

CHAPTER I

INTRODUCTION

The interest in fiber-reinforced composite materials has been growing steadily since the Second World War. Their potential in terms of specific strength, stiffness, and lightness makes them a competitive substitute for metal and wood in many applications. Today, glass fiber is the most widely used fiber for reinforcing composite materials. However, many studies (Sanadi *et al.* (1986), Jain *et al.* (1993), Mohanty *et al.* (1995), Zhu *et al.* (1995), and Devi *et al.* (1997)) have shown that natural cellulose fibers may be used as reinforcements in polymer composites. Natural fibers have advantages over glass fibers in being of lower cost, density, and abrasion, having excellent mechanical properties, and using less energy in production. There are however two major problems in the applications of cellulose fibers. One is the lack of adequate interaction between the cellulosic reinforcement and the polymer matrix. The other is the hygroscopic nature of the natural fibers which results in low durability. In this study, the use of kenaf fiber to reinforce polyester matrix is studied by using the hand lay-up method for preparation of the composites. The readily available silane coupling agents were used to treat the fiber surface. Two types of silane coupling agent with different organofunctional groups were selected to provide two different types of interactions with the polymer matrix. The effect of these different interactions on the strength of the composite will be studied.

1.1 Unsaturated Polyester Resin Matrix

Unsaturated polyester resins or polyester thermosets are polymers formed by condensation polymerization, with ester bonds linking the repeating units. The resins are able to form crosslinks because each repeat unit contains an active carbon-carbon double bond that can react with a crosslinking monomer (also unsaturated) by the addition polymerization mechanism. A repeat unit of an unsaturated polyester resin containing a carbon-carbon double bond is shown in Figure 1.1.

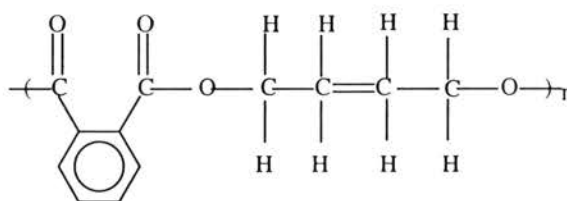


Figure 1.1 Segment of an unsaturated polyester resin chain, showing the active C=C site.

Reaction of carbon-carbon double bonds by addition polymerization requires that a free radical be formed and attack the double bond to initiate the reaction sequence. Usually a heat-activated or time-activated peroxide or some other free radical source is added to the resin as an initiator at a concentration of 1-2 % to initiate the crosslinking reaction. The resin is normally cured at room temperature followed by a post cure at higher temperature to complete the reaction. In general, direct bonding between polyester molecules are difficult because of the steric hindrance of the bulky phenylene group. Therefore, solvents are added to allow the polymer molecules to move more freely, thus facilitating the crosslinking reaction. The most common solvents used are styrene, vinyl toluene, and methyl methacrylate. These solvents also act as a cross-linking agent for the resin as shown in Figure 1.2.

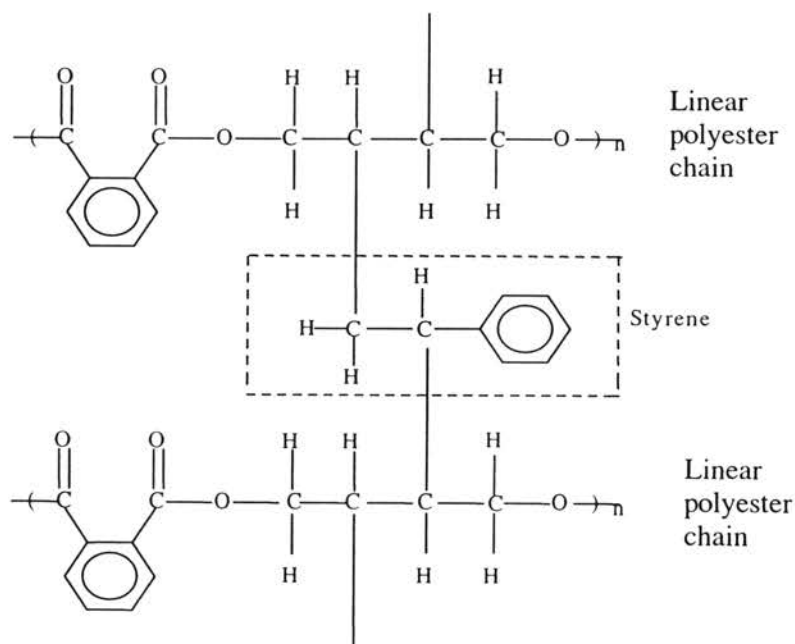


Figure 1.2 Crosslinking of linear unsaturated polyester resin with styrene, (Strong, 1996).

1.2 Kenaf Fiber

1.2.1 Structure

Kenaf is a kind of bast fiber obtained from the stem of the plant *Hibiscus sabdariffa* Linn. *Var. altissima*. The fiber bundle consists of strands of individual fibers held together by intercellular material. The fiber strands are usually 1.5 to 2 m long. The cross section of each individual fiber is a five- or six-sided polygon with a central oval lumen. The secondary wall of individual fibers is fibrillar in nature with the fibrils helically disposed along the length of the fiber although with a smaller helical angle and with no structural reversals along the fiber length as found in cotton, (Mark *et al.*,

1989). Moreover, fibers are not convoluted so the spiral angle is not distorted with respect to the fiber axis as in cotton. Therefore bast fibers are generally stronger and stiffer with less extensibility than cotton. The morphology of kenaf fibers is similar to wood fibers, as depicted in Figure 1.3.

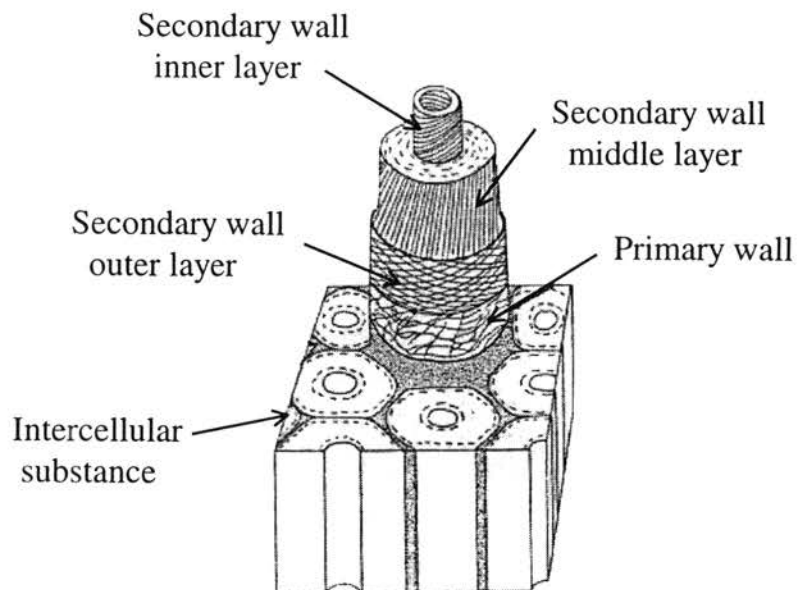


Figure 1.3 Schematic representation of the fine structure of a wood fiber, (Mark *et al.*, 1989).

1.2.2 Chemical Composition

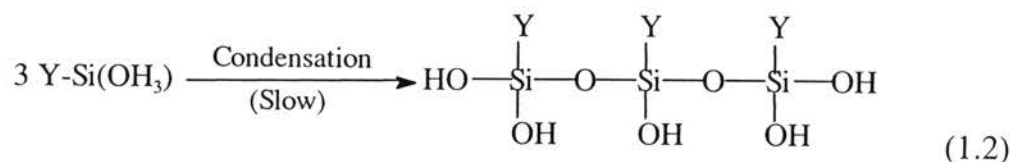
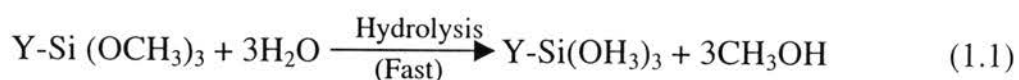
Individual kenaf fibers consist mainly of cellulose with intercellular materials binding them together. The intercellular materials is composed mainly of hemicellulose and lignin with a small amount of pectin and other extractives (Mark *et al.*, 1989). The chemical composition of kenaf fibers is shown in Table 1.1.

Table 1.1 Chemical composition of kenaf fibers, wt%.

Cellulose	Hemicellulose	Lignin	Pectin	Extractives
72.8	13.4	11.8	0.2	1.8

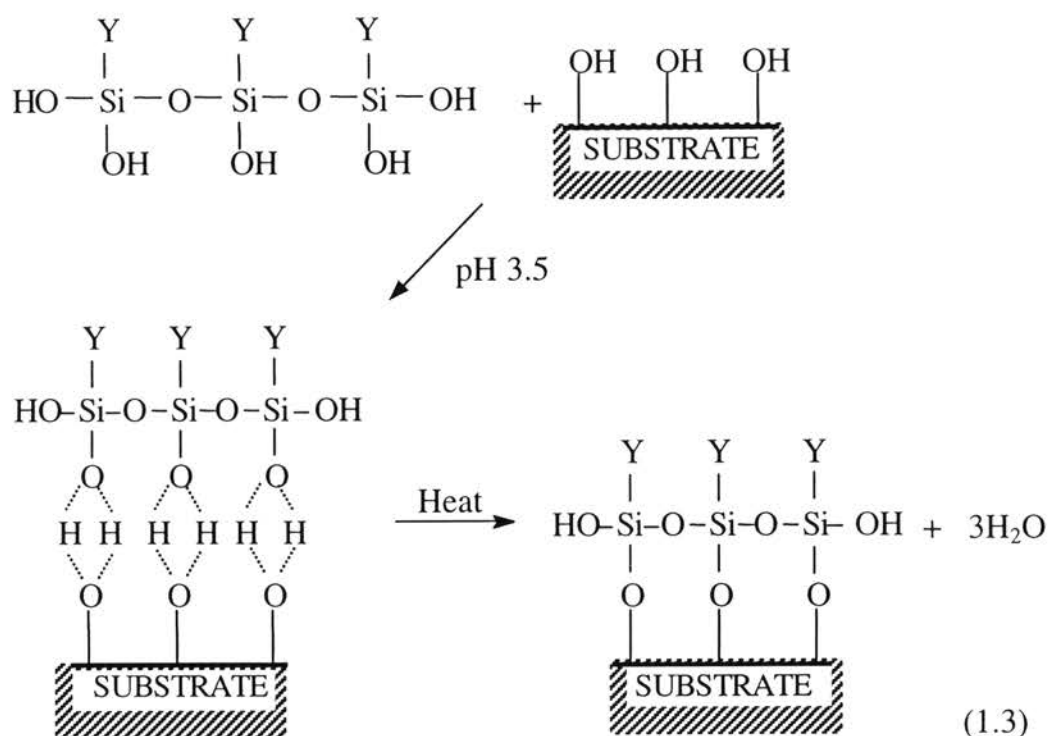
1.3 Organosilane Coupling Agents

In composite materials, coupling agents are generally employed to improve the bonding between the polymer matrix and the reinforcing fibers. Organosilane coupling agents are widely used for glass fiber reinforcement. They have the general structure of $Y-Si(X)_3$, where Y is an alkyl or group containing an alkyl function and X is a hydrolyzable group such as alkoxy or halide group. These coupling agents are used in the form of silane triols after the hydrolysis of X, which is fast in dilute acid (Eq. 1.1). The silanols that form have to be used quickly to avoid condensation which will lead to the formation of polysiloxane oligomers (Eq. 1.2). These oligomers are insoluble in aqueous medium and will precipitate out.



Silanetriols can react directly with the hydroxyl groups of a substrate to form oxane bonds or they may condense with the surface hydroxyl groups, and with each other, to form an organosilicone-bonded surface (Eq. 1.3). The siloxane thus formed on the substrate surface can act as a linking agent



between fiber and polymer matrix via the interaction between organofunctional groups (Y) and polymer matrix.



The organofunctional group, Y, in the coupling agent can interact with the polymer matrix to form an IPN (interpenetrating network). The final curing reaction of a silane treated substrate in a composite enhances the adhesion of the polymer matrix to the fiber substrate. Structures of some commercial silane coupling agents are presented in Table 1.2. In composite processing, the Y group should be chosen according to its chemical reactivity with the resin for maximum interaction. In the present work, styrene-diluted polyester resin was used as the polymer matrix in the composite. Plueddemann (1982) found that in such a system the performance of silanes showed good correlation with their relative reactivity in copolymerizing with styrene. Therefore, γ -methacryloxypropyltrimethoxysilane (γ -MPS) and vinyltriethoxysilane

(VTS) were chosen for this work as representatives of high and low activity silanes respectively.

Table 1.2 Characteristics of representative commercial silane coupling agents.

Organofunctional group	Chemical structure	Applied polymers
Vinyl	$\text{CH}_2=\text{CHSi}(\text{OCH}_3)_3$	UP
Vinyl	$\text{CH}_2=\text{CHSi}(\text{OC}_2\text{H}_5)_3$	UP, PE, DAP, PP, EPM, EPM
Chloropropyl	$\text{ClCH}_2\text{CH}_2\text{CH}_2\text{Si}(\text{OCH}_3)_3$	EP
Epoxy	 $\text{CH}_2\text{CH}(\text{O})\text{CH}_2\text{OCH}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$	EP, PA, PC, PF, PVC, PUR
Methacrylate	 $\text{CH}_2=\text{C}(\text{CH}_3)\text{COO}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$	UP, PE, PP, DAP, EPM, EPDA
Primary amine	$\text{H}_2\text{N}(\text{CH}_2)_3\text{Si}(\text{OC}_2\text{H}_5)_3$	UP, PA, PC, PI, MF, PF, PUR, MPF
Diamine	$\text{H}_2\text{N}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$	UP, PA, PC, PI, MF, PF, PUR, MPF
Cationic styryl	$\text{CH}_2=\text{CHC}_6\text{H}_4\text{CH}_2\text{N}^+\text{H}_2(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3\text{Cl}^-$	All polymers
Phenyl	$\text{C}_6\text{H}_5\text{Si}(\text{OCH}_3)_3$	PS, addition to amino silanes
Mercapto	$\text{HS}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$	EP, EPDM, SBR, PUR

1.4 Composite Material

A composite material can be defined as a material in which strong, high modulus fibers, with diameters of the order of 10 - 100 μm , are embedded in a polymeric matrix material. The objective is to obtain improved mechanical or physical properties that neither individual component has or to compensate for certain shortcomings of the individual materials.

The matrix generally performs the function of a binder to transfer stress to the reinforcing fibers and ensure their cooperative interaction. Both thermosetting and thermoplastic polymers can be used as the matrix for polymer-based composites.

Reinforcing fibers can either be man-made fibers or vegetable fibers, although glass fiber is the most widely used reinforcing agent at present. The strength of the composite is the combination of the strength of the reinforcing fiber and the matrix.

1.5 Hand Lay-Up Process

This method is the most widely used processing method for molding fiber reinforced materials. Its major advantage is that it is a very simple process with no special equipment needed. The first step is to coat the mold with releasing agent such as wax and polyvinyl alcohol (PVA) to prevent the molding sticking to it. This is followed by brushing catalysed resin onto the mold and then placing layers of fiber mat in the mold by hand with resin applied to each layer in succession. In this step, a brush and roller are used to consolidate the mat and remove entrapped air. The mold is then closed and the resin allowed to cure at room temperature, but heat is sometimes applied to the

mold in order to accelerate the curing process. The hand lay-up method was employed in the present study.