Effect of dentin surface modification using carbon nanotubes on dental bonding and antibacterial ability

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This study developed carbon nanotube coatings for the dentin surface and investigated the bonding strength and the in vitro antibacterial properties of carbon nanotube-coated dentin. Single-walled carbon nanotubes and multi-walled carbon nanotubes were first modified and then characterized using Fourier-transform infrared spectroscopy, scanning electron microscope, and transmission electron microscopy. Second, dentin samples were coated using either single-walled carbon nanotubes or multi-walled carbon nanotubes and observed under a scanning electron microscope. Then, the shear bonding strength and antibacterial properties of the dentin samples were tested. The results showed that both modified single-walled carbon nanotubes and multi-walled carbon nanotubes formed a stable coating on the dentin surface without affecting the shear bonding strength. Moreover, the antibacterial properties of the single-walled carbon nanotube-coated samples was obviously superior to those of the multi-walled carbon nanotube-coated samples. Consequently, single-walled carbon nanotube coating may be an antibacterial agent for potential application in the dental bonding field.

Keywords: Carbon nanotubes, Dentin, Bonding, Streptococcus mutans

INTRODUCTION

Carbon nanotubes (CNTs), a nanomaterial with unique properties, is attracting considerable attention. CNTs consist of a rolled-up sheet of graphite in single-walled (SWCNTs) or multi-walled forms (MWCNTs). CNTs can improve strength of composite materials, increase cell adhesion and proliferation and affect the nucleation of hydroxyapatite. Therefore, considerable attention has focused on the use of CNTs as biomedical materials in dentistry.

Moreover, CNTs have a strong attachment with collagen, which makes them a good candidate for tissue engineering scaffolding. Akasaka et al. found that MWCNTs could easily adhere to tooth slice surfaces after the slices were suspended in a CNT-dispersed solution, suggesting that CNTs have an excellent interaction with dentin collagen. Meanwhile, MWCNTs adhere uniformly to the surface of a collagen-covered medium, and SWCNTs can wrap tightly with collagen molecules. Collagen is the main organic component of dentin; it forms a fiber mesh structure for deposition, crystallization and the ordered arrangement of minerals. During the dentin bonding process, acid etching exposes the dentin collagen and allows the primer and adhesive resin monomers to permeate into the fiber mesh structure. However, collagen fibrils, which serve as the basic structure of dentin collagen, have gaps of only 20–30 nm. Priyanka’s study showed that the general adhesive resin monomer could not fully penetrate these gaps, which led to the formation of nanoleakage and the occurrence of secondary caries. Meanwhile, CNTs (which are 0.4–5.0 nm in diameter) can penetrate the gaps between collagen fibers. This evidence suggests the possibility of developing a CNT coating for the dentin surface. Moreover, in the study by Akasaka et al., the micro tensile strength test showed that CNT coating did not affect the immediate strength of dentin bonding; therefore, CNT coating may be further applied in dentin bonding.

Secondary caries, a bacterial disease, are common on the dentin bonding surface after restorative treatment. One reason for this high incidence is that resin composites and dentin bonding systems are not able to prevent secondary caries. Therefore, several trials have examined the possibility of adding antibacterial materials to the dentin bonding system. As a novel nanomaterial, CNTs also have antibacterial activity. In 2007, Kang et al. showed that Escherichia coli was killed by SWCNT suspension. Later, that group also found that highly dispersed MWCNTs had antibacterial activity. Numerous other researchers have confirmed that CNT suspension is a potential antimicrobial agent. Bai et al. found that CNT suspensions could inhibit the activity of Streptococcus mutans (S. mutans), which is the main cariogenic bacterium. Furthermore, SWCNTs in water can penetrate S. mutans and kill the cell, and MWCNT suspension also had antibacterial activity against S. mutans. These findings raise the possibility of adding CNTs to the dentin bonding system as an antibacterial. Nevertheless, to our knowledge, there are few reports regarding the antibacterial activity of CNTs applied to the dentin surface. Therefore, in this study, we coated the dentin surface with SWCNTs.
and MWCNTs to investigate whether they could serve as antibacterial agents when applied to the dental bonding.

The present study examines the use of SWCNT and MWCNT coatings on dentin surfaces. These coating may have antibacterial activity without affecting the immediate shear strength of dentin bonding. In this study, the effects of these coatings on dentin bonding and \textit{S. mutans} were investigated. It is expected that CNT coatings could be applied to the dental bonding system as antibacterial agents.

**MATERIALS AND METHODS**

### The modification and characterization of CNTs

SWCNT powder with an average particle diameter of 1–2 nm and an average length of 5–30 mm and MWCNT powder with an average particle diameter of <8 nm and an average length of 1–10 mm (Chengdu Organic Chemicals, Chengdu, China) were calcined in a porcelain furnace to remove carbon impurities (SWCNT: 440°C, 30 min; MWCNT: 400°C, 1 h). Then, a solution of 10 mL 70% HNO\textsubscript{3} and 40 mL 98% H\textsubscript{2}SO\textsubscript{4} was used to dissolve the SWCNT powder to remove metal impurities. After ultrasonic treatment (200 W, 40°C, 2 h), the solution was diluted with deionized water and centrifuged (3,000 rpm, 10 min) and the precipitates was washed with 4 mol·L\textsuperscript{-1} NaOH and centrifuged again; this process was repeated three times. A mixture of 30 mL 98% H\textsubscript{2}SO\textsubscript{4} and 10 mL 30% H\textsubscript{2}O\textsubscript{2} was then added to the obtained precipitates to activate the CNTs. The suspension was stirred overnight at room temperature and centrifuged. The precipitates were cleaned and diluted with deionized water, and the pH was adjusted to 7. After ultrasonic treatment (200 W, 40°C, 2 h) and dialysis for 24 h in deionized water, an SWCNT suspension was obtained. The MWCNT suspension was prepared in the same way. The two types of suspensions were further centrifuged, filtered, and washed twice, the obtained precipitates were lyophilized, and the powders were used for the following experiments.

Unmodified SWCNT powder, unmodified MWCNT powder, the above-mentioned modified SWCNT powder and the above-mentioned modified MWCNT powder were dispersed in deionized water, and the optical density (OD) of these suspensions was measured with a UV-vis spectrophotometer operating at 600 nm. The phase compositions of the unmodified SWCNTs, unmodified MWCNTs, modified SWCNTs and modified MWCNTs were determined using Fourier-transformed infrared spectroscopy (FTIR, MX-1E, Nicolet, Madison, WI, USA). The morphology of the unmodified SWCNTs, unmodified MWCNTs, modified SWCNTs and modified MWCNTs was characterized using a scanning electron microscope (SEM, KYKY-2800 microscope, KYKY Technology Development, Beijing China). Transmission electron microscopy (TEM, FEI, Hillsboro, OR, USA) was adopted to analyze the particle size. (For the unmodified SWCNT and unmodified MWCNT, the supernatants of them were used for the spectroscopic and morphological observations.)

#### Interaction of CNTs with collagens

Type I collagen fibers (MP, Billerica, MA, USA) were purchased. Twenty milliliters HCl solution (13 mmol/L), 20 mL HCl solution (13 mmol/L) with 20 mg modified SWCNTs, and 20 mL HCl solution (13 mmol/L) with 20 mg modified MWCNTs were sonicated at 200 W for 30 min. Each of the three solutions obtained (labeled the control group, SWCNT group and MWCNT group) were added to 3 mg type I collagen fibers and then activated (stirring speed: 300 rpm) for 3 h. Subsequently, the collagen fibers were removed from the three solutions, dried at room temperature and observed under SEM.

#### Preparation and characterization of CNT-coated dentin samples

Sixty intact molars or premolars extracted because of periodontitis were collected. This study was performed with the approval of the ethical committee of the West China Hospital of Stomatology, Sichuan University. The teeth were cut into 6×4×2 mm cubic samples to expose the dentin surface. Then, the dentin samples were etched with 32% phosphoric acid (Bisco, Schaumburg, IL, USA) for 30 s to expose the dentin collagen network.

The dentin samples were labeled the control group, SWCNT group and MWCNT group. The samples in the SWCNT group and MWCNT group were soaked and stirred (stirring speed: 300 rpm) for 1 h in the modified SWCNT water suspension or the modified MWCNT water suspension at room temperature for 3 h and then dried in the air. The control group received no treatment. Finally, the surface morphology of 3 randomly selected samples from each group was observed under SEM after being coated with gold particles to improve conductivity.

#### Shear bond strength (SBS) test of the dentin bonding samples

As mentioned above, the dentin samples in the SWCNT group and the MWCNT group had been etched with 32% phosphoric acid and soaked in the modified SWCNT water suspension or the modified MWCNT water suspension. The dentin samples of the control group only received acid etching. Then, all dentin samples were air-dried. Subsequently, randomly selected samples (n=15) from each group were bonded with composite resin (Coltene, Altstätten, Switzerland). The bonding procedure was performed as follows: First, Non-Rinse Conditioner (Coltene) was brushed uniformly on the dentin surfaces. Second, adhesive A and B (1:1) mixture (Coltene) was applied uniformly to the surface and then light-cured for 5 s. Third, composite resin was placed on the dentin bonding surface to form a resin cube, the thickness of which was 2 mm. After trimming, the resin was light-cured for 20 s and subsequently polished.

The prepared samples were fixed in a universal testing machine (LY-1066A, Shenzhen Bory Technology Service, Shenzhen, China) and then a SBS test was performed. The SBS test was performed using a universal testing machine (LY-1066A, Shenzhen Bory Technology Service, Shenzhen, China) and then a SBS test was performed.
performed. A blade loader (loading speed: 1 mm/min) was loaded on the sample until the sample broke. The maximum loads were recorded, and the SBS values were obtained (Formula 1).

Formula 1: \[ \tau = \frac{F}{S} \] \[ \tau: \text{SBS (MPa)}; F: \text{the maximum load value (N); S: the bonded area of dentin surface and composite resin (mm²)} \]

**Antibacterial testing of the CNT coating**

1. Cultivation and observation of *S. mutans* attached to dentin samples
Dentin samples (n=3) from each group and 500 mL liquid Brain Heart Infusion (BHI) medium (Shanghai Biochemicals, Shanghai, China) containing 4% (mass percentage) sucrose were prepared and sterilized. Cultures of *S. mutans* were grown on nutrient agar overnight, and one to two colonies were transferred from the culture into 2 mL BHI liquid medium to obtain an initial OD of 0.1 at 660 nm (OD<sub>660</sub>). One microliter of the suspension was inoculated into 14 mL BHI liquid medium corresponding to approximately 3x10<sup>7</sup> colony-forming units (CFU) per mL. The obtained suspension and the dentin samples were added to sterilized bacteria culture tubes (denoted as control group, SWCNT group, and MWCNT group). The culture tubes were cultivated in anaerobic incubation at 37°C for 48 h. Then, the dentin samples were washed with phosphate buffer saline (PBS), fixed with 2.5% glutaraldehyde overnight and sequentially dehydrated with 30, 40, 50, 60, 70, 80, 90 and 100% alcohol. Finally, the surfaces of the samples were observed under SEM.

2. Determination of viable cell numbers by plating method
The number of viable *S. mutans* cells on the CNT coatings was determined using a CFU assay. Briefly, *S. mutans* cells on the dentin samples (n=3) of three groups after cultivation for 48 h were scraped and introduced into 1-mL micro centrifuge tubes. Two hundred microliters PBS was added into the tubes and then the tubes were kept rotating on a shaker at 200 rpm for 20 min at room temperature. A 100-µL aliquot from each tube was diluted serially with 900 µL PBS, and the dilution was spread evenly onto solid BHI-agar plates for anaerobic cultivation at 37°C. After 48 h, the number of colonies on each plate was counted.

3. Fluorescence imaging
Dextran conjugate stain (Alexa Fluor647, Invitrogen, Life Technologies, Eugene, OR, USA) and green fluorescence nucleic acid stain (Syto9, Invitrogen, Life Technologies) were used to image the activity of *S. mutans*. Dextran conjugate stain is a bright, far-red-fluorescent dye with excitation ideally suited for the 594 or 633 nm laser lines, and the peak emission wavelength is 665 nm. It can be conjugated with a variety of antibodies, peptides and proteins for cellular labeling and detection. The green fluorescence nucleic acid stain is an excellent green-fluorescent nuclear and chromosome counterstain that permeates both prokaryotic and eukaryotic cell membranes. It has a high affinity for DNA and exhibits enhanced fluorescence upon binding, with an excitation maximum of 483 nm and a fluorescence emission maximum of 503 nm. It is particularly useful as a nuclear counterstain for bacterial assays since it stains both live and dead gram-positive and gram-negative bacteria. The dentin samples (n=3) were added to 3 different sterilized bacteria culture tubes (marked control group, SWCNT group, and MWCNT group), each of which contained 15 mL *S. mutans* suspension, corresponding to approximately 3x10<sup>8</sup> CFU per mL, and 1 µL diluted dextran conjugate stain (1 mg/100 µL). The three tubes were cultivated at 37°C under aerobic conditions for 48 h. Later, the dentin samples were washed, and 100 µL diluted (1:100) green fluorescence nucleic acid stain was added to the surfaces of the samples. After staining for 15 min, the samples were washed and observed with a confocal scanning laser microscope (CSLM, Leica, TCS SP2, Wetzlar, Germany).

4. Statistical analysis
The data were analyzed with the SPSS 16.0 software (SPSS, Chicago, IL, USA). One-way analysis of variance and the Student-Newman-Keuls test were used to determine the differences in OD value, SBS test and the number of viable *S. mutans* cells among the control group, SWCNT group and MWCNT group. The statistical significance set was at p<0.05.

**RESULTS**

**Characterization of CNTs**
As shown in Fig. 1, after ultrasonic treatment, the modified CNT suspensions were stable without agglomeration compared with the unmodified CNTs. Figure 1 also shows the dispersion ability of the modified CNTs in terms of UV-vis absorbance at the wavelength of 600 nm. With the modification, the dispersion activity of the SWCNTs and MWCNTs was higher than that of the unmodified CNTs.

Figure 2 shows the FTIR spectra of the unmodified CNT coatings.
SWCNTs, modified SWCNTs, unmodified MWCNTs and modified MWCNTs. The corresponding characteristic groups of the modified SWCNTs, such as C-O, C-C, C=O and O-H, were observed at 1,137, 1,634, 1,730 and 3,438 cm\(^{-1}\), respectively. Similarly, the corresponding characteristic groups of the modified MWCNTs, such as C-O, C-C, C=O and O-H, were observed at 1,122, 1,635, 1,731 and 3,438 cm\(^{-1}\), respectively.

Figure 3 presents the SEM and TEM images of the unmodified SWCNTs, modified SWCNTs, unmodified MWCNTs and modified MWCNTs. As shown, the unmodified SWCNTs and unmodified MWCNTs accumulated large numbers of non-uniform tubular particles. In contrast, the modified SWCNTs and modified MWCNTs had more uniform tubular particles with fewer agglomerations. However, the distribution of the SWCNTs was more uniform than that of the MWCNTs. Furthermore, TEM characterization also suggested that the unmodified SWCNTs and unmodified MWCNTs accumulated easily, even after ultrasonic
treatment, but the modified SWCNTs and MWCNTs were uniformly dispersed. Furthermore, the number of tubular structures in the modified SWCNTs was greater than that in the modified MWCNTs.

Interaction of CNTs with collagens
The configuration of type I collagen in the control group was well defined, and light and dark stripes were visible (Fig. 4A). In the SWCNT group, the collagen fibers were completely wrapped by the SWCNTs, and the stripe structures of the collagen were almost invisible (Fig. 4B). Compared with the SWCNT group, the MWCNTs did not uniformly wrap around the collagen fibers, and partial stripe structures could be observed (Fig. 4C).
Table 1 Shear bonding strength test results of dentin samples of the three groups (n=15)\(^a\)

| Parameters                  | Sample                      |
|-----------------------------|-----------------------------|
| Shear bonding strength (N/mm\(^2\)) | Control group | single-walled NT group | multi-walled NT group |
| 24.72±1.10                  | 23.84±1.69                 | 24.10±1.37              |

\(^a\)The data are expressed as mean±standard deviation (SD).

The surface morphology of CNT-coated dentin samples
As seen in Fig. 5, in both the SWCNT group and MWCNT group, tubular SWCNTs and MWCNTs covered the dentin surface. However, in higher magnification images, the number of tubular structures on the SWCNT-coated dentin sample was greater than that on the MWCNT-coated dentin sample, and the distribution of SWCNTs on the dentin surface was more uniform than the distribution of the MWCNTs.

SBS test
As shown in Table 1, the results of the SBS test indicated that the mean value of the control group (M=24.72, SD=1.10) did not significantly differ from that of the SWCNT group (M=23.84, SD=1.69) or the MWCNT group (M=24.10, SD=1.37). Meanwhile, there was no significant effect of SWCNT coating or MWCNT coating on dentin bonding at the \(p<0.05\) level for the three groups \([F(2,33)=1.238, p=0.303]\).

Antibacterial activity of CNT coating
The antibacterial activity of the CNT coating was first investigated by observing the adhesion of \(S.\) mutans. Figure 6 shows that a large number of \(S.\) mutans cells attached to the dentin surface of the control group and the MWCNT group. However, the number of \(S.\) mutans cells attached to the SWCNT group was few, and the cell volume was also smaller than that of the other two groups. This phenomenon was further confirmed by the CFU assays. Figure 6D shows that the number of viable cells in the SWCNT group was decreased compared with the control group, and the mean value of the MWCNT group did not significantly differ from that of the control group. In particular, the viable cell numbers attached to the SWCNT coating represented a 5.5-log reduction.

To confirm the reliability of the CFU assays, fluorescent dyes, as an indicator of \(S.\) mutans activity, were used to assess the antibacterial activity of CNT coating against \(S.\) mutans. The bacterium with strong activity were appeared as red. The CSLM images showed that the red areas of the control group and the MWCNT group were clearly larger than that of the SWCNT group, and there were no significant differences between the control group and the MWCNT group (Fig. 7).

DISCUSSION
The unique properties of CNTs make them widely used in the field of dentistry; in particular, CNTs can penetrate the dentin’s collagen fiber network\(^6\). Meanwhile, coating technology is a common method used to apply CNTs in the biomedical field\(^10\). Therefore, this study attempted to introduce CNT coating into the dentin bonding system.

Good dispersion of CNTs is the basis of uniform coating, but unmodified CNTs are intrinsically inert, often aggregate or entangle, and contain impurities\(^20\). In general, some surface-active groups can be grafted onto the surface of CNTs to increase dispersion\(^21\). Impurities in the CNTs may also affect dispersion\(^10\). In this study, purchased CNTs were calcined in air to remove impurities, treated with acid and dialyzed\(^22\); the resulting CNTs had good dispersion (Figs. 1 and 3). FTIR is an important tool for characterizing the functional
groups on CNTs\textsuperscript{20}. The peaks of the modified SWCNTs and modified MWCNTs shown in Fig. 2 were most likely caused by the formation of carboxylic acid groups, which contributed to the good dispersion of CNTs\textsuperscript{23}.

It is noteworthy that there was a difference between SWCNTs and MWCNTs when they directly interacted with collagens; specifically, SWCNTs had a stronger affinity for collagen than MWCNTs (Figs. 4B and C). A plethora of in vitro evidence shows that CNTs have good interaction with collagens\textsuperscript{24,25}. In our experiment, SWCNTs showed better interaction with collagens than MWCNTs. This might be due to the smaller diameter of SWCNTs, which have a larger superficial area with more grafted reactive groups\textsuperscript{26}.

In our experiment, the etched dentin samples were soaked and stirred in a CNT suspension to obtain stable coatings on the surfaces. However, the distribution of the SWCNT coating was more uniform than that of the MWCNT coating (Fig. 5). This phenomenon might be the result of the structure of CNTs\textsuperscript{20}. The walls of MWCNTs are thin and easily broken compared with SWCNTs, so after modification with high temperatures and strong acid, the MWCNT coating showed few tubular structures and low dispersion\textsuperscript{20}.

The SBS test is the most commonly used dentin bond strength test\textsuperscript{27}. Some scholars have argued that the in vitro SBS test is affected by many factors, such as unnecessary stresses; thus, the results are poorly relative to clinical situations\textsuperscript{28}. However, the relative values among different treatment groups are worthy of reference\textsuperscript{28}. This study illustrated that CNT coating did not impact the SBS of dentin bonds. Therefore, SWCNTs and MWCNTs may be added to the resin matrix like other nanomaterials to exert particular effects on dentin bonding\textsuperscript{29}.

Many foreign scholars have found that a CNT suspension could inhibit the activity of \textit{S. mutans}\textsuperscript{13,17,18}, while the results shown in Figs. 6 and 7 suggest that the activity of \textit{S. mutans} could be inhibited by SWCNT coating on the dentin surface but not by MWCNT coating. The reason may be the destruction of the tubular walls of MWCNT when they are modified, as stated above\textsuperscript{20}. However, tubular SWCNTs were unbroken and were distributed uniformly on the dentin surface, which gave SWCNTs more contact with \textit{S. mutans}. As a result, the number of \textit{S. mutans} cells attached to the dentin samples in the SWCNT group was much lower than that of the other groups (Fig. 6). Similarly, Fig. 7 suggests that
the activity of *S. mutans* could be inhibited by SWCNT coating on the dentin surface but not by MWCNT coating. In brief, these results indicate that the colonization of *S. mutans* can be inhibited by SWCNT coating. However, to further determine the antibacterial activity of MWCNTs, more studies are needed.

**CONCLUSION**

In summary, by means of calcination, acid treatment and carboxylation, we successfully fabricated CNTs with good dispersion. SWCNTs showed better interaction with collagens than MWCNTs, and both SWCNTs and MWCNTs formed a stable coating on the dentin surface. Moreover, SWCNT coating and MWCNT coating do not affect the immediate shear strength of the dentin bond. However, the colonization and activity of *S. mutans* on the dentin surface were only inhibited by the SWCNT coating. Consequently, our study indicates that SWCNT coating may be a potential antibacterial material when applied to dental bonding.

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