Eight-year Microtensile Bond Strength to Dentin and Interfacial Nanomechanical Properties of a One-step Adhesive

Keiichi Hosaka[a] / Antonin Tichy[b] / Daisuke Araoka[c] / Wurihan[d] / Yo Shibata[e] / Masaomi Ikeda[f] / Celso Afonso Klein Jr[g] / Junji Tagami[h] / Masatoshi Nakajima[i]

Purpose: To evaluate the microtensile bond strength (μTBS) of a one-step self-etch adhesive (1-SEA) to dentin and its interfacial nanomechanical properties after 8 years of water storage.

Materials and Methods: Flat coronal dentin surfaces of extracted human third molars were bonded with a 1-SEA (Clearfil S3 Bond Plus, CS3+) and built up with a hybrid resin composite (Clearfil AP-X). After storage in water for 24 h or 8 years, non-trimmed stick-shaped specimens were fabricated from the central part of each bonded tooth and subjected to the μTBS test at a crosshead speed of 1.0 mm/min. Failure modes and the morphology of debonded interfaces were analyzed using a scanning electron microscope (SEM). In addition, the elastic modulus (E) and hardness (H) of the adhesive layer and the resin composite were determined by an instrumented nanoindentation test. The acquired μTBS, E, and H data were statistically analyzed using t-tests to examine the effect of storage time ($\alpha = 0.05$).

Results: The 8-year μTBS was slightly lower than that after 24 h, but the difference was not significant ($p = 0.123$). The SEM observation of debonded surfaces after 8 years revealed extrusions and lacunas. E and H of the adhesive layer and the resin composite significantly decreased over the 8-year water storage ($p < 0.001$).

Conclusions: Although 8 years of water storage did not decrease the μTBS of CS3+ significantly, the observed failure mode patterns and significantly decreased nanomechanical properties indicated resin degradation of the adhesive and the resin composite.

Keywords: bond durability, microtensile bond strength, one-step adhesive, dentin, resin composite, nanohardness, elastic modulus, failure mode.

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Direct composite restorations, from simple fillings to large, complex restorations, have been widely accepted as a valid minimally invasive treatment option.[4,18,35,40] In the past decades, adhesives have been simplified and optimized for longevity of composite restorations.[11,12,21] Recently, one-step self-etch adhesives (1-SEAs) have gained popularity in the field of restorative dentistry over conventional multistep systems. However, the bonding durability of

[a] *Professor and Chair, Department of Regenerative Dental Medicine, Tokushima University Graduate School of Biomedical Sciences, Tokushima, Japan. Idea, hypothesis, experimental design, performed the bond strength test, wrote the manuscript.*

[b] *PhD Candidate, Department of Cariology and Operative Dentistry, Graduate School of Medical and Dental Sciences, Tokyo Medical and Dental University, Tokyo, Japan; Institute of Dental Medicine, First Faculty of Medicine of the Charles University and General University Hospital in Prague, Czech Republic. Contributed substantially to discussion, co-wrote the manuscript.*

[c] *Adjunct Lecturer, Department of Cariology and Operative Dentistry, Graduate School of Medical and Dental Sciences, Tokyo Medical and Dental University, Tokyo, Japan. Performed SEM observation, proofread the manuscript.*

[d] *Postdoctoral Fellow, Department of Conservative Dentistry, Division of Biomaterials and Engineering, Showa University School of Dentistry, Tokyo, Japan. Performed the nanoindentation test, proofread the manuscript.*

[e] *Professor, Department of Conservative Dentistry, Division of Biomaterials and Engineering, Showa University School of Dentistry, Tokyo, Japan. Consulted on idea and experimental design, proofread the manuscript.*

[f] *Junior Associate Professor, Department of Oral Prosthetic Engineering, Graduate School of Medical and Dental Sciences, Tokyo Medical and Dental University, Tokyo, Japan. Performed the statistical analysis, proofread the manuscript.*

[g] *Professor, Department of Operative Dentistry, Lutheran University of Brazil, Cachoeira do Sul, RS, Brazil. Contributed substantially to discussion, proofread the manuscript.*

[h] *Professor Emeritus, Department of Cariology and Operative Dentistry, Graduate School of Medical and Dental Sciences, Tokyo Medical and Dental University, Tokyo, Japan. Consulted on idea, contributed substantially to discussion, proofread the manuscript.*

[i] *Adjunct Lecturer, Department of Cariology and Operative Dentistry, Graduate School of Medical and Dental Sciences, Tokyo Medical and Dental University, Tokyo, Japan. Consulted on idea and experimental design, contributed substantially to discussion, proofread the manuscript.*

Correspondence: *Professor Keiichi Hosaka, Department of Regenerative Dental Medicine, Tokushima University Graduate School of Biomedical Sciences, 3-18-15, Kuramotocho, Tokushima, 770-8504, Japan. Tel: +81-88-633-7339; e-mail: hosaka@tokushima-u.ac.jp*

Table 1 Composition of materials used in this study

Material	Composition	Manufacturer's instructions
Clearfil S3 BOND Plus (CS3+, Kuraray Noritake; Tokyo, Japan)	10-MDP, HEMA, bis-GMA, hydrophilic dimethacrylate, hydrophobic methacrylate, water, ethanol, colloidal silica, CQ, new photo initiator, photo polymerization accelerator, chemical polymerization accelerator, sodium fluoride	Apply for 10 s Dry with mild air pressure Light cure for 10 s
Clearfil AP-X (Kuraray Noritake)	Bis-GMA, TEG-DMA, CQ, photo initiators, pigments, silanated barium glass, silanated silica	Light cure for 40 s

10-MDP: 10-methacryloloxydecyl dihydrogen phosphate; HEMA: 2-hydroxyethyl methacrylate; bis-GMA: bisphenol A glycidyl methacrylate; CQ: Camphorquinone; TEG-DMA: triethylene glycol dimethacrylate.

1-SEAs may diminish over time because of their higher hydrophilicity, which is mainly associated with the formula, which includes polar acidic functional monomers, as well as hydrophilic monomers, such as 2-hydroxyethyl methacrylate (HEMA), and water.[31] In particular, HEMA-containing 1-SEAs are susceptible to water absorption,[13] and previous studies revealed a significant reduction in their elastic modulus and tensile strength after storage in water.[13,14] The hydrated adhesive layer is prone to degradation, resulting in decreased bond strengths or, ultimately, in debonding under repeated loading.[17]

The durability of 1-SEAs was examined in several studies which reported a significant decrease in bond strengths of most HEMA-containing 1-SEAs after 1 year,[6,10,15,20,27,34] 2 years,[24,38] and 5 years[5] of storage in water. The decrease in bond strength was generally accompanied by an increase in the number of failures in the adhesive resin, and two studies observed a deterioration of the adhesive-composite interface.[10,27] These findings indicate that the long-term reduction in bond strength is due to the degradation of the hydrated adhesive layer, so its mechanical properties should be examined. Given the thin adhesive layers of 1-SEAs,[1,29] standard macro-scale mechanical tests are not appropriate for detailed assessment of resin-dentin interfaces. Instead, nano-scale testing methods, such as nanoindentation, which enable very precise selection of the measurement location, should be preferred, because they ensure that the desired local material properties of heterogeneous structures are measured.[26] Dos Santos et al[3] found that the nanomechanical properties of interfaces between an etch-and-rinse adhesive and dentin decreased over 6 months of storage. In contrast, Freitas et al[7] reported no significant difference in elastic moduli or nanohardness of four adhesives after 6-month water storage. However, to the best of our knowledge, the nanomechanical properties of 1-SEAs after long-term water storage have not been examined.

The purpose of this in vitro study was to evaluate the durability and interfacial nanomechanical properties of a HEMA-containing 1-SEA after 8 years of water storage using the microtensile bond strength (μTBS) test and instrumental nanoindentation. The null hypotheses tested were that the 8-year water storage would not affect: 1. the μTBS to dentin, or 2. the nanomechanical properties of the adhesive layer.

MATERIALS AND METHODS

Specimen Preparation

Ten extracted caries-free human molars were collected according to protocol number 571, and approved by the Human Research Ethics Committee of Tokyo Medical and Dental University. They were stored frozen until use, which was within one month of extraction. First, mid-coronal flat dentin surfaces were exposed, leaving the surrounding enamel; then the dentin surfaces were prepared using a 600-grit silicon carbide (SiC) paper to create a standardized smear layer. A HEMA-containing 1-SEA (Clearfil S3 Bond Plus [CS3+], Kuraray Noritake; Tokyo, Japan) was applied to the prepared dentin surfaces with an enamel margin, air dried, and light cured for 10 s, according to the manufacturer's instructions (Table 1). The bonded surfaces were built up with a hybrid resin composite (Clearfil AP-X, shade A2, Kuraray Noritake) to a height of 6 mm in three increments. Each increment was light cured for 40 s using an LED light-curing unit (Valo, Ultradent; South Jordan, UT, USA) with a light output of ~1000 $mWcm^{-2}$. The specimens leaving the surrounding enamel (ie, bulk specimens) were then randomly divided into two experimental groups (n = 5) and stored in distilled water at 37°C for either 24 h or 8 years. The distilled water was changed every two months.

Microtensile Bond Strength (μTBS) Test

After the respective storage periods, the specimens were sectioned into sticks (bonded area: 1.0 mm x 1.0 mm) using a low-speed diamond saw (Isomet, Buehler; Lake Bluff, IL, USA) under water cooling. Five sticks from the central area of each tooth underwent testing, adding up to a total of 25 sticks per experimental group. The sticks were individually bonded to a microtensile testing jig with a cyanoacrylate glue (Model Repair II Blue, Dentsply-Sankin; Tokyo, Japan), and stressed in tension at a crosshead speed of 1.0 mm/min until failure using a table-top testing

Table 2 Microtensile bond strength results (mean ± SD) and failure mode distribution

Storage time	24 h	8 years
μTBS (MPa)	59.1 ± 10.0	53.6 ± 11.7
Failure mode distribution (%) (CoD / Ad-D / CoAd / Ad-RC / CoRC)	32 / 8 / 0 / 60 / 0	8 / 12 / 4 / 64 / 12

μTBS: microtensile bond strength; CoD: cohesive failure in dentin; Ad-D: interfacial failure between dentin and the adhesive; CoAd: cohesive failure in the adhesive layer; Ad-RC: interfacial failure between the adhesive and the resin composite; CoRC: cohesive failure in the resin composite.

machine (EZ-SX, Shimadzu; Kyoto, Japan). The fractured specimens were desiccated for 24 h, mounted on brass stubs, coated with a 3-nm layer of gold, and observed using a scanning electron microscope (SEM; JSM-5310, JEOL; Tokyo, Japan) to assess their failure modes and the morphology of debonded interfaces. The failure modes were classified as follows: 1. cohesive failure in dentin (CoD); 2. interfacial failure between the adhesive and dentin (Ad-D); 3. cohesive failure in the adhesive layer (CoAd); 4. interfacial failure between the adhesive and the resin composite (Ad-RC); 5. cohesive failure in the resin composite (CoRC). The proportions of failure modes at each debonded surface were determined using SemAfore software (version 5.2, Insinooritoimisto Rimppi; Ojakkala, Finland), and a mean percentage of each failure mode was calculated.

Nanoindentation Test

Four additional teeth were prepared as described in the section on specimen preparation, and randomly divided into two experimental groups (n = 2) according to the length of water storage. After 24 h or 8 years, the bulk specimens were embedded in epoxy resin (Epoxicure Resin, Buehler) and sectioned perpendicular to the resin-dentin interfaces with a low-speed diamond saw. Then, each half of the each specimen was polished with a series of SiC papers (up to 2000 grit) and diamond pastes with a particle size decreasing down to 0.25 μm (Struers; Ballerup, Denmark).

Nanoindentation tests were performed at the resin-dentin interface using a diamond pyramidal Berkovich tip attached to a quantitative nanomechanical testing instrument (TI 950 TriboIndenter, Hysitron; Eden Prairie, MN, USA) interfaced with an atomic force microscope (AFM), according to the method presented by Sato et al.[26] Fused quartz was used as the standard calibration material. The indenter was positioned to the central region of the adhesive layers, and 5 measurements were performed per specimen for total of 20 measurements per experimental group. The measurements were performed no closer than 10 μm apart to avoid any influence of the residual stress from adjacent indentations. Similarly, the measurements were performed in the resin composite near the interface. The thermal drift rate with a minimum preloading force of 2 μN was measured for 40 s prior to the indentation to correct the thermal drift indentation depth based on the mean thermal drift rate over the final 20 s. The acquired indentation data for each sample in the contact depth range of 20–200 nm were used by considering the effective range of the tip area function. To determine the effective range of the nanoindentation depths for each sample, the elastic moduli of all the samples were measured as a function of depth using the partial unloading technique with a load function containing a total of 20 partial unloading cycles, each comprising a 1-s loading segment, a 1-s holding segment, and a 1-s unloading segment to a maximum loading force of 1000 μN.

Statistical Analysis

The Shapiro-Wilk test indicated a normal distribution of the acquired data ($p > 0.05$). T-tests at a significance level of 0.05 were used to investigate the effect of storage time on μTBS and nanomechanical properties (elastic modulus and hardness) of the adhesive layer and the resin composite. The analyses were performed in SPSS Statistics 22 (IBM; Armonk, NY, USA).

RESULTS

Table 2 presents the results of the μTBS test and the failure mode analysis. Statistical analysis revealed that there was no significant difference ($p = 0.123$) in the μTBS between the 24-h group (59.1 ± 10.0 MPa) and the 8-year group (53.6 ± 11.7 MPa). After both storage periods, the predominant failure mode was at the interface between the adhesive and the resin-composite. However, the typical appearance of the debonded interfaces after 24 h was markedly different from the 8-year storage group. In the 24-h group, flat surfaces covered with the adhesive and the resin composite were observed on dentin and composite sides, respectively (Fig 1). In contrast, the 8-year specimens exhibited numerous extrusions on the dentin side and corresponding lacunae on the composite side (Fig 2). At magnification 200X and 5000X, filler particles covered with resin matrix could be identified on the surface of the extrusions and lacunae (Fig 2). Besides interfacial failures, cohesive failures in dentin were commonly observed after 24 h. However, their proportion decreased markedly after 8 years, and more cohesive failures in the adhesive and the resin composite were observed instead. No pre-test failures occurred in this study.

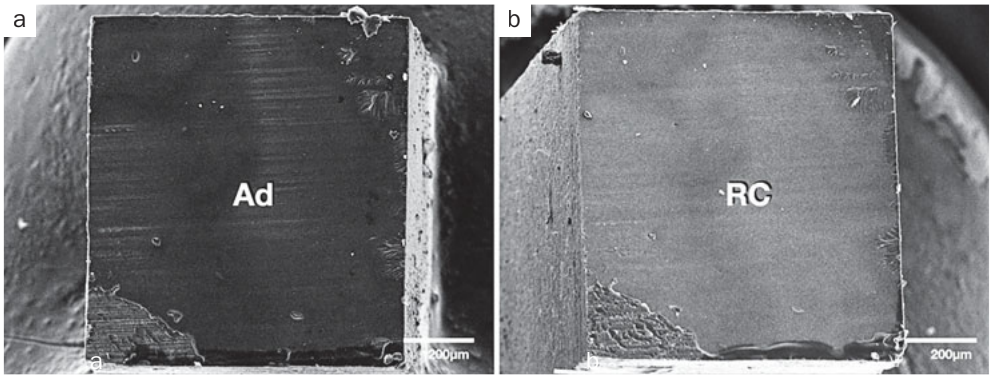

Fig 1 Representative SEM images of interfacial failure between the adhesive and the resin composite after 24 h of water storage. Dentin side (a) and composite side (b), magnification 70X. Ad: adhesive layer; RC: resin composite (RC).

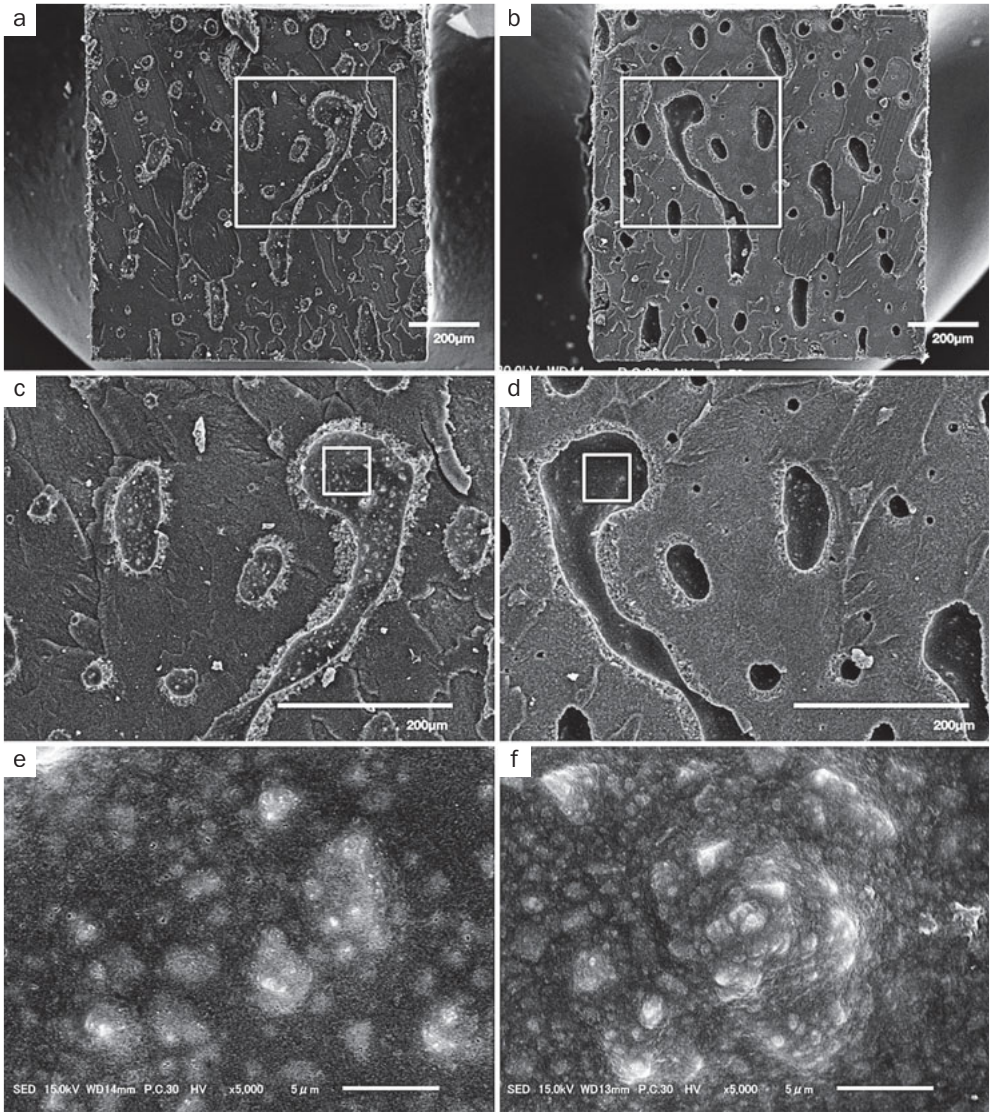

Fig 2 Representative SEM images of interfacial failure between the adhesive and the resin composite after 8 years of water storage. Dentin side (a) and composite side (b), magnification 70X. Matching extrusions on the dentin side and lacunae on the composite side were observed. Images (c) and (d) show an extrusion from image (a) and a lacuna from image (b) at higher magnification (200X). The area within white squares in (c) and (d) at 5000X magnification is presented in (e) and (f), respectively. Filler particles covered with resin matrix can be observed.

Tables 3 and 4 summarize the elastic moduli and hardness of the adhesive layer and the resin composite. Statistical analysis revealed that the 8-year water storage significantly decreased the elastic moduli and the hardness of both the resin composite and the adhesive resin ($p < 0.001$).

Table 3 Elastic modulus of the adhesive layer and the resin composite (mean ± SD)

Elastic modulus [GPa]	24 h	8 years
Adhesive layer	14.7 ± 1.8^{A}	7.0 ± 1.2^{B}
Resin composite	21.9 ± 0.9^{A}	10.2 ± 2.3^{B}
Different superscript letters in rows indicate statistically significant differences between the storage periods in each group ($p < 0.001$).		

Table 4 Hardness of the adhesive layer and the resin composite (mean ± SD)

Hardness (GPa)	24 h	8 years
Adhesive layer	1.27 ± 0.12^{A}	0.31 ± 0.02^{B}
Resin composite	1.33 ± 0.13^{A}	0.75 ± 0.19^{B}
Different superscript letters in rows indicate statistically significant differences between the storage periods in each group ($p < 0.001$).		

DISCUSSION

The ability to achieve a reliable, durable bond between dental resins and tooth structures is an important determinant for the lasting clinical success of adhesive restorations. Several artificial aging procedures including long-term water storage, thermocycling, or thermomechanical cycling are conducted in in vitro studies to simulate stresses to which dental materials are exposed in the oral environment.[19] A recent study examined the long-term μTBS of resin composite restorations performed in vivo and in vitro, and concluded that thermomechanical cycling and long-term water storage were useful to predict the durability of resin-dentin bonds.[22] It was also reported that the 5-year retention rate of Class V restorations was correlated with the aged but not the immediate bond strength.[36] This suggests that the results of immediate testing do not always correspond to the clinical performance of the adhesives and thus that more laboratory effort should be dedicated to aging procedures, rather than simply evaluating initial bond strengths.

In this study, bulk specimens bonded using a 1-SEA were stored in water for 8 years to evaluate the long-term stability of μTBS to dentin and mechanical properties of the adhesive layer. Sectioned specimens were aged in most previous studies to accelerate the degradation, but the storage of bulk specimens with enamel margins was preferred in this study, because it simulates the clinical situation more authentically. Generally, the surrounding enamel is believed to protect the more vulnerable resin-dentin bond from degradation.[2,37] However, our previous study reported significantly reduced μTBS of Clearfil S3 Bond (CS3; a predecessor of the CS3+ used in the present study) to dentin after 1-year water storage despite the presence of bonded enamel margins.[34] This could be attributed to the inferior etching ability of 1-SEAs and thus a weaker resin-enamel bond compared to multistep adhesives.[8,37] A significant decrease in the μTBS of CS3 was also observed in another study after 5 years of simulated pulpal pressure.[5] In contrast, the 8-year μTBS of CS3+ was not significantly different from the 24-h values in this study. Given the presence of a new photo-initiator in CS3+, this could be due to the higher degree of conversion (DC) of the adhesive layer created by CS3+ compared to CS3.[23] The higher DC of adhesive resins decreases their susceptibility to water sorption and hydrolytic degradation, while improving their initial bond strengths and contributing to the durability of the resin-dentin bond.[25,32] On the other hand, the slight decrease in μTBS after 8-year water storage accompanied by a marked decrease in the proportion of cohesive failures in dentin suggest bond degradation. It is possible that the statistical analysis of μTBS did not reveal significance because of the relatively small sample size and thus insufficient statistical power, which can be seen as a limitation of this study.

Failure mode analysis showed that the predominant failure mode was interfacial failure between the adhesive and the resin composite in both storage groups. A previous study reported that the oxygen-inhibition layer of CS3+ could not reach maximum DC even after the overlying resin composite Clearfil AP-X was light cured, which could result in its decreased mechanical properties and contribute to failures at this interface.[22] However, while flat debonded surfaces were observed after 24 h (Fig 1), extrusions of resin composite and corresponding lacunae were observed after 8-year water storage (Fig 2). This finding is in accordance with the reported degradation of the adhesive's oxygen-inhibition zone at the adhesive-resin composite joint after water storage for 100 or more days.[9] The composite structures observed with SEM on the fractured surfaces indicated that the crack propagated not only at the interface, but also through the matrix of the resin composite. In a

previous study by Feitosa et al,[5] transmission electron microscopy (TEM) also revealed polymer degradation in the adhesive layer of CS3 after 5 years of simulated pulpal pressure. In addition, the loss of silica filler particles from the adhesive layer was observed at high magnification,[5] indicating degradation at the filler-matrix interface.

The resin degradation observed with SEM at the failed surfaces was confirmed by the significant decrease in elastic moduli and hardness of the adhesive layer and the resin composite after 8 years of water storage. This finding is noteworthy, because to the best of our knowledge, nanomechanical properties of 1-SEAs and resin composites at the interface after such long water storage have not been investigated. The decrease in nanomechanical properties agrees with the fact that cohesive failure in the adhesive layer and in resin composite were observed only in the 8-year specimens. The present results corroborate previous studies which reported that the absorption of water decreased the mechanical properties of polymerized 1-SEAs.[13,14] Given that mechanical properties of HEMA-containing 1-SEAs were shown to be affected by water storage more than a HEMA-free 1-SEA,[30] we assume that long-term water infiltration and subsequent hydrolytic degradation of the resin matrix contributed to the deterioration of mechanical properties of the HEMA-containing CS3+ used here.

The first null hypothesis that 8-year water storage would not affect the μTBS of the HEMA-containing CS3+ to dentin had to be accepted, because no significant difference was found between the 24-h and 8-year values. The fact that only one 1-SEA was tested and that the measurements were performed only at two time points do not permit generalization of the obtained results, but they suggest that the long-term durability of the 1-SEA improved and that its use is clinically appropriate. On the other hand, 8-year water storage significantly decreased the mechanical properties of the adhesive layer, so the second null hypothesis was rejected. Moreover, signs of resin degradation were observed during the fractographic analysis. These observations were attributed to hydrolysis, and they reaffirmed the need to reduce the hydrophilicity of 1-SEAs after polymerization to improve their long-term durability. In the course of this 8-year study, this was achieved by extended air drying, especially if using warm air,[28,39] or the coating of 1-SEAs with a hydrophobic bonding agent.[33] Recently, it was also reported that the substitution of HEMA with methacrylamide monomers leads to lower water absorption, higher μTBS[16] and higher DC[32] of 1-SEAs. Nevertheless, further research is necessary to enhance the properties of 1-SEAs and to evaluate their aging behavior under various conditions.

CONCLUSION

Although 8-year water storage did not decrease the μTBS of a HEMA-containing 1-SEA, the observed failure mode patterns and significantly decreased nanomechanical properties indicated resin degradation of the adhesive and the resin composite at the adhesive-composite interface.

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REFERENCES

1. Araoka D, Hosaka K, Nakajima M, Foxton R, Thanatvarakorn O, Prasansuttiporn T, Chiba A, Sato K, Takahashi M, Otsuki M, Tagami J. The strategies used for curing universal adhesives affect the micro-bond strength of resin cement used to lute indirect resin composites to human dentin. Dent Mater J 2018;37:506–514.
2. Carvalho RM, Manso AP, Geraldeli S, Tay FR, Pashley DH. Durability of bonds and clinical success of adhesive restorations. Dent Mater 2012; 28:72–86.
3. Dos Santos PH, Karol S, Bedran-Russo AK. Long-term nano-mechanical properties of biomodified dentin-resin interface components. J Biomech 2011;44:1691–1694.
4. Federation FDIWD. FDI policy statement on Minimal Intervention Dentistry (MID) for managing dental caries: Adopted by the General Assembly: September 2016, Poznan, Poland. Int Dent J 2017;67:6–7.
5. Feitosa VP, Sauro S, Zenobi W, Silva JC, Abuna G, Van Meerbeek B, Sinhoreti MAC, Correr AB, Yoshihara K. Degradation of Adhesive-Dentin Interfaces Created Using Different Bonding Strategies after Five-year Simulated Pulpal Pressure. J Adhes Dent 2019;21:199–207.
6. Foxton RM, Melo L, Stone DG, Pilecki P, Sherriff M, Watson TF. Long-term durability of one-step adhesive-composite systems to enamel and dentin. Oper Dent 2008;33:651–657.
7. Freitas PH, Giannini M, Franca R, Correr AB, Correr-Sobrinho L, Consani S. Correlation between bond strength and nanomechanical properties of adhesive interface. Clin Oral Investig 2017;21:1055–1062.
8. Giannini M, Makishi P, Ayres AP, Vermelho PM, Fronza BM, Nikaido T, Tagami J. Self-etch adhesive systems: a literature review. Braz Dent J 2015;26:3–10.
9. Hashimoto M, Fujita S, Endo K, Ohno H. In vitro degradation of resin-dentin bonds with one-bottle self-etching adhesives. Eur J Oral Sci 2009; 117:611–617.
10. Hashimoto M, Fujita S, Kaga M, Yawaka Y. In vitro durability of one-bottle resin adhesives bonded to dentin. Dent Mater J 2007;26:677–686.
11. Heintze SD, Rousson V. Clinical effectiveness of direct class II restorations – a meta-analysis. J Adhes Dent 2012;14:407–431.
12. Heintze SD, Rousson V, Hickel R. Clinical effectiveness of direct anterior restorations – a meta-analysis. Dent Mater 2015;31:481–495.
13. Hosaka K, Nakajima M, Takahashi M, Itoh S, Ikeda M, Tagami J, Pashley DH. Relationship between mechanical properties of one-step self-etch adhesives and water sorption. Dent Mater 2010;26:360–367.
14. Hosaka K, Tagami J, Nishitani Y, Yoshiyama M, Carrilho M, Tay FR, Agee KA, Pashley DH. Effect of wet vs. dry testing on the mechanical properties of hydrophilic self-etching primer polymers. Eur J Oral Sci 2007;115: 239–245.
15. Itoh S, Nakajima M, Hosaka K, Okuma M, Takahashi M, Shinoda Y, Seki N, Ikeda M, Kishikawa R, Foxton RM, Tagami J. Dentin bond durability and water sorption/solubility of one-step self-etch adhesives. Dent Mater J 2010;29:623–630.
16. Kuno Y, Hosaka K, Nakajima M, Ikeda M, Klein Junior CA, Foxton RM, Tagami J. Incorporation of a hydrophilic amide monomer into a one-step self-etch adhesive to increase dentin bond strength: Effect of application time. Dent Mater J 2019;38:892–899.
17. Liu Y, Tjäderhane L, Breschi L, Mazzoni A, Li N, Mao J, Pashley DH, Tay FR. Limitations in bonding to dentin and experimental strategies to prevent bond degradation. J Dent Res 2011;90:953–968.
18. Loomans BAC, Kreulen CM, Huijs-Visser HECE, Sterenborg BAMM, Bronkhorst EM, Huysmans MCDNJM, Opdam NJM. Clinical performance of full rehabilitations with direct composite in severe tooth wear patients: 3.5 years results. J Dent 2018;70:97–103.
19. Mair L, Padipatvuthikul P. Variables related to materials and preparing for bond strength testing irrespective of the test protocol. Dent Mater 2010;26:e17–23.
20. Marchesi G, Frassetto A, Mazzoni A, Apolonio F, Diolosa M, Cadenaro M, Di Lenarda R, Pashley DH, Tay FR, Breschi L. Adhesive performance of a multi-mode adhesive system: 1-year in vitro study. J Dent 2014;42:603–612.
21. Peumans M, De Munck J, Mine A, Van Meerbeek B. Clinical effectiveness of contemporary adhesives for the restoration of non-carious cervical lesions. A systematic review. Dent Mater 2014;30:1089–1103.

22. Rego HM, Alves TS, Bresciani E, Niu LN, Tay FR, Pucci CR. Can long-term dentine bonding created in real life be forecasted by parameters established in the laboratory? Sci Rep 2016;6:37799.
23. Sakano W, Nakajima M, Prasansuttiporn T, Foxton RM, Tagami J. Polymerization behavior within adhesive layer of one- and two-step self-etch adhesives: a micro-Raman spectroscopic study. Dent Mater J 2013;32: 992–998.
24. Sampaio PCP, Kruly PC, Ribeiro CC, Hilgert LA, Pereira PNR, Scaffa PMC, Di Hipolito V, D'Alpino PHP, Garcia FCP. Comparative bonding ability to dentin of a universal adhesive system and monomer conversion as functions of extended light curing times and storage. J Mech Behav Biomed Mater 2017;75:41–49.
25. Sato K, Hosaka K, Takahashi M, Ikeda M, Tian F, Komada W, Nakajima M, Foxton R, Nishitani Y, Pashley DH, Tagami J. Dentin bonding durability of two-step self-etch adhesives with improved of degree of conversion of adhesive resins. J Adhes Dent 2017;19:31–37.
26. Sato M, Fujishima A, Shibata Y, Miyazaki T, Inoue M. Nanoindentation tests to assess polymerization of resin-based luting cement. Dent Mater 2014;30:1021–1028.
27. Shinoda Y, Nakajima M, Hosaka K, Otsuki M, Foxton RM, Tagami J. Effect of smear layer characteristics on dentin bonding durability of HEMA-free and HEMA-containing one-step self-etch adhesives. Dent Mater J 2011; 30:501–510.
28. Taguchi K, Hosaka K, Ikeda M, Kishikawa R, Foxton R, Nakajima M, Tagami J. The effect of warm air-blowing on the microtensile bond strength of one-step self-etch adhesives to root canal dentin. J Prosthodont Res 2018;62:330–336.
29. Tajima K, Nikaido T, Inoue G, Ikeda M, Tagami J. Effects of coating root dentin surfaces with adhesive materials. Dent Mater J 2009;28:578–586.
30. Takahashi M, Nakajima M, Hosaka K, Ikeda M, Foxton RM, Tagami J. Long-term evaluation of water sorption and ultimate tensile strength of HEMA-containing/-free one-step self-etch adhesives. J Dent 2011;39: 506–512.
31. Tay FR, Pashley DH. Have dentin adhesives become too hydrophilic? J Can Dent Assoc 2003;69:726–731.
32. Tichy A, Hosaka K, Abdou A, Nakajima M, Tagami J. Degree of conversion contributes to dentin bonding durability of contemporary universal adhesives. Oper Dent 2020;45:556–566
33. Tichy A, Hosaka K, Bradna P, Ikeda M, Abdou A, Nakajima M, Tagami J. Subsequent application of bonding agents to a one-step self-etch adhesive – Its effect with/without previous light-curing. Dent Mater 2019;35: e299–e309.
34. Torkabadi S, Nakajima M, Ikeda M, Foxton RM, Tagami J. Bonding durability of HEMA-free and HEMA-containing one-step adhesives to dentine surrounded by bonded enamel. J Dent 2008;36:80–86.
35. Tyas MJ, Anusavice KJ, Frencken JE, Mount GJ. Minimal intervention dentistry – a review. FDI Commission Project 1-97. Int Dent J 2000;50:1–12.
36. Van Meerbeek B, Peumans M, Poitevin A, Mine A, Van Ende A, Neves A, De Munck J. Relationship between bond-strength tests and clinical outcomes. Dent Mater 2010;26:e100–121.
37. Van Meerbeek B, Yoshihara K, Yoshida Y, Mine A, De Munck J, Van Landuyt KL. State of the art of self-etch adhesives. Dent Mater 2011;27:17–28.
38. Walter R, Swift EJ, Jr., Nagaoka H, Chung Y, Bartholomew W, Braswell KM, Pereira PN. Two-year bond strengths of "all-in-one" adhesives to dentine. J Dent 2012;40:549–555.
39. Yonekura K, Hosaka K, Tichy A, Taguchi K, Ikeda M, Thanatvarakorn O, Prasansuttiporn T, Nakajima M, Tagami J. Air-blowing strategies for improving the microtnsile bond strength of one-step self-etch adhesives to root canal dentin. Dent Mater J 2020;39:892–899.
40. Zimmerli B, De Munck J, Lussi A, Lambrechts P, Van Meerbeek B. Long-term bonding to eroded dentin requires superficial bur preparation. Clin Oral Investig 2012;16:1451–1461.

Clinical relevance: Eight years of water storage significantly decreased nanomechanical properties of a one-step self-etch adhesive but not its bond strength to dentin. Resin degradation of the adhesive and the resin composite led to failures at the adhesive-composite interface.