The addition of carbon nanotubes to orthodontic adhesives: an in vitro study

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Objective: The evolution of adhesive dentistry and the addition of nanoparticles has heralded an improvement in the mechanical properties of adhesives. Thus, the aim of the present study was to evaluate the effects produced by carbon nanotubes (CN) added to two adhesives used for indirect bonding by an examination of the shear bond strength (SBS) and location of bond failure.

Methods: One hundred and sixty bovine incisors were randomly divided into eight groups (N = 20): (1) indirect bonding with Sondhi adhesive; (2), (3) and (4) indirect bonding with Sondhi adhesive into which CN at 0.5%, 0.25% and 0.05% concentrations were incorporated; (5) indirect bonding with Concise adhesive; (6), (7) and (8) indirect bonding with Concise adhesive into which CN at 0.5%, 0.25% and 0.05% concentrations were incorporated. Following etching with 37% phosphoric acid and the placement of brackets, maximum shear bond strength (SBS) was measured with a mechanical testing machine. The location of bond failure was evaluated using the Adhesive Remnant Index (ARI). The SBS between groups and ARI scores were statistically analysed ($p < 0.05$).

Results: There was no statistical difference ($p > 0.05$) in SBS or ARI.

Conclusions: CN addition to Concise and Sondhi adhesives did not influence the SBS and the ARI of the brackets. Therefore, in the conditions of this experiment, there was no benefit in the addition of CN to orthodontic adhesives.

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Introduction

Developments in the field of dental materials, specifically in dental adhesives, have led to the replacement of banding procedures by the bonding of orthodontic attachments.[1] The bond strength of the enamel/composite/bracket interface must be strong enough to support functional loads and mechanical forces resulting from the orthodontic treatment.[2] Despite recent improvements in the development of new adhesives, studies have shown that there is still a 2% to 6% bond failure rate, which leads to extra material costs, increased chair time and increased total treatment time.[3,4]

An improvement in the mechanical properties of dental composites has been found in the incorporation of filler particles.[5] While hybrid, microhybrid and flowable composites have been used, the addition of nanoparticles to the composite resins has been shown to be advantageous.[6] A benefit of nanoparticles compared with other larger types of particles is the production of an increased interfacial area per volume between the host polymer and the nano-element. When a uniform dispersion of nano-sized filler particles in organic matrix is achieved, the intrinsic properties of the nanoparticles may be transferred to the unit formed.[7]

A relatively new class of nanoparticles, discovered by Iijima[8] in 1991, is the class of carbon nanotubes

(CN). The CN are thin and long cylinders of graphite, formed by one or more layers of carbon atoms organised in a hexagonal lattice. There are two types of CN identified as multi-walled varieties, which are formed by several cylindrical walls (one within another), or single-walled varieties, which are formed by only a single cylindrical wall.[8] This type of nanoparticle has a length between 10 and 1000 nm, a diameter between 10 and 50 nm, and a weight percentage that may vary between 0.005 and 5.0 nm.[9] The CN/organic matrix bonding is mediated by weak van der Waals forces.[7] Nevertheless, the adhesion strength generated between the CN/organic matrix is very large, such that mechanical compression and fatigue resistance are enhanced. The bond strength, toughness, elasticity, durability, dimensional stability and coefficient of thermal expansion of the composite are also improved.[7,10] The CN are considered the best material for the reinforcement of resins[8] and their addition to orthodontic adhesives is predicted to increase the bond strength between tooth enamel and brackets.

Silverman et al.[11] introduced indirect bonding in Orthodontics in 1972. This unique technique is divided into two steps requiring the positioning of brackets on the working model and their subsequent transfer to the patient's dentition using fabricated, individual trays.[12] Bracket positioning is considered to be more accurate and requires less clinical time for attachment.[13] However, it has been suggested that the interface between the pre-cured composite resin and the adhesive may be weak and result in a higher number of clinical failures.[14,15]

The present study aimed to evaluate the changes that might result from the addition of carbon nanotubes (CN) to two types of adhesives commonly used in indirect bracket bonding by examining the shear bond strength (SBS) and the amount of adhesive remaining on the teeth after debonding.

Materials and methods

Sample size calculation

The sample size for the study was calculated based on a formula advocated by Pandis,[16] for a significance level of 0.05 and a power of 90% to detect a clinically meaningful difference of 2 Mpa (±2 Mpa) for the shear bond strength between groups. A power analysis showed that 20 specimens were needed for the study.

Adhesives preparation

A 10% (w/v) solution of sodium dodecyl sulfate (SDS) (Reagen, Colombo, Brazil) was prepared as a surfactant to stabilise CN in water and to promote bonding to the monomers of the Sondhi Rapid Set adhesive base pastes (3M Unitek, CA, USA) and Concise (3M Unitek, CA, USA).[17-19] Single-walled CN (Sigma Aldrich, São Paulo, Brazil) measuring 0.7 to 1.1 nm in diameter and 800 nm in length were added to the solution to obtain a concentration of 1 mg/ml. Homogenisation was achieved by ultrasound (80 Hz, Elmasonic P, Germany) for 10 minutes, together with heating.[18,19] The prepared solution was included in the adhesive base paste to obtain 0.1%, 0.5% and 1% CN concentrations, so that when base and catalyst pastes were combined, the total adhesive concentration of CN was 0.05%, 0.25% and 0.5%.[20] Finally, the base pastes were homogenised again by using ultrasound for 10 minutes, and excess water was removed in a vacuum oven at 70° C for 24 hours.[18,19]

Specimen preparation

One hundred and sixty bovine incisors were randomly divided into eight groups (N = 20). The teeth were disinfected in 0.1% thymol solution for one week[21] and, subsequently, stored in distilled water until use (maximum time of one month).

Before bonding, groups of five teeth were aligned and embedded in type III dental plaster (Vigodent, Rio de Janeiro, Brazil) (Figure 1).

Alginate impressions (DencrilGel, Dencril, Pirassununga, Brazil) were taken of the sets of teeth and poured in orthodontic plaster (Vigodent, Rio de Janeiro, Brazil), after which the models were left to set for one hour (Figure 2).

Bonding procedure

Transbond XT composite resin (3M Unitek, CA, USA), and maxillary incisor brackets (Edgewise Standard, Morelli, Sorocaba, Brazil) were used for all groups. The brackets were pressed to the most convex buccal surface of the plaster models using a 450 g load,[3] measured with a dynamometer (Morelli, Sorocaba, Brazil) and positioned using a parallelometer (Bio-Art, São Carlos, Brazil). Excess resin was removed using a scaler, and each bracket was light cured for 20 seconds from the cervical aspect and 20 seconds

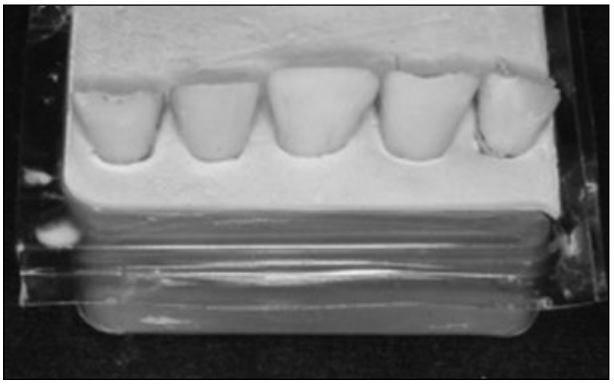

Figure 1. Bovine teeth embedded in dental plaster.

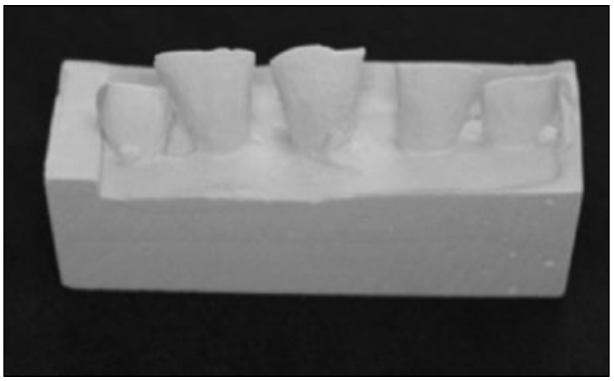

Figure 2. Plaster models for indirect bonding.

from the incisal aspect. For all groups, light curing was performed with a previously calibrated light-emitting diode (LED) unit (ThreeH X-lite II, China) at 1100 mW/cm^2 irradiance. For indirect bonding, transfer trays were made with condensation silicone (Perfil, Rio de Janeiro, Brazil). After sufficient curing time (five minutes), excess silicone was removed using a scalpel blade (15C, Lamedid, São Paulo, Brazil), following which the transfer trays with the embedded brackets were removed from the models (Figure 3). The custom resin bases were sandblasted (Micro-etcher, Bio-Art, Brazil) with 90 µm aluminum oxide particles (Bio-Art, São Carlos, Brazil) at a 1 cm distance for two seconds.

All teeth were cleaned with a rubber cup and pumice stone, rinsed with water for 10 seconds and dried with oil-free compressed air. Phosphoric acid (Condac 37, FGM, Joinville, Brazil) at a concentration of 37% was applied to the buccal surface of each tooth for 30 seconds. The samples were then rinsed with water for 10 seconds and air-dried.

The various adhesives were applied according to the manufacturer's instructions.

Group 1: (Sondhi Control): Sondhi Rapid Set (3M Unitek, CA, USA).

Group 2: (0.5% Sondhi): Sondhi Rapid Set (3M Unitek, CA, USA) with the addition of 0.5% CN.

Group 3: (0.25% Sondhi): Sondhi Rapid Set (3M Unitek, CA, USA) with the addition of 0.25% CN.

Group 4: (0.05% Sondhi): Sondhi Rapid Set (3M Unitek, CA, USA) with the addition of 0.05% CN.

Group 5: (Concise Control): Concise (3M Unitek, CA, USA).

Group 6: (0.5% Concise): Concise (3M Unitek, CA, USA) with the addition of 0.5% CN.

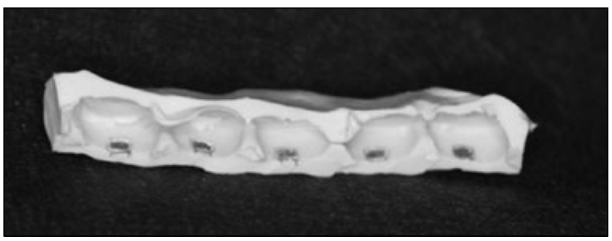

Figure 3. Transfer tray containing the brackets.

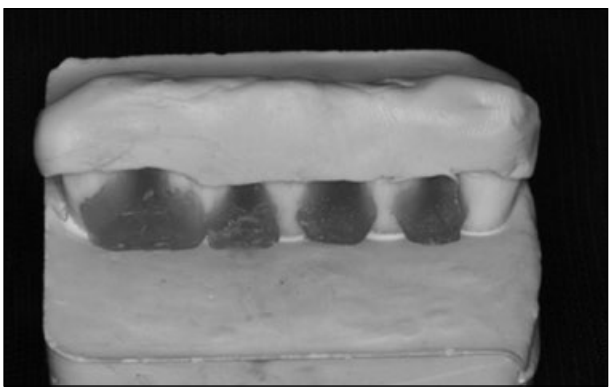

Figure 4. Transfer tray positioned on the sample.

Group 7: (0.25% Concise): Concise (3M Unitek, CA, USA) with the addition of 0.25% CN.

Group 8: (0.05% Concise): Concise (3M Unitek, CA, USA) with the addition of 0.05% CN.

After the application of adhesive, the transfer trays were immediately positioned on the samples (Figure 4). Following curing and the removal of the trays, the teeth were immersed in distilled water at room temperature for 72 hours[14] prior to the SBS tests.

Debonding procedure

The brackets were debonded using a mechanical testing machine (TA HD Plus - Stable Micro System)

Table I. SBS means and standard deviations of groups 1, 2, 3, and 4 (ANOVA).

Group	N	Mean (MPa)	Standard deviation (MPa)
1 (Sondhi Control)	20	7.5584 [b]	5.8806
2 (0.5% Sondhi)	20	9.2863 [a]	4.5663
3 (0.25% Sondhi)	20	9.9766 [a]	5.0413
4 (0.05% Sondhi)	20	8.1691 [a]	4.6611

[a,b] Similar letters indicate no statistically significant differences between groups ($p > 0.05$).

Table II. SBS means and standard deviations of groups 5, 6, 7 and 8 (ANOVA).

Group	N	Mean (MPa)	Standard deviation (MPa)
5 (Concise Control)	20	8.4444 [a]	5.1728
6 (0.5% Concise)	20	6.9272 [a]	5.0053
7 (0.25% Concise)	20	6.9941 [a]	4.4447
8 (0.05% Concise)	20	9.5794 [a]	4.3720

[a] Similar letters indicate no statistically significant differences between groups ($p > 0.05$).

with a 1 kilo-newton load and at a speed of 0.5 mm per minute. The results were reported in Newtons (N) and converted to MPa considering the area of the bracket base (0.16 cm^2).

Adhesive remnant index (ARI)

After bracket debonding, the amount of composite remaining on the enamel was classified using the adhesive remnant index (ARI) following examination under a light microscope (Carl-Zeiss, São Paulo, Brazil) at 10 times magnification.[22] ARI scores were classified as 0 = no adhesive remnant on enamel; 1 = less than 50% adhesive on enamel; 2 = more than 50% adhesive on enamel; 3 = all adhesive left on enamel. SBS tests and ARI scoring were conducted by a single operator blinded to the sample group assignment.

The ARI replicability was tested by a second evaluation of the specimens, 30 days after the first classification. The results were statistically compared using the Wilcoxon test. Data showed excellent reproducibility ($p > 0.05$).

Statistical analysis

Means and standard deviations were calculated for each group. SBS data were analysed using the D'Agostino normality test and normal distribution was verified. One-way ANOVA and the Tukey test were applied to detect possible differences in SBS between groups. The non-parametric Kruskal-Wallis test and the Dunn post-test were used to detect differences in ARI scores. The level of significance was set at 0.05.

Results

Table I shows the results of SBS tests for groups 1, 2, 3 and 4 (Sondhi adhesive). There was no statistically significant difference between the groups ($p > 0.05$).

Table II shows the results of SBS tests of groups 5, 6, 7 and 8 (Concise adhesive). There was no statistically significant difference between the groups ($p > 0.05$).

Table III shows ARI scores for groups 1, 2, 3 and 4 (Sondhi adhesive). There was no statistically significant difference between the groups ($p > 0.05$).

Table IV shows the ARI scores for groups 5, 6, 7 and 8 (Concise adhesive). There was no statistically significant difference between the groups ($p > 0.05$).

Discussion

Nanoparticles have recently been included in dental composite resins to improve their mechanical properties.[5,20] Meguid[23] reported that resin debonding characteristics were positively affected by varying the percentage weight of nanofillers. Argueta-Figueroa et al.[24] reported that SBS was significantly higher

Table III. ARI scores for groups 1, 2, 3 and 4 (Sondhi).

Group	Score 0	Score 1	Score 2	Score 3
1 (Sondhi Control)[a]	16	1	1	2
2 (0.5% Sondhi)[a]	15	1	2	2
3 (0.25% Sondhi)[a]	12	3	1	4
4 (0.05% Sondhi)[a]	14	2	2	2

[a] Similar letters indicate no statistically significant differences between groups ($p > 0.05$).

Table IV. ARI scores for groups 5, 6, 7 and 8 (Concise).

Group	Score 0	Score 1	Score 2	Score 3
5 (Concise Control)[a]	14	2	1	3
6 (0.5% Concise)[a]	16	0	2	2
7 (0.25% Concise)[a]	12	4	1	0
8 (0.05% Concise)[a]	16	3	1	0

[a] Similar letters indicate no statistically significant differences between groups ($p > 0.05$).

for brackets bonded directly using an orthodontic adhesive with copper nanoparticles. In 2012, Turagam et al.[20] reported no polymerisation shrinkage of polymethyl methacrylate (PMMA) resin incorporated with 0.5 wt% of CN, and a significant reduction in 0.25 wt% and 0.125 wt% concentrations. Similarly, Yeung et al.[25] reported lower polymerisation shrinkage in PMMA resins incorporated with CN compared with unmodified PMMA resins. Lewis and Mladsi[26] reported improved stiffness and tensile strength of PMMA resin incorporated with 0.5 wt% of CN compared with the incorporation of 0.25 wt% and 0.125 wt%.

In adhesion studies,[27,28] bovine enamel has been shown to be a reliable substitute for human enamel, since both have similar composition and physical properties. In addition, the low cost and availability made the bovine incisors the teeth of choice for this study.

CN were included in the adhesive base pastes, because the catalyst paste contains benzoyl peroxide, which triggers polymerisation and, in contact with water, forms original acids/alcohols, which would block the reaction.[17] No reports in the literature have demonstrated changes due to the addition of CN to a dental adhesive.

The use of full-arch trays for indirect bonding might result in non-uniform positioning of the tray and, consequently, inconsistent adhesive layers,[29] which might affect the bond strength at the bracket/enamel interface.[14] When filler particles are included, thicker composite layers could be used over tooth surfaces.[5] It has been reported that different CN concentrations incorporated into resins leads to an improvement in various mechanical properties, such as impact strength, tensile strength and stiffness, as well as reduced polymerisation shrinkage.[30] Furthermore, the addition of CN increases the viscosity and the elastic melt properties of the composites.[7] Accordingly, the addition of CN was carried out using two common types of adhesive used in indirect bracket bonding.

There were no statistically significant differences in SBS between groups 1, 2, 3 and 4 (Table I). There were also no statistically significant differences between groups 5, 6, 7 and 8 (Table II). The SBS values obtained in the eight groups of the present study ranged from 6.9 MPa to 9.9 MPa. Reynolds[31] estimated that 5.9 to 7.8 MPa was required for satisfactory orthodontic adhesion; therefore, the SBS values obtained in the present study were considered adequate. Nevertheless, the minimum in vitro bond strength necessary for a reliable orthodontic bond is still unknown.[3] Although CN are considered the best material to reinforce resins,[8] the addition of CN investigated in the present study did not result in an increase in SBS for the two indirect bonding adhesives (Tables I and II).

Transfer trays for five teeth were used in an attempt to reproduce the clinical conditions, in which full-arch trays are invariably used.[12] However, the use of full-arch trays often leads to difficulties in uniform seating over areas under which the brackets are located.[3,32]

There were no statistically significant differences in ARI scores between groups 1, 2, 3 and 4 (Table III). There were also no statistically significant different scores between groups 5, 6, 7 and 8 (Table IV). Most test specimens had scores of 0 or 1, which is similar to the results reported by Kanashiro et al.[3] and Linn et al.[33] Low ARI scores may indicate that the addition of CN to the adhesives did not increase the bond strength at the enamel/composite interface.

The use of nanoparticles in dentistry has several advantages,[20,24] but the CN examined in the present study, at the filler amounts evaluated, did not improve the expected performance of the adhesives used in indirect bracket bonding. The search for improvement could be related to the use of other types of CN (multi-walled varieties), nanoparticles of different chemical elements (Cu, Ag, Au),[24] or even their incorporation into the composite resin used for bonding.

Conclusion

The addition of CN to the Concise and Sondhi adhesives, at the concentrations used, did not improve SBS or the amount of adhesive remnant remaining on enamel (ARI) following bracket removal.

Conflict of interest

The authors declare no conflicts of interest with respect to the authorship and/or publication of this article.

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