

Development of a self-sterilizing lancet coated with a titanium dioxide photocatalytic nano-layer for self-monitoring of blood glucose

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Abstract

A photocatalyst was applied to a lancet for pricking the finger to obtain an antibacterial property. A photocatalytic and uniform nano-layer of titanium dioxide (TiO₂) on the surface of the lancet (0.36 mm × 24.5 mm) was formed by sputtering and annealed for crystallization of the TiO₂ layer. By elementary analysis of the TiO₂ layer, titanium and oxygen were detected. Next, for the estimation of the antibacterial properties resulting from the photocatalytic effect, the lancet was packed into a capillary tube filled with a suspension of *Escherichia coli* K-12 (non-spore-forming bacterium), and was continuously rolled in a continuous UV-irradiation system under black-light irradiation. Distinct antibacterial effects after irradiation at 0.5 mW cm⁻² for 45 min were observed in the crystallized TiO₂ layer on the lancet. Finally, lancing resistances obtained by pricking an artificial skin sheet were examined using control lancets, and lancets with an unannealed TiO₂ layer or an annealed TiO₂ layer. The results showed almost the same lancing resistances for the control (0.53 ± 0 N, *n* = 3) and the lancet with an annealed TiO₂ layer (0.51 ± 0.018 N), while the lancet with an unannealed TiO₂ layer showed a high lancing resistance compared with the other lancets (0.62 ± 0.05 N). In conclusion, the lancet coated with a crystallized, velvety nano-layer of TiO₂ obtained by annealing had antibacterial properties and a similar lancing resistance compared with the bare lancet, and showed potential for application in monitoring blood glucose in diabetes.

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1. Introduction

Sterile needles, including lancets, are required in clinical and medical settings, and their use in self-monitoring of blood glucose (SMBG) (Nakamura and Karube, 2003; Newman and Turner, 2005; Kaimori et al., 2006) is increasing due to the rise in diabetes (WHO, 2004).

In the development of needles, both low piercing resistance and low-cost, safe sterilizing methods are required. In a conventional method to reduce piercing resistance, a silicone compound was applied to a metal surface in the form of an adhesive coating

material comprising a siloxane unit with an amino group and an organosiloxane unit (Schwieger, 1971). However, there is a possibility that, when the needle is used, the coating may be peeled off due to insufficient curing of the adhesive material. There is also the problem that gamma-ray irradiation cannot be used for sterilization of needles with this adhesive coating because the piercing resistance of the needle is increased by gamma-rays.

A coating method using silane with an epoxy group on the needle surface was then developed and has excellent curing characteristics (Kurita, 1986). However, the piercing resistance of these needles cannot be reduced sufficiently to reduce patient pain because the coating is too hard. A coating method involving a specific polyorganosiloxane with an amino group and a specific polyorganosiloxane, and a curing method including gamma-ray irradiation, has been applied to the surface of needles, and a low piercing resistance is obtained (Horie et al., 1995). However, the sterilization method is limited to gamma-ray irradiation.

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A mixture of a polyorganosiloxane with amino groups or an alkoxysilane with epoxy groups, and the reaction product of a polyorganosiloxane with an alkoxysilane with epoxy groups has been applied to the surface of a needle (Arimatsu and Nizuka, 2000). Although the sterilization method for this needle is not limited to gamma-ray irradiation, an organic thin film is required to lighten patient's load.

Several sterilizing techniques for needles used in the clinical and medical fields have been investigated and put to practical use, and the most widely used method is gamma-ray irradiation (Reid, 1995; Brinston and Wilson, 1993). However, gamma-ray irradiation requires special equipment and protection to prevent gamma-ray leakage. Thus, the antibacterial properties resulting from the photocatalytic effects of titanium dioxide (TiO_2) have been studied (Fujishima and Honda, 1972). In general, the UV spectra for observing antibacterial properties are from 190 to 300 nm, and the maximum value is approximately 260 nm. However, titanium dioxide photocatalysis can be observed at around 360 nm, a wavelength that is harmless to human eyes. Therefore, the equipment required for sterilization by photocatalysis can be simplified by using an inexpensive black light (UV light). The antibacterial properties achieved by photocatalytic effects were studied using *Escherichia coli*, a TiO_2 -coated glass plate, and a black light (Sunada et al., 2003). In this study, it was suggested that the photo-killing reaction that demonstrated the antibacterial properties was initiated by partial decomposition of the outer membrane, followed by disordering of the cytoplasmic membrane, resulting in cell death. Such antibacterial properties are currently widely used in, for example, toilets, operating rooms, medicine (Fujishima, 2003), and catheters (Ohko et al., 2001). Recently, a micro-syringe was made from titanium and the surface was oxidized by oxygen gas (Tsuchiya et al., 2005). In this study, the biocompatibility of TiO_2 could be utilized; however, the photocatalytic property could not be utilized because UV light could not reach the inside of the syringe for photocatalytic reaction.

The antibacterial properties resulting from the photocatalytic effect on lancets can be achieved at a low cost with a simplified system of sterilization that is extremely safe. In addition, a velvety nano-layer can be achieved with low lancing resistances. In this study, a photocatalytic and uniform nano-layer of TiO_2 was coated onto the surface of lancets by sputtering, and was investigated by electron microscopic observation, elementary analysis, and characterization of antibacterial properties and lancing resistances. In addition, a system for conducting antibacterial tests was developed in this study (Shinohara et al., 2005).

2. Experimental

2.1. Materials

The lancets (CL-CRV 28G; SUS304 medical grade; 0.36 mm \times 24.5 mm) were kindly donated by Facet Technologies Co. (USA). To form a TiO_2 layer on the surface of the lancet, the plastic holder was carefully removed to avoid any damage to the tip (Fig. 1). The chemicals used in this study were of reagent grade.

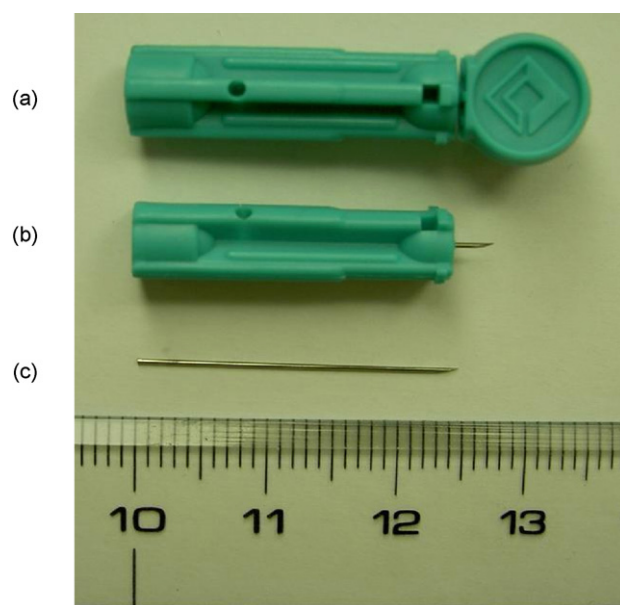


Fig. 1. Photocatalytic lancet. (a) Lancet packaged with a plastic holder and cap. (b) Lancet fixed with a plastic holder. (c) Bare lancet.

2.2. Preparation of photocatalytic nano-layer

The photocatalytic nano-layer of TiO_2 on the lancet was formed by Be-Sputter Co. Ltd. (Japan) and a direct-current (DC) magnetron sputtering system (company's own model). The sputtering procedure was as follows. To achieve a needle-like shape, a nano-layer of TiO_2 was applied twice, on the front and the back of the lancet surface. The TiO_2 nano-layer was applied to a thickness of about 300 nm under the following sputtering conditions: power, 1.0 kW; Ar gas flow, 65–68 ccm; O_2 gas flow, 32–35 ccm; distance between lancet and titanium target, 100 mm; sputtering time, 80 min. The lancet was then annealed at 300 °C for 15 min to crystallize the TiO_2 layer.

2.3. Observation and elementary analysis

The tip of the lancet coated with TiO_2 nano-film was mounted onto a specimen stage and its surface was observed with a field emission scanning electron microscope (FE-SEM; JSM-7700F, JEOL Co., Japan) to obtain secondary electron images (SEI). Elementary analysis of the TiO_2 layer on the surface of the lancet was carried out by energy-dispersive X-ray spectroscopy (EDS) provided in the FE-SEM.

2.4. Antibacterial tests

The antibacterial properties of the photocatalytic lancet were examined using the non-spore-forming bacterium *E. coli* K-12 (MG1665). *E. coli* K-12 was incubated in two steps of pre-culture and present-culture under aerobic conditions, using a Luria–Bertani (LB) medium comprising (amounts in g l^{-1}) NaCl, 5.0, Bacto TriptoneTM, 10, and yeast extract, 5.0, adjusted to pH 7.2, and autoclaved (121 °C, 2 atm, 20 min). In the pre-culture, *E. coli* K-12 was incubated in 2 ml LB medium in a test

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