Clinical adhesiveness of selected dental materials

An *in-vivo* study

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An investigation was conducted to determine the clinical adhesiveness of some commonly used dental materials. Contact angle measurement data, obtained under clinical conditions, were used to calculate the *in vivo*-critical surface tensions for enamel, gold, stainless steel, and a resin polymer. The results of this study showed that the adhesive properties of the artificial surfaces studied were considerably modified in the oral environment. Thus, after two hours these solids with diverse original surface chemistries were observed to produce *in vivo*-critical surface tensions common to those recorded for enamel. The observed changes were attributed to be adsorbed biofilm (acquired pellicle), which forms on all solid surfaces in the oral cavity.

Key-words: Pellicle; biofilm adhesiveness; clinical

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The surface chemistry of teeth and selected dental materials has been studied by several investigators; (2, 3, 4, 5, 7, 11, 13, 15, 18). These studies have similar findings; the tooth structures themselves are normally covered by low energy substances and the metallic dental restorative materials have hydrophilic surfaces of relatively high free energy levels. Further, it is generally recognized that an organic film, the acquired pellicle, forms on teeth in the oral cavity by adsorption of salivary glycoproteins (12). Baier & Glantz (2) have studied these oral films on various well defined artificial surfaces *in vivo.* Among other things they found that substrates of different surface free energy levels produced different molecular packing patterns and different film thicknesses. On the other hand Jendresen & Glantz (11) have shown that the clinical adhesiveness of normal tooth enamel surfaces was at a common level over a wide range of ages and in both sexes.

Now the question arises as to whether or not foreign material surfaces in the oral cavity, after the adsorption of organic film-pellicles, will exhibit similar or different adhesive behaviour from that of normal tooth surfaces. In other words, is there a possible difference in the adhesive bonding potential to micro-organisms, biological debris or dental plaque, between normal tooth

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surfaces and restorative and prosthetic materials? Bearing the above in mind it was considered worthy to study the clinical adhesiveness (8) of selected dental materials.

MATERIALS AND METHODS

Four males between the ages of 25 and 28 years were chosen as test persons. They considered themselves to be in good general health, and were found to be in good dental health upon clinical examination.

The three dental materials selected were pure gold foil, stainless steel (Stainless steel matrix material, type 3/16" x .002», Crescent Dental Manufacturing Company, Chicago, Illinois) and a dental polymer (Nitrocellulose Cavity Varnish, batch 4551–138, S. S. White Dental Health Products, King of Prussia, Pennsylvania).

The gold foil in sheet form was cut to size and placed over the entire labial surface of the test person's central incisor. Cavity varnish was placed on the tooth surface in order to lute the gold foil in place for the duration of the test. The gold foil was then burnished to contour with the use of a dental cotton roll.

The stainless steel material was placed in a similar fashion by burnishing the matrix band around the contours of a central incisor and retained by the mechanical retention afforded by the adjacent contacting tooth.

The resin polymer suspended in ether was painted on to the labial surface of a central incisor. Following the immediate evaporation of the ether vehicle, a thin continuous film of the polymer remained on the surface for testing.

The determinations of the *in vivo*-critical surface tension for the surfaces studied were performed two hours after their placement in the oral cavity. No food or drink was consumed during this time. The method used has been presented in detail by Glantz, Jendresen & Baier (8). Briefly, this method employed contact angle measurements on the solid surfaces in question. Pure liquids of known surface tensions, Table 1, were delivered to the surface in microdrops via a clean platinum wire. After the drop had reached a mechanical equilibrium, the angle of contact between the liquid and solid was measured. From the contact angles (θ) obtained, the critical surface tensions according to Zisman (19) were calculated using the equations and graphic modifications described by Glanz et al (8).

Three individual experiments were performed for each test person and material. Prior to their placement into the oral environment the critical surface tensions were also determined for the individual restorative materials. From the metal surfaces the contact angles were obtained immediately after cleaning with ethanol and subjection to an alcohol flame, which allowed the surfaces to attain a red heat. The polymer was evaluated immediately after having been applied to a clean glass slab. Studies on some of the possible sources of error in this method have previously been reported by Glanz et al (8).

RESULTS

The means of the measured contact angles obtained from the three selected dental materials prior to and after their placement into the oral environment are presented in Table 1. The spontaneous spreading of both the polar and non-polar liquids on the metal surfaces, found at the preclinical experiments, demonstrate resultant high energy surfaces. The resultant critical surface tensions of the selected resin (37.2 dynes/cm) was considerably lower than that of the cleaned metal surfaces. The calculated mean critical surface tensions for both polar (γ_c^p) and non-polar (γ_c^d) test liquids for the surface of the materials after two hours in the oral cavity are presented in Table 2.

An analysis of variance from these data, using the polar and non-polar liquids, showed that there was no significant difference in critical surface tension of the films formed on the different materials within the same test person. However, statistically significant differences ($p < 0.01$) were observed between the four test persons using the non-polar liquids. The data were analysed by the computer program entitled: Statistical Package for the Social Sciences (SPSS), Sperry Univac 1100, exec. 8, Version H, September 1978.

The results indicate that the studied materials, when placed in the oral environment and thus covered by a biofilm (acquired pellicle), exhibit the same clinical adhesiveness in spite of the considerable differences between their basic surface chemical properties.

DISCUSSION

All dental restorative and prosthetic materials are solids and may be divided into two broad categories relative to their specific surface free energies. Hard solids have surface free energies ranging from 500 ergs/cm^2 to several thousand ergs/cm^2. The high values relate to materials with a high melting point and high hardness values. Conversely, soft solids generally have specific surface free energies below 100 ergs/cm^2. Most liquids have similar low surface free energies and will normally spread freely on solids of high but not on solids of low surface energies. Materials in these two classes may be referred to as possessing «high-energy surfaces», and «low-energy surfaces» (1).

A liquid will generally spread on any high energy solid unless a film, with a surface tension lower than that of the liquid, has formed on the surface. Since an adsorbed film with a thickness of a couple of nm or more will impart its surface chemical properties to the underlying solid, solids of different specific surface free energies will exhibit the same critical surface tension, when covered by a thick enough common film.

Saliva is in contact with all exposed solid surfaces within the oral cavity. This complex liquid is composed of both organic and inorganic substances and has a surface tension of about 53 dynes/cm (6, 9, 15), its mean dispersion force component being about 29 dynes/cm at the time of salivary stimulation (6). These reported surface tensions indicate that saliva will, to some degree, wet all solid surfaces normally present in the mouth. Thereby it is given an opportunity to form salivary films. Such films or acquired pellicles have been found to form on solid surfaces immediately after their exposure to saliva *in vivo* (2) and are considered to be derived mostly from salivary glycoproteins (16, 17).

The results from the *in vitro* parts of this study indicate that clean, smooth, hard surfaces of gold and stainless steel have high energy surfaces. These results agree with those previously reported by Glantz (5, 6).

Further, the inherent critical surface tension for the tested dental polymer was found to be 37.2 dynes/cm. This is very close to the values reported by Jarvis, Fox & Zisman (10) and Glantz (6) of 39 and 41.2 dynes/cm, respectively, for polymethyl methacrylate.

When these different materials with widely different critical surface ten-

Table 1. *Mean contact angles (Θ) recorded in vitro and in vivo for four test persons (I-IV) between certain test liquids (γ_{LV}) and enamel/selected dental materials*

Test liquid: Water (γ_{LV} = 72.4 dynes/cm)

Material	Test person I			Test person II			Test person III			Test person IV			In vitro
	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	Θ
Enamel	5	26.4°	1.98°	4	27.8°	2.68°	4	20.5°	13.68°	5	26.8°	6.00°	–
Gold	3	28.9°	0.53°	3	24.3°	3.18°	3	31.6°	1.24°	3	27.4°	7.60°	Spreading
Dental Polymer	3	29.0°	2.38°	3	27.1°	1.24°	3	29.1°	2.30°	3	31.4°	5.13°	62.2°
Stainless Steel	3	30.4°	2.30°	3	14.3°	20.15°	3	29.5°	4.95°	3	14.1°	19.98°	Spreading

Test liquid: Glycerol (γ_{LV} = 63.7 dynes/cm)

Material	Test person I			Test person II			Test person III			Test person IV			In vitro
	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	Θ
Enamel	4	44.8°	2.77°	4	41.5°	10.19°	4	49.3°	3.38°	5	43.0°	4.09°	–
Gold	3	40.3°	13.08°	3	42.3°	3.18°	3	57.6°	1.94°	3	52.0°	0.71°	Spreading
Dental Polymer	3	44.6°	2.67°	3	37.4°	7.25°	3	55.5°	7.07°	3	53.3°	3.89°	52.2°
Stainless Steel	3	39.1°	16.44°	3	47.9°	6.19°	3	45.8°	8.48°	3	40.4°	8.30°	Spreading

Test liquid: Formamide (γ_{LV} = 58.5 dynes/cm)

Material	Test person I			Test person II			Test person III			Test person IV			In vitro
	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	Θ
Enamel	4	26.8°	3.31°	4	21.6°	14.53°	4	30.8°	4.15°	5	32.9°	1.90°	–
Gold	3	31.4°	0.88°	3	31.3°	8.13°	3	26.6°	9.01°	3	30.6°	0.88°	Spreading
Dental Polymer	3	27.9°	3.15°	3	33.0°	7.78°	3	16.4°	23.16°	3	35.6°	6.19°	40.0°
Stainless Steel	3	28.4°	0.53°	3	17.3°	24.40°	3	17.6°	24.93°	3	19.5°	10.60°	Spreading

Test liquid: Thiodiglycol (γ_{LV} = 53.5 dynes/cm)

Material	Test person I			Test person II			Test person III			Test person IV			In vitro
	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	Θ
Enamel	4	27.9°	1.16°	4	31.0°	2.21°	4	34.0°	2.28°	5	30.4°	1.24°	–
Gold	3	31.1°	7.96°	3	34.9°	0.53°	3	36.4°	1.23°	3	31.6°	0.88°	Spreading
Dental Polymer	3	30.2°	7.07°	3	28.5°	1.41°	3	35.6°	1.23°	3	30.9°	4.77°	38.0°
Stainless Steel	3	29.1°	4.78°	3	25.0°	4.24°	3	32.3°	6.36°	3	27.5°	4.60°	Spreading

Test liquid: Methylene iodide (γ_{LV} = 51.7 dynes/cm)

Material	Test person I			Test person II			Test person III			Test person IV			In vitro
	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	Θ
Enamel	4	44.4°	2.95°	4	45.0°	1.08°	4	58.3°	3.56°	5	46.6°	6.16°	–
Gold	3	48.5°	3.89°	3	53.4°	2.65°	3	51.3°	5.30°	3	52.4°	3.36°	Spreading
Dental Polymer	3	40.6°	1.04°	3	47.0°	3.18°	3	51.5°	2.82°	3	52.1°	6.19°	36.2°
Stainless Steel	3	49.0°	6.01°	3	58.1°	0.17°	3	58.4°	12.90°	3	56.5°	8.13°	Spreading

Test liquid: S-tetrabromoethane (γ_{LV} = 49.8 dynes/cm)

Material	Test person I			Test person II			Test person III			Test person IV			In vitro
	n	Θ	S.D	n	Θ	S.D.	n	Θ	S.D	n	Θ	S.D.	Θ
Enamel	4	35.5°	5.18°	4	35.9°	4.94°	4	48.0°	2.15°	5	33.8°	12.08°	–
Gold	3	36.1°	6.89°	3	40.6°	5.13°	3	47.9°	10.78°	3	46.8°	12.37°	Spreading
Dental Polymer	3	34.0°	4.79°	3	37.1°	15.02°	3	36.9°	5.48°	3	36.3°	5.66°	25.0°
Stainless Steel	3	39.6°	0.88°	3	41.5°	3.18°	3	45.6°	11.14°	3	52.3°	5.66°	Spreading

Test liquid: Propylene carbonate (γ_{LV} = 41.8 dynes/cm)

Material	Test person I			Test person II			Test person III			Test person IV			In vitro
	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	Θ
Enamel	4	31.4°	5.83°	4	29.5°	2.26°	4	34.3°	2.16°	5	32.1°	1.46°	–
Gold	3	31.0°	8.13°	3	36.5°	3.18°	3	37.8°	6.18°	3	38.5°	5.65°	Spreading
Dental Polymer	3	29.8°	3.86°	3	30.0°	6.36°	3	35.8°	1.77°	3	35.8°	6.36°	20.8°
Stainless Steel	3	36.6°	0.53°	3	32.13°	11.84°	3	33.8°	7.42°	3	35.9°	9.37°	Spreading

Test liquid: Dicyclohexyl (γ_{LV} = 32.7 dynes/cm)

Material	Test person I			Test person II			Test person III			Test person IV			In vitro
	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	Θ
Enamel	4	Spreading		4	3.4°	6.88°	4	12.2°	8.22°	5	Spreading		–
Gold	3	Spreading		3	7.5°	10.61°	3	17.3°	4.24°	3	Spreading		Spreading
Dental Polymer	3	Spreading		3	Spreading		3	14.0°	4.60°	3	Spreading		Spreading
Stainless Steel	3	5.5°	7.79°	3	7.6°	10.78°	3	15.9°	1.94°	3	Spreading		Spreading

Test liquid: Hexadecane (γ_{LV} = 27.6 dynes/cm)

Material	Test person I			Test person II			Test person III			Test person IV			In vitro
	n	Θ	S.D	n	Θ	S.D.	n	Θ	S.D.	n	Θ	S.D.	Θ
Enamel	–			4	Spreading		4	Spreading		–			–
Gold	–			–	Spreading		3	Spreading		–			–
Dental Polymer	–			–	Spreading		3	Spreading		–			–
Stainless Steel	3	Spreading		3	Spreading		3	Spreading		–			–

Table 2. *Calculated mean in-vivo-critical surface tensions (γ_c^p & γ_c^d) for human enamel and selected dental materials*

Material	n	γ_c^p (dynes/cm)		γ_c^d (dynes/cm)	
		mean	S.D.	mean	S.D.
Human enamel	17	46.8°	15.00°	31.9°	1.68°
Gold	12	39.5°	8.99°	31.4°	1.66°
Dental Polymer	12	39.9°	15.68°	31.6°	1.67°
Stainless Steel	12	37.3°	23.51°	31.9°	1.65°

sions were placed in the mouths of human subjects, their surface properties were modified in such a manner as to be indistinguishable from the surface chemistry of normal tooth structure.

These results lead to the conclusion that all these surfaces of extremely different original chemistry are being brought to the same surface state by a mechanism of surface film adsorption.

Since the surface film characteristics, measured by contact angles, in the relevant clinical setting are the same as those obtained on natural dentition, the surface properties that were observed must be imparted by the adsorbed film.

The data then endorse and enhance the findings from the work on human dental enamel surfaces, including the high but somewhat varying precisions, of the methods used (8, 11). The results also eliminate any speculation that the observed low energy surface properties might have been contributed by some diffusion peculiarity from within the tooth structure, rather than by some component in the saliva.

The original surface properties of the materials tested: enamel, gold, stainless steel, and a resin polymer, could not in themselves have imparted a common low energy property to their surfaces. Therefore, one must conclude that the nature of the film formed brought these different materials to the same free surface energy state, and thus to the same surface adhesiveness.

These results lead to several practical conclusions about adhesive events in the oral cavity with respect to the fouling potential of plaque and debris. Different materials are neither more nor less beneficial than the natural dentition in preventing or enhancing biological adhesion as a precursor for periodontal disease and caries.

A microorganism coming close to a tooth surface will therefore find no differences in adhesive properties depending on whether the surface is natural or restored.

In this context it must, however, be strongly emphasized that even if the adhesiveness is found to be exactly the same for all the films on all the tested materials, there can be considerable and important differences between other surface chemical qualities of these films. Their packing distributions as well as the work of adhesion between the films and their respective underlying solids can be different, as has been shown by Baier & Glantz (2) and Glantz (6, 7). These factors will influence the possibilities for desorption of the film material as well as the maximum amount of adhering material, such as plaque (6).

Due to the fact that none of the presently existing materials with bioabhesive properties (1) have mechanical characteristics enabling them to be tested under the experimental conditions of this study, this complex problem could not be further explored.

Further, as has been shown by Sönju

& Glantz (17) and Norman, Mehra, Swartz & Phillips (14) there could be differences in the composition of the inner parts of the films. Such differences might influence on the metabolism of microorganisms retained to the film.

Finally, the surface roughness of a material may be transmitted to the film surface. A resulting surface texture difference may contribute to or, in some instances, modify the adhesive properties of the adsorbed film.

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REFERENCES

1. Baier, R. E. Surface properties influencing biological adhesion. In: Manly, R. S., ed. Adhesion in Biological Systems, Academic Press, N. Y., 1970, pp. 15 - 48
2. Baier, R. E. & Glantz, P.-O. Characterization of oral in vivo films formed on different types of solid surfaces. Acta Odontol. Scand. 1978, 36, 289 - 301
3. Brauer, G. M. & Wiedeman, W. H. Measurement of the surface area and heats of wetting of dentine powders. J. Dent. Res. 1964, 43, 921
4. Glantz, P.-O. & Nyquist, G. The effect of topical fluorine application on the free surface energy of dentine. Odontol. Revy 1966, 17, 332 - 335
5. Glantz, P.-O. On wettability and adhesiveness. Odontol. Revy 1969, 20, Suppl. 17, 1 - 132
6. Glantz, P.-O. The surface tension of saliva. Odontol. Revy 1970, 21, 119 - 128
7. Glantz, P.-O. The adhesiveness of teeth. J. Coll. Interf. Sci. 1971, 37, 281 - 290
8. Glantz, P.-O., Jendresen, M. D. & Baier, R. E. A clinical method for the study of in vivo adhesiveness of teeth. Acta Odontol. Scand., 1980, 38, 371 - 378
9. Hodgman, C. D. ed. Handbook of chemistry and physics, Ed. 44 Chemical Rubber Publishing Company, Cleveland, 1962
10. Jarvis, N. L., Fox, R. B. & Zisman, W. A. Surface activity at organic-liquid air interfaces. V. Effect at partial fluorinated additives on the wettability of solid polymers. In: Gould, R. F. ed. Contact angle, wettability and adhesion. Advances in Chemistry Series No. 43, American Chemical Society, Washington D.C. 1964
11. Jendresen, M. D. & Glantz, P.-O. Clinical adhesiveness of the tooth surface. Acta Odontol. Scand., 1980, 38, 379 - 383
12. Meckel, A. H. The formation and properties of organic films on teeth. Arch. Oral Biol. 1965, 10, 585 - 593
13. Newman, G. V. & Sharpe, L. H. On the wettability of tooth surfaces. J. N. J. Dent. Soc. 1966, 37, 289 - 306
14. Norman, R. D., Mehra, R. V., Swartz, M. L. & Phillips, R. W. Effects of restorative materials on plaque composition. J. Dent. Res. 1972, 51, 1596 - 1601
15. O'Brien, W. J. & Ryge, G. Wettability of poly-(methyl methacrylate) treated with silicon tetrachloride. J. Prosthet Dent. 1965, 15, 304 - 308
16. Rölla, G. Formation of dental integuments - some basic chemical considerations. Swed. Dent. J. 1977, 1, 241 - 251
17. Sönju, T. & Glantz, P.-O. Chemical composition of salivary integuments formed in vivo on solids with some established surface characteristics. Arch. Oral Biol. 1975, 20, 687 - 691
18. Uy, K. C. & Chang, R. Approaches to achieve adhesion. In: Public Health Publication No. 1494, U. S. Dept. Health, Education and Welfare, Washington D.C
19. Zisman, W. A. Relation of the equilibrium contact angle to liquid and solid constitution. In: Gould, R. F. ed. Contact angle, wetting and adhesion. Advances in Chemistry Series No. 43, American Chemical Society, Washington D. C. 1964, pp. 1 - 51