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복합레진 수리에 있어 열순환이 층으로 형성된 시편의 물리적 성질에 미치는 영향

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〈Abstract〉

Effect of Thermocycling on the Mechanical Properties of Layered Specimens Formed for Resin Repair

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본 연구는 열순환이 두 층으로 형성된 복합레진 시편의 물리적 성질에 어떤 영향을 주는지 평가하기 위함이다. 실험을 위해 5개의 복합레진을 선택하고 두 층을 이루는 시편을 만들기 위하여 하부층은 열 순환된 4종류의 복합레진으로 그리고 상부층은 1 종류의 복합레진으로 제작하였다. 미세경도는 하부층으로 사용되는 복합레진만으로 단층의 시편을 제작한 다음 측정하였다. 두 층을 이루는 시편은 하부층으로 사용되는 복합레진을 4°C 와 60°C 로 5000 회 열순환 한 다음 상부층 복합레진으로 수복하여 제작하였다. 이후 굴곡강도 및 굴고계수와 압축 강도 및 압축계수를 측정하였다. 실험결과 미세경도는 열순환 후 측정된 시편들에 있어 증가 또는 감소를 보였다. 모든 경우에 상부층 복합레진으로 단층으로 제작한 시편이 두 층으로 제작한 시편들 보다 훨씬 높은 굴곡 및 압축강도를 보였다. 그러나 열순환 후 굴곡 및 압축강도는 유의하지 않은 증가 혹은 감소를 보였다. 열순환 후 굴곡강도의 최소값(92 MPa)은 ISO 4049 요구값(〉80 MPa) 보다는 컸고, 굴곡계수는 유의하게 증가된 반면 (7.4-105.2%) 압축계수는 유의하지 않은 증가 혹은 감소를 보였다. 결론적으로 물리적 성질에 있어 하부층을 열순환 후 상부층에 하부층보다 높은 굴곡 및 압축강도를 가진 복합레진으로 수복한 시편이 그들을 단층으로 제작한 시편보다 기계적 변화가 일정하지 않다.

Key words : Thermocycling, Layered specimen, Microhardness, Mechanical properties

Ⅰ. INTRODUCTION

The restoration of teeth damaged by accident or by caries-related diseases is a routine process in dentistry to restore inherent tooth function and anatomy. Of the various dental materials available, dimethacrylate-based composite resins are most widely used due to their excellent aesthetics and mechanical properties that are compatible with those of host teeth, ease of handling, and short polymerization time. However, composite resins used for restoration are continuously exposed to various stresses in forms of food, heat, and mechanical loads.

Usually the tooth cavity shows only slight pH changes due to the buffering effect of saliva, but pH within the tooth cavity can be readily changed from acid to alkali by food or the acids produced by oral bacteria when they metabolize carbohydrates (Loesche, 1986; McCabe, 1990;

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Edgar & Mullane, 1996). Temperatures in teeth can change widely due to the consumptions of cold and hot liquids or foods or by smoking. Temperature fluctuations depend on age and dietary habits, but generally are within the range 0°C to 60-70°C (Spierings et al., 1987; Barclay et al., 2005). Many components of the tooth cavity can be changed dynamically due to such temperature fluctuations. Among them, composite resin discoloration due to foods and beverages (Malhotra et al., 2011; Lepri and Palma-Dibb, 2012; Ren et al., 2012) and wear and degradation due to diverse mastication loads modify initial surface conditions (Wonglamsam et al., 2008; Durner et al., 2010; Palaniappan et al., 2011), and result in the need for repair or replacement. However, replacement of entire restored composite resins is impractical, because the interface between composite resins and tooth margins is unclear (due to their similar shades), and thus, repair of discolored or damaged portions of composite resin is a more attractive alternative. Recently, many reports have been issued on resin repair and resultant bond strengths between repaired composite resins for different bonding agent options (Maneenut et al., 2011; Staxrud and Dahl, 2011; Giachetti et al., 2012; Daur and Ilie, 2013).

During the repair of previously restored composite resins, color (shade) matching between over- and underlying composite resins is important to maintain consistency between layers. The purpose of the present study was to evaluate changes in the mechanical properties of specimens subjected to thermocycling and then layered with composite resin to mimic repair situations. Furthermore, the overlying resin product has a much higher strength and modulus than the underlying resin.

II. MATERIALS AND METHODS

1. Specimens and light-curing unit (LCU)

For this study, five composite resins [for overlying: Filtek Z350XT (ZX); for underlying: Palfique Estelite LV-HF (PH), Palfique Estelite LV-LF (PF), Filtek P90 (P90), and Premise Packable (PR)] were used to make layered specimens. Resin characteristics are listed in Table 1. Polymerization of specimens was performed using a LED light-curing unit (LCU) (L.E.Demetron, Kerr, Danbury, CT, USA] at an output light intensity of approximately 900 mW/cm^2 as measured using a built-in radiometer.

2. Microhardness measurement

To measure the surface microhardnesses (Hv) of

Table 1. Materials tested in the present study

	Composition	Filler content vol%/wt%	Manufacturer
ZX	Bis-GMA, Bis-EMA, TEGDMA, UDMA, PEGDMA, zirconia/silica filler & nanoclusters	63.3 / 78.5	3M ESPE St. Paul, MN, USA
PH	Bis-GMA, TEGDMA, Silica-zirconia, silica-titania filler	50 / 68	Tokuyama, Tokyo, Japan
PL	Bis-EMA, Bis-GMA, TEGDMA, Silica-zirconia, silica-titania filler	50 / 65	Tokuyama, Tokyo, Japan
P90	Silorane, Silanized quartz, yttrium fluoride	55 / 76	3M ESPE, St. Paul, MN, USA
PR	Bis-EMA, TEGDMA, Barium glass, SiO_2	71 / 84	Kerr, Orange, CA, USA

ZX: Filtek Z350XT; PH: Palfique Estelite LV-HF (High Flow); PL: Palfique Estelite LV-LF (Low Flow); P90: Filtek P90; PR: Premise Packable
wt%: according to the information supplied by the manufacturers

non-layered specimens used as underlying material, resin was filled into a metal mold (4×2×3 mm) and light cured for 40 s using the LCU. Cured specimens were removed from the mold and aged for 24 h in a 37°C dry, dark chamber. The microhardnesses of top (z=0) and bottom (z=3 mm) surfaces were measured using randomly selected specimens using a Vickers hardness tester (MVK-H1, Akashi, Tokyo) by evaluating the sizes of microindentations (n=12 for each test condition). To make these microindentations, a 200 gf load and 10 s dwell time conditions were applied. Another microhardness measurement of aged specimens was performed in conjunction with the thermocycling process using themocycling machine (DC-TC005, Dongsung, Busan, Korea). Briefly, aged specimens were immersed in an alternating manner in two water baths at 4°C and 60°C 5000 times with a 30 s dwell time in each bath. After thermocycling, microhardness measurements were performed using the measurement conditions mentioned above.

3. Evaluation of flexural properties

A three-point bending test was performed before and after thermocycling to determine flexural properties [flexural strength (FS) and modulus (FM)]. To produce specimens for the before thermocycling condition (non-layered), stainless steel molds (25×2×2 mm) were filled with each resin product (ISO 4049, 2000). Both top and bottom surfaces were then flattened using two thin glass slides, and resins were light cured for 200 s (40 s × 5) by overlapping exposures. Cured specimens were then removed from molds and aged for 24 h in a 37°C dry, dark chamber. To produce specimens for the after thermocycling condition (layered), stainless steel molds (25×2×1 mm) were filled with resins (except ZX), top and bottom surfaces were then flattened using two glass slides, and resins were light cured for 200 s (40 s × 5) by overlapping exposures. Cured resins were then removed from molds and aged for 24 h in a 37°C dry, dark chamber. After aging, specimens were thermocycled 5000 times between 4°C and 60°C with a 30 s dwell time in each bath. Thermocycled specimens were then plugged into the bottom of the 2-mm thick mold, the upper remaining space was filled with ZX, covered using a thin glass slide, pressed firmly, and light cured totally for 200 s using five overlapping exposures. After light curing, specimens were removed from molds and aged for 24 h in a 37°C dry, dark chamber. Specimens for before (n=7) and after (n=7) thermocycling conditions were loaded into a universal test machine (Instron 3345, Grove City, PA, USA) and tested at a crosshead speed of 1 mm/min. FS (σ_f in MPa) was obtained using the following formula:

$$\sigma_f = 3DP/(2WH^2)$$

where D is the distance between supports (20 mm), P is maximum failure load (N), W is sample width (2 mm), and H is sample height (2 mm). FM (E in GPa) values were calculated using the following formula:

$$E = (P/D) \cdot (D^3/(4WH^3))$$

where P/D is the slope of the linear portion of the load-displacement curve.

4. Evaluation of compressive properties

To measure compressive properties [compressive strength (CS) and modulus (CM)] of specimens before and after thermocycling, two matching stainless steel hemicylinders (inner diameter: 3 mm, outer diameter: 10 mm, height: 3 and 6 mm) were manufactured. To produce specimens for the before thermocycling condition (non-layered), the empty inner space of the hemicylinders (height: 6 mm) was filled with a resin product, and the resin was then exposed to light on its top and bottom surfaces for 5 s. One of the two hemicylinders was then removed, and the uncovered lateral surface was exposed to light for 40 s. The remaining hemicylinder was then removed and its uncovered lateral surface was exposed to light in the same manner. After curing, the specimen was removed from the mold and aged for 24 h in a 37°C dry, dark chamber. To produce specimens for the after thermocycling condition (layered), resin (except ZX) was filled in the inner space of the hemicylinders (height: 3 mm), and exposed to light on its top and bottom surfaces for 5 s. One of the two hemicylinders was then removed, and the uncovered lateral surface was exposed to light for 40 s. The remaining hemicylinder was then removed and its uncovered lateral surface was exposed to light in the same manner. After light curing, specimens were removed from the mold and aged for 24 h in a 37°C dry, dark chamber. Aged specimens were thermocycled 5000 times in the two water baths between 4°C and 60°C with 30 s dwell time in each bath. The thermocycled specimens were then plugged into the bottom of the 6-mm high mold, the upper space was filled with ZX, covered with a glass slide, pressed firmly to make a flat surface, and light cured for 5 s. Light curing of ZX was performed as described above. After light curing, specimens were removed from the mold and aged for 24 h in a 37°C dry, dark chamber. Specimens for before (n=7) and after (n=7) thermocycling were loaded into a universal test machine and compression tested at a crosshead speed of 1 mm/min. CS (σc in MPa) values of specimens were obtained using the following formula

$$\sigma_c = P/A$$

where P is the maximum failure load (N) and A is specimen cross-sectional area. CM (in GPa) was defined as the slope of the linear portion of the load-displacement curve.

5. Statistical analysis

Differences between the before and after thermocycling results were analyzed with the student's t-test using SPSS Ver. 21.0 (SPSS, Chicago, IL, USA). Statistical significance was accepted for *p* values 〈 0.05.

Ⅲ. RESULTS

The microhardnesses of non-layered specimens before and after thermocycling are shown in Table 2. Initial microhardnesses of top surfaces were significantly changed (*p*〈0.05) by thermocycling, but those of bottom surfaces were not significantly changed. Two flowable resins (PH and PL) showed slight increases in microhardness after thermocycling.

Table 3 shows the flexural properties (FS and FM) of specimens. After thermocycling, the FS values of layered specimens changed insignificantly, which were obtained

Table 2. Microhardness (Hv) of the non-layered specimens before and after thermocycling

		PH	PL	P90	PR	t-test
Top	24 h	42.9 ± 0.4	10.8 ± 0.5	57.9 ± 0.6	66.9 ± 0.4	SD
	thermocycling	33.4 ± 0.2	11.2 ± 0.3	51.3 ± 0.2	51.4 ± 0.3	
Bottom	24 h	14.0 ± 1.6	5.9 ± 0.3	46.9 ± 0.4	56.3 ± 0.6	NSD
	thermocycling	16.3 ± 2.3	6.9 ± 0.2	44.4 ± 0.4	46.6 ± 0.2	

PH: Palfique Estelite LV-HF (High Flow); PL: Palfique Estelite LV-LF (Low Flow); P90: Filtek P90; PR: Premise Packable
SD: Significantly different; NSD: Not significantly different

Table 3. Flexural properties (FS and FM) of layered specimens with ZX before and after thermocycling

		PH	PL	P90	PR	t-test
FS (MPa)	24 h	185.2±6.0	134.5±20.2	136.5±11.3	128.7±15.0	NSD
	thermocycling	166.1±28.4	173.6±29.0	92.2±23.1	118.3±9.3	
FM (GPa)	24 h	11.05±0.60	5.41±0.64	14.70±0.79	13.63±1.43	SD
	thermocycling	14.32±2.45	11.10±0.60	15.79±1.46	16.18±0.77	

* After aging 24 h, FS and FM of ZX were 188.1±7.8 MPa and 17.04±1.19 GPa, respectively.
PH: Palfique Estelite LV-HF (High Flow); PL: Palfique Estelite LV-LF (Low Flow); P90: Filtek P90; PR: Premise Packable

Table 4. Compressive properties (CS and CM) of layered specimens with ZX before and after thermocycling

		PH	PL	P90	PR	t-test
CS (MPa)	24 h	323.0±35.7	319.0±45.0	302.2±35.2	334.3±40.1	NSD
	thermocycling	352.1±40.0	294.3±21.5	289.8±59.9	345.2±18.6	
CM (GPa)	24 h	4.29±0.08	2.90±0.39	4.08±0.21	4.24±0.15	NSD
	thermocycling	3.96±0.50	3.33±0.37	4.12±0.36	4.10±0.12	

* After aging 24 h, CS and CM of ZX were 388.0±39.0 MPa and 4.83±0.05 GPa, respectively.
PH: Palfique Estelite LV-HF (High Flow); PL: Palfique Estelite LV-LF (Low Flow); P90: Filtek P90; PR: Premise Packable

from their non-layered state. Except PL, which showed a 29.1% increase in FS, the other specimens showed 8.1-32.4% decreases in initial FS values. On the other hand, FM values were significantly increased by thermocycling (7.4-105.2%, which were obtained from their non-layered state) ($p<0.05$).

Table 4 summarizes the compressive properties (CS and CM) of specimens. Initial CS and CM values in the non-layered state were inconsistently changed by thermocycling. Among the specimens, PH and PR showed slight increases in CS, but slight decreases in CM after thermocycling. CS and CM were not significantly changed ($p>0.05$) by thermocycling before (non-layered state) and after thermocycling (layered state).

Ⅳ. DISCUSSION

Restored composite resins in the tooth cavity are inevitably subjected to wear, staining, and degradation due mastication activity and the absorption of water-based fluids (Wonglamsam et al., 2008; Durner et al., 2010; Malhotra et al., 2011; Palaniappan et al., 2011; Lepri and Palma, 2012; Ren et al. 2012). As a result, repair or replacement is often requested during restoration. In terms of cost and ease of work, the repair of damaged or discolored portions of restored composite resins often provides an optional solution. In addition, since composite resins are frequently withdrawn from the market, matching using same resin products is not always possible. Furthermore, if the brand name of the previously used resin product is not known, users have no option but select what they consider a suitable restoration composite resin. The layered specimens in the present study reflect such situations

In the present study, microhardness was evaluated to determine how repeated thermocycling affects polymerized specimens. As was expected, flowable resins had low microhardnesses on top and bottom surfaces due to their low filler contents. After thermocycling, PL showed a slight increase in top surface microhardness, and PH and PL (flowable resins) showed slight increases in bottom surface microhardness. The present study shows that repeated thermal cycling and aging produce different patterns of change in the mechanical properties of flowable and other composite resins. Since the thermocycling process involves repeated heating and cooling, it affects the lifetimes of monomer radicals. Heating may produce free radical and reduce their half-lives, as occurs during post-curing (Wu and Fanconi, 1983). On the other hand, subsequent cooling process reduces free radical production but extends half-lives (Burtscher, 1993). Also, due to short water bath dwell time and low polymer thermal conductivity, specimens temperature are not fully achieved. However, 5000 cycles of 30 s in the two baths took more than 3 days, and thus, additional aging (curing) could have occurred during thermocycling than that of the only 24 h aged (non-thermocycled). The slightly increase in the microhardnesses of flowable resins observed was probably due to curing of residual monomers.

The flexural properties (FS and FM) measured by three-point bend testing are measures of a specimen's ability to resist stresses without fracture. The FS value obtained from non-layered specimens ranged from 128.7 MPa to 188.2 MPa, and PH and ZX had much higher (185 and 188 MPa, respectively) FS values than the others (129-137 MPa). When resin products were thermocycled and then layered with ZX, layered specimens had much lower FS values than upper and underlying resin products, except PL. The FS values of layered PL were much increased by thermocycling, and values were greater than the underlying, but lower than the overlying resin product. PH and PL (the layered flowable resins) had higher FS values close to the overlying ZX. However, P90 and PR had lower FS value than that of each of their non-layered state. In the case of FM, initial FM values were significantly increased by thermocycling and pre- and post-thermocycling FM values were linearly correlated (R=0.98). Thus, when the FM value of an overlying resin product (ZX) was greater than that of the underlying, the FM value of the layered product was greater than that of the non-layered underlying resin product. However, FS and FM values of layered specimens were greater than the ISO 4049 requirement (greater than 80 MPa) and lower than dentin (17-25 GPa), respectively, after thermocycling (Xu et al., 1998; Mahoney et al., 2000; Kinney et al., 2003; Leprince et al., 2011).

The compressive properties (CS and CM) of non-layered specimens changed inconsistently (increased or decreased) by thermocycling of each resin products and then layered with ZX. After thermocycling, layered PL and P90 had lower CS values than the non-layered underlying resin products (although overlying ZX had much greater CS values than that of others). On the other hand, PH and PR showed slightly CS increases after thermocycling (although their values were still lower than that of overlying ZX). In the case of CM, the situation was reversed. Layered PL and P90 with ZX which had much higher CM values than the others showed non-significantly higher CM values after thermocycling. Like FM, CM values before and after thermocycling were also linearly correlated (R=0.95). Furthermore, the CM values of thermocycled and then layered specimens are much lower than that of dentin (11.0-18.5 GPa) (Craig and Peyton, 1958; Watts et al., 1987) and were highly correlated (R=0.98) with the FM values of the same thermocycled and layered specimens.

V. CONCLUSIONS

Within the limitations of the present study, the following conclusions could be reached:

1. Initial low microhardnesses, such as, the top surface of PL and the bottom surfaces of PH and PL, were slightly increased by thermocycling.
2. As compared with non-layered specimens before thermocycling, the FS and CS values of thermocycled and then layered specimens changed inconsistently

and insignificantly. FM values significantly increased after thermocycling, whereas CM values changed insignificantly.

3. Nonetheless, under all circumstances, the flexural and compressive properties of thermocycled specimens subsequently layered with ZX satisfied ISO 4049 requirements and were compatible with dentin due to their low mechanical properties.

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