Long-Term Strength of Ceramics for Biomedical Applications

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Abstract: The influence of slow crack growth on the initiation of radial cracks at the lower surfaces of ceramic layers bonded to polymeric substrates is studied, with particular relevance to biomechanical systems, e.g., dental crowns and hip replacement prostheses. Critical loads are measured as a function of loading rate (dynamic fatigue) for model bilayers fabricated by epoxy-bonding selected clinical ceramics to polycarbonate bases. Radial crack initiation is observed *in situ* by viewing from below the transparent base during loading. Declines in the critical loads with diminishing load rate are consistent with slow crack growth of intrinsic flaws prior to radial crack pop in. A simple fracture mechanics relation incorporating a crack velocity function is used to analyze the data. Extrapolation beyond the data range enables long-lifetime (10 yr) estimates of sustainable loads. The procedure provides a basis for ranking ceramic types, and in particular for eliminating vulnerable candidate materials, for use in biomechanical systems. While slow crack growth is an important factor in failure, other mechanisms could operate in concert and even dominate under severe testing conditions, especially under cyclic loading. © 2004 Wiley Periodicals, Inc.* J Biomed Mater Res Part B: Appl Biomater 69B: 166–172, 2004

Keywords: bilayers; brittle coatings; crack velocity; critical loads; dental crowns; lifetime; fracture

INTRODUCTION

Hard coating layers afford mechanical, thermal, and chemical protection to soft underlayers in biomechanical structures (teeth, dental crowns, hip prostheses, crustaceans, and seashells). Such bilayers are subject to fatigue damage at the ceramic top and bottom surfaces from sustained concentrated loads.[1–4] Of the various competing damage modes, radial cracks initiating from flaws at ceramic lower surfaces are especially deleterious because they can spread easily and thereby fracture the ceramic layer. Radial cracks have been identified as a primary source of premature failure in all-ceramic dental crowns.[5–7] These (and other) cracks are enhanced by slow-crack-growth (SCG),[8] generally associated with the chemical influence of water.[9,10] There is an attendant implication that moisture has access to the ceramic lower surfaces at the interface with the underlying soft substrate, either from preexisting water content in the substrate material or by diffusion from the external environment.[6,8]

When ceramic components are used in biomechanical applications, long-term reliability is a major concern.[11] The question arises as to what extent SCG may contribute to failure in clinical structures. In a recent study, Lohbauer et al.[12] conducted stressing rate tests on free-standing plates of two dental ceramics, a feldspathic porcelain and a glass-infiltrated alumina, in four-point flexure. They projected declines in strengths of more than a factor of 2 over a period of 1 year. However, it is not immediately apparent how such results pertain to the fatigue of ceramic/polymer bilayer (sometimes multilayer) configurations representative of dental crowns, total hip replacements (THRs), and other layered clinical structures. Testing on ceramic/polymer bilayers with spherical indenters—in simulation of occlusal function or hip articulation—is closer to the physical reality.[4,7,13] By choosing a transparent polymer material for the substrate, critical loads for initiation of subsurface radial cracks can be measured *in situ* from below during a loading cycle, making for simple data accumulation;[14] and, by varying the loading rates, kinetic factors can be quantified and slow crack growth parameters evaluated.[8] Such a methodology affords a convenient means of screening and ranking ceramics for optimum performance in biomechanical applications.[4,15]

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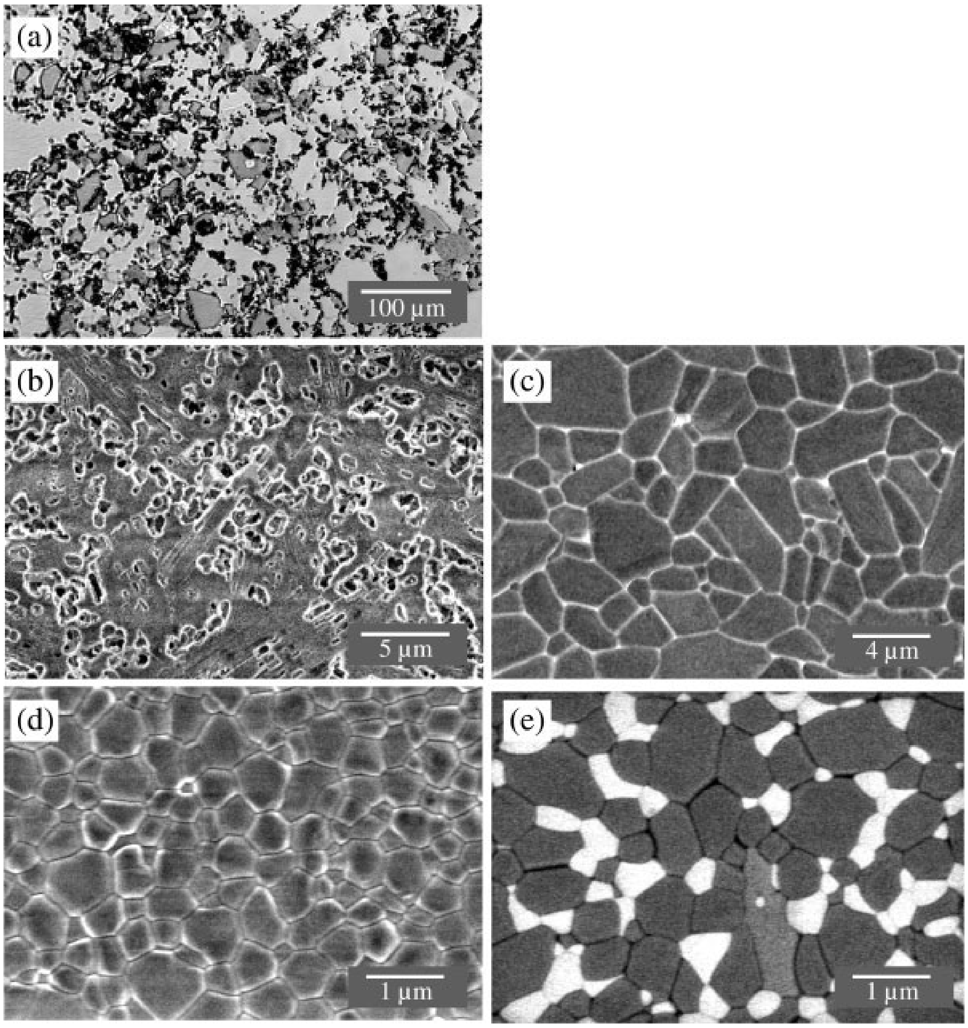

Figure 1. Microstructures of ceramics investigated in this study (see Table I). (a) Fluorospathic leucite porcelain. Acid-etched surface, optical micrograph. Coarse grains are residual crystalline phases in (partly etched) silica glass matrix, with porosity. (b) Glass-ceramic. Acid-etched surface, scanning electron micrograph (SEM). Elongate features are lithium disilicate grains in glassy matrix. (c) Alumina (Al_2O_3). Thermally etched surface, SEM. Structure is 99.5 wt.% pure, equiaxed, and fine grained. (d) Yttria-stabilized zirconia (Y-TZP). Thermally etched surface, SEM. (e) Alumina-matrix composite. Thermally etched surface, SEM. Equiaxed, fine-grained alumina matrix with zirconia inclusions (light phase).

In this article, results of Hertzian contact tests on flat-layer ceramic/polymer bilayers are presented. Five clinically relevant ceramic materials are evaluated: porcelain, glass ceramic, fine-grain alumina, yttria-stabilized zirconia, and alumina-matrix composite. Critical loads to produce radial cracking at the ceramic lower surfaces are measured as a function of fixed loading rates (dynamic fatigue). The resulting data are analyzed in terms of a slow-crack-growth model, and crack velocity exponents thereby determined. Fitted dynamic fatigue relations are used to extrapolate the data to long-term (1 year or 10 year) operational conditions. Predictions for bilayer configurations representative of ceramic layers on dentin (crown) and ceramic liners in acetabular cups (THRs) are used to evaluate the survivability of each material type. The potential for fatigue mechanisms other than slow crack growth is discussed.

MATERIALS AND METHODS

Materials and Testing

Clinically relevant ceramics covering a broad range of properties were selected for study as coating materials for bilayers: a dental veneering fluoroapatite porcelain (brand name IPS d.Sign, Ivoclar-Vivadent, Schaan, Leichtenstein); a lithium disilicate glass-ceramic used in all-ceramic dental restorations (Empress II, Ivoclar-Vivadent, Schaan, Leichtenstein);[16] a dense, 99.5% pure, fine-grain alumina (AD995, CoorsTek, Golden, CO) representative of a wide range of crown and THR aluminas; a 3 mol% yttria-stabilized zirconia (Prozyr Y-TZP, Norton, East Granby, CT) and an alumina-matrix composite with 25 vol% zirconia (AMC DC-25, CeramTec, Plochingen, Germany) as ultra-strong ceramics for dental and hip applications. Micrographs of these ceramics are shown in Figure 1. Note the relatively fine, homogeneous, and equiaxed structures of the Y-TZP and AMC. Clear polycarbonate (Hyzod, AlN Plastics, Norfolk, VA) was chosen as a model substrate material, representative of tooth dentin and ultra-high-molecular-weight (UHMW) polyethylene liners in acetabular cups. Pertinent properties of these materials are listed in Table I.[15]

Flat-layer bilayer specimens were fabricated according to procedures described in a previous study.[15] Ceramic plates with minimum surface dimension 25 × 25 mm were ground and polished (1 μm surface finish) to thicknesses $d = 1$ mm or less. Radial cracks tend to be the dominant mode of damage in the submillimeter thickness range. The plates were bonded by epoxy resin (Harcos Chemicals, Bellesville, NJ) onto polycarbonate substrates 12.5 mm thick, with resultant adhesive layers ≈10 μm. The thickness of the epoxy resin interlayer is not crucial in the present study, because the elastic modulus of epoxy resin is similar to that of the polycarbonate base (Table I).

The ceramic/polycarbonate bilayers were loaded at their top surfaces with a tungsten carbide (WC) indenting sphere of radius 3.18 mm mounted into the cross head of a screw-driven Instron machine (Model 5500R, Instron Corp., Canton, MA). Loading was increased monotonically at prescribed constant rates, over a range $\dot{P} = dP/dt = 0.01$ to 100 N s^{-1}, with load data recorded electronically at intervals of 0.002 s. The initiation of radial cracking in the lower surfaces of the ceramic plates was monitored from below the contact through the transparent polycarbonate and adhesive using a video camcorder (Canon XL1, Canon, Lake Success, NY) equipped with a microscope zoom system with on-screen magnification 10 × (Optem, Santa Clara, CA). These observations enabled direct measurement of the critical loads P_R to radial fracture and corresponding test durations t_R. Load drops in the $P(t)$ responses (typically 5% of P_R) proved a useful adjunct method at faster loading rates.

TABLE I. Properties of Materials Used in Current Study

Material	Modulus E (GPa)	Strength σ (MPa)	Crack Velocity Exponent (N)
Coating			
Zirconia (Y-TZP)	205	1400	25 ± 2
Alumina-matrix composite (AMC)	350	1150	54 ± 9
Alumina (AD995)	372	572	26 ± 6
Glass-ceramic (lithium disilicate)	104	320	20 ± 3
Porcelain	68	101	17 ± 2
Glass	73	110	16 ± 1
Adhesive/substrate			
Epoxy resin	3.5	–	
Polycarbonate	2.3	–	
Tooth dentin	16	–	
Polyethylene	2	–	

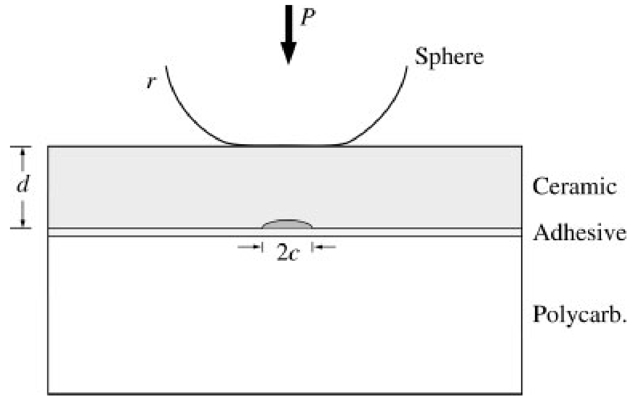

Figure 2. Schematic of radial crack of characteristic dimension c at lower surface of brittle layer of thickness d on compliant substrate, loaded at top surface with sphere of radius r at load P. Other potential damage modes not shown.

Fracture Mechanics

Consider bilayers consisting of a ceramic layer of thickness d and Young's modulus E_c bonded onto a thick complaint substrate of modulus E_s, loaded at the top surface with a concentrated force P, as in Figure 2. The load induces a flexural tensile stress σ at the center of the coating lower surface,[14,17]

$$\sigma = (P/Bd^2) \log (E_c/E_s) \quad (1)$$

within the limits of linear elasticity, where $B = 1.35$.[18,19] Radial cracking pops in from a dominant starting flaw in the ceramic at or close to the central location of maximum tension.

The starting flaw is subject to slow crack growth prior to instability, generally enhanced by applied stress intensity and intrusion of water molecules from the immediate environment into the flaw. This growth can be expressed by a crack velocity relation $v \sim K^N$,[20] where N is a characteristic exponent and $K \sim \sigma c^{1/2}$ is a stress-intensity factor for a crack of length c under tensile stress σ.[21] Combining these basic relations along with the dynamic fatigue condition $P = \dot{P}t$ in Eq. (1) and integrating c between initial and final (instability) lengths and t between 0 and t_R yields

$$P_R/d^2 = [A'(N+1)\dot{P}/d^2]^{1/(N+1)}, \quad (2)$$

where A' is a load-, time-, and thickness-independent quantity.[8] Note the d^2 dependence in the load terms. Or, alternatively,

$$P_R/P_0 = (t_0/t_R)^{1/N}, \quad (3)$$

where P_0 and t_0 are reference parameters relating to nominal short-term tests.

Other modes of damage that might contribute to fatigue will be considered in the discussion section.

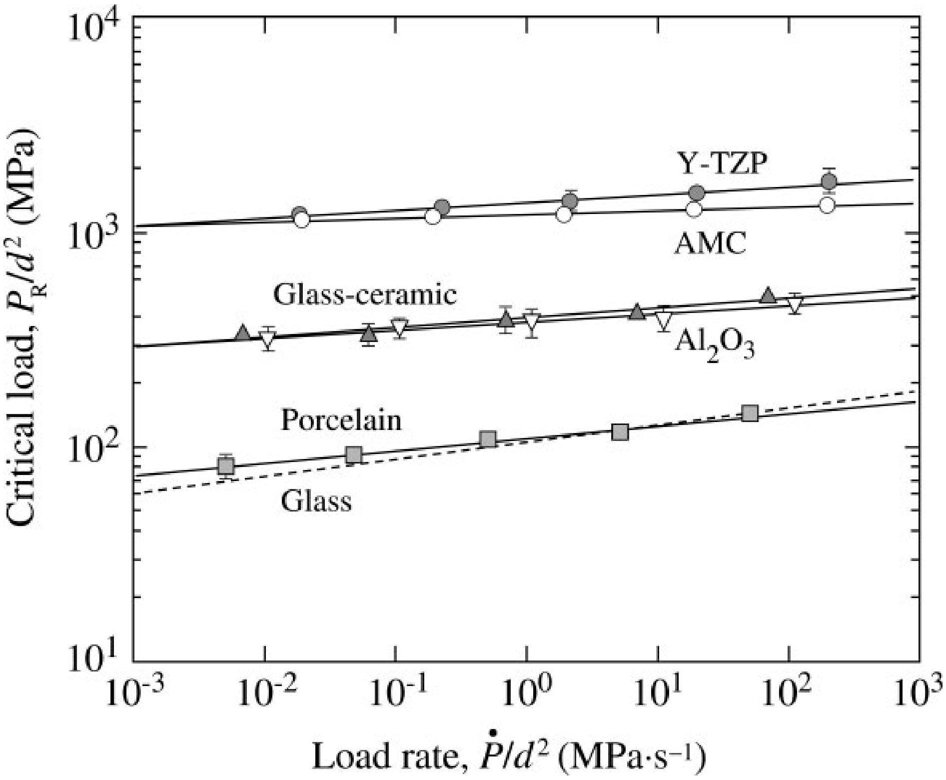

Figure 3. Contact load versus loading rate for selected ceramic layers epoxy-bonded to polycarbonate substrates. Ordinates are normalized to d^2 to accommodate data from specimens of different ceramic thickness. Data are means and standard deviations for onset of radial cracking at ceramic lower surface, minimum five tests (error bars smaller than symbols in some cases). Solid lines are logarithmic regression fits to raw data, in accordance with slow-crack-growth analysis. Dashed line represents data fit for glass from an earlier study.[8]

RESULTS AND ANALYSIS

Figure 3 shows results of the dynamic fatigue tests as P_R/d^2 versus $\dot{P}/d^2$ in logarithmic coordinates. Normalization to d^2 enables direct comparison of critical loads for ceramic coatings of different d, effectively reducing all data to one nominal thickness. Data are means and standard deviations for a minimum of five tests at each point for the ceramics listed in Table I. Solid lines are regression power-law fits to Eq. (2). The dashed line is a reference baseline for (surface-abraded) soda-lime glass from an earlier study.[8] The critical loads P_R at any prescribed loading rate $\dot{P}$ and layer thickness d differ by more than an order of magnitude between porcelain or glass (weakest) and AMC or Y-TZP (strongest), with alumina and lithium disilicate glass-ceramic intermediate. These material groupings correlate primarily with bulk strengths (Table I). Exponents N evaluated from the (inverse) slopes of the fitted lines vary from $N = 16$ (most susceptible) for glass or porcelain to 54 for AMC (least susceptible) (Table I). It is not practicable to make direct comparisons with literature values of N from independent crack velocity measurements for each material type because of variabilities in material characteristics (grain size, composition, porosity) and test procedures. Although the range of values is typical of glasses and polycrystalline ceramics, there is evidence from a preceding study that the N values from bilayer tests may slightly underestimate true crack velocity exponents because of contributions to the fatigue response from other dissipative processes; for example, creep in the substrate or adhesive layers.[8]

Figure 4 replots the critical loads in Figure 3 as a function of test duration t_R, using $t_R = P_R/\dot{P}$ to convert the load-rate data. In this plot the data are plotted without error bars, for clarity. The solid lines are regression fits to the raw data in accordance with Eq. (3), with 95% confidence bounds. Note that the confidence bounds are relatively tight for the two strongest ceramics, Y-TZP and AMC, consistent with the more homogeneous grain structures in these materials (Figure 1). Extrapolations of the regression lines to 1 and 10 years enable estimates of long-term lifetimes for any given contact load and ceramic layer thickness. The falloff in sustainable loads amounts to a factor of 2–4 over this time period, depending on N values—highest falloffs for glass and porcelain, lowest for AMC.

Figure 5 is a replot of the data in Figure 4 but with effective strength S replacing critical load P_R on the ordinate, obtained by substituting $\sigma = S$ at $P = P_R$ into Eq. (1). The term *effective* is used because values of S from bilayer tests do not always correspond absolutely to bulk strengths from flexure tests, for a variety of reasons to be discussed below.[19] The groupings of the materials are similar to those in Figure

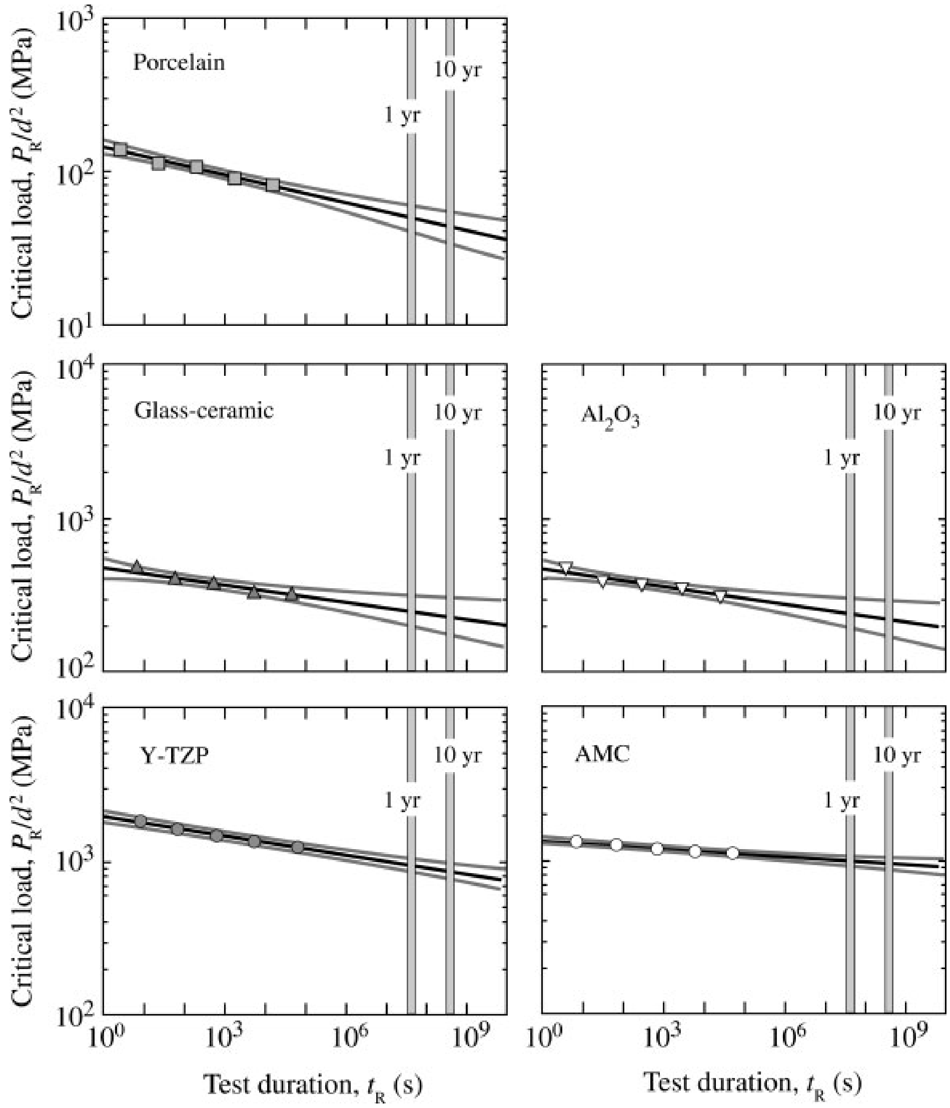

Figure 4. Replot of data in Figure 3 as load for radial cracking versus test duration. Data points are means. Solid lines are regression best fits; shaded bands are 95% confidence bounds.

3, except for small relative shifts in the data associated with the E_c modulus dependence in Eq. (1).

DISCUSSION AND CONCLUSIONS

The results presented in this study confirm the susceptibility of ceramic layers on compliant substrates to degradation by slow crack growth from sustained concentrated loading. The focus has been on one particularly deleterious fracture mode, radial cracking at the ceramic lower surfaces. It is implied that water has access to the ceramic/substrate interface in the bilayer configuration.[8] A fracture mechanics analysis incorporating a crack velocity equation provides the basis for quantifying the rate dependence of the critical loads for radial fracture, and for ranking different ceramic types for use in bilayer structures. Thus in Figure 3 the dynamic fatigue data for ceramic/polycarbonate bilayers fall into three distinct groups: low-strength ceramics, porcelain and soda-lime glass, with relatively strong rate dependence (low N); intermediate-strength ceramics, alumina and lithium disilicate glass-ceramic, with moderate rate dependence; and high-strength ceramics, Y-TZP and AMC, with relatively low rate dependence in the case of AMC. By replotting the critical load data as a function of test duration in Figure 4, direct extrapolations can be made to estimate potential lifetimes at any given loading level. Typically, the capacity for the ceramics to sustain prolonged loads diminishes by a factor of 2–4 over a period of years.

The data in Figures 3 and 4 pertain specifically to radial cracking in ceramic layers of thickness $d \sim 1$ mm on polycarbonate substrates. With fits to these data used as reference baselines, Eqs. (1)–(3) can be used to predict critical loads for prospective biomechanical systems with different ceramic thicknesses and substrate materials, over extended operational durations. Figure 6 shows critical loads $P_R = P_{10}$ at

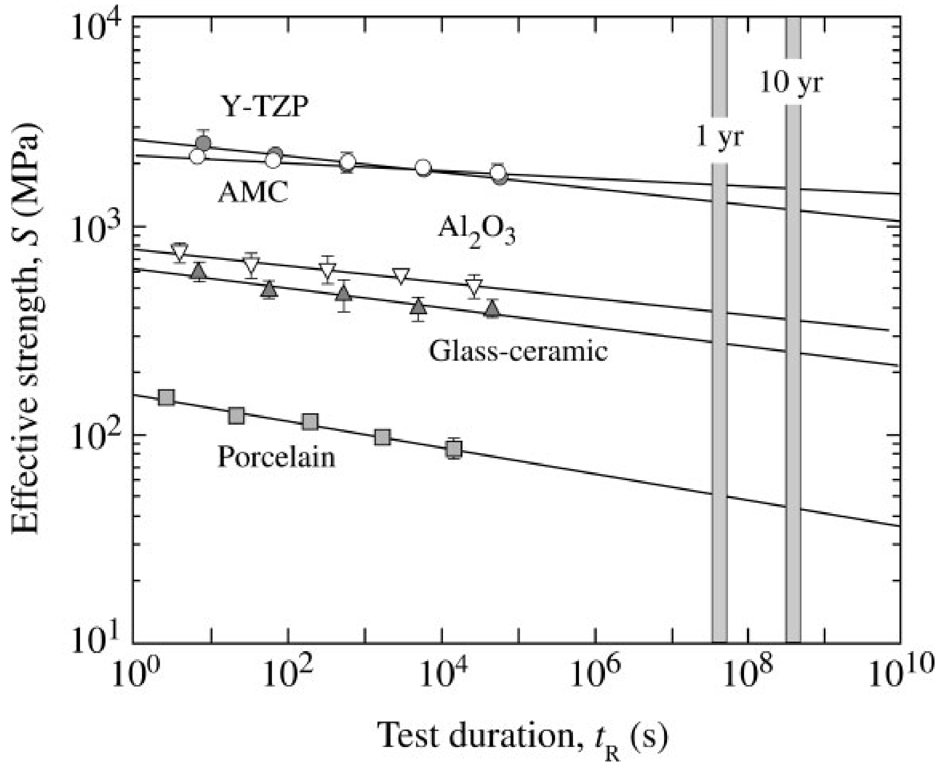

Figure 5. Replot of data in Figure 4 as strength versus test duration, with Eq. (1) to convert critical load data. Data points are means and standard deviations.

$t_R = t_{10} = 10$ yr for two such systems: (a) ceramic dental crowns on natural dentin substrates, thickness $d \sim 1.5$ mm and modulus $E_s \sim 16$ GPa; (b) ceramics liners for THR acetabular cups with UHMW polyethylene backing, thickness $d \sim 5$ mm and modulus $E_s = 2.0$ GPa. The bars indicate means and 95% confidence bounds. Shaded areas in this figure indicate extreme ranges of operational forces, $P = 0\text{–}400$ N for crowns and $P = 0\text{–}5$ kN for THRs. Clinically, it is necessary to ensure that the values of P_{10} remain above the shaded regions. These computations ignore any curvature of the ceramic surface in the clinical systems, but nevertheless indicate which materials are most likely to survive. Porcelains are clearly vulnerable in monolithic form. Alumina and lithium disilicate glass-ceramic are in the intermediate range, and may be vulnerable to occasional load spikes or to other fatigue mechanisms (see below). Y-TZP and AMC would appear to be comparatively immune to radial cracking. However, Y-TZP is suspect for reasons of chemical and thermal instabilities, specifically from moisture- and heat-induced phase transformations, leaving AMC and other strong materials as preferable candidates.[11] Noting the dependence $P_R \propto Sd^2$ at $\sigma = S$ in Eq. (1), it is crucial to maintain a minimum nominal thicknesses d in each of the clinical systems as well as high strengths.

It should be reiterated that the current analysis assumes radial cracking to be the dominant mode of failure in the bilayers of interest. Radial cracking is most likely for hard ceramics with thin layers ($d < 1$ mm) and characteristically blunt contacts ($r > 4$ mm).[15] As mentioned earlier, this has

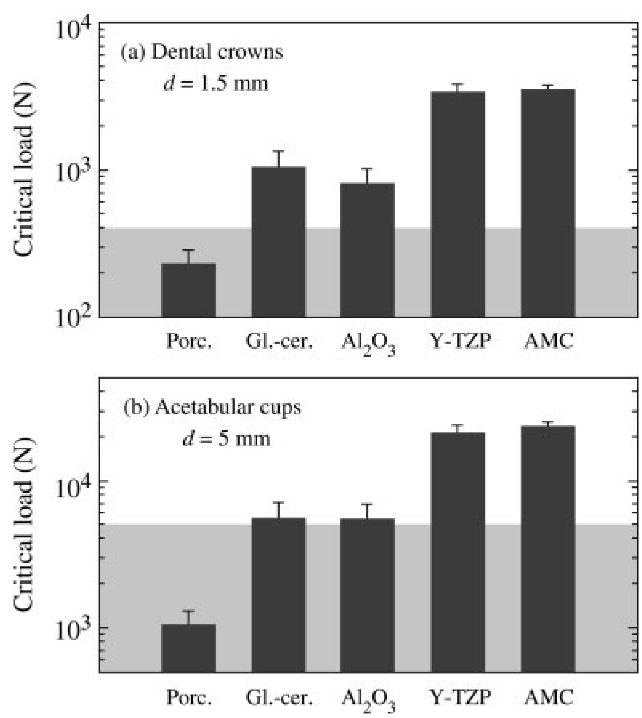

Figure 6. Bar chart showing predicted critical loads for radial cracking in bilayers for two biomechanical applications: (a) ceramic/dentin for dental crowns, $d = 1.5$ mm; (b) ceramic/polyethylene for ceramic liners in acetabular cups, $d = 5$ mm. Data computed from Eq. (2) at sustained load for $t_R = 10$ yr. Error bars are 95% confidence limits. Shaded areas indicate typical operational loads in biomechanical function. Note different scales in (a) and (b).

been identified as the primary mode of failure in all-ceramic dental crowns.[5,6] Other damage modes may operate in certain instances: in thick ceramic layers and sharp contacts, top-surface damage in the form of cone cracking or quasiplasticity in the near-contact region;[22] in softer materials, subsurface damage—quasiplasticity at the ceramic bottom surface or viscoelasticity in the near-interface region of the substrate.[8,19] In actual biomechanical structures, chipping at the specimen edges or margins can also become a factor. All of these modes are similarly susceptible to rate effects. Quasiplasticity modes are particularly deleterious in cyclic loading (mechanical fatigue),[23,24] and warrant further attention in some material systems.

Analysis of the critical load data in Figures 3 and 4 enable evaluation of controlling material parameters, strength S (Figure 5) and crack velocity exponent N (Table I). Allusion has been made to the fact that these quantities may not always correspond exactly to values from measurements on free-standing bulk ceramic specimens. Such discrepancies can arise from several factors: flaw statistics, by restricting the availability of large flaws within the localized tensile region at the ceramic lower surfaces;[25] residual intralayer stresses developed during fabrication and function;[26] inapplicability of Eq. (1) associated with departures from strict point-force loading in the (Hertzian) contact at the top surface;[19] and nonlinear effects associated with quasiplasticity or viscoelasticity in the material components. Nonetheless, the data provide a useful guide to relative strengths, and are in any case closer to the bilayer configurations that characterize actual crown and hip prosthesis applications.

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REFERENCES

1. Jung YG, Wuttiphan S, Peterson IM, Lawn BR. Damage modes in dental layer structures. J Dent Res 1999;78:887–897.
2. Lawn BR et al. Damage-resistant brittle coatings. Adv Eng Mater 2000;2:745–748.
3. Lawn BR. Ceramic-based layer structures for biomechanical applications. Current Opinion Solid State Mater Sci 2002;6: 229-235.
4. Lawn BR, Deng Y, Miranda P, Pajares A, Chai H, Kim DK. Overview: Damage in brittle layer structures from concentrated loads. J Mater Res 2002;17:3019–3036.
5. Kelly JR. Ceramics in restorative and prosthetic dentistry. Annu Rev Mater Sci 1997;27:443–468.
6. Kelly JR. Clinically relevant approach to failure testing of all-ceramic restorations. J Prosthet Dent 1999;81:652–661.
7. Lawn BR, Deng Y, Thompson VP. Use of contact testing in the characterization and design of all-ceramic crown-like layer structures: A review. J Prosthet Dent 2001;86:495–510.
8. Lee CS, Kim DK, Sanchez J, Miranda P, Pajares A, Lawn BR. Rate effects in critical loads for radial cracking in ceramic coatings. J Am Ceram Soc 2002;85:2019–2024.
9. Wiederhorn SM, Bolz LH. Stress corrosion and static fatigue of glass. J Am Ceram Soc 1970;53:543–548.
10. Wiederhorn SM. A chemical interpretation of static fatigue. J Am Ceram Soc 1972;55:81–85.
11. Willmann G. Improving bearing surfaces of artificial joints. Adv Eng Mater 2001;3:135–141.
12. Lohbauer U, Petschelt A, Greil P. Lifetime prediction of CAD/CAM dental ceramics. J Biomed Mater Res 2002;63:780–785.
13. Lawn BR. Indentation of ceramics with spheres: A century after Hertz. J Am Ceram Soc 1998;81:1977–1994.
14. Rhee YW, Kim HW, Deng Y, Lawn BR. Contact-induced damage in ceramic coatings on compliant substrates: Fracture mechanics and design. J Am Ceram Soc 2001;84:1066–1072.
15. Deng Y, Lawn BR, Lloyd IK. Characterization of damage modes in dental ceramic bilayer structures. J Biomed Mater Res Appl Biomater 2002;63:137–145.
16. Höland W, Schweiger M, Frank M, Rheinberger V. A comparison of the microstructure and properties of the IPS Empress®2 and the IPS Empress® glass-ceramics. J Biomed Mater Res 2000;53:297–303.
17. Chai H, Lawn BR, Wuttiphan S. Fracture modes in brittle coatings with large interlayer modulus mismatch. J Mater Res 1999;14:3805–3817.
18. Kim HW et al. Effect of flaw state on the strength of brittle coatings on soft substrates. J Am Ceram Soc 2001;84:2377–2384.
19. Miranda P, Pajares A, Guiberteau F, Deng Y, Lawn BR. Designing damage-resistant brittle-coating structures: I. Bilayers. Acta Mater 2003;51:4347–4356.
20. Wiederhorn SM. Subcritical crack growth in ceramics. In: Bradt RC, Lange FF, Hasselman DPH, editors. Fracture mechanics of ceramics. New York: Plenum. p. 613.
21. Lawn BR. Fracture of brittle solids. Cambridge: Cambridge University Press; 1993.
22. Lawn BR, Padture NP, Cai H, Guiberteau F. Making ceramics "ductile." Science 1994;263:1114–1116.
23. Kim DK, Jung YG, Peterson IM, Lawn BR. Cyclic fatigue of intrinsically brittle ceramics in contact with spheres. Acta Mater 1999;47:4711–4725.
24. Jung YG, Peterson IM, Kim DK, Lawn BR. Lifetime-limiting strength degradation from contact fatigue in dental ceramics. J Dent Res 2000;79:722–731.
25. Miranda P, Pajares A, Guiberteau F, Cumbrera FL, Lawn BR. Role of flaw statistics in contact fracture of brittle coatings. Acta Mater 2001;49:3719–3726.
26. Lakshminarayanan R, Shetty DK, Cutler RA. Toughening of layered ceramic composites with residual surface compression. J Am Ceram Soc 1996;79:79–87.