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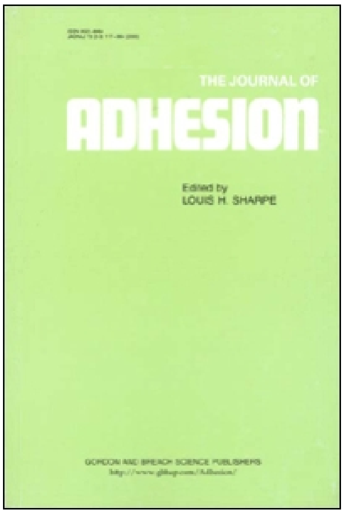

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The Strength of Liquid Bridges Between Dissimilar Materials

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The strength of the liquid bridge between a sphere and a plate of dissimilar materials was studied. An equation was derived using the surface energy approach. For small amounts of liquid, the force of adhesion f was

$$f = 2\pi R\gamma(\cos\theta_1 + \cos\theta_2)$$

where R is the sphere radius, γ is the surface tension, and θ_1, θ_2 the contact angles. In the derivation, major simplications about the meniscus shape were possible.

The equation was experimentally tested with water, ethyl alcohol, aniline and iodobenzene using factorial combinations with different solids. Force of adhesion measurements were carried out using a tensile testing machine at controlled loading rates. Excellent agreement was obtained in the experimental and predicted adhesion values. The McFarlane–Tabor equation was identified as correct only for small amounts of liquids and similarly wet solids.

INTRODUCTION

Traditionally, the strengths of thin liquid bridges between solids has been treated as a capillary phenomenon. The Fisher equation for the tensile strength (force) of liquid bridges between two spheres contains two terms[1]:

$$f = \pi R^2 \Delta P \sin^2\psi + 2\pi R\gamma \sin\psi, \qquad (1)$$

where

R is the radius of the sphere
ΔP is a reduction in pressure beneath the meniscus
ψ is the half-centriangle to the meniscus
γ is the liquid surface tension

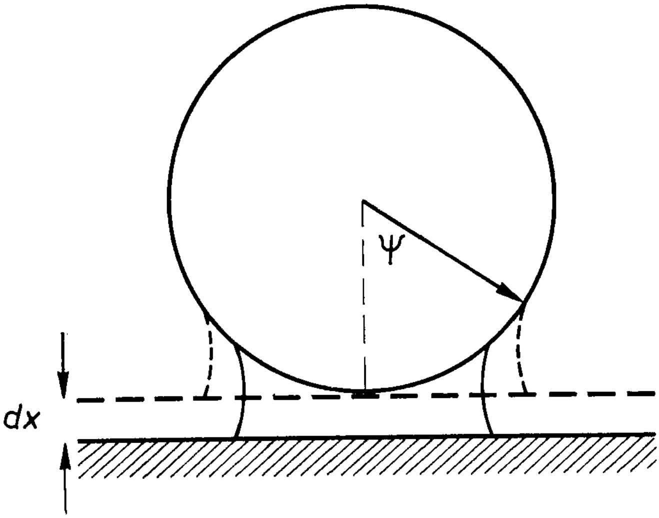

FIGURE 1 Liquid bridge between lens and flat plane.

The first term is the reduced pressure component due to the curvature of the meniscus whereas the second expresses the surface tension pull effect. An equation for the strength of a liquid bridge between a sphere and a plate was derived and tested by McFarlane and Tabor[2]:

$$f = 4\pi R\gamma \cos\theta. \tag{2}$$

Their equation does not contain a surface tension pull term. The case of two dissimilar solids held together with a liquid bridge has not been treated because of the complex shape of the meniscus.

Another approach used successfully in treating capillary phenomena is the surface energy method.[3] This method focuses attention on surface energy changes taking place at interfaces rather than on the meniscus curvature. O'Brien, Craig and Peyton[4] have recently used this method to derive an equation for the penetration of liquids between dissimilar materials which resisted solution by the curvature approach.[5]

The purpose of this study was to develop and test an equation for the strength of liquid bridges between a sphere and a plate composed of dissimilar materials using the surface energy approach.

The equation for the strength of this bridge based on an increased surface tension may be derived with reference to Figure 1. The increased liquid surface tension, $\gamma_L{}^*$, is given by the relation[3]

$$\gamma_L{}^* = \gamma_L + \frac{(\gamma_{SV} - \gamma_{SL})A_{SL}}{A_{LV}} \tag{2}$$

where $(\gamma_{SV} - \gamma_{SL})$ represents the free energy term on wetting and A_{SL} and A_{LV} are the solid–liquid and liquid–vapor interfacial areas. The value of γ^* is obtained from Eq. (2) using the following geometrical approximations for low ψ values

$$A_{LV} = \pi^2 sb, \tag{3}$$

and

$$A_{SL} = 2\pi s^2. \tag{4}$$

Substituting $\gamma \cos \theta$ from the Dupre equation for $(\gamma_{SV} - \gamma_{SL})$ gives the following

$$\gamma^* = \gamma + \frac{2s\gamma \cos \theta}{\pi b} \tag{5}$$

The work necessary to stretch the meniscus film by an amount δb is equated to the increase in surface energy of the meniscus

$$f\delta b = \gamma^* \pi^2 s \delta b, \tag{6}$$

where $\pi^2 s\delta b$ is the increase in surface area.

Substituting the value of γ^* from Eq. (5), the following is obtained

$$f = \gamma\pi^2 s + \frac{2\gamma \cos \theta \pi s^2}{b} \tag{7}$$

At angles of ψ of a few degrees

$$s^2 = 2Rb, \tag{8}$$

giving

$$f = \gamma\pi^2 s + 4\pi R\gamma \cos \theta. \tag{9}$$

With low liquid volumes, the first term is minor and the final equation is identical to the Tabor equation[2]

$$f = 4\pi R\gamma \cos \theta. \tag{10}$$

In the case of two dissimilar materials

$$f = 2\pi R\gamma (\cos \theta_1 + \cos \theta_2) \tag{11}$$

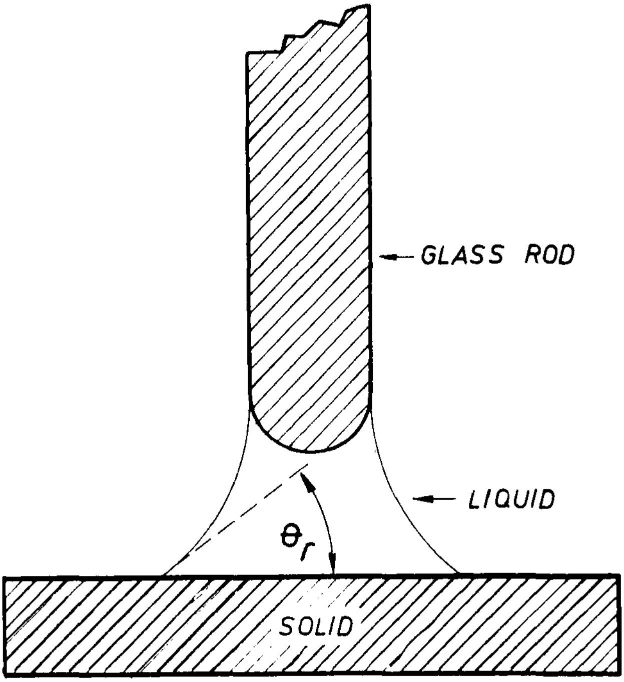

FIGURE 2 Method of measuring receding contact angle θ_r.

MATERIALS AND METHODS

Measurement of parameters

1) *Contact angles* Receding contact angles for the different solid–liquid interfaces were measured by the following method. A drop of liquid was placed at the end of a glass rod which was freely suspended. The plane solid surface was raised until it was in contact with the glass rod. The plane solid was then slowly lowered. This position is illustrated in Figure 2. The receding contact angle was measured by means of a coordinate cathetometer which was equipped with a protractor eyepiece on a telemicroscope.†

The liquids used were carbon dioxide free distilled water,‡ absolute ethyl alcohol,§ reagent grade iodobenzene‡ and aniline.‡ Solids on which the

---

† Model 1238–1818 Cathetometer; Gaertner Scientific Corp.; Chicago, Ill.

‡ Distilled Water; Iodobenzene; Aniline; Reagent Grade; Fisher Scientific Company; Chicago, Ill.

§ Absolute Ethyl Alcohol; Reagent Grade; U.S. Industrial Chemicals Corp., New York, N.Y.

liquids were placed were plane glass plates†, acrylic resin‡ and Teflon sheets,‡ and silicone treated glass.§

2) *Surface tension* The Wilhelmy Plate principle was employed to measure the surface tension of the liquids.|| A wettable blade is immersed in the liquid and then slowly withdrawn. The liquids were kept at 20°C during all measurements.

3) *Radius of lens curvature* The radius of curvature of the plano-convex glass lens was determined by means of a spherometer.††

4) *Force of adhesion* A tensile testing instrument,‡‡ as shown in Figure 3, was used to measure the adhesion forces at controlled strain rates. The strain rates were 0.01, 0.005 and 0.002 inches per minute. These different strain rates were used to check the influence of the viscosity of the liquid on the force of adhesion.

The adhesive systems consisted of a plano-convex glass lens resting on a plane solid surface. The plano-convex glass lens was attached to the load cell of the instrument by means of a rigid brass holder. The flat solid was mounted on a brass socket which was kept at 20°C by circulating water of the given temperature through this metal base. The experimental arrangement is shown in Figure 4. A known amount of liquid, 0.020 ml, was placed on the glass lens with a syringe and the two solids were brought together. The two solids were then separated at a preselected strain rate and the force of adhesion recorded on a strip chart recorder.

Special care was taken with respect to the cleanliness of the surfaces of the solids. The glass surfaces were boiled in a cleaning solution§§ containing sodium dichromate sulfuric acid for at least two hours. The glassware was then rinsed under a jet of hot tap water and successively rinsed with distilled water. The surfaces of the acrylic and Teflon sheets were only rinsed with distilled water. After cleaning, the solids were dried and stored in a desiccator which was purged with dry and purified nitrogen gas.

RESULTS

1) *Contact angles* Six contact angle measurements were taken for each solid-liquid combination. The values of the receding contact angles and

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† Glass Plates; Plano-Convex Glass Lenses, Lando Optical Corp., Milwaukee, Wisc.

‡ Acrylic and Teflon Sheet; Cadillac Plastics Company; Milwaukee, Wisc.

§ Silicone Resin; Dri-Film 88; General Electric; Waterford, N.Y.

|| Roller Smith Precision Balance; Federal Pacific Electric Co.; Newark, N.J.

†† Spherometer; Model 73360; Central Scientific Co.; Chicago, Ill.

‡‡ Tensile Testing Instrument; Model TM; Instron Corp.; Canton, Mass.

§§ Cleaning Solution; Fisher Scientific Co.; Chicago, Ill.

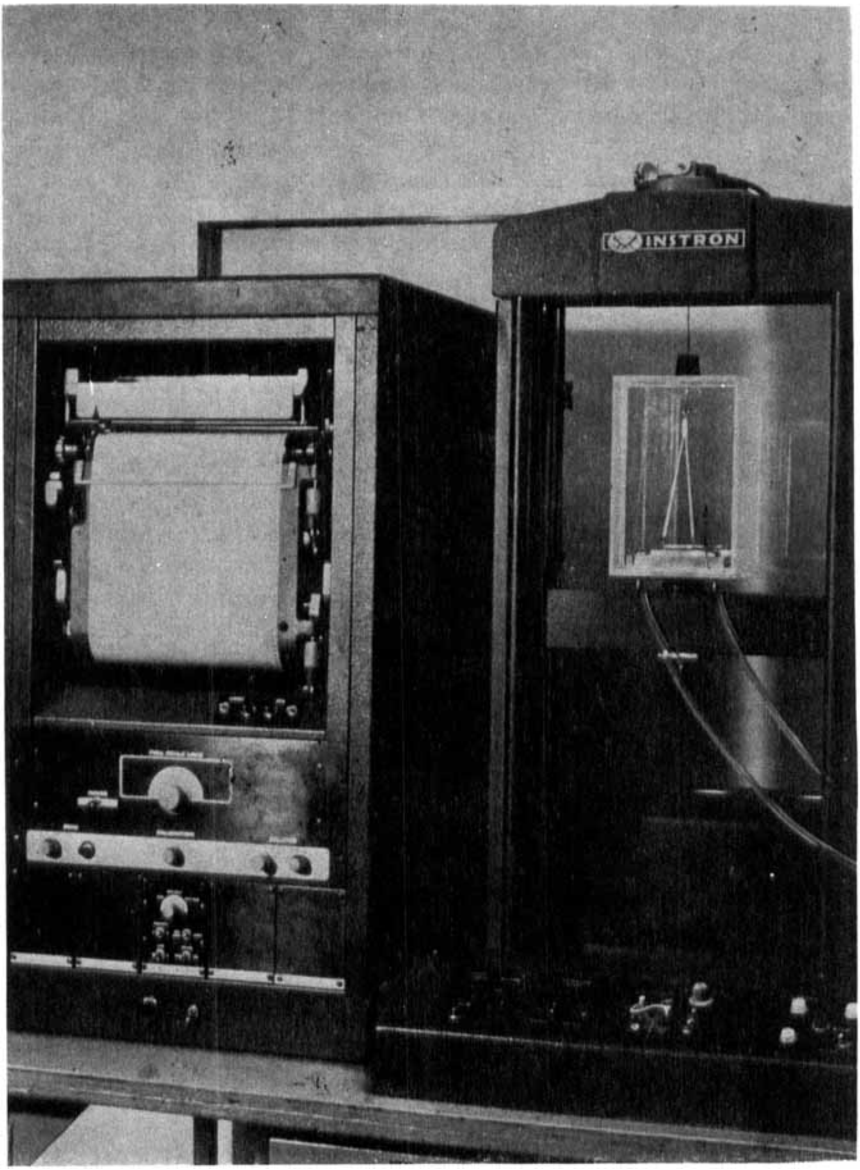

FIGURE 3 Tensile testing equipment used for adhesion measurements.

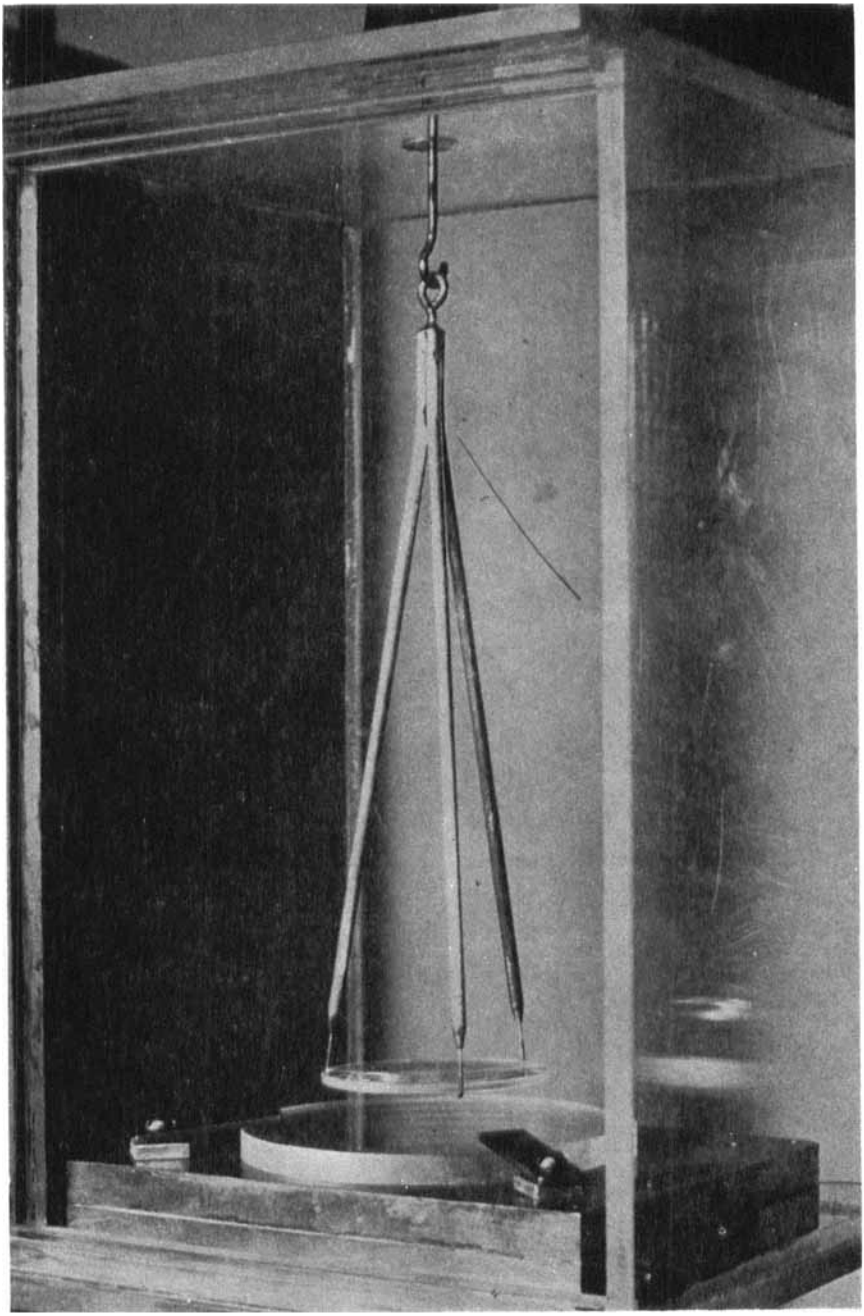

FIGURE 4 Arrangement for measuring adhesion between lens and plane surface.

TABLE I

Receding contact angles of liquids on various solids at 20°C.

Solid	Liquid	θ_r Degrees	s.d. Degrees
Glass	Water	6	3
Acrylic	Water	56	1
Silicone	Water	92	1
Teflon	Water	95	1
Glass	Ethanol	Spread	
Acrylic	Ethanol	Spread	
Teflon	Ethanol	24	2
Glass	Iodobenzene	Spread	
Teflon	Iodobenzene	52	4
Glass	Aniline	16	3
Teflon	Aniline	55	3

standard deviations are listed in Table I. Ethanol completely spread on glass and acrylic, thus forming a zero contact angle. Teflon and silicone coated glass are poorly wetted by water, their respective receding contact angles being 95° and 92°.

2) *Surface tension* The surface tension of water, ethanol, iodobenzene, and aniline were found to be 72.2, 22.3, 39.3 and 42.7 dynes per centimeter at 20°C. Values reported in literature[7] are 72.8, 22.3, 39.7 and 42.9 dynes per centimeter at 20°C.

TABLE II

Calculated and mean observed values for glass lens of radius 24.2 cm. Standard deviation for nine measurements. Values in dynes $\times 10^{-3}$

		Glass			
	Water			Aniline	
Glass	Acrylic	Silicone	Teflon	Glass	Teflon
21.8[a]	17.1[a]	10.5[a]	9.9[a]	12.5[a]	10.0[a]
20.6[b]	16.6[b]	10.5[b]	8.8[b]	12.1[b]	9.2[b]
0.072[c]	0.228[c]	0.147[c]	0.184[c]	0.171[c]	0.151[c]

		Glass			Silicone
	Ethanol		Iodobenzene		Water
Glass	Acrylic	Teflon	Glass	Teflon	Acrylic
6.8[a]	6.8[a]	6.5[a]	12.0[a]	9.7[a]	5.9[a]
6.8[b]	6.8[b]	5.8[b]	11.8[b]	9.7[b]	5.7[b]
0.170[c]	0.148[c]	0.155[c]	0.098[c]	0.349[c]	0.252[c]

[a] Calculated values
[b] Mean observed values
[c] Standard deviation

3) *Radius of lens curvature* Two plano-convex glass lenses were selected. Their radius of curvature was found to be 24.2 cm and 13.2 cm.

4) *Force of adhesion* Adhesion forces for the systems are listed in Table II and Table III. Also included in Table II and Table III are the expected values for the force of adhesion which were calculated by substituting the experimental values of the receding contact angles, surface tension and radius of lens curvature into Eq. (11). The experimental data listed in Table II was obtained using the glass lens of radius 24.2 cm. The calculated film thickness in all systems was 1.60 x 10^{-2} cm. The calculated and observed forces of adhesion and standard deviation values are in dynes and are to be multiplied by 10^3. The mean observed forces are calculated using nine measurements. The mean observed forces are calculated using nine measurements. Three measurements were taken for each of the three previously mentioned strain rates. No significant difference in the adhesion forces was found for the different strain rates in any of these systems. Thus, it was concluded that, with the low strain rates of 0.01, 0.005 and 0.002 inches per minute, the viscosity of the liquids had no influence on the forces of adhesion.

TABLE III

Calculated and mean observed values for glass lens of radius 13.2 cm. Standard deviation for three measurements. Values in dynes × 10^{-3}

	Glass				
Water			Aniline		
Glass	Acrylic	Silicone	Teflon	Glass	Teflon
11.9^a	9.3^a	5.7^a	5.4^a	6.8^a	5.4^a
11.7^b	9.2^b	5.7^b	5.4^b	6.8^b	5.2^b
0.098^c	0.559^c	0.133^c	0.057^c	0.098^c	0.057^c

	Glass				Silicone
Ethanol			Iodobenzene		Water
Glass	Acrylic	Teflon	Glass	Teflon	Acrylic
3.7^a	3.7^a	3.5^a	6.5^a	5.3^a	3.1^a
3.5^b	3.7^b	3.5^b	6.6^b	5.3^b	2.7^b
0.098^c	0.000^c	0.000^c	0.057^c	0.000^c	0.100^c

[a] Calculated values
[b] Mean observed values
[c] Standard deviation

The experimental adhesion forces listed in Table III were measured using a glass lens of radius 13.2 cm and represent a mean value composed of three individual measurements. The calculated film thickness for these systems was

2.20×10^{-2} cm. The strain rate employed for these measurements was 0.002 inches per minute. Again, all numbers in Table III are to be multiplied by 10^3 in order to obtain the proper values in dynes. Figure 5 illustrates the

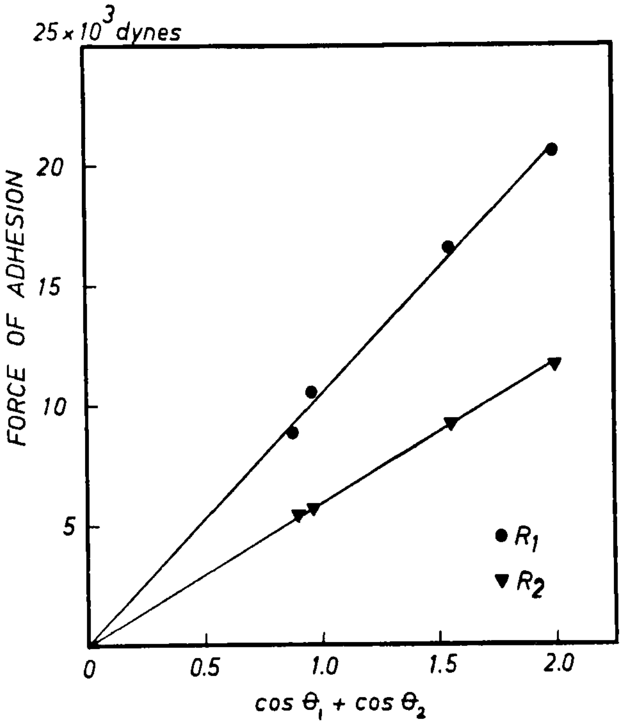

FIGURE 5 Adhesive force as a function of ($\cos \theta_1 + \cos \theta_2$).

linear relationship between the force of adhesion and the sum of the cosines of the two contact angles. The two curves in this figure are graphical representations of the data obtained for the systems employing water as the adhesive, with each curve corresponding to a particular lens radius.

The evaluation of the derived equation for the force of adhesion between dissimilar solids may be performed by appraising its predictive ability. The experimentally determined adhesion forces are compared with the values which were obtained by substituting the values of receding contact angles, liquid surface tension and radius of lens curvature into the equation

$$f = 2\pi R\gamma (\cos \theta_1 + \cos \theta_2) \tag{11}$$

and solving for the force of adhesion. The experimental and calculated adhesion forces are given in Table II and Table III. In Figure 6, the calculated adhesive forces are plotted against those that were experimentally determined.

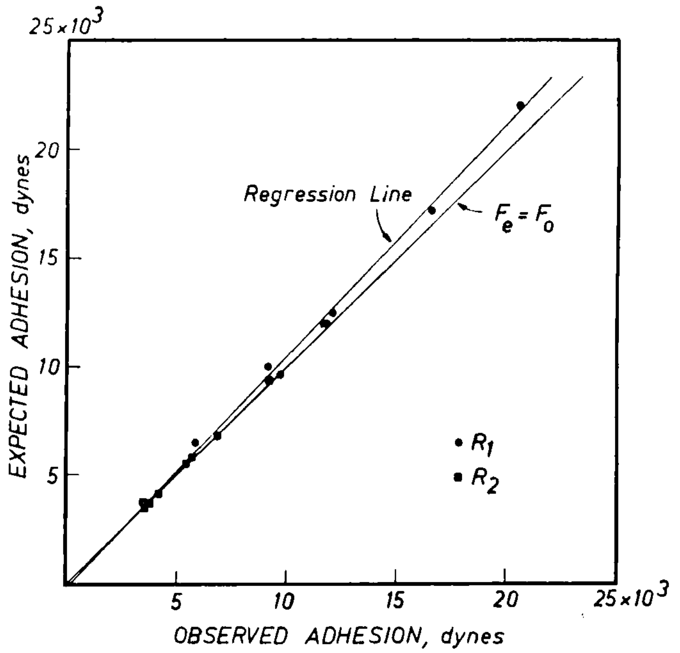

FIGURE 6 Regression of expected adhesion forces on observed adhesion forces.

The degree of association between the calculated and experimental adhesion forces was first tested by determining the equation for the regression line that best conformed to all data points. Using the least squares method, the regression line for the calculated values on the observed values was found to be:

$$f_c = 1.04 f_e - 0.10 \tag{12}$$

Here, the subscripts *c* and *e* refer to calculated and experimental, respectively. The regression line is shown in Figure 6 along with the line for the calculated force as equal to the observed force. The slope of the regression line is 1.04. This figure, being close to unity, indicates a close 1:1 correspondence of the expected to the observed values.

The absolute magnitude of the regression coefficient alone is not sufficient to determine whether a correlation is strong or weak. The correlation coefficient was obtained from an analysis of variance.

The correlation coefficient for all the data listed in Table II and Table III was calculated to be 0.99. Therefore, the expected values correlate highly and significantly with the observed values in almost a 1:1 ratio and the newly derived Eq. (11) appears to be satisfactory.

DISCUSSION

The surface energy approach led to an equation for the strength of liquid bridges between a sphere and a solid:

$$f = 2\pi R\gamma (\cos\theta_1 + \cos\theta_2) \qquad (11)$$

The McFarlane–Tabor equation is a special case when ψ is low and $\theta_1 = \theta_2$,

$$= f4\pi R\gamma \cos\theta. \qquad (2)$$

The experimental data were in reasonable agreement with Eq. (11) when tested for small ψ values and neglecting the first term.

As with capillary equations derived using the surface energy approach, major simplifications about the same shape of the meniscus are possible in deriving the traditionally accepted equation. The Fisher equation was recently derived using this approach.[8] The derivation casts doubt on the traditional practice of treating interfacial curvature as the driving force for capillary phenomena.

Acknowledgement

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