Bonding Efficacy of a New Self-adhesive Restorative onto Flat Dentin versus Class-I Cavity-bottom Dentin

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Purpose: This study investigated the bonding efficacy of a new so-called 'self-adhesive composite hybrid' onto flat ('FLAT') and high C-factor class-I cavity-bottom ('CAVITY') dentin.

Materials and Methods: The 'immediate' and 'aged' (50k thermocycles) micro-tensile bond strength (μTBS) to FLAT and CAVITY dentin of the experimental self-adhesive bulk-fill restorative (K-0180 ASAR pilot: 'ASAR-pilot'; Dentsply Sirona) was compared to that of two universal adhesives applied in self-etch mode and combined with a bulk-fill composite (Prime&Bond Elect/QuiXfil: 'P&Be/QuiXF'; Prime&Bond Active/QuiXfil: 'P&Ba/QuiXF'; both from Dentsply Sirona), two pre-conditioned materials (Activa Bioactive-Restorative (Pulpdent): 'Activa'; Fuji II LC Improved (GC): 'Fuji2LC'), and one bulk-fill glass-hybrid restorative (Equia Forte Fil (GC): 'EquiaF'). Statistically significant differences were recorded using Welch's ANOVA with Games-Howell contrast ($p<0.05$).

Results: No significant difference in immediate μTBS was recorded when the restorative materials were applied onto FLAT dentin, except for Activa_FLAT and EquiaF_FLAT. When bonded onto CAVITY dentin, the significantly highest μTBS was recorded for Fuji2LC_CAVITY (layer-filled), solely not being significantly different from P&Ba/QuiXF_CAVITY. Upon aging, the highest μTBS to flat dentin was achieved by ASAR-pilot_FLAT, not being significantly different from P&Be/QuiXF_FLAT and Fuji2LC_FLAT. No significant difference between immediate and aged μTBS was recorded for ASAR-pilot when bonded onto FLAT or CAVITY dentin, the latter however associated with a low bond strength.

Conclusion: A favorable bonding performance was found for the new self-adhesive bulk-fill composite hybrid ASAR-pilot when bonded onto flat dentin. Much lower bond strength was however recorded when ASAR-pilot was bonded to high C-factor cavity-bottom dentin.

Keywords: Bond strength, Bulk-fill, Composite, Dentin, Durability

Clinical Relevance: A favorable bonding performance can be achieved when the new self-adhesive bulk-fill composite hybrid ASAR-pilot is bonded to flat dentin. However, its clinical use into deep cavities remains to be further addressed.

INTRODUCTION

Resin-based composite has widely been applied in dental practice since it was introduced into dentistry in the 1950's.[19] When combined with a modern dental adhesive, adhesively restoring teeth in a reliable, predictable and durable way can today be considered a reality. However, research-and-development in dental adhesive technology remains to be challenged, for instance now to fabricate adhesives that are also effective in conditions of suboptimal field control or complex cavity configurations, or to develop economic, easy-to-place and preferentially self-adhesive 'true' amalgam alternatives. In this decade, self-adhesive tooth-colored restorative materials that do no longer need pre-treatment with a separate adhesive are vigorously being developed for their clinical appeal of desired shorter application time and lower technique sensitivity.[7,38,41]

Self-adhesive restorative composites can be considered as the logical succession of self-adhesive composite cements, which since quite some time have been commercialized as easy-to-use 'compromise' cements to lute semi-direct/indirect ceramic and composite CAD/CAM restorations. Regarding bonding effectiveness, these self-adhesive luting composites were generally documented to be less effective than their etch-and-rinse and self-etch counterparts that respectively require a separate etch-and-rinse or self-etch adhesive as luting pre-treatment.[13,50] With a consistency and probably composition very alike that of luting composites, flowable self-adhesive restorative materials have been developed first, among which some were commercialized, yet with mixed success.[38,49,51] Self-evidently, self-adhesiveness is easier to achieve with a flowable than a more viscous paste-like consistency, this already solely because of better surface-wetting potential of less viscous composite formulations.

Self-adhesive composite that has the adhesive inserted into the restorative material needs to be combined with bulk-fill potential to become an amalgam-like dental restorative material. Bulk-filling cavities in posterior teeth enable clinicians today to fill 4- to 5-mm deep boxes in one single bulk.[17,43] Nevertheless, previous research demonstrated that bulk-filling does not always provide favorable research data, as for instance some bulk-fill composites were shown to result into higher polymerization shrinkage and larger interfacial gaps than incrementally layered composites.[2,21]

Recently, a so-called 'self-adhesive bulk-fill restorative composite hybrid' (coded as 'K-0180 ASAR pilot' and being referred to as 'ASAP-pilot' in this study; Dentsply Sirona; Konstanz, Germany) was developed and later commercially introduced as Surefil One (Dentsply Sirona). To our knowledge, few

in-vitro studies reported on the performance of self-adhesive bulk-fill restoratives. In this study, we investigated the 'immediate' and 'aged' bonding effectiveness of the self-adhesive bulk-fill restorative hybrid when applied both onto flat and class-I cavity-bottom dentin, the latter serving as a 'worst-case' condition due to the high C-factor involved. Representative adhesive/composite combinations as well as conventional/resin-modified glass-ionomer cements (GICs) were likewise applied, serving as references/controls. The null hypotheses tested were that (1) the immediate bonding efficacy onto flat dentin or cavity-bottom dentin of the new self-adhesive bulk-fill composite hybrid ASAR-pilot did not significantly differ from that obtained when the other restorative materials were bonded onto flat (1a) or high C-factor class-I cavity-bottom dentin (1b), and (2) the bonding efficacy did not decrease upon substantial artificial aging when again applied following the two substrate conditions (2a,b).

MATERIAL AND METHODS

Tooth preparation

Ninety-six non-carious human third molars were collected (following informed consent approved by the Commission for Medical Ethics of KU Leuven under file number S57622), stored in 0.5% chloramine T/water at 4°C and used within 1 month after extraction. All teeth were randomly subdivided in 12 experimental groups (n=8 per experimental group). For the six 'FLAT' groups, the crown was cut 4 mm below the cusp tips, ending with a surface at mid-coronal dentin. For the six 'CAVITY' groups, the teeth were first built up using the flowable composite G-ænial Universal Flo (GC; Tokyo, Japan) after etching enamel with phosphoric acid (DeTrey Conditioner 36, Dentsply Sirona) and subsequent application of the universal adhesive Prime&Bond Active (Dentsply Sirona). A flat surface was made at the level of the cusp tips, upon which standard box-type class-I cavities (3.5×3.5×4 mm) were prepared with the cavity bottom again ending at mid-coronal dentin, as for the FLAT dentin surfaces. In this way, the effect of regional variability on micro-tensile bond strength (μTBS) was kept minimal. All preparations were made using the MicroSpecimen Former (University of Iowa; Iowa, IA, USA), equipped with a high-speed medium-grit (107 μm) diamond bur (882, Komet; Lemgo, Germany). Onto the flat dentin surfaces, a 3.5×3.5×4-mm buildup with the same dimensions as those of the class-I cavities, was made by applying the respective restorative material in bulk/layers in an addition-silicone mould (Aquasil medium body, Dentsply Sirona), while the class-I cavities were likewise bulk/layer-filled with the

restorative materials. The experimental tooth-preparation protocol is schematically explained in Fig. 1.

All restorative materials were applied strictly following the respective manufacturer's instructions (Table 1). Bonding efficacy of the experimental self-adhesive bulk-fill composite hybrid (1: 'ASAR-pilot') was compared with the adhesive/composite combinations consisting of the universal adhesives Prime&Bond Elect (Dentsply Sirona) and Prime&Bond Active (Dentsply Sirona) applied in self-etch mode prior to the application of the paste-like 'full-body' bulk-fill composite QuiXFil (Dentsply Sirona) (2: 'P&Be/QuiXF'; 3: 'P&Ba/QuiXF'), the so-called 'bioactive ionic resin with reactive glass filler' (Activa Bioactive-Restorative, Pulpdent; Watertown, MA, USA) following phosphoric acid-etching using DeTrey Conditioner 36 (Dentsply Sirona) (4: 'Activa'), the resin-modified glass-ionomer restorative Fuji II LC Improved (GC; Tokyo, Japan) applied in three layers following conditioning with the poly-alkenoic glass-ionomer conditioner Dentin Conditioner (GC) (5: 'Fuji2LC'), and the 'bulk-fill glass-hybrid restorative' Equia Forte Fil (GC) (6: 'EquiaF'). The composite buildups/restorations were light-cured using a light-emitting diode (LED) curing light (Bluephase 20i, Ivoclar Vivadent; Schaan, Liechtenstein) with an output of 1200 mW/cm^2, when used in 'high mode', as determined and confirmed regularly during the experiment using a Marc Resin Calibrator (BlueLight Analytics; Halifax, Canada). The light output was measured each time before the start and at the end of a test.

Specimen preparation for μTBS testing

Once the bonded macro-specimens were prepared, they were kept for 1 hr at 100% humidity prior to being immersed and stored for 1 week in distilled water at 37°C. After 1-week water storage, all specimens were sectioned perpendicular to the interface using a water-cooled diamond saw (Accutom-50, Struers; Ballerup, Denmark) to obtain rectangular sticks (4 central micro(μ)-specimens per tooth: 1×1 mm wide; 8-9 mm long). During μ-specimen preparation, alginate was used to support the thin slabs after the first cut, this in an attempt to prevent pre-test failures as much as possible. For each experimental group, 16 non-trimmed μ-specimens (2 μ-specimens per tooth) were immediately tested to determine the IMMEDIATE μTBS and another 16 μ-specimens were aged for 50k thermocycles between two water baths at 5°C and 55°C using a THE-1200 thermocycler (SD Mechatronik; Munich, Germany) prior to testing to determine the AGED μTBS (Fig. 1). For the actual μTBS test, the specimens were fixed to a BIOMAT jig with cyanoacrylate-based glue (Model Repair II

Blue, Dentsply Sirona Sankin; Tochigiken, Japan) and stressed at a crosshead speed of 1 mm/min until failure in a LRX testing device (LRX, Lloyd; Hampshire, UK) using a load cell of 100N. When specimens fail before actual testing, they were recorded as pre-test failures (ptf's) and included as 0 MPa to calculate the mean μTBS. The whole test protocol followed the Academy of Dental Materials guidelines for μTBS testing.[3]

Fractographic analysis by light microscopy (LM) and scanning electron microscopy (SEM)

After the μTBS test, all fractured specimen halves (both the dentin and restoration sides) were observed under 50x magnification using a stereo-microscope (Stemi 2000-CS, Zeiss; Oberkochen, Germany) to classify the mode of failure as either 'cohesive failure in dentin', 'cohesive failure in composite, 'adhesive (interfacial) failure', or 'mixed failure'.

Representative fractured surfaces, exhibiting the most frequently recorded failure mode and originating from a μTBS close to the mean, and the pre-test failures were further processed for high-magnification SEM examination (JSM-6610LV, Jeol; Tokyo, Japan). These SEM specimens were fixed using 2.5% glutaraldehyde, gradually dehydrated in increasing concentrations of ethanol, and chemically dried using hexamethyldisilazane (HMDS; Acros Organics, Thermo Fisher Scientific; Geel, Belgium).[35] Finally, the specimens were thinly gold-coated using a gold-sputter machine (JFC-1300, Jeol). SEM photomicrographs were taken at 85-95×, 2000×, and 9000× original magnification under an accelerating voltage of 5 kV.

Statistical Analyses

Statistical analysis was carried out using SPSS 23 (IBM; Chicago, IL, USA) with statistical significance set at α=0.05. As some μTBS data of the experimental groups revealed unequal variance, Welch's ANOVA with Games-Howell contrast was used to check for statistically significant differences (p<0.05).

Results

μTBS testing

The bonding effectiveness data (including ptf number) are shown in Fig 2. No significant difference in immediate μTBS was recorded when the restorative materials were applied onto flat dentin, except

for Activa_FLAT and the conventional GIC EquiaF_FLAT, both exhibiting a significantly lower bond strength (for Activa_FLAT mainly due to a high ptf number). The self-adhesive bulk-fill restorative hybrid ASAR-pilot did not significantly underscore the other restoratives. In addition, when bonded onto class-I cavity-bottom dentin, the significantly highest μTBS was recorded for the pre-conditioned resin-modified GIC Fuji2LC_CAVITY (layer-filled), solely not being significantly different from P&Ba/QuiXF_CAVITY. A significantly lower μTBS onto cavity-bottom dentin along with a high ptf number was recorded for ASAR-pilot_CAVITY.

Upon aging, the highest aged μTBS to flat dentin was achieved by ASAR-pilot_FLAT, not being significantly different from P&Be/QuiXF_FLAT and Fuji2LC_FLAT. The significantly lowest aged μTBS to flat dentin was again recorded for Activa_FLAT and EquiaF_FLAT' (mainly due to a high ptf number). Bonding to cavity-bottom dentin was significantly lower upon aging than to flat dentin for all restorative materials, among which also for ASAR-pilot_CAVITY, but not for P&Ba/QuiXF_CAVITY. All P&Be/QuiXF_CAVITY and Activa_FLAT/CAVITY specimens failed prior to testing (ptf).

Furthermore, no significant difference between immediate and aged μTBS was recorded for ASAR-pilot when bonded onto flat or cavity-bottom dentin, the latter ASAR-pilot_CAVITY however associated with a high ptf number, while no ptf's were recorded for ASAR-pilot_FLAT. After aging, P&Ba/QuiXF_FLAT, EquiaF_FLAT, Fuji2LC_CAVITY and EquiaF_CAVITY presented a lower μTBS than their immediate μTBS (p<0.05).

Failure-mode analysis

The failure-mode analysis in percentage is graphically presented in Fig 3. Overall, the analysis clearly revealed that most fractured surfaces failed adhesively at the interface. Also the pre-test failures predominantly were interfacial failures, clearly indicating very low bond strength. The higher bond strength recorded for ASAR-pilot_FLAT, Fuji2LC_FLAT and Fuji2LC_CAVITY went along with a higher percentage of cohesive failures in composite and mixed failures (Fig 3A). Upon aging, most de-bonded surfaces again represented adhesive interfacial failures, with the exception of ASAR-pilot_FLAT, EquiaF_FLAT and EquiaF_CAVITY, for which also mixed failures and cohesive failures in composite/GI in a higher percentage were recorded (Fig 3B).

SEM fracture analysis

Representative SEM photomicrographs of fractured µ-specimens representing ASAR-pilot_FLAT and ASAR-pilot_CAVITY upon 1w water storage ('immediate') and 50k TC ('aged') are shown in Figs 4 and 5, respectively. Immediate ASAR-pilot_FLAT failed partially at the interface and partially within the composite. Aged ASAR-pilot_FLAT failed at the interface. The scratches produced by the diamond bur were filled with the restorative material, which consisted of filler particles with sizes ranging from less than 1 µm up to around 8 µm (Figs 4c and 4d). However, immediate ASAR-pilot_CAVITY revealed mainly cohesive failures with partially adhesive interfacial failures. Higher magnification photomicrographs (2000× and 9000×) illustrated that dentin remained covered by hybrid layer and smear plug, indicating less effective bonding performance (Figs 5a and 5b). Furthermore, aged ASAR-pilot_CAVITY failed cohesively within the composite, which was accompanied with some air bubbles (Figs 5c1 and 5d1, 95× original magnification).

Representative SEM photomicrographs of the dentin side (except Fig 7d: resin side) of fractured µ-specimens representing the different experimental groups (restorative materials) bonded to flat and cavity-bottom dentin upon 1w water storage ('immediate') or 50k TC ('aged') are shown in Figs 6 and 7, respectively. Most fractured surfaces (Fig 6) failed at the interface, except for immediate P&Be/QuiXF_CAVITY and ACTIVE_CAVITY (mixed failures). Upon ageing, higher magnification SEM showed porosities within the adhesive layer for P&Ba/QuiXF_FLAT (Fig 6d3). Interfacial failure was recorded for Fuji2LC when bonded to a polyalkenoic-acid pre-conditioned flat dentin surface (Figs 7a1 and 7c1, 85-90× original magnification). However, a small amount of Fuji2LC remained on the surface, which contained filler particles with sizes ranging from less than 2 µm to over 10 µm (Figs 7c2 and 7c3). Also noteworthy are the small cracks present in 'EquiaF' (Figs 7e and 7f, 2000× original magnification), which in the first place should be regarded as dehydration artifacts of the water-containing GIC. Immediate EquiaF_FLAT failed cohesively within GIC, while EquiaF_CAVITY failed at the interface. Circular scratches produced during preparation of the standard box-type class-I cavity can be observed (Fig 7f1, 90× original magnification).

DISCUSSION

In this study, we investigated the bonding effectiveness of K-0180 ASAR pilot ('ASAR-pilot'), the

experimental pre-cursor of a new self-adhesive bulk-fill restorative material that has recently been commercialized under the brand Surefil One (Dentsply Sirona). According to manufacturer's information, this so-called 'self-adhesive composite hybrid' claims to offer the dentist an innovative filling concept for posterior teeth.[14] Surefil one is allegedly a 'forgiving material that combines the simplicity of a glass-ionomer cement (GIC) with the stability of a conventional resin-based composite (RBC) without sacrificing aesthetic outcome'. Key features of the new posterior restorative material are self-adhesiveness and bulk-filling. Considering the product description, the claims made by the manufacturer and the new material's key features, a representative but diverse group of restorative materials were included in this study (Table 1). As one of the first bulk-fill composites, although not called 'bulk-fill' composite at the time of its market introduction, the RBC QuiXFil (Dentsply Sirona)[15] was applied in this study, like ASAR-pilot, following its manufacturer's instructions in 4-mm bulk. Not being a self-adhesive composite, QuiXFil was applied after adhesive (pre-)treatment with either the universal adhesive Prime&Bond Elect ('P&Be/QuixF') or the successor universal adhesive Prime&Bond Active ('P&Ba/QuixF'), both bonded in self-etch mode. While P&Be contains dipentaerythritol pentacrylate phosphate (PENTA), P&Ba contains 10-methacryloyloxydecyl dihydrogen phosphate (10-MDP) as functional monomer. Claimed to be a 'bioactive' restorative material with alleged mineralization potential,[39] Activa Bioactive-Restorative (Pulpdent; 'Activa') is documented by the manufacturer as a dual-cure resin-based material with GIC components (modified polyacrylic acid, reactive glass filler). According to the original manufacturer's instructions, Activa was in this study applied in 4-mm bulk and in a semi/self-adhesive mode after solely phosphoric acid-etching dentin (at the time of publication of this paper, the manufacturer recommends to additionally use a bonding agent of own choice). Being a resin-modified GI, Fuji II LC Improved (GC; 'Fuji2LC') was applied according to its manufacturer's instructions in three 1.8-mm layers following polyalkenoic-acid (pre-)conditioning. Finally, representing a conventional GIC, also being referred by its manufacturer to as 'bulk fill glass hybrid restorative system', Equia Forte Fil (GC; 'EquiaF') was applied in this study following its manufacturer's instructions in 4-mm bulk and in a full self-adhesive mode as dentin did not receive any pre-treatment.

To measure bonding efficacy, a micro-tensile bond strength (μTBS) test was selected with the six restorative systems being applied to low C-factor flat (mid-coronal, bur-cut) dentin versus high C-factor class-I cavity-bottom (mid-coronal, bur-cut) dentin. The latter bonding condition should be regarded

as the 'worst-case' condition, especially for the restorative materials that are applied in a full self-adhesive and bulk-fill application mode (ASAR-pilot, EquiaF). The test involving bonding to class-I cavity-bottom dentin has been applied in previous research[10,11,30,40,46] and appeared especially useful to discriminate bonding efficacy of bulk-fill composites.[44,45]

Finally, the test design also enabled to compare 'immediate' with 'aged' bonding efficacy, the latter in this study involving long-term 50k thermo-cycles, much more than the 500 thermo-cycles required by the ISO/DTS 11405 standard. Moreover, thermocycling is the most commonly used method to simulate *in-vivo* aging, which results in contraction, expansion stress and hydrolytic degradation at the bonded interface.[12,48] Challenging bond-strength specimens by exposing them to long-term water storage or even thermo-cycling is as a laboratory test in prediction of clinical bond longevity a must to measure bonding efficacy of adhesives, self-adhesive cements and restorative materials.

While a self-adhesive bulk-fill restorative application procedure can be regarded as the simplest filling technique, hereby approximating the amalgam filling technique, logically a lower bonding efficacy is expected when no separate adhesive with better wetting and more intensive adhesive interaction is beforehand used. However, the findings of this study did not reveal significant difference in immediate μTBS when the restorative materials were applied onto flat dentin, except for Activa_FLAT and EquiaF_FLAT that both exhibited a significantly lower bond strength. Therefore, the tested null hypothesis part (1a) that the immediate bonding effectiveness onto flat dentin of the new self-adhesive bulk-fill composite ASAR-pilot did not significantly differ from that obtained when dentin was bonded with the other restorative materials, was accepted. Additionally, there was no significant difference in immediate μTBS when bonded onto class-I cavity-bottom dentin, except for only Fuji2LC_CAVITY that exhibited a significantly higher bond strength. Hence, the tested null hypothesis part (1b) that the immediate bonding effectiveness onto cavity-bottom dentin of the new self-adhesive bulk-fill composite ASAR-pilot did not significantly differ from that obtained when dentin was bonded using the other restorative systems, was accepted for five out of the six restorative systems tested, and failed to be accepted regarding the bonding efficacy onto cavity-bottom dentin for ASAR-pilot_CAVITY versus Fuji2LC_CAVITY. Upon long-term thermo-cycling aging, the aged μTBS of 8 restorative system/substrate conditions did not significantly decrease as compared to the 1w immediate μTBS, while a significant decrease in μTBS upon aging was recorded for the four restorative system/substrate conditions P&Ba/QuiXF_FLAT, Fuji2LC_CAVITY, EquiaF_FLAT and EquiaF CAVITY.

Therefore, the tested null hypothesis parts (2a) and (2b) that the bonding efficacy did not decrease upon substantial artificial aging when applied following the two substrate conditions, were accepted except for the four restorative system/substrate conditions mentioned above.

Regarding ASAR-pilot, no significant difference in μTBS was recorded as compared to the highest recorded μTBS onto flat dentin by the pre-conditioned, layer-filled resin-modified GIC Fuji2LC, confirming ASAR-pilot's quite favorable self-adhesiveness. To achieve self-adhesiveness, the restorative material should contain functional mono/polymers to interact with tooth structure, producing micro-retention by etching and/or realizing primary chemical interaction.[20,31,37] The latter two main bonding mechanisms can self-evidently only be effective if the restorative material adequately wets the surface as primary requirement for adhesion. According to the compositional data released by its manufacturer, ASAR-pilot contains polycarboxylic and acrylic acid, and bifunctional acrylate. The latter acrylate monomers ensure the formation of a 'crosslinked' resin network following polymerization reaction and provide increased mechanical strength.[4,22,26] Besides superficial etching resulting in a submicron hydroxyapatite-rich hybrid layer, ionic bonding potential has been attributed to polycarboxyl acid, constituting the main bonding mechanisms of (resin-modified) GICs.[28,52] As disclosed by SEM (Figs 4a and 4b), exposed collagen fibers can be clearly seen, hereby suggesting that ASAR-pilot may have etching potential. To fully elucidate, however, if ASAR-pilot's self-adhesiveness should be ascribed to both micro-mechanical interlocking through etching along with primary chemical bonding, interfacial TEM characterization combined with chemical XRD and NMR analysis is required in further research.

However, the μTBS of ASAR-pilot dropped significantly when bonded to class-I cavity-bottom dentin (ASAR-pilot_CAVITY), the latter associated with a high proportion of pre-test failures (ptf). This is in agreement with many previous studies that found a decrease in bond strength when bonded to class-I cavity-bottom dentin as compared to bonding onto a flat dentin surface.[33,57] The differences can be ascribed to many factors, among which most likely the cavity-configuration factor, commonly being referred to as the C-factor, being most determining. Polymerization shrinkage stress within cavities is well known to be largely affected by the C-factor.[18] As calculated in previous research by Van Ende *et al.*,[44] the C-factor of flat dentin, with the same dimensions (3.5×3.5×4 mm) as used in this study, is 0.18, while that of the class-I cavity (3.5×3.5×4 mm) is 5.8. This significantly increased C-factor restricted the free flow of the polymerizing/shrinking resin-based material, since more material was constrained by

being bonded to the rather stiff cavity walls;[27] resin-based material could shrink nearly unrestricted when bonded to a flat dentin surface.[30] Therefore, more shrinkage stress could have acted on the bond to cavity-bottom dentin, correlating positively with a higher proportion of interfacial gaps[21] and affecting bonding efficacy. In previous similar experiments investigating bonding to high C-factor class-I cavity bottom dentin, only the bonding efficacy of the bulk-fill flowable composite SDR (Dentsply Sirona) appeared insensitive, while other bulk-fill composites with a paste-like consistency suffered much more from the high shrinkage stress generated in the high C-factor cavity.[25,45]

Besides the effect of C-factor, another plausible reason that may have contributed to the significantly different 'FLAT surface' versus 'CAVITY' data recorded for ASAR-pilot may be related to control of dentin moisture. Although containing water, the self-adhesive bulk-fill composite hybrid ASAR-pilot requires some moisture to activate the included functional acids.[8] Consequently, the manufacturer instructed that the dentin surface should not be desiccated. However, it is much more difficult to control surface moisture in a deep and narrow cavity than at a flat surface, so to guarantee the adequately needed moisture amount at the surface.[11] Furthermore, using a relatively high-viscous bulk-fill composite, surface wetting could have been compromised, having produced porosities at the bonded interface. Being not a liquid, also intratubular and inter-collagen infiltration is more difficult for a material with a paste-like consistency, as compared to what can be achieved by liquid adhesive solutions.

The first bulk-fill RBC QuixF combined with the self-etch adhesive P&Be resulted in a relatively high μTBS when bonded to flat dentin (QuixF/P&Be_FLAT). No significant reduction in μTBS onto flat dentin was recorded upon 50k thermo-cycling. However, when bonded to cavity-bottom dentin (QuixF/P&Be_CAVITY), almost all μ-specimens failed before testing (ptf's), indicating that the polymerization shrinkage stress developed within the high C-factor cavity must have led to interface de-bonding at the cavity bottom. Interestingly, when QuixF was combined with the self-etch universal adhesive (successor) P&Ba, a significantly better bonding efficacy was recorded in the class-I cavities (QuixF/P&Ba_CAVITY). Compared with the PENTA-containing universal adhesive P&B Elect, P&B Active contains the functional monomer 10-MDP. Considering its chemical structure with five methacrylate groups versus one phosphate group, PENTA should rather be regarded as a cross-linking monomer than a monomer with good ionic bonding potential to Ca of hydroxyapatite; the latter interaction potential of PENTA's phosphate group may even be expected to be sterically hindered by the

surrounding methacrylate groups.[1,47] However, much research has demonstrated that the functional monomer 10-MDP is one of the best performing functional monomers,[54,55] the main reason because of which most of the newest generation of universal adhesives contain 10-MDP.[32,36] Besides etching capability, providing surface micro-retention, and primary ionic bonding potential to Ca of hydroxyapatite, 10-MDP has been documented to uniquely self-assemble into so-called nano-layers of stable 10-MDP_Ca salts.[53,56] This difference in functional monomer is the most logic explanation for the superior bonding efficacy of QuixF/P&Ba versus that of QuixF/P&Be recorded in this study. Furthermore, the manufacturer claimed that P&Ba is well balanced for hydrophobic/hydrophilic properties, promoting surface wetting and resin infiltration at various moisture conditions.

When it comes to Activa, Activa's bioactive ionic resin is claimed to facilitate diffusion of calcium, phosphate and fluoride ions across the restoration-dentin interface,[8,58] hereby stimulating hydroxyapatite formation and remineralization at the bonded interface. This claimed interfacial bioactivity, for which at the time of publication no hard (independent) evidence has been published in scientific literature, can self-evidently only work if the restorative material makes direct contact with the dentin substrate, thus requiring self-adhesiveness. Pre-etching with phosphoric acid, as originally instructed by Activa's manufacturer, will not prevent the claimed interfacial bioactivity. However, the newly released application instructions to additionally use a bonding agent will block direct Activa-dentin contact, by which any interfacial bioactive interaction becomes very questionable. The adapted instructions-for-use, now also requiring the (pre-)application of a separate adhesive, were released because of a recently documented insufficient self-adhesiveness of Activa. A randomized clinical trial indeed revealed that the use of Activa in class-I/II cavities, applied as instructed by the manufacturer after a short phosphoric-acid pretreatment without adhesive, resulted in a non-acceptable very high failure frequency after a one-year period of clinical service.[42] The authors concluded that further studies involving Activa should be conducted using a bonding agent. This insufficient self-adhesiveness of Activa was confirmed in this study, when the significantly lowest μTBS along with a very high incidence of pre-test failures (only ptf's upon aging) was recorded when Activa was bonded both onto pre-etched flat and cavity-bottom dentin. Although during μ-specimen preparation alginate was used to support the thin slabs after the first cut, many Activa μ-specimens failed during the second cutting action (perpendicular to the first one). Reasons for this inferior bonding performance of Activa must be multifactorial. The most plausible explanation is Activa's weak self-adhesiveness to dentin,

considering the low μTBS and high ptf incidence recorded even when Activa was bonded onto flat dentin (in contrast to all other restorative systems investigated, except also for EquiaF). In addition, the initial bond strength of Activa could not withstand the polymerization shrinkage stress developed in the class-I cavity. Hence, apart from developing materials with potential bioactive properties, primary properties such as mechanical strength and bonding potential remain essential.

Overall, in terms of bonding efficacy, the best performing restorative system was Fuji2LC in this study. When bonded both onto flat and class-I cavity-bottom dentin, a high immediate and aged μTBS were recorded for the pre-conditioned resin-modified GIC applied in incremental layers. Our finding is in concordance to previous research that showed that an incremental layer-filling technique reduces contraction stress and improves adhesion into cavities with tight internal adaptation.[2,5,34] As resin-modified GIC, Fuji2LC's self-adhesiveness should be attributed to combined micro-mechanical interlocking within submicron hydroxyapatite-rich hybrid-layer formation and primary chemical bonding of carboxylic groups with calcium in hydroxyapatite.[24,28] Previous research also reported that GIC is more capable of reducing contraction stress during early setting than RBC, increasing the possibility of realizing a durable bond to the cavity walls.[9]

Less favorable bonding efficacy was recorded for the conventional GIC EquiaF, which was applied to non-conditioned dentin. As a low μTBS was recorded to both flat as class-I cavity-bottom dentin, these inferior data should most logically be attributed to cohesive failure within the GIC rather than to actual bond failure, as this is typical of GIC's when subjected to bond-strength testing.[6,23] However, failure-mode analysis in this study revealed that most μ-specimens revealed interfacial and mixed failures for 'aged' EquiaF_FLAT. Other reasons for the low bond strength of EquiaF are (1) that dentin was not pre-etched, providing less effective micro-mechanical interlocking along with potential smear-layer interference, (2) that the restorative was solely self-cured, potentially having reached lower cohesive strength, and (3) the fact that the resin-based coating agent Equia Forte Coat (GC) was not used. Equia Forte Coat (GC) is a nano-filled resin coating agent with high hydrophilicity and low viscosity, which not only fills surface cracks and porosities, but also protects the early setting against the outer water environment.[29] The additional application of the resin coating has indeed previously been shown to increase abrasion and erosion resistance, to provide protection against water sorption, and to increase mechanical strength.[16,29,59]

CONCLUSION

In this study, we investigated the bonding efficacy and bond durability of the new 'self-adhesive bulk-fill composite hybrid' K-0180 ASAR pilot (later commercialized as Surefil One, Dentsply Sirona) to flat and class-I cavity-bottom dentin. When applied onto flat dentin, favorable immediate μTBS was recorded for ASAR-pilot; its self-adhesiveness to flat dentin resisted 50k thermo-cycling aging. However, ASAR pilot suffered from ptf's when bonded following the worst-case scenario to high C-factor class-I cavity-bottom dentin, although upon aging ASAR pilot did not underscore any other restorative system investigated.

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Table 1. List of materials investigated in this study.

Materials	Code	Composition[1,2]	Filler loading	Application procedure	Batch No.
K-0180 ASAR pilot (Dentsply Sirona)	**ASAR-pilot**	Aluminum-phoshor-strontium-sodium-fluoro-silicate glass, highly dispersed silicon dioxide, ytterbium fluoride, polycarboxylic acid, bifunctional acrylate, acrylic acid, iron oxide pigments, water, titanium dioxide pigments, camphorquinone, stabilizer, self-cure initiator	Not specified	Upon application in 4-mm bulk, light-cure for 20 sec with an output of 1200 mW/cm^2.	Not specified
QuiXfil (Dentsply Sirona)	**QuiXF**	UDMA, TEGDMA, di- and trimethacrylate resins, carboxylic acid modified dimethacrylate resin, BHT, UV stabilizer, camphorquinone, phosphate silicate glass, ethyl-4-dimethylaminobenzoate, silanated strontium aluminum sodium fluoride,	66 vol% 86 wt%	1. Application of Prime&Bond Elect (P&Be; PENTA, urethane dimethacrylate monomer, 2-hydroxy-3-acryloyloxypropyl methacrylate, HEMA, trimethylolpropane trimethacrylate, acetone) or Prime&Bond Active (P&Ba; MDP, PENTA, Bisacrylamide 1, Bisacrylamide 2, propan-2-ol, 4-(dimethylamino)benzonitrile) in self-etch mode (both adhesives from Dentsply Sirona); 2. Upon application in 4-mm bulk, light-cure for 10 sec with an output of 1200 mW/cm^2.	1710000818
Activa Bioactive-Restorative (Pulpdent)	**Activa**	Blend of diurethane and other methacrylates with modified polyacrylic acid, silica, amorphous, sodium fluoride	56 wt%	1. Etch prepared dentin for 5 sec with DeTrey Conditioner 36 (Dentsply Sirona; 25– 50% phosphoric acid), rinse well and lightly dry; 2. Upon application in 4-mm bulk, light-cure for 20 sec with an output of 1200 mW/cm^2.	171102

Fuji II LC Improved (GC)	**Fuji2LC**	HEMA, polybasic carboxylic acid, UDMA, dimethacrylate, others	76 wt%	1. Apply Dentin Conditioner (GC; 20% polyacrylic acid, 3% aluminum chloride, distilled water) for 20 sec, rinse thoroughly with water and dry gently; 2. Upon successive application in layers of max. 1.8-mm thickness (3 layers), light-cure each layer for 20 sec with an output of 1200 mW/cm^2.	170713A
Equia Forte Fil (GC)	**EquiaF**	Powder: fluoroaluminosilicate glass, polyacrylic acid, iron oxide Liquid: polybasic carboxylic acid, water	Not specified	No pre-treatment; self-cure, wait for 2.5 min (prior to further specimen processing).	170807A

[1]According to information provided by the respective manufacturer; [2]Abbreviations: ***BHT****: Butylated hydroxy toluene;* ***HEMA****: 2-hydroxyethyl methacrylate;* ***MDP****: 10-Methacryloyl-oxydecyl-dihydrogenphosphate;* ***PENTA****: Dipentaerythritol pentaacrylate phosphate;* ***TEGDMA****: Triethyleneglycol dimethacrylate;* ***UDMA****: Urethane dimethacrylate*

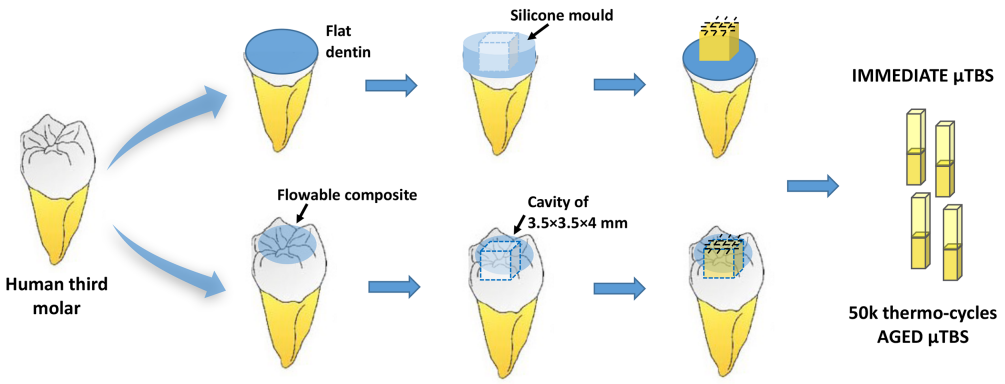

Fig 1 Schematic illustrating the specimen-preparation protocol for micro-tensile bond strength (µTBS) testing.

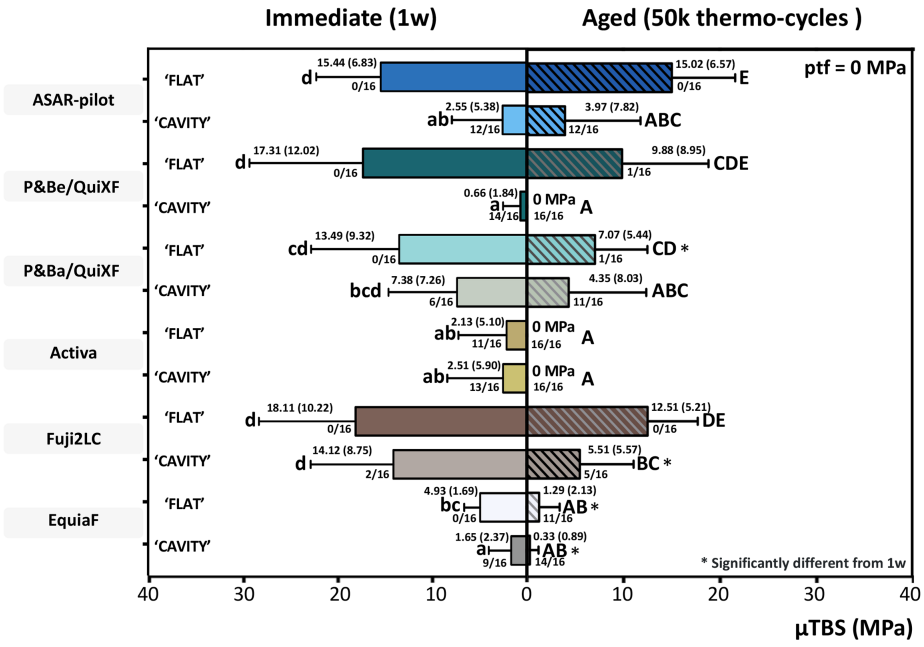

Fig 2 'Immediate' and 'aged' µTBS of the six restorative systems to flat ('FLAT') and class-I cavity-bottom dentin ('CAVITY'). The mean µTBS with the standard deviation (SD) and the number of pre-test failures (ptf's) per total number of micro(µ)-specimens are mentioned at each bar. Groups with the same small (immediate µTBS) or capital (aged µTBS) letter are not statistically significantly different (p>0.05). An asterisk indicates that there exists a significant difference between the immediate and aged µTBS (p<0.05).

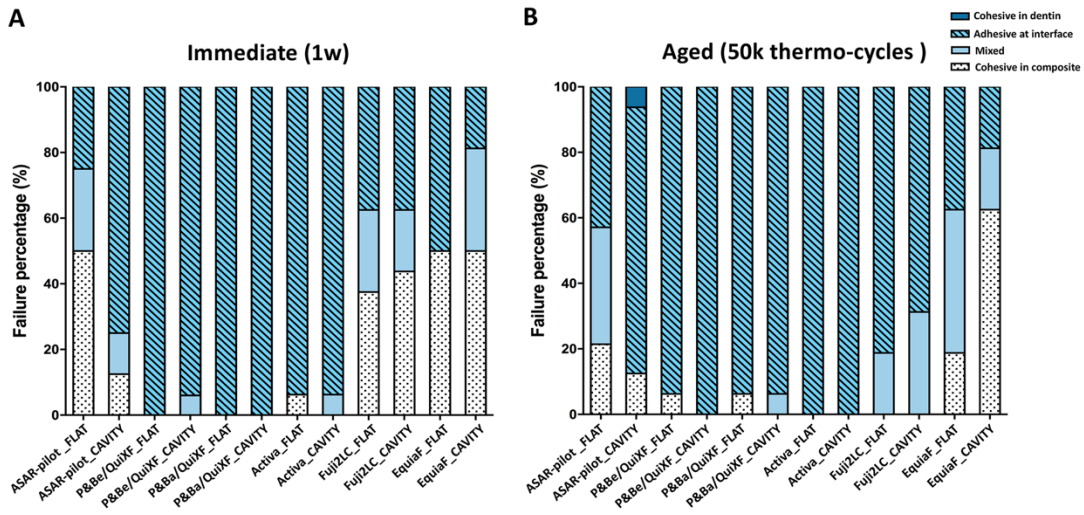

Fig 3 Light microscopy (LM) failure analysis of all μ-specimens of the different experimental restorative systems investigated regarding the immediate μTBS in (A) and the aged μTBS in (B).

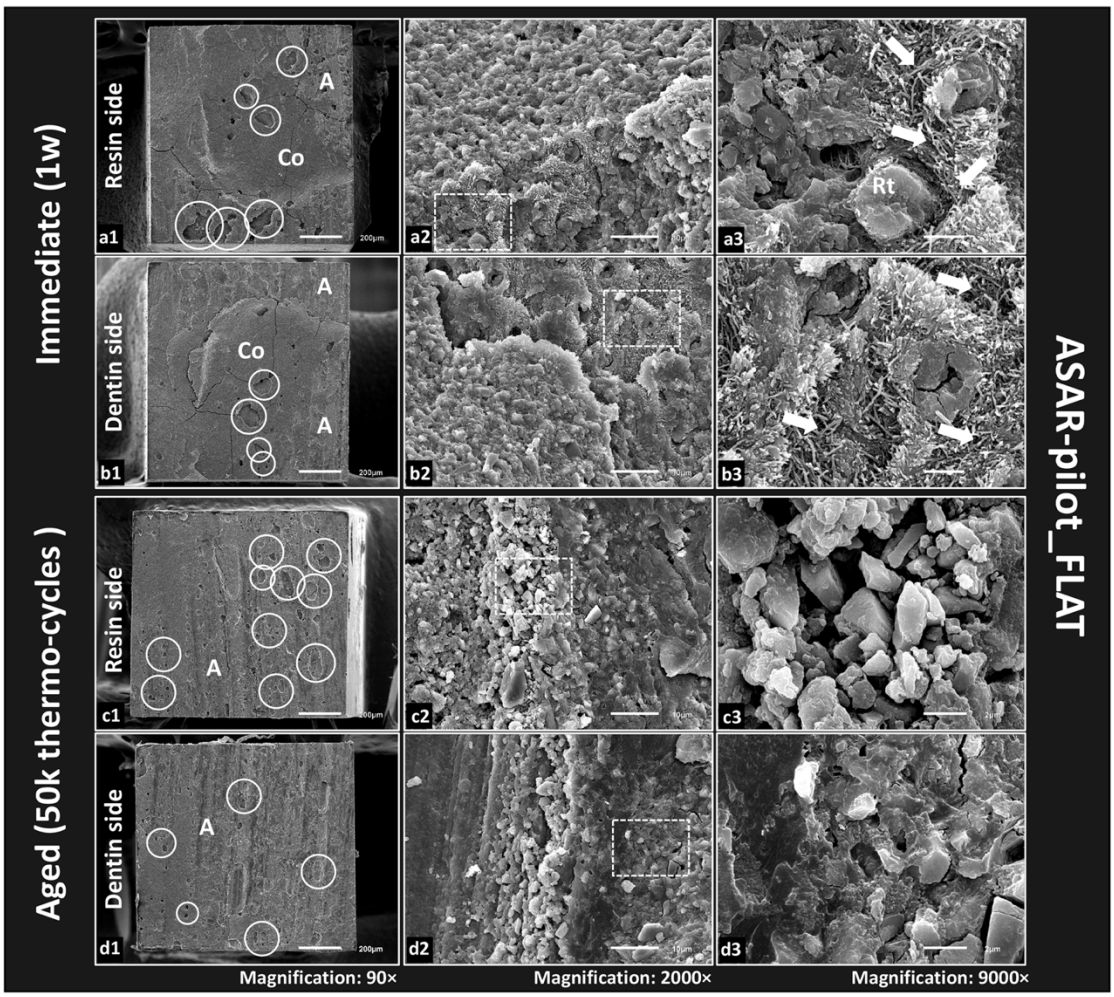

Fig 4 Representative SEM photomicrographs illustrating the fractured sides of ASAR-pilot_FLAT. **(a1 and b1)** Fractured surface (resin and dentin side, respectively) revealing a mixed failure mode with ASAR-pilot remnants (Co) having remained attached to the dentin (b1) side, while also some interfacial porosities were disclosed at both the resin (a1) and dentin (b1) side (white circles) (low-magnification image: 90× original magnification). **(a2 and a3)** Higher magnification of the fractured resin side in (a1), revealing a structure that may represent a resin tag (Rt) and intertubular collagen fibrils (white arrows). **(b2 and b3)** Higher magnification of the fractured dentin side in (b1), revealing intertubular collagen fibers (white arrows). **(c1 and d1)** Fractured surface revealing an adhesive (A) interfacial failure mode with diamond-bur scratches clearly observable at both the resin (c1) and dentin (d1) side, while also interfacial porosities were exposed, in particular at the resin side in (c1) (white circles). **(c2 and c3)** Higher magnification of the fractured resin side in (c1), revealing filler particles in different sizes, ranging from less than 1 μm to around 8 μm. **(d2 and d3)** Higher magnification of the fractured dentin side in (d1), revealing bur-cut scratches filled with restorative material. A: adhesive failure; Co: composite; Rt: resin tag.

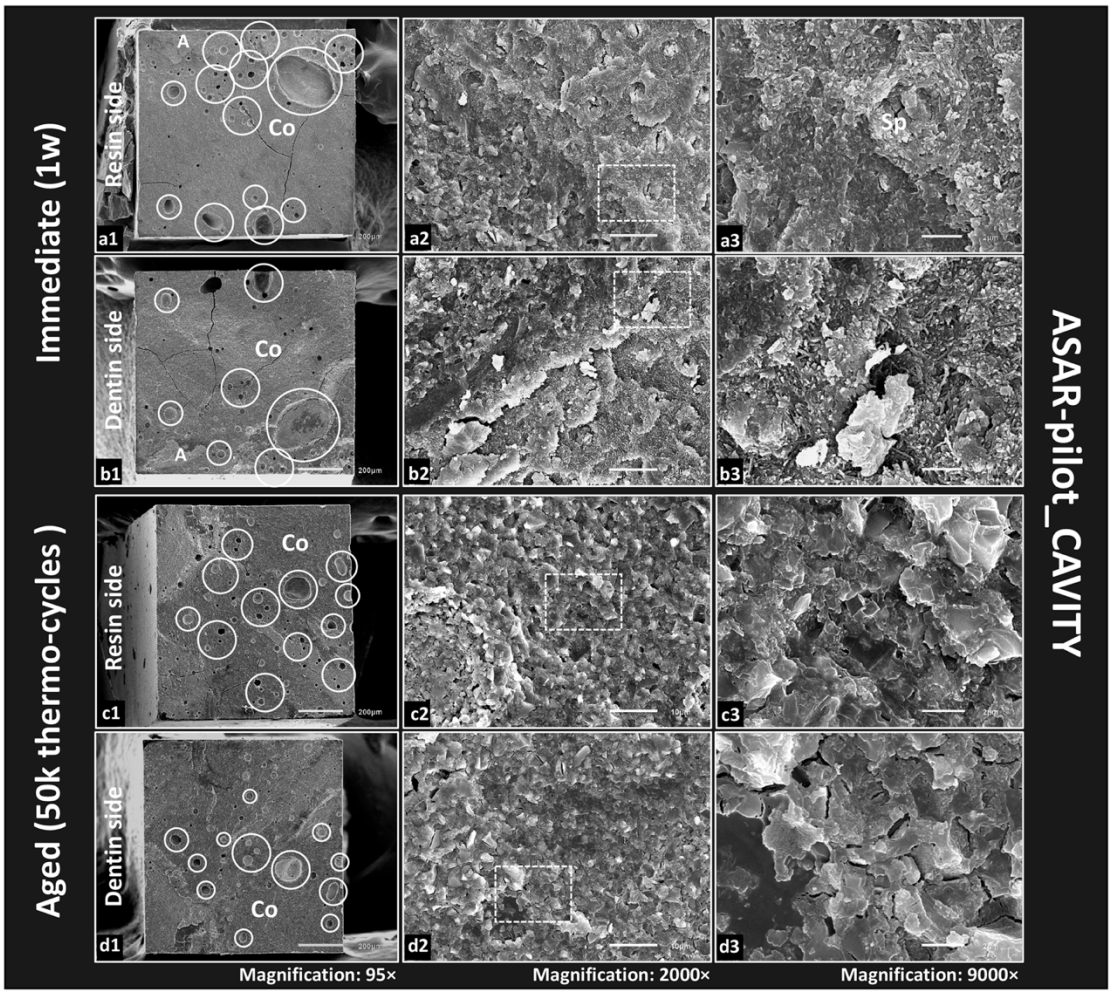

Fig 5 Representative SEM photomicrographs illustrating the fractured sides of ASAR-pilot_CAVITY. **(a1 and b1)** Fractured surface (resin and dentin sides, respectively) mainly revealing a cohesive failure mode in composite (Co), while also interfacial porosities were exposed at both the resin (a1) and dentin (b1) side (white circles) (low-magnification image: 95× original magnification). **(a2 and a3)** Higher magnification of the fractured resin side in (a1), revealing dentin tubules obstructed by a smear plug (Sp). **(b2 and b3)** Higher magnification of the fractured dentin side in (b1). **(c1 and d1)** Fractured surface revealing a cohesive failure mode within composite (Co) and interfacial porosities (white circles). **(c2 and c3)** Higher magnification of the fractured resin side in (c1), revealing the size of most filler particles in this area being less than 5 μm. **(d2 and d3)** Higher magnification of the fractured dentin side in (d1). A: adhesive failure; Co: composite; Sp: smear plug.

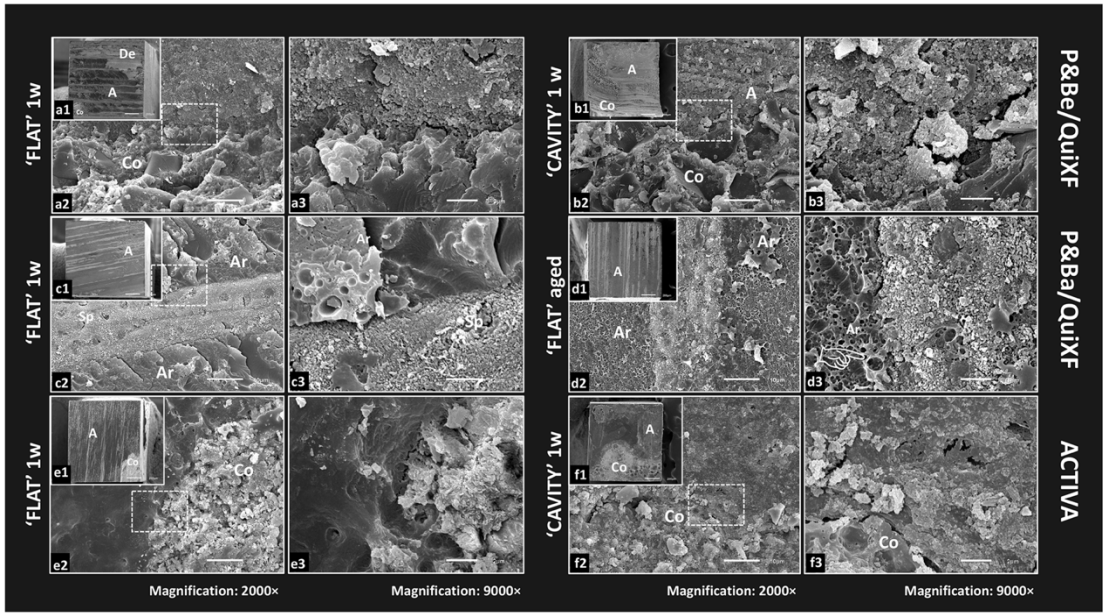

Fig 6 SEM photomicrographs of the fractured dentin side of the experimental restorative systems P&Be/QuiXF, P&Ba/QuiXF and ACTIVA, imaged at low magnification in **(a-f1)** (inserts: 85× original magnification) and at high magnification in **(a-f2)** and **(a-f3)** (2000× and 9000× original magnification, respectively). **(a/a1)** P&Be/QuiXF_FLAT: 'Immediate' fractured surface revealing a predominantly adhesive (A) interfacial failure mode. **(a2/3)** Higher magnification of (a1), revealing composite (Co) that remained attached to the dentin surface. **(b/b1)** P&Be/QuiXF_CAVITY: 'Immediate' fractured surface revealing a mixed failure mode which failed partially adhesively (A) at the interface and partially within the composite (Co). **(b2/3)** Higher magnification of (b1), revealing the size of most filler particles being around 10 µm. **(c/c1/c2)** P&Ba/QuiXF_FLAT: 'Immediate' fractured surface revealing an adhesive (A) interfacial failure mode. **(c3)** Higher magnification of (c1/2), revealing a dentin tubule obstructed by a smear plug (Sp), while also porosities (white circle) within the adhesive resin (Ar) were disclosed. **(d/d1/d2)** P&Ba/QuiXF_CAVITY: 'Aged' fractured surface revealing an adhesive (A) interfacial failure mode. **(d3)** Higher magnification of (d1/2), revealing porosities (white circles) within the adhesive layer (Ar). **(e1-3)** ACTIVA_FLAT: 'Immediate' fractured surface mainly revealing an adhesive (A) interfacial failure mode with remnants of restorative material (Co). **(f/f1)** ACTIVA_CAVITY: 'Immediate' fractured surface revealing a mixed failure mode. **(f2/3)** Higher magnification of (f1), revealing the size of most filler particles being

around 3-4 µm. A: adhesive failure. Ar: adhesive resin; Co: composite; De: dentin; Sp: smear plug.

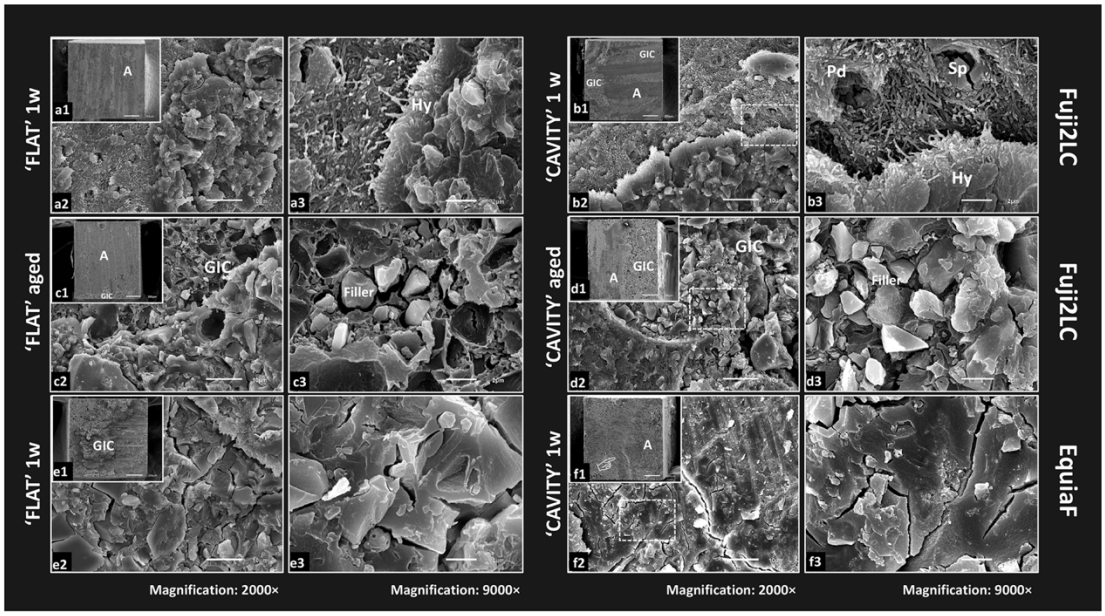

Fig 7 SEM photomicrographs of the fractured dentin side (except for 'aged' Fuji2LC_CAVITY: resin side) of the experimental restorative systems Fuji2LC and EquiaF, imaged at low magnification in **(a-f1)** (inserts: 85× original magnification) and at high magnification in **(a-f2)** and **(a-f3)** (2000× and 9000× original magnification, respectively). **(a/a1)** Fuji2LC_FLAT: 'Immediate' fractured surface revealing an adhesive (A) interfacial failure mode. A typical bur-cut pattern can be observed. **(a2/3)** Higher magnification of (a1), most likely revealing the hybrid layer, indicating clear interaction of Fuji2LC with pre-conditioned dentin. **(b/b1)** Fuji2LC_CAVITY: 'Immediate' fractured surface revealing a mixed failure with glass-ionomer cement (GIC) remaining attached onto the dentin surface. **(b2/3)** Higher magnification of (b1), revealing smear-plugged (Sp) dentin tubules, peritubular dentin (Pd) and most likely the hybrid layer (Hy). **(c/c1)** Fuji2LC_FLAT: 'Aged' fractured surface revealing an adhesive (A) interfacial failure mode. **(c2/3)** Higher magnification of (c1), revealing filler particles in different sizes, ranging from less than 2 µm to more than 10 µm. **(d1-3)** Fuji2LC_CAVITY: 'Aged' fractured surface (resin side) revealing a mixed failure. **(e/e1)** EquiaF_FLAT: 'Immediate' fractured surface revealing a cohesive failure in GIC. **(e2/3)** Higher magnification of (e1),

revealing dehydration artifacts of the water-containing GIC. **(f1-3)** EquiaF_CAVITY: 'Immediate' fractured surface revealing an adhesive (A) interfacial failure mode. Circular scratches represent the bur-cut smear layer produced at the class-I cavity-bottom dentin (f1, 90× original magnification). A: adhesive failure; Hy: hybrid layer; Pd: peritubular dentin; Sp: smear plug.