BRITTLE MATRIX COMPOSITES
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A Conception of a Structural Theory of Composite Materials

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ABSTRACT

A fundamental step towards the characterisation of the various mechanisms responsible for the deformation and strength behaviour of composite materials is the determination of decisive criteria and parameters and the regions of their applicability.

On the basis of an analysis of the behaviour of various types of composites two fundamental criteria have been defined: the 'structurality' of the system and the inner specific surface. A number of examples are given, in which these criteria are discussed and illustrated.

In accordance with the deformation processes taking place in the system the composites are classified into three basic types. Their similarities and differences are discussed and five boundary systems are defined. This definition lays the foundations of a general structural theory of composite materials. The application of this theory is illustrated on an actual structural model and the quantitative description of deformation behaviour of composites of the second and third types. The model enables the introduction of both the geometrical arrangement of the structure and the interphase interaction including the effect of external environment.

I. INTRODUCTION

We are well aware of the complexity of composite materials, their deformation behaviour and the difficulties in predicting of properties or

their composition to suit required purposes. However, not all of us are equally well aware of the number of diverse mechanisms existing in the formulation and application of composites, their mutual intermingling, their mutual, often synergic, interaction, and the meaning of the individual mechanism for the particular type of composite under investigation. This is probably one of the reasons for the existence of so many different theories and methods for describing the deformation behaviour, strength and other necessary characteristics and also why none of them are generally valid. Even more serious is the fact that only very few of such theories define the scope of their validity by some objective criteria or parameters describing the analysed system. The enormous diversity of the previously mentioned composites necessarily results in the fact that in different systems various deformation and failure mechanisms are of varying significance and importance. What is essential in one system, is negligible in another, what is a determining factor in one system, can be merely a modifying factor in another. For those reasons a first fundamental step in an endeavour to conceive any sort of objective theory will obviously be:

(a) the selection of decisive criteria
(b) the definition of the fields of their validity, and to endeavour, on this synthetic basis, to classify the composites into certain groups or classes and only within these classes make an endeavour to analyse them.

The first question naturally is, whether such decisive criteria can be recognized and the extent of their validity can be defined.

If we analyse the behaviour of various composites, usually differentiated according to the relative representation of phases, without taking into account the geometry or morphology of phases, we can discern quite clearly the influence of superstructural arrangement.

2. EXAMPLES OF TWO SOLID-PHASE COMPOSITES

Let us illustrate the influence of superstructural arrangements by a single example of a composite consisting of two solid phases in the whole possible extent of their representation in the system, with or without the presence of a single fluid phase according to Fig. 1. The first extreme system is a quasihomogeneous system consisting of the first solid phase, the other extreme system is a two-phase non-cohesive system consisting
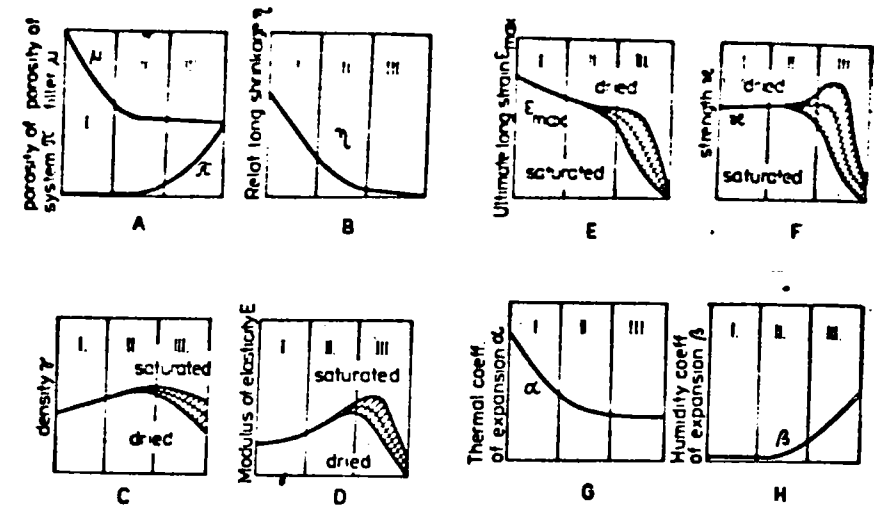
Fig. 1.

only of the second solid phase and the fluid phase. We see that we find not an accidental, but a regularly variable change of all the physical characteristics. The first obvious similarity of the dependence of all these relationships on the volume representation of the individual phases (regardless of their type and geometrical form) lies in the fact that their changes are markedly different in three regions and that these regions are identical for all the observed characteristics.

The first and third regions are characterised by a slow and continuous change of properties. However in the second region, they change abruptly. It is obvious that this change is not due to either the volume representation of phase or to their geometry, but to the structure of the composite, which is, next to the volume representation of phases, also a function of their geometrical and physical parameters and is responsible also for the course of force flows and the mechanisms of microdeterioration, micro-creep, micro-relaxation and/or micro-plasticity.

If we analyse in some detail the example of strength (Fig. 1F) the relationship between strength and change of the superstructure, expressed for the sake of simplicity by the relative weight representation of solid phases according to Fig. 2, we can see that the internal stresses change in relation to the external manifestation of the material—its strength. The inclusion of the dispersed phase in the matrix causes the growth of the internal state of stress from the beginning to the moment, in which the
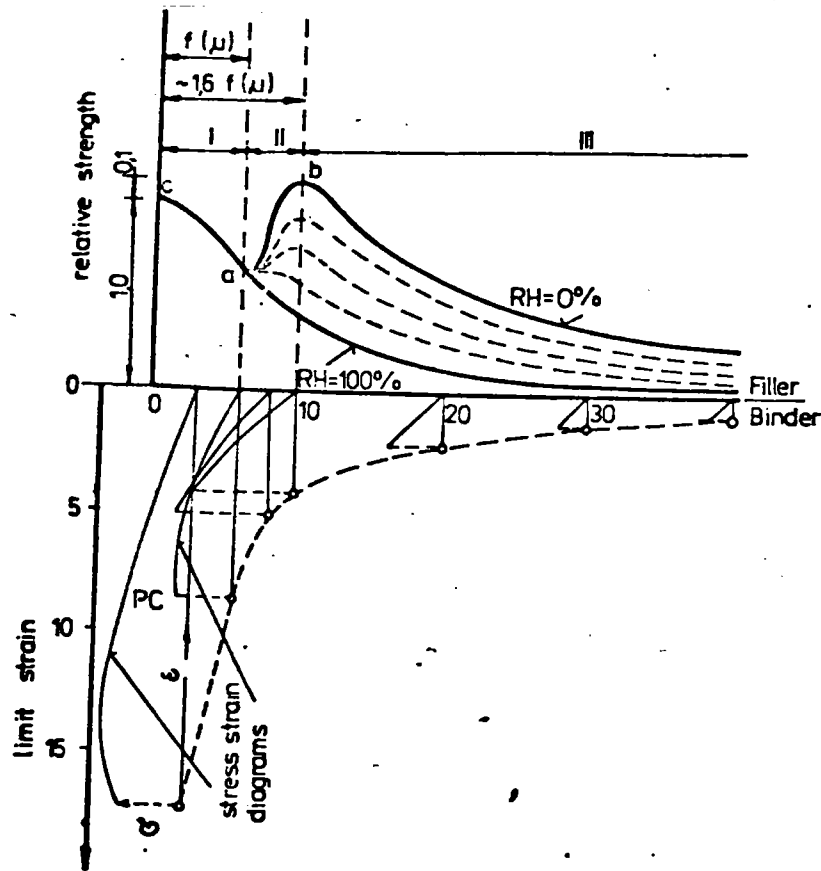
Fig. 2a.

Fig. 2b.

integration of stress concentrations to the inclusions with small mutual spacing, causes failure of the bridges of the matrix or the interphase contacts. The growth of the internal state of stress brings about local creep or plastic deformation of the matrix. The microfailure, on the other hand, results in the release (relaxation of internal stresses) and growth of the reserve of bond energy for the transfer of external global loads.

If the fluid phase is present in the structure from the very beginning, the inner state of stress and the extent of microfailure are higher in the case of a liquid, and lower in the case of a gas.

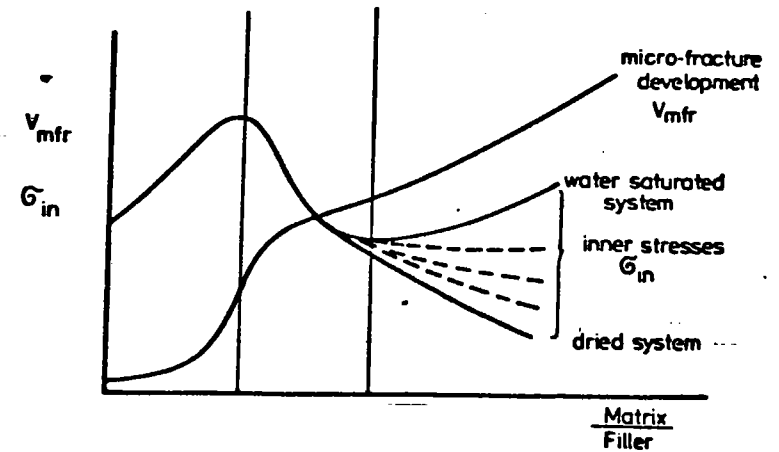In the first region, if the relative quantity of inclusions increases, the extent of microfailures is small. In the second region, when the critical spacing of inclusions has been attained, it grows steeply, and later, in the third region, in which the fluid phase is sufficiently represented, it once again changes very little.

Apart from relaxation due to the origin of microfailures a reduction of the internal state of stress is also contributed to, in the later phase by the presence of the fluid phase, particularly if it is a gaseous one. The individual microformations of the superstructure, whose rigidity is lower, have a greater possibility of movement and are capable of taking over a considerable quantity of energy by their elastic deformation.

These and further mechanisms act simultaneously in the structure, mutually overlapping and even causing synergic effects. It is no wonder, therefore, that experimental results obtained and theories based on them are so different. It would not be reasonable to endeavour to cover all mechanisms in their entirety, as it would make theoretical relationships entirely incomprehensible. It is more sensible to find those regions of superstructural arrangement in which the individual mechanisms are dominant, and not to consider other mechanisms in this region. The results will not be entirely accurate, but can be very characteristic and will prevent any unexpected surprises.

The magnitude of influence of the superstructure on the properties of the composite can be illustrated with another example, shown by Uriev [2]. The microfiller, mixed in current methods with a matrix (resin), will afford a superstructure consisting of coagulated clusters of particles bound

by physical forces. The supply of further energy, e.g. by high-frequency vibration, can achieve the disintegration of this primary superstructure and the creation of a new one, characterised by a regularity of dispersal of the individual particles, structural uniformity. This will reduce, for instance, the effective viscosity of the fresh mix by 7-8 orders, as shown in Fig. 3. As a consequence of the considerably more homogeneous field of inner deformations and stresses, however, also the strength and other characteristics will increase significantly, e.g. toughness after solidification of such a system.
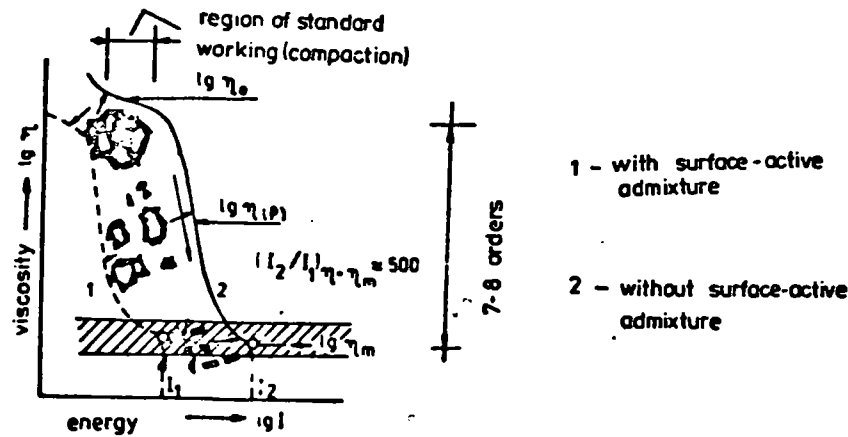

Fig. 3.

region of standard working (compaction)

1 - with surface-active admixture

2 - without surface-active admixture

3. STRUCTURALITY OF THE SYSTEM AND INNER SPECIFIC SURFACE

From what we have shown, the *first* decisive criterion follows—*the structurality of the system* (superstructurality). In the first place, it is decisive whether the dispersed phase is segregated in the dispersing phase (without mutual force contact), or whether it is aggregated (capable of transferring directly a force flow from one particle to another, even across an intermediate layer of the matrix). Another factor is the quantity and type of the fluid phase (gas, liquid) and the degree of its continuity with ambient environment.

Apart from the structurality of the system, the further no less important criterion is the existence of discernible boundaries of phases or the *inner specific surface* which determines the position of the individual regions on the axis of the volume representation of phases, not only by its magnitude, but also by the quality of interface.

In their contacts the individual phases are bound mostly (but not exclusively) by physical links. In the close proximity of dispersed particles, which are tougher than matrix, a change in the matrix structure takes

Fig. 4.

place. There originates a so-called transition layer, which originates even in the case when no chemical reaction takes place. For example, as a consequence of the effect of physical and physico-chemical forces the orientation of the polymer (crystallinity) originates in the proximity of the particles, shown in the micro photograph on Fig. 4, which results in a significant change of its deformation and strength characteristics in comparison with its bulk properties. Micromeasurements have shown [4] for instance, that, the modulus of elasticity varies within two orders depending on the distance from the particle (Fig. 5).
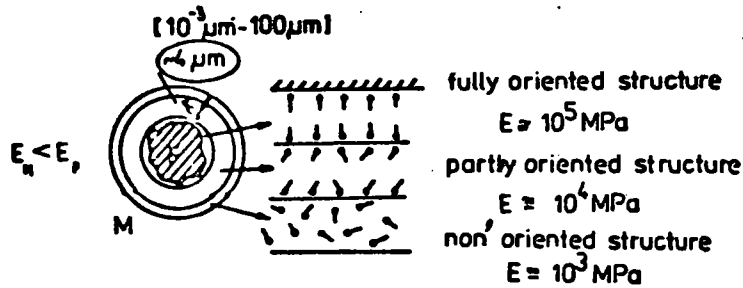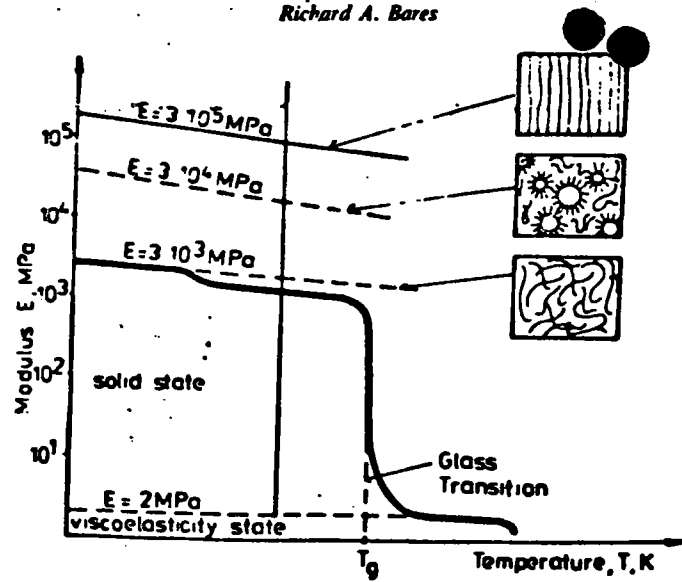
Fig. 5.

4. DEVELOPMENT OF INTERNAL STRESSES

In the formation of a composite high stresses originate at the interface as a consequence of the shrinkage of the matrix in the course of solidification and possible considerable difference in the coefficients of thermal expansion in the course of cooling from the solidification temperature. At the boundaries of the particles, most frequently in the enveloping (modified) layer, but also in the particles themselves, microfailures (microcracks) frequently originate (Fig. 6), which are
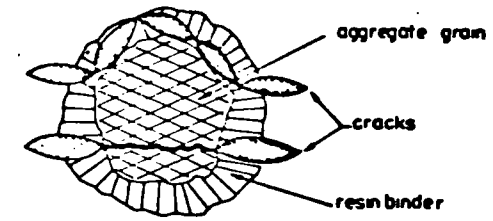
Fig. 6.

responsible for a marked reduction of the strength of the composite as compared with the strength corresponding with the energy of links attained.

On the other hand, if the dispersed particles are less rigid than the matrix (and sufficiently small. i.e. below 1 μm or, sometimes. even below 0.1 μm), their deformation initiate the origin of crazes in the adjoining matrix (Fig. 7), dissipating a considerable quantity of energy. This contributes to an often considerable increase of the toughness of the system.

A combination of both preceding mechanisms can be ensured by the dispersion of rigid particles enveloped by a pliable layer of sufficient thickness to ensure the creation of microcrazes even in a brittle matrix. In this way we approach the so often sought ideal combination of properties: higher rigidity and simultaneously higher strength and toughness than those of the matrix alone (Fig. 8).

If the interphase cohesion is low, microdisplacements along contact surface occur already in the early phases of loading, and the further
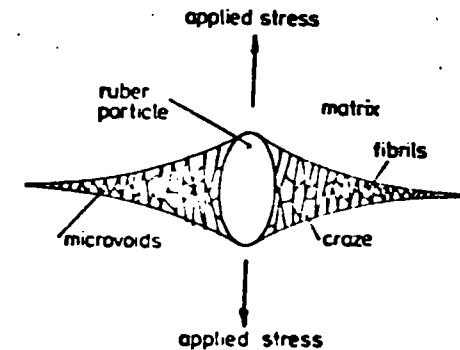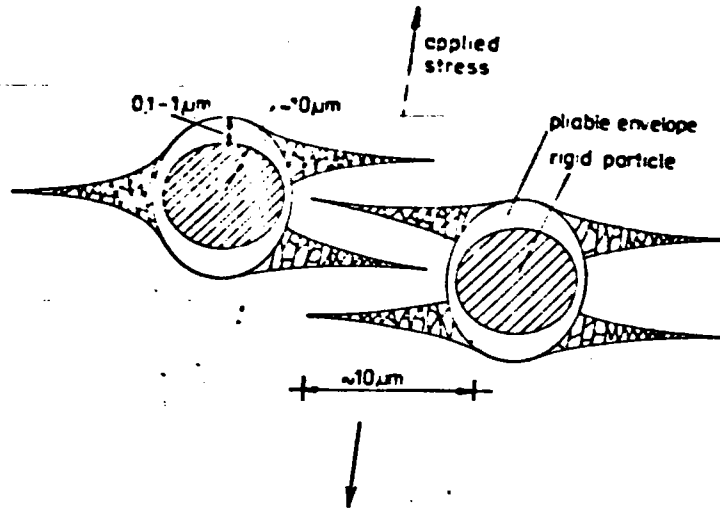

Fig. 7.

coplied
stress

0.1-1μm   .-*0μm

pliable envelope

rigid particle

~10μm

Fig. 8.
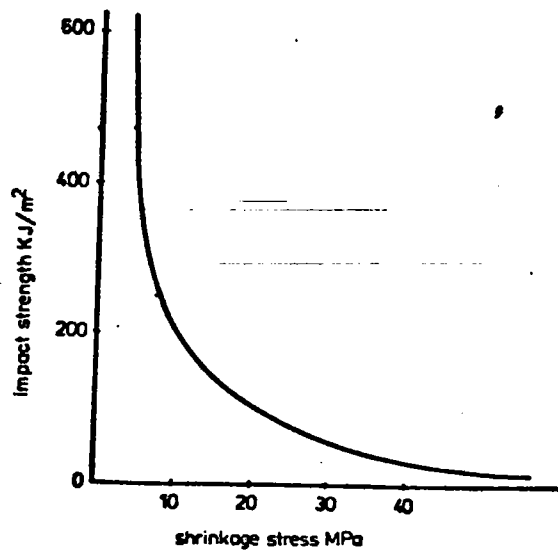

Impact strength KJ/m²

shrinkage stress MPa

Fig. 9.

transmission between phases is possible only by means of friction. If the coefficient of thermal expansion of the dispersed phase is considerably higher than that of the matrix, the danger of their separation and the elimination of friction forces arises. The coefficient of thermal expansion of the dispersed phase. however, must not be too low, as in this case high stresses originate in the matrix, capable of causing cracks in it.

The limits of the differences of coefficients of thermal expansion of both solid phases, in which either excessive transverse or excessive longitudinal stresses arise, capable of causing failure, can be determined from the stress strain equilibrium at the boundaries. These limits are relatively narrow and the mis-match of thermal expansion coefficients between both phases most frequently causes the failure of composites with a brittle matrix, such as reinforced ceramics or concrete.

The reduction of internal stresses due to solidification is of extraordinary importance. For example, in the C E composites the reduction of · shrinkage stresses below the value of 10 MPa increases its toughness exponentially from the initial tens of kJ sq.m to hundreds (200–600) kJ,sq.m, as it is shown in Fig. 9.

5. SUPERPOSITION OF STRUCTURAL AND INNER SURFACE INFLUENCES

The joint influences [8] of structurality and the inner surface can be best demonstrated on a polystructural system, i.e. a system with two or more independent, phase and geometrically continuous structures, the model of which is shown in Fig. 10. A practical example of such a system is the PIC or the C/C/SiC ceramics. If we create, for example, in accordance with Fig. 11, two systems with the same matrix volume in unit volume, once a non-porous PC and the other time a PIPC system by impregnation of the continuously porous PC, we shall introduce into the system in the latter case, apart from two mutually interspersed bearing structures, an additional number of interfaces. The new deformation and failure mechanisms of two mutually independent, chemically identical, but physically, morphologically and crystallographically different infrastructures are capable of absorbing a great quantity of energy. Therefore, the second system has also more advantageous external characteristics, as it is shown in Fig. 12.

In comparison with the system, whose pores have been filled with a liquid, the strength will increase considerably by the filling of the pores
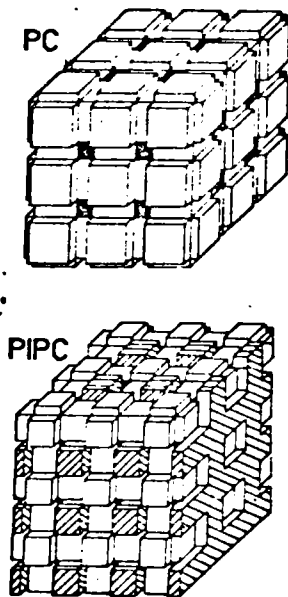
Fig. 10.

by a rigid matrix with continuous infrastructure, and will exceed the values attainable by a single-structure, although even perfectly dried system. Analogous case is represented by the PIC: the various mechanisms by means of which various authors explain the improvement of characteristics in comparison with non-impregnated cément concrete, such as a better cohesion of phases, chemical influencing of primary matrix, the densification of the system, etc., certainly exist, but are not decisive. Of decisive significance are the creation of an independent infrastructure in the pores of primary matrix, geometrically arranged as if along fibres of
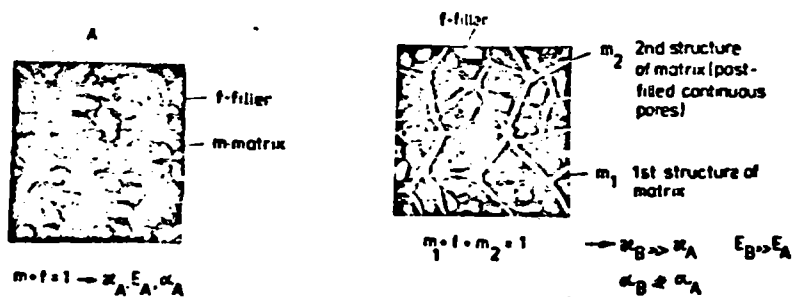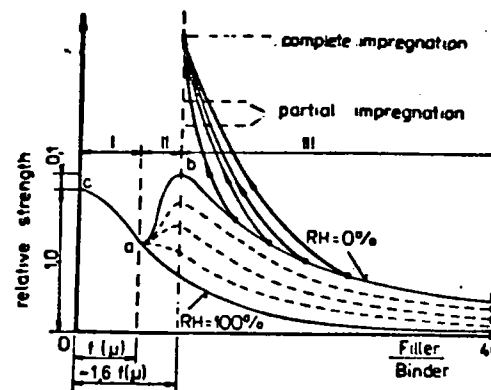

Fig. 11.

Fig. 12.

microscopic dimensions, and a large number of interfaces in which the physical characteristics of the matrix differ considerably from the characteristics in bulk. This changes significantly the character of stress flows, causes a more favourable stress redistribution, localisation and prevention of crack propagation, etc. In short the mechanical interaction of both structures plays the decisive role.

Other experiments have shown the influence of the quantity of the inner surface. We shall show it, once again, on the example of a composite system consisting of two solid phases. The characteristic change of strength, determined by the relations among the interstructural mechanical processes depending on the relative quantities of both solid phases in the system will not change. However, in accordance with the quantity of inner surface, the individual processes will manifest themselves for other phase ratios, as it is shown in Fig. 13.
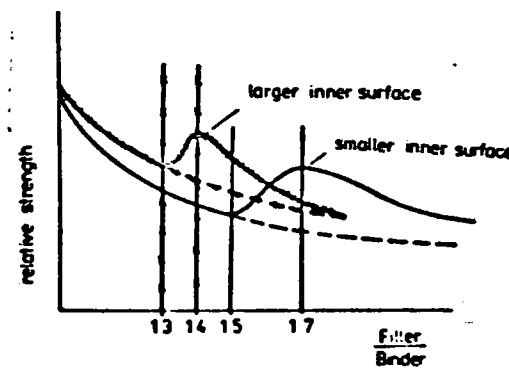

Fig. 13.

6. APPROACH OF THE GENERAL THEORY OF COMPOSITE MATERIALS (GTCM)

On the basis of these studies. illustrated by a few examples. it is possible to outline the basic conception of a general theory of composites, schematically represented in Fig. 14.

Let us analyse here one of the principal quantities which generally describes composite materials. viz. their structure. Apart from the *super-structure* which describes the arrangement of the whole system. there are in a composite more or less regular structures of systems on various levels. as it is shown in Fig. 15. The *infrastructure* is created by the prepared phase in its original arrangement in the composite; every phase
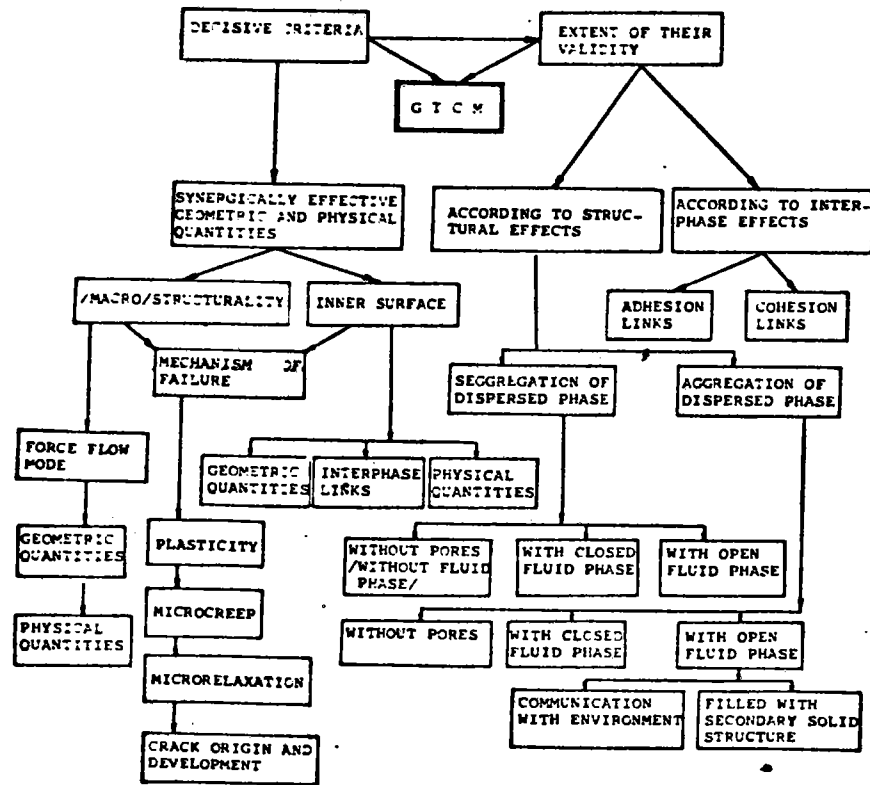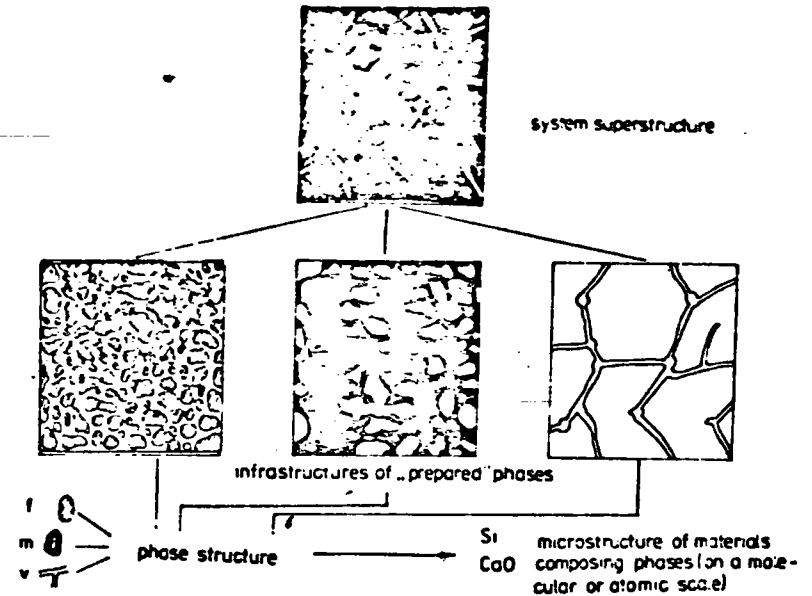

Fig. 14.

Fig. 15.

has its own *structure*. Let us try now to define the individual types of composites in accordance with their superstructure.

For the sake of simplicity let us consider. once again, a system consisting of two solid phases. On the basis of foregoing conclusions it is possible to find several types, differing by the mutual ratio and arrangement of phases. There is a whole number of various systems filling completely the given volume, ranging from pure matrix (i.e. a quasihomogeneous system) as the *first boundary system* (Fig. 16a) as the *second boundary system*. The determining factor of the geometric arrangement in these systems is the *segregation* of particles, the determining component of properties is the matrix. The whole group between the first and the second boundary system can be classified as *Type I Composites*.

It has already been said and we shall accentuate it now that Type I composites fill completely the volume they take up. From the second boundary system of Type I composites onwards it is no longer possible to change the volume of the dispersed phase in the given body (the dispersed phase remains aggregated) and any further change of the ratio of both phases is possible only by a reduction of the matrix, which is replaced in the system by the third phase; the fluid phase (Fig. 16d).

First boundary system

Second boundary system

Composites of the first type

Composites of the second type

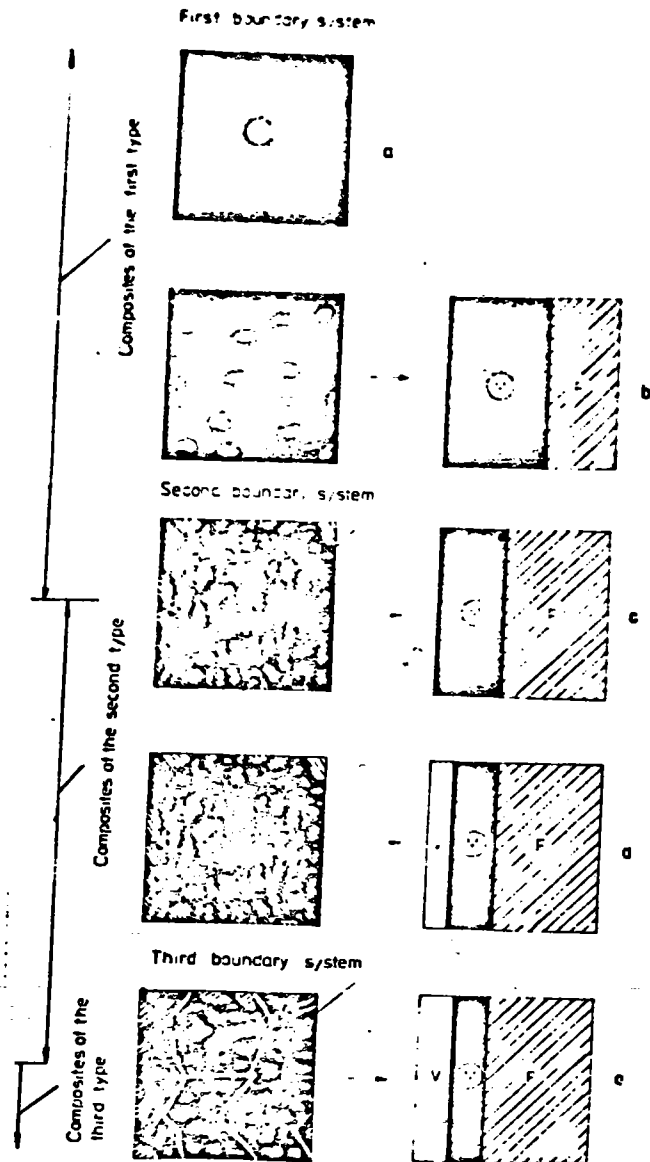Composites of the third type

Third boundary system

Fig. 16.

Closed pores originate in the matrix and the overall ratio of the volume of the solid phase to the volume of the body drops below one and continues to decrease. There originates a system consisting of at least three phases, all characteristics of which will begin to differ diametrally from the preceding systems. The composites of this type form another separate group which we classify as *Type II composites*. Theoretically this region terminates with the system in which the third, fluid phase is continuous and forms a *separate infrastructure*, thus connecting the system continuously and reversibly with ambient environment (Figs. 16e, 17a). Practically, however, this boundary, i.e. the *third boundary system*, cannot be sharp, as the individual pores connect into continuous channels gradually. The solid phase in these systems, as distinct from Type I composites, does not occupy the whole space defined by the material. If the fluid phase in the vacancies of the matrix is gaseous, it is easily compressible, if it is liquid, it is entirely incompressible. The primary internal state of stress of the system (due to the origin of the material), similarly as the secondary internal state of stress (due to external factors) become different with the origin of vacancies, similarly as the dissipation of energy, etc. If the pores are filled with a gaseous phase, the system has a greater possibility of transverse deformation and greater energy dissipation, which results, for example, in high impact and fatigue strengths and longer life. The number of parameters determining the characteristics includes, apart from the factors applicable to Type I composites (ratio of volume and properties of solid phases, magnitude and properties of the specific surface of the dispersed phase), yet other parameters: *the ratio of the fluid and the solid phases and the magnitude and characteristics of the specific surface of the fluid phase*.

As soon as the porosity of the system becomes continuous (without any change of arrangement of the dispersed phase which remains permanently in an aggregated state as at the beginning of the Type II composite group) another quantity (often of decisive character) comes into play, viz. the *composite interaction with ambient environment*. Once again this factor changes decisively the behaviour of the system which is characteristic of Type III composites (Fig. 17b). Another indispensable parameter, without which the description of these systems cannot get along, is, consequently, the *ambient environment*, its characteristics and the way of interaction with the inner surface of the composite. The solid phases occupy an ever decreasing portion of the overall (external) volume of the composite, the matrix decreases until its primary structure distintegrates, loses phase continuity, which is the fourth boundary system
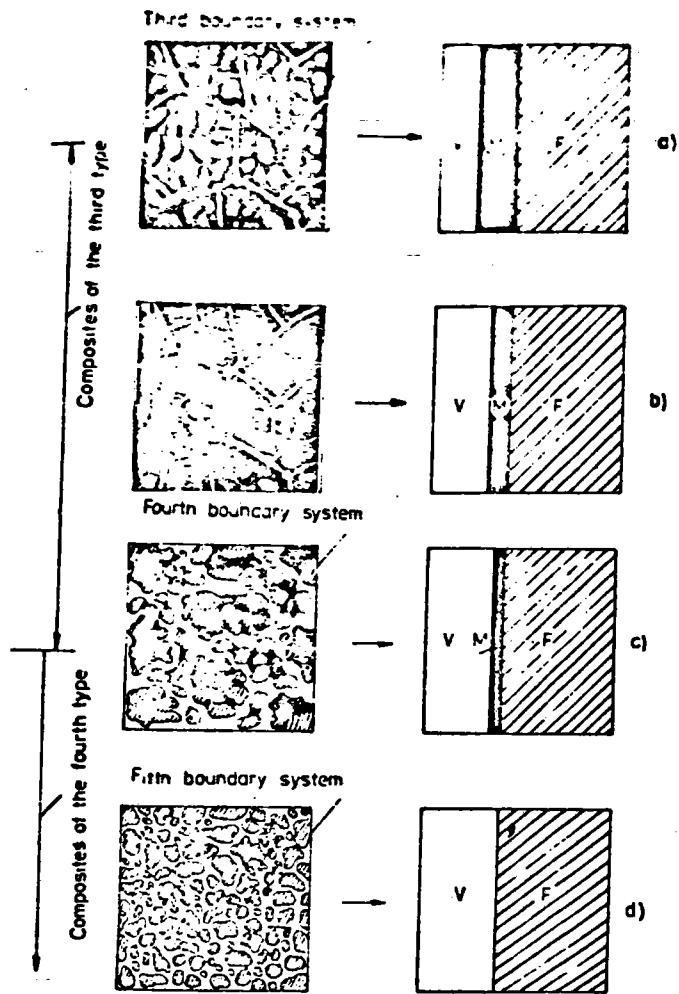
Third boundary system

Fourth boundary system

Fifth boundary system

Composites of the third type

Composites of the fourth type

a)

b)

c)

d)

V M F

V M F

V F

Fig. 17.

(Fig. 17c). The material becomes loose and its properties are determined primarily by its fluid phase (as the secondary dispersing phase). This group could be classified as Type IV composites. The boundary of this group consists in the fifth boundary system in which, the primary dispersing phase (the matrix) is entirely absent, so that the system contains only one solid phase, dispersed in fluid environment (Fig. 17d). Type IV composites are of no significance as structural materials and are encountered in the form of soils in the majority of cases.

7. BASIC CLASSIFICATION OF STRUCTURAL COMPOSITES

Thus we can schematically present the basic classification of structural composites in Fig. 18. Only if their investigation, description and experimental research is broken up in accordance with this classification, we have some hopes to arrive at more unequivocal and chiefly objective, reproducible and uncontradictory results.

Let us note that the physical description of the structure comprises the physical characteristics of every component of the system, their interaction and interphase boundaries. As the physical constants (functions) of a composite are random spatial functions, they can be described only statistically. For this purpose it is necessary to know a complete geometric description of the structure and the physical characteristics of the components, and not only the volume representation of the individual phases.
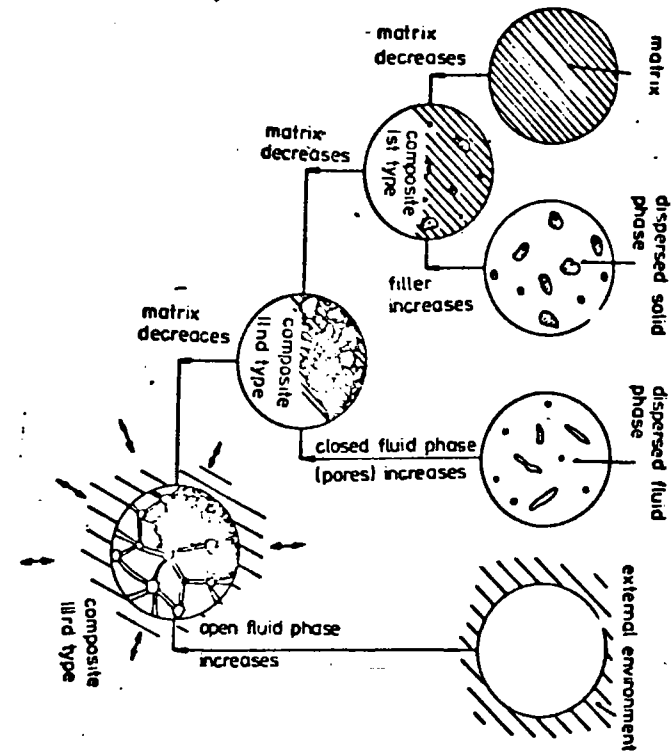

Fig. 18.

8. MECHANISMS OF THE DEFORMATION PROCESS

The mechanical actions which result from the relations of geometric and physical parameters follow in the structure two fundamental mechanisms:

—deformations processes acting as shear flows along interphase boundaries and resulting in the changes of mechanical energy by dissipation, into thermal energy or in mechanical energy used for the formation of new surfaces (cracks, crazes) in the structure. The quota of other types of energy (chemical energy, changes of electromagnetic field) is—with regard to their resulting effect—negligible. These deformation processes are typical of Type I composites;

—deformation processes resulting in the formation of force configurations (force flows) in the structure and, consequently, in the origin of loaded and relieved regions in the structure. The external loads are then resisted by a skeleton of force ways originated in the structure, often of an orthogonal or hexagonal character, and mechanical energy is consumed for the deformation of the skeleton, the deformation of the elements of the skeleton.

These processes are characteristic particularly of Type II and Type III composites. The most significant parameter is the particle size distribution, the particle concentration distribution, the phase continuity and the ratio of physical characteristics of the phases. These deformation processes represent also one of the reasons explaining the remarkable properties of polystructural composites.

We have shown that the introduction of structurality (and, consequently, the inner surface) enables the description of the elastic deformations of the system or its thermal and electrical properties with far greater accuracy and truthfulness than that enabled by mathematical, phenomenological or rheological models.

The introduction of a simple structural model in the form of a system of cubes and prisms as elementary units (Fig.19) made it possible to determine the elastic constants of systems of various compositions, with different relative representation of solid phases, with a different contents of the fluid phase, i.e. composites of all three types, ranging from compact materials to continuous foams. It made it possible to include also the effect of co-operation of the solid and fluid phase at interphase boundaries and the influence of ambient environment on the system. An example of moduli of elasticity and Poisson's coefficients of cement concrete and polymer concrete shows the agreement of experimental and theoretical
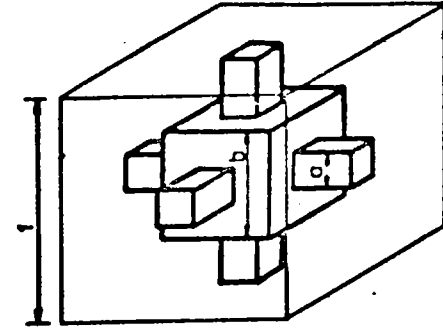
Fig. 19.

values in the whole extent of the relative representation of the individual phases (Figs. 20–22).

The contemporary studies have shown that also the description of strength characteristics following this way is hopeful, although an exhaustive conclusion cannot be made so far in this respect. There are only some conclusions of qualitative character, which still await their transcription into a mathematical form.

In the field of strength a substantial role is played by technology and the occurrence of weak spots in the matrix, the dispersed phase and the phase contact, further by the volume of the body and its surface as the most frequent source of failures. In all places of microscopic as well as macroscopic defects of materials there originate strong concentrations c
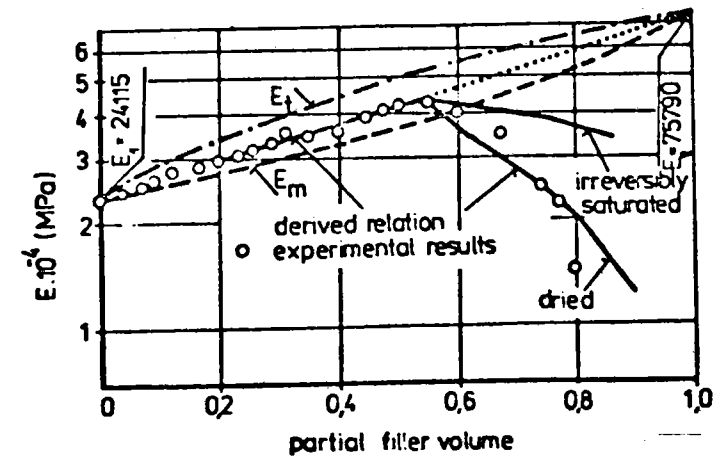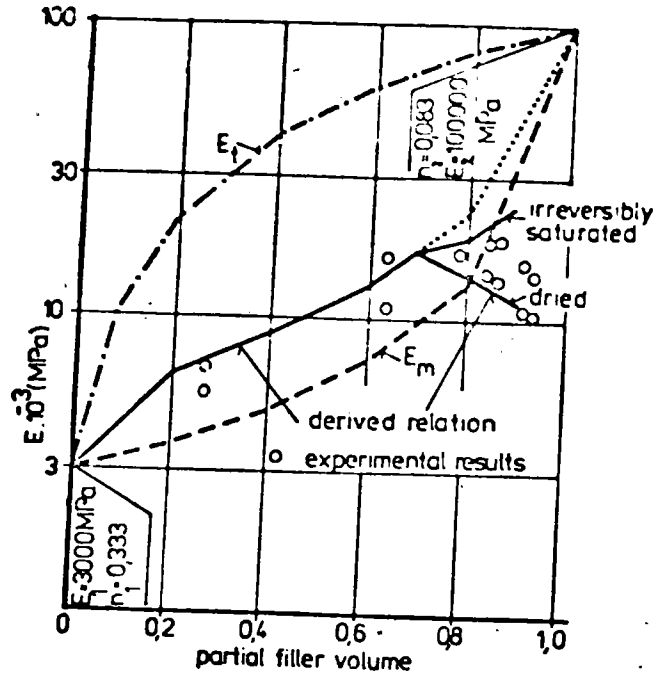

partial filler volume

Fig. 20.

Fig. 21.

Fig. 22.

damages of the structure and the initial points of microcrack development. From the coalescence of pores, defects and plastic deformation concentrations the whole system of microcracks and/or crazes originates, as a rule, after a complex incubation period. Some of these microdamages grow, leaving the place of their origin in the form of small cracks already. Some small cracks are dampened again, but one or several of them pass across the whole cross section of the body as macrocracks and result, in the end, in fracture. The damage develops on various dimensional levels within the scope of at least 4 orders, its velocities fluctuating between merely 10^{-10}cm.s^{-1} in the case of subcritical microcrack and crazes growth and 10^5cm.s^{-1} in the case of brittle fracture in a macrovolume of the body.

A number of sophisticated experimental investigations has shown that the process of deterioration is of a step character. As a rule, it has a random character, depending on the initial defects and weak spots of the structure. Also in composites with a seemingly brittle matrix large non-reversible deformations took place, especially if pliable particles are dispersed. This is due to the origin of crazes and microcracks, accompanied
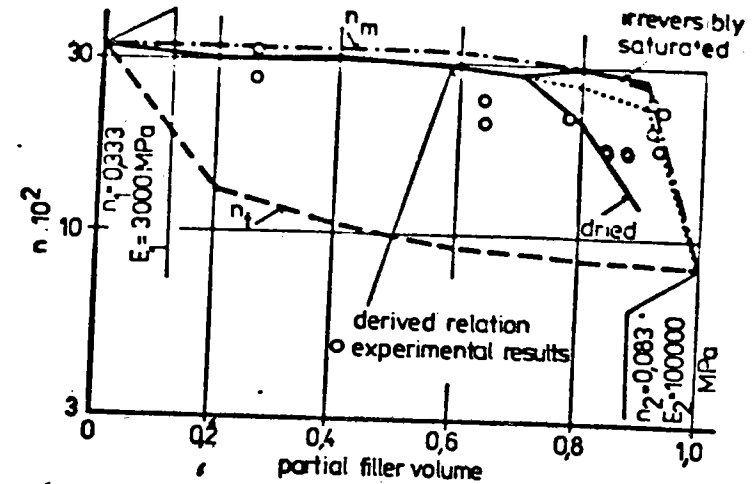
with the softening of the region, stress relaxation and more intensive transfer of energy to its surroundings. The fact that a number of crazes and microcracks originates first, brings about—apart from energy dissipation—also the advantage of origin of a new large inner surface without an excessive prolongation of a single crack which would be connected with a high deformation concentration at its head. In the next phase the crazes and microcracks first increase their density, and only later the bridges between them are torn and the cracks capable of further growth are formed. In every phase of the loading (or life of the material) the mechanisms of propagation of structural failures are different, similarly the barriers and the threshold values of propagation are different.

An objective description of strength, therefore, is exceedingly difficult. It will be necessary to incorporate into the proposed structural model both the principles of modern fracture mechanics, and the Weinbull's statistical conception of strength. The Weinbull's statistical theory will have to be enriched with the influence of interaction factors and based not on the theory of weakest spots, but on the theory of a whole cloud of weak spots, including also the dynamic process of development of weak spots and the effects of failure barriers, i.e. the consideration of principal technological aspects.

However, what we can apply immediately from the aforementioned achievements to the formulation of composite materials is the ensurement of an optimum state of internal state of stress both by technological

means and. primarily. by the formation of an adequate microporous structure (structure with a sufficient quantity of free inner surfaces) by an analogy with the initial state of origin of microcracks under stress. This state corresponds with Type II composites (Fig. 2b). The magnitude of the contribution of the reduction of internal state of stress to strength increase has already been shown by the example of C E composite.

9. CONCLUSION

In conclusion a wish is expressed that the outlined conception of a general theory of composite materials. based on the structural aspect and a synthetic rather than analytical approach. may serve the composite specialists as an effective means to understanding and mastering of a purpose-oriented formulation of composites. This goal is particularly important. as in the future development of mankind. accompanied with growing limitations of material and energy resources. composites will play a role of decisive importance in all branches and industries, as among the available materials they have the highest specific effectiveness.

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Curved Thermal Crack Growth in Self-Stressed Models of Fibre Reinforced Materials with a Brittle Matrix

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ABSTRACT

Combined fracture phenomena in thermally loaded plane models of fibre reinforced materials with relatively low fibre volume fractions have been studied experimentally as well as theoretically. Thereby curved matrix cracks running along special principal stress trajectories as well as interface cracks in the material interfaces of such compound models are considered. Further, the determination of thermal stress states produced in the composite structures by a cooling procedure has been performed by applying the method of photoelasticity: Moreover. finite element calculations have been undertaken in order to determine fracture mechanical data like crack opening displacements and strain energy release rates. respectively. Thereby the latter have been used for the prediction of further crack growth, especially for branched crack systems consisting of a combination of matrix and interface cracks.

1. INTRODUCTION

Investigations of thermal fracture phenomena in nonhomogeneous materials are of significance in the failure analysis of multiphase solids because high-strength composites used in space travel and aircraft technology are often subjected to variable temperature fields. Thereby the arising time-dependent thermal stress fields can cause thermal fracture if the ultimate strength of the corresponding material is reached. Further, the appearance