

## EIS AND SURFACE INVESTIGATION IN COMPARING DENTAL COMPOSITE RESIN AND DENTAL IONOMER CEMENT

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*Se prezintă o comparație între două materiale dentare larg utilizate în activitatea clinică curentă: compozitul dentar cu microumplutură și cimentul ionomer. Configurațiile suprafețelor fiecărui dintre aceste tipuri de materiale definesc multe dintre proprietățile acestora. După obținerea unor mostre similare, acestea au fost caracterizate din punct de vedere al rugozității de suprafață și integrate cu rezultatele clinice. Datele EIS fac legătura cu informațiile legate de suprafața materialului și pot fi corelate cu diferite simptomatologii clinice.*

*A comparison between two dental materials currently in full clinical use is discussed. The surface configurations of each of the two types of materials define many of their properties. After obtaining the test samples with quantifiable sizes, the surface roughness of each material was observed, and the data were compared to clinical results and experience. The EIS testing of the mentioned materials goes further to show structure characteristics that can be linked to different clinical symptomatology.*

**Keywords:** Cement, Dental resin composite, AFM testing, EIS testing, roughness, Fluoride release, surface aspect

### 1. Introduction

The use of composite resins and ionomer cements has a long tradition in dentistry. These materials have brought great improvements in the way dental affections were and are being treated, consequently in the way each of us looks and functions today. In the beginning the Ionomer cement following in the footsteps of the silicate cement meant a revolution in the field of dental esthetics[1]. Shortly after, the dental resin composite materials brought a new high to dental esthetics with one of the first self-curing resins named Evicrol. Many clinical in vivo and ex-vivo studies have been performed on these materials in order to better understand their clinical behavior and even better, to formulate prognoses on the materials lifespan [2]. For example the use of composite resins

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for dental fillings does in time succumb to the appearance of marginal infiltration[2]. The ionomer cement presents usually a lower rate of marginal infiltration but a higher discoloration issue [3]. These materials are exposed in the oral cavity to all kinds of chromatophoric substances contained in food and beverages. The discoloration effect of coffee, tea and other similar substances has long been demonstrated.

The ionomer cement has entered its clinical use in 1972, starting out as one of the best solution for anterior restorations and offering the only alternative to silver amalgam. Its use had been extended to subgingival restorations in the belief that it can better harden in moist conditions, fact that has been proven false later on. The main advantage of this material is the high fluoride ion release over time[4]. Fluoride is a very efficient antibacterial substance and also helps remineralize the dental tissues. The widely spread commercial form of the material is a bicomponent one containing a powder and a liquid that mixed together form a creamy substance. Another commercial type is that represented by capsules containing powder and liquid separated by a polymeric membrane. When the capsule is activated the membrane perforates and the two components mix with the help of an amalgamator device at 4500 vibrations/min. There is also another option represented by two paste-like components.

The dental composite resin has started its clinical use around the 1990's bringing a whole new promise to dental practitioners everywhere. The material was much harder than ionomer cement and it was more esthetic. It was comprised of a BisGMA matrix in which different sized particles were placed (nano to macro). It was easier to use in terms of in practice handling and it could offer a better final result. The dental composite resin was a totally different material than the ionomer cement regarding bonding to the dental tissue. While the ionomer cement contained a polyalkenoic acid that demineralized the hard dental tissue allowing for a  $5\mu\text{m}$  penetration and binding interface, the composite dental resin required a new three-step approach. The first step was applying an acid in order to etch the dentinal surface thus cleaning the smear layer and effecting the permeabilization of the dentinal tubuli in dentine and creating micro retention in enamel. After etching the primer was applied in order to decrease the surface tension of the bonding agent applied in the third step. Classically after each step excluding the acid etch a light curing device was used to chemically activate the used substances. The commercial forms were chronologically auto curing liquid-paste or paste paste compounds to modern day light curing dental composite resins. Although all companies presently use BisGMA matrix a Japanese GC (General Chemistry) company introduced in 2011 a new kind of UDMA matrix (Urethane dimethacrylate).

The current article wishes to compare two different kinds of materials regarding the surface roughness measured by AFM and inner structure behavior using Electrical Impedance Spectroscopy.

## 2. Materials and method

Samples for each material have been obtained using commercially bought substances placed in plastic conformers to obtain identical testing forms.

In the case of the ionomer cement its setting mechanism is special in the respect that the setting reaction, initiated by the mixing of the powder with the liquid, consists of three phases that overlap each other.

Phase 1: When the powder and liquid are mixed, hydrated protons (hydrogen ions) are formed from the ionization of the polyacrylic acid in water. These ions attack the peripheries of the glass particles causing the release of calcium, aluminum, and fluoride ions and the formation of a silica-based hydrogel around the involved glass particles.

Phase 2: In the second phase of the reaction, the  $\text{Ca}^{+2}$  and  $\text{Al}^{+3}$  ions migrate from the silica hydrogel into the aqueous cement phase where, as the pH increases, they precipitate out as polysalts (specifically as polycarboxylates). The polycarboxylates ionically crosslink the polyanion chain and cause the cement to harden. Calcium polycarboxylates form first for several reasons: 1. they are released in greater quantity by the action of the hydrogen ions because attack on the glass particles occurs preferentially at the calcium-rich sites; 2. the calcium ions have a bivalent, rather than trivalent, charge which allows them to migrate faster into the aqueous cement phase; and 3. the calcium cations do not form stable complexes with the fluoride ions as do the aluminum cations. This means that the calcium is more readily available to crosslink the polyanion chains. The calcium polycarboxylates form over the first 5 minutes while the stronger and more stable aluminum polycarboxylates form over 24 hours. As a result, the cement has relatively poor physical properties at first. These properties improve, however, as the aluminum polycarboxylates form. The fluoride ions initially released from the glass particles along with the calcium and aluminum ions do not take part in the matrix-forming stage, but remain available in the matrix.

Phase 3: A slow hydration of both the silica-based hydrogel and the polycarboxylates occurs which results in a further improvement in the cement's physical properties. This phase of the reaction may continue for several months. Two clinically important results of this reaction are that the physical properties of the glass-ionomer cements take a relatively long time to fully develop because of the cement's long setting reaction and that the cement is sensitive to moisture

contamination and to desiccation because the glass particles are covered with a hydrogel.

The surface characterization was obtained using A.P.E Research A-100 AFM microscope. Electrical impedance spectroscopy was used to characterize the diffusion of electric charge through the test materials.

Gradia Direct dental composite resin and GC Fuji IX ionomer cement were the materials used for testing. The materials mechanical strength properties can be found in Table 1 for Gradia composite and Table 2 for Fuji IX ionomer cement.

Table 1

**Statistical data regarding Gradia GC composite strength[13]**

Product	GC Gradia
Flexural strenght (MPa)	124
Flexural modulus (Mpa)	6.92
Flexural energy (MPa)	1.92
Occlusal wear(microns) after 200000 cycles with 1.70 MPa load	8.7

Table 2

**Statistical data regarding Fuji IX ionomer strength**

Procedure	Condition	Data Values
Mixing Time(seconds)	23 degrees Celsius	10 sec
Working Time(min:sec)	23 degrees Celsius	1min.15sec
Final Finishing Commencing Time (min:sec)	37 degrees Celsius after starting mix	3min
Surface Hardness (Hv)	After 1 day	74(3)
Compressive strenght (MPa)	After 1 day After 1 week	268(10) 274(11)
Modulus of elasticity (GPa)	After 1 day	8.6(0.3)
Diametral tensile strenght (MPa)	After 1 day	23(2)
Flexural strenght (MPa)	After 1 day	26(3)
Solubility(%)	Distilled water 1mmol/L Lactic Acid	0.02 0.21
Radiopacity(mm)		3.7(0.3)
Bonding strenght (MPa) with Cavity Conditioner	To Bovine enamel after 1 day To Bovine dentin after 1 day	6.9(1.6) 5.8(2.2)

For the AFM testing both materials were pressed between two very smooth glass plates in order to obtain a straight surface. The ionomer cement was contained in a modern capsule with exact reproducible amounts of material and it was mixed using the SDI Ultramat 2 amalgamator.

For the EIS testing a plastic conformer was used for material deposition thus creating identical material shapes after the hardening process. Before the final hardening was over, a platinum wire was introduced into both samples in the exact center of the shape. Testing was carried out using Autolab PGSTAT by

Metrohm using AC current 10mV and varying the frequency between 1-100kHz. The working electrodes (samples) were introduced into an electrochemical cell with ultrapure distilled water. This solvent was preferred because we wanted to observe how the two materials perform in a very low conductive medium compared to the oral cavity. We believe that saliva having a high electrical conductivity is not precise enough to show small variations. Also the used materials are only partially in contact with oral saliva and a study using a model was required. Reference electrode was Ag/AgCl and the counter electrode was a Platinum (Pt) electrode. The testing results were then digitally fitted and interpreted using the Nova 1.7.8 software.

### **3. Results and Discussion**

#### **Ionomer Cement**

Viewing the ionomer cement via AFM it presented a rough surface with a medium roughness of 41.9nm [Fig.2]. This value of roughness accounts for the clinical behavior and the high discoloration rate determined by bacterial colonization [4]. This character explains the good retention offered to composite resin in sandwich techniques based on the surface being determined by heterogeneous glass particles that fuse together [Fig. 2]. Also the surface hints to a more aerated structure of the whole material thus the expected higher diffusion rate through such a material. The actual roughness of the ionomer is influenced greatly by the intra oral pH value [5][6]. If the pH decreases it also affects the surface of the ionomer causing surface dissolution and increased roughness. The important advantage of the material is that with the wear of one of its surface layers a new high concentration fluoride releasing area is exposed from underneath. Also it is very important to add that in basic pH and in high fluoride environment the Ionomer actually attracts fluoride ions to its surface [7][8]. The EIS investigation performed on the sample reveals the character described via AFM [Fig. 2]. The fitting circuit used for the corroboration of data presented three phases [Fig. 1].

The third phase corresponds to interaction between the platinum electrode and subsequent material, the second phase corresponds to the bulk materials

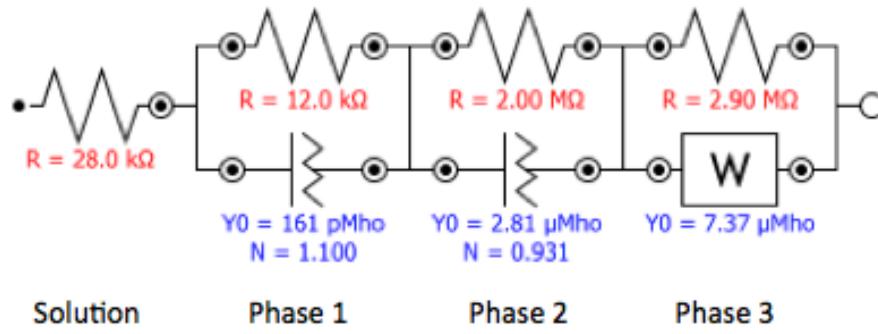


Fig.1 Equivalent EIS circuit used for Ionomer Cement data fitting

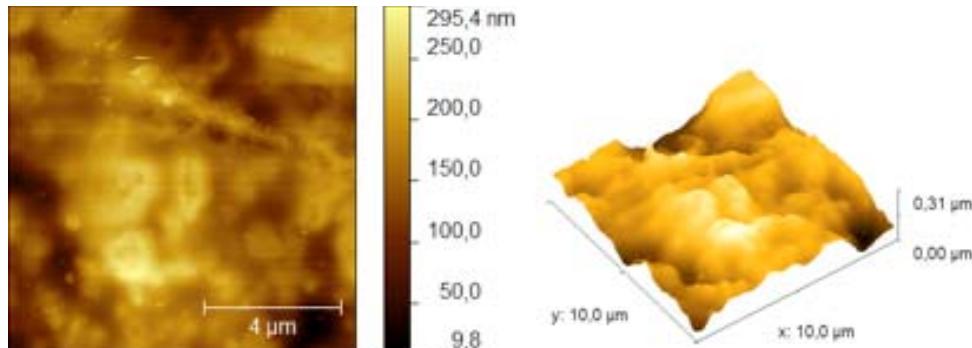


Fig. 2 AFM image of ionomer cement sample

properties and the first phase corresponds to surface phenomena and interaction between the solution and surface of sample. The high value of electrical resistance of the second phase of the equivalent circuit shows the non-conduciveness of the sample. The ionomer cement presents an electrical diffusion effect showing a less dense, less compacted structure than the dental composite resin [Fig.4]. The ionomer cement also presents a fluoride release mechanism that insures a theoretical high fluoride concentration in the dental tissue over the next 12 months [9].

The effect of fluoride on the dental surface is extremely beneficial, it decreases bacterial growth and can help in the process of re-mineralization[10]. The EIS data coupled with clinical observation show that the ionomer cement has a more life-like, tooth like behavior compared to the composite resin. The

advantage of a material that is similar to the tooth structure used to “repair” the dental cavities is huge taking into account the tooth-filing processes involved [6][11].

### Dental Composite Resin

The micro particle dental resin used for testing has presented a much lower value of roughness around 20.7nm[Fig.3]. Thus the surface is more homogenous with moderated peaks and high values of only 844.3nm. Clinically the dental resin is visibly more even, less rugged to direct touch with the dental probe. The lower roughness also reduces the colonization effect of bacteria on such a surface helping self-cleaning of oral elements. The lower roughness offers a better polishing surface for a more natural life-like finish of the dental filling making it as similar as possible to the natural tooth [7]. The surface character also hints to a denser structure and confirms the findings of the EIS sample investigation [Fig.5].

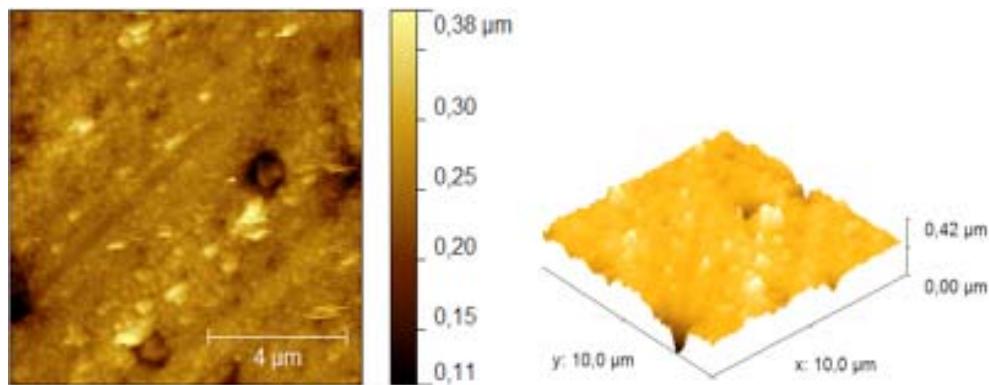


Fig.3 AFM image of Dental Composite Resin

The Dental Composite resin presents a very compact structure almost “impermeable” to electrical diffusion. It presents a very high electrical resistance showing only scattered data points. The high degree of micro particle compaction and the uniform disposition of the said particles in the matrix are partly responsible for the great isolation capabilities of the composite resin. These characteristics are clinically evident in the case of sensible teeth, reducing pain. The silica micro particles offer very good isolation from the oral cavity and different stimuli that act on the tooth decreasing sensibility and pain, reducing the amount of nerve inflammation and decreasing post-op pain. The EIS diagram presented clearly shows that even at a very high AC frequency there is almost no electron diffusion through the material showing a non-conductive substance [Fig.5].

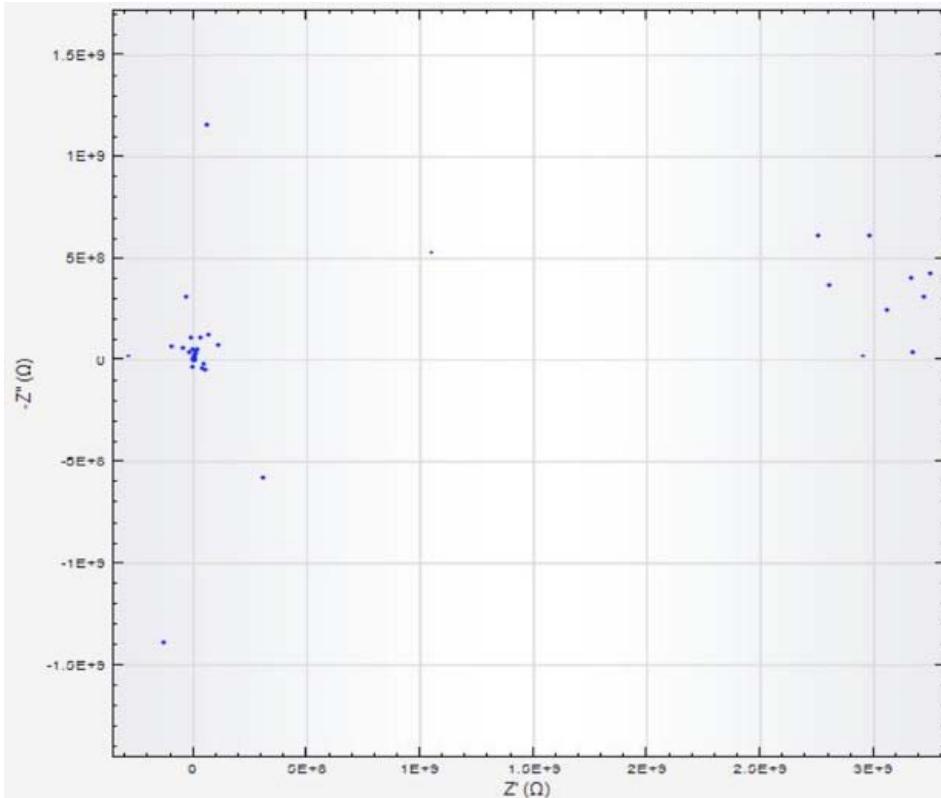


Fig.5 EIS Nyquist diagram presenting Dental Composite data

Both tested materials presented different qualities that clearly define each class and indicate the areas of use. The Ionomer Cement distinguishes itself by great surface roughness, fact that is quite disadvantageous in the oral cavity, because it offers a perfect surface for bacterial colonization. This is countered by long-term fluoride release [9] beneficial for tooth. Seldom are caries found under an ionomer filling. Although the surface exposed to the oral cavity releases fluoride ions inhibiting bacterial colonization [10], the rough surface has a higher degree of wear than the natural tooth, requiring periodical replacement or coating with a dental composite resin. Diffusion through the ionomer cement coupled with the liquid up-take show that the cement acts life-like and similar to the tooth structure. Based on the surface characteristics described above, it can be ascertained that some chemical elements and molecules could diffuse through the cement structure to the tooth underneath having a beneficial effect.

Another great advantage of the ionomer cement is its capability to recharge the fluoride ion concentration in certain advantageous environments. In

high pH environments or at high NaF concentration the surface of the ionomer greatly increases its Fluoride contents [6][7].

The dental composite resin - a polymeric substance - greatly differs from the structure of the ionomer cement. It has a much denser, much more compacted structure. It is mostly neutral toward the dental structure. Clinically it leaks a very small quantity of primer after hardening, having a minimal invasive effect on the vital pulp organ [12]. The EIS data reveal a dense structure that protects the dental organ from different external stimuli but presents no salivary interaction or molecular exchange.

## 6. Conclusions

This study links the AFM data obtained in both ionomer cement and dental composite resin test samples to the EIS data, correlating intrinsic properties to clinical proofs. This type of integration, between clinical correlations, electrochemical technique and surface microscopy, could lead to a better understanding of dental materials and their indications. Thus, it could improve the outcome of classical dental treatment and offer a new insight regarding structure modification of oral materials with time and wear.

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