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THE OUTLINE OF THE GENERAL THEORY OF COMPOSITES

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Abstract

The fundamental step towards the characterization of the varied 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 is given, on 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 made possible the quantitative description of deformation behaviour of composites introducing both the geometrical arrangement of the structure and the interphase interaction including the effect of external environment.

In the qualitative region the problems connected with deterioration and strength are further discussed.

1. Introduction

Though the title of this conference implicates an idea of civil, machine or other structures per se it includes in the reality even mechanics of microstructures in the present-day conception of mechanics of materials. Every material, namely composite one, is a complex structure in which deformation and strength relationships in the main control similar laws being common in mechanics of structures although affected by plenty active, often even synergical influences /physical, chemical, physico-chemical/.

In the short space of the present article are mentioned only the basic principles and namely the philosophy of the approach of the general theory of composites /1/, understood as structural system composed of several phases /at least one of which is solid with microscopically distinguishable boundary/; such a system makes it possible to obtain new properties or a combination of properties which are not obtainable by any component separately or by simple addition of them.

Several methods and theories have already been derived, mostly in the empiric or model basis, for description of the deformation and mechanical behaviour and failure, but they usually apply only to single or limited number of systems.

That way doesn't lead obviously to a goal in a consequence of

a lot of combinations of structural, geometrical, component and interbound arrangement. On the other hand, it is not possible to overlook that for different types of composites the same basic principles, there is a defined similarity, defined basic criteria which all can be identified by a detailed "synthetic" study of the already analysed data. Difficult but necessary it is to separate fundamental and non-fundamental, critical and modifying aspect, and to define decisive parameters for single groups or classes of composites. To find and recognize those basic classes is after them the first and fundamental task of any theory should it be working in sufficient extent.

Clearly a basic step in an attempt to understand the behaviour of composites and hence to be able to predict their properties is /fig. 1/

- a/ classification of the critical criteria
- b/ classification of the areas of their validity.

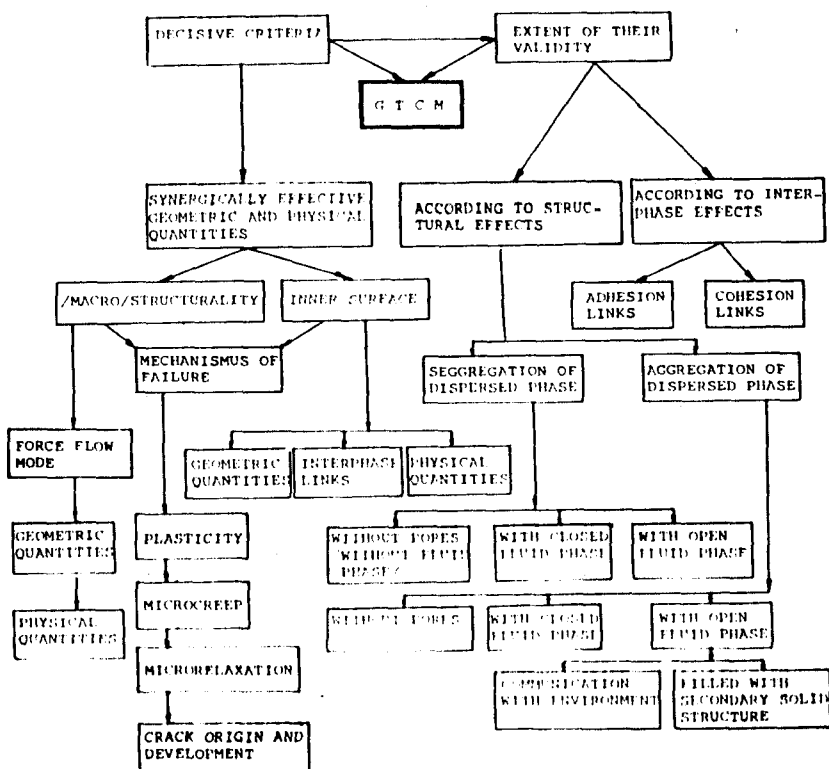


Fig. 1 Basic criteria for the description of compositions, and the areas where they are valid

2. Critical criteria and the areas of their validity

If, using a simple example of a composite consisting of two solid phases with or without the presence of a liquid phase, we study the properties of the composite with different phase compositions /including the extremes, i.e. on the one hand a quasihomogeneous system consisting of one solid phase, and on the other hand a two-phase non-coherent system consisting only of the second solid phase and a liquid phase/, we can find a non-random, systematic change in all physical in-

dices, as shown in Fig. 2 /2/. The evident similarity of all these dependences on the phase composition of the system lies in the fact that they are markedly different in three regions and that these regions are always the same.

The reasons for this change is not only the phase composition nor only the phase geometry but also the superstructure of the composite, which is, of course, a function of the phase composition and geometry, but is also a function of other physical and geometrical parameters of the system and of the phases.

Let us show by another example /Fig. 3/ how great an influence the superstructure has on the properties of a composite. A fine particle filler mixed using normal methods, with a binder /resin/ will give a structure formed by aggregations of coagulated particles bound by physical forces. By more energetic mixing. e.g. by high frequency vibration, it is possible to achieve a breakdown of this primary structure and the formation

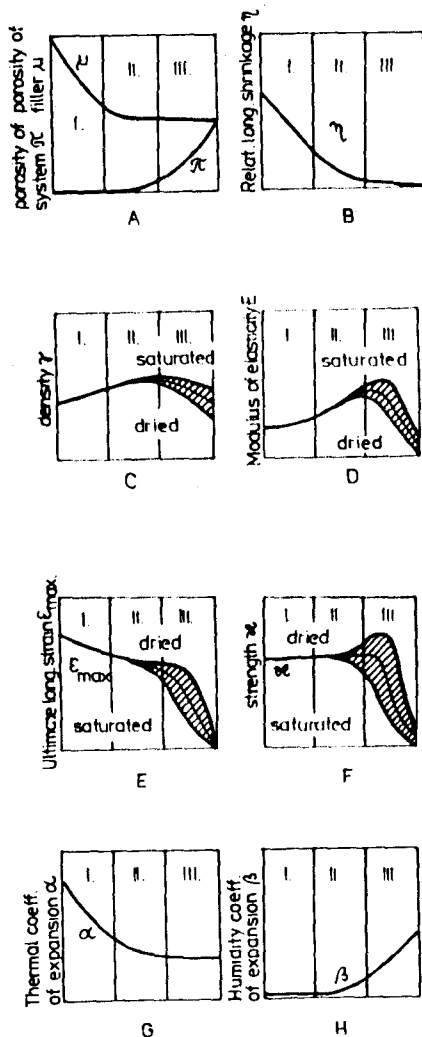


Fig. 2 The change in the properties of possible systems comprising two solid phases with or without a fluid phase, in dependence on volume representation of phases in unit volume

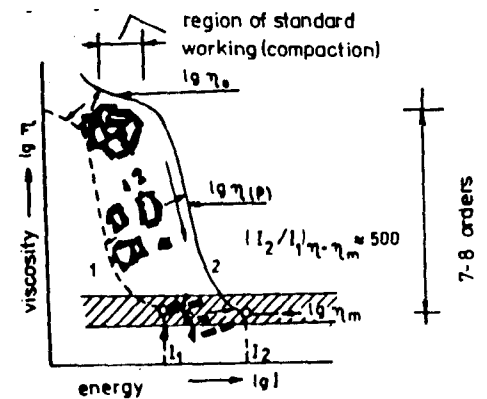


Fig.3 Structural uniformity achieved by breakdown of the primary structure
 1. with lubricant
 2. without a lubricant

of a new structure, exhibiting uniform dispersion of individual particles, also structural uniformity. In this way we can, for example, reduce the true viscosity of a fresh mix by 7-8 orders, increase its mechanical properties after curing several-fold, etc. /3/.

It would be possible to give several other examples of the decisive influence of the degrees of structure.

They are all examples of a multi-structure system /Fig. 4/, i.e. a system in which there are two or more independent structures extending through the phases and the geometry, as is the case with PIC /Polymer Impregnated Concrete/, impregnated ceramics /e.g. the C/C/SiC system/, etc.

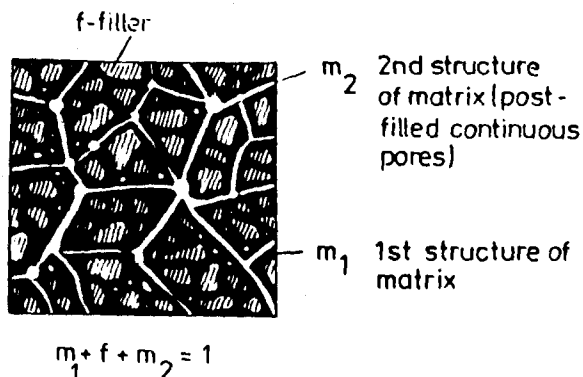


Fig. 4 Bistructural composite system

chemically similar to the original binder but physically, morphologically and crystallographically different. The geometric arrangement of the impregnated phase, as if in microscopic fibres, and the large amount of interfaces, near which the physical properties of the binder differ markedly from the properties in the mass, appreciably alter the nature of the stress flows and breakdown, cause more advantageous stress redistribution, localisation and hindered growth of faults /cracks/, etc. As Fig. 5 shows; for example, the strength of the material when the pores are filled with solid binder /as opposed to filling with a liquid/ increases above the values achieved with a single structure system, even when completely dried.

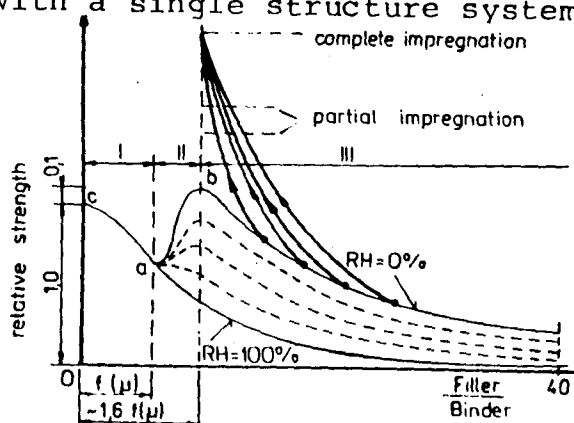


Fig. 5 Change in strength of PC after impregnation with the same polymer /PIPC/, as a function of the mass ratio between the solid phases

We have shown, for example, in the case of PIPC /Polymer Impregnated Polymer Concrete/ that with a certain amount of binder /polymer/ in the system much more advantageous properties are obtained if two independent continuous binder structures are formed than if only one is formed; this will be achieved for example, if, after the formation of PC /Polymer Concrete/ with a deficiency of binder and hence with continuous porosity, more binder is added to the system /by impregnation/, the latter forms in the pores an independent infrastructure,

The above discussion clearly reveals the first critical criterion - the degree of structure of the system /Fig. 1 A/. It is particularly critical whether the dispersed phase in the continuous phase /the binder/ is segregated /without mutual force contact/ or is aggregated /capable of directly transferring a force from one particle to another, possibly through an intermediate layer of binder/. Another consideration is the amount and type of fluid phase /gas, liquid/ and the degree of its continuity with the external medium /Fig. 1 B/.

Besides the degree of structure of the system, a no less important criterion is the existence of detectable phase boundaries, or an internal surface /Fig. 1 A/. The internal surface determines

the position of the individual regions, shown in Fig. 2, on the axis for the phase composition /by volume or mass/, not only by its magnitude but also by the quality of the boundary /Fig. 6/. The individual phases in contacts are bound predominantly /but not exclusively/ by physical bonds.

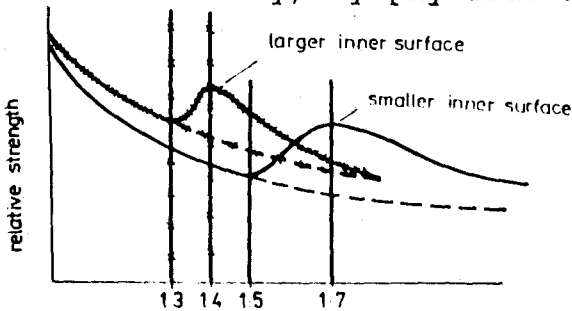


Fig. 6 Influence of the internal surface on the change in the strength of PC, as a function of the mass ratio between the solid phases

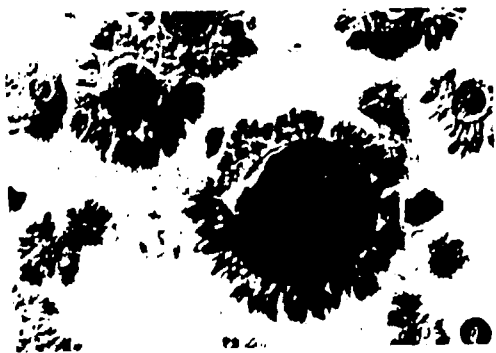


Fig. 7 Formation of a transition layer on the phase boundary by orientation of polymers

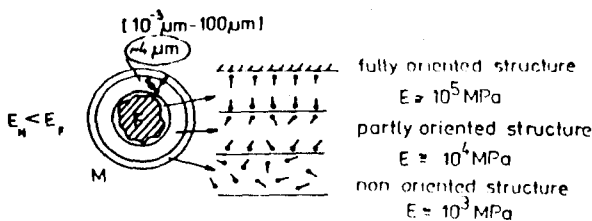
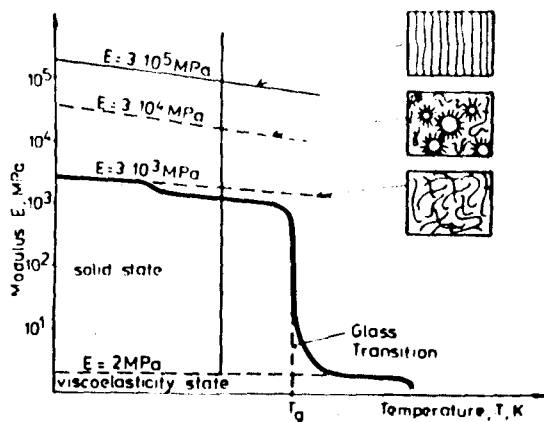


Fig. 8 Properties of the transition layer on the phase boundary

In the direct proximity of the dispersed phase the structure of the binder changes as a "transition layer", is formed, as also in the case when no chemical reaction occurs. For example, under the action of physical and physicochemical forces, orientation of the polymer occurs /crystallinity/ in the proximity of the particles /as the photomicrograph 7 shows/. These changes on the phase boundary then result in a marked change in the deformation and strength properties of the polymer /see Fig. 8/. Micromerements have shown that, for example, the elastic modulus at different distances from the particle varies over two orders /4/.

In the formation of composites, during the transition to the solid state large stresses occur on the phase boundary as a result of shrinkage of the binder usually large difference between the coefficients of thermal expansion of the components. At the borders of the particle /Fig. 9/, most often in the envelope /modified/ layer but also in the particles, microfaults can occur /microcracks or crazing/, which are responsible for composites having much lower strength given by the energy of the bonds formed.

Some experiments have confirmed that by reducing the internal stress due to shrinkage during polymerization on the properties of a composite can be greatly improved, e.g. in the case of a composite composed of carbon fibres and an epoxy, with reduction in the shrinkage stress to below 10 MPa the impact strength increases exponentially, from initial values of tens of kJ/m² to hundreds /200-600 kJ/m²/ as Fig. 10 shows.

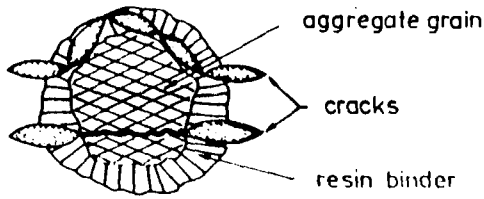


Fig. 9 Formation of micro-defects in the structure in the proximity of dispersed particles /grains, fibres/ under the influence of shrinkage during the transition to the solid stage and of the difference in the coefficients of thermal expansion

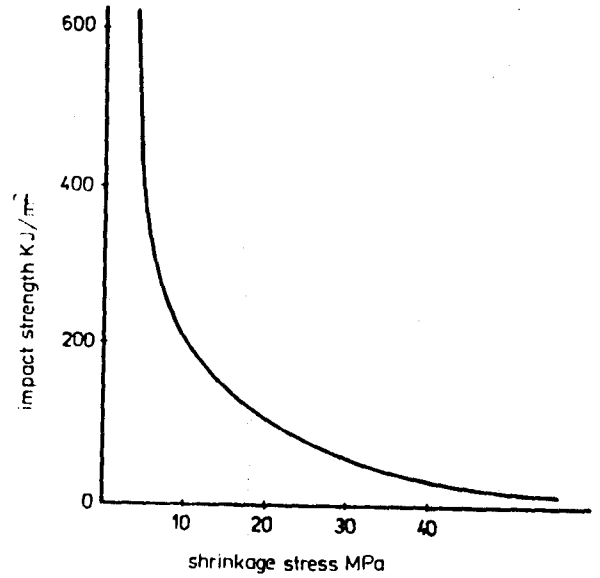


Fig. 10 Influence of internal stress due to shrinkage and during the transition to the solid state and cooling from the solidification temperature on the impact strength of a composite consisting of carbon fibres and on epoxy resin

Together with the superstructure /sometimes also called the macro-structure/, which expresses the arrangement of the system as a whole, a composite contains a more or less regular structure of systems on different levels /Fig. 11/: each phase has its own structure. The infrastructure is formed by the prepared phases in

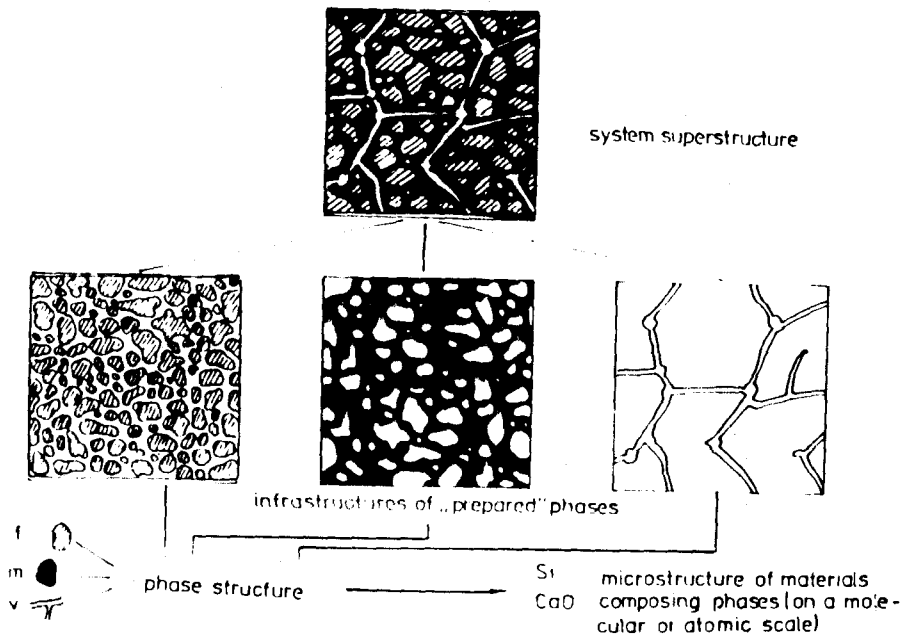


Fig. 11 Structure of systems of various levels

their original arrangement in the composite. In the system of which the phases are composed, each substance has its own microstructure.

3. Definition of composites on the basis of their structure

For simplicity, let us for the moment consider a system comprising two solid phases. Then, taking into account the previous conclusions, we can find several types differing in the ratio and arrangement of the phases.

There are several different systems which completely fill a given space /Fig. 12/, ranging from a pure binder /i.e. a quasi-homogeneous system/ as the first limiting system via dispersed particles, to a system with the dispersed phase in the densest possible arrangement, /with aggregated particles/ the second limiting system; the critical aspect of the geometrical arrangement in these systems is the segregation of the particles, and the central component of the properties is the binder. The whole of this group, between the first and second limiting system, will be termed "Type I composites". The parameters which determine their properties are the volumetric ration and properties of the solid phases, the magnitude and properties of the specific surface of the dispersed phase.

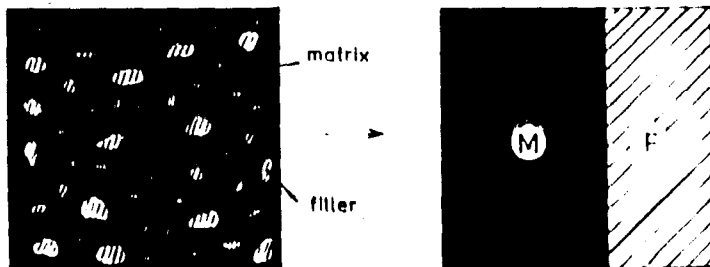


Fig. 12 Composite material of first type

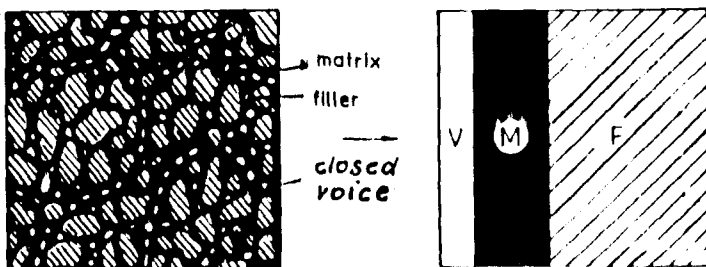


Fig. 13 Composite material of the second type

From the final /boundary/ type I composite onwards it is already impossible to change the volume of the dispersed phase /the dispersed phase remains aggregated/, and further change in the volumetric ratio between the two phases is only possible by reduction of the binder in place of which a third fluid phase is introduced into the system /Fig. 13/. Closed pores are formed in the binder and the overall ratio of the volume of solid phase to the volume of the test specimen falls below unity and continues to fall. A system is formed which is at least three phase, all of whose properties will start to differ diametrically from the preceding system. Composites of this type form another independent group, which we shall term "Type II composites". Theoretically this region finishes with a system in which a third, liquid phase is introduced and forms an independent infrastructure so that the system is continually and reversibly connected to

the external medium. In practice, of course, this boundary, i.e. the third limiting system, cannot be precise, since the combining of individual closed cells into continuous channels is gradual. The solid phase in these systems, by contrast to type I, does not occupy the whole space delineated by the material. If the fluid phase in the free spaces in the binder is gaseous it is easily compressible, if it is liquid it is completely incompressible. The primary internal stress in the system /resulting from the formation of the material/, just like the secondary internal stress /resulting from external influences/, changes when free spaces are formed, the energy dissipation is different etc. If the pores are filled with gaseous phase, the system has greater capacity for transverse deformation, prevent the development of micro-defects and dissipate larger amounts of energy, those give an increase in impact strength, fatigue strength and service life. In the parameters which determine the properties, in addition to the factors which apply to type I composites, other parameters appear: the ration between the fluid and solid 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 any change in the arrangement of the dispersed phase, which remains in the aggregated state as at the start of type II composites/ a further value /often critical/ becomes relevant: internal interaction of the composite with the external medium. This again decisively alters the behaviour of the system, which is characteristic of Type III composites /Fig. 14/. Another essential parameter for describing

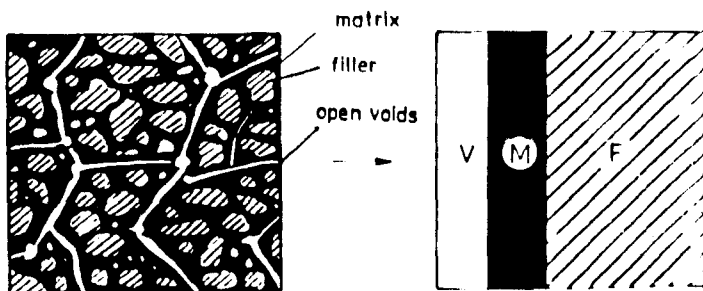


Fig. 14 Composite material of third type

the fluid phase /as the secondary dispersing phase/. We shall term the group "Type IV composites". The boundary of this system is the fifth limiting system, in which the primary continuous phase /the binder/ is completely absent, so that the system contains only one solid phase, dispersed in a fluid medium. Type IV composites are not important for design materials, and are encountered mostly as soils.

Thus, in a schematic and very simplified way we can represent the sub-division of constructional composites /as the basis of a general theory/ in terms of structural arrangement as shown in Fig. 16. Only if the study, description and experimental investigation

these systems is therefore the external medium, its properties and the method of interaction with the internal surface. The solid phase occupies a continually decreasing part of the total /external/ volume of the composite, and there is a loss of binder up to the moment when the primary structure of the binder breaks down and it loses its phase continuity /this is the fourth limiting system/. The material becomes free-flowing /Fig. 15/ and its properties are determined mainly by

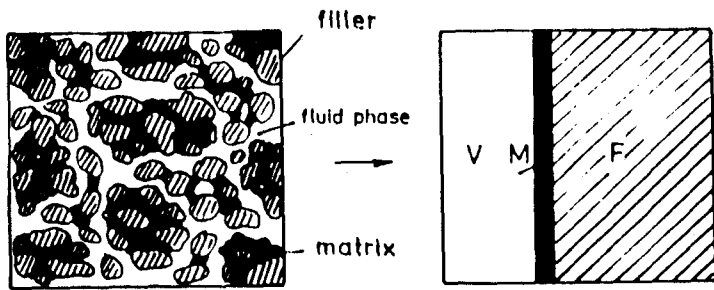


Fig. 15 Composite material of the fourth type

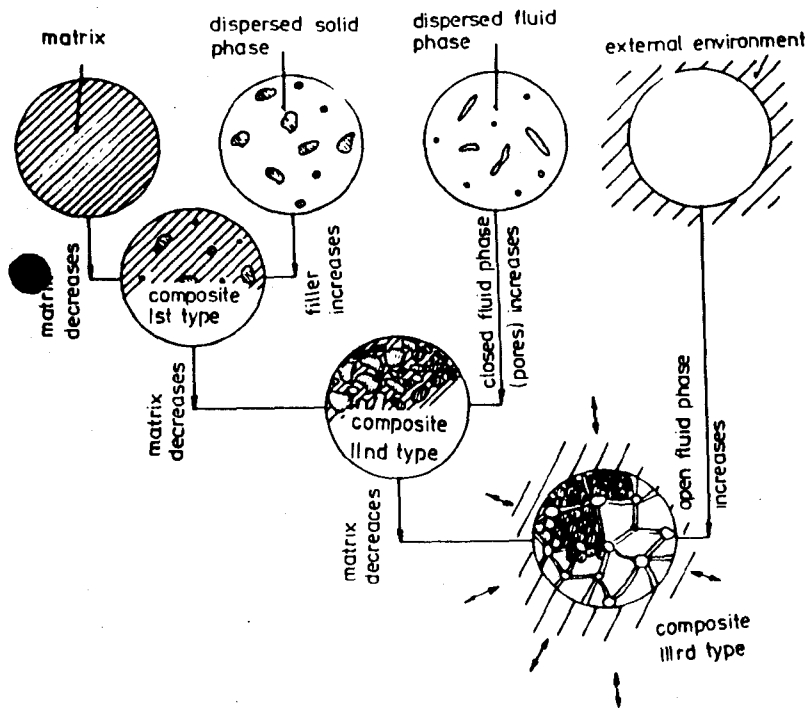
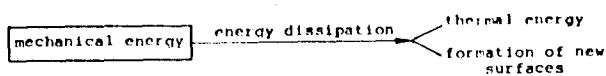


Fig. 16 Schematic division of composites in terms of the /macro/structural arrangement

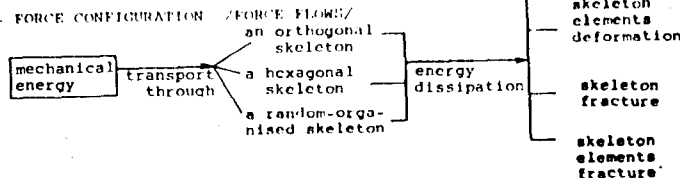
TWO BASIC MECHANISMS OF DEFORMATION PROCESSES IN THE STRUCTURE:

- SHEAR FLOWS ALONG PHASE BOUNDARIES



/mostly composites of the 1st type/

- FORCE CONFIGURATION



/mostly composites of the 2nd and 3rd types/

Fig. 17 Two basic mechanisms of deformation processes in a structure

are sub-divided on this basis is there any prospect of obtaining less ambiguous results, which are objective, reproducible and non-contradictory.

Let us also remember that a physical description of the structure comprises physical indices of each component of the system, of their interaction and phase boundary. Because the physical constants /functions/ of the composite are random spatial functions, they can only be described statistically. For this, however, it is necessary to know the complete geometrical description of the structure and the physical properties of the components, a knowledge of the volumetric phase composition alone is not adequate.

The mechanical processes, which are a function of the relationship of the geometric and physical parameters, take place in the structure through two basic mechanisms /Fig. 17/:

- A. Deformation processes acting by shear flows along the phase boundaries and causing changes of mechanical energy of dissipation into thermal energy or into mechanical energy expended on the formation of new surfaces /crazes, microcracks or cracks/. These deformation processes are typical of type I composites.

A typical example of the stress transfer in the matrix following primarily the indirect, namely shear flow mechanisms along the interphase boundaries, is shown in Fig. 18 /5/ for the Type I composite model made of bronze circular inclusions in epoxy matrix investigated by the photoelastic method.

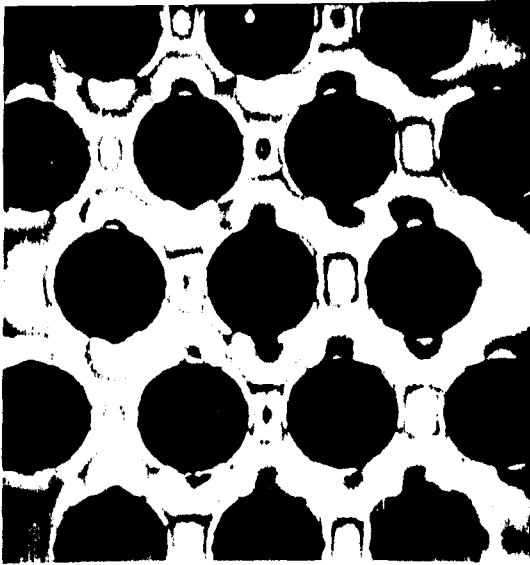


Fig. 18 The shear flow mechanisms along the interphase boundaries in the Type I composite model

with the same geometric arrangement as former but differing in the spacing of the individual inclusions given in Fig. 19 /5/ show near the direct force transfer from grain to grain.

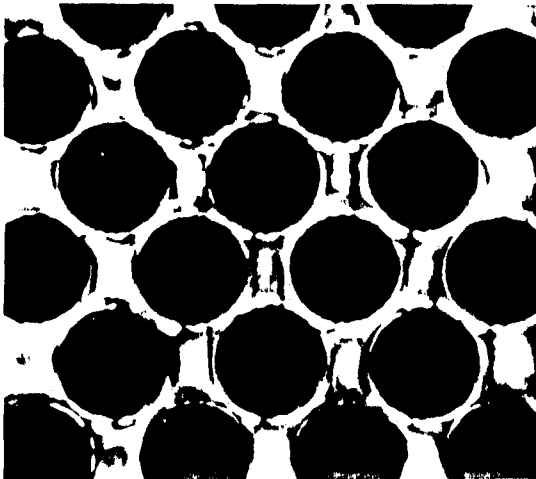


Fig. 19 The direct force flow mechanism among the dispersed particles in the Type II/III composite model

multi-structural composites is therefore one of the reasons for the markedly different properties of such systems.

4. Elastic constants of composites

On the basis of the above described proposal for a theory of composites, equations have already been derived which accurately characterize the elasticity of these systems. In addition to the interaction of the individual phases and their intensive participation in the process /as is normal for practically all so-called mixing rules/, the degree of structure of a system with a definite internal surface and its interaction with the ambient medium have also been introduced. This makes it possible also

B. Deformation processes causing the formation of force configurations and force flows in the structure and hence the occurrence of stressed and unstressed areas in the structure. Then the external load is resisted by a framework of force paths which is formed in the structure, is often orthogonal or hexagonal and is depending also on interface properties.

Mechanical energy is consumed for the deformation of the internal skeleton /incl. the interphase region/, for the failure of the skeleton and the failure of the individual skeleton particles. The results of photoelastic investigation of the analogic model

The origin of closed pores improves the inner stress state /primary and secondary/ similarly as in Type I composite; from the mechanical point of view, therefore, their presence in the superstructure could be advantageous.

This process is particularly important for type II and III composites. The most important parameters are the size distribution, the concentration distribution, the phase continuity and the ratio of the physical properties of the phases. The fact that both deformation processes evidently act in

to take account of a fluid phase in the system, not only in terms of its volume and elastic modulus, but also in terms of the magnitude of the internal surface to which it is bound /6/.

5. Strength of composites

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 /7/.

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 concentration of 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 at least 4 orders, its velocities fluctuating between merely 10^{-10} cms⁻¹ in the case of subcritical microcrack and crazes growth and 10^5 cms⁻¹ in the case of brittle fracture in a macrovolume of the body.

A number of sophisticated 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 soft 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 and 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 as also 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 factor 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 effects of failure barriers, i.e. the consideration of principal technological aspects.

However, what we can apply immediately from the afore mentioned 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 surface/ by an analogy with the initial state of origin of microcracks under stress. This state corresponds with Type II composites /Fig. 13/. 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.

6. Conclusion

The above described outline of the general theory of composites, based on a structural survey and a synthetic rather than analytical approach, should provide users of composite materials with an effective aid to understanding and mastering planned formation of composites. The author considers this aim particularly important because in the future growth of population much more attention should be given to composites since of current materials they have the highest specific efficiency. Only in that way it is possible to ensure adequate saving of matter and energy dictated by restriction of their sources.

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