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Evaluation of surface characteristics of dental composites using profilometry, scanning electron, atomic force microscopy and gloss-meter

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Abstract The aim of this *in vitro* investigation was to compare various roughness and topography measurement methods to characterize the surface quality in several types of resin composites. The initial surface quality of several resin composites was compared. The materials evaluated were of three categories: i) *hybrid:* TPH Spectrum; ii) *reinforced microfill:* Micronew and iii) *microhybrid:* Synergy Duo, Esthet-X, Point.4 and Palfique Estelite. Three Groups of identical disk-shaped specimens (10 × 1.5 mm) were prepared from each material ($n = 6$) and polished with Soflex discs. Macro-roughness (Ra) was measured with Group 1 by 2-D profilometry. Atomic Force Microscopy (AFM) gave 3-D images and micro-roughness (Ra) of Group 2. Surface optical gloss at 60° was determined for Group 3. Specimens of each material were also studied by scanning electron microscopy. Macro-Ra values (μm) ranged from 0.30 to 0.56. Micro-Ra values ranged from 0.03 to 0.14 and they differed from macro-Ra values in ranking order. Percentage Gloss values ranged from 30.6 to 70.1%. The results revealed that micro-roughness showed a high correlation with gloss values ($r = 0.93$), whilst macro-roughness did not ($r = 0.62$). Moreover, the AFM method showed higher capability to distinguish surface roughness compared with the 2-D profilometry and to reveal more detailed definition of surface texture than the examination under SEM.

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Clinical significance

Reinforced microfill composite exhibited similar gloss and roughness values with microhybrid resin composites but illustrated higher gloss and lower roughness values compared with the hybrid resin-composites evaluated. Data recorded by the AFM method described more reliably the surface quality of resin composites.

Introduction

Aesthetic concepts have been particularly important in driving the development of dental restorative materials in the last few years. A glossy and perfectly smooth surface is a requirement for desirable aesthetic appearance. It also needs to remain like this for a long period within the oral environment. The smooth surface, apart from enhancing the aesthetic result, prevents the formation of discoloring films and plaque retention due to the absence of micro-roughness. Moreover, surface smoothness decreases the coefficient of friction and subsequently this may reduce wear rate [1], which compromises the clinical performance of the restorations. Surface quality also affects the fracture resistance in brittle materials such as the resin composites [2].

The quality of a polished resin composite surface is related to intrinsic material properties and to the finishing/polishing procedure applied. A type of resin composite is known as *microhybrid*. These materials incorporate a high volume-fraction of filler particles, with a mean size below 1 μm, along with a narrow particle size distribution. Manufacturers claim that the characteristics of microhybrid resin composites include improved handling properties, adequate cohesive strength, high-gloss, ceramic-like smooth polished surface and optical properties which mirror the natural enamel and

dentin [3]. However, independent *in vitro* and *in vivo* studies, especially on the aesthetic appearance of the microhybrid composites, in terms of their smoothness and gloss, are limited.

Gloss is an important property and is used primarily as a measure of surface shine [4]. The gloss of a surface may be defined as its degree of approach to a mirror surface. A perfect mirror surface is said to have maximum gloss [5].

Various techniques can be used for assessing surface roughness. Research on surface roughness in dental materials has involved qualitative methods such as optical and scanning electron microscopy and quantitative methods, such as surface profile analysis. Contact diamond and non-contact laser modes as well as laser reflectivity measuring systems are commonly applied for surface profile measurements [6–8]. Several shortcomings with respect to the sensitivity of the methods and overall limitations of surface profilometry have been described [6, 9, 10]. In the last decade, the newer technique of Atomic Force Microscopy (AFM) has been employed in the dental materials research field. AFM is capable of providing three-dimensional detailed topographical images of surface roughness at a nanometer resolution. Although these features make AFM a promising technique for surface quality evaluation of dental materials [7, 11, 12] limited applications could be found to date [13].

The aim of this study was to assess surface characteristics of resin composites classified as microhybrid, compared with other resin types, using the following instruments: mechanical profilometer, atomic force, scanning electron microscopes and a gloss-meter. The research hypotheses tested were that i) the methods applied for surface analysis are equally effective to determine the surface quality in resin composites and ii) hybrid, microhybrid and reinforced microfill resin-composites present similar surface quality.

Materials and methods

Six materials were studied (Table 1). The classification of the products and the tabulation of the composition of the filler particles were based on the manufacturers' data.

Three Groups, with six specimens each one ($n = 6$) per material for all surface measurements were prepared. The shape of specimens was disk (10 mm in diameter and 1.5 mm in thickness), and were prepared as follows. A polyethylene mold was used and filled with each resin-composite paste. The free surface was covered with a transparent cellulose strip and pressed with a microscope glass slide to remove the material excess. Then, the top surface was photo-polymerized for 40 s, using a light-curing unit (Elipar TriLight, 3M ESPE, St Paul, MN, USA) operated in standard mode and emitting 840 mW/cm^2 irradiance, as measured with a curing radiometer (Model 100,

Table 1 The resin composites tested and their composition*

Resin composite	Batch number	Manufacturer	Filler composition	Filler content (% ww/vol)	Filler size (μm)
Micronew (*reinforced microfill*)* *Shade A3*	0200007732	Bisco, Inc, Schaumburg, IL, USA	Amorphous silica, strontium alumino silicate	70/-	Mean = 0.5 Range: –
TPH Spectrum *(hybrid)* Shade A3*	200011106	DeTrey/Dentsply, Konstantz, Germany	Colloidal silica, barium-alumino-boro silicate	76/58	Mean: above 1 Range: 0.04–2
Esthet.X *(microhybrid)* Shade A3*	011120	DeTrey/Dentsply, Konstanz, Germany	Silicon dioxide, barium-alumino-boro-fluoride silica	77/60	Mean: 0.6–0.8 Range:0.04–2.5
Synergy Duo *(microhybrid)* Shade A3/D3*	LJ142	Coltene/Whaledent Inc, Mahwah, NJ, USA	Amorphous silica, strontium-alumino silicate	74/59	Mean: 0.6 Range:0.04–2.5
Point.4 *(microhybrid)* Shade A3*	205553	Kerr, Orange, CA, USA	Fumed silica dioxide, barium-alumino-boro silicate	76/58	Mean = 0.4 Range: 90% below 0.8 μm
Palfique *Estelite (microhybrid)* Shade A3*	YE805112	Tokuyama Dental Corp, Taitu-Ku, Tokyo, Japan	Spherical silico- zirconia (sol-gel)	82/71	Mean = 0.2 Range: –

*According to manufacturers' data.

Demetron Corp., Danbury, CT, USA). The mold was then removed and the directly irradiated surfaces were polished sequentially with a complete series of Soflex polishing discs (3M ESPE, St Paul, MN, USA). A single operator, using a low-speed handpiece at approximately 4,000–5,000 rpm, performed the polishing procedure. After that, the polished surfaces were water-rinsed with an air-water syringe for 60 s, to remove any surface debris left and then were air-dried for 30 s. The surfaces were immediately examined under a metallographic microscope (ME 600 Eclipse, Nikon-Kogaku, Tokyo, Japan) to assure the absence of any defects. The surface properties assessed included the roughness average (R_a) by a two-dimensional profilometer and (S_a) by an atomic force microscopy (AFM) as well as the gloss. Furthermore, surface texture evaluation was performed under AFM and scanning electron microscopy (SEM).

A calibrated, mechanical 2-D profilometer (Diavite DH-5, Asmeto, Richterswill, Germany) was used to measure the R_a for each material, according to DIN 4768. A diamond stylus of 5 μm and stylus angle 90° was traversed in a length of 1.25 mm and with cut-off length 0.25 mm. Six measurements in the center of each sample at crossing directions were performed. Three-dimensional images of each polished surface per material were obtained at 100 μm scan sizes using a multimode scanning probe microscope (Nanoscope IIIa, Digital Instruments, Santa Barbara, CA, USA). This was equipped with a scanner of maximum ranges 120 μm × 120 μm × 7 μm, in *x, y* and *z* directions respectively, and an optical microscope to locate the region of interest by monitoring the sample on a TV screen. The images were acquired in contact mode by using a 10 nm etched silicon nitride probe at 320 KHz oscillating frequency and 5 min acquisition period. One region in the center of each specimen, with no visual defects, was analyzed in order to avoid the bias of sensitivity readings of the AFM cantilever. The AFM roughness analysis software was used to evaluate the surface roughness parameter S_a, which is equivalent to the line profilometric parameter described above. Six specimens were analyzed from each material by performing two recordings per specimen's surface at 100 μm scan size.

Qualitative evaluation of the polished surfaces was made by observation under scanning electron microscopy (Quanta 200, FEI, Hillsboro, OR, USA). The surfaces were imaged by low vacuum SEM, operating at 1 Torr pressure, and 20 KVa. Four randomly selected specimens—half from 2-D profilometer and AFM groups-per resin-composite were observed and representative images at 800× magnification were taken.

The surface gloss was measured at 60° incidence angle, using a calibrated infrared gloss-meter (IG 330, Horiba Ltd., Kyoto, Japan). The average of six measurements was recorded per surface.

Statistical analysis of the roughness values, assigned by both techniques, and of the gloss values was performed to define differences among the materials tested, using one-way analysis of variance (ANOVA) and Scheffe tests, at $\alpha = 0.05$. The same statistical analysis was applied to compare the two measurement techniques in terms of R_a. Regression analysis was applied to determine any possible correlation among the R_a measurement techniques and between R_a and gloss values. Statistical analyses were performed by SPSS Version 11.0 (SPSS Inc, Chicago, IL, USA).

Results

The R_a, S_a and surface gloss values recorded are presented in Table 2.

Overall, the AFM method gave lower roughness values (S_a) than the 2-D profilometer (R_a) for all resin composites. Moreover, different rankings of the materials with respect to the roughness values were found with the two measuring techniques. The higher roughness values were found for TPH Spectrum and Synergy Duo and the lower for Micronew, Point.4 and Palfique Estelite when the assessment was facilitated by AFM. Considering those obtained by the 2-D profilometer, the higher values were recorded for Synergy Duo, Palfique Estelite and Point.4 and the lower for Micronew and Esthet.X. No correlation was found between the S_a values recorded by AFM and the R_a values by the 2-D profilometer ($r = 0.59$).

Gloss values ranged from 30.6 percent for TPH Spectrum to 70.1 for Palfique Estelite. A high correlation coefficient ($r = 0.93$) was found between gloss and S_a values (Fig. 1), whereas such correlation was not established for R_a and gloss ($r = 0.62$).

Under SEM evaluation, Palfique Estelite and Point.4 showed more homogeneous surface textures although several narrow scratches were found on a Point.4 surface (Fig. 2). White small spots, randomly distributed, were only noted on Palfique Estelite surface. Microphotographs revealed a relatively uniform surface for Synergy Duo. Filler particles were

Table 2 Surface roughness values (S_a) measured by AFM and 2-D profilometer (R_a) and gloss values at 60°

Materials	S_a (AFM)	R_a (2-D profilometer)	Gloss
Palfique Estelite	0.03 (0.01) b	0.53 ± 0.06 b	70 ± 11.9 c
Micronew	0.04 (0.01) b	0.38 ± 0.03 a	62 ± 9.7 c
Point.4	0.05 (0.01) b	0.52 ± 0.06 b	61 ± 8.9 c
Esthet.X	0.10 (0.02) a	0.30 ± 0.03 a	48 ± 6.1 b
Synergy Duo	0.12 (0.03) a	0.56 ± 0.07 b	46 ± 5.8 b
TPH Spectrum	0.14 (0.05) a	0.39 ± 0.05 a	30 ± 4.3 a

-within a column, values with similar lower case letters are statistically equivalent.

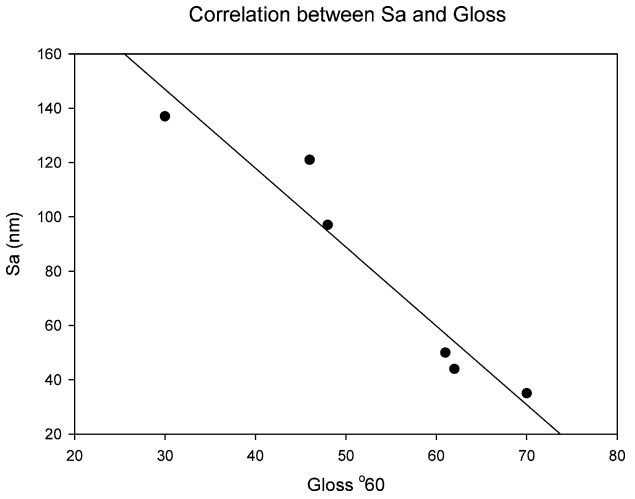


Fig. 1 Graph correlation between AFM roughness (S_a) and Gloss (60°) values.

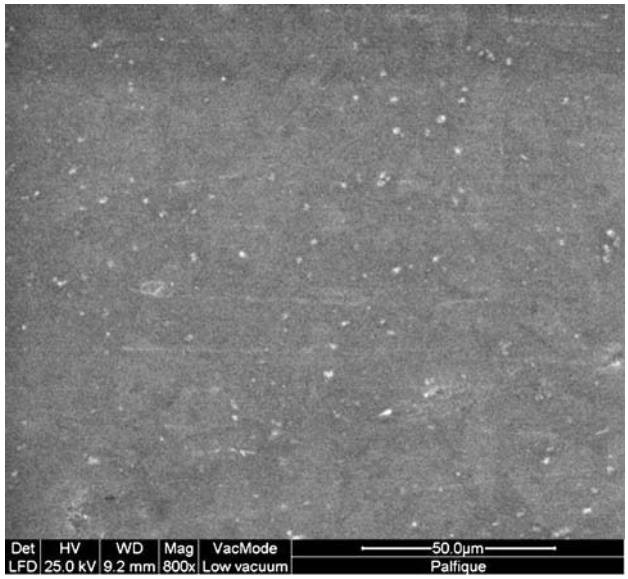
exposed from the matrix of TPH Spectrum, whereas Esthet.X established a moderate relief (Fig. 3). Larger fillers with TPH Spectrum than with Esthet.X surfaces were noticed. Black, areas abnormal in shape and with several large projected fillers ($\geq$5 μm) were detected on Micronew surface (Fig. 4).

AFM images for TPH Spectrum showed a non-uniform surface with distinct sharp projections dotted with pores (Fig. 5). Narrow, deep scratch lines crossed the Synergy Duo surface causing an irregular area (Fig. 6). Esthet.X demonstrated a moderate irregular surface with heights and valleys. A low profile, interrupted by randomly located rounded-off projections, characterized the Micronew surface (Fig. 7). Moderate and slight relief with shallow scratches, were displayed on Point.4 and Palfique Estelite, respectively.

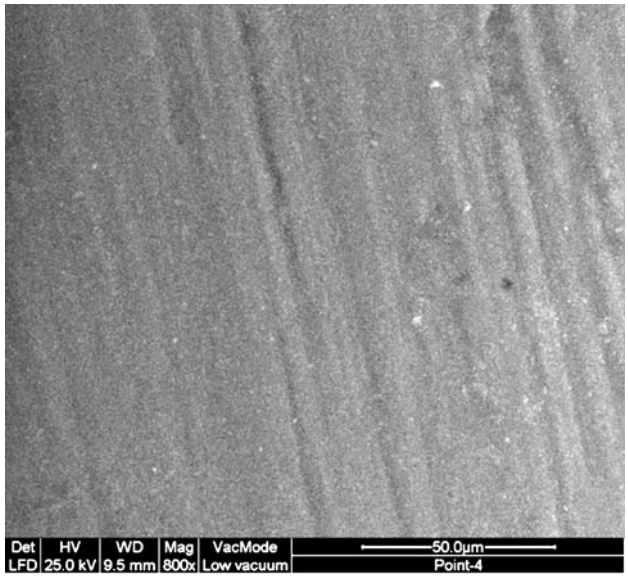
Discussion

The term surface quality reflects a set of widely different properties such as gloss, roughness, colour, polarity and morphology. In the present study, the characteristics of morphology, roughness and gloss were selected to assess polished resin composite surfaces. It has been established previously that the resin composite surface quality is material- and polishing procedure-related. Although each manufacturer for most of the materials evaluated recommends specific polishing systems, the same polishing procedure was applied for all the materials, in the current study, to avoid any differences that might be caused by different polishing systems.

The series of Soflex discs was the system of choice. Aluminum oxide discs have been suggested as standard protocol [14] because of their capability to produce non-destructive, smooth polished surface on a variety of resin composites included microfills as well as heavy filled materials [15]. Concerning the experimental procedure, every effort was applied



(a)

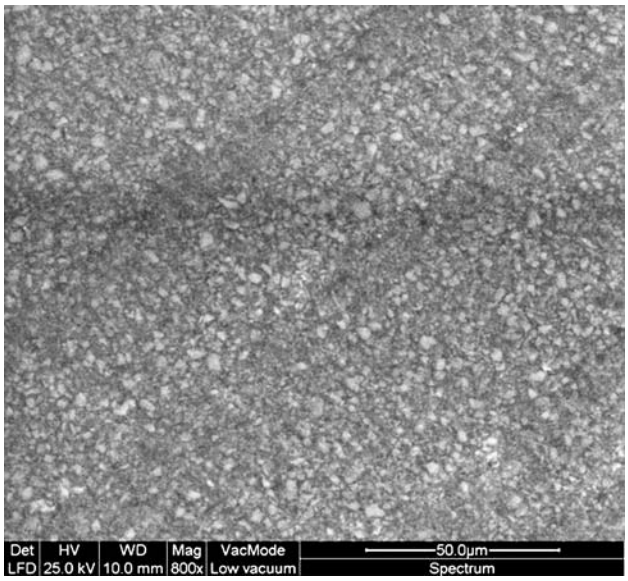


(b)

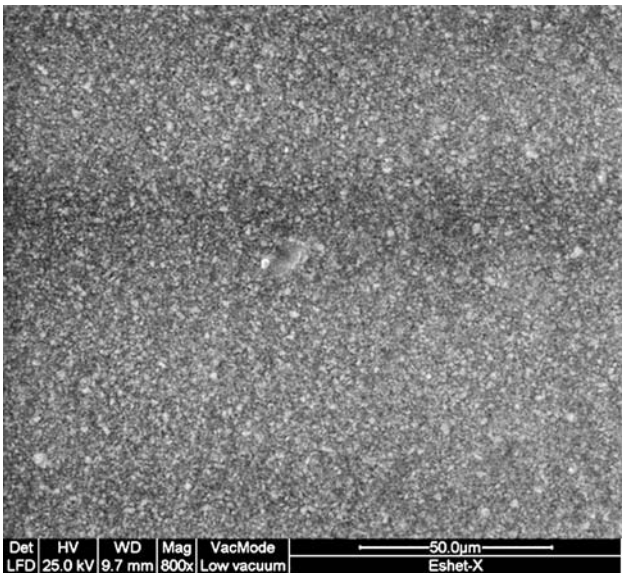
Fig. 2 SEM images of the surfaces (a) Palfique Estelite and (b) Point.4. Small white spot areas are observed on Palfique Estelite and slight lines on Point.4.

to standardize the polishing, in terms of the number, direction and duration of the strokes, and by using one operator to prepare all the specimens.

In this study, the surface characteristics were defined by both qualitative evaluations, assessed by atomic force microscopy and scanning electron microscopy, and by quantitative measurements conducted by 2-D and 3-D profilometry. Both null hypotheses tested in this study were rejected. AFM proved to be a more accurate method for determining the surface quality in resin composites. Also, differences were established among the microhybrid resin composites evaluated.



(a)



(b)

Fig. 3 SEM images of the surfaces TPH (a) Spectrum and (b) Esthet.X. Projected fillers—smaller on Estet.X than TPH Spectrum—are noted.

The qualitative examination of the surface state enabled discrimination between the inherent roughness of the material and the destructive effect of the finishing/polishing instruments. SEM is commonly used to observe surface scratches and defects produced on a surface. SEM images revealed an absence of such defects from the resin composites tested, except for slight grinding lines noticed on a Point.4 surface. However, SEM has limitations in defining surface topography [16]. The electron beam technique does not allow visualization of three-dimensional surface texture. Also, because in beam techniques the contrast relies on the different emission of electrons,

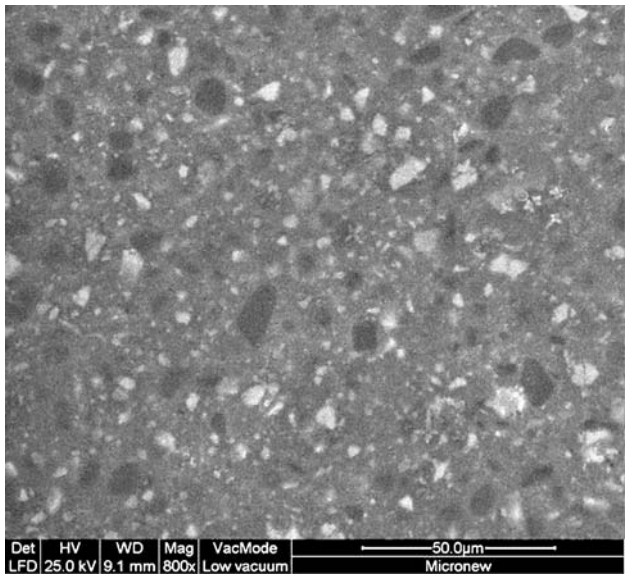


Fig. 4 SEM image of the Micronew surface, characterized by rarely disturbed exposed fillers (large, white areas).

these cannot give contrast on flat homogeneous surface materials.

AFM can be applied for qualitative measurements as well, and provides three-dimensional data. Thus, polishing scratches were detected on TPH Spectrum, Synergy Duo and Esthet.X surfaces and detached fillers were distinguished on TPH Spectrum. These features, were not visible in the SEM images. The differences found between SEM and AFM techniques suggest that AFM can offer more detailed definition of surface topography.

In general, roughness values obtained by profilometers facilitate a quantitative measure of the surface irregularities. The surface roughness, in the current study, was assigned by R_a parameter. Although R_a is considered as a poor indicator of surface texture, this is the most frequently recorded value to verify surface topography in dental materials [17, 18].

Surface topography is three-dimensional in nature. Therefore, the measurement of 3-D surface topography can represent the natural characteristics of a surface, while the measurement of a 2-D profile does not achieve this. The parameters obtained from 3-D are more realistic than those obtained from 2-D profiles [19]. The information that can be obtained by 3-D measurement gives a complete description of surface topography and is more comprehensive than the 2-D measurement [20]. Stylus type profilometers used in the present study provide definitions of surface features for a scale size related to the probe dimensions. Compared to the $5\,\mu$m diamond stylus of the 2-D profilometer, the AFM equipped with a $0.01\,\mu$m SiN_3 tip, permits more precise tracings. Obviously, because of its size, 2-D profilometer cannot penetrate certain micro-irregularities [21] and could not represent

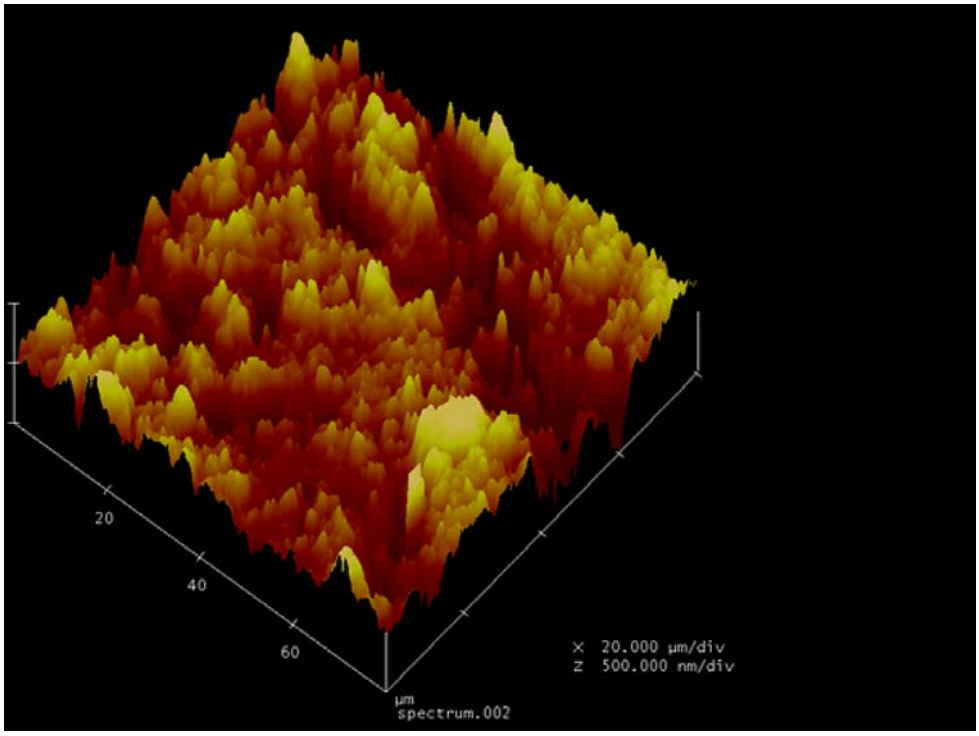


Fig. 5 3-D AFM image of TPH Spectrum showing strong irregular surface with dislodged fillers.

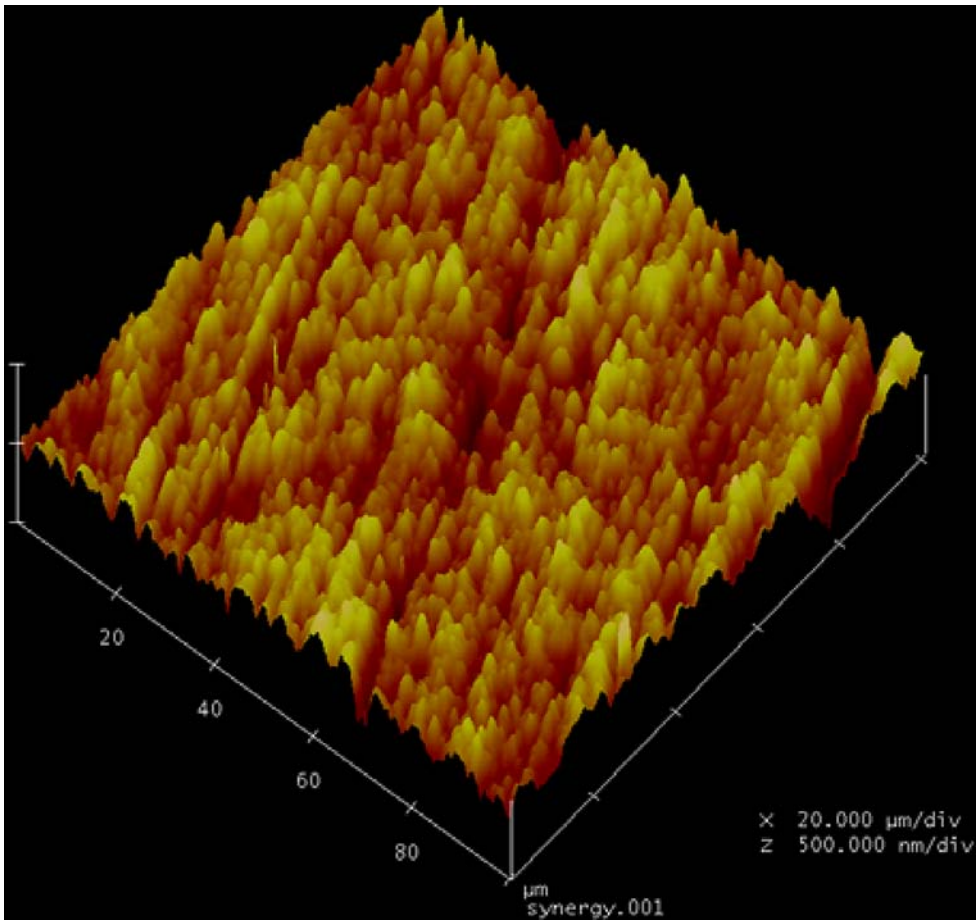


Fig. 6 3-D AFM image of Synergy Duo.

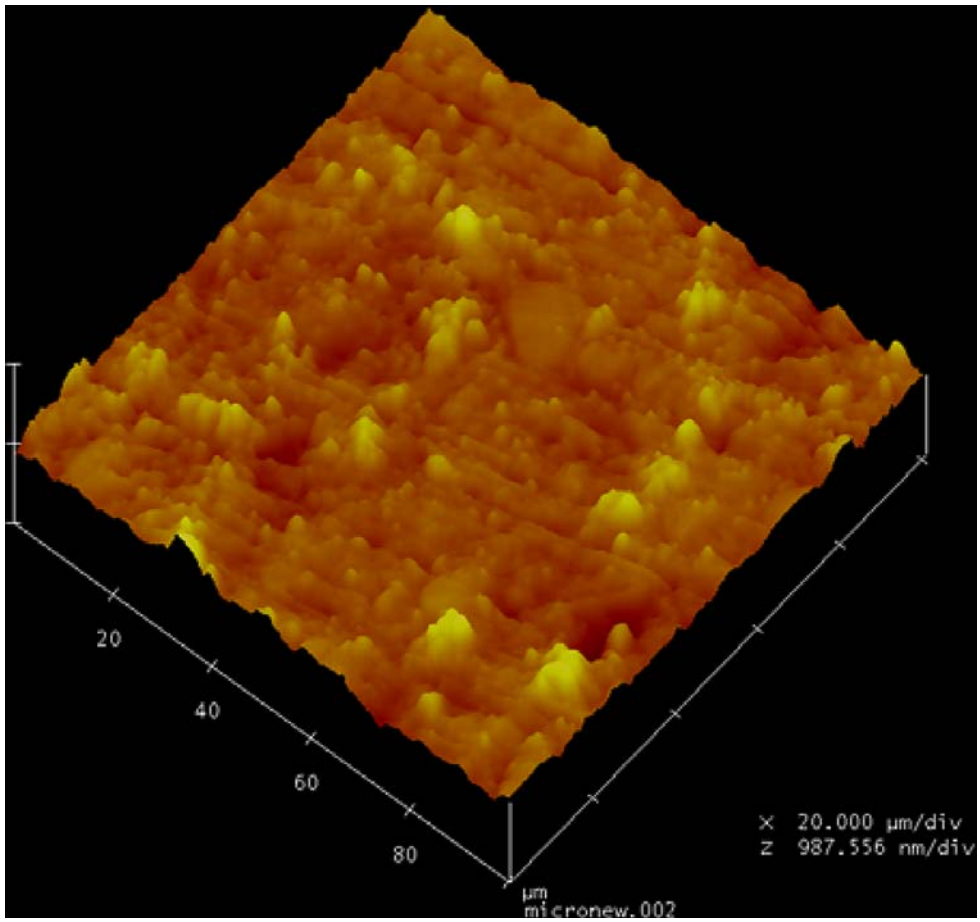


Fig. 7 3-D AFM image of Micronew. Low surface profile is interrupted by rounded shinny projections on surface.

surface features, which were narrower than the stylus. The latter may result in underestimation of the surface roughness [6].

The ranking of the resin composites in terms of S_a, provided by AFM, largely corresponds to the surface appearance of the materials, as depicted by AFM and SEM images. A similar relationship could not be established when R_a was measured by the 2-D profilometer. These findings again highlight the higher capability of the AFM method to distinguish surface texture compared with the 2-D profilometer.

The significantly lower arithmetic values of S_a recorded by AFM compared with 2-D profilometer R_a may be partly attributed to the smaller sample area studied by AFM ($100 \times 100\,\mu$m). It is likely low R_a values were obtained, because the value of each surface parameter depends on the size of the area examined. In addition, the 2-D surface profilometer determines line roughness, in either horizontal or vertical directions, while AFM identifies area roughness monitored on an entire surface. It is therefore unwarranted for arithmetic R_a values from a 2-D profilometer to be compared with AFM analogues.

Comparison of R_a values from this study by the 2-D profilometer with those reported in other investigations is problematic, since several experimental factors influence R_a measurements [17]. Such comparison is not yet possible for S_a values facilitated by AFM because so far such data are not available in the literature for resin composites.

Although a threshold for unacceptable surface roughness has not yet been agreed, Bollen & others [22] have reported that 2-D surface roughness (R_a) above $0.2\,\mu$m results in an increase of plaque accumulation and higher risk for caries and periodontal inflammation. But other reports have found no appreciable differences in plaque accumulation on surfaces with R_a values ranged from 0.7 to $1.4\,\mu$m [23–25]. Chung [26] found that when R_a was lower than $1\,\mu$m the surfaces were visibly smooth. Considering that all R_a values obtained in the current study were above $0.2\,\mu$m and below $0.7\,\mu$m independently of the measurement method, the resin composite surfaces evaluated may be considered to have demonstrated a smooth surface, from the clinical point of view, which presents no risk of plaque accumulation.

During finishing/polishing, the softer resin matrix is subjected to a preferential loss between the harder glass particles and the unsupported fillers are then exposed [14]. It can be assumed that large fillers cause rough surfaces on polished

resin composite. Thus, differences were found among the materials tested, with regard to the roughness and texture established by AFM, which may reflect variations in composition and size distribution of the fillers.

Palfique Estelite and Point.4, which contain the smaller filler particles, exhibited overall superior surface quality compared with the rest of the materials. This suggests the significant benefit of sub-micron filler particles. However, although Synergy Duo and Esthet.X also contain sub-micron fillers, these demonstrated inferior surface characteristics compared with the other resins classified as microhybrid. A wider size distribution of the fillers along with the slightly higher mean particle size may cause this behavior. The round shape of the fillers in Palfique Estelite may be an additional contributor factor for the lowest surface relief observed.

The white spot projected areas distinguished on Palfique Estelite surface may represent alumina oxide particles removed from Soflex discs during polishing, which subsequently were embedded into the resin matrix. More profound surface relief on the other resin composites may not permit visual detection of these particles. Scratches observed on several resin composites can be mostly attributed to the grinding effect of the dislodged fillers. Thus, resin composites composed of smaller fillers exhibited narrower and shallower lines.

Micronew is a highly reinforced microfill composite, since apart from the predominately amorphous silica particles, a small percentage of larger strontium aluminum fillers are incorporated as a reinforcing phase. The latter is possible to cause the rounded projections found on its surface.

Gloss is an important property and is used primarily as a measure of surface shine [4]. A perfect mirror surface is said to have maximum gloss. Gloss is the number assigned for the reflectance value of a surface. As more direct light is reflected, higher gloss value is recorded which indicates a smooth and high-luster surface [27, 28]. It is obvious that high gloss for a resin composite gives a natural, aesthetic appearance to a restoration [29].

Generally, gloss values obtained by various measurement methods depend on experimental conditions such as spectral distribution of the light and viewing angle. According to ISO 2813, ASTHD 523 and 2457 and DIN 67530 semigloss surfaces should be measured with 60° angle of illumination, which was applied in the current study. Although a wide distribution of values was recorded, all the materials tested can be characterized as being "semigloss" because the data fall into the range of 10 to 70 units. Furthermore, 60° angle gloss measurements are considered more reliable from a clinical perspective since it is closer to the angle from which the average person will observe the surface. Smoother resin materials with lower surface relief, as found under AFM evaluation, gave higher gloss values. It is an interesting finding that gloss values were directly linked to S_a values obtained by AFM but not to R_a. Thereby, the validity of the AFM method for surface roughness measurements, was further emphasized.

Conclusions

(a) Micro-roughness showed a high correlation with gloss values, whilst macro-roughness did not.
(b) The AFM method was more suitable to distinguish surface roughness than compared was 2-D profilometry, and was able to give a more detailed definition of surface texture than SEM.
(c) Microhybrid resin-composites presented higher gloss and lower roughness values, compared with the hybrid resin composite evaluated, but were comparable to the reinforced microfill.

References

1. A. TJAN and C. CHAN, *J. Prosth. Dent.* **61** (1989) 138.
2. N. DE JAGER, A. FEILZER and C. L. DAVIDSON, *Dent. Mater.* **16** (2000) 381.
3. http://www.dentsply.co.uk/products/products/esthetx.htm.
4. P. CAMPBELL, W. JOHNSTON and W. J. O'BRIEN, *J. Dent. Res.* **65** (1986) 892.
5. D. JUDD and G. WYSZECKI, in "Color in Business, Science and Industry.' (John Wiley & Sons, Inc, New York, 1975) p. 397.
6. S. WHITEHEAD, A. SHEARER, D. C. WATTS, N. H. F. WILSON, et al., *Dent. Mater*. **15** (1999) 79.
7. A. WENNERBERG, R. OHLSSON, B. G. ROSEN and B. ANDERSSON, *Med. Eng. Phys.* **18** (1996) 548.
8. S. JONIOT, G. GREGOIRE, A. M. AUTHER and Y. M. J. ROQUES, *Oper. Dent.* **25** (2000) 311.
9. S. WHITEHEAD, A. SHEARER, D. C. WATTS and N. H. F. WILSON, *J. Oral. Rehab.* **22** (1995) 421.
10. S. WHITEHEAD, A. SHEARER, D. C. WATTS and N. H. F. WILSON, *Dent. Mater.* **12** (1996) 315.
11. G. MARSHALL, M. BALOOCH, R. J. TENCH, M. KINNEY and S. J. MARSHALL, *Dent. Mater.* **9** (1993) 265.
12. N. SILIKAS, A. R. LENNIE, K. E. R. ENGLAND and D. C. WATTS, *Microscopy and Analysis* **82** (2001) 19.
13. A. YAP and B. MOK, *Oper. Dent.* **27** (2002) 161.
14. T. RYBA, W. DUNN and D. F. MURCHISON, *Oper. Dent.* **27** (2002) 243.
15. A. WILDER, E. SWIFT, K. N. MAY, J. Y. THOMPSON and R. A. MCDOUGAL, *J. Dent.* **28** (2000) 367.
16. B. VAN MEERBEEK, M. VARGAS, S. INOUE, Y. YOSHIDA, J. PERDIGAO, P. LAMBRECHTS and G. VANHERLE, *Am. J. Dent.* **13** (2000) 3D.
17. N. ABU-BAKR, L. HAN, A. OKAMOTO and M. IWAKU, *Dent. Mater. J.* **20** (2001) 172.
18. L. MARIGO, M. RIZZI, G. LA TORRE, G. RUMI, 3-D. *Oper. Dent*. **26** (2001) 562.
19. K. STOUT and L. BLUNT., in "Three-Dimensional Surface Topography" (Penton Press, London, 2000) p. 178.

20. F. TAY, S. SIKDAR and M. MANNAN, *J. Materials Processing Technology* **120** (2002) 243.
21. R. WASSELL, J. F. MCCABE and A. W. WALLS, *Dent. Mater.* **10** (1994) 269.
22. C. BOLLEN, P. LAMBRECHTS and M. QUIRYNEN, *Dent. Mater.* **13** (1997) 258.
23. R. WEITMAN and W. EAMES, *Oral Health* **65** (1975) 29.
24. R. WEITMAN and W. EAMES, *J. Am. Dent. Assoc.* **91** (1975) 101.
25. H. SHINTANI, J. SATOU, H. HAYASHIHARA and T. INOUE, *Dent. Mater.* **1** (1985) 225.
26. K. CHUNG, *Dent. Mater.* **10** (1994) 325.
27. F. BILLMEYER and R. ABRAMS, *J. Paint. Technol.* **45** (1973) 23.
28. I. BRIDGEMAN, in "The nature of light and its interaction with matter. Colour physics for industry" (H Charlesworth & Co Ltd, Huddersfield, 1987) p. 1.
29. R. HUNTER, in "The measurement of appearance" (Wiley, New York, 1975) p. 65.