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In Vitro Electrical Impedance Spectroscopy of Human Dentine: The Effect of Restorative Materials

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The influence of different restorative materials on in vitro dielectric properties of sound dentine was investigated. The studied samples were three-layer materials consisting of successive disks of dentine and silver amalgam or nanohybrid composite resin. Before being tested, the samples were maintained in physiological solution never more than 48 h from the extraction. Also, sections of intact dentine were similarly prepared for electrical measurements. Complex dielectric permittivity of these specimens was determined in a wide frequency range using the parallel-plate capacitor technique. Very similar dielectric responses of intact dentine and amalgam-dentine material were observed. This is explained on the basis of high dc conductivity exhibited by both samples. In contrast, resin-dentine specimen revealed a much more insulating behavior. A simple theoretical model for heterogeneous systems could be applied to these dental three-layer materials. The dielectric properties of restored dentine are strongly dependent on the kind of restorative material employed in each case. This suggests that electrical data should be used carefully in caries diagnosis on restored teeth. Bioelectromagnetics 29:163–168, 2008. © 2007 Wiley-Liss, Inc.

Key words: dentine; restorative materials; electrical impedance spectroscopy; dielectric properties

INTRODUCTION

Early detection of occlusal caries has become a further complicated task for clinicians [Pitts, 1991; Kidd et al., 1995]. Although traditional routine methods are efficient in the detection of irreversible lesions, they are often inadequate for non-cavitated caries. To improve the efficiency in diagnosing these lesions, new techniques have been developed during the last 15 years which aim to provide quantitative methods for accurate detection and monitoring progression. Electrical resistivity measurements, digital radiography, light scattering, and methods based on fluorescence are some successful examples [Wenzel et al., 1991; Angmar-Månsson and ten Bosch, 1993; Huysmans et al., 1998; Attrill and Ashley, 2001; Bamzahim, 2005].

In particular, electrical measurements have been one of the most investigated techniques due to their easy application and very good reported results [Wenzel et al., 1991; Longbottom and Huysmans, 2004]. The physical fundamentals of the method are based on the phenomenon that resistivity of teeth is significantly reduced by the creation of pores which are filled with conductive fluids. Clinical trials have been carried out using the Electronic Caries Monitor (ECM), a device that measures the bulk resistance of teeth by a specially designed probe tip [Ashley et al., 2000]. The ECM works with a single low frequency, which constitutes an important drawback since the measured data could be strongly affected by polarization effects due to the very low frequency used (21 Hz) [von Hippel, 1954]. Measurements on a wide range of frequencies, the so-called impedance spectroscopy (EIS), give a more

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complete characterization since the electrical response of teeth, as any material, significantly changes at different frequencies. Some in vitro tests have demonstrated the excellent ability of this technique to differentiate sound, non-cavitated carious and cavitated dental surfaces [Huysmans et al., 1996; Longbottom and Huysmans, 2004]. Other studies have evaluated the formation of microleakages in several composite resin restorations using electrochemical techniques to determine the electrical properties as a function of frequency [Pradelle-Plasse et al., 2004]. Moreover, electrical impedance spectroscopy appears as a promissory technique to be applied to in vivo measurements, using adequate contact probes.

When teeth are restored all their physical properties are modified, including the electrical ones. For this reason the behavior observed for sound teeth could be very different from that exhibited by restored specimens. If we consider the problem that means for current dental practice secondary caries (lesions which occur at the margins of an existing restoration) [Fontana and González-Cabezas, 2000] the importance of knowing the electrical response of restored teeth is evident. In addition, the radiographic techniques used for diagnosing secondary caries can lead to confusing results, since the employed restorative materials commonly hide the affected zone [Tveit and Espelid, 1992]. Previous studies have reported in vitro electrical measurements on restored teeth evaluating the marginal integrity of sealant restorations [Verdonschot et al., 1995]; however, these measurements were performed only at a single frequency (using the ECM device). Electrical impedance spectroscopy on restored specimens could provide much more information because it shows the electrical behavior in a wide frequency range, avoiding spurious polarization effects and improving the electrical method as a diagnostic tool.

To obtain applicable results it is essential to suitably characterize the dielectric properties of sound teeth (restored and non-restored) to serve as a reliable comparison with the carious specimens. Moreover, a dielectric characterization of restored dentine may be useful to determine the effect of electromagnetic waves on the stability and performance of restorative materials. In this way, the dielectric characterization could complement conventional studies performed on dental materials, such as biocompatibility and mechanical tests.

In this work some results concerning the dielectric behavior of human sound dentine are reported to provide experimental data that help establish its standard response. A comparison between the electrical properties of dentine with different restorative materials is also carried out.

MATERIALS AND METHODS

Sample Preparation

The material consisted of sound human premolar teeth which were extracted by orthodontic prescription. After extraction, the specimens were immediately placed in physiological solution, and stored at 5–6°C. All samples were prepared by the same researcher (a dentist). After removing soft tissues with ultrasound, these specimens were sectioned perpendicularly to the long axis using a diamond-blade saw. Sections with a thickness of 0.5 mm were obtained from the dentinal region between the occlusal surface and the pulp camera. To assure that the integrity of dentinal tubules was conserved after the cutting procedure, the prepared layers were analyzed by scanning electron microscopy (SEM). Figure 1 shows a typical micrograph of the dentine sections, confirming that the characteristic tubular structure remained intact.

Three-layer composite materials were prepared by interposing a 0.5 mm layer of restorative material between two dentinal sections. Silver amalgam (Tytin®) or nano-hybrid composite resin (Grandio-Voco®) were used. The resulting materials were gently polished to obtain smooth and totally parallel surfaces. Dental sections without restorative material were similarly prepared for dielectric measurements. The resulting samples were labeled as dentine-amalgam (DA), dentine-resin (DR), and intact dentine (ID). A total of five specimens of each class were prepared.

Electrical Impedance Measurements

One of the most common methods for measuring complex dielectric permittivity is submitting the material to an ac voltage between two capacitor plates

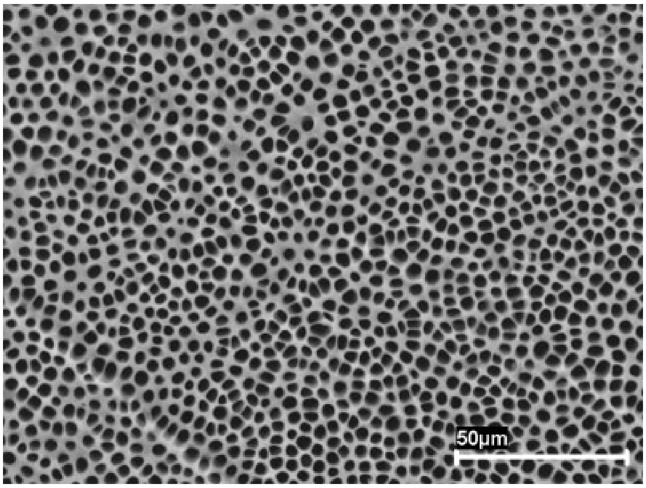

Fig. 1. SEM micrograph showing the tubular structure of the obtained dentine sections.

(electrodes). The electrical resistance (R) and capacitance (C) are determined as a function of frequency. These data are used to calculate both real (ε_r') and imaginary (ε_r'') parts of the complex relative dielectric permittivity, according to the following equations:

$$\varepsilon_r' = \frac{C}{\varepsilon_o} \cdot \frac{d}{A} \quad \varepsilon_r'' = \frac{1}{\varepsilon_o.\omega.R} \cdot \frac{d}{A}, \tag{1}$$

where d and A are the thickness and the area of the sample, respectively, ε_0 is the permittivity of the free space (8.85×10^{-12} F/m), and ω is the angular frequency of the oscillation ($\omega = 2\pi v$ in rad/s, v being the frequency in Hz, using Standard International units). From the equations above, the real part of permittivity is proportional to capacitance, which evidences the strong link between ε_r' and the level of polarization in the material. In contrast, the imaginary part is inversely proportional to resistance, being a direct indication of the conductivity of the sample. This comprises the electrical conduction by free-carriers (ions, electrons) as well as temporal charge variation related to purely dielectric relaxation processes.

Complex dielectric permittivity of the samples was measured at frequencies from 20 to 1×10^6 Hz using a home-made device connected to an Agilent 4284A LCR-meter. Basically, the sample-holder is a two parallel-plate capacitor coupled to a temperature controller, which allowed fixing a temperature of 37 °C (Fig. 2). Before placing the sample between the electrodes, both material surfaces were covered with silver paint to assure a good electrical contact with the electrodes. All measurements were made immediately after taking out the specimens from physiological solution and externally drying with tissue paper.

The sample-holder was pre-heated before measurement to avoid delays that could cause significant variations of water content in the tested materials. All the samples' dimensions were taken with a caliper with an accuracy of 0.02 mm. An elliptical geometry was assumed for the areas calculation.

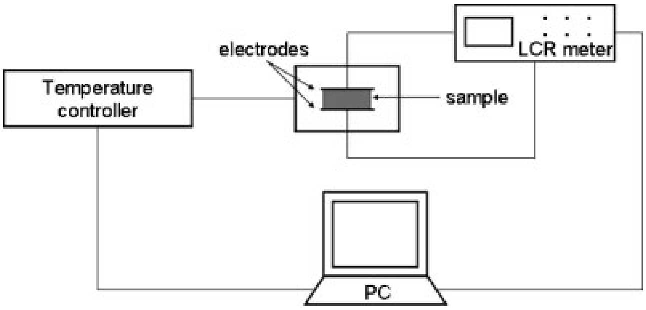

Fig. 2. Scheme of the experimental setup used for electrical measurements.

A maximum of six permittivity measurements per specimen was performed, giving dispersion values as low as 1.5 and 5% for ε_r' and ε_r'', respectively, which are very close to the limit of accuracy of our impedance analyzer. On the other hand, the dispersion of the permittivity observed for different specimens of each studied material (DA, DR, and ID) resulted in higher values. For frequencies above 5 kHz the mean dispersion was 7% for ε_r' and 10% for ε_r''. For frequencies lower than 5 kHz both values increased up to 10% and 14%, respectively, probably due to electrode polarization effects. The curves in all figures show the average response of the whole set of specimens.

RESULTS

Figure 3 shows the real part of the relative permittivity (dielectric constant, ε_r') of the samples as a function of frequency. For sample DR, an almost flat dielectric response (with values of ε_r' between 20 and 50) can be observed, indicating a poorly conductive behavior. This is consistent with the insulating characteristics of the used resin, which is composed of a polymeric matrix filled with SiO_2 particles (both well-known insulating materials). In contrast, the curves obtained for samples ID and DA show much higher ε_r' values together with a decrease of several orders of magnitude in the whole measured frequency range. At about 1×10^4 Hz both curves split up, showing a larger relaxation for sample ID.

Figure 4 displays the imaginary part of the relative permittivity (ε_r'') as a function of frequency for the three

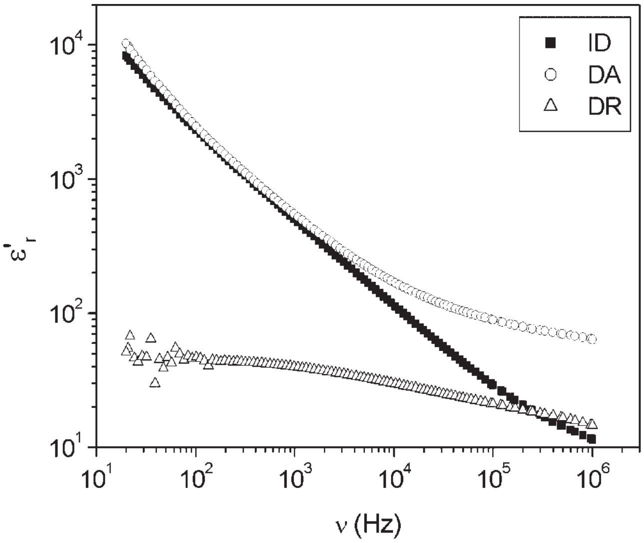

Fig. 3. Real part of the relative dielectric permittivity, ε_r', as a function of frequency for intact dentine (ID), dentine-amalgam (DA), and dentine-resin (DR).

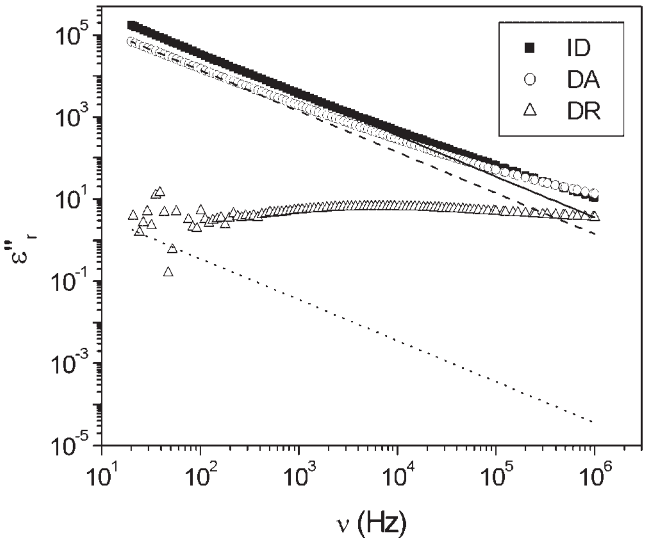

Fig. 4. Imaginary part of the relative dielectric permittivity, ε''_r, as a function of frequency for samples ID, DA, and DR. Straight lines represent the contribution from dc conduction.

studied samples. For ID and DA a very similar behavior can be observed. Their ε''_r values are much higher than those registered for sample DR, especially at low frequencies. The straight lines in Figure 4 represent the contribution to ε''_r of free-carriers conduction, which is due to ions (mainly in ID sample) and ions and electrons (in sample DA). This was calculated using the following expression: $\varepsilon''_{r,cond} = \sigma_{dc}/\varepsilon_0 2\pi\nu$, where σ_{dc} is the dc electrical conductivity. This variable was estimated from resistance data measured at the lowest frequency (20 Hz). The contribution of free carriers is prevalent for sample ID and for sample DA at frequencies lower than 1×10^4 Hz. Conversely, the dc conduction contribution for sample DR is negligible, suggesting that the observed losses are exclusively due to dielectric relaxation effects.

DISCUSSION

The permittivity values measured for the intact dentine perfectly agree with those reported for fluid saturated bones studied in a similar frequency range [Kosterich et al., 1983]. The dependence of ε'_r with frequency (a continuous decrease from 10^4 to 10, between 20 Hz and 1 MHz) coincides with that observed for freshly excised samples of bone, which indicates that the methodology employed to conserve the human tissue was really appropriate.

The results shown above indicate that the dielectric behavior of intact dentine is more similar to dentine-amalgam, rather than the behavior exhibited by dentine-resin material. It is important to remark here that all the samples were measured after being immersed in physiological solution. Such experimental condition is necessary to simulate the in vivo state of the dental composite material, taking into account that 30% of dentine is composed of liquid [Bascones, 1998]. In this way the aqueous solution inside the dentinal tubules strongly affects the dielectric response of the dentine in all the samples.

The sample composed only of dentine retains more solution volume because of its higher porosity. Samples containing restorative material (amalgam or resin) are denser and consequently they absorb less solution volume. In fact, absorption of saline solution was measured for all the specimens. Samples were weighted before and after 10 days of being dried at room temperature. The results indicate that specimen ID absorbs 100% more solution than DA and 70% more than DR. The aqueous solution dramatically enhances both values of ε'_r and ε''_τ, since a high water content (whose $\varepsilon'_r \sim 78$ in the investigated frequency range) produces an increase of dipolar polarization as well as a faster mobility of the ions in the sample. The relatively high values of ε'_r and ε''_r registered for sample DA can be explained by the metallic nature of the amalgam, which presents high dc conduction (mainly electronic), as shown in Figure 4.

The observed behavior for the studied samples can be understood by considering the restored specimens as series combinations of both capacitive and resistive elements. Sample DR consists of a combination of a highly resistive component (hybrid resin) and a highly conductive one (dentine). By contrast in sample DA both components have significant electric conductivities (metallic amalgam and dentine). This is the main reason for the similarity observed in the response of ID and DA, since both samples have similar electric conductivities given by ionic or electronic carriers.

The studied three-layer materials can be interpreted in terms of the Maxwell–Wagner model for interfacial polarization [von Hippel, 1954; Albella Martín and Martínez Duart, 1984]. In this model, a capacitor formed by two dielectric materials, with conductivities σ_1, σ_2 and dielectric permittivities ε_1, ε_2 is considered. Such a system gives a relaxation spectrum (ε'_r vs. ν) indistinguishable from that expected for a single-phase dielectric which obeys the well-known Debye model. However, the relaxation process occurs at a characteristic frequency that depends on the properties of both individual components. In the ε''_r versus ν curve, significant differences with Debye model are found. The imaginary part contains a conductivity term owing to both phases, which noticeably modifies the curve shape, especially at low frequencies. Figure 5 summarizes several dielectric responses theoretically

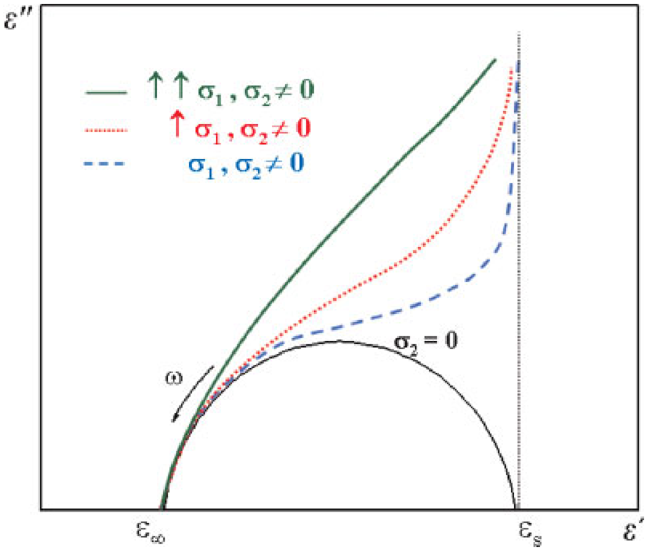

Fig. 5. Cole–Cole plot showing theoretical dielectric responses for a two-phase system according to Maxwell–Wagner model. [The color figure for this article is available online at www.interscience.wiley.com.]

predicted by the Maxwell–Wagner model. These are graphically shown in a Cole–Cole plot (ε''_r vs. ε'_r) to better visualize the influence of conductivity on the dielectric behavior of a two-phase capacitor. When a system is composed of a conductive phase ($\sigma_1 \neq 0$) embedded in an insulating phase ($\sigma_2 = 0$), the observed response agrees with the ideal Debye model, that is, Cole–Cole plot results in a semicircular arc, whose diameter is the difference between real permittivities at $v = 0$ (ε'_r) and $v \to \infty$ (ε'_∞). As σ_2 grows, the ideal semicircular curve's shape disappears, showing a steep rise in ε''_r.

Figure 6 shows Cole–Cole plots obtained for samples ID, DA, and DR. Clearly distinct responses are observed for the studied samples. A behavior completely different to that predicted by the Debye model can be seen for samples DA and ID. This is a consequence of the high conductivity produced by the free carriers existing in theses specimens. On the contrary, the curve obtained for dentine-resin material resembles the semicircular arc, approaching the behavior predicted by the Maxwell–Wagner model for two phases with $\sigma_2 \approx 0$ (Fig. 5). In this case, phase 1 is the intact dentine (with a relatively high conductivity due to the presence of saline solution in the tubules) and phase 2 is the composite resin, whose conductivity is negligible. Actually, a pear-like shaped curve rather than a semicircle is obtained. This is due to the effect of the finite conductivity value of the used resin [Jonscher, 1983], which has absorbed some conductive solution according to the results obtained from the experiments. For sample DA both phases (dentin and amalgam) have relatively high conductivities and consequently the behavior is entirely different to that exhibited by dentine-resin sample and much more similar to that revealed by the intact dentine.

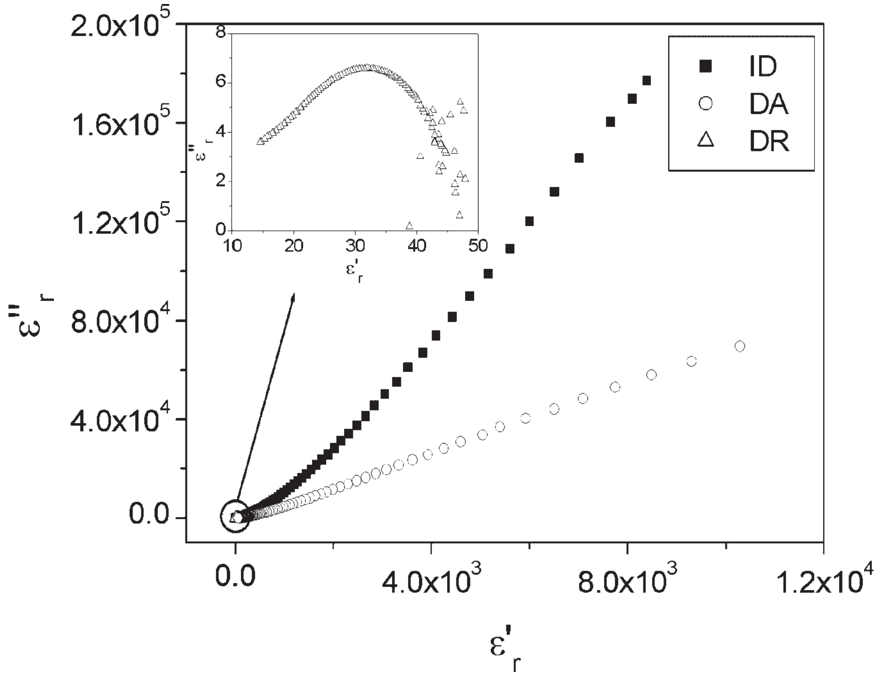

Fig. 6. Cole–Cole plot for samples ID, DA, and DR. The inset shows an enlargement of the curve corresponding to sample DR.

CONCLUSIONS

The in vitro dielectric behavior of non-restored and restored (with a silver amalgam and a nanohybrid resin) human dentine has been determined by a simple experimental set up. This method allows obtaining the real and imaginary part of the dielectric complex permittivity, which are parameters intrinsic to the nature of studied material and independent of the dimensions of the measured specimen.

From a dielectric point of view, the properties of non-restored dentine and amalgam-dentine are very similar: both exhibit relatively high conductivities and dielectric constants. In contrast, specimens restored with resin show a much more insulating response. Despite the complexity of the investigated systems, these can be qualitatively described by the two-layer Maxwell–Wagner model.

Although the experimental design and model used in this work are different from those in clinical practice, the results obtained prove that the dielectric properties of dentine could critically depend on the type of material used for its restoration. In this way, electrical experimental data already reported on non-restored teeth can not be simply extrapolated to those restored. Further experiments using these and other restorative materials should be performed for a comprehensive electrical evaluation of restored teeth.

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REFERENCES

Albella Martín JM, Martínez Duart JM. 1984. Física de dieléctricos. Barcelona: Marcombo.

Angmar-Månsson B, ten Bosch JJ. 1993. Advances in methods for diagnosing coronal caries—A review. Adv Dent Res 7:70–79.

Ashley PF, Ellwood RP, Worthington HV, Davies RM. 2000. Predicting occlusal caries using Electronic Caries Monitor. Caries Res 34:201–203.

Attrill DC, Ashley PF. 2001. Occlusal caries detection in primary teeth: A comparison of DIAGNOdent with conventional methods. Br Dent J 190:440–443.

Bamzahim M. 2005. Evaluation of the DIAGNOdent method for detection and quantification of cariuos lesions—In vitro and in vivo studies. PhD thesis, Institute of Odontology, Karolinska Institutet, Stockholm.

Bascones A. 1998. Tratado de Odontología: Tomo I. Madrid: Smithkleine Beeckam.

Fontana M, González-Cabezas C. 2000. Secondary caries and restoration replacement: An unresolved problem. Compend Contin Educ Dent 21:15–26.

Huysmans MCDNJM, Longbottom C, Pitts CNB, Bruce PG. 1996. Impedance spectroscopy of teeth with and without approximal caries lesion—An in vitro study. J Dent Res 75:1871–1878.

Huysmans MCDNJM, Longbottom C, Pitts CNB. 1998. Electrical methods in occlusal caries diagnosis: An in vitro comparison with visual inspection and bitewing radiography. Caries Res 32:324–329.

Jonscher AK. 1983. Dielectric relaxation in solids. London: Chelsea Dielectrics Press.

Kidd EA, Ricketts DN, Pitts NB. 1995. Occlusal caries diagnosis: A changing challenge for clinicians and epidemiologists. J Dent Res 74:1206–1211.

Kosterich JD, Foster KR, Pollack SR. 1983. Dielectric permittivity and electrical conductivity of fluid saturated bone. IEEE Trans Biomed Eng 30:81–86.

Longbottom C, Huysmans MCDNJM. 2004. Electrical measurements for use in caries clinical trials. J Dent Res 83:C76–C79.

Pitts NB. 1991. The diagnosis for dental caries: Diagnostic methods for assessing buccal, lingual and occlusal surfaces. Dent Update 19:393–396.

Pradelle-Plasse N, Wenger F, Picard B, Colon P. 2004. Evaluation of microleakeage of composite resin restorations by an electrochemical technique: The impedance methodology. Dent Mater 20:425–434.

Tveit AB, Espelid I. 1992. Class II amalgams: Interobserver variations in replacement decisions and diagnosis of caries and crevices. Int Dent J 42:12–18.

Verdonschot EH, Rondel P, Huysmans MCDNJM. 1995. Validity of electrical conductance measurements in evaluating the marginal integrity of sealant restorations. Caries Res 29:100–106.

von Hippel A. 1954. Dielectrics and waves. London: Artech House.

Wenzel A, Larsen MJ, Fejerskov O. 1991. Detection of occlusal caries without cavitation by visual inspection, film radiographs, xeroradiographs, and digitized radiographs. Caries Res 25:365–371.