Technical note

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Hardness and Fracture Toughness of Commercial Core Composite Resins

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One important mechanical property of core composite resins is fracture toughness, K_{IC}, which expresses serviceability in the oral cavity, such as the resistance to marginal fracture. K_{IC} values of eight commercial core composite resins were examined by the single-edge notched-beam (S. E. N. B.) method. Two composites containing about 80wt% Si_3N_4 fillers exhibited both the highest K_{IC} value of around 2.0 MN•m$^{-3/2}$ and the highest hardness value. The other six composites containing 66 to 86wt% SiO_2-based fillers had K_{IC} values of around 1.2 to 2.0 MN•m$^{-3/2}$, and there was a tendency toward higher K_{IC} values as hardness increased. It was speculated that the clinical acceptability of core composite resins could be broadened, if dental clinicians selected composites with higher K_{IC} values.

Key words : Core composite resin, Hardness, Fracture toughness

INTRODUCTION

Endodontically treated teeth are usually re-constructed with post and core restorations[1−3]. Although metal cores are generally recognized as superior in durability and reliability, there exists a wide-spread trend toward increased usage of composite resin cores due to ease in handling and shorter chair time[3]. The drawback of composite resin cores is, however, their weakness, leading to bulk fracture and interfacial separations between composite and metal screw and between composite and remaining teeth[4,5].

Fracture toughness, K_{IC}, provides a quantitative measure of strain-energy absorption ability during structural deformation, or a measure of the resistance to brittle fracture[6]. The K_{IC} of composite resins is important in evaluating their usefulness. Although the K_{IC} of direct filling[7−9] and indirect[10] composites have been measured, there have been few studies in which the K_{IC} of core composites was examined.

This study, therefore, determined the K_{IC} values of eight commercial core composite resins and two core cements (as reference) by the single-edge notched-beam method. Knoop and Micro Brinell hardness were also measured. Furthermore, the fillers in the composites were investigated by thermogravimetric thermal analysis and X-ray diffraction.

MATERIALS AND METHODS

Materials

Table 1 lists the eight core composite resins and two glass ionomer-based cements used for core build-up that were examined in this study. The six chemically-cured composites and two

Table 1 Eight commercial core composite resins and two core glass ionomer-based cements

Type	Sample code
Chemically cured core composite	MC[@1]
Chemically cured core composite	CL[@2]
Chemically cured core composite	CC[@3]
Chemically cured core composite	CM[@4]
Chemically cured core composite	BC[@5]
Chemically cured core composite	HD[@6]
Visible-light-cured core composite	PC[@7]
Visible-light-cured core composite	BL[@8]
Glass ionomer-based core cement	MM[@9]
Glass ionomer-based core cement	CS[@10]

[@1] Microrest Core, GC Co., Tokyo, Japan (Lot No.: base=180991, catalyst=191211)
[@2] Corelite, GC Co., Tokyo, Japan (Lot No.: base=300511, catalyst=071291)
[@3] Clearfil Core, Kuraray Co., Osaka, Japan (Lot No.: universal=RU-1846, catalyst=RC-1743)
[@4] Core Max II, Sankin Kougyou Co., Tochigi, Japan (Lot No.: powder=229, liquid=218)
[@5] Belfeel Core, Kanebo Co., Tokyo, Japan (Lot No.: universal=1NJ9N, catalyst=1NJ9N)
[@6] Belfeel Core HD, Kanebo Co., Tokyo, Japan (Lot No.: universal=19B10, catalyst=10B1D)
[@7] Clearfil Photo Core, Kuraray Co., Osaka, Japan (Lot No.: 1154)
[@8] Blue Core, Teledyne Getz Co., Ill. U. S. A. (Lot No.: 76070)
[@9] Miraclemix, GC Co., Tokyo, Japan (Lot No.: powder=010812, liquid=271111)
[@10] Chelon-silver, ESPE Co., Germany (Lot No.: powder=0120, liquid=0026)

core cements were mixed with plastic spatulas on mixing pads, following the manufacturers' instructions, and, successively, filled into teflon molds, 4mm high×3mm wide×20mm long with two open longitudinal surfaces which were covered with slide glasses. The two visible-light-cured composites were directly condensed into the similar teflon molds, the two open surfaces of which were covered by plastic strips, and photocured with a visible light source*. The tip of the light source (5mm in diameter) was first located on one end of the specimen, and photo-cured for 40 s. Following the first photo-cure, the tip of the light source was moved 2.5mm toward the other end of the specimen and irradiated with visible-light. This step-wise photo-curing was repeated until the tip of the light source reached the other end of the specimen. The plate specimen was then turned upside-down, and successively photo-cured from the reverse side. After setting, surfaces of all specimens were metallographically polished to a final finish with 1500 grit sand paper. For each material, five samples were made. After determination of the K_{IC}, the plate specimens, broken into two, were used for hardness measurements.

* Quick Light, Model VL-1, Morita Co., Kyoto, Japan.

Fracture toughness measurements

Fig. 1 shows the sample configuration and loading condition for three-point-bending test, by which K_{IC} values were determined. A notch, about 0.3mm wide and 1.6mm deep, was carved at the center of the long axis of the specimen, using a dental diamond disk* driven by a dental micro-engine#. The flexure tests were performed in a universal testing machine## at a crosshead speed of 0.5mm/min. K_{IC} was calculated by the following formula[6].

$$K_{IC}=3PL/2BW^2 \cdot a^{1/2} \cdot \{1.93-3.07\ (a/W)+14.53\ (a/W)^2-25.11\ (a/W)^3+25.80\ (a/W)^4\}$$

Where a=depth of the notch (mm)

W=height of the specimen (mm)

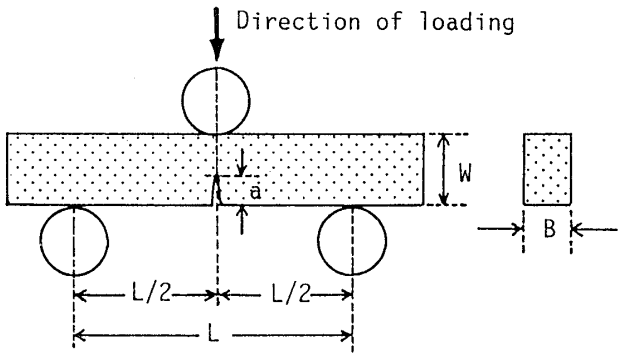
B=width of the specimen (mm)

L=span of the specimen (mm)

P=breaking load (N)

Hardness measurements

Knoop hardness, Hk, was measured by indenting the surface of the specimen at a load of 9.8 N (1 kgf) for 30 s with a microhardness tester@. Micro Brinell hardness, MBH, was determined by pressing the surface of the specimen with a steel ball indenter, 5.0mm in diameter, at a load of 5 kgf for 30 s with a hardness testing machine@@. For each material, five hardness measurements were taken.



a : depth of the notch = 1.6±0.2 mm
W : height of the specimen = 4.0±0.1 mm
B : width of the specimen = 3.0±0.1 mm
L : span of the specimen = 16.0 mm

Fig. 1 Sample configuration and loading condition for three-point-bending test, which measured fracture toughness, K_{IC}, using the single-edge notched-beam method.

* Gemflex thin, Jelenco Co., N.Y., U.S.A.
\# Volver 8, Nakanishi Dental Machine Co., Tochigi, Japan
\#\# Autograph, Shimadzu Co., Kyoto, Japan
@ HMV-2000, Shimadzu Co., Kyoto, Japan
@@ Mori Testing Machine Co., Tokyo, Japan

Characterization of fillers

To determine the inorganic filler contents in set core composite resins, thermogravimetric thermal analysis (TG) was performed with a thermal analyzer[$], under the following experimental conditions: sample weight, 20±1 mg; reference material, α-alumina; TG sensitivity, 20mg; heating rate, 5°C/min; temperature range, room temperature to 800°C; and atmosphere, air under a flow of 30 ml/min N_2. The weight of the composite specimen at 575°C was recorded here as the filler content with relative accordance to international standard[11].

The composite resin paste was dissolved in acetone and centrifuged, the liquid was removed, and the residue was freeze-dried. To identify the phase of the inorganic filler, X-ray diffraction (XRD) analysis of the extracted fillers was performed with a diffractometer[$$], under the following experimental conditions: diffraction beam, Cu•K_α ray; accelerated voltage, 30 kV; current, 10 mA; scan rate (2θ/min), 2°/min; and scan range (2θ), 10 to 50°. The phase of the filler was labelled by matching with J. C. P. D. S. (Joint Committee on Powder Diffraction Standards) files[12].

RESULTS

Fig. 2 indicates the K_{IC} of eight core composites and two core cements. Three composites (CM, BC and HD) demonstrated the maximum K_{IC} at around 2.0 MN•m$^{-3/2}$, followed by those of three composites (CL, CC and PC), while two composites (MC and BL) showed the minimum K_{IC} at around 1.2 MN•m$^{-3/2}$. The two core cements (MM and CS) had much smaller K_{IC} values at around 0.6 MN•m$^{-3/2}$, one third to one half those of core composite resins.

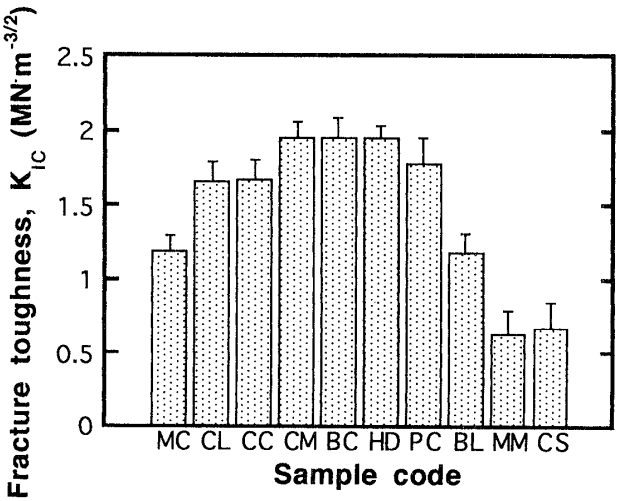


Fig. 2 Fracture toughness of eight core composite resins and two core cements.

[$] DT-30, Shimadzu Co., Kyoto, Japan
[$$] Geigerflex D-2, Rigaku Co., Tokyo, Japan

Fig. 3 shows the Hk and MBH hardness of the eight core composites and two core cements. The two hardness values were basically analogous to each other, although there were larger variations in the Hk data. One composite (HD) was the hardest with an MBH of about 68, four composites (CL, CM, BC and PC) belonged to the group with the second highest MBH score ranging from 30 to 40, while three composites (MC, CC and BL) and the two core cements (MM and CS) were in the softest group with MBH values between 20 and 30.

Table 2 summarizes the contents and main phases of fillers in eight core composite resins. The phase identification of the fillers in two composites (MC and CL), presumably labelled as SiO_2−based glass+BaO, is still under confirmation. The filler content of MC was the smallest, at around 66wt%, while those of CL and BL exceeded 80wt%. Three composites (CC, CM and PC) contained quartz fillers with concentrations of 74 to 87wt%. Two composites (BC and HD) had Si_3N_4-based fillers with contents of around 80wt%.

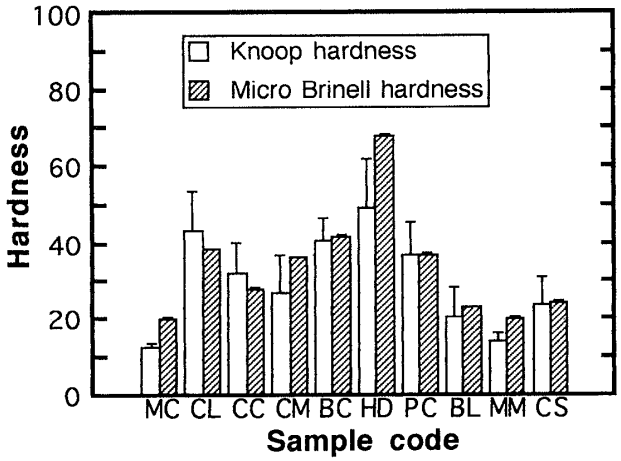


Fig. 3 Knoop hardness, Hk, and Micro Brinell hardness, MBH, of eight core composite resins and two core cements.

Table 2 Contents and main phases of fillers in eight core composite resins

Sample code	Content at 575°C (wt%)	Main phase
MC	65.64±1.13	Glass+BaO (?)
CL	80.03±1.97	Glass+BaO (?)
CC	81.35±1.78	Quartz
CM	73.66±1.04	Quartz
BC	81.45±0.07	Si_3N_4
HD	79.28±1.22	Si_3N_4
PC	86.53±0.13	Quartz
BL	84.28±0.65	Glass

DISCUSSION

The single-edge notched-beam method is a popular way to determine the K_{IC} of core composite resins[7-10]. Considering the K_{IC} values of natural enamel (at around 1.0 MN•$m^{-3/2}$)[13,14] and those of natural dentin (at around 3.0 MN•$m^{-3/2}$)[15], it was noted that K_{IC} values of commercial core composites lie between those of enamel and dentin. To reduce the likelihood of brittle fracture or marginal fracture under tension of core composites, selecting composites with a higher K_{IC} value, such as one of the two composites containing Si_3N_4 fillers or one of those containing SiO_2-based fillers with a high filler content of more than 80wt%, is recommended.

Yamauchi *et al.*[16] already reported the Hk hardness of core composite resins. Our data were consistent with theirs. Hardness of all core composites and cements examined were comparable to, or lower than, those of natural dentin (Hk of about 60). Hk hardness readings had larger data variation, compared with those of MBH hardness. This might be attributed to the fact that the indenter of the latter hit larger and more homogeneous areas including both filler and resin matrix. Although Yamauchi *et al.*[16] found a linear correlation between filler content and Hk, we could not obtain the same results. However, we noticed a weak linear correlation between MBH hardness and K_{IC} values (linear correlation coefficient, $r = 0.742$). The reason for this is not clear. It appears that not only the filler content[17] but also the resin matrix plays a significant role in determining both hardness and K_{IC}.

The two core cements had very low K_{IC} values. When selecting core cements, dental clinicians must consider advantages other than mechanical properties, such as anti-cariogenic action from the delayed release of fluoride ions or strong radiopacity from metal ingredients.

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結果，1日目では，GICへHY剤を1.5%配合することによって，接着強さを向上させることができた．経時的には，いずれの配合割合のものでも接着強さは低下し，HY剤を配合したものも，しないものでも，各配合割合間での差は認められなくなった．フッ化ジアンミン銀の併用によってGICの接着強さは向上し，HY 0およびHY 1.5では，経時的な接着強さの低下も防止できた．

微小圧子圧入法による歯科用陶材の疲れ寿命の推定

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Radial Crackの，水中及び大気中での，成長速度を測定することにより，炉冷，空冷した歯科焼付用歯冠色陶材の疲れ寿命を推定した．測定したCrackはVickers圧子を9.8 Nの荷重で陶材表面に圧入して導入した．応力腐食係数nはcrack長さを5ヶ月まで測定して求めた。計算されたn値から，陶材の使用応力の評価を試みた．その結果，炉冷後，水中保存したガラス質及び長石系陶材の使用応力は，寿命が10年以上になるためには，その破壊強さの47，69%であると計算された．風冷強化（空冷）した陶材では，それぞれ58，67%であった．以上のことから，風冷により導入された残留応力はガラス質陶材の使用寿命を伸ばすのには有効であるが，長石系陶材では有意な違いが生じないことが明らかになった．

直接的な観察法によるコンポジットレジンの口腔内磨耗像

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本研究の目的は，コンポジットレジンの磨耗を口腔内で評価するための簡便な方法を開発し，磨耗の進行過程で果たすフィラーの役割を明らかにすることである．2種類の異なるフィラーシステム，81 wt%の粉砕フィラー（従来型）及び73 wt%の有機複合フィラー（MFR型），を有する試作光重合型コンポジットレジンが用いられた．なお，試作レジンには，Bis-GMA（50 wt%）とTEGDMA（50 wt%）からなるレジンモノマーが用いられた．Au-Pd合金製クラウンの咬合接触部（OCA）と非接触部（CFA）に設けた直径2 mmの円筒形窩洞に試作レジンを充塡し，ボランティアの口腔内にクラウンを仮着した．クラウンは1月毎に撤去され，SEMによる連続的観察が行われた．その結果，新しく開発された方法は，コンポジットレジンの口腔内磨耗パターンの観察に有用なことが明かとなった．また，異なるフイラーシステムを有する試作コンポジットレジンは，著しく異なる口腔内磨耗パターンを示した．

市販コアー用コンポジットレジンの硬さと破壊靱性値測定

宮脇博正，平　雅之，豊岡博夫，若狭邦男，山木昌雄

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8種類の市販コアー用コンポジットレジンと2種類の市販コアー用グラスアイオノマーセメントについて，片側切り欠き試験片（S. E. N. B. 法）による破壊靱性値（K_{IC}）測定を行った．また，Knoop硬さとMicro Brinell

硬さ測定を行った．その結果，Si_3N_4 を 80 wt%程度含有する 2 つのコンポジットレジンが最大の K_{IC}値（＝約 2.0 #$MNm^{-3/2}$）と最大の硬さを有することが判明した．他の 6 種類のコンポジットレジンは 66 から 86 #wt%の SiO_2 系フィラーを含有し，硬い試料程大きな K_{IC}値（1.2 から 2.0 $MNm^{-3/2}$）を有する傾向を示した．2 種類のコアー用セメントの K_{IC} 値は，コアー用コンポジットレジンの半分以下であった．これらの材料を臨床応用する際には，縁端部の破折等を防止する目的で，高めの K_{IC} 値を有する材料を選択すべきことが示唆された．

歯科用接着剤含有 Methacryloyloxydecyl Dihydrogen Phosphate (MDP)とりん脂質リポソーム相互作用の NMR スペクトロスコピー研究（続報）

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MDP がりん脂質リポソームとどのように相互作用するかを見出すために，DPPC/MDP リポソーム及びジラウロイルフォスファチジルエタノールアミン（DLEA)/コレステロール（CS)/MDP リポソーム両系の NMR ケミカルシフトの変化を pH 7.0 で研究した．この結果 DPPC 系でリポソームの流動化を伴って MDP の ^1H 遮蔽が見られた．DLEA/CS 系で MDP の ^1H シグナルは飽和して観測できなかった．一般にイオン化する化合物はりん脂質 2 重層より成る生体膜を透過しにくい．本研究から，pH 7.0 でイオン化した MDP のリポソーム相互作用が大きいことが明らかになった．これは MDP 分子中のデカメチレングループとりん脂質のアシル鎖との疎水性相互作用に起因すると推察された．