

PROCEEDINGS OF INTERNATIONAL SYMPOSIUM ON COMPOSITE MATERIALS AND STRUCTURES

June 10–13, 1986 Beijing, China

Edited by:

T. T. LOO &
C. T. SUN

The Chinese Society of Theoretical and Applied Mechanics

American Astronautical Society

European Association for Composite Materials

TECHNOMIC
PUBLISHERS, INC.

General Theory of Composite Materials

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Abstract: If we are to formulate a general theory describing the behaviour of composite materials, it is necessary to base it on the fundamental principles which are responsible for the synergic effects bringing about the very advantages of composite materials. Their number includes, in the first place, the structurality on various levels, the inner surface determining interactions (physical, chemical, etc.) of the structures and/or phases present and the continuity of the material system with ambient environment and the purpose for which it is used. Thus we can define three fundamental types of composites differing by the mutual ratio and arrangement of phases, and five boundary systems.

Introduction

Until recently the composite materials have been considered a simple combination of two or more materials. Actually, however, the composites represent a material system consisting of several phases, of which at least one is a solid phase with macroscopically perceivable boundaries, and which makes it possible to obtain new properties or a combination of properties not attainable by any of its components separately or by their mere sum.

The study of composite materials must be necessarily interdisciplinary. For the description of the deformation and mechanical behaviour and failure of composites a number of methods and theories have already been derived. However, there are certain identical fundamental principles which apply to all composites. There are certain similarities, and a marked progress can be made only by a synthetic theory based on the hitherto disintegrated data from various branches and disciplines of science about various individual materials. The scope of this relatively short paper makes it possible to mention only the fundamental principles of the general theory of composite materials, to emphasize primarily the philosophy of approach, without describing the analytical process.

The existing theories have been derived usually by empiric methods for the individual concrete types of composite materials. If a single substance is changes or if the production process is modified in any way, the theory ceases to function. Further process consisted, as a rule, in the modification of the theory to make the intention correspond with the result, instead of it being the other way about.

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The reason for this unsuccessful procedure is the fact that in the majority of cases no fundamental criteria have been respected, that the substantial has been mixed with the unsubstantial, the determining factors with the modifying factors, particularly that different classes of composites are mixed which originate and operate in different conditions.

The obvious fundamental step in the endeavour to understand the behaviour and consequently obtain the possibility of predicting the characteristics of composites is (Fig. 1)

- a) the selection of decisive criteria
- b) the definition of the fields of their validity

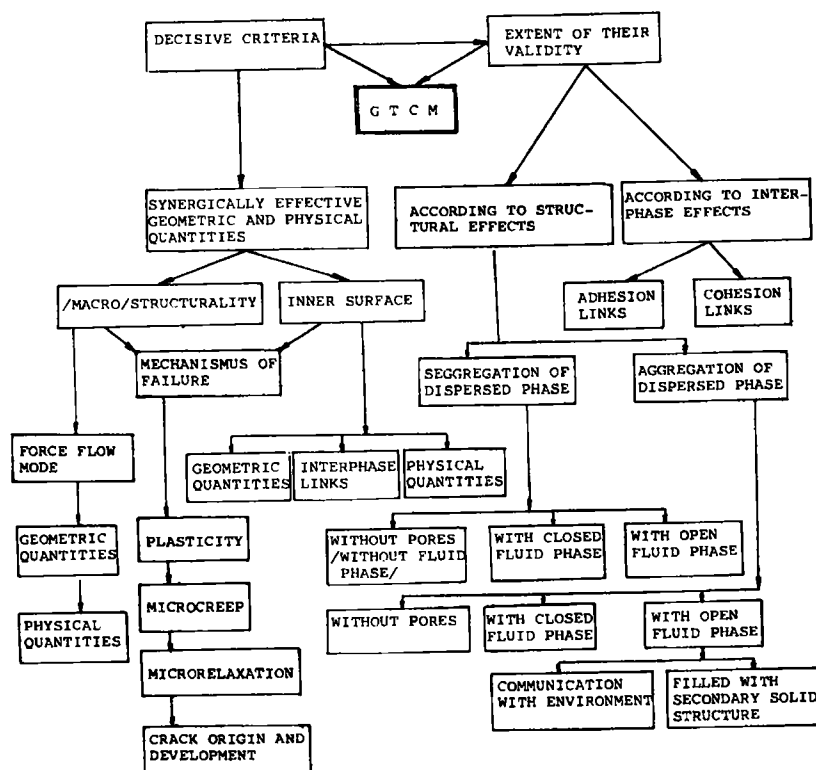


Fig. 1
Fundamental criteria for the description of composites and the field of their validity

Decisive Criteria and Field of Their Validity

If we observe, on an example of a composite consisting of two solid phases with or without the presence of one fluid phase, its properties for various representations of the individual phases (incl. the extremes, i.e. a quasihomogeneous system consisting of a single solid phase on the one hand, and a two-phase non-cohesive system consisting of the other solid phase and the fluid phase on the other hand), we can find not a random, but a regularly variable change of all physical characteristics which is shown in Fig. 2 [1]. The obvious similarity of all these relations in dependence on the representation of the phases in the system lies in that they are markedly different in three regions and that these regions are always the same.

This change is due not only to the representation of the phases or their geometry, but also to the superstructure of the composite which is, naturally, the function of the representation and geometric parameters of the phases as well as of further physical and geometric parameters of the system and its phases.

The magnitude of the influence of the superstructure on the characteristic of the composite can be illustrated by another example (Fig.3). Microfiller mixed by any ordinary method with a matrix (resin) affords a structure consisting in coagulated clusters of particles linked by physical forces. By a

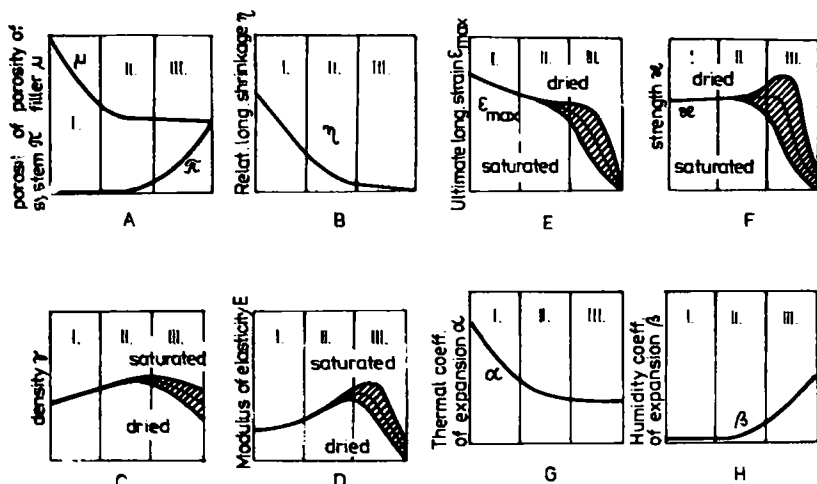


Fig. 2 Change of properties of possible systems consisting of two solid phases with or without the presence of a fluid phase in dependence on the weight ratio of solid phases

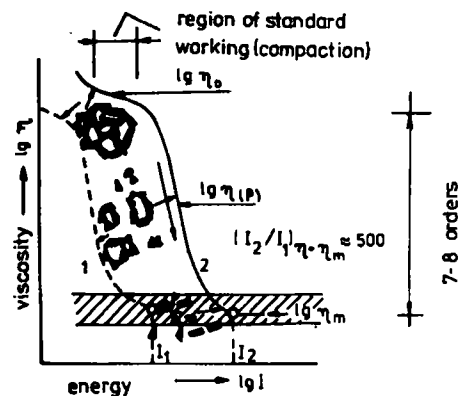


Fig. 3 Structural uniformity achieved by the disintegration of the initial (primary) structure

- 1 - with surface-active admixture
- 2 - without surface-active admixture

more strenuous mixing, e.g. by high-frequency vibration, this primary (initial) structure can be disintegrated and a new structure created, characterized by the homogeneity of dispersal of individual particles - structural uniformity. In this way we reduce, for example, the effective viscosity of the fresh mix by 7 - 8 orders, increase the values of mechanical properties after curing several times, etc. [2].

It would be possible to quote a number of further examples of the decisive influence of structurality. Let us represent them by an example of a polystructural system (Fig. 4), i.e. a system in which there are two or more independent, phase- and geometrically continuous structures, such as it is the case of PIC⁺, impregnated ceramics (e.g. C/C/SIC system), etc. For example we have proved in the PICP⁺⁺ that for a certain quantity of the matrix (polymer) in the system considerably more favourable properties can be obtained, if we create two independent continuous structures of the matrix than if there is only a single structure. This is achieved, for example,

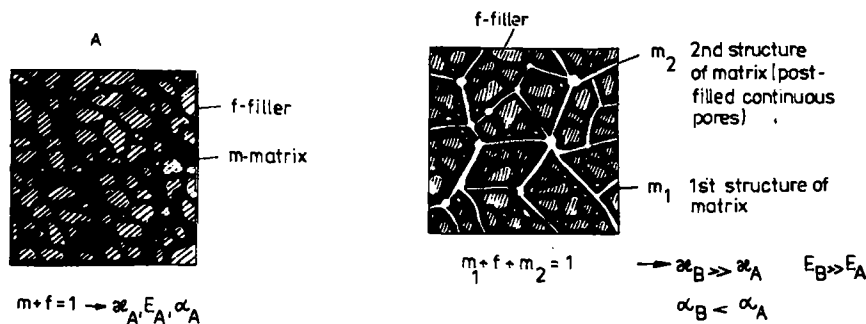


Fig. 4 Comparison of a polymer-impregnated polymer concrete (PICP) with a polymer concrete (PC) with identical matrix quantities in unit volume

⁺) PIC - Polymer Impregnated Concrete
⁺⁺) PICP - Polymer Impregnated Polymer Concrete

by adding - after the formation of the PC⁺ (with shortage of matrix and, consequently, with continuous porosity) - to the system another batch of the matrix (by impregnation) which forms in the pores of the primary system another, independent infrastructure, chemically identical with the initial matrix, but physically, morphologically and crystallographically entirely different from it. The geometric arrangement of the impregnated phase as if in fibres of microscopic dimensions, and the large quantity of interfaces, in which the physical characteristics of the matrix differ considerably from its characteristics in bulk, change significantly the character of stress flows and failure, result in a more favourable stress redistribution, localization and prevention of crack propagation, etc. As it is shown in Fig. 5 the strength, for example, grows by the filling of pores with a rigid matrix in contrast to the filling of the

pores with some liquid considerably above the values attained in the case of a single-structure, even fully dried system.

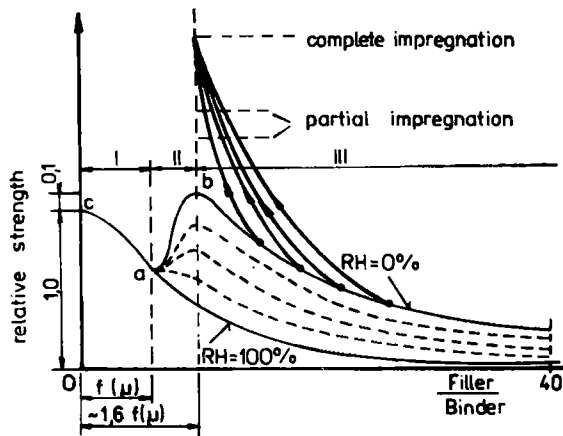


Fig. 5 Change of strength of polymer concrete (PC) after impregnation with the same polymer (PICP) in dependence on the weight ratio of solid phases

From what we have shown there follows explicitly (Fig. 1) the first decisive criterion - the structurality of the system. In the first place, it is decisive whether the dispersed phase is segregated in the dispersing phase (matrix), i.e. without mutual force contact, or whether it is aggregated, i.e. capable of transferring directly a force flow from one particle to another, even through an intermediate layer of the matrix. The further circumstance is the quantity and type of fluid phase (gas, liquid) and the degree of its continuity with the ambient environment.

Apart from the structurality of the system the further, no less important criterion is the existence of discernible phase boundaries or the specific inner surface. The specific inner surface determines the position of the individual regions shown in Fig. 2 on the axis of the volume or weight representation of phases, not only by its magnitude, but also by the quality of the of the phase boundary (Fig. 6). The individual phases are bound in their contacts mostly (but not exclusively) by physical links.

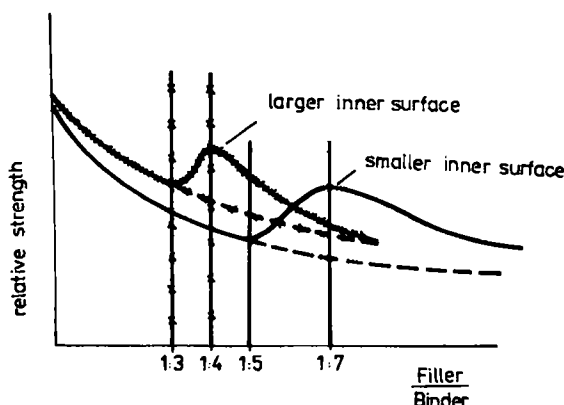


Fig. 6 Influence of the specific inner surface on the change of strength of a PC in dependence on the weight ratio of solid phases

+) PC - Polymer Concrete (concrete with polymer binder only)

In the close proximity of dispersed particles the matrix structure changes and the so-called transition layer is formed even in the case that no chemical reaction takes place. For example, as a result of the effect of physical and physico-chemical forces, the orientation of the polymer (crystallinity) occurs in the proximity of the particles, shown in the microphotograph in Fig.7. These changes in the interphase contact result in significant changes of the deformation and strength characteristics of the polymer shown in Fig. 8. Microm measurements have shown that, for example, the modulus of elasticity varies with the distance from the particle within the scope of two orders [4].



Fig. 7 Formation of a transition layer at phase boundary by the orientation of polymers

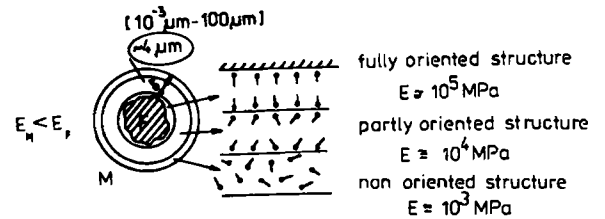
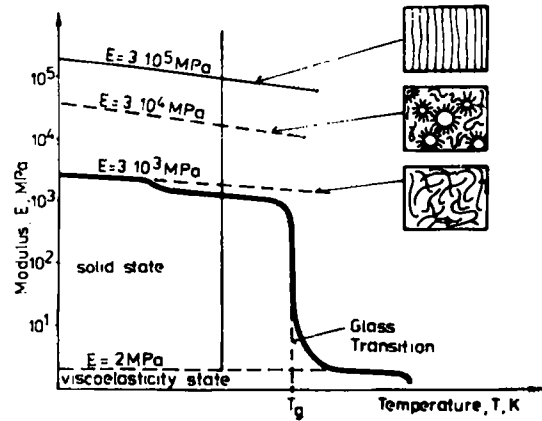


Fig. 8 Properties of a transition layer at phase boundary

During the composite solidification great stresses arise at the interphase boundaries as a result of matrix polymerization shrinkage and, as a rule, of considerably different thermal expansion coefficients of components. At particle boundaries (Fig. 9), most frequently in its enveloping (modified) layer, but also in the particles themselves, microfailures (microcracks or crazes) may arise, which are responsible for the considerably lower strength of the composite than the strength determined by the linkage energy of arising links.

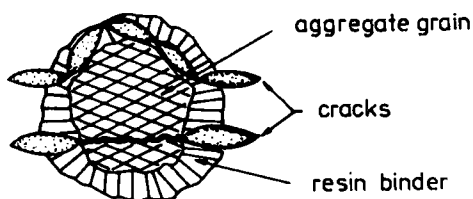


Fig. 9a Origin of structural microdefects in the proximity of dispersed rigid particles (grains, fibres) due to the solidification shrinkage and different coefficients of thermal expansion

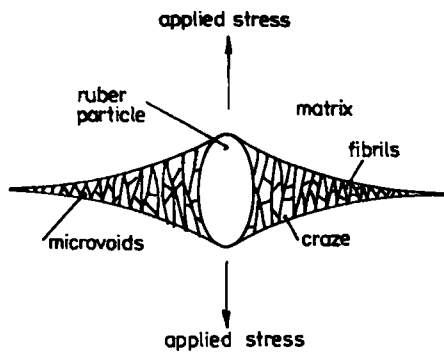


Fig.9b Crazes in the matrix
In the vicinity of
pliable dispersed
particles

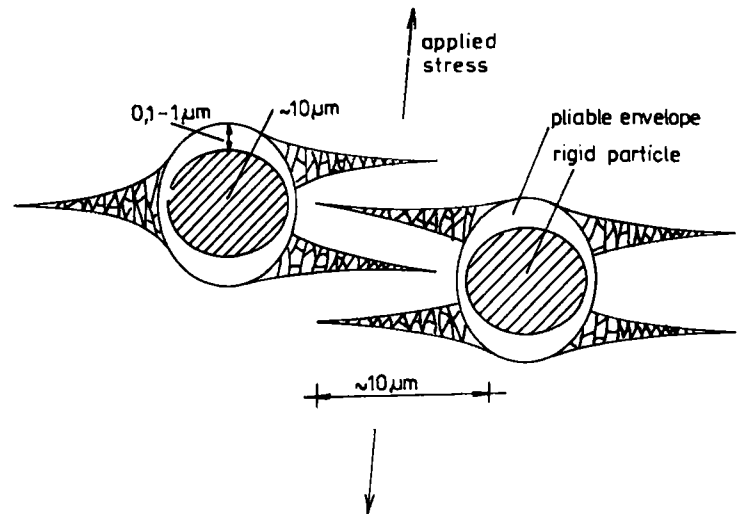
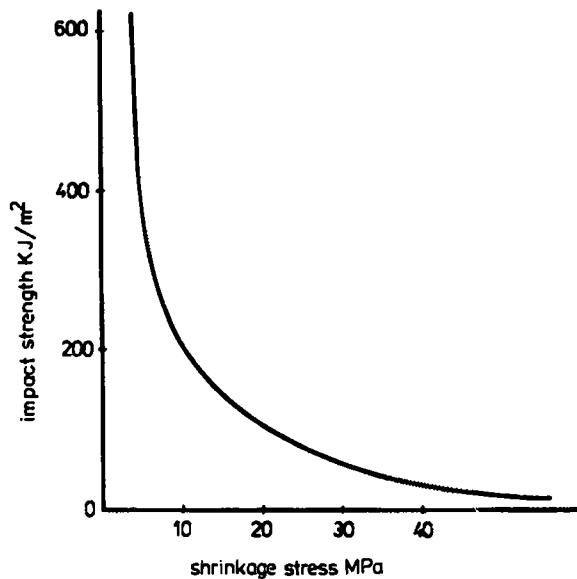


Fig.9c Microcrazes in the matrix in the vicinity of rigid dispersed particles enveloped with a pliable layer

Recent experiments have confirmed that a reduction of the inner stresses due to polymerization shrinkage can result in an enormous improvement of the properties of the composite; for example, in the case of a C/E composite the toughness grows with the shrinkage stress reduction below 10 MPa exponentially from the initial tens of kJ/sq.m to hundreds (200 - 600) kJ/sq.m., as it is shown in Fig. 10.



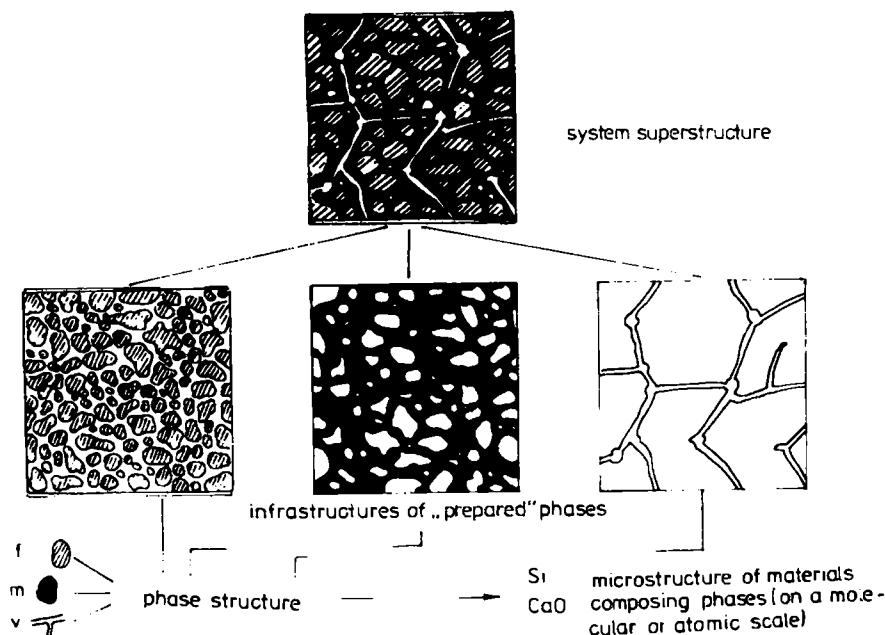
Together with the superstructure (sometimes called also macrostructure or suprastructure), expressing the arrangement of the whole system, there exist in the composite more or less regularly also the structures of systems on different levels (Fig. 11). Every phase has its own structure. The prepared phases in their original arrangement in the composite form its infrastructure. Every substance present in the system, of which the phases consist, has its own microstructure.

Fig. 10 Influence of the inner state of stress due to solidification shrinkage and cooling from the solidification temperature on impact strength of a C/E composite

Composite Definition According to Their Structure

Let us consider, for the sake of simplicity, once again a system consisting of two solid phases. With reference to the conclusions justified in the preceding text, it is possible to find several types of composites, differing in the mutual ratio and arrangement of phases.

There is a whole number of systems, fully filling the given space, from pure matrix (i.e. a quasihomogeneous system) (Fig.12a) as the first boundary system, over dispersed particles (Fig. 12b) to the system with dispersed phase in the densest possible arrangement with aggregated particles (Fig.12c)



as the second boundary system. The determining criterion of geometric arrangement in these systems is the segregation of particles, the determining component being the matrix. The whole group between the first and the second boundary systems is classified as Type I composites.

Fig. 11
System structure on different levels

From the final (boundary) type I composite 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 volumes of both phases is possible only at the cost of matrix reduction, which is replaced in the system by the third phase, viz. the fluid phase (Fig. 12d). Closed pores arise in the matrix and the overall ratio of the solid phase volume to the body volume drops below one and diminishes. Thus a system of at least three phases originates, whose all properties begin to differ diametrically from the preceding systems. The composites of this type form another separate group, classified as Type II composites. Theoretically this region terminates with a system in which the third, fluid phase becomes continuous and forms a separate infrastructure, thus connecting the system continuously and reversibly with the ambient environment (Fig. 12e). Practically, however, this boundary, i.e. the third boundary system, cannot be sharp, as the connection of the individual closed pores into continuous channels proceeds gradually. In contradistinction to Type I composites the solid phase in these systems does not occupy the whole space defined by the material. If the fluid phase in the vacancies is gaseous, it is easily compressible, if it is a liquid, it is entirely non-compressible. The primary inner state of stress (due to the origin of the material), similarly as the secondary inner state of stress (due to external effects) become different with the origin of vacancies, similarly as energy dissipation, etc. If the pores are filled with a gaseous phase, the system has a greater possibility of transverse deformations, prevention of the development of microfailures, and dissipation of a greater amount of energy, which results, for example, in an increase of impact and fatigue strength and longer life. The number of parameters determining properties includes, apart from the factors applied in the case of Type I composites (i.e. the ratio of volume and characteristics of solid phases, the magnitude and characteristics of the specific surface of the dispersed phase) further parameters: ratio of the solid and the fluid phases and the magnitude and properties of the specific surface of the fluid phase.

As soon as the porosity of the system becomes continuous (without changing the arrangement of the dispersed phase, which remains permanently in aggregated state like at the beginning of Type II composites) a further (often

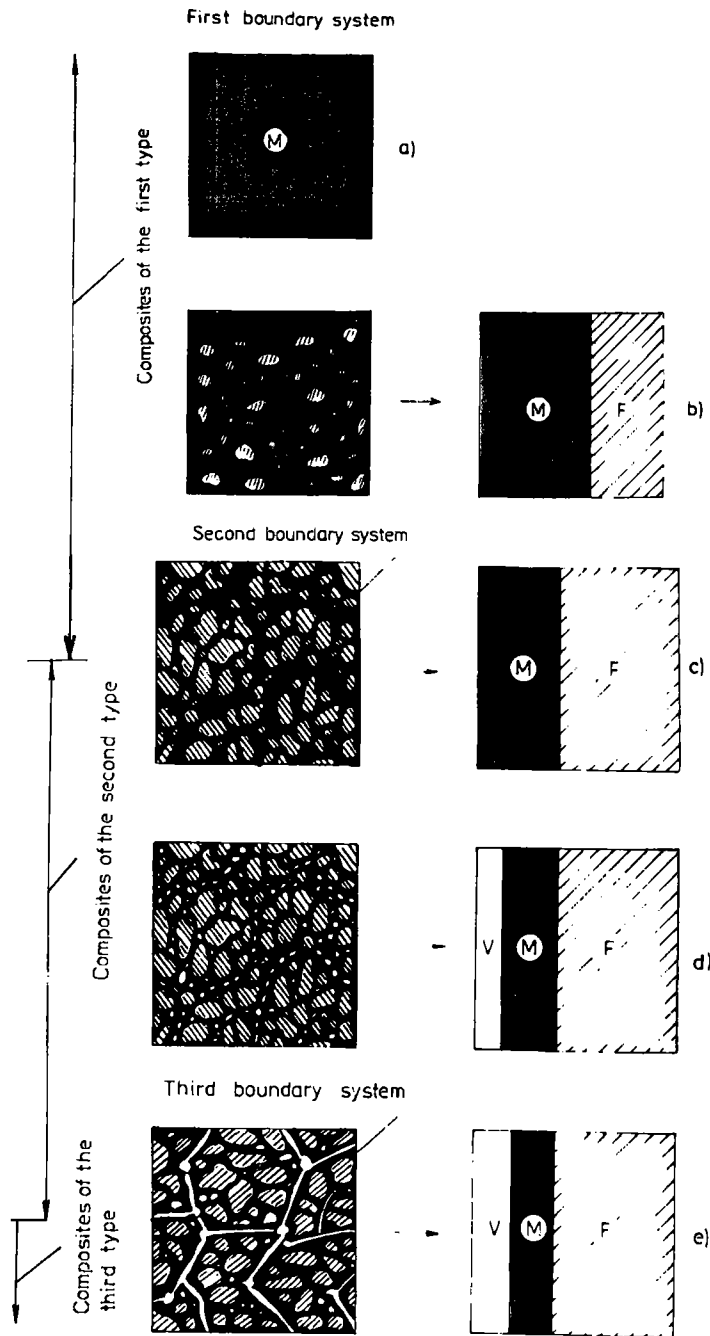


Fig. 12 Composite systems filling differently the given volume and their classification into fundamental types according to their (macro)structural arrangement

decisive) quantity makes itself felt, viz. the inner interaction of the composite with its ambient environment (Fig. 13a). Once again, this quantity changes decisively the behaviour of the system into that which is characteristic of Type III composites. Another necessary parameter without which the description of these systems cannot get along is, consequently, the ambient environment, its characteristics and way of its interaction with the inner surface of the composite. The solid phase occupies an ever decreasing portion of the overall (outer) volume of the composite, the matrix decreases until the initial primary structure of the matrix disintegrates, loses phase continuity (which represents the fourth boundary system). The material becomes loose (Fig. 13b) and its properties are determined primarily by the fluid phase (as the boundary dispersing phase). This group is classified as Type IV composites. The boundary of this system is the fifth boundary system (Fig. 13c), in which the primary dispersing phase (matrix) is entirely absent, so that the system comprises only one solid phase dispersed in a fluid environment. Type IV composites are of no significance as structural materials and we encounter them mostly as soils.

In accordance with structural arrangement we can thus represent the classification of structural composites schematically and with great simplification (as a basis of the general theory of composite materials) as it is shown in Fig. 14. Only if their investigations, description and experimental research are classified in accordance with the system, we have some hope of

arriving at more explicit and chiefly objective, reproducible and uncontradictory results.

Let us note yet that the physical description of the structure comprises the physical characteristics of every component of the system, their interaction and phase boundaries. Since the physical constants (functions) of a

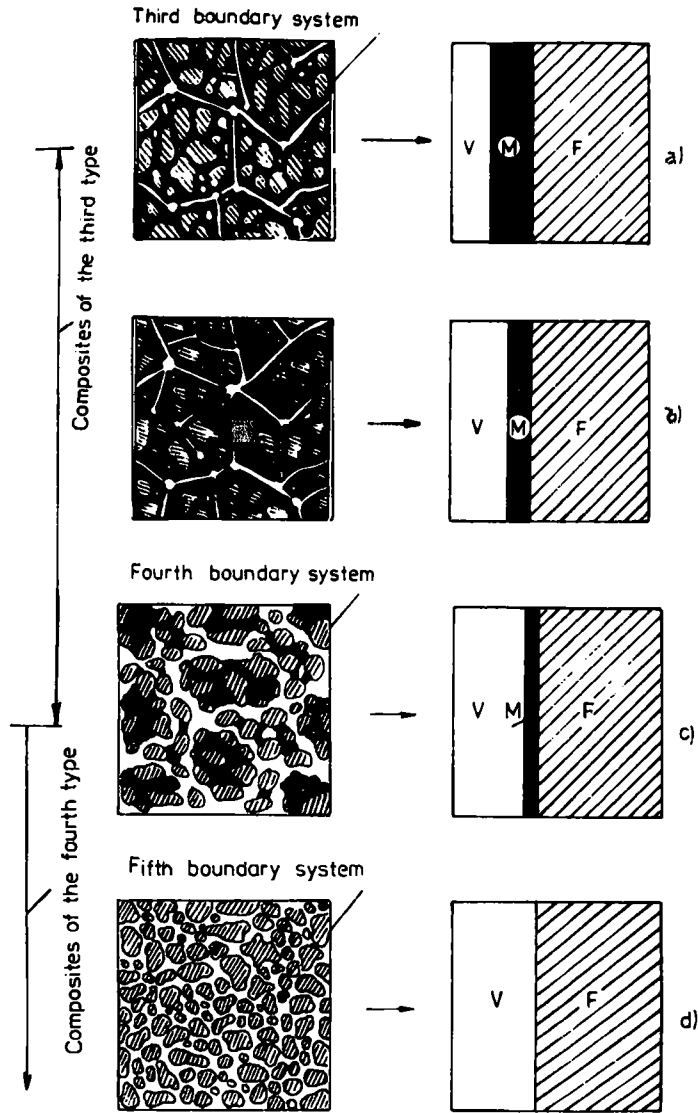


Fig. 13 Composite systems filling differently the given volume and their classification into fundamental types according to their (macro)structural arrangement

composite are random spatial functions, they can be described only statistically. For this purpose, however, 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 phases.

Mechanical processes which result from the relations of geometric and physical parameters, proceed in the structure in accordance with two fundamental mechanisms

- deformation processes acting as shear flows along phase boundaries and resulting in the changes of mechanical energy by its dissipation into thermal energy or mechanical energy lost in the formation of new surfaces (cracks) in the structure. The quota of other types of energy (chemical energy, changes of electric field, etc.) is negligible with regard to the resulting effect. 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 of the structure.

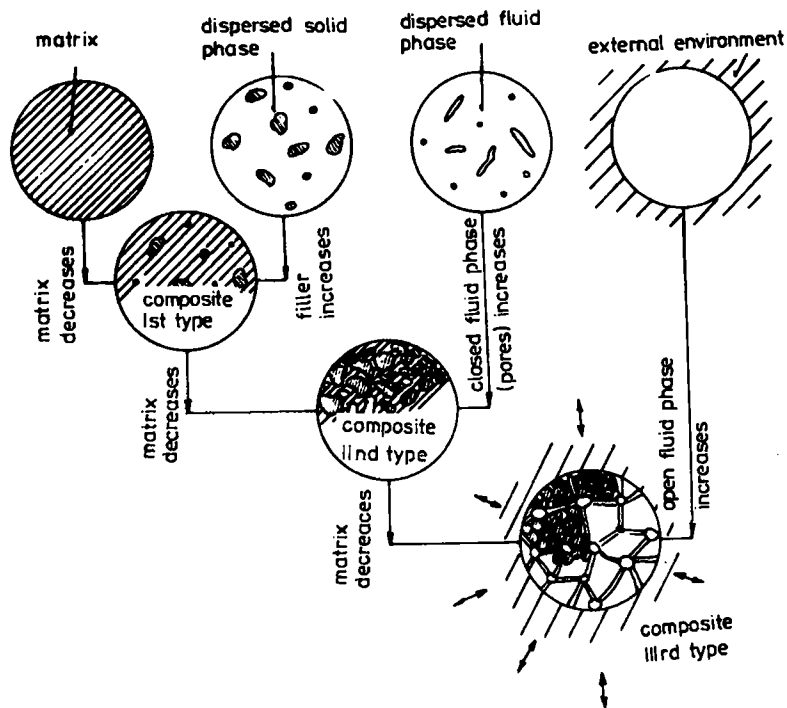
External loads are resisted by a skeleton of force routes originating in the structure, often of orthogonal or hexagonal character. This process is significant particularly for Type II and Type III composites. The

most important parameter is the size distribution, concentration distribution, phase continuity and the ratio of physical characteristics of the phases.

The circumstance that both deformation processes take place in polystructural composites is also one of the reasons explaining the remarkable characteristics of such systems.

Elastic Properties of Composites

On the basis of the briefly described fundamental conception of the general theory of composites some relations have been already derived characterizing truthfully and realistically the elasticity of these systems [3]. Apart from interaction of the characteristics of the individual phases and the intensity of their representation in the process (as it is customary in practically all so-called blending rules) also the structurality of the system



with a certain inner surface and its interaction with ambient environment have been introduced. This affords the possibility of respecting also the fluid phase of the system not only by its volume and modulus of elasticity, but also by the magnitude of its inner surface with which it is connected.

Fig. 14

Schematic classification of composites according to their (macro)structural arrangement

Conclusion

It is the author's wish that the outlined conception of the general theory of composite materials, based on a structural aspect and a synthetic rather than analytical approach, may serve the composite specialists as an effective means of understanding and mastering a purpose-oriented formulation of composites. The author considers this goal particularly important, as in the future development of mankind, accompanied with the growing limitations of material and energy resources, the composites will play a role of decisive importance in all branches and industries, since they have the highest specific (energy) efficiency of all existing materials.

Bibliography

- [1] Bares, R.A., et al., Practical application of synthetic constructive materials as a result of exact definition of material properties Symposium on Recherche et reception des materiaux de synthese utilises dans la construction, University of Liege, 1965
- [2] Uriev, N.B., Dubinin, I.C., Colloidal cement mortars (in Russian), Leningrad, 1980
- [3] Bares, R.A., Relation between geometric and physical structure and properties of granular composites, Journal of Materials Science, 20(1984), 471-481
- [4] Czarnecki, L., et al., Some Polish investigation on properties and applications of composite PC materials. In: Advance in Composite Materials, Paris, 1980, 1310-1316