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A fracture mechanics approach to ceramo–metal bond evaluation

D. R. WILEY*, N. E. WATERS
Department of Dental Materials Science, UMDS Guy's Campus, London SE1 9RT, UK

The incidence of mechanical failure of cast metal/porcelain dental restorations has promoted a study to develop reliable methods of characterizing and improving bond strengths when produced under standard dental laboratory conditions. Single-edge notch beam specimens were prepared by firing porcelain from a single blended frit to either side of a central metal coupon and subjecting these to four-point bending at 20 ± 2° C and 0.5 mm min^{-1} crosshead speed. Specimens of three representative dental alloys and an experimental alloy were prepared in two specimen sizes and three notch widths. The fracture toughness (K_{Ic}) values were independent of notch width and specimen size over the range of these variables examined and good discrimination was obtained. The fracture toughness of all bonds was improved by a commercial hot isostatic pressure cycle. SEM examination of the fractured surfaces suggested that this improvement was due to the reduction in microporosity at the interface.

1. Introduction

In the past decade increasing use has been made of porcelain fused to metal restorations, for such structures combine the aesthetic qualities of the all porcelain restoration with the strength and marginal integrity of the cast restoration. This combination of properties enables such restorations to be used for both crowns and bridgework which are subjected to the high stresses imposed by mastication.

The incidence of mechanical failure of cast metal porcelain restorations both during fabrication and under service conditions has prompted a study to develop reliable methods of characterizing and improving bond strengths when produced under standard laboratory conditions. The present report is concerned with the measurement of bond strength.

Although considerable ingenuity has been used to develop reliable test methods in the past, no generally accepted method has been evolved Jones [1]. Recently, however, Elssner and Pabst [2] have successfully used a fracture mechanics approach to evaluate the bond between polycrystalline alumina and niobium foil using a four-point bend test specimen provided with a notch at the interface (see Fig. 1). As this approach should yield a single unambiguous parameter for the bond strength it was decided to investigate this method using a representative range of dental alloys. All alloys used in the project were cast under standard laboratory conditions in order that the test-pieces produced simulated normal production material.

2. Materials and methods

Three dental alloys representing the precious, semi-precious and non-precious alloys commercially available and an experimental alloy produced in collaboration with Johnson Matthey (Technology Centre, Blounts Court, Sonning Common, Reading) were used in conjunction with a single porcelain frit of one blend of Vita opaque body porcelain obtained from Vita Ltd (H. Rauter Gm. BH., Bad Sackingham, West Germany). This frit was of unknown composition, but generally the opaque bonding porcelains have the approximate composition 60% SiO_2, 20% Al_2O_3, 6% K_2O, 5% TiO_2, the remainder CaO and B_2O_3, included for liquid phase sintering.

The alloys examined and the casting parameters used were as shown in Table I.

All dental castings were carried out by the lost wax method, wax patterns for the coupons required for the single edged notched beam (SENB) specimen being cut from sheet wax nominally 0.25 mm in thickness. All samples were sprued with 4 mm diameter wax and invested in a mild steel casting ring lined with asbestos. A phosphate bonded investment was used (Jelenko Complete, Austenal Dental Products, Harrow, Middlesex). Mixing was carried out under partial vacuum for 1 min using a constant powder/liquid ratio of 60 g of powder to 11 mm^3 of liquid and vibrated into the mould, the wax pattern having previously been wetted with a proprietary wetting agent (Kerrs Debubblizer, Kerr UK Ltd, Peterborough). Moulds were allowed to set for 1 h prior to removing the rubber crucible former and scraping the glazed upper surface, which if left can cause gas entrappment leading to casting porosity. Wax burnout of the mould was undertaken at 350° C for 30 min prior to transfer of the mould to another oven for heat soaking for 1 h. Castings were all carried out centrifugally using the parameters given

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* *Present address*: Department of Restorative Dentistry, University College Dental School, Mortimer Market, London WC1E 6JD, UK.

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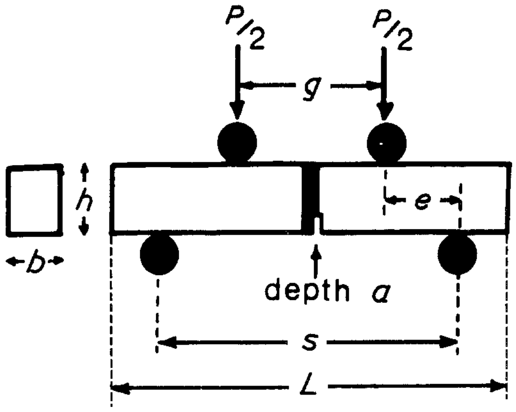

Figure 1 Four-point bend test specimen. $K_{Ic} = 3Pea^{1/2}\,[Y/bh^2]$. $Y = 1.99 - 2.47(a/h) + 12.97(a/h)^2 - 23.17(a/h)^3 + 24.8(a/h)^4$. Conditions $g \geqslant 2h$; $s \geqslant 4h$; $L \geqslant 4.2h$; $2 \leqslant h/b \leqslant 8$.

in Table I. The gold and silver–palladium alloys were melted using a heating coil and the higher temperature alloys by high frequency induction heating. The coupons after casting were glass bead blasted to remove investment remnants, sectioned, subjected to ultrasonic cleaning and finally washed in a degreasing agent, Inhibisol (Penetone Ltd, Northumberland) and water in preparation for porcelain bonding. All specimens were finished to 600 grade SiC paper, and oxidized under controlled conditions (Table II).

Jigs were constructed to enable the production of a block of the same depth and length as the SENB specimen, but of greater width from which individual specimens were obtained by the longitudinal sectioning. The porcelain was applied to the oxidized alloy coupon prior to its insertion in the location slots provided at 90° to the long axis. The remaining porcelain was then applied, vibrated and dried with cotton gauzes. After careful disassembly, further drying was allowed before insertion in the furnace for additional drying. An additional jig was used without a location slot for the preparation of the opaque porcelain bars.

Firing was carried out to the temperature–time cycles specified by the manufacturers and given in Table III with additional accurate temperature measuring equipment.

Initially in order to simulate normal dental production material, individual specimens were prepared and finished by grinding and lapping using a jig with hardened carbon steel faces. After considerable experimentation this procedure was abandoned in favour of the block technique detailed below, because of the poor reproducibility of the results, which persisted despite seemingly adequate controls in the firing, grinding, lapping and measurement stages.

Longitudinal sectioning to produce individual specimens was carried out on a diamond cutting machine with 6.5 inch diameter wheel and water cooling. The method developed for holding samples accurately, such that dimensional accuracy was obtained with a consistent surface finish which eliminated the necessity for further lapping, was as follows.

A brass stock was heated on a hot plate, dental sticky wax applied, and a glass microscope slide stuck to the surface followed by wax and a second slide. The specimen was then mounted with wax on the top side and the assembly allowed to cool. With this technique the specimens were rigidly mounted in the jaws of the cutter vice by the brass stock and could be cut through completely without movement and furthermore no brass was picked up on the cutting edges. Specimens were removed by heating on a hot plate and dissolving the wax with Inhibisol. Specimens were then washed with water and dried.

Notches of various widths located at one ceramo-metal interface were placed with the aid of a special jig which held a dental handpiece in which diamond discs with nominal thicknesses of 120, 200, 290 μm could be inserted. Notching was carried out at 5000 r.p.m. under water cooling.

Fracture toughness measurements were carried out on an Instron Universal Testing machine at 20 ± 2° C

TABLE I Alloy compositions and casting parameters

Alloy	Composition	Supplier	Mould heat soak (°C)	Crucible	Casting temperature (°C)
Au	88Au, 5Pt, 4Pd, 1.5In remainder Ag	Johnson Matthey Medical Ltd, Birmingham	850	Carbon	1180
Ag Pd	54Pd, 38Ag, 8Sn	Davis Schottlander & Davis, London EC1.	850	Carbon	1270
Ni Cr	67Ni, 13C, 7.5Ga, 7Mo remainder Fe	Howmedica International Ltd, London N16.	900	Ceramic	1485
Experimental	78Pd, 20Ag, 2Al	Johnson & Mattey Medical Ltd, Birmingham	850	Ceramic	1320

TABLE II Alloy oxidation conditions

Alloy	Furnace temperature at insertion (°C)	Rate of heating (°C min^{-1})	Maximum temperature (°C)	Hold time (min)	Bench cooled from (°C)
Au	400	200	1000	3	500
Ag Pd	400	200	1000	4	500
Ni Cr	400	200	1093	0	600
Experimental	400	200	1000	4	500

TABLE III Porcelain firing conditions

Alloy insert	Maximum temperature (° C)		Hold time (min)	Removed from furnace after slow cooling to (° C)
	Drying out	Firing		
Au	300	960	3	400
Ag Pd	300	960	3	400
Ni Cr	300	975	3	400
Experimental	300	960	3	400

and 0.5 mm min^{-1} cross-head speed using a specially constructed four-point loading rig designed as advocated by Pabst [3] which allowed accurate alignment to minimize uneven load distribution and torsional effects and reduced any frictional effects at the loading and support points. The jig dimensions were as follows: mean separation of support roller centres, 19 mm; mean separation of load roller centres 7 mm.

Each alloy-porcelain bond was prepared in two specimen sizes with different notch depths (Table IV). These were separated into two batches, one for testing without further treatment and the other for examining the effect on the bond strength of a suitable hot isostatic processing (HIP) treatment in which the pressure vessel filled with Argon at 100 MPa was taken up to 750° C, held at this temperature for 20 min and then allowed to cool. The duration of this temperature–time cycle was 2 h. This commercial cycle was selected as the most appropriate of those available for Vita porcelain with a nominal glass transition temperature (T_g) reported to be between 575–750° C depending on the cooling rate [4–6].

The fracture toughness K_{1c} was calculated from the standard formula given in Fig. 1 [7, 8].

3. Results and discussion

The concept that the strength of a bond between two dissimilar materials may be uniquely characterized by a single fracture toughness parameter is, for obvious reasons, a very attractive one. The validity of any particular test geometry for supplying this information, however, requires a thorough examination both theoretically and experimentally. Although a detailed stress analysis of the composite SENB specimen is not yet available, it is clear from first principles that the stress field in the crack tip region must be asymmetric because of the differences between the elastic constants of the two materials. This asymmetry although inevitable with any test geometry will be affected by the thickness of the alloy insert; for this reason this thickness was kept constant. The effect of alloy coupon thickness on the fracture toughness of such bonds will, nevertheless, require examination, for this may complicate any comparison of fracture toughness results with those obtained by other methods.

The test variables that have been examined are specimen size, and notch width. It should be noted that the notch depth also varied since there was no means of adjusting this parameter as the cutter width varied.

A summary of the fracture toughness results obtained is presented in Table IV for the Vita opaque porcelain alone and for the normal and hipped porcelain/alloy bonds. Statistical analysis showed that the K_{1c} results were independent of notch size for a given specimen size for all alloy bonds; the results for a given bond could therefore be pooled.

A linear regression analysis carried out on the K_{1c} results for different notch widths for the opaque porcelain alone showed that the slope of the regression line was significantly different from zero ($p = 0.022$). A one-way analysis of variance for K_{1c} against notch

TABLE IV Summary of fracture toughness results

Condition	Alloy–porcelain bond	Specimen size reference	Number of specimens	Mean K_{1c} (MN m$^{-3/2}$)	SD	Pooled means	Duncan grouping
Normal	Vita porcelain	1	18	2.1039	0.2883	2.1039	A
	Au	2	18	2.3697	0.5385	2.2199	A
	Au	4	14	2.0275	0.3818		
	AgPd	2	10	2.5627	0.5609	2.4070	A
	AgPd	4	12	2.2772	0.3817		
	NiCr	4	10	1.3529	0.3059	1.3529	B
	NiCr	3	10	2.0167	0.6173	RD*	
	Experimental	2	9	1.3861	0.3192	1.3861	B
	Experimental	3	9	2.3103	0.2705	RD	
Hipped	Au	2	6	2.4129	0.1674	2.6622	C
	Au	5	5	2.9614	0.2476		
	AgPd	2	5	2.6980	0.4356	2.8390	C
	AgPd	4	5	2.9799	0.3193		
	NiCr	4	5	2.0799	0.5288	2.0799	D
	Experimental	5	6	2.6214	0.2057	2.6214	C

*RD = Results discarded.

Specimen dimensions (height × breadth (mm)) size reference (1) 3.65 × 1.75; (2) 2.54 × 1.25; (3) 2.15 × 1.19; (4) 2.875 × 1.235; (5) 3.15 × 1.43.

Notch depth (mm) Normal. (1) 1.46; (2) 0.535; (3) 0.874; (4) 0.95.
Hipped. (2) 0.86; (4) 1.12; (5) 1.95.

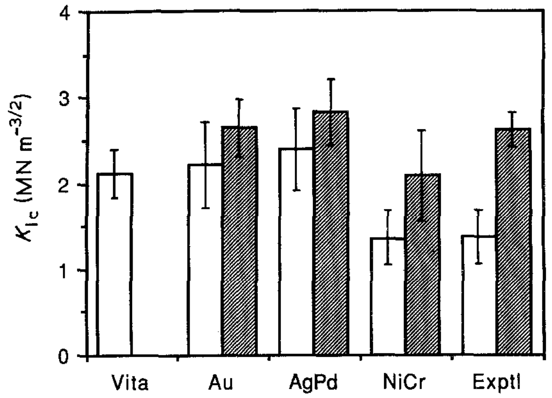

Figure 2 Fracture toughness of normal and hipped alloy bonds.

width, however, showed that there was no significant difference between the results for different notch widths, and this was considered sufficient justification for pooling these results.

The finding that the K_{Ic} was independent of notch width was unexpected, since such a dependency has been reported in the literature for a notch beam test when used on ceramic materials [9–11] and overcome by extrapolating the linear regression line for K_{Ic} on notch width to obtain the fracture toughness at zero notch width.

Duncan's multiple range analysis indicated that there was no significant difference between the mean K_{Ic} results for specimen sizes 2 and 4 for the normal Au or AgPd bonds, so that the results for each of these bonds could be pooled. The analysis showed, however, that there was a statistically significant difference between sizes 3 and 4 for the NiCr and sizes 2 and 3 for the experimental alloy bonds. On further examination it was found that (due to an error) the size 3 specimens were slightly below the accepted lower limit for the breadth : height ratio for the SENB test-piece. If the size 3 results are eliminated from the analysis on these grounds, the results for the normal bonds fall into two groups with the porcelain, Au and AgPd bonds superior to the NiCr and experimental alloy bonds, there being no significant difference between the materials in either group.

The number of specimens subjected to the HIP treatment had to be restricted and, in addition, a number of specimens failed in the process. The greatest incidence of failure was found for the NiCr bonds, where approximately half the specimens were lost.

A comparison of the normal and hipped bond pooled mean results is given in Fig. 2, where the hipped results are shown hatched. Once again analysis showed that the results were independent of specimen size (Au and AgPd bonds only) so that the results for each of these two alloy bonds could be pooled. Ignoring, as before, the size 3 results for the experimental alloy, it may be seen that the HIP treatment has significantly improved the fracture toughness of all bonds. Further analysis using Duncan's multiple range test showed that the hipped results fell into two groups, with the NiCr significantly lower than the other three alloys.

Figure 3 SEM of Au alloy bond (normal) fracture surface.

The drawback of all fracture tests in which once a crack is initiated catastrophic rupture occurs is that the process of rupture is controlled by the largest flaw in the notch tip region. The grouping together by Duncan's multiple range test in the present study of the porcelain with Au and AgPd bonds before hipping inevitably poses the possibility that the measurements are merely reflecting the fracture strength of the porcelain rather than that of the bond itself.

An attempt was therefore made to categorize the failure modes of all bonds by eye and with low power optical microscopy. The summarized results are shown in Table V. The predominant mode of failure of the normal bonds in the higher range of strengths using Duncan's test was through the oxide–porcelain boundary, with some isolated small areas of porcelain cohesive failure which increased in density towards

TABLE V Failure modes of alloy porcelain bonds (percentage of total number tested)

Mode	Alloy							
	Au		AgPd		NiCr		Experimental	
	N*	H	N	H	N	H	N	H
Alloy oxide	–	–	–	–	15	100	5.5	–
Oxide	3.1	–	–	–	–	–	–	–
Oxide porcelain	81.2	100	81.8	100	70	–	55.5	100
Porcelain	9.4	–	18.2	–	15	–	35	–

*N = Normal; H = hipped.

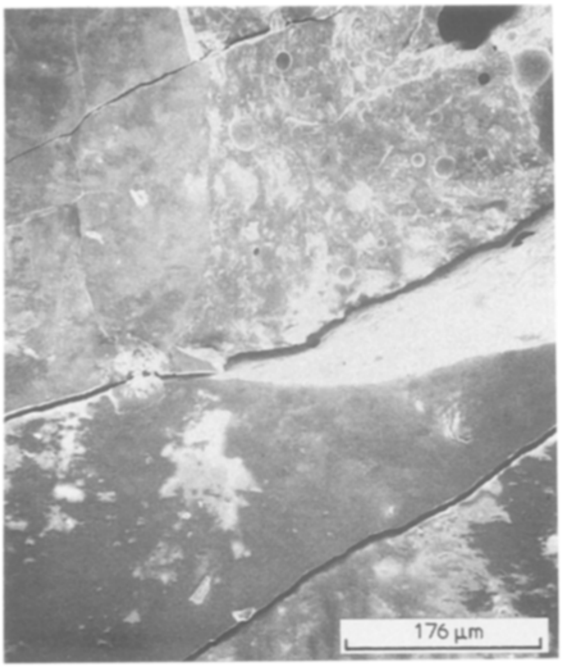

Figure 4 SEM of AgPd alloy bond (normal) fracture surface.

the upper surface of each specimen. For the bonds shown by the statistic to be in the lower strength range, i.e. the NiCr and experimental bonds, failure occurred most frequently at the alloy–oxide interface and seemed to be associated with a higher incidence of microporosity than was present with the bonds in the superior group. After hipping, fracture at the oxide–porcelain interface was the sole failure mode except for the NiCr bonds which all exhibited failure at the alloy–oxide boundary. SEM examination of the fracture surfaces confirmed that with the normal bonds there was considerable porosity at the interface, as may be seen in Figs 3, 4 and 5 for the Au, AgPd and NiCr alloy bonds, respectively, which are all taken at relatively low magnification.

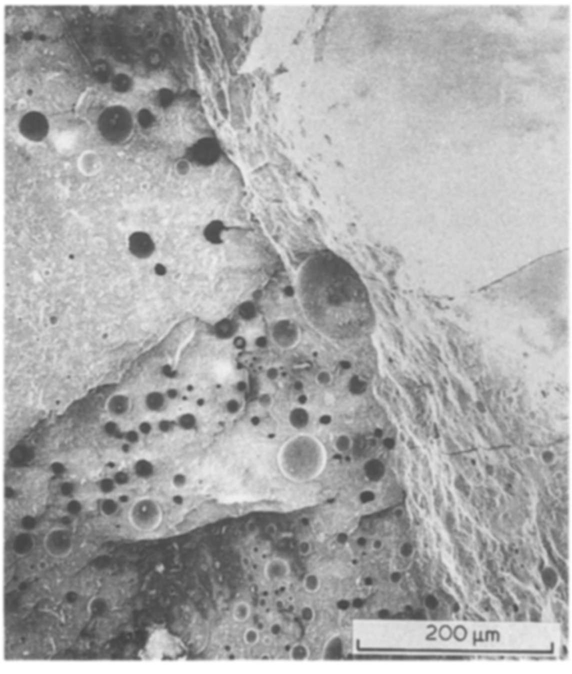

Figure 5 SEM of NiCr alloy (normal) fracture surface 7.6 cm wide × 8.8 cm deep.

Figure 6 SEM of Au alloy bond (hipped) fracture surface.

SEM examination of the fracture surfaces of the hipped specimens suggested that this treatment had produced, in the main, a reduction of porosity in the bond region. For example, Fig. 6 shows the fracture surface of the Au alloy bond at a magnification of × 350 where only a few pores are visible. As might be expected the hipping treatment caused a densification of the porcelain. The appearance of examples of the normal and hipped porcelain fracture surfaces at the same magnification (× 200) are shown in Figs 7 and 8.

An increase, both in density and fracture toughness in microfine alumina after a HIP treatment has been observed by Sclosa *et al.* [12]. It was suggested that this implied that radial creep, occurring by grain-boundary sliding had filled voids and shortened cracks left by the prefiring process. It seems reasonable that

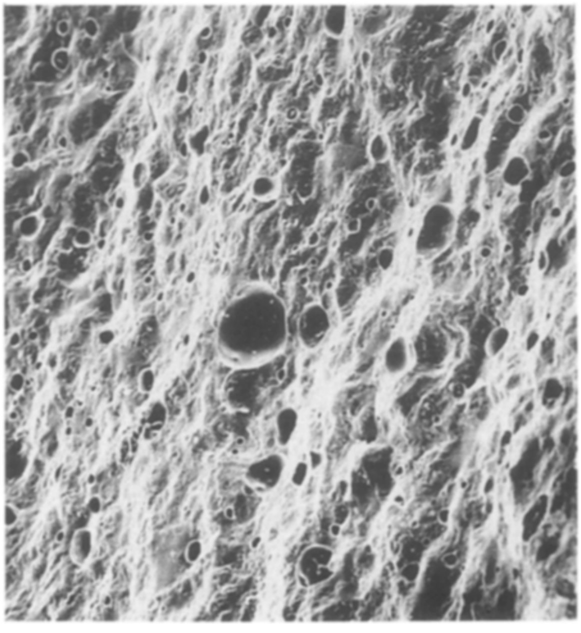

Figure 7 SEM of porcelain (normal) fracture surface. 7.8 cm wide × 8.3 cm deep.

Figure 8 SEM of porcelain (hipped) fracture surface.

the same mechanism is responsible for the improvement obtained with the dental porcelain.

The high failure rate during hipping is tentatively attributed to the ingress of argon under pressure through open surface pores in the interfacial region. Prior coating of the bond with a low fusing porcelain was found to restrict this type of failure. The average coefficient of variation obtained without hipping by the methods developed was 20 and 22% for the Au and AgPd bonds and 35 and 29% for the weaker NiCr and experimental bonds. The coefficient of variation was reduced by the HIP treatment to 13.7, 13.0, 7.8 and 25.4%, respectively. These figures compare favourably with other data in the literature for bond tests as opposed to fracture of porcelain alone.

It is important to note that the commercial HIP cycle used was chosen as the most appropriate of those available with regard to the reported glass transition temperature range of the porcelain used and may not necessarily be the pressure–temperature–time cycle required for maximum enhancement of the bond strength. If the hypothesis that failure of some specimens during the hip cycle is caused by the ingress of argon under pressure through open pores in the interfacial region is correct, it is possible that an initial heating treatment under evacuation might prove beneficial. Clearly the rate of cooling adopted in the final part of the cycle will affect the relief of any residual stresses present due to thermal mismatch between the alloy and the porcelain used.

Finally, it is felt that the difficulty experienced in the project in obtaining reliable results by preparing individual specimens for the SENB test piece, which has not been previously commented upon in the literature, may indicate either that the fracture toughness is extremely sensitive to minor temperature variations during the firing process or perhaps more significantly to the influence of a mixed fracture stress involving both modes I and III due to slight non-parallelism of the upper and lower faces of the specimen producing a torsional element to the stress at the notch on loading.

4. Conclusions

On the basis of the results reported the SENB test method appears to warrant serious consideration as a means of providing reliable comparative data on the fracture toughness (K_{Ic}) of ceramo–metal bonds. The K_{Ic} values obtained were independent of notch width and specimen size over the range of these variables examined and showed good discrimination. Further examination of the effects of specimen size, alloy coupon thickness and notch depth is clearly necessary. The K_{Ic} of all bonds as measured by this method was improved by a commercial HIP treatment and the coefficient of variation of all bonds thus treated was improved. SEM examination of the fractured surfaces strongly suggested that this improvement was due to a reduction in porosity at the interface.

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