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Dental adhesives and strategies for displacement of water/solvents from collagen fibrils

Larissa Sgarbosa de Araújo Matuda[a,b,*], *Giselle Maria Marchi*[a], *Thaiane Rodrigues Aguiar*[b], *Ariene Arcas Leme*[b], *Gláucia M.B. Ambrosano*[c], *Ana Karina Bedran-Russo*[b]

[a] *Department of Restorative Dentistry Piracicaba Dental School, University of Campinas, 13414-903 Campinas, São Paulo, Brazil*

[b] *Department of Restorative Dentistry, University of Illinois at Chicago, 60612 Chicago, IL, USA*

[c] *Department of Community Dentistry Piracicaba Dental School, University of Campinas, 13414-903 Campinas, São Paulo, Brazil*

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ABSTRACT

Objectives. To evaluate the influence of temperature of evaporation in adhesive systems with different solvents on the apparent modulus of elasticity and mass change of macro-hybrid layers modified by proanthocyanidins (PACs).

Methods. Adhesive resin beams (A) from Single Bond Plus (SB), Excite (EX) and One Step Plus (OS) were prepared after solvent evaporation at 23 °C or 40 °C ($n = 12$). Macro-hybrid layers (M) ($n = 12$) were prepared using demineralized dentin beams sectioned from extracted human third molars. The demineralized dentin specimens were infiltrated with each one of the three adhesive systems at 23 °C or 40 °C; with or without prior dentin treatment with PACs for 10 min. The apparent modulus of elasticity (E) and mass change (W_{mc}, %) of adhesives beams and resin-infiltrated specimens were assessed in dry and wet conditions after immersion in water (24 h, 1, 3 and 6 months). The E was statistically analyzed by Tukey–Kramer test and the W_{mc}, % by Kruskal Wallis, and Dunn ($\alpha = 0.05$).

Results. Solvent evaporation at 40 °C resulted in higher E values for adhesive resin beams at all storage conditions, regardless of the adhesive system ($p < 0.05$). Increased mass loss (3 months: −0.01%; 6 months: −0.05%) was observed in One Step resin beams ($p \leq 0.05$). In the macro-hybrid layer models the pretreatment with PACs along with solvent evaporation at 40 °C increased E and decreased the W_{mc}, % (3 months: −2.5; 6 months: 2.75%) for adhesives evaluated over time ($p < 0.05$). No significant differences in ratio (resin/dentin) were found for the macro-hybrid layers ($p > 0.05$).

Significance. Improved solvent evaporation at higher temperature, and increased collagen cross-linking induced by PACs, enhanced the mechanical properties resulting in highly stable macro-hybrid layers over 6 months storage.

* *Corresponding author at*: Departamento de Odontologia Restauradora, Faculdade de Odontologia, UNICAMP, Av. Limeira, 901-Areião, CEP-13414-903 Piracicaba, SP, Brazil. Tel.: +55 19 2106 5200; fax: +55 19 3421 0144.
E-mail address: larissasgarbosa@hotmail.com (L.S.d.A. Matuda).

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1. Introduction

The clinical success and longevity of dental adhesive restorations depends on the stability of the resin–dentin interfacial components. The infiltration of resin monomers into a mineral depleted dentin matrix, results in the formation of a hybrid layer. Thus, the presence and the quality of the hybrid layer are believed to be essential for adhesion to dentin [1,2]. However, there is no evidence that water in the intra and inter-microfibrillar compartments of the collagen matrix is completely replaced by resin [3–5]. The remaining water present in dentin during resin infiltration increases the water absorption of hydrophilic resins [6], plasticize polymers, and accelerate degradation of the adhesive interface [7].

The resin co-monomers are dissolved in organic solvents that facilitate monomer diffusion within the collagen fibrils, and assist with the removal of water during solvent evaporation [8,9]. Therefore, the residual water in the dentin [5,10] and the organic solvents in adhesive systems [11] need to be adequately volatilized as they can interfere in the mechanical properties of the hybrid layer [1]. Clinical techniques using heat for evaporation of solvents result in higher mechanical properties [12–16] and decreases interfacial micro permeability [13,15]. However, despite the fact that these techniques have increased the evaporation of the solvents, there are limited long-term studies evaluating such techniques. Although evaporation of excess solvents can promote a highly cross-linked resin polymer, the hydrophilic nature of the monomer would not change, thus water absorption could plasticize the polymer chains and degrade the adhesive interface at the long-term [13].

The modification of dentin by plant-derived proanthocyanidins (PACs) modify the structure of collagen by increasing inter- and intra-molecular cross-links [17], enhancing the mechanical properties of the hybrid layer [18] and increasing the hybrid layer resistant to degradation [19]. The enhanced mechanical properties may also be due to the type of bonds between PACs and collagen, decreasing the hydrophilicity of the dentin matrix [20,21]. This could decrease the degradation of the adhesive interface caused by long-term water absorption. In this context, flexural tests of macro-hybrid layer models can provide valuable information regarding the quantification of matrix shrinkage and resin uptake, as well as mechanical properties of the hybrid layer after infiltration of the adhesive [22–25].

Therefore, this study evaluated the association of tissue biomodification and temperatures for solvent evaporation to minimize the effects of water from the dentin substrate and excess of solvents from adhesives systems on the apparent modulus of elasticity and mass change of macro-hybrid layers over a 6-month storage period. The null hypothesis tested was that the biomodification of dentin matrix associated to warm temperature of solvent evaporation (40 °C) would not affect the mass change and modulus of elasticity of macro-hybrid layers created by different adhesive systems.

2. Materials and methods

2.1. Adhesive systems

Commercially available etch-and-rinse adhesive systems with different solvents were selected: Single Bond Plus [SB – ethanol and water based], 3M/ESPE, St. Paul, MN, USA, One Step Plus [OS – acetone based], Bisco, Schaumburg, IL, USA and Excite F [EX – ethanol based], Ivoclar Vivadent AG, Bendererstrasse, Schaan, Liechtenstein. The composition of the adhesive systems is described in Table 1.

2.2. Adhesive resin beam preparation

One mL of each adhesive system was dispensed in a circular-plate and their initial mass was measured using an analytical balance (XP 504DR, Mettler Toledo Inc., Columbus, OH, USA). Each circular plate with the adhesive system was repeatedly weighed after 15, 30, 45 and 60 min until weight was stabilized. To keep traces of solvent in the adhesive, the time of volatilization was set at 30 min (based on pilot studies). The adhesive systems were kept in the dark for 30 min in an oven at 40 ± 1 °C or at room temperature (23 ± 1 °C). After time elaspsed, the adhesives were placed in polyvinyl siloxane molds (0.28 mm × 1.5 mm × 7.0 mm) and were immediately light-cured for 60 s on each side. Photo-activation was performed using a halogen-based unit (Optilux 501, Kerr Corp., Orange, CA, USA) with light intensity of 800 mW/cm^2. Then, all adhesive resin beams (n = 12 per group) with final dimensions 0.28 mm thickness × 1.5 mm width × 7.0 mm length were

Table 1 – Compositions of adhesive systems tested.

Adhesive systems	Composition	Batch number
Excite F (Ivoclar Vivadent)	15% HEMA, dimethacrylates, Phosphonic acid acrylate, 25% Bis GMA highly dispersed silicone dioxide, initiators, stabilizers, potassium fluoride, 20% ethanol	R78651
Adper Single Bond Plus (3M ESPE)	25–35% ethyl alcohol 10–20% BIS-GMA 10–20% silane treated silica 5–15% HEMA 5–10% copolymer of acrylic and itaconic acids 5–10% glycerol 1,3-dimethacrylate <5% water 1–5% UDMA	N446453
One Step Plus (Bisco)	15–40% BPDM 10–40% HEMA 40–70% acetone photoinitiators	1200007653