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Effect of Evaporation on the Shelf Life of a Universal Adhesive

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Clinical Relevance

Evaporation of adhesive ingredients due to repeated opening of the bottle adversely influences the shelf life of a universal one-step adhesive only when more than 50% of the solvent and evaporable ingredients have been evaporated.

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SUMMARY

Objectives: The purpose of this study was to evaluate how evaporation affects the shelf life of a one-bottle universal adhesive.

Methods: Three different versions of Scotchbond Universal (SBU, 3M ESPE, Seefeld, Germany) were prepared using a weight-loss technique. SBU0 was left open to the air until maximal weight loss was obtained, whereas SBU50 was left open until 50% of evaporation occurred. In contrast, SBU100 was kept closed and was assumed to contain the maximum concentration of all ingredients. The degree of conversion (DC) was determined by using Fourier transform infrared spectroscopy on different substrates (on dentin or glass plate and mixed with dentin powder); ultimate microtensile strength and microtensile bond strength to dentin were measured as well.

Results: DC of the 100% solvent-containing adhesive (SBU100) was higher than that of the 50% (SBU50) and 0% (SBU0) solvent-containing adhesives for all substrates. DC of the adhesive applied onto glass and dehydrated dentin was higher than that applied onto dentin. Even though the ultimate microtensile strength of SBU0 was much higher than that of SBU50 and SBU100, its bond strength to dentin was significantly lower.

Conclusions: **Evaporation of adhesive ingredients may jeopardize the shelf life of a one-bottle universal system by reducing the degree of conversion and impairing bond strength. However, negative effects only became evident after more than 50% evaporation.**

INTRODUCTION

In dental clinical practice, the shelf life of adhesives is important. Shelf life can be defined as the period during which adhesive systems retain optimum bonding efficacy. Because the composition of adhesives gradually changes over time, manufacturers always provide an expiration date.[1] The adhesive should not be used after this date, as the efficacy can no longer be guaranteed. More specifically, the adhesive composition may change over time due to hydrolysis or polymerization of the monomers, degradation of the additives (initiators/stabilizers), or evaporation of ingredients.[2]

Thanks to their easy application procedure, one-step self-etch adhesives are commonly used in dental practice today. A long shelf life is important for their bonding effectiveness, but because of their specific composition, this class of adhesives may be more prone to reduced shelf life than conventional multi-step systems. First, one-step self-etch adhesives typically have their adhesive monomers mixed with water, which may lead to hydrolysis of the ester bond of the methacrylic group.[3,4] Moreover, the acidic pH in one-step self-etch adhesives may accelerate this hydrolysis process.[3,5] Long storage times and high temperatures may also promote hydrolysis.[3,5] Previous research showed that hydrolysis of the monomers may indeed reduce the bonding efficacy of self-etch systems.[1]

Second, ingredients may evaporate by repeated opening of the bottle. Not only do organic solvents easily evaporate due to their high vapor pressure but small quantities of low-molecular-weight monomers do also.[6] Evaporation is also enhanced by high temperatures. It is clear that the amount of solvent in an adhesive may be different between the first and last use. Unlike composites, dental adhesives contain solvents, which serve different purposes, like dissolving the monomers, ionizing the functional monomers, and facilitating infiltration of the resin in dentin.[2] Perdigao and coworkers[7] studied the effects of repeated opening of a two-step etch-and-rinse adhesive on bond strength. They left the bottle open for one minute, two times a day for three weeks, and determined the bond strength. They found lower bond strengths after three weeks for the acetone-containing adhesive and concluded that solvent evaporation may reduce the shelf life of adhesives.

Apart from that study, only a few studies have evaluated how ingredient evaporation affects the bonding effectiveness of adhesives. The objective of this study was to evaluate how ingredient evaporation affects degree of conversion (DC), ultimate microtensile strength (UμTS), and bond strength of a commercial, universal, one-component, one-step self-etch adhesive. To better understand how ingredient evaporation affects the shelf life of this adhesive, polymerization efficacy was investigated on different substrates. The null hypothesis tested was that evaporation does not affect DC, UμTS, and microtensile bond strength.

METHODS AND MATERIALS

Adhesive Preparation

Three different versions of Scotchbond Universal (SBU, 3M ESPE, Seefeld, Germany) were prepared (Table 1). First, the content of a fresh bottle of SBU was divided into three amber vials, which were protected from light by aluminum foil. The first vial was capped immediately. This version was assumed to have 100% solvent and was called SBU100. The second vial was left open in a dark room at room temperature (20°C) until its weight was constant for at least two days (this took more than 14 days). This vial was assumed to have 0% solvent (SBU0). The last vial was left open until it had 50% of the weight loss as calculated from the second vial, which took more than five days at room temperature. This adhesive version was called SBU50. According to the manufacturer, SBU contains around 20 to 30 wt% solvent (ethanol and water), and the percentage of weight loss in SBU0 and SBU50 was 22.7 wt% and 11.3 wt%, respectively.

All adhesives were then kept capped in the refrigerator until use. The pH of SBU100, SBU50, and SBU0 was 2.93, 2.79, and 2.15, respectively (measured in triplicate, Inolab pH Level 2, WTW GmbH, Weilheim, Germany).

Degree of Conversion

Each adhesive was applied onto six different substrates and the DC of the adhesives was measured by Fourier transform infrared spectroscopy (FTIR). Four samples were prepared per group (n=4).

- Group 1 (glass): The adhesives were rubbed on a glass slide for 20 seconds and then air-blown for 10 seconds according to the manufacturer's instruc-

Table 1: *Composition of the Universal Adhesive Used in This Study*

Adhesive	Composition	Instructions for Application
Scotchbond Universal (SBU) Lot 468651	BisGMA 15-25 wt%, HEMA 15-25 wt%, DMDMA 5-15 wt%, ethanol 10-15 wt%, water 10-15 wt%, silane-treated silica 5-10 wt%, copolymer of acrylic and itaconic acid 1-5 wt%, methyl ethyl ketone <0.5 wt%, CQ ~2 wt%, EDMAB <2 wt%	Apply the adhesive and rub it in for 20 seconds, gently air-blow for 5 seconds until no more movement of the adhesive, then light-cure for 10 seconds.

Abbreviations: BisGMA, bisphenol A diglycidyl ether dimethacrylate; CQ, camphorquinone; DMDMA, decamethylene dimethacrylate; EDMAB, ethyl 4-(dimethylamino)benzoate; HEMA, 2-hydroxyethyl methacrylate.

tions. The adhesive was covered by another glass slide to avoid an oxygen-inhibition layer, and light-cured.

- Group 2 (glass 37°C): The adhesives were applied as in group 1 onto a glass slide, which was preheated on a hot table up to 37°C, and then covered by another preheated glass slide.
- Group 3 (dentin): Human third molars with no caries (collected after obtaining informed consent approved by the Commission for Medical Ethics of Katholieke Universiteit Leuven) were stored in 0.5% chloramine/water at 4°C and used within three months after extraction. Midcoronal dentin discs were cut from extracted human molars. The adhesives were applied as in group 1, covered by a glass slide, and light-cured.
- Group 4 (dentin 37°C): The adhesives were applied as in group 3 to a dentin disc preheated on a hot table up to 37°C, and then covered by another preheated glass slide.
- Group 5 (dehydrated dentin): The adhesives were applied as in group 3 to a dentin disc, which was dehydrated in an oven at 37°C for 24 hours, and then kept in vacuum for at least 1 hour.
- Group 6 (dentin powder): Dentin powder was prepared by grinding human dentin with a grinder (A10, IKA, Staufen, Germany) according to a previously described protocol.[8] After mixing the adhesive with 10 wt% dentin powder, it was further treated as in group 1.

The specimens were cured with a Bluephase 20i light-emitting diode unit (Ivoclar-Vivadent, Schaan, Liechtenstein) for 10 seconds at 1000 mW/cm^2 in high mode and stored at 37°C for 24 hours in a dry condition. DC was measured by attenuated total reflectance FTIR (Vertex 70, Bruker Optik GmbH, Ettlingen, Germany) in triplicate and calculated as the ratio of peak intensities of the aliphatic 1640 cm^{-1} and aromatic 1610 cm^{-1} peak in cured and uncured materials. FTIR spectra between 4500 and 400 cm^{-1} were recorded at room temperature at 32 scans per sample and a resolution of 4 cm^{-1}. DC was calculated according to the following formula:

$$DC = \left(1 - \frac{R\ \text{cured}}{R\ \text{uncured}}\right) \times 100$$

where R is the ratio of intensities of the peak at 1640 cm^{-1} and 1610 cm^{-1}.

Ultimate Microtensile Strength

Ten polymerized adhesive specimens were prepared (n=10) to determine the UμTS. Each adhesive was applied in a $2 \times 2 \times 8$ mm^3 silicone mold, which was covered with a glass slide. The specimens were cured for 20 seconds through the glass slide with the Bluephase 20i (Ivoclar-Vivadent) light-emitting diode unit with the output of 1000 mW/cm^2. After dry storage for 24 hours in the dark at 37°C, the adhesive sticks were trimmed at the middle of the stick to an hourglass shape with a diameter of 1.1 mm using a cylindrical, extra-fine grit (15 μm) diamond bur in a water-cooled high-speed handpiece mounted in a MicroSpecimen Former (University of Iowa, Iowa City, IA, USA).[9] For SBU0, additional samples with a diameter of 0.8 mm were trimmed. The diameter of each specimen was measured using a stereomicroscope with a resolution of 1 μm at a magnification of 20× (400-NRC, Leitz, Germany). A cross-section area of about 1 mm^2 (SBU100 and SBU50) and about 0.5 mm^2 (SBU0) was obtained. Each specimen was attached to a modified notched Ciucchi's jig with cyanoacrylate glue (Model Repair II Blue, Dentsply-Sankin, Tochigi, Japan) and stressed at a crosshead speed of 1 mm/min until failure in a universal testing device (LRX, Lloyd, Hampshire, UK) to determine the UμTS.

Microtensile Bond Strength (μTBS)

After removal of the occlusal third of the crown of human third molars with a diamond saw (Isomet 1000, Buehler, Lake Bluff, IL, USA), a standardized bur-cut smear layer was created by means of a high-speed medium-grit diamond bur (100 μm; 842, Komet, Lemgo, Germany) mounted in a MicroSpecimen former (University of Iowa).[10] Per adhesive

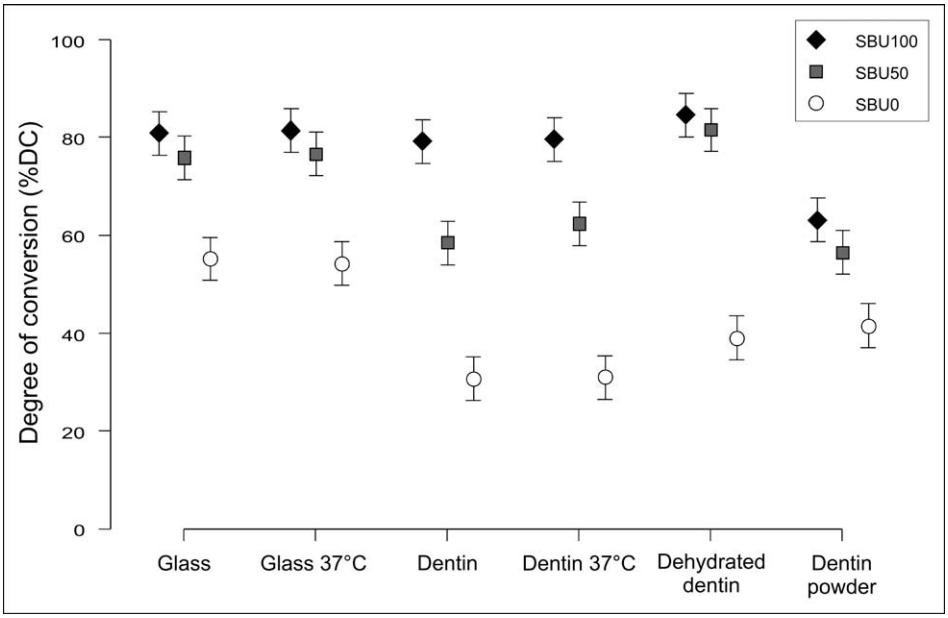

Figure 1. *Degree of conversion (DC in %) of the adhesive versions: 100% solvent-containing adhesive (SBU100), 50% solvent-containing adhesive (SBU50), and 0% solvent-containing adhesive (SBU0) (mean and standard deviation).*

group, eight teeth were used (n=8). As per the manufacturer's instructions, the adhesives were applied onto dentin, rubbed for 20 seconds, air-blown for 10 seconds, and light-cured for 10 seconds (output of 1000 mW/cm^2, Bluephase 20i, Ivoclar Vivadent). Composite build-ups were made with Z100 MP composite (shade A3; 3M ESPE) in three layers, the first being 1-mm thick and then two subsequent 2-mm thick layers. Each composite layer was cured for 20 seconds. After 24 hours of storage in distilled water at 37°C, the teeth were sectioned perpendicularly to the interface by means of an automated water-cooled precision diamond saw (Accutom-50, Struers, Ballerup, Denmark) to produce rectangular 1×1 mm sticks. Up to six central sticks were used for μTBS testing. Each stick was attached to a modified notched Ciucchi's jig[9] with cyanoacrylate glue and stressed at a crosshead speed of 1 mm/min until failure in the LRX (Lloyd) testing device to determine the μTBS. The mode of failure was determined by stereomicroscopy. Representative composite and dentin fragments were observed by scanning electron microscopy (SEM; JSM-5600, JEOL, Tokyo, Japan).

Statistical Analysis

To statistically assess DC, a linear mixed effects model taking into account multiple testing of the same specimens was constructed using statistical software (R 2.13.2 and nlme package, R Foundation for Statistical Computing, Vienna, Austria). In this model substrate, adhesives and their interaction were included. To further investigate these interactions, 95% confidence intervals were calculated (R 2.13.2 and AICcmodavg package). All tests were performed at a significance level of $\alpha=0.05$.

The UμTS and μTBS data were analyzed by one-way analysis of variance and Tukey's multiple comparison test ($\alpha=0.05$).

RESULTS

Degree of Conversion

Irrespective of the group, SBU100 always exhibited the highest DC, and SBU0 had the lowest DC (Figure 1). The linear mixed-effect model showed that both adhesive and substrate significantly affected DC ($p<0.0001$ for both). However, depending on the adhesive, a different effect was observed on the various substrates, as a significant interaction ($p<0.0001$) was observed.

SBU100 had the highest DC when applied on dehydrated dentin substrate (DC=84%) and the lowest DC when mixed with dentin powder (DC=63%). DC in this last group was significantly lower than that in the other groups. There were no statistically significant differences between the other groups: glass, glass 37°C, dentin, dentin 37°C, and dehydrated dentin.

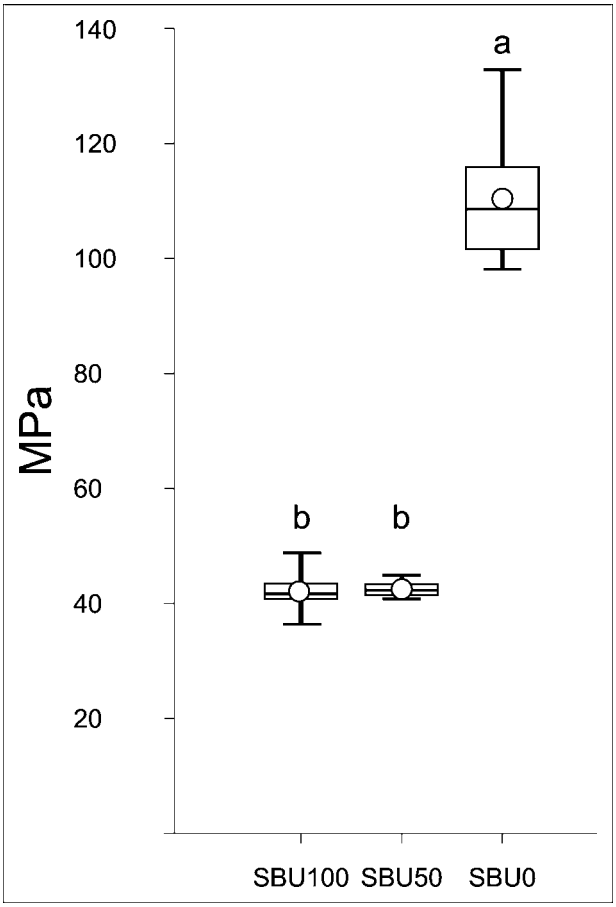

Figure 2. *Ultimate microtensile strength of the adhesive versions: 100% solvent-containing adhesive (SBU100), 50% solvent-containing adhesive (SBU50), and 0% solvent-containing adhesive (SBU0) (mean and standard deviation). The same letters indicate no statistically significant difference (*p*>0.05).*

For SBU50, the highest DC was also observed when applied on dehydrated dentin (DC=81%), and this DC was not significantly different from that in the group with glass and glass 37°C substrates (Figure 1). DCs on glass, glass 37°C, and dehydrated dentin were significantly higher than the DCs on dentin or dentin 37°C and when mixed with dentin powder. The lowest DC, which was 56%, was observed in the dentin powder group.

For SBU0, the highest DC was obtained when applied on a glass substrate (DC=55%), and this was not significantly different from that in the glass 37°C group. When applied on dehydrated dentin and mixed with dentin powder, DC was significantly lower than in the glass and glass 37°C groups, but still higher than the DC in the dentin and dentin 37°C group. The lowest DC observed was 30% when applied on dentin.

Ultimate Microtensile Strength

The UμTS of SBU0 adhesive samples with a 1-mm constriction could not be determined as the cyanoacrylate glue always failed first. Therefore, the UμTS of SBU0 was determined in additional samples with 0.8-mm constriction. With a UμTS of 110.4 (±10.4) MPa, the UμTS of SBU0 was more than twice as high as that of SBU50 and SBU100 (UμTS of 42.5 ± 1.4 and 42.1 ± 3.3, respectively) (Figure 2). The UμTS of SBU0 was significantly higher than that of SBU100 and SBU50.

Microtensile Bond Strength

The μTBS of the different experimental adhesives to dentin are shown in Figure 3. No pretesting failures were recorded. The μTBS of SBU100 and SBU50 was not statistically significantly different. However, SBU0 obtained a significantly lower bond strength than SBU50 and SBU100.

DISCUSSION

So-called universal adhesives, which have been claimed to be applicable onto different substrates (tooth, alloys, ceramics, composites) have recently regained attention, as the latest generation of universal adhesives consists of only a single bottle. With their simple and short application procedure, they represent an attractive substitute for previous universal systems that were often very laborious and required different application steps and primers depending on the bonding substrate. Because this new class of adhesives meets the demands of dentists, who no longer need to buy different adhesives, it can be foreseen that they will probably become very popular. Considering the still increasing share of adhesive applications used in general dental practice, the way evaporation affects shelf life may be very important for universal adhesives, as this type of adhesives will be opened very frequently.

In this study, we left the bottle open until there was no longer weight loss due to evaporation (SBU0). In total, there was 23% weight loss compared with the original adhesive. Assuming that the main cause for the weight loss was the evaporation of the solvent, this corresponded well to the amount of solvent (20-30 wt%) in the adhesive according to the manufacturer.

First, we tested how evaporation affects DC. In general, evaporation led to decreased DC, which corresponds to previous research, in which it was reported that small amounts of solvent increase the polymerization degree.[11,12] The negative effect of

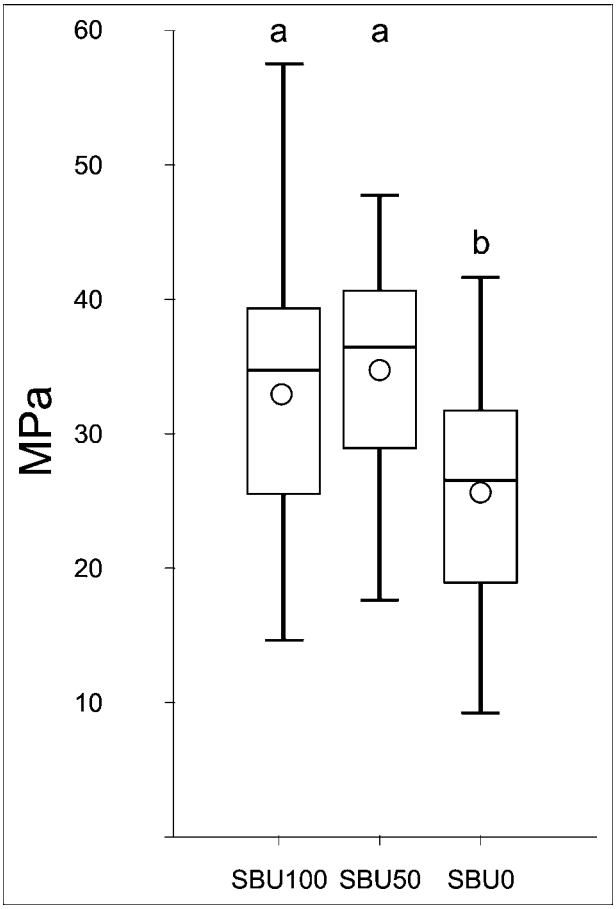

Figure 3. *Microtensile bond strength of the adhesive versions: 100% solvent-containing adhesive (SBU100), 50% solvent-containing adhesive (SBU50), and 0% solvent-containing adhesive (SBU0) (mean and standard deviation). The same letters indicate no statistically significant difference (*$p > 0.05$*).*

reduced solvent concentrations may be attributed to increased viscosity, with hampered diffusion of monomers, growing polymer-chain segments, and radicals.[13] Alternatively, this phenomenon could be explained by a decreased glass-transition temperature when a solvent is mixed with the adhesive monomers.[14] At a constant temperature, mixtures with a lower glass-transition temperature will obtain higher degrees of polymerization.[14] To verify this hypothesis, we added 10 wt% of ethanol (absolute ethanol, 99.99%; CAS: 64-17-5; VWR, Haasrode, Belgium) to SBU0 and applied it on a glass plate as in group 1.This resulted in a dramatic increase of the DC of SBU0 from 55.1% (±1.2) to 74.8% (±1.8), which confirmed that solvent evaporation jeopardized polymerization and that the lower DC was not due to monomer degradation, premature cure, or any other side effect of the evaporation.

However, the effect of evaporation was highly dependent on the type of substrate, which demonstrates that the conversion rate depends on a complex interplay of factors. When applied on a glass plate, 50% evaporation did not significantly reduce DC, whereas on dentin, DC was significantly decreased. This should be attributed to the inhibitory effect of remaining water in dentin[15] as SBU50 did not exhibit reduced DC on dehydrated dentin. The inhibitory effect of water on the DC can be explained by the fact that in SBU50, ethanol had most probably been evaporated due to its high vapor pressure and that any remaining solvent would have been water. As result, the solvent in SBU50 could not be removed anymore, and further dilution of the monomers due to additional water uptake from dentin led to a reduced DC.

Nevertheless, when all solvent was evaporated, SBU0 performed equally on hydrated and dehydrated dentin. We hypothesized that the drop in DC on dehydrated dentin compared with SBU50 and compared with SBU0 on glass was due to the fact that the dehydrated dentin absorbed all remaining water upon application of the solvent, thereby increasing the viscosity to the point that polymerization was hindered. A balanced amount of solvent in the adhesive thus seems imperative to obtain high conversion degrees.[13,16]

To evaluate the effect of the constituents (collagen, hydroxyapatite)[17] of dentin on the polymerization rate, we mixed dentin powder with the adhesive.[18,19] However, the DC of SBU100 mixed with dentin was significantly lower than on glass or dentin, probably due to the fact that the dentin powder acted as filler particles,[20] which may prevent activated polymer strands from meeting. As reported in literature, temperature is an important factor that influences the degree of polymerization.[21] Preheating the substrate did not seem to influence DC in this study. Whereas a significant positive effect of higher temperatures on DC was previously reported in studies[21,22] in which DC was measured shortly after curing, DC of the samples in our study was only determined 24 hours after light-curing.

We also tested how evaporation affects the intrinsic mechanical strength of the adhesive resin. Unlike SBU100 and SBU50, the UμTBS of SBU0 could not be tested, as the intrinsic mechanical strength of these adhesive samples surpassed that of the cyanoacrylate glue to fix samples to the μTBS jig. Therefore, we also tested adhesive samples with a 0.8 mm constriction, even though it was previously reported that the bonding surface may influence the

bond strength.[23] To our surprise, the UμTS of SBU0 was more than twice as high as that of SBU50 and SBU100, in spite of its inferior DC. A plausible explanation may be the higher monomer/volume ratio in SBU0 compared with the other versions. In addition, it is also conceivable that the solvent in SBU50 and SBU100, which was difficult to remove with air-blowing once the adhesive was applied in the mold, significantly deteriorated the mechanical properties of the adhesive resin. It was previously reported that remaining solvent softened the polymer.[11,15,24] The remarkably higher mechanical strength may also be due to a higher cross-linked polymer structure in SBU0, as small low-molecular weight monomers, like 2-hydroxyethyl methacrylate (HEMA), may have evaporated. HEMA is a monomethacrylate, which will lead to a less cross-linked polymer[2] and thus to polymers with inferior mechanical strength.

Nevertheless, in spite of the superior UμTS of SBU0, the bond strength to dentin was significantly worse than that of SBU100 and SBU50. This result once again shows that adhesion to tooth tissue is not related to mechanical strength alone[25,26] but also to the interaction with the tooth substrate and the DC. Complete evaporation of water from an adhesive with a self-etch strategy would be problematic as the functional monomers need to be ionized before they can interact with hydroxyapatite in dentin.[27] In addition, the increased viscosity after evaporation may adversely influence the interaction with dentin.[28] Last, the inferior bonding performance of SBU0 may also be partially associated with the low DC on dentin.

To conclude, evaporation of adhesive ingredients did affect the DC, UμTS, and microtensile bond strength of a universal adhesive, and the null hypothesis must thus be rejected. However, repeated opening of the bottle will only jeopardize the clinical performance of a universal one-step adhesive when more than 50% of the solvent and evaporable compounds have been evaporated. In our study, it took more than five days before 50% was evaporated (at room temperature), in spite of using an open bottle. It is thus unlikely that shelf life in a clinic will be impaired by evaporation, especially when the adhesive is used according to the manufacturer's instructions and when the adhesive bottle is recapped after every use.

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Conflict of Interest

The authors of this manuscript certify that they have no proprietary, financial, or other personal interest of any nature or kind in any product, service, and/or company that is presented in this article.

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