PREDICTING COHESIVE STRENGTH OF A BONDED JOINT FROM PROPERTIES OF BULK ADHESIVE

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Abstract: This paper shows that the strength of an adhesive tested in bulk correlates well with its cohesive strength in a bonded joint if moisture equilibrium has been established, making it possible to predict the cohesive strength of a bonded joint from the equilibrium strength of the bulk adhesive. Previous investigators failed to arrive at this conclusion because they did not allow sufficient time for the moisture content in the adhesive to equilibrate with the RH of the environment. The findings from the present study opens the door to a more general design approach in which the tensile properties of polymeric materials can be used — along with those of steel, concrete, and various fiber materials — to predict the strength of composite members.

Keywords: Adhesive, bonded, bulk, equilibrium, humidity, joint, moisture, neat, shear, temperature, tension.

Introduction

The durability of epoxies, which are commonly used as a matrix in FRP composites and to adhesively bond FRP composites to other elements of structural members, depends greatly on their resistance to moisture and temperature. Despite the many known benefits, the loss of strength in adverse environments is preventing the application of adhesives to primary load-carrying members. Even in the aerospace industry, with its high standards of fabrication and quality control, adhesives are mainly used for bonding secondary members. Long-term durability must be better understood before adhesives can be reliably used for primary members.

The strength of a bonded joint may gradually deteriorate when the joint is exposed to relative humidities (RH) and temperatures that are high but still within the service environment of the structure. Although the detrimental effect of sustained loading is well known, the physical mechanisms of structural degradation have not been satisfactorily explained.

A related problem is the lack of correlation between the properties of adhesive tested as a bulk (neat) specimen and in a bonded joint. In the opinion of one researcher, "no correlation has been found with either neat adhesive coupon data or with quality-control tests like the single-lap or wedge-crack test [1]." The lack of correlation has been attributed to the widely different causes: uniaxial stress state in bulk specimens versus triaxial stress state in bonded specimens, initial curing stresses in bonded specimens, or differences in adhesive chemistry in bulk and bonded specimens. But no conclusive data have been presented in support of these explanations.

In the authors' opinion, the previously reported lack of correlation in the long-term strength of bulk and bonded specimens stems, to a large degree, from not allowing sufficient time for the moisture content in the adhesive to equilibrate with the RH of the environment surrounding the specimen [2,3].

Surface preparation of adherends is also important in ensuring bonded joint strength. Moisture penetrates the bondline, gradually degrades interfaces, and weakens the bond. As a result, premature adhesive failure can prevent the joint from reaching its full cohesive strength.

This paper will show that (1) moisture and temperature greatly reduce the strength of a bonded joint and (2) bulk and cohesive bond properties correlate well if moisture equilibrium has been established. This opens the door to a more general design approach in which the basic properties of polymeric materials can be used — along with those of steel, concrete, and FRPs used — to predict the strength of composite members.

Experimental Procedures

Test Matrix: The following properties of bulk adhesive were measured in the present study: (1) rapid loading stress-strain curve at 27°C/50% RH; and (2) equilibrium stress-strain curves at -4°C/50% RH, -4°C/90% RH, 27°C/10% RH, 27°C/50% RH, 27°C/90% RH, 49°C/10% RH, 49°C/50% RH, and 49°C/90% RH. The temperature and RH values are nominal; the actual values are listed in the figures.

For comparison with the results from the bulk specimen tests, the following properties of the adhesive in bonded joints were measured: (1) rapid-loading tensile and shearing strengths at 27°C/50% RH and 49°C/90% RH; and (2) creep tensile and shearing strengths at 27°C/50% RH and 49°C/90% RH.

Adhesive: American Cyanamid's FM-300, the adhesive chosen for this study, is a modified epoxy film with a tight-knit moisture-resistant polyester carrier. The manufacturer lists the following features and benefits of FM-300 structural adhesive: "superior metal-to-metal peel strength, composite-to-composite bonding, and composite-to-metal joints; extensively used as surface ply for composite materials;

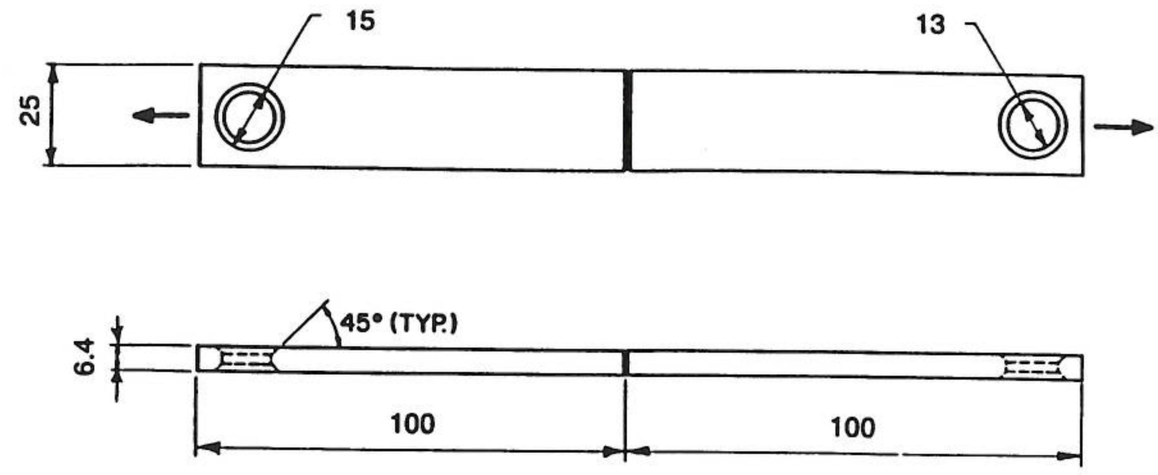

Fig. 1. Bonded Specimen for Creep Tensile Tests; all Dimensions in mm

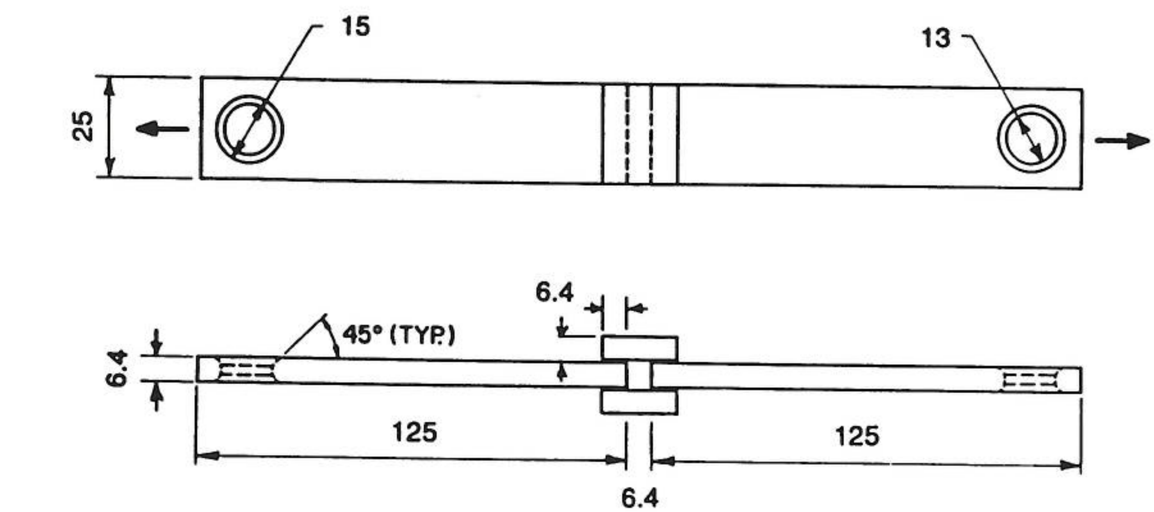

Fig. 2. Bonded Specimen for Creep Shearing Tests; all Dimensions in mm

service temperature from -55 to 150 °C; excellent moisture and corrosion resistance in high humidity environments with no reduction in material properties; and high elongation and toughness with high ultimate shear strength, making it particularly suitable to redistribute the high stress concentrations of graphite epoxy-to-metal bonds and allowing it to accommodate the low interlaminar shear strength of the composite." Of the 24 adhesives the authors have tested, FM-300 is the best suited for possible application to steel highway bridges [2,3].

Specimens: The specimens for measuring the tensile stress-strain curves for bulk adhesive consisted of 100 mm long by 5 mm wide strips cut with a heated knife from 0.25 mm thick films. The films were cured for 60 minutes at 180 °C.

All bonded specimens were made of High-Strength Low-Alloy Structural Steel with 345 MPa minimum Yield Point to 100 mm Thick, ASTM designation A588. The bonded specimens for the creep tensile tests, shown in Fig. 1, consisted of 6.4 X 25 X 100 mm butt-bonded adherends. The thickness of the rapidly loaded specimens was increased to 9.5 mm because in these tests more time was available for moisture in the bondline to reach equilibrium with the RH of the environment.

Similarly, the specimens for the creep shearing tests, shown in Fig. 2, consisted of two 6.4 X 25 X 125 mm main adherends that were spliced with two 6.4 X 25 X 19 mm strap adherends; the lap length was 6.4 mm. For the rapid-loading shearing tests, the main adherend thickness and lap length were increased to 12.7 mm. The bondlines of all bonded specimens were 0.25 mm thick, the same as the thickness of the bulk specimens.

Preparation: The adherends were bonded as follows: (1) wiped the contact surfaces with a cloth, (2) degreased the surfaces with trichloroethylene, (3) cut the adhesive film to size and placed it on one contact surface, (4) placed the parts in a jig, (5) clamped the bonded surfaces, (6) cured the specimens in an oven by incrementally raising the temperature over a 30-minute period to 180 °C and held it for 60 minutes, and (7) removed the specimens from the oven and let them cool at room temperature.

Environmental Chambers: The bulk and bonded specimens were conditioned and tested in environmental chambers built by the authors. The temperature was controlled to ± 1 °C with heating lamps and cooling coils, and the RH to ± 1.5% with silica gel previously conditioned to the desired environment. The exceptions were the bonded specimens for the rapid-loading tests. These were kept in a chamber until moisture equilibrium was reached. They were then taken out of the chamber the bondline was sealed, and the specimens were tested outside of the chamber.

Loading Equipment: All bulk tensile specimens were tested in tensiometers of 200-N load capacity designed and built by the second author [2], the bonded creep specimens in spring-loaded portable frames donated by At&T Bell Laboratories, and the bonded rapid-loading specimens in an Instron machine of 22-kN capacity.

Equilibrium Stress-Strain Curves

The FM-300 adhesive was selected for the present study based on its stress-strain curves measured under rapid-loading at 27°C/50% RH and under equilibrium loading at 49°C/90% RH. The latter was taken as potentially the most severe environment to which a steel bridge might be exposed. Fig. 3 compares the two curves. Both specimens were equilibrated in their respective environments for four days before the tests began.

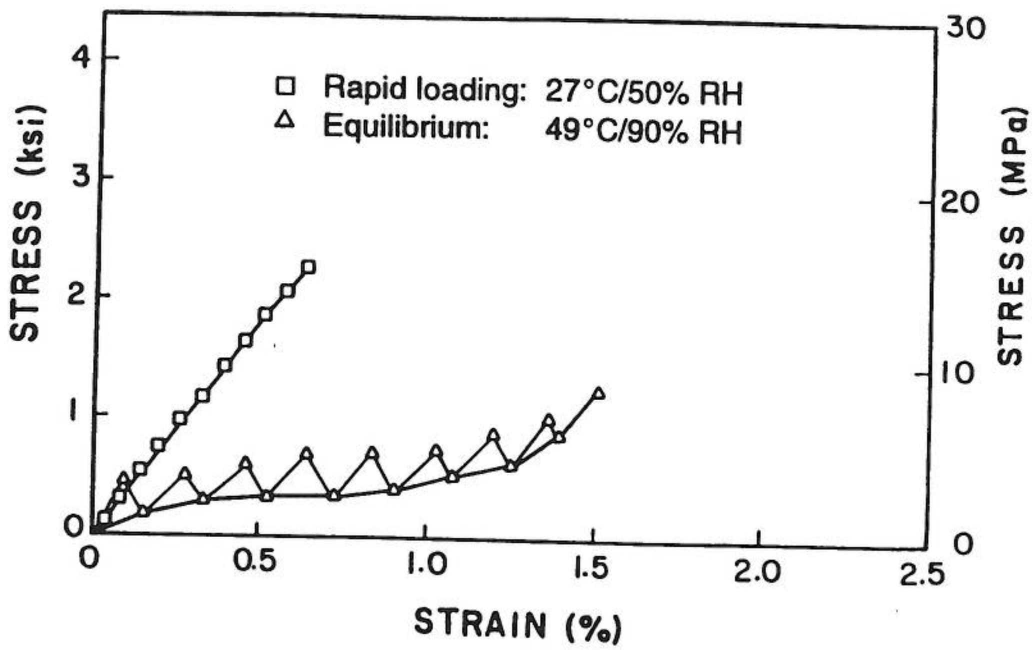

Fig. 3. Screening Test Results for FM-300 Adhesive

In the rapid-loading tests, which lasted about one minute, the strain was incremented in steps of $\Delta\epsilon = 0.0005$ until the specimen failed at $\epsilon_u = 0.0063$ and $\sigma_u = 16$ MPa. The failure strain of the adhesive was about 3.6 times the yield strain of 345-MPa yield strength steel. The rapid-loading stress-strain curve is linear, with a slope of $E = 2,400$ MPa.

In the equilibrium tests, the strain was incremented in steps of $\Delta\epsilon = 0.0012$ and, after each strain increment, was allowed to stress relax for four days under quasi-fixed strain. The test was not entirely strain-controlled because the load cell in the tensiometer, which consisted of an instrumented cantilever beam, deformed under load. This explains why the strain increased by a small amount during stress relaxation. The specimen eventually failed at $\epsilon_u = 0.0151$ and $\sigma_u = 8.6$ MPa. The shape of the stress-strain curve resembles that of a rubbery material which consists of three distinct portions: an initial rise followed by a plateau and ending with a second rise steeper than the first rise. Equilibrium loading, in combination with the severe environment, greatly reduced strength.

The lowest points of all stress-relaxation segments lie on what the authors call the *equilibrium stress-strain curve*, meaning that the moisture content in the adhesive is in equilibrium with the RH of the environment and internal reorganization has taken place as discussed next.

Three possible mechanisms appear to cause polymers to stress relax. One is additional water absorption from the surrounding environment after a strain increment. Tensile loading increases specimen volume and hence water capacity. As the polymer absorbs more water, it swells and stress relaxes under fixed strain.

The second possible mechanism is internal reorganization of water molecules that are lodged between polymer molecules or form weak hydrogen bonds with the polymer molecules. Pressure exerted by lateral contraction under Poisson's effect can force water molecules to rotate into positions that would allow additional lateral contraction and, hence, stress relaxation under fixed strain.

The third mechanism is breakage of cross links, allowing polymer molecules to flow until new links are formed.

The authors believe that additional water absorption and internal reorganization are the dominant mechanisms in the linear-elastic region of the stress-strain curve. Breakage of cross links becomes increasingly pronounced as the "yield" point of the adhesive is approached. The first two mechanisms are largely reversible, the last is not. There is, at present, no method of quantifying the separate contributions of each mechanism to the total stress relaxation.

Stress relaxation at fixed strain — or conversely strain increase at fixed stress — is neither a new observation nor is limited to polymeric materials. Upton [4] referred to it as early as 1916 by writing that for ductile materials "There is a small time factor in elastic deformations. The bulk of the information occurs as instantaneously as the application of the load. There is a slight further increase of deformation with time as the load remains, known to physicist as the 'elastic after effect.' The 'elastic after effect' is of no importance in engineering." Upton was right about the elastic after effect but did not foresee its importance in designing structural members containing polymeric materials.

Civil engineers typically neglect time-dependency in designing steel structures for sustained loading because its effect is small, but account for it in concrete structures by reducing the modulus of elasticity. The more a material absorbs moisture, the more its strength is reduced. Strains resulting from expansion/contraction as RH increases/decreases must be accounted for in predicting polymer strength [2,3]. Such environmental strains can be as large as those induced by mechanical loading [5]. This is equally true for adhesives as it is for the various types of FRP materials that engineers are now specifying for structural members.

Results for Bulk Specimens

Figs. 4 to 6 show the equilibrium stress-strain curves for FM-300 adhesive; the saw-tooth-like load/relaxation segments, which were drawn in Fig. 3, are left out in Figs. 4 to 6. Each figure is for one nominal temperature and varying RHs. As a reference, the rapid-loading stress-strain curve measured at 27°C/50% RH was added as a dashed line.

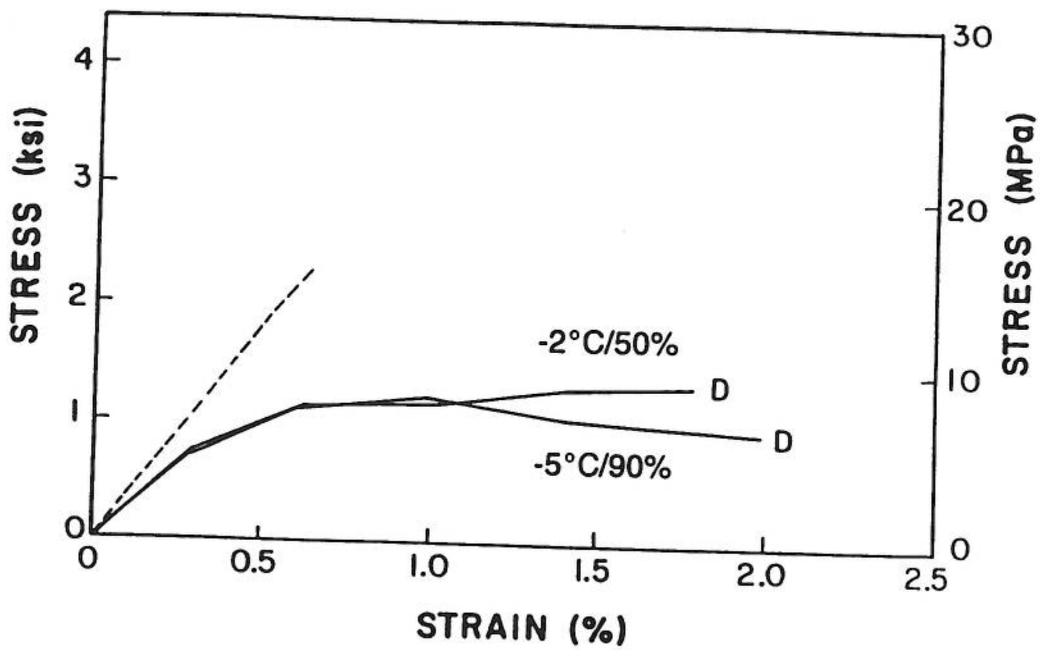

Fig. 4. Effect of RH on Stress-Strain Curve of Bulk Adhesive in -4°C Environment

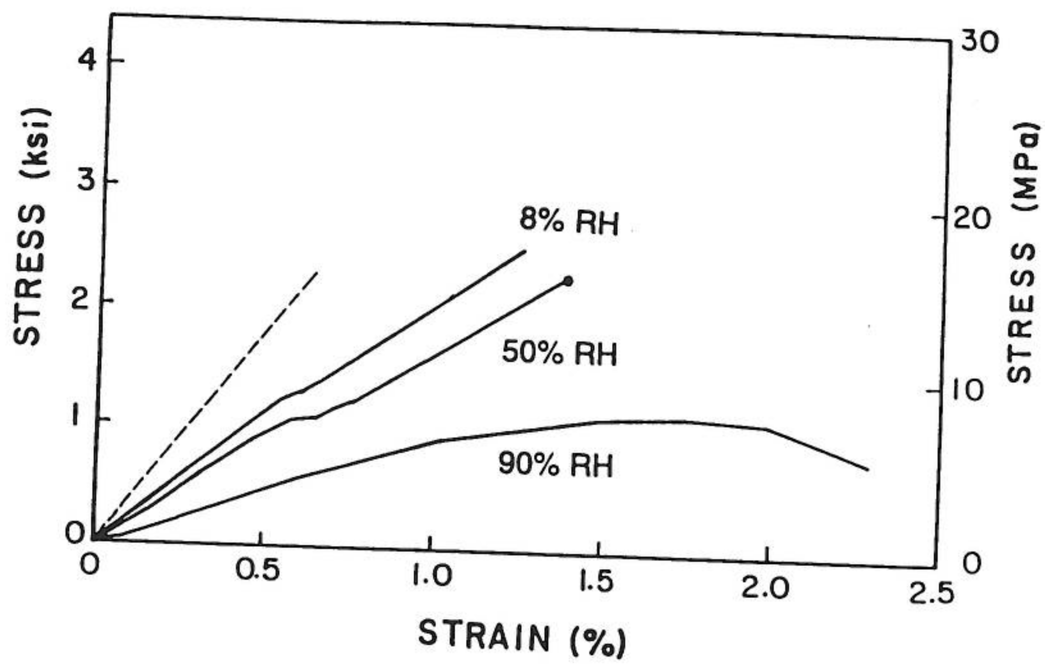

Fig. 5. Effect of RH on Stress-Strain Curve of Bulk Adhesive in 27°C Environment

At -4°C (Fig. 4), increasing the RH from 50 to 90% had little effect on the equilibrium stress-strain curve. The specific humidities in these two environments, defined as kg of water per kg of dry air, are SH = 0.0018 and 0.0033. These values are very

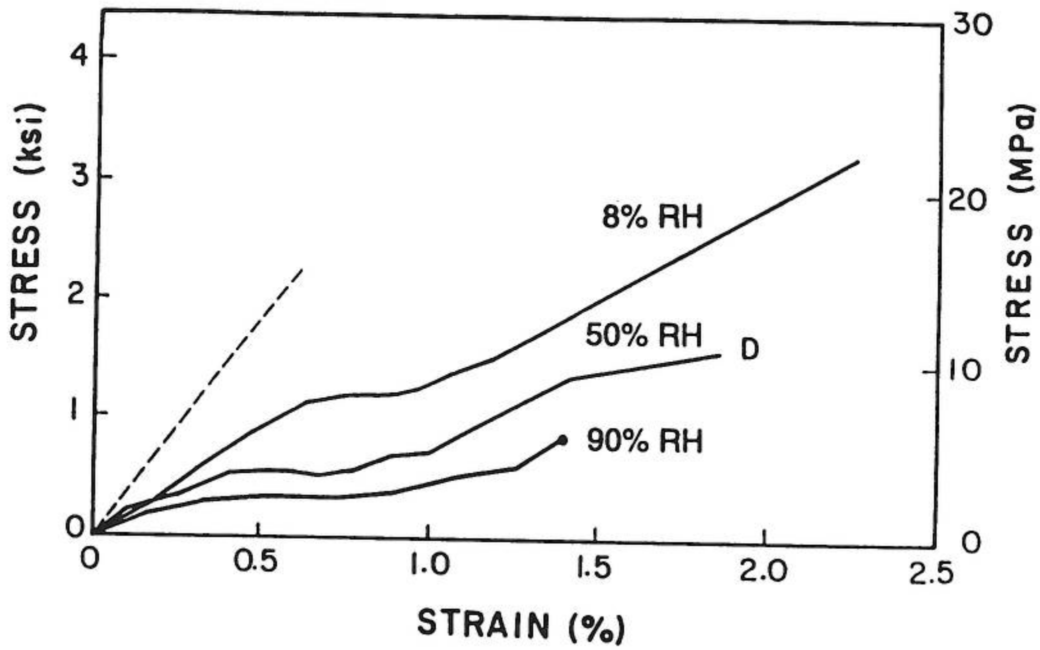

Fig. 6. Effect of RH on Stress-Strain Curve of Bulk Adhesive in 49°C Environment

small. In comparison, the air in a typical indoor environment of 27°C/50% RH has a SH of 0.0113, on average five times higher. Even though the 90% RH suggests a humid environment, the SH is very low at the cold temperature of -4°C. As a result the air and the adhesive contain little moisture at both 50 and 90% RH. This explains the similarities between the two curves. Both are typical of an elastic-plastic stress-strain curve with an elastic modulus of E = 1,700 MPa and a pronounced yield plateau at σ = 8.0 MPa. The tests were discontinued (D) at a strain of about ϵ = 0.002, before the specimens ruptured into two parts.

At 27°C (Fig. 5), increasing the RH from 8 to 50% had a moderate effect on strength. Both of these equilibrium stress-strain curves exhibited some yielding at 7.3 and 8.6 MPa, respectively, at values similar to the 8.0-MPa plateau in the tests performed at -4°C. After the short plateau, the specimens strain hardened until they ruptured at σ_u = 17.2 and 16.8 MPa. But increasing the RH from 50 to 90% greatly lowered and plasticized the equilibrium stress-strain curve (Fig. 5). Its plateau peaked at 7.9 MPa, a value similar to the yield plateaus cited above. The elastic moduli were E = 1,550, 1,200, and 690 MPa at 8, 50, and 90% RH.

At 49°C (Fig. 6), the equilibrium stress-strain curve at all three RHs rose at first elastically, then leveled out, and finally rose again in a manner resembling stress-strain curves of elastomeric materials. The elastic moduli varied from 700 to 1,200 MPa at 8 and 90% RH.

In separate bulk specimen tests of FM-300 adhesive whose results are reported in [3], the authors also measured water absorption by weight as well as expansion as a function of increasing RH at a constant temperature of 27°C. Knowing the dry mass and density of the specimen, the mass gain was converted to volumetric strain. The moisture expansion was also converted to volumetric strain. Both were found to be proportional to each other. Clearly, the link between moisture absorption, RH-induced strain, and adhesive strength is unmistakable. As the moisture content rises with RH, the adhesive is plasticized and becomes more ductile. Ductility is gained at the expense of strength.

This paper presents the data in terms of the RH effect at constant temperature. The same data could be replotted to illustrate the temperature effect at constant RH. This is not done here for lack of space. Suffice it to say that over ranges of RH and temperature found in typical service environments, RH affects the strength of adhesives more than does temperature. A companion paper examines the combined hydro/thermal/mechanical behavior of bulk adhesive [5].

Results for Bonded Creep Tensile Specimens

The specimens for the creep tensile and shearing tests, shown in Figs. 1 and 2, were bonded and placed in environmental chambers with environments of 27°C/50% RH and 49°C/90% RH. As test frames became available — there were fewer test frames than specimens — specimens were loaded and returned to the chambers.

Seven tensile creep tests were performed in each environment. The specimens were loaded to a tensile stress higher, equal to, or lower than the equilibrium bulk tensile strengths in the same environments. The bulk tensile strengths were 15.9 and 5.9 MPa in the 27°C/50% RH and 49°C/90% RH environments. They are shown as solid circles in Figs. 5 and 6, and as dashed lines in Figs. 7 and 8.

Two points are plotted in Figs. 7 and 8 for each specimen; the open circles indicate the total time a specimen stayed in the chamber in the unloaded and loaded states, and the adjacent solid circles indicate the portion of the total time in the chamber during which a specimen was under load, in other words, the duration of the creep test. An arrow identifies the data points for the specimens that did not fail at the end of the project. The findings for the creep tensile strength were as follows:

- As expected, most specimens (8 of 9) failed when stressed to the equilibrium bulk tensile strength or higher. Conversely, most specimens (4 of 5) did not fail when stressed less than the equilibrium bulk tensile strength even after 430 days, a time 3 to 4 times longer than is needed for the moisture content in the 6.4 mm wide bondline to equilibrate with the RH of the environment.
- The 49°C/90% RH environment was much more severe than the 27°C/50% RH environment.

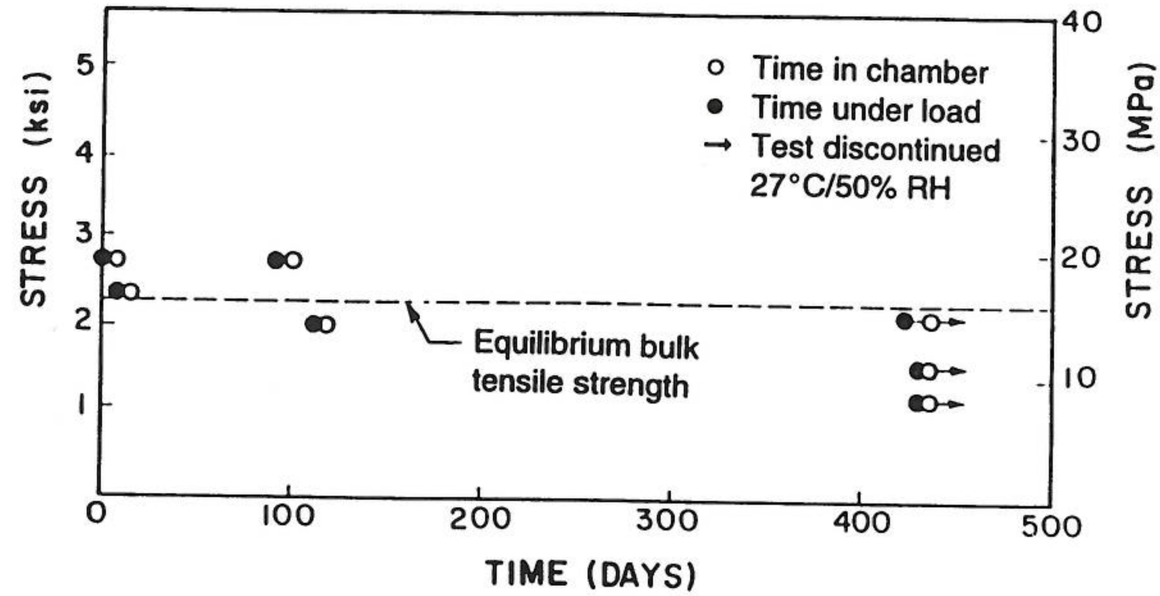

Fig. 7. Creep Tensile Strength of Bonded Specimen in 27°C/50% RH Environment

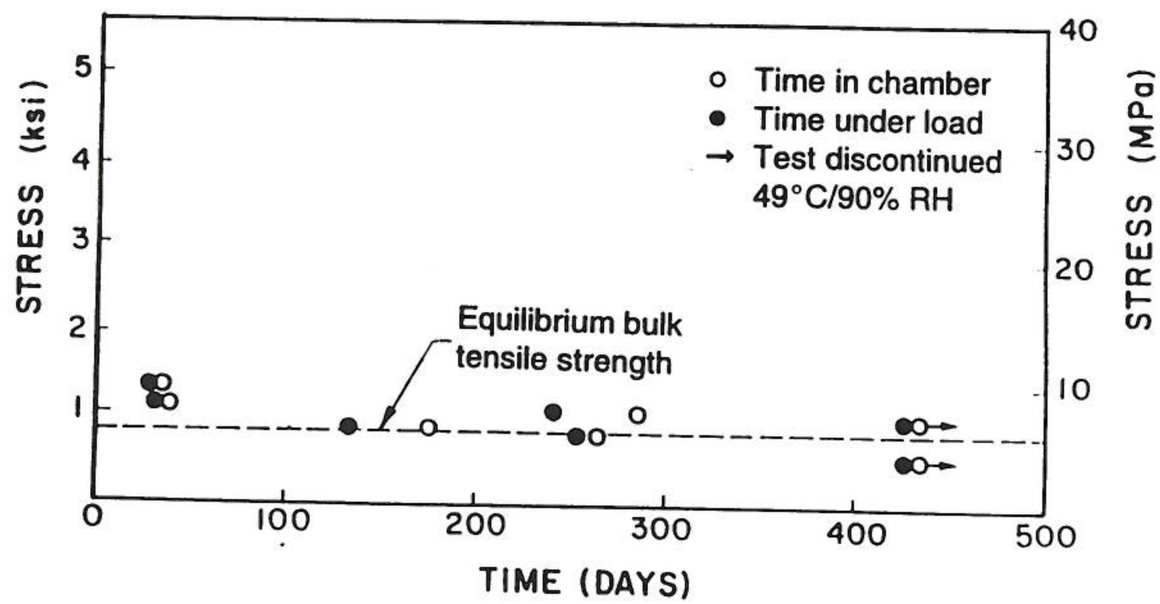

Fig. 8. Creep Tensile Strength of Bonded Specimen in 49°C/90% RH Environment

- Creep failures occurred predominantly in adhesion (Fig. 9), that is, along the interface between the adhesive and the steel adherend. But one specimen tested at 49°C/90% RH failed in 90% cohesion. It was stressed to 9.8 MPa, a value higher than the equilibrium tensile strength. The tests support the authors' believe that specimens stressed to the equilibrium tensile strength or higher will eventually fail in creep.

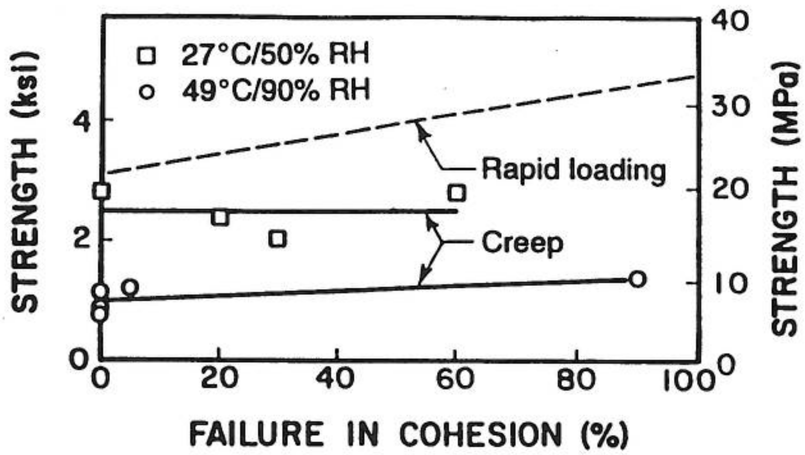

Fig. 9. Effect of Failure Type on Creep Tensile Strength of Bonded Specimen

The equilibrium bulk tensile strength is an upper bound on the creep tensile strength of the adhesive in a bonded joint. It should come as no surprise when bonded tensile specimens fail in creep when loaded to a stress equal to or greater than the equilibrium tensile strength. If an adhesive does not resist such stresses in bulk, it cannot be expected to do so in a bonded joint. Yet the literature contains the results of numerous tests in which the sustained load was greater than the equilibrium strength. In the authors' opinion, such tests would not have been needed if the equilibrium strength had been determined. The creep tensile strengths were much lower than the 28.1- and 21.7-MPa rapid-loading tensile strengths of specimens bonded with FM-300 in the same environments, which the authors reported in [3].

Such predictions are valid only for the cohesive strength of the joint. The adhesive strength depends on surface preparation and cannot be predicted from cohesive strength data. The strength of a bonded joint cannot exceed the cohesive strength of the adhesive. But it can be less if improper surface preparation lowers the adhesive strength to a value smaller than the cohesive strength.

Results for Bonded Creep Shearing Specimens

In a second series of tests, the creep shearing strength was determined for the bonded joints shown in Fig. 2. Seven specimens were tested at 27°C/50% RH and seven specimens at 49°C/90% RH. Lacking equilibrium stress-strain curves in shear, the equilibrium shearing strength was assumed to be the same as the equilibrium tensile strength. This seemed justified given that many manufacturers reported similar values of tensile and shearing strengths in their product literature [2].

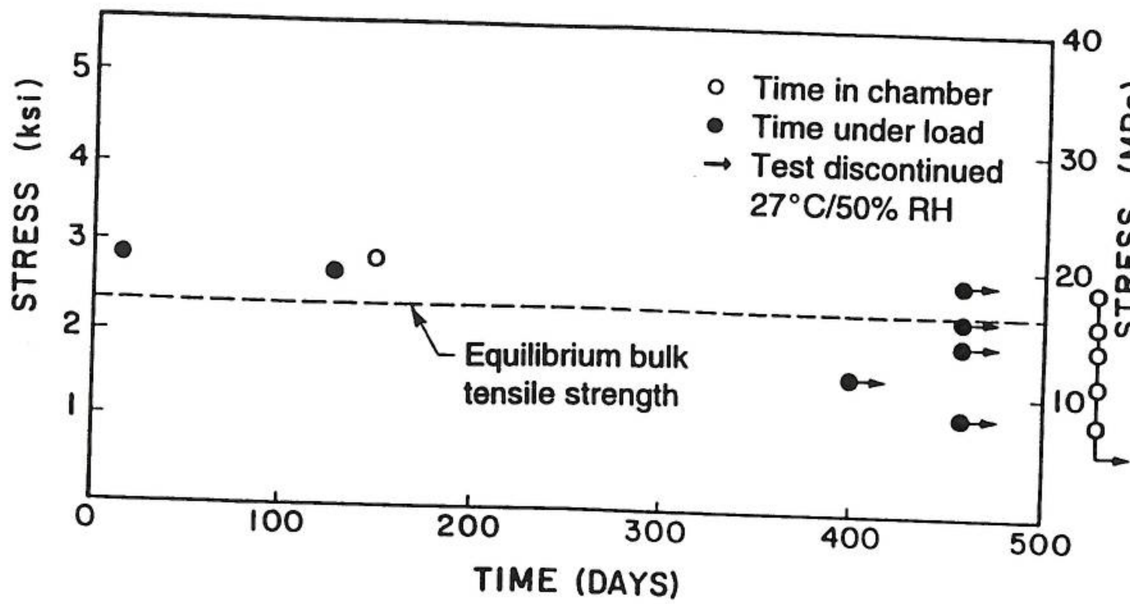

Fig. 10. Creep Shearing Strength of Bonded Specimen in 27°C/50% RH Environment

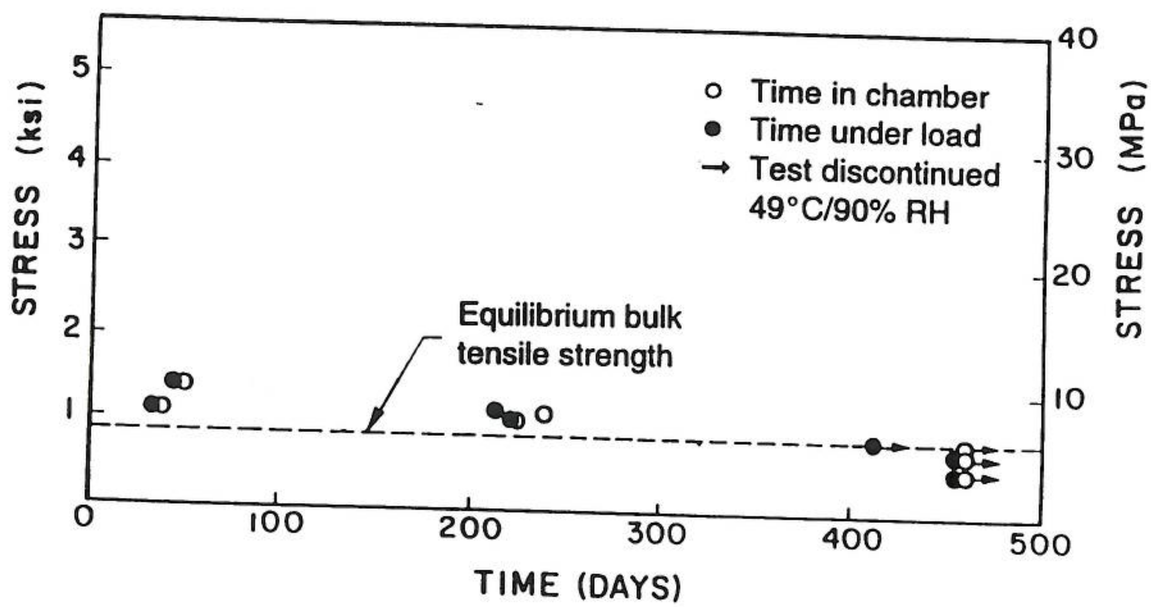

Fig. 11. Creep Shearing Strength of Bonded Specimen in 49°C/90% RH Environment

Figs. 10 and 11 show the results of the creep shearing tests of joints bonded with FM-300 and tested at 27°C/50% RH and 49°C/90% RH. The following was found:

- As in the tensile tests, most specimens tested in shear (6 of 8) failed when stressed to the equilibrium bulk tensile strength or higher, while all specimens (6 of 6) did not fail when stressed less than the equilibrium tensile strength

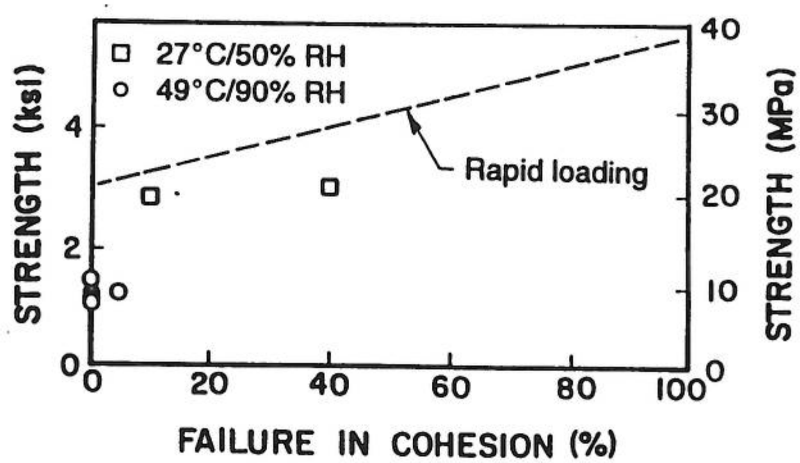

Fig. 12. Effect of Failure Type on Creep Shearing Strength of Bonded Specimen

even after up to 457 days, a time much longer than the narrow bondline needs to reach moisture equilibrium.

- Again, the bondlines failed mostly in adhesion (Fig. 12).
- Changing the environment from 27°C/50% RH to 49°C/90% RH decreased the creep shearing strength about the same as in the creep tensile tests.

Conclusions

- The equilibrium bulk tensile strength of an adhesive gives an upper bound on its cohesive strength in a bonded joint.
- The cohesive strength under sustained loading (creep) can be predicted from the equilibrium bulk tensile strength. In the authors' opinion, past inability to correlate the strength of adhesive in bulk with that of adhesive in a bonded joint resulted from not allowing sufficient time for the moisture content in the bondline to equilibrate with the RH of the environment.
- Increasing the RH and, to a lesser degree, the temperature greatly decreases the strength of adhesives.
- The adhesive strength of a bonded joint depends largely on surface preparation. It cannot be predicted from the cohesive strength of the bulk adhesive.
- Adhesives appear to have similar tensile and shearing strengths.
- Tests of an adhesive for application in a structure exposed in a given environment must be performed under conditions typical of the application. The bondline must be given enough time in the test to absorb the amount of moisture that it would in the service environment of the structure.

The adhesive and cohesive strengths of a bonded joint involve different physical phenomena that must be treated separately. An analogy from steel structures is the behavior of slip-critical connections in which preparation of the contact surfaces (mill scale versus blast cleaning or painting) determines the friction coefficient and hence the slip resistance, while the tensile strength of the bolt determines the shearing strength ($F_v = 0.60F_u$). Likewise, surface preparation of the adherend surfaces in a bonded joint determines the adhesive strength while the equilibrium bulk tensile strength determines the cohesive strength.

Irrespective of whether joints are bonded or bolted, surface preparation should be optimized so that the joint can reach its full shearing strength. This paper addressed the cohesive strength of bonded joints.

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