Failure criteria of dentin–resin adhesion – The J -integral approach

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In dentistry, current tests to determine the bond strength of dentin–resin adhesives are simple but result in large variations in the cleavage load. Our numerical results show that these variations could be minimized by using linear elastic fracture mechanics to analyze tensile specimens into which a subcritical precrack in the adhesive layer which initiates crack propagation has been introduced. In our model, the minimum fracture toughness was obtained when 11% of mode II fracture accounted for the differences in the elastic moduli of the dentin and the resin.

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Adhesive bonding agents are extensively applied in dentistry, where restorative composite resins are frequently bonded to natural tooth materials such as dentin and enamel. Therefore, determining the interfacial bond strength is likewise important, because reliable clinical performance of adhesive agents is predicated on the ability to accurately measure bond strengths [1]. The most popular methods for determining bond strength in dental research are shear and tensile tests, which define adhesive strength as the measured failure load divided by the cross-sectional area of the bonded surface and presumes an approximately uniform stress distribution at the cross-section [2]. However, finite element analysis has revealed the non-uniformity of the stress distribution at the interface [3]. In fact, the “average stress” of conventional bond strength tests does not accurately represent the interfacial failure stress. This conceptual flaw may contribute to the large variations noted among the results of numerous studies [4].

It has been suggested that a more promising approach to determining bond strength should be based on interfacial fracture mechanics [5,6], which represents the local effort of creating a new crack surface. The theoretical and experimental advances that have been made in the application of interfacial fracture mechanics in

general engineering suggest that these ideas can be carried over into the field of biomaterials and biomechanics, and should help rationalize the biomechanical successes and failures of bonded biological structures such as the dentin–enamel complexes [7].

In 1998, Lin et al. first proposed the failure criteria of a dentin–resin tensile bond test through a fracture mechanics approach [8]. In that study, an analytical method was used by deriving fracture mechanics-based equations to obtain the mode I fracture toughness, K_{IC} , and Y , a dimensionless stress intensity factor coefficient. However, it is advisable that applications of fracture mechanics concepts to the failure criteria of bonding in dentistry be numerically justified before proceeding further towards the experimental stage. A pathway-independent method for approximating the strain concentration of interfacial cracks known as the J -integral was introduced by Rice in 1968 [9]. The purpose of this study was to use the J -integral method to numerically justify the theoretical concepts and the failure criteria of the dentin–resin tensile bond tests proposed by Lin et al. [8].

The configuration of the dentin–resin tensile bond test specimen and a diagram of the bond interface cross-section are shown in Figure 1. A pathway-independent method for approximating the strain concentration of interfacial cracks is known as the J -integral [9]. This method can be expressed in the following equation:

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Figure 1. (a) Configuration of a conventional tensile bond test used in dentistry. (b) Diagram of the bonded area. D , diameter of the original bonded area; a_0 , precrack length; a , crack length.

$$J = \int_{\Gamma} \left(W dy - \vec{T} \cdot \frac{\partial \vec{u}}{\partial x} ds \right) = \int_{\Gamma} \left(W dy - T_i \frac{\partial u_i}{\partial x} ds \right) \quad (1)$$

where Γ is any path surrounding the crack tip, W is the strain-energy density, u is the displacement vector, s is the distance along path Γ and T is the traction vector. This method avoids direct computation of the stress in close proximity to the crack tip. In addition, the J -integral can be used to represent the stress concentration at the fracture point. For elastic–plastic materials, it can be proven that the J -integral is indeed path independent and can explain the elastic–plastic discrepancies.

In cases of linear elasticity, the J -integral can be expressed by the following:

$$J = G = \frac{K^2}{E'} \quad (2)$$

where G is the strain energy release rate, K is the stress intensity factor, and E' is E (elastic modulus) in plane stress and $E/(1 - \nu^2)$ in plane strain.

With regard to how fractures propagate along boundaries of different materials, Malyshev and Salganic [10] formulated a model that relates G to K in the following way:

$$G = 2\beta K \bar{K} \quad (3)$$

$$\beta = \frac{1}{16 \cosh^2(\alpha\pi)} \left[\frac{\kappa_1 + 1}{\mu_1} + \frac{\kappa_2 + 1}{\mu_2} \right] \quad (4)$$

where $K = K_I + iK_{II}$. The bulk modulus and shear modulus of the two materials are represented by k and μ , respectively. When linear elasticity is involved, the following relationship can be applied:

$$J = G = 2\beta K \bar{K} \quad (5)$$

Finite element analysis was conducted on a graphics workstation using ABAQUS (edition 6.3) (Hibbitt, Karlsson & Sorensen, Inc., Pawtucket, RI). I-DEAS 8.0 (EDS, Plano, TX) was used in conjunction with ABAQUS to construct the finite element models. The dimensions of the model were taken from experimental test specimens. A thin strip (less than 20 μm in thickness) of adhesive tape with a punched hole (3 mm in diameter) is normally utilized to precisely define the bonding area. A constant, gentle bonding pressure (10 N) was generated by the composite rod (4 mm in diameter) onto the bonding area shown in Figures 1

and 2(a), where a_0 is the precrack length due to the sharp/thin edge of the adhesive interface. While the model dimensions may be insufficient in size for the test to constitute a plane strain, light-curing limitations in clinical practice dictate that specimens cannot exceed 4 mm in diameter. A uniformly distributed tensile load of 10 N was applied at the unbonded composite cylinder top surface. The nodes at the dentin base were restrained for all degrees-of-freedom. In this analysis, the assumed Young's moduli and Poisson's ratios were: $E_d = 18.3$ GPa and $\nu_d = 0.30$ for human dentin, and $E_r = 16.6$ GPa and $\nu_r = 0.24$ for composite resin, respectively [2]. The composite resin is composed of polymer matrix and inorganic filler, where the inorganic filler occupies 77–84 wt.% of the composite [2]. The non-linear properties of dental composite resins are not available and previous related studies all assumed that the composite resin has linear elastic properties [11,12]. Moreover, the currently used dental adhesive is mainly composed of dimethacrylate oligomers while the composite resin is composed of polymer matrix and inorganic filler particles [2]. Because the adhesive exhibits a similar composition and lower viscosity to the composite resin, the adhesive was not modeled in this study. The hybrid layer, which principally contributes to the bond strengths and forms between the resin and dentin after bonding, is only 2–5 μm in thickness.

While finite element analyses have become increasingly popular because they eliminate variations inherent with experimentation, they are still essentially approximations. Error analyses were conducted to eliminate contour dependence and ensure that the mesh was sufficiently accurate in the details and structural response without exceeding the limitations of the computational hardware. Theoretically, crack extension was simulated and the resulting energy loss was calculated. This energy loss is the energy required for cracking and represents the physical meaning of the J -integral. In practice, the evaluation is the virtual motion of a block of material surrounding the crack tip. Each material block is defined by its contours, where each contour is a ring of elements surrounding the crack tip. These rings of elements are then defined recursively to surround the previous contours. Sharp cracks are usually modeled using small-strain assumptions. Focused meshes should normally be used for small-strain fracture mechanics evaluations. However, for sharp cracks, the strain field becomes singular at the crack tip, as shown in Figure 2. Based on the

Figure 2. (a) Cross-sectional diagram of the bonded area. a_0 , precrack length = 0.5 mm. The dashed arrow indicates the fracture path. (b) Typical focused mesh for fracture mechanics evaluation. $\mathbf{n}$, the normal crack plane; $\mathbf{q}$, the virtual crack extension direction.

node sets given to define the crack tip, ABAQUS was able to determine the elements which formed each ring. The crack tip was constructed using 20-node bricks in conjunction with a collapsed face. As the displacements vary with $r^{1/2}$ near the crack tip and $r^{-1/2}$ at the crack tip, square root and $1/r$ singularity elements were created by setting the midside nodes for the two elements at the quarter points and midpoints, respectively. The nodes on the collapsed face were then constrained to move together using multi-point constraints.

After element construction, the finite element analysis was conducted using ABAQUS to obtain the J -integral, and K_I and K_{II} values for various normalized crack lengths. The phase angle was also calculated because interfacial elastic mismatches are inherent in interface fracture problems. Essentially a measure of the ratio with which mode I and the mixed mode influence the fracture process, the phase angle can be expressed by the following equation:

$$\text{Phase angle} = \tan^{-1}(K_{II}/K_I) = \tan^{-1}[\Delta\delta_x/\Delta\delta_y]$$

where K_I and K_{II} are mode I and mixed-mode fracture toughness, respectively, and $\Delta\delta_x$ and $\Delta\delta_y$ are the relative displacements of two neighboring points on the crack surfaces behind the crack tip [13].

Using fracture mechanics equations, the stress intensity factor, K , and stress intensity factor coefficient, Y , corresponding to each normalized crack length were subsequently obtained.

J -integral values as a function of the normalized crack length for tensile loading are shown in Figure 3. Upon comparison, the minimum J -integral was observed at $\alpha = 0.15$, after which the curve increased monotonically. We further ascertained that at $\alpha = 0.15$ the critical stress intensity factor and minimum stress intensity factor coefficient could also be found. This result implies that $0.15D$ is actually the critical crack length for the specimen geometry modeled in this study, and is similar to the value proposed by Lin et al. [8] though they left the peripheral precrack out of their consideration. Thus, the minimum J -integral which occurs at the critical crack length can be considered the critical J -integral and corresponds to the critical load of the tensile test. In other words, the tensile test specimen under-

Figure 3. J -integral (J) as a function of the normalized crack length ($\alpha = a/D$).

Table 1. K_I , K_{II} , K_{II}/K_I and phase angle values for various α

α	K_I (MN m ^{-3/2})	K_{II} (MN m ^{-3/2})	K_{II}/K_I	Phase angle ψ (rad)
0.15	0.04101	0.00460	0.1121	6.395
0.175	0.04228	0.00240	0.0566	3.242
0.2	0.04663	0.00205	0.0439	2.512
0.225	0.05056	0.00182	0.0359	2.058
0.25	0.05477	0.00158	0.0289	1.654
0.275	0.05981	0.00121	0.0202	1.156
0.3	0.06568	0.00077	0.0118	0.675
0.325	0.07253	0.00030	0.0041	0.236
0.35	0.08030	-0.00015	-0.0019	-0.108

goes steady-state crack growth until $0.15D$, after which catastrophic fracture ensues. Thus, it is imperative that the critical crack length be determined before conducting any tensile bond tests to ensure that the obtained critical load accurately reflects the fracture toughness of the tested material.

K_I , K_{II} , K_I/K_{II} and the phase angle are listed in Table 1. When $\alpha = 0.15$, the phase angle was approximately 6.4° , which suggests that the stress condition was not entirely mode I, while K_{II} was about 11% of K_I . As the normalized crack length, α , increased, however, the phase angle decreased noticeably. In fact, when $\alpha = 0.325$, K_{II} was less than 0.5% of K_I , which indicates that the effect of K_{II} diminished and mode I dominated the observed stress condition following the normalized crack length increase. Nonetheless, it is because the minimum J -integral was at $\alpha = 0.15$ that the experimental peak load could not be measured at $\alpha = 0.325$. The introduction of the mode II fracture condition was due to the differences in the elastic moduli between the dentin and the resin. In the mode II fracture condition, shear force was introduced and the measured maximal force was reduced compared with that of the pure tensile test. As a result, the measured maximal force may tend to be underestimated.

The finite element analysis results, fracture mechanics theory and historical review were referenced to obtain the minimum stress intensity factor coefficient.

$$J_{IC} = \frac{K_{IC}^2}{E^*} \quad (6)$$

$$K_{IC} = \frac{P_c}{D^{1.5}} f\left(\frac{a_c}{D}\right) \quad (7)$$

Here, P_c is the critical load, $f(a_c/D)$ is the minimum stress intensity factor coefficient, D is the bonding diameter, a_c is the critical crack length, K_{IC} is the critical stress intensity factor, J_{IC} is the critical J -integral and E^* is the combined elastic modulus.

$$J_{IC} = \frac{P_c^2}{D^3 \cdot E^*} f^2\left(\frac{a_c}{D}\right) \quad (8)$$

$$Y_m = f\left(\frac{a_c}{D}\right) = 1.0135 \quad (9)$$

In Eq. (7), $D = 4$ mm, $P_c = 10$ N and $1/E^* = 0.5(1/E_d + 1/E_r)$. After some calculations, E^* was determined to be 15,000 MPa [6]. Putting this value into Eq. (6), Y_m can be obtained using Eq. (9), where Y_m is the minimum stress intensity factor coefficient. This study showed that if P_c and Y_m are known, then the mode I fracture toughness,

K_{IC} , can be obtained. The experiment to validate Y is difficult to perform and should be controlled very precisely. Nonetheless, it is worth doing in the future to calibrate the coefficient.

Large variations have been reported in bond strength values from different investigators, with coefficients of variation of between 65% and 111% [4]. As shown in Figure 3, the stress intensity factor coefficients increased rapidly for $\alpha \leq 0.15$. The experiments simulated in this study were under displacement control. Therefore, we found that energy release decreased monotonically before increasing. Furthermore, the condition of the bond at the edge of the adhesive interface modeled in this study was ideal in that the amount of adhesive was neither excessive nor insufficient, thus eliminating any environmental effects resulting from specimen preparation. These findings suggest that the measurement of bond strength or fracture toughness is strongly affected by the length of the initiating crack as well as the local conditions near the edge of the adhesive interface. In fact, the large variations in fracture toughness observed in previous studies may have depended, in part, on these two factors. Therefore, including a precrack of less than $0.15D$ in length and $20 \mu\text{m}$ in thickness in conventional tensile bond tests may help reduce variations in experimental measurements, promote test stability and facilitate the assessment of fracture toughness, though it is not a pure mode I fracture toughness.

While it is appealing to use a conventional tensile bond test to determine the interfacial bond strength due to its simplicity, the imprecise and theoretically flawed nature of the test tends to produce inaccurate data with large variations. The results suggest that the large variations observed in various studies may be associated with the local bond conditions in proximity to the

edge of the adhesive interface. Thus, a precrack, not exceeding the critical crack length and sharp/thin enough to initiate crack propagation, should be included in the specimen to eliminate any environmental effects resulting from specimen preparation. Furthermore, the data also showed that while the critical stress intensity factor derived from the calculated minimum J -integral consisted predominantly of mode I, a small amount of mode II component (with a 6.4° phase angle) was also included. This finding implies that the conventional tensile bond test is inherently not a pure assessment of mode I fracture toughness, and other test methods should be considered to ensure a purely mode I measurement.

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