Cells and Materials

Volume 7 | Number 2 Article 2

1997

Fractals, Surface Roughness, and Fracture Toughness of Dental Composite and Unfilled Resin Fracture Surfaces

James L. Drummond
University of Illinois

Cells and Materials Vol. 7, No. 2, 1997 (Pages 95-102) 1051-6794/97$5.00+.25
Scanning Microscopy International, Chicago (AMF O'Hare), IL 60666 USA

FRACTALS, SURFACE ROUGHNESS, AND FRACTURE TOUGHNESS OF DENTAL COMPOSITE AND UNFILLED RESIN FRACTURE SURFACES

James L. Drummond

Department of Restorative Dentistry, University of Illinois at Chicago,
337A College of Dentistry, m/c 555, 801 S. Paulina Street, Chicago, Illinois 60612-7212
Telephone number: 312-413-3790 / FAX number: 312-996-3535 / E-mail: drummond@uic.edu

(Received for publication April 1, 1997 and in revised form October 2, 1997)

Abstract

The intent of this project was to evaluate whether or not there exists a correlation between fractal dimension, surface roughness, and fracture toughness of an unfilled resin and a filled composite. The fracture surfaces of the unfilled resin and the filled composite were examined using atomic force microscopy (AFM) and a surface roughness measuring device. The specimens examined were aged in distilled water or air at 37°C for up to 12 months. Line scans of 1.4 μm for the atomic force microscope and 0.25 mm for the surface analyzer across the fracture surface were conducted on each specimen. Comparisons were made between the fracture toughness (K_{Ic}) of the specimens, the fractal dimensional increment (D^*), and surface roughness (R_a). No correlation was found.

Key Words: Fractal, surface roughness, fracture surface, dental composite, fracture toughness.

Introduction

The original project from which this data evolved was to investigate the fracture characteristics of a single resin system with different weight percentages of glass filler aged for 6 to 12 months in air and distilled water at 37 °C (Drummond *et al.*, 1995). Since the fracture toughness and flexure strength of these glass filled resins were similar to the properties of lithia based glass ceramics (Anusavice and Zhang, 1995) which had demonstrated a relationship between fracture toughness and fractal dimension (Hill *et al.*, 1995; Naman *et al.*, 1994; Thompson *et al.*, 1995), the study was expanded to determine if such a relationship existed for these glass filled resins. The use of fractals to explain the geometry of fracture is well known (Mandelbrot, 1983; Mandelbrot *et al.*, 1984). Mecholsky's group has suggested that there is a correlation between an increase in the fractal dimensional increment (D^*) and an increase in the fracture toughness (K_{Ic}) (Mecholsky, 1991; Mecholsky and Plaia, 1992; Mecholsky *et al.*, 1989). Scanning tunneling microscopy has been used for fractal analysis of fatigue surfaces (Lankford and Longmire, 1991; Mitchell and Bonnell, 1990) for fine scale detail and topographical analysis. Other studies have used atomic force microscopy (AFM) in conjunction with the slit-edge technique to determine the fractal dimension and the correlation with fracture toughness on glass ceramics (Hill *et al.*, 1995; Naman *et al.*, 1994; Thompson *et al.*, 1995). These results have shown a positive relationship between the fractal dimensional increment and fracture toughness, i.e., the tougher the material, the more tortuous the fracture surface produced during catastrophic failure. Also, the two methods of determining the fractal dimensional increment, AFM and slit-edge, gave similar results. A study by Baran *et al.* (1992) found no correlation between the fractal dimension and fracture toughness of glass and dental porcelains. The intent of this project was to determine if a relationship exists between the fractal dimensional increment, surface roughness, and fracture toughness for a unfilled resin and a filled composite.

Experimental Procedure

The present study investigated the relatively smooth unfilled resin fracture surface and the fracture surface of a dental composite made of resin and glass filler particles. The materials used in this investigation were specially prepared resin-matrix composites (Bisco Dental Products Inc., Itasca, IL), similar to commercially available visible light cured systems (O'Brien, 1997). The resin was a mixture by weight of 60% Bis-GMA (2,2 bis[4(2-hydroxy-3-methacryloloxypropyloxy)-phenyl] propane) and 40% TEGMA (triethylene glycol dimethacrylate). The inorganic phase (filler) was strontium glass (small particle 1-8 μm) with colloidal silica (microfill size 0.04 μm) in a ratio of 9 (small particle) to 1 (microfill). The glass filler was silanated by the manufacturer. The dental composite was 25% resin and 75% filler by weight. Volume fraction was not determined for these composites, but based on similar commercial products would be about 64%. The composite and unfilled resin specimens were fabricated in bars (4 mm x 2 mm x 70 mm), cured in a light curing oven (Triad II, Dentsply/York Division, York, PA) for 2 minutes on each side, and hand ground on 120 and 240 SiC grit papers. A sandwich technique was employed to fabricate the samples such that no force was required to remove the cured materials. The split mold was placed on a Plexiglas slab with a strip of mylar covering the specimen, the mold filled with the composite, a second mylar strip placed on the other side of the composite, a second Plexiglas slab placed on top, the mold tightened, and then cured. The specimens were aged in sealed polyethylene containers with 500 ml of deionized, distilled for up to 12 months at 37°C. The specimens aged in air at 37°C were also sealed in polyethylene containers. The control specimens were not aged, but were either fractured in air or in water.

A second group of specimens was subjected to further processing after the initial light curing: (1) heating to 125°C under 0.6 MPa in water (CW) (Concept's System, Williams Dental Division of Ivoclar North America, Amherst, NY), and (2) heating in air in an oven at 125°C (CA). Both treatments were held at the prescribed conditions for 30 minutes with the entire cycle of heating and cooling requiring 60 minutes. The additional processing was not conducted until 2-4 weeks after specimen preparation, but these specimens were subjected to the same aging conditions as the original specimens (C). The 2-4 week delay in processing was due to time constraints required to fabricate all of these specimens to be able to have all specimens receive the same exact post-processing treatment.

Mode I fracture toughness tests were performed on single edge notched specimens (prismatic bars with dimensions 2 mm x 4 mm x 70 mm) in four point loading with a lower span length of 50 mm and an upper span length of 16 mm. A detailed explanation of the specimen preparation is provided in Drummond *et al.* (1995) in which it was demonstrated that for these bar specimens with a 60° V-notch, there was no variation in the fracture toughness for the notch depth ratio, *a*/H, between 0.1 to 0.5. A 60° V-notch of 0.5 mm in depth was machined at the mid-span of the bars. All experiments were conducted on a screw-driven testing system (MTS, Minneapolis, MN) controlled by digital electronics (Instron Corporation, Canton, MA) at room temperature (21°C) in laboratory air. A 100 N load cell (Instron) was used to maximize the sensitivity of the outputs. Mode I fracture toughness, K_{1c}, was evaluated with the use of the fracture mechanics formula for three point bend specimens:

$$K_{1c} = \{PLf_1\ (a/H)\} / (BH^{1.5}) \qquad (1)$$

Here P is the load at specimen fracture, L is the support span (50 mm), B is the specimen thickness, H is the specimen height, and *a* is the notch depth. The function is a correction factor appropriate to the specimen geometry (Gross and Strawley, 1965). The number of specimens for the fracture toughness was 12-25 per variable.

The specimens used for the fractal dimension and surface roughness determination were taken from a more extensive study on the characterization of fracture properties of dental composites (Drummond *et al.*, 1995). The AFM experiment was performed with an ARIS-3300 Personal Atomic Force Microscope (Burleigh Instruments, Inc., Fisher, NY). Line scans of 1.4 μm for the dental resin and composite across the fracture surface were conducted on each specimen to determine the surface roughness, and scans of 1.4 μm x 1.4 μm were utilized to determine the fractal dimension. Due to the roughness of the composite surface and the limitations of the AFM, the scans were limited to 1.4 μm x 1.4 μm. The fractal dimension was determined using the Hurst fractal surface analysis program of Russ (1994). This approach determines the largest difference between the height values on the surface as a function of their separation distance and direction. These data are then used to measure the slope of the Hurst plot (maximum distance versus distance) as a function of direction and shows the result as a plot which gives the fractal dimension. The fractal dimension was only determined from the AFM scans. All scans, 6-8 per specimen, were made adjacent to the tip of the V-notch of the bars. The fractal dimensional increment (D^*) is the fractal dimension (D) minus 2; i.e., $D^* = D - 2$.

A second determination of surface roughness at a larger scale, millimeters versus micrometers, was conducted using a Surftest 201 surface analyzer (Mitutoyo

Table 1. Measured values of fractal dimension (D; dimensionless), surface roughness (R_a), and fracture toughness (K_{Ic})

	N	D	R_a (nm) AFM	R_a (μm) Surface Analyzer	(K_{Ic}) (MPa-$m^{0.5}$)
Resin					
0 m control air	6	2.27 ± 0.08[a]	9.3 ± 2.9[a]	0.25 ± 0.17[b]	1.0 ± 0.2[b]
6 m air	6	2.24 ± 0.07[a]	11.1 ± 1.8[a]	0.13 ± 0.15[b]	1.0 ± 0.1[b]
12 m air	8	2.20 ± 0.11[a]	6.9 ± 1.8[b]	0.18 ± 0.04[b]	0.9 ± 0.1[b]
0 m control water	8	2.07 ± 0.06[b]	9.3 ± 1.9[a]	0.45 ± 0.23[a,b]	1.2 ± 0.2[a]
6 m water	8	2.15 ± 0.11[a]	6.4 ± 1.1[b]	0.15 ± 0.07[b]	0.7 ± 0.1[c]
12 m water	7	2.18 ± 0.12[a]	9.3 ± 1.8[a]	0.70 ± 0.38[a]	0.7 ± 0.1[c]
Glass Filled Resin					
0 m control air	6	2.09 ± 0.01[a]	71.2 ± 31.4[a]	2.15 ± 0.19[a,b]	1.4 ± 0.1[b]
12 m air	6	2.11 ± 0.02[a]	78.5 ± 27.2[a]	2.41 ± 0.28[a,b]	1.5 ± 0.1[a]
12 m air (CW)	6	2.11 ± 0.02[a]	38.4 ± 16.0[a]	2.59 ± 0.34[a]	1.6 ± 0.1[a]
12 m air (CA)	7	2.09 ± 0.02[a]	16.0 ± 8.0[a]	1.67 ± 0.09[b]	1.6 ± 0.1[a]
0 m control water	6	2.09 ± 0.01[a]	42.4 ± 10.5[a]	2.33 ± 0.24[a,b]	1.5 ± 0.1[a]
12 m water	6	2.11 ± 0.03[a]	45.0 ± 13.9[a]	2.14 ± 0.25[a,b]	1.2 ± 0.2[c]
12 m water (CW)	6	2.11 ± 0.02[a]	95.5 ± 53.5[a]	1.99 ± 0.30[a,b]	1.1 ± 0.1[c]
12 m water (CA)	6	2.10 ± 0.02[a]	54.5 ± 19.4[a]	2.52 ± 0.49[a]	1.2 ± 0.1[c]

Mean values with the same letter were not statistically significant at the 0.05 level; $D^* = D - 2$

Manufacturing Company, Ltd., Tokyo, Japan). The scans, 6-8 per specimen, were conducted over a 0.25 mm length from the tip of the V-notch inward. Statistical analysis consisted of a one-way analysis of variance followed by a multiple means comparison, Tukey analysis (Wilkinson, 1989) when needed. The fracture surfaces of the specimen were coated with gold-palladium and observed at an accelerating voltage of 15 kV using a JOEL 35C scanning electron microscope (JOEL, USA, Peabody, MA).

Results and Discussion

The data presented here are from a preliminary investigation of a possible relationship between fractal dimensional, surface roughness, and fracture toughness. No attempt was made to reproduce the fractal dimension other than to take 6-8 scans of the fracture surface. All scans were taken in essentially the same position on the fracture surface, i.e., adjacent to the notch in the middle of the specimen. The exact location of the AFM scanning tip was not known, such that, the scans could have been on either of the glass filler particles, the resin, or the resin coating the glass fillers. The extreme roughness of the fracture surface in the z-direction of the filled composite resin prevented the determination of the fractal dimension at different magnitude of scale. The data obtained from the unfilled resin and composite specimens for the different aging times and processing are given in Table 1. Graphical representation of the fractal dimensional increment versus the fracture toughness is shown in Figure 1 and the fracture toughness versus the surface roughness is presented in Figures 2 and 3. Even though the fracture toughness values showed a significant difference, and significant differences were obtained within the fractal dimension increment

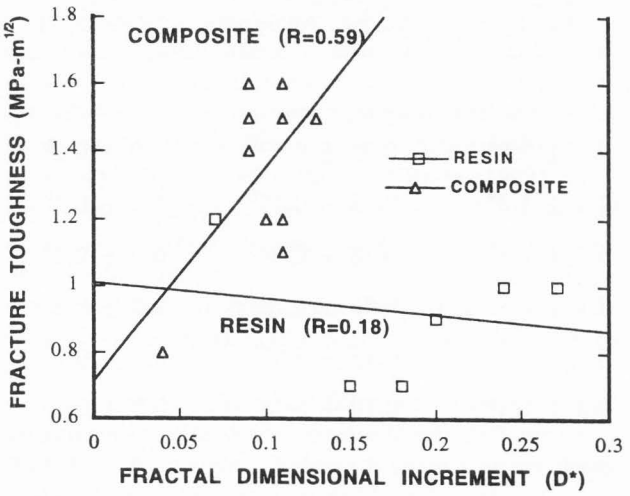


Figure 1. Fracture toughness versus fractal dimensional increment.

---

and surface roughness, no consistent correlation was observed between fracture toughness and either surface roughness or fractal dimensional increment. The values for correlation coefficients, R, are presented on Figures 1, 2 and 3. The glass filled resin had a higher surface roughness and fracture toughness than the unfilled resin, but the fractal dimensional increment showed the opposite effect. This increase in fracture toughness and surface roughness with the addition of a glass filler is expected since it requires more energy to fracture the glass filled resin. The surface roughness showed a scale effect, that is, the larger the size of the scan, the higher the surface roughness. This correlation with scan size has been previously observed (Majumdar and Bhushan, 1995).

The scanning electron micrographs of fracture surfaces from resin and glass filled composite are shown in Figures 4 and 5, respectively. As can be observed, the resin surface is smoother than the glass filled composite. The addition of the filler not only improves the mechanical properties of the dental composite, but increases the fracture surface roughness.

Possible explanations for the lack of correlation between the fracture toughness and the fractal dimensional increment for the atomic force microscopy analysis is that the scale of the scans is less than that of the glass particles which have a significant influence on the fracture toughness. Also, the fractal algorithm may be inappropriate for this material. Other studies that have

FIGURE 2: FRACTURE TOUGHNESS VERSUS ROUGHNESS FOR RESIN

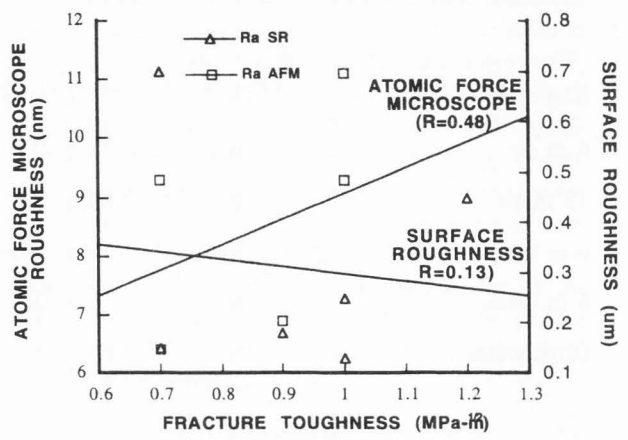
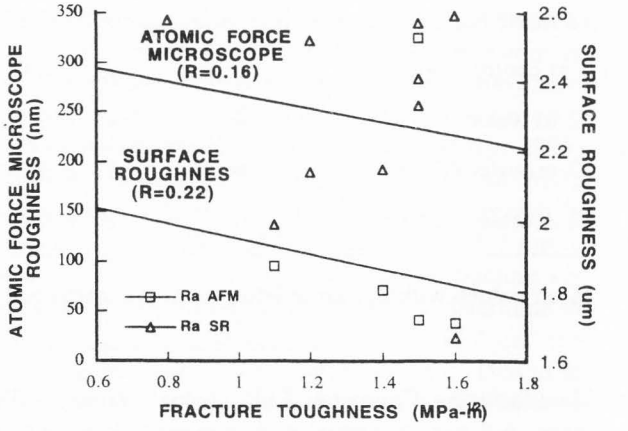


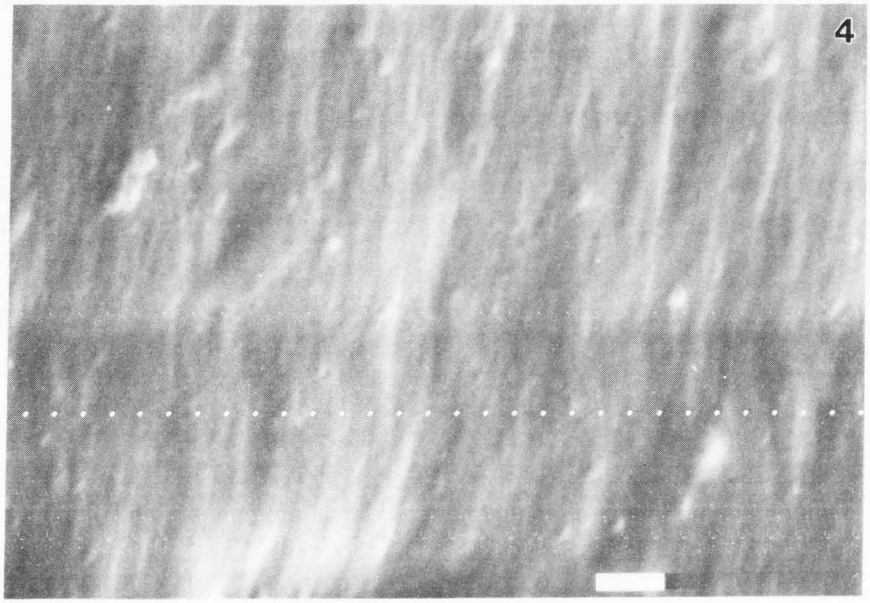
FIGURE 3: FRACTURE TOUGHNESS VERSUS ROUGHNESS FOR COMPOSITE

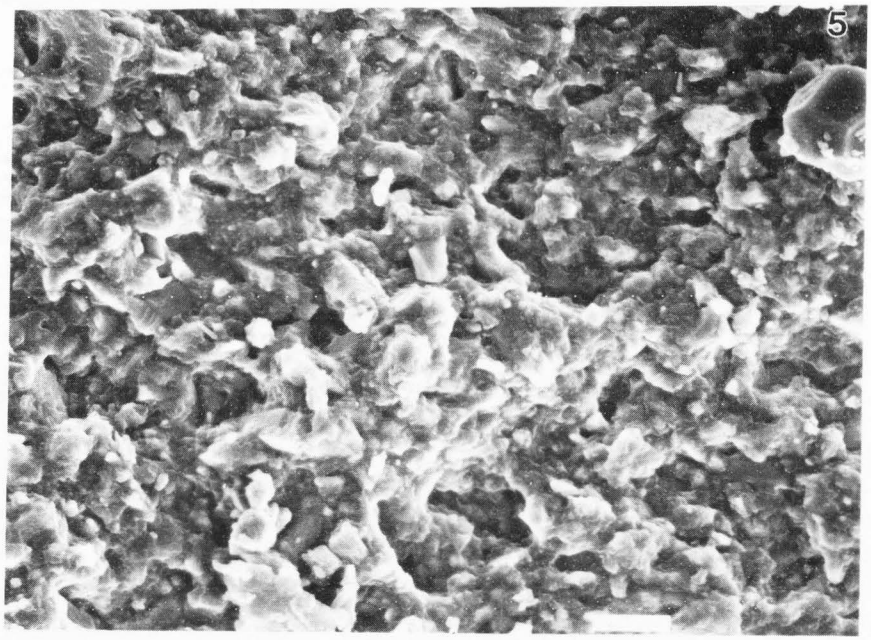


Figures 2 and 3. Fracture toughness versus surface roughness of resin (**Fig. 2, top**) and composite (**Fig. 3, bottom**).

---

demonstrated a correlation between fracture toughness and fractal dimension have all used the slit edge technique to determine the fractal dimension. Even though these materials are similar to the glass ceramics discussed in the **Introduction**, the glass ceramic materials did not have a resin interface or coating on or between the ceramic particles and maybe this approach is not appropriate for resin-ceramic materials, but only for ceramic-ceramic materials.





Figures 4 and 5. Scanning electron micrographs of resin (**Fig. 4, top**) and composite (**Fig. 5, bottom**) fracture surfaces. Bars = 10 μm.

Even though several investigators have shown that fracture toughness correlates with the fractal dimensional increment, this correlation is mainly empirical. It would be possible to obtain either a positive or a negative correlation. For example, if one takes the case of a composite where the reinforcement is random inclusions, a positive correlation between fracture toughness and fractal dimension would be obtained. If one takes a material with random voids, then a negative correlation would be obtained. On the other hand, the fractal dimension increases in both cases with the increase in volume fraction of the inclusions and voids (Duxbury, 1990). Most likely, the relationship of a positive correlation between fracture toughness and the fractal dimensional increment is related to the specific material and is not a general trend.

Conclusions

Although for some glass ceramics a correlation has been observed between fracture toughness and the fractal dimensional increment of the fractal dimension, no correlation was observed in this study on a resin and a brittle dental composite.

Acknowledgments

The authors wish to thank Christopher Zuiker and Alan Krauss at Argonne National Laboratory for assistance with the atomic force microscope and the Electron Microscope Facilities at the Research Resource Center at the University of Illinois at Chicago for the scanning electron micrographs. This project was supported by grant RO1-DE07979 from National Institute of Dental Research.

References

Anusavice KJ, Zhang N-H (1995) Effect of crystallinity on strength and fracture toughness of lithia-based glass-ceramics. J Dent Res (Spec Issue #1656) **74**: 218 (abstract).

Baran GC, Roques-Carmes C, Wehbi D, Degrange M (1992) Fractal characteristics of fracture surfaces. J Am Ceram Soc **75**: 2687-2691.

Drummond JL, Botsis J, Zhao D (1995) Fracture mechanics of dental composites. In: Encyclopedia of Biomaterials and Bioengineering. Marcel Dekker, New York. pp. 2813-2834 (Chap. 56).

Duxbury PM (1990) Breakdown of diluted and hierarchical systems. In: Statistical Models for Fracture of Disordered Media. Herrmann HJ, Roux S (Eds.). Elsevier Science Publishers, Amsterdam. pp. 189-228.

Gross B, Strawley JE (1965) Stress intensity factors for single-edge notch specimens in bending and tension by boundary collocation of a stress function. NASA Lewis Research Center (Cleveland, Ohio), Technical Note D2603.

Hill TJ, Mecholsky JJ, Anusavice KJ (1995) Fracture toughness and fractal dimension versus crystallization for baria-silicate glass-ceramics. J Dent Res (Spec Issue #1657) **74**: 219 (abstract).

Lankford J, Longmire M (1991) Application of scanning tunneling microscopy to fatigue and fracture, J Mater Sci **26**: 1131-1136.

Majumdar A, Bhushan B (1995) Characterization and modeling of surface roughness and contact mechanics. In: Handbook of Micro/Nano Tribology. Bhushan B (ed.). CRC Press, Boca Raton, FL. pp. 109-165.

Mandelbrot BB (1983) The Fractal Geometry of Nature. W.H. Freeman, San Francisco, CA. pp. 1-34.

Mandelbrot BB, Passoja DE, Paullay AJ (1984) Fractal character of fracture surfaces of metals. Nature (London) **308**: 721-722.

Mecholsky JJ, Passoja DE, Feinberg-Ringel KS (1989) Quantitative analysis of brittle fracture surfaces using fractal geometry. J Am Ceram Soc **72**: 60-65.

Mecholsky JJ (1991) Quantitative Fractography: An Assessment in Fractography of Glasses and Ceramics. Amer. Ceramic Society, Columbus, OH. pp. 413-451.

Mecholsky JJ, Plaia JR (1992) Fractal analysis on fracture surfaces of glass using replication techniques. J Non-Cryst Solids **146**: 249-255.

Mitchell MW, Bonnell DA (1990) Quantitative topographic analysis of fractal surfaces by scanning tunneling microscopy. J Mater Res **5**: 2244-2254.

Naman A, Anusavice KJ, Mecholsky JJ (1994) Fractal analysis of lithia-based glass-ceramics. J Dent Res (Spec Issue #755) **73**: 196 (abstract).

O'Brien WJ (Ed.) (1997). Dental Materials and Their Selection, 2nd ed. Quintessence Publishing Co., Carol Stream, IL. pp. 97-114.

Russ JC (1994) Measurement of Fractal Dimensions in Fractal Surfaces. Plenum Press, New York. pp. 89-97.

Thompson JY, Anusavice KJ, Balasubramaniam B (1995) Effect of microcracking on the fracture toughness and fracture surface fractal dimension of lithia-based glass-ceramics. J Am Ceram Soc **78**: 3045-3049.

Wilkinson L (1989). SYSTAT: The System for Statistics. Systat, Inc., Evanston, IL. pp. 446-471.

Discussion with Reviewers

Reviewer V: It appears that not enough representative regions were taken on the AFM scans. What is the number of points per line? How many lines per scan? The area of the scan was 1.4 μm x 1.4 μm. Thus, the AFM may not have measured the appropriate area for a proper fractal dimension measurement. It is important to make fractal dimension measurements across several orders of magnitude. The range selected may be in between the large deviations, which are obvious from the scanning electron micrographs, and the very fine deviations of the molecules. Thus, it is most likely the fractal dimension obtained from the limited scan range is not representative of the material. Just judging from the scanning electron micrographs in Figures 4 and 5, the fracture surface shown in Figure 5 should have a larger fractal dimension than that shown in Figure 4. The fact that it does not indicates a problem with the measurement technique using the AFM.

Author: There were 200 lines per scan. With regards to whether the appropriate area was measured to make the fractal dimension measurement, approximately the

same area was examined in relation to the notch machined in each of the specimens. The roughness of the glass filled composite made it impossible to increase the scan area.

Reviewer V: The explanations offered for possible differences in the author's findings and several other investigations are not well presented. The first is the scale of the scans. This is probably the most likely explanation, but not because the scan is less than the glass particles. If enough scans were taken, the particles would have been sampled unless the entire fracture surface is in the resin. Is it? In any case, if enough of the surface is sampled, a representative value would have been obtained. If the author really thinks that this may be a possible explanation for the differences, why were larger scans not taken? Regarding the argument involving random inclusions versus random voids, I assume that the author wishes to imply that random voids would have a lower toughness than random inclusions. However, this is not necessarily the case. Fine porosity dispersed through a matrix can increase fracture toughness because of local deviations of the crack front. So the example they provide means that the toughness could increase with fractal dimension in both cases. Because of this fact, I do not understand their point. Finally, the relationship for "a positive correlation between fracture toughness and the fractal dimensional increment" has been shown for intermetallics, polymers, polycrstalline ceramics, inorganic glasses, and glass ceramics. It is difficult to think that the author has found a brittle material that does not behave in a similar manner. However, it is possible. The author should explain why with better arguments.

Author: The possibility exists that all of the glass particles are covered with resin since seldom in the scanning electron micrographs were fractured glass particles observed. The size of the scan was limited, the roughness of the glass-composite surface prevented any larger scans due to the limited response in the z-direction of the AFM. Our experience with these materials regarding voids always results in a lowering of the fracture toughness and we were exploring reasons pertaining to these glass filled composites. Baran *et al.* (1992) also found no correlation between fractal dimension and fracture toughness.

Reviewer V: The author uses several treatments which can chemically alter the crack tip, but does not address this possibility. Also, it is not mentioned whether the cracks started in the same manner for all of the tests. In other words, if the chemistry at the crack tip was altered by the treatments, then perhaps a sharp crack could not be obtained for some of the conditions and could be obtained for other conditions. Did the author verify that the cracks started in a uniform manner in all tests so that the formulae presented were applicable? If the author argues that the cracks was put in after the treatments so there should be no difference, then it is argued that the material is different because of the treatments and could result in different crack tip geometries. The sharpness of the crack before propagation has a greater impact on the value recorded for toughness.

Author: The specimens were notched before the treatments, but scanning electron microscopy of the fracture surface of the machined notch indicated that the crack appeared to start in the same manner for all specimens.

Reviewer V: The materials treated differently may be, in effect, different materials. Thus, there may not be an unique correlation between the toughness and the fractal dimension. The general relation of toughness increasing with fractal dimension increment would be expected to be valid, but the materials would not necessarily be expected to be on the same line, i.e., have the same slope. With this viewpoint, all of the resin points should not be graphed as one material in Figure 1 (similarly for the composite). If this is the case, the author does not disagree with the literature. Unfortunately, the author does not discuss the effect of the treatments on the structure, strength, residual stress, etc., so the reader does not know what the effect is and whether or not the treatments should be considered different materials.

Author: The comment that the aging altered the materials such that the comparison is on different materials instead of the same material is a mater of debate, but for this study, the materials were assumed to be the same. The effect of treatments is discussed in detail in Drummond *et al.* (1995).

Reviewer V: If the surface is self-affine, then a profile analysis will not result in an accurate representation of the fracture structure. An analysis that cuts the plane would be better. This difference in measurement is discussed in Russ (1994).

Author: The technique employed was what was available. The discussion in Russ (1994) examines the history, controversy, and methods utilized to examine for a possible relationship between fractal geometry and fracture surfaces. This summary presents some of the correlations, both positive and negative, between fractal dimension and fracture properties.

W.A. Brantley: Does the author think that different values of the fractal dimension would have been obtained if the fracture surfaces of the two resins had been analyzed at the range of magnifications customarily used with the SEM to study the microstructures of dental resins?

Author: The fractals dimension values might change if the range of magnification is modified, but also this would entail a different approach to obtain the fractal dimension rather than the atomic force microscope and, most likely, a different program to determine the fractal dimension. All of these changes in analysis could lead to obtaining a different value for the fractal dimension.

W.A. Brantley: What microstructural scale does the author consider most relevant for the fracture processes in these dental resins?

Author: The most relevant microstructural scale for fracture processes would probably be one in which the filler particles are readily observed to determine if the fracture occurs though or around the glass filler particles.