Scanning electron microscopy evaluation of composite inlays luted with different adhesive systems

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The composite biomaterials used for manufacturing indirect inlay restorations are advanced materials that offer a superior alternative in operative dentistry. The compatibility of the composite inlay with the resin cement and adhesive system is important in achieving a good seal at the tooth-restoration interface, thus ensuring its durability in time. This *in vitro* study aimed to evaluate the ability of some experimental adhesive systems to create a good bond between the dental structures and Barodent inlays together with the luting resin cement used for inlay cementation. Forty composite inlays were manufactured using the Romanian composite for indirect restorations, Barodent (ICCRR). For luting the restorations, two experimental adhesive systems were used in combination with a dual cured resin cement. Two well recognized adhesive systems were used as control, in order to compare the results for the experimental products. The tooth -luting cement-inlay interfaces were investigated using scanning electron microscopy analysis. The results showed some similarities between the interfaces obtained with the experimental adhesives and those obtained with the trade-mark adhesives.

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1. Introduction

Composite resins and adhesive systems have been available in dentistry for decades. They continued to develop regarding their mechanical, chemical and optical properties thus becoming widely used in anterior and posterior restorations because of the growing aesthetic demands of the patients. However these advanced materials still have some undesirable properties like the polymerisation shrinkage which is one of their most important limitations. It affects the quality of marginal adaptation by producing defects in the composite- tooth bond, leading to failure associated with microleakage, postoperative sensitivity and recurrent caries. Indirect inlay restorations minimise the polymerisation shrinkage, except for the amount associated with the layer of composite resin luting cement. The presence of leakage at the margins of inlay restorations is correlated with the type of dental substrate, the etching procedures and the type of the used adhesive system [1-7].

In spite all the improvements in the composite materials' properties and adhesive systems' formula, the bond strength and the marginal adaptation of composite restorations to dentin is less resistant than bonding to enamel. More frequently the proximal caries in posterior teeth go subgingivally, so that the limits of the preparations will be situated in dentin or cement that may cause problems in assuring an optimal marginal seal [8-12]. Thus the longevity of composite inlays depends on many factors i.e. quality of the marginal adaptation, presence of a continuous interface between the restoring material and the dental tissues, tightness of the tooth - restoration interface that should be able to prevent microleakage and restoration breakdown [13-15]. Marginal adaptation of bonded restorations is influenced by the inherent properties of the materials, the polymerisation contraction and the stress caused by this contraction on the dental tissues. Also the marginal seal is influenced by factors like the operative technique in use i.e. size and form of the cavity, sectioning angle of the enamel prisms and dentinal tubules, fabrication protocol of the restorations, polymerisation protocol, method used for the dental substrate conditioning or properties of the luting cement [15-18]. The choice of one adequate restoring material with good compatibility with the adhesive system in order to obtain an optimal marginal adaptation to the dental tissues is very difficult.

Our previous studies showed that the Romanian composite Barodent [19, 20] could be successfully used as restorative material for manufacturing inlays. The purpose of this *in vitro* study was to evaluate the sealing ability of two experimental adhesive systems in combination with a dual-cured resin cement for luting Barodent composite inlays. Two well recognized adhesive systems were used as control, in order to compare the results for the experimental products. The quality of the tooth-luting cement-inlay interface was investigated using scanning electron microscopy analysis.

2. Experimental

Forty extracted, sound permanent third molars were selected for this study and they were kept in distilled water at 4°C for maximum 3 month before use. Standard class II (proximo-occlusal) cavities for inlays were prepared according to the clinical protocol. Composite inlays were prepared by the indirect procedure using Romanian composite, *Barodent* (Raluca Ripan Institute for Chemical Research) as described in our previous research [20].

Sample teeth were randomly divided into 4 groups of 10 teeth each. In the first two groups, two Romanian experimental adhesive systems, manufactured at the Raluca Ripan Institute for Chemical Research (ICCRR) were used for the inlay luting procedure (Table 1). The two-step self-etch adhesive *AD-3A* and the one-step self-etch adhesive *AD-3B* were used in Group 1 (G1) and Group 2 (G2), respectively. In the other sample groups, two similar trade-mark self-etch adhesives manufactured by Kerr Corporation were used as control. *OptiBond® Solo Plus™ Self-Etch Adhesive System* and *OptiBond® All-In-One* were used in Group 3 (G3) and respectively Group 4 (G4). For the luting procedure, the dual-cure resin cement *NX3 /*Kerr Corp was used in all four restorations groups.

The adhesive systems were applied on the prepared cavities, according to the manufacturer' instructions and the composite inlays were cemented. For this, the luting material was applied on the cavity surfaces, the inlays were set into place, and the excess of luting material was removed. All restorations were light-cured (*Optilux* 501/Kerr Corp) for 40 seconds on each tooth surface. All restorations were finished and polished using diamond burs and flexible discs (OptiDisc/Kerr Corp).

The restored teeth were kept for 24 hours in distilled water at room temperature, then embedded in self-curing acrylic resin (Duracryl /SpofaDental, Kerr Company) and sectioned mesio-distally with a low speed diamond saw under cooling water (Isomet, Buehler Ltd, USA). Specimens of 1,5 mm width were obtained.

All specimens were analysed by scanning electron microscopy (SEM Inspect S, FEI, Netherlands) at different magnifications to put in evidence the quality of the adhesive interface.

3. Results and discussion

In this study four different adhesive systems were used for luting Barodent composite inlays that is two experimental Romanian self-etch adhesives *AD-3A* and *AD-3B* (Table 1) which were compared with two well recognized self-etch adhesives, *OptiBond® Solo Plus™ Self-Etch Adhesive System* and *OptiBond® All-In-One* from Kerr Company, taken as control.

Table 1. The composition of the investigated experimental adhesive systems.

Group	Code	Bonding System	Composition of the adhesive systems	Characteristics
G1	AD-3A	Two-step self-etch adhesive	Primer: acidic monomers, polyacrylic acid, HEMA, Ethanol, 4-ethyl-dimethylamino benzoate, CQ;	Appearance: yellowish liquid (primer) and white-yellowish paste (adhesive) pH=1.5-2.0 (primer) and 2.5 (adhesive) Water sorption: 20.5 µg/mm^3
			Adhesive: 70 wt%- Bis-GMA, TEGDMA, HEMA, acidic monomer, CQ, dimethylaminoethyl methacrylate & *Filler:* 30% (wt) - hydroxyapatite	
G2	AD-3B	One-step self-etch adhesive	Primer&Adhesive: acidic monomers, polyacrylic acid, HEMA, Bis-GMA, ethanol, CQ, 4-ethyl-dimethylamino benzoate & *Filler:* 30 wt% - hydroxyapatite	Appearance: white-yellowish paste, slightly fluid pH=2.5 Water sorption: 19.8 µg/mm^3

The Romanian *self-etch* adhesive systems contain polyacrylic acid, which has the role to demineralise the dentin surface, and to create optimal conditions for the adhesive to penetrate the dentin and form a homogeneous hybrid layer that improves the adhesion quality. They both contain about 30 % hydroxyapatite that diminishes the material porosity. Adhesives with hydroxyapatite absorb of about 2.5 times less water than the materials without filler.

The SEM images for specimens in Group 1 (G1) of restored teeth revealed a continuous interface between tooth-luting cement and luting cement- composite inlay as well (*Fig. 1A- 200 × magnification)*. An uniform and homogeneous adhesive layer could be observed between the luting cement and the dental substrate. This one infiltrated the superficial layer from the dentin surface, also known as smear-layer (*Fig. 1 B- 800 × magnification*) and forming a thin hybrid layer (*Fig. 1 C- 3000 × magnification*).

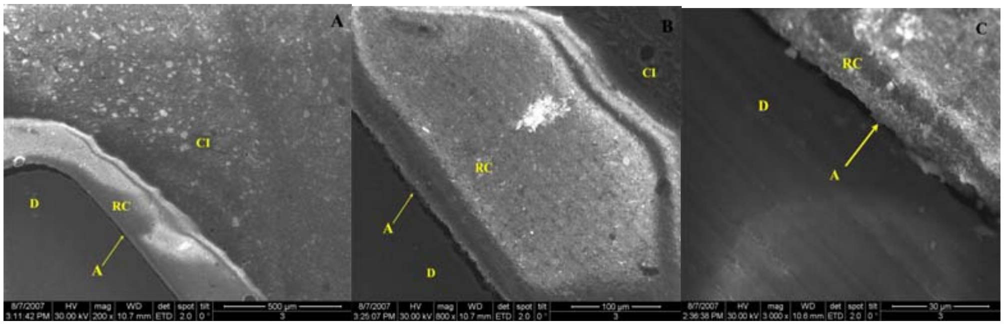


Fig.1. SEM images for G1 (AD-3A+NX3) in different investigated regions and with different magnifications CI-composite inlay; RC- resin cement; D-tooth(dentin); A- adhesive.

SEM investigation of Group 2 (G2) of samples shows the presence of a continuous, gap-free interface between the tooth and the luting resin cement as well as a good adhesion between the resin cement and the composite inlay (*Fig. 2A- 200 × magnification*). The microscopy images illustrate the uniformity of the adhesive layer between the tooth and the luting resin cement (*Fig. 2B-700 × magnification)*. The presence of a thin and uniform hybrid layer along the tooth-adhesive interface could be also observed at larger magnifications (*Fig.2C-1600 × magnification)*.

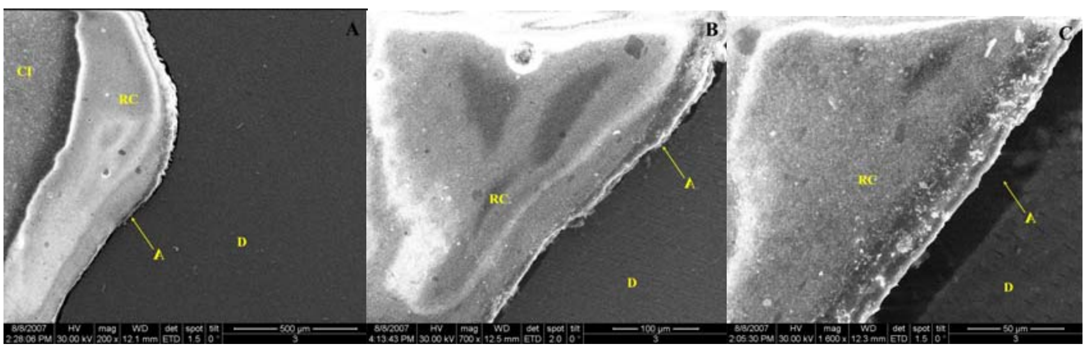
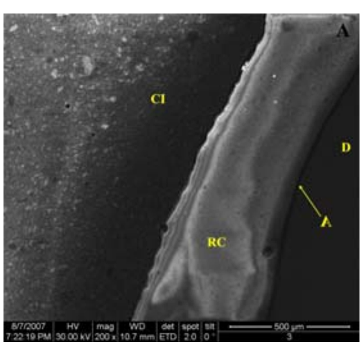
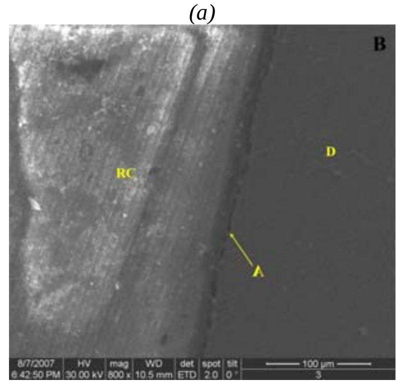


Fig.2 SEM images for G2(AD-3B+NX3) in different investigated regions and with different magnifications CI-composite inlay; RC- resin cement; D-tooth(dentin); A- adhesive.

For Group 3 (G3) of indirect restorations, a good adaptation between tooth and luting cement could be observed without gaps and a continuous adaptation between the resin cement and the composite inlays (*Fig.3 A- 200 × magnification*). At larger magnifications SEM micrographs showed the good adaptation of the luting cement to the dental substrate and the uniformity of the adhesive layer. The formation of a hybrid layer resulting from the adhesive infiltration into the underlying dentin was put in evidence (*Fig.3B- 800 × magnification*).



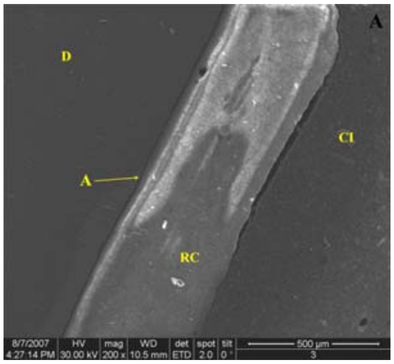
(a)



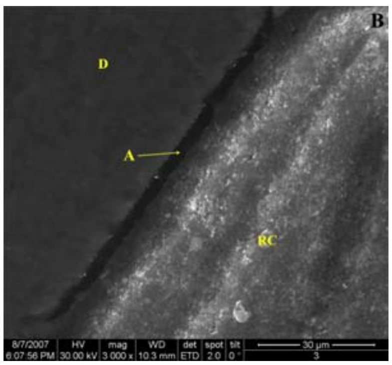
(b)

Fig.3 SEM images for G3(Optibond SoloPlus Self-Etch Adhesive System +NX3 CI-composite inlay; RC- resin cement; D-tooth(dentin); A- adhesive.

SEM images for Group 4 (G4) revealed a good adaptation, lack of gaps along the interface between the resin cement and the tooth (*Fig. 4A- 200 × magnification)* and the formation of a thin hybrid layer along this interface (*Fig. 4B- 3000 × magnification*).



(a)



(b)

Fig.4 SEM images for G 4(OptiBond® All-In-One +NX3) CI-composite inlay; RC- resin cement; D-tooth(dentin); A- adhesive.

Self-etch adhesives simplify the adhesive techniques by acting simultaneously as a conditioning agent and primer. Their way of action on the dental substrate, both enamel and dentin, the efficiency of realising the hybrid layer influence the marginal fit of indirect restorations.

In *Group 1*, the hybrid layer was thinner as compared with the control Groups 3 and 4. The experimental AD-3A product is a mild self-etch adhesive with a relatively low pH value, probably due to the small amount of acidic monomers in the adhesive system. It is known that the acidic monomers can have a reduced conditioning effect with the formation of a thinner hybrid layer [21]. For this reason, hydroxyapatite was introduced as filler in the composition of AD-3A adhesive. Thus the decrease of the micromechanical adhesion caused by the thin hybrid layer should be compensated by the chemical bond between the monomers, hydroxyapatite and the collagen from dental structures [21].

For *Group 2*, a very thin adhesive layer was observed at the interface between the resin cement and the dentin. Probably because of a lower viscosity of the adhesive, a very small adhesive layer remains on the dentinal surface after the solvent evaporation. A thin hybrid layer has negative effects on adhesion, because of the polymerisation taking place in the atmosphere; the oxygen will inhibit the polymerisation in the superficial layers of the adhesive, so that a thin layer of unpolymerised adhesive could remain at surface, affecting the adhesion resistance in time [21]. AD-3B is an all-in- one adhesive with a pH of 2.5, that could explain the formation of a very thin hybrid layer. It could be supposed that by adding hydroxyapatite to its composition the adhesion could become stronger by formation of chemical bonds.

For *Group 3 and 4*, used as control, the results of the SEM analysis showed the presence of continuous interfaces between tooth- luting cement-inlay. Thus All-in One® (Kerr*)* adhesive system produces a homogeneous and uniform adhesive layer between the resin cement and the dentinal surface. The adhesive infiltrated the smear-layer and the underlying dentin (observed at larger magnifications), forming a thin hybrid layer in this case. Regarding the two- step self-etch adhesive system Optibond® SoloPlus ™, a thicker and uniform hybrid layer was observed between the dentin and the resin cement.

The two step self-etch adhesives formed more consistent hybrid layers than the all-in-one adhesives. Thus, Optibond Self-Etch created a thicker hybrid layer than Optibond All-in-One, and AD-3A a thicker hybrid layer than AD-3B, even if the one- step self-etch adhesives are more acids than the two-step self-etch adhesives. This could be explained by the fact in this that the concentration of the acidic monomers in all-in-one adhesive systems is reduced due to dilution with solvent and hydrophilic/ hydrophobic resin monomers in the same solution [22].

The results show that self-etch adhesives can hardly form a hybrid layer along the dentinal surface. For the experimental adhesives, the hydroxyapatite filler exhibits little influence on adhesion, as long as only the smear-layer is infiltrated. The hydrophilic properties of the monomers in these adhesives rise questions upon their resistance in a ‘wet’ environment like dentin, with permanent internal and external sources of water.

For the adhesives that lead to the formation of a hybrid layer, this one was very thin so that it does not influence the immediate resistance of the adhesion, but rises questions regarding the resistance in the wet oral environment, under masticatory forces. In fact, there are studies that proved that, the adhesion values of these types of adhesives decrease with about 30-40% in 6 months [23]. However two-step self-etch adhesives appear to produce better adhesive bonds than all-in-one adhesives.

4. Conclusions

Scanning Electron Microscopy analysis performed on composite inlays luted with different adhesive systems helped to evaluate the tooth-resin cement-inlay interfaces and to put in evidence the differences regarding the quality of the adaptation of the investigated restorations. In most of the cases, the presence of continuous interfaces tooth / resin cement and resin cement/inlay, as well as a good adaptation of the composite inlays was observed. Both experimental adhesives, the two-step self-etch adhesive (AD-3A) and the one-step self-etch adhesive (AD-3B) produced a good adhesion to the dental substrate, forming an uniform interface between the tooth and the luting resin cement and a thin hybrid layer.

This study showed that the Romanian experimental self-etch adhesive systems are capable to produce a good adaptation to the dental substrate and have a good compatibility with the resin cement used for luting the composite inlays. The results prove that the quality of the experimental adhesives is comparable to that of the trade-mark adhesive systems used as control.

Self-etch adhesives assure an immediate, good bond to the dental tissues, but the big concentration of acidic monomers and the thin hybrid layer may have a negative effect on the adhesion stability and on its resistance in oral environment. The two-step self-etch adhesives produced a deeper and a much homogeneous hybrid layer than all-in-one adhesives.

The results of this *in vitro* study emphasize also the benefits of the self-etch adhesives regarding their simplified clinical protocol.

References

[1] D. A. Gerdolle, E. Mortier, C. Loos-Ayav, B. Jacquot, M. Panighi. J Prosthet Dent **93**, 563 (2005).
[2] W. J. O'Brien, Dental Materials and their selection, 3rd ed, Quintessence, 2002.
[3] M. Irie, K. Suzuki. Dent Mater **17**, 347 (2001).
[4] U. Pallesen, V. Qvist, Clin. Oral Invest. **7,** 71 (2003).
[5] O. S. De Andrade, M. F. De Goes, Dental Materials **23**, 279 (2007).
[6] I. Krejci, F. Lutz, L. Gautschi. J. Prosthet. Dent. **72**, 233 (1994).
[7] M. Federlin, S. Schmidt, K. A Hiller, B. Thinemann, G. Schmalz. Operative Dentistry **29**(5), 560 (2004).
[8] C. J. Soares, L. Celiberto, P. Dechichi, R. B. Fonseca, L. R. M. Martins, Braz. Oral Res. **19**(4), 295 (2005).
[9] C. J. Soares, L. R. Martins, A. J. Fernandes Neto, M. Giannini, Operative Dentistry **28**(6), 689 (2003).
[10] D. Fortin, E. J Swift Jr, G. E Denehy, J. W Reinhardt, Dental Materials **10,** 253 (1994).
[11] R. Liberman, A. Ben-Amar, L. Herteanu, H. Judes, J. Oral Rehabil, **24,** 26 (1997).
[12] S. Karakaya, A. Sengun, F. Ozer, J. Oral Rehabil. **32**, 448 (2005).
[13] K. Hikita, B. Van Meerbeek, J. De Munck, T. Ikeda, K. Van Landuyt, T. Maida, P. Lambrechts, M. Peumans. Dental Materials **23,** 71 (2007).
[14] A. K. B. Bedran de Castro, P. E. C. Cardoso, G. M. B Ambrosano, L. A. F. Pimenta. Operative Dentistry **29**(1), 42 (2004).
[15] S. D Heintze, J. Adhes. Dent., **9**(1), 77 (2007).
[16] A. K. B Bedran de Castro, P. N. R. Pereira, L. A. F. Pimenta, J. Y. Thompson, Operative Dentistry **29**(2), 150 (2004).
[17] A. R Yazici, Ç. Çelik, G. Özgünaltay, B. Dayangaç, Operative Dentistry **32**, 166 (2007).
[18] S. Deliperi, D.N Bardwell, C. Wegley. Operative Dentistry, **32**-2, 179 (2007)
[19] A. Popovici, D. Borzea, A. Pop, A. Roman, O. Pastrav, M. Moldovan, C. Prejemerean, Eng. of Biomaterials **47-53**(VII), 15 (2005).
[20] A. Popovici, O. Fodor, M. Moldovan, A. Roman, D. Borzea. Optoelectron. Adv. Mater. – Rapid Comm. **2**(12), 891(2008).
[21] K. Van Landuyt, J. De Munck, E. Coutinho, M. Peumans, P. Lambrechts, B. Van Meerbeek, Interfacial Phenomena and Related Properties, eds. G Eliades, DC Watts, T Eliades, Springer Verlag, 2005.
[22] Y. Torii, K. Itou, R. Hikasa, S. Iwata, Y. Nishitani, J. Oral Rehabil. **29,** 528 (2002).
[23] A. Reis, V. Grandi, L. Carlotto, G. Bortoli, R. Patzlaff, M. L Rodrigues Accorinte, A. D Loguercio, J. Dent. **33,** 549 (2005).

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