Contact damage of curved zirconia/epoxy bi-layers

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Abstract. Contact damage of a curved bi-layer system, consisting of a brittle zirconia coating (Y-TZP) on a compliant polymeric substrate from indentation by a hard tungsten carbide sphere is investigated. The specimens are loaded axially at their top surface. The failure evolution and critical loads to initiate radial cracking in curved zirconia/epoxy bi-layer specimens are compared to those of flat bi-layers with the same thickness. The onset of fracture is observed in situ using a video camera. Finite element modelling is used to evaluate the stress distribution in the zirconia coating, and to confirm experimental data. It is demonstrated that in all specimens, cone cracking occurs prior to radial cracking, with the latter being defined as the primary mode of failure. Increasing the curvature has little effect on the critical load to initiate radial cracking, but causes unstable crack propagation towards the extremities of the specimen resulting in catastrophic failure. The results of this study provide useful guidelines from the data trends, and for extending the system to tri-layers.

Keywords: Zirconia, radial cracks, bi-layers, brittle coatings, contact damage

1. Introduction

High performance materials in layered structures are finding increasing usage in a wide range of engineering applications, for example, coated cutting tools, thermal barrier coatings [1–5], and biomechanical replacements such as dental crowns, hip and knee prostheses, heart valves and bone implants [5–7]. The combination of a hard, brittle outer layer and a compliant, tough inner layer offers high wear resistance and damage tolerance, factors that are crucial to the lifetime of such structures. Generally, these structures include more than one material type – ceramic, metal, and polymer – in layered or composite configurations. In biomechanical prostheses ceramics are replacing metals as a material of choice, where biocompatibility, chemical durability, and aesthetics are critical issues. Yttria-stabilized zirconia (Y-TZP) and alumina-matrix composites are replacing metals as core support materials in dental crowns and other biomechanical replacements [8]; however, these materials require special attention because of their brittleness. Currently, the lifetimes of ceramic-based layered structures are unacceptably short, limited by any one of a number of damage modes that may develop in either the coating or the substrate [9–12]. These modes include cone cracking (C) at the coating upper-surface, and radial cracking (R) at the coating under-surface, from flexure induced by concentrated loads at the top surface. Radial cracks have been identified as the most deleterious in the context of system survival [9,13]. A considerable amount of experimental and analytical investigations on various contact damage modes have been carried out, but virtually all have concentrated on flat surfaces [3–8]. Regimes for different
damage modes have been determined, but it is not known how these might be affected by curvature. However, in recent studies of dome-shaped brittle (glass) shell structures filled with a compliant polymer (epoxy resin and epoxy composite) [1,9,14–16], it was demonstrated that surface curvature can play an important role in the radial crack evolution, initially by inhibiting initiation and subsequently – in the case of convex curvature – by strongly enhancing propagation to failure. Most interestingly, the studies revealed the existence of a new mode of fracture, “margin cracks” starting at the specimen edges and propagating into a characteristic “semi-lunar” fracture around one side of the dome, with this achieved by using compliant soft indenters [16]. Fractures of this type have been reported in the clinical dental literature [11,12]. To the best of our knowledge, no previous studies of contact damage in curved (zirconia/polymer) systems have been carried out and it is not known how strong and tough ceramics – zirconia is the material chosen in this study – fail in more complex geometries. Controlled indentation testing with spheres on curved (convex) specimens is used in this study to simulate the basic geometrical shapes in biomechanical and engineering structures, as well as flat surfaces for comparison. The results from this study of bi-layer yttria-stabilized zirconia (Y-TZP) systems provide useful guidelines for extending the system to tri-layers.

Controlled indentation testing with hard spheres on dome-shaped structures is used to study the effect of surface curvature on the evolution of damage from concentrated top-surface loading. Y-TZP of 0.5 mm thick is fabricated by slip casting producing half-sphere shells having inner diameter of 14 mm. The shells are then bonded to an epoxy resin base to produce bi-layers with convex surface geometry. A tungsten carbide sphere of radius 3.175 mm is used to apply an axisymmetric load on the specimen top surfaces. Cone and radial crack evolution is observed in situ using a video camera system; some specimens were loaded in step-loading and viewed after unloading due to the opacity of the zirconia and the indenter obstructing the views, which made it difficult to observe exactly when cone cracks initiated during loading. Results are interpreted using finite element modelling (FEM) of the stress states at the brittle layer under-surfaces, and compared with previous studies on glass/polymer bi-layer systems similar to the system under consideration here. It is demonstrated that for the zirconia/polymer bi-layer system, surface curvature (convex) has no effect on the load to initiate the first radial crack, but the same curvature can diminish the loads to ultimate failure. Furthermore, unlike the glass/polymer system, in both the flat and curved zirconia/polymer specimens, cone cracking initiates at the top surface prior to radial cracking. The extent of quasi-plastic damage was minimal in the experimental tests on both flat and curved zirconia/epoxy specimens.

2. Experimental procedures

2.1. Specimens fabrication

Curved and flat zirconia/epoxy bi-layers, were fabricated for indentation testing in configuration of Fig. 1. Properties of relevant materials are shown in Table 1. The zirconia shells were prepared at “Rojan Ceramics Co, Henderson, Western Australia” [17] using the slip-casting technique in two configurations, convex shells having 14 mm inner diameter and 0.5 mm thick, and flat geometry 20 × 20 × 0.5 mm in dimension. A slip mixture containing zirconia particles (ZY3, Millennium Performance Chemicals, Australia) was poured into a mould and then removed after a shell of 0.65 mm thick was formed. A Vernier calliper was used to measure the thickness of the solid layer as it was deposited. After casting, the specimens were initially fired in a kiln at 850°C for 30 minutes to reduce the risk of chipping.
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Fig. 1. Schematic showing indentation with a spherical indenter of radius $r_i$, load $P$ on a dome structure of inner radius $r_s$ consisting of a brittle hemispherical shell of thickness $d$ supported by polymeric base extending depth $h$ below the margin edges. The symbol $L$ represents the width of the flat specimen.

Table 1
Materials properties for input into FEM

<table>
<thead>
<tr>
<th>Material</th>
<th>Young’s modulus (GPa)</th>
<th>Poisson’s ratio</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Indenter</td>
<td>(E_I) 613</td>
<td>0.22</td>
<td>Elastic</td>
</tr>
<tr>
<td>Coating</td>
<td>(E_C) 200</td>
<td>0.20</td>
<td>Elastic</td>
</tr>
<tr>
<td>Substrate</td>
<td>(E_S) 3.4</td>
<td>0.33</td>
<td>Elastic</td>
</tr>
</tbody>
</table>

the brittle material during handling, polishing and shape forming through “green-machine” [18]. Shape forming or green-machining is conducted on the specimens before final sintering, using polishing paper (P240 grit size, ~60 µm particles) to remove any irregularities at the edges. The top and bottom surfaces of the flat layers were polished with P400 grit size paper (~35 µm particles) to ensure the thickness was consistent. The specimens (zirconia layer) were sintered at 1500°C to remove porosities and strengthen the bonds between the grains in the material [19], resulting approximately in a 70% increase in density and linear shrinkage of 20%. After sintering, the flat specimens reduced in size to a thickness of 0.5 mm, and the curved specimens to a thickness of 0.5 mm and inner radius of 7 mm. The amount of densification and shrinkage for the curved zirconia layers after slip-casting, initial firing and sintering are calculated, as indicated in Appendix.

The zirconia plates were then fitted into a mould of the same lateral dimensions. Epoxy resin (ADR2512/ADH2407, ATL Composites, Australia) was then poured into the mould to form a substrate with depth of $r_s + h$ (12 mm) for the curved specimens, and $d + h$ ($\approx 5$ mm) for the flat geometry
tests, with intermittent vibration to remove air bubbles. The resulting specimen configuration is depicted schematically in Fig. 1. To avoid residual stresses – from the shrinkage of the epoxy during the curing process – the epoxy was applied according to the manufacturer’s specifications in thin layers <5 mm, allowing each successive layer to cure under the action of a slow hardener for 24 hours before adding the next. Residual stresses in the zirconia coating can therefore be neglected; this procedure was used successfully in preceding studies [14–16]. Polished sections through the specimen revealed a strong bonding without any sign of delamination from the substrate (see Fig. 5).

2.2. Testing

Indentation tests were carried out in air (at room temperature) using a tungsten carbide sphere of radius 3.175 mm mounted in a screw-driven testing machine (Instron 4301, Instron Corp., Canton, MA). The specimens were loaded axially and monotonically at cross-head speed 0.005 mm min$^{-1}$ ($\sim$10 N min$^{-1}$); loads of up to 2000 N were applied. Five to ten separate tests were conducted on the flat and convex specimens, and 3 to 6 step loading tests were carried out to determine cone crack critical loads. During loading, the specimens were viewed from the side and slightly from above using a video camera, such that the contact and side walls of the specimens were within the field of view at all times (although not as clearly close to indentation area, where the sphere obstructed the surface view). A light source was placed behind the specimens to optimise crack visibility. A single-cycle axisymmetric indentation was performed on each specimen. Critical loads to initiate radial cracks at the zirconia under-surfaces, and cone cracks at the zirconia upper-surface were monitored.

2.3. Finite element modelling

Similar to the previous studies [1,9,14–16], finite element modeling was used to compute the state of stresses at the zirconia layer. Details of the FEM procedures, using Abaqus V6.5 software, have been well documented [20,21]. The curved model specimen model has a 14 mm inner diameter and thickness of 0.5 mm, and the support thickness $h$ below the hemispherical base was 5 mm. The bi-layer flat model is represented by a rectangle having a height ($h+d$) and a width $L=10$ mm, with $h$ and $L$ large enough compared to the zirconia layer thickness $d$ for the solution to be independent of the dimensions $h$ and $L$ and the substrate can be regarded as a half space [1,3]. Properties of relevant materials for input into FEM are shown in Table 1. Contact was assumed to be frictionless. The meshes were systematically refined, particularly in the critical region – zirconia coating near the contact, until the solutions attained convergence. The mesh consisted of up to 25,500 elements, with a minimum element size 10 $\mu$m near the contact zone.

3. Results

Representative photographs from video sequences demonstrating the main effects of surface curvature on radial ($R$) and cone ($C$) crack evolutions were selected and included in this paper. In all the systems under consideration in this study, cone cracking was found to be the first mode of cracking – cone cracking initiates at the coating upper-surface prior to radial cracking at the under-surface of the coating, as shown in Fig. 2. Both radial and cone cracks can be observed, however the segmentation from the radial cracks indicates the prior presence of cone cracks (see Fig. 3(c)). Audible clicks could be heard at lower loads $P \approx 300$ N for both the flat and curved specimens without any visible sign of damage,
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Fig. 2. Contact fractures in epoxy-filled zirconia shells of thickness \(d = 0.5\) mm, indented at axis of symmetry with TC sphere of radius \(r_1 = 3.175\) mm load \(P\): (a) 680 N (flat geometry), and (b) 1400 N (curved geometry). Showing failure from radial cracks (b). Views taken after unloading, multiple radial and cone cracks are observed.

Fig. 3. Contact fractures in epoxy-filled zirconia domes of inner radius 7 mm, indented with TC spherical indenter at load \(P\): (a) 600 N, and (b) 800 N (critical load), (c) specimen unloaded after segmentation at reduced load 750 N. Showing failure from radial cracks. Note crack location, and how multiple cone propagate fully through the zirconia shell.

which could suggest the initiation of cone cracks. However the extent of damage from cone cracking was minimal. Consequently, radial cracking is the focus of this study.

Figure 2 highlights the effect of curvature on contact damage in zirconia/epoxy bi-layer specimens, in comparison with the flat specimens. Generally, similar loads were required to initiate radial cracks in both the flat and curved specimens for the given geometries and dimensions, but the ensuing “pop-in” of the radial arms was more abrupt in the curved specimens. The radial crack densities also tended to be higher than in the flat specimens, and most interestingly in the curved specimens, radial cracks popped-in without interruption to the specimen extremities, resulting in instantaneous failure with a sudden decrease in load <100 N. After further loading <50 N, more radial arms appear to initiate after others in similar fashion. In the flat specimens, the radial cracks propagated steadily in uneven lengths, but nonetheless at a detectable rate, towards the edge with a mean difference of approximately 50 N recorded between the critical load and failure load. As can be seen from Fig. 2(b) the curved geometry is more likely to fail catastrophically from radial cracking. This is in accord with the experimental observations from curved glass/polymer bi-layers [9].
Figure 4. Contact fractures in epoxy-filled glass domes of inner radius 8 mm, 0.5 mm thick, indented with TC sphere radius 4 mm at load \(P\): (a) 200 N, (b) 300, and (c) 700 N. Showing failure from radial cracks. Note how radial cracks initiate (a) and propagate steadily toward extremities of the specimen. Cone crack observed only in (c) after unloading.

Figure 3 shows a sequence of video clips during indentation of a convex surface with zirconia coating inner radius \(r_s = 7\) mm, and coating thickness of 0.5 mm. Note in (a) the indenter obstructed the view, which made it difficult to observe exactly when cone cracks initiated during loading. As mentioned earlier, the radial cracks popped-in and propagated without interruption to the base. Also of interest is the initiation of well developed cone cracks which can be seen in Fig. 3(c). More interestingly, radial cracks do not cross the cone crack boundaries. This can be seen also from Fig. 5, which suggests that cone cracks initiate and propagate well before radial arms, also that radial cracks, originating in the zirconia under-layer, could be anywhere between the indentation area and the specimen edge, depending on the availability of starting flaws.

Similar to the sequence of video clips (Fig. 3), representative photographs from video sequences in Fig. 4 demonstrate the typical failure of glass coatings – representing less stiff porcelain (Young’s Modulus of 73 MPa) [22], for the same coating thickness 0.5 mm, and same substrate materials. In this case, the first crack initiated from the near-contact zone, followed by steady propagation towards the base of the dome and through the thickness of the glass layer [2] – this being a consistent feature in the evolution of damage in such a system. At higher loads, the radial cracks linked up at their base, causing separation and dislodgement of triangular glass segments (Fig. 4(c)) – note similarity of failure pattern in the zirconia coatings (Fig. 3(c)). Top surface ring cracks were apparent in the unloaded specimens, but again were preceded by the radials in the glass system [2,9,14,22], unlike the zirconia coating in the present study where cone cracks initiate before radial cracks (Fig. 3(c) and Fig. 5).

Figure 5 shows a cross-sectional view of an epoxy-filled zirconia dome of inner radius 7 mm and coating thickness of 0.5 mm. The pattern of a well developed cone crack is clearly evident from the cross-sectional view, suggesting that cone cracks initiate before radial cracks.

Critical loads \(I\) to initiate subsurface radial cracks (responsible for the structure failure) in the curved zirconia surfaces were measured as a function of specimen geometry. Data are plotted in Fig. 6 for flat and curved zirconia coatings, as well as for glass coatings from a previous study [22] for comparison. As explained earlier after initiation of the radial crack, with further increase in load more cracks initiated and linked with neighbouring radial arms, causing portions of the coating to separate from the substrate and ultimately material dislodgment. This final stage in the failure process is indicated in Fig. 6 and represented by \(F\) bars “Failure loads”. Note that for zirconia coatings, \(I \approx F\), and the extended \(F\) loads for the glass coatings, due to the steady crack growth after initiation.
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Fig. 5. Cross-section view of epoxy-filled zirconia dome of inner radius 7 mm. Note the initiation of cone cracks at the top surface of the coating.

Fig. 6. Histogram showing critical loads to initiate radial cracks \((I)\) and loads to propagate these same cracks to failure \((F)\). Flat and curved geometries having the same coating thickness \(d = 0.5\) mm. Glass shells data for as-polished under-surface from previous study [22].

Figure 7 shows FEM simulations of maximum principal stresses at an applied load of 800 N, for flat specimens \((a)\) and curved \((b)\). Tensile stresses are displayed – a lighter shade indicates higher tensile stress. The plots clearly show the effect of curvature. Of primary interest is the extended tensile region outside the contact zone towards the specimen edge (see Fig. 7(b)). Note the concentration of contours under the contact zone in the case of flat geometry in Fig. 7(a). The extensions of contours around the dome side in Fig. 7(b) are a direct consequence of coating flexure, suggesting that crack initiation could occur anywhere in this region, depending on the availability of starting flaws.

Stress distributions around the inner surface of the zirconia coating are shown in Fig. 8, for both flat and curved specimens. The maximum tensile stresses are plotted as a function of longitudinal coordinates \(S\) (see insertion in Fig. 8), for load increment of 800 N. Considering the stress variation along the coordinate \(S\), FEM computations shed some insight into the influence of surface curvature. The tensile stresses maxima along the contact axis are close between the flat and curved specimens, accounting for the relatively similar critical loads for radial crack initiation in Fig. 6. Of interest, the curves in the plot cross each other (see arrow).
4. Discussion

The failure of brittle dome structures from axial loading with hard indenters has been investigated. The results show that surface curvature (convex) has little effect on critical loads to initiate the first radial crack for the dome geometry and dimensions under consideration, but this same curvature can diminish the loads to ultimate failure. Furthermore, unlike the glass/polymer system, cone cracking in both the
Fig. 8. Plot of maximum principal stresses as a function of latitudinal coordinate S measured from the contact axis along inner surface, for epoxy-filled zirconia thickness 0.5 mm, curved domes of inner radius 7 mm and flat geometries. Indentation load $P = 800$ N. Note crossover of curves, indicated by arrow.

flat and curved zirconia/epoxy resin specimens initiated at the top surface prior to radial cracking. Most prior studies have been conducted on flat bi-layer specimens [3–11] or, more recently, on curved dome structures with glass coatings [9,14–16,22]. In those studies the confined contact between hard indenter and brittle surface favoured a flexural mode of fracture, leading to dominant radial cracks or near-contact cone cracking.

The results highlight the importance of surface curvature in the fracture behaviour. As mentioned, radial cracking is widely believed to be the principal mode of failure of such systems [9,13]. Convex surfaces would seem to be especially vulnerable to sudden unstable pop-in from the subsurface flaws at a critical initiation load, and crack propagation through the entirety of the structure. Again, tests on flat layers may be used to provide a conservative baseline for estimating failure loads. In this case, the fractures are more likely to remain localised around the contact site rather than sudden propagation to the edges of the specimen as in curved structures. Further more, the zirconia used in this study is much stiffer and stronger than traditional porcelain used in dental crowns – hence, the argument that radial cracks initiate first in thinner coatings [2–9,13–16] stands for certain materials, and cannot be generalised (see Fig. 3(c) and Fig. 5). It is necessary to consider all the variables in the design of brittle coating layered structures, as each of the variables – coating thickness, modulus mismatch, indenter size and materials stiffness, etc. – has a significant effect on one or more of the critical conditions and the evolution of damage in these systems. As such, experimental and analytical investigations based on fixing one of the key entities cannot be generalised, as the scaling of the results for different dimensions is not straightforward [9,15,22,23]. Furthermore, variation in two or more of these variables causes the behaviour of the system to become quite complex.

Use of the present data to make quantitative predictions of failure is more ambitious. In this study, particular variables and conditions remained constants: using only 0.5 mm thickness coatings, single-cycle loading and bi-layer system – zirconia is more commonly used as a support layer in tri-layer structures. Nevertheless, the methodology does provide useful guidelines for data trends and extending the system to the tri-layer. The long term goal with the ongoing experimental studies on material layer systems is to characterise the variables affecting damage. In doing so, the conditions in which safe
operation can be achieved for a given system and the geometry and properties that optimise damage resistance can be determined.

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Appendix. Densification and shrinkage in the curved zirconia layers during preparation

<table>
<thead>
<tr>
<th>Stage</th>
<th>Inner radius, ( r_s ) (mm)</th>
<th>Thickness, ( d ) (mm)</th>
<th>Ave linear shrinkage ( a ) (%)</th>
<th>Volume ( (\text{mm}^3) )</th>
<th>Mass ( (g) )</th>
<th>Density, ( \rho ) ( (g/mm^3) )</th>
<th>Increase in density ( d ) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1)</td>
<td>9.00</td>
<td>0.65</td>
<td>–</td>
<td>615</td>
<td>1</td>
<td>0.00630</td>
<td>–</td>
</tr>
<tr>
<td>(2)</td>
<td>8.25</td>
<td>0.60</td>
<td>8</td>
<td>477</td>
<td>1</td>
<td>0.00210</td>
<td>29</td>
</tr>
<tr>
<td>(3)</td>
<td>7.00</td>
<td>0.50</td>
<td>16</td>
<td>286</td>
<td>1</td>
<td>0.00349</td>
<td>66</td>
</tr>
<tr>
<td>Total</td>
<td>–</td>
<td>–</td>
<td>23</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>115</td>
</tr>
</tbody>
</table>

(a) After slip-casting; (2) After initial firing; (3) After sintering.

(a) Average linear shrinkage = \( \frac{(r_s1-r_s2)+(d_1-d_2)}{r_s1+d_1} \cdot 100 \% \).

(b) Density, \( \rho = \frac{\text{mass}}{\text{volume}} \ [g/mm^3] \).

(c) Increase in density = \( \frac{\rho_2-\rho_1}{\rho_1} \cdot 100 \% \).

(d) Total shrinkage = \( \frac{(r_s1-r_s3)+(d_1-d_3)}{r_s1+d_1} \cdot 100 \% \).

Total densification = \( \frac{(\rho_3-\rho_1)}{\rho_1} \cdot 100 \% \).

References


