



Structural/Chemical Characterization and Bond Strength of a New Self-Adhesive Bulk-fill Restorative

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Purpose: The material structure and chemical elemental composition of a new self-adhesive composite hybrid were investigated. The bonding performance when applied on flat (FLAT) vs high C-factor class-I cavity-bottom (CAVITY) dentin and in light-cure (LC) vs self-cure (SC) mode was determined.

Materials and Methods: The self-adhesive bulk-fill composite Surefil One (Su-O; Dentsply Sirona) was compared with the resin-modified glass-ionomer Fuji II LC Improved (Fuji2LC; GC) and the ion-releasing alkasite material Cention N (CentionN; Ivoclar Vivadent). The material structure was examined with SEM and TEM, while the chemical elemental composition was analyzed using EDS. The immediate and aged microtensile bond strength (μ TBS) of Su-O_LC/SC was compared to that of Fuji2LC applied without any pre-treatment, and to that of CentionN applied following bonding with Adhese Universal (AU) (Ivoclar Vivadent) in self-etch mode (AU/CentionN). All restorative materials were bonded onto FLAT and CAVITY dentin. Statistical analysis was performed with the Kruskal-Wallis nonparametric test.

Results: EDS analysis revealed that Su-O was richer in C and P than the reference restorative materials. Applied to FLAT dentin, the significantly highest immediate and aged μ TBS were recorded for AU/CentionN, which were not significantly different only from Su-O_LC. Applied to CAVITY dentin, the significantly highest immediate μ TBS was recorded for AU/CentionN, which did not differ significantly only from Su-O_SC. Su-O_LC bonded to CAVITY dentin suffered from a high incidence of pre-test failures.

Conclusion: While Su-O_LC bonded effectively and durably to FLAT dentin, Su-O_SC bonded more favorably than Su-O_LC in class-I cavities, which was probably related to shrinkage stress variously challenging the respective bond.

Keywords: adhesion, aging, composite, curing, EDS, TEM.

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Self-adhesive tooth-colored restorative materials are greatly desired to further simplify clinical filling procedures.^{14,30,45} Self-adhesive composites do not require a

separate adhesive or any kind of surface pretreatment to bond to tooth substrate.^{19,38} One of the first commercial self-adhesive composites is the self-adhesive flowable com-

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posite Vertise Flow (Kerr; Orange, CA, USA). Its self-adhesiveness depends on two constituents: the functional monomers phosphoric-acid ester methacrylate and glycerol-phosphate dimethacrylate (GPDM).¹⁴ Although Vertise Flow (Kerr) was found to exhibit less microleakage than conventional fissure sealants,³⁹ a clinical trial of sealants revealed a disappointing retention rate of only 62.9% after two years of clinical service.²⁷ In-depth ultramorphological interfacial characterization showed that the bond strength of the self-adhesive composite Vertise Flow (Kerr) to aprismatic enamel and smear-covered tooth surface did not equal that of contemporary adhesives.³² In that study, interfaces at flat surfaces were investigated, while in clinical situations, less favorable cavity configurations challenge the self-adhesively obtained interface to a much greater extent. The sub-optimal interfacial findings correlated with inferior bond strength and interfacial fracture toughness data reported in three other studies.^{9,10,38} Previous *in vitro*⁶ and short-term 6-month clinical⁸ research on another self-adhesive flowable composite, ie, Fusio Liquid Dentin (Pentron, Orange, CA, USA), revealed both a low bond strength and low (33%) clinical success rate when bonded to dentin, indicating unsatisfactory self-adhesiveness. In contrast, a tighter interface with both enamel and dentin was ultrastructurally imaged for the experimental 3M Oral Care (Seefeld, Germany) Exp. 564 self-adhesive filling material, which contains phosphoric acid-6-methacryloxy-hexylesters as self-adhering functional monomers.¹⁹ Unfortunately, actual bond strength measurements were not reported, although they are needed to substantiate the findings of the exploratory interfacial characterization study.¹⁹

Recently, a new self-adhesive bulk-fill composite was commercially introduced as Surefil One (Su-O, Dentsply Sirona; Konstanz, Germany).¹¹ The primary objectives of this study were to ultrastructurally and chemically characterize the filler-matrix configuration of Su-O, as well as to measure its dentin bond strength and durability to flat and high C-factor cavity-bottom dentin when used in light- and self-cure mode. The self-adhering resin-modified glass-ionomer cement (GIC) Fuji II LC Improved (Fuji2LC, GC; Tokyo, Japan) and the recently introduced amalgam-replacing alkasite restorative material Cention N (CentionN, Ivoclar Vivadent; Schaan, Liechtenstein) served as reference materials. The null hypotheses tested were that: 1. there was no structural or chemical difference in filler-matrix configuration between Su-O and the two reference materials; and 2. the immediate and aged bond strength to flat and high C-factor cavity-bottom dentin did not significantly differ from that obtained by the reference materials.

MATERIALS AND METHODS

Preparation of Restorative Material Disks

Four 2-mm-thick disks with a 7-mm diameter were prepared from each restorative material. Two Su-O disks were self-cured by allowing them to set for 6 min in the dark. The two other remaining disks were light cured for 30 s on each

side using the LED light-curing unit Bluephase 20i (Ivoclar Vivadent) with an output of 1200 mW/cm² when used in high mode, as determined and confirmed regularly during the experiment using a Marc Resin Calibrator (BlueLight Analytics; Halifax, Canada). Once prepared, the specimens were kept for 1 h at 100% humidity prior to being embedded in epoxy resin using an 18-mm-thick, 25-mm-diameter Teflon mold. Next, the embedded disks were sequentially polished with silicon carbide papers P320 (Hermes; Hamburg, Germany), P1200, and P4000 (Struers; Ballerup, Denmark), after which they were polished further using 3- and 1- μ m diamond suspensions (Kemet International; Maidenstone, UK) on a synthetic rayon cloth (MicroCloth, PSA, Buehler; Lake Bluff, IL, USA), all using a grinding/polishing machine (Beta Grinder-Polisher, Buehler).

Structural SEM Characterization of Restorative Materials

Two metallographically polished specimens for each light-cured material were coated with gold using a gold-sputter coater (JFC-1300, JEOL; Tokyo, Japan) and imaged using an SEM (JSM-6610LV, JEOL) at an accelerating voltage of 5 kV. SEM photomicrographs were obtained at 5000X original magnification with a working distance of around 10 mm.

SEM/Energy Dispersive X-ray Spectroscopic (SEM/EDS) Elemental Analysis of Restorative Materials

SEM coupled with EDS (FEI-Nova Nanosem 450, FEI; Eindhoven, The Netherlands) was used to evaluate the elemental composition and distribution of two polished disks ($n = 2$) prepared from each material. Disk surfaces of Fuji2LC and CentionN were coated with a thin layer of platinum (Q150T S, Quorum, United Kingdom). In order to avoid peak overlapping of the Pt coating with the phosphorus contained in self-cured Su-O, a thin gold layer was applied instead using a gold-sputter coater (JFC-1300, JEOL). On each disk, two areas of 20 x 20 μ m with a representative filler-matrix configuration were mapped for the chemical elements expected, as based on technical compositional data obtained for the three restorative materials investigated. Drift correction was enabled to avoid drifts that might have occurred on a nanoscale for nonconductive specimens. Areas of 10 x 10 μ m on Su-O disks were additionally characterized using multi-element EDS mapping.

Ultrastructural Transmission Electron Microscopy (TEM) of Restorative Materials

To disclose the ultrastructure of the experimental self-adhesive restorative material Su-O and compare it to that of the two reference restoratives, TEM specimens were prepared by embedding cured material into silicon molds using TEM epoxy resin (Sigma-Aldrich; St Louis, MO, USA). Specimens were light cured for 60 s (30 s per side) using the LED curing light Bluephase 20i (Ivoclar Vivadent) with an output of 1200 mW/cm² when set to high mode. After the specimens were kept for 1 h at 100% humidity, they were processed for TEM, employing routine TEM-specimen preparation and processing, as previously described in detail by Van Meerbeek et al.⁴⁸

Table 1 Restorative materials examined in this study

Materials	Code batch No.	Type	Composition	Application	Filler loading
Surefil One (Dentsply Sirona)	Su-O 1807004175	Self-adhesive bulk fill composite	Aluminum-phosphor-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	Light cure for 20 s with an output of 1200 mW/cm ² Self-cure for 6 min (prior to further specimen processing)	77 wt% 58 vol%
Fuji II LC Improved (GC)	Fuji2LC 171012A	Resin-modified glass-ionomer	2-hydroxyethyl methacrylate, polybasic carboxylic acid, urethane dimethacrylate, dimethacrylate, calcium-aluminum-fluoro-silicate glass, others	Light cure up to 1.8-mm thickness for 20 s with an output of 1200 mW/cm ²	76 wt%
Cention N (Ivoclar Vivadent)	CentionN W94184	Alkaside restorative	Liquid: dimethacrylates, initiators, stabilizers and additives Powder: calcium fluoro-silicate glass, barium glass, calcium-barium-aluminum fluoro-silicate glass, iso-fillers, ytterbium trifluoride, initiators and pigments	1. Application of AdheSE Universal in self-etch mode 2. The mixing ratio for larger cavities: 2 measuring scoops of powder and 2 drops of liquid (corresponding to a weight ratio of 4.6:1) 3. Light cure up to 4-mm thickness for 20 s with an output of 1200 mW/cm ²	78.4 wt% 57.6 vol%

Microtensile Bond Strength (μ TBS) of Restorative Materials to Dentin

A total of 64 noncarious human third molars were collected following informed consent approved by the Commission for Medical Ethics of KU Leuven (file number S57622), stored in 0.5% chloramine-T/water and used within 1 month after extraction. All teeth were randomly subdivided into 8 experimental groups. For the four FLAT groups, the crown was cut 4 mm below the cusp tips, ending with a flat surface in midcoronal dentin. For the four CAVITY groups, the cusp tips were first flattened, upon which a standard box-type class-I cavity (3.5 x 3.5 x 4 mm) was prepared with the cavity bottom again ending in midcoronal dentin. All preparations were made using the MicroSpecimen Former (University of Iowa; Iowa, IA, USA), equipped with a high-speed medium-grit (107 μ m) diamond bur (882, Komet; Lemgo, Germany). A 3.5 x 3.5 x 4 mm bulk buildup of one of the restorative materials was made on the flat dentin surfaces using an addition silicone mold (Aquasil medium body, Dentsply Sirona), while the cavities were likewise bulk-filled with one of the restorative materials as well. The restorative material was applied strictly following the respective manufacturer's instructions (Table 1). Besides application of the experimental self-adhesive bulk-fill restorative material Su-O, resin-modified GIC Fuji2LC was applied without prior poly-alkenoic acid conditioning, while the alkaside restorative material CentionN was applied to dentin previously self-etched using AdheSE Universal (AU) (Ivoclar Vivadent). All

specimens of the three FLAT and three CAVITY experimental groups were light cured using the LED light-curing unit (Bluephase 20i, Ivoclar Vivadent) with an output of 1200 mW/cm². Specimens of two additional experimental groups were made by applying Su-O to FLAT and CAVITY dentin that was allowed to self-cure for 6 min.

Subsequently, the bonded specimens were kept for 1 h 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 micro[μ]-specimens per tooth: 1 x 1 mm wide). For each group, 16 μ -specimens (2 μ -specimens per tooth) were immediately tested, while another 16 μ -specimens were aged for 50,000 thermocycles between two water baths at 5°C and 55°C using a THE-1200 thermocycler (SD Mechatronik; Munich, Germany) prior to testing. For the μ TBS test, the specimens were fixed to a BIOMAT jig with cyanoacrylate 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 failed before actual testing, they were recorded as pre-test failures (ptfs) with each ptf assigned a bond strength of 0 MPa.¹ Statistical differences were examined using Kruskal-Wallis nonparametric statistical analysis with significance set at $\alpha = 0.05$.

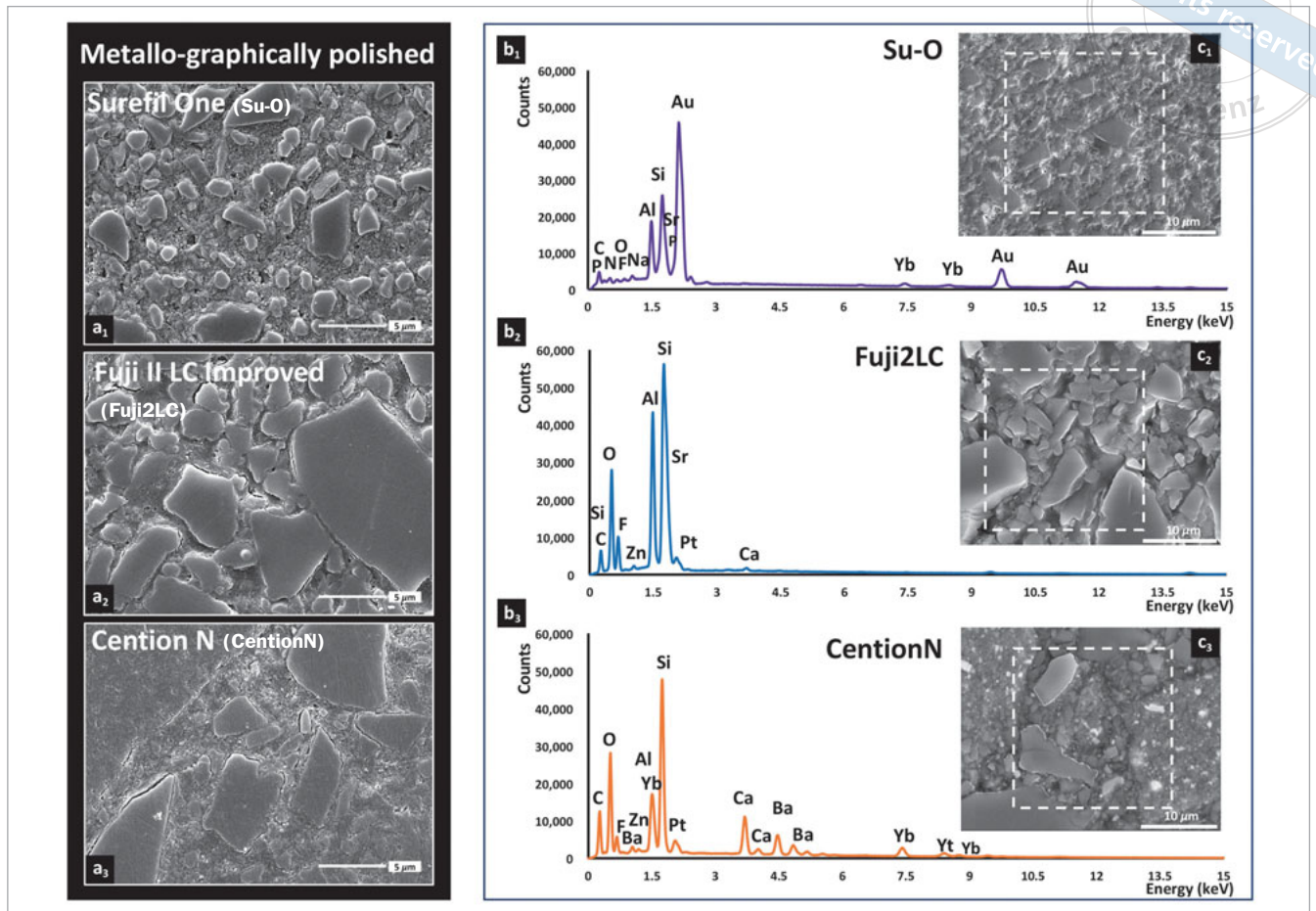


Fig 1 SEM photomicrographs and EDS spectra of the three restorative materials investigated. Metallographically polished surfaces are shown for the self-adhesive bulk-fill composite restorative Surefil One (Su-O) (Dentsply Sirona) in a₁, the resin-modified GIC Fuji II LC Improved (Fuji2LC) (GC) in a₂, and the alcasite restorative Cention N (CentionN) (Vivadent Ivoclar) in a₃. Representative SEM-EDS spectra and high-magnification SEM photomicrograph of the material surfaces are presented in b₁ and c₁ for Su-O, in b₂ and c₂ for Fuji2LC, and in b₃ and c₃ for CentionN. The white square in the high-magnification SEM photomicrograph represents the selected 20 x 20 μm area, at which the EDS spectra were captured. The elements Au and Pt were detected, which should be attributed to the gold and platinum sputter-coating to make the specimens conductive for SEM.

Fractographic Analysis of μTBS Specimens

The fractured specimens were evaluated using stereomicroscopy (Stemi 2000-CS, Zeiss; Oberkochen, Germany) at 50X magnification to classify the failure mode as either cohesive failure in dentin, cohesive failure in composite, adhesive (interfacial) failure, or mixed failure. Representative fractured surfaces exhibiting the most frequent failure mode and originating from specimens with a μTBS close to that of the mean, or from ptf specimens, were selected for SEM observation (JSM-6610LV, JEOL). After fixation using 2.5% glutaraldehyde, the SEM specimens were gradually dehydrated in ethanol and dried with hexamethyldisilazane (Acros Organics, Thermo Fisher Scientific; Geel, Belgium). Afterwards, specimens were coated with gold using the gold-sputter coater (JFC-1300, JEOL). SEM photomicrographs were originally taken at 85-90X, 2000X, and 9000X magnification with a working distance of ca 10 mm.

RESULTS

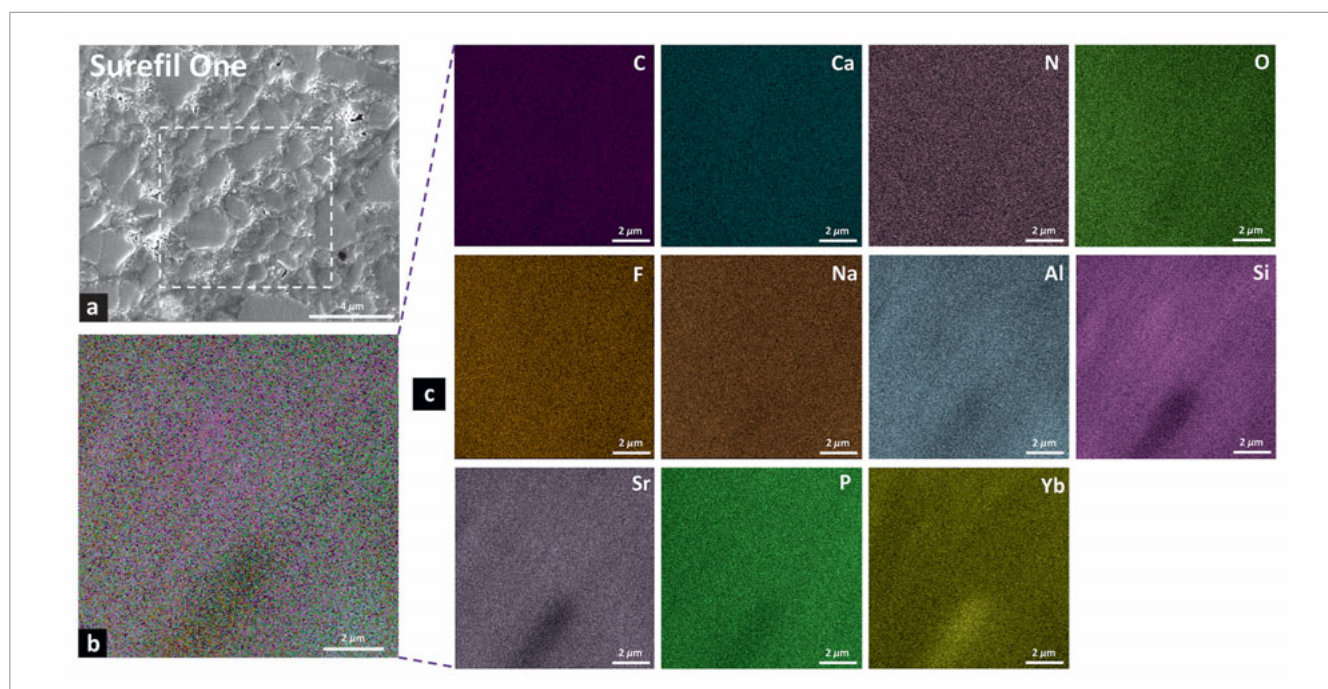
Structural SEM Characterization of Restorative Materials (Fig 1)

The three restorative materials contained filler particles that differed in shape and size (Fig 1a₁₋₃). Visual inspection showed that Su-O presented the most homogeneous filler-matrix configuration with particles that were generally smaller (Fig 1a₁) than those of Fuji2LC (Fig 1a₂) and CentionN (Fig 1a₃). At high magnification (Fig 1c₁₋₃), the maximum filler size in Su-O was estimated to be around 5 μm (Fig 1c₁), while the filler size in Fuji2LC ranged widely (between 1 and 10 μm; Fig 1c₂), and CentionN contained filler particles up to 15 μm (Fig 1c₃). Filler particles were rounder in Su-O (Fig 1a/c₁), while Fuji2LC's filler particles appeared sharper (Fig 1a/c₂), somewhat typical of GICs, and CentionN's particles varied most in shape and size (Fig 1a/c₃). Morphologically, the

Table 2 EDS analysis of elements (mean and standard deviation in wt%) detected in the different restorative materials investigated

	C	Ca	N	O	F	Na	Zn	Al	Si	Sr	Zr	P	Ba	Yb
Su-O	28.2 ± 0.9	0.3 ± 0.6	4.8 ± 1.2	9.5 ± 2.3	3.5 ± 0.7	0.6 ± 0.3	–	7.1 ± 0.8	13.9 ± 0.3	10.6 ± 0.3	–	16.0 ± 0.7	–	5.5 ± 1.7
Fuji2LC	19.7 ± 3.1	0.6 ± 0.1	–	26.9 ± 0.6	8.6 ± 0.7	–	0.2 ± 0.2	11.4 ± 0.4	13.8 ± 0.7	17.8 ± 1.0	1.3 ± 1.5	–	–	–
CentionN	19.0 ± 1.9	7.7 ± 0.8	–	28.2 ± 1.0	4.9 ± 0.6	–	1.5 ± 0.3	2.5 ± 0.2	16.6 ± 0.5	–	–	–	11.2 ± 0.7	8.6 ± 0.7

See Discussion (above) for details of results in bold.

**Fig 2** SEM/EDS mapping of the self-adhesive bulk-fill composite Su-O, with an SEM photomicrograph presented in a, indicating the selected EDS mapping area of 10 x 10 μm, the superimposed EDS map in b, and individual maps for C, Ca, N, O, F, Na, Al, Si, Sr, P and Yb in c, with higher color intensity indicating higher element content. In particular, C, O, Si and P were detected more intensively.

filler load appeared to decrease in the order of Fuji2LC > Su-O > CentionN, although according to the technical information provided by the manufacturers, the filler load of the three restoratives does not differ substantially (Table 1).

SEM/EDS Elemental Analysis of Restorative Materials

Representative SEM/EDS spectra revealed different chemical compositions of the three restorative materials (Fig 1b). Further details per element in weight percentage (mean and standard deviation of four measurements) are presented in Table 2 for each restorative material. The main elements C, O, and Si were detected in all materials, with Su-O being richer in C and poorer in O than the reference restoratives.

Elements detected in Su-O but not detected in the reference restoratives are P (substantial amount), N, and Na (small amounts). All three restoratives contain F, decreasing in the order of Fuji2LC > CentionN > Su-O. Both Su-O and Fuji2LC contain Sr. The relatively high Ca and Ba content is typical of CentionN. Semi-quantitative elemental EDS mapping confirmed the relatively high concentrations of C, O, Si, Sr, and P in Su-O (Fig 2).

Ultrastructural TEM of Restorative Materials

TEM of Su-O disclosed the presence of three kinds of filler particles (Fig 3 and Table 1). Al-P-Sr-Na-F-Si glass exhibited an irregular polyhedral shape (Fig 3b,c: open arrows). YbF particles were irregular and highly electron-dense, with a

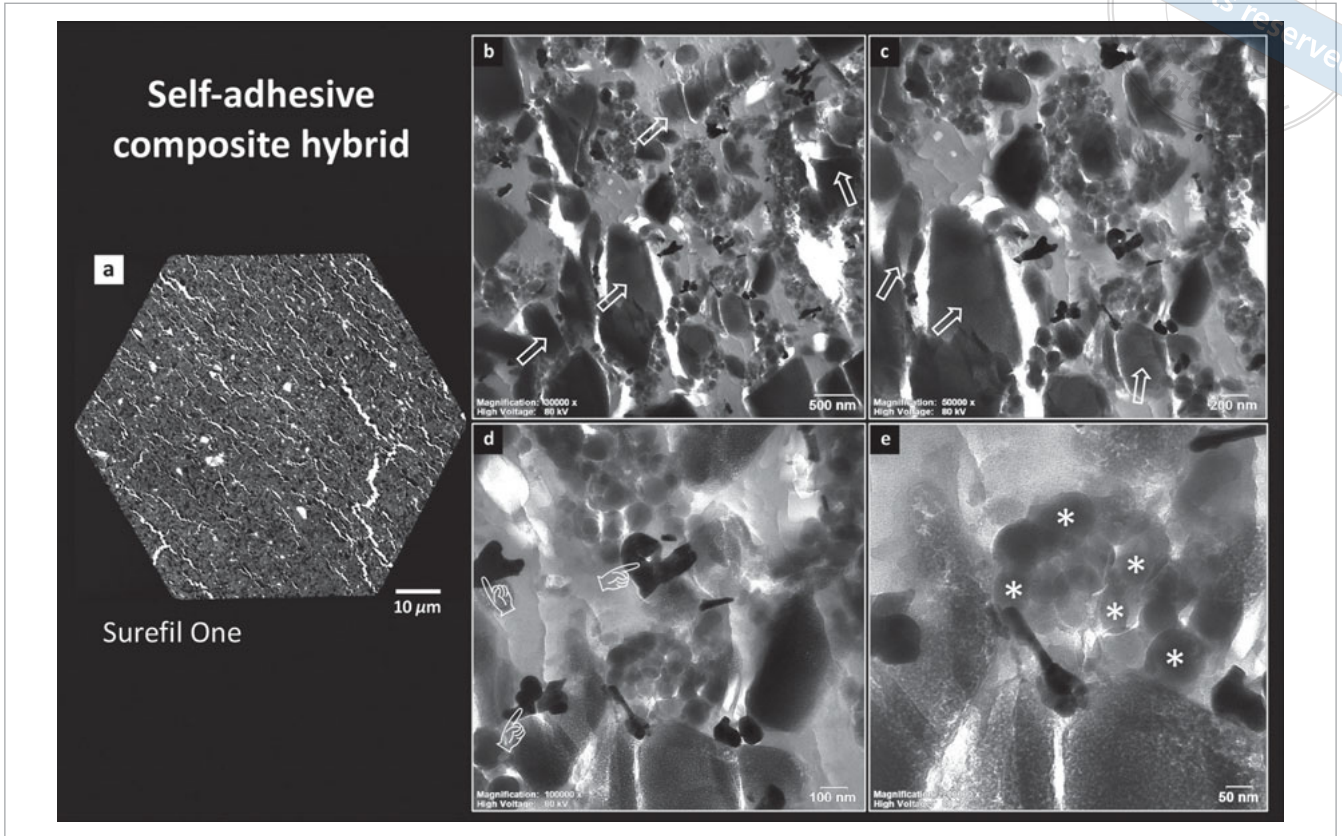


Fig 3 TEM photomicrographs of the self-adhesive bulk-fill composite Su-O at low magnification in a and at increasingly higher magnifications in b-e. Open white arrows: Al-P-Sr-Na-F-Si glass with an irregular polyhedral shape; handpointers: irregular and strongly electron-dense YbF particles with a size of around 200 nm; asterisks: highly dispersed SiO₂ filler particles.

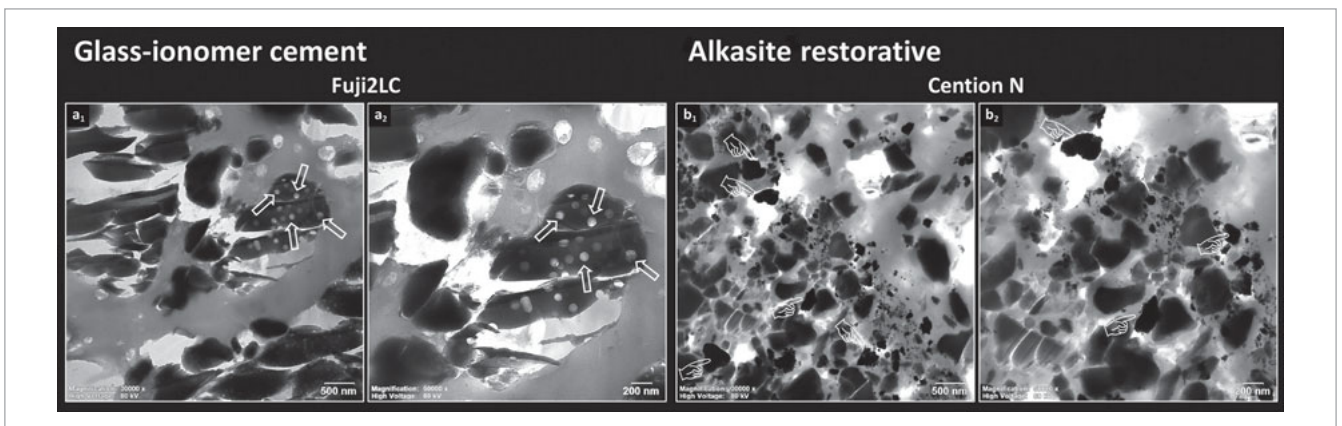


Fig 4 TEM photomicrographs of Fuji2LC at low and higher magnification in (a₁) and (a₂), respectively, and likewise of Cention N in (b₁) and (b₂), respectively. Open with arrows: electron-lucent globules within relatively large GIC glass filler; Handpointers: electron-dense 150-200 nm YbF particles.

size of ca 200 nm (Fig 3d: handpointers). Highly dispersed SiO₂ was found between the larger filler particles (Fig 3e: asterisks). Typical GIC filler was detected in Fuji2LC, with the filler size ranging between 0.5 and 2 μm (Fig 4a). At high magnification, electron-lucent globules were discovered

within the relatively large glass filler particles (Fig 4a: open arrows). High quantities of YbF (Fig 4b: handpointers) were interspersed between larger (0.1-1 μm), irregular, and rounded filler particles, representing Ca-F-Si and Ca-Ba-Al-F-Si glass, as described by the manufacturer (Fig 4b).

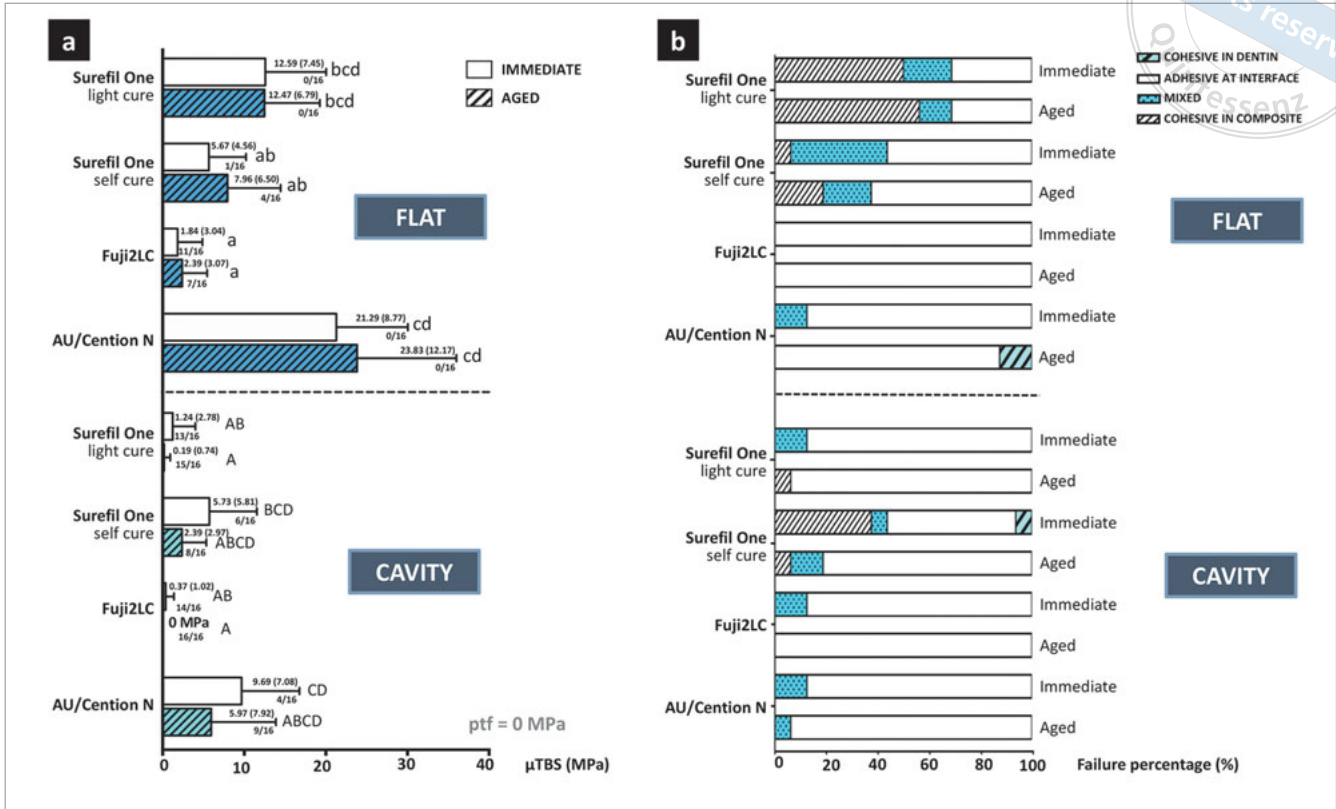


Fig 5 Immediate and aged μ TBS of the three restorative materials to flat (FLAT) and class-I cavity-bottom dentin (CAVITY) in a, with the failure analysis data of all (micro) μ -specimens graphically presented in b. Besides the mean μ TBS, the standard deviation is given within brackets, along with the number of pre-test failures (ptfs) per total number of μ -specimens. μ TBSs with the same small (bonded to FLAT dentin) or capital letter (bonded to CAVITY dentin) are not statistically significantly different ($p > 0.05$). The predominant failure mode recorded was adhesive interfacial failure for all experimental groups, except for light-cured Su-O when bonded to flat dentin and tested both immediately and upon aging.

μ TBS of Restorative Materials to Dentin

The immediate and aged bond strengths to flat and class-I cavity-bottom dentin are graphically presented in Fig 5a.

When bonded to flat dentin, the highest immediate μ TBS was recorded for AU/CentionN; this was only non-significantly different from light-cured Su-O. Fuji2LC presented the lowest immediate μ TBS, mainly due to a high ptf number. Fuji2LC's immediate μ TBS did not significantly differ from the immediate μ TBS recorded for self-cured Su-O. No significant difference in μ TBS was found between light- and self-cured Su-O. Upon aging, the μ TBS did not significantly decrease for any of the experimental groups. Overall, the order of aged μ TBS followed that of immediate μ TBS.

Compared to the restoratives bonded to flat dentin, when bonded to class-I cavity-bottom dentin, lower immediate and aged μ TBS, along with higher ptf numbers, were generally recorded. The highest immediate μ TBS was still recorded for AU/Cention N, which did not significantly differ only from self-cured Su-O. Again, no significant difference in μ TBS was found between light- and self-cured Su-O. Again, no significant difference between immediate and aged μ TBS was measured for any of the experimental groups.

Fractographic Analysis of μ TBS Specimens

When the restoratives were bonded to flat or class-I cavity-bottom dentin, the μ -specimens failed predominantly adhesively at the interface (Fig 5b). Figure 6 depicts representative SEM photomicrographs of fractured immediate (1-week water storage) and aged (50,000 TC) μ -specimens (dentin side) from the different experimental groups when the restoratives were bonded to flat dentin. Low magnification photomicrographs (85-90X original magnification) revealed many voids at the interface (Fig 6a₁, a₃₋₄; arrows). Furthermore, fractured light- and self-cured Su-O surfaces often revealed a particle-filled substance that remained attached to dentin, along with cohesively fractured dentin with obvious collagen-fibril exposure (Fig 6a_{1-d2}). AU/CentionN often presented interfacial failures, as evidenced by the bur scratches clearly observable especially at low magnification (Fig 6e_{1-f2}).

Representative SEM photomicrographs of fractured immediate (1-week water storage) and aged (50,000 TC) μ -specimens (dentin side) from the different experimental groups when the restoratives were bonded to class-I cavity-bottom dentin are shown in Fig 7. Fractured light- and self-cured Su-O surfaces often revealed smear-layer areas that

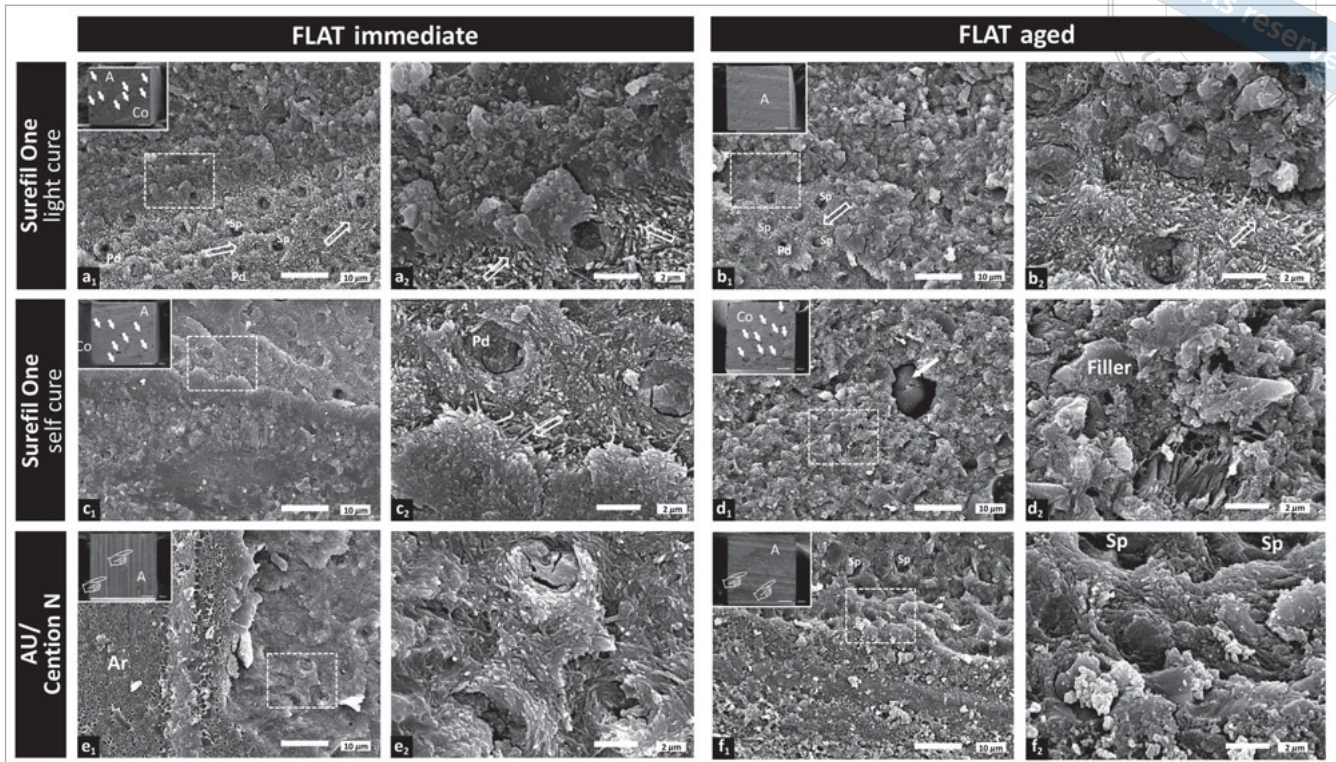


Fig 6 Representative SEM photomicrographs illustrating the fractured dentin sides of the three different experimental materials bonded to flat (FLAT) dentin upon 1-week water storage and 50,000 TC. (a₁) Fractured surface of an immediate light cured Su-O μ -specimen, revealing a mixed failure mode exhibiting a particle-filled substance that remained attached to dentin along with cohesively fractured dentin with clear collagen-fibril exposure (open white arrows). Dentin remained covered with smear debris and was potentially infiltrated by resin. The low-magnification image in the insert (90X original magnification) revealed some interfacial porosities (arrows). (a₂) Higher magnification of the white rectangle in a₁, confirming the particle-filled substance, most likely representing smear-layer parts potentially mixed with Su-O fragments. (b₁) Fractured surface of an aged light cured Su-O μ -specimen, revealing a similar interfacial structure as observed in a_{1,2}. No interfacial porosities were detected in the low-magnification image in the insert (90X original magnification). (c_{1,2}) Fractured surface of an immediate self-cured Su-O μ -specimen, revealing a similar mixed failure mode as revealed for light cured Su-O, along with some interfacial porosities observable in the low-magnification image in the insert (90X original magnification, arrows). (d_{1,2}) Fractured surface of an aged self-cured Su-O μ -specimen, revealing most likely a mainly cohesive failure mode with part of the restorative material having remained attached to the dentin side, along with some interfacial porosities observable in the low-magnification image in the insert (90X original magnification, arrows). (e_{1,2}) Fractured surface of an immediate AU/CentionN μ -specimen, revealing an interfacial failure mode with bur scratches clearly visible (handpointers) in the low-magnification image in the insert (90X original magnification). Failure seems to have occurred close to the actual interface with areas of fractured dentin (e₂) adjacent to areas covered by the adhesive resin (Ar). Abundant micropores were detectable in Ar, most likely representing water droplets absorbed through osmosis in Ar. (f₁) Fractured surface of an aged AU/CentionN μ -specimen, revealing an interfacial failure mode with bur scratches (handpointers) detectable in the low-magnification image in the insert (90X original magnification). Areas of fractured dentin can be observed next to areas covered by a particle-filled substance, most likely representing smear-layer fragments. A: adhesive failure; Ar: Adhesive resin; AU: Adhese Universal; Co: composite; Pd: peritubular dentin; Sp: smear plug.

were potentially mixed with Su-O fragments and together remained attached to dentin (Fig 7a₁₋₂, d₁₋₂). AU/CentionN often presented large areas of fracture at the actual interface, as based upon observation of circular bur scratches at low magnification, and with exposed dentin tubules obstructed by smear plugs, as was observed at high magnification (Fig 7e₁₋₂).

High-magnification SEM photomicrographs of the fractured restorative materials revealed the different shapes and sizes of the restoratives' filler particles, thus confirm-

ing the filler-particle details characterized by SEM, EDS, and TEM. More specifically, the overall filler-particle size of Su-O – with particles < 2 μ m along with nanofiller – was smaller than that of Fuji2LC and CentionN (Fig 8).

DISCUSSION

Thanks to their esthetic potential, composites are the main plastic filling materials currently used in daily dental prac-

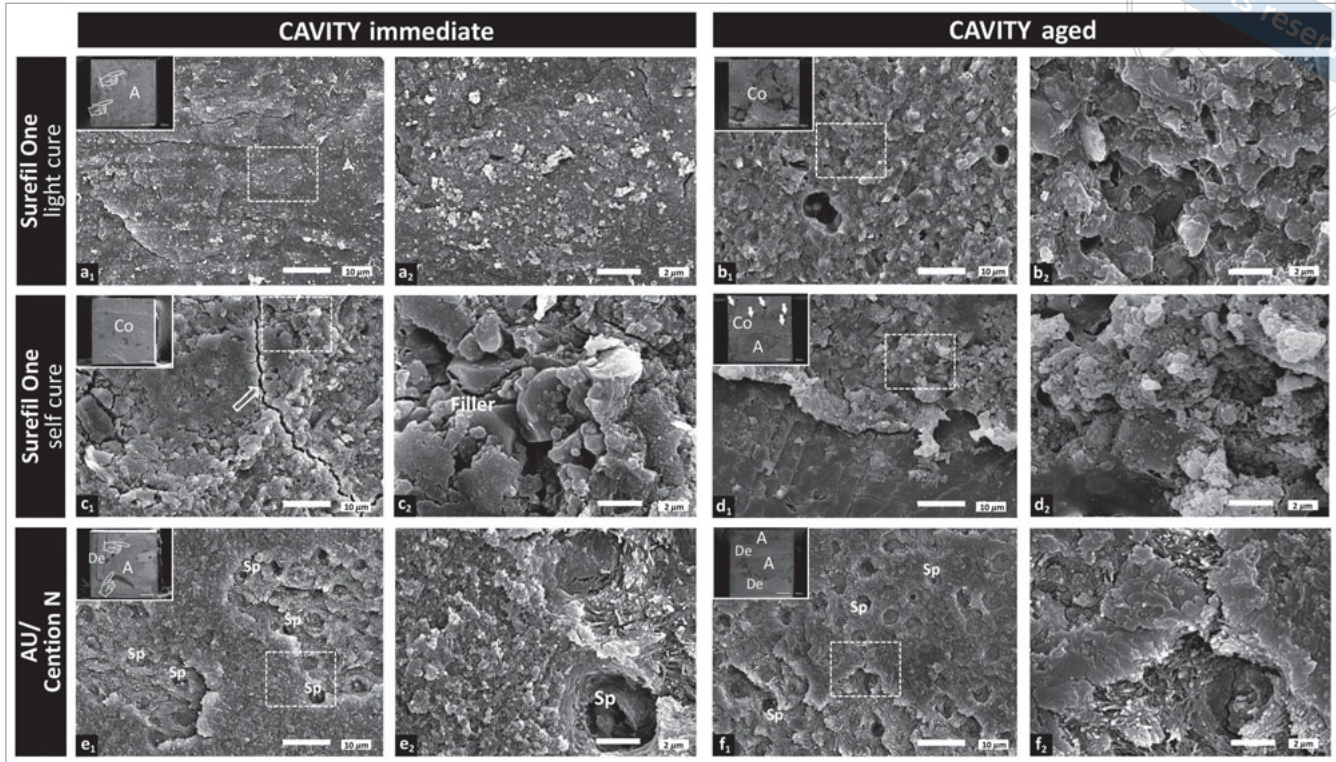


Fig 7 Representative SEM photomicrographs illustrating the fractured dentin sides of the three different experimental materials bonded to class-I cavity-bottom dentin (CAVITY) upon 1-week water storage and 50,000 TC. (a₁) Fractured surface of an immediately light cured Su-O μ -specimen, revealing most likely an interfacial failure mode, as based upon observation of circular bur scratches (handpointers) in the low-magnification image in the insert (90X original magnification). (a₂) Higher magnification of the white rectangle in a₁, revealing most likely smear-layer fragments potentially mixed with Su-O filler particles. (b_{1,2}) Fractured surface of an aged light cured Su-O μ -specimen, revealing a cohesive failure mode within Su-O. This special cohesive failure with partially fractured flat surface could be ascribed to apply two layers of Su-O, which was not common and may be happened when a thin layer of Su-O was bonded onto the cavity and the Capsule Extruder could not continue to squeeze out more material. (c₁) Fractured surface of an immediate self-cured Su-O μ -specimen, revealing a cohesive failure within Su-O. The crack detected within Su-O (open white arrow) should most likely be attributed to a dehydration artifact. (c₂) Higher magnification of the white rectangle in c₁, revealing fractured fragments entailing smaller filler particles (asterisk). (d_{1,2}) Fractured surface of an aged self-cured Su-O μ -specimen, revealing a mixed failure mode with most likely smear-layer parts that were potentially mixed with Su-O fragments and together remained attached to dentin, along with some interfacial porosities (arrows) observable in the low-magnification image in the insert (90X original magnification). (e₁) Fractured surface of an immediate AU/CentionN μ -specimen, revealing a mixed failure mode with large parts having fractured at the actual interface, as based upon observation of circular bur scratches (handpointers) in the low-magnification image in the insert (90X original magnification). (e₂) Higher magnification of e₁, having exposed dentin tubules obstructed by smear plugs. (f_{1,2}) Fractured surface of an aged AU/CentionN μ -specimen, revealing a mixed failure mode with large fractured parts with exposed dentin tubules. A: adhesive failure; AU: Adhese Universal; Co: composite; De: dentin; Sp: smear plug.

tive.⁴⁰ As a new generation, self-adhesive composites are being developed to simplify their clinical application, often claiming to be true amalgam replacements.^{3,18,38} In this study, the recently introduced self-adhesive composite hybrid Surefil One (Su-O) (Dentsply Sirona), allegedly combining the simplicity of GICs with the stability of resin-based composites,¹¹ was investigated. As the first of two reference restorative materials, the resin-modified GIC Fuji2LC was selected, since conventional and resin-modified GICs also auto-adhere to tooth tissue, an adhesion mechanism

based on shallow hybridization combined with ionic interaction of polyalkenoic acid with its multiple carboxyl groups binding to Ca in hydroxyapatite.^{37,47} Second, the recently introduced alkasite amalgam-replacing restorative material CentionN was tested when combined with the universal adhesive AU applied in self-etch mode.

Representing a new generation of self-adhesive restorative materials, the filler-matrix configuration was structurally and chemically characterized using SEM, TEM, and SEM/EDS. Another primary property that was investigated

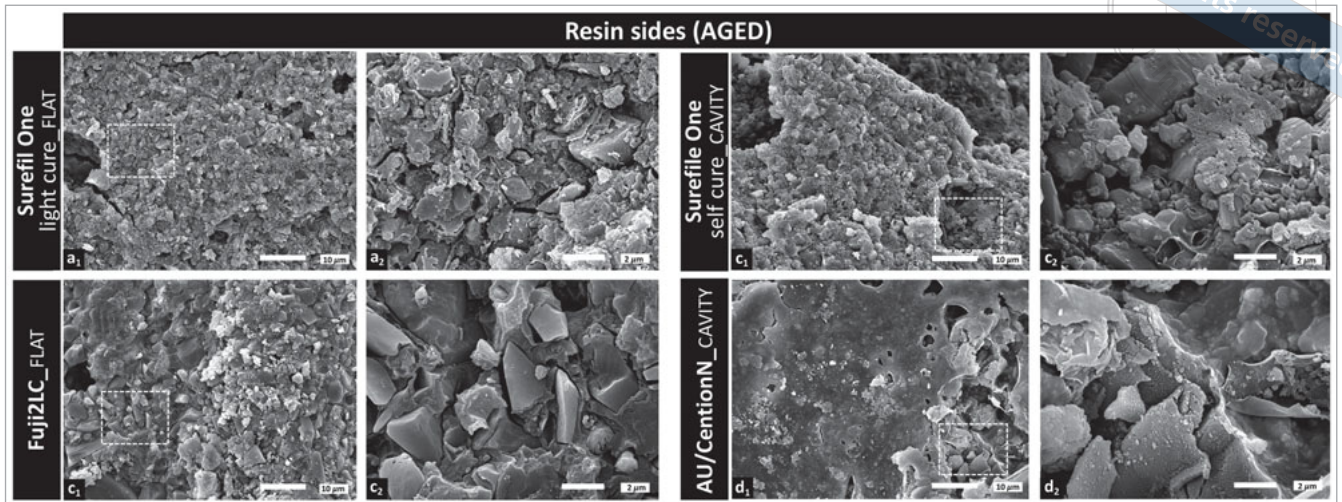


Fig 8 Representative SEM photomicrographs illustrating the fractured resin sides of aged μ -specimens of the different experimental materials bonded to flat dentin (FLAT) or class-I cavity-bottom dentin (CAVITY). (a₁) Cohesively fractured light cured Su-O (2000X original magnification). (a₂) Higher magnification (9000X original magnification) of the white rectangle in a₁, revealing filler particles in different sizes, ranging from less than 1 μ m to around 5 μ m. (b₁) Cohesively fractured self-cured Su-O (2000X original magnification), revealing some porosities within Su-O. (b₂) Higher magnification of the white rectangle in (b₁), revealing filler particles in different sizes, ranging from around 0.5 μ m to 5-6 μ m. (c₁) Cohesively fractured Fuji2LC. (c₂) Higher magnification of the white rectangle in (c₁), revealing the size of most filler particles being around 3-5 μ m. (d₁) Fractured AU/CentionN μ -specimen. (d₂) Higher magnification of the white rectangle in d₁, revealing that the filler particle size of CentionN was larger than the ones of Su-O and Fuji2LC.

in this study was its bond strength and durability upon artificial, accelerated aging. The self-adhesive composite is theoretically expected to interact with tooth substrate through its acidic functional mono/polymer ingredients, not only to provide microretention by etching the substrate surface, but also to ionically bind with Ca in hydroxyapatite. In-depth ultrastructural TEM along with chemical interfacial analysis is needed to fully elucidate the self-adhesiveness of Su-O. A clinically relevant research question this study endeavored to answer was whether Su-O's self-adhesiveness was sufficient to bond to bur-cut and thus smear-layer covered dentin. As an amalgam alternative to bulk-fill posterior cavities, bond strength to flat dentin in the most ideal condition was compared with bonding in high C-factor class-I cavities as a worst-case scenario, with high shrinkage stress severely challenging the bond to cavity-bottom dentin.^{42,43} As Su-O self-cures as well as light cures, both the dual-curing and solely chemical curing mode were investigated. Self-curing composites are known to produce lower polymerization-contraction stress in high C-factor cavities and thus may pose less of a challenge to the bond to cavity-bottom dentin than the dual-cure polymerization mode.^{22,26}

The structural and chemical elemental characterizations using SEM/TEM and EDS, respectively, revealed that the three restorative materials vary widely in filler-particle nature, size, and shape, contrary to the technical information provided by the manufacturers which states that the filler loads of the three materials are quite similar. Hence, the

first null hypothesis, that there would be no structural or chemical difference in filler-matrix configuration between Su-O and the two reference materials, failed to be accepted.

Previous studies reported that filler size and shape greatly influence various properties, including mechanical strength and esthetics.^{20,25} Moreover, filler size and especially filler load – in addition to well-known composite matrix parameters such as type of monomer, their molecular weight and concentration – will together have an effect on polymerization efficacy and resultant polymerization shrinkage.^{5,28} Thus, the present structural and chemical characterization has shed light on the filler-matrix configuration of the three restorative materials investigated here. SEM revealed that Su-O contained relatively large filler particles with a maximum size of ca 5 μ m, while high-resolution TEM disclosed numerous small filler particles ranging between 50 nm and 1 μ m. This finding is confirmed by the technical information provided by Su-O's manufacturer (unpublished communication), revealing that the d₅₀ filler size is 2 μ m with addition of nanofiller. In contrast, Fuji2LC and CentionN consisted of generally larger filler particles, often exceeding 10 μ m, while also containing nanofiller of about 100 nm particle size. These filler characterization data confirmed the trend that dental composites have continuously evolved from initially macrofiller-based composites towards nanohybrid compositions.^{14,20} In general, small filler sizes can result in enhanced filler/matrix surface interactions,⁴ thus also resulting in greater light scattering. On the other hand, Fujita et al⁴⁵ demonstrated that a decrease in particle size of silica

filler can lead to increased transmission of visible light. The latter finding may indicate that the smaller filler size of Su-O might have improved light-penetration depth, which would facilitate a sufficiently high degree of conversion.

Semi-quantitative EDS revealed that most common element found in the three restorative materials was C, with Su-O containing about 28 wt% C, which is substantially higher than the nearly 19 wt% C present in the two reference materials. Since C is part of the organic resin network,¹⁷ the high C concentration in Su-O appeared to suggest that Su-O has a lower filler content than the two other materials. However, this was not evident in the filler-load data provided by the manufacturers, as presented in Table 1. The filler load of the three restorative materials was indeed quite similar. The fact that Si was found in relatively high amounts in the three restorative materials was expected, as silica filler and/or silicon-based glass filler are very common fillers of many dental restorative materials. The significantly lower O content in Su-O was striking. This finding disagrees with reports by other authors, who observed 20-40 wt% O in commonly used composites.^{36,41} Characteristic of Su-O is its relatively high P content, which should be largely attributed to P-containing glass filler, but could also be related to the bifunctional acrylate mentioned by the manufacturer, potentially referring to a phosphate-based functional monomer added to produce Su-O's self-adhesiveness. The presence of N in Su-O may point to the addition of some acrylamide monomers, known for their better hydrolytic resistance. Other elements, such as Al, P, Sr, Na, F, and Si, are part of Su-O's aluminum-phosphor-strontium-sodium-fluoro-silicate glass filler; Si is also part of the silica filler. Both kinds of filler are mentioned by the manufacturer. To achieve radiopacity, Yb was added to Su-O. A selected area of Su-O, containing both resin matrix and filler, was additionally subjected to EDS multi-elemental mapping (Fig 2); area differences for signals representing filler and surrounding resin were quite low, indicating a homogeneous filler-matrix configuration.

As expected, Fuji2LC, as resin-modified GIC, contains F, Al, and Sr as part of GC's classical GIC filler. Of all three restorative materials investigated, Fuji2LC contains F in the highest concentration, which upon release renders Fuji2LC anti-cariogenic.²⁹ Peculiar to the alkasite material, CentionN contains a relatively large amount of Ca, most likely in keeping with the alleged remineralization potential. When the oral environment's pH is low (acidic), the released Ca ions are claimed to slow tooth demineralization.^{23,24} Because of their high atomic number, Ba and Yb were added to CentionN to achieve adequate radiopacity.^{17,25,31}

An essential test for a self-adhering restorative material is to assess its immediate and aged bond strength to dentin. More specifically, the best bonding performance was found when the restorative materials were bonded to flat dentin, where the bond was hardly challenged by polymerization shrinkage of the material in this low C-factor model. Previous research calculated a C-factor as low as 0.18 when employing a similar study set-up.⁴³ While the separate application of a self-etch adhesive resulted in a better

bonding performance for CentionN, the relatively high variance recorded for it did not make its bond strength significantly better than that recorded for light-cured Su-O. However, both their immediate and aged μ TBS were significantly higher than that of self-cured Su-O and Fuji2LC, with the latter applied without previous polyalkenoic-acid conditioning, which is known to promote its bonding effectiveness.^{7,12} The better self-adhesiveness of light- than self-cured Su-O may be due to the lower mechanical properties produced by self-curing. A μ TBS test should be regarded as a strength test of the whole μ -specimen assembly, including the restorative material in this particular case. This hypothetical explanation should be confirmed by additional mechanical strength testing. It is worth mentioning that all restorative materials resisted accelerated aging, since no drop in the optimum bond strength was recorded after 50,000 thermocycles, which lasted about 6 weeks and thus should be seen as a rather severe aging regime.

As the manufacturer intends Su-O to be used as a bulk-fill restorative material, its immediate and aged bond strength to class-I cavity-bottom dentin (high C-factor, high polymerization shrinkage) was additionally determined in this study. The additional polymerization shrinkage clearly challenged the bond to cavity-bottom dentin,^{33,43,44} since significantly lower μ TBS along with high ptf numbers were recorded for the different restorative materials. The highest μ TBS and lowest ptf number was recorded for AU/CentionN, suggesting that a separately applied adhesive appeared to better resist the polymerization-shrinkage challenge. In contrast to bonding to flat dentin, self-cured Su-O outperformed light-cured Su-O, which most likely should be attributed to the slower polymerization and the lower shrinkage stress developed during polymerization. Previous studies indeed demonstrated that light-cured composites produced more contraction stress in high C-factor class-I cavities, as shown by a greater tendency to separate from the cavity wall than their self-cured counterparts.^{16,22} In fact, light-initiated polymerization is fast, during which restorative (and luting) composites undergo a short gel stage, not allowing the materials to flow fluently and the resin molecules to easily dissipate the tension generated by polymerization contraction.^{5,26} The reason why the self-cured composites produce a lower polymerization-contraction stress should principally be attributed to a lower polymerization rate.^{13,26} Therefore, when Su-O bonded to flat dentin and shrank nearly unrestrictedly, the light-cure mode resulted in better bonding performance than the self-cure mode, while the opposite effect was recorded when Su-O was bonded to a high C-factor class-I cavity. Furthermore, self-curing Su-O requires more time to reach maximum polymerization, so that dentists would need to wait longer, thus somewhat contradicting the concept of simplified treatment. Nevertheless, it should finally be mentioned that with the exception of the bulk-fill flowable composite SDR (Dentsply Sirona), many commercially marketed bulk-fill composites tested in previous research failed in the same class-I cavity μ TBS test model,^{43,44} indicating that this test indeed involves a worst-case scenario.

It is noteworthy that Fuji2LC performed worst when bonded to both flat and cavity-bottom dentin; in the latter situation, all μ -specimens failed upon aging prior to testing (100% ptf). If dentin had been pretreated with a polyalkenoic-acid conditioner, the low bond strengths for Fuji2LC recorded in this study could have been higher, since the relatively thick/compact bur smear layer would have interfered less and enabled the GIC to have interacted more intimately with the underlying dentin surface.^{7,47}

Nevertheless, it is not surprising that good bonding performance was recorded for AU/CentionN. Three main factors may have contributed to this. First, the universal adhesive AU must have wetted the dentin surface better, enabling the acidic functional monomer 10-methacryloyloxydecyl dihydrogen phosphate (10-MDP) to chemically interact with Ca in hydroxyapatite.^{49,50} In addition, SEM fracture analysis disclosed the formation of a hybrid layer along with resin tags, indicating that AU partially demineralized the dentin surface and interdiffused into partially exposed collagen fibrils. Second, previous studies found that a relatively thick adhesive layer with shock-absorbing or elastic potential^{34,36} was less affected by the C-factor when bonded to class-I cavity-bottom dentin.⁴² The adhesive acted as an intermediary stress reliever to partially compensate for the shrinkage stress induced.² While universal adhesives like AU generally have a rather thin film thickness, their separate application and polymerization may have contributed to better withstanding the high polymerization shrinkage induced in a class-I cavity. Such a stress-absorbing role of adhesives cannot be taken over by the self-adhesive material. A third factor that may explain the higher bond strength of AU/CentionN might be the special photoinitiator Ivocerin contained in Cention N, which is a dibenzoyl germanium derivative (manufacturer's data).²³ Previous research pointed out that the germanium-based photoinitiator has a higher photocuring reactivity than camphorquinone, with a higher light-absorption potential in the 400 to 450 nm wavelength range.^{21,35}

Altogether, the second null hypothesis – that the immediate and aged bond strength to flat and high C-factor cavity-bottom dentin did not significantly differ from that obtained by the reference materials – failed in part to be accepted, in particular as compared to AU/CentionN.

CONCLUSION

Ultrastructural/chemical characterization revealed that the self-adhesive composite hybrid Su-O mainly consisted of small-sized filler particles, which were rich in Al, P, Sr, and Si. Good bonding performance of Su-O was achieved when applied in light-curing mode on flat dentin. Nevertheless, Su-O applied in light-curing mode presented lower bond strength when bonded following the worst-case scenario onto class-I cavity-bottom dentin. Self-cured Su-O bonded more effectively to cavity-bottom dentin, as did CentionN when it was bonded using the separately applied and light-curing universal adhesive UA. Since self-adhesive bulk-fill

composites are relatively new materials, more *in vitro* experiments, eg, to fully elucidate the underlying mechanisms of self-adhesiveness, but also short- and long-term clinical data are urgently needed.

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Clinical relevance: Filling class-I cavities in bulk without adhesive is more favorable when the self-adhesive composite hybrid is allowed to self-cure.