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Influence of radiopaque fillers on physicochemical properties of a model epoxy resin-based root canal sealer

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ABSTRACT

Objective: To verify the influence of radiopaque fillers on an epoxy resin-based sealer. Material and Methods: Experimental sealers were formulated by adding 20%, 40%, 60%, 80%, 100% and 120% of calcium tungstate, ytterbium trifluoride or barium sulphate by weight to an epoxy-resin-base. Setting time, flow, film thickness, radiopacity, sorption, solubility, pH and push-out bond strength were evaluated. Results: The setting time ranged from 373 to 612.66 min, the flow varied from 13.81±0.49 to 22.49±0.37 mm, and the film thickness ranged from 16.67±5.77 to 33.33±11.54 µm. The lowest pH was 5.47±0.53, and the highest was 6.99±0.03. Radiopacity varied from 0.38±0.04 to 2.57±0.21 mmAl and increased with the amount of filler. Calcium tungstate sealers had a higher sorption and solubility than other sealers. There was no significant difference in the push-out bond strength among the fillers at the 120% concentration. Conclusion: The inorganic fillers evaluated and their concentrations affect the physicochemical properties of an epoxy resin-based root canal sealer.

Key words: Tungsten. Ytterbium. Barium. Root canal filling materials.

INTRODUCTION

The success of endodontic therapy depends on a complete cleaning and three-dimensional filling of the root canal system. For ideal obturation of the canal, sealers must have certain characteristics, such as high radiopacity, flowability, adhesion to canal walls, reliable working time, biocompatibility and low solubility. Although no cement has all of these properties, some sealers, such as zinc oxide-eugenol-based, calcium hydroxide-based, glass-ionomer and resin-based, are clinically acceptable and widely used[23,30]. Despite this, epoxy resin-based cement presents suitable physicochemical properties[2,7] and satisfactory clinical outcomes[13]. One of the most important properties of endodontic sealers is radiopacity. Cement radiopacity enhances diagnostic procedures and facilitates the diagnosis in Endodontics. To reach an acceptable radiopacity, inorganic fillers are added to endodontic sealers. Furthermore, these fillers improve the physical, chemical and mechanical properties of the sealer, such as viscosity and film thickness.

The radiopacity of dental materials depends on the composition of the materials (e.g., radiopaque agents) and the filler concentration. X-rays are either absorbed and/or scattered by material to produce radiographic images. Adequate radiopacity in images is necessary to avoid overtreatment and false-positive results and to obtain the proper clinical diagnosis. The chemical structures of agents that produce radiopacity in dental materials should be considered to obtain an acceptable material because the atomic number, density and size of fillers influence the radiopacity[3]. Elements with a high atomic number can absorb or reflect more X-rays leading to increased radiopacity[1]. Besides the effects on cement radiolucency, the inorganic fillers

could affect the polymerization shrinkage[11], degree of conversion[4], polymerization rate[11] rheological properties[4] and water absorption[14] when added to resins.

Calcium tungstate ($CaWO_4$) and barium sulphate ($BaSO_4$) are already used as fillers in commercial sealers with epoxy-resin and gutta-percha cones[12,27], providing high level of radiopacity[27]. Ytterbium trifluoride (YbF_3) contains ytterbium, which is in the lanthanide series with a high atomic number leading to a high opacity to X-rays. In addition, this filler is very translucent, which is a good optical characteristic for dental materials. Ytterbium trifluoride has already been incorporated in methacrylate-based cements[5] and has shown favourable results for an endodontic sealer. However, additions of these radiopaquing fillers on epoxy resin-based root canal sealers properties were barely studied.

The purpose of this study was to evaluate the influence of the addition of different inorganic fillers on the physicochemical properties of an epoxy-resin based root canal sealer.

MATERIAL AND METHODS

Endodontic cements were prepared using an epoxy resin and inorganic fillers. The resin is composed of a bisphenol-A (Araldite® LY 1564, Huntsman Advanced Materials Química Brasil Ltda, Taboão da Serra, SP, Brazil) and a cycloaliphatic amine (Aradur 2963, Huntsman Advanced Materials Química Brasil Ltda, Taboão da Serra, SP, Brazil), base and catalyst, respectively. Base and catalyst were added at 2:1 ratio by weight. Calcium tungstate ($CaWO_4$) (American Elements, Los Angeles, CA, USA), ytterbium trifluoride (YbF_3) (American Elements, Los Angeles, CA, USA) and barium sulphate ($BaSO_4$) (Labsynth Produtos para Laboratório Ltda, Diadema, SP, Brazil) at 20%, 40%, 60%, 80%, 100% and 120%, in weight, were added to the amount of epoxy resin base. Colloidal silica nanoparticles (7 nm) (Aerosil 380, Evonik Industries AG, Hanau, Hesse, Germany) were added at 0.5% by weight to adjust the viscosity of cements. Base, catalyst and inorganic fillers were manually mixed for 90 s on a glass plate until obtaining a homogeneous paste, checked visually. A control group without radiopaquing agent filler was included. The sealers were subjected to laboratory tests to characterize selected physicochemical properties, as following.

Particle size

The particle size distribution of each filler was assessed using a laser diffraction particle size analyser (CILAS 1180 Particle-Size-Analyzer, Compagnie Industrielle des Lasers, Orleans, Loiret, France).

Setting time

The setting time of the experimental sealers was analysed according to ISO 6876:2001[15]. The experimental sealer was placed in a silicone matrix that had an internal diameter of 10 mm and a height of 1 mm. An indenter with a mass of 100±0.5 g and a flat end with a diameter of 2.0±0.1 mm was placed vertically on the horizontal surface of the sealer for 2 s. The surface was then visually evaluated for indentations. Indentations were repeated every 30 min until no indentation was observed, and at this moment the setting time was recorded. The mean value of three measurements for each group was recorded for the setting time of the material.

Flow

The flow test was conducted according to ISO 6876:2001[15]. A total of 0.5 ml of each experimental sealer was placed on a glass plate (40x40x5 mm) using a graduated 1.5 ml syringe. Another plate with a mass of 20±2 g and a load of 100 g was applied to the top of the material for 180±5 s after mixing. Ten minutes after mixing, the load was removed, and the major and minor diameters of the compressed material were measured using a digital caliper. The results were recorded if both measurements were consistent to within 1 mm. If the major and minor diameter discs were not uniformly circular or did not agree within a range of 1 mm, the test was repeated. For each experimental group, the test was conducted three times and the mean value was recorded for the flow.

Film thickness

The film thickness was evaluated according to ISO 6876:2001[15]. Two glass plates that were 5 mm and 10 mm thick were placed together and their combined thickness was measured. A total of 0.5 mL of experimental sealer was placed at the centre of one of the plates, and a second plate was placed on top of the material. A 150 N load was applied vertically on the top of the glass plate 180±5 s after mixing. Ten minutes after the start of mixing, the thickness of the two glass plates and the sealer film was measured using a digital caliper. The difference in the thickness of the two glass plates, with and without sealer, was the film thickness of the experimental sealer. The mean value of three measurements for each sealer was recorded for the film thickness of the material.

Radiopacity

The radiopacity of the experimental sealers was tested according to ISO 6876:2001[15]. Five specimens that were 10.0±0.1 mm in diameter and

1.0±0.01 mm thick were produced. Radiographic images were obtained using a phosphor plate digital system (VistaScan, Dürr Dental GmbH & Co. KG, Bietigheim-Bissingen, Baden-Württemberg, Germany) at 70 kV and 8 mA with a 0.2 s exposure time and a 400 mm focus-film distance. One specimen with the same percent filler from each group was positioned with a specimen from the control group for each film; there were a total of four specimens *per* film. In all images, an aluminium step-wedge was simultaneously exposed with the specimens. The aluminium step-wedge thickness ranged from 0.5 to 9.0 mm in increments of 0.5 mm. The aluminium alloy used was Al 99.12, Fe 0.47, Mg 0.41 and <0.1 of Cu (% by weight) according to ISO 6876. The images were saved in TIFF format and analysed using Photoshop (Adobe Systems Incorporated, San Jose, CA, USA). The means and standard deviations of the grey levels (pixel density) of the aluminium step-wedge and the specimens were obtained in a standardised area of 1.5 mm^2 [5].

Sorption and solubility

Water sorption and solubility tests were performed according to ISO 4049:2009[16], except for the specimen dimensions. The specimens had a diameter of 10±0.1 mm and a thickness of 1.0±0.1 mm. The specimens were placed in desiccators containing silica gel at 37°C. Each specimen was weighed repeatedly at 24 h intervals on an analytical balance (AUW220D, Shimadzu Philippines Manufacturing Inc, Cavite, Calabarzon, Philippines) until a constant mass (m_1) was obtained (i.e., until the specimen's decrease in mass was no more than 0.1 mg in a 24 h period). The diameter and thickness were measured with a digital caliper to calculate the volume (V) in mm^3. Then, the specimens were placed in a light-free container with distilled water at 37°C for 7 days. The specimens were removed from the liquid, and each specimen was weighed after being dried slightly. The weight was recorded (m_2). The procedures to obtain m_1 were repeated to obtain m_3. Water sorption and solubility in micrograms *per* cubic millimetre were calculated according to a previous study[6].

pH

The pH of the sorption and solubility water was evaluated using a digital pHmeter (pH 21, Hanna Instruments, São Paulo, SP, Brazil). Five measurements were performed *per* group.

Push-out bond strength

Thirty bovine incisors were sectioned transversely 15 mm from the apex, the pulp tissue was removed and the chemo-mechanical preparation was performed. Teeth were divided into 3 groups, and the canals were obturated by lateral condensation technique with gutta-percha points and the 120% concentration of calcium tungstate, ytterbium trifluoride and barium sulphate experimental cements. The roots were stored at 100% humidity and 37°C for 7 days. Subsequently, the roots were sectioned transversely into 7 slices that were approximately 0.7 mm thick using a low speed disc (Isomet, Buehler Ltd, Lake Bluff, IL, USA) with constant water cooling. The internal diameter of canal of each slice was measured with a digital caliper (Digimess, 100.174BL, Digimess Instrumentos de Precisão Ltda, São Paulo, SP, Brazil) and the contact area between the filling and dentin of each slice was calculated. Each slice was placed with the apical side up on a mechanical testing machine (DL-2000, EMIC Equipamentos e Sistemas de Ensaio Ltda, São José dos Pinhais, PR, Brazil). A force was placed on the shutter towards the apical-neck using a 500 N load cell and a cross-head speed of 1 mm/min with a 1 mm diameter cylindrical device. The bond strength (MPa) was obtained by dividing the required force (N) to displace the filling material by the adhesive area (mm^2).

Statistical analysis

Data normality was checked by the Kolmogorov-Smirnov test. Differences among filler compositions and control group were detected using ANOVA and the Tukey *post-hoc* test. Linear regression was performed to determine the influence of filler composition on radiopacity. A significance level of 5% was used for analysis.

RESULTS

The mean particle sizes of calcium tungstate, ytterbium trifluoride and barium sulphate were 17.79 µm, 14.37 µm and 4.86 µm, respectively. Table 1 shows the setting time, flow, film thickness, sorption, solubility and water pH of experimental sealers. The setting time of experimental sealers ranged from 373 to 612.66±4.71 min, the flow of experimental sealers ranged from 13.81±0.49 to 22.49±0.37 mm and the film thickness ranged from 16.67±5.77 to 33.33±11.54 µm. The pH of experimental sealers ranged from 5.47±0.53 to 6.99±0.03.

Radiopacity ranged from 0.38±0.04 to 2.57±0.21 mmAl (Table 2) and increased with the amount of filler ($CaWO_4$ r^2=0.996, YbF_3 r^2=0.983, $BaSO_4$ r^2=0.994; $p<0.05$). Table 3 show the water sorption and water solubility of experimental groups compared to the controls. The push-out bond strengths for groups containing 120% of filler were 3.26±1.34 MPa ($CaWO_4$), 4.48±1.66 MPa (YbF_3) and 4.73±1.53 MPa ($BaSO_4$) and there was

Table 1- Means ± standard deviation for the setting time, flow, film thickness and pH of experimental sealers

	Filler concentration					
	20%	**40%**	**60%**	**80%**	**100%**	**120%**
setting time (min)						
$CaWO_4$	451.66±4.71	430±8.17	453±8.17	509±0	501±0	448.66±11.79
YbF_3	612.66±4.71	570±7.07	546.66±4.71	373±0	388±0	378±0
$BaSO_4$	468.66±6.13	478.66±6.13	486.66±6.24	451.66±10.21	384.66±2.36	398.33±10.27
flow (mm)						
$CaWO_4$	20.26±0.51	18.92±2.16	19.28±0.57	18.10±1.28	15.02±1.81	15.85±0.25
YbF_3	20.93±0.17	18.41±0.35	18.50±0.92	15.50±0.38	13.81±0.49	14.59±0.70
$BaSO_4$	22.49±0.37	21.36±0.37	19.43±0.21	17.96±0.21	18.08±1.21	17.69±0.17
film thickness (μm)						
$CaWO_4$	23.33±5.77	30±0	26.67±5.77	26.67±5.77	33.33±5.77	23.33±11.55
YbF_3	16.67±5.77	26.67±5.77	33.33±5.77	30±0	30±0	33.33±5.77
$BaSO_4$	33.33±11.54	23.33±5.77	46.67±5.77	20±0	16.67±5.77	20±0
pH						
$CaWO_4$	6.31±0.13	6.69±0.08	6.66±0.08	6.82±0.08	6.84±0.05	6.99±0.03
YbF_3	6.74±0.08	5.47±0.53	5.69±0.30	6.11±0.22	6.25±0.21	6.31±0.21
$BaSO_4$	6.28±0.09	6.27±0.10	6.37±0.12	6.40±0.10	6.45±0.16	6.51±0.19

Table 2- Means ± standard deviation for the radiopacity (mmAl) of experimental sealers*

	Filler concentration					
	20%	**40%**	**60%**	**80%**	**100%**	**120%**
$CaWO_4$	0.44±0.04	0.91±0.06	1.24±0.15	1.56±0.14	1.98±0.10	2.43±0.05
YbF_3	0.38±0.04	0.76±0.20	1.47±0.37	1.63±0.11	2.07±0.05	2.57±0.21
$BaSO_4$	0.44±0.06	0.63±0.21	1.01±0.10	1.29±0.09	1.62±0.16	2.00±0.07

*Radiopacity increases with the amount of filler ($CaWO_4$ r^2=0.996, YbF_3 r^2=0.983, $BaSO_4$ r^2=0.994; $p<0.05$)

Table 3- Means±standard deviation for the water sorption and solubility of experimental sealers

	20%	**40%**	**60%**	**80%**	**100%**	**120%**
Sorption (μg/mm³)						
$CaWO_4$	145.57±2.81*	182.08±12.15*	167.53±12.09*	236.25±22.46*	183.21±6.35*	200.72±3.67*
YbF_3	69.24±6.54	79.14±5.10*	80.10±1.93*	62.36±4.18	67.75±5.97	70.01±7.06
$BaSO_4$	60.84±7.21	54.52±2.67	51.73±2.41*	52.36±2.48*	51.26±5.87*	45.20±11.26*
Solubility (μg/mm³)						
$CaWO_4$	105.18±10.45*	151.18±25.53*	157.14±10.96*	180.79±31.50*	200.10±11.92*	264.95±11.63*
YbF_3	15.42±1.83*	14.00±3.08*	9.96±0.64	8.16±1.12	10.71±1.61	11.83±1.93
$BaSO_4$	10.44±2.58	10.84±2.17	10.02±1.10	13.16±0.68	13.15±0.92	8.11±8.7

*Statistical difference ($p<0.05$) against control group. Water soprtion and solubility of control group are 64.28±3.79 μg/mm³ and 9.93±2.20 μg/mm³, respectively

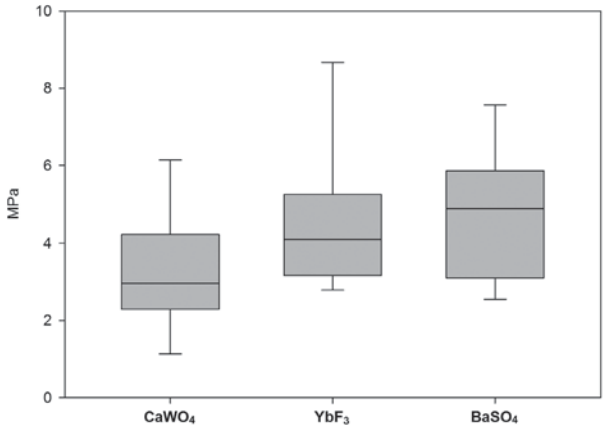

Figure 1- Push-out bond strength (MPa) of the groups with 120% filler by weight. There are no differences between groups (p>0.05)

no significant difference between the experimental groups as shown in Figure 1.

DISCUSSION

Improved physical properties of materials and diagnoses could lead to a higher success rate for root canal treatments. The addition of radiopaque fillers to root canal sealers could interfere with the physical and mechanical properties of the sealers[4,20]. In this study, the concentration of the filler influenced the setting time, flow, film thickness, radiopacity, sorption and solubility of root canal sealers.

The setting time of endodontic sealers should be long enough to allow the filling of the entire root canal system without displacement of the obturation material and the formation of gaps to avoid subsequent bacterial leakage, which could decrease the longevity of the treatment. In this study, the setting time of the sealers ranged from approximately 6 to 10 h. There is no standard for the setting time of endodontic sealers according to ISO 6876. However, the setting time of experimental sealers was suitable to permit root canal filling within an adequate time.

The ability to penetrate accessory canals, dentinal tubules and constitute a thin layer between core materials (e.g. guta-percha points), in order to fill the smallest voids and prevent fluid percolation through the root canal system, is a concern in Endodontics, and this ability is related to flow and film thickness of sealers. All groups present flow nearest to 20 mm and film thickness under 50 µm, which are within the parameters outlined by ISO 6876. The flow and film thickness are directly influenced by the composition of the sealer, temperature and relative humidity. The literature presents different flow and film thickness values for commercial materials[26,28]. The flow of the other two commercial resin-based sealers also had flow and film thickness values lower than those outlined in ISO 6876. In addition, the flow of the sealer cannot be too high due to a possible periapical extrusion, which could compromise apical healing and lead to decreased tooth longevity[24]. The film thickness of experimental sealers was similar to widely used commercial sealers, which have film thicknesses of approximately 50 µm[10].

The addition of radiopaque agents to root canal filling materials should ideally enable their visualisation and assessment on a radiograph without altering their chemical properties. The high atomic numbers of ytterbium (z=70), barium (z=56) and tungsten (z=74) could explain the increase in radiopacity as the amount of filler increases. Elements with high atomic numbers can absorb more X-rays, leading to a radiopaque image[1,21]. Experimental groups containing 120% radiopaque agent by weight were more radiopaque than 2 mm of aluminium, which allowed the sealer to be feasibly identified on a radiograph.

In this study, the sorption and solubility values of experimental sealers were influenced by the addition of radiopaque filler. Calcium tungstate groups had higher sorption and solubility values than other groups. These results may be explained by the calcium tungstate particle size, which has a higher mean diameter than ytterbium trifluoride and barium sulphate. The increased particle size causes the $CaWO_4$ particles to be more soluble in water even though the solubility coefficient of calcium tungstate (2.39 mg/100 ml) is lower than those of ytterbium trifluoride (5.77 mg/100 ml) and barium sulphate (0.24 mg/100 ml). Water sorption and solubility have a significant influence on the mechanical properties and degradation of endodontic sealers. Sealers degrade over time as a result of the sorption/solubility process, which could promote resin/filler lixiviation[9] and consequently cause porosities on obturation mass. The ISO 6876 details the normalisation of root canal sealing materials but does not consider resin-based material. For this reason, in this study we adopted ISO 4049, a standard for polymer-based filling and restorative and luting materials, even though it is not specific for root canal filling materials. According to ISO 4049, the water sorption of resin-based material cannot be higher than 40 µg/mm^3 and the water solubility must be up to 7.5 µg/mm^3. The values obtained from sealers containing different fillers, especially the calcium tungstate sealers, do not meet these standards. However, the ytterbium trifluoride and barium sulphate sealers had sorption and solubility values that met ISO 4049 standards. The matrix properties of root canal sealers are important features that predict the solubility of cements. The literature presents a wide range of solubility data for different compositions of sealers

showing that the epoxy matrix is more resistant to water diffusion than other matrices, such as ionomeric and methacrylate, and water-based cements[7]. Furthermore, the process of periapical repair requires favourable conditions, such as the absence of microorganisms and an adequate pH[8]. Root canal sealers that are in close contact with periapical tissues could interfere with the periapical repair. Lixiviation of sealers as result of solubility could also cause changes in the pH of the periapical environment. An alkaline or neutral pH provides the best conditions for the healing process. All sealers tested in this study had a pH that was close to neutral.

The fluid leakage at root canal sealers interface is a concern to root canal filling longevity. To analyze the dentin-sealer interface, push-out bond strength test showed a good correlation with microleakage elsewhere[22,29]. In this study, the push-out bond strengths were tested for the groups with high radiopacity (120% filler by weight), and no significant differences were observed. The mean values of bond strength agreed with studies that evaluated commercial epoxy-based root canal sealers[17]. Theoretically, resin-based root canal sealers could have micromechanical retention with dentin substrate leading to a more stable interface that prevents degradation over time.

Despite the fact of being an *in vitro* study, the present study clearly identified differences in properties as function of filler type and concentration. The Dental Materials field has the obligation of research development of new materials and provide the experimental explanation of mechanisms involved in phenomena[4,5,18-20,25]. Furthermore, identifying these differences between fillers provides data to clinicians to take more evidence-based decisions regarding the acquisition of root canal sealers.

CONCLUSION

The inorganic fillers evaluated and their concentrations affect the physicochemical properties of an epoxy resin-based root canal sealer. Ytterbium trifluoride and barium sulphate at 120% of concentration showed adequate properties to be used as fillers at epoxy resin-based root canal sealers.

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