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Original Article

The effect of acrylate-based dental adhesive solvent content on microleakage in composite restorations

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ABSTRACT

Background: This study aimed to evaluate the effect of different percentages of ethanol solvent of an experimental methacrylate-based dentin bonding agent containing polyhedral oligomeric silsesquioxanes (POSS) on the microleakage of resin composite restorations.
Materials and Methods: In this experimental study, 42 extracted human premolar teeth used and 84 standard Class V cavities were prepared on the buccal and lingual surfaces of the teeth. The teeth were divided into 6 groups of 7. Experimental bonding agents with different percentages of solvent were used in 5 groups and Single Bond® as a control. The teeth were restored with resin composite and subjected to thermal cycling test. Teeth were then immersed in a solution of 2% basic fuchsine dye for 24 h and sectioned buccolingually and scored using stereomicroscope with ×32 magnification. Microleakage data were analyzed using the Kruskal–Wallis, Mann–Whitney U, and Wilcoxon tests.
Results: There were significant differences between the microleakage enamel margins ($P = 0.036$) and dentinal margins ($P = 0.008$) in all the groups. These significant differences were seen between the control group and groups containing 46 wt% solvent ($P = 0.011$), 46 wt% and 31 wt% solvent in dentinal ($P = 0.027$), 31 wt% and 0 wt% in enamel ($P = 0.021$), also 0 wt% and control in enamel ($P = 0.039$), and dentinal margins microleakage ($P = 0.004$). The microleakage in dentinal margins was higher than enamel margins ($P < 0.001$). In the groups with 46 wt% solvent ($P = 0.103$), 0 wt% ($P = 0.122$), and control group ($P = 0.096$), however, this difference was not significant.
Conclusion: The adhesive containing 31 wt% solvent showed the least marginal microleakage, presence of POSS filler may also result in the reduction of microleakage.

Key Words: Bonding agent, leakage, micro, polyhedral, oligomeric silsesquioxanes methacrylate

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INTRODUCTION

Microleakage is one of the most important reasons for the secondary decay, inflammation, and necrosis of the pulp.[1] In resin composite restorations, microleakage occurs between cavity wall and restorative materials usually due to the polymerization shrinkage. Marginal microleakage can be prevented with a good bond to the tooth structure.[2] Composition of the bonding agents is one of the most important prerequisites to ensure a

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good seal and stable adhesion to the tooth tissue.[3] Main components of bonding agents include resin monomers and solvent. The most common solvents recently used are acetone, ethanol, and water.[4,5] The purpose of adding a solvent to the dental bonding agent is to dilute it to create a thin layer on the surface of etched tooth, and displacement of water from the collagenous network and penetration of resin monomers into these spaces.[6-8] Although the presence of the solvent in bonding agent is necessary, the entire solvent must be completely eliminated from bonding before curing the bonding agent because its presence can have adverse effects on polymerization of the resin monomers.[9,10] When the solvent's concentration is too high, reduced photoinitiator concentration and occupation of spaces between the monomer molecules will cause a reduction in the conversion of monomers to polymers and reduces the mechanical properties and strength of the bonding.[10-14] Therefore, after bonding resin applications, the solvent is removed from the resin using air-stream.[15] Acrylate in dental adhesives makes an important contribution in polymerization and creating an appropriate bonding.[16]

On the other hand, polyhedral oligomeric silsesquioxanes (POSS) are nanostructural blocks that have recently been used in dental materials.[17,18] POSS as cross-linking agents in bioadhesive acrylate affect the contractile behavior and improve their physical and mechanical properties.[19,20]

Wang *et al.*,[4] observing the hybrid layer with scanning electron microscope, reported that among the different concentrations of ethanol as solvent (10%, 30%, and 50%), the concentration of 30 wt% was the optimum percentage of the solvent and concentrations of less than 10 wt% and over 50 wt% resulted in poor quality hybrid layer.

Holmes *et al.*[9] reported that in a resin matrix which prepared with six different concentrations of acetone and ethanol, the highest conversions of monomer to polymer were observed in acetone and ethanol with concentrations of 5 mol and 2.5 mol, respectively. However, in higher concentrations of the solvents, the polymer conversion rate was reduced quickly so that at 13 mol approached zero.

As the solvent concentration plays an important role in the efficiency of dentin binding agents, the present study was designed to evaluate the effect of ethanol concentration as a solvent in experimental acrylate-based dental adhesives containing POSS, on the microleakage of dental composite restoration.

MATERIALS AND METHODS

In this experimental study, 42 extracted sound noncarious human premolars without restorations, abrasion, and cracks were selected and stored for 2 months in 0.2% thymol solution before performing the study. Standard Class V cavities (H × W × D = 3 × 4 × 2) were prepared with 0.8 mm cylindrical diamond burs using a high-speed handpiece and water coolant in the buccal and lingual sides with the occlusal margin in enamel and the cervical margin in dentin. The teeth were randomly divided into 6 experimental groups of 7. In all groups, teeth were acid etched by 35% phosphoric acid gel (Ultradent, USA) for 15 s, rinsed for 15 s and blot-dried for 2 s. In group 1, as the control group, Adper Single Bond®, 3M, USA was used The adhesives of all the test groups contained Di and trimethacrylate monomers, POSS, and photoinitiator system but the concentration of ethanol, as the solvent, in groups 2 through 6 varied and was 0%, 20%, 31%, 39%, and 46% by weight, respectively. The adhesives were prepared in Iran Polymer and Petrochemical Institute, Tehran, Iran. The adhesives were applied and light cured for 20 s at an intensity of 600 mW/cm^2 (Litex 695C, Taiwan). Cavities in all control an test groups were restored with Filtek Z350 nanocomposite (3M ESPE, USA) in 3 increments. Each increment of composite resin was cured for 40 s. Finishing and polishing were done with flamed shaped polishing bur and disks (Soflex, 3M ESPE, USA). All samples were thermocycled between 5°C and 55°C for 1000 cycles. Then, wax was used to seal apics of all specimens. All the surfaces of teeth up to 1 mm around the restorations were covered with two layers of nail varnish. Then, specimens were immersed in 2% basic fuchsine (Merck, Germany) solution at room temperature for 24 h. The teeth were then rinsed thoroughly with distilled water. CNC cutting section machine (Fanavaran Pars, Iran) was used to section the specimens in halves through the middle of the restoration. Dye penetration in each section was scored under stereomicroscope (MBC-10, Hp, USA) with ×32 magnification.

The following dye penetration standard scoring[21] was used:

0 = No microleakage
1 = Penetration up to one-third of the cavity depth
2 = Penetration up to two-third of the cavity depth
3 = Penetration up to more than two-third of the cavity depth
4 = Penetration up to axial wall or toward the pulp.

The results of microleakage were analyzed with Kruskal–Wallis, Mann–Whitney U-test, and Wilcoxon tests while significance was predetermined at $P < 0.05$.

RESULTS

Table 1 shows the number and percentage of microleakage scores at enamel and dentinal margins.

Illustrates the representative microscopic images showing the scores of microleakage, taken under a stereomicroscope (at a ×32 magnification).

According to Kruskal–Wallis test, there were significant differences in microleakage in enamel margins ($P = 0.036$) and in dentinal margins ($P = 0.008$) between all the tested groups. According to Wilcoxon test, microleakage in dentinal margins was higher than enamel margins ($P < 0.001$). In groups with 46 wt% solvent ($P = 0.103$), 0 wt% ($P = 0.122$), and control group ($P = 0.096$), this difference was not significant.

Microleakage results in the enamel and dentinal margins for the study groups are shown in Table 2.

DISCUSSION

In the present study, an acrylate-based experimental bonding agent with varied solvent concentrations was used. The experimental bonding agent also contained POSS nanostructures to improve the mechanical properties of the adhesive. The rate of resin composite microleakage was checked out in dentinal and enamel margins.

In the study, ethanol solvent in experimental groups was the same as the solvent applied in the control group. The reason for using ethanol is the benefits of this solvent. Because of the higher boiling point of ethanol than acetone and establishment of hydrogen bonds with water remaining in dentin, its volatility is less, but still has more evaporation power than water.[22-24]

Abate *et al.*[25] have shown that the bonding agents containing acetone will lose the solvent rapidly, but bonding agents containing water are almost stable. Due to the rapid evaporation of acetone and lack of ability to penetrate the dry collagen network of dentin, the dentin should have sufficient moisture before using the bonding agents containing acetone. While ethanol is less sensitive to moisture and this will make it easier to use.[9,26] Furthermore, bonding agents containing water, due to a low evaporation rate of water leads to a nonuniform hybrid layer and the remaining water prevents resin monomer polymerization, so the bond strength will be decreased. Therefore, bonding agents with ethanol compared to acetone needs more time to dry and there is a possibility of phase separation. Thus, they should be well shaken before use.[26]

In this study, microleakage was significantly higher, in both enamel and dentinal margins, in group containing 0 wt% solvent, compared to the control group. It is attributed to the high resin viscosity in the 0% group and its lack of penetration into enamel porosities and intertubular dentin spaces.[27]

By increasing the solvent concentration, the microleakage decreased so that the leakage in enamel margins of the group containing 31 wt% solvent reduced significantly compared to the group with 0 wt% solvent and the lowest leakage was observed in this group (even less than the control group). In dentinal margins by increasing solvent, microleakage decreased and the bonding with 31 wt% solvent assigned the lowest microleakage between the

Table 1: Number and percentage of scores at enamel and dentinal margins

Variable studied	Score	Solvent percentage					Control (%)	Total (%)
		46	39	31	20	0		
Enamel margin	0	6 (42.9)	8 (57.1)	12 (85.7)	8 (57.1)	5 (35.7)	10 (76.9)	50 (59.5)
	1	5 (35.7)	6 (42.9)	1 (7.1)	5 (35.7)	5 (35.7)	2 (35.7)	24 (15.4)
	2	1 (7.1)	0	1 (7.1)	1 (7.1)	1 (7.1)	1 (7.7)	5 (6)
	3	2 (14.3)	0	0	0	2 (14.3)	0	4 (4.8)
	4	0	0	0	0	1 (7.1)	0	1 (1.2)
Dentinal margin	0	1 (7.1)	2 (14.3)	5 (35.7)	3 (21.4)	1 (7.1)	7 (50)	19 (22.6)
	1	6 (42.9	7 (50)	7 (50)	5 (35.7)	5 (35.7	5 (35.7)	35 (41.7)
	2	3 (21.4)	4 (28.6)	1 (7.1)	3 (21.4)	2 (14.3)	1 (7.1)	14 (16.7)
	3	2 (14.3)	1 (7.1)	1 (7.1)	2 (14.3)	3 (21.4)	1 (7.1)	15 (11.9)
	4	2 (14.3)	0	0	1 (71)	3 (21.4)	0	6 (7.1)

Table 2: Mean rank of microleakage in the enamel and dentinal margins for the study groups

Variable studied	Group (%)	*n*	Mean rank	χ^2	df	*P*
Enamel margin microleakage	Control	14	34.46	11.923	5	0.036
	0	14	54.57			
	20	14	42.39			
	31	14	31.82			
	39	14	41.36			
	46	14	50.39			
	Total	84				
Dentinal margin microleakage	Control	14	27.86	15.469	5	0.008
	0	14	55.93			
	20	14	44.86			
	31	14	31.71			
	39	14	42.75			
	46	14	51.89			
	Total	84				

experimental bonding groups. In enamel and dentinal margins, further increase in the solvent up to 46 wt%, the microleakage increased again. At the higher solvent concentrations, the solvent occupies the space between the monomers and will reduce the degree of conversion, mechanical properties of the polymer, and resin-dentin bond strength.[9,28]

The results of this study are in agreement with findings in the other studies stating that the solvent facilitates the penetration of resin in collagenous matrix by reducing the bonding agent viscosity. Wang *et al.*[4] studied the effect of different concentrations of ethanol on the quality of the hybrid layer and concluded that among the 10%, 30%, and 50% concentrations of solvent, the concentration of 30% was the best. In bonding agents with 10% ethanol, the bonding agent monomers are not able to penetrate into dentin because of higher viscosity of the adhesive. When the solvent concentration is 30%, more resin penetration occurred, whereas at the higher solvent contents 50%, penetration of the monomers decreases again greatly due to the high dilution bonding agent or severe dehydration which causes collapse of nanochannels between collagen fibrils. The concentration of 50% was not perfect due to the intense evaporation of the solvent and leaving a porous hybrid layer. If the concentration of the solvent becomes more than adequate, it will prevent polymerization of adhesive monomers, and mechanical properties of adhesives are reduced.[9]

Holmes *et al.*[9] studied a mixture of acetone and ethanol with six different concentrations. By adding solvents, the rate of monomer conversion to polymer increased compared to solvent-free resin, and the highest rate of polymerization was in 5 mol concentration of acetone and in ethanol, the maximum monomer conversion was observed in 2.5 mol concentrations. Increasing the concentration of solvent (for ethanol more than 2.5 and for acetone more than 5 M) led to a decrease of degree of conversion so that in concentration of 13, it reached the zero. By increasing the thickness of the bonding agent, the solvent trapped and prevented complete polymerization of adhesive and reduced the bond strength. Therefore, in clinical work using air-stream is necessary to prevent the accumulation of the solvent of the bonding agents.[29]

In a study by Aw *et al.*,[30] however, three different bonding agents, containing ethanol and water as solvent and one without solvent, were applied on teeth by the integration of other variables, and after following the study for 1 year, they found that the restorations were the same in terms of retention, marginal integrity, marginal discoloration, and post-treatment sensitivity. They believed that the solvent-free adhesive by creating thicker layer makes better thermal protection and reduces shear stresses, but if the thickness of layers becomes too high, may lead to the weakening of the bond strength.

Ye *et al.*[31] believe that solvent concentrations greater than 20 wt% reduce the degree of conversion by increasing the physical space between reactive species during polymerization.

It has been shown by Cadenaro *et al.*[32] that increasing the ethanol concentration up to 10 wt% and 20 wt% led to increased DC, but by adding 30 wt% during 1st s of light exposure lowered DC and then increased DC after 40 s exposure. The residual solvent results in higher degree of conversion through the decrease in the viscosity of the monomer blend which allows the macroradicals to propagate before the polymerization reaction being diffusion controlled.

In the present study, the POSS nanostructures were added to experimental dental adhesive. POSS particles contain reactive methacrylate functional groups which take part in the copolymerization with the resin monomers increasing the cross-link density and consequently the mechanical properties of the adhesive layer. The nanostructures can also enhance the physical, chemical, and thermal properties of bonding agent and therefore impact on the leakage.[33,34]

Mousavinasab *et al.*,[35] by comparing the amount of leakage between experimental adhesives containing nanoclay fillers, after 24 h and 6 months in Class V restorations, concluded that these adhesives reduced enamel leakage in short term than control group (Adper Single Bond). However, no success was observed in dentinal microleakage in the short-term (24 h) and long-term (6 months). The cause of more microleakage in these adhesives was attributed to its high concentration of 2-hydroxyethyl methacrylate.

Sadat-Shojai *et al.*,[36] by adding nano-hydroxyapatite fillers to experimental dental acrylic-base bonding and 40 wt% ethanol, concluded that the incorporation of 0.2%–0.5% by weight of the nano-hydroxyapatite fillers, enhanced physical properties compared to the group without filler.

Fadaie *et al.*[20] showed the effect of POSS nanoparticles on improving physical and mechanical properties and hydrolytic stability of cyanoacrylate adhesives. A reduction in the rate of solubility and water absorption was observed using POSS nanostructures in the cyanoacrylate adhesives.

CONCLUSION

According to the results, it can be concluded that in the bonding agents, an optimum percentage of solvent is required to have a minimum microleakage. The optimum solvent concentration was shown to be 31 wt% in the studied experimental bonding agent which led to the reduction in marginal microleakage. Furthermore, the presence of POSS fillers may also result in a reduction in microleakage due to the lower hydrophilicity of the nanostructures and the higher cross-link density of the adhesive in the presence of the multi-acrylate POSS.

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Conflicts of interest
The authors of this manuscript declare that they have no conflicts of interest, real or perceived, financial or non-financial in this article.

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