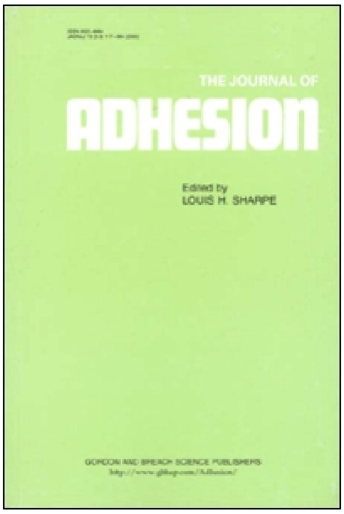
This article was downloaded by:
On: *22 January 2011*
Access details: *Access Details: Free Access*
Publisher *Taylor & Francis*
Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



The Journal of Adhesion

Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713453635

The Effect of Contamination on Adhesive Strength: Wettability Characterization by the CSC Method

Abraham Marmur[a]; Hanna Dodiuk[b]; Dora Pesach[a]
[a] Department of Chemical Engineering, Technion-Israel Institute of Technology, Haifa, Israel and Rafael [b] Department of Chemical Engineering, Haifa, Israel

To cite this Article Marmur, Abraham , Dodiuk, Hanna and Pesach, Dora(1987) 'The Effect of Contamination on Adhesive Strength: Wettability Characterization by the CSC Method', The Journal of Adhesion, 24: 2, 139 — 153
To link to this Article: DOI: 10.1080/00218468708075423
URL: http://dx.doi.org/10.1080/00218468708075423

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: http://www.informaworld.com/terms-and-conditions-of-access.pdf

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

J. Adhesion, 1987, Vol. 24, pp. 139–153
Photocopying permitted by license only

Printed in the United Kingdom

The Effect of Contamination on Adhesive Strength: Wettability Characterization by the CSC Method

ABRAHAM MARMUR,[1] HANNA DODIUK[2] and DORA PESACH[1]

Department of Chemical Engineering,[1] Technion—Israel Institute of Technology, Haifa 32000, Israel, and Rafael,[2] P.O. Box 2250, Haifa 31201, Israel

(Received July 10, 1986; in final form March 14, 1987)

The correlation between the adhesive strength of joints of aluminum alloys and the wettability of their surfaces was studied. Two alloys, Al-1100 and Al-2024, were used, with two commercial epoxy adhesives, FM-73 and FM-300K, and the BR-127 primer. The wettability of the surfaces was modified by applying coatings of silicone oil and stearic acid at various concentrations. Wettability was characterized by the "complete spreading concentration" (CSC) method and by advancing contact angles. The CSC method was proven to be more reliable than contact angle measurements in detecting variations in the surface energy of the aluminum alloys studied. The adhesive strength was measured by the lap shear strength (LSS) and by the T-peel strength. The adhesive strength is only mildly sensitive to the concentration of silicone oil for the two adhesive systems. For FM-300K, the LSS decreases when the concentration of stearic acid in the coating increases. For FM-73, the LSS is only mildly sensitive to the concentration of stearic acid, but the T-peel strength shows appreciable sensitivity. The failure mode becomes more adhesive as the concentration of stearic acid and thereby the CSC increase. It is only mildly dependent on the concentration of silicone oil. The results indicate that silicone oil probably interacts with either the primer or an adhesive component in a way which counteracts the expected decrease in adhesive strength due to the reduction in surface energy and wettability of the adherends. However, the effect of stearic acid on the adhesive strength is associated with decreased wettability. All cases for which a pronounced decrease in the adhesive strength was measured are associated with contact angles larger than 90° and with either high CSC values or nonspreading situations.

KEY WORDS Effect of contamination; epoxy film adhesives; joint strength; silicone oil; stearic acid; wettability characterization.

INTRODUCTION

It is generally accepted[1-10] that adhesion is closely related to the interfacial properties of the adherends. As the purpose of an adhesive is to bridge between two solid surfaces by forming an intimate contact with each of them, good wetting of the solids by the adhesive is a natural perequisite.

To establish a correlation between the adhesive strength and the wettability of the surfaces of the adherends, reliable measures of these properties are required. While reasonable standardization exists regarding measures of adhesive strength,[7] the characterization of wettability is still fraught with some difficulties. Contact angle measurements have been frequently used for characterizing the surface energy of solids,[11] however a major difficulty is caused by the phenomenon of contact angle hysteresis, which may lead to a large range of possible contact angles. The interpretation of the measured contact angle is therefore uncertain, especially under conditions which enhance hysteresis, such as surface roughness or contamination, which frequently occur in industrial practice.

A method which has gained popularity in characterizing the surface energy of solids was developed by Zisman and coworkers.[12] This method involves measurements of contact angles of a homologous series of liquids on the solid to be characterized. The results are plotted in the form of $\cos\Theta$ *vs.* γ, where Θ is the measured contact angle and γ is the surface tension of the liquid. Extrapolation of the curve to $\cos\Theta = 1$ defines a corresponding surface tension, which is termed the critical surface tension. The latter is used to characterize the surface of the solid, in the sense that every liquid of γ smaller than the critical surface tension will completely wet the solid. This method involves the same uncertainty as mentioned above, regarding the identification of the true value of the contact angle within the hysteresis range.

A possible way to overcome the uncertainty in contact angle measurements is to look for a liquid, the equilibrium contact angle of which is exactly zero on the surface which is studied. The equilibrium zero contact angle which is required for the characterization of a surface should not be confused with the apparent zero contact angle, which may be shown by a liquid of sufficiently low surface tension, while dynamically spreading on the solid. It is unlikely, however, to find a suitable pure liquid for each solid surface, which will have $\Theta = 0$ at equilibrium. Therefore, it is natural to solve the problem by using a binary mixture of liquids, one which completely spreads on the solid surface and one which does not. By adjusting the concentration of the mixture, it is possible to reach the transition point between non-zero contact angles and complete spreading, which may be assumed to represent the situation of $\Theta = 0$ at equilibrium. The surface energy of the solid can then be empirically characterized by the lowest concentration of the less polar component which induces complete spreading. This concentration is termed the "complete spreading concentration" (CSC). Two typical mixtures which may cover a wide range of situations are water-ethanol and ethanol-octane. The CSC for the former is defined as the lowest ethanol concentration which leads to complete spreading, and for the latter it is the lowest octane concentration. The use of mixtures rather than pure liquids may involve some additional complications, such as selective adsorption or evaporation of one of the components. Therefore, the method should be carefully studied and empirically tested.

The CSC method has been used in industry, but its sensitivity and reliability have been tested only in a limited number of cases.[13–15] For polyethylene, for example, it was found that the CSC method was much more sensitive to variations in the surface energy due to oxidation than contact angle measurements.[13] The CSC method appears similar to Zisman's method, however two major differences exist between them: (a) the point of $\Theta = 0$ is experimentally identified in the CSC method, rather than by graphical extrapolation; (b) no instrumental measurement of contact angles is being made in the CSC method, since the CSC can be easily determined by visual inspection. Zisman and coworkers[12] also used mixtures of liquids in their studies, in addition to homologous series of pure liquids. However, they were used to characterize surfaces which were lower in surface energy than the surface tension of either of the components in the mixture. Thus, the condition $\Theta = 0$ was again found by extrapolation and not by direct experimental observation, as in the CSC method.

Recently, a series of studies on surface characterization of aluminum alloys (Al-1100 and Al-2024) treated by various anodizing processes has been published. The microstructure of aluminum oxide and aluminum oxide-primer-adhesive interfaces have been evaluated, using various spectroscopy techniques (ESCA, SIMS, Auger, FTIR, SEM and EDXA), and its correlation with the adhesive strength has been elucidated.[16–20] It is of practical interest to correlate the adhesive strength of these aluminum adhesive joints with the wettability of the treated surfaces, since the latter can be characterized by techniques which are much simpler than the above mentioned spectroscopy methods.

The purpose of this paper is thus twofold: first, to establish the use of the CSC method and its reliability in characterizing surfaces of aluminum alloys treated with various controlled contaminations; and mainly, to correlate the CSC with the adhesive strength.

EXPERIMENTAL

Contact angle measurement

The measurement of contact angles was performed with a direct reading contact angle meter (Kernco Instruments Co. Inc.). A drop of distilled water, about 3 μl in volume, was placed on the test surface, using a micrometric syringe. The drop volume was slowly increased by adding water, and the advancing angle was recorded. The latter was defined as the highest equilibrium contact angle which could be assumed by the drop, given an initial drop volume. Three samples were used for each measurement.

Complete spreading concentration (CSC) determination

The determination of the CSC was based entirely on visual inspection, since it is easy and accurate to determine the existence of complete spreading. This was accomplished by noting the large basal area of the drop and the time required for spreading. In situations involving a non-zero contact angle, the equilibrium configuration is usually attained within less than a second. In complete spreading situations, the motion usually continues for about 10 sec or more.

Ethanol-water mixtures of various concentrations were prepared and carefully stored, to avoid significant changes due to evaporation. A drop of each was put on a test surface (one drop only on each surface), in a monotonic order of concentrations, until the lowest concentration of ethanol which led to complete spreading was identified. The surface was characterized at this point by two limiting solutions: one with the lowest ethanol concentration which led to complete spreading, and the other with the highest ethanol concentration which still led to a non-zero contact angle. Fine tuning between these limiting solutions could be made by decreasing the concentration intervals between two successive solutions, until satisfactory differentiation was reached. Three samples were used for each final measurement. The CSC values reported in the tables below are the concentrations of the limiting solutions. The CSC values are given in volume percent of ethanol, referring to the volumes of the liquids prior to their mixing.

Adherends and adhesives

Two adherends were used for the present study: Al-1100 and Al-2024, both chromic acid anodized without sealing, in accordance with MIL-B-8625.

Two commercial film adhesives were studied: FM-73, a toughened, supported, modified epoxy film adhesive for 250°F service; and FM-300K, a supported, modified epoxy film adhesive for 350°F service (Bloomingdale Division of American Cyanamid). The primer was BR-127 (Bloomingdale Division of American Cyanamid).

A thin layer of the BR-127 primer was applied by spraying and cured for 30 min at room temperature, followed by 1 hour at 120°C. FM-73 was cured at 120°C and 0.25 MPa for 1.5 hours. FM-300K was cured at 177°C and 0.25 MPa for 2 hours.

Mechanical tests

Tensile lap shear specimens were prepared according to ASTM D-1002-72. T-peel specimens were prepared according to ASTM D-3167-73T. Five specimens were fabricated for each test, using a special mold under compression. Bondline thickness for all specimens was 0.10 + 0.03 mm.

The bond strength was measured by using an Instron Mechanical

Tester (crosshead speed was 2 mm/min for lap shear strength and 200 mm/min for T-peel strength) at 25°C. The mode of failure (adhesive or cohesive) was evaluated by visual inspection.

Cleaning and coating procedures

Cleaning was achieved by immersing the samples in air plasma at 0.2 torr, formed by applying 3 kV AC, at 50 Hz, for a predetermined period.

The coating solutions consisted of silicone oil or stearic acid, dissolved in chloroform. The concentration of each coating solution is expressed by volume percentage, in terms of the volumes of the constituents prior to their mixing. Coating of the samples for the lap shear tests was performed by dipping into the solution for 10 min. The T-peel samples were large, and therefore their coating was done by slowly pouring about 75 ml of solution on each side.

RESULTS AND DISCUSSION

The main purpose of the present paper is to present the correlation between the adhesive strength and the wettability of the surfaces of the adherends, measured in terms of the "complete spreading concentration" (CSC). However, it is first of interest to establish the usefulness of the CSC method for characterizing the surface energy of treated surfaces of aluminum alloys, in comparison with the well known technique of contact angle measurement.

Figure 1 presents the CSC *vs.* the advancing contact angles for two aluminum alloys, Al-1100 and Al-2024, and two types of controlled contamination, silicone oil and stearic acid. It is clear that, in general, a reasonable correlation exists between the two measurements, in the sense that the advancing contact angle tends to increase when the CSC increases. Specifically, both methods show that the surface energy of the stearic acid coating is lower than that of the silicone oil coating. However, a detailed examination of these results, with reference to the concentrations of contaminants as reported in Tables I and II, reveals further details. It can be seen that, in some cases, very different contact angles correspond to the same CSC. The reason is that the CSC varies monotonically with

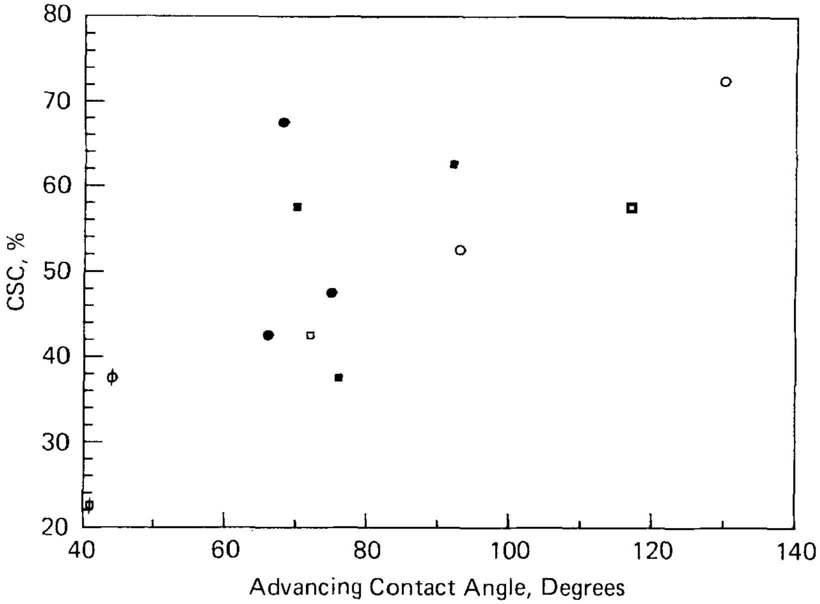


FIGURE 1 The correlation between the CSC and the advancing contact angle. ○—Al-1100 samples. □—Al-2024 samples. Full symbols—silicone oil. Empty symbols—stearic acid. Divided symbols—plasma cleaning.

the concentration of the contaminant, while the advancing contact angle is not always a unique function of the latter. For example, the CSC for Al-1100 increases with the concentration of silicone oil in the coating solution, while the contact angle for 1% silicone oil is smaller than for 10^{-2}%. Thus, the CSC is more reliable in characterizing the wettability of the surfaces studied here than the advancing contact angle, in accordance with previously published data on other systems.[13,14]

It is also of interest to note that the advancing angles reported here for high concentrations of stearic acid are higher than previously reported for packed monolayers of similar species.[12] This can be explained in terms of contact angle hysteresis, by realizing that the solid surfaces used here were rougher than those usually used for monolayer deposition, and that the present coating procedure most probably led to a nonuniform coating. This observation may serve to emphasize again the advantage of the CSC

TABLE I
Wettability, lap shear strength (LSS) and failure mode[a] of treated Al-1100

Surface Treatment[b]	θ°	CSC %	LSS, MPa FM73 + BR127 (Failure mode)	LSS, MPa FM300K + BR127 (Failure mode)
Plasma Cleaning	44	35–40	2.51 ± 0.17 (60%A)	2.03 ± 0.02 (70%A)
SO 10^{-5}%	66	40–45	3.35 ± 0.08 (20%A)	2.40 ± 0.07 (35%A)
SO 10^{-2}%	75	45–50	3.17 ± 0.06 (15%A)	2.51 ± 0.06 (40%A)
SO 1%	68	65–70	3.01 ± 0.03 (20%A)	2.33 ± 0.05 (50%A)
SA 10^{-5}%	93	50–55	3.48 ± 1.45 (20%A)	2.42 ± 0.02 (50%A)
SA 10^{-2}%	130	70–75	2.82 ± 0.10 (35%A)	0.87 ± 0.12 (95%A)
SA 10^{-1}%	113	NS[c]	2.94 ± 0.17 (70%A)	0.66 ± 0.06 (100%A)

[a] Failure mode in %A (percentage adhesive failure) is defined as the percentage of exposed substrate area which is devoid of adhesive.

[b] Abbreviations used in identifying surface coatings are: SO—silicone oil; SA—stearic acid. Contaminants were dissolved in chloroform, at the volumetric percentage indicated. Coating was performed by dipping.

[c] NS—no spreading even with pure ethanol.

TABLE II
Wettability, lap shear strength (LSS) and failure mode[a] of treated Al-2024

Surface Treatment[b]	θ°	CSC %	LSS, MPa FM73 + BR127 (Failure mode)	LSS, MPa FM300K + BR127 (Failure mode)
Plasma Cleaning	41	20–25	3.06 ± 0.10 (20%A)	2.27 ± 0.13 (30%A)
SO 10^{-5}%	76	35–40	2.97 ± 0.10 (20%A)	2.42 ± 0.05 (45%A)
SO 10^{-2}%	70	55–60	3.28 ± 0.12 (25%A)	2.59 ± 0.06 (50%A)
SO 1%	92	60–65	3.00 ± 0.51 (30%A)	2.44 ± 0.13 (50%A)
SA 10^{-5}%	72	40–45	3.17 ± 0.15 (25%A)	2.48 ± 0.02 (50%A)
SA 10^{-2}%	117	55–60	3.06 ± 0.09 (35%A)	2.06 ± 0.19 (80%A)
SA 10^{-1}%	155	NS[c]	2.88 ± 0.11 (70%A)	0.65 ± 0.11 (100%A)

[a] Failure mode in %A (percentage adhesive failure) is defined as the percentage of exposed substrate area which is devoid of adhesive.

[b] Abbreviations used in identifying surface coatings are: SO—silicone oil; SA—stearic acid. Contaminants were dissolved in chloroform, at the volumetric percentage indicated. Coating was performed by dipping.

[c] NS—no spreading even with pure ethanol.

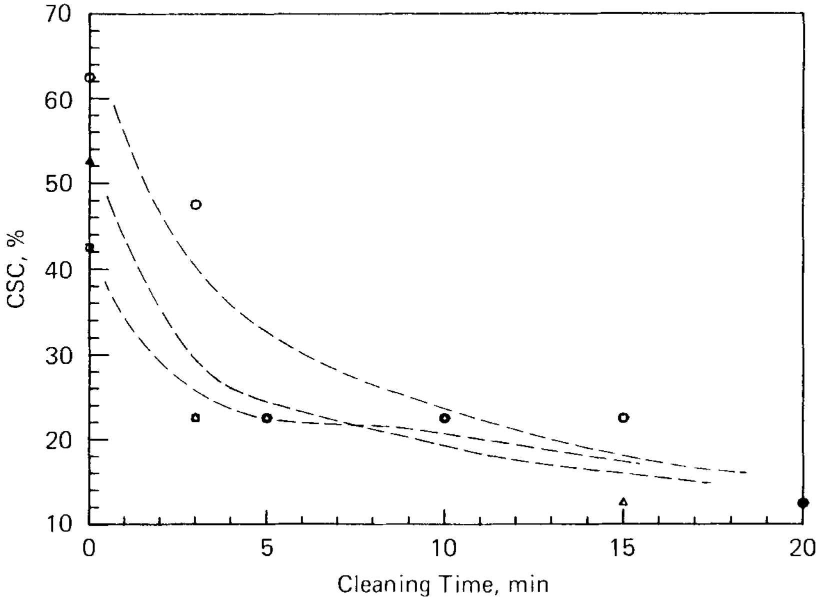


FIGURE 2 The effect of cleaning time in plasam on the CSC. Samples initially contaminated by silicone oil at various concentrations: ○—10^{-1}%, △—10^{-2}% and □—10^{-3}%.

method for systems of practical interest, where contact angle hysteresis may interfere with surface characterization due to roughness and chemical heterogeneity.

To further establish the usefulness of the CSC method and its sensitivity to surface treatments, the cleaning of Al-1100 substrates was followed. Figure 2 shows the CSC *vs.* the cleaning time in plasma, for various initial concentrations of silicone oil. The decrease in CSC with the progress of the cleaning process is clearly seen.

Having established the usefulness of the CSC method for the present systems, the main goal of this paper i.e., the elucidation of the correlation between the adhesive strength and the CSC, can be discussed. The results are summarized in Tables I–III and Figures 3 and 4. In order to present a complete picture, the advancing contact angles were also measured for the samples used for the Lap Shear Strength (LSS). Unfortunately, the T-peel samples were too large for the contact angle goniometer. The correlation of the adhesive

TABLE III
Wettability, peel strength (*T*-peel) and failure mode[a] of treated Al-2024

Surface coating[b]	CSC %	*T*-peel, N/mm FM73 + BR127 (Failure mode)
"As received"	40–45	8.12 ± 0.25 (0%A)
SO 10^{-5}%	45–50	8.49 ± 0.21 (0%A)
SO 10^{-2}%	75–80	10.82 ± 0.26 (0%A)
SO 1%	NS[c]	10.59 ± 0.21 (0%A)
SA 10^{-5}%	55–60	8.31 ± 0.14 (0%A)
SA 10^{-2}%	95–100	7.32 ± 0.30 (20%A)
SA 10^{-1}%	NS	2.99 ± 0.33 (95%A)

[a] Failure mode in %A (percentage adhesive failure) is defined as the percentage of exposed substrate area which is devoid of adhesive.

[b] Abbreviations used in identifying surface coatings are: SO—silicone oil; SA—stearic acid. Contaminants were dissolved in chloroform, at the volumetric percentage indicated. Coating was performed by dipping.

[c] NS—no spreading even with pure ethanol.

strength with the CSC is presented only in the tables and not in the figures, since some of the pronounced changes in the adhesive strength occurred at non-spreading situations, *i.e.* beyond a CSC of 100%.

The results show that the LSS for the FM-73 adhesive is only mildly sensitive to the contaminant concentration, for the two coatings which were studied. With the FM-300K adhesive, the LSS is mildly sensitive to the contaminant concentration for the samples coated with silicone oil, while a marked decrease in the LSS is seen for high concentrations of stearic acid. Similar trends, in terms of the dependence of the adhesive strength on the presence of contamination, were previously observed,[8] although for different adherends and with different specific dependence on the type of contamination. As Figures 3 and 4 show, a decrease in LSS is correlated with contact angles higher than 90°. From Tables I and II

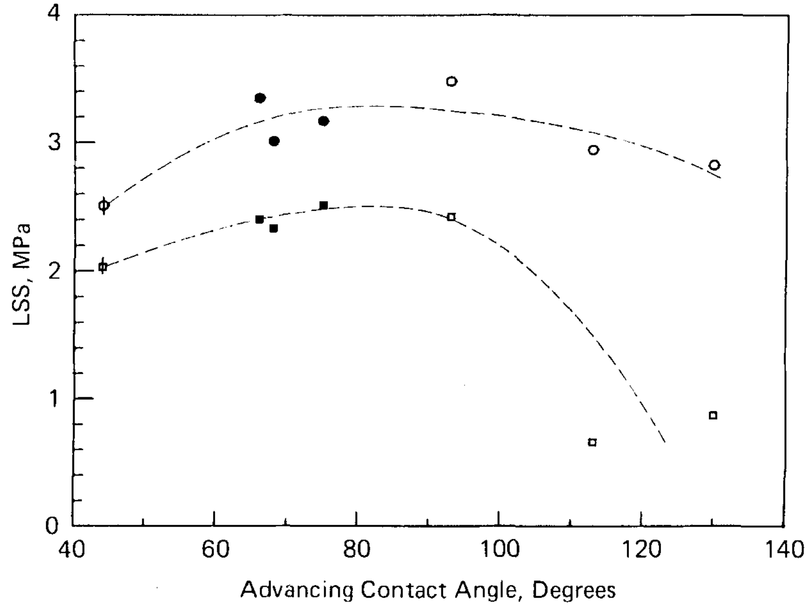


FIGURE 3 The dependence of LSS on the advancing contact angle for Al-1100. ○—FM73. □—FM300K. Full symbols—silicone oil. Empty symbols—stearic acid. Divided symbols—plasma cleaning.

it can also be concluded that the decrease in LSS is associated with high CSC values of over 70%, and mostly with non-spreading situations. These observations indicate that a decrease in the LSS is clearly associated with a diminished wetting of the solid by the adhesive. However, the low sensitivity of the LSS to the concentration of the contaminant in some cases needs to be explained. In general, a low sensitivity may stem either from the mechanism of adhesion or from the test method itself. For rough surfaces, the LSS may be increased by mechanical interlocking of the adhesive in the surface grooves, thus masking the decrease in strength, which should have resulted from lowering the wettability of the adherends by the contaminants. On the other hand, a low sensitivity of the LSS to the concentration of a contaminant may also be due to an interaction between the contaminant and either the primer or an adhesive component, which may compensate for the decreased

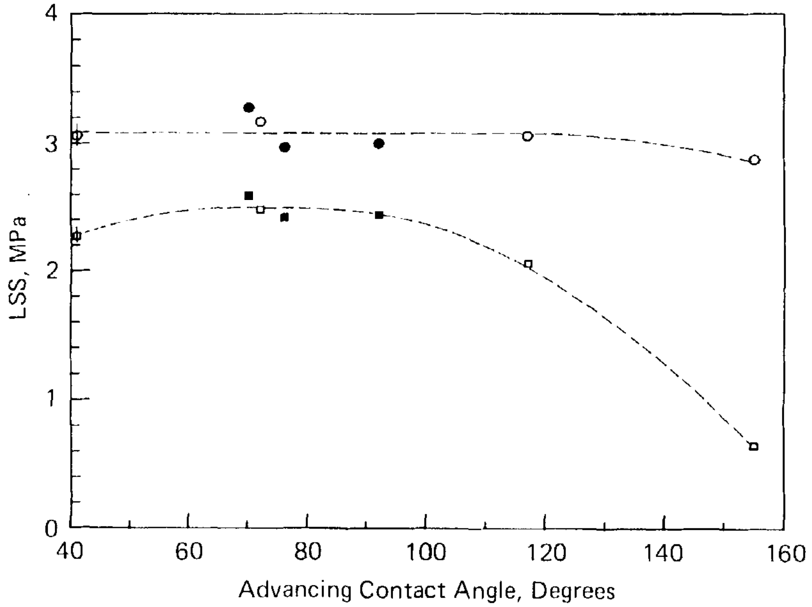


FIGURE 4 The dependence of LSS on the advancing contact angle for Al-2024. Symbols are as in Figure 3.

wettability. Clearly, some additional information is required in order to resolve this question, as discussed below.

It is also interesting to notice that the Al-1100 cleaned by plasma performed worse than the coated samples of this material. This may be attributed to the adsorption of contamination from the environment sometime during the handling of the samples. This explanation is supported by the CSC value of 35–40, which is much higher than usual for clean Al-1100, as shown above (Figure 2). These results demonstrate the possible effect of random contamination on the adhesive strength, and the usefulness of the CSC method in identifying its existence.

The failure mode is presented in Tables I and II in terms of the percentage of adhesive failure. These tables show that the percentage of adhesive failure increases with the concentration of stearic acid, but depends only mildly on the concentration of silicone oil. The failure mode is also shown to be more adhesive for coatings of stearic acid than for silicone oil. These observations suggest a clue to the question which was posed above. Since both the LSS and the failure mode for the samples coated with silicone oil depend only mildly on its concentration, it is reasonable to assume that some interaction exists between the silicone oil and either the primer or an adhesive component, which counteracts the effect of decreased wettability. In contrast, the effect of stearic acid seems to be directly related to the reduction in the surface energy (and thereby in the wettability) of the adherends, since the failure mode becomes more adhesive, even when the LSS is not much decreased.

Table III presents the results for the T-peel strength. Only the FM-73 adhesive was used, since the FM-300K adhesive is known to have a very low peel strength. Similarly to the LSS, the T-peel strength depends only mildly on the concentration of silicone oil. However, an appreciable decrease in strength is apparent for high concentrations of stearic acid. The T-peel strength is expected to be more sensitive to surface contamination than the LSS, since it is less influenced by mechanical interlocking than the latter. Thus, the hypothesis that the insensitivity of the adhesive strength to the presence of silicone oil is due to some interaction with either the primer or an adhesive component, is supported by the fact that the T-peel strength is also insensitive to this coating. In addition, since the T-peel strength is more sensitive to the concentration of stearic acid than the LSS, it can be concluded that the effect of stearic acid is indeed to decrease the surface energy of the solid and thereby its wettability by the adhesive. The dependence of the failure mode on the concentration of contamination, as determined by testing T-peel failure surfaces, is similar to the corresponding results for the lap shear tests: for the samples coated with stearic acid, the failure mode becomes more adhesive as the CSC increases.

SUMMARY AND CONCLUSIONS

1. The CSC method was proven to be sensitive to variations in the surface energy of the aluminum alloys studied. It was shown to be more reliable than contact angle measurements, while the performance of the test is much easier and simpler than the latter. In

addition, the CSC method is not limited by the size and geometry of the samples, in contrast with contact angle measurements, where these factors may determine the feasibility and convenience of the measurements.

2. The LSS as well as the T-peel strength are only mildly sensitive to the concentration of silicone oil in the systems studied. These observations suggest some interaction of the silicone oil with either the primer or an adhesive component, in a way which compensates for the decrease in surface energy and wettability of the adherends.

3. The LSS is markedly sensitive to the concentration of stearic acid for the FM-300K adhesive, and mildly sensitive to the concentration of stearic acid for FM-73. However, the peel strength for the latter shows appreciable sensitivity. Since the T-peel strength is more sensitive to the wetting of the adherend by the adhesive than the LSS, these observations indicate that the effect of stearic acid on the adhesive strength is related to the reduction in surface energy of the adherends.

4. The failure mode becomes more adhesive as the concentration of stearic acid and thereby the CSC increase. It is almost insensitive to the concentration of silicone oil.

Acknowledgement

The authors wish to express their sincere gratitude to N. Fin for technical assistance, and to L. Drori for helpful discussion.

References

1. J. R. Huntsberger, in *Contact Angle, Wettability and Adhesion,* Adv. in Chem. Ser., **43** (ACS, Washington, D.C., 1964), p. 180.
2. L. H. Sharpe and H. Schonhorn, *Ibid.*, p. 189.
3. J. J. Bikerman, *The Science of Adhesive Joints,* 2nd. ed. (Academic Press, N.Y., 1968).
4. R. E. Baier, E. G. Shafrin and W. A. Zisman , *Science* **162,** 1360 (1968).
5. Y. Kitazaki and T. Hata, *J. Adhesion* **4,** 123 (1972).
6. J. R. Huntsberger, *J. Adhesion* **7,** 289 (1976).
7. W. C. Wake, *Adhesion and the Formulation of Adhesives* (Applied Science Publishers Ltd, London, 1976).
8. T. Smith, in *Structural Adhesives and Bonding* (Structural Adhesives and Bonding Conference, El Segundo, California, March 1979), p. 190.
9. J. R. Huntsberger, *J. Adhesion* **12,** 3 (1981).
10. L. S. Penn and E. R. Bowler, *Surface and Interface Analysis* **3,** 161 (1981).

11. R. J. Good, in *Surface and Colloid Science,* Vol. 11, R. J. Good and R. R. Stromberg, Eds. (Plenum Press, N.Y., 1979), pp. 1–29.
12. W. A. Zisman, in *Contact Angle, Wettability and Adhesion,* Adv. Chem. Ser., **43** (ACS, Washington, D.C., 1964), p. 1.
13. A. Marmur, M. Narkis and W. Woogen, *J. Appl. Polym. Sci.* **25,** 1253 (1980).
14. A. Marmur, D. Pesach, H. Dodiuk and L. Drori, in *Fouling and Cleaning in Food Processing,* (Second International Conference on Fouling and Cleaning in Food Processing, Madison, Wisconsin, July 1985), p. 340.
15. A. Marmur, W. Chen and G. Zografi, *J. Coll. Interf. Sci.,* **113,** 114 (1986).
16. I. E. Klein, *et al., Int. J. Adhesion Adhesives* **3,** 159 (1983).
17. A. E. Yaniv, I. E. Klein, J. Sharon and H. Dodiuk, *Surf. Interf. Anal.* **5,** 93 (1983).
18. A. E. Yaniv, N. Fin, H. Dodiuk and I. E. Klein, *Appl. Surf. Sci.* **20,** 538 (1985).
19. H. Dodiuk, *et al., Appl. Surf. Sci.* **25,** 137 (1986).
20. N. Fin, H. Dodiuk, A. E. Yaniv and L. Drori, *Appl. Surf. Sci.,* in press.