Dental Materials Journal 2009; 28(1): 102—112

Technical Report

Examination of composite resins with electron microscopy, microhardness tester and energy dispersive X-ray microanalyzer

Rogelio José SCOUGALL-VILCHIS[1], Yasuaki HOTTA[2], Masato HOTTA[1], Taizo IDONO[1] and Kohji YAMAMOTO[1]

[1]*Department of Operative Dentistry, Division of Oral Functional Science and Rehabilitation, School of Dentistry, Asahi University, 1851 Hozumi, Mizuho City, Gifu 501-0296, Japan*
[2]*Central Research, Institute of Oral Science, School of Dentistry, Asahi University, 1851 Hozumi, Mizuho City, Gifu 501-0296, Japan*
Corresponding author, Rogelio José SCOUGALL-VILCHIS; E-mail: rogelio_scougall@hotmail.com

This study was conducted to examine the ultrastructures of eight recently improved light-cure restorative composite resins with scanning and transmission electron microscopes (SEM and TEM). Additionally, Vickers hardness, volume/weight fraction of filler, and chemical composition were analyzed. Composite resins selected for evaluation were Beautifil II, Clearfil AP-X, Clearfil Majesty, Estelite Σ, Filtek Supreme, Filtek Z250, Solare, and Synergy. SEM and TEM images revealed a great diversity in ultrastructure, and Vickers hardness test showed significant differences amongst all the composite resins (except between Clearfil Majesty and Estelite Σ, and between Filtek Supreme and Filtek Z250). By means of EDX, similar elements such as C, O, and Si were detected, but the concentration was different in every composite resin. Results obtained in this study served to validate that the methods employed in this study — SEM and TEM at high magnification — were useful in examining the ultrastructures of composite resins. It was also found that the ultrastructure, size of filler particles, volume/weight fraction of filler, and chemical composition of the composite resins had an effect on Vickers hardness. Given the great diversity of ultrastructures amongst the composite resins, which stemmed from the different revolutionary technologies used to manufacture them, further studies are warranted in the search of clinical applications that optimally match the differing properties of these materials.

Key words: Ultrastructure, Microhardness, X-ray microanalysis

Received Apr 3, 2008: Accepted Jun 16, 2008

INTRODUCTION

The goal of minimally invasive dentistry is to conserve sound tooth structure during the preparation and restorative phases. In parallel with the widespread acceptance and adoption of minimally invasive dentistry is the proliferation of adhesive dentistry techniques. On the development of restorative materials for adhesive procedures, the introduction of composite resins to the dental profession by Bowen has since generated much discussion from clinicians, researchers, and manufacturers[1]. Upping the ante is an increasing need for esthetic tooth-colored restorative materials, and this trend has brought about a dizzying galore of esthetic composite resins into the dental market[2].

Composite resins have proven to be predictable and reliable restorative materials for the treatment of traumatized dentin or dental caries in the anterior region[3,4]. Recent longevity data on resin-based restorations have shown that the quality and longevity of these materials have increased[5]. Against this background, they have been suggested as substitutes for amalgam to restore posterior teeth because of these advantages: they offer improved esthetics and they do not contain mercury[3-5]. On this note, it has been stated that 21st century ultrafine compact-filled composite resins may be the materials of choice for restoring posterior cavities[2]. Therefore currently, resin composites are undisputedly one of the most widely used materials in restorative dentistry[6].

Basically, a dental composite consists of four major components: an organic polymer matrix, inorganic filler particles, a coupling agent, and an initiator-accelerator system. The resin forms the matrix of the composite material, binding the individual filler particles together through the coupling agent. The most commonly used monomer is bisphenol-A-glycidyl methacrylate (Bis-GMA) diluted with triethyleneglycol dimethacrylate (TEGDMA)[4,7]. Fillers are added to the polymeric part of the composite to provide strengthening, increase stiffness, reduce dimensional changes, and improve handling. Currently, most of the composites are filled with silicate particles based on oxides of barium, strontium, zinc, aluminum, or zirconium[8].

Optimum mechanical properties could be achieved by incorporating high concentrations of filler particles of varied sizes into the resin[2,8,9]. Several classifications based on the average size of filler particles have been suggested[1]. However, despite the wide variety, there is no superiority of any specific filler because every type of filler reveals

its advantages and disadvantages during clinical use[8]. Besides, it is believed that one of the main factors in reinforcing composites is a suitable bond between the fillers and the resin matrix[4,6].

The microstructure and properties of dental resin composites have been extensively studied[9]. As for the filler particles, the scanning electron microscope (SEM) has been successfully used to evaluated their numbers, shapes, and sizes[3,10]. However, to the best of our knowledge, there are no comparative studies of newly developed restorative composite resins using electron microscopy to examine their ultrastructures. The term 'ultrastructure' is used to describe the level of organization that can be seen only with an electron microscope. A recent study of orthodontic composites showed a great diversity in ultrastructure, chemical composition, and microhardness[11].

With a view to providing complementary information to the study of composite resins, this research was conducted to examine the ultrastructures of eight recently improved light-cure restorative composite resins using SEM and transmission electron microscopy (TEM). In addition, Vickers hardness, volume/weight fraction of filler, and chemical composition were also measured.

MATERIALS AND METHODS

The eight light-cure restorative composite resins evaluated in this study are listed in Table 1. All composite resins examined were of shade A3.

SEM

Several specimen blocks were made by injecting the composite resin into a metal mold (4×4×1 mm). The surface was then covered with a microslide glass and light-cured for 60 seconds (BlueLEX, Yoshida Dental, Tokyo, Japan). The borders of each block were rounded with a cutter, and the specimens were mounted in acrylic resin. The surfaces of the restorative composite resins were gently polished with sandpaper sheets (Fuji Star 1000-, 2000-, 4000-, 6000-, 10,000-grit, Sankyo Rikagaku Co., Saitama, Japan) such that the fillers were exposed for observation under SEM. Subsequently, a grinder polisher (Minimet 1000, Buehler, Lake Bluff, IL, USA) was used by adding 6-μm and 0.025-μm diamond pastes for 10 minutes (MetaDi II, Diamond Polishing Compound, Buehler). The surfaces were also slightly etched with a solution of 0.8% (wt/vol) H_3PO_4 for 10 seconds to obtain a clearer image during SEM observation. After which, the specimens were ultrasonically cleansed for 5 minutes, placed on

Table 1 Restorative composite resins examined in the present study

Material (Category)	Batch No.	Manufacturer
Beautifil II (Light-cure fluoride releasing dental restorative material)	040722	Shofu Inc., Kyoto, Japan
Clearfil AP-X (Ligth-cure dental restorative composite resin)	01063A	Kuraray Medical Inc., Tokyo, Japan
Clearfil Majesty (Ligth-cure dental restorative composite resin)	0054AA	Kuraray Medical Inc., Tokyo, Japan
Estelite Σ (Ligth-cure resin-based dental restorative material)	009067	Tokuyama Dental Corp., Osaka, Japan
Filtek Supreme (Ligth-cure universal restorative composite resin)	6BCJ	3M ESPE, Dental Products, St Paul, USA
Filtek Z250 (Ligth-cure universal restorative composite resin)	6MMJ	3M ESPE, Dental Products, St Paul, USA
Solare (Ligth-cure dental restorative composite resin for anterior)	0706051	GC Corporation, Tokyo, Japan
Synergy (Ligth-cure resin-based dental restorative material)	0113324	Coltène Whaledent AG, Altstätten, Switzerland

aluminum stubs with conductive tape, coated with osmium for 10 seconds (HPC-1S, Vacuum Device, Ibaragi, Japan), and observed under SEM (S-4500, Hitachi, Tokyo, Japan) with backscattered electron signal.

TEM

Specimens were prepared by placing the composite resin directly into a silicon rubber plate for embedding. Their surfaces were then covered with a microslide glass and light-cured for 60 seconds. After which, the specimens were cut with a 45° diamond knife (Diatome, Biel, Switzerland) positioned in an ultramicrotome (MT2-B, Ivan Sorvall Inc., Newtown, CT, USA) to obtain ultra-thin sections of 80 nm thickness. These sections were placed on fine grid meshes (F-200, Nisshin EM, Tokyo, Japan) and observed under TEM (H-7100, Hitachi).

Microhardness test

A total of 32 disks were prepared by placing the composite resin into a plastic mold of 10 mm diameter × 1 mm height. Both surfaces were covered with slide glasses and light-cured for 60 seconds, using a light beam directed at the top and bottom surfaces for 30 seconds each. The set specimens were then stored dry in a black receptacle at room temperature for five days. After which, Vickers hardness was evaluated with a microhardness tester (HMV-2, Shimadzu Corp., Kyoto, Japan).

A load of 2.942 N was applied to the resin disks for 10 seconds, and the scores were recorded in hardness Vickers (HV). The test was performed 40 times for every restorative composite resin, and the procedure was divided into 10 indentations for each resin disk. Descriptive statistics, including the mean and standard deviation, were calculated, and Scheffé's *post hoc* multiple comparison test (one-way analysis of variance) with significance predetermined at $P<0.05$ was carried out.

Volume/weight fraction of filler

Five specimens of each composite resin were prepared in the same way as that described for TEM observation. The specimens were weighed (approximately 0.45 g) in an analytical balance with an accuracy of 0.0001 g to determine the mass of the polymerized composite resins (Wa). To eliminate the organic phase, the specimens were burned in a furnace at 700°C for 60 minutes so as to obtain the mass of the inorganic filler. After the furnace was turned off, the specimens were slowly cooled for 90 minutes to room temperature. The mass of the inorganic filler was then obtained by using an analytical balance (Wb). The volume/weight fraction of inorganic filler was calculated using the following equation:

$$(Wb/Wa) \times 100\ wt\%$$

The mean values were compared using one-way ANOVA and Scheffé's *post hoc* multiple comparison test, with significance defined at $P<0.05$.

Energy dispersive X-ray microanalysis (EDX)

Resins blocks of 4×4×1 mm were prepared as described before, and the specimens were placed on carbon stubs. The specimens were coated with osmium for 5 seconds and analyzed using EDX (Super Xerophy, S-817XI, Horiba Stec, Kyoto, Japan), which was attached to SEM S-4500. An area of approximately 20×15 μm was selected for 2D analysis, which included both the resin matrix and filler particles. Relative values were obtained after 300 seconds of measurement.

RESULTS

SEM

Observation with backscattered electron signal provided an adequate contrast between the resin matrix and fillers (Fig. 1). The shapes and sizes of the filler particles were different amongst the composites. The traditional appearance of compact-filled composite resins[2] was observed in Beautifil II and Clearfil AP-X; however, a greater amount of filler particles was presented in Clearfil AP-X. For Clearfil Majesty and Solare, the backscattered images showed that they were alike — where their bigger fillers particles were observed in black color, as opposed to the traditional composites filled with observable particles in white color[11]. For Filtek Supreme, Filtek Z250, and Synergy, homogeneous smaller filler particles were shown because these composite resins were filled with nanoparticles; nonetheless, some microparticles were observed in Filtek Supreme. For Estelite Σ, the backscattered image showed a view which was the most dissimilar among the eight composite resins.

TEM

Images of the restorative composite resins under TEM are shown in Fig. 2. Great diversity in the ultrastructure was observed among the eight composites. For Beautifil II, Clearfil AP-X, Filtek Z250, and Synergy, views of their ultrastructures were consistent at both low and high magnification modes. For Clearfil Majesty (Fig. 2C) and Solare (Fig. 2G), the filler particles dispersed in their resin matrices were comparable to those observed in Beautifil II, Clearfil AP-X, and Synergy. For Estelite Σ (Fig. 2D), it seemed to be filled with homogeneous spherical nanoparticles with the inclusion of a few traditional filler particles.

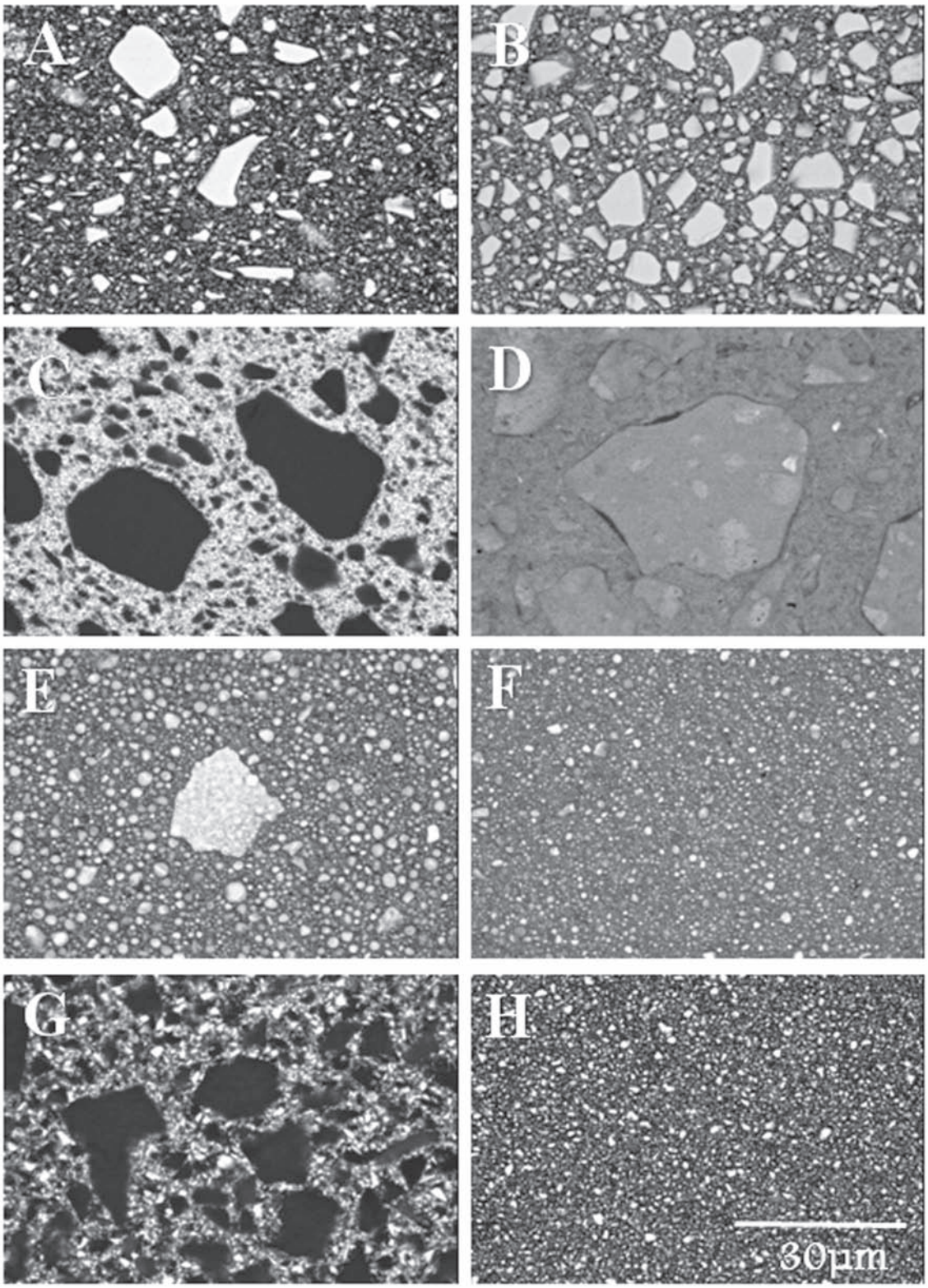

Fig. 1 SEM backscattered images of the restorative composite resins: (A) Beautifil II; (B) Clearfil AP-X; (C) Clearfil Majesty; (D) Estelite Σ; (E) Filtek Supreme; (F) Filtek Z250; (G) Solare; and (H) Synergy (original magnification ×1000).

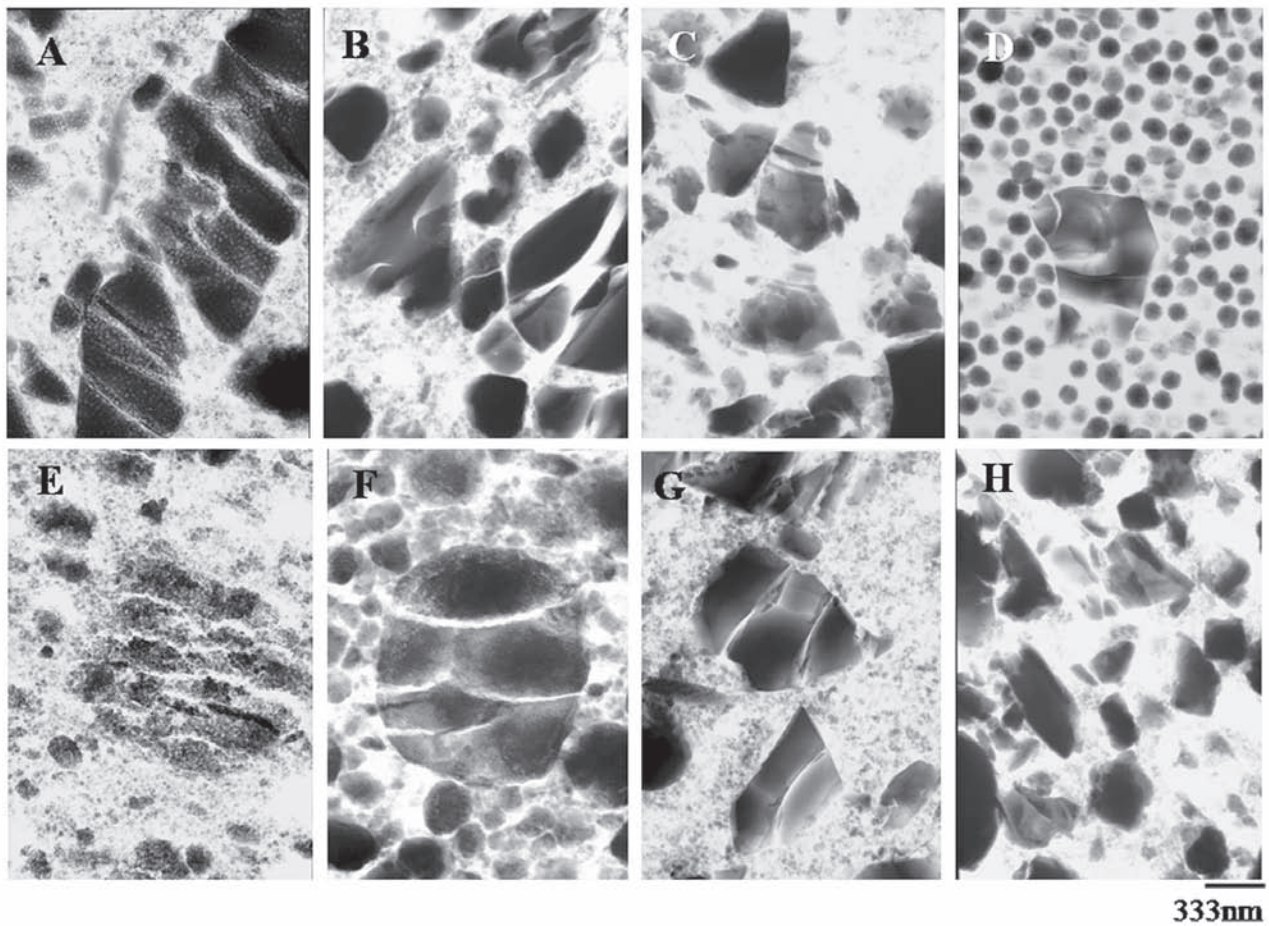

Fig. 2 TEM images of the restorative composite resins: (A) Beautifil II; (B) Clearfil AP-X; (C) Clearfil Majesty; (D) Estelite Σ; (E) Filtek Supreme; (F) Filtek Z250; (G) Solare; and (H) Synergy (original magnification ×30,000).

It is interesting to note that Clearfil Majesty (Fig. 3) and Solare (Fig. 4) presented the most complex ultrastructures, whereby black fillers particles observed with SEM backscattered electron signal were observed as gray color with TEM. These particles seemed to be organic filler particles including smaller nanoparticles of inorganic fillers. For Filtek Supreme, although it has an ultrastructure similar to that of Filtek Z250, it also contained some apparent splintered pre-polymerized nanofiller complexes[1] (Fig. 5).

Microhardness test

The mean values of Vickers hardness are shown in Table 2. It must be mentioned that in all cases, the size of the indentations was larger than the filler particles. As such, the score recorded was the average value of both the resin matrix and filler. Large differences in microhardness were observed among the composites. The results were statistically significant in all comparisons except between Clearfil Majesty and Estelite Σ, and betweem Filtek Supreme and Filtek Z250. Clearfil AP-X showed the highest mean value (112.0±9.3 HV), whereas Solare presented the lowest score (42.8±2.5 HV).

Volume/weight fraction of filler

Table 3 shows the mean values of the volume/weight fraction of filler expressed in percentages. The inorganic filler concentrations of Filtek Supreme and Synergy were comparable. However, statistically significant differences were seen among all the other composite resins.

EDX

The chemical compositions of the restorative composite resins, including the elements with relative values, expressed in weight percentage are presented in Table 4. Similar elements, such as C, O, and Si, were detected; however, the concentration

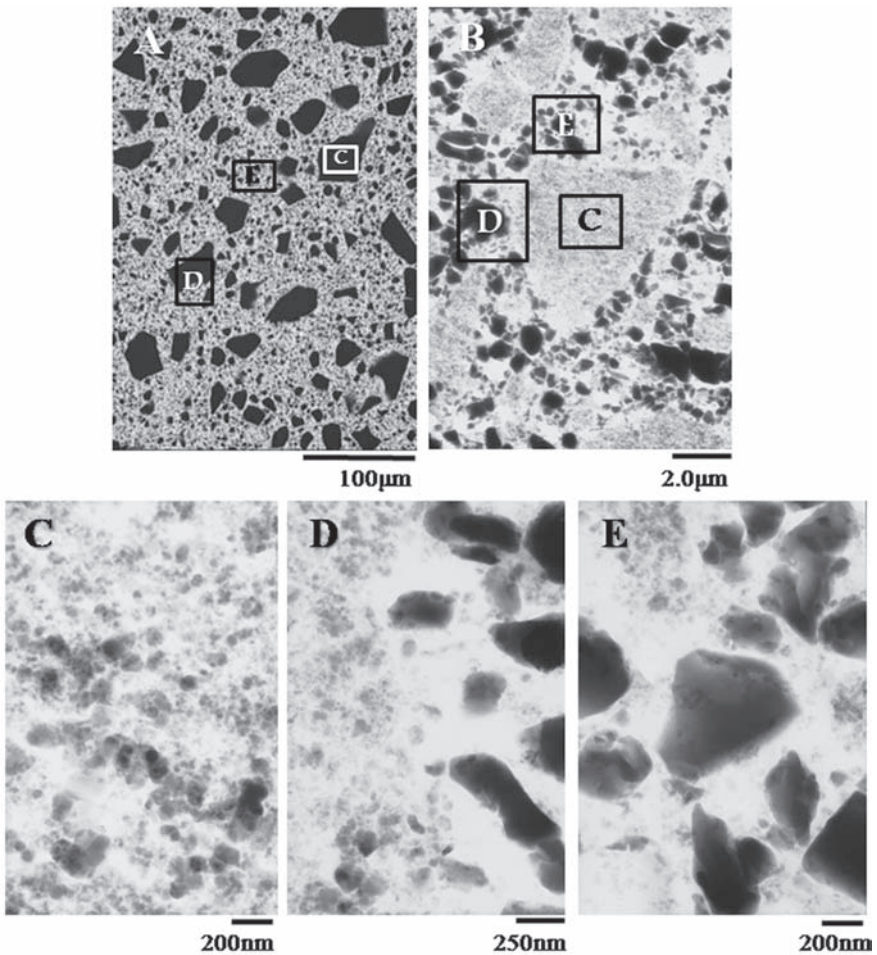

Fig. 3 Clearfil Majesty viewed under SEM and TEM. (A) SEM backscattered image (original magnification ×300). (B) General view of Clearfil Majesty under TEM (original magnification ×5000). (C) Representative TEM image of 'C' rectangle in figures A and B, showing the larger filler particles (original magnification ×50,000). (D) TEM image of the 'D' rectangle in figures A and B, showing the interface between figures C and E (original magnification ×40,000). (E) TEM image of the 'E' rectangle in figures A and B, showing the filler particles dispersed in the resin matrix (original magnification ×50,000).

was different in every composite resin. The filler contents showed interesting differences in elemental composition and concentration; nevertheless, Si seemed to be a common filler component.

DISCUSSION

Composite resins are routinely classified on the basis of filler particle size for the purposes of research, clinical applications, and communications[1,3,12]. However, gaping variations exist when defining the average particle size or range of sizes of the particles for their material grouping[1,3]. The investigation of filler technology is particularly important because filler content has been shown by numerous authors to strongly correlate with the mechanical and physical properties of composites resins[2,3,6,8,9,12]. In this study, the SEM and TEM images of the restorative composite resins showed a great diversity of ultrastructures with regard to filler particle size, shape, and content. These findings were consistent with the significant differences in filler particle numbers and sizes of composite resins reported by Jaarda *et al*.[3].

The size and characteristics of filler particles have been considered as significant factors in the

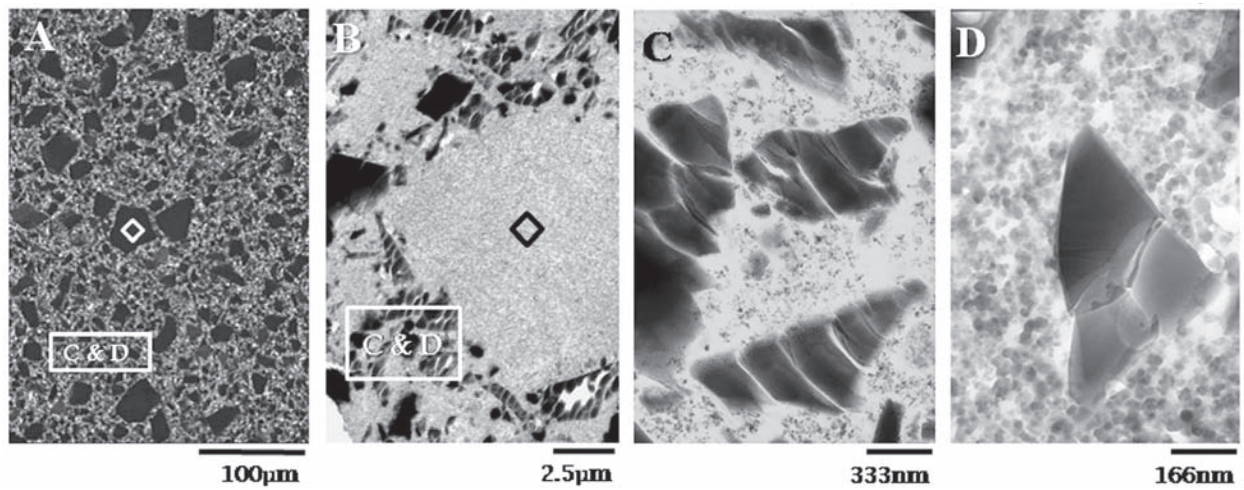

Fig. 4 Solare viewed under SEM and TEM. (A) SEM backscattered image (original magnification ×300). (B) General view of Solare under TEM (original magnification ×4000). The rhomboids and rectangles labeled as 'C' and 'D' in figures A & B indicate the same kind of filler particles; however, the color contrast observed with SEM is opposite to the view with TEM. (C) and (D) Representative TEM images of the rectangles labeled 'C' and 'D' in figures A and B, showing the filler particles dispersed in the resin matrix (original magnification ×30,000 and 60,000 respectively).

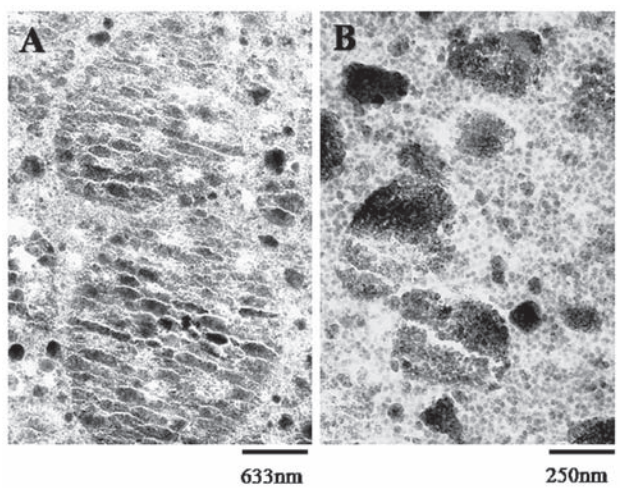

Fig. 5 View of Filtek Supreme under TEM showing apparent splintered pre-polymerized microfiller complexes. (A) Original magnification ×12,000; and (B) Original magnification ×40,000.

rate of wear of composites[3,10,13,14]. Although the property of wear was not evaluated in this study, practitioners should keep in mind that wear is the loss of material through different processes: abrasion, adhesion, fatigue and corrosive effects which act in various combinations depending on the properties of the materials. Abrasion and attrition have been identified as the main clinical wear mechanisms for dental composites[13]. It has been reported that the addition of filler particles to a composite increases its wear resistance[10], but that optimal enhancement of wear resistance can be achieved only if the particles are well bonded to the resin matrix[4,10,13,15]. For this reason, silane coupling agents have been introduced to provide good bonding between fillers and resin matrix components in dental composites, but this bond can be degraded by water absorbed by the composites[4,10].

In addition, inter-particle spacing — which depends on filler particle size and filler load by volume — has proven to be another key factor that affects the wear resistance of composites[10,16]. The shorter the distance between the particles, the more the matrix will be protected against erosive activities[16]. In the present study, sectioning on an ultramicrotome using a diamond knife could have produced some cracks in the fillers[11]. These cracks were lines produced by the mechanical sectioning of the ultramicrotome and diamond knife, and they were relatively parallel to each other or in the same direction. On the use of TEM to analyze the resin-filler interface and inter-particle spacing, it was thus employed in this study because it has been highly recommended in other reports[10,16]. Based on the TEM images shown in Fig. 2, all restorative composite resins exhibited acceptable resin-filler interface. However, inter-particle spacing was different in each case due to diversity in shape, size, and content of the filler particles. In this connection, microfilled composites possess the highest percentage of inorganic fillers and exhibit the most homogeneous

Table 2 Vickers hardness of the restorative composite resins

Composite resin	Mean	SD	Scheffèr Test*
Beautifil II	77.0	6.2	A
Clearfil AP-X	112.0	9.3	B
Clearfil Majesty	57.7	6.5	C
Estelite Σ	58.4	2.3	C
Filtek Supreme	84.5	3.2	D
Filtek Z250	87.2	2.8	D
Solare	42.8	2.5	E
Synergy	66.7	2.7	F

*Adhesives with different letters are significantly different from each other.

Table 3 Mean values expressed in percentage of the volume/weight fraction of filler content

Composite resin	Mean	SD	Scheffèr Test*
Beautifil II	76.6	0.8	A
Clearfil AP-X	82.9	0.3	B
Clearfil Majesty	57.1	0.6	C
Estelite Σ	67.4	0.4	D
Filtek Supreme	73.3	0.1	E
Filtek Z250	78.0	0.1	F
Solare	45.5	0.3	G
Synergy	73.8	0.8	E

*Adhesives with different letters are significantly different from each other.

Table 4 Energy dispersive X-ray micro analysis of the composite resins with relative values expressed in weight percentages

	C	O	F	Na	Al	Si	K	Ti	Co	Sr	Zr	Ba
Beautifil II	42.56	29.09	2.54	0.64	6.82	7.21	----	----	----	11.12	----	----
Clearfil AP-X	37.12	30.11	----	----	2.46	16.11	----	----	0.08	----	----	14.12
Clearfil Majesty	39.40	31.40	----	----	2.07	15.44	----	----	0.01	----	----	11.67
Estelite Σ	35.95	34.72	----	0.49	----	21.02	----	0.22	----	----	7.59	----
Filtek Supreme	33.86	34.38	----	----	----	22.76	----	----	----	----	9.00	----
Filtek Z250	37.69	33.21	----	0.17	----	19.77	----	----	----	----	9.16	----
Solare	45.11	33.46	----	----	2.67	16.02	2.73	----	----	----	----	----
Synergy	38.24	30.06	----	----	2.60	14.28	----	----	0.10	----	----	14.73

dispersion of fillers. Coupled with the strongest chemical bond between the fillers and resin matrix, microfilled composites should have the best wear resistance[10]. In contrast, larger filler particles offer less favorable wear resistance properties[17].

Although flexural strength was not measured in this study, it has been considered as another important property of restorative dental materials. Zandinejad *et al.*[4] found that porosity increased flexural strength significantly but did not affect flexural modulus and diametral tensile strength. Therefore, porous fillers can be considered as an important and feasible way to reinforce dental composites. On the overall, the TEM images in Fig. 2 show that the filler particles of the evaluated composite resins were slightly porous. However, Beautifil II, Filtek Supreme, and Filteck Z250 seemed to present a higher degree of porosity with their larger filler particles. Consequently, the greater porosity of the latter type of particles can promote a better penetration of resin into the fillers, thereby enabling a stronger bond between the two components[4].

On Vickers hardness, the microhardness test

revealed significant difference among the composites (Table 2). These findings were consistent with the mean values of volume/weight fraction of filler (Table 3). In general, higher values of Vickers hardness were found in composite resins with higher volume/weight fraction of filler values. However, Synergy indicated otherwise by presenting a significantly lower value of Vickers hardness than Filtek Supreme, although both had comparable mean values of volume/weight fraction of filler. This result could be explained in terms of the particular combination of filler particle size and chemical composition of Synergy. The chemical compositions of Clearfil AP-X and Synergy were alike (Table 4); however, their Vickers hardness values were significantly different, probably because the larger filler particles of Clearfil AP-X made it harder (Fig. 1). On the other hand, the sizes of the filler particles of Filtek Z-250 and Synergy were similar, and the volume/weight fraction of filler values of both Filtek Supreme and Synergy were almost the same; nonetheless, Synergy showed a significantly lower Vickers hardness value in both comparisons. Therefore, the chemical composition suggested that a higher concentration of Si combined with Zr could make a composite resin harder. In brief, the results of this study demonstrated that these identified factors had an impact on Vickers hardness: ultrastructure of composite resin as observed with both SEM and TEM, size of filler particles, volume/weight fraction of filler, and chemical composition.

Kim *et al.*[12] stated that composite resins with higher hardness produced a positive effect on wear resistance. In this study, Clearfil AP-X presented the significantly highest value, followed by Filtek Z250, Filtek Supreme, and Beautifill II. Therefore, accordingly, these composite resins can be successfully used as universal restoratives, especially for posterior teeth and anterior teeth areas that must withstand high occlusal forces. The microhardness of Filtek Z250 (specimens with 1 mm thickness) has shown a comparable value (83.0±3.2 HV) after being cured with halogen light for 40 seconds[18]. On the other hand, Synergy, Estelite Σ, and Clearfil Majesty could also be used for both anterior and posterior restorations; nevertheless, additional studies are required to evaluate their performance as stress-bearing restorations. As for the significantly lowest microhardness value of Solare, this result could be explained on the basis that this composite is prescribed for anterior restorations.

In a study by Rode *et al.*[18], it was reported that current light curing units promote a similar degree of conversion and microhardness for the same composite resin. However, the shape of filler particles, as well as particle size and filler content, can significantly affect the light transmittance characteristics[19]. Apart from filler properties, Emami *et al.* suggested that the characteristic of incident light also affected light absorbance by dental composites[20]. In the same vein, Arikawa *et al.*[21] concluded that markedly inhomogeneous light emitted from a light curing unit could result in inhomogeneous polymerization in some areas of the restoration below the light guide tip.

The composition of composite resins is complex. By means of EDX in this study, general information on the elemental compositions of the composite resins evaluated is obtained[11]. Common resin matrix elements such as C and O were detected in all the materials with comparable concentrations. Nevertheless, Solare presented the highest concentration of C, suggesting that this composite was slightly lower-filled than the others — and this could be related with its significantly lowest Vickers hardness value. The filler contents showed great diversity, whereby elements such as Na, Al, K, Ti, Co, Sr, Zr, and Ba were detected. Nevertheless, in agreement with previous reports[13], Si seemed to be a common filler component.

Secondary caries at the tooth-restoration margin is the most frequently cited reason for restoration replacements[22]. Based on previous studies that measured fluoride release from dental materials, fluoride has been shown to be effective in preventing caries development[22,23]. Among the composite resins evaluated in this study, fluoride was detected only in Beautifill II, which contained surface pre-reacted glass ionomer (S-PRG) filler[11].

One of the most interesting findings of this study was the comparison between Clearfil AP-X and Clearfil Majesty. This was because both composites were produced by the same manufacturer and they presented similar compositions after EDX analysis (Table 4). To examine their ultrastructures, SEM and TEM were employed in this study. With the SEM method, the contrast obtained with the back-scattered electron signal was very useful for showing the differences between the resin matrix and filler particles. Consequently, the SEM backscattered images showed a dramatic dissimilarity between the ultrastructures of Clearfil AP-X and Clearfil Majesty. However, it is noteworthy that the color contrast was opposite to each other (Figs. 1B and 1C). TEM images revealed that Clearfil Majesty had a more complex ultrastructure (Figs. 2B and 2C; Fig. 3). At this juncture, it must also be mentioned that Clearfil AP-X presented a significantly higher Vickers hardness value (Table 2). Taken together, these results suggested that whereas composite resins could present similar compositions, the different technologies employed to manufacture them could cause them to possess dramatically different properties and ultrastructures.

The TEM images in Figs. 4 and 5 illustrate the ultrastructures of Solare and Filtek Supreme respectively. The complex ultrastructure of Solare was comparable to that observed for Clearfil Majesty. As for Filtek Supreme, TEM clearly showed that it contained splintered pre-polymerized nanofiller complexes.

With the aim of combining the advantages of hybrid and microfilled restorative materials, various new composites have been developed based on nanoparticle filler technology. Nanocomposites claim to provide the esthetic properties required for anterior restorations, as well as mechanical properties necessary for posterior, stress-bearing restorations. On the latter claim, nanocomposites have indeed been demonstrated to possess favorable mechanical properties. In addition, previous scientific data from *in vitro* investigations indicated that the majority of nanofilled composites led to higher surface quality and superior polish retention. However, in a study by Jung *et al.*, Filtek Supreme (which is filled with nanoparticles) showed a surface quality that was no better than that of a traditional hybrid composite after polising[24].

CONCLUSIONS

Within the limitations of the current study, the following conclusions were drawn:

(1) The revolutionary technologies employed to manufacture composite resins have dramatically changed their properties, especially in terms of the size, shape, and distribution of the filler particles.

(2) SEM and TEM methods used in this study were useful for examining the ultrastructures of composite resins. However, the ultrastructure of nanofillers could be observed only with TEM at high magnifications.

(3) The ultrastructure, size of filler particles, volume/weight fraction of filler, and chemical composition of a composite resin had an impact on its Vickers hardness.

(4) Although recently improved restorative composite resins could offer the properties required of dental restorations, the great diversity among them warrants further studies to ensure that clinical applications optimally match their differing properties.

REFERENCES

1) Lang BR, Jaarda M, Wang RF. Filler particle size and composite resin classification systems. J Oral Rehabil 1992; 19: 569-584.
2) Willems G, Lambrechts P, Braem M, Vanherle G. Composite resins in the 21st century. Quintessence Int 1993; 24: 641-658.
3) Jaarda MJ, Lang BR, Wang RF, Edwards CA. Measurement of composite resin filler particles by using scanning electron microscopy and digital imaging. J Prosthet Dent 1993; 69: 416-424.
4) Zandinejad AA, Atai M, Pahlevan A. The effect of ceramic and porous fillers on the mechanical properties of experimental dental composites. Dent Mater 2006; 22: 382-387.
5) Ottenga ME, Mjör I. Amalgam and composite posterior restorations: curriculum *versus* practice in operative dentistry at a US dental school. Oper Dent 2007; 32: 524-528.
6) da Silva GR, Simamoto-Júnior PC, da Mota AS, Soares CJ. Mechanical properties of light-curing composites polymerized with different laboratory photo-curing units. Dent Mater J 2007; 26: 217-223.
7) Domingo C, Arcís RW, Osorio E, Osorio R, Fanovich MA, Rodríguez-Clemente R, Toledano M. Hydrolytic stability of experimental hydroxyapatite-filled dental composite materials. Dent Mater 2003; 19: 478-486.
8) Faltermeier A, Rosentritt M, Faltermeier R, Reicheneder C, Müssig D. Influence of filler level on the bond strength of orthodontic adhesives. Angle Orthod 2007; 77: 494-498.
9) Xu HH. Whisker-reinforced heat-cured dental resin composites: effects of filler level and heat-cure temperature and time. J Dent Res 2000; 79: 1392-1397.
10) Lim BS, Ferracane JL, Condon JR, Adey JD. Effect of filler fraction and filler surface treatment on wear of microfilled composites. Dent Mater 2002; 18: 1-11.
11) Scougall Vilchis RJ, Hotta Y, Yamamoto K. Examination of six orthodontic adhesives with electron microscopy, hardness tester and energy dispersive X-ray microanalyzer. Angle Orthod 2008; 78: 655-661.
12) Kim KH, Ong JL, Okuno O. The effect of filler loading and morphology on the mechanical properties of contemporary composites. J Prosthet Dent 2002; 87: 642-649.
13) Atai M, Yassini E, Amini M, Watts DC. The effect of a leucite-containing ceramic filler on the abrasive wear of dental composites. Dent Mater 2007; 23: 1181-1187.
14) Faltermeier A, Rosentritt M, Reicheneder C, Müssig D. Experimental composite brackets: influence of filler level on the mechanical properties. Am J Orthod Dentofacial Orthop 2006; 130: 699.e9-699.e14.
15) Faltermeier A, Behr M, Müssig D. Esthetic brackets: the influence of filler level on color stability. Am J Orthod Dentofacial Orthop 2007; 132: 5.e13-5.e16.
16) Vankerckhoven H, Lambrechts P, van Beylen M, Vanherle G. Characterization of composite resins by NMR and TEM. J Dent Res 1981; 60: 1957-1965.
17) Venhoven BA, de Gee AJ, Werner A, Davidson CL. Influence of filler parameters on the mechanical coherence of dental restorative resin composites. Biomaterials 1996; 17: 735-740.
18) Rode KM, Kawano Y, Turbino ML. Evaluation of curing light distance on resin composite microhard-

ness and polymerization. Oper Dent 2007; 32: 571-578.
19) Arikawa H, Kanie T, Fujii K, Takahashi H, Ban S. Effects of inhomogeneity of light from light curing units on the surface hardness of composite resin. Dent Mater J 2008; 27: 21-28.
20) Emami N, Sjödahl M, Söderholm KJ. How filler properties, filler fraction, sample thickness and light source affect light attenuation in particulate filled resin composites. Dent Mater 2005; 21: 721-730.
21) Arikawa H, Kanie T, Fujii K, Takahashi H, Ban S. Effect of filler properties in composite resins on light transmittance characteristics and color. Dent Mater J 2007; 26: 38-44.
22) Xu HH, Weir MD, Sun L, Takagi S, Chow LC. Effects of calcium phosphate nanoparticles on Ca-PO_4 composite. J Dent Res 2007; 86: 378-383.
23) Chan WD, Yang L, Wan W, Rizkalla AS. Fluoride release from dental cements and composites: a mechanistic study. Dent Mater 2006; 22: 366-373.
24) Jung M, Eichelberger K, Klimek J. Surface geometry of four nanofiller and one hybrid composite after one-step and multiple-step polishing. Oper Dent 2007; 32: 347-355.