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ORIGINAL ARTICLE

Influence of the curing mode on the degree of conversion of a dual-cured self-adhesive resin luting cement beneath ceramic

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Abstract

Objective. To evaluate the effect of the delayed photoactivation and ceramic barrier on the degree of conversion (DC) of self-adhesive resin cement. ***Materials and methods.*** Circular specimens (5 mm in diameter × 1 mm in thickness) of the RelyX U-100 resin cement were made using the following curing protocols ($n = 10$): (G1) 40 s beneath a IPS Empress II ceramic; (G2) 40 s of direct photocuring; (G3) 80 s beneath the ceramic; (G4) 80 s of direct photocuring; (G5) self-curing; (G6) 5 min in the absence of light (self-curing) followed by transceramic photocuring for 40 s; (G7) 5 min in the absence of light (self-curing) followed by transceramic photocuring for 80 s. All the specimens were photoactivated by LED (800 mW/cm^2). After 24 h of dry storage, the DC was measured by FTIR, on the top surface of the specimens. Data were submitted to one-way ANOVA and Tukey test ($p \leq 0.05$). ***Results.*** Direct photocuring with no ceramic interposition, regardless of the curing time (40 s and 80 s) promoted the highest conversion mean (56.79 ± 1.19 and 59.98 ± 2.93, respectively) and the 5 min delay time for the transceramic photocuring presented a similar mean compared to the immediate transceramic photocuring. The DC was influenced by the ceramic barrier, decreasing the conversion values (49.72 ± 1.91 for 40 s and 52.36 ± 2.50 for 80 s), with no statistical difference from the groups with the previous 5 min of photoactivation delay. The self-cure only showed the worst DC values. ***Conclusion.*** Direct photocuring provided a higher degree of conversion for the self-adhesive resin cement. The delayed light activation did not influence the degree of conversion for the resin cement tested.

Key Words: *resin cement, conversion degree, ceramic, photocuring*

Introduction

Dual-cured resin luting cements are widely used in clinical practice, due to their ability to bond indirect restorations to enamel and dentin [1,2]. When using the resin cement for dental restorations, some adverse effects from direct resin composite are minimized, such as polymerization shrinkage stress magnitude of the composite and marginal and internal gap formation at the tooth/restoration interface [3,4]. Moreover, a satisfactory monomer conversion of the resin luting cement is needed in order to guarantee an adequate bond strength and mechanical properties [5,6], since indirect restorations attenuate the light that passes through and reaches the luting material [2,5]. The reduction of light intensity beneath the indirect restoration is caused by the scattering, reflecting and absorbing properties of the ceramic or indirect resin [7] and it can affect the degree of conversion (DC). Thus, many problems are generated by an insufficient degree of conversion, such as deficient physical and mechanical properties of the resin luting cement [6,8,9] and consequently increasing second caries susceptibility [4], which can jeopardize the durability of the indirect restoration.

In an attempt to overcome the light attenuation promoted by the indirect restoration, dual-cured resin

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cements were developed, presenting characteristics of auto- and light-curing components, which can provide an adequate cure in places with reduced light penetration [7]. Also, the self-adhesive resin cements, a new modality of dual-cured luting material, present acidic monomers in their composition that are claimed to have a chemical interaction with the basic inorganic fillers [10]. Thus, these cements have an additional acid-base setting reaction in addition to the free radical polymerization of the material [10].

Moreover, choosing the correct photocuring approach for the luting process of dual-cured resin cements needs attention, since the physical-mechanical properties of the polymer matrix depend on the adequate degree of monomer conversion. In this sense, a delay period before photocuring, which can allow an initial chemical cure, was shown to reduce shrinkage stress and rate of polymerization, without compromising the final degree of conversion of dual-cured resin cements, when compared to the immediate photocuring [11]. Also, Faria-e-Silva et al. [12] found that the delay photoactivation approach (DPA) promoted similar bond strength values when compared to the immediate photoactivation. However, little information is known about the DC of dual-cured self-adhesive resin cements when cured with the delay photoactivation approach under a ceramic restoration. Furthermore, due to the attenuation promoted by the indirect materials, some authors [13,14] have suggested an extended photoactivation time to guarantee an adequate monomer conversion of the resin cement. It is claimed that when increasing the energy dose the DC also increases, since more free radicals are generated by the photoinitiator system. Notwithstanding this, it is important to investigate whether the over-exposure can compensate for the attenuation of light that reaches the resin cement.

Thus, the aim of this study was to evaluate the effect of the delayed photoactivation approach and ceramic barrier on the conversion degree of dual-cured self-adhesive resin cement. The tested hypotheses are that (1) a delayed (5 min) photoactivation approach would increase the DC of the resin cement and (2) the extended transceramic photoactivation time would improve the DC of the resin cement.

Materials and methods

The self-adhesive luting cement RelyX U-100 (3M ESPE, St Paul, MN) was tested.

The composition of the resin cement used is presented in Table I. Seventy disk-shaped specimens (n = 10, 5 mm in diameter and 1 mm thick) were made using the RelyX U-100 (U100). The resin cement was mixed and inserted in a silicon mold (Express XT, 3M ESPE, St Paul, MN) and then covered with a mylar strip and a glass slide, in order to form a plane surface. A disk (10 mm in diameter and 2 mm thick) of a lithium disilicate reinforced ceramic (IPS Empress II, Ivoclar Vivadent, Schaan, Liechtenstein) shade A2 was prepared to simulate the indirect restoration.

Table I. The resin cement used in this study with composition and manufacturer's information.

Resin cement	Composition	Manufacturer
RelyX U-100 (Batch #270644)	*Base*: glass fibers, methacrylated phosphoric acid esters, dimethacrylates, silanated silica, sodium persulfate, photoinitiators, self-cure initiators. *Catalyst*: glass fibers, dimethacrylates, photoinitiators, self-cure initiators, silanated silica, p-toluene sodium sulfate, calcium hydroxide.	3M ESPE, St Paul, MN

Equal amounts of base and catalyst pastes (27 g) were mixed for 15 s and then cured by the following methods in the groups: (G1) 40 s beneath ceramic; (G2) 40 s of direct photocuring; (G3) 80 s of photocuring beneath ceramic; (G4) 80 s of direct photocuring; (G5) self-curing; (G6) 5 min in the absence of light (self-curing) followed by transceramic photocuring for 40 s; and (G7) 5 min in the absence of light (self-curing) followed by transceramic photocuring for 80 s. All the groups were photocured by a single-peak light-emitting diode (LED FlashLite 1401, 800 mW/cm^2, Discus Dental, Culver City, CA). The irradiance of the curing source was measured with a radiometer before light activation (Demetron, Kerr Corporation, Orange, CA). The light spectrum of the LED unit is shown in Figure 1. With the interposition of the lithium disilicate ceramic, the irradiance decreased, reaching 430 mW/cm^2.

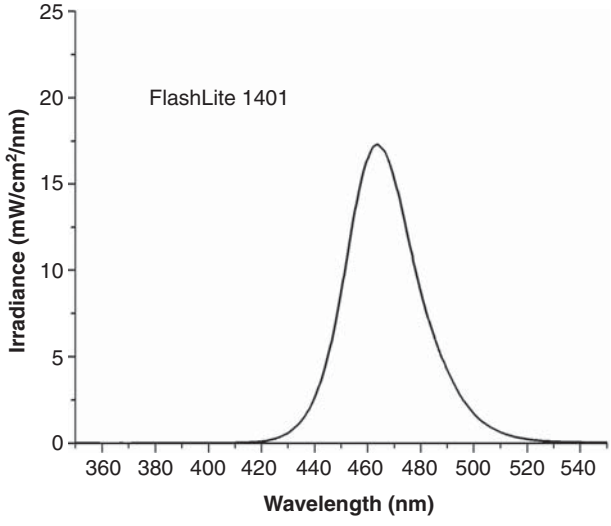

Figure 1. Spectrum of the light curing unit used in this study.

Degree of conversion

After 24 h of dry storage at 37°C, the degree of conversion (DC) was measured by Fourier Transformed Infrared Spectroscopy (FTIR) with an attenuated total reflectance (ATR) device (Spectrum 100, PerkinElmer, Shelton, CT) at 24°C and dry storage. Specimens were placed on the surface of a zinc selenide pellet (PerkinElmer) and the absorption spectra of non-polymerized and polymerized adhesive resins were obtained from the region between 1800–1500 cm^{-1}, with 32 scans at 4 cm^{-1}. Since the resin cement presented aromatic vinyl bonds of bisphenol and aliphatic bonds of the methacrylate functional group, the aliphatic carbon-to-carbon double-bond absorbance peak intensity (located at 1638 cm^{-1}) and that of the aromatic component (located at 1608 cm^{-1}; reference peak) were obtained. The degree of conversion was measured with the following equation: DC (%) = 100 × [1 − (R polymerized/R non-polymerized)], where R represents the ratio between the absorbance peak at 1638 cm^{-1} and 1608 cm^{-1}.

Data were submitted to one-way ANOVA and post-hoc Tukey's test, with a pre-set alpha of 0.05.

Results

The results of the one-way ANOVA showed statistical difference among the groups ($p \leq 0.001$). Comparisons according to the Tukey's test are presented in Table II. For the direct light curing, using either 40 s or 80 s, the resin cement showed the highest degree of conversion among the groups. The self-cure alone exhibited the lowest monomer conversion values compared to all tested groups. There was no difference of the DC values for the 40 s/80 s immediate or delayed transceramic curing protocols. The extended transceramic photoactivation time did not improve the monomer conversion for RelyX U-100.

Table II. Means (standard deviations, SD) of the degree of conversion for the tested groups.

Photocuring protocol	Degree of conversion (%)
40 s direct photocuring	56.79 (1.19) A
80 s direct photocuring	59.98 (2.93) A
40 s transceramic	49.72 (1.91) B
80 s transceramic	52.36 (2.50) B
5 min self-cure + 40 s transceramic photocuring	48.89 (2.48) B
5 min self-cure + 80 s transceramic photocuring	49.84 (3.29) B
Self-cure only	43.93 (2.03) C

Different letters mean statistical differences.

Discussion

This study used the self-etching, self-adhesive resin cement RelyX U-100, which is similar to RelyX Unicem, previously launched on the market. The main advantage of this material over other self-etching, self-adhesive resin cements is that the phosphate groups of the functionalized monomer included in the monomer mixture are claimed to react with the hydroxyapatite of the tooth substrate, resulting in additional retention through chemical bonding [10]. Indeed, a chemical interaction between the cement and the Ca^{+} of hydroxyapatite has been reported [15]. Thus, efforts should be made to improve physical properties of RelyX U-100, especially the DC. Optimal monomer conversion of resin cements is crucial to assure adequate physical properties and durable clinical performance of the indirect ceramic restoration on the tooth [14].

It was thought that immediate exposure to light would lead to rapid formation of a cross-linked polymer, causing entrapment of the reactive species such as activators and initiators needed for the pos-polymerization self-cure reaction, decreasing the DC of the resin cement [16]. In this sense, a delay period between cement mixing and light activation would increase total free-radical concentration, which would lead to a higher overall DC. However, the delayed photoactivation approach (DPA) did not improve the DC of U-100, regardless of the photoactivation time, rejecting the first hypothesis tested.

In fact, the ceramic beneath the curing-light tip promotes light attenuation, decreasing the irradiance that reaches the resin cement' surface in the transceramic photoactivation modes. Since more cross-linked polymer networks have been observed when higher irradiance reaches the surface of the resin-based material [17], it can be speculated that immediate transceramic photoactivation was not capable of forming a highly cross-linked polymer in the present investigation, favoring similar DC to those provided by DPA mode. Moreover, the lack of differences for the DC between immediate and delayed photoactivation modes might be attributed to the continued chemical curing occurred during 24 h storage, which could have increased the DC of samples immediately photoactivated. If the DC measurement had been performed after 5 or 10 min after mixing, the immediate photocuring might have promoted higher monomer conversion, compared to the groups with delayed photoactivation approach.

Despite the fact that DPA did not improve the DC of the resin cement tested in this study, it might provide reduced shrinkage stress, as observed elsewhere for the other conventional resin cement RelyX ARC [11], and might yield long-lasting indirect restorations. Further studies are necessary to confirm this assumption. Also, it is probable that the highest

DC of the resin cement has been obtained quickly after the minimal energy dose tested in this study for direct photoactivation (32 J/cm^2 – provided by 40 s) in comparison with the highest energy dose tested for direct photoactivation (64 J/cm^2 – provided by 80 s). That is, there were no statistically significant differences among the specimens photoactivated directly for 40 s and 80 s, rejecting the second hypothesis tested in this study. Nevertheless, the absence of an indirect restorative material is not clinically possible, so this condition was evaluated in this study only as a control condition.

The efficacy of light over-exposing on the DC of resin cement photoactivated through an indirect ceramic restoration was tested, which would be clinically suitable. However, as well as for the direct photoactivation, the extended photoactivation time of 80 s did not improve the 24 h DC of the resin cement. Although the irradiance decreased by the ceramic restoration, it is also likely that the minimal energy dose provided by 40 s indirect photoactivation (17 J/cm^2) was sufficient to provide the highest 24 h DC in comparison with that provided by 80 s indirect photoactivation (34 J/cm^2). In fact, it has been demonstrated that the DC of a resin cement irradiated using 20 J/cm^2 and 30 J/cm^2 energy doses did not show statistically significant differences [18]. Also, another explanation for the lack of differences among the samples, directly or indirectly photoactivated for 40 s and 80 s, is that the DC was evaluated 24 h after the photoactivation, allowing for a chemical cure in the absence of light [19]. That is, the observation of any statistically significant differences among the groups that used the photoactivation for 40 s and 80 s would be noted only if the evaluation of the DC had been made even after photoactivation.

Regardless of the photoactivation time, a direct photoactivation method allows high light irradiance to reach the resin cement, since there is no intervening material between the light and the resin cement. This was responsible for the highest DC of the specimens that were directly photocured. Also, the results concerning the low DC of the resin cement tested for the self-curing mode in comparison with the dual-cured modes are in accordance to the findings obtained elsewhere [9,10,20].

The self-etching, self-adhesive resin cement tested contains acidic monomers in its composition. Acidic monomers have been shown to negatively affect the DC of dual-cured materials, especially in the self-curing mode of polymerization, since they seem to interact chemically with the amine initiator which is contained in the polymer matrix. The changes in viscosity during the auto-polymerization reaction may reduce the ability of radicals to migrate and continue the conversion reaction [21]. Moreover, the slow polymerization promoted by the self-curing mode might have been impaired by water produced during the neutralization reaction of phosphate monomers with basic filler within the cement [9]. Thus, proprietary activator/initiator systems should be included in the composition of self-adhesive resin cements to overcome chemical incompatibility with acidic monomers [10]. Moreover, a lower DC could not be avoided in the self-curing mode. This implies that clinicians should avoid the complete absence of light during the cementation of fixed prosthesis using the self-etching, self-adhesive resin cement tested.

The method used for the assessment of the DC was Fourier Transformed Infrared Spectroscopy (FTIR), which is a well-established technique in the relevant literature [7,10,22,23]. However, taking into account that this is an *in vitro* study, further clinical trials should be performed to evaluate the longevity of indirect ceramic restorations cemented using the curing modes tested in the present investigation.

In conclusion, direct photocuring yielded a higher degree of conversion for the self-adhesive resin cement. The lithium disilicate reinforced ceramic promoted light attenuation, decreasing monomer conversion, and the extended transceramic photoactivation time did not exhibit improvements on the conversion values. The delay photoactivation approach can be clinically used since it promotes a similar degree of conversion compared to the conventional continuous photoactivation procedure.

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