

# Introduction

Composite materials are as much an engineering concept as they are a specific material. They represent an idea which to the best of my knowledge was first recognized in engineering in the “composite” construction of the latter Clipper ships in the middle of the nineteenth century where wooden planking was set on iron frames. This was a binary composite—some ships carried in addition copper outer sheathing to combat marine organisms—a tertiary composite. The iron was used because it was strong, stiff, and durable and being cast to shape could make the frames. Iron was not available at all easily in sheet form—wood of course was—as sawn planks. So to use iron’s strength and stiffness a composite was necessary.

The modern term composite arose in the aircraft industry in the middle part of the twentieth century. Its advent can be viewed either as means of using plastics in aircraft construction or as a means of utilizing the stiffness and strength of fibers which were first made in the late 1950s and early 1960s. The first of these views emphasized the desirable properties of plastics which were becoming available in the late 1930s. The modern synthetic plastics were first discovered in the early 1900s. The desirable properties were ease of formability, very low density compared with metals, excellent surface finish, and lack of corrosion. They lacked stiffness and so some stiffening agent was necessary. The leading idea was to use plastics for aircraft construction.

Glass fibers were not available then, being produced in quantity only from about 1940 or so. Naturally grown fibers such as flax or jute were tried as additions to stiffen the plastic. Another possible addition would have been asbestos because chrysotile asbestos is very stiff and strong but it is not available in long continuous lengths. A problem of course with natural fibers, as with wood, which is in fact a natural fiber composite of cellulose fibers in lignin, is that moisture attacks the material and degrades the properties. The addition of flax to an organic resin does make a fibrous composite in the modern sense.

This view of fibrous composites as a means of greatly improving the mechanical properties of plastics remains a very powerful perspective. Almost all plastics in use contain fillers of various kinds, often particulate, which are added to plastic to extenuate the material and hence lower the cost—they are often cheap and plentiful materials such as chalk—and to add stiffness. The addition of fibers is a natural progression.

The normal means of incorporating a filler into plastic involves blending or mixing so as to produce a homogeneous material in which little or no attention is paid to the relative arrangement of the filler particles. While such a procedure will suffice with roughly equiaxed particles of spheroidal or cuboidal form, it will not suffice with fibers if the advantages of incorporating fibers are to be maximized; a fiber is strong and stiff in a direction parallel to its axis, so only in one direction.

The breaking strength and stiffness of a fiber of given chemical constitution can be understood in terms of the geometric form which often governs the strength, molecular construction, and internal structure of the fiber which governs the stiffness. For simple chemical substances such as pure silica ( $\text{SiO}_2$ ) or alumina, a fiber is in principle, and it can be shown in practice, no stronger than a large piece, provided that the large piece has smooth surfaces. The stiffness of the fiber—along its axis—can be understood with some precision—say within 50%—from knowledge of the binding forces between the component atoms. The fiber is strong because the surface is likely to be smooth—in general the thinner the smoother.

For other modern fibers such as those covered in Volume 1 of this work, carbon (graphite), or aramid, control of the internal constitution (the microstructure) is vital for determining the axial stiffness. The strength follows from this and the method of production, drawing through a die at a crucial stage, ensures surface smoothness. To some extent in these cases, provided that the surface is “fairly smooth,” the internal microstructure can also add to or subtract from the strength. For metal wires or fibers the axial stiffness can be understood in terms of the binding forces between atoms. High strength demands very close control of the microstructure, besides attention to surface smoothness and perfection.

These factors controlling the strength and stiffness of fibers were recognized in the 1960s and very stiff and strong fibers of boron and graphite were made for the first time. They were strong and stiff, but also, because they were made from chemical elements of low atomic weight, they are much less dense than most metals.

The second way of looking at composite materials then arises if one asks how to utilize these attractive properties of a fiber: stiffness, strength, and low density. By themselves they can only be used for their mechanical properties in the form of a rope and even for that application some facility for twisting the fibers to form a handleable tow will be necessary. To make them really useful they must be incorporated into some material—a matrix. Clearly one of the most attractive is a resin. They must also be handled before being put into a matrix. Fibers provide the stiffest material when all aligned parallel to one another and this is the form in which they are used in the highest performance applications. Fibers are also useful in woven form and if used thus must be capable of withstanding modern weaving processes.

The outlines and contents of this set of volumes are now clear. They are about the properties of strong stiff fibers and how they are made and how and why the fibers affect the properties of the matrices which are used to bind them together. The fibers require a matrix in order to be useful to the engineer. Since the mechanical properties of the fibrous composite which depend upon properties of the (usually) two components and their relative volume fractions are describable rather generally, so Volume 1 contains both descriptions of the properties of commonly available fibers and consideration of the thermoelastic properties of fibrous composites. Modern methods of describing and characterizing the distribution and orientation of the fibers must also be included and consideration of the nonmechanical properties of composites.

Of course the idea of using a pair of materials each of known properties in order to obtain an effect shown by neither alone is a very general one and one would hope, would occur to any competent engineer. The bimetallic construction devised by Harrison to eliminate the effects of differential thermal expansion in a chronometer is an obvious example. Reinforced concrete is another where steel bars are introduced into concrete in order to bear tensile loads which cement, concrete, or other masonry cannot. Such examples are not referred to as composite materials primarily because the two components are clearly recognizable as distinct from one another and the size of the pieces of each large enough so that one is considering an engineering “structure.”

The principles of interaction of the two material pieces in the above examples are, however, composite principles. The latter example of “reinforced concrete” behaves very similarly to some of the ceramic composites described in Volume 4. A material is usually described as being a composite material when the size of the individual units is so small, usually less than 100  $\mu\text{m}$  (0.1 mm), so they are not visible to the naked eye. Here, importantly, they are so small that a useful material—one handleable by the engineer—contains many millions of units.

The size of the units here are crucial. The thermomechanical properties of the composite described in these volumes—resins and rubbers in Volume 2—metals in Volume 3—ceramics in Volume 4—depend on the microstructure—sometimes called the mesoscopic phase—in the materials on a scale of size between atomic dimensions (i.e., up to a few nanometers at most) and several micrometers (ten thousand nanometers). Over these length scales metals, ceramics, etc. are not homogeneous. Conventional processing (heating, drawing, dissolving, precipitating) of such materials alters this microstructure and redistributes the constitutive chemical elements so as to produce different phases (metals and ceramics) or different conformations (plastics). However, such materials are not to be regarded as composites.

However, when particles, be they fibers, whiskers, voids, or other shapes, of a completely different material are introduced extraneously on a similar scale of size, i.e. much smaller than the diameter of the fibers described here, the resulting material may quite rightly be described as a nanocomposite and the chapter in Volume 1 on Whiskers and Particulates might be described as discussing nanocomposites. The engineering idea of the nanocomposite is of growing importance for two reasons: materials processing capability based on modern microscopes, micromanipulators, and sensitive transducers is now carried out on this scale of size which was not possible a few years ago—nanotechnology—microlithography—and so the principle of combining two (or more) materials in order to obtain new properties shown by neither alone can now be applied on a much smaller scale of size. The nonmechanical, physical properties of such nanocomposites (e.g., magnetic and electrical) I believe to be perhaps more important than the thermomechanical ones. Carbon nanotubes, an important recently discovered form of carbon, are an exemplar.

The upper size limit at which one speaks of a composite material is also important. This arises for two reasons, both ultimately connected with the design of structures. First, as has been pointed out for many years, most living structures of animals and plants are composites in the sense used in these volumes, strong fibers (cellulose in the case of wood, hydroxyapatite in the case of animals’ bony structures) surrounded by a matrix, lignin, or collagen. The matrix fulfils the mechanical properties necessary and in biological structures also permits the passage of fluids and so its transport properties are essential. The strong fibers bear the load. Composite mechanics can explain some of the properties of biological materials and since many biological structures, e.g., birds wings or fishes bodies, are found to be extremely well designed in terms of light weight or aerodynamic efficiency, there is a desire to copy them.

They, like an engineering structure, particularly the civil engineering structure, are grown into place; they develop the composite material as a structure. Conceptually the fibers are placed first in the most economical way (least weight) to carry the load. There are various theorems, Michells' famous one is well known, concerning the arrangement of fibers necessary to bear the loads. The matrix should then be placed around the fibers to fulfill other tasks. In order to proceed like this in more than a rather superficial sense for a structure with varying and dynamic loads and possibly many degrees of freedom, the designer needs efficient computer codes to define and work out the problem—codes which define the response of the structure and of the component materials. These are becoming available—they are mentioned particularly in Volume 6.

Since composite materials can claim to be the first materials to be designed in the sense of building together the two components of a structure and (due to its size) calling it a material, we have thought it timely and correct to review the subject in depth. That is the reason for the appearance of these volumes now. The application of computer codes referred to above require some material properties as input. Sufficient is now known so that one can be quite definitive in reviewing how these may be obtained and used in a large number of cases.

Volume 1 then deals with fibers used in high-performance composites and goes on to cover the general theories of composite properties which apply independently of the precise fiber or matrix used.

Of course the properties of real composites depend on how each is made. Processing is of paramount importance because whether or not a particular structure of a composite with the properties described in say the relevant chapter of Volume 1 or elsewhere can be attained must depend on the processing procedure. If this is complicated or expensive the properties may not be achieved. Usually, but certainly not always, fiber composites are manufactured by making the fibers separately from the matrix and then combining the two, usually with some additive in order to ensure the two come together properly. There are three generic types of matrix: polymer (and rubber), metal, ceramic (and inorganic glass). Ceramics are taken to include composites with a carbon matrix. Composites made with these three matrices, respectively, are described in Volumes 2, 3, and 4 of this comprehensive review. Of the thousands of tons of composite material manufactured today, the great majority have a polymer matrix (Volume 2). The manufacturing techniques used with thermosetting resins are different from those with thermoplastic substances. It follows that a discussion is necessary on the selection of manufacturing routes. The composite fabrication industry is, like other materials manufacturing industries, much concerned with measuring property evolution during manufacture and to control this—the concept of smart or intelligent processing. This relates the material in Volume 2 to that on Smart Composites in Volume 5.

Metal matrix composites (MMCs) are the most recent to be developed—if one omits, as is usually done, to consider cutting tools made of so-called “hard metal” and its derivatives. MMCs have arisen since the 1960s. The reason for the technological interest in MMCs at that time and since arose because the really stiff fibers of low density only became available in the very late 1950s and early 1960s. Boron, discovered in 1958, and graphite fiber, discovered in 1963, were not in fact stiffer than wires of tungsten but the density is almost an order of magnitude less. This predicated the advantage to the aerospace industry which was eager to receive the fibers in order, among other things, to make aluminum—the principal aircraft constructional material—stiffer. Naturally the early composites made with the much stiffer and lighter fiber used a resin as the matrix. However, boron in aluminum was experimented with extensively in the USA and very large pieces were made. When carbon fibers were first discovered they were difficult to make compatible with aluminum due to electrochemical effects and this is still the case to some extent. The advent of silicon carbide fibers in the early 1970s which were quite compatible with aluminum has led to a developing interest in MMCs.

Particles of a very stiff inclusion in a weak elastic matrix if present in sufficient volume fraction can of course give a very stiff composite and so in the case of the metallic matrix particulate composites are produced in larger quantities than those containing fibers.

An additional point is that the metallurgist or materials scientist specializing in metals is very used to dealing with particles inside metals. So particulate composites with a metal matrix seem a natural development. In the case of the metal matrix there is then a case for adding both forms of stiff material to a metal matrix and indeed some composites contain both stiff fibers and stiff particles. The fibers are not usually continuous.

The principal advantages conferred upon a metal by the addition of materials such as silicon carbide, boron, or other strong solid are: increased stiffness, decreased coefficient of thermal expansion, and better resistance to wear. Whether or not the breaking strength of the material is increased is not the most important factor.

The reinforcement of metals though leading to much smaller market sizes than polymer matrix composites has played an important part in the development of the field of composite materials generally. In the years after World War II, although the advantages of a composite material were well known to some

people, metals dominated the construction market, particularly for aerospace and defense which are often the drivers of new developments. Work at NASA initially intended to provide metals of higher temperature performance, e.g., by putting tungsten wires into nickel alloys, led to the understanding of some principles of reinforcement and this understanding was picked up by the metallurgical community.

This community formed the principal group of engineers and scientists concerned with the development of new and improved materials. Experiments in universities and other research institutes led to a wider dissemination of the knowledge of how composite materials could show improved properties compared with metallic monoliths. When the stiffer fibers became available the materials engineering community was therefore much better able to appreciate quickly the possible advantages of the composite approach.

We referred earlier to the fact that making the fibers and matrix separately and then combining them into the composite material is not the only way to fashion a composite. In some alloy systems, e.g. cobalt, chromium alloys containing carbon fibers of a stiff strong phase may be produced *in situ* in a matrix. This is done by various forms of heat treatment and can produce aligned and very regular fibers. Work on such systems, aimed at replacing metal alloy for high temperature service, did not produce commercially useful products but was again of importance in demonstrating the advantages of “thinking composites.”

We now know how to grow *in situ* reinforcements in all three of the matrix types: polymer, metal, and ceramic.

Returning to the point that composite materials are as much an engineering concept as they are a specific material, one can think of introducing voids into a matrix, i.e. using empty space as the second material. Doing so automatically lowers the density and greatly reduces the thermal conductivity while in some cases altering the transmission of sound and the sound velocity. A foam can be a very useful form of material in many cases and metallic foams are dealt with in Volume 3.

The very strong and stiff fibers described in Volume 1 contain principally the chemical elements occurring early in the Periodic table, B, C, N, O, Si, Al—the chemicals which form the basis of the inorganic solids. Of course carbon is always a special case, forming the backbone of the strong organic fibers, polyamide, polyester, aramid, besides providing itself a stiff fiber—maybe the stiffest possible—and being a constituent of some strong inorganics. The six elements just listed comprise indispensable chemical elements in the constitution of carbon, cement, and ceramic matrices. So in this case fibers of very similar chemical composition to that of the matrix are being introduced into it. The reason is that in these cases the introduction of fibers is often, though not always, in order to ameliorate a failing of the matrix rather than to realize a property of the fiber. The principal “failing” of the matrix is of course its brittleness.

The market for composites with a matrix of this type is very much larger than that for metal matrix composites and is of two types. Fibers introduced into cheap widely used materials, e.g., cement or clays (which have a big market) or fibers introduced into specialist materials, such as various ceramics, e.g. silicon nitride ( $\text{Si}_3\text{N}_4$ ).

We should distinguish in passing between reinforced concrete introduced in the early part of the twentieth century where steel wires and or bars are used to enable concrete to bear tensile loads and fiber reinforced cements. In the former case we are dealing with a structure, and the wires may be variously stretched and then anchored in place as in the so-called Freysinnet system. Although some of the theory may be read over and has relevance for composite theory, when prestressing of the fibers, either by accident or by design occurs, reinforced concrete is not a composite material. Asbestos cement and cement containing polypropylene or carbon fibers is. Chapters on reinforced cement appear in Volume 4.

Here the fibers are introduced into the cement, usually Portland cement, in order to prevent it cracking under tensile loads. The use of fibers to prevent otherwise brittle materials from cracking has been recognized for millennia. The Holy Bible (*Exodus*, Chapter 5) refers to the need to introduce straw into clay to make bricks and many early building materials are of this type and are still made. Some of the earliest ancient Egyptian vessels were built from glass fibers wound by hand round a core of shaped clay. Eskimos are familiar with the important fact that ice containing moss is safe to stand on because although it may crack locally under pressure the cracks do not extend as they do in ice containing no vegetation. The effect is quite dramatic and during World War II a large project concerned with the introduction of wood fibers (sawdust) into ice in order to build artificial islands yielded much important data. The theory which explains this type of behavior, namely how it is that fibers prevent the spreading of cracks, is of course given in Volume 4 and is now quite sophisticated and used in the understanding of the composites described in Volume 2 and in the design of personal and of other types of armor.

Ceramics and glasses are materials which do not burn in air at very high temperatures. To prevent oxidation at high temperature one must use either a noble metal or something already oxidized so that the process can go no further. The material must be either a stable oxide or one which is covered with an oxide

(a nitride) or some sort of impervious cover itself stable against oxidation (and nitridation). Many ceramics fulfil these conditions. SiC, for instance, resists oxidation under many conditions due to the formation of a very stable film of silica. Fibers of silicon carbide—almost stoichiometric silicon carbide—were produced due to the efforts of Yajima in Japan in the mid-1970s. At the same time a fiber of aluminum oxide was produced. Such fibers, like carbon and boron fibers, are very stiff and although somewhat denser than the first two are nonetheless much less dense than all metals capable of high-temperature service (with the possible exception of beryllium which is toxic).

At about this time the feasibility of developing materials for service at elevated temperatures under substantial loads in air was assuming great industrial significance. This significance remains today. The advent of the fibers just mentioned, however, gave impetus to the concept of producing a strong composite of very high temperature capability. Ceramics were the natural matrix.

A good deal of research effort has therefore gone into understanding in further detail the mechanics of ceramic matrix composites containing ceramic fibers. A large part of this is the subject matter of Volume 4. Important though it is in advancing understanding, no new material of high-temperature capability superior to that of the nickel alloys—about 1000 °C—has yet emerged. The major reason for this is the lack of a fiber with a high strength at that temperature which can be maintained for many hours.

A feature of many ceramic materials not shared by metals and thermoplastics is that they do not melt, but rather sublime, e.g. C, SiC, and BN. Consequently, it is usually very difficult to form them into solid pieces without voids. Volume 4 therefore considers specifically composites with a matrix containing voids. The difficulty of diffusion in many ceramics and the wish to avoid a matrix full of holes (say by using vapor deposition) means in general that the processing of ceramic composites is very slow compared with others and this adds to the cost of production. Strenuous efforts are being made to change this situation.

Long fiber composites differ from most other materials of engineering construction in being anisotropic and comprising at least two distinct phases or materials. The tensile testing and particularly the compressive testing of very anisotropic materials have demanded the evolution and general acceptance of new test methods for composite materials. The test methods required for a composite usually of plate form and taking into account end effects are reviewed in the first part of Volume 5. In addition standardized test methods concerned with the detailed interaction between fiber and matrix are reviewed.

Composite materials made with a resin matrix are usually susceptible to moisture uptake and this does not occur in metals. Durability tests are therefore of special significance and again Volume 5 deals with these.

The properties of a real material depend on its structure after it has been processed. The properties obtained usually differ from the ideal required. The real properties must be measured without dismantling the piece, therefore, nondestructive evaluation (NDE) is required. The methods used in practice with composite materials are also included in Volume 5. Some of these are similar to those used in evaluating metals—but many are different or so qualitatively different that they require different apparatus. For example, eddy current testing of a metal is commonplace because of the high electrical conductivity. For composite materials based on a resin containing a nonelectrically conducting fiber the technique is of much less importance. This is not because of the low conductivity, because this leads to a larger penetration depth in a composite, but due to the ambiguity of the response.

Of course the development of composite materials as an engineering concept and its realization has advanced at the same time as the development of other engineering materials. Until recently NDE was used almost exclusively for the detection of microscopic flaws (usually cracks) after a material had been created and was in service. Nowadays it embraces all aspects of materials production and application.

One very big concomitant advance contemporaneous with the development of composite materials has been that of optical fiber communication. This highly developed technique evolved very rapidly since the discovery of very low loss optical fibers in the mid-1970s. Optical fibers produced for long distance telecommunication are so important because the methods used to check them and those proposed for this task make the fibers themselves extremely sensitive sensors. Since fiber composites by definition contain fibers, it seems natural to incorporate these into a material so that it might be able to tell one “how it feels.” Optical fiber sensing techniques and the ways of integrating optical fibers into composites (the fibers are usually of much greater diameter than the stiff fibers in the composite) are dealt with in Volume 5.

The use of fibers embedded in a composite so as to monitor structural change can be described as the passive use—producing a perceptive material. A fully intelligent material or “smart material” might try to correct a defect in itself or to alter its form in response to external stimuli. This involves self-measurement of its condition and an active response. Again the bimetallic strip is an extremely simple example. To produce an active response an actuator must be incorporated. The three forms of these piezoelectric, magnetostrictive and those based on what are called shape memory alloys (an imposed shape change is accomplished by means of a crystal transformation which may be reversed) are all described in Volume 5. The piezoelectric actuators of power are often composite materials themselves.

The societal drive to develop modern composite materials arose from a military imperative as we have briefly seen. Hence many of the leading applications of composite materials are in military hardware of some form. The extant and developing applications of composite materials are described in Volume 6. It is clear from the foregoing that component design involving composites will inevitably be *sui generis* due to the anisotropy of the material and to the fact that it contains at least two components. An introductory chapter therefore covers some of the generalities of design.

Composite materials provide stiff, strong, lightweight materials of high corrosion resistance, excellent fatigue resistance, and may be electrically conducting. Provided they can be fashioned to shape at acceptable cost there are many applications emerging. They are gaining ground in civil engineering where the light weight and excellent corrosion resistance makes them the preferred materials for repair of the transportation infrastructure: bridges, roads, tunnels, etc.

Cost is of less importance in areas such as military hardware, highly competitive, high-publicity sports activities, and in medical devices. These are areas where new composite materials have been developed and tried. As happens with all “new” materials, their availability enables the making of some artifacts which would be impossible without them. An example of this type for composite materials would be a man-powered aircraft. More usually the development enables much improved performance of a given artifact so that the performance could not be obtained without use of the material. The increase in performance becomes so great that eventually one considers it natural to use that material. The chassis of Formula 1 racing cars, some aspects of stealth aircraft, and vaulting poles are examples of these for composite materials.

To repeat that composite materials are an engineering concept that is now becoming widely appreciated and will be better appreciated as the result of the publication of these volumes is to recognize that the composite idea, though presently of great importance for constructional materials, has great pregnancy for functional materials, for example in the manufacture of computers, particularly of substrate and packaging materials for silicon chips. Composites provide the possibility of varying combinations of thermal expansion, thermal conductivity, and dielectric constant which are not possible with a single material. Some of this is hinted at in Volume 6 and will develop in the years to come.

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