AN INTERMEDIATE ADHESION QUALITY MODEL FOR THE EVALUATION OF STRENGTH AND INHERENT FLAW OF RESIN / FILLER SYSTEMS

By

G. Bourkas

National Technical University of Athens
Faculty of Applied Sciences
Department of Mechanics
Lab. of Strength and Materials
Zografou Campus,
GR-157 73, Athens, Greece

ABSTRACT

The tensile strength of particulate composites has been evaluated for the case that adhesion exists between matrix and filler by using a model consisting of four components, on the basis of cube –within-cube formation. Additionally the "inherent flaw" of the composite has been estimated with respect to the "inherent flaw" of the matrix, for the case of resin / filler systems. By comparing the theoretical results of the strength with experimental data for untreated particles in epoxy / filler systems, the model under consideration has been characterized as corresponding to intermediate adhesion quality between matrix and filler, since a satisfactory agreement between theory and experiments has been observed for the case of untreated particles of intermediate strength values. The systems used to compare the strength were resin / iron and resin / glass particulate composites. The strength values predicted by the present procedure are in agreement to those provided by an existing evaluation method in the literature.

INTRODUCTION

Although there is a considerable number of theories' describing the behaviour of elastic modulus of filled polymer systems, a satisfactory treatment of the behaviour of strength of composites reinforced with rigid particles has not yet been developed [1].

Ref. [2] presents the existing models for the estimation of the strength of particulate composites. In ref. [3] there is an extensive report for the stress intensity factor, K_{ic}, of resin / particles systems.

Although the addition of rigid particles to a polymer matrix tends to cause reduction of the strength of the filled material, the fracture toughness [3], K_{ic}, as well as the elastic modulus, are increased.

In ref. [4,5] treated and untreated particles have been used to examine the influence of the adhesion quality between matrix and filler on the strength. It was observed that the strength increases by improving the adhesion quality between matrix and filler. Additionally it was found that when there is adhesion between matrix and filler by increasing the filler content, the strength at first decreases then reaches a minimum value and then increases. In ref. [3] the highest fracture toughness, K_{ic}, for composites in which poor particle – matrix interphacial adhesion was also observed. In ref. [6] the highest level of toughness was achieved by introducing rubber particles in particulate – filled polymer in which simultaneous crack pinning effect (the crack propagation is embedded be a filler group) and localized plastic deformation occurred.

In this paper the tensile strength of particulate composites has been evaluated by using a four-components model on the basis of cube-within-cube formation. It has been found that increasing the filler content the strength initially decreases, for $u_f \cong 20\%$ the strength attains a minimum and for greater values of u_f the strength increases steadily with a slow rate. It was also found that the theoretical values of the strength are close to the experimental results of untreated epoxy/iron and epoxy/glass particulate composites. Moreover assuming a linear relations of the stress intensity factor, K_{ic}, with respect to filler volume fraction in epoxy/filler particulate composites an equation was developed which gives the inherent flaw of the composite.

THEORETICAL CONSIDERATIONS

The theoretical analysis is based on the following assumptions.

1) The particles are perfectly cubic.
2) The matrix volume distribution of each filler is also cubic.
3) The volume fraction of the particles is sufficiently low, so that there is no interaction between the stress fields around neighboring particles.
4) The particles are uniformly distributed in the matrix, so that homogeneity can be assumed.
5) Both the matrix and the inclusion are prepared from perfectly homogeneous, elastic and isotropic materials of known mechanical properties.
6) The matrix is brittle and the stress-strain linearity is maintained up to the failure of the composite.
7) There is no transverse variation of the strains in the components, which are connected in parallel and have the same length in the load direction.
8) The stresses do not vary in the direction of the applied load for the components, which are connected in series and have the same cross sections.

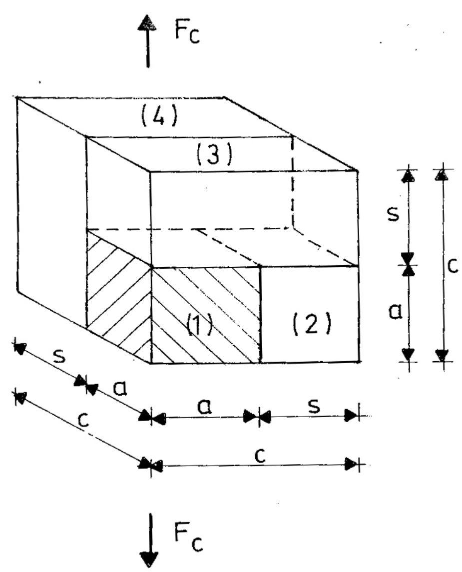

Fig. 1. A schematic representation of the four-components model.

The model presented in fig. (1) consists of four components. The components (1) and (2) are connected in parallel and the resulting element is connected in series with the component (3). All the above components are connected in parallel with the component (4).

As it is shown in fig. (1) the filler volume fraction is given by

$$u_f = a^3 / c^3 \tag{1}$$

For uniaxial load in the direction shown in fig. (1) the equations for the stresses and the strains in the composite are

$$\sigma_c = \sigma_3 u_f^{1/3} + \sigma_4 \left(1 - u_f^{1/3}\right) \tag{2}$$

$$\sigma_3 u_f^{1/3} = \sigma_1 u_f^{2/3} + \sigma_2 \left(u_f^{1/3} - u_f^{2/3} \right) \tag{3}$$

$$\varepsilon_1 = \varepsilon_2 \tag{4}$$

$$\varepsilon_4 = \varepsilon_c \tag{5}$$

$$\varepsilon_c = \varepsilon_1 u_f^{1/3} + \varepsilon_3 \left(1 - u_f^{1/3}\right) \tag{6}$$

where the indexes 1,2,3,4 and c correspond to the components (1), (2), (3), (4) and the composite respectively.

The constitutive equation for each component and the composite are given by Hooke's law;

$$\sigma_1 = \varepsilon_1 \ E_f \tag{7}$$

$$\sigma_2 = \varepsilon_2 \ E_m \tag{8}$$

$$\sigma_3 = \varepsilon_3 \ E_m \tag{9}$$

$$\sigma_4 = \varepsilon_4 \;\; E_m \tag{10}$$

$$\sigma_c = \varepsilon_c \;\; E_c \tag{11}$$

where the indexes m and f correspond to the matrix and the filler respectively. From the above equations it comes out that

$$\sigma_1 > \sigma_3 > \sigma_c > \sigma_4 > \sigma_2 \tag{12}$$

Assuming that failure in the component (3) of the matrix corresponds to failure of the whole composite from the above relations one obtains

$$\sigma_{cu} = \sigma_{mu} \left(1 - \frac{u_f^{2/3} - u_f}{\frac{1}{m-1} + u_f^{1/3}} \right) \tag{13}$$

where σ_{mu} and σ_{cu} are the strengths of the matrix and the composite respectively and $m = {E_f}/{E_m}$.

Similarly the fracture strain is given by

$$\varepsilon_{cu} = \varepsilon_{mu} \left(1 - \frac{u_f}{\frac{u_f^{1/3}}{m-1} + u_f^{2/3}} \right) \tag{14}$$

Alternatively when the values of ε_{cu} and E_c are known the strength can be evaluated by Hooke's law [1].

$$\sigma_{cu} = E_c \cdot \varepsilon_{cu} \tag{15}$$

Finally the elastic modulus of the presented model is given by [7]

$$E_c = E_m\left(1+\frac{u_f}{\frac{1}{m-1}+u_f^{1/3}-u_f^{2/3}}\right) \quad (16)$$

where $m = {E_f}/{E_m}$.

Inherent flaw

In resin / filler particulate systems it has been found that the stress intensity factor, K_{ic} has a linear faction of the filler content u_f [3]. Thus one can write

$$K_{ic}^c = K_{ic}^m + bu_f \quad (17)$$

where the indexes m and c correspond to the matrix and composite respectively and b is a constant depended on the properties of the matrix, the filler and the interphase.

For the evaluation of the "inherent flaw" of the composite the expression

$$K_{ic} = y \cdot \sigma_u a^{1/2} \quad (18)$$

will be used, where y is a geometrical factor, σ_u is the strength and a is the inherent flaw. Writing this equation separately for the matrix and for the composite and introducing the resulting values of K_{ic}^m and K^{ic} in eqs. (17) one obtains

$$\frac{a_c}{a_m} = \left\{\frac{\sigma_{cm}}{\sigma_{cu}}\left(1+\frac{bu_f}{y\sigma_{um}a_m^{1/2}}\right)\right\}^{1/2} \quad (19)$$

Eqs. (19), as $\sigma_{cm} > \sigma_{cu}$ provides that $a_c > a_m$, fact that is explained by the crack pinning effect.

RESULTS AND DISCUSSION

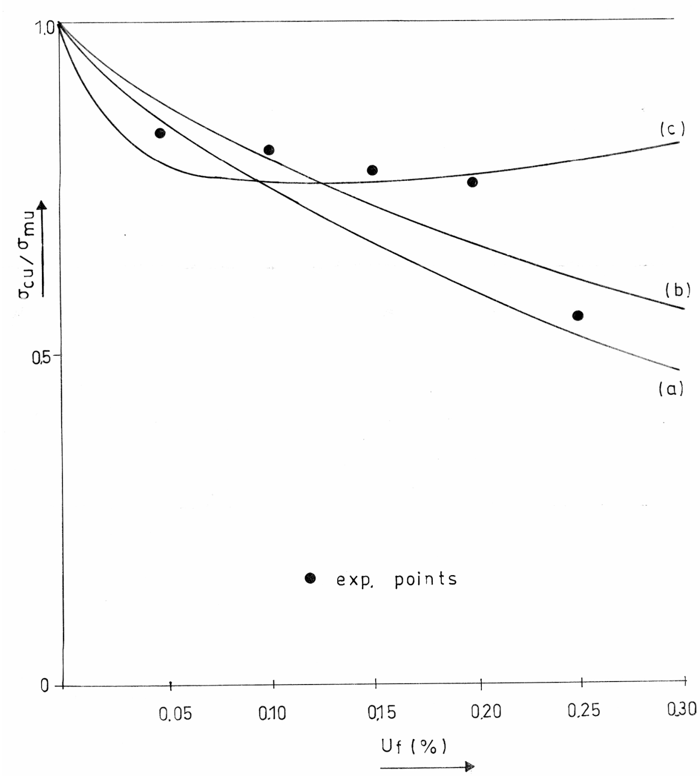

Fig. 2. Ratio σ_{cu}/σ_{mu} (strength of the composite over strength of the matrix.) versus filler volume fraction, u_f, in epoxy / iron particulate composites. (a); Nicolais and Narkis (eqs. A.4), (b); Nielsen (eqs.A.1), (c); Present model, Nielsen, (eqs. A.2 and A.3).

In fig. (2) the strength of epoxy / iron particulate composites is plotted versus the filler content. Curve (c) represents the theoretical values of the strength predicted by the presented model, while curves (a) and (b) represent the strength predicted by existing equations in the literature. The shown experimental values come from tensile experiments carried out in our laboratory. In fig. (2) one can observe that there is a satisfactory agreement between the strength obtained from the present model and from the experiments. The discrepancy that is observed at high filler volume fractions probably is owed to the appearance of agglomerations in the used specimens.

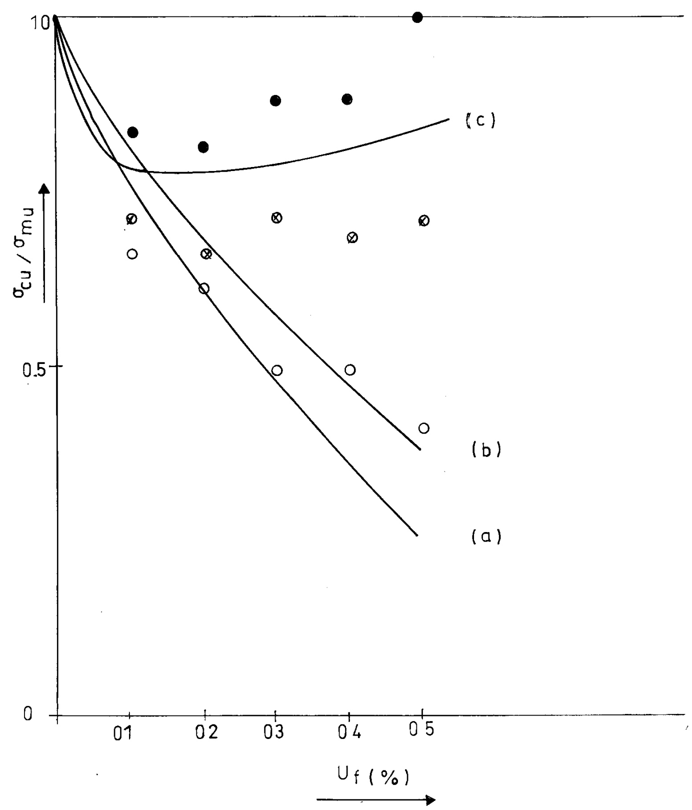

Fig. 3. Ratio σ_{cu}/σ_{mu} as a function of volume fraction of filler, u_f, 4,5 μ_m particles
● treated with A187, ⊗ untreated, O DC1107 treated. (After Spanoudakis and Young).
(a); Nicolais and Narkis, (b); B. Nielsen (eqs.A.1), (c); Present model, Nielsen, (eqs. A.2 and A.3).

In figs. (3) and (4) the strength of epoxy / glass particulate composites is plotted against filler volume fraction. The shown experimental results are taken from ref. [4,5] and correspond to i) treated particles with an improvement of the kind of adhesion between matrix and filler (•) ii) untreated particles (⊗) and iii) treated particles so that there is no adhesion between matrix and filler (O). In figs. (3) and (4) it is observed that the strength predicted by the present model is in agreements with the experimental results of untreated particles shown in fig. (4). Consequently the model is characterized as corresponding to an intermediate adhesion quality, which appears in composites with untreated particles.

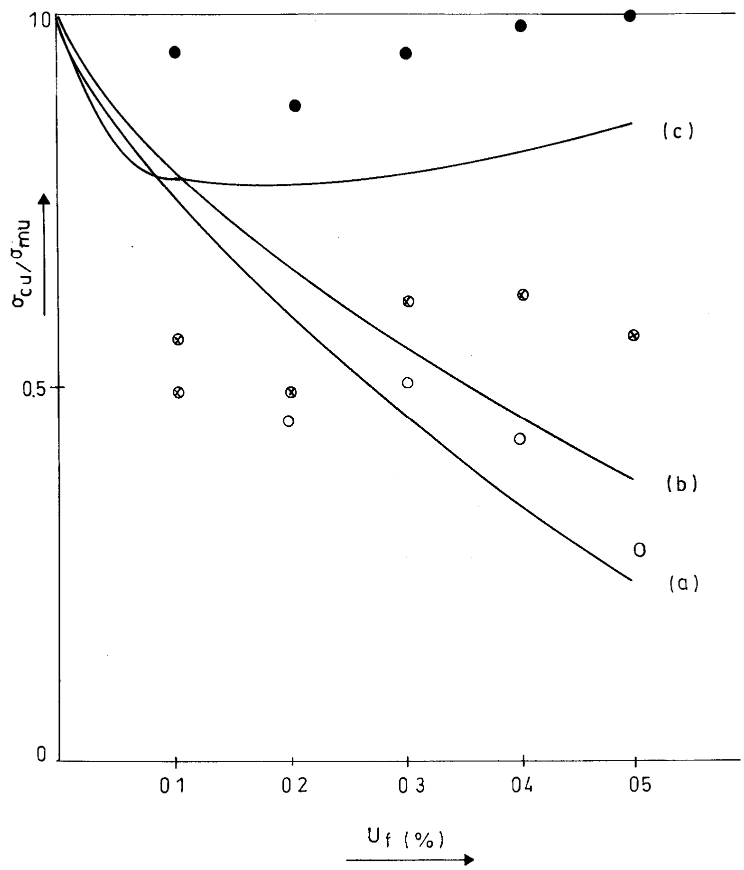

Fig. 4. Ratio σ_{cu}/σ_{mu} as a function of volume fraction of filler, u_f, 62 μ_m particles
● treated with A187, ⊗ untreated, O DC1107 treated. (After Spanoudakis and Young).
(a); Nicolais and Narkis, (b); Nielsen (eqs.A.1), (c); Present model, Nielsen, (eqs. A.2 and A.3).

It is noteworthy that the strength predicted by the presented model is in agreement with the strength predicted by the procedure proposed by Nielsen in which

$$\sigma_{cu} = \varepsilon_{cu} E_{cu}$$

where

$$\varepsilon_{cu} = \varepsilon_{mu}\left(1 - u_f^{1/3}\right) \tag{20}$$

This can be explain because ε_{cu} given by the present model is close to ε_{cu} resulting by the above relation. One can assume that discrepancy from Nielsen procedure appears only when the fracture deformation is different than the fracture deformation evaluated by the above relation $\varepsilon_{cu} = \varepsilon_{mu}\left(1-u_f^{1/3}\right)$.

Strength and Ultrasounds

It is known that the strength is related with the hardness predicted by Brinnel's method. The existing relation is [8]

$$\sigma_{cu} = KBHN_{30} \tag{21}$$

where BHN is Brinnel's hardness and K is a constant of the material. The index 30 corresponds to the relation

$$\lambda = 30 = \frac{p}{D^2} \tag{22}$$

where P is the applying load on the specimen and D is the diameter of the penetration.

It is also known that the hardness can be obtained by means of ultrasonic measurements. The constant K of eqs. (21) can be determinate by this method.

The present study presents a model for the evaluation of the strength, which can be used to comparison with the strength evaluated by ultrasonic measurement and eqs. (21).

CONCLUSIONS

From this study the following conclusions result.

1) The presented model correspond to an intermediate adhesion quality between matrix and filler as the predicted values of the strength are close to intermediate experimental values of untreated particles.
2) The strength evaluated by the presented procedure is in agreement to the strength evaluated by Nielsen's procedure.

3) The equation of the inherent flaw, a_c, of the composite yields $a_c > a_m$. The constant b of this equation must evaluate experimentally.
4) The theoretical values of the strength can be used to compare with the strength evaluated by ultrasounds method.

REFERENCES

1. L. E. NIELSEN, J. Appl. Pol. Sci, Vol. 10, p. 97 (1966).
2. S. AHMED, F. R. JONES, J. Mat. Sci, Vol. 25, p. 4933 919900.
3. A. C. MOLONEY, H. H. KAUSCH, T. KAISER, H. R. BEER, J. Mat. Sci. Vol. 22, p. 381 (1987).
4. J. SPANOUDAKIS and R. J. YOUNG, Ibid. 19, p. 487 (1984).
5. J. SPANOUDAKIS and R. J. YOUNG, J. Mat. Sci, Vol. 19, p. 487 (1984).
6. A. J. KINGUCH, D. L. MAXWELL, B. J. YOUNG, J. Mat. Sci, Vol. 20, p. 4169 (1985).
7. G. BOURKAS, E. SIDERIDIS, V. KYTOPOYLOS and J. PRASSIANAKIS, THE FIFTH NATIONAL N.D.T. CONF. OF THE H.S.N.T., NOVEMBER, 18-19, 2005, NTUA, ATHENS, GREECE.
8. J. N. PRASSIANAKIS, S. K. KOURKOULIS, EXPERIMENTAL STRENGTH OF MATERIALS, EDITOR SYMMETRIA, ATHENS, GREECE, 1999.
9. L. NICOLAIS and M. NARKIS, Ibid. 11, p. 114 (1974)

APPENDIX

Equations used for comparison:

1) Nielsen’s equation (no adhesion) [1]

$$\sigma_{cu} = \sigma_{mu}\left(1 - u_f^{2/3}\right)\cdot \mathrm{K} \qquad \text{(A.1)}$$

where K is a stress concentration factor.

2) Nielsen’s equation (adhesion)

$$\sigma_{cu} = \varepsilon_{mu} \mathrm{E}_c \tag{A.2}$$

where

$$\varepsilon_{cu} = \varepsilon_{mu} \left(1 - u_f^{2/3}\right) \tag{A.3}$$

3) Nicolais and Narkis equation (no adhesion) [8]

$$\sigma_{cu} = \sigma_{mu} \left(1 - 1{,}21 u_f^{2/3}\right) \tag{A.4}$$