In vitro Accelerated Aging of Composites and a Sealant

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The in vitro *accelerated aging of conventional and microfilled composite restorative materials and a sealant was studied. Volume loss/surface area ranged from 2.0 to 7.3 x 10^{-3} mm^3/mm^2 after 900 h of aging. Surface morphology changed more dramatically for the conventional composites than for the other materials. Changes in surface chemistry as measured by FTIR-ATR were observed.*

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Introduction.

Surface degradation of composite restorative materials has been studied *in vitro* by accelerated aging in a weathering chamber.[1] Aging for 900 h caused erosion of the resin matrices and exposure of filler particles. Crazing of the resin matrices was similar to that reported *in vivo* by O'Brien and Yeh.[2] Differences in surface profiles of the aged composites suggested that the materials eroded at different rates.

The purpose of this study was to evaluate weight loss, volume loss, and changes in surface chemistry and morphology of conventional and microfilled composites and a sealant after accelerated aging *in vitro*.

Materials and methods.

The *in vitro* accelerated aging of eight commercial composite restorative materials and a pit and fissure sealant was evaluated. The code, shade, batch number, and manufacturer are listed in Table 1 for each of five conventional composites (A, AR, C, PR, and S), three microfilled composites (I, PF, and SF), and a sealant (D).

Five samples (20 mm diameter and 1mm thick) were prepared for each material by polymerization in a die. The samples were placed in an oven at 37°C within 90 s after initiating the mix and were stored for 24 h before drying to a constant weight over freshly regenerated silica gel in a desiccator. The balance* was accurate to 0.1 mg.

The samples were exposed to conditions of accelerated aging for 900 h in a weathering chamber† at 43°C and 90% relative humidity. One surface of each sample was subjected continuously to the radiation of a 2500 watt light source‡ filtered by borosilicate glass and to an intermittent water spray for 18 min every two h. The intensity of the uv radiation was measured with a uv meter§ to be about 11.2 mWatts/cm^2 at the sample position. After aging the samples were dried again to constant weight. The change in weight caused by aging was calculated and used to determine volume loss. Density was determined on one new sample of each material by a water displacement technique. The exposed surface area of a sample was calculated by geometry to be 377 mm^2.

Surface morphology of the materials was observed after 900 h of aging by scanning electron microscopy.// Changes in the surface chemistry of AR and SF were studied by Fourier transform infrared spectroscopy

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The infrared spectra were run by Digilab, Inc., as a courtesy.

*Model H 14, Mettler Instrument Corp., Princeton, NJ 08540

†Weather-Ometer 25-WR, Atlas Electric Devices Co., Chicago, IL 60613

‡12-2881 Xenon Burner, Atlas Electric Devices Co., Chicago, IL 60613

§Model J-221, Ultraviolet Products, Inc., San Gabriel, CA 91778

//JSM-U3, Japan Electron Optics Laboratory, Japan

TABLE 1
CODE, NAME, BATCH NUMBERS, and MANUFACTURER OF PRODUCTS TESTED

Code	Product Name (Shade)	Batch Numbers		Manufacturer
Conventional Composites:				
A	Adaptic (Universal)	base catalyst	– 8C 004 – 8C 004	Johnson & Johnson Dental Products Division East Windsor, NJ 08520
AR	Adaptic Radiopaque (Universal)	base catalyst	– 8H 109 – 8H 109	Johnson & Johnson
C	Concise (Universal)	paste A paste B	– 80 23 – 8L 23	3M Company St. Paul, MN 55101
PR	Profile (Shade 55)	base catalyst	– 47812 – 37901	S.S. White, Div. of Pennwalt Philadelphia, PA 19102
S	Simulate (Universal)	base catalyst	– 91037 – 92044	Kerr Manufacturing Co. Div. of Sybron Corp. Romulus, MI 48174
Microfilled Composites:				
I	Isopast (Shade 22)	base catalyst	– 721078 – 630179	Vivadent (USA), Inc. Buffalo, NY 14214
PF	Phaseafill (Shade GO)	base catalyst	– 026 – 044	Phasealloy, Inc. El Cajon, CA 92021
SF	Superfil (Universal)	universal catalyst	– 80338 – 80329	Harry J. Bosworth Co. Skokie, IL 60076
Sealant:				
D	Delton	universal catalyst	– 5001 2792 EA – 5001 2792 DA	Johnson & Johnson

TABLE 2
PARAMETERS DESCRIBING AGING OF CONVENTIONAL AND MICROFILLED COMPOSITES AND A SEALANT

Code	Weight Loss/ Surface Area, mg/cm^2	Density g/cm^3	Volume Loss/ Surface Area, 10^{-3} mm^3/mm^2
Conventional Composites:			
A	1.16 (0.06)*	1.960	5.9 (0.3)†
AR	0.70 (0.11)	1.982	3.6 (0.6)
C	1.09 (0.10)	2.098	5.2 (0.5)
PR	0.46 (0.10)	2.112	2.1 (0.4)
S	1.03 (1.29)	2.046	5.1 (1.4)
Microfilled Composites:			
I	0.28 (0.10)	1.392	2.0 (0.7)
PF	0.51 (0.08)	1.462	3.5 (0.6)
SF	1.03 (0.21)	1.407	7.3 (1.6)
Sealant:			
D	0.46 (0.04)	1.188	3.9 (0.4)

*Mean value of five replications with standard deviations in parentheses. The Tukey interval at the 95% level of confidence was 0.26 mg/cm^2.

†The Tukey interval at the 95% level of confidence was 1.5×10^{-3} mm^3/mm^2.

(FTIR)[#] in an attenuated total reflectance (ATR) mode using a thallium bromide-iodide crystal (KSR-5). Reflectance spectra were obtained at zero and 900 h for a representative sample of AR and SF between 400 and 4000 cm^{-1}. Difference spectra were obtained by subtraction of 900-hour from zero-hour spectral data using a computer program. The spectra were factored to cancel sharp C-H peaks in the subtraction. The resolution of the spectrophotometer was 0.08 cm^{-1}.

[#]Model FTS-20C/D, Digilab, Inc., Cambridge, MA 02139

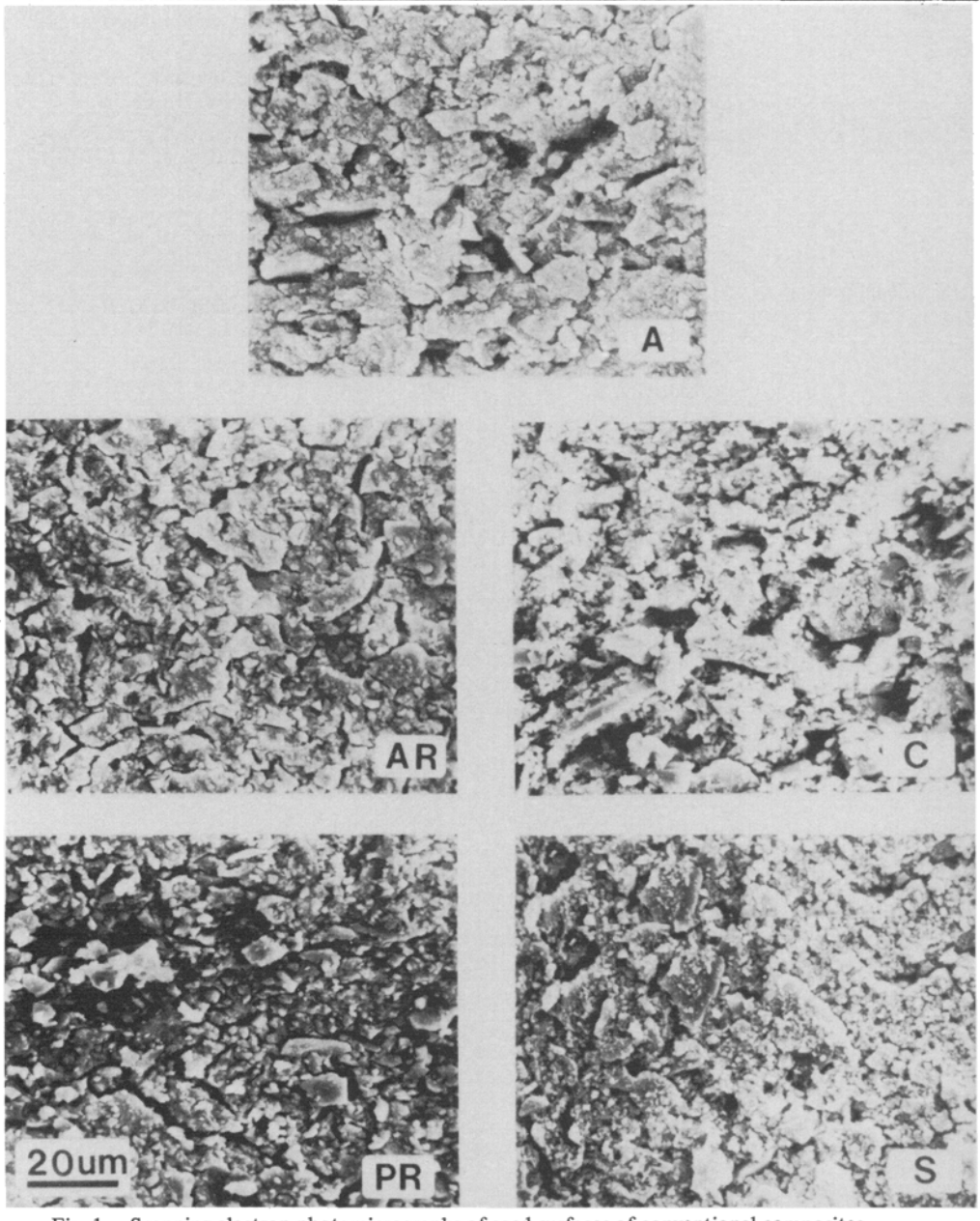

Fig. 1 – Scanning electron photomicrographs of aged surfaces of conventional composites.

Results.

Parameters describing the aging of conventional and microfilled composites and a sealant are listed in Table 2. Weight loss per exposed surface area after 900 h of aging ranged from 0.46 to 1.16 mg/cm^2 for the conventional composites and from 0.28 to 1.03 mg/cm^2 for the microfilled composites and was 0.46 mg/cm^2 for the sealant. Volume loss per exposed surface area was determined from the data of weight loss, density, and surface area. It ranged from 2.1 to 5.9 x 10^{-3} mm^3/mm^2 for the conventional composites and from 2.0 to 7.3 x 10^{-3} mm^3/mm^2 for the microfilled composites and was 3.9 x 10^{-3} mm^3/mm^2 for the sealant. Values of Tukey's interval at the 95% level of confidence were 0.26 mg/cm^2 and 1.5 x 10^{-3} mm^3/mm^2, respectively.

Scanning electron photomicrographs of the aged surfaces of the conventional composites and the microfilled composites and sealant are shown in Figs. 1 and 2, respectively. Crazing of the surface and exposure of filler particles were observed for the conventional composites. The surfaces of the microfilled composites also showed crazing. The surface of the sealant (D) appeared unchanged. The surfaces of all materials at baseline appeared smooth and without characteristic morphology, much like the surface of D in Fig. 2.

Infrared ATR spectra at zero and 900 h and difference spectra (zero minus 900 h) of AR and SF are shown in Figs. 3 and 4, respectively. Changes in the spectra between zero and 900 h (Fig. 3) were observed for AR and were different from those observed for SF (Fig. 4).

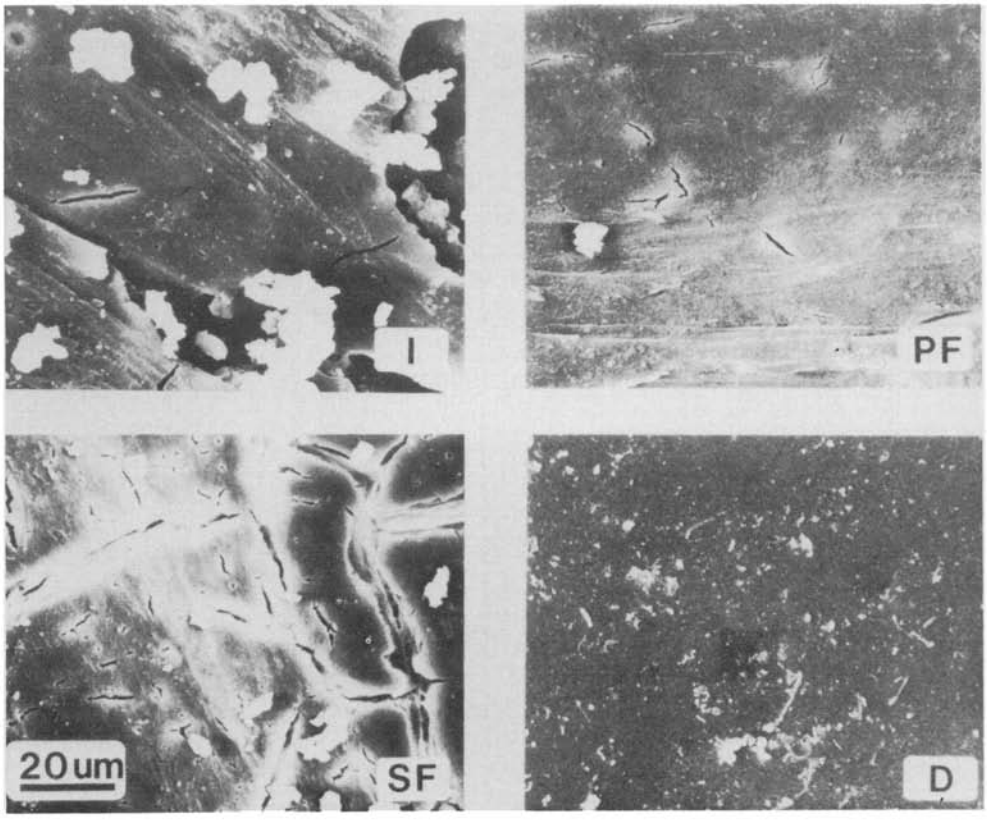

Fig. 2 – Scanning electron photomicrographs of aged surfaces of microfilled composites and a sealant (D).

Discussion.

Similarities in the relative peak heights of spectra of AR and SF before and after aging confirm that loss of material from the surface by dissolution is occurring. Peaks observed at 1715 and 1170 cm^{-1} in the difference spectrum of AR (Fig. 3), however, are characteristic of an aliphatic carbonyl absorption. Changes in these peaks could be explained by photo-oxidation of the resin of AR. Conjugation resulting from condensed carbonyl compounds could explain changes in color reported for AR during accelerated aging.[3] Clinically, photo-oxidation is an unlikely mechanism, but oxidation of the resin could occur in the presence of activated oxygen that might form chemically in the oral environment.

In vitro wear characteristics of commercial composites exposed to conditions of accelerated aging have been shown to differ from those of the unaged materials.[4] Changes in wear track width, tangential force, and morphologies of surface failure caused by single-pass sliding were attributed to surface degradation. The weight loss data and preliminary FTIR-ATR spectra further support a hypothesis that changes in the surface chemistry of the composites occur during accelerated aging.

Comparisons of the weight loss and volume loss data (Table 2) do not suggest superiority of either the conventional or microfilled composites as a group, whereas two-body abrasion data[5] demonstrate that conventional composites are more wear resistant *in vitro*. It is probable that wear of composites *in vivo* is affected by both mechanical and chemical mechanisms of degradation. For example, clinical data[6] of composites A and S in Class 1 posterior restorations in humans suggest that initial wear results in loss of occlusal contact with opposing teeth and loss of margin integrity, with S changing more rapidly than A. After 18 mo, however, no difference in loss of anatomic form can be detected between A and S. The present data suggest that the aging processes of A and S are similar, although A has been demonstrated to be superior to S in resistance to mechanical wear.[7]

Further research is necessary to more completely characterize the chemical degradation of composites that occurs as a

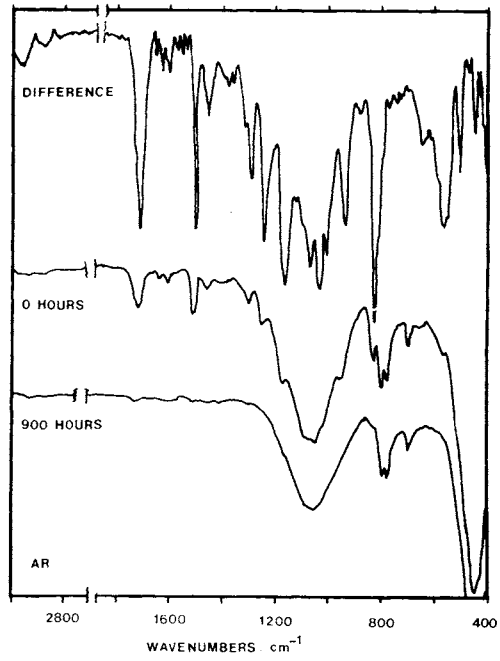

Fig. 3 – Infrared ATR spectra at zero and 900 h and difference spectrum (zero minus 900 h) of AR.

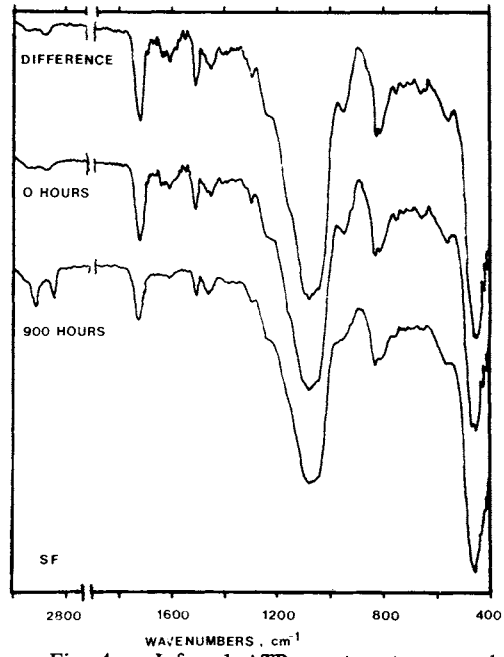

Fig. 4 – Infrared ATR spectra at zero and 900 h and difference spectrum (zero minus 900 h) of SF.

result of accelerated aging *in vitro* and that which may occur *in vivo*.

Conclusions.

The *in vitro* accelerated aging of conventional and microfilled composite restorative materials and a sealant was studied. Volume loss/surface area ranged from 2.0 x 10^{-3} mm^3/mm^2 for I to 7.3 x 10^{-3} mm^3/mm^2 for SF after 900 h of aging. Surface morphology of the conventional composites was characterized by crazing and exposure of filler particles. The surfaces of the microfilled composites also showed crazing. The surface morphology of the sealant appeared unchanged. Comparisons of infrared ATR spectra between zero and 900 h of aging showed that slight chemical changes occurred at the surface of AR but not SF.

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