

ORIGINAL ARTICLE

Application of Titanium Dioxide Nanotubes to Tooth Whitening

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Synopsis

The titanium oxide nanotube (TNT) is a novel high-functional material with photocatalytic performance inherent to titanium oxide improved by combining the diverse properties of the specific nanotube structure and titanium oxide crystal. Basic research was conducted on TNT for its application as a dental bleaching agent.

TNT was chemically synthesized, and the amount of generated radicals was determined by the ESR method. In addition, the oxidation-reduction reaction on UV irradiation was investigated using methylene blue to examine photocatalytic properties. As a result, TNT generated a larger amount of radicals than TiO₂, and showed improved photocatalytic properties. Furthermore, a bleaching experiment was conducted using a colored enamel model. Then, sufficient bleaching effects were demonstrated with visible light, suggesting the potential application of TNT to dental bleaching.

Key words: whitening, titanium oxide nanotube (TNT), TiO₂

Introduction

With a growing awareness of esthetics in clinical settings, an increasing number of patients are requesting orthodontics and tooth whitening. The demands for tooth whitening have also markedly increased. However, hydrogen peroxide (H₂O₂) is used at a high concentration (30-35%) in the office bleaching method, which raises concerns about hyperesthesia [1, 2] and its adverse effects on dental hard and periodontal tissues [3-7]. To minimize these adverse effects, titanium dioxide (TiO₂), used in white pigments, cosmetics, paint, and food additives, has been applied to tooth whitening [8-10].

TiO₂ is less expensive because it is abundant in nature, and is safe for the human body because it is chemically stable. TiO₂, used as a photocatalyst, causes a strong redox reaction through holes and electrons generated by light absorption. TiO₂ becomes super-hydrophilic when coated as a membrane. In the presence of water, multiple types of radicals are known to be generated by the redox power of a photocatalyst [11]. TiO₂ absorbs light above band gap energy (approximately 3.1 eV), and electrons are excited to the conduction band. The excited electrons reduce oxygen, generating superoxide radicals, such as O₂⁻. The holes generated in the valence

band reduce hydroxide ions, generating hydroxyl radicals (Figure 1). These radicals have been shown to discolor tooth-coloring organic compounds through oxidation and degradation [12]. This is a principle of tooth whitening. The properties of functional materials, such as photocatalysts, are greatly influenced by reactions with surface-adsorbed molecules. Thus, materials with smaller particle sizes and larger specific surface areas exhibit markedly better properties. Studies to identify new functionalities have recently been actively conducted by utilizing nanosized and other forms of materials [13-16]. A representative material is the titanium oxide nanotube (TNT) [17]. TNT has an excellent photocatalytic ability and physicochemical functions through specific co-existing nanotube structures and diverse titania crystals [18].

The purpose of the present study was to utilize TNT to develop a highly efficient tooth whitening agent that exerts high performance in a short time at a low H₂O₂ concentration. The amounts of radicals generated in TNT and the photocatalytic properties and whitening effects of apatite pellets were herein investigated.

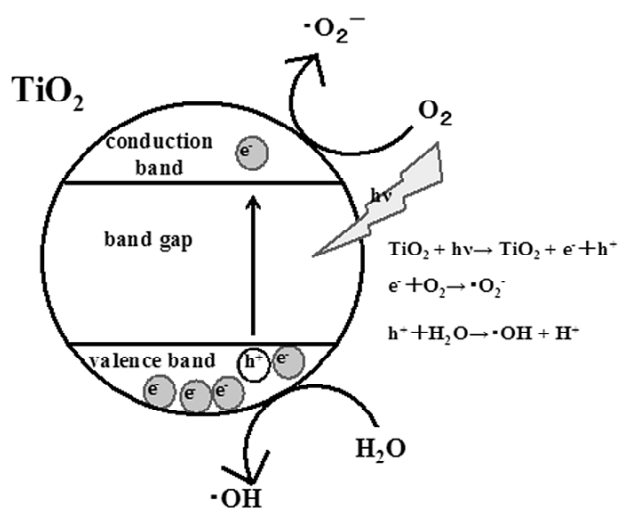


Fig. 1 Photocatalytic action of titanium oxide

Materials and Methods

1. TNT synthesis

TNT was chemically synthesized as reported by Kasuga *et al.* [17] (Figure 2). One gram of commercially available TiO₂ (P-25, NIPPON AEROSIL CO., LTD., Tokyo, Japan) was added to 100 mL of 10 M/L aqueous solution of sodium hydroxide and stirred under reflux for 24 hours at 110°C. The resulting slurry was subjected to solid-liquid separation by suction filtration. Ion-exchanged water was subsequently added to wash the powder by suction filtration. These procedures were repeated until the filtrate conductivity reached <70 μS/cm. Suction filtration was then conducted with 0.1 M/L hydrochloric acid, followed by washing with deionized water until conductivity reached 5 μS/cm in order to remove the sodium used in the reaction solution. The resulting powder was dried to obtain TNT. Half of the powder was heated at 400°C for 2 hours in the atmosphere to prepare two types of TNT.

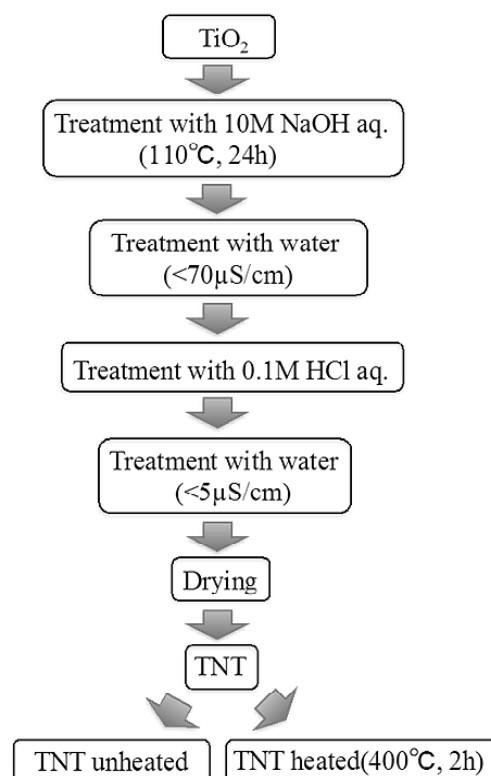


Fig. 2 Synthesis of TNT

TNT was synthesized by a low temperature chemical synthesis method. The TNT collected was unheated or heated for 2 hours at 400°C in the atmosphere.

The resulting TNT was examined by transmission electron microscopy (TEM) and X-ray diffraction (XRD).

2. Measurement of radical generation

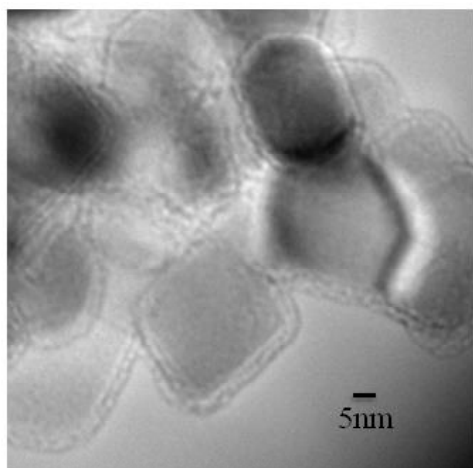
The amounts of radicals generated in three samples (commercially available TiO₂ and unheated and heated synthesized TNT) were measured by electron spin resonance (ESR).

Each of the TiO₂ and unheated and heated TNT samples was added into a glass tube, followed by UV irradiation from the side at 1-minute intervals for 10 minutes to measure the amounts of radicals in each sample with the ESR equipment (ESP350E, BRUKER Co., Germany). ESR was measured under the following conditions: central magnetic field: 3400 G, magnetic

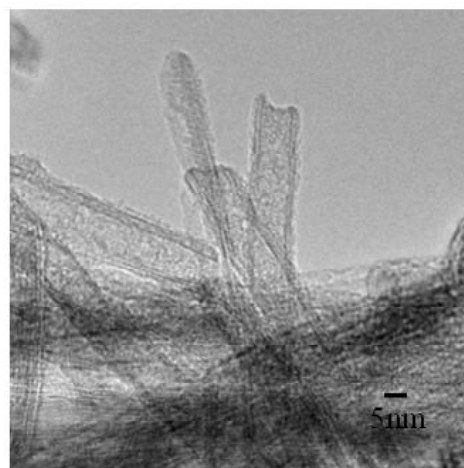
field sweep width: 400 G, microwave power: 9.48 GHz, 0.1 mW (TiO₂), 9.48 GHz, 0.025 mW (unheated and heated TNT), modulation frequency: 100 kHz, and ambient temperature: 20 K. An ultra-high pressure mercury lamp was used as the UV irradiation device (USH-1005D, Ushio, Tokyo, Japan) at a wavelength of 300-400 nm and illumination intensity of 10 mW/cm².

3. Evaluation of photocatalytic properties

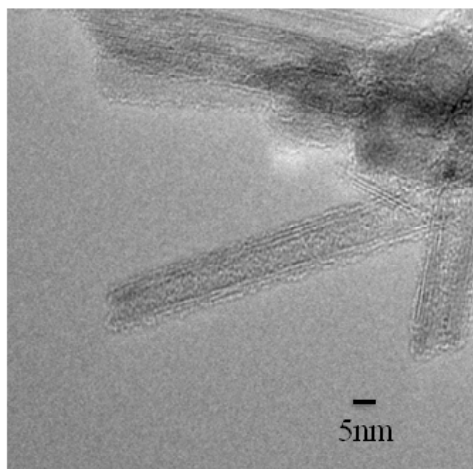
The photocatalytic properties of unheated and heated TNT were evaluated using methylene blue (MB). MB is a representative reagent used to examine redox reactions. Blue MB becomes colorless when reduced, allowing the reducing power of a catalyst to be determined [19].



(a) TiO₂



(b) TNT unheated



(c) TNT heated

Fig. 3 TEM images of TiO₂, TNT unheated and TNT heated TiO₂ particles accumulated (a). Both unheated and heated TNT accumulated with a tube structure (b), (c).

MB solutions (20 mg/L in 100 mL) were prepared. Ten milligrams of each sample powder was added to the MB solutions and stirred for 24 hours in the dark. Twenty-four hours later, the MB solutions were UV-irradiated to determine changes in absorbance in the solutions at specified time points using an ultraviolet-visible spectrophotometer (UV-mini1240, Shimadzu Corporation, Kyoto, Japan). The time points examined were 0, 5, 15, 30, 60, 120, 240, and 480 minutes. UV irradiation was conducted with a UV irradiation device (UVF-204S, SAN-EI ELECTRIC CO., LTD., Osaka, Japan).

4. Evaluation of whitening effects

Commercially available hydroxyapatite ceramics (APP-100, PENTAX, Tokyo, Japan) were immersed in a 5 g/L MB solution for two weeks to generate a virtually colored enamel model [20, 21]. Ten milligrams of each sample (TiO₂ and unheated and heated TNT) was added to 1 mL of 3% H₂O₂ and stirred to prepare a whitening reagent. In the present study, a commercially available TiO₂-containing tooth whitening agent with the lowest H₂O₂ concentration was used as a reference. The concentration of H₂O₂ was determined to be 3% to provide a highly efficient tooth whitening agent with a low H₂O₂ concentration. Each whitening reagent, prepared using a colored enamel model, was applied, followed

by light irradiation for 5 minutes using a dental LED irradiator (Cosmo Blue, GC, Tokyo, Japan). These procedures were repeated three times.

Color changes were measured with a dental colorimeter (ShadeEye[®] NCC, SHOFU, Kyoto, Japan). The CIEL*a*b* color system was employed for color measurements. Whitening effects were evaluated using the resulting L*, a*, and b* values and color difference ΔE^*_{ab} . The measurement was conducted four times (including once before whitening). A one-way analysis of variance was conducted for the color difference ΔE^*_{ab} , followed by statistical processing by Tukey's test ($p < 0.05$, $n = 5$).

Results

1. TNT synthesis

In the TEM observations, the starting material, TiO₂, had a particle size of approximately 25 nm and a tube structure with 10-nm outer and 7-nm inner diameters after being chemically synthesized (Figure 3). Even after heating at 400°C, the same tube structure as that of unheated TNT was observed (Figure 3).

In XRD, a peak was detected at a TNT-specific diffraction angle of 10°, demonstrating TNT synthesis. A peak nearing that of anatase TiO₂ was detected for heated TNT (Figure 4).

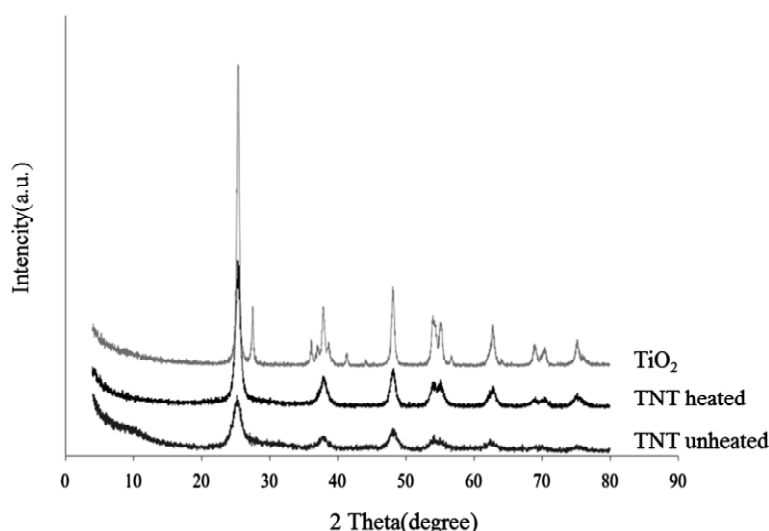


Fig. 4 XRD patterns of TiO₂, TNT unheated and TNT heated

The waveform peak of anatase type TiO₂ was detected for TiO₂.

A broad peak at a diffraction angle of approximately 10° was detected for unheated TNT, while a smaller peak was detected for TiO₂.

The broad peak at a diffraction angle of approximately 10° disappeared for heated TNT, nearing the peak of anatase type TiO₂.

2. Measurement of radical generation

Radical generation by UV irradiation was confirmed for all TiO₂ and unheated and heated TNT samples by the electron spin resonance method. The amounts of radicals generated at 10 minutes were 1.6×10^{15} and 2.6×10^{15} for TiO₂ and unheated TNT, respectively. The maximum amount of radicals generated for heated TNT was 9.0×10^{15} at 10 minutes, which was approximately 5-fold that for unheated TNT (Figure 5).

3. Evaluation of photocatalytic properties

Absorbance of approximately 2.2 before UV irradiation decreased to approximately 0.3 and 0.8 at 480 minutes after UV irradiation for unheated and heated TNT, respectively. The absorbance of an MB solution decreased for both unheated and heated TNT samples. The concentration of MB solution decreased more rapidly for heated TNT than for unheated TNT (Figure 6).

4. Evaluation of whitening effects

a* indicates the degrees of green and red colors in the minus and plus directions relative to the reference (0), respectively. b* indicates the degrees of blue and yellow colors in the minus and plus directions relative to the reference (0), respectively. In the graph of a* and b*, the value neared 0 (colorless) for all samples as the number of whitening increases (Figure 7). L* indicates brightness (0: black, 100: white). In the graph of L*, the value became larger and the color turned white for all samples as the number of whitening increased (Figure 8). ΔE^*_{ab} represents the color difference. In all samples, the color difference became larger as the number of whitening increased. Heated and unheated TNT showed significantly larger values than the controls and TiO₂ (Figure 9).

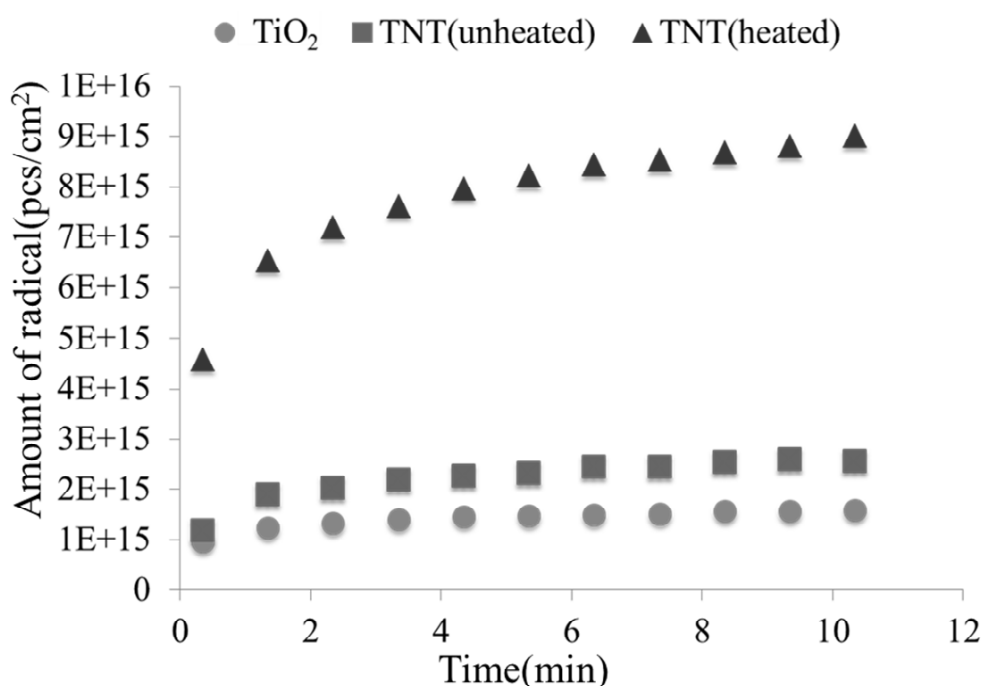
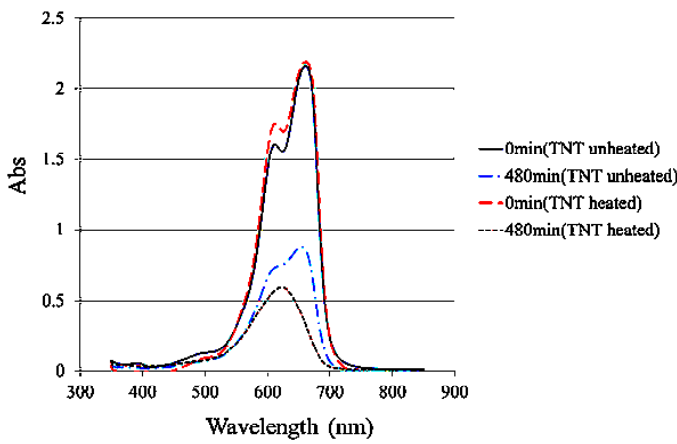


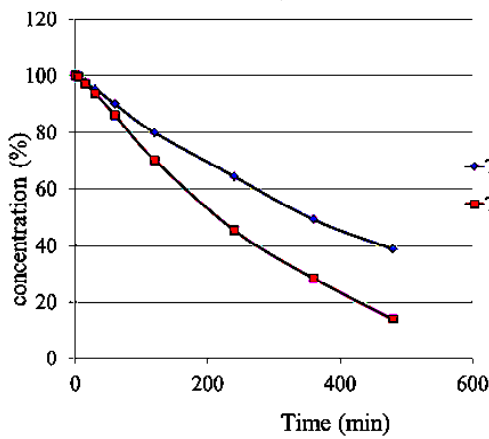
Fig. 5 Radical generation

Radicals were generated in each sample by UV irradiation.

TNT generated a larger amount of radicals than TiO₂. Heated TNT generated a significantly larger amount of radicals.



(a)



(b)

Fig. 6 Change of MB concentration

(a) Absorbance waveform of MB before and 480 minutes after light irradiation
 (b) Changes in MB by the measurement time at an absorbance 663 nm

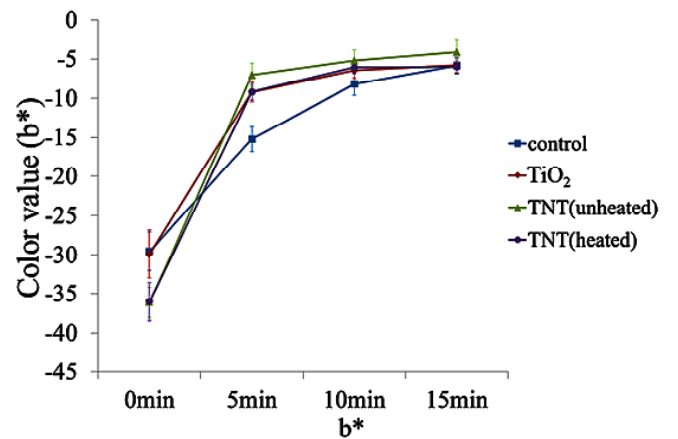
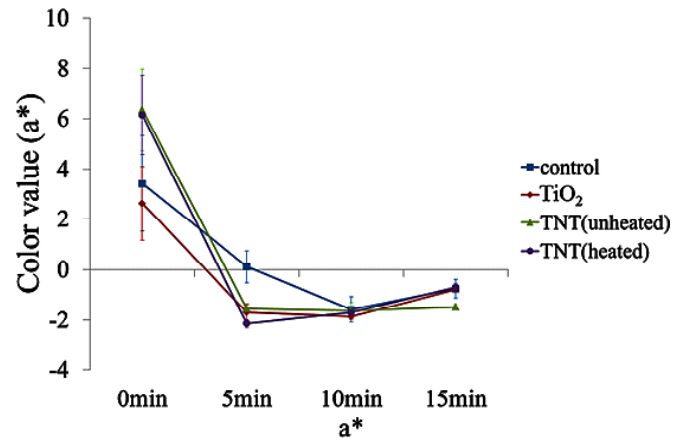


Fig. 7 Change of color value a* and b*

Change in a* for each sample after whitening. The value neared 0 with repeated whitening.
 Change in b* for each sample after whitening. The value neared 0 with repeated whitening.

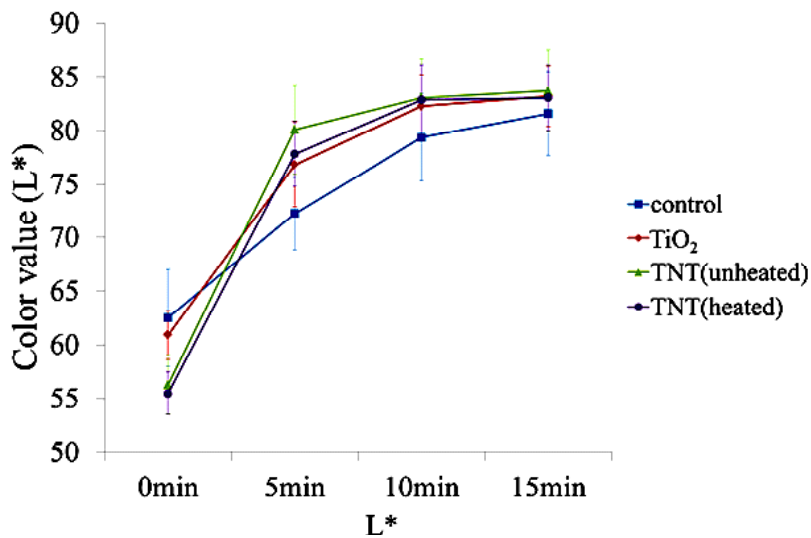


Fig. 8 Change of color value L*

Change in L* for each sample after whitening. The value increased with repeated whitening. The value changed more significantly for catalyst-containing samples than for the control.

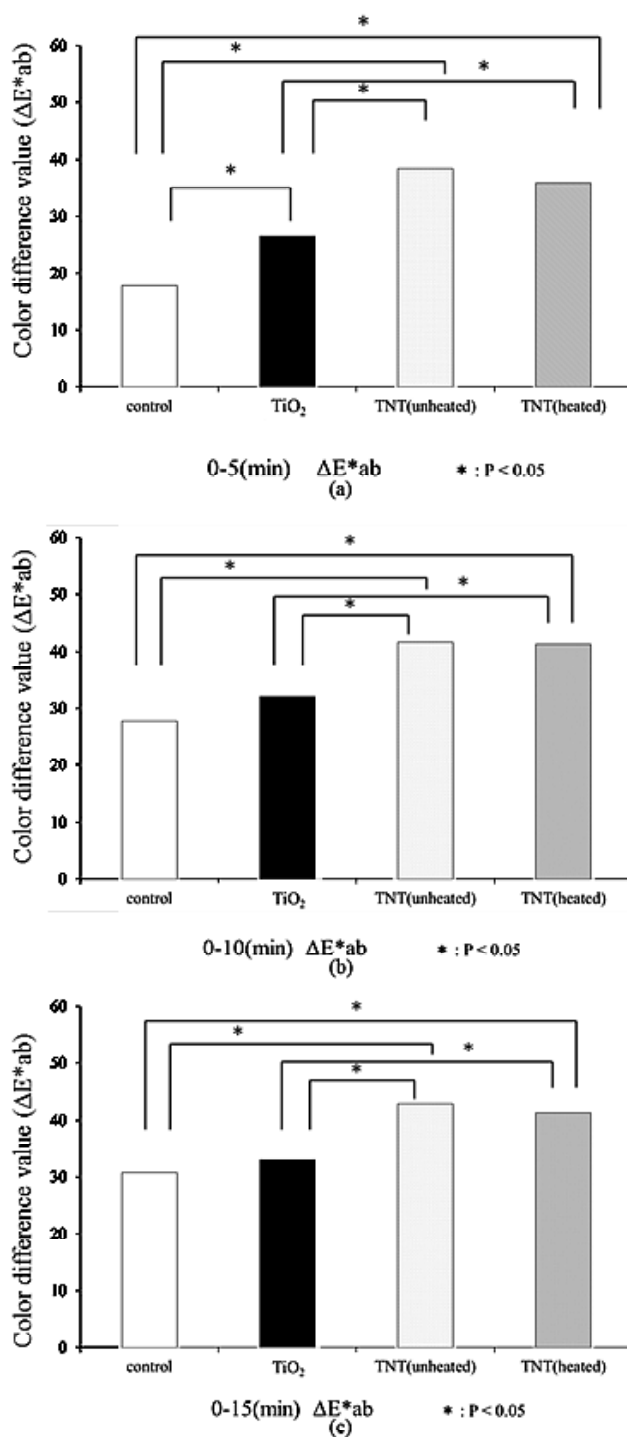


Fig. 9 Change of color difference value
 (a) Color difference ΔE^*_{ab} between before whitening and after the first whitening.
 (b) Color difference ΔE^*_{ab} between before whitening and after the second whitening.
 (c) Color difference ΔE^*_{ab} between before whitening and after the third whitening.

Discussion

TiO_2 is a photocatalyst that causes a redox reaction (i.e., radical generation) and decomposes organic compounds when light-irradiated [12]. TNT has a larger specific surface area than TiO_2 and light catalytic action due to its tube structure [17, 18]. Furthermore, TNT is an environment-friendly material that has adsorption effects and is applied in various fields. In this study, the application of TNT to tooth whitening was investigated.

1. TNT synthesis

Chemically synthesized TiO_2 was observed by TEM, showing the tube structure of TNT. The tube structure was maintained for heated TNT. TNT forms a layered sheet of TiO_2 through an amorphous formation with sodium hydroxide at a high concentration. The sheet is washed with deionized water to remove sodium ions between the layers and peel off the sheet. At that time, the sheet is rounded, forming a multilayer tube structure (Figure 10). The peak of XRD in TNT has been characterized by the multilayer part of the tubular structure at a diffraction angle of approximately 10° [17, 18]. A broad peak was detected at a diffraction angle of approximately 10° for unheated TNT. This peak demonstrated the synthesis of TNT. However, the peak at a diffraction angle of approximately 10° had almost disappeared for heated TNT, nearing the peak of anatase TiO_2 . This result suggested that

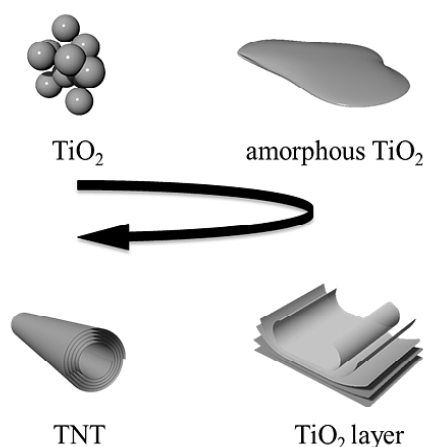


Fig.10 Formation of Titanium dioxide nanotube

TiO_2 forms a layered structure containing sodium ions between its layers through an amorphous formation with sodium hydroxide at a high concentration. The sheet is washed with deionized water to remove sodium ions between the layers and peel off the sheet, self-organizing a tube structure.

crystallinity was improved by the heat treatment and that the peak at a diffraction angle of approximately 10° was attenuated by the contraction of TNT layers, nearing the peak of anatase TiO_2 .

2. Measurement of radical generation

The ESR measurement demonstrated an ESR signal, which indicated the presence of unpaired electrons immediately after UV radiation, for TiO_2 and unheated and heated TNT. The analysis of g values demonstrated the generation of radicals ($\text{O}_2^{\cdot -}$). The amounts of radicals generated were measured, and the results obtained revealed that TNT generated a larger amount of radicals than TiO_2 .

Heated TNT generated 5-fold more radicals than unheated TNT, suggesting a significant increase due to the heat treatment. TNT has a larger specific surface area due to its tubular structure [17, 18]. TNT presumably had a larger specific surface area by taking a multi-layer tube structure, thereby absorbing light efficiently, and generated a larger amount of radicals than TiO_2 because of the increased number of reaction active sites for radical generation. Heated TNT generated a significantly larger amount of radicals. This was explained by heating increasing the crystallinity of TNT, which facilitated photoexcitation, and also that by the recombination of photoexcited electrons and holes being suppressed. The exciton lifetime due to the photoexcitation of TNT was examined previously, and the lifetime of radicals and electrons generated by the photoexcitation of TNT was found to be five- to ten-fold longer than that of typical titanium oxide particles [22]. This finding was attributed to TNT allowing the stable arrangement of active species, such as generated radicals, on the surface because of its long one-dimensional structure, suggesting the superiority of a specific one-dimensional nanostructure. The results obtained in the present study may have been derived from such unique nanostructure, suggesting the superiority of titania with a nanotube structure.

3. Evaluation of photocatalytic properties

To investigate the photocatalytic properties of TNT, a whitening experiment was conducted

with MB solutions. MB is a representative reagent used to examine redox reactions. Blue MB becomes colorless when reduced, thereby allowing the reducing power of a catalyst to be determined [19]. The photocatalytic properties of TNT were examined using a change in absorbance at 663 nm, a characteristic absorption peak of MB, as an indicator [19]. Peak absorbance before photoirradiation was shared by unheated and heated TNT. Peak absorbance 480 minutes after light irradiation was lower for heated TNT than for unheated TNT. This result suggested that heated TNT decomposed and decolorized MB, i.e., heated TNT exhibited higher photocatalytic activity than unheated TNT. This is plausible in view of the amount of radicals generated. Thus, heated TNT exhibited higher crystallinity and improved photocatalytic properties.

4. Evaluation of whitening effects

H_2O_2 is now commonly used for tooth whitening. The concentration of H_2O_2 used in the office bleaching method is as high as approximately 35%. Thus, hypersensitivity and its adverse effects on hard and periodontal tissues are areas of concern. H_2O_2 produces radicals when decomposed. Dental coloring materials are organic substances that become colorless when decomposed by the oxidation action of a radical [12]. This is the principle of tooth whitening. Thus, tooth whitening products that efficiently generate radicals even at a low concentration of H_2O_2 have recently been developed by utilizing TiO_2 as a photocatalyst [9, 10, 23, 24]. Thus, TNT was applied to H_2O_2 at a low concentration in order to examine whitening effects in an MB-colored enamel model. In all samples, both a^* and b^* values neared 0 with repeated whitening. The L^* value for brightness also increased during repeated whitening. The ΔE^*_{ab} , which indicates the degree of the color change, showed a significantly larger value for the TNT sample than for the control and TiO_2 samples. In this experiment, unheated TNT provided the highest whitening effects, and this was attributed to visible light being employed for the dental LED irradiator. TiO_2 of the anatase type generates electrons in the conductor through photoexcitation and holes in the valence band when irradi-

ated with ultraviolet light (<380 nm) at a band gap energy of >3.1 eV. This causes a redox reaction in the conductor and valence band, generating radicals. However, the central wavelength of the dental LED illuminator was 405 nm (3.06 eV). This generates light energy below the band gap energy of TiO₂. The applications of visible light responsiveness to TiO₂ and TNT are currently being investigated. Nitrogen- or metal ion-doped titanium oxide and TNT provide responsiveness to visible light with a wavelength of >380 nm [25-27]. Nitrogen- or metal ion-doped titanium oxide and TNT provide excellent responsiveness to visible light through the coloring of titanium oxide, as a result of energy levels formed in the presence of such dopants [28]. Chemical bonds, such as H₂O₂-derived Ti-OO-Ti, were previously shown to be formed on the TiO₂ surface in the presence of titanium oxide and H₂O₂, providing photocatalytic properties due to visible light [29]. In the present study, the properties of TNT may be improved by a reaction to visible light through the redox action of H₂O₂. Furthermore, unheated TNT had a larger specific surface area than heated TNT, and was more strongly influenced by the effects of H₂O₂ and more responsive to visible light, resulting in the highest whitening effects.

We herein demonstrated that TNT facilitated whitening with H₂O₂ at a low concentration. In addition, TNT mixed with H₂O₂ increased responsiveness to visible light. In the future, the mechanism of the TNT response to H₂O₂ will be elucidated in more detail, and if visible-light-responsive TNT becomes available, whitening agents that exert effects on visible light irradiation will be developed. Environmentally friendly light catalysts that efficiently utilize sunlight or room light will also be developed. The photocatalytic action of TiO₂ has been investigated for its applications to cancer treatment. The applications of TNT as a biomaterial should be developed, including for cancer treatments and DDS carriers utilizing a tube structure.

Acknowledgments

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