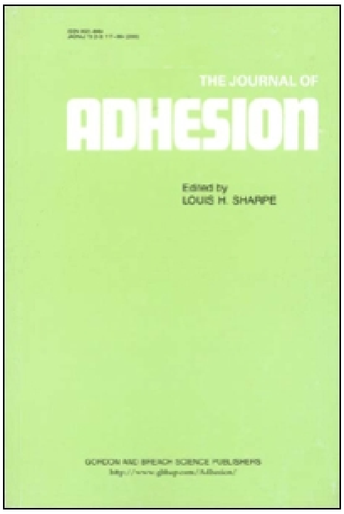

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The Effect of Scrim Material on the Elasticity of Adhesives Under Bond-Normal Tension

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NOTE

The Effect of Scrim Material on the Elasticity of Adhesives Under Bond-Normal Tension

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KEY WORDS Adhesive materials; elastic response; non-linear response; scrim dewetting; damage accumulation; isotropic behavior.

INTRODUCTION

An essential ingredient in the stress analysis of bonded joints and the estimation of potential failure occurrence are the (thermo) mechanical properties of the adherends and of the adhesive. These properties are universally determined through standard engineering tests on uniaxial tensile coupons.

Structural adhesives are commercially available with a scrim or carrier material which has the multiple purpose of serving as a facilitator for handling, as a spacer in the bond forming operation, and as an escape guide (along the fiber-adhesive interface) for residual solvent or other cure-related gases to prevent porosity of the bonded adhesive. This carrier material comes in different forms, usually as a random mat of short fibers on the order of several inches in length, or as a knitted material; in either case, it gives the adhesive certain characteristics of a composite material. Because the carrier material is initially located in the midplane of the adhesive film, the lamination of the film into test specimens for property determinations and subsequent testing always generates tests in which a substantial portion of the fibers are oriented along the tension direction. However, in use the adhesive is laminated between two (metallic) adherends and the forces tending to destroy the bond are applied in shear and/or tension *across* the bond, that is, normal to the plane in which the fibers are located. One would naturally question, therefore, whether the elastic characterization of an adhesive in the plane of the scrim material is appropriate for stress analysis purposes.

The carrier fibers are usually made of nylon or polyester, materials that are not well known for their excellence in adhesion to other polymers. Thus, one readily appreciates that stressing the adhesive-scrim assembly normal to its plane may elicit a response that is different from that encountered when the composite material is tested in-plane and more or less parallel to the scrim fibers: In the latter case, one would expect that even in the event of imperfect bonding between fibers and matrix there exists sufficient load transfer to provide a load-carrying contribution by the fibers. However, if "less than perfect" bonding exists between fibers and matrix the generation of disbonds should elicit a significantly different stiffness response for the scrimmed and unscrimmed material, because after fiber debonding the adhesive becomes essentially a solid containing numerous, elongated cavities. An additional consideration regarding the proclivity to develop fiber-matrix disbonding is the fact that fibers tend to shrink more on cool-down from processing temperatures than the adhesive material by itself; this differential volume shrinkage would foster unbonding prior to loading or under low loads.

In view of this situation it appears reasonable that one should examine the potentially anisotropic properties of such scrimmed adhesives. However, in our exposure to structural adhesive work we have not, on the one hand, come across a single attempt to examine this question; yet, on the other hand, we have routinely experienced stress analyses for bond failure investigations which treat the adhesive layer without question as an isotropic medium. Should adhesives exhibit substantially anisotropic behavior (in disagreement with popular assumption) then it would be high time that such properties be accounted for in future stress analyses of bonded joints. On the basis of this possibility it appears, therefore, that at least a limited study is in order to assess the degree to which such an isotropy assumption may be justified. This observation provides thus the motivation for the following work. At the same time it should be noted that the subsequent work does not fully address the question of anisotropy in the sense of providing measurements of the, say, orthotropic characteristics of the "adhesives." The objective of this report is to document a more limited material response which addresses succinctly the question of fiber unbonding and interfacial damage under stress. Of particular interest then is whether the introduction of fibers into the adhesive matrix reduces the stiffness of the composite transverse to the scrim plane. If such reduction does occur to a significant level it would seem appropriate to begin characterizing "adhesives" as orthotropic materials.

SPECIMEN PREPARATION

It is clear that for comparison purposes it was necessary to prepare specimens that contain scrim material and others that do not. In order to make meaningful measurements it is desirable to deal with fairly large or standard engineering-sized specimens. For this reason we opted for specimens of the shape and typical

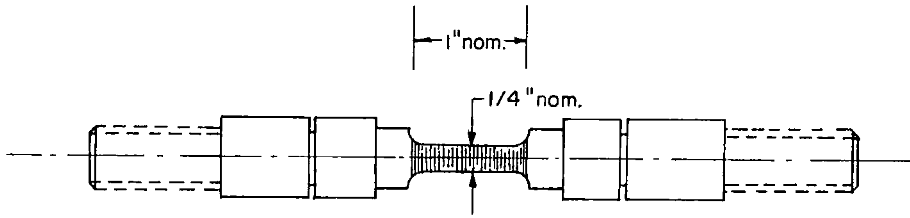

FIGURE 1 Geometry and nominal (planned) dimensions of specimen.

dimensions shown in Figure 1. We note that these dimensions are typical, because the manufacturing process did not always allow close control over them; this fact posed no serious problem because the dimensions could always be measured after machining.

An absolutely mandatory requirement for successful specimen manufacture was the elimination of air and other gases generated prior to and during the cure process. This fact required the following manufacturing process involving both evacuation and pressure. Figure 2 shows the components from which the specimens were prepared: End rods (a) were surface-prepared (phosphoric anodize etched) for good bonding to the adhesive. Discs were cut from five-mil (0.127 mm) thick adhesive sheets and stacked to fill the central portion of the tube (b) which was provided with a valve for connection to vacuum prior to and during heating.† Insertion of the end rods into the tube under a compression of

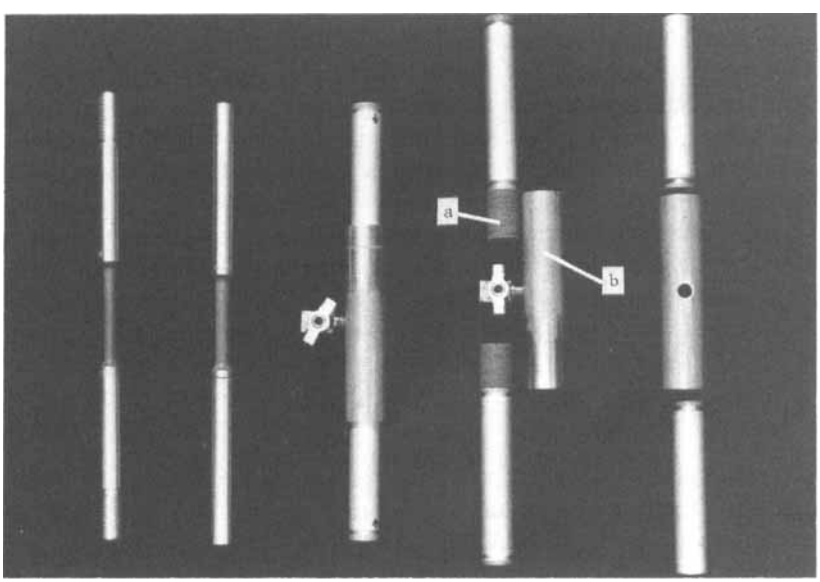

FIGURE 2 Specimen preparation process. (a) End rods, ultimately machined with screw thread, (b) Forming tube with vacuum valve.

† The assistance of Dr. W. B. Jones in the construction of the mold is gratefully acknowledged.

2700 psi (18.6 MN/m^2) for the time and temperature recommended by the adhesive manufacturer effected a solid bond between all components. The specimen, as shown in Figure 1, was then machined from this assembly. Since ultimately all specimens were stressed to failure it could be determined which ones possessed internal defects.

The use of such a high manufacturing pressure raises an important question regarding "easy" debonding of the matrix from the fibers, which was mentioned in the introduction as the possibly primary contribution to the mechanical response across the adhesive layer. Under such high pressures it is possible to effect stronger bonds between fibers and matrix than under pressures of three or four atmospheres,[1] so that the answer to the question posed in the introduction may not be fully given. However, inasmuch as there is really not much choice in manufacturing tension specimens without significant porosity in such a relatively large volume of adhesive, we need to accept this potential limitation to this investigation.

Since this study addressed only the deformation response, and not the failure behavior of the materials, those specimens which showed visible internal flaws were eliminated from the data base under the assumption that the other specimens yielded reliable deformation response data. In this way, specimens without and with scrim normal to the specimen axis were manufactured. Five "materials" were studied, namely unscrimmed FM73 (FM73U), with a random mat of nylon scrim (FM73M), and the standard material with a nylon stocking weave scrim cloth (FM73)† as well as AF163 which contained a non-woven scrim material (AF163-2M) and an unscrimmed version (AF163-2U). Scrimmed and unscrimmed samples were prepared in identical fashion.

It should be pointed out that machining the specimens from blanks was a delicate matter. For this reason normal finishing operations to generate very smooth surfaces were not followed. Thus, specimens would often show (very) small circumferential toolmarks that would be considered of possibly detrimental influence in a study of "ultimate properties" if one were to choose this test geometry for failure characterizations. However, in the present study which emphasizes the deformation/stiffness characterization it is felt that these tool marks are of no relevance.

SINGLE LAYER AND MULTILAYER COMPOSITE

It is of interest to note that the chosen test geometry employing many layers of adhesive does not conform to the normal use of adhesive films which are typically in the form of single layers between adherends. In raising the question whether this difference in the test geometry can imply potential differences in the

† The author is indebted to Mr. Ray Krieger of the American Cyanamid Company for supplying the FM73 materials in laboratory quantities. Dr. Alphonsus Pocius of the 3M Company was kind enough to assist in procuring the requisite amounts of the AF163 adhesives.

measured properties is tantamount to questioning the very existence of material properties; material properties are quantities that are independent of the size of the material element under consideration, unless microstructural considerations enter. In the present case, the dimension of a single adhesive layer is comparable to the thickness of the scrim fibers. Thus the question of response differences in the test geometries and in an adhesive bond is not without justification. Although there are apparently indications that the adhesive stiffness in an adhesive bond can be somewhat different from that encountered in bulk testing, it has never been quite clear whether such differences are due to the differences in processing history imparted to adhesives in the manufacture of different geometries. While we have information from within our own laboratory on differences in fracture behavior of polymers in an adhesive *versus* bulk application, we have no such knowledge regarding the stiffness characteristics; in view of the results to be presented later it would seem, however, that they would not represent first-order effects.

TEST PROCEDURE AND DATA ACQUISITION

Tests were conducted in a standard way with an Instron Tester at a single cross head rate of 0.254 mm per minute at room temperature (23°C). The uniaxial strains were determined with a clip-on Instron extensometer possessing a 25.4 mm gage length. One might ask whether the viscoelastic properties of the adhesive might not induce time dependent effects that could influence the results in a significant way. Although that may be the case, there were two reasons why the study was not extended to study explicitly time or rate sensitive phenomena.

While time dependence of structural response is an important aspect of failure/life estimation, time or rate dependence is often a phenomenon that is superposed on other material behavior. Thus, physical characteristics of the type investigated here at one time scale are, most likely, also evident at other time scales. We would expect that such rate dependent effects would be dominant when the adhesives are heated to temperatures where they are not normally used for structural purposes. Alternately, such time dependent effects would be expected to be significant under load levels where the nonlinearly viscoelastic properties come into play; this load range would imply situations where an adhesive is close to the failure point, and that behavior is, perhaps, studied more appropriately as part of a damage/failure investigation. Here we are interested strictly in the mechanical response as it may affect conventional stress analyses. As a corollary, we note that time/rate dependence is absent at the "lower" temperatures indicative of many use-conditions so that the present results should be indicative of the effect of scrim material on the deformation/stiffness response of structural adhesives. Moreover, it will be seen that there is surprisingly little difference in the stiffness between the scrimmed and the non-scrimmed materials, so that in light of this finding the present results seemed definitive enough of the general situation without need for a much more costly and extensive time- or rate-parametric study.

Upon initial test evaluations it became clear quickly that, for unidentified reasons, a small amount of material "shakedown" was required to generate repeatable results. This finding was true for both scrimmed and unscrimmed materials. Thus, all results presented here are deduced from tests that subjected the specimen to at least five loading/unloading cycles prior to any data recording. Inasmuch as the physical reasons for material changes under initial loading are rarely identified with specificity, and not really known in the present case, material "shakedown" is a rather non-quantitative description. However, experience showed that the indicated cycles at 0–10 MPa produced reasonable repeat cycles of material response.

As mentioned in the beginning, one of the suspected effects of the scrim material on the transverse stiffness of the adhesive is that the interface between scrim fibers and adhesive may become separated. If this occurs on a massive scale, softening of the adhesive layers would result. Moreover, repeated load or strain cycling may exacerbate this softening characteristic, and it appeared imperative, therefore, to study the stiffness issue under repeated loading. Moreover, adhesives are exposed to inhomogeneous stress states which may elicit different responses according to stress levels encountered. It appears more meaningful to access the response of the adhesive at different stress magnitudes through cyclic loading in various stress ranges and to record results through a kind of differential stiffness as outlined below.

Consequently, each specimen was subjected to at least twenty strain cycles in order to examine whether repeated loading would register a noticeable change in

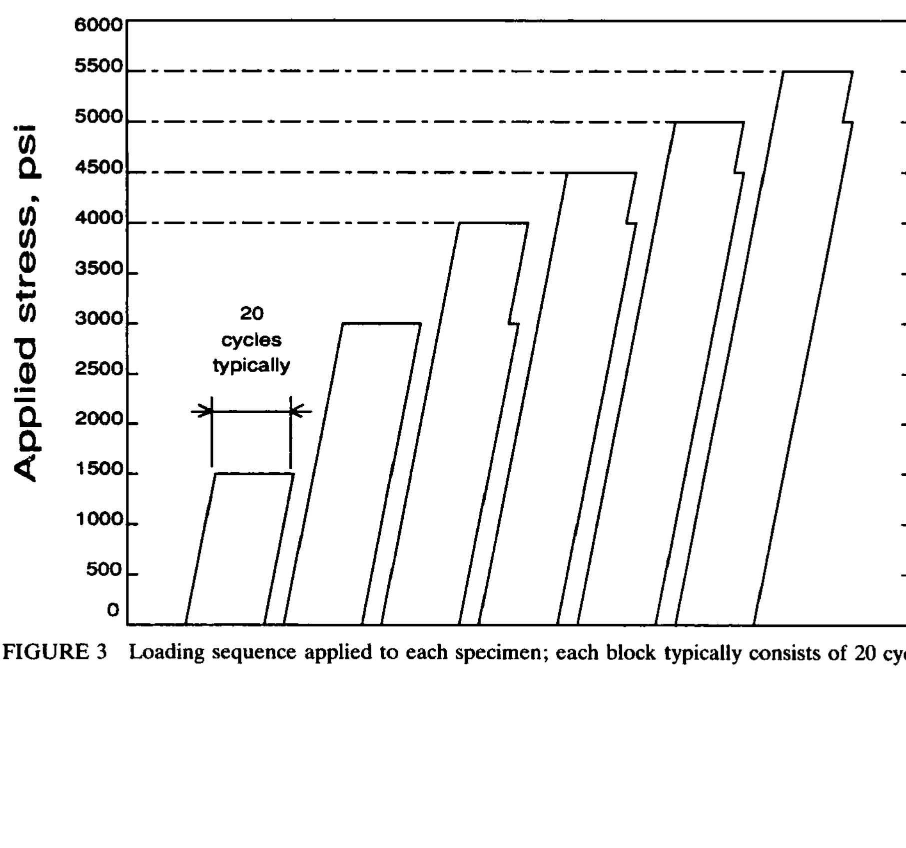

FIGURE 3 Loading sequence applied to each specimen; each block typically consists of 20 cycles.

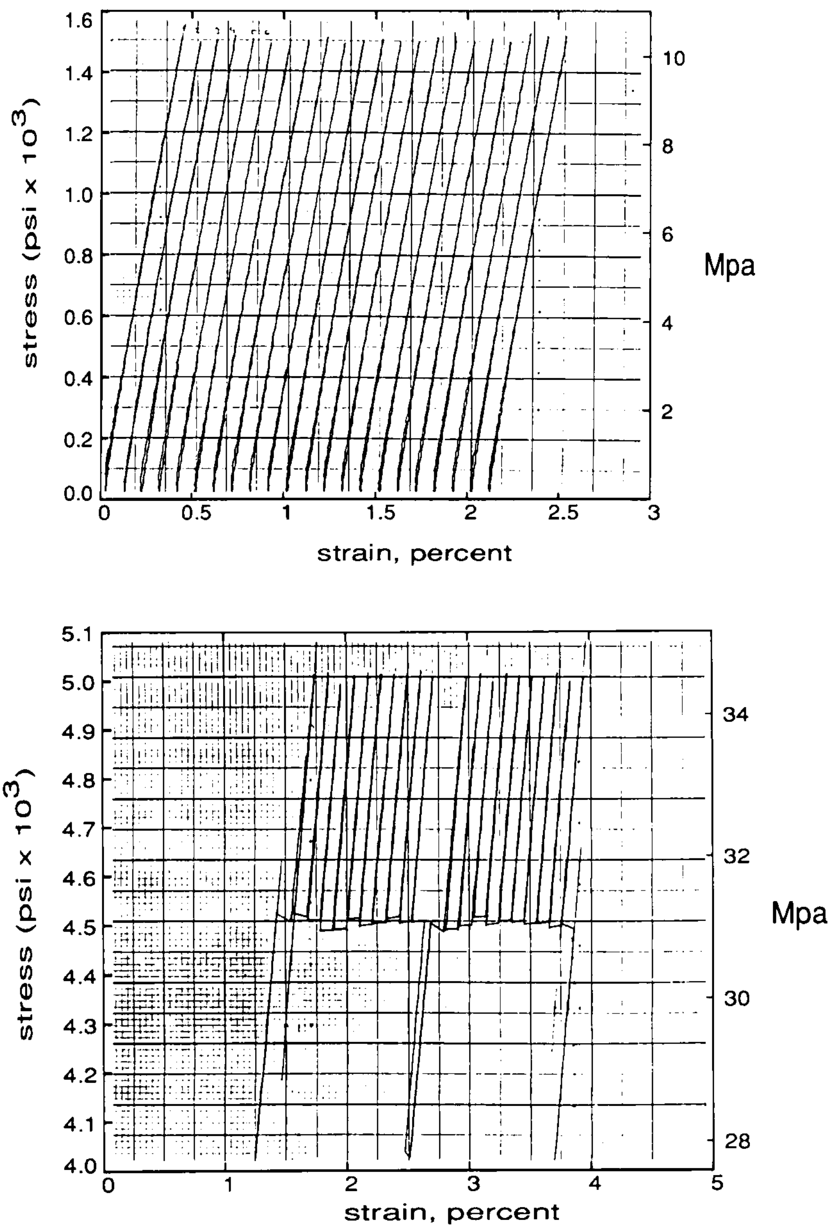

FIGURE 4 Typical stress-strain test history at low (upper graph) and high (lower graph) stress levels.

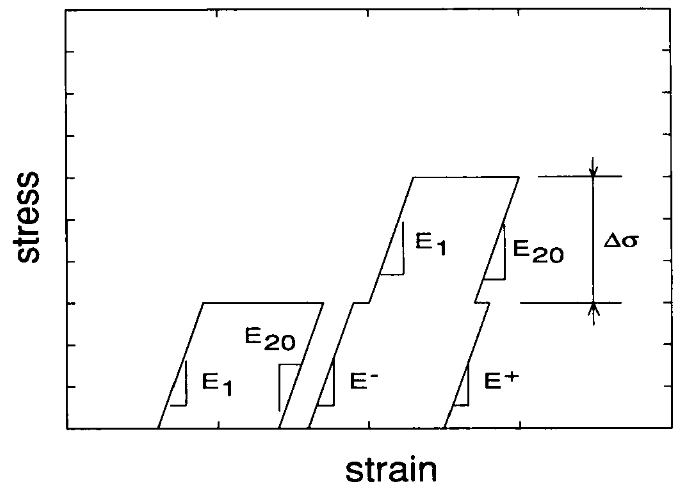

FIGURE 5 Identification of stress–strain behavior in cyclic tests.

the overall stiffness of the materials. In addition, in order to examine whether such prospective behavior would occur at different rates (per cycle) as the load level was increased (approaching maximum loads commensurate with failure loads), load cycling was performed at increasing levels of maximum load; in order to conserve testing time, load cycling at the higher load levels did not include zero load as the minimum value. This measure of expediency is unlikely to influence the conclusions from this work. In general, the following sequence of loading on each specimen was performed. Following the initial shakedown each specimen was cycled successively twenty times between

0 to 10.3 MN/m^2	(0 to 1500 psi)
0 to 20.7 MN/m^2	(0 to 3000 psi)
20.7 to 27.6 MN/m^2	(3000 to 4000 psi)
27.6 to 31.0 MN/m^2	(4000 to 4500 psi)
31.0 to 34.5 MN/m^2	(4500 to 5000 psi)
34.5 to 37.9 MN/m^2	(5000 to 5500 psi)

This sequence is shown graphically in Figure 3. Typical stress strain response graphs are shown in Figure 4. It is clear that this kind of investigation generates a large data base, not all of which can be considered here. In Figure 5 are defined the moduli recorded for further evaluation. Detailed tabulations of these reference values are given in Ref. 2.

It turns out that the loading/unloading cycle resulted usually in a small amount of hysteretic response, which was such that the unloading cycle produced a more nearly straight line response between stress and strain, while the loading phase did not necessarily do so. Examples of this difference in behavior are **not** reflected in Figure 4. Inasmuch as the purpose of this work is to effect a comparison of properties, rather than provide engineering values for them, and in order to ease the choice of property description for reporting purposes, modulus data are typically related to the unloading portion of the stress/strain behavior.

DATA EVALUATION AND RESULTS

There are two principal questions which motivate this investigation: One addresses the differences in the stiffness of scrimmed and unscrimmed material, and the second relates to changes associated with stress level. There are several ways in which such an evaluation can be accomplished; however, let us confine our attention to essentially two sets of data, namely (a) the comparison of the magnitude of moduli at low loads (0 to 1500 psi, 0 to 10.3 MN/m^2) and (b) to a comparison of the effect of increasing load level, with the data normalized by the modulus at low loads. For the latter comparison the fractions were formed from average values. Typically 8 to 10 specimens were used to provide these averages.

The most important results of this study are summarized in Figures 6 and 7. The first of these shows a comparison of the moduli at small loads for the five composite materials examined. Both moduli for initial loading and unloading after 20 cycles are shown with indications of data scatter. Taking the latter into account one notes that there exists surprisingly little difference between scrimmed and unscrimmed materials. This result is, perhaps, not surprising if one assumes that perfect bonding exists between the fibers and the adhesive matrix. The result is, however, unexpected if one considers, as observed the introduction, that the difference in thermal expansion of the fibers and matrix and the less than optimal

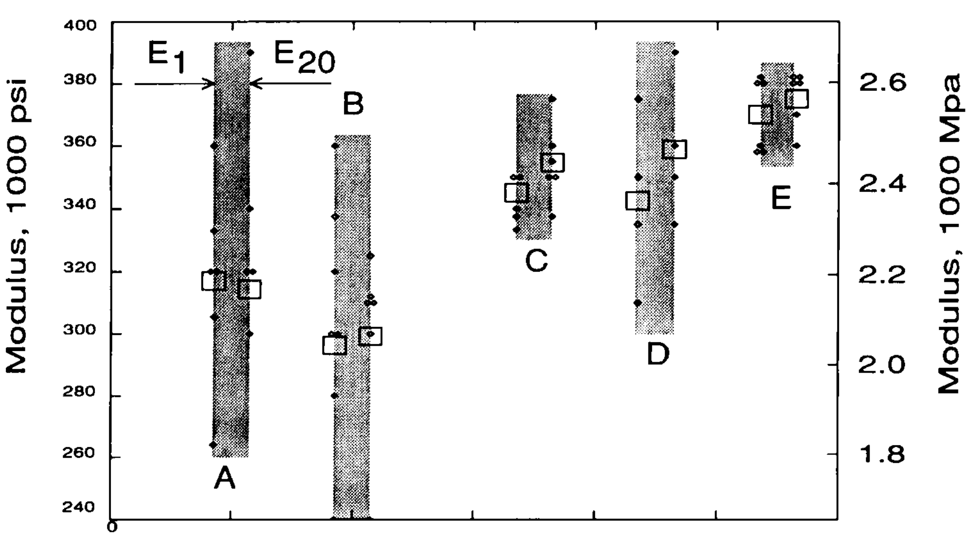

FIGURE 6 Average stiffness (square symbols) at low load levels at beginning (E_1) and end (E_{20}) of 20-cycle test. A = AF163 scrimmed, B = AF163 unscrimmed, C = FM73 polyester knit scrim, D = FM73 unscrimmed, E = FM73 random scrim.

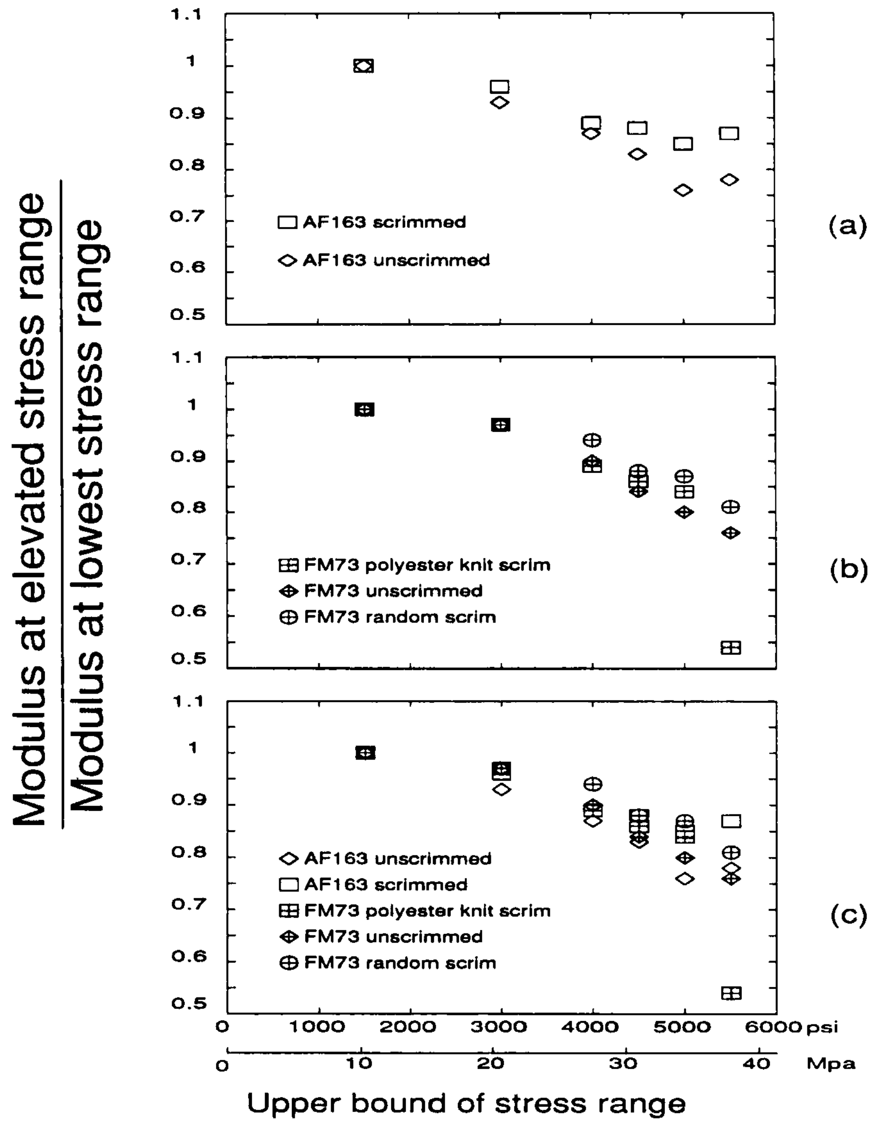

FIGURE 7 Normalized loading moduli as a function of stress level for (a) AF163; (b) FM73; (c) combination of both.

bonding characteristic of nylon and polyester are likely to promote dewetting and softening. That behavior did not, apparently, result in the specimens prepared in this study.

Figure 7 renders a comparison of behavior at increasing load levels, the unloading moduli of Figure 6 serving for normalization of the ordinate, with the abscissa indicating the maximum stress level achieved in a cycle sequence. If any difference between scrimmed and unscrimmed materials emerges from these plots, it points towards a lower stiffness for the unscrimmed materials, rather than a higher one as one might have suspected on the basis of the simple micromechanics arguments of fiber/matrix separation alluded to in the introduction. In fact, there is not much of a fundamental difference between any of the five materials considered with respect to the softening process under cyclic loading as evidenced in the superposed plot of Figure 7c, where the differences are on the order of 10%. Bearing in mind that these differences are well within the range of data scatter it follows that one observes that:

a) The introduction of scrim fibers does not materially change the bond-normal stiffness of the adhesive; in particular at "lower" stress levels fiber-matrix separation does not appear to operate strongly. One might deduce, therefore, that isotropic representation of the adhesive for stress analysis purposes is reasonable.

b) In the material samples studied this observation appears to hold true also at stress levels typically in the range of 50% to 75% of the gross ultimate stress, although "softening" of all samples occured at these elevated stress levels.

The last observation raises the question whether the reduction in stiffness with increasing stress levels is the result of intrinsically non-linear material response or the result of strain-induced micro damage. The conclusive answer to this question would be an autopsy under a microscope for possible damage identification—this examination was not performed because the unloaded state would make microcracking difficult to detect. However, one notes that generally intrinsic material nonlinearity in these types of materials is associated with notable hysteresis, while microcracking of an otherwise linearly elastic solid produces nearly linear behavior under cyclic load with minimal increase in hysteresis losses.

It was noted that, at the higher stress, levels the loading and unloading behavior was more often than not different, indicating significant hysteretic behavior. Some test samples showed no particular hysteresis and moderate softening; it appears reasonably consistent with these data that damage associated with non-linear material behavior comes into play as part of the softening behavior, such as micro cracking and non-linear material behavior in the high strain fields around these microcracks.

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