

論文題目 **Development of Nanostructured Titanium Oxide Film by Wet Corrosion Process and Application for Biodevices**
(湿式溶解プロセスによる酸化チタンナノ構造体薄膜の作製とバイオデバイスへの応用)

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Abstract

The primary purpose of this dissertation is to establish new type of process for synthesizing nanostructures incorporating titanium, and to explore application for nanostructured titanium oxide film as biodevices. It will be relevant applications made the use of nanostructures and property of biodevices discussed in this thesis; that is to say, the geometry characteristic, electrical transport ability, and biocompatibility obtained from synthesized titanium oxide film were used for investigating one-spot electrode for the third generation biosensor and the geometry characteristic, FET-like operation were used for new type ion sensor.

This dissertation is described of the overall consideration composed of the following parts:

- A. Suggest the method for synthesizing nanostructured titanium oxide films with wet corrosion process (WCP) in alkali solution
- B. Characterized the synthesized nanostructured titanium oxide films including geometry, morphology, structural properties, electrical transport ability and so on
- C. Designed the pseudo FET ion sensor using hydrogen ion, and demonstrated analysis method of the signal
- D. Designed the one-spot bioelectrode induced direct electron transfer (DET) from between enzyme and electrode for biosensor and systemically investigated terms for DET in this system with condition of electrode morphology and absorbed enzyme

In chapter 2, the nanostructured titanium oxide film by wet corrosion process (WCP) using various concentration of KOH aqueous solution was explored, and diverse properties of obtained nanostructured titanium oxide film were characterized.

Nanostructured K-doped TiO₂ were fabricated by using a wet corrosion process (WCP) with various concentrations of the KOH solutions. The structure and the morphology of the product were controlled by the volume of the doped potassium content at various KOH concentrations. The nanotubular was synthesized at a dopant concentration of <0.27%; however, at heavy doping, the nanotubular structure disappeared. The K-doped TiO₂ exhibited a distinct electrical behavior with a high conductivity and p-type characteristics. This electrical behavior was governed by the volume of the dopant when the dopant concentration was <0.10% and by the volume of the TiO₂ phase when the dopant concentration was >0.18%. The nanostructured K-doped TiO₂ is expected to have potential applications for nanostructured electronic devices. On the basis of understanding about basic preparation and characterization of obtained products is cornerstone of the whole research, application for biodevices were achieved through diverse properties union and demonstrate in Chapter 3 and Chapter 4.

In Chapter 3, on the possibility of realization of field-effect-transistor (FET) based biosensor with obtained nanostructured titanium oxide film was examined. Utilized properties were 3-D geometry, electrical transport ability, especially p-type characteristic. Fabricated nanostructured titanium oxide films FET was investigated and the pH sensitivity is discussed based FET effect consideration. Fabricated nanostructured titanium oxide films FET revealed turn on/ off behavior, which indicating transistor characteristic. Since the gate region has a nanotubular structure and a 3-D porous structure increases the effective adsorption surface on the channel area, which, in turn, transmits the ion effect to channel. The response of these structures to pH reveals the change in turn off voltage, which recorded 100mV per pH. Thus, the turn off voltage is one of the parameter as detecting signal in this system. In addition, the fabricated nanostructured titanium oxide films FET is not limited to turn off voltage, but the slope of the current–voltage characteristics dramatically increases, leading to high values of sensitivity. This fabricated nanostructured titanium oxide films FET has a potential for ion sensing device.

In Chapter 4, we designed direct electron transfer (DET) based biosensor using obtained nanostructured titanium oxide film as for one-spot electrode. The structure of DET based biosensor is complicated due to separated electron transfer part, matrix part, and DET reaction part. To overcome these problems, we will design no barrier DET based biosensor, namely one-spot electrode for biosensor, using bulk nanostructured titanium oxide film, and systematically investigate the terms for DET induced by

between electrode and immobilized enzyme on the surface. In here, utilized properties were geometry, conductivity, and biocompatibility. Bulk nanostructured titanium oxide films FET with good conductivity were synthesized by WCP in terms of various KOH concentrations and systematically investigated as terms for DET using various nanostructured titanium oxide films FET morphology and concentrations of enzyme solution. It is pointed out that reaction-site size and adsorbed enzyme condition are contributed to the DET, which are defined as follows. The reaction-site is below 100 nm which observing csupidal of network texture, and the surface coverage ratio of HRP is below 1.0. The surface coverage is meaning of surface which was covered with HRP, it calculated ratio amounts of adsorbed HRP / surface area. Thus, the ratio 1.0 indicates the surface is full covered by HRP. We also evaluated the redox catalysis ability of HRP/NPT/Ti-electrode as a biosensor. The catalytic current value was linear according to increase in the concentration of H₂O₂ up to 100 mM and the least concentration of H₂O₂ is 10 μ M at HRP/70 nm-NPT/Ti-electrode in this system. From these results, fabricated bulk NPT/Ti-electrodes have potential to apply for nanobio electronic devices based on DET between enzyme and electrode.

From these demonstration, it is a expected that nanostructured titanium oxide films which were synthesise by wet corrosion process (WCP) can be used in divers bioeletronic devices, such as nanotubular-base single cell endoscopy, nanotubular transistor for the detection, stimulation, and inhibition of neuronal signal propagation, due to unique geometry, biocompatibility, electrical transport ability. Furthermore, not limited biofields, NPT will expecting for direct solar to fuel conversion system, integrated nanophotonics etc.