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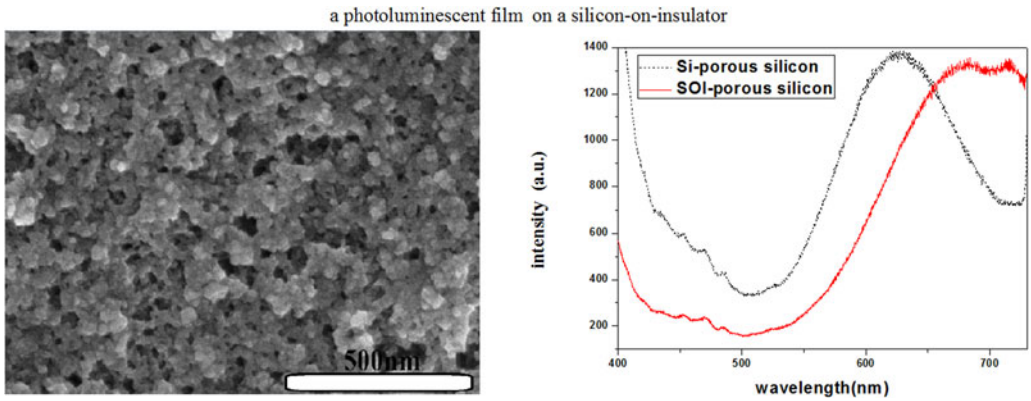

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Preparation of a Photoluminescent Film on a Silicon-On-Insulator Device for the Simple, Rapid, and Quantitative Detection of a Hydatid Disease Diagnostic Protein Marker

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Abstract: A silicon-on-insulator (SOI) device is an important integrated circuit technology containing an insulating material. In this paper, an SOI wafer consisting of n-type silicon grown on the surface of a SiO_2 layer was adopted. Porous silicon on the SOI surface, prepared by an electrochemical etching method that connected the anode and cathode to the surface of the SOI wafer, displayed strong photoluminescence properties. A hydatid disease diagnostic protein was used as a target molecule to test the detection ability of the device. After immersing the film in different concentrations of the target protein, which resulted in the simple adsorption of the protein by the porous structure, the photoluminescence intensity of the film decreased after adsorption of the protein, and with an increasing protein concentration, the photoluminescence further decreased. This photoluminescence SOI-based porous silicon film provided the rapid quantitative detection of a protein and may be a promising silicon-based optoelectronic material.

Index Terms: Silicon-on-insulator, biosensor, porous silicon, photoluminescence.

1. Introduction

A silicon-on-insulator (SOI) device is an important semiconductor technology in which an insulator layer is positioned between a silicon layer and a semiconductor substrate. Various optical devices, such as widely used optical waveguides, are prepared using SOI technology for integrated optics [1]–[3]. In recent years, SOI photonic chips have attracted significant attention in optical biochemical sensing, as these materials are not limited to the traditional telecom field [4]–[6], which is due to

their many advantages, such as submicron-scale features, high packing densities, low cost and compatibility with existing complementary metal-oxide semiconductor (CMOS) technology.

Porous silicon materials have been extensively used in optoelectronic applications, such as biological sensing, due to their unique properties [7], [8]. Many studies have focused on the fabrication of porous silicon integrated on the surface of SOI photonic devices, combining the advantages of SOI technology and porous silicon [9]–[11]. Conventional electrochemical etching methods are used to prepare porous silicon with fluorescent properties or porous silicon photonic crystals with periodic refractive index changes; however, the preparation of porous silicon on SOI is difficult using these methods [11]. Non-electrochemical etching methods, such as vapor-phase stain etching, have been adopted to prepare porous silicon films on SOI devices [12], [13], and Zhixuan Xia *et al.* reported a magnesiothermic reduction process for the fabrication of SOI resonators capped with porous silicon, but the process required a high experimental temperature [11]. In our previous work, we obtained multilayered porous silicon via a novel electrochemical etching method by simultaneously connecting the positive and negative poles on the surface of the SOI wafer, representing a simple and low-cost procedure [14], [15]. However, the SOI surface layer used to prepare porous silicon was an epitaxial p-type silicon layer, which was rich in holes and easily electrochemically anodized. Additionally, the layer did not show the unique photoluminescence properties of typical porous silicon.

In the present study, porous silicon films with strong luminescent properties were prepared on top of an SOI device with an epitaxial n-type silicon layer, and this device was used as a substrate for a biosensor chip to determine a diagnostic protein of hydatid disease. Many studies have been interested in the unique photoluminescence characteristics of porous silicon to detect biological molecules [16]–[18]. Additionally, we previously detected hydatid disease using reflection and fluorescence spectra of conventional porous silicon devices [19], [20]; however, to the best of our knowledge, a study of the photoluminescence properties of SOI-based porous silicon has not been reported. Hydatid disease is a serious parasitic disease in many regions worldwide, and diagnostic protein markers with high specificities are expected to lead to new detection techniques, which is currently being studied by the Xinjiang Key Laboratory of Echinococcosis. Extensive testing is required in the research process of diagnostic protein markers. Therefore, herein, a photoluminescent SOI-based porous silicon biosensor was fabricated, and a diagnostic protein for hydatid disease was employed as a target. When protein is added into the SOI-based porous silicon, the luminescence intensity is reduced according to the concentration of the protein. These successful endeavors will be helpful for studying hydatid disease, and these strategies can be applied in practice in many fields.

2. Material and Methods

2.1 Material and Thin Film Fabrication

The photoluminescent SOI-based porous silicon film was obtained using an anodic-etching method under illumination from an SOI wafer consisting of a 60-μm thick epitaxial n-type silicon(100) layer with a resistivity of 3-3.5 Ω·cm, grown on a thin 2.0-μm buried oxide layer on a silicon substrate. The electrolyte solution was a 1:3 mixture of hydrofluoric acid (HF, 49%) and ethanol (95%). The anodic-etching method employed a current density of 50 mA/cm^2 and etching time of 600 s. All the prepared samples were placed in air for 72 hours.

2.2 Characterization and Measurements

The relationship between the photoluminescence reduction of the SOI-based porous silicon surface and the concentration of the immobilized hydatid disease diagnostic protein was analyzed. Photoluminescence spectra were measured before and after protein adsorption by excitation with a Xe lamp (Hitachi F-4600, Japan) at an excitation wavelength of 370 nm. The adsorptions were performed by submerging the photoluminescent films into hydatid disease diagnostic protein solutions

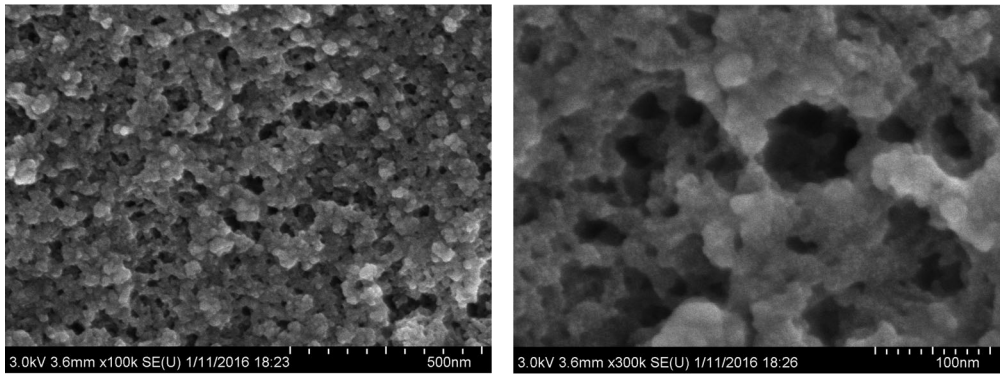

Fig. 1. Top view SEM images of SOI-based porous silicon. The current density used in the etch is 50 mA/cm^2. Pore are 20-100 nm in diameter.

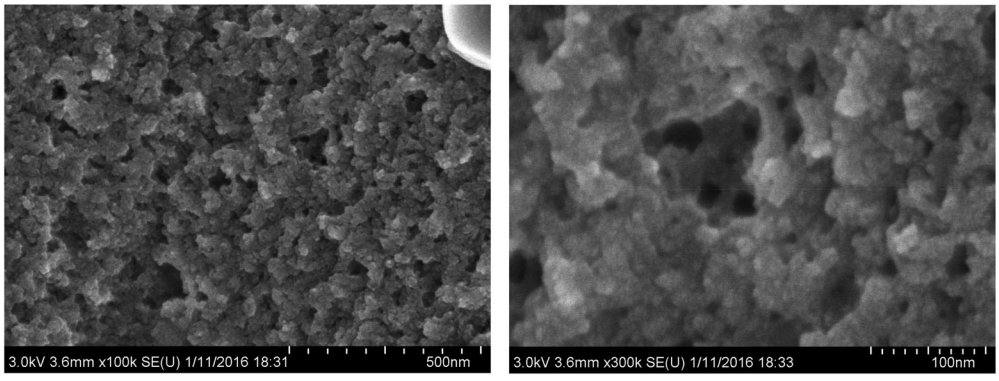

Fig. 2. Top-view SEM images of SOI-based porous silicon immobilized with the hydatid disease diagnostic protein (2×10^{-3} mg/ml). Pore are 10-60 nm in diameter.

with concentrations of 2×10^{-3} mg/ml, 2×10^{-4} mg/ml, 2×10^{-5} mg/ml, 2×10^{-6} mg/ml, 2×10^{-7} mg/ml and 2×10^{-8} mg/ml. The chemical reaction was analyzed by Fourier-transform infrared (FTIR) spectroscopy in a wavenumber range of 500-4000 cm^{-1}.

3. Results and Discussion

3.1 SEM Observation of the Surface Morphology

Generally, the pore size of n-type silicon is larger than p-type porous silicon. In agreement with this generality, Fig. 1 shows the SEM images of SOI-based porous silicon, displaying pores with diameters larger than 20 nm, which are sufficiently wide to allow easy infiltration of the hydatid disease diagnostic protein.

Fig. 2 shows the SEM images of SOI-based porous silicon immobilized with the hydatid disease diagnostic protein. From Fig. 2, the diameter of the porous silicon surface decreased significantly after exposure to the protein. Most of the holes on the surface of porous silicon were filled by the adsorption of the protein. The SEM images also show that the complex pore structure allows for efficient adsorption of the macromolecules.

3.2 FTIR Spectra Measurement

The chemical compositions of the samples were analyzed by FTIR. In Fig. 3, the SOI-based porous silicon sample before the protein treatment shows Si-H stretching vibrations at approximately 2082 cm^{-1} and 2107 cm^{-1} and an Si-H wag at 769 cm^{-1}, as well as an Si-O band at approximately

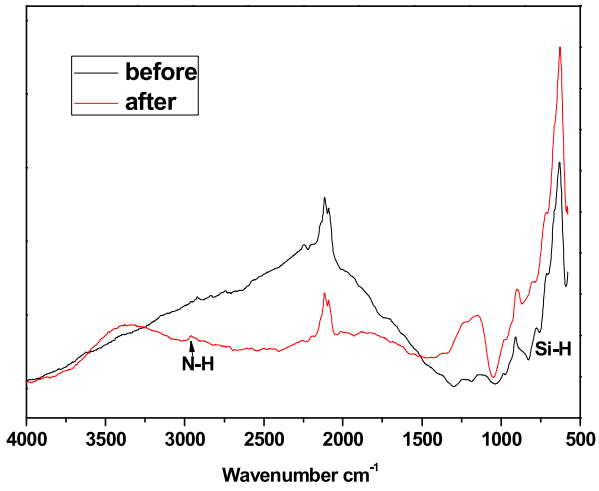

Fig. 3. FTIR spectra of the photoluminescent SOI-based porous silicon biosensor. The black curve is FTIR before any treatment. The red curve is FTIR after treatments with the hydatid disease diagnostic protein (2×10^{-3}mg/ml).

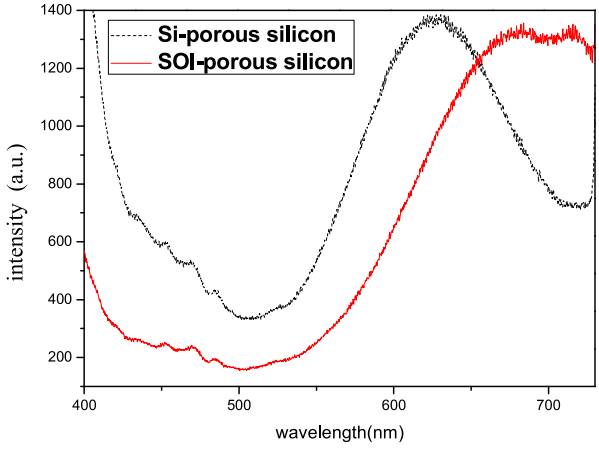

Fig. 4. Photoluminescence spectra of the porous silicon-on-silicon and SOI-based substrates. The black curve is photoluminescence of Si-based porous silicon. The red curve is photoluminescence of SOI-based porous silicon.

1126 cm^{-1} due to natural oxidation in air. After formation of the hydatid disease diagnostic protein-treated photoluminescent layer, the sample showed small bands at approximately 3000 cm^{-1}, which were attributed to N-H bonds, and a C = O band at approximately 1870 cm^{-1} [16], [17].

3.3 Photoluminescence Data Acquisition

The photoluminescence properties of the porous silicon-on-silicon and SOI-based substrates under the same conditions are compared in Fig. 4. The experimental conditions for the electrochemical etching and the excitation wavelength were the same for both substrates. As shown in Fig. 4, a significant redshift of the SOI-based porous silicon photoluminescence peak was observed, which was induced by the different electrode-loading method used in the electrochemical etching procedure. In the electrochemical etching process, the production of some electrically active holes at the interface of the silicon wafer and HF is necessary. Upon connecting the positive and negative poles on the surface of the SOI wafer, the hole replenishment rate is relatively slow, leading to a decrease of the porosity and in turn an increase of the refractive index of porous silicon. Optimization

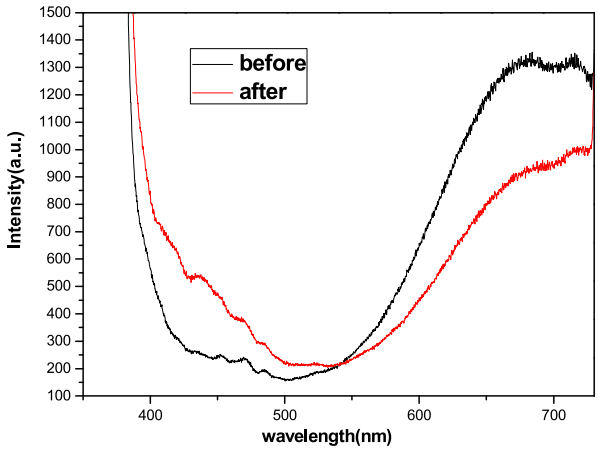

Fig. 5. Photoluminescence spectra of the SOI-based porous silicon biosensor. The black curve is photoluminescence before any treatment. The red curve is photoluminescence after treatments with the hydatid disease diagnostic protein (2×10^{-4} mg/ml).

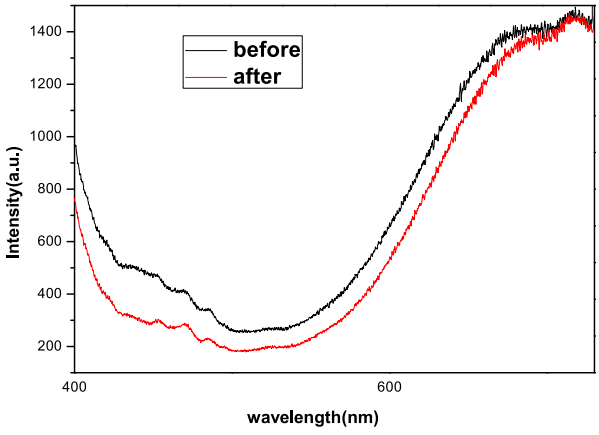

Fig. 6. Photoluminescence spectra of the SOI-based porous silicon biosensor. The black curve is photoluminescence before any treatment. The red curve is photoluminescence after treatments with 10 g/L NaCl solution.

experiments are currently in progress to investigate the photoluminescence mechanism of the SOI-based porous silicon.

Fig. 5 shows the photoluminescence intensity of SOI-based porous silicon after immersion into the 2×10^{-4} mg/ml hydatid disease diagnostic protein solution. The intensity reduction is approximately 318. Additionally, Fig. 5 shows that a slight redshift of the photoluminescence spectrum occurred due to the protein adsorption [16].

Control experiments were performed using a 10 g/L NaCl solution. Fig. 6 shows that no reduction in the photoluminescence intensity was detected after immersion into NaCl solution, indicating that no absorption of small molecules inside the large silicon pores occurred. A small redshift of the photoluminescence peak in Fig. 6 was observed, likely caused by the small change in refractive index after a small amount of molecular adsorption.

Fig. 7 shows the reduction of the photoluminescence intensity as a function of the concentration of the hydatid disease diagnostic protein (all data points are triplicate measurements), showing a constant decrease of the photoluminescence intensity with an increasing concentration. From Fig. 7, we obtain a linear relationship ranging from 2×10^{-3} mg/ml to 2×10^{-8} mg/ml with a coefficient of correlation 0.959 and detection limit of 0.02 ng/ml due to the lower concentration is near the

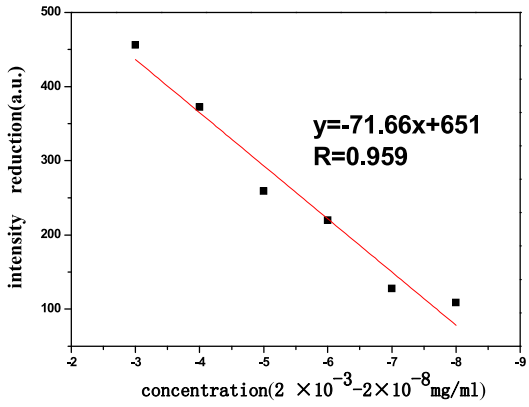

Fig. 7. Photoluminescence intensity reduction after immersion of the samples into different concentrations of the hydatid disease diagnostic protein. A linear relationship ranging from 2×10^{-3} mg/ml to 2×10^{-8} mg/ml with a coefficient of correlation 0.959.

TABLE 1
Sensitivity and Reproducibility of the SOI-Based Porous Silicon Biosensor After Exposure to Different Concentration of Hydatid Disease Diagnostic Protein in Fig. 7

	$\bar{y} \pm s$
Blank samples	44 ± 3.6
2×10^{-3} mg/ml	456 ± 3.2
2×10^{-4} mg/ml	372.6 ± 4.1
2×10^{-5} mg/ml	259 ± 2.6
2×10^{-6} mg/ml	220 ± 2.3
2×10^{-7} mg/ml	127.9 ± 3.4
2×10^{-8} mg/ml	109 ± 2.9

$\bar{y}$ is average value of photoluminescence intensity reduction, s is standard deviations.

level of the blank sample in the experiment. The average value of photoluminescence intensity reduction in Fig. 7 and standard deviations can be seen in Table 1. Experiments are currently in progress to optimize the parameters and increase the number of reproducibility experiments for reducing the impact of accidental error and investigate the relationship between the reduction of the photoluminescence intensity and the protein concentration.

We found that some large molecules, such as BSA, also change the photoluminescence intensity of the film. The quenching of luminescence by molecular adsorbates was observed with simple detection process and higher sensitivity. However, this platform has not shown higher specificity because of the simply physical adsorption of molecules, not specific bingding. So far, specific antibodies of hydatid disease diagnostic protein marker are in the process of preparation. Before this, the platform can not be used for the detection of real life samples, because the types of molecules in the actual samples are too complex.

Next, experiments are in progress to prepare specific antibodies of hydatid disease diagnostic protein marker. Once the antibody was produced successfully, the specificity should be improved by using immunoreaction with a crosslink method. Combined with immunoassay, this platform can not only be used for the detection of hydatid disease diagnostic protein, but can also be extended to other real life application.

Furthermore, hydatid disease is a serious parasitic disease in many regions worldwide, and extensive testing is required in the research process of diagnostic protein markers. Compared to conventional enzyme-linked immunosorbent assay (ELISA) techniques, our photoluminescent sensor is lower cost, has shorter detection time and is more convenient.

4. Conclusion

A photoluminescent porous silicon film on an SOI substrate was successfully fabricated and used as a biosensor chip for the simple, rapid and quantitative detection of a hydatid disease diagnostic protein marker. Compared with the other label free protein screening technologies such as plasmonic biosensors, photonic crystal biosensensors and resonator based biosensing, this photoluminescent biodetection platform is simple and low-costly. Also, it's the first time porous silicon films with strong luminescent based on SOI have been fabricated by electrochemical anodization. This study discloses two significant ramifications. First, these findings are significant for the development of SOI-based fluorescent optoelectronic devices. Second, the developed SOI-based porous silicon film has potential to be an excellent fluorescent biosensor for the simple, rapid and quantitative detection of various biomolecules.

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