

# An Extended Gate Field Effect Transistor (EGFET) pH Microsensor

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**Abstract**—Here, an extended – gate field effect transistor (EGFET) pH microsensor was developed for use in fast and sensitive pH measurement applications. The system consisted of two components; a pH sensitive modified gold electrode and a simple and cheap metal oxide semiconductor field-effect transistor (MOSFET). Polypyrrole, a semiconductor and pH responsive polymer, was formed by electropolymerization of pyrrole monomer at the surface of the gold electrode in galvanostatic mode. Then, measurements were made in PBS at different pH values using the pH sensitive electrode. In this context, the pH sensitivity of polypyrrole with respect to electropolymerization and incubation time were studied. According to the results, the EGFET pH microsensor formed by 4-min pyrrole electropolymerization showed at pH 6-12 the highest pH sensitivity with 67 mV/pH.

**Keywords**—EGFET, pH microsensor, electropolymerization, MOSFET, polypyrrole.

## I. INTRODUCTION

Proper measurement and control of pH is essential for many industrial applications and living organisms due to its relevance to many chemical and biochemical processes. Today, different electrochemical and non-electrochemical methods are used to measure pH values [1]. There are various methods for pH measurement, such as pH paper strips that change color depending on pH, potentiometric methods based on glass electrodes, fluorescence, amperometry and conductimetry. Although glass electrodes are widely used, they have some disadvantages such as difficulty of miniaturization, fragility, frequent recalibration prior to use, pressure and temperature dependence. Solid-state electrodes have been developed to overcome such limitations [2]. Solid state sensors can provide a cheap, robust and miniaturizable alternative for pH measurements, as Ion Sensitive Field-Effect Transistors (ISFETs) based pH probes. These features can be used to make the sensor system with low power consumption, low cost, and ease of operation [3]. Although ISFETs have small size and fast response time, the need for (glass) reference electrode remains a disadvantage for these sensors. Complex gate encapsulation procedures and costly clean room processes required to produce an ISFET increase the cost of manufacture [4]. An extended – gate field effect transistor (EGFET) is a device which can be applied to chemical sensing devices and also can be used as pH sensing in which the chemically sensitive

electrode region, coaxial cable, and buffer are on a single integrated structure [5], [6]. Activity of target analyte results in an additional chemical contribution to the threshold voltage in EGFET. It is because of separating the sensing membrane physically and preserving the gate region of MOSFET (Metal Oxide Semiconductor Field Effect Transistor). Applications of EGFET are especially associated with measurement of pH, ionic species, urea and glucose through the functionalization of the gate surface [7]. Popularity of conducting polymers are increasing in the field of sensors due to their low cost, robustness and versatility [3]. Polypyrrole is a conducting polymer that is especially used in potentiometric sensors due to its biocompatibility, high conductivity, easy polymerization, high pH sensitivity and stability [8], [9]. In this context, Lakard et al. developed a potentiometric miniaturized pH sensor by modifying platinum electrodes with electrosynthesized polypyrrole films. One electrode of the sensor is covered with a polypyrrole film and the other one was made of a silver film as a reference electrode. pH change was almost linear in the range of 3 to 11 [8]. In another work, Naser et al. designed an electrochemical flow cell pH sensor for aqueous solutions based on the working principle of an EGFET. Two different sensitivities were obtained in the pH range of 8.00 to 10.80 on the first day and on the 15th day. It was stated that the sensitivity, which was 52.31 mV/pH on the first day, decreased to 42.65 mV/pH on the fifteenth day [4].

Since polypyrrole contains a significant portion of –NH groups that can be protonated or deprotonated as a function of pH changes [10], an EGFET pH microsensor integrating a MOSFET with a polypyrrole coated electrode was fabricated and reported in this study. The EGFET pH microsensor produced with a new approach was small, simple, cheap, fast and sensitive. In addition, it is expected that the microsensor can be used for local pH measurement in small volume environments, biological samples or corrosion studies.

## II. MATERIAL AND METHODS

### A. Material

Phosphate buffered saline (PBS) (Sigma Aldrich, USA), sodium hydroxide (NaOH) (Sigma-Aldrich, USA), hydro-

gen chloride (HCl) (Sigma-Aldrich, USA), pyrrole (Sigma-Aldrich, USA), lithium perchlorate ( $\text{LiClO}_4$ ) (Sigma-Aldrich, USA), perchloric acid ( $\text{HClO}_4$ ) (Supelco, USA), silver paste (Sigma-Aldrich, USA), n-type MOSFET (Metal Oxide Semiconductor Field Effect Transistor) (IRFZ44N, International Rectifier, USA). Potentiostat (Autolab PGSTAT204) was used in the electropolymerization of pyrrole on the produced sensor. The field effect transistor (FET) analysis unit (B2901A Precision Source/Measure Unit) was used for electrochemical measurements and characterization of the produced sensor.

### B. Microchip production

A glass slide was spin coated with S1818 positive photoresist at 2000 rpm for 30 sec. After prebaking, the photoresist was patterned via contact lithography. The exposed areas of the photoresist was removed using a developer solution (CD-26, Microchem, USA). Ti and Au films were sputtered on the glass slide with patterned photoresist. Lastly, the remaining photoresist was removed using acetone to define the Au patterns of the microchip.

### C. Pyrrole solution preparation and electropolymerization

Pyrrole solution was prepared using 0.2 M pyrrole, 0.2 M lithium perchlorate ( $\text{LiClO}_4$ ) and 0.1 M perchloric acid ( $\text{HClO}_4$ ). Electropolymerization of the pyrrole solution was carried out in galvanostatic mode by applying a current of 0.1  $\mu\text{A}$  to the working electrode (gold electrode) for 4 minutes by potentiostat/galvonastat (PGSTAT204, Metrohm, Switzerland). In addition, 0.1  $\mu\text{A}$  current was applied in galvanostatic mode for different times (1, 3, 4, 5, 10 min) to see the effect of coating time during the electropolymerization process. In Table I, effect of different time intervals to pH sensitivity was given. According to result obtained by this, pH measurements were continued by chip 3. A two-electrode electrochemical cell was used for electropolymerization. The Au electrode and an Ag electrode printed on the chip using Ag paste were used as the working and reference electrodes, respectively. The electrode coated with polypyrrole was incubated for 40 minutes at room temperature. Then, PBS solutions at different pH (5.53 - 7.08 - 7.96 - 8.57 - 9.82 - 12.07) were prepared by adjusting the pH of PBS using NaOH and HCl and used to determine the pH sensitivity of the EGFET pH microsensor.

### D. Use and characterization of EGFET pH microsensor in pH measurement after integration with MOSFET

The measurement capacity and characterization of the EGFET pH microsensors were performed in PBS solutions at different pH using a field effect transistor (FET) analysis unit (B2901A Precision Source/Measure Unit). While the source and drain of the n-type MOSFET were connected to the source and drain inputs of the FET analysis unit, the gate is connected to the EGFET pH microsensor as can be seen in Figure 1. In order to make the measurement more sensitive and stable, pH measurements were carried out with the printed Ag electrode as the reference electrode. Measurements were carried out by immersing the prepared pH microsensor into solutions at

different pH values (5.53 - 7.08 - 7.96 - 8.57 - 9.82 - 12.07). The potential applied to the gate was scanned over a wide range (0 to +2 V) while a constant potential was applied between the source and the drain (+0.5 V). As a result of the measurement, current-voltage curves (*i-v*) were obtained.

## III. RESULT

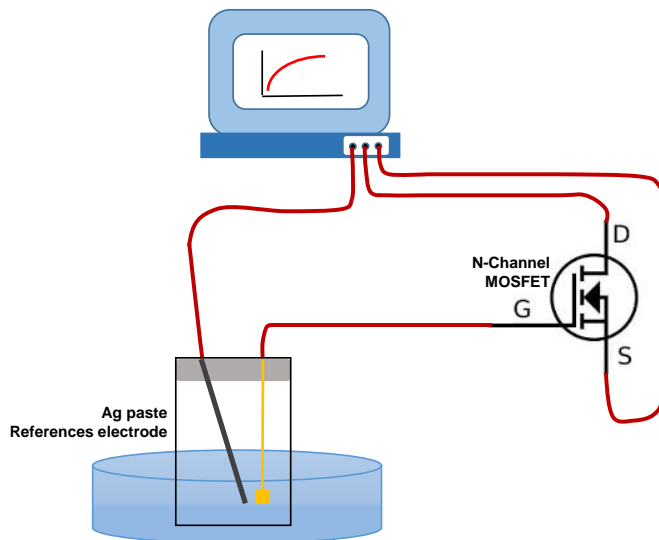


Fig. 1: EGFET pH microsensor constructed through the integration of a polypyrrole coated electrode and a MOSFET.

Electropolymerization of polypyrrole using a solution containing 0.2 M pyrrole, 0.2 M lithium perchlorate and 0.1 M perchloric acid was achieved in galvanostatic mode with a constant current of 0.1  $\mu\text{A}$ . A two-electrode cell was used for the electropolymerization process. In addition, in order to see the effect of the electropolymerization time, 0.1  $\mu\text{A}$  current was applied to the working electrode for 1, 3, 4, 5, 10 min in galvanostatic mode. When the Au microelectrode was imaged using a microscope (Top Ring Led Illuminated Stereo Microscope), it was observed that the color of the Au microelectrode changed to black (Figure 2). The result clearly demonstrates that the electropolymerization of the pyrrole onto the Au microelectrode was successful.

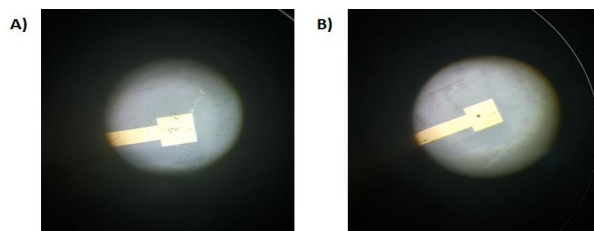


Fig. 2: Image of gold microelectrode (A) before and (B) after electropolymerization of pyrrole on chip.

The EGFET pH microsensor was connected to an n-type MOSFET and current-voltage ( $I_D-V_{GS}$ ) curves were obtained in PBS solutions at different pH values. As can be seen in

TABLE I: Parameters of polymerizing PPy and result of the chips.

PARAMATERS					RESULTS		
No	Current	Time	Incubated	Temperature	Slope (mV/pH)	Linear Range (pH)	Linear Correlation Coefficient (%)
1	0.1 uA	1 min	at room temperature during 40 minutes	25	29	5-12	91,26
2	0.1 uA	3 min	vacuum desicator for 2 hours	25	39	2-12	89,82
3	0.1 uA	4 min	at room temperature during 40 minutes	25	66	5-12,5	96,23
4	0.1 uA	4 min	not incubated	25	40	5-11	96,35
5	0.1 uA	5 min	at room temperature during 40 minutes	25	34	5-12,5	96,20
6	0.1 uA	10 min	not incubated	25	27	5-11	96,22

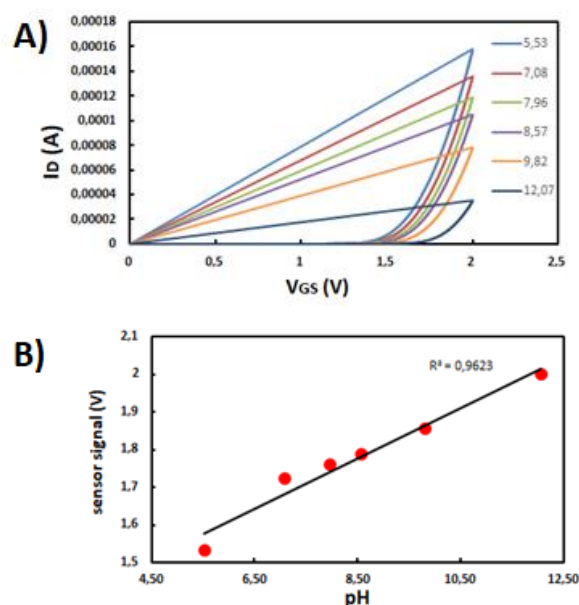


Fig. 3: I-V curves obtained at different pH values (A). obtained using  $I_D$  values measured at  $V_{GS}$  for each pH level (B). Calibration curve

Figure 3, the obtained  $I_D$ - $V_{GS}$  curves differed depending on the pH of the measured solution. The potential of the EGFET affects the gate voltage of the MOSFET, and the potential of the sensing layer may appear as the potential source. It means that the resistance of the sensing layer will affect the pH sensitivity of the EGFET pH microsensor. Different pH values changed the conductivity of the polypyrrole and thus affecting the potential applied to the channel between source and drain of the MOSFET. This situation caused different  $I_D$ - $V_{GS}$  curves to be obtained at different pH values. The calibration curve based on the voltage values obtained from  $I_D$ - $V_{GS}$  graph with respect to different pH values shows that the relationship between pH and voltage values was linear and the pH sensitivity of the microsensor was 67 mV/pH.

As seen in Table 1, in addition to the different electropolymerization times, the effects of incubating after the

polymerization process were also investigated. Due to the pH response of the 1 min and 10 min coated chips were lower than the other chips, in this study proper coating time was observed as 4 minutes. Next, to examine the effect of incubation on chips coated for 4 minutes, two types of chips were prepared; incubated for 40 min and not incubated (chip 3 and chip 4). The pH response slopes of chips 3 and 4 were obtained as 66 mV/pH and 40 mV/pH, respectively. According to these results, it was observed that the chip electropolymerized for 4 minutes with 0.1  $\mu$ A current and incubated for 40 minutes at room temperature had good pH sensitivity and linear correlation coefficient.

Results prove that the response of the EGFET pH microsensor in a relatively wide range of pH 5 to 12 was accurate. The pH microsensor developed in this study has linear pH sensitivity and good reliability. In addition, EGFET-based microsensors can be used in bioanalytical applications for pH measurement, with advantages such as miniaturization, convenient integration into different micro-systems, stability to environmental changes such as light and temperature, high conductivity and sensitivity. Therefore, the pH microsensor developed in this study is suitable to be a pH sensor for applicability to solid state sensing device with its simple manufacture, low cost and good reliability.

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