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Research Paper

Mini-interfacial fracture toughness as a new validated enamel-bonding effectiveness test

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ABSTRACT

Today's most commonly applied bonding effectiveness tests are criticized for their high variability and low reliability, the latter in particular with regard to measuring the actual strength of the adhesive interface. Objectives: in continuation of previous research conducted at dentin, we hereby aimed to validate the novel mini-interfacial fracture toughness (mini-iFT) test on its applicability to assess bonding effectiveness of contemporary adhesives when bonded to enamel. Methods: The 3-step etch&rinse (E&R) adhesive OptiBond FL (Kerr), the 2-step self-etch (SE) adhesive Clearfil SE Bond (Kuraray Noritake) and the two multi-mode adhesives Clearfil S^3 Bond Plus (Kuraray Noritake) and Scotchbond Universal (3M ESPE), both used following a 2-step E&R and 1-step SE mode, were applied to clinically relevant, flattened enamel surfaces. A composite (Filtek Z100; 3M ESPE) build-up was made in layers. After 1-week water storage at 37 °C, all specimens were sectioned perpendicular to the interface to obtain rectangular sticks. A mini-iFT notch was prepared at the adhesive-enamel interface using a thin diamond blade under water cooling. Finally, the specimens were loaded in a 4-point bending test until failure. Results: the mini-iFT onto human enamel was significantly higher for the adhesives applied in E&R mode versus those applied in SE mode. The lowest mini-iFT was found for the adhesives applied following a 1-step SE approach. SEM fracture analysis revealed that all fractures originated at the adhesive-enamel interface and that the induced crack propagated preferentially along this interface. Conclusion: mini-iFT appeared a valid alternative method to assess the mechanical properties of adhesive-enamel interfaces.

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1. Introduction

Assessing bonding effectiveness of adhesives to enamel is commonly done using either macro-shear (Braga et al., 2010), micro-shear (μSBS) (Walter et al., 2011; Meharry et al., 2013; Tedesco et al., 2014; McLean et al., 2015) or micro-tensile bond-strength (μTBS) testing (Hanabusa et al., 2012; De Munck et al., 2013; de Goes et al., 2014; Yaman et al., 2014). Such bond-strength data generally vary widely, mainly because of non-uniform stress distribution and/or stress that not always is concentrated at the actual adhesive-enamel interface. Hence, a strong demand exists for a fracture mechanics approach (Scherrer et al., 2010; Soderholm, 2010), so to measure interfacial fracture toughness (iFT) that better reflects the interfacial properties. However, specimen preparation for iFT testing is commonly regarded as more laborious, time consuming and technique sensitive.

iFT testing can be done in various ways, this using a short rod chevron notch (Tam and Pilliar, 1993; Armstrong et al., 1998), a notchless triangular prism (Far and Ruse, 2003) or a chevron notch beam (CNB) (De Munck et al., 2013, 2015). In 2013, we designed a CNB iFT test; a significant ($p=0.011$) correlation of 92% was found between the CNB iFT data and the data obtained with the today most popular μTBS test (De Munck et al., 2013). However, a major disadvantage of this conventional iFT test method is that it requires a relatively large interfacial area. Therefore, a new mini-iFT test was recently developed and validated on its potential to assess bonding effectiveness to dentin (Pongprueksa et al., 2016); specimen preparation was found to be easier, entailing a much smaller interfacial area. Moreover, a significant ($p<0.001$) correlation of 80% was found between the mini-iFT data obtained at dentin and the corresponding μTBS data; the method revealed a higher discriminative power and appeared to generate more reliable data associated with failure at the actual interface. Hence, this mini-iFT test appeared also promising to assess bonding effectiveness to enamel, in particular since the interfacial area is four times smaller than that of the conventional iFT test.

A new trend in dental adhesive technology is 'universal', enabling the dentist to use one and the same adhesive for direct and indirect restorative procedures, but also to opt for either an 'etch-and-rinse' (E&R) or 'self-etch' (SE) approach (Chen et al., 2015). Predating the latter universal trend, the extension of a one-step self-etch adhesive into a multi-step adhesive revealed a slightly improved bonding effectiveness when a hydrophobic bonding agent was additionally used; adding a preceding etching step was beneficial for enamel but should be avoided for dentin as it decreased bond strength and even jeopardized bond durability for the particular adhesive that was tested (Van Landuyt et al., 2006a, 2006b). Laboratory and clinical evidence today favors a full three-step E&R approach or an alternative three-step protocol consisting of selective enamel etching followed by a mild two-step SE approach (Van Meerbeek et al., 2003, 2010, 2011; Peumans et al., 2005a, 2005b, 2010, 2014; Van Meerbeek and Yoshihara, 2014). Essential with regard to the latter 'mild' SE bonding approach to dentin is the use of an adhesive that contains a functional monomer with a high chemical affinity to hydroxyapatite (Yoshihara et al., 2010, 2011). Among different monomers investigated, 10-MDP (10-methacryloyloxidecyl dihydrogen phosphate) appeared most effective, but is still regarded as being sensitive to hydrolysis (Salz et al., 2005; Van Landuyt et al., 2008). Very striking is that most of the today commercially available 'universal' adhesives contain 10-MDP as functional monomer (Chen et al., 2015; Munoz et al., 2015; Rosa et al., 2015). While 10-MDP has principally been regarded of importance with regard to bond durability thanks to its intense chemical interaction with HAp, recent NMR spectroscopy also revealed a relatively stable interaction of 10-MDP with dentinal collagen (Hiraishi et al., 2013). Hence, the 10-MDP-containing universal adhesives appear also to benefit from 10-MDP-collagen complexation when applied following the optional E&R bonding protocol.

In literature, to our knowledge only one study applied iFT onto enamel (Tam and Pilliar, 1993); they used bovine teeth, on which a much larger enamel area is available than in case of human teeth. In light of the recently developed mini-iFT test (Pongprueksa et al., 2016), we aimed with this study to determine the mini-iFT of adhesive-enamel interfaces prepared using dental adhesives representing the different adhesive classes, in a similar way as in our previous study. The hypothesis advanced was that no difference in mini-iFT at enamel exists for the different adhesives tested.

2. Materials and methods

2.1. *Mini-iFT*

Thirty non-carious human premolars (collected following informed consent approved by the Commission for Medical Ethics of KU Leuven under the file number S57622) were stored in 0.5% chloramine T/water at 4°C and used within 3 months after extraction. Each crown was divided into a mesial and distal part, after which the root of the tooth was removed at the dentin–enamel junction with a slow-speed diamond saw (Isomet 1000; Buehler, Lake Bluff, IL, USA). The flat enamel surface was wet-sanded with 320-grit SiC paper (Buehler-Met II; Buehler) to produce a standard smear layer with a surface topography resembling that of bur-cut enamel (Pashley et al., 1988). All prepared enamel surfaces were carefully inspected using a stereo-microscope (Stemi 2000 CS; Carl Zeiss, Jena, Germany). A three-step E&R adhesive (3-E&Ra: OptiBond FL; Kerr, Orange, CA, USA), a two-step SE adhesive (2-SEa: Clearfil SE Bond; Kuraray Noritake, Tokyo, Japan), and two 'universal' adhesives (Clearfil S^3 Bond Plus; Kuraray Noritake; Scotchbond Universal; 3M ESPE, Seefeld, Germany), applied either following a 2-step E&R (2-E&Ru) or 1-step SE (1-SEu) mode (Table 1), were selected; they were applied following the respective manufacturer's instruction and light-cured using a polywave LED light-curing unit (Bluephase 20i; Ivoclar Vivadent, Schaan, Liechtenstein) with an output of around 1100 mW/cm^2, as measured by a MARC Patient Simulator (BlueLight Analytics, Halifax, NS, Canada). A composite (Filtek Z100; 3M ESPE: shade A2, lot N459523) build-up was made in layers using a polytetrafluoroethylene (Teflon) mold ($6\times8\times6$ mm); a similar composite build-up was built at the pulpal side using the SE adhesive Clearfil SE Bond. After 1-week water storage at 37°C, the specimens were sectioned perpendicular to the interface using a semi-automated diamond saw (Accutom-50; Struers, Ballerup,

Table 1 – List of materials tested.

Adhesive	Manufacturer	Adhesive class	Application method	Lot number
OptiBond FL	Kerr, Orange, CA, USA	3-step E&R (3-E&Ra)	Application of phosphoric-acid gel etchant on enamel for 15 s, followed by rinsing for 15 s and gentle air-drying.	Gel etchant: 5007742
			Rubbed application of primer for 15 s, followed by 5-s air-drying.	Primer: 5002746
			Application of adhesive, followed by 10-s light-curing.	Adhesive: 5009539
Clearfil SE Bond	Kuraray Noritake, Tokyo, Japan	2-step SE (2-SEa)	Rubbed application of primer for 20 s, followed by 5-s mild air-drying.	Primer: 6J0038
			Application of the adhesive, followed by gentle air-drying and 10-s light-curing.	Bond: 6k0063
Clearfil S^3 Bond Plus	Kuraray Noritake	'UNIVERSAL' used in: 2-step E&R mode (2-E&Ru) 1-step SE mode (1-SEu)	Application of phosphoric-acid gel etchant on enamel for 15 s, followed by rinsing for 15 s and gentle air-drying.	Gel etchant: 5007742
			Rubbed application of the adhesive for 10 s, followed by 5-s gentle air-drying and 10-s light-curing (solely this step in case of 1-SEu).	Adhesive: 00012A
Scotchbond Universal	3M ESPE, Seefeld, Germany	'UNIVERSAL' used in: 2-step E&R mode (2-E&Ru) 1-step SE mode (1-SEu)	Application of phosphoric-acid gel etchant on enamel for 15 s, followed by rinsing for 15 s and gentle air-drying.	Gel etchant: 5007742
			Rubbed application of the adhesive for 20 s, followed by 5-s gentle air-drying and 10-s light-curing (solely this step in case of 1-SEu).	Adhesive: 514260

E&R=etch&rinse; SE=self-etch.

Denmark) to obtain one or two rectangular sticks (1.5 × 2.0 mm wide; 14–16 mm long).

A single mini-iFT notch tip was prepared under a stereo-microscope (Leica M715; Wetzlar, Germany) at the adhesive-enamel interface using a water-cooled diamond saw mounted with an ultra-thin 150 μm diamond blade (M1DO8; Struers) and set at a feed speed of 0.015 mm/s and wheel speed of 1,000 rpm. The notch width was less than 0.3 mm and incorporated the adhesive-enamel interface. The mini-iFT notch tip angle was 45 degrees. The tip of the single-notch mini-iFT was located on the long surface at 0.24–0.48 mm from the bottom left corner (Fig. 1). The opposite part of the mini-iFT notch did end at less than 0.2 mm from the top back side. The specimen was immediately transferred to the universal testing machine (5848 Micro Tester; Instron, Norwood, MA, USA), placing the specimen with the notch tip down in the test fixture. The specimen was tested in a 4-point bending test setup with a crosshead speed of 0.05 mm/min; the outer and inner span were 10 and 5 mm, respectively. After testing, all fractured surfaces were processed for SEM (JSM-6610LV; JEOL, Tokyo, Japan) using common specimen processing, including fixation, dehydration and gold-sputter coating, this to determine the fracture location, the crack propagation and possible specimen imperfections. Finally, the exact dimensions of the mini-iFT notch were measured using a measuring optical microscope (400-NRC; Leitz, Wetzlar, Germany) at 250 × magnification, after which K_{Ic} was calculated in MPa $m^{1/2}$, as described in detail in the previous study (Pongprueksa et al., 2016).

2.2. *Finite element analysis (FEA)*

A finite element model of the mini-iFT specimen was created in Abaqus 6.11 (Simulia, Providence, RI, USA). The adhesive layer was assumed to be 30 μm thick (Pongprueksa et al., 2008). The element size in the adhesive layer was 15 μm, as well as in the adjacent enamel and composite, and gradually increased to 285 μm at both ends of the specimen, this to limit computational cost. The total number of quadratic hexahedral elements (C3D20 element in Abaqus) and nodes of the model was 7186 elements and 39,985 nodes. The contact interfaces between each part of the model (dentin–enamel-adhesive-composite interface) were assumed to be fully bonded to each other; as such, no sliding was allowed.

Boundary conditions were set according to the mechanical testing conditions. At the bottom of the specimen, all nodes located at 5 mm from the adhesive-enamel interface were assumed to be fully constrained to mimic the support span of 10 mm. The total compression force of 30 N was applied perpendicular to the specimen at 2.5 mm from the adhesive-enamel interface to form the loading span of 5 mm. The material properties of dentin, enamel, composite and adhesive were, respectively, 25.1, 46.8, 24.4 and 8.4 GPa for modulus of elasticity and, respectively, 0.31, 0.3, 0.25 and 0.35 for Poisson's ratio, these as according to previous studies (Wright and Yettram, 1979; Pongprueksa et al., 2008; Soderholm et al., 2012). The maximum principal stress in tensile, compression and shear, as imposed to the adhesive layer at the adhesive-enamel interface, was analyzed.

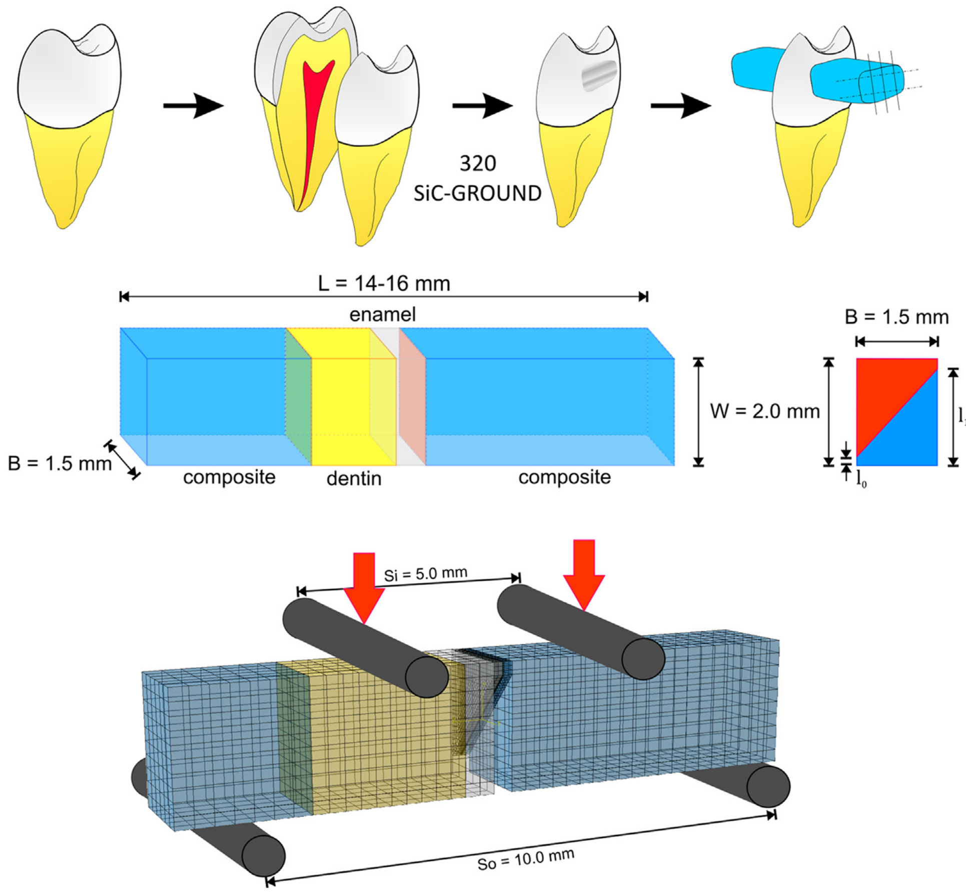

Fig. 1 – Schematic explaining the specimen-preparation methodology and test setup for measuring the mini-iFT to enamel.

2.3. *Statistical analysis*

The mini-iFT data were statistically analyzed by Weibull analysis (Quinn and Quinn, 2010); pivotal confidence bounds were calculated using Monte Carlo simulation (Symynck and De Bal, 2011). The different groups were compared at the 10% unreliability level and at the characteristic strength (63.2% unreliability). All tests were performed at a significance level of $\alpha=0.05$ using a software package (R3.01 and abrem package, R Foundation for Statistical Computing, Vienna, Austria).

3. Results

The mini-iFT data are summarized in Table 2 and graphically presented in Fig. 2. The mini-iFT at the Weibull characteristic strength (63.2% unreliability) revealed a significantly higher value for the adhesives applied in E&R mode than in SE mode. The mini-iFT of the 3-E&Ra (Optibond FL) was similar to that of the 2-E&Ru adhesives (Clearfil S^3 Bond Plus in E&R mode, Scotchbond Universal in E&R mode). The mini-iFT of the 2-SEa (Clearfil SE Bond) was higher than that of the 1-SEu adhesives (Clearfil S^3 Bond Plus in SE mode, Scotchbond Universal in SE mode). The Weibull modulus was excessively high (16.14) for Clearfil S^3 Bond Plus when applied in SE mode, despite the Weibull modulus of all other adhesives varied between 6.32 and 8.75.

SEM failure analysis of representative mini-iFT specimens are presented in Figs. 3 and 4. Overall, failure analysis disclosed that the mini-iFT test resulted in failures at the actual adhesive-enamel interface. More specifically, the notch tip of all mini-iFT specimens failed at the adhesive-enamel interface; enamel prisms could be observed at higher magnification (Figs. 3,3b and 4,4b: hand pointer). For the SEa/u, the crack propagated along the adhesive-enamel interface; near the end of the notch, however, the crack often deviated towards the adhesive layer. This fracture profile differed from that recorded for the E&Ra/u, which also presented with a higher mini-iFT; then, failure began at the adhesive-enamel interface, but the crack propagated through the adhesive and composite layer from about the middle of the notch.

The FEA results are presented in Fig. 5. The distribution of principal stress disclosed that high tensile stress was concentrated at the notch tip, while compressive stress was imposed towards the end of the notch. The maximum tensile stress within the specimen was considerably higher than the maximum compressive stress. Overall, the amount of shear stress was minimal (not shown). At the same location along the notch tip, the tensile stress at the interface was higher at the adhesive-enamel interface than at the adhesive-composite interface.

4. Discussion

Bonding effectiveness of adhesives to human enamel was assessed using a new mini-iFT test; the test results confirmed

Table 2 – Mini-interfacial fracture toughness (mini-iFT) at enamel.

ADHESIVE[1]	Mean (K_{Ic}) mini_iFT[2]	SD	B[3](m)	η[4]	b10[5]	Characteristic strength[6]	ptf/n
OptiBond FL (3-E&Ra)	2.15	0.38	6.32	2.30	1.61(1.23–1.91)[a,b]	2.30(2.10–2.53)[a]	0/15
Clearfil S[3] Bond Plus (2-E&Ru)	2.29	0.43	6.73	2.44	1.75(1.26–2.08)[a]	2.44(2.20–2.71)[a]	0/11
Scotchbond Universal (2-E&Ru)	1.95	0.29	7.79	2.07	1.55(1.12–1.82)[a,b]	2.07(1.87–2.30)[a]	0/9
Clearfil SE Bond (2-SEa)	1.63	0.23	8.75	1.72	1.33(1.09–1.50)[a,b]	1.72(1.61–1.84)[b]	0/15
Clearfil S[3] Bond Plus (1-SEu)	1.19	0.09	16.14	1.23	1.07(0.95–1.14)[b,c]	1.23(1.19–1.28)[c]	0/14
Scotchbond Universal (1-SEu)	1.16	0.22	6.90	1.24	0.89(0.70–1.04)[c]	1.24(1.14–1.35)[c]	0/15

[1] E&Ra=etch&rinse adhesive; SEa=self-etch adhesive; E&Ru=E&R universal; SEu=SE universal.
[2] Mini-interfacial fracture toughness K_{Ic} in $MPam^{1/2}$.
[3] Beta, shape, slope or modulus of Weibull parameter.
[4] Eta, characteristic life or scale of Weibull parameter.
[5] Estimation and 95% confidence interval at 10% probability of failure; groups with the same superscript letter are statistically not different.
[6] 95% confidence interval at characteristic strength (=63.2% unreliability); groups with the same superscript letter are statistically not different.

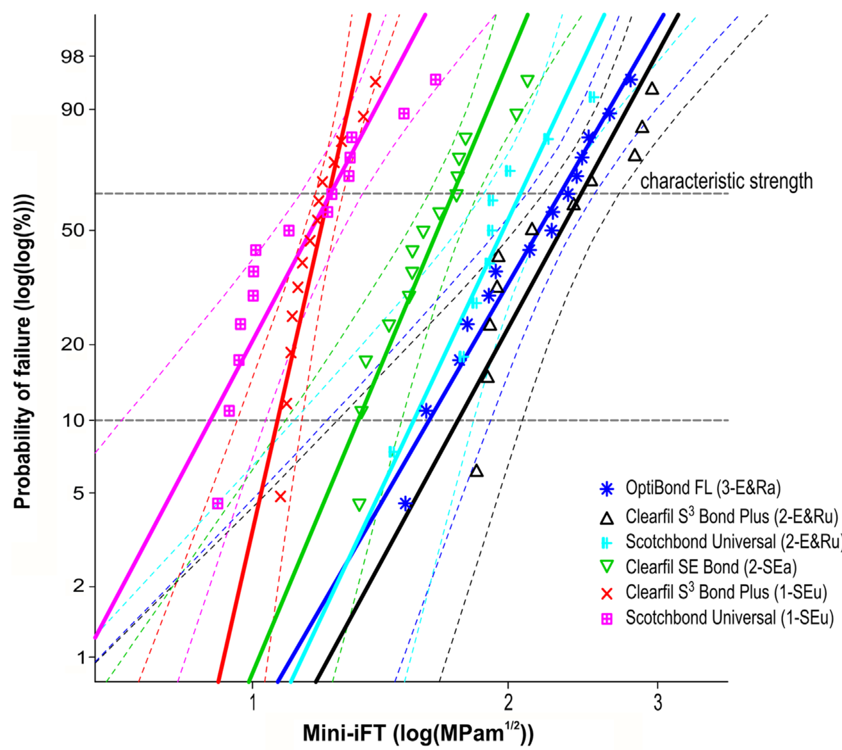

Fig. 2 – Weibull plot of the mini-interfacial fracture toughness (mini-iFT) data. The dotted lines represent the 95% confidence bounds as calculated from Monte-Carlo simulation.

that in terms of bonding effectiveness enamel is best etched with phosphoric acid following an E&R bonding protocol: a significantly higher mini-iFT was recorded than for a (mild) SE bonding protocol. Following an E&R mode, no difference in mini-iFT was found between a 3- and a 2-step E&R approach, while 2-SEa presented with a significantly higher mini-iFT at enamel than 1-SEu.

The new mini-iFT test was before validated to assess bonding effectiveness to dentin (Pongprueksa et al., 2016); this study now proved that the mini-iFT test can also differentiate among different adhesive approaches on their bonding effectiveness to enamel. The finding that an E&R approach is more effective at enamel than a SE bonding protocol correlates well with previous enamel bonding-effectiveness studies (El Zohairy et al., 2010; Juloski et al., 2012; Schlueter et al., 2013; de Goes et al., 2014). Other studies that employed μSBS or μTBS test protocols were however not always able to distinguish between both approaches (El Zohairy et al., 2010; Walter et al., 2011; Meharry et al., 2013; Tedesco et al., 2014; Yaman et al., 2014). This study also demonstrated that multi-mode or universal adhesives, which up to the dentist's choice can be applied following either an

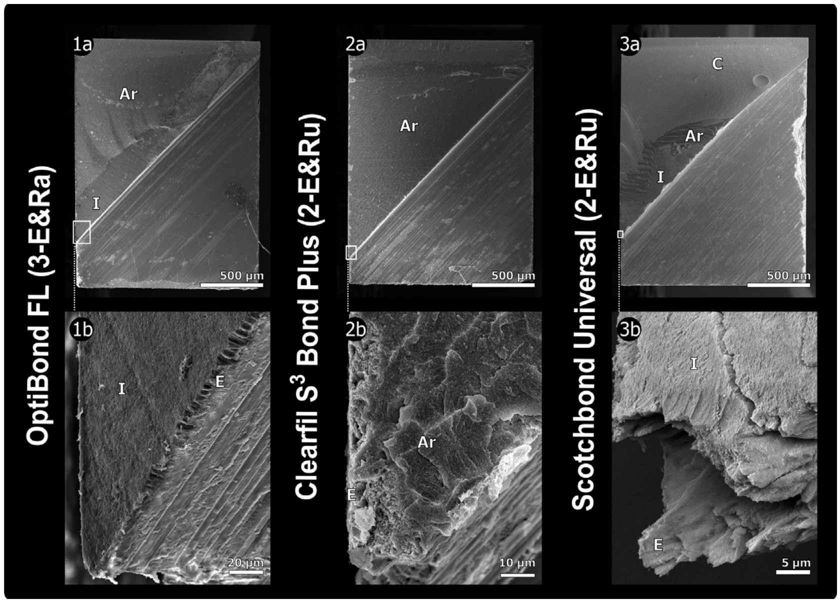

Fig. 3 – SEM fracture-surface analysis of representative mini-iFT specimens prepared following an etch&rinse (E&R) approach applied onto enamel. Ar: Adhesive resin; C: Composite; E: Enamel; I: Interface. (1a) Overview photomicrograph of the fractured surface of a specimen prepared using the 3-step E&R adhesive (OptiBond FL, Kerr). Despite the notch tip was positioned and initiated at the interface (I), the crack deviated away from the adhesive-enamel interface (I) through the adhesive resin (Ar). (1b) High-magnification photomicrograph of the notch tip from an oblique angle at 45 degrees, showing the notch configuration in detail and that the fracture was initiated exactly at the tip. (2a) Overview photomicrograph of the fractured surface of a specimen prepared using a multi-mode adhesive applied as a 2-step E&R adhesive (Clearfil S^3 Bond Plus, Kuraray Noritake). The entire specimen was covered with the adhesive resin (Ar). (2b) High-magnification photomicrograph of the notch tip; the fracture initiated exactly at the adhesive-enamel interface. Subsequent failure occurred within the thin adhesive resin (Ar), but very close to the interface with enamel. (3a) Overview photomicrograph of the fractured surface of a specimen prepared using the universal adhesive applied as a 2-step E&R adhesive (Scotchbond Universal, 3M ESPE). The crack initiated at the interface (I) and then deviated through the adhesive resin (Ar) and composite (C). (3b) High-magnification photomicrograph of the notch tip using backscatter electron imaging from an oblique angle at 45 degrees, confirming the presence of exposed enamel (E).

E&R or SE mode, benefit most from phosphoric-acid etching of enamel; to effectively bond to enamel, universal adhesives are best applied following an E&R mode.

In general, adhesion to enamel is regarded as less challenging than to dentin and mainly relies on micro-mechanical interlocking of resin within enamel micro-retentions (Van Meerbeek et al., 1996). Modern SE adhesives were documented to benefit also from additional chemical interaction of the functional monomer with hydroxyapatite. Despite the proven chemical interaction potential of the functional monomer MDP with HAp (Yoshida et al., 2004; Yoshihara et al., 2010, 2011, 2013; Zhang et al., 2013), the SE bonding protocol appeared insufficient to reach a mini-iFT at enamel in line with that achieved with an E&R bonding protocol. The three most plausible reasons are: (1) the enamel bond depends largely on micro-mechanical interlocking in deep etch pits created by phosphoric-acid etching, while the etching capacity of functional monomers like MDP is insufficient to achieve adequate micro-retention at enamel; (2) MDP appears less capable of reaching/interacting with Ca within enamel HAp that has a higher crystallinity and is less crisscross oriented than dentinal HAp; at enamel, despite its higher mineral content, less MDP-Ca salt is produced than at dentin (Yoshihara et al., 2011; Yokota and Nishiyama, 2015); (3) the mild self-etch bonding is compromised by the enamel-smear complex, the latter being effectively removed by phosphoric-acid etching and thus not interfering with bonding in case the more surface-aggressive E&R approach is applied (Mine et al., 2010). Bond-strength testing comparing MDP-free with MDP-containing adhesives showed nevertheless that chemical interaction of MDP with HAp, while perhaps less relevant at short time, is crucial in maintaining interfacial stability over time. Very recent research confirmed this even in case an E&R approach is employed (Tsuchiya et al., 2016).

Comparing the multi-step SE with the single-step SE approach, this study showed that the simplified one-step application procedure coincides with a significant decrease in enamel mini-iFT. This is in agreement with previous studies that assessed the mechanical properties of adhesive-enamel interfaces (Beloica et al., 2010; Walter et al., 2011; Hanabusa et al., 2012; Goracci et al., 2013; Meharry et al., 2013; de Goes et al., 2014; McLean et al., 2015), although some conventional bond-strength studies reported similar values for 2-SEa and 1-SEa (Reis et al., 2013; de Goes et al., 2014).

Studies correlatively using μSBS and μTBS to assess bonding effectiveness to enamel concluded that μSBS is more accurate to differentiate adhesives on their bonding potential to enamel than μTBS (Ishikawa et al., 2007; Beloica et al., 2010; El Zohairy et al.,

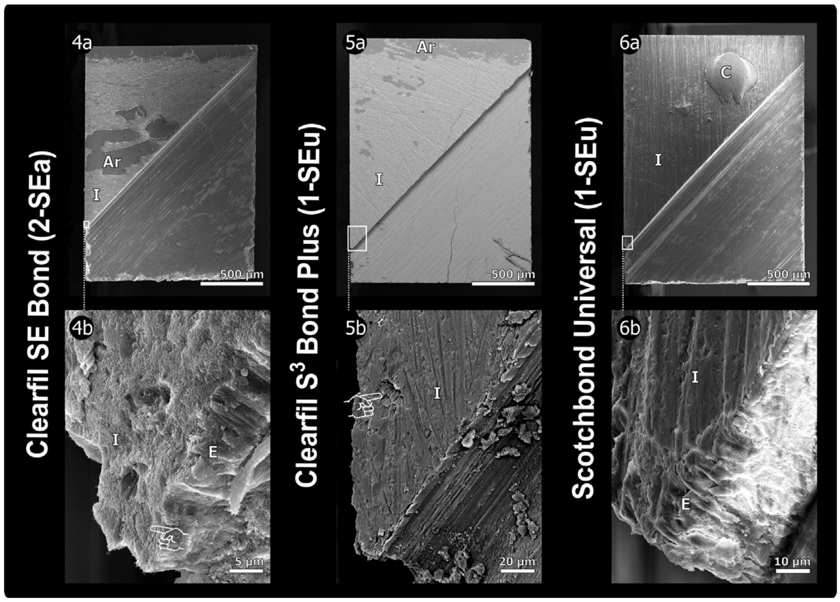

Fig. 4 – SEM fracture-surface analysis of representative mini-iFT specimens prepared following a self-etch (SE) approach applied onto enamel. Ar: Adhesive resin; C: Composite; E: Enamel; I: Interface. (4a) Overview photomicrograph of the fractured surface of a specimen prepared using the 2-step SE adhesive (Clearfil SE Bond, Kuraray Noritake). Despite the specimen initially fractured along the adhesive-enamel interface (I), the crack deviated towards the adhesive resin (Ar). Further on, the crack however returned to the interface, which may be a sign of favorable stress distribution during the test. (4b) High-magnification photomicrograph of the notch tip imaged from an oblique angle at 45 degrees; the fracture initiated exactly at the adhesive-enamel interface and exposed enamel prisms (hand pointer). (5a) Overview photomicrograph of the fractured surface of a specimen prepared using a multi-mode adhesive applied as a 1-step SE adhesive (Clearfil S^3 Bond Plus, Kuraray Noritake) using backscatter electron imaging. Although the entire specimen failed at the adhesive-enamel interface (I), the crack deviated towards to the adhesive resin (Ar) near the end of the specimen. (5b) High-magnification photomicrograph of the notch tip imaged from an oblique angle at 45 degrees, showing the sharp notch and that the specimen fractured exactly at the adhesive-enamel interface (I) close to the enamel substrate (hand pointer). (6a) Overview photomicrograph of the fractured surface of a specimen prepared using a universal adhesive applied as a 1-step SE adhesive (Scotchbond Universal, 3M ESPE). Despite the entire specimen failed at the adhesive-enamel interface (I), the crack deviated towards the composite (C), and then returned to the interface towards the end of the notch. (6b) High-magnification photomicrograph of the notch tip imaged from an oblique angle at 45 degrees, illustrating that the fracture initiated at enamel (E) and the adhesive-enamel interface (I).

2010). Reasons advanced are that, as compared to μSBS, a μTBS approach often results in a large amount of pre-testing failures, for instance varying in one study from 28–68.3% (Beloica et al., 2010), and/or still a large number of specimens tend to fail cohesively in enamel (El Zohairy et al., 2010). Both tendencies should most probably be attributed to interfacial defects that are incorporated within individual μTBS specimens (Ferrari et al., 2002), this along with the high brittleness of enamel. In addition, the relatively aggressive cutting method employed to prepare μTBS specimens not necessarily imposes the same amount of stress to each micro-specimen. On the other hand, several μTBS studies revealed only a limited number of pre-testing failures; most failures were reported to have occurred adhesively at the interface or were 'mixed' adhesive-cohesive failures that always included the interface (Poitevin et al., 2010; Hanabusa et al., 2012; Yaman et al., 2014). This once again confirms the high variance in μTBS data gathered at different research centers. A μTBS test is rather technique sensitive. Specimen preparation for a μTBS test should be done not only in a very accurate, but also in a controlled and standardized way. Devices like a semi-automated and programmable diamond saw (e.g. Accutom 50, Struers; also used for the preparation of mini-iFT specimens in this study) and a so-called Micro-Specimen Former (University of Iowa, Iowa, IA, USA) to prepare an interfacial constriction (to better concentrate tensile stress at the actual interface) are hence preferred as they can minimize differences in specimen preparation in contrast to when the micro-specimens would be prepared by hand. A strong argument against the alternative shear bond-strength approach, including the μSBS protocol, is that stress imposed during shear loading is commonly documented to concentrate more within the base material, usually dentin, than at the actual adhesive interface (Della Bona and van Noort, 1995; Versluis et al., 1997; Braga et al., 2010; Jongsma et al., 2012). Therefore, a test that better controls loading of the actual interface and thus assesses the strength of the interface itself is highly desired; the innovative fracture mechanics-based mini-iFT test introduced recently to assess bonding effectiveness to dentin (Pongprueksa et al., 2016) and with this study now also employed to assess bonding effectiveness to enamel, appeared to meet this objective closely. It should nevertheless be clear that this innovative mini-iFT test method requires time and proper skills of the operator to prepare the specimens in a very accurate manner.

Overall, SEM failure analysis of mini-iFT specimens revealed defect-free and rather sharp notch tips that all

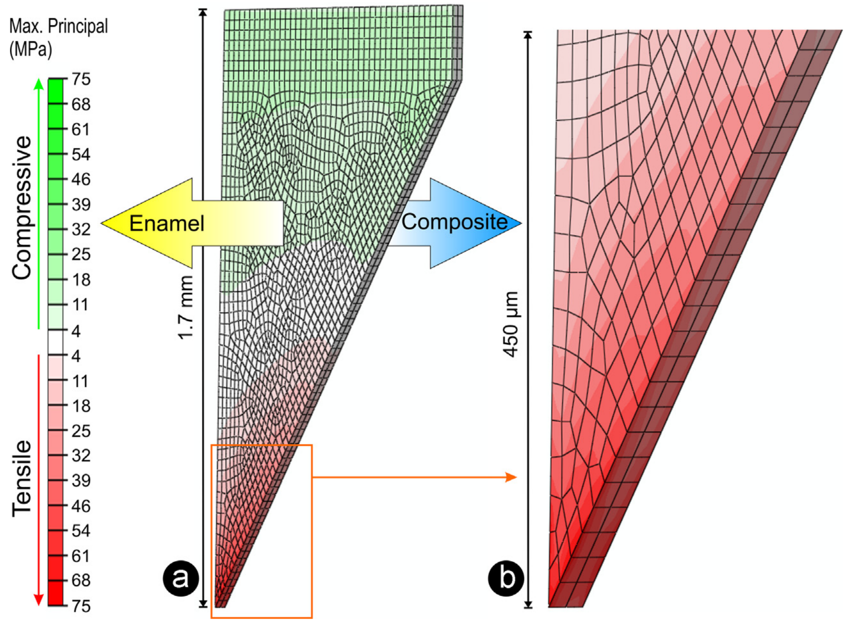

Fig. 5 – Finite element analysis (FEA) of the mini-interfacial fracture toughness (mini-iFT) notch geometry. Graphical presentation of the FEA model and the mesh used can be found in Fig. 1. Only the adhesive resin part of the model is shown to visualize the stress distribution along the adhesive-enamel boundary. (a) Principal stress analysis revealed mainly tensile stress (red color) at the tip of the flaw, while minor compressive stress (green color) can be observed from about the middle of the specimen and further on. (b) High magnification of the principal stress at the tip of the flaw showed a favorable stress concentration at the adhesive-enamel interface, which was higher than at the adhesive-composite interface. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

seemed to have been prepared in a very consistent way. The fracture of all mini-iFT specimens was initiated at the actual interface near the notch tip. The track, along which the crack propagated, was on the other hand found to vary and appeared to be associated with the magnitude of the resultant mini-iFT. For SEa, the crack typically propagated along the adhesive-enamel interface and deviated towards the adhesive layer near the end of the notch. A similar trend was before observed when the mini-iFT of SEa at dentin was measured; it was also supported by the FEA of stress distribution along the adhesive-dentin interface in the previous study (Pongprueksa et al., 2016). This crack-propagation track recorded for SEa differs from that recorded for E&Ra; regarding the latter, the crack initiated at the adhesive-enamel interface near the notch tip and propagated more through the adhesive and composite layer; once about the middle of the specimen was reached, the crack often deviated completely inside the composite. This phenomenon was documented before and termed as 'compression curl'; it has been related to the crack that propagated from the tensile loading side of the specimen (bottom part) into the compression loading side of the specimen (top part) during the 4-point bending (Quinn, 2007). The size and location of the compression curl appeared again to be related to the magnitude of the mini-iFT; for specimens that presented with a higher mini-iFT, the compression curl tended to start closer to the notch tip.

FEA of the mini-iFT specimen design revealed stress to concentrate between the adhesive and the enamel substrate at the notch tip, similarly as was shown before for the interface with dentin (Pongprueksa et al., 2016). A slight difference is that the stress amount was equal at the adhesive-dentin and adhesive-composite interface near the notch tip of the dentin mini-iFT specimens; FEA of the enamel mini-iFT specimens in this study revealed higher tensile stress at the adhesive-enamel than the adhesive-composite interface. Moreover, following the same loading conditions of 30 N, the maximum principal stress at the adhesive-enamel interface (68.4 MPa in this study) was slightly higher than at the adhesive-dentin interface (60.6 MPa; according to Pongprueksa et al., 2016); this must probably be ascribed to the higher elastic modulus of enamel as compared to that of dentin and composite. This difference in maximum principal stress at enamel versus dentin might explain to some extent the lower mini-iFT values measured for the same adhesives at enamel than at dentin. The measured difference in mini-iFT was, however, 20–30% lower at enamel (Pongprueksa et al., 2016), suggesting a clearly lower fracture toughness at enamel. The distribution of stress, as revealed by FEA, also corroborated very well the SEM fracture analysis data. Crack propagation was indeed found to occur preferentially along the adhesive-enamel interface, which should be considered a prerequisite to determine the strength of the actual adhesive interface (Scherrer et al., 2010). In addition, it is remarkable that areas along the inner edge of the notch, where the tensile stress concentration is higher, failed more at the enamel-adhesive interface than at the outer edge where FEA disclosed clearly less stress concentration, as can be seen in Figs. 3,1a and 3,3a.

The notch shape of the mini-iFT specimens used in this study was based on that employed in the single gradient notched beam (SGNB) fracture toughness setup (Wan et al., 2009); this is considered a simplified version of the chevron notch beam (CNB) fracture toughness test specimen (ISO24370, 2005). Furthermore, the mini-iFT test was shown to correlate well with the popular μTBS test and appeared even more discriminative in assessing bonding effectiveness to dentin (Pongprueksa et al., 2016). Based on the present favorable results, the mini-iFT now also turned out to be a valid alternative test method to determine interfacial bonding effectiveness to human enamel.

The new mini-iFT test is thought to discriminate adhesives better in terms of bonding effectiveness than conventional bond-strength tests, this with the ultimate goal to be able to better predict clinical effectiveness in the laboratory.

5. Conclusion

An E&R application mode presented with a higher interfacial fracture toughness to enamel than a SE approach. The additional application of a separate adhesive resin did not increase the interfacial fracture toughness to enamel for an E&R application mode (3-step versus 2-step), but did so for a SE application mode (2-step versus 1-step). FEA suggested a favorable stress distribution along the adhesive-enamel interface during loading, which was corroborated by the SEM fracture analysis. It can therefore be concluded that the mini-iFT test method appears a valid alternative to assess the strength of the interface of an adhesive bonded to human enamel.

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