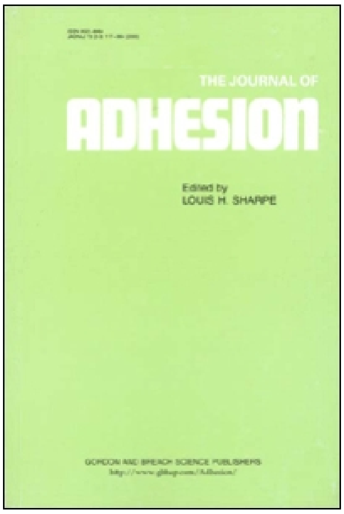

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Some Thoughts About the Mechanical Response of Composites†

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Some criticism is directed at current approaches to composite response which do not include a mechanical model of the region in the vicinity of the interface. An inter*phase* model based on inter*face* roughness is suggested.

INTRODUCTION

What I intend to do, in this brief introduction to the series of papers on Polymeric Composites, is simply to say something about some approaches to composites or composite structure behavior which I think we are neglecting.

There are really two quite different, extreme approaches to composite mechanical response at the present time. There is the approach which emphasizes the role of the interface between the different elements of the composite or structure as the determining factor in its response. By nature, it is a molecular or chemical approach. It attempts to link a change in composite response directly and solely to a change in molecular structure *at* the interface. It says that molecular structures *at* the interface which change the interaction energies between the two phases of the composite act directly and solely to change the mechanical response of that composite. This approach, therefore, assumes that it is possible to identify, isolate and assign direct, simple cause

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and effect relationships between interfacial structure and bulk mechanical response in a composite. It is what we might call an interface intensive approach.

The other extreme approach is that in which the role of the interface is, in essence, ignored and an attempt is made to understand composite response in terms of the bulk response of the two phases and a geometry. In this approach, the interface is not really totally neglected. It is simply relegated to a status in which it is one of two types—well bonded or poorly bonded. If it is a well bonded interface, meaning that failure does not appear visually to occur there, then it is considered not to enter into composite response and its presence is neglected. If it is a poorly bonded interface, meaning that it looks as if the failure occurred there, then the response is considered to be determined by the interface and not by bulk mechanical properties.

It is quite evident, at least from the descriptions of the extreme positions as I have presented them, that there are aspects of similarity between the two approaches which center on the interface. The main dissimilarities are those which one would expect between the approach of a chemist—molecular structure, interaction energies, bonding—and of a mechanical engineer-macroscopic response and fracture.

The questions which arise from all of this are:

1) What are the logical shortcomings of the two approaches just discussed?

2) Are there aspects of the two approaches which might be combined and/or modified to provide a more useful, general approach?

3) Are there some new approaches worth looking into?

APPROACHES TO COMPOSITE RESPONSE

Let us look at these in order. What are the major shortcomings of the chemical approach? Well, it seems to me that if one is going to point to the interface as the source of strength or weakness in a composite, one is going to have to come to grips with the question of "how strong" and "how weak." That is, one is going to have to develop means to describe quantitatively the relationship between interface structure (or energetics) and composite mechanical response, including ultimate strength, in whatever mode of failure. Furthermore, if one is going to point solely to the interface as the source of strength or weakness, then one is going to have to develop strong arguments as to why one can neglect the bulk response of the elements of the composite in arriving at an explanation of its behavior. Until such time as means are developed to relate postulated interfacial structures and interactions quantitatively to composite response, one has to consider that such approaches are, at best, a qualitative rationalization of observed behavior. In all fairness however, I must add that these approaches seem to have proven useful, e.g., in providing guidance as to the types of materials likely to be useful as coupling agents. I say *seem*, because one cannot be sure how much guidance occurred before success, relative to rationalization following success.

Approaches which concern themselves solely with the bulk behavior of the elements of the composite are limited in their ability to produce true descriptions of response because, like the interface model, they focus on a single aspect of response. As we have pointed out,[1] one cannot always consider adhesive joints to be "simple" composite structures of adherend 1, adherend 2 and adhesive—that is, one cannot always consider them to consist only of three bulk solid phases, each characterized by its own single set of material constants. The reason is that solids are generally themselves composite layer structures and this can modify their mechanical response when a layer (e.g., a surface layer), having response different from the bulk, is an integral part of a composite structure (or a composite) containing such a solid. We know that such boundary layers exist. They are oxides on metals. They are transcrystalline regions in semi-crystalline polymers. They are diffusion regions between polymers solidified in contact with each other. And so on. We know that such layers differ structurally from the bulk materials with which they are in contact. The must, therefore, differ mechanically. If they differ mechanically, and we are concerned with mechanical response of a composite, we simply have to concern ourselves with their influence on response.

Let me say very clearly that I am not supporting the pervasive use of the notion of weak boundary layers. It has been used indiscriminately and despite the fact that sometimes it involves a circular argument. I would like to emphasize, however, that boundary layers do exist, that they can influence composite structure behavior and I would like to suggest that perhaps some attention ought to be given to how best we can attempt to describe and understand them.

It seems to me that by discarding the notion of an inter*face* in thinking about composite structures (or composites) and substituting instead the notion of an inter*phase*, a region of more or less variable composition, and therefore, in general behavior, intermediate to the two bulk contiguous phases, we might be able to make some headway. First of all, such a notion is probably closer to the real situation in an actual system than the notion of an interface, which constrains one to think in terms of a sharp boundary—a geometric plane or a smooth curved surface between two contiguous solid materials—when we know that the boundary between two contiguous technological solid phases is seldom flat to more than a few thousand angstroms or so. Let us examine the mechanical consequences of this roughness.

A ROUGHNESS MODEL OF THE INTERPHASE

Consider a model of two solids *A* and *B* in complete contact. *A* is initially solid and *B* is brought into contact with it, while in the liquid stage, and caused to solidify without shrinkage and without residual stress. Assume that the roughness of *A* can be characterized by a series of planes of irregular shape, joined at their edges, arranged at random angles to each other, with dimensions varying from perhaps a thousand to several thousands of angstroms. The important aspect of the model, Figure 1, is that the geometry

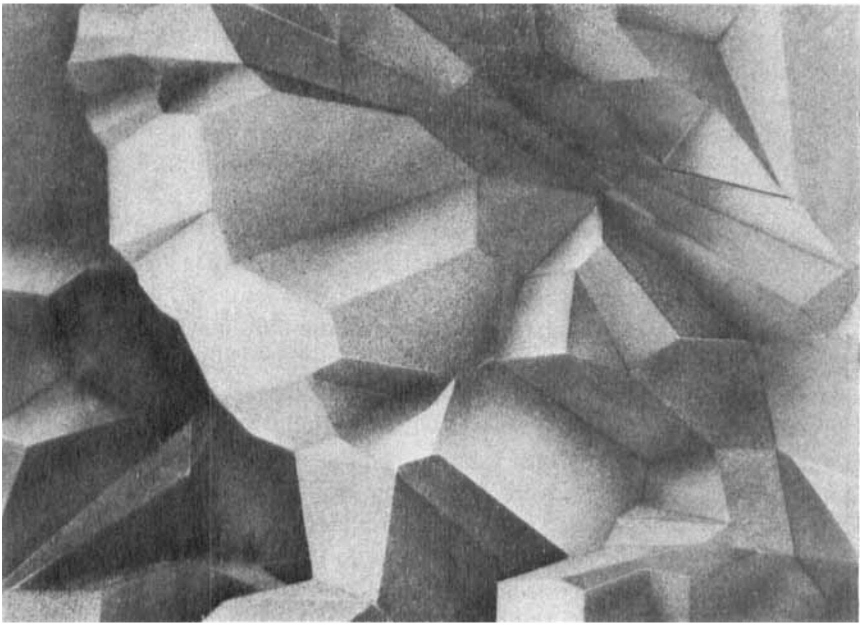

FIGURE 1 Roughness model.

changes more or less abruptly in every direction along the surface of *A*, and that the extent of individual planes is large enough so that the mechanical response of aggregates of molecules of *B* attached to any plane and associated with it can be considered as characteristic of bulk response of *B*.

Consider now that an external load is applied to a macroscopic section of the *A–B* composite. The external load produces, in the interface region, local modes of loading, therefore stresses and strains, which are determined, in part, by the *local* geometries. Since these local geometries vary, the *local* stresses and strains will vary. The interface region of the materials will, therefore, deform and, perhaps, fail in a manner characteristic of the local geometries and constraints and not of the far field materials. In other words, there is possible a boundary layer, resulting only from the roughness, which is forced to behave in a manner different from the bulk.

The situation is, of course, far more complicated than I have pictured it. For example, on the scale that I have been talking about it is possible that stress fields in one local geometry interact with fields in surrounding geometries, inducing material behavior which, because of the scale of deformations induced on the constraints, simply may not be found in macroscopic sections. I suggest that perhaps some computations should be done on this model to see if it is capable of leading to sizeable effects. At the very least, it is a reasonable model of the interface—more correctly, the interface region or interphase—which, in principle, is capable of analysis. This is more than can be said of any other interface model. One should realize, however, that because of the small volume of the region in which the effect is likely to occur (relative to the macroscopic dimensions of any test specimen) it probably cannot be detected satisfactorily in a stress-strain experiment. The effect would only be expected to show up in failure experiments because, in those, changes in the *local* stress distribution could influence the failure process markedly and result in measurable changes in the average breaking stress of a test specimen.

Incidentally, I might mention that I think the model of interphase behavior which I have been discussing may be worthwhile exploring as an explanation of the effect of substrate roughness on joint strength of bonded structures, on the adherence of coatings and on the failure behavior of filled systems.

So far, I have discussed interphases as being thin discrete layers different from bulk materials in a composite structure and as thin layers which, because of a scale of roughness in an interfacial region, may also behave differently from the bulk.

There is a third case of materials behavior near an interface which, perhaps, we should consider. If we join materials of different moduli and Poisson's ratios and apply a load to the joint, we of course find that a stress concentration develops at the edges of the joint. This stress concentration will be higher the greater the difference in moduli and Poisson's ratios, no matter what the mode of loading. Large stress concentrations induced by the differential strain of the two materials can cause a composite to fail at loads which produce mean stresses, over the joint area, far below characteristic failure stresses for the isolated bulk materials of the structure. Additionally, the higher modulus material would be expected to exert some restraint on the lower modulus material, this restraint being localized near the interface. This localization of stress could cause the lower modulus (the "weaker") material, to fail in a local region near the interface, creating the illusion of an interfacial or "weak boundary layer" failure, although it may be neither.

The point of all this discussion of interphases is simply to draw attention to their existence and to show that it may be possible to work them into a theory of composite response.

SOME FINAL THOUGHTS ABOUT COMPOSITE RESPONSE

I am sure it is well known that the overall response of a composite material or structure is not simply determined but is a function of a complex of interacting factors. Therefore, to approach the development of an understanding of its response by focussing on a single factor is not productive unless one can justify eliminating other factors from consideration. Unfortunately, this is not very often, if ever, possible.

It seems to me that we are failing to face the fact that a composite or composite structure is a system. The problem of response which faces us is a systems problem and it ought to be treated as such. That is, we need to be concerned with describing, explaining and finally understanding how the responses of the individual, not necessarily independent, parts of the system interact to determine response of the system as a whole.

It seems to me also that we need to be able to describe the system behavior phenomenologically, and be sure of that, before we can proceed to make sense of system response in a fundamental way. In fact, we may very well have to proceed through a hierarchy of levels of aggregation before we can finally reach the fundamental or molecular explanations of composite response which so many workers seem to be searching for. What I am saying is that morphologies of one or more levels of scale may intervene between the molecular level and the macroscopic level for each of the materials in a composite. If this is so, then each of the materials has to be viewed as being itself a composite material, the response of which has to be described. But it is generally true that the description of the response of these materials is no better than phenomenological. Then how can it be reasonable to propose fundamental explanations of composite response when one does not have a fundamental understanding of the response of the elements of the composite?

Since I have been critical, I should also have something constructive to offer. Let me suggest, if you will, one way in which I think we ought to proceed in order to get at answers to the composite response problem. First, we seem to know most about the phenomenology of materials. Our first step, therefore, ought to be to develop sufficiently realistic models and techniques so as to be able to describe, on a consistent basis, composite mechanical response in terms of bulk properties of the materials comprising the composite, an interphase region, a geometry and perhaps some other mechanical elements that I do not know about. Until we achieve a facility at this kind of thing we stand very little chance of learning something fundamental about composite response. If we cannot describe behavior on a macroscopic level, how can we isolate, identify and assign causes and effects on a fundamental level? For example, how can we say things in detail about the mechanism of action of coupling agents in changing composite mechanical behavior, when we do not even consider, much less use, a mechanical model of the interface region in arriving at our conclusions? It seems to me that we are going to have to work ourselves down the hierarchy of structure, determining how each successive level determines response, rather than to try to bypass the hierarchy and attempt to relate molecular structure directly to mechanical response. I feel that only if we proceed in a stepwise manner can we achieve a deep, fundamental understanding of the relationship between interfacial bonding and fracture in composites, if indeed a meaningful one exists.

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