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FAILURE PROCESSES GOVERNING LONG TERM RELIABILITY OF CARBON FIBRE COMPOSITE STRUCTURES

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Advanced composites are now used in many critical civil applications for which long term reliability is an absolute necessity. Testing based on more traditional metal structures has been shown to be inappropriate and there is a pressing need to develop new tests based on an understanding of damage processes in composite structures. Multi-scale modelling which takes into account the characteristics of the fibres, matrix and fibre/matrix interfaces is now well developed. It allows the kinetics of fibre failure during monotonic loading and sustained loading of composites to be explored and identifies the critical damage levels leading to failure for these loading conditions. This information allows safety factors based on the intrinsic properties of the composites to be determined and quantified.

ПРОЦЕССЫ РАЗРУШЕНИЙ, ОПРЕДЕЛЯЮЩИХ ДОЛГОВРЕМЕННУЮ НАДЁЖНОСТЬ КОНСТРУКЦИЙ, АРМИРОВАННЫХ УГЛЕВОЛОКНОМ

Современные композиты используют в настоящее время во многих критически важных гражданских приложениях, для которых долговременная надёжность абсолютно необходима.

Методы испытаний, применяемые для более традиционных металлических конструкций, как сейчас ясно, не дают достаточно надёжных результатов, будучи применёнными к композитным конструкциям. Поэтому очевидна необходимость разработки новых методов, основанных на понимании процессов повреждения композитных элементов. Многоуровневое моделирование, учитывающее характеристики волокон, матрицы и границы раздела, к настоящему времени достаточно развито. Оно позволяет исследовать накопление повреждений на стадии их устойчивого роста и определить критическую величину повреждения, определяющую разрушение конструкции в заданных условиях нагружения. Эта информация позволяет определить количественно коэффициент безопасности на основе знания характеристик структуры композита.

Introduction

Advanced composites made up of carbon fibres in an epoxy matrix are now being used in many applications for which long term reliability is an absolute necessity. Such materials are replacing traditional materials or enabling the manufacture of structures hitherto difficult or impossible to make due to their superior characteristics, amongst which are; stiffness and strength linked to light weight; ease of manufacture and corrosion resistance. The still high cost of carbon fibres means that these composites are being primarily used in critical structures the failure of which must be avoided. An important example is their use in the storage of gases such as natural gas and hydrogen at very high pressures. Such pressure vessels have been used for more than a decade for storing natural gas at 20 MPa as fuel for buses and other vehicles. Such pressure vessels have become one of the biggest markets for the carbon fibre industry. The forthcoming hydrogen economy will require much higher pressures of up to 90 MPa so as to ensure sufficient autonomy for the vehicles. Such high pressures mean that reliability must be absolute as failure could be devastating and would put lives at risk. As has previously been discussed the present international standards for assessing the reliability of pressure vessels are based on the failure processes in metal, usually steel, vessels (1). However the failure processes in composites are very different from those encountered in metals which fail by a major crack developing whereas composites fail by a more diffuse process of fibre breakage. In an acknowledgement of this, such pressure

vessels are over designed with little quantifiable evaluation of damage processes controlling long term reliability. The cost of making a carbon fibre composite pressure vessel is dominated by the cost of the fibres which can represent seventy-five percent of the total cost. This could mean that the development of the technology is being hindered by unnecessary high costs which a better understanding of the mechanisms of failure could alleviate.

It has been shown that even the most stable form of carbon fibre composite, consisting of all the fibres aligned in the loading direction, continues to experience fibre failure during periods of steady state or cyclic loading (2, 3). As the fibres support all but around one percent of the applied load this means that long term loading leads to the progressive deterioration of the composite and can lead to delayed failure (4). This is directly pertinent for the reliability of pressure vessels as the fibres are wound on geodesic paths so that when the vessels are under pressure the fibre experience only tensile forces within the plane of the walls of the pressure vessels. The most critical part of the composite envelope of the pressure vessels is the layers in which the carbon fibres are place at right angles to its major axis as these circumferentially wound layers determine the circumferential failure stress of the pressure vessel. The rate of fibre failure in pressure vessels and unidirectional specimens, as detected by acoustic emission, has been seen, at first sight, to be similar in form to that of a typical creep curve, although no overall macroscopic creep can be measured. Under a constant load or constant amplitude cyclic loading damage increases in a logarithmic manner as a function of time and does not stop. It has been proposed that such behaviour, as measured by the rate of damage detected by acoustic emission as a function of time, could be used to predict time to failure and so be used as a means of determining future reliability of the structure (1, 6). Multi-scale modelling of failure processes in these composites has revealed how the elastic carbon fibres can progressively fail due to the viscoelastic relaxation of the resin in the vicinity of pre-existing fibre breaks (5 - 9). However practical constraints and in particular the extraction of the relevant information from acoustic emission monitoring means that the direct application of the monitoring technique for testing composite pressure vessels is not straight forward.

The present paper shows how critical damage levels can be determined for carbon fibre composites under different loading conditions. It is proposed to use such an understanding to determine lifetimes of composite structures under load and to identify the load levels which lead to acceptable lifetimes. This is an essential step in determining safety factors so as to increase the reliability and optimisation of carbon fibre composite pressure vessels.

Modelling of damage processes in composites

Unidirectional composites or indeed the fibres in the walls of pressure vessels experience tensile forces when under load. The reliability of the structure depends on the capacity of the fibres to withstand these loads and for inservice lives of tens of years the fibres must be able to withstand the applied stresses over these periods. Carbon fibres seem to be perfectly elastic so that they do not show any time dependent behaviour (10). Embedded in a matrix these fibres support all but a small percentage of the applied load and are often assumed to dominate long term behaviour. However it has long been known that carbon fibres in such composites do progressively fail (2). This is due to the viscoelastic nature of the matrix in which the fibres are embedded and the relaxation of which allows the stress field around pre-existing fibre breaks to evolve. This leads to a progressive local increase in load in intact fibres neighbouring fibre breaks. As there are often many millions of fibres in a composite structure and the strength of the fibres is stochastic the result of these local stress increases is that some fibres eventually break. The rate at which the fibres break under steady applied loads or cyclic loading can be modelled analytically and the time to failure of a structure such as a pressure vessel can be deduced. This has been proposed as a means of monitoring the ageing of pressure vessels but the uncertainties of interpreting the acoustic emission means that other approaches need to be explored.

The model of this type of damage accumulation both in monotonic tests to failure and under steady loads is now quite mature and has been validated on both laboratory specimens and pressure vessels. It is a multi-scale model which allows the effects of applied stress to be examined at the level of groups of single fibres taking into account the stochastic nature of fibre strength, the viscoelastic nature of the epoxy resin in which they are embedded and also the debonding of fibre/matrix interfaces around fibre breaks. This requires the determination of the <u>Representative</u> <u>Volume Element or RVE</u> which has been shown to contain thirty two fibres (11). The composite is therefore made up of many RVEs and their combined behaviour is simulated using a finite element model which homogenises and

sums the behaviour at the level of the RVEs. In this way the problem of summing all the details within the RVEs, which would involve an impossible level of computing power to do so for a complete structure, is circumvented.

Details of the model can be found in several publications (1, 9 and 12) and only improvements of the model will be described here.

This time dependent behaviour has been modelled and shown to be due to the viscoelastic behaviour of the matrix (8). Experimental verification of delayed failure of unidirectional carbon fibre composites has been reported by Bunsell et al. (5). The progressive accumulation of fibre breaks when unidirectional carbon fibre composites are under load has been observed using high resolution tomography by Scott et al. which has revealed the creation of clusters of breaks occurring just before composite failure (13 - 15). The model has been shown to accurately predict the effects of changes to the rate of pressurisation of carbon fibre epoxy composite pressure vessels and to be able to quantify the changes in burst pressures which are observed experimentally (16).

The observation of damage development at the level of individual fibres, each of which has a diameter of $7\mu m$, in a composite structure is fraught with difficulties as there are so many of them. Notwithstanding the remarkable results shown by Scott et al (13 – 15), using high resolution tomography, the model has been shown to be sufficiently detailed that it can be used to explore the kinetics of damage accumulation in composite structures (16) It is less limited in specimen size than would be necessary in experimentally testing composite structures.

The modelling has therefore been aimed at determining if critical damage levels can be determined for different loading conditions so as to define loading conditions which could guarantee that in-service failure could never occur.

Kinetics of damage accumulation in carbon fibre composite structures

Two types of loading will be considered in the case of unidirectional carbon fibre reinforced epoxy resin which, as discussed above, is analogous to the composite structure in a pressure vessel. The first type of loading is monotonic to failure which occurs within a few minutes at most. Under these conditions the viscoelastic nature of the matrix has a negligible effect on overall behaviour and the main process is fibre failure. The second type of loading involves taking the composite to a predetermined fraction of its ultimate failure stress, as determined in monotonic tests, and then holding the load constant. The model then allows the effects of steady loading over a simulated period of many years or decades, typically twenty years, to be determined. The composite is then loaded directly to failure so as to determine the level of damage which produces damage under these conditions. In this latter case the viscoelastic nature of the matrix become dominant in determining damage accumulation.

The model as described above has been extended so as to determine the kinetics of fibres failing, including the clustering nature of breaks as the load is increased. The number of fibre breaks in the RVE is given by i-plets, where 'i' represents the number of broken fibres in a RVE, so that when no fibre is broken it is described as a 0-plet, two breaks are described as a 2-plet and so on until all thirty two fibres are broken, which is described as a 32-plet.

On loading, fibres fail randomly throughout the composite. Their points of failure reflect the random nature of defects controlling fibre failures and this stochastic behaviour can be modelled using Weibull statistics and introduced into the model using a Monte Carlo simulation. This means that initially and in reality for most of the loading, low level i-plets are created involving one or two associated breaks. However a point is reached at which clusters of fibre breaks begin to be created and this is the point at which the composite begins to become unstable. Figure 1 shows how the tensile behaviour of the composite remains linear nearly up to the point I in the Figure which is the point at which clustering of breaks begins. Very few breaks are involved so that only 4% of the fibres in the RVEs making up the structure are broken and only 3% of all possible 32-plets have developed at this point. Almost immediately after the point I is reached the composite becomes unstable at point J and failure occurs. At this point clusters of 32 fibre breaks have occurred somewhere in the composite and failure inevitably follows. At the failure point J only around 8% of all possible fibres breaks have occurred in all the RVEs making up the composite and 70% of all possible RVEs have not been damaged at all with only around 6% of all possible 32-plets having developed. It was also seen that approximately 80% of the total number of fibre breaks is the critical damage process controlling failure. Figure 1 shows that failure occurs at or just after the point J is reached but indicates an instantaneous



Figure 1. A simulated tensile curve showing that failure occurs at point J immediately after when large clusters of breaks form around point I which is the last point which can be discerned experimentally. The plateau represents instant failure of all the fibres in the section containing the largest cluster and the final stage (C-D) the failure of the resin Расчетная кривая растяжения, показывающая, что разрушение возникает в точке J, которая оказывается последней, могущей быть зарегистрированной экспериментально. Плато отражает мгновенное разрушение всех волокон в сечении, содержащем наибольший кластер. Финальная стадия (C-D): разрушение полимера

extension of the composite until all the fibres in the section containing the critical 32-plet cluster are broken leaving only a contribution of the matrix which then breaks. This is similar to behaviour predicted by the ACK model and is not considered to be realistically observable [17, 18]. The Point J is the point of instability. However, as mentioned by Thionnet et al. [4], it is considered that Point I represents the practical limit of damage for the composite as the critical level at Point J is too quickly attained to be observable. Then, by analysing the state of damage at the Point I, a critical damage state in the case of monotonic loading can be defined as around 5% of all possible fibres breaks leading to around 3% of all possible 32-plets and about 20% of the material (all possible RVEs) contains at least one broken fibre. The point of final instability occurs when about 8% are broken and failure follows immediately afterwards. An analogy can be made with the development of a critical crack in a metal structure which leads to failure under the applied stress.

Figure 2 shows the evolution of i-plets during monotonic tensile loading. All the RVEs in the composite are considered as being intact at the onset and little change is seen until around 50% of the breaking load above which the 0-plets decrease in number but 75% are still intact when 32-plets develop and failure occurs almost immediately afterwards.

Under steady loading the accumulation of damage has been shown to be of a different nature. The composite is, by necessity, loaded to a lower level than the breaking load and can therefore sustain greater damage without breaking than that which caused failure in the monotonic test. The viscoelastic nature of the resin causes the matrix material around fibre breaks to relax resulting in a progressive increase in stress in the intact fibres neighbouring the breaks. Some of these fibres break and clusters of fibres can develop early during sustained loading (19). In order to determine if the critical damage level is different under these loading conditions the simulated loading has examined the effects of holding the composite at different percentages of the ultimate monotonic tensile failure load, from one percent to ninety percent of the breaking load, for a simulated period of twenty years and then, without unloading, taking the composite to failure. Figure 3 shows a schematic representation of the results of this calculation. In contrast to the monotonic loading case sustained loading leads to a much more diffuse damage so that when failure



Figure 2. All RVEs are originally intact and little damage occurs until around 50% of failure load at which point low level *i*-plets appear leading to a fall in 0-plets however approximately 75% are still intact when clusters of 32-plets are created and failure occurs immediately afterwards. The strain is seen to increase linearly until failure.

Все представленные объёмные элементы (RVE) не имеют разрушений, лишь небольшие разрушения волокон наблюдают при нагружении до примерно 50% от разрушающего, когда возникают кластеры, содержащие небольшие і числа обрывов волокон (i-plets) при этом 75% RVE всё ещё не имеют разрушений. Когда возникают 32-plets, разрушение наступает немедленно. Деформация, как видно, растёт линейно вплоть до разрушения.



Figure 3. During sustained loading damage in the form of clusters of breaks is accumulated throughout the test and on being taken to failure after a simulated period of twenty years the damage level can be identified. В течение периода устойчивого роста повреждений, кластеры разрушений накапливаются. После 20-летнего срока вычислительного эксперимента уровень повреждений может быть определён.

occurs at one point there are many other points throughout the composite which are almost just as vulnerable. The damage level at the point of instability under sustained loading is approximately twice that which is calculated for monotonic loading, irrespective of the sustained loading level, as can be seen in Table 1. Under these conditions approximately 6% of all fibres in all of the RVEs are broken at the point I and at the point J 15% are broken. The role of the viscoelastic nature of the matrix is clearly demonstrated under long term loading.

Table 1

Numbers of all possible fibre breaks in the RVEs and percentage of all possible 32-plets under monotonic loading and also sustained loading, to different percentages of the monotonic failure load, for a simulated period of twenty years followed by loading to failure

Monotonic loading conditions		
	Fibre breaks within all RVEs	Percentage of all
	(%)	possible 32-plets (%)
	8.44	6.65
Sustained loading conditions then taken to failure after a simulated 20 years		
Sustained load level as	Fibre breaks within all RVEs	Percentage of all
percentage of monotonic	(%)	possible
breaking load (%) = stress		32-plets (%)
(MPa)		
1.0 %/ 29 MPa	13.19	11.46
2.0 %/ 58 MPa	13.26	11.52
3.0 %/ 88 MPa	13.38	11.64
4.0 % / 117 MPa	13.41	11.69
5.0 % / 146 MPa	13.56	11.82
10.0 %/ 292 MPa	14.03	12.29
20.0 %/ 584 MPa	14.11	12.37
30.0 %/ 876 MPa	14.44	12.72
40.0 %/ 1168 MPa	14.66	12.94
50.0 %/ 1468 MPa	15.18	13.42
60.0 %/ 1753 MPa	16.13	14.29
70.0 %/ 2045 MPa	18.34	16.32
80.0 %/ 2337 MPa	21.58	18.29
82.0 %/ 2395 MPa	17.00	15.43
84.0 %/ 2454 MPa	20.83	17.99
86.0 %/ 2512 MPa	17.12	15.57
87.0 %/ 2541 MPa	15.82	14.52
88.0 %/ 2570 MPa	15.62	14.38

Time to failure

The accumulation of damage as identified above clearly takes the composite towards ultimate failure and this can be quickly demonstrated by loading unidirectional carbon fibre epoxy specimens to near their monotonic tensile failure load. Figure 4 shows the failure of such specimens loaded to 96% of the breaking load. The specimens showed a range of lifetimes reflecting the stochastic nature of failure in these specimens. Similar tests at lower loads show a quickly increasing time to failure so that these tests have been simulated using the above mentioned model. Figure 5 shows that as the loads are reduced the times to failure quickly increase as does the scatter predicted in the time to failure (5). These results raise the possibility of defining loading levels below which failure becomes increasingly unlikely and it becomes possible to identify a minimum safety factor based on the intrinsic



Figure 4. Acoustic emission monitoring of damage accumulation in unidirectional carbon fibre composites loaded in the fibre direction to 96% of breaking load. The specimens broke over a range of lifetimes from around one hour to fourteen hours

Рост сигналов акустической эмиссии, отражающий накопление повреждений в однонаправленном композите с углеволокнами, нагружаемом в направлении армирования напряжениями до 98% от разрушающего. Времена разрушений образцов - от ~1 до 14 час



Figure 5. Lifetimes of unidirectional carbon fibre composites loaded to different fractions of ultimate breaking load showing how the lifetimes increase as does the scatter in lifetimes as the loads are reduced Времена жизни однонаправленного композита, нагружаемого до различных величин относительно предельного напряжения при кратковременном разрушении. Можно также видеть разброс времён жизни

characteristics of the components making up the composite. This does not include other possible damage processes such as manufacturing faults or poor handling. Failure is a probabilistic process so it is not logical to say that failure can never occur but the model can be used to calculate loading conditions which lead to a statistical value of failure which would preclude in-service failure over lifetimes much greater than those required. For this reason the model has been used to calculate the load level at which a probability of failure of one in a million over fifteen years and also over one hundred and fifty years. When these studies are extended to even lower probabilities of failure it becomes clear that the intrinsic safety factor for these carbon fibre composite structures lies between 1.4 and 1.6.

Conclusions

Modern computing techniques have allowed the kinetics of damage accumulation in advanced composites to be ascertained. It has been shown that, in the case of carbon fibre reinforced composite structures, in which the fibres are subjected to tensile forces, as in unidirectional and internally filament wound pressure vessels and pipes, damage is initially random on loading. When the composites are loaded fibres break at weak points and these breaks are randomly distributed throughout the structure unless there are particular stress concentrations. During monotonic tensile tests a point is reached where clusters of fibre breaks begin to accumulate and failure occurs quickly afterwards. The number of broken fibres is small with only around 4 or 5% broken, of all the fibres in the RVEs making up the structure, when the observable point of instability is reached. Under sustained loading, which can be over periods of decades, the viscoelastic nature of the matrix material induces increasing stresses in intact fibres in the composite experiences lower stresses than at failure in a monotonic tensile test and can support the presence of clusters of fibre breaks, at least for some time. The critical damage level in such composite structures after prolonged loading has been shown to be approximately twice that which causes failure in monotonic tests with around 8% of all fibres being broken at the observable point of instability.

The quantification of critical levels of damage under both monotonic and sustained loading allows the identification of critical damage limits which have been used to identify intrinsic safety factors for composite structures. In the absence of manufacturing defects or damage caused by mishandling it has been shown that the present safety factors are excessive and could be reduced.

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