STRETCHING THE ENDURANCE BOUNDARY OF COMPOSITE MATERIALS: PUSHING THE PERFORMANCE LIMIT OF COMPOSITE STRUCTURES

Very stiff fibres woven into engineering's future: a long-term perspective

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Abstract An account is given of the development of the discipline and practice of composite materials from the 1960s to the present as seen by the author. The present time is so pregnant because composite materials are now displacing metallic materials as the prime material for the construction of the airframes of the new large commercial airliners. Observations are made on the advantages of the Airbus A380, Boeing's 787 and the planned Airbus A350. Some of the difficulties are dealt with and solutions proposed for problems of testing and deterioration. And some final remarks are made on environmental concerns.

Introduction

The best definition I have noticed of a composite material is that contained in the Oxford English Dictionary. It is; *composite: material made of constituents that remain recognisable*. The first use of the term in Engineering refers to the composite construction of the Clipper ships—those wonderful vessels constructed to bring wool from Australia and tea from China to England and there it referred to wooden planking on iron frames. The iron frame was the novel element at the time on the conventional wooden plank. And today, in the marvellous huge wings of the Airbus A380 we have familiar aluminium skins on composite ribs and frames.

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The growth of the business of high-performance composite structures may be linked to the growth of the demand for carbon fibre (Fig. 1). In 2007, carbon fibre production is some 27 thousand tonnes, worth some \$1.3B hence selling at \$50/kg. The global sales of carbon fibre reinforced plastics are some \$10B and Airbus based in Europe and Boeing based in North America absorb some 50% the total small diameter tow of carbon fibre. In view of the importance of carbon fibre I will tell a little of its discovery since I was lucky enough to meet all of the groups responsible and was well acquainted with members of two of these.

History-the property stiffness

In the late 1950s, fibre composite materials were known to many engineers and their advantage in terms of corrosion resistance, light weight and great strength were apparent. Composite aircraft propellers had been made in the early 1920s. Glass-reinforced plastic was common place and in terms of strength per unit weight was superior to aluminium by a large factor (Table 1). Indeed, in unidirectional applications coupled with one-off uses, such as a rocket motor case, it was superb and was used in the Polaris missile programme—showing that a real military interest was aroused.

However, there was lacking a really stiff material, which was weavable and of light weight. At about that time there arose a quickened research interest in composites because of the discovery at Bell Telephone Laboratories of whisker crystals of the lighter materials such as alumina, beryllia and silicon nitride. These, while still denser (density of $3-4 \text{ Mg m}^{-3}$) than aluminium (density 2.6 Mg m⁻³), were very much stiffer with values of Young's modulus of up to

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Table 1 Fibres and composites available prior to 1960

Fibre or composite	Modulus E (GPa)	Tensile strength σ (MPa)	Specific gravity ρ	E/ρ (GPa)	σ/ρ (MPa)
Flax	103	690	1.5	69	460
Hemp-wet	34	-	1.5	23	-
Hemp-dried under tension	85	_	1.5	57	-
Ramie-wet	19	-	1.5	13	-
Ramie-dried	51	758	1.5	34	505
Asbestos-chrysotile	159	1,379	2.6	61	530
E-glass	69	3,447	2.54	27	1,357
Unidirect. Flax phenolic resin (Aerolite)	34	345	1.35	25	256
Unidirect. E-glass/epoxy	41	1,241	2.05	20	605
Asbestos/phenolic partly aligned "Durestos"	17	138	1.27	13	109
Aluminium alloy	70	600	2.8	25	214

Data from: P McMullen, Composites, 15, 222-230 (1984) Fibre-resin composites for aircraft primary structures: a short history

700 GPa—much larger of that of aluminium—70 GPa and hence much stiffer specifically. Asbestos, particularly chrysotile, had been found also to be stiff but toxicity prohibited its use and continues to inhibit it.

In 1958, Talley working at the Texaco laboratories, deposited boron on tungsten wires and produced a fibre of stiffness 400 GPa, wonderfully stiff but too thick to weave. In the early 1960s, Roger Bacon at Union Carbide grew some whisker crystals of graphite. We knew that stiff graphite could be produced by hot working at high temperature and the elastic moduli of graphite crystals were being measured accurately. The importance of the stiffness of the whisker crystals was fervently emphasised by J. E. Gordon who had expert knowledge of composite materials, having built composites of them during World War II. At a meeting of the Royal Society on New Materials in 1963, I asked why there was no stiff fibre of graphite and, as a result of this remark according to Willie Watt, the principal British inventor of carbon fibre, he returned to the Royal Aircraft Establishment (RAE), Farnborough and decided to try and make carbon fibre by carbonising a polymer. It is only fair to record that Phillips, his co-inventor with Johnson, always denied this.

Anyway, they were astute to choose the polymer PAN (polyacrylonitrile $(CH_2CHCN)_n$) to carbonise, which forms a ladder polymer on heating, and clever enough to realise that they should heat PAN fibre in an oven before carbonising at high temperature. And during this stage, while the fibre is being slightly oxidised, preventing the fibres from shrinking by having wound them on a frame. This was and is the most significant step and it is due to British workers at the RAE. Bacon hot stretched a cellulosic fibre and produced a stiff carbon fibre, which he called Thornel. Akio Shindo in Japan had previously made a fibre a good

Fibre or composite	Modulus E (GPa)	Strength σ (MPa)	Specific gravity ρ	Specific stiffness E/p	Specific strength σ/ρ
S-glass fibre	86	4,481	2.49	35	1,780
Unidirect. S-glass/epoxy	52	1,793	2.08	25	862
Boron fibre	379	2,758	2.69	141	1,025
Unidirectional Boron/epoxy	269	1,345	1.97	137	683
Carbon (HM) fibre	379	1,724	2.0	189	862
Unidirectional Carbon/epoxy	131	1,517	1.55	85	979
Kevlar 49 fibre	117	2,758	1.45	81	1,902
Kevlar/epoxy	83	1,931	1.35	61	1,430

Table 2 Some fibres and composites available since 1965

Data from: P McMullen, Composites, 15, 222-230 (1984) Fibre-resin composites for aircraft primary structures: a short history

deal stiffer than normal textile fibre and since the Japanese had heard of the efforts in the UK and USA, he also chose to carbonise PAN. Bacon was unlucky in choosing cellulose. PAN is a much better characterised fibre and produced in quantities. Shindo was lucky in getting the company Toray, a major producer of PAN fibre, to make available the best grade for carbonising. At about the same time, the Rolls-Royce company developed a carbon fibre independently. The book by R. M. Gill—*Carbon Fibres in Composite Materials*—published by The Plastics Institute (1972) gives a good account of these early researches.

As a result of these early works, a stiff fibre of small thickness and of great length could be made; hence, it was weavable. That was effectively the end of whiskers, or was it? Carbon nano-tubes are the present day example. The fact that the PAN-based carbon fibre was weavable was almost as important as its stiffness and its strength. Shortly afterwards, Stephanie Kwolek and colleagues at the DuPont Research Station, Wilmington, Delaware invented Kevlar fibre, also weavable. Some fibres available since 1965 are listed in Table 2. So the fibres were stiff and the resulting composites very stiff but would the composite be tough?

History—the property toughness

At the time, there was distrust in structural engineering circles of the pursuit of very high strength in materials due to the memory of the catastrophic failure of some all-welded ships—the Liberty ships of WW II—and the disaster of the de Haviland's commercial jet aircraft Comet in the early 1950s. In one sense, the mistrust was well placed in view of the fragility under impact conditions of the large fan blades for the RB211 Rolls-Royce jet engine, which were made of carbon fibre reinforced plastic known by the name HyFil[®]. And the notable failure of the Team Phillips round the world carbon fibre composite catamaran in 2003.

The essential "trick" by which a composite of brittle carbon fibres in a brittle epoxy resin provides a tough composite is (as in the case of wood) due to the interaction between the two components. (Cellulose fibres and lignin in the case of wood). They break at quite different strains and then if the matrix fails first it may be traversed by a set of cracks. When the composite finally parts fibres may be pulled from their sockets in the matrix. The first of these processes requires the provision of the surface energy of the cracks; the second requires work to be done in overcoming the sliding friction between fibre and matrix; both together can provide much larger values of energy per unit of weight than is provided by a tough metal such as steel. In some cases, the work of fracture of composite materials can approach the values shown by steel on an energy per unit volume basis. For a detailed discussion of toughness by fibre pull-out, see [1]. The essential difference between the way in which composites requires energy to be broken and the way in which a metal absorbs energy is illustrated by the crushing of tubes, Fig. 2.

So, well-designed fibre composites are both stiff and tough and a conventional compression stress-strain curve of such may provide a form beguilingly similar to that shown by structural steel. The steel achieves its toughness by plastic flow, an internal mechanism, which rearranges the atoms inside the piece and leaves the surface almost (but certainly not entirely) as it was-but on the whole impervious. The stiffness of the material is essentially unchanged. The toughness provided by the composite is wholly different involving cracking and splitting at the interface and of the two components. As the material density decreases with the accumulation of matrix cracks, the surface is likely to become pervious, and the elastic modulus decreases as the amount of "damage" increases. These are clearly undesirable features, certainly in comparison with the provision of the ductility by plastic flow. However, much energy is absorbed. These features are made use of in the design of crash-worthy structures made





of composites, for instance in helicopters, crash barriers and some types of armour.

Strength and stiffness of laminates

The strength and stiffness of carbon fibres are provided of course, in just one direction in space-along the axis of the fibre. The fibres must be linked together by a matrix or used in woven form and this latter usually also requires to be embedded in a matrix—usually a thermosetting polymer-epoxy resin for example. To provide all-round properties parallel sheets of fibre in a matrix are stacked together to form a laminate with the fibres within the individual component laminae running in different directions. The proportion of fibres in the various directions may be altered at will for a particular design requirement. Computer codes are adequate to provide reliable estimates of the elastic and other physical properties in the various directions in-plane and through the thickness (ttt). The laminating process, of course, produces interlaminar stresses within the laminate when it is subject to external force. These must be understood and controlled.

Laminate theory is adequate for predicting properties within the laminate in-plane but is much less adequate near the edges and for *ttt* properties. Here, good predictive schemes are necessary to understand edge effects. Although the elastic properties of a laminate are easily predicted, almost no effort is put into estimating the fracture toughness of a laminate, which has led to a number of spectacular failures in the past as we have noted.

Although the elastic properties are fully appreciated, the process of making laminates, usually in an autoclave, requires much further advances be made. If the cost of moulds were reduced, control of the curing process improved, and observation of the exotherm and other variables improved, a large reduction in the cost of processing would ensue.

By proper choice of lamination stacking architecture, startling and useful effects can be produced such as very large Poisson's ratios, negative Poisson's ratios, and negative thermal expansion coefficient from materials where both fibre and matrix show positive values. Matching of any thermal expansion in a plane may be achieved [2]. These properties have application in switches and thermo mechanical control.

New artefacts

So, because of the high stiffness, low weight and lack of corrosion combined with durability against fatigue, carbon and other fibre composites are becoming common place. Examples of applications are legion: microlight (man powered) aircraft, Formula 1 car chassis, unstayed masts, high-performance racing sails, vaulting poles, squash and tennis racquets, fishing rods, and golf clubs, (each of which has raised the modern standards of the game to new heights), helicopter rotor blades, light-weight construction and repair in civil engineering. There are many others. There is the B-2 bomber.

Large aircraft by Airbus and Boeing

And, of course, in aeroplanes. Military aircraft have incorporated composite components for many years but now civil aircraft do so in increasing quantity. At the time of writing, the proportion of weight in the airframe of large international airliners is about to pass the 50% mark. And this is accompanied by strong competition between designers notably Airbus and Boeing for the best design. Boeing are now making and hopefully successfully flying in 2009 a large civil airliner, with more than 50% of the airplane made of composite. It is interesting that Boeing is aiming to take the lead since Airbus may legitimately claim to have pioneered the use of advanced composite materials with the A300B in 1972 incorporating them in secondary structures such as tail fin leading edges. The A340-600 saw the first use of composites in crucial primary structures such as the rear pressure bulkhead and the keel beam. Other components made from composites on this aircraft include the fin and rudder, horizontal tail plane and wing trailing edge moving surfaces as well as the floor panels in the passenger deck.

Boeing premiered the first 787 on July 8 2007 (cf. US way of setting the date). The airframe materials by weight are 50% composite, 20% aluminium, 15% titanium, 10% steel and 5% other. This might be contrasted with the same break-down for the Airbus A380, the world's largest civil airliner now flying in service since the beginning of 2008, of 22% composite (or 25% if the 3% of Glare is included), 10% steel and titanium and 61% aluminium. In contrast, Boeing's 777 possesses 50% aluminium and only 12% composite. By volume, the 787 will comprise 80% composite. Each 787 contains 35 tonnes of CFRP made from 23 tonnes of carbon fibre. The lighter weight provides greatly reduced fuel burn and a side advantage is that high humidity in the passenger cabin is possible because composites do not corrode like aluminium. Other innovations are an automatic active gust alleviation system developed for the B-2 bomber. Boeing has an agreement with Toray Industries to purchase \$6B worth of carbon fibre.

The most striking innovation is the 787's all-composite fuselage to be made by filament winding of (I believe) four barrel sections, which will be joined end to end to form the fuselage; this will eliminate the use of 50,000 fasteners and will allow a higher cabin pressure during flight compared to that attainable using aluminium. The Airbus riposte is to announce the A350 as a direct competitor (replacing the A330-lite—the initial Airbus response) with an airframe of 52% composite, 20% Al–Li, 14% titanium, 7% steel and 7% miscellaneous. Once again, an all-composite fuselage but with the fuselage made of curved longitudinal sections. A composite fuselage offers the greatest advantage for highly integrated design concepts and greater reduction in production costs. Much potential is seen in thin walled sandwich structures leading to higher bending stiffness than single skin designs.

The A380 is flying well and is successful technically, having a wing span of 80 m and length of 73 m (cf. 60 m and 56 m, respectively, for the 787). As said, composite materials make up 25% of the airframe by weight. CFRP, GFRP and quartz FRP are used extensively in wings, fuselage sections, tail surfaces and doors. It is the first commercial airliner with a central wing box made of CFRP and the first to have a wing cross section smoothly contoured-which allows maximum aerodynamic efficiency-where thermoplastics are used in the leading edges of the slats. Glare (GLAss-REinforced fibre metal laminate) is used in the upper fuselage and in the stabilizers leading edges. Glare has better fatigue, corrosion and impact resistance than conventional aluminium alloys and can be repaired using conventional repair techniques. Laser beam welding is used to eliminate riveting in much of the construction.

Solving problems in composite design

The complexity of failure of large composite structured assemblies such as these represents a major challenge and the problems are too complicated and too interactive to admit of solution by practical testing and in any case the structures are too large. It follows that judicious testing coupled with well-substantiated computer modelling is the only way forward at present. Bird strike, the fatal nemesis of the brave RB211 fan jet project can now be adequately modelled so that the EASA (European Aviation Safety Agency) will accept bird strike simulation. And some forms of virtual testing are being accepted. But the difference from design and testing with metal structures is still vast. Data sheets used for metal structures in the certification process are not suitable for composites because there is too little data; hence no data sheets.

Cooperation in research and development

A feature of modern composite materials engineering with large or very important new structures is the manner in which university groups and research institutes, ex-house to the main contractor, are involved both in designcertainly for the last stages—and in testing activities. As an illustration of this involvement let me mention that the production project for the end section of the A350 involves the following specific requests for work.

On the production method

- Getting it right first time and quality control of the manufacturing process.
- Effects of impact damage—impact due to falling or struck objects (at up to 900 km/h) may not immediately give visual evidence of the event on the exterior surface of the composite—delamination may be produced and/ or peeling at the inner surface. It turns out that curved sections appear to be more susceptible to damage than are planar ones.
- Damage tolerance is a most important concept and much credit will be given to some reliable quantitative statement of how much may be allowed with safety.
- For both of the last two, NDT methods for checking the adhesive bond failure are important. Fokker aircraft developed a method using ultrasound which according to Robert Crane—one of the world authorities in the field—is capable of detecting the dreaded "kissing bond" where no substantial gap appears, yet the laminae are not adhering.
- Modelling of impact damage can save much time and effort and I have already referred to the acceptance of the bird strike simulation.

The Polytechnic University of Madrid has developed a virtual testing procedure for drop weight impact tests using finite element methods (Fig. 3). Recent advances allow the inclusion of complex constitutive equations and their manipulation with fast computers. The main damage mechanisms known, by experiment to occur (these are

intralaminar and interlaminar failure), are explicitly taken into account and used to predict the change in elastic constants and other parameters. The microstructure of the undamaged laminate is included in detail for each ply. Behaviour of the interface elements may be controlled using a simple cohesive crack model. The agreement between simulations and experiments is very good and maximum load at failure and absorbed energy can be accurately predicted. Another example is the development at Cranfield University in England of Z-pinning and of tufting to prevent easy delamination under peel loading.

Most, if not all of these difficulties stem from the fact that how a composite structure, particularly an aligned one, fails in compression, is not understood in sufficient detail. A composite panel may be damaged by impact, as we have seen, and this damage may not be visible. However, it is weakened and the strength in compression reduced. When subject to oscillating stress, as it will be in service, involving tensile and compressive loads the damage may spread leading to fatigue.

Fatigue of composites

In a metal undergoing fatigue, damage takes the form of the initiation and propagation of isolated cracks; the growth of which is governed by the local maximum tensile stress; and the most important of these is at the surface of the component. We know that cracks grow and we can control life by monitoring the growth of the longest crack. The behaviour under multiaxial stresses can be quantified by the use of Goodman diagrams.

These diagrams are not feasible for a composite structure and though it can be said that composite structures are



Fig. 3 Illustrating simulation of impact on a laminated structure-courtesy

more resistant to fatigue than are metal ones, this is only strictly the case in a tensile—tensile situation. In most cases, in contrast to metals, when polymer matrix composites are fatigued, damage takes the form of numerous micro-cracks predominantly in the matrix material or at the fibre-matrix interface and often most importantly by fibre fracture. The damage is sustained and spreads through significant parts of the bulk material and structure. There is no dominant crack and so it is not feasible with *present* knowledge to access the nature of the damage simply by microscopic examination.

To overcome this uncertainty the designer reduces the allowable stress on the material so that it then becomes overweight or overcost and I have personal experience of specific examples of where this occurred. Some suggestions have been made to use the measurement of Poisson's ratio of the fatigued structure as a monitor of the damage. The value of Poisson's ratio is a more sensitive indicator of the presence of cracks than is direct measure of the other elastic constants. It has been argued against this in that the principal Poisson's ratio of a carbon-epoxy aligned composite is very small and hence changes in its value difficult to detect. One answer to this objection is that a composite has a number of Poisson's ratios and so while a very small one may occur between one particular pair of directions, it will be accompanied by a much larger value shown when another pair of directions is taken for the measurements. The latter pair should be chosen. An angle ply laminate has a very large value of the principal Poisson's ratio.

How to assess damage

How new methods of assessment operate may be illustrated by Bunsell's way of controlling the residual life time of CFRP filament wound composite pressure vessels (cf. the 787 fuselage). With metallic structures the conventional test would be to subject the vessel to a pressure of 1.5 times the design pressure but this cannot be used with a composite structure as it would weaken the material.

In the case of filament wound pressure vessels damage occurs not by macroscopic crack propagation but by fibre failure distributed more or less at random throughout the composite. As carbon fibre dominates the behaviour, it could be supposed that if the fibres survive the initial loading they will prevent any further damage. However, like unidirectional specimens loaded parallel to the fibres the composite continues to accumulate fibre failure after loading to a constant level. This can be detected by acoustic emission. A finite element model based on first principles which takes into account load transfer between single broken fibres, and their neighbours can predict the tensile failure well. The model is used to calculate a master curve, which corresponds to a pressure vessel having exactly the desired service life.

A threshold damage level is determined experimentally. The damage accumulation shown in the master curve reaches this threshold level after the required service life is attained. Subsequently tests on a vessel in service are carried out periodically so as to predict residual life times. As it has been shown that the rate of damage is only a function of applied stress and the number of damage sites a period of steady pressure will give a rate of damage which describes the state of accumulated damage in the pressure vessel. The test involves monitoring of damage events by ultrasound over a period of several hours and comparison with the number predicted by the master curve. Bunsell has obtained results after more than 2 years in service and the results show that the pressure vessels will survive for more than 20 years of service.

It will be apparent from these examples how research and contract groups and university departments could become involved.

Omissions

I have described composites from the point of view of a material scientist/structural engineer; hence, production methods are not dealt with. There are many additional aspects of composites not mentioned in this review. One is the idea of a smart material, where the material itself becomes an actuator. And related to this, the use of embedded fibres within a composite so as to provide a form of structural health monitoring. In the longer term this may well be used to overcome some of the difficulties in designing against damage sustained in service of the type described in the section above-solving problems. Neither have I mentioned the importance of composites with a metallic matrix, where particles rather than fibres are used-my own special interest at present. A very significant omission is the use of composites for the repair of civil engineering structures, particularly tunnels, sites with limited access. Here it is the essential light weight of composite panels compared with the (usually) steel alternative that enables the composite to displace steel despite the composite's much higher cost. This is because panels may be fitted without the use of cranes and other heavy moving equipment.

Environmental concerns and sustainable development

To some extent such problems are not so much a problem for composites as a problem for the component materials. Part integration and the elimination of fasteners—one of the driving forces for the use of composites (cf. the examples of the reduction of rivets in the A380 and that of fasteners in the 787)—mitigate against re-cycling because of the great difficulty of dismantling. This problem is being addressed by manufacturers of composite aircraft through the formation in 2006 of the Aircraft Fleet recycling Association aiming to deal with the disposal of large composite structures.

A fibre form of any material, where this is made by a non-biological route, always produces a more expensive material than the same material produced either in bulk or in sheet form. And usually requires more energy. Carbon fibre is extremely energy-expensive. Composites may be made from renewable fibrous forms and many essays in this direction are published. The main problem is lack of uniformity in a biological product. This, of course, harks back to the use of some of the fibres listed in Table 1. Efforts will continue to be made to produce environmentally friendly resins and those not based on oil.

However, the overwhelming contribution from composite structures is enormously positive by providing light weight, non-corrodable structures for moving parts and hence greatly reducing the energy cost of moving anything. Ceramic matrix composites may greatly increase the efficiency of power generation.

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