COBEM2005-0946 – SEM/EDS ANALYSIS OF MICROSTRUCTURE OF DENTAL RESTORATIVE SYSTEMS: INFLUENCE ON OPTICAL DENSITY

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Abstract. *The aim of this study is to evaluate and compare the microstructure and chemical composition of dental restorative systems and its influence on optical density using the technique of scanning electron microscopy (SEM) associated to electron dispersive spectroscopy (EDS). Samples of the composite resins Durafill VS® and Z250® and the glass ionomer cement Ketac-Fil®chemically cured, thickness of 1 to 4mm, had been made using matrices n Teflon® of previously determined diameter. The metallic covering of the samples was carried out to allow the evaluation by SEM and EDS microprobe. Three radiographies were taken from samples of each material. After development, the optical density of the material images was messured by mean of a transmission densitometer. Three readings were taken from each radiographic image. The optical density value for each specimen was the arithmetic mean of the three readings. The statistics anlysis by Kruskall-Wallis, ANOVA and Mann-Whitney (p< 0,05) revelead that, for all materials, increase in thickness showed reduction on optical density and these values are statistically significant. Durafill VS® showed lower radiopacity than Z250® and Ketac-Fil®. Variations in microstructure and in chemical composition obtained by SEM-EDS analysis would explain the variation on radiographic aspects of the materials evaluated.*

Keywords *:dental composite, optical density, radiopacity, glass ionomer, microstructute*

1. Introduction

In routine clinical practice, the diagnosis of recurrent caries, the assessment of excess restorative material on the cervical margins of proximal surfaces, the identification of internal voids and contacts with adjacent teeth continues to be a problem (1). So, periodical clinical evaluation is frequently associated to radiographic examination.

Radiopacity is a desirable feature for most dental materials. Inadequate radiopacity of restorative materials decreases X-ray diagnostic information and can be a contributing factor in faulty interpretation (2). Filler particles composed by high atomic number elements (barium, zircon, zinc and lanthanum metal) have been incorporated to organic matrix of polymeric materials in order to improve radiographic quality (3). The question of how radiopaque a restorative material should be, for optimal diagnostic utility, has been investigated by several authors and there is no clear agreement on the degree of radiopacity that provides the best conditions for radiographic detection of caries and defects adjacent to

restorations (2). While a composite with a radiopacity grater than dentin meets ISO standard 4049, most studies concluded that a restorative material should be approximately as radiopaque as enamel (4).

The aim of this study is to evaluate the microstructure and chemical composition of dental restorative systems and its influence on optical density using the technique of scanning electron microscopy (SEM) associated to electron dispersive spectroscopy (EDS).

2. Materials and methods

The selection of the restorative systems it was made on basis in the commercial marks routinely used by dentistry. It had been analyzed two light-cured composite resins (Durafill® – Haerus-Kulzer, and Filtek-Z250® – 3M/ESPE, respectively a microfiller and a microhybrid composite resin) and one glass ionomer cement chemically cured (Ketac-Fil® – 3M/ESPE) for cavity liner selected for its physical properties similar to dentin. All the samples had been maked in the color A3 (VITA scale), so that this not became one variable to influence the results.

2.1. Making of samples

Samples of light cured composite resins Filtek-Z250® and Durafill VS® were obtained using Teflon® matrices of previously diameter and thickness of 1 to 4mm. This variation on thickness is justified by the diversity in depth of carie lesions restored with these materials. For light-cured composite resins, the samples of 1mm and 2mm had been gotten from insertion of one increment in the respective matrices, and its light cure in a Ultralux® device (Dabi Atlante) duly calibrated for Demetron® radiometer to supply the energy praised for the manufacturer. The fulfilling of the matrices of 3mm and 4mm in two increments resulted in samples in these thicknesses. This procedure was adopted aiming to guarantee that these samples suffered uniform polymerization in all its thickness. The selected composite resins had been inserted with appropriate instrument and polymerized in accordance with manufacturer instructions for 40 seconds, being overcome the care to design the superior part of the samples with a glass plate. The samples of glass ionomer cement had been obtained from proportion (powder:liquid) and manual manipulation in cooled glass plate, in accordance with manufacturer instructions. To minimize the incorporation of internal bubbles in these samples, the material was inserted in only increment with a Centrix® device. Soon observe the loss of surface brightness; the samples had been protected to minimize the effect of ambient conditions. It was waited the initial setting time (7 minutes) for removal of the samples of matrices.

2.2. Microstructural analysis

For the study of microstructural features (size, form and distribution of filler particles) of the selected materials, it was used the Scanning Electron Microscopy (SEM). For this, the samples had received metallic covering with Au to make possible the electron beam propagate and formation of images. For the chemical elementary analysis of materials it was selected the microprobe EDS (Energy Dispersive Spectroscopy). To allow to this, the samples had received covering with carbon. In this analysis, five distinct points in surface of each sample had been evaluated and from this a half-quantitative of the main elements/composition was obtained.

2.3. Optical density by the conventional method – Direct densitometry

How widely described in literature filler particles of determined nature (chemical composition, size and form), composed for high atomic number elements (as barium, zircon, zinc, ytterbium and lanthanum metal), have been incorporated to dental materials in order to improve radiographic quality, getting a similar or superior radiopacity of the dental enamel permitting assessment of interproximal contour of restorations, identifies internal voids within the material as well as secondary caries or decalcified dentin. Three radiographs were obtained by the arrangement of the samples in a periapical film (Ektspeed Plus® - Eastman Kodak) exposed for 0,4 seconds at 70kV, 8,0mA, 50/60Hz and a 200,0mm target-film distance with a X-ray machine (Dabi-Atlante®). Films were developed automatically and the radiographic images submitted to reading in a transmission densitometer (X-Rite®- Black and Transmission Densitometer) calibrated by 301 Industrex® - Calibrated Step Tablet - KODAK in order to determine the optical density by direct method. Five readings were taken from each specimen's radiographic image. The optical density value for each specimen was the arithmetic average of the five readings.

3. Results and discussion

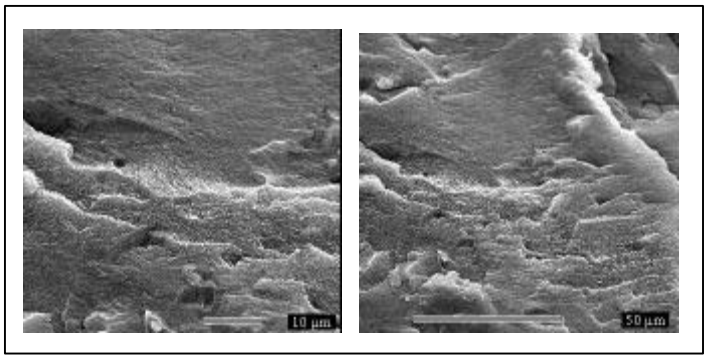

Figure 1: SEM of composite resin Durafill VS® – 750X and 1500X

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 Number of Oxygen Atoms = 10          Chi_square = 1029.9919

Element Weight%  Cation    ZAF      Z      A      F
   SO3    8.296  0.2325  1.0128 1.0957 0.9241 1.0003
   CO2   90.170  4.5977  2.2774 1.0366 2.1971 0.9999
 Al2O3    0.861  0.0379  1.1052 1.0030 1.1024 0.9995
  SiO2    0.673  0.0251  0.9116 0.9952 0.9168 0.9991
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Total   100.000  4.8932
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Figure 2: Chemical analysis by EDS – Durafill VS®

Table 1: Optical density – Durafill VS®

RADIOGRAPHIC	THICKNESS			
	1,0mm	2,0mm	3,0mm	4,0mm
T1	1,786	1,712	1,654	1,494
T2	1,692	1,642	1,534	1,42
T3	1,762	1,708	1,59	1,454
Média	1,746	1,687	1,592	1,454

* Average values – Three measures by densitometer

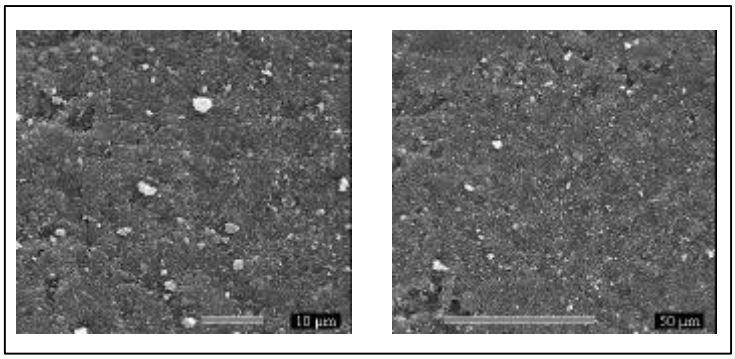

Figure 3: SEM of composite resin Filtek-Z250® – 750X and 1500X

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 Number of Oxygen Atoms =  0          Chi_square = 1779.5602

Element Weight%  Cation    ZAF      Z      A      F
  SiO2   64.993  0.0000  0.8912 0.9834 0.9088 0.9972
  ZrO2   11.324  0.0000  1.5471 1.3381 1.1562 1.0000
 Al2O3    2.532  0.0000  1.0514 0.9916 1.0721 0.9890
  Na2O    0.066  0.0000  1.3696 1.0337 1.3261 0.9991
   CO2   21.085  0.0000  6.0649 1.0271 5.9051 1.0000
-------------------------------------------------------------
Total   100.000  0.0000
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Figure 4: Chemical analysis by EDS – Filtek-Z250®

Table 2: Optical density – Filtek-Z250®

RADIOGRAPHIC	THICKNESS			
	1,0mm	2,0mm	3,0mm	4,0mm
T1	1,130	0,716	0,570	0,450
T2	1,100	0,670	0,560	0,462
T3	1,074	0,726	0,564	0,430
Média	1,101	0,704	0,564	0,447

*Average values – Three measures by densitometer

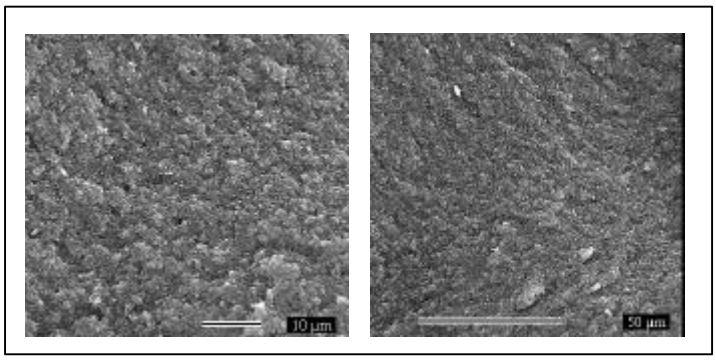

Figure 5: SEM of glass-ionomer Ketac-Fil® – 750X and 1500X

Number of Oxygen Atoms = 1 Chi_square = 459.0441

Element	Weight%	Cation	ZAF	Z	A	F
Al2O3	11.553	0.0699	1.1619	0.9798	1.1928	0.9941
SiO2	52.913	0.2716	0.9947	0.9717	1.0238	0.9999
BaO	12.004	0.0241	1.1729	1.2019	0.9758	1.0000
CO2	23.281	0.1632	5.0211	1.0153	4.9456	1.0000
CaO	0.249	0.0014	0.9730	0.9811	1.0042	0.9876
Total	100.000	0.5302				

Figure 6: Chemical analysis by EDS – Ketac-Fil®

Table 3: Optical density – Ketac-Fil®

RADIOGRAPHIC	THICKNESS			
	1,0mm	2,0mm	3,0mm	4,0mm
T1	1,072	0,824	0,622	0,514
T2	1,040	0,804	0,614	0,502
T3	1,105	0,820	0,564	0,536
Média	1,072	0,816	0,600	0,517

* Average values – Three measures by densitometer

The statistic analysis (ANOVA, Kruskal-Wallis and Mann-Whitney tests) reveals for all materials an inverse relation between thickness and optical density: lower values of density have represented greater radiopacity and these results were significantly different. These results are showed in Tables 1, 2 and 3. The composite Durafill VS® presented for all thickness higher values of optical density than the observed for Filtek-Z250® and Ketac-Fil® indicating lower radiopacity, disfavoring the contrast with dental structure and areas of carie lesions. Data of elementary chemical composition (EDS) is showed in Figures 2, 4 and 6. It can be observed that the composite Filtek-Z250® presented 11,324% of ZrO_2, higher levels (64,993%) of SiO_2 when comparated with Durafill VS® and Ketac-Fil® suggesting that these compounds are responsible for the highest radiopacity observed. So that Durafill VS® has lower rates of Al_2O_3 (0.861%) and SiO_2 (8,296%). In chemical composition of Ketac-Fil were observed BaO (12,004%) and SiO_2 (52,913%). By analysis of optical density values the glass ionomer cement showed similar radiopacity when compared to Filtek-Z250®. The images obtained by SEM (Figures1, 3 and 5) have showed variations in the size and distribution of filler particles, suggesting that it could influence the propagation of X-rays through the material, intervening in the values of optical density and, consequently, in the radiographic aspect of the studied composite resins.

4. Conclusions

On the basis of the results obtained in this experiment, we can be concluded that:
1 - The SEM/EDS analysis showed variations on microstructure of the materials selected.
2 – Differences on the chemical composition of the evaluated materials suggest influence on radiographic behavior.
3 - For all the materials evaluated in this study, how much higher the thickness of the samples lower the optical density.
4 - A composite resin Filtek-Z250® showed lower values of optical density, therefore is presented more radiopaque, mainly when compared to composite Durafill VS®, one revealed less radiopaque. The glass ionomer cement Ketac-Fil® evaluated in this study had been showed optical density sufficiently similar to the ones of a composite resin (Filtek-Z250®).

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6. References

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