Optical effects of graphene addition on adhesives for orthodontic lingual retainers

Received: 17 September 2023 | Accepted: 18 November 2023

DOI: 10.1111/eos.12966

European Journal of Oral Sciences | NOF Scandinavian Division IADR | WILEY

ORIGINAL ARTICLE

Optical effects of graphene addition on adhesives for orthodontic lingual retainers

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Abstract

The objective of this study was to determine the effects on the colour of adding increasing concentrations of graphene to orthodontic fixed retainer adhesives and to evaluate changes in optical transmission during light curing and the resultant degree of conversion. Two different types of adhesives commonly used for fixed retainers were investigated: A packable composite (Transbond) and a flowable composite (Transbond Supreme). Graphene was added to the adhesives in three different concentrations (0.01, 0.05, and 0.1 wt%). Adhesives without graphene addition were set as control groups. A Minolta colourimeter was used to measure the colour and translucency parameters. Irradiance transmitted during curing was quantified using MARC Light Collector. Fourier-transform infrared spectroscopy was used to record degree of conversion. Data were statistically analysed with the Student's *t*-test and one-way ANOVA with Tukey's tests ($\alpha = 0.05$). The findings showed that incorporating graphene darkened the adhesive colour significantly and reduced translucency. As the graphene concentration reached 0.1 wt%, samples became opaque; yet, no adverse effect on degree of conversion was observed. The addition of graphene reduces optical transmission of lingual retainer adhesives; the effect increases with graphene concentration.

KEYWORDS
bonding, colour, degree of conversion, Orthodontic adhesives

INTRODUCTION

Maintaining the teeth in an optical aesthetic and functional position after treatment is important in orthodontics [1]. Fixed lingual retainers were introduced to prevent relapse and are widely used [2–4]. Typically, lingual retainers are attached to the teeth with resin-based composites applied around the retainer as adhesives [5]. The adhesives have an important role in avoiding the failure of retainers, which is a major concern for patients and orthodontists. According to in vitro studies, detachment between the retainer and composite is the most frequent type of retainer failure [6–9]. Thus, improving the properties of composites is important to avoid the risk of retainer failure with the possibility of relapse.

Depending on the flowability of the material, resin-based composites can be divided into packable and flowable

materials. The first type is usually highly filled and reported to have better mechanical properties and durability [10], while the other is easier to handle but demonstrates a lower bond strength [5, 11, 12]. It has been shown that adding some types of inorganic fillers, especially nanoparticles can increase both the mechanical properties and biological behaviour of composites [13]. Recently, graphene and its derivatives have been applied in dental materials as they are able to bring significant improvement in properties with small addition [14, 15]. Some studies have shown that graphene family materials can be used to improve both mechanical and antimicrobial properties of dental resin composites, thus improving the survival of the composites in the complex environment of the oral cavity [15–17]. Meanwhile, graphene family materials tend to be cytocompatible when used as fillers in biomedical materials [18].

However, considering the black colour of graphene, its application in dentistry is greatly restricted. Researchers try to find the optimal graphene concentration that can bring improvement in mechanical performance and have minimal aesthetic effects [19]. In addition to the colour effect, addition of different materials to resin matrices could affect the curing under exposure to visible light. In composites, the organic phase has a lower refractive index than the inorganic phase. The greater the mismatch, the more pronounced the effect on light scattering, according to Monte Carlo theory [20, 21]. In general, the mismatch is greater for unpolymerised monomers as the degree of conversion increases, the refractive index of the organic matrix increases and the mismatch with the filler decreases [22]. Given the refractive index of graphene is usually higher than organic matrix [23, 24], the refractive index of fillers increases, and it can be predicted that the addition of graphene will have an effect on the overall refractive index.

To understand the effect of graphene on the optical properties and the degree of conversion of materials, it is necessary to understand how light passes through composites. On the surface of the composites, part of the light is reflected, while the other part penetrates to activate the photosensitizer to start the polymerization. During the penetration, the energy is absorbed and scattered by the composite, which determines the depth of cure [25]. The attenuation coefficient describes this character of composites and the depth of penetration [26]. According to the Beer-Lambert law, absorption and scattering cause the light to attenuate exponentially with distance [27, 28]. The relation between parameters can be expressed by the following equation:

$$I = I_0 (1 - R^2)\exp(-\alpha x) \quad (1)$$

where I is the irradiance transmitted from the bottom of the samples, I_0 is the incidence optical power, R is the Fresnel reflectance of the sample surface, α is the attenuation coefficient, and x is the thickness of the sample.

The objective of this study was to determine the impact of increasing concentrations of graphene addition on colour, and to evaluate its effect on optical properties during the light curing of retainer resin adhesives. The relationship between light transmission and the degree of conversion was also investigated. The null hypotheses were that different concentrations of graphene addition will not influence (1) the colour and translucency parameter, (2) light irradiance, and (3) the degree of conversion of orthodontics retainer adhesives.

MATERIAL AND METHODS

Preparation of the resin composites

Two commercial materials, a packable composite (Transbond; 3 M ESPE) and a flowable composite (Transbond Supreme; 3 M ESPE), were used in this study. The composition and other product information are listed in Table 1. The graphene was provided by the Centre for Advanced 2D Materials, National University of Singapore as characterized elsewhere [29]. Graphene powder was added to the adhesives in three different concentrations (0.01, 0.05, or 0.1% wt) and mixed with the Speedmixer (DAC 150.1 FVZK; Haushild) at 3000 rpm for 5 min. Adhesives without graphene were the controls.

Surface microscopy

Disc-shape resin blocks after degree of conversion measurement were embedded in epoxy resin, then polished successively under water up to 1200-grit silicon-carbide papers at 250 rpm. The surfaces of the samples were imaged wither using a light microscope (Revolve, Echo; A BICO) at 20× magnification or scanning electron microscope (SEM, Quanta 250; FEI). Images were obtained in backscattered electron mode at 500× and 2000× magnification.

Colour measurement

Samples (1 mm thickness x 4 mm diameter) were made using teflon moulds. The materials were inserted in the moulds and covered with mylar strips and glass slides (top and bottom surfaces) were used to extrude the excess materials. The samples were cured after the removal of the top glass slide using a light-curing unit (Elipar S10; 3 M ESPE, wavelength range: 430–480 nm, peak wavelength: 455 nm) at an output of 1200 mW/cm^2 for 20 s. The colour measurements were performed with a colorimeter (Minolta Chroma Meter CR-221) according to the CIE-Lab (Commision Internationale de l'Eclairage, L*, a*, b*) coordinates with a D65 standard light

TABLE 1 The manufacturers' compositional information.

Product	Manufacturer	Composition
Transbond LR	3 M-ESPE	Silane treated quartz: 75%−85% Bisphenol A diglycidyl ether dimethacrylate (Bis-GMA): 5%−15% Triethylene glycol dimethacrylate (TEGDMA): < 10% Silane treated silica: < 2% N,N-Dimethylbenzocaine: < 0.5%
Transbond Supreme LV	3 M-ESPE	Silane treated ceramic: 50%−60% TEGDMA: 10%−20% Bis-GMA: 10%−15% Silane treated zirconia: 1%−10% Bisphenol A polyethylene glycol diether dimethacrylate (Bis-EMA-6): 1%−5% Silane treated silica: < = 5% Reacted polycaprolactone polymer: < 5% Diphenyliodonium hexafluorophosphate: < 0.5% N,N-Dimethylbenzocaine: < 0.3%

source [30]. The colorimeter was calibrated with a standard white plate with a 3 mm diameter measuring area and 45° illumination angle, and 0° viewing angle. Following measurement of three samples per group after 24 h of storage at room temperature, the results were averaged. The colour difference, ΔE of each group compared with the control group (0% wt of graphene) was calculated as follows [31]:

$$\Delta E = \left[(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2\right]^{1/2} \quad (2)$$

where ΔL^*, Δa^*, and Δb^* are the differences in lightness and colour between specimens of each group and control group.

The translucency parameter was calculated with the measurements against a black and white background as follows [32]:

$$TP = \sqrt{(L_1 - L_2)^2 + (a_1 - a_2)^2 + (b_1 - b_2)^2} \quad (3)$$

where L_1, a_1 and b_1 are values measured against the black background, while L_2, a_2 and b_2 are values measured against the white background.

Measurement of irradiance during curing

A Managing Accurate Resin Curing System- Light Collector (MARC-LC) system (Bluelight Analytics) with two 4 mm cosine corrector sensors (top and bottom sensor) was used to collect real-time irradiance and radiant exposure received on the top and bottom surfaces of the composite specimens [33]. The same light curing unit (Elipar S10; 3 M ESPE) was powered cordlessly with a full charge and mounted on the AMRC fixing arm. A matrix strip was placed on the sensor with no sample to adjust the irradiance to 1200 mW/cm^2 while the irradiance was shown in real-time.

Specimens were fabricated with customized dark teflon moulds with 4 mm diameter and three different thicknesses (0.3, 1.3, and 3 mm) as described before. Real-time data for the instantaneous irradiance during curing were displayed on the laptop. Light irradiance was continuously measured during the curing at both top and bottom of the specimens with three different thicknesses, and three replicates of each thickness were tested.

Calculation of optical parameters

The reflectance and attenuation coefficient instantaneously after curing and post-irradiation were fitted according to Equation (1) [34]. In this study, it can be assumed that the reflectance is independent of the polarization angle and is the same for entering or leaving a medium with a high optical density, resulting in the same surface reflectance at the top and bottom of each sample.

Measurement of degree of conversion during curing

The degree of conversion ($n = 3$) was measured by a Fourier-transform infrared (FTIR) spectrometer (ALPHA II; Bruker) with a single attenuated total reflection (ATR) accessory. The spectrum was obtained at a resolution of 4 cm^{-1} in a range of 4000–400 cm^{-1} wavelengths after storage in room temperature for 24 h. A mould was placed over the FTIR-ATR crystal and uncured composite paste was dispensed into the mould. The specimen was pressed from the top with a Mylar strip, followed by a glass slide to remove air bubbles. After removing the glass slide, the spectrum of uncured composites was collected first. Then, the same light curing unit of mean irradiance 1200 mW/cm^2 was immediately

used to cure specimens for 20 s. The spectrum was collected using OPUS SOFTWARE (VERSION 8.1; BRUKER OPTIK). To calculate the degree of conversion, the peak heights of the aliphatic C = C absorbance peak at 1637 cm^{-1} and the aromatic C-C absorbance peak at 1608 cm^{-1} for uncured and cured samples were utilised according to the following equation [35]:

$$DC\% = \left[1 - \frac{\left(\frac{aliphatic\ C=C}{aromatic\ C-C}\right)_{polymerized}}{\left(\frac{aliphatic\ C=C}{aromatic\ C-C}\right)_{unpolymerized}}\right] \times 100\% \quad (4)$$

Statistical analysis

Sample size calculations were performed using the G*POWER SOFTWARE (V. 3.1.9.2; Heinrich Heine University), with $\alpha = 0.05$, power $(1–\beta) = 80\%$, and data from the pilot study. All data were analysed with SPSS (VERSION 25.0, SPSS). The normality of the data distributions was confirmed using the Shapiro-Wilk Test ($p < 0.05$). Results are therefore presented using mean values ± SD. One-way analyses of variance (ANOVA) with Tukey's post hoc test were used to test for significant differences in ΔE, translucency parameter, light transmission, and degree of conversion between groups of the same material with different graphene concentrations. The Student's *t*-test was used to compare the effect of different materials. The level of significance was set at $\alpha = 0.05$. Correlation of colour change with graphene concentration and correlation of translucency parameter, transmission and degree of conversion were tested with Spearman correlation coefficient test.

RESULTS

Surface microscopy

Optical microscopic images (20 ×) of each group of materials are shown in Figure 1. Different filler patterns were observed in the two composite types. For the packable composite, larger and different sizes of fillers were found, while the fillers were not obvious in the flowable composite. In all composites with the graphene addition, the graphene was uniformly distributed and graphene agglomerations with a size of about 10 μm could be identified. It appeared that as the graphene content increased, the agglomerated graphene also increased.

Representative SEM images under backscattered electron signal of polished cured resin composites are presented in Figure 2, the adequate contrast between resin matrix and fillers was observed. The shapes and sizes of the filler particles were different between packable and flowable composite. The packable composite showed particles ranging from 2 to 50 μm with irregular shapes, while the flowable composite featured homogeneous, smaller filler particles with spherical shapes, approximately 1 μm in size.

Colour measurement

The CIE L^*, a^*, and b^*values for specimens are presented in Table S1. Graphene addition had a significant effect on the colour (Figure 3A). The colour differences were mainly influenced by variations in lightness (L* values), as the graphene was darker than both the packable and flowable composite. Furthermore, an increase in graphene content led to a corresponding increase in ΔE. Statistically significant correlation was found between ΔE and graphene addition was found in both packable composite ($\rho = 0.95$, $p = 0.001$) and flowable composite ($\rho = 0.96$, $p = 0.003$). The highest ΔE was identified in the 0.1 wt% graphene addition group in both packable and flowable composites (ΔE = 53.7 and 33.5, respectively).

Figure 3B presents the translucency parameter (TP) of the tested samples. All groups with graphene addition showed a statistically significantly lower translucency parameter than the control group, even with the lowest concentration (0.01 wt%) ($p < 0.05$). When the graphene content increased to 0.1 wt%, both packable and flowable composite became almost opaque (TP = 1.7 ± 0.1 and 3.7 ± 0.1, respectively). It was also observed that in the control groups, the translucency parameter of packable composite was significantly higher than that of flowable composite ($p < 0.05$). Conversely, in the other groups where graphene was added, the translucency parameter of packable composite was significantly lower than that of flowable composite ($p < 0.05$).

Measurement of irradiance during curing

Changes in the light irradiance passing through the materials with different graphene concentrations and thicknesses are shown in Figure 4. For irradiance measurement, statistically significant differences in transmitted irradiance between different concentrations of graphene addition can be identified for each thickness ($p < 0.05$). The transmitted light decreased as the graphene concentration and sample thickness increased. The statistically significantly highest values of transmitted irradiance can be identified for the group of 0.3 mm thickness ($p < 0.05$); among them, the significantly highest transmitted irradiance is detected in the groups with no graphene addition ($p < 0.05$). No light was detected in either of the packable or flowable composites with 0.1 wt% graphene

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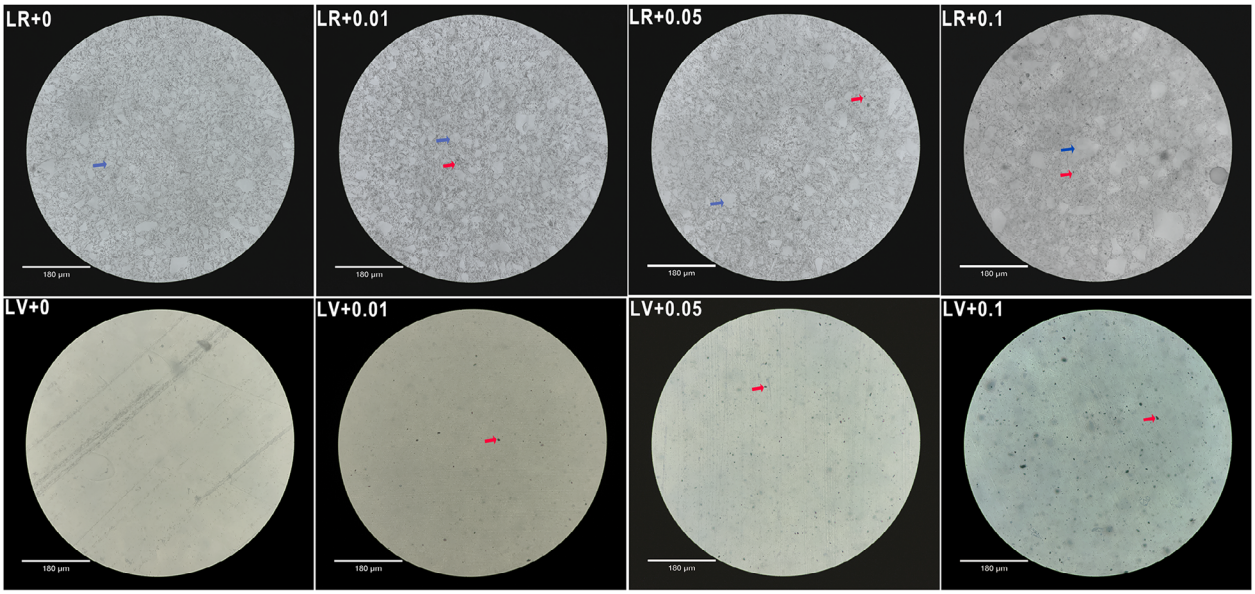

FIGURE 1 Representative microscopic images of the samples in the different groups. Blue arrows point to inorganic fillers, red arrows point to graphene agglomeration. LR, packable composite (Transbond LR); LV, flowable composite (Transbond Supreme LV); + indicates the %wt of graphene added to the composites. Bar = 180 μm.

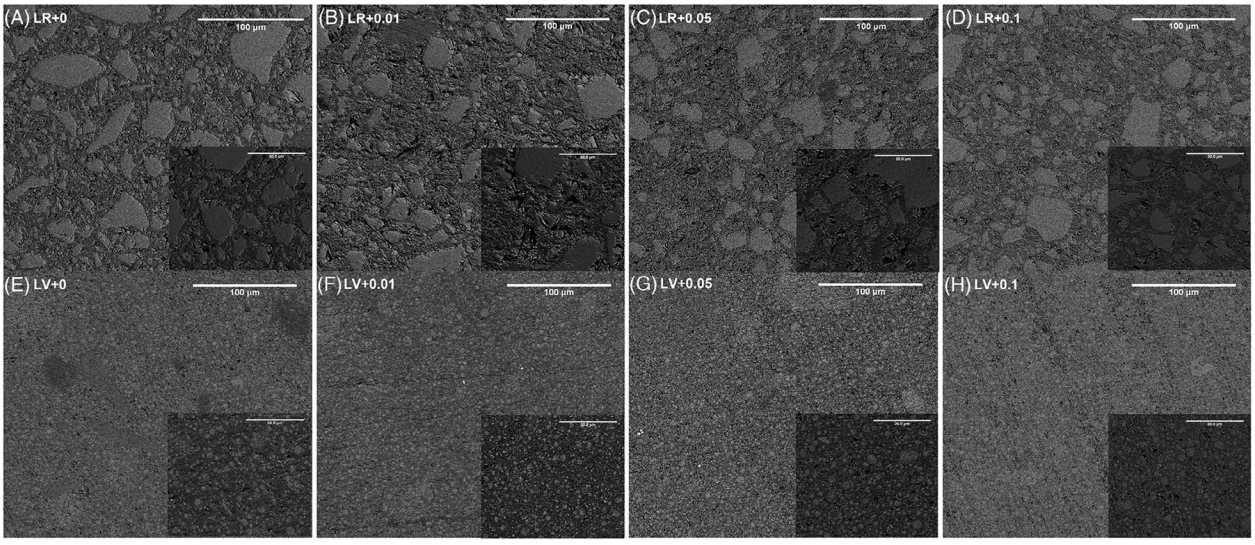

FIGURE 2 Representative SEM images of samples in the different groups. LR, packable composite (Transbond LR); LV, flowable composite (Transbond Supreme LV); + indicates the %wt of graphene added to the composites. Bars = 100 and 30 μm (inserts).

at 3 mm thickness, which means the absolute opacity was reached.

Calculation of optical parameters

The irradiance received at the bottom of the material at the instant of light initiation and post-irradiation is shown in Figure 5. It can be seen that with the increase of specimen thickness, the irradiance decreased rapidly. Furthermore, among specimens of the same thickness, a higher irradiance was detected in the group with no graphene added. As the content of graphene increased, the irradiance decreased.

The irradiance emerging from the light curing unit is reflected at both the incidence and the exitance surfaces and absorbed within the resin composites, which can be described by the Equation (1). The reflectance and attenuation coefficients of the material estimated from the fit are shown in Table 2.

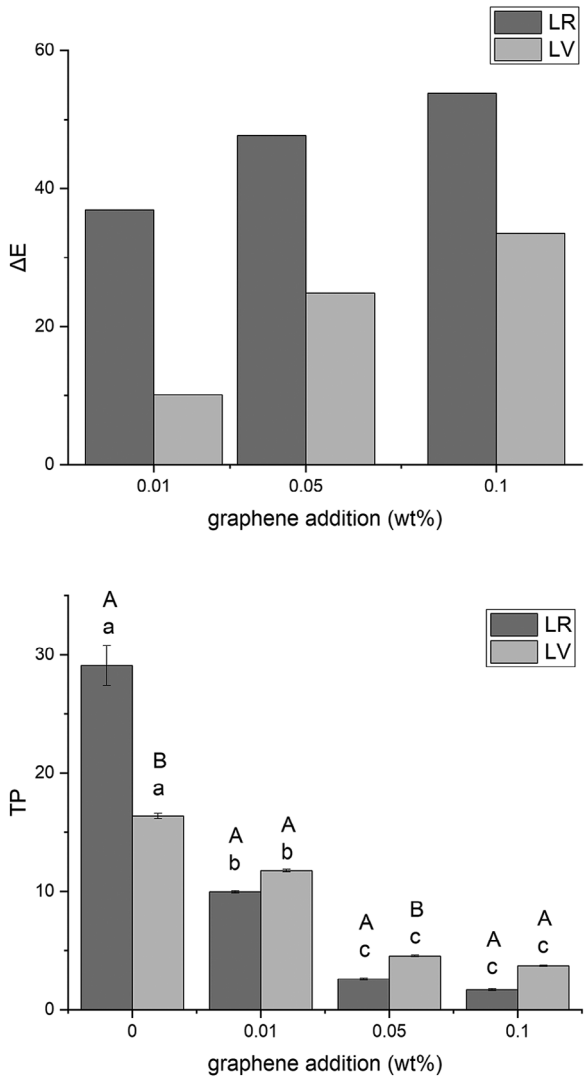

FIGURE 3 Mean values of the optical properties for different composites with different graphene additions. Top panel: The colour difference (ΔE), Lower panel: The translucency parameter (TP). Same upper-case letters indicate no statistically significant differences between different types of composites ($p > 0.05$); same lower-case letters indicate no significant differences between different graphene concentrations of each adhesive ($p > 0.05$). LR is packable Transbond LR composite and LV is flowable Transbond Supreme LV.

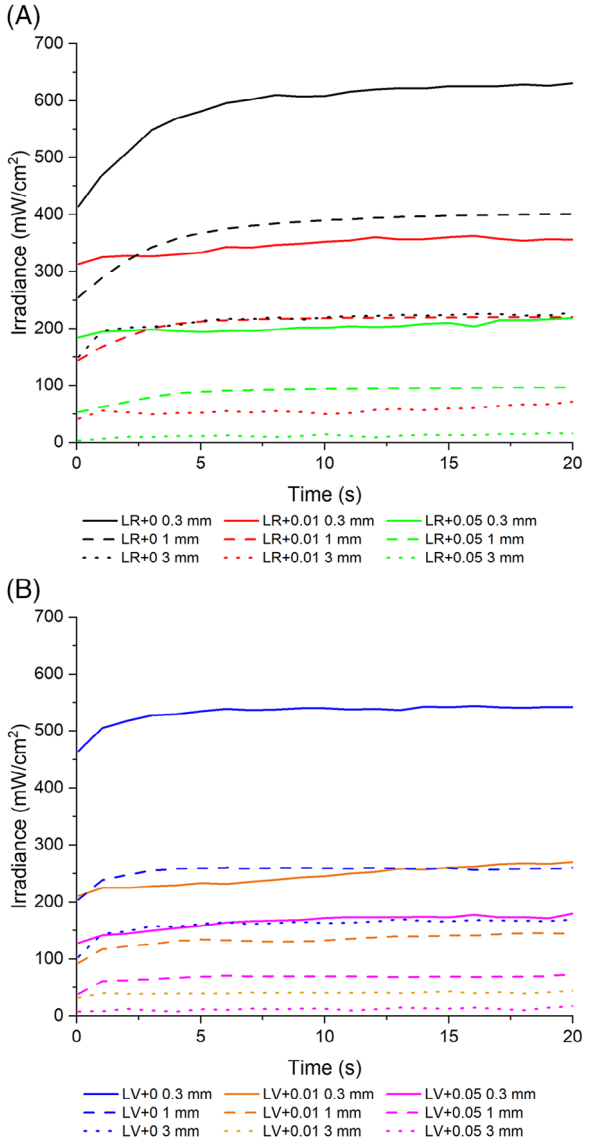

FIGURE 4 Curves depicting the instantaneous light irradiance during the light exposure through different thickness specimen. (a) LR, packable composite (Transbond LR) with different amounts of graphene, (b) LV, flowable composite (Transbond Supreme LV) with different amounts of graphene.

Measurement of degree of conversion

The mean and standard deviation of the degree of conversion in specimens of each group after 24 h post-curing is presented in Table 3. Except for the packable composite with 0.01 wt% graphene, which showed a statistically higher degree of conversion than the control group, graphene addition and material type had no significant effect on the degree of conversion of the remaining groups ($p < 0.05$). No correlation was found between the degree of conversion and the translucency parameter after 24 h curing of the material and the light transmittance at the end of curing. Transmittance and transparency were statistically positive correlated in both packable ($\rho = 0.94$, $p = 0.005$) and flowable ($\rho = 0.94$, $p = 0.005$) composite. However, at each graphene content, the transmittance of the packable composite was consistently higher than that of the flowable composite regardless of the translucency parameter.

DISCUSSION

This study evaluated the colour change of composite materials after adding different contents of graphene, using a colourimeter with the CIE L*a*b* system, which can detect

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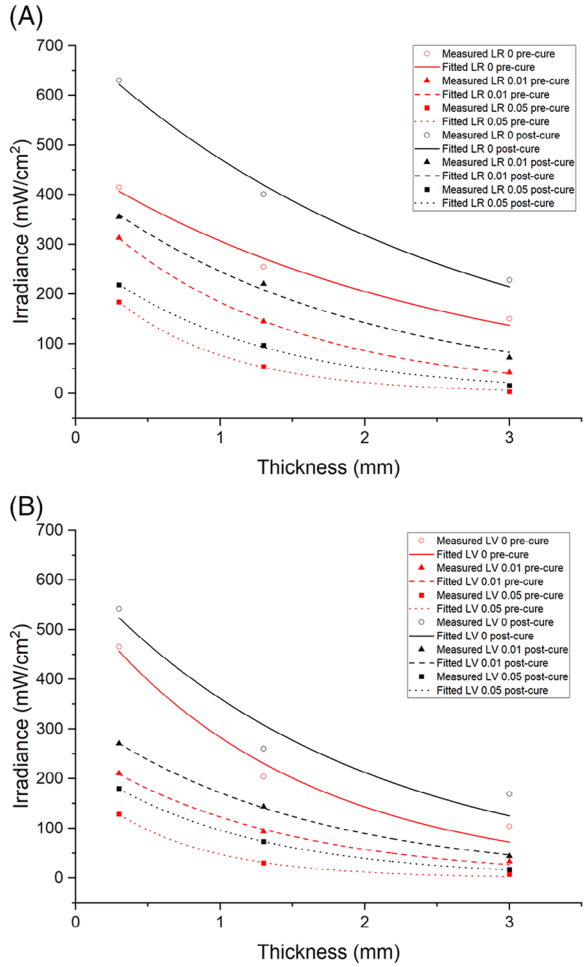

FIGURE 5 Curves depicting the irradiance of specimens of different thickness. Top panel: LR, packable composite (Transbond LR) with different amounts of graphene. Bottom panel: LV, flowable composite (Transbond Supreme LV) with different amounts of graphene.

TABLE 2 Reflectance R and attenuation coefficient α at the instant of light initiation and post-irradiation, calculated from the fitted output.

Materials	R_0	α_0 (mm^{-1})	R^2	R_1	α_1 (mm^{-1})	R^2
LR+0	0.38	0.40	0.98	0.24	0.39	0.99
LR+0.01	0.43	0.76	0.99	0.41	0.54	0.99
LR+0.05	0.53	1.25	0.99	0.51	0.86	0.99
LV+0	0.32	0.68	0.95	0.28	0.41	0.99
LV+0.01	0.53	0.76	0.99	0.48	0.65	0.99
LV+0.05	0.60	1.41	0.99	0.56	0.89	0.99

Note: Numbers in Material column indicate the amount of graphene added (wt%). R_0 and α_0 are values at the instant of light initiation, while R_1 and α_1 are values post-irradiation. R^2 is the goodness of fit. LR is packable transbond LR and LV is flowable transbond supreme LV composite.

TABLE 3 Mean (standard deviation) of the translucency parameter (TP) and degree of conversion (DC) at 24 h post-irradiation and transmittance (T) after light exposure of specimen with 1 mm thickness.

Materials	TP	T (%)	DC (%)
LR+0	28.9 (1.2) ^{Aa}	37.4 (1.4) ^{Aa}	72.9 (3.2) ^{Aa}
LR+0.01	10.0 (0.1) ^{Ab}	23.8 (1.4) ^{Ab}	80.1 (0.8) ^{Ab}
LR+0.05	2.6 (0.1) ^{Ac}	8.1 (0.3) ^{Ac}	75.7 (0.4) ^{Aab}
LR+0.1	1.7 (0.3) ^{Ac}	–	77.5 (1.8) ^{Aab}
LV+0	16.4 (0.2) ^{Ba}	29.9 (1.2) ^{Ba}	77.8 (5.6) ^{Aa}
LV+0.01	11.6 (0.3) ^{Bb}	14.9 (2.2) ^{Bb}	80.7 (4.3) ^{Aa}
LV+0.05	4.5 (0.2) ^{Bc}	7.4 (1.0) ^{Ac}	76.0 (5.3) ^{Aa}
LV+0.1	3.7 (0.1) ^{Bd}	–	73.1 (4.6) ^{Aa}

Note: Numbers in Material column indicate the amount of graphene added (wt%). Same upper-case letters indicate no significant differences between different type of adhesive ($p > 0.05$); same lower-case letters indicate no significant differences between different graphene concentrations of each adhesive ($p > 0.05$). LR is packable composite Transbond LR and LV is flowable composite Transbond Supreme LV.

the colour changes that are not visible to the human eye and express colour differences in units that may be related to visual perception and clinical significance [36]. The result of this study indicated that the addition of graphene significantly changed the colour of materials (Figure 3A). The impact was greater with increasing graphene concentration. The greatest variations were found in L*, which may be due to graphene being significantly darker than the resin composite. The ΔE value under 1.6 was considered undetectable by the human eye. Most studies reported ΔE value under 3.3 as clinically acceptable [37–39]. In this study, even the lowest graphene addition of the two commercial composites exceeded this value (ΔE = 36.9 and 10.1). Clinically, when the colour of dental composites does not match that of the natural tooth, it can impact aesthetics and lead to patient rejection. In this study, the adhesives studied were applied to the lingual surface of teeth, which is less likely to affect aesthetics. However, to expand the use of graphene in dentistry, it is important to understand its effect on colour.

Many studies have evaluated the translucency of resin composite using the translucency parameter [40–43]. The translucency parameter refers to the colour difference between the colour over a white background and a black background. In present studies, the translucency parameter values of all groups decreased as the increase of graphene concentration (Figure 3B), therefore, the first null hypothesis was rejected. Although research has proven the exceptional optical transmittance of graphene and its potential for transparent film fabrication, the optical transmittance is still adversely affected by the unavoidable agglomeration of graphene when it is mixed with resins [44, 45]. According to the Rayleigh theory, the light scattering increases with the increasing filler

particle size and filler/matrix refractive index mismatch [46, 47]. Graphene has a higher refractive index compared with resin matrix and other fillers [48, 49]. The increased mismatch decreased the translucency of materials. It was also noted that the packable composite was significantly more translucent than the flowable composite at the baseline. This can be due to the fact that the refractive index of bisphenol A-glycidyl methacrylate (Bis-GMA) (1.55) is closer to the silica filler than that of triethylene glycol dimethacrylate (TEGDMA) (1.46) and ethoxylated bisphenol A glycol dimethacrylate (Bis-EMA) (1.54). Additionally, variations in translucency can arise from the use of different fillers in the two materials. It can be seen from Figure 4 that the addition of graphene has a greater impact on the translucency parameter and colour of packable composite than of flowable composite. This could be due to packable composite having a greater difference in refractive index and particle size with graphene than seen for the flowable composite. However, more research is needed to confirm this inference.

When curing resin composites, light may experience reflected, absorbed, scattered or transmitted. The attenuating effects of light while passing through the composites are summarised together with absorption, and reflected by attenuation coefficient α. The optical analysis of this study is mainly based on Beer-Lambert's law, which is well-established for solids and solutions absorbing radiation. Beer-Lambert's law has also been applied to understand the light phenomenon of dental composites [46, 50, 51]. From the result of this study, groups with graphene addition showed a lower light transmittance, and higher reflectance and attenuation coefficient, the second null hypothesis was rejected (Figure 5 and Table 2). Studies have shown that graphene has a strong optical absorbance and can be used as photocatalytic material [52]. The mismatching between graphene and matrix may cause enhanced light scattering, thus a higher attenuation coefficient and lower light transmission [53]. It was also observed that as the light curing proceeded, the reflectance and attenuation coefficient decreased (Table 2). This may be due to a decrease in the refractive index mismatch between the resin and filler as curing proceeds. However, as the refractive index is still much higher than matrix phase, the reflectance and attenuation coefficient were higher in graphene added groups compared with control group.

The two commercial composites evaluated in this study showed significantly different results in their optical properties (Figures 3–5). This could be due to their different fillers and matrix components. The optical properties of resin composites are determined by various factors, including the type of resin matrix, fillers and other additives, and the concentration of fillers. Both the two commercial composites in this study contain Bis-GMA and TEGDMA as the matrix, however, with different concentrations, while the flowable composite also contains Bis-EMA. Bis-GMA is the majority of the matrix in dental resin composites [54]. However, it has a high viscosity that is unable to incorporate the inorganic filler. Therefore, TEGDMA and Bis-EMA are usually used as co-monomers to reduce the viscosity and increase the degree of conversion [55, 56]. Fillers are also important in determining the properties of composites. Ideally, the refractive indices of the fillers should be similar to the resin matrix to provide sufficient the degree of conversion [57]. The size, shape and content were also different in the two types of commercial composites. The size and distribution of fillers affect the light scattering during photo-polymerization. Generally, the difference between the refractive indices of the inorganic filler phase and the organic monomer/polymer matrix leads to a reduction in the transmission of light due to light refraction and diffraction at the filler-matrix interface, and results in lower degree of conversion and depth of cure [58].

No correlation between the degree of conversion and the translucency parameter or the light transmission was found. This contradicted previous research findings [59, 60]. Although the addition of graphene caused a significant reduction in the light transmission of the material, it had no adverse effect on the degree of conversion. The packable composite with 0.01 wt% graphene added even showed a higher degree of conversion than the control ($p < 0.05$), thus, the third null hypothesis was rejected. This may be due to the complexity of the factors affecting the degree of conversion. Previous studies have found materials that contain graphene to have high thermal conductivity, leading to better polymerization [61]. Moreover, compared with other studies, the graphene content used in this study was relatively low. This lower graphene content might have contributed to a reduced adverse effect on the degree of conversion. A positive correlation was found between the translucency parameter and transmittance for different graphene concentrations of each material.

This study analysed the optical characteristic of graphene added experimental dental composites. However, as the composition of commercial composites are variable, experiments with different composite resins are needed to refine the conclusions. Moreover, deformation and change in light transmission due to material shrinkage during the polymerization were not taken into account in this study. For future work, a refined model is needed. Additionally, it should be considered that many compounds such as fluoride [62], bleaching agents [63] and other contaminants [64] could have an influence on bond strength of orthodontic adhesives. Therefore, the effect of graphene addition on bond strength should be evaluated in future studies.

In conclusion, the addition of graphene adversely affected the optical properties of the composites in this study. The experimental results showed that as the concentration of graphene increases, the colour of the material becomes darker and the transmittance decreases. No transmitted light was detected when the concentration of graphene reached 0.1 wt%.

Within the selected concentrations, the addition of graphene had no adverse effect on the degree of conversion.

AUTHOR CONTRIBUTIONS

Conceptualization: Shiyao Liu; Ahmed EI-Angbawi; Ruidong Ji; Nikolaos Silikas. **Methodology**: Shiyao Liu; Ahmed EI-Angbawi; Vinicius Rosa; Ruidong Ji; Nikolaos Silikas. **Software**: Shiyao Liu. **Validation**: Shiyao Liu. **Formal analysis**: Shiyao Liu. **Investigation**: Shiyao Liu. **Data curation**: Shiyao Liu. **Writing-original draft**: Shiyao Liu. **Writing-review and editing**: Shiyao Liu; Ahmed EI-Angbawi; Ruidong Ji; Nikolaos Silikas. **Visualization**: Shiyao Liu. **Supervision**: Ahmed EI-Angbawi; Vinicius Rosa; Nikolaos Silikas. **Resources**: Vinicius Rosa. **Project administration**: Nikolaos Silikas.

ACKNOWLEDGMENTS

The research was not supported financially.

CONFLICT OF INTEREST STATEMENT

The authors declare that they have no competing interest.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

How to cite this article: Liu S, El-Angbawi A, Ji R, Rosa V, Silikas N. Optical effects of graphene addition on adhesives for orthodontic lingual retainers. Eur J Oral Sci. 2023;e12966. https://doi.org/10.1111/eos.12966