

Full Length Article

Incorporation of hydrogel as a sensing medium for recycle of sensing material in chemical sensors



Yunjung Hwang^a, Jeong Yong Park^b, Oh Seok Kwon^c, Seokwon Joo^b, Chang-Soo Lee^{c,d,*}, Joonwon Bae^{a,*}

^a Department of Applied Chemistry, Dongduk Women's University, Seoul, 02748, Republic of Korea

^b College of BioNano Technology, Gachon University, Seongnam City, 13120, Republic of Korea

^c Hazards Monitoring Bionano Research Center, Korea Research Institute of Bioscience & Biotechnology (KRIBB), 125 Gwahak-Ro, Yuseong-Gu, Daejeon, 34141, Republic of Korea

^d Nanobiotechnology (Major), University of Science & Technology (UST) 125 Gwahak-Ro, Yuseong-Gu, Daejeon, 34141, Republic of Korea

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ABSTRACT

A hydrogel, produced with agarose extracted from seaweed, was introduced as a reusable medium in ultrasensitive sensors employing conducting polymer nanomaterials and aptamers. A basic dopamine (DA) sensor was constructed by placing a hydrogel, containing a sensing material composed of aptamer-linked carboxylated polypyrrole nanotubes (PPy-COOH NTs), onto a micropatterned gold electrode. The hydrogel provided a benign electrochemical environment, facilitated specific interactions between DA and the PPy-COOH NT sensing material, and simplified the retrieval of PPy-COOH NTs after detection. It was demonstrated that the agarose hydrogel was successfully employed as a sensing medium for detection of DA, providing a benign environment for the electrode type sensor. PPy-COOH NTs were recovered by simply heating the hydrogel in water. The hydrogel also afforded stable signal intensity after repeated use with a limit of detection of 1 nmol and a clear, stable signal up to 100 nmol DA. This work provides relevant information for future research on reusable or recyclable sensors.

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1. Introduction

Interest in hydrogels has become significant in all fields of science and technology. Hydrogels are an attractive class of materials used in biological and biomedical applications including drug delivery, tissue engineering, and artificial skin development [1,2]. Microscale hydrogels have been developed for use in electronic devices [3] and extensive research efforts have been devoted to the development of more elegant hydrogel materials [4].

Hydrogels are three-dimensional insoluble polymer networks with a relatively open matrix structure and consequently high internal volume. They are also relatively hydrophilic and therefore compatible with most biomolecules [5]. As a result of these properties, sensor development [6–9] has become one of the most popular fields within the diverse and competitive application space of hydrogels. For example, conductive hydrogels have been employed effectively for monitoring glucose [10–12]. Hydrogels that respond to changes in the external environment have also been used as

photonic sensors [13,14]. Mechanical sensors using soft and elastic hydrogels have also been reported [15–17].

The need for reusable and recyclable sensors is continuously growing due to environmental concerns and cost-efficiency. Hydrogels are considered one of the most promising candidates for reusable or recyclable sensors as they are naturally abundant, nontoxic, and easy to handle. In particular, agarose hydrogels, produced with seaweed extracts, are appealing because they can be easily reshaped by simply heating and cooling in water [18–22].

In this study, an agarose hydrogel was introduced as a reusable sensing medium for an ultrasensitive aptasensor. The hydrogel accommodated a sensing material, aptamer-linked carboxylated polypyrrole nanotubes (PPy-COOH NTs), for the detection of dopamine (DA). It was expected that the hydrogel would be advantageous for transporting target molecules, suppressing signal noise, and simplifying the retrieval of sensing material. A basic sensor was constructed to demonstrate the role of a hydrogel as a sensing medium by placing a hydrogel containing PPy-COOH NTs linked with aptamers onto a patterned gold electrode. Sensor performance was monitored every minute. This article provides information for future research on hydrogel-based, reusable, and recyclable sensors.

* Corresponding authors.

E-mail addresses: cslee@kribb.re.kr (C.-S. Lee), joonwonbae@gmail.com (J. Bae).

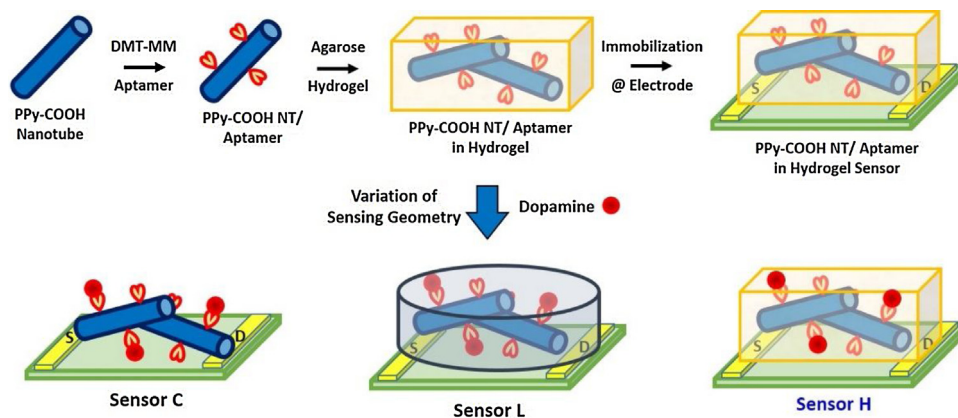


Fig. 1. A schematic shows the production of sensors of varying geometry employing hydrogels containing aptamer-linked PPy-COOH NTs.

2. Experimental

2.1. Materials

Pyrrole (Py), pyrrole-3-carboxylic acid (Py-COOH, 95%), sodium bis(2-ethylhexyl) sulfosuccinate (AOT, 98%), agarose, and hexane (99%) were purchased from Aldrich. A condensing agent, 4-(4,6-dimethoxy-1,3,5-triazin-2-yl)-4-methylmorpholinium chloride (DMT-MM) was also purchased from Aldrich. A 57-mer dopamine-binding DNA aptamer, having the sequence 5' amine GTC TCT GTG TGC GCC AGA GAC ACT GGG GCA GAT ATG GGC CAG CAC AGA ATG AGG CCC 3' was, designed as reported and synthesized by Bioneer (Daejeon, Republic of Korea). Stock aptamer solutions were prepared with DDW and stored at 20 °C before use.

2.2. Fabrication of carboxylated polypyrrole (PPy-COOH) nanotubes (NTs)

A surfactant, AOT (15 mmol), was dissolved in hexane (40 mL) and the solution was stirred for 30 min. An aqueous FeCl₃ solution (5 M, 1 mL) was subsequently introduced into the AOT/hexane solution to generate reverse-cylindrical micelles containing iron cations [23]. Finally, the Py (7.5 mmol) and Py-COOH (0.25 mmol) mixture was added drop-wise into the micelle solution. The oxidative polymerization of Py/Py-COOH monomer proceeded for 3 h at room temperature. The resulting product was thoroughly washed with excess ethanol to remove the surfactant and other residual reagents. The final products were obtained after drying under vacuum at room temperature.

2.3. Fabrication of agarose hydrogel containing carboxylated polypyrrole (PPy-COOH) nanotubes (NTs) linked with aptamer

The obtained PPy-COOH NTs were dispersed in distilled water and the aptamer solution and a condensing agent (DMT-MM) were added drop-wise. The coupling reaction, which served to attach the aptamer to PPy-COOH NTs, proceeded for 1 h.

A 2 wt% solution of agarose in distilled water was prepared by heating the mixture at 100 °C on a hot plate until the agarose was completely dissolved. The solution was cooled to ~50 °C, where the solution remained in a liquid state. The aptamer-linked PPy-COOH NTs were then added to the agarose solution and stirred for 30 min. Then the whole solution was cooled to room temperature. The weight fraction of PPy-COOH NTs in the resulting hybrid was approximately 5%. As the temperature decreased, the viscosity of the mixture increased quickly due to gelation and the material gradually changed to black. The obtained gel was washed with

fresh distilled water at least three times and dried under ambient conditions.

2.4. Construction of electrode type sensor

A gold microelectrode was patterned onto a glass substrate using a lithographic process. A hydrogel containing aptamer-linked PPy-COOH NTs was manually placed on the surface of the gold electrode using tweezers. Variations in measurement strategy are shown in Fig. 1.

2.5. Recovery of sensing material, carboxylated polypyrrole (PPy-COOH) nanotubes (NTs) linked with aptamer

After the completion of electrochemical measurements, the sensing material was retrieved by thermally melting the hydrogel in distilled water to 80 ~ 90 °C. The resulting solution was stirred for 30 min and 100 mL of hot water added. The mixture was centrifuged at 3000 rpm for 15 min, resulting in a black precipitate that was removed and dried [24]. Another hydrogel was prepared as described above, incorporating the recovered aptamer-linked PPy-COOH NTs.

2.6. Instrumentation

Electrical measurements were obtained with a Keithley 2612 B SourceMeter and probe (MS TECH, Model 4000). Changes in electrical current were normalized as $\Delta I/I_0 = (I - I_0)/I_0$, where I_0 and I are the initial and detected current, respectively. A sample chamber (0.25 mL volume) was designed and used (sensor L in Fig. 1) for solution-based measurements. A fluorescence microscope (Olympus, model IX2-RFA), with excitation and emission wavelengths of 532 and 488 nm, respectively, was used to detect aptamers. FT-IR spectra were recorded on a Perkin Elmer Spectrum One spectrometer. SEM micrographs were obtained with a JEOL 6700 scanning electron microscopy at an accelerating voltage of 10 kV.

3. Results and discussion

Conducting polymers boast the combined advantages of polymers and semiconductors. Their large conjugated structures allow intermolecular charge transport. As a result, conducting polymers have been used extensively as sensing materials in chemical sensors. The schematic in Fig. 1 shows the overall sensor architecture used in the current study. Starting from monomer, PPy-COOH NTs with an average diameter of several hundred nanometers, as shown in Fig. 2a, were prepared using a reverse micelle system [23]. Note that the presence of COOH functional groups decreased the

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