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On the Definition of Adhesion

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I. INTRODUCTION

The definition of the word, Adhesion, in Webster's International Dictionary, 3rd edition, contains the following pertinent items. (The numbering is Webster's; the omitted definitions are not relevant to science or technology).

2) The action or state of adhering, specifically, a sticking together of substances (as of glue to wood).

7a) A grip or sticking effect produced by friction.

7b) The force that must be developed to overcome this grip.

8) The molecular attraction that must be exerted between the surfaces of bodies in contact—distinguished from *cohesion*.

The Oxford English Dictionary gives the following:

The action of sticking (to anything) by physical attraction, viscosity of surface, or firm grasping.

The ASTM gives the following¹:

Adhesion, n: The state in which two surfaces are held together by interfacial forces which may consist of valence forces or interlocking action, or both. (See also *adhesion*, *mechanical* and *adhesion*, *specific*.)

Adhesion, *mechanical*: adhesion between surfaces in which the adhesive holds the parts together by interlocking action. (See also *adhesion*, *specific*.)

Adhesion, *specific*: adhesion between surfaces which are held together by valence forces of the same type as those which give rise to cohesion. (See also *adhesion*, *mechanical*.)

This paper is published with the express purpose in mind of stimulating discussion.
Editor.

There is not space, here, to review all the attempts that have been made, to define "adhesion"; the three definitions given above are quite representative of the range that exists. All the available definitions are unsatisfactory for several reasons, which will be discussed below.

The most fundamental criticism is a physical one. Physically, adhesion between two phases has both macroscopic (i.e., mechanical) and microscopic aspects, which are complementary to each other. The macroscopic aspect involves properties such as elastic modulus and plastic compliance, and processes such as the transfer or dissipation of quantities of work which can be measured by ordinary laboratory instruments. Stress is a property which has meaning only when the area to which force is applied is large enough that the atomic and molecular structure of the material can be ignored, and the system treated as if it were a continuum.

The microscopic phenomena that are pertinent to adhesion are the binary atomic and molecular contacts, and the short- and long-range interatomic attractions and repulsions, that are present in the systems where the macroscopic phenomena are observed. The microscopic processes are, in most cases, known only by scientific inference; i.e., they cannot be observed directly in the same way that the macroscopic phenomena are observed.

The available definitions of adhesion ignore the difference between these two *physical* aspects, or give the impression that they can be subsumed under a single set of verbal terms. Consequently, they actually interfere with the scientific elucidation of adhesion, and hence with the technological exploitation of the phenomenon.

A second, very fundamental criticism is that, heretofore, definitions have not recognized the fact that there is a serious mechanical and topological difference between systems in which there is only one interface present, *vs.* those in which two interfaces exist. See below, where we discuss the distinction between adhesion and adhesive joint strength.

To return to the dictionary definitions themselves, terms such as "gripping" or "sticking" are employed in such a way as to indicate that the lexicographers considered them self-explanatory. (If they are not self-explanatory, there is danger of circularity. Thus, the dictionaries define "sticking" by use of the word "adhesion".) So there is, logically, a need to replace these undefined words which refer to mechanical processes, with mechanical representations, i.e., in terms of force and energy.

We also note that the noun, adhesion, is derived from the Latin verb, *haerere*, to stick. This gives us the hint that the verb, "to adhere", should be defined before the noun.

Finally, we must restrict our present task, of the defining of "adhesion", to solid systems. This is legitimate both lexicographically and physically. For example, in a system in which one or both phases are liquid, the

mechanical properties of the system as a whole are so different from those of a solid-solid system that, while the same word, adhesion, is commonly used, it requires a separate definition[†]; and limitations on space prevent us from treating such systems in detail here.

II. DEFINITIONS

We will first subdivide the definitions of the verb, to adhere, according to macroscopic and microscopic usages.

A. Macroscopic²

1) *Adhere*. v., i., Two elastic solid bodies are said to adhere to each other when mechanical force or work can be transferred from one to the other, in tension, in shear, or in any combination of tension and shear, across an interface where intimate contact exists, the interface being perpendicular to the stress in the case of pure tension, or parallel to the stress direction in the case of pure shear.

2) For a system comprising two bodies, one of which is plastic or visco-elastic and the other elastic or plastic or visco-elastic, the bodies are said to adhere, when mechanical force or work can be transferred from one to the other, as in definition no. 1, with the following further qualifications: Either, in the period of time under consideration, there is negligible plastic deformation; or plastic deformation occurs only in regions that are significantly removed from the interface.

Comments

a) The macroscopic definitions can be extended to systems in which one (or both) of the bodies is liquid, by noting the fact that a liquid has zero shear strength. But the absence of shear strength, in a liquid, has the consequence that it would be necessary to specify that there be macroscopic restraint (e.g., confinement in a cylinder with the solid phase at one end) against gross shear deformation, in order for stress or work to be transmissible in tension. Such a stipulation would make adhesion strongly dependent on the gross geometry of the system; and for *solids*, observation tells us that adhesion is not dependent on such *gross* geometry.

b) One purpose of the specification about plastic deformation is, the recognition of the fact that a system of two solids such as an ice skate on ice is not commonly considered to be an adhering system.

[†] A more extensive treatment of the definition of "adhesion" is in preparation, and will be published in *Treatise on Adhesion and Adhesives*, Vol. 5, R. L. Patrick, ed., Marcel Dekker, Inc., New York, (in press).

c) If both phases are plastic or visco-elastic, and if the phases adhere to each other, the interface itself may deform quite appreciably, by expansion, contraction, flexing or distortion, with maintainance of its topological integrity. The reference locus for the stress is, then, the local tangent plane of the interface. The deformation considerations stated in Definition 2 apply, but the second of the two qualifications is revised as follows: In the time period under consideration, the plastic deformation in material on either side of and close to the interface is no greater than that in the corresponding interfacial region.

d) The accepted thermodynamic definition of a phase is, a region of space which is sensibly homogeneous in regard to composition and physical properties. Hence if "weak boundary layer" material³ is present, it must be regarded as a separate phase. The system, then, is a three-phase composite, with the interlayer phase being thin and also weak. Such a system differs topologically (*i.e.*, in the number of interfaces present) from a two-phase adhering system; and it differs mechanically from an adhesive joint (which see, below) in that the mechanical or wetting properties of the interlayer phase are so poor that the overall system is weak.

B. Microscopic

3) Two bodies in the form of condensed phases are said to adhere to each other if an interface of intimate contact exists (over an appreciable period of time) and if the contact is close enough, over an appreciable region of the interface, that interatomic or intermolecular forces between the atoms or molecules in one phase and those in the other phase, when summed over all atoms or molecules, are appreciable. In this case, "appreciable" means that the total force, or energy of interaction, is commensurate with or larger than the loading to which the bodies are subjected, or the energy which is supplied externally.

Comments

a) Valence forces acting across an interface constitute a specific case, under Definition 3.

b) The description of forces in terms of the Lifshitz theory of attractions⁴ is a refinement on the theory of interaction of discrete atoms or molecules in separate phases. It includes cooperative effects in the electronic behavior of the atoms or molecules in a single phase, as manifested in the interaction across an interface or between bodies separated by a layer of a different phase. In terms of Definition 3, the existence of forces of the type described by the Lifshitz theory is explicitly included.

d) The term, to adhere, is also commonly used in a microscopic sense, as describing systems such as a single atom colliding inelastically with a solid surface. The atom may, for example, become chemisorbed to the surface. We are justified in excluding such systems from the definition of adhesion which we are considering here, for such molecules are not commonly said to be in a "state of adhesion", but in an "adsorbed state".

C. Definition of Adhesion

The noun, adhesion, can now be defined as follows:

Adhesion, n. 1) Macroscopic definition: The condition in which two macroscopic bodies adhere to each other, or one to the other, in accordance with Definitions 1 or 2, above.

2) Microscopic definition: The condition in which two phases adhere to each other, in the microscopic sense given above in Definition 3.

"Adhere" is occasionally used as an active verb.

Adhere, v., a. 1) An adhesive may be said to *adhere* one body to another if it converts the system in which the two bodies are separate (and no force or work can be transmitted from one to the other, except by gross mechanical interlocking such as the links of a chain) into a system in which such mechanical work or force can be so transmitted.

2) A person may be said to adhere to one body to another by coating one or both of the facing surfaces with an adhesive, bringing them into contact, and accomplishing the cure or setting of the adhesive. The same term may be used when one of the bulk bodies is of such a nature that no interlayer is needed, to bring the one body into a state of adhesion to the other.

III. ASSOCIATED TERMS

Much of the confusion that is apparent, in the multiplicity of common usages, can be resolved by formally assigning a number of concepts to other words, in accordance with accepted practice. For example, it is appropriate to separate out the concept of an adhesive joint.

An adhesive joint is a system in which *two* effectively parallel interfaces exist. It consists of two solids of macroscopic dimensions, which may or may not be of the same material, and a layer of adhesive, which may be of microscopic thickness, but in any case must be thin relative to the dimensions of the two solids that are joined. The composition of the adhesive is different from that of at least one of the bodies. The condition of adhesion exists with respect to the first body and the adhesive, and with respect to the second body and the adhesive. One does not say that the two bodies in an

adhesive joint adhere to each other. The shear modulus and strength of the cured adhesive, *in situ*, are properties which have prime importance, in conferring strength on an adhesive joint, even though the adhesive as a bulk phase may or may not have extremely high modulus or strength.

On the other hand, we commonly speak of the adhesion of one phase to another, in the absence of any thin interlayer which has a composition different from either bulk phase. There is only *one* interface, in such an adhering system. If one phase is of microscopic thickness, or at any rate, thin relative to the other phase, we call the thin phase a coating. It is obvious that the absence of a second bulk phase, and of a second interface, renders the system mechanically quite different from an adhesive joint.

Strength of adhesion and *adhesive joint strength* are to be distinguished by the specification that, with respect to the former term, the system consists of two adhering bodies with an interface between them; and for the latter, it consists of two macroscopic bodies with a thin layer of another material in between them, two interfaces being present in the system.

The thermodynamic property, *work of adhesion*, is defined by the Dupre equation⁵,

$$W_{12}^a \equiv -\Delta G_{12}^a = \gamma_1 + \gamma_2 - \gamma_{12} \quad (1)$$

where ΔG^a is the free energy change when one cm² of interface between phases 1 and 2 is created, out of one cm² of free surface of phase 1 and one cm² of free surface of phase 2. γ_1 is the specific surface free energy of phase 1, γ_2 is that of phase 2, and γ_{12} is that of the interface between phases 1 and 2.

Operationally, thermodynamic work of adhesion cannot be measured or even qualitatively observed, for a solid-solid system. (It bears no relation to the "work" that can be transmitted across a solid-solid interface in tension or in shear.) It has operational meaning only with respect to solid-liquid or liquid-liquid systems, and we have already shown that there is good reason not to include such systems in a definition that is relevant to solid-solid systems. See the recent comments of the author⁶ on the correlation between work of adhesion and force of adhesion.

IV. DISCUSSION OF "MECHANICAL ADHESION" AND "HOOKING"

The ASTM definition of "mechanical adhesion", quoted in Section I, refers to one function which an adhesive in a joint may, in many cases, perform. However, the relation of this function to the mechanical strength of the system may be misinterpreted; we will now examine the nature of this misinterpretation.

The words, "by an interlocking action", employed by ASTM D907-70, imply that the contribution of the adhesive is analogous to that in the interlocking in a dovetail joint, or the action of a hook or a chain link. The same implication is present in the discussion by Bikerman,³ under the heading of "hooking" adhesion. This analogy is not a valid one, for the following reasons:

First, in such macroscopic configurations as dovetailing, hooking, etc., there is negligible area of real interfacial contact. The boundaries of the miniscule regions of contact are lines where, under tensile or shear loading, there are concentrations of stress, which lead to separation under very small loading or absolute displacement.

Second, the mechanical strength of an interlocking or dovetailed system arises from geometric relations (and for chain loops, topological relations) *in combination with the existence of certain bulk properties*: high shear modulus and stiffness of the *bulk* phases, *independent* of the presence of an *adhesive*. As opposed to this, an adhesive, when tested in bulk, is often found to have relatively low (but nonzero) stiffness and shear modulus. Low values of these bulk mechanical properties are irrelevant to the *adhesion* that exists *between the adhesive and either bulk phase*.

Third, in simple mechanical hooking or interlocking, an immediate result of a reversal of stress is the loss of contact in those regions where intimate contact exists. This is followed, sooner or later, by establishment of contact in other regions of the surfaces of the two phases. This sequence of events needs to be distinguished from that in which, on load-reversal, intimate contact is maintained, at the phase boundary. Such a distinction can most easily and logically be made by restricting the term, adhesion, to the latter situation.

Since we have denied the direct relevance of bulk properties of a solid adhesive to *adhesion*, it is incumbent on the author to state what role bulk mechanical properties do play, in adhering systems. With respect to "mechanical adhesion" (the term introduced in the ASTM definition), the physical role that is played is to eliminate re-entrant voids at the interface. When this elimination is complete, there are no longer any stress concentrations (under loading) at the edges of voids. This contributes a great improvement in the mechanical strength of the system. The bulk mechanical property, which is involved here, is dilational strength of the adhesive, i.e., the negative pressure it can sustain without cavitation. The boundaries of a void that is filled are, essentially, fixed; so shear deformation is constrained, and dilational strength is converted to tensile strength. Thus, a region of extreme local weakness, due to stress concentration, is replaced by a region

of higher local strength. The contribution of the adhesive is far greater than what it could contribute by means of its practical, bulk shear strength or stiffness—which may, as noted above, be relatively low.

If we examine the ASTM definition closely, we see that it includes direct reference to “two surfaces” being “held together”; and in the *mechanical adhesion* definition, it explicitly mentions the presence of an adhesive. So we see that the ASTM did not distinguish between the *adhesion* in a two-phase system, and *adhesive joint strength*, and indeed was directed rather more to the joint-strength phenomenon. (See our discussion of this distinction, above.) For the purposes of scientific investigation of the causal factors that control strength of adhesion, we should isolate and exclude those factors that depend *directly* on the bulk properties of the adhesive *regardless of the presence of a second phase*, and on the *gross* geometric properties of the system. (This recommendation is simply the ancient principle of the scientific method, to “isolate the variables”.)

Accordingly, we recommend that the term, “mechanical adhesion”, be dropped from the scientific and engineering vocabulary and from ASTM D907. We recommend that a separate definition be introduced by the ASTM, pertaining to systems containing two bulk phases and an interlayer—e.g., “adhesive joint strength”. If the concept of a component of strength attributable to interlocking action is to be retained, it should be in the joint-strength definition, and not with regard to “adhesion”.

V. GENERAL DISCUSSION

A primary requirement, which must be observed particularly carefully with technical definitions, is freedom from circularity. A notable example of circularity is to define adhesion as a “sticking together”; see above. The mechanical definitions given above eliminate this logical defect: They are given in terms that are independent of the phenomenon.

A final and very important comment must be made on dualism: Do the macroscopic and microscopic definitions correspond to two distinct phenomena? This can be answered with the aid of reference to the modern theory of light and of the electron. An electron (and correspondingly, a photon) behaves like a particle in some experiments, and like a wave in others. Yet there are not two distinct entities. In the one case, it is useful to think of the electron as analogous to a macroscopic particle, e.g., a baseball with an electric charge; and in the other case, one may usefully employ the analogy of a wave on water. For adhesion, there are two different kinds of analogies that are (tacitly) employed by workers in the field. With respect to the microscopic definition, the analogy is to a chemical reaction involving

two atoms or molecules. (The ASTM refers to "valence forces" in its definition of "specific adhesion". With respect to the macroscopic definition, the analogy is to a one-phase solid which has appreciable tensile and/or shear strength. A full understanding of adhesion requires a comprehension of both the microscopic and the macroscopic aspects, just as a full understanding of the electron requires a comprehension of both its wave and its particle character.

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