Influence of the size and distribution of filler particles on the colour of resin composites

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Abstract

Resin composites are the most used biomaterials in dental restoration. A general new methodology based in high contrast Fe-SEM (Field emission scanning electron microscope) images is proposed to characterize the distribution and size of the particles of resin composites for the evaluation of their influence on the color of these materials. The digital images were treated and analyzed using the public domain software ImageJ. The color coordinates in CIELAB were obtained using a spectroradiometer with the experimental conditions recommended by the CIE. The results show that the color coordinates of resin composites are more influenced by the particle size than by the amount of filler particles or the filler distribution. This proposed method can be used for the analysis of other biomaterials in which the color is an important parameter.

Introduction

The photoactivated resin composites are the most employed materials in dental restoration. In dentistry, posterior class I or II restorations require composites that show high mechanical properties, while anterior restorations need composites that have superior aesthetics. Therefore, a suitable and correct dental restoration requires that the optical characteristics of these biomaterials are similar to those of the recovered tooth.[1,2,3] Between these characteristics it is possible to emphasize the colour, because only when the colour of composite equals the colour of the tooth, the restoration will be considered satisfactory by the patient.[4]

The actual resin composites are composed basically of two components: 1.) organic matrix (continuous phase); 2.) inorganic filler particles (disperse phase). The two components are very different; among them chemical connection does not exist, obtaining its union through the denominated phase of connection (silane). The hardening of the material is obtained with photoactivation. The inorganic filling improves the properties of the material from a mechanical point of view (reduction of the contraction of polimerization, diminution of the water absorption, increases the hardness and increases the resistance to the fracture) and also is responsible, to a great extent, of its optical characteristics, according to the described recently for traditional composites. Recently, a study[5] show that, by comparing modeled surfaces generated by Matlab for different materials, it is possible to determine how different variables such as filler type, filler surface treatment and light source affect light attenuation.

For that reason, in the last decades, the investigation and, therefore, the advances in this field have taken place basically to expenses of the improvement of the inorganic filler. For this reason, the characteristics of this phase (size of the filling particle, volumetric fraction and morphology) are allowing the classification of the different resin composites.

A recent work[6] seems to indicate that the colour coordinates of the resin composites especially L^*, might be influenced by the filler distribution. Therefore, the colour study of current dental materials, just as the development of biomimetic restorative materials needs the knowledge of the filler distribution. In this study[6], experimental resins with an ideal inorganic filler with a homogeneous distribution of particles with same size are employed and they are very different from commercial resin composites, reason why results about the colour of composites cannot be conclusive.

On the other hand, a new study[7] has evaluated the weight percentage of filler of the inorganic fraction by thermogravimetric analysis and the morphology of the filler particles with scanning-electron microscopy in nanofilled, universal and microfilled composites, in order to determine the mechanical properties. However, in this work the filler distribution of the resin composites is not analyzed and the filler shape is studied with a not quantitative method.

The aim of this work is to propose a new method for the evaluation of the filler inorganic distribution employing high contrast Fe-SEM (Field emission scanning electron microscope) images and digital images treatments that allow to know the influence of the filler distribution on the resin composites colour.

Materials and methods

In this study, three commercial resin composites have been evaluated: IntenS, A110 and Z100. The main characteristics of those materials are indexed in the Table1. The resin composites have the same polymeric matrix (BisGMA, BisEMA, UDMA and TEGDMA) and the same type of filler particle types (zirconia/silica), but the particle size is different for each resin composite.

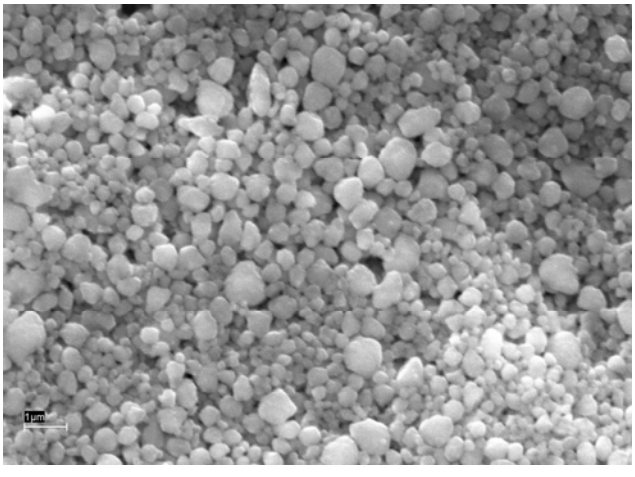
In our study we evaluate microfilled, hybrid and microhybrid resin composites[8]. The fillers morphology was determined using SEM. Unpolymerized monomers were removed by a washing technique: approximately 0.5 g of each composite was dissolved in acetone pro analysi (ACS, ISO, 14.100; Merck, Darmstadt, Germany), mixed and centrifuged for 30 min at 3000 rpm. This procedure was then repeated three times to completely remove remnants of resin matrix. The supernatant was again removed and the remaining filler particles dried at 37°C for 12 h. Finally, the powder was ultrasonically agitated to reduce agglomeration of filler particles. All powders were screened under Fe- SEM to confirm complete filler isolation and to investigate filler morphology.

A110	
Resin composite type	Microfill
Organic composition	Bis-GMA/TEGDMA
Filler size from manufacturer	0,01 – 0,09 µm
Manufacturer	3M
Z100	
Resin composite type	Microhybrid
Organic composition	Bis-GMA/TEGDMA
Filler size from manufacturer	0,01 – 3,5 µm
Manufacturer	3M
IntenS	
Resin composite type	Hybrid
Organic composition	Bis-GMA/UDMA
Filler size from manufacturer	0,2 – 7 µm
Manufacturer	Ivoclar-Vivadent

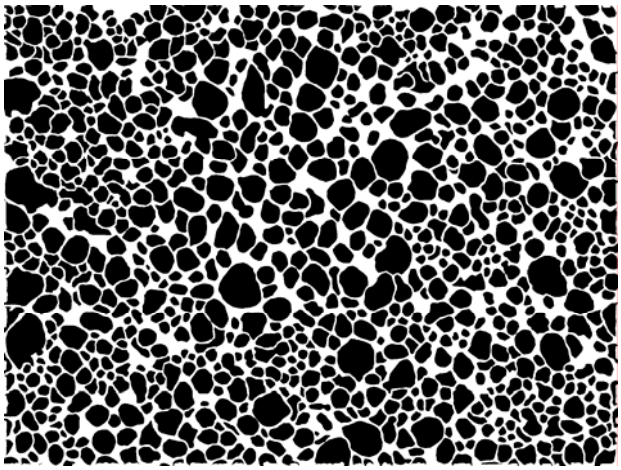
Table1: Characteristics of the three studied resin composites

The Fe-SEM (Field emission scanning electron microscope) original images are grey scale images with a 2048x1536 pixels resolution. A digital image treatment was applied using the public domain software for image analysis ImageJ (NIH, Bethesda, Maryland, USA). In order to facilitate the particle selection, the brightness, contrast and levels of the original images were modified. The particle selection was made employing a professional drawing tablet, an optical pen and the ImageJ software. Then, the images were translated to binary images, where black meant particle and white meant background. The measurement steps, including particle counting, area measurements, and filler distribution, were performed with the software for image analysis ImageJ (Figure1).

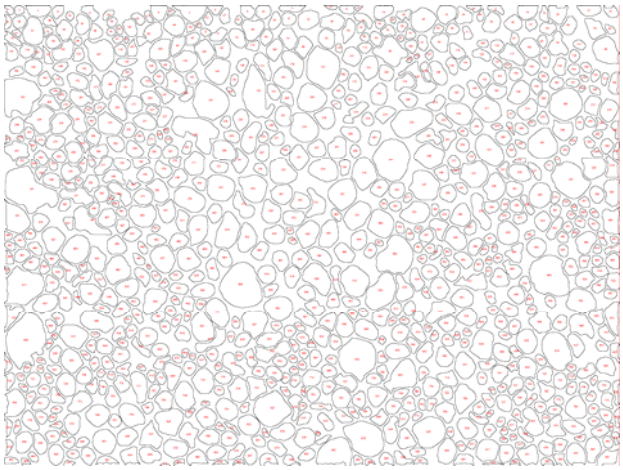
Figure 1. Example of digital image analysis process of resin composite A110



a.) Original image with high contrast



b.) Binary image with particle selection



c.) Final image with particle counting obtained with the ImageJ software

For the colour measurement, the resin composites were manipulated according manufacturers instructions and packaging in a silicon mold of 5mm diameter and 4mm thickness polymerized with a halogen light curing unit. It was then polished with a 2000 grit SiCa paper, a habitual polish in clinical practice.

Colour measurements were made with a spectroradiometer (SpectraScan PR-704, Photo Research Inc. Chatsworth, USA) with a 4% of accuracy. Samples were placed in a colour assessment cabinet (CAC portable, Verivide Limited, Leicester LE3 5AG, England), with a source simulating the relative spectral irradiance of CIE standard illuminant D65. A CIE 45º/dº geometry and the CIE 1964 10º supplementary standard colorimetric observer were employed. Measurements were repeated three times for each resin composite.

Results and discussion

The CIELAB chromaticity coordinates for each material are indexed in Table2

Resin composite	L^*	a^*	b^*
A110	74,7	3,1	7,0
Z100	64,7	5,5	16,6
IntenS	66,4	6,14	18,9

Table2: CIELAB chromaticity coordinates for the three resin composites

Figures from 2 to 4 show the distribution in size (% particles of specific area) for each one of the studied materials.

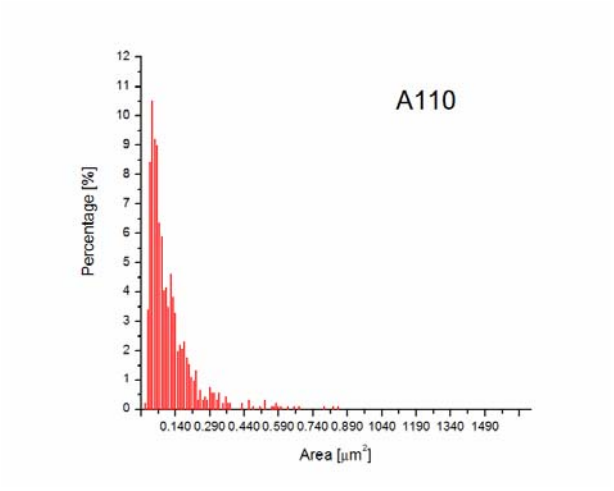


Figure2: A110 Filler Distribution

From Fig.3 and Fig.4, IntenS and Z100 show a total percentage (sum of percentages) of 84.14% and 91.31% for particles smaller than 1µm^2, respectively. However, in the case of A110, considering only particles smaller than 0.35µm^2, we obtain a total percentage of 97%. This result indicates that there is an important difference between the size of the particles of microfilled resin composite and the particles of the hybrid and microhybrid resin composites.

It is noteworthy that the distribution of the particles bigger than 1µm^2 is different between hybrid and microhybrid resin composites. In the case of Z100, there is a minor number of particles with an area bigger than 1µm^2 compared with the case of IntenS, where the distribution is more homogeneous.

The data on acceptability and perceptibility limits in the dental literature are somewhat arbitrary. However some research results[11,12] provide useful orientation; a ΔE^*_{ab}=1 was found to be 50:50% perceptibility threshold under controlled conditions (50% of observers will not detect colour difference), while a ΔE^*_{ab}=2.7 was found to be a 50:50% acceptability threshold (50% of observers will reject the restoration because of colour mismatch) for monochromatic specimens in laboratory conditions (viewing booth). A recent study, performed on polychromatic denture teeth in relatively controlled clinical conditions reported ΔE^*_{ab} of 2.6 and 5.5 as perceptibility and acceptability thresholds[13]. This large discrepancy with previous findings probably originates from the polychromatic nature of compared teeth, difference in evaluated area between the instrumental measurements (D=1mm) and visual comparisons (whole labial surface) and possible lack of controlled conditions in visual comparisons (illuminant, optical geometry and visual angle).

Considering the values of L*, a* and b* for each studied specimen, the colour difference obtained between IntenS and Z100 (ΔE^*_{ab}=2,92) is close to the perceptibility threshold (ΔE^*_{ab}=2,60). Nevertheless, the colour differences between A110 and IntenS and Z100 are ΔE^*_{ab}=14,82 and ΔE^*_{ab}=14,06 respectively. These values are much bigger than perceptibility and acceptability thresholds for dental materials.

In the paragraph above, A110 presents a notable difference in particle size respect to the other studied materials. Thus, our results seem to show that the major influence in the colour of the composites is due to the size of the filler particles, and not to its amount or its distribution as reported by Lim et al.[6] This difference can be due to the different scattering pattern. A study which involves more materials will be necessary to confirm these preliminary results.

To conclude, we want to highlight that this new method can also be used in the study of other types of biomaterials, like microcellular solids and hidrogels.

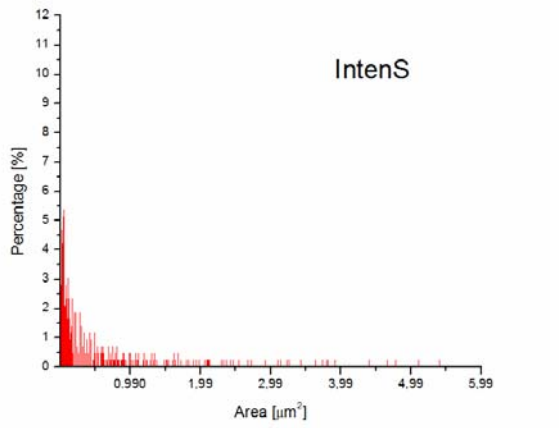


Figure3: IntenS Filler Distribution

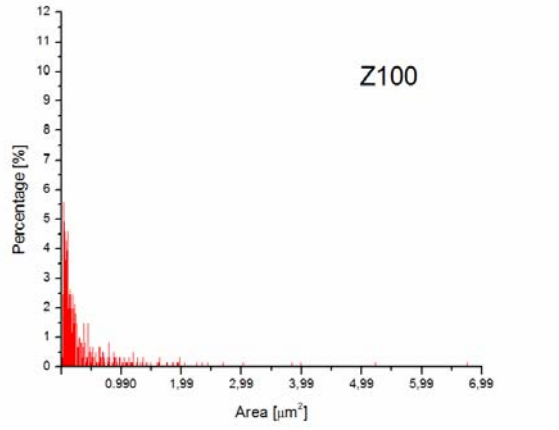


Figure4: Z100 Filler Distribution

Acknowledgements

The authors would like to thank the manufacturers (3M and Ivoclar-Vivadent) for supplying materials and light curing unit.

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Razvan Ghinea received his BS in Physics from the Craiova University in 2007. He joined the Biomaterials Optics Laboratory of Granada University in October 2007. He has been working in determining the optical properties of the resin composites. Actually he is a student of Master in Methods and Advanced Techniques in Physics (UGR). He is funded by a Student Mobility in the frame of Socrates Programme: Higher Education (Erasmus) scholarship.