Eur. J. Prosthodont. Rest. Dent., Vol.16, No. 1, pp 10-14
Printed in Great Britain

Effect of Artificial Accelerated Aging on Color Stability and Surface Roughness of Indirect Composites

Fabíola Rejane Zanin*, Lucas da Fonseca Roberti Garcia*, Luciana Assirati Casemiro† and Fernanda de Carvalho Panzeri Pires-de-Souza†

Abstract - *Direct and indirect composite resins have different forms of polymerization. Some materials require a post-cure system associating light and heat enhancing clinical properties. This study assessed the changes in color and surface roughness of three indirect composite resins after accelerated aging. Twelve specimens (15x2mm) were obtained for each tested material. Subsequently, the first measurements for roughness tests and colorimetric spectrophotometry (CIE L*a*b* scale) were performed. Specimens were subject to accelerated aging for 384 hours. New measurements were then performed to evaluate the resulting change. Accelerated aging produced color change and increased surface roughness in all composite resins. Solidex® resin showed color changes above the clinically accepted value (ΔE = 4.31±0.22), and roughness values (Ra= 0.088±0,008 μm) statistically lower than that of Artglass® (Ra=0.141±0.026 μm) and Targis® (Ra=0.124±0.02 μm) (p<0.001). All the indirect resins tested showed color change and increased roughness after accelerated aging. Solidex® showed color stability above a quantitative level considered clinically acceptable and lower roughness values compared to the other resins.*

KEY WORDS: Material aging, Artificial aging, Colour stability, Surface roughness, Ceromers

INTRODUCTION

Indirect composite resin systems have been introduced with the attempt to solve problems concerning ceramic restorations (high abrasiveness causing the wear of antagonist teeth) and conventional resins (low resistance to wear and changes in color). These composites, called ceromers (CERamic Optimized polyMER), present a high density of inorganic ceramic particles, in comparison to the traditional direct and indirect composite resins, in addition to changes in the resin matrix. Moreover, they are an alternative material for esthetic restorations using porcelain and direct resins.

Color stability is crucial to the success of any type of esthetic restoration; in fact, color change is the major reason for replacing anterior restorations[1]. Researchers agree that direct restorations using composite resins suffer color changes with time[2-5]. Optical properties change with time, especially related to surface degradation[6], chemical reaction of the tertiary amine accelerator in composites[4,7], filler movement to the surface, and/or resin degradation[8]. Three types of composite resin discoloration are usually described: 1) external discolorations due to accumulation of plaque and stains[9,10]; 2) surface changes that imply on surface degradation and slight penetration and reaction of staining agents with the superficial layer of the composite resin[11,12]; 3) intrinsic discolorations due to physical-chemical reactions in the deep portions of the restoration, such as visible light and UV irradiation, and changes in temperature and humidity[13]. Physical-chemical stress induces the degradation of the composite surface and sub-surface. As a consequence, this will allow for the formation of microfractures on the restoration due to the pre-existence of micropores on the interface between the inorganic filler and the matrix. These microcavities represent a preferable pathway for the penetration of stains[14].

The composition of the composite resin matrix also has an influence over intrinsic discoloration[15]. The affinity of the resin for stains is modulated by its proportion of conversion of monomers into polymers[16] and by its chemical characteristics. Changes in the resin matrix because of water sorption lead to their discoloration and reduction in desirable physical qualities[17]. An insufficient resin conversion proportion will contribute to the absorption of staining substances. Chemical additives to the composite, mainly those that do not suffer reaction, as initiators, accelerators, and ultraviolet filters, may also degrade color components[18]. The characteristics of the filler particles also have a straight impact on the surface smoothness of the resin[19,20] and, consequently, on the susceptibility of the restoration to extrinsic staining[21]. The smoother the restoration, the smaller the capacity of stains impregnating on resin surface, due to smaller surface roughness.

Most ceromers use a post-cure system, with the association of light, heat, vacuum and pressure. Post-cure heating has been found to enhance surface hardness[22-24], flexure strength[24-27], color stability[23], and minimal polymerization shrinkage. It also contributes to the relaxation of the matrix-filler interface tensions[16], and to the conversion of monomers into polymers[18].

Thus, the purpose of this study was to assess the effect of artificial accelerated aging on color stability and surface roughness of three indirect composites. The hypothesis tested was that the post-cure process results in a minimal color change and low surface roughness after accelerated aging.

* DDS, MSD
† DDS, MSD, PhD

Table 1. *Composition of composite resins evaluated in the present study.*

Material	*Filler size (μm)*	*Filler weight (%)*	*Monomer composition*	*Polymerization Technique*
Artglass®	0.7 – 2.0	72	UDMA	UniXS® polymerization unit (Heraeus Kulzer®, Hanau, Germany): 180 seconds in light cycles generated by 2 xenon lamps (450-500 nm)
Targis®	0.03 – 1.0	80	Bis-GMA, TEGDMA	Targis® Quick fast polymerization unit (Ivoclar Vivadent®, Schaan, Liechtenstein): 40 seconds for pre-polymerization between increments. Targis Power® high-performance polymerization unit (Ivoclar Vivadent®): Visible light (8 high-power cold lamps, 400-450nm) and heat (95°C) in a 25 minutes.
Solidex®	0.16 – 7.0	53	UDMA	Solidite® light polymerization unit (Shofu®, Kioto, Japan): 180 seconds.

UDMA – Urethane-dimethacrylate;
Bis-GMA - Bisphenol glycidyl methacrylate;
TEGDMA – triethyleneglycol-dimethacrylate.

MATERIAL AND METHODS

Three indirect composites were selected for this study: Artglass® (Heraeus Kulzer®, Hanau, Germany); Solidex® (Shofu®, Kioto, Japan); Targis® (Ivoclar Vivadent, Schaan®, Liechtenstein), all of them in the color B2 for dentin (Table 1). Twelve specimens (15x2mm) were fabricated for each resin, using a stainless steel matrix. The material was inserted in the matrix by 2 increments of 1mm, and polymerized in specific polymerization units following the manufacturer's instructions (Table 1). The last increment was leveled with a glass plate so the excess of resin material could be removed.

The specimens had their surface polished (Sof-Lex®, 3M Dental Products®, St Paul, MN, USA), in the sequence of higher to lower abrasiveness. All specimens were coded and stored (100% humidity and absence of light for 24h). After this period, the first measurements were made for the roughness and colorimetric spectrophotometry tests.

Roughness:

The needle of the profilometer (Surfcorder SE 1700 profilometer®, Kosakalab®, Tokio, Japan) was positioned on each specimen, and three readings were performed in different places of the surface. The instrument presented the mean surface roughness values, considered as the initial roughness values or control. After the process of accelerated aging, new roughness readings were performed following the same method. The values were submitted to 1-way ANOVA with level of significance at 5%.

Colorimetric Spectrophotometry:

A PCB 6807 BYK GARDNER® spectrophotometer (Geretsried®, Germany) was used for color reading. Specimens were placed on the white standard block (Standard for 45°, 0° Reflectance and Color Gardner Laboratory Inc. ®, Bethesda, Maryland 20014) and the first reading was made. The observation pattern simulated by the spectrophotometric colorimeter follows the system CIE L*a*b*. Initial color readings were performed before the specimens were subject to the artificial accelerated aging process and color stability was determined by the difference (ΔE) between the coordinates obtained for the specimens before and after the aging process, by the formula:

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}$$

Values of $\Delta E \geq 3.3$ are considered clinically unacceptable[5]. The ΔE values were compared using 1-way analysis of variance (level of significance at 5%).

Artificial accelerated aging:

Specimens were subject to artificial accelerated aging (Accelerated Aging System for non-metallic C-UV®, Comexim Matérias Primas Ltda®, São Paulo, Brazil) for 384 hours, by exposure to 4 hours to UV-B light at 50° C (280/320nm) and 4 hours of water condensation at 50° C.

Table 2. *Means (standard deviation) of color change value (ΔE)*

Materials	*ΔE*
Artglass®	2.99(1.33)^{ns}
Solidex®	4.31(0.22)^{a}
Targis®	3.01(0.86)^{ns}

ns – no statistically significant difference (p>0.05),
a – statistically different (p<0.05)

Table 3. *Difference of means (μm) and standard deviation of roughness values (Ra) before and after artificial accelerated aging*

Materials	*Roughness*
Artglass®	0.141(0.026)^{a}
Solidex®	0.088(0.008)^{ns}
Targis®	0.124(0.02)^{a}

ns – no statistically significant difference (p>0.05),
a – statistically different (p<0.001)

RESULTS

Color change

All indirect resins submitted to the process of artificial accelerated aging showed color changes (Table 2). Solidex® showed greater color variation. Such values were above those considered clinically accepted (Table 2). Differences were statistically significant ($p<0.05$) compared to the other composite resins, which presented color variation within the clinically accepted range ($\Delta E<3.3$).

Roughness

By assessing the mean roughness values (Ra) before and after artificial accelerated aging, it could be observed that there was an increase in the roughness of the specimens after aging (Fig. 1 and Table 3).

Artglass® and Targis® suffered the greatest surface changes, showing a statistically significant increase in roughness after artificial accelerated aging ($p<0.001$). Changes in Solidex® were not statistically significant ($p>0.05$).

DISCUSSION

The materials used in this study are recommended for use in indirect restorations and suffer some additional polymerization during the curing process. The properties of these materials are different from those of direct composite resins. This is especially true for polymerization shrinkage, which may be better controlled in indirect materials, since it occurs outside the mouth.

Various mechanisms have been suggested for changes due to aging. The Accelerated Aging System has been adopted to test the color stability of dental resins since 1978[28] and intended to reproduce the weathering effects that occur when materials are exposed to sunlight and moisture. Although the manufacturer of the device estimates that 300 hours of exposure is equivalent to 1 year of service, it is not known how an outdoor simulation such as weathering would translate to intraoral conditions[29]. Therefore, even though the method mimics situations different from the oral environment, it was chosen because the intention was to induce property changes associated with moisture and heat. The oral environment is severe and this study had the purpose to evaluate the chromatic changes of materials submitted to degradation processes. The UV-B lamp emits radiation around 300nm. It provides a severe aging process, which produces rapid polymer degradation and often causes degradation by mechanisms that do not occur when materials are exposed to sunlight[30].

The CIE L*a*b* system uses the three parameters L*, a* and b* to define color[31]. The degree of lightness and darkness corresponds to L*, while a* and b* are on the chromatic scale and ranges from red (+a*) to green (-a*) and yellow (+b*) to blue (-b*). Authors have documented that ΔE values between 2–3 are perceptible to the naked eye[24] and that ΔE values greater than 3.3 are clinically unacceptable[5]. Hence, in terms of color stability, the present study results show that Targis® and Artglass® show a perceptible, but not unacceptable, color change. The average color changes were of 2.99 and 3.01 units of ΔE, respectively, which is in agreement with the results of a previous study[8]. On the other hand, Solidex® indirect resin showed the highest color instability with mean ΔE equal to 4.31, a critical color change value.

The variations in values of L*, a*, and b* after artificial accelerated aging were specific for each tested resin. Solidex® resin showed a reduction in the value L*, becoming darker, and with an increase in the value b*, which makes it more yellow after aging. Value a* decreased, causing the resin to discolor. Targis® and Artglass® resins showed an increase in luminosity (value L*) and a reduction in the values of chroma. Artglass® resin showed a reduction only for the value b*, losing the yellow tone. Whereas Targis® resin did not show a statistically significant difference for the value b* after aging, and continued practically with the same tone of yellow, but it did lose some tone of red with the reduction of value a*. These results are different from other reports[17], but a different method for accelerated aging was used in which the samples were immersed in 60°C distilled water for 8 weeks.

Some authors[32] have reported that the physical characteristics of filling resins deteriorate when immersed in water because water sorption leads to their discoloration and reduction in desirable physical qualities. However, composite materials have different levels of water sorption depending on the type of monomer they are made of, and different monomer might also cause the materials to vary in color[17].

In this study, two materials contain UDMA and one contains Bis-GMA/TEGDMA, and color changes were observed in all materials. However, for the polymerization of Targis resin, which contains Bis-GMA, there is a post-curing heating process. Moreover, color change was within clinically

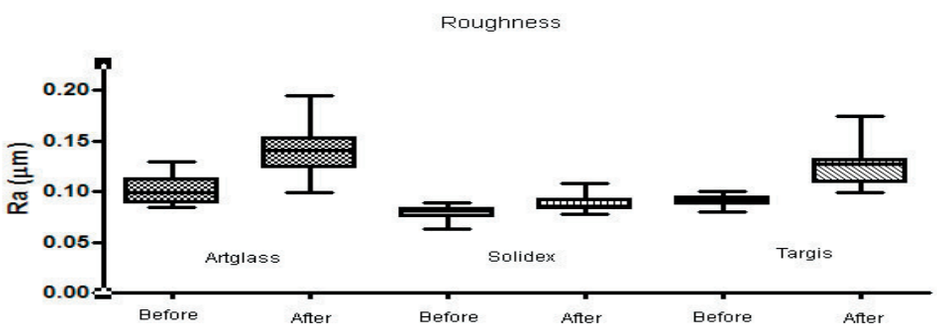

Figure 1. *Mean values and standard deviation of roughness Ra for the tested resins, before and after artificial accelerated aging*

accepted levels, which may have resulted from the better monomer conversion into polymer as a result of this heating. In a previous study[33], authors verified that post-cure heating significantly decreases the amounts of unreacted monomer remaining after the initial light curing stage, and that at 50°C there was a 75% reduction in remaining unreacted Bis-GMA[33].

In the present study, the composites with the lowest filler contents had poor color stability, supporting results of previous work[34,35]. The results demonstrated that Solidex®, with 53% of inorganic particles in weight (39% in volume), showed the greatest color change, followed by Targis® resin, which presents 80% of inorganic particles in weight (68% in volume). Artglass® resin, which showed color stability similar to that of Targis® resin, presents 72% of inorganic particles in weight (58% in volume).

Color change can also be explained due to filler exposure and degradation of polymer material[36]. Authors have suggested that the aging process increased the C:O, C:Si, and C:Ba ratios at least threefold, suggesting that some inorganic materials were lost from the surface. Besides, the process can modify the bonding interface between filler and resin (silane treatment). Elemental oxygen is lost when barium oxide and silica fillers are depleted, leaving more organic material on the surface. Surface deterioration has been shown to increase lightness and decrease chroma of restorative resins[4]. The results of presented research sustain this assertion.

Increased surface roughness after accelerated aging has been attributed to wear of the resin[6,37] or exposure of interior porosities[38,39]. Composite surface roughness may be influenced by the size and composition of filler particles[40]. After the process of aging, all tested resins showed an increase in roughness. Artglass® and Targis® resins showed the highest roughness values after artificial accelerated aging, whereas Solidex® resin showed statistically lower roughness values as compared to the other resins.

Before aging, Solidex® resin had the lowest Ra value lower (although not significantly lower) than that of the other resins and after aging of the structure, there is a chance of exposing the inorganic particles on the surface of the specimens causing an increase in roughness[41]. Since Solidex® shows a smaller amount of filler particles in its composition, it might be suggested that this was why it showed better results than Artglass® and Targis® resins. The latter have greater matrix volume, which may have caused greater particle exposure after artificial accelerated aging, leading to higher surface roughness. However, microscopic images are needed in order to confirm this theory. Higher surface roughness causes more light scattering and loss of gloss[8]. Campbell *et al.*[42] reported a linear relationship between the optical scattering coefficient and the filler concentration. This was not confirmed in the present study. Solidex® resin demonstrated the lowest roughness values, but the highest color change values.

By analyzing the study's results and conditions, the hypothesis that resins subject to post-cure heat treatment could present lower color change and smoother surfaces can be partially accepted. Targis presented clinically accepted color change ($\Delta E<3.3$), but increase in surface roughness

Other factors are associated with these properties. Further investigations are needed in order to draw conclusions about the proportion of conversion of this material's monomers, as to analyze its influence over the properties studied in the present experiment.

CONCLUSIONS

All resins showed color change and an increase in roughness after the process of artificial accelerated aging. Artglass® and Targis® resins showed color changes within the acceptable limit ($\Delta E <3.3$) whilst Solidex® resin showed the greatest color change among the resins, being clinically unacceptable ($\Delta E >3.3$). This material showed the smallest change in surface roughness after artificial accelerated aging.

ADDRESS FOR CORRESPONDENCE

Profa. Dra. Pires-de-Souza, Faculdade de Odontologia de Ribeirão Preto – USP, Departmento de Materiais Dentários e Prótese FORP-USP, Avenida do Café, s/n Bairro Monte Alegre, CEP 14040-904, Ribeirão Preto, São Paulo, Brasil. E-mail: ferpanzeri@forp.usp.br

REFERENCES

1. Kroeze, H.J., Plasschaert, A.J., Van 't Hof, M.A., Truin, G.J. Prevalence and need for replacement of amalgam and composite restorations in Dutch adults. *J. Dent. Res.*, 1990; **69**:1270-1274.
2. Dietschi, D., Campanile, G., Holz, J., Meyer, J.M. Comparison of the color stability of ten new-generation composites: an in vitro study. *Dent. Mater.*, 1994; **10**:353-362.
3. Powers, J.M., Bakus, E.R., Goldberg, A.J. In vitro color changes of posterior composites. *Dent. Mater.*, 1988; **4**:151-154.
4. Powers, J.M., Fan, P.L., Raptis, C.N. Color stability of new composite restorative materials under accelerated aging. *J. Dent. Res.*, 1980; **59**:2071-2074.
5. Ruyter, I.E., Nilner, K., Moller, B. Color stability of dental composite resin materials for crown and bridge veneers. *Dent. Mater.*, 1987; **3**:246-251.
6. Powers, J.M., Fan, P.L. Erosion of composite resins. *J. Dent. Res.*, 1980; **59**:815-819.
7. Uchida, H., Vaidyanathan, J., Viswanadhan, T., Vaidyanathan, T.K. Color stability of dental composites as a function of shade. *J. Prosthet. Dent.*, 1998; **79**:372-377.
8. Schulze, K.A., Marshall, S.J., Gansky, S.A., Marshall, G.W. Color stability and hardness in dental composites after accelerated aging. *Dent. Mater.*, 2003; **19**:612-619.
9. Guler, A.U., Kurt, S., Kulunk, T. Effects of various finishing procedures on the staining of provisional restorative materials. *J. Prosthet. Dent.*, 2005; **93**:453-458.
10. Villalta, P., Lu, H., Okte, Z., Garcia-Godoy, F., Powers, J.M. Effects of staining and bleaching on color change of dental composite resins. *J. Prosthet. Dent.*, 2006; **95**:137-142.
11. Imazato, S., Tarumi, H., Kato, S., Ebisu, S. Water sorption and colour stability of composites containing the antibacterial monomer MDPB. *J. Dent.*, 1999; **27**:279-283.
12. Lee, Y.K., Lim, B.S., Powers, J.M. Color changes of dental resin composites by a salivary enzyme. *J. Biomed. Mater. Res. B Appl. Biomater.*, 2004; **70**:66-72.
13. Ferracane, J.L., Moser, J.B., Greener, E.H. Ultraviolet light-induced yellowing of dental restorative resins. *J. Prosthet. Dent.*, 1985; **54**:483-487.
14. Mair, L.H. Staining of in vivo subsurface degradation in dental composites with silver nitrate. *J. Dent. Res.*, 1991; **70**:215-220.
15. Miettinen V.M., Narva, K.K,. Vallittu, P.K. Water sorption, solubility and effect of post-curing of glass fibre reinforced polymers. Biomaterials, 1999; **20**:1187-94.
16. de Gee, A.J., ten Harkel-Hagenaar, E., Davidson, C.L. Color dye for identification of incompletely cured composite resins. *J. Prosthet. Dent.*, 1984; **52**:626-631.

17. Nakamura, T., Saito, O., Mizuno, M,. Tanaka, H. Changes in translucency and color of particulate filler composite resins. *Int. J. Prosthodont.*, 2002; **15**:494-499.
18. Asmussen, E. Factors affecting the quantity of remaining double bonds in restorative resin polymers. Scand. *J. Dent. Res.*, 1982; **90**:490-496.
19. Tjan, A.H., Chan, C.A. The polishability of posterior composites. *J. Prosthet. Dent.*, 1989; **61**:138-146.
20. van Noort, R., Davis, L.G. The surface finish of composite resin restorative materials. *Br. Dent. J.*, 1984; **157**:360-364.
21. Shintani, H., Satou, J., Satou, N., Hayashihara, H., Inoue, T. Effects of various finishing methods on staining and accumulation of Streptococcus mutans HS-6 on composite resins. *Dent. Mater.*, 1985; **1**:225-227.
22. Bausch, J.R., de Lange, C., Davidson, C.L. The influence of temperature on some physical properties of dental composites. *J. Oral. Rehabil.*, 1981; **8**:309-317.
23. Wendt, S.L. Jr. The effect of heat used as secondary cure upon the physical properties of three composite resins. II. Wear, hardness, and color stability. *Quintessence Int.*, 1987; **18**:351-356.
24. Ferracane, J.L., Condon, J.R. Post-cure heat treatments for composites: properties and fractography. *Dent. Mater.*, 1992; **8**:290-295.
25. Asmussen, E., Peutzfeldt, A. Mechanical properties of heat treated restorative resins for use in the inlay/onlay technique. Scand. *J. Dent. Res.*, 1990; **98**:564-567.
26. Loza-Herrero, M.A., Rueggeberg, F.A. Time-temperature profiles of post-cure composite ovens. *Gen. Dent.*, 1998; **46**:79-83.
27. Loza-Herrero, M.A., Rueggeberg, F.A., Caughman, W.F., Schuster, G.S., Lefebvre, C.A., Gardner, F.M. Effect of heating delay on conversion and strength of a post-cured resin composite. *J. Dent. Res.*, 1998; **77**:426-431.
28. Powers, J.M., Dennison, J.B., Koran, A. Color stability of restorative resins under accelerated aging. *J. Dent. Res.*, 1978; **57**:964-970.
29. Douglas, R.D. Color stability of new-generation indirect resins for prosthodontic application. *J. Prosthet. Dent.*, 2000; **83**:166-170.
30. ASTM STANDARDS G154-00A. Standard practice for operating fluorescent light apparatus for UV exposure of nonmettalic materials. Annual Book of ASTM Standards, v. 14.04. ASTM-Standards. Pennsylvania: United States, 2006 p. 646-654.
31. C.I.E. Commission Internationale de l'Eclarigage. Recommendations on uniform color spaces. Color difference equations, and metric color terms. *Suppl No. 2* to Pub No. 15. Paris: Bureau Central de la CIE, 1978.
32. Drummond, J.L., Botsis, J., Zhao, D., Samyn, J. Fracture properties of aged and post-processed dental composites. *Eur. J. Oral Sci.*, 1998; **106**:661-666.
33. Bagis, Y.H,. Rueggeberg, F.A. The effect of post-cure heating on residual, unreacted monomer in a commercial resin composite. *Dent. Mater.*, 2000; **16**:244-247.
34. Eldiwany, M., Friedl, K.H., Powers, J.M. Color stability of light-cured and post-cured composites. Am. *J. Dent.*, 1995; **8**:179-181.
35. Vichi, A., Ferrari, M., Davidson, C.L. Color and opacity variations in three different resin-based composite products after water aging. *Dent. Mater.*, 2004; **20**:530-534.
36. Schulze, K.A., Tinschert, J., Marshall, S.J., Marshall, G.W. Spectroscopic analysis of polymer-ceramic dental composites after accelerated aging. *Int. J. Prosthodont.*, 2003; **16**:355-361.
37. Davis, B.A., Friedl, K.H., Powers, J.M. Color stability of hybrid ionomers after accelerated aging. *J. Prosthodont.*, 1995; **4**:111-115.
38. Gladys, S., Van Meerbeek, B., Braem, M., Lambrechts, P., Vanherle, G. Comparative physico-mechanical characterization of new hybrid restorative materials with conventional glass-ionomer and resin composite restorative materials. *J. Dent. Res.*, 1997; **76**:883-894.
39. Mathis, R.S., Ferracane, J.L. Properties of a glass-ionomer/resin-composite hybrid material. *Dent. Mater.*, 1989; **5**:355-358.
40. Tanoue, N., Matsumura, H., Atsuta, M. Wear and surface roughness of current prosthetic composites after toothbrush/dentifrice abrasion. *J. Prosthet. Dent.*, 2000; **84**:93-97.
41. Cho, L.R., Yi, Y.J., Heo, S.J. Effect of tooth brushing and thermal cycling on a surface change of ceromers finished with different methods. *J. Oral Rehabil.*, 2002; **29**:816-822.
42. Campbell, P.M., Johnston, W.M., O'Brien, W.J. Light scattering and gloss of an experimental quartz-filled composite. *J. Dent. Res.*, 1986; **65**:892-894.