UNIVERSIDADE ESTADUAL DE CAMPINAS

FACULDADE DE ODONTOLOGIA DE PIRACICABA

THAIANE RODRIGUES AGUIAR

CIMENTOS RESINOSOS CONVENCIONAIS E AUTOADESIVOS: CARACTERIZAÇÃO DAS PARTÍCULAS DE CARGA, ULTRAMORFOLOGIA E RESISTÊNCIA DA UNIÃO RESINA-DENTINA

TESE DE DOUTORADO APRESENTADA À FACULDADE DE ODONTOLOGIA DE PIRACICABA/UNIVERSIDADE ESTADUAL DE CAMPINAS PARA A OBTENÇÃO DO TÍTULO DE DOUTORA EM CLÍNICA ODONTOLÓGICA, NA ÁREA DE DENTÍSTICA

ORIENTADOR: PROF. DR. MARCELO GIANNINI

ESTE EXEMPLAR CORRESPONDE À VERSÃO FINAL DA TESE/DISSERTAÇÃO DEFENDIDA PELA ALUNA, E ORIENTADA PELO PROF. DR.

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PIRACICABA, 2011

FICHA CATALOGRÁFICA ELABORADA PELA
BIBLIOTECA DA FACULDADE DE ODONTOLOGIA DE PIRACICABA
Bibliotecária: Marilene Girello – CRB-8ª. / 6159

Ag93c

Aguiar, Thaiane Rodrigues.
Cimentos resinosos convencionais e autoadesivos: caracterização das partículas de carga, ultramorfologia e resistência de unlão resina-dentina / Thaiane Rodrigues Aguiar. -- Piracicaba, SP: [s.n.], 2011.

Orientador: Marcelo Giannini.
Tese (Doutorado) – Universidade Estadual de Campinas, Faculdade de Odontologia de Piracicaba.

1. Cimentação. 2. Compostos inorgânicos. 3. microscopia eletrônica de varredura. 4. Microscopia confocal . 5. resistência à tração. I. Giannini, Marcelo. II. Universidade Estadual de Campinas. Faculdade de Odontologia de Piracicaba. III. Título.
(mg/fop)

Título em Inglês: Conventional and self-adhesive resin cements: filler particles characterization, ultramicromorphology and dentin-resin bond strength

Palavras-chave em Inglês (Keywords): 1. Cimentation. 2. Inorganic chemicals. 3. Microscopy, Electron, Scanning. 4. Microscopy, Confocal. 5. Tensile Strength

Área de Concentração: Dentística

Titulação: Doutor em Clínica Odontológica

Banca Examinadora: Marcelo Giannini, Alessandro Dourado Loguercio, Paulo Francisco Cesar, Americo Bortolazzo Correr, Mario Alexandre Coelho Sinhoreti

Data da Defesa: 20-05-2011

Programa de Pós-Graduação em Clínica Odontológica

UNIVERSIDADE ESTADUAL DE CAMPINAS
Faculdade de Odontologia de Piracicaba

A Comissão Julgadora dos trabalhos de Defesa de Tese de Doutorado, em sessão pública realizada em 20 de Maio de 2011, considerou a candidata THAIANE RODRIGUES AGUIAR aprovada.

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DEDICATÓRIA

À Deus, por iluminar o meu caminho e tornar tudo possível.

Aos meus pais, Antonio e Mauraci, pelo exemplo de amor e dedicação. Obrigada por estarem sempre ao meu lado e por serem pessoas tão especiais para mim.

À minha querida irmã, Laira, pelas constantes palavras de apoio e carinho.

Ao meu marido, Fábio, por aceitar e apoiar as minhas escolhas, dedicando amor, carinho e compreensão.

Obrigada por tudo!

AGRADECIMENTO ESPECIAL

Ao Prof. Dr. Marcelo Giannini por ter me acolhido como sua orientada durante o Mestrado e o Doutorado. A sua dedicação à ciência, profissionalismo e seriedade tenho como exemplo para minha vida profissional. Não posso deixar de agradecer os muitos ensinamentos transmitidos, a sua disponibilidade em ajudar e pelas muitas oportunidades oferecidas que contribuíram tanto para o meu crescimento profissional como pessoal. Muito obrigada pela confiança em mim depositada e pela sua amizade.

AGRADECIMENTOS

À Faculdade de Odontologia de Piracicaba (FOP), da Universidade Estadual de Campinas (UNICAMP), representada na pessoa do diretor Prof. Dr. Jacks Jorge Junior e do diretor associado Prof. Dr. Alexandre Augusto Zaia;

À coordenadora dos cursos de Pós-graduação da FOP-UNICAMP Profa. Dra. Renata Cunha Matheus R. Garcia e ao Prof. Dr. Márcio de Moraes, coordenador do programa de Pós-graduação em Clínica Odontológica;

À Profa. Ana Karina Bedran-Russo, por ter gentilmente aceito me orientar durante o estágio de doutoramento na University of Illinois at Chicago;

À Profa. Dra. Paula Mathias, pela amizade e apoio à pesquisa. O seu incentivo aprimorou o meu interesse pela vida acadêmica e hoje, também dedico essa vitória a você;

Ao Prof. Cesar Arrais, pela muitas horas de convivência na Universidade de Guarulhos. Muito obrigada pela sua disponibilidade, atenção e pela participação nos trabalhos desenvolvidos durante o Doutorado;

Aos Professores que estiveram presentes na banca de exame de qualificação, Prof. Dr. André Reis, Profa. Dra. Regina Puppin e Prof. Dra. Vanessa Cavalli, pela valiosa contribuição à este trabalho;

A Coordenação de Aperfeiçoamento Profissional de Nível Superior (Capes) pela concessão da bolsa de estudo no primeiro semestre de Doutorado e à Fundação de Amparo à Pesquisa o Estado de São Paulo – Fapesp, pela bolsa de estudo subsequente (processo 09/51281-4) e auxílio pesquisa (09/51674-6);

Ao corpo docente do Programa em Clínica Odontológica da Faculdade de Odontologia de Piracicaba, em especial aos professores da Área de Dentística, Prof. Dr. Marcelo Giannini, Profa. Dra. Giselle Maria Marchi Baron, Prof. Dr. Flávio Henrique Baggio Aguiar, Prof. Dr. Luis Roberto Marcondes Martins, Prof. Dr. Luís Alexandre Maffei Sartini Paulillo e ao Prof. Dr. José

Roberto Lovadino pela oportunidade de aprimorar os meus conhecimentos durante a realização do Doutorado;

A Profa. Dra. Gláucia Maria Bovi Ambrosano pela realização das análises estatísticas;

Às empresas 3M ESPE e Kuraray Medical Inc. pela doação dos materiais utilizados neste trabalho;

Ao centro de Microscopia Eletrônica de Varredura da FOP-UNICAMP e aos funcionários Adriano Luis Martins e Eliene Orsini N. Romani, pela atenção e disponibilidade em ajudar;

Aos funcionários da Área de Dentística, Mônica e Pedro Justino, pela presteza e dedicação;

A todos os colegas do curso de Pós-Graduação em especial aos amigos do curso de Doutorado, Giulliana, Adriano, Maria, Cíntia, Bruno, Marina, Gisele e Anderson pela amizade e por compartilhar as alegrias e as dificuldades durante este período;

Às amigas Adriana e Sandrine pelos conselhos e companheirismo em todos os momentos;

À amiga Carolina Bosso, pela participação no desenvolvimento desse trabalho em todos os momentos que precisei;

A todos que torceram por mim, em especial aos familiares e amigos, que mesmo distantes sempre estiveram me apoiando;

MEUS SINCEROS AGRADECIMENTOS!

"Quero, um dia, dizer às pessoas que nada foi em vão... Que o amor existe, que vale a pena se doar às amizades e às pessoas, que a vida é bela sim e que eu sempre dei o melhor de mim... e que valeu a pena."

(Mário Quintana)

RESUMO

O emprego dos cimentos resinosos na fixação de peças protéticas têm sido avaliado em muitos estudos, entretanto, trabalhos que simulem os desafios presentes na cavidade bucal e o comportamento a longo prazo dos cimentos resinosos autoadesivos ainda são escassos na literatura. Diante disso, o presente estudo teve como objetivo: 1) verificar a composição inorgânica das partículas de carga e caracterizá-las quanto ao tipo e morfologia; 2) analisar a ultramorfologia da interface de união dentina-cimento resinoso através da microscopia confocal por varredura a laser e da microscopia eletrônica de varredura (MEV); 3) avaliar o efeito do armazenamento por 1 ano ou da ciclagem mecânica (50.000 ciclos) na resistência de união dentina-cimento resinoso. Quatro cimentos resinosos foram avaliados: dois autoadesivos (RelyX Unicem, 3M ESPE e Clearfil SA Luting, Kuraray Medical Inc.) e dois convencionais (RelyX ARC/Adper Scotchbond Multi-Purpose Plus, 3M ESPE e Clearfil Esthetic Cement/Clearfil DC Bond, Kuraray Medical Inc.). Para analisar os principais componentes inorgânicos e avaliar as características das partículas de carga, o conteúdo orgânico dos cimentos resinosos foi removido por solventes orgânicos e as amostras analisadas em MEV/EDX (Espectroscopia de energia dispersiva por raios-X). Terceiros molares humanos foram utilizados na avaliação da ultramorfologia da interface dentina-cimento resinoso e no ensaio de resistência de união à dentina. Na análise da ultramorfologia da área de união, corantes (fluoresceína e rodamina) foram previamente adicionados aos materiais estudados para possibilitar a obtenção das imagens. Para o ensaio de resistência de união, discos de resina composta indireta foram confeccionados visando simular as restaurações indiretas. Após a cimentação, os dentes restaurados foram submetidos a três tratamentos e ao ensaio de microtração: 1- espécimes testados após 24 h (grupo controle); 2- ciclagem mecânica e em seguida testados; 3- armazenados em saliva artificial por 1 ano na forma de "palitos" e testados. Os dados obtidos a partir do ensaio de resistência de união foram analisados estatisticamente (ANOVA e teste Tukey, $p<0,05$). Nos outros dois estudos, a análise descritiva dos dados obtidos a partir das imagens microscópicas foi realizada. Na avaliação MEV/EDX notou-se elevado conteúdo de silício em todos os materiais. O cimento RelyX ARC apresentou partículas de carga esféricas e irregulares, enquanto os outros materiais demonstraram partículas com formato irregular. Nas fotomicrografias, a hibridização dentinária somente foi observada para os sistemas de cimentação convencional. No ensaio de resistência de união, os tratamentos não influenciaram a resistência de união dos cimentos resinosos à dentina. Os cimentos autoadesivos apresentaram médias de resistência de união significantemente superiores aos chamados convencionais. Embora os materiais avaliados apresentem partículas de carga com distintas composições inorgânicas e interagem com a dentina através de diferentes mecanismos, os tratamentos de envelhecimentos propostos não reduziram a resistência de união à dentina, quando comparados ao grupo controle. Os cimentos autoadesivos não formam camada híbrida na dentina intertubular, nem *tags* de resina nos túbulos dentinários, entretanto, apresentam partículas de carga com maior tamanho e demonstraram os maiores valores médios de resistência de união à dentina entre os cimentos estudados.

Palavras-chave: Cimentos resinosos, Interface adesiva, Resistência de União, Partículas de Carga, Microscopia Confocal.

ABSTRACT

Resin luting materials to fixed prosthetic restorations have been evaluated in many studies, however, little is known about self-adhesive resin cements, mainly involving long-term durability and when these materials have been submitted to challenges of oral environment. Thus, the purpose of this study was: 1- to verify the filler particles components and to characterize them according to type and morphology; 2- to analyze the ultramorphology of the dentin-resin interfaces using confocal laser scanning microscopy and scanning electron microscopy (SEM) and 3- to evaluate the effect of the storage for 1 year or the mechanical cycling (50.000 cycles) on bond strength of resin cements to dentin. Four resin cements were used in this study: two self-adhesive resin cements (RelyX Unicem, 3M ESPE and Clearfil SA Luting, Kuraray Medical Inc.) and two conventional resin cements (RelyX ARC/Adper Scotchbond Multi-Purpose Plus, 3M ESPE and Clearfil Esthetic Cement/Clearfil DC Bond, Kuraray Medical Inc.). For indentifying the main inorganic components and to evaluate the filler particles characteristics, the organic phase of resin cements was removed using organic solvents and the sample was analyzed by SEM/EDX (Energy-dispersive X-ray spectroscopy). Human third molars were used for micromorphology analysis of resin-dentin interfaces and microtensile bond strength test. For micromorphology analyses of resin-dentin interfaces, two fluorescent dyes (fluorescein and rhodamine B) were incorporated into the materials previously. For microtensile bond strength, pre-polymerized resin discs were prepared to simulate indirect restorations. After bonding procedures, the restored teeth were submitted to three treatments: 1- control group (tested 24 h after specimen preparation); 2- tested after mechanical cycling; 3- beams were storage for one year in artificial saliva and tested. Bond strength data were statistically analyzed by two-way ANOVA and Tukey test ($\alpha<0.05$). Qualitative analysis was performed for two other studies using microscopic images. For SEM/EDX measurements, high amount of silicon was identified for all cements.

The resin cement RelyX ARC showed spherical and irregular particles, while other cements presented irregular shape fillers. Hybrid layer formation was observed only for conventional resin cements in micromorphology analyses. For microtensile test, the treatments did not reduce the bond strength between dentin-resin cement. Self-adhesive resin cements provide significantly higher bond strength than those conventional materials. Despite the resin cements showed filler particles with differences in the inorganic composition and different bonding mechanisms, the mechanical cycling and storage for one year did not affect the bond strength to dentin when it was compared to control group. The self-adhesive resin cements showed no hybrid layer formation in intertubular dentin and nor resin tags in the dentinal tubules were detected. However, these materials showed the biggest filler particles and higher bond strength to dentin than conventional resin cements studied.

KEYWORDS: Resin cements, Adhesive interface, Bond strength, Filler particles, Confocal microscopy.

SUMÁRIO

INTRODUÇÃO

A cimentação adesiva de peças protéticas (inlays, onlays, coroas, facetas e laminados) constitui uma parcela representativa dos tratamentos de reabilitação e estéticos restauradores atuais (Hikita, *et al*., 2007). Fatores estéticos associados aos conceitos de preservação da estrutura dental, através do uso da técnica adesiva, têm gerado o crescente emprego e o aprimoramento das técnicas de cimentação adesiva (Abo-Hamar *et al*., 2005).

O emprego de cimentos resinosos de dupla polimerização (ativação química e foto-ativação) na cimentação adesiva visa garantir que a reação química de polimerização ocorra mesmo na ausência de luz, possibilitando a indicação desses materiais em diferentes situações clínicas como na fixação de pinos de fibra e de restaurações indiretas (Rosenstiel *et al*., 1998; Braga *et al*., 1999; Arrais *et al*., 2007; Aguiar *et al*., 2010). Os cimentos resinosos de dupla polimerização apresentam reduzida sorção de água e a baixa solubilidade, quando comparados aos cimentos odontológicos tradicionais e elevada estabilidade de cor, quando comparados aos cimentos apenas autopolimerizáveis (Tanoue *et al*., 2003).

A classificação atual dos cimentos resinosos de dupla polimerização em convencionais ou autoadesivos envolve a utilização ou não de um agente de união durante os procedimentos adesivos. Os cimentos convencionais são empregados em combinação com um sistema adesivo, que pode ser do tipo *etch-and-rinse* ou autocondicionante (Hikita *et al*., 2007; Duarte *et al*., 2008; Viotti *et al*., 2009). Por outro lado, os cimentos resinosos autoadesivos dispensam o pré-tratamento dentinário (condicionamento ácido, aplicação do *primer* e adesivo) uma vez que sua matriz orgânica contém monômeros multifuncionais de metacrilato derivados do ácido fosfórico, que interagem quimicamente com a hidroxiapatita presente no tecido dentário (De Munck *et al.*, 2004, Abo-Hamar *et al.*, 2005, Hikita *et al.*, 2007). Assim, a simplificação do protocolo adesivo permite reduzir o tempo de atendimento clínico, além de facilitar a técnica de cimentação, pois, etapas consideradas críticas, como a aplicação do ácido fosfórico, a sua remoção com água, o controle da umidade dentinária pós-condicionamento e a aplicação do sistema adesivo são eliminadas (De Munck *et al.*, 2004; Abo-Hamar *et al.*, 2005; Yang *et al.*, 2006; Goracci *et al*., 2006; Piwowarczyk *et al.*, 2007; Han *et al.*, 2007), reduzindo as dificuldades inerentes à técnica adesiva, até então relatada somente para os cimentos resinosos convencionais (Piwowarczyk *et al.,* 2004).

Trabalhos científicos têm demonstrado a influência das características das partículas de carga nas propriedades físico-mecânicas de materiais resinosos (Chung *et al.*, 1990; Kim *et al.*, 2002; Han *et al.*, 2007; Polydorou *et al.*, 2009). Weiner (2007) descreve os cimentos resinosos como compósitos de baixa viscosidade, que apresentam reduzido conteúdo de carga envolvido numa matriz orgânica monomérica. Entretanto, estudos que apresentam informações detalhadas sobre a morfologia e a composição das partículas de carga dos cimentos resinosos autoadesivos são escassas na literatura.

Estudos têm avaliado a resistência de união dos cimentos autoadesivos à dentina, comparando com cimentos tradicionalmente empregados na Odontologia. Piwowarczyk *et al*., em 2004, mostraram que os cimentos resinosos convencionais e autoadesivos apresentaram valores de resistência de união superiores aos dos cimentos de fosfato de zinco, de ionômero de vidro e dos cimentos de ionômero de vidro modificado por resina. De Munck *et al*. (2004), Hikita *et al*. (2007) e Aguiar *et al*. (2008) observaram valores de resistência de união semelhantes entre cimentos resinosos autoadesivos e os cimentos resinosos convencionais. Entretanto, a longevidade das restaurações indiretas, principalmente relacionada ao uso dos sistemas autoadesivos é um dado importante ainda pouco estudado.

A longevidade das restaurações indiretas está intimamente relacionada à efetividade da união, que envolve o tecido dentinário, o agente de união, o cimento resinoso e a restauração indireta. O estudo da longevidade dos procedimentos restauradores em laboratório é feito através do envelhecimento *in vitro* das amostras restauradas que podem ser feito por meio do armazenamento da amostra em solução aquosa (Yamauti *et al*., 2003; Uceda-Gómez *et al.,* 2007; Abdalla *et al.,* 2008; de Morais *et al.,* 2008; Reis *et al.*, 2008) como também, através da ciclagem mecânica (Nikaido *et* *al.*, 2002; de Paula *et al.*, 2008; Lodovici *et al.*, 2009, Naumann *et al.*, 2010). O armazenamento das amostras restauradas visa avaliar o comportamento dos materiais com relação à degradação hidrolítica, enquanto, o teste de ciclagem mecânica busca simular o dente restaurado em função oclusal, mostrando os efeitos da mastigação na restauração e no dente.

Considerando-se que o conhecimentos das partículas de carga fornece importantes informações sobre as propriedades mecânicas dos materiais resinosos, o presente estudo teve como objetivos avaliar a composição inorgânica e às características morfológicas das partículas de carga dos cimentos resinosos. Adicionalmente, foram avaliadas a morfologia da interface de união e a resistência da união dentina-cimento resinoso-resina indireta imediata e após o envelhecimento das amostras (ciclagem mecânica ou o armazenamento em saliva artificial por 1 ano). Desta forma, o estudo *in vitro* teve como objetivo geral avaliar quatro cimentos resinosos (convencionais e autoadesivos) quanto às características das partículas de carga, ultramorfologia e resistência de união entre a restauração indireta-cimento resinoso e o tecido dentinário.

Os objetivos específicos foram:

1. Analisar os componentes inorgânicos dos cimentos resinosos e caracterizar as partículas de carga presentes em cada material, empregando a microscopia eletrônica de varredura (MEV)/Microanálise de energia dispersiva de raios X (EDX);
2. Avaliar a ultra-morfologia da área de união dentina-restauração indireta formada por sistemas de cimentação, utilizando o microscópio confocal de varredura laser (MCVL) e MEV;
3. Avaliar a resistência da união dentina-restauração indireta formada por sistemas de cimentação que foram testados após três tratamentos: controle (teste 24 horas após a confecção das restaurações); imediatamente após ciclagem mecânica; armazenamento por 1 ano em saliva artificial, utilizando o ensaio de microtração;

Inorganic composition and filler particles morphology of conventional and self-adhesive resin cements by SEM/EDX

Running title: Inorganic composition and filler analysis of resin cements

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Key Words: resin luting cement, inorganic composition, filler characterization, EDX.

ABSTRACT

The purpose of this study was to characterize the inorganic components and morphology of filler particles of conventional and self-adhesive dual-curing resin luting cements. The main components were identified by energy dispersive X-ray spectroscopy microanalysis (EDX) and filler particles were morphologically analyzed by scanning electron microscopy (SEM). Four resin cements were used in this study: two conventional resin cements (RelyX ARC/3M ESPE and Clearfil Esthetic Cement/Kuraray Medical Inc.) and two self-adhesive resin cements (RelyX Unicem/3M ESPE and Clearfil SA Luting/Kuraray Medical Inc). The materials (n=5) were manipulated according to manufacturers' instructions, immersed in organic solvents to eliminate the organic phase and observed under SEM/EDX. Although EDX measurements showed high amount of silicon for all cements, differences in elemental composition of materials tested were identified. RelyX ARC showed spherical and irregular particles, whereas other cements presented only irregular filler shape. In general, self-adhesive cements contained higher filler size than conventional resin luting cements. The differences in inorganic components and filler particles were observed between categories of luting material and among them. All resin cements contain silicon, however, other components varied among them.

INTRODUCTION

Resin-based cements have been routinely used for cementation of indirect esthetic restorations. The advantages of resin cements include bonding to tooth structure (Aguiar, 2010a; Arrais, 2008; Hikita, 2007; Viotti, 2009), dual-curing mode (Aguiar 2010b; Arrais, 2008, De Menezes, 2006), low water sorption, low solubility and high color stability (Tanoue, 2003). These materials can be classified in two categories according to adhesive cementing technique: conventional and self-adhesive resin cements. Conventional adhesive cementation technique requires the pretreatment of tooth surface with a bonding adhesive agent (etch-and-rise or self-etching), while the self-adhesive cements do not require phosphoric acid treatment or any special adhesive system for bonding to tooth structure (Abo-Hamar, 2005; De Munck, 2004; Duarte, 2008; Gerth, 2006).

For self-adhesive resin cements, multifunctional phosphoric acid methacrylate monomers were incorporated for developing self-adhesive characteristics (De Munck, 2004). According to Weiner (2007), resin cements are low-viscosity composites, containing reduced filler content and a resin matrix based on different monomers (Berger, 2009), such as Bis-GMA, TEGDMA and dimethacryates. Other components like glasses and/or ceramic fillers that contain chemical elements such as barium, strontium and zirconium can be added to provide radiopacity characteristics (Anusavice, 2004). The incorporation of filler particles into a resin matrix and filler characteristics (i.e. radiopacity, filler distribution, shape and size) changes the physical properties, such as elastic modulus, compressive and tensile strength (Chung, 1990; Kim, 2002; Leprince, 2010; Polydorou, 2009, Shinkai, 2001).

Bond mechanism and mechanical properties of conventional and self-adhesive resin cements have been the topic of recent investigations (Aguiar, 2010a, b; De Munck, 2004; Viotti, 2009). However, their inorganic composition and respective filler particles morphology between conventional and self-adhesive resin cements are not widely reported and need more characterization. Therefore, the purpose of this study was to investigate if there are differences in the inorganic composition of filler particles of conventional and self-adhesive resin cements by energy dispersive X-ray spectroscopy microanalysis (EDX). In addition, the filler particles characteristics were determined using scanning electron microscopy (SEM). The research hypothesis tested was that there are differences in the inorganic composition of conventional and self-adhesive resin cements. In addition, it was hypothesized that there are not differences in filler morphology characteristics for all materials.

MATERIALS AND METHODS

Experimental Groups

Two conventional (RelyX ARC, 3M ESPE, Saint Paul, MN, USA and Clearfil Esthetic Cement, Kuraray Medical, Kurashiki, Japan) and two self-adhesive resin cements luting (RelyX Unicem, 3M ESPE, Seefeld, Germany and Clearfil SA Cement, Kuraray Medical, Kurashiki, Japan) were selected for this study (Table 1). In order to evaluate the inorganic composition and characteristics of the fillers, the resin cements were manipulated according to manufacturers' instructions.

Sample preparation for inorganic analyses and filler characterization

An amount of 60 ± 1 mg was used from each material (n=5). Immediately after mixing, the unpolymerized resin luting cements were dissolved in 6 mL of acetone (99.5%), and centrifuged for 5 min. This procedure was repeated three times with a 24h interval. After that, the chloroform solution (99.8%) was used the same manner (Sabbagh, 2004). The remaining filler particles were immersed in 6 mL absolute ethanol for one day followed by air-drying overnight at 37° C. Samples were fixed in plastic stubs, sputter coated with carbon (MED 010, Balzers, Balzer, Liechtenstein) to eliminate the charging effects. After that, the samples were observed by scanning electron microscope (SEM)/ energy dispersive x-ray analysis (EDX).

The EDX was used to detect the main inorganic components of tested materials. Specimens were identified by using a SEM operating with a Vantage System (Noran Instruments, Middleton, WI, USA). The spectra for EDX measurements were obtained for 100s livetime (voltage: 15 kV; dead time 20-25%; working distance: 20 mm).

For characterization of the inorganic fraction, specimens were observed using a SEM (VP 435, Leo, Cambridge, UK). It was used five repetitions (n=5) of each resin cement to analyze the filler particles morphology. SEM images of filler particles were taken at x1.000 and x3.000 magnifications from each resin cement sample (voltage: 15 kV; beam width: 25 a30 nm; working distance: 10-15 mm). Therefore, it was obtained five images at X1,000 and five at X3,000 per group to be analyzed. The five images with 3,000X magnification were used to calculate the fillers sizes for each cement. The measurement of fillers sizes for each resin cement was performed using the scale markers of the figures.

RESULTS

Inorganic elements identified by scanning electron microscope/energy dispersive X-ray spectroscopy microanalysis are demonstrated in Figure 1. The elemental composition of RelyX ARC by EDX shows zirconium and high amounts of silicon (Figure 1A). EDX measurements showed that Clearfil Esthetic Cement contain barium, aluminum and silicon elements (Figure 1B). RelyX Unicem presents fluoride, sodium, aluminum, silicon, calcium and lanthanum in its inorganic composition (Figure 1C). Clearfil Esthetic showed the same composition of Clearfil SA cement, except for sodium element (Figure 1D).

Filler particles examination by SEM showed morphological variations among resin cements. Figures 2 to 5 show the filler particles of RelyX ARC, Clearfil Esthetic Cement, RelyX Unicem and Clearfil SA Luting, respectively. SEM micrograph of RelyX ARC resin cement showed spherical and irregular aggregates of various sizes (Figure 2A). Irregular-shaped particles can be observed for Clearfil Esthetic Cement, RelyX Unicem and Clearfil SA Luting resin cements (Figures 3 to 5).

In the same magnification, conventional cements showed smaller filler size than self-adhesive resin cements. For conventional resin cements, RelyX ARC showed particle size less than 4 μm (Figure 2B), while Clearfil Esthetic Cement presented irregular shaped fillers ranging from 0.5 to 3 μm (Figure 3A e 3B). For self-adhesive resin luting cements, the average size was less than 5 μm for RelyX Unicem (Figure 4A e 4B), while Clearfil SA Luting contained small particles (around 2 μm) and many particles larger than 5 μm (Figure 5A e 5B).

DISCUSSION

Inorganic composition, which was investigated by energy dispersive X-ray spectroscopy microanalysis (SEM/EDX), is described in Figure 1. In this study, the monomers or the organic matrix were removed from the resin cements using organic solvents to maintain only the filler particles. According to Sabbagh (2004), fillers are easier to observe when this technique is used. As shown in Figure 1, all materials showed higher amount of silicon, which is used to impart radiopacity characteristics. Moreover, the silane treated fillers (silanization process) is important to promote bonds between methacrylate groups in the resin matrix and filler particles (Sabbagh, 2004).

As illustrated in Figure 1 (A, B and D), RelyX ARC contains zirconium and the two Kuraray Medical products tested (Clearfil Esthetic Luting and Clearfil SA Luting) contain barium element, can provide radiopacity. For RelyX Unicem (Figure 1C), higher amount of aluminum components was observed. Aluminum has a similar optical density to tooth structure and it is present in the compositions of glass ionomer cements. In general, the glass ionomer cements contain calcium fluoroaluminosilicate glass, which is attacked by polyacrylic acid to release cations and fluoride ions. Many studies have shown that the fluoride ion is effective to inhibit the demineralization and enhance remineralization of the hard dental under specific dosages (Mukai, 2002; Tenuta, 2009).

The manufacturer reports that the RelyX Unicem combines glass ionomer, adhesive and composite technology. In accordance with manufacturer, this study identified silicon, aluminum and fluoride components for RelyX Unicem resin cement. Thus, it is possible to assume that the RelyX Unicem contains the main components of glass ionomer technology. For the remaining materials, aluminum and fluoride were not present in RelyX ARC cement, however, Clearfil Esthetic Luting and Clearfil SA Luting showed smaller amount of aluminum.

In this study, simplified self-adhesive Clearfil SA luting did not show fluoride element, in spite of the MSDS (material safety data sheet) of this material informs that it contains sodium fluoride, without show the concentration of sodium fluoride in the formulation of the resin cement. EDX measurement identified sodium, however it was not enough sensible to detect the fluoride ion in the composition of Clearfil SA Cement. This fluoride salt is present in the composition of this resin cement and it is important to control the demineralization around restorations in cases of high caries risk.

The amount of inorganic fillers contained in RelyX Unicem and RelyX ARC is approximately 70% and 67.5% by weight, respectively (information supplied by the manufacturer). According to this manufacturer, one part of the fillers of RelyX Unicem is silanated, while another part (glass powder) is available for neutralization reaction with the phosphoric acid groups of the methacrylate monomers. This reaction occurs during the setting of the material with the basic fillers and is related to the bonding mechanism to the dental structures (De Munck, 2004; Hikita, 2007).

Adhesive restorative materials have shown better properties due to the incorporating of fillers, and also because of the bonding established between fillers and resin matrix, which is provided by silane coupling agent (Polydorou, 2009; Shinkai, 2001, Sabbagh, 2004). As shown in Figure 2, RelyX ARC resin cement contains spherical shape filler, while other materials showed predominantly irregular-shaped particles (Figures 3, 4 and 5). RelyX ARC resin cement showed spherical agglomerates of various sizes. The mixture of large and small particles promotes reduction of space between particles, increasing maximum amount of filler into a resin matrix and yielding positive effects on the mechanical properties (Anusavice, 2003). Another type of filler particles combination is between spherical and irregular-shaped particles as observed for RelyX ARC resin cement (Figure 2). In addition, the self-adhesive resin cements showed higher filler size than conventional resin cements and the lower viscosity for self-adhesive resin cements, such as RelyX Unicem, could be related to presence of larger filler particles size.

The present results describe the main inorganic components and filler characterization of the resin cements studied. In vitro complementary studies are needed to verify the organic matrix phase, other chemical components and their function in each product. Also, the investigations about the distribution of the filler particles in the organic matrix and the volumetric content of particles would complement the characterization of the inorganic content of these resin cements.

Based on the findings of this study, EDX microanalyses showed differences in inorganic compositions among the resin cements, except for silicon element. Filler particles examination showed morphological variations between cements and in general, self-adhesive cements contained higher filler size than conventional resin luting cements. Thus, the research hypothesis tested that there are differences in the inorganic composition of conventional and self-adhesive resin cements was accepted, while, the hypothesis that there are not differences in filler morphology characteristics among the materials was not confirmed.

ACKNOWLEDGEMENTS

This study was supported by grants from FAPESP, Brazil (09/51281-4 and 09/51674-6).

REFERENCES

1. Abo-Hamar SE, Hiller KA, Jung H, Federlin M, Friedl KH, Schmalz G. 2005. Bond strength of a new universal self-adhesive resin luting cement to dentin and enamel. Clin Oral Investig 9:161-167.

2. Aguiar TR, Di Francescantonio M, Ambrosano GMB, Giannini M. 2010a. Effect of curing mode on bond strength of self-adhesive resin luting cements to dentin. J Biomed Mater Res B Appl Biomater 93B:122-127.

3. Aguiar TR, Francescantonio MD, Arrais CAG, Ambrosano GMB, Davanzo C, Giannini M. 2010b. Influence of curing mode and time on degree of conversion of one conventional and two self-adhesive resin cements. Oper Dent 35:295-299.

4. Anusavice K. 2003. Philips` Science of Dental Materials. UK: Elsevier; 832 p.

5. Arrais C, Rueggeberg F, Waller J, Degoes M, Giannini M. 2008. Effect of curing mode on the polymerization characteristics of dual-cured resin cement systems. J Dent 36:418-426.

6. Berger SB, Palialol AR, Cavalli V, Giannini M. 2009. Characterization of water sorption, solubility and filler particles of light-cured composite resins. Braz Dent J 20:314-318.

7. Chung KH. 1990. The relationship between composition and properties of posterior resin composites. J Dent Res 69:852-856.

8. De Menezes M, Arrais C, Giannini M. 2006. Influence of light-activated and auto- and dual-polymerizing adhesive systems on bond strength of indirect composite resin to dentin. J Prosthet Dent 96:115-121.

9. De Munck J, Vargas M, Van Landuyt K, Hikita K, Lambrechts P, Van Meerbeek B. 2004. Bonding of an auto-adhesive luting material to enamel and dentin. Dent Mater 20:963-971.

10. Duarte S, Botta A, Meire M, Sadan A. 2008. Microtensile bond strengths and scanning electron microscopic evaluation of self-adhesive and self-etch resin cements to intact and etched enamel. J Prosthet Dent 100:203-210.

11. Gerth H, Dammaschke T, Zuchner H, Schafer E. 2006. Chemical analysis and bonding reaction of RelyX Unicem and Bifix composites—A comparative study. Dent Mater 22:934-941.

12. Hikita K, Van Meerbeek B, Demunck J, Ikeda T, Van Landuyt K, Maida T, Lambrechts, P; Peumans, M. 2007. Bonding effectiveness of adhesive luting agents to enamel and dentin. Dent Mater 23:71-80.
13. Kim K, Ong J, Okuno O. 2002. The effect of filler loading and morphology on the mechanical properties of contemporary composites. J Prosthet Dent 87:642-649.
14. Leprince J, Palin WM, Mullier T, Devaux J, Vreven J, Leloup G. 2010. Investigating filler morphology and mechanical properties of new low-shrinkage resin composite types. J Oral Rehabil 37:364-376.
15. Mukai Y, Ten Cate J. 2002. Remineralization of advanced root dentin lesions in vitro. Caries Res 36:275-280.
16. Polydorou O, König A, Hellwig E, Kümmerer K. 2009. Long-term release of monomers from modern dental-composite materials. Eur J Oral Sci 117:68-75.
17. Sabbagh J, Ryelandt L, Bachérius L, Biebuyck J, Vreven J, Lambrechts P, Leloup G. 2004. Characterization of the inorganic fraction of resin composites. J Oral Rehabil 31:1090-1101.
18. Shinkai K, Suzuki S, Katoh Y. 2001. Effect of filler size on wear resistance of resin cement. Odontology 89:41-44.
19. Tanoue N, Koishi Y, Atsuta M, Matsumura H. 2003. Properties of dual-curable luting composites polymerized with single and dual curing modes. J Oral Rehabil 30:1015-1021.
20. Tenuta L, Zamataro C, Del Bel Cury A, Tabchoury C, Cury J. 2009. Mechanism of fluoride dentifrice effect on enamel demineralization. Caries Research 43:278-285.
21. Viotti RG, Kasaz A, Pena AE, Alexandre RS, Arrais CAG, Reis AF. 2009. Microtensile bond strength of new self-adhesive agents and conventional mutlstep systems. J Prosthet Dent 102:306-312.
22. Weiner R. 2007. Dental Cements: a review and update. Gen Dent 55:357-364.

Figure legends:

Figure 1. Elements identified by energy dispersive X-ray spectroscopy microanalysis for RelyX ARC resin cement (A), Clearfil Esthetic Cement (B), RelyX Unicem (C) and Clearfil SA Luting (D).

Figure 2. SEM micrograph of RelyX ARC resin cement (original magnification x1.000 (A) and x3.000 (B).

Figure 3. SEM micrograph of Clearfil Esthetic Cement resin cement (original magnification x1.000 (A) and x3.000 (B).

Figure 4. SEM micrograph of RelyX Unicem resin cement (original magnification x1.000 (A) and x3.000 (B).

Figure 5. SEM micrograph of Clearfil SA resin cement (original magnification x1.000 (A) and x3.000 (B).

Table 1: Classification, composition, manufacturer and batch number of resin cements used in this study (Information Supplied by the Manufacturer).

Resin cement (manufacturer)	Classification	Composition (batch number)
RelyX ARC (3M ESPE, St. Paul, MN, USA)	Total-etch Resin Cement	Paste A: silane treated ceramic, TEGDMA, Bis-GMA, silane-treated silica, functionalized dimethacrylate polymer; 2-benzotriazolyl-4-methylphenol, 4-(dimethylamino)-benzeneethanol; Paste B: silane treated ceramic, TEGDMA, Bis-GMA, silane-treated silica, functionalized dimethacrylate polymer, 2-benzotriazolyl-4-methylphenol, benzoyl peroxide (GE9JG).
Clearfil Esthetic Cement (Kuraray Medical, Kurashiki, Japan)	Self-etching Resin Cement	Paste A & B: Bis-GMA, TEGDMA, hydrophobic aromatic dimethacryate, hydrophilic aliphatic dimethacrylate, silanated silica filler, silanated barium glass filler, colloidal silica, dl-camphorquinone, catalysts, accelerators, pigments (0008AA).
RelyX Unicem (3M ESPE, St. Paul, MN, USA)	Self-adhesive Resin Cement	Liquid: methacrylated phosphoric acid esters, TEGDMA, substituted dimathacrylate Powder: silanized glass powder, silane treated silica, calcium hydroxide, substituted pyrimidine, sodium persulfate (365945).
Clearfil SA Cement (Kuraray Medical, Kurashiki, Japan)	Self-adhesive Resin Cement	Paste A & B: Bis-GMA, sodium fluoride, TEGDMA, 10-MDP, hydrophobic aromatic dimethacryate, hydrophilic aliphatic dimethacrylate, silanated barium glass filler, silanated colloidal silica, dl-camphorquinone, initiators, catalysts, pigments, others (0004AB).

Abbreviations: TEGDMA: triethylene glycol dimethacrylate; Bis-GMA: bisphenol A diglycidyl ether methacrylate; 10-MDP: 10-methacryloyloxydecyl dihydrogen phosphate.

FIGURES:

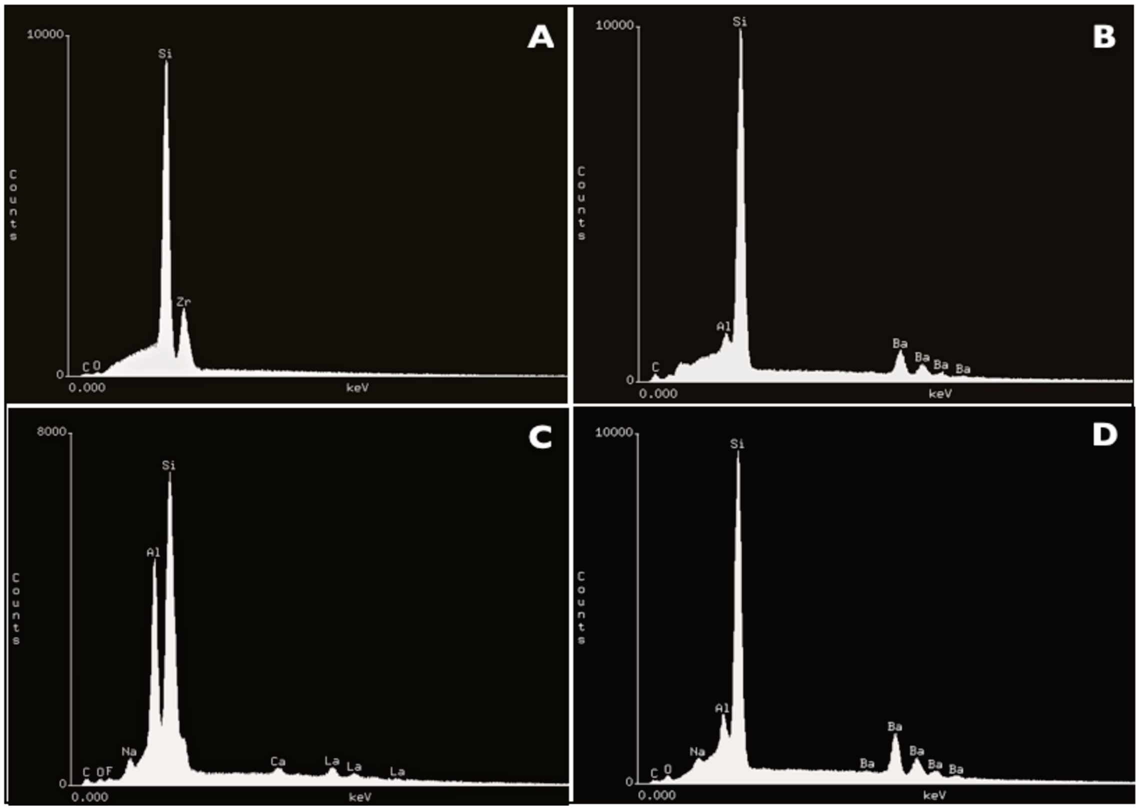

Figure 1. Elements identified by energy dispersive X-ray spectroscopy microanalysis for RelyX ARC resin cement (A), Clearfil Esthetic Cement (B), RelyX Unicem (C) and Clearfil SA Luting (D).

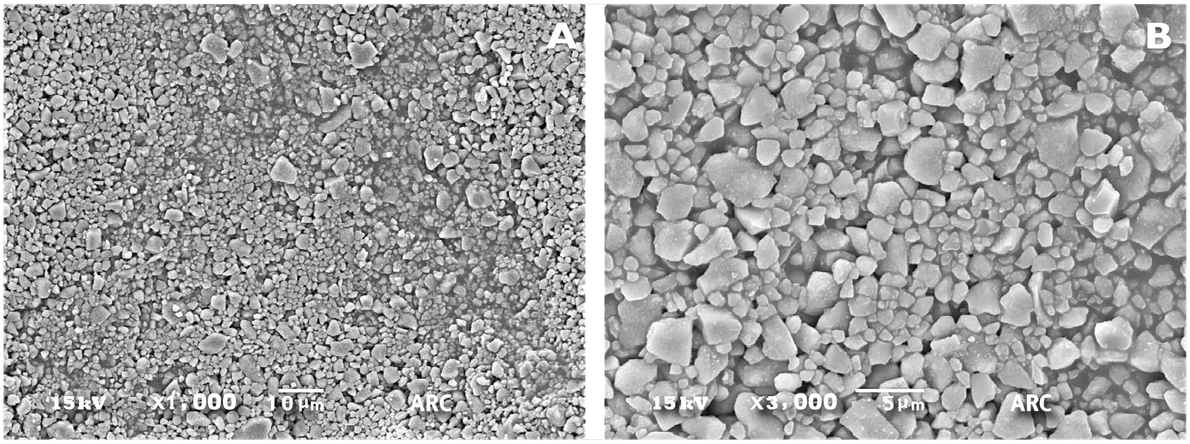

Figure 2. SEM micrograph of RelyX ARC resin cement (original magnification x1.000 (A) and x3.000 (B).

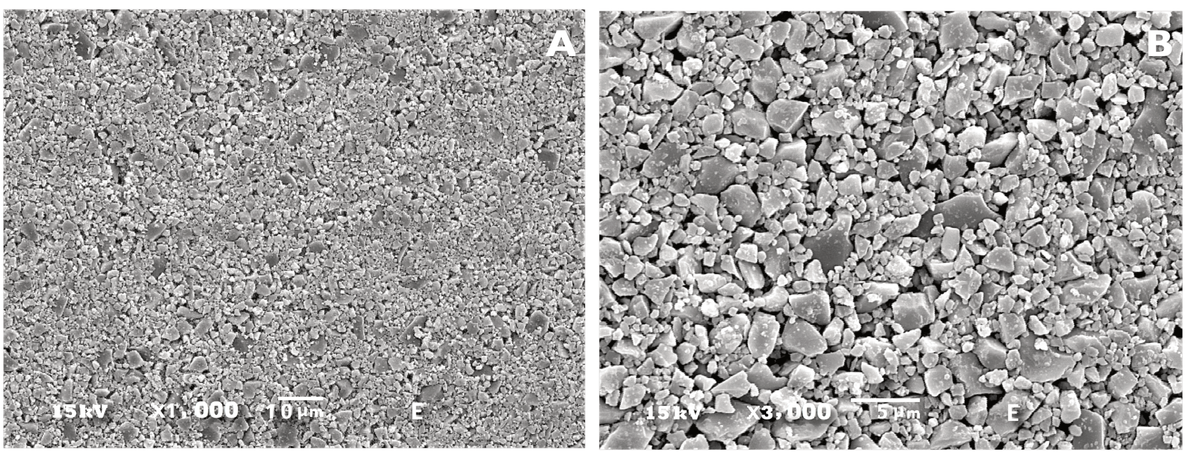

Figure 3. SEM micrograph of Clearfil Esthetic Cement resin cement (original magnification x1.000 (A) and x3.000 (B).

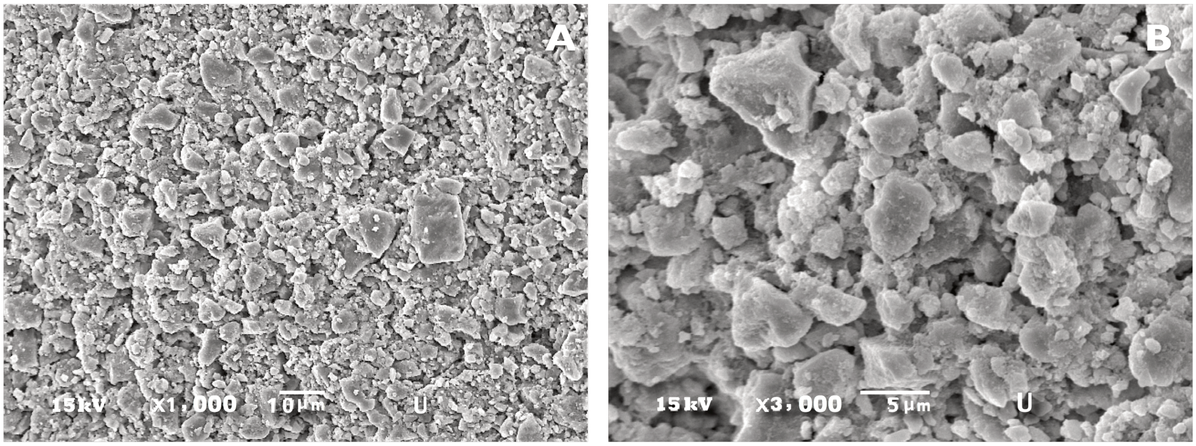

Figure 4. SEM micrograph of RelyX Unicem resin cement (original magnification x1.000 (A) and x3.000 (B).

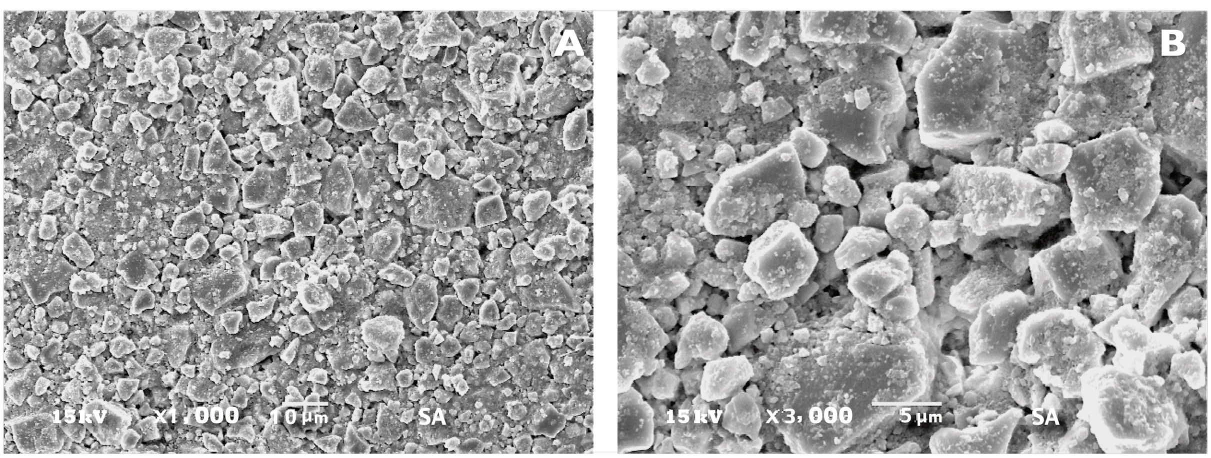

Figure 5. SEM micrograph of Clearfil SA resin cement (original magnification x1.000 (A) and x3.000 (B).

Micromorphology of resin-dentin interfaces using self-adhesive and conventional resin cements: a confocal laser and scanning electron microscope analysis

Short title: Morphological characteristics of resin cement-dentin interfaces

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KEY WORDS: resin cements, confocal laser scanning microscopy, SEM, hybrid layer, dentin.

ABSTRACT

Purpose: The aim of this study was to evaluate the resin-dentin morphology created by four dual-cured resin cements. Materials and Methods: Two self-adhesive resin cements (RelyX Unicem, 3M ESPE and Clearfil SA Luting, Kuraray Med.) and two conventional resin cementing systems (RelyX ARC, 3M ESPE and Clearfil Esthetic Cement, Kuraray Med.) were evaluated. Occlusal dentin surfaces of 32 extracted human third molars were flattened to expose coronal dentin. Teeth were assigned to 8 groups (n=4), according to resin cement products and microscope analysis (SEM: scanning electron microscope or CLSM: confocal laser scanning microscopy). For CLSM, two different fluorescent dyes, fluorescein isothiocyanate-dextran and rhodamine B, were incorporated into the adhesive system and resin cement, respectively. The resin cements were applied to indirect composite resin discs, which were cemented to dentin surface according to manufacturer's instructions. After 24h, all restored teeth were vertically sectioned into 1-mm-thick slabs for SEM or CLSM analyses. Results: RelyX ARC and Clearfil Esthetic Cement systems showed bonding agent and resin tags penetration into the dentin tubules. The hybrid layer formed by the Scotchbond Multi-Purpose Plus/RelyX ARC cementing system was thicker than that created by DC Bond/Clearfil Esthetic Cement. Also, short resin tags were observed when the DC Bond self-etch adhesive was used. No hybrid layer was detected for self-adhesive resin cements. Conclusion: Representative SEM and CLSM images provided resin-dentin interfaces variability between resin cements studied.

INTRODUCTION

Resin cements are the most indicated luting materials in the cementation of indirect composite resins and ceramic restorations to tooth structures. The increased demand of esthetic treatment with metal-free restorations and the evolution of bonding/adhesive techniques are responsible for the widespread use of resin cements. These luting systems can be classified according to the bonding strategies: self-adhesive resin cements, which do not require a bonding agent and the conventional resin cements, which are used after an adhesive application.[5,14,12,21,22]

The conventional cementing systems are used with etch-and-rinse adhesives or self-etching primers. For the etch-and-rinse technique, a 30 to 40% phosphoric acid conditioner demineralizes the dentin surface and totally removes the smear layer and smear plugs to allow the monomer infiltration into the intertubular dentin and dentin tubules, and create the hybrid layer.[19] The resin cements that combine self-etching primers application prior to luting procedure do not require the conditioning and rinsing steps, because the self-etching primers contain acidic monomers.[20] The etching aggressiveness of each self-etching adhesive depends on the type of acidic functional monomers, such as carboxyl or phosphate groups.[8,11] On the other hand, the self-adhesive resin cement has been recently development and did not require any dentin pretreatment.[8,10,21]

Some methodologies, such as confocal laser scanning microscopy (CLSM) and scanning electron microscopy (SEM) have been routinely used to evaluate interfacial structures and can be used to show the interaction between self-adhesive cements and dentin surface.[4,17] The CLSM provides more detailed information than SEM.[4] This method does not require elaborate specimen preparation, decreasing the risk of dehydration, shrinking and other artifacts, such as gap formation during coating or under SEM observation.[16] In addition, it allows researchers to capture images from the specimen subsurface, reducing the effects of surface contamination.[7]

The purpose of this study was to evaluate by SEM and CLSM the features of resin-dentin interfaces of indirect resin composite restorations created by self-adhesive resin cements or bonding agents combined with their respective dual-curing cementing systems. The tested hypothesis was that conventional and self-adhesive resin cements form different interfacial structures, regardless of the use of bonding agents for conventional resin cement.

MATERIALS AND METHODS

Specimen Preparation and Experimental Groups

This research protocol was approved by the Institutional Review Board of the Piracicaba School of Dentistry, Campinas State University (089/2009). Thirty-two freshly extracted, erupted, human third molars were stored in a saturated thymol solution for no longer than 3 months. The teeth were then transversally sectioned in the middle of the crown using a diamond blade saw (Buehler Ltd; Lake Bluff, IL) on an automated sectioning device (Isomet 2000; Buehler Ltd.) under water irrigation to expose areas of middle-depth dentin. The exposed dentin surfaces were wet polished by machine (APL-4, Arotec, Cotia, SP, Brazil) using 600-grit SiC paper to create a flat surface with standardized smear layer formation before application of bonding agents. The prepared teeth were then randomly assigned to four groups according to products, so four teeth were prepared for SEM analysis and four teeth were prepared for CLSM analysis.

Four commercial dual-curing resin cements were used in this study: two self-adhesive resin cements (RelyX Unicem, 3M ESPE, Seefeld, Germany and Clearfil SA Luting, Kuraray Medical Inc., Kurashiki, Japan) and two conventional resin cements (RelyX ARC, 3M ESPE, St. Paul, MN, USA and Clearfil Esthetic Cement, Kuraray Med.), which were combined with a three-step, etch-and-rinse adhesive (Adper Scotchbond Multi-Purpose Plus, 3M ESPE) and a single-step, self-etching adhesive (Clearfil DC Bond, Kuraray Med.), respectively. The composition of resin cements, classification, manufacturers, shade and lot number are described in Table 1.

Thirty-two pre-polymerized, light-cured composite resin discs with 2 mm thick and 10 mm in diameter (B2D shade, Sinfony; 3M ESPE) were prepared to simulate overlying laboratory-processed composite resin restorations.[1] The surface of each disc was airborne-particle abraded with 50 µm aluminum oxide (Danville Engineering, Danville, VA, USA) for 10 s (air pressure: 0.552 MPa; distance from the tip: 1.5 cm) and silanated using coupling agents according to manufacturer directions (RelyX Ceramic Primer, 3M ESPE or Clearfil Ceramic Primer, Kuraray Med.).

When RelyX ARC was used, Adper Scotchbond Multi-Purpose Plus was previously applied to the dentin surface. According to 3M ESPE manufacturer, dentin was etched with 37% phosphoric acid (Scothbond Etchant) for 15 s, followed by rinsing and surface moist control. Afterwards, the Activator, Primer, and Catalyst were consecutively applied to the etched dentin surface. For Clearfil DC Bond/Clearfil Esthetic Cement, equal amounts of DC Bond Liquid A & B were mixed for 10 s. The mixed solution was applied to the dentin surface with a microbrush and the mixture was left undisturbed for 20 s. After air blowing (10 s), the self-etching adhesive was light-cured for 20 s prior to resin cement application and the resin cement Clearfil Esthetic Cement were manipulated and applied in indirect restoration.

RelyX Unicem self-adhesive resin cement was applied in the resin disk after base and catalyst pastes were dispensed on the mixing pad and mixed during for 20 s. For Clearfil SA Luting, base and catalyst pastes were dispensed with the automix syringe and applied to the indirect resin composite. The self-adhesive resin cements do not require any dentin pre-treatment**.**

The mixed resin cements pastes were applied to the resin composite pre-polymerized disc, which was placed on the dentin surface with 500 g load, the excess was removed and the restoration was light-cured from their buccal and lingual aspects for 40 s (XL 3000; 3M ESPE, St. Paul, MN, USA). The output intensity of 580mW/cm^2 was used and was constantly measured with a radiometer Curing Radiometer, Model 100, Kerr Corp Orange, CA, USA). After removal of the load, additional 40 s of light exposure was performed also on mesial, distal and occlusal surfaces. The restored teeth were dark stored in relative humidity for 24 h at 37° C.

Scanning Electron Microscopy Analysis

To observe dentin interface by SEM, restored teeth were sectioned in the mesial-distal direction under water cooling into several 1.2-mm thick slabs with a slow-speed diamond saw (Isomet 1000, Buehler Ltd., Lake Bluff, IL, USA). Each slab was wet polished with 600-, 1200- and 2000-grit SiC paper. Afterwards, the specimens were polished using soft cloths and diamond pastes of decreasing abrasiveness (6, 3, 1 and ¼ µm). Slabs were etched with 37% phosphoric acid solution for 10 s to remove the mineral content, washed with water and immersed in 5% sodium hypochlorite for 5 min to remove exposed collagen from the dentin surface, respectively. Finally, specimens were subjected to ultrasonic bath with distillated water for 10 min and allowed to dry overnight at 37° C. Specimens were then sputter coated with gold (MED 010, Balzers Union, Balzers, Liechtenstein) and observed using a scanning electron microscope (VP 435, Leo, Cambridge, England) at x1.000 magnification.

Confocal Laser Scanning Microscope Analysis

For this analysis, two different fluorescent dyes, fluorescein isothiocyanate-dextran (Fluorescein-Isothiocyanate-Dextran, Sigma, St. Louis, MO, USA) and rhodamine B (Rhodamine B, Sigma), were incorporated into the adhesive systems and resin cements, respectively. Fluorescein was incorporated into adhesive systems tested (40 μg/mL).[3] This amount of fluorescein was added to each bottle of Adper Scotchbond Multi-Purpose Plus Adhesive (Activator, Primer and Catalyst) and to Clearfil DC Bond (Liquid A & B). The dye was mixed directly in the supplied bottle using a mixing device (Vortex Machine, Scientific Industries, New York, NY, USA) during 6 h to measure the complete dye dissolution. Rhodamine was added to the base resin cement paste of all resin cements and mixed to obtain a paste with uniform shade (0.32 μg/mg).[3]

The teeth were restored as previously described. Restored teeth were stored in vegetable oil for 24 h and were vertically sectioned under vegetable oil lubrification (Liza Pure Vegetable Oil, Cargill Agrícola S.A., Mairinque, SP, Brazil) into several 1.2-mm thick slabs with a slow-speed diamond saw (IsoMet 1110, Buehler Ltd.). Afterwards, slabs were wet polished with 600-, 1200- and 2000-grit SiC paper, were storage in vegetable oil and were analyzed under CLSM (LSM 510 Meta Confocal Microscope, Zeiss; Göttingen, Germany). An argon laser at 488 nm and He-Ne laser at 543 nm provided excitation energies, so images were obtained in dual fluorescence mode using a 25X objective and were analyzed by AxioVision LE software (Zeiss).

RESULTS

Representative SEM and CLSM images are shown in Figures 1 to 4. Once this study was based on a qualitative analysis, only visual differences among groups were considered and described as findings. For Adper Scotchbond Multi-Purpose Plus/RelyX ARC, hybrid layer and resin tag formation was clearly visible (Fig. 1d), while short resin tags were observed for Clearfil DC Bond self-etching system (Fig. 2d). In addition, the hybrid layer created by Adper Scotchbond Multi-Purpose Plus Adhesive/RelyX ARC (Fig. 1d) was thicker than that formed by Clearfil DC Bond/Clearfil Esthetic Cement (Fig. 2d). No dentin hybridization process and no resin tags inside the dentinal tubules were noted for self-adhesive resin cements. Also, the intimate and uniform contact was not seen between dentin and resin cement, because some bubbles or voids were presented at the interfaces (Figs. 3a and 4a).

CLSM images allowed visualization the adhesive system (Figs. 1a and 2a) and resin cements (Figs. 1b and 2b). The adhesive, which was labeled with fluorescein (Fig 1a and 2a), showed green fluorescence, while the resin cement, that was stained with rhodamine B, exhibited red fluorescence (Fig 1b, 2b, 3b and 4b). For the Adper Scotchbond Multi-Purpose Plus / RelyX ARC, it was not possible to distinguish the adhesive and resin cement layers, when the fluorescence dyes were added to the bonding agent and resin cement and when they were analyzed together (Fig 1c). The resin cement diffusion into adhesive layer and its mixture with labeled adhesive produced orange color fluorescence at dentin-resin cement bonded interface.

Clearfil DC Bond/Clearfil Esthetic Cement showed monomer difusion into the dentin surface (Figs. 2a e 2c) and into the resin cement layer (layer above) with green color predominance (Fig 2c). For this cementing system, it was possible to detect the bonding between adhesive and resin cement. A thin orange line can be seen, which corresponded to a overlap (Fig. 2c).

For self-adhesive resin cements, SEM and CLSM images showed the interaction between the dentin and the luting materials (Figs. 3 and 4). No diffusion of rhodamine (and resin cement) into the intertubular dentin or dentinal tubules were observed. Thus, the formation of hybrid layer and resin tags was not detected for RelyX Unicem (Figs. 3a and 3b) and Clearfil SA Luting (Figs. 4a and 4b).

DISCUSSION

Studies have focused in micromorphological analyses of bonded interfaces between tooth structures and adhesive systems[9,17,18,] or resin cements[3,4] to provide further information about the correct use of bonding agents and resin cements in order to improve bonding efficiency and durability. Fluorescent agents promote specific emission wavelength for each resinous component when they are excited by laser with specific wavelengths.[3] Rhodamine B is frequently used as fluorochrome for the analysis of bonded interfaces,[3,4,7,17] it is stable under various pH conditions, soluble in organic solutions[17] and displays a red-color characteristic.[7] On the other hand, fluorescein isothiocyanate-dextran was added to the adhesive composition emitted a green color.[3]

The type of bonding agent is determinant for the formation of interfacial structures. While self-etching etching adhesives promote the formation of a thin hybrid layer (0.5 to 2 microns), etch-and-rinse systems produced hybridization higher than 5 microns.[2,15] Representative SEM images of the Adper Scotchbond Multi-Purpose Plus Adhesive/RelyX ARC showed thicker hybrid layer and longer resin tags than those created by DC Bond/Clearfil Esthetic Cement. In general, this thick hybrid layer is equivalent to the demineralization depth promoted by phosphoric acid etching. On the other hand, the thickness of the interaction zone for self-etching primers depends on etching aggressiveness of acidic monomers from bonding agents within this category, which range from mild to strong.[20]

The orange layer observed in Figure 1c was created by the mixture of uncured bonding agent (Adper ScotchBond Multi-Purpose Plus) and resin cement (Rely X ARC). According to 3M ESPE manufacturer, adhesive components must be applied and left in the uncured state prior to the RelyX ARC application, consequentely the mixture of all agents occurs prior to light activation. As rhodamine penetrated into dentinal tubules, the cured of the mixture of these materials formed the resin tags. Also, some components of the resin cement seemed to form the hybrid layer, since the red fluorescence dye was presented in this interface area. Arrais et al. (2009)[3] also reported resin cement penetration within the hybrid layer and into the dentin tubules, depending on the category of cement system.

Conversely, the light-activation of Clearfil DC Bond self-etching bonding agent did not allow a massive penetration of the components of the Clearfil Esthetic Cement into dentin. However, the adhesive was able to infiltrate and blend with the resin cement, which is a function of this type of bonding agent. This mixture resulted in predominance of green fluorescence (Fig. 2c). The self-etching adhesive contains self-curing components or co-initiators, such as the dibenzoyl peroxide, which are important to increase the degree of conversion of resin cements.[6] A thin orange line represents the mixture between adhesive from oxygen-inhibited layer and resin cement (Fig. 2c).

In the current study, SEM and CLSM images showed only superficial interaction with dentin for self-adhesive resin cements. No diffusion of the rhodamine B dye into the dentin was observed using CLSM methodology. Also, no hybrid layer formation and resin tags were detected for RelyX Unicem (Figs. 3a and 3b) and Clearfil SA Luting (Figs. 4a and 4b).

Studies have suggested that RelyX Unicem show the best performance when used in wet dentin[13] and when a strong seating load of restoration is applied during the luting procedure.[8,11] However, the seating load used in this study (500 g) was not enough to improve superficial contact between dentin and resin cements, since some voids or bubbles were seen at the interface.

The bonding mechanism of self-adhesive resin cements to mineralized dental tissues is related to chemical reaction with hydroxyapatite. The Clearfil SA Luting contains 10-MDP monomer (10-Methacryloyloxydecyl dihydrogen phosphate), which has the ability to form strong ionic bond with calcium of enamel and dentin.[23] For RelyX Unicem, the bonding mechanism of self-adhesive resin cements involves phosphoric acidic methacrylates that react with the basic components, i.e., filler particles in the luting cement and hydroxyapatite in tooth tissue. After initial mixing, the cement is very acidic, however, the pH-value tends to increase according to the changes from hydrophilic to a hydrophobic features of resin cement.[12] Thus, the interfacial structure of resin cements depends on the approach of each luting material and can vary from micromechanical retention provided by hybridization to superficial interaction with chemical reaction between resin-based material and mineralized dental tissues. Therefore, the hypothesis tested in the current study must be rejected. In general, no hybridization was detected for self-adhesive resin cements, while conventional systems showed typical hybrid layer and resin tags formation.

ACNOWLEDGEMENTS

This study was supported by grants 09/51281-4 and 09/51674-6 from FAPESP, Brazil.

CLINICAL RELEVANCE

The SEM and CLSM images identified different interfacial structures for self-adhesive resin cements and conventional cementing systems. If the self-adhesive resin cements would produce efficiency interaction with the dentin, their clinical use could be widespread since they are user-friendly.

REFERENCES

1. Aguiar TR, Di Francescantonio M, Ambrosano GMB, Giannini M: Effect of curing mode on bond strength of self-adhesive resin luting cements to dentin. J Biomed Mater Res B Appl Biomater 2010; 93B:122-127.

2. Arrais CA, Giannini M: Morphology and thickness of the difussion of resin through demineralized or unconditioned dentinal matrix. Pesqui Odontol Bras 2002;16:115-120.

3. Arrais CAG, Miyake K, Rueggeberg FA, Pashley DH, Giannini M: Micromorphology of resin/dentin interfaces using 4th and 5h generation dual-curing adhesive/cement systems: a confocal laser scanning microscope analysis. J Adhes Dent 2009;11:15-26.

4. Bitter K, Paris S, Mueller J, Neumann K, Kieldassa AM: Correlation of scanning electron and confocal laser scanning microscopic analyses for visualization of dentin/adhesive interfaces in the root canal. J Adhes Dent 2009;11:7-14.

5. Cantoro A, Goracci C, Papacchini F, Mazzitelli C, Fadda GM, Ferrari M: Effect of pre-cure temperature on the bonding potential of self-etch and self-adhesive resin cements. Dent Mater 2008;24:577-583.

6. Cavalcanti SCSXB, Arais CAG, Oliveira MT, Giannini M: The effect of the presence and presentation mode of co-initiators on the microtensile bond strength of dual-cured adhesive systems used in indirect restorations. Oper Dent 2008;33:682-689.

7. D'Alpino PHP, Pereira JC, Svizero NR, Rueggeberg FA, Pashley DH: Factors affecting use of fluorescent agents in identification of resin-based polymers. J Adhes Dent 2006;8:285-292.

8. De Munck J, Vargas M, Van Landuyt K, Hikita K, Lambrechts P, Van Meerbeek B: Bonding of an auto-adhesive luting material to enamel and dentin. Dent Mater 2004;20:963-971.

9. de Oliveira MT, Arrais CA, Aranha AC, de Paula Eduardo C, Miyake K, Rueggeberg FA, Giannini M: Micromorphology of resin-dentin interfaces using one-bottle etch&rinse and self-etching adhesive systems on laser-treated dentin surfaces: a confocal laser scanning microscope analysis. Lasers Surg Med 2010;42:662-670.
10. Duarte S, Botta A, Meire M, Sadan A: Microtensile bond strengths and scanning electron microscopic evaluation of self-adhesive and self-etch resin cements to intact and etched enamel. J Prosthet Dent 2008; 100:203-210.
11. Goracci C, Cury AH, Cantoro A, Papacchini F, Tay FR, Ferrari M: Microtensile bond strength and interfacial properties of self-etching and self-adhesive resin cements used to lute composite onlays under different seating forces. J Adhes Dent 2006;8:327-335.
12. Holderegger C, Sailer I, Schuhmacher C, Schlapfer R, Hammerle C, Fischer J: Shear bond strength of resin cements to human dentin. Dent Mater 2008;24:944-950.
13. Mazzitelli C, Monticelli F, Osorio R, Casucci A, Toledano M, Ferrari M: Effect of simulated pulpal pressure on self-adhesive cements bonding to dentin. Dent Mater 2008;24:1156-1163.
14. Monticelli F, Osorio R, Mazzitelli C, Ferrari M, Toledano M: Limited decalcification/diffusion of self-adhesive cements into dentin. J Dent Res 2008;87:974-979.
15. Pashley DH, Ciucchi B, Sano H, Horner JA: Permeability of dentin to adhesive agents. Quintessence Int 1993;24:618-631.
16. Pioch T, D'Souza PD, H.J.Staehle, Duschner H: Resin-dentin interface studied by SEM & CLSM. Micros Anal 1996;42:15-16.
17. Pioch T, Stotz S, Staehle HJ, Duschner H: Applications of confocal laser scaning microscopy to dental bonding. Adv Dent Res 1997;11:453-461.
18. Reis AF, Bedran-de-Castro AK, Giannini M, Pereira PN: Interfacial ultramorphology of single-step adhesives: nanoleakage as a function of time. J Oral Rehabil 2007;34:213-221.
19. Shimada Y, Harnirattisai C, Inokoshi S, Burrow MF, Takatsu T: In vivo adhesive interface between resin and dentin. Oper Dent 1995;20:204-210.
20. Tay FR, Pashley DH: Aggressiveness of contemporary self-etching systems. I: Deph of penetration beyond dentin smear layers. Dent Mater 2001;17:296-308.

21. Viotti RG, Kasaz A, Pena AE, Alexandre RS, Arrais CAG, Reis AF: Microtensile bond strength of new self-adhesive agents and conventional mutlstep systems. J Prosthet Dent 2009;102:306-312.
22. Yang B, Ludwig K, Adelung R, Kern M: Micro-tensile bond strength of three luting resins to human regional dentin. Dent Mater 2006;22:45-56.
23. Yoshida Y, Nagakane K, Fukuda R, Nakayama Y, Okazaki M, Shintani H, Inoue S, Tagawa Y, Suzuki K, De Munck J et al: Comparative study on adhesive performance of funtional monomers. J Dent Res 2004;83:454-458.

Table 1: Cementing systems, manufacturer, classification, shade, compositions and batch number of resin systems used in this study (Information Supplied by the Manufacturer).

Cementing systems (manufacturer)	Classification	Shade	Composition (batch number)
RelyX ARC/ Adper™ Scotchbond™ Multi-Purpose Plus (3M ESPE, St. Paul, MN, USA)	Dual-cured resin cement/ 3-step etch-and-rise adhesive system	A1	Scotchbond Multi-Purpose Plus: Activator: ethyl alcohol, sodium benzenesulfinate (N123538); Primer: water, HEMA, copolymer of acrylic and itaconic acids (N124894); Catalyst: bis-GMA, HEMA, benzoyl peroxide (N130621); RelyX ARC Paste A: silane treated ceramic, TEGDMA, bis-GMA, silane-treated silica, functionalized dimethacrylate polymer; 2-benzotriazolyl-4-methylphenol, 4-(dimethylamino)-benzeneethanol; Paste B: silane treated ceramic, TEGDMA, bis-GMA, silane-treated silica, functionalized dimethacrylate polymer, 2-benzotriazolyl-4-methylphenol, benzoyl peroxide (N149151).
Clearfil Esthetic Cement/ Clearfil DC Bond (Kuraray Medical, Kurashiki, Japan)	Dual-cured resin cement/ 1-step self-etching adhesive system	Clear	Clearfil DC Bond: Liquid A: HEMA, bis-GMA, dibenzoyl peroxide, 10-MDP, colloidal silica, dl-camphorquinone, initiators, others (00271A); Liquid B: ethanol, water, accelerators, catalysts (00146B); Clearfil Esthetic Cement: Paste A & B: bis-GMA, TEGDMA, hydrophobic aromatic dimethacryate, hydrophilic aliphatic dimethacrylate, silanated silica filler, silanated barium glass filler, colloidal silica, dl-camphorquinone, catalysts, accelerators, pigments (0015AB).
RelyX Unicem (3M ESPE, St. Paul, MN, USA)	Dual-cured self-adhesive resin cement	A2	Base: glass power, methacrylated phosphoric esters, TEGDMA, silane-treated silica, sodium persulfate. Catalyst: glass power, dimethacrylate, silane-treated silica, TEGDMA, dental glass (394291)
Clearfil SA Cement (Kuraray Medical, Kurashiki, Japan)	Dual-cured self-adhesive resin cement	A2	Paste A & B: bis-GMA, sodium fluoride, TEGDMA, 10-MDP, hydrophobic aromatic dimethacryate, hydrophilic aliphatic dimethacrylate, silanated barium glass filler, silanated colloidal silica, dl-camphorquinone, initiators, catalysts, pigments, others (0005AB).

Abbreviations: HEMA: 2-hydroxyethyl methacrylate; TEGDMA: triethylene glycol dimethacrylate; bis-GMA: bisphenol A diglycidyl ether methacrylate; 10-MDP: 10-methacryloyloxydecyl dihydrogen phosphate.

Figures:

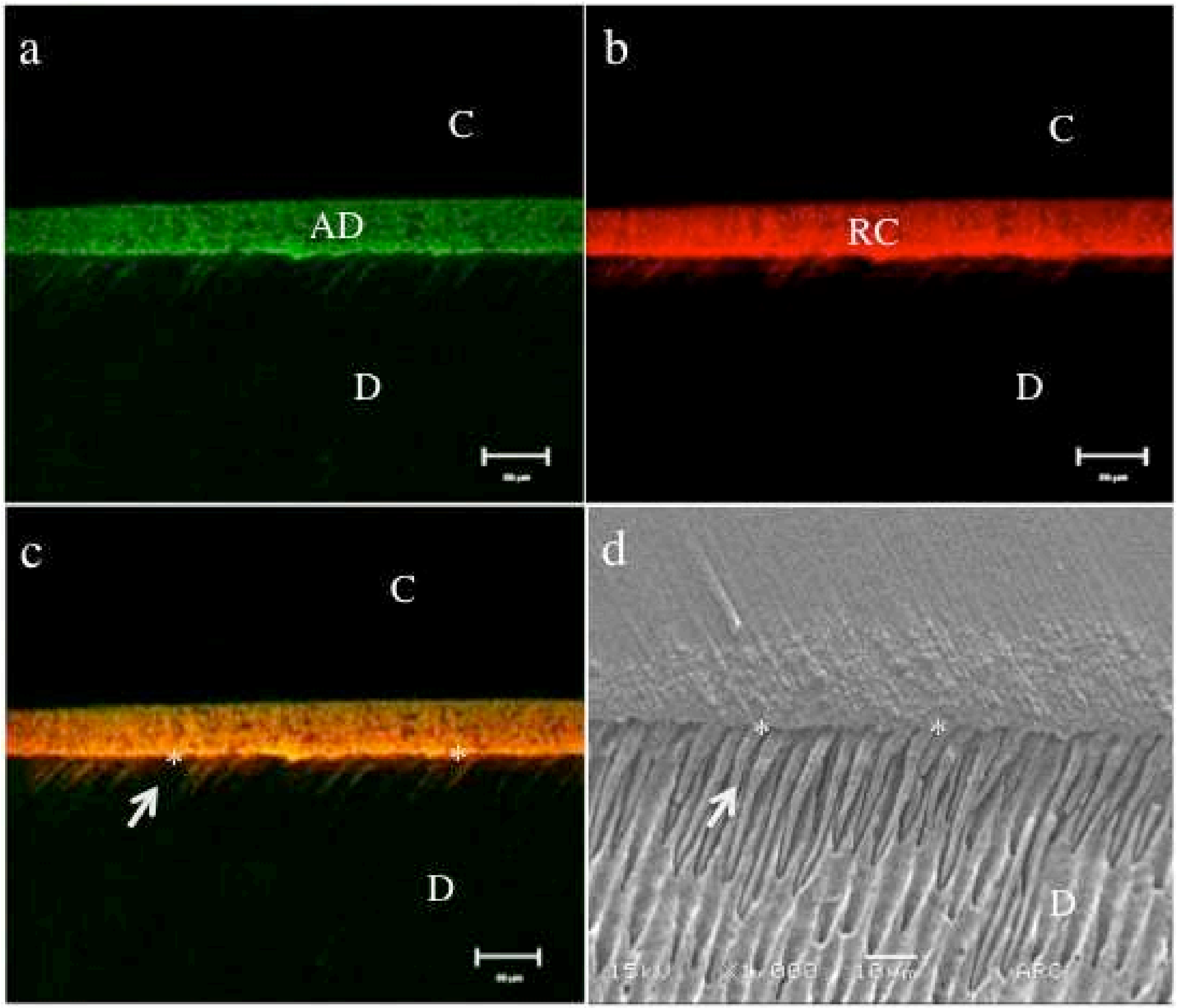

Figure 1. Bonding of indirect composite (C) to dentin (D) using Adper Scotchbond Multi-Purpose Plus adhesive system (AD) and RelyX ARC resin cement (RC). For CLSM images, the adhesive was labeled with fluorescein (Fig 1a) and the resin cement was stained with rhodamina B (Fig 1b), showing green and red fluorescence colors, respectively. The Figure 1c demonstrates an orange layer, which correponded to the mixture bewteen the adhesive (green) and resin cement (red), showing no distinction between adhesive and resin cement. The hybrid layer (*) and tag formation (arrows) can be clearly observed (Figs. 1c and 1d).

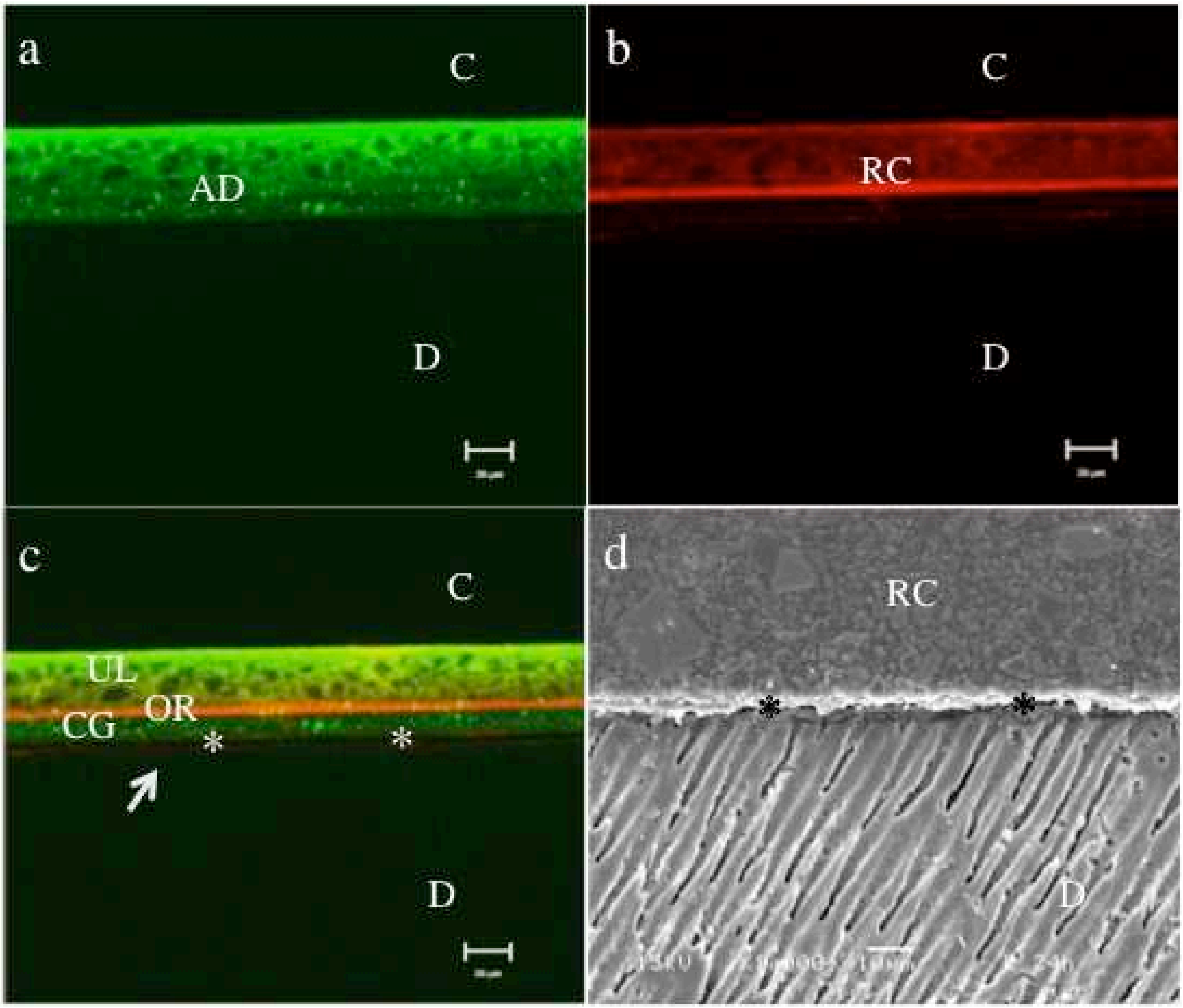

Figure 2. Bonding of indirect composite (C) to dentin (D) using Clearfil DC Bond / Clearfil Esthetic Cement (RC). For CLSM images, the adhesive was labeled with fluorescein (Fig. 2a) and the resin cement was stained with rhodamina B (Fig. 2b), showing green and red fluorescence color respectively. The Figure 2c and 2d demonstrates hybrid layer (*) and short resin tags (arrow) formation of the green-labeled adhesive. Also, it is possible to verify three layers, according to the green intensity: upper dark green layer (UL) correspond to the resin cement infiltrated by adhesive, meddium layer (orange line - OR) is the top of the polymerized adhesive (green) and red-labeled resin cement, and the clear green layer (CG) (botton) that is the self-etch in contact with dentin.

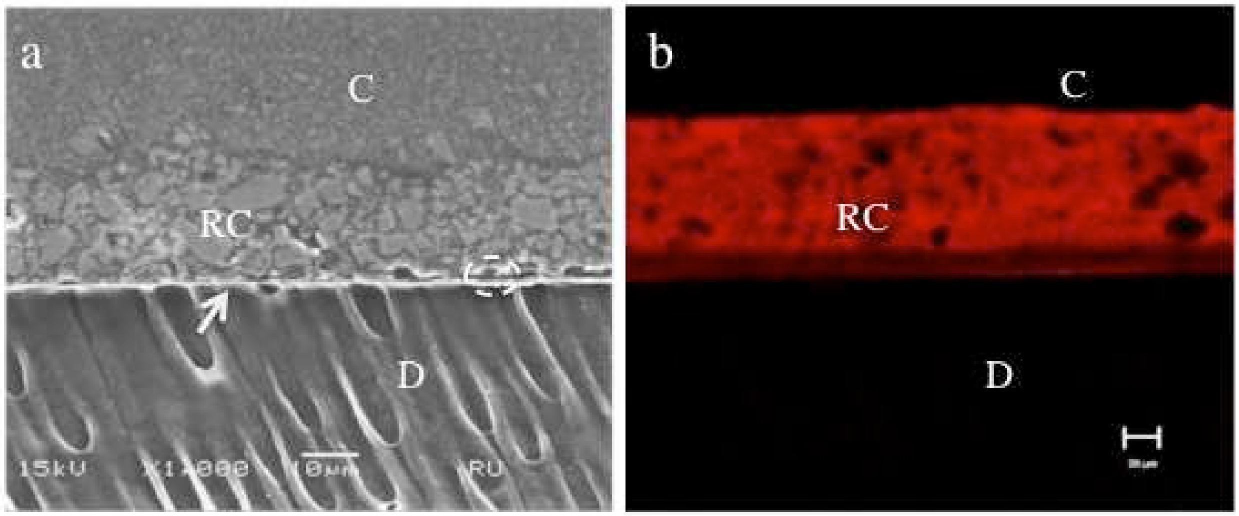

Figure 3. Bonding of indirect composite (C) to dentin (D) using RelyX Unicem self-adhesive resin cement (RC). No distinction hybrid layer and resin tags were detected. The arrows show the contact between the dentin and resin cement and the formation of voids (ring).

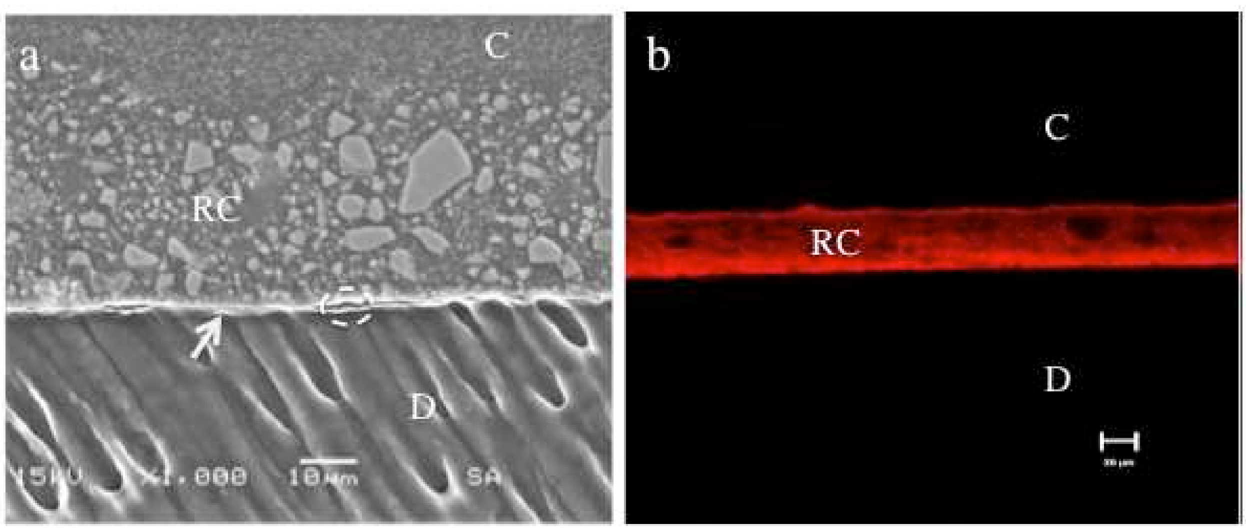

Fig 4. Bonding of indirect composite (C) to dentin (D) using Clearfil SA Luting resin cement (RC). No distinction hybrid layer and resin tags were detected. The arrows show the contact between the dentin and resin cement and the formation of voids (ring).

CAPÍTULO TRÊS

Effect of aging methods on dentin bond strength of conventional and self-adhesive resin luting cements

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Running title: Bond strength of cementing system

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Aguiar TR, André CB, Correr-Sobrinho L, Arrais CAG, Ambrosano GMB, Giannini M. Effect of storage time and mechanical load cycling on dentin Bond strength of conventional and self-adhesive resin luting cements. Eur J Oral Sci

Abstract: This study evaluated the tensile bond strength of conventional and self-adhesive dual-curing resin cements to dentin after artificial aging methods. Occlusal dentin surfaces of 96 extracted human third molars were flattened to expose coronal dentin. Twelve groups (n=8) were evaluated, according to resin cement products and artificial aging methods. Materials tested included two conventional (RelyX ARC/Adper Scotchbond Multi-Purpose Plus and Clearfil Esthetic Cement/DC Bond) and two self-adhesive resin cements (RelyX Unicem and Clearfil SA Cement). Resin cements were applied to pre-polymerized resin discs, which were subsequently bonded to the dentin surfaces. Restored teeth were divided into three treatments: 1- control groups (tested 24 h after specimen preparation); 2- mechanical load cycling; 3- storage in artificial saliva for one year. Bonded beams (1.0 mm^2) were obtained from these restored teeth and tested in tension (0.5 mm/min) until failure. Fracture modes were examined by SEM. Data was analyzed by two-way ANOVA and Tukey test (α= 0.05). The bond strength of all resin cements did not reduce after aging methods. Self-adhesive resin cements provide significantly higher bond strength than conventional luting materials. The Clearfil Esthetic Cement showed the lowest bond strength to dentin.

Key words: bond strength, resin cement, dental adhesive, dentin

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The use of self-adhesive, dual-polymerizing resin cements simplifies the adhesive cementation procedures. This type of resin cement does not require any pretreatment of tooth surface with bonding agent and the bonding mechanism of self-adhesive resin cements to mineralized dental tissues is related to chemical reaction between acid monomer or phosphoric acid ester with calcium of enamel and dentin (1). Conversely, conventional cementing systems involve a complex clinical protocol with multi-step applications and rather technique sensitive (2, 3).

Studies have compared the bond strength values of conventional and self-adhesive resin cements and the results have shown some advantages of self-adhesive resin cements (1, 4-8). The *in vitro* artificial aging methods have been suggested to simulate some challenges of restorative materials in the oral environment. The long-term storage of restored teeth, thermal or mechanical load cycling is often used to accelerate the degradation of resin-dentin interfaces and analyze the performance of restorative materials (8-10).

The purpose of this *in vitro* study was to evaluate the tensile bond strength of resin cements to dentin. The teeth were restored with conventional cementing systems or self-adhesive resin cements and tested following three treatments (1- control groups (tested 24 h after specimen preparation), 2- submitted to mechanical load cycling (MLC) and 3- storage in artificial saliva for one year). The research hypothesis tested was that bond strength values would be significantly lower when the restored teeth were stored for one year in artificial saliva or when subjected to MLC, regardless the type of resin cement.

Material and methods

Specimen Preparation and Experimental Groups

Ninety-six freshly extracted, erupted, non-carious human third molars were used. The research protocol was approved by the Institutional Review Board of the Piracicaba Dental School –State University of Campinas (089/2009). The teeth were stored in a saturated thymol solution at 5°C for no longer than 3 months. They were then transversally sectioned in the middle of the crown, using a diamond blade saw (Buehler Ltd, Lake Bluff, IL, USA) on an automated sectioning device (Isomet 2000, Buehler Ltd) under water irrigation, exposing areas of middle-depth dentin. The exposed dentin surfaces were wet polished by machine (APL-4, Arotec Ind. Com. Ltda, Cotia, SP, Brazil) using #600-grit SiC paper, under constant running water. The prepared teeth were then randomly assigned to 12 experimental groups (n=8), according to the treatment group (control group, storage in artificial saliva for one year and mechanical load cycling) and resin cement type.

Four dual-cured resin cements were tested: two self-adhesive resin cements (RelyX Unicem, 3M ESPE, Seefeld, Germany and Clearfil SA Cement, Kuraray Medical Inc., Kurashiki, Japan) and two conventional cementing systems, one combine a three-step etch-and-rinse adhesive (RelyX ARC/ Adper Scotchbond Multi-Purpose Plus, 3M ESPE, St. Paul, MN, USA) and one that uses an one-step self-etching adhesive (Clearfil Esthetic Cement/DC Bond, Kuraray Medical Inc). Chemical composition, manufacturer, classification and shade of the tested materials are listed in Table 1.

Bonding procedures

Ninety-six pre-polymerized, light-cured composite resin discs, 2 mm thick and 10 mm in diameter (B2D shade, Sinfony, 3M ESPE, St. Paul, MN, USA), were prepared to simulate overlying laboratory-processed composite resin restorations (6). The surface of each disc to be bonded to the prepared tooth was airborne-particle abraded with 50-µm aluminum oxide (Danville Engineering Inc, San Ramon, CA, USA) for 10 seconds (air pressure: 0.552 MPa; distance from the tip: 1.5 cm) (6). After that, each disc was silanated using coupling agents according to manufacturer directions. The same manufacturer of resin cements was used to avoid chemical bias (Ceramic Primer, 3M ESPE or Clearfil Ceramic Primer, Kuraray Medical Inc).

All cementing systems were manipulated and applied according to manufacturers' instructions. The tooth surface was prepared with adhesive systems (Adper Scotchbond Multi-Purpose Plus and Clearfil DC Bond) for conventional resin cements (RelyX ARC and Clearfil Esthetic Cement, respectively). Resin cement pastes were applied to the sandblasted surface of the pre-polymerized composite resin disc after which the disc was placed on the dentin surface. The excess was removed, a 500 g load was applied and the restoration was light-cured from their buccal, lingual and occlusal aspects for 40 seconds (XL 3000, 3M ESPE, St. Paul, MN, USA). The output intensity of 580mW/cm^2 was used. After removal of the load, additional 40 seconds irradiations were performed also the mesial and distal surfaces. Thereafter, 3-mm-thick block of autopolymerizing composite resin (Concise, 3M of Brazil, Sumaré, SP, Brazil) was then added to the untreated pre-polymerized composite resin surface to facilitate specimen gripping length while bond testing was performed.

Artificial Aging Methods and Microtensile Bond Strength Test

Following restorative procedures, the restored teeth were further divided into three subgroups according to artificial aging treatments:

1- control groups (specimens tested 24 h after cementation),

2- submitted to mechanical load cycling (MLC),

3- storage in artificial saliva for one year (at 37°C / artificial saliva changed every 15 days).

For MLC (group 2), root teeth were involved with polyether impression material (±0.2 mm) (Impregum Soft, 3M ESPE, St. Paul, MN, USA) to simulated periodontal ligament and were embedded in acrylic resin cylinder (JET, Campo Limpo Paulista, SP, Brazil). The cycling was performed in a Mechanical Loading Machine (Erios International, São Paulo, SP, Brazil). Restored teeth were subject to 50.000 cycles at an axial force of 80N at a frequency of 1.0 Hz. After MLC, the restored teeth were prepared for microtensile testing.

For the groups 1 and 3, restored teeth were vertically sectioned under running water into several 1.0-mm thick slabs with a slow-speed diamond saw (IsoMet 1110, Buehler Ltd, Lake Bluff, IL, USA) after 24 hs. Each slab was further sectioned perpendicularly to produce bonded sticks approximately 1.0 mm^2 in cross-section and the bonded sticks were tested or stored for one year in artificial saliva, respectively (group 1 and 3). The bonded surface area was calculated using a digital caliper (mod. 727-6/150, Starret Ind. e Com. Ltda., Itu, SP, Brazil). Each bonded stick was attached to the grips of a microtensile testing device with cyanoacrylate glue (Super Bonder, Henkel/Loctite, Diadema, SP, Brazil). The tensile testing was performed in a universal testing machine (EZ Test; Shimadzu Corp, Kyoto, Japan), at a crosshead speed of 0.5 mm/min until failure. Five beams were selected from each sample (restored tooth) and the average value (MPa) was calculated for teeth. Data were analyzed by two-way analysis of variance (ANOVA) (4 resin cements and 3 treatments) and Tukey post-hoc test (α =0.05).

Failure pattern analysis

The fractured specimens were carefully removed from the grips and the fracture modes examined by SEM. Fractured specimens were mounted on aluminum stubs and allowed to air-dry overnight at 37° C. After, samples were sputter coated with gold (MED 010, Bal-Tec AG, Balzers, Liechtenstein) and examined by a single individual using a scanning electron microscope (VP 435, Leo, Cambridge, England). Failure modes were classified into 1 of 6 types (6): Type 1 - cohesive failure in dentin; Type 2 - adhesive failure along the dentin surface; Type 3 - cohesive failure within adhesive layer for conventional cements; Type 4 - cohesive failure within the resin cement; Type 5 - adhesive failure along the pre-polymerized composite–resin cement interface, Type 6 - mixed failure. Representative areas of the failure patterns were photographed at X500 and X1000.

Results

Two-way ANOVA indicated that only resin cement factor ($p < 0.001$) significantly influenced tensile strength results. Summary statistics for the different experimental groups are shown in Table II. When looking at data with respect to differences in treatments, the bond strength of groups submitted to storage for one year and MLC were similar to control groups. Among the resin cements, Clearfil Esthetic Cement/DC Bond resulted in significantly reduction in bond strength to dentin in all simulations. Self-adhesive resin cements (RelyX Unicem and Clearfil SA Luting) yielded higher bond strength than conventional cementing systems (RelyX ARC and Clearfil Esthetic Cement) for all treatments of restored teeth.

Figure 1 shows the proportional prevalence (%) of the failure patterns in all experimental groups. Adhesive failures along the dentin surface were observed for all resin cements tested (Figure 2A, 2B and 2C). However, this classification was not the most predominant pattern for Clearfil Esthetic Cement after MLC and for RelyX Unicem and Clearfil SA Luting after one year of artificial saliva storage. Failure analysis of these cements showed high incidence of cohesive fracture within the resin cement. Mixed failure was noted for most of experimental groups, except for Clearfil Esthetic Cement submitted to MLC.

Discussion

The bond strength longevity of indirect adhesive procedures is influenced by the physic and mechanical properties of materials and the bonding effectiveness between luting agent-tooth (11). In addition, clinical factors such thermal and mechanical stress, malocclusion (12), saliva, dentinal fluid, acids beverages and organic biofilm (13) may affect the adhesive interface. According to BRESCHI *et al.* (14), the clinical longevity of adhesive procedures depends on physical and chemical factors; however; it is difficult to reproduce all these factors in *in-vitro* studies.

In this current study, artificial aging methods (storage for one year in artificial saliva and mechanical load cycling tests) did not significantly affect the bond strength performance of resin cements tested. It was expected that fatigue load stress would contribute to accelerate the bonding degradation between resin cement and dentin. Laboratory studies have suggested that fatigue stress can reduce the bond durability (12, 15, 16); however, after MLC, the restored teeth showed similar bond strength to untreated control group (12). According to NIKAIDO *et al.* (12), it is possible that the resin composite and the resin cement may function as a shock absorber, distributing the force on the dental structure and preserving the adhesive interface.

Little information is available in the dental literature regarding the clinical behavior of indirect restoration cemented with self-adhesive resin cements and the bond strength of these luting materials to dentin during and after MLC. NAUMANN *et al.* (8) analyzed the performance of resin cements when specimens were long-term stored or exposed to thermo/mechanical cycling. The authors reported that no significant differences were found between self-adhesive and the conventional resin luting cement. BOUILLAGUET *et al.* (17) and NIKAIDO *et al.* (12) indicated that cavity configuration factor and depth dentin affected the bond strength values after MLC. Other factor that can affect the results is the varied cycling parameters described in articles, according to cycle frequency, load and number of cycles used (15, 18).

The storage of bonded beams for one year in artificial saliva did not reduce the bond strength for resin cements. Studies have reported that smaller interfacial bonding area, such as, used in this study (less than 1.0 mm^2) and the peripheral enamel removal around the specimens can facilitate and accelerate the degradation process of dentin structure and resin-dentin interface (7, 19). Long-term *in vitro* studies involving adhesive (20) and cementing systems (21) reported that bond strength to dentin are brand-dependent. In this study, the storage of bonded beam specimens in artificial saliva for one year was not enough to promote any resin-dentin degradation detected by microtensile bond strength test. The research hypothesis that bond strength values will be significantly lower when the restored teeth were stored for one year in artificial saliva or when subjected to MLC, regardless the type of resin cement was rejected.

The self-adhesive resin cements (RelyX Unicem and Clearfil SA Luting) provided significantly higher bond strength to dentin than those conventional materials (Rely X ARC and Clearfil Esthetic Cement). Similarly, the bonding performance to dentin has been reported between RelyX Unicem and conventional resin cements after 24 hours (1, 4, 5, 22) and after aging methods (8). However, in the self-curing mode, the RelyX Unicem exhibited lower bond strength than other cements (3, 23) and it can be explained by the fact that this cement showed lower degree of conversion when it was not light-activated (24).

RelyX Unicem contains phosphoric acidic methacrylates, which react with basic fillers in the cement and the calcium ions of the hydroxyapatite from tooth tissue promoting the bonding to dentin and enamel (5). The 10-MDP monomer (10-Methacryloyloxydecyl dihydrogen phosphate) is the main component of Clearfil SA Luting. YOSHIDA *et al.* (25) showed that this acidic monomer has an ability to form strong ionic bond with calcium of enamel and dentin. Thus, the bonding mechanism of self-adhesive resin cements to dentin is based on chemical reactions between dentin and resin cement, while the bonding mechanism of conventional cementing system depends on the type of the bonding agent used in combination with this system.

RelyX ARC/Adper Scotchbond Multi-Purpose Plus cementing system showed bond strength values between 16.3 to 17.1 MPa that corroborated with ASMUSSEN & PEUTZFELDT (26) (18.0 MPa). This three-step etch-and-rinse adhesive promotes the hybrid layer formation, which is the bonding mechanism of this category of bonding agent. The Clearfil Esthetic Cement uses a one-step self-etching adhesive (Clearfil DC Bond) as bonding agent of this system. The adhesive also contain 10-MDP, however, it showed the lowest bond strength among the resin cements.

Regarding the failure mode, specimens failed predominantly adhesively along the dentin surface, which is in accordance with the results of other fractographic investigations (6, 23). RelyX Unicem and Cleafil SA Cement when storage for one year and Clearfil Esthetic Cement after MLC showed higher incidence of cohesive failure within the resin cement. The tubule orifices were opened and tags were not presented for the Rely X ARC resin cement (Fig. 2A). For the Clearfil Esthetic Cement, some tubules were not opened, showing remains of hybridized smear plugs. In contrast, SEM micrograph for self-adhesive resin cements showed that smear layer covered the dentin surface and the dentinal tubules were occluded by smear plugs (Fig. C). The failure patterns observed for resin cements seem to vary according to the bonding mechanism of each material tested.

The bond strength of restored teeth with indirect composite and resin cements did not reduce by storage in artificial saliva for one year or by mechanical load cycling. The self-adhesive resin cements showed higher bond strength than conventional cementing systems that use a bonding agent to bond the luting material to the tooth structure.

Acnowledgements

This study was supported by grants 09/51281-4 and 09/51674-6 from FAPESP, Brazil.

REFERENCES

1. DE MUNCK J, VARGAS M, VAN LANDUYT K, HIKITA K, LAMBRECHTS P, VAN MEERBEEK B. Bonding of an auto-adhesive luting material to enamel and dentin. *Dent Mater* 2004; **20**: 963-971.

2. MAK YF, LAI SC, CHEUNG GS, CHAN AW, TAY FR, PASHLEY DH. Micro-tensile bond testing of resin cements to dentin and an indirect resin composite. *Dent Mater* 2002; **18**: 609-621.

3. HOLDEREGGER C, SAILER I, SCUHMACHER C, SCHLAPFER R, HAMMERLE C, FISCHER J. Shear bond strength of resin cements to human dentin. *Dent Mater* 2008; **24**: 944-950.

4. ABO-HAMAR SE, HILLER KA, JUNG H, FEDERLIN M, FRIEDL KH, SCHMALZ G. Bond strength of a new universal self-adhesive resin luting cement to dentin and enamel. *Clin Oral Investig* 2005; **9**: 161-167.

5. HIKITA K, VAN MEERBEEK B, DE MUNCK J, IKEDA T, VAN LANDUYT K, MAIDA T, LAMBRECHTS P, PEUMANS M. Bonding effectiveness of adhesive luting agents to enamel and dentin. *Dent Mater* 2007; **23**: 71-80.

6. AGUIAR TR, DI FRANCESCANTONIO M, AMBROSANO GMB, GIANNINI M. Effect of curing mode on bond strength of self-adhesive resin luting cements to dentin. *J Biomed Mater Res B Appl Biomater* 2010; **93B**: 122-127.

7. DE MUNCK J, VAN LANDUYT K, PEUMANS M, POITEVIN A, LAMBRECHTS P, BRAEM M, VAN MEERBEEK B. A critical review of the durabiliy of adhesion to tooth tissue: methods and results. *J Dent Res* 2005; **84**: 118-132.

8. NAUMANN M, STERZENBACH G, ROSENTRITT M, BEUER F, FRANKENBERGER R. In vitro performance of self-adhesive resin cements for post-and-core build-ups: Influence of chewing simulation or 1-year storage in 0.5% chloramine solution. *Acta Biomater* 2010; **6**: 4389-4395.

9. SKOVRON L, KOGEO D, GORDILLO LA, MEIER MM, GOMES OM, REIS A, LOGUERCIO AD. Effects of immersion time and frequency of water exchange on durability of etch-and-rinse adhesive. *J Biomed Mater Res B Appl Biomater* 2010; **95**: 339-346.

10. SABÓIA V, SILVA F, NATO F, MAZZONI A, CADENARO M, MAZZOTTI G, GIANNINI M, BRESCHI L. Analysis of differential artificial ageing of the adhesive interface produced by a two-step etch-and-rinse adhesive. *Eur J Oral Sci* 2009; **117**: 618-624.

11. FURUKAWA K, INAI N. TAGAMI J. The effects of luting resin bond to dentin on the strength of dentin supported by indirect resin composite. *Dent Mater* 2002; **18**: 136-142.

12. NIKAIDO T, KUNZELMANN KH, CHEN H, OGATA M, HARADA N, YAMAGUCHI S, COX CF, HICKEL R, TAGAMI J. Evaluation of thermal cycling and mechanical loading on bond strength of a sef-etching primer system to dentin. *Dent Mater* 2002; **18**: 269-275.

13. TAY FR, PASHLEY DH. Have dentin adhesives become too hydrophilic? *J Can Dent Assoc* 2003; **69**: 726-731.

14. BRESCHI L, MAZZONI A, RUGGERI A, CADENARO M, DILERNARDA R, DESTEFANODORIGO E. Dental adhesion review: Aging and stability of the bonded interface. *Dent Mater* 2008; **24**: 90-101.

15. MITSUI FH, PERIS AR, CAVALCANTI AN, MARCHI GM, PIMENTA LA. Influence of termal and mechanical load cycling on microtensile bond strengths of total and self-etching adhesive systems. *Oper Dent* 2006; **31**: 240-247.

16. AGGARWAL V, LAGONI A, JAIN V, SHAH N. Effect of cyclic loading on marginal adaptation and bond strength in direct vs indirect class II MO composite restorations. *Oper Dent* 2008; **33**: 587-592.

17. BOUILLAGUET S, CIUCCHI B, JACOBY T, WATAHA JC, PASHLEY D. Bonding characteristics to dentin walls of class II cavities, in vitro. *Dent Mater* 2001;**17**: 316-321.

18. BEDRAN-DE-CASTRO AK, PEREIRA PN, PIMENTA LA, THOMPSON JY. Effect of thermal and mechanical load cycling on microtensle bond strength of a total-etch adhesive system. *Oper Dent* 2004; **29**: 1501-1556.

19. REIS AF, GIANNINI M, PEREIRA PN. Effects of a peripheral enamel bond on the long-term effectiveness of dentin bonding agents exposed to water in vitro. *J Biomed Mater Res B Appl Biomater* 2008; **85**: 10-17.

20. ERHARDT MC, SHINOHARA MS, BEDRAN-DE-CASTRO AK, AMARAL CM, PIMENTA LA. Effect of long-term water storage on etch-and-rinse and self-etching resin-dentin bond strengths. *Gen Dent* 2008; **56**: 372-377.

21. AGUIAR TR, CAVALCANTI AN, FONTES CM, MARCHI GM, MUNIZ L, MATHIAS P. 24 hours and 3-months bond strength between dual-cured resin cements and simplified adhesive systems. *Acta Odontol Latinoam* 2009; **22**: 171-176.

22. GORACCI C, CURY AH, CANTORO A, PAPACCHINI F, TAY FR, FERRARI M. Microtensile bond strength and interfacial properties of self-etching and self-adhesive resin cements used to lute composite onlays under different seating forces. *J Adhes Dent* 2006; **8**: 327-335.

23. YANG B, LUDWIG K, ADELUNG R, KERN M. Micro-tensile bond strength of three luting resins to human regional dentin. *Dent Mater* 2006; **22**: 45-56.

24. AGUIAR TR, FRANCESCANTONIO MD, ARRAIS CAG, AMBROSANO GMB, DAVANZO C, GIANNINI M. Influence of curing mode and time on degree of conversion of one conventional and two self-adhesive resin cements. *Oper Dent* 2010; **35**: 295-99.

25. YOSHIDA Y, NAGAKANE K, FUKUDA R, NAKAYAMA Y, OKAZAKI M, SHINTANI H, INOUE S, TAGAWA Y, SUZUKI K, DE MUNCK J, VAN MEERBEEK B. Comparative study on adhesive performance of funtional monomers. *J Dent Res* 2004; **83**: 454-458.

26. ASMUSSEN E, PEUTZELDT A. Bonding of dual-curing resin cements to dentin. *J Adhes Dent* 2006; **8**: 299-304.

TABLES

Table 1: Cementing systems, manufacturer, classification, shade, compositions and batch number of resin systems used in this study (Information Supplied by the Manufacturer).

Cementing systems (manufacturer)	Classification	Shade	Composition (batch number)
RelyX ARC/ Adper™ Scotchbond™ Multi-Purpose Plus (3M ESPE, St. Paul, MN, USA)	Dual-cured resin cement/ 3-step etch-and-rise adhesive system	A1	Scotchbond Multi-Purpose Plus: Primer: water, HEMA, copolymer of acrylic and itaconic acids (9CC); Activator: ethyl alcohol, sodium benzenesulfinate (9LB); Catalyst: bis-GMA, HEMA, benzoyl peroxide (9BF); RelyX ARC Paste A: silane treated ceramic, TEGDMA, bis-GMA, silane-treated silica, functionalized dimethacrylate polymer; 2-benzotriazolyl-4-methylphenol, 4-(dimethylamino)-benzeneethanol; Paste B: silane treated ceramic, TEGDMA, bis-GMA, silane-treated silica, functionalized dimethacrylate polymer, 2-benzotriazolyl-4-methylphenol, benzoyl peroxide (GE9JG).
Clearfil Esthetic Cement/ Clearfil DC Bond (Kuraray Medical, Kurashiki, Japan)	Dual-cured resin cement/ 1-step self-etching adhesive system	Clear	Clearfil DC Bond: Liquid A: HEMA, bis-GMA, dibenzoyl peroxide, 10-MDP, colloidal silica, dl-camphorquinone, initiators, others (00013A); Liquid B: ethanol, water, accelerators, catalysts (0009A); Clearfil Esthetic Cement: Paste A & B: bis-GMA, TEGDMA, hydrophobic aromatic dimethacryate, hydrophilic aliphatic dimethacrylate, silanated silica filler, silanated barium glass filler, colloidal silica, dl-camphorquinone, catalysts, accelerators, pigments (0008AA).
RelyX Unicem (3M ESPE, St. Paul, MN, USA)	Dual-cured self-adhesive resin cement	A2	Liquid: methacrylated phosphoric acid esters, TEGDMA, substituted dimathacrylate Powder: silanized glass powder, silane treated silica, calcium hydroxide, substituted pyrimidine, sodium persulfate (365945).
Clearfil SA Cement (Kuraray Medical, Kurashiki, Japan)	Dual-cured self-adhesive resin cement	A2	Paste A & B: bis-GMA, sodium fluoride, TEGDMA, 10-MDP, hydrophobic aromatic dimethacryate, hydrophilic aliphatic dimethacrylate, silanated barium glass filler, silanated colloidal silica, dl-camphorquinone, initiators, catalysts, pigments, others (0004AB).

Abbreviations: HEMA: 2-hydroxyethyl methacrylate; TEGDMA: triethylene glycol dimethacrylate; bis-GMA: bisphenol A diglycidyl ether methacrylate; 10-MDP: 10-methacryloyloxydecyl dihydrogen phosphate.

Table II. Summary statistics [mean (sd)] of tensile bond strength among test groups (in MPa).

Cementing Systems	Immediate (24 h)	Storage (1 year)	Mechanical Loading
RelyX ARC / Scotchbond Multi-Purpose Plus	16.3 (2.0) Ab	17.3 (5.2) Ab	17.1 (5.9) Ab
Clearfil Esthetic Cement / Clearfil DC Bond	12.6 (3.7) Ac	12.9 (3.8) Ac	13.3 (0.9) Ac
RelyX Unicem	18.3 (1.9) Aa	21.0 (3.8) Aa	21.3 (4.0) Aa
Clearfil SA Cement	19.9 (2.4) Aa	21.1 (2.3) Aa	20.8 (4.3) Aa

Groups having similar letters (upper case: row; lower case: column) are not significantly different.

FIGURES

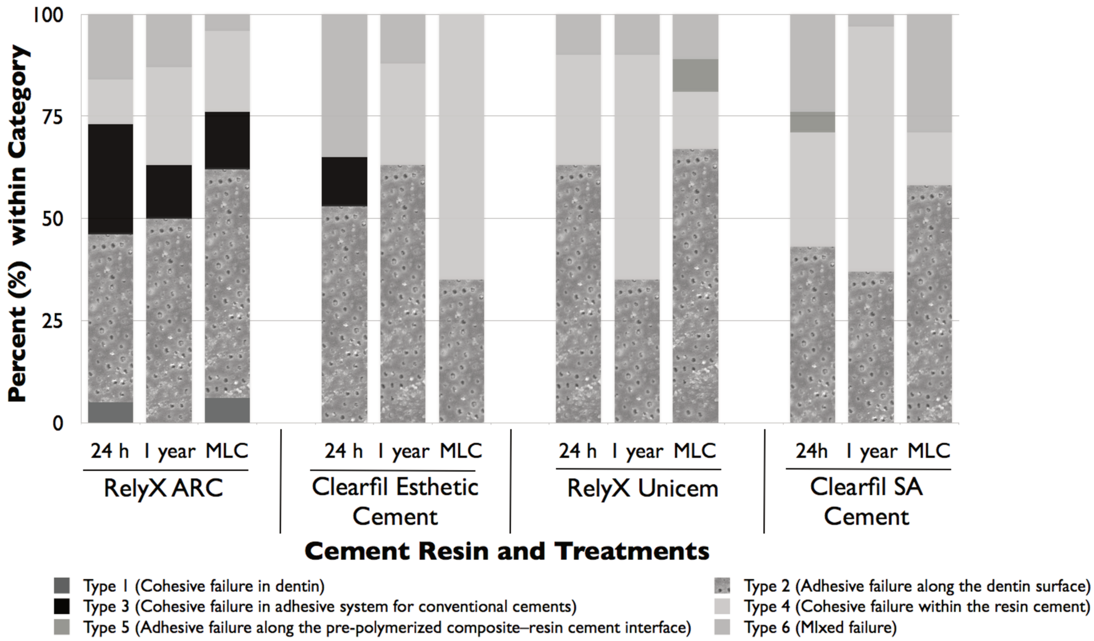

Figure 1. Failure modes of experimental groups.

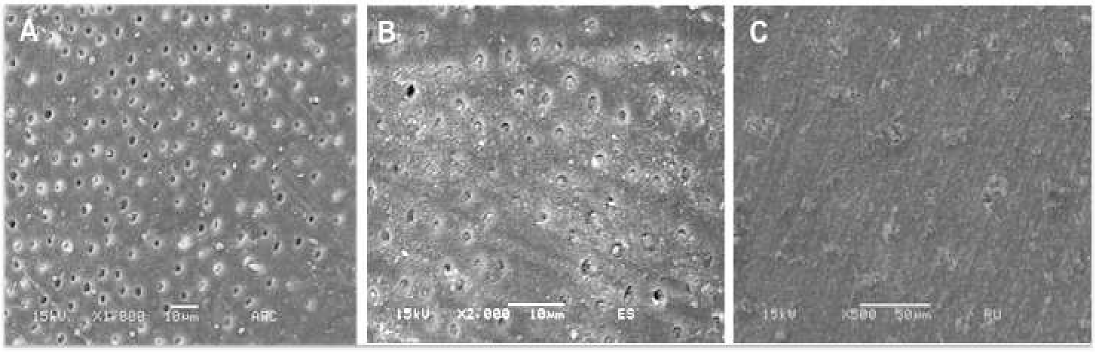

Figure 2. SEM micrographs showing adhesive failures along the dentin surface (type 2): (A) RelyX ARC/ Scotchbond Multi-Purpose Plus (original magnification X1000). The smear plugs seems to be removed and most of dentin tubules were opened; (B) Clearfil Esthetic Cement/DC bond (original magnification X1000). The dentin surface bonded with self-etching adhesive system showed some smear debris remaining inside the tubules. (C) Self-adhesive resin cement (RelyX Unicem) (original magnification X500). The dentin surface seemed cover by smear layer and the tubule orifices occluded with smear plugs.

CONSIDERAÇÕES GERAIS

Os cimentos resinosos autoadesivos foram disponibilizados no mercado odontológico com o intuito de simplificar a técnica de cimentação adesiva, promovendo a redução no número de passos presentes no protocolo clínico. Informações detalhadas quanto às propriedades mecânicas, o mecanismo e a durabilidade da união dos cimentos resinosos autoadesivos representam um tópico relevante e atual da Odontologia Restaurada, principalmente quando comparados aos cimentos resinosos convencionais (De Munck *et al*., 2004; Hikita *et al*., 2007). Assim, o presente estudo avaliou 4 cimentos resinosos (2 autoadesivos e 2 convencionais) quanto à composição inorgânica e às características morfológicas das partículas de carga, utilizando MEV/EDX; analisou a ultramorfologia da interface de união dentina/cimento resinoso empregando o microscópio confocal de varredura laser e a microscopia eletrônica de varredura. Adicionalmente, foi avaliado o efeito do armazenamento por 1 ano ou da ciclagem mecânica na resistência de união dentina-cimento resinoso.

Na avaliação das partículas de carga buscou-se identificar os principais componentes inorgânicos e caracterizar as partículas de carga presente nos cimentos resinosos. Para Han *et al*., (2007), a concentração e o tamanho das partículas de carga exercem influência nas propriedades mecânicas dos materiais. No presente estudo, o cimento resinoso convencional RelyX ARC apresentou partículas de carga esféricas e irregulares, enquanto os outros materiais demonstraram somente partículas com formato irregular. Essas partículas apresentam-se com diferentes dimensões, o que proporciona uma melhor compactação entre as mesmas, influenciando positivamente suas propriedades mecânicas. Para os cimentos resinosos autoadesivos, partículas com maior dimensão foram observadas, entretanto, ainda não se sabe como esta característica pode interferir nas propriedades físicas e mecânicas quando comparado aos cimentos convencionais.

Na avaliação em MEV/EDX, elevado conteúdo de sílico foi identificado em todos os materiais estudados (convencionais e autoadesivos). A sílica promove características radiopacas para esses materiais, o que facilita a identificação dos cimentos resinosos em exames radiográficos e evita possíveis dúvidas quanto ao diagnóstico da doença cárie. Com a mesma finalidade, os elementos zircônio e bário foram identificados no cimento RelyX ARC e nos produtos fabricados pela Kuraray Medical Inc. (Clearfil Esthetic Cement e Clearfil SA Esthetic). Segundo o fabricante, o cimento autoadesivo RelyX Unicem apresenta características semelhantes ao cimento de ionômero de vidro. Concordando com o exposto, este estudo demonstrou a presença de alumínio e fluoreto na composição do RelyX Unicem, o que pode estar relacionado com as partículas de vidro de flúor-alumínio-silicato presentes na porção ionomérica deste material.

Os cimentos resinosos avaliados além de apresentarem partículas de carga com diferentes composições inorgânicas, demonstraram distintas formas de interagir com o tecido dentinário. Concordando com outros estudos (De Munck *et al*., 2004; Goracci *et al*., 2006; Cantoro *et al*., 2008; Yang *et al*., 2006; Monticelli *et al*.,2008), os cimentos resinosos autoadesivos não formam *tags* resinosos e nem camada híbrida, notando-se uma área de íntimo contato na interface de união entre os cimentos autoadesivos e a superfície dentinária. Segundo De Munck *et al*. (2004), a limitada capacidade de difusão desses materiais está relacionada com a alta viscosidade dos mesmos. Assim, a aplicação de pressão durante o assentamento da peça protética visa proporcionar o aumento da capacidade de molhamento dos cimentos autoadesivos, gerando melhor contato com a superfície e melhor adaptação da peça ao tecido dentinário (De Munck *et al*., 2004; Goracci *et al*., 2006).

Similarmente ao estudo realizado por Aguiar *et al*. (2010), foi aplicado uma carga de 500 g após o assentamento da restauração indireta. Entretanto, a aplicação dessa pressão parece não ser suficiente para evitar a presença de bolhas na área de união entre o cimento resinoso autoadesivo e a dentina como demonstrado nas imagens em MEV. Por outro lado, a hibridização do tecido dentinário foi observada para os sistemas convencionais (RelyX ARC e Clearfil Esthetic Cement). Para o cimento RelyX ARC, que foi associado ao sistema adesivo *etch-and-rinse* de 3 passos (condicionamento ácido, *primer* e *bond*), notou-se *tags* resinosos longos e em maior quantidade. O uso do condicionamento ácido promove a desmineralização do tecido dental para posterior infiltração da resina hidrófila e fluida (Nishiyama *et al*., 1996; Hashimoto *et al*., 2000), entretanto, o cimento Clearfil Esthetic Cement por preconizar o uso do sistema adesivo autocondicionante de um único passo, permite que a infiltração do agente de união e a desmineralização do substrato dental ocorra ao mesmo tempo. Apesar das distintas formas de interagir com a dentina, estudos têm questionado a resistência e a estabilidade da união dos novos sistemas de cimentação adesiva (autoadesivos e autocondicionantes).

O presente estudo também avaliou a resistência da união imediata e após o envelhecimento das amostras em dentina. Para tal, as restaurações indiretas foram submetidas à ciclagem mecânica ou seccionadas em forma de "palitos" e armazenadas em saliva artificial durante 1 ano. A ciclagem mecânica possibilitou a aplicação de uma força oclusal sobre a restauração, visando simular a presença de forças mastigatórias e promover a degradação da união restauração indireta-cimento resinoso (Nikaido *et al*., 2002; Mitsui *et al*., 2006; Aggarwal *et al*., 2008). Por outro lado, o armazenamento das amostras em forma de "palitos" foi realizado com o intuito de promover maior contato da interface adesiva com a solução de armazenamento, o que teoricamente permite uma maior degradação hidrolítica e consequente redução dos valores de resistência de união.

Discordando do exposto, os sistemas de cimentação estudados resistiram aos desafios propostos (ciclagem mecânica e armazenamento) e não demonstraram diferença estatística quando comparados com o grupo controle. Pressupõe-se que o tempo de armazenamento e a quantidade de ciclos utilizados no estudo não tenha sido suficiente para promover o envelhecimento das amostras e gerar diferenças no comportamento adesivo desses cimentos. Em geral, os cimentos resinosos avaliados apresentaram características inorgânicas e mecanismos da união distintos, entretanto, os cimentos autoadesivos apresentaram valores estatisticamente superiores de resistência de união à dentina, quando comparado aos convencionais.

CONCLUSÃO

A partir dos resultados obtidos nos três capítulos observa-se que os cimentos resinosos analisados apresentaram composições distintas e diferentes modos de estabelecer união à dentina. Em cada capítulo pode-se concluir que:

1- Os cimentos resinosos apresentaram composições inorgânicas diferentes, com exceção do elemento silício. A morfologia das partículas de carga também variou de acordo com o material, sendo os cimentos autoadesivos àqueles que apresentaram as maiores partículas de carga;

2- Os cimentos convencionais utilizados com agentes de união formam camada híbrida e *tags* resinosos, enquanto, os autoadesivos apresentam apenas interação na superfície dentinária;

3- Para os cimentos resinosos estudados, o armazenamento das restaurações em saliva artificial por 1 ano ou a ciclagem mecânica não influenciaram a resistência de união à dentina. Entretanto, os cimentos resinosos autoadesivos apresentaram valores superiores de resistência de união, quando comparado aos sistemas convencionais.

REFERÊNCIAS*

1. Braga RR, Ballester RY, Carrilho MRO. Pilot study on the early shear strength of porcelain-dentin bonding using dual-cure cements. J Prosthet Dent. 1999; 81: 285-289.
2. De Moraes RR, Marinmon JL, Schneider LF, Sinhoreti MA, Correr-Sobrinho L, Bueno M. Effects of 6 months of aging in water on hardness and surface roughness of two microhybrid dental composites. J Prosth 2008; 17(4): 323-6.
3. De Paula AB, Duque C, Correr-Sobrinho L, Puppin-Rontani RM. Effect of restorative technique and thermal/mechanical treatment on marginal adaptation and compressive strength of esthetic restorations. Oper Dent 2008; 33:434-40.
4. Hashimoto M, Ohno H, Endo K, Kaga M, Sano H, Oguchi H. The effect of hybrid layer thickness on bond strength: demineralized dentin zone of the hybrid layer. Dent Mater. 2000; 16: 406-411.
5. Lodovici E, Reis A, Geraldeli S, Ferracane JL, Ballester RY, Filho LER. Does adhesive thickness affect resin-dentin bond strength after thermal/load cycling? Oper Dent 2009; 34: 58-64.
6. Nishiyama N, Suzuki K, Asakura T, Nakai H, Yasuda S, Nemoto K. The effects of pH on N-methacryloyl glycine primer on bond strength to acid-etched dentin. J Biomed Mater Res. 1996; 31: 379-784.
7. Rosenstiel SF, Land MF, Crispin BJ. Dental luting agents: A review of the current literature. J Prosthet Dent. 1998; 20: 280-301.
8. Uceda-Gómez N, Loguercio AD, Moura SK, Grande RH, Oda M, Reis A. Long-term Bond strength of adhesive systems applied to etched and deproteinized dentin. J Appl Oral Sci. 2007; 15: 475-479.

* De acordo com a norma da UNICAMP/FOP, baseada na norma do International Committee of Medical Journal Editors – Grupo de Vancouver. Abreviatura dos periódicos em conformidade com Medline.

9. Yamauti M, Hashimoto M, Sano H, Ohno H, Carvalho RM, Kaga M, Oguchi H, Kubota M. Degradation of resin-dentin bonds using NaOCl storage. Dent Mater, 2003; 19: 399-405.

ANEXOS

ANEXO 1- Certificado do Comitê de Ética em Pesquisa, FOP-UNICAMP

COMITÊ DE ÉTICA EM PESQUISA
FACULDADE DE ODONTOLOGIA DE PIRACICABA
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CERTIFICADO

O Comitê de Ética em Pesquisa da FOP-UNICAMP certifica que o projeto de pesquisa **"Estudo dos cimentos resinosos convencionais e autoadesivos: Ultramorfologia e resistência de união à dentina imediata e a longo prazo"**, protocolo nº 089/2009, dos pesquisadores Marcelo Giannini e Thaiane Rodrigues Aguiar, satisfaz as exigências do Conselho Nacional de Saúde - Ministério da Saúde para as pesquisas em seres humanos e foi aprovado por este comitê em 08/07/2009.

The Ethics Committee in Research of the School of Dentistry of Piracicaba - State University of Campinas, certify that the project **"Study of conventional and self-adhesive resin luting cement: Ultramorphology and bond strength to immediate and long-term dentin"**, register number 089/2009, of Marcelo Giannini and Thaiane Rodrigues Aguiar, comply with the recommendations of the National Health Council - Ministry of Health of Brazil for research in human subjects and therefore was approved by this committee at .

Prof. Dr. Pablo Agustin Vargas
Secretário
CEP/FOP/UNICAMP

Prof. Dr. Jacks Jorge Junior
Coordenador
CEP/FOP/UNICAMP

Nota: O título do protocolo aparece como fornecido pelos pesquisadores, sem qualquer edição.
Notice: The title of the project appears as provided by the authors, without editing.

Anexo 2

Preview

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Subject: European Journal of Oral Sciences - EOS-5176-OA-11

Body: @@date to be populated upon sending@@

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