Fracture of ceramic/ceramic/polymer trilayers for biomechanical applications

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Abstract: Fracture damage in trilayers consisting of outer and inner brittle layers bonded to a compliant (polycarbonate) substrate and subjected to concentrated surface loading is analyzed. The principal mode of fracture is radial cracking at the undersurface of the inner (core) layer, even in the strongest of core ceramics—other damage modes, including radial cracking in the outer (veneer) layer, are less invasive in these all-brittle coating systems. Tests on simple trilayer structures fabricated from glasses, sapphire, and dental ceramics are used to examine the dependence of the critical load for radial fracture in terms of relative outer/inner layer thickness and modulus, and inner layer strength. An explicit relation for the critical load, based on a flexing plate model in which the outer/inner bilayer is reduced to an "equivalent" monolithic coating with "effective" composite modulus, is used to examine these dependencies. The theoretical relation describes all the major trends in the critical load data over a broad range of variables, thus providing a sound basis for trilayer design. Relevance of the analysis to dental crowns and other biomechanical applications is a central theme of the study. © 2003 Wiley Periodicals, Inc. J Biomed Mater Res 67A: 828–833, 2003

Key words: brittle coatings; cracking; critical loads; dental crowns; trilayers

INTRODUCTION

An issue receiving current attention is that of the failure from concentrated loading of all-ceramic coating layers bonded to compliant substrate bases. Such layer systems are relevant to a wide range of engineering applications, and especially to biomechanical structures.[1,2] Principal examples of the latter are dental crowns[3–5] and acetabular cups in hip replacements.[6] The ceramic coating layers shield an otherwise vulnerable substrate from external loads and provide wear resistance; the compliant substrates cushion impacts and help to constrain any damage within the coating layers. Most work to date has been performed on model bilayers consisting of single-layer glass or ceramic coatings on compliant polymer[7–11] or soft metal[12,13] substrates. More recently, work has been extended to trilayers in which a strong and stiff inner core layer is inserted to provide support to a weak but functional outer veneer layer.[14,15] In dental crowns, ceramic cores offer superior aesthetics and biocompatibility over more traditional high-modulus metals. However, in real life, the brittleness of all-ceramic crown systems compromises the lifetime.[3,4] There is a need to improve the design of these systems—what are the best material combinations, and what are the optimum relative thicknesses?

The principal damage mode in brittle layer structures subjected to contact loading is radial cracking at the ceramic undersurfaces.[2,8,16] In trilayers with all-ceramic coatings it is the stiff (and generally stronger) support cores that are most vulnerable, because most of the tensile stress is transferred to the lower layers in flexure mode.[14] Radial cracks are especially dangerous

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because they can occur at relatively low loads, especially in thinner coating systems, and can spread rapidly to failure.[1,2] Other damage modes have been identified:[2,15] cone cracking in the near-contact region at the outer top surface (thick specimens); quasiplasticity at the top surface or adjacent to interlayer interfaces (softer, tougher ceramics); radial cracking in the outer layer or interface delamination (exacerbated by quasiplasticity in the core or plasticity in the substrate). However, such additional modes are generally secondary in the brittle coating systems used in most biomedical applications—and although there is some suggestion that these modes may be enhanced under certain exacting conditions (thin layers, cyclic loading), they will not be considered in great detail here.

Whereas explicit relations for the critical loads to activate different damage modes have been well documented for bilayers,[2,11] extension to trilayers has proved problematic. Analytical solutions for stress distributions in trilayers are not readily available. Nonetheless, by reducing the trilayer system to an effective bilayer in which the outer/inner composite is regarded as a hypothetical monolithic coating with appropriately averaged "effective modulus," an analogous critical load relation for core radial cracking has been derived.[15] This relation expresses explicit dependence on the relative modulus and thickness of the outer and inner layers, as well as on the strength of the inner layer, providing a basis for predictive design of trilayer composites. However, systematic experimental studies of these controlling variables in the critical load functions remain to be conducted.

The goal in this study is to confirm the critical load relations for core radial cracking in brittle coating trilayers. Model trilayer systems consisting of various combinations of glasses and sapphire for simple specimen fabrication, covering a range of relative elastic moduli and layer thicknesses, are used in an experimental study. *In situ* observation methods are used to measure the critical loads for core radial cracking as a function of relative ceramic layer modulus and thickness. Additional tests on systems containing dental ceramic cores are used to examine the dependence on core strength, and to illustrate the general usefulness of the analysis.

MATERIALS AND METHODS

A schematic of the trilayer system under consideration is given in Figure 1. A hard sphere of radius r is applied to the specimen top surface under load P in a single cycle. The specimen consists of outer and inner layers of thicknesses d_o and d_i and moduli E_o and E_i on a substrate of modulus E_s. The outer/inner and inner/substrate interfaces are assumed to be well bonded. Radial cracks R initiate preferentially at the undersurface of the inner core layer at a critical load.

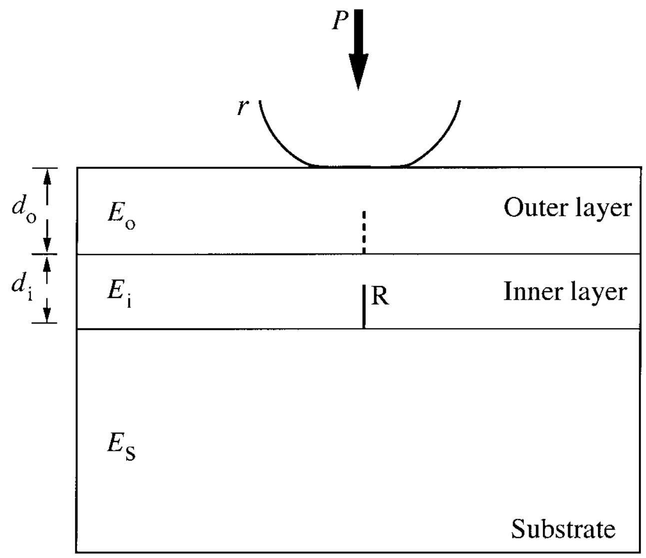

Figure 1. Schematic of trilayers with brittle outer layer of thickness d_o and modulus E_o and brittle inner layer of thickness d_i and modulus E_i on compliant substrate of modulus E_s, in contact at top surface with sphere of radius r at load P. Radial cracking R occurs preferentially at the lower surface of the inner core layer.

Material systems

Model trilayers were fabricated by bonding inner and outer ceramic plates of prescribed thicknesses up to 2 mm and minimal lateral dimension 25 mm to polycarbonate substrate bases of thickness 12.5 mm for contact testing. For the ceramic plates, glasses of different compositions listed in Table I plus sapphire and selected dental ceramics from previous studies[10,14] were used. The glasses and sapphire were selected because of their well-documented behavior as model brittle materials, the dental ceramics for their clinical relevance. The core soda-lime glass undersurfaces were preabraded with 600 SiC grit to provide a uniform density of starting flaws for radial crack initiation;[8] all other surfaces were polished to 1μm diamond finish. Polycarbonate has been previously established as a well-behaved compliant base material—transparent (for *in situ* viewing) and elastic (minimal viscoelasticity).[8] Essential properties of the materials are included in Table II: E from ultrasonic measurements (Grindosonic MK5, J.W. Lemmens Inc., St. Louis, MO), effective strengths S from radial crack data on ceramic/polycarbonate bilayer control specimens (see Results).

Two distinct sets of ceramic-coating trilayer systems were then prepared from the materials in Table II, to examine different variables:

Relative outer/inner thickness and modulus

The first set of specimens was fabricated with glasses and sapphire as the outer layer and soda-lime glass as a common inner core layer, to examine the influence of relative layer thickness d_o/d_i and modulus E_o/E_i for systems with invariant core strength S_i. Total ceramic thickness was fixed at $d = d_o$

TABLE I
Glass Compositions wt(%)

Glass	B_2O_3	Na_2O	SiO_2	PbO	K_2O	CaO	MgO	Al_2O_3	BiO_2	BeO
Boron	86.5	9.1	4.4							
Lead			30	70						
Soda-lime		14	72		1	7	4	2		
Bismuth							22	20	45	13

$+ d_i = 2$ mm. All layers were simply bonded with a thin (<10 μm) interlayer of epoxy resin adhesive (Harcos Chemicals, Bellesville, NJ) with elastic properties similar to that of polycarbonate. Previous studies[14] have confirmed that flexibility in the adhesive at either interface has little effect on the critical load to initiate radial fracture in the core; but that in some systems any adhesive between the outer and inner layers may cause premature fracture in the outer layer from enhanced flexure.[8] Abraded soda-lime glass is ideal as a core material in these studies because its strength is sufficiently low to guarantee that fracture nearly always occurs first in the core; also, the modulus of soda-lime glass lies in the intermediate range in Table II, enabling study of outer/inner modulus ratios both greater and less than unity.

Inner core strength

The second set of specimens was fabricated with a common soda-lime-glass outer layer (similar modulus to dental porcelain[10]) bonded to the dental ceramics listed in Table II, to investigate the role of strength of the core material. Total coating thickness was $d = 1.5$ mm. Because of the high strengths of the dental ceramics relative to soda-lime glass, extra steps were taken to avoid premature fracture in the outer layer: glass-transfer tape (Vitta Corp., Bethel, CT) was used to provide a higher stiffness bond at the glass/Y-TZP and glass/glass–ceramic interfaces (glass/alumina could not be prepared in this way because of thermal expansion mismatch between the two materials); and the outer soda-lime surfaces were preetched in 4% HF to remove surface flaws.

In both specimen sets, bilayers with coatings consisting solely of the nominal core glass or ceramic and of the same total thickness as in the corresponding trilayer specimens ($d_i = d = 2$ or 1.5 mm, $d_o = 0$), were prepared as control specimens.

Fracture tests

The finished trilayers were subjected to contact loading at their top surfaces with WC spheres of radius $r = 3.18$ or 3.96 mm. A thin plastic sheet was placed between sphere and specimen surface to smooth out local stress concentrations from asperities on the indenters, and thus to minimize premature surface cone cracking damage in specimens with particularly strong inner core materials.

The incidence of fracture in the trilayer specimens was observed by *in situ* viewing from the side (glass layers) or from below (glass and ceramic) using an optical telescope arrangement with video recording facilities.[8,9] These tests were used to confirm radial cracking in the inner core as the principal mode of fracture in our systems, and to measure the critical loads.

Critical load relations

The starting point for our analysis is the relation for radial cracking in a *bilayer* of coating material i of thickness d, modulus E_i and strength S_i on substrate of modulus E_s (valid within $1 < E_i/E_s < 100$)

$$P_B = BS_i d^2/\log(E_i/E_s) \tag{1}$$

where B is a dimensionless coefficient.[9,11] An analogous expression has been derived for *trilayers* by replacing the composite outer/inner layer by a hypothetical monolithic layer of same total thickness $d = d_o + d_i$ and "effective modulus" E^* (valid within $1 < E^*/E_s < 100$):[15]

$$P_T = BS_i d^2/[(E_i/E^*)\log(E^*/E_s)] \tag{2}$$

The appearance of the prelogarithmic factor E_i/E^* takes into account the difference in modulus between the actual core material and its monolithic replacement. The effective modulus depends on modulus ratio $\varepsilon = E_o/E_i$ and thickness ratio $\delta = d_o/d_i$ according to a semiempirical function[15]

TABLE II
Properties of Materials

Material	Modulus E (GPa)	Strength S (MPa)
Model materials		
Boron glass	32	
Lead glass	55	
Soda-lime glass (abraded)	73	110
Bismuth glass	140	
Sapphire	417	
Dental materials		
Glass-ceramic (lithium disilicate)	104	414
Alumina (glass-infiltrated)	270	550
Zirconia (Y-TZP)	205	1710
Substrate and adhesive		
Polycarbonate	2.3	
Epoxy	3.5	

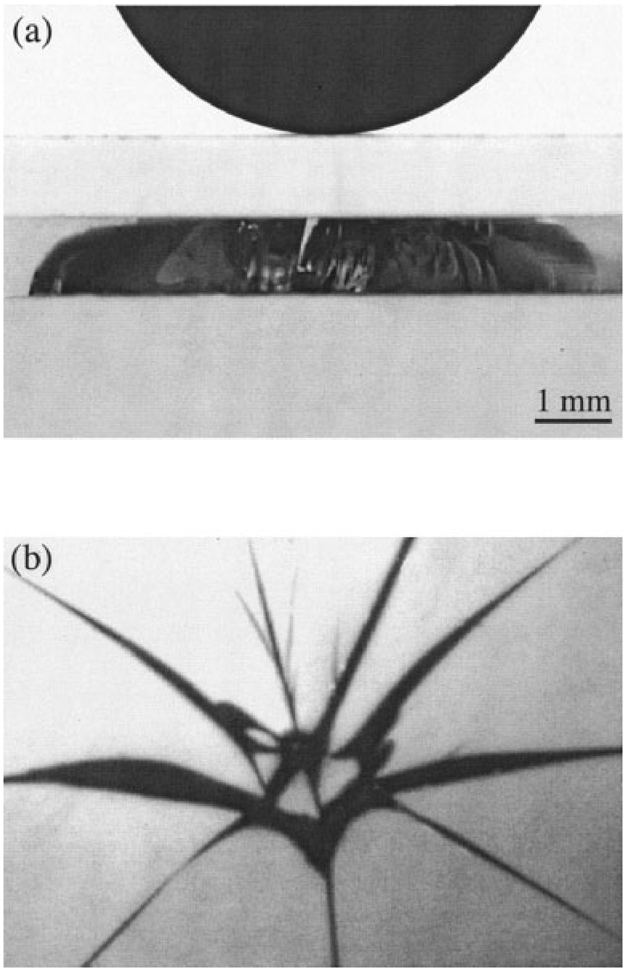

Figure 2. Micrograph showing (a) side and (b) bottom views of radial crack in inner layer of sapphire/glass/polycarbonate trilayer with $d_o = d_i = 1$ mm at threshold contact load $P_T = 700$ N.

$$E^* = E_i\{1 + \varepsilon^2\delta^3 + \varepsilon\delta(5.66 + 2.18\delta)\}/\{1 + 1.97\delta + \varepsilon\delta[(5.66 - 1.97) + 2.18\delta + \delta^2]\} \quad (3)$$

In the bilayer limit $\varepsilon = 1$ (like materials) or $\delta = 0$, we have $E^* = E_i$ in eq. (3), in which case eq. (2) reduces to eq. (1).

RESULTS AND ANALYSIS

Figure 2 are *in situ* side and bottom views of a sapphire/glass/polycarbonate trilayer specimen with $d_o = d_i = 1$ mm at threshold contact load $P_T = 700$ N. The view shows elongate radial cracks fully contained within the soda-lime glass inner core. Combined side and subsurface views were sufficient to confirm preferential radial cracking in the inner cores of all systems studied, except in very special circumstances indicated below. The elongate crack geometry in Figure 2 was typical of all specimens examined.

For the critical load measurements, control tests were first run on the bilayer specimens to "calibrate" essential parameters in the above relations. A minimum of five indentation tests were used to determine mean and standard deviation bounds for the critical radial cracking load P_B in each bilayer system. From these measurements, along with $B = 1.35$ from finite element analysis,[11] effective strengths S_i could be evaluated from eq. (1) for each core material. These values are included in Table II.

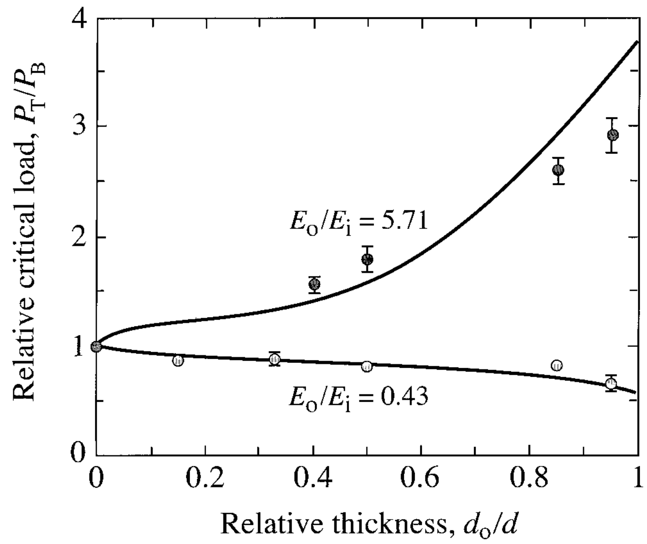

Figure 3. Relative critical load P_T/P_B for inner core radial cracking as function of relative outer-layer thickness d_o/d, for model trilayers. Results for sapphire/soda-lime-glass/polycarbonate ($E_o/E_i = 5.71$) and boron-glass/soda-lime-glass/polycarbonate ($E_o/E_i = 0.43$). Data points are experimental data, solid curves are theoretical predictions.

Results for trilayers with fixed soda-lime glass inner

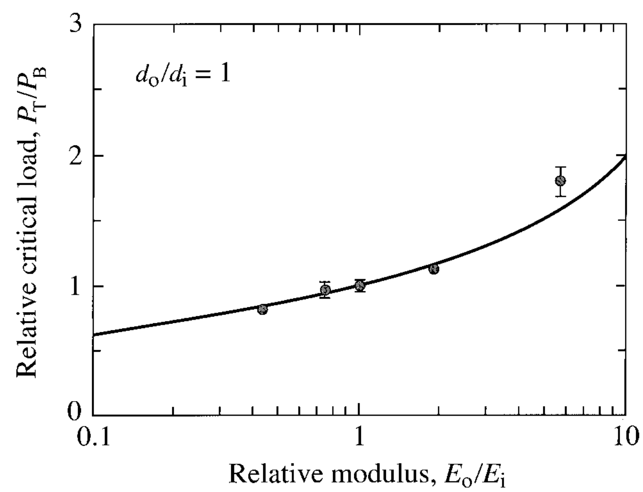

Figure 4. Relative critical load P_T/P_B for inner core radial cracking as function of relative modulus E_o/E_i, for trilayers with various outer layers and common soda-lime glass inner core layers. Results for fixed thickness ratio $d_o/d_i = 1$. Data points are experimental data, solid curves are theoretical predictions.

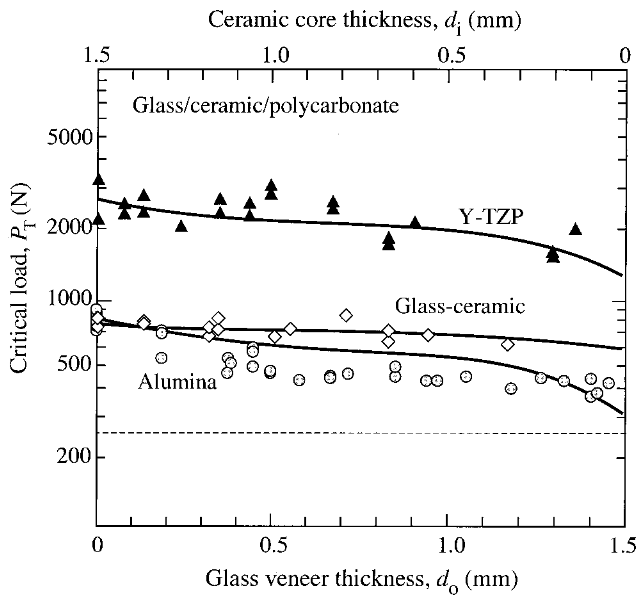

Figure 5. Critical load P_T for inner core radial cracking as function of outer core thickness d_o (or inner thickness d_i), for trilayers with common soda-lime glass outer layers and indicated inner core ceramic layers. Results for fixed net thickness $d = d_o + d_i = 1.5$ mm. Data points are experimental data, solid curves are theoretical predictions.

layers and glass or sapphire outer layers are shown in Figures 3 and 4. The critical loads are plotted in reduced form P_T/P_B: as a function of relative layer thickness d_o/d at fixed $E_o/E_i = 0.43$ and 5.71 (Fig. 3); and as a function of relative modulus E_o/E_i at fixed $d_o/d_i = 1$ mm/1 mm = 1 (Fig. 4). The bilayer limit $P_T/P_B = 1$ corresponds to $d_o/d = 0$ in Figure 3 and $E_o/E_i = 1$ in Figure 4. Points with error bars are experimental data with standard deviation bounds, for first radial fracture in the inner layer. In only limited cases, at $d_o/d < 0.4$ for $E_o/E_i > 1$, did first radial cracking occur first in the outer layer, in this case due to the presence of the epoxy adhesive (hence, the omission of data in this region in Fig. 3). Solid curves are predictions from eqs. (1)–(3).

Figure 5 shows absolute critical loads P_T for trilayers with fixed soda-lime glass outer layer and various stiff ceramic inner support layers as a function of absolute outer layer thickness d_o, at fixed $d = d_o + d_i = 1.5$ mm. Points are individual experimental data for first radial cracking in the inner layer. The solid curves are corresponding predictions from eqs. (1)–(3) using S_i values calibrated from the bilayer tests.

Agreement between predictions and experimental data in Figures (3)–(5) is within 20% over the data range covered.

DISCUSSION

Experiments have been conducted on the critical contact loads to activate fracture in model trilayer systems consisting of ceramic/ceramic coating layers on polycarbonate bases. These experiments demonstrate the vulnerability of even the strongest inner ceramic core layers to lower surface radial fracture from flexure on the compliant substrate. Explicit relations for the critical loads are given in terms of relative outer/inner ceramic thickness and modulus, and inner ceramic strength. When normalized in terms of limiting bilayer data, these relations account for basic trends in the experimental data, with absolute predictions within 20%.

For systems in which the inner core material is fixed, the results in Figure 3 indicate optimal thickness ratios: when the outer layer is stiffer than the inner layer (upper curve), it is better to use a thinner inner layer; conversely, where the outer layer is more compliant (lower curve), it is better to use a thicker inner layer. Note that the lower curve in Figure 3 indicates relative insensitivity to d_o/d in the intermediate region $0.25 < d_o/d < 0.75$, a result of some comfort in the context of dental crowns with stiff inner support layers—errors in fabrication are not so likely to lead to catastrophic strength losses in practice. For systems in which the inner/outer thickness ratio is fixed, Figure 4 demonstrates that it is better for the outer layer to be relatively stiff. Recall, however, if the outer layer is made *too* stiff, then the outer layer itself becomes suceptible to radial cracking.

The results in Figure 5 show that the curves for different core materials with common outer layer shift primarily according to the effective strengths (Table II). Note that for all systems P_T remains relatively insensitive to d_o/d (cf. lower curve in Fig. 3). Y-TZP gives the highest values of P_T; lithium disilicate glass–ceramic and alumina have comparable values of P_B. However, P_T for alumina diminishes somewhat more rapidly than the glass–ceramic as d_o increases, as more stress transfers to the stiffer inner core. So while high strength is a principal requirement for the core material, a low modulus is also beneficial. These results would indicate that superior dental crowns might be fabricated with Y-TZP cores—however, other issues, such as long-term stability under exacting body function;[6] need to be addressed in the total design package.

Although the radial crack relations given here appear to provide a sound basis for layer design, they are nevertheless subject to some intrinsic uncertainties. Equations (1) and (2) are approximate solutions and eq. (3) is determined empirically.[11,15] It is assumed in the derivation of eqs. (1) and (2) that the concentrated loading is ideal point-contact, that the starting flaws from which the radial cracks initiate remain small relative to the layer thickness, and that there is an abundance of such starting flaws in the tensile zone beneath the contact.[2,11,15] These factors become especially important in thinner coatings. Also, eqs. (1) and

(2) are accurate only within a limited modulus range (in fact, the alumina bilayer data point in Figure 5 lies just outside of this range). In addition, the critical loads are subject to rate effects from slow crack growth, amounting to typical reductions of a factor of two or more over a period of a year.[17] Accordingly, the quantities S_i deconvoluted from bilayer critical load data in eq. (1) should be regarded as "effective' strengths that may differ somewhat from bulk strengths using conventional test procedures. Further, competing quasiplastic damage modes may become active under certain conditions, particularly in cyclic loading, leading to premature failure.[2] However, even allowing for these and any other systematic sources of uncertainty, the formulation presented here remains a sound foundation for materials design in biomechanical applications.

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