Micromechanical modelling of polymeric composites*

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The community of scientists and engineers associated with composite materials has reached a level of understanding of composite material systems such that we can now consider how composite material systems *should* be made, not just how they *can* be made. In particular, we can now address this question not only for quasi-static stiffness characteristics, but also in order to achieve long-term performance characteristics such as desired remaining strength and life, resistance to environment, and reliability and safety. However, this opportunity presents challenges that are substantial and complex. The present paper addresses an approach to the problem of combining our understanding, and our fundamental science and technology, in such a way that we can make such global predictions with reasonable accuracy for engineering utility. The approach proposed is the use of micromechanical models that represent the strength of composite material systems in terms of the parameters that control the manner in which the systems are synthesized and manufactured.

(Keywords: composites; micromechanical modelling; performance)

INTRODUCTION

It has long been known that the manner in which composite material systems 'come apart' is determined by the manner in which they are 'put together'[1–4]. It would appear that this obvious truism points a simple path to the solution of the problem of attempting to predict long-term behaviour of composite material systems. However, the simplicity of this idea belies the difficulty of the actual task. The anisotropy and inhomogeneity of composite material systems give rise to many complex effects under long-term exposure to mechanical, thermal and chemical applied conditions. The properties and performance of such systems generally change in various modes and manners as a function of time and, in some cases, as a function of rate. These changes occur as a result of 'processes' which may be complex from the standpoint of mechanics. Although documentation of the micro-events that constitute these processes is extensive and even profuse, the interpretation of those observations and the modelling of the fundamental aspects of the processes are difficult. One must decide on the scale of observation and representation that is appropriate; information obtained from non-destructive and destructive tests must be related to mechanics analysis, and results must be related to the remaining strength and life in a way that correctly represents the changes in properties and performance that may be occurring continuously.

The present paper develops the thesis that an approach based on representations of the 'principal strengths' of micromechanical composite systems (which are reinforced by continuous fibres in the present case) can be used as a mechanism to approach this difficult question. The approach to be discussed is an attempt to construct such micromechanical representations in terms of the properties, geometries and arrangements of the constituent material in such a way as to represent changes in material state, and the influence of those changes on changes in local stress state. It should be noted that this approach is separate and distinct from the 'classic' approach of developing micromechanical representations of effective elastic (quasi-static) properties as reported by numerous authors over the years[5,6]. The first major difference is that we are attempting to address the question of constructing a model which anticipates the *strength* of a composite material system, a property which is dramatically different and inherently more difficult to understand than the stiffness of a composite material system (which is relatively insensitive to the microdetails of the constituents). Secondly, we are attempting to discuss the long-term behaviour of composite material systems in which the global properties of the composites are not constant, and the properties and arrangement of the constituents may be changing as a function of the applied conditions, especially mechanical loads and aggressive environments.

Fracture in polymeric composite material systems is usually not a single event, but is a sequence of events, i.e. a 'process'. This failure process generally affects both the state of stress and the state of material in some sequence, until a critical condition is reached. Micromechanical modelling of the strength of polymeric composites must address both of these changes together. Moreover, the physical nature of the processes which bring about those changes must be carefully determined as a basis for setting correct boundary value problems, at a correct dimensional scale.

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In our brief discussion, we present a few examples of micromechanical models of (remaining) strength in the presence of the long-term application of mechanical loading, both quasi-static and cyclic. We then present a very brief discussion of the manner in which those micromechanical models can be used to address the opportunity that motivates our effort, that is, the opportunity to provide assistance to the 'designers' of composite material systems in determining how those systems should be made to achieve specific long-term performance. It is passively assumed that the synthesis and material science community is able to provide answers to the question of how such systems *can* be made; we address the responsibility of the engineering community only, which is to indicate how those systems *should* be made.

We discuss just two examples of what we shall call 'principal strengths' of a polymeric composite material system. These principal material strengths, which are engineering definitions, are defined by our ability to measure those quantities uniquely. Generally, these strengths are defined in 'material directions', in terms of a tensile and compressive strength under unidirectional loading in the direction of reinforcement (X_t and X_c, respectively), tensile and compressive strength perpendicular to the fibre direction under unidirectional loading (Y_t and Y_c, respectively) and the shear strength, S (for in-plane loading). These principal material strengths are a unique set of independent constants when considering the strength of unidirectional materials in response to loading applied in a two-dimensional plane, a situation that is common to many engineering applications. We could also attempt to address questions of strength for three-dimensional situations, an even more complex question. The two examples considered are the tensile strength in the direction of the fibres, and the compressive strength in the direction of the fibres under uniaxial loading.

DISCUSSION

We begin with consideration of the tensile strength of a unidirectional laminate in the direction of the reinforcing fibres subjected to uniaxial mechanical loading. In polymer composites, such strength is generally fibre-controlled, with important contributions of the matrix to the sequence of fibre fracture, and the local stress concentrations in the region of fibre fractures (which control the tendency for subsequent fibres to break). The tensile properties of the fibres, the tensile and shear properties of the matrix, and the fibre–matrix interface or interphase region, as well as the geometry and arrangement of the fibres, matrix and interphase regions, all influence potential tensile strength significantly. The fundamental principles that govern the process of fibre fracture that eventually results in global fracture under tensile loading in the fibre direction are as follows. The statistical distribution of the strength of the fibres determines the manner in which fibre fractures accumulate as a function of the level of applied load. If the statistical distribution of strength of the fibres is great, fibres begin to break at very low stress levels. Second, stress is transferred back into the fibres that break by the shearing action of the matrix around those broken fibres. If the matrix (in the surrounding region) is stiff, this transfer is very quick and the region of disturbance around a broken fibre (called the 'ineffective length') is small; broken fibres which are located near one another do not tend to interact to produce global failure, i.e. the composite material has a high 'bundle strength'. However, in that situation, the local stress concentration in the region in which the build-up happens is very high, since the matrix must transfer stress back into the broken fibre quickly, and the local region around that break experiences a high stress concentration. This last factor counteracts the bundle strength tendency and would suggest that the matrix material (in the composite itself) should be *less* stiff so that the local stress concentration is smaller. Hence, these tendencies must be balanced by a correct representation of the local mechanics to estimate the correct stiffness (and strength) of the surrounding matrix material and, if it dominates, the stiffness and strength of the interphase region that controls the fibre–matrix interaction. Indeed, our understanding of this tensile fracture event presents us with the opportunity to determine the correct answer to the question 'how stiff and strong should the matrix and interphase region be to optimize the tensile strength of the fibre-reinforced composite material system?' That is, we can, if we construct a correct mechanical model, correctly predict how a strong composite material *should* be made.

To this end, Gao and Reifsnider[7] have constructed a micromechanical model of tensile strength in such composite material systems. The model is based on an axisymmetric representation of a core of broken fibres surrounded by matrix material, a ring of next-nearest fibres, a second matrix region, and the surrounding body of composite material as shown in *Figure 1*. *Figure 1* shows that there is also a region of fibre–matrix slip which may be associated with matrix plasticity, fibre–matrix debonding, or the slipping of an interphase region between the fibre and the matrix. This region of slip is joined to the surrounding elastic region by continuity conditions between the two-dimensional solutions for the stresses in each of those regions. The statistical nature of fibre fracture within the broken bundle of fibres is controlled by a model developed by Batdorf and modified by the author to account for non-linear material behaviour, and the non-linearity of the problem is handled by incremental loading and the interaction of solutions[7,8]. The model calculates tensile strength under an increasing load by estimating the first fibre fracture, calculating the local stresses and allowing for matrix or interphase deformation, adjusting the size of the slip

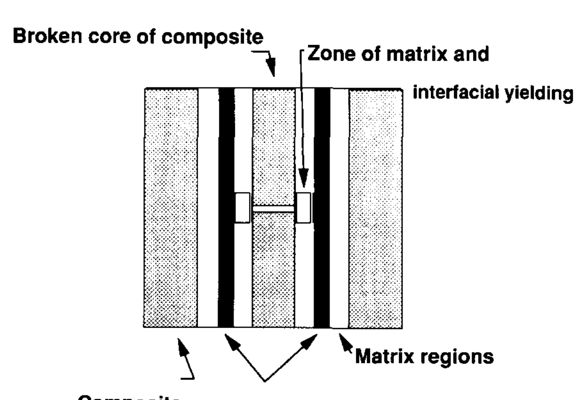

Figure 1 Schematic showing microfeatures of a composite that enter the strength model developed by Gao and Reifsnider

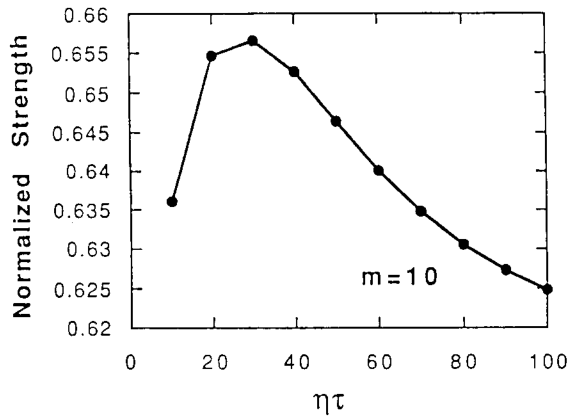

Figure 2 Predicted variation of composite strength *versus* matrix (or interphase) shear strength in a fibrous composite

X_t *Parameters:*	*Affects:*
$E_{ij}^{f,m,i}$	**stress distributions**
r_f, v_f	**stress state**
τ_m	**length of local plastic zone**
β, α	**statistics of fiber strength**

Figure 3 Material and microgeometry parameters that enter the fibre direction tension strength model

region until local equilibrium is achieved, determining if the next-nearest fibre will break in the presence of that local stress, fracturing those fibres if that stress exceeds their strength, and continuing with the solution of the matched boundary value problems at the local level until statistical failure of the fibres dictates global instability and defines global strength.

The model is a representation of the balance between competing effects of the 'ineffective length', as discussed earlier. This model has been validated by comparison with numerous datasets[7,8]. *Figure 2* shows the prediction of composite strength as a function of the plastic flow stress of the matrix or of the interphase region between the matrix and the fibres in a graphite epoxy composite system. It is seen that the model predicts an 'optimum' value for the matrix or interphase strength, that is, the model predicts a correct balance between bundle strength concepts and the local stress concentration limitations discussed earlier. We have shown that such an optimum is consistent with the observation of experimental data, but have been unable to find data that can be directly compared with our observations[7-9].

The parameters that appear in the tensile model are shown in *Figure 3*. (The non-linear model is beyond the scope of our present presentation.) As one might expect, the elastic moduli of the constituents (fibre, matrix, interphase region) play a major role in determining the local stress distributions in the region of the fibres that are broken, as well as determining the manner in which the load is shared by the various constituent materials. Stiff constituents bear a greater local portion of the load than less stiff ones. In addition, it is not surprising that the volume fraction and the radius of the fibres play an important part in the tensile strength since the materials we are considering are fibre-dominated. In addition, one sees that the shape factor of the Weibull distribution of strength of the fibres (which is proportional to the spread of strength of the fibres) and the location parameter, β, which is proportional to the average strength of the fibres, play a prominent role in the global tensile strength model. However, *Figure 3* also shows that the shear strength of either the matrix material around the broken fibres or of the interphase region between the fibres and the matrix also enters the model. That is a surprisingly large influence on the global tensile strength and can be 'optimized' to adjust the balance between the tendency for fibre fractures to join together and lower the bundle strengths and the tendency for broken fibres to break neighbouring fibres due to the stress concentration effect.

In addition to these quasi-static considerations, it is also possible to ask the question, 'how does the global strength change during the application of various long-term environments?' In fact, upon examination of *Figure 3*, one can clearly see that the long-term strength can be discussed in terms of changes in the constituent properties and their arrangement, as represented by variations in the parameters that enter the model, shown in *Figure 3*. Hence, if we are successful in representing the tensile strength (or any other principal material strength) in terms of parameters that are related to how the material is made, we can suggest how a composite material should be made to sustain any given long-term applied conditions on the basis of our observations of the influence of those service conditions on the parameters in our model. Hence, the micromechanical approach is the key not only to the prediction of quasi-static and long-term properties, but also to the challenge of attempting to suggest a proper design for composite material systems for both short-term and long-term performance.

Modelling of compressive strength is even more difficult than for tensile strength in composite material systems. There are at least three distinct failure modes that are commonly observed in various types and forms of polymer composite materials[10-16]. The simplest failure mode consists of 'crushing' of the fibres and the matrix to create a simple failure in compression in the manner that one might expect of any brittle material such as concrete, i.e. a crushing-shear failure at angles of roughly 45° to the load axis in which each constituent material contributes to the strength of the composite in rough proportion to the volume fraction of their presence. This 'rule of mixtures' strength is a common engineering model that is widely used for the approximation of composite behaviour. However, it neglects any 'mutual influence' involved in the action of the fibres and the matrix, and especially any influence of the interphase region between the fibres and matrix materials. In many situations, these interactions, sometimes called 'the composite effect', control and dominate the compressive strength of the system.

Under those conditions, two fibre-dominated compressive failure modes are widely discussed: one in which microbuckling of the fibres controls the global strength, and one in which 'kinking' controls the global strength. In the case of microbuckling, the strength of the composite material is controlled by the stability of the fibres, and the surrounding matrix and composite materials provide a 'foundation' which supports the fibres to inhibit their tendency to buckle. Hence, the matrix material should be stiff so that the fibres are prevented from buckling. However, micromechanical buckling models involve a variety of parameters; indeed, a total

X_c **Parameters:**	**Affects:**
$E_{ij}^{f,m,i}$	stress distributions constraint to buckling
r_f, V_f	stress state; resistance to buckling
$\tau_{m,i}$	resistance to fiber/matrix slipping
η	bonding / debonding

Figure 4 Material and microgeometry parameters that enter the fibre direction compression strength model

of 10 parameters commonly appear in such models in the literature[10–16]. *Figure 4* illustrates a typical set of parameters for a micromechanical model developed by Xu and Reifsnider[13]. The expression they obtain for the prediction has the following form:

$$\sigma_{\mathrm{comp}} = G_{\mathrm{m}}\left[V_{\mathrm{f}} + \frac{E_{\mathrm{m}}}{E_{\mathrm{f}}}(1 - V_{\mathrm{f}})\right]\left\{2(1 + \nu_{\mathrm{m}}) \times \sqrt{\frac{\pi\sqrt{\pi}\eta r_{\mathrm{f}}}{3\frac{E_{\mathrm{m}}}{E_{\mathrm{f}}}\left(V_{\mathrm{f}}\frac{E_{\mathrm{m}}}{E_{\mathrm{f}}} + 1 - V_{\mathrm{f}}\right)[1 + V_{\mathrm{f}}\nu_{\mathrm{f}} + \nu_{\mathrm{m}}(1 - V_{\mathrm{f}})]}} + B\left(1 - \xi - \frac{\sin\pi\xi}{2\pi}\right)\right\} \qquad (1)$$

where $\xi = 2s/L$ (percentage of matrix slippage)[6]. From the expression, it is seen that for a given composite system, the possible factors affecting the composite compressive strength are fibre radius r_{f}, fibre volume fraction V_{f}, Young's moduli of constituents E_{m} and E_{f}, Poisson's ratios of constituents ν_{m} and ν_{f}, fibre debonding η (open-mode debonding) and matrix slippage ξ (slide-mode debonding). The last two factors are directly related to the interfacial strength of the composite.

As shown in *Figure 4*, the elastic properties of the fibre, matrix and interphase region enter this strength model as well. The influence of the geometry of the fibre is much greater in the case of compressive strength since the fibre acts as a 'beam' or 'column'; therefore, the moment of inertia of the fibre is of critical importance. The volume fraction of the fibres is also important to the compressive strength. However, in *Figure 4* we also see that the resistance to fibre–matrix slipping and the length over which any debonding or slipping occurs also enter the model in a prominent manner. In fact, if the fibre and matrix slip, the model shows that a loss of strength of the order of 40 or 50% for just a small slip distance is typical[13]. This large influence of fibre–matrix interaction is critical to a proper representation of the compressive strength and to a proper design of material systems for optimum compressive strength. Hence, this is another example of the manner in which a mechanics representation can provide valuable information regarding how a composite material system should be designed. As in the case of tensile strength, one can see from *Figure 4* that it is also possible to follow the change in global compressive strength by following the changes in the parameters which appear in the compression model as a function of the applied conditions and their history of application.

BRINGING THE MODEL TOGETHER

We will briefly describe the manner in which such micromechanical models can be used to anticipate the long-term performance of polymer composites. We assume that the events associated with the properties and performance of the composites and their long-term conditions are uniformly distributed such that we can define a 'representative' volume which has 'average' properties of the composite material (*Figure 5*). We also assume that the state of material and state of stress in the representative volume are typical of all other such volumes in the material. We introduce a unique concept at this point. We postulate that the representative volume can be divided into a 'critical element' that remains intact and contiguous until the global failure occurs. That is, failure of the critical element defines failure of the representative volume and global failure of the component. The remainder of the material in the representative volume is said to be a 'subcritical element' (or elements) in the sense that their failure does not cause failure at the global level. However, failure of the subcritical elements (which may involve cracking, delamination, debonding or other micro-events) will generally cause changes in the local stress state which controls the conditions in which the critical element must operate. We choose to describe changes in the 'material state' in the critical element using continuum constitutive relationships, and we describe stress state changes at the local level associated with damage in subcritical elements using micromechanical treatments in the classic sense. It should be noted that the scale of the critical element may be very small, as in the case of fibre-controlled tensile failure discussed above, or it may be very large as in the case of global buckling controlled by local ligaments of the laminate which may be millimetres in thickness. Careful experimental observation is essential to determine the scale of details of the critical element which must be used to set a proper boundary value problem for our micromechanical representation of global strength.

This concept of a critical element is associated with specific and distinct failure modes, and is the most important step in our effort to interpret information obtained from laboratory experiments. It is important to note that we have taken the position that it is not possible to construct micromechanical models of strength which are general; we construct micromechanical models for each independent and distinct failure mode. Luckily, the

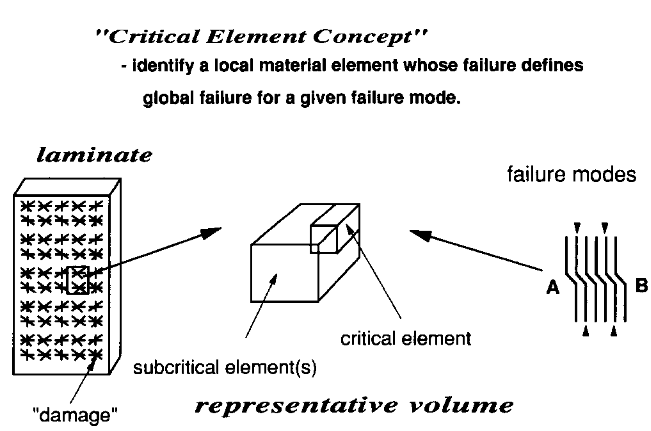

Figure 5 Schematic of the 'critical element concept' for the representation of distributed damage in composite systems

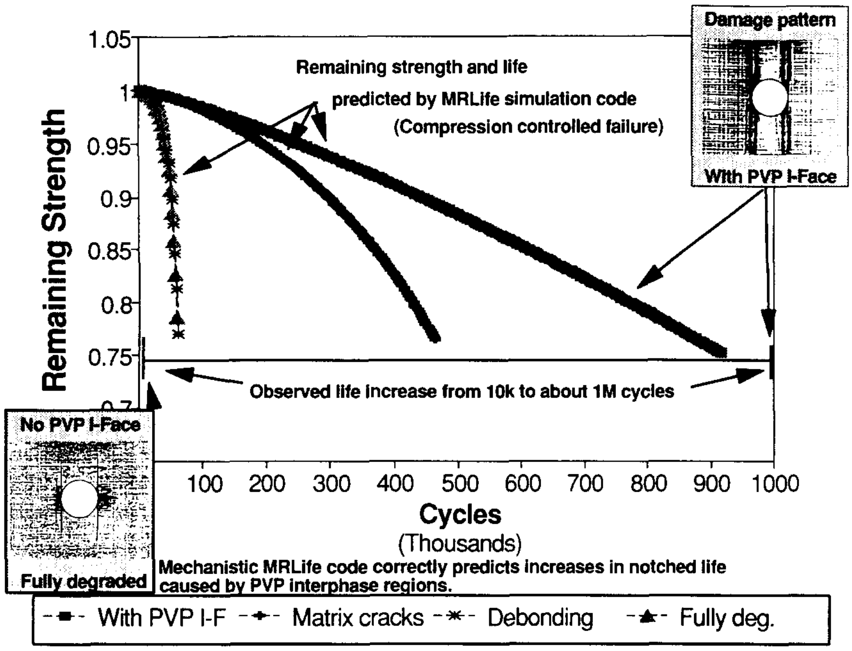

Figure 6 Predicted and observed variations of the fatigue behaviour of graphite epoxy coupons with and without PVP fibre coatings

number of such distinct and unique failure modes in fibre-reinforced polymer composite materials is relatively small[17–20]. For each individual failure mode of interest, we must solve an individual and distinct mechanics problem to interpret our understandings and our data.

The mathematical mechanism used to achieve that interpretation is a 'strength evolution equation' of the following form:

$$\text{Remaining strength} = 1 - \int \left\{1 - \frac{S_a(n)}{S_u[X(n)]}\right\} i \left(\frac{n}{N}\right)^{i-1} \times \mathrm{d}\left(\frac{n}{N}\right) \qquad (2)$$

This expression represents the local state of stress, S_a, in terms of its arguments, which are the values of local stress in the critical element as a function of cycles, n; of course, the local stress in the critical element may be changing as damage develops in the subcritical elements around it. The function S_u in equation (2) represents the local state of strength in the critical element, written in terms of the principal material strength, X_{ij}, as modelled earlier in our discussion. These principal material strengths are also a function of generalized time since the strength in the critical element may be degraded by various processes such as oxidation, chemical degradation or other damage events. The rates associated with the processes that control long-term performance in a given failure mode enter the generalized 'life' of the critical element under current conditions and are represented in equation (2) by N. The parameter i is generally regarded as a material parameter, which is adjustable depending upon the rate of degradation of the material. The equation calculates the normalized remaining strength, S_r, at some instance of time or cycles by evaluating the integral which subtracts the degradation of strength associated with the combined effects of mechanical, thermal and chemical involvements up to that time. It is clear to see that the principal material strengths that we modelled earlier will cause a degradation of global strengths if they are diminished in the denominator of the arguments of equation (2).

Figure 6 shows an example of how these computations can be used to predict the remaining strength of a notched graphite epoxy coupon which is subjected to a fully reversed tension–compression cyclic (fatigue) loading. The figure shows X-ray radiographs of two such specimens after they have been cyclically loaded to a point which is close to their total life time. In the lower left-hand side of the figure, the radiograph shows a damaged specimen (the load axis is vertical) in which fibre fracture has developed near the notch in directions perpendicular to the load axis along the cross-section of the specimen that is sustaining the maximum stress concentrations. In contrast, at the top right-hand side of the figure, one sees a radiograph of a specimen in which damage development in the transverse direction is significantly less, and damage development along lines tangent to the centre hole and parallel to the load axis dominates the observed damage patterns. Physically, the only difference between these two specimens was the introduction of poly(vinyl pyrolidone) (PVP) as a sizing on the fibres. That coating of the fibres, which accounts for less than 1% by volume of the constituents, caused a dramatic change in the damage and failure mode of the coupons under cyclic loading, and that change in damage and failure mode caused a change in total life from about 10000 to over 900000 cycles. This change in behaviour was modelled using the compression micromechanics model discussed earlier, in connection with the evolution equation presented in equation (2). The 'critical element' in this case was a local region of fibres in which microbuckling was modelled, and the representative volume of material which controls final behaviour was chosen as a small micro-element of material along a line parallel to the load axis next to the notch in the cross-section of the specimen. For the material in which the fibres were not sized, damage development, as shown in the bottom left of *Figure 6*, was modelled with matrix cracking, debonding of the fibres and matrix, and subsequent microbuckling of the fibres. As seen from *Figure 6*, the predicted life was less than 50 000 cycles compared to an observed life of the order of 10 000 cycles. For the case in which the fibres were sized with PVP, damage was modelled by allowing the shear damage along tangent lines to relax the stress concentration in the specimen, and by oppressing the matrix cracking in fibre–matrix debonding in the representative volume along the line perpendicular to the load axis through the centre of the notch. In that case, the model predicted an increase in the life of the specimen of nearly two orders of magnitude, which corresponds well to the variation observed in the laboratory. It should be mentioned that the model also predicted a compression failure rather than a tension failure, which is also in good agreement with our laboratory observations.

SUMMARY

We have suggested an approach to the solution of the question of how polymeric composite material systems should be made in order to achieve optimum long-term performance, especially damage tolerance and durability. Our approach is based on attempts to use micromechanics to represent principal material strengths in terms of constituent properties, geometries and arrangements, using parameters which are directly associated with the manner in which the constituents are synthesized and in which the composite is manufactured. This approach has the advantage of providing a clear opportunity to 'design' a material system for specific performance under specific applied conditions. Moreover, it provides the opportunity to apply our understandings and information from the laboratory regarding the constituent materials and their behaviour under long-term conditions to achieve an understanding and prediction of long-term composite behaviour.

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REFERENCES

1 Reifsnider, K. L. (Ed.) 'Damage in Composite Materials', ASTM STP 775, American Society for Testing and Materials, Philadelphia, 1982
2 Reifsnider, K. L. (Ed.) 'Fatigue of Composite Materials', Elsevier, London, 1991
3 Aboudi, J. *Composite Sci. Technol.* 1988, **33**, 79
4 Hashin, Z. and Rotem, A. *Mater. Sci. Eng.* 1978, **34**, 147
5 Halpin, J. C. and Tsai, S. W. 'Effects of Environmental Factors on Composite Materials', AFML-TR.67-423, Wright Patterson AFB, 1969
6 Christensen, R. 'Mechanics of Composite Materials', John Wiley, New York, 1979
7 Gao, Z. and Reifsnider, K. L. *J. Composites Technol. Res.* 1992, **14**(4), 201
8 Reifsnider, K. L. and Gao, Z. in 'Proceedings of the International Conference on Spacecraft Structures and Mechanical Testing', Noordwijk, The Netherlands, 24–26 April 1991, European Space Agency SP-321, 1991, pp. 653–658
9 Gao, Z., Reifsnider, K. L. and Carman, G. *J. Composite Mater.* 1992, **26**(11), 1992
10 Budiansky, B. and Fleck, N. A. *J. Mech. Phys. Solids* 1993, **41**(1), 183
11 Slaughter, W. S., Fleck, N. A. and Budiansky, B. *J. Eng. Mater. Technol.* in press
12 Argon, A. S. 'Treatise of Materials Science and Technology', Vol. 1, Academic Press, New York, 1972
13 Xu, Y. L. and Reifsnider, K. L. *J. Composite Mater.* 1993, **27**, 572
14 Lo, K. H. and Chim, E. S. M. *J. Reinforced Plastics Composites* 1992, **II**, 839
15 Caiazzo, A. A., Sullivan, B. J. and Rosen, B W. in 'Proceedings of the Sixth Technical Conference of American Society for Composites' 1991, pp. 659–670
16 Stief, P. S. *J. Composite Mater.* 1988, **22**, 819
17 Reifsnider, K. L. in 'Proceedings of International Colloquium on Durability of Polymer Based Composite Systems for Structural Applications', Free University of Brussels, August 1990, pp. 27–31
18 Reifsnider, K. L. in 'Mechanics of Damage and Fatigue', (Eds S. R. Bodnen and Z. Hashin), Pergamon Press, New York, 1986, pp. 739–750
19 Reifsnider, K. L. in 'Proceedings of Spacecraft Structures Conference', Noordwijk, The Netherlands, 24–26 April 1991, European Space Agency SP-238
20 Reifsnider, K. L. and Stinchcomb, W. W. in 'Composite Materials: Fatigue and Fracture' (Ed. H. T. Hahn), ASTM STP 907, American Society for Testing and Materials, Philadelphia, 1986, pp. 298–313