In Vitro Assessment of Solvent Evaporation from Commercial Adhesive Systems Compared to Experimental Systems

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Solvents should be properly evaporated after application to dental substrates. The aim of this study was to assess the evaporation of commercial, experimental and neat solvents. The tested null hypotheses were that there are no differences in solvent evaporation regardless of its formulation and over time. Evaporation from commercial adhesive systems (Scotchbond Multipurpose Primer, Scotchbond Multipurpose Adhesive, Prime & Bond NT, Multi Bond, Excite, Single Bond 2, Adhese Primer, Adhese Bond, Xeno III A and Xeno III B) and experimental primers (35% HEMA plus 65% acetone or ethanol or water v/v) were compared to neat solvents (acetone, ethanol and water). Samples (10 µL) of these products were dripped into glass containers placed on a digital precision balance. Evaporation was assessed at 0, 5, 10, 15, 30, 60, 120, 300 and 600 s times to calculate mass loss. Data were analyzed statistically by ANOVA and Bonferroni's correction (α=0.05). Acetone-based products exhibited a remarkable capacity to evaporate spontaneously over time. Neat acetone evaporated significantly more than the HEMA-mixtures and the commercial formulations (p<0.05). The incorporation of monomers and other ingredients in the commercial formulations seem to reduce the evaporation capacity. Solvent evaporation was time and material-dependent.

Key Words: dentin bonding agents, solvents, evaporation.

INTRODUCTION

Biological and chemical factors are both relevant to obtain an optimal adhesion to tooth structure, mainly to dentin substrate (1). Based on the principles of hybrid layer formation, dentin demineralization followed by adequate resin infiltration and polymerization are key steps in the adhesive protocol to ensure the longevity of restorations (1). One of the greatest challenges in adhesion is related to the need of dentin being slightly moist before being properly bonded (1). Water is an essential component of dentin matrix to prevent the collapse of the collagen network after etch step. However, excessive moisture can adversely affect hybrid layer durability due to degradation of either collagen fibrils or resin material (2).

Different solvents presented in primer component or in simplified bonding agents are responsible for either carrying excess water out or infiltrating resin monomers into interfibrilar dentin (3). As solvent is necessary to provide a proper infiltration of the resin monomers into demineralized collagen matrix, the bonding process still depends on its capacity (3,4). Benefits offered by solvents rely on their properties of improving substrate wetting, aiding to impede the collagen fibrils collapse or to stiffen them (3). However, solvents must be elimi-

nated after having completed their function because it has been demonstrated that residual solvent can lead to deterioration of the adhesive interface (2) by interfering with resin polymerization (1,3,5) and decreasing mechanical properties (4-9).

Water, ethanol and acetone are basically the main solvents in commercial formulations (3,6-8,10). A combination of some physical and chemical factors, namely vapor pressure, molar fractions, molar weight and solubility, is considered to determine the evaporation capacity of different solvents (8,11-14). Although non-solvated resins may establish a more stable and less fragile adhesion to dentin compared to simplified solvated products, bonding durability to this substrate is still a clinical challenge (13,15).

Previous studies have shown differences in the capacity of either experimental solvents or commercial formulations to evaporate (14), but no comparative study is currently available. Such an investigation would be interesting because commercial formulations contain ingredients like initiators, co-monomers and/or nanoparticles that can affect solvent evaporation rate. Although previous reports have assessed experimental primers based mainly on HEMA and solvent, their performance might be different from commercial formulations since the latter present other ingredients that modify evaporation rate. Therefore, the aim of this study was to evaluate the evaporation of solvents from commercial adhesive systems, experimental primers and neat solvents. The following null hypotheses were tested: 1. There is no difference in solvent evaporation regardless of its formulation; 2. There is no difference in solvent evaporation over time.

MATERIAL AND METHODS

A list of the commercially available systems, experimental systems and neat solvents that were used in the present study is shown in Table 1. HEMA-based mixtures and neat solvent were considered control groups to commercial materials.

Using a micropipette (Gilson SA, Villers-le-Bel, France), 10 μL of each product obtained from the original container was transferred to a small glass receptacle of known weight placed on a digital precision balance (model 2104N; Bioprecisa, Curitiba, PR, Brazil) at controlled temperature and humidity conditions (21°C and 65% relative humidity). This mass registration was

considered as the initial mass (time 0). It is important to highlight that this evaporation was spontaneous, with no strategy to facilitate the evaporation.

Thereafter, the balance was opened to facilitate solvent evaporation and the mass was registered after different intervals (5, 10, 15, 30, 60, 120, 300 and 600 s) to calculate the percentage loss of mass over time using the initial mass as a reference. The mass of each time was subtracted from the initial mass and divided by initial time. Mass monitoring was repeated 6 times *per* product. Mass loss of the tested products was normalized to percentage for better comparison of their evaporation capacity over time. Data were submitted to statistical analysis by ANOVA and multiple test comparisons were performed by Bonferroni's correction according to time to each material and among products to each evaluated time. Significance level was preset at 5%.

RESULTS

There was statistical significance for time, material and interaction between both factors ($p<0.001$). For better interpretation of the results, the tested materials were grouped according to the main solvent present in the formulation (acetone, ethanol or water).

Figure 1 presents the percent mass loss of neat acetone, ethanol and water within 600 s of spontaneous evaporation. These products served as a control for the tested commercial and experimental formulations.

For the acetone-based materials, mass loss decreased significantly ($p<0.05$) over time with a remarkable difference between the mass registered at all time points and the initial mass (time 0). At the end of study, acetone-based materials, that is, neat acetone, acetone-HEMA, Prime & Bond NT and Multi Bond Uno presented total mass loss of 79.16, 23.42, 42.02
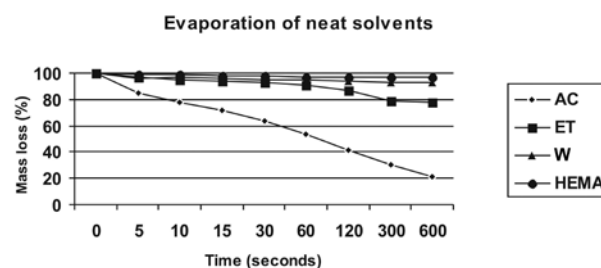

Figure 1. Mass loss (%) of the 3 neat solvents over time compared to HEMA.

Table 1. Tested materials*.

Material	Type	Manufacturer	Batch	Composition
Adper SBMP - primer	ERP	3M/ESPE, St Paul, MN, USA	3NL (2006/10)	Polyalkenoic acid, HEMA (30- 40%), W
Adper SBMP - adhesive	ERBA	3M ESPE, St Paul, MN, USA	5PJ (12/2008)	Bis-GMA (60-70%), HEMA (30- 40%), CQ
Prime & Bond NT	SERA	Dentsply, York, PA, USA	494615 (03/2007)	UDMA, PENTA, R5-62-1 resin, T-resin, D-resin, Butylated hydroxitoluene, EDMAB, Cetylamine hydrofluoride, AC, Nanofiller, IS
Multi Bond Uno	SERA	DFL, Rio de Janeiro, RJ, Brazil	06111659 (11/2008)	PMGDM, HEMA, PHFA, AC, CQ, EDMAB, BHT
Excite	SERA	Ivoclar/Vivadent, Schaan, Liechtenstein	F61706 (2006/01)	Phosphonic acid acrylate (<12%), HEMA (<21%), dimethacrylates (<45%), ET (<26%), SiO2, IS
Adper Single Bond 2	SERA	3M ESPE, St Paul, MN, USA	4BR (2007/11)	Ethyl alcohol (25-35%), Bis-GMA (10-20%), silica-treated nanoparticles (10-20%), HEMA (5-15%), glycerol 1, 3 dimethacrylate (5-10%), acrylic acid copolymer and itaconic acid (5-10%), diurethane dimethacrylate (1-5%), W (<5%)
AdheSE - primer	SEP	Ivoclar/Vivadent, Schaan, Liechtenstein	F-46035 (2006/03)	HEMA (<25%), dimethacrylates (<75%), phosphonic acrylic acid (<40%), IS
AdheSE - adhesive	SEBA	Ivoclar/Vivadent, Schaan, Liechtenstein	F-46035 (2006/03)	HEMA (<25%), dimethacrylates (<75%), SiO2, IS
Xeno III	SES	Dentsply, York, PA, USA	0305001039 (2005/04)	Liquid A: HEMA, purified water, ET, UDMA resin, BHT, highly dispersed silicon dioxide Liquid B: Phosphoric acid modified polymethacrylate resin, mono fluoro phosphazene modified methacrylate resin, UDMA resin, BHT, CQ, EDMAB
AC	S	Synth, Diadema, SP, Brazil	840889 (07/2008)	-
ET	S	Dinâmica, São Paulo, SP, Brazil	20161 (12/2009)	-
W	S	-	-	-
HEMA	RM	Sigma-Aldrich, St. Louis, MN, USA	S24239-215 (07/2010)	-
HEMA + AC	EM	-	-	35% AC, 65% HEMA
HEMA + ET	EM	-	-	35% ET, 65% HEMA
HEMA + W	EM	-	-	35% W, 65% HEMA

*Manufacturers' information. AC = Acetone; ET = Ethanol; W = Water; HEMA = 2-hidroxyethyl methacrylate; ERP = Etch-and-rinse primer; ERBA = Etch-and-rinse bonding agent; SERA = Simplified etch-and-rinse agent; SEP = Self-etching primer; SEBA = Self-etching bonding agent; SES = Self-etching system; S= Solvent; RM = Resin monomer; EM = Experimental mixture; Bis-GMA = Bisfenol diglycidyl dimethacrylate; CQ = Camphoroquinone; IS = Initiators and stabilizers; EDMAB = Ethyl 4-dimethyl amino benzoate; UDMA = Urethane dimethacrylate; BHT = butylated hydroxytoluene; PMGDM= pyromellitic dianhydride and glycerol dimethacrylate; PHFA= potassium hexafluoroantimonate.

and 42.55%, respectively (p<0.05) (Fig. 2A). The mass loss of HEMA, Prime & Bond NT and Multi Bond Uno (acetone-based materials) differed significantly from that of neat acetone, at time 0 (initial mass), 5 s, 30 s and 300 s to respectively. Considering all tested products regardless of the solvent type, ethanol-, water- and ethanol/water-based materials presented a remarkably lower solvent evaporation capacity than that of the acetone-base materials with mass loss ranging from 2.15 to 21.80%.

The ethanol-based products, that is, neat ethanol, ethanol-HEMA mixture and Excite, presented significant (p<0.05) mass loss of 21.80, 5.14 and 6.65%, respectively (Fig. 2B). Only neat ethanol showed statistically significant difference in mass loss over time (at 300 and 600 s).

Water was the solvent of water-HEMA mixture, Scotchbond Multipurpose primer and Adhese 1. In the same as the other neat solvents, water showed a great mass loss (7.18%), followed by Adhese 1 (6.17%) and Scotchbond Multipurpose primer (3.60%). The experimental water-HEMA mixture showed a mass loss of 2.43% at the 600-s evaluation period (Fig. 2C).

Xeno III and Single Bond 2 present both water and ethanol in their composition and were analyzed separately. These systems lost 8.04% and 6.10% of their mass, respectively, at the end of the evaluation period (Fig. 2D). HEMA evaporation at the evaluated times showed statistical significance only at 600 s. Total mass retention (%) within the 600-s evaluation period is presented graphically in Figure 3.

DISCUSSION

Due to the role attributed to solvents in the adhesive protocol, their evaporation capacity from different experimental products has been investigated (14). There are also few studies with commercial adhesive systems, such as those of Abate et al. (6) and Lima et al. (8), who have monitored this property directly from the product vials. In every clinical practice, adhesive system vials are frequently left opened during the restorative procedure, which facilitates solvent volatization. As the solvent evaporates from the open vial, the viscosity of the adhesive increases and the amount of residual vehicle to carry the resin monomer within the demineralized
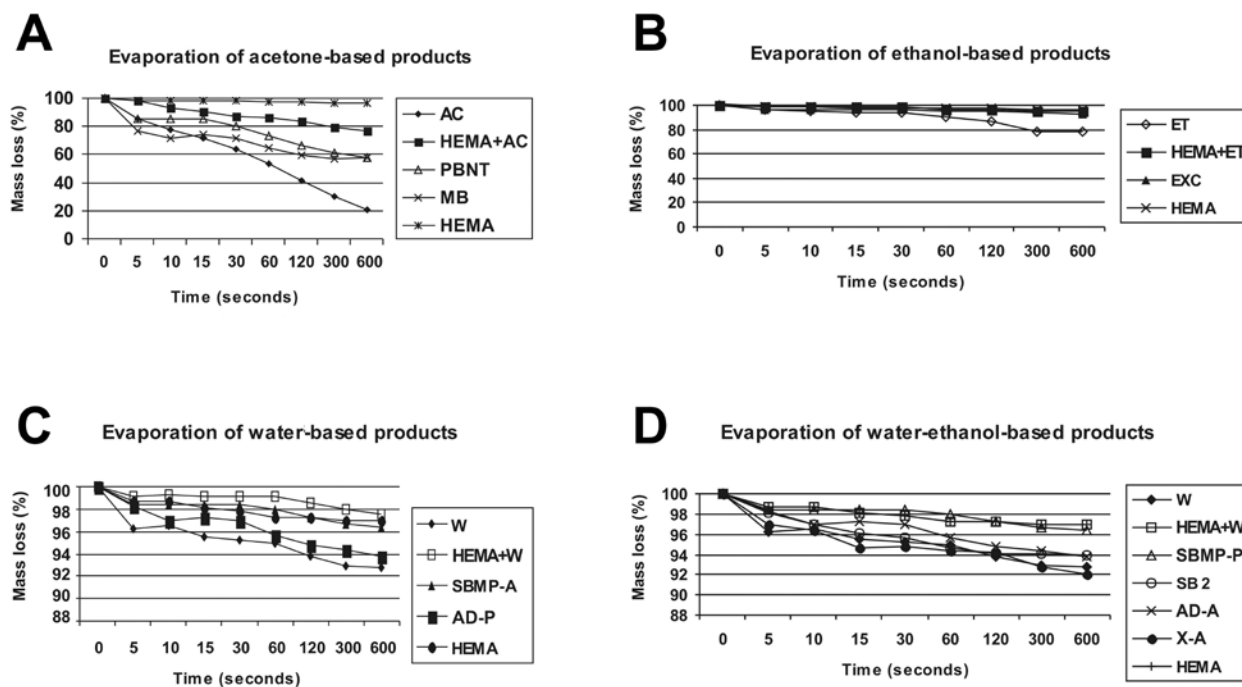

Figure 2. Mass loss (%) of products over time. A= Acetone-based products; B= Ethanol-based products; C= Water-based products; D= Water-ethanol-based products.

dentin matrix and involve collagen fibrils may not be sufficient, compromising the quality of the adhesive protocol (8,16). On the other hand, solvent evaporation after application to dentin is extremely important because failure to remove excess solvent by gentle air drying may contribute to the degradation of the adhesive interface over time (2-9).

In the present study, the tested products were grouped according to solvent type to allow better comparison. Based on the results of the proposed experimental design, it was observed that the acetone-based materials presented a greater evaporation capacity compared to all other products. These results are in accordance with those of Yiu et al. (15) after examining the solvent and water retention in dental adhesive blends after evaporation. Commercial formulations presented less solvent evaporation compared to neat solvent, especially for the acetone-based products. In this study, solvent evaporation appeared as the main responsible for product mass loss.

It is clear that other components of the adhesive system have potential to limit spontaneous evaporation of the solvent. Pashley et al. (14) have claimed for the effect of monomer (HEMA) as a solute. As solvents evaporate over time, HEMA concentration increases, which lowered the pressure vapor of the experimental bonding mixture in the present study. By observation of the HEMA-acetone profile, HEMA contributed for less evaporation when compared to neat solvent. Commercial acetone-based products presented intermediary values, which can indicate a less concentration of HEMA and greater acetone content. It seems to occur in a similar manner when ethanol and water-mixtures were compared to their respective neat solvents and experimental primers. The results of the present investigation indicate that HEMA is an adhesive system component that potentially interferes with solvent evaporation.
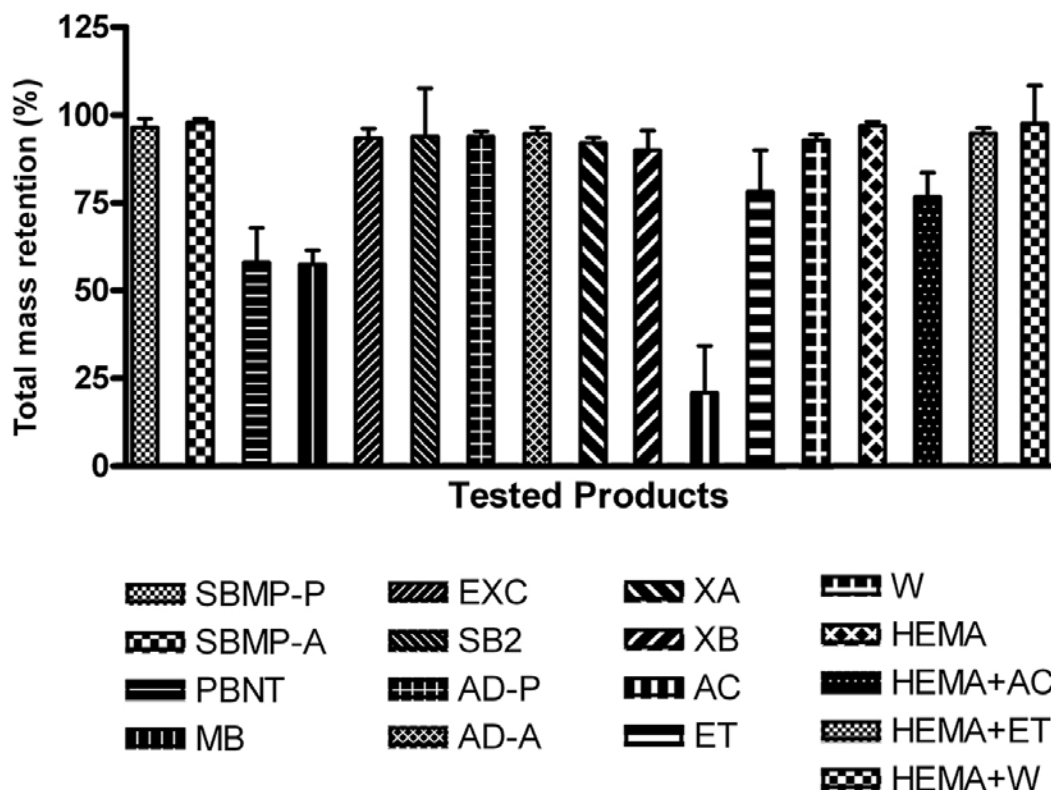
Although it was clear that commercial formula-

Figure 3. Total mass retention (%) within the 600-s evaluation period. Same letters indicate no statistically significant differences among the products.

tions had less evaporation than neat solvents, the limitations of the methodology did not allow determining any specific influence of the action of photoinitiators, nanoparticles and co-monomers as possible solutes. In simplified adhesive systems, nanofiller incorporation has the role of improving the viscosity in order to prevent over-thinning of unfilled solvated adhesive layers and to being helpful to reduce polymerization shrinkage (17).

Solvent evaporation depends on different factors, mainly molecular weight and vapor pressure (11,12). The higher the molecular and the vapor pressure, the greater the evaporation rate. Regarding the characteristics of the solvents and monomers presented in the commercial formulations, solvent can be more or less bound to the water presented in dentin. Clinically, solvent binding to the collagen fiber network contributes to reduce its evaporation (15). This solvent solubility is of great relevance because it influences the capacity to solvate the proteoglycans that involve collagen fibrils (11). The solvent should breaks the hydrogen bound to the collagen fibrils, leading resin monomers to infiltrate within the collagen network. Acetone-based products need water at higher percentage to interact with these fibrils compared to ethanol (11).

In addition to the properties of the main solvent and other components of each adhesive system, other technique factors such as cavity configuration and drying time can also influence solvent evaporation from adhesive systems. A recent study (16) has shown the effect of cavity configuration. Even when long air-drying was employed, there was evidence of ineffective drying and pooling of adhesive when applied in narrow Class I cavities, which resulted in solvent retention.

There seems not to exist a consensus regarding the recommendation for the drying time and manner of drying after application of adhesive systems to dentin. According to Jacobensen et al. (18) dentin adhesive systems after 10 s drying time or longer seem to perform better under microtensile tests. Significant increase in gap-free restorations was observed, attesting the relevance of allow solvent evaporation after application to dentin.

The findings of this study reinforce the need of following an accurate protocol in clinical situations such as root canal bonding treatment (13). The canal design is challenging for spontaneous evaporation of solvents, mainly water and ethanol. Based on a water sorption analysis, Fabre et al. (19) showed differences among commercial formulations of bonding agents by, indicating the presence of solvent as a condition to exacerbate water sorption.

Additionally, the presence of oxygen acts negatively on adhesive polymerization, contributing to make dental bonding a clinical challenge (20). These properties are determinant to degradation resistance.

The performance of the tested commercial adhesive systems suggests that they act within the expected evaporation of main solvent-content. Although other ingredients seem to act as solute as HEMA does, they contribute to makes spontaneous evaporation more difficult. Lima et al. (8) is one of the few studies that correlated solvent volatization characteristics with properties like microleakage. Poor results were observed when solvents are evaporated from the bottles before applied to dentin. Studies using commercial formulations are necessary to clarify the interferences of factors others than solvent.

In conclusion, solvent evaporation should be facilitated as spontaneous evaporation was not adequate to any tested material over time. Within the limitations of this study, the null hypotheses were rejected. Solvent evaporation varied over time and according to the different commercial adhesive systems.

RESUMO

O solvente deve ser adequadamente evaporado após aplicação ao substratos dentários. O objetivo deste estudo foi avaliar a evaporação de formulações comerciais, primers experimentais e solventes puros. As hipóteses nulas testadas foram de que não há diferenças da quantidade evaporada independentemente do material e tempo. Evaporação dos sistemas adesivos comerciais (Scotchbond multipurpose primer, Scotchbond multipurpose adhesive, Prime & Bond NT, Multi Bond, Excite, Single Bond 2, Adhese Primer, Adhese Bond, Xeno III A e Xeno III B) e primers experimentais (35% HEMA associado com 65% acetona, etanol ou água v/v) foram comparadas a solventes puros (acetona, etanol e água). Amostras (10 µL) de cada produto foram dispensadas em balança de precisão digital. As massas nos tempos 0, 5, 10, 15, 30, 60, 120, 300 e 600 s foram registradas. Os dados foram analisados estatisticamente por ANOVA e Bonferroni (α=0,05). Produtos a base de acetona exibiram maior capacidade de evaporação espontânea ao longo do tempo. Acetona pura evaporou significantemente mais que as misturas de HEMA e formulações comerciais (p<0,05). A incorporação de monômeros e outros ingredientes nas formulações comerciais reduzem a capacidade de evaporação. A evaporação é dependente do produto e do tempo.

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