Functional design of electrolytic biosensor

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Abstract. A novel amperometric biosensor based on conjugated polypyrrole (PPy) deposited on a Pt modified ITO (indium tin oxide) conductive glass substrate and their performances are described. We have presented a method of developing a highly sensitive and low-cost nano-biosensor for blood glucose measurements. The fabrication method proposed decreases the cost of production significantly as the amount of noble metals used is minimized. A nano-corrugated PPy substrate was developed through pulsed electrochemical deposition. The sensitivity achieved was 325 mA/(Mcm²) and the linear range of the developed sensor was 50-60 mmol/l. Then the application of the electrophoresis helps the glucose oxidase (GOx) on the PPy substrate. The main reason behind this high enzyme loading is the high electric field applied across the sensor surface (working electrode) and the counter electrode where that pushes the nano-scale enzyme particles floating in the phosphate buffer solution towards the substrate. The novel technique used has provided an extremely high sensitivities and very high linear ranges for enzyme (GOx) and therefore can be concluded that this is a very good technique to load enzyme onto the conducting polymer substrates.

1. Introduction

The commencement of 21st century have witnessed the advanced visual communication-oriented society which is beginning more and more complicated leading to the emergence so-called "ubiquitous" network. In order to meet the demands of such a visual communication-oriented society, it is essential to realize the functional diversity and the functional integration based on the hyperfine processing. However, the techno-scientific innovation in the semiconductor electronics seems to exhibit a downfall owing to the reachable physical limits.

Organic materials are quite fascinating due to availability of many metastable states which is considered to play a pivotal role for the next-generation electronics [1]. Especially, "Organic Iontronics" which are dealing with the devices and applications using functions of ions etc. attracts much attention [2-5]. The word "Iontronics" is still unfamiliar words and has been created by the fusion of two words "Ion" and "Electronics". Though it may be said that "Iontronics" is discipline which deals with phenomena and device applications based on the behavior of the electron and ion, most people may feel like there is something a little off with in comparison with words infiltrating to the general public widely so-called "Electronics". However, the most familiar area biotechnology which is based on amicable use of unique biological functions towards betterment of human life and
environmental conservation cannot deny the important role played by the ions. “Iontronics” does not mean to simply mimic electronics but it is intended to create possible novel and precise features assisted by ions. In order to realize the molecular devices such as functional element of the next-generation electronics, a thought of "Iontronics" will be indispensable.

In this article, the problems that should solve about the characteristic improvement such as the performance of the biosensor which is one of the bio-related "Iontronics" devices are introduced concretely. The biosensor is the measurement device which can be used or imitated by the high material recognition function to have of the bio-material. Since Clark and Lyons proposed the glucose biosensor for the first time in 1962, by putting together with various signal conversion devices and bio-elements, those have been developed and their applications are expected in many fields such as medical care, food, and environment [6].

Figure 1 shows a basic principle of biosensor and their application fields [7]. The biosensor is constructed in the substrate recognition region called the receptor which recognizes a substrate (measurement target material), and in the signal conversion region called the transducer which causes a chemical change at that place and finally converts the change into an electrical signal. In the substrate recognition department, an enzyme, an antibody, a cell, a microbe and so on are used and in the signal conversion region, an electrode detecting substances change, thermistor detecting a heat change, piezoelectric elements detecting a weight change, and so on are employed. The current biosensor is mainly constructed with the enzyme electrode consisting of an enzyme as the substrate recognition region, and the electrode as the signal conversion region.

The most popular enzyme used in the glucose biosensors is glucose oxidase (GOx). Various immobilization approaches such as physical adsorption, covalent binding, crosslinking, and coentrapment, have been developed for immobilizing GOx [8, 9]. As one of effective methods of fabricating glucose biosensors, a combined procedure of physical adsorption and coentrapment of GOx in conductive polymer matrix may become important [10, 11]. To date, various conductive polymers have been synthesized, but polypyrrole (PPy), the molecular structure of which is shown in Figure 2, is the most stable after doping and is one of the
most attractive materials, together with polyaniline (PAn), because of their good environmental stability and relatively high electrical conductivity. Then to develop nerve stimulation electrodes of the inside of the body burial type, PPy of biocompatibility was mainly evaluated in the present study [12].

The sensitivity of a biosensor mainly depends on the amount of enzyme loaded onto the reacting surface and the ability to collect the electrons released by the reaction. Therefore increased effective reacting area is one way to obtain a higher sensitivity. Different research groups apply many different techniques to increase the effective surface area including the use of materials like carbon nanotubes and processes like nano-templating [10, 13-17]. In this research, we have investigated developing a nano-scale corrugated conductive polymer surface suitable for immobilizing glucose oxidase (GOx) for the development of a low-cost nano-biosensor. That is, traditional biosensors use noble metals for their electrodes and they contribute to a significant portion of their production cost. In contrast, conductive polymer based biosensors use minimum amount of noble metals and therefore reduce the cost of production. Further, the fabrication process involved with this novel nano-biosensor is comparatively simple and low-cost.

2. Electrolytic polymerization technique

2.1. Electrolytic polymerization condition

In general, electrolytic polymerization technique is performed with the electrolytic polymerization equipment shown in Figure 3 [18]. If the aromatization compound monomer that is to be polymerized is dissolved into a solvent containing a suitable supporting electrolyte and an appropriate voltage is applied to the electrode pair immersed into this solution, the monomer is oxidized or reduced on the surface of the anode or cathode, respectively, and it polymerizes in the form of a powder or a film and

![Figure 2. Molecular structures of polypyrrole (PPy), polyaniline (PAn), and poly(3, 4-ethylenedioxy thiophene) (PEDOT).](image)

![Figure 3. Electrolytic polymerization cell.](image)
at times may show arborization. The case in which the monomer is oxidized and polymerized on the anode surface is called electrolytic oxidation polymerization.

On the other hand, the case in which the monomer is reduced and polymerized on the surface of the cathode is called electrolytic reduction polymerization. The reference electrode might be immersed in the solution if necessary.

In order to obtain good-quality conductive polymer films by the electrolytic polymerization technique, it is necessary to examine the above predominant factors in detail and to understand the optimal electrolytic polymerization condition. Moreover, the conductive polymer can be reversibly electrochemically doped and dedoped and the dopant concentration can be widely adjusted by limiting the voltage, enabling a film that has an arbitrary electric conductivity to be obtained.

2.2. Reaction mechanism of electrolytic polymerization

The reaction mechanism of conductive polymer synthesis by electrolytic polymerization has not been clarified, because various factors, such as the composition of the electrolyte and the electrolytic conditions, interact very complexly with the electrode reaction. Therefore, the polymerization reaction condition differs for each individual conductive polymer and the optimum conditions are determined through trial and error. However, qualitatively, the following reaction mechanisms are accepted in general. The dynamic movement of the molecule, which takes part in the monomer, and the electrolytic ion formed by the transfer of an electron from the solvent are caused at the interface between the electrode and the electrolyte in the electrolytic polymerization technique. Usually, 2–2.5 electrons are consumed in the electrolytic polymerization reaction, and ultimately, two pieces of electrons are used in the polymerization reaction and the remainder is used in the doping. As a result, the same number of protons (H⁺) that as the number of electrons used for the polymerization will accumulate in the polymerization liquid. Therefore, the polymerization reaction occurs as a result of the removal of the electron from the monomer, and the coupling reaction, in which the generated radical cation (positive ion) is assumed to be the activated species, and the deprotonation reaction are thought to progress repeatedly. The polymerization reaction is thought to be either reaction 1, the parent electron substitution coupling reaction, or reaction 2, the radical coupling reaction (Here, M indicates a monomer), as shown in Figure 4.

\[
\begin{align*}
\text{Reaction 1: electrophilic substitution coupling reaction} & \\
\text{Reaction 2: radical coupling reaction} & \\
\end{align*}
\]

Figure 4. Reaction mechanism of electrolytic polymerization.

However, the structure analysis of the obtained polymer is difficult because the polymer is nonfusible and nonsoluble. Then, the polymerization reaction mechanism itself becomes very complex and a united opinion is not obtained, because electrolytic polymerization is a reaction of a nonuniform system that progresses in a limited area in the neighborhood of the electrode, and it is necessary to
consider the supply of monomers and electrolytic ions to that area. That is, the electrolytic polymerization is thought to begin with the oxidizing or reducing reaction of the monomer in the electrolyte upon applying potential, followed by the generation of the radical cation or the radical anion in the aromatic compounds, and subsequently, the polymerization progresses repeated the coupling and the deprotonation reaction. It is necessary to prevent the solvent from undergoing the electrolytic reaction with the monomer at a low voltage, because the effects of the various factors such as the type of solvent, the supporting electrolyte, the polymerization voltage, and the polymerization temperature, on the electrolytic polymerization reaction are not yet fully clarified [18].

3. Experimental
Polymerizing a nano-corrugated conductive polymer substrate matching the size of the enzyme size is not an easy task. In the past we have done this through nano-templating using a nano-porous almina template [10, 11]. However the cost involved with the nano-porous alumina template was a significant factor in the cost of production. Therefore in this research we try to develop a different technology to produce a matching conductive polymer surface for the enzyme immobilizing.

3.1. Materials
Glucose oxidase (E.C.1.1.3.4., 210 U/mg, GOx) from Aspergillus niger, pyrrole monomer, ascorbic acid, uric acid and sodium tetrafluorophosphate were purchased from Wako, Japan. α-D glucose was obtained from Sigma-Aldrich. Pyrrole monomer was distilled and sodium tetrafluorophosphate solution was freshly prepared before use. A 0.05 mol/l phosphate buffer solution (PBS) was freshly prepared using Potassium Dihydrogenphosphate (KH2PO4) and Disodium Hydrogenphosphate (Na2HPO4) at pH 6.5. The pH values of the buffer solutions were adjusted using a Cyberscan pH 100/RS232 portable meter. A stock solution of 1 mol/l glucose was prepared and allowed anomers to reach equilibrium at aqueous state, before use. That is, glucose solutions made from α-D glucose (prepared with a 0.05 mol/l PBS, pH 6.5) were left at rest for more than 12 hours before use and allowed to stabilize. All the chemicals were of analytical grade and throughout the experiment temperature was maintained at 25 ± 2 ºC unless otherwise stated.

ITO (indium tin oxide) electrodes were Pt coated by plasma sputtering using JEOL quick coater (JFC 1,500). Hokuto Denko automatically polarization system (HSV-100) was used to perform all electrochemical studies including polymerization and current response measurement.

3.2. Electrolytic polymerization equipment

In general, as shown in Figure 3, a conventional three-electrode cell consisting of a working electrode (Pt coated ITO), a platinum counter electrode and an Ag/AgCl reference electrode was used for
electrochemical synthesis and measurements. A magnetic stirrer was used during the amperometric measurements to ensure convective transport. However, the schematic experimental setup we used for our work is shown in Figure 5. The ITO electrode was modified by plasma vacuum deposition of a 50 nm thick Pt layer on the ITO surface using JEOL quick auto coater. This electrode was placed on one contact terminal of the test ring and a Pt electrode was placed on the other contact terminal where the Pt/ITO surface facing the Pt counter electrode. The gap between them was 1 mm and the common area between them was 1 cm². The test rig was positioned to have the electrodes horizontal and an adequate amount of 0.05 mol/l aqueous pyrrole solution doped with 0.1 mol/l NaPF₆ was carefully injected in between the electrodes.

3.3. Fabrication of nano-porous PPy electrodes

Normally, the sensitivity of a biosensor is determined by several factors such as the match between surface porosity and physical dimensions of target enzyme, effective surface area and charge collecting backbone material on the substrate etc. Therefore, it is very important to increase the effective surface area in order to obtain a good sensor. However, the surface roughness of the artificial porosity should match with the physical dimensions of the target enzyme [9].

When a PPy layer is electrodeposited on a planar electrode, polymerizing starts with random nuclei and keeps on growing out of them as long as the polymerization current continues. However, if the polymerization current is interrupted instantaneously, there is no guarantee that it will restart from the same point it stopped before and most probably start from a different physical location on the working electrode where there is a low contact resistance. We have effectively utilized this random polymerization phenomenon to develop an artificially porous conductive polymer surface for our working electrode to entrap GOx. The physical dimensions of the porosity can be controlled with the applied voltage, mark to space ratio and pulse width of the polymerization potential. In this research we have tested various pulse timing and mark to space ratios and optimized our sensor performance with 1 s pulse time with 1 : 1 mark to space ratio of 1 V polymerization potential. Figure 6 shows a schematic experimental diagram of the pulsed electrolytic polymerization system.

3.4. Surface morphology of PPy electrodes

Different pulse timing and pulse voltages were used to optimize the above parameters. Figure 7 (a) and (b) shows the nano-scale morphological changes of the PPy substrates electrodeposited under pulsed voltage and constant voltage respectively from these trials. Pulse voltage was 1 V and deposition was carried out for 1 s and turned off for the next 1 s period. This was repeated for 175 cycles. Pt modified ITO substrates were used for all experiments. As shown in Figure 7 (a) and (b), clearly, these surface morphologies of these PPy electrodes are greatly different each other. That is, the surface area of pulsed deposition PPy electrode (a) is larger than that of constant voltage deposited PPy electrode (b).
3.5. Immobilization of GOx on the PPy electrode

Once NaPF₆ doped pyrrole monomer is in place, twenty times for 1 V pulses of 1 s were applied to the terminals where the Pt/ITO acted as the working electrode and Pt as the counter electrode. After polymerization the remaining pyrrole monomer was removed and both the electrodes were carefully washed in pH6.5 phosphate buffer solutions (PBS). Then the electrode assembly was positioned again to have the electrodes horizontally and a little less than adequate amount of GOx solution was injected in between the electrodes.

An aliquot of glutaraldehyde was injected to the same chamber to mix with GOx and a potential of 1 V was applied to the system for 20 minutes constantly. This provides the environment for the GOx to get deposited under a high electric field of 1 kV/m. This electrophoretic deposition helps the GOx deposition on the PPy electrode. After the period the sensor and the counter electrode were carefully washed in pH6.5 PBS.

3.6. Biosensor fabrication

The sensor assembly was mounted vertically for the testing where the open end of the electrodes facing downwards. The sensor electrode and counter electrode were gently submersed in a slowly steady stirring PBS of pH6.5 up until the 1 cm × 1 cm area was covered by PBS. Fabricated sensor (as working electrode) and the Pt counter electrodes were connected to Hokuto Denko HSV-100...
electrochemical system in a three-electrode configuration where the reference electrode was Ag/AgCl. The GO\textsubscript{x}/pulse-deposited PPy/Pt electrode was maintained at room temperature at a polarization potential of 0.38 V vs. Ag/AgCl in the air-saturated sensing solution to yield a stable background current. The system was applied a potential of 0.38 V and was allowed to settle down before the testing started. When the system reached stability a known concentration of glucose was added to the system and allowed to settle. This was repeated for different glucose concentration and all data were logged.

In the next round of testing the system was applied a potential of 0.38 V and was allowed to settle down before the testing started. When the system reached stability, 5 mmol/l of glucose was added to the solution at constant time intervals and the response of the system was continuously recorded through the HSV-1000.

The sensitivity of a sensing electrode is represented by the following.

\[
sensitivity = \frac{\text{(current response)}}{\text{(concentration of glucose)}} \times \text{(initial area of the electrode)}
\]

All sensors, PBS, and GO\textsubscript{x} were kept at 4 \(^\circ\)C while in storage and were allowed to settle to the room temperature before the experiments.

3.7. Equipment which assists experiments

Electrochemical impedance spectroscopy (EIS) measurements system (Hokuto Denko Co., Ltd., HZ-5000) was used to confirm the surface modification under GO\textsubscript{x} deposition stage. That is, two EIS measurements, one before and one after GO\textsubscript{x} deposition were carried out to make sure an effective GO\textsubscript{x} deposition was taken place.

If the enzyme was loaded on to the PPy electrode, it should modify the nanoscale morphology of the sensor substrate and hence the contact impedance. The changes in real and imaginary impedance show that the enzyme deposition has taken place successfully. However, EIS measurement in our experiment is only adhesion confirmation of GO\textsubscript{x} onto the PPy electrode. The details of the EIS measurement based on the electrophoretic electrodeposition of GO\textsubscript{x} will be reported somewhere in the near future.

4. Results and discussion
This research revealed that the developed novel glucose sensor has comparatively a high sensitivity and a very good linear range for this class of a nano-biosensor. This was achieved through the novel technique of electrophoretic enzyme deposition. Same sensor substrate deposited with glucose oxidase, again with the same electrophoretic enzyme deposition resulted in a very high sensitivity and a good linear range [19]. Both these tests revealed that the new enzyme deposition technique has a very high impact on the amount of enzyme loaded on to a unit linear surface area.

4.1. Optimization of biosensor’s sensitivity

Our purpose of using nano porous corrugated PPy as our base electrode was, to retain its original nano porosity until PPy polymerization stage allowing PPy film to be porous artificially. Therefore controlling the thickness of PPy by optimizing the polymerization time (i.e., application time of the pulsed voltage, in other word, the charge passed during polymerization) was crucial in this regard. With GOx enzyme having particles around 10-100 nm in size, adequate size of pores for GOx entrapment was also regarded to be in a closer range.

Figure 10 shows good performances around a polymerizing time (application time of the pulsed voltage t (s)) of around 1 s giving evidence for ideal pore size to serve our purpose. The SEM image in Figure 7 provides clear clues of this PPy growth. The efficiency of immobilization of the enzyme is influenced by this changed effective surface area available due to nano-pores.

4.2. Time response of the sensor for different glucose concentrations

The sensor was tested for its time response under known different glucose concentration. It displayed very similar and consistent time responses for different glucose concentrations. Results revealed that the sensor reaches its stable state in about 40 s, which is little longer than normal value for a glucose biosensor but still acceptable for a low-cost biosensor.

The current-time curves shown in Figure 11 shows that the GOx/pulse-deposited PPy/Pt electrodes have a response and high sensitivity to glucose. At a polarization potential of 0.38 V vs. Ag/AgCl and in an air-saturated sensing solution, the anodic current increased dramatically and reached a steady state within 40 s as seen the inset in Figure 11 after the injection of a certain amount of a glucose solution. By applying a polarization potential of 0.38 V, polarization current, that is, background current was reached a steady state. And then, only the response current after the injection of a certain amount of glucose solution was examined.
Figure 11 shows the response of the fabricated sensor against increase of glucose concentration by 5 mmol/l at a step. The system was allowed to settle for extensive amount of time in order to minimize background noise from the PBS, but it can be used with much less stabilization if appropriate noise filtering is employed.

The current response of a PPy film polymerized by constant voltage on a Pt coated ITO electrode is shown in Figure 11 for similar concentration of glucose. It has been revealed that only a little amount of GOx is entrapped in the PPy film deposited by constant voltage on the Pt/ITO electrode without a porous structure compared to the highly porous new pulse-deposited PPy electrode.

The results illustrated in Figure 11 reveal that the sensor has a very high sensitivity and a high linear range. Figure 12 was derived from data in Figure 11 and this shows that the linear range spans from 0 to 60 mmol/l, which is more than double the range of a blood glucose sensor. Further, the sensitivity value calculated from Figure 11 for this sensor is 325 mA/(Mcm²). This is a very high sensitivity enables designing of low sensitivity front-end electronics otherwise needed very high sensitivity measuring circuits [21, 22].

As it can be seen from the Figure 11, the response current tends to be fluctuating at higher glucose concentrations. This can be due to the physical arrangement of the sensor electrodes with only 1 mm apart from each other, not allowing a homogeneous mixture of glucose to enter the nature gap at higher concentrations. At the same time, the reaction product is not removed at a linear rate due to narrow opening and surface tension of the liquid. Both these factors contribute at higher glucose concentrations to produce a fluctuating current. This can be overcome by introducing a planar geometry, but will have to sacrifice the sensitivity at the same time.

Though the sensor responded for even higher glucose concentrations, the sensitivity has a higher standard deviation beyond 55 mmol/l and the linear range ends nearly at 60 mmol/l of glucose concentration. The extraordinary sensitivity and the very high linear range resulted by the pulsed deposition of PPy film suggests that this newly developed biosensor can be applied in industrial applications as well.
5. Conclusion

The commencement of 21st century have witnessed the advanced visual communication-oriented society which is beginning more and more complicated leading to the emergence so-called "ubiquitous" network. In order to meet the demands of such a visual communication-oriented society, it is essential to realize the functional diversity and the functional integration based on the hyperfine processing. However, the techno-scientific innovation in the semiconductor electronics seems to exhibit a downfall owing to the reachable physical limits. Organic materials are quite fascinating due to availability of many metastable states which is considered to play a pivotal role for the next-generation electronics.

In this research, highly sensitive glucose oxidase (GOx) electrodes were fabricated on the basis of nanostructured polypyrrole (PPy) films using pulse electrodeposition. The nanostructures of the PPy films had a nano-porous morphology like voids. GOx was immobilized in nanostructured PPy films coated on a ITO substrate electrode. The GOx/PPy/Pt electrode showed a linear response of glucose concentration in the range of 0 – 50 or 60 mmol/l at a potential of 0.38 V (vs. Ag/AgCl). Its sensitivity was measured to be approximately 325 mA/(Mcm²) at room temperature. In comparison, the sensitivity of the GOx/PPy/Pt electrode based on a flat PPy film was only approximately 0.3 mA/(Mcm²) under the same conditions.

The nano-biosensor fabricated shows excellent performance with the optimized polymerization parameters. These results show a cost effective way of developing a highly effective nanostructured working electrode surface for nano-biosensor application. This method can be used to develop different nano-biosensor working electrodes by fine-tuning the electrochemical deposition parameters such as time interval, mark to space ratio and pulse shape etc., to adjust the PPy cavity sizes to suit the chosen enzyme.

The electrochemical deposition of pyrrole under pulsed electric field has created many different PPy islands on the planar Pt modified ITO surface otherwise it would have been a linear growth from the original nuclei. Eventually this produced an artificially corrugated nanostructure for the newly formed PPy substrate. From these results, it can be concluded that the PPy layer electrodeposited on the planar surface of Pt modified ITO has got suitable size of nano-cavities to hold the GOx enzyme molecules.
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