

ML TDR 64-85

FOREWORD

This report was prepared in the Materials Information Branch, Materials Applications Division, of the AF Materials Laboratory, Research and Technology Division under Project 7381, Task 738105, "Ceramic and Graphite Technical Information." This report was written to provide a summary of the principles influencing composites.

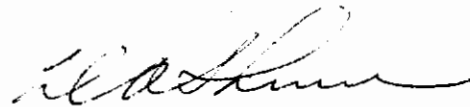
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ABSTRACT

The high strength of thin metallic and inorganic fibers, whiskers, and flakes can be exploited if they are properly protected and bonded together by a suitable adhesive or matrix material. The principles which appear to influence the strength of thin specimens and their mechanical behavior in such a matrix are reviewed in this report. Qualitative attention is given to series and parallel failure mechanisms in the fibers, to the description of brittle behavior, to bonding and the mechanisms of stress transfer, and to testing methods and evaluations. Semi-quantitative relations are adduced to suggest optimization of mechanical properties, and composites containing SiC are discussed to illustrate these relations.

On the basis of the principles discussed, suggestions are made for selecting future composite materials and designing them for specific applications.

This technical documentary report has been reviewed and is approved.



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INTRODUCTION

Because of the success of glass fiber-reinforced plastics and the discovery of very high strengths in metallic and ceramic filaments and whiskers (Reference 1), research and development in the field of fibrous composites has increased greatly in the past ten years.

It is perhaps unfortunate that so large a proportion of this effort until recently has been confined to laminates of glass fibers in resin matrices. Although these composites have now been developed sufficiently to exceed all other materials in tensile strength to weight, the principles peculiar to them have too often been generalized and used to measure the potential of other materials and other geometries. It is the purpose of this report to review the status of materials knowledge in this field as a whole, so that realistic plans can be made for developing improved composites for U. S. Air Force applications.

In a properly designed composite, each constituent performs a particular function and modifies the environment to which other parts of the system are exposed. For the purposes of this report, only those composite systems will be discussed in which the highest achievable strength (or elastic modulus) can be achieved constituent with minimal density. Such pertinent criteria as refractoriness, chemical stability, and fabrication techniques will be considered in discussing each type of composite.

BACKGROUND THEORY

The Peculiarities of Materials of High Strength or High Modulus

The elastic moduli of a solid substance are resultant from the forces holding its constituent atoms together (bond strength) and the spatial arrangement of these atoms (structure), at, of course, certain conditions of temperature and pressure. For most crystalline compounds, it is possible to calculate theoretical modulus values, and good agreement with observed moduli has been noted with simple metallic and ceramic crystals. With corrections for crystallite boundary characteristics and proportion, reasonably good agreement has been noted even for some polycrystalline substances.

On the argument that a perfect crystal cannot be pulled apart until the imposed stress is comparable to the bonding strength of the crystal, attempts have been made to predict the "ideal strength" of several materials. No experimental data can be offered to confirm these high ideal strength values, however, since real specimens break at far smaller loads. It has been observed, however, that the maximum measured strength for monocrystalline whiskers and filaments approaches about five percent of the calculated Young's modulus. For this reason it has been suggested (Reference 2) that the probability of being able to attain the greatest possible tensile strength (in an ideal crystal) will be proportional to the calculated modulus for the material.

Manuscript released by the author 8 April 1964 for publication as an RTD Technical Documentary Report.

Since the strongest interatomic bonds are the shortest bonds, and since the atomic density of a material of a given structure is proportional to the mean atomic weight of the elements in it, it follows that:

those substances having the highest potential strength to density ratio will be those having the lightest possible atoms in the closest possible packing.

The same criterion holds, of course, for maximum modulus to density ratio.

Caution must be used in applying this rule. The tensile strength actually obtained even in a thin single crystal is subject to the weakening influence of pores, impurity and vacancy sites, dislocations, and in some instances, locked-in residual stresses. Nor can any solid body be divorced from the influence which its own surface must produce; the surface atoms of a crystal must possess either unsatisfied valencies or chemisorbed foreign ions, even when gross surface flaws are not present.

It can be stated with some assurance, however, that a "figure of merit" for most materials can be computed which will serve to select the most promising candidates for future work. This has been done for some compounds and for a few elements in Table 1 by dividing the ideal modulus of each by its atomic density.

Despite the difficulties of achieving ideal tensile strength in any real specimen, the critical problem is that of exploiting the real strengths which have already been obtained. These are amply high: graphite single crystals have exhibited strengths of about 4×10^6 psi; BeO and Al_2O_3 nearly 2×10^6 psi; and glass ribbons resisting more than 10^6 psi have been reported in axial tension.

Important considerations in the selection of materials involve correctly assessing the ability of the material to be formed into a useful shape, and assuring that in this shape it will exhibit the properties for which it was selected, and that in service it will be exposed to the environment for which it was designed. It has been found that these probabilities decrease as the size of the specimen, and its modulus, increase.

The Influence on Tensile Strength of Specimen Size

The observation that a filament of glass is stronger (in tension, torsion, and flexure) than a rod, which in turn is stronger than a thick bar, illustrates one of the important peculiarities of brittle behavior. That a thin specimen exhibits higher nominal strength than a thick one is customarily explained by the greater probability that a critical flaw (Reference 3) will exist in a larger gage volume (Reference 4).

The wide variation in nominal fracture strength usually observed in brittle materials prevents any rigorous relation between size and fracture stress values. The popular theory of Weibull predicts that the relative tensile strengths of two specimens of identical material should be:

$$\frac{X_1}{X_2} = \frac{V_2^{\frac{1}{m}}}{V_1}$$

TABLE 1

CALCULATED MODULUS TO DENSITY RATING FOR SELECTED CRYSTALS

Crystal	Modulus million psi	Density lb/cu.in.	Fig. of Merit million/in.	Strength million psi
diamond	170	.126	1350	8.5
$3AlB_{12}SiC$	75	.091	825	3.8
B_4C	66	.090	734	3.3
Be	45	.065	692	2.2
SiC	71	.115	616	3.5
B	51	.083	615	2.6
TiB_2	94	.162	580	4.7
Be_2C	46	.087	529	2.3
Al_2O_3	76	.144	526	3.8
BeO	51	.108	472	2.6
TiC	72	.177	406	3.6
ZrB_2	64	.202	317	3.2
MgO	35	.126	278	1.8
FeS_2	50	.180	276	2.5
Si	23	.083	275	1.7
$MoSi_2$	55	.217	244	2.8
ZrC	50	.246	203	2.5
NbC	50	.282	177	2.5
WC	104	.580	173	5.2
Steel	28	.282	99	1.7
W_2C	62	.625	99	3.1
W	60	.696	86	3.0

where X is strength, V is volume, and m is the Weibull "flaw density factor" in the equation:

$$F(x) = 1 - e^{-\frac{X - X_u}{X_o}^m}$$

where X_u is the highest imposed tensile stress at which the body will never break (sometimes called the "zero strength") and X_o is a normalizing factor (Reference 5).

This theory assumes that the flaw density m for a given material is constant and independent of size. If m is infinite, the material is classically perfect, and the value of m has been used to indicate the brittleness of the material tested. Salmassy (Reference 6) suggests the values given in Table 2.

TABLE 2

FLAW DENSITY EXPONENTS

	m
Glass fibers	1.3
Nickel-cemented TiC	7
Hydrostone plaster	15
Steel at liquid air temperature	24
Spark plug porcelain	35
Steel at room temperature	58

From the data given it can be seen that the value of m is not necessarily unique for a given material; indeed, separate batches of brittle ceramic materials almost always show different values of m.

It is not the writer's intention to review the many theories which have been advanced to describe brittle fracture phenomena; this has already been well done (Reference 7). It should be noted, however, that most of these theories presume that brittle fracture is initiated by the concentration of stress at a critical flaw and that failure ensues by propagation of one or more cracks issuing from this flaw, and that the propagating energy is derived from elastic strain.

The "strength" of a brittle material, then, depends not only on atomic cohesion but upon the number and location of critical flaws in it and upon energy considerations in the formation and extension of cracks. For this reason, it is highly improbable that any two specimens of a brittle material will fail at exactly the same stress, and statistical methods are used to describe the probability of fracture, rather than a mean or average strength value.

Unfortunately, a large number of specimens, identically treated, need to be broken to obtain a reasonably accurate picture of the distribution of failure stress values and the probability of fracture. The picture is further complicated by a situation which, in the opinion of the writer, is important and not often recognized. This is the internal error in the tests by which these values are obtained.

The Dependence of Strength on Testing Methods

To obtain the strength and/or the stress-strain relation for a material, it is necessary to have some method of relating the load on the specimen to the stresses in it. In one of the simplest of mechanical tests, that of a prismatic rod under axial tension, the physical assumption given by Saint-Venant is made that, if the bar is sufficiently long, all stress distributions on the ends of the bar will produce a uniform stress in the center of the bar.

With truly brittle materials, however, the applicability of this principle is open to question for all specimens of reasonable length. Suppose that the material in question is totally brittle, and possesses no mechanism for plastic deformation. In this event a typical cylindrical rod specimen, gripped at its outer end surfaces, will be in tension throughout, but the axial tension will be greater on its outer surface throughout than at its core. This could be measured by applying strain gages to both inner and outer surfaces of an axially stressed cylinder, but this appears not to have been tried.

The axial stress at the center of such a tensile specimen will be lower than that at its surface by an amount which:

increases with modulus,

decreases with specimen length, and

decreases with gage diameter.

How serious an error in calculating the stress imposed on tensile specimens this may have been can be judged only after careful experimental work has been done with well-characterized, high-modulus materials. It may well be sufficiently large to explain the abnormally high sensitivity of strength to the surface imperfections of some brittle materials, and partially to explain the unusual increase in measured strength observed when these materials are tested as thin whiskers, filaments, and ribbons.

A second problem arises from the difficulty of obtaining, in actual test, pure axially in tension without superimposed bending or torsion. The importance of axially is greatest with brittle materials; it can be shown that the proportion of increased stress produced is eight times the ratio of eccentricity to diameter for a circular rod specimen.

The problems relating to performing a satisfactory uniaxial tension test are formidable. Recent tests (Reference 8) have shown that increases of up to 20 percent in mean nominal strength of brittle oxides are obtainable when "gas bearings" are employed in the load train of a tensile test machine to minimize bending and torsion moments.

A moment of reflection on the importance of this observation is worthwhile. Obviously, a change in testing technique does not make the material stronger or weaker, yet it can make a significant change in the nominal stress value at which the material fails. The answer must be, of course, in the "nominal" nature of the imposed stress.

It has long been noted that when a bar of brittle material is broken in transverse rupture, the calculated "ultimate fiber tensile strength" is greater than the uniaxial tensile strength for the same material. It has often been suggested that this apparent increase in strength is largely due to frictional forces which provide a resisting couple and a consequent increase in the apparent strength of the specimen by as much as 30 percent. Clearly this resistance due to friction, being proportional to the true fracture loads, introduces a systematic error in the measurement.

Nevertheless, when the highest values of the refined uniaxial tension test are compared with flexural loading test data obtained with roller supports or corrected for frictional restraint, the values are commensurate. (Materials such as concrete or polycrystalline graphite, which may exhibit compressive moduli appreciably different from their Young's moduli, cannot be safely compared in flexural rupture with axial tension).

Other methods of stressing specimens in tension have been developed which minimize restraint and multiaxial stress. Two of these are the "brittle ring" test adopted by Bortz et al., (Reference 9) and the Stanford Research Institute uniform internally pressurized ring tension test (Reference 10). Both tests result, for a given material, in higher computed mean tensile strengths than those obtained in uniaxial tension tests, even with "gas bearings;" more importantly, both result in significantly smaller deviations from the mean values of modulus and strength.

For a long time it has been assumed that the wide variation in strength observed with all brittle materials is attributable to their inherent inhomogeneity. Since the tests referenced above were performed with a wide variety of brittle materials ranging from graphite and brittle resins to polycrystalline oxide ceramics and in all cases produced smaller deviations than those observed in axial tension and flexural tests, it would appear that a large part of the apparent spread of strength values is, in fact, caused by localized (or at least mathematically unresolved) stress concentrations induced by interactions between the specimen and imposed stresses.

In the case of the brittle ring specimens, the imposed stress gradients (tangent to the axis of compression) are so steep that critical flaws outside this area are no longer critical; in consequence, the specimen acts like a specimen of smaller volume and consequent higher strength. In the case of the SRI test, the internal stresses are equally imposed upon the whole unrestrained thin-walled cylinder so that areas of different strength may be most nearly compensated by elastic bending.

It appears, then, that the term "strength" when applied to brittle materials has a meaning which must cautiously be applied. It is obvious illogic to expect a tension member or transverse beam of a brittle material to display the same strength as that of the same material in a highly refined test such as those described here. The illogic of the converse statement, however - that the strength values obtained by ordinary testing techniques are sufficiently precise or meaningful to serve to evaluate the potential value of brittle materials or to serve the designer's needs - has yet to be recognized.

Whether very thin specimens appear to be strong because they contain fewer flaws or because they, like the SRI cylinder, can be more uniformly stressed remains to be established by exhaustive studies of the testing techniques themselves. It may, however, be argued that since thin specimens promptly lose virtually all of their great strength when

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they are restrained, the second explanation will be found to account for the observation that filaments, whiskers, and ribbons increase in nominal tensile strength most rapidly when one of their dimensions is smaller than about 10 microns.

The Parallel Model

The parallel model has received only a fraction of the attention paid to the series "weakest-link" model. In the latter it is assumed that failure occurs when a stress concentration occurs anywhere exceeds the ultimate tensile strength of the local material, that this is most apt to occur at the site of a critical flaw, and that the stored elastic strain in the material adjacent to this flaw is sufficient to supply the energy required to enlarge the flaw and propagate it as a crack.

In the parallel model, one may visualize a bundle of n threads on which is imposed a tensile load S . If this stress is equally distributed throughout the bundle, each thread will feel a load S/n , and the bundle will not break so long as a number of threads k have strengths exceeding S/k (Reference 11). This is a rather complex way of saying that the strength of the bundle will be as great as the summation of its strongest fibers rather than their mean.

It may be helpful to consider the parallel model as a means for explaining the failure of the flaw-density relation to account for the very high strengths of very thin specimens, using measured strengths and m values calculated from the equation in section B for thin ribbons of pyrex glass (Reference 12).

It is worthy of note that the widest deviations in observed strength values occurred with the 4-micron ribbons (from 36,700 to 1,480,000 psi) and were somewhat smaller for thinner and thicker ribbons. Higher strengths (averaging 17,000 psi for ribbons 4 microns thick) and smaller deviations (from 97,000 to 317,000 psi) were observed for fresh soda-lime glass ribbons.

Most notable in the tabled values is the significantly increased value of m , the flaw density coefficient, for the thinnest flake. Although microscopic examination of these flakes revealed many serious flaws (largely bubbles or holes and striae) in all of them, the increase in strength due to size reduction was greater than that predicted by the theory of random flaw distribution. It must therefore be concluded that the greatly increased tensile strength observed with very thin specimens must be attributable in part to one or more other factors than that of flaw density. The reasons for this conclusion are summarized below:

1. As specimen thickness is decreased, all materials increase in nominal tensile strength. The phenomenon is not, therefore, exclusively attributable to brittle materials, but the increase in strength observed is more spectacular with substances of high modulus. Nor is increased strength peculiar to glasses or single crystals; similar results are achieved with polycrystalline filaments.
2. Very thin specimens fail in the fashion characteristic of brittle materials. whether the bulk material is brittle or ductile; elongation and necking are seldom observed.
3. Axiality of loading is less critical with thin specimens than with thick ones; this is demonstrated by the straightening of moderately curved specimens under load without serious loss in strength.

TABLE 3

STRENGTH OF PYREX GLASS RIBBONS

Thickness (microns)	Mean Tensile Strength (psi)	m
2	193,000	1.9 ± 0.25
4	134,000	1.4 ± 0.2
6	100,000	1.3 ± 0.15
8	79,700	1.5 ± 0.1
12	61,000	1.35 ± 0.05
80	15,000	

4. Despite the relative increase in surface-to-volume ratio which must occur in thinner specimens, the strength increase for "surface-sensitive" materials like glass is as great as that observed with less sensitive materials (such as sapphire) as the specimens are made thinner.

In order to provide an adequate background for discussing means of exploiting the very high strength of thin filaments and fibers, it may be profitable to discuss these observations as they relate to the parallel model.

The classically perfect, ductile substance can be visualized as a bundle of thin specimens, each attached to its neighbors by bonding forces indistinguishable from those in the filament itself. As a tensile load is applied, each filament supports a portion of it, reached nearly at the same stress level. The outermost fibers, being under somewhat greater stress, yield first (and this will be more notable in substances of high elastic modulus or when surface imperfections exist). The elongation of the outer fibers provides a shear component which is oriented toward the center of the specimen and necking begins, terminating in the familiar double-cone or cup-and-cone failure in shear.

For a flawed material, the ductile behavior is similar except that each flaw acts as a point or volume of stress concentration about which the shear vectors for adjacent fibers are centered. As a consequence, only the unflawed filaments can accept their full proportion of the stress; these are strained to their elastic limit at a smaller load than that of the ideal case. The presence of flaws (except of the kind which hinder dislocation generation and movement) will thus reduce both the modulus and strength in proportion to their number and size in the volume of the specimen.

As the elastic modulus of the (isotropic) substance is increased, a larger stress in each filament is necessary to produce proportionate strain, and the mechanism of shear-stress transfer is proportionately denied at the surface of the specimen. As a consequence the inward compressive vector is diminished, and necking becomes impossible at strain-energy levels below those of the cohesive bonds in the filaments themselves. A similar

weakening effect is observed near each flaw.

In substances which are brittle, then, the explanation for their brittleness lies in the diminution or denial of the shear-stress mechanism of transferring stress from a strained filament to those around it at energy levels safely below the bonding energy of the material. (This definition of brittleness will be implied throughout this presentation. It must be emphasized that a material is not necessarily brittle because its elastic moduli are high, nor is a brittle material necessarily weak.)

Since the shear-stress mechanism requires mass diffusion (dislocation or twin generation, dislocation movement and slip, or the formation of new phases) materials of high bond-strength and structures in which such diffusion is restricted will tend to be brittle at all ordinary temperatures. (The ductile-brittle transition temperature is related to the energy threshold at which a principal diffusing mechanism can occur.)

Brittleness in the parallel fiber model, then, will occur whenever the shear-stress mechanism for transferring tensile stress is achievable only at strain energies comparable to the cohesive energy of the filaments themselves. Upon this basis, the reasons why thin filaments of brittle materials are very strong is most easily understood through recognition of the weakening influence of increasing size.

The finest imaginable filament is a single crystal (or molecular chain) of a length infinitely greater than its thickness. Such a crystal would everywhere resist tension with the bond forces within it, and its strength, if it could be measured, would approach the theoretical elastic moduli given in Table 1.

If this filament is surrounded by - and attached to - precisely similar crystals in perfect array it would exhibit a similar strength if a method could be found for distributing the tensile stress uniformly across its cross-section. To the extent that crystals can be elastically distorted in three dimensions (such as cubic-rhombohedral distortion) the strength would not be greatly diminished.

The presence of impurity ions or structural imperfections must of course reduce the ability of these crystals to deform elastically or to resist throughout stresses comparable to their bond strength, and weakening would then be observed, even when the flaws are far smaller than those regarded as "critical" by Griffith.

In a real crystal of finite size, as outer crystallites are deformed they attain energy values which can be dissipated either by mass diffusion mechanisms or by the creation of new surfaces, e.g., shear failure. As a consequence, there is a limit of strength achievable by any monolithic material which is determined by the stress to modulus ratio at which the monolith is just separated by shear forces into groups of crystallites of nearly ideal structure. This limit appears to be equivalent to strength values variously estimated at from 3 to as much as 10 percent of the modulus (depending on the elastic distortion permissible) in the ideal crystal. The values listed in Table 1 were calculated on the common basis of five percent of the estimated moduli).

It should be noted that the "ideal filament" described here represents a ceiling value for tensile strength based entirely upon principles which can be postulated from crystal data. It remains to be decided whether strengths approaching those of the ideal filament can be approached and whether certain materials are better candidates than others for continued development.

In the crystalline monolith under tension, separation of the crystallites adjacent to each other into "packets" or mechanical domains achieves a reduction in strain energy by forming boundaries in which mass diffusion can more easily and rapidly occur. These boundaries will tend to capture impurity atoms and vacancy sites and act as dislocation sources. The stress which can be transferred across them will be safely less than the cohesion of the crystals (Reference 13). That such domain boundaries do form is indicated by increased gas diffusion through strained crystals.

These intercrystallite boundaries are the source of real strength of the material in tension. Formed along directions of easiest shear (determined by crystallite orientation and by tensile direction) they may, depending on the diffusion mechanisms available to them, act to produce dislocations, extend to form slip planes, or serve as nuclei for the formation of more favorably oriented crystals.

In any of these mechanisms, part of the tensile strain energy must be absorbed either by transfer (as elastic strain) to an adjacent crystallite, or by diffusion of mass in a way which is analogous to plastic flow.*The result in both cases is an elongation of the specimen under increased tension.

In a ductile material, the forces necessary to supply an inward pressure on the specimen are provided by dislocation movement and slip along shear planes. If this movement is prevented (Reference 14) these forces are unable to act and mass diffusion can no longer occur.**

It can be seen, then, that both ductile behavior and the achievement of maximum strength capacity in any material depend upon diffusion mechanisms which permit elongation; if diffusion is slowed to a rate less than that demanded by the rate of loading, the material will fail in a brittle fashion.

Whether or not a material is really weakened by becoming brittle is not known with certainty; it is certainly true that all ceramics are weakened at sufficiently elevated temperatures, but the relation between their strength and modulus is, in the absence of phase transitions, usually unchanged.

It would appear from these considerations that the problem of brittle behavior might be more easily attacked if some care is given to the measurement of tensile strength in metals at temperatures just above and below the ductile-brittle transitions. (There is little advantage in choosing alloys for this work, since they are nearly as complex in their atomic structure and microstructure as the polycrystalline ceramics; however, the relative ease in forming and machining most metals makes them attractive for such a study.)

*Dr. Weyl has, on a similar basis, adduced evidence to show that glasses also have a 2-phase structure.

**This has been demonstrated in an ingenious way. By brazing closely spaced copper washers around a mild steel tension specimen, Dr. Shanley was able to achieve brittle failure at higher than normal ultimate strengths.

***It should be noted that most brittle materials are (apparently) strengthened and more ductile where they are broken in tension under sufficient isostatic pressure. The relation of this observation to this discussion is self-evident.

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In concluding this part of the discussion it should be pointed out that continued elongation without adequate radial diffusion constitutes triaxial tension. Unfortunately, it is difficult to design an experimental loading system capable of producing triaxial tension. It appears, since many materials which are adequately ductile in uniaxial tension are subject to brittle failure in biaxial tension near their transition temperatures, that triaxial failure would be still more abrupt. Experience with thin pressure vessels would support this contention.

It is concluded, therefore, that the increased tensile strength which can be achieved in a sufficiently thin filament is the result of the following causal conditions:

1. Critical flaws are fewer in the thin specimen,
2. Biaxial and triaxial stresses are less apt to be obtained in a thin specimen,
3. The stress gradient across or through the thin specimen is necessarily smaller in the thin specimen,
4. Locked-in or residual stresses are less apt to occur or are smaller in a thin specimen, and
5. The thin specimen, since it is not restrained by external material of the same modulus, can adjust itself along the line of tension so that minor eccentricities of loading can be tolerated.

Considerations Influencing Composite Design

In the interests of brevity, this report will be limited to the application of high-modulus materials for structural purposes. The enormous potential strength and high modulus of these materials, coupled with the low density of some of them are attractive for structural applications. It has been shown that this combination of desirable properties can be achieved in some of them if at least one dimension can be very small. It follows from this that the best approach for exploiting these materials is that of combining thin fibers or flakes with one or more other less brittle materials into a composite structure.

The structural applications for which these materials would be best suited are:

1. Tension members, requiring maximum strength per unit weight,
2. Stiffeners, requiring maximum rigidity per unit weight, and
3. Flexural or torsion members requiring both strength and stiffness.

Each of these applications will be separately considered here.

Composite Design for Maximum Tensile Strength

It has already been shown that the maximum tensile strength which could be achieved would be that of a bundle of fine fibers. Unfortunately, filaments of brittle materials are so sensitive to surface damage and to local flexure that they must be protected against contact with each other. This is normally done by attaching a thin coating to the fibers or filaments immediately after they are formed.

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For structural purposes, however, a yarn or rope of brittle filaments is neither practical nor effective; no two filaments are identical in size or strength, and no end attachment system has yet been devised which will distribute the stress among the filaments either equally or in proportion to their strength.

The most practicable solution appears to be the formulation of a composite in which the fibers are separated and protected in a matrix of a second material. (For convenience, the symbol F will hereafter be used to indicate the fibers, flakes, or filaments of the stronger, more brittle material and the symbol M for the matrix.)

Alternatively, the same system might be described as one in which M acts as an adhesive or binder to join the fibers together. Whether it is regarded as a matrix or an adhesive, it is obvious that material M must act as a stress transfer agent between adjacent fibers, and provide thereby a way out of the difficulties involved in making the fibers as long as the tension member. Indeed, if M possesses the proper characteristics, it can provide for the distribution of stresses more equally among the fibers than would be possible in any other way now known.

It should be possible, then, to postulate an "ideal" material M in terms of its role in a fibrous composite. This role will require that:

M should have as high a shear strength as possible,

M should have as low a compressive modulus as possible and

M should bond tightly (or be capable of shear stress transfer by an other mechanism) to material F.

These requirements are worthy of discussion in some detail, since they are not always recognized and are often misunderstood.

The requirement for high shear strength is an obvious one, but is, by itself, easily achieved. In measuring the very high tensile strengths of laboratory filaments, the technique is frequently resorted to of attaching the ends of the filament to the loading bar with an ordinary gummed label. If this attachment is sufficiently strong to break the filament, it is obvious that adequate shear strength is being provided.

It should be remembered, however, that both the shear strength and the shear modulus of any material appear to increase with decreasing thickness to a maximum value. This thickness will hereafter be called the critical thickness; it will, of course, be different for different F and M materials.

It will also be affected by the geometry of the system. In any bundle of cylindrical fibers, the closest distance of approach between two fibers is along the plane through their axes. If this distance is greater than the critical thickness of M, the full ability of the adhesive to transfer stress between them is not realized; if this distance is equal to the optimum thickness, maximum stress transfer will be realized along this plane but nowhere else.

If the content of M is reduced and compaction increased, the M layer may be reduced between fibers to a thickness smaller than the critical value. If this happens de-wetting may occur, permitting contact, or the apparent modulus of the shear layer will rise to

so high a value that each of the fibers restrains the other as in the monolithic structure.

The problem is inherent in the packing of the fibers, and can be attacked only by providing fiber cross-sections which can be so packed together that intervening layers of M are essentially flat. The cross-sections which will permit this are the square, rectangle, rhombus, hexagon, or flattened hexagon. In view of the serious stress concentrations associated with acute-angled specimens, it is doubtful that the rhombus or square can be considered; the flattened hexagon and the rectangle can be approached by ribbons, flakes, and oval cross-section filaments.

The second requirement, for low compressive modulus, stems from the requirement that each filament be as free as possible to adjust itself to its stress environment. This requirement seems at first to be incompatible with the attainment of high shear stress transfer for any isotropic substance.

The matrix or binder phase M, however, is in reality a thin film interposed between parallel fibers; its compressive modulus is less affected by film thickness than its shear modulus. In consequence, for any geometry and composition of fibers and matrix there will be a well-defined optimum proportion for each constituent at which the tensile strength will be highest in the laminate. In the absence of a third phase (pores, fillers, etc.) the laminate proportion which exhibits the highest strength can be used to calculate critical thickness values for M.

The first requirement for M is that its shear strength be as high as possible. In the transfer of unit stress from one fiber to another, the relative elongation of the adhesive film will be approximately inverse to the ratio of the Young's modulus of M to that of F. If the matrix material is insufficiently strong to survive this elongation it will fail by shear, and the high potential strength of the filaments F will not be exploited. The relation of this requirement to the familiar "extensibility" of polymers or the "percent elongation" in metals, is immediately apparent, but often oversimplified.

In transfer of stress, it is generally stated that F and M must be strongly bonded together; from this many investigators have assumed that secondary valence forces at least must be utilized in obtaining adequate bonding. Yet, McGarry (Reference 15) and others have shown that most fibers in glass-fiber-resin laminates are held by mechanical hoop forces exerted by the high-shrinkage resins; in many of the strongest laminates the resultant hoop tension in the resin is sufficient to produce microcracks parallel to the fibers. Since these cracks are normal to the planes of shear stress, they do not, in themselves, reduce the tensile strength of the laminate in the direction of the fibers with which they are associated, but it is obvious that they must reduce the mean resin strength in other directions, and provide corridors through which moisture or other weakening vapors may attack the fibers.

Similar frictional restraint is achieved in the mechanism by which concrete is reinforced with metal rods or mesh, and similar microcracking is observed. The purpose of using knurled or roughened reinforcing metal is that of increasing friction.

It is unwise to assume, however, that very fine filaments can be similarly roughened; attempts to accomplish this by etching fibrous glass and beryllium wire (Reference 16) have resulted in serious degradation of the filaments themselves.

It is equally unwise to assume that continued improvements in laminate strength can be achieved with resins of maximum curing shrinkage (or metal matrices of maximum thermal contraction on cooling). It is obvious that this shrinkage, while it results in a beneficial compressive stress upon the high-modulus fiber, must be accompanied by comparable tensile strains in the matrix material itself. For a cylindrical fibrous system this might not be too serious, but for ribbon or flake geometries, the tension in the matrix would be biaxial, and hence would cause not only weakening of the laminate but bending moments in the flakes.

It therefore is not always reasonable to expect laminates made with flakes or ribbons to be improved in strength by pretreatments or coatings which have accomplished this purpose on cylindrical fibers. By the same token, it is unreasonable to expect that flake laminates can be as strong in one direction as fibrous laminates having the same proportion of F to M.

A recapitulation of the role of the matrix or adhesive must include the following principles:

1. The proportion of matrix to fibers (or whiskers, filaments, flakes) should be as small as is consistent with the method of layup or fabrication; any further increase in matrix proportion will result in reduced strength.
2. The matrix material, for a unidirectional bundle of fibers, should be capable of adhering to the fibers firmly without being weakened by residual longitudinal stresses; shrinkage stresses can be tolerated and may be relieved by microcracking with cylindrical fibers and whiskers.
3. The matrix material should have the highest cohesive (shear) strength achievable with low compressive modulus.

Composite Design for Maximum Rigidity to Weight Ratio

There are three ways of building rigid structures. The first involves the use of gross geometries which insure that bending moments will be resisted by a maximum thickness of material; I- or T- section beams and rigid foams cellular or honeycomb sandwich structures are examples of this for bending loads, and cylinder or L-sections for torsion.

The second also involves geometrical considerations but includes the possibility that two or more materials can be used. For maximum resistance to bending in a single direction, the upper flange of the I-beam is under longitudinal compression only, and the substitution in it of a material of higher compressive modulus will often permit a reduction in weight.

The third involves the use everywhere in the structure of materials having the highest possible moduli (in the direction of imposed stress).

The simplest illustration might be the simple column under a pure compression load; if a maximum load at the least weight is to be borne it would seem that a monolithic column of one of the materials from Table 1 might be chosen on the basis of its figure of merit. (In certain instances such as those when a diamond indenter is used for micro-hardness measurement, this is done).

For columns of fairly large size, the monolithic approach becomes untenable; the compressive strength of brittle materials decreases with increasing specimen size just as the tensile strength does. The reason is, of course, the shear vector of compression, which in an isotropic material is resolved as tension along a surface inclined about 45° from the compression axis. Under compressive loading, relatively tiny radial forces can produce shear failure or spalling.*

For any real monolith, another source of brittle failure is the tensile component of thermal stress. This will be proportional to the modulus, thermal gradient, and thermal expansion of the material; it results in a tensile stress which, when added to that produced by the shear component, decreases its apparent load-bearing strength.

If to these are added the stress concentrations of inhomogeneities and residuals, it will become obvious that the monolithic column in compression must be seriously overdesigned to provide safe support of pure compressive loads. It will be equally obvious that utilizing the high-modulus material to best advantage will require that the material, thin in at least one dimension, be properly used in a composite.

While filaments, fibers, and ribbons have been shown to be best suited for uniaxial tension, they are not so well suited for compression. This arises largely through the interaction of the geometry of the filament with the geometry of packing.

Around each filament in a parallel bundle under axial compression, the shear vector at the interface is one of radial tension. Thus each fiber is stressed in compression in such a way that it tends to force all neighboring fibers away from it. Because no two fibers are equally strong, equally stressed, or surrounded by equally distributed matrix material, failure will finally occur by delamination, or by buckling of the fibers most strongly stressed, or both.

This failure can be minimized, as in the case of the tensile specimen, by making certain that all of the matrix is present in layers of optimum thickness. This can perhaps be done with hexagonal or square cross-section fibers; it can certainly be done with flakes.

It has been stated that there is, for any matrix material, a critical thickness such that its effective shear strength and modulus is highest for a given compressive modulus. In a fibrous system, this thickness cannot everywhere be achieved.

In a composite composed of flakes, however, the parallel flakes ideally can be separated by layers of matrix of the optimum thickness. As a consequence, a greater proportion of the flake material can be used in a given volume of composite; since the flakes are the high-modulus material, the moduli of the composite itself are thereby increased.

* The spalling failure of marble and granite columns can often be attributed to the stresses imposed by weathered detritus in (initial) Griffith flaws near the top. Similar effects are observed with glass columns in moist air or in rods exposed to freezing and thawing.

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In a parallel-flake laminate, then, the compressive modulus is increased in every direction in the plane of the flakes and is minimal in the direction normal to that plane. (A similar anisotropy on the atomic scale is perceived in the strongly-layered structures of graphite, MoS_2 , BN, and somewhat less notably in the illite and montmorillonite clays, gypsum, and layered interstitials like MoSi_2 and the higher borides).

That this anisotropy is significant cannot be doubted; a typical flake glass laminate can be made in which the longitudinal Young's modulus is close to six million psi while the flexural modulus is less than half as much. A laminate of this kind in an L- or T-section would certainly exceed the stiffness to weight ratio now available in metal if it can be made with flakes of the better materials of Table 1.

It has been claimed that flake laminates, since they cannot achieve the tensile strengths of which fiber laminates are demonstrably capable, deserve no further study (Reference 17). It should be noted, however, that most attempts to develop or improve flake glass systems have been based on the pretreatments, coatings, compositions, and laminating procedures which had been developed to produce maximum uniaxial tensile strengths with fibrous materials. In addition, there has been relatively little attention paid to parameters which are vital to the application for which the composite is intended.

It seems important, therefore, to compare flake and fiber systems more fully in order to demonstrate the probable virtues and faults of each. This can most easily be done by examining the design of a structural material which is to be both strong and rigid at minimum weight.

Maximum Strength and Rigidity at Minimum Weight in Tension

Since it is highly improbable that diamond fibers or flakes will soon be available, the candidate material for this example will be beta-SiC (silicon carbide), chosen because it is refractory, oxidation-resistant, relatively inexpensive, and already capable of being reproducibly formed in whisker, flake, and compound-fiber forms. Slightly heavier than B or Be, its theoretical bond strength and achievable modulus are commensurately higher.

Since single crystals of SiC have demonstrated tensile strengths well above 2 million psi, it will be assumed here that with future development, a practical strength average of about 1 million psi can certainly be achieved. Other mechanical properties are: Young's modulus, at least 50 million psi, shear modulus 21 million psi, Poisson's ratio 0.2, bulk modulus about 14 million psi. (For comparison, the fibrous glasses approach one-fourth of these moduli respectively with about half the strength; the Poisson's ratio of most glasses is close to .25.)

It is not so easy to report meaningful mechanical property data for the matrix. The data shown in Table 3 illustrate the difficulty encountered.

Table 3 (Reference 18)

Tensile Strength of a Titanium Alloy Reinforced With Mo Fibers

Temperature (°F)	Ultimate Tensile Strength (psi)		
	Ti-6Al-4V	20 Vol. % Mo wire	Ratio
75	112,000	140,000	1.25
1000	70,000	95,000	1.35
1200	40,000	68,000	1.70
1400	15,000	42,000	2.80

It is apparent that the stronger, stiffer chopped molybdenum wire contributes only slightly to the strength of the alloy at low temperatures. As the composite is heated, however, the modulus and strength of the matrix diminish, and the molybdenum fibers carry a greater proportion of the load.

In this example, however, both materials are metals of reasonable ductility. What would happen if one of them were brittle? The answer (and perhaps some insight into the directions best taken by future development) is contained in Table 4 and Figure 1.

Table 4

Tensile Strength of Silver Reinforced with Al₂O₃ Whiskers (Reference 19)

Temperature (°F)	Ultimate Tensile Strength (psi)			
	Silver	Silver with 35 v/o Al ₂ O ₃		Ratio
		$\ell/d = 100$	$\ell/d = 300$	
75	17,000	67,500	74,500	3.97, 4.38
800	(8500)	-----	72,500	(8.55)
980	6300	45,000	-----	7.14
1280	(3000)	(43,500)	(70,000)	(14.5, 23.4)
1400	2000	(42,000)	(66,000)	(21.0, 33.0)
1600	(1000)	38,200	-----	(38.2)
1690	(650)	-----	48,000	(73.9)
1700	(500)	-----	40,600	(81.5)
1720	-----	25,000	-----	-----

Note: parenthetic values obtained by graphical interpolation.

Figure 1 shows graphically the information given in Table 3. The lowest curve shows the ultimate tensile strength of silver (S_{uAg}) as a function of temperature. Note that the relation is linear to about 1250°F. (The temperature threshold of self-diffusion in pure silver has been reported to be 1280°F.) Above this temperature the metal achieves a measure of plasticity at ordinary loading rates, and the plastic deformation increases, with decreasing strength, to the melting temperature (1760°F). These temperatures and the annealing temperature (400°F) are indicated at the bottom of the graph. The tensile

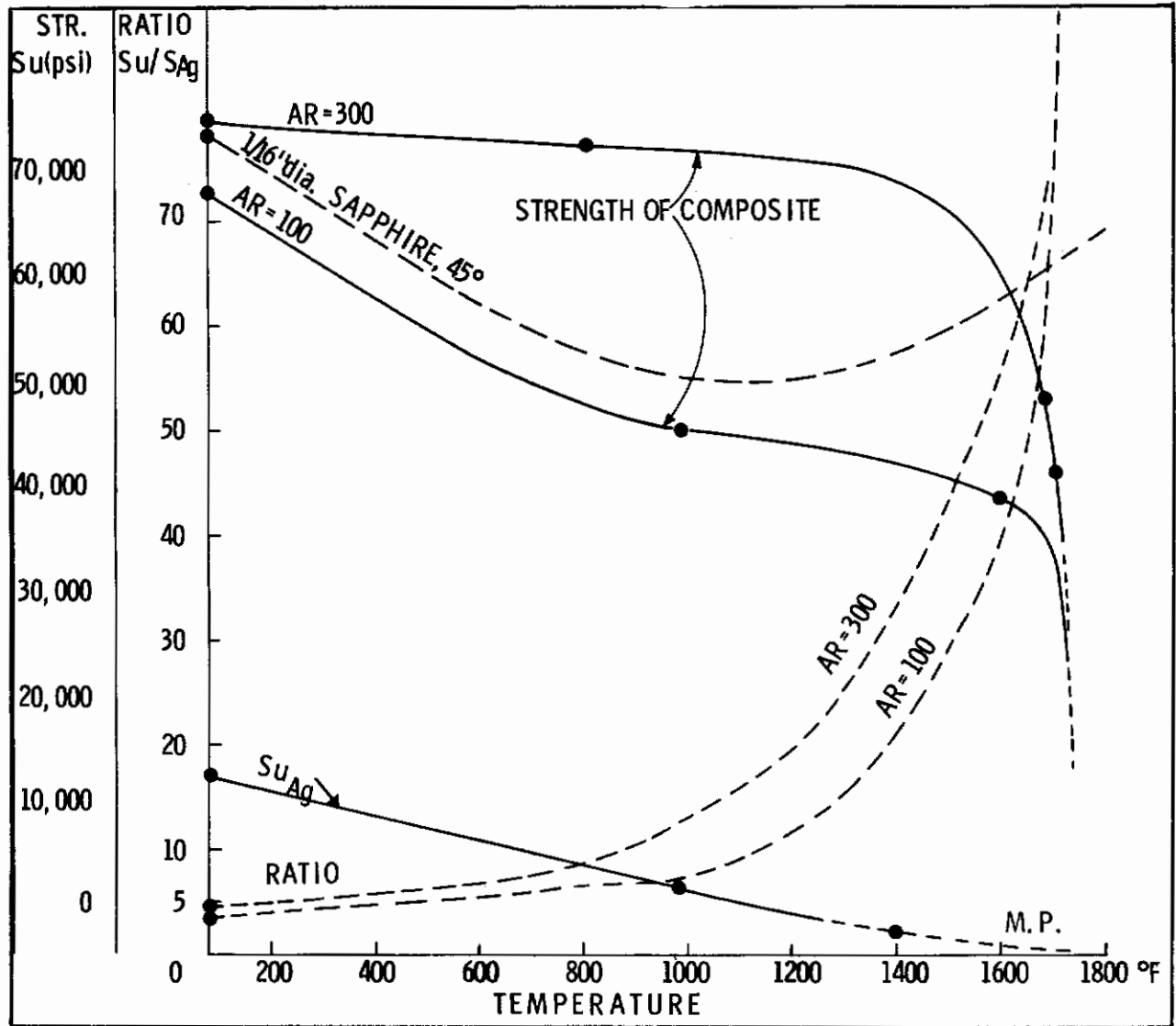


Figure 1. Strength Relations in Composites of Ag-Al₂O₃ Whiskers

elongation of pure silver at room temperature is around 48 percent.

The intermediate solid curve shows the ultimate tensile strength of a composite of pure silver with about 30-35 volume percent of single crystal alumina whiskers in it. The whiskers were selected (by a flotation process) so that their aspect ratio (length to diameter ratio) is close to 100.

At room temperature, this composite is almost four times as strong as silver itself. Since the length of the tensile specimen is many times longer than that of the whiskers, there is no doubt that this strength is contributed by the whiskers, stress among them being transferred by shear through the silver matrix.

At any cross-section of the test specimen, the stressed area consists of about one-third alumina and two-thirds silver. If each makes an additive contribution to the strength, that of the shorter alumina whiskers at room temperature, 1000°, 1400°, and 1600°F would be 157,000, 117,000, 116,000, and 109,000 psi, respectively. At the same temperature levels, the calculated strength contributions of the longer fibers are 181,000, 193,000, 192,000, and 167,000 psi, respectively.

Obviously the longer fibers are no stronger than the short ones. Nor is there a mechanism operating within them which increases the strength of the longer fibers alone at intermediate temperatures. The explanation for these observations must lie in the effectiveness with which stresses are transferred by the matrix among the fibers.

Also on Figure 1 is shown the average measured tensile strength of carefully prepared single-crystal Al_2O_3 (sapphire) rods over this temperature range. It will be observed that the curve closely parallels that of the short-fibered composite, decreasing to about 1000°F. Above this temperature it increases and is still increasing at the melting point of silver. It may be asserted that the observed decrease in strength is in some way associated with flaws and residual stresses in crystalline Al_2O_3 ; as thinner specimens are used the minimum in the curve diminishes and it is not observed in very fine filaments or whiskers. The similarity in the strength-temperature relation for the alumina rod and the short-fibered composite is either sheer coincidence, or it results from some strength-limiting mechanism in the short-fibered composite and the rod which is not reflected in the composite containing the longer whiskers.

Any valid explanation for this behavior must account for: (1) an apparent increase in the strength contribution of the long fibers between room temperature and nearly 1300°F without a similar increase for the short fibers, (2) the relatively rapid decrease in strength of the short-fibered composite to 1000°F and the far less rapid weakening above this temperature, and (3) the "ratio" data shown in the lowest curves.

It might still be argued that the decrease in strength (with increasing temperature) of these composites must be attributed to the decreasing strength of the silver matrix. If this were so, however, the strength of the composite must continue to decrease with further heating. Since this does not occur, one must not only look for another explanation for the flattening of the curve above 1000°F, one must also presume that an increase in strength is occurring in the alumina to compensate for the continued weakening of the matrix metal. (Note that no substantial increase in effective shear modulus of the metal is possible when all the specimens contain nearly the same volume percentage of ingredients.)

Before proceeding to a detailed investigation of the Ag-Al₂O₃ system, certain noteworthy aspects of it should be mentioned. Since silver has a much higher thermal expansion coefficient than Al₂O₃ (22×10^{-6} vs. $4 \times 10^{-6}/^{\circ}\text{C}$), it might be argued that large tensile stresses in the silver are caused by cooling. The annealing temperature of silver, marked at the bottom of the curve, is about 400°F; since there are no observable changes in strength near this temperature it seems safe to expect that the residual strains are quickly dissipated at temperatures higher than 500°F and are quite small at lower temperatures. It is worthy of note that there is a small change in the modulus-temperature curve of silver near 750°F (Reference 20).

Self-diffusion begins in Al₂O₃ at 1275°F: this might be regarded as an annealing temperature, but no inflection in the strength-temperature or modulus-temperature curves are observed near it. Since the composites are formed above this temperature, it is improbable that interface bonding or penetration of the whiskers by the silver is greatly changed by the temperature of measurement.

The rapid loss in strength of both composites near 1700°F is, of course, due to melting of the silver which is complete at 1760°F. (For composites containing a larger proportion of whiskers, this softening would occur at a higher temperature.)

The fibers are certainly no less restrained in the AF = 300 specimens at a given temperature than in specimens with shorter fibers. Being, on the average, three times as long, they are more certain to be parallel than shorter ones. Since there are only one-third as many ends in a given volume of long fibers as short ones, the probability of introducing stress concentrations between two adjacent butt-joined fibers is reduced significantly; it is almost certainly responsible for the small increase in strength observed at room temperature. That this effect should increase at elevated temperatures is absurd, since the importance of stress concentrations (or other local flaws) must decrease with the decreasing moduli of both materials.

It has been pointed out that the apparent strength of alumina rods decreases to a minimum near 1000°F and thereafter increases. This phenomenon might be attributed (Reference 21) to a static fatigue effect; at sufficiently low temperatures the rate of "corrosion" reactions necessary to accelerate bond failure in alumina is too small to be observed at any but the slowest loading rates, while at sufficiently high temperatures the corrosion rate is so high that it is not influenced by added stress (Reference 22).

Somewhere near 1000°F, therefore, one can expect the rate-of-strain sensitivity of Al₂O₃ to diminish; at or above about 1250°F, self-diffusion is sufficient to permit some unpinning of dislocations and result in increased strength. This effect persists to about 3200°F, at which recrystallization and creep become significant.

MODEL OF A FIBROUS PARALLEL COMPOSITE

Knowing that the alumina fibers are sufficiently refractory and inert to be dehydrated but otherwise unaffected by the heating necessary to infuse them with molten silver, that little diffusion of silver into the alumina will occur at any lower temperature, that no phase changes will occur in either of them over the range from room temperature to the melting point of silver, and finally that the alumina whiskers in these composites are sufficiently protected by the silver matrix from external corrosion so that minor

differences in rate of loading will not introduce systematic errors, it appears that this system is an excellent choice as a model for attempting to evaluate the factors which govern the modulus and strength of fibrous composites.

The Ideal Model

Most attempts to derive stress-strain relations in a model of fibers are based upon the transfer of stress from a cylindrical fiber in an external cylinder of matrix (Reference 23), with the assumption that each fiber is so situated and stressed, and that the butt-joints between fibers are so staggered, that an analysis of tensile stress in the whisker and of shear stress in the surrounding matrix is symmetrical from the center of the fiber. Despite these simplifying assumptions, the mathematical solutions are ponderous and imprecise.

The model sketched in Figure 2 is more amenable to analysis. It consists of a hexagonal array of parallel hexagonal whiskers, all having a common "diameter" or least dimension d , length ℓ , and separation by a common radial distance f .

If a horizontal plane is drawn at the end of any "A" fibers, it will intersect three "B" fibers, spaced 120° around it, at two-thirds of their length (measuring down); it will also intersect three "C" fibers, also at 120° around it, at one-third of their length. One may say that each fiber overlaps three neighbors with a common length of $\ell/3$, and three equally distant neighbors with a common length of $2\ell/3$.

Two other slightly rearranged cells like this can be used with it to form a solid body in all directions.

The space between whiskers is filled with matrix. Tensile shear operates on that portion of the matrix lying between the facing parallel sides of adjacent fibers; the shape of the shear unit is a rectangular prism of width $d/\sqrt{3}$ and thickness $f-d$. (For convenience, the shear prisms may also be divided into lengths of $\ell/3$ and $2\ell/3$.)

The material spacing of whiskers f is determined by the volume of fibers in the composite. If the unit cell shown is chosen, its width (to the centers of the fibers) is D and its area A is $0.866 D^2$. Of this area there are one whole fiber and six one-third segments of adjoining fibers of area $0.866 d^2$, and the relation between volume composition and the interfiber spacing f is:

$$\text{Volume \% fibers} = 100 \times \frac{3d^2}{D^2} = 100 \times \frac{d^2}{f^2} = 100 V_f$$

The "unit cell" for this model is a triangular prism containing one-sixth of each of three adjacent fibers, one-half of the three rectangular, shear prisms joining them, and a triangular prism of matrix material among them of side $(f-d)$. It is assumed that the material in this triangle does not take part in the elastic behavior of the composite, but acts as a reservoir of material to permit plastic flow when the elastic limit of any shear prism is exceeded.

Since the overlap of the C fibers with the A fibers is only $\ell/3$, one would expect the elastic limit of the three A-C prisms to be reached first. For fibers of length ℓ and hexagonal cross-section $a = 0.866d^2$, the interfacial unit area of stress transfer is $0.192d\ell$. The ratio of areas $.192d\ell/a = .221\ell/d$. If ℓ/d (the aspect ratio of the

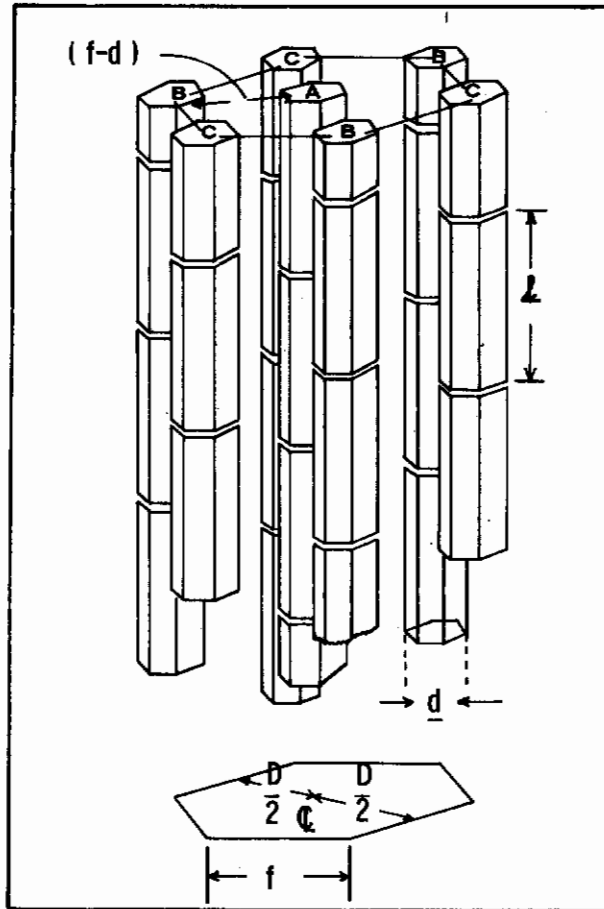


Figure 2. Ideal Whisker Packing

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whisker) is 100, this ratio is 22.2, for whiskers with an aspect ratio of 300, the area ratio would be 65.5.

It is quite obvious that these area ratio values cannot be directly converted into the more familiar modulus ratios for the same materials without taking into account the fact that the ends of the fibers are incapable of being highly stressed by the matrix around them. As a consequence, the effective length of the fibers is somewhat shortened by a proportion which depends on their diameter, their true length, the matrix proportion, and, of course, the moduli ratio of whiskers and matrix material.

It is nevertheless true that below the elastic limit of the matrix, the strains at the interface between whisker and matrix must be equal, and one can therefore equate the moduli ratio to the area ratio, solving for a new value of ℓ .

Table 5 gives the data necessary to do this; since the modulus of Al_2O_3 changes with temperature far less than that of silver, raising the test temperature has the effect of increasing the modulus ratio E_f/G_m .

Table 5
Change in Elastic Constants with Temperature

Temp	E_f	E_m^*	G_m	E_f/G_m
(°F)	(psi x 10 ⁻⁶)	(psi x 10 ⁻⁶)	(psi x 10 ⁻⁶)	
75	55	11.5	4.1	13.4
1000	51	8.5	3.0	17.0
1400	49	6.6	2.4	20.4
1600	48	5.2	1.8	25.7

*(See Reference 27.)

From the equation:

$$\frac{E_f}{G_m} = 0.221 \frac{\ell_{eff}}{d}$$

one obtains, for 10-micron thick whiskers, values of 605μ at 75°F and 780μ at 1000°F.

Dow (Reference 23, 24) indicates a similar but somewhat smaller decrease in effective length based on moduli ratio; Figure 3 is taken from this work. It shows that higher efficiencies in stress transfer result from decreased E_f/G_m ratios and larger whisker or fiber proportions.

The fibers in the real composites described in Figure 1 are neither perfectly aligned, spaced, nor entirely of one size; it is probable that as much of the lowered efficiency of

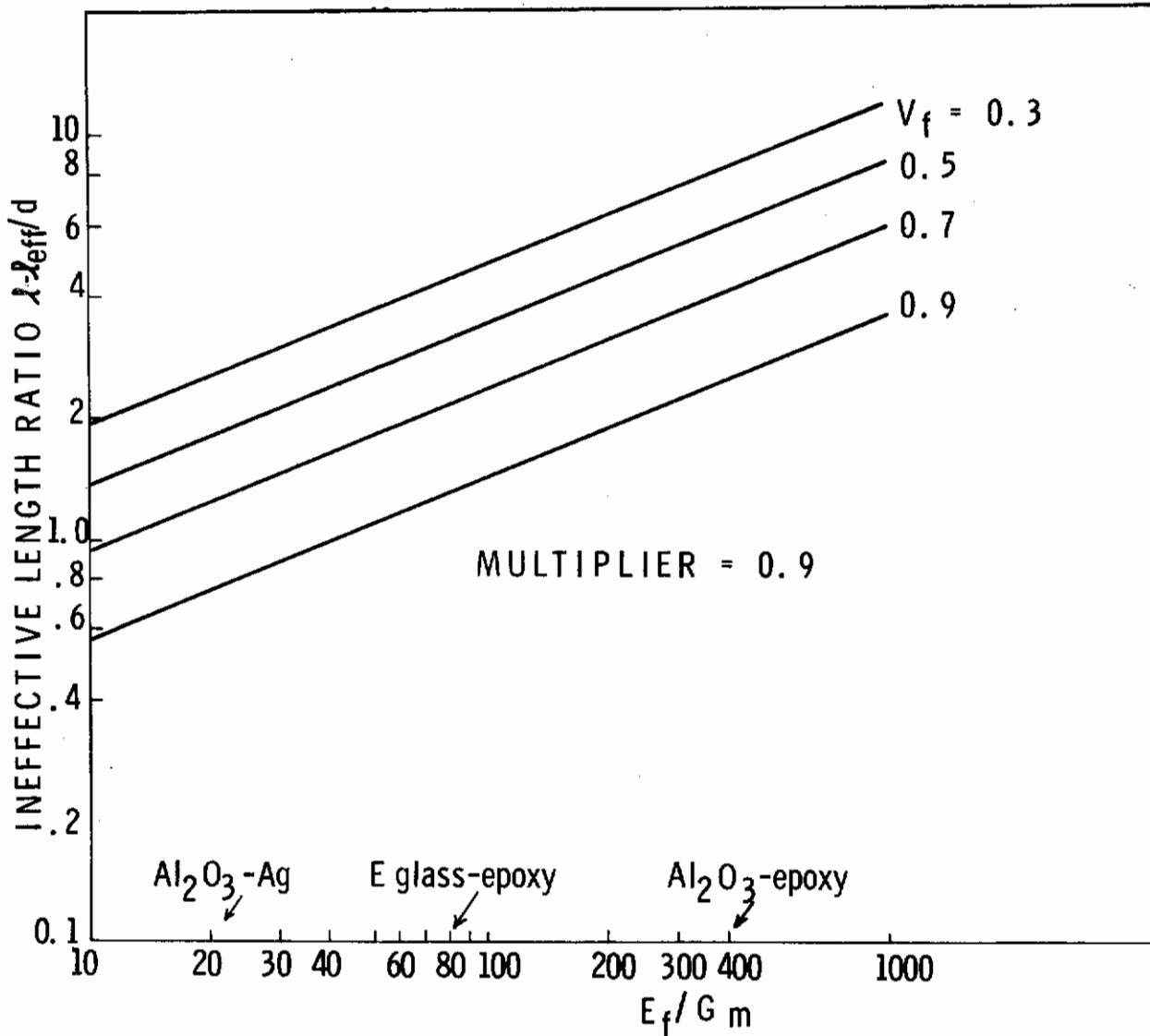


Figure 3. Relation Between "Ineffective Fiber Length" and Fiber/Matrix Moduli Ratio

elastic stress transfer is attributable to this as to the "ineffective length" phenomenon. It will be recalled that at 1500°F the 1000 μ fibers exhibited an average strength of about 115,000 psi while the 3000 μ fibers exhibited an apparent strength of nearly 180,000 psi. On the basis of this model it can only be concluded that the orientation, spacing, and thickness variations in the short-fibered composite collectively result in lowering the effective strength of the shorter fibers from the computed value of about 140,000 psi.

As pulling continues, the strains along the shared length of the interfacial material become larger at the ends than in the middle. In theory the very high interfacial shear stresses in the matrix will at some point be relieved by yielding. For cases where the shear strength of the matrix is low and its modulus is also very low, the second whisker cannot acquire much stress because the elastic strain in the binder is too small to stretch it appreciably. If the binder begins to yield, however, "the load is transferred to the filament in a relatively short distance, perhaps 16 to 20 filament diameters even when the binder is massive compared to the filament. The shear stresses produced near the end of the filament are of the same order of magnitude as the tensile stresses applied to the binder" (Reference 24).

On this basis, the difference between the behavior of the 1000-micron and the 3000-micron composites is readily understood. The former are simply too short for effective stress transfer at low temperatures; shearing stresses are therefore induced in the silver matrix which are so high that failure occurs in the relatively few whiskers capable of carrying the load or at the interface along them. The effect, while large, is comparable to increasing the modulus of the matrix rather than to true fiber reinforcement. This will be discussed more fully in a later section of the memorandum.

At higher temperatures, the ratio E_f/G_m is substantially increased, and plastic shear begins in the matrix before many of the whiskers break. As a consequence, the whiskers tend to accept a proportion of the load over a longer proportion of their length; in the hexagonal ideal model, the load begins to be picked up elastically between shared two-third segments when the E_f/G_m ratio reaches about 23. In the 1000-micron whisker composite above about 1000°F and the 3000-micron composite at all temperatures, and as the temperature continues to increase, E_f/G_m increases and ultimately becomes so large that the composite fails through shear-tensile yielding in the matrix. It is important to note, however, that both composites exhibit respectable ultimate strengths at 98 percent of the melting temperature of silver; elongations at failure are of the order of two percent.

It follows that efforts to develop high-strength fibers will not be particularly effective unless:

1. The fibers are very thin,
2. The fibers can be evenly distributed in the composite,
3. The fibers are sufficiently long to accept stress at the lowest E_f/G_m ratio encountered, and
4. The matrix used is sufficiently low in shear modulus for effective stress transfer, but sufficiently strong in shear to withstand loads approaching the failure load of the fibers in unrestrained tension.

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Deviations from the Ideal in Real Composites

In the ideal composite pictured in Figure 2, it is assumed that the load transferred between adjacent fibers is wholly transferred by the prism of matrix material between them. In a real composite this stress condition may be approached on the average, but local areas must exist where misalignment or poor distribution of fibers are such that an unbalanced bending moment is exerted on certain fibers. Under such conditions, the fibers near this discontinuity act as if they are weaker than they really are.

In the ideal model, all fibers are assumed to be equal in cross-section and in strength. In practical fiber manufacture or composite fabrication these conditions will not be satisfied. To compensate for the inevitable range of fiber diameters and strengths, it is necessary to consider the probability that any fiber will exceed or fall below the "average" fiber strength. This can be done by exhaustive, repetitive tensile testing of representative fibers, but for the present a multiplier which accounts for misalignments, discontinuities, and strength deviations can be used for semi-quantitative evaluation of experimental composites. In some of the Al_2O_3 -Ag composites above, this multiplier is about 0.8; about 80 percent of the strength of the alumina whiskers is realized.

A more serious defect in real composites arises from variation in diameter among the whiskers or fibers used. A thicker whisker must be appreciably longer than the others to carry its proportionate share of the load. It is less able to do so because it will, on the average, be weaker. Still more important is the physical impossibility of obtaining the same "critical" binder thickness in a composite containing fibers of different dimensions. This will be amplified later.

Finally, it will be apparent that while it is possible to make a real composite in which the length of all fibers is identical, it is highly improbable that all or most of them can be arranged so that joint discontinuities are well distributed. It should be pointed out, however, that this objection is not overly serious when all of the fibers are initially long enough so that when they break, the remaining portions will have an aspect ratio in excess of the critical value computed for the E_f/G_m ratio. This is well shown by a simple experiment performed by Rosen, Dow, and Hashin (Reference 23) in which a number of glass filaments were bonded parallel to each other in a thin sheet of modified epoxy resin. Figure 4 shows how, as these filaments break, the total load supported continues to rise, the relation becoming approximately linear after about half of the filaments have broken at least once.

The art of binding high-strength fibers together in a matrix of lower rigidity, then, consists of balancing the mechanical properties of each and the geometry of the composite so that each fiber will carry as much of the load as it can, and when the weakest fiber breaks, the stresses across the break will be transferred to other fibers in the most effective way.

The answer to the perennial question, "How long should reinforcing fibers be for optimum strength?" is one which is dictated principally by the ratio of the modulus of filament to that of binder, and is limited by the strength of the latter. In a properly designed composite, the answer is that longer-than-optimum fibers do no harm (except where they may limit techniques of fabrication or the placement of other fibers).

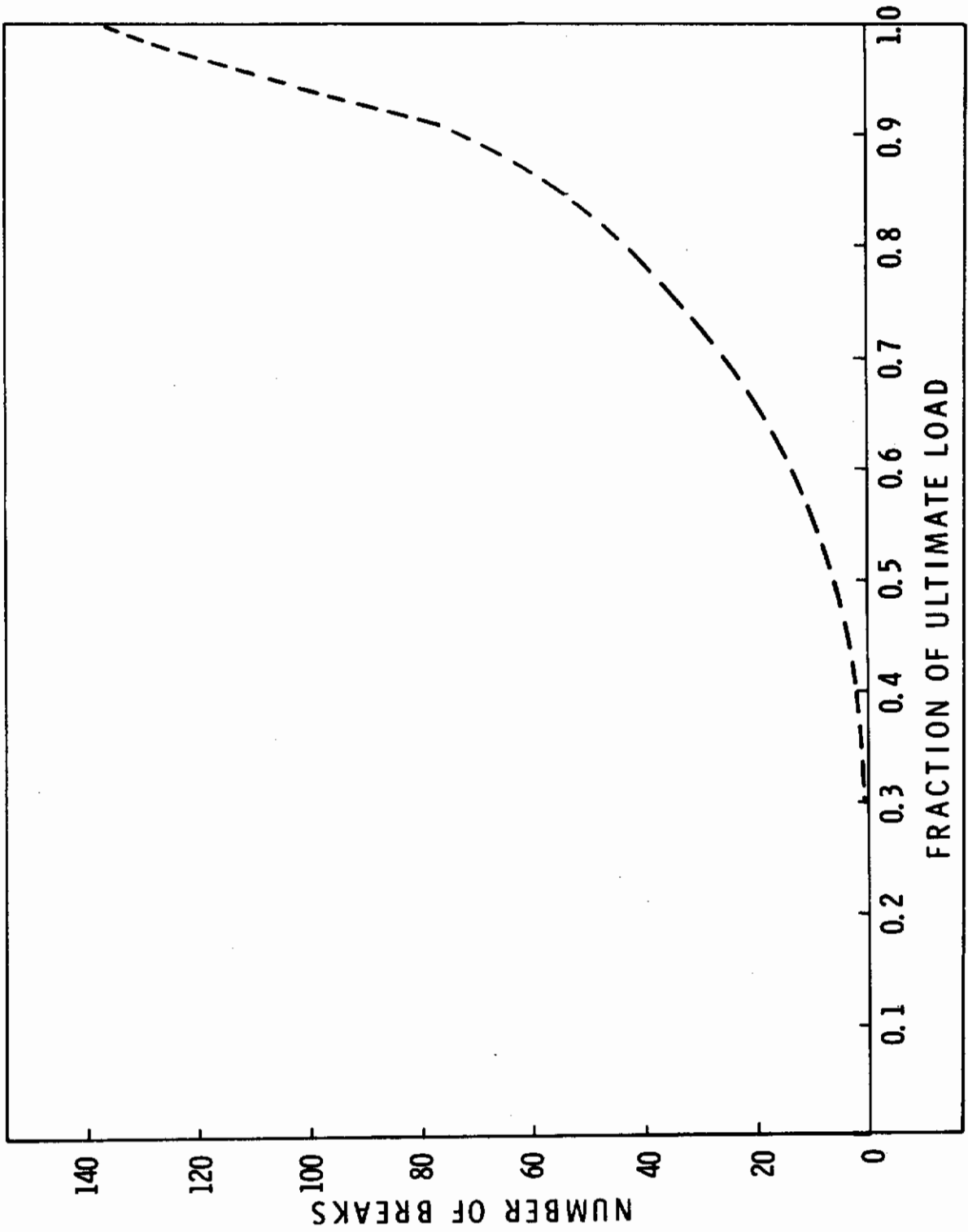


Figure 4. Load Fracture Vs Fiber Breaks

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Throughout this discussion it has been presumed that the mechanical properties of the fibers and of the matrix are truly those of the bulk material. It will be noted that the Young's modulus of 55×10^6 psi for bulk Al_2O_3 (single crystal or polycrystalline corrected to zero porosity) has been used rather than the static modulus measured on the fibers themselves, in excess of 65×10^6 psi. In the same way, the shear modulus for silver was calculated from its Young's modulus and Poisson's ratio.

There is no doubt that this oversimplification will lead to error if one of the constituents, but not the other, is significantly changed by its conversion to a fiber or its use as a binder.

For example, fine glass fibers are less dense, lower in modulus, and less hard than annealed glass of the same composition. When they are heated they tend to increase in density, modulus, and hardness; these changes are accompanied by a serious decrease in strength. It would be a serious source of error, therefore, to predicate the properties of glass fibers upon those of bulk glass. A similar decrease is observed when work-hardened metal fibers are annealed.

The problem is made more acute by the difficulties inherent in accurate measurements of the elastic properties of very thin specimens. Changes in dimensions, flaws, and variations in structure and surface must be observed by microscopic scanning. If flexural modulus is determined by observing the fiber as a cantilever beam, the average of deflections measured when the fiber is turned over should be used. The accuracy of measurement of the smallest dimension is, of course, critical.

Even when the fiber or whisker is in all other respects identical to the bulk material (as is the case with most metals and non-glassy substances), it is noted that the elastic constants of the fiber or whisker are higher. In some cases this effect is real; an example is the increase in modulus of steel wool or drawn metals due to work hardening.

A classical example of potential error in estimating elastic constants for fibers and flakes is inherent in their high surface to volume ratio. Where the surface properties can or do differ sharply from those of the bulk of the material, care must be taken to make certain that the surface condition of a fiber or flake is the same at the time of measurement as it will be in the composite. Siliceous glasses, for instance, tend to acquire a coating of chemically-bound water or hydroxyl ions very quickly after drawing; as a consequence, glass fibers are weakened appreciably with age (unless stored in a dry atmosphere), and this weakening effect is physically irreversible. When the glass fibers are combined with resins in a laminate, the water generated by condensation tends to be chemisorbed at the glass surface. Since this water film is far lower in modulus than the glass, it acts to reduce the stress transfer between particles by the resin.

To minimize this unwanted effect, the glass fibers may be treated, immediately after drawing, with a silicone or metallic coating; a second hydrophobic coating may also be used (vinyl resins and aluminum metal are examples). In such cases, the error in applying moduli based on the glass alone is appreciable.

A more consistent approach is that of rendering the fiber less sensitive to surface reactions. The familiar "E" glass owes much of its worth to its unusually low free-alkali content, and glasses in which relatively unreactive magnesium, strontium, or

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barium have been substituted for alkali elements are appreciably stronger in laminate form. In the grinding wheel industry, free lime or other dehydrating agents are combined with resins to absorb water which would otherwise attack the glass fiber reinforcement.

A similarly absorbed water layer forms on many oxide surfaces. On Al_2O_3 and ZrO_2 , however, this water layer can be driven off at temperatures insufficient to weaken the fiber; it is largely for this reason that the Al_2O_3 -Ag system has been used here to illustrate the elastic-plastic requirements of a high-strength laminate.

The Concept of Critical Binder Thickness

The fibrous ingredient of the laminate is not the only feature which should be scrutinized, however. It has been pointed out that the shear modulus of any substance appears to increase with decreasing thickness. (There is little doubt that Al_2O_3 -Ag composites containing more than about 50 v/o of alumina would be both stronger and more brittle, but that the temperature at which they would still support some load would be still higher than for those discussed here.)

This increase in shear modulus must be taken into account whenever the geometry of the composite is such that the matrix or binder cross-section is locally thin. While it is not presently possible to predict its magnitude, it is necessary to recognize its importance.

In classical mechanics, the shear modulus of an isotropic solid is given by:

$$G = E/2(1 + \mu)$$

where G is the modulus of rigidity in shear, E is the modulus in tension, and μ is the Poisson's ratio.

In the case of an adhesive layer bonding two plates of more rigid material, it is invariably found that both the strength and the rigidity of the joint are maximum for a given "critical" thickness of the adhesive layer. Thinner layers apparently do not adequately fill the joint or wet both mating surfaces, while thicker layers are limited by the strength and cohesion of the softer material.

This effect has been observed not only with organic adhesives but with glass-to-metal seals, brazing alloys, solders, and refractory mortars. A familiar instance is the excellent adhesion obtained between gage blocks or polished glass plates with a thin water film.

The most widely accepted explanation is one which recognizes the interfacial energy of wetting between the adhesive and the substrates. In a thick film the adhesive can deform in response to axial tension or shear on the plates, and its response to tensile stress will be dictated by its tensile and shear moduli. If the plates are plane and parallel, the thick adhesive layer will deform by a process comparable to necking.

As the adhesive fiber is made thinner, a point is reached at which the restraint against inward movement of the adhesive provided by its bond to the plates is sufficient so that necking cannot occur. The plastic flow associated with necking is thereby

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denied, and the adhesive exhibits a higher modulus and strength than are apparent in the thicker joints. A somewhat more complex explanation is required to explain the similar sensitivity of sealing glasses to thickness in the bonding of ceramics to each other or to high-modulus metals because of the requirement that the glassy layer be sufficient to wet and to dissolve both ceramic and metal oxide to establish an adequate interfacial bond, but the same general trend of increasing strength and apparent modulus is observed as the thickness of the glassy layer is reduced.

In essence, then, a very thin layer of material can be so modified by attachment to adjacent surfaces that it no longer behaves in an isotropic way. Under these conditions, Poisson's ratio ceases to have a real meaning, the shear modulus approaches the tensile modulus value, and the adhesive layer breaks in a brittle fashion in tension.

It is not generally recognized that the strength and apparent modulus of a composite can be so drastically increased by thinning the adhesive; the reason for this lack of recognition is readily apparent if one looks at the geometry of the laminates with which most investigators have worked.

In an assembly of close-packed uniform cylinders, the void volume is 9.3 percent of the total volume, regardless of cylinder diameter. In a perfectly packed system of uniform glass fibers and binder, then, the fibers will just touch each other when they occupy 90.7 percent of the bundle volume. At this level of binder (if it can be achieved), the binder thickness is certainly less than the critical depth necessary for stress transfer.

An increase in binder content can be regarded as adding an annulus of binder around each fiber. If d_f is the fiber diameter and $D-d_f$ is the distance between them, then:

$$V_f = 0.907 d_f^2 / D^2$$

from which the "minimum binder film thickness" $D-d_f$ can be calculated for different composite proportions and different fiber sizes. The results are shown in Figure 5 for all compositions of 2, 10, and 20-micron fibers.

Suppose that the "critical" binder film thickness is, for a given combination of fiber and matrix materials, close to 7 microns. It can be seen that for bundle composites of 20-micron fibers this is approached at $V_f = 50$ v/o, for 10-micron fibers at 31 v/o, and for 2-micron fibers at less than 10 v/o. In terms of establishing a similarity of binder behavior, then, the tendency toward comparing a number of laminates containing fibers of different sizes is unrealistic. More importantly, the report "strength" contributions made by the fibers will be systematically in error.

In most real bundle laminates, both the ideal hexagonal model and the close-packed cylinder model give inexact solutions, the first overestimating the stress-transfer matrix volume, the second underestimating it. For the Al_2O_2 -Ag composites, the hexagonal model is quite exact because most alumina whiskers are hexagonal in cross-section. For 10-micron fibers, the hexagonal model gives a binder thickness of 6.9 microns; the cylinder model 7.4 microns. The error is not large, and can be corrected for. It is completely overshadowed by the larger errors introduced when composites of differing fiber diameters are compared without recognition of the profound influence of binder thickness on stress transfer and strength.

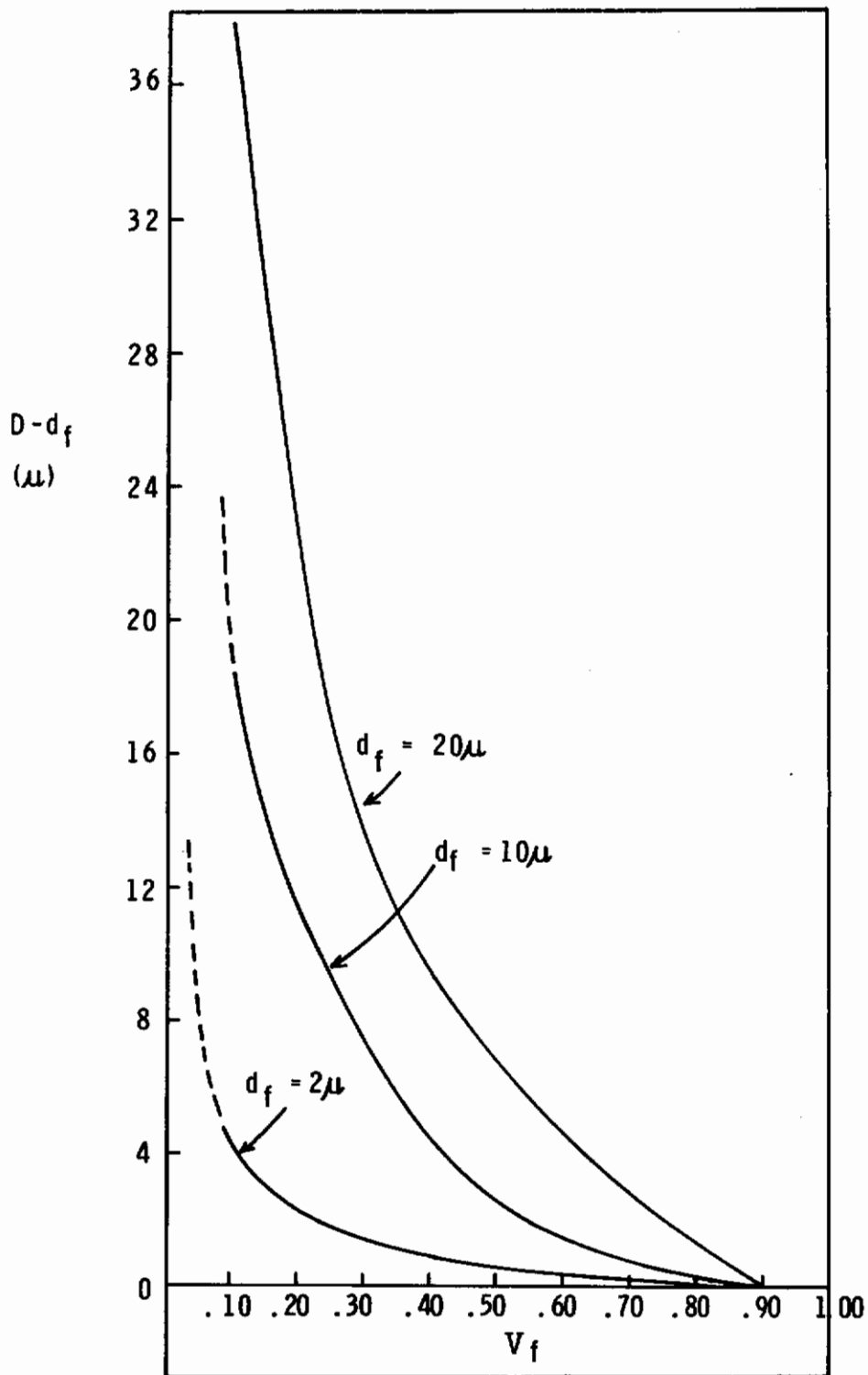


Figure 5. Minimum Binder Film Thickness Vs Volume Fraction of Fibers

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It should be noted that changes in fiber length can be made without introducing similar errors, even when the fiber length is only slightly more than that dictated by the E_f/G_m ratio.

It cannot be said that a bundle laminate has yet been made which optimizes the potential strength of the fibers or whiskers now available. It has been shown here that the Al_2O_3 -Ag laminates possess a more favorable balance of elastic properties at about 1000°F than at room temperature; at that temperature, however, the silver is beginning to weaken. To provide thinner silver stress-transfer volumes requires finer whiskers or lesser proportions of silver in the matrix; either or both of these should lead to the development of stronger composites. The substitution of Ni for Ag will, of course, improve high-temperature strength but will add to low-temperature brittleness.

A similar situation exists in most fibrous glass-resin laminates, in most of which the E_f/G_m ratio is even less favorable than that of the Al_2O_3 -Ag system. In most instances, the inordinately large resin proportion is recognized, and intensive work has been done to develop "high-modulus" resins to achieve more favorable E_f/G_m ratios. Since most of these resins are relatively more viscous than those of lower modulus, the results have not been encouraging. It seems reasonable, therefore, to pursue an opposite course by working with resins of minimum viscosity in laminates containing smaller resin proportions if maximum strength is to be achieved together with high modulus.

Design of a Laminate using SiC Fibers or Flakes

Starting with the knowledge that the Young's modulus of beta-silicon carbide (the most easily vapor-deposited form) is nearly 70×10^6 psi at room temperature and about 60×10^6 psi at 2000°F, one can postulate that whiskers or flakes of it having any dimension smaller than about 12 microns should be capable of developing tensile strengths comparable to that of the 10-micron alumina whiskers - about 10^9 psi. Thinner whiskers or flakes should, of course, be stronger, but more difficult to handle. The advantage of SiC over Al_2O_3 is, of course, its higher potential strength to weight ratio.

In selecting a suitable matrix material, the processes of mixing, forming, and curing must be considered. Silicon carbide after long exposure to air is normally coated with a thin film of amorphous SiO_2 . This can easily be removed by washing with dilute HF solution. Other pretreatments may be suggested according to the matrix material used.

In view of the fragility of the whiskers or flakes, it would appear that the latter might best be handled from a slurry or suspension of volatile fluid. For shapes of revolution a centrifugal casting process can be used; for complex shapes vacuum forming or slip casting in a porous mold would appear to be appropriate. In all cases, the flake geometry will insure parallel deposition if the rate of evaporation of the suspending fluid is balanced properly with the rate of flake deposition so that bubbles or blisters do not form.

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For whiskers or fibers, a means of insuring parallelism is needed. Mechanical orientation by transferring them from a longitudinally stretched belt to a tacky surface may be required. Longer whiskers will be more easily aligned.

Forming the laminate must involve methods which will permit optimum packing with minimum breakage. Parallel tape layups and screen or film transfer techniques can be used for whiskers; the flakes may be combined with resin or with inorganic colloid binders from the slurry stage.

The importance of curing shrinkage and thermal coefficient of expansion has already been discussed. The latter increases in influence with increases in modulus and, of course, with broadening of the range of temperatures to which the composite will be exposed.

The selection of the matrix material is critical; the criteria to be considered must include the following:

1. The mechanical properties of the matrix must be such that the high strength and modulus and low density of the fibers or flakes will be exploited. This criterion involves theoretical and experimental examination of the matrix to establish its modulus to temperature relation and its modulus to thickness relation (see later discussion of evaluation techniques).
2. The chemical characteristics of the matrix must be such that it will survive the necessary processing steps, will wet and adhere strongly to the aggregate material without undue interdiffusion at maximum temperatures anticipated, and will maintain its characteristic properties under storage and service conditions.
3. Candidate matrix materials should be considered on the bases of density, availability, cost, and processing requirements.

For use with SiC, a variety of matrix materials are worthy of consideration. The listing below is not exhaustive but is intended to indicate such a selection process.

- a. Materials of maximum refractoriness: Refractory metals (other than Mo and W), Si_3N_4 , BN, SiC foam, carbon and graphite, mullite, alumina hydrate colloids, zirconia colloids, reconstituted micas, Au, Pt, etc.
- b. Materials of maximum modulus to density ratio; beryllium, magnesium, and aluminum alloys and light-metal intermetallics, titanium and vanadium alloys, carbons, graphite, BN, AlN, AlP, etc.
- c. Materials for maximum strength to density ratio composites: Cr, Si, Co, Ni, Ti, V, Rh, Ag, Pt, Au, Al, Mg (doubtful) alloys, over-cured polyesters, fused silica, FA resins, glasses (CaO.FeO.SiO_2), keratin proteins, polyamide-epoxides, reconstituted proteides, polyimides, phenolics, etc.

It will be obvious that the application for which the composition is intended will bear most strongly upon the selection of candidate matrices. The list can further be shortened by eliminating materials which can be emplaced only by techniques which would damage the aggregate material, SiC in this instance.

Having selected several candidate matrix materials, experiments to establish wetting, adhesion, interdiffusion, and mixing, forming, and processing techniques may be applied as required.

TESTS AND PROPERTY DATA FOR COMPOSITES

For the simplest structural applications, the evaluation of critical properties is usually performed through well-established testing techniques. Of these perhaps the best-known are tests for tensile, flexural, compressive, and torsional strength and modulus values. Somewhat more complex tests for creep, fatigue, notch toughness, impact-resistance, thermal-stress resistance, and hardness have been developed as needed.

In almost all cases, these tests have been developed for and applied to relatively ductile substances. It has only recently been recognized that, unless applied with great care, these tests (and the calculated results from them) are of doubtful value. (It is understandable, for instance, that the strength of a brittle material should be sensitive to size effects, but the finding that elastic moduli are also affected by size suggests that either the finding, or the concept of modulus, should be reviewed)

The importance of stress relaxation mechanisms (mass diffusion, dislocation generation and movement, stress-induced polarization, vibration coupling, and thermal effects) has been recognized in connection with strain-rate sensitivity, fatigue, and notch toughness. The absence of many of these mechanisms in brittle materials is not so widely recognized.

It is often assumed, since virtually all structural composites consist of high-modulus members or aggregates joined by material of lower modulus, that the composite can be treated as if it were homogeneous and ductile. The discussion earlier in this report will have served its purpose if these assumptions are hereafter questioned.

There appear to be no easy, shortcut approaches to obtaining an understanding of the mechanical behavior of these composites. Attempts to obtain meaningful measurements of such properties as elastic moduli and density can be justified for preliminary screening, but the uninhibited use of these property values for evaluation among materials of different structure has undoubtedly obscured some of the principles discussed here.

A single test can be used to illustrate this. Perhaps the most common procedure for measuring the strength of brittle materials is the familiar "modulus of rupture" or "bend" test.

This test, hereafter called the flexural test, consists simply in loading a beam of the material at one or more points between two supports. The maximum stress at fracture is defined as the "flexural strength". This maximum stress is defined as:

$$S_{\max} = \frac{M_c}{I}$$

where M is the bending moment at the point of rupture, c the distance from the extreme fibers (where failure is initiated) to the "neutral axis" of the beam, and I is the moment

of inertia of the cross-section about the neutral axis. For a rectangular beam of thickness d and width b supported over a span L the formula:

$$S_{\max} = \frac{3PL}{2bd^2}$$

is frequently used, P being the failure load imposed at the midpoint of the specimen.

It is not always recognized that this formula presumes the following:

1. The original transverse plane of the specimen from which moment calculations are made remains a plane, normal to all longitudinal fibers during bending. This assumption is justifiable only if deflection at failure is very small.
2. The material of the beam follows Hooke's law of linearity between stress and strain so that the stress distribution is linear across the bent beam, and is everywhere directly proportional to the distance from the neutral axis. This assumption cannot safely be made for inhomogeneous materials and is certainly invalid for the anelastic behavior of the matrix material.
3. The stresses on the beam are pure bending; that is, neither shearing, torsional, nor tensional forces are additionally applied. It can be shown that unresolved shearing forces exist as a wedging action in ordinary mid-point loading, that torsional forces exist unless unusual care is taken to insure against them, and that tension forces on the loaded surface are introduced by frictional restraint, introducing a bending moment opposing that being applied and measured. While any or all of these non-bending stresses can be more or less compensated for, the "corrected" values are always open to some question and are seldom reported in the literature. In some cases the corrections are large; for a coefficient of friction between beam and supports of 0.4, a correction of 12 percent should be applied to the ultimate fiber stress for 3-point loading, and for 4-point loading the calculated stress should be reduced by 16 percent.
4. The material behaves equally under tension or compression; its compressive and tensile elastic moduli are equal. It is in this respect that most fibrous composites do not accord with the idealized, homogeneous, isotropic substances for which the flexural strength equation was developed.

It is not the writer's intention to suggest that flexural testing be discontinued; indeed, bend testing is frequently the most practical and direct way of comparing a variety of solid materials with each other. It is strongly recommended, however, that bend tests in which the deflection at failure is several times the thickness dimension of the specimen be regarded with suspicion, and that "ultimate fiber strength" values obtained in bending (after correction for wedging and friction) which are much larger than the highest uniaxial tensile strengths for that material should be a cause for reappraisal of the testing technique used.

It has been noted earlier that in composites, each of the components is likely to affect the other, leading to "residual stresses" affecting the stress situation at the time of testing. It has been shown (Reference 25), for instance, that in a glass-reinforced resin composite, the curing shrinkage of the resin is opposed by the high-modulus glass, leading to residual tension in the resin layer adjacent to it. In a uniaxially stressed bundle composite, the superimposed tension load will be added to this longitudinal residual stress, and the bundle may be weakened.

In flexure, however, the axial compression on the loaded side of the specimen will act in the same direction as this residual stress; the combined compression along the fibers may introduce bending moments in them sufficient to cause buckling and delamination.

The danger, then, in applying conventional mechanical tests to laminates lies not in the data obtained but in the possibility of application of principles and interpretations of doubtful validity. Until an adequate body of knowledge can be applied to such data they should be acquired with unusual care and applied with extreme caution.

SUMMARY

It is doubtful that the state-of-the-art of the macrolaminates discussed here will benefit from increased theoretical studies in micro-mechanics. In the simplest of systems, a longitudinally reinforced ideal beam, the application of a bending load can be shown to introduce stress and strain distributions in fibers and matrix which are presently beyond rigorous mathematical solution. The fibers are multiple elastic inclusions in the elastic-plastic matrix, and are subjected to load only because of their interaction with it. The loads upon them, and their contribution to the elastic properties of the beam, depend on their elastic properties, their size, number, distribution, shape, and length as well as on the elastic properties of the matrix and of the bond between them.

An attempt has been made in this discussion to show that many of the principles needed to isolate and define the most important parameters for composite development can be obtained by precise experimentation with carefully selected materials. It is strongly recommended that such work continue to be supported.

It has been amply demonstrated that brittle materials are less forgiving toward minor errors in alignment, finish, dimensional tolerance, and discriminating care in selection, handling, and mechanical loading than are ductile ones. It is strongly recommended that the whole practice of mechanical testing (and the calculating and reporting of property data) be critically reviewed both for brittle materials and for composites containing them. As a part of this critical review, certain techniques and practices should be advanced as tentative standards.

It cannot be too strongly emphasized that the probability of achieving an optimum composite system will be greatly improved through recognition of the following principles:

1. A designed composite consists of two or more materials which, in interacting with each other, permit the whole to achieve some property or properties not possible with either material separately. It follows that any composite must and should be tailored toward the performance of a specific task; it also follows that there is little probability of optimizing any composite system for a wide variety of applications.
2. The simple laws of volume proportionality, although applicable for characterizing such properties as density and elastic moduli, are not applicable to composites in which the shape, size, size distribution, and/or the orientation of either or both components can be varied. It follows that each of these must be adequately defined and controlled.
3. Interactions between the two or more components of a composite may be sufficiently important to negate or reverse the behavior expected on the basis of bulk behavior. It is only infrequently true, for instance, that the mechanical behavior of a brittle polycrystalline solid can be predicted from or related to the mechanical properties of the single crystal.

4. For achieving maximum strength, the high-modulus material must itself exhibit great strength. This is greatest when one or two of its physical dimensions is as small as possible, certainly less than 12 microns (one-half mil).

5. To exploit the high strength of the strong component, it must be evenly distributed in a matrix with which it is chemically and physically compatible. A principal requirement for the matrix is that it can be formed or shaped in the presence of the strong component.

6. Geometrical considerations in any laminate may dictate the system to be employed for optimized properties. For example, no system will exceed the well-packed fiber bundle for uniaxial tensile strength, but if high shear strength or biaxial strength is required, a flake laminate (or combined flake and fibrous laminate) will be better.

7. The illogic of expecting a laminate to develop higher strength in two or more dimensions than in one must be recognized.

8. As the proportion of matrix material is reduced the effective shear modulus of the matrix increases. It follows that great benefits can be derived from decreasing the matrix proportion as well as from developing matrices of higher modulus.

9. Techniques for aligning fibers, whiskers, and flakes require development if optimum composite structures are to be achieved. The infiltration of these with matrix will always be difficult. Efforts should be made to precoat the aggregate with matrix prior to consolidation to insure optimum matrix film thickness, and efforts to obtain fibers, whiskers, and flakes of optimum and controlled dimensions are vital.

10. The "ideal" composite will undoubtedly be achieved when the aggregate fibers or flakes can be precipitated or grown in situ in the laminate. Mechanical working of the mixture to obtain proper aggregate shape and alignment will undoubtedly be required.

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