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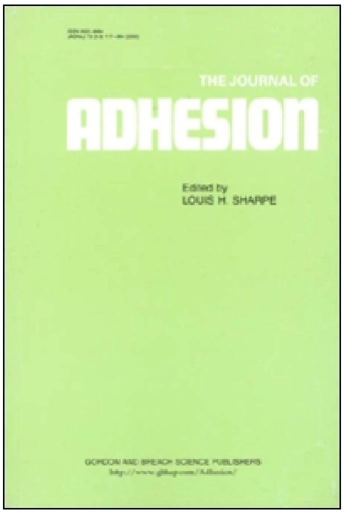

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The Strength of Adhesive Joints†

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The main rules pertaining to the strength of adhesive joints are: (1) This strength is a mechanical (or rheological) property. The local stress which causes the extension of a pre-existing crack can be determined only if the stress pattern in the whole adhint is known and the intensification of stress at flaws is taken into account. (2) The rupture occurs *in* a material, not *between* two materials. Consequently, the molecular forces across the adhesive—adherend interface are irrelevant, and the "adhesion tension" does not determine the adhint strength.

In this summary only the main rules of the science of adhesive joints can be presented; for a more detailed review the author's recent book[1] should be consulted.

Rule I. An adhesive joint (or, for short, adhint) is a solid, and its strength must be studied in the same manner as that of other solids. In the determination of strength, generally the external force F (just sufficient to cause rupture) is measured. In some instances, this force may be adequate to indicate whether the production adhints will, or will not be found satisfactory but it is not directly usable for understanding the cause of fracture or for predicting the strength of adhints having different geometrical shapes.

The rupture of a solid almost always starts at one point, namely at the point where the local stress first overcomes the local strength. The accent is on the word local. Consequently, the absolute value of the force F applied, perhaps, several centimeters away from the source of fracture has no direct

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relation to the stress σ_o which really causes the pre-existing flaw or crack to grow and to split the solid in two parts. The translation of F into σ_o may be considered as being done in two successive steps.

First the stress pattern produced by F everywhere in the solid must be calculated or determined, assuming each phase of the adhint to be completely homogenous. This procedure was followed for several geometrical types of adhint. The simplest instance is supplied by a butt joint in which the adhesive material has the main mechanical properties (the modulus of elasticity and the Poisson ratio) identical with those of the adherends. In this system, the tensile stress would be simply equal to F/A, if A is the cross section of the adhint (perpendicular to the external force). An analogous approximation results in the equation

$$F = kw\,\sigma_m\,(E/E_1)^{0.25}\,\delta^{0.75}\,h^{0.25} \tag{1}$$

in which F is the force required to maintain steady peeling, k is a numerical constant depending on the Poisson ratios of the adherend ribbon and the adhesive (usually it is between 0.4 and 0.7), w is the width of the ribbon, σ_m the tensile strength of the adhesive; E and E_1 are the moduli of elasticity of, respectively, the ribbon and the adhesive; δ is the thickness of the adhesive film, and h that of the ribbon. Both the ribbon and the adhesive are supposed to be Hookean solids, and the rupture interface is supposed to be in the plane of the external force F.

Even when these two assumptions are permissible, equation (1) still affords no value for the stress σ_o. The tensile strength σ_m of any solid is smaller than the "crucial" stress σ_o because no solid is truly homogeneous. The second step of the path from F to σ_o is determination of σ_o when the stress pattern, as it would have been in a uniform solid, is known.

Because each solid has different mechanical properties at different points (at a dislocation, a vacancy, a chain fold, a grain boundary, an inclusion, etc.), the actual (local) stress in many spots is smaller than the average stress calculated from F and the bulk properties of the solid; these low stresses are irrelevant as far as fracture is concerned because, in general, they do not originate a break. In some other spots, the local stress is much greater than the average, and at *a* spot it precipitates the growth of a crack. Equation (2) shows how much the local stress σ_o may exceed the average stress σ_a. If an ellipse with the half-axes a and b is cut in a wide sheet so that a is perpendicular and b is parallel to the average stress, then

$$\sigma_o = \sigma_a\left(1 + \frac{2a}{b}\right). \tag{2}$$

If the ellipse is narrow enough to deserve the name of a crack, the ratio $2a/b$ is great and σ_o is much greater than σ_a.

In the first step of the above procedure, it is often found (especially in lap joints) that stresses σ_a have directions different from that of the external force and that their intensities in many directions are much higher (for instance by a factor of 10) than would be guessed before studying the stress pattern. In the second step, the value of σ_0 often is found to exceed that of σ_a by a factor of 10, 100, or even greater. Thus break is caused by an external force which, at a superficial inspection, would be considered far too small to overcome the forces of cohesion. These forces can be calculated, for instance, for alkali halide crystals, and they usually are 500–1000 times as great as the experimental tensile strength of these crystals.

The estimate of σ_0 for adhints is a little more complicated than for single solids because there are two moduli of elasticity, two Poisson ratios and so on to consider, and because the interface between the two phases gives rise to a more complex stress pattern. Composite materials (such as glass-reinforced plastics) share this inconvenience with adhints.

As far as the author is aware, no serious criticism of rule I has ever been published. Nevertheless the rule often is disregarded. The main reason for this sorry state of affairs is that adhesives are made by chemists and adhints commonly are tested by the makers of the adhesives. If a chemist compares the performance of an adhesive containing, say, a nitro group in the molecule (or a monomer) with that of an analogous adhesive free of NO_2, it is natural for him to attribute the difference observed to the direct intervention of the nitro radical. In reality every change of the chemical composition alters the macroscopic mechanical properties of the system and thus affects the stress pattern produced by the outside force; and the kind and frequency of the flaws present in the solid before the rupture also are changed when the composition is. Thus both steps leading to σ_0 from F become steeper or shallower. There exists also a different connection between composition and F; it is explained later in this talk.

Rule II. Rupture so rarely proceeds exactly between the adhesive and an adherend, that these events (that is, "failure in adhesion") need not be treated in any theory of adhesive joints. Practically always separation occurs in one of the solid materials present in the adhint. This is a result of probability. The boundary between the adhesive and an adherend is a predetermined surface of a very complicated shape because of the surface roughness of all solids. The probability of a crack following this surface is about as small as that of tearing a sheet of paper and finding that the tear spells the experimenter's name.

Apparent "failures in adhesion" are quite common but they do not contradict rule II. In many instances, the separation takes place so near the interface (few microns in the instance of a purified polyethylene adhering to an aluminum ribbon) that the adhesive remaining on the adherend after the rupture is not visible to the unaided eye. In many other instances, a weak boundary layer is present. In these adhints, no interface between adhesive and adherend exists at all; and, of course, no rupture can advance along a non-existing interface. Instead, a third material keeps the adhesive apart from the adherend. This material may be air, or impurities "bleeding out" of the adhesive, or a reaction product of adhesive and adherend, and so on. Since it is much weaker (mechanically !) than the two main components of the adhint, it is likely to break first on the application of the external force.

When a weak boundary layer (WBL) is present, the force F necessary for rupture may be only a small fraction (say, one fiftieth) of that required to break an otherwise identical adhint free of WBL. Probably the easiest way of obtaining a pair of adhints for such a comparison is to use a hot-melt adhesive and to prepare a joint (a) by using a temperature which is too low or a formation time which is too short, and (b) by using time and temperature guaranteeing a complete contact. The adhint (a) will contain air pockets between the two main phases, the separation will occur chiefly in the air, and force F will be much smaller than in system (b).

The possibility of introducing or eliminating a weak boundary layer by an alteration of the chemical composition of the adhesive is the other aspect of the relation between composition and F; see above. The adhesive A may contain impurities which tend to aggregate near the interface and thus to form a WBL. When a nitro group is introduced in the polymer, the new adhesive B may be a better solvent for these impurities than A is; hence, no "bleeding out" and no formation of a WBL takes place. Or, polymer A may be incapable of a chemical reaction with the adherend and thus may give rise to long lasting joints but the NO_2 group of polymer B may cause some corrosion, and the corrosion products may constitute a zone of weakness. Whenever WBL is a possibility, chemists are needed; mechanical engineering which is adequate for proper adhints (those free of WBL) is helpless when the adhints are improper (i.e., contain WBL).

Since rupture does not occur between two phases, the forces acting across their interface (and discussed on the first day of the Symposium) do not determine the breaking load F. Thus, the "molecular" theories of adhesion, which attribute the strength of adhints to these forces, cannot be correct. We may predict that the F of adhints such as "ice—solid argon—ice" will be small; but it will be small not because the attraction between ice and argon is small but because the cohesion of argon is much less than that of polymeric adhesives.

Since rupture is not a surface event, the theories connecting the strength of adhints with the "adhesion tension" and wetting also cannot be trusted. Good wetting of the adherend by the adhesive is necessary to avoid formation of WBL consisting of air pockets; but as soon as uninterrupted contact of adhesive and adherend is achieved, the surface energies of the two phases (if these energies exist) cease to be of interest.

As a general conclusion, it may be stated that the problem of the strength of adhesive joints is a problem of applied mechanics or rheology; but often chemistry is needed to find out what solid phases are encountered in the system.

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1. J. J. Bikerman, *The Science of Adhesive Joints* (Academic Press, New York, 1968).