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Laboratory Research

The Use of Warm Air Stream for Solvent Evaporation: Effects on the Durability of Resin-dentin Bonds

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Clinical Relevance

In terms of silver nitrate uptake, the use of a warm air stream may improve bonding interface resistance to degradation over time. In terms of bond strength values, a warm air stream only improved the resin-dentin bond strengths of the ethanol/water-based system.

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SUMMARY

This study evaluated the effect of a warm (W) or cold (C) air-dry stream for solvent evaporation on the immediate (IM) and six-month (6M) resin-dentin bond strength (μTBS) and silver nitrate uptake pattern (SNU) of two-step etch-and-rinse adhesive system (Adper Single Bond [SB] and Prime & Bond 2.1 [PB]). The adhesives were applied on demineralized dentin surfaces and a warm or cold air-dry stream (10 seconds) was applied followed by light-activation (10 seconds). After 24-hours of water storage, the specimens were serially sectioned in the "x" and "y" directions to obtain bonded sticks around 0.8 mm² to be tested immediately or after six months of water storage. The specimens at each period were immersed in a 50% solution of silver nitrate, photodeveloped and analyzed by SEM for SNU.

Higher IM μTBS values were observed for SB under W conditions. Both adhesives showed reductions in μTBS after 6M in both air temperatures. Regarding SEM, a low silver nitrate uptake was observed in the W groups either in IM or 6M for both adhesives.

INTRODUCTION

The major drawback of contemporary adhesive restoratives is their limited durability *in vivo*,[1] which has been attributed to loss of retention[1] and marginal adaptation.[2-4] This short-term durability seems to result from suboptimal longevity of the hybrid layer. Since the hybrid layer is created by a mixture of dentin organic matrix, residual hydroxyapatite crystallites, resin monomers and solvents, aging may affect each of the individual components or it may be due to synergistic combinations of degradation phenomena occurring within the hybrid layer. The degradation of unprotected collagen fibrils[5-9] recently attributed to host-derived matrix metalloproteinases;[10-11] the elution of resin monomers due to suboptimal polymerization[12] and degradation of resin components[14-15] are some of the factors involved in the reduction of the resin-dentin bonds observed by some authors.[16-22]

Hence, any approach to prolong the clinical lifetime of adhesives might focus on improving stability of the bonding interface of these biomaterials to tooth tissue. As a result, different clinical approaches have been proposed, such as increased application times of bonding agents,[23] multiple adhesive coating,[24] delayed polymerization,[21,25] adhesive rubbing[26-27] and longer exposure times of bonding systems.[12]

Another recent attempt was the use of a warm air stream to evaporate solvents from simplified etch-and-rinse adhesives.[28] The authors have demonstrated significantly higher bond strength values and lower silver nitrate penetration along the adhesive layer when a warm, instead a cold air-dry stream is used. However, one should consider that the percentage of hydrophilic monomers that are responsible for high degrees of permeability after polymerization and high silver nitrate uptake expression over time[20] cannot be changed by simple application of a warm air stream. Therefore, one should investigate whether this clinical procedure is effective in preventing or even retarding degradation of resin-dentin bonds.

The current study compared the effects of air stream temperature for solvent evaporation on the immediate and six-month microtensile resin-dentin bond strengths (μTBS) and nanoleakage pattern (SEM) of an ethanol/water-based and acetone-based simplified etch-and-rinse adhesive system.

METHODS AND MATERIALS

Microtensile Testing

Twenty extracted, caries-free human third molars were used. The teeth were collected after obtaining the patients' informed consent under a protocol approved by the University Estadual de Ponta Grossa Institutional Review Board. The teeth were disinfected in 0.5% chloramine, stored in distilled water and used within six months of extraction. A flat dentin surface was exposed after wet grinding the occlusal enamel on #180 grit SiC paper. The exposed dentin surfaces were further polished on wet #600-grit silicon-carbide paper for 60 seconds to standardize the smear layer.

Two simplified etch-and-rinse adhesive systems were tested: Adper Single Bond (SB, 3M ESPE, St Paul, MN, USA), an ethanol/water-based system and Prime & Bond 2.1 (PB, Dentsply Caulk, Milford, DE, USA) an acetone-based system. The composition, application mode and batch number are described in Table 1.

After acid etching with the respective etchants of each adhesive system, the surfaces were rinsed with distilled water for 20 seconds and air-dried for 20 seconds. The surfaces were then rewetted with water. Two coats of adhesive were lightly applied for 10 seconds. After each coat, solvent evaporation was performed either with warm (60 ± 2°C) or cold air (20 ± 1°C) for 10 seconds at a distance of 10 cm. In both cases, the air stream was generated by a commercial hair-dryer (SC831, Black & Decker, Uberaba, MG, Brazil). The air speed of the stream was 5.50 m/s and the air flow was 0.0138 m^3/s.

Table 1: *Adhesive Systems: Composition, Application Mode and Batch Number*

Adhesive Systems	Composition	Application Mode	Batch #
Adper Single Bond (3M ESPE)	1. Scotchbond Etchant–35% phosphoric acid 2. Adhesive–Bis-GMA, HEMA, dimethacrylates, polyalkenoic acid copolymer, initiators, water and ethanol	*a,b,c,d,e,f,e,f,g*	5FE
Prime & Bond 2.1 (Dentsply)	1. Caulk tooth Conditioner Gel–34% phosphoric acid 2. Adhesive–UDMA, PENTA, Bis-GMA, butylated hydroxytoluene, 4-ethyl dimethyl aminobenzoate, cetylamine hydrofluoride, initiator and acetone	*a,b,c,d,e,f,e,f,g*	707608

a–acid-etching (15 seconds); b–rinsing (15 seconds); c–air-drying (30 seconds); d–dentin rewetted with water; e–one coat of adhesive; f–air-dry for 10 seconds at 20 cm for solvent evaporation; g–light-curing (10 seconds–600 mW/cm²).

Bis-GMA: bisphenol A diglycidyl methacrylate; HEMA: 2-hydroxyethyl methacrylate; UDMA–urethane dimethacrylate; PENTA–dipentaerythritol pentaacrylate monophosphate.

The adhesives were light-cured for the respective recommended times using a quartz-tungsten halogen light set at 600 mW/cm^2 (VIP, BISCO, Schaumburg, IL, USA) (Table 1). Resin composite buildups (Z250, shade A2, 3M ESPE) were constructed on the bonded surfaces in three increments of 1 mm each, which were individually light-cured for 30 seconds with the same light intensity. All the bonding procedures were carried out by a single operator at a room temperature of 24°C and a constant relative humidity. Five teeth were used for each combination of adhesive system and air temperature.

After the restored teeth were stored in distilled water at 37°C for 24 hours, they were longitudinally sectioned in both a mesio-to-distal and buccal-to-lingual direction across the bonded interface with a diamond saw in a Labcut 1010 machine (Extec Corp, Enfield, CT, USA) to obtain sticks per tooth, each with a cross-sectional area approximately 0.8 mm^2. The number of premature debonded sticks per tooth during specimen preparation was recorded. Specimens that originated from areas immediately above the pulp chamber had their remaining dentin thickness measured with a digital caliper and recorded (Absolute Digimatic, Mitutoyo, Tokyo, Japan). The cross-sectional area of each stick was measured with a digital caliper to the nearest 0.01 mm for calculation of the actual bond strength values (BS).

Half of the bonded sticks of each tooth were assigned for testing immediately or after six months of water storage in microtensile testing. Each bonded stick was attached to a modified device for microtensile testing with cyanoacrylate resin (Zapit, Dental Ventures of North America, Corona, CA, USA) and subjected to a tensile force in a universal testing machine (EMIC, São José dos Pinhais, PR, Brazil) at a crosshead speed of 0.5 mm/minute. The failure modes were evaluated at 400x (HMV-2, Shimadzu, Tokyo, Japan) and classified as cohesive (failure exclusive within the dentin or resin composite), adhesive (failure at the resin/dentin interface) or adhesive/mixed (failure at the resin/dentin interface that included cohesive failure of the neighboring substrates).

The mean bond strength of all sticks from the same half-tooth was averaged for statistical purposes. The prematurely debonded specimens were included in the tooth mean. The average value attributed to specimens that failed prematurely during preparation was arbitrary and corresponded to approximately half of the minimum bond strength value that could be measured in the current study (ca 4.3 MPa).[29] The BS mean for every testing group was expressed as the average of the five tooth halves used per group. The microtensile bond strength data was subjected to a two-way repeated measures analysis of variance (Air temperature vs Storage time) for each adhesive system and a post-hoc Tukey's test at α=0.05 for pairwise comparisons. The storage time was the repeated measure.

Scanning Electron Microscopy for Silver Nitrate Uptake Evaluation

Two bonded sticks from each tooth half used for each experimental condition were coated with two layers of nail varnish applied to within 1 mm of the bonded interfaces. A total of 10 bonded sticks were analyzed for silver nitrate uptake, as five tooth halves were used per the experimental conditions. The specimens were rehydrated in distilled water for 10 minutes prior to immersion in the tracer solution. Ammoniacal silver nitrate was prepared according to the protocol previously described by Tay and others.[30] The sticks were placed in ammoniacal silver nitrate in darkness for 24 hours, rinsed thoroughly in distilled water and immersed in photo developing solution for eight hours under a fluorescent light to reduce the silver ions into metallic silver grains within voids along the bonded interface.

All the sticks were wet-polished with 600-grit SiC paper to remove the nail varnish. The specimens were polished with 800-, 1000-, 1200-, 1500-, 2000- and 2500-grit SiC paper and 1 µm and 0.25 µm diamond paste (Buehler Ltd, Lake Bluff, IL, USA), using a polishing cloth. They were ultrasonically cleaned, air dried, mounted on aluminum stubs and sputter-coated with carbon only. The resin-dentin interface was analyzed in a scanning electron microscope (Jeol 5800, Tokyo, Japan) operated in the backscattered electron mode. The working distance was 8 mm, with an accelerating voltage of 12 Kv.

Three pictures from each specimen were taken. The first picture was from the center of the stick. The other two pictures were subsequently taken 0.3 mm left and 0.3 mm right of the first one by a technician who was not aware of the experimental conditions under evaluation. Only the most representative picture among the three was chosen per stick. The relative percentage of silver nitrate uptake within the adhesive and hybrid layer was measured using UTHSCSA ImageTool 3.0 software (Department of Dental Diagnostic Science at The University of Texas Health Science Center, San Antonio, TX, USA). The data was submitted to two-way ANOVA and Tukey's test for comparison of the means for each adhesive system (alpha=0.05).

The mean of silver nitrate update of all pictures and sticks from the same tooth was averaged for statistical purposes.

RESULTS

Microtensile Bond Strength Testing

The mean cross-sectional area to specimens for microtensile bond strength ranged from 0.82 to 0.98 mm^2 and no difference among the groups was detected (p>0.05). The percentage of specimens with premature debonding and the frequency of each fracture pattern mode are shown in Table 2. Table 3 depicts the overall

Table 2: *Number of Specimens and Their Respective Percentages (%) Distributed According to the Fracture Pattern Modes As Well As the Percentage of Premature Debonded Specimens for Each Experimental Condition*

Adhesive	Air Temperature	Storage Time	Adhesive/Mixed	Cohesive	Debonded
Single Bond (SB)	Cold	Immediate	33 (73.3)	4 (8.9)	8 (17.8)
		6-month	37 (92.5)	0 (0)	3 (7.5)
	Warm	Immediate	29 (64.4)	7 (15.6)	9 (20)
		6-month	29 (76.3)	0 (0)	9 (23.7)
Prime Bond 2.1 (PB)	Cold	Immediate	28 (68.3)	5 (12.2)	8 (19.5)
		6-month	32 (80)	0 (0)	8 (20)
	Warm	Immediate	25 (73.6)	3 (8.8)	6 (17.6)
		6-month	36 (81.8)	0 (0)	8 (18.2)

Table 3: *Overall Microtensile Bond Strength Values and the Respective Standard Deviations (MPa) Obtained in Each Experimental Condition (*)*

	Air Temperature			
	Cold		Warm	
Adhesive	Immediate	6-month	Immediate	6-month
SB	35.7 ± 7.3 b	28.3 ± 2.6 c	50.1 ± 7.3 a	37.3 ± 3.7 b
PB	34.1 ± 6.2 A,B	24.2 ± 4.2 C	45.8 ± 6.3 A	26.5 ± 3.8 C

(*) Comparisons can be made only within rows. The same lowercase and uppercase letters indicate statistically similar means (p>0.05).

Table 4: *Overall Mean Percentage of Silver Nitrate Uptake Within Hybrid Layer and Adhesive Layer and the Respective Standard Deviations (MPa) Obtained in Each Experimental Condition (*)*

	Air Temperature			
	Cold		Warm	
Adhesive	Immediate	6-month	Immediate	6-month
SB	24.8 ± 5.2 a	25.7 ± 5.3 a	3.7 ± 2.9 b	5.0 ± 2.6 b
PB	19.8 ± 5.5 A	20.8 ± 7.6 A	6.5 ± 2.9 B	8.2 ± 2.9 B

(*) Comparisons can be made only within rows. The same lowercase and uppercase letters indicate statistically similar means (p>0.05).

means and respective standard deviations of the resin-dentin bond strengths (MPa) for all the experimental groups.

For SB, the interaction Air Temperature vs Storage time was not statistically significant ($p>0.05$), while the main factors, Air Temperature and Storage time, were statistically significant ($p=0.001$). Higher bond strength values were observed for SB when the solvent evaporation step was performed with a warm air stream, regardless of the storage period. An approximate 28.7% improvement in bond strengths was observed in the immediate time when warm instead of cold air dry was used. Reductions in bond strength values were observed after six months for both air temperatures; however, the BS mean under the warm condition was higher than the cold ones.

For PB, neither the interaction Air Temperature vs Adhesive nor the main factor Air Temperature was statistically significant ($p>0.05$). Only the main factor Storage time was statistically significant ($p=0.001$). Significant reductions in bond strength values were observed after six months of water storage for both groups (warm and cold group).

Silver Nitrate Uptake

The interaction Air Temperature vs Adhesive and the main factor Time were not statistically significant ($p>0.05$) for both adhesives. Only the main factor Temperature was statistically significant for both adhesive systems tested ($p < 0.001$). Significantly lower silver nitrate uptake within the hybrid and adhesive layer was observed when the solvent was evaporated with a warm air stream instead of cold air. For SB, the warm air-dry stream decreased the silver nitrate staining approximately 85% and 80.5% for the immediate and six-month periods, respectively. For PB, these drops were 67.2% and 60.5% for the immediate and six-month periods (Table 4).

Representative SEM images at the resin-dentin interfaces for the experimental conditions are depicted in Figures 1 and 2. Single Bond, after solvent evaporation with a cold air-dry stream, showed a poor seal, as many dentinal tubules were filled with silver nitrate (Figure 1a). In addition, the entire thickness of the hybrid layer and some spots in the adhesive layer formed under this condition were impregnated with silver nitrate. This situation got worse after six months of water storage (Figure 1b), as more areas of the adhesive layer became impregnated with silver nitrate. Under the warm air-dry condition, this was observed neither immediately (Figure 1c) nor at six-months (Figure 1d).

Similarly, Prime & Bond 2.1 showed a very dense deposition of silver nitrate penetration at the hybrid layer and within the dentin tubules when the solvent was

evaporated with a cold air-dry (Figure 2a). This situation was practically the same after six months of water storage (Figure 2b). However when the cold air-dry conditions were compared to the warm groups at their respective storage times, significantly lower silver nitrate penetration (Figures 2c and 2d) can be observed, with practically no deposition within the dentin tubules.

DISCUSSION

Simultaneous chemical cross-linking and solvent evaporation are the main processes that govern formation and quality of the majority of the bonding coatings. Solvents act as a transport medium and lower resin viscosity in adhesive solutions, allowing for greater penetration of resins into the microporosites of the prepared tooth surface,[31] enhancing the mobility of radicals and growing polymer chains.[32] On the other hand, although solvents seem to be an indispensable ingredient of adhesives systems, they should be removed to a sufficient degree, otherwise residual solvent inhibits polymerization and weakens the mechanical properties of the adhesive resin,[12,33-35] which explains the attempt of the current investigation to employ a warm air dry to improve the solvent evaporation rate.

In fact, the incomplete evaporation of solvents from primer solutions has been blamed for the significant reductions in the ultimate tensile strength of adhesives when the bonding resins are mixed with their respective primer solutions.[34,36] The complete evaporation of solvents is hard to achieve, even by thorough air drying. As water/solvent evaporates from the adhesive, monomer density is found to increase sharply, creating a monomer concentration gradient that acts as a barrier for further solvent evaporation and, thus, reduces the ability of water and solvents to evaporate from the adhesive.[37-38]

This situation is even worse for simplified adhesives, such as the two-step etch-and-rinse adhesives evaluated in the current investigation, since the extent of solvent and water retention in polymer networks seems to be directly correlated with the hydrophilicity of the resin blends,[35] which is higher for these simplified systems. In addition, the recommended clinical time for solvent evaporation is rather short, since some studies have demonstrated that only periods of time longer than 12-20 minutes can ensure adequate solvent evaporation.[25,39]

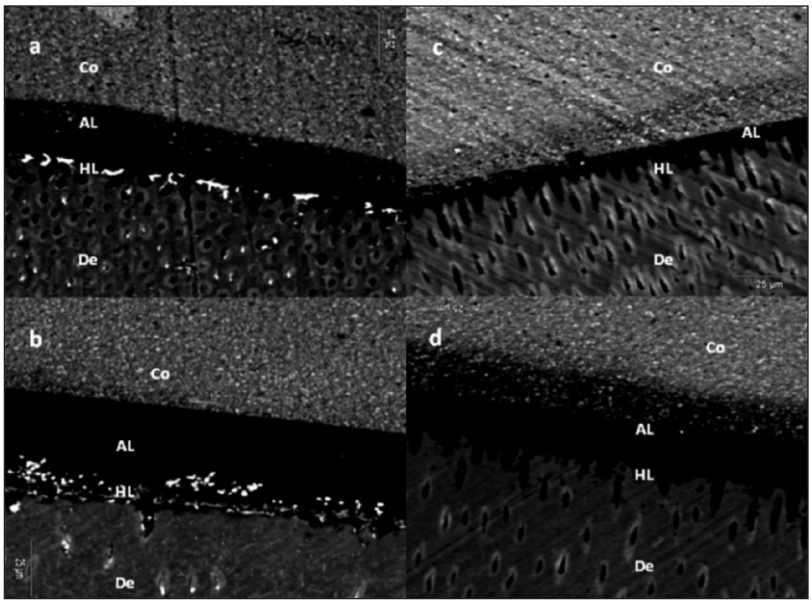

Figure 1. *Representative backscattered SEM images of the resin-dentin interface bonded with Single Bond. In Figures 1a and 1b, the solvent was evaporated with a cold air-dry stream, while in Figures 1c and 1d, a warm air-dry stream was employed. The amount of silver penetration in Figure 1a (cold air-dry stream) was higher in comparison with Figure 1c and 1d (warm air-dry stream immediate and after six months, respectively). It can be seen that, when cold air-dry stream was performed after six months, the amount of silver penetration was higher and occurred within the HL and as part of the AL (Figure 1b) (Co = composite; AL = adhesive layer; HL = hybrid layer and De = dentin).*

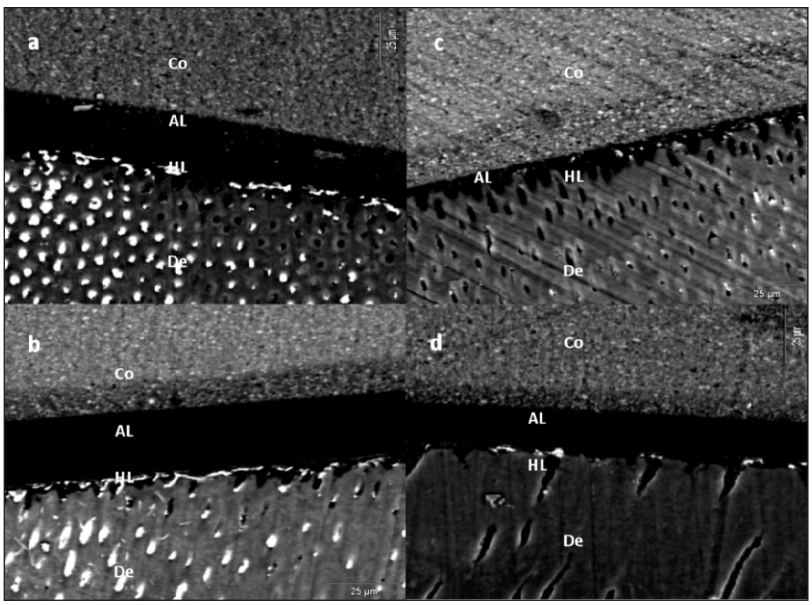

Figure 2. *Representative backscattered SEM images of the resin-dentin interface bonded with Prime & Bond 2.1. In Figures 2a and 2b, the solvent was evaporated with a cold air-dry stream, while in Figures 2c and 2d, a warm air-dry stream was employed. The amount of silver penetration in Figure 2a (cold air-dry stream) was higher, compared with Figures 2c and 2d (warm air-dry stream immediate and after six months, respectively). It can be seen that, when a cold air-dry stream was performed after six months, the amount of silver penetration was higher and occurred within the HL (Figure 2b) (Co = composite; AL = adhesive layer; HL = hybrid layer and De = dentin).*

A previous study that evaluated resin-dentin bond strength only in immediate time demonstrated that one way to improve solvent evaporation without increasing the solvent evaporation time was to use a warm air-dry stream at approximately 60°C.[28] These findings were confirmed in the current investigation for the ethanol/water-based system. Higher immediate bond strength means were obtained when the warm air stream was applied instead of the cold air-dry. When heat is delivered to a substance, it may cause an increase in the kinetic energy of the molecules. This leads to a temperature increase or a change in the substance state,[40] which is basically dependent on the substance and temperature. By observing the resin-dentin bond strength results, one can notice that SB responded better to the warm air-dry application than PB, even after six-months of water storage. This could be attributed to differences in the solvents presented in both systems. SB is an ethanol-based system, while PB is an acetone-based system. The vapor pressure and boiling temperature of ethanol is respectively lower and higher than acetone, thus, heat from the warm air-stream might have improved the evaporation rate of ethanol from the SB system but had a less pronounced effect in terms of evaporation rate for acetone presented in the PB system.[28]

Contrary to the six-month bond strength findings, the silver nitrate uptake evaluation showed that the warm groups performed somewhat better than the cold groups for both adhesives. This could theoretically mean that a better sealing of the cavity can be achieved with this technique. One could not attribute the good performance of the warm air-dry groups to a higher degree of conversion of the polymer network inside the hybrid layer, since a previous study demonstrated that use of a warm or cold air-stream did not affect this adhesive property.[28]

The percentage of silver nitrate penetration was significantly higher in the specimens that were cold air-dried. This situation got worse after six months of water storage and was likely due to the presence of solvent-rich pores and highly hydrophilic domains inside the adhesive and hybrid layer, as can be seen in the SEM findings of the current study. Bond degradation is known to be preceded by water sorption in a polymer matrix. Water sorption is enhanced by the presence of hydrophilic and ionic resin monomers,[41-42] which, in turn, facilitate ion movement within a polymerized resin matrix.[42-43]

This water movement causes hydrolytic degradation and likely leaching of the hydrolyzed components of porous dentin adhesive coatings with an associated increase in permeability, thus creating a vicious cycle that increases deterioration of the mechanical properties of the adhesive coatings.[44] If solvent is not adequately removed, it represents areas of incomplete polymerization and/or hydrogel formation within the adhesive layer,[30,44] similar to what could have occurred in the cold air-dried groups. Thus, it serves as immediate channels within the bonding interface, which may speed up the transport of extrinsic water. These sites work as one of the sources of silver nitrate uptake expression within adhesive interfaces and are highly prone to the deposition of silver nitrate, as can be seen in the micrographs of the current investigation after six months.

Based on these findings, the good results attained with the groups that were air dried with a warm air-dry could be attributed to the removal of these solvent channels. This could have allowed for close contact of the polymer chains by reducing the spaces that would work as sites for the accumulation of water. The nature of the adhesive (hydrophilicity) was not altered by use of a warm air-dry stream. However, the authors of the current study might presume that the clinical approach used (warm air for solvent evaporation) could have altered the polymer network chain topology/morphology, thus reducing the intrinsic fraction of nanopores within the polymer network.[45]

CONCLUSIONS

Based on the results of the current investigation, when assessing silver nitrate uptake, the authors can consider that the use of a warm air-stream may improve the bonding interface over time, mainly for the ethanol/water-based system.

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