Series on Biomechanics, Vol.37, No.2 (2023), 43- 48
DOI: 10.7546/SB.05.02.2023

Long-term microtensile bond strength of universal adhesives with human dentin: a mini review

E. Koytchev[a*]

[a] *Dept. of Biomechanics, Institute of Mechanics, Bulgarian Academy of Sciences; koytchev@imbm.bas.bg*

Received: 15 May 2023 *Accepted: 27 June 2023* **Corresponding Author*

Abstract
Objectives: This mini review aimed at evaluating the long-term bonding effectiveness of universal dental adhesives (UA) with human dentin.
Methods: The web databases of PubMed and Web of Science from were scanned using the keywords: (universal adhesives, long-term microtensile bond strength - μTBS, human dentin). After following the inclusion criteria, a total of 16 articles were retained for the purposes of this review.
Results: Overall, the different articles investigated various factors and their influence on the μTBS. The main variables included: dentin moisture levels, etching times and modes, inclusion of antibacterials and dentin crosslinkers, air-blowing and bond application times. The majority of studies were consistent in their findings that a long-term stable bond is achievable by using the adhesives in self-etching protocol. Dentin cross-linkers were beneficial to improve the longevity of UA systems. The effects of antibacterials and water storage times, as well as air blowing were material dependent.
Conclusions: Generally, although with well-studied short-time μTBS performance, the UAs need further clinical and *in vivo* validation studies to evaluate bond strength over time.

Keywords: Universal adhesives, long-term microtensile bond strength, human dentin

1. Introduction

The evolvement of dental adhesive technology is often associated with continuous improvement of the adhesive systems, and a constant pursuit of the manufacturers to reduce the clinical steps and hence – the technique sensitivity. The adhesive systems which are now used in modern dentistry are currently in their 8th generation [1]. These constitute the so-called universal adhesives (UA) and can be used in either etch and rinse or self-etch regimes, depending on the personal preference of the clinicians. The UAs can also be combined with selective enamel etch strategy [2]. This multi-approach opportunity allows the dental professionals to be flexible in choosing the right method according to the clinical situation [3]. Although the UAs can be seen as one-step self-etch adhesives, they should not be confused with the previous 7th generation dental adhesive systems, which were prone to nanoleakage after aging and had limited durability [4]. The UAs have very complex chemistry since they are designed to work well with both direct and indirect teeth restorations. They must contain specific functional monomers, which are capable to co-polymerize with composite resin materials and at the same time interact with dentin. Additionally, the UAs should have acidic character to be active in self-etch mode and simultaneously be not so acidic to interact with the initiators for the polymerization of self- and dual-cure cements [5]. Despite some fine nuances that vary from manufacturer to manufacturer, such as pH, solvent chemistry and others, all of the UAs use adhesive functional monomers. One of them is the proven throughout the years 10-MDP (methacryloyloxy-decyl-dihydrogen-phosphate). This phosphate ester was developed and synthesized by Kuraray Noritake Company (Osaka, Japan) more than 40 years ago. The 10-MDP monomer today is part of the composition of many UAs. It is the most hydrophobic among all functional monomers that are commonly used in dental adhesives and can create chemical bonds to methacrylate composite resins and cements [6]. The UAs are supposed to exhibit both, hydrophilic and hydrophobic properties at the same time, since the hard tooth tissues substrate is inherently wet and the restorative materials need the hydrophobic ends of the functional monomers. This subtle balance between hydrophobic and hydrophilic monomers is achieved by the various manufacturers by mixing some of the commonly used monomers in adhesive dentistry like bis-GMA (bisphenol A-glycidyl methacrylate), which is hydrophobic and HEMA (hydroxyethylmethacrylate), which has hydrophilic properties. The final goal after setting of the universal adhesives should be the creation of a matrix that is well-bonded to the tooth tissues and composite restorations at the same time [7]. The pH of contemporary UAs (table 1) can be between 2.2 and 3.2 and accordingly they are subdivided to mild pH>2), extra-mild (pH>2.5) and intermediately strong (pH>1.2). The acids used in the self-etching bonding systems have the purpose of creating a demineralized zone with exposed collagen fibrils, which are infiltrated with resins that will be further polymerized. This will create the thin layer of resin-infiltrated dentin – the so-called hybrid layer (Fig.1). Nakabayashi et al. first described this layer in 1982 [8]. Human dentin is a complex, anisotropic biocomposite with distinct hierarchical structure. Its composition of 50% vol % mineral phase, 30 vol % collagen and 20 vol % water [9]. Bonding to dentin is challenging task since the mineral and organic phases, as well as the moist conditions should be considered.

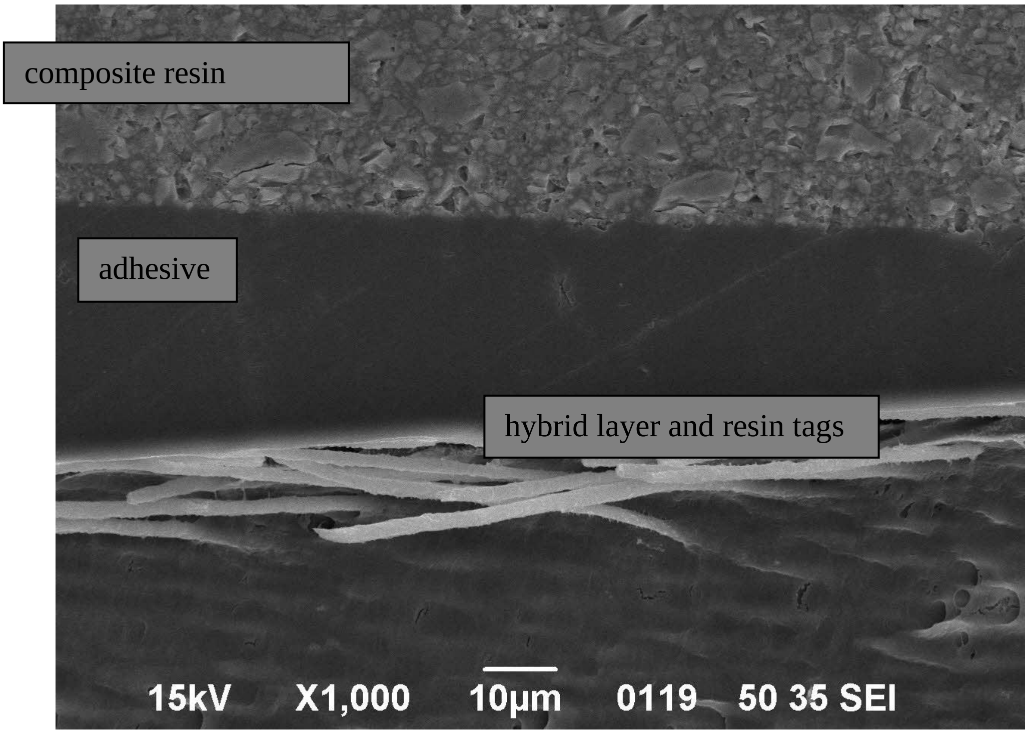

Fig1. SEM picture of interaction of UA (Clearfil Universal Bond Quick) with dentin and restorative composite resin material. The hybrid layer and associated resin tags form the foundation of the adhesive interface and represents the first in a series of links that together form a bonded assembly between the tooth tissues and resin-based restoratives and cements (SEM image – courtesy of the author).

The microtensile bond strength (μTBS) test was first introduced by Sano et al. in 1994 [10]. Nowadays, the method is modified, so that it employs beam-shaped specimens with 1x1 mm cross-sectional area. The specimens for the μTBS test have three main components: dentin, adhesive and some restorative material [11].

More than thousand publications are available when the Web is searched for μTBS (Google scholar and PubMed). Overall, the μTBS method is well-established for testing the adhesion of various restorative materials to dentin (Fig 2.).

The main ingredients of UAs and classification, according to pH, proposed by van Meerbeck et al. are presented in Table 1 [1].

Table 1
Main compositions and pH classification of universal adhesives

Classification	pH	Name	Manufacturer	Main components
Ultra-mild	3.2	All-Bond Universal	Bisco; Schaumburg,IL, USA	Bisphenol A diglycidylmethacrylate, ethanol, MDP,2-hydroxyethyl methacrylate
Mild	2.7	Single BondUniversal	3M Oral Care; St Paul,MN, USA	2-hydroxyethyl methacrylate, bisphenol A diglycidyl etherdimethacrylate, decamethylene dimethacrylate, ethanol, silanetreated silica, water, 2-propenoic acid, 2-methyl-, reactionproducts with 1,10-decanediol and phosphorous oxide,copolymer of acrylic and itaconic acid, dimethylamino ethylmethacrylate, camphorquinone, dimethylaminobenzoate, 2,6-di-tert-butyl-P-cresol
	2.5	Adhese Universal	voclar Vivadent;Schaan, Liechtenstein	2-hydroxyethyl methacrylate, bisphenol A diglycidyl etherdimethacrylate, ethanol, 1,10-decandiol dimethacrylate,methacrylated phosphoric acid ester, campherquinone,2-dimethylaminoethyl methacrylate
	2.5	Prime&Bond Elect	Dentsply Caulk;Milford, DE, USA	Acetone, urethane dimethacrylate resin, dipentaerythritolpentaacrylate phosphate, polymerizeable dimethacrylate resin,polymerizeable trimethacrylate resin
	2.4	OptiBond XTR Primer	Kerr; Orange, CA, USA	Acetone, 2-hydroxyethyl methacrylate, ethanol
		OptiBond XTRAdhesive	Kerr	Ethanol, 2-hydroxyethyl methacrylate, 2-hydroxy-1,3-propanediylbismethacrylate, propylidynetrimethanol, ethoxylated, esterswith acrylic acid, alkali fluorosilicates
	2.3	Futurabond M+	VOCO; Cuxhaven,Germany	Bisphenol A diglycidylmethacrylate, ethanol, acidic adhesivemonomer, catalyst.
	2.3	Clearfil UniversalBond	Kuraray Noritake;Okayama, Japan	Bisphenol A diglycidylmethacrylate, 2-hydroxyethylmethacrylate, ethanol, 10-methacryloyloxydecyl dihydrogenphosphate, hydrophilic aliphatic dimethacrylate, colloidal silica,dl-camphorquinone, silane coupling agent, accelerators,initiators, water
Intermediately strong	1.5	G-aenial Bond	GC; Tokyo, Japan	Acetone, dimethacrylate, phosphoric acid ester monomer,dimethacrylate component, photoinitiator, butylatedhydroxytoluene.
	1.2	Peak Universal BondPrimer	Ultradent; SouthJordan, UT, USA	Ethyl alcohol, methacrylic acid, 2-hydroxyethyl methacrylate
	1.2	Peak Universal BondAdhesive	Ultradent	Ethyl alcohol, 2-hydroxyethyl methacrylate, methacrylic acid,chlorhexidine di(acetate)

The main ingredients, included in the Table 1 are according to the respective manufacturer. It should be noted that sometimes the companies are not willing to disclose all the components.

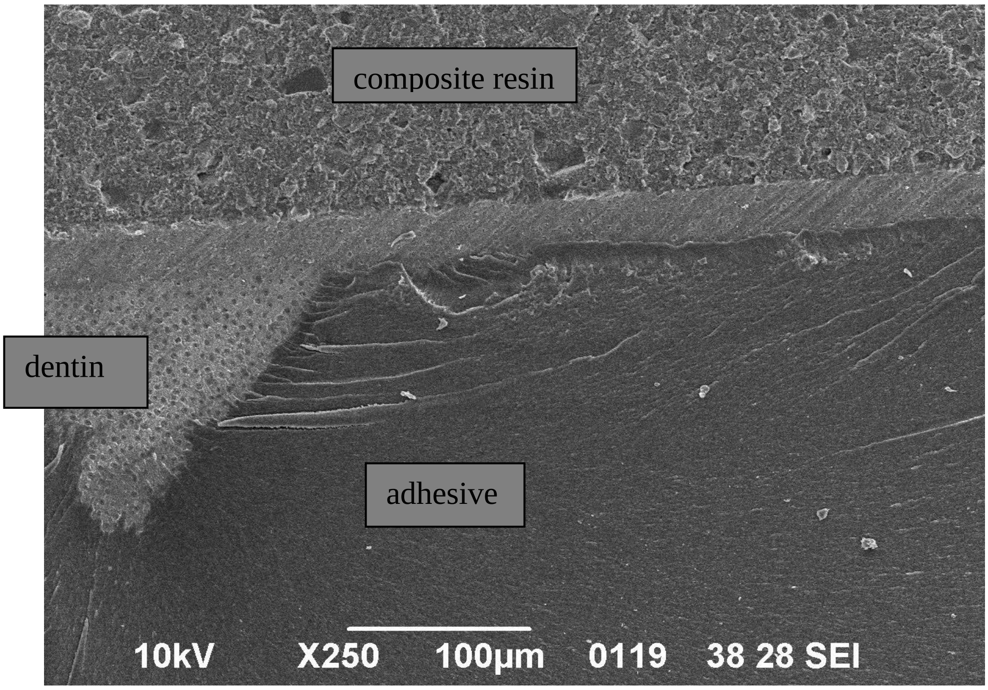

Fig.2. Typical (mixed) adhesive fracture surface of a µTBS test (SEM image – courtesy of the author).

2. Methods

For the purposes of this review paper, the databases of PubMed and Web of science were scanned, using the key words (universal adhesives; long-term microtensile bond strength; human dentin). Both databased turned out a total of 40 papers. Publications before 2010 were intentionally excluded, since a widely accepted classification of dental adhesives [1], places UAs as 8th generation adhesives that appeared on the market, starting from 2010. The inclusion criteria for the review were set to retain articles that used human dentin as a bonding substrate and the µTBS as a main method of testing. After applying the inclusion criteria and excluding the duplicates, the number of documents was narrowed down to 16 and the full texts were retrieved and analysed.

3. Results

The different articles included in the present review, investigated various factors and their influence on the µTBS to dentin. These included: pulpal pressure simulation, dentin moisture levels and etching times, inclusion of antibacterials, inclusion of dentin cross-linkers, air blowing time and bond application times.

Six studies were consistent with their conclusions that a long-term stable bond is achived when a self-etch protocol is followed [12, 13, 14, 15]. One of them come up with conclusions that selective etching with conventionally used H_3PO_4 [16] for 3 sec. produce superior long-term dentin bonding with improved effectiveness, while longer etching times should be avoided. Two studies [17, 18], employed antibacterials – chlorhexidine and 12-MDPB, respectively and reported a statistical difference in µTBS when compared to control groups. However, one study investigating the antibacterial agent tt-Farnesol reported alteration of bonding properties.

Dentin cross-linkers were investigated in two studies [19, 20] and were reported to be beneficial to improve the longevity of UA systems and to preserve the stability of the adhesive interface.

The effects of air blowing time and water storage were reported to be material-dependent [21]. Moreover, water storage induced approximately 50% reduction in dentin bond strength, regardless of adhesive strategy employed [22].

Dry surfaces facilitated optimal bonding for HEMA-free adhesives [23]. The application time was shown to be also important and compromise the performance of UAs when shortened time protocols were applied [24].

4. Discussion

The UAs have complex chemistry, which has to provide for their hydrophobic and hydrophylic properties. They differ from each other in ingredients, solvents and acidity (Table 1). Water is a main component in all UA compositions and it is next to impossible to be removed from the adhesive. The reason for this is the decreased vapour pressure, caused by the resin functional monomers and the osmotic gradient, which is responsible for resins' hydrolysis and disruption of collagen fibres [25, 26]. It has long been considered that the use of 10-MDP monomer can produce calcium salts (10-MDP-Ca) and the phenomenon called nanolayering contributes for collagen protection and less bond degradation. Currently, it can be accepted that after aging in water, the UAs that work in self-etch mode, produce more durable bonds than those in etch and rinse mode [13].

5. Conclusions

Although the short-term μTBS of the UAs is well-studied [27] and the main tendencies and strategies for enhancing bond durability are clearly defined, further clinical and *in vivo* studies are warranted to validate all the preliminary conclusions regarding the use of universal adhesives.

References

[1] Van Meerbeek, B., Yoshihara, K., Van Landuyt, K., Yoshida, Y., Peumans, M., 2020. From Buonocore's Pioneering Acid-Etch Technique to Self-Adhering Restoratives. A Status Perspective of Rapidly Advancing Dental Adhesive Technology. J Adhes Dent 22, 7-34.
[2] Muñoz, MA., Luque-Martinez, I., Malaquias, P., Hass, V., Reis, A., Campanha, NH., Loguercio, AD., 2015. In vitro longevity of bonding properties of universal adhesives to dentin. Oper Dent 40, 282-92.
[3] Alex, G., 2015. Universal adhesives: the next evolution in adhesive dentistry? Compend Contin Educ Dent 36, 15-26.
[4] Van Meerbeek, B., Yoshihara, K., Yoshida, Y., Mine, A., De Munck, J., Van Landuyt, K.L., 2011. State of the art of self-etch adhesives. Dent Mater. 27, 17-28.
[5] Suh, B.I., Feng, L., Pashley, DH., Tay, FR., 2003. Factors contributing to the incompatibility between simplified-step adhesives and chemically-cured or dual-cured composites. Part III. Effect of acidic resin monomers. J Adhes Dent 5,4, 267-82.
[6] Suh BI., 2013. Principles of Adhesive Dentistry: A Theoretical and Clinical Guide for Dentists. Newtown, PA: AEGIS Publications.
[7] Suh, BI., Chen, L., Brown, DJ., 2011. A novel concept: the introduction of crosslinking monomers into a self-etch adhesive to create a more hydrophobic and durable bond. Oral Health 94, 62-66.
[8] Nakabayashi, N., Kojima, K., Masuhara E., 1982. The promotion of adhesion by the infiltration of monomers into tooth substrates. J Biomed Mater Res 16, 265-273.
[9] Nakabayashi, N., Pashley, D.H., Hybridization of dental hard tissues. Quintesence Publishing; Chicago, 1998.
[10] Sano, H., Shono, T., Sonoda, H., Takatsu, T., Ciucchi, B., Carvalho, R., Pashley, DH., 1994. Relationship between surface area for adhesion and tensile bond strength--evaluation of a micro-tensile bond test. Dent Mater. Jul;10236-40.
[11] Sano, H., Chowdhury, AFMA,. Saikaew, P., Matsumoto, M., Hoshika, S., Yamauti, M., 2020. The microtensile bond strength test: Its historical background and application to bond testing. Jpn Dent Sci Rev. 56, 24-31.
[12] Lezaja Zebic, M., Dzeletovic, B., Miletic, V., 2018. Microtensile bond strength of universal adhesives to flat versus Class I cavity dentin with pulpal pressure simulation. J Esthet Restor Dent 30, 240-248.
[13] Zhang, ZY., Tian, FC., Niu, LN., Ochala, K., Chen, C., Fu, BP., Wang, XY., Pashley, DH., Tay, FR., 2016. Defying ageing: An expectation for dentine bonding with universal adhesives? J Dent 45, 43-52.
[14] Maciel Pires, P., Dávila-Sánchez, A., Faus-Matoses, V., Nuñez Martí, JM., Lo Muzio, L., Sauro, S., 2022. Bonding performance and ultramorphology of the resin-dentine interface of contemporary universal adhesives. Clin Oral Investig 26, 4391-4405.
[15] Nonato, RF., Moreira, PHA., Silva, DOD., Ferreira, MWC., Reis, A., Cardenas, AFM., Loguercio, AD., Siqueira, FSF. 2022. Long-term Evaluation of Bonding Performance of Universal Adhesives based on Different Dentinal Moisture Levels. J Adhes Dent. 24, 395-406.
[16] Stape, THS., Wik, P., Mutluay, MM., Al-Ani, AAS., Tezvergil-Mutluay, A., 2018. Selective dentin etching: A

potential method to improve bonding effectiveness of universal adhesives. J Mech Behav Biomed Mater 86, 14-22.
[17] Giacomini, MC., Scaffa, PMC., Gonçalves, RS., Zabeu, GS., Vidal, CMP., Carrilho, MRO., Honório, HM., Wang, L., 2020. Profile of a 10-MDP-based universal adhesive system associated with chlorhexidine: Dentin bond strength and in situ zymography performance. J Mech Behav Biomed Mater 110:103925.
[18] Belmar da Costa, M., Delgado, A.H., Amorim Afonso, T., Proença, L., Ramos, AS., Mano Azul, A.,2021. Investigating a Commercial Functional Adhesive with 12-MDPB and Reactive Filler to Strengthen the Adhesive Interface in Eroded Dentin. Polymers (Basel). Oct 15;13(20):3562.
[19] Comba, A., Maravić, T., Villalta, V., Tozzola, S., Mazzitelli, C., Checchi, V., Mancuso, E., Scotti, N., Tay, FR., Breschi, L., Mazzoni, A., 2020. Effect of an ethanol cross-linker on universal adhesive. Dent Mater 36, 1645-1654.
[20] Dávila-Sánchez, A., Gutierrez, M.F., Bermudez, JP., Méndez-Bauer, ML., Hilgemberg, B., Sauro, S., Loguercio, AD., Arrais, CAG., 2020. Influence of flavonoids on long-term bonding stability on caries-affected dentin. Dent Mater 36, 1151-1160.
[21] Saikaew, P., Fu, J., Chowdhury, AFMA., Carvalho, RM., Sano, H., 2019. Effect of air-blowing time and long-term storage on bond strength of universal adhesives to dentin. Clin Oral Investig 23, 2629-2635.
[22] Vinagre, A., Ralhoa, A., Ramosa, N., Messiasb, A., Ramos, JC., 2019. Bonding performance of a universal adhesive: effect of hydrophobic resin coating and long-term water storage. Revista Portuguesa de Estomatologia, Medicina Dentária e Cirurgia Maxilofacial. DOI:10.24873/j.rpemd..11.463.
[23] Saeed, NA., Tichy, A., Kuno, Y., Hosaka, K., Tagami, J., Nakajima, M., 2021. Effect of Surface Moisture on Bur-cut Dentin on Bonding of HEMA-free and HEMA-containing Universal Adhesives with or without Methacrylamide Monomer. J Adhes Dent 23, 327-334.
[24] Saikaew, P., Matsumoto, M., Chowdhury, A., Carvalho, RM., Sano, H., 2018. Does Shortened Application Time Affect Long-Term Bond Strength of Universal Adhesives to Dentin? Oper Dent 43, 549-558.
[25] Pashley, EL., Zhang, Y., Lockwood, PE., Rueggeberg, FA., Pashley, DH., 1998. Effects of HEMA on water evaporation from water-HEMA mixtures. Dent Mater 14, 6-10.
[26] Tay, FR., Pashley, DH., Suh, BI., Hiraishi, N., Yiu, CK., 2005. Water treeing in simplified dentin adhesives--déjà vu? Oper Dent 30, 561-79.
[27] Koytchev, E., Takahashi, Y., Suzaki, N., Okamoto, M., Hayashi, M., 2020. Bonding performance of a newly developed one-step single-bottle adhesive on sound human dentin with different tubule orientations. Series on Biomechanics 34, No.1, 3-11.