JOURNAL OF MATERIALS SCIENCE 27 (1992) 1811–1817

Deterministic method of describing rupture probability application to the analysis of high-modulus carbon fibres

E. BOURGAIN, J. J. MASSON
Institute for Materials Research, DLR, D-5000 Köln 90, Germany

The fitting of experimental fibre strength distribution by a Weibull distribution function must theoretically allow the prediction of strength distribution in other conditions as, for example, other gauge lengths, because it is the hypothesis of the Weibull model that the material is well described. It may be assured that this last assumption is almost always fully unrealistic, and applied to the case of aluminium-coated high-modulus carbon fibres, the Weibull statistics do not lead to any convenient prediction of the size effect. A new approach is proposed. It is based on a precise description of the real population of defects and no longer on an idealized and a priori simplified distribution as proposed by the Weibull model. It is completely deterministic and does not require any parameter adjustment. It permits an extremely precise prediction of the experimental strength distribution at other gauge lengths, exactly describing, for example, the experimentally observed jumps in the curve (P_r–σ_r). Moreover, it results in a very significant optimization of testing procedure, in determining only the necessary gauge lengths at which strength measurements have to be made in order to allow a complete description of the defect population and thus a confident strength prediction at every gauge length. Therefore, by giving a quantitative representation of the critical defect distribution in the material by a function of their failure probability, the damage, caused by annealing treatment or chemical reaction, may be thoroughly analysed as a function of the microstructure of the material and particularly at the level of the critical defects, whose evolution (increasing of failure propensity) may be easily followed. This method has been applied very successfully in the case of aluminium-coated high-modulus HM35 carbon fibres.

1. Introduction

The mechanical properties of polymer-, metal- or ceramic-matrix composites are widely correlated to those of their reinforcement. In the particular case of metal-matrix composites (MMC) made by hot-pressing of prepreg layers, their quality mainly depends on the quality of the precursor and then of the fibres. The experimental results presented below concern C/Al MMC [1].

A rough statistical characterization of the fibres (the Weibull parameter, m, values about 6) leads to the assumption that the composite must break when only 15% of the fibres are broken, because the bonding between fibres and matrix is weak (Coleman's model [2]). The fracture surface observations confirm this assumption (Fig. 1). The study of the C/Al MMC strength can then amount to the study of the reinforcing fibres. The optimization of the quality of the MMC by its manufacturing can come down to establishing how the fibre strength is degraded by reaction with aluminium during hot-pressing.

Single fibres, which are representative of the reinforcing fibres in the MMC, have been subjected to tensile tests at six different gauge lengths. The Weibull statistical approach has been used to analyse the strength measurements and to allow the prediction of strength distributions at other gauge lengths, for example, at greater gauge length, to calculate a real structure as well as elementary length in a fracture model of composite material [3]. The Weibull statistics have been shown here to be inadequate to give a good prediction of the size effect.

A new statistical mechanics approach of the fracture, presented below, has therefore been developed. It allows an extremely precise prediction of the experimental strength distributions at other gauge lengths. Moreover, it allows one to reduce very significantly the testing procedure necessary for confident strength prediction at every gauge length. It is based on a precise description of the real population of critical defects (able to lead to rupture) and no longer on an idealized and a priori simplified distribution, as proposed by the Weibull model.

Consequently, the damaging of the carbon fibre by reaction with aluminium may be followed separately for each defect, according to its failure propensity (size, shape, kind of defect, etc). As an application it is now possible to make a qualitative and quantitative prediction of the damage undergone at the level of more critical defects, which cannot be observed experi-

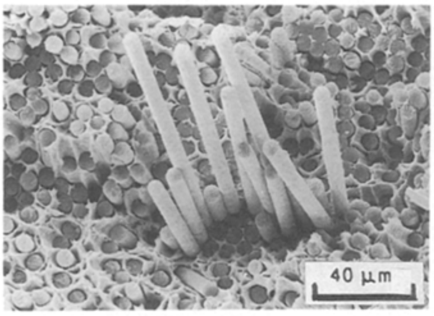


Figure 1 Fracture surface of C/Al composite.

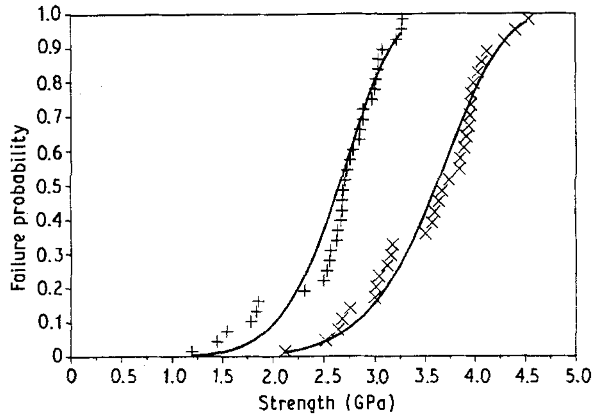


Figure 2 Weibull correlation curves of experimental strength distributions at the two extreme gauge lengths. $L = (+)$ 40 mm, $(\times)$ 2.5 mm.

mentally through tensile tests, because it would require too small gauge lengths, but which may control the rupture of the MMC.

2. Size effect: the Weibull approach

Single high-modulus HM35-fibres have been subjected to an annealing treatment with aluminium, simulating temperature and atmosphere conditions of composite processing [1]. They are denoted "HM35/Al" below. Tensile tests were then made using a method similar to ASTM-D 3379 [4], at six different gauge lengths, L. The two extreme experimental strength distributions (cumulative failure probability function) as well as their Weibull correlations, $F(\sigma)$, are reported in Fig. 2.

$$F(\sigma) = 1 - \exp\left[-\left(\frac{\sigma}{\sigma_0(L)}\right)^m\right] \quad (1)$$

As the Weibull statistics fit the failure distribution of HM35/Al well, the Weibull parameter m must be constant from one length to the other and the scale parameter, $\sigma_0(L)$, must verify Equation 2

$$\ln \sigma_0(L) = K - \frac{1}{m}\ln L \quad (2)$$

where K is a constant. It has already been demonstrated [5] that a great error (more than 35%) may be expected in the determination of the parameter m when it is deduced from a distribution at fixed length. The estimation of the parameter $\sigma_0(L)$ is, however, always very confident (< 5% error).

In the case of HM35/Al, the Weibull parameter, m, of the calculated strength distribution at fixed length varies between 4.4 and 7.5. The average value of $m = 6$ may be retained for characterizing the HM35/Al fibres, the experimental results being within the expected interval of estimation error (Fig. 3a). According to Equation 2, another determination of this parameter has been made from the regression line describing the effect of the real size on the scale parameter (Fig. 3b, c). The new estimation is then $m^* = 11.5$, almost twice the previous one. Moreover, the correlation factor $r = 0.85$, is relatively bad. Another approach, that includes a threshold stress, σ_u, has been attempted

$$F(\sigma) = 1 - \exp\left[-\frac{L}{L_0}\left(\frac{\sigma - \sigma_u}{\sigma_0}\right)^m\right] \quad (3)$$

The physical meaning of this additional parameter can be explained as follows: the rupture stress of a very long fibre corresponding to 63% cumulative failure probability would not be equal to zero, but be $\sigma_u > 0$. In this way, the correlation factor is much better: $r = 0.99$ when $\sigma_u = 2617$ MPa, but the new regression parameter, $m^* = 0.94$, is absolutely unrealistic as a Weibull parameter describing the experimental distribution.

In this particular case of carbon fibres, as well as in other cases of carbon, alumina and SiC fibres already tested, the Weibull statistics prove unable to predict confidently the size effect, i.e. the decrease in the measured strength at given cumulative failure probability with increasing gauge length.

3. A new deterministic approach to rupture

The Weibull statistics is based on an idealized and a priori simplified distribution of critical defects in the material. It may now be assumed that this representation is insufficient and, indeed, completely inadequate for any precise prediction of the material behaviour. The new approach, proposed here is based on the idea that the material may be characterized by a continuous distribution of critical defects that are peculiar to the studied material and which may result from the manufacturing as well as from subsequent treatment or damage.

Thus, the HM35/Al surface shows a complete population of critical defects, more or less large, of various shapes (Fig. 4a). It may be assumed that, among these defects, some are severe enough to be able to lead to fracture. The observation of HM35/Al fracture surfaces justifies such an assumption (Fig. 4b).

3.1. Formulation of the model

Let us imagine an infinite fibre. It contains an exhaustive population of critical defects (defects which can lead to fracture when the fibre is under load). Each defect may then be characterized by the critical stress, σ_c, which would be sufficient to extend it up to fibre

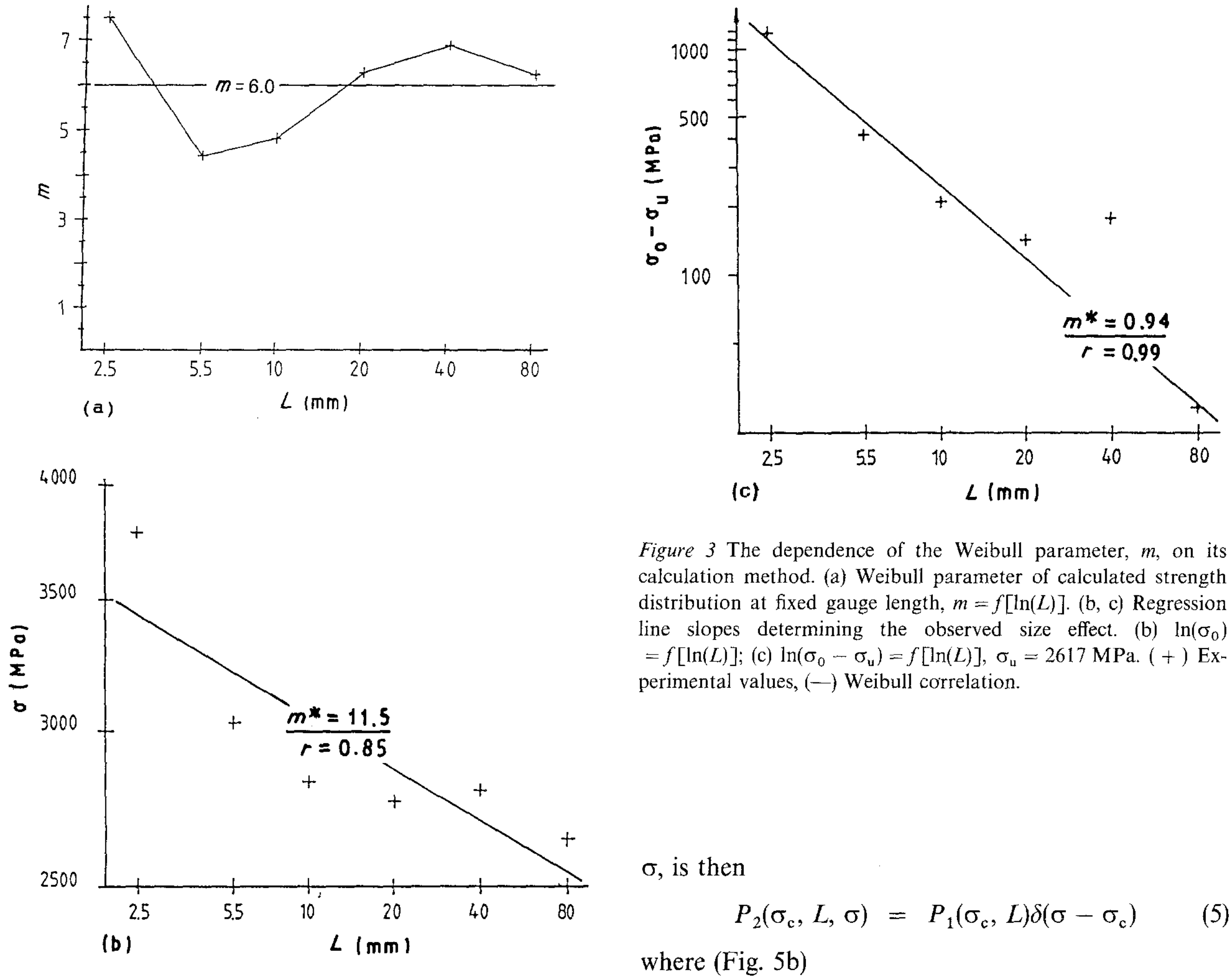


Figure 3 The dependence of the Weibull parameter, m, on its calculation method. (a) Weibull parameter of calculated strength distribution at fixed gauge length, $m = f[\ln(L)]$. (b, c) Regression line slopes determining the observed size effect. (b) $\ln(\sigma_0) = f[\ln(L)]$; (c) $\ln(\sigma_0 - \sigma_u) = f[\ln(L)]$, $\sigma_u = 2617$ MPa. (+) Experimental values, (—) Weibull correlation.

fracture. It is then denoted by "σ_c-defect". The σ_c-defects (σ_c fixed) distribution (or appearance probability) may be given by the characteristic length $L(\sigma_c)$, schematically separating two such defects (Fig. 5a).

The strength measurements are practically made with fibres of finite length, L, denoted "L-fibres". A L-fibre contains $N(L)$ defects which may be ordered according to their respective critical stresses: $\{\sigma_c(1) \leq \ldots \leq \sigma_c[N(L)]\}$. The $\sigma_c(1)$-defect is then the most critical defect of the L-fibre and $\sigma_c(1)$ must be thus the L-fibre strength.

By definition, the presence probability of a σ_c-defect in an L-fibre has the value

$$P_1(\sigma_c, L) = \varepsilon\left[\frac{L}{L(\sigma_c)}\right] \qquad (4)$$

where

$$\varepsilon(x) = \begin{cases} x \text{ if } x \leq 1 \\ 1 \text{ otherwise} \end{cases}$$

The probability that the σ_c-defect may lead to the fracture of an L-fibre subjected to the tensile loading, σ, is then

$$P_2(\sigma_c, L, \sigma) = P_1(\sigma_c, L)\delta(\sigma - \sigma_c) \qquad (5)$$

where (Fig. 5b)

$$\delta(x) = \begin{cases} 0 \text{ if } x < 0 \\ 1 \text{ otherwise} \end{cases}$$

The probability that an L-fibre is broken at the loading level, σ, is then

$$P_r(L, \sigma) = 1 - \prod_{\sigma_c}[1 - P_2(\sigma_c, L, \sigma)] \qquad (6)$$

which may be rewritten in case of a continuous distribution

$$P_r(L, \sigma) = 1 - \exp\left(\int_0^{\sigma} \ln\left\{1 - \varepsilon\left[\frac{L}{L(\sigma_c)}\right]\right\} d\sigma_c\right) \qquad (7)$$

Let us assume that tensile tests have been made at the gauge length L_0, and then, that the strength distribution at this length $P_r^0(\sigma)$ is known. It follows from Equation 7 that the strength distribution at the new gauge length, L, is given by

$$P_r(L, \sigma) = 1 - \exp\left[\int_0^{\sigma} \ln\left(1 - \varepsilon\left\{\frac{L}{L_0}\left[1 - \exp - \frac{\frac{dP_r^0}{d\sigma}(\sigma_c)}{1 - P_r^0(\sigma_c)}\right]\right\}\right) d\sigma_c\right] \qquad (8)$$

3.2. Discussion

This statistical approach of fracture allows us to predict the failure probability at every length from the strength distribution measured at fixed length, but only within the strength dispersion band observed at this length (in fact, where $P_r^0(\sigma)$ is known). Thus, to predict the failure probability at any length, corresponding to rupture stress smaller than the smallest or higher than the highest observed one, it would be necessary to make a hypothesis on the defect distribution shape in this unobserved stress field portion. This is what the Weibull statistics does directly, implicitly and with very strong and unrealistic hypotheses.

Moreover, if the experimental distribution, $P_r^0(\sigma)$, is determined for too great a length, L_0, such as there exists a value σ_c^* verifying $L(\sigma_c^*) < L_0$, no rupture of fibres can thus be observed over σ_c^*. To let the entire defect distribution appear, it would consequently be necessary to test the fibres at the smallest possible gauge length.

However, with a limited number of tensile tests at such a small gauge length, the probability is very small to reveal very critical defects, that are rare in the material. Consequently, a large number of tests must be performed at this length to give a chance to observe all the defects. The most sparing testing work method is thus to perform tensile tests at different complementary gauge lengths.

3.3. Application to the experimental results of HM35/Al

According to the previous discussion, two experimental distributions would have been enough to give the present description of the HM35/Al material: 2.5 mm, the smallest tested gauge length and 20 mm, which allows optimal scanning of the critical defects dispersion (Fig. 6). Indeed, the σ_c-defect ($\sigma_c \approx 3.0$ Gpa) corresponding to the failure probability of 20% for the gauge length 2.5 mm, correspond to a failure of 75% for the gauge length 20 mm. The more critical defects ($\sigma_c < 3.0$ GPa) are observed through the strength measurements at the gauge length 20 mm, the less critical defects ($\sigma_c > 3.0$ GPa) through the strength measurements at 2.5 mm. To be able to give such a precise description of the defect distribution in the fibre, it would be necessary to make more than 1000 tests at a fixed gauge length between 2.5 and 20 mm, instead of about 60 tests in the method used here. The new approach thus allows savings of about 95% of otherwise needed tensile tests. To describe a new complementary part of the critical defect distribution, it would be optimal to test 30 HM35/Al single fibres at the new gauge length of 150 mm which is practically unrealizable.

It is of great interest to emphasize that the observed jumps in the experimental curves no longer result only from a statistical error, as classically considered, but essentially reveal the real concentrations of defects around some critical stress levels (here, for example, 1.8, 2.4, 2.7, 3, 3.2 and 4 GPa). The prediction of the distribution at a new gauge length from an experimental distribution must consequently reproduce these jumps. For example, the distribution at 2.5 mm has been calculated from the experimental one at 20 mm, and the perfect fitting with the experimental curve at 2.5 mm is noteworthy (Fig. 7).

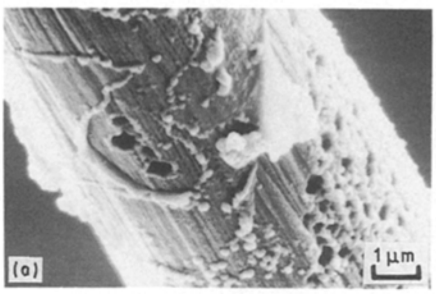
4. Analysis of damage by annealing treatment of HM35/Al fibres

4.1. Interface reaction of HM carbon fibres with aluminium

High-modulus carbon fibres react with aluminium at temperatures above 500 °C in a vacuum to form aluminium carbide, Al_4C_3 [6]. During the initial stage of the reaction the carbides are produced from active sites on the fibre surface, e.g. at edges of carbon-based planes perpendicular to the fibre surface or at geometrical irregularities on the surface. In a second stage they grow both into the matrix and into the fibre. The bonding at the interface between carbides and matrix is more stable than between carbides and fibre [7]. Therefore, the roots of carbide crystals act as notches on the fibre surface (see Fig. 4), which has a weakening effect on the fibre strength. Carbide formation is diffusion-controlled and therefore dependent on time and temperature [8].

4.2. Experiments

In order to examine the influence of fibre/matrix reaction on the mechanical properties of HM35/Al, single fibres were tested in tension after different annealing treatments (at 550, 600 and 650 °C for 10, 30 or



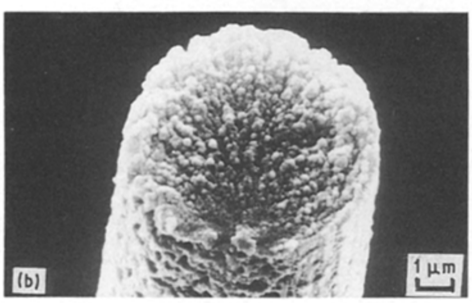
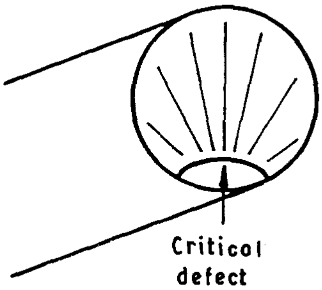


Figure 4 Critical defects distribution in the material. (a) HM35/Al fibre surface (after annealing treatment and dissolution of aluminium matrix). (b) Crack propagation.



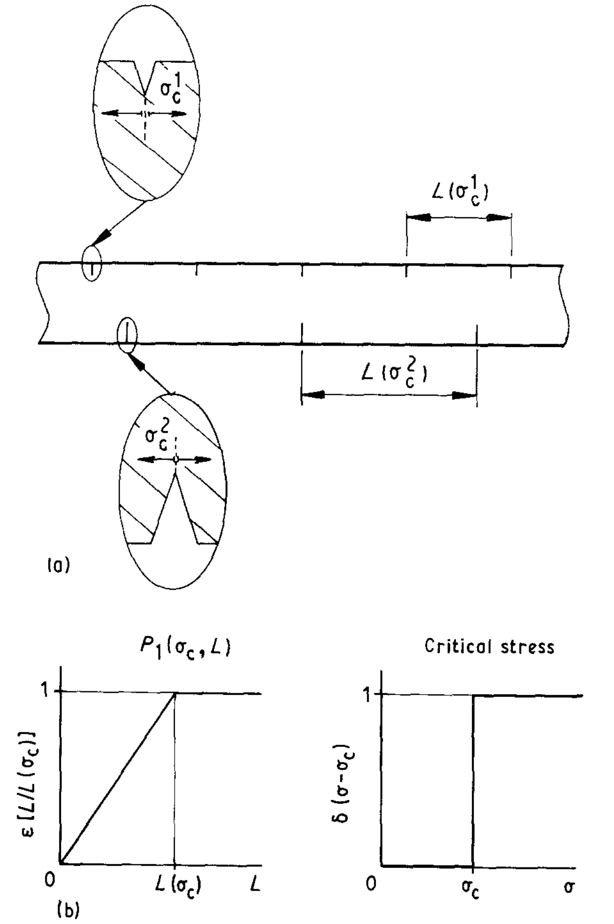


Figure 5 Deterministic approach of the rupture. (a) Representation of the defects distribution, L (σ_c). (b) Model of rupture.

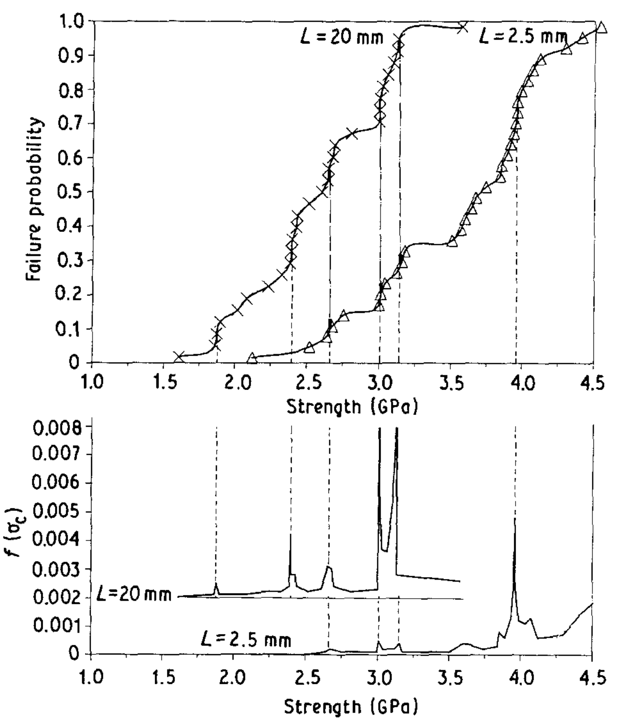


Figure 6 Strength and observed defect distributions at different gauge lengths.

60 min), which simulated the temperature and atmosphere conditions of composite processing. Tests were performed at a fixed gauge length, L, of 5.5 mm. More details on the experimental procedure are reported elsewhere [1, 9].

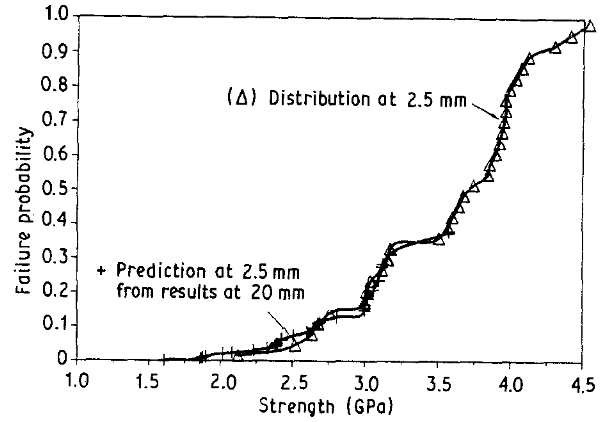


Figure 7 Application of the approach to the prediction of strength distribution at other gauge lengths.

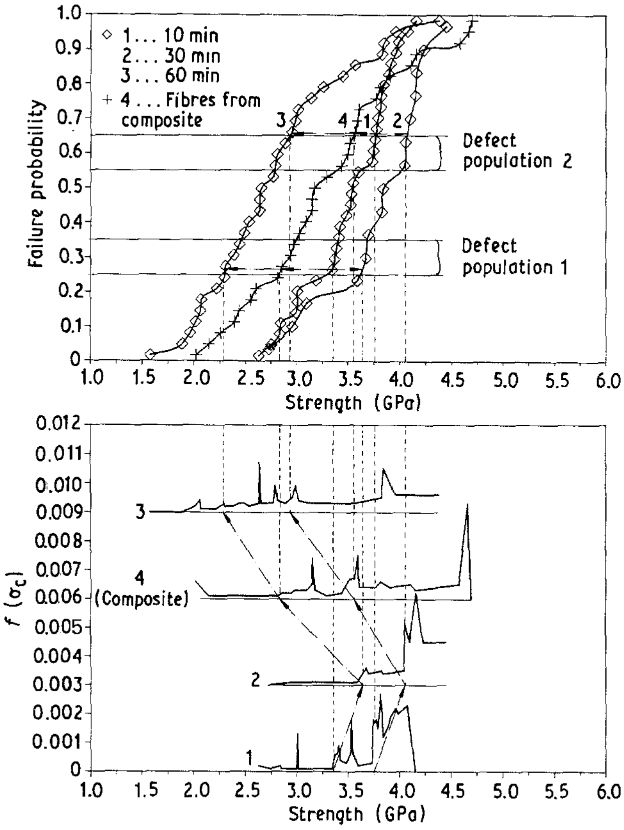


Figure 8 Strength and defect distributions after annealing treatment at 600 °C. (◇): 1, 10 min; 2, 30 min; 3, 60 min. (+): 4, fibres from composite.

4.3. Results and discussion

As an example, Fig. 8 shows cumulative strength distributions obtained after annealing treatment at 600 °C for different times. Each step observed in the distribution curves corresponds to a σ_c-defect population, whose probability to appear (i.e. concentration) along the fibre is high, as demonstrated in Section 3. Each peak of the defect distribution corresponds to a jump in the strength distribution (with the model proposed here, corresponding defect distributions have been calculated, Fig. 8).

In the initial strength distribution (here 600 °C, 10 min), each σ_c-defect appears in a certain band of failure probability, corresponding to a jump whose

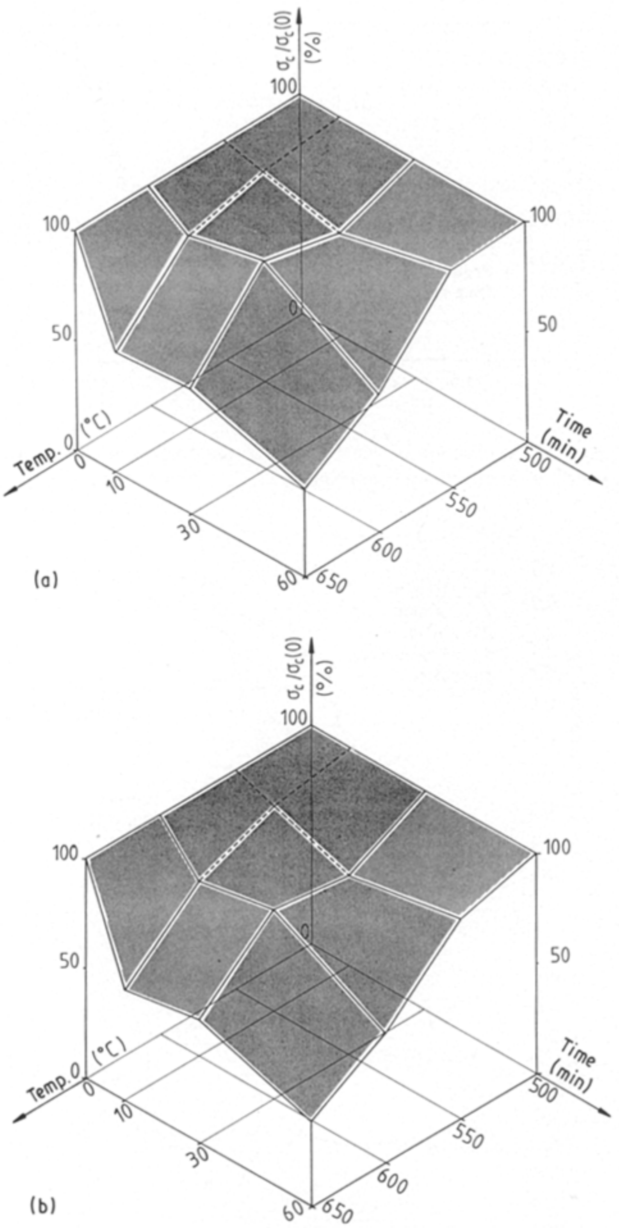


Figure 9 Relative critical stress of "$P_{\Delta P}$-defects" as a function of annealing temperature and time. (a) Population 1, $0.25 \leqslant P_r(\sigma_c) \leqslant 0.35$; (b) population 2, $0.55 \leqslant P_r(\sigma_c) \leqslant 0.65$.

average level is the failure probability, P, and the height is ΔP. These values are correlated, respectively, with the order of appearance of the defect and its concentration. These defects are denoted "$P_{\Delta P}$-defects". Because all tests were performed at the same gauge length, the defect populations previously denoted "σ_c-defects" correspond here to "$P_{\Delta P}$-defects".

When the same batch of fibres undergoes a stronger annealing treatment, the carbide crystal roots grow. Thus a $P_{\Delta P}$-defect is growing in size, causing a variation of its critical stress. However, in an ideal case where all $P_{\Delta P}$-defects are growing in the same relative properties, it can be assumed that their order of appearance and their concentration in the strength distribution remain unchanged. Only the critical stress of these defects will vary. Therefore, the shape of the distribution will be the same, with the same steps. However, the distribution curve will be translated horizontally. That is what is observed in Fig. 8 for failure probabilities between 0.25 and 0.75.

Two $P_{\Delta P}$-defect populations have been chosen in order to plot their relative critical stress as a function of annealing temperature and time: population 1 (or 2) corresponding to failure probability between 0.25 and 0.35 (or 0.55 and 0.65). The stresses are normalized through the critical stress of the same defects in untreated fibres. Fig. 9 is a three-dimensional representation of the results, whereas Fig. 10 shows lines of iso-relative critical stress. The influence of annealing treatment is similar for both populations of defects. The relative critical stress has a local maximum at 30 min, 600 °C. At a constant temperature of 600 °C, for instance, the strength decreases between 0 and 10 min, increases between 10 and 30 min, and then decreases again after 30 min. This should be related to the mechanism of carbide formation. As a hypothetical sequence we propose: (1) formation of carbide

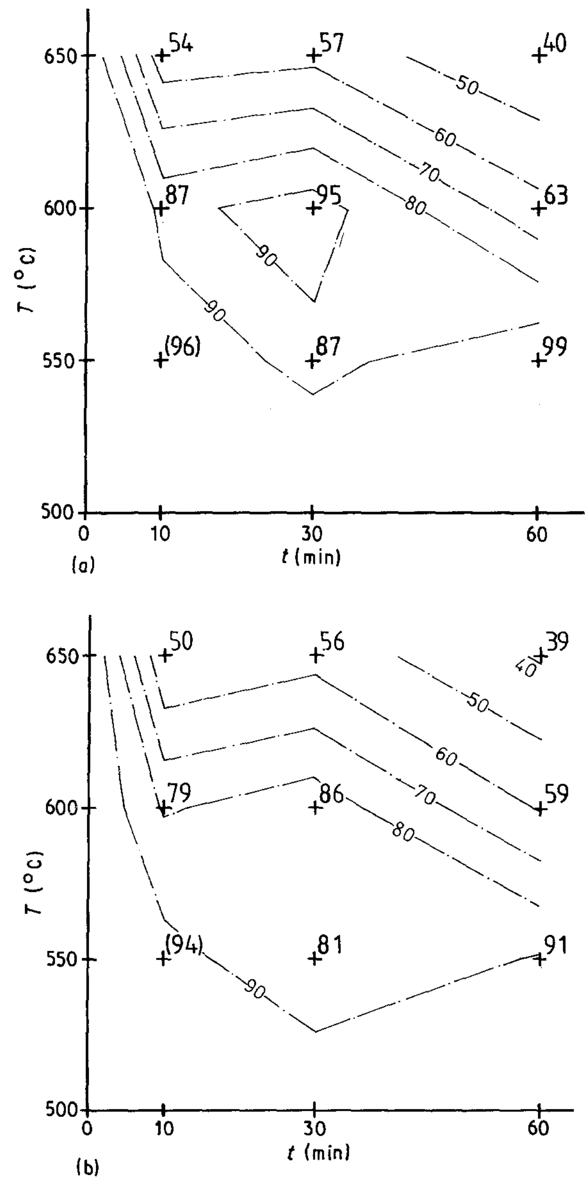


Figure 10 Iso-relative critical stress of $P_{\Delta P}$-defects. (a) Population 1, $0.25 \leqslant P_r(\sigma_c) \leqslant 0.35$; (b) population 2, $0.55 \leqslant P_r(\sigma_c) \leqslant 0.65$.

roots in the fibre acting as notches (strength decrease); (2) diminution of stress concentration at the notch tip by smoothing of the defect due to growth crystals parallel to the fibre axis (strength increase); (3) carbide crystal growth into the fibre (strength decrease).

All these tests were performed to simulate the influence of composite processing temperature and time on the strength of fibres. To check this approach, individual tensile tests have been performed on fibres directly extracted from a composite plate. The results are also plotted in Fig. 8. Composite processing occurred by solid-phase diffusion bonding at 600 °C for 1 h. The shape of the distribution obtained is similar to that of aluminium-coated fibres annealed at 600 °C for 1 h, however, the distribution curve is translated horizontally to higher strengths. This may be an influence of the real processing temperature. Based on Figs 8 and 10b, and according to the fibre strength distribution, the real processing temperature of the composite must have been about 575 °C, instead of 600 °C (i.e. an error less than 5% in manufacturing temperature).

5. Conclusion

A new approach describing fracture probability has been proposed and has been applied very successfully in the case of HM35/Al fibres: the effect of fibre gauge length on the measured strength could be predicted very precisely, and the representation of the defect distribution was very helpful in the interpretation of results obtained after different annealing treatments.

The new method is based on the analysis of interpolated experimental results and no longer on the simple fitting by an a priori chosen function (e.g. the Weibull distribution). It leads, then, to an unbiased description of the defect distribution in the material and to a prediction of its fracture probability. It also indicates the limits of any prediction of the material's behaviour under conditions other than the observed ones (other gauge lengths, for example).

Nevertheless, it has been demonstrated that the new method offers a significantly more precise description of actual fracture strength distribution and it offers better predictive capability than do the standard Weibull statistics.

Acknowledgements

The authors thank Mr N. Dade, University of Bordeaux, France, for his meticulous experimental work, and Professor G. E. Welsch, University of Ohio, USA, for help in the preparation of the manuscript. The S. E. P., Bordeaux, France, and the European Community, Contract no. MA1E/0063-C, are thanked for funding this work. Additional thanks go to Dr K. Schulte and Professor H. Nowack for helpful discussions.

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Received 26 November 1990
and accepted 10 April 1991