Influence of light intensity on surface-free energy and dentin bond strength of single-step self-etch adhesives

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In this study, we investigated the influence of light intensity on the surface-free energy and dentin bond strength of single-step self-etch adhesives. The adhesives were applied to the dentin surfaces of bovine mandibular incisors and cured with light intensities of 0 (no irradiation), 200, 400, and 600 mW/cm². Surface-free energies were determined by measuring the contact angles of three test liquids placed on the cured adhesives. Dentin bond strengths of the specimens were also measured. Polymerization with a higher light intensity resulted in a lower surface-free energy of the cured adhesives. The greatest bond strength was achieved when a light intensity of 400 mW/cm² or greater was used. Our data suggest that the surface-free energy and dentin bond strength of single-step self-etch adhesives are affected by light intensity of the curing unit.

Keywords: Light intensity, Surface-free energy, Dentin bond strength, Single-step self-etch adhesive

INTRODUCTION

Current dental adhesive systems feature simplified application methods to achieve optimum clinical results¹⁾. Single-step self-etch adhesives that combine the functions of a self-etching primer and a bonding agent have been developed²⁾. This kind of adhesive system combines acidic functional monomers, hydrophilic monomers, hydrophobic monomers, fillers, water, and organic solvent in a single bottle³⁾. The use of water as a solvent is indispensable for single-step self-etch adhesives to ensure the ionization of the acidic functional monomers, and the organic solvents are added to facilitate mixing of the hydrophilic and hydrophobic components⁴⁾. However, it has been suggested that water and the solvents mixed in the adhesive resin lead to reduced mechanical properties and accordingly poorer bonding performance⁵⁾. Therefore, removing these components from single-step self-etch adhesives is important for optimal bonding.

Single-step self-etch adhesives reportedly have a high hydrophilicity even after polymerization, which may increase their solubility and water uptake compared with those of conventional multi-step adhesives⁶⁾. Long-term leaching of water into the material may further compromise its mechanical properties, leading to poor bonding performance of single-step self-etch adhesives to dentin⁷⁾. These phenomena may be dependent upon the degree of conversion of the adhesive, which is strongly influenced by light intensity during curing.

Light intensity during curing is dependent upon the clinical circumstances, such as cavity preparation, tooth position, or the existence of adjacent teeth⁸⁾. When the distance between the curing light tip and cavity wall increases, light intensity reaching the adhesive surface decreases⁹⁾. It has been demonstrated that reduced light intensity causes lower bond strengths between self-etch adhesives and dentin¹⁰⁾. In addition,

after the light irradiation of adhesives at a reduced light intensity, the top surface of the adhesive is covered with a relatively thick, uncured adhesive layer due to the oxygen inhibition¹¹⁾. In the case of single-step self-etch adhesives, residual water and acidic functional monomers may be present within this oxygen-inhibited layer¹²⁾. The presence of water and acidic functional monomers may compromise the bonding durability of single-step self-etch adhesives. Therefore, the bonding performance of single-step self-etch adhesives may be influenced by light intensity during curing to a greater degree than the conventional multi-step adhesive systems with solvent-free bonding agents. However, there is little information on the mechanism by which the light irradiation of single-step self-etch adhesives affects dentin bond strength; therefore, it is necessary to determine the influence of light intensity on the bonding performance of these adhesives.

Previous studies^{13,14)} have shown that the interfacial characteristics of self-etch adhesives can be analyzed on the basis of the surface-free energy measurement. Generally, the surface-free energy characteristics of polymers make a significant contribution to their interfacial interactions with liquids, with which they can form hydrogen bonds. Wetting properties and the hydrophilicity of adhesives may be expressed in terms of the surface-free energy and its components. Thus, analysis of the surface-free energy of single-step self-etch adhesives may provide an insight in the development of their bonding performance.

In this study, we examined the influence of light intensity during curing on the surface-free energy of adhesive-treated dentin and bond strength of single-step self-etch adhesives to bovine dentin. Our null hypothesis was that there would be no significant differences in surface-free energy or bond strength at different light intensities.

MATERIALS AND METHODS

Adhesives used

As presented in Table 1, the single-step self-etch adhesive systems used were: Scotchbond™ Universal Adhesive (3M ESPE, St. Paul, MN, USA); Clearfil™ Tri-S Bond (Kuraray Noritake Dental, Tokyo, Japan); and G-Bond™ Plus (GC, Tokyo, Japan).

A visible-light curing unit (Optilux 501, Demetron Kerr, Danbury, CT, USA) was connected to a variable-voltage transformer (Type S-130-10, Yamabishi Electric, Tokyo, Japan). The light intensities used were 100, 200, 400, and 600 mW/cm²; these values were determined using a dental curing radiometer (Model 100, Demetron Kerr). The curing unit was placed in a jig to maintain the distance between the light tip end and specimen surface (2 mm).

Surface-free energy measurement

Mandibular incisors were extracted from 2- to 3-year-old cattle and frozen until use. After removing the roots of each tooth with a slow-speed saw equipped with a diamond-impregnated disk (IsoMet®, Buehler, Lake Bluff, IL, USA), the pulp was removed, and the pulp chamber was filled with cotton to avoid penetration of the embedding medium. After ultrasonic cleaning for 1 min in distilled water to remove debris, the surface of each tooth was washed and then dried with oil-free compressed air. The labial surface was ground with wet 240-grit silicon carbide (SiC) paper to create a flat dentin surface. Each tooth was then mounted in self-curing acrylic resin (Tray Resin II, Shofu, Kyoto, Japan)

to expose the flattened area, and placed under tap water to reduce the temperature increase caused by the exothermic polymerization reaction of the acrylic resin. The final finish was created by grinding with wet 320-grit SiC paper. Finally, the dentin surface of each tooth was dried with oil-free compressed air.

Each adhesive was applied to the dentin surface according to the manufacturers' instructions, followed by light irradiation for 10 s. Light irradiation was performed at intensities of 0 (no irradiation), 200, 400, and 600 mW/cm². Surface-free energy was determined by measuring the contact angle with the surface of three test liquids, 1-bromonaphthalene, diiodomethane, and distilled water, each of which has known surface-free energy parameters (Table 2). The surface-free energy of five specimens per light intensity group for each adhesive-treated dentin surface was determined by measuring their contact angle (Drop Master DM500, Kyowa Interface Science, Saitama, Japan). The device was fitted with a charge-coupled device camera, which allowed automatic measurement of the contact angle (Fig. 1). For each test liquid, the equilibrium contact angle (θ) was measured, using the sessile-drop method at 23±1°C, for five specimens of each adhesive¹⁵. The surface free-energy parameters of the solids were then determined based on the fundamental concepts of wetting.

The Young-Dupré equation describes the work of adhesion (W) between a solid (S) and a liquid (L) in contact as follows:

Table 1 Single-step self-etch adhesives used in this study

Adhesive	Code	Lot No.	Main components	Manufacturer
Scotch Bond™ Universal	SU	541424	MDP, Bis-GMA, HEMA, Vitrebond copolymer, polyethylene glycol, water, initiator, colloidal silica, aluminium oxide	3M ESPE, St. Paul, MN, USA
Clearfil™ Tri-S Bond	TS	00038B	MDP, Bis-GMA, HEMA, ethanol, water, initiator, silanated colloidal silica	Kuraray Noritake Dental, Tokyo, Japan
G-Bond™ Plus	GB	1312131	MDP, 4-MET, methacrylate monomer, acetone, water, initiator, silica filler	GC, Tokyo, Japan

MDP: 10-methacryloyloxydecyl di-hydrogen phosphate, Bis-GMA: 2,2-bis[4-(2-hydroxyl-3-methacryloyloxypropoxy)phenyl]propane, HEMA: 2-hydroxyethyl methacrylate, 4-MET: 4-methacryloyloxyethyl trimellitate

Table 2 Surface free energy and their components values of test

Liquid	Lot No.	Manufacturer	γ_L	γ_L^d	γ_L^p	γ_L^h
1-Bromonaphthalene	ALH4513	Wako Pure Chemical Industries	44.6	44.4	0.2	0.0
Diiodomethane	ALL2310	Wako Pure Chemical Industries	50.8	46.8	4.0	0.0
Distilled water	—	—	72.8	29.1	1.3	42.4

Unit: mN/m.

γ_L : surface free energy, γ_L^d : dispersion force, γ_L^p : polarity force, γ_L^h : hydrogen bonding force

Fig. 1 Drop Matser DM500 apparatus fitted with a charge-coupled device camera (a) allowing automatic measurements of the contact angles to be made (b).

$$W_{SL} = \gamma_L + \gamma_S - \gamma_{SL} = \gamma_L (1 + \cos\theta)$$

Here, γ_{SL} is the interfacial free energy between the solid and liquid, γ_L is the surface-free energy of the liquid, and γ_S is the surface-free energy of the solid. By extending the Fowkes equation, the γ_{SL} can be expressed as follows:

$$\gamma_{SL} = \gamma_L + \gamma_S - 2(\gamma_L^d \gamma_S^d)^{1/2} - 2(\gamma_L^p \gamma_S^p)^{1/2} - 2(\gamma_L^h \gamma_S^h)^{1/2}$$

$$\gamma_L = \gamma_L^d + \gamma_L^p + \gamma_L^h, \quad \gamma_S = \gamma_S^d + \gamma_S^p + \gamma_S^h$$

where γ_L^d , γ_L^p , and γ_L^h are components of the surface-free energy (γ) arising from the dispersion force, the polar (permanent and induced) force, and the hydrogen-bonding force, respectively. Surface-free energy (γ) values were determined for the three test liquids, and the surface energy parameters of the treated dentin surfaces were calculated based on these equations using add-on software and an interface measurement and analysis system (FAMAS, Kyowa Interface Science).

Bond strength measurement

The shear-bond strength between the single-step self-etch adhesives and the dentin surface was measured by a notched-edge shear-bond strength test, as described by ISO 29022¹⁶⁾. A total of 120 mandibular incisors from cattle were prepared as described in the preceding section. The adhesives were applied according to the manufacturers' instructions, followed by light irradiation for 10 s. Light irradiation was performed at intensities of 0 (no irradiation), 200, 400, and 600 mW/cm². The specimens were clamped in a bonding jig (Ultradent Bonding Jig, Ultradent Products, South Jordan, UT, USA), and plastic molds (2.4 mm in internal diameter, 2.5 mm in height) were used to form and hold the resin composite on the adhesive-treated dentin surface. The resin composite (Clearfil™ AP-X, Kuraray Noritake Dental) was condensed into the mold and light cured for 30 s at a light intensity of 600 mW/cm². The finished specimens were transferred to distilled water, and stored at 37°C for 24 h.

Ten specimens per light intensity were tested in shear mode using bond-testing notched-blade semi-circular apparatus (Ultradent Products) in a universal testing machine (Type 5500R, Instron, Norwood, MA, USA) at a crosshead speed of 1 mm/min. Bond strength values (MPa) were calculated from the peak load at failure divided by the specimen surface area. After testing, the specimens were examined under an optical microscope (SZH-131, Olympus, Tokyo, Japan) at a magnification of $\times 10$ to determine the location of bond failure. The type of failure was classified based on the percentage of substrate-free material as follows: adhesive failure; cohesive failure in the dentin; or cohesive failure in the composite.

Statistical analysis

The dentin bond strength data obtained were analyzed using a commercial statistical software package (SigmaStat Version 3.1, SPSS, Chicago, IL, USA). Because the data were normally distributed (Kolmogorov-Smirnov test), two-way analysis of variance (ANOVA) was used to analyze the effects of light intensities and the adhesive systems that were used. Multiple comparisons were then conducted using the Tukey-Kramer test, with a significance level of 0.05.

Scanning electron microscopy

The resin-dentin interface was ultrastructurally observed by scanning electron microscopy (SEM). Bonded specimens from each group ($n=5$) were stored in distilled water maintained at 37°C for 24 h, embedded in self-curing epoxy resin (Epon812, Nisshin EM, Tokyo, Japan), and stored at 37°C for an additional 12 h. The embedded specimens were sectioned through the diameter of the composite resin post, and the surfaces of the cut halves were polished with an Ecomet 4/Automet 2 (Buehler) using SiC papers with a grit size of 600, 1,200, and 4,000 in succession. The surface was finally polished by a soft cloth using diamond paste (Buehler) with a grit size of 1.0 μ m. All SEM specimens were dehydrated in ascending concentrations of tert-butanol

(50% for 20 min, 75% for 20 min, 95% for 20 min, and 100% for 2 h) and transferred to a critical-point dryer for 30 min. These surfaces were subjected to Argon ion-beam etching (Type EIS-200ER, Elionix, Tokyo, Japan) for 30 s, with the ion beam (accelerating voltage, 1.0 kV, ion current density, 0.4 mA/cm²) directed perpendicularly to the polished surface. Surfaces were coated in a vacuum evaporator (Quick Coater Type SC-701, Sanyu Denshi, Tokyo, Japan) with a thin film of gold and observed by SEM (ERA 8800FE, Elionix) at an accelerating voltage of 10 kV. All of the specimens were observed under SEM.

RESULTS

The surface free energies and their components, for each cured adhesive, are shown in Table 3. Total surface-free energy ($\gamma_s = \gamma_s^d + \gamma_s^p + \gamma_s^h$) values decreased with increasing light intensity, and were significantly lower when adhesives were irradiated with a light intensity of 400 mW/cm². For all surfaces, the estimated γ_s^d values

remained relatively constant, in the range of 38.3–39.8 mN/m. Decreased γ_s^p and γ_s^h values were observed for specimens irradiated with a greater light intensity.

The influence of light intensity during curing on shear bond strength is shown in Table 4. Two-way ANOVA revealed that both adhesive type ($p < 0.001$) and light intensity ($p < 0.001$) significantly influenced bond strength, although there was no significant interaction between the two factors ($p = 0.601$). For each adhesive, greater bond strength was achieved by irradiation with a light intensity of 400 mW/cm² or greater. Statistically significant differences were evident between the values obtained, except for those obtained at light intensities of 400 and 600 mW/cm² ($p < 0.005$). Failure type was not associated with bond strength: the predominant failure type was adhesive failure.

SEM observations of the resin-dentin interface are shown in Fig. 2. The dentin-resin interface of each groups showed excellent adaptation, with the formation of a transitional layer between the adhesive resin and

Table 3 Influence of light intensity on surface free energies and their components for single-step self-etch adhesives

		γ_s	γ_s^d	γ_s^p	γ_s^h
SU	0	73.5 (3.2)	38.5 (0.8)	10.6 (2.0)	24.4 (2.3)
	200	68.0 (3.1)	39.1 (0.7)	8.3 (2.0)	20.6 (2.2)
	400	55.1 (3.4)	39.6 (1.0)	5.3 (2.7)	10.2 (1.8)
	600	54.8 (2.5)	39.8 (0.8)	4.9 (2.0)	10.1 (1.2)
TS	0	72.2 (3.1)	38.9 (0.6)	9.8 (2.2)	23.5 (2.1)
	200	67.7 (3.6)	38.7 (1.2)	9.6 (2.8)	19.4 (1.9)
	400	57.2 (3.5)	38.9 (1.1)	7.7 (2.7)	10.6 (1.9)
	600	57.1 (3.0)	39.1 (0.9)	6.8 (2.2)	11.2 (1.8)
GB	0	69.0 (3.3)	38.5 (1.2)	10.2 (2.2)	20.3 (2.2)
	200	64.3 (3.2)	38.3 (0.8)	9.3 (2.2)	16.7 (2.2)
	400	57.2 (2.4)	39.8 (0.9)	5.4 (1.5)	12.0 (1.7)
	600	57.6 (2.0)	39.9 (1.2)	5.1 (1.9)	12.6 (2.0)

Unit: mN/m, values in parenthesis are standard deviations ($n=5$).

Values connected by vertical lines indicate no significant difference ($p > 0.05$).

Table 4 Influence of light intensity on dentin bond strength of single-step self-etch adhesives

Adhesive	Light intensity			
	0	200	400	600
SU	18.8 (3.2) ^{a,A} [10/0/0]	24.8 (3.7) ^{a,B} [10/0/0]	30.3 (3.7) ^{a,C} [10/0/0]	29.2 (3.2) ^{a,C} [10/0/0]
TS	15.5 (2.8) ^{b,A} [10/0/0]	19.2 (4.4) ^{b,B} [10/0/0]	27.9 (3.8) ^{b,C} [10/0/0]	27.3 (3.2) ^{b,C} [10/0/0]
GB	15.3 (3.5) ^{b,A} [10/0/0]	19.0 (3.4) ^{b,B} [10/0/0]	25.3 (3.9) ^{b,C} [10/0/0]	26.0 (3.0) ^{b,C} [10/0/0]

Unit: MPa, values in parenthesis are standard deviations ($n=15$).

Values with same letter indicate no significant difference ($p > 0.05$).

[] : failure mode [adhesive failure/ cohesive failure/ mixed failure]

Fig. 2 Representative SEM photomicrographs of the dentin-resin interface (Original magnification, $\times 5,000$). Thickness of adhesive was indicated by white arrows in each SEM. (a) SU with 0 mW/cm², (b) TS with 0 mW/cm², (c) SU with 200 mW/cm², (d) TS with 400 mW/cm², (e) SU with 400 mW/cm², (f) TS with 400 mW/cm², (g) SU with 600 mW/cm², (h) TS with 600 mW/cm².

tooth structure. The thickness of the adhesives was lesser for the specimens irradiated with a weaker light intensity of <200 mW/cm².

DISCUSSION

Generally, the surface-free energy of organic substances (γ_s) has three component forces: the dispersion (γ_s^d), polarity (γ_s^p), and hydrogen-bonding (γ_s^h) forces¹⁷. The dispersion force represents London interactions between apolar molecules, whereas the polar (nondispersion) force represents electrostatic, metallic, and dipolar interactions. Further to these two parameters of the γ_s , we also calculated the hydrogen-bonding force, which is related to water and hydroxyl components. Because the hydration of the adherend is of great importance to wettability—which is, in turn, key to dentin bonding—polar interactions and hydrogen-bonding characteristics must be accurately determined to clarify the interactions between the adhesives and water¹⁸. An estimation of dipolar interactions separately from hydrogen-bonding interactions may provide a novel insight into the mechanisms contributing to wettability and the bonding characteristics of the adhesives.

The γ_s^d values of the dentin surfaces remained relatively constant (38.3–39.8 mN/m) regardless of the adhesive used, and there were no significant differences in γ_s^d between light intensity groups. Reportedly, the γ_s^d values of many oxidized surfaces measured using this method are approximately 40 mN/m^{12,15,19}, whereas those of surfaces coated with polytetrafluoroethylene are around 20 mN/m.

The γ_s^p values of all specimens were relatively low (from 4.9 to 10.6 mN/m), and decreased with increasing light intensity. The γ_s^h values ranged from 16.7 to 24.4 mN/m at light intensities of 0 and 200 mW/cm², and from 10.1 to 12.6 mN/m at light intensities of 400 mW/cm² or greater. The γ_s^p value, which reflects polar interactions, and the γ_s^h value, which relates to water and hydroxyl components, together measure hydrophilic interactions. The functional monomers of the adhesives used have three distinct structural components: functional, spacer, and polymerizable groups²⁰. The functional group exhibits hydrophilic properties that enhance the wettability and demineralization of the tooth surface²¹. The spacer group influences the properties of the monomer, including water solubility, viscosity, wettability, and penetration²². However, after light irradiation at sufficient light intensities, the polymerizable groups of the functional monomers become polymerized, increasing the hydrophobicity of the cured adhesive²³. Therefore, sufficient light energy may enhance the polymerization reaction and encourage the double-bond conversion of resin monomers in the adhesives. Conversely, incomplete polymerization may lead to residual water within the adhesive²⁴. Irradiation of the adhesive at a lower light intensity may impair cross-linking within the adhesive as a result of insufficient cross-linked functional monomers and excess water within the adhesive; this may lead to higher γ_s^p and γ_s^h values in adhesives irradiated at a lower light intensity.

When the adhesives were cured at a greater light intensity, the resultant dentin bond strength values

were higher than those obtained when the adhesives were polymerized with a light intensity of 200 mW/cm² or less. From SEM observation of the dentin-resin interface, the thickness of the adhesive was much less in the group irradiated with a weaker light intensity. These data suggest that the adhesives acquire inferior mechanical properties as a result of insufficient polymerization by inadequate light energy, in turn reducing bond strength. Furthermore, irradiation of single-step self-etch adhesives at a lower light intensity retards the polymerization process, leading to the creation of a thicker, uncured adhesive layer that includes an oxygen-inhibited layer²⁵. Presumably, residual acidic functional monomers, water, and solvent are present in this oxygen-inhibited layer¹². These remnants may impair the secondary polymerization of the adhesive and the polymerization of the filler resin, because tertiary amines in the adhesives and filler resin composite may be neutralized by the acidic functional monomers in the uncured adhesive²⁶. Acidic conditions affect polymerization driven by dl-camphorquinone/amine catalysts, resulting in poor polymerization and attenuating interactions with the filler resin composite²⁷.

In this study, when the surface-free energy of single-step adhesives decreased following irradiation at a greater light intensity, dentin bond strength values increased. Optimal wettability is essential to enable the materials to spread across the entire surface and establish adhesion²⁸. Although in general, the surface-free energy of the adherend surface must be maximized, the maximum bond strength was evident in properly polymerized adhesives, which have a lower surface-free energy¹⁴. Therefore, full polymerization of the adhesive has a greater influence on shear bond strength than the surface-free energy of the adhesive. This suggests that the degree of polymerization of adhesives with a lower surface-free energy significantly influenced dentin-bond strength. Our null hypothesis that there would be no significant differences in surface-free energy and bond strength at different light intensities, can therefore be rejected.

CONCLUSION

Regarding the single-step self-etch adhesives evaluated in this study, polymerization with a greater light intensity reduced the surface-free energy of adhesive-treated dentin surfaces, because conversion of the polymerizable group of functional monomers was enhanced. Sufficient light energy may enhance the polymerization reaction and perpetuate the double-bond conversion of resin monomers in the adhesives, leading to higher dentin bond strength. When the single-step self-etch adhesives were polymerized with a light intensity of 200 mW/cm² or less, stronger polarity and hydrogen-bonding forces were evident in the adhesives, leading to inferior bonding performance, possibly due to suboptimal polymerization or the influence of residual acidic functional monomers and

water. These data suggest that dentin bond strength and the polymerization behavior of single-step self-etch adhesives are affected by light intensity during curing.

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