-AEF-MAPE composite.

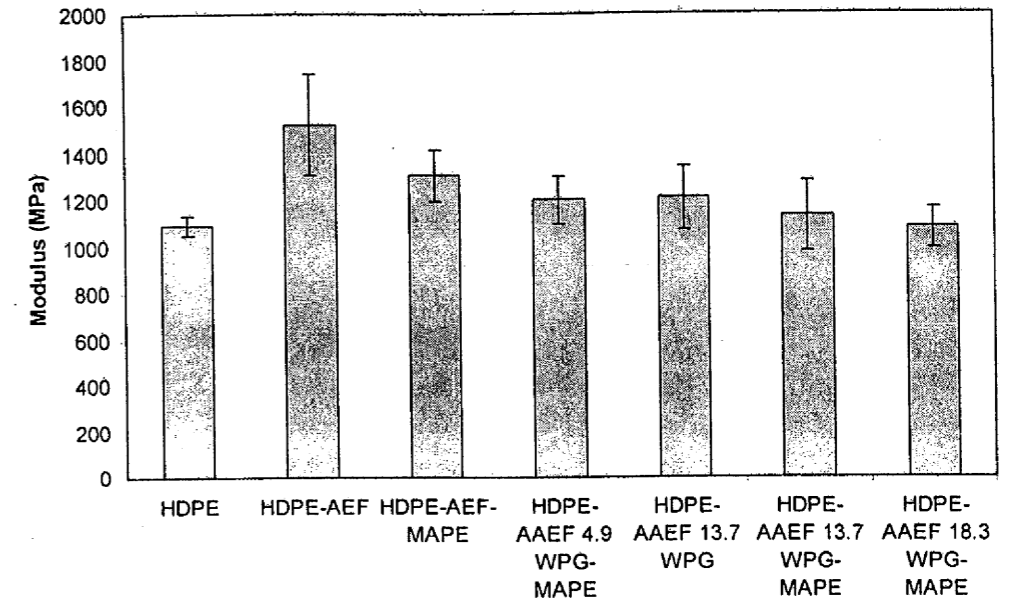


Figure 4.20 Modulus results for HDPE, HDPE-AEF, HDPE-AEF-MAPE, HDPE-AAEF, and HDPE-AAEF-MAPE composites.

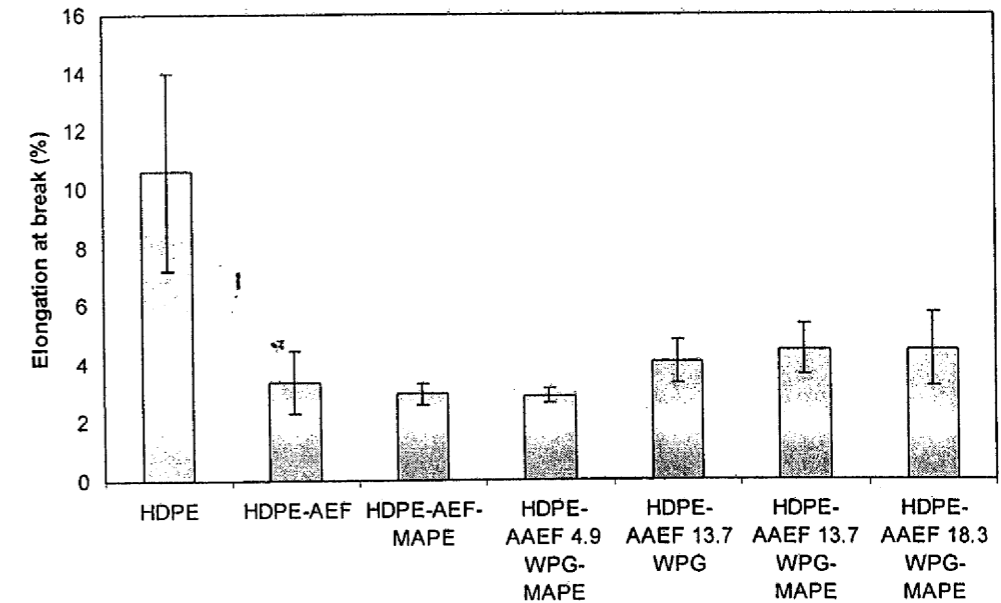


Figure 4.21 Elongation at break results for HDPE, HDPE-AEF, HDPE-AEF-MAPE, HDPE-AAEF, and HDPE-AAEF-MAPE composites.

To support this argument that the free unreacted MAPE acts as a plasticizer for HDPE, melt flow index was determined for each composite system. The results are shown in Figure 4.22. Significant increase in MFI is observed when MAPE is added to HDPE-AAEF composites of increasing WPG of the fiber due to increasing amount of free unreacted MAPE present in the composite systems to act as a plasticizer.

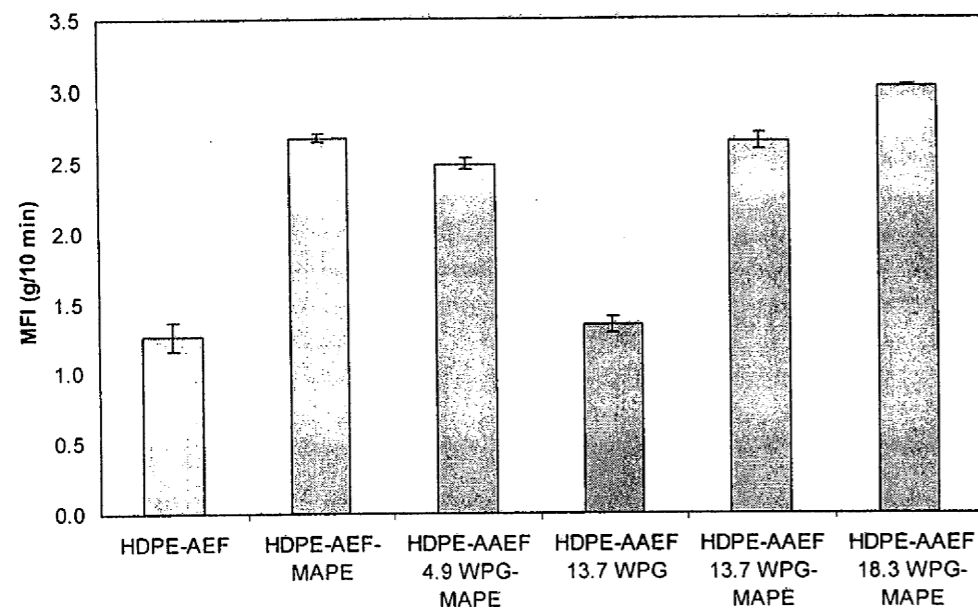


Figure 4.22 Melt flow index values for HDPE, HDPE-AEF, HDPE-AEF-MAPE, HDPE-AAEF, and HDPE-AAEF-MAPE composites.

It is too obvious from the Figure that addition of MAPE to HDPE-AEF composites has resulted in an increase in the MFI value of about 120%. The same observation is also significant for HDPE-AAEF 13.7 WPG-MAPE composite that experienced 115% improvement in the MFI value. This is probably due to 1) improved fiber dispersion and 2) reduced fiber-fiber as well as fiber-matrix attrition. The improved

fiber dispersion is attributed to the presence of PE chains of MAPE on the fiber surface that reduces intermolecular hydrogen bonding between the fibers. The reduced fiber-fiber and fiber-matrix attrition is also attributed to the PE chains of MAPE. The PE chains reduce shearing between the individual fibers and also between the fibers and HDPE matrix resulting in better composite flow.

4.2.1.4 Effect of compatibilizer

Since addition of MAPE to HDPE-AAEF composites did not show positive effects on tensile properties, an attempt was made to study the effect of adding MAPE to HDPE-AEF composites. For this study, filler loading was varied from 10 to 40 wt. % and MAPE content was fixed at 5 wt.% (based on fiber dry weight).

Figure 4.23 shows significant improvement in tensile strength (as compared to pure HDPE) with increasing filler content when MAPE was added to HDPE-AEF composites. At filler content of 10 wt.%, the improvement is not significant because AEF act more as flaw than reinforcing fillers. However, increase in filler content to 30 wt.% improves the tensile strength significantly. Beyond 30 wt.% filler content, the property begins to drop. At low filler loading HDPE matrix is not restrained by enough AEF fibers and highly localized strains occurs in the matrix at low stresses causing the bond between HDPE and AEF to break. As filler content increases, the stress is more evenly distributed and an improvement in the tensile strength becomes more prominent suggesting the ability of AEF to perform as reinforcing filler. This is due improved interaction between AEF and HDPE with the addition of MAPE that can be observed from Figures 4.18 (B) and 4.19 (A). Improved interaction facilitates fiber wetting by the matrix polymer

resulting in acceptable interface for stress transfer to occur in a composite. The maximum improvement of about 29% is observed at 30 wt.% filler content.

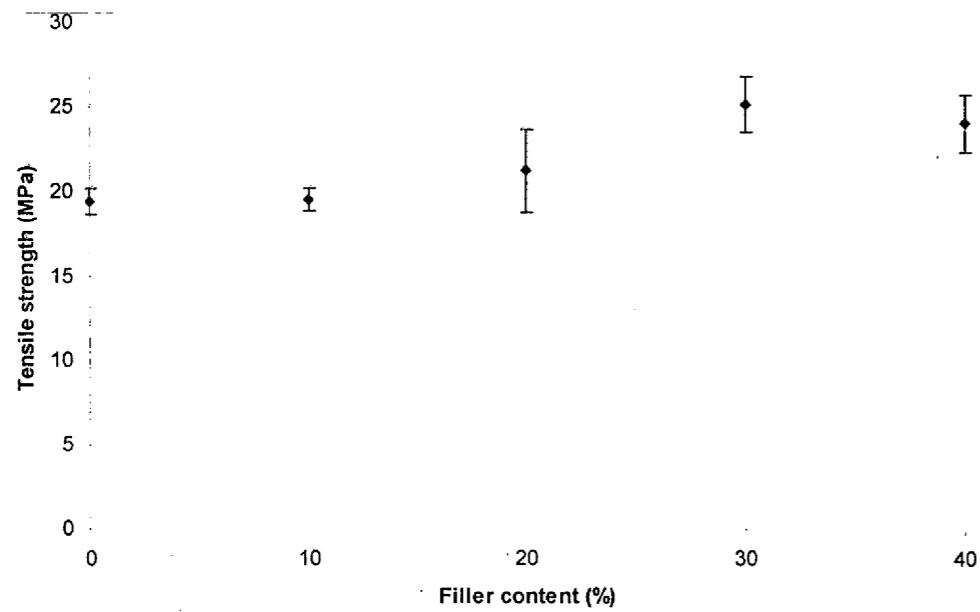


Figure 4.23 Effect of filler content on the tensile strength of HDPE-AEF-MAPE composites.

As for tensile modulus, significant improvement in property is observed with increasing filler content (Figure 4.24). In contrast for elongation at break (Figure 4.25), the property decreases with increasing filler content.

A summary of the tensile properties of HDPE and HDPE-AEF-MAPE composites is given in Table 4.6.

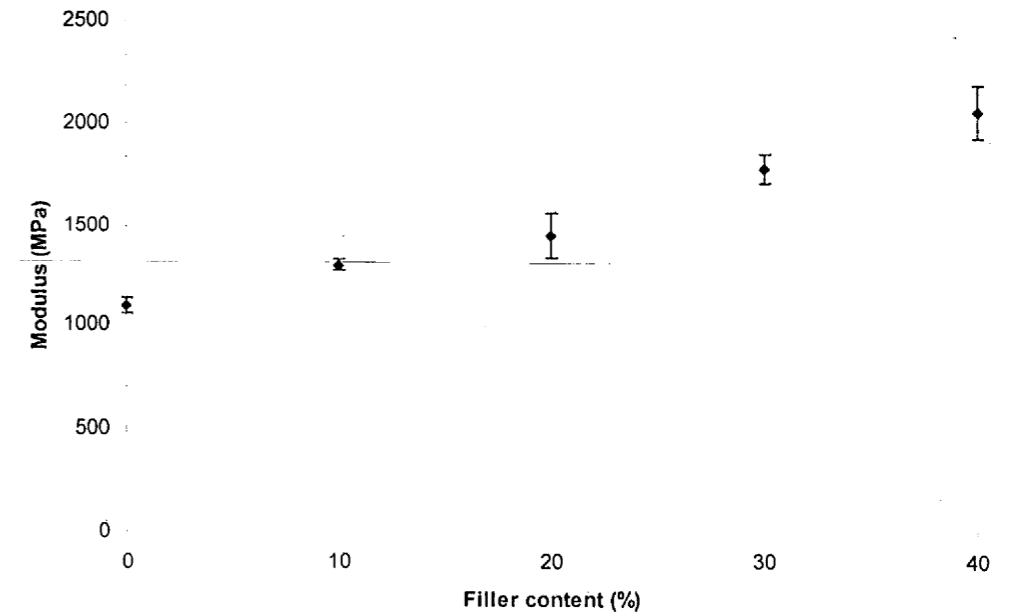


Figure 4.24 Effect of filler content on the modulus of HDPE-AEF-MAPE composites

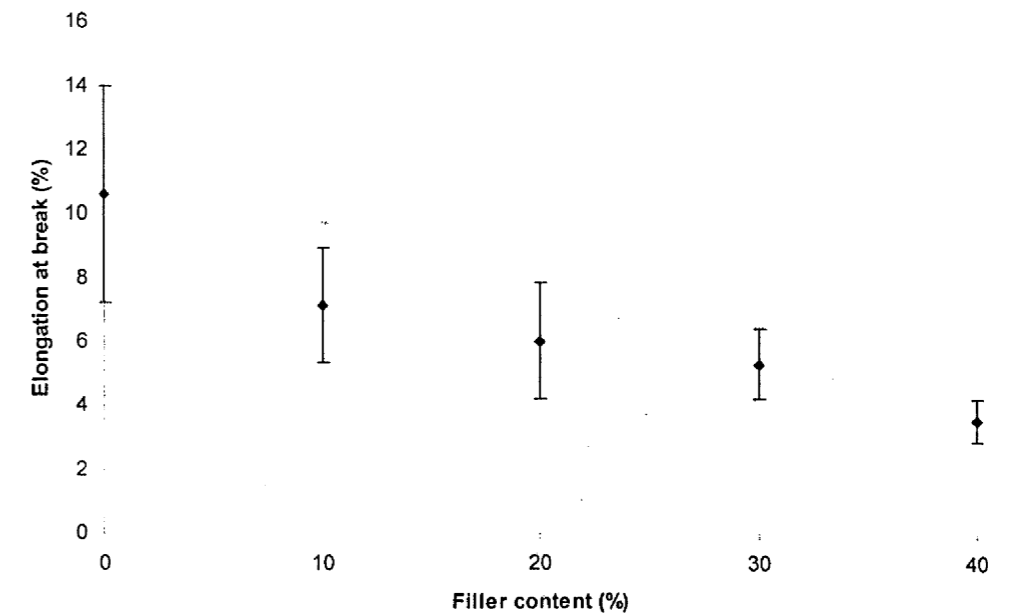


Figure 4.25 Effect of filler content on the elongation at break of HDPE-AEF-MAPE composites.

Table 4.6 Tensile properties of HDPE and HDPE-AEF-MAPE composites*.

Filler content (wt. %)	Filler content (wt. %)		
	Tensile strength (MPa)	Modulus (MPa)	Elongation at break (%)
0	19.44 ± 0.76	1096.20 ± 41.40	10.59 ± 3.38
10	19.52 ± 0.67 (+0.4%)	1301.33 ± 26.98 (+19%)	7.14 ± 1.78 (-48%)
20	21.23 ± 2.44 (+9%)	1451.93 ± 111.28 (+32%)	6.02 ± 1.80 (-43%)
30	24.15 ± 1.63 (+24%)	1782.33 ± 72.61 (+63%)	5.29 ± 1.11 (-50%)
40	24.01 ± 1.67 (+23%)	2059.07 ± 127.80 (+88%)	3.53 ± 0.67 (-67%)

* Numbers in the parentheses are percent increase (+) or decrease (-) as compared to pure HDPE.

4.2.2 Water absorption study

4.2.2.1 Effect of filler content

4.2.2.1.1 Water uptake

Figure 4.26 shows water uptake for HDPE and HDPE-AEF composites as a function of immersion time in days. Each data point represents the average of six specimens.

HDPE as seen in Figure 4.26, does not exhibit appreciable amount of water uptake after 30 days of water immersion at room temperature. This is explained by the hydrophobic nature of the material. HDPE-AEF composites on the other hand show appreciable amount up take of water with increasing filler content. Since HDPE does not

absorb water, it is then reasonable to assume that the water uptake is due to uptake of water by the fibers, AEF, and also voids. Uptake of water by AEF is expected since AEF has hydroxyl and other oxygen-containing groups such as esters that can interact with water molecules via hydrogen bonding.

All water uptake curves show kind of similar trend, i.e. kind of a linear increase of water uptake in the beginning followed by a gradual increase and then a plateau. It is also observed that composites filled with higher filler content reach saturation earlier than other composites with lower filler content. A strong linear relationship is observed between equilibrium water uptake and filler content with R^2 of about 0.9838 (Figure 4.27).

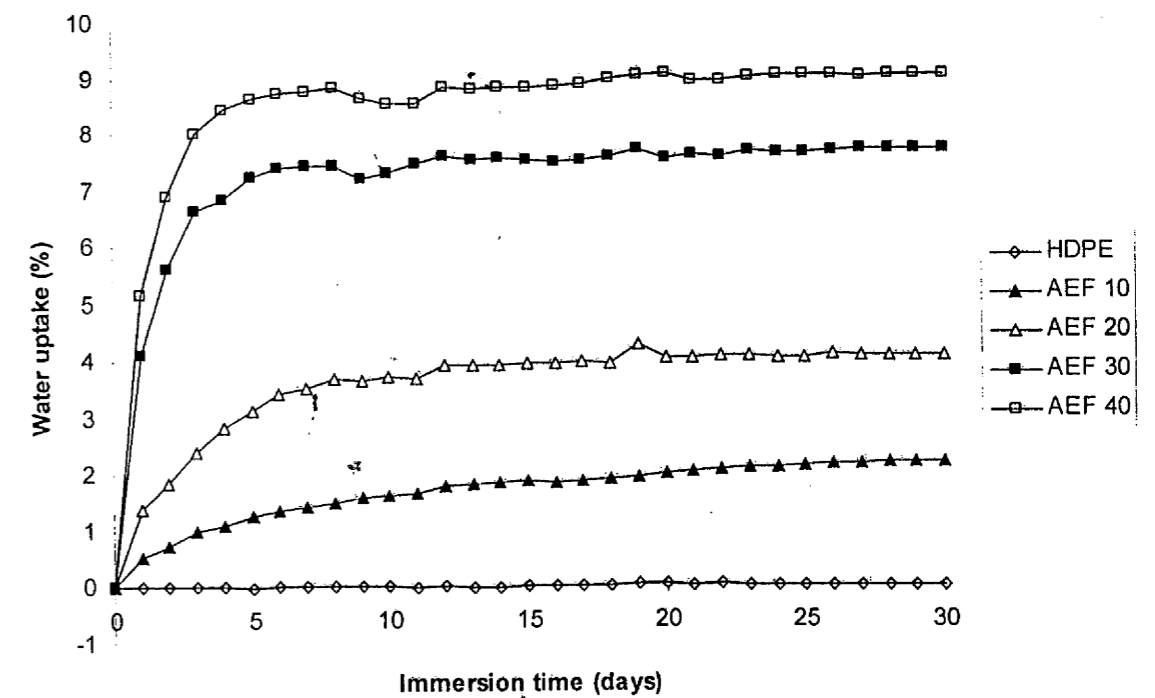


Figure 4.26 Water uptake for HDPE and HDPE-AEF composites immersed in water at room temperature for 30 days.

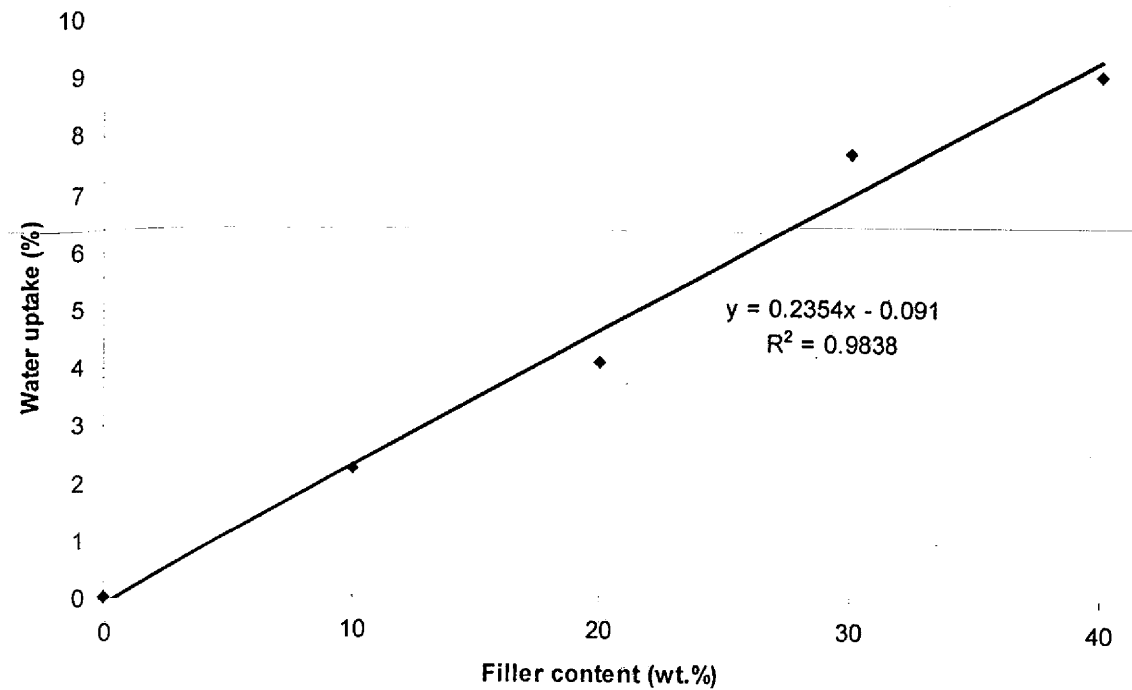


Figure 4.27 Equilibrium water uptake versus filler content for HDPE-AEF composites.

4.2.2.1.2 Tensile properties

After 30 days of water immersion, it is observed that the tensile strength of HDPE is hardly affected (Figure 4.28). This is explained by the negligible amount uptake of water by the hydrophobic material. As for HDPE-AEF composites a slight decrease in the property is observed but not that significant. Composites often show a decrease in the tensile strength due to absorption of water because of degradation of the fiber-matrix interface and microcrack formation as a result of fiber swelling. Degradation of fiber-matrix interface is not likely to occur as already shown in Figure 4.7 for example that AEF does not interact well with HDPE to form acceptable interface for stress transfer to occur. As for microcracks, swelling of AEF would not cause adjacent matrix under high stress as there are voids between them due to poor wetting of AEF by HDPE.

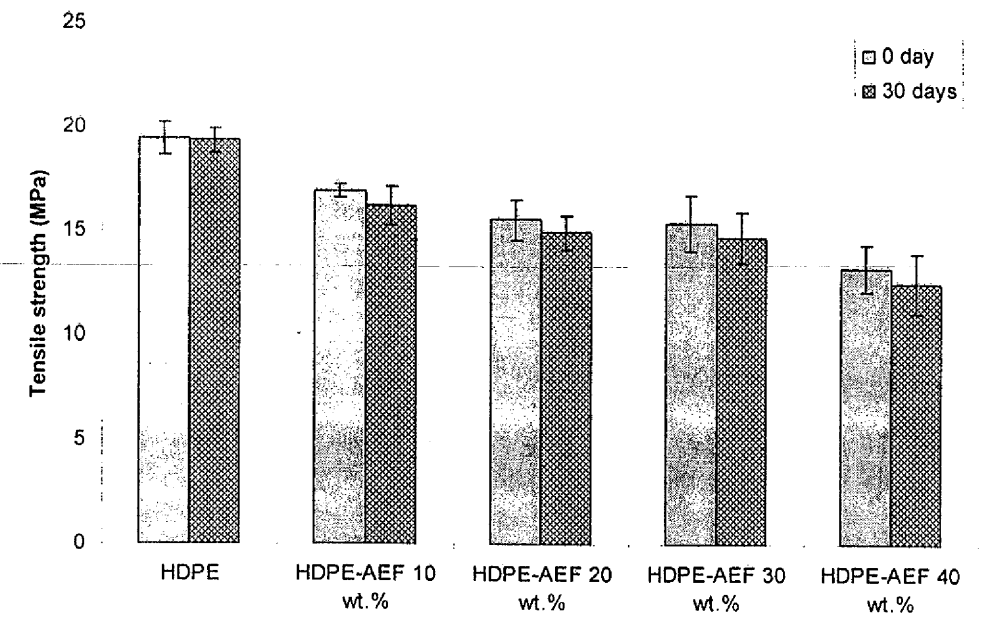


Figure 4.28 Effect of water absorption on the tensile strength of HDPE and HDPE-AEF composites.

In contrast with modulus (Figure 4.29), the absorption of water has resulted in a decrease in the property for HDPE-AEF composites. The decrease is becoming more prominent with increasing filler content. It is too observed that with increasing filler content the percent decrease is more and significant. Since HDPE hardly absorbs water and the modulus remains unaffected after 30 days of water immersion, it is suggested that the decrease is due to the effect of water molecules absorbed by AEF on its stiffness. The higher the filler content, the greater decrease in the modulus.

As for elongation at break (Figure 4.30), the proper increases with absorption of water by HDPE-AEF composites. Percent increase increases with increasing filler content. It is probably due to plasticization effect of water molecules on AEF.

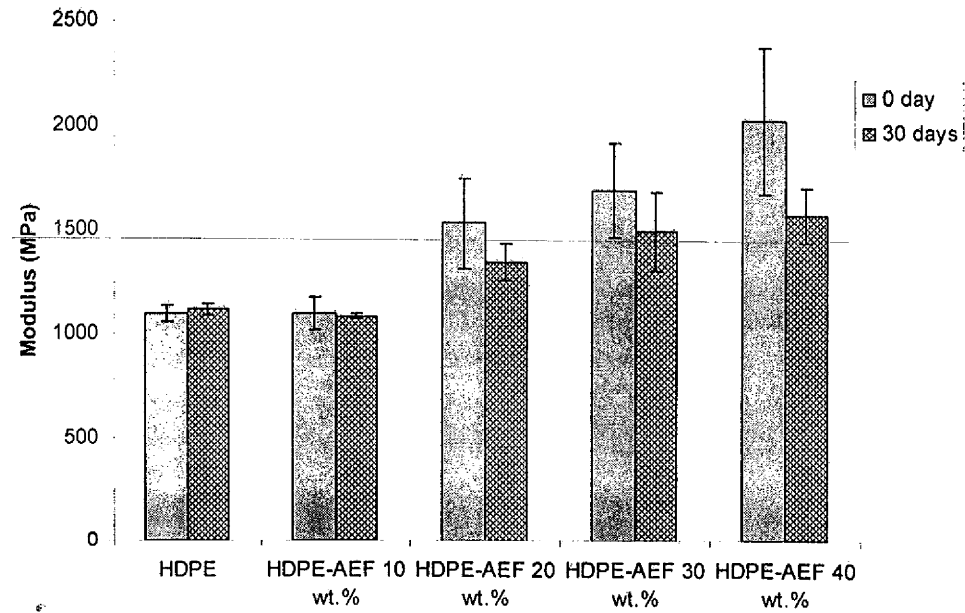


Figure 4.29 Effect of water absorption on the modulus of HDPE and HDPE-AEF composites.

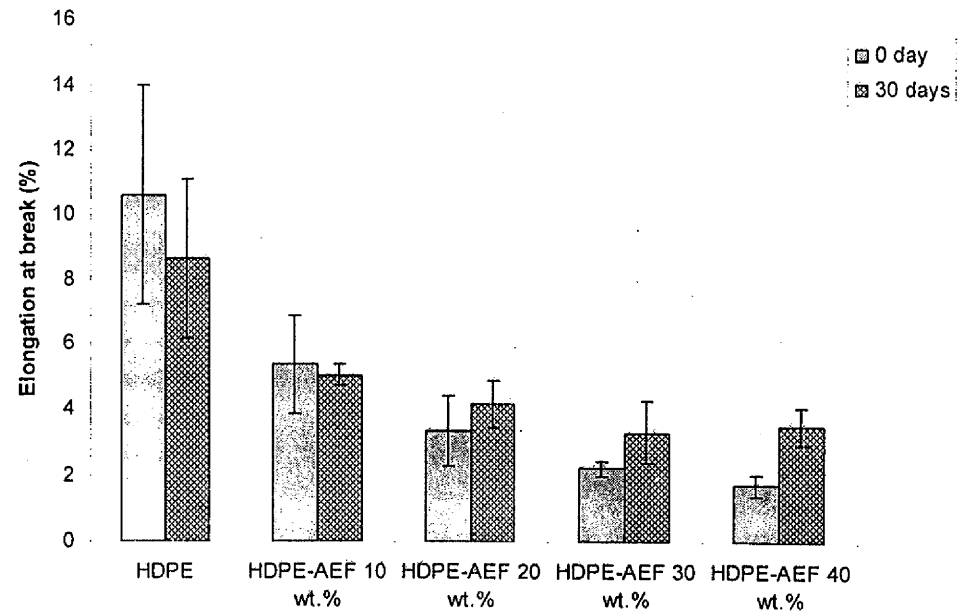


Figure 4.30 Effect of water absorption on the elongation of HDPE and HDPE-AEF composites.

4.2.2.2 Effect of acetylation

For this study, filler content of all composites was fixed at 40 wt.%. Tensile test specimens were immersed in distilled water at room temperature for 30 days. Tensile test was carried out on samples after 2, 4, and 6 days of water immersion.

4.2.2.2.1 Water uptake

Figure 4.31 shows the water uptake for HDPE, HDPE-AEF and HDPE-AAEF composites as a function of immersion time in days. Each data point represents the average of six specimens.

HDPE as seen in Figure 4.31, does not exhibit appreciable amount of water uptake after 30 days of water immersion at room temperature. This is explained by the hydrophobic nature of the material. As for HDPE-AEF and HDPE-AAEF composites, appreciable amount of water uptake are observed. The water uptake curves, all shows kind of similar patterns, i.e., linear increase in water uptake followed with a gradual increase and finally a plateau. As HDPE is hydrophobic and does not absorb water appreciably, it is then reasonable to assume that all the water was absorb by AEF and AAEF and also voids. Like other wood fibers, AEF and AAEF are also hydrophilic. This is because they too have the same cell wall polymers that can interact with water molecules via hydrogen bonding. In WPCs, voids can exist particularly at the fiber/matrix interface when the fiber is poorly wet by the matrix polymer due to inferior fiber/matrix interaction. HDPE-AEF and HDPE-AAEF composites have inferior fiber/matrix interface. This is evident from the SEM micrographs of the controls as in Figure 4.14 which show smooth pulled-out of AEF and AAEF and the existence of voids between the

fibers and the HDPE matrix. As a result, transportation of water molecules via capillary action to voids as well as flaws at the fiber/matrix interface can be expected which contributes to the total amount of water uptake by the composites.

It is also obvious from Figure 4.31 that HDPE-AEF composites reach saturation earlier (approximately after 5 days of water immersion as compared to 27 days) than HDPE-AAEF composites. Other observations are the decrease in the initial rate as well as the level of water uptake when the composites were filled with AAEF of increasing WPG. The initial rate at which water molecules were absorbed and the level of water uptake of a WPC are largely dependent on the amount of accessible hydroxyl groups that can form hydrogen bonds with water molecules. The higher the amount of these hydroxyl groups, the higher the initial rate as well as the level of water uptake. Due to acetylation between the anhydride group of acetic anhydride and the hydroxyl groups of AEF, some of these hydroxyl groups were acetylated reducing the amount of hydroxyl groups available to come in contact and interact with water molecules. With increasing WPG, higher amount of hydroxyl groups were reacted leaving fewer accessible hydroxyl groups. As a result a lower amount of water uptake as well as initial rate are observed when the composites were filled with AAEF of higher WPG. This can be understood by considering relationship between equilibrium water uptake and WPG. A linear relationship is obtained with R^2 of about 0.8624 (Figure 4.32).

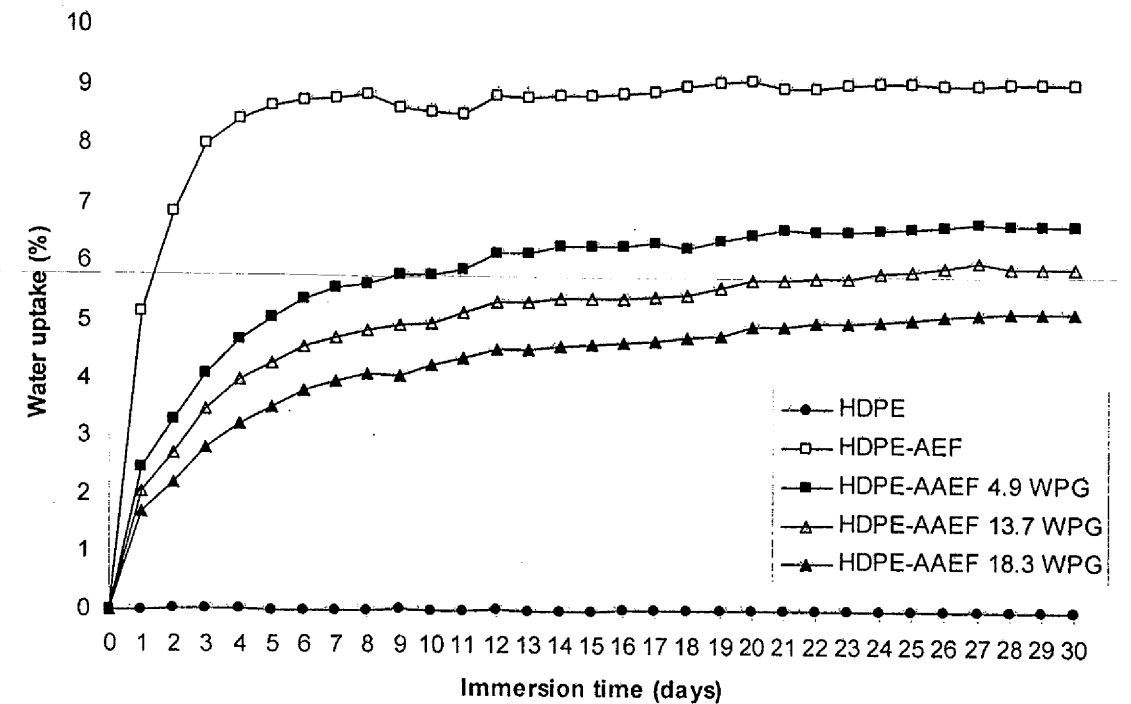


Figure 4.31 Water uptake for HDPE, HDPE-AEF and HDPE-AAEF composites immersed in water at room temperature for 30 days.

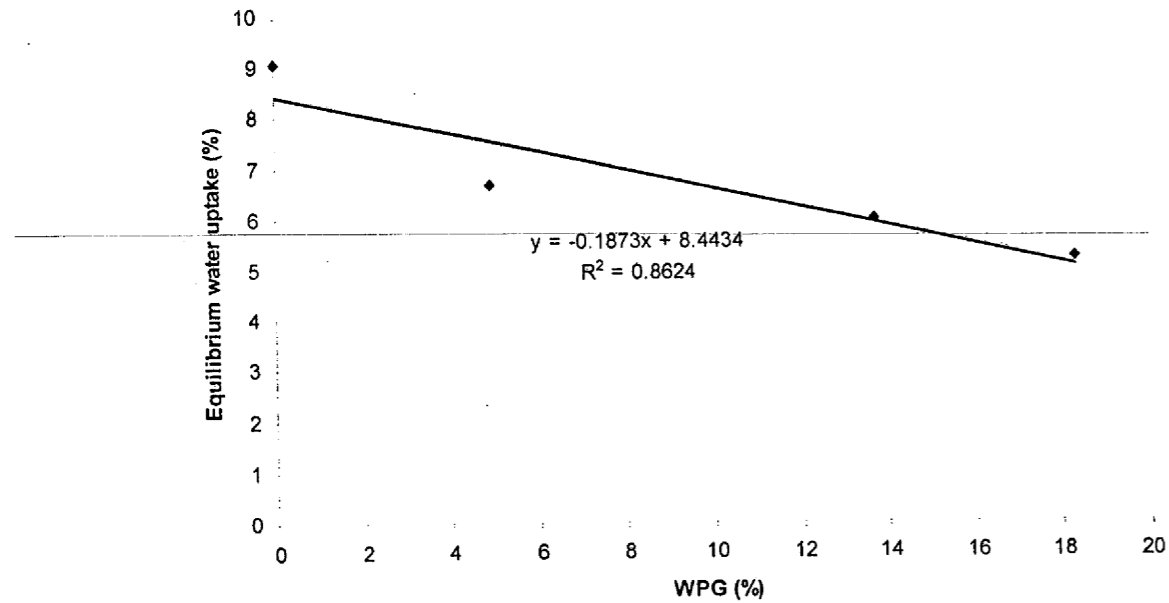


Figure 4.32 Equilibrium water uptake versus WPG for HDPE-AAEF composites.

4.2.2.2.2 Tensile properties

As seen in Figure 4.33, HDPE-AEF composites exhibit abrupt decrease of 24.9% (Table 4.7) in maximum stress after 2 days of water immersion. HDPE-AAEF composites too show a decrease in the property but the decrease is obvious only after 4 days. In order to understand the detrimental effects of water on the mechanical properties of a composite, it is necessary to investigate its effect on the constituent of the composites, i.e., the matrix, the fiber, and the fiber/matrix interface. HDPE-AEF as well as HDPE-AAEF composites have inferior fiber-matrix interaction (Figure 4.14). The decrease in maximum stress therefore could not be possibly due to degradation of the fiber/matrix interface as a result of water absorption. Too it could not be due to HDPE matrix as the material is hydrophobic. Water molecules can interact with wood fibers via hydrogen bonding resulting in swelling of the fiber cell wall. Such interaction has been

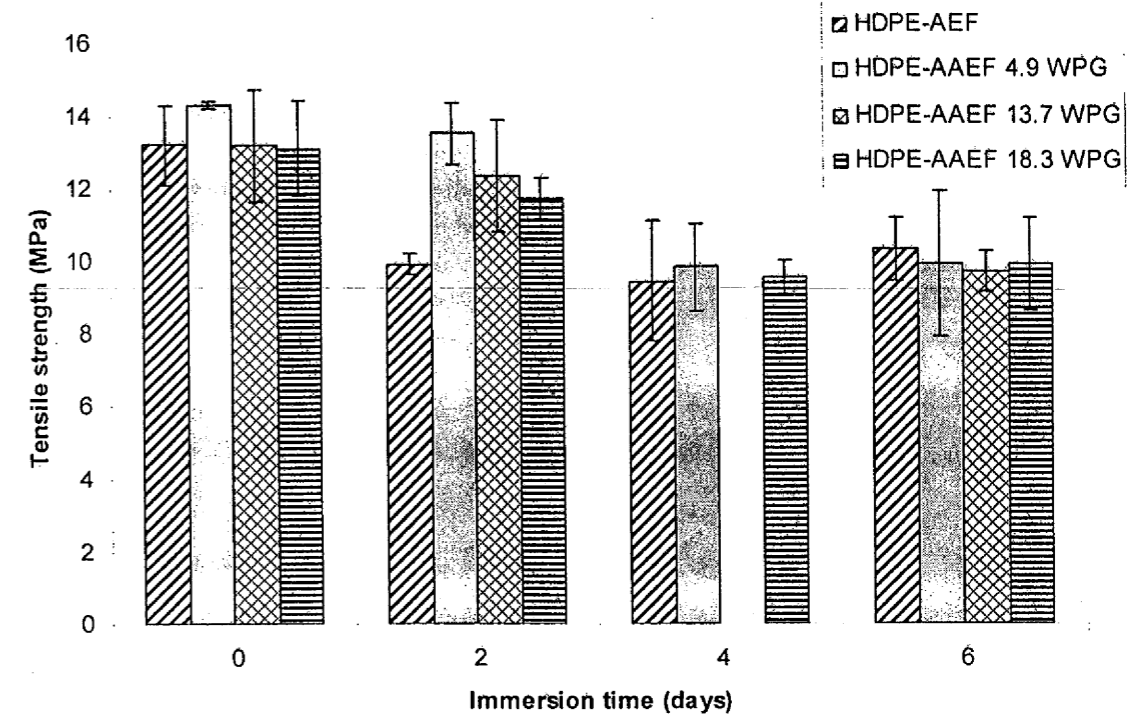


Figure 4.35 Tensile strengths of HDPE-AEF and HDPE-AAEF composites after 2, 4, and 6 days of water immersion.

Table 4.7 Percent decrease in the tensile properties of HDPE-AEF and HDPE-AAEF composites after 2, 4, and 6 days of water immersion¹.

Tensile property	Immersion time [days]	HDPE-AEF	HDPE-AAEF 4.9 WPG	HDPE-AAEF 13.7 WPG	HDPE-AAEF 18.3 WPG
Tensile strength	2	24.9	NA	NA	NA
	4	28.4	31.4	-	27.2
	6	21.8	31.0	26.3	24.4
Modulus	2	38.3	20.1	16.0	10.8
	4	46.3	30.3	-	31.73
	6	49.0	38.4	40.4	31.4

¹ Values are all in percentage.
NA = no statistical difference in the properties at alpha level of 0.05.

found to adversely affect the mechanical strength of wood fibers. This eventually would affect, i.e., decrease the strength of WPC's. The same argument could be used to explain the negative effect observed on the tensile strength of HDPE-AEF and HDPE-AAEF composites. In addition, other factors such as microcracks due to fiber swelling may also cause the property to decrease.

Although the number of accessible hydroxyl groups to interact with water molecules was reduced due to acetylation and further decreased with increasing WPG of AAEF, the decrease in maximum stress seems independent of these factors (Figure 4.33). This is probably due to the inferior fiber-matrix interaction that allows free movement of water molecules within the composite structures specifically along the fiber/matrix interface which increases the probability of water molecules to come in contact and interact with the accessible hydroxyl groups of AAEF.

As for tensile modulus (Figure 4.34), similar observation is observed as with the tensile strength. Abrupt decrease of 38.3% (Table 4.7) is observed for HDPE-AEF composites after 2 days of water immersion. As for HDPE-AAEF composites the decrease too is obvious but after 4 days of immersion in water. The tensile modulus of a composite is determined in part by the stiffness of each component, i.e., the fiber and the matrix. Since HDPE does not absorb water, it can be assumed that its stiffness is hardly affected by water absorption. The decrease in tensile modulus of HDPE-AEF and HDPE-AAEF composites therefore might be attributed to the decrease in stiffness of AEF and AAEF as a result of water absorption. It has been reported that water molecules can act as plasticizers to soften the fiber cell wall polymers. Composites filled with increasing WPG of AAEF exhibit lower decrease in tensile modulus. The higher the WPG of AAEF,

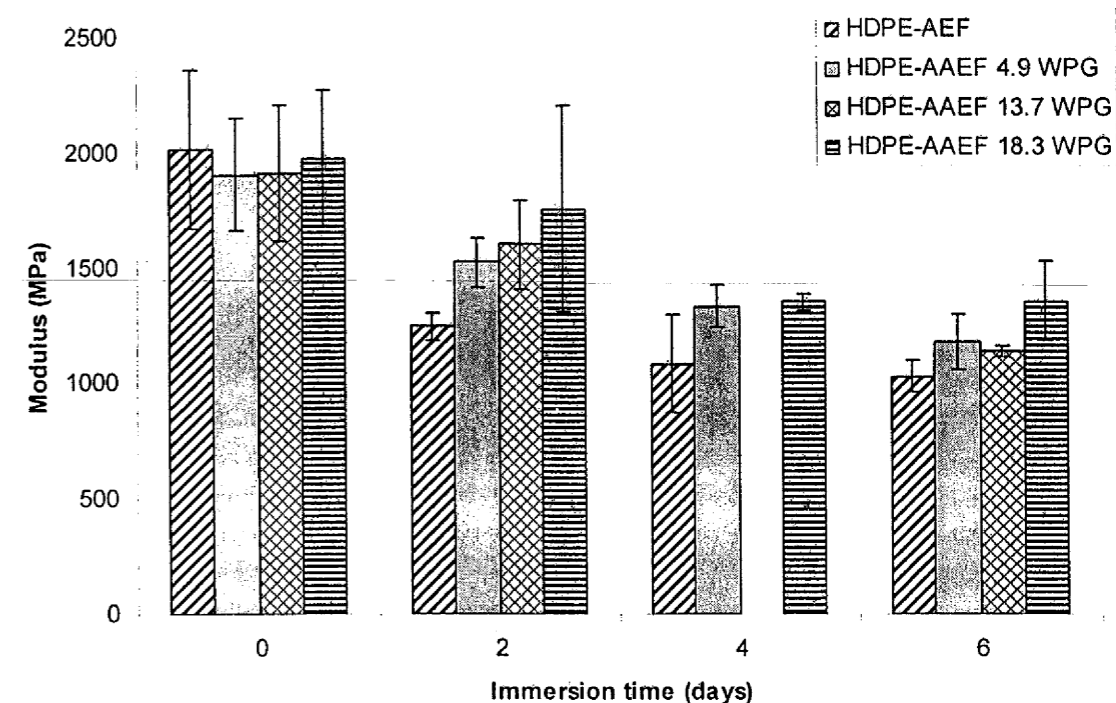


Figure 4.34 Modulus of HDPE-AEF and HDPE-AAEF composites after 2, 4, and 6 days of water immersion.

the lower the water uptake (Figure 4.31) and the lower the number of water molecules that could act as plasticizers to affect the fiber modulus.

For further understanding on the influence of water molecules on tensile modulus, percent decrease in tensile modulus was plotted over water uptake (Figure 4.35). Data were gathered from HDPE-AEF and HDPE-AAEF composites after 2 and 4 days of immersion in water. Data for after 6 days of water immersion were excluded because the change in tensile modulus after 4 and 6 days of water immersion was not obvious (Figure 4.34). Another reason was that the increase in water uptake after 6 days of immersion in water was probably more due to transportation of water molecules into voids and flaws rather than the uptake of water molecules by AEF or AAEF. From Figure 4.35, a linear

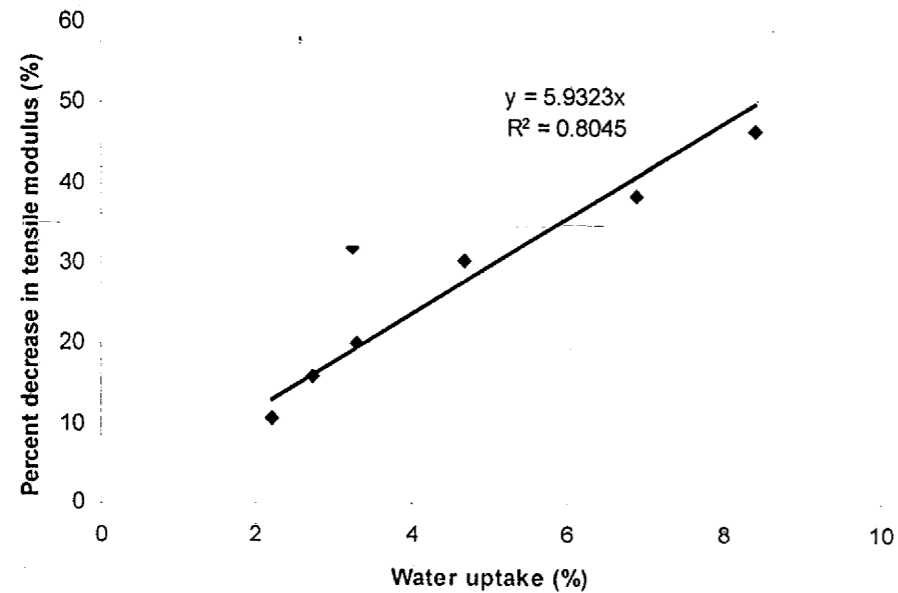


Figure 4.35 Percent decrease in tensile modulus versus water uptake.

relationship between percent decrease in tensile modulus and water uptake is observed with R^2 of about 0.8045.

Percent decrease in maximum stress and tensile modulus for HDPE-AEF and HDPE-AAEF composites are showed in Table 4.7. HDPE-AEF and HDPE-AAEF composites experience larger decrease in tensile modulus than maximum stress. This is expected as wood fiber modulus have been claimed to suffer a decrease of two-thirds or even more due to water absorption but not fiber strength, which is less affected.

Elongation at break of HDPE-AEF composites is influenced by the extent of water exposure (immersion time). This can be seen from Figure 4.36 which shows increasing values of the property with increasing immersion time. Elongation at break of HDPE-AAEF composites, however, is slightly affected without any consistent trend.

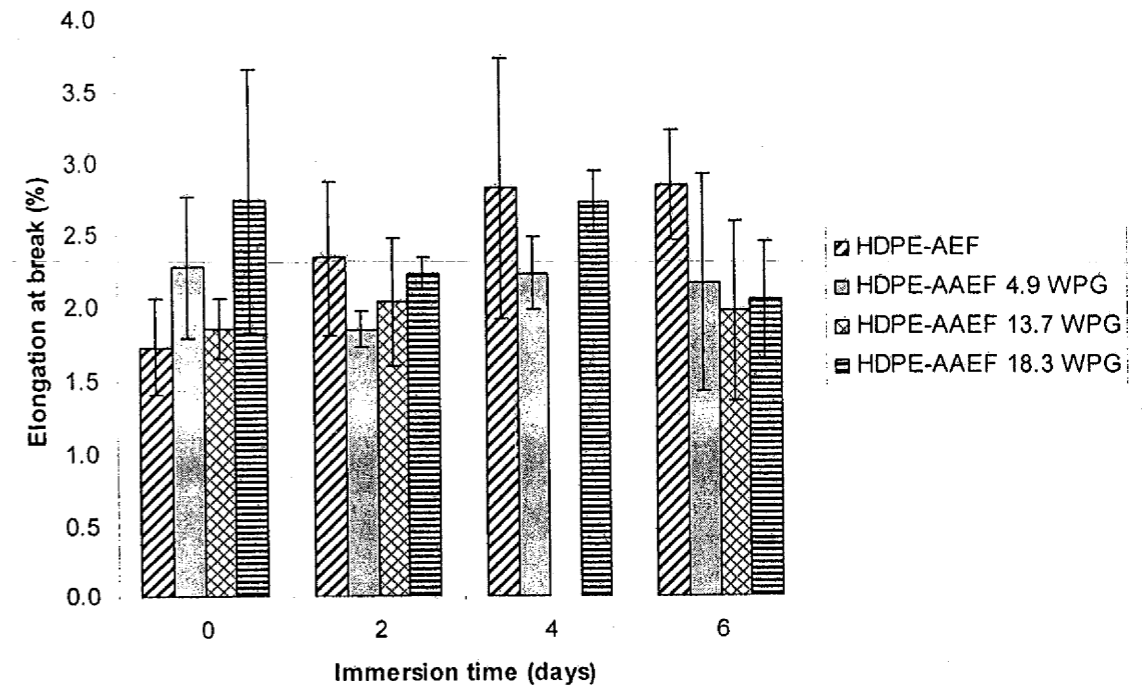


Figure 4.36 Elongation at break of HDPE-AEF and HDPE-AAEF composites after 2, 4, and 6 days of water immersion.

4.2.2.3 Effect of acetylation and compatibilizer

Water absorption study was also carried out on HDPE-AEF-MAPE and HDPE-AAEF-MAPE composites. For this study filler content was fixed at 20 wt. %.

4.2.2.3.1 Water uptake

As shown in Figure 4.37, addition of MAPE to HDPE-AEF composite has resulted in a slight decrease in water uptake of about 7% (equilibrium water uptake). The same trend is observed for HDPE-AAEF-MAPE composites. This can be seen by comparing the water uptake curve of HDPE-AAEF 13.7 WPG with that of HDPE-AAEF 13.7 WPG-MAPE composites with the percent decrease of about 44% (equilibrium water

uptake. With increasing WPG of AAEF, further decrease in water uptake is observed. MAPE can interact with free accessible hydroxyl groups of AEF or AAEF and form ester bonds. This reaction reduces the amount of free accessible hydroxyl groups for these composites. As a result a decrease in water uptake especially the equilibrium water uptake is expected. The formation of ester bonds too eliminates voids at the fiber-matrix interface. This restricts movement of water molecules along the fiber-matrix interface to penetrate deeper inside the composite structures and limits the probability of the water molecules to come in contact and interact with the free accessible hydroxyl groups of AAEF or AAEF.

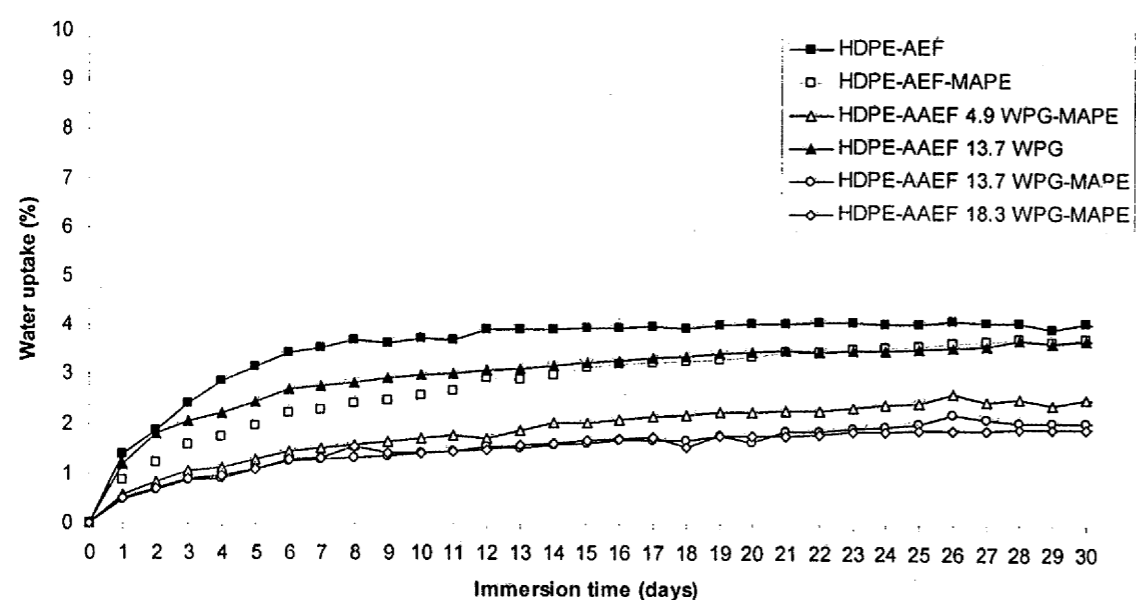


Figure 4.37 Effect of compatibilizer on the water uptake for HDPE, HDPE-AEF and HDPE-AAEF composites immersed in water at room temperature for 30 days.

With increasing WPG of AAEF, only a slight decrease in water uptake is observed with the addition of MAPE. This is probably due to the limited interaction of MAPE with the free accessible hydroxyl groups AAEF as discussed in 4.2.1.3. At high WPG of AAEF, MAPE has been shown to hardly promote fiber-matrix interaction between AAEF and HDPE (Figure 4.19). Because of this the amount of free accessible hydroxyl groups of AAEF are not much reduced and the inferior fiber-matrix interface facilitates the movement of water molecules to come in contact and form hydrogen bonding with these hydroxyl groups.

4.2.2.3.2 Tensile properties

After 30 days of water absorption, all composite samples were tested for tensile properties. It is seen from Figure 4.38 that composites with MAPE show higher tensile strength values than composites without MAPE. This is clear by comparing HDPE-AAEF 13.7 WPG with HDPE-AAEF 13.7 WPG-MAPE composites. The property, however, is increasingly lower when the composite is filled with AAEF of higher WPG.

As for the modulus, the effect of adding MAPE to HDPE-AEF and HDPE-AAEF 13.7 WPG is not obvious (Figure 4.39). Among HDPE-AAEF-MAPE composites, HDPE-AAEF 4.9 WPG-MAPE shows the highest value of the modulus after 30 days of water immersion. As for elongation at break, the effect of adding MAPE is negligible (Figure 4.40).

Based on water uptake behavior and the tensile properties after 30 days of water immersion, it can be concluded that HDPE-AAEF 4.9 WPG-MAPE composite performed the best. This however confirmed at filler loading of 20 wt.%.

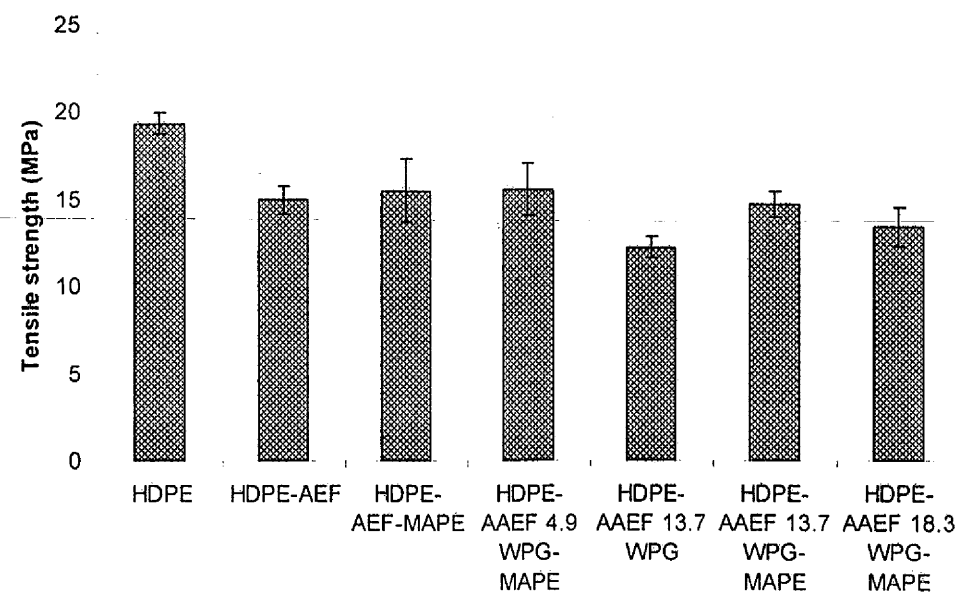


Figure 4.38 Effect of compatibilizer on the tensile strength values of HDPE, HDPE-AEF, and HDPE-AAEF composites after 30 days of water absorption.

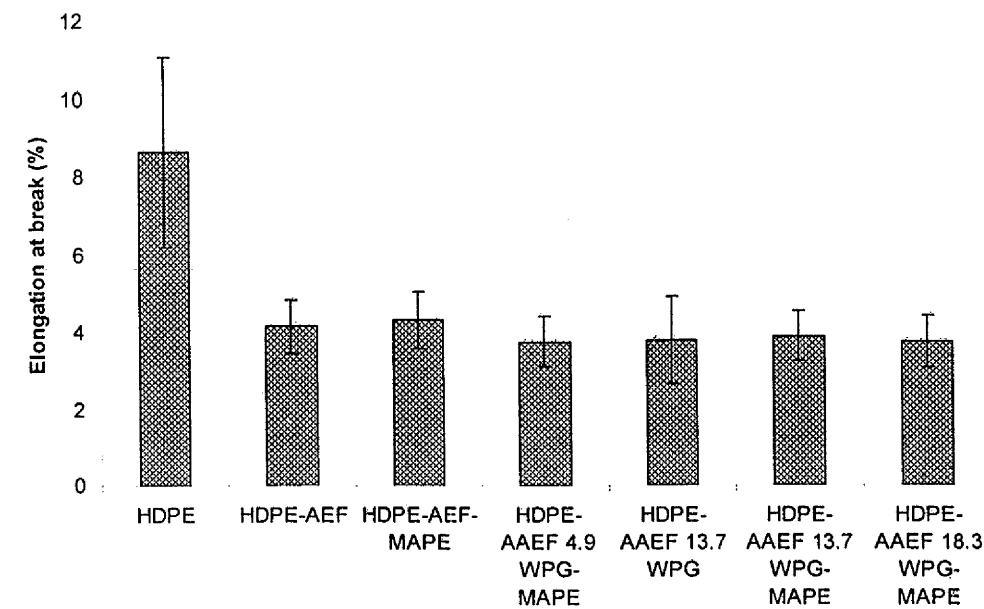


Figure 4.40 Effect of compatibilizer on the elongation at break values of HDPE, HDPE-AEF, and HDPE-AAEF composites after 30 days of water absorption.

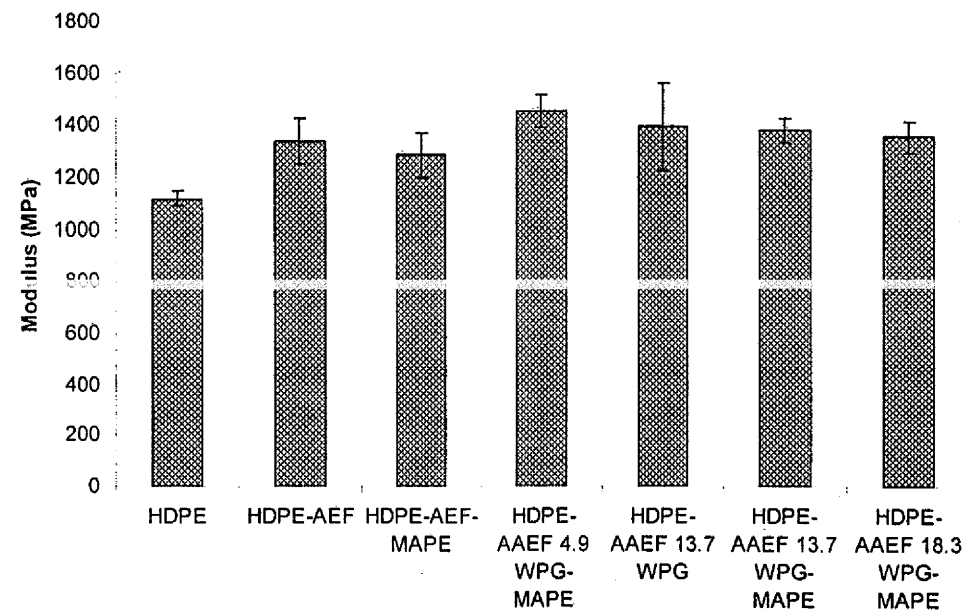


Figure 4.39 Effect of compatibilizer on the modulus values of HDPE, HDPE-AEF, and HDPE-AAEF composites after 30 days of water absorption.

5.0 CONCLUSIONS

In designing high water resistance wood-thermoplastic composites, absorption of water by the wood fiber as well the fiber-water interaction are among the important factors that need to be considered. Absorption of water by the wood fiber can adversely affect the fiber properties. As for fiber-matrix interaction, it can be deteriorated because of water absorption by the composite system. Acetylated AEF (AAEF) did not interact well with HDPE and this had resulted in a decrease in the tensile properties of the composites. Addition of MAPE did not help in improving the tensile strength particularly for composites filled with AAEF of higher WPG. However, a significant improvement in the property was observed when AEF was used instead of AAEF in the compatibilized composite systems. Absorption of water by HDPE-AEF composites was due to

hydrophilic nature of AEF. In this work although AAEF did reduce the amount of water absorbed by the composite systems, the composites showed poor retention of the tensile properties as water molecules are free to move within the composite structures (because of poor fiber-matrix interface) to interact with the accessible hydroxyl groups of AAEF. For compatibilized HDPE-AAEF composite systems (filler loading at 20 wt.%), the best is HDPE-AAEF 4.9 WPG-MAPE composite as the composite shows the lowest uptake of water and the highest tensile properties after 30 days of water immersion.

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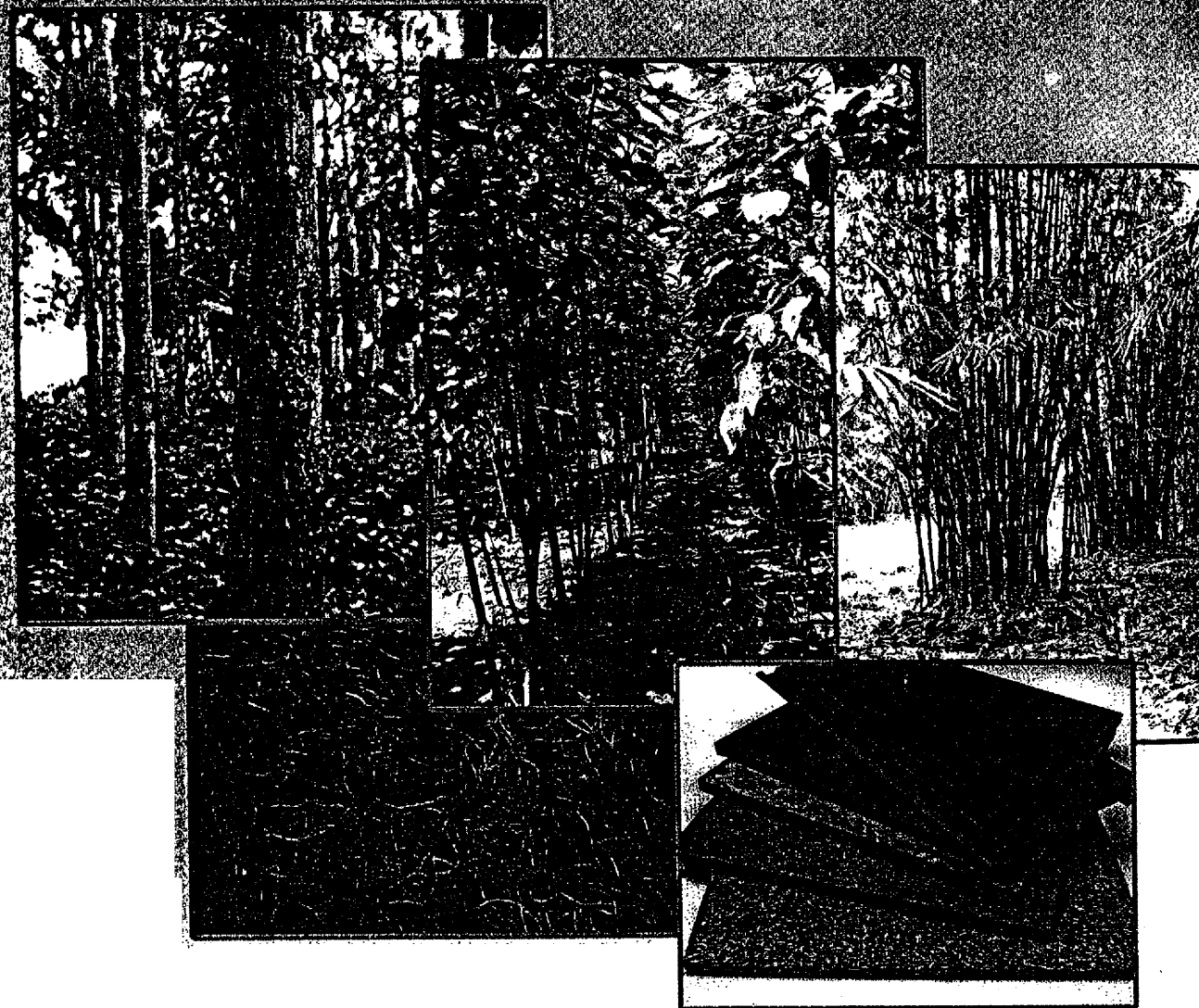
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Effect of Immersion Time in Water on the Tensile Properties of Acetylated Steam-exploded *Acacia mangium* Fibers Filled Polyethylene Composites

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ABSTRACT

Steam-exploded fibers were derived from steam explosion of *Acacia mangium* wood chips. The fibers, after undergoing water and alkali extractions (AEF) were acetylated with acetic anhydride. Weight percent gain (WPG) of the acetylated fibers (AAEF) were 4.9, 13.7, and 18.3. Composites of high-density polyethylene (HDPE) and acetylated steam-exploded *Acacia mangium* fibers were prepared using 2-roll mill and compression molding. The composites (tensile specimens) were immersed in water at room temperature for 30 days. All composites absorbed water but the water uptake was found to decrease with increasing WPG of AAEF fibers. Acetylation had decreased the number of accessible hydroxyl groups of AAEF fibers to interact with water molecules. The tensile properties of the immersed samples were tested after 2, 4, and 6 days of water immersion. The tensile properties of HDPE-AEF and HDPE-AAEF composites were observed to decrease with immersion time. HDPE-AEF composites showed a reduction in maximum stress and tensile modulus after 2 days of water immersions. HDPE-AAEF composites, on the other hand, showed a distinct decrease in both properties after 4 days of water immersion. All composites exhibited higher percent decrease in tensile modulus than maximum stress. The decrease in tensile modulus in particular was lower for composites filled with AAEF fibers of higher WPG. Tensile modulus too was found to decrease linearly ($R^2=0.80$) with increasing water uptake of the composite system. Acetylation did not improve fiber-matrix interaction but did reduce the amount of water absorbed by the composite system.

Keywords: steam-exploded fiber, acetylation, polyethylene, water absorption, tensile properties, immersion time

INTRODUCTION

Wood fibers are candidates for use as fillers or reinforcement to replace traditional inorganic fillers or fibers for thermoplastic matrices. The advantages of using wood fibers include low volumetric cost, low specific density, renewable and biodegradable. Wood fibers, however, are hydrophilic. The cell wall polymers, i.e. cellulose, hemicelluloses, and lignin, have hydroxyl and oxygen containing groups that can interact with water molecules via hydrogen bonding (Rowell *et al* 1993). Fiber-water interaction leads to fiber swelling. Fiber swelling will result in dimensional instability of composites filled with wood fibers (wood-plastic composites, WPCs). Fiber-water interaction too has been reported to adversely affect the mechanical properties of wood fibers (Winandy and Rowell 1984). As a result a decrease in the mechanical properties of WPCs can be expected. In order to decrease the water absorption of wood fibers, cell wall polymers (i.e. hemicelluloses and lignin) that are mainly responsible for the property can be removed to some extent via steam explosion treatment

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followed by water and alkali extractions (Glasser *et al* 1999). Water absorption of wood fibers can also be reduced by reducing the number of accessible hydroxyl groups to interact with water molecules (Mat Taib *et al* 2006). This can be achieved through chemical surface modification of wood fibers with chemicals such as acetic anhydride. WPCs can also exhibit inferior mechanical properties due to the degrading impact of water molecules on fiber-matrix interface.

In this study, both steam-explosion and acetylation were used to produce wood fibers of improved water resistance. The fibers were melt-blended with high density polyethylene. The water absorption behavior was determined. The reduction in tensile properties of the composites was followed after 2, 4, and 6 days of water immersion.

EXPERIMENTAL

Materials

High density polyethylene (HDPE), (ETILINAS HD5218AA from POLYETHYLENE MALAYSIA Sdn. Bhd.) was used as the polymer matrix. Its density and melt flow index were specified as 0.952 g/cm³ and 18 g/10min (230°C/2.16 kg), respectively.

Steam-exploded fibers (AEF) were generated via steam explosion of *Acacia mangium* wood chips at a severity of log R_o 4.23. The severity factor is defined elsewhere (Overend and Chronet 1987). The fibers, after undergoing water and alkali extractions were acetylated with acetic anhydride at 109°C for 6, 24, and 72 h. Acetylated fibers (AAEF) of 4.9, 13.7 and 18.3 WPG were produced, respectively. AAEF fibers then were continuously washed with water, oven-dried at 105°C for 24 hours and stored over a silica gel desiccant until use.

Compounding and molding

HDPE and AAEF fibers were melt-blended using 2-roll mill at 160°C. The proportion of AAEF fibers was maintained at 40 weight percent. Following compounding, the composite sheets were prepared by melt pressing chips of compounded material on a hot press at 160°C and 14.3 MPa for 10 minutes to form 1 mm thick plates. In addition, composites of HDPE and unreacted steam-exploded fibers (AEF) were also prepared for comparison.

Water absorption study

Tensile specimens were cut from the compression molded plates and used for the measurements of water absorption. After being vacuum dried at 70°C to a constant weight, the specimens were immersed in an immersion tank filled with deionized water at room temperature for 30 days. The weights of the specimens were determined at regular intervals using an analytical balance and the water uptake at any time (W_t) was calculated according to the following equation:

$$\text{Water uptake, } W_t(\%) = \frac{W_1 - W_2}{W_2} \times 100$$

where W_t and W_2 are the weight after the exposure time t and the dry weight, respectively.

Tensile test

Tensile specimens were retrieved after 2, 4, and 6 days of water immersion and tested for tensile properties. The tensile tests were performed at room temperature according to ASTM D 638, using a computerized laboratory tensile tester (M500 Tensometric AZ), at a crosshead speed of 1.5 mm/min. Tensile properties for unaged (control) specimens were

also determined for comparison. A minimum of six samples of each composite system were tested to obtain an average value.

RESULTS AND DISCUSSION

Water uptake curve

Figure 1 shows the water uptake for HDPE, HDPE-AEF and HDPE-AAEF composites as a function of immersion time in days. Each data point represents the average of six specimens.

HDPE as seen in Figure 1, did not exhibit appreciable amount of water uptake after 30 days of water immersion at room temperature. This is explained by the hydrophobic nature of the material. As for HDPE-AEF and HDPE-AAEF composites, appreciable amount of water uptake were observed. As HDPE did not absorb water, it is then reasonable to assume that all the water was absorbed by AEF and AAEF fibers and also voids. AEF and AAEF fibers have hydroxyl groups that can interact with water molecules via hydrogen bonding. It is also obvious from the figure that the initial rates and levels of water uptake were lower for composites filled with AAEF fibers of increasing WPG. This can be explained as follows. Due to acetylation between the anhydride group of acetic anhydride and the hydroxyl groups of AEF fibers, some of these hydroxyl groups were acetylated reducing the amount of free hydroxyl groups available to interact with water molecules. With increasing WPG, higher amount of hydroxyl groups get acetylated and fewer hydroxyl groups are available. This consequently results in a lower amount of water uptake as well as its initial rate.

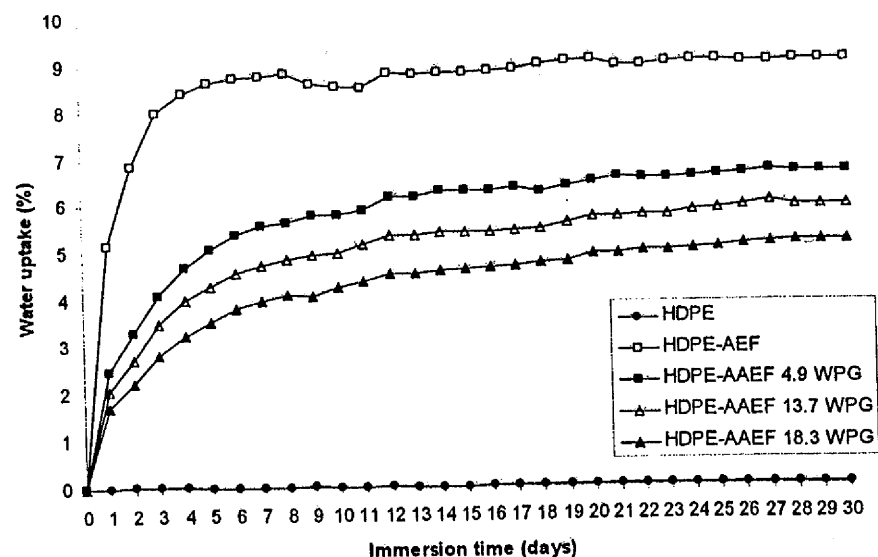


Figure 1. Water uptake for HDPE and HDPE-AAEF composites immersed in water at room temperature for 30 days

Tensile strength

As shown in Figure 2, HDPE-AEF composites exhibited an abrupt decrease of 24.9% (Table 1) in maximum stress after 2 days of water immersion. HDPE-AAEF composites too showed a decrease in the property but the decrease was obvious only after 4 days. HDPE-AEF as well as HDPE-AAEF composites were found to show inferior fiber-matrix interaction (Figure 3). The decrease in maximum stress therefore could not be possibly due to the degradation of the interface as a result of water absorption. Too it could not be due to HDPE matrix as the material is hydrophobic. Water molecules can interact with wood fibers via hydrogen bonding which resulted in fiber swelling. In composites, fiber swelling can lead to the

formation of microcracks that adversely affect composite properties including maximum stress. This explains the decrease observed in maximum stress for HDPE-AEF and HDPE-AAEF composites. Although the number of accessible hydroxyl groups to interact with water molecules is reduced due to acetylation and further decrease with increasing WPG of AAEF fibers, the decrease in maximum stress seems independent of these factors (Figure 2). This is probably due to the inferior fiber-matrix interface that allows free movement of water molecules within the composite structure. This consequently increases the probability of water molecules to interact with the accessible hydroxyl groups of AAEF fibers.

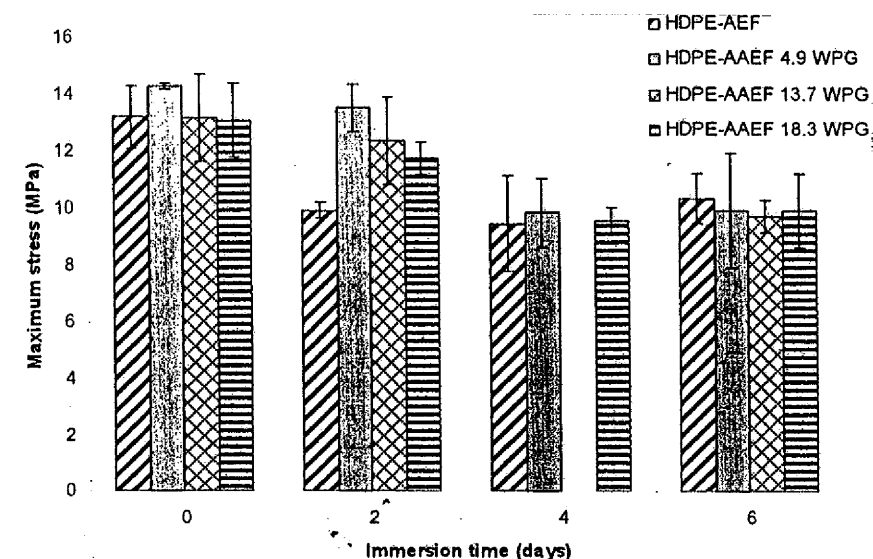


Figure 2. Maximum stress of HDPE-AEF and HDPE-AAEF composites after 2, 4, and 6 days of water immersion

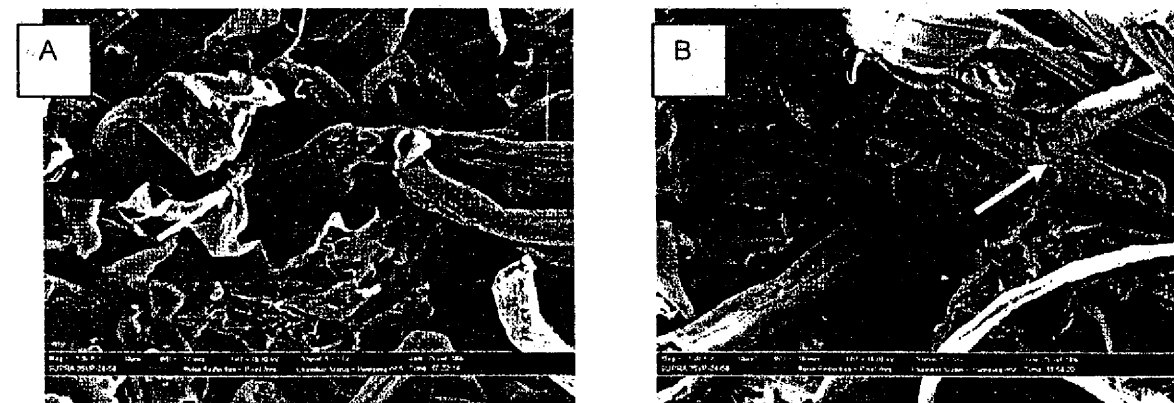


Figure 3. SEM micrographs of the tensile fracture surface of HDPE-AEF (A) and HDPE-AAEF 18.3 WPG (B) composites. The arrows show void that exist around AEF and AAEF fibers suggesting inferior fiber-matrix interaction

As for tensile modulus (Figure 4), similar observation was observed as with the maximum stress. An abrupt decrease of 38.3% (Table 1) was observed for HDPE-AEF composites after 2 days of water immersion. As for HDPE-AAEF composites the decrease too was obvious but after 4 days of immersion in water. The decrease in tensile modulus can be attributed to plasticizing effect of water on AEF and AAEF fibers. Composites filled with increasing WPG of AAEF fibers exhibited smaller decrease in tensile modulus. The higher the WPG of AAEF fibers, the lower the water uptake and the lower the number of water

molecules that could act as plasticizers to affect the fiber modulus. For further understanding on the influence of water molecules on tensile modulus, the percent decrease in tensile modulus was plotted over water uptake (Figure 5). Data were gathered from HDPE-AEF and HDPE-AAEF composites after 2 and 4 days of immersion in water. Data for after 6 days of water immersion were excluded because the change in tensile modulus after 4 and 6 days of water immersion was not obvious (Figure 4). Another reason was that the increase in water uptake after 6 days of immersion in water was probably more due to the transportation of water molecules into voids rather than uptake of water molecules by AEF or AAEF fibers. From Figure 5, a linear relationship between percent decrease in tensile modulus and water uptake was observed with R^2 of about 0.80.

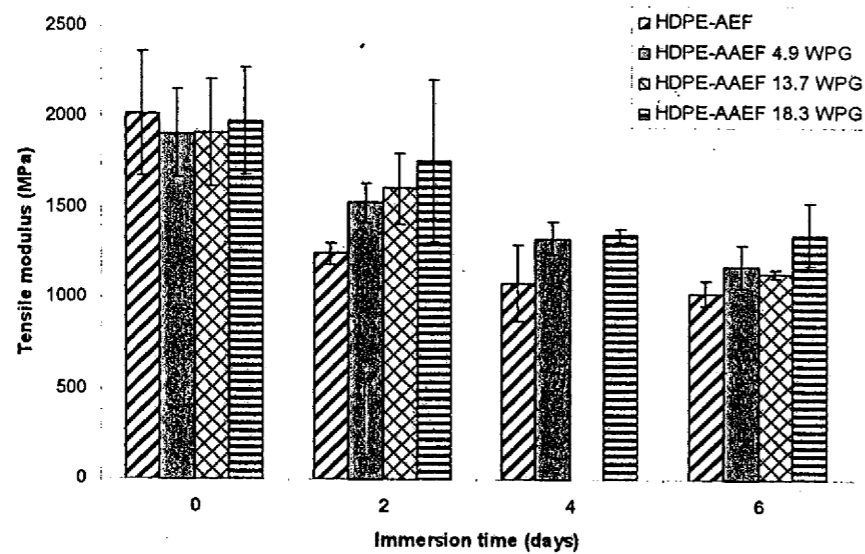


Figure 4. Tensile modulus of HDPE-AEF and HDPE-AAEF composites after 2, 4, and 6 days of water immersion

The percent decrease in maximum stress and tensile modulus for HDPE-AEF and HDPE-AAEF composites are shown in Table 1. HDPE-AEF and HDPE-AAEF composites experienced larger decrease in tensile modulus than maximum stress. This is expected as wood fiber modulus has been claimed to suffer a decrease of two-thirds or even more due to water absorption but not fiber strength, which is less affected (Zadorecki and Flodin 1985).

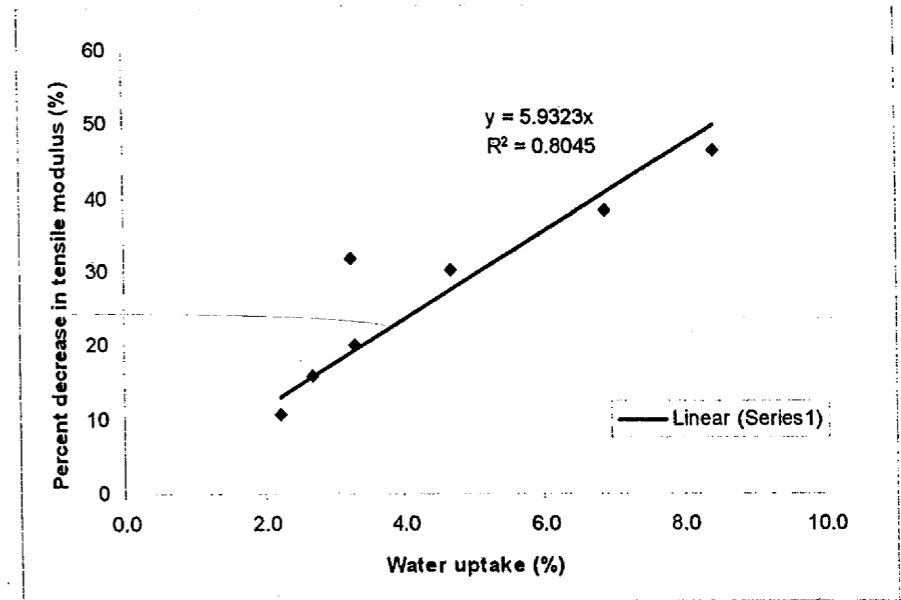


Figure 5. Percent decrease in tensile modulus versus water uptake

Table 1. Percent decrease in tensile properties of HDPE-AEF and HDPE-AAEF composites water exposure¹.

Tensile property	Immersion time [days]	HDPE-AEF	HDPE-AAEF 4.9 WPG	HDPE-AAEF 13.7 WPG	HDPE-AAEF 18.3 WPG
Maximum stress	2	24.9	NA	NA	NA
	4	28.4	31.4	-	27.2
	6	21.8	31.0	26.3	24.4
Tensile modulus	2	38.3	20.1	16.0	10.8
	4	46.3	30.3	-	31.73
	6	49.0	38.4	40.4	31.4

¹ Values are all in percentage.
NA = no statistical difference in the properties at alpha level of 0.05.

The elongation at break of HDPE-AEF composites was influenced by the extent of water exposure (immersion time). This can be seen from Figure 6 which shows increasing values of the property with increasing immersion time. Elongation at break of HDPE-AAEF composites, however, was only slightly affected.

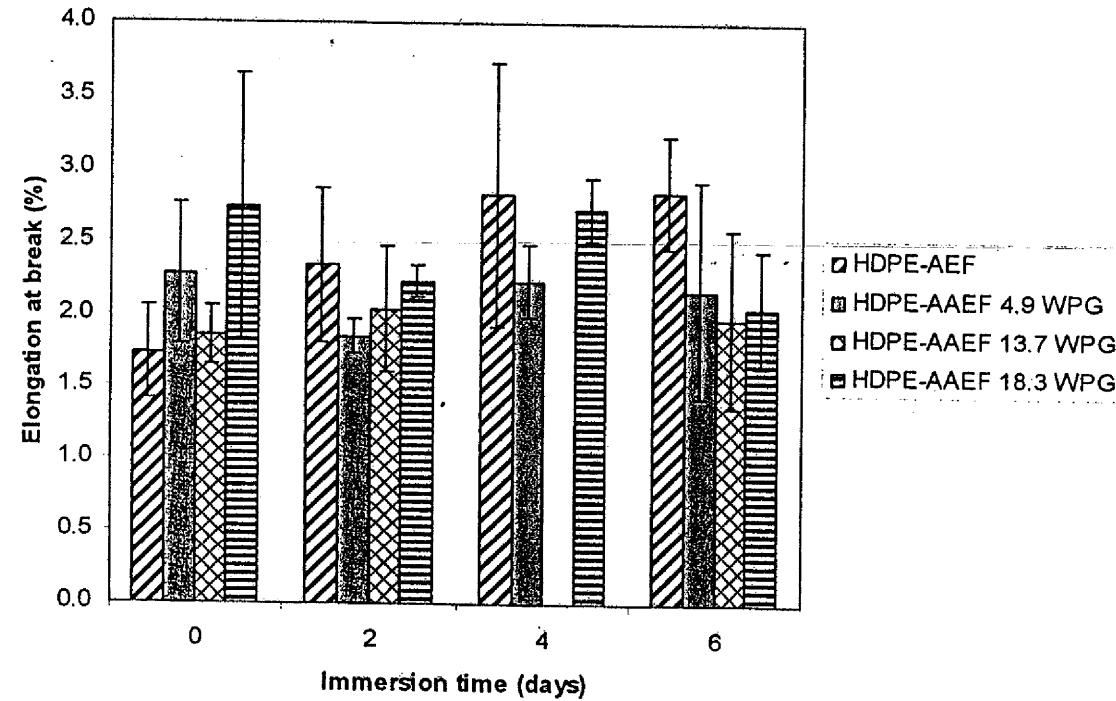


Figure 6. Elongation at break of HDPE-AEF and HDPE-AAEF composites after 2, 4, and 6 days of water immersion

CONCLUSION

This study examined the water uptake behavior of HDPE-AEF and HDPE-AAEF composites. All composites absorbed water but the water uptake was found to decrease with increasing WPG of AAEF fibers. Tensile properties of HDPE-AEF and HDPE-AAEF composites were determined after 2, 4, and 6 days of water immersion. The properties were found to adversely affected by water absorption. Maximum stress was less affected than tensile modulus. Composites filled with AAEF fibers of increasing WPG showed a lower decrease in tensile modulus. Linear relationship between percent decrease in tensile modulus and water uptake was observed.

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