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TAJUK PROJEK : **MECHANICAL PROPERTIES OF OIL PALM EMPTY FRUIT BUNCH IMPACT-MODIFIED UNPLASTICISED POLY(VINYL CHLORIDE) COMPOSITES : EFFECT OF FILLER CONTENT AND PROCESSING CONDITIONS**

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MECHANICAL PROPERTIES OFOIL PALM EMPTY FRUIT BUNCH IMPACT-MODIFIED UNPLASTICISED POLY(VINYL CHLORIDE) COMPOSITES: EFFECT OF FILLER CONTENT AND PROCESSING CONDITIONS

(SIFAT MEKANIK KOMPOSIT BERTERTULANG PENGISI TANDAN BUAH KELAPA SAWIT: KESAN KANDUNGAN PENGISI DAN KEADAAN PEMPROSESAN)

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ABSTRACT

MECHANICAL PROPERTIES OFOIL PLAM EMPTY FRUIT BUNCH IMPACT-MODIFIED UNPLASTICISED POLY(VINYL CHLORIDE) COMPOSITES: EFFECT OF FILLER CONTENT AND PROCESSING CONDITIONS

(*Keywords : Mechanical properties, composites, PVC, EFB filler, screw speed, processing temperature, fusion*)

Interest in the use of natural fibers as fillers has grown over the past few years due to the advantages it offers. The use of oil palm empty fruit bunch (EFB) as fillers in the unplasticised poly (vinyl chloride) (PVC-U) is a new attraction in the polymer composite technology. The objectives of the study is to investigate the effects of EFB fillers, screw speeds and processing temperature on processability and mechanical properties of filled and impact modified PVC-U composite extrudates.

To produce the composite extrudates, the PVC resin and the additives were firstly dry blended using high speed mixer before being extruded into sheets using single screw extruder. Test specimens were prepared and shaped using cutter machine after which impact, flexural and tensile properties were determined. The results showed that only EFB-filled PVC-U composite incorporated with 10 phr of filler can be extruded with the smoother surface whereas at 20 phr the extrudates surfaces changed to be rough.

The impact strength of acrylic-impact modified and EFB-filled acrylic-impact modified extrudates increased while CPE-impact modified and EFB-filled extrudates decreased as the screw speed increased. Meanwhile the flexural properties extrudates decreased as the screw speed increased. In terms of temperature effects, the maximum impact strength of impact modified occurred at temperature of 180°C while EFB-filled and EFB-filled acrylic-impact modified occurred at 185°C. The flexural modulus slightly increased while the flexural strength slightly decreased with the increase of temperature. The yield stress and yield strain of extrudates were insignificantly affected by screw speeds but slightly increased as temperature increased. The yield stress, yield strain and fusion level of extrudates increased with temperature. The overall results showed that the temperature more influence the processability and mechanical properties of both the unmodified of and impact modified extrudates compared to screw speed.

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ABSTRAK

SIFAT MEKANIK KOMPOSIT BERTERTULANG PENGISI TANDAN BUAH KELAPA SAWIT: KESAN KANDUNGAN PENGISI DAN KEADAAN PEMPROSESAN

(Katakunci: Sifat mekanik,komposit,PVC, pengisi EFB, halaju skru, suhu pemprosesan, pelakuran)

Kecenderungan menggunakan gentian semulajadi sebagai pengisi berkembang berapa tahun kebelakangan ini disebabkan oleh kelebihan yang ada pada gentian ini. Penggunaan tandan kelapa sawit (EFB) sebagai pengisi di dalam poli(vinil klorida) tanpa pemplastik(PVC-U) adalah merupakan satu tarikan baru pada teknologi komposit polimer. Objektif kajian adalah untuk menyelidik kesan pengisi EFB, halaju skru dan suhu pemprosesan terhadap kebolehanproses dan sifat mekanik komposit PVC-U berpengsisi dan komposit PVC-U terubahsuai-impak.

Untuk penghasilan komposit, resin PVC and bahan tambah telah terlebih dahulu diadun dengan menggunakan pencampur berkelajuan tinggi sebelum di semperit ke bentuk kepingan dengan menggunakan penyemperit satu skru. Spesimen ujian telah dibentuk dan disediakan dengan menggunakan alat pemotong yang mana selepas itu sifat impak, lenturan dan regangan ditentukan. Keputusan telah menunjukan komposit terisi-EFB dengan 10 phr hanya yang boleh disemperit dengan permukaan yang licin, sebaliknya pada 20 phr, permukaan semperit (komposit) telah bertukar menjadi kasar.

Kekuatan impak terubahsuai-impak akrilik dan terisi-EFB terubahsuai-impak akrilik meningkat, sementara impak terubahsuai-CPE and terisi-EFB menurun apabila halaju skru meningkat. Sementara itu sifat lenturan menurun apabila halaju skru meningkat. Kesan suhu pula, kekuatan impak maksimum terubahsuai-impak telah berlaku pada suhu 180°C sementara terisi-EFB terubahsuai impak-akrilik telah berlaku pada 185°C. Modulus lenturan sedikit meningkat sementara kekuatan lenturan sedikit menurun apabila suhu bertambah. Tegasan alah dan tarikan alah adalah tidak banyak dipengaruhi oleh halaju skru tetapi meningkat sedikit apabila suhu bertambah. Tegasan alah, terikan alah dan paras perlakuran meningkat dengan pertambahan suhu. Keseluruhan keputusan telah menunjukkan bahawa kebolehanproses dan sifat mekanik adalah lebih dipengaruhi oleh suhu berbanding halaju skru.

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LIST OF ABBREVIATIONS AND SYMBOLS

PVC	-	Poly(vinyl chloride)
PVC-U	-	Unplasticised Poly(vinyl chloride)
EFB	-	Oil Palm Empty Fruit Bunch
CPE	-	Chlorinated Polyethylene
E	-	Tensile Modulus
G	-	Shear Modulus
E_B	-	Flexural Modulus
K	-	Compression Modulus
GPa	-	Giga Pascal
MPa	-	Mega Pascal
$g.cm^{-}3$	-	gram per centimeter cube
$W.m^{-1}.K^{-1}$	.	Watt per meter per Kelvin
μm	-	micron meter
N	-	Newton
rpm	-	revolution per min
phr	-	part per hundred resin
$CaC0_3$	.	Calcium Carbonate
UV	-	Ultraviolet
MBS	-	Methacrylate-Butadiene Styrene
ACR	-	All acrylic
RHA	-	Rice Husk Ash
ABS	-	Acrylonitrile Butadiene Styrene
HDPE	-	High Density Polyethylene
PP	-	Polypropylene
PMMA	-	Poly(methyl methacrylate)
CaSt	-	Calcium streate

HSt - Stearic Acid

PA - Processing Aid

TiO_2 - Titanium Dioxide

Sn - Tin

W - Ultimate Failure Load

L - Span Length

ΔW - Load Increment

ΔS - Deflection Increment

σ_y - Yield Stress

ε - Strain Rate

A - Constant

ΔH - Activation Energy

k - Boltzmann's constant

γ - Stress Concentration Factor

T - Absolute Temperature

v - Impact velocity

d - Mean thickness of the specimens

CHAPTER 1

INTRODUCTION

1.1 Overview

Poly (vinyl chloride) (PVC) has grown into one of the major thermoplastics material since it was first produced in the 1930's. More than one-half of the PVC that is produced is used in the building industry (Mengeloglu, 2001). Because of its blending versatility (ability to be modified through the introduction of various additives) and cost-effectiveness, PVC is one of the most worldwide used thermoplastics and its plays an important role in the plastic industry as a major component of composite manufacturing (Wenguang and La Mantia, 1996)

Cost reduction is achieved when fillers are incorporated into the PVC matrix. In addition to reducing cost, other typical reasons for using fillers are stiffness enhancement and thermal properties alteration of vinyl products. Currently, the most important fillers for PVC are inorganic materials such as glass fibre, calcium carbonate, agalmatolite, talc (Saad, *et al.* 1999, Wee, 2001, Pedro, *et al.* 2001, Xie, *et al.* 2001). These high-density inorganic fillers offer wide property changes in the composites but on a volumetric basis, their use is not cost-effective. In addition, these inorganic fillers cause wear of equipment during processing. Therefore the use of natural fillers such as wood has been suggested as an alternative to mineral materials (Nabi Saheb and Jog, 1999).

Interest in the use of natural fillers has grown during the last decade and most of the natural fibres used in thermoplastics composites are derived from wood. The use of natural fillers in USA plastic industry in 2000 was estimated 0.4 billion pounds (7%) (Gurram, *et al.* 2002). In fact, industry experts believe that the demand for natural fillers for plastics composite applications will grow at least six-fold in the next 5 to 7 years (Stokke, 2002). The rationale behind this interest is that the use of natural fibres offers several benefits, including lower cost, less abrasiveness to equipment, high specific properties such as higher specific modulus (modulus/specific gravity), renewable nature and biodegradability (Nabi Saheb and Jog, 1999).

Oil palm (*Elaeis Guineensis*) originated from the tropical forest of West Africa. It has now become a major cash crop and is cultivated commercially in Malaysia. Empty fruit bunch (EFB) is one of the natural fillers obtained from oil palm. Many million tons of EFB on dry weight basis are produced annually throughout the world. This is an industrial waste which is left unutilized after the removal of the oil seeds for oil extraction. In order to minimize the abundant of this waste and environmental problems, therefore new applications are urgently required for EFB to be more useful (Sareekala, *et al.* 1996/97). The utilization of this EFB as a reinforcement in polymers has economical as well as ecological importance.

The incorporation of EFB as a reinforcing component in the polymers composites has received much intention (Sareekala, *et al.*, 1996/97; Rozman, *et al.*, 1998, 2000, 2001; Sarani, *et al.*, 2001; Abdul Khalil, *et al.*, 2001). Most of the research works on EFB so far deals with polymers such as polypropylene, polyester polystyrene and phenol-formaldehyde resin. The researchers focused on the effect of filler loading, filler particle size distributions and filler treatment on the mechanical properties of the composites. Like other natural fillers, many issues have been identified in the processing of the composites. These include low compatibility between EFB fibre and polymer matrix, higher moisture intake, poor distribution of fibre in the matrix and effect of oil residue on mechanical properties.

Although EFB have been useful as fillers in various polymers as mentioned above the use of EFB filler in PVC has not been studied extensively. Most of the researchers have used wood flour, wood sawdust and rice husk ash as fillers in PVC matrix (Matuana, 1998; Matuana,*et al.*, 2001; Sivaneswaran, 2002; Sombatsompop, *et al.*, 2003). The use of these fibres in the PVC matrix produces products that are more brittle than neat PVC. The incorporation of the natural fibres alters the ductile mode of failure of the matrix. The brittleness of PVC/wood fibre composites compared to the unfilled PVC may prevent this emerging class of materials to capture their full market potential in applications such as door and window frame (Mengelollu, 2001).

One of the most important aspects in the development of PVC compound by using filler is to achieve a good combination of properties and processability at the moderate cost. As far as mechanical properties are concerned, the main target is to strike a balance of stiffness, strength and toughness (Mohd Ishak, *et al.*, 2000). The incorporation of filler in thermoplastic such as PVC is associated with an enhancement in stiffness and strength decrease the toughness. The incorporation of impact modifiers in PVC, on the other hand will result in a significant improvement in toughness at the expense of stiffness and strength.

The enhancement of impact properties of PVC-U by introduction of a rubbery phase is well known phenomenon which has been widely exploited commercially. Among those in widespread use are chlorinated polyethylene (CPE) and various modified acrylic. Hiltner (1984) reported that CPE modifier forms a network enclosing PVC primary particles in order to develop the desired impact strength whereas Robinovic (1983) found that acrylic modifier forms a distinct disperse phase in the continuous PVC phase. Besides that, the dispersed rubber particles of acrylic are independent of the processing time and temperature. On the contrary, the CPE network structure is dependent of those factors.

Although by different mechanisms, the studies have concluded that the function of the impact modifier used (whether in the form of discrete phase or network structure) is that to increase the toughness of PVC by facilitating the ability of PVC matrix to shear yield and/or craze. This means the PVC will deform rather than fracture when subjected to a sudden load (Petrich, 1973; Lutz, 1993). Meanwhile, Stevenson (1995) has pointed out that the impact modifiers can also be used to correct the detrimental effects of other additives such fillers by boosting the impact performance of the material.

1.2 Significance of the Study

Based on the previous findings as mentioned above, there is no study has been reported the useful of EFB fibre in PVC matrix. Most of studies involved investigation of the mechanical properties of the natural fibre polymer composites in terms of natural fibre content, various treatments of the fibre, whereas very few have investigated processability of the composites. According to Sombatsompop *et al.* (2003), area for PVC/wood fibre composites still open especially in terms of processability-related mechanical properties of the composites.

The property of PVC-U without impact modifier and filler is brittle, therefore impact modifier is added to increase the ductility of PVC-U. However, instead of ductility increases, the impact modifier used is costly and lack of the PVC-U stiffness. Fillers are used to overcome the imbalance of the impact strength and stiffness of modified PVC-U. The environmental issue is also seriously highlighted, and then several agricultural by products are used as attractive filler to reinforce with polymers to produce filled thermoplastics composites. Thus, in this study the strategies to combine both filler and impact modifier are to develop PVC composites with good stiffness without sacrificing the desirable toughness and with increasing the cost-effectiveness. Therefore, towards to understand the properties of the new development of PVC composite which make EFB waste more useful, thus in this study the properties of composites reinforced with EFB filler were investigated

1.3 Objectives of the Study

The main objective of the study is to produce EFB-filled PVC-U composites with good combination of properties. Therefore the effect of processing temperatures and screw speeds on the impact, tensile and flexural properties of the composites extudates. The influence of processing temperatures on fusion level of composites is also determined.

CHAPTER 2

LITERATURE REVIEW

2.1 Poly (vinyl chloride)

PVC emerged as the second largest of thermoplastics (polypropylene is a first in the list) being used as raw material in the plastic industry. The world market reported that growth rate of the world production and the growth rate of world consumption of PVC thermoplastic from 1999 to 2005 will be 3.30 % and 3.83 %, respectively (Murphy, 2001). It shows that although PVC has been remarked as a toxic material but its excellence properties and versatility cannot be denied.

2.1.1 Definitions and General Feature Properties

Poly (vinyl chloride), commonly known as PVC, is a predominantly amorphous polymer with a basic repeat unit [-CH_2-CHCl-]. Based on its chemical structure, PVC is structurally very similar to polyethylene (PE), with a chlorine atom substituted for a hydrogen atom on alternating carbons in the chain. The number of chlorine molecules attaching to theses components governs overall characteristics of PVC polymer or compound to a substantial extent (Penn, 1971).

The chlorine atom is many times larger than hydrogen and greatly reduces the ability of the molecules to rotate and flex. The base polymer is therefore quite stiff and low creep (long term deflection under stress) properties at room temperature. Due to high electronegativity, chlorine atoms create polarity and are electrically repulsive to each other. This feature results in further immobility, inflexibility and low level of crystallinity. Therefore, the impact resistance of the basic vinyl chloride is quite poor but relatively good resistance to organic solvents and flame. The polarity of PVC is responsible for its remarkable ability to form compatible compositions with many other polymers and plasticizers as well as other formulating ingredients (Coaker and Wypart, 1993). These advantageous properties offer to rigid PVC the opportunity to be used as base polymer matrix for reinforced version. This can be explained by their characteristics as listed in Table 2.1. Its main limitations are its low softening temperature (82°C) and glass transition (80-84°C), the tendency to degrade at elevated temperature.

Under heat or ultraviolet light, the chlorine atoms become unstable and break out of the molecules. This heat sensitivity makes the base polymer extremely difficult to be processed by conventional melt processes. However, due to its affinity for additives, this undesirable property of PVC is overcome by the development of suitable stabilizer in the early 1930's (Butters, 1982). Despite poor thermal stability, when correctly formulated, the PVC is widely used, as it is relatively cheap, inherently flame retardant, strong and stiff, and good chemical and weathering resistance (Whelan and Goff, 1989).

2.1.2 Morphology of PVC

The PVC resin particle is unusual in structure and to understand the properties and processing of this unique polymer requires an understanding of its structure or morphology. Suspension PVC resin particles usually posses a pericellular "skin" or membrane" which extends almost continuously over the entire outer surface of the grain. This skin is formed during the early stages of polymerization.

Table 2.1: Some physical properties of moulding PVC products and compounds (Titow, 1991)

Properties	Typical values or values range for PVC Rigid PVC
Density (g.cm^{-3})	1.30-1.45
Tensile strength (MPa)	31-60
Elongation at break (%)	2-40
Flexural strength (MPa)	62-100
Young's modulus (GPa)	
a. in tension, E	2.5-3.5
b. in shear, G	1.0-1.8
c. in flexural, E_B	2.0-3.5
d. in compression,K	2.2-3.5
Thermal conductivity(Wm^{-1}K^{-1})	0.14-0.28

Suspension-polymerized PVC in a powder form possesses various level of morphology. Powder particles, which are visible to the naked eyes, are known as grains. Each grain is an assembly of primary particles, which are loosely packed together. Each primary particle appears to be made up of smaller structure called domains. These are illustrated in Figure 2.1 (Hattori, *et al.* 1972).

There are the arguments among researchers in defining the terminology to describe PVC particle morphology. Thus, to avoid any confusing and to obtain consistency of the PVC terminology, a table with standard terminology was summarized and published by Geil (1977). This standard PVC nomenclature is shown in Table 2.2 and it will be used for further discussion on the morphology.

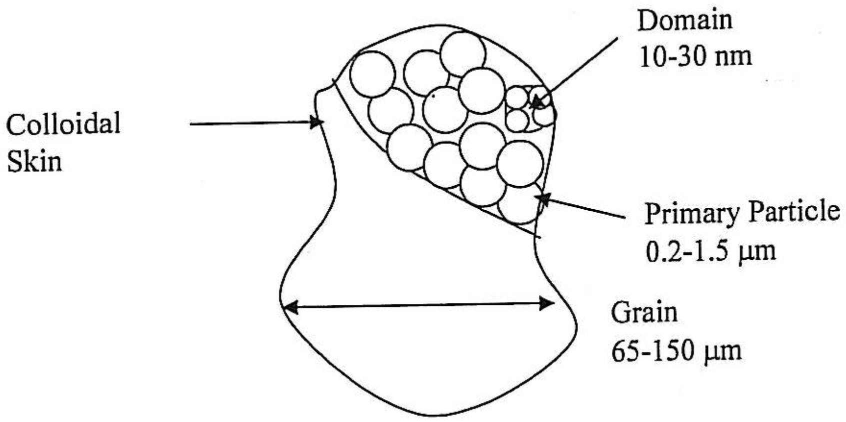


Figure 2.1: PVC particle morphology (Gilbert, 1985)

Although PVC is amorphous polymer, it has been reported that it contains about 5-10% of crystallinity. Based on small angle X-ray scattering (SAXS) data, the crystalline structure has been proved by the existence of crystalline noduli with size of 2.7 nm and spacing of 10nm of virgin PVC (Blundell, 1979). This crystalline nucleus surrounded by constrained amorphous has been associated with the basic micro-domain structure of PVC (Soni, *et al.* 1982 and Gilbert, 1985).

Summers (1981) studied the nature of crystallinity of PVC and found that the crystallites in the micro-domain structures were connected by tie molecules and have a 100 A spacing. These crystallites act as crosslink in three-dimensional network and prevent primary particles interaction at low temperature. However, at high temperature (177° C) some of the crystallites were melted and then recrystallization after cooling leads to strong ties between primary particles. It also revealed that PVC contains two kinds of crystallinity, namely primary crystallinity was formed during polymerization whereas secondary crystallinity formed during recrystallization (Lacatus and Rogers, 1986).

Table 2.2: PVC nomenclatures (Geil, 1977)

Term	**Range (µm)**	**Average (µm)**	**Description**
Grain	50-250	130	Visible constituents of free flowing powders, made up of more than 1 monomer droplet
Sub-grain	10-150	40	Polymerized monomer droplet
Agglomerate	2-10	5	Formed during early stage of polymerization by coalescence of primary particle (1-2 µm). Grows with conversion to size shown.
Primary particle	0.6-0.8	0.7	Grows from domain. Formed at low conversion (less than 2 %) by coalescence of micro-domain: grows with conversion to size shown.
Domain	0.1-0.2	0.2	Nucleus of primary particle. Only observed at low conversion (less than 2 %) or after mechanical working. It becomes primary particle as soon as growth starts.
Micro-domain	0.01-0.02	~0.02	Smallest species so far identified. Aggregate of polymer chains.

2.1.3 PVC Fusion

PVC must undergo fusion in every fabrication process in order to achieve its best mechanical properties. It is generally agreed by the researchers that the level of fusion has vital influence on the mechanical, physical and chemical properties of the PVC. To obtain optimum properties an appropriate level of fusion is needed (Covas, *et al.* 1988)

The fusion process for PVC is the subject of great deals of discussion, and there are many views about the exact courses of this process. Although there are several ideas as mentioned by researchers but one conclusion to describe the fusion process of PVC can be made, namely the fusion process is the breakdown process of the PVC grains and affected by a combination of such external parameters such as temperature, pressure and shear. The methods used to breakdown the PVC grains, whether Brabender torque rheometer, two roll mill, single screw extruder or twin screw extruder, resulting in the different breakdown mechanisms (Benjamin,1980; Parey and Manges;1981; Portingell, 1982).

Benjamin (1980) has defined that the fusion of PVC as the process of removing the boundaries between base particles by which a continuous mass formed having a homogeneous molecular structure. The grains have broken down to the so-called primary particles. The further broken down of the primary particles to the subprimary particles occurred as the sufficient energy was applied. The structure of this subprimary the particles was said to be continuous network of molecular chains which, to some extent, were bound together by a crystalline nucleus. The subprimary particles contain approximately 10 % crystalline material.

Parey and Manges (1981) concluded that the fusion of PVC proceeds in two stages. In the first stage, the PVC dry-blend was changed into a material capable of fusing under the influence of temperature and particularly of shear. In the second stage, mainly through the effect of temperature, this material becomes fused. The shear serves here only to homogenize the material. While, Portingell, (1982) has defined the fusion process was a destruction process of grains and primary particles and establishment of an entanglement network locked by a network of crystallites. This destruction process is accomplished by the combined action of heat, pressure and shear during processing. The extent of destruction of particulate of PVC is known as the degree of fusion.

2.2 PVC Composites

There has been increasing interest in commercialization of natural fibre composites and their use, especially for automobile industry. Composites based on polyolefins are now commercially available. It is reported that these composites offer advantages of 20 % reduction in processing temperature and 30 % of weight reduction (Nabi Saheb and Jog, 1999). Meanwhile, the analysis of the cellulosic filler cost/performance has shown that advantage of wood fibre over glass fibre and mica filler (Raj, *et al.* 1990)

2.2.1 Composite: Definitions and Categories

The word ‘composite’ means ‘consisting of two or more distinct parts’. A composite material is formed when two or more materials are combined so that the properties of the composite are different from, and usually better than, those of the individual constituents. Composites are made up of continuous and discontinuous mediums. The continuous medium is called matrix and the discontinuous medium, which is usually the harder and the stronger one, is called reinforcement. The properties of a composite are dependent on the properties of the constituent materials, and their distribution and interaction (fibre-matrix interface)(Mahmood Husein Datoo, 1992).

Composite materials can be broadly classified into three categories: particulate composites, fibrous composites and laminated composites (Mahmood Husein Datoo, 1992). The definition of fibrous composites is referred to the fibre which has the particle size longer than 100 μm with a length to diameter ratio lesser (aspect ratio) than 10, whereas the opposed particle size as stated above is referred to the particulate composites. (Katz and Brandmaier, 1987)

2.2.2 Matrix

The main roles of the matrix are to bind and transmit and distribute stresses among the individual fibres, and to maintain the fibres separated and desired orientation. The matrix also provides protection against both fibre abrasion and fibre exposure to moisture or other environmental conditions, and causes the fibres to act as a team in resisting failure and deformation under load. The maximum service temperature of the composites is limited by the matrix ((Katz and Brandmaier, 1987; Miller, 1996)

Although the thermoset polymers have some great virtues when used as matrices in plastic composites such as low viscosity to facilitate thorough wetting of reinforcing particles, economical for large components and high softening points, however recent years have seen rapid growth in the used of reinforced thermoplastic polymers. The major advantage of thermoplastic matrix is that forming is possible by injection moulding or extrusion techniques. These are the most economical processes when cheap and precise manufacture of very large quantities of components is required (McCrum, *et al.*1997). However, some special consideration must be made for thermoplastics because their poor high-temperature capability (Collyer and Clegg, 1986).

Most of the PVC is used as composite matrix in the mineral filler-filled PVC composite. Calcium carbonates ($CaCO_3$) have been extensively used in PVC composites to enhance mechanical and thermal properties of PVC, such as hardness, stiffness and heat resistance. However, the use of fillers at high loading leads to a decrease in yield stress, tensile strength and ductility compared with the based polymer (Saad, *et al.* 1999). Recently, PVC matrix is used as the matrix for nano $CaCO_3$. The impact strength, flexural modulus and softening point temperature of PVC are significantly enhanced after addition of 0-15 phr nano $CaCO_3$ but the yield strength and elongation at break of PVC are decreased (Chen, *et al.* 2004).

The previous findings as mention above show that PVC thermoplastic is also competent to be a good matrix in the composite system. Although unlike thermosetting resins, if more ductile and versatile PVC thermoplastic is modified with some consideration on heat processing, the potential to be used PVC as matrix in the thermoplastic composite is become wide.

2.2.3 Filler and Reinforcement

The basic purpose of fillers is to 'fill' a plastics that is increase the bulk at low cost, to improve the economics. With good adhesion between filler and polymer, consequently the filler begins to play a role in improving mechanical properties (Murphy, 2001). Therefore, recently the primary purpose for using fillers in thermoplastics is to modify properties such as stiffness, hardness, thermal stability and shrinkage, instead of reducing the material cost. In this case, the fillers function as the reinforcing fillers in the composites. Reinforcing fillers are additives that improve the tensile strength and tear resistance of composite; fillers (also called extenders) reduce both of these properties. In general, the less fibrous (more particulate) fillers which have the lower aspect ratio actually provide less reinforcement (Schwartz, 1987).

Whether extender or reinforcing filler, the effect of fillers on the composite properties are largely dependent on the mean particle size, particle shape, particle size distribution of filler and the strength of the bond between filler particle and polymer matrix (Wypych, 1999). Most filler do not consist of particles which are all the same size and the same shape. The shape of the particles affects the modulus and other mechanical properties significantly. Spherical particles are less likely to initiate cracking than more slender particles. However, flat platelet-type particles tend to produce stronger bond between the filler particle and matrix, due to increased surface area available for bonding. One measurement of shape is the aspect ratio. The greater aspect ratio, the greater the increase in bonding area is available which usually improved mechanical properties. (Miller, 1996).Example of aspect ratio calculation is illustrated in Figure 2.2.

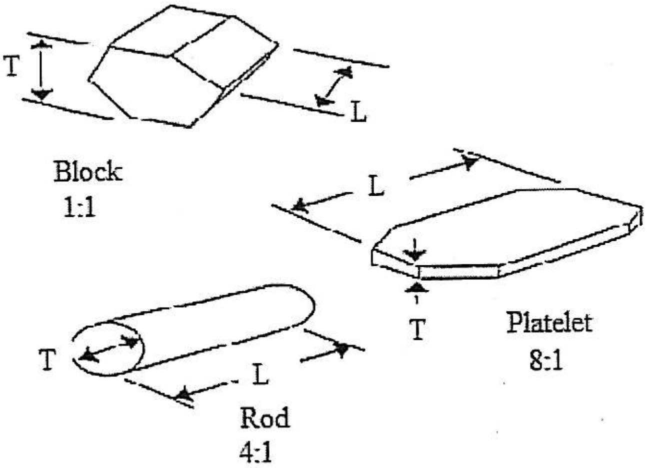


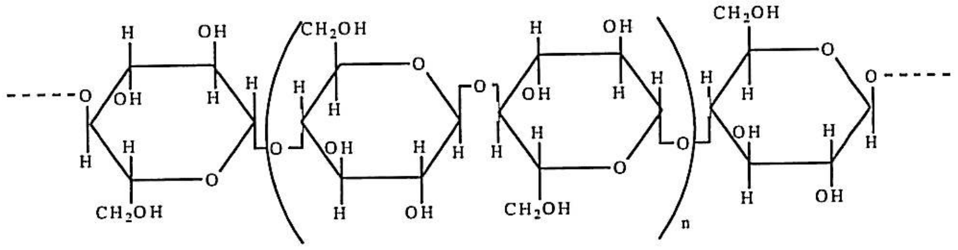
Figure 2.2: Aspect ratio calculation (Wiebking 1986).

According to Wiebking (1986), the flexural modulus or stiffness of PVC composite is directly proportional to the aspect ratio of the filler being used. A high aspect ratio can also increase the tensile properties of PVC composite by providing more surfaces to contact the polymer. As a material is made stiffer it also becomes more brittle. Miller (1996) reported that the tensile strength depends on the interfacial bond strength that determines the distribution of stress between filler and matrix a. He concluded that, in general, the smaller the average size of the filler particles, the greater their surface area and the greater the bonding, resulting composite with good tensile strength.

Organic fillers such as wood flour, pulp fibre, rice husk are also used in PVC compounds. They possess low specific gravities (about 1.4) and when used to extend resins they offer a significant cost saving. However, the use of cellulosic fillers in the PVC materials is not as common as in polyolefins (polypropylene and polyethylene).

Organic filler or natural fillers are biological structures composed mostly of holocellulose (combination of cellulose and the hemicellulose.) and lignin, and other traces of plant materials, namely inorganic(ash content) and extractives (a group of cell wall chemicals mainly consisting of fats, fatty acids, fatty alcohol and so forth). The inorganic content can be quite high in plants containing large amounts of silica. The extractives can be removed by one or several extraction procedures (Han and Rowell, 1997).

These polymers are rich in hydroxyl groups, which are responsible for moisture sorption through hydrogen bonding, biodegradation and thermal degradation of the fiber whereas lignin is thermally stable but is responsible for the UV degradation. Generally, without non-wood fibers, the fibers contain 60-80% cellulose, 5-20 % lignin and up to 20% moisture. The hydrogen bonds and other linkages provide the stiffness to the fibers (Saheb and Jog, 1999). Figure 2.3 shows the structure of cellulose molecules. They have a tendency to form intra and intermolecular hydrogen bonds.



n= repeat unit

Figure 2.3: Cellulose molecular structure (Han and Rowell, 1997)

Low thermal stability of natural fillers is the primary drawbacks. The thermal degradation of natural fillers is a two-stage process, namely degradation of hemicellulose and cellulose at low temperature (220 –280°C) and lignin at high temperature (280-300° C). The suggested processing temperatures are thus limited to about 200°C. The lower processing temperature permissible is due to the possibility of natural fillers degradation and/or the volatiles emissions that could affect the composites properties. This factor will limit the type of the thermoplastics that can be used with agro-fibres to commodity thermoplastics

The other primary drawback is the high moisture absorption of the natural fillers. The moisture absorption can result in swelling the fibres, dimensional variation in composites and affects the mechanical properties of the composites. The moisture content, during processing of composites based on the thermoplastics, can lead the poor processability and porous product. (Sanadi, *et al.*1997).

Uniform distribution and interaction of natural fillers is also important, so that as many polymer chains as possible can be bound to the free filler surface. However, natural fillers have poor interactions with the polymer matrix and tend to aggregate due to strong hydrogen bonding, which holds the filler together, thus fibres do not disperse well in a hydrophobic polymer matrix. Due to the poor adhesion at the fibre-matrix interface, the fillers do not function as an effective reinforcement system. Modification or surface treatment of the fibres with suitable chemical reagents prior to processing leads to good dispersion and adhesion and significantly improved mechanical properties of the composites and also reduced water absorption.

Thus, the important factors which need to be considered in the application of natural fibres as fillers or reinforcements in the composite system are;

- thermal stability of the fillers
- surface adhesion characteristics of the fillers
- dispersion of the fillers

The incorporation of wood fibre derived from cellulose into PVC matrix has been studied extensively by Matuana, *et al.* (1997, 1998, and 2001). The main reason for using wood fibre or fibrous cellulose materials lies in the fact that these fibres are offer a number of advantageous over currently used reinforcing inorganic materials in terms of cost on unit volume basis, flexibility during processing and stiffness.

Matuana, *et al.* (2001) studied the photo-aging and stabilization of rigid PVC/wood-fibre composites. The experimental results showed that wood fibres are effective sensitizers and that their incorporation into a rigid PVC matrix has accelerated the photo-degradation of the PVC matrix when exposed to UV radiation. They also reported that the light stability of these composites could be improved quite efficiently with the addition of rutile titanium dioxide photoactive pigment during formulation. In their previous investigation, the incorporation of wood fibre increased stiffness and melts viscosity of the composites and limited the potential of producing a foamed composite with high void fraction (greater than 20 %). However with good treatment of fibre (silane), the foamed PVC composites with void fraction larger than 20 % were successfully achieved (Matuana, *et al.* 1997)

Mengeloglu (2001) has studied the effects of impact modifiers such as methacrylate-butadiene-styrene (MBS), all acrylic (ACR) and CPE on the properties of rigid PVC/wood fibre composites. The wood fibre from hardwood maple was used. Based on the mechanical property testing, the author concluded that the MBS and ARE were more efficient and effective in improving the impact resistance of the composites than CPE modifier. Meanwhile the tensile strength and modulus of composites were significantly decreased by the addition of impact modifier.

The effect of rice husk ash (RHA) in the PVC-U has been studied by Sivaneswaran (2002). The RHA-filled PVC has been impact modified with ABS and coupling agent modified with LICA 12. The results showed that the sample containing 8 phr ABS and 40 phr RHA fillers treated with LICA 12 was found to be the most effective formulation in terms of flexural modulus, impact strength and cost.

Sombatsompop, *et al.* (2003) have used saw dust as a filler in the PVC to examine the sawdust content on structural and thermal changes, and rheological and mechanical properties of composite. The results revealed that the extrudate swell monotonically decreased up to 33.3 wt% sawdust content and became independent and sawdust content at greater than 33.3 %. Meanwhile, tensile, impact, flexural and hardness properties considerably decreased with up to 16.7% sawdust content before levelling off for higher sawdust loading. The decreases in mechanical properties are associated with the presence of moisture, interfacial defects and fibre dispersion in PVC matrix. Thermal degradation of composite also decreased with increasing sawdust content, which caused by chlorine atoms cleavage due to strong hydrogen bonds of wood fibre and PVC molecules. However the glass transition temperature was found to improve after adding 16.7 % sawdust due to strong hydrogen bonds and polar-polar interaction between PVC and sawdust molecules.

2.2.4 Filler-Matrix Interface

One of the factors controlling the performance of the composite materials is the bond between the filler and the matrix. The localized stresses are usually highest at or near the interface, which may be the point of premature failure of the composite. As a result, a high interfacial strength is desirable to maximize the overall composite strength. The filler-matrix interface bond act as a bridge to allow the dispersed phase (filler) to 'communicate' with matrix to transmit the stress or strain due to a mechanical load from one phase to the other (Shackelford, 1996).

The interface must have appropriate chemical and physical features to provide the necessary load transfer from the matrix to the filler. The use of a coupling agent can provide improved interfacial condition (Katz and Brandmaier, 1986).

2.3 Oil Palm Empty Fruit Bunch -Polymer Composites

As mentioned before most of the published papers have reported the use of EFB fibre or filler as reinforcement in the thermoplastics polymers such as polyethylene (Mohd Ishak, *et al.* 1998), polypropylene (Rozman, *et al.*2001) and polystyrene (Sarani, *et al.* 2001) and in thermosets such as polyester (Hill and Abdul Khalil, 2000 ; Abdul Khalil, *et al.* 2001) and phenol-formaldehyde (Sreekala *et al.* 1996, Sreekala and Thomas, 2003). However, areas for using of EFB fibres or fillers in the PVC composites are still wide, especially in terms of the processability, thermal stability-related to mechanical properties of the composites (Sombatsompop, *et al.* 2003)

2.3.1 Oil Palm Empty Fruit Bunch Fibre and its Chemical Composition

Oil palm empty fruit bunch (EFB), one of the by-products of oil palm milling process, can be found in abundance in palm oil mills in Malaysia. Compared to other by-products such as oil palm trunk (OPT) and oil palm fronds (OPF), the EFB has the economical advantage of being collected at the oil palm mills. Currently, Sabutek Sdn. Bhd, Ipoh, Perak, Malaysia has commercially produced the pulverized EFB fibres and fillers. Because of this reason, EFB was chosen as the filler for PVC-U composites in this study.

The EFB fibres are derived from oil palm tree *(Elais guineensis)* component. The chemical composition of untreated EFB fibre strands has been reported by Peng (2002) as shown in Table 2.3.

Table 2.3: Chemical analysis of EFB

Chemical Composition	Percentage (%)
Extractive	2.3
Holocellulose	82.5
α-cellulose	60.6
Lignin	17.2
Hemicellulose	32.5
Ash content	5.4

2.3.2 Utilization of EFB Fibre in Polymer Composites

Utilization of short EFB as a reinforcement in phenol-formaldehyde (PF) resins has been studied by Sreekala *et al.*(1996). Four types of fibre length of 20, 30, 40 and 50 mm with various fibre loading were used for composite fabrication by means of hand lay-up. . The tensile, flexural and impact properties showed improvement upon reinforcing with the fibre. The elongation, brittle nature and buckling characteristics of PF resin were considerably improved by incorporating the fibre. The fibre length of 40 mm and at 38 % fibre loading has produced the maximum tensile and flexural strength. Fibre entanglement at higher fibre length was the reason for the decrease in properties beyond 40 mm fibre length. Meanwhile the impact properties showed linear enhancement with increase in fibre length up to 50 mm. However, for the best balance of mechanical properties, the optimum fibre length and fibre loading were considered to be 40 mm and 30 %, respectively.

EFB fibre is also utilized in thermoplastic composites. Rozman, *et al.* (1998) have studied the effect of compounding techniques, effect of oil palm EFB filler loading and the effect of coupling agents on the mechanical properties of EFB/PP composites. The study revealed that the incorporation of EFB into PP matrix resulted in the improvement in the tensile modulus. However, the tensile strength, elongation at breaks and impact strength decreased with increasing filler loading. Composites produced by internal mixer have displayed higher mechanical properties than with those produced by extrusion. The better performance has been attributed to the effectiveness of the internal mixer in improving the wetting of the filler surface. Meanwhile, the incorporation of compatibilizer and coupling agent, that is, maleic anhydride-modified PP (Epolene E-43) and 3-Aminopropyl triethoxysilane(3-APE), respectively, have produced composites with improved tensile and impact strength. The presence of coupling agent and compatibilizer has also affected the amount of water absorbed. The water absorption decreased as the loading of coupling agents increased.

Besides the effect of chemical modified oil palm EFB, fibre-matrix interaction of composites, the effect of silane-based coupling agents treated EFB, and acrylic acid based compatibilizers on the mechanical properties of other polymer matrix, such as high density polyethylene (HDPE) also has been investigated by Mohd Ishak *et al.*(1998).They have found that tensile modulus of EFB-HDPE without chemical treatment showed an increase, whereas tensile strength, elongation at break and impact strength decreased with the increasing filler loading. The strong tendency of EFB to exist in the form of fibre bundles, poor filler-matrix interaction was believed to be responsible for the poor strength displayed by the composites. Meanwhile, almost all chemical treatments increased the stiffness of the composites but limited improvement has been observed in the case of tensile strength. This has been attributed to the presence of fibre bundles that remain intact even after several types of chemical treatment have been carried out. Scanning electron microscopy (SEM) micrograph revealed that the main energy-absorbing mechanisms contributing towards toughness enhancement was through the fibre bundle pull-out process.

Rozman *et al.* (2001) in their recent paper reported the effect of oil extraction of the oil palm EFB on the mechanical properties of PP/EFB/Glass Fibre Hybrid composites. The results showed that composites with oil-extracted EFB produced significantly higher flexural and tensile strength and toughness than those without extraction. It revealed that the efficiency of coupling agent was enhanced with the removal of oil from the EFB surface. The main reason is the oil extraction has resulted in the formation of continuous interfacial region (good adhesion) between EFB and PP matrix.

Abdul Khalil *et al.* (2001) have used other method to improve the compatibility between oil palm EFB and the polyester polymer matrix, where EFB was chemically modified using non-catalysed reaction with acetic, propionic and succinic anhydrides. The effects of various anhydride modifications on mechanical properties and water absorption of oil palm reinforced polyester composites were investigated. The properties were improved for modified fibres whereas unmodified EFB fibres exhibited poor mechanical properties and higher water absorption. It was concluded that the chemical modification resulted in hydrophobic fibre and hence improved the fibre-matrix bonding.

Another research, oil palm EFB was used in the polystyrene matrix as reported by Sarani Zakaria, *et al.*(2001). The benzylation process treatment was used in order to decrease the hydrophilic property of EFB fibres where the hydroxyl groups were replaced by aromatic rings from benzyl groups to produce benzylated EFB fibres. The results showed that the modified EFB/polystyrene produced better mechanical and physical properties than unmodified EFB/polystyrene composites, due to the enhancement of compatibility between EFB fibres and polystyrene.

Recently, Sreekala and Thomas (2003) studied the effect of fibre surface modification on water-sorption characteristics of EFB fibres. Surface modification such as mercerization,latex coating, gamma irradiation, silane treatment, isocyanate treatment, acetylation and peroxide treatment were used to reduce the hydrophilicity of fibres. Treatment reduced overall water uptake at all temperatures (30, 50, 70 and 90°C). The decrease was due to its physical and chemical changes occurred to the fibre on modifications. The effect of sorption on the mechanical performance of the treated and untreated fibres was also investigated and it was found that the properties fibres decrease upon sorption and regains on desorption.

Based on the literatures reviews, the following conclusions are derived. The addition of EFB fibres or other natural fillers as reinforcement generally has increased the modulus of the composites while decreased the strength, elongation and water absorption.

2.4 Mechanical Properties of PVC-U Extrudates

The mechanical properties of PVC-U extruded using single screw extruder have been studied by Covas and Gilbert (1992). Effect on fusion and mechanical properties were to be the main aim in their study. Owing to the significant levels of shear present in single screw extrusion, the route to fusion seems to be different from that observed in other processing techniques, namely twin screw extrusion. Therefore, they have proposed the hypothesis for the fusion of PVC-U compounds in single screw extruder was developed from that originally derived by Allsopp (1982). This mechanism of fusion was illustrated in Figure 2.4. The melting of the solid material occurred either by comminution of grains with internal structure substituting or by ruptures of internally fused grains. In other words, the comminution and compaction and internal fusion of grains could be observed simultaneously in the machine. Progressive fusion and gradual increase in melt homogeneity were postulated. A network structure of entanglements and primary and secondary crystalline was present upon cooling.

It is well known in the PVC profile extrusion industries that the mechanical properties of the product are related to the processing conditions. Shear and temperature are generally agreed to be considered as that conditions in PVC processing. Both parameters are strongly related to PVC fusion. Increasing shear in an extruder can be done by increasing the screw speed. Unfortunately the residence time is decreased at the same time counteracting the effect of the shear increase. In the previous finding, Summers (1982) found that the morphology and toughness of a PVC compound in single screw extrusion was effected only slightly by shear rate but was dominated by melt temperature. He found that the toughness of PVC compound went through a maximum then decreased with increasing melt temperature. The decreased of this toughness was due to melt fracture as a result of the failure of lubrication system. Similar result was also reported by Lacatus and Rogers (1986), in which the better toughness was observed at 177°C melt temperature (lower fusion level) than a melt temperature at 204°C. The melt fracture that observed at 204°C was a reason for toughness decreasing.

The mechanical properties, namely tensile and impact, Covas and Gilbert (1992) found that the yield parameters were insensitive to variations in the degree of fusion whereas the impact strength was increased and exhibited a maximum value. Similar results have also been extensively reported by other researchers (Benjamin, 1980, Uitenham and Geil, 1981 and Terselius and Jansson, 1981, 1984, 1985). Based on the previous findings, the conclusion can be made that in order to obtain optimum mechanical properties an appropriate level of fusion was needed. It means that although on processing the original particles was progressively change into a network of entanglements and primary and secondary crystallinity, but the presence of some remaining primary particle was necessary for optimum properties. Meanwhile, the independent of yield to the degree of fusion was related to molecular relaxation processes which independent to the relative coherence of the entanglement network and number of crystalline junctions.

The interrelation of yield stress and toughness is vital importance. The materials with the highest yield stresses will prone to fail in a brittle manner. This is generally true for all materials, those which are classified as being strong (high yield stress) most often brittle such as alloy steels, titanium and so forth. In the polymer family (excepting rubbers which are both strong and ductile) this truth is generally bear out in practice. As a broad generalization, plastics with yield strengths greater than the yield for PVC-U are considered to be brittle (Marshall, 1982). The yield stress of the polymers is not constant; its values will change depending on the strain rate and temperature of the testing. The Eyring equation, which describes the temperature and strain rate dependence of the yield stress, has been chosen by many researchers as their starting point for tensile yield behavior investigation (Ward, 1971, Marshall, 1982, Povolo *et al.* 1996).

Wee (2001) has also applied the Eyring equation in his tensile yield studies to predict the tensile yield stress of calcium carbonate ($CaCO_3$)-filled PVC-U at impact velocity of 3 m/s. Tensile yield studies are made by determining the yield stress at the variety of strain rate. By using the Eyring equation, the yield stress increased linearly with the logarithm of tensile strain rate. By applying linear regression and extrapolating the data, the yield stress value at $3 m.s^{-1}$ was obtained. Similar method had previously used by other researchers (Clavert *et al.*1991, Azman Hassan, 1996).

CHAPTER 3

MATERIALS AND METHODS

In this section the materials and blend formulation are descirbed. The dry blending, extrusion processing, mechanical testing and DSC are described.

3.1 Materials

The PVC used in this study was a suspension resin with solution viscosity K-value 66, and trade name MH-66. This homopolymer PVC is a medium molecular weight resins designed for general purpose, rigid and flexible applications. It was supplied by Industrial Resin Malaysia (IRM) Sdn Bhd. The specifications of PVC used are summarized in Table 3.1.

The additives used in the PVC formulations are shown in Table 3.2. Most of the additives used were also supplied by IRM Sdn. Bhd except for impact modifiers and filler. Core and shell impact modifier with a trade name of Paraloid KM355P was supplied by Kureha Chemicals. It consists of poly(methyl methacrylate) (PMMA) as a shell while poly butyl acrylate (PBA) as a core. Meanwhile, chlorinated polyethylene (CPE) for thermoplastic applications with trade name Tyrin 702P and containing 36 % chlorine contents was supplied by Dupont Dow Elastomers. The pulverized Oil Palm Empty Fruit Bunch (EFB) in the powder form was purchased from Sabutek Sdn. Bhd., Teluk Intan Perak. The chemical analysis of untreated EFB as shown in Table 2.3 (Chapter 2) could be used as the specified chemical analysis of EFB filler in this study.

Table 3.1: Specifications of PVC suspension resin MH-66

Appearance	White Powder
Degree of polymerization	1000 ± 50
K-value	66
Specific gravity	1.4
Bulk density (g/cm^3)	0.50±0.05
Volatile Matter (max) (%)	0.5
Foreign Matter (grain/100g)	15
Particle size (%)	0.3

Table 3.2 : Types of additives

Additives	**Types**
Internal lubricant	Calcium Stearate (CaSt)
External lubricant	Stearic Acid (HSt)
Processing aid	(PA-Acrylic)
Pigment	Titanium Oxide (TiO_2)
Heat Stabilizer	Tin (Sn)
Impact Modifier	Acrylic
	Chlorinated Polyethylene (CPE)
Filler	Oil Palm Empty Fruit Bunch (EFB)

3.2 Filler Preparations

The EFB filler was supplied in bulk with unknown sizes by the supplier. Thus, in this study the Restsch shaker was used to separate the EFB fillers into different sizes. The shaking time and vibration speed were set up at 10 minutes and 60 Hz, respectively. The main filler size used in this study in range of 0-75 µm with filler distribution and filler morphology is shown in Figure 3.1. EFB filler distribution was determined by using Malvern Instruments Mastersizer. Another filler sizes used as the minor part of this study in order to investigate the effect of filler size on the mechanical properties were 0-53 µm and 75-150 µm. EFB fillers were oven dried at temperature of 105°C for about 24 hours until the weight of the EFB fillers was constant, the weight loss then being calculated. The moisture content of the EFB filler was found to be around 5 %.

The temperature and time duration used for drying process in this study were referred to previous researchers who used natural fibre in their studies (Zaini, *et al.* 1996 and Hill and Abdul Khalil, 2000). The dried EFB filler was placed into desiccators for cooling and it was left in desiccators before dry blended with PVC and other additives to produce the dry blends of PVC-U compounds.

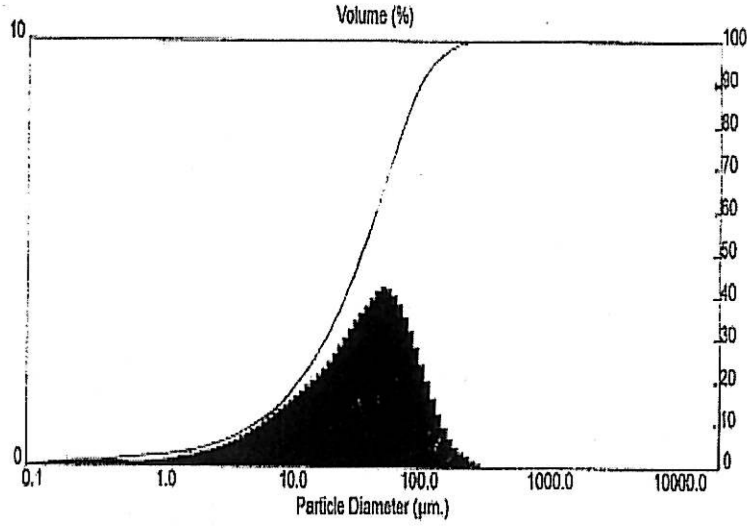


Figure 3.1 : EFB filler distribution (0-75µm)

3.3 Blend Formulations

The PVC-U dry blend formulations are shown in Table 3.3, which was based upon typical commercial PVC window formulations with some modifications. The basis of this formulation was also used by previous researchers such as Wee (2001) and Sivaneswaran (2002). The additives loading were unchanged in the formulations except for EFB filler, CPE and acrylic impact modifiers.

Table 3.3 : Blend Formualtions

Ingredients	Formulations								
	S_0	S_3	S_7	S_9	S_{10}	S_{21}	S_{22}	S_{37}	S_{38}
PVC	100	100	100	100	100	100	100	100	100
CaSt	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
HSt	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6
PA-Acr	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
TiO_2	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0
Sn	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.0
EFB	0	0	0	10	20	10	20	10	20
Acrylic	0	9	0	0	0	9	9	0	0
CPE	0	0	9	0	0	0	0	9	12

3.4 Dry Blending

The purpose of dry blending is to obtain the good homogeneity of the blends. In other word to ensure all the incorporated additives are well dispersed and distributed in the PVC resin. Before the dry blending takes place the correction proportion of the resin and the additives have to be weighed and mixed appropriately. The dry blending of PVC resin and additives was done using a high-speed mixer of the impeller type and water jacketed. The formulations were dry blended in 4 kg batches, using the following sequence: 1 min at low speed and followed by another 1 min at high speed. The temperature was risen, due to the generation of frictional heat, thus the mix was discharged from the mixer chamber after 2 min in order to avoid the temperature exceed than 60°C.

3.5 Extrusion Processing

Each PVC-U dry blend was oven dried at temperature of 105°C for 24 hour before they were compounded on a single screw extruder at two conditions as shown in Table 3.4. In the first condition, the extrudates were extruded at constant screw speed with different processing temperatures whereas the second condition was vice versa. The extrudate in the stripe form with approximately dimension of 3mm in thickness and 100 mm in width was cooled in the ambient temperature. The extrudates were cut into bars by using a band saw before they used in the specimen moulds. . Specimens for impact flexural and tensile tests were shaped according their dimensions by using cutter machine. The cutting side of specimens test was polished by using sand paper before carrying out the testing.

Table 3.4: The conditions of extrusion processing

Constant Screw Speed				
Screw Speed (rpm)	**Processing Temperature (°C)**			
	Feed Zone	**Compression Zone**	**Metering Zone**	**Die**
60	160	170	180	180
60	165	175	185	185
60	170	180	190	190
60	175	185	195	195
Constant Processing Temperature				
50	160	170	180	180
60	160	170	180	180
70	160	170	180	180

3.6 Izod Impact Testing

Sample test bars for impact test used was 62.5 x 13x 3 mm^3 according to the size specified in ASTM D256 (A). Notching was carried out manually using a Davenport notch cutting apparatus. The notch depth was fixed at 2.5 ± 0.02 mm with angle 45°.The notched samples impact tested in the Izod mode was carried out by using Toyo-Seiki Pendulum Impact Tester with impact velocity of 3.0 $m.s^{-1}$ and 150° swing angle using a 31.87 N of hammer in room temperature. The reported values for the tests were average values of seven specimens.

3.7 Flexural Testing

Sample test bars for flexural test were also extruded from the extruder. The flexural test was conducted on the Instron Machine with static load cell of 30kN according to ASTM D790, a three point bending system. The samples with dimensions 125 x 13x 3 mm^3 were tested at crosshead speed of 3$mm.min^{-1}$. The support span for the flexural test was 51 mm.

Flexural modulus was represented by the slope of the initial linear portion of the stress-strain curve. It was envisaged the resistance to elastic deformation under applied stress. Meanwhile flexural strength was represented the resistance to plastic deformation and failure under applied stress. They were calculated according to the equations as follows:

$$\text{Flexural modulus} = \frac{L^3 \Delta W}{4bd^3 \Delta S} \qquad (3.8)$$

$$\text{Flexural strength} = \frac{3WL}{2bd^2} \qquad (3.9)$$

Where W is the ultimate failure load (N), L is the span between the centers of support, b is the mean width of the specimens (m),. d is the mean thickness of the specimens of the sample, a ΔW is the increment in load (N) and ΔS is the increment in deflection. The reported values for flexural modulus and flexural strength were the average values of seven specimens.

3.8 Tensile Testing

The tensile properties, in accordance with the ASTM D638-91 method, were measured by using a Lloyd Instruments model EZ20 with four variations of crosshead speed, namely of 5.0, 20, 80 and 100 mm.min^{-1} and cell load of 20kN. The gauge length used was 60 mm. Likewise impact and flexural testing, the reported values for the test were also average values of seven specimens.

3.9 Differential Scanning Calorimetry (DSC) Study

Mettler Toledo DSC instrument Model 821 with STAR system was used.The extrudates with difference of processing temperatures were analysed in order to determine heat of fusion. DSC was carried out according to ASTM D 3418-82 at a heating rate of 10 °C min^{-1} to ensure high resolution of the DSC curve, and at temperature ranging from 30°C to 250°C. The liquid nitrogen rate was maintained at 50ml.min^{-1}. The calorimeter was calibrated with an indium reference. Meanwhile, samples in range of 8.0-8.4 mg were placed in a sealed aluminium pan and empty aluminium pan was used as reference. The DSC data was presented as heat flow against temperature plot.

CHAPTER 4

RESULTS AND DISCUSSION

In this section the influence of extruder screw speeds and processing temperatures on the impact, flexural and tensile properties of extrudates were studied. Several models of relative modulus were applied in order to compare the experimental with the predicted values. The prediction of yield stress at impact velocity of 3 $m.s^{-1}$ using Eyring equation was also performed.

4.1 Extrudates Characteristics

The dry blends, particularly for the dry blends contained EFB filler, must be sufficiently oven dried and left them to cold down for a few minutes in the closed oven before they were applied for extrusion processing. Insufficient drying process, the air was trapped during compounding and consequently the extrudate surface produced not smooth. These air bubbles were generated from the moisture of the dry blends and they were trapped in the extrudate due to a vent port unavailable at the compression zone of the extruder. Shortly after exit from the die, the air trapped inside the melt starts to migrate to the top of the surface and form the air bubbles. The air bubbles randomly distributed on the extrudate surface are obviously shown in Figure 4.1.

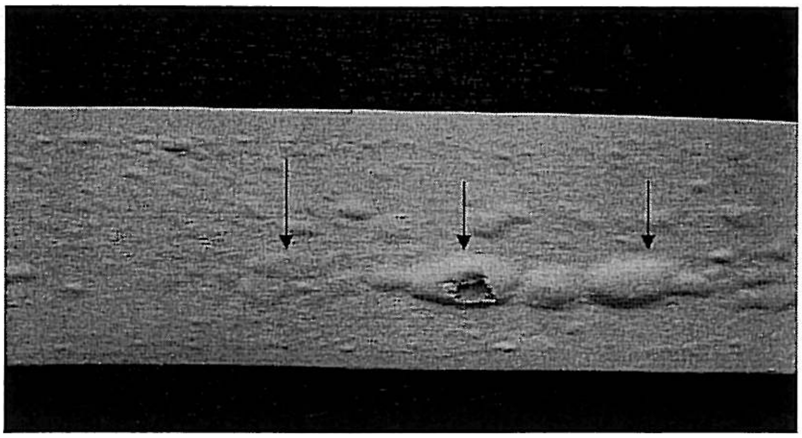


Figure 4.1 : Composite extrudate of S_9 with air bubbles on surface.

Besides air bubbles, Figure 4.2 shows that the defect of bambooing on the melt of EFB-filled CPE-impact modified PVC-U composite extrudate (S_{37}) at die temperatures of 180°C and 185°C shortly after exit from the die. Perhaps the10 phr of EFB filler and 9 phr of CPE in the S_{37} reduced the extent of fusion process for PVC, and thus increased the viscosity of the melt. Furthermore, as the melt is flowing, it develops a flow profile with the centre of the melt flowing faster than the edges, because of the friction of the melt against the edges of the die. The melt in the centre of the flow slides against itself easier than it can slide against the die walls. Consequently, as the melt left (extrudate) the die, it is distorted somewhat like bamboo because of the difference velocity between edges and centre. If the applied tensile stress, which is generated after the melt left the die, exceeds the melt strength (tensile strength) the edges of extrudate are ruptured (Figure 4.2). The bombooing defect was greatly eliminated as the die temperature increased to 190°C and 195°C whereby the extrudates with smooth surface are produced. Higher temperature in the die is eliminated the bambooing defect (Strong, 2000). Unlike S_{37}, other extrudates such as S_0,S_3,S_7,S_9,S_{21} did not experience this defect for all processing temperatures and screw speeds condition as shown in Figure 4.3.

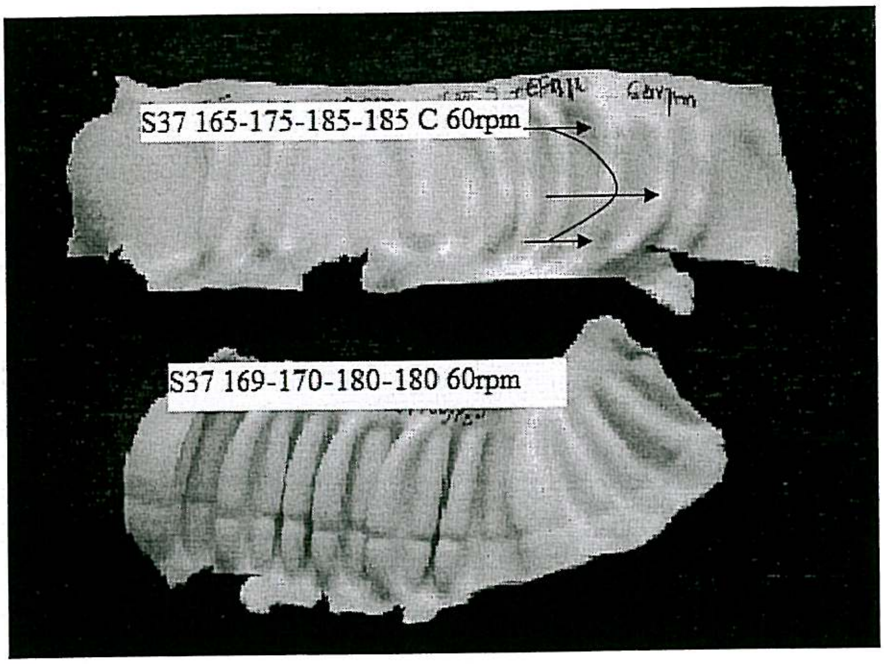


Figure 4.2: Composite extrudates of S_{37} at die temperature of 180°C and 185°C at constant screw speed of 60 rpm.

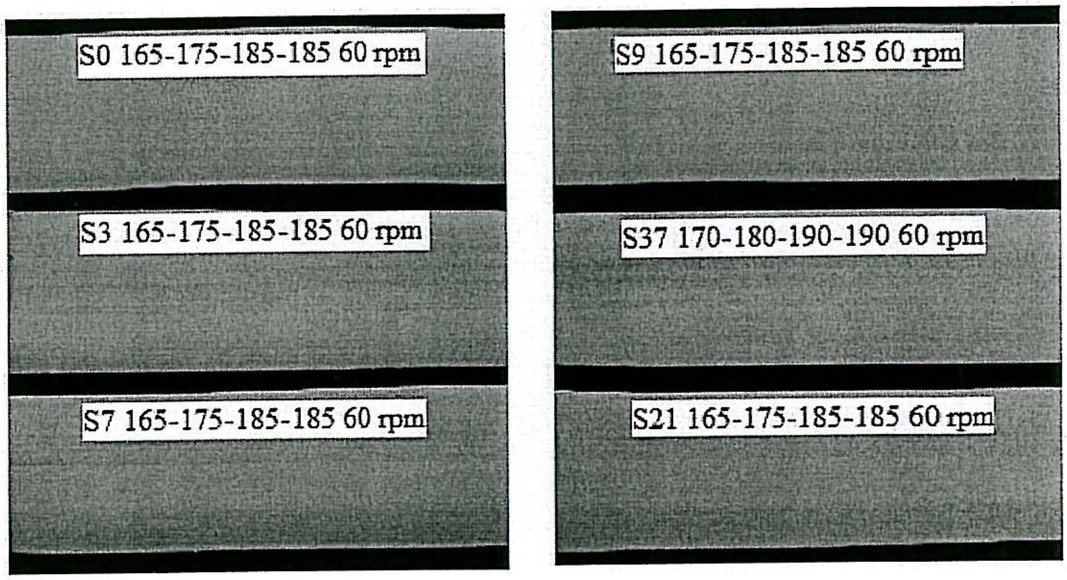


Figure 4.3: Extrudates of S_0, S_3,S_7,S_9 and S_{21} at die temperature of 185°C, and extrudate of S37 at die temperature of 190°C.

As EFB filler content increased to 20 phr in the dry blends such as S_{10}, S_{22} and S_{38}, the extrudates with rough surface were produced. The roughness of bottom surface was greatly noticeable than the top surface of composite extrudates. Any action to vary the processing temperatures and screw speeds of extruder was done but still did not change the quality of composite extrudates produced as shown in Figure 4.4. Based on visual inspection of the composites extrudates surface, it found that EFB fillers were not well distributed and dispersed throughout the matrix and precipitated at the bottom surface. Perhaps the shorter of L/D and single screw used in this study were the two reasons to describe the occurring of those defects. Therefore, in this study the mechanical properties of these composite extrudates were not evaluated.

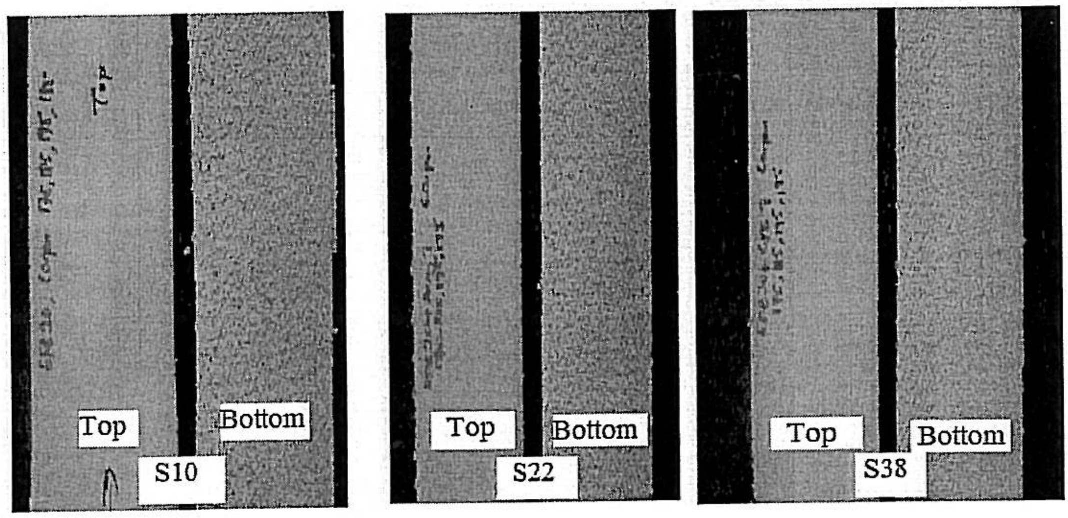


Figure 4.4: Composites extrudates of S_{10},S_{22} and S_{38} at Die Temperature of 195°C.

4.2 Impact Properties

Figure 4.5 shows the influence of extruder screw speed on the impact strength of PVC-U extrudates. Acrylic-impact modified PVC-U extrudate (S_3), the number of samples undergone hinge breaks at 50 rpm were 5 samples, while 4 samples at 60 rpm. At 70 rpm, all the samples have undergone hinge breaks. For PVC-U extrudate (S_0)

At 70 rpm, all the samples have undergone hinge breaks. For PVC-U extrudate (S_0) and CPE-impact modified PVC-U extrudate (S_7), all the samples were completely broken, nevertheless the impact strength values of S_7 at all screw speeds were higher than PVC-U. The greater of acrylic in enhancing the impact strength of the PVC-U extrudates compared to CPE was observed in this study.

Figure 4.5 also shows that the increase of screw speed from 50 to 70 rpm at constant of processing temperatures resulted in the impact strength of S_0 and S_3 increase, while slightly decreases in the impact strength for S_7. The increase of screw speed is simultaneously increased shear heating but reduced conductive heating during extrusion process. Furthermore, the residence time as shown in Table 4.1 also decreased at the same time counteracting the effect of the shear increased. On the contrary, the residence time of for all extrudates as shown in Table 4.2 insignificantly changed as the processing temperature increased at the constant screw speed. As a result the impact strength of extrudates is affected differently at different extruder screw speed. The increase of impact strength may be due to the less progressive fusion of the primary crystallites and increment of free volume as screw speed increased. According to *Cora et al.* (1999), the extent to which a matrix can yield to dissipate impact energy depends on the level of free volume in the matrix. As screw speed increased the three dimensional crystalline network is less fused. This resulted in a less packed matrix with higher free volume, leading to increase the toughness of the material. The decreasing in impact strength of CPE as the screw speed increased, perhaps the network to enclose PVC primary particles is not yet developed, and resulting the CPE impact modifier's being present as "globs" dispersed in the PVC matrix. In this configuration, impact strength is not developed.

Table 4.1 : Residence time of extrudates at constant die temperature of 180°C

Speed (rpm)	**Residence Time (s)**				
	S_0	S_9	S_3	S_7	S_{21}
50	60	61	62	63	63
60	55	53	53	54	50
70	41	43	43	42	44

Die Temp. (°C)	Residence Time (s)					
	S_0	S_9	S_3	S_7	S_{21}	S_{37}
180	55	53	53	54	53	none
185	53	54	57	56	50	none
190	54	55	58	57	53	60
195	53	53	58	58	53	55

Figure 4.6 shows the impact strength of composites with respect to the extruder screw speeds. The impact strength of S_9 decreases with increasing of screw speeds. Decreasing in impact strength may be due to the EFB fillers probably not well dispersed in the PVC-U matrix as the residence time decreased during processing. However, the addition of 9 phr of acrylic impact modifier in the S_{21} has improved the impact properties composites. Acrylic has capability to compensate the detrimental effect of EFB filler and thus improved the impact properties of the composites.

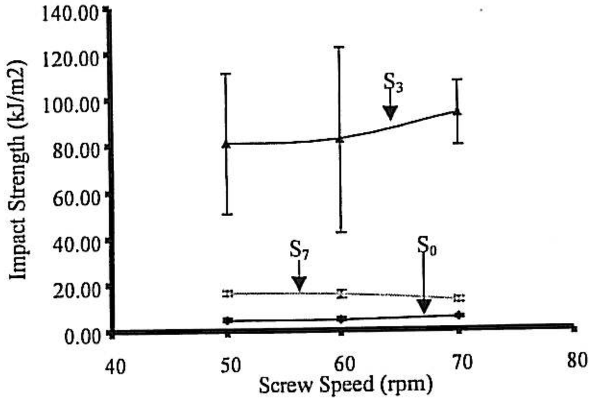


Figure 4.5: Effect of screw speed on the impact strength of S_0, S_3 and S_7.

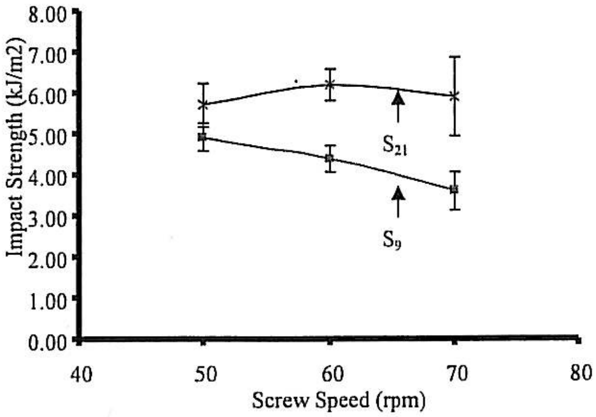


Figure 4.6: Effect of screw speed on the impact strength of S_9 and S_{21}

Figure 4.7 illustrates the effect of extrusion processing temperature on the impact strength of extrudates at extruder screw speed of 60 rpm. A number of samples of S_3 undergone hinge break at all die temperatures except at 195°C. Four out of seven samples were hinge break at temperature of 180 °C and 190 °C, but only two samples at temperature of 185 ° C. Generally, the impact strength of S_7 decreases as the temperature increased but its values still higher compared to other extrudates. Other extrudates such as S_0 and S_7 were completely broken at all processing temperatures. Figure 4.7 also shows that the optimum temperature of S_3 and S_7 extrudates to produce maximum impact strength is achieved at temperature of 180°C, whereas the optimum temperature of S_0 is 185°C. The shorter of fusion time and the higher heat of fusion S_3 and S_7 than S_0 (Table 4.12) show that the S_3 and S_7 firstly fused and firstly melted as temperature increased. It means that the contribution of primary particles to enhance the impact strength of S_3 and S_7 is firstly occurred to both compounds as compared to S_0 as temperature increases from 180 to 195°C. However, it shows that in general a moderate processing temperature is required to attain higher impact strengths. The results obtained are comparatively with other researchers (Benjamin, 1980, Gilbert 1985, Terselius, *et al.* 1985) who found that the maximum impact strength has also occurred at the moderate processing temperature.

According to Gilbert (1985), the reason for decreasing in impact strength at higher temperature was not fully understood but it was appeared to when the primary particles have disappeared to produce a reasonably homogeneous matrix. Perhaps the presence of some primary particles was beneficial in providing a mechanism for stress distribution. In case of impact modified whether coherent as for CPE modifier or dispersed particulate as for acrylic modifier should form a network embedding the primary particles. The network morphology was obtained at moderate processing temperatures. At higher temperatures the primary particles fused, the rubbery network was broken up and the impact strength was lost (Terselius, *et al.*1985).

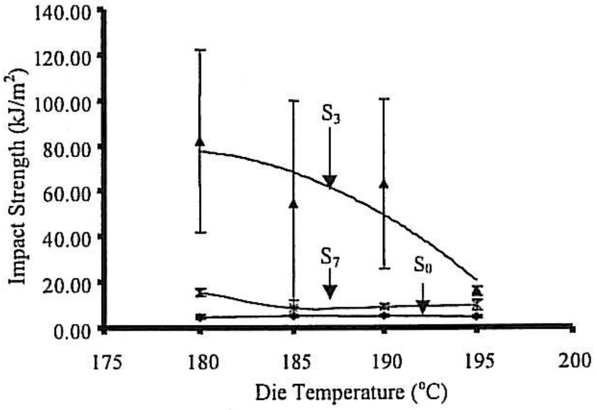


Figure 4.7: Effect of temperature on the impact strength of S_0, S_3 and S_7.

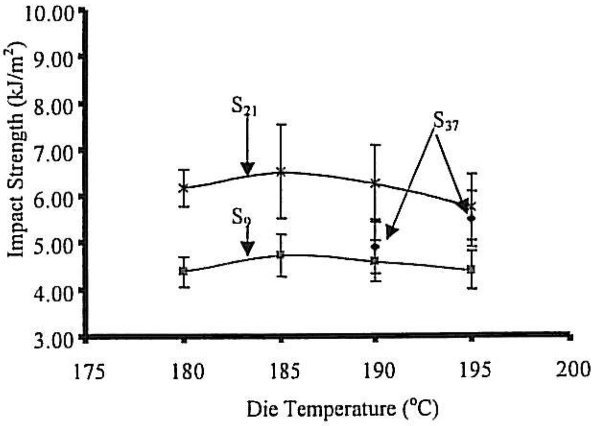


Figure 4.8: Effect of temperature on the impact strength of S_9,S_{21} and S_{37}.

Figure 4.8 shows the impact strength of composites (S_9, S_{21} and S_{37}) with respect to the extrusion processing temperature. It indicates that a maximum value of impact strength of composites is achieved, but its position is shifted to higher temperature as compared to impact modified compounds (S_3 and S_7) in Figure 4.7. The maximum impact strength of S_9 and S_{21} is achieved at temperature of 185°C. The incorporation of EFB filler (10 phr) delayed the fusion time of S_{21} and thus the contribution of primary particles to enhance the impact strength is more effective at 185°C. Meanwhile EFB filler in S_9 does not affect the optimum temperature at 185°C but decreased the impact strength of S_0.

The combination of 9 phr of CPE with 10 phr of EFB resulted in S_{37} could not be processed at temperatures of 180 °C and 185°C. This is due to the composite extrudate produced was formed melt bambooing just after out from the die as shown in Figure 4.2 .The combination increases the melt viscosity and retards the fusion process of PVC to occur thoroughly. Composite extrudate of S_{37} with smooth surfaces can be produced as temperature increased from 190 to 195°C. Meanwhile Figure 4.8 shows that the impact strength of S_{37} increases as processing temperature increased due to the increase of fusion level which is shown in Figure 4.19.

4.3 Flexural Properties

Figure 4.9 shows the effect of extruder screw speed on the flexural modulus of extrudates of S_0, S_3, S_7 , S_9 and S_{21}. The S_9 shows the greater flexural modulus. As mentioned in Chapter 3, the ability of EFB filler to increase the resistance of elastic deformation by restricting the PVC chains mobility could be one of the factors affected the composites modulus. However, the addition of 9 phr of acrylic (S_3 and S_{21}) or CPE (S_7) decreased the stiffness of PVC-U due to the softening effect that generated by both impact modifiers.

Figure 4.9 also shows that the flexural modulus of extrudates decreases with increasing of extruder screw speed. Decreasing in stiffness properties may be due to the increment of free volume of the matrix and void contents as the residence time decreased. This is resulted in the rigidity of matrix reduced. Besides that EFB fillers are not well dispersed in the PVC-U matrix as the screw speed increased. Therefore the standard deviation value for this composite is shown substantial scatter, particularly at 70 rpm as shown in Figure 4.9.

Figure 4.10 shows that EFB inability to increase the flexural strength of PVC-U. The randomly dispersed, cling together in bundles, weak filler-matrix interfaces and void contents are key factors for the contribution to the flexural strength reduction. As the screw speed increased those factors are exaggerated and brought to the great reduction in flexural strength. It can be seen that the flexural strength is drastically decreased as the screw speed increased from 60 to 70 rpm.

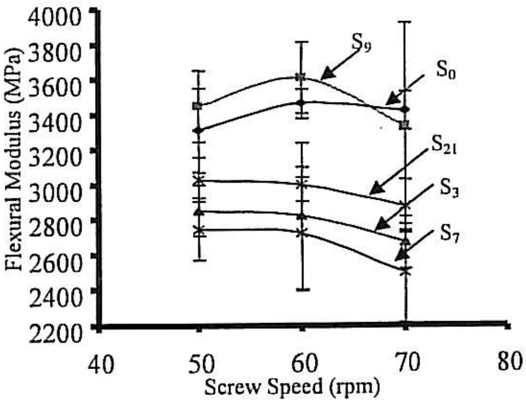


Figure 4.9: Effect of screw speed on the flexural modulus of S_0,S_3,S_7,S_9 and S_{21}.

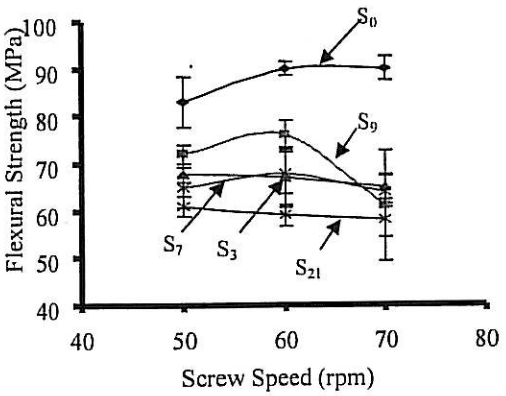


Figure 4.10: Effect of screw speed on the flexural strength of S_0,S_3,S_7,S_9 and S_{21}.

The effects of extrusion processing temperature on the flexural properties of extrudates are shown in Figure 4.11 and Figure 4.12. As expected, the incorporation of EFB filler is slightly increased the flexural modulus but decreased the flexural strength extrudates. Meanwhile the addition of impact modifier decreased the flexural properties of the extrudates.

The flexural modulus increased due to the mobility of PVC molecular chains are restricted by the fillers. This is because of the PVC free volumes are occupied by the fillers and consequently the stiffness of PVC increased. In case of flexural strength, the inability of EFB fillers to support the applied stress due to weak interface of filler-matrix is a most important factor to the reduction of flexural strength of PVC-U extrudate. The softening effect generated by impact modifier in increasing the flexibility of polymer matrix to enhance the toughness was adversely affected the flexural properties.

The flexural properties of extrudates are insignificantly affected as the processing temperature increased, although there is somewhat increment in flexural modulus while somewhat decrement in flexural strength. It is believed that the processing temperature is little affected to the flexural properties of extrudates.

Generally the flexural modulus of the PVC-U extrudate is more affected by processing temperature compared to other extrudates. This is because of the progressive fusion of primary crystallites, leading to the reduction of free volume and thus increased the modulus of PVC-U (Cora, *et al.* 1999).

The similar reason as mentioned above can also be applied for the increasing of flexural modulus of the impact modified PVC-U extrudates (Figure 4.11). In the compatible blends such as S_3 and S_7, the flexural modulus is also increased as the processing temperature increased. One possible explanation for this, the PVC may have more experienced the fusion. As result, the PVC primary particles had lost their particulate nature and became more miscible with the impact modifier. This result is in good agreement with the flexural modulus, showing that the modulus of the impact modified PVC-U extrudates slightly increased with the temperature (Wimolmala, *et al.* 2001).

Meanwhile, the flexural strength decreases as the processing temperature increased may be due to the presence of weak regions at the boundaries of the primary particles. The presence of weak regions of low entanglement density at the boundaries of the primary particles has been reported by Terselius *et al.* (1985). It may be assumed that weak region could act as stress concentrations during flexural testing. Thus, the network distribution of weak regions could locally initiate crack thereby decreased the flexural strength.

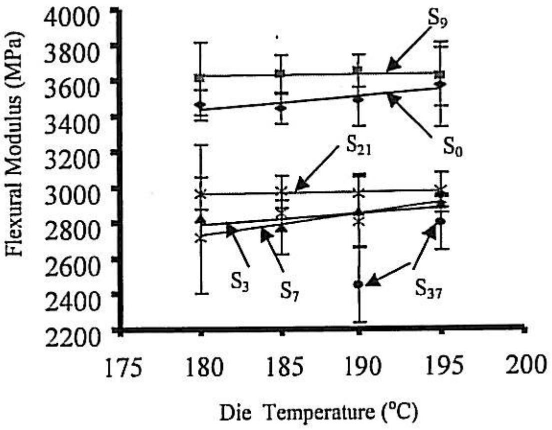


Figure 4.11: Effect of temperature on the flexural modulus of S_0,S_3,S_7,S_9,S_{21} and S_{37}.

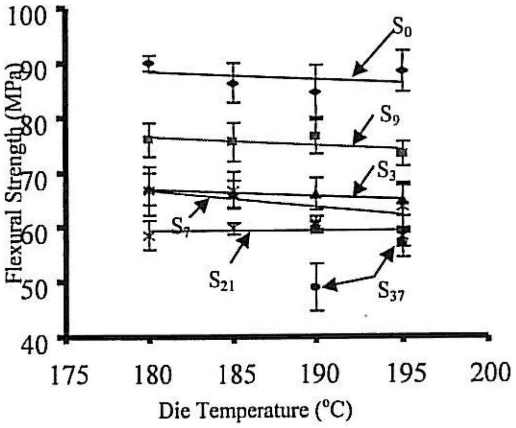


Figure 4.12 : Effect of temperature on the flexural strength of S_0,S_3,S_7,S_9,S_{21} and S_{37}.

4.4 Tensile Properties

Tensile test was done in order to determine the yield stress, strain at yield and strain at break of the extrudates with varying the extruder screw speed or processing temperature. The crosshead speed was fixed at 20 mmmin^{-1}

Table 4.3 and Table 4.4 show that the yield stress and yield strain of extrudates are insignificantly affected by the screw speeds. Meanwhile the yield stress

Table 4.3 and Table 4.4 show that the yield stress and yield strain of extrudates are insignificantly affected by the screw speeds. Meanwhile the yield stress and yield strain Table 4.5 and Table 4.6 show somewhat increment as temperature increased from 180°C to 185°C and thereafter nearly remains constant. It means that yield parameters, namely yield stress and yield strain are insensitive (independent) to the variations in the screw speeds and are more influenced by melt temperature. The domination of melt temperature and independent of screw speed on the PVC properties also reported by Summers, *et al.*(1982).

Table 4.3 and Table 4.5 show the yield stress of S_3, S_7, S_9 and S_{21} is lower compared to S_0. It indicates that impact modifier or EFB filler capable to reduce the yield stress of PVC in order for yielding process to occur. In case of impact modifier lowers the yield stress of the PVC, facilitating cold drawing and thus increasing energy absorption has also been reported by other researchers (Petrich, 1973, Stevenson, 1995 and Yanagase, 1996). The yield stress can be lowered by impact modification has also reported by Marshall (1982). Impact modifier provides sites of stress concentration that occur in the PVC matrix around an impact modifier particle upon application of the tensile stress to the material. The well dispersed of these localized concentrations throughout the PVC matrix at which shear yielding of polymer can be initiated easily upon tensile.

Meanwhile, in case of the effectiveness of EFB filler lowers the yield stress of PVC can be explained by the increase in the numbers of voids which act as stress concentration sites for yielding initiated more easily (Nakamura, *et al.* 1998 and Liang, *et al.* 2000). Unlike impact modifier (rubbery particles), fillers basically is not deformed as external stress applied to specimen because the modulus of filler was greater than that of the matrix. When the applied stress exceeds the interfacial adhesion strength between EFB and PVC, debonding at the interfaces will occur first, leading to the formation of micro-voids. In this case, the deformation restraint of the matrix around the filler is released, resulting in the shear yielding (shear yielding is favored than crazing due to poor interfacial bonding) and absorbing strain energy of PVC without blocking the propagation of cracks (Liang, *et al.* 2000). Therefore, although void can also provide stress concentrations to facilitate the shear yielding but it is still ineffectively as impact modifier. It elucidates that void can also act as a defect which indicates the break of the specimen. This observation appears to be the reason why the yield stress of PVC lowered as incorporated with EFB filler. Similar reason (void as a defect) can also be used to explain why the yield strain of S_9 and S_{21} as shown in Table 4.4 and Table 4.6 is lower compared to other extrudates.

The percentage strain at break which indicates of tensile toughness of the materials is shown in Table 4.7 and Table 4.8. Since the impact strength depends on the ductility of the specimen, the results obtained in these tables are well correlated with the impact strength results as shown in Figure 4.4, Figure 4.5, Figure 4.6 and Figure 4.7. The most of the maximum values of the impact strength are tally with the maximum values of strain at break for both effects, namely screw speed and processing temperature. In other words, the trend of impact strength values is similar with the trend values of strain at break.

The strain at break decreases with the incorporation EFB filler in the same manner as yield strain. Likewise to the yield strain, the detrimental effect of EFB filler and void on the S_9 and S_{21} are the main reasons for the strain break reduction compared to other extrudates. Meanwhile, as expected the addition of impact modifier in the S_3 ad S_7 has promoted the PVC matrix to experience cold drawing after yielding and thus increasing the energy absorption. The capability of impact modified PVC matrix to enhance the energy absorption by cold drawing (plastic deformation) has been discussed from various view points (Bucknall, 1977, Newman and Stella, 1965). More pertinent to the behaviour of impact modified PVC has been suggested by Newman and Stella. They suggested that cold drawing of the glassy matrix resulted in the increase of free volume of the glassy matrix around rubbery particle because of the resistance of the particle to change in volume. The expansion of matrix surrounding the particle generated heat that risen the temperature of the matrix so that it approached its glass transition temperature and deformed more easily and absorbed more energy in viscous flow process.

Table 4.3: Effect of extruder screw speed on the yield stress of extrudates at constant die temperature of 180°C.

Speed (rpm)	Yield Stress (MPa)				
	S_0	S_9	S_3	S_7	S_{21}
50	43.0 ± 3.7	38.9 ± 1.4	43.2 ± 1.2	41.6 ± 2.5	31.7 ± 1.4
60	43.8 ± 2.3	40.9 ± 1.3	42.1 ± 1.4	41.5 ± 0.4	34.3 ± 1.6
70	43.8 ± 2.7	40.4 ± 2.9	44.3 ± 1.8	43.4 ± 2.0	32.9 ± 2.6

Table 4.4: Effect of extruder screw speed on the yield strain of extrudates at constant die temperature of 180°C.

Speed (rpm)	Yield Strain (%)				
	S_0	S_9	S_3	S_7	S_{21}
50	6.0 ± 1.0	5.2 ± 0.9	9.0 ± 1.3	8.2 ± 1.7	4.6 ± 0.9
60	6.0 ± 0.2	5.1 ± 0.6	8.7 ± 1.0	8.4 ± 2.0	4.9 ± 0.6
70	6.8 ± 0.7	4.9 ± 0.4	8.8 ± 1.0	7.6 ± 1.4	4.9 ± 0.5

Table 4.5: Effect of processing temperature on the yield stress of extrudates at constant screw speed of 60 rpm

Die (°C)	Yield Stress (MPa)					
	S_0	S_9	S_3	S_7	S_{21}	S_{27}
180	43.8 ± 2.0	40.9 ± 1.0	42.1 ± 1.4	41.0 ± 1.7	34.3 ± 1.4	
185	55.4 ± 2.1	49.8 ± 1.9	44.1 ± 0.5	42.7 ± 3.9	39.8 ± 0.7	
190	57.6 ± 1.6	48.9 ± 1.0	47.3 ± 1.8	42.2 ± 2.7	38.9 ± 0.4	32.9+2.5
195	55.7 ± 3.4	47.7 ± 1.1	47.3 ± 1.5	44.5 + 1.0	38.9 ± 0.9	33.5+3.5

Table 4.6: Effect of processing temperature on the yield strain of extrudates at constant screw speed of 60 rpm.

Die (°C)	Yield Strain (%)					
	S_0	S_9	S_3	S_7	S_{21}	S_{27}
180	6.0 ± 0.4	5.1 ± 0.6	8.7 ± 1.0	8.4 ± 1.7	4.9 ± 0.6	
185	7.8 ± 0.7	6.0 ± 0.4	9.1 ± 0.6	8.9 ± 3.9	5.8 ± 1.0	
190	7.6 ± 0.5	5.6 ± 0.5	8.2 ± 0.9	8.0 ± 2.7	5.7 ± 0.9	6.3±0.7
195	7.1 ± 0.3	5.6 ± 0.5	9.0 ± 0.9	7.6 + 1.0	5.9 ± 0.7	6.8±0.7

Table 4.7: Effect of screw speed on the percentage strain at break at constant die temperature of 180°C.

Speed (rpm)	Strain at Break (%)				
	S_0	S_9	S_3	S_7	S_{21}
50	14.7 ± 7.1	6.9 ± 1.3	99.7 ± 65.4	23.5 ± 5.7	5.0 ± 1.0
60	10.7 ± 5.2	6.2 ± 0.8	132.1 ± 77.9	38.4 ± 3.0	4.9 ± 0.6
70	16.7 ± 6.2	6.0 ± 0.5	172.3 ± 57.3	36.9 ± 11.3	4.6 ± 0.9

Table 4.8: Effect of processing temperature on the percentage strain at break at constant screw speed of 60 rpm.

Die (°C)	Strain at Break (%)					
	S_0	S_9	S_3	S_7	S_{21}	S_{27}
180	10.7 ±5.1	6.2 ± 0.8	132.1 ± 65.4	38.4 ± 3.0	4.9 ± 0.6	
185	19.3 ± 5.0	7.6 ± 0.9	103.4 ± 77.9	21.1 ± 9.6	11.1 ± 1.8	
190	15.1 ± 1.7	6.0 ± 0.6	62.5 ± 57.3	19.4 ± 5.4	9.7 ± 2.1	8.0 ± 1.2
195	6.9± 0.7	6.9± 0.7	29.2±10.5	20.7+2.0	10.2+2.4	8.2 ± 1.8

4.4.1 Yield Stress Analysis

In this section, the yield stress analysis was done to predict the yield stress of the extrudates at impact velocity of 3 ms^{-1} which was used for pendulum impact testing in Izod mode. The samples were chosen from exrudates at constant screw speed of 60 rpm at 180°C and at 190°C. To obtain the yield stress at this velocity, thus a linear regression was performed on the yield stress data at lower strain rate (Table 4.6) corresponding with the Eyring theory of plastic yielding. This method has been previously done by other researchers to predict the yield stress values of impact-modified PVC-U at high strain rate (Calvert *et al.*1991, Azman Hassan,1996 and Wee, 2001).

The Eyring theory has described that the yield behavior of polymers was very dependent on temperature and rate of testing. In the high stress region where yield occurs the Eyring theory gives the yield stress (σ_y) in terms of the strain rate at yield ($\dot{\varepsilon}$) as :

$$\frac{\sigma_y}{T} = \frac{k}{\gamma\nu}\left[\frac{\ln \varepsilon}{A} + \frac{\Delta H}{kT}\right] \qquad 4.1$$

where, A is a constant, υ is the activation volume, ΔH is a activation energy for the low process, *k* is Boltzmann's constant, γ is a stress concentration factor and T is a absolute temperature. The plots of σ_y for all extrudates at temperature of 180 °C and 190 °C against natural log tensile strain rate are increased linearly as shown in Figure 4.13, Figure 4.14,Figure 4.15, Figure 4.16, Figure 4.17 and Figure 4.18. The equation obtained by linear regression and extrapolating is used to predict the σ_y .at constant testing temperature. The value of slope is represented for value of $k/\gamma\upsilon A$ while the intercept is represented for value of $\Delta H/k$. Thus, to obtain σ_y, the strain rate at impact velocity is firstly determined.

In order to determine the apparent strain rate of the impact test in three point bending, both equations from the flexural strength and flexural modulus are used and rearranged using Hooke's Law and resulting in the following equation:

$$\varepsilon = \frac{6\ vd}{L^2} \qquad 4.2$$

where v is the impact velocity, d is the mean thickness of the specimens of the sample and L is the span between the centers of support. By substituting the relevant data, the strain rate for impact velocity of 3 ms^{-1} was found to be $20.76s^{-1}$. It is substituted into the regression equation obtained from the graph. The predicted σ_y of extrudates at 3 ms^{-1} are shown in Table 4.10 and Table 4.11. From the results, the predicted yield stress showed that the incorporation of EFB filler and impact modifier is decreased the yield stress of the filled and impact modified PVC extrudates. The ability of EFB filler and impact modifier in reducing the yield stress is discussed previously.

The effect of strain rate on the yield stress of the extrudates can be seen in Table 4.9. It shows that the yield stress increases as the strain rate increased where the yield stress of S_0 arestill higher than others at all strain rate. The increase of yield stress as the strain rate increased was also found by Wee (2001). It indicates that the yielding process being suppressed with the increase of the strain rate, and thus the fracture will tend to change from ductile to brittle. It can be explained in molecular aspect, where during yielding the molecular chains are uncoiled and slip past each and gradually align closer in the reaction of the tensile stress. As the strain rate increased, the rate of extension molecular chains to align and stiff become faster and then the stress on the molecular chains increased. Eventually, the covalent bonds of the main chains break and fracture of the material occurred (Smith, 1996).

Table 4.9 : Effect of strain rate on the yield stress of extrudates

Crosshead speed (mm/min)	Strain Rate (s^{-1})	Strain Rate (ln)	Yield Stress (MPa)										
			S_0		S_9		S_3		S_7		S_{21}		S_{27}
			180 (°C)	190 (°C)	180 (°C)	190 (°C)	180 (°C)	190 (°C)	180 (°C)	190 (°C)	180 (°C)	190 (°C)	190 (°C)
5	0.001	-6.571	38.8	53.7	37.6	46.4	40.8	44.1	37.5	40.3	31.7	36.5	26.3
20	0.006	-5.185	43.8	57.6	40.9	48.9	42.1	47.3	41.0	42.2	34.3	38.9	26.9
80	0.022	-3.808	45.1	60.9	43.6	51.8	46.0	50.1	43.3	45.6	35.2	41.3	28.1
100	0.028	-3.583	45.2	61.7	44.1	52.2	46.6	49.3	42.8	46.2	37.2	41.7	29.0

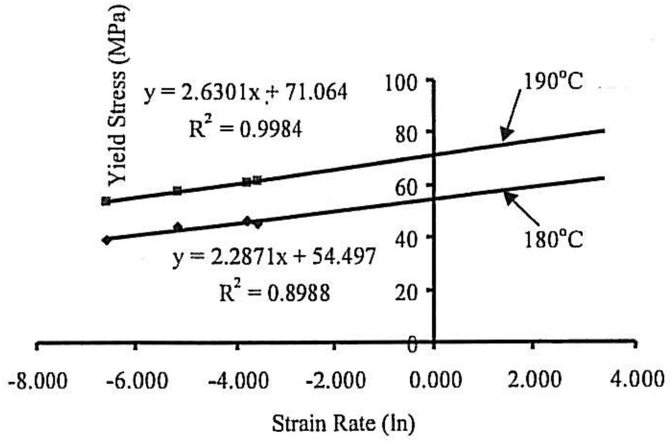


Figure 4.13: Predicted yield stress values at 3m/s for S_0

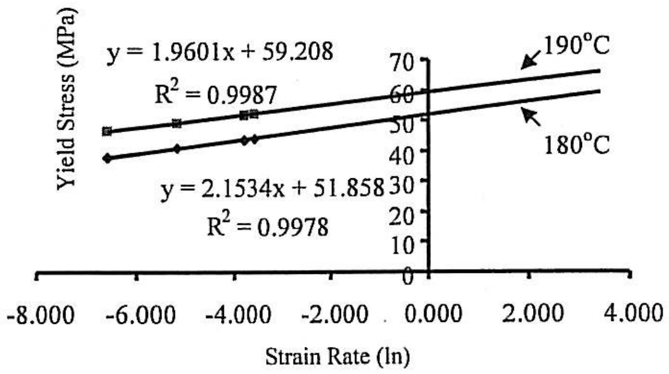


Figure 4.14 : Predicted yield stress values at 3 m/s for S_9

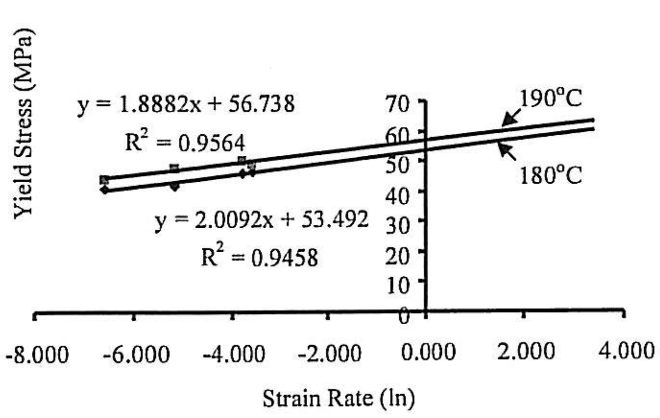


Figure 4.15 : Predicted yield stress values at 3 m/s for S_3

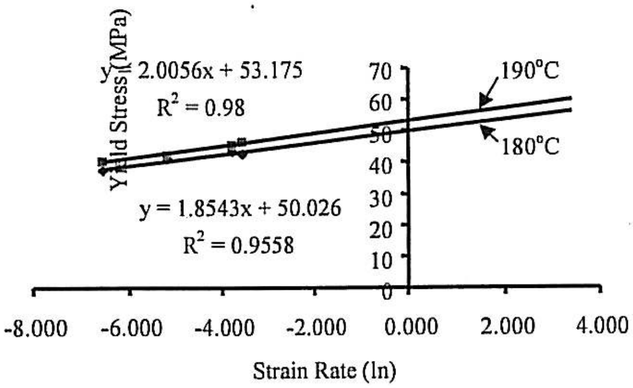


Figure 4.16: Predicted yield stress values at 3 m/s for S_7

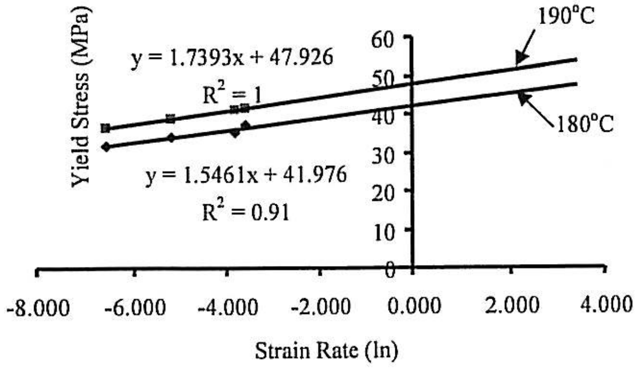


Figure 4.17: Predicted yield stress values at 3 m/s for S_{21}

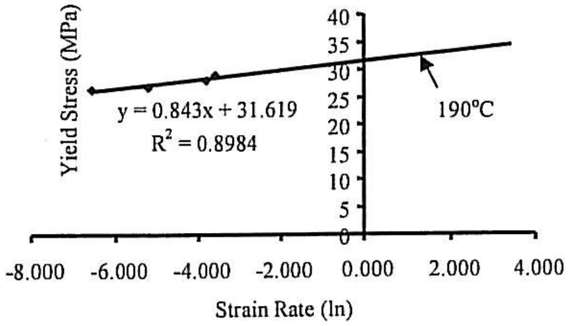


Figure 4.18 : Predicted yield stress values at 3 m/s for S_{27}

Based on the equations obtained from linear regression the predicated yield stress values of extrudates at impact velocity of 3 m/s or strain rate of 20.67s^{-1} is calculated.

Table 4.10: Predicted yield stress of extrudates at temperature of 180°C and screw speed of 60 rpm

Extrudate	Equation	Predicted Yield Stress (MPa)
S_0	$\sigma_y = 2.2871(\ln \varepsilon) + 54.497$	61.42
S_9	$\sigma_y = 2.1534(\ln \varepsilon) + 51.853$	58.38
S_3	$\sigma_y = 2.0092(\ln \varepsilon) + 53.492$	59.58
S_7	$\sigma_y = 1.8543(\ln \varepsilon) + 50.026$	55.64
S_{21}	$\sigma_y = 1.5461(\ln \varepsilon) + 41.976$	46.67

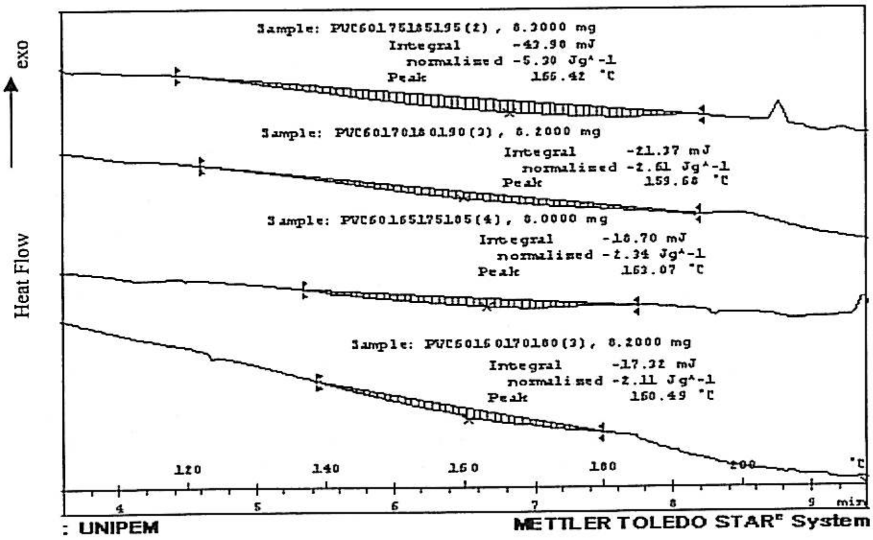
Table 4.11 : Predicted yield stress of extrudates at temperature of 180°C and screw speed of 60 rpm

Exrudate	Equation	Predicted Yield Stress (MPa)
S_0	$\sigma_y = 2.6301(\ln \varepsilon) + 71.064$	79.03
S_9	$\sigma_y = 1.9601(\ln \varepsilon) + 59.208$	65.15
S_3	$\sigma_y = 1.8882(\ln \varepsilon) + 56.738$	62.46
S_7	$\sigma_y = 2.0056(\ln \varepsilon) + 53.175$	59.25
S_{21}	$\sigma_y = 1.7393(\ln \varepsilon) + 47.926$	53.19
S_{27}	$\sigma_y = 0.8430(\ln \varepsilon) + 31.619$	34.17

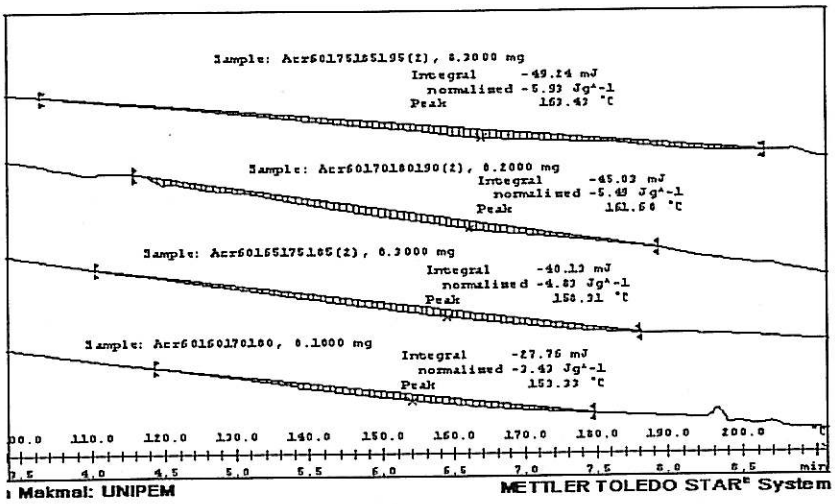
4.5 DSC Analysis.

As described by Summers (1982), the nature of PVC crystallinity played a significant role in the toughness of extruded products. During melt processing, the melt flow units are usually made of PVC primary particles. A PVC primary particle is composed of about a billion molecules of PVC held together by structure of crystallites and tie molecules. The crystallites act as crosslinks in a three-dimensional network. As melt temperature increases some primary crystallites experience melting and diffusion, and concomitant loss of order effectively blurs interparticle boundries. In this study this process was observable by DSC analysis.

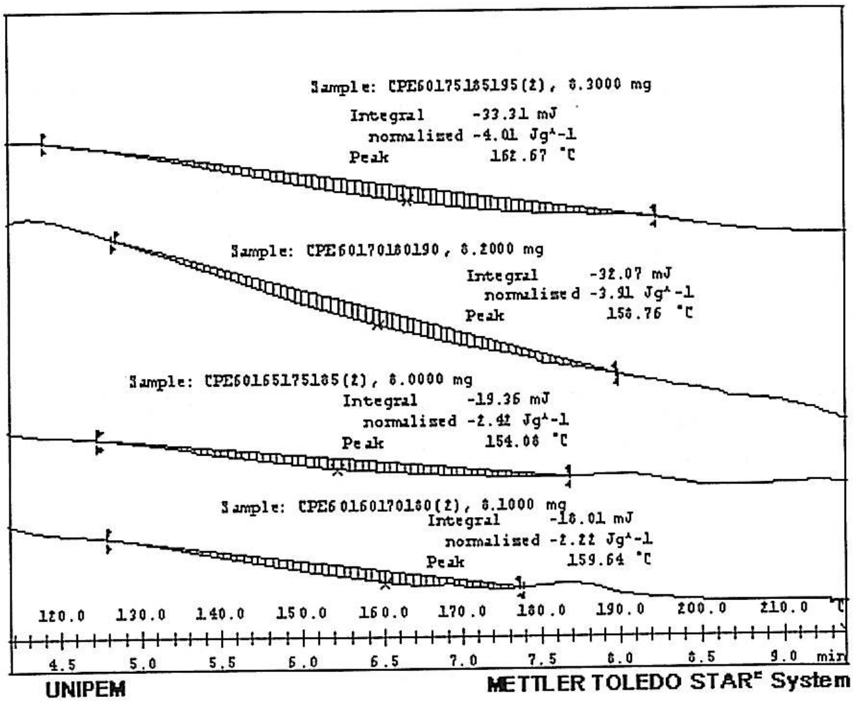
Figure 4.19 clearly reveals that the heat of fusion of endotherm is becoming bigger as the processing temperature increased. Similar results were also found other researchers (Gilbert *et al.* 1983). The increase of the area of endotherm (heat of fusion) is related to the increase of degree of fusion. As the fusion level increases, the three-dimensional network structure related to primary crystallites disappears progressively, leading to a matrix with an increasing number of free PVC molecules. It probably results in a more packed matrix structure with less free volume and any reduction in free volume should result in increase of the stress at which matrix yields (Cora *et al.*1999). The Table 4.10 and Table 4.11 (predicted) clearly shows that the stress at yield somewhat increases with increasing melt temperature whereas the impact strength of extrudates decreases. Although the results obtained are contradicted with other researchers as mentioned in Chapter 3 but in agreement with the results obtained by Cora *et al.* 1999)



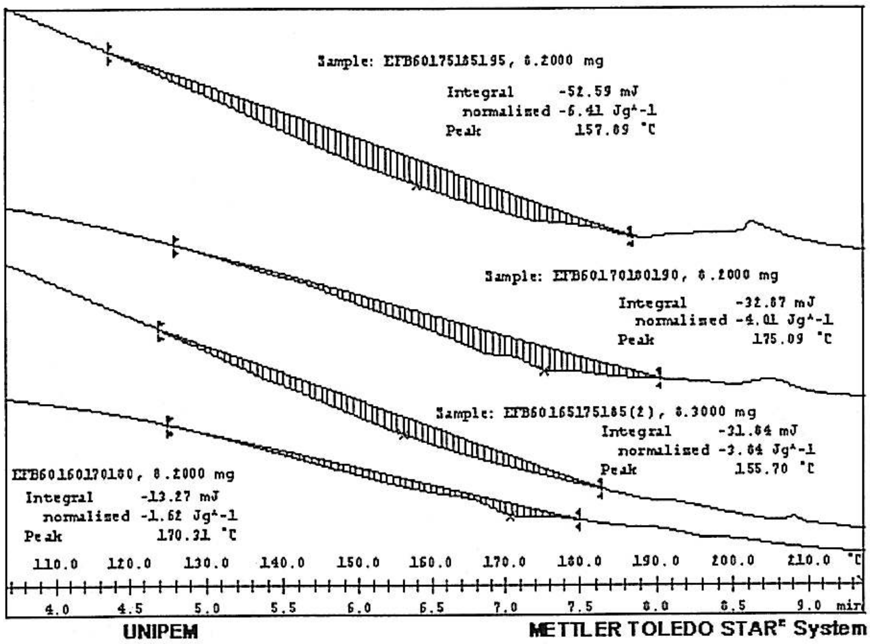
(a)


Sample: Acr60175185195(2), 8.3000 mg
Integral -49.24 mJ
normalised -5.93 Jg^-1
Peak 163.43 °C
Sample: Acr60170180190(2), 8.2000 mg
Integral -45.03 mJ
normalised -5.49 Jg^-1
Peak 161.66 °C
Sample: Acr60165175185(2), 8.3000 mg
Integral -40.13 mJ
normalised -4.83 Jg^-1
Peak 158.31 °C
Sample: Acr60160170180, 8.1000 mg
Integral -27.76 mJ
normalised -3.43 Jg^-1
Peak 153.33 °C
00.0
110.0
120.0
130.0
140.0
150.0
160.0
170.0
180.0
190.0
200.0
°C
.5
4.0
4.5
5.0
5.5
6.0
6.5
7.0
7.5
8.0
8.5
min
Makmal: UNIPEM
METTLER TOLEDO STAR^e System

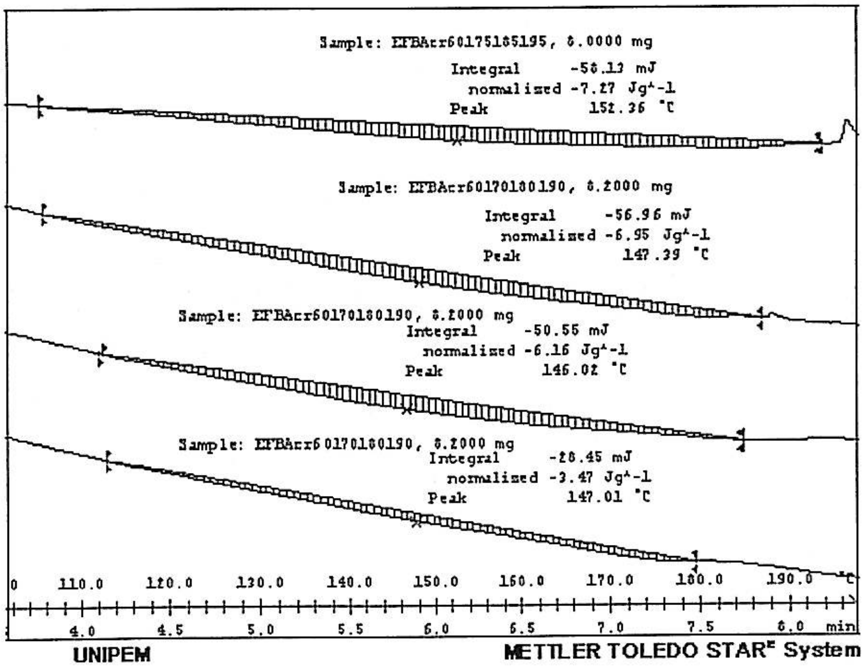
(b)


Sample: CPE60175185195(2), 8.3000 mg
Integral -33.31 mJ
normalised -4.01 Jg^-1
Peak 162.67 °C
Sample: CPE60170180190, 8.2000 mg
Integral -32.07 mJ
normalised -3.91 Jg^-1
Peak 158.76 °C
Sample: CPE60165175185(2), 8.0000 mg
Integral -19.36 mJ
normalised -2.42 Jg^-1
Peak 154.08 °C
Sample: CPE60160170180(2), 8.1000 mg
Integral -18.01 mJ
normalised -2.22 Jg^-1
Peak 159.64 °C
120.0
130.0
140.0
150.0
160.0
170.0
180.0
190.0
200.0
210.0
°C
4.5
5.0
5.5
6.0
6.5
7.0
7.5
8.0
8.5
9.0
min
UNIPEM
METTLER TOLEDO STAR^e System

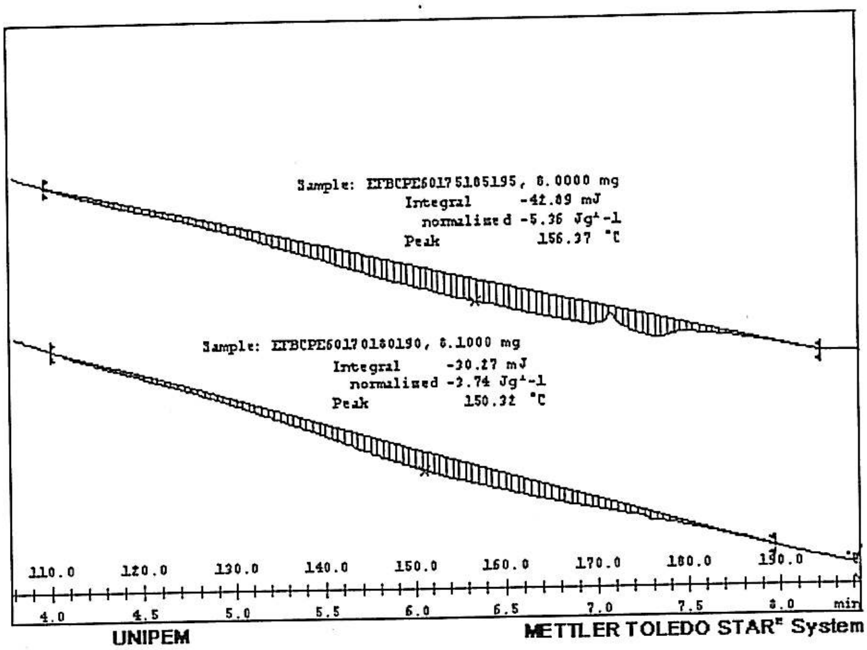
(c)


Sample: EFB60175185195, 8.2000 mg
Integral -52.59 mJ
normalized -6.41 Jg^-1
Peak 157.89 °C
Sample: EFB60170180190, 8.2000 mg
Integral -32.87 mJ
normalized -4.01 Jg^-1
Peak 175.09 °C
Sample: EFB60165175185(2), 8.3000 mg
Integral -31.84 mJ
normalized -3.84 Jg^-1
Peak 155.70 °C
EFB60160170180, 8.2000 mg
Integral -13.27 mJ
normalized -1.62 Jg^-1
Peak 170.31 °C
110.0
120.0
130.0
140.0
150.0
160.0
170.0
180.0
190.0
200.0
210.0
°C
4.0
4.5
5.0
5.5
6.0
6.5
7.0
7.5
8.0
8.5
9.0
min
UNIPEM
METTLER TOLEDO STAR^e System

(d)


Sample: EFBAcr60175185195, 8.0000 mg
Integral -58.13 mJ
normalized -7.27 Jg^-1
Peak 152.36 °C
Sample: EFBAcr60170180190, 8.2000 mg
Integral -56.96 mJ
normalized -6.95 Jg^-1
Peak 147.39 °C
Sample: EFBAcr60170180190, 8.2000 mg
Integral -50.55 mJ
normalized -6.16 Jg^-1
Peak 146.02 °C
Sample: EFBAcr60170180190, 8.2000 mg
Integral -28.45 mJ
normalized -3.47 Jg^-1
Peak 147.01 °C
0
110.0
120.0
130.0
140.0
150.0
160.0
170.0
180.0
190.0
°C
4.0
4.5
5.0
5.5
6.0
6.5
7.0
7.5
8.0
min
UNIPEM
METTLER TOLEDO STAR^e System

(e)



(f)

Figure 4.19: DSC thermograms for extrudates (a) S_0 (b) S_3 (c) S_7 (d) S_9 (e) S_{21} and (f) S_{37}

The fusion level of extrudates increases with increasing of processing temperature as shown in Table 4.12. The fusion levels are in range of 20-95 %. Both impact modifiers accelerated the fusion process of PVC but acrylic as mentioned previously is also more effective as a fusion promoter than CPE. The higher of heat of fusion of filled impact modified composites (S_{21}, S_{37}) than unfilled impact modified compounds (S_3,S_7) was expected.. The result evidently shows that the combination of 10 phr of EFB filler with 9 phr of acrylic (S_3) or with 9 phr of CPE (S_7) capable to accelerate the fusion time of the composite extrudates.

Table 4.12: Heat of fusion of extrudates at screw speed of 60 rpm

Die Temperature (°C)	Heat of Fusion (J/g)					
	S_0	S_3	S_7	S_9	S_{21}	S_{27}
180	2.23	4.15	2.68	1.91	4.52	
185	2.53	5.81	2.93	4.49	8.02	
190	2.82	5.49	4.72	4.72	9.05	4.89
195	5.71	5.93	4.82	7.73	9.38	6.92

CHAPTER 5

CONCLUSIONS

The mechanical properties and fusion analysis of extrudates were studied. Based on the results obtained and discussion, there are several conclusions can be made as follows:

(a) Only EFB-filled PVC-U composite incorporated with 10 phr of filler can be extruded with the smooth surfaces whereas at 20 phr the extrudate surfaces change to be rough. Poor distribution of filler throughout matrix due to the shorter of L/D of screw was attributed a main cause.

(b) The combination of 10 phr of EFB and 9 phr of CPE in the EFB-filled CPE impact modified PVC-U composite only can be extruded at die temperatures of 190 and 195$_0$C. The extrudates were experienced melt bambooing below these temperatures. The increase of melt viscosity was a main reason.

(c) The increases of screw speed were increased the impact strength of S_0, S_3 and S_{21} while decreased the impact strength of S_7 and S_9. Meanwhile the flexural properties extrudates decreased as the screw speed increased except for S_0. The yield stress and yield strain of the extrudates insignificantly affected by screw speeds.

(d) The maximum impact strength of S_3,S_7 was obtained at temperature of 180°C while S_0,S_9,S_{21} at temperature of 185°C. The flexural modulus slightly increased while the flexural strength slightly decreased with the increasing of temperature. The yield stress and yield strain slightly increased as temperature increased.

(e) The crosshead speed had a proportional relationship with yield stress where the predicted yield stress increased with crosshead speed. Likewise experimental yield stress, the predicted yield stress increased with temperature.

(f) The fusion level of extrudates increased with the increasing of temperature. EFB filler had capability to accelerate the fusion process of extrudates particularly combined with acrylic impact modifier.

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