Effect of storage conditions on mechanical properties of resin composite blanks for CAD/CAM crowns

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The objective of this study was to examine the effects of storage conditions on the flexural strengths of resin composite materials for CAD/CAM restorations. Seven commercially available resin composite CAD/CAM blanks were examined. Rectangular specimens (4.0×1.2×4.0 mm) of each material were trimmed from the blanks and subjected to thermal cycling between 5°C and 55°C in deionized water at 10,000 cycles or stored in 37°C deionized water for one week or air for one day ($n=10$ for each condition). The difference in storage condition between water immersion and thermal cycling did not affect the flexural strengths of resin composite materials for CAD/CAM examined in this study. The resin composite block CS300 made from Bis-MEPP and UDMA showed the greatest flexural strength under all storage conditions and less deterioration of strength by thermal cycling and water immersion among the resin composite blocks tested.

Keywords: Resin composite block for CAD/CAM crowns, Flexural strength, Storage conditions, Thermal cycling, Elastic modulus

INTRODUCTION

In recent years, crown restorations are apt to be metal-free due to increased concerns about the incidence of metal allergy with metallic restorations and increased consciousness of esthetic appearance among patients. In addition, with the rapid development of digital technologies, the application of CAD/CAM in fabricating dental restorations has become widespread^{1,2)}. Metallic materials, ceramic materials and resin composites are used for crown restorations. Since the use of resin composite materials for CAD/CAM as posterior restoration materials was approved under the Japanese social health insurance system in 2014, many manufacturers are trying to develop new resin composite materials for CAD/CAM. This is not only for the Japanese domestic market; the use of resin composite materials for CAD/CAM will spread to other countries if these materials offer acceptable properties for posterior crown restorations.

Resin composites for CAD/CAM are made into block shapes by a polymerization process, placing a polymer matrix resin containing fillers under high pressure and high temperature conditions³⁾. As resin composite blocks (or blanks) for CAD/CAM can possess a higher degree of polymerization and greater content of filler, these materials show greater and more stable strengths in comparison to direct restorative resin composites. Previous studies on resin composite blanks have examined mechanical properties such as wear behavior, hardness, flexural strength, flexural modulus, glossiness after polishing, and adhesive strength to tooth structures using resin cements⁴⁻¹⁵⁾. Hamakubo *et al.*⁴⁾ reported that CAD/CAM resin blocks exhibited lower compressive strength but comparable flexural strength in comparison with prosthodontic resins or

crown and bridge. Lauvahutanon *et al.*^{5,6)} found that after 10,000 thermocycles the flexural strength of CAD/CAM resins was reduced but their amount of wear was lower than conventional posterior resin composites. Kamonkhantikul *et al.*¹⁰⁾ reported that although some CAD/CAM resin blocks showed increased surface roughness after a toothbrush wear test with increased brushing cycles, the majority of CAD/CAM blocks showed significantly reduced values of both surface roughness and degree of gloss after brushing.

As for the adhesion of resin cements to CAD/CAM resin blocks, silane treatment after sandblasting of CAD/CAM resin surfaces showed greater adhesive strength than silane or sandblasting treatment with no surface treatment¹²⁾. Cleaning using an ultrasonic cleaner or phosphoric acid was reported not to affect the adhesive strength of resin cements¹³⁾. Cekic-Nagas *et al.*¹⁴⁾ found that surface treatment using 9.6% hydrofluoric acid solution did not increase the adhesive strength between CAD/CAM resin blocks and resin cements.

Materials for crown restorations require high mechanical strength and durability. In the oral cavity, deterioration of resin composite restorative materials is caused by hydrolysis of the silane bonds between the matrix resin and filler during extended periods of water contact, resulting in separation of the filler particles from the matrix¹⁶⁻¹⁹⁾. In addition, water sorption into composite materials was reported to reduce their mechanical properties^{20,21)}. This deterioration behavior found in resin composite filling materials would likely also occur in resin composite restorations made by CAD/CAM. Therefore, investigation of the deterioration behavior, particularly as it affects the mechanical properties, of these materials in water will be useful in understanding the longevity of materials in the oral cavity and in improving and developing new materials.

The objective of this study was to examine the mechanical properties of resin composite materials for CAD/CAM, including a newly-developed resin composite material intended for use in molar restorations. We conducted a thermal cycling experiment to investigate the mechanical durability of the resin composite materials for CAD/CAM blanks.

The hypothesis to be tested in this study was that the flexural strength of CAD/CAM resin blocks is affected by different storage conditions, *i.e.*, whether the specimens are kept in air or water, or subjected to thermal cycling.

MATERIALS AND METHODS

Materials

Six commercially-available resin composite CAD/CAM blanks and a newly-developed resin composite CAD/CAM blank (Cerasmart 300, GC, Tokyo, Japan) were examined (Table 1). Table 1 also shows the composition data from the manufacturers, lot numbers and other information.

Methods

1. Hardness measurements

The hardness value of each resin composite blank (12.0×14.0×18.0 mm) was determined using a micro-Vickers hardness tester (HM-102, Mitsutoyo, Kanagawa, Japan) at 0.98 N of loading for 30 s at 23±2°C and 50±10% relative humidity in air. Five measurement points were randomly selected on the lateral walls of blank surfaces. The measured values were averaged for each blank, and five blanks for each resin composite were examined. The

results were analyzed using one-way ANOVA followed by Scheffé's test at $\alpha=0.05$ (SPSS Statistics 20, IBM, Armonk, NY, USA and StatMate V, ATMS, Tokyo, Japan).

2. Flexural strength measurements

The flexural strength of CAD/CAM resin blocks was determined according to ISO6872:2015²²⁾ and JADMAS 245:2017²³⁾. Rectangular specimens measuring 4.0±0.2×1.2±0.2×14.0 mm were cut from the resin composite blanks. The surfaces of specimens were finished using No.2000 SiC abrasive paper. Thirty specimens were prepared for each resin composite. Specimens were randomly divided into three groups of ten specimens each.

Of the 30 specimens, ten specimens were kept in air at 23±2°C for one day. Another ten specimens were subjected to thermal cycling between 5°C and 55°C in deionized water for 10,000 cycles. Cycling was conducted for 60 s at each temperature with 30 s of dwell time in air at 23±2°C between different temperature conditions (total of 3 min per cycle). The remaining ten specimens were immersed in deionized water at 37±2°C for 7 days.

The specimens after thermal cycling or storage in either water or air were subjected to flexural strength measurements. The flexural strengths of specimens were determined using a three-point bending jig (12.0 mm span length with a 2.0 mm diameter supporting or loading rod) at 1.0 mm/min of crosshead speed in a universal testing machine (Instron 3366, Instron, Boston, USA) in air at 23±2°C. The flexural strength of each specimen was calculated from the fracture load of the specimen using the following equation.

Table 1 Resin composite blocks for CAD/CAM crowns used in this study

Product	Manufacturer	Lot No.	Composition	Color	Code
Resin composite					
Cerasmart 300	GC	17062310	Monomer: Bis-MEPP, UDMA Filler: SiO ₂ , Ba glass	A3-LT	CS300
Cerasmart 270	GC	17062310	Monomer: Bis-MEPP, UDMA Filler: SiO ₂ , Ba glass	A3-LT	CS270
Shofu Block HC	Shofu	0117310	Monomer: UDMA, TEGDMA Filler: SiO ₂ , Microfumed SiO ₂ , Zirconium silicate	A3-LT	HC
KZR-CAD HR2	Yamakin	20021713	Monomer: UDMA, TEGDMA Filler: SiO ₂ - Al ₂ O ₃ - ZrO ₂ , SiO ₂	A3	KZR
Katana Avencia Block	Kuraray-Noritake	000510	Monomer: UDMA Filler: SiO ₂ , Al ₂ O ₃	A3-LT	AVE
Estelite Block	Tokuyama Dental	044067	Monomer: UDMA, TEGDMA Filler: SiO ₂ , Zirconia filler	A3-LT	EST
Hybrid ceramic					
Vita Enamic for Kavro Arctica	Kavo Dental	60870	Monomer: UDMA, TEGDMA Filler: Feldspar ceramic enriched with Al ₂ O ₃	3M2-T	ENA

$$\delta = 3Pl/2wb^2$$

where δ is flexural strength (MPa), P is fracture load (N), l is the span length between support rods, w is specimen width (mm) and b is specimen thickness (mm).

The results were analyzed using two-way ANOVA followed by Scheffé's test at $\alpha=0.05$.

3. Flexural modulus measurements

The flexural modulus of each specimens was calculated from the load–deflection curve when the flexural strength of the specimen was obtained in the universal testing machine. The results were analyzed using one-way ANOVA followed by Scheffé's test at $\alpha=0.05$.

4. Determination of inorganic filler contents in resin composites

A specimen measuring 2.0×1.2×2.0 mm was cut from a flexural strength test specimen stored in air after the strength test. After the mass of flexural strength test specimen which was subjected to air storage condition was measured, the specimen was heated in air using TG-DTA (Thermo plus EVO2 TG8120, Rigaku, Tokyo, Japan) at 10°C/min of heating rate up to 900°C with a 3-h holding time and the mass of specimen was recorded. The mass of specimen after heating was divided by that before heating to obtain mass% of inorganic filler content because the residue of specimen after heating was believed to be the inorganic components. Three specimens for each resin composite material were examined.

The results were analyzed using ANOVA followed

by Scheffé's test at $\alpha=0.05$.

5. Scanning electron microscopic observation

The fractured surfaces of specimens after the flexural strength test were examined using a scanning electron microscope (JSM-6360LV, JEOL, Tokyo, Japan). SEM observation was performed on the compressed side (upper side) and tension stressed side (lower side) of the specimens. The specimens were coated with gold film (20 nm thick) and observed at 20 kV of acceleration voltage.

RESULTS

Hardness

Vickers hardness numbers of the resin composite blanks examined are listed in Table 2. The results of one-way ANOVA indicated significant differences among the resin composite blanks ($F=2963.47$, $p<0.0001$, Table 3).

The resin composite ENA showed the greatest hardness value among the materials examined ($p<0.05$). The resin composites CS300 and EST were significantly harder than CS270, HC, KZR and AVE ($p<0.05$). There were no significant differences in hardness values between CS300 and EST, between HC and AVE, between KZR and AVE, or among CS270, KZR and AVE ($p>0.05$).

Flexural strength

Figure 1 shows the flexural strengths of the resin composites. Flexural strength of the resin blocks ranged from 87 to 290 MPa. Significant effects of the resin block products ($F=405.004$, $p<0.0001$) and the

Table 2 Vickers hardness numbers of resin composite blocks used in this study

Product	Vickers hardness number
CS300	94.2 (2.0) ^a
CS270	82.7 (2.8) ^b
HC	71.6 (2.5) ^c
KZR	82.1 (3.2) ^{b,d}
AVE	76.3 (2.9) ^{b,c,d}
EST	96.4 (3.3) ^a
ENA	254.2 (1.5)

(): SD

Same lower-case letters indicate no statistical differences ($p>0.05$) among the resin composite blocks.

Table 3 Summary of analysis of variance for Vickers hardness numbers of resin composite blocks for CAD/CAM crowns

Source	Type III Sum of squares	df	Mean Square	F-value	p-value
Resin composite block	126,641.342	6	21,106.890	2,963.647	0.000
Error	199.414	28	7.122	—	—
Total	126840.756	34	—	—	—

Fig. 1 Flexural strengths of resin composite blocks for CAD/CAM crowns after different storage conditions. Same lower-case letters indicate no significant differences ($p>0.05$) among the different storage conditions for each resin composite block. Same upper-case letters indicate no significant differences ($p>0.05$) among the different resin composite blocks for each storage condition.

Fig. 2 Elastic moduli of resin composite blocks for CAD/CAM crown after different storage conditions. Same lower-case letters indicate no significant differences ($p>0.05$) among the different storage conditions for each resin composite block. Same upper-case letters indicate no significant differences ($p>0.05$) among the different resin composite blocks for each storage condition.

Table 4 Summary of analysis of variance for flexural strength of resin composite blocks for CAD/CAM crowns

Source	Type III Sum of squares	df	Mean Square	F-value	<i>p</i> -value
Resin composite block	455,814.549	6	75,969.091	405.004	0.000
Storage condition	19,212.780	2	9,606.390	51.213	0.000
Resin composite block $\times$ Storage condition	3,744.574	12	312.048	1.664	0.078
Error	35,451.867	189	187.576	—	—
Total	514,223.770	209	—	—	—

storage conditions ($F=251.213$, $p<0.0001$) were found for the flexural strengths, but there was no significant interaction between the resin block products and storage condition ($F=1.664$, $p>0.05$) (Table 4).

The resin composite blank CS300 had greater flexural strength than the other blanks in all storage conditions ($p<0.05$). HC showed the least flexural strength among the resin composite blocks in all storage conditions ($p<0.05$). CS300 and ENA did not vary significantly in flexural strength for all storage conditions ($p>0.05$). The flexural strengths of other resin blocks, CS270, HC, KZR, AVE and EST, stored in air were significantly greater than those when stored in water for 7 days and thermocycled ($p<0.05$). No resin blocks showed significant differences in flexural strength between storage in water for 7 days and the thermal cycling ($p>0.05$). In the storage condition, the flexural strengths of the resin composite blanks stored in air showed significantly greater than those when stored in water for 7 days and thermocycled ($p<0.05$).

The difference in storage condition between water immersion and thermal cycling did not affect the flexural strengths of resin composite materials for CAD/CAM

examined in this study.

Elastic modulus

The elastic modulus values obtained by flexural strength testing are shown in Fig. 2. The values of elastic modulus ranged from 6.4 to 30.1 GPa. ANOVA indicated significant effects of the resin block products ($F=12821.373$, $p<0.0001$) and the storage conditions ($F=419.113$, $p<0.0001$) on the elastic modulus values. A significant interaction was also found between the resin block products and the storage conditions ($F=23.761$, $p<0.0001$) (Table 5). In all storage conditions, ENA had the highest and HC had the lowest elastic modulus values among the resin blocks ($p<0.05$). No significant differences in elastic modulus values were found for CS270 among the different storage conditions ($p>0.05$). There were no significant differences in the elastic modulus values of CS300 between storage in air and 7 days' immersion in water or between 7 days' immersion and thermal cycling ($p>0.05$). The elastic modulus values of HC, KZR, AVE, EST and ENA were significantly affected by the storage conditions ($p<0.05$).

Table 5 Summary of analysis of variance for elastic modulus of resin composite blocks for CAD/CAM crowns

Source	Type III Sum of squares	df	Mean Square	F-value	p-value
Resin composite block	8,760.313	6	1,460.052	12,821.373	0.000
Storage condition	95.454	2	47.727	419.113	0.000
Resin composite block × Storage condition	32.469	12	2.706	23.761	0.000
Error	21.523	189	0.114	—	—
Total	8,909.759	209	—	—	—

Table 6 Summary of analysis of variance for content of inorganic filler of resin composite blocks for CAD/CAM crowns

Source	Type III Sum of squares	df	Mean Square	F-value	p-value
Resin composite block	1,469.378	6	244.896	2,508.816	0.000
Error	1.367	14	0.098	—	—
Total	1,470.745	20	—	—	—

Fig. 3 Inorganic filler contents of resin composite blocks for CAD/CAM crown. An asterisk symbol (*) indicate significant differences ($p < 0.05$) for each resin composite block.

Inorganic filler contents of resin composite blanks

Inorganic filler contents in the blocks are shown in Fig. 3. The amounts of inorganic filler content ranged from 57.4 to 85.2 mass%. The results of 1-way ANOVA indicated significant differences among the resin blocks ($F = 2508.816$, $p < 0.0001$) (Table 6). ENA had the highest filler content and AVE the least, among the resin blocks ($p < 0.05$). There were significant differences in filler content among the resin block products ($p < 0.05$).

Relationships of the inorganic filler content to flexural strength and elastic modulus

Figure 4 shows the relationships between inorganic filler content and flexural strength of all blocks examined. No significant correlation was found between inorganic filler content and flexural strength ($p > 0.05$). The relationships between inorganic filler content and elastic modulus values are shown in Fig. 5. A significant

Fig. 4 Relationship between content of inorganic filler and flexural strength of resin composite blocks for CAD/CAM crowns.

Fig. 5 Relationship between content of inorganic filler and elastic modulus of resin composite blocks for CAD/CAM crowns.

positive correlation was found between filler content and elastic modulus value ($r=0.877$, $p<0.05$).

Observation of fracture surfaces of specimens fractured in flexural strength tests

Figures 6 to 9 show representative fractured surfaces of specimens after the flexural strength tests. Figure 6

Fig. 6 Fractured surfaces of specimens after flexural strength tests by scanning electron microscope (CS300).

Fig. 7 Fractured surfaces of specimens after flexural strength tests by scanning electron microscope (HC).

Fig. 8 Fractured surfaces of specimens after flexural strength tests by scanning electron microscope (AVE).

Fig. 9 Fractured surfaces of specimens after flexural strength tests by scanning electron microscope (SENA).

shows the fractured surfaces of CS300 specimens. The specimen stored in air revealed a surface fractured in a brittle manner and flat. In comparison with the specimen stored in air, the specimens stored in water and after thermal cycling showed rougher surfaces and cracks on the compressed side of the specimen.

Figure 7 shows the fractured surfaces of HC specimens after flexural strength tests. Spherical filler shapes were observed on the fractured surfaces. The specimens stored in air and in water for 7 days had rougher surfaces. The specimens after thermal cycling also showed a rougher surface and less filler was exposed than for the specimens stored in air or in water for 7 days.

Figure 8 shows the fractured surfaces of AVE specimens after flexural strength tests. Compared to other resin blocks, cracks were observed at the compressed side of specimens for all storage conditions. The specimens stored in air and in water for 7 days showed hackle-shape or mirror-like cracks on the tension stress side, as often seen in brittle fractures²⁴.

Figure 9 shows the fractured surfaces of ENA specimens after flexural strength tests. The specimens stored in air and in water for 7 days both exhibited a rough surface with sharp edges. The fractured surface of the specimen after thermal cycling had a smoother appearance compared to those stored in air and in water for 7 days.

DISCUSSION

The present study determined the flexural strengths of resin composite materials. The results indicated that CS300 had the greatest strength and HC had the least strength under all experimental conditions ($p < 0.05$). Regarding the flexural moduli, ENA showed the highest values and HC the lowest values under all experimental conditions ($p < 0.05$). Except for CS300 and ENA, the resin blocks examined revealed lower flexural strengths after 7-day water immersion or 10,000 thermal cycles than after storage in air at room temperature ($p < 0.05$).

In this study, the specimens were subjected to thermal cycling between 5°C and 55°C in deionized water for 10,000 cycles. Each cycle was 60 s at each temperature with 30 s of dwell time in air at 23±2°C between temperature conditions. These experimental conditions were selected based on previous studies^{25–29}. The 10,000 cycles in this study were considered to be equivalent to approximately 2 years and 6 months in the oral cavity, assuming 10 thermal cycles in the oral cavity per day³⁰.

The flexural strengths of resin blocks stored in air at room temperature ranged from 133 to 270 MPa. Conventional dental composite resins and hybrid resins for crown and bridge are reported to have flexural strengths ranging from 169.1 to 196.0 MPa⁴. Some of the resin blocks we examined showed greater flexural strength than the conventional dental composite resins for crown and bridge (resin jacket crown). The flexural moduli of the resin blocks in this study ranged from 9.0 to 29.3 GPa, whereas those of the conventional dental composite resins for crown and bridge (resin jacket crown) are reported to range from 9.4 to 21.4 GPa⁴. This suggests that the strength and elastic moduli of the resin blocks examined in this study are acceptable for clinical application.

The present study showed no significant differences in flexural strength between the resin blocks stored in water for 7 days or thermocycled for 10,000 cycles and those stored in air ($p > 0.05$).

Ferracane³¹ reported that polymers used in dental restorative materials are subject to both hygroscopic and hydrolytic effects that may influence their mechanical properties, dimensional stability and biocompatibility.

The water absorptions of resin composite were previously reported by many researchers^{32–36}. Fan *et al.*³² reported that the water sorption in direct restorative resins occurs in the organic resin matrixes. Thus, a more valid expression of water sorption values is the amount of water absorbed per unit weight of organic matrix content (resin matrix). It is more indicative of the water sorption characteristics of the resin matrix. Sideridou *et*

*al.*³³⁾ reported that the water sorption at equilibrium for poly (Bis-EMA), poly (Bis-GMA), poly (UDMA) and poly (TEGDMA) were 1.8, 2.6, 2.9 and 6.3 wt%, respectively. Polymers made with the UDMA monomer exhibit similar or slightly less water sorption than those prepared from Bis-GMA, however, UDMA polymers show significantly more water uptake than polymers based on non-hydroxylated Bis-GMA analogues such as Bis-EMA.

Musanje and Darvell³⁴⁾ reported that water sorption not only affects physical and mechanical properties, especially of composite resins but also decreases surface hardness and elastic modulus. Ito *et al.*³⁵⁾ reported that the least hydrophilic resin (E-Bis-GMA and TEGDMA) absorbed 0.55 wt% water and showed a 15% decrease in modulus after 3 days and the most hydrophilic experimental resin (Bis-GMA and HEMA) absorbed 12.8 wt% water and showed a 73% modulus decrease during the same periods.

Therefore, the resin block used this study is considered to have low water absorption of the resin block because the main monomers are UDMA and Bis-MEPP. Under the present experimental conditions, water sorption was considered not to noticeably influence the flexural strength of the resin blocks examined.

As for the main resin monomers used in the materials, HC and AVE are composed of UDMA, and ENA is composed of UDMA and TEGDMA. Both CS300 and CS270 employ Bis-MEPP and UDMA. All the resin composites in this study were made from similar resin matrix monomers, but CS300 and CS270 used Bis-MEPP in addition to UDMA. This difference in resin monomer composition from other materials possibly increased the flexural strength of these two materials. The composites based on dimethacrylates without hydroxy groups (Bis-MEPP) showed a relatively greater flexural strength than those with hydroxy groups (Bis-GMA) under wet conditions³⁷⁾. Bis-MEPP is also known to be a rigid monomer³⁸⁾.

The stable and greater flexural strength of CS300 in this study could be explained by the use of Bis-MEPP, having lower water sorption, as a matrix component and filler content in the resin composite block. Furthermore, the production process for CS300 may be different from other resin blocks, increasing the degree of polymerization and density.

Although CS270 contained a similar matrix resin and filler to CS300, CS270 showed lower flexural strength. This difference in strength may have stemmed from a difference in filler content in the resin composite block.

Water sorption of matrix components and reduction of bonding force between resin matrix and filler are known to affect the deterioration of resin composite materials¹⁶⁻¹⁹⁾. The resin components of CS300 and CS270 blocks are mainly composed of Bis-MEPP, which shows hydrophobic behavior, and UDMA, which is still hydrophobic but rather hydrophilic compared to Bis-MEPP³⁷⁾. Although water likely penetrated into the resin blocks by immersion in water, the matrix resin components would absorb less water because of their

hydrophobic nature. Therefore, the water may have had a greater effect reducing the bonding of resin matrix and filler in CS300 and CS270 resin blocks.

Filler content determinations in resin composites should be measured by volume ratio. In recent years, fillers incorporated in resin composites were included both organic composite fillers and inorganic fillers. In the previous report^{5,8)}, the mass of the specimen after heating, as the mass of inorganic fillers in the composite material, was divided by the initial mass of the specimen to obtain the amount of inorganic filler as a simple method for measuring the filler content. Therefore, we calculated the inorganic filler content based on the mass fraction and considered that it could be effective in examining the relationship between the strength and filler content of the resin composites.

From the results of filler content determinations, the inorganic filler content ranged from 57.4 to 85.2 mass%, and ENA had the largest filler content among the materials examined in this study. It is generally believed that an increase in filler content increases the strength and the hardness of resin composite materials³⁹⁻⁴²⁾. ENA showed the greatest Vickers hardness value, while that of HC was the lowest ($p < 0.05$). We found a significant positive correlation between Vickers hardness and filler content ($p < 0.05$, $r = 0.850$), and a trend that an increase in filler content increased the Vickers hardness. However, there was no significant correlation between flexural strength and filler content in this study ($p > 0.05$, Fig. 4). Regarding the flexural moduli of materials, ENA showed the greatest value and HC the lowest, as seen in the Vickers hardness experiments ($p < 0.05$). A significant positive correlation was found between flexural modulus and filler content ($p < 0.05$, $r = 0.877$). The results suggested that the flexural moduli were influenced by the filler contents and the filler compositions; ENA, which had the greatest filler content and contained feldspar glass with a large amount of Al_2O_3 , revealed a greater flexural modulus value than the other resin composites containing quartz as filler. Masouras *et al.*⁴³⁾ found that the elastic moduli of some resin composite materials were positively correlated to their filler contents. Randolph *et al.*⁴⁴⁾ and Mainjot *et al.*⁴⁵⁾ reported that modern resin composites with high inorganic filler content were associated with higher flexural modulus and strength. The results of the present study supported these previous reports.

The flexural strengths did not correlate to the filler content, and are considered to be affected more by the composition of the resin matrix than by the filler content and composition.

Resin composite materials were reported to deteriorate due to residual stress yield³⁰⁾. Since the polymerization of resin matrix components continues after setting, both polymerization shrinkage and shrinkage by cooling water during thermal cycle process can cause residual stress in the resin matrix. In addition, because of the large difference in thermal expansion coefficient between the filler and the resin matrix, further complex residual stress is generated at the resin matrix/filler interface. These residual stresses

are considered to induce the deterioration of materials that fracture in a brittle manner.

Ferracane *et al.*⁴⁶⁾ reported that water penetration into the matrix generates softening of the polymerized matrix when immersing resin composites in water. Druck *et al.*⁴⁷⁾ reported that water immersion led to hydrolysis of silane coupling agents at the interface between filler and matrix, and that hydrolysis notably occurred in resin composites using zirconia fillers. In HC, KZR and EST, which use zirconium silicate, $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-ZrO}_2$, and zirconia, respectively, as fillers, the silane coupling agents at the matrix/filler interface could be hydrolyzed by water immersion, resulting in a decrease in flexural strength by reducing the bonding force between filler and matrix. AVE, which does not use zirconia as a filler component, employs UDMA as a matrix component. Since UDMA is rather hydrophilic compared to Bis-MEPP, as mentioned above, the flexural strength of AVE was possibly lower due to enhanced hydrolysis of the silane coupling agent at the matrix/filler interface by water penetration through the matrix.

In fracture surface observations, all of the resin blocks revealed somewhat rough surfaces fractured in a brittle manner. It could be speculated from this finding that the resin blocks examined absorbed less water in the matrix. The result that, except for CS300 and ENA, the resin blocks showed decreased flexural strength after 10,000 thermal cycles compared to storage in air at room temperature, would be due to an effect of hydrolysis of the coupling agents at the matrix/filler interface rather than direct water absorption in the matrix resin.

Under the present experimental conditions, the resin composite block CS300 exhibited the greatest flexural strength among all storage conditions and less deterioration of strength by thermal cycling and water immersion among the resin composite blocks examined. None of the CAD/CAM resin blocks showed significant differences in flexural strength between storage in water and thermal cycling ($p>0.05$). Therefore, the experimental results rejected the proposed hypothesis.

CONCLUSIONS

Under the present experimental conditions, the following conclusions were drawn:

1. The flexural strengths differed among the resin composite materials for CAD/CAM due to differences in matrix and/or filler compositions.
2. The difference in storage condition between water immersion and thermal cycling did not affect the flexural strengths of resin composite materials for CAD/CAM examined in this study.
3. The resin composite block CS300 showed the greatest flexural strength for all storage conditions and less deterioration of strength by thermal cycling and water immersion among the resin composite blocks.

CONFLICT OF INTEREST

No conflict of interest exists for any of the authors of this paper. This research received no specific grant from any funding agency in the public, commercial or not-for-profit sectors.

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