

PART 1

# Fundamentals of Constituents for Composites Manufacturing

# Introduction

Advanced composite materials have been used to fabricate many structural parts in engineering applications. This is due to their many attractive characteristics such as light weight, high strength, high stiffness, good fatigue resistance and good corrosion resistance. Also, the ability to manufacture parts with complicated geometry using fewer components enables manufacturers to save cost as compared with the same parts made of conventional metallic materials. Before presenting the fundamental aspects of manufacturing and different techniques used for composites manufacturing, it is appropriate to present composite structural parts currently in use and the main techniques that have been used to fabricate them.

#### 1. EXAMPLES OF PRODUCTS MADE USING DIFFERENT MANUFACTURING TECHNIQUES

Figure 1.1(a) shows a schematic of an Airbus 380 airplane (the largest airplane in the world as of 2008). This airplane has more than 50% of its structure made of composite materials. These components include the flaps, ailerons, rudder, radome etc. Most of these components are flat in shape and they are usually made using hand-lay-up (HLU) and autoclave molding techniques. Figure 1.1(b) shows a schematic of the hand-lay-up fabrication technique and a representative lay-up sequence. Autoclave molding is a well-established method for composites used in the aero-space industry with certified resins and fibers. A photograph of an autoclave is shown in Figure 1.1(c). Autoclave Molding will be discussed in detail in Chapter 4.



FIGURE 1.1(a) Airbus 380 with its composite component (from http://www. specialchem4adhesives.com/home/editorial.aspx?id=752).



*FIGURE 1.1(b)* Schematic of the hand-lay-up fabrication method and a representative lay-up sequence. Individual layers can be cut by hand or by a computerized machine cutter. The layers can be stacked one on top of the other by hand or by a robot.



FIGURE 1.1(c) Photograph of an autoclave (courtesy of ASC Ltd.).

Figure 1.2(a) shows a pressure vessel made of composite materials using the combination of hand-lay-up and filament winding processes. Composite pressure vessels are light weight and can contain pressures higher than those contained by metallic vessels. These components are made using the filament winding process [Figure 1.2(b)]. Figure 1.2(c) shows a photograph of a filament winding machine. The filament winding process will be discussed in detail in Chapter 5.



*FIGURE 1.2(a)* Composite pressure vessel made by combination of hand-lay-up and filament winding.



FIGURE 1.2(b) Schematic of the filament winding process (courtesy of Wiley Interscience).



*FIGURE 1.2(c)* A two-spindle winder with a carriage-mounted resin bath and a free-standing creel in the background (courtesy of *Composites Technology* magazine, August 2005).



FIGURE 1.3(a) A composite pultruded connector.

Figure 1.3(a) shows a component made using pultrusion. Pultrusion is used to make many structures for civil engineering applications. Figure 1.3(b) shows the schematic of the pultrusion process, and Figure 1.3(c) shows a photograph of a lab scale pultrusion machine. Pultrusion will be discussed in Chapter 6.

Figure 1.4(a) shows a composite component made using the liquid composite molding (LCM) method (5 piece). LCM has been used to make automobile composite components. Figure 1.4(b) shows a schematic of the liquid composite molding process and Figure 1.4(c) shows a pump, a mold and accessories for the liquid composite molding hardware. Liquid composite molding will be discussed in Chapter 7.



FIGURE 1.3(b) Schematic of the pultrusion process (courtesy of Springer).



FIGURE 1.3(c) A lab pultrusion machine.



FIGURE 1.4(a) A curved piece made by Liquid Composite Molding (LCM) method.



FIGURE 1.4(b) Schematic of the liquid composite molding process.



*FIGURE 1.4(c)* Instrumentation for LCM: pump, mold and accessories. Resin is filled into the vertical cylinder, then pumped into the mold cavity on the left-hand side.



FIGURE 1.5(a) A thermoplastic composite wing box panel made by compression molding.

Figure 1.5(a) shows a composite wing box panel made using thermoplastic composites and compression molding method. Figure 1.5(b) shows the schematic for the thermoplastic composite molding process. Figure 1.5(c) shows a compression molding machine. Molding of thermoplastic composites will be discussed in Chapter 8.



 Male mold on the bottom
 Female mold on the bottom

 FIGURE 1.5(b)
 Schematic of the thermoplastic composite molding process.



FIGURE 1.5(c) A compression molding machine.

Figure 1.6(a) shows a thermoplastic composite tube made by the fiber placement process. Figure 1.6(b) shows the schematic of the thermoplastic composite placement process, and Figure 1.6(c) shows a photograph of a fiber placement machine. Fiber placement of thermoplastic composites will be discussed in Chapter 8.



FIGURE 1.6(a) A thermoplastic composite tube made by the fiber placement process.



FIGURE 1.6(b) Schematic of the thermoplastic composite placement process.



*FIGURE 1.6(c)* A fiber placement machine (courtesy of Aerospace Manufacturing Technology Center, National Research Council of Canada).

A few specific features can be extracted from the above components and the different manufacturing techniques used to fabricate them.

Normally structural components can be classified according to their shape, and the manufacturing technique used depends significantly on the shape of the component as follows:

- *Relatively thin flat plate or shallow shell with free edges.* Normally aerospace components have these types of shapes. These are usually made using the hand-lay-up method. The autoclave is the common tool used for making aerospace composite components having these shapes.
- Components of revolution, such as cylindrical or spherical pressure vessels and pipes. These structures usually have no free edges (except for the end openings). These are usually made using the filament winding method.
- Components having constant cross section such as tubes, rods, or even components with complex but constant cross section along the length such as door frames. These are usually made using the pultrusion method.
- *Components having complex 3-D configurations.* These can be thick or thin. These are usually made using the liquid composite molding (LCM) method.
- *Large structures such as boat hulls, wind turbine blades* etc. These are made using a modified form of LCM such as vacuum-assisted LCM. A special process called SCRIMP (seaman composite resin infusion molding process) is usually used to make boat hulls.
- *Small and large components, either without free edges or with free edges.* These can be made by the fiber placement method. These machines are versatile but require a large amount of capital investment (on the order of several millions of dollars).

# 2. GENERAL CHARACTERISTICS OF MANUFACTURING USING COMPOSITES

Generally, manufacturing using composites involves the processing of two main ingredient materials to make a final product. The ingredients involve the matrix and fiber materials. This processing requires the following:

· Good bonding between matrix and fibers

- Proper orientation of the fibers
- · Good amount of volume fraction of fibers
- Uniform distribution of fibers within the matrix material
- · Proper curing or solidification of the resin
- · Limited amount of voids and defects
- · Good dimensional control for the final part

The implications of the above are as follows.

Good bonding between matrix and fibers. To provide reinforcement so that properties such as strength and stiffness can be enhanced, the fibers need to be bonded to the matrix. If at a certain location, the fibers are not properly bonded to the matrix, dry spots will occur. At this location, there is no proper shear transfer of load between fiber and matrix and the domino effect (as will be discussed in Section 3.1 of this chapter) will occur. These locations will also serve as nuclei for cracks to form. However, there are situations, such as the requirement to absorb impact energy, where partial dry spots may enhance the energy absorbing capability of the composite.

Proper orientation of the fibers. Proper orientation of the fibers is important since properties such as stiffness and strength are very sensitive to fiber orientation. If the fiber orientation deviates by about  $10^{\circ}$  from the  $0^{\circ}$  direction, the stiffness can drop by more than 30%. Fiber orientation may be deviated from the intended orientation due to improper position of the layer during the lay-up step, or due to the flow of liquid resin that pushes the fibers during the filling period in liquid composite molding.

Good amount of volume fraction of fibers. In composite materials, the fibers provide stiffness and strength. Therefore the greater the amount of fibers, the better will be these properties. The amount of fibers is usually expressed in terms of volume fraction,  $v_f$  which is defined as:

$$v_f = \frac{V_f}{V_c} \tag{1.1}$$

where  $V_f$  is volume of fibers and  $V_c$  is volume of the composite material.

Properties such as stiffness of a unidirectional composite along the axial direction is given by the rule of mixtures:

$$E_c = E_f v_f + v_m E_m \tag{1.2}$$

Where subscript *f* refers to fiber, and *m* refers to matrix.

The fiber volume fraction and matrix volume fraction are related by:

$$1 = v_f + v_m + v_v \tag{1.3}$$

Where the last term refers to volume fraction of voids.

For good quality composites, the amount of voids should be minimum (less than 1%) and Equation (1.3) can be approximated to be

$$1 = v_f + v_m \tag{1.4}$$

Using Equation (1.4) in Equation (1.2), the modulus is expressed as:

$$E_{c} = (E_{f} - E_{m})v_{f} + E_{m}$$
(1.5)

The modulus is linearly proportional to the fiber volume fraction. Therefore the larger the fiber volume fraction, the better the mechanical properties. It should be noted, however that the fiber volume fraction cannot be 1 because this would mean that there is no matrix material which, in turn, would mean dry bundles of fibers and the domino effect as mentioned in Section 3.1 will prevail.

Uniform distribution of fibers within the matrix material. Figure 1.14 shows a cross section of a unidirectional composite layer. The white dots show the cross section of the fibers and the dark area represents the matrix. One can see that at the fine scale, the distribution of the fibers is uniform in some region but not in other regions. The region where there is more matrix than fiber is called a *resin rich area*. It is not a good idea to have large or many resin rich regions because there will also be weak areas. Under loading, these areas can serve as locations for crack to nucleate.

*Proper curing of the resin.* In the manufacturing of polymer matrix composites, the resin first occurs in the form of low viscosity liquid so that it can flow and wet the surface of the fibers. After wetting has been completed, the resin needs to solidify and harden. For thermoset resin, this is called *curing*; and for thermoplastic resin, this is called *solidifica*-

tion. In both cases, the resin needs to be hard and stiff for the reinforcement effect to take place. If there are regions where the resin is not hard enough, they will be weak and can serve as crack nucleation areas.

Limited amount of voids and defects. Voids and defects may be formed during the manufacturing of composites. Voids can arise due to lack of compaction of many layers together, or due to low pressure in the resin during curing. The amount of voids needs to be a minimum to be acceptable. A limit of about 1% is commonly used. Defects such as delamination between layers, cracks, fiber mis-orientation, or nonuniform fiber distribution may not be acceptable.

Good dimensional control for the final part. Polymeric resins shrink when they change from liquid state to solid state. The degree of shrinkage can be between about 5%–8% depending on the type of materials. This shrinkage of the material may cause residual stresses in the part, and also out-of dimensions or warping. For a large structure such as the wing of an aircraft, a few percentages of shrinkage of the material can translate into significant deformation of the structure. Another problem that may occur is the surface finish of parts such as automobile panels which may be adversely affected by this shrinkage. Resins with Low Profile Additives are usually used to control shrinkage.

## 2.1 Metal versus Composite Manufacturing

Manufacturing using composites has differences from manufacturing using metals:

- In metals such as steel or aluminum, materials with finished form such as rods, slabs, or sheets are available. The making of a finished product such as a car body or the box frame for a computer only requires working on these finished forms. Processes such as cutting, bending, forming, welding, or drilling are used on these finished forms to make the finished product.
- In composites, the steps that transform the finished form to the final structure are usually bypassed. A manufacturer using composite materials has to work directly from the ingredients of fiber and matrix to make the finished product itself. Figure 1.7(a) shows the different stages of existence of composite constituents up to the final product:

--Stage a: At this stage, the materials appear in raw basic form. For fibers, these consist of fiber either in the form of filaments or fi-



FIGURE 1.7(a) Stages of existence of constituents in the manufacturing of composites.

ber bundles. Fibers may also be woven into fabrics or braided into braided perform. For matrix, the material usually appears in liquid form for thermoset resin or in granular form in the case of thermoplastics.

- --Stage c: At this stage, the layers in stage b are stacked on top of each other to make flat plate laminates. This intermediate step is important for the analysis where material properties are tested or calculated. However this step is usually bypassed in the manufacturing process of practical composite parts.
- *—Stage d:* This is the final stage where the final product configuration is formed.

The involvement of these stages in the different manufacturing processes is as follows:

- Hand-lay-up (with or without autoclave): Stages *a*, *b* and *d* are involved. Stage *c* is bypassed.
- Filament winding: Stages *a* and *d* are involved. Stages *b* and *c* are bypassed.
- Pultrusion: Stages *a* and *d* are involved. Stages *b* and *c* are bypassed.
- Liquid composite molding: Stages *a* and *d* are involved. Stages *b* and *c* are bypassed [Figure 1.7(b) shows stages for Liquid Composite Molding].
- Thermoplastic composites: Stages *a* and *d* are involved. Sometimes stage *b* and even stage *c* may be involved.

The mentality of working with metals therefore cannot be applied when manufacturing using composites.



FIGURE 1.7(b) Stages of existence of constituents in the LCM process.

# 3. FUNCTIONS OF THE CONSTITUENTS OF COMPOSITES

There are two main constituents making up advanced composites. These are fibers and matrix. The interface between the fiber and matrix is critical for the function of the composite material. The interface may be considered as a third constituent of the material. Each of these constituents will be presented in the following.

#### 3.1. Fibers

Fibers provide strength and stiffness to the composite materials. Fiber materials are usually glass, carbon or Kevlar. One may ask the question why do composites appear in fiber form. There are many reasons for this as follows.

#### 3.1.1. Advantages of the Fiber Form

#### 3.1.1.1. Strength of Material in Fiber Format is Better as Compared to Bulk Format

Materials can appear in different forms. These can be bulk form (relatively large volume), fiber form (diameter of about 10  $\mu$ m and length from a few millimeters to a few meters) powder form (more spherical shape with diameter on the order of micrometers), or flake form (thin sheets). As a rule, the smaller the volume of a certain piece of material, the less defects there are in that volume, because there is less chance for defects to occur when a smaller volume of material is made. As such, bulk-form pieces have smaller strength than fiber-form pieces. The difference in strength of materials in bulk form and in fiber form is illustrated by the comparison between the properties of glass in fiber form and in plate (bulk) form. While the moduli of E glass (72 GPa) and plate glass (70 GPa) are about the same, their strengths are very different. E glass has a strength of 3448 MPa (and S glass has a strength of 4585 MPa) while plate glass has a strength of only 70 MPa.

Materials in powder form do have small volume, however, the reinforcement effect is not as good as that in fiber form. This is because the reinforcement effect depends on the aspect ratio (ratio of l/d) where l is the length of the reinforcement and d is its diameter. If the aspect ratio is smaller than a certain critical value (under uniform shear stress distribution assumption, equal to  $\sigma/2\tau$  where  $\sigma$  is the tensile strength of the reinforcement material and  $\tau$  is the shear bond strength between the

reinforcement and the matrix), failure will occur due to slipping between the reinforcement and the matrix, making the reinforcement ineffective. For the case of glass/epoxy where  $\sigma = 3448$  MPa and  $\tau = 20.5$  MPa, the critical aspect ratio is 84. Reinforcements in powder form having aspect ratios on the order of 2 or 3 do not give the same reinforcement effect as fibers with small diameter and long length. If the diameter of a fiber is about 10 µm, then a length of about 1 mm would be sufficient. However due to stress magnification at the end of the fibers, the smaller the number of ends of the fibers, the better the reinforcement effect. As such, long continuous fibers give better reinforcement than short fibers.

Materials in flake form are also available (such as mica or clay sheets). However these usually occur naturally and are limited in their variety.

#### 3.1.1.2. Availability of More Fabrication Techniques

The fiber format allows fiber processing steps that are difficult or impossible in bulk. Examples of this are stretching and orientation (carbon, polyaramide, Kevlar, and polyethylene fibers), vapor deposition (boron fibers), solvent removal (polyaramide and kevlar-type fibers), and rapid oxidation (carbon fibers). Hence, the fibers used in advanced composite structures frequently represent unique materials that are not possible or at least difficult to achieve in bulk. The same explanation goes for the reduction of the strength of the fiber as the diameter increases (as shown in Figure 1.8).



FIGURE 1.8 Effect of fiber diameter on strength [1] (courtesy of ASTM).

#### 3.1.1.3. Flexibility in Forming

The fiber format allows formation of very complex shapes out of strong and stiff materials at very low forces and without breaking the fibers. This is because at these very small diameters, the fibers may conform to complex shapes by essentially elastic bending. For example, the maximum axial strain in a fiber of diameter d bent to a radius  $\rho$  under typical elastic assumptions is:

$$\left|\varepsilon_{\max}\right| = \frac{d}{2\rho} \tag{1.6}$$

Hence, if a fiber of diameter 10  $\mu$ m is bent to a radius of 2.54 mm (0.10 in), the axial strain will be  $1.97 \times 10^{-3}$ , or about an order of magnitude smaller than the typical strain at breaking for a glass fiber (5% or 0.05). Hence very small features may be molded into advanced composite parts without damaging the fibers.

The fiber format provides many advantages as mentioned above. However, the fiber format also presents difficulties and disadvantages that need to be addressed. These are described below.

#### 3.1.2. Disadvantages of the Fiber Form

#### 3.1.2.1. Requirement of a Large Number of Fibers

Fibers have very small diameter (about 10  $\mu$ m, while the diameter of one hair is about 100  $\mu$ m). In order to make something of a good dimension for engineering applications, one needs to make components with thickness on the order of millimeters or centimeters (about 1000 times the diameter of a fiber, and of width in the order of decimeters or meters). Therefore, one needs millions and millions of these fibers to make an engineering component of significant size. Individual fibers by themselves are very flexible and fragile. The fibers tend to curl and form entangled pieces if not aligned. Figure 1.9 shows a photograph of three tows of entangled fibers.

The fibers need to be aligned, and slightly tensioned in order for their properties to be effectively utilized. In order to withstand loads of significant magnitude, millions of fibers need to be aligned and work simultaneously. Not only that, fibers need to be straight and a small amount of tension may be required to keep them straight. Special techniques and care are required to attain this configuration.



*FIGURE 1.9* Photograph of entangled bunches of fibers. Each shows a tow with fibers aligned and fibers that are frayed.

# *3.1.2.2. Fibers Need to be Bonded Together to Provide Good Mechanical Properties*

Fibers used in composite materials can have a significant variation in their strength because strength depends on the microstructure of the material and is very sensitive to the presence of defects. Fibers are brittle materials and their strength exhibits a significant amount of variation. Figure 1.10 shows the variation in the strength of graphite fibers. This figure shows that graphite fibers can have strength that varies from 0.14 MPa–0.4 MPa. The strongest fiber can have strength that is about three times more than that of the weakest fiber. If the fibers are aligned but not bonded together, the strength of the whole bundle of fibers would be governed by the strength of the weakest fibers.

The strength of a dry bundle of fibers can be much less than the average strength of a bundle of fibers. A dry bundle of fibers means that the fibers are not bonded together by the matrix material. The following example illustrates why a dry bundle of fibers has much lower strength than that of the average strength of the fibers and why the use of adhesive bond between the fibers can improve the strength of the bundles.



FIGURE 1.10 Typical strength distribution for graphite fibers.



FIGURE 1.11 A dry bundles of 5 fibers.

#### Example of Domino Effect of Strength of a Dry Bundle Of Fibers

For illustration purposes, a dry bundle of five fibers is shown in Figure 1.11. Assume that this bundle of fibers is held fixed at the top and all five fibers are joined together at the bottom by a common bar. This bar is in turn subjected to a load P. Due to the variation in the properties of the fibers, assume that the strength of the five fibers is as follows:

Load required to break fiber $1 = 0.30$ N	
Load required to break fiber $2 = 0.35$ N	
Load required to break fiber $3 = 0.25$ N	
Load required to break fiber $4 = 0.40$ N	
Load required to break fiber $5 = 0.50$ N	(1.7)

The sum total of the above five loads is 1.80 N. However, if the load *P* were to increase slowly from 0 N, the whole bundle of fibers will break when the load reaches 1.25 N, much less than the total value of 1.80 N. The reason for this is as follows:

- a. When the total load *P* reaches 1.25 N, the load in each of the fibers is 0.25 N (1.25 N/5). This is the load at which fiber 3 breaks.
- b. After fiber 3 breaks, only 4 fibers remain to sustain the load of 1.25 N. The average of 1.25 N over 4 fibers is 0.31 N. This load in turn is more than the breaking load of 0.30 N of fiber 1. So fiber 1 breaks and leaves only 3 fibers to sustain the load of 1.25 N.
- c. The average of 1.25 N over 3 fibers is 0.42 N. This is more than the breaking load of 0.35 N of fiber 2 so this fiber breaks.
- d. The average of 1.25 N over 2 fibers is 0.63 N. This is more than the breaking loads of 0.40 N and 0.50 N of fibers 4 and 5 and therefore these fibers also break.

The domino effect above results in the total bearing load of the bundle being controlled by the strength of the weakest fiber in the bundle. For this situation, the stronger fibers cannot contribute much to the enhancement of the strength of the material.

On the other hand, if the fibers were bonded together by some adhesive matrix such that the load from one fiber can be transferred to another through the adhesive mechanism, the situation is different. To illustrate this point, Figure 1.12 shows again the fiber bundle as in Figure 1.11 but in this case the fibers are bonded together via matrix adhesive.



FIGURE 1.12 Bundles of five fibers bonded together via adhesive.

Assume that we also have the same individual fiber failure loads as in Equation (1.7). Now assume that the load is increased slowly from 0-1.25 N. Assume also that the matrix has negligible (zero) tensile strength (matrix can have good adhesive shear strength but low tensile strength). As such, fiber 3 will break at a location within its length. A small crack is shown in Figure 1.12. For fiber 3, load can no longer be transferred between the two pieces of the fiber above and below the crack. However, load can still be transferred by shear action between fiber 3 and fiber 4 and between fiber 2 and fiber 3. The two segments of fiber 3 above and below the small crack therefore do not become totally useless. In fact they still contribute to the bearing of the load and the remaining structure that supports the load P is more than just the four fibers 1, 2, 4, and 5. The stress in the surrounding fibers may be more than before, as shown in Figure 1.13. In Figure 1.13(a), the adhesive is shown as the binder. The middle fiber has a crack. The shear stress  $\tau$  at the interface between fiber and binder shows a maximum close to the cracked end of the fiber. The normal stress  $\sigma$  is zero at the fiber end and increases as one moves away from the end. In Figure 1.13(b), the presence of the crack in the middle fiber causes the normal stress  $\sigma$  in the fiber on the right to increase a little in the vicinity of the crack. Depending on the strength of the fiber on the right at that location, that fiber may or may not break. If one assumes that the normal stress in the fiber to the right and left of the cracked fiber increases by 8% (0.02 N) due to occurrence of the crack in the middle fiber, the maximum stress in the remaining fibers can be as follows:

Fiber 1: 1.25/5 = 0.25 N Fiber 2: 0.25 + 0.02 = 0.27 N Remaining of Fiber 3: Less than 1.25/5 = 0.25 N (assumed to be 0.21 N) Fiber 4: 0.25 + 0.02 = 0.27 N Fiber 5: 0.25 N

Total sum = 1.25 N

(The lower stress in fiber 3 is due to the presence of crack which relaxes the stress in this fiber and the load is shifted to the other fibers.)

Comparing the strengths of the fibers as shown in Equation (1.7), it can be seen that the crack will not propagate at 1.25 N load. Load needs to be increased if further cracks are to happen. The presence of the adhesive therefore allows the stronger fibers to participate in the load bearing action. This is because after the occurrence of the first crack, fiber 3 does



FIGURE 1.13 Stress redistribution after a fiber cracks.

not become totally useless. It can be seen in Figure 1.13(b) that even though the normal stress  $\sigma$  becomes 0 at the crack, it picks up as one moves away from the location of the crack. This occurs on both portions of fiber 3 above and below the crack. As such the loss of load bearing of fiber 3 is not 100%.

Note that the assumption of 8% increase in the load in surrounding fibers and 16% reduction in the load in the broken fiber are assigned for this illustration only. The real values of the modified stresses (increase or decrease) depend on the particular arrangement of fiber and matrix materials and need more rigorous analysis to be accurate. The adhesive is therefore essential for the strength of the composites. At locations where there is no adhesive (the so called dry spots), cracks may appear and propagate and premature failure may happen. One of the challenges for manufacturing using composites is to assure that the matrix adhesive surrounds each and every one of the fibers (this is the so-called wetting action).

## 3.1.2.3. The Need for a High Fiber Volume Fraction, $v_f$

Since the fibers provide strength and stiffness for the composite materials, it is essential that one has as much fiber as possible in a composite material. The modulus of the composite along the fiber direction is proportional to the fiber volume fraction as expressed in Equation (1.2).

In Figure 1.14, the white dots represent the fiber cross section and the dark area represents the matrix material. The volume fraction of the fibers can be obtained from the micrograph by determining the ratio of area of fiber over area of the material. It can also be calculated based on some idealized arrangement. Figure 1.15 shows a square array of fibers.



FIGURE 1.14 Micrograph of a cross section of a unidirectional composite sample.



FIGURE 1.15 Square array of fibers.

Based on this arrangement, the fiber volume fraction can be calculated as:

$$v_f = \frac{\text{Volume of fiber}}{\text{Total volume}} = \frac{n^2 \pi d^2}{4(nd+n\delta)^2}$$
(1.8)

where *d* is the fiber diameter and  $\delta$  is the shortest space between two fibers. In the limit when the fibers are touching ( $\delta = 0$ ), one has:

$$v_f = \frac{\pi}{4} = 0.785 \tag{1.9}$$

So the limit for fiber volume fraction in the case of square array is 0.785.

One can also assume the fiber arrangement to be hexagonal such as those shown in Figure 1.16. The maximum fiber volume fraction is 0.875 for open packing and 0.907 for closed packing.

Normally the fiber volume fraction may not reach the high levels cal-



*FIGURE 1.16* Hexagonal packing arrangement of fibers: (a) open packing, (b) closed packing.

culated using the ideal arrangement. Fiber volume fractions achieved in practice is around 68% for hand-lay-up using autoclave molding and may be 70% for pultrusion. Note that there should always be a layer of resin in between two fibers, otherwise dry spots will occur and dry spots are points of weakness. Therefore one important thing to remember in the case of composite materials is:

For composite materials, one wants to have as much fiber content as possible as long as the fibers do not touch each other.

This can be put another way as:

For composite materials, one wants to have as little matrix material between two fibers as possible, but not zero.

## 3.1.2.4. Small Interfiber Spacing

One important consequence of the high fiber volume fractions for advanced composites is a small interfiber spacing. For example, if the actual microstructure is approximated as a square array (Figure 1.15) where the maximum allowable fiber volume fraction is  $\pi/4$ , the average interfiber spacing  $\delta$  can be calculated as:

$$\delta = d \left[ \sqrt{\frac{\pi / 4}{v_f}} - 1 \right] \tag{1.10}$$

where d is the fiber diameter.

Hence for a typical case (graphite/epoxy) with  $d = 10 \,\mu\text{m}$  and  $v_f = 0.68$ , one gets  $\delta = 0.74 \,\mu\text{m}$ .

#### Important Consequences of this Small interfiber Spacing

1. *Stress concentration*. In the solid composite, the resin is highly constrained in small volumes between the fibers, which results in stress concentrations and reduced strength in the matrix-dominated directions.

During processing, the small interfiber spacing also has important consequences:

2. *Fiber-to-fiber contact.* Because there is much variation in fiber spacing for real composites, a small average  $\delta$  suggests considerable fiber-to-fiber contact. This can make the fiber bundle

load-bearing when compressed in the transverse direction. This means that when a load is applied on a bundle of fibers containing liquid resin, fibers may support the load through their contacts as shown in Figure 1.17. A direct consequence of this can be reduced resin pressure during cure, which can lead to potential voids in the matrix.

- 3. Large shear resistance of prepregs. Another important effect is the resultant large shear resistance of the prepregs (Prepregs are fibers impregnated with partially cured resin). This affects properties such as drape, which translates into poor handling properties during manufacture. *Drape* is a term used to denote the ability of the fiber fabric to conform to the shape of the tool.
- 4. *Small permeability values.* Permeability is a characteristic of the bed of fibers that indicates the ease (or difficulty) for the resin to penetrate into the bed of fibers. Permeability depends on the space between the fibers and can be shown to scale roughly as  $\delta^2$ . As such, the small interfiber spacings result in very small permeability values. The effects of increasing liquid volume fraction  $v_R$  (or decreasing interfiber spacing and therefore decreasing  $v_f$ ) on the axial permeability S<sub>11</sub> and the transverse load-carrying capacity of the aligned fiber beds are shown in Figure 1.18. It can be seen that the higher is the liquid volume fraction (or the lower is the fiber volume fraction), the higher is the values of the permeability.



*FIGURE 1.17* Contact between fibers allows fibers to partially support the applied load (sharing with the resin).



Liquid volume fraction V<sub>R</sub>

*FIGURE 1.18* Effect of liquid volume fraction on the axial permeability of an aligned fiber bed,  $S_{11}$  (reproduced from Reference [2], with permission from John Wiley and Sons).

The above four factors act to limit the maximum obtainable fiber volume fraction, generally making it much below the theoretical maximum values of 0.785 for square packing, and 0.907 for hexagonal packing.

#### 3.1.2.5. Anisotropic Behavior

Figure 1.19 shows a representative element of an aligned fiber bundle. It exhibits anisotropic behavior (properties depending on direction). Apart from mechanical properties such as stiffness and strength, there are also implications of anisotropic behavior for manufacturing. One particular influence is the anisotropy of the permeability of liquid resin into the interstices between the fibers. For the graphite/epoxy system shown in the figure, the ratio between elastic modulus along the fiber direction over elastic modulus transverse to the fiber direction  $E_{11}/E_{22}$  is about 16. Similarly, the ratio of axial to transverse resin permeabilities



FIGURE 1.19 Representative element for an aligned fiber bundle.

for the fiber bundle,  $S_{11}/S_{22}$ , is on the same order. Figure 1.20 shows the transverse permeability of an aligned fiber bundle, which is about one twentieth the values as those along the axis of the fibers, as shown in Figure 1.18.



**FIGURE 1.20** Transverse permeability of aligned fiber bed. Note that the unit for permeability is multiplied by  $10^{-11}$  cm<sup>2</sup>, as compared to Figure 1.18 where the unit is multiplied by  $10^{-10}$  cm<sup>2</sup>. The ratio between axial permeability and transverse permeability is about 19 (reproduced from Reference [2], with permission from John Wiley and Sons).

# 3.2. Matrix Materials

It was mentioned at the beginning of Section 3.1 that fibers are made of strong and stiff materials and that they can provide strength and stiffness for the composite materials. However fibers by themselves cannot provide these properties to the composites. This is because fibers exist in tiny quantities (the diameter of a fiber is about 7 mm). In order to make composite structures of a dimension of engineering significance, parts of dimension in the order of centimeters (0.01 m) or decimeters (0.1 m)need to be made. Matrix materials serve the function of making this possible. Matrix materials usually have low normal strength (tensile or compressive) but they can provide good adhesive shear strength. The tensile strength of epoxy resin is about 35–130 MPa (as compared to the tensile strength of carbon fiber of about 3000 MPa). The shear strength of epoxy adhesive is about 20 MPa. Even though this number (20 MPa) seems to be small as compared to the tensile strength of carbon fiber (3000 MPa), the aspect ratio of the fibers (length over diameter) is usually large. This provides comparative shear load as compared to tensile load (more analysis to illustrate this effect is given later in this chapter). As such, matrix materials in composites are utilized such that shear is the main mode of loading. The matrix serves the following functions.

### 3.2.1. Aligning the Fibers

It can be seen in Figure 1.9 above that a bundle of dry fibers consists of fibers that can faze. Individual fibers can take random orientation and may not align with each other well. In order to make an engineering component out of composite materials with a certain significant dimension, the fibers need to be aligned. One can grasp the fibers and align them, however in order to keep them aligned, some form of glue (adhesive) needs to be used. The matrix material serves the function of the glue. It is essential that the matrix resin (glue) surrounds the total surface of each individual fiber.

## 3.2.2. Transfer the Load Between the Fibers

In Section 3.1, it was mentioned that fibers need to be bonded together so that their strengths can be utilized effectively. Otherwise the domino effect will take place and the strength of the composite material is governed by the strength of the weakest fibers. The bond is provided by the matrix material, since the matrix material serves as a glue. This bonding action serves to transfer the load from one fiber to the matrix and then



FIGURE 1.21 Shear load transfer between fiber and matrix.

from the matrix to the next fiber. While the fiber supports the load via its tensile strength, the matrix provides the load transfer via shear strength (Figure 1.21).

When a load is imposed on the matrix portion of the material, this load is transferred to the fiber in the form of shear. Usually the shear stress at the interface is maximum at the end of the fiber and minimum toward the center of the fiber [Figure 1.13(a)]. If one assumes that the shear stress is constant to simplify the calculation, and assuming that the diameter of the fiber is *d* and its length is *l*, equilibrium of the broken segment gives:

$$\frac{\pi}{4}d^2\sigma_f = \pi d\frac{l}{2}\tau_i \tag{1.11}$$

yielding

$$\frac{l}{d} = \frac{l}{2} \frac{\sigma_f}{\tau_i} \tag{1.12}$$

Equation (1.11) shows the balance between load provided from tensile resistance of the fiber and load provided from shear resistance at the interface, where  $\sigma_f$  represents the tensile stress in the fiber and  $\tau_i$  represents the shear stress at the interface. Equation (1.12) shows the aspect ratio of

the fiber as a function of the two strengths. If the aspect ratio is as given in Equation (1.12), then failure will occur by both fiber breaking and interface slipping simultaneously. If the aspect ratio is larger than that given in Equation (1.12), then the fiber is longer than the critical length and failure will occur by fiber breaking. If the aspect ratio is less than that given in Equation (1.12), then failure will occur by slipping at the interface. In this case, the fiber is not well utilized. In order to fully utilize the strength of the fiber, it is important that the fiber be longer than the critical length given as:

$$l_c = \frac{1}{2} \frac{\sigma_f}{\tau_i} d \tag{1.13}$$

# 3.2.3. Assisting the Fibers in Providing Compression Strength and Modulus to the Composites

Fibers are long and slender pieces that behave like strings. Individual fibers resist tension well but they cannot resist compression. In order to possess good compressive properties, fibers need to be aligned. Matrix materials assist in aligning the fibers and providing the composite materials with compressive properties.

# 3.2.4. Assisting the Fibers in Providing Shear Strength and Modulus to the Composites

Similar to the above, a bunch of individual fibers cannot provide good shear properties because the fibers can slide relative to each other. The presence of the matrix material provides the shear transfer between the fibers and this also provides good shear properties for the composite material.

#### 3.2.5. Protecting the Fibers from Environmental Attack

Fibers such as carbon and glass usually have high surface energies. Moisture can easily adsorb on the surface of these fibers. With adsorption of water, it is difficult for the matrix material to adhere to the fiber to make a good bond. The presence of the matrix on the surface of the fiber prevents moisture from adhering to the fiber surface. Also water attacks and creates cracks in glass fiber over a long time. The presence of the resin on the surface of the fiber prevents glass fibers from being attacked by moisture in the surrounding environment.

#### 3.3. Interface

It was mentioned in the previous section that the matrix needs to be bonded to the fibers. The bond between the fiber and the matrix constitutes the interface between them. The interface area within a certain volume of a composite material made up of aligned fibers of diameter d can be estimated as:

$$\frac{\text{Interface area}}{\text{Composite volume}} \approx \frac{4v_f}{d}$$
(1.14)

Equation (1.14) is obtained by considering a unit cell containing one single fiber (which can be extracted from Figure 1.15). The interface area is  $\pi d$  while the composite volume is  $(\pi d^2/4)v_f$ .

For a composite 1 m × 1 m × 0.02 m with  $v_f = 1/2$  and  $d = 8 \times 10^{-6}$  m, the interface area is:  $(0.02 \text{ m}^3)(4 \times 0.5)/(8 \times 10^{-6} \text{ m}) = 5000 \text{ m}^2!$  To develop this interface, the resin must come into intimate contact with the fibers. A good interface is needed for a coherent structure that transfers loads around broken fiber ends and carries transverse loads. In general, one does not want the strongest possible bond. Separation between the fiber and the resin can be an important energy absorbing mechanism during the failure of the composite. This idea is used to great advantage, for example, when making composites that stop ballistic projectiles. In order for this interface to develop, there are two requirements that a manufacturing process needs to satisfy: availability and compatibility.

#### 3.3.1. Availability of the Resin at the Surface of the Fibers

For the interface to develop between the resin and the fiber, the resin needs to be available at the surface of the fiber. This may seem obvious, but the concept is important for manufacturing. Recall that the diameter of a fiber is about 10  $\mu$ m. Assuming a fiber volume fraction of 0.6, the interfiber distance can be calculated from Equation (1.10) to be 0.14  $\mu$ m. The dimension of the unit cell would be 10.14  $\mu$ m. If one were to make a laminate for an aircraft wing about 3 mm thick and 500 mm wide, there are about 15 million fibers over the cross section of the part. One needs to get the resin onto the surface of each and every one of these fibers to assure a good quality part.

To bring the matrix to the surface of the fiber, the matrix needs to be in the form of a liquid with low viscosity. The bulk flow is generally pressure driven by an external pressure source, with the final degree of wetting accomplished by capillarity. Often the bulk flow may be complicated by the inhomogeneity of the fiber distribution. For example, in woven and braided fabrics, the different length scales that characterize the fiber structure result in different time scale for resin flow. The basic event of flow of resin through a fiber bundle can be modeled as flow through an anisotropic porous medium. The flow of the liquid through a bed of these fibers normally is estimated using Darcys law of flow through a porous medium as:

$$q_i = \frac{S_{ij}}{\mu} \frac{\partial p}{\partial x_i}$$
(1.15)

Here the vector  $q_i$  is the so-called filter velocity, which consists of the three components of the volumetric flow rate divided by the cross-sectional area. Where  $\mu$  is the Newtonian viscosity and  $\partial p/\partial x_j$  is the pressure gradient vector. The term  $S_{ij}$  constitutes the permeability matrix, with units of (length)<sup>2</sup>.

When Darcys law is written in the material coordinate system, such as the direction shown in Figure 1.19 for a bundle, the permeability matrix is diagonalized and Sij takes the form:

$$S_{if} = \begin{bmatrix} S_{11} & 0 & 0 \\ 0 & S_{22} & 0 \\ 0 & 0 & S_{33} \end{bmatrix}$$
(1.16)

where  $S_{ii}$  are the principal values corresponding to each of the three principal directions. It is usually further assumed that the fiber bundle is transversely isotropic which leads to the simplification that  $S_{22} = S_{33}$ .

For the majority of cases, the flow is along one direction (normal to the plane of the composite component), for this case the equation simplifies to:

$$q_z = \frac{S_T}{\mu} \frac{dp}{dz} \tag{1.17}$$

where z represents the direction along the normal to the plane and  $S_T$  represents the transverse permeability of the fiber bed.

A faster flow rate [large value of  $q_z$  in Equation (1.17)] can bring the liquid resin matrix to the surface of the fiber faster. If the liquid resin has viscosity like water (1 centipoise) then it runs well. If the liquid resin has

viscosity like honey at room temperature ( $10^5$  centipoise), then the flow is not as rapid.

The viscosity of the matrix has important implications on the type of matrix materials that are commonly used. Thermoset resins such as polyesters, vinyl esters and epoxies have reasonably low viscosities at reasonably low temperatures (from room temperature up to about 200°C). Viscosities of polyesters are about 1 Pa-sec (1000 centipoise) at room temperature whereas those of epoxies are about 50 centipoise (0.05 Pa-sec) at 115°C. On the other hand, viscosities of thermoplastic resins such as polyetheretherketone (PEEK) (1000 Pa-sec or 10<sup>6</sup> cP at 400°C) are very high. As result, there are more composite materials made of thermoset matrix than with thermoplastic matrix in spite of the fact that composites with thermoplastic matrix can offer better properties such as fracture toughness and environmental resistance.

#### 3.3.2. Compatibility Between the Fiber and the Matrix Materials

The second important requirement for good bonding is that the matrix and fiber be thermodynamically compatible. The required intimate contact can take place spontaneously by a wetting process. The thermodynamics of wetting can be illustrated by calculating the surface and interface energies for a wetting system as illustrated in Figure 1.22.

Surface energy = 
$$A\gamma_i$$
 (1.18)

where *A* is the area,  $\gamma_j$  is the specific surface energy, and the subscript j = s (solid), *l* (liquid) and *ls* (liquid-solid) interface.



FIGURE 1.22 Illustration of (a) non-wetting and (b) wetting.

The equation of equilibrium for the situation shown in Figure 1.22 is:

$$\gamma_{sv} = \gamma_{lv} + \gamma_{ls} \cos\theta \tag{1.19}$$

where  $\gamma_{sv}$  is the surface tension of the solid surface,  $\gamma_{lv}$  is the surface tension of the liquid surface,  $\gamma_{ls}$  is the surface tension of the liquid/solid interface and  $\theta$  is the contact angle. Situation (a) represents the case where the liquid does not want to spread over the solid surface and situation (b) represents the case where the liquid wants to spread on the solid surface. For the case of complete wetting,  $\theta = 0$  and  $\gamma_{sv} = \gamma_{lv} + \gamma_{ls}$ .

For wetting to occur, one needs to have:

$$\gamma_{sv} \succ \gamma_{lv} \tag{1.20}$$

Simply put, low-energy liquids wet high-energy solids. For many polymer composite systems, the liquid resin will naturally wet the solid fibers. This is because the fibers are high surface energy elements, whereas the liquid polymers have much lower surface energy. Exceptions to this rule can occur when we try to wet solid polymeric polymers with other liquid polymers, or dirty and/or wet solid fibers with liquid polymers. With metal matrix composites, it is difficult for the liquid metal to wet the fibers, since the molten metal may have a higher surface energy than do the solid fibers. The surface energy of metals varies from 400-2000 dyne/cm, that of clean glass is about 500 dyne/cm, graphite fiber is about 50, and that of polymers is about 30-45 dyne/cm. As such, polymeric liquids have lower surface tensions than most fibers, making the polymeric liquids compatible with the fibers. This facilitates bonding between the polymeric liquids and the solid fiber materials. This explains why, in practice, there are more polymer matrix composites than metal matrix composites even though metal matrix composites may provide higher temperature resistance than polymer matrix composites.

In summary, in order for the liquid matrix materials to bond to the solid fibers, two conditions need to be satisfied:

1. The liquid matrix has to be available at the surface of the solid fibers. Various means have been designed to do this and this has resulted in different forms of manufacturing techniques. This is governed by the viscosity of the liquid matrix material  $\mu$ , the permeability of the fiber beds *S* (or *K*), the amount of pressure applied

to move the matrix  $\Delta p$  and the distance that the liquid matrix has to flow  $\Delta x$ . Even though there are many forms of manufacturing techniques, these can all be understood from this simple principle.

2. The liquid matrix and solid fiber materials have to be thermodynamically compatible with each other. To put in simple terms, the surface energy of the liquid matrix has to be less than that of the solid fiber. This principle limits the number of material systems that can be used to make composites and it also explains why there are more polymer matrix composites as compared to metal matrix composites.

Composite materials consist of two groups, based on the length of the fibers. These are short fiber composites and long fiber composites. Short fiber composites consist of those where fibers are about 1.25 cm or less, whereas long fiber composites are those consisting of fibers that are continuous and long. Short fibers are usually incorporated into the polymer system. Traditional processing techniques used for regular polymeric materials such as injection molding or compression molding are also used to process polymers reinforced with short fibers. The principle governing the processing of regular polymers also applies to the processing of polymer reinforced with short fibers. The focus of this book is on long fiber composites.

### 4.0 REFERENCES

- 1. Metcalf A.G. and Schmitz K.G. ASTM Proc. 64, 1974, p. 1075.
- Lam R.C. and Kardos J.L. The permeability and compressibility of aligned and cross-plied carbon fiber beds during processing of composites, *Polymer Engineering and Science*, 1991, Vol. 31, No. 14, pp. 1064–1070.

## 5.0 APPENDIX

#### **Volume Fraction and Weight Fraction**

In the manufacturing of composites, weights and weight fractions are usually used. However, in the determination of different properties for the composite materials in terms of the properties of the fiber and the matrix, volume fraction is usually used. The following gives relations between volume fractions and weight fractions. The following symbols will be used:

 $V_{f} = \text{Volume of fibers}$   $V_{m} = \text{Volume of matrix}$  V = Volume of the composite  $v_{f} = \text{Fiber volume fraction}$   $w_{m} = \text{Matrix volume fraction}$   $w_{m} = \text{Matrix weight fraction}$  W = Weight of the composite  $W_{f} = \text{Weight of fibers}$   $\rho_{f} = \text{density of fibers}$   $\rho_{m} = \text{density of matrix}$   $\rho = \text{density of composite materials}$ 

The volume fraction of fibers can be written as:

$$v_f = \frac{V_f}{V}$$

This can be expanded as:

$$v_f = \frac{\frac{W_f}{\rho_f}}{\frac{W_f}{\rho_f} + \frac{W_m}{\rho_m}} = \frac{\frac{w_f}{\rho_f}}{\frac{w_f}{\rho_f} + \frac{w_m}{\rho_m}}$$

This can be written as:

$$\frac{1}{v_f} = 1 + \frac{\rho_f w_m}{\rho_m w_f} \qquad \text{or} \qquad v_f = \frac{\rho_m w_f}{\rho_m w_f + \rho_f w_m} \tag{1.21}$$

The weight fraction of the fiber can be written as:

$$w_f = \frac{W_f}{W}$$

This can be expanded as follows:

$$w_f = \frac{V_f \rho_f}{V_f \rho_f + V_m \rho_m} \quad \text{and} \quad w_f = \frac{v_f \rho_f}{v_f \rho_f + \rho_m (1 - v_f)} \quad (1.22)$$

#### Percent (%) and Parts per Hundred (phr)

In the discussion for relative amounts of different constituents (particularly for the case of matrix material where curing agents are involved), two types of quantities are used: percent and parts per hundred (phr). The following example illustrates the difference between the two quantities.

A mixture containing 100 g of epoxy and 40 g of amine curing agents is to be made. The percent of amine curing agent in the mixture is 40/140= 29% and the parts of amine per hundred of epoxy is 40 phr.

If 10 g of certain fillers are added into the mixture, the percent of the amine now is 40/150 = 27% while the parts of amine per hundred epoxy remains 40 phr.

The use of phr is convenient in that the phr for a particular constituent does not change when one adds some other components into the mixture.

#### 6.0 HOMEWORK

- 1. Indicate three main functions provided by the matrix in a composite material. Indicate the main functions provided by the fiber in a composite material.
- 2. Indicate the four most common manufacturing processes for making components using advanced thermoset composite materials. For each process, indicate in which sector of the industry the process is used the most. Give a reason why this is so.
- 3. Indicate in what way manufacturing using advanced composites is different from manufacturing using metals.
- 4. Why is wetting of the fibers important? What are the two most important aspects that affect wetting of the fibers?
- 5. Using the square packing array, show that the interfiber spacing is given by:

$$\delta = d_f \left( \sqrt{\frac{\pi}{4v_f}} - 1 \right)$$

where  $\delta$  is the interfiber spacing,  $d_f$  is the fiber diameter, and  $V_f$  is the fiber volume fraction.

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- 6. What is the general difference between the viscosity of a thermoset composite and a thermoplastic composite. What is the implication of this phenomenon on the manufacturing strategy?
- 7. What are the three important advantages of a material (like glass material) existing in fiber form over the same material in bulk form?