An evaluation of the radiopacity of composite restorative materials used in Class I and Class II cavities

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The radiopacity of 28 shades of 18 composite brands, recommended for use in Class I and Class II cavities, and one amalgam were tested in accordance with the instructions in the latest draft standards of ISO for resin-based filling materials. The composition of the inorganic fillers in the materials was analyzed by optical emission spectroscopy. Twelve composites showed radiopacity greater than enamel, for five the radiopacity was lower than that of dentin, and for two materials the radiopacity was between that of enamel and dentin. The optical emission spectroscopy analyses showed a large variety in the composition of the fillers. The elements added to increase radiopacity in the composite materials are barium, strontium, zinc, zirconium, and ytterbium. □ *Dental materials; optical emission spectroscopy; radiography*

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The dental profession has long sought for a restorative material to replace amalgam. The use of composite resin materials in Class I and Class II cavities has increased during the past decade. These composites vary from the earlier radiolucent resins to the more recently developed composites, to which radiopaque elements with high atomic number, such as barium, strontium, and zinc, have been added (1–3). However, the high mole percentages of some of these elements required to give radiopacity may be disadvantageous, as they can result in high solubility (4). Adequate radiopacity in composites, especially those used in posterior teeth, is necessary for differentiation from primary caries and for detection of recurrent caries and marginal defects. According to the latest ISO draft standards for resin-based materials (5), a material claimed to be radiopaque by the manufacturer should have a radiopacity greater than that of the same thickness of aluminium.

The purpose of this study was to measure the radiopacity of 18 composite brands recommended by the manufacturers for use in Class I and Class II cavities and to analyze the composition of the inorganic fillers in the composite materials.

Materials and methods

The materials studied are listed in Table 1. For eight of the composite resin materials darker shades of the resins were tested in addition to the universal color. Five specimens, each 15 mm in diameter and 2.0 mm thick, were prepared of each shade of the different composites investigated. The specimens were prepared in accordance with the instructions in the latest draft standard of ISO for resin-based filling materials (5). Five amalgam specimens, 7 mm in diameter and 2 mm thick, were prepared by condensation of the material in clear acrylic moulds. A newly extracted human permanent lower molar was sectioned mesiodistally with a rotating and water-cooled diamond disc. A 2-mm (±0.1) thick slice was then obtained by wet polishing with silicon-carbide abrasive paper.

Table 1. Dental restorative materials used in the investigation

Material	Color	Code	Batch no.	Manufacturer
Adaptic II	U	A	665111	Johnson and Johnson Ltd., East Windsor, N.J., USA
Brilliant DI	U	B	071287-10	Coltène AG, Altstätten, Switzerland
Cavex Clearfil Posterior	U	CCP	43112	Cavex Holland B.V., Haarlem, The Netherlands
Cavex Clearfil Ray	US UY	CCR	1047A 2023	Cavex Holland B.V.
Condensit	U	C	120886	Austenal Intern. Inc., Sweden
Dispersalloy amalgam		DA	2K881	Johnson and Johnson
Distalite	U	D		Johnson and Johnson
Estilux Posterior	L XR1 XR2 YO	EP	171 1.87 3.87 042 3.87 036	Kulzer and Co. GmbH, Wehrheim, FRG
Ful-fil	LY U	F	0710863 870408	De Trey/Dentsply, Zurich, Switzerland
Heliomolar Radiopaque	U	HR	393601	Vivadent, Schaan, Liechtenstein
Herculite	U Y	H	61354 61094	Kerr, Sybron AG, Basel, Switzerland
Isomolar	U	I	3331/3482	Vivadent
Occlusin	S DY	O	UN60 0387 AO70 1186	ICI, PLC, Pharmaceuticals Div., Cheshire, U.K.
P10	U	P10	6D1/6K1	3M Dental Prod., St. Paul, Minn., USA
P30	U Y	P30	6T2 6J77P	3M Dental Prod.
P50	U Y	P50	72P 73P	3M Dental Prod.
Profile TLC	55 57	P	35609	SS White, Philadelphia, Pa., USA
Visiodispers	S	VD	0079	ESPE, Seefeld, FRG
Visiomolar	S	VM	0010N352	ESPE

Radiopacity

Each specimen was placed near the center of a group-D radiographic film (Kodak Ultraspeed occlusal film) together with an aluminum step-wedge, a 2-mm-thick strip of lead, and a 2-mm-thick slice of a human permanent lower molar (Fig. 1). The step-wedge of 99.5% pure aluminum consisted of five 1-mm steps. The radiographic film was positioned on a 2.5-mm-thick sheet of lead, to minimize back-scatter. Radiographs were taken with a calibrated dental X-ray source (Minray DC, Soredex, Helsinki, Finland) under standardized conditions at 65 kVp and a target–film distance of 40 cm. The films were developed in an automatic processor (Velopex, Phillips, Stockholm, Sweden) operating at 27°C. The radiographs were exposed so that a region of the film near the specimen and aluminum had a photographic

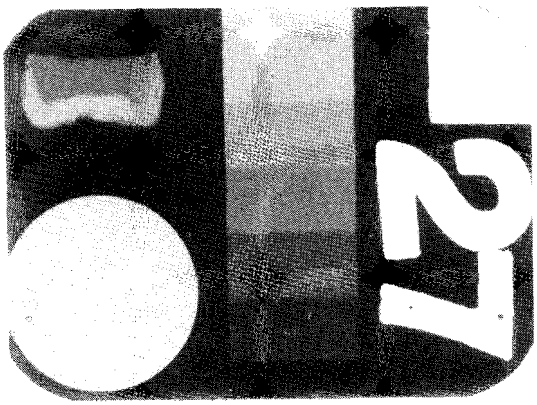

Fig. 1. Radiographic record of 2-mm specimen of composite material compared with a 2-mm-thick section of a lower molar, an aluminum alloy step-wedge (nominal depth of each step, 1 mm) and a 2-mm-thick strip of lead.

density of between 1.5 and 2.0. The optical densities of the images of the specimen, five aluminum steps, enamel, dentin, lead, and background were measured with a transmission densitometer (Macbeth, model TD-502). Calibration curves were made for each film to enable transposition of the measured optical densities to an equivalent thickness of aluminum ($\log(D-D_{base\ and\ fog})$ versus mm Al). $D_{base\ and\ fog}$ is the optical density of unexposed film due to the absorption of light in the film materials and the grains that were exposed by, for example, cosmic radiation before the X-ray exposure. Thus, after correction, the logarithm of the film density is nearly linearly related to the logarithm of the exposure, which in turn is nearly linearly inversely related to the thickness of aluminum in the reference step-wedge. Means and standard errors of the means of the radiopacity values of composite materials, enamel, and dentin in terms of equivalent thickness of aluminum were calculated. The enamel and dentin values were tested with a one-way analysis of variance (6).

Filler characteristics

The inorganic portion of each composite remaining after combustion at 575 ± 5°C for 30 min was analyzed by optical emission spectroscopy (Hilger Quartz Spectrograph E 492, Hilger & Watts, Ltd., London, England). Emission spectroscopy is a semi-quantitative method that can identify and roughly estimate the elemental composition of different filler particles.

Results

Radiopacity

The means and standard errors of the means of the radiopacity values for 2-mm-thick samples of the composite materials, calculated in terms of equivalent thickness of aluminum, are presented in Table 2. The materials are ranked in order of decreasing radiopacity. The values varied between 0.53 (VD) and 6.72 mm Al equivalent (P color 57). The mean values for enamel and dentin are given separately for each of the test groups. Fourteen composites showed radiopacity greater and five less than an equal thickness of aluminum. For 12 composites the radiopacity exceeded that of an equal thickness of enamel, for five the radiopacity was lower than that of dentin, and for two the radiopacity was between that of enamel and dentin. The material EP behaved as two materials, with two colors, YO and L, displaying lower radiopacity than the other two colors, XR1 and XR2. The logarithm of optical density of the step-wedge above base and fog varied linearly with the thickness of aluminum over the 1- to 5-mm range. The standard errors of the means of the five samples for each color of each material were less than 0.1 mm Al in all but three cases and of the order of 1–4% of the mean values in most cases. The radiopacity of enamel did not vary significantly between the different radiopacity measurements, whereas that of dentin varied significantly (Table 2).

The amalgam tested in this study showed an extrapolated value of 23.9 mm Al, which is clearly higher than that for the composites.

Filler characteristics

The results of the optical emission spectroscopy analysis of the inorganic fillers and types of fillers are given in Table 3.

Table 2. Mean values and standard errors of the means of radiopacity values, calculated in terms of equivalent thickness of aluminum, for the amalgam and the composite materials

		Material		Enamel		Dentin	
Material	*n*	$\bar{x}$	(SEM)	$\bar{x}$	(SEM)	$\bar{x}$	(SEM)
Dispersalloy (amalgam)	5	23.9	(0.79)	4.10	(0.04)	1.97	(0.03)
Profile TLC (57)	5	6.72	(0.04)	4.29	(0.06)	2.01	(0.04)
Profile TLC (55)	5	6.53	(0.08)	4.17	(0.09)	2.10	(0.07)
Occlusin (DY)	5	6.40	(0.07)	4.09	(0.07)	2.00	(0.04)
Occlusin (S)	5	6.36	(0.06)	4.10	(0.06)	2.11	(0.04)
Brilliant DI	5	6.09	(0.09)	4.00	(0.06)	2.00	(0.05)
P50 (U)	5	5.82	(0.23)	4.17	(0.05)	2.03	(0.05)
Condensite (U)	5	5.53	(0.26)	4.11	(0.06)	2.10	(0.08)
P50 (Y)	5	5.50	(0.07)	4.21	(0.06)	2.10	(0.02)
Ful-fil (LY)	5	5.20	(0.06)	4.17	(0.05)	2.11	(0.03)
Ful-fil (U)	5	5.08	(0.14)	4.09	(0.03)	2.12	(0.04)
Herculite (U)	5	4.86	(0.07)	4.25	(0.04)	2.07	(0.01)
Herculite (Y)	5	4.86	(0.04)	4.20	(0.09)	2.05	(0.04)
AD II	4	4.82	(0.06)	4.10	(0.03)	1.99	(0.05)
Distalite (U)	5	4.54	(0.08)	4.00	(0.08)	2.03	(0.05)
Heliomolar R (U)	5	4.49	(0.06)	4.19	(0.08)	2.13	(0.03)
P30 (U)	5	4.48	(0.09)	4.18	(0.08)	2.07	(0.06)
Estilux Post (XR1)	5	4.37	(0.06)	4.10	(0.07)	2.05	(0.04)
Estilux Post (XR2)	5	4.26	(0.04)	4.12	(0.03)	2.12	(0.03)
P30 (Y)	5	4.24	(0.07)	4.09	(0.06)	2.07	(0.05)
Clearfil Post (U)	5	2.45	(0.06)	4.15	(0.04)	2.11	(0.03)
Estilux Post (YO)	5	2.40	(0.04)	4.13	(0.05)	2.07	(0.04)
Estilux Post (L)	5	2.19	(0.03)	4.16	(0.08)	2.11	(0.02)
Clearfil Ray (UY)	5	1.06	(0.03)	4.16	(0.08)	2.14	(0.04)
Clearfil Ray (US)	5	1.00	(0.03)	4.18	(0.03)	2.11	(0.03)
P10 (U)	5	1.00	(0.03)	4.22	(0.06)	2.11	(0.03)
Visiomolar	5	0.98	(0.02)	4.10	(0.06)	1.97	(0.04)
Isomolar (U)	5	0.56	(0.04)	4.17	(0.04)	2.11	(0.06)
Visiodispers (S)	5	0.53	(0.02)	4.22	(0.05)	2.11	(0.07)
One-way analysis of variance		Mean		4.14		2.07	
		F		1.21		2.05	
		df		28, 115		28, 115	
		p		>0.05		<0.01	

Discussion

There was good precision of measuring radiopacity in all but three cases. The source of the variation in these three cases is not known but may be due to the lack of homogeneity in the samples. The precision of the measurements of enamel and dentin was also good, but the values of the radiopacity of dentin varied among the cases. As the radiopacity of enamel did not vary among the 29 cases, the variation in dentin radiopacity may be due to the inhomogeneity of dentin.

Different methods have been used to investigate the radiopacity of dental materials (1, 7–9). The data obtained in this study were determined in accordance with the specifications suggested in the latest draft of ISO international standards (5). These draft standards recommend that if the manufacturer claims that a material is radiopaque, the radiopacity must be greater than that of the same thickness of aluminum, when determined in accordance with clause 6:10. According to this criterion, 14 of the composites tested in this study can be considered to be radiopaque. However, this degree of radiopacity is of limited clinical significance, as was shown by Sewerin et al. (7). In Class

Table 3. Composition of inorganic fillers expressed as weight percentage

Material	Major elements	5–20%	0.5–10%	0.1–1%	0.01–0.1%	0.001–0.01%	Type of filler
Adaptic II	Si, Al, Ba		Zr	B, Sr	Mg, Fe	Ti	Ba glass
Cavex Clearfil Posterior Universal	Si		Al		Mg, Fe		Quartz
Cavex Clearfil Posterior Catalyst	Si, Al, Zr		Li, Zn, (La, P)		B, Mg, Ti, Fe, Ta, Co		LiAl silicate
Cavex Clearfil Ray	Si			Al	Ti	Mg, Fe	Quartz
Condensit	Sr	Al, Si	Ca, Na	Mo, Ba, B	Sb, Mg, Ti, Fe	Pb, Cu	Sr glass
Distalite	Si		Yb, Zr	Al	Mg, Fe, Sb	Ti	Amorphous silica
Estilux Posterior L	Si, Al, Zr		Li, Zn	Ba, B, Mg	Ti, Ta, Fe		LiAl silicate
Estilux Posterior XR1	Si, Al, Ba		Zr	B	Mg, Ti, Fe, Sr		Ba glass
Estilux Posterior XR2	Si	Ba, Al	Zr	B	Mg, Ti, Fe		Ba glass
Ful-Fil	Si, Al, Ba		Zr	B, Sr	Mg, Fe, Sb, Sn	Ti	Ba glass
Heliomolar Radiopaque 393601	Si		Yb, Zr	Al	Mg, Ti, Fe, Sb		Amorphous silica, YbF_3
Herculite Y	Si, Al, Ba		Zr	B	Mg, Fe, Ca, Ti, Sb, Sr		Ba glass
Isomolar Base	Si		Zr	Al, Ti	Mg, Fe		Amorphous silica
Isomolar Catalyst	Si		Zr	Al, Ti	Mg, Fe		Amorphous silica
Occlusin S	Si, Al, Ba		Zr	B, Sr	Mg, Ti, Fe		Ba glass
P-10 Catalyst (B)	Si			Al	Mg, Ti, Fe, Sb		Quartz, aluminum
P-10 Universal A	Si			Al	Sb, Mg, Fe		Quartz, aluminum
P-30 Y	Si, Al, Zn		Zr, Ba	B	Mg, Fe, Ca, Sr, Sb	Ti	Zn glass
P-50 Y	Si, Zr		Al	Ti	Mg, Fe, Sb		SiO_2/ZrO_2
Profile TLC 55	Si, Al, Sr		Na	B, Ti, Zr, Ba	Mg, Ca, Fe, Sb		Sr glass
Visiomolar (L)	Si			Al	Mg, Fe		Amorphous silica
Visiodispers (U)	Si, Ca			Al	Mg, Fe, Sb		Amorphous silica

II fillings the X-rays must pass through two layers of tooth structure, buccal and lingual segments, which are contiguous with the cavity preparation, before reaching the film. These two layers of tooth structure partially mask the composite. If the composite is to be made radiopaque to facilitate the detection of enamel decay or marginal defects in Class II cavities, which are surrounded by enamel (10), the material must have a radiopacity not less than that of the enamel. Twelve of the composites meet this standard.

Two studies have attempted to establish the optimum range of radiopacity required for the clinical use of composites in Class II cavities (11, 12). Tveit & Espelid (11) showed that a moderate radiopacity of a dental restorative material was preferable to the radiopacity of amalgam. The frequencies of both under- and over-scoring recurrent caries and simulated marginal defects adjacent to the posterior composite P30 were lower than those for caries and defects near amalgam. P30 had a slightly higher radiopacity value than enamel in the present study. Espelid et al. (12) showed that radiopacity of P30 was better than the higher or lower radiopacity of three experimental composites with regard to the possibility of diagnosing recurrent proximal caries in extracted premolars. According to these studies (11, 12), the optimal radiopacity for a posterior composite should be slightly greater than that of enamel.

The results from the optical emission spectroscopy analyses of the inorganic fillers showed a large variety in composition. The elements with high atomic number in the fillers of the composite materials are underlined in Table 3. The elements added to increase radiopacity in the composite materials are barium, strontium, zirconium, zinc, and ytterbium, of which barium gives the highest radiopacity. These elements are incorporated in the glass powder (3). Watts (13) showed that radiopacity values higher than that of enamel can be achieved in composites with a filler loading of approximately 70% by volume, when the mass percentage of radiopaque oxide in the filler particles exceeds about 20%. Composites containing barium and strontium glass filler mixtures with greater than 35% barium glass or with more than 40% strontium glass fulfill this condition. For 10 of the 12 composite materials for which the radiopacity exceeded that for an equal thickness of enamel, the weight percentage of the radiopaque oxide exceeded 20% (Table 3). The radiopacity of the other two materials (D, HR) derived from the incorporation of less than 10% ytterbium and zirconium. The incorporation of large mole percentages of inorganic ions, to give radiopacity, may be disadvantageous. After storage of cured composites in water or weak acids in laboratory investigations (4, 13), leakage of the filler elements has been shown to occur. Öysaed & Ruyter (4) showed that the highest total leaching of inorganic ions after 110 days' storage in water occurs with zinc, barium, and strontium glasses. It may thus be expected that the radiopacity of composites will decrease with time owing to this leaching. However, Omar et al. (15) showed that there was no significant decrease in the radiopacity of 20 posterior composites after storage in buffered aqueous solutions of acidic, alkaline, and neutral pH for 12 weeks. Further studies are needed to investigate whether the leaching of inorganic ions, especially in the outer part of proximal filling surfaces, may reduce the radiopacity of composites to levels lower than enamel values.

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