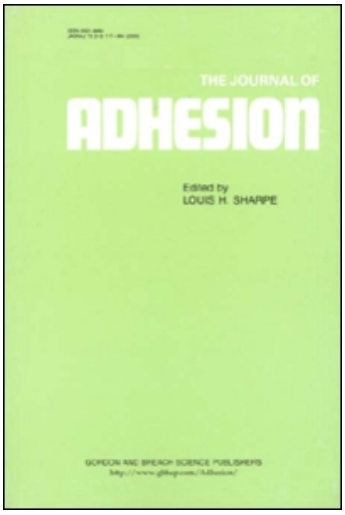

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Controlling Adhesion of Orthodontic Adhesives Through Adjustment of the Interphase Mechanical Properties

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Controlling Adhesion of Orthodontic Adhesives Through Adjustment of the Interphase Mechanical Properties*

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Adhesives for ceramic orthodontic brackets are so strong that instances of enamel fracture and bracket fracture have occurred during removal. Our approach to minimize the potential enamel damage was to modify the mechanical properties of the adhesive, a BIS GMA-silica composite, by use of diethyl phthalate which is a common plasticizer. The plasticizer, used in amounts up to 20% of the adhesive weight, significantly decreases the adhesive modulus and tensile strength. One objective of this research is to evaluate plasticizer stability in the adhesive *via* functional testing in a simulated oral environment. A second objective was to simulate, by use of finite element analysis, clinical loading conditions during orthodontic treatment and removal. The finite element analysis determined the changes in computed stresses due to plasticization. After 25 days in an artificial saliva solution held at 60°C, the bracket removal torque was lower for the 10% plasticized adhesive group than that for the non-plasticized group. The 3-D linear elastic finite element anlaysis found that plasticization should not lead to premature failure when typical treatment loadings were applied. The torsional loading conditions simulating bracket removal reported peak stresses in excess of the plasticized adhesive tensile strength in the corner regions. Thus, modelling of the adhesive as a layer with distinct mechanical properties appears reasonable.

KEY WORDS dental materials; BIS GMA; diethyl phthalate (DEP); modulus; torsional adhesion; plasticizer; orthodontics; ceramic brackets; bonding; adhesives.

INTRODUCTION

Single crystal and polycrystalline ceramic orthodontic brackets have improved the aesthetics during treatment.[1] While ceramic brackets have not completely replaced

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stainless steel brackets, they are becoming widely used. When ceramic brackets are bonded to brittle tooth enamel using heavily ceramic-filled adhesives, the joint is extremely strong and brittle which is desirable during treatment but leads to problems at the end of the treatment period. Among the biggest problems has been the difficulty in removing ceramic brackets after the treatment period is complete.[1-3] Clinically measured increases in the removal force have been documented over that required to remove stainless steel brackets.[2] As a result, clinical problems have included both bracket and enamel fractures.

There have been a series of published efforts recently to make bracket removal easier and more predictable. They have included heating the resin,[4-7] removing the silica reinforcement[8] and introducing modifiers such as plasticizers.[9,10] All of these have shown varying degrees of success as well as their own unique drawbacks.

For example, heating the bis glycidyl methacrylate [BIS GMA with a glass transition temperature above 100°C] resin will definitely reduce the stiffness. Unfortunately, the resin retains mechanical rigidity well above 60°C, considered to be an upper limit before tooth pulp damage occurs. Removal of ceramic filler will also reduce the modulus of the adhesive but, clinically, orthodontists find that without the filler, the resin is more difficult to use. Our efforts to reduce the stiffness of the adhesive have been to treat the adhesive as a separate layer with its own distinct mechanical properties between the adherends and to reduce its stiffness by adding plasticizers. Significant reductions in elastic modulus and torsional adhesion have been shown.[9,10] However, questions remain as to whether leaching of plasticizer by saliva will reduce the benefit of the plasticizer additions and how much plasticizer can be added to improve the removal characteristics without compromising the adhesive strength of the bonded bracket during treatment.

In previous work,[9,10] we have demonstrated statistically-significant reductions in the elastic modulus of a 75% silica-filled BIS-GMA composite [from 12.0 GPa without any plasticizer to below 3.0 GPa by adding up to 20% of the adhesive weight of diethyl phthalate (DEP), as measured at 10 Hz by dynamic mechanical spectroscopy]. In addition, when brackets were bonded to a standard stainless steel wire mesh substrate [made by Rocky Mountain Orthodontics, (RMO) Denver, CO], statistically-significant reductions in the torsional strength were observed due to the plasticizer additions. Maximum torsion values ranged from 0.20 N-m (1.76 in-lbs) for the non-plasticized adhesive to 0.07 N-m (0.66 in-lbs) when additions of up to 20% DEP by weight were made.

Conceptually, the mechanical properties of a particulate-reinforced composite can be described by the rule of mixtures as, for example, for the elastic modulus,[11]

$$E^n_{composite} = (X)\, E^n_{polymer} + (1 - X)\, E^n_{reinforcement}$$

where X is the weight fraction of polymer, $E_{polymer}$ and $E_{reinforcement}$ are the elastic moduli of the polymer and reinforcement materials, and n is an exponent ranging from -1 to 1 between the isostrain and the isostress cases, respectively. The exponent, n, is usually near 0 for particulate composites.[11] Plasticizers can reduce the elastic modulus of the polymer, $E_{polymer}$. Therefore, if the polymer in a reinforced composite adhesive resin can be plasticized, the reductions in the adhesive modulus, $E_{composite}$, will be due to polymer-plasticizer interactions. $E_{polymer}$ is temperature

dependent due to the free volume dependence on temperature. The plasticizer should not interact with the ceramic reinforcement.

This paper summarizes our efforts to modify the mechanical properties of the bulk adhesive and to alter the resulting bond strength. In particular, the question of how much the adhesive stiffness can be reduced while retaining viable adhesive properties during orthodontic treatment and allowing for easier removal is considered. The finite element (FE) model treats the adhesive zone as a separate layer with its own distinct properties. The results of salivary exposure studies are presented to determine the effectiveness of plasticization during exposure to an artificial saliva solution at two temperatures.

EXPERIMENTAL

Methods and Materials

The adhesive samples were based on a commercial orthodontic resin system (Reliance Orthodontics Phase II, Itasca, IL). This is a two-part, silica-filled acrylic resin using a peroxide curing agent and a tertiary amine activator. The filler content is nominally 75% of the adhesive by weight. Samples for mechanical property testing were fabricated by mixing the prescribed amount of plasticizer with equal parts of the two-part adhesive system. The mixture was then deposited into casting plates to make cured samples of the modified adhesive resin for dynamic mechanical property and tensile strength testing. Modulus measurements were made at various temperatures but reported at 30°C using a Polymer Labs DMTA equipped with a shear head in a single cantilever mode at 10 Hz while applying the appropriate end correction factor. The modulus measurements were taken as baseline values for the finite element modelling and analysis. The tensile strength measurements were made using an Instron TT Machine set at 0.127 cm/minute and were used for comparison with the stresses computed in the finite element model. At least 4 samples were used for the tensile strength measurements and at least 3 samples were used for the modulus measurements at each condition. Standard deviations were computed based on a student t statistical analysis. Adhesion tests were performed using the procedure below.

Groups of at least 5 orthodontic brackets for each plasticizer condition (RMO Denver, "Signature" Brackets) were bonded to a standard stainless steel wire mesh bonding disk made by RMO and used generally throughout the industry. The disk was prepared by first applying a sealant layer of unfilled resin to the disk. The thin sealant layer was a two-part acrylic resin with no filler. After allowing the sealant layer to set (5–10 minutes), the 2-part, silica-filled adhesive and desired amounts of DEP were mixed together by hand and a small amount was applied to each bracket. The bracket was then affixed to the bonding substrate. Again, all of the adhesives were modifications of the original commercially-available orthodontic resin. Care was taken to remove excess adhesive from around the edges of the bracket, as the adhesive set, without dislodging the bracket. This reasonably approximates what is performed by the clinician.

After the brackets were fixtured to each disk, salivary exposure studies were performed using an artificial saliva solution to determine how bond strength is affected by the interaction with saliva. The saliva solution was based on a composition originally outlined by Marek *et al.*[12] and exposures were performed at both room temperature and at 60°C. After periodic exposure times, sample disks were removed, gross water removed, and the bond strength tested in torque using a Sturtevant 0.569 N-m (5 in-lb) measurement device. Measurements of the maximum torque required for removal were recorded. Again, standard deviations were computed based on a student t distribution. Ideally, measurements of the plasticizer extracted by the saliva solution would have been made; however, there was such a small amount of plasticizer used for the bond tests that extraction was not a viable option. Only an indirect approach such as measuring bond strength before and after exposure was thought to be a reasonable approach.

Finite Element Modelling

A 3-D linear elastic finite element model was constructed for the orthodontic bracket and adhesive. The bracket was modelled to provide a mechanism to introduce load into the adhesive. Because the stiffness of the sapphire bracket is large compared with that of the adhesive, the adhesive will encounter the most deformation. The curvature and the microporous topography of the tooth were not modelled. Although the bracket and adhesive layer have geometric symmetry, a full model of the bracket and adhesive were assembled in anticipation of introducing non-symmetric tooth curvature in future studies. The interface between the tooth and the adhesive was represented as being rigid. The mechanical properties introduced into the model were generated by the measurements done in the first part of this study.

A boundary representation of the finite element model and the loading conditions for the two cases are shown in Figure 1. The adhesive pad is 2.65 mm wide and 3.54 mm long with a uniform adhesive thickness of 0.25 mm. The lower section of the model is the adhesive and the upper section is the bracket. All materials were modelled with 20-node brick elements which are not shown. The bracket has 1200 elements total with 2 layers through the thickness. All loads were applied to the bracket section.

The stresses were computed using the finite element program ABAQUS.[13] Two distinct load cases were examined. The top illustration in Figure 1 shows the shear loading of 3.92 N applied as a uniform force across the surface of the bracket. The shear load is applied in the + Y direction and it simulates the insertion and loading of the nickel/titanium wire in the bracket groove. The shear force is indicative of forces typically used in orthodontic treatment. The second loading condition shown in the lower illustration of Figure 1 represents a 0.169 N-m torque acting about the global Z-axis. The torque value is typical of bracket removal torques.[9,14] The von Mises stress in the adhesive was determined for both loading conditions in the unplasticized and 10% plasticized condition.

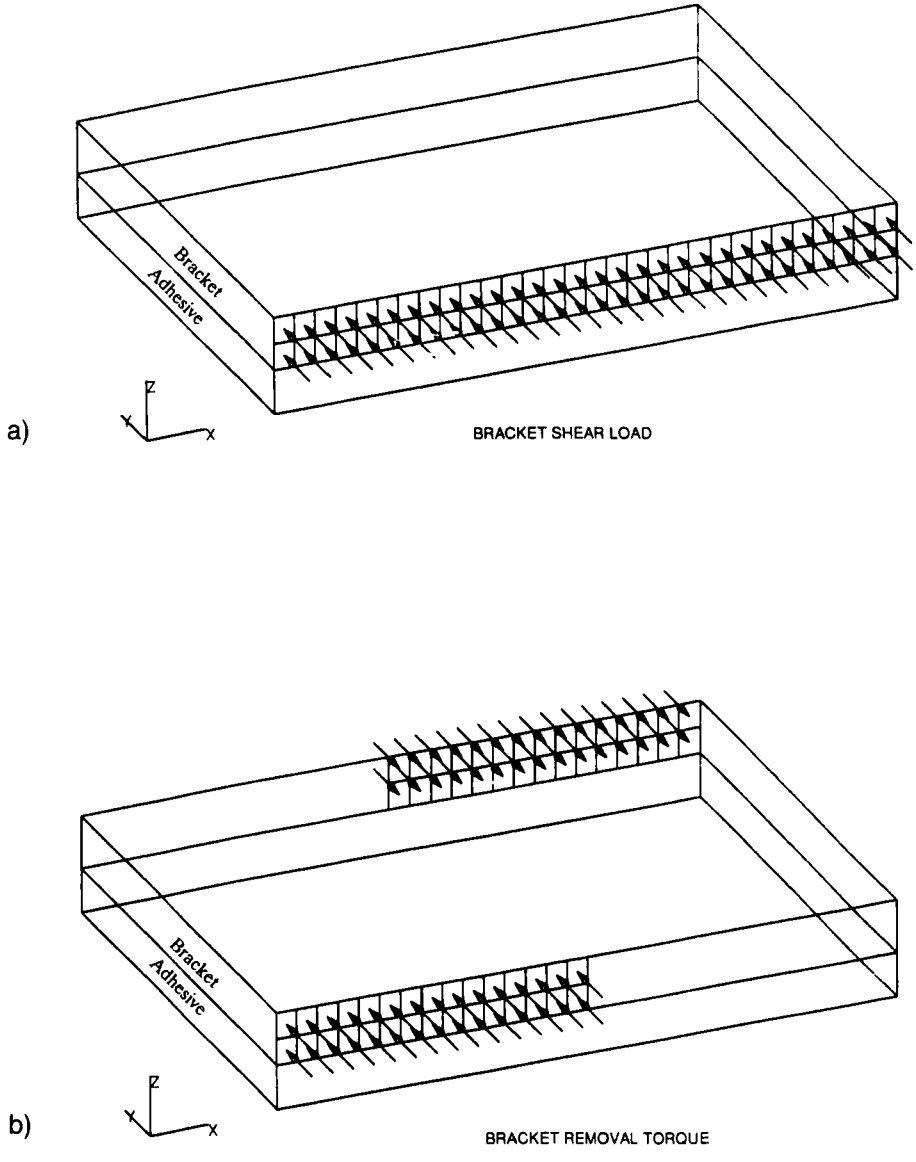

FIGURE 1 FEM model of the bracket and adhesive region including simulated loading conditions under shear "treatment" (a) and under torsional "removal" (b).

TABLE I
Mechanical property measurements *vs* plasticizer content

Sample (% DEP)	In-phase dynamic modulus, E' (GPa)	1 SD (GPa)	Tensile strength N/mm^2 (KSI)
0*	12.9	0.2	42.9 (6.2)
5	7.6	0.1	
10*	7.1	0.5	18.6 (2.7)
15	3.6	0.7	
20	3.0	1.9	

*Conditions inputted into the ABAQUS FEM Program

RESULTS AND DISCUSSION

Mechanical Property Measurements

A summary of the in-phase dynamic mechanical modulus measurements, E', and the tensile strengths is shown in Table I. The mechanical properties of the non-plasticized and 10% plasticized adhesive resins were used in comparing the finite element model. Significant reductions in both modulus and tensile strength occur with increasing plasticization. There should be a significant swelling effect due to polymer/solvent (plasticizer) interaction since DEP has nearly the same solubility parameter as methyl methacrylate, the main monomer segment. The plasticized polymer should also have decreased resistance to segmental motion and hence a lower modulus. One important result of this work is that the mechanical properties of the composite resin can be affected through plasticizer interactions with one of its constituents.

The modulus values from the dynamic mechanical measurements were used in the modelling work at two conditions, 0% plasticizer and 10% diethyl phthalate by weight. One major concern is how long the decreased mechanical properties are retained. This aspect is addressed by the exposure studies in the artificial saliva solution.

Salivary Exposure Results

The salivary exposure results at room temperature are shown in Table II and at elevated temperature in Table III. All bond failures occurred either at the bracket/adhesive interface or within the adhesive. At room temperature, the torsional removal force using the non-plasticized adhesive samples remains statistically higher than for the 10% plasticized sample group at 0 and at 30 days exposure. While the general trend for all conditions is toward lower torsional strength at increased plasticizer content, the results for the 5% plasticized adhesive group were not considered to be statistically lower than for the non-plasticized group. There is no statistically-significant increase over time in the torsional strength either for the 5% or for the 10% plasticized groups. An increase would have been indicative of salivary leaching of plasticizer.

At 60°C, more salivary leaching of the plasticizer from the adhesive would be expected. The torsional force results for the 5% and 10% plasticized samples indicate no statistically-significant change over time in the artificial saliva solution. The

TABLE II
Torsional debond strength as a function of time in the synthetic saliva solution (room temperature)

Torsional debond strength (N = m ± 1 S.D.)

Days (at room temp.)	% Plasticizer		
	0	5	10
0	0.20 ± 0.02	0.15 ± 0.05	0.10 ± 0.02
15	0.16 ± 0.05	0.15 ± 0.08	0.11 ± 0.05
30	0.21 ± 0.04	0.17 ± 0.06	0.09 ± 0.01

TABLE III
Torsional debond strength as a function of time in the synthetic saliva solution (60°C)

Torsional debond strength (N = m ± 1 S.D.)

Days (at 60°C)	% Plasticizer		
	0	5	10
0	0.20 ± 0.02	0.15 ± 0.05	0.10 ± 0.02
15	0.13 ± 0.03	0.13 ± 0.03	0.09 ± 0.01
20		0.16 ± 0.03	0.08 ± 0.02
25		0.13 ± 0.05	0.08 ± 0.01

torsional force results for the plasticized adhesives are not statistically different from their corresponding room temperature exposure values in the artificial saliva solution. There is an interaction between the non-plasticized adhesive and the saliva as measured by a reduction in the torsional force with time in the artificial saliva solution. This result is consistent with work by Beatty *et al.* who found moisture absorption in unfilled resin systems which reduced the overall hardness.[15] We were surprised not to see a similar effect with the plasticized adhesives.

There are several potential mechanisms for the lower bond strength at long times in the artificial saliva solution. The first is that the plasticizer interacts with the polymer over time to increase free volume, decrease the elastic modulus, and reduce the resultant bond strength. This is the case before the samples are exposed to the artificial saliva solution.[9] After salivary exposure, three mechanisms of reduced bond strength are possible. If there is no interaction between saliva and the plasticized adhesive, a continued reduction in bond strength is expected. Of course this indirect approach does not rule out the possibility of an exchange of saliva for plasticizer given that they may both have some affinity for the resin. There is also a chance that plasticizer extraction by saliva would create voids where extraction occurred in the vicinity of the interface, lowering the total bond area and resulting in continued lower bond strength even though some plasticizer is extracted.

Nevertheless, lower bond strength from plasticization is more of a permanent effect even after exposure to an artificial saliva solution. Over the time of our experimentation, there was no change in bond strength of the plasticized adhesive specimens that could be attributed to leached plasticizer as a result of exposure to the artificial saliva solution.

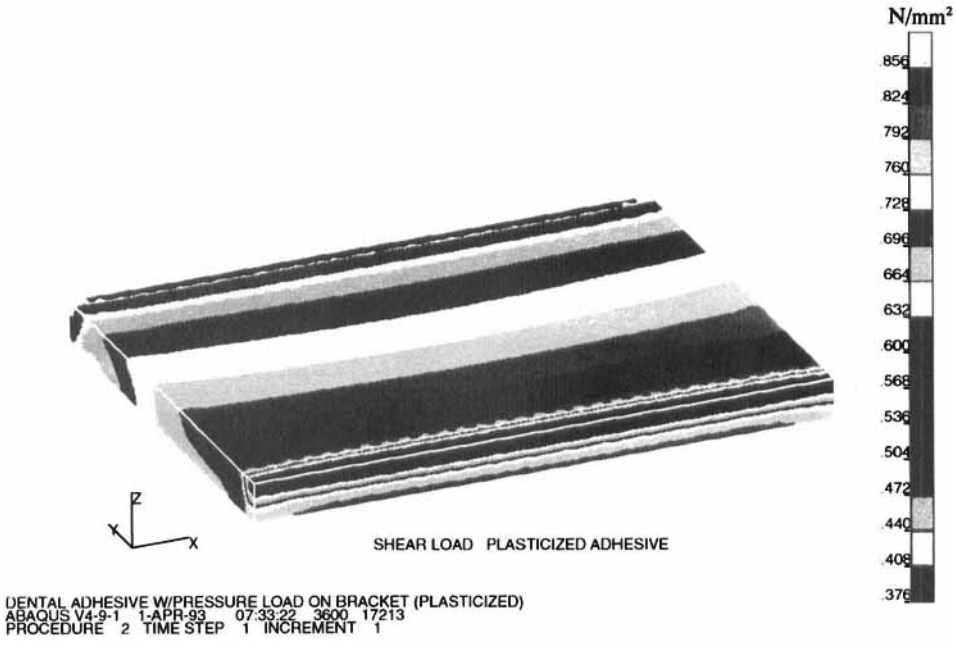

FIGURE 2 FEM von Mises stress distribution for the plasticized region applying the shear force conditions (simulating orthodontic "treatment force"). See Color Plate I.

Finite Element Results

Results from the FEA indicate that treatment of the adhesive as a separate and distinct layer with its own mechanical properties is satisfactory for describing the strength behavior of these bracket samples. The results of the simulation with 3.92 N orthodontic "treatment force" corresponds to very low computed stress values within the adhesive layer. The peaks of maximum stress are 0.84 N/mm^2 for the 10% plasticized adhesive and 0.93 N/mm^2 for the non-plasticized adhesive. A color contour plot for the computed stress state of the plasticized adhesive under this simulated loading condition is shown in Figure 2. Conceptually, the results are similar for the unplasticized case. These results indicate that a 10% DEP plasticized adhesive should survive the orthodontic loadings and should not result in premature failure during treatment.

Even more interesting are the modelling results when simulated torque loads nearly equal to the experimental failure torques are entered into the model. These results are shown in the color contour plot in Figure 3 for plasticized adhesive. The computed peak stress generated at the corners of the adhesive from the application of a 0.169 N-m (1.5 in-lb) loading is 22.8 N/mm^2 (3300 PSI) for the non-plasticized adhesive case and 20.3 N/mm^2 (2950 PSI) for the 10% plasticized adhesive case. Noting the reported tensile strengths for each adhesive (ultimate tensile

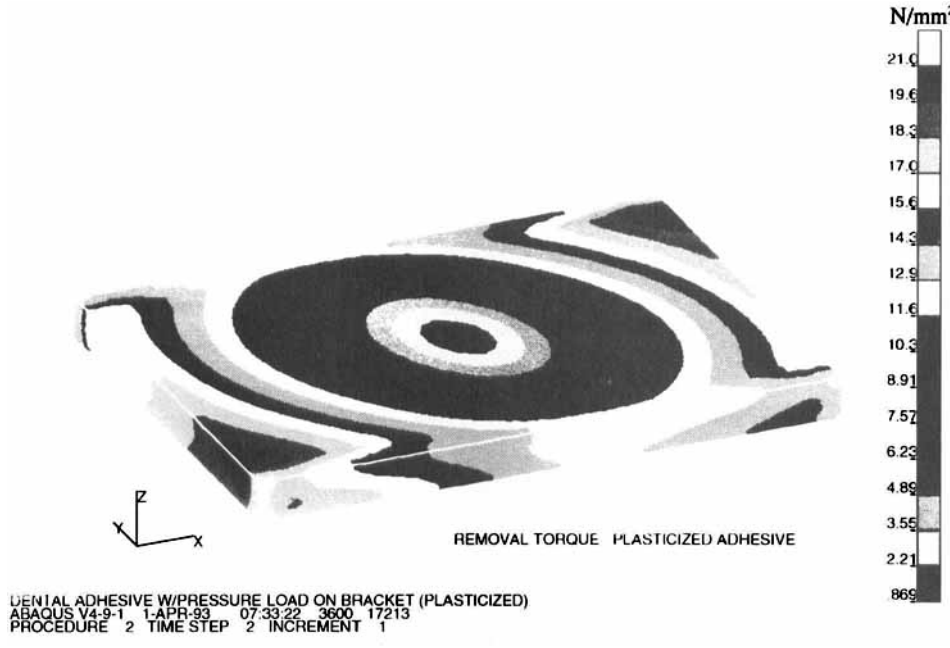

FIGURE 3 FEM von Mises stress distribution for the plasticized region applying typical torque removal force conditions (simulating "removal"). See Color Plate II.

strength = 18.6 N/mm^2 for 10% plasticized, 42.9 N/mm^2 for non-plasticized[9]), the modelling predicts cohesive failure within the adhesive when the torsion conditions are applied to the 10% plasticized adhesive. If the goal of adhesive modification is to make bracket removal easier/more predictable, then lowering the adhesive tensile strength to stress levels that are achievable during removal appears reasonable. Treatment of the adhesive as an interphase region with its own distinct and separate mechanical properties is reasonable from the finite element modelling and stress analyses.

Other factors need consideration. Tooth curvature and bracket curvature have not been included in our analytical model. In addition, the sealant layer is modelled as filled resin. Thus, the true adhesive zone will not be completely reinforced throughout the adhesive layer as has been modelled. In addition, the elastic modulus and tensile strength values for the adhesive would be more accurate using samples which were exposed to an oral environment. Also, there may be stress concentrators in our adhesion construction due to the wires in the wire mesh bonding disk which could be initiator points for failure. Finally, concerns about plastic deformation suggest that the 3-D linear elastic model might be inappropriate given the computed stresses. Nevertheless, the results are very much in line with what is experimentally observed.

CONCLUSIONS

This study leads to the following conclusions:

1. The modulus and tensile strength of these adhesive resins are significantly affected by the addition of DEP. These properties may only require slight modification since the adhesive is still required to withstand loading conditions during treatment and normal chewing forces. Thus, the highest amounts of plasticization may be undesirable for clinical use.
2. Salivary leaching leading to higher observed removal forces after treatment is not a major concern with this adhesive. Torsional bond strength measurements on the 10% plasticized samples are significantly lower than for the unplasticized samples even after exposure in the artificial saliva for 25 days at 60°C. More scatter is apparent in the 5% plasticized samples; however, it appears that plasticization leading to lower adhesive strength is a permanent effect.
3. The finite element analysis has shown that typical stresses calculated for the simulated treatment conditions should not lead to premature debonding with the 10% DEP plasticized resins. The treatment stresses computed from our finite element model are between 0.8 and 1.0 N/mm^2, far below the measured tensile strength for either the non-plasticized or 10% plasticized adhesives.
4. The treatment of the adhesive layer as an interphase region with mechanical properties of its own is reasonable. If the mechanical properties of the 10% plasticized adhesive are inserted into the FE model for the simulated torsional removal conditions, the computed stresses in the corners exceed the adhesive tensile strength where cohesive fracture was experimentally found. The peak stresses under the same conditions for the unplasticized adhesive are still well below the unplasticized adhesive tensile strength.

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References

1. M. L. Swartz, Ormco Technical Bulletin, April 1–15 (1988).
2. A. J. Gwinnett, *Am. J. Orthod. Dentofac. Orthop.* **93,** 346–348 (1988).
3. J. Odegaard and D. Segner, *Am. J. Orthod. Dentofac. Orthop.* **94,** 201–206 (1988).
4. F. A. Rueggeberg and P. E. Lockwood, *The Angle Orthodontist* **62**(1), 45–50 (1992).
5. J. J. Sheridan, G. Brawley and J. Hastings, *Am. J. Orthod. Dentofac. Orthop.* **89,** 21–27 (1986).
6. J. J. Sheridan, G. Brawley and J. Hastings, *Am. J. Orthod. Dentofac. Orthop.* **89,** 141–145 (1986).
7. K. Strobl, T. L. Bahns, L. Willham, S. E. Bishara and W. C. Stwalley, *Am. J. Orthod. Dentofac. Orthop.* **101,** 152–158 (1992).
8. A. J. Ostertag, V. B. Dhuru, D. J. Ferguson and R. A. Meyer, *Am. J. Orthod. Dentofac. Orthop.* **100,** 251–258 (1991).
9. K. E. Starling, Jr. and B. J. Love, *J. Clin. Ortho.,* in press (1993).
10. B. J. Love, R. F. Hochman and K. E. Starling, Jr., *Proc. 15th Annual Adh. Soc. Mtg.,* Feb. 1992, Hilton Head, SC. **15,** pp. 46–47.

11. J. F. Shackelford, *Introduction to Materials Science for Engineers,* 2nd ed. (MacMillan, New York, 1988), pp. 474–477.
12. M. Marek and E. Topfl, *Abstract # 1192. J. Dent. Res.* **65,** 301 (1986).
13. ABAQUS is a commercially-available finite element analysis program from Hibbitt, Karlsson, and Sorenson, Inc. Providence, Rhode Island, USA.
14. K. R. Williams and J. T. Edmundson, *Biomaterials* **5,** 347–351 (1984).
15. M. W. Beatty, M. L. Swartz, B. K. Moore, R. W. Phillips and T. A. Roberts, *J. Biomed. Mater. Res.* **27,** 403–413 (1993).