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# Use of the Blister Test to Study the Adhesion of Brittle Materials. Part II. Application

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Using a modified form of the blister test, where the adhesive layer was between the substrate and a massive base, instead of as a continuous sheet on top of the substrate, we determined the interfacial fracture energy  $F$  for a series of interfaces where a brittle material (ice) was adhering to various substrates. Fracture energies obtained were compared with work of adhesion values measured for water on the same substrates. Fracture energy, which contains within it both a reversible contribution due to intermolecular interactions across the interface (work of adhesion) and an irreversible contribution due to collective dissipative processes, was found to rise rapidly with modest increases in work of adhesion. The observed relation suggests that the irreversible contribution to fracture energy is influenced strongly by the intermolecular interactions at the interface.

**KEY WORDS** Ice adhesion; work of adhesion; interfacial fracture energy; dissipative fracture processes; blister test.

## I INTRODUCTION

The adhesion of ice is of considerable practical importance in situations involving roadways, power lines, and ship and aircraft structures, to give just a few examples. Although this problem has been addressed over the years, there is still no clear understanding of which substrate properties are the dominant ones and how they influence ice adhesion. A major reason for this is that severe technical difficulties have interfered with scientific investigations of ice adhesion in the laboratory. These difficulties include conducting tests and using measuring instruments at low temperature, machining and gripping such a brittle material as ice, and preparing specimens where one of the constituents must be put in place as a very low viscosity liquid.

Fundamentally, there are several mechanisms that could influence the magnitude of ice adhesion. These are chemical bonding, mechanical lock and key

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interaction, interdiffusion, and intermolecular interactions between the ice and the substrate. For ice on most substrates, chemical bonding and interdiffusion do not contribute to adhesive bonding. The relevant mechanisms, then, are the lock and key interactions and the intermolecular interactions.

In the work described in this paper, we sought to study the adhesion brought about by intermolecular interactions alone, without the complicating effects of lock and key interactions. To do this, we used smooth substrates with a range of chemical structures. The smoothness eliminated lock and key interactions at the interface from consideration, and the different chemistry provided a range of intermolecular interactions.

## II BACKGROUND

The molecular adhesion across a planar interface is the summation of forces of interaction over all interacting sites. The types of forces that can play a role include dispersion, dipole-dipole, dipole-induced dipole, ion-dipole, acid-base, and hydrogen bonding. The energy of adhesion, or "work of adhesion," is the work required to overcome these forces and to move the two surfaces (*i.e.*, the collective interacting sites) an infinite distance apart in a reversible process. In doing this, two new surfaces are created, each having a characteristic surface energy. The work of adhesion  $W_{adh}$  is the difference between their summed individual surface energies,  $\gamma_1 + \gamma_2$ , and the energy  $\gamma_{12}$  at their interface when they are in the joined state. It is given by Eq. (1) below:

$$W_{adh} = \gamma_1 + \gamma_2 - \gamma_{12} \quad (1)$$

For solid materials the quantities on the right hand side of Eq. (1) cannot be determined directly. (This is because the process of separation is not a reversible one). Fortunately, there is another way to determine work of adhesion between two adhering materials. When one of the materials is in the liquid phase, the work of adhesion can be determined directly by wetting behavior.

For a liquid contacting a planar solid surface, the work of adhesion is given by:

$$W_{adh} = \gamma_L(1 + \cos \theta_a) \quad (2)$$

where  $\gamma_L$  is the surface tension (numerically equal to the surface energy) of the liquid, and  $\theta_a$  is the advancing contact angle made by the liquid on the solid surface. As long as the adhering liquid does not undergo a chemical change in the process of solidification, the work of adhesion given by Eq. (2) should be equivalent to that given conceptually by Eq. (1).

From the above discussion, we see that substrate surface energy is only part of the picture. In spite of the fact that substrate surface energy is often proposed as a key parameter in adhesive performance, by itself it cannot be regarded as an adequate indicator of the intermolecular interactions leading to adhesion. Because the properties of both of the adhering materials contribute to their

mutual adhesion, the work of adhesion as defined above is the appropriate quantity to use.

To be sure, in the art of adhesion, use is made of the assumed relation between substrate surface energy and adhesive bonding. To improve a weak bond, surface treatments that increase the surface energy of the substrate are often used. To prevent sticking, mold releases of low surface energy are often used. The success of these procedures probably partially explains the emphasis placed by practitioners on surface energy alone.

For assessing adhesive performance between two solid materials, the experimental work to separate unit area of interface must be determined. This is the interfacial fracture energy  $F$ , which differs from the work of adhesion in that it contains not only a reversible contribution (equivalent to the work of adhesion) but also an irreversible contribution associated with plastic deformation or other dissipative processes that occur at the crack tip as the two materials are separated. Obviously, measured values of  $F$  will exceed the work of adhesion by an amount depending on the size of the irreversible contribution to the separation process.

For the study of ice adhesion, the chief requirements are a mechanical test that provides a value for interfacial fracture energy and a fracture plane at the ice-substrate interface. The blister test, specially modified for bimetals systems where one constituent is brittle or fragile, is able to meet both of these requirements, and is described in Part I of this paper.

In past studies, these requirements have not been met. Tests were used which provided only the "breaking stress," a quantity of dubious value because it is dependent on such things as substrate thickness, ice layer thickness, substrate modulus, etc. In addition, much of the past data on joint failure was for cohesive fracture within the ice itself. While this might be of practical interest, it provides little insight into the nature of adhesion at the ice-substrate interface.

The requirement for interfacial failure is not easy to meet because of the tendency for ice-substrate joints to exhibit cohesive failure in the ice.<sup>1,2,3</sup> Cohesive failure in the ice occurs in preference to interfacial failure at temperatures below about  $-13^{\circ}\text{C}$  in a wide variety of joint geometries designed for both shear<sup>4-12</sup> and tension.<sup>11-13</sup> However, above  $-13^{\circ}\text{C}$  predominantly interfacial failure occurs, allegedly due to the formation of a "liquid-like layer." In our work, wanting to avoid the higher temperatures where such special effects come into play, we attempted to achieve interfacial failures at temperatures below  $-13^{\circ}\text{C}$ .

Originally we used the most straightforward version of the blister test, with the substrate surmounted by a sheet of ice, but found that changes were needed to achieve interfacial failure. We then inverted the order of materials, placing the substrate of interest on top of the ice. This modification made possible the collection of consistent and reproducible interfacial failure data for several different ice-substrate interfaces.

The details of the inverted blister test configuration and the fracture mechanics analysis are presented in Part I of this paper. The operative equation comes

directly from the fracture mechanics analysis and is shown below:

$$P_c = \sqrt{(EF)} \sqrt{f(h/c)/c} \quad (3)$$

where

$$f(h/c) = \frac{1}{(1-\nu^2)} \left\{ \frac{3}{32} \left[ \left( \frac{c}{h} \right)^3 + \left( \frac{c}{h} \right) \frac{4}{1-\nu} \right] + \frac{2}{\pi} \right\}^{-1}$$

In this equation  $P_c$  is the critical pressure (failure),  $E$  is the substrate elastic modulus,  $\nu$  is the substrate Poisson's ratio,  $h$  is the substrate sheet thickness, and  $c$  is the initial crack radius. The quantity  $\sqrt{f(h/c)/c}$  can be regarded as a geometry factor, and can be made to vary by using different values of  $h$  and/or  $c$  in the test geometry. A plot of  $P_c$  versus  $\sqrt{f(h/c)/c}$  will produce a good straight line whose slope is equal to  $\sqrt{(EF)}$ . The value of  $F$  can be computed directly from this slope.

### III EXPERIMENTAL

#### A. Materials used

For ice, ordinary tap water was used without any special preparation. Substrates were obtained in sheet form, as per the details given in Table I. They were washed with mild detergent before use.

#### B. Specimen preparation and testing procedure

The preparation and testing of the specimen shown in Figure 1 is described in Part I of this paper. Testing was done in a cold room at  $-20^\circ\text{C}$ . Both initial crack radius  $c$  and substrate thickness  $h$  were varied.

#### C. Modulus measurements

For the blister test in the configuration used here, with the substrate as the deformable layer on top, modulus values for each substrate were obtained at  $-20^\circ\text{C}$  as described in Part I of this paper.

TABLE I  
Information on substrates (sheets) used in blister test

Substrate	Thickness, mm	Comments
Stainless steel	0.30, 0.38	Shim stock, Precision Co., Downers Grove, IL
PMMA	1.90	Plastic sheet, Cadillac Plastic & Chem. Co., New York, NY
Polycarbonate	1.68	Plastic sheet, Cadillac Plastic & Chem. Co., New York, NY
Polystyrene	1.53	Plastic sheet, Cadillac Plastic & Chem. Co., New York, NY
Teflon <sup>®</sup>	1.58	Plastic sheet, Cadillac Plastic & Chem. Co., New York, NY

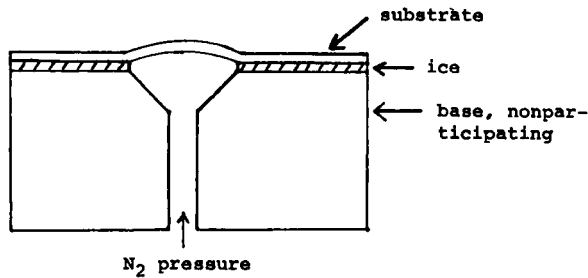


FIGURE 1 Blister test in the inverted configuration. The substrate, in the form of a thin continuous sheet, is on top. The brittle adhesive is present as a thin interlayer between the substrate and a massive base.

#### D. Work of adhesion determination

Contact angles were determined on the substrates of interest by the Wilhelmy wetting force method.<sup>14,15</sup> In our use of this method, a small piece of the substrate, in the form of a fiber or wire, was hung vertically from the arm of a recording electronic balance. A reservoir of the wetting liquid (water, surface tension = 72.4 dynes/cm) was brought into contact with the free end of the fiber, and was slowly raised so that the liquid surface advanced along the length of the fiber. The liquid travel rate was 0.167 mm/min, slow enough so that it did not affect the value of the contact angle. In this way, wetting force *versus* position along the fiber length was recorded. A single wetting force value, obtained by averaging the wetting force along several mm of fiber length, was used to compute advancing contact angle cosine,  $\cos \theta_a$ , for each fiber from the following relation:

$$\cos \theta_a = \frac{Mg}{\gamma_L \pi D} \quad (4)$$

where  $Mg$  is the wetting force of the liquid advancing along the fiber,  $\gamma_L$  is the surface tension of the probe liquid (measured independently), and  $\pi D$  is the perimeter of the fiber. Contact angle cosines from 8–10 fibers were averaged for each substrate. The average cosine values were then used in Eq. (2) to compute the work of adhesion,  $W_{adh}$ , between water and the substrate.

## IV RESULTS

Table II shows the modulus values measured at  $-20^\circ\text{C}$  for all the substrate materials. Although the low temperature values are the ones used in the computation of  $F$ , room temperature values were also measured and are shown in Table II for comparison. As expected, most moduli were not very temperature sensitive over the range tested. All values shown are averages of at least eight specimens.

TABLE II  
Modulus values for substrate materials (ave. ± 1 std. dev.)

Substrate	GPa at 20°C	GPa at -20°C
Teflon®	0.57 ± 0.02	1.32 ± 0.14
Polystyrene	1.20 ± 0.09	1.30 ± 0.07
Polycarbonate	1.45 ± 0.07	1.43 ± 0.12
Polymethyl methacrylate	1.66 ± 0.03	1.77 ± 0.24
Stainless steel	33.5 ± 2.9	33.4 ± 4.0

Figure 2-6 show the data, in the form of plots of  $P_c$  versus  $\sqrt{(f(h/c)/c)}$ , for ice adhered to five substrates. The plots give good straight lines, a fact that indicates that bulk viscoelastic effects can be neglected over the  $h/c$  range studied.

Table III presents the work of adhesion values for water on the substrates. The left hand column contains our values, obtained from the wetting force measure-

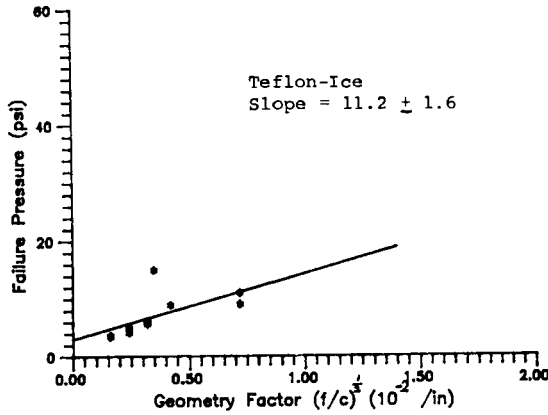


FIGURE 2 Critical pressure versus geometry factor for the Teflon®-ice interface.

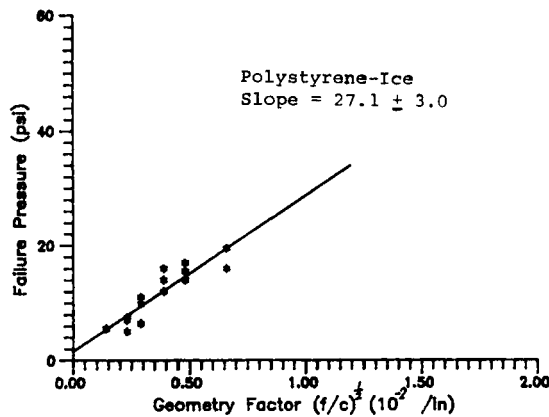


FIGURE 3 Critical pressure versus geometry factor for the polystyrene-ice interface.

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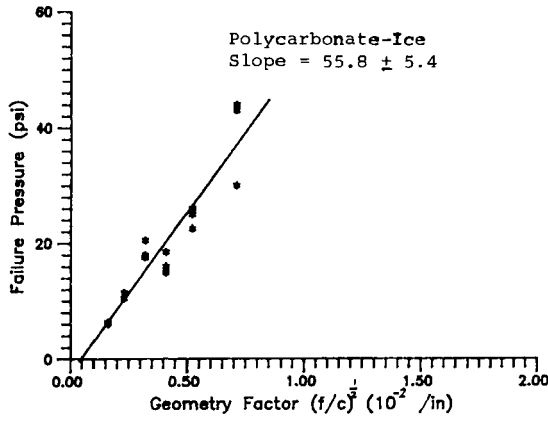


FIGURE 4 Critical pressure *versus* geometry factor for the polycarbonate-ice interface.

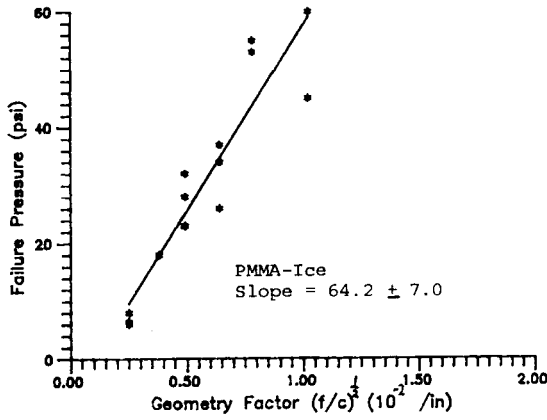


FIGURE 5 Critical pressure *versus* geometry factor for the PMMA-ice interface.

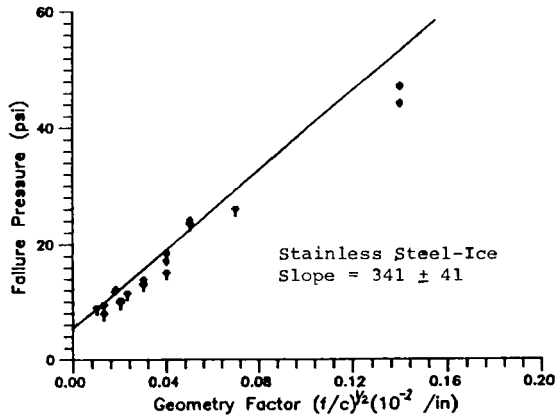


FIGURE 6 Critical pressure *versus* geometry factor for the stainless steel-ice interface. Data from two thicknesses of stainless steel sheet are plotted, for 0.30 mm and for 0.38 mm, with best fit line through all points.



TABLE III  
Work of adhesion of water on smooth substrates (ave.  $\pm$  1 std. dev.)

Substrate	$W_{adh}$ , mJ/m <sup>2</sup> †	$W_{adh}$ , mJ/m <sup>2</sup> ‡
Teflon®	45 $\pm$ 1	62
Polystyrene	70 $\pm$ 1	79
Polycarbonate	102 $\pm$ 2	128
PMMA§	—	132
Steel	107 $\pm$ 5	—

† Our data.

‡ From Ref. 11, no standard deviations given.

§ Not available in fiber form for wetting force measurements.

TABLE IV  
Slopes and values for  $F$  computed from inverted blister test data (ave.  $\pm$  1 std. dev.)

Substrate	$\sqrt{(EF)}$ , psi $\sqrt{\text{in}}$ †	C.V., %‡	$F$ , mJ/m <sup>2</sup>
Teflon®	11.2 $\pm$ 1.57	14	81
Polystyrene	27.1 $\pm$ 2.97	11	481
Polycarbonate	55.8 $\pm$ 5.42	9.7	1850
PMMA	64.2 $\pm$ 7.03	11	1980
Steel	341 $\pm$ 41	12	2220

† To convert to kPa  $\sqrt{\text{m}}$  multiply by 1.1.

‡ Coefficient of variation.

ments. For comparison, we have also included, in the right hand column, work of adhesion values from another laboratory, obtained by sessile drop measurements.<sup>1</sup> The key point made by Table III is that work of adhesion values for chemically different materials range over only a few decades. In fact, Eq. (2) points out that the  $W_{adh}$  values have an upper limit set by the work of cohesion of the contacting liquid itself. For water, the upper limit is about 144 mJ/m<sup>2</sup>.

Table IV lists the slopes extracted from the fracture data, and the values of  $F$  computed from the slopes for the substrate-ice interfaces. Note that the  $F$  values span a range of nearly two orders of magnitude, much larger than the work of adhesion range.

## V DISCUSSION

The goal of our work was to evaluate the role played by intermolecular interactions in ice adhesion. This role is embodied in Figure 7, where  $W_{adh}$  represents intermolecular interactions and  $F$  represents ice adhesion. Figure 7 shows that interfacial fracture energy appears to be a very strong function of work of adhesion. Even if the reversible contribution to the interfacial fracture energy is subtracted, the remaining irreversible contribution still rises rapidly with work

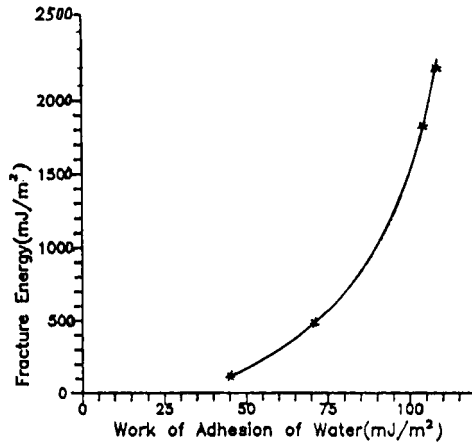


FIGURE 7 Interfacial fracture energy *versus* work of adhesion for an array of interfaces. The relation is by no means linear.

of adhesion. This suggests that the intermolecular interactions at the interface somehow influence the size of the irreversible processes that occur during fracture.

None of the work done by us so far gives an indication of the identity of the energy consuming process, *i.e.*, whether it is localized plastic flow, temperature rise, fractoemission, or other mechanisms that are not occurring in the bulk adhesive or adherend. Nor is it clear exactly how stronger interactions across the interface would act to increase the irreversible contribution. One speculation would be that stronger interactions at the interface induce a long range order penetrating into the ice or substrate bulk and that this order changes the way in which energy is dissipated near the crack tip. However, this speculation would seem to be naive since a material more ordered would seem to be less able to dissipate energy.

The results presented above offer a compelling arena for future work. Not only must it be discovered how energy is dissipated, but also if the influence of work of adhesion on fracture energy is a general phenomenon for all materials and not just for ice.

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