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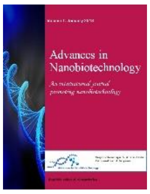

Design, synthesis and application of gold nanoparticles in tooth whitening process

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ABSTRACT

In this present work, gold nanoparticles coated with citrate was first synthesized and then it was used as a nanocomposite catalytic both for oxidizing and for reducing hydrogen peroxide (H_2O_2), to whiten teeth. Dyes were used to stain the tooth specimens for three days and subsequently bleached using H_2O_2 alone, H_2O_2 plus gold nanoparticles. The results of bleaching teeth with and without Luma care irradiation were compared.

1. Introduction

Nanotechnology is currently driving the dental materials industry to substantial growth, thus reflecting on improvements in materials available for oral prevention and treatment. New developments in nano-technology are applied to dentistry, focusing on the use of nanomaterials for improving the quality of oral care, the perspectives of research in this arena, and discussions on safety concerns regarding the use of dental nanomaterials [1-3].

Gold nanoparticles have recently been introduced in a variety of applications in diagnosis and treatment. Several studies reported the synthesis of gold nanorods, nanocages, nanoshells, and nanospheres [4], generating promising compounds to be used as contrast agents, radiosensitizers, photothermalagents, and drug delivery nanocarriers [5]. The main benefits are their low cytotoxicity and wide variety of possible chemical functionalizations. Nanotechnology has induced the generation of innovative and cost-effective dental materials and devices.

Dental materials have evolved with the advent of nanotechnological research focusing on the production and application of nanoparticles with high-quality structural characteristics. The incorporation of amyriad of nanoparticles into dental materials (e.g., quartz, colloidal silica, zirconia, zincoxide) represents an innovation by manufacturers to improve the chemical and physical properties of these materials [6]. This area of research has been coined ‘nanodentistry’, defined as the ‘science and technology of diagnosing, treating and preventing dental diseases, relieving pain, preserving and improving dental health using nanostructured materials [7].

Nanodentistry is used of polymeric (e.g., polyethyleneglycol, solid lipids, nanogels, dendrimers, chitosan), metallic (e.g., silver, gold, copper), and inorganic nano-based materials (e.g.,zirconia, silica, titanium dioxide, hydroxyapatite, quantumdots, nanocarbons).

Catalytic wet peroxide oxidation (CWPO) is based on the action of hydroxyl and hydroperoxyl radicals produced, in this case, upon catalytic decomposition of hydrogen peroxide under relatively mild operating conditions in the presence of a solid catalyst. Current trends are addressed to the development of supported and unsupported metal nanoparticles, being the most studied iron, manganese and gold nanoparticles [8].

Recent studies on the catalytic applications of gold nanoparticles show that CWPO is a promising area in that respect [9-12]. So far, gold nanoparticles deposited on hydroxyapatite [9], diamond [11] or activated carbon [12] have shown fairly high activity. However, frankly different efficiencies of hydrogen peroxide consumption were observed with those catalysts, which are attributed to the nature of the support, crucial for the activity of gold [12].

CWPO proceeds according to the following steps: (i) adsorption of hydrogen peroxide on the gold-support interface, (ii) production of hydroxyl and hydroperoxyl radicals mainly on the gold surface though in some extension also occurs on the carbon surface and (iii) reaction between hydroxyl radicals and other OH- molecules on the gold surface. According to the mechanism, hydrogen peroxide decomposition into hydroxyl and hydroperoxyl radicals in the presence of gold nanoparticles.

Recently, the work of Martín et al. [13], working with gold-on-nanosized diamond [14], demonstrated that small gold nanoparticles supported on diamond nanoparticles are an efficient catalyst in the sense of achieving adequate biodegradability of phenolic waste water with values of H_2O_2 to phenol molar ratio. Herein, an insight into the origin of the catalytic properties of gold in wet peroxide oxidation is accomplished.

Having beautiful white teeth is a goal of many people. According to the American Academy of Cosmetic Dentistry, more than half of the people undergoing orthodontics treatment programs want to change the color of their teeth, which indicates the level of demand by the general public for tooth whitening. Tooth color is determined by intrinsic and extrinsic factors. Intrinsic tooth color is associated with the light scattering and absorption properties of the enamel and dentine. Extrinsic color is associated with the absorption of materials (e.g., tea) onto the surface of enamel. Discolored teeth, especially in the anterior region, can result in considerable cosmetic impairment. In addition to invasive therapies, such as dental crowns and veneers, the noninvasive whitening of teeth is an alternative therapy. Tooth whitening is divided into two broad categories: in-office whitening, which is done by a dentist using high-concentration whitening agents, and at-home bleaching, done by the patient using lower-concentration whitening agents in special trays. For more than a decade, tooth whitening has been done in dental offices using products that are extensively based upon hydrogen peroxide chemistry.

Both clinical and laboratory studies [15–18] report that tooth whitening after a single power bleaching session using 35% concentration hydrogen peroxide (H_2O_2) is effective. There is clear evidence that, within 10–30 minutes at room temperature, the H_2O_2 molecules are able to penetrate to the dentine through faults in the enamel. Photoactivation might be an ideal vehicle for generating radicals since visible light can penetrate deep into the tooth [19-22]. To improve or accelerate the whitening process, several different types of irradiation sources, such as heat, light, and lasers, have been used to accelerate the professional bleaching procedure [23–25]. The low molecular mass of H_2O_2 and its subproducts favors its diffusion through mineralized dental tissue to reach the pulp chamber, where it causes oxidative stress and pulp inflammation, which trigger the most prevalent bleaching induced side effect: tooth sensitivity [26,27].

In-office whitening generally uses relatively high concentrations of bleaching agents (25–35% H_2O_2) on the teeth for a fast effect. Some studies have reported superficial enamel alterations promoted after a bleaching session with 35% H_2O_2 [28,29]. Some recent approaches use a direct-current cold atmospheric pressure plasma microjet (PMJ) increased the production of hydroxyl (OH) radicals to improve tooth whitening [30,31].

Future technologies for whitening teeth might involve using activating agents to improve the performance of H_2O_2.

Gold nanoparticles (GNPs), a new class of two-dimensional carbon nanostructure, has recently been extensively studied because of its high conductivity, large surface-to volume ratio, and superior chemical and mechanical stability.

In this work, we synthesized gold nanoparticles (GNPs). We used these nanoparticles as a catalyst for tooth bleaching and compared its efficacy with that of whitelight irradiation in the present study.

2. Materials and Methods

2.1. Reagents and Instruments

UV-visible (UV-vis) absorption spectroscopic measurements were recorded on a UV-vis spectrometer (Agilent Cary 100), using quartz cells of 1 cm path length and water as the reference solvent at room temperature. Transmission electron microscopy (TEM) was performed using a Zeiss EM 900 and used to investigate morphology and size of GNPs. The concentrations of GNPs in µg/ml were measured by the inductively coupled plasma optical emission spectrometry (ICP-OES). The surface charge was determined by a ZEN 3600 nanosizer (Malvern, UK). The hydrodynamic size of the GNPs was measured via Dynamic Light Scattering method (DLS). Incandescent lamps (wavelength: 310 nm) to lightactivate bleaching products were in a photochemical reactor (Systems PR-2000; Panchum, Taiwan).

Reagents required for the synthesis were purchased from Sigma-Aldrich. All the chemical compounds used in this research were purchased from Merck, Germany and Fluka, Switzerland. Hydrogen tetrachloroaurate (III) trihydrate ($HAuCl_4.3H_2O$, 99.5% purity) and sodium citrate dihydrate ($C_6H_5Na_3O_7.2H_2O$) were purchased from Merck. The commercial tea was purchased at a local retail grocery. All other reagents were locally purchased and were of analytical grade.

2.2. Synthesis of gold nanoparticles (GNPs)

Gold nanoparticles (GNPs) were synthesized as follows [5]: all glassware was cleaned in aqua regia (3 parts HCl, 1 part HNO_3), rinsed with H_2O, and then oven-dried prior to use. A mixture containing 5 mL of $HAuCl_4$ (0.2%, w/w) and 90 mL of water was mixed and refluxed under stirring. 5 mL of sodium citrate trihydrate solution (1%, w/w) was then added quickly. The solution color changed from pale yellow to deep red. After the color changed, the solution was refluxed for an additional 15 min, allowed to cool to room temperature.

2.3. Preparing Teeth and Commercial Tea

According the previous method [32], fifteen fully developed adult molars with roots and crowns were perpendicularly cut from the occlusal surface into three pieces using a low-speed diamond saw. The natural form of the enamel on the upper portion of the teeth was maintained, but the bottom of the incision and the root dentin portions were polished. The polished teeth were cleaned in an aqueous solution of 35% phosphoric acid (H_3PO_4) to remove the smear layer, and then rinsed with distilled water for 30 seconds. The tea solution was produced by soaking 8.0 g of black tea in 50 ml of hot water for 24 hours.

2.4. Bleaching Tooth Stains

Tooth specimens were placed in 0.15 mM tea and stained for three days. After the teeth had been stained and subsequently bleached, they were divided into two groups: (1) 1.5 ml of 17% H_2O_2; (2) 1.5 ml of 17% H_2O_2 plus 1.5 ml of 3.0 3 1023 M GNPs. Then each group was again divided into three reaction condition groups: (1) in a dark room; (2) in open-system photoirradiation (wavelength: 254 nm); (3) in closed-system photoirradiation (wavelength: 310 nm).

3. Results and Discussion

3.1. Characterization of Gold nanoparticles (GNPs)

GNPs solutions were synthesized by the protocols described before. UV-visible spectroscopy and TEM images are presented in Fig. 1 and 2. UV-visible spectrometry was utilized to characterize the synthesized GNPs. It is clear that the GNPs displays a typical surface plasmon broadened band at 520 nm, indicative of the formation of GNPs (the unmodified gold nanoparticles display a characteristic surface plasmon absorption at 520 nm).

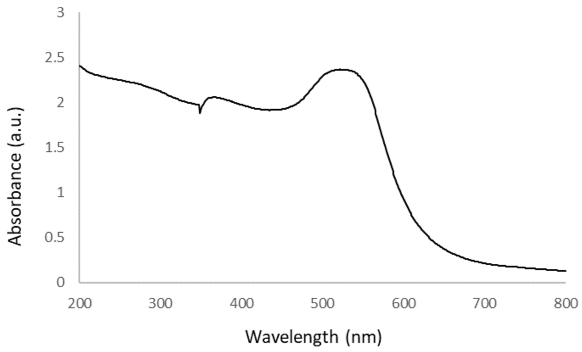

Fig. 1. UV-Vis absorption spectra of the aqueous solutions of gold nanoparticles.

The size and morphology of the formed GNPs were characterized using TEM (Figure 2). It can be noted that the formed GNPs have a spherical shape with mean diameter of ~13 nm. The hydrodynamic sizes of the GNPs were measured to be 22.4 nm via DLS (Figure 3). Zeta potential measurements were also employed to confirm the good reaction. We showed the surface potential of the GNPs (-28.5 mV) (Figure 3).

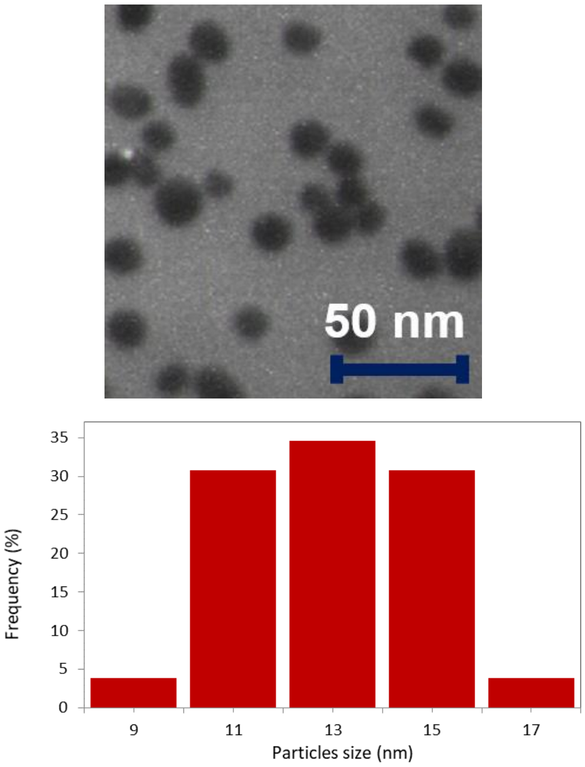

Fig. 2. TEM images of gold nanoparticles and size distributions of GNPs.

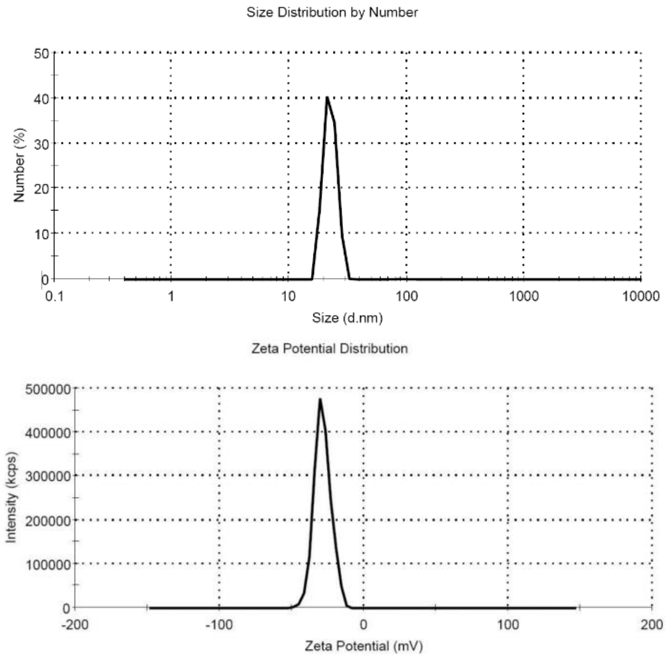

Fig. 3. Dynamic light scattering measurements (DLS) of GNPs (peak: 22.4 nm) and Zeta potential measurements of GNPs (peak: -28.5 mV).

3.2. Dying and Whitening the Tooth Specimens

On the basis of the previous work [32], the removal of black tea, which were bleached without and with photoirradiated H_2O_2 for 1–24 hours. Tooth specimens discolored by black tea in the closed system were significantly whiter after 0.5–2 hours of being bleached with photoirradiated H_2O_2 in the closed system.

Tooth specimens that had been soaked in black tea for 0.5–50 hours and then bleached using H_2O_2 alone for 1–2 hours in a dark room showed significantly more discoloration than did specimens bleached using H_2O_2/GNPs. All three groups bleached and photoirradiated in the closed system showed greater color changes than did the three groups without photoirradiation. We found that GNPs increased the efficacy of whitening teeth stained with black tea. All color images of tooth specimen stain development and bleaching were taken after the specimens had been soaking for 3 days.

The in situ formation of surface-bound gold ions is thermodynamically favored by electron transfer from the gold nanoparticle core to the surface of the oxide layer, which reacts with H_2O_2 to produce highly reactive hydroxyl radicals (•OH) with strong oxidative ability. In contrast, the alternative electron transfers from the gold nanoparticles via the conduction band of the gold nano shell to reduce H_2O_2 was spontaneously driven by the contact with gold nanoparticles.

The bleaching ability of H_2O_2 is based on the generation nascent state oxygen such as excited singlet states ($^1O_2$), and free radicals. Singlet oxygen is a better oxidant than ground state oxygen.These nascent state oxygen [O] and free radicals attack the C=O, C=C, and C=C=C unsaturated double bonds of long molecules in the colored molecules or reduce the colored metallic complexes like gold nanoparticles, which causes the chromogens to decompose. Active radicals produced by H_2O_2 degradation are believed to be extremely short-lived. Consequently, we believe that the bleaching agent H_2O_2 must first penetrate the tooth, and then the radicals must be generated in the dentin by an appropriate mechanism. We used photoirradiation as an ideal vehicle for the generation of radicals: because light can penetrate deep into the tooth, photoactivation of H_2O_2 plus GNPs might increase the reaction rates between H_2O_2 and stain molecules [32].

4. Conclusion

We selected black tea, 2 types of whitening (H_2O_2 alone, H_2O_2/GNPs), and 2 different light sources (not photoirradiated, photoirradiated with light cure), based on the results of this study, the gold nanoparticles were used as a catalyst for tooth bleaching, and it significantly increased the bleaching efficiency of H_2O_2.

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