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Effect of Light Intensity on Polymerization of Light-cured Composite Resins

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The depths of cure and the distributions of degree of conversion (DC), polymerization conversion (PC) and percent pendant double bonds (PDB) of light-cured composite resins cured under various intensities of light were investigated. When the total amount of exposure, represented by the product of the light intensity and the irradiation time, was kept constant, each of the depth of cure and the distributions of DC, PC and PDB were the same for each material regardless of the light intensity and irradiation time. The depth of cure could be expressed as a logarithmic function of the total amount of exposure. From regression analysis, the attenuation coefficient and the critical total amount of exposure capable of initiating polymerization of each composite resin could be determined.

Key words: Light-cured composite resin, Light intensity, Depth of cure

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INTRODUCTION

The intensity of most light sources reduces with time during radiation. There are many variables that can influence the intensity of light during exposure and affect the properties of the set restorative material[1−4]. Light-cured materials have a limited depth of cure[1,5,6] since light of a certain intensity is required to activated polymerization[5,7]. The purpose of this study was to investigate the influence of light intensity on polymerization of composite resins. The depth of cure and the distributions of degree of conversion (DC), polymerization conversion (PC) and percent pendant double bonds (PDB) were measured. PC is the proportion of double bonds at which monomer molecules are converted into polymers.

MATERIALS AND METHODS

Visible light-cured composites

The three dental composite resins used were abbreviated as Z100*, SP** and CL#. These were commercially available visible light-cured composites based on BisGMA-TEGDMA. The light source## used for this research was equipped with a light tip 12 mm in diameter. This unit was chosen because of the little reduction of light intensity with time, as shown in Fig. 1.

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* Z100, 3M Co., Minn., USA

** Silux Plus, 3M, Co., Minn., USA

\# Clearfil Photo Posterior, Kuraray Co., Ltd., Okayama, Japan

\#\# New light VLII, GC Co., Tokyo, Japan

Light intensity measurement

The light intensity was measured with a lux meter@. A light-tight black cap with an aperture 4.0 mm in diameter at its center was placed on the measuring surface of the lux meter. This aperture size was the same as the cavity diameter of the stainless steel mold used for preparation of specimens. The light tip and the lux meter were set so that the detector head of lux meter was parallel to and under the center of the light tip. When the exit window of the light tip made contact with the black cap of the detector head, the value of light intensity through the 4.0-mm aperture was standard, abbreviated as I. The light intensity at the surface of the lux meter could be reduced to one-half, one-quarter or one-eighth of the standard value by varying the distance between the tip of the activating device and the lux meter. The distance was measured with a slide caliper. This pre-measured distance was applied when the composite resin was polymerized.

As shown in Fig. 1, the light intensity of this light unit was approximately constant after 30s from the beginning of radiation. Even though it was switched off and instantly on repeatedly, the intensity immediately matched that at the time the unit was switched off. Consequently, for each test section, exposure light was taken as the incident light intensity after 30s from the time that the lamp was switched on. The irradiation periods were then applied after 30 s exposure.

Depth of cure measurement

A stainless steel split mold with a cylindrical cavity 4 mm in diameter and 8 mm in depth was used. The cavity was slightly overfilled with the composite. The material was covered with polyester strips@@ (51 μm in thickness) and was subsequently evened with a glass plate, which was then removed. The tip distance to the surface of the strips was that pre-

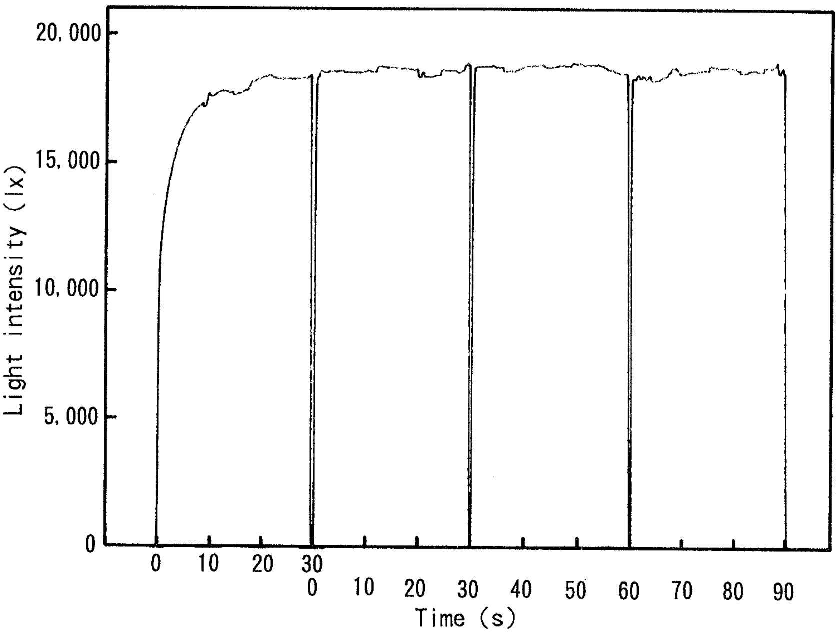

Fig. 1 Variation of the light intensity with irradiation time for the light source used.

@ Illuminance meter T-1H, Minolta Camera Co., Ltd., Tokyo, Japan

@@ Polyester matrix tape, 3M Co., Minn., USA measured for the various intensities of light, and the center of the tip was fitted to the center of the resin. Thereafter, the material was exposed to visible light at various intensities for either 10, 20, 40 or 80 seconds. Samples were stored at 23°C in the dark. After 180 s from completion of exposure, the specimen was removed from the mold and the uncured material was gently removed with a plastic spatula. The height of the cylinder of cured material was measured with a micrometer. Five replications were made for each test condition.

Evaluation of degree of conversion, polymerization conversion and percent pendant double bonds

A method similar to that used to prepare the specimens has been described previously[8,9]. The composites were cured as described above and stored at 37°C in the dark for 23h without being removed from the molds. Subsequently, a thin composite film about 0.2 mm in thickness was sectioned perpendicular to the irradiated surface with a cutting machine under a water stream and polished in water. The thickness of specimen finally reached about 0.15 mm. Three specimens were prepared for each condition.

After 24h from the end of the irradiation period, infrared (IR) spectra of microareas ($100 \times 100\ \mu m^2$) were measured at 0.2-mm intervals along the midline from the irradiated surface to the bottom of the specimen using a Fourier transformed infrared spectrometer[\$] equipped with a microscopic unit and an automatic mapping stage. IR spectra were measured in order to evaluate the quantities of carbon-carbon double bonds remaining in the specimens.

Specimens were immersed in methanol to extract residual monomers. After elutable components were completely extracted, the specimen was freed of solvent. Then IR measurement were repeated at the same location.

In these IR spectra, two absorbance peaks appeared in the range from 1600-1650 cm^{-1}. The peak at 1637 cm^{-1} was assigned to C=C stretching vibrations of methacryloyl groups and that at 1608 cm^{-1} to stretching vibrations of aromatic rings. An appropriate baseline was drawn, and these two absorption peaks were separated into two Lorentzian curves. The intensities of absorption were determined by the areas under the peaks.

DC, PC and PDB were calculated using the following equations:

$$DC\ (\%) = 100 - \{(bc/ad) \times 100\}$$

$$\begin{aligned} PC\ (\%) &= 100 - [\{(c-e)/c\} \times \{(bc/ad) \times 100\}] \\ &= 100 - [\{(c-e)/c\} \times (100 - DC)] \end{aligned}$$

$$\begin{aligned} PDB\ (\%) &= (e/c) \times \{(bc/ad) \times 100\} \\ &= (e/c) \times (100 - DC) \end{aligned}$$

Where the intensities a-e are defined as follows:

a: C=C absorbance peak at 1637cm^{-1} of resin pastes;
b: aromatic absorbance peak at 1608cm^{-1} of resin pastes;
c: C=C absorbance peak at 1637cm^{-1} of cured resin before extraction;
d: aromatic absorbance peak at 1608cm^{-1} of cured resin before extraction; and,
e: C=C absorbance peak at 1637cm^{-1} of cured resin after extraction.

[\$] JIR-100, Japanese Electron Optics Laboratory Co., Ltd., Tokyo, Japan

RESULTS

Light intensity

Fig. 2 shows the relationship between the tip distance to the lux meter and the light intensity. For the light intensity at the detector head of the lux meter, the inverse-square law held.

Depth of cure

The depths of cure for the investigated resins irradiated with the various light intensities and irradiation times are shown in Tables 1-3. Table 1 is for Z100, Table 2 is for SP and Table

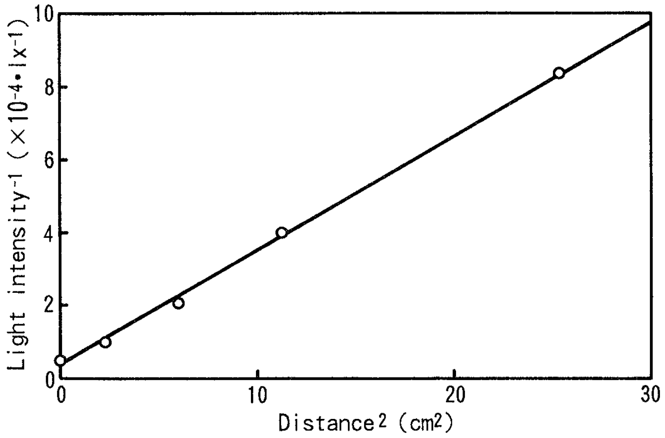

Fig. 2 Relationship between the tip distance to the lux meter and the light intensity.

Table 1 Variation of the depth of cure for Z100

Irradiation time (s)	Depth of cure (mm)			
	Intensity of light			
	I	I/2	I/4	I/8
10	4.10^{a} (0.10)	3.16^{b} (0.10)	2.42^{c} (0.10)	1.83 (0.06)
20	5.07^{d} (0.06)	4.08^{a} (0.06)	3.09^{b} (0.02)	2.52^{c} (0.01)
40	6.08^{e} (0.00)	5.04^{d} (0.08)	3.92^{a} (0.03)	3.27^{b} (0.10)
80	6.90 (0.02)	6.00^{e} (0.04)	4.99^{d} (0.02)	4.13^{a} (0.11)

numbers in parentheses : standard deviation
I : standardized intensity of light
a, b, c, d, e : the same amount of irradiation, respectively

Table 2 Variation of the depth of cure for SP

Irradiation time (s)	Depth of cure (mm)			
	Intensity of light			
	I	I/2	I/4	I/8
10	2.93^{a} (0.06)	2.35^{b} (0.01)	1.79^{c} (0.04)	1.36 (0.01)
20	3.47^{d} (0.03)	2.89^{a} (0.06)	2.30^{b} (0.03)	1.86^{c} (0.03)
40	4.06^{e} (0.04)	3.38^{d} (0.07)	2.74^{a} (0.01)	2.34^{b} (0.03)
80	4.59 (0.08)	3.94^{e} (0.06)	3.24^{d} (0.04)	2.81^{a} (0.03)

numbers in parentheses : standard deviation
I : standardized intensity of light
a, b, c, d, e : the same amount of irradiation, respectively

3 is for CL. Letters adjunct to the results indicate the same total amount of exposure. The total amount of exposure is represented by the product of the light intensity and the irradiation time. Each depth of cure was the same for each material regardless of the light intensity and irradiation time when the total amount of exposure was kept constant.

DC, PC and PDB

Fig. 3 shows the DC as a function of distance from the exposed surface for each irradiaion condition. Fig. 4 shows the PC and Fig. 5 shows the PDB. The total amount of exposure was constant at 10 • I. The destributions of DC, PC and PDB coincided with those obtained for different conditions of irradiation regardless of the light intensity and irradiation time when the total amount of exposure was kept constant.

DISCUSSION

When the total amount of exposure was kept constant, the cured composites had not only the

Table 3 Variation of the depth of cure for CL

Irradiation time (s)	Depth of cure (mm)			
	Intensity of light			
	I	I/2	I/4	I/8
10	4.29^{a} (0.03)	3.54^{b} (0.02)	2.76^{c} (0.01)	2.10 (0.01)
20	5.27^{d} (0.01)	4.30^{a} (0.02)	3.43^{b} (0.05)	2.76^{c} (0.03)
40	6.17^{e} (0.05)	5.22^{d} (0.02)	4.20^{a} (0.03)	3.37^{b} (0.06)
80	7.01 (0.11)	6.18^{e} (0.03)	5.00^{d} (0.08)	4.07^{a} (0.01)

numbers in parentheses: standard deviation
I: standardized intensity of light
a, b, c, d, e: the same amount of irradiation, respectively

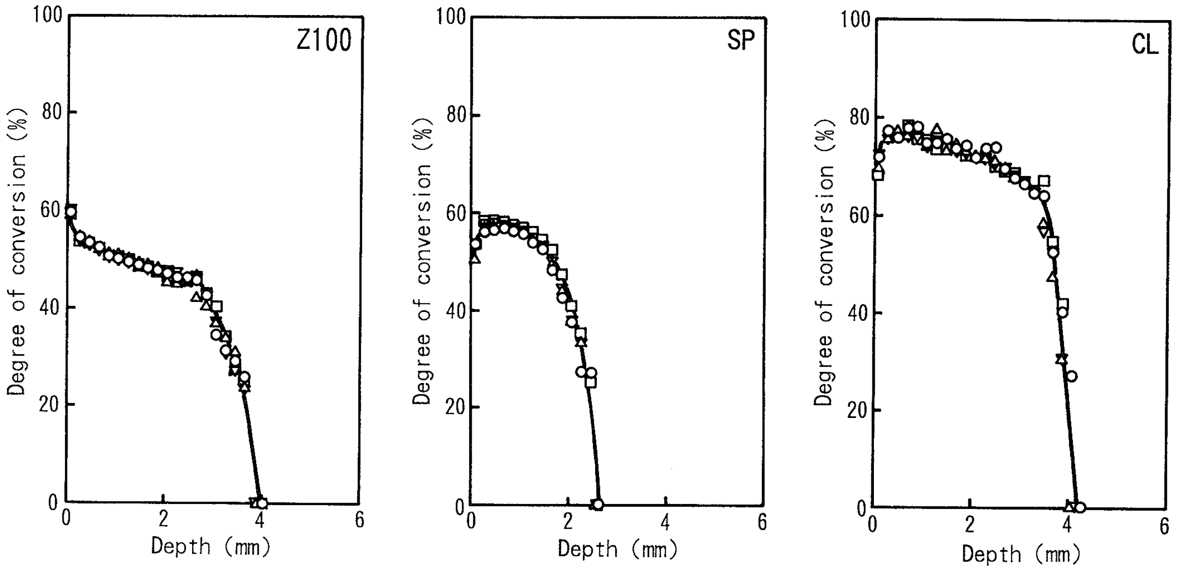

Fig. 3 Variation of the degree of conversion with depth.
irradiated for ○: I × 10 s; △: (I/2) × 20 s;
▽: (I/4) × 40 s; □: (I/8) × 80 s
I: standardized intensity of light

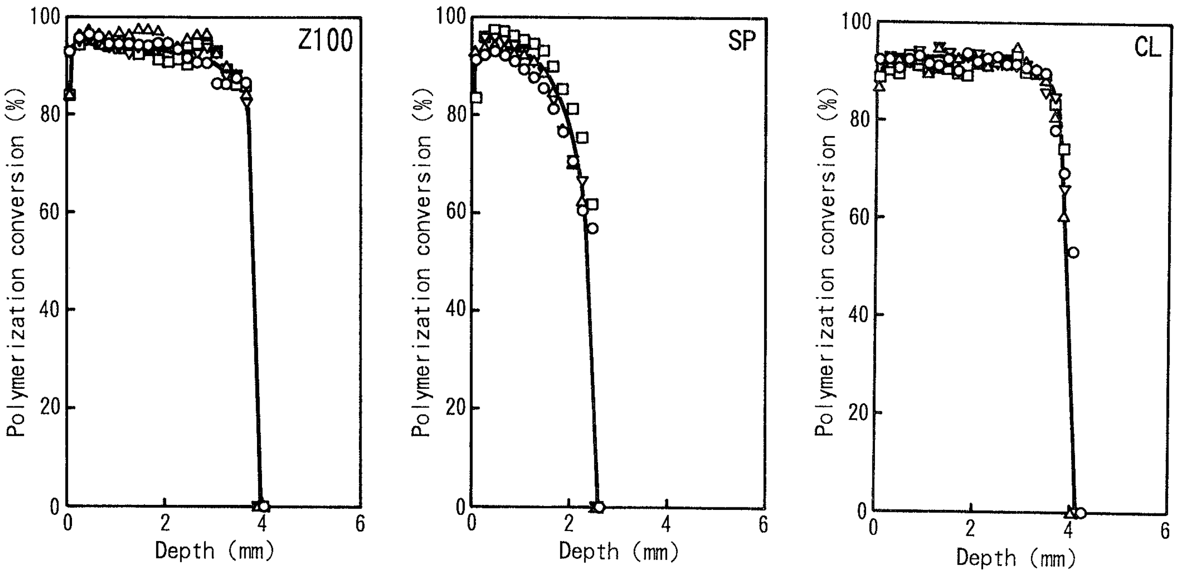

Fig. 4 Variation of the polymerization conversion with depth.
irradiated for ○ : I × 10 s ; △ : (I/2) × 20 s ;
▽ : (I/4) × 40 s ; □ : (I/8) × 80 s
I : standardized intensity of light

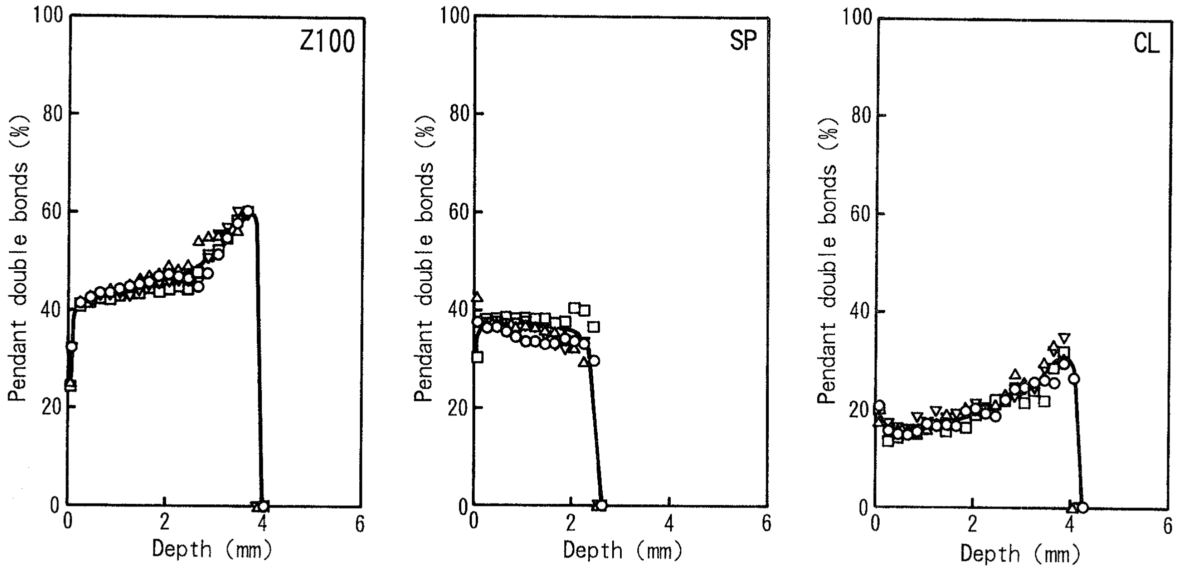

Fig. 5 Variation of the percent pendant double bonds with depth.
irradiated for ○ : I × 10 s ; △ : (I/2) × 20 s ;
▽ : (I/4) × 40 s ; □ : (I/8) × 80 s
I : standardized intensity of light

same depths of cure, but the same distributions of DC, PC and PDB. This finding is in accordance with those reported previously by Cook[5,7]. Cook derived the following relationship for the depth of cure (*L*) on the basis of analysis of the kinetics involved in the photopolymerization of dimethacrylate resins and on the assumption that the Lambert's law holds and polymerization does not occur until all of the inhibitor has been consumed by the radicals

Table 4 Results of statistical analysis

Material	ε (mm^{-1})	E (lx • s)	γ
Z100	0.345	7100	0.995
SP	0.557	4260	0.996
CL	0.359	4800	0.994

ε : attenuation coefficient
E : critical total amount of exposure
γ : correlation coefficient

generated in the immediate vicinity.

$$L=\frac{1}{\varepsilon}\log t+\frac{1}{\varepsilon}\log\left(\frac{2.303K\phi I_0\varepsilon_s S_0}{\chi_0}\right) \tag{1}$$

Where t is the irradiation time, I_0 is the intensity of the incident light at the surface of the material, ε is the attenuation coefficient of the composite resin, ϕ is the quantum yield, ε_s is the molar absorptivity, S_0 and χ_0 are the concentration of the photoinitiator and inhibitor, respectively, and K is a constant. Since ϕ, ε_s, χ_0 and S_0 are inherent in the composite resin, Equation(1) can be simplified to the following form :

$$L=\frac{1}{\varepsilon}\log(A\cdot I_0\cdot t) \tag{2}$$

Where $A=(2.303K\phi\varepsilon_s S_0)/\chi_0$, *i. e.*, is a constant. By virtue of Equation (2), it follows that L depends on $I_0\cdot t$, that is, the total amount of exposure. Equation (2) thereby shows that the depth of cure is dependent on the product of light intensity and irradiation time.

Assuming that the change of the attenuation coefficient during polymerization and the difference of the attenuation coefficient which was due to the differences of the cross-linked polymers in a cured composite were negligibly small, the intensity of light transmitted by the cured composite is defined by the Equation (3) :

$$I=I_0\cdot 10^{-\varepsilon L} \tag{3}$$

by the applying Lambert's law. For this law, ε is an absorption coefficient but is referred to as an attenuation coefficient because the heterogeneity in dental composite resin influences the optical properties. Light is not only absorbed by matrix resin and fillers but also scattered at the interfaces between the filler and resin phases. When the sample is irradiated for time-t, the amount of light transmitted is defined by the Equation (4) :

$$E=I\cdot t=I_0\cdot t\cdot 10^{-\varepsilon L} \tag{4}$$

E is defined as the critical total amount of exposure and represents a minimum total amount of exposure required to activate polymerization. Equation (4) can be transformed :

$$L=\{\log(I_0\cdot t)-\log E\}/\varepsilon \tag{5}$$

Equation (5) indicates that a linear relationship exists between the depth of cure and the logarithm of total amount of exposure. The attenuation coefficient and the critical total amount of exposure are defined by this equation.

Table 4 lists the results of regression analysis for each material. The attenuation coefficient of SP was larger than those of other materials, indicating that SP attenuates light to a greater extent than Z100 and CL. When the critical total amount of exposure was reached for each sample, the surfaces of Z100 and CL were slightly cured but crumbled during measurement of depth of cure, and SP was not cured, validating the above inference.

CONCLUSION

The depths of cure and the distributions of degree of conversion (DC), polymerization conversion (PC) and percent pendant double bonds (PDB) of light-cured composite resins cured at various light intensities were measured. The influence of light intensity and irradiation time on polymerization of composite resins were investigated. Different intensities of light were obtained by varying the distance between the exit window of the light activation unit and the surface of the specimen.

When the total amount of exposure, represented by the product of the intensity of light and the irradiation time, was kept constant, the depths of cure and the distributions of DC, PC and PDB were the same for each material regardless of the light intensity and irradiation time. The depth of cure could be expressed as the logarithmic function of the total amount of exposure, and the attenuation coefficient and the critical total amount of exposure, which could initiate polymerization of each composite resin on the light activation unit used, could be determined from the regression equation.

ACKNOWLEDGMENT

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HEMAプライマーへの各種銅塩の添加効果を検討した．ウシ象牙質表面を10％リン酸水溶液で処理した後，銅塩を含む35％HEMA水溶液プライマーを塗布し，MMA-TBBレジンでアクリル棒を接着した．銅塩の種類・濃度を変えて接着したところ，プライマー中の銅塩の最適濃度は0.5-3.0μmol/gであった．検討した8種類の銅塩の中で，硫酸，メタクリル酸，メタクリロイルオキシエチルフタル酸，メタクリロイルオキシエチルコハク酸の銅塩が特に有効であり，14.2-16.1MPaの平均接着強さおよび最低の接着強さの平均値として9.9-11.7MPaが得られた．これらの接着強さは，市販の4-META/MMA-TBBレジン接着システムで得られる値よりも大きかった．ある種の銅塩存在下で重合したポリMMAの分子量が増加したことから，いくつかの銅塩ではMMA-TBBレジンに対するその分子量増加効果によって接着強さが向上したことが示唆された．

光重合コンポジットレジンの重合性に及ぼす光強度の影響

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照射距離を変化させて得られた種々の光強度で光重合コンポジットレジンを重合し，硬化深さを初め硬化体内部の反応率，重合率およびペンダント二重結合量の深さ方向に対する分布を測定し，これら重合特性に及ぼす光強度の影響を調べた．照射する照射量（光強度と照射時間の積）を一定にすると光強度，照射時間に関わらず硬化深さはほぼ一定の値を示し，硬化体内部の反応率，重合率およびペンダント二重結合量の各分布もよく一致した．硬化深さは照射量の対数で表され，この直線回帰式から使用した光照射器に対する材料の吸光係数および材料が硬化するのに必要な照射量（臨界照射量）を求めることができた．

チタン鋳造におけるガスパージの効果

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横山有紀，柿川　宏，小園凱夫

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溶解雰囲気と鋳型内のガス清浄化を目的としたダブルガスパージを特徴として開発されたチタン専用鋳造機を用い，ダブルパージの効果について検討することを目的に，溶解室雰囲気中ならびに純チタン鋳造体中の酸素濃度分析，鋳造体の硬さ試験および鋳造性試験を行った．ダブルパージを採用することで溶解雰囲気の極低酸素化が可能となり，純チタン鋳造体中の酸素量は他機種に比べて著しく低くなった．さらにチタン鋳造体表面の硬さもパージを採用することで減少した．しかし，鋳造性に対しては影響はみられなかった．以上の結果から，ダブルパージを採用して開発されたチタン鋳造機により酸素の混入を抑えたチタン鋳造体を作製できることが示された．